A tricritical model with an experimentally accessible ordering field

1975 J. Phys. A: Math. Gen. 8 913

(http://iopscience.iop.org/0305-4470/8/6/010)

J. Phys. A: Math. Gen., Vol. 8, No. 6, 1975. Printed in Great Britain.

A tricritical model with an experimentally accessible ordering field

M R H Khajehpour†, Robert A Kromhout† and Yung-Li Wang‡
Department of Physics, The Florida State University, Tallahassee, Florida 32306, USA

Received 4 October 1974

Abstract. The Blume–Capel model in a transverse field is studied in the mean field approximation. We find that a tricritical point exists in the transverse field–temperature plane for a range of values of anisotropy to exchange interaction ratio. This is in contrast with the original Blume–Capel model which shows the tricritical phenomena only at a single value of this ratio. The most interesting feature, however, is that the ordering field, which is the externally applied field along the longitudinal direction, is experimentally accessible and can be set at one's will. This allows an experimentalist to investigate all aspects of a system describable by this model. Equations of the lines of critical points including the wing boundaries are derived. Phase diagrams for the whole range of values of anisotropy are described, and the experimental situation is discussed.

1. Introduction

Great progress has been made in understanding critical phenomena in recent years. While behaviour near the critical point caught the early attention and has been studied extensively and thoroughly, tricritical phenomena have received the attention they deserve only recently, after Griffith (1970) pointed out the different features of a system around the tricritical point in comparison with an ordinary critical point. Tricritical phenomena, however, prevail in many physical systems, notably: metamagnets, ^{3}He–^{4}He mixtures, multi-component fluid mixtures, NH_4Cl, and certain anisotropic ferromagnets. In the space of thermodynamic fields, the tricritical point separates a first-order transition line from a line of critical points. A conceptually more advantageous picture can be obtained, as noted by Griffith (1970), by introducing the thermodynamic ordering 'field' η, conjugate to the order parameter. Then in the η–T–H space, for example, for a metamagnet, there are three coexistence surfaces each bounded by a critical line intersecting along the first-order transition line. The boundaries of the surfaces meet together at a point, the tricritical point. In the ordinary metamagnet such as $FeCl_2$, η is the fictitious staggered field. Because of the unphysical nature of η, the only experimentally accessible region in this space is the original $\eta = 0$ plane. However, very recently (Blume *et al* 1974) it was found that η is not necessarily fictitious, but can manifest itself in a class of antiferromagnets in which DAG is a representative. Because of the peculiar symmetry of these crystals, a staggered field is induced as an external magnetic field is applied. The persistence of such a staggered field caused the phase transition observed in a magnetic field to be a first-order phase transition on one of the 'wings' bifurcating from the first-order transition line; in this instance the $\eta = 0$

† Supported in part by the Committee on Faculty Research Support (FSU).
‡ Supported in part by National Science Foundation under Grant No GH-40174.

plane becomes experimentally inaccessible and the experimentally observable space is a curved surface on which $\eta = \eta(H_z, T)$. The staggered field is thus not really a freely adjustable field and as a result, the tricritical point is not approachable in such a crystal in the laboratory.

To allow for a complete investigation of the behaviour near the tricritical point, it is highly desirable to obtain a tricritical system in which the ordering field η is entirely at one's disposal. Indeed, such models exist and the simplest one is the Blume–Capel model (Blume 1966, Capel 1966) in a transverse field. The Blume–Capel model assumes a ferromagnetic Ising exchange interaction for a system of spins of magnitude one and a single ion anisotropy $D(S^z)^2$ which gives rise to a zero field splitting. D is taken to be positive so the system is one of the singlet ground state systems.

It has been shown that in the Blume–Capel model a first-order phase transition occurs in a narrow range of values of the anisotropy to exchange interaction ratio and the tricritical phenomena arise only at a single value of this ratio. However, in a transverse field the tricritical point develops into a line. Thus, instead of a single value there is a range of values of the anisotropy for which the system would display the tricritical phenomena. Unlike the case of a metamagnet, the ordering field η here is the externally applied magnetic field along the z axis; the disordering field is clearly the transverse field H_x. In the space of H_z, H_x and T, we obtain, in the plane $H_z = 0$, a first-order transition line which joins to a critical line at the tricritical point, and two coexistence surfaces for $H_z \neq 0$, the 'wings', which bifurcate from the first-order transition line. This clearly is the familiar diagram for a tricritical system. The important feature of the model considered here is, however, that all the fields are physical and can be set at one's will. This makes possible a complete investigation of the system in the laboratory.

In this paper we study theoretically the behaviour of the Blume–Capel model in a transverse field. The mean field approximation (MFA) is used. Concentrating on tricritical phenomena we obtain the critical curves bounding the coexistence surfaces including the 'wings'. Phase transitions on the wings are also studied. Varying the fields, the transition point can be pushed towards the wing critical end point; the size of the discontinuous jump in magnetization is shown to decrease towards zero. The different behaviours for the whole range of anisotropy constant are discussed in detail. The organization of the paper is as follows.

In the next section we introduce the Hamiltonian of the Blume–Capel model in a transverse field and study the eigenenergy levels in MFA. Through a Landau expansion of the free energy, in § 3, the equation of the critical line in the plane of zero ordering field is found, and location of the tricritical point obtained. The critical lines in finite ordering field (wing critical end points) are obtained by using an expansion around the critical end point. In § 4 a short discussion of the typical magnetization curves and parallel susceptibility is given. A detailed discussion of phase diagrams for the full range of positive values of anisotropy is given in § 5. Finally in § 6 the main results are summarized and the experimental situation is reviewed.

2. Model Hamiltonian and energy levels

We consider an Ising model with zero field splitting (the Blume–Capel model) in a transverse field H_x:

$$\mathcal{H} = -J\sum_{i,j} S_i^z S_j^z + \Delta \sum_i (S_i^z)^2 - \mu H_z \sum_i S_i^z - \mu H_x \sum_i S_i^x \tag{2.1}$$

where both J and D are assumed positive. In the MFA the Hamiltonian (1) in units of $J(0)$ can be written as

$$\mathcal{H}_m = -2\langle S^z\rangle S^z + D(S^z)^2 - h_z S^z - h_x S^x + \langle S^z\rangle^2 \tag{2.2}$$

where

$$D = \frac{\Delta}{J(0)}, \qquad h_z = \frac{\mu H_z}{J(0)}, \qquad h_x = \frac{\mu H_x}{J(0)} \qquad \text{and} \qquad J(0) = \sum_j J_{ij}.$$

Energy levels of the system in MFA are found by diagonalizing the Hamiltonian (2.2):

$$E_i = D - \lambda_i + \langle S^z\rangle^2 \tag{2.3}$$

where λ_i are the three roots of the cubic equation:

$$\lambda_i^3 - D\lambda_i^2 - (h_x^2 + \alpha^2)\lambda_i + D\alpha^2 = 0 \tag{2.4}$$

$$\alpha = h_z + 2\langle S^z\rangle. \tag{2.5}$$

The λ_i can be found as a power series in α. For small α (eg $h_z = 0$ and $\langle S^z\rangle \to 0$ near a continuous phase transition) an appropriate truncation in series enables us to construct a Landau expansion of free energy:

$$E_i = \epsilon_i + a_i\alpha^2 + b_i\alpha^4 + c_i\alpha^6 + \langle S^z\rangle^2 \tag{2.6}$$

where:

$$\begin{aligned} \epsilon_0 &= D \\ \epsilon_\pm &= \tfrac{1}{2}[D \pm (D^2 + 4h_x^2)^{1/2}] \end{aligned} \tag{2.7}$$

and

$$a_i = \frac{\epsilon_i}{3\epsilon_i^2 - 4D\epsilon_i + D^2 - h_x^2} \tag{2.8}$$

$$b_i = \frac{a_i + (2D - 3\epsilon_i)a_i^2}{3\epsilon_i^2 - 4D\epsilon_i + D^2 - h_x^2} \tag{2.9}$$

$$c_i = \frac{b_i + 2a_i b_i(2D - 3\epsilon_i) - a_i^3}{3\epsilon_i^2 - 4D\epsilon_i + D^2 - h_x^2}. \tag{2.10}$$

At zero temperature, for $h_z = 0$ and small $\langle S^z\rangle$, E_- is the ground state energy of the system:

$$E_- = \epsilon_- + (4a_- + 1)\langle S^z\rangle^2 + 16b_-\langle S^z\rangle^4 + 64c_-\langle S^z\rangle^6. \tag{2.11}$$

For D less than a certain value D_t, $\langle S^z\rangle$ tends to zero continuously as h_x is increased, and the system evolves into a paramagnetic state with energy ϵ_-. This transition happens when $4a_- + 1 = 0$. For $D = D_t$, the transition to the paramagnetic state occurs continuously at $h_x = h_t$. D_t and h_t are found from the following equations:

$$\begin{aligned} 4a_- + 1 &= 0 \\ b_- &= 0. \end{aligned} \tag{2.12}$$

Solving equation (2.12) for D_t and h_t, one gets $D_t = 0{\cdot}584$ and $h_t = 1{\cdot}20$. For $D > D_t$, $\langle S^z\rangle$ does not vanish continuously, and drops to zero from a finite value at a critical field.

When h_z is finite, no phase transition is expected for $D < D_t$. For $D > D_t$ a first-order phase transition may occur. The jump in moment in this instance decreases as h_x and h_z approach certain critical values depending on D and finally vanishes at h_z^c and h_x^c. To determine the behaviour near this critical point, the energy levels should be expanded about $\alpha_c = h_z^c + 2\langle S^z\rangle^c$. The expansion involves odd order terms in $(\alpha - \alpha_c)$, and the coefficients are α_c dependent. If we let

$$\alpha = \alpha_c + \delta\alpha$$

then the energy levels are:

$$E_i = \varepsilon_i + a_i\delta\alpha + b_i\delta\alpha^2 + c_i\delta\alpha^3 \tag{2.13}$$

where ϵ_i are the solutions of the cubic equation:

$$\varepsilon_i^3 - 2D\varepsilon_i^2 + (D^2 - h_x^2 - \alpha_c^2)\varepsilon_i + Dh^2 = 0 \tag{2.14}$$

and a_i, b_i and c_i are given by:

$$a_i = \frac{2\alpha_c\varepsilon_i}{f_i} \tag{2.15a}$$

$$b_i = -\frac{a_i^2(3\varepsilon_i - 2D) - 2\alpha_c a_i - \varepsilon_i}{f_i} \tag{2.15b}$$

$$c_i = -\frac{2a_ib_i(3\varepsilon_i - 2D) + a_i^3 - a_i - 2\alpha_c b_i}{f_i} \tag{2.15c}$$

$$f_i = 3\varepsilon_i^2 - 4D\varepsilon_i + D^2 - h_x^2 - \alpha_c^2. \tag{2.15d}$$

We now proceed to find the free energy and study the critical behaviour at finite temperatures.

3. Critical lines and tricritical point at finite temperature

When h_z approaches zero, we can use the energy levels given by equation (2.6) to find the free energy as a Taylor expansion in α. The partition function Z is written up to sixth order in α,

$$Z = (Z_0 + Z_2\alpha^2 + Z_4\alpha^4 + Z_6\alpha^6)\exp(-\beta\langle S^z\rangle^2) \tag{3.1}$$

where

$$Z_0 = \sum_i e^{-\beta\epsilon_i} \tag{3.2a}$$

$$Z_2 = -\beta\sum_i a_i e^{-\beta\epsilon_i} \tag{3.2b}$$

$$Z_4 = -\beta\sum_i (\ell_i - \tfrac{1}{2}\beta a_i^2) e^{-\beta\epsilon_i} \tag{3.2c}$$

$$Z_6 = -\beta\sum_i (c_i - \beta a_i\ell_i + \tfrac{1}{6}\beta^2 a_i^3) e^{-\beta\epsilon_i} \tag{3.2d}$$

and $\beta = 1/T$, the inverse of temperature which is measured in units of $J(0)/k_B$. From (3.1) the free energy is found as

$$\tilde{G} = -\frac{1}{\beta}\ln Z = -\frac{1}{\beta}\ln(Z_0 + Z_2\alpha^2 + Z_4\alpha^4 + Z_6\alpha^6) + \langle S^z\rangle^2. \tag{3.3}$$

Expanding the logarithm in α and keeping terms up to sixth order:

$$\tilde{G} = G_0 + \langle S^z\rangle^2 + \tfrac{1}{2}A\alpha^2 + \tfrac{1}{4}B\alpha^4 + \tfrac{1}{6}C\alpha^6 \tag{3.4}$$

where

$$G_0 = -\frac{1}{\beta}\ln Z_0 \tag{3.5a}$$

$$A = -\frac{2Z_2}{\beta Z_0} \tag{3.5b}$$

$$B = -\frac{4}{\beta}\left[\frac{Z_4}{Z_0} - \frac{1}{2}\left(\frac{Z_2}{Z_0}\right)^2\right] \tag{3.5c}$$

$$C = -\frac{6}{\beta}\left[\frac{Z_6}{Z_0} - \frac{Z_2Z_4}{Z_0^2} + \frac{1}{3}\left(\frac{Z_2}{Z_0}\right)^3\right]. \tag{3.5d}$$

To find the equilibrium values of the parameter $\langle S^z\rangle$, the free energy should be minimized with respect to $\langle S^z\rangle$:

$$\left(\frac{\partial\tilde{G}}{\partial\langle S^z\rangle}\right)_{T,h_z} = 0. \tag{3.6}$$

Thus the equilibrium values of $\langle S^z\rangle$ are given by the equation:

$$\langle S^z\rangle + A\alpha + B\alpha^3 + C\alpha^5 = 0. \tag{3.7}$$

From equation (3.7) one can find $\langle S^z\rangle$ as a function of h_x, h_z and T for fixed D. Substituting for $\langle S^z\rangle$ in equation (3.4) one obtains the Gibbs free energy $G(h_x, h_z, T)$. The Legendre transformation of $G(h_x, h_z, T)$ with respect to the ordering field h_z, is the Helmholtz free energy:

$$F(\langle S^z\rangle, h_x, T) = G(H, T) - \langle S^z\rangle h_z \tag{3.8}$$

where

$$h_z = -\frac{\partial F}{\partial\langle S^z\rangle}. \tag{3.9}$$

To find α as a function of $\langle S^z\rangle$ we invert equation (3.7):

$$\alpha = -\frac{1}{A}\langle S^z\rangle + \frac{B}{A^4}\langle S^z\rangle^3 + \left(\frac{C}{A^6} - \frac{3B^2}{A^7}\right)\langle S^z\rangle^5 + \ldots \tag{3.10}$$

and

$$h_z = \alpha - 2\langle S^z\rangle = -\left(\frac{1}{A} + 2\right)\langle S^z\rangle + \frac{B}{A^4}\langle S^z\rangle^3 + \left(\frac{C}{A^6} - \frac{3B^2}{A^7}\right)\langle S^z\rangle^5. \tag{3.11}$$

And the free energy $F(\langle S^z\rangle; h_x, T)$ can be written as

$$F(\langle S^z\rangle; h_x, T) = F_0 + F_2\langle S^z\rangle^2 + F_4\langle S^z\rangle^4 + F_6\langle S^z\rangle^6 \tag{3.12}$$

where

$$F_2 = -\left(1+\frac{1}{2A}\right) \tag{3.13a}$$

$$F_4 = \frac{1}{4}\frac{B}{A^4} \tag{3.13b}$$

$$F_6 = \frac{1}{A^6}\left(\frac{C}{6}-\frac{1}{2}\frac{B^2}{A}\right). \tag{3.13c}$$

The equations for the λ lines (lines of critical points) can be found from

$$\frac{\partial^2 F}{\partial\langle S^z\rangle^2} = \frac{\partial^3 F}{\partial\langle S^z\rangle^3} = 0 \qquad \frac{\partial^4 F}{\partial\langle S^z\rangle^4} > 0 \tag{3.14}$$

with solutions

(i) $$F_2 = 0, \qquad h_z = 0 \tag{3.15a}$$

(ii) $$F_2 = \frac{3}{5}\frac{F_4^2}{F_6}, \qquad h_z = \pm\frac{16}{25}\frac{F_4^2}{F_6}\left(-\frac{F_4}{5F_6}\right)^{1/2}. \tag{3.15b}$$

In terms of A, B and C, the equations of the λ lines are:

(i) $$A = -\tfrac{1}{2}, \qquad h_z = 0 \tag{3.15c}$$

(ii) $$\frac{9}{40}\frac{B^2A^3}{CA-3B^2}+\frac{1}{2A}+1 = 0, \qquad h_z = \pm\frac{16}{15}\left(\frac{A}{6B}(1+2A)^3\right)^{1/2}. \tag{3.15d}$$

Equation (3.15*c*) gives the equation of the λ line in the $h_z = 0$ plane, and (3.15*d*) those of the wing critical end lines for small h_z. The three λ lines given by equation (3.15*c*) and equation (3.15*d*) meet at a tricritical point $(h_z = 0, h_x^{(t)}, T^{(t)})$ determined by:

$$\begin{aligned} A(h_x^{(t)}, T^{(t)}) &= -\tfrac{1}{2} \\ B(h_x^{(t)}, T^{(t)}) &= 0. \end{aligned} \tag{3.16}$$

To find the critical end points on the wings, we use the energy levels as given by equation (2.13) to construct the free energy about α_c:

$$F = -\frac{1}{\beta}\ln Z = -\frac{1}{\beta}\ln Q + \langle S^z\rangle^2 \tag{3.17}$$

where

$$Q = \sum_i \exp[-\beta(\varepsilon_i + a_i\delta\alpha + b_i\delta\alpha^2 + c_i\delta\alpha^3 + d_i\delta\alpha^4 + \ldots)]. \tag{3.18}$$

Expanding equation (3.18) in powers of $\delta\alpha$:

$$Q = Q_0(T,\alpha_c) + Q_1\delta\alpha - Q_2\delta\alpha^2 + Q_3\delta\alpha^3 + Q_4\delta\alpha^4 + \ldots \tag{3.19}$$

where:

$$Q_1 = -\beta\sum_i a_i\, e^{-\beta\varepsilon_i} \tag{3.20a}$$

$$Q_2 = -\beta\sum_i (b_i - \tfrac{1}{2}\beta a_i^2)\, e^{-\beta\varepsilon_i} \tag{3.20b}$$

$$Q_3 = -\beta \sum_i \left(c_i - \beta a_i b_i + \frac{\beta^2}{3!} a_i^3 \right) e^{-\beta \varepsilon_i} \tag{3.20c}$$

$$Q_4 = -\beta \sum_i \left(d_i - \tfrac{1}{2}\beta b_i^2 + \frac{\beta^2}{2} a_i^2 b_i - \frac{\beta^3}{4!} a_i^4 \right) e^{-\beta \varepsilon_i} \tag{3.20d}$$

where Q_0, Q_1, Q_2, Q_3 and Q_4 depend only upon α_c and T explicitly. Imposing the equilibrium conditions on the free energy equation (3.17):

$$2\langle S^z \rangle = \frac{1}{\beta} \frac{1}{Q} \left(\frac{\partial Q}{\partial \langle S^z \rangle} \right)_{T, h_z} \tag{3.21}$$

or

$$h_z = \alpha_c + \delta\alpha - \frac{1}{\beta} \frac{1}{Q} \left(\frac{\partial Q}{\partial \langle S^z \rangle} \right)_{T, h_z} \tag{3.22}$$

Substituting for Q from equation (3.19):

$$h_z = h_z^c - \frac{1}{\beta Q_0} \left(4Q_2 - \frac{2Q_1^2}{Q_0} - \beta Q_0 \right) \delta\alpha - \frac{1}{\beta Q_0} \left(6Q_3 - \frac{6Q_1 Q_2}{Q_0} + \frac{2Q_1^3}{Q_0^2} \right) \delta\alpha^2 + \ldots \tag{3.23}$$

where

$$h_z^c = \alpha_c - \frac{2Q_1}{\beta Q_0}. \tag{3.24}$$

Now

$$\frac{\partial h_z}{\partial \langle S^z \rangle} = \frac{-[4Q_2 - (2Q_1^2/Q_0) - \beta Q_0]}{4Q_2 - (2Q_1^2/Q_0)} + O(\delta\alpha) \tag{3.25}$$

$$\frac{\partial^2 h_z}{\partial \langle S^z \rangle^2} = \frac{\beta Q_0 [6Q_3 - (6Q_1 Q_2/Q_0) + (2Q_1^3/Q_0^2)]}{[4Q_2 - (2Q_1^2/Q_0)]^2} \left(\frac{\partial h_z}{\partial \langle S^z \rangle} + 2 \right) + O(\delta\alpha). \tag{3.26}$$

At $\alpha = \alpha_c$, ie at critical end point:

$$(4Q_2 - \beta Q_0) Q_0 - 2Q_1^2 = 0 \tag{3.27}$$

$$6Q_3 Q_0^2 - 6Q_1 Q_2 Q_0 + 2Q_1^3 = 0. \tag{3.28}$$

Equations (3.27) and (3.28) together with equation (3.24) determine the coordinates of the wing boundaries in (h_z, T, h_x) space (for fixed D). In the limit of $h_x \to 0$ one gets:

$$h_z^c = \pm T \ln \frac{1 - T + (1 - \frac{3}{2}T)^{1/2}}{[\frac{1}{2}T(2T-1)]^{1/2}} \mp 2(1 - \tfrac{3}{2}T)^{1/2} \tag{3.29}$$

$$D = \tfrac{1}{2} T \ln[8T/(2T-1)]. \tag{3.30}$$

This special case was obtained by Blume *et al* (1971).

4. Magnetization and susceptibility

We only discuss briefly the magnetization $\langle S^z \rangle$ and the parallel susceptibility $\chi_{zz} = \partial \langle S^z \rangle / \partial h_z$ as we approach the critical end line.

As wing coexistence surfaces are crossed magnetization $\langle S^z \rangle$ changes discontinuously. This jump becomes smaller and smaller as the wing critical end point is approached. A typical set of magnetization curves at a constant temperature (and for different values of transverse field) as functions of parallel field is given in figure 1. For $T = 0.5$ and $D = 0.85$, the critical transverse field is $h_x^c = 0.6494$. For $h_x = 0.62$ a drop in $\langle S^z \rangle$ is observed as the coexistence surface is crossed at $h_z = 0.008$. The magnetization curves for $h_x > h_x^c$ are typical of a paramagnetic regime. The two curves labelled A and B in figure 1 are stability (spinodal) and coexistence (binodal) curves respectively. Figure 2 shows the behaviour of the inverse parallel susceptibility at a

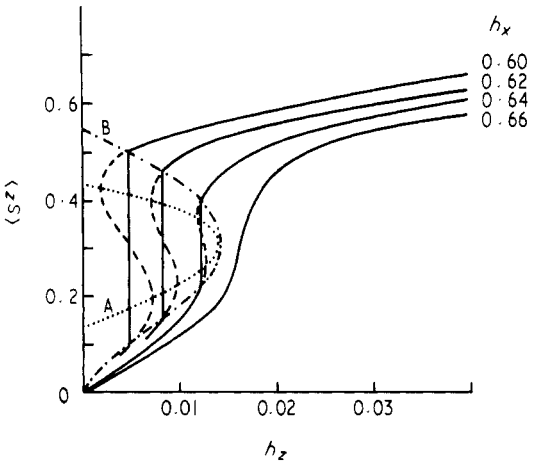

Figure 1. Behaviour of $\langle S^z \rangle$ (full line) as h_z varies at a constant temperature for various values of transverse field. Here $T = 0.5$ and $D = 0.85$. The critical field is $h_x^c = 0.6494$. For $h_x < h_x^c$ a first-order phase transition takes place as h_z increases and hits the wing. The discontinuity decreases as h_x increases and pushes the transition point towards the wing boundary; at $h_x = h_x^c$ the transition is continuous. For $h_x > h_c$ no phase transition is expected. Stability (dotted) and coexistence (chain) curves are denoted by A and B respectively.

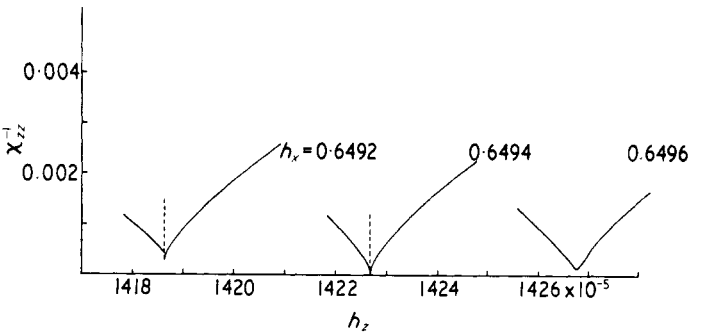

Figure 2. Inverse susceptibility χ_{zz}^{-1} as a function of h_z for various values of h_x. Here $D = 0.85$ and $T = 0.5$. For $h_x = 0.6492$ which is less than the critical value, χ_{zz}^{-1} changes discontinuity as the field h_z crosses the wing. At $h_x = 0.6494$, the critical value, χ_{zz}^{-1} goes to zero as h_z increases and approaches the wing critical end point. A case with h_x above the critical value is also shown ($h_x = 0.6496$); the system is always in the paramagnetic phase but χ_{zz}^{-1} dips still as h_z goes over the wing edge.

constant temperature ($T = 0{\cdot}5$), near a wing critical end point for $D = 0{\cdot}85$. We note that the inverse parallel susceptibility χ_{zz}^{-1} vanishes for $h_x = 0{\cdot}6494$ and $h_z = 0{\cdot}01423$, where the wing critical end point resides.

5. Phase diagrams

The shape of a phase diagram depends on the magnitude of the anisotropy D. For D less than a certain value (figure 3(*a*)) the phase diagrams consist of a single critical line for all ranges of temperature below a certain $T_c(h_x = 0)$. In the notation of Chang *et al* (1973) this line is designated as ${}^2\mathrm{R}_1$; it is the boundary of a coexistence surface (CXS) in the (h_x, T) plane. For slightly larger values of anisotropy D, only a part of the boundary of CXS at $h_z = 0$ is a critical line. The critical line bifurcates at a tricritical point (TCP). Beyond the bifurcation point we have a line of first-order phase transitions in the $h_z = 0$ plane (figure 3(*b*)). For even higher values of anisotropy no critical line in the $h_z = 0$ plane will exist. Any phase transition in this plane (figure 3(*c*)) from ordered to disordered phase will take place in a discontinuous manner.

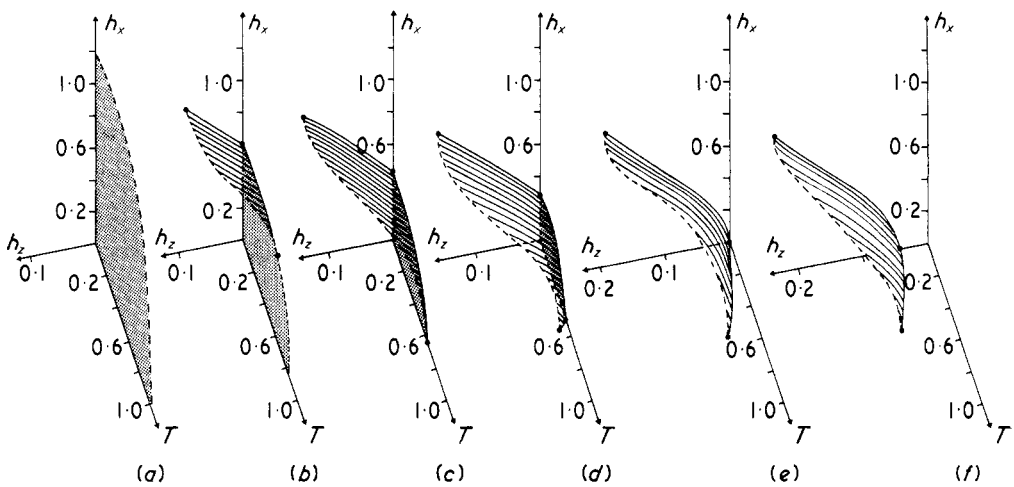

Figure 3. Phase diagrams in the (h_z, T, h_x) space for different D values. Note that only one of the wings is drawn in the figure.

(*a*) For $D \leqslant 0{\cdot}583$, a single critical line bounds the (T, h_x) coexistence plane.

(*b*) For $0{\cdot}583 < D < 0{\cdot}9242$, the critical line in the (T, h_x) plane bifurcates into two critical lines which bound the two coexistence surfaces for $h_z \gtrless 0$, the two wings. The location of the tricritical point, where the three critical lines meet, depends on the value of D.

(*c*) For $D = 0{\cdot}9242$ the tricritical point is on the T axis and the whole line in the (T, h_x) plane is a first-order transition line.

(*d*) For $0{\cdot}9242 < D < 1$, the wing critical lines intersect the (h_z, T) plane at two separate points but the $h_z = 0$ plane is still a coexistence plane.

(*e*) For $D = 1$, the coexistence surface in the plane $h_z = 0$ shrinks to a point at the origin.

(*f*) For $D > 1$, the two coexistence surfaces are completely separated from each other.

The range of values of anisotropy which gives a tricritical point in the phase diagram can be found easily. One limit is found by considering the free energy as $T \to 0$. We obtain (cf § 2) $D_t = 0{\cdot}583$. For $D < D_t$ the phase transition will always be of second

order, and will only occur in the $h_z = 0$ plane (figure 3(*a*)). The other limit is when the bifurcation into the wings happens on the T axis, ie $h_x = h_z = 0$, at a point $T = T_b$ (figure 3(*c*)). Recalling the work of Capel (1966), T_b is the point where the nature of phase transition changes for $h_x = h_z = 0$. The corresponding value of anisotropy D_b is equal to 0·9242. One can also find D_b from equations (3.15*c*, *d*) by putting $h_x = 0$.

For $0{\cdot}583 < D < 0{\cdot}924$, the critical line in the (h_x, T) plane will end in a tricritical point (h_{xt}, T_t) and there it bifurcates into the wing critical lines (figure 3(*b*)). The location of the TCP is a function of anisotropy D. In this interval the phase diagram looks similar to those of metamagnets, ^{3}He–^{4}He mixture and other systems having a TCP. It consists of three two-dimensional CXS, $^2X_2$ (Chang *et al* 1973) meeting in a line of triple points, $^3X_1$, with one of the CXS in the $h_z = 0$ plane. Each of the three CXS is bounded by a one-dimensional CRS of order 2, $^2R_1$. The three CRS meet at the tricritical point, which is also the end point of the line of triple points. At any point on the line of triple points three phases, two ferromagnetic and one paramagnetic, coexist. At TCP the three phases are indistinguishable. A cross section of the wings in the (h_x, h_z) plane at $T = 0{\cdot}5$ is shown in figure 4 for $D = 0{\cdot}85$. We note that the angle between the wings decreases to zero as TCP is approached.

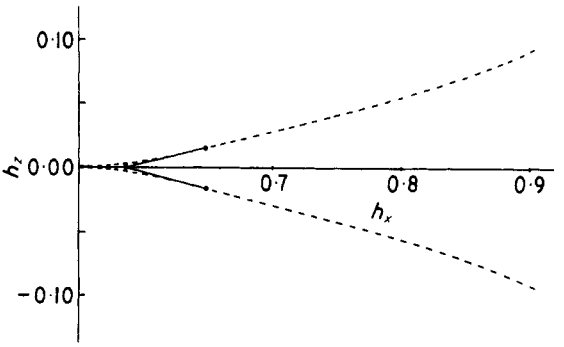

Figure 4. A typical phase diagram in the (h_x, h_z) plane for $D = 0{\cdot}85$. The full line is a wing cross section at $T = 0{\cdot}5$. The dashed curve is the projection of the critical end points on (h_x, h_z) plane.

Figure 3(*c*) shows the situation at $D = 0{\cdot}9242$; the critical line characterized by equation (3.15*c*) shrinks to a point on the T axis (at T_b) where the two wing critical lines meet. The phase transition line in the $h_z = 0$ plane is totally of first order. At this limiting point T_b only two critical lines meet. Although the reflection operation $h_x \to -h_x$ will result in two more critical lines below the $h_x = 0$ plane, each of these lines can be considered as the continuation of the corresponding line above the plane. Thus still three phases become indistinguishable at T_b.

For $0{\cdot}9242 < D < 1$ while the CXS in the $h_z = 0$ plane still persists, the critical lines no longer meet. Figure 3(*d*) shows a typical phase diagram in this range of anisotropy D. The wing critical lines intersect the (h_z, T) plane at two points which are determined by equations (3.29) and (3.30).

At $D = 1$ the CXS in the $h_z = 0$ plane shrinks to a point at the origin where the two wing CXS touch. This is shown in figure 3(*e*). Finally for values of anisotropy $D > 1$ the two CXS are completely separated from each other as shown in figure 3(*f*). The intersection points of wing critical lines with the (h_z, T) plane are again given by equa-

tions (3.29) and (3.30). As $D \to \infty$ the coordinate T of these points tends to a limiting value 0·5, and h_z approaches infinity. Finally it is clear that if we include the anisotropy as a coordinate in the thermodynamic field space we generate a line of tricritical points terminating at T_b.

6. Conclusion

A study of the Blume–Capel model in a transverse field in the mean field approximation (MFA) was performed in this paper. Although MFA is a poor approximation for the study of critical behaviour we expect that the qualitative features of the phase diagram are nevertheless correct. In contrast to the models studied before, while the model does show a TCP, the thermodynamic ordering field is a field available in the laboratory unlike the situation in metamagnets or ^{3}He–^{4}He mixtures. In addition, the disordering field is controllable by the experimenter (unlike the Blume–Capel model where D is the disordering field). Consequently, the whole thermodynamic field space including the wing critical end points and TCP is experimentally available. We think that this feature alone justifies a search for physical systems describable by the model.

Physical systems which can be described by the Blume–Capel model might be found in compounds of Ni^{2+} or V^{3+} (both of $S = 1$). Indeed, α-$NiSO_4.6H_2O$ (Stout and Hadley 1964) and $Ni(NO_3)_2.6H_2O$ (Herweijer and Friedberg 1971) both with a small orthorhombic distortion, have been found to have the non-magnetic state as the ground state ($D > 0$). The exchange interactions are however weak and antiferromagnetic. Furthermore there are complications that each unit cell contains more than one Ni^{++} and that their local z axes are not parallel to each other. Recently $NiSiF_6.6H_2O$ and $NiSnCl_6.6H_2O$ (Meier *et al* 1974) have been studied. These compounds have only one $[Ni(H_2O_6)]^{++}$ complex per unit cell and the departure of the crystal field from octahedral symmetry is purely axial. D is negative for the first compound but can be made positive by applying a hydrostatic pressure. The crystals have rather weak exchange interactions and appear to be antiferromagnetic again. Although none of the above compounds shows the desirable property that fits this model, there is some indication that a suitable compound in the latter family may be found. Perhaps the best candidates that we know of are at present nickel lanthanum double nitrate and the dilute compounds with some of the nickel ions replaced by magnesium ions studied by Mess *et al* (1969). They become ferromagnetically ordered at very low temperatures. The pure compound has $T_c = 0{\cdot}393$ K. D is positive and is of the same order as the exchange coupling parameter. However, there are two different kinds of Ni^{++} ions in the crystal. This complicates the problem somewhat. But the fact that ions of the second kind are rather isolated from their nearest-neighbour magnetic ions may enable one to consider only one kind of ion in a first approximation. It would be of great interest to examine these compounds in the presence of a transverse field.

We should note also that it is not essential for the exchange interaction to be Ising like. As an immediate generalization of the idea presented in this paper, a more general uniaxial ferromagnetic model considered by Wang and Khajehpour (1972) can be analysed in a transverse field in a similar manner. All features described in this paper will be retained, plus many other new features due to the possibility of the existence of additional phases. The more general nature of this model enhances the probability of finding a compound to fit the model exactly. A detailed analysis of this more general model will be given in a later publication.

References

Blume M 1966 *Phys. Rev.* **141** 517
Blume M, Corliss L M, Hastings J M and Sciller E 1974 *Phys. Rev. Lett.* **32** 544
Blume M, Emery V G and Griffith R B 1971 *Phys. Rev.* A **4** 1071
Capel H W 1966 *Physica* **32** 966
Chang T S, Hankey A and Stanley H E 1973 *Phys. Rev.* B **8** 346
Griffith R B 1970 *Phys. Rev. Lett.* **24** 715
Herweijer A and Friedberg S A 1971 *Phys. Rev.* B **4** 4009
Meier D, Kornezos M and Friedberg S A 1974 *Magnetism and Magnetic Materials, Part* 2 (New York: American Institute of Physics) p 1316
Mess K W, Lazendijk E, Zimmermann H J, Van Duyneveldt A J, Giesen J J and Huiskamp W J 1969 *Physica* **43** 165
Stout J W and Hadley W B 1964 *J. Chem. Phys.* **40** 55
Wang Y L and Khajehpour M R H 1972 *Phys. Rev.* B **6** 1778