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Unconventional phase transitions in liquid crystals

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Contents

Abstract. According to classical textbooks on thermodynamics or statistical physics, there are only two types of phase transitions: continuous, or second-order, in which the latent heat L is zero, and first-order, in which $L \neq 0$. Present-day textbooks and monographs also mention another, stand-alone type — the Berezinskii–Kosterlitz–Thouless transition, which exists only in two dimensions and shares some features with first- and second-order phase transitions. We discuss examples of nonconventional thermodynamic behavior (i.e., which is inconsistent with the theoretical phase transition paradigm now universally accepted). For phase transitions in smectic liquid crystals, mechanisms for nonconventional behavior are proposed and the predictions they imply are examined.

Keywords: fluctuations, phase transitions, liquid crystals

1. Introduction. Problem formulation

The theory of second-order phase transitions, whose foundations were laid by Landau in the 1930s, crucially relies on the concept of the so-called order parameter [1], which reflects the fact that a second-order phase transition relates to the symmetry properties of the material. Namely, as the temperature decreases, spontaneous symmetry breaking occurs in the system, related to the fact that the mean value of the order parameter becomes nonzero. In the neighborhood of a second-order phase transition, fluctuations of the order parameter, i.e., its spontaneous changes due to thermal motion, are important, and their significance increases in approaching the transition point. In a certain neighborhood of the phase transition point, fluctuations destroy the simple mean-field picture. This occurs if $|T - T_c|/T_c < \mathrm{Gi}$, where Gi is the so-called Ginzburg number and T_c is the phase transition temperature. Thus, for mean field theory to apply in a certain region, the Ginzburg number must be small.

As predicted by the theory of phase transitions (see, e.g., Refs [1–6]) and shown experimentally [7, 8], various thermodynamic quantities exhibit singular behavior in the region of strong fluctuations. The spatial behavior of the order parameters is characterized by the critical radius r_c, which depends on the distance to the transition temperature in accordance with a power law,

$$r_c \propto |T - T_c|^{-\nu}\,, \tag{1}$$

with a critical exponent ν. The value of ν is positive, and hence the critical radius tends to infinity as the temperature approaches the transition point. In the region of strong fluctuations, the singular contribution to the heat capacity behaves as

$$C_{\mathrm{sing}} \propto |T - T_c|^{-\alpha}\,. \tag{2}$$

Such is the current picture of continuous phase transitions in the case where the correlation radius r_c is the largest spatial scale in the system under consideration. Under these conditions, the ε-expansion method (where $\varepsilon = d - d_c$, d being the dimension of the system and d_c the so-called upper critical dimension, for which only logarithmic corrections to the Landau mean-field theory exist) allows calculating (or more precisely, estimating) all critical exponents [1–6]. The critical exponents are uniquely determined by the spatial dimension and the number of the order parameter components (all of which are tabulated and presented in textbooks; see Refs [1, 7, 8]). The situation with first-order phase transitions (for example, gas–liquid transitions or the melting of solids) looks even simpler, and the theoretical picture, in its general features, remains the same as given in van der Waals's doctoral dissertation [9]. Van der Waals's results apply to most model and real systems exhibiting strong first-order phase transitions. From a quantitative standpoint, the term 'strong first-order phase transition' means that the latent heat L is large, i.e., if calculated per molecule, it is of the order of $k_B T_c$ or even greater (where k_B is Boltzmann's constant).

Fortunately for phase transition physicists, history is not over yet, and things are not that dull: the paradigm we discussed above relates to simple systems, whereas modern

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physics (in particular, the physics of soft matter) deals with ever more complex systems, in which r_c is not always the largest length in experimentally relevant conditions. In addition, a first-order phase transition is not at all bound to always being strong. In what follows, we discuss how such a nonconventional phase transition picture arises and what physical experimental consequences this nonconventionality implies with the example of phase transitions in smectic liquid crystals. Clearly, nonconventional phase transitions can occur not only in liquid crystals, and the reason for choosing them here is primarily that their experimental study does not require any extreme conditions and therefore is apparently easier to perform.

The rest of this paper is structured as follows. In Section 2, we discuss the phase transition from an orthogonal smectic A to a tilted smectic C, a transition in which a nonconventional behavior arises on spatial scales where orientational anisotropy is important. Section 3 illustrates nonconventional behavior at smectic-A–hexatic-smectic transitions; this behavior relates to the fact that the continuous phase transition being studied is close to another, weak first-order transition, namely, the crystallization of a hexatic smectic. In Section 4, we summarize the results and discuss some implications.

2. Smectic-A–smectic-C phase transition

The term liquid crystal refers to systems that exhibit broken orientational symmetry [characterized by a subgroup of the full rotational symmetry group O(3)] but are not three-dimensional crystals. The simplest and most common type of liquid crystals is called nematic (N). The uniaxial symmetry of the N phase, $D_{\infty h} \times T(3)$, is specified by a unit vector (director) $\mathbf{n}$ [T(3) is the full three-dimensional translational symmetry]. As the temperature decreases, the N phase usually transforms into a smectic liquid crystal in which, in addition to orientational symmetry, one-dimensional translational symmetry is also broken, with the result that the smectic is a system of equidistant layers. If the layers of a smectic liquid crystal are isotropic (the $D_{\infty h}$ symmetry with an infinite-order axis along the layer normal), then such a smectic is called a smectic A [10, 11].

To describe a smectic A, a single director $\mathbf{n}$ (aligned along the layer normal in this case) is insufficient. To describe the layered structure of the smectic, it is appropriate to introduce a scalar function $W(x, y, z)$ such that the condition $W = \text{const}$ determines equidistant layers. The free energy of the smectic (which attains a minimum for such a system of layers) can be written in covariant form as [10–12]

$$F_{\text{sm}} = \frac{1}{2} B\left[l^2 (\nabla W)^2 - 1\right]^2 , \tag{3}$$

where l is the equilibrium layer separation and B is the phenomenological Frank elastic modulus. Energy (3) should be supplemented by Frank's orientational elasticity energy [10, 11]

$$F_{\text{F}} = \frac{1}{2}\left[K_1 (\operatorname{div} \mathbf{n})^2 + K_2 (\mathbf{n} \operatorname{rot} \mathbf{n})^2 + K_3 (\mathbf{n} \times \operatorname{rot} \mathbf{n})^2\right] , \tag{4}$$

where K_1, K_2, and K_3 are the Frank elastic moduli.

In terms of the function W, the normal to the smectic layers is specified by ∇W, and in a smectic A the director is parallel to the layer normal, $\mathbf{n} = \nabla W / |\nabla W|$. As the temperature decreases, the smectic A layers lose their isotropy, and hence the uniaxial symmetry group $D_{\infty h}$ breaks. If a preferred direction appears in the layers, which is traditionally called the c-director, then they acquire the D_{2h} symmetry. Such smectics are known as smectics C. Figure 1 outlines the molecular structures in orthogonal smectics A and tilted smectics C. A preferred direction in smectic layers can arise due to the cooperative tilt of the director with respect to the layer normal. In a smectic C, molecular forces fix only the polar tilt angle (i.e., specify the projection of the vector $\mathbf{n}$ onto ∇W). Hence, the energy is independent of the azimuthal angle between $\mathbf{n}$ and ∇W, and is therefore a Goldstone variable for a smectic C. Introducing a vector ϕ_i by

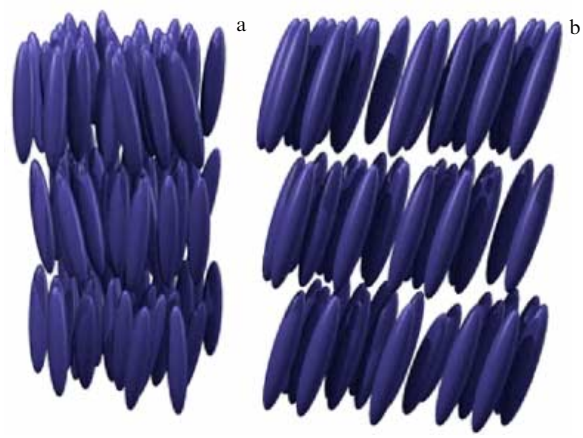

Figure 1. Schematic of molecular ordering in (a) orthogonal smectics A and (b) tilted smectics C.

$$\phi_i = \epsilon_{ikj} n_k v_j , \tag{5}$$

(where $\mathbf{v} \equiv \nabla W / |\nabla W|$ is the unit normal vector to the layers and ϵ_{ikj} is the totally antisymmetric tensor), we can express the director $\mathbf{n}$ as

$$\mathbf{n} = \left(\nabla W \left(1 - |\boldsymbol{\phi}|^2\right) + [\nabla W \times \mathbf{v}]\right) , \tag{6}$$

and then the Frank energy (4) takes the form

$$F_{\text{F}} = \frac{1}{2} K_1 (\epsilon_{\alpha\beta} \nabla_\alpha \phi_\beta)^2 + \frac{1}{2} K_2 (\nabla_\alpha \phi_\alpha)^2 + \frac{1}{2} K_3 (\nabla_z \phi_\alpha)^2 , \tag{7}$$

where the z axis is chosen to be along the layer normal and $\phi_\alpha = (\phi_x, \phi_y)$.

Thus, the phase transition from a smectic A to a smectic C is described by a two-component order parameter and, according to the traditional paradigm of second-order phase transitions [1–6], the behavior of all thermodynamic quantities at this transition should be of the same universality class as in the case of the phase transition to the superfluid state of helium-4. However, gradient energy (7) is anisotropic ($K_1 \neq K_2 \neq K_3$), and therefore the universal critical behavior is realized only in a very narrow neighborhood of the transition, $|T - T_c| / T_c \leqslant 10^{-3}$, in which the fluctuations (in the second(!) order of the ε-expansion) effectively isotropize the gradient energy [13]. Outside this region, we have a nonconventional behavior with critical exponents dependent on the orientational elastic anisotropy [13].

The reason for the nonconventional behavior in the above example is the anisotropy of the nematic orientational Frank energy, which plays an important role in a broad neighbor-

hood of the smectic-A–smectic-C transition point. The smectic order parameter W and the nematic director $\mathbf{n}$ are defined in the real coordinate space, and therefore this phase transition depends essentially on the anisotropy of the gradient energy (in analogy with magnetic phase transitions, it can be argued that this phase transition is characterized by a strong spin–orbit coupling). A totally different reason for nonconventional behavior is discussed in Section 3 with the example of the smectic-A–hexatic-smectic phase transition. In that case, the reason for the nonconventional behavior is the proximity of the second-order smectic-A–hexatic-smectic phase transition to the transition consisting in a weak crystallization of the hexatic smectic.

3. Smectic-A–hexatic-smectic phase transition

There are smectic liquid crystals in which decreasing the temperature reduces the $D_{\infty h}$ symmetry of the isotropic layers of a smectic A to the hexagonal symmetry D_{6h} (see, e.g., Refs [10, 11, 14, 15]). Such smectic liquid crystals are called hexatic smectics (or simply hexatics) (Fig. 2). Hexatics are not just exotic structures in some liquid-crystal materials. Such phases (long-range or quasi-long-range hexagonal orientational order or short-range positional order) are well known in the world of two-dimensional crystals [16] and are observed in dust plasmas [17–20] and lipid membranes [21, 22].

The reason for considering liquid crystals in this paper is that, first, they are the subject of the author's work and, second, they do not require extreme conditions for experimenting on them and are easier to bring to equilibrium.

The natural order parameter for the smectic-A–hexatic phase transition is the irreducible, symmetric, sixth-rank tensor Q_{injklm}. The irreducible nature of this tensor means that its contraction over any pair of indices yields zero, $Q_{iijklm} = 0$. Besides, the tensor order parameter Q_{injklm} relates to the plane of a smectic layer and is therefore orthogonal to the layer normal. These conditions leave us with only two independent order parameter components [23], which can be taken to be Q_{xxxxxx} and Q_{xxxxxy} or their complex combination

$$\Psi = Q_{xxxxxx} + \mathrm{i}Q_{xxxxxy} \,. \tag{8}$$

Because of the high (hexagonal D_{6h}) symmetry of a hexatic, the gradient part of the Landau free energy is isotropic (unlike the anisotropic energy (7) for the phase transition from a smectic A to a low-symmetry (D_{2h}) smectic C). Therefore, according to the theory of second-order phase transitions and the universality paradigm, the thermodynamic behavior at the phase transition from a smectic A to a hexatic should be described by the universality class of helium-4. In particular, a low negative critical exponent of the heat capacity, $\alpha = -0.01$, typical of this universality class [8, 24], should be expected. However, all calorimetric experimental data on the smectic-A–hexatic phase transition (see, e.g., Refs [25–29]), yield a large positive heat capacity exponent, $\alpha = 0.5-0.7$, giving a clear example of nonconventional behavior. Clearly, this does not mean that there is something wrong with the Nobel Prize-winning theory of phase transition. Nor does this suggest any crude systematic errors in the experimental data. The resolution of this paradox turns out to be less dramatic and lies in the simple fact that theory and experiment relate to different ranges of parameters and conditions.

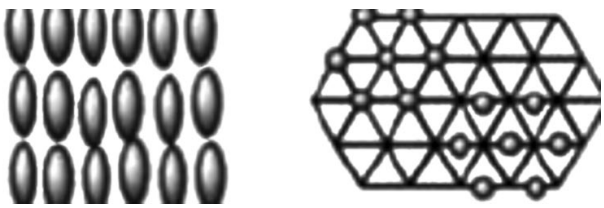

Figure 2. Schematic of molecular ordering in hexatic smectics.

Directly measuring the hexatic order parameter and the hexatic correlation length r_h is very difficult, if at all possible. The high (sixth-order axial) symmetry of a hexatic virtually prevents using optical methods, whereas X-ray scattering is sensitive to the electron density rather than to the orientational order parameter or r_h. The precision X-ray methods currently available [30–32] give a very large value of the positional correlation length r_{tr}: the value of r_{tr}, which is 1–2 nm in the smectic phase, increases to 5–10 nm near the smectic-A–hexatic phase transition, reaching 20 – 30 nm deep in the hexatic phase. For the hexatic correlation length r_h to reach such values, the system has to be in a sufficiently close vicinity of the phase transition point. The theoretical estimate in [33] shows that the universality criterion of second-order phase transitions is satisfied in the neighborhood $(T - T_c)/T_c \leqslant 10^{-3} - 10^{-4}$. Outside this narrow temperature region, a nonconventional critical behavior should be expected.

We discuss this behavior in greater detail. In the region $r_{tr} \geqslant r_h$, not only fluctuations of the hexatic order parameter but also fluctuations of the translational (positional) order parameter should be taken into account. Such an order parameter is given by the short-wavelength density modulation $\delta\rho$ (with a characteristic wave vector $q_0 \approx a^{-1}$, where the molecular size is $a = 0.1-0.3$ nm). The experimental fact we have mentioned above that $r_{tr} \gg a$ implies that a hexatic smectic (liquid crystal) is on the verge of crystallization (which is a transition to a molecular crystal). This deceptively simple fact implies the applicability of the theory of weak crystallization [34–36].

According to the theory of weak crystallization, on approaching a weak first-order phase transition, the structural factor of a liquid $S(\mathbf{q})$ (i.e., the Fourier image of the pair correlation function $\langle\delta\rho\delta\rho\rangle$) has the form

$$S(\mathbf{q}) = \frac{T}{\Delta + b(q_\perp - q_0)^2} \,, \tag{9}$$

where b is the Landau free energy expansion coefficient before the gradient term for the weak first-order crystallization transition [36], and the parameter Δ, known as the gap, satisfies the self-consistency equation

$$\Delta = a + \frac{Tq_0\lambda}{4\sqrt{b\Delta}} \,, \tag{10}$$

(where λ is the coefficient in the Landau free energy expansion at the fourth power of density $(\Delta\rho)^4$ [36]) and describes the softness (i.e., easy excitability) of the short-wavelength density fluctuations in the vicinity of the full circle $q_\perp - q_0 = 0$ in reciprocal space (and not at the isolated point $q_\perp = 0$, as in conventional second-order phase transitions with a long-wavelength order parameter). It is from this fact, trivial at first glance, that all the nonconventional features of

weak crystallization originate. For example, it follows from Eqn (10) that the gap Δ can never vanish [at arbitrarily low temperatures (!)], i.e., the liquid phase always remains at least metastable. This observation illustrates the important role of thermal fluctuations. Although individual density fluctuations are, as it were, weak due to the large volume of their easy excitation region (in the case of hexatics, the neighborhood of the circle $q_\perp - q_0 = 0$), they always have a dramatic effect.

Having the smectic-A–hexatic-smectic phase transition in mind, we should now take the interaction between the density fluctuations (i.e., of the positional order parameter) and the hexatic order parameter into account. In the first approximation (assuming this interaction to be weak), the correction to the gap Δ is

$$\delta\Delta = -\frac{1}{2T}\int \frac{\mathrm{d}^3 q}{(2\pi)^3}\, F(\mathbf{q}) S(\mathbf{k}+\mathbf{q})\,, \tag{11}$$

where the wave vector $\mathbf{k}$ refers to the density fluctuations, and we therefore have short-wavelength fluctuations in the vicinity of the circle (in our case of layered smectics, a cylinder) $k_\perp = q_0$, whereas the magnitude of the wave vector q is much smaller, $k \gg q$. Using this inequality and the weak crystallization condition, we obtain

$$\delta\Delta = -\frac{1}{8\pi^2\sqrt{b\Delta}}\int \mathrm{d}q_z\, \mathrm{d}q_\perp\, F(\mathbf{q})\,. \tag{12}$$

To calculate the heat capacity, we need to know the derivative of the gap with respect to temperature,

$$\frac{\partial}{\partial T}\Delta \propto |T - T_\mathrm{h}|^{\nu\eta - 1}\,. \tag{13}$$

This quantity determines the second derivative of the free energy, i.e., the heat capacity,

$$-T\frac{\partial^2 F}{\partial T^2} = \frac{V}{8}\frac{\partial a}{\partial T}\frac{T^2 q_0}{l b^{1/2}\Delta^{3/2}}\frac{\partial\Delta}{\partial T}\,. \tag{14}$$

Formula (14) makes an additional contribution to the heat capacity due to the interaction of the orientational (hexatic) and translational (positional) order parameter fluctuations. Our derivation shows that in the vicinity of the smectic-A–hexatic transition we are considering, the critical contribution to the temperature dependence of the heat capacity is determined by two terms: one with a small critical exponent α, consistent with the corresponding universality class, and the other with a large exponent (close to unity, $1 - \nu\eta$), which arises due to the interaction with weak-crystallization-related fluctuations of the positional order parameter.

The theoretical scheme proposed above allows representing the heat capacity at a smectic-A–hexatic phase transition in the form

$$C = \begin{cases} \dfrac{p_1}{|x|^{-0.013}} + \dfrac{p_3}{|x|} + p_5\,, & x < 0\,, \\[2ex] \dfrac{p_2}{x^{-0.013}} + \dfrac{p_4}{x} + p_5\,, & x > 0\,, \end{cases} \tag{15}$$

where $x = (T - T_\mathrm{h})/T_\mathrm{h}$, and the value of the exponent $\alpha = -0.013$ is chosen in accordance with the standard theory of second-order phase transitions with a two-component order parameter. The experimental data are ideally fitted by Eqn (15) with the following values of the parameters involved: $T_\mathrm{h} = 341.11$ K, $p_1 = -48.09599$, $p_2 = -48.19495$, $p_3 = 0.0008$, $p_4 = 0.00064$, and $p_5 = 91.60242$. It is important to note that although the parameters p_3 and p_4 are very small, they lie within the numerical accuracy of the procedure and support the above assumption that the interaction between the orientational and positional order parameters is weak. In Fig. 3, we compare the results for the heat capacity of a liquid crystal 65OBC measured in Ref. [28] (circles) and calculated by theoretical formula (15) (solid line). The agreement appears to be almost ideal (and better than one would expect from the first-order perturbation theory). A similar agreement can be achieved for measurements with other materials.

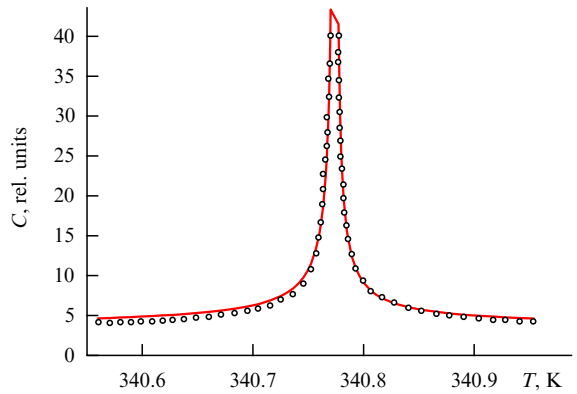

Figure 3. Experimental calorimetric data [28] (circles) and the theoretical results based on Eqn (15) (solid line) for the liquid crystal material 65OBC (n-hexyl-4′-n-pentyloxybiphenyl-4-carbxylate).

4. Conclusion

The main assertion in this paper is that the standard theoretical paradigm of phase transitions offers an adequate description of second-order phase transitions under conditions where there is only one largest length scale, the correlation length of order parameter fluctuations. The traditional scheme for describing first-order phase transitions is valid if the latent transition heat is not small compared to $k_\mathrm{B}T_\mathrm{c}$, the characteristic energy for a phase transformation occurring at a temperature T_c. There are, however, many real systems, both interesting from a fundamental perspective and attractive for applications, in which these conditions for conventional behavior either are not fulfilled at all or are fulfilled in a very narrow transition neighborhood not very important for the description of experimental data.

We have discussed two examples of such nonconventional behavior of liquid crystals. The first example is the phase transition from an orthogonal smectic A to a tilted low-symmetric smectic C. Due to the anisotropy of layers in a smectic A, the gradient energy, which is important for describing thermal fluctuations in the neighborhood of a continuous phase transition, turns out to be anisotropic (i.e., is characterized by three Frank orientational elastic constants of different magnitudes). Therefore, in a broad temperature range, the critical exponents for the transition from a smectic A to a tilted low-symmetry smectic C turn out not to be universal (contrary to conventional theory) but rather are dependent on the ratio of the elastic constants. It is only in a very narrow neighborhood $|T - T_\mathrm{c}|/T_\mathrm{c} \leqslant 10^{-3}$ of the transi-

tion point, where the fluctuation-assisted isotropization of the gradient energy occurs, that the universal picture of second-order phase transitions is restored.

In the second example of the phase transition from a smectic A to a hexatic smectic, the gradient energy is isotropic due to the high (sixth-order axial) symmetry of the hexatic. However (in fact, also because of the high orientational symmetry), such smectics are close to a crystalline state, and hence the positional correlation length (determining the proximity to the crystalline state) is very large throughout the region of existence of the hexatic smectic, much larger than the characteristic atomic or molecular scale. For the same reason, the condition that the correlation length for the orientational (hexatic) order parameter fluctuations be the only and largest spatial scale is fulfilled in a very narrow neighborhood of the transition. At the same time, in a broad temperature range of importance for the experimental study of hexatics, the critical behavior is controlled by the interaction of two fluctuating order parameters, the orientational (hexatic) one, whose fluctuations are determined by the corresponding universality class, and the positional (crystalline) one, whose fluctuations are described by the theory of weak crystallization and are important in the neighborhood of a circle in Fourier space.

Undoubtedly, such reasons for nonconventional thermodynamic behavior are not determined by any exotic property of liquid crystals. Similar phenomena should also be observed in many other real systems that have one or more complex order parameters and undergo phase transitions.

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