PHYSICAL REVIEW B VOLUME 4, NUMBER 9 1 NOVEMBER 1971

Critical Behavior of a Magnetic Alloy

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(Received 19 February 1971)

A classical lattice model for a binary alloy which consists of a ferromagnetic and a nonmagnetic component is investigated. Two coupling constants, a magnetic and a nonmagnetic one, describe the interactions between particles on nearest-neighbor sites. For vanishing magnetic coupling the model can be reduced to the Ising model from which the coexistence surface is determined exactly. The critical behavior is discussed extensively within the mean-field approximation. The critical line and the coexistence surface define three classes of directions (in the space of intensive variables) with respect to which the various critical exponents are calculated and classified.

I. INTRODUCTION

A classical configurational-lattice model for a binary alloy has been proposed recently.[1,2] The model consists of a ferromagnetic component A and a nonmagnetic component B, and neighboring A spins are exchange coupled to each other by a magnetic coupling constant ϵ_1 while a nonmagnetic coupling constant ϵ_2 favors the occurrence of AA neighbor pairs (we assume ϵ_1, ϵ_2 non-negative). An external magnetic field h is coupled to the A spins and, furthermore, the two components are assigned different chemical potentials μ_A and μ_B.

This model is suitable to describe the mutual influence of two different phase transitions. On the one hand, a ferromagnetic transition occurs at a magnetic ordering temperature if the A-atom concentration is sufficiently high and, on the other hand, a miscibility gap becomes evident at low temperatures. The two order parameters, which correspond to these phase transitions, are interrelated in general.

The study of this model is rather complex. So far, only partial results have been obtained. In one dimension, where no phase transitions occur, the model was solved exactly.[2,3] The one-dimensional dilute Ising model, which corresponds to the case $\epsilon_2 = 0$, has been solved earlier.[4,5] The case $\epsilon_2 = 0$ was proposed to describe the λ curve and the critical behavior of a He^3-He^4 mixture.[6] Mean-field solutions for this system are given in Refs. 6 and 7. Exact high-temperature series expansions were performed for zero field,[8] from which the location of the critical points was extrapolated. For the general case ($\epsilon_1 > 0$, $\epsilon_2 > 0$) the stable mean-field solutions and exact series expansions will be discussed in forthcoming papers.[9]

Here we first give the mathematical description of the model and formulate the mean-field equations for the general case (Sec. II). In Sec. III we solve the special case $\epsilon_1 = 0$ by reducing it to the Ising model. The coexistence surface is given exactly in terms of the intensive variables $\mu = \mu_A - \mu_B$, h, and T. In Sec. IV the behavior in the neighborhood of the critical line is discussed by calculating explicitly all the interesting critical exponents within the mean-field approximation. A simple classification scheme for the critical exponents is introduced. Section V contains some additional remarks.

II. MODEL

The two kinds of particles A and B are located on the sites of a regular lattice. The occupation of site i is described by the site variable S_i ($i = 1, 2, \ldots, N$):

$$S_i = \pm 1, \quad A \text{ particle at site } i \text{ with spin } \begin{cases} \text{up} \\ \text{down} \end{cases}$$

$$= 0, \quad B \text{ particle at site } i \text{ (no spin)} .$$

The Hamiltonian of the system,

$$H = -\epsilon_1 \sum_{\langle ij \rangle} S_i S_j - \epsilon_2 \sum_{\langle ij \rangle} S_i^2 S_j^2 - h \sum_{i=1}^{N} S_i - \mu \sum_{i=1}^{N} S_i^2 , \tag{1}$$

depends on a magnetic coupling constant ϵ_1, a nonmagnetic coupling constant ϵ_2, the external magnetic field h, and the difference of the chemical potentials $\mu = \mu_A - \mu_B$ of the A and B particles, respectively. $\sum_{\langle ij \rangle}$ indicates a sum over nearest-neighbor pairs.

The thermodynamic behavior of the system is described by the free energy

$$F = -T \ln Z , \tag{2}$$

where the partition function Z is given by

$$Z = \sum_{\{S_i\}} e^{-H/T} . \tag{3}$$

(Throughout this paper we put $k_B = 1$.)

In the mean-field approximation (MFA), which is equivalent to the Bragg-Williams approximation, the model is described by two coupled nonlinear self-consistency equations

$$m = \rho \tanh\left(\frac{\epsilon_1 m + h}{T}\right) , \tag{4}$$

$$\frac{\rho}{1-\rho} = 2 \exp\left(\frac{\epsilon_2 \rho + \mu}{T}\right) \cosh\left(\frac{\epsilon_1 m + h}{T}\right) . \tag{5}$$

Here $m = \langle S_i \rangle$ is the magnetization, $\rho = \langle S_i^2 \rangle$ the density (of the A particles)—the brackets denote ensemble averages in the grand canonical scheme—and $\epsilon_{1,2} = z\epsilon_{1,2}$ (z is the number of nearest neighbors). In every point of the space of intensive variables ("fields") μ, h, and T, Eqs. (4) and (5) determine $m(\mu, h, T)$ and $\rho(\mu, h, T)$. If several solutions are possible, the stable one is singled out by the requirement that the MFA free energy

$$F(\mu, h, T) = \tfrac{1}{2}\epsilon_1 m^2 + \tfrac{1}{2}\epsilon_2 \rho^2 + T \ln(1-\rho) \tag{6}$$

is minimized:

$$F = \text{minimum (at constant } \mu, h, T) . \tag{7}$$

Even in the MFA the general case shows a rather complex behavior. New types of critical points will occur, as, for example, the so-called tricritical point, discussed in Ref. 6 for the case $\epsilon_2 = 0$. An extensive analysis will be given in Ref. 9.

III. SOLUTION OF CASE $\epsilon_1 = 0$

As a preliminary step it is useful to study some special cases. The case $\epsilon_2 = 0$ has been discussed by several authors.[6,7]

In this paper we solve the alternative case $\epsilon_1 = 0$. Here the A spins are coupled to an external magnetic field only, so that the model describes, for instance, a magnetic alloy without exchange coupling or a paramagnetic lattice gas.

Using a generalization of an argument given in Ref. 10, this problem can be reduced to the Ising problem. Putting $\sigma_i = 2S_i^2 - 1$, the sum over all configurations $\{S_i\}$ is split into a sum over $S_i = \pm 1$ (for all i with $\sigma_i = +1$) followed by a sum over $\{\sigma_i\}$. Then the partition function (3) can be written as

$$Z = \sum_{\{\sigma_i\}} \prod_{\langle ij \rangle} \exp\left[\frac{\epsilon_2}{T}\left(\frac{\sigma_i+1}{2}\right)\left(\frac{\sigma_j+1}{2}\right)\right] \times \prod_{i=1}^{N} \left[2e^{\mu/T} \cosh\left(\frac{h}{T}\right)\right]^{(\sigma_i+1)/2} . \tag{8}$$

Therefore the *exact* free energy per site is related to the corresponding one of the Ising model:

$$F_{\epsilon_1=0,\epsilon_2}(\mu, h, T) = F_J^{\text{Ising}}(h^*, T) - h^* + \tfrac{1}{8} z \epsilon_2 , \tag{9}$$

where

$$J = \tfrac{1}{4}\epsilon_2 , \tag{10}$$

$$h^* = \tfrac{1}{2}\{\mu + \tfrac{1}{2} z \epsilon_2 + T \ln[2 \cosh(h/T)]\} . \tag{11}$$

From Eq. (9) it follows that the coexistence surface Σ of our model is described by

$$h^* = 0, \quad T < T_c^{\text{Ising}} . \tag{12}$$

It is symmetric with respect to $h = 0$. The phase diagram in the (μ, h, T) space is shown in Fig. 1. On passing through Σ the system undergoes a first-order transition, i.e., the "densities" ρ, m, and s (entropy) exhibit a discontinuity. At $T = T_c^{\text{Ising}}$, Σ terminates in the line of critical points (critical line) Γ, which is therefore given by

$$h^* = 0, \quad T = T_c^{\text{Ising}} . \tag{13}$$

Note that the MFA gives the same equation for Σ. The location of T_c, however, is shifted with respect to the Ising value ($T_c^{\text{MFA}} = \frac{1}{2}$).

With the help of Eq. (9) all thermodynamical quantities of the model can be expressed in terms of the Ising quantities. For the densities (first-order derivatives of the free energy) we obtain

$$\rho \equiv -\left(\frac{\partial F}{\partial \mu}\right)_{h,T} = \tfrac{1}{2}(m^{\text{Ising}} + 1) , \tag{14}$$

$$m \equiv -\left(\frac{\partial F}{\partial h}\right)_{\mu,T} = \tfrac{1}{2}(m^{\text{Ising}} + 1)\tanh(h/T) , \tag{15}$$

$$s \equiv -\left(\frac{\partial F}{\partial T}\right)_{\mu,h} = s^{\text{Ising}} + \tfrac{1}{2}(m^{\text{Ising}} + 1) \times \{\ln[2\cosh(h/T)] - (h/T)\tanh(h/T)\} . \tag{16}$$

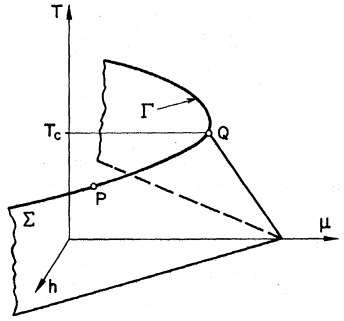

FIG. 1. Exact phase diagram for $\epsilon_1 = 0$ in the intensive variable space.

The second-order derivatives, i.e., the heat capacity $c_{\mu,h}$, the analog of the compressibility $\kappa_{h,T}$, and the magnetic susceptibility $\chi_{\mu,T}$ are expressed as follows:

$$c_{\mu,h} \equiv T\left(\frac{\partial s}{\partial T}\right)_{\mu,h} = c_{h^*}^{\text{Ising}} + T\left(\frac{\partial m^{\text{Ising}}}{\partial T}\right)_{h^*}\frac{\partial h^*}{\partial T} + T(m^{\text{Ising}}+1)\frac{\partial^2 h^*}{\partial T^2}\ , \tag{17}$$

$$\kappa_{h,T} \equiv \left(\frac{\partial \rho}{\partial \mu}\right)_{h,T} = \tfrac{1}{4}\chi_T^{\text{Ising}}\ , \tag{18}$$

$$\chi_{\mu,T} \equiv \left(\frac{\partial m}{\partial h}\right)_{\mu,T} = \tfrac{1}{4}\chi_T^{\text{Ising}}\tanh^2(h/T) + \frac{m^{\text{Ising}}+1}{2T\cosh^2(h/T)}\ . \tag{19}$$

IV. CRITICAL BEHAVIOR

The critical properties of the model are described by the behavior of the thermodynamic quantities near a critical point (on Γ). Through Eqs. (14)–(19) this critical behavior is given, in principle, by that of the Ising model. The critical exponents of the Ising quantities in question, however, are not all known. In order to get a complete table of exponents, we therefore calculate and discuss the MFA critical exponents, which are determined analytically by Eqs. (4), (5), (12), and (13) (with $e_1 = 0$ and T_c^{Ising} replaced by $T_c^{\text{MFA}} = \frac{1}{2}$). It turns out that they can be classified within a simple scheme. For this purpose we define at a critical point P (on Γ) three classes C_i $(i = 1, 2, 3)$ of directions. C_1 contains the directions parallel to Γ, C_2 all directions parallel to the coexistence surface Σ (but not parallel to Γ), and C_3 all directions not parallel to Σ. Furthermore, we let F_i and F_{ik} denoted first- and second-order derivatives of F with respect to any direction of class C_i and of classes C_i and C_k, respectively. On approaching P asymptotically in a direction of class C_l, the critical exponents corresponding to F_i and F_{ik} depend only on the index pair $\{i, l\}$ and the index triplet $\{i, k, l\}$, respectively.[11,12]

The three different ways of approaching P are denoted as follows: case 1, asymptotically parallel to Γ; case 2, asymptotically parallel to Σ but not parallel to Γ; and case 3, not parallel to Σ. The position of P on Γ is irrelevant for the critical behavior of the model.

In a critical point $P(\mu_c, h_c, T_c)$ with $h_c \neq 0$ the densities ρ, m, and s are all of type F_3. On approaching P they behave as

$$|\rho-\rho_c|,\ |m-m_c|,\ |s-s_c|$$

$$\sim |\mu-\mu_c|^{2/3} \text{ or } |h-h_c|^{2/3} \quad \text{(case 1)}$$

$$\sim |\mu-\mu_c|^{1/2} \text{ or } |h-h_c|^{1/2} \text{ or } |T-T_c|^{1/2} \quad \text{(case 2)}$$

$$\sim |\mu-\mu_c|^{1/3} \text{ or } |h-h_c|^{1/3} \text{ or } |T-T_c|^{1/3} \quad \text{(case 3)}.$$

The second-order derivatives $c_{\mu,h}$, $\kappa_{h,T}$, and $\chi_{\mu,T}$ are of type F_{33} and diverge as

$$c_{\mu,h},\ \kappa_{h,T},\ \chi_{\mu,T}$$

$$\sim |\mu-\mu_c|^{-4/3} \text{ or } |h-h_c|^{-4/3} \quad \text{(case 1)}$$

$$\sim |\mu-\mu_c|^{-1} \text{ or } |h-h_c|^{-1} \text{ or } |T-T_c|^{-1} \quad \text{(case 2)}$$

$$\sim |\mu-\mu_c|^{-2/3} \text{ or } |h-h_c|^{-2/3} \text{ or } |T-T_c|^{-2/3} \quad \text{(case 3)}.$$

In the point $Q(\mu_c, h_c = 0, T_c)$ the magnetization m is of type F_1 and therefore has a different critical behavior ($m_c = 0$, $h_c = 0$):

$$m \sim h^1 \quad \text{(case 1)}$$

$$\sim |\mu-\mu_c|^1 \text{ or } h^1 \text{ or } |T-T_c|^1 \quad \text{(cases 2 and 3)}$$

($m \equiv 0$ if Q is approached in the plane $h = 0$).

The susceptibility $\chi_{\mu,T}$ in Q is of type F_{11} and approaches a finite value as

$$|\chi_{\mu,T} - \chi_{\mu,T}^c| \sim |h|^{2/3} \quad \text{(case 1)}$$

$$\sim |\mu-\mu_c|^{1/2} \text{ or } |h|^{1/2} \text{ or } |T-T_c|^{1/2} \quad \text{(case 2)}$$

$$\sim |\mu-\mu_c|^{1/3} \text{ or } |h|^{1/3} \text{ or } |T-T_c|^{1/3} \quad \text{(case 3)}.$$

Thus F_{11} behaves like the density F_3.

We finally note that "nondiagonal" quantities like $c_{\rho,h}$ or $\kappa_{m,T}$ are of type F_{13} and diverge *more weakly* than their diagonal analogs (compare Ref. 12). As an example we have in Q

$$\kappa_{m,T} \sim h^{-2/3},\quad \kappa_{h,T} \sim h^{-4/3} \quad \text{(case 1)}.$$

V. REMARKS

(i) The idea of a different (singular) behavior of the thermodynamic quantities, as a critical point is approached in different ways, was introduced as a general postulate in the theory of phase transitions[11,12] and has been discussed in a recent paper.[13] The above calculations on our model reveal a simple classification scheme of the critical

indices.

(ii) For the special case $\epsilon_1 = 0$ no tricritical point occurs, in contrast to the case $\epsilon_2 = 0$. The exceptional behavior of the thermodynamical quantities in the critical point $Q(\mu_c, 0, T_c)$ is explained by the fact that here Γ is parallel to the h axis.

(iii) The model may be used for several different physical systems. For instance, if $S_i = 0$ denotes a vacancy on site i, a *paramagnetic lattice gas* is described. The susceptibility diverges even in the absence of ferromagnetic coupling if a critical point P ($\neq Q$) is approached. The critical exponents of the van der Waals gas for the compressibility and the specific heat coincide with ours.

*Supported by the Schweizerischer Nationalfonds.

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PHYSICAL REVIEW B VOLUME 4, NUMBER 9 1 NOVEMBER 1971

Neutron Orbital Cross Section for a Tight-Binding Model of Paramagnetic Nickel

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(Received 5 February 1971)

The orbital contribution to the magnetic partial differential neutron cross section is calculated for a realistic band model of paramagnetic nickel within the tight-binding scheme. The orbital contribution is generally less than one-quarter that of the spin contribution in an energy range up to ~ 0.15 eV. At higher energies it exceeds the spin contribution and should be observable.

I. INTRODUCTION

In the past few years there have been several theoretical and experimental studies aimed at understanding the generalized electron-spin susceptibility $\chi_s(\vec{\kappa}, \omega)$ of magnetic metals.[1-4] $\chi_s(\vec{\kappa}, \omega)$ measures the response of electrons to an external perturbation, of frequency ω and wave vector $\vec{\kappa}$, that couples to their spin; i.e., it describes electron-spin dynamics. Efficient and accurate band-structure calculations have made realistic calculations of χ_s possible. This work has been stimulated by experimental studies, especially thermal neutron scattering experiments.

The neutron-electron interaction evaluated to leading order in the reciprocal of the neutron mass is the sum of two terms,[5] the spin and orbital interactions. The contribution to the neutron cross section from the former is simply related to $\chi_s(\vec{\kappa}, \omega)$. For small scattering wave vectors, Elliott[1] has argued that the matrix element of the orbital interaction operator is a factor m_e/m^* smaller than that of the spin interaction, and it has been assumed to be negligible in experimental analysis. Since fine detail can be measured by neutron scattering, and measurements are not restricted to small wave vectors, the orbital contribution to the neutron cross section now warrants a more complete study. As a first step we have calculated the orbital contribution to the magnetic neutron cross section for a tight-binding model of paramagnetic nickel and compared it with the spin contribution. The latter dominates for small $\vec{\kappa}$ and ω. The two contributions become comparable for $\hbar\omega \sim 0.15$ eV