CRITICAL PROPERTIES OF RANDOM SYSTEMS

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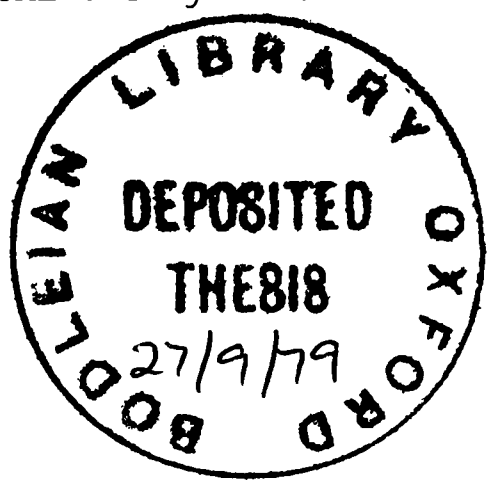

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ABSTRACT

Calculations are presented for a series of interrelated problems in the theory of disordered solids.

The simple mean field theory of tricriticality in the layered Ising metamagnet is modified by inclusion of the Bethe-Peierls equation of state for the planar interactions. The approximation allows for a model of dilution with a finite percolation concentration for the layers. The calculated behaviour as a function of dilution for anisotropic coupling strengths allows comparison with experimental results on dilute ferrous chloride. Estimation is made of the effects of treating fluctuations by theories with mean field singularities.

A discussion is given of first order phase transitions in disordered systems. A mean field theory of the implications of introducing quenched disorder to a system undergoing a transition of first order is effected by reformulating the problem in terms of a translationally invariant one via the replica method. The conclusions are examined in terms of a simple scaling theory and criteria derived for smearing of the singularities.

Dynamic and thermodynamic properties of diluted magnetic insulators near the percolation concentration are considered in terms of simple geometric models of the percolating cluster as introduced by de Gennes. New scaling relations for the spin wave stiffness and conductivity exponents are derived and differences from previous relations interpreted geometrically.

The scaling models of the percolating cluster so found, in which correlations propagate locally via effectively one-dimensional paths, are applied to the determination of the mobility edge for spin wave excitations in a dilute Heisenberg magnet near the percolation threshold. A prediction for the functional form of the mobility edge is made by means of results known for the Anderson model and a localisation length derived from an exact solution of a random problem by Dyson.

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Introduction

There is currently great interest in developing the theory of the solid state to encompass materials that are not perfectly crystalline. While effective medium approximations such as the Coherent Potential Approximation (see Elliott, Krumhansl, Leath 1974) give good agreement with experiment in many cases,there are situations where they are less successful: especially in the prediction of critical singularities and in transport phenomena. On the other hand theories of scaling and the renormalization group that have elucidated critical properties of systems with translational invariance have been applied to random systems with some success (reviewed by Aharony 1978). The drawbacks of such theories is that the quantities most readily determined, such as critical exponents may be difficult to measure experimentally. This thesis concerns aspects of disordered systems that are not reliably predicted by meanfield-like theories but are difficult to treat by the more formal methods of the renormalization group.

The first chapter presents a calculation of the phase diagram for a dilute Ising metamagnet with anisotropy in inter- and intra-planar coupling strengths. The simplest mean field theory of tricriticality is modified by including planar fluctuations within a Bethe-Peierls approximation while conserving the simple mean field theory for inter-planar correlations. A desirable feature of the Bethe-Peierls approximation is that it provides a sensible model for dilution with percolative aspects. The results of the calculation are compared to qualitative features indicated by experiments in diluted crystals of ferrous chloride.

Quantitative discrepancies are related to the mean-field nature of the approximations employed.

One question about the phase diagram of the dilute metamagnet that cannot be answered in terms of the theory presented in Chapter 1 is the effect of random fluctuations on the singular behaviour on the line of first order phase transitions. This problem is taken up in Chapter 2 with a general discussion of the effects of introducing disorder to a system with a first order phase transition. The replica method is used to circumvent the loss of translational symmetry and a mean field approach applied to an equivalent translationally invariant problem. The validity of the conclusions is examined by construction of a simple scaling theory for the effects of disorder and criteria for smearing of the transition derived.

The most striking effects of dilution on phase diagrams such as that calculated in Chapter 1 are those caused by the qualitative change in connectivity at percolation concentrations. The percolation problem has been intensively studied by the methods of modern critical phenomena and the renormalization group. Random dilution of magnetic crystals provides experimental tests of percolation theories. Many of the experimentally accessible variables, however, such as correlation lengths and excitation spectra, cannot easily be calculated by formal renormalization group techniques. It is valuable therefore to develop scaling models of percolation consistent with known critical singularities yet simple enough to allow predictions of measurable quantities. The third chapter is devoted to such models with application primarily to dynamic and thermodynamic properties of dilute Heisenberg magnets near the percolation threshold.

In Chapter 4 the same approach is made to the behaviour of the mobility edge in a dilute Heisenberg magnet near percolation. Conclusions from this work have been published (Ziman and Elliott 1978). They illustrate the capacity of simple scaling models to make definite predictions of properties that are both difficult to treat by the more rigorous renormalization group methods and unsatisfactorily treated by present effective medium treatments.

CHAPTER 1

TRICRITICAL POINTS IN DILUTE METAMAGNETS

1.1 Tricritical Points.

In a system with a critical point there may be a parameter which when varied produces a line of critical points or "critical line". Griffiths (1970, 1973) defined a "tricritical point" as a point in parameter space where three critical lines intersect. Tricritical behaviour is thought to occur in layered metamagnets, where the parameter space includes uniform magnetic field H, staggered magnetic field H_s and temperature T; and in He^3 - He^4 mixtures with parameters the field H conjugate to the superfluid order parameter, the difference $\mu_3 - \mu_4$ in chemical potential of the two species and temperature T. Tricritical properties are reviewed by Stryjewski and Giordano (1977) for metamagnets and Kincaid and Cohen (1975) for helium mixtures and metamagnets. In neither of these systems can the whole phase diagram be explored experimentally - in the space of experimentally accessible parameters the tricritical point is identified as the point of intersection of a line of second order phase transitions and a line of first order phase transitions. For the metamagnet the experimentally accessible space is the plane $H_s = 0$ in figure 1.1. From now on we consider only the parameters H and T and discuss the phase diagram in terms of the metamagnet with order parameter the difference in sublattice magnetisation.

A simple Landau theory for tricritical points is as follows

(Griffiths 1970). We consider that the Gibbs free energy may be expanded

$$G(T,H,H_s)=a(T,H)s^2+b(T,H)s^4+c(T,H)s^6-H_s s \qquad (1)$$

where s is the value minimising the expression. IF c > 0, b(T,H) = b(H) then for fixed H, b(H) > 0, the free energy shows a second order phase transition when $H_s = 0$, a(T,H) = 0, but for b(H) < 0 minimising G with respect to s for $H_s = 0$ gives a first order transition at $a = b^2/4c$. S_{min}, the value of s minimising the expression (1), changes discontinuously by $(-B/2C)^{\frac{1}{2}}$ as shown in figure 2. Within Landau theory we expect the tricritical point at

$$a(T_t,H_t)=b(T_t,H_t)=0\ ,\ c(T_t,H_t)>0 \qquad (2)$$

At each point on the line of second order phase transitions critical exponents are defined in the usual way (see for example Toulouse and Pfeuty 1975) and within Landau theory take values $\beta = \frac{1}{2}$, $\nu = \frac{1}{2}$, $\delta = 3$ in the usual notation. For temperatures close to the tricritical temperature T_t and at field $H = H_t$, G is expanded in Landau theory:

$$G(T,H_t,H_s)=a'(T-T_t)s^2+cs^6-H_s s \qquad (3)$$

leading to equation of state

$$H_s=2a'(T-T_t)s+6cs^5 \qquad (4)$$

From (4) we have exponents called "tricritical exponents"

$$\beta_t=\frac{1}{4}\ ,\ \gamma_t=1\ ,\ \delta_t=5$$

Even in mean field theory therefore there are new critical exponents associated with the tricritical point.

Renormalisation group techniques have stimulated interest in the critical behaviour at second order phase transitions (Wilson and Kogut 1974, Toulouse and Pfeuty 1975) and the tricritical point (Riedel and Wegner 1972). Nelson and Fisher (1975) studied the Ising metamagnet in detail by ε-expansion techniques. One conclusion of such studies is that the power law singularities of thermodynamic quantities will be as predicted by Landau theory for dimensionalities d greater than the upper critical dimensionality d_u where d_u satisfies Josephson's scaling law

$$d\nu = 2-\alpha \qquad (5)$$

and α, ν take the mean field values. For the second order phase transitions described as above the Gaussian approximation gives $\nu = \frac{1}{2}$ and $\alpha = 0$, implying $d_u = 4$. From the tricritical exponents found above and Rushbrooke's scale relation:

$$\alpha + 2\beta + \gamma = 2 \qquad (6)$$

we find $\alpha_t = \frac{1}{2}$ and substituting this with the Gaussian value $\nu = \frac{1}{2}$ equation (5) is satsfied by $d_u = 3$. Thus in three dimensions, while the critical exponents for the second order phase transitions cannot be as in mean field theory, the tricritical exponents may have mean field values and additionally, since the physical dimensionality is equal to d_u, logarithmic corrections are expected.

The scaling properties of tricritical phenomena are consequently of great interest for the study of the effects of marginal dimensionality and crossover from tricritical exponents to the exponents on the critical line. Complications in the scaling behaviour have recently been discussed by Fisher and Sarbach (1978).

A metamagnet thought to have a tricritical point is anhydrous ferrous chloride $FeCl_2$. Studies of its structural and magnetic properties are discussed by Birgeneau et al. (1972), tricritical behaviour by Dillon, Chen and Guggenheim (1978). At zero field and temperatures less than the Néel temperature $T_N = 23.7^{o}K$ the ground state has alternate planes of ferromagnetically aligned Fe^{++} spins with a weaker coupling between planes causing the moments of the planes to alternate in sign. Application of a uniform field perpendicular to the planes induces a phase transition to the paramagnetic state, with phase diagram qualitatively as in figure 1.1. Experiment and calculations of crystal fields and spin orbit coupling agree that there are three low-lying ^5D states of the Fe^{2+} ion with the lowest two degenerate (Alben 1969). Birgeneau et al. (1972) found that the magnon spectrum at low temperatures gives a good fit to a two-dimensional s = 1 Heisenberg Hamiltonian with large anisotropy in the single ion term and exchange. A fit to the planar Hamiltonian is (Birgeneau et al. 1972, Rastelli, Tassi and Reatto 1974)

$$\mathcal{H} = -2J_1 \sum_{NN} \underset{\sim}{S_i} \cdot \underset{\sim}{S_j} - 2J_2 \sum_{NNN} \underset{\sim}{S_i} \cdot \underset{\sim}{S_j} - D \sum_i (S_i^z)^2 - 2K_a \sum_{NN} S_i^z S_j^z \qquad (7)$$

with $2J_1 = 7.9$ K

$2J_2 = -1.04$ K

$D = 12.5$ K

$2K_a = 2.1$K and summation over nearest and next nearest neighbours of the planar triangular lattice. Fit to the transition field at low temperatures gives an antiferromagnetic interplanar exchange

$$2J_1' = 0.4 \text{ K}$$

From the structure each ion interacts with 6 others via J_1'.

A simple model Hamiltonian for such a layered metamagnet suitable for studying qualitative aspects of the phase diagram is an Ising model with lattice anisotropy:

$$\mathcal{H} = -J_1 \sum_{<ij>} S_i S_j + J_1' \sum_{<ik>'} S_i S_k - H \sum_i S_i \qquad (8)$$

$$J_1'/J_1 \sim 1/20 \qquad S_i = \pm 1$$

For each i the summation <ij> is over the six nearest neighbours, <ik>' six nearest neighbours in adjacent planes. The Hamiltonian (7) is simplified to (8) by including only the two lowest spin states, the strongest (nearest neighbour)intraplanar coupling and the interplanar coupling responsible for the metamagnetic transition. This is clearly not very accurate but one would expect it to fall in the same universality class as (7) and therefore show the same scaling properties. Comparison of the phase diagrams of the experimental and calculated systems should be made with the differences borne in mind.

The thermodynamics of Ising systems of the form (8) have been studied by series expansions (Harbus and Stanley 1973), computer simulation (Arora and Landau 1973) and mean field theory (reviewed by Kincaid and Cohen 1975). All show, for appropriate J_1 and J_1',the phase diagram of figure 1.1 but there are numerical discrepancies to be discussed later.

1.2 Effects of Dilution.

The tricritical properties of a metamagnet such as ferrous chloride may be understood, at least qualitatively, in terms of a simple model

Ising Hamiltonian. We now consider the effects of disorder on tricriticality In three dimensions we have seen that the tricritical exponent α_t is strictly positive, $\alpha_t = \frac{1}{2}$. Therefore, from the Harris criterion, (Harris 1974) disorder should strongly affect the tricritical properties. Bergman, Aharony and Imry 1978 have argued that the tricritical point in dirty metamagnets is the most favourable experimental situation for studying critical effects in the presence of disorder since α_t is known to be relatively large and positive in three dimensions.

A simple form of quenched disorder, for both experiment and theory, may be introduced by site dilution of the magnetic ions. Recently, solid solutions $Fe_pMg_{1-p}Cl_2$ with iron ions randomly replaced by non-magnetic magnesium ions have been grown (Garton and Walker 1976) and an optical study of the tricritical behaviour initiated (Wood and Day 1977) There have been studies of $Fe_pCd_{1-p}Cl_2$ by Egbert et al. (1978). The first step in the study of dilution effects is to determine the phase diagram as a function of dilution. Wood and Day measured the tricritical temperature and field with varying dilution

We consider then a system with the model Hamiltonian (8) on a randomly site-diluted lattice. The pure crystal has phase diagram as shown in figure 1.1. As p decreases from 1 T_N decreases and vanishes at the percolation probability $p_c(3d)$ for the three-dimensional lattice, as for an Ising ferromagnet with lattice anisotropy. This is apparent as in zero field the metamagnetic Hamiltonian (8) is mapped onto a ferromagnetic Hamiltonian by the transformation $S_i \rightarrow -S_i$ in every alternate layer. The critical field H_c at zero temperature is determined by competition between a ferromagnetically ordered ground state and the layered antiferromagnetic ground state. By summing over the energy per

spin on each plane this is close to $pH_c(p=1)$ and does not vanish at the three-dimensional percolation probability. It is clear that the phase diagram cannot shift uniformly towards the origin H = T = 0 as in the simplest mean field dilution:

$$J_1 \longmapsto p J_1$$
$$J_1' \longmapsto p J_1' \qquad (9)$$

Furthermore one might expect changes in the thermodynamic properties in the vicinity of the percolation probability $p_c(2d)$ of the planes, since below that concentration long range order is sustained via the weak interplanar coupling. The Néel temperature should therefore decrease rapidly but smoothly near the two-dimensional percolation concentration. A more quantitative discussion follows.

We wish to calculate the phase diagram and in particular the coordinates $(H_t(p), T_t(p))$ of the tricritical point as a function of dilution p. We shall use the simplest approximation expected to give features of an exact solution that are absent in the simplest mean field treatment. For comparison with the metamagnet $FeCl_2$ we treat the Hamiltonian (8) with coupling anisotropy $J_1'/J_1 \sim \frac{1}{20}$. It is appropriate to treat the interlayer coupling as a perturbation sufficiently weak to be included within a mean field approximation, at least for temperatures T satisfying T >> A. As the Néel temperature is of order the strong coupling J_1 this should be valid for all but the lowest temperature region of the phase diagram, at least until the reduced connectivity in the diluted plane depresses T_N well below the mean field temperature $6J_1$.

In this approximation the Helmholtz potential A^c of the metamagnet is written as a function of the magnetisations of each sublattice

$$A^c(m_1, m_2, T) = A^{2d}(m_1, T) + A^{2d}(m_2, T) + A m_1 m_2 \tag{10}$$

where A^{2d} is the Helmholtz potential for a two-dimensional plane. $A = Z_{int} J_1'$ and Z_{int} is the number of nearest neighbours in adjacent planes. The equations of state for external fields H_1 and H_2 on each sublattice are

$$H_1 = \frac{\partial A^c(m_1, m_2, T)}{\partial m_1} = \frac{\partial A^{2d}}{\partial m_1}(m_1, T) + A m_2 \tag{11}$$

$$H_2 = \frac{\partial A^c(m_1, m_2, T)}{\partial m_2} = \frac{\partial A^{2d}}{\partial m_2}(m_2, T) + A m_1$$

For uniform applied field $H = H_1 = H_2$ we deduce coupled equations of state

$$\begin{aligned} m_1 &= m(H - Am_2) \\ m_2 &= m(H - Am_1) \end{aligned} \tag{12}$$

where $h \to m(h)$ is the equation of state for the two-dimensional system in a field.

The problem then reduces to the solution of two coupled equations involving the equation of state for a two-dimensional Ising model in a field. This equation of state can be written explicitly within a mean field approximation, but this is insufficient to reproduce essential effects of reduced connectivity for strong dilution.

The approximate equation of state employed will be that obtained within the Bethe-Peierls approximation (Bethe 1935, Peierls 1936). This at least gives a sensible model for percolation: the critical temperature is given by (See Appendix A, A4)

$$\tanh \frac{J}{kT_c} = \frac{1}{z-1} \tag{13}$$

where z is the coordination number. For the dilute case substitution of an effective coordination number for the connected cluster

$$z(p) = 1 + p(z - 1) \tag{14}$$

predicts a critical temperature vanishing at $p_c = \frac{1}{z-1}$ as

$$kT_c = \frac{2J}{\ln\left(\frac{2}{(p-p_c)(z-1)}\right)} \tag{15}$$

(15) has the correct asymptotic dependence on $(p - p_c)$. (see Chapter 3) and is also exact for the dilute Bethe lattice (Matsubara 1974).

The procedure is to solve equations (12) with a Bethe-Peierls approximation for the planar equation of state, and to introduce dilution effects by varying the coordination of the plane z(p) and interplanar coupling A(p) as

$$z(p) = 1 + p(z-1) \tag{16}$$

$$A(p) = pA \tag{17}$$

(17) follows from the zero temperature value of the critical field.

1.3 Simple Mean Field Theory of Tricriticality in the Metamagnet.

To illustrate the means of locating the critical line and tricritical point we consider first the solution when the mean field equation of state is substituted into equations (12). This follows Kincaid and Cohen (1975). From equation 1 we expect within Landau theory that the Gibbs free energy can be expanded in powers of the order parameter

$$G(T,H) = a(T,H)s^2 + b(T,H)s^4 + c(T,H)s^6 \qquad (18)$$

and s is determined by the minimum of (1) or

$$\frac{\partial G(T,H)}{\partial s} = 0 \qquad (19)$$

$$s(2a(T,H) + 4b(T,H)s^2 + 6c(T,H)s^4) = 0 \qquad (20)$$

(20) has a paramagnetic solution s = 0 and antiferromagnetic solutions

$$2a(T,H) + 4b(T,H)s^2 + 6c(T,H)s^4 = 0 \qquad (21)$$

The critical line is defined by $a(T_\lambda,H_\lambda) = 0$, $b(T_\lambda,H_\lambda) > 0$ and the tricritical point

$$a(T_t,H_t) = b(T_t,H_t) = 0 \qquad (22)$$

(21) and (22) imply that the tricritical point can be found from the expansion of the equation of state for the antiferromagnetic state about the paramagnetic state s=0.

Consider the mean field equation of state substituted in (12):

$$m_1 = \tanh[\beta(H - Am_2) + \beta J m_1]$$
$$m_2 = \tanh[\beta(H - Am_1) + \beta J m_2] \qquad (23)$$

with $J = zJ_1$.

Transforming to variables $r = \frac{m_1+m_2}{2}$ $s = \frac{m_1-m_2}{2}$ and finding the sum and difference of the two equations,

$$\tanh^{-1}(r+s) - \tanh^{-1}(r-s) = (2\beta J + 2\beta A)s$$
$$\tanh^{-1}(r+s) + \tanh^{-1}(r-s) = 2\beta H + 2\beta(J-A)r \qquad (24)$$

In the vicinity of the critical line we expand (24) in powers of s:

$$\left(\frac{2}{1-r^2}\right)s + \left[\frac{8}{(1-r^2)^3} - \frac{6}{(1-r^2)^2}\right]\frac{s^3}{3} + \left[\frac{5}{(1-r^2)^3} - \frac{20}{(1-r^2)^4} + \frac{16}{(1-r^2)^5}\right]\frac{2s^5}{5}$$

$$= 2\beta(J+A)s$$

$$2\tanh^{-1}r + \frac{2r}{(1-r^2)^2}s^2 + 2\left[\frac{2}{(1-r^2)^4} - \frac{1}{(1-r^2)^3}\right]rs^4$$

$$+2\left[\frac{3}{(1-r^2)^4} - \frac{16}{(1-r^2)^5} + \frac{16}{(1-r^2)^6}\right]\frac{rs^6}{3} = 2\beta H + 2\beta(J-A)r \qquad (25)$$

There are then two equations for s and r

$$\left[2\beta(J+A) - \frac{2}{1-r^2}\right]s - \left[\frac{8}{(1-r^2)^3} - \frac{6}{(1-r^2)^2}\right]\frac{s^3}{3}$$

$$- \left[\frac{5}{(1-r^2)^3} - \frac{20}{(1-r^2)^4} + \frac{16}{(1-r^2)^5}\right]\frac{2s^5}{5} = 0 \qquad (26)$$

$$\left[\frac{2r}{(1-r^2)^2}\right]s^2 + 2\left[\frac{2}{(1-r^2)^4} - \frac{1}{(1-r^2)^3}\right]rs^4 + 2\left[\frac{3}{(1-r^2)^4} - \frac{16}{(1-r^2)^5} + \frac{16}{(1-r^2)^6}\right]\frac{rs^6}{3}$$

$$= 2\beta H + [2(\beta J - \beta A)r - 2\tanh^{-1}r \qquad (27)$$

The paramagnetic solution s = 0, which clearly satisfies (26), has paramagnetic equation of state from (27)

$$\beta H - [\tanh^{-1} r - (\beta J - \beta A) r] = 0 \qquad (28)$$

The function $f(r) = \tanh^{-1} r - (\beta J - \beta A) r$ has derivative satisfying, for $\beta J \leq 1$

$$f'(r) = \left[\frac{1}{1-r^2} - (\beta J - \beta A) \right] \geq [1 - (1 - \beta A)] = \beta A > 0$$

Above the two-dimensional ordering temperature f is monotonic, therefore (28) defines a unique paramagnetic solution

$$r = r_{para}(H,T)$$

(28) may be differentiated with respect to H to give

$$\beta = \left[\frac{1}{1-r^2} - (\beta J - \beta A) \right] \left(\frac{\partial r_{para}}{\partial H} \right)_T \qquad (29)$$

On the critical line the paramagnetic solution becomes unstable and the antiferromagnetic phase stable. From (26) the antiferromagnetic solution $s \neq 0$ has expansion

$$\left[2\beta(J+A) - \frac{2}{1-r^2} \right] - \left[\frac{8}{(1-r^2)^2} - \frac{6}{(1-r^2)^2} \right] \frac{s^2}{3}$$
$$- \left[\frac{5}{(1-r^2)^3} - \frac{20}{(1-r^2)^4} + \frac{16}{(1-r^2)^5} \right] \frac{2s^4}{5} + \ldots = 0 \qquad (30)$$

On the critical line, where the transition is continuous, as $s \to 0$ $r \to r_{para}$; therefore the first bracket is proportional to a(T,H) as defined

in (21). The critical line is therefore defined by

$$\frac{1}{1-r_\lambda^2} = \beta_\lambda (J+A) \tag{31}$$

and (28). (31) can be rewritten

$$r_\lambda = \left(1 - \frac{1}{\beta_\lambda (J+A)}\right)^{1/2} \tag{32}$$

Eliminating r from (28), the critical line has equation

$$\beta H_\lambda = \tanh^{-1}\left(1 - \frac{1}{\beta_\lambda (J+A)}\right)^{1/2} - \beta_\lambda (J-A)\left(1 - \frac{1}{\beta_\lambda (J+A)}\right)^{1/2} \tag{33}$$

The Néel temperature ($H_\lambda = 0$, T_N) is given, as expected, by

$$\beta_N (J+A) = 1 \quad ; \quad kT_N = J+A \tag{34}$$

The critical line is also determined, putting (31) into (29) as

$$\left(\frac{\partial r_{para}}{\partial H}\right)_T = \frac{1}{2A} \tag{35}$$

The tricritical point is now determined by expanding the antiferromagnetic equations (30),(27) in powers of s around the critical point ($H = H_\lambda$, $r = r_\lambda$, $s = 0$) keeping at constant temperature, to determine the sign of the coefficient b of (21). From (27) s^2 is of order $(r - r_\lambda)$; we therefore keep terms of this order.

Expansion of (27) gives

$$\left.\frac{2r_\lambda}{(1-r_\lambda^2)^2}+\dots\right)s^2+2\left[\frac{1}{(1-r_\lambda^2)^4}-\frac{1}{(1-r_\lambda^2)^2}\right][r_\lambda+(r-r_\lambda)]s^4+\dots$$

$$=2\beta(H-H_\lambda)-\left[\frac{2}{(1-r_\lambda^2)}-2(\beta J-\beta A)\right](r-r_\lambda)$$

(36)

or to order s^2

$$\frac{2r_\lambda}{(1-r_\lambda^2)^2}s^2=2\beta(H-H_\lambda)-2\left[\frac{1}{(1-r_\lambda^2)}-(\beta J-\beta A)\right](r-r_\lambda)$$

(37)

and

$$\left[2\beta(J+A)-\frac{2}{1-r_\lambda^2}-\frac{4r_\lambda}{(1-r_\lambda^2)^2}(r-r_\lambda)\right]-\left[\frac{8}{(1-r_\lambda^2)^3}-\frac{6}{(1-r_\lambda^2)^2}\right]\frac{s^2}{3}=0$$

(38)

On the critical line $\frac{1}{1-r_\lambda^2}=\beta(J+A)$ and (38) can be written

$$-4r_\lambda(\beta(J+A))^2(r-r_\lambda)-[8(\beta(J+A))^3-6(\beta(J+A))^2]\frac{s^2}{3}=0$$

$$(r-r_\lambda)=-\left(\frac{4\beta(J+A)-3}{6r_\lambda}\right)s^2$$

(39)

Substituting into (37) gives the expression

$$\left\{\frac{r_\lambda}{(1-r_\lambda^2)^2}-\frac{[4\beta(J+A)-3]2\beta A}{6\quad r_\lambda}\right\}s^2+\dots=\beta(H-H_\lambda)$$

(40)

On the critical line we expect a stable antiferromagnetic solution for $H < H_\lambda$, from (40) this implies

$$\tilde{b}(\lambda) = \left\{ \frac{r_\lambda}{(1-r_\lambda^2)^2} - \frac{(4\beta(J+A)-3)2\beta A}{6 \quad r_\lambda} \right\} < 0 \tag{41}$$

For $\tilde{b}(\lambda) > 0$ there is no antiferromagnetic solution with $s \to 0$ as $H \to H_\lambda^-$; the solution with $s \to 0$ as $H \to H_\lambda^+$ corresponds to a relative maximum near the origin (as in figure 1.2c). By comparison with (22) $(\beta, \tilde{b}(\lambda))$ is proportional to

$$\left(2 \frac{\partial a}{\partial H}(T_\lambda, H_\lambda) \quad , -4b(T_\lambda, H_\lambda)\right) .$$

The tricritical point is defined by $\tilde{b}(\lambda) = 0$, or

$$\frac{r_\lambda}{(1-r_\lambda^2)^2} - \frac{(4\beta(J+A)-3)}{6} \frac{2\beta A}{r_\lambda} = 0 \tag{42}$$

Expression (31) for r_λ allows (42) to be rewritten

$$\beta_t(J+A) = \frac{J}{J-A/3} \qquad kT_t = (J+A)\left(1 - \frac{A}{3J}\right) \tag{43}$$

$$T_t/T_N = 1 - \frac{A}{3J} \tag{44}$$

From (32) and (33)

$$\beta_t H_t = \tanh^{-1}\left(\frac{A}{3J}\right)^{1/2} - \beta_t(J-A)\left(\frac{A}{3J}\right)^{1/2} \tag{45}$$

Series expansions and Monte Carlo simulation (Harbus and Stanley 1973, Arora and Landau 1973) indicate that, for the isotropic case at

least (44) predicts a value of T_t/T_N considerably higher than the exact value. For a simple cubic lattice mean field theory (44) gives

$$T_t/T_N = 1 - \frac{2}{3.4} = \frac{5}{6}$$

Whereas $T_t/T_N = 0.58 \pm 0.01$ from series and Monte Carlo studies. Reasons for this will be discussed later.

1.4 Improved Mean Field Theory for Tricriticality.

We now follow the procedure of 1.3 but now including the equation of state from the Bethe-Peierls approximation for the planes, namely (A1,A2)

$$m(h) = \tanh\left[\frac{\beta H_o(h) z - \beta h}{z-1}\right] \qquad (46)$$

Where $H_o(h)$ is defined as the solution of

$$e^{2\beta h} = e^{2\beta H_o}\left(\frac{\cosh(\beta J_1 - \beta H_o)}{\cosh(\beta J_1 + \beta H_o)}\right)^{z-1} \qquad (47)$$

In Appendix A it is demonstrated that for $\beta < \beta_{2d} = \frac{1}{J_1}\tanh^{-1}(\frac{1}{z-1})$ $H_o(h)$ is well defined by (47) and differentiable.

Coupled equations (12) are then

$$m_1 = \tanh\left[\frac{\beta H_o(H - Am_2) z - \beta(H - Am_2)}{z-1}\right] \qquad (48)$$

$$m_2 = \tanh\left[\frac{\beta H_o(H - Am_1) z - \beta(H - Am_1)}{z-1}\right] \qquad (49)$$

or $$\tanh^{-1} m_1 - \tanh^{-1} m_2 = \frac{\beta z}{z-1}\left[H_0(H-Am_2) - H_0(H-Am_1)\right] + \frac{\beta A}{z-1}(m_2-m_1$$

(50)

$$\tanh^{-1} m_1 + \tanh^{-1} m_2 = \frac{\beta z}{z-1}\left[H_0(H-Am_2) + H_0(H-Am_1)\right] - \frac{2\beta H}{z-1} + \frac{\beta A}{z-1}(m_1+m_$$

(51)

defining $m_1 = r+s$, $m_2 = r-s$, we find

$$\tanh^{-1}(r+s) - \tanh^{-1}(r-s) = \frac{\beta z}{z-1}\left[H_0(H-Ar+As) - H_0(H-Ar-As)\right] - \frac{\beta A}{z-1} 2s$$

(52)

$$\tanh^{-1}(r+s) + \tanh^{-1}(r-s) = \frac{\beta z}{z-1}\left[H_0(H-Ar+As) + H_0(H-Ar-As)\right] - \frac{2\beta H}{z-1} + \frac{2\beta Ar}{z-1}$$

(53)

The paramagnetic solution s=0 satisfies (52) trivially and (53) defines r_{para}

$$2\tanh^{-1}(r_{para}) = \frac{\beta z}{z-1} 2H_0(H-Ar_{para}) - \frac{2\beta H}{z-1} + \frac{2\beta A r_{para}}{z-1}$$

(54)

As in 1.3 we locate the critical line as the line of instability of the paramagnetic phase and determine the order of the transition by finding an expansion in powers of s at this line. (52) and (53) expanded in powers of s are

$$\frac{2}{(1-r^2)} s + \left[\frac{8}{(1-r^2)^3} - \frac{6}{(1-r^2)^2}\right]\frac{s^3}{3} + \cdots = \frac{\beta z}{z-1}\left[2As\frac{\partial H_0}{\partial h}(h=H-Ar) + \frac{2(As)^3}{3!}\frac{\partial^3 H_0}{\partial h^3}(h=H-Ar)\right] - \frac{\beta A}{z-1} 2s$$

(55)

and

$$2\tanh^{-1}r + \frac{2r}{(1-r^2)^2}s^2 + 2\left[\frac{2}{(1-r^2)^4} - \frac{1}{(1-r^2)^3}\right]rs^4 + \cdots$$

$$= \frac{\beta z}{z-1}\left[2H_0(H-Ar) + \frac{2(As)^2}{2!}\frac{\partial^2 H_0}{\partial h}(h:H-Ar) + \frac{2(As)^4}{4!}\frac{\partial^4 H_0}{\partial h^4}\right] - \frac{2\beta H}{z-1} + \frac{2\beta Ar}{z-1}$$

(56)

On approaching the critical line from the antiferromagnetic phase the antiferromagnetic solution tends continuously to the paramagnetic solution. At the critical line (55) is satisfied for $s \to 0$, but $s \neq 0$, thus:

$$\frac{2}{1-r_\lambda^2} = \frac{\beta z}{z-1} 2A \frac{\partial H_0}{\partial h}(H_\lambda - Ar_\lambda) - \frac{\beta 2A}{z-1} \qquad (57)$$

or

$$\frac{1}{\beta A} = (1-r_\lambda^2)\left[\frac{z}{z-1}\frac{\partial H_0}{\partial h}(H_\lambda - Ar_\lambda) - \frac{1}{z-1}\right]$$

(58)

Equations (58) and (54) define the critical line. They can be solved numerically to give the critical line and equation of state for the paramagnetic phase $(\beta_\lambda, H_\lambda, r_\lambda)$.

We now expand around the line in powers of s just as in 1.3. Factoring out the paramagnetic solution $s = 0$ from (55) and retaining terms of order s^2, we have

$$\left\{\frac{1}{3}\left[\frac{4}{(1-r^2)^3} - \frac{3}{(1-r^2)^2}\right] - \frac{\beta z}{z-1}\frac{A^3}{3!}\frac{\partial^3 H_0}{\partial h^3}(h:H-Ar)\right\}s^2$$

$$= \frac{\beta z}{z-1}\left[A\frac{\partial H_0}{\partial h}(h=H-Ar)\right] - \frac{\beta A}{z-1} - \frac{1}{(1-r^2)}$$

(59)

As in section 1.3 we work to order $(r - r_\lambda)$, s^2, $(H - H_\lambda)$. The right hand side of (59) is zero at the critical line, and to the required order is

$$\frac{\beta z}{z-1} A \frac{\partial H_0}{\partial h}(H - Ar) - \frac{\beta A}{z-1} - \frac{1}{(1-r^2)}$$

$$= \left[-\frac{\beta z}{z-1} A^2 \frac{\partial^2 H_0}{\partial h}(h_\lambda) - \frac{2r_\lambda}{(1-r_\lambda^2)^2} \right](r - r_\lambda) + \frac{\beta z}{z-1} A \frac{\partial^2 H_0}{\partial h^2}(h_\lambda)(H - H_\lambda$$

(60)

where $h_\lambda = H_\lambda - Ar_\lambda$.

To this order the bracket in the left hand side of (59) is evaluated at (r_λ, H_λ) and (59) becomes

$$\left\{ \frac{1}{3}\left[\frac{4}{(1-r_\lambda^2)^3} - \frac{3}{(1-r_\lambda^2)^2} \right] - \frac{\beta z}{z-1} \frac{A^3}{3!} \frac{\partial^3 H_0}{\partial h^3}(h_\lambda) \right\} s^2$$

$$= \frac{\beta z}{z-1} A \frac{\partial^2 H_0}{\partial h^2}(h_\lambda)(H - H_\lambda) - \left[\frac{\beta z}{z-1} A^2 \frac{\partial^2 H_0}{\partial h^2}(h_\lambda) + \frac{2r_\lambda}{(1-r_\lambda^2)^2} \right](r - r_\lambda)$$

(61)

(61) is then used to eliminate $(r - r_\lambda)$ in (56) to obtain an expansion in powers of s^2. (56) is first written

$$B_o + B_1 s^2 + B_2 s^4 + \ldots\ldots\ldots = 0 \qquad (62)$$

with

$$B_0(r,H) = 2\tanh^{-1} r - \frac{2\beta z}{z-1} H_0(H-Ar) + \frac{2\beta H}{z-1} - \frac{2\beta A r}{z-1}$$

$$B_1(r,H) = \frac{2r}{(1-r^2)^2} + \frac{2\beta z}{z-1}\frac{A^2}{2!}\frac{\partial^2 H_0}{\partial h^2} \quad (h = H-Ar)$$

To order $(r - r_\lambda)$ and $(H - H_\lambda)$

$$B_0(r,H) = B_0(r_\lambda,H_\lambda) + (r-r_\lambda)\frac{\partial B_0}{\partial r}(r_\lambda,H_\lambda) + (H-H_\lambda)\frac{\partial B_0}{\partial H_\lambda}(r_\lambda,H_\lambda) + \ldots \tag{63}$$

$B_0(r_\lambda,H_\lambda) = 0$ from (54), differentiating B_0 we find

$$B_0(r,H) = (r-r_\lambda)\left\{\frac{2}{1-r_\lambda^2} + \frac{2\beta z}{z-1} A \frac{\partial H_0}{\partial h}(h_\lambda) - \frac{2\beta A}{z-1}\right\} + (H-H_\lambda)\left\{-\frac{2\beta z}{z-1}\frac{\partial H_0}{\partial h}(h_\lambda) + \frac{2\beta}{z-1}\right\} \tag{64}$$

To order s^2 (62) is, from (64) and (61)

$$\left[\frac{2}{1-r_\lambda^2} + \frac{2\beta z}{z-1} A \frac{\partial H_0}{\partial h}(h_\lambda) - \frac{2\beta A}{z-1}\right] R(H-H_\lambda, s^2) + \left[-\frac{2\beta z}{z-1}\frac{\partial H_0}{\partial h}(h_\lambda) + \frac{2\beta}{z-1}\right](H-H_\lambda) + \left[\frac{2r_\lambda}{(1-r_\lambda^2)^2} + \frac{2\beta z}{z-1}\frac{A^2}{2!}\frac{\partial^2 H_0}{\partial h^2}(h_\lambda)\right] s^2 = 0$$

$$\mathcal{R}(H-H_\lambda, s^2) = \frac{\left[\left\{\frac{1}{3}\left(\frac{4}{(1-r_\lambda^2)^3} - \frac{3}{(1-r_\lambda^2)^2}\right) - \frac{\beta z}{z-1}\frac{A^3}{3!}\frac{\partial^3 H_0}{\partial h^3}\right\} s^2 - \left\{\frac{\beta z}{z-1} A \frac{\partial^2 H_0}{\partial h^2}\right\}(H-H_\lambda)\right]}{-\left[\frac{\beta z}{z-1} A^2 \frac{\partial^2 H_0}{\partial h^2}(h_\lambda) + \frac{2r_\lambda}{(1-r_\lambda^2)^2}\right]}$$

(65)

or $A(0)(H - H_\lambda) + A(1)s^2 + \ldots = 0$ (66)

where

$$A(0) = \left[-\frac{2\beta z}{z-1}\frac{\partial H_0}{\partial h}(h_\lambda) + \frac{2\beta}{z-1}\right] + \frac{\left[\frac{2}{1-r_\lambda^2} + \frac{2\beta z}{z-1} A \frac{\partial H_0}{\partial h}(h_\lambda) - \frac{2\beta A}{z-1}\right]\frac{\beta z A}{z-1}\frac{\partial^2 H_0}{\partial h^2}(h_\lambda)}{\left[\frac{\beta z}{z-1} A^2 \frac{\partial^2 H_0}{\partial h^2}(h_\lambda) + \frac{2r_\lambda}{(1-r_\lambda^2)^2}\right]}$$

(67)

$$A(1) = \left[\frac{2r_\lambda}{(1-r_\lambda^2)^2} + \frac{2\beta z}{z-1}\frac{A^2}{2!}\frac{\partial^2 H_0}{\partial h^2}(h_\lambda)\right] - \frac{\left[\frac{2}{(1-r_\lambda^2)} + \frac{2\beta z}{z-1} A \frac{\partial H_0}{\partial h}(h_\lambda) - \frac{2\beta A}{z-1}\right]\left\{\frac{1}{3}\left[\frac{4}{(1-r_\lambda^2)^3} - \frac{3}{(1-r_\lambda^2)^2}\right] - \frac{\beta z}{z-1}\frac{A^3}{3!}\frac{\partial^3 H_0}{\partial h^3}\right.}{\left[\frac{\beta z}{z-1} A^2 \frac{\partial^2 H_0}{\partial h^2}(h_\lambda) + \frac{2r_\lambda}{(1-r_\lambda^2)^2}\right]}$$

(68)

As in 1.3 the condition that the critical line represent a line of second order phase transitions is $A(1)/A(0) > 0$

and the tricritical point is given by A(1) = 0.

The numerical procedure was then to solve (58) and (54) at a series of points starting at (T_N, H=0) and find the constants A(0) and A(1) at each point along the line, using (67) and (68) and the derivatives of H_o as found in Appendix A.

This procedure was followed for the metamagnet in a simple cubic lattice with isotropy with respect to bond strengths in order to compare with the results of series and computer simulation of Harbus and Stanley (1973) and Arora and Landau (1973). Results are summarised in table 1.

Table 1. Simple Cubic Lattice $J_1 = J_1' = 100$.

	T_N	T_t	T_{2d}	T_t/T_N	$H_t/H_c(T=0)$
Series*	451	260	226	.58	.95
Theory of 1.4	535	369	289	.68	.90
Simple mean 1.3 field theory	600	500	400	.83	.63

*Series results for (T_t, H_t) for Harbus and Stanley (1973) T_N and T_{2d} from Domb (1974) T_{2d} is the exact value from Onsager's solution.

It is apparent that even in the isotropic case, where an interplanar mean field approximation is questionable, inclusiòn of a Bethe Peierls approximation improves agreement with series values considerably. This is partly due to the more accurate value of T_{2d} found by the Bethe Peierls method.

The critical line and tricritical point were calculated for parameters $A/J = \frac{6x1}{20}$, $\underline{z} = 6$ appropriate for comparison to the metamagnet $FeCl_2$.

The critical line for $J_1 = 100$ A = 30 = $H_c(T = 0)$ is shown in figure 1.3. The Néel temperature and tricritical points have coordinates $T_N = 530$, $T_t = 525$, $H_t = 6.6$. Mean field results for the same parameters are $T_N = 630$, $T_t = 619.5$, $H_t = 4.5$. The ratio $T_t/T_N = 1-0.010$, greater than the mean field value $T_t/T_N = 1-0.017$. Both values are considerably greater than the experimental value for $FeCl_2$ quoted by Dillon, Chen and Guggenheim (1978): $(T_t/T_N) = 0.91$. The same magneto-optical studies of the phase diagram of $FeCl_2$ gave a tricritical field $H_t \simeq 7.2$ kOe and $H_t/H_c(T = 0) \simeq 7.2/10.8 = 0.67$ much greater than the values reported above. Of course it is not possible to say how much of the discrepancies can be attributed to the simplifications of the model Hamiltonian (8) and how much to approximation in the solution of its tricritical properties. At temperatures close to the tricritical temperature $\sim$ 21 K the anisotropy D = 12.5 K of the Hamiltonian (7) is insufficient to prevent excitation to the higher $s_i^z = 0$ state in the ground state triplet of the ferrous ion. The spin $\frac{1}{2}$ Hamiltonian (8) does not include such effects.

It will be argued later, however, that a major part of the discrepancies outlined above results from the approximations employed in finding the tricritical point. Whereas in the isotropic case the Bethe-Peierls approximation for the planes gave reasonable agreement with series values, in the limit of weak interlayer coupling the critical line lies close to the two-dimensional ordering temperature. The Bethe-Peierls approximation

and simple mean field theory show the same (incorrect) singularities near the critical temperature. In section 1.6 the effects of critical fluctuations in the plane will be discussed in more detail.

1.5 The Tricritical Point in the Dilute System.

In section 1.4 we developed a treatment of the tricritical point in a system with model Hamiltonian (8). We now consider the Hamiltonian (8) for occupied sites on a randomly diluted lattice, where sites are occupied with probability p. We saw in 1.2 that effects of reduced coordination in the plane may be modelled by the Bethe-Peierls equation of state with $z(p) = 1 + p(z-1)$ and interplanar couplings $A(p) = pA$ as in (16) and (17). With these parameters the critical line was found for a range of values of p between 1 and the percolation concentration of the Bethe lattice $(1/(z-1))$. The variations of the tricritical point $(T_t(p), H_t(p))$ and Néel temperature $T_N(p)$ as a function of p are shown in figures 1.4 and 1.5. It is seen that the tricritical point initially falls linearly with p but then rises and apparently converges to the point $(T = 0, H = p_c A)$ on the $T = 0$ axis. The approximation indicates that as dilution approaches the two-dimensional percolation threshold the phase diagram is qualitatively as in figure 1.6, i.e. the line of first order transitions collapses to temperatures low compared to T_N which stays finite at p_c, as one would expect for an exact solution as T_N vanishes only at the three-dimensional percolation probability.

The position of the tricritical point in $Fe_pMg_{1-p}Cl_2$ has been measured for various concentrations p from 1 to 0.7 by Wood and Day (1977)

who found behaviour as in figure 1.7. Extrapolated to lower concentrations, the data were taken to indicate a tricritical temperature vanishing at $p = .6$, close to the planar percolation probability $p = 0.59$ rather than that for the three-dimensional lattice. The tricritical field appeared to level off to a value only 30% reduced from that of pure $FeCl_2$. This is in accord with the theoretical conclusion presented above, that the phase diagram at the two-dimensional percolation concentration is as in figure 1.6 with a tricritical temperature close to zero and the tricritical field near the zero temperature value. The increase in tricritical field at intermediate concentrations found in the theoretical calculations is presumably a result of a value of the tricritical field H_t that is too low in the pure case. As the zero temperature value p_cA is fixed, $H_t(p)$ must increase in some range if it falls below this value.

1.6 Critical Fluctuations in the Plane.

It is desirable to improve the theory of the tricritical point presented in 1.4, in particular to estimate the effects of fluctuations on the position of the tricritical point in (H,T) space. As the critical line is at temperatures close to the two-dimensional ordering temperature it may be adequate to treat the interplanar coupling by mean field theory, at least when the two-dimensional critical temperature is much greater than the interplanar coupling. The most important failure in the theory of 1.4 therefore is the failure of the Bethe-Peierls approximation to reproduce an equation of state for the Ising plane of the correct analytic form. In this section we try to judge what effects inclusion of a correct equation of state would have.

As before the coupled equations are

$$m_1 = m(H - Am_2)$$
$$m_2 = m(H - Am_1) \qquad (69)$$

where

$$m: h \rightarrow m(h) \qquad (70)$$

is the equation of state for the two-dimensional Ising model in a field. With definitions $r = \frac{m_1+m_2}{2}$, $s = \frac{m_1-m_2}{2}$, (69) can be rewritten

$$\beta \bar{m}^1(r+s) - \beta \bar{m}^1(r-s) = 2\beta As \qquad (71)$$

$$\beta \bar{m}^1(r+s) + \beta \bar{m}^1(r-s) = 2\beta(H - Ar) \qquad (72)$$

where $m^{-1}: x \rightarrow m^{-1}(x)$ is the inverse function of (70) for fixed temperature or β. The paramagnetic phase is defined by $s = 0$ or

$$2m^{-1}(r_{para}) = 2H - 2Ar_{para} \qquad (73)$$

On the critical line the paramagnetic solution is unstable with respect to antiferromagnetic ordering by a second order transition; therefore on this line (71) has a solution for $s \rightarrow 0$. Staying at constant temperature, comparing terms of order s in (71) gives

$$2\beta \frac{\partial \bar{m}^1(r)}{\partial r} s = 2\beta As$$

$$\left.\frac{\partial \bar{m}^1(r)}{\partial r}\right|_T = A \qquad (74)$$

Condition (74) can be written in terms of the equation of state for the paramagnetic state by differentiating (73) with respect to r at constant temperature

$$\left(\left.\frac{\partial \bar{m}^1(r)}{\partial r}\right|_T + A \right) = \left(\frac{\partial H}{\partial r_{para}} \right)_T$$

or, with (74),

$$\left(\frac{\partial r_{para}}{\partial H}\right)_T = \frac{1}{2A} \tag{75}$$

The tricritical point is located by expanding in powers of s around the critical line.

Let us define

$$\alpha_i = \beta\left(\frac{\partial^i \bar{m}^{-1}(r)}{\partial r^i}\right)_T \tag{76}$$

Then (71) and (72) can be expanded

$$\alpha_1 s + \frac{\alpha_3}{3!} s^3 + \frac{\alpha_5}{5!} s^5 + \dots = (\beta A) s \tag{77}$$

$$\frac{\alpha_2}{2!} s^2 + \frac{\alpha_4}{4!} s^4 + \dots = \beta H - [\beta \bar{m}^{-1}(r) + \beta A r] \tag{78}$$

On the critical line $H = H_\lambda(\beta)$, $r = r_\lambda(\beta)$ $H - (m^{-1}(r_\lambda) + A r_\lambda) = 0$ from (73); (77) and (78) to order $(r - r_\lambda)$ and s^2 are

$$s\left(\alpha_1 + \frac{\alpha_3}{3!} s^2\right) = (\beta A) s \tag{79}$$

$$\frac{\alpha_2}{2!} s^2 = \beta (H - H_\lambda) - \left\{\beta \frac{\partial}{\partial r}(\bar{m}^{-1}(r) + Ar)\Big|_{r=r_\lambda}\right\}(r - r_\lambda)$$

$$= \beta (H - H_\lambda) - 2\beta A (r - r_\lambda) \tag{80}$$

(79) has paramagnetic solution s = 0 and antiferromagnetic solution

$$(\alpha_1(r) - \beta A) + \frac{\alpha_3}{3!} s^2 = 0 \tag{81}$$

From (74) $\alpha_1(r_\lambda) = \beta A$, therefore to order $(r - r_\lambda)$ (81) is written

$$\alpha_2(r_\lambda)(r-r_\lambda) + \frac{\alpha_3}{3!} s^2 = 0$$

$$(r-r_\lambda) = -\frac{\alpha_3(r_\lambda)}{3!\alpha_2(r_\lambda)} s^2 \qquad (82)$$

Substituting (82) into (80) eliminates $(r - r_\lambda)$

$$\beta(H-H_\lambda) + \left[2\beta A \frac{\alpha_3(r_\lambda)}{3!\alpha_2(r_\lambda)} - \frac{\alpha_2(r_\lambda)}{2}\right] s^2 + \ldots = 0 \qquad (83)$$

The condition that there is a second order phase transition as a function of field at H_λ to an antiferromagnetic state is

$$2\beta A \frac{\alpha_3(r_\lambda)}{3!\alpha_2(r_\lambda)} - \frac{\alpha_2(r_\lambda)}{2} > 0 \qquad (84)$$

and the tricritical point is given by

$$\frac{\alpha_2(r_\lambda)}{2} = \frac{\beta A \alpha_3(r_\lambda)}{3\,\alpha_2(r_\lambda)} \qquad (85)$$

The problem therefore involves the first three derivatives of the equation of state.

The scaling theory for critical phenomena predicts that in the vicinity of the critical point the equation of state can be written (Widom 1965, Brézin, Le Guillou, Zinn-Justin 1976)

$$\beta_{(H)} m^{-1}(r) = r^{-\delta} f\left(\frac{t}{r^{1/\beta}}\right)$$

$$t = \frac{T-T_c}{T_c} \qquad (86)$$

where $f(x)$ is regular at $x = 0$, $f(0) > 0$ $f(-1) = 0$, $f(x) \sim x^\gamma$ as $x \to \infty$ with $\gamma = \beta(\delta-1)$. For the two-dimensional Ising model $\delta = 15$, $\beta = \frac{1}{8}$, $\gamma = \frac{7}{4}$, whereas for mean field theories $\delta = 3$, $\beta = \frac{1}{2}$, $\gamma = 1$.

Unfortunately f(x) is not known in general except within the ε expansion although numerical approximations have been found (reviewed by Domb 1974). At the Néel temperature $r \to 0$ and the asymptotic form of f is applicable. Equation (74) can be written, using (86)

$$\beta_N A = a_1 t^{\gamma} = \beta \chi_{2d}(T)^{-1} \tag{87}$$

as it must, since in the mean field approximation for the interplanar coupling the susceptibility is given by

$$\chi = \frac{\chi_{2d}}{1 - A\chi_{2d}} \tag{88}$$

The Néel temperature, from (87), depends on the strength of the interplanar coupling as

$$T_N = T_c \left(1 + \left(\frac{\beta_N A}{a_1}\right)^{1/\gamma}\right) \tag{89}$$

Since $\gamma = 7/4$ for the two-dimensional Ising model while mean field theories, such as the Bethe-Peierls approximation predict $\gamma = 1$, one effect of using a mean field theory instead of an equation of state of the correct analytic form is to decrease

$$\left(\frac{T_N - T_c}{T_c}\right)_{mf} \sim \left(\frac{A}{k_B T_c}\right) << \left(\frac{A}{k_B T_c}\right)^{4/7} \qquad \begin{matrix} A \to 0 \\ T_N \to T_c \neq 0 \end{matrix} \tag{90}$$

If we consider the next term in the expansion of f for large x, namely that given by Griffiths (1967)

$$f(x) = \sum_{p=1}^{\infty} a_p x^{\gamma - 2(p-1)\beta}$$

$$= a_1 x^{\gamma} + a_2 x^{\gamma - 2\beta} + \ldots \tag{91}$$

then the equation of state (86) is

$$\beta h = a_1 t^{\gamma} r + a_2 t^{\gamma-2\beta} r^3 + \ldots \tag{92}$$

We can then find the expression in (84)

$$2\beta A \frac{\alpha_3}{3!\alpha_2} - \frac{\alpha_2}{2} = \frac{1}{3}\beta A \frac{6a_2 t^{\gamma-2\beta}}{6a_2 t^{\gamma-2\beta} r} - \frac{6a_2 t^{\gamma-2\beta} r}{2}$$

$$= \frac{1}{r}\left[\frac{1}{3}\beta A - 3a_2 t^{\gamma-2\beta} r^2\right] \tag{93}$$

On the critical line we have condition (74)

$$\alpha_1(r) = \beta A \Rightarrow a_1 t^{\gamma} + 3a_2 t^{\gamma-2\beta} r^2 = \beta A \tag{94}$$

(93) can then be written

$$\frac{1}{r}\left[a_1 t^{\gamma} - \frac{2}{3}\beta A\right] \tag{95}$$

To this approximation this would give a tricritical point for $a_1 t^{\gamma} = 2\beta A/3$ which combined with (87) predicts $\frac{t_t}{t_N} = \left(\frac{2}{3}\right)^{1/\gamma}$

If t_t and t_N are of the same order of magnitude as this implies, then from (90)

$$T_t/T_N = \frac{1+t_t}{1+t_N} \simeq 1 - (t_N - t_t)$$

$$1 - T_t/T_N \sim (\beta_2 A)^{1/\gamma} \tag{96}$$

and the incorrect value of γ in a mean field approximation gives a value of

T_t/T_N that is too high in the limit $\beta_c A \to 0$. This approximation also gives

$$r^2 = \frac{\beta A - a_1 t^{\gamma}}{3 a_2 t^{\gamma-\beta}}$$

from (94)

Substituting the value t_t making (95) vanish gives

$$r_t^2 = \frac{\frac{1}{3}\beta A}{3a_2}\left(\frac{2}{3a_1\beta A}\right)^{-\left(\frac{\gamma-2\beta}{\gamma}\right)}$$

$$= (\beta A)^{2\beta/\gamma}\left(\frac{2}{3a_1}\right)^{\left(\frac{2\beta}{\gamma}-1\right)}\frac{1}{9a_2}$$

$$r_t \propto (\beta_t A)^{\beta/\gamma} \qquad (97)$$

The expansion parameter $x = t/r^{1/\beta}$ then takes a value $\sim \frac{(\beta A)^{1/\gamma}}{(\beta A)^{1/\gamma}}$ of order 1. Thus we cannot find the tricritical point self consistently within the large x asymptotic series. Note, however that the value of r_t found would give a tricritical field

$$\beta h_t \propto (\beta A)(\beta A)^{\beta/\gamma}$$

$$\frac{h_t}{A} \propto (\beta A)^{\beta/\gamma} \qquad (98)$$

The exponent β/γ is in mean field theory $\frac{1}{2}$ but for the two-dimensional Ising mode $\frac{1}{8}/\frac{7}{4} = \frac{1}{14}$. Mean field theory would therefore give a value for h_t/A much less than would a correct equation of state in the plane.

The tricritical temperatures and fields found above are not consistent with the large x assumption under which truncation of series (91) is valid. Nevertheless they illustrate how the very different exponents of mean field theory and low dimensional equations of state predicted from (69) may tend

to depress the tricritical field and raise the ratio T_t/T_N for strong anisotropy in the exchange strengths.

Throughout this work we have relied on a mean field approximation for the interplanar coupling. This is questionable but can give qualitatively correct behaviour for highly anistropic systems. Consider, for example, an estimation for the critical temperature in a highly anisotropic two-dimensional Ising model, or a two-dimensional metamagnet with planes collapsed to lines, in zero field. Simple mean field theory gives $T_N = 2J_1 + 2J_1'$, whereas if the exact equation of state for the strongly coupled ferromagnetic chains is taken with a mean field approximation for the weak tranverse bonds the susceptibility is predicted

$$\chi = \frac{\chi_{1d}}{1-2J_1'\chi_{1d}} \tag{99}$$

Substituting $\chi_{1d}(T) = \beta e^{2\beta J_1}$ (100)

produces a divergence at

$$T_N \simeq \frac{2J_1}{\ln(J_1/J_1')} \tag{101}$$

Onsager's exact solution $\sinh\frac{2J_1}{KT_c}\sinh\frac{2J_1'}{KT_c} = 1$ predicts $T_N \simeq \frac{2J_1}{\ln(2J_1/J_1')}$ showing that (101) is asymptotically correct, in contrast to the simple mean field value $2J_1 + 2J_1'$ which is completely wrong. The Bethe-Peierls approximation for the chain z = 2 has the exact low temperature form (100) and hence also predicts (101). The reason that the mean field approximation (99) gives a good result for T_N is that the T_N so predicted is much greater than J_1'. (99) should be valid for temperatures greater than those of the order of J_1'.

1.7 Beyond Mean Field Theories.

We saw in 1.5 indications both numerical and experimental that near percolation in the plane the tricritical temperature approaches zero and the tricritical field approaches the zero temperature value. From the discussion in section 1.6 we would expect the mean field treatment of the interplanar coupling to be invalid for temperatures comparable to A and we conclude the phase diagram may be as drawn in figure 1.8 with $T_t \sim A << T_N$. An approach to the tricritical properties near the planar percolation threshold might be made in terms of geometric models of the percolating cluster as discussed in Chapter 3. At the percolation threshold correlations spread essentially one dimensionally, therefore we might expect the phase diagram of the metamagnet to be close to that of a two-dimensional metamagnet with ferromagnetically coupled chains rather than planes. The Bethe-Peierls dilution models percolation as exactly that - at p_c the correlation z is reduced to that of a chain. As the Bethe-Peierls approximation for $z = 2$ has the correct singularities near $T = 0$ the theoretical treatment may be qualitatively correct (see however the next section). Whether T_t is actually zero or finite of order A might be predicted by more accurate treatment of the two-dimensional metamagnet.

From renormalization group ideas (e.g. Toulouse and Pfeuty 1975) we expect critical and multicritical properties of a thermodynamic systems to depend on dimensionality and local symmetries of the Hamiltonian. The nature of the phase diagram should be the same for different Hamiltonians in the same universality class. If we consider the metamagnet in an external field close to the critical field there are three states of low

energy, namely the two degenerate antiferromagnetic ground states and the ferromagnetic ground state with moment in the direction of the field. The phase transition at the critical field is caused by the crossing of energies of the possible orderings. For $H > H_c$ there is a singlet ground state and two degenerate states of higher energy, for $H < H_c$ two degenerate ground states and one state of higher energy. We might then expect the Hamiltonian to fall in the same universality class as a 3 state Potts model(as defined in Chapter 2) with an external field applied in the direction of one of the states (Fig. 1.9)

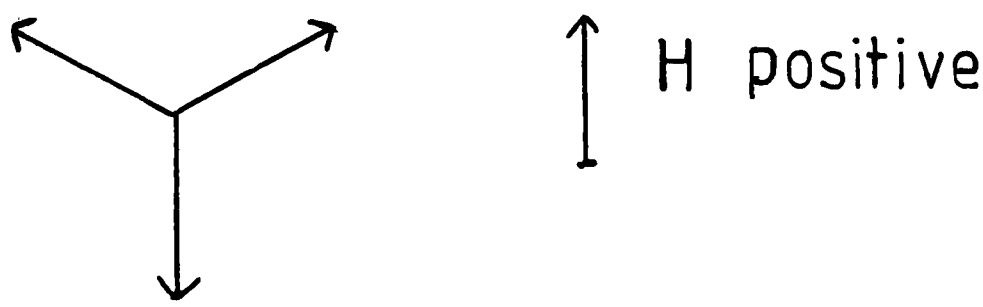


Figure 1.9. Vector representation of the 3-state Potts model in a field.

That model has been studied by mean field theory, by the renormalization group, analytically in two dimensions and experimentally in cubically anisotropic ferromagnets in a field (Straley and Fisher 1973, Baxter 1973, Mukamel, Fisher, Domany 1976, Barbara, Rossignol and Bak 1978). While their conclusions are not definite the indications are that the phase diagrams are as follows: either there is an "anomalous tricritical point" P at zero field, and a critical Ising line at positive fields (Fig. 1.10), as is suggested by exact solutions in zero field to be the case in two dimensions, or a quadrupole point Q at zero field and tricritical point T_1 at finite positive field (Figure 1.11) as found in mean field theory.

There is experimental evidence that figure 1.10 is appropriate for three dimensions from the study of a cubic ferromagnet in a near diagonal magnetic field. Thus it seems that fluctuations drive the

tricritical point to the symmetric axis H = 0 at two dimensions. If the metamagnetic Hamiltonian were in the same universality class as the symmetric Potts model in a field then the line QT_1C of figure 1.11 might correspond to the line of transitions in the phase diagram of the metamagnet. In that case the change of the phase diagram of figure 1.11 in three dimensions to that of figure 1.10 in two would be analogous to the change of figure 1.1 to that of Figure 1.6. Immediate difficulties are apparent: the point P at finite temperature is unlikely to correspond to the point ($H = H_c$, $T = 0$) of the metamagnet and an analogue of the line WQ does not appear in the phase diagram of the metamagnet.

Such speculations might be tested by renormalization group techniques and analogy, however weak, may serve to illustrate the difficulty of applying mean field results in low dimensions. It must be emphasised that the "two-dimensional metamagnet" discussed here is not the same as the Ising model of a metamagnet found to have tricritical behaviour by Nienhuis and Nauenberg 1976, using real space scaling techniques. They discuss a two-dimensional lattice with nearest neighbour antiferromagnetic coupling and next to nearest neighbour ferromagnetic coupling. The ferromagnetic couplings connect a two-dimensional sublattice with finite critical temperature in contrast to our two-dimensional metamagnet in which the ferromagnetically coupled chains have zero critical temperature.

1.8 Effects of Disorder.

In the treatment of the phase diagram of the dilute metamagnet presented, nowhere has randomness appeared - the dilution p occurs only as a parameter in the interlayer coupling constant and in the number

of nearest neighbours. As seen in 1.6 thermodynamic fluctuations may change the phase diagram ; disorder should also be important. One way the effects of disorder might be included is within a mean field interlayer approximation by including the equation of state for a dilute Ising system. By the Harris criterion (1974) we expect that the critical behaviour of the dilute Ising model may be the same as that of the pure if the specific heat exponent α_{pure} is negative but it may not if α_{pure} is positive. As $\alpha = 0$ in the two-dimensional Ising model the argument is inconclusive. In renormalization group language (Aharony 1976) α/ν is the dimension of the operator associated with randomness: $\alpha = 0$ corresponds to a marginal operator.

Near the two-dimensional percolation concentration p_c there will be fluctuations in the local geometry of the infinite cluster. In section 1.7 we argued that the Bethe-Peierls dilution might give a reasonable model of the percolating infinite cluster but clusters that are connected via ferromagnetic couplings to only a finite number of occupied sites may contribute to long range order via antiferromagnetic couplings. Therefore geometric fluctuations may make the part of the $p = p_c$ phase diagram at temperatures comparable to A different from that of the two-dimensional metamagnet of the last section.

The critical line of the metamagnet has the exponents of the three-dimensional Ising model which in the pure case has positive specific heat exponent $\alpha = 0.12 > 0$ (e.g. Domb 1974). Exponents for the dilute case should differ and be those for a random Ising model, as calculated within the ε expansion (Aharony 1978, Khmel'nitzkii 1975). There is also the question of the effects of fluctuations on the sharpness of the first order phase transitions. The general problem of the effects of disorder on

first order phase transitions will be discussed in Chapter 2 with the conclusion that close to the tricritical point there will be a sharp first order phase transition. Presence or absence of smearing could not easily be judged by experiment in dilute metamagnets as demagnetising effects spread the first order transition over a range of applied fields in the pure system. The sharp first order phase transition is recovered only if correction is made for demagnetising effects that may not be separable from random effects.

As mentioned in 1.2 the Harris criterion implies that the tricritical scaling properties should be strongly affected by presence of disorder. Bergman, Aharony and Imry (1978) have suggested a percolative type transition in the ransom case. Stephen (1976) on the other hand, found a random tricritical point differing from the pure fixed point in three dimensions in its logarithmic corrections to mean field theory. It does not seem consistent with Harris' argument that such a fixed point with $\alpha_t = \frac{1}{2}$ could describe criticality in a random model.

1.9 Conclusions.

The simple mean field theory for the critical line and tricritical point in Ising metamagnets has been improved by treating the planar fluctuations within a Bethe-Peierls approximation, giving substantially better agreement with numerical results for isotropic coupling strengths. The approximation allows treatment of dilution with the result, for anisotropy in the coupling strengths, that the tricritical field approaches the zero temperature transition field as dilution approaches the

percolation threshold for the plane. This is in agreement with experiments on dilute $FeCl_2$. The quantitative discrepancy in predicted values of tricritical field and temperature for pure $FeCl_2$ is related to the failure of the Bethe-Peierls approximation to produce correct critical singularities. Examination of the approximations employed allow the conclusion that at the planar percolation concentration the phase diagram is as in Figure 1.8 with T_t (of order J_1') much less than T_N (of order $2J_1/\ln (J_1/J_1')$).

Appendix A. The Bethe-Peierls Equation of State

The Bethe-Peierls equation of state m(H) is found by calculating the partition function Z_p for the cluster consisting of a single spin in the field H surrounded by z nearest neighbours, each in a fictitious field H_o and interacting via exchange J

$$Z_p(\beta, H, H_o) = \sum_{\substack{S_0 = \pm 1 \\ S_i = \pm 1 \\ i = 1, \ldots z}} e^{\beta J \sum_{i=1}^{z} S_0 S_i + \beta H_0 \sum_{i=1}^{z} S_i + \beta H S_0}$$

$$= \sum_{S_0 = \pm 1} e^{\beta H S_0} \prod_{i=1}^{z} \left(\sum_{S_i = \pm 1} e^{\beta J S_0 S_i + \beta H S_0 S_i} \right)$$

$$= e^{\beta H} 2^z \cosh^z(\beta J + \beta H_0) + e^{-\beta H} 2^z \cosh^z(-\beta J + \beta H_0)$$

The field H_o is determined as a function of H by requiring that the expectation value of the central spin s_o equal that of each of its neighbours

$$\langle S_0 \rangle = \frac{1}{\beta Z_p} \frac{\partial Z_p}{\partial H} = \langle S_i \rangle = \frac{1}{z \beta Z_p} \frac{\partial Z_p}{\partial H_0}$$

$$e^{\beta H} \cosh^z(\beta J + \beta H_0) - e^{-\beta H} \cosh^z(-\beta J + \beta H_0)$$

$$= e^{\beta H} \cosh^{z-1}(\beta J + \beta H_0) \sinh(\beta J + \beta H_0) - e^{-\beta H} \cosh^{z-1}(\beta J - \beta H_0) \sinh(\beta J - \beta H_0)$$

$$e^{\beta H}\cosh^{z-1}(\beta J+\beta H_0)\left(\cosh(\beta J+\beta H_0)-\sinh(\beta J+\beta H_0)\right)$$

$$= e^{-\beta H}\cosh^{z-1}(\beta J-\beta H_0)\left(\cosh(\beta J-\beta H_0)-\sinh(\beta J-\beta H_0)\right)$$

$$e^{2\beta H}\cosh^{z-1}(\beta J+\beta H_0)\,e^{-(\beta J+\beta H_0)} = \cosh^{z-1}(\beta J-\beta H_0)\,e^{-(\beta J-\beta H_0)}$$

$$e^{2\beta H} = e^{2\beta H_0}\left(\frac{\cosh(\beta J-\beta H_0)}{\cosh(\beta J+\beta H_0)}\right)^{z-1} \qquad \text{(A1)}$$

The equation of state is found by substituting for H_o from A1.

$$\langle S_o\rangle = \frac{e^{\beta H}\cosh^{z}(\beta J+\beta H_0)-e^{-\beta H}\cosh^{z}(\beta J-\beta H_0)}{e^{\beta H}\cosh^{z}(\beta J+\beta H_0)+e^{-\beta H}\cosh^{z}(\beta J-\beta H_0)}$$

$$= \frac{e^{2\beta H}\left(\frac{\cosh(\beta J+\beta H_0)}{\cosh(\beta J-\beta H_0)}\right)^{z}-1}{e^{2\beta H}\left(\frac{\cosh(\beta J+\beta H_0)}{\cosh(\beta J-\beta H_0)}\right)^{z}+1}$$

$$= \frac{e^{2\beta H}\,e^{2\beta(H_0-H)\frac{z}{z-1}}-1}{e^{2\beta H}\,e^{2\beta(H_0-H)\frac{z}{z-1}}+1}$$

From A1

$$= \tanh\left[\frac{\beta H_0 z-H}{z-1}\right]$$

The equation of state is therefore

$$m(H) = \tanh\left[\frac{\beta H_0(H) z - \beta H}{z-1}\right] \qquad \text{(A2)}$$

where $H_o(H)$ is defined by (A1). (A1) can be differentiated with respect to H at constant β.

$$2e^{2\beta H} = \frac{dH_0}{dH}\left[2e^{2\beta H} - (z-1)\tanh(\beta J - \beta H_0)e^{2\beta H} - (z-1)\tanh(\beta J + \beta H_0)e^{2\beta H}\right]$$

$$= 2e^{2\beta H}\frac{dH_0}{dH}\left[1 - \frac{(z-1)\sinh(2\beta J)}{2\cosh(\beta J - \beta H_0)\cosh(\beta J + \beta H_0)}\right]$$

$$\frac{dH_0}{dH} = \frac{1}{1 - \left(\frac{(z-1)\sinh(2\beta J)}{\cosh 2\beta J + \cosh(2\beta H_0)}\right)} \qquad \text{(A3)}$$

Equation (A1) has at least one solution for each value of H since A1 defines a continuous function $H(H_o)$ with $H(H_o \to -\infty) = -\infty$ $H(H_o \to +\infty) = +\infty$, thus $H(H_o)$ takes all intermediate values. There will be a uniquely defined solution provided $\frac{dH}{dH_o} > 0$. But for $\tanh\beta J < \frac{1}{z-1}$

$$\frac{(z-1)\sinh(2\beta J)}{\cosh(2\beta J) + \cosh(2\beta H_0)} \leq (z-1)\frac{\sinh 2\beta J}{\cosh 2\beta J + 1}$$

$$= (z-1)\tanh\beta J$$

$$< 1$$

From A3 $\tanh\beta J < \frac{1}{z-1}$ implies $\frac{dH}{dH_o} > 0$. Also $\frac{dH_o}{dH}$ diverges and therefore $m'(H)$ at $H_o = H = 0$ and

$$\tanh\left(\frac{J}{kT_c}\right) = \frac{1}{z-1} \qquad \text{(A4)}$$

For $T > T_c$ $\frac{dH_o}{dH}$ is defined from (A3), which can also be differentiated to give

$$\frac{d^2H}{dH_o^2} = \frac{(z-1)(\sinh 2\beta J)\, 2\beta \sinh 2\beta H_o}{(\cosh 2\beta J + \cosh 2\beta H_o)^2}$$

$$\frac{d^3H}{dH_o^3} = \frac{(2\beta)^2(\cosh 2\beta H_o)(z-1)\sinh 2\beta J}{(\cosh 2\beta J + \cosh 2\beta H_o)^2}$$

$$- \frac{2(2\beta)^2(\sinh 2\beta H_o)^2(\sinh 2\beta J)(z-1)}{(\cosh 2\beta J + \cosh 2\beta H_o)^3}$$

$$= \frac{(2\beta)^2(z-1)\sinh 2\beta J}{(\cosh 2\beta J + \cosh 2\beta H_o)^3}\left[1 + \cosh 2\beta J \cosh 2\beta H_o - (\sinh 2\beta H_o)^2\right]$$

$\frac{d^2H_o}{dH^2}$ and $\frac{d^3H_o}{dH^3}$ are found from

$$\frac{d^2H_o}{dH^2} = \frac{-\left(\frac{d^2H}{dH_o^2}\right)}{\left(\frac{dH}{dH_o}\right)^3}$$

$$\frac{d^3H_o}{dH^3} = -\frac{d^3H}{dH_o^3}\left(\frac{dH}{dH_o}\right)^{-4} + 3\left(\frac{d^2H}{dH_o^2}\right)^2\left(\frac{dH}{dH_o}\right)^{-5}$$

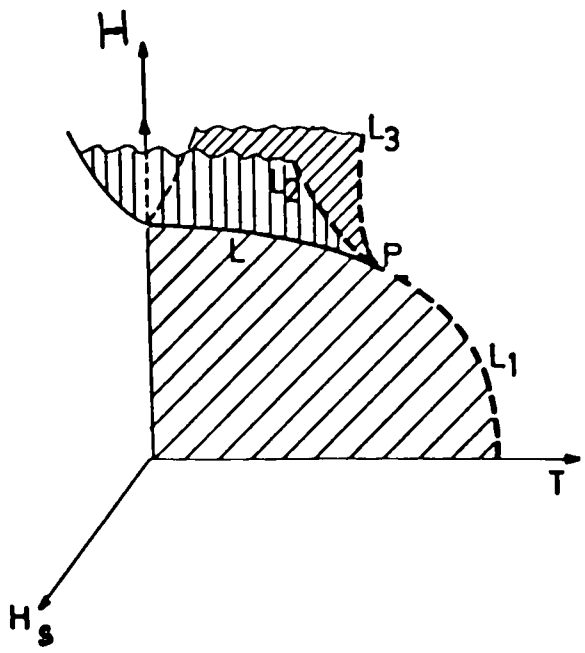
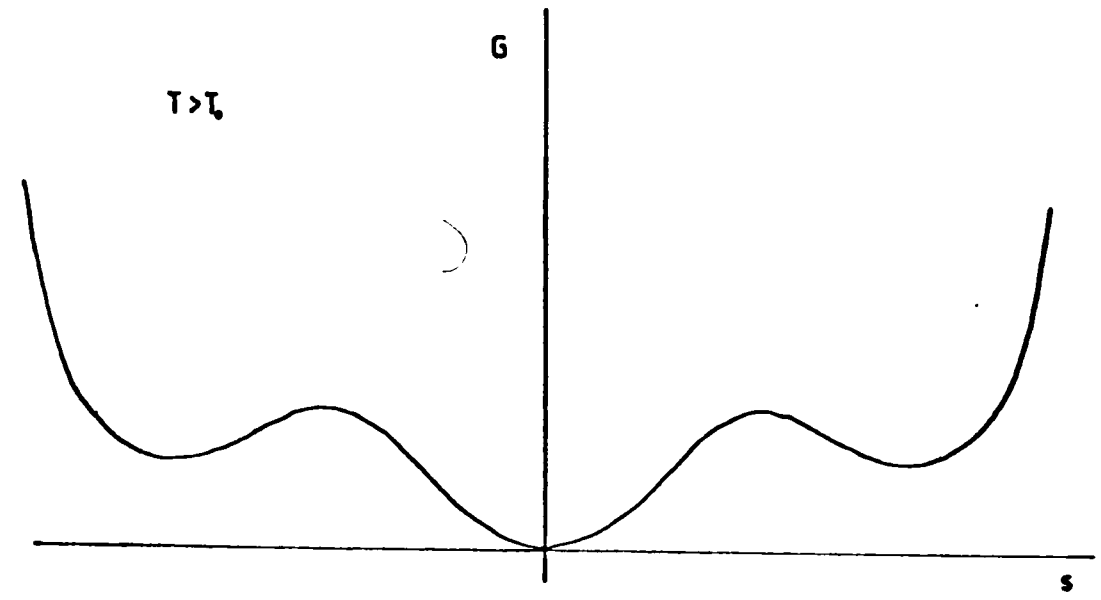
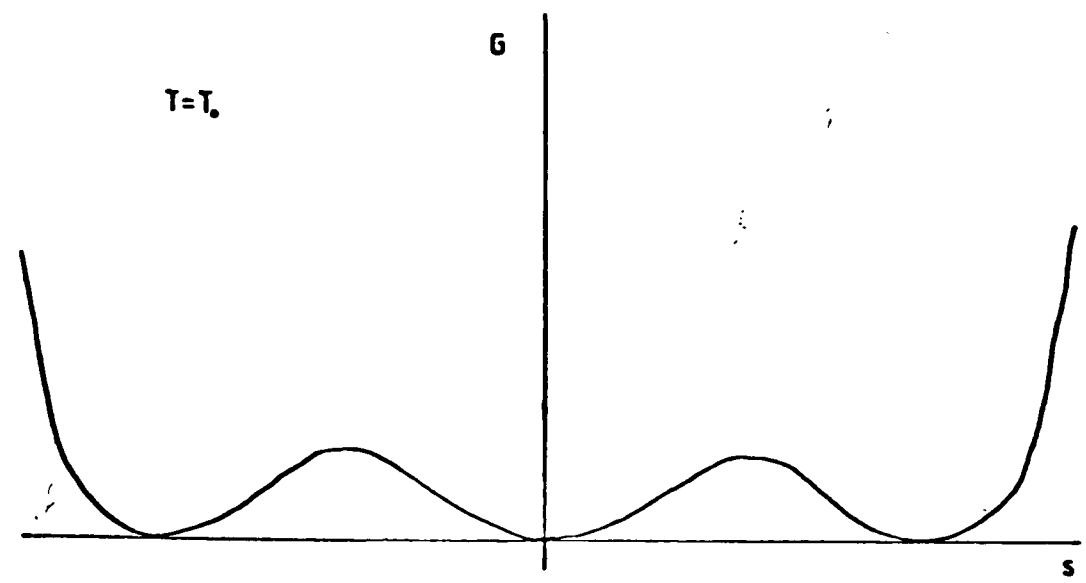


Figure 1.1. Phase diagram with tricritical point P. L_1, L_2, L_3 are lines of second order phase transitions, L is a line of first order phase transitions, the shaded surfaces are surfaces of first order phase transitions. (From Toulouse and Pfeuty 1975).





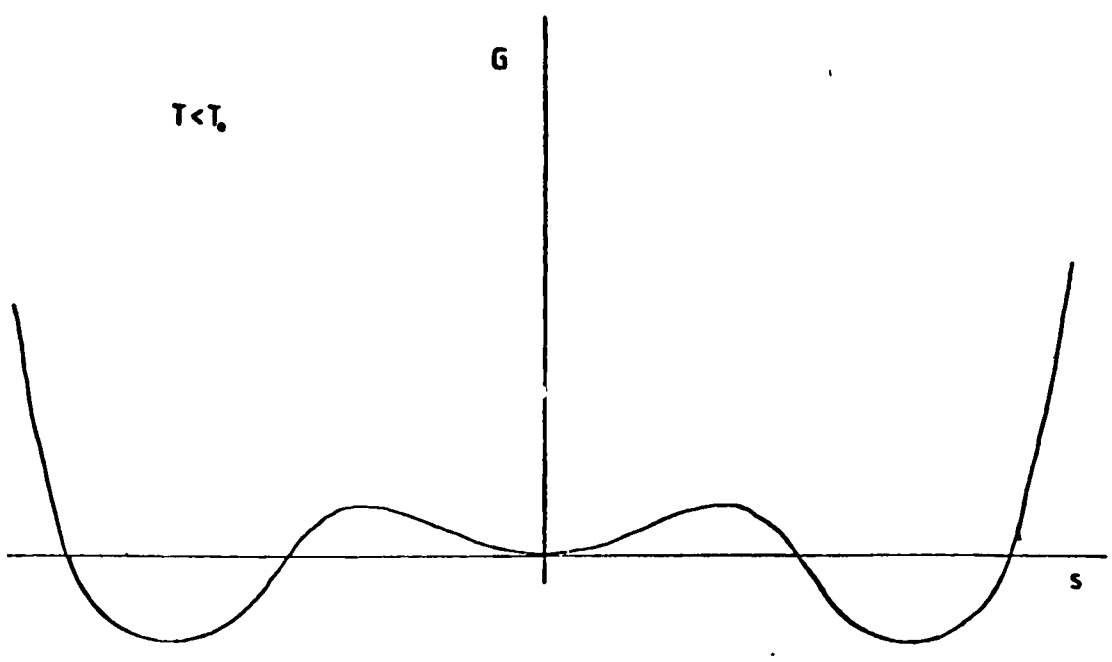


Figure 1.2. Landau theory for a first order phase transition at temperature $T = T_o$.

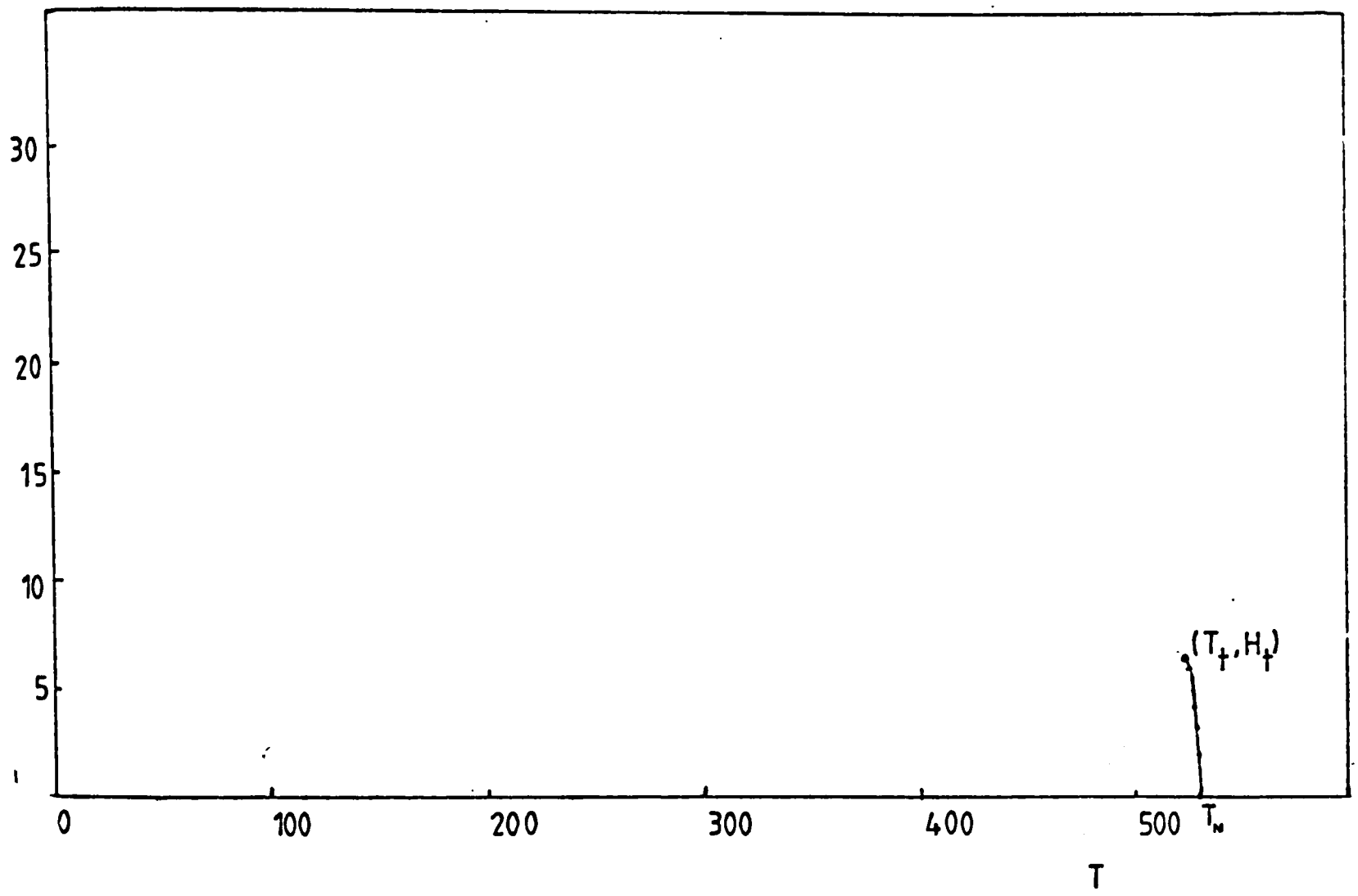


Figure 1.3. The critical line for $J_1 = 100$, $A = 30$, $z = 6$. $T_N = 530$, $T_t = 525$, $H_t = 6.6$, $H_c(T = 0) = 30$.

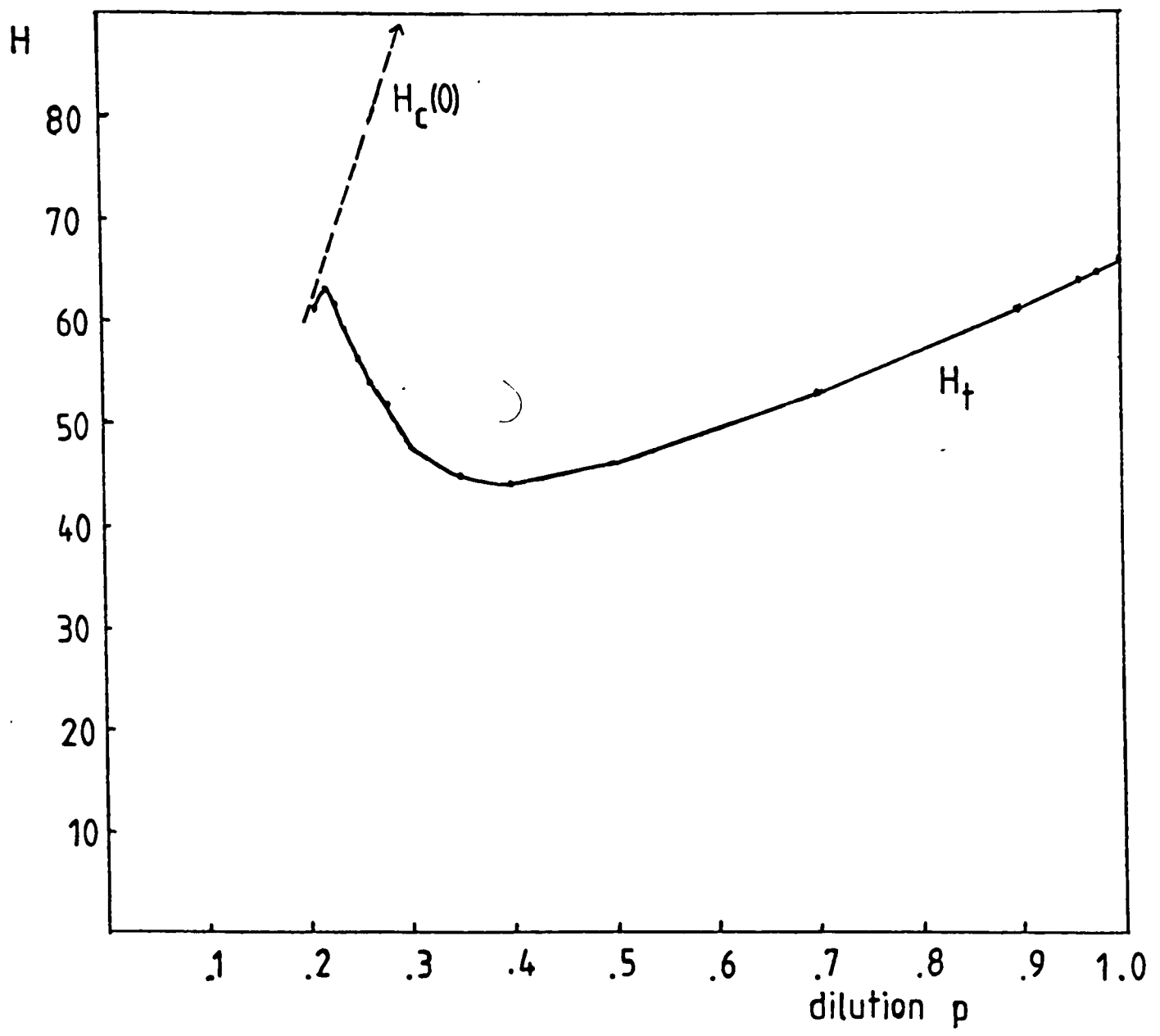


Figure 1.4. The tricritical field $H_t(p)$ as a function of dilution p.

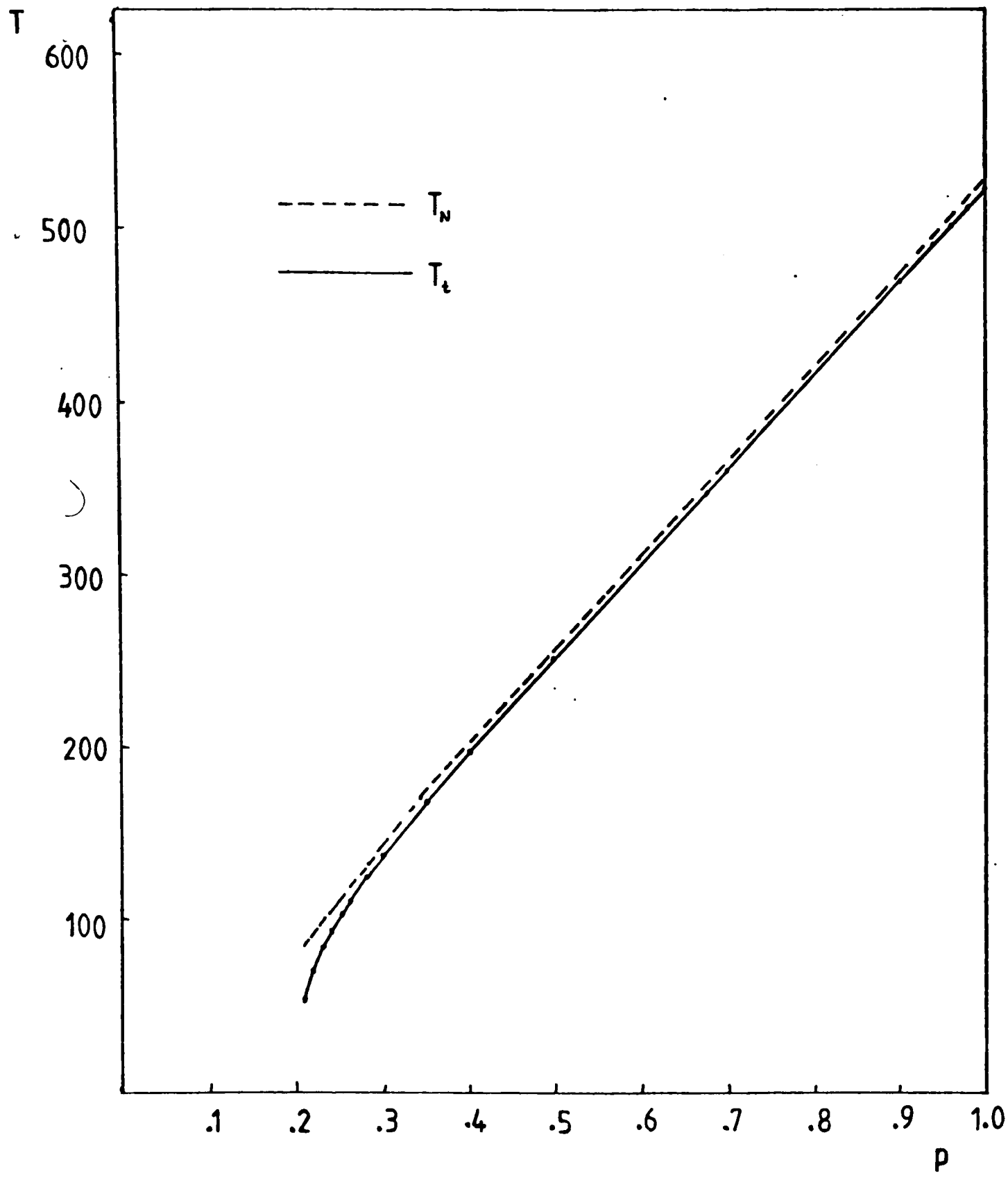


Figure 1.5. The Néel temperature $T_N(p)$ and the tricritical temperature $T_t(p)$ as a function of dilution p.

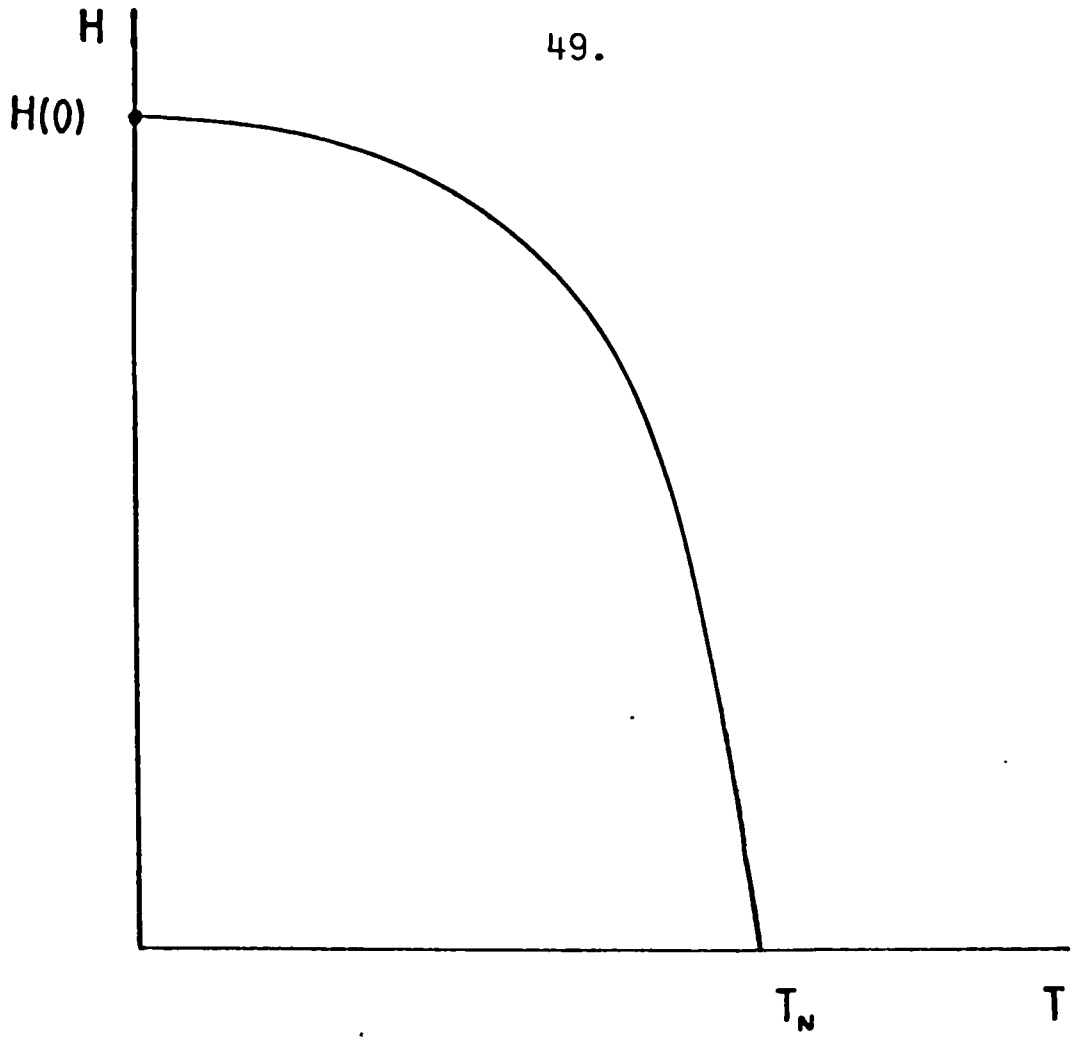


Figure 1.6. Tricritical phase diagram in the limit $T_t = 0$, $H_t = H(0)$.

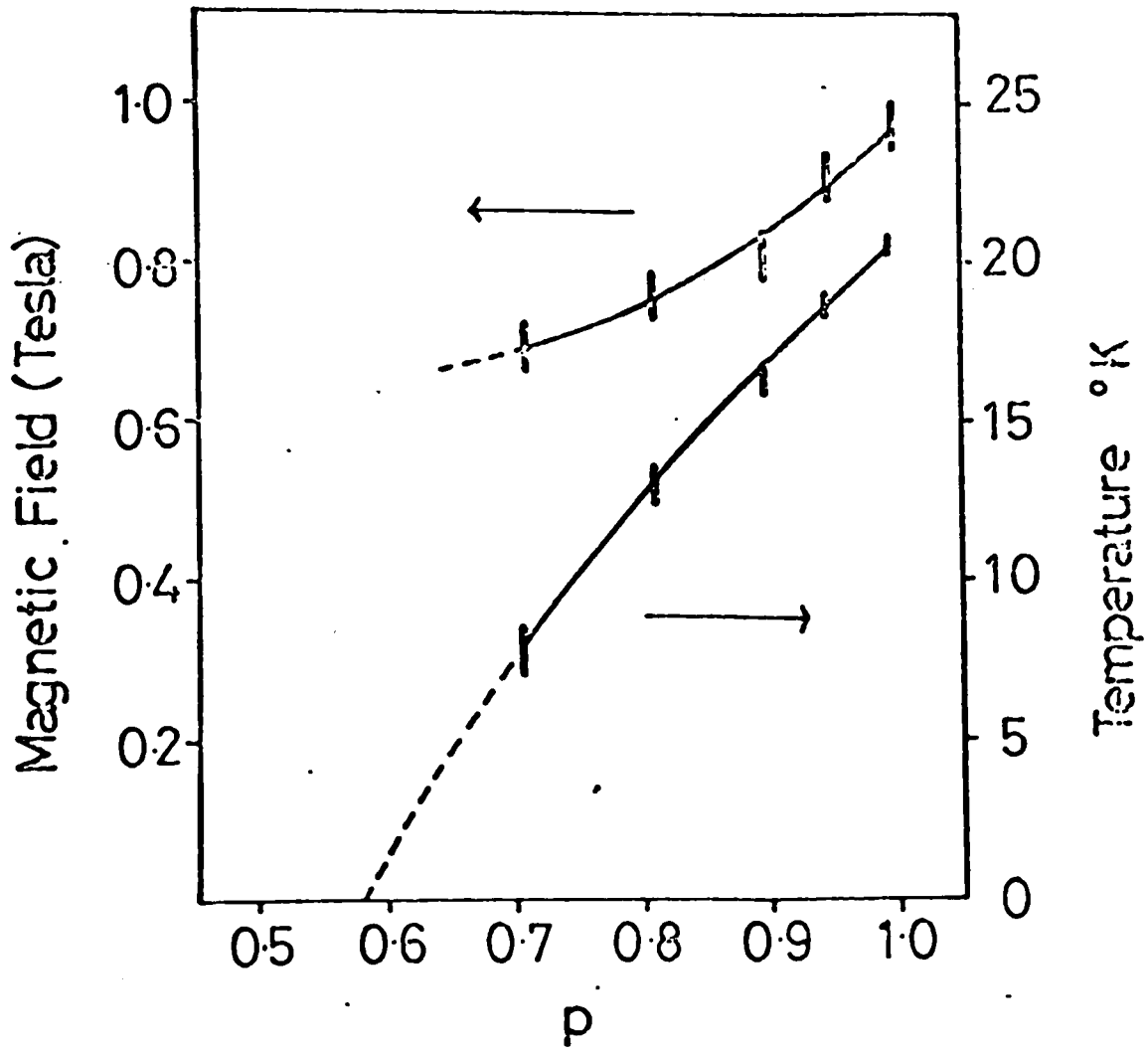


Figure 1.7. Experimentally determined tricritical field and temperature in the dilute metamagnet $Fe_pMg_{1-p}Cl_2$ from Wood and Day (1977).

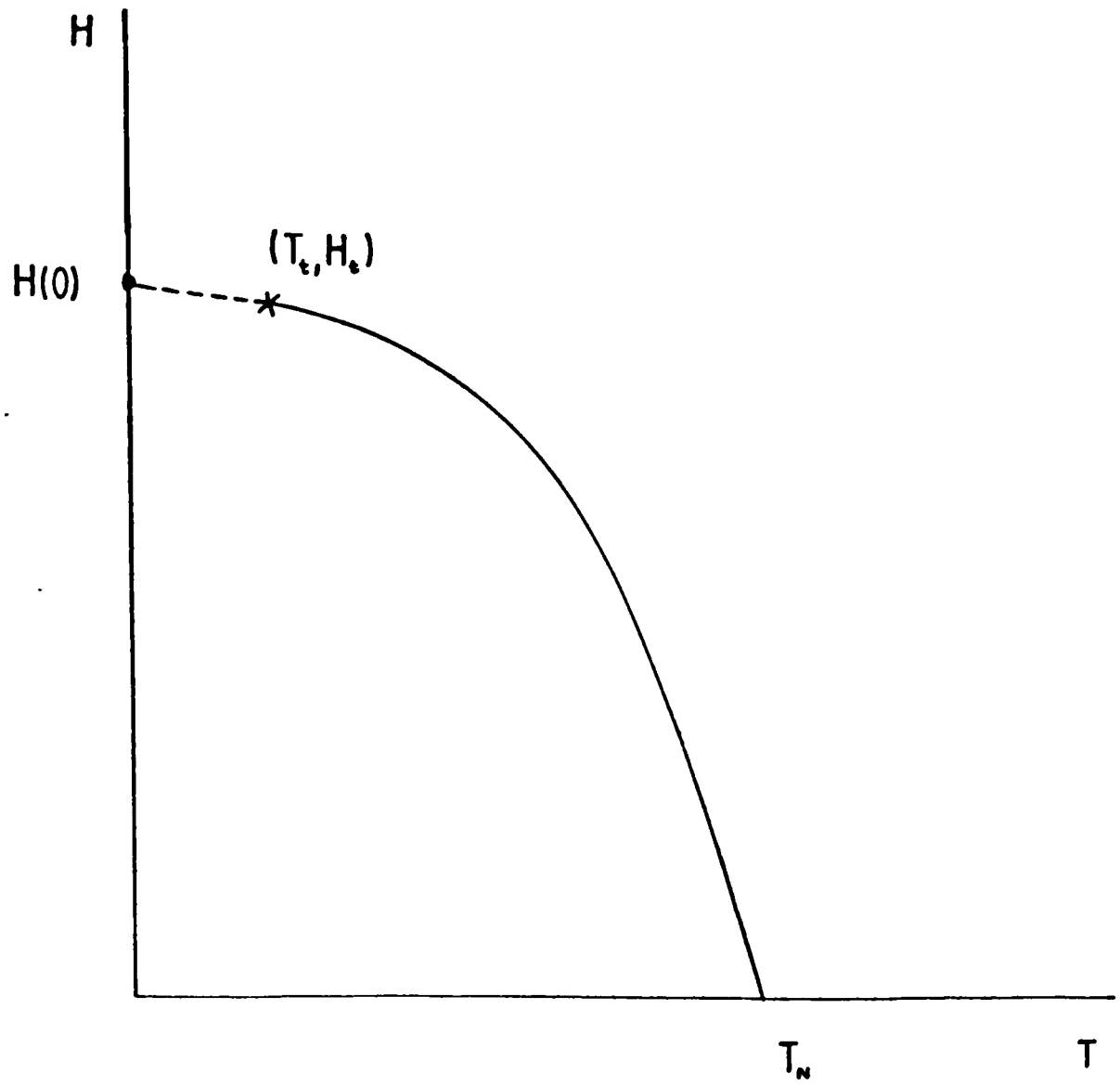


Figure 1.8. Proposed phase diagram for dilutions close to the percolation concentration of the plane. T_t is of order A, much less than T_N.

Figure 1.9. See text, page 34.

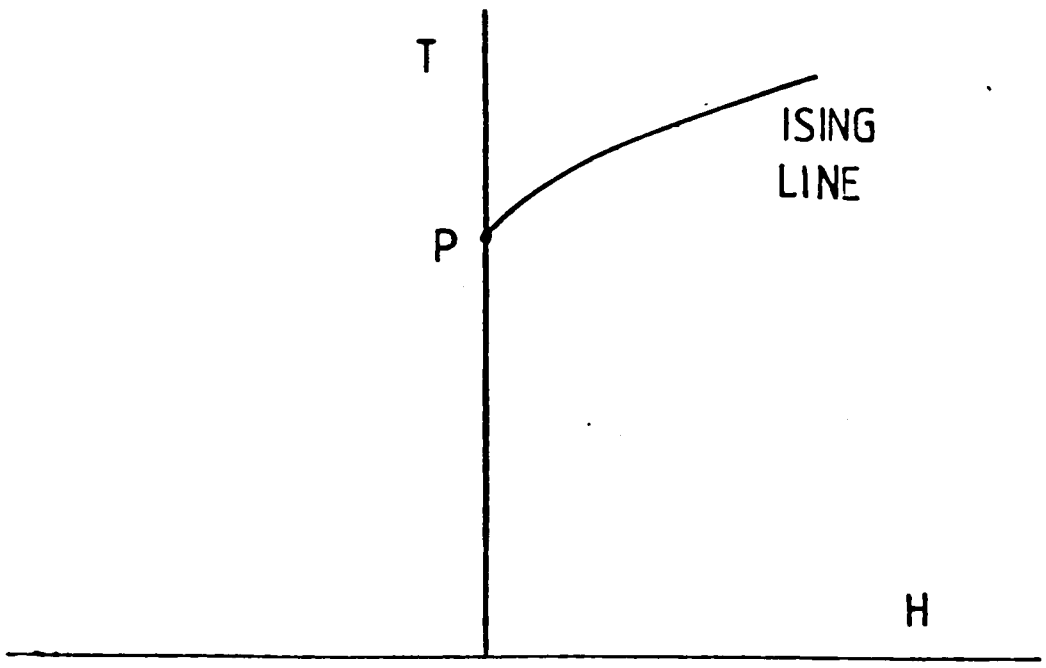


Figure 1.10. Possible phase diagram for two-dimensional 3-state Potts model in a field. P is the "anomalous tricritical point".

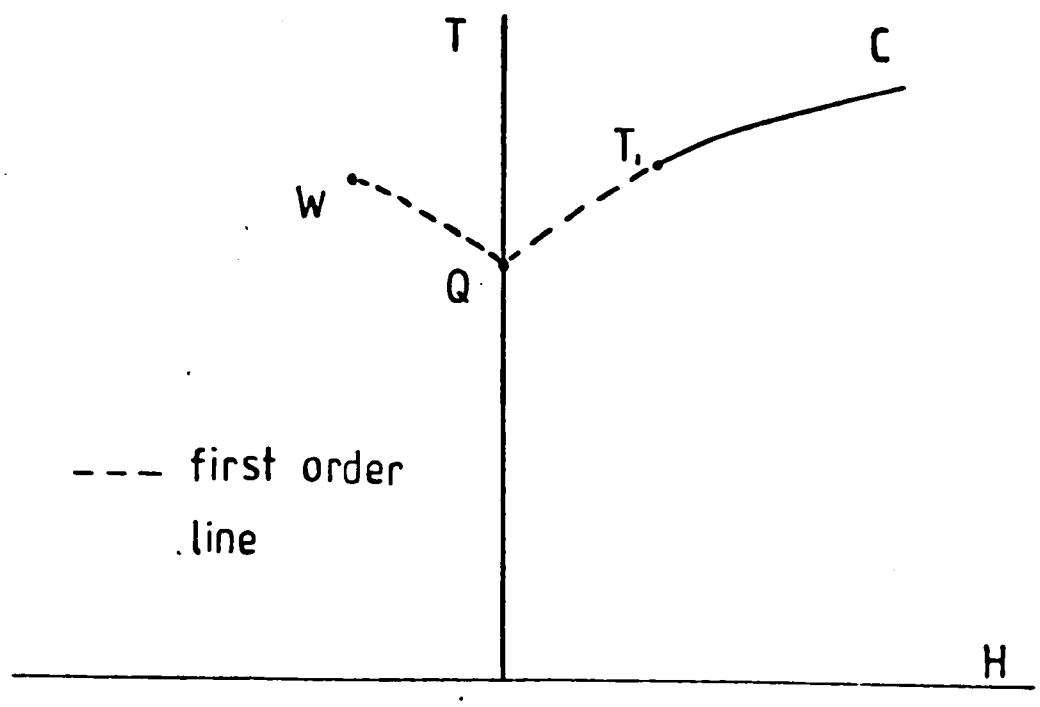


Figure 1.11. Mean field phase diagram for 3-state Potts model in a field. Q is a quadrupole point, WQ and QT lines of first order transitions, T_1 a tricritical point and C an Ising line.

CHAPTER 2

FIRST ORDER PHASE TRANSITIONS IN DISORDERED SYSTEMS

2.1 First Order Phase Transitions in Pure Systems.

While the theory of second order phase transitions has been greatly stimulated by ideas of scaling and the renormalization group, relatively little attention has been paid to phase transitions of first order. Theoretical problems in describing second order phase transitions arise from the divergent correlation length producing divergences in perturbative theory. Renormalization group techniques exploit the existence of a diverging correlation length, however, to provide universality arguments of great power. First order phase transitions, for which discontinuities occur in the first derivatives of the free energy, do not involve divergent correlation lengths so that universality arguments cannot be applied in the same way. Generally the mathematical structure remains obscure, although there have been some developments (Klein, Wallace and Zia 1976)..

Discussion of first and second order phase transitions in terms of Landau theory is straightforward and has been reviewed in Chapter 1. For a system symmetric under the transformation $m \leftrightarrow -m$ of the order parameter the Gibbs free energy is expanded

$$G(T,h) = am^2 + bm^4 + cm^6 - hm \qquad (1)$$

and m is defined by the value minimising the expansion. For $h = 0$, $a = a(T)$ (1) implies a second order transition for $a = 0$, $b, c > 0$

but for $b < 0$, $a = b^2/4c$, $c > 0$ there is a discontinuity in the equilibrium m and hence a discontinuity in the derivative of the free energy as a function of temperature.

A conclusion of the modern theory of critical phenomena is that Landau theory is incorrect. The functional form of Landau reappears, however, in the Ginsburg-Landau or Hamiltonian formulation of statistical mechanics, in which the partition function is written as a functional integral (see, for example Toulouse and Pfeuty 1975)

$$\beta = \frac{1}{k_B T}$$

$$Z = \int [\mathcal{D} m(x)] \exp\left[-\beta F_L\{m(x)\}\right] \quad (2)$$

where the free energy density functional $F_L\{m(x)\}$ can be written, for a single component order parameter,

$$F_L\{m(x)\} = \int \left[a m^2(x) + b m^4(x) + c m^6(x) + \ldots + (\nabla m(x))^2\right] d^d x \quad (3)$$

Landau theory is recovered by a saddle point approximation by including only the contribution of the configuration $\{m_o(x)\}$ minimising $\beta F_L\{m(x)\}$. The Gibbs free energy, to this approximation, is given by

$$G(T,H) = -\frac{1}{\beta} \ln Z$$

$$= F_L\{m_o(x)\} \quad (4)$$

The configuration $m_o(x)$ is clearly constant in space and equal to the value of m minimising

$$V(m) = am^2 + bm^4 + cm^6 + \ldots\ldots\ldots$$

Thus Landau theory is recovered. The first order phase transition corresponds to the crossing of the energies of two configurations

$\{m_1(x)\}$ and $\{m_2(x)\}$ as a function of temperature or external field.

Fluctuations can be important in changing the order of the transition or destroying it: consider a one component spin in a magnetic field with free energy density

$$F_L\{m(x)\} = \int [a(T)m^2(x) + bm^4(x) + (\nabla m(x))^2 - hm(x)]\, d^d x \qquad (5)$$

where $a = a'(T - T_o)$, $b > 0$. Then in Landau theory there is a second order phase transition at $h = 0$, $T = T_o$ and a first order phase transition as a function of field for all $a < 0$, since the configuration $\{m_o(x)\}$ minimising $\beta F_L\{m(x)\}$ changes discontinuously as h passes through zero. In renormalization group theory the effects of fluctuations are included by loop expansion about the saddle point. The conclusions are that for $d > 1$ there is a phase transition with onset of long range order but at a critical temperature $T_c < T_o$, and for $d < 4$ with a modified singularity in $m_o(T)$, the spontaneous magnetisation, for $T \to T_c$ from that predicted by Landau theory for $T \to T_o$. Thus for fixed $a < 0$ there is a first order transition with field for a sufficiently negative but fluctuations reduce the magnitude of the magnetisation discontinuity, for $T = T_c$ change the transition to second order with $m(h) = \pm|h|^{1/\delta}$ for $h \to 0^{\pm}$, and for $T_c < T < T_o$ remove the singularity in m(h) altogether.

Generally then a system showing a first order transition in mean field theory may, when fluctuations are included show a first order, second order or no phase transition. An example of a system with a first order phase transition as a function of temperature in mean field theory is the s-state Potts model for $s > 2$: a d-dimensional discrete lattice system with s possible states $\{S_i^{\alpha}\ \alpha=1,.....s\}$ at each point and Hamiltonian (Potts 1952)

$$\mathcal{H} = -K \sum_{\substack{<ij> \\ \text{nearest neighbours}}} \delta_{\alpha\beta} S_i^{\alpha} S_j^{\beta} \qquad (6)$$

For s=2 this is the Ising model which has a second order transition for d > 1, but for s > 2 mean field theory includes a cubic term and therefore predicts a first order transition (Straley and Fisher 1973) Baxter (1973) proved that in two dimensions the transition is of second order in zero field for $s \leq 4$. In higher dimensions a continuous spin version of the s = 3 Potts model has been studied by the renormalization group and for small $\varepsilon = 4-d$ predicted to be first order (Amit and Shcherbakov 1974, Rudnick 1975). In three dimensions for s = 3 both renormalization group calculations (Golner 1973, Burkhardt, Knops and den Nijs 1976) and series expansions (Ditzian and Oitmaa 1974, Straley 1974) have given conflicting results. Mukamel, Fisher and Domany (1976) have argued that ferromagnetic crystals with cubic anisotropy may show a phase transition as a function of diagonal external field described by a 3 state Potts model and suggested that experiment might resolve the question. Experiments on $DyAl_2$ (Barbara, Rossignol and Bak 1977) support the conclusion that the transition is of first order in three dimensions. In any case the picture emerging is that for each s, for sufficiently large dimension, including $d \geq 3$ for s = 3 if the experimental evidence is accepted, mean field theory is correct in predicting the first order nature of the transition but at low dimensions, including d = 2 for $s \leq 4$ the transition is of higher order.

As already mentioned the application of renormalization group techniques to first order transitions is less illuminating than it is to

second order because of the lack of universality. Nevertheless scaling transformations can be applied to the free energy density and the flow diagram and fixed point structure used to predict thermodynamic properties. Bak, Krinsky and Mukamel (1976, Bak and Mukamel 1976) argued that the lack of a stable fixed point within the ε expansion for certain anisotropic Hamiltonian with order parameter of dimensionality $n \geq 4$ indicated that fluctuations induce a first order phase transition rather than the second order transition found in mean field theory. They showed that under recursion an initial free energy functional was transformed into one showing a first order phase transition in mean field theory. By this reasoning they explained observed first order phase transitions in Cr, Eu, UO_2, MnO, which have order parameters of dimension $n \geq 4$ corresponding to antiferromagnetic or incommensurate ordered structures. A difficulty is that absence of a stable fixed point has also been interpreted as indicating a smeared transition in certain problems of disorder (discussed by Aharony 1976). Furthermore inability to find a stable fixed point within an ε expansion may not preclude existence of one with coordinates outside the range of validity of the recursion relations. It is important to look closely at the flow to decide whether the expected transition is first order or smeared. In studying the effects of disorder on first order transitions such difficulties should be acute.

A slightly different approach has been that of Nienhuis and Nauenberg (1975) who claimed that an unstable fixed point with magnetic eigenvalue b^d under scale transformation $L \rightarrow L/b$ is associated with a discontinuity in bulk equilibrium magnetisation. Aharony (1977) has suggested that a thermal exponent $\nu = 1/d$ might similarly be interpreted

as showing a discontinuity in the interal energy. An exponent $\nu < 1/d$ is unphysical, as from the hyperscaling relation $\alpha = 2 - d\nu$ it implies $\alpha > 1$ giving a non-integrable specific heat singularity. Aharony proposed that the fact that his exponents calculated for an s-state Potts model tended to unphysical values $\nu < 1/d$ for large s corresponds to the transition's becoming first order.

2.2 Disordered Phase Transitions of First Order.

We consider now the problem of the effects of introducing disorder in the form of impurities to a system undergoing a first order phase transition. In the pure system at the transition there is a discontinuity in the first derivative of the free energy with respect to the varied external parameter. For a discontinuity induced by an external field this gives a discontinuity in magnetisation, for a thermally induced transition a discontinuity in the internal energy. The question is, what effect do quenched random impurities have on the discontinuity - do they smear it over a finite temperature range or do they simply reduce its amplitude while leaving it sharp?

Experimental systems to which this question is relevant include mixtures of perovskites having a first order structural phase transition, such as $Rb_xK_{1-x}MnF_3$ (Borsa, Benard, Walker and Baviera 1977); dilute metamagnetic systems in a field below the tricritical temperature, such as $Fe_xMg_{1-x}Cl_2$ (Chapter 1), and weakly anisotropic antiferromagnets such as $Mn_xZn_{1-x}F_2$ in a field at the spin-flop transition (King, Rezende,

Jaccarino, 1977) as well as disordered examples of supposed fluctuation induced phase transitions such as the antiferromagnetic to paramagnetic transition in impure Chromium (Bak 1976). Bak concluded from the recursion relations that such fluctuation induced first order transitions would be smeared for arbitrarily small concentrations of impurity. We shall consider the more usual case, as in the first three examples quoted, in which the pure systems are predicted by mean field theory to be first order and we discuss primarily the case of a small concentration of impurities.

To formulate the problem mathematically we consider a pure system that can be described by a free energy density functional

$$F_L^0\{m(x)\} = \int [a(m(x))^2 - b(m(x))^4 + c(m(x))^6 + (\nabla m(x))^2]\, d^d x \qquad (7)$$

As before a saddle point approximation to the partition function gives a Landau theory with free energy expansion

$$G(T,m) = a(T)m^2 - bm^4 + cm^6 \qquad (8)$$

If we take $a(T) = a'(T - T_o) + b^2/4c$ (9)

the Landau theory predicts a first order transition at $T = T_o$.

Presence of a random distribution of impurities may be modelled by a perturbing term

$$F'\{m(x)\} = \int \psi(x)\, m^2(x)\, d^d x \qquad (10)$$

where $\{\psi(x)\}$ is a random function with short range correlation $\langle\psi(x)\psi(x')\rangle \alpha\ \delta^{(d)}(x - x')$. For example $\psi(x)$ may be a continuum version of a function taking values $\psi(x_i) = \psi_i$ at each point of a lattice

independently with probability density function

$$p(\psi_i) = r\,\delta(\psi_i) + (1-r)\,\delta(\psi_i - \psi_o)$$

$$P(\{\psi(x_i)\}) = \prod_i p(\psi(x_i)) \qquad (11)$$

representing a quenched mixture of systems with critical temperatures T_o and $T_o - \frac{\psi_o}{a'}$ from (8) and (9). The total free energy density functional is

$$F_L\{m(x);\psi(x)\} = \int [(a+\psi(x))m^2(x) - bm^4(x) + cm^6(x) + (\nabla m(x))^2]d^dx \qquad (12)$$

The free energy is found, for such a quenched problem, by averaging the free energy for a particular impurity distribution $\{\psi(x)\}$ over all such distributions (Brout 1959, Aharony, Imry and Ma 1976)

$$G = \int \mathcal{D}\psi(x)\, P(\{\psi(x)\})\, G(T;\psi(x)) \qquad (13)$$

where

$$G(T;\psi(x)) = -\frac{1}{\beta} \ln Z(T;\psi(x))$$

$$Z(T;\psi(x)) = \int \mathcal{D}m(x) \exp[-\beta F_L\{m(x);\psi(x)\}] \qquad (14)$$

Unlike the pure case, it is not clear what a saddle point approximation to the functional integral (14) will be: minimising $F_c[\{m(x);\psi(x)\}]$ with respect to configurations $\{m(x)\}$ entails satisfying the conflicting requirements that $m(x)$ should vary slowly to minimise the $|\nabla m(x)|^2$

term and yet should be close to the local minimum of

$$V(m;\psi) = (a+\psi)m^2 - bm^4 + cm^6 \tag{16}$$

to minimise the other terms. Loss of translational invariance in the microscopic free energy density makes even the mean field theory difficult.

A technique developed to overcome the difficulty of evaluating the partition function of a Hamiltonian without translational invariance is the replica or $n \to 0$ method (Emery 1975). This was applied to renormalization group calculations for second order phase transitions in random systems (Lubensky 1975, Aharony, Imry and Ma 1976) and to spin glass calculations (Edwards and Anderson 1975). We apply this to the free energy density above, following notation of Aharony, Imry and Ma as applied to the second order case. We have

$$G = \int \mathcal{D}\psi(x)\, P(\{\psi(x)\})\, G(T;\psi(x)) \tag{17}$$

where $P(\{\psi(x)\})$ is the continuum limit of

$$P(\{\psi(x_i)\}) = \prod_i p(\psi(x_i)) \tag{18}$$

We now define a function g by

$$e^{-g(y)} = \int_{-\infty}^{\infty} d\psi\, p(\psi) e^{-y\psi} \tag{19}$$

and consider an n-component model with free energy density

$$F_{Ln}\{m_i(x)\} = \int d^d x \left\{ \sum_{i=1}^{n} \left[a m_i^2(x) - b m_i^4(x) + c m_i^6(x) + (\nabla m_i(x))^2 \right] + g\left(\sum_{i=1}^{n} m_i^2(x) \right) \right\}$$

and free energy G_n. Then, as in the second order case (Aharony, Imry and Ma (1976)

$$\lim_{n\to 0}\frac{G_n}{n} = -\frac{1}{\beta}\lim_{n\to 0}\frac{1}{n}\ln\left[\int\prod_{i=1}^{n}(\mathcal{D}m_i(x))\, e^{-\beta F_{Ln}\{m_i(x)\}}\right] \quad (20)$$

$$= -\frac{1}{\beta}\lim_{n\to 0}\frac{1}{n}\ln\int\mathcal{D}\psi(x)\,P(\{\psi(x)\})\times \left(\int\mathcal{D}m(x)e^{-\beta F_L[m(x);\psi(x)]}\right)^n \quad (21)$$

$$= -\frac{1}{\beta}\int\mathcal{D}\psi(x)P(\{\psi(x)\})\ln\int\mathcal{D}m(x)e^{-\beta F_L\{m(x);\psi(x)\}} \quad (22)$$

where the step (20) → (21) follows from (19) and the continuum limit of

$$e^{-\beta\int d^dx\, g(\sum_{i=1}^{n} m_i^2(x))} \simeq e^{-\beta\sum_\alpha g(\sum_1^n m_i^2(\alpha))\,\delta V_\alpha}$$

$$= \prod_\alpha e^{-\beta g(\sum_1^n m_i^2(\alpha))\,\delta V_\alpha}$$

$$= \prod_\alpha\int d\psi_\alpha\, p(\psi_\alpha)e^{-\beta(\sum_1^n m_i^2(\alpha))\psi_\alpha\delta V_\alpha}$$

$$= \int\left(\prod_\alpha d\psi_\alpha\right)\left(\prod_\alpha p(\psi_\alpha)\right)e^{-\beta\sum_\alpha\psi_\alpha(\sum_1^n m_i^2(\alpha))\,\delta V_\alpha}$$

$$\hookrightarrow \int\mathcal{D}\psi(x)P(\{\psi(x)\})\exp\left[-\beta\int d^dx\,\psi(x)\sum_{i=1}^{n}m_i^2(x)\right]$$

and the step (21) → (22) is simply

$$\frac{1}{n}\log\langle Z^n\rangle = \frac{1}{n}\log\langle 1 + n\log Z\rangle$$

$$= \frac{1}{n}\cdot n\log Z = \log Z \quad \text{for } n\to 0$$

Therefore in the limit n → 0 the translationally invariant free energy density (20) gives the same free energy per component as the random free

energy density (17). As (20) is translationally invariant we can find the saddle point approximation to it by taking $m_i(x) = m_i$ independent of x where $\{m_i\}_{i=1,n}$ minimises

$$V(m_1,\cdots,m_n) = \sum_{i=1}^{n} (am_i^2 - bm_i^4 + cm_i^6) + g\left(\sum_{i=1}^{n} m_i^2\right)$$

For distribution (11) $p(\psi_i) = r\delta(\psi_i) + (1 - r)\delta(\psi - \psi_u)$ the function

$$g(y) = -\ln\left[r + (1-r)e^{-y\psi_0}\right]$$

$$= (1-r)y\psi_0 \qquad (23)$$

to lowest order in y. For m_i^2 bounded $\sum_{i=1}^{n} m_i^2$ should be of order n for small n and $g(\sum_{i=1}^{n} m_i^2)$ can be approximated by its leading term and V can be written

$$V(m_1,\cdots,m_n) = \sum_{i=1}^{n} (a + (1-r)\psi_0)\, m_i^2 - bm_i^4 + cm_i^6 \qquad (24)$$

Therefore in the limit $n \to 0$ we expect the solution to be the analytic continuation of that with $m_i\{x\} \equiv m$ for all x and i and m the minimum of $V(m) = (a - (1 - r)\psi_0)m^2 - bm^4 + cm^6$. We expect, then, a first order phase transition at a transition temperature defined by, from (9)

$$a(T_c) + (1-r)\psi_0 = a(T_0)$$

$$T_c = T_0 - (1-r)\frac{\psi_0}{a'} \qquad (25)$$

A mean field approximation applied to the translationally invariant free energy density (20) in the limit $n \to 0$ therefore yields a sharp first

order transition for the random system. The theory could clearly be extended to higher dimensional order parameters with the same results.

The treatment obviously glosses over mathematical difficulties of the replica method such as the interchange of the $n \to 0$ limits and the thermodynamic limit (Aharony 1978). Analytic continuation of expressions obtained for integral n may lead to exchange of relative minima and maxima in function space (Thouless, Anderson and Palmer 1977). We exchange the difficulties of treating a non-translationally invariant problem with those of analytic continuation. The result agrees, however, with the intuitive notion that the two extremal configurations corresponding to high and low temperature minima are widely separated in function space and when perturbed may exchange roles of absolute minimum and metastable local minimum at a perturbed temperature but cannot transform continuously one to the other.

The question remains, when is such a mean field approximation valid? We would expect it to be for a weak impurity or small concentration provided the dimension of space is sufficiently great. How great the dimension must be and how weak the disorder will be discussed in a scaling theory in the next section.

2.3 Scaling Theory for Disordered First Order Phase Transitions.

In the theory of second order phase transitions of simple model systems there appear upper and lower critical dimensions d_u and d_L. For dimensions $d > d_u$ mean field theory is qualitatively correct in that the transition occurs with exponents predicted by mean field theory.

Fluctuations may be treated formally by a convergent loop expansion around the zero loop saddle point approximation (Brézin, Le Guillou, Zinn-Justin 1976), and depress the temperature of the transition but do not change its nature. For $d_L < d < d_u$ a phase transition occurs but with different exponents from those predicted by mean field theory. For $d < d_L$ the transition is suppressed by fluctuations altogether. For example an Ising spin system with short range interactions has $d_u = 4$, $d_L = 1$, while a classical isotropic Heisenberg spin system has $d_u = 4$ and $d_L = 2$. While the problem may be studied formally in terms of the equivalent field theory, one can evaluate the upper and lower critical dimensions by simple physical arguments: the Ginzburg criterion for the self consistency of mean field exponents successfully predicts d_u for the above, as well as more complicated examples such as dipolar coupled uniaxial ferromagnets in which $d_u = 3$ (see review by Als-Nielson and Birgeneau 1977). The lower critical dimension can be predicted by the existence of scale invariant excitations destroying the ordered phase above zero temperature (Migdal 1975). Scaling ideas may be a useful guide then to behaviour as a function of dimension.

A random problem for which a scaling argument has been developed is that of a spin system with a random field h(x) conjugate to the order parameter (Imry and Ma 1975), giving a contribution to the energy density

$$\Delta F_i \{\underset{\sim}{m}(x)\} = -\int d^d x \, \underset{\sim}{h}(x) \cdot \underset{\sim}{m}(x) \qquad (26)$$

h(x) is a random function of zero mean and short range correlations

$$\langle h_i(x_1) h_i(x_2) \rangle = \sigma_h^2 \delta_{x_1 x_2} \qquad (27)$$

We shall talk in terms of a ferromagnetic ordered state although the discussion is clearly applicable to more general orderings with a random field coupled linearly to the order parameter. If we consider that for temperatures T less than the critical temperature T_c the spin system without the random fields has a first order transition as a function of external field $\underset{\sim}{H}$ at $\underset{\sim}{H}_t = 0$, then inclusion of ΔF randomly shifts a local transition field

$$\underset{\sim}{H}_t(x) = -\underset{\sim}{h}(x) \tag{28}$$

Imry and Ma argued that the quenched random fields destroy ferromagnetic long range order for low dimensions. They compared the energy gain of formation of domains of linear scale L lattice spacings in the direction of the random field averaged over L^d lattice points at H = 0

$$\sim L^{d/2}\sigma_h \tag{29}$$

to the cost of forming domain walls in an Ising system

$$\sim L^{d-1} \tag{30}$$

or in an isotropic Heisenberg system

$$\sim L^{d-2} \tag{31}$$

Comparison of terms (29) with (30) or (31) shows that for

$$\frac{d}{2} > d-1 \quad \text{or} \quad \frac{d}{2} > d-2 \tag{32}$$

respectively there will for arbitrarily small σ_h be a length scale L sufficiently great on which it is energetically favourable to

break up into domains of scale L. The critical dimensions so obtained are $d_c = 2$ for an Ising spin and $d_c = 4$ for an anisotropic. The model has been studied in field theory (Imry and Ma 1975, Grinstein 1976, Young 1977) and found to have an upper critical dimensionality $d_u = 6$ and a general shift in critical properties, for the Heisenberg case and $d > 4$, at least, of $d_{pure} \leftrightarrow (d-2)_{random}$. Of course instability of the ferromagnetic ground state does not preclude some other ordered state such as a spin glass. If the dimension is greater than the lower critical dimension for spin glass ordering a spin glass transition might occur at finite temperature. It is not yet clear what the lower critical dimension for spin glass ordering is: Anderson and Pond (1978) argued 3 for vector spin glass and 2 for Ising; Reed, Moore and Bray (1978) 4 for Ising.

The critical dimension defined above is the dimension below which an arbitrarily weak random field causes instability of the ordered ground state. For higher dimensions sufficiently strong random fields can lead to instability on some scale. Aharony and Imry (1978b)proposed that for such dimensions there will be a transition in the ground state as a function of random field strength H_o, where h has probability density function

$$p(h) = \frac{1}{2}\delta(h+H_o) + \frac{1}{2}\delta(h-H_o) \qquad (33)$$

They find a phase diagram in (H_o, T) space as in figure (2.1). If their diagram is correct then considering the three-dimensional parameter space (H_o,T,H) where H is a uniform magnetic field, then we would expect the first order transition as a function of field H for fixed T at H=0 to persist for $T < T(H_o)$ and for $T > T(H_o)$ to be smeared.

The scaling ideas of Imry and Ma may be adapted for application to more usual first order transitions as a function of temperature. The unsmeared transition found in section 2.2 implies an abrupt jump from a homogeneous low temperature phase to a homogeneous high temperature phase at some averaged temperature for the disordered case. The self-consistency of this result will be examined by considering the cost in free energy of forming domains of the opposite phase on a homogeneous phase. This amounts to extending the search for a local minimum in function space of $F_i\{m(x);\psi(x)\}$ by looking at configurations homogeneous with domains of linear scale L. The first point to note is that one expects domain walls of finite thickness and thus an Ising like domain wall energy varying as L^{d-1} as $L \to \infty$. The L^{d-2} dependence take for the isotropic spin case by Imry and Ma depends on the continuous energy-preserving symmetry by which the $H \to 0^-$ and $H \to 0^+$ equilibrium states may be connected. In other words in the ordered phase the transverse correlation length is infinite from the Goldstone theorem (Brézin, Le Guillou, Zinn-Justin 1976). Domain walls then occur with thickness comparable to the scale of the domains. For discrete or anisotropic systems domain walls are restricted to have thickness no more than the correlation length ξ. One would not expect a continuous energy preserving symmetry relating say the low and high temperature phases at the critical temperature for a first order structural phase transition. For weak anisotropy the domain wall energy varies as

$$\begin{aligned} &\sigma_0 L^{d-1} \quad \text{for} \quad L \geq \xi \\ &\sigma_0 \xi L^{d-2} \quad \text{for} \quad L \leq \xi \end{aligned} \tag{34}$$

where σ_o is defined as the surface tension.

Any breaking into domains is driven by the spread in local transition temperature caused by fluctuations in the number of impurities. For a given volume with L^d sites the local transition temperature is the average of L^d random variables and differs from the mean with a distribution of width

$$\frac{\Delta T_c}{T_c} \sim D L^{-d/2} \qquad (35)$$

where D is the fluctuation per unit volume. In the notation of section 2.2 D is the standard deviation of ψ normalised by $a(T_o)$. For instance for a mixture of p species with transition temperature T_1 to (1 - p) of T_2 we have

$$D \sim (p(1-p))^{1/2} \left(\frac{T_1 - T_2}{pT_1 + (1-p)T_2} \right) \qquad (36)$$

The gain is free energy by formation of domains of the high T phase of scale L at T_c for $(\Delta T)_{loc} > 0$ on the low T homogeneous phase is

$$\Gamma \Delta T \sim \Gamma D T_c L^{-d/2} \qquad (37)$$

where Γ is the discontinuity in the temperature derivative of the free energy per unit volume, i.e. the latent heat. The cost of domain formation per site is

$$\frac{\sigma_o L^{d-1}}{L^d} = \frac{\sigma_o}{L} \quad \text{for } L \gtrsim \xi \qquad (38)$$

Comparing (37) and (38) we expect instability with respect to a phase inhomogeneous on length scale L if

$$\Gamma D T_c L^{-d/2} > \frac{\sigma_o}{L}$$

$$D > \left(\frac{\sigma_0}{\Gamma T_c}\right) L^{\frac{d}{2}-1} \qquad (39)$$

For d < 2 the criterion (39) is satisfied for L sufficiently large for arbitrarily small D. The lower critical dimension below which any amount of disorder smears the transition is 2. In three dimensions the sharp discontinuity in the internal energy found in mean field theory will therefore persist for slight disorder. In parts of the crystal there may be improbably large fluctuations in the impurity density allowing isolated regions to transform at a different temperature from the bulk. In this case the internal energy as a function of temperature shows incipient smearing as well as a finite discontinuity with slightly reduced amplitude (see Figure 2.2b). As the amount of disorder increases the smearing increases and the amplitude of the discontinuity decreases until the transition is no longer first order. This is qualitatively as is seen in birefringence measurements on $Rb_xK_{1-x}MnF_3$ as x increases from 0 to 6% (Borsa, Benard, Walker and Baviera 1977).

We can estimate how much disorder is required to smear the transition by noting that within the above criterion for 2 < d < 4 domain formation is most likely on a length scale comparable to the correlation length. This is because for $L \leq \zeta$ the domain wall energy varies as L^{d-2} from (34) and (39) becomes

$$D > \left(\frac{\sigma_0 \xi}{\Gamma T_c}\right) L^{\frac{d}{2}-2} \qquad L \leq \xi \qquad (40)$$

which for $d < 4$ is more likely to be satisfied by L as large as possible, and (39) for $L \geq \zeta$ is more likely to be for L as small as possible. The criterion for the disorder D to smear the transition is

$$D \geq \left(\frac{\sigma_0}{\Gamma T_c}\right) \xi^{d/2 - 1} \qquad (41)$$

If the inequality (41) is not satisfied the transition remains sharp in the presence of disorder but the transition of isolated regions with improbable fluctuations in the degree of disorder may reduce the amplitude of the discontinuity. The proportion by which the discontinuity is reduced may be estimated by considering the probability that a finite volume of the scale of the correlation length has an averaged transition temperature T_{loc} sufficiently far from the mean T_o that it is energetically favourable for it to transform independely in spite of the cost in domain energy. Consider the low temperature homogeneous phase at temperature $T = \bar{T_o}$. A correlation volume with averaged transition temperature T_{loc} gains energy $\Gamma(T_o - T_{loc})$ per site by transforming to the high temperature phase but loses domain energy $\frac{\sigma_o}{\xi}$ per site. It is energetically favourable for it to do so if

$$\Gamma(T_o - T_{loc}) > \frac{\sigma_o}{\xi}$$

$$T_{loc} - T_o < -\frac{\sigma_o}{\xi\Gamma} \qquad (42)$$

T_{loc} is the average of ξ^d random variables: for ξ at least a few lattice spacings it will have a normal distribution with mean T_o and

standard deviation $D\Gamma_c\xi^{-d/2}$. The probability that (42) is satisfied is then given by

$$\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{-N} e^{-x^2/2}\,dx \sim \frac{e^{-N^2/2}}{\sqrt{2\pi}\,N} \quad , N\to\infty$$

where $N = \frac{\sigma_o}{\xi\Gamma}\quad\frac{\xi^{d/2}}{D\Gamma_c}$ is the number of standard deviations from the mean of the upper limit in (42). Thus in the limit $D \to 0$ the first order discontinuity is reduced in amplitude by an amount

$$\frac{2}{\sqrt{2\pi}}\;\frac{\exp\left[-\frac{1}{2D^2}\left(\frac{\sigma_o}{\Gamma T_c}\right)^2\xi^{d-2}\right]}{\left(\frac{\sigma_o}{\Gamma T_c D}\xi^{d/2-1}\right)} \qquad (43)$$

Recently Imry (private communication) has constructed arguments similar to those advanced here with similar conclusions. In particular he considered the case in which variation of a parameter μ changes the first order transition to second order of $\mu = \mu_c$. If exponents are defined such that the correlation length at the first order transition diverges as

$$\xi(\mu) \sim (\mu-\mu_c)^{-\nu} \qquad (44)$$

and the latest heat as

$$\Gamma(\mu) \sim (\mu-\mu_c)^{1-\alpha} \qquad (45)$$

where the exponent is defined to imply $\frac{d\Gamma}{d\mu} \sim (\mu-\mu_c)^{-\alpha}$ in analogy to $\frac{d\Gamma}{dt} = C_u \sim (T - T_c)^{-\alpha}$ and the surface tension varies as

$$\sigma_o \sim (\mu-\mu_c)^{\nu(d-1)} \qquad (46)$$

(46) is obtained by equating the energy of excitation of a domain of linear scale ξ by a thermal fluctuation $kT_c(\mu)$ and assuming as $\mu \to \mu_c$ $kT_c(\mu)$ tends to a finite $kT_c(\mu_c)$. (44), (45), (46) substituted into (41) give the criterion

$$D \gtrsim (\mu - \mu_c)^{\nu(d-1)+\alpha-1-\nu(d/2-1)}$$

$$D \gtrsim (\mu - \mu_c)^{d/2} \qquad (47)$$

provided that hyperscaling $d\nu = 2-\alpha$ is observed. For $\alpha > 0$ an arbitrarily small amount of disorder smears the transition sufficiently close to the second order transition, in accordance with the Harris (1974) argument. For $\alpha < 0$ disorder is irrelevant as $\mu \to \mu_c$.

As an example of this, we consider the line of first order transitions approaching the tricritical point in a three-dimensional metamagnet (Chapter 1). Here $\mu - \mu_c = H - H_t$ and the exponent α for approach along the first order line is the renormalized exponent α_u (in the notation of Toulouse and Pfeuty 1975) satisfying

$$2 - \alpha_u = \frac{2-\alpha_t}{\phi_t} \qquad (48)$$

with ϕ_t the tricritical crossover exponent. In three dimensions critical behaviour is mean field up to logarithmic corrections and $\alpha_t = \frac{1}{2}$, $\phi_t = \frac{1}{2}$, $\alpha_u = -1$. We conclude that in the dilute metamagnet we expect a sharp first order transition close to the tricritical point but possible smearing far below the tricritical point.

The nature of the smeared transition is unclear. If for a smeared transition the system can be thought of as splitting into domains of a characteristic length ξ transforming independently at local transition temperatures, one would expect a form of percolation transition with

a non-analyticity in the specific heat as $\sim (\Delta T)^{\beta_P}$ as suggested by Bergman, Aharony and Imry (1978) for disordered second order phase transitions with $\alpha > 0$. Loss of a first order discontinuity in the free energy may not preclude higher order singularities.

The conclusions of this Chapter are then the following. Via the replica method, mean field theory indicates that in the presence of disorder a first order transition may remain sharp although at a different temperature. Simple scaling ideas lead to the conclusion that in three dimensions mean field conclusions are correct for a small amount of disorder apart from the possibilities of incipient smearing from improbable fluctuations. Increasing disorder gradually decreases the discontinuity until the transition is of higher order at a measure of disorder given by the scaling relation (41).

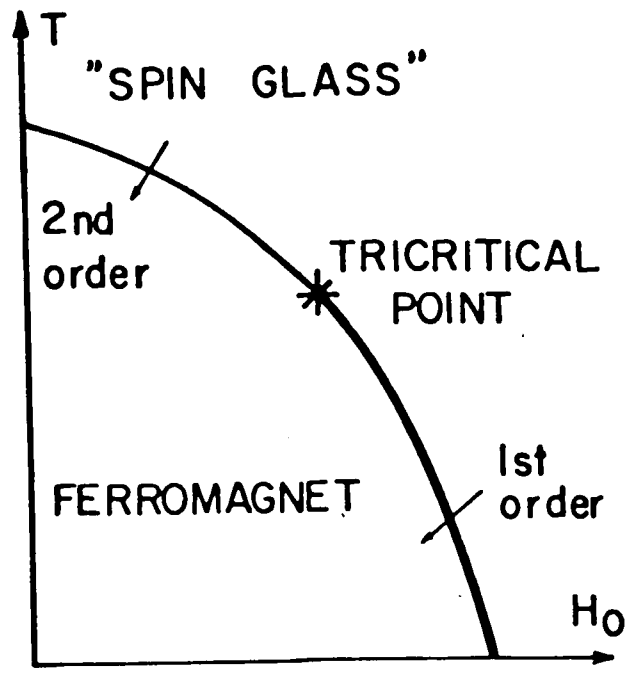


Figure 2.1. Phase diagram for an Ising model in a random magnetic field $\pm H_o$. (Aharony and Imry 1978b).

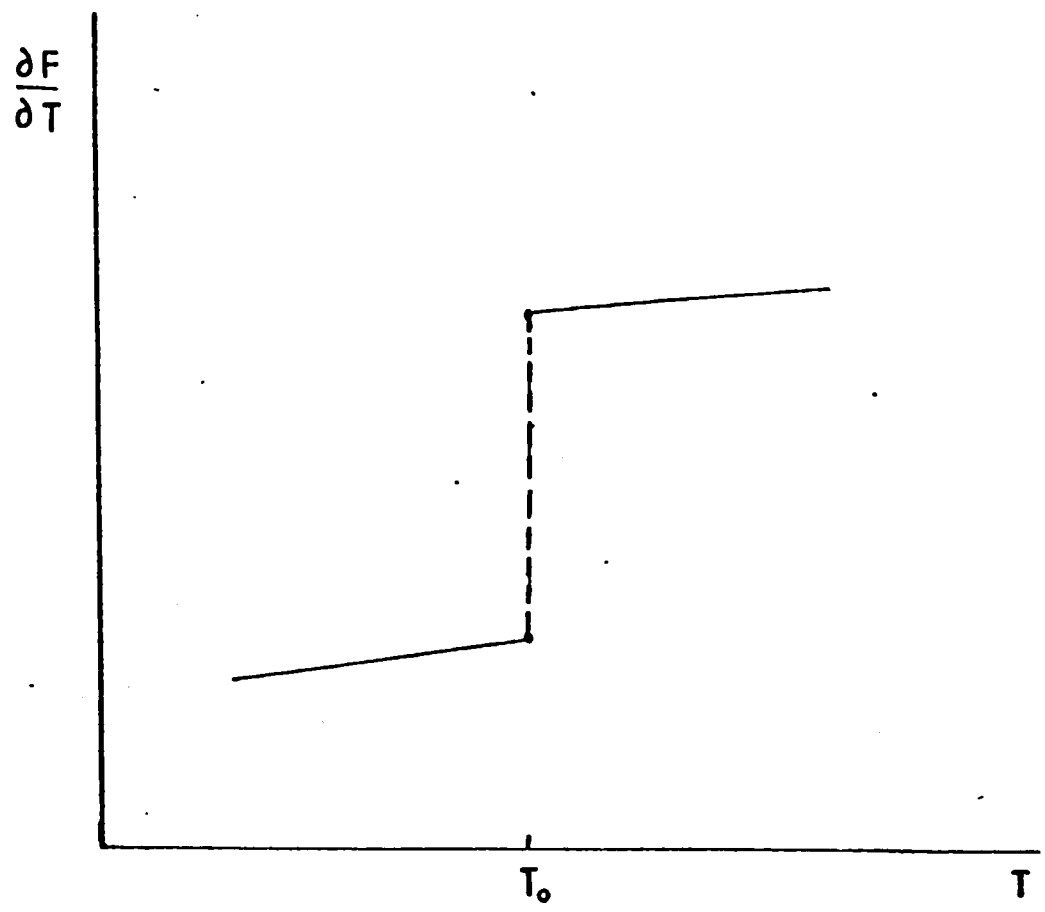


Figure 2.2a. The first derivative of the free energy in a pure crystal with first order transition at $T = T_o$.

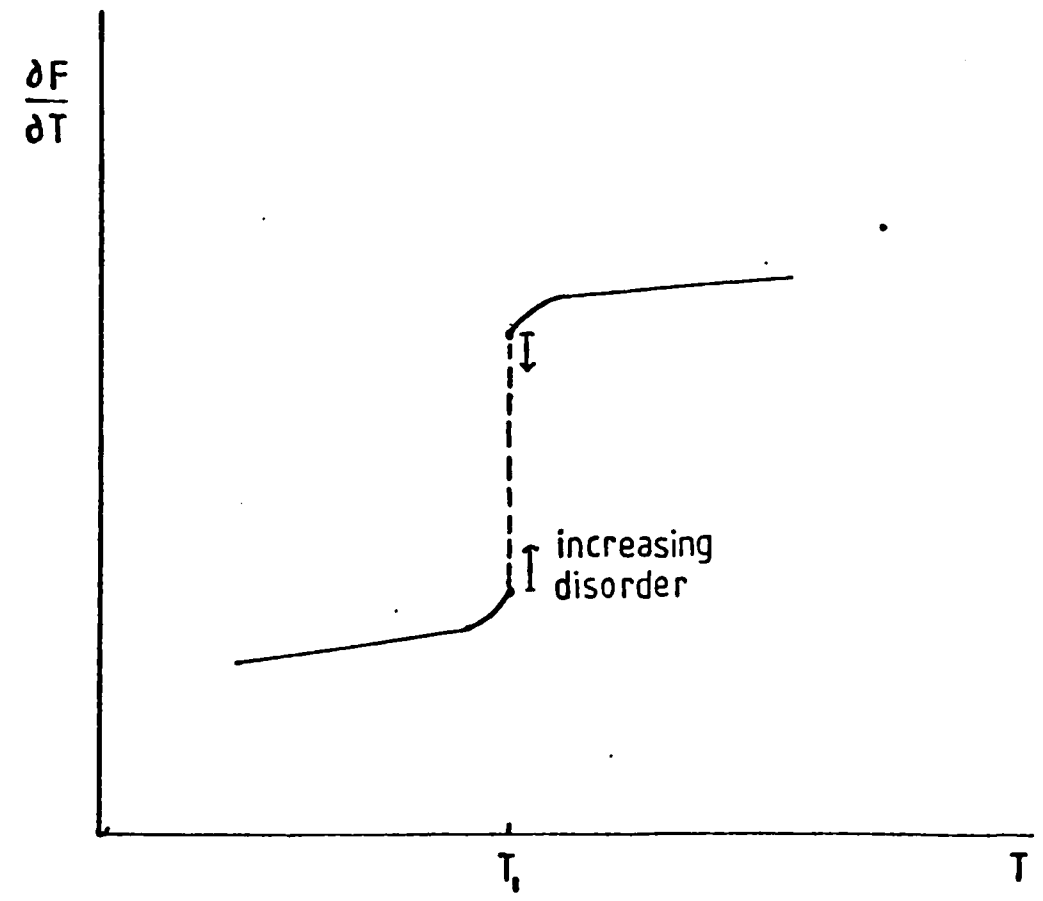


Figure 2.2b. The first order transition in a slightly disordered crystal. The transition temperature T_1 is shifted from T_o and the discontinuity reduced.

CHAPTER 3

EXCITATIONS OF DILUTE MAGNETS NEAR THE PERCOLATION THRESHOLD

3.1 Percolation.

One of the simplest means of introducing disorder into a theory of the solid state, and one that is experimentally accessible, is random dilution. In a magnetic insulator, for instance, this may be achieved by substituting a proportion of magnetic ions randomly by chemically similar but non-magnetic ions. The resolution of the geometry of the resulting structure: a regular lattice in which each site is occupied or unoccupied with probability p or (1-p) respectively constitutes a site percolation problem (see reviews by Shante and Kirkpatrick 1971, Essam 1972). A different but closely related problem is that of bond percolation, in which the bonds between nearest neighbours on a regular lattice are randomly severed. This is a model for a random resistor network: an infinite lattice with neighbouring points connected by conductors, the conductance of each an independent random variable with probability density function

$$f(\sigma) = p\,\delta(\sigma - \sigma_0) + (1-p)\,\delta(\sigma) \qquad (1)$$

In each case, two occupied sites in the dilute lattice are defined to belong to the same cluster if they are connected by a chain of occupied sites or bonds. The size and number of clusters depends on the dilution p. There is a critical p_c below which all clusters are finite; for $p > p_c$ there is a cluster of infinite extent. P(p) is defined to be the

probability that a given occupied site belongs to the infinite cluster, so that $P(p) > 0$ for $p > p_c$. $S(p)$ is the mean size of finite clusters. A "free energy" $K(p)$ is defined as the mean number of finite clusters per site.

Such functions of the random geometry have been studied by series expansions (Essam 1972) and computer simulation (Kirkpatrick 1973) as well as analytically on a Cayley tree (Essam 1972) and on some two-dimensional lattices for p_c. It is concluded that at p_c $S(p)$ diverges as $(p-p_c)^{-\gamma_p}$, $P(p)$ vanishes as $(p-p_c)^{\beta_p}$, thereby defining exponents β_p and γ_p and suggesting analogy to the singular forms of thermodynamic quantities in a system near the critical temperature for a second order phase transition.

Such an analogy was made formal by Kasteleyn and Fortuin (1969) who showed that the bond percolation problem corresponds to the $s \to 1$ limit of an s-state Potts model with Hamiltonian

$$\mathcal{H}^s = -J \sum_{\langle ij \rangle} s(\delta_{n_i n_j} - 1) - h \sum_i s(\delta_{n_i 1} - 1) \tag{2}$$

with

$$P(p) = \lim_{s \to 1} \frac{1}{s-1} \left. \frac{\partial G}{\partial h} \right|_{h=0=s-1} \tag{3}$$

$$S(p) = \lim_{s \to 1} \frac{1}{s-1} \left. \frac{\partial^2 G}{\partial h^2} \right|_{h=0=s-1} \tag{4}$$

where $1-p = e^{-\beta J}$; G the free energy per site. This Hamiltonian formulation was used to study the critical properties of the percolation problem by renormalization group techniques: including expansions about 6 dimensions, above which exponents are mean field like. (Harris, Lubensky,

Holcomb and Dasgupta 1975). From the Hamiltonian formulation and the usual renormalization group ideas one expects power law exponents in the above quantities with exponents satisfying scaling relations and universality hypotheses such as equality of bond and site exponents and dependence of exponents on dimension and not on lattice symmetry. A pair connectedness correlation length $\xi(p)$ diverges at p_c as

$$\xi(p) \sim |p - p_c|^{-\nu_p} \tag{5}$$

$K(p)$, analogous to the free energy, has a singularity $K(p) \sim (p-p_c)^{2-\alpha_p}$. Renormalization group methods have also been applied without recourse to the Hamiltonian formulation by rescaling probabilities, as in Young and Stinchcombe (1975).

Table 3.1 presents values for the percolation exponents defined here, as quoted by Harris and Kirkpatrick (1977) as the most accurately determined values, with scaling relations assumed as indicated.

Table 3.1

Exponent	Definition	2D	3D	Mean Field ($d > 6$)
β_p	$P(p)\sim(p-p_c)^{\beta_p}$	0.14 ± 0.005	0.39 ± 0.005	1
γ_p	$S(p)\sim\|p-p_c\|^{-\gamma_p}$	2.3 ± 0.1	1.8 ± 0.05	1
α_p	$K(p)\sim\|p-p_c\|^{2-\alpha_p}$	-0.65 ± 0.1	-0.58*	-1
ν_p	$\xi(p)\sim\|p-p_c\|^{-\nu_p}$	1.29	0.86*	$\frac{1}{2}$

*assuming scaling relations $\alpha_p + 2\beta_p + \gamma_p = 2$ or $d\nu_p = (2\beta_p + \gamma_p)$.

Naturally there is more to the geometry than the quantities defined above; in particular attention has recently been paid to the distribution of finite clusters and their perimeters just below the percolation concentration, for which proposed scaling hypotheses have been tested by computer simulation (Leath 1976, Leath and Reich 1976). This chapter essentially concerns geometric features of the incipient infinite cluster just above the percolation threshold and their physical consequences.

Apart from functions of the geometry one can define, for example, the conductance Σ of the random resistor network, vanishing at p_c as $(p-p_c)^{\sigma}$. Σ was measured experimentally and compared to an effective medium theory with good agreement except at the percolation threshold (Watson and Leath 1974). A conductivity Σ_o was calculated exactly on a Bethe lattice by Stinchcombe (1974), although later work (de Gennes, 1976, Stephen 1978) suggests that the calculated Σ_o corresponds to a "microscopic" conductivity rather than the macroscopic conductivity Σ and is closer to the spin wave stiffness in a finite dimensional lattice. Dasgupta, Harris and Lubensky (1978) and Stephen (1978) derived Hamiltonian formulations for the random resistor network and found the critical exponent for dimensions near six, the upper critical dimension. In lower dimensions the critical behaviour $\Sigma(p)$ has been studied by real space scaling (Stinchcombe and Watson 1976) and computer simulation (Kirkpatrick 1973).

In principle if the geometry is completely determined Σ should be calculable. Conversely simple models of the geometry can be tested by calculating Σ. The simplest idea was that the effect of dilution is simply to remove the contribution of finite clusters to the conductivity.

This would then imply that

$$\Sigma(p) = D P(p) \qquad (6)$$

where D is constant. Relation (6) disagrees with results obtained as above, implying that the geometry of the infinite cluster is not simply that of a pure crystal of reduced volume. Disagreement is particularly marked at the percolation threshold, where in three dimensions for example, P(p) vanishes with infinite slope, $\beta < 1$, while Σ vanishes with zero slope, $\sigma > 1$. This disagreement is the first motivation for considering scaling properties of the percolating cluster, as will be discussed in the next section (3.2).

Another physical quantity related to Σ is defined as the resistance of a random network of superconductivity links and links of finite resistance(Straley 1976), i.e. a random resistor network in which the resistance of each link is a random variable with probability density function

$$f(R) = p\delta(R) + (1 - p)\delta(R - R_o) \qquad (7)$$

In this case, for $p > p_c$ the resistance of the network is zero and for $p < p_c$ the conductance diverges as $\Sigma' \sim (p - p_c)^{-s}$. Stephen's (1978) field theory allows calculation of s near 6 dimensions. The Hall coefficient of a resistor network was predicted from a scaling picture of the infinite cluster to show a power law divergence in three dimensions by Skal and Shklovskii (1974).

This chapter concerns the dynamics and thermodynamics of dilute Heisenberg magnets with nearest neighbour ferromagnetic or antiferromagnetic exchange. The interest of such systems is that crystals such as $Rb_2Mn_pMg_{1-p}F_4$, a two-dimensional isotropic antiferromagnet (Birgeneau et al.

1976) and $Mn_pZn_{1-p}F_2$, a three-dimensional anisotropic antiferromagnet (Cowley et al. 1977) have been studied experimentally for a range of dilutions including values near percolation. The percolation threshold is particularly interesting because it is there that mean field theories such as the Coherent Potential Approximation, which give good results well above percolation (see Elliott, Krumhansl, Leath 1974), fail to reproduce correct singularities.

The existence of an infinite cluster is vital to the ordering of a magnetic structure as it is only in the thermodynamic limit that a ground state of broken symmetry is stable. Series expansions by Elliott, Heap, Morgan and Rushbrooke (1960) showed that the critical temperature of both Ising and Heisenberg ferromagnets tend to zero as $p \to p_c$ from above. More recently Stanley et al. (1976) and Stauffer (1975) have considered scaling theories in which the percolation point is considered as a multicritical point in the space of thermodynamic variables and dilution parameter p. The way in which the critical temperature $T_c(p)$ tends to zero is determined in formal terms by a crossover exponent from the geometrically driven percolation transition to the thermally driven critical transition.

The aim of this chapter is to discuss how observations of thermodynamic correlations coupled with the scaling behaviour of Σ and other quantities such as the constants in the low energy dispersion relations in the ferro- and antiferromagnets can be used to suggest simple geometric models of the percolating cluster. The scaling laws derived are expressions of the implications of different geometries which may then be tested by inclusion of exponents from more rigorous theories and experiment. The virtue of developing such models of the geometry is that they may then be applied to

predict, with relative ease, the scaling behaviour of the excitation spectrum, susceptibilities and thermodynamic correlations for which a more formal calculation would be difficult. These applications are treated in sections 3.5 and 3.6, both for the simplest model, as employed by Lubensky (1977) for some aspects of the thermodynamics, and for generalisations found desirable in sections 3.2 and 3.3. In Chapter 4 the same geometrical ideas are used to discuss localisation of excitations near the percolation threshold - again a difficult subject to treat by more rigorous means.

3.2 Geometric Model of the Percolating Cluster.

Just above the percolation threshold the infinite cluster is weakly connected in the sense that removal of a further small proportion of magnetic sites splits it into finite clusters. A model of the percolating cluster has been introduced (de Gennes 1976, Skal and Shklovskii 1974) emphasising this property. The infinite cluster (see Figure 3.1) is considered as a superlattice of small clusters set on nodes of typical spatial separation the correlation length $\xi(p) \sim (p - p_c)^{-\nu_p}$ and connected by random paths of length typically longer

$$L \sim (p - p_c)^{-\zeta} \qquad (8)$$

The paths are random but may not intersect other parts of the infinite cluster and consequently are shorter than self avoiding walks of the same endpoint separation $\xi(p)$. This implies that L is restricted to

$$\xi(p) \leq L \leq \xi(p)^{1/\nu_s} \qquad (9)$$

where ν_s is the self avoiding walk exponent for the dimension of the lattice. Inequality (9) restricts ζ to the range

$$\nu_p \leq \zeta \leq \nu_p/\nu_s \tag{10}$$

Stanley et al. (1976) have discussed a number of indications both experimental and theoretical that ζ should be taken to equal to the upper limit. If we accept this, Stanley's "Self Avoiding Walk Ansatz", then ζ is expressed in terms of numerically well-defined parameters. The value ν_p/ν_s will be denoted by ζ_{SAW}.

The highly simplified model of figure 3.1 will now be used to predict scaling relations in order to test how good an approximation to the actual infinite cluster it is. Comparison to more rigorous results from renormalization group, computer simulation and experimental results will suggest generalizations.

de Gennes introduced the model in a discussion of the conductivity of a randomly diluted resistance network. The resistance of the model network would be

$$\Sigma = \frac{1}{L}\xi^{2-d}$$

implying that the conductivity vanishes as

$$\Sigma \sim (p - p_c)^{\sigma} = (p - p_c)^{\zeta+(d-2)\nu_p} \tag{11}$$

This gives a scaling relation

$$\sigma = \zeta + (d-2)\nu_p \tag{12}$$

We define ζ_r as the exponent determined when independent values of σ and ν_p are substituted into (12).

The model infinite cluster of figure 3.1 has itself a percolation probability r(p) tending to one as $p \to p_c$ with leading term, after a short calculation,

$$1 - r(p) \sim (p - p_c)^{\zeta} \text{ as } p \to p_c \qquad (13)$$

This relation, with r(p) for the actual infinite cluster defines a value for ζ denoted $\zeta = \zeta_{perc}$.

The thermodynamic multiscaling hypothesis (Lubensky 1977) is that the free energy in the neighbourhood of $p = p_c$ and $T = 0$ varies as

$$G(t,p) = (p - p_c)^{2-\alpha_p} f_d \left(\frac{\xi_1(T)}{(p-p_c)^{-\zeta}} \right) \qquad (14)$$

where $f_d(r)$ is the free energy for a d-dimensional system with thermodynamic correlation length r and $\xi_1(T)$ is the one-dimensional correlation length. Clearly this form is consistent with the model cluster of figure 3.1. The form (14) predicts critical temperatures vanishing as

$$KT_c \sim \frac{2J}{\zeta_{MI} \ell n |p-p_c|} \qquad (15)$$

$$KT_c \sim J(p - p_c)^{\zeta_{MH}} \qquad (16)$$

for Ising and Heisenberg spin systems. The values of ζ defined by (15) and (16) are denoted ζ_{MI} and ζ_{MH}.

We have defined four exponents ζ_r, ζ_{perc}, ζ_{MI} and ζ_{MH} which we should expect to be equal if our model lattice had all the essential features of the geometry of the infinite cluster. If we accepted the Self Avoiding

Walk Ansatz they would all equal ζ_{SAW}. Values quoted for ν_p, ν_s and ζ_{SAW} are shown in table 3.2. Unfortunately there are strong indications that the ζ's are not all equal and in two dimensions may not all obey the inequality (10).

Table 3.2

	d=2	d=3	Mean Field(d $\geq$ 6)
ν_p^*	1.29	0.86	$\frac{1}{2}$
$\nu_s^\dagger$	.75	.60	$\frac{1}{2}$(d $\geq$ 4)
ζ_{SAW}	1.72	1.43	1

*From Harris and Kirkpatrick 1977 (assuming scaling relations)

†From McKenzie 1976.

As already mentioned Stanley et al. (1976) have advanced plausibility arguments favouring the Self Avoiding Walk Ansatz and experimental evidence in $Rb_2Mn_pMg_{1-p}F_4$ is in accord with this, while that in $Mn_pZn_{1-p}F_2$ is less clear-cut. It must be emphasised that the self avoiding walk nature of correlations in $Rb_2Mn_pMg_{1-p}F_4$ has been found below percolation, we assume that it will persist above percolation as in the Self Avoiding Walk Ansatz. With this qualification the experimental evidence is taken

as evidence that $\zeta_{MH} = \zeta_{SAW}$. From computer simulations of the conductivity Kirkpatrick (1976) quotes values $\zeta_r = 1.1 \pm 0.1$ and 0.7 ± 0.1 in two and three dimensions in agreement with renormalization group results in two dimensions by Stinchcombe and Watson (1976) $\zeta_r = 1.13 \pm 0.09$, and experimental results on ZnO/ZnS powder mixtures in three dimensions $\zeta_r = 0.8 \pm 0.2$ (Clarke, Orton and Guest 1978). There is some doubt however as recent series expansions on an equivalent field theory for $p < p_c$ suggest different values $\zeta_r = 1.43 \pm 0.03$, 1.12 ± 0.2 (Fisch and Harris 1978). ζ_{perc} is easily seen to be one in all dimensions by considering successive dilution of a pure crystal first by p and then by

$$p_c/p = \frac{p_c}{p_c+p-p_c} \simeq 1 - \frac{p-p_c}{p_c} \qquad (17)$$

to the percolation point. Comparison with (13) gives $\zeta_{perc} = 1$.

In two dimensions ζ_{MI} is also 1 from the rigorous result of Bergstresser (1978). Furthermore ζ_{MI} is 1 in all dimensions $1 \leq d \leq 6$ to all orders in perturbation theory from the field theory of the $s \to 1$ Potts model. (Stephen and Grest 1977, Wallace and Young 1978).

Resolution of these difficulties within a geometrical model lies in considering the effects of sites in the infinite cluster not included in the simple model of figure 3.1. The model cluster of figure 3.1 has a total volume per unit volume of the undiluted crystal

$$P^s(p) \sim (p - p_c)^{\beta_s} = (p - p_c)^{d\nu_p - \zeta}$$

$$\beta_s = d\nu_p - \zeta \qquad (18)$$

The actual infinite cluster however has volume proportional to $P(p) \sim (p - p_c)^{\beta_p}$. If we take $\zeta = \zeta_{SAW}$, β_s and β_p are as shown in table 3.3. The model cluster includes only a fraction of the number of sites of the infinite cluster. Extra sites may occur as dangling bonds or parallel paths. Such features affect the quantities considered above in different ways, as we shall see in the following section.

Table 3.3

	d=2	d=3	Mean Field($d \geq 6$)
β_p^*	0.14	0.39	1
$\beta_s^\dagger$	0.9	1.1	2

*From Harris and Kirkpatrick 1977.

†$d\nu_p - \zeta_{SAW}$ as in Table 3.2.

3.3 Scaling Relation for the Spin Wave Stiffness.

We have seen that de Gennes' scaling (12) for the conductivity exponent σ implies a value for ζ_r in disagreement with other values of ζ. Here we derive a new scaling relation for σ and discuss the geometric significance of the differences with (12).

The conductivity of a resistor network $\Sigma(p)$ is related to the spin wave stiffness $D(p)$ of spin waves in the dilute isotropic ferromagnet by (Harris and Kirkpatrick 1977).

$$\Sigma(p) = D(p)P(p) \tag{19}$$

Thus if we define the exponent for spin wave stiffness by

$$D(p) \sim (p - p_c)^t \tag{20}$$

then we have the exact result

$$t = \sigma - \beta_p \tag{21}$$

Now we shall derive a scaling relation for t and hence σ by considering spin wave excitations on the model lattice. As far as the equations of motion for spin waves are concerned, the model lattice of fig. 3.1 is equivalent to a superlattice with nodes separated by straight paths of length L as in fig. 3.2. This lattice has low frequency excitations with dispersion

$$\begin{aligned} \omega(Q) &= J \frac{L+1}{2L} Q^2 \qquad Q \to 0 \\ &\simeq \frac{J}{2} Q^2 \end{aligned} \tag{22}$$

where Q is the momentum. The momentum q of a spin wave in the model lattice of fig. 3.1 is related to Q by

$$q = \begin{cases} Q_c^{\nu_p/\zeta} \, Q/Q_c & q < q_c = \frac{2\pi}{\xi} \sim (p - p_c)^{\nu_p} \\ Q^{\nu_p/\zeta} & q \geq q_c \end{cases} \tag{23}$$

since for distances L' less than $L \sim (p - p_c)^{-\zeta}$ the spin wave propagates by a randomly directed path travelling a Euclidean distance $L'^{\nu_p/\zeta}$. Since $Q_c^{\nu_p/\zeta}/Q_c \sim (p - p_c)^{\nu_p - \zeta}$, the dispersion relation in terms of the measured momentum becomes

$$\omega(q) = \frac{J}{2} Q^2$$

$$= \begin{cases} \frac{J}{2} q^{2\zeta/\nu_p} & q \geq q_c \text{ but small} \\ \frac{J}{2}(p - p_c)^{2(\zeta-\nu_p)} q^2 & q \leq q_c \end{cases} \qquad (24)$$

The dispersion (24) is of the "hydrodynamic" form proposed by Stauffer (1976)

$$\omega(q) = q^2 f(q\xi)$$
$$f(y) \sim y^{z-2} \quad y \to \infty$$
$$f(0) = D(p) \qquad (25)$$

but here the "dynamical exponent" takes the value $z = 2\zeta/\nu_p$ and is interpreted geometrically. Thus the spin wave stiffness is given by

$$D(p) = \frac{J}{2}(p - p_c)^{2(\zeta-\nu_p)} \qquad (26)$$

and we find scaling relations

$$t = 2(\zeta - \nu_p) \qquad (27)$$

$$\sigma = 2(\zeta - \nu_p) + \beta_p \qquad (28)$$

(28) is a new scaling relation, different from (12). If we take the value $\zeta = \zeta_{SAW}$ that may be appropriate for a continuous spin system (28) gives values for σ of 1.00 and 1.53 in 2 and 3 dimensions in good agreement with Kirkpatrick's results.

An apparent difficulty with (28) is that for mean field values of ζ, ν_p and β_p it implies $\sigma = 2$ rather than the mean field value $\sigma = 3$ (Stephen 1978). Conversely the de Gennes relation with $\zeta = \zeta_{SAW}$ gives correct mean field behaviour but fails in dimensions 2 and 3. Let us analyse where (28) and (12) differ. The model network has volume vanishing as $P^s(p) \sim (p - p_c)^{\beta_s}$. If we substitute $\beta_s = d\nu_p - \zeta$ from (18) and $t = 2(\zeta - \nu_p)$ into (28) we obtain $\sigma = \zeta + (d - 2)\nu_p$, the de Gennes relation (12). The difference between (28) and (12) is then that by using β_p in (28) we allow the sites in the infinite cluster not included in the simple model of fig. 3.1 to contribute to the conductivity whereas in (12) they do not. The relative successes of the different scaling relations in different dimensions leads us to a generalised geometric model with introduction of a dimension d^*, $3 < d^* < 6$, at which the geometry changes qualitatively. This is shown in Figure 3.3 and is as suggested by Dasgupta et al. (1978). For dimensions $d < d^*$ there is a diverging number of random paths between each pair of nodes of the superlattice, whereas for $d > d^*$ the extra sites contribute to dangling paths. It seems natural to take $d^* = 4$ since for dimensionalities above 4 the self avoiding walk model has mean field exponents and excluded volume effects are unimportant. (de Gennes 1972). Branchings from paths connecting nodes of the cluster are therefore unlikely to encounter other parts of the infinite cluster above four dimensions and do not contribute to the conductivity. Dead end branchings do, however, affect the spin wave

stiffness. For example, consider the dispersion relation for a linear ferromagnetic chain with branches of length n as in Figure 3.4. This chain has spin wave stiffness

$$D_n = \frac{J}{(n+1)} \qquad (29)$$

where J is the interaction strength, as can be seen by solving the coupled equations of motion or from the relation $\Sigma = DV$. By comparison of the chain without branches n = 0

$$V_n = (n+1)V_o$$

$$\Sigma_n = \Sigma_o \qquad (30)$$

leading to (29). Failure of the scaling relation (27) in high dimensions indicates a diverging number of sites in dangling paths in high dimensionality.

We conclude therefore that it is possible to reconcile observed exponents for conductivity with the Self Avoiding Walk Ansatz by generalising the geometric model slightly as in Figure 3.3. The spin wave stiffness and dynamic exponent are given geometrical interpretations in terms of the spread of local correlations,

3.4 Scaling Relations for Anisotropic Spins.

We have seen what geometrical conclusions may be drawn from the values of ζ_r and ζ_{MH}. It remains to interpret the value $\zeta = 1$, apparently true for all dimensions for ζ_{perc} and ζ_{MI}. The existence of parallel linkage as in figure 3.3a does not affect ζ_{perc}: it merely implies logarithmic corrections to the form (13). Anisotropic spin systems and

dilution properties are sensitive to different geometric features from conductivity and isotropic spin correlations. Consider the chain in figure 3.5, a double chain with one of every nth pair of horizontal bonds missing (with n = 6 in figure 3.5). The conductivity of such a chain is that of a single chain of the same length and conductivity per unit length

$$\sigma' = 2\sigma \frac{n}{n+1} \tag{31}$$

Calculating the probability r(n) of breaking a chain of length n given a probability r of breaking each horizontal link we find

$$1 - r(n) = (1 - r^2)^{n-1}(1 - r) \tag{32}$$

Thus for fixed n, $r \to 0$ the chain is equivalent to one of length 1/n, or in other words for calculation of percolation probabilities the single links dominate. When we consider spin correlations along such a chain isotropic spin systems show the same dependence on single or multiple linkage as the conductivity and anisotropic spin systems the same as percolation, as expected from the radiation $\Sigma = DV$ and formal analogies between Ising and percolation problems (e.g. Coniglio et al. 1977), or from the fact that the percolation problem can be expressed as the $s \to 1$ limit of the (discrete) s-state Potts model. Intuitively this is because of the forms of the correlation functions as $T \to 0$

$$\xi_{IS}(T) = e^{2J/T}$$

$$\xi_{H}(T) = 2J/T \tag{33}$$

In a low temperature approximation, two parallel bonds may be considered equivalent to a single bond of twice the strength. Correlations in the

two cases over n links, (n - 1) of which are double as in Figure 3.5 decay as

$$\langle S(0)S(n)\rangle \simeq (1 - e^{-2J/T})(1 - e^{-4J/T})^{n-1}$$

$$\simeq (1 - e^{-2J/T}) \qquad (34)$$

$$\langle S(0)S(r)\rangle \simeq (1 - T/2J)(1 - T/4J)^{n-1}$$

$$\simeq \left(1 - \frac{T^{\frac{n+1}{n}}}{4J}\right)^{n} \qquad (35)$$

Thus Ising (or anisotropic) spin correlations are much more sensitive to finite parallelisms along a chain than are continuous spins.

It may then be possible to interpret $\zeta_{perc} = \zeta_{IS} = 1$ as the exponent for the divergence of the number of single links in the one-dimensional paths with finite width as in Figure 3.1 or 3.3 which have total length diverging with ζ_{SAW}.

3.5 Excitations of the Dilute Isotropic Ferromagnet.

We have seen that it is possible to construct a geometric model of the percolating cluster that is consistent with the Self Avoiding Walk Ansatz and observed exponents. The advantage of this is to suggest at least qualitatively the behaviour of other physical quantities that might be difficult to calculate more rigorously. From now on, as we shall be concerned with isotropic spins, we shall use ζ without subscript to denote ζ_{SAW}.

Consider first the excitation spectrum of spin waves from the ground state of a dilute Heisenberg spin system with isotropic ferromagnetic exchange between nearest neighbours. For the simplest model system (of figure 3.1) the density of spin wave modes is, omitting constant factors

$$\rho(\omega) = \begin{cases} \dfrac{\omega^{\frac{d-2}{2}}}{2(D(p))^{d/2}} & \omega << \omega_c \\ \dfrac{c(p)}{\omega^{1/2}} & \omega_c << \omega << J \end{cases} \qquad (36)$$

where ω_c is the spin wave frequency of an excitation of wavelength L as seen along the random chain

$$\omega_c = J(p - p_c)^{2\zeta} \qquad (37)$$

For wavelengths shorter than the node-node distance the lattice has the same spectrum as a collection of weakly coupled one-dimensional paths; only at long wavelengths is the fully d-dimensional nature of the lattice apparent. C(p) is determined from the condition that the integrated density of states equal the total number of sites

$$\int_0^{\omega_f} \rho(\omega)d\omega \sim P^s(p) = (p - p_c)^{d\nu} p^{-\zeta} \qquad (38)$$

$$C(p) \sim (p - p_c)^{d\nu} p^{-\zeta}/\omega_f^{\frac{1}{2}} \qquad (39)$$

where $\omega_f = 4J$ as $p \to p_c$. Substituting the scaling forms (26) and (39) into (36) shows that the different asymptotic forms for $\rho(\omega)$ match at ω_c. Naturally even for the simple lattice of figure 3.1 (36) is only approximate near ω_c and should be understood as a local average density

with band structure details smoothed. As a model of a random percolating cluster this may not be a disadvantage.

In section 3.2 we saw that the simple model of fig. 3.1 can include only a small proportion of the sites in the percolating cluster, consequently the density of states (36) is a fraction of the true density. Let us consider a density of states functionally as in (36) but with modified coefficients. Equating the integrated density of states to the total number of sites implies

$$C(p) \propto P(p)/\omega_f^{\frac{1}{2}}$$

$$\rho(\omega) = \frac{P(p)}{(\omega\omega_f)^{\frac{1}{2}}} \qquad \omega_c << \omega << J \tag{40}$$

This is of the form

$$\rho(\omega) = P(p)\rho_{1d}(\omega) \tag{41}$$

corresponding to the view that short wavelength modes propagate by effectively one-dimensional chains. The low frequency density is assumed unchanged

$$\rho(\omega) = \frac{\omega^{\frac{d-2}{2}}}{2(D(p))^{d/2}} \tag{42}$$

corresponding to the uniqueness of the zero energy mode. For D(p) given by (26) as is appropriate in three dimensions, $\rho(\omega)$ now shows a discontinuity at ω_c, emphasising the point that at $\omega \simeq \omega_c$ $\rho(\omega)$ is only to be understood in an averaged sense. It is here that we differ from Shender (1976) who made the assumption that the functional form of $\rho(\omega)$ for $\omega >> \omega_c$ was of a power law with exponent determined by matching

the asymptotic forms of $\rho(\omega)$ at ω_c.

Once $\rho(\omega)$ is known the critical temperature T_c at which long range order is destroyed by excitation of spin waves may be estimated by comparing the number of thermally excited spin waves with independent Bose distributions to the total number of spin sites

$$\int_0^\infty \frac{\rho(\omega)}{e^{\frac{\omega}{K_B T}} - 1} d\omega \sim P(p) \qquad \text{at } T = T_c \tag{43}$$

giving

$$KT_c \sim \frac{P(p)}{\int_0^\infty \frac{\rho(\omega)}{\omega} d\omega} = \frac{P(p)}{\frac{1}{d-2}\rho(\omega_c^-) + 2\rho(\omega_c^+)} \tag{44}$$

In the limit $p \to p_c$ (40) and (42) imply

$$KT_c \sim (\omega_f \omega_c)^{\frac{1}{2}} \sim J(p - p_c)^{\zeta} \tag{45}$$

agreeing with Lubensky's (1977) argument comparing correlation length to the one-dimensional chain length.

The low temperature form of the specific heat of the infinite cluster is also sensitive to the density of states

$$C_{inf}(T) = \frac{d}{dT}\int_0^\infty \frac{\rho_{inf}(\omega)\omega}{e^{\frac{\omega}{K_B T}} - 1} d\omega \tag{46}$$

For

$$T << \omega_c \quad C_{inf}(T) \propto \frac{1}{(D(p))^{d/2}} T^{d/2} \tag{47}$$

Thus the d-dimensional contribution to the specific heat has a diverging

coefficient $\frac{1}{(D(p))^{d/2}}$ in agreement with Shender (1976). Finite clusters do not contribute as they have an excitation gap. For temperatures T $\omega_c << T << \omega_c^{\frac{1}{2}}$ $C_{inf}(T)$ has the form (in disagreement with Shender)

$$C_{inf}(T) \sim (k_B T/J)^{\frac{1}{2}} \tag{48}$$

The spin wave stiffness also appears in the expression for the parallel susceptibility for low temperatures

$$\chi_{||}(T) \sim K_B T \left(\frac{h_{||}}{D(p)}\right)^{\frac{d-4}{2}} \qquad h_{||} \to 0,\ T \to 0 \tag{49}$$

So far there has been no consideration of the effects of disorder while the percolating cluster is highly disordered. One effect of disorder is to modify the low frequency form of $\rho_{1d}(\omega)$ appropriate to (41) from the pure $\omega^{-\frac{1}{2}}$ behaviour. It will be argued in Chapter 4 that the appropriate singularity for a disordered chain of spins is

$$\rho_{1d}(\omega) \sim \frac{1}{\omega(\ell n \omega)^{r+2}} \qquad r > 0 \tag{50}$$

If this form is substituted into condition (43) for the loss of long range order the critical temperature is lowered to

$$kT_c \sim \omega_c(\ell n \omega_c)^{r+2} \tag{51}$$

It is also argued in Chapter 4 that disorder will localise most of the excitations in the percolation cluster. If the loss of long range order is

associated with the excitation of mainly localised modes this may affect the sharpness of the phase transition.

3.6 Excitations of the Dilute Isotropic Antiferromagnet.

The dilute magnets that have been studied experimentally have been almost all antiferromagnetic. We therefore consider the arguments for spin wave spectrum and thermodynamics for isotropic antiferromagnets. As for the ferromagnet we discuss first the dispersion relation and excitation spectrum for the simple model of figure 3.1 and then modifications in including a model for the full cluster as in figure 3.4. Finally the important effects of fluctuations in the geometry on the dispersion and perpendicular susceptibility are studied.

The linear coefficient of spin wave dispersion B(p) for the model lattice of figure 3.1 is determined by matching

$$\omega(q) = \begin{cases} B(p)q & q \leq q_c \sim (p - p_c)^{\nu_p} \\ J|\sin q^{\zeta/\nu_p}| & q \geq q_c \end{cases} \qquad (52)$$

at q_c, giving

$$B(p) \sim J(p - p_c)^{\zeta - \nu_p}$$
$$\sim J(p - p_c)^{s} \qquad (53)$$

and B(p) vanishes with exponent

$$s = \zeta - \nu_p = t/2 \qquad (54)$$

The relation (54) will be modified by fluctuations as we shall see later. The density of states is determined for the model as before:

$$\rho(\omega) = \begin{cases} \dfrac{\omega^{d-1}}{B(p)^d} & \omega << \omega_o \\ (p - p_c)^{\beta_s}/(1 - (\omega/J)^2)^{\frac{1}{2}} & \omega_o << \omega \end{cases} \qquad (55)$$

with

$$\omega_o = B(p)q_c \sim J(p - p_c)^{\zeta} \qquad (56)$$

Inclusion of the rest of the sites of the infinite cluster may modify the density of states to

$$\rho(\omega) = \begin{cases} \dfrac{\omega^{d-1}}{B(p)^d} & \omega << \omega_o \\ P(p)\rho_{1d}(\omega) & \omega >> \omega_o \end{cases} \qquad (57)$$

Note that we differ again from Shender (1978) who assumed that $\rho(\omega)$ could be matched at ω_o to determine the functional form for frequencies $\omega >> \omega_o$. The critical temperature $K_B T_c$ for the destruction of long range order is determined by

$$\int_0^{\omega_f} \frac{\rho(\omega)d\omega}{\gamma(\omega)(e^{\omega/K_BT} - 1)} \sim P(p) \qquad (58)$$

where $\gamma(\omega)$ is a factor for the Bogoliubov transformation varying as ω for $\omega \to 0$ (for example see Kittel 1963). Substituting in the density of states (57) gives

$$K_BT_c P(p) \frac{\rho_{1d}(\omega_o)}{\omega_o} \sim P(p) \qquad (59)$$

$$K_BT_c \sim \omega_o \qquad (60)$$

since $\rho_{1d}(\omega_o)$ is constant for this regular model as $\omega_o \to 0$. If (56) is satisfied we recover

$$\frac{J'}{K_BT_c} \sim (p - p_c)^{-\zeta} \qquad (61)$$

in agreement with the simple correlation length argument of Lubensky (1977).

The low temperature specific heat of the percolating cluster is given by

$$C_{inf}(T) = \frac{d}{dt} \int_0^{\omega_f} \frac{\rho_{inf}(\omega)}{e^{\omega/K_BT} - 1} \omega d\omega$$

$$\propto \frac{T^d}{(B(p))^d} \qquad (62)$$

as given by Shender (1978).

Let us now consider fluctuation effects. Random dilution of the antiferromagnet dilutes the ground state sublattices independently; locally then there will be a net magnetic moment. It is this that causes the perpendicular susceptibility of the infinite cluster to diverge in three dimensions as

$$\chi_{\perp}(p) \sim (p - p_c)^{-\tau} \qquad (63)$$

as found by Harris and Kirkpatrick (1977) who quote a numerical value from computer simulation $\tau = 0.5$. It should be noted that the Curie-type paramagnetic susceptibility of finite clusters with net magnetisation at zero temperature may mask this effect in experimental systems. The existence of a local net moment will also affect the spin wave dispersion as the relation from the phenomenological theory of spin waves would suggest. (Harris and Kirkpatrick 1977)

$$B(p) = \left(\frac{\Sigma(p)}{\chi_{\perp}(p)}\right)^{\frac{1}{2}} \qquad (64)$$

Matching the simple dispersion (52) to give (54) ignores such an effect. Substitution of (54) into (64) determines $\chi_{\perp}(p)$ as

$$\chi_{\perp}(p) = \frac{\Sigma(p)}{(B(p))^2} \sim \frac{(p - p_c)^{t+\beta}}{(p - p_c)^{t}}$$

$$\sim (p - p_c)^{\beta} \qquad (65)$$

giving a susceptibility that vanishes rather than diverging. We shall try to understand the effects of fluctuations by considering propagation of spin waves on a locally ferrimagnetic chain. The low energy dispersion for a ferrimagnetic chain with net moment $|S_a - S_b|$ per unit cell is

$$\omega(q) = \frac{J}{|S_a - S_b|} q^2 \tag{66}$$

If the net moment at each point in the chain is an independent random variable with mean zero, then averaged over a length λ there will be a net moment per unit length of order $\lambda^{-\frac{1}{2}}$. For a spin wave of wavevector q it would be appropriate to average over a wavelength, giving an average moment

$$\langle |S_a - S_b| \rangle_{1/q} \sim q^{\frac{1}{2}} \tag{67}$$

Suggesting a dispersion

$$\omega(q) = \frac{J}{q^{\frac{1}{2}}} q^2 = Jq^{3/2} \tag{68}$$

If we include this speculative dispersion relation and follow a matching $q = q_c$, we find instead of (52)-(54)

$$\omega(q) = \begin{cases} B(p)q & q \leq q_c \\ Jq^{3\zeta/2\nu_p} & q \geq q_c \end{cases} \tag{69}$$

$$B(p) = (p - p_c)^{3\zeta/2-\nu_p} \tag{70}$$

or

$$S = 3\zeta/2-\nu_p \tag{71}$$

From the expression (64) we deduce

$$\frac{1}{\chi_\perp} = \frac{B(p)^2}{\Sigma(p)} \sim \frac{(p - p_c)^{3\zeta - 2\nu_p}}{(p - p_c)^{t+\beta}}$$

$$= (p - p_c)^{\zeta - \beta} \qquad (72)$$

provided $t = 2(\zeta - \nu_p)$ as was found in three dimensions.

Comparison with (63) gives

$$\tau = \zeta - \beta \qquad (73)$$

Relation (73) for τ agrees with that of Harris and Kirkpatrick (1977) who gave a scaling relation for $\chi_\perp$ directly.

A dispersion relation of the form (69) has

$$\rho_{1d}(\omega) \sim \frac{1}{\omega^{1/3}} \qquad (74)$$

and this would affect calculations of T_c. Full inclusion of randomness in the percolating cluster may make the singularity in ρ_{1d} even stronger as argued for the ferromagnet, and again randomness may localize spin waves.

3.7 Conclusions.

We have demonstrated that the scaling properties of the percolating cluster are consistent with quite simple geometric models of the cluster. A new scaling relation for the conductivity has been presented and its

success and failure in low and high dimensions interpreted geometrically. Differences between isotropic and anisotropic spin systems are stressed and given a tentative geometric explanation. The scaling picture so developed has been applied to the dispersion and thermodynamics of isotropic spin systems. The form of the dispersion and density of states so derived make explicit reference to properties of the one-dimensional local connectivity. This has the advantage that the effects of fluctuations on one-dimensional properties immediately suggest modifications in scaling relations for percolation. In particular speculation on the dispersion relation for a ferrimagnet with fluctuating local moment leads to a scaling form for the perpendicular susceptibility in agreement with previous arguments.

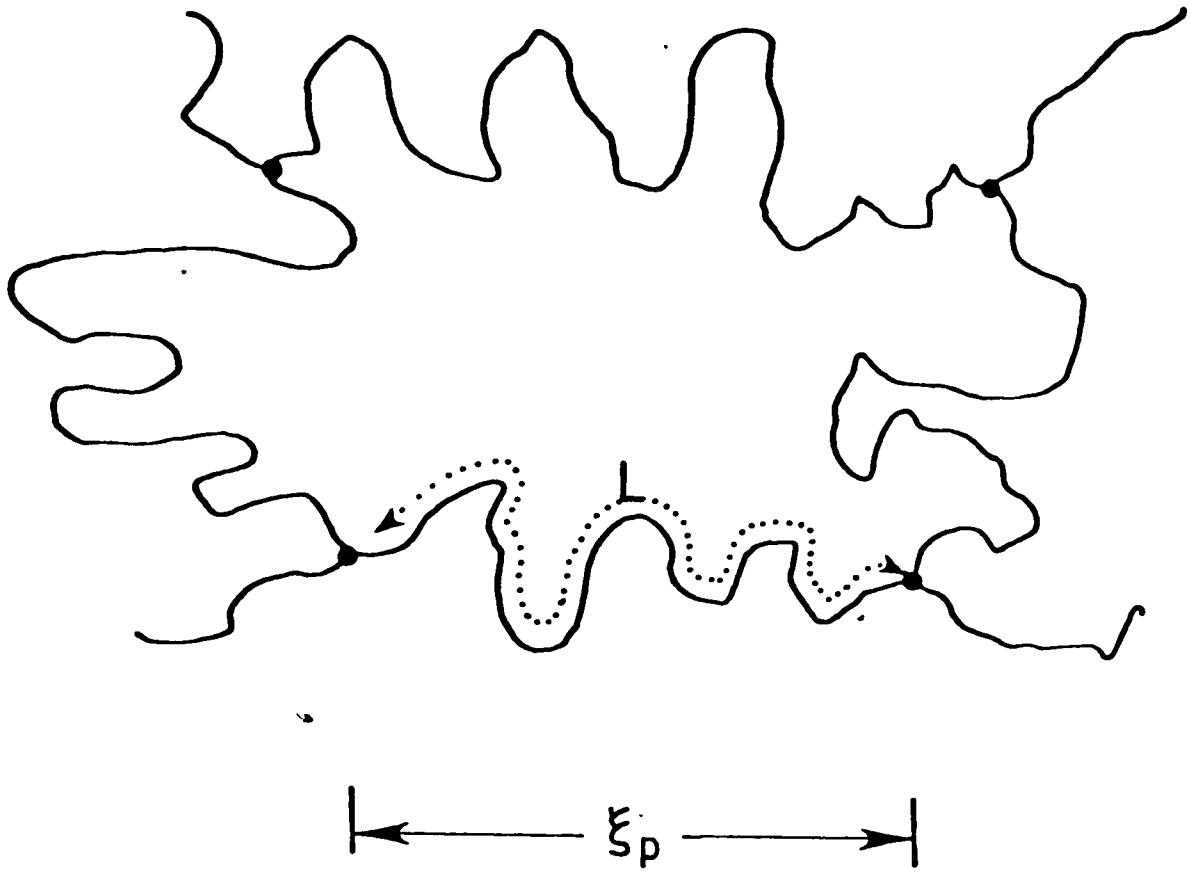


Figure 3.1. Scaling model of the percolating cluster.

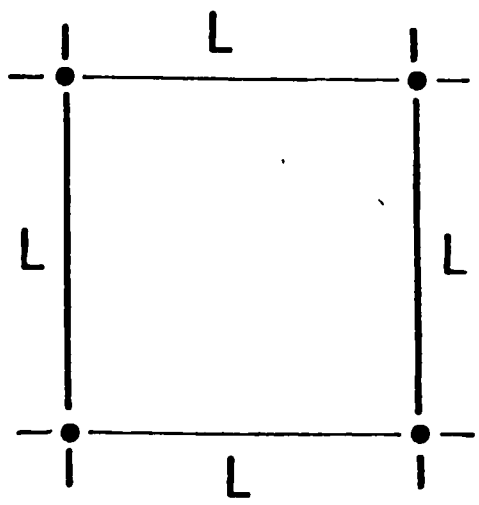


Figure 3.2.

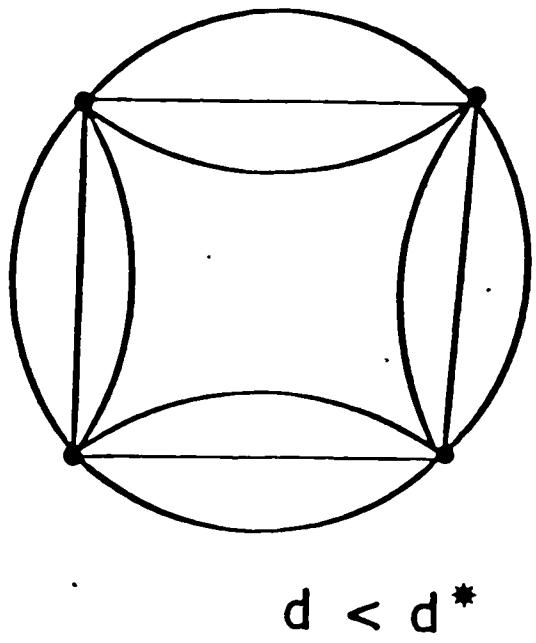


Figure 3.3a. Generalised model of the percolating cluster for low dimensions.

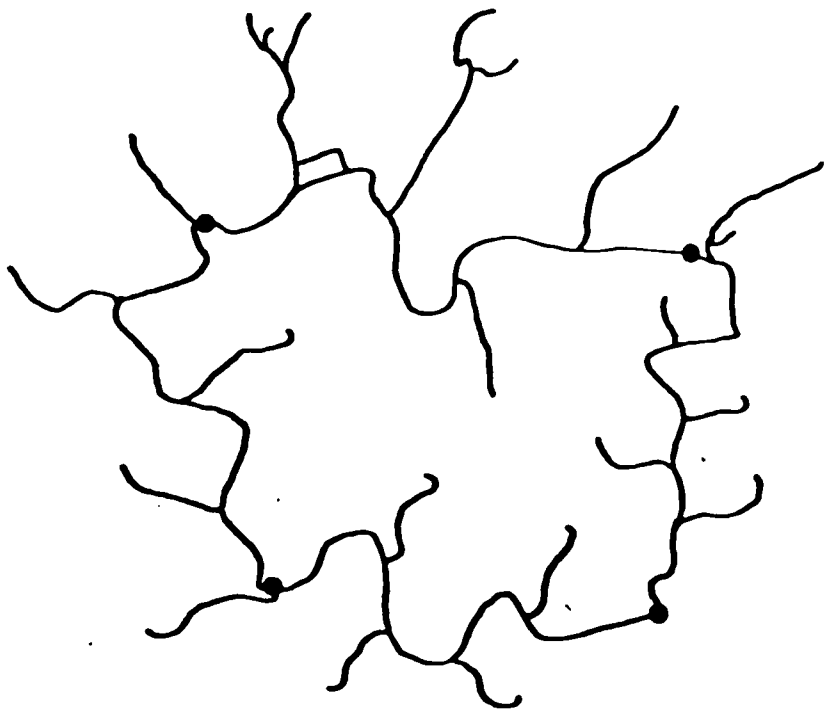


Figure 3.3b. Generalised model of the percolating cluster for high dimensions.

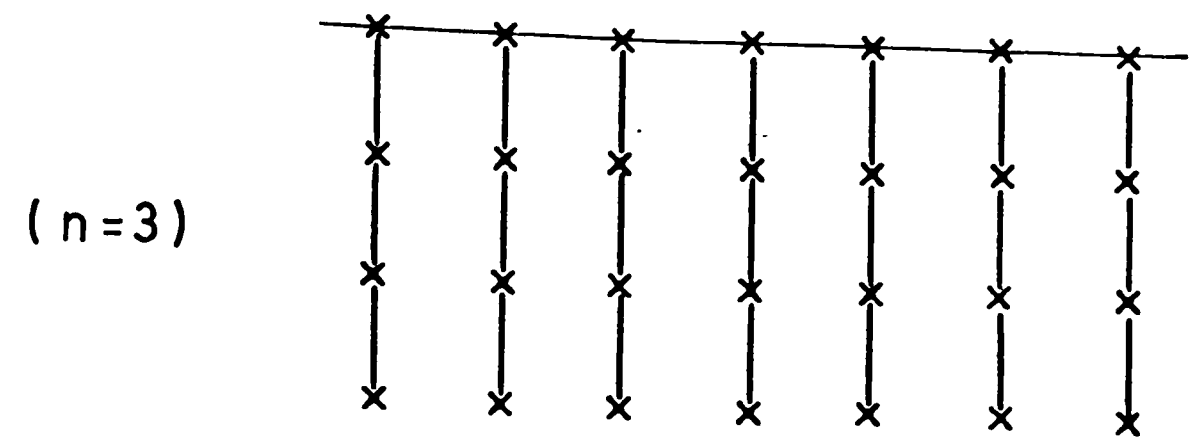


Figure 3.4. Chain with dead end branchings.

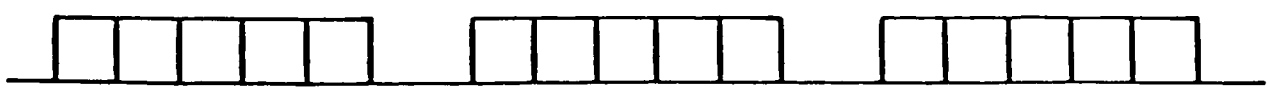


Figure 3.5. Chain with parallel linkage.

CHAPTER 4

LOCALISATION OF EXCITATIONS NEAR THE PERCOLATION THRESHOLD

4.1 Introduction.

The question of the localised or extended nature of excitations in random solids has attracted much attention since Anderson's paper on the subject (Anderson 1958, discussed by Thouless 1970, 1974). Attention has focussed primarily on the localisation of wavefunctions for electrons in disordered metals and semiconductors where localisation affects the electronic transport properties. Experimental study of localisation has been most detailed for Si/SiO_2 interfaces (Mott, Pepper, Pollitt, Wallis and Adkins 1975), in which both the fermi energy and the degree of disorder can be varied in a controlled way. Less attention has been paid to the localisation of spin wave excitations in magnetic insulators, presumably because of difficulties in observing spin diffusion even in pure crystals (see however Benner 1978). Furthermore the chemical potential is zero and cannot be varied in any obvious manner., On the other hand the problem is clearly important as experiments in random magnets develop and predictions can be tested by computer simulation.

This chapter concerns localisation of spin waves in a randomly dilute isotropic Heisenberg ferromagnet with nearest neighbour exchange, as defined in Chapter 3. Spin excitations can propagate only via occupied sites; consequently the percolative effects considered in Chapter 3 are important. Below percolation all excitations are localised on finite

clusters while above the percolation concentration there is an infinite cluster supporting at least one extended mode: the Goldstone mode at zero energy corresponding to the symmetry of the system with respect to uniform rotation of all the spins in the infinite cluster. Thus above the percolation concentration we expect a mobility edge $E_\ell(p)$ for excitations of the infinite cluster such that for $E > E_\ell(p)$ all are localised. At the percolation concentration there is a finite width to the density of states even ignoring the tail extending to the pure bandwidth corresponding to improbably large fluctuations (Lifshitz 1964). Immediate questions are posed: what is $E_\ell(p)$ as a function of p and, in particular for $p \to p_c^+$ does $E_\ell(p)$ stay a finite fraction of the total width or does it tend to zero; if so, how?

Elementary spin wave excitations of the dilute Heisenberg model are described by linear combinations of spin deviations on the occupied sites i and are created by operators

$$S_E^- = \sum_i a_i(E) S_i^- \qquad (1)$$

where the coefficients a_i satisfy a difference equation (see, for example Elliott, Krumhansl, Leath 1974)

$$(E - \Sigma_i) a_i(E) = -2 \sum_j S_i J(i, j) a_j(E) \qquad (2)$$

where S_i is the expectation value $\langle S_i^z \rangle$ in the ground state and

$$\Sigma_i = 2 \sum_j S_j J(i,j) \qquad (3)$$

Summation in (2) and (3) extends over occupied sites j nearest neighbours to i. Formally the difference between this problem and that considered by

Anderson is that here there is correlated diagonal and off-diagonal disorder; correlation (3) ensuring that there is always the zero energy solution $a_i(E) = S_i$. In this chapter we consider localisation properties of solutions to (2). First we review what is known about localisation, in sections 4.2 and 4.3, the latter when the effects of reducing the lattice connectivity to that of one-dimensional system are included.

4.2 Localisation in the Anderson Model.

We shall be guided in our inquiry by properties of the Hamiltonian written by Anderson in 1958.

$$\mathcal{H} = \sum_i \varepsilon_i a_i^\dagger a_i + \sum_{ij} V_{ij} a_j^\dagger a_i \qquad (4)$$

where ε_i is a random variable with a distribution of width W representing a random site energy, V_{ij} a constant V for nearest neighbours only: a hopping probability. In principle the Hamiltonian can be diagonalised to give eigenfunctions $\psi_\alpha(i) = a_i^\alpha$ of energy eigenvalue E_α. For a periodic system, from Bloch's theorem, $|\psi_\alpha(i)|^2$ summed over each unit cell is a constant $\frac{1}{N}$ where N is the number of unit cells (assumed equal to the number of sites in the following), but for a random system this need no longer be true: eigenfunctions may have appreciable amplitude in a localised region of the solid.

There are several means of characterising localisation (see review by Thouless 1974). A conceptually simple one is by the inverse participation ratio

$$A_\alpha = \frac{\sum_{i=1}^{N} |a_i^\alpha|^4}{\left(\sum_{i=1}^{N} |a_i^\alpha|^2\right)^2} \qquad (5)$$

which is of order $\frac{1}{N} \to 0$ for extended states with amplitude of order $\frac{1}{\sqrt{N}}$ on each site, and is of order a non-zero constant $\sim 1/L$ for $N \to \infty$ for states with amplitude on L states only. This is useful if the eigenstates can be found explicitly, in computer simulation for example.

Anderson (1958) considered the function $G_{oo}(t)$ (in Thouless' (1970) notation), the site diagonal time dependent Green's function. If an eigenstate is localised we expect an excitation at time $t = 0$ at one of the sites o within the amplitude envelope to remain there with some probability $|G_{oo}(t)|^2$ as $t \to \infty$. Ignoring degeneracy, denote $\psi_\alpha(0) = a_o^\alpha$ where $\sum_\alpha |a_o^\alpha|^2 = 1$, then for $G_{oo}(0) = 1$

$$G_{oo}(t) = \sum_\alpha |a_o^\alpha|^2 e^{-iE_\alpha t} \qquad (6)$$

with Fourier transform

$$G_{oo}(E) = \sum_\alpha \frac{|a_o^\alpha|^2}{E - E_\alpha} \qquad (7)$$

Then

$$\lim_{t\to\infty} |G_{oo}(t)|^2 = \lim_{t\to\infty} \sum_\alpha \sum_\beta |a_o^\alpha|^2 |a_o^\beta|^2 e^{i(E_\alpha - E_\beta)t}$$

$$= \sum_\alpha |a_o^\alpha|^4 \qquad (8)$$

From $\lim_{t\to\infty} e^{i(E_\alpha - E_\beta)t} = \delta_{E_\alpha, E_\beta}$ in a distributional sense.

Thus (8) averaged over sites 0 is the inverse participation ratio averaged over sites α. In the limit $N \to \infty$ $\lim_{t\to\infty} |G_{oo}(t)|^2$ is non-zero if and only if some of the states are localised. It is apparent that localisation properties are lost if the single particle Green's function $G_{oo}(t)$ is averaged with respect to site:

$$\langle G_{oo}(t) \rangle = \frac{1}{N} \sum_{o} \sum_{\alpha} |a_o^\alpha|^2 e^{-iE_\alpha t}$$

$$= \frac{1}{N} \sum_{\alpha} e^{-iE_\alpha t} \to \int \rho(E) e^{-iEt} dE \qquad (9)$$

The averaged site diagonal Green's function is the Fourier transform of the density of states and gives no direct information on localisation. For that either the distribution function for $G_{oo}(t)$ or else averaged properties of higher order Green's functions such as $\langle |G_{oo}(t)|^2 \rangle$ are required.

Anderson (1958, Abou-Chacra, Anderson and Thouless 1973) considered the Fourier transform of $G_{oo}(t)$, the site diagonal Green's function $G_{oo}(E)$ or rather the self energy $S_o(E)$

$$G_{oo}(E) = \frac{1}{E - \varepsilon_o - S_o(E)} \qquad (10)$$

$$G_{oo}(t) = \int_{-\infty}^{\infty} e^{iEt} \frac{1}{E - \varepsilon_o - S_o(E)} dE \qquad (11)$$

For $S_o(E)$ with imaginary part for $\mathrm{Im}E \to 0$, $G_{oo}(t)$ acquires an exponentially damped contribution indicating a current-carrying extended state. Anderson studied $S_o(E)$ by means of a perturbation series renormalised by multiple scattering theory. Abou-Chacra et al. wrote

this as a sum of terms over nonrepeating paths as

$$S_i(E) = \sum_{j \neq i} V_{ij} \frac{1}{E - \varepsilon_j - S_j^{(i)}(E)} V_{ji}$$

$$- \sum_{j \neq i} \sum_{k \neq i,j} V_{ik} \frac{1}{E - \varepsilon_k - S_k^{(ij)}} V_{kj} \frac{1}{E - \varepsilon_j - S_j^{(i)}} V_{ji}$$

$$+ \ldots \qquad (12)$$

$S_K^{(ij..)}$ is the self energy at site k as defined by a series similar to (12) but excluding terms involving sites ij... Multiple scattering formalism avoids divergences reflecting the crossing of energy levels rather than extension. Each term in (12) has vanishing imaginary part for ImE → 0; therefore Anderson took the criterion for extended states to be the divergence of the renormalised perturbation series (12). He analysed this by approximating the effects of self energy terms as acting as effective cut-offs on small denominators, using percolation theory to estimate the number of terms of order L as K^L with K the connective constant and studying the distribution of the sum of terms of each order by assuming them statistically independent (see criticism by Thouless 1970).

An apparently different approach to the study of the perturbation theory was that of Abou-Chacra, Anderson and Thouless (1974) who considered only the first term of the series (12), or more precisely that for $S_i^{(j)}(E)$ and assumed $S_i^{(jk)}$ to have the same distribution as the $S_i^{(j)}$ in the series. The probability distribution for the self energy term $S_i(E)$ at each lattice point i is required to satisfy the self consistent equation

$$S_i(E) = \sum_{j=1}^{K} V_{ij} \frac{1}{E - \varepsilon_j - S_j(E)} V_{ji} \qquad (13)$$

K is the connective constant. This approximation to a real finite dimensional lattice is exact on a Bethe lattice with Z = k+1 where there is at most one path between any two points and therefore $S_i^{(jk)}(E) = S_i^{(j)}(E)$ in the series (12) and there are no closed nonrepeating paths of length greater than 2. The results of the calculations were nonetheless similar to the earlier ones of Anderson. This may be related to the assumption of statistical independence of the K^L terms of order L.

Both methods gave a condition for convergence at the band centre within certain approximations of

$$\frac{W}{V} > 4K \ln \frac{W}{2V} \qquad (14)$$

Site disorder is predicted, then, to localise all the eigenstates of the Hamiltonian when the distribution width is several times the band width. In three dimensions (14) agrees with numerical estimation (Thouless 1974).

An unphysical aspect of the Bethe lattice is that it leads to narrower bands for given coordination Z > 2 than do finite dimensional lattices. Solution of (13) for a regular Hamiltonian leads to a band bounded by $\varepsilon_i \pm 2\sqrt{K}V$. This is particularly distressing for a spin wave Hamiltonian with $\varepsilon_i = ZV$, since the zero energy excitation is no longer at the band edge. This can happen because the uniform mode is no longer

the limit of current-carrying k-dependent excitations owing to the loss of k-space.

Localisation can be studied other than through the site diagonal Green's function. If localised states ψ_α have envelopes falling off exponentially about r_α say, the Green's function $G_{rr'}(E)$ can be written as a sum of terms, for $|r - r_\alpha| \to \infty$ $|r'-r_\alpha| \to \infty$

$$G^{\alpha}_{rr'}(E) = \frac{1}{E - E_\alpha} \exp\left[-\lambda_\alpha |r-r_\alpha| - \lambda_\alpha |r'-r_\alpha|\right] \qquad (15)$$

in terms of inverse localisation length λ_α. This motivates analogies to decay of thermodynamic correlations with λ_α analogous to the inverse correlation length. Attempts have been made to formalise this. Aharony and Imry (1978a) have drawn an analogy to the thermodynamics of spin glass models. Unfortunately spin glasses are not well understood either.

It is not certain that localised states always decay exponentially or symmetrically about a finite region as in (15). Last and Thouless (1971) suggested a power law decay close to the mobility edge. Yoshino and Okazaki (1978) found evidence from computer simulation for chainlike modes in the centre of the band for an Anderson model with ε_i having a binary alloy distribution

$$f(\varepsilon_i) = p\,\delta(\varepsilon_i) + (1-p)\,\delta(\varepsilon_i - \Delta) \qquad (16)$$

This was studied in the limit $\Delta \to \infty$. For energies off the band centre they found ridge-like excitations.

A localisation criterion with less rigorous justification than Anderson's is that proposed by Economou and Cohen (1972). They considered the perturbation series (12) and with assumptions about correlations between the terms of each order and their relative signs, defined a localisation function L(E) less than 1 or greater than 1 for energies of localised or extended states respectively, and approximated L(E) by

$$L(E) = KV \exp\left[- < \log |E - E_\alpha| >\right] \qquad (17)$$

As discussed by Elliott, Krumhansl and Leath (1974), the criterion (17) depends only on the density of states which, as in (9) is found from the averaged single particle Green's function $\langle G_{oo}(t) \rangle$, yet such averaging loses localisation properties. On the other hand there is a connection between the density of states and likelihood of localisation in that as the site diagonal disorder increases, for a given value of V, so does the width in the density of states.

Economou (1972) and Lyo (1972) modified the Economou argument to give a localisation criterion for substitutionally disordered anti-ferromagnets to compare with neutron scattering measurements of Buyers et al. (1971) on $K(Co,Mn)F_3$ amd $(Co,Mn)F_2$. Their criteria suffer again from lack of rigour in that they deduce localisation properties from an averaged single particle Green's function. The results agreed well with experimental data but then the experimentalists simply used the dispersion of the modes to deduce localisation. Neither theory nor experiment therefore concerned localisation properties directly.

Economou and Antoniou (1977) developed a similar theory for general uncorrelated off-diagonal and diagonal disorder, concluding that pure

off-diagonal disorder is unlikely to localise excitations, although presence of off-diagonal disorder may modify the width of diagonal disorder needed to localise. Their predictions are borne out in part by numerical calculations of Weaire and Srivastava (1977) who found that for pure off-diagonal disorder the mobility edge closely followed the CPA band edge.

4.3 Localisation in One Dimension.

It was first suggested by Mott and Twose (1961) that in one dimension almost all eigenstates are localised for arbitrarily weak disorder, in marked contrast to the situation in higher dimensions. This has been verified by various means (see review in Ishii (1973)). The ease of localisation in one dimension might be expected from the collapse of the perturbation expansion (12) to a single term. Analogies to percolation theory and phase transitions also make the result plausible.

In one dimension equation (13) is exact with a sum over K = 1 term

$$S_i^{(i-1)}(E) = \frac{V_{i,i+1}^2}{E - \varepsilon_{i+1} - S_{i+1}^{(i)}(E)} \qquad (18)$$

Abou-Chacra, Anderson and Thouless (1973) were able to show that for K = 1 the consistency condition on the distribution function for $S_i(E)$ implied by (18) implied localisation for arbitrarily weak disorder in the ε_i, as expected. Equation (18), considered as a mapping from S_i (E) to $S_i(E)$ (dropping superscripts), can be written in the form of a linear fractional transformation with associated matrix, for $V_{i,i+1} \equiv V$

$$T_i = \begin{pmatrix} 0 & 1 \\ -1 & \frac{E-\varepsilon_i}{V} \end{pmatrix} \qquad (19)$$

The self energies S_i and S_{i+N} are related by a product of N linear fractional transformations determined by the product of the random matrices T_i. For the Anderson model with ε_i random the condition that two matrices of the form (19) commute is

$$T_1 T_2 = \begin{pmatrix} 0 & 1 \\ -1 & \frac{E-\varepsilon_1}{V} \end{pmatrix}\begin{pmatrix} 0 & 1 \\ -1 & \frac{E-\varepsilon_2}{V} \end{pmatrix} = \begin{pmatrix} -1 & \frac{E-\varepsilon_2}{V} \\ -\frac{E-\varepsilon_1}{V} & \left(\frac{E-\varepsilon_1}{V}\right)\left(\frac{E-\varepsilon_2}{V}\right)-1 \end{pmatrix}$$

$$= T_2 T_1 \iff \varepsilon_1 = \varepsilon_2 \qquad (20)$$

Thus for arbitrarily weak disorder not all the matrices commute and the relation of self energies of distant sites depends on the product of independent random non-commuting unimodular matrices, properties of which product were studied by Matsuda and Ishii (1970) who related the results to the localisation. (They however formulated the problem in terms of eigenstate amplitudes rather than self energies).

For a spin model, as in (2), the $V_{i,i+1}$ of (18) are independent random variables and the ε_i are correlated $\varepsilon_i = V_{i-1,i} + V_{i,i+1}$, so (18) can be written

$$S_i(E) = \frac{V_{i,i+1}^2}{E - V_{i,i+1} - V_{i+1,i+2} - S_{i+1}(E)} \qquad (21)$$

Denoting $Z_i(E) = V_{i,i+1} + S_i(E)$, the Z_i have a recursion relation

$$Z_i(E) = \frac{Z_{i+1}(E) - E}{\frac{Z_{i+1}(E)}{V_{i,i+1}} + 1 - \frac{E}{V_{i,i+1}}}$$

a linear fractional transformation with associated unimodular matrix

$$T_i = \begin{pmatrix} 1 & -E \\ \frac{1}{V_{i,i+1}} & 1 - \frac{E}{V_{i,i+1}} \end{pmatrix} \qquad (22)$$

Therefore as for the Anderson problem with site disorder, the self energies of widely separated sites are related via a linear fractional transformation corresponding to a product of independent random matrices. The commutation condition is:

$$T_1 T_2 = \begin{pmatrix} 1 & -E \\ \frac{1}{V_1} & 1 - \frac{E}{V_1} \end{pmatrix} \begin{pmatrix} 1 & -E \\ \frac{1}{V_2} & 1 - \frac{E}{V_2} \end{pmatrix}$$

$$= \begin{pmatrix} 1 - \frac{E}{V_2} & -E\left(2 - \frac{E}{V_2}\right) \\ \frac{1}{V_1} + \frac{1}{V_2} & \left(1 - \frac{E}{V_1}\right)\left(1 - \frac{E}{V_2}\right) - \frac{E}{V_1} \end{pmatrix} = T_2 T_1$$

$$\Longleftrightarrow \quad \text{either } E = 0 \text{ or } V_1 = V_2 \qquad (23)$$

Therefore for non-zero E the self energies depend on the product of non-commuting random unimodular matrices and the eigenstates are localised, but as $E \to 0$ the commutator vanishes indicating that the extended uniform mode should be the limit of states with a diverging localisation length. It is interesting that at $E = 0$

$$\prod_{i=1}^{n} T_i(E=0) = \begin{pmatrix} 1 & 0 \\ \sum_1^n \frac{1}{V_i} & 1 \end{pmatrix} \qquad (24)$$

Distributions for which the moment $<\frac{1}{V}>$ does not exist might be expected to show low energy properties dominated by the diverging moment as in the models considered by Bernasconi, Alexander and Orbach (1978).

In section 4.2 it was emphasised that, in general, properties of the averaged single particle Green's function will not give direct information on localisation properties. In one dimension, however, there is a dispersion relation relating the density of states to the exponential decay of eigenstates (Herbert and Jones 1971, Thouless 1972) Thouless explained the existence of such a dispersion relation in the terms that the rate of exponential fall-off of an eigenstate is essentially the imaginary part of a complex wavevector whose real part gives the integrated density of states from node-counting.

Thouless considered the G_{1N} element of the matrix $G_{ij}(E) = <i|(E-\mathcal{H})^{-1}|j>$ for a chain $i,j=1,....N$. $\mathcal{H}$ is the Hamiltonian. If $\mathcal{H}$ is tridiagonal, with site energies and nearest neighbour interactions $V_{i,i+1}$ only, the G_{1N} element can be found by matrix inversion

$$G_{1N}(E) = \prod_{i=1}^{N-1} V_{i,i+1} \Big/ \det(E-\mathcal{H})$$

$$= \prod_{i=1}^{N-1} V_{i,i+1} \Big/ \prod_{\alpha=1}^{N} (E-E_\alpha) \qquad (25)$$

The pole at energy E_β with corresponding eigenstate a_i^β has residue $a_1^\beta a_N^\beta$ which from (25) gives

$$\ln|a_1^\beta a_N^\beta| = \sum_{i=1}^{N-1} \ln|V_{i,i+1}| - \sum_{\alpha\neq\beta} \ln|E_\beta - E_\alpha| \qquad (26)$$

If an eigenstate has amplitude decreasing as $a_j^\beta = \exp(-\lambda_\beta |i_\beta - j|)$, $1 \leq i_\beta \leq N$, then $a_1^\beta a_N^\beta = \exp(-\lambda_\beta (N-1))$ defining an inverse localisation length λ_β. For the limit $N \to \infty$

$$\lambda(E_\beta) = \lim_{N\to\infty} \left\{ \frac{1}{N-1} \sum_{\beta \neq \alpha} \ln|E_\beta - E_\alpha| - \frac{1}{N-1} \sum_{i=1}^{N-1} \ln|V_{i,i+1}| \right\}$$

$$= P_i \int \rho(x) \ln|E_\beta - x| \, dx - \langle \ln|V| \rangle \qquad (27)$$

Differentiating (27) we find

$$\frac{d\lambda(E)}{dE} = P_i \int \frac{\rho(x)}{E - x} \, dx$$

$$= \mathrm{Re}\, G(E) \qquad (28)$$

where $G(E) = \langle G_{oo}(E) \rangle$ is the average site diagonal Green's function, $\rho(E) = \frac{1}{\pi} \mathrm{Im} G(E)$. Thus $\lambda'(E)$ and $\rho(E)$ are real and imaginary parts of the Green's function $G(E)$. In the case of a system with a symmetry such as (3) guaranteeing the existence of an extended mode at $E=0$, $\lambda(E=0)=0$ and

$$\lambda(E) = \int_0^E \mathrm{Re}\, G(E) \, dE \qquad (29)$$

In this case then, one expects a localisation length diverging as $\lambda^{-1}(E)$ as $E \to 0$.

A random problem with such a symmetry and for which $G(E)$ can be found was solved by Dyson (1953). He considered a chain of one-dimensional harmonic oscillators with equations of motion

$$-M_j\omega^2 x_j = K_j(x_{j+1}-x_j) + K_{j-1}(x_{j-1}-x_j) \qquad (30)$$

or, with definitions $\lambda_{2j-1} = K_j/m_j$; $\lambda_{2j} = K_j/m_{j+1}$

$$(\omega^2 - (\lambda_{2j-1}+\lambda_{2j-2}))x_j = -\lambda_{2j-1}x_{j+1} - \lambda_{2j-2}x_{j-1} \qquad (31)$$

With the substitution $\omega^2 \to E$, (31) is similar to equation (2) if $\lambda_{2j-1} = \lambda_{2j} = J_{j,j+1}$. Equation (2) is found by a decoupling approximation inexact especially in one dimension. The analogy between a spin system and a chain of oscillators can be made exact, however, for a spin $\frac{1}{2}$ XY model, equivalent to non-interacting fermions as shown by Lieb, Schultz and Mattis (1961). Smith (1970) and Bulaevskii et al. (1972) used this to deduce properties of the disordered spin $\frac{1}{2}$ XY antiferromagnetic chain.

Dyson defined a characteristic function for the problem

$$\Omega(x) = \lim_{N\to\infty} \frac{1}{2N-1} \sum_{j=1}^{N} \ln(1+x\omega_j^2)$$

$$= \int_0^\infty \ln(1+x\mu)\, D(\mu)\, d\mu \qquad (32)$$

with $D(\mu)$ the probability density function for eigenvalue solutions ω^2.

Now define

$$\omega(z) = \Omega(-1/z) \qquad (33)$$

Then

$$\omega'(z) = \frac{1}{z^2}\Omega'(-1/z)$$

$$= \frac{1}{z^2}\int_0^\infty \frac{\mu}{1-\mu/z}\, D(\mu)\, d\mu$$

$$= \frac{1}{z}\int_0^\infty \frac{\mu\, D(\mu)}{z-\mu}\, d\mu$$

$$= \frac{1}{z}\int_0^\infty \frac{-(z-\mu)+z}{z-\mu} D(\mu)\,d\mu$$

$$= -\frac{1}{z} + \int_0^\infty \frac{D(\mu)}{z-\mu}\,d\mu$$

$$= -\frac{1}{z} + G(z)$$

where G(z) is the averaged single particle Green's function. Therefore from Dyson's solution we can extract the site diagonal Green's function

$$G(z) = \omega'(z) + \frac{1}{z} \tag{35}$$

Dyson found an integral equation for $\Omega(x)$ which he could solve exactly for the family of cases in which each λ_j is an independent random variable with probability distribution function

$$G_n(\lambda) = \frac{n^n}{(n-1)!}\lambda^{n-1}e^{-n\lambda} \tag{36}$$

He wrote the solution as

$$\omega(z) = \frac{2L_n(z)}{K_n(z)} \tag{37}$$

with leading term as $z \to 0$

$$\omega(z) = \frac{(\gamma + \mathrm{Log}\,nz)^2 + \pi^2/6 - (S_{n-1}^2 + t_{n-1})}{-(S_{n-1} + \gamma + \mathrm{Log}\,nz)} \tag{38}$$

γ is Euler's constant, $S_j = \sum_{i=1}^{j} \frac{1}{i}$, $t_j = \sum_{i=1}^{j} \frac{1}{i^2}$, Log nz is defined to have a cut across the positive real axis with value $\log|nz| + i\pi$ as $\mathrm{Im}z \to 0^+$, $\mathrm{Re}z > 0$. Taking the asymptotic form (41) for $z \to 0$ we can find the Green's function from (35)

$$\omega'(z) = \frac{-2(\gamma+\operatorname{Log} nz)}{(S_{n-1}+\gamma+\operatorname{Log} nz)}\frac{1}{z} + \frac{\left[(\gamma+\operatorname{Log} nz)^2 + \frac{\pi^2}{6} - (S_{n-1}^2 + t_{n-1})\right]}{(S_{n-1}+\gamma+\operatorname{Log} nz)^2}\frac{1}{z}$$

$$= -\frac{\left[2(\gamma+\operatorname{Log} nz)^2 + 2S_{n-1}(\gamma+\operatorname{Log} nz) - (\gamma+\operatorname{Log} nz)^2 - \frac{\pi^2}{6} + S_{n-1}^2 + t_{n-1}\right]}{z(S_{n-1}+\gamma+\operatorname{Log} nz)^2}$$

$$= -\frac{1}{z(S_{n-1}+\gamma+\operatorname{Log} nz)^2}\left[(\gamma+\operatorname{Log} nz+S_{n-1})^2 - (\pi^2/6 - t_{n-1})\right]$$

(39)

$$G(z) = \omega'(z) + \frac{1}{z}$$

$$= \frac{(\pi^2/6 - t_{n-1})}{z(S_{n-1}+\gamma+\operatorname{Log} nz)^2}$$

(40)

Letting $\eta = \operatorname{Im} z \to 0^+$, $\operatorname{Re} z \to E \geq 0$ $\operatorname{Log} nz = \underline{\log}\, nE + i\pi$

$$G(E+i\eta) = \frac{\left(\frac{\pi^2}{6} - t_{n-1}\right)}{E\left[(S_{n-1}+\gamma+\log nE)^2 - \pi^2 + 2i\pi(S_{n-1}+\gamma+\log nE)\right]}$$

$$\frac{1}{\pi}\,\mathrm{Im}\,G(E) \simeq \frac{-2\left(\pi^2/6 - t_{n-1}\right)\left(S_{n-1}+\gamma+\log nE\right)}{E\left[\left(S_{n-1}+\gamma+\log nE\right)^4+\pi^4+2\pi^2\left(S_{n-1}+\gamma+\log nE\right)^2\right]}$$

$$\sim \frac{-2\left(\pi^2/6 - t_{n-1}\right)}{E\left(S_{n-1}+\gamma+\log nE\right)^3} \qquad (42)$$

$$\mathrm{Re}\,G(E) = \frac{\left(\pi^2/6 - t_{n-1}\right)\left[\left(S_{n-1}+\gamma+\log nE\right)^2-\pi^2\right]}{E\left[\left(S_{n-1}+\gamma+\log nE\right)^4+\pi^4+2\pi^2\left(S_{n-1}+\gamma+\log nE\right)^2\right]}$$

$$\sim \frac{\left(\pi^2/6 - t_{n-1}\right)}{E\left(S_{n-1}+\gamma+\log nE\right)^2} \qquad (43)$$

The terms (42) and (43) are the dominant singularities in the real and imaginary parts of G(E) for E → 0. Equation (29) now predicts an inverse localisation length vanishing as

$$\lambda(E) \sim \frac{\left(\pi^2/6 - t_{n-1}\right)}{\log\left(\frac{1}{nE}\right)} \qquad (44)$$

A logarithmically divergent localisation length for the model has previously been quoted by Theodorou and Cohen (1976) who estimated λ(E) by approximating the integral (27) rather than using the real part of the Green's function as above.

Thus in one class of random models with symmetry guaranteeing an extended mode at E = 0 the localisation length diverges logarithmically. This may be true more generally.. If we consider a general disordered spin wave Hamiltonian on a chain as in (2). G(E) is likely to show power law and logarithmic singularities at E = 0. We consider then Green's functions with singularity

$$G(E) \sim E^{-a}(\log E + i\pi)^{-(1+r)} \qquad (45)$$

The strongest such singularity compatible with the integrability of G(E) is that with a = 1, r > 0. In a general random problem it is plausible that all possible singularities are present in G(E) which will then be dominated by the strongest

$$G(E) \sim \frac{1}{E(|\log E| + i\pi)^{1+r}} \tag{46}$$

The real and imaginary parts of such a function behave as

$$\frac{1}{\pi} \operatorname{Im} G(E) \sim \frac{1}{E(\log \frac{1}{E})^{2+r}} \tag{47}$$

$$\operatorname{Re} G(E) \sim \frac{1}{E(\log \frac{1}{E})^{1+r}} \tag{48}$$

$$\lambda(E) = \int_0^E \operatorname{Re} G(E)\, dE$$

$$\sim (\log \tfrac{1}{E})^{-r} \tag{49}$$

Dyson's solution is compatible with this when r = 1.

4.4 Localisation of Spin Waves Near the Percolation Threshold.

We return now to the problem of the mobility edge in a dilute ferromagnet defined in section 4.1. Equation (2) has correlated diagonal and off-diagonal disorder. From the discussion in section 4.2 we expect

that provided the connectivity of the dilute lattice is close to that of a three-dimensional lattice, the off-diagonal disorder is relatively unimportant in localising excitations and as the diagonal disorder Σ_i is no more than a fraction of the bandwidth of the pure crystal, it is unlikely that a significant proportion of the modes will be localised. Of course correlation effects may be important but as we have seen they act at low energies to maintain the extended state at zero energy. Recent numerical studies for a dilute two dimensional ferromagnet by Fujiwara (1978) support the conclusion that well above percolation almost all the excitations are extended.

Just above the percolation threshold, however, the connectivity of the lattice is greatly reduced, as discussed in Chapter 3. From section 4.3 of this chapter we expect that excitations in a lattice of low connectivity are much more readily localised. To predict the behaviour of the mobility edge just above percolation we take models of the infinite cluster discussed in Chapter 3, which give good account of the effects of reduced connectivity on thermodynamic correlations. We consider then propagation in the model lattice of figure 3.1, with random paths of length $L \sim (p - p_c)^{-\zeta}$ connecting nodes of a superlattice separated by $\xi_p \sim (p - p_c)^{-\nu_p}$. Although essentially one-dimensional the paths have dangling bonds and local parallelisms. Such features have the effect of introducing random energy dependent self energies $\Sigma_i(E)$ and bond strengths $J_{ij}(E)$ into the equations of motion of spin waves on the paths. The local spin dynamics are modelled then by an effective one-dimensional random Hamiltonian, which must still preserve the Goldstone symmetry (3) in the limit $E \to 0$. For such a random Hamiltonian, taken in the limit of an infinite chain, we can define a density of states

$\rho(E)$ and inverse localisation length $\lambda(E)$ as in section 4.3, with $\lambda(E)$ non-zero except at $E = 0$. The mobility edge in the infinite cluster for fixed dilution p just above p_c is determined by considering the possibility of tunnelling from states localised on chains of length L(p) via coupling at the nodes of the superlattice. For a given energy E, on a chain of length L there will be discrete eigenvalues randomly spaced by energies of order $\frac{1}{L\rho(E)}$. As each eigenstate is localised with exponential decay $\lambda(E)$ the overlap of eigenstates on chains adjoining at a given node will be of order $J \exp[-\lambda(E)L]$. By comparison with known results for the Anderson model the excitations will remain localised in spite of the coupling at the nodes provided that the width of the distribution in energies of the closest eigenvalue of each finite chain $\frac{1}{L\rho(E)}$ is much greater than the overlap $J \exp[-\lambda(E)L]$. The localisation criterion is then

$$J \exp[-\lambda(E)L] < C/L\rho(E) \qquad (50)$$

where C is a constant characteristic of the superlattice. The mobility edge $E_\ell(p)$ is assumed given by an equality in (51). As $p \to p_c$ L diverges and consequently $\lambda(E_\ell) \to 0$, therefore $E_\ell(p) \to 0$. The model therefore predicts that the mobility edge falls to zero at the percolation concnetration in spite of the finite bandwidth.

To predict the manner in which $E_\ell(p)$ vanishes a knowledge of $\rho(E)$ and $\lambda(E)$ as $E \to 0$ is necessary. In 4.3 we discussed a random model with the appropriate symmetry for which

$$\rho(E) \sim \frac{1}{E(\log(1/E))^3} \qquad \lambda(E) \sim \frac{1}{\log(1/E)} \qquad (51)$$

If the singular forms (51) are substituted into (50) the mobility edge is found to vanish as

$$E_\ell(p) \sim \exp\left[-\frac{A}{(p - p_c)^{\zeta/2}}\right] \qquad (52)$$

A is constant. If the more general singularities (47) and (48) are assumed then $F_\ell(p)$ vanishes in a similar fashion:

$$E_\ell(p) \sim \exp\left[-\frac{A}{(p-p_c)^{\zeta/1+r}}\right] \qquad (53)$$

In Chapter 3 we discussed modification to the effective cluster geometry suggested by reconciliation of known exponents. In particular in three dimensions a modified model cluster (as in figure 3.3a) was found to give more satisfactory predictions. The effect of such a geometry would be from (14) to replace the constant C by a factor with a power law divergence in $(p - p_c)$. Such a modification has no effect on the dominant behaviour (52) and (53).

We find then that the combined effects of disorder and reduced connectivity drive the mobility edge to zero at the percolation threshold as in equation 53, drawn in figure 4.1

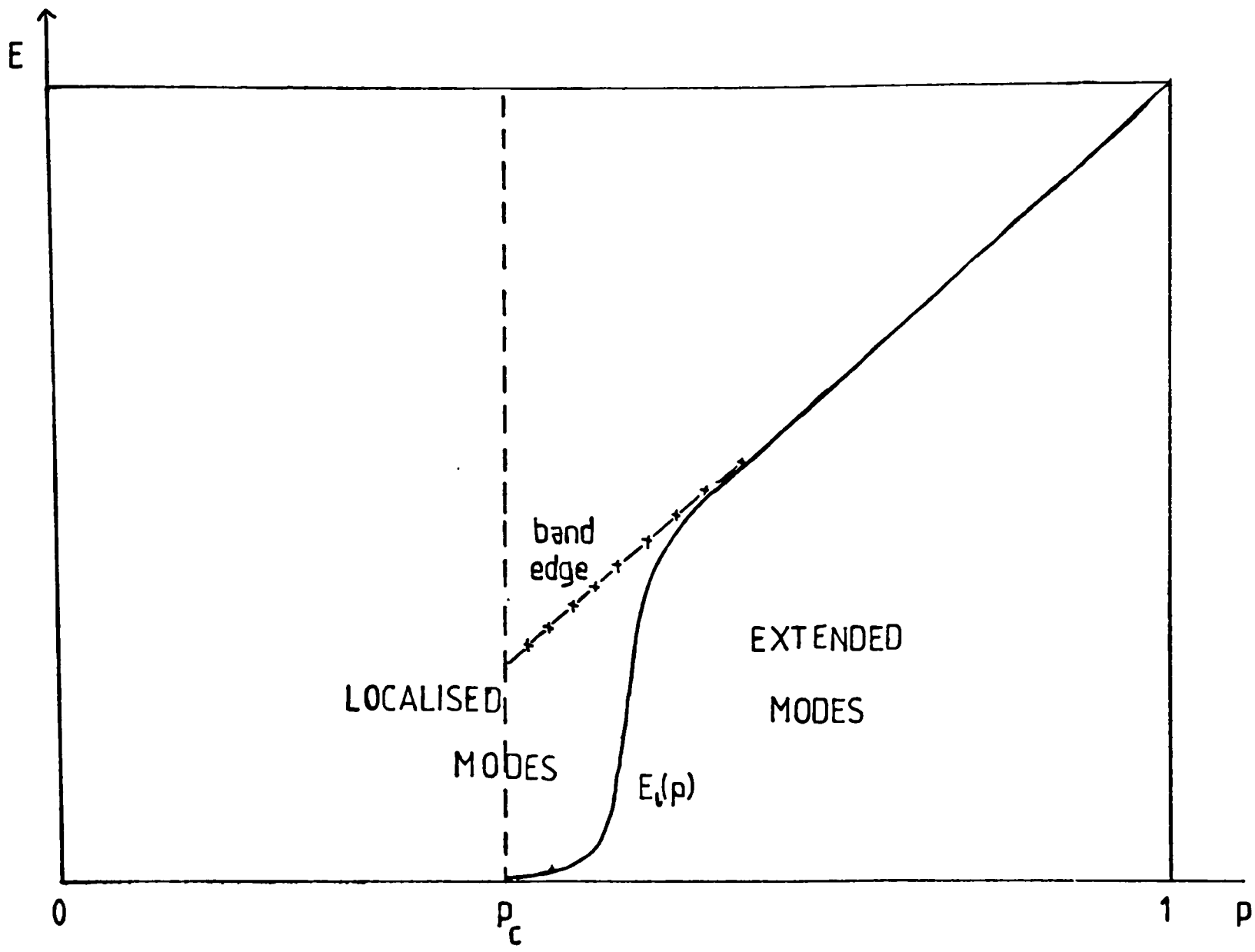


Figure 4.1. The mobility edge in a dilute ferromagnet near the percolation concentration.

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