

System Exhibiting a Critical Point of Order Four: Ising Planes with Variable Interplanar Interactions*

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The first phase diagram explicitly exhibiting intersecting lines of tricritical points is presented. Their point of intersection is a critical point of order 4. The system is a simple assembly of ferromagnetic Ising planes coupled with an arbitrary interplanar interaction, with Hamiltonian

$$\mathcal{H} = -J \left(\sum_{\langle ij \rangle} s_i s_j + \mathcal{R} \sum_{(ij)} s_i s_j \right) - \mu H \sum_i s_i - \mu H' (-1)^\eta \sum_i s_i,$$

where $J > 0$, $s_i = \pm 1$, and μ is the magnetic moment per spin. The first sum is over nearest-neighbor (nn) spins in an x - y plane, the second sum over nn spins coupled along the z direction, H is the magnitude of a uniform magnetic field, and H' is a staggered magnetic field which acts oppositely on adjacent planes of constant z ($\eta = 0$ on even planes, $+1$ on odd planes). The Hamiltonian is invariant under $\mathcal{R} \rightarrow -\mathcal{R}$, $H \rightarrow H'$, and $s_i \rightarrow (-1)^\eta s_i$, so that the Gibbs potential is also invariant, $G(T, H, H', \mathcal{R}) = G(T, H', H, -\mathcal{R})$. Using this symmetry, we make a scaling hypothesis about the special point $\tau \equiv T - T_c (\mathcal{R} = 0) = H = H' = \mathcal{R} = 0$, namely, that the Gibbs potential obeys the functional equation $G(\lambda^a \tau, \lambda^{a_H} H, \lambda^{a_{H'}} H', \lambda^{a_{\mathcal{R}}} \mathcal{R}) = \lambda G(\tau, H, H', \mathcal{R})$; the four scaling powers are found to be $a_\tau = \frac{1}{2}$, $a_H = a_{H'} = \frac{15}{16}$, $a_{\mathcal{R}} = \frac{7}{8}$.

I. CONCEPT OF ORDER AND MODEL HAMILTONIAN

In 1970 Griffiths proposed the concept of a tricritical point (TCP)—the intersection of three critical lines.¹ It has come to be increasingly appreciated that this point has special physical properties and occurs in several realizable systems. Tricritical points—and tetracritical points^{2,3} (defined as the intersection of four critical lines)—have similar physical properties; both are examples of “critical points of order 3” (where an ordinary critical point is a critical point of order 2). In this work we present the first example of a system whose phase diagram exhibits intersecting lines of *tricritical* points. We call this point of intersection a critical point of order 4.

Our example is an Ising model on a simple cubic lattice with a fixed ferromagnetic interaction J in the xy planes, and a variable (ferro- or antiferromagnetic) interaction $\mathcal{R}J$ between the planes. The Hamiltonian is

$$\mathcal{H} = -J \left(\sum_{\langle ij \rangle} s_i s_j + \mathcal{R} \sum_{(ij)} s_i s_j \right) - \mu H \sum_i s_i - \mu H' (-1)^\eta \sum_i s_i. \quad (1)$$

Here $J > 0$, $s_i = \pm 1$, and μ is the magnetic moment per spin. The first sum is over nearest-neighbor (nn) spins in an x - y plane, the second sum is over nn spins coupled along the z direction, H is a uniform magnetic field, and H' is a staggered mag-

netic field which acts oppositely on adjacent planes of constant z ($\eta = 0$ on even planes, 1 on odd planes). For $\mathcal{R} > 0$, one has a three-dimensional ($d = 3$) Ising ferromagnet, for $\mathcal{R} < 0$ a model “metamagnet”⁴ (ferromagnetic planes coupled antiferromagnetically), while for $\mathcal{R} = 0$ the lattice reduces to a stack of uncoupled two-dimensional ($d = 2$) square lattices.

The notation proposed by Griffiths and Wheeler^{5a} has been generalized to facilitate treatment of more complex phase diagrams. Their notation is CXS for a coexistence hypersurface, and CRS for a critical hypersurface. For subspaces of the field space where different phases coexist, we use the notation ${}^p X_d$, where p is the number of phases and d is the dimensionality of the space.^{5b} For example, a line of points (such as the vapor pressure curve of a simple fluid) where two phases coexist is denoted by ${}^2 X_1$. For spaces of critical points we introduce the concept of order of a critical point. Ordinary critical points are defined to be of order 2, and points where lines of points of order θ intersect are defined to be of order $(\theta + 1)$ ⁶. The notation used for critical spaces is ${}^\theta R_d$, where θ refers to the order and d to the dimension of CRS.

In a metamagnet the lines of ordinary critical points ${}^2 R_1$ intersect at a tricritical point which is of order 3, a ${}^3 R_0$. For the Hamiltonian (1), such ${}^3 R_0$ occur in the field space spanned by T , H , and H' for any fixed $\mathcal{R} \neq 0$ (cf. Fig. 1). In the full four-dimensional field space obtained when \mathcal{R} is allowed to vary, each TCP becomes a ${}^3 R_1$, a line of tri-

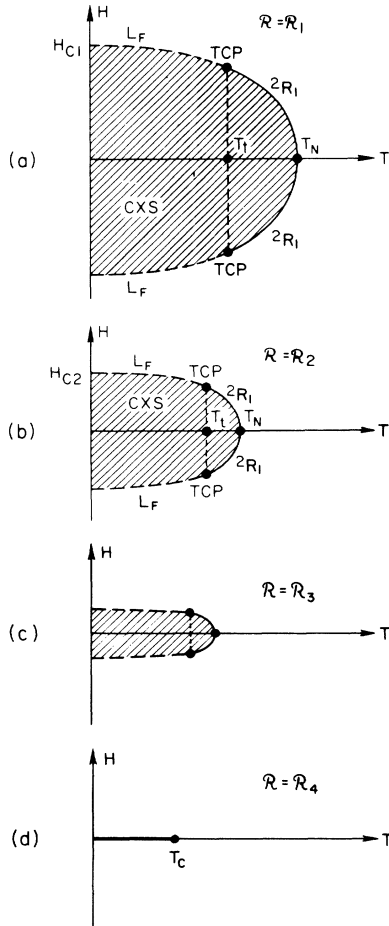


FIG. 1. (a) Section of the four-dimensional field space (T, H, H', \mathcal{R}) for $H' = 0$ and $\mathcal{R} = \mathcal{R}_1 < 0$. L_F is a first-order phase transition line where three phases coexist (3X_1). (b) Same as (a), except that $\mathcal{R} = \mathcal{R}_2 < 0$, with $|\mathcal{R}_2| < |\mathcal{R}_1|$. Note that H_c , T_t , and T_N have all decreased in magnitude. (c) Phase diagram for $\mathcal{R} = \mathcal{R}_3 < 0$, $|\mathcal{R}_3| < |\mathcal{R}_2|$. Here $|\mathcal{R}_3|$ is nearly zero. (d) Phase diagram for $\mathcal{R} = \mathcal{R}_4 = 0$, the two-dimensional limit.

critical points; these intersect and terminate at a 4R_0 where four phases become critical simultaneously.

II. DISCUSSION OF PHASE DIAGRAM

To justify these remarks, we begin by noting that the Hamiltonian (1) has an important symmetry under the operation \mathcal{S} defined as follows: $\mathcal{R} \rightarrow -\mathcal{R}$, $H \rightarrow H'$, $H' \rightarrow H$, and $s_i \rightarrow (-1)^n s_i$. This operation reverses the sign of the interplanar interaction, exchanges direct and staggered external fields, and exchanges direct and staggered magnetizations. Since \mathcal{H} [of Eq. (1)] is invariant under \mathcal{S} , the Gibbs potential is also invariant,

$$G(T, H, H', \mathcal{R}) = G(T, H', H, -\mathcal{R}). \quad (2)$$

The phase diagram for (1) will necessarily re-

flect this symmetry. In particular, the pair of TCPs in the $H-T$ ($H' = 0$) plane [Fig. 1(a)] for $\mathcal{R} = \mathcal{R}_1 < 0$ maps into a pair of TCP's in the $H'-T$ ($H = 0$) plane for $\mathcal{R} = -\mathcal{R}_1 > 0$. What happens as $|\mathcal{R}|$ is decreased from $|\mathcal{R}_1|$ is depicted in Fig. 1(b). The TCP in Fig. 1(a) will approach the Néel point T_N . Also, T_N , the tricritical temperature T_t , and the $T = 0$ critical field $H_c(T = 0)$ will all decrease in magnitude. Figure 1(c) depicts a still smaller value of $|\mathcal{R}|$. Finally, $\mathcal{R} = 0$ is depicted in Fig. 1(d). Here we reduce to the two-dimensional Ising ferromagnet. There is no longer a phase transition in finite field, and the entire phase diagram collapses into a first-order line along the T axis terminating at the Curie point $T_c(0)$, which is seen to be the limit of the sequence of tricritical points for decreasing $|\mathcal{R}|$.

Similar statements hold for $\mathcal{R} > 0$ with the roles of H and H' reversed. We shall focus on the $\mathcal{R} < 0$ case in the following discussion.

The $T = 0$ critical field is trivially given by $\mu H_c(0) = 2|\mathcal{R}|J$. That the Néel temperature decreases as \mathcal{R} decreases is well known. For example, simple molecular-field theory gives

$$k_B T_N(|\mathcal{R}|) = 4J + 2|\mathcal{R}|J. \quad (3)$$

More accurate series-expansions work⁷ on crossover between three- and two-dimensional Ising ferromagnetic lattices shows that a scaling-law prediction of the form

$$T_c(\mathcal{R}) - T_c(0) \sim \mathcal{R}^{4/7} \quad (4)$$

is obeyed for $\mathcal{R} > 0$. The symmetry of the Hamiltonian then implies that $T_c(\mathcal{R}) = T_N(-\mathcal{R})$.

Mean-field theory for two-sublattice Ising antiferromagnets⁸ gives a tricritical point for the model of Eq. (1) for all $|\mathcal{R}| < \frac{4J}{3}$, with the following relation for the ratio of T_t/T_N :

$$T_t/T_N = 1 - \frac{1}{6}|\mathcal{R}| \quad (5)$$

Thus $T_t/T_N \rightarrow 1$ as $|\mathcal{R}| \rightarrow 0$, the two-dimensional ferromagnetic limit.

When Eq. (5) is coupled with Eq. (3) above, we see that

$$k_B T_t = 2J \left(2 + \frac{2}{3}|\mathcal{R}| - \frac{1}{6}|\mathcal{R}|^2 \right). \quad (6)$$

Thus mean-field theory gives T_t as a monotonically increasing function of $|\mathcal{R}|$ in the region of validity of Eq. (3). We shall be concerned only with behavior near $\mathcal{R} = 0$, i. e., for $|\mathcal{R}| \ll 1$. In this region, therefore, mean-field theory supports the behavior we have sketched in Fig. 1.

That T_t behaves in this way may also be obtained from the following physical consideration: The tricritical temperature occurs when the phase transition on a sublattice, produced by increasing the magnetic field H at constant temperature T_0 , changes from first order (for $T_0 < T_t$) to second

order (for $T_0 > T_t$). The effect of increasing the coupling between the plane of spins for which the magnetization changes sign at the transition, and the adjacent planes, on which the magnetization remains of the same sign, will be to increase the effective coupling between adjacent spins in the planes which change sign. Thus one would expect it to increase the temperature T_t at which the sublattice transition changed from first order to second order.

In accordance with all the above considerations,

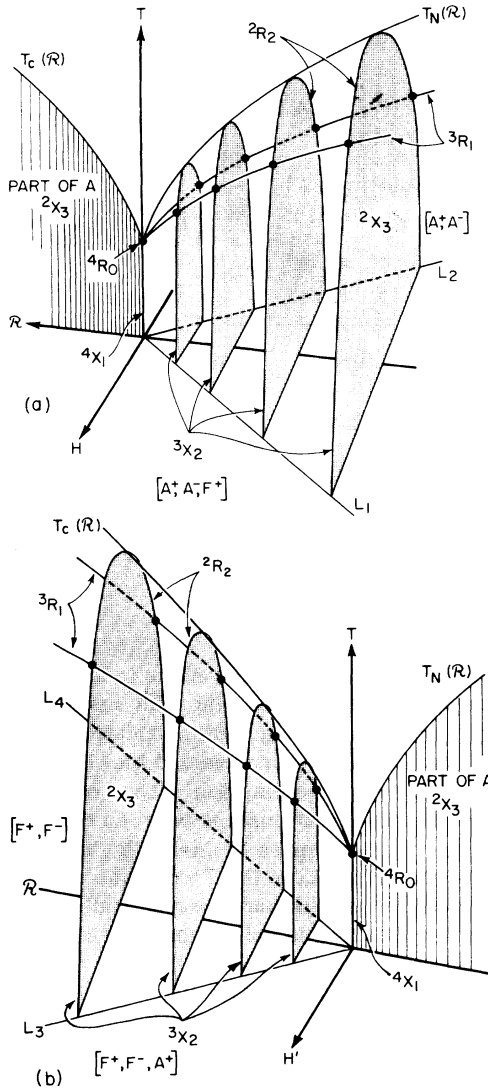


FIG. 2. (a) Section of the (T, H, H', R) field space with $H' = 0$. The reader should note how each 0R_d and 2X_d of Fig. 1 becomes a ${}^0R_{d+1}$ or ${}^2X_{d+1}$: thus the point 3R_0 becomes a line 3R_1 , separating a "snow-covered" surface [${}^2R_1 - {}^2R_2$] and a "tree-covered" surface [$L_F - {}^3X_2$]. (b) Section of the (T, H, H', R) field space with $H = 0$. Now the "mountain" occurs for positive R .

we have represented the $H' = 0$ section of the complete $T-H-H'-R$ space phase diagram in Fig. 2(a), and the $H = 0$ section in Fig. 2(b). Figure 2(a) was obtained by combining Figs. 1(a)-1(d) for different values of R . Figure 2(b) was obtained from Fig. 2(a) by using the symmetry embodied in Eq. (2), and is a mirror image of Fig. 2(a) with H and H' reversed. The essential features of Fig. 2(a) may perhaps be most easily visualized as a coexistence volume ("mountain") capped on top by a surface of critical points ("snow"). The snowcap stops at "snow lines" which are lines of tricritical points. The mountain is a 2X_3 in which A^+ and A^- , the two antiferromagnetic phases corresponding to opposite values of the staggered field, coexist. At $T = 0$ the 2X_3 is bound by the $T = 0$ plane and the lines L_1 and L_2 . On L_1 the three coexisting phases are A^+ , A^- , and a ferromagnetic phase pointing parallel to a positive H , F^+ , while on L_2 , A^+ , A^- , and F^- coexist. For increasing T , the 2X_3 (the mountain) is bounded on either side by 3X_2 —the continuations to finite T of L_1 and L_2 . The 2X_3 is bounded from above by a 2R_2 (the snow cap). The two 3X_2 meet the 2R_2 at lines of TCP's—indicated as 3R_1 . At $R = 0$, all four phases (A^+ , A^- , F^+ , F^-) are in equilibrium from $T = 0$ to $T = T_c(R = 0)$ on the T axis; this line is a 4X_1 . Note that the line of Néel points $T_N(R)$ lies at the crest of the snow cap in the $H = 0$ plane, but is of no particular significance with respect to the rest of the critical 2R_2 surface.^{5a, 5c}

For $R > 0$ in Fig. 2(a), the phase diagram is very simple. There is a critical line $T_c(R)$ bounding a coexistence plane at $H = 0$. However, if one considers the $H = 0$ section of the full $T-H-H'-R$ phase diagram as in Fig. 2(b), the mountain occurs for $R > 0$, and the simple line of critical points for $R < 0$.

A mental superposition of the two mountains in the four-dimensional $T-H-H'-R$ space reveals four different 3R_1 meeting at the point $T_c(R = 0)$, $H = H' = 0$. This then, is a 4R_0 .

We have not depicted the additional coexistence surfaces, called "wings," in Fig. 2(a) [Fig. 2(b)] because they occur at nonzero H' [H]. A naive analysis might suppose that there were eight wings, two corresponding to each (3X_2 , 3R_1) pair. To understand correctly the relationships between the 3R_1 and all the 2R_2 which intersect at them, we have to analyze the system in the $T = 0$ hyperplane.

III. $T = 0$ PHASE DIAGRAM AND ITS IMPLICATIONS FOR $T > 0$

The $T = 0$ phase diagram is depicted in Fig. 3. In the ground state, the surfaces where two phases are in equilibrium occur at places where the energies of the two phases are equal. The energies of the

ferromagnetic phases are given by $E_F^\pm = -\mathcal{R} \mp H$ and the energies of the antiferromagnetic ("metamagnetic") phases are given by $E_A^\pm = +\mathcal{R} \mp H'$ where we have put $J = \mu = 1$. Setting these energies pairwise equal, we obtain the six different surfaces given in Table I. The equations of the four lines where three surfaces intersect and where three phases are in equilibrium can be similarly obtained and are also given in Table I. Because of the *linear* forms of E_F^\pm and E_A^\pm , the surfaces are flat and the lines are straight.

The $H' = 0$ plane of Fig. 3 coincides with the $T = 0$ plane of Fig. 2(a). As one moves from Fig. 3 to Fig. 2(a), it can be seen that a space of dimension d where p phases coexist (pX_d) gives rise to a space of dimension $(d + 1)$ where p phases coexist. For instance the lines L_1 and L_2 in Fig. 3 become the 3X_2 in Fig. 2(a); similar considerations of $[A^+, A^-]$ or the 4X_0 convinces one of the validity of the equation

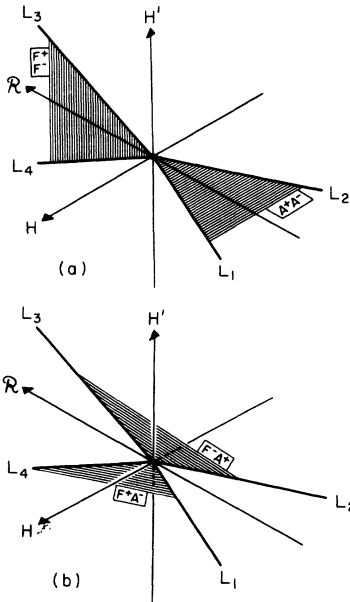


FIG. 3. Section of the (T, H, H', \mathcal{R}) field space with $T = 0$. (a) Shown here are two of the six 2X_2 planes (where two phases coexist) and the four 3X_1 lines (where three phases coexist) which bound these planes. The plane bounded by L_1, L_2 lies in the $H - \mathcal{R}$ plane and separates phases A^\pm , where A^+ is the phase that is stable as $H' \rightarrow +\infty$, and A^- is stable as $H' \rightarrow -\infty$. It generates, as T increases, the mountain (2X_3) and, eventually, the snow cap (2R_2) of Fig. 2(a). The plane bounded by L_3, L_4 lies in the $H' - \mathcal{R}$ plane and separates the two ferromagnetic phases F^\pm . At finite T it generates the picture in Fig. 2(b). (b) Shown are two more of the six 2X_2 planes where two planes coexist [cf. Table I and Fig. 3(a)]. These two 2X_2 become, for $T > 0$, two of the four wings discussed in the text.

TABLE I. Equations for the six coexistence surfaces 2X_2 , where two phases coexist, and for the four coexistence lines 3X_1 , where three phases coexist.

Coexistence surfaces	(Planes 2X_2)
$[F^+, F^-]$ [Fig. 3(a)]	$H = 0$
$[A^+, A^-]$ [Fig. 3(a)]	$H' = 0$
$[F^+, A^-]$ [Fig. 3(b)]	$2\mathcal{R} + H + H' = 0$
$[F^-, A^+]$ [Fig. 3(b)]	$2\mathcal{R} - H - H' = 0$
$[F^+, A^+]$	$2\mathcal{R} + H - H' = 0$
$[F^-, A^-]$	$2\mathcal{R} - H + H' = 0$
Coexistence lines	(3X_1)
$[F^+, F^-, A^+]$	$H = 0$
	$2\mathcal{R} - H' = 0$
$[F^+, F^-, A^-]$	$H = 0$
	$2\mathcal{R} + H' = 0$
$[A^+, A^-, F^+]$	$H' = 0$
	$2\mathcal{R} + H = 0$
$[A^+, A^-, F^-]$	$H' = 0$
	$2\mathcal{R} - H = 0$

$${}^pX_d (T = 0 \text{ hypersurface}) \rightarrow {}^pX_{d+1} (T > 0). \quad (7a)$$

The fate of a ${}^pX_{d+1}$ in Fig. 2(a) is particularly simple as T increases. The 2X_3 ends in a surface of ordinary critical points—a 2R_2 ; the 3X_2 ends in a line of critical points of order three—a 3R_1 ; the 4X_1 ends in a 4R_0 . Thus we find

$${}^pX_{d+1} \rightarrow {}^pX_d. \quad (7b)$$

Equations (7a) and (7b)⁹ combine to show that for this model there is a homeomorphic mapping (i. e., a continuous, nonsingular transformation) between the pX_d in the $T = 0$ hyperplane and the pR_d (where $\mathcal{O} = p$) in the full phase diagram. Thus one can deduce topological relationships between the various pR_d in the full field space by inspecting the relationships between the pX_d in the $T = 0$ phase diagram. Since the full space is four dimensional and the $T = 0$ phase diagram is only three dimensional, this is of considerable value.

The first deduction we may make is that because there are six coexistence surfaces (2X_2) in the $T = 0$ phase diagram, there are also six distinct surfaces of critical points (2R_2) in the complete phase diagram. Two of these six are shown in Figs. 2(a) and 2(b); these are the "snowcaps" labeled 2R_2 . The snowcap of Fig. 2(a) corresponds to the surface in the $T = 0$ phase diagram of Fig. 3(a) on which the phases $[A^+, A^-]$ coexist, while the snowcap of Fig. 2(b) corresponds to the coexistence surface labeled $[F^+, F^-]$ of Fig. 3(a).

The remaining four surfaces, $[F^+, A^+]$, $[F^-, A^-]$, $[F^+, A^-]$, and $[F^-, A^+]$, of which the last two are shown in Fig. 3(b), connect L_1, L_2 (for $\mathcal{R} < 0$) to L_3, L_4 (for $\mathcal{R} > 0$). The four lines L_1, L_2, L_3 , and L_4 meeting at the origin in Figs. 3(a) and 3(b) correspond [in the sense of Eqs. (7a) and (7b)] to the four lines of tricritical points meeting at the 4R_0 in the full phase diagram. Thus we deduce that as the temperature is increased, the four surfaces $[F^+, A^+]$ evolve into the critical surfaces bounding the wings, and that these boundaries connect those tricritical lines (3R_1) for $\mathcal{R} < 0$ shown in Fig. 2(a) to those tricritical lines for $\mathcal{R} > 0$ shown in Fig. 2(b). Each of the four tricritical lines of Figs. 2(a) and 2(b) is thereby connected to each of the other tricritical lines by a 2R_2 surface. Indeed, as we initially pointed out, there are $(\frac{4}{2}) = 6$ critical surfaces.

IV. SCALING HYPOTHESIS FOR THE 4R_0

We can make a scaling hypothesis at the 4R_0 . It is believed¹⁰ that in the variables of Fig. 2(a), a hypothesis of the form $G(\lambda^{\mathcal{R}}\tau, \lambda^{\mathcal{R}}H, \lambda^{\mathcal{R}}\mathcal{R}) = \lambda G(\tau, H, \mathcal{R})$ is valid where $\tau \equiv T - T_c$ ($\mathcal{R} = 0$). However, Eq. (2) indicates that if a scaling hypothesis is valid in H , it is also valid in H' . Hence we make the full scaling hypothesis at the 4R_0 ,

$$G(\lambda^{\mathcal{R}}\tau, \lambda^{\mathcal{R}}H, \lambda^{\mathcal{R}}H', \lambda^{\mathcal{R}}\mathcal{R}) = \lambda G(\tau, H, H', \mathcal{R}) . \quad (8)$$

This scaling hypothesis has four principal directions of scaling and correspondingly four scaling powers ($a_\tau, a_H, a_{H'}$, and $a_{\mathcal{R}}$). This situation will always occur at spaces of points where a special scaling hypothesis is valid, and there are four independent directions out of the critical space (here the critical space, a 4R_0 , is a point). In the present model the 4R_0 occurs because four lines of tricritical points intersect, but in general more complex possibilities may exist.

In the particular case considered here, there is a very large degree of symmetry, and this reduces the number of independent scaling powers. Since the symmetry given by Eq. (2) interchanges H and H' , we expect

$$a_H = a_{H'} . \quad (9a)$$

There is a second relationship between the scaling powers that may be obtained from the result¹¹ $\varphi = \gamma$, where φ is the crossover exponent describing change of lattice dimensionality and γ is the

susceptibility exponent for the $\mathcal{R} = 0$ system. In terms of scaling powers,¹² $\varphi \equiv a_{\mathcal{R}}/a_\tau$ and $-\gamma = (1 - 2a_H)/a_\tau$. We therefore obtain

$$a_{\mathcal{R}} = 2a_H - 1 . \quad (9b)$$

From (9a) and (9b), we see that there are only two (not four) independent exponents at the special point (e. g., a_τ and a_H).

In the two-dimensional H, T plane, where $\mathcal{R} = 0$ and $H' = 0$, the model is a set of decoupled Ising planes and the exponents at the critical point on the temperature axis are therefore equal to the exponents for the $d = 2$ Ising model. For this model, we can use the rigorous values¹³ $\alpha = 0$ and $\beta = \frac{1}{8}$ together with the scaling relations¹²

$$-\alpha = (1 - 2a_\tau)/a_\tau, \quad \beta = (1 - a_H)/a_\tau \quad (10)$$

to obtain actual numerical values for all four scaling powers. These are

$$a_\tau = \frac{1}{2}, \quad a_{\mathcal{R}} = \frac{7}{8}, \quad a_H = a_{H'} = \frac{15}{16} . \quad (11)$$

Note that from (11), all possible thermodynamic exponents can be exactly calculated.

From (11) we observe that the scaling powers satisfy

$$a_\tau < a_{\mathcal{R}} < a_H = a_{H'} . \quad (12)$$

Since a_τ is the smallest scaling powers, it is to be expected that all lines and surfaces on which singularities of the Gibbs function occur and which extend to the critical point of order four, will approach the point of order four tangentially to the T axis. This is entirely analogous to the prediction of how lines of critical points approach tricritical points.¹⁴ In Fig. 2, we have, therefore, depicted the lines of tricritical points as approaching the 4R_0 tangentially to the T axis.

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other publications and meeting talks. Here we shorten the notation to pX_d and oR_d in order that superscripts or subscripts not be attached to an acronym; (c) R. B. Griffiths, in *Critical Phenomena in Alloys, Magnets and Superconductors*, edited by R. E. Mills, E. Ascher, and R. I. Jaffee (McGraw-Hill, New York, 1971), pp. 377-391.

⁶An alternative definition of order of a critical point may be formulated in terms of the number of phases becoming critical simultaneously at the point. These two definitions coincide for the model discussed here. However, the alternative definition may be more appropriate for complex fluid mixtures. See, e.g., R. B. Griffiths and B. Widom (unpublished); T. S. Chang, A. Hankey, and H. E. Stanley, *Phys. Rev. B* **8**, 346 (1973); A. Hankey, T. S. Chang, and H. E. Stanley, *Phys. Rev. B* **8**, 1178 (1973).

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⁹It must be emphasized that both Eqs. (7a) and (7b) have exceptions. For example, (7a) is not valid in a $d = 1$ Ising model with a finite-range interaction, where the "critical

point" is at $T = 0$. An exception to (7b) is provided by binary liquid mixtures. Here the line of liquid-liquid-vapor coexistence points (3X_1) does not usually end in a 3R_0 . Rather the 2R_1 line of liquid-liquid critical points intervenes and meets the gas-liquid coexistence surface at an interior point. We should like to thank Professor J. C. Wheeler for indicating this last point.

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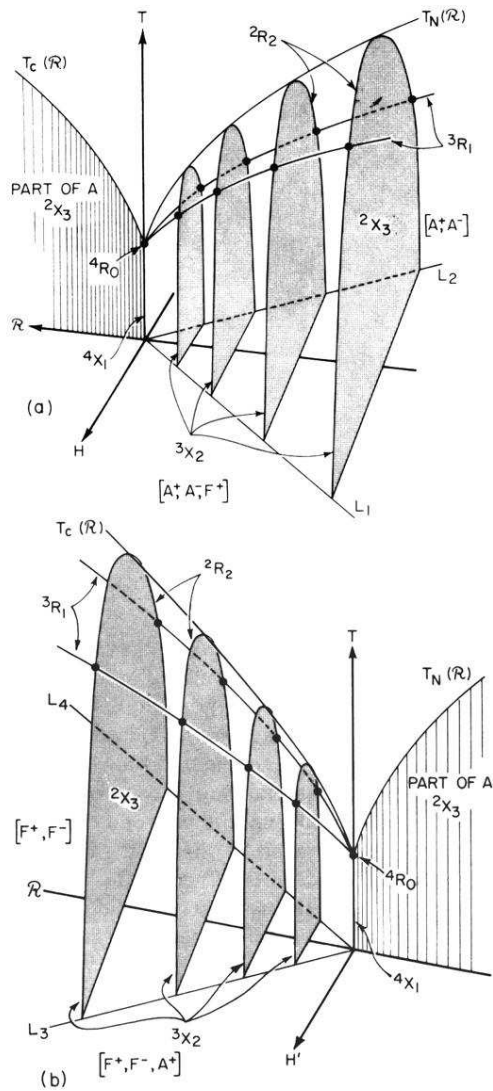


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