

Disorder- driven gradual transition of the continuous symmetry- breaking phase transition

Z naključnim neredom spodbujen postopni fazni prehod sistema z zveznim faznim prehodom, ki zlomi simetrijo

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Abstract: We study the influence of random anisotropy type quenched disorder on the phase behavior of the system, which exhibits in undistorted case the 2nd order continuous symmetry breaking phase transition. Invoking the central limit theorem we express the free energy of the system in terms of the order parameter η and the characteristic length ξ of the gauge field ϕ . The latter exhibits the Goldstone fluctuation mode and is consequently extremely susceptible to the imposed disorder. In case of negligible distribution width ΔT of the local transition temperatures the disorder converts the 2nd order transition into a discontinuous one for $W < W_C$, where W represents the disorder strength. Above the critical disorder strength W_C the transition becomes gradual. However for the finite width ΔT the transition becomes gradual for any $W > 0$. We demonstrate that for large enough values of ΔT the system behavior is dominated by the distribution of temperatures, while the details of the random field interaction term play a secondary role. The influence of distribution of local (quasi) phase transitions is in most theoretical approaches dealing with randomly perturbed systems neglected from the outset.

Key words: quenched disorder; phase transition; symmetry breaking; gradual phase transition

Povzetek: Obravnavamo vpliv naključnega nerada na fazno obnašanje sistema, ki vsebuje v nemoteni fazi fazni prehod drugega reda. Z uporabo izreka o »centralni limiti« izrazimo prosto energijo sistema na podlagi ureditvenega parametra η in karakteristične dolžine ξ umeritvenega polja ϕ . Slednji se podreja Goldstone-ovim fluktuacijskim načinom in je izredno občutljiv na motnje. V primeru zanemarljive porazdelitve lokalnih temperatur faznega prehoda, nered pretvori fazni prehod drugega reda v nezvezni prehod za vrednosti $W < W_C$, kjer je W jakost nerada. Nad kritično vrednostjo jakosti nerada W_C postane prehod postopen. V primeru, ko imamo lokalno več različnih temperatur faznega prehoda – neko porazdelitev temperatur s širino ΔT , pa postane prehod postopen za vsako vrednost $W > 0$. V članku pokažemo, da pri zadosti velikih vrednostih ΔT , fazno obnašanje določa porazdelitev temperatur, podrobnosti interakcij naključnega polja tukaj igrajo sekundarno vlogo.

Ključne besede: naključni nered; fazni prehod; zlom simetrije; postopni prehod

1. Introduction

For some years we can observe an intensive interest in how various types of disorder influence the character of

phase transformations [1, 2, 3, 4, 5]. Of particular interest are continuous symmetry- breaking (CSB) transitions. They are extremely susceptible [2] to disorder due to the existence of Goldstone mode in the broken symmetry

phase, whose amplitude diverges in the long wavelength limit. Thermal fluctuations of the mode can drive the 2nd order phase transition into the 1st order one even in a pure sample if the fluctuations are strongly coupled to the order parameter. This phenomenon is referred to as the Halperin-Lubensky-Ma (HLM) effect [6, 7, 8].

The influence of noncorrelated disorder on CSB transitions has been mostly studied theoretically using minimal universal models, e.g. Heisenberg spin systems [9]. The disorder was either introduced via random field [10] or anisotropy random field [11] terms, exhibiting one-fold and two-fold field axis symmetry, respectively. It has been shown that the 1st order phase transitions become gradual as disorder exceeds the critical value [12, 13]. On the other hand, the impact of disorder on 2nd order transitions is still open because of contradicting claims. Harris [10] has shown (the so called Harris criterion) that the bond disorder can change the critical properties of the transition if $\alpha > 0$, where α stands for the critical coefficient describing the thermal behavior of the specific heat. The renormalization study of the random anisotropy magnetic system has shown [14] that the fixed point corresponding to the non-random critical behavior is unstable even with respect to infinitesimally weak disorder strength. This instability indicates either smoothening/broadening of the transition or transformation into the transition, which displays a 1st order character. The latter arises due to the static disorder affecting the Goldstone (also called gauge) type continuum field in a reminiscent way [5] as the thermal fluctuations of this field trigger the HLM effect [6, 7]. Note that the static disorder is much more efficient in comparison to the thermal one. It was shown that random field fluctuations behave like thermal fluctuations with the lower marginal dimensionality increased by two [2]. Further, the renormalization study of Radzihovsky and Toner [15] on a similar model suggests that for a weak enough disorder a sharp transition into a Bragg-glass type phase could take place. However, the starting point of renormalization studies [16] assumes a translationally invariant effective Hamiltonian of the system, i.e. inhomogeneities are discarded at the starting point.

Regarding the structure of the disordered phase some studies predict that the broken phase exhibits a domain type structure characterized by a single characteristic length scale ξ_d [2, 9, 13, 5]. In that case the disordered phase exhibits a short range order (SRO). However, there exist several studies that predict for random anisotropy models and weak enough disorder strength the algebraic decay of correlations [3, 4], the hallmark of quasi long range order (QLRO). Note that even in this case a characteristic length ξ_d can exist indicating the distance above which the correlations decay rather weakly [3]. The

numerical simulations also yield controversial results, supporting either QLRO [17, 18] or SRO [19].

Theoretical studies on randomly perturbed systems were initially motivated to understand random magnets [9, 20]. However, in the last decades various liquid crystal (LC) phases appeared as particularly adequate experimental systems to study universal phenomena in such systems. This is due to experimental accessibility of LC phases owing to their fluidity, softness and optical anisotropy [21]. In such systems the disorder is commonly introduced either by random confinement of LCs to various inert porous matrices or by adding aerosil nanoparticles to a LC host phase [22]. In the latter case three qualitatively different random regimes are encountered by changing the concentration of aerosils [23].

In this contribution we study the influence of noncorrelated random anisotropy field disorder on a second order CSB phase transition. For illustrative purpose we consider the impact of disorder on a second order nematic-smectic A (N-SmA) phase transition. We show that an arbitrary weak disorder can convert the continuous transition into discontinuous one. However, due to spatial variations of the local phase changes the transition appears gradual on the macroscopic scale. This behavior is rather robust and for strong enough smearing independent on random field interaction details. The plan of the paper is as follows. In Sec. II we present the model. We express the effective free energy of the system, averaged over the uncorrelated RA disorder. In Sec. III the results are presented and in the last section we summarize our results.

2. Model

In this section we derive the effective free energy of a disordered system, where the disorder free energy term exhibits the two fold random anisotropy symmetry. The free energy is expressed in terms of the order parameter field η and the gauge field ϕ . The order parameter field distinguishes between the high and low temperature phase of the pure thermotropic system, separated by the continuous phase transition. If a local perturbation distorts η , it recovers its equilibrium value roughly over the distance given by the order parameter correlation length ξ_η . The gauge field ϕ describes the symmetry-breaking structure of the low temperature phase. In a pure system ϕ typically responds to a local perturbation on a scale given by geometrical constraints within the system. For example, in LC phases the pair $(\eta; \phi)$ corresponds i) in the uniaxial nematic phase to the uniaxial order parameter and nematic director field, ii) in the smectic A phase to the smectic translational order parameter and the smectic phase factor, iii) in the smectic C phase to the zenithal and azimuthal cone angle, respectively. Note that the isotropic-

nematic (I-N) transition is weakly first order, while the nematic-smectic A (N-SmA) and nematic-smectic C (N-SmC) are in general of second order.

In the following we express a typical effective free energy expression for a randomly perturbed system, described with continuum fields η and ϕ . For illustrative reasons we consider the case of the 2nd order N-SmA phase transition.

2.1. Effective free energy

We focus on LC exhibiting a continuous N-SmA phase transition in the pure (i.e. nonperturbed) case. We express the free energy in terms of the complex smectic order parameter $\psi = \eta e^{i\phi}$ and the nematic director field \vec{n} . These fields determine the local degree of translational ordering and the average orientation of the uniaxial ordering of the system, respectively. Using the Landau-Ginzburg type approach we write the free energy $f = f_c + f_e$ of the pure LC system as the sum of the condensation (f_c) and elastic (f_e) term. Using the standard Landau expansion they are expressed as [24, 25]

$$f_c = a_0(T - T_C)|\psi|^2 + \frac{b}{2}|\psi|^4 + \frac{c}{3}|\psi|^6 \quad (1a)$$

$$f_e = C_{II}|\vec{n} \cdot \nabla - iq_0\psi|^2 + C_{\perp}|\vec{n} \times \nabla\psi|^2. \quad (1b)$$

Here positive quantities a_0, b, c stand for the phenomenological coefficients of the Landau expansion, T_C stands for the N-SmA phase transition of elastically non-distorted LC, $q_0 = 2\pi/d_0$ is the smectic wave vector amplitude, corresponding to the average smectic layer thickness d_0 . The elasticity of the system is given in terms of positive compressibility (C_{II}) and smectic bend (C_{\perp}) elastic constant. We henceforth neglect the elastic anisotropy of the system and set $C = C_{\perp} \sim C_{II}$.

We further assume that at random sites the smectic translational ordering is perturbed due to the presence of phase pinning centers. The corresponding free energy density f_p is given by

$$\bar{f} \sim a_0(T - T_C^{(eff)})\bar{\eta}^2 + \frac{b}{2}\bar{\eta}^4 + \frac{c}{3}\bar{\eta}^6 + C\frac{\bar{\eta}^2}{\xi_d^2} - W_0\bar{\eta}\frac{A_d}{V_d}\left(\frac{\xi_p}{\xi_d}\right)^{3/2} \quad (5)$$

Here

$$T_C^{(eff)} = T_C - C\frac{|\nabla\gamma|}{a_0} \quad (6)$$

stands for the effective phase transition temperature. Note that the value of $\nabla\gamma$ exhibits spatial variations within the

$$f_p = -(W\psi + W^*\psi^*), \quad (2)$$

where $W = W_0 e^{-i\phi_p}$. The positive real constant W_0 determines the translational anchoring strength, favoring the phase ϕ_p .

We focus on the influence of disorder on translational degrees of freedom. Therefore we locally express the continuum fields describing the ordering of the LC system as $\vec{n} = \vec{e}_{z'}$, $\phi = q_0 z' + \varphi$ where the unit vector points along the local z' -coordinate parallel to the smectic layer normal and φ measures the departures from the uniform stack of layers along the z' -axis. With this in mind it follows

$$f_e \sim C(\eta^2|\nabla\varphi|^2 + |\nabla\eta|^2) \quad (3)$$

In the following we express the effective free energy density of the system. For this purpose we set that the disorder breaks the system into a domain type pattern characterized by a single characteristic length ξ_d . This length estimates a typical scale on which the field φ changes. Therefore, on average it roughly holds $|\nabla\varphi|^2 \sim 1/\xi_d^2$. We express the smectic ordering as $\eta = \gamma \bar{\eta}$, where the over-bar stands for the spatial averaging, and $\bar{\gamma} \sim 1$.

The average contribution of layer pinning centers is given by

$$\bar{f}_p = -\bar{\eta}W_0\overline{\cos(\phi - \phi_p)}\frac{A_d}{V_d}, \quad (4)$$

where A_d describes the average surface area of the LC-perturber interface within the average domain volume $V_d \sim \xi_d^3$. The effectiveness of the averaging within V_d is estimated using the central limit theorem. For this purpose we set that the phase factor, on average, exhibits an apparent non-correlated change at a distance $\xi_p \ll \xi_d$.

Thus, $\overline{\cos(\phi - \phi_p)} \sim \frac{1}{N_d} \sim \left(\frac{\xi_p}{\xi_d}\right)^{3/2}$, where N_d estimates the number of pinning centers within V_d . With this in mind we express \bar{f} as

system. Consequently one expects a finite distribution of the transition temperatures.

To avoid singularities for $\xi_d = 0$, we introduce the dimensionless length ξ defined as $\xi^2 + 1 = (\xi_d/\xi_p)^2$. Thus, the distances are measured in units of ξ_p . We express \bar{f} in the dimensionless form as

$$\tilde{f} = \frac{\bar{f}}{f_0} = r \tilde{\eta}^2 + \frac{\beta}{2} \tilde{\eta}^4 + \frac{1}{3} \tilde{\eta}^6 + \kappa \frac{\tilde{\eta}^2}{\xi^2 + 1} - \Lambda \frac{\tilde{\eta}}{(\xi^2 + 1)^{3/4}}, \quad (7)$$

where

$$r = \frac{T - T_C^{(eff)}}{T_C^{(eff)}} \quad (8)$$

is the effective reduced temperature, $\tilde{\eta} = \frac{\eta}{\eta_0}$, $f_0 = c \eta_0^6$, $T_C^{(eff)} = \frac{c \eta_0^4}{a_0}$, $\beta = \frac{b}{2 c \eta_0^2}$, $\kappa = \frac{c}{c \eta_0^4 \xi_p^2}$, $\Lambda = \frac{W_0 A_d}{c \eta_0^2 V_d}$. We henceforth skip the tildes.

2.2. Specific heat

The specific heat at constant pressure is defined as $c_p = -T \left(\frac{\partial^2 f}{\partial T^2} \right)_p$, where p is pressure. In the dimensionless form we express it as $\tilde{c}_p = c_p \frac{T_C^{(eff)}}{f_0}$. In the following we skip the tilde over c_p . In the appendix we express an alternative form for c_p :

$$c_p = (1+r) \frac{\left(\frac{\partial^2 f}{\partial r \partial \eta} \right)^2}{\left(\frac{\partial^2 f}{\partial \eta^2} \right)} \quad (9)$$

This form enables us to express c_p analytically in terms of η .

We assume further, that $T_C^{(eff)}$ i.e. the reduced temperature r exhibits some spatial variations within the system. In the model we take this into account via an ensemble of perturbed systems, in which each of them exhibits the 2nd order transition at spatially independent temperature $T_C^{(eff)}$ in case that the perturbation is absent. The values of the critical transition temperatures obey the Gaussian distribution

$$P(T_C^{(eff)}) = \frac{2}{\sigma \sqrt{\pi}} \exp\left(-\frac{(T_C^{(eff)} - T_0)^2}{\sigma^2}\right) \quad (10)$$

$$\sigma = \frac{\Delta T}{2 \ln 2} \quad (11)$$

Here ΔT describes the half height width of the distribution $T_0 = \langle T_C^{(eff)} \rangle$ and $\langle \dots \rangle$ stands for the ensemble average.

The corresponding average global order parameter and specific heat of the system are given by

$$\langle \eta \rangle = \int P(T_C^{(eff)}) \eta dT_C^{(eff)} \quad (12)$$

$$\langle c_p \rangle = \int P(T_C^{(eff)}) c_p dT_C^{(eff)} \quad (13)$$

3. Phase behavior

We proceed by analyzing the phase behavior of the system. Note that the case $\Delta T = 0$ has already been studied for the expansion in the order parameter up to the quartic term in f_c and $\kappa = 1$ (see ref. [5]).

We first consider the case with the negligible dispersion of transition temperatures, i.e. $\Delta T = 0$. Afterward we study the influence of a finite value of ΔT on the order parameter and specific heat temperature behavior. Finally we compare the model predictions and the available experimental results.

The obtained results give useful information about any randomly perturbed system exhibiting the 2nd order continuous symmetry breaking phase transition in the pure (i.e. non-perturbed) case. The necessary conditions are that (i) the low temperature structure is well described by the order parameter and Goldstone field (η and ϕ , respectively), and that (ii) in the elastic part of the free energy a term of the form $\eta^2 |\nabla \phi|^2$ plays the dominant role.

To stress this generally we refer to the high and low temperature phase for $\Lambda = 0$ as the *disordered* and *ordered* phase, respectively. For $\Lambda > 0$ and $\Delta T = 0$ we refer to the high temperature phase as the *paraordered* phase because of the finite value of the order parameter. The low temperature phase is referred to as the *speroordered* phase due to its domain-type structure characterized by $\xi > 0$.

3.1. Non-dispersed system

For a given value $T_C^{(eff)}$ and in the limit $\xi \rightarrow \infty$ one reproduces the undistorted bulk free energy density, corresponding to $\Lambda = 0$. By minimizing the bulk contribution one obtains $\eta(r \geq 0) = 0$ and $\eta(r < 0) = \left(\sqrt{\frac{\beta^2}{4} - r - \frac{\beta}{2}} \right)^2$. In perturbed samples, i.e., $\Lambda > 0$, a finite value of ξ appears in the speroordered phase. It reflects the competition between the elastic term (favoring $\xi \rightarrow \infty$) and the surface field term (favoring $\xi \rightarrow 0$). Minimizing $f(\xi, \eta)$ with respect to ξ yields

$$\xi = \begin{cases} 0, & \text{for } \eta \leq \eta_0 \\ \left[\left(\frac{\eta}{\eta_0} \right)^4 - 1 \right], & \text{for } \eta > \eta_0 \end{cases}, \quad (14)$$

where

$$\eta_0 = \frac{3\Lambda}{4\kappa} \quad (15)$$

stands for the crossover value of the order parameter. Therefore, the free energy density can be expressed solely in terms of η as

$$f = \begin{cases} r\eta^2 + \frac{\beta}{2}\eta^4 + \frac{1}{3}\eta^6 + \kappa\eta^2 - \Lambda\eta & , \eta \leq \eta_0 \\ r\eta^2 + \frac{\beta}{2}\eta^4 + \frac{1}{3}\eta^6 - \frac{\kappa}{3}\left(\frac{3\Lambda}{4\kappa}\right)^4 \eta^{-2} & , \eta > \eta_0 \end{cases} \quad (16)$$

Using (9) we can now calculate also the specific heat. It follows

$$c_p = \frac{2(r+1)\eta^2}{r + 3\beta\eta^2 + 5\eta^4 + \frac{81\lambda^4}{256\kappa^3\eta^4}}. \quad (17)$$

By minimizing f with respect to η we obtain the equation determining the order parameter. In Fig. 1a, Fig. 1b, and Fig. 1c we plot η , ξ and corresponding specific heat temperature dependence as the other parameters are varied. One finds that even an infinitesimal strength of the RA field converts the 2nd order transition into a 1st order one [5].

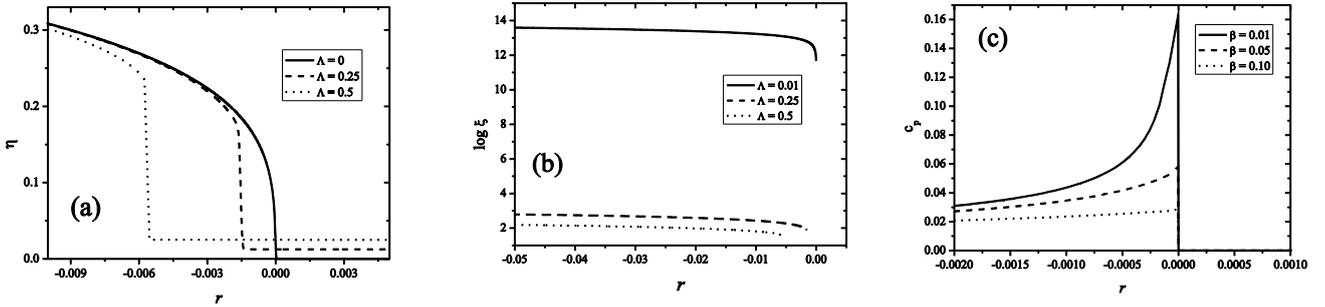


Figure 1. Temperature dependences of (a) $\eta(T)$, (b) $\xi(T)$ and (c) $c_p(T)$ as Λ or β are varied. A finite, but small enough value of Λ triggers the 1st order transition. Above the critical value of the disorder strength $\Lambda \equiv \Lambda_c$ the transition becomes gradual. $\kappa = 100$ for (a) and (b); $\beta = 0.01$

A finite value of Λ gives rise to the finite value of η above the transition characterizing the speroordered phase. The behavior of the order parameter jump $\Delta\eta$ at the transition is shown in Fig's. 2a and 3a. In general (observed for $\kappa \leq 1$), with increasing Λ the jump $\Delta\eta$ first increases and afterward decreases (see Fig. 2a). At the critical value $\Lambda \equiv \Lambda_c$ the transition is smeared out and $\Delta\eta$ drops to zero. In Fig. 2a we can infer also that if the

parameter β gets smaller the critical value of Λ increases. On the other hand when, the elastic constant κ is reduced, the transition ceases to exist at lower Λ_c . If we increase the elastic constant above the value 1.2, the system cannot get rid of the order parameter jump at the phase transition (see Fig. 3a). As we can see on the Fig. 3b the phase transition temperature would reach 0K ($r_c = -1$) before $\Delta\eta$ would drop to zero.

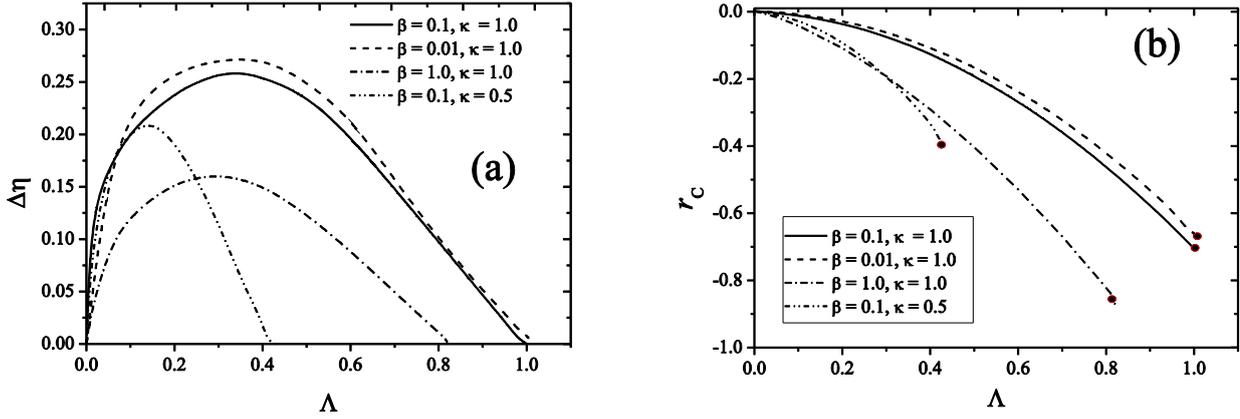


Figure 2. (a) Temperature driven jump of the order parameter and (b) phase diagram in the (r, Λ) free energy parameter space for $\kappa \leq 1$. The critical points Λ_C are marked with circles.

The corresponding phase diagram in the (r, Λ) free energy parameter space is shown in Figs. 3a and 3b.

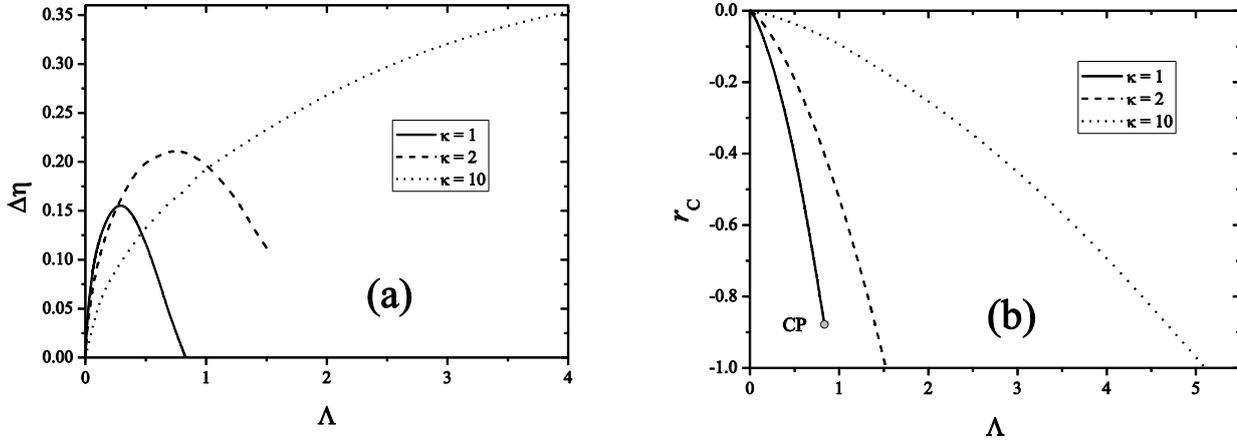


Figure 3. (a) Temperature driven jump of the order parameter and (b) phase diagram in the (r, Λ) free energy parameter space for $\kappa \geq 1$. The critical point Λ_C is marked with a circle.

The phase transition temperature T_C monotonically decreases as the disorder strength is increased until $\Lambda = \Lambda_C$. To obtain the analytic expression for Λ_C we focus on the transition line. At $\Lambda \equiv \Lambda_C$ it holds $\xi = 0$, $\Delta\eta = 0$ and consequently $\eta = \eta_0$. Taking into account $\frac{\partial f}{\partial \eta}(\eta_0) = \frac{\partial^2 f}{\partial \eta^2}(\eta_0) = 0$ we obtain

$$\Lambda_C = \frac{2\kappa}{3} \left[\sqrt{\beta^2 + \frac{16}{3}\kappa - \beta} \right]^{1/2} \quad (18)$$

3.2. Dispersed system

We next consider the influence of ΔT on the order parameter and specific heat temperature evolution, which is depicted on Figs. 4a and 4b. A finite distribution width ΔT washes out discontinuities that each member of the ensemble displays for $\Lambda < \Lambda_C$. The specific heat peak progressively broadens and decreases in height with increasing ΔT . Therefore, a finite distribution width destroys the global phase transition for any positive value of Λ .

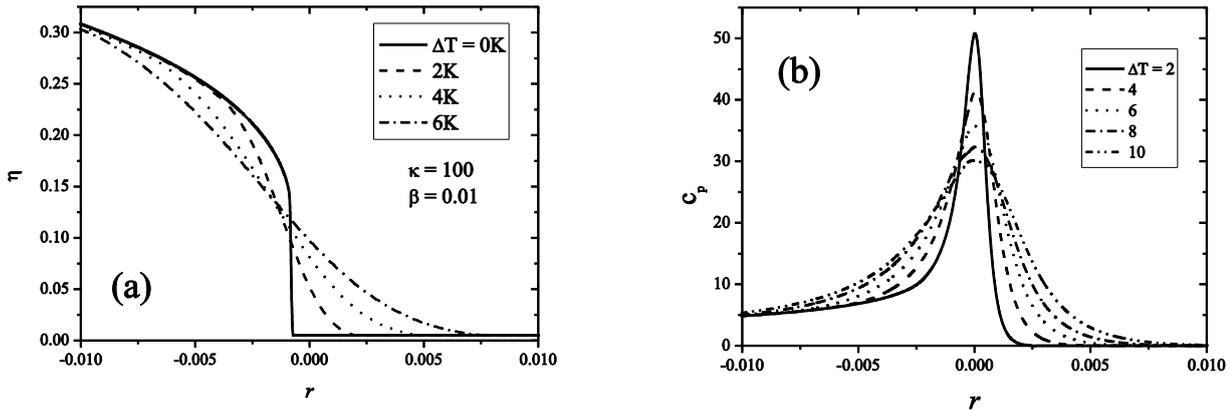


Figure 4. The (a) order parameter and (b) specific heat temperature evolution as ΔT is varied. A finite value of ΔT washes out the singularities anticipated for $0 < \Lambda < \Lambda_C$.

We next show that this behavior is robust. For this purpose we consider published experimental results for aerosil-LC mixtures. In such systems essentially randomly distributed aggregates of aerosil particles introduce a kind of random disorder to the enclosing LC phase. The results [26] suggest that the disorder strength is linearly proportional to the density of aerosil particles ρ_s . There are several publications reporting high resolution calorimetric measurements at the N-SmA and SmA-SmC phase transitions that exhibit the 2nd order transitions in pure samples. It was shown [27,28] that in the N-SmA case the specific heat is dominated by finite-size effects. The finite-size scaling is well obeyed if the characteristic finite-size length is set to be equal to the mean separation between aerosil particles $l_0 \sim 2/a\rho_s$. Here $a \sim 300 \text{ m}^2/\text{g}$ stands for

the average surface per gram area of the aerosils. On the other hand, this scaling is not observed (i.e. does not dominate the behavior) at the SmA-SmC phase transition [29], suggesting that the disorder might play the dominant role. In addition, this transition can be well described with the mean-field type model [30].

In Figs. 5a and 5b we plot the height of c_p at the SmA-SmC transition. Both, the experimental and theoretical results are superimposed. In Fig. 5a the elastic constant is varied and the distribution of phase transition temperatures within the ensemble is constant. In the case of Fig. 5b at the fixed elastic constant the distribution ΔT is altered. In both cases the theoretical and experimental results are very similar.

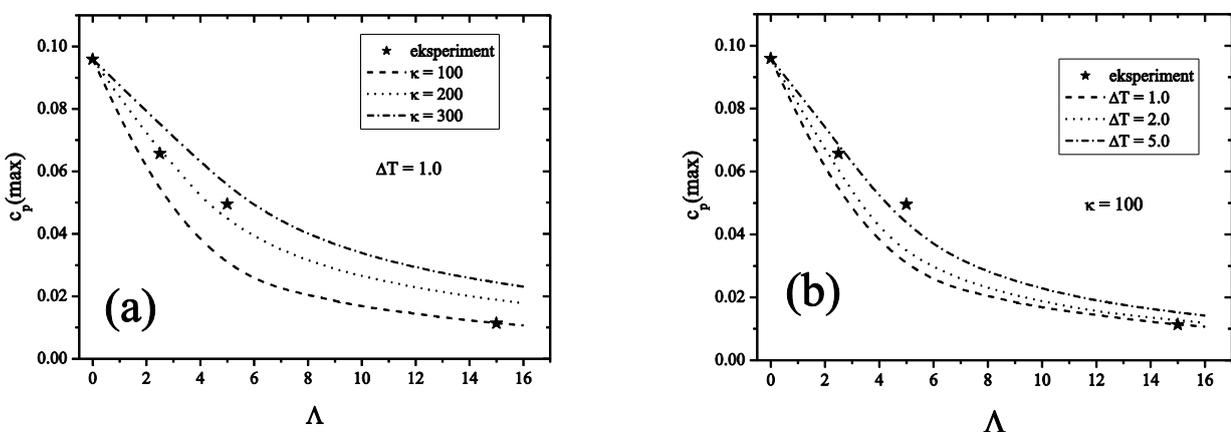


Figure 5. The excess specific heat height c_p at the paraorder - speroorder transition. Theoretical and experimental measurements are compared. In the experimental case we consider the influence of aerosil particles on the 2nd order SmA – SmC transition which is encountered in the pure LC sample. In (a) we alter the elastic constant κ , in (b) the dispersion of transition temperatures ΔT is altered.

4. Conclusions

We study the influence of the random anisotropy (RA) type disorder on the 2nd order phase transition in which a continuous symmetry is broken. For illustrative purposes we consider thermotropic LC phases in which the symmetry broken phase is characterized by an order parameter η and a gauge field ϕ exhibiting a Goldstone fluctuation mode. Due to the disorder the gauge field experiences spatially random variations that result in the characteristic length ξ . Therefore, the variational parameters of our approach are η and ξ . The disorder also gives rise to spatial variations of local quasi-phase transitions. We take this into account via an ensemble of systems with the Gaussian distribution of phase transition temperatures. In our model the disorder is introduced via the dimensionless strength Λ and also by the spreading ΔT of phase transition temperatures.

Appendix. Specific heat

We start with the thermodynamic definition of the specific heat density at constant pressure p :

$$c_p = -T \left(\frac{\partial^2 f}{\partial T^2} \right)_p, \quad (\text{A1})$$

where f is the Gibbs free energy density. We consider the case where $f = f(T, \eta, p)$ and assume a homogeneous order parameter η spatial profile. Therefore $df = \left(\frac{\partial f}{\partial T} \right)_{\eta, p} dT + \left(\frac{\partial f}{\partial \eta} \right)_{T, p} d\eta + \left(\frac{\partial f}{\partial p} \right)_{\eta, T} dp$ and $\left(\frac{\partial f}{\partial T} \right)_p = \left(\frac{\partial f}{\partial T} \right)_{\eta, p} + \left(\frac{\partial f}{\partial \eta} \right)_{T, p} \left(\frac{\partial \eta}{\partial T} \right)$. After partial derivation with respect to T and assuming an equilibrium state, we get

$$\frac{\partial^2 f}{\partial \eta \partial T} + \frac{\partial^2 f}{\partial \eta^2} \frac{\partial \eta}{\partial T} = 0, \quad (\text{A2})$$

and it follows

$$\left(\frac{\partial^2 f}{\partial T^2} \right)_p = \left(\frac{\partial^2 f}{\partial T^2} \right)_{\eta, p} + \left(\frac{\partial^2 f}{\partial \eta \partial T} \right)_p \frac{\partial \eta}{\partial T} \quad (\text{A3})$$

From Eq. (A2) we can infer that

$$\frac{\partial \eta}{\partial T} = - \frac{\frac{\partial^2 f}{\partial T \partial \eta}}{\frac{\partial^2 f}{\partial \eta^2}}. \quad (\text{A4})$$

With this in mind we obtain the specific heat as

$$c_p = T \frac{\left(\frac{\partial^2 f}{\partial T \partial \eta} \right)^2}{\frac{\partial^2 f}{\partial \eta^2}}. \quad (\text{A5})$$

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