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Phenomenological order parameter and local parameters fluctuation far beyond the critical region of the continuous phase transition

Vladimir Dmitriev [a,b,*]

[a] SNBL at ESRF, CS40220, 38043 Grenoble, France
[b] Southern Federal University, 344092 Rostov-on-Don, Russia

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ABSTRACT

In the framework of an extended phenomenological approach to phase transitions, we show that existing nonlinear relation between local critical atomic parameters and phenomenological order parameter induces the corresponding nonlinear temperature scaling transformation, and find the explicit form for such a transformation. The theoretically predicted uniform function reproduces well the experimentally observed behavior of order parameters in different systems.

1. Introduction

It is well established in the literature that, despite the fact that the phenomenological Landau theory of phase transitions [1,2] deals with various types of variational free energy, it predicts, in fact, a single set of the critical point exponents ($\alpha=0$, $\beta=1/2$, $\gamma=1$ and $\delta=3$) and thus belongs to the single "mean field" universal class [3]. In what follows we use the terminology of the theory of *critical phenomena* for the *phenomenological theory*, although the latter does not treat critical phenomena properly. The phenomenological theory characterizes a system by the critical exponents both outside and within the critical region, where temperature behavior of a general function $f(t)$ can be approximated by a simple power function $f(t)=At^{\lambda}$, with λ as a critical point exponent, and $t=|T-T_C|/T_C$ as a dimensionless variable to measure the temperature difference with the critical temperature T_C. A considerable body of experimental data indicates that the real systems show regular deviation from the behavior predicted in the framework of the Landau phenomenological theory, and different universality classes were found experimentally in such systems. It is convenient to consider such a discrepancy as caused by two main reasons: (i) the phenomenological theory neglects critical fluctuations, i.e. one assumes that the order parameter can be characterized by a single value at any temperature, and (ii) the above theory uses the dimensionality for the fluctuation space lower than the marginal dimensionality ($d < d*$) (see, for example, [4–6]). These reasons both relate to the *critical* fluctuations and are valid in the critical region. However, the existence of background hetero- and homophase fluctuations [7,8] was not considered in the analysis of the above discrepancies. Here we highlight yet another reason for the divergence of the phenomenological calculations and the corresponding experimental data. Specifically, a transcendental relation between a phenomenological order parameter and the corresponding local atomic variables along with the nonlinear temperature scaling transformation result in a deviation of the experimental values for the critical exponents from those predicted by the Landau theory. We show how the nonlinear transformation parameters depend on the fluctuation properties of the real systems *far beyond* the critical region.

2. Microscopic and macroscopic order parameters

2.1. Order parameter space

The Landau theory uses the increment $\delta\rho(\boldsymbol{r})$ of the probability density, expressing the difference between the initial density in high-symmetry parent phase, $\rho_0(\boldsymbol{r})$, and the final low-symmetry

* Correspondence address: SNBL at ESRF, 71, avenue des Martyrs, 38000 Grenoble, France. Fax: +33 476 88 2694.
E-mail address: dmitriev@esrf.fr

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phase, $\rho_d(\boldsymbol{r})$, expanded as a function of the basis functions of an irreducible representation (IR) τ_{kj} of the space group G_0 of the parent phase [1,2]. This expansion has the form:

$$\delta\rho(r) = \rho_d(r) - \rho_0(r) = \sum_{k,j,i} \eta_{kj}^{i} \varphi_{kj}^{i}(r) \tag{1}$$

The wave vector $\boldsymbol{k}$, located in the first Brillouin zone, characterizes the translational symmetry of the basis functions $\varphi_{kj}^{i}(\boldsymbol{r})$, which are the *linear* combinations of the local atomic functions, associated with the crystalline structure. The index j labels the representations $\mathfrak{a}_{kj}$ of G_0, and the index i ($i=1,\ldots,n$) runs over the distinct basis functions spanning the n-dimensional IR τ_{kj}. For a given j, the set of scalar coefficients η_{kj}^{i} defines the *order parameter* (OP), which describes the total distortion of the initial structure at the transition. Usually a single irreducible OP breaks the symmetry in a phase transition, so we only keep index i in Eq. (1).

The linear coupling between η_j and $\varphi_j(\boldsymbol{r})$ in Eq. (1) allows choosing either of these two quantities as a forming basis of the relevant IR. As a consequence, the non-equilibrium thermodynamic potential, associated with the transition, $\Phi_L(T,p,\delta\rho)$, can be considered as a function of the η_i instead of the $\varphi_i(\boldsymbol{r})$. The OP components define the order parameter space ε_n that is irreducible invariant space by the group G_0. The $\delta\rho$ variation of the probability density, associated with a phase transition, can be considered as a vector in the representation ε_n-space, and the components of $\eta = \{\eta_i^{eq}\}$ invariant vector, in the basis of this space, are the values of the OP that minimize the thermodynamic potential (for details see [9–11] and references therein).

Naturally, we consider the symmetry identity of η_i and $\varphi_i(\boldsymbol{r})$ as a general property which is also valid for the more general renormalization group approach. Indeed, a linear *projection operator* of a space group induces basis functions for the relevant IR in the form of the linear combinations of local atomic functions. The latter are linked to the phase transition mechanism and were selected as a result of a *regular* renormalization transformation, separating critical and non-critical variables [6,12,13]. The integral over the non-critical variables gives the equilibrium part of the free energy Φ_0, and unintegrated part forms the variational free energy (Landau potential) $\Phi_L(\delta\rho)$.

The crystal geometry analysis of the different displacive type structural phase transitions, in particular, martensitic transformations, shows that there exists a transcendental functional relation between the value of the phenomenological OP η_i and the magnitude of local atomic shifts, or the periodic character of its distortions [11,14–16]. Same type of non-linear periodic dependence was obtained for η_i as a function of probability density variation for the segregation type phase transitions [17]. One can conclude thus that the order-parameter space, denoted hereafter σ_n, in general case (i.e. for the full range of the OP variation), conceptually differs from the order-parameter space ε_n used earlier in the description of continuous phase transitions. While ε_n is a n-dimensional *vectorial* space, σ_n is a n-dimensional closed *functional* space *with boundary*, whose structure depends on the type of the variational parameters that identify the transition mechanism [11].

2.2. Phenomenological order parameter and essential variational parameters

We derive the general form of the function $\eta_i(\xi_j)$, where the set of η_i is a long-range phenomenological order parameter and ξ_j represent variational local atomic parameters, i.e. short-range order parameters (variation of probability for the segregation or disorder-order transformation, magnitude of atomic displacements for displacive type transitions etc.), by considering the problem in the functional order-parameter space σ_n.

One makes use of the usual scheme for calculating $\eta(\xi)$ by finding the solution of the Euler's variational equation $\{\delta\Phi/\delta\eta(\xi)\}=0$ that minimizes the free energy functional. The appropriate choice for the latter in the case of continuous phase transition is the classical Landau–Ginzburg functional

$$\Phi(\eta) = \int_{V_\xi} d\xi \left\{ a_1\eta(\xi)^2 + a_2\eta(\xi)^4 + g\left(\frac{d\eta}{d\xi}\right)^2 \right\} \tag{2}$$

where the integral is over a volume in the *OP space*. For the sake of simplicity we treat a single-component or effective OP, while the conjugate external field is neglected. The coefficient a_1 is conveniently assumed to be a regular function of thermodynamic variables (the temperature, pressure, etc.) and the remaining coefficients are regarded as temperature- and pressure-independent parameters. The corresponding Euler's equation takes the form:

$$\frac{\delta\Phi}{\delta\eta(\xi)} = a_1\eta(\xi) + 2a_2\eta(\xi)^3 + g\frac{d^2\eta}{d\xi^2} = 0 \tag{3}$$

The boundary conditions are $\eta(0)=0$ and $\eta'(0)=1$. The first condition indicates a coincidence of the origin points for the variables η and ξ. The second one ensures their identical behavior close to T_C, i.e. it justifies the change of variables $\xi\to\eta$ in the Landau theory. The differential Eq. (3) has exact general solution expressed as

$$\eta = \eta_0 \cdot sn[\mu(\xi - \xi_0), k] \tag{4}$$

In the above equation $\mathrm{sn}[\mu(\xi-\xi_0),\kappa]=\mathrm{sn}u$ is the elliptic sine of Jacobi, and μ and ξ_0 are the arbitrary constants [18]. With the applied boundary conditions, $\mu=1$ and $\xi_0=0$. The parameter $\kappa=\sqrt{(a_2/|g|)}$ is the *modulus* of the elliptic integral of the first kind

$$u = F(\xi, \kappa) = \int_0^{\xi} \frac{d\xi}{\sqrt{1 - \kappa^2 \sin^2 \xi}} \tag{5}$$

The phase diagram of this system can be obtained by minimizing the Landau potential

$$\Phi_L(\eta) = a_1\eta^2 + a_2\eta^4 \tag{6}$$

where the OP η has the form of Eq. (4). The minimization of Φ_L with respect to the actual variational parameter ξ is expressed by $\frac{\partial\Phi_L}{\partial\xi} = \frac{\partial\Phi_L}{\partial\eta}\cdot\frac{\partial\eta}{\partial\xi}$, giving the equation of state

$$\eta\eta'\left\{a_1 + 2a_2\eta^2\right\} = snu \cdot cnu \cdot dnu\left\{ a_1 + 2a_2 sn^2 u \right\} = 0 \tag{7}$$

where $\mathrm{cn}u = \sqrt{1-\mathrm{sn}^2 u}$ and $\mathrm{dn}u = \sqrt{(1-\mathrm{k}^2\ \sin^2\xi)}$. Eq. (7) yields three possible stable states: (i) The parent phase I for $\mathrm{sn}u=0$ (origin of the space σ); (ii) The limit, non-Landau, phase II, given by $\mathrm{cn}u=0$ ($\mathrm{sn}u=1$) (boundary of the space σ), corresponding to the fixed values η_0 of the OP; (iii)"'Landau'" phase corresponding to the standard minimization of Φ_L with respect to the OP η, whose value $\eta_2=-a_1/2a_2$ varies between 0 and η_0 (interior of the space σ). The function $\mathrm{dn}u$ has zeros only if $\kappa=1$, however, even in this case $\mathrm{dn}u$ vanishes simultaneously with $\mathrm{cn}u$ and no different solution of Eq. (7) exists.

The stability condition has the form:

$$\frac{\partial^2\Phi_L}{\partial\eta^2}\left(\frac{\partial\eta}{\partial\xi}\right)^2 + \frac{\partial\Phi_L}{\partial\eta}\frac{\partial^2\eta}{\partial\xi^2} \geq 0 \tag{8}$$

The resulting phase diagram in the plane of thermodynamic parameters (a_1,a_2) is shown in Fig. 1(a). The second-order phase transition line $a_1=0$ separates parent I and Landau III phases. The stability regions of Landau III and limit II phases adjoin along the

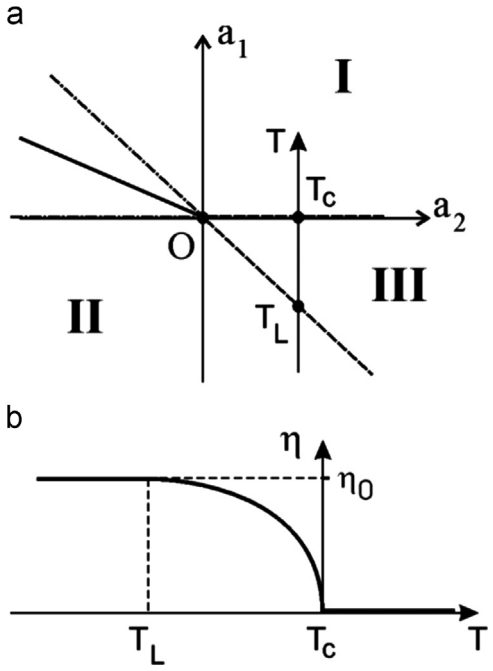

Fig. 1. (a) Phase diagram of the model Eq. (6). Solid, dashed and dashed-dotted lines are respectively discontinuous first-order transition, continuous transition, and limit of stability lines. *O* is the three-phase point. (b) Temperature dependence of the order parameter for the path indicated in (a).

line $a_1=-2a_2$. The first-order transitions line $a_1=-a_2$ between parent *I* and limit *II* phases meet in the three-phase point *O* two latter lines. Notably, in the classical Landau theory the mandatory positivity of the coefficient a_2 in the potential (6) ensures the global stability of the phases and the convexity of the potential for large values of the OP. However, Eq. (4) shows that magnitude of the OP cannot be arbitrarily large. This restriction raises the possibility of the global stable phase diagrams, even for negative values of a_2 (Fig. 1(a)) [11].

Importantly, one can see in Fig. 1(a) that the temperature range of the OP variation, that is, the stability region of Landau phase *III*, is limited. Denoting the low-temperature stability limit for the Landau phase by T_L (Fig. 1(a)), the order parameter has the value 0 above T_C, and varies between 0 and η_0 for $T_C>T>T_L$, finally reaching the saturation value η_0 (conventionally, $\eta_0=1$) at T_L and then remains invariable for $T<T_L$ (Fig.1(b)). As no conventional reasons give $T_L=0$ K, one makes use of a generalized form for the dimensionless variable $t_n=(T-T_C)/(T_C-T_L)$. The latter distinguishes the Landau approach, in which any phase has its thermodynamic limit of stability at a finite temperature (or pressure) $T_L\neq 0$ K, defined by Eq. (8) $\partial^2\Phi_L/\partial\xi^2=0$, where the corresponding energy minima disappear, and the Gibbs type consideration where the set of minima exists at any temperature up to $T=0$ K and the system finds a lowest one for any given *T*.

3. Temperature scaling transformation

3.1. OP temperature behavior

As discussed above, the Landau theory uses identical temperature dependencies for both a phenomenological OP η and atomic variable parameters ξ. This means that the Landau potential [Eq. (6)] can be introduced for the latter as well: $\tilde{\Phi}_L(\xi)=\alpha_1\xi^2+\alpha_2\xi^4$. By minimizing variational free energy $\tilde{\Phi}_L$ and assuming $\alpha_1=\alpha_{10}\cdot t$ (α_{10}=const) one finds $\xi=\sqrt{(-\alpha_1/2\alpha_2)}\propto\sqrt{t}$. In Section 2.2 we stressed the existence of the complex relation

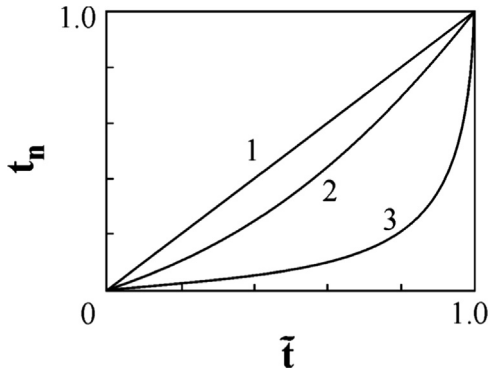

Fig. 2. Scaling ratio between temperature variables t_n and $\tilde{t}$ for different values of the modulus κ: 1 – 0, 2 – 0.8, 3 – 0.999.

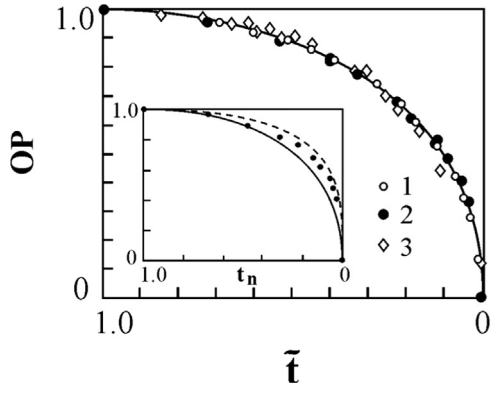

Fig. 3. Reduced values of the order parameters for (1) Sn [19], (2) β-brass [25], and (3) $PrAlO_3$ [22] as a function of rescaled temperature, and compared to the theoretical curve (solid line). For the inset see the text.

between essential variational parameters ξ_i and measurable phenomenological OP η_j. Thus, if one approximates $\xi=\tilde{\alpha}_{10}t_n^{1/2}$, then a phenomenological OP η from Eq. (4) takes form

$$\eta(t_n)=\eta_0 sn(\tilde{\alpha}_{10}\sqrt{t_n};\,k) \tag{9}$$

We consider the elliptical sine as a circular sine with the argument scaled with the integral transformation (5): $\mathrm{sn}(x,\kappa)=\sin\tilde{x}$ (factor $\pi/2$ is included in the definition of the argument) [18]. Making use of temperature as a variational thermodynamic parameter one can convert Eq. (5) into nonlinear temperature scaling transformation

$$\sqrt{\tilde{t}}=\int_0^{t_n}\frac{d(\sqrt{t_n})}{\sqrt{1-\kappa^2\,\sin^2(\sqrt{t_n})\frac{\pi}{2}}} \tag{10}$$

Nonlinearity of such a temperature scaling transformation is fully defined by the modulus κ, varying from 0 to 1. Fig. 2 shows the scaling ratios between variables t_n and $\tilde{t}$ for different values of the modulus κ.

When $\kappa=0$ the transformation is linear. It is worth reminding that the corresponding sinusoidal form for OP as a function of ξ was already employed earlier in the phenomenological models of displacive type reconstructive phase transitions in crystals [11,14–16] and of segregation transformations in complex fluids [17]. The presented discussion thus complements this approach with the temperature-controlled fluctuations.

To confirm the applicability of the presented approach to real physical systems we choose three crystals undergoing continuous phase transitions of different nature. Importantly, this choice is supported by the existence of the reliable experimental data.

(i) In metallic Sn the transition to superconducting state takes place at $T_C=3.782$ K [19]. The microscopic OP for such a transition is an effective wave function $\Psi(\mathbf{r})$ which is proportional to the local value of the energy-gap parameter Δ [20,21]. The measurable phenomenological OP is then the average normalized energy gap in the elementary excitation spectrum of a superconductor [19].

(ii) The cooperative Jahn–Teller transition with the ordering in the electronic degrees of freedom reduces the symmetry of the perovskite $PrAlO_3$ crystal from tetragonal to monoclinic at $T_C=151$ K. The electronic functions of the lowest crystal-field double degenerated level E_g of the Pr^{3+} (in the parent cubic field) form the basis functions of the relevant IR. The observable phenomenological OP is then either the splitting between the doublet levels in the ordered phase or the linearly coupled acoustic- and optical-phonon modes of the same symmetry [22].

(iii) Disorder-to-order transformation in β-brass at $T_C=740$ K [23–25]. The deviation of average probability of atoms to occupy positions in a crystal lattice can be considered as the phenomenological OP for ordering type transitions. The local variation of the probability of occupation or, equivalently, population of the corresponding sites is then the essential variable parameter for the transition.

The temperature T_L, limiting the stability region of the Landau phase, and the modulus κ, characterizing nonlinearity of the temperature scaling transformation, are used as the fitting parameters. Fig. 3 shows the best fit for different OPs plotted versus *scaled normalized* temperature $\tilde{t}$ and compared with the worked out function $\eta(\tilde{t}) = \sin\frac{\pi}{2}\sqrt{\tilde{t}}$. Only the temperature range where $\eta\neq$const is displayed. The corresponding fitting parameters were fixed as follows: Sn ($T_L=1.236$ K; $\kappa=0.05$), $PrAlO_3$ ($T_L=61$ K; $\kappa=0$) and β-brass ($T_L=490$ K; $\kappa=0.77$).

Clearly, the predicted uniform function reproduces well the experimentally observed behavior of OPs in these different systems. To emphasize the role of the temperature scaling transformation, the experimental spots for the β-brass OP [24,26] are plotted (inset of Fig. 3) versus the *non-scaled* normalized temperature t_n. Two curves in the inset display functions $\eta_1 = \sin(\frac{\pi}{2}t_n^{1/2})$ (solid line) and $\eta_2 = \sin(\frac{\pi}{2}t_n^{1/3})$ (dashed line). It is clear that close to T_C the approximate function $\eta_2 \propto t_n^{1/3}$ better reproduces experimentally observed OP behavior, and we deduce the critical exponent $\beta\approx 1/3$ for β-brass.

3.2. Modulus of the temperature scaling transformation

At this point we see that the phenomenological approach deals with two temperature scales. One can be termed *real-temperature*. This temperature t (or, equivalently, t_n) is the *measurable* thermodynamic variable for the observable functions, describing the behavior of a system. Another, termed *rescaled* temperature $\tilde{t}$, appears in the phenomenological approach after the renormalization procedure that eliminates the local microscopic variables and replaces them with the macroscopic averaged variables, the components of the phenomenological OP. More precisely, they are the rescaled variables in the Landau theory for the temperature behavior of different physical quantities or the corresponding critical exponents. The example of β-brass (Fig. 3) clearly shows that the values for the OP critical exponent depending on the temperature scale. To compare experimental results and phenomenological predictions it is important to take into account the temperature scaling transformation Eq. (10). Fig. 4 shows the real-temperature OP critical exponent β as a function of the modulus κ of such transformation.

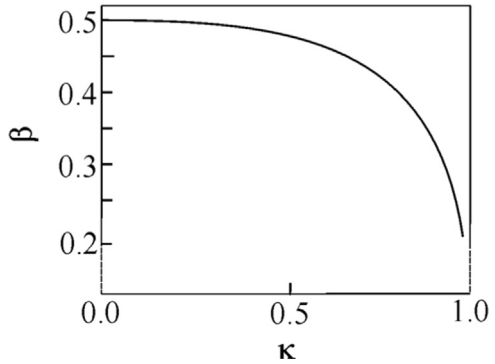

Fig. 4. Measurable real temperature order parameter critical exponent as a function of the transformation modulus κ.

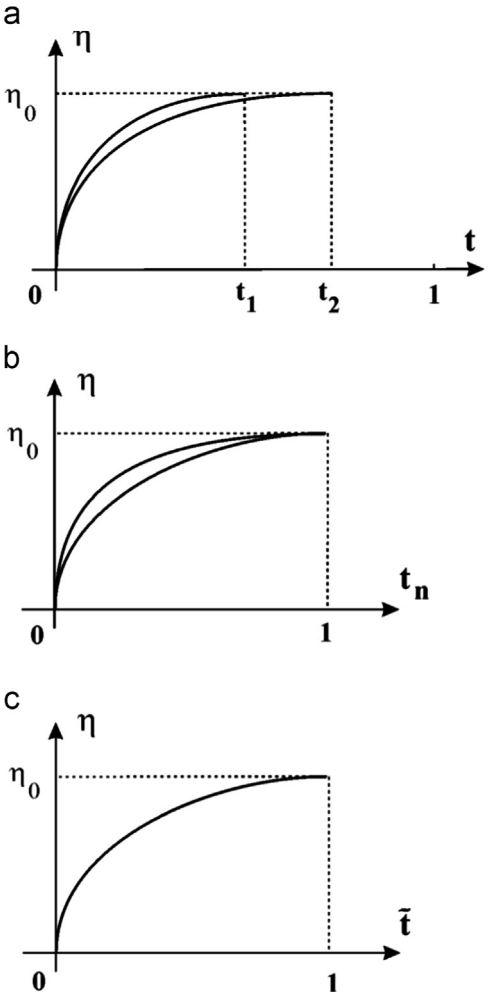

Fig. 5. Order parameter variation curves presented in different temperature scales. (a) OP as a function of a dimensionless variable $t=|T-T_C|/T_C$, (b) as a function of $t_n=(T-T_C)/(T_C-T_L)$, and (c) in function of the nonlinearly rescaled variable $\tilde{t}$.

Despite the fact that $\beta=1/2$ is unambiguously predicted in the Landau theory, we expect to obtain the *different* values for the critical exponent measured in the real-temperature scale.

Fig. 5 summarizes the modification steps passed by the experimentally measured curve $\eta(t)$ to be compared with the predictions of a phenomenological theory. Customary normalization of the temperature scale by the corresponding critical temperature T_C ($T\to t$) brings different curves to the single origin (Fig. 5(a)). Then modified normalization introduced above merges the curves at the origin $t_n=0$ ($T=T_C$) and at the saturation point $t_n=1$ ($T=T_L$) of the OP variation range (Fig. 5(b)). Finally, nonlinear temperature scaling transformation $t_n\to\tilde{t}$ fits experimental points for different systems in the uniform curve $\eta(\tilde{t}) = \sin\frac{\pi}{2}\sqrt{\tilde{t}}$ (Fig. 5(c)), predicted

in the extended phenomenological approach.

To elucidate the physical meaning of the modulus κ of the nonlinear transformation (10) we consider the response function $\tilde{G}(\xi) = (4\pi g\xi)^{-1}e^{-q\xi}$, where $q^{-2} = g\chi$, and χ is the generalized susceptibility [26]. The quantity $r_c = q^{-1}$ is the correlation length of the short-range order. The latter can be directly measured in a scattering experiment when the radiation couples to the OP [5]. By incorporating a generalized external field in Eq. (6) it is easy to calculate the function $\chi = (a_1 + 3a_2\eta^2)^{-1}$ and then derive the modulus $\kappa^2 = a_2/|g| = \varepsilon \cdot r_{c0}^{-2}$, where r_{c0} is the correlation length far from the phase transition point ($t \sim 1$) and ε is the normalizing factor. The modulus κ thus characterizes the *background* fluctuational property of a physical system (i.e. far beyond the critical region).

3.3. Landau-to-limit phase transition

We next discuss the specific features of the non-Landau limit phase and manifestation of the Landau-to-limit-state transformation. The limit phase is defined above (Section 2.2) with respect to the classical Landau phase by the OP that retains the maximal value $\eta_0 = \text{const}$ independent of the temperature variation. A considerable experimental data unambiguously indicate that such states do exist in various physical systems. The examples of Section 3.1 provide typical cases of the temperature behavior, predicted here (Fig. 1(b)). It should be stressed that for a given system in the possible but not necessarily perfect limit state the degree of order $\eta_0 \leq 1$ can be realized. This means that the limit phase is the state with the temperature-independent stable equilibrium between perfect order and partial disorder. One deals with a *saturated* state, perfectly ordered is a particular case. The latter property along with the absence at T_L of a singularity in the function $\eta(t)$ provides the emergence of the Landau-to-limit-state transition. One can show that the measurable thermodynamic functions such as entropy, latent heat or specific heat are proportional to $\partial\eta/\partial t = \eta_0 \cos \frac{\pi}{2}\tilde{t}$. However, this latter has no singularities and goes to zero at T_L (or, equivalently, at $\tilde{t} = 1$). Thus, these functions don't show any jump-like behavior, divergence or different singularities at the corresponding transition point. From this, it is clear that the Landau-to-limit-state transformation can be identified neither as first nor as second order phase transition.

4. Conclusions and outlooks

Two assumptions should be considered to ensure the generality of the presented approach. Firstly, only the case of the single-dimensional OP was treated in this work. However, the temperature behavior of the OP in the Landau-type phenomenological approach is controlled by the coefficient a_1 [see Eq. (2) and (6)]. It is associated with the invariant quadratic in OP components $a_1 I_1 = a_1(\eta_1^2 + ... + \eta_n^2)$ of the free energy expansion, and a_1 specifies the uniform functional form of the temperature dependence $\eta_i(t)$ for each of the multiple OP components as well as for the corresponding single-component effective OP. The Fig. 3 further confirms this as the uniform function there reproduces the temperature behavior of the single- (β-brass), two- (Sn) and three-component ($PrAlO_3$) phenomenological OPs. Secondly, here we deal with the free energy expansion, restricted to the fourth degree term in OP components, while the above argument on the uniqueness of the coefficient a_1 is still valid. Including the higher order terms modifies the stability conditions and the form of stability boundaries and transition lines in the phase diagram, but does not affect the temperature-dependent part of the free energy.

Summarizing, we have shown in the framework of an extended phenomenological approach that there exists a uniform function describing the OP temperature variation. The renormalization procedure, eliminating the local atomic variables and replacing them by the phenomenological order parameter components, is accompanied by the nonlinear temperature scaling transformation. *Rescaled* temperature is an essential thermodynamic variational parameter of the Landau theory with respect to which mean-field critical exponents are predicted unambiguously while the corresponding experimental values are obtained in the *real-temperature* scale. The general form of such transformation is found to be the elliptic integral of the first kind.

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