Hexatic-herringbone coupling at the hexatic transition in smectic liquid crystals: 4-ϵ renormalization group calculations revisited

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Simple symmetry considerations would suggest that the transition from the smectic-*A* phase to the long-range bond-orientationally ordered hexatic smectic-*B* phase should belong to the *XY* universality class. However, a number of experimental studies have reported over the past twenty years ''novel'' critical behavior with non-*XY* critical exponents for this transition. Bruinsma and Aeppli argued [Phys. Rev. Lett. **48**, 1625 (1982)], using a 4-ϵ renormalization-group calculation, that short-range molecular herringbone correlations coupled to the hexatic ordering drive this transition first order via thermal fluctuations, and that the critical behavior observed in real systems is controlled by a ''nearby'' tricritical point. We have revisited the model of Bruinsma and Aeppli and present here the results of our study. We have found two nontrivial strongly coupled herringbone-hexatic fixed points apparently missed by these authors. Yet, these two nontrivial fixed points are unstable, and we obtain the same final conclusion as the one reached by Bruinsma and Aeppli, namely that of a fluctuation-driven first-order transition. We also discuss the effect of local twofold distortion of the bond order as a possible ''extra'' order parameter in the Hamiltonian.

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I. INTRODUCTION

The nature of phase transitions in two dimensional (2D) systems has been the subject of numerous investigations over the past three decades. According to the Mermin-Wagner-Hohenberg theorem [1], the continuous symmetry of the *XY* and Heisenberg models cannot be spontaneously broken at finite temperature, and there can be no long-range magnetic order. However, Kosterlitz and Thouless (KT) [2] argued that there is a new type of phase transition from a high temperature phase with exponential decay of the correlations to a low temperature phase with power law decay of the correlations. The idea of KT has been extended by Halperin and Nelson [3] and Young [4] (HNY) to the 2D melting problem. One of the main results of the KTHNY theory is the prediction of an intermediate 2D phase called the hexatic phase for systems that have a sixfold (hexagonal) symmetry in their crystalline ground state. This hexatic phase displays short-range positional order, but quasi-long-range bond-orientational order, which is different from the true long-range bond-orientational and quasi-long-range translational order of a 2D solid phase [3,5]. The hexatic phase can be characterized by a bond-orientational order parameter defined by $\Psi_6=|\Psi_6|\exp(i6\psi_6)$. Assuming that the hexatic state exists and is not preempted by a direct first-order melting transition from the solid to the isotropic liquid phase, the system should, in the simplest scenario for 2D, display either a KT transition or a first-order transition from the hexatic state to the isotropic liquid phase [5].

It was soon realized after the proposal of the KTHNY theory that novel hexaticlike phases with short-range positional order but true long-range bond-orientational order might exist in highly anisotropic three-dimensional (3D) systems. Specifically, Birgeneau and Lister [6] applied the notion of a hexatic state of the 2D melting theory to 3D liquid crystal phases consisting of stacked 2D liquid layers. They proposed that some of the experimentally observed smectic liquid crystal phases could be physical realization of 3D hexatics. Birgeneau and Lister suggested that the (weak) interlayer interaction could promote the quasi-long-range order of 2D hexatic layers to true long-range bond-orientational order in 3D.

Stimulated by these theoretical advances, numerous experimental efforts have been undertaken to test theoretical predictions in different liquid crystal materials candidate for displaying hexatic phases [7]. An x-ray study of the liquid crystal compound 65OBC (*n*-alkyl-4′-*m*-alkoxybiphenyl-4-carboxylate,$n=6,m=5$) [8] provided the first indication of the existence of the 3D analog of the 2D hexatic phase. It was also found that in addition to the hexagonal pattern of diffuse spots of scattered intensity, which is the signature of the hexatic phase, there are some broader peaks corresponding to correlations in the molecular orientations about their long axes [8]. The positions of these peaks show that, locally, the molecules are packed according to a herringbone pattern perpendicular to the smectic layer stacking direction (see Fig. 1). Despite the indication of short-range herringbone correlations, this phase is simply denoted as the hexatic-*B* (Hex*B*) phase. Upon increasing temperature, this phase loses its long-range bond-orientational order and undergoes a transition to the smectic-*A* (Sm*A*) phase, which essentially consists of a stack of 2D liquid layers. Upon cooling, the Hex*B* phase transforms via a first-order phase transition into the crystal-*E* (Cry*E*) phase, which exhibits both long-range translational order and long-range herringbone orientational order in the orientations of the molecular axes.

According to the U(1) symmetry of the Ψ_6 bond-orientational order parameter, one would naively expect to find *XY*-like critical exponents at the Sm*A*-Hex*B* transition in 3D. However, heat-capacity investigations near the

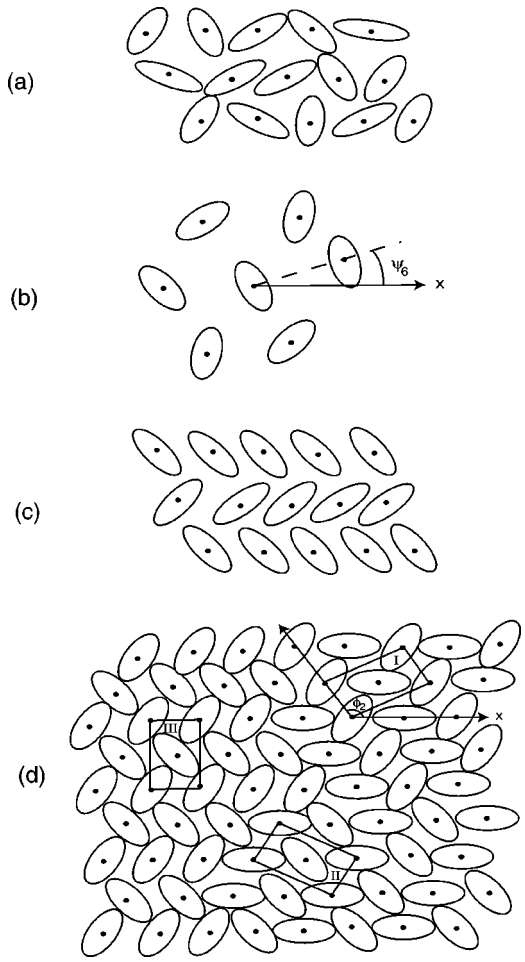

FIG. 1. Local hexagonal coordination of the molecules ''as seen'' along the stacking direction of the smectic layers. The elliptical shape of the molecules as seen along the stacking direction is meant to represent the ''wide'' benzene rings present on most thermotropic liquid crystal molecules. (a) With local hexagonal (hexatic) correlations, but without herringbone correlations. (b) With both local hexagonal and herringbone correlations. (c) shows the orientation of the molecular axis within a smectic layer as seen along the stacking direction of the smectic layers. (d) In a hexatic phase, there are three possible orientations of only short-range ordered herringbone domains. The angle ϕ_2 used to parametrize the complex herringbone order parameter $\Phi_2=|\Phi_2|\exp(i2\phi_2)$ is the angle between a fixed axis x in the laboratory frame and the direction in real space along which the molecular axes are parallel [see domain I in (d), for example].

SmA-Hex$B$ transition of 65OBC [7–9] and subsequent calorimetric studies on many other components in the nmOBC homologous series [7,10] have been reporting continuous (second-order) SmA-Hex$B$ transitions with very large values for the heat-capacity critical exponent, $\alpha\approx 0.6$. This is drastically different from the 3D XY critical exponent $\alpha=-0.007$ [11]. As well, thermal conductivity and birefringence experiments have allowed the determination of other static critical exponents, all of which differ systematically from the 3D XY value, while they, together, obey the standard scaling relationships expected for a genuine second-order phase transition [7].

In light of the existence of the short-range herringbone fluctuations, detected in the x-ray diffraction studies [8], Bruinsma and Aeppli (BA) [12] formulated a Ginzburg-Landau theory that includes both the hexatic and the herringbone order. Because the Hex$B$ phase exhibits only short-range positional order, BA suggested that the herringbone order can also be represented by an XY order parameter described by $\Phi_2=|\Phi_2|\exp(i2\phi_2)$ (see Fig. 1). At the microscopic level, it is the molecular anisotropy and the resulting anisotropy of the intermolecular pair potential that creates a coupling between the hexatic bond order and the herringbone molecular order [13,14]. At the phenomenological Ginzburg-Landau level, this coupling is minimally described by a hexatic-herringbone interaction term $V_{\text{hex-her}}=h\text{Re}(\Psi_6^*\Phi_2^3)$.

BA constructed an appropriate free energy density based on symmetry considerations and investigated the effects of fluctuation corrections to the mean-field behavior for 3D systems. In the mean-field approach, their results indicate that the SmA-Hex$B$ transition should be continuous. However, 4-ϵ renormalization-group (RG) calculations, which include thermal fluctuations and the coupling term $h\text{Re}(\Psi_6^*\Phi_2^3)$, show that short-range molecular herringbone correlations coupled to the hexatic ordering drive this transition first order, which becomes second order at a tricritical point [12].

Interestingly, heat-capacity measurement studies of (truly two-dimensional) two-layer free standing films of different nmOBC compounds yield very sharp heat-capacity peaks near the SmA-Hex$B$ transition which can be parametrized by a critical exponent $\alpha\approx 0.3$ [7,15]. This is in sharp contrast with the usual broad and nonsingular specific heat hump associated with the KT transition in the 2D XY model, or yet the first-order transition that could occur in a physical system where the vortex core energy is less than some critical value [5,16]. This $\alpha\approx 0.3$ result in 2D films further suggests that the SmA-Hex$B$ cannot be described by a simple model with a unique (critical) XY-like order parameter. In this context, there have been some numerical simulations aimed at obtaining more insight into the nature of the SmA-Hex$B$ transition in 2D systems. The model used in the simulations [17,18] consists of a 2D lattice of coupled XY spins based on the BA Hamiltonian. The simulation results suggest the existence of a new type of phase transition in which the two different orderings are simultaneously established through a continuous transition. It is interesting to note here that, in a seemingly different context, there have also been numerous theoretical and numerical attempts to identify ''novel chiral'' universality classes for systems such as frustrated XY model and Ising-XY coupled model [19].

Certainly, for three dimensions, the scenario of a fluctuation-driven first-order SmA-Hex$B$ transition due to hexatic-herringbone coupling would appear reasonable for the SmA-Hex$B$ transition in 65OBC which, upon further cooling, undergoes a Hex$B$-Cry$E$ transition that establishes long-range herringbone and positional order. However, the mixture of 3(10)OBC and 4-propionyl-4′-n-heptanoyloxy-azobenzene (PHOAB) exhibits a very large temperature range for the Hex$B$ phase above the crystallization temperature to the Cry$E$ phase. If there were herring-

bone fluctuations near the Sm*A*-Hex*B* transition in that mixture, one could expect them to be quite small because of the large temperature range over which liquid crystalline Hex*B* exists before long-range herringbone order develops at the Hex*B*-Cry*E* transition. In such a case, the Sm*A*-Hex*B* transition could then possibly be continuous, and belong to the (then naively expected) *XY* universality class. However, the fact that the Sm*A*-Hex*B* transition in the 3(10)OBC-PHOAB mixture is first order does not support this simple minded argument [7]. Following the same type of reasoning, recent x-ray diffraction studies on 75OBC [7] show that the intensity of the herringbone peaks is weaker than those of 65OBC. In principle, if one assumes that 65OBC is near a tricritical point, 75OBC should therefore be further removed from such a tricritical point (due to the weaker herringbone diffraction peaks, and consequently, weaker hexatic-herringbone coupling $V_{\text{hex-her}}$), with again the possibility to recover 3D *XY* critical behavior. Yet, the same (unconventional) heat-capacity critical exponents are found for these two materials.

The experimental results above could be interpreted as a possible indication of an underlying (non-*XY*) stable fixed point that controls the Sm*A*-Hex*B* transition when herringbone correlations are present, however small they might be. The apparent lack of progress on the theoretical side of the Sm*A*-Hex*B* problem has led us to reinvestigate the model of BA and to, specifically, look for a possible calculation error. First, it is important to note that the conclusion of a fluctuation-driven first order transition within a $4\text{-}\epsilon$ calculation is acutely depending on numerics and not constrained by symmetry consideration: a small error (such as a factor 2 instead of 4 here or there) can change the renormalization-group flow and the conclusion of a fluctuation-driven first order transition. Second, and more specific to the BA problem, we show in the following section, when describing the Ginzburg-Landau free energy density for the Sm*A*-Hex*B* transition, that some terms in the RG equations, to first order in ϵ, were missed in the work of BA. Third, based on our RG equations, we find two nontrivial strongly coupled herringbone-hexatic fixed points, apparently missed by those authors. However, these two nontrivial fixed points are unstable, and we reach the same final conclusion as the one found by BA, namely that of a fluctuation-driven first-order transition. We also discuss the possibility of a third and *a priori* possibly physically pertinent order parameter in the Hamiltonian model of the Sm*A*-Hex*B* transition. Because of local distortion of the bond-orientational order induced by the anisotropy of the intermolecular potential [13,14] and the herringbone correlations, one may generalize the Hamiltonian to the case with three XY-like order parameters, in which two of them are twofold symmetric, one for the herringbone correlations, Φ_2, and one for the local twofold distortion, Ψ_2, and a third-order parameter with sixfold symmetry, Ψ_6, related to hexatic ordering. We discuss both mean-field and RG calculations for this new three order parameter model.

The rest of this paper is organized as follows. In Sec. II A, we reintroduce the BA model and present the result of our RG calculations. In Sec. II B, we generalize the Hamiltonian to the case of three order parameters and discuss the mean-field theory and RG results. The discussions and conclusions appear in Sec. III.

II. MODELS AND RG CALCULATIONS

A. BA Hamiltonian

To formulate the Ginzburg-Landau (GL) free energy, which describes both the hexatic and the herringbone order, one recalls that the hexatic order is sixfold symmetric, while rotating a herringbone pattern by 180° leaves it unchanged. Consequently, the appropriate GL free energy ought to be invariant with respect to the transformation $\phi_2(r)\rightarrow\phi_2(r)+n\pi$ and $\psi_6(r)\rightarrow\psi_6(r)+m(2\pi/6)$ where n and m are integers. Thus to lowest order in Ψ_6 and Φ_2, the BA Hamiltonian [12] is

$$\beta F=\int d^3x\left[\frac{r_6}{2}|\Psi_6|^2+\frac{1}{2}|\nabla\Psi_6|^2+\frac{r_2}{2}|\Phi_2|^2+\frac{1}{2}|\nabla\Phi_2|^2 + u_6|\Psi_6|^4+u_2|\Phi_2|^4+w|\Phi_2|^2|\Psi_6|^2+h\,\mathrm{Re}(\Psi_6^*\Phi_2^3)\right]. \tag{2.1}$$

The condition for thermodynamic stability of F for $w=0$ is $h^{4/3}<(4^{4/3}/3)u_2u_6^{1/3}$ [12]. This condition can be obtained by minimizing the free energy density on the critical isothermal line $r_2=r_6=0$ and requesting that $\beta F>0$. As discussed in Ref. [12], in the mean-field approximation, for $w=0$ and $h=0$ the phase diagram in the r_2-r_6 plane includes four distinct phases and phase transitions: an isotropic (Sm*A*) phase with $\Psi_6=\Phi_2=0$, a hexatic (Hex*B*) phase with no herringbone order with $\Psi_6\neq0,\Phi_2=0$, a ''putative'' herringbone (or nematic) liquid crystal phase with $\Psi_6=0,\Phi_2\neq0$ [20], and a fully ordered state with $\Phi_2\neq0,\Psi_6\neq0$ [21]. Beyond mean-field level, and for $h=0$ and $w=0$, all these transitions are in the *XY* universality class. For $h\neq0$, the transition from Hex*B* to the fully ordered phase with both Ψ_6 and Φ_2 order belongs to the three-states clock (Potts) universality class and is first order in three dimensions. If $h\neq0$, the herringbone liquid crystal state with no hexatic order ($\Phi_2\neq0,\Psi_6=0$) is eliminated because Φ_2 acts as a symmetry-breaking field on Ψ_6. Within mean-field theory, the transition lines between the isotropic Sm*A* and ordered phases remain second-order for $h\neq0$, and terminate together with the first-order line separating the Hex*B* and the fully ordered phase at a multicritical point [12].

We now discuss the RG flow equations and the stability of the fixed points (FPs). Our calculations show that the RG equations to first order in $\epsilon=4-d$ are

$$\frac{dr_2}{dl}=2r_2+\frac{16K_4u_2}{1+r_2}+\frac{4K_4w}{1+r_6},$$

$$\frac{dr_6}{dl}=2r_6+\frac{16K_4u_6}{1+r_6}+\frac{4K_4w}{1+r_2},$$

$$\frac{du_2}{dl} = \epsilon u_2 - 40K_4 u_2^2 - 2K_4 w^2 - 9K_4 h^2,$$

$$\frac{du_6}{dl} = \epsilon u_6 - 40K_4 u_6^2 - 2K_4 w^2, \tag{2.2}$$

$$\frac{dw}{dl} = \epsilon w - 16K_4 w u_2 - 16K_4 w u_6 - 8K_4 w^2 - 18K_4 h^2,$$

$$\frac{dh}{dl} = \epsilon h - 24K_4 h u_2 - 12K_4 h w,$$

where $K_4 = 1/8\pi^2$. The above RG equations differ from those found by BA in Ref. [12] in two important ways

(1) The first set of differences are the $4K_4 w/(1+r_6)$ and $4K_4 w/(1+r_2)$ terms in the first and second equations, while BA have $2K_4 w/(1+r_6)$ and $2K_4 w/(1+r_6)$. The extra factor 2 comes from the fact that the fields Ψ_6 and Φ_2 are complex and the related correlations have two-component. For $h=0$, Eq. (2.2) (with factors $4K_4 w$) reproduces the RG equations of coupled two component two-vector model as in previous studies [23,24]. We therefore believe that the above RG equations for dr_2/dl and dr_6/dl are correct.

(2) Compared to the BA equations, we also obtain two extra and important terms, $-18K_4 h^2$, in the fifth equation and $-12K_4 hw$ in the sixth equation, which can be easily checked using Feynman diagram technique. Specifically, these two terms come from the connected diagrams in the second-order perturbative RG obtained by multiplication of the relevant diagrams of $h\Psi_6^* \Phi_2^3$ with $h\Psi_6 \Phi_2^{*3}$ for the RG equation for dw/dl, and of $w|\Phi_2|^2|\Psi_6|^2$ with $h\mathrm{Re}(\Psi_6^* \Phi_2^3)$ for the RG equation for dh/dl, respectively.

Because of the two extra terms in the RG equations for dw/dl and dh/dl, we obtain, in addition to the simple decoupled FP $(r_6^* = r_2^* = -\epsilon/5,\ u_6^* = u_2^* = \epsilon/(40K_4),\ w^* = h^* = 0)$, two fixed points such that $(w^* \neq 0,\ h^* = 0)$ and $(w^* \neq 0,\ h^* \neq 0)$. The first nontrivial FP is given by $h^* = 0$, $r_6^* = r_2^* = -\epsilon/4$, $u_6^* = u_2^* = \epsilon/(48K_4)$, and $w^* = \epsilon/(24K_4)$. This FP, akin to the one found in minimally coupled two-component two-vector model [23,24], was not discussed by BA.

However, and most interestingly, we find another nontrivial mixed herringbone-hexatic FP with all the couplings being nonzero:

$$\begin{aligned} r_6^* &= -0.248\,455\,66\epsilon, \\ r_2^* &= -0.240\,189\,95\epsilon, \\ u_6^* &= 0.019\,414\,03\epsilon/K_4, \\ u_2^* &= 0.018\,380\,82\epsilon/K_4, \\ w^* &= 0.046\,571\,69\epsilon/K_4, \\ h^* &= \pm 0.007\,665\,19\epsilon/K_4. \end{aligned} \tag{2.3}$$

Therefore, based on our RG calculations, there is a FP with $h^* \neq 0$, which was not found in the previous work of BA. Linearizing the recursion relations in the vicinity of the FPs yields the following for the FP with $(w^* \neq 0,\ h^* = 0)$: $y_1 = 2 - \epsilon/2$, $y_2 = 2 - \epsilon/6$, $y_3 = -\epsilon$, $y_4 = -2\epsilon/3$, $y_5 = y_6 = 0$. Thus, two eigenvalues are marginal, compatible with what has been found in similar minimally coupled two-component two-vector model [23]. The eigenvalues for the FP with $(h^* \neq 0,\ w^* \neq 0)$ above are

$$\begin{aligned} y_1 &= 2 - 0.488\,829\epsilon, \\ y_2 &= 2 - 0.115\,889\epsilon, \\ y_3 &= -0.997\,894\epsilon, \\ y_4 &= -0.537\,266\epsilon, \\ y_5 &= +0.121\,467\epsilon, \\ y_6 &= +0.040\,239\,2\epsilon. \end{aligned} \tag{2.4}$$

These results show that there are four positive eigenvalues, and the above nontrivial FP with $(h^* \neq 0,\ w \neq 0)$ is therefore unstable. The two largest (most positive) eigenvalues, y_1 and y_2 correspond, respectively, to the thermal eigenvalue and the ''relative'' coupling strength that places the system in coupling parameter space and determines what sequence of phase transition occurs; namely, isotropic→(hexatic + herringbone) via a unique phase transition or isotropic →hexatic→(hexatic+herringbone) via two distinct phase transitions. The four eigenvalues y_3, y_4, y_5, and y_6 essentially control the flow in the w-h-u_2-u_6 plane. y_5 and y_6 are positive, rendering the above nontrivial mixed herringbone-hexatic FP unstable. We have further confirmed explicitly that the new FP is unstable by direct numerical integration of the RG equations. We found that the RG flow goes to the unstable region identified above, which we interpret as the transition being driven first-order by fluctuations (see discussion for fluctuation-driven first-order transitions in Chap. II.4 of Ref. [22]). Therefore, while we have indeed found some discrepancies between our RG equations and those of BA, and recovered two extra coupled fixed points, we at the end still reach the same physical conclusion of BA, namely that of a fluctuation-driven first-order Sm*A*-Hex*B* transition. Consequently, in the r_2-r_6 plane, the overall phase diagram that emerges is the same as that of Bruinsma and Aeppli [Fig. 2(b) of Ref. [12]] where thermal fluctuations close to the mean-field multicritical point drive the Sm*A* to Hex*B* transition first order.

B. Generalized Hamiltonian

We expect physically the local molecular anisotropy (e.g., from the anisotropic nature of benzene rings found in most thermotropic liquid crystal materials) present in the intermolecular pair potential to couple to the local bond direction, and to create a local twofold distortion of the otherwise perfect local sixfold symmetric nearest-neighbor bond order

[13,14]. Consequently, we now discuss the GL free energy, which describes both the hexatic and the herringbone order, as well as the local twofold distortion of the bond order. If we assume that the distortion of lattice has twofold symmetry, with the order parameter $\Psi_2=|\Psi_2|\exp(i2\psi_2)$, then the resulting free energy is invariant under the transformation $\phi_2(r)\rightarrow\phi_2(r)+n\pi$, $\psi_6(r)\rightarrow\psi_6(r)+m(2\pi/6)$, and $\psi_2(r)\rightarrow\psi_2(r)+p\pi$, where n, m, and p are integers.

Aside from the above physical motivation, there is also a strictly theoretical motivation to include Ψ_2 in the theory. From an RG point of view, our motivation to expand the symmetry of our Hamiltonian stems from the observation that in N coupled two-vector models a stable fixed point (called mixed fixed point [23,24]) appears in the coupling parameter space (when $N>2$). Thus, to lowest order in Ψ_6, Φ_2, and Ψ_2, we have $\beta F=\beta F_0+U$, where the Gaussian part is given by

$$\beta F_0=\frac{1}{2}\int d^3x[r_6|\Psi_6|^2+r_2|\Phi_2|^2+\tilde{r}_2|\Psi_2|^2+2r\mathrm{Re}(\Phi_2\Psi_2^*)+|\nabla\Psi_6|^2+|\nabla\Phi_2|^2+|\nabla\Psi_2|^2+2g\mathrm{Re}(\nabla\Phi_2\nabla\Psi_2^*)], \tag{2.5}$$

and the perturbative Hamiltonian has the following form [25]:

$$\begin{aligned}U=\int d^3x[&u_6|\Psi_6|^4+u_2|\Phi_2|^4+\tilde{u}_2|\Psi_2|^4+w_1|\Psi_6|^2|\Phi_2|^2\\&+w_2|\Psi_6|^2|\Psi_2|^2+w_3|\Phi_2|^2|\Psi_2|^2+h_1\mathrm{Re}(\Psi_6^*\Phi_2^3)\\&+h_2\mathrm{Re}(\Psi_6^*\Psi_2^3)+h_3\mathrm{Re}(\Phi_2^2\Psi_2^{*2})+v_1\mathrm{Re}(\Psi_6^*\Phi_2\Psi_2^2)\\&+v_2\mathrm{Re}(\Psi_6^*\Phi_2^2\Psi_2)+v_3|\Psi_6|^2\mathrm{Re}(\Phi_2\Psi_2^*)\\&+v_4|\Phi_2|^2\mathrm{Re}(\Phi_2\Psi_2^*)+v_5|\Psi_2|^2\mathrm{Re}(\Phi_2\Psi_2^*)].\end{aligned} \tag{2.6}$$

For the case that $r_2=\tilde{r}_2$, one can simply diagonalize the Gaussian part of the Hamiltonian using the transformation $\Phi_2=(\tilde{\Phi}_2+\tilde{\Psi}_2)/\sqrt{2}$ and $\Psi_2=(\tilde{\Phi}_2-\tilde{\Psi}_2)/\sqrt{2}$, and then do the RG calculations. The RG calculations for the case that $\Psi_6=0$ were done by Yosefin and Domany [26] in the study of the phase transitions in fully frustrated XY models.

In the mean-field approach, there are now four distinct phases for $r\neq 0$ ($\Psi_6=\Phi_2=\Psi_2=0;\Psi_6=0,\Phi_2\neq 0,\Psi_2\neq 0;\Psi_6\neq 0,\Psi_2=\Phi_2=0;\Psi_6\neq 0,\Psi_2\neq 0,\Phi_2\neq 0$). One should note that for the phases where both Φ_2 and Ψ_2 are nonzero, we have the condition of local stability of the free energy, $r_2\tilde{r}_2<r^2$. In addition, the singularity of the propagators at zero wave vector ($\mathbf{q}=0$) is for $r_2\tilde{r}_2=r^2$, which is the Gaussian critical point of the theory.

To obtain further insight into the specific situation where both Φ_2 and Ψ_2 go simultaneously critical (soft), we perform a RG calculation. To simplify the calculations, we consider the case that $r_2=\tilde{r}_2$, so that the fields Φ_2 and Ψ_2 are simultaneously critical (soft), and that they are equally coupled to the Ψ_6 hexatic field ($u_2=\tilde{u}_2$, $w_1=w_2$). We further need to require that the full theory, after diagonalization of the Gaussian part, is self-consistent with no new RG-generated terms. This imposes that $h_1=h_2$ and $v_1=v_2$. Using the above mentioned transformation for Φ_2 and Ψ_2 and rescaling the fields, one can rewrite the Hamiltonian as

$$\begin{aligned}\beta F=\int d^3x\Big[&\frac{r_6}{2}|\Psi_6|^2+\frac{r_2'}{2}|\tilde{\Phi}_2|^2+\frac{\tilde{r}_2'}{2}|\tilde{\Psi}_2|^2+\frac{1}{2}|\nabla\Phi_6|^2\\&+\frac{1}{2}|\nabla\tilde{\Phi}_2|^2+\frac{1}{2}|\nabla\tilde{\Psi}_2|^2+u_6|\Psi_6|^4+u_2'|\tilde{\Phi}_2|^4\\&+\tilde{u}_2'|\tilde{\Psi}_2|^4+w_1'|\Psi_6|^2|\tilde{\Phi}_2|^2+w_2'|\Psi_6|^2|\tilde{\Psi}_2|^2\\&+w_3'|\tilde{\Phi}_2|^2|\tilde{\Psi}_2|^2+h_1'\mathrm{Re}(\Psi_6^*\tilde{\Phi}_2^3)+h_3'\mathrm{Re}(\tilde{\Phi}_2^2\tilde{\Psi}_2^{*2})\\&+v_1'\mathrm{Re}(\Psi_6^*\tilde{\Phi}_2\tilde{\Psi}_2^2)\Big],\end{aligned} \tag{2.7}$$

where the new (primed) coefficients can be written in terms of old (unprimed) coefficients. To first order of ϵ, the RG equations are given by

$$\frac{dr_6}{dl}=2r_6+\frac{16K_4u_6}{1+r_6}+\frac{4K_4w_1'}{1+r_2'}+\frac{4K_4w_2'}{1+\tilde{r}_2'}, \tag{2.8}$$

$$\frac{dr_2'}{dl}=2r_2'+\frac{16K_4u_2'}{1+r_2'}+\frac{4K_4w_1'}{1+r_6}+\frac{4K_4w_3'}{1+\tilde{r}_2'},$$

$$\frac{d\tilde{r}_2'}{dl}=2\tilde{r}_2'+\frac{16K_4\tilde{u}_2'}{1+\tilde{r}_2'}+\frac{4K_4w_2'}{1+r_6}+\frac{4K_4w_3'}{1+r_2'},$$

$$\frac{du_6}{dl}=\epsilon u_6-40K_4u_6^2-2K_4w_1'^2-2K_4w_2'^2,$$

$$\frac{du_2'}{dl}=\epsilon u_2'-40K_4u_2'^2-2K_4w_1'^2-2K_4w_3'^2-9K_4h_1'^2-2K_4h_3'^2,$$

$$\frac{d\tilde{u}_2'}{dl}=\epsilon\tilde{u}_2'-40K_4\tilde{u}_2'^2-2K_4w_2'^2-2K_4w_3'^2-2K_4h_3'^2-K_4v_1'^2,$$

$$\frac{dw_1'}{dl}=\epsilon w_1'-16K_4u_6w_1'-16K_4u_2'w_1'-8K_4w_1'^2-4K_4w_2'w_3'-18K_4h_1'^2-2K_4v_1'^2,$$

$$\frac{dw_2'}{dl}=\epsilon w_2'-16K_4u_6w_2'-16K_4\tilde{u}_2'w_2'-8K_4w_2'^2-4K_4w_1'w_3'-4K_4v_1'^2,$$

TABLE I. The FPs of RG equations for the generalized Hamiltonian.

$r_6'^*/\epsilon$	$-1/5$	$-3/11$	$-2/7$	0	-0.2761944898	-0.2583527792	-0.2727272730
$r_2'^*/\epsilon$	$-1/5$	$-3/11$	$-2/7$	$-1/5$	-0.2761944898	-0.2756996328	-0.2727272730
$\tilde{r}_2'^*/\epsilon$	$-1/5$	$-3/11$	$-2/7$	$-1/5$	-0.2566490108	-0.2756996328	-0.2727272724
$u_6'K_4/\epsilon$	1/40	1/44	1/56	0	0.0202792491	0.0237809941	0.0227272727
$u_2'^*(K_4/\epsilon)$	1/40	1/44	1/56	0.0125	0.0202792491	0.0194412033	0.0170454546
$\tilde{u}_2'^*(K_4/\epsilon)$	1/40	1/44	1/56	0.0125	0.0238702512	0.0194412033	0.0170454546
$w_1'^*(K_4/\epsilon)$	0	1/44	1/28	0	0.0405584982	0.0170262066	0.0227272727
$w_2'^*(K_4/\epsilon)$	0	1/44	1/28	0	0.0164217503	0.0170262066	0.0227272727
$w_3'^*(K_4/\epsilon)$	0	1/44	1/28	0.05	0.0164217503	0.0430587966	0.0454545454
$h_1'^*(K_4/\epsilon)$	0	1/44	0	0	0	0	0
$h_3'^*(K_4/\epsilon)$	0	1/44	0	$\pm$ 0.025	0	± 0.0041763898	± 0.0113636364
$v_1'^*(K_4/\epsilon)$	0	1/44	0	0	0	0	0

$$\frac{dw_3'}{dl}=\epsilon w_3'-16K_4u_2'w_3'-16K_4\tilde{u}_2'w_3'-8K_4w_3'^2-4K_4w_1'w_2'-16K_4(h_3')^2-4K_4v_1'^2,$$

$$\frac{dh_1'}{dl}=\epsilon h_1'-24K_4h_1'u_2'-12K_4w_1'h_1'-4K_4v_1'h_3',$$

$$\frac{dh_3'}{dl}=\epsilon h_3'-8K_4u_2'h_3'-8K_4\tilde{u}_2'h_3'-16K_4w_3'h_3'-6K_4h_1'v_1',$$

$$\frac{dv_1'}{dl}=\epsilon v_1'-8K_4v_1'\tilde{u}_2'-4K_4w_1'v_1'-8K_4w_2'v_1'-8K_4w_3'v_1'-12K_4h_1'h_3'.$$

We have found a number of FPs for the above RG equations that fulfill the above condition ($r_2'^*=\tilde{r}_2'^*$, and for which $u_2'^*=\tilde{u}_2'^*$, $w_1'^*=w_2'^*$, $h_1'^*=h_2'^*$, $v_1'^*=v_2'^*$, and $v_4'^*=v_5'^*$). Some of these are given in Table I, where each column corresponds to a different FP (the nontrivial mixed herringbone-hexatic FP found in Sec. II A occurs here as well, but is again unstable). As in the simplest case of Hamiltonian (2.1), none of the FPs correspond to a nontrivial stable fixed point: each fixed point (column) shows more than two positive eigenvalues. Given the complexity of those nonlinear equations we cannot be 100% sure that we have found all the (unstable) FPs in the $(u_6,u_2',\tilde{u}_2',w_1',w_3',h_1',h_3',v_1')$ plane. However, a numerical investigation of the RG flow in that plane starting from a large number of initial values for the coupling parameters always failed to converge towards a stable and attractive fixed point. Therefore, we reach the same conclusion obtained in the preceding section, namely that of a fluctuation-driven first-order transition for this expanded symmetry Hamiltonian, and the stabilization of fixed point in N coupled two-vector models for $N>2$ does not appear to occur in this generalized Hamiltonian due to the extra Ψ_2 order parameter and associated coupling parameters h_1, h_2, and h_3.

III. DISCUSSION

In Sec. II A, we found that there are two fixed points (FPs) in the minimal Ginzburg-Landau model of the smectic-*A* to hexatic-*B* phase transition, which were missed in the previous work of Bruinsma and Aeppli [12]. However, those FPs are unstable, and we reach the same final conclusion as Bruinsma and Aeppli, namely that the Sm*A*-Hex*B* transition is driven first order by fluctuations in the BA Hamiltonian. We also discussed a slightly modified simple model that considers not only the hexatic and the herringbone order, but also one that involves the local twofold deformation of the bond correlations induced by the herringbone correlations. We assumed that this deformation can be represented by a twofold symmetry order parameter, as in the case of herringbone order, and wrote the free energy based on symmetry arguments. We were not able to find a stable FP which could possibly result in unconventional (new) second order critical exponents.

It is not clear to us in what directions to pursue the paradoxical puzzle of ''new universality'' (non 3D XY transition) in hexatic liquid crystal materials. In the work presented here, we have found a ''mixed'' hexatic-herringbone fixed point in the theory, but which is unstable to first order in ϵ. One notes that the two positives eigenvalues y_5 and y_6 in Eq. (2.4) are only very slightly positive for $\epsilon=1$. This observation may open the possibility that in a calculation that includes hexatic-herringbone coupling, there is no stable fixed point to lowest order in ϵ, but that the fixed point may be stabilized in a theory that goes beyond an $O(\epsilon^1)$ calculation. This is what happens, for example, in the normal to superconducting phase transition where the ϵ expansion to order ϵ^1 predicts a fluctuation-driven first-order phase transition [27] while theoretical arguments and Monte Carlo simulations strongly argue for a second-order (inverted 3D XY) phase transition [28].

Having said that, one should note that there are experiments on liquid crystal materials that do not display any herringbone correlations [29,30] but still show a Sm*A*-Hex*B* transition with exponents that differ from the critical behavior expected for a 2D [29] or 3D [30] XY critical behavior. That may suggest that the whole idea of hexatic-herringbone

interactions is a red herring (no pun intended). Another possibility is that a more subtle ''hidden'' order parameter distinct from the herringbone order characterizes the Sm*A*-Hex*B* transition in real materials, and that the coupling between this hidden order parameter and the hexatic order parameter Ψ_6 produces a stable fixed point. Clearly, more experimental studies are needed to shed light on this problem. In particular, high resolution scattering experiments would seem necessary to search for extended short-range correlation in either molecular correlations or distortion of hexagonal coordination to shed some light on what such hidden order parameter(s) may be.

We finally note that a related (unconventional critical behavior) situation arises in the context of layered systems of smectic liquid crystals studied by Defontaines and Prost [31]. These authors have argued that critical points that do not involve any symmetry change can define a set of new universality classes in layered systems. Possibly, considerations of some features of the Defontaines Prost theory may be useful in further investigations of the Sm*A*-Hex*B* problem.

We hope that our work and reinvestigation of the long standing Sm*A*-Hex-*B* transition in smectic liquid crystal materials will motivate further theoretical, numerical, and experimental investigations of this very interesting problem.

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