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Applicability of Landau's theory to phase transitions in the vicinity of a tetracritical point

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The structure of the more and of the less symmetric phases between which a phase transition occurs are regarded as being distorted with respect to a still more symmetric phase, which is called the parent phase. The distortions of structure of the phases are described by means of a single irreducible representation of the symmetry group of the parent phase. It is shown that when these distortions are small, i.e., when the phase transitions are close to a multicritical point on the phase diagram, the constants of Landau's theory for the transitions under consideration have anomalous values, which show up both in the temperature variations of thermodynamic quantities and in the narrowness of the range of inapplicability of Landau's theory. Within the framework of the Landau theory, these phase transitions can be only transitions of second order. The treatment is carried through for the case of an arbitrary type of two-dimensional irreducible representation of the symmetry group of the parent phase.

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It is known that Landau's[1] phenomenological theory applies well to structural phase transitions (so far it has not been possible to detect with certainty, for any crystal, a range of inapplicability of this theory). Moreover, the material constants of crystals contain no small parameter that might enable us to demonstrate a physical reason for the narrowness of this region, as is the case, for example, for phase transitions into the superconducting state.[2] As will be shown below, it is possible to demonstrate such a reason for a quite broad class of structural phase transitions. It is known that the structures of many crystals are often only slightly distorted with respect to more symmetric structures: the so-called pseudocubic and pseudohexagonal crystals and others. Such a slight distortion can also serve as the small parameter that determines the good applicability of Landau's theory to the structural phase transitions under consideration. In other words, the set of constants that characterizes the region of inapplicability of Landau's theory is a small quantity in proportion to the smallness of the distortion; and the other constants, determining the temperature variations of thermodynamic quantities, are anomalous.

Phase transitions from a more symmetric phase to a less symmetric will be treated below within the framework of Landau's theory; not, however, on the basis of a thermodynamic potential written for the more symmetric phase, but on the basis of the thermodynamic potential of a still more symmetric phase—the "parent phase,"[3] with respect to whose structure the more and the less symmetric phases are only slightly distorted.

For definiteness, we shall consider phase transitions between phases that correspond to a single two-dimensional irreducible representation of the parent phase.

All possible two-dimensional (mathematically) irreducible representations of the space groups of crystal symmetry can be classified according to the degree of one of the two independent invariants. Designating the basis functions of the representation by η and ξ and introducing suitable polar coordinates ρ and φ,

$$\eta = \rho \cos \varphi, \quad \xi = \rho \sin \varphi, \tag{1}$$

we can express the independent invariants I and I' in the form[4]

$$I = \rho^2, \quad I' = \rho^n \cos n\varphi, \tag{2}$$

where n is an integer; $n \geq 3$. Thus the two-dimensional irreducible representation, designated hereafter by E_n, can be characterized by the number n.

The thermodynamic potential Φ for the representation E_n can be written as a series of powers of the invariants I and I' of (2):

$$\Phi = \alpha I + \beta I^2 + \gamma I^3 + \ldots + \alpha' I' + \beta' I'^2 + \ldots + \beta'' II' + \ldots \tag{3}$$

To this potential, as follows from the condition that Φ shall be an extremum with respect to the variable φ,

$$\partial \Phi / \partial \varphi = \Phi_\varphi = \Phi_{I'} I_\varphi' = (\alpha' + 2\beta' I' + \beta'' I)(-n\rho^n \sin n\varphi) = 0,$$

correspond four types of solution:

$$
\begin{aligned}
&0)\ \rho = 0, \\
&1),\ 2)\ \sin n\varphi = 0, \quad \cos n\varphi = \mp 1, \\
&3)\ \cos n\varphi = -\frac{\alpha' + \beta'' \rho^2}{2\beta' \rho^n},
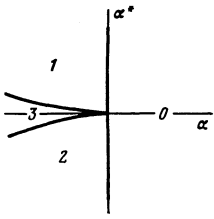\end{aligned}
\tag{4}
$$

FIG. 1. Stability limits of phases 0, 1, 2, and 3 in the $\alpha\alpha^*$ plane ($\alpha^* = \alpha' + \beta''\rho^2$) for the thermodynamic potential (3); $\beta > 0$, $\beta' > 0$.

corresponding to four phases: the parent phase 0 and the three less symmetric phases 1, 2, and 3. The symmetry groups of phases 1, 2, and 3 are subgroups of the symmetry group of phase 0 ($G_{1,2,3} \subset G_0$); furthermore $G_3 \subset G_{1,2}$, but the groups G_1 and G_2 are not subgroups of each other. It is obvious that there are n domains in each of phases 1 and 2 (n different values of φ: $\varphi = \pi(2m+1)/n$ in phase 1 and $\varphi = \pi 2m/n$ in phase 2, where $m = 0, 1, \ldots, (n-1)$) and $2n$ domains in phase 3.

Figure 1 shows the phase diagram in the plane α, α' (or $\alpha'' = \alpha^* = \alpha'' + \beta''\rho^2$) for fixed values of the remaining coefficients of the thermodynamic potential (3). The phase diagram in the temperature-pressure plane will be topologically similar. We note a peculiarity of the phase diagram, the existence of a tetracritical point, determined by the coordinates $\alpha = 0$, $\alpha' = 0$ (see Fig. 1). We emphasize that the phase diagram with tetracritical point represented here differs from the phase diagrams considered by Liu and Fisher[5] and by Bruce and Aharony[6]; there η and ξ transform according to two different one-dimensional representations of the symmetry group of the ordered phase (a phase diagram with a tetracritical point was first considered for two one-dimensional representations in Landau's paper[7]; see also Lifshitz[8] and Landau and Lifshitz[1]).

The stability limit of phase 0 is $\alpha = 0$. The stability limits of phase 3 coincide with its existence limits, which are determined by the conditions $\cos n\varphi = \mp 1$ or, from (4), by the conditions

$$\alpha' + \beta''\rho^2 \mp 2\beta'\rho^n = 0. \tag{5}$$

Phase 3 is stable if the inequality

$$\beta' \geqslant \beta''^2/4(\beta + 3\gamma\rho^2) > 0, \tag{6}$$

is satisfied; this follows from the conditions

$$\Phi_{\varphi\varphi} = I_\varphi'^2\Phi_{I'I'} = (n\rho^n \sin n\varphi)^2 2\beta' \geqslant 0,$$

$$\Phi_{\rho\rho}\Phi_{\varphi\varphi} - \Phi_{\rho\varphi}^2 = I_\rho^2 I_\varphi'^2(\Phi_{II}\Phi_{I'I'} - \Phi_{II'}^2)$$

$$= (2\rho)^2(n\rho^n \sin n\varphi)^2[(2\beta + 6\gamma\rho^2)2\beta' - \beta''^2] \geqslant 0.$$

The stability limits of phases 1 and 2 coincide with the stability limits of phase 3 and of phase 0; the latter is correct for $n > 3$ and $\beta > 0$. When $\beta < 0$, the phase transition from phase 0 is of first order, and a tetracritical point does not exist. For $n = 3$, a tetracritical point exists, but it is an isolated point of transitions of second order.[9,10] (A phase diagram with a nonisolated tetracritical point for a two-dimensional representation, of the type E_4, was considered in Ref. 3.) We note that if the Lifschitz condition is not satisfied, i.e. if there is a gradient invariant of the form[1]

$$\eta\xi_x - \xi\eta_x = \rho^2\varphi_x, \tag{7}$$

then there is no tetracritical point on the phase dia-

gram, because in this region of phase space an incommensurable phase will be stable (see Ref. 11 for E_4 and Ref. 12 for E_6). We shall suppose hereafter that the coefficient β and the coefficient β' of (6) are positive.

We are interested in the phase transitions $1 \to 3$ and $3 \to 3$. For their occurrence, as is seen from Fig. 1, a sufficient assumption is that only the coefficient α' varies with temperature (at fixed pressure), according to the linear law

$$\alpha' = \alpha_T'(T - \Theta). \tag{8}$$

Then the transition temperatures Θ_1 and Θ_2 are determined from the condition (5):

$$\Theta_{1,2} = \Theta - \frac{\beta''}{\alpha_T'}\rho_s^2 \pm \frac{2\beta'}{\alpha_T'}\rho_s^n. \tag{9}$$

We expand the thermodynamic potential (3), near the points $T = \Theta_{1,2}$ of phase transitions $1, 2 \to 3$, in the derivation of φ from its equilibrium values in phases 1, 2; we denote the deviation also by φ. As a result we get

$$\Phi = \Phi_0 + A\varphi^2 + B\varphi^4 + D(\nabla\varphi)^2,$$

where

$$A = \pm\tfrac{1}{2}n^2\rho_s^n\alpha_T'(T - \Theta_{1,2}), \quad B = \tfrac{1}{6}n^4\rho_s^{2n}\beta', \quad D = \delta\rho_s^2,$$

$$\Phi_0 = \alpha\rho_s^2 + \beta\rho_s^4 + \gamma\rho_s^6 \mp \alpha'\rho_s^n + \beta'\rho_s^{2n} \mp \beta''\rho_s^{n+2} + \delta(\nabla\rho_s)^2. \tag{10}$$

Here, in addition to (3), we have taken into account the gauge invariant

$$\delta[(\nabla\eta)^2 + (\nabla\xi)^2].$$

In the vicinity of the tetracritical point, i.e., for values of the coefficient α that are not too large in magnitude, we may neglect the coefficients δ'' and γ and also, in a number of cases, the coefficients α' and β'. Then for the spontaneous value ρ_s we obtain, in all the phases 1, 2, and 3, the expression

$$\rho_s^2 = -\alpha/2\beta, \tag{11}$$

and by (10), $\Phi_0 = -\alpha^2/4\beta$. The value (11) of ρ_s is small and decreases with approach to the tetractitical point on the phase diagram.

As is evident from the expression (10), we may take as order parameter for the transitions $1, 2 \to 3$ the quantity φ (or $\rho_s\varphi$). We emphasize a peculiarity of the phase transitions $1, 2 \to 3$. They can be (within the framework of the Landau theory and near the tetracritical point) only transitions of second order, as long as the coefficient B of φ^4 in the thermodynamic potential (10) is positive. Otherwise ($\beta' < 0$) the phase 3 would be unstable by (6), and a phase transition $1 \to 2$ of first order would occur.

The applicability of the Landau theory to the phase transitions $1, 2 \to 3$ is determined by a dimensionless parameter[13,2,1] which, as is evident from the thermodynamic potential (10), has the value

$$\frac{B^2\Theta_{1,2}}{|A_T|D^3} = \frac{\beta'^2\Theta_{1,2}}{|\alpha_T'|\delta^3}\frac{n^6}{8}\rho_s^{3(n-2)}. \tag{12}$$

The parameter may be anomalously small. In fact, it is proportional to the small quantity $\rho_s^{3(n-2)}$, which de-

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creases on approach to the tetracritical point according to the law $|\alpha|^{3(n-2)/2}$. In the immediate vicinity of the tetracritical point, besides fluctuations of the order parameter φ, fluctuations of the value of ρ_s begin to play a role in the phase transitions 1, 2, →3 (because of the closeness of the phase transitions 0→1, 2, 3). But it may be expected that there will be a region of phase space in which the fluctuations of ρ_s are already insignificant, while the parameter (12), which determines the role of the fluctuations of the order parameter φ for the phase transitions 1, 2→3, is still anomalously small. This region is obviously larger, the larger the number n.

The closeness of the phase transitions 1, 2→3 to the tetracritical point determines, along with the parameter (12) that describes the applicability of the Landau theory, anomalous values of other thermodynamic quantities also; of these, we shall consider the specific heat and the susceptibility. The discontinuity ΔC of the specific heat in the phase transitions 1, 2→3 is determined from (10):

$$\Delta C = A_T^2 \Theta_{1,2}/2B = \alpha_T'^2 \Theta_{1,2}/2\beta' ; \qquad (13)$$

it is independent of ρ_s and should therefore have the usual orders of magnitude.

The susceptibility $\chi = \partial\rho_s\varphi/\partial h$, where h is the intensity of the field conjugate to the order parameter η, ξ for the transitions 0→1, 2 (a term $-\rho_s\varphi h$ in the thermodynamic potential (10)), is determined in phases 1, 2, as follows from (10), by the relation

$$\chi^{-1} = n^2 \rho_s^{n-1} \alpha_T'(T - \Theta_{1,2}). \qquad (14)$$

The Curie constant

$$C = (n^2 \rho_s^{n-1} \alpha_T')^{-1}$$

in the Curie-Weiss law (14) is inversely proportional to the small quantity ρ_s^{n-2} and therefore is anomalously large. We emphasize that for phase transitions of the shift type, the inverse susceptibility χ^{-1} of (14) is proportional to the elasticity of the normal oscillations $\rho_s\varphi$; in other words, the square of the frequency of a soft mode is anomalously small for the transitions 1, 2→3.

If the quantity $\rho_s\varphi$ is a component of a polarization vector or of a deformation tensor, i.e. if the representation E_n is contained in a vectorial representation or in a symmetrized square of a vectorial representation of the symmetry group of phase 0, then χ of (14) is a dielectric or mechanical susceptibility, easy to measure experimentally. If this is not so, then the expression (14) does not determine either a dielectric or a mechanical susceptibility; these may, however, be determined by other expressions (see below). In this connection, those quantities are of considerable interest that occur linearly in the thermodynamic potential (3), coupled with an arbitrary power of components of the order parameter η, ξ (or ρ, φ). In order to find such quantities, we ask how arbitrary symmetrized powers of the representation E_n transform. This can be seen from tables compiled for the E_n with the smallest n, odd ($n=3$) and even ($n=4$); tables for other E_n, for example for E_6, have a similar form (see Table I). Since pro-

TABLE I. Transformation of the quantities $\rho^m \cos m\varphi$ and $\rho^m \sin m\varphi$ for the two-dimensional representations $E_n (n = 3, 4, 6)$ according to which the quantities $\rho \cos\varphi = \eta$ and $\rho \sin\varphi = \xi$ of (1) transform.

Representation*	Degree of the basis of the representation**							
	2	1	2	3	4	5	6	...
n = 3								
A	$\rho^2 c_0$			$\rho^3 c_3$			$\rho^6 c_6$	
B_3	$\rho^2 s_0$			$\rho^3 s_3$			$\rho^6 s_6$	
E_3		ρc_1 / ρs_1	$-\rho^2 c_2$ / $\rho^2 s_2$		$\rho^4 c_4$ / $\rho^4 s_4$	$-\rho^5 c_5$ / $\rho^5 s_5$		...
n = 4								
A	$\rho^2 c_0$				$\rho^4 c_4$			
B_3	$\rho^2 s_0$				$\rho^4 s_4$			
E_4		ρc_1 / ρs_1		$-\rho^3 c_3$ / $\rho^3 s_3$		$\rho^5 c_5$ / $\rho^5 s_5$		...
B_2			$\rho^2 c_2$				$\rho^6 c_6$	
B_1			$\rho^2 s_2$				$\rho^6 s_6$	
n = 6								
A	$\rho^2 c_0$						$\rho^6 c_6$	...
B_3	$\rho^3 s_0$						$\rho^6 s_6$	...
E_6		ρc_1 / ρs_1				$-\rho^5 c_5$ / $\rho^5 s_5$		
E_3			$\rho^2 c_2$ / $\rho^2 s_2$		$-\rho^4 c_4$ / $\rho^4 s_4$			
B_2				$\rho^3 c_3$				
B_1				$\rho^3 s_3$				

*$B_{1,2,3}$ are one-dimensional representations; A is the identical representation.
**For conciseness of notation, the abbreviations $c_m \equiv \cos m\varphi$ and $s_m \equiv \sin m\varphi$ are used.

ducts of arbitrary powers of the cosine and sine can be expressed in terms of cosines and sines of multiple angles, the tables show how products of arbitrary powers of η and ξ transform. The antisymmetric square of the representation E_n, i.e. the quantity $\eta\xi - \xi\eta = \rho^2 \sin\theta\varphi$, transforms according to the representation B_3.

In the phase 3 of lowest symmetry, those quantities are different from zero, i.e. have spontaneous values, that transform according to any of the representations of the corresponding table. In phase 1 or 2, as is evident from the solutions (4), quantities transforming according to the one-dimensional representations B_3 and B_2 or B_1 are zero, i.e. do not have spontaneous values. Hence it follows that these quantities (we denote them by P_3, P_2, P_1) are linearly coupled with the order parameter φ. In other words, the thermodynamic potential (10) contains the invariants

$$\mp A_3 P_3 \varphi \mp A_{2,1} P_{2,1} \varphi,$$

$$A_3 = a_3 n \rho_s^n, \qquad A_{2,1} = a_{2,1} \frac{n}{2} \rho_s^{n/2}. \qquad (15)$$

These invariants are obtained from the mixed invariants

$$a_1 P_1 \rho^{n/2} \sin \tfrac{1}{2}n\varphi + a_2 P_2 \rho^{n/2} \cos \tfrac{1}{2}n\varphi + a_3 P_3 \rho^n \sin n\varphi, \qquad (16)$$

which, as follows from the tables, are contained in the thermodynamic potential (3) (for odd n there is obviously only the invariant with the coefficient a_3). There are also other mixed invariants (for $n > 4$; see Table I), but they are of no interest here.

The quantities P_1, P_2, and P_3, which transform according to the one-dimensional representations B_1, B_2,

and B_3 and assume spontaneous values in the phase transitions $0\to1,2,3$, we shall call "improper" quantities with respect to these transitions; it is now customary to apply that term to the transitions themselves in the literature on ferroelectric and ferroelastic materials.[14] The quantities η and ξ we shall call "proper" with respect to the transitions $0\to1,2,3$. The quantity φ, or $\rho_s\varphi$, is proper for the phase transition $1\to3$ (or $2\to3$). The quantities P_2 and P_3 (or P_1 and P_3) may be called "pseudoproper" with respect to the transition $1\to3$ (or $2\to3$). These quantities transform like the order parameter φ, according to the same one-dimensional representation of the symmetry group of phase 1 (or of phase 2).

The one-dimensional representations $B_{1,2,3}$ may be contained in a vector representation or a symmetrized square of a vector representation of the symmetry group of phase 0; that is, the quantities $P_{1,2,3}$ may have the meaning of components of a polarization vector or of a strain tensor. We shall find the susceptibility

$$\chi_{1,2,3}=\frac{\partial P_{1,2,3}}{\partial E_{1,2,3}},$$

we designate by $E_{1,2,3}$ the intensities of the fields conjugate to the quantities $P_{1,2,3}$. Adding to the thermodynamic potential (3), and consequently also to (10), and (15), terms

$$\varkappa_1 P_1{}^2+\varkappa_2 P_2{}^2+\varkappa_3 P_3{}^2-P_1E_1-P_2E_2-P_3E_3,\qquad(17)$$

we find in phase 1

$$\chi_2(E_{1,3}=0)=\frac{A_2{}^2}{8\varkappa_2{}^2A_T(T-\Theta_1)}+\frac{1}{2\varkappa_2}=\frac{a_2{}^2}{16\varkappa_2{}^2\alpha_T{}'(T-\Theta_1)}+\frac{1}{2\varkappa_2},$$
$$\chi_3(E_{1,2}=0)=\frac{A_3{}^2}{8\varkappa_3{}^2A_T(T-\Theta_1)}+\frac{1}{2\varkappa_3}=\frac{a_3{}^2\rho_s{}^n}{4\varkappa_3{}^2\alpha_T{}'(T-\Theta_1)}+\frac{1}{2\varkappa_3}\qquad(18)$$

and similarly in phase 2 with the substitution $1\leftrightarrow2$. The susceptibility χ_1 in phase 1 (or χ_2 in phase 2) is not of interest, because the Curie-Weiss law is not obeyed. We note that when the terms (17) are taken into account in the thermodynamic potential, the expressions for the transition temperatures $\Theta_{1,2}$ in Eq. (14) for $\chi(E_{1,2,3}=0)$ and in Eq. (18) differ from (9) by a term

$$\pm\frac{A_{2,1}^2}{4\varkappa_{2,1}A_T}\pm\frac{A_3{}^2}{4\varkappa_3A_T}=\pm\frac{a_{2,1}^2}{8\varkappa_{2,1}\alpha_T{}'}\pm\frac{a_3{}^2\rho_s{}^n}{2\varkappa_3\alpha_T{}'}.$$

The Curie constants $C_{2,1}=a_{2,1}/16\varkappa_{2,1}\alpha_T{}'$ for the susceptibilities $\chi_{2;1}$ of (18) are independent of ρ_s and should therefore have the usual orders of magnitude. The Curie constant $C_3=a_3{}^2\rho_s{}^n/4\varkappa_3{}^2\alpha_T{}'$ for χ_3 of (18) is proportional to $\rho_s{}^n$ and should assume anomalously small values. For transitions of the shift type this is due, as is evident from (18), to the fact that in phase 1 (or in phase 2) the contributions of the soft mode $(\rho_s\varphi)$ to P_2 (or to P_1) and to P_3, i.e. the soft-mode oscillator strengths, are proportional to $\rho_s{}^{n-2}$ and to $\rho_s{}^{2n-2}$ respectively [see (15)], whereas the square of the soft-mode frequency is proportional to $\rho_s{}^{n-2}$ by (14).

Thus, we emphasize, while the square of the soft-mode frequency in phase transitions $1,2\to3$ of the shift type, which is determined by the expression (14), has an anomalously small value, the Curie constant in the Curie-Weiss law for a dielectric or mechanical susceptibility may have either an anomalously large value (14) (a proper ferroelectric or ferrelastic phase transition) or ordinary or anomalously small values (18) (a pseudoproper transition).

An analogous treatment can be carried out for representations or more than two dimensions. It is also obvious that all the results presented above are valid also for magnetic transitions (in a region still far removed from saturation ρ_s), when η and ξ transform according to two-dimensional irreducible representations of the magnetic space groups. Such phase transitions are usually called spin-reorientation transitions.[15]

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