

## **TWO POINT SCALING APPROACH TO PHASE TRANSITIONS**

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### **Abstract**

The two-point scaling approach is introduced by the assumption that the thermodynamic potentials are generalized homogeneous functions with respect to the reduced temperature variable and to the fields conjugated to the order parameters, however, the singularities are related to the stability points in contrast to the conventional scaling where the fixed point is identified with the phase transition temperature.

The extended scaling theory is illustrated in the case of the pyroelectric function behaviour in the neighbourhood of ferro-paraelectric phase transitions. The method is successfully applied to the description of the melting and surface melting phenomena. Applications to liquid crystals and mixtures of solvents can be predicted as fruitful but they still remain open for considerations.

**Keywords:** phase transition, thermodynamics, two-point scaling approach

### **Introduction**

The scaling hypothesis is based on the assumption that the critical parts of thermodynamic functions are generalized homogeneous functions with respect to the reduced temperature variable whose value is scaled to the point in which the thermodynamic functions become singular and to the fields conjugated to the corresponding order parameters.

The conventional approach assumes that the singularities are connected with the critical temperature at which the phase transition occurs. The idea intuitively introduced [1-2] was confirmed by the theoretical considerations within the renormalization group theory and widely applied to various problems of phase transitions [2-4].

The scaling hypothesis turned out a very fruitful method for numerous investigations not only for the phase transitions but for other aspects of the thermodynamics. Sometimes, it is called the fourth thermodynamical law. The idea was developed for different generalizations while its background is well established.

The conventional scaling approach describes in fact the continuous phase transitions although the idea is extended to the case of discontinuous order parameter behaviours at the phase transition point. One of the ways to this purpose

is the so-called two-point scaling model which is based on the non-conventional predictions which allow us to take into account the intuitively evident features of thermodynamically inhomogeneous samples.

The two-point scaling approach seems a simple and natural extension of the scaling theory. It is formulated by the assumption that the thermodynamic potential functions are generalized homogeneous functions with respect to the reduced temperature variable and fields conjugated to the order parameters. Now, however, the singularities are related to the stability conditions. In this case we can see that the fixed points are related to the stability points of a considered phase, so that the scale of the reduced variables is different for different phases. The phase transition temperature is then determined by the natural conditions of the equality of the thermodynamical potentials and belongs to the phase coexistence interval defined by the stability points.

The extended form of the scaling hypothesis was originally introduced in order to describe the systems with fluctuations which make a system thermodynamically inhomogeneous [5–6]. Next, the two-point scaling construction was applied to the systems with restricted dimensions where the notion of the phase coexistence region plays an essential role. The surface of a sample leads then to the temperature interval which vanishes when the sample becomes infinite [7].

Recently, the discussed approach is suggested to be applied for crystals in order to describe their thermodynamic behaviour in the case of melting and pre-melting phenomena [8]. The present contribution brings also further considerations concerning the surface melting description.

Applications to liquid crystals and mixtures of solvents can be predicted as fruitful but they still remain open for considerations.

## Two-point scaling construction

The extended scaling hypothesis is based on the assumption that the thermodynamic potentials are generalized homogeneous functions with respect to their thermodynamic variables, i.e. the reduced temperature variables  $\varepsilon_s$  and fields  $h$  corresponding to the order parameters  $m_s$ .

The condition which is added in order to extend the scaling hypothesis can be formulated as the stability condition satisfied by the considered thermodynamic potential.

In the generalized formulation of the two-point scaling approach we consider a system which can appear in  $n$  phases confined by the stability points  $T_{S\nu}$ ,  $\nu \in (1, n)$ , respectively. The phase  $\nu$  is then described by means of the thermodynamic functions

$$G_\nu(\varepsilon_\nu, h, m_\nu) = G(\varepsilon, h, m_\nu) \quad (1)$$

for which the equations of state read

$$\delta G_\nu(\varepsilon_\nu, h, m_\nu) = 0 \quad (2)$$

while the order parameter  $m_v$  determining the phase  $v$  is one of  $n$  solutions for  $\delta G=0$ . It is simultaneously given by the thermodynamic relation

$$m_v = - \frac{\partial G_v}{\partial h} \quad (3)$$

Next, the stability condition is of its usual form

$$\delta^2 G_v(\varepsilon_v, h, m_v) = 0 \quad (4)$$

Assuming that,  $G_v$  is a homogeneous function with respect to  $\varepsilon_v$ ,  $h$ ,  $m_v$  we obtain as the result, consequent to the homogeneity assumption, that the equation of state (2), the order parameter (3), as well as the stability condition (4) are also homogeneous functions of the considered variables.

The homogeneity relation satisfied by  $G_v(\varepsilon_v, h, m_v)$  can be expressed as

$$\lambda G_v(\varepsilon_v, h, m_v) = G(\lambda^{a_v} \varepsilon_v, \lambda^{b_v} h, \lambda^{c_v} m_v) \quad (5)$$

for an arbitrary  $\lambda$  and three indices,  $a_v$ ,  $b_v$ ,  $c_v$  non vanishing simultaneously. Thus,

$$\delta G(\varepsilon_v, h, m_v) = \lambda^{c_v-1} \delta G(\lambda^{a_v} \varepsilon_v, \lambda^{b_v} h, \lambda^{c_v} m_v) \quad (6)$$

and

$$\delta^2 G(\varepsilon_v, h, m_v) = \lambda^{2c_v-1} \delta^2 G(\lambda^{a_v} \varepsilon_v, \lambda^{b_v} h, \lambda^{c_v} m_v) \quad (7)$$

with

$$m_v(\varepsilon_v, h) = \lambda^{b_v-1} (\lambda^{a_v} \varepsilon_v, \lambda^{b_v} h) \quad (8)$$

where  $c_v + b_v = 1$ . Substituting  $\lambda = m_v^{-1/c_v}$  we obtain

$$m^{(1-c_v)/c_v} \delta G \left( \frac{\varepsilon_v}{m_v^{a_v/c_v}}, \frac{h}{m_v^{b_v/c_v}}, 1 \right) = 0 \quad (9)$$

and

$$m_v^{(1-2c_v)/c_v} \delta^2 G \left( \frac{\varepsilon_v}{m_v^{a_v/c_v}}, \frac{h}{m_v^{b_v/c_v}}, 1 \right) = 0 \quad (10)$$

hence, for  $h=0$ , the homogeneity properties lead to the result

$$m_v = c_1^{c_v/b_v} \varepsilon_v^{c_v/b_v} \quad (11)$$

and

$$m_v = c_2^{c_v/a_v} [\epsilon_v(T_{sv})]^{c_v/a_v} \quad (12)$$

where  $c_1$  and  $c_2$  are the solutions of the equations  $\delta G(c_1, 0, 1) = 0$  and  $\delta^2 G(c_2, 0, 1) = 0$ , respectively. In general,  $c_1 \neq c_2$ , the relation (12) is then satisfied only for the scaling temperature  $T_s = T_{sv}$ , where  $T_{sv}$  denotes the stability temperature for the phase  $v$ . In this way, we derived that the thermodynamic potential is a homogeneous function with respect to  $\epsilon = |T - T_s|/T_s$ ; it really is, and then and only then if the scaling temperature  $T_s = T_{sv}$  denotes the stability temperature for the phase  $v$ .

Taking into account the above considerations we can see that the scaling point can be equal to the phase transition point  $T_c$  only in the case when  $T_{sv} = T_c$  which is satisfied for the conditions of the conventional scaling approach. In this sense we can speak of the present construction as an extension of the conventional scaling theory.

The case  $T_c \neq T_{sv}$  implied that  $G_v \neq G_{v'}$ , in general, but the condition

$$G_v(\epsilon_v(T_c), h) = G_{v'}(\epsilon_{v'}(T_c), h) \quad (13)$$

determines the phase transition temperature between the phases  $v$  and  $v'$  in every case. From the physical point of view the assumption (13) is natural and it reflects the fundamental property of thermodynamic potential. Within the scaling approach we can derive that

$$G_v(1, 0) \left( \frac{T_v - T_c}{T_v} \right)^{1/a_v} = G_{v'}(1, 0) \left( \frac{T_c - T_{v'}}{T_{v'}} \right)^{1/a_{v'}} \quad (14)$$

for  $h=0$ . In particular case when  $a_v = a_{v'} = a$  we obtain

$$T_c = (1 + g^a) T_{s2} / \left( 1 + \frac{T_{s2}}{T_{s1}} g^a \right) \quad (15)$$

for  $g = G_{s2}(1, 0)/G_{s1}(1, 0)$  and  $T_{s2} > T_{s1}$ . The phase transition temperature  $T_c$  belongs to the interval  $(T_{s1}, T_{s2})$ . In the case when  $T_{s1} = T_{s2}$  we obtain  $T_{s1} = T_c = T_{s2}$  which corresponds to the phase transition of the second order where the order parameter is continuous at the phase transition point  $T_c$ . This result obtained within the two-point scaling construction reduces the present construction to its homologous case derived from the conventional scaling theory.

## Order parameter shape coefficient

The basic thermodynamic functions and parameters can be now discussed in the light of the considered construction. The most typical quantities are the specific heat, the order parameter, the generalized susceptibility and the correla-

tions whose behaviour is tightly connected with the susceptibility shape and the stability points.

The illustrative picture for the temperature dependence of the specific heat and the magnetic susceptibility is presented in Fig. 1 and Fig. 2, respectively. One of the curves corresponds to the conventional scaling which shows its typical and well accepted result that the considered quantities are singular at  $T_c$  and the specific heat does not appear in the phase above  $T_c$ . The second curve represents the typical result of two-point scaling which predicts maximum of the specific heat as well as the susceptibility at  $T_c$ . The decrease of the specific heat above  $T_c$  is also expected but it is still appearing in contrast to the conventional description. Comparing these characteristic curves with experimental data representative for almost all the systems [e.g. 9–10] we find that the agreement between theory and experiment is favourable for the extended form of the scaling hypothesis. We can also interpret the measurements as still not sufficiently perfect but, on the other hand, the specific heat non appearing in a wide interval of temperature does not seem physically reasonable.

A similar situation can be analysed for the order parameter behaviour. Its typical character can be reflected by means of the order parameter shape coefficient whose temperature dependence is more evident and convincing for the shape of the order parameter close to  $T_c$ . The order parameter shape coefficient is introduced as

$$\lambda(\varepsilon_s, h) = \left| \frac{dm(\varepsilon_s, h)}{d\varepsilon} \frac{1}{T_s} \right| \quad (16)$$

and it can be measured independently way from the measurements of the order parameter. In particular, the coefficient  $\lambda(\varepsilon_{s1}, h)$  is very useful in the case of ferroelectrics and then it is identified with the pyroelectric function. On Fig. 3 we show its temperature dependence related to the order parameter shape. We can see that the characteristics of the results given by the considered forms of the scaling theory are of the same type as for the specific heat behaviour. A comparison of the results predicted by the two-point scaling procedure with the experimental points seems to suggest the assumption made already for the extended scaling construction (the illustrative results are fitted for ferroelectric Si Sb samples [11–12]). Thus, the assumption is confirmed in self-consistent way.

One of interesting applications of the shape coefficient analysis is connected with the prediction of a special type of phase transitions. Namely, we can consider the continuous phase transition in the sense of the order parameter behaviour, but the diffuse phase transitions – in the sense of the coexistence temperature interval.

## Application to ferroelectrics

The presented method is particularly convenient in the case of the phase transition description in ferroelectrics when the observations are performed by

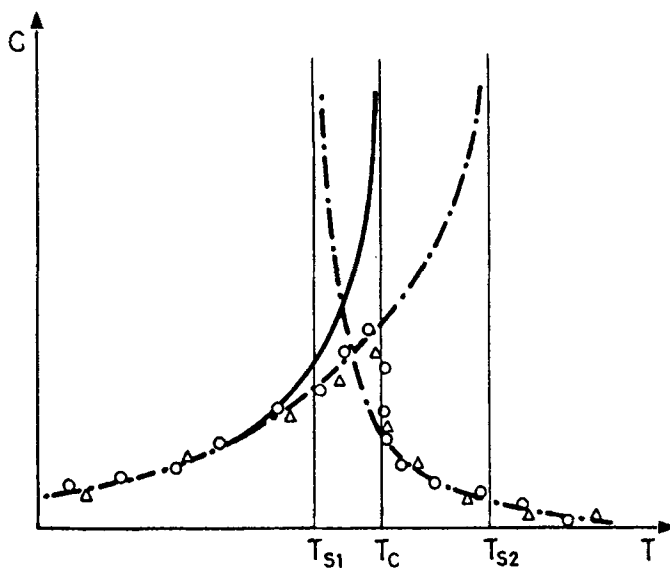


Fig. 1 Specific heat behaviour corresponding to the conventional scaling (continuous curve) and to the two-point scaling (dashed curve) compared with experimental points [9]

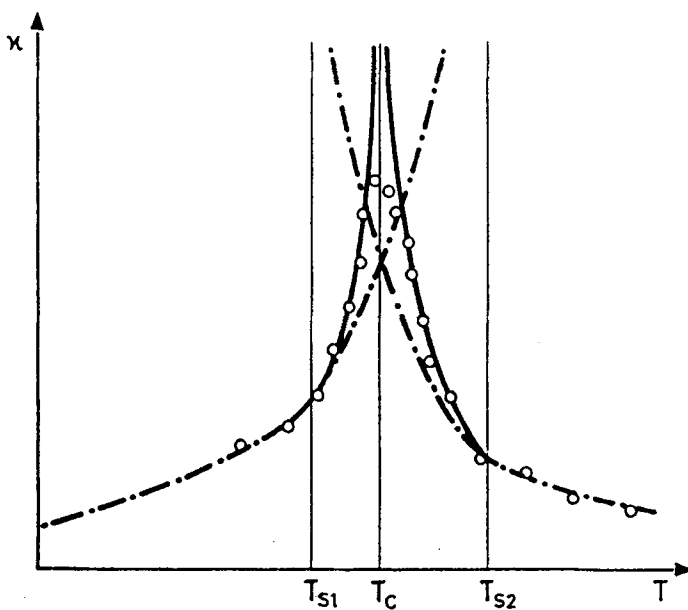


Fig. 2 Magnetic susceptibility behaviour corresponding to the conventional scaling (continuous curve) and to the two-point scaling (dashed curve) compared with experimental points [10]

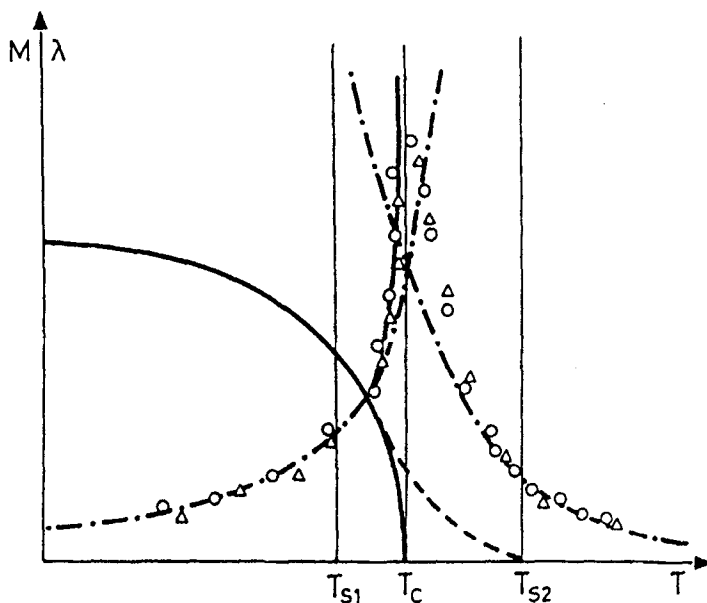


Fig. 3 Temperature dependence of the order parameter shape coefficient corresponding to the order parameter and described by the conventional scaling (continuous curve) as well as by the two-point scaling (dashed curve) compared with experimental data for the spontaneous polarization [11-12]

means of the pyroelectric function measurements. From the experimental point of view the pyroelectric coefficient in ferroelectrics is related to the dc discharge current from a sample of a known electrode area subjected to a controlled rate of the change of temperature [11]. The measurement results are determined then by the absolute value of the polarization derivative with respect to the reduced temperature (16).

It is worth-while stressing that the pyroelectric function allows us to determine the tail of the polarization with the high precision. For this reason the approach based on the pyroelectric coefficient measurements is fruitful for the analysis of the diffuse phase transition effects.

We can see now that the pyroelectric function has interesting properties for its discussion within the two-point scaling approach describing the behaviour of ferroelectrics. Namely, the temperature dependence of the pyroelectric function reflects directly the polarization behaviour which is given by the same coefficient  $m_v(1, 0)$ , the same exponents  $\beta_v = (1 - b_v)/a_v$  (c.f. (8)) as well as the reference temperatures  $T_{av}$ . Moreover, taking into account that the maximum of the pyroelectric function occurs at  $T_c$ , we obtain some relations between the parameters characterizing the considered sample. In this case the two point scaling approach allows us to compare the experimental behaviour of the pyroelectric function and the spontaneous polarization derived from another

method and to confirm that the pyroelectric function can be considered in fact as the polarization derivative. First of all, however, we can see that the pyroelectric function (16) on Fig. 3 reflects its experimental shape. It seems to us that this fact evidently confirms the intuitively introduced hypothesis.

### Application to surface melting

The present construction was also applied to the description of the coexistence conditions of the solid-liquid system with an intermediate phase which is observed as a solid-like or liquid-like layer in the surface melting effect. This description has required the extension of the presented approach to the case of three-phase coexistence. The construction is discussed in connection with the generalized compressibility and the specific heat behaviour [8].

The standard methods show that the dependence between the free energies for the solid and liquid phase in the stability interval is an almost linear function of temperature related to the latent heat  $L_m$  for melting per unit volume. We can write [13-14]

$$\Delta G = G_l^{(T)} - G_s^{(T)} = L_m \left( \frac{T_m - T}{T_m} \right) \quad (17)$$

or

$$\Delta G = G_l(T_m) - G_s(T_m) + T_m \left( \frac{\partial}{\partial T} \right) (G_l(T_m) - G_s(T_m)) \left( \frac{T_m - T}{T_m} \right) + 0 \left( 1 - \frac{T}{T_m} \right) \quad (18)$$

hence

$$L_m = T_m (\partial/\partial T) (G_l(T_m) - G_s(T_m)) \quad (19)$$

On the other hand, the latent heat can be expressed by means of the thermodynamic potential when the scaling procedure is taken into account. We obtain [8]

$$a \frac{L_m}{T_m} = \left( \frac{1}{T_s} \varepsilon_s^{1/\kappa-1}(T_m) \right) G_s(1, 0) - \left( \frac{1}{T_l} \varepsilon_l^{1/\kappa-1}(T_m) \right) G_l(1, 0) \quad (20)$$

while the jump of the specific heat is then given by

$$\frac{\Delta C_m}{T_m} \frac{a^2}{1-a} = \left( \frac{1}{T_s^2} \varepsilon_s^{1/\kappa-2}(T_m) \right) G_s(1, 0) - \left( \frac{1}{T_l^2} \varepsilon_l^{1/\kappa-2}(T_m) \right) G_l(1, 0) \quad (21)$$

where  $T_l$  and  $T_s$  stand for the stability temperatures of the liquid and solid phases, respectively.

The Eqs (20) and (21) allow us to estimate  $G_s(1, 0)$  and  $G_l(1, 0)$  by means of the physical characteristics of a considered sample, namely, a jump of the specific heat  $\Delta C_m$  and the latent heat  $L_m$ . Substituting the solutions for  $G_s(1, 0)$



and  $G_1(1, 0)$  into (20) we find the equation for  $T_m$  which can serve to fit also  $T_s$  and  $T_l$  or the exponent  $a$  when the melting temperature is assumed to be the third characteristic of a sample.

One of the properties seems to be of particular interest, namely, the fact that the phase transition temperature  $T_{m1}$  between the surface melting phase and the melting phase turns out always higher than the bulk melting temperature  $T_m$  obtained in the case when the surface effects are not considered. The problem is not so simple, but its nature is rather clear and due to the size effect which appears in the systems with restricted dimensions. The discussion concerning the global character of the melting point allows us to conclude that the surface influences the melting by shifting the bulk melting temperature towards lower temperatures. The temperature  $T_{m1}$  results then from a competition between the size, or surface, and the proximity effects. This fact can be predicted to be verified empirically when we take into account that the different shift of the phase transition temperature with respect to the crystallographic orientation is expected. The compensation does not occur then for all the orientations simultaneously. In consequence, the empirical melting temperature should depend on the geometry of experiment and the method used for the measurements.

On Fig. 4 we can see the established behaviour of the order parameter related to the density variable for the solid and liquid phases as well as the pre-

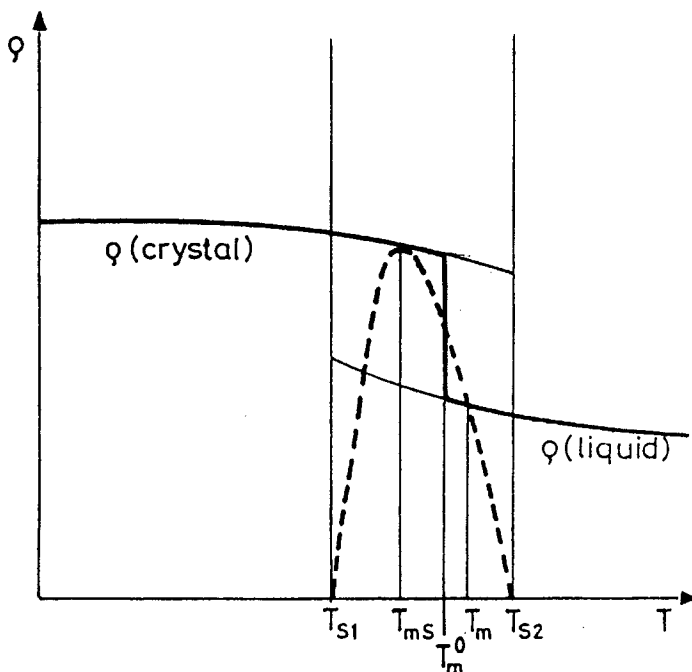


Fig. 4 The behaviour of density in the neighbourhood of phase transition. The point  $T_m^0$  corresponds to the transition with a jump of the density from  $\rho_c$  to  $\rho_l$ . The dashed curve corresponds to the surface melting phase with the transition at  $T_{ms}$  and  $T_m$  points

dicted shape inside the intermediate phase where the fragment of the trajectory between the pre-melting and the melting points is only realised. We can also illustrate the physical interpretation for the change of the phase transition character. The curve with a jump at  $T_m$  representing the first order transition is replaced by the curves which are tangent at  $T_{om}$  and/or  $T_{im}$ . In this case the phase transition at the surface becomes the transition of the second type. This property is expected in experiments [13].

## Conclusions

The generalized scaling hypothesis discussed during the present lecture is grounded on the fact that the singularity points of thermodynamic functions do not agree with the physical points of phase transitions but they coincide with the stability points. Only in some particular cases the stability and phase transition points appear at the same point and then the two-point scaling reduces to the conventional scaling theory.

The presented approach is controversial from the point of view of traditional formulations of the thermodynamics but it is verified now by numerous applications in practical studies. Therefore, it seems to me that this hypothesis should be open for further discussion.

From the point of view of the reality of a physical system the objects considered in statistical physics cannot be free of deviations from the state of homogeneity. The spatial inhomogeneities in any real system are its immanent feature. They are, of course, of different character.

The inhomogeneities of a system of static character are connected with geometrical structure which is determined by the surfaces, structural defects and dislocations. A physical structure can be another cause of static inhomogeneities. It is also worth-while mentioning here, for instance, the impurities leading to a stoichiometric disorder in alloys and local deviations of interaction constants. The static inhomogeneities can be caused by external factors, like applied fields or a temperature gradient. The spatial distribution of static inhomogeneities does not depend on time or it changes very slowly with time in comparison with the time of the measurements. This can be met in the case of the migration process for dislocations or diffusion of impurities.

The presented construction is also applicable to the case of dynamic scaling. The inhomogeneities of dynamic character depend on the geometrical stability of a system. Any physical system is inhomogeneous due to the permanent motion of particles in a fluid as well as to thermal oscillations of atoms around their equilibrium positions or other collective excitations in solids. First of all, local stochastic fluctuations are a source of dynamic inhomogeneities. A local fluctuation is comprehensible as a deviation of a certain thermodynamic variable from its mean value in certain volume element of a system with the spatial distribution which is of dynamical character with time characteristics given by the fluctuation decay and its growth parameters. In general, the characteristics

of local fluctuations depend on intensive variables like temperature, pressure, and external fields. Particularly, the temperature dependence is intuitively understandable in the case of temperature phase transitions when the local fluctuations play an essential role in the vicinity of phase transition temperature.

In the light of the above considerations any real system is spatially inhomogeneous due to local fluctuations. The fluctuations grow and decay in time and instantaneous picture of inhomogeneities changes but the system remains incessantly inhomogeneous although the average local fluctuation disappears; however, the non-zero mean square local fluctuation can be determined. Thus, the thermodynamic quantities which are non-linear, at least quadratic functions of fluctuating parameters, depend on dynamic inhomogeneities due to local fluctuations. This dependence can be observed for an arbitrarily long time, independently of the fact that the average fluctuation is equal to zero. A confirmation of such a dependence can be found in the observations of critical opalescence phenomenon as well as in the case of surface melting studies in connection with the solid-like and liquid-like layers and the transition between them. Other examples of the experimental verification refer to magnetic, electric, structural and size dependent effects in the neighbourhood of phase transitions.

The presented approach does not remain in contrast to the conventional theories which discuss the phases and phase transitions of surfaces and interfaces in various aspects. One of them is to emphasize the symmetry characterisation of surfaces and interfaces [15], another one concerns the roughening transitions [16] which are competitive to the melting and particularly surface melting phenomena. The interface between the ordered and the disordered phases becomes delocalised as in the melting transition. In this case critical surface phenomena can be described by the critical exponents but their definition requires to consider systems which undergo a first order bulk transition whose behaviour is convenient to be described in terms of the scaling theory [17]. A rigorous theory of finite-size scaling at the first order phase transitions is then formulated [18]. The starting point comes then from the obvious observation that the number of stable phases has a local maximum at the transition point, which means no singularities in it. The relation between the singular part of the energy and the singularity of the correlation length [19] in the context of the algebra for fluctuation operators [20] leads to an additional argument that the fixed point following from the identity between the singular points of energy and correlation is delayed in general to the stability temperature. This fact remains in full analogy with the main feature of the present construction.

The picture of inhomogeneous system brings a complementary description of the background for the hypothesis of two-point scaling. However, independently of these additional arguments, the reported formal remarks and, first of all, the interesting, empirically regular convergences between the two-point scaling conclusions and the behaviour of physical objects can be really stimulating for a deeper reflection concerning the presented considerations.

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The present paper is financially supported by the State Committee for Research grant (KBN grant 2 0194 91 01/92–94). Warm thanks are due to Professor S. Taniewska-Osińska for her encouraging discussion. I am also grateful to Professor H. Piekarski for his kind invitation to pres-ent this contribution at the VI Conference of Calorimetry and Thermal Analysis in Zakopane '94.

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**Zusammenfassung** — Unter der Annahme, daß die thermodynamischen Potentiale verallgemeinerte homogene Funktionen im Bezug auf die reduzierte Temperaturvariable und die zu den Ordnungsparametern gehörenden Felder sind, wurde die Zwei-Punkte-Scaling-Näherung eingeführt, Besonderheiten werden im Gegensatz zu der herkömmlichen Scaling-Methode, wo der Fixpunkt durch die Phasenumwandlungstemperatur festgelegt wird, den Stabilitätspunkten zugeordnet.

Die erweiterte Scaling-Theorie wird im Falle des Verhaltens der pyroelektrischen Funktion in der Nachbarschaft der ferro-paraelektrischen Phasenumwandlung illustriert. Die Methode wird erfolgreich zur Beschreibung von Schmelz- und Oberflächenschmelzerscheinungen angewendet. Anwendungen bei Flüssigkeitskristallen und Lösungsgemischen können als fruchtbar vorausgesagt werden, bedürfen jedoch noch weiterer Betrachtungen.