

Critical Behavior of a Noncritical Field: Destruction of Smectic Order at the Smectic A-C Tricritical Point and Implications for de Vries Behavior

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Critical behavior near the smectic A-C tricritical point is studied using renormalization group techniques. Critical fluctuations induce a singular softening of the smectic bulk modulus in the A phase. At the tricritical point, the quasi-long-range positional order of the smectic layers is destroyed. Despite this loss of order, dislocations remain bound so that smectic elasticity is retained but becomes anomalous, i.e., length scale dependent. The critically induced large layer fluctuations lead to negative thermal expansion of the layers in the A phase and may explain the origin of de Vries behavior. Experimental predictions are given for the temperature dependence of the smectic bulk modulus, x-ray structure factor, and layer spacing in the A phase.

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Since the development of the renormalization group, an enormous amount of work has been carried out to investigate critical behavior at phase transitions. The variety of systems exhibiting critical behavior is incredibly rich: ferromagnets, antiferromagnets, superconductors, liquid crystals, superfluids, and disorder driven metal-insulator transitions, to name a few. In many of these systems, there is a coupling between the critical order parameter (e.g., magnetization) and a noncritical field (e.g., underlying lattice elasticity), and the interplay between the two is a topic of significant importance [1]. It is known that critical fluctuations can induce weak singularities in the noncritical field; see, e.g., Ref. [2]. Until now, however, despite the wide range of systems studied, these singularities have been shown to be sufficiently weak so that the noncritical field retains its order; e.g., the lattice retains long range positional order at the paramagnetic-ferromagnetic transition.

In this Letter, we show that it is possible for critical fluctuations of the order parameter to *significantly* affect the order of an underlying noncritical field. The phase transition exhibiting this novel and dramatic behavior is the well known tricritical smectic A-C transition in liquid crystals, and the noncritical order that is destroyed is the quasi-long-range positional order (QLRPO) of the smectic layers [3]. Aside from the significance of this result in the general context of phase transitions and critical behavior, it has important implications, both scientific and technological, for the AC transition. In particular, we believe it may explain the origin of the much debated de Vries behavior (an unusually small change in layer spacing at the AC transition) [4].

Smectics have density modulated along one direction (\hat{z}), as shown in Fig. 1. Their elongated molecules tend to align their long axes along a common direction \hat{n} . In the A phase, $\hat{n} = \hat{z}$, while in the lower temperature C phase, \hat{n}

lies at an angle θ to \hat{z} . The C phase order parameter \mathbf{c} is the projection of \hat{n} onto the layering plane. Following de Gennes's observation [5] that the AC transition should belong to the 3d XY universality class, further analyses were performed [1,6,7], all of which incorporated layer fluctuations. As shown in Fig. 1, the displacement field $u(\mathbf{r})$ describes the layer deviation, at position \mathbf{r} , from its ground state. Layer fluctuations are measured by the correlation function $g(\mathbf{r}_\perp, z) \equiv \frac{q_L^2}{2} \langle [u(\mathbf{r}_\perp, z) - u(\mathbf{0}, 0)]^2 \rangle$, where the angled brackets denote a thermal average and $q_L = 2\pi/a$, with a the layer spacing. It was found that layering fluctuations should not affect the 3d XY universality classification and also [7] that the A phase layering behavior is unaffected by \mathbf{c} 's critical fluctuations; i.e., $g(\mathbf{r}_\perp, z)$ still grows logarithmically with distance:

$$g(\mathbf{r}_\perp, z) \approx \begin{cases} \eta \ln r_\perp^2 & z = 0 \\ \eta \ln z & r_\perp = 0, \end{cases} \quad (1)$$

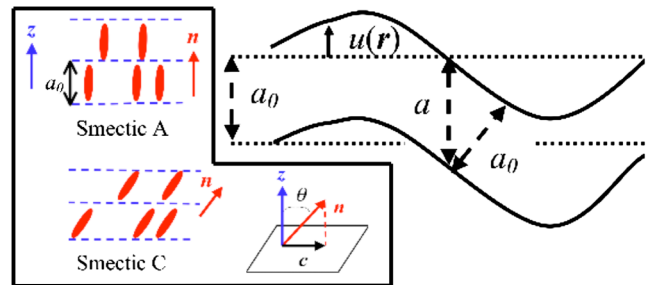


FIG. 1 (color online). Inset: The smectic A and C phases and order parameter \mathbf{c} . Also shown is a layer (solid line) undulating about its ground state (dotted line). The displacement is represented by the field $u(\mathbf{r})$. The local layer spacing, e.g., a , is measured along the z direction. Thus, the average layer spacing of the undulating state is larger than that of the ground state (a_0).

where η is a nonuniversal, weakly T dependent exponent. The slow, logarithmic (as opposed to algebraic) growth of fluctuations corresponds to the well known QLRPO with associated quasisharp (as opposed to delta-function) Bragg peaks in x-ray scattering. For a peak centered at $mq_L \hat{\mathbf{z}}$ (with m an integer), the intensity scales with wave vector $\mathbf{q} \approx mq_L \hat{\mathbf{z}}$ like

$$I(\mathbf{q}) \sim \begin{cases} q_{\perp}^{-4+2m^2\eta} & q_z = mq_L \\ (q_z - mq_L)^{-2+m^2\eta} & q_{\perp} = 0. \end{cases} \quad (2)$$

The analyses [1,6,7] did not consider AC transitions that are near a tricritical point (where first and second order phase boundaries meet). Early work on tricritical points in magnetic systems [8] established that tricritical fluctuations are larger than those for nontricritical transitions. Thus, it is of interest to understand how these strong fluctuations affect the already soft smectic elasticity. Experiments, e.g., Ref. [9], indicate that many transitions occur near a tricritical point and display significant fluctuation effects, including a critical suppression of the smectic bulk modulus B . Indeed, most de Vries smectics have a transition at or close to tricriticality [10].

Here, we present the first renormalization group (RG) analysis of the effects of critical fluctuations on layering in the A phase near a tricritical AC transition. Unlike transitions away from tricriticality, these effects lead to very large layer fluctuations, with important experimental and technological consequences. We show that B is *singularly* suppressed upon approach to the tricritical point. As a result, the correlation function $g(\mathbf{r})$ and peak intensity $I(\mathbf{q})$ are still described by Eqs. (1) and (2), but with a strongly T dependent exponent $\eta(T)$. In $d = 3$, $\eta(T) \propto (T - T_c)^{-1/4}$, with T_c the transition temperature. The intensity peaks are correspondingly broadened as the transition is approached. In fact, all divergent peaks will be replaced by finite, cusp singularities for $T_c < T < T_d$, where T_d is given by $\eta(T_d) = 2$. At the tricritical point ($T = T_c$), B displays anomalous elasticity; i.e., it becomes length scale dependent, resulting in the destruction of QLRPO. The correlation function $g(\mathbf{r})$ now grows algebraically with distance:

$$g(\mathbf{r}_{\perp}, z) \sim \begin{cases} r_{\perp}^{(10-3d)/2} & z = 0 \\ z^{(10-3d)/d} & r_{\perp} = 0, \end{cases} \quad (3)$$

giving exponents $1/2$ and $1/3$ in $d = 3$. Correspondingly, the intensity peaks are now finite and without cusps. We also investigate the effects of B 's singular softening on layer spacing behavior in the A phase and predict negative thermal expansion as the transition is approached. Last, we argue that de Vries behavior in the C phase is due to the large layer fluctuations near the tricritical point.

Our model's Hamiltonian is

$$H = \int d^d r \left[\frac{1}{2} t |\mathbf{c}|^2 + \frac{1}{2} K_c |\nabla \mathbf{c}|^2 + v_c |\mathbf{c}|^4 + s_c |\mathbf{c}|^6 + \frac{1}{2} B \gamma^2 + \frac{1}{2} K_s (\nabla_{\perp}^2 u)^2 + g \gamma |\mathbf{c}|^2 \right], \quad (4)$$

where $t \propto (T - T_c)$. We use a single elastic constant K_c , rather than separate splay, bend, and twist constants [11]. B and K_s are the smectic bulk and bend elastic moduli, and $\gamma \equiv \partial_z u - \frac{1}{2} (\nabla_{\perp} u)^2$. The $\partial_z u$ piece corresponds to the fractional change in the layer spacing and is positive for expansion. The second part of γ is required to satisfy rotational symmetry and leads to weak anomalous elasticity [12]. The coupling between the layering and \mathbf{c} is controlled by $g > 0$. The $|\mathbf{c}|^6$ term, with $s_c > 0$, is required to stabilize the system in the C phase. In the A phase, which is the focus of our analysis, this term has two unimportant effects. The first is a finite renormalization of v_c . The second effect (small because in $d = 3$, the s_c term is only marginally relevant in the RG sense) is a negligible logarithmic correction to the leading order T dependence of the specific heat [13].

Setting $\gamma = \partial_z u$ (and thus ignoring purely u dependent anharmonic terms) and integrating out u fluctuations from the partition function results in a purely \mathbf{c} dependent H with v_c replaced by $v'_c = v_c - g^2/2B$ [1]. v'_c is independent of K_s because the effect of the anisotropy of the smectic elasticity [i.e., $(\partial_z u)^2$ vs $(\nabla_{\perp}^2 u)^2$] vanishes in the long length scale limit. This means that the critical behavior of the order parameter \mathbf{c} is unaffected by its coupling to u . (While not obvious, we have shown that this remains true even when anharmonicities in u are retained.) Thus, one can focus on the effects of \mathbf{c} on u , without worrying about the reverse.

One can gauge these effects by using perturbation theory. The anharmonic term $g \gamma |\mathbf{c}|^2$ leads to a negative (softening) correction to B :

$$\delta B = -2k_B T g^2 \langle |\mathbf{c}|^2(\mathbf{q}) |\mathbf{c}|^2(-\mathbf{q}) \rangle_{q=0} \propto t^{-\alpha}, \quad (5)$$

where $|\mathbf{c}|^2(\mathbf{q}) = V \int (d^d p / (2\pi)^d) \mathbf{c}(\mathbf{p}) \cdot \mathbf{c}(-\mathbf{p} - \mathbf{q})$, with V the volume and $\mathbf{c}(\mathbf{q})$ the Fourier transform of $\mathbf{c}(\mathbf{r})$. The correlation function $\langle |\mathbf{c}|^2(\mathbf{q}) |\mathbf{c}|^2(-\mathbf{q}) \rangle_{q=0}$ scales like $t^{-\alpha}$, where α is the specific heat exponent. Away from the tricritical point (i.e., at the Wilson-Fisher fixed point), $\alpha < 0$ and B receives an innocuous, finite correction. However, for a tricritical transition, $\alpha = (4 - d)/2$, and for $d < 4$, the downward correction to B diverges at the tricritical point [14].

To deal with this apparent divergence, we employ the standard momentum shell RG transformation. We separate the fields into high and low wave vector components: $\mathbf{c}(\mathbf{r}) = \mathbf{c}_{<}(\mathbf{r}) + \mathbf{c}_{>}(\mathbf{r})$ and $u(\mathbf{r}) = u_{<}(\mathbf{r}) + u_{>}(\mathbf{r})$. The $>$ fields [e.g., $\mathbf{c}_{>}(\mathbf{q})$] have support in the high wave vector

range $\Lambda e^{-\ell} < q < \Lambda$, where $\Lambda = 1/a$ is the ultraviolet cutoff and ℓ is RG “time.” The $>$ fields are integrated out, and the length is then rescaled isotropically as $r = r' e^\ell$ to restore the UV cutoff back to Λ . The $<$ fields are also rescaled: $\mathbf{c}_<(\mathbf{r}) = \mathbf{c}'_<(\mathbf{r}') e^{\chi_c \ell}$ and $u_<(\mathbf{r}) = u'_<(\mathbf{r}') e^{\chi_u \ell}$. The choice of χ_c and χ_u is arbitrary and does not affect the physical results, so we choose $\chi_c = (2-d)/2$ and $\chi_u = (4-d)/2$ to keep K_c and K_s fixed. By perturbatively integrating out the $>$ fields to one-loop order, we obtain the RG flow equations for $\ell \gg 1$:

$$dt/d\ell = 2t + 4(n+2)(1-t/K_c\Lambda^2)\mu, \quad (6a)$$

$$dB/d\ell = (2-w)B, \quad (6b)$$

$$d\mu/d\ell = [\epsilon - 4(n+8)\mu]\mu, \quad (6c)$$

$$dw/d\ell = [\epsilon - w - 8(n+2)\mu]w, \quad (6d)$$

where $\epsilon = 4-d$, and $n = d-1$ is the number of components of \mathbf{c} . The dimensionless parameters $\mu \equiv k_B T (S_d \Lambda^{d-4} / (2\pi)^d K_c^2) (v_c - (g^2/2B) + (3s_c(n+4))/(d-2))$ and $w \equiv 2nk_B T (S_d \Lambda^{d-4} / (2\pi)^d) (g^2/BK_c^2) > 0$, where S_d is the surface area of a d -dimensional unit-radius sphere [15]. The RG flows of μ and w are shown in Fig. 2 for $d < 4$. The two $w = 0$ fixed points correspond to an absence of \mathbf{c} - u coupling (i.e., $g = 0$) and do not concern us. The Wilson-Fisher (WF) fixed point controls $3d$ XY critical behavior. It is the tricritical (TC) fixed point at $w^* = \epsilon$ that controls the behavior of the system at the tricritical point. As expected, TC lies on the separatrix ($\mu = 0$) dividing parameter space into a domain of first order transitions (with runaway $\mu < 0$ flows) and the $\mu > 0$ domain with

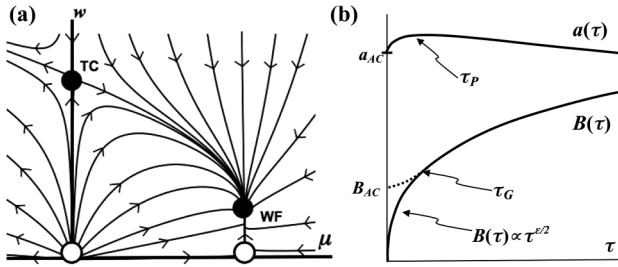


FIG. 2. (a) RG flows for μ and w . The $w = 0$ fixed points correspond to zero \mathbf{c} - u coupling. WF at $\mu^* = \epsilon/4(n+8)$ and $w^* = \epsilon(4-n)/(n+8)$ is the usual Wilson-Fisher fixed point controlling $3d$ XY critical behavior. The fixed point TC at $\mu^* = 0$ and $w^* = \epsilon$ controls tricritical behavior. (b) Bulk modulus $B(\tau)$ and layer spacing $a(\tau)$ vs reduced temperature $\tau = (T - T_c)/T_c$. Near a tricritical transition, $B(\tau) \propto \tau^{\epsilon/2}$, where $\epsilon = 4-d$. For a transition close to, but not at, tricriticality, B 's softening is cut off below the Ginzburg temperature τ_G , and B (dotted line) remains finite. Large layer fluctuations induced by B 's softening cause $a(\tau)$ to exhibit negative thermal expansion for $\tau > \tau_p$. As the proximity of the transition to tricriticality decreases, the value of τ_p increases.

flows to the WF fixed point controlling $3d$ XY critical behavior.

Having found the tricritical fixed point, we use Eqs. (6) to analyze the A phase layer fluctuations, characterized by the correlation function $G(\mathbf{q}) \propto \langle u(\mathbf{q})u(-\mathbf{q}) \rangle$, which can be related to the renormalized G by matching

$$G(\mathbf{q}, t, K_{c/s}, B, w) = e^{(d+2\chi_u)\ell^*} G(\mathbf{q} e^{\ell^*}, t(\ell^*), K_{c/s}(\ell^*), B(\ell^*), w(\ell^*)), \quad (7)$$

with t , K_c , K_s , B , and w the physical parameters. We choose ℓ^* such that the renormalized correlation length $\xi(\ell^*) = \sqrt{K_c(\ell^*)/t(\ell^*)} = \Lambda^{-1}$, and the renormalized G is evaluated in the noncritical region. Near the transition, $\ell^* \gg 1$, so that $w(\ell^*) \approx w^* = \epsilon$, giving

$$G(\mathbf{q}) = \frac{k_B T}{B(T)q_z^2 + K_s q_\perp^4}, \quad T > T_c, \quad (8)$$

with $B(T) = B\tau(T)^{\epsilon/2}$, where B is the unrenormalized bulk modulus, and $\tau(T) = t(T)/K_c\Lambda^2 = (T - T_c)/T_c$. Using $G(\mathbf{q})$, one obtains $g(\mathbf{r})$, given by Eq. (1) with

$$\eta(T) = \frac{\pi k_B T}{2a^2 \sqrt{K_s B(T)}}, \quad (9)$$

which, in $3d$, gives $\eta(T) \propto (T - T_c)^{-1/4}$. Using Eqs. (2) and (9), we find the T below which divergent peaks are absent: $T_d = T_c [1 + (\eta/2)^4]$, where η is given by Eq. (9), with the bare B , and $T = T_c$. At the tricritical point, $t = 0$, and we choose ℓ^* so that $q e^{\ell^*} = \Lambda$ and find

$$G(\mathbf{q}) = \frac{k_B T}{B(q)q_z^2 + K_s q_\perp^4}, \quad T = T_c, \quad (10)$$

where the anomalous elasticity $B(q) = Bq/\Lambda$ vanishes as $q \rightarrow 0$. Correspondingly, at tricriticality, the u - u correlation function $g(\mathbf{r})$ diverges as $r \rightarrow \infty$, according to Eq. (3), implying the destruction of QLRPO for $d < 10/3$.

We also predict that even for a transition that is close to, but not at, tricriticality, there will be a window $\tau_G < \tau < \tau_B$, within which there is significant (but nonsingular) softening of B and large layer fluctuations. Standard analysis [16] gives $\tau_B = (w/w_{*TC})^{2/\epsilon}$ and $\tau_G = (\mu/\mu_{*WF})^{2/\epsilon}$, with $w_{*TC} = \epsilon$ and $\mu_{*WF} = \epsilon/4(n+8)$. The Ginzburg temperature τ_G is the crossover from tricritical Gaussian behavior (analyzed in this Letter) to nonsingular behavior controlled by the WF fixed point. Figure 2 shows the corresponding $B(\tau)$ crossover.

The destruction of QLRPO at the tricritical point does not necessarily mean that the smectic melts, i.e., that the system no longer displays smectic elasticity. In general, the loss of smectic elasticity is the result of dislocation proliferation, so to assess the stability of the smectic at the tricritical point, we must determine if dislocations

remain bound when B vanishes at long wavelengths. Fortunately, such a nontrivial analysis has been carried out in the context of a transition between A phases of different layer spacings [17] and shows that a vanishing B does not lead to dislocation unbinding. Thus, despite the destruction of QLRPO, the system still possesses smectic elasticity, but of an anomalous, wave vector dependent variety [3].

The behavior of the layer spacing $a(T)$ near tricriticality in the A phase can also be investigated using our RG treatment. The fractional change in layer spacing $\Delta a/a$ is given by $\langle \partial_z u \rangle$, so that

$$\frac{\Delta a}{a} = \frac{1}{B} \left[-g \langle |\mathbf{c}(\mathbf{r})|^2 \rangle + \frac{B}{2} \langle |\nabla_{\perp} u(\mathbf{r})|^2 \rangle \right]. \quad (11)$$

The $\langle |\mathbf{c}(\mathbf{r})|^2 \rangle$ piece corresponds to the layer contraction due to tilting of the molecules and, for transitions far from tricriticality, it dominates the T dependence of $a(T)$, which in $d=3$ scales like $a(T) = a(T_c) + A\tau(T)^{1/2}$, where $A \sim nk_B T_c g / K_c B$. Thus, as such a transition is approached, $a(T)$ decreases and exhibits a cusp singularity at T_c . For a tricritical transition, one must also take into account the singular softening of B and growth of layer fluctuations. The latter effect results in layer undulations (i.e., nonzero $\langle |\nabla_{\perp} u(\mathbf{r})|^2 \rangle$) which, as shown in Fig. 1, tend to *increase* layer spacing a . We calculate $\Delta a/a$ near the tricritical point using matching and find

$$a(T) = a(T_c) + A \left[\tau(T)^{1/2} - \frac{2\tau(T)^{3/4}}{3\tau_p^{1/4}} \right], \quad (12)$$

where $\tau_p \sim (na(T_c)K_S^{1/2}g/B^{1/2}K_c)^4$. The extra $\tau(T)^{3/4}$ contribution is due to the layer fluctuations. As shown in Fig. 2, for $\tau(T) > \tau_p$, it tends to increase $a(T)$ as the transition is approached. This negative thermal expansion (NTE) is observed in most de Vries materials, which are known to exhibit transitions at or near tricriticality. This is particularly puzzling when NTE is accompanied by a decrease in orientational order upon approach to the transition [4]. Thus, we propose that NTE is due in significant part to the layer fluctuations, which are particularly strong near the tricritical point. Using a generalized Landau theory [18], it can be shown that $\tau_p \propto S^4$, with S the bare orientational order where, by ‘‘bare,’’ we mean far from the AC transition. In de Vries smectics, S is unusually small (due to the absence of a nematic phase), which would make τ_p small, perhaps small enough that the window ($0 < \tau < \tau_p$) may be difficult to observe.

Finally, we discuss the layering behavior on the low T side of the transition, i.e., in the C phase. It is well known [16] that, even in the absence of coupling to noncritical fields, RG analysis on the low T side of the transition is considerably more difficult for $n > 1$ due to the Goldstone mode of the ordered phase. Nonetheless, experimental

work [9], showing a significant softening of B on the low T side, helps to compensate for the lack of rigorous theoretical analysis. From the analysis presented here, it is reasonable to expect that this softening of B will be accompanied by large layer fluctuations which, as shown above, tend to increase the layer spacing. Thus, upon entry to the C phase, the expected decrease in layer spacing due to the development of \mathbf{c} order will be offset by the increase in layer spacing due to the critically induced layer fluctuations. This may go some way to explaining de Vries behavior (the unusually small change of layer spacing upon entry to the C phase), which generally occurs in materials with a transition at or near tricriticality. It should be pointed out that by design, the RG is used to model long length scale effects. Thus, small length scale effects such as molecular conformational change and layer nanosegregation cannot be modeled by the RG but could also have a role to play in de Vries behavior. The importance of the large layer fluctuations, critically induced at long length scales, should be tested using our predictions for the x-ray structure factor.

In conclusion, we have presented the first example of a system in which critical fluctuations destroy the underlying noncritical order. Moreover, we have shown that the associated softening of the bulk modulus and the large layer fluctuations near a tricritical point may explain the origin of de Vries behavior in smectics.

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