

Phase transitions with coupled order parameters

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A broad set of phenomena in which the coupling of several order parameters is essential is examined from a unitary standpoint on the basis of the fluctuational theory of phase transitions. The coupling of an order parameter with other degrees of freedom can lead to the appearance on the phase transition lines of singular (polycritical) points, near which the character of the anomalies changes. We analyze numerous experimental studies near polycritical points, whose results as a whole confirm the conclusions of fluctuational theory. The studied objects include ^3He – ^4He solutions, antiferromagnetics, ammonium halides, liquid crystals, and mercury near the liquid-gas critical point. The main result of studying systems with coupled order parameters is a substantial expansion of the possibility of a universal description of real phase transitions, including those that have been objects of more or less separate fields of physics, owing to their complexity.

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1. INTRODUCTION

Only several years have passed since the solution of the problem of critical phenomena (second-order phase transitions)—one of the most difficult problems of modern macroscopic physics. The universality of the behavior of objects of the most varied nature is the most attractive result of the fluctuational theory of phase transitions (see Refs. 1–5). Critical anomalies are characterized by a set of universal constants (critical exponents) that depend only on the type of symmetry of the order parameter. As a rule, only relatively simple models have been studied theoretically (e.g., the

Ising and Heisenberg models). In particular, when the order parameters of a model and a real system have the same symmetry, universality implies that the singularities of the corresponding thermodynamic quantities must be the same (isomorphic). Therefore, differences in the behavior of the thermodynamic quantities of real and model systems (if they exist) can be associated with the coupling of the order parameter in real systems with other degrees of freedom that were not taken into account in the simplest models.

In recent years, a rather large number of both theoretical and experimental studies has been devoted to the

problem of taking into account the coupling of the order parameter with any other variables. We can briefly characterize their fundamental results as follows: coupling of the order parameter with other degrees of freedom leads to:

- 1) appearance of second-order phase-transition lines where isomorphism is maintained;
- 2) appearance on these lines of singular (polycritical) points, near which the character of the anomalies changes;
- 3) breakdown of the second-order transition into a first-order transition.

We should stress that the experimental situation near polycritical points is far more complicated than in objects with ordinary critical points, where the accuracy of the determination of the critical exponents is sometimes better than the accuracy of the theoretical predictions.⁶ Hence we have restricted the treatment solely to sufficiently reliable experimental data. From among a large number of objects having polycritical points, we have selected only the ones that are most characteristic and well studied experimentally.

This includes first of all the $^3\text{He}-^4\text{He}$ solution, which has become a classical example of a system with a tricritical point, as well as the relatively well studied antiferromagnetics and ammonium halides (near their orientational transitions).

Moreover, we have treated in this review certain phase transitions which, at first glance, have nothing in common with polycritical points: the transition to the liquid-crystalline state, the liquid-vapor critical point of mercury, and the triple points in the ammonium halides. As it has turned out, in all these cases the concept of coupling of the order parameters can serve as the basis for understanding the character of these phenomena, while we can consider the transitions themselves to be distorted to some extent by polycritical points.

2. PHASE DIAGRAMS WITH POLYCRITICAL POINTS

a) Mixture of helium isotopes

Figure 1 shows the phase diagram of $^3\text{He}-^4\text{He}$ solutions under the pressure of the saturated vapors.⁷ A second-order phase transition occurs in the isotope ^4He at the temperature $T = 2.17\text{ K}$ to the superfluid state (λ -transition). Addition of the isotope ^3He lowers the tem-

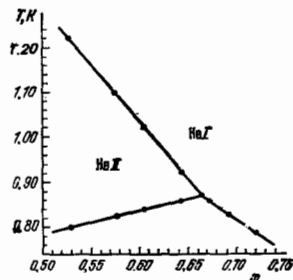


FIG. 1. Phase diagram of the $^3\text{He}-^4\text{He}$ mixture.

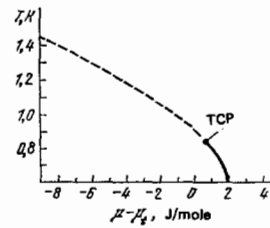


FIG. 2. Phase diagram of the $^3\text{He}-^4\text{He}$ mixture in coordinates of temperature vs chemical potential. The dotted and solid lines are respectively the second- and first-order phase transitions.

perature of the λ -transition. At a content of ^3He above 67 atom percent, the transition to the superfluid state occurs by a jump accompanied by demixing into two phases of differing composition. The superfluid phase is enriched in the isotope ^4He , and the normal phase in ^3He . The point at which the second-order phase-transition line is supplanted by a first-order transition line is called a tricritical point (Landau critical point).⁸

In Fig. 2 the same diagram⁹ is drawn in the coordinates of temperature vs chemical potential¹¹ (μ is the quantity thermodynamically conjugate to the concentration). The tricritical point (TCP) of the $^3\text{He}-^4\text{He}$ solution is the experimentally best studied object among the tricritical points.

b) Ammonium halides

The ammonium halides manifest phases with varying orientational order of the ammonium ions, depending on the temperature and the pressure (P). Figure 3 shows the phase diagram of NH_4Br .¹⁰ Three phases coexist at the triple point (β is disordered, δ shows ferromagnetic-type ordering, and γ shows antiferromagnetic-type ordering). The transitions among the phases near the point 0 are all first-order. Therefore we should not confuse this point with a tricritical point.

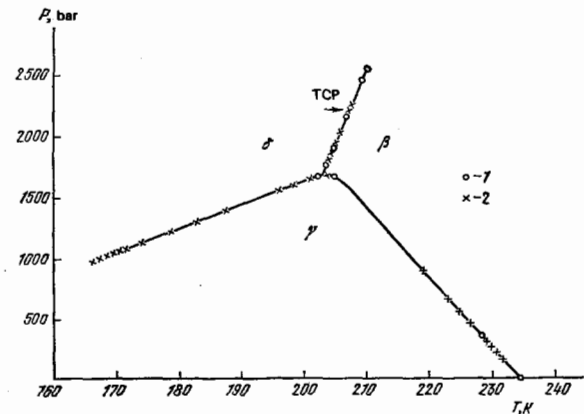


FIG. 3. Phase diagram of NH_4Br : 1—point of maximum C_p (T), 2—breakdown of the adiabatic.

¹¹More exactly, the difference of chemical potentials of the ^3He and ^4He isotopes.

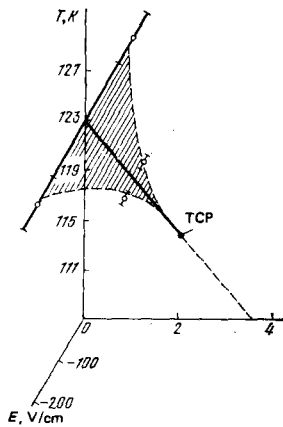


FIG. 4. Phase diagram of KH_2PO_4 . The dotted and solid lines are respectively the second- and first-order phase transitions. ///-paraelectric-ferroelectric first-order transition.

However, with increasing pressure, the β - δ transition becomes a second-order transition, and hence a TCP arises. The pressure is the same in the coexisting phases. Therefore the diagram in Fig. 3 is an analog of the diagram for ${}^3\text{He}$ - ${}^4\text{He}$ in the coordinates of T and μ . The same type of TCPs have been found on the β - δ transition lines in the other ammonium halides as well.

c) Ferroelectrics and antiferromagnetics

TCPs have also been found in ferroelectrics. KH_2PO_4 has been best studied experimentally. Figure 4 shows the phase diagram for KH_2PO_4 in the coordinates T , P , and E (electric field). A characteristic and seldom encountered feature of this case is the fact that the field E , which is thermodynamically conjugate to the order parameter (the polarization), can be realized experimentally. At the TCP three second-order transition lines converge: two lines for $E \neq 0$ and one line for $E = 0$. Hence we get the name of this point: tricritical.¹²

In strongly anisotropic antiferromagnetics, e.g., dysprosium aluminum garnet (DAG), FeCl_2 , and DyPO_4 at temperatures above the tricritical point, a continuous phase transition occurs from a homogeneously magnetized paramagnetic to the antiferromagnetic state. At lower temperatures, this transition is accompanied by a jump in the magnetization. Figure 5 shows the phase diagram of FeCl_2 in coordinates of temperature vs magnetization.^{13,14} We see that it qualitatively coincides with the diagram of the ${}^3\text{He}$ - ${}^4\text{He}$ solution (see

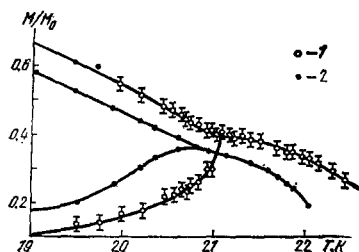


FIG. 5. Phase diagram of FeCl_2 , M_0 —saturation magnetization. 1—data of Ref. 13, 2—data of Ref. 14.

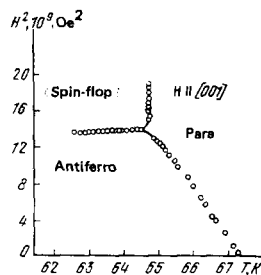


FIG. 6. Phase diagram of MnF_2 .

Fig. 1). The magnetization plays the role of the ${}^3\text{He}$ concentration here. In the coordinates of temperature vs magnetic field, the phase diagram of similar antiferromagnetics correspondingly has the same form as the diagram of Fig. 2.

In weakly anisotropic antiferromagnetics (e.g., MnF_2), the situation differs. As the magnetic field H is increased, when the antiferromagnetic ordering becomes unfavorable, the mean magnetizations of the sublattices deviate from the axis of easiest magnetization. The transition from this (spin-flop) phase to a homogeneously magnetized phase is a second-order transition. Yet the spin-flop-antiferromagnetic transition is first order. Consequently we have the phase diagram shown in Fig. 6.¹⁵ The point at which two second-order transition lines and one first-order transition line meet in this way is called a bicritical point.¹⁶

In more complicated cases, the appearance of another (intermediate) phase can occur.¹⁷ Here four second-order transition lines meet at a polycritical point. Therefore this point has been termed a tetracritical point.¹⁶

d) Solid solutions

An intermediate phase is realized in two-component solid solutions of antiferromagnetics, e.g., $\text{Fe}_x\text{Mn}_{1-x}\text{WO}_4$. The crystal structures of the two pure components FeWO_4 and MnWO_4 are very similar. At the same time, the elementary magnetic cells completely differ¹⁸ (Fig. 7). The neutron scattering of this mixture shows that reflections of both structure (a) and structure (b) are present in a certain range of concentrations x ($0.12 < x < 0.32$) (see Fig. 7).

In particular, the phase diagram of the solution $\text{K}_2\text{Mn}_{1-x}\text{Fe}_x\text{F}_4$ has been studied.¹⁹ Figure 8 shows this diagram in the coordinates of T vs x . Four second-order transition lines intersect at the point O , i.e., at a tetracritical point.

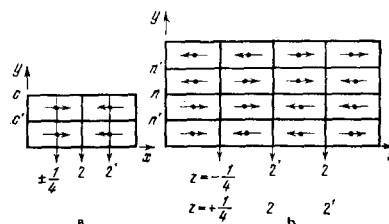


FIG. 7. Magnetic structures of FeWO_4 (a) and MnWO_4 (b).

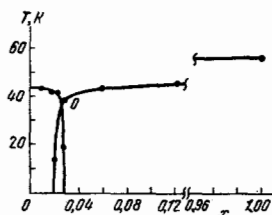


FIG. 8. Phase diagram of $K_2Mn_{1-x}Fe_xF_4$.

e) Multicomponent liquid solutions

In multicomponent liquid solutions (with a number of components $N \geq 3$), higher-order critical points exist, at which three or more phases become identical.²⁰ When three phases coincide (e.g., two liquids and one gas), this point proves to be a tricritical point.²¹

3. PHENOMENOLOGICAL DESCRIPTION OF POLYCRITICAL PHENOMENA

a) The Landau theory

Let us examine the conditions for appearance of polycritical points within the framework of the Landau theory of phase transitions,²² in which the thermodynamic potential has the form of an expansion in powers of the order parameter ϕ :

$$\Phi = \Phi_0 + \frac{1}{2} A \phi^2 + \frac{1}{3} B \phi^3 + \frac{1}{4} C \phi^4 + \frac{1}{5} D \phi^5 + \frac{1}{6} E \phi^6 + \dots \quad (3.1)$$

The coefficients of the expansion depend on the temperature, the pressure, and the other field variables (which assume identical values in coexisting phases).

First let us examine the symmetric case in which symmetry considerations rule out invariants of odd order (e.g., ϕ is a vector). If the coefficient $C > 0$, then, when $A = 0$, a second-order transition occurs. When $C < 0$, the phase transition proves to be first-order. Consequently, the thermodynamic "surface" defined by the conditions

$$A = 0, \quad C = 0, \quad (3.2)$$

is a "surface" of tricritical points. If there are only three independent variables (P, T, ϕ), then the conditions (3.2) define a single TCP (the number of degrees of freedom is $i = 0$) (Fig. 9a). If we go from any field variable to its thermodynamic conjugate (the so-called "density"), e.g., from pressure to volume, from magnetic field to magnetization, or from chemical potential to concentration, then the phase diagram having a TCP

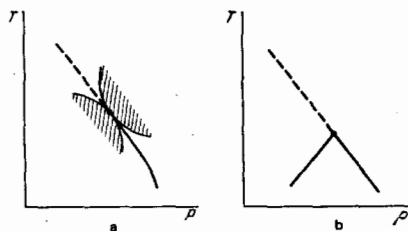


FIG. 9. Phase diagrams of systems having a tricritical point in the Landau theory in the variables of temperature vs field (a) and temperature vs "density" (b). The dotted and solid lines are respectively second- and first-order phase transitions. The region of tricritical behavior is cross-hatched.

has the form shown in Fig. 9b. The following conditions define the location of fourth-order critical points, etc. (in the terminology of Stanley *et al.*):²³

$$A = 0, \quad C = 0, \quad E = 0. \quad (3.3)$$

Here the third-order critical-point lines meet at a fourth-order critical point. In general, lines of points of order θ meet at a point of order $\theta + 1$. We note that a critical point of order θ is characterized by the fact that the first nonvanishing term in the thermodynamic potential of (3.1) is $\phi^{2\theta}$. In this terminology an ordinary critical point is a second-order critical point.

According to the Gibbs phase rule,⁸ we have

$$i = n + 1 - f. \quad (3.4)$$

Here n is the number of independent thermodynamic variables, and f is the number of coexisting phases. In the (symmetric) case being studied, we should assign the following value to the number of phases at a critical point of order θ ²³:

$$f = \theta + 1. \quad (3.5)$$

If the invariants of odd order are not identically zero, then we have²⁴

$$f = 2\theta - 1. \quad (3.6)$$

Equation (3.4) implies that the number of independent variables satisfies $n \geq f - 1$. Since f is larger in the nonsymmetric case ($2\theta - 1 \geq \theta + 1$), then the appearance of high-order critical points in the nonsymmetric case requires a larger number of independent variables than in the symmetric case. For example, a tricritical point ($\theta = 3$) in liquid solutions is possible only with a number of components equal to three (four independent variables).

Now let us examine the more general case of two coupled order parameters. We can represent the thermodynamic potential in the form

$$\Phi^*(\varphi_1, \varphi_2) = \Phi_0 + \Phi_1(\varphi_1) + \Phi_2(\varphi_2) + \Phi_{int}(\varphi_1, \varphi_2). \quad (3.7)$$

Three cases can occur: 1) ϕ_1 is a vector and ϕ_2 is a scalar; 2) ϕ_1 and ϕ_2 are both vectors; 3) ϕ_1 and ϕ_2 are both scalars.

1) ϕ_1 is a vector and ϕ_2 is a scalar:

$$\Phi^* = \Phi_0 + \frac{1}{2} A_1 \varphi_1^2 + \frac{1}{4} C_1 \varphi_1^4 + \frac{1}{6} E_1 \varphi_1^6 + \frac{1}{2} A_2 \varphi_2^2 + \lambda \varphi_1^2 \varphi_2. \quad (3.8)$$

Here we have $A_1 = a_1 \tau_1 \equiv a_1(T - T_{cl})/T_{cl}$, $A_2 = a_2 \tau_2 \equiv a_2(T - T_{c2})/T_{c2}$, and the coupling constant is $\lambda = -(a_1/2T_{cl})(dT_{cl}/d\phi_2)$. We have neglected the higher-order terms in ϕ_2 , since we assume that $T_{c2} < T_{cl}$. That is, a transition in ϕ_2 independent of the transition in ϕ_1 is not realized. In this case we can reduce the two-parameter potential of (3.8) to a one-parameter potential of the form of (3.1). Actually, we find the following expression from the condition $\partial \Phi^*/\partial \phi_2 = h_2$, where h_2 is the field thermodynamically conjugate to ϕ_2 :

$$\varphi_2 = \frac{h_2 - \lambda \varphi_1^2}{a_2 \tau_2}.$$

Let us transform to a potential that depends on the field variable h_2 :

$$\begin{aligned} \Phi(\varphi_1, h_2) &= \Phi^*(\varphi_1, \varphi_2) - h_2 \varphi_2 \\ &= \Phi_0 + \frac{1}{2} \left(a_1 \tau_1 + \frac{2h_2 \lambda}{a_2 \tau_2} \right) \varphi_1^2 + \frac{1}{4} \left(C_1 - \frac{2\lambda^2}{a_1 \tau_1} \right) \varphi_1^4 + \frac{1}{6} E_1 \varphi_1^6. \end{aligned} \quad (3.9)$$

Upon comparing this with (3.1), we find

$$A = a_1\tau_1 + \frac{2h_2\lambda}{a_2\tau_2}, \quad C = C_1 - \frac{2\lambda^2}{a_2\tau_2}, \quad E = E_1, \quad B = D \equiv 0.$$

As the temperature is lowered, τ_2 declines and the coefficient C can change sign. The conditions $A=0, C=0$ correspond to the TCP. For a fixed ϕ_2 , the TCP is determined from the conditions

$$A_1 = -\frac{2h_2\lambda}{a_2\tau_2}, \quad C_1 = \frac{2\lambda^2}{a_2\tau_2}.$$

Then the phase diagram has the form shown in Fig. 9b.

2) ϕ_1 and ϕ_2 are both vectors:

$$\Phi^* = \Phi_0 + \frac{1}{2} A_1 \phi_1^2 + \frac{1}{4} C_1 \phi_1^4 + \frac{1}{2} A_2 \phi_2^2 + \frac{1}{4} C_2 \phi_2^4 + \frac{1}{2} \lambda \phi_1^2 \phi_2^2. \quad (3.10)$$

If T_{c1} and T_{c2} depend on any variable g , then the transition lines can intersect. The intersection point will be bicritical (Fig. 10a) when $\lambda^2 \geq C_1 C_2$ and tetracritical (Fig. 10b) when $\lambda^2 < C_1 C_2$ (see Ref. 25). We note that the condition for appearance of a bicritical point is strong coupling of the order parameters.

If we minimize with respect to one of the order parameters, we again arrive at a one-parameter potential of the form of (3.1). Here, in spite of the absence of odd-order invariants in the two-parameter potential, it may turn out that such invariants will appear in the one-parameter representation. The number of independent variables in the studied case is four (T, g, ϕ_1 , and ϕ_2). The intersection point of the critical lines is invariant ($i=0$), i.e., $f=n+1=5$. In the symmetric case we have $\theta=f-1=4$, and in the nonsymmetric case $\theta=(f+1)/2=3$. It has been shown²⁶ that the tricritical-point lines meet at a bicritical point in the T, g, h_1, h_2 space, and according to Ref. 23, the bicritical point is at the same time a fourth-order critical point. No detailed study has been made of the tetracritical point.

3) ϕ_1 and ϕ_2 are both scalars:

$$\Phi^* = \Phi_0 + \Phi_1(\phi_1) + \Phi_2(\phi_2) + \lambda\phi_1\phi_2. \quad (3.11)$$

This case is characteristic of liquid mixtures. The conditions of stability have the form

$$\frac{\partial^2 \Phi^*}{\partial \phi_1^2} > 0, \quad (3.12^I)$$

$$\frac{\partial^2 \Phi^*}{\partial \phi_2^2} > 0, \quad (3.12^{II})$$

$$\frac{\partial^2 \Phi^*}{\partial \phi_1^2} \frac{\partial^2 \Phi^*}{\partial \phi_2^2} - \left(\frac{\partial^2 \Phi^*}{\partial \phi_1 \partial \phi_2} \right)^2 > 0. \quad (3.13)$$

In the previous cases we have $\partial^2 \Phi^* / \partial \phi_1 \partial \phi_2 = 0$ from the side of the disordered phase, and the stability was de-

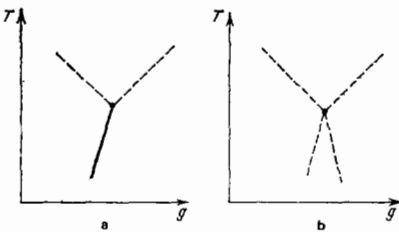


FIG. 10. Phase diagrams of systems having bicritical (a) and tetracritical (b) points in the Landau theory. The dotted and solid lines are respectively second- and first-order phase transitions.

termined by one of the expressions of (3.12). In the case under discussion with bilinear coupling of the order parameters, we have $\partial^2 \Phi^* / \partial \phi_1 \partial \phi_2 \neq 0$, and the stability is determined by the expression (3.13). That is, generally one cannot determine which of the order parameters is involved in the breakdown of stability.

The differences are revealed only in the presence of a noncritical phase, i.e., at the so-called finite critical points. For two-component mixtures the situation is simplest, since ϕ_1 can be defined in terms of the composition of the mixture (x), and ϕ_2 in terms of the density (ρ). Two types of critical points can exist in binary mixtures: liquid-gas and liquid-liquid equilibria. Both of these types are equivalent thermodynamically, and we can choose either of the parameters x or ρ as the critical parameter. However, in describing liquid-gas critical points it is convenient to choose the density as the critical parameter, since the critical-point lines end at the critical points of the components of the mixture. Upon transforming to the field variables—the chemical potential μ (or the pressure P)—we obtain the one-parameter potential $\Phi(T, \rho, \mu)$ [or $\Phi(T, x, P)$]. For a fixed value of μ (or P), this is isomorphic with the potential of an ideal system having an isolated critical point: $\Phi = \Phi(T, \rho)$ for a one-component liquid, or $\Phi = \Phi(T, x)$ for an incompressible binary mixture. For more details on the isomorphism, see Refs. 27 and 28. We must stress that isomorphism is conserved everywhere on the critical lines of two-component mixtures, and higher-order critical points are absent. In a three-component mixture, we are now dealing with critical surfaces, and correspondingly, with lines of finite critical points. These lines can meet to form a critical point of order $\theta=3$, i.e., a TCP. This is the only singular point on the critical surface. The fundamental differences of this point from a symmetric TCP are the following: there are no separate critical-point lines; one cannot single out the critical and non-critical order parameters. That is, the singularities of all the corresponding thermodynamic quantities pertaining to the different order parameters are the same. With a larger number of components, critical points of order $\theta > 3$ can arise in addition to those of order $\theta=3$. Thus, in a five-component mixture a fourth-order critical point can occur.

b) The field of applicability of the Landau theory

As we know, the Landau theory, which neglects fluctuations of the order parameter, is inapplicable in the immediate vicinity of a critical point. The Ginzburg criterion, which defines the field of applicability of the Landau theory, has the form³

$$\frac{\langle (\Delta\phi)^2 \rangle}{\phi_0^2} \ll 1. \quad (3.14)$$

Here the $\langle (\Delta\phi)^2 \rangle \sim 1/a |\tau| r_c^d$ are the fluctuations in a volume having linear dimensions of the order of the correlation radius r_c , and ϕ_0 is the equilibrium value of the order parameter. The correlation radius, as calculated under the assumption that the fluctuations are small (the Ornstein-Zernike approximation),⁸ is

$$r_c = r_0 |\tau|^{-1/2}. \quad (3.15)$$

Here r_0 is the direct correlation radius measured in units of the distance between the particles. Then the criterion (3.14) assumes the form

$$\frac{a|\tau|^{(d-2)/2}}{r_0^d \phi_0^2} \ll 1. \quad (3.16)$$

As is implied by the expansion of (3.1) with $B=D \equiv 0$, the region of tricritical behavior has $C\phi^4 \ll E\phi^6$, and the equilibrium value of the order parameter is

$$\varphi_0 = \left(\frac{a|\tau|}{E} \right)^{1/4}. \quad (3.17)$$

The Landau theory then is applicable under the condition

$$|\tau|^{(3-d)/2} \gg \frac{E^{1/2}}{a^{3/2} r_0^d}. \quad (3.18)$$

This condition is satisfied always when $d > 3$. When $d = 3$, as will be shown below, taking fluctuations into account leads to the appearance of logarithmic corrections to the results of the Landau theory. That is, the behavior remains "almost classical".

In the region of critical behavior ($C\phi^4 \gg E\phi^6$), we have

$$\varphi_0 = \left(\frac{a|\tau|}{C} \right)^{1/2}. \quad (3.19)$$

Then the Landau theory is applicable if

$$|\tau|^{(4-d)/2} \gg \frac{C}{a^2 r_0^d}. \quad (3.20)$$

This is always satisfied if $d > 4$. When $d = 3$, we have $|\tau| \gg C^2/a^4 r_0^6$, and a region of applicability of the Landau theory appears either upon a decrease in the constant C or when $r_0^2 \gg 1$ (long-range action). A broad set of objects (liquids, helium, ferromagnetics) has $r_0 \approx 1$. Then a region of applicability of the Landau theory is absent, and the fluctuational theory gives a correct description.^{3,5} The situation is also analogous near bicritical and tetracritical points.

c) The scaling hypothesis. Crossover phenomena

A characteristic feature of all the systems that we have examined is the existence of an isolated singular point (polycritical point) on the phase-transition line $T_c(g)$. Here we have introduced the generalized thermodynamic field g , which can differ in meaning for different systems (the chemical-potential difference of the components in the ^3He - ^4He mixture; the magnetic field, or more exactly, H^2 , in antiferromagnetics, etc.). The polycritical point arises when $g = g_0$. We shall denote the corresponding temperature $T = T_c(g_0)$ as T_0 .

The singularities of the thermodynamic quantities on the phase-transition lines and at the polycritical point differ. Isomorphism to an ideal system is conserved on the phase-transition lines, i.e., a system whose thermodynamic potential depends only on the temperature and on the order parameter. Isomorphism breaks down at a polycritical point. As we approach a critical point $T_c(g)$ that is close to but not coincident with the polycritical point T_0 , the behavior of the system must change from polycritical to critical. In order to describe such a transition behavior, we can employ the hypothesis of homogeneity of the thermodynamic func-

tions.²⁹ This hypothesis is a generalization of the scaling hypothesis,¹ on which the fluctuational theory of the critical point is based.

Let us represent any thermodynamic quantity A as the sum of a regular and a singular component. We shall assume that:

1) the singular component of the quantity A is a function of the dimensionless deviations of the temperature from the transition point $\tau = [T - T_c(g)]/T_c(g)$ and of the field g from the polycritical value $\Delta g = (g - g_0)/g_0$:

2) the singular component of the quantity A is a homogeneous function of its variables:

$$A_s = \tau(g)^{-a_0} f(z), \quad z = \frac{\Delta g}{\tau(g)^{1/\psi}}. \quad (3.21)$$

Here a_0 is the critical exponent that characterizes the behavior of the quantity A at the polycritical point, and ψ is the so-called crossover exponent²⁹ that characterizes the width of the transition region from polycritical to critical behavior. Here $f(z)$ is a nonanalytic function. Let us find the conditions that this function must satisfy near the polycritical ($z \ll 1$) and critical ($z \gg 1$) points. When $z \ll 1$, the anomaly of the quantity A is characterized by the exponent a_0 , i.e.,

$$f(z) = \text{const.}$$

When $z \gg 1$, the singular component of the quantity A is characterized by the exponent $a_c \neq a_0$. That is, $A_s \sim |\tau(g)|^{-a_c}$, where a_c is the critical exponent of the quantity A in an ideal system, and we have

$$f(z) = \text{const.} \cdot z^{(a_0 - a_c)\psi}.$$

The equation of the critical line is determined by the exponent ϕ and it has the form

$$T_c(g) = T_0 (1 + b\Delta g^\phi + \dots). \quad (3.22)$$

In addition to the principal nonanalytic term that we have written out, there are also terms regular in Δg .

The transition from polycritical to critical behavior occurs at $z \sim 1$, i.e., at $\tau \sim \Delta g^\phi$. The Landau theory of the tricritical point has $\phi = 2$, since the transition from the critical to the tricritical region occurs when $\tau \sim C^2/aE$, while the quantity C plays the role of Δg . The Landau theory has $\phi = 1$ at bicritical and tetracritical points.

A very essential point is that the thermodynamic functions found experimentally must be homogeneous functions of the type of (3.21) only in the case in which the experiment was actually performed at a fixed value of the field g (e.g., the difference of the chemical potentials of ^3He and ^4He). As was shown in Ref. 28, only a fixing of the field variables, as contrasted with the "densities", allows one to maintain isomorphism. Sometimes it is impossible to satisfy this condition. Thus, in the ^3He - ^4He system, all the measurements are performed at a fixed value of the concentration x of ^3He . In this case all the quantities must be expressed in terms of the parameter $\tau(x) = [T - T_c(x)]/T_c(x)$. Here the critical exponents are renormalized.

Thus the exponent that characterizes the quantity A can vary for two reasons near a polycritical point:

1) transition from the polycritical to a critical region (first type of crossover phenomena);

2) transition to a renormalization region upon approaching a critical or polycritical point along a non-isomorphic path (second type of crossover phenomena).

In the polycritical region the renormalized values of the exponents are³⁰

$$a_0^* = \begin{cases} a_0/(1-\alpha_0) & \text{when } \varphi(1-\alpha_0) > 1, \alpha_0 > 0. \\ a_0\varphi & \text{when } \varphi(1-\alpha_0) \leq 1, \varphi > 1. \\ a_0 \text{ (no renormalization)} & \text{when } \varphi(1-\alpha_0) > 1, \alpha_0 < 0 \text{ or} \\ & \varphi(1-\alpha_0) \leq 1, \varphi < 1. \end{cases} \quad (3.23)$$

In the critical region the exponent a_c is renormalized in a unique manner:

$$a_c^* = \begin{cases} a_c/(1-\alpha_c) & \text{when } \alpha_c > 0. \\ a_c \text{ (no renormalization)} & \text{when } \alpha_c < 0. \end{cases} \quad (3.24)$$

This is because $z \gg 1$ near a critical point.

The width of the renormalization region when g is the chemical potential is determined, as shown in Ref. 27, by the parameter

$$\xi \sim x \left(\frac{1}{T_c} \frac{\partial T_c}{\partial x} \right)^2.$$

For example, near a critical point renormalization arises in the region

$$\xi \tau (x)^{-\alpha_c} \gg 1. \quad (3.25)$$

Experimental study of systems near polycritical points is highly impeded, precisely by the existence of these crossover phenomena. Practically all the experiments to study polycritical points have been performed either in the first or the second transition regions.

The problem of the microscopical theory is to calculate all the critical exponents (both at the critical and the polycritical points) and the crossover function $f(z)$. In the experimental testing of this theory, it is most expedient to study the degree of homogeneity of all the thermodynamic functions, rather than the "effective" critical exponents, which vary as we approach the transition point.

4. THEORETICAL STUDY OF PHASE TRANSITIONS IN SYSTEMS HAVING SEVERAL ORDER PARAMETERS

a) Formulation of the problem

All real second-order phase transitions occur on lines or surfaces, since other degrees of freedom exist in addition to the order parameter. The only exception is the critical point of a one-component liquid. In principle, we can treat any equilibrium degree of freedom as being an additional order parameter. Thus, we can represent the Hamiltonian of the system in the form of the sum of the Hamiltonians of noncoupled subsystems, each of which is characterized by its own order parameter ϕ_m , plus the term \mathcal{H}_{int} , which takes account of the interaction between them:

$$\mathcal{H} \{ \varphi_1, \dots, \varphi_n \} = \sum_{m=1}^n \mathcal{H}_m \{ \varphi_m \} + \mathcal{H}_{\text{int}} \{ \varphi_1, \dots, \varphi_n \}. \quad (4.1)$$

Here we have

$$\mathcal{H}_m \{ \varphi_m \} = \mathcal{H}_{0m} \{ \varphi_m \} + \int d^d r \left[\sum_{k=1}^{\infty} \frac{u_m^{(k)}}{k!} \varphi_m^{2(k+1)}(r) - h_m \varphi_m(r) \right], \quad (4.2)$$

$$\mathcal{H}_{0m} \{ \varphi_m \} = \frac{1}{2} \int d^d r \left[\tau_m \varphi_m^2(r) + (\nabla \varphi_m(r))^2 \right], \quad \tau_m = \frac{T - T_{cm}}{T_{cm}}. \quad (4.3)$$

The integration is performed over the entire space of dimensionality d , and the T_{cm} are the temperatures of the transitions with respect to the parameters ϕ_m in the absence of coupling. The Hamiltonian \mathcal{H}_m is the well-known Landau Hamiltonian,⁸ while the form of \mathcal{H}_{int} is determined by the particular type of symmetry of the order parameters.

No extra restrictions are imposed on the ϕ_m in (4.1); this implies that we are operating at fixed values of the thermodynamically conjugate variables ϕ_m of the fields h_m .

According to the fluctuational theory of second-order phase transitions,^{2,3} the constants of the original Hamiltonian of (4.1) are renormalized as we approach the critical point and approach certain universal values. In the language of the theory, this implies the existence of a stable fixed point (FP) of the transformations of the renormalization group. The character of a phase transition, in particular the critical exponents, is fully determined by the character of the stable FP. As a rule, taking into account the extra variables ϕ_m leads to the appearance of new FPs, each of which has its own region of stability with respect to the constants of the Hamiltonian of (4.1). Here one of the following three cases can occur:

1) The FP corresponding to the ideal system remains stable. The phase transition proves to be isomorphic with the transition in the ideal system.

2) The old FP becomes unstable. A new FP is stable, and the character of the phase transition is altered. In the phase diagrams this corresponds to the appearance of polycritical points.

3) There are no stable FPs at all. Breakdown of a second-order into a first-order transition occurs. This case has been studied in detail in Refs. 31–33.

b) Coupling of two order parameters. Conditions for appearance of polycritical points

Let us treat the coupling Hamiltonian \mathcal{H}_{int} in the form

$$\mathcal{H}_{\text{int}} \{ \varphi_1, \varphi_2 \} = \lambda \int d^d r Q_1(r) Q_2(r). \quad (4.4)$$

Here λ is the coupling constant, and the operators $Q_m(r)$ depend only on the $\phi_m(r)$. All the cases of interest to us have the operator $Q_1(r) = \phi_1^2(r)$. Depending on the symmetry, the operator $Q_2(r)$ can be equal either to $\phi_2(r)$ (if ϕ_2 is a scalar), or to $\phi_2^2(r)$ (if ϕ_2 is a vector).

The two cases subsequently lead to substantially different results:

1) The critical temperatures of the noncoupled subsystems T_{c1} and T_{c2} differ. As we shall show below, a tricritical point can arise here in the phase diagram.

2) The critical temperatures T_{c1} and T_{c2} are the same. This condition leads to a bicritical or tetracritical point, depending on the number of components of the order parameters ϕ_1 and ϕ_2 .

In the former case ($T_{c1} \neq T_{c2}$), only one of the order

parameters proves to be critical (say, ϕ_1). Let us integrate the partition function of the system

$$Z = \int d\varphi_1 d\varphi_2 \exp(-\beta \mathcal{H}\{\varphi_1, \varphi_2\}) \quad \left(\beta = \frac{1}{k_B T}\right) \quad (4.5)$$

for $\tau_1 \ll 1$ over the noncritical mode ϕ_2 .³⁴ Thus we reduce the problem of calculating the partition function of (4.5) of a two-component system to calculating the partition function of a one-component system having the effective Hamiltonian $\mathcal{H}_{\text{eff}}\{\phi_1\}$, which is defined by the formula

$$\mathcal{H}_{\text{eff}}\{\phi_1\} = -\frac{1}{\beta} \ln \int d\varphi_2 \exp(-\beta \mathcal{H}\{\varphi_1, \varphi_2\}). \quad (4.6)$$

Upon expanding (4.6) in terms of \mathcal{H}_{int} , we find

$$\begin{aligned} \mathcal{H}_{\text{eff}}\{\phi_1\} &= \mathcal{H}_1\{\phi_1\} \\ &- \frac{i}{\beta} \sum_l \frac{(-i)^l}{l!} (\beta \lambda)^l \int B_2^{(l)}(r_1, \dots, r_l) \prod_{i=1}^l Q_1(r_i) d^d r_i. \end{aligned} \quad (4.7)$$

Here the $B_2^{(l)}$ are the irreducible l th-order means of the operators $Q_2(r)$:

$$B_2^{(l)}(r_1, \dots, r_l) = \langle\langle Q_2(r_1) \dots Q_2(r_l) \rangle\rangle. \quad (4.8)$$

The averaging in calculating the $B_2^{(l)}$ is performed using the distribution function $\exp(-\beta \mathcal{H}_2\{\phi_2\})$. The fundamental contribution to the integrals of (4.7) arises from the regions of scale $r_{\infty} \ll r_{c1}$. (The r_{c1} are the correlation radii of the fluctuations of ϕ_1 and ϕ_2 respectively). Therefore, in the principal approximation with respect to r_{∞}/r_{c1} , we can replace the products of operators $\prod_{i=1}^l Q_1(r_i)$ with $Q_1^{(l)}(r)$ in (4.7). Then the effective Hamiltonian will amount to a polynomial in the critical order parameter ϕ_1 that is analogous to the Hamiltonian $\mathcal{H}_1\{\phi_1\}$ of the ideal system, with constants that depend on the temperature and on the field thermodynamically conjugate to the parameter ϕ_2 . Naturally this leads to the appearance in the phase diagram of a line of critical points $T_c(h_2)$ of the transitions with respect to the order parameter ϕ_1 . When

$$\Delta\tau \equiv \frac{T_c(h_2) - T_{c2}}{T_{c1}} \sim 1$$

the $B_2^{(l)}$ are regular functions of T and h_2 , and the Hamiltonian $\mathcal{H}_{\text{eff}}\{\phi_1\}$ is isomorphic with the Hamiltonian $\mathcal{H}_1\{\phi_1\}$. In renormalization-group language, this means that the stable FPs of the Hamiltonian $\mathcal{H}_{\text{eff}}\{\phi_1\}$ (perturbed system) and of $\mathcal{H}_1\{\phi_1\}$ (ideal system) coincide. We must stress that isomorphism is conserved almost throughout the phase diagram and breaks down only at the singular points.

Let us find the conditions for breakdown of isomorphism. As h_2 varies, so does $T_c(h_2)$; here $T_c(h_2)$ and T_{c2} can approach, one another, and the constants of the Hamiltonian $\mathcal{H}_{\text{eff}}\{\phi_1\}$ approach the boundaries of stability of the FP. If the emergence beyond the stability region occurs at a finite (albeit small) value of $\Delta\tau$, a tricritical point will arise in the phase diagram.

Let us explain why a TCP arises as $T_c(h_2)$ approaches T_{c2} . The FP of the ideal system is stable as long as the coefficient of ϕ_1^4 is positive. Equation (4.7) implies that the increment to the fourth-order term is negative and is associated with the correlator $B_2^{(2)}$ (we recall

that $Q_1 = \phi_1^2$), which increases with decreasing τ_2 . Here the effective coefficient u_{eff} decreases and can become negative. In the case of a scalar order parameter ϕ_2 , we have $Q_2 = \phi_2$, and

$$u_{\text{eff}} = u_1 - \frac{1}{2} \beta \lambda^2 G_2(\tau_2, h_2). \quad (4.9)$$

Here $G_2(\tau_2, h_2)$ is the Green's function of the fluctuations of ϕ_2 .

The stability boundary of the FP that characterizes the phase transition in the ideal system corresponds to the vanishing of u_{eff} at a certain temperature T_t :

$$u_{\text{eff}} = u_1 - \frac{1}{2} \beta \lambda^2 G_2(\tau_{2t}, h_{2t}) = 0, \quad \tau_{2t} = \frac{T_t - T_{c2}}{T_{c2}}. \quad (4.10)$$

In other words, at the point in the phase diagram, whose temperature and field h_2 are determined by Eq. (4.10) and by the extra condition

$$T_c(h_{2t}) = T_t, \quad (4.11)$$

the second-order phase-transition curve $T_c(h_2)$ goes over into a first-order transition curve. Here T_t and h_{2t} are the coordinates of the TCP.

One can perform the analysis presented above only when the transition points T_{c1} and T_{c2} of the subsystems do not coincide. Where the two critical-point lines intersect, we have $T_{c1} = T_{c2}$, both order parameters are critical, and a transformation to an effective Hamiltonian that depends on only one of the variables is impossible. Isomorphism breaks down. As soon as we leave the intersection point, the condition $T_{c1} = T_{c2}$ no longer holds, one of the order parameters becomes noncritical, and isomorphism is conserved along the lines. Whether the intersection point is bicritical or tetracritical depends on the number of components of the order parameters.³⁴

We shall not treat in detail the breakdown of a second-order into a first-order transition. We only note three possible mechanisms of such a breakdown.

1) The transition is second order in the isomorphic variables, and first-order in the nonisomorphic. A characteristic example has been treated in Ref. 31, where the coupling of the order parameter with phonons was taken into account (Goldstone mode, i.e., everywhere critical). Here the physical variables (P, T) proved to be nonisomorphic, and the constant u_{eff} becomes negative upon approaching the transition point.

2) A stable FP exists, but the constants of the original Hamiltonian lie outside its region of stability.³²

3) There are no stable FPs. For example, this case is realized in the coupling of critical order parameters of the same dimensionality. This leads to the appearance of additional fourth-order invariants, in particular, the square of the scalar product of the order parameters.³³

5. TRICRITICAL POINTS

a) Theory

Let us introduce the tricritical exponents, or the exponents of the power-function dependences of the ther-

modynamic quantities near the TCP, following Ref. 35 (Table I). The signs + and - pertain respectively to the right-hand and left-hand branches of the first-order transition in the coordinates T and ϕ_2 (see Fig. 1 and Table II). The exponent κ defines the form of the critical lines at zero ordering field h_1 . In all the remaining cases in Table I we assume that $h_1 = 0$.

Among the 19 tricritical exponents that we have introduced, only three, e.g., α_t, γ_t , and ϕ , are independent. The rest can be calculated by using the following relationships:

1) The relationships of scaling theory¹:

$$\begin{aligned} \alpha_t + 2\beta_t + \gamma_t &= 2, \quad \beta_t(\delta_t - 1) = \gamma_t, \quad 2 - \alpha_t = d\nu_t, \\ \gamma_t &= \nu_t(2 - \eta_t), \quad \gamma_+ = \gamma_- = \gamma_u, \quad \delta_+ = \delta_-, \quad \beta_+ = \beta_- = \beta_u, \\ d_u + 2\beta_u + \gamma_u &= 2, \quad \beta_+(\delta_+ - 1) = \gamma_+. \end{aligned} \quad (5.1)$$

2) The relationships that stem from the renormalization upon transforming to nonisomorphic variables^{30,2)}

$$\gamma_t^* = \gamma_t \varphi, \quad \nu_t^* = \nu_t \varphi, \quad \kappa = (\beta_t \delta_t \varphi)^{-1}, \quad \gamma_u = \alpha_t \varphi, \quad \beta_u = (1 - \alpha_t) \varphi. \quad (5.2)$$

Let us study the form of the phase diagram and the thermodynamics near the TCP by starting with the effective Hamiltonian of (4.7). In momentum space the latter has the form³⁾

$$\begin{aligned} \tilde{\mathcal{H}}_{\text{eff}}\{\varphi_1\} &= \int d^d k \left\{ \frac{1}{2} [\tau_1(h_2) + k^2] |\varphi_1(k)|^2 + \frac{u(h_2)}{4!} \prod_{i=1}^4 \varphi_1(k_i) \delta\left(\sum_{i=1}^4 k_i\right) + \frac{v}{6!} \prod_{i=1}^6 \varphi_1(k_i) \delta\left(\sum_{i=1}^6 k_i\right) \right\} - h_1 \varphi_1(k=0). \end{aligned} \quad (5.3)$$

We must retain the term proportional to ϕ_1^6 , owing to the sign change of $u(h_2)$ upon passing through the TCP.

Let us transform in (5.3) to the new variable

$$\varphi_1(k) = \tilde{\varphi}(k) + \sigma_0 \delta(k).$$

Also let us choose σ_0 in such a way that the cubic term in $\tilde{\varphi}$ in the Hamiltonian $\tilde{\mathcal{H}}_{\text{eff}}\{\tilde{\varphi}\}$ vanishes. Then, in addition to the λ -line

$$\sigma_0 = \tau_1(h_2) = h_1 = 0 \quad (\text{for } u(h_2) > 0), \quad (5.4)$$

we find that two other second-order phase-transition lines meet at the TCP when $h_1 \neq 0$:

$$\begin{aligned} \sigma_0 &= \pm \left(-\frac{6u}{v}\right)^{1/2}, \quad h_1 = \pm \left(-\frac{6u}{v}\right)^{1/\kappa} \cdot \frac{v}{45}, \quad \kappa = \frac{2}{5} \\ & \quad (\text{for } u(h_2) < 0). \end{aligned} \quad (5.5)$$

Since the exponent κ satisfies $\kappa < 1$, all three critical lines meet at the TCP.

In calculating the critical exponents and the equations of state, it is convenient to single out the terms in the Hamiltonian of (5.3) that correspond to the mean value of the order parameter $\phi_{10} = \phi_1(k=0)$. Then we eliminate the quantity $\tau_1(h_2)$ from the equations for the ordering field h_1 and the susceptibility χ_1 :

$$h_1 = \left\langle \frac{\partial \tilde{\mathcal{H}}_{\text{eff}}}{\partial \varphi_{10}} \right\rangle, \quad \chi_1^{-1} = \frac{\partial h_1}{\partial \varphi_{10}}. \quad (5.6)$$

²⁾As was noted in Sec. 2, such a renormalization occurs under the condition $\phi(1 - \alpha_t) \leq 1$, which is satisfied at the TCP ($\phi = 2$, $\alpha_t = 1/2$).

³⁾The further treatment in this section mainly follows Ref. 36, to which we refer the reader for details.

TABLE I. Tricritical exponents.

Thermodynamic quantity	Symbols	Thermodynamic path	Functional relationship
a) Quantities associated with the critical order parameter			
Susceptibility	$\chi_1 = \frac{\partial \varphi_1}{\partial h_1}$	$h_2 = h_{2t}$ $\varphi_2 = \varphi_{2t}$	$\tau^{-\nu_t}$ $\tau^{-\nu_t^*}$
Order parameter	φ_1	$h_2 = h_{2t}, T < T_t$ $h_2 = h_{2t}, T = T_t$	τ^{β_t} $h_1^{\nu_t}$
Heat capacity	C_{h_2}	$h_2 = h_{2t}$	$\tau^{-\alpha_t}$
Correlation radius of the fluctuations	r_{c1}	$h_2 = h_{2t}$ $\varphi_2 = \varphi_{2t}$	$\tau^{-\nu_t}$ $\tau^{-\nu_t^*}$
Correlation function	$G_1(k)$	$h_2 = h_{2t}, T = T_t$	$k^{2-\eta_t}$
Critical lines in nonzero ordering field	$T_c - T_t$	—	$h_1^{1/\kappa}$
First-order transition lines for $h_1 = 0$	$T_{tr}(h_2) - T_c(h_2)$	—	$(h_2 - h_{2t})^\varphi$
b) Quantities associated with the noncritical order parameter			
Susceptibility	$\chi_2 = \frac{\partial \varphi_2}{\partial h_2}$	$\varphi_2 = \varphi_{2t}, T > T_t$ $\varphi_2 = \varphi_{2t}^+, T < T_t$ $\varphi_2 = \varphi_{2t}^-, T < T_t$	$\tau^{-\nu_u}$ $\tau^{-\nu_+}$ $\tau^{-\nu_-}$
Order parameter	$\varphi_2^+ - \varphi_2^-$ $\varphi_2^+ - \varphi_{2t}$ $\varphi_{2t} - \varphi_2^-$ $\varphi_2 - \varphi_{2t}$ $\varphi_{2t} - \varphi_2$	$T < T_t$ $T < T_t$ $T < T_t$ $\varphi_2 > \varphi_{2t}, T = T_t$ $\varphi_2 < \varphi_{2t}, T = T_t$	τ^{β_u} τ^{β_+} τ^{β_-} $(h_2 - h_{2t})^{1/\delta_+}$ $(h_2 - h_{2t})^{1/\delta_-}$
Heat capacity	C_{φ_2}	$\varphi_2 = \varphi_{2t}$	$\tau^{-\alpha_u}$

Thus we obtain the parametric equation of state proposed by Migdal to describe the singularities of the thermodynamic quantities near second-order transition points:

$$h_1 = \chi_1^{-1-\beta/\nu} \psi(m), \quad \varphi_{10} = \chi_1^{-\beta/\nu} m. \quad (5.7)$$

Here we have

$$\psi(m) = m - g(\chi_1) m^3 - \frac{4}{5!} p(\chi_1) m^5. \quad (5.8)$$

In the averaging in (5.6), the bare coupling constants $u(h_2)$ and v in (5.3) are renormalized, and they become universal functions of the susceptibilities $\tilde{u}(\chi_1)$ and $\tilde{v}(\chi_1)$, each of which has its own scaling dimensionality. The transformation to the new universal functions

$$g(\chi_1) = \tilde{u}(\chi_1) \chi_1^{(4-d)/2}, \quad p(\chi_1) = \tilde{v}(\chi_1) \chi_1^{d-3}, \quad (5.9)$$

whose scaling dimensionalities are zero, leads to the simplest notation for the equation of state (5.7), since the scaling dimensionalities of the functions m and ψ are also zero.

The equation of state (5.7) describes the entire neighborhood of the TCP (including the λ -line and the first-order transition line). However, one can write

TABLE II. Correspondence between solutions of the helium isotopes and antiferromagnetics.

	³ He- ⁴ He	Antiferromagnetics
ϕ_1	ϕ -superfluid order parameter*	l -antiferromagnetic order parameter
ϕ_2	x - ³ He concentration	M -magnetization
h_1	h -field conjugate to ϕ (nonphysical)	H_{st} -staggered field
h_2	μ -difference of chemical potentials of ³ He and ⁴ He	H_l -internal magnetic field

*Here ϕ is a wave function having two components (amplitude and phase) and is therefore equivalent to a two-component vector. Since x is a scalar quantity, the coupling of the order parameters has the form $\phi^2 x$, which leads to a TCP (see Sec. 3).

it in such a simple form only by neglecting the exponent of the correlation function η , i.e., under the condition

$$\frac{\beta_c}{\gamma_c} = \frac{\beta_t}{\gamma_t}, \quad (5.10)$$

Here the symbols c and t pertain respectively to the exponents at the critical and tricritical points. When η_c is taken into account, the fifth-order term in m is changed. Therefore it exceeds the accuracy of the treatment to preserve this term in the approximation $\eta_c = 0$. However, since $\eta_t = 0$, the equation of state (5.7) gives a correct description of the behavior of the substance in the tricritical region of interest to us, while the smallness of η_c ($\eta_c \approx 0.05$)⁶ assures us that the deviations in the critical region are not too large.

The functions $g(\chi_1)$ and $p(\chi_1)$ are defined by nonlinear differential equations analogous to the equations of the renormalization group. These equations have two FPs, one of which corresponds to critical, and the other to tricritical behavior. In order to find the singularities of the thermodynamic quantities near the critical or tricritical points, one must substitute the values of g and p at the corresponding FPs into the equation of state (5.7), (5.8). The character of the variation of g and p near the FP is usually governed by the nonasymptotic increments to the thermodynamic quantities. It affects the asymptotic singularities of the thermodynamic quantities only at a certain, so-called limiting dimensionality d_c of the space. For a critical point we find $d_c = 4$, and for a tricritical point $d_c = 3$. The FP corresponding to a TCP is determined by the following expressions (for $d = 3$):

$$g_t = 0, \quad p_t \sim \ln^{-1} \chi_1 \rightarrow 0. \quad (5.11)$$

An account taken of the logarithmic dependence of (5.11) leads to the appearance of logarithmic increments

TABLE III. Results of a theoretical study of the tricritical behavior of the heat capacity and the order parameter.

		Self-consistent field (Landau theory)	Fluctuational theory
C_{h_2}	$T > T_t$	const	$\tau^{-1/2} \ln^{-1/2} \frac{1}{\tau}$
	$T < T_t$	$ \tau ^{-1/2}$	$ \tau ^{-1/2} \ln^{1/2} \frac{1}{ \tau }$
ϕ_1	$T < T_t$	$ \tau ^{1/4}$	$ \tau ^{1/4} \ln^{1/4} \frac{1}{ \tau }$

to the pure power-function (scaling) singularities of certain thermodynamic quantities (Table III). Both above and below the TCP, logarithmic corrections to the heat capacity exist, yet they differ: when $\tau > 0$ the singularity is weaker than a pure radical singularity, and stronger when $\tau < 0$. The ratio of the coefficients at the singularities of the heat capacity below and above the TCP is universal and equal to $5\sqrt{5/\pi}$.

Let us list the fundamental differences between the predictions of the fluctuational theory and the results of the classical Landau theory.

1) Singularities of the thermodynamic quantities exist in the disordered phase.

2) Certain singularities are not of pure power-function type.

3) In the T, ϕ_2 plane, all three transition lines (see Fig. 1) approach the TCP with different slopes, in contrast to the conclusions of the Landau theory. This is explained by the fact that the quantity $(\partial \phi_2 / \partial h_2)_T$ [$(\partial x / \partial \mu)_T$ in the case of a ³He-⁴He mixture] has in the disordered phase a singular ($\sim \tau^{-\alpha}$) component just as the heat capacity does.²⁸

We should note that all the differences from the Landau theory arise upon taking systematic account of the logarithmic corrections. Without an account taken of the logarithmic term in (5.11), the TCP is fully described by the classical theory.

b) Experiment

Tricritical points have been found in systems that undergo phase transitions of the most varied types: magnetic,^{38,39} orientational,⁴⁰⁻⁴⁶ liquid-crystalline,⁴⁷⁻⁵⁰ etc. Correspondingly, the distorting factors that lead to deviation of the experimentally determined exponents from the true, asymptotic values also differ. For example, in magnetics, one of the main difficulties is taking the demagnetizing fields correctly into account. For specimens of strictly ellipsoidal shape, the demagnetizing field is homogeneous and can be taken into account. When the shape deviates from ellipsoidal, an inhomogeneous field arises that qualitatively alters the behavior in the immediate vicinity of the transition point. In all likelihood, this circumstance has led to values of the exponents differing from the theoretical ones near the TCP in the antiferromagnetic $\text{CsCoCl}_3 \cdot 2\text{D}_2\text{O}$.⁵¹ Here the magnitude of the inhomogeneous demagnetizing field was not at all small (~ 25 Oe),³⁹ owing to the irregular shape of the specimen.

Another factor that affects the value of the exponents is crossover. If the measurements are not performed in the asymptotic region, the behavior of the thermodynamic quantities is described by certain effective exponents. Thus, the obtained value of the effective exponent for the heat capacity in the orientational transition in NH_4Cl varies smoothly with varying pressure⁴⁰ (Fig. 11). The behavior of the critical order parameter ϕ_1 in ND_4Cl has been studied.⁴¹ It turned out that the value of the exponent β depends considerably on the path of approach to the phase transition point. For example,

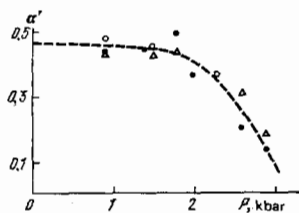


FIG. 11. Pressure-dependence of the tricritical exponent of the heat capacity of NH_4Cl . The differently shaped symbols pertain to different methods of processing the experimental results.

near the critical point $T_c = 280$ K, one finds $2\beta_c = 0.61 \pm 0.04$ for $P = P_c$, and $2\beta_c = 0.50 \pm 0.04$ for $T = T_c$. An analogous discrepancy was found also near the TCP: $2\beta_t = 0.36 \pm 0.01$ for $P = P_t$, and $2\beta_t = 0.28 \pm 0.01$ for $T = T_t$. This is explained by the fact that the path $T = \text{const}$ lies mostly in the transition region, owing to the great steepness of the transition line $P(T)$ (Fig. 12).

We have noted two factors that affect the value of the exponents: distorting factors and crossover. There is another factor that involves the fact that tricritical behavior is not described by pure power-function relationships, and processing of the results of the measurements without taking the logarithmic factors into account can lead to false values of the exponents.

This is manifested especially explicitly in the determination of the exponent β_t , which is small in magnitude. Even with a single choice of T_t , the difference between the pure power-function dependence $\phi_1 \sim |\tau|^{\beta_t}$ and the theoretical relationship $\phi_1 \sim |\tau|^{1/4} |b/\tau|$ in the interval $|\tau| = 10^{-4}$ to 10^{-1} does not exceed 2% for $\beta_t = 0.20$, and 4% for $\beta_t = 0.18$. Hence it is not surprising that the neutron-scattering intensity data in ND_4Cl ⁴¹ (accuracy ~7%) are equally well described by a pure power-function relation and by the theoretical formula.⁴⁰

The study of the tricritical behavior in the ^3He - ^4He mixture and in many-component liquid mixtures is characterized by the fact that the experiment is performed at constant concentration, i. e., at $\phi_2 = \text{const}$, and one must take into account the renormalization of the exponents. The pertinent experimental values are given in Table V and they agree with the theoretical values. When we take the remarks made above into ac-

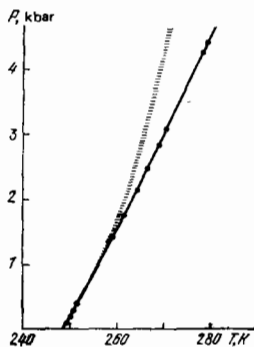


FIG. 12. Phase diagram of ND_4Cl . The region of the transition from critical to tricritical behavior is dotted.

TABLE IV. Tricritical exponents. Comparison with experiment.

System	α_t	α'_t	β_t	γ_t	ν_t	δ_t
DyPO_4	0.50 ± 0.04 ⁷¹	0.46 ± 0.04 ⁷¹	—	—	—	—
KH_2PO_4	—	—	≈ 0.25 ⁵²	1.04 ± 0.02 ⁵³	—	5.1 ± 0.8 ⁵³
NH_4Cl	—	0.45 ± 0.07 ⁴⁰ 0.57 ± 0.07 ⁴²	—	—	—	—
ND_4Cl	—	0.50 ± 0.07 ⁴²	0.18 ± 0.01 ⁴¹	1.05 ± 0.20 ⁴¹	0.52 ± 0.08 ⁴¹	—
NH_4Br	—	0.49 ± 0.01 ⁴⁶	≈ 0.25 ⁴³⁻⁴⁵	—	—	—
Fluctuational theory	0.18 ± 0.06 ⁵⁴ (0.5*)	0.35 ± 0.03 ⁵⁴ 0.5	$0.18 - 0.20$ ⁵⁷ 0.25	1 ⁵⁵ 1	0.5 ⁵⁸ 0.5	5

*The Landau theory has $\alpha_t = 0$.

count, the values of the non-renormalized exponents also agree with the theory (see Table IV).

The form of the phase diagram in the coordinates T and h_2 is determined by the crossover exponent ϕ . All the existing experimental data (see Table VI) agree with the theoretical value $\phi = 2$. Moreover, it has been confirmed experimentally^{60,67} that the transition line $T(h_2)$ has a continuous first derivative and discontinuous second derivative at the TCP. Thus, it was found in Ref. 67 that the transition line in DAG has the following form near the TCP (the signs \pm pertain to $T \gtrless T_t$):

$$h_{2\pm} - h_{2t} = A_{\pm}(T - T_t) + B_{\pm}(T - T_t)^2.$$

Here A_+ and A_- coincide to an accuracy of 5%, while $B_+ \neq B_-$ ($B_+/B_- = 3.9 \pm 1.4$). It is difficult to find the critical exponents in non-zero ordering field h_1 , since the latter is nonphysical in the mixture of helium isotopes, while for antiferromagnetics h_1 is the staggered field H_{st} . Fortunately, owing to the special symmetry in DAG, application of a magnetic field along the $[111]$ axis induces H_{st} ,⁷⁰ and one can find⁶⁹ the form of the critical lines for $H_{st} \neq 0$. The value of the critical exponent κ agrees with the theoretical value (see Table VI). In the ferroelectric KH_2PO_4 , the role of h_1 is played by the electric field E (see Fig. 4), and one can construct the whole phase diagram. However, the existing experimental data do not suffice for finding the exponent κ .

The numerous studies^{7,58,60,61} of the phase diagram of the ^3He - ^4He mixture in the coordinates of T and x all agree with the theory (as an example, Fig. 1 shows the phase diagram taken from Ref. 7). We note that, although all the lines approach T_t linearly, there is a discrepancy (~15–20%) in the slopes found in the different studies. In all probability they involve the gravitational effect.⁶¹

Figure 5 shows the phase diagram of FeCl_2 as obtained by different methods: by neutron diffraction¹³ and from measurements of the magnetic circular di-

TABLE V. Renormalized tricritical exponents.

System	γ_t^*	γ'_t^*	ν_t^*	ν'_t^*
^3He - ^4He	—	—	1 ⁶¹	—
C_6H_6 - $\text{C}_6\text{H}_5\text{OH}$ - $-\text{H}_2\text{O}$ - $(\text{NH}_4)_2\text{SO}_4$	2.20 ± 0.12 ⁶³	1.98 ± 0.11 ⁶³	1.15 ± 0.1 ⁶⁴ 1.06 ± 0.08 ⁶⁵	1.03 ± 0.06 ⁶³
Theory	2	2	1	1

TABLE VI. Exponents characterizing the form of the phase diagram near the TCP.

System	ψ	κ
He ³ -He ⁴	1.95 ± 0.08 **	—
DAG	1.95 ± 0.11 **	0.41 ± 0.03 **
Theory	2	0.4

chromism.¹⁴ In both studies the lines T_λ and T_* approach T_t linearly and have different slopes, as predicted by the theory. However, the forms of the T_* lines differ completely. The exponent β_* is 0.36¹³ and 1.13 ± 0.14.¹⁴ This discrepancy in the value of the exponent may arise from the difference of almost 2% in the position of the TCP. Unfortunately, the reasons for the discrepancy in T_t are not clear. Apparently, the main reason may involve the difficulty of taking the demagnetizing fields into account.

The behavior of the noncritical order parameter ϕ_2 near the TCP has been studied most fully in two systems: dysprosium aluminum garnet and the ³He-⁴He mixture. Figures 13 and 14 show the results of measuring, respectively, the magnetization of DAG along the tricritical isotherm and the susceptibility with respect to the noncritical order parameter for $M = M_t$.⁶⁶ We see that the data agree with the theoretical values of the exponents $\delta_+ = 2, \gamma_u = 1$. Figures 15 and 16 show the analogous results for the mixture of helium isotopes.⁶⁰ In general we should note that all the experimental data confirm the theoretical values of the exponents $\delta_+ = 2, \gamma_u = \gamma_+ = 1$ (Table VII). There is only one exception known to us, which can serve as an example of the need for taking a thorough account of the effect of distorting factors. In Ref. 62 the behavior of $\partial x / \partial \mu$ in a ³He-⁴He mixture was studied by means of light scattering, and the value $\gamma_- = 1.67 \pm 0.1$ was obtained. A little later, the same authors obtained $\gamma_- = 1.00 \pm 0.05$.⁶³ It turned out that the region of asymptotic power-function behavior of the quantity $\partial x / \partial \mu$ in the superfluid phase is very small: $\Delta T = T_t - T \leq 10$ mK. However, a gravitational effect was manifested in Ref. 62 at $\Delta T < 10$ mK. This distorted the singularity and impeded finding the asymptotic value of the exponent γ_- .

The heat capacity at constant ³He concentration $C_{P,x}$ has also been measured in the ³He-⁴He mixture. Upon approaching the tricritical point, along with the decline in the discontinuity in the unmixing-type first-order

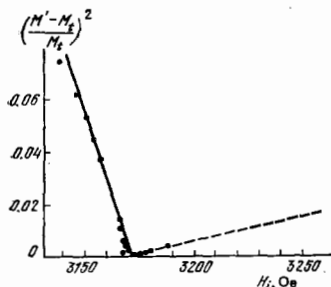


FIG. 13. Magnetization of DAG at $T = T_t$.

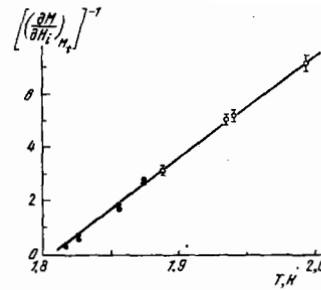


FIG. 14. Susceptibility of DAG at $M = M_t$.

transition, the value of $C_{P,x}$ along the λ -line also declines⁷ (Fig. 17). Along the tricritical "isochore" $x = x_t$, the heat capacity $C_{P,x}$ decreases linearly⁵⁹ (Fig. 18). This agrees with the theoretical value of the exponent $\alpha_u = -1$. An analogous value has been obtained by processing the light-scattering data by using the scaling relationships among the exponents.⁶⁴ Naturally, the value $\alpha_u = -1$ corresponds to $\alpha_t = 1/2$. Direct measurement of the heat capacity at constant chemical potential $C_{P,\mu}$ (i.e., C_{N_2}) is impossible in practice. However, a recalculation from $C_{P,x}$ to $C_{P,\mu}$ has been performed in Ref. 59. The results (Fig. 19) agree with the value $\alpha_t = 1/2$.

6. BICRITICAL AND TETRACRITICAL POINTS

a) Theory

We have shown in Sec. 4 that the existence of two parameters with noncoincident transition points leads to a renormalization of all the constants of the effective Hamiltonian. The character of this renormalization depends on the character of the coupling between the variables ϕ_1 and ϕ_2 . In particular, in systems of the type of ³He-⁴He [the coupling operator has the form of (4.4) with $Q_2 = \phi_2$], the fluctuations of the noncritical mode mainly renormalize the quaternary coupling constant u_1 . Yet the temperature of the transition with respect to the variable ϕ_1 is practically independent of the fluctuations of ϕ_2 . This leads to the appearance of a TCP in the phase diagram.

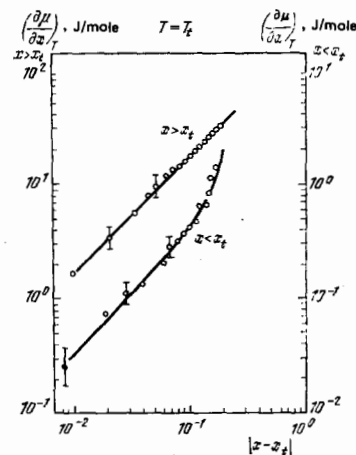


FIG. 15. Concentration-dependence of the susceptibility with respect to the noncritical order parameter $(\partial x / \partial \mu)_T$ in a ³He-⁴He mixture at $T = T_t$.

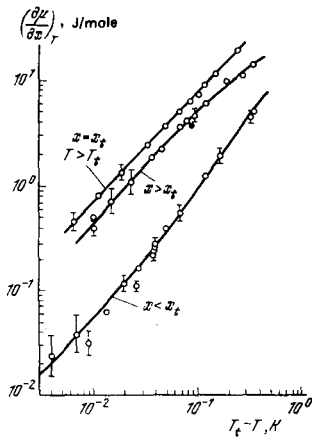


FIG. 16. Temperature-dependence of the susceptibility $(\partial\mu/\partial x)_T$ in a ^3He - ^4He mixture at $x = x_c$ and along the first-order transition lines.

Another type of coupling is realized, e. g., in antiferromagnetics with weak anisotropy,⁷² and also in certain materials that undergo crystallographic transitions (SrTiO_3)⁷³. The crossover term in ϕ_1 and ϕ_2 in these systems has the form

$$\mathcal{H}_{\text{int}} = \lambda \int d^d r \varphi_1^2(r) \varphi_2^2(r). \quad (6.1)$$

With this choice of \mathcal{H}_{int} , one can easily show that the fluctuations of the noncritical variable ϕ_2 ; 1) shift the temperature of the transition with respect to the parameter ϕ_1 ; and 2) renormalize the bare quaternary coupling constant u_1 . Upon substituting $Q_1 = \phi_1^2(r)$ and $Q_2 = \phi_2^2(r)$ into (4.7) and (4.8), we obtain in the weak-coupling limit (the general case has been treated in Ref. 74):

$$\begin{aligned} \Delta T_{c1} &\sim \lambda \int d^d k \langle |\varphi_2(k)|^2 \rangle, \\ \Delta u_1 &\sim \lambda^2 \int d^d r d^d r' \langle \langle \varphi_2^2(r) \varphi_2^2(r') \rangle \rangle. \end{aligned} \quad (6.2)$$

The increment to u_1 is proportional to the correlator of energies of the parameter ϕ_2 , which is equal to the heat capacity of the system described by the Hamiltonian $\mathcal{H}_2\{\phi_2\}$. In the case of separated transition points,

TABLE VII. Exponents of the noncritical order parameter.

System	β_u	β_+	β_-	ν_u	γ_+
^3He - ^4He	1.00 ± 0.05^9	1^9	1^9	1.02 ± 0.03^{63}	1.00 ± 0.02^{63}
DAG	0.97 ± 0.04^{69}	1^{66}	1^{66}	1.01 ± 0.07^{66}	1^{67}
FeCl_2	1.11 ± 0.11^{14}	1.03 ± 0.05^{14}	1.13 ± 0.14^{14}		
fluctuational theory	1	1	1	1	1
Landau theory	1	1	0	0	0
System	γ_-	δ_+	δ_-	α_u	
^3He - ^4He	1.00 ± 0.05^{63}	2.05 ± 0.1^9	2.07 ± 0.1^9	-1^{59}	-0.9 ± 0.2^{44}
DAG	1^{67}	2.12 ± 0.24^{66}	2.14 ± 0.26^{66}		
FeCl_2	1	2	2		
Fluctuational theory	1	2	2	-1	
Landau theory	1	1	2		0

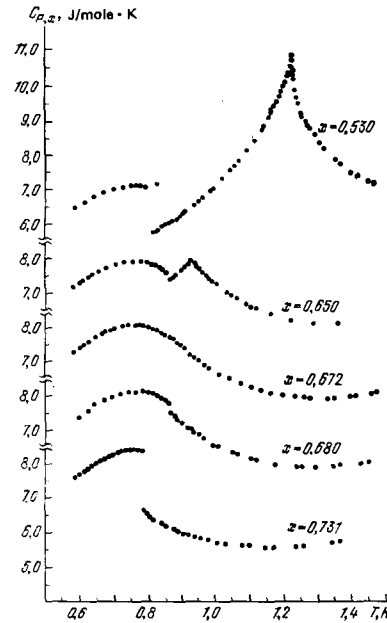


FIG. 17. Heat capacity $C_{P,x}$ of ^3He - ^4He mixtures.

the expressions of (6.2) are analytic functions of their argument $\Delta\tau(T_{c1} - T_{c2})/T_{c2}$, and the effective Hamiltonian proves to be isomorphic with the Hamiltonian $\mathcal{H}_1\{\phi_1\}$.

However, as $\Delta\tau \rightarrow 0$, both the effective quaternary coupling constant and the shift in the transition temperature are nonanalytic functions of the difference $\Delta\tau$ between the critical temperatures. Here the phase transition is nonisomorphic with the "ideal" phase transition.

Let us find the singularities of the thermodynamic quantities in this case. As the main approximation, it is now more convenient to select a model whose Hamiltonian has the form (4.1) with transition temperatures in the variables ϕ_1 and ϕ_2 that differ weakly from one another:

$$T_{c1}(g) = T_0 + a\Delta g, \quad T_{c2}(g) = T_0 - b\Delta g, \quad (6.3)$$

Here Δg is the deviation of the disordering field from a certain value g_0 that is defined by the equations

$$T_{c1}(g_0) = T_{c2}(g_0) \equiv T_0, \quad (6.4)$$

$$a = \frac{An_2}{n}, \quad b = \frac{An_1}{n}, \quad n = n_1 + n_2, \quad A > 0. \quad (6.5)$$

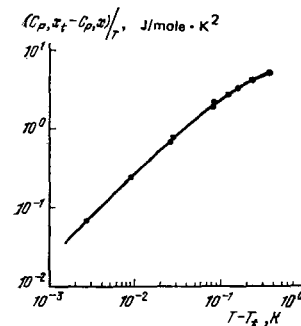


FIG. 18. Temperature-dependence of the heat capacity $C_{P,x}$ of a ^3He - ^4He mixture at $x = x_c$.

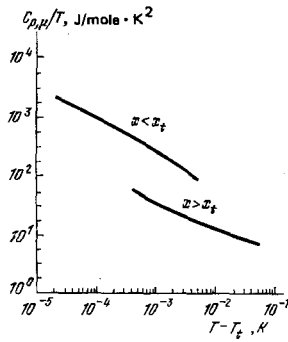


FIG. 19. Temperature-dependence of the heat capacity $C_{p,\mu}$ of ^3He - ^4He mixtures at $\mu = \mu_t$.

Here n_1 and n_2 are the numbers of components of ϕ_1 and ϕ_2 , respectively. \mathcal{H}_{int} has the form (6.1). In the case of an antiferromagnetic (see Fig. 6), the transition temperatures are functions of the magnetic field ($g \sim H^2$). $T_{c1}(H^2)$ is the temperature of the transition to the spin-flop phase, and $T_{c2}(H^2)$ is the temperature of the transition to the antiferromagnetic phase $\Delta g = H^2 - H_0^2 \approx 2H_0\Delta H$.

When $\Delta g > 0$, the order parameter ϕ_1 is critical, while the order parameter ϕ_2 is critical when $\Delta g < 0$. When $T < T_0$ and Δg changes sign, a first-order transition occurs in the system: a phase having $\phi_1 \neq 0$, $\phi_2 = 0$ is supplanted by a phase with $\phi_1 = 0$, $\phi_2 \neq 0$ (a more complicated case of an intermediate phase is also possible).

When $\Delta g = 0$, both order parameters are critical, and a transformation to an effective Hamiltonian that depends only on one of the variables is impossible. In this case the singularities of the thermodynamic quantities can be found by the renormalization-group method within the framework of the ε -expansion. If we assume that the dimensionality d of the space is close to four: $d = 4 - \varepsilon$ (we have singled out the dimensionality $d = 4$ since all the fluctuations in four-dimensional space prove to be Gaussian, while the singularities of the thermodynamic quantities are classical), we can employ perturbation theory in the small parameter ε . Here all the coupling constants are renormalized and also prove to be of the order of ε .

The equations for the complete Green's functions $G_i(k) = \langle |\phi_i(k)|^2 \rangle$ in the first order in ε have the form

$$\begin{aligned} G_1^{-1} &= \tau_1 + (n_1 + 2) \tilde{u}_1 G_1^{-1} \ln G_1^{-1} + \frac{n_2}{2} \tilde{\lambda} G_2^{-1} \ln G_2^{-1} \\ G_2^{-1} &= \tau_2 + (n_2 + 2) \tilde{u}_2 G_2^{-1} \ln G_2^{-1} + \frac{n_1}{2} \tilde{\lambda} G_1^{-1} \ln G_1^{-1}. \end{aligned} \quad (6.6)$$

The exponents if the susceptibilities $\gamma_{1,2}$ (with respect to the first- and second-order parameters) are obtained by substituting the values of the renormalized coupling constants at the FP into Eq. (6.6):

$$\begin{aligned} \gamma_1 &= 1 + (n_1 + 2) \tilde{u}_1 + \frac{n_2}{2} \tilde{\lambda}, \\ \gamma_2 &= 1 + (n_2 + 2) \tilde{u}_2 + \frac{n_1}{2} \tilde{\lambda}. \end{aligned} \quad (6.7)$$

Near the intersection point of the curves $T_{c1}(g)$ and $T_{c2}(g)$, all the thermodynamic quantities have the characteristic "homogeneous" form of (3.21), in particular,

$$G_1^{-1} = \tau^{\nu_1} f_1 \left(\frac{\Delta\tau}{\tau^{1/\varphi}} \right), \quad G_2^{-1} = \tau^{\nu_2} f_2 \left(\frac{\Delta\tau}{\tau^{1/\varphi}} \right). \quad (6.8)$$

Here we have $\tau = (T - T_{tr})/T_0$, and the crossover exponent ϕ in the first order in ε is equal to

$$\varphi = \frac{1}{\phi} = 1 - \frac{1}{2} \tilde{\lambda} (n_1 + n_2). \quad (6.9)$$

The transition temperature T_{tr} and the functions f_1 and f_2 are determined by Eq. (6.6). In particular, $T_{tr} - T_0 \sim \Delta\tau^\phi$.

A bicritical or a tetracritical point will be realized, depending on the number of components of the order parameters.³⁴ Thus, when $n = n_1 + n_2 < 4$, a bicritical point will exist. In this case we have

$$\begin{aligned} \tilde{u}_1 = \tilde{u}_2 &= \frac{\tilde{\lambda}}{2} = \frac{\varepsilon}{2(n+8)}, \\ \gamma_1 = \gamma_2 &= 1 + \frac{\varepsilon(n+2)}{2(n+8)}, \quad \varphi = 1 + \frac{\varepsilon n}{2(n+8)}. \end{aligned} \quad (6.10)$$

Since $\phi > 1$, in contrast to the Landau theory, the critical lines are tangent to each other at the bicritical point.

When $n > 4$, the intersection point will be tetracritical, i. e., an intermediate phase arises. However, we must distinguish two cases:

- 1) Here we have $(n-8)(n+16) > (n_1 - n_2)^2$ —an "uncoupled" tetracritical point;
- 2) Here we have $4 < n < 8 + (n_1 - n_2)^2/(n+16)$.

In the former case we find

$$\begin{aligned} \tilde{\lambda} &= 0, \quad \tilde{u}_1 = \frac{\varepsilon}{2(n_1+8)}, \quad \tilde{u}_2 = \frac{\varepsilon}{2(n_2+8)}, \\ \gamma_1 &= 1 + \frac{\varepsilon(n_1+2)}{2(n_1+8)}, \quad \gamma_2 = 1 + \frac{\varepsilon(n_2+2)}{2(n_2+8)}, \quad \varphi = 1. \end{aligned} \quad (6.11)$$

That is, the system at an "uncoupled" tetracritical point behaves like two noninteracting subsystems, and the transition lines intersect at this point at an angle ($\phi = 1$).

In the latter case we have $\tilde{\lambda} > 0$, $\phi > 1$, and the transition lines are tangent to each other at the tetracritical point. The exponents also turn out to be different.³⁴

b) Experiment

The experimental study of bicritical points is at present in its very initial stage. The relatively small number of studies is mainly restricted to measuring the form of the phase-transition lines near a bicritical point, i. e., determining the single critical exponent ϕ [see (6.9)]. All the systems that have been studied are anisotropic antiferromagnetics with an "easy axis" type of anisotropy (the phase diagram is given in Fig. 6).

Along with the ordinary experimental difficulties of studying phase transitions in solids (inhomogeneous samples, long relaxation times, etc.), another difficulty exists here that is precisely specific for the given experiments: the need for exact coincidence of the direction of the external magnetic field H and the axis of easiest magnetization (AEM). When the direction deviates from the AEM, the phase diagram is greatly complicated.²⁶ In particular, the transition from the antiferromagnetic phase to the spin-flop phase ceases

to be a first-order transition.⁷⁵ In all likelihood, it makes sense to perform measurements at various angles between the field H and the AEM and to extrapolate the results of the measurements to zero angle.⁷⁶ In Ref. 76, the phase transition lines of $GdAlO_3$ were determined from the maxima of the magnetic susceptibility, and the crossover exponent ϕ was found from the expressions⁷⁷

$$H^2 - H_b^2 - p t_{||, \perp} = \omega_{||, \perp} [t_{||, \perp} + q (H^2 - H_b^2)]^\phi. \quad (6.12)$$

Here we have

$$t_{||, \perp} = \frac{T_c^{||, \perp} - T_b}{T_b}, \quad p = T_b \left(\frac{\partial H_{SF}}{\partial T} \right)_b, \quad q = -\frac{n+2}{3n} \frac{1}{T_b} \left(\frac{\partial T_c}{\partial H^2} \right)_{H=0}.$$

Here the $T_c^{||, \perp}$ are respectively the critical temperatures of the paramagnetic-antiferromagnetic and the paramagnetic-spin flop phase transitions, and $\omega_{||}$ and ω_{\perp} are nonuniversal coefficients whose ratio is universal and depends only on the total number of components of the order parameters $n = n_1 + n_2$. The subscripts b and SF pertain to the bicritical point and to the antiferromagnetic-spin-flop phase transition line. It was assumed in processing the data⁷⁶ that $\omega_{||} = \omega_{\perp}$. That is, a total dimensionality of the order parameters $n = 2$ was directly imposed (Table VIII). The exponent ϕ proved to be 1.25 ± 0.07 , which agrees reasonably with the theoretical value.⁷⁸ In Ref. 76 an attempt was made also to determine two other critical exponents: γ_u —the exponent of the noncritical susceptibility, and β_u —the jump in the total magnetization for $T < T_b$. The experimental value of the exponent β_u agrees well with the theoretical prediction. Yet the values that were obtained of the exponent γ_u proved to be somewhat lower than the theoretical estimate. In all likelihood this involves the effect of crossover.

Measurements of the form of the phase-transition lines in MnF_2 ⁷⁹ yield a result intermediate between $n = 2$ and $n = 3$. The value of the exponent ϕ agrees well with the theoretical value for $n = 2$, whereas the universal ratio $\omega_{\perp}/\omega_{||}$ lies between the theoretical values for $n = 2$ and $n = 3$ (see Table VIII).

The experimental data near tetracritical points are qualitative in nature, and they consist mainly of the proof of existence of an intermediate phase. The accuracy of the experiments evidently does not suffice for determining the form of the phase-transition lines (i. e., the exponent ϕ). Among the other critical exponents, only the exponent β , which describes the behavior of the order parameter has been determined,¹⁹ but this was done along the critical lines rather than at the tetracritical point itself.

TABLE VIII. Results of experimental study of bicritical points.

Субстрат	ϕ	$\omega_{\perp}/\omega_{ }$	β_u	γ_u
$GdAlO_3$	1.25 ± 0.07 ⁷⁶	1 ⁷⁶	0.92 ± 0.03 ⁷⁶	0.15 ⁷⁶
MnF_2	1.279 ± 0.031 ⁷⁹	1.56 ± 0.35 ⁷⁹	—	—
$NiCl_2 \cdot 6H_2O$	1.24 ± 0.04 ⁸⁰	1 ⁸⁰	—	—
$CsMnBr_3 \cdot 2D_2O$	1.20 ± 0.03 ⁸¹	—	—	—
$RbMnF_3$	1.278 ± 0.02 ⁸²	—	—	—
Theory	$n = 2$ $n = 3$	1 2.51	0.85 ± 0.04 0.85 ± 0.04	0.33 ± 0.07 0.40 ± 0.07

7. DISTORTED POLYCRITICAL BEHAVIOR

a) Tricritical behavior in the isotropic liquid-nematic liquid crystal phase transition

Tricritical points have been found⁴⁷⁻⁵⁰ in liquid-crystalline materials in the transition from a nematic or cholesteric phase to a smectic phase (Fig. 20).

There have been attempts⁸³ to explain the appearance of a tricritical point within the framework of the Landau theory by the coupling of the smectic order parameter ψ (two-component vector) with the nematic order parameter $Q = \langle \frac{1}{2} (3 \cos^2 \theta - 1) \rangle$, where θ is the angle between the orientation of the axis of the molecule and the preferred direction.^{84, 4)} The term in the thermodynamic potential that takes into account the coupling of the order parameters has the form $\lambda |\psi|^2 Q$, as in the 3He - 4He mixture (see Sec. 3a).

In contrast to the nematic-smectic transition, the isotropic liquid-nematic liquid crystal phase transition ($I \rightleftharpoons N$) is everywhere a first-order transition and is traditionally described by the Landau theory:

$$\Phi - \Phi_0 = \frac{1}{2} a \tau Q^2 - \frac{1}{3} B Q^3 + \frac{1}{4} C Q^4. \quad (7.1)$$

The smallness of the constant $B \approx 0.06$ ⁸⁵ has the result that the jump in entropy is small, and the transition is close to a second-order transition ($B = 0$).

The constant C also has proved to be unexpectedly small. This has posed the question of the closeness of the $I \rightleftharpoons N$ transition to a tricritical point ($C = 0$).⁸⁵ The solid line in Fig. 21 shows the heat-capacity curve that arises from the Landau theory with account taken of fluctuational corrections in the Ornstein-Zernike approximation.⁵⁴ The experimental curve fundamentally differs from the theoretical curve in the absence of a jump in the regular components and in the large magnitude of the anomaly in the isotropic phase. Upon comparing the temperature dependences near the $I \rightleftharpoons N$ transition and near the TCP in NH_4Br , their striking similarity catches one's eye⁸⁶ (Fig. 22). At the same time, an attempt to describe the temperature-dependence of the heat capacity with a law having a fixed exponent $\alpha = 0.5$ (as at a tricritical point) yields an inadequate model. The model becomes adequate upon vary-

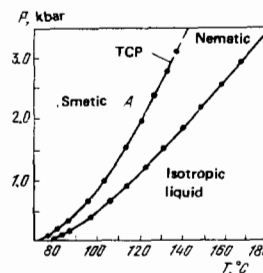


FIG. 20. Phase diagram of the liquid crystal CBNA.⁴⁷

⁴⁾Strictly speaking, the nematic order parameter is a second-order tensor, and the presented treatment is a substantial simplification.

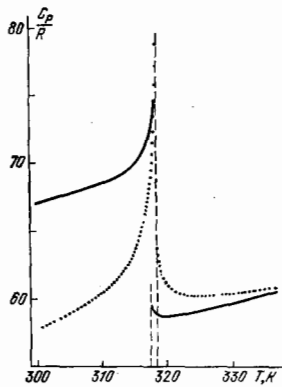


FIG. 21. Experimental values of the heat capacity of MBBA and the results of the Landau-de Gennes theory with allowance for the fluctuational corrections in the Ornstein-Zernike approximation (solid line).

ing the critical exponents, though the values of α and d (see Table IV) do not fit into any of the theoretical schemes. We can explain this result, first, by crossover from tricritical to critical behavior (see Sec. 3), and second, by the need for taking account of the logarithmic factors. Therefore the data on the heat capacity of methoxybenzylidenebutylaniline (MBBA) and butylmethoxyazoxybenzene (BMOAB) have been analyzed⁵⁴ for agreement of the crossover model of (3.21) with the function $f(z)$, which was chosen in the simplest form

$$f(z) = K_0 [1 + K_2 z^{\alpha_1 - \alpha_0}]^{-1}. \quad (7.2)$$

That is, the experimental data were processed by the formula

$$C_P = |K_1| \tau^{0.5} + K_2 |\tau|^{-1} + (C_P)_{\text{reg}}. \quad (7.3)$$

The best agreement is attained for $\alpha = 0.1$. Here the model of (7.3) adequately describes the data in both phases. In all cases the constant K_2 is smaller by a factor of 15 to 30 than K_1 . Therefore the second term in (7.3) is comparable with the first only when $|\tau| < 10^{-3}$. Consequently tricritical behavior governs throughout the experimental temperature range, both in the nematic and in the isotropic phase. As regards

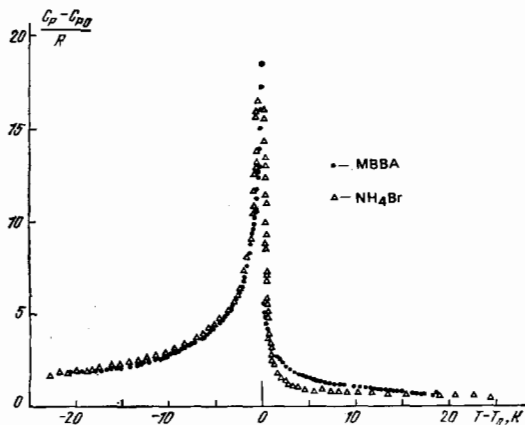


FIG. 22. Comparison of the temperature-dependences of the heat capacity of MBBA and NH_4Br .

the logarithmic factors, one can neglect them compared to the crossover background at the existing accuracy of calorimetric experimentation.

Essentially the short-range intermolecular forces are responsible for the $I \rightleftharpoons N$ transition. Therefore it was difficult from the very outset to expect agreement of the Landau theory with experiment. The "classical" behavior of the susceptibility $\sim (T - T_c)^{-1}$ and of the correlation radius $r_c \sim (T - T_c)^{-1/2}$, which followed from rather accurate experiments,⁵⁶ was even more surprising. The idea expressed above that the $I \rightleftharpoons N$ transition is close to tricritical enables one to explain without contradiction the entire set of experimental data: the "classical" behavior of the exponents γ and ν (see Table IV) and the value of the exponent $\beta \approx 1/4$, which was first noted by Keyes.^{87 5)}

The proposed closeness of the $I \rightleftharpoons N$ transition to a TCP poses many problems. Although one can obtain tricritical behavior in the region $\tau \gg B^{4.3}$, $\tau \gg C^{2.85}$ within the framework of the Landau theory, the causes of this behavior are not evident. We can suppose that the nematic order parameter couples with the other degrees of freedom (e.g., the intramolecular ones).⁶⁾ It is not ruled out that this coupling causes the coefficients B and C simultaneously to be small both by allowing the closeness of the transition to a second-order transition and by giving rise to the features of tricritical behavior.

An explanation of the experimental results has been proposed⁸⁹ that is based on the idea that the $I \rightleftharpoons N$ transition is close to a special second-order transition whose possibility in principle has been demonstrated in Refs. 88 and 89, rather than to a TCP. This transition must be characterized by a special set of critical exponents. However, the calculated values of the critical exponents differ substantially from the experimental values. It seems highly unlikely to us that a more accurate calculation of the exponents will lead, as proposed in Ref. 89, to agreement with experiment.

b) The liquid-gas coexistence curve near the critical point of mercury

Features of the equation of state have been found experimentally near the critical point of mercury that sharply distinguish it from nonconductive liquids.⁹⁰ Mercury is characterized by failure of the law of corresponding states and by an unusually strong deviation of the liquid-gas coexistence curve from linearity (Fig. 23). One can explain these features⁹¹ by the coupling of two order parameters: the density ρ , which characterizes the liquid-gas transition, and the concentration n

⁵⁾The experimental data indicate rather the value $\beta = 0.18 - 0.20$.⁵⁷ As we have noted above, one can easily raise this value to 0.25 by taking into account the logarithmic corrections. Moreover, one should treat the given values of β with caution, since the temperature-dependence of the order parameter was determined in a narrow temperature range (smaller than an order of magnitude with respect to τ).

⁶⁾Apparently this hypothesis has recently been confirmed experimentally.⁹⁹

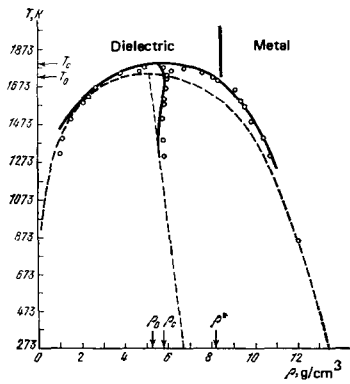


FIG. 23. Phase diagram of mercury near the critical point.

of electrons in the conduction band, which characterizes the metal-dielectric transition. This coupling has been treated in the approximation of the self-consistent field.⁹¹ The thermodynamic potential of the interaction Φ_{int} was chosen in the simplest form allowed by symmetry considerations, and which was derived from model considerations in Ref. 92:

$$\Phi_{\text{int}} = -B\rho n. \quad (7.4)$$

Figure 23 shows the experimental points⁹⁰ and the theoretical coexistence curve⁹¹ (solid line) of mercury. The heavy line at $\rho = \rho^*$ shows the metal-dielectric transition. The dotted line is an extrapolation from the low-temperature region without taking into account the coupling of the order parameters (see Ref. 90). Taking the coupling into account substantially alters the coexistence curve. The critical point of the liquid-gas transition is "pulled" toward the metal-dielectric transition line. However, in this case a TCP is not realized, owing to the following circumstance. It is common to speak provisionally of the metal-dielectric "transition" in the transcritical region, having in mind the line at which the electrical conductivity varies almost jumpwise. We should also note that, even if the metal-dielectric transition were a real phase transition, nevertheless it could not be a second-order transition. This is prevented by the existence in the thermodynamic potential of the system of a coupling term in (7.4) that is linear in n .

c) The intersection point of the oriented-phase-transition lines in ammonium bromide

If the β - δ and β - γ transitions in NH_4Br (see Fig. 3 and Sec. 2) were second-order, then the triple point O would be a bicritical point, and the heat of the δ - γ transition would approach zero at the point O . However, near the point O the β - δ and β - γ transitions are first-order, and we can view their intersection point as "bicritical" only to the extent that they are close to second-order, i. e., to the extent that the ratio $\lambda/RT \leq 0.1$ is small. Nevertheless this closeness suffices, as shown by experiment,⁹³ to cause a sharp decline in the heat of the δ - γ transition. In NH_4Br there are two critical order parameters: one is characterized by parallel, and the other by antiparallel orientation of the ammonium tetrahedra in the crystal structure. Their

coupling should lead to a bicritical point. However, the coupling of the critical order parameters with the non-critical degrees of freedom causes a breakdown into a first-order transition (a TCP on the β - δ line). Unfortunately, it is not yet clear which degrees of freedom are responsible for this breakdown.

8. CONCLUSION

The main result of the study of systems with coupled order parameters is the substantially expanded possibility of universal description of real phase transitions in the language of more or less simple models. Previously, real systems were treated from the standpoint of distortion of a model having a single order parameter (e. g., the Ising model with an impurity, taking account of asymmetry in the model of a lattice gas, etc.). Now already a set of models having several coupled order parameters can play the role of "ideal systems". Thus one can include in the physics of critical phenomena topics that, owing to their complexity, were heretofore the object of special fields (physics of magnetic phenomena, physics of liquid crystals, physical chemistry of solutions, etc.). And while currently a physically clear and theoretically rigorous description of tricritical behavior exists only for the ^3He - ^4He mixture in a model of coupled order parameters (wave function and concentration), we are assured of the generality of this approach to polycritical phenomena.

Numerous objects whose behavior apparently can also be explained by the coupling of order parameters have remained outside the scope of our review. First of all, these include the recently discovered⁹⁴ transitions to the superfluid state in ^3He , which merit a special review. We have also not treated the so-called Lifshits critical points, which separate the lines of commensurate and incommensurate phase transitions,^{95,96} and phase transitions in nonintrinsic ferroelectrics, to which a special review has been devoted.⁹⁷

Recently, great interest has been aroused in dissipative structures.⁹⁸ The study of the coupling of several order parameters in these systems should lead to analogs of the polycritical points for nonequilibrium processes (a promising topic in this regard is convective instability in mixtures).

LIST OF THE MAIN ABBREVIATIONS AND SYMBOLS

TCP	-tricritical point,
FP	-fixed point of the transformations of the re-normalization group,
AEM	-axis of easiest magnetization,
DAG	-dysprosium aluminum garnet,
NLC	-nematic liquid crystal,
MBBA	-methoxybenzylidenebutylaniline,
BMOAB	-butylmethoxyazoxybenzene,
T	-temperature,
T_c	-critical temperature (the symbol c refers to critical parameters),
T_0	-temperature of the polycritical point,
T_t	-temperature of the tricritical point (the symbol t pertains to tricritical parameters),

T_b	-temperature of the bicritical point (the symbol b pertains to bicritical parameters),							
P	-pressure,							
μ	-chemical potential,							
x	-concentration,							
E	-electric field,							
H	-magnetic field,							
H_i	-internal magnetic field,							
H_{st}	-staggered magnetic field,							
M	-magnetization,							
M_0	-saturation magnetization,							
d	-dimensionality of the space,							
\mathcal{H}	-Hamiltonian,							
Φ	-thermodynamic potential,							
τ	$-(T - T_c)/T_c$,							
ϕ	-order parameter,							
h	-field thermodynamically conjugate to ϕ ,							
G	-correlation function of the fluctuations,							
r_c	-correlation radius of the fluctuations,							
r_0	-direct correlation radius,							
χ	-susceptibility,							
C	-heat capacity,							
n_i	-number of components of the orderparameter							
n	$-\sum_i n_i$,							
k	-wave vector,							
ϕ	-crossover exponent,							
κ	-exponent that determines the form of the critical lines in nonzero ordering field near a TCP,							
α	<table> <tr> <td rowspan="7">-critical exponents of the</td> <td>heat capacity,</td> </tr> <tr> <td>order parameter,</td> </tr> <tr> <td>susceptibility,</td> </tr> <tr> <td>critical isotherm,</td> </tr> <tr> <td>correlation radius,</td> </tr> <tr> <td>correlation function.</td> </tr> </table>	-critical exponents of the	heat capacity,	order parameter,	susceptibility,	critical isotherm,	correlation radius,	correlation function.
-critical exponents of the			heat capacity,					
			order parameter,					
			susceptibility,					
			critical isotherm,					
			correlation radius,					
			correlation function.					
	β							
γ								
δ								
ν								
η								

The primed exponents (α' , etc.) pertain to the ordered phase, renormalization exponents are denoted in addition by an asterisk (α^* , α'^* , etc.), and exponents for quantities involving a noncritical order parameter are marked by the subscripts u (γ_u , etc.).

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