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STATISTICAL THEORY OF
FERROMAGNETS WITH IMPURITIES

報告番号乙第1276号

宮 島 佐 介

主論文 Statistical Theory of Ferromagnets with
Impurities

(不純物磁性体の統計理論)

(主論文のうち印刷公表したもの)

1. A Statistical Model for the Dilute Ferromagnet

(稀薄強磁性の統計的モデル)

Progress of Theoretical Physics

Vol. 36, NO. 6

昭和41年12月

1. A Statistical Model of Ising Spin with Five
Transition Points

(5つの相転移点をもつイジングスピンの統計的モデル)

Progress of Theoretical Physics

Vol. 40, No. 1

昭和43年7月

1. Modified Slater Model in Kagome Lattice

(かごめ格子における変型スレーターモデル)

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1. Three Phase Transitions of Ising Model

(3つの相変化のあるイジングモデル)

Progress of Theoretical Physics

Vol. 40, No. 3

昭和43年9月

1. Ising Lattices with Random Arrangements of

Ferromagnetic and Antiferromagnetic Bonds

(強磁性的な結合と反強磁性的な結合がランダムに
あるイジング格子)

Progress of Theoretical Physics

Vol. 42, No. 1

昭和44年7月

1. On the Neel Temperature of a Mixed Crystal

Consisting of Two Kinds of Antiferromagnetic

Substances

(2種類の反強磁性体の混晶のネール点について)

Progress of Theoretical Physics

Vol. 42, No. 1

昭和44年7月

1. Orderings in Certain Decorated Ising Lattices
(ある りつきイジング格子の秩序について)

Journal of Physical Society of
Japan Vol. 26, Supplement
昭和44年9月

1. Localized Spin Wave Spectrum of Antiferromagnet
with Two Different Impurities
(2種の不純物のある反強磁性体の局在スピン波の
スペクトル)

Journal of Physical Society of
Japan Vol. 32, No. 5
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1. On Critical Behavior of Dilute Ferromagnet with
Four-spin Interaction
(4体相互作用のある稀薄強磁性体の臨界現象について)

Progress of Theoretical Physics
Vol. 47, No. 6
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Progress of Theoretical Physics
Vol. 49, No. 2
昭和48年2月

(主論文のうち未公表のもの)

1. Localized Magnon States in Ferrimagnets

(フェリ磁性体での局在マグノンの状態)

Progress of Theoretical Physics

投 稿 中

1. Phase Transition in n -Layer Ising Lattices
with Impurities

(不純物を含む n 層イジング格子の相変化)

Journal of Physical Society of

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投 稿 中

Statistical Theory of Ferromagnets with Impurities

Sasuke Miyazima

Department of Applied Physics, Faculty of Engineering,
Osaka University, Suita, Osaka

Synopsis

Thermodynamic properties of ferromagnet with impurities are discussed and in particular those in the critical region are investigated. The critical temperature, the critical concentration and the critical exponent for dilute ferromagnets and magnetic solid solutions are calculated on the basis of the Ising model. There are four kinds of models in the statistical problem of random system, the annealed bond, the annealed site, the quenched bond and the quenched site models. The grand partition functions of annealed bond models for the dilute ferromagnets and the magnetic solid solutions are exactly calculated and the expressions for the critical temperature, the critical concentration and the critical exponents are obtained. The critical temperature decreases monotonically in the dilute ferromagnet as the concentration of impurities increases and vanishes at the critical concentration. Several types of variation of the critical temperature are obtained in the magnetic solid solutions, depending on the exchange coupling of impurities. The specific heat has the cusp singularity in contrast with the logarithmic singularity in the two-dimensional Ising Model solved by Onsager. The renormalized exponents are constant in the annealed bond model.

In the case of the quenched bond and the quenched site models, the zero field susceptibilities are calculated by the high temperature series expansion, and the critical temperature and the critical exponent are obtained by the ratio method. The critical temperature versus concentration curves are similar to those of the annealed models both in the dilute ferromagnet and in the magnetic solid solution. The numerical results of the above calculations are investigated in comparison with the rigorous theorems. As for the critical exponent of the dilute ferromagnet we present other two models for which the rigorous

result can be obtained. The critical exponents of the above models satisfy the scaling relation. The size-effect on the critical temperature is discussed for the quenched site model, considering n -layer Ising systems of the square lattice, where $n = 1, 2, \dots, 6$ and ∞ . A simple relation between shift of the critical temperature and n is obtained.

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Chapter I Introduction

Thermodynamic properties of ferromagnet with impurities are discussed, putting stress on the critical phenomena or phase transition temperature, critical exponent, specific heat and so on.

The investigation of physical systems with an impurity arised from the study for excitation modes of lattice vibration by Lifshitz¹⁾ Recently the coherent potential approximation²⁾ is used in order to investigate a number of physical systems. There are many other methods for the study of physical systems with an impurity. The purpose and the situation of this paper as well as some related methods are explained briefly.

§1-1 Present situation of the impurity problem

There are many investigations on a system with a single impurity.^{*}) Though a single impurity has no effect on the thermodynamic properties, it is important to study the microscopic mechanism of the interaction of host atoms and an impurity and to study elementary excitations in the system with an impurity. And it may be possible to develop an exact formalism in the single impurity system. There exists also the study of localized and resonance levels^{**)} induced in systems with an impurity, for example, many-electron problem²⁾, lattice vibration¹⁾

^{*}) In this paper we focus our discussions upon the mixtured crystal with the substituted impurities and will not discuss glassic solids, liquid metals etc.

^{**)} There are many discussions concerning the distribution of energy levels and localizability of wave function. For example, Tanaka, Terakawa and Miyazima calculated the energy and the wave function of localized spin wave in one-dimensional magnetic systems.

and magnon in crystals³⁾ with an impurity. And the experimental investigations by the Raman scattering, far-infrared absorption and the neutron scattering⁴⁾ have been developed and have obtained the informations with regard to the impurity level.

There exist also some advanced investigations for the system with many impurities. In this case, however, the exact treatments in the mathematical form are very difficult. As for the investigation of magnetic insulator with two impurities, there have been three reports, that is, Frikkee's study for the fcc ferromagnet,⁵⁾ Miyazima and Okiji's one for the bcc antiferromagnet⁶⁾ and Tonegawa's one for the one-dimensional ferromagnet.⁷⁾

There is a new method of CPA²⁾ (coherent potential approximation) by which many-impurity problem can be treated, even though the mathematical exactness recedes a little. This method has been formulated by Soven, and analyzed to be nearly equivalent to the molecular field approximation. The Hamiltonian H consists of two parts,

$$H = H_0 + V \quad , \quad (1-1)$$

where H_0 is an unperturbed Hamiltonian for a certain regular system and V is assumed to have only the diagonal randomness. If the Green's function for the unperturbed system is given by

$$G_{ij}^0 \equiv \left\langle a_i \frac{1}{E - H_0} a_j^+ \right\rangle \quad , \quad (1-2)$$

where $\langle A \rangle = \text{tr} A e^{-H_0/kT} / \text{tr} e^{-H_0/kT}$. Then the Green's function for the total system is expressed by

$$\begin{aligned} G_{ij} \equiv \left\langle a_i \frac{1}{E - H} a_j^+ \right\rangle &= G_{ij}^0 + \sum_l G_{il}^0 V_l G_{lj}^0 \\ &+ \sum_{ll'} G_{il}^0 V_l G_{ll'}^0 V_{l'} G_{l'j}^0 + \dots, \end{aligned} \quad (1-3)$$

where a_i and a_i^+ are the annihilation and creation operators of some kind of excitons. Now if we put

$$\begin{aligned} t_1 &= V_1 + V_1 G_{11}^0 V_1 + V_1 G_{11}^0 V_1 G_{11}^0 V_1 + \dots \\ &= V_1 / (1 - G_{11}^0 V_1) , \end{aligned} \quad (1-4)$$

we obtain

$$\begin{aligned} G_{ij} &= G_{ij}^0 + \sum_l G_{il}^0 t_l G_{lj}^0 \\ &+ \sum_{l \neq 1} G_{il}^0 V_l G_{ll}^0 V_l G_{lj}^0 + \dots . \end{aligned} \quad (1-5)$$

Here if we determine H_0 such as

$$\langle t_1 \rangle_c = 0 , \quad (1-6)$$

then

$$\langle G_{ij} \rangle_c = G_{ij}^0 + \sum_{l \neq 1} G_{il}^0 (G_{ll}^0 G_{ll}^0)^2 G_{lj}^0 \langle t_1^2 \rangle_c^2 + \dots , \quad (1-7)$$

where $\langle \rangle_c$ means the summation over all the configurations. As shown in (1-7), the first correction term to G_{ij}^0 is the fourth order of t_1 . And it is possible to make the correction term smaller. In CPA it is important to choose the unperturbed Hamiltonian so that the local t -matrix comes to be zero. In the virtual-crystal approximation, the average potential is chosen as a part of unperturbed Hamiltonian. This method is utilized successfully to calculate the density of states, the dispersion relation and so on in the alloy. When impurities with finite concentration are substituted in, there appear some effects on the critical phenomena. However there are no trials to calculate the phase transition temperature by CPA. This is one of problems in the future.

§ 1-2 Ferromagnet with impurities

We confine ourselves to the Ising model with impurities, where non-magnetic, ferromagnetic and antiferromagnetic impurities can be treated. Exact expressions of the critical temperature and the critical concentration, at which the critical temperature vanishes, have been obtained by Syozi, Miyazima and Kasai for several models.⁸⁾ These are explained in chapters II and III.

In 1959 Brout⁹⁾ gave the mathematical expressions for the two systems of dilute ferromagnet, i.e. an annealed system and a quenched system. The free energy of the two systems is given by

$$-kT \left\langle \ln \left\langle \exp(-H/kT) \right\rangle_s \right\rangle_c \quad \text{for the quenched system} \quad (1-8)$$

and

$$-kT \ln \left\langle \left\langle \exp(-H/kT) \right\rangle_s \right\rangle_c \quad \text{for the annealed system,} \quad (1-9)$$

where H is the Hamiltonian for the Ising spin system with impurities k is the Boltzmann factor and T is the absolute temperature. $\langle \rangle_s$ and $\langle \rangle_c$ mean the average over the spin states and the configurations of impurities respectively.

At first the appearance of long range order in the dilute ferromagnet is explained qualitatively. The general proof of the existence of ferromagnetic long range order is given by Griffiths and Lebowitz¹⁰⁾ and the critical temperature for the two-dimensional Ising model is actually calculated in chapters II and III. When many impurities are substituted in and the concentration of magnetic ions p is smaller than $1/z$, where z is the number of nearest neighbor lattice point, the state of long range order is impossible. For it is impossible to link all neighbors into macroscopically long chains in the random

situation. When p is slightly greater than $1/z$ there is a finite probability of forming infinite clusters and making ferromagnetism possible.

Brout calculated the critical temperature of quenched system by the cumulant expansion method as follows,

$$z \tanh (J/kT_c) = 1/p . \quad (1-10)$$

In the equation (1-10), there is no solution of T_c for $p \leq 1/z$, which is equivalent qualitatively to the above-mentioned discussion.

Now we explain the Syozi model for dilute ferromagnet which is the only one exactly soluble model of dilute ferromagnet at the present time. As this model will be discussed in § 2-1, even though a little modification is added, we only give an introduction of the Syozi model. Consider a decorated Ising square lattice. There is an Ising spin at each matrix lattice point which is represented by the variable $\mu = \pm 1$, where the upper sign corresponds to the up spin and the low sign does to the down spin. The variable σ ($= \pm 1$ and 0) is introduced at each decorating lattice point. $\sigma = \pm 1$ correspond to the up and down spins respectively and $\sigma = 0$ corresponds to no spin at the decorating lattice point. Either magnetic or non-magnetic ion can occupy the decorating lattice point in the present model. This decorating Ising lattice is reduced to the ordinary Ising lattice solved by Onsager,¹¹⁾ utilizing the extended iteration transformation introduced by Syozi. Thus the exact expressions for several thermodynamic quantities can be obtained. In particular the critical concentration and the cusp type specific heat which forms a striking contrast with the logarithmic divergence must be noted.

In this paragraph we discuss the percolation problem which is closely related with the dilute ferromagnet, following to Broadbent and Hammersley's¹²⁾ paper. A system introduced in the percolation problem consists of lattice points and bonds connecting two lattice points, being satisfied with certain conditions. Two particles are connected by a bond, if they are put at the nearest neighboring sites. And thus a large cluster can be made. The magnitude of clusters depends on the ratio p of particles to total lattice point. It is apparent that the total lattice points become a cluster when $p = 1$ in the system with finite lattice points. There is a possibility of occurrence of infinite large cluster in an infinite system. The critical concentration (probability) p_c is defined as follows. If a particle which occupies a lattice point belongs to an infinitely large cluster with the probability $P(p)$, $P(p)$ becomes non-zero and finite at $p = p_c$, which is called the critical probability. There are close correspondences between the percolation problem and the dilute ferromagnet which are discussed in § 5-2.

Other related problems are the lattice gas and the binary alloys. These models may be used to investigate the distribution of grain size in sands and photographic emulsions.¹³⁾ These theories as mentioned above put stress on the subsystem of the lattice points in the percolation problem, which is called the site percolation problem. On the other hand we have a following problem which lays stress on the bond rather than the lattice point, which is called the bond percolation problem.

A bond has the function as a pipe which flows water from an edge of the bond to another edge. Adding bonds to a considered lattice

and flowing water, then many lattice points become wet and at last the infinite number of lattice points become wet at the critical concentration p_c of bonds. Otherwise the cluster of lattice points is called the maze. According to the approximate calculations the critical concentration is about 0.58 for the square lattice and about $2/z$ for the three-dimensional lattices.

As an application of the bond percolation problem, the conductor-insulator transition was discussed by Ziman.¹⁴⁾ The bond between two lattice sites is supposed to be open or closed to a classical electron, with the probability p of an open bond depending on the energy of the electron. When $p \leq p_c$ the electrons can only migrate locally but when $p > p_c$ the crystal becomes conducting. As other bond problems there are, for example, the gelation of polymers and the infection of the disease in an orchard.

Chapter II The dilute ferromagnetism

In this part we discuss the magnetic properties in the ferromagnetic substances which contain non-magnetic impurities. As discussed in the introduction we have two different systems, annealed and quenched systems. And we have two different approaches to these random magnets, that is, considering as a bond problem and as a site problem. The latter classification has not only mathematical means but also physical correspondences. In the case of solid solution $\text{Mn}(\text{Cl}_x\text{Br}_{1-x})_2 \cdot 2\text{H}_2\text{O}$ it is better to consider it as a bond problem. On the contrary it is better to consider it as a site problem in the case of $\text{Mn}_x\text{Ni}_{(1-x)}\text{Cl}_2 \cdot 2\text{H}_2\text{O}$.

As a result we can treat the dilute ferromagnetism as the following four models, annealed bond, quenched bond, annealed site and quenched site models.

§ 2-1 The Syozi model (annealed bond model)

In 1965 Syozi proposed a decorated Ising lattice as a model of the dilute ferromagnetism which was exactly soluble by using the extended iteration transformation. The spin variables μ ($= \pm 1$) at the matrix lattice points correspond to up and down state of spin and those $\sigma = \pm 1$ and 0 at the decorating lattice points correspond to up, down spins and existence of non-magnetic impurity on the bond, which means no exchange interaction between two spins at the vertices of bond. The Syozi model for the dilute ferromagnetism gave the exact expression of the phase transition temperature, the critical concentration and other thermodynamic quantities.

The Syozi model is the intermediate model of the site and bond problems. Here we give the brief explanation of this model with a modification, where it is treated as a pure bond problem, but the

essential points are same as the Syozi model. A part of bond configurations is represented in Fig. 2-1.

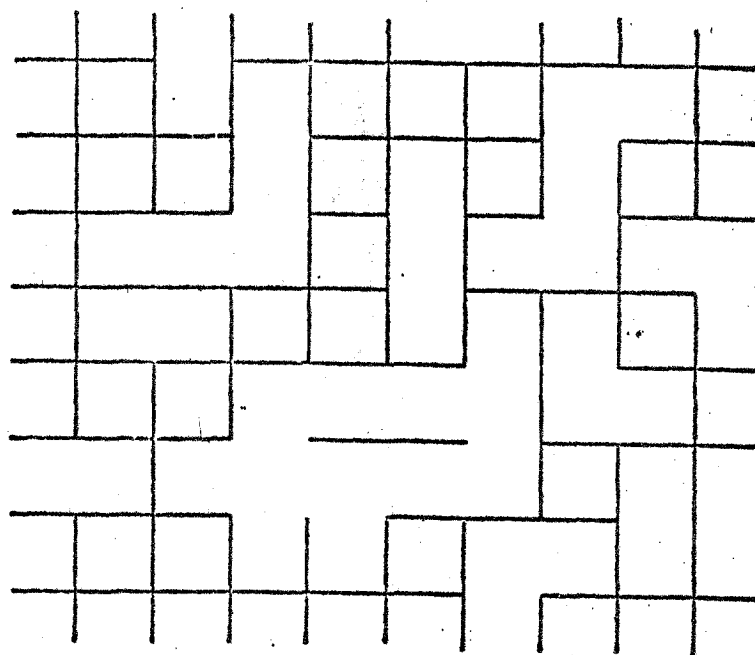


Fig.2-1 One of the bond configurations in the bond problem for the dilute ferromagnet is represented. The bond means the exchange interaction between a pair of spins at the vertices.

Assuming the exchange integral between a pair of nearest neighboring spins, The Hamiltonian is expressed as

$$H = - J \sum_{\langle ij \rangle} \mu_i \sigma_{ij} \mu_j, \quad (2-1)$$

where μ_j ($= \pm 1$) is an Ising spin variable and σ_{ij} ($= 1$ and 0) is introduced in order to express whether a bond between the i -th and j -th lattice points exists or not. The summation is over all the nearest neighboring pairs. By using the parameter ξ , the grand

canonical partition function is given by

$$\tilde{Z} = \sum_{\mu} \sum_{\langle ij \rangle} \prod \exp(L \mu_i \sigma_{ij} \mu_j + \xi \sigma_{ij}), \quad (2-2)$$

where $L = J/kT$, the first summation is over all spin configurations and the second one is over all bond configurations.

At first summing over all bond configurations in (2-2) we obtain

$$\tilde{Z} = A^{Nz/2} \sum_{\mu} \prod_{\langle ij \rangle} \exp(K \mu_i \mu_j) \quad (2-3)$$

where N is the total number of lattice points, z is the coordination number and A and K are given by the following equation,

$$A^2 = (1 + e^{+L+\xi})(1 + e^{-L+\xi}) \quad (2-4)$$

$$e^{2K} = (1 + e^{+L+\xi}) / (1 + e^{-L+\xi}) \quad (2-5)$$

Now the second factor in (2-3) is the partition function $Z_0(K)$ for the matrix Ising model with the exchange coupling constant K . Thus

$$\tilde{Z} = A^{Nz/2} Z_0(K) \quad (2-6)$$

From this grand partition function, the average number of bonds $\frac{zNp}{2}$ is calculated derivating by ξ ,

$$\frac{zNp}{2} = \frac{\partial \ln \tilde{Z}}{\partial \xi} = \frac{zN}{2} \frac{\partial \ln A}{\partial \xi} + \frac{\partial \ln Z_0(K)}{\partial K} \frac{K}{\xi} \quad (2-7)$$

Eliminating ξ by (2-5), we have

$$p = \frac{1}{2} \frac{1 - e^{-2K}}{e^{2L} - 1} \left\{ e^{2L}(1 + \xi(K)) + e^{2K}(1 - \xi(K)) \right\} \quad (2-8)$$

where ξ is the correlation function for a pair of the nearest neighboring Ising spins given by

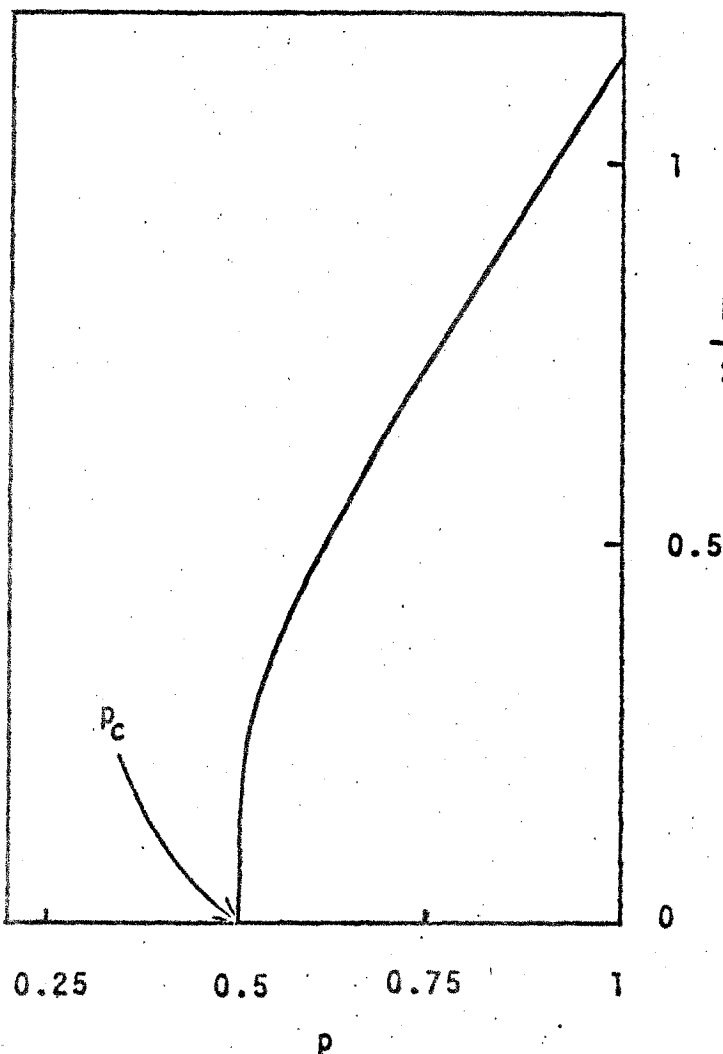
$$\xi = \frac{2}{zN} \frac{\partial \ln Z_0(K)}{\partial K} \quad (2-9)$$

Now if we utilize the critical values for the square lattice

$$\exp(-2K_c) = \sqrt{2} - 1, \quad \xi_c = \sqrt{2} / 2 \quad (2-10)$$

and (2-7), we obtain the critical temperature for the present dilute ferromagnet,

$$e^{2L_c} = 1 + \sqrt{2}/(2p-1) \quad (2-11)$$



The behavior of critical temperature given by (2-11) is represented by Fig.2-2. As shown in Fig.2-2, the critical temperature $\frac{kT_c}{2J}$ vanishes at $p = 0.5$ for the square lattice. The concentration at which

Fig. 2-2 The critical temperatures of the annealed model for the dilute ferromagnet in the square lattice versus the concentration of magnetic atoms.

the critical temperature vanishes is called the critical concentration. It is also shown by Syozi that if p_c is the critical concentration for an Ising lattice, then the critical concentration of its dual lattice p_c^* is given by

$$p_c^* = 1 - p_c. \quad (2-12)$$

It is also important that the logarithmic singularity of the specific heat in the ordinary Ising model reduces to the cusp singularity with finite value at the critical temperature. This is shown in a following way. The specific heat per bond $C (= kL^2 d \mathcal{E} / dL)$ is calculated as following,

$$C = kL^2 \left[\frac{d\xi}{dK} \frac{dK}{dL} - \left\{ \left(1 - \frac{dK}{dL} \right) \cosh(L-K) (\xi \cosh K - \sinh K) + \sinh(L-K) \cdot \frac{dK}{dL} \cdot (\xi \sinh K - \cosh K + \frac{d\xi}{dK} \cosh K) \right\} / \sinh L + \sinh(L-K) (\xi \cosh K - \sinh K) \cosh L / \sinh^2 L \right] \quad (2-13)$$

where

$$\frac{dK}{dL} = \left\{ 2p - (1 - e^{-2K})(1 + \xi) e^{2L} / \left[e^{-2K} \{ e^{2L}(1 + \xi) + e^{2K}(1 - \xi) \} + (e^{2K} - 1)(1 - \xi) + (1/2)(1 - e^{-2K})(e^{2L} - e^{2K}) \frac{d\xi}{dK} \right] \right\}. \quad (2-14)$$

In the case of the square lattice,

$$\xi = \coth 2K (\pi/2 + k'K(k))/\pi \quad (2-15)$$

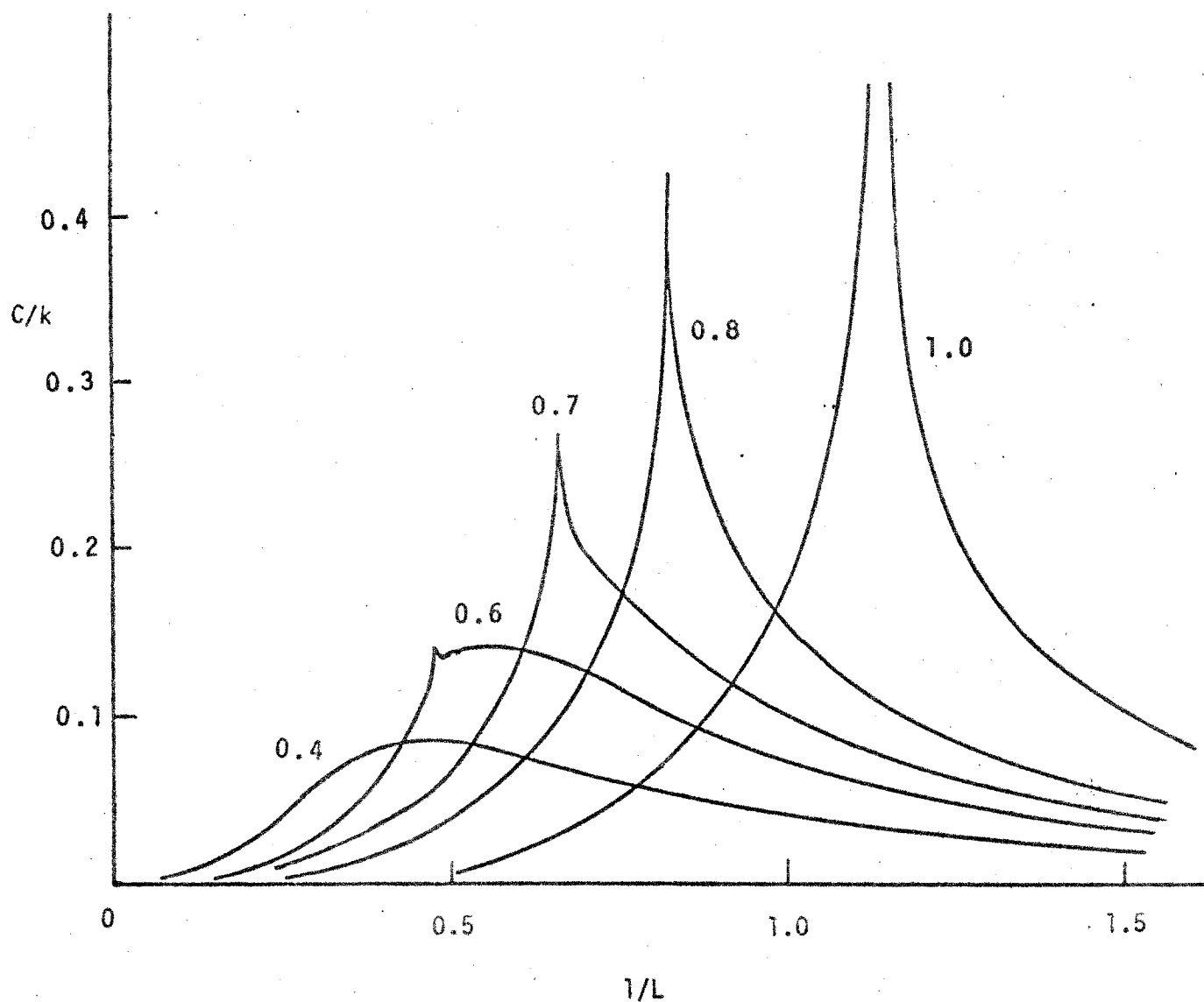


Fig. 2-3(a) The specific heats of the annealed model for the dilute ferromagnet in the square lattice are given. The numbers are the concentrations.

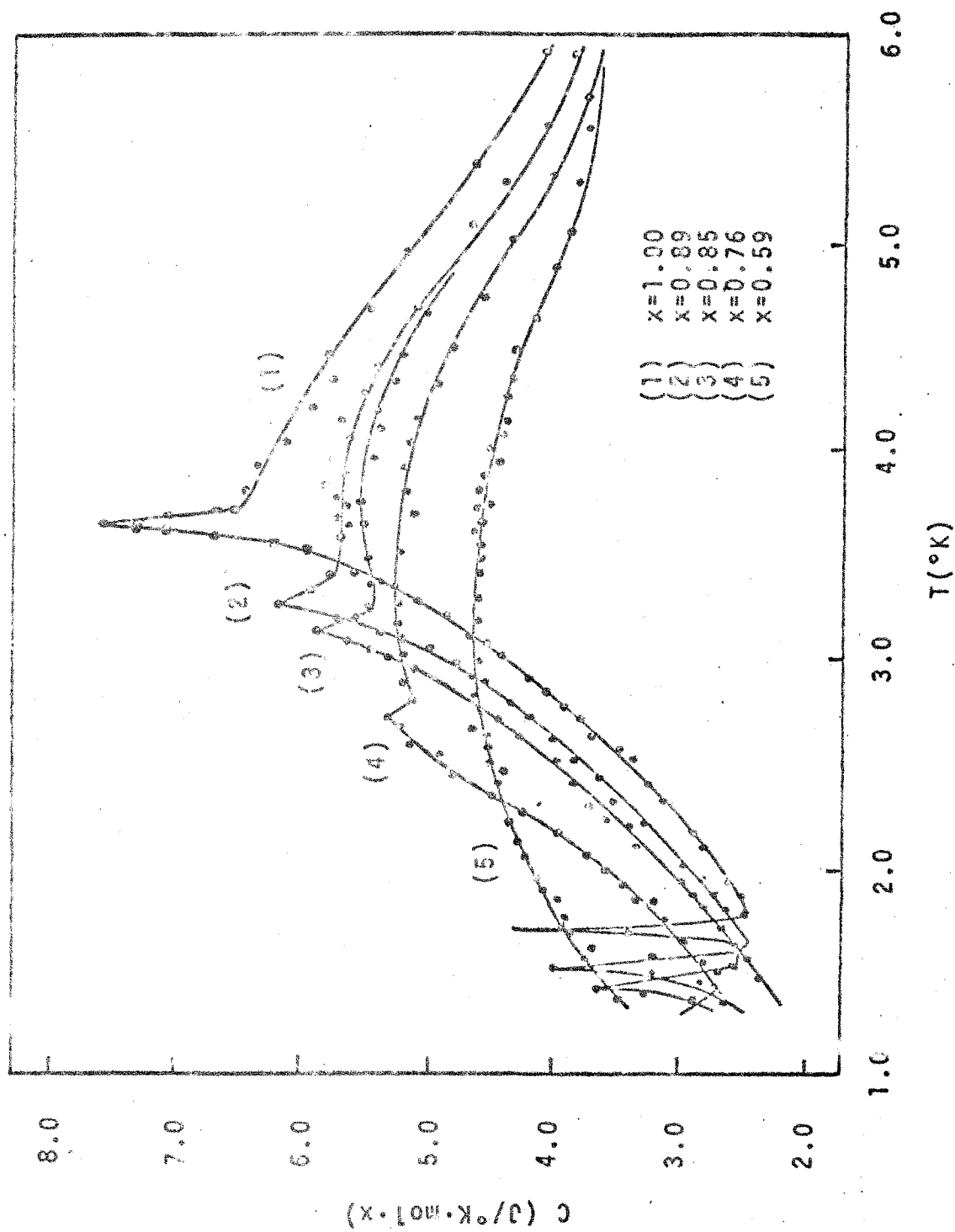


Fig. 2-3(b) The specific heats of $\text{Mn}_x\text{Mg}_{1-x}(\text{CHOO})_2$ are represented.

and

$$\frac{d\varepsilon}{dK} = \coth^2 2K \left\{ 2K(k) - 2E(k) - (1-k') \left(\frac{\pi}{2} + k'K(k) \right) \right\} / \pi, \quad (2-16)$$

where

$$k = \frac{2 \sinh 2K}{\cosh^2 2K}, \quad k' = \pm (1-k^2)^{1/2} = 2 \tanh^2 2K - 1 \quad (2-17)$$

and $K(k)$ and $E(k)$ are the first and second complete elliptic integrals respectively. The specific heat C versus $1/L$ is plotted in Fig.2-3(a). It is clearly shown in Fig.2-3(a) that the logarithmic singularity at $p = 1$ reduces to the cusp singularity for $0.5 < p < 1$. In the Fig.2-3(b) the specific heat of $Mn_x Mg_{(1-x)}(HCOO)_2$ is shown.¹⁶⁾

§ 2-2 A quenched model for the dilute ferromagnet

In the previous section we discussed the annealed bond model for the dilute ferromagnetism. In this section we discuss the quenched site and bond models by the series expansion of the zero field susceptibility. The critical phenomena are seemed to depend on the dimensionality, the interaction range and the type of interaction. In order to investigate the effect of dimensionality to the critical phenomena which is called hereafter size-effect simply, we consider two-dimensional layer Ising lattices, i.e. single-layer, two-layer, three-layer, ..., and simple cubic lattices. Of course the dilution brings the similar, not the same, effects of change of dimensionality. A complex effect of the dilution and size is also found.

The Hamiltonian of the present model in the presence of magnetic field H_0 is given by

$$H = - J \sum_{i,j} \mu_i \mu_j - m H_0 \sum_i \mu_i, \quad (2-18)$$

where m and H_0 denote the magnetic moment per spin and the field.

The zero field susceptibility χ_0 ($= \lim_{H_0 \rightarrow 0} M/H_0$, where M is the magnetization) can be expressed as¹⁵⁾

$$(kT/m^2) \chi_0 = 1 + \sum_{r=1}^{\infty} a_r(p) v^r, \quad (2-19)$$

where $v = \tanh J/kT$ and $a_r(p)$ is the number of graphs of r -th order with probability p^k . Here k is the number of sites for the considered magnetic graph^{*}) in the case of site problem and k is the number of bonds in the case of bond problem. p is the probability that a site is occupied with an Ising spin in the site problem and that two ends of a bond are occupied with two Ising spins in the bond problem, respectively. $a_r(p)$ for the quenched site model and the quenched bond model are given in Table 1 and Table 2 respectively. In the case of no impurity, that is, $p = 1$, these coefficients are same as those calculated by Ballentine¹⁷⁾ and Allan.¹⁸⁾

From this series, the critical temperature and the critical exponent γ can be estimated by the ratio method.

When we assume that $a_r = a_r(p) \sim \phi(r)/v_c^r$ in (2-19), where

$$\lim_{r \rightarrow \infty} [\phi(r)]^{1/r} = 1, \quad (2-20)$$

and $v_c = \tanh J/kT_c$, we can fit $\phi(r)$ by an expression of the form

$$\phi(r) \sim A r^g. \quad (2.21)$$

*) This terminology was introduced by Oguchi¹⁹⁾ and this high temperature counting problem was discussed by Fisher and Sykes²⁰⁾ in detail. See also Domb's review article.²¹⁾

Table 1. Coefficients for the high temperature series expansion of zero field susceptibility in the quenched site model.

| $a_r(p)$ | $n = 1$ | $n = 2$ | $n = 3$ | $n = 4$ | $n = 5$ | $n = 6$ | $n = \infty$ |
|------------|-----------|------------|-----------------------|------------|--------------|-----------------------|--------------|
| $a_1(p)$ | $4p^2$ | $5p^2$ | $5\frac{1}{3}p^2$ | $5.5p^2$ | $5.6p^2$ | $5\frac{2}{3}p^2$ | $6p^2$ |
| $a_2(p)$ | $12p^3$ | $20p^3$ | $23\frac{1}{3}p^3$ | $25p^3$ | $26p^3$ | $26\frac{2}{3}p^3$ | $30p^3$ |
| $a_3(p)$ | $36p^4$ | $80p^4$ | $102\frac{2}{3}p^4$ | $114.5p^4$ | $121.6p^4$ | $126\frac{1}{3}p^4$ | $150p^4$ |
| $a_4(p)$ | $100p^5$ | $304p^5$ | $433\frac{1}{3}p^5$ | $506p^5$ | $550p^5$ | $579\frac{1}{3}p^5$ | $726p^5$ |
| $a_5^1(p)$ | $284p^6$ | $1168p^6$ | $1841\frac{1}{3}p^6$ | $2254p^6$ | $2509.6p^6$ | $2680\frac{1}{3}p^6$ | $3534p^6$ |
| $a_5^2(p)$ | $-8p^4$ | $-16p^4$ | $-18\frac{2}{3}p^4$ | $-20p^4$ | $-20.8p^4$ | $-21\frac{1}{3}p^4$ | $-24p^4$ |
| $a_6^1(p)$ | $780p^7$ | $4348p^7$ | $7625\frac{1}{3}p^7$ | $9824p^7$ | $11232.8p^7$ | $12180p^7$ | $16926p^7$ |
| $a_6^2(p)$ | $-32p^5$ | $-96p^5$ | $-128p^5$ | $-144p^5$ | $-153.6p^5$ | $-160p^5$ | $-192p^5$ |
| $a_6^3(p)$ | $-8p^4$ | $-16p^4$ | $-18\frac{2}{3}p^4$ | $-20p^4$ | $-20.8p^4$ | $-21\frac{1}{3}p^4$ | $-24p^4$ |
| $a_7^1(p)$ | $2172p^8$ | $16336p^8$ | $31726\frac{2}{3}p^8$ | $42990p^8$ | $50530.4p^8$ | $55664p^8$ | $81390p^8$ |
| $a_7^2(p)$ | $-160p^6$ | $-696p^6$ | $-1010\frac{2}{3}p^6$ | $-1178p^6$ | $-1278.4p^6$ | $-1345\frac{1}{3}p^6$ | $-1680p^6$ |
| $a_7^3(p)$ | $-32p^5$ | $-96p^5$ | $-128p^5$ | $-144p^5$ | $-153.6p^5$ | $-160p^5$ | $-192p^5$ |
| $a_7^4(p)$ | $-8p^4$ | $-16p^4$ | $-18\frac{2}{3}p^4$ | $-20p^4$ | $-20.8p^4$ | $-21\frac{1}{3}p^4$ | $-24p^4$ |

Table 2. Coefficients for the high temperature series expansion of zero field susceptibility in the quenched bond model.

| $a_r(p)$ | $n = 1$ | $n = 2$ | $n = 3$ | $n = 4$ | $n = 5$ | $n = 6$ | $n = \infty$ |
|------------|-----------|------------|-----------------------|------------|--------------|-----------------------|--------------|
| $a_1(p)$ | $4p$ | $5p$ | $5\frac{1}{3}p$ | $5.5p$ | $5.6p$ | $5\frac{2}{3}p$ | $6p$ |
| $a_2(p)$ | $12p^2$ | $20p^2$ | $23\frac{1}{3}p^2$ | $25p^2$ | $26p^2$ | $26\frac{2}{3}p^2$ | $30p^2$ |
| $a_3(p)$ | $36p^3$ | $80p^3$ | $102\frac{2}{3}p^3$ | $114.5p^3$ | $121.6p^3$ | $126\frac{1}{3}p^3$ | $150p^3$ |
| $a_4(p)$ | $100p^4$ | $304p^4$ | $433\frac{1}{3}p^4$ | $506p^4$ | $550p^4$ | $579\frac{1}{3}p^4$ | $726p^4$ |
| $a_5^1(p)$ | $284p^5$ | $1168p^5$ | $1841\frac{1}{3}p^5$ | $2254p^5$ | $2509.6p^5$ | $2680\frac{1}{3}p^5$ | $3534p^5$ |
| $a_5^2(p)$ | $-8p^4$ | $-16p^4$ | $-18\frac{2}{3}p^4$ | $-20p^4$ | $-20.8p^4$ | $-21\frac{1}{3}p^4$ | $-24p^4$ |
| $a_6^1(p)$ | $780p^6$ | $4348p^6$ | $7625\frac{1}{3}p^6$ | $9824p^6$ | $11232.8p^6$ | $12180p^6$ | $16926p^6$ |
| $a_6^2(p)$ | $-32p^5$ | $-96p^5$ | $-128p^5$ | $-144p^5$ | $-153.6p^5$ | $-160p^5$ | $-192p^5$ |
| $a_6^3(p)$ | $-8p^4$ | $-16p^4$ | $-18\frac{2}{3}p^4$ | $-20p^4$ | $-20.8p^4$ | $-21\frac{1}{3}p^4$ | $-24p^4$ |
| $a_7^1(p)$ | $2172p^7$ | $16336p^7$ | $31726\frac{2}{3}p^7$ | $42990p^7$ | $50530.4p^7$ | $55664p^7$ | $81390p^7$ |
| $a_7^2(p)$ | $-160p^6$ | $-696p^6$ | $-1010\frac{2}{3}p^6$ | $-1178p^6$ | $-1278.4p^6$ | $-1345\frac{1}{3}p^6$ | $-1680p^6$ |
| $a_7^3(p)$ | $-32p^5$ | $-96p^5$ | $-128p^5$ | $-144p^5$ | $-153.6p^5$ | $-160p^5$ | $-192p^5$ |
| $a_7^4(p)$ | $-8p^4$ | $-16p^4$ | $-18\frac{2}{3}p^4$ | $-20p^4$ | $-20.8p^4$ | $-21\frac{1}{3}p^4$ | $-24p^4$ |

If (2-21) is valid,

$$a_r/a_{r-1} \sim (1/v_c) (1 + \frac{g}{r}) \quad (2-22)$$

and we should obtain a straight line of plotting a_r/a_{r-1} versus $1/r$ whose intersection with $1/r = 0$ determines v_c , and whose slope determines g . This means that the singularity in the susceptibility at the Curie point is of the form $(1 - T_c/T)^{-1-g}$. 22)

Some parts of $a_r/a_{r-1} \sim 1/r$ curves for the quenched site model are shown Fig. 2-4. The critical temperatures estimated by these plots are shown in Fig. 2-5. The critical temperature curve for $n = 1$ shows a good fit with that for the Syozi model. As seen in Fig. 2-5, p_c for the square lattice and s.c. lattice in the present calculation are a little larger than those values in the Syozi model. The initial gradients of $T_c - p$ curves in the present model are larger than those in the Syozi model. These are natural differences which occur between the site problem and the bond problem. Such inclinations are explained in the chapter V.

The critical exponent γ is plotted in Fig. 2-6 against p .²³⁾ The variation of critical exponent in the present model is different from that in the Syozi model. The critical exponent increases as p decreases. This result might be acceptable from the following two facts. One is the decrease of effective dimensionality by the dilution. The other is that the critical exponent for two-dimensional Ising lattice is $7/4$ and that for three-dimensional lattice is $5/4$. The same kinds of behaviors can be seen in the other systems. The critical exponents in the Syozi model will be discussed in chapter IV, and they are shown to be constant for $0 < p < p_c$ and to have gaps at $p = 1$. The present results by high temperature series expansion contradict with

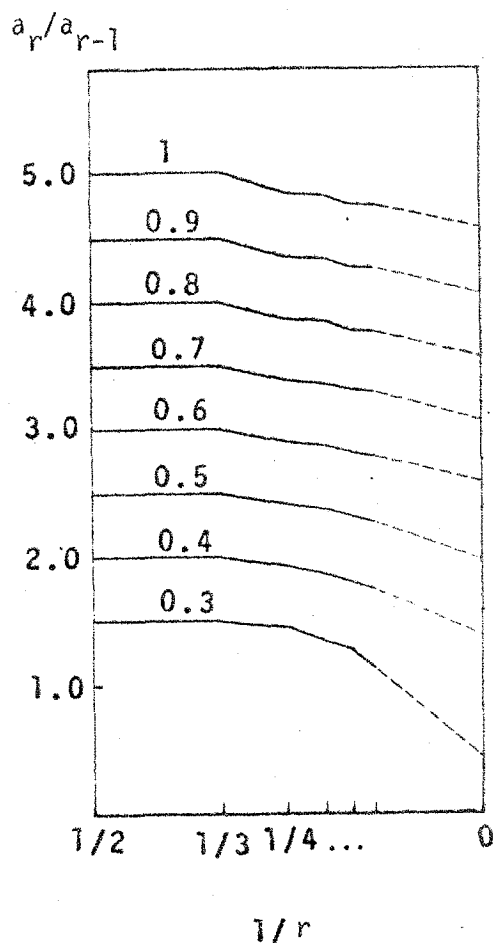


Fig.2-4 The ratio $a_r(p)/a_{r-1}(p)$ of a simple cubic lattice is given against $1/r$. The number in the figure means the concentration of magnetic atoms.

$$\frac{kT_c}{2J}$$

Fig.2-5 The critical temperature for the quenched site model versus the concentration of magnetic atoms is represented.

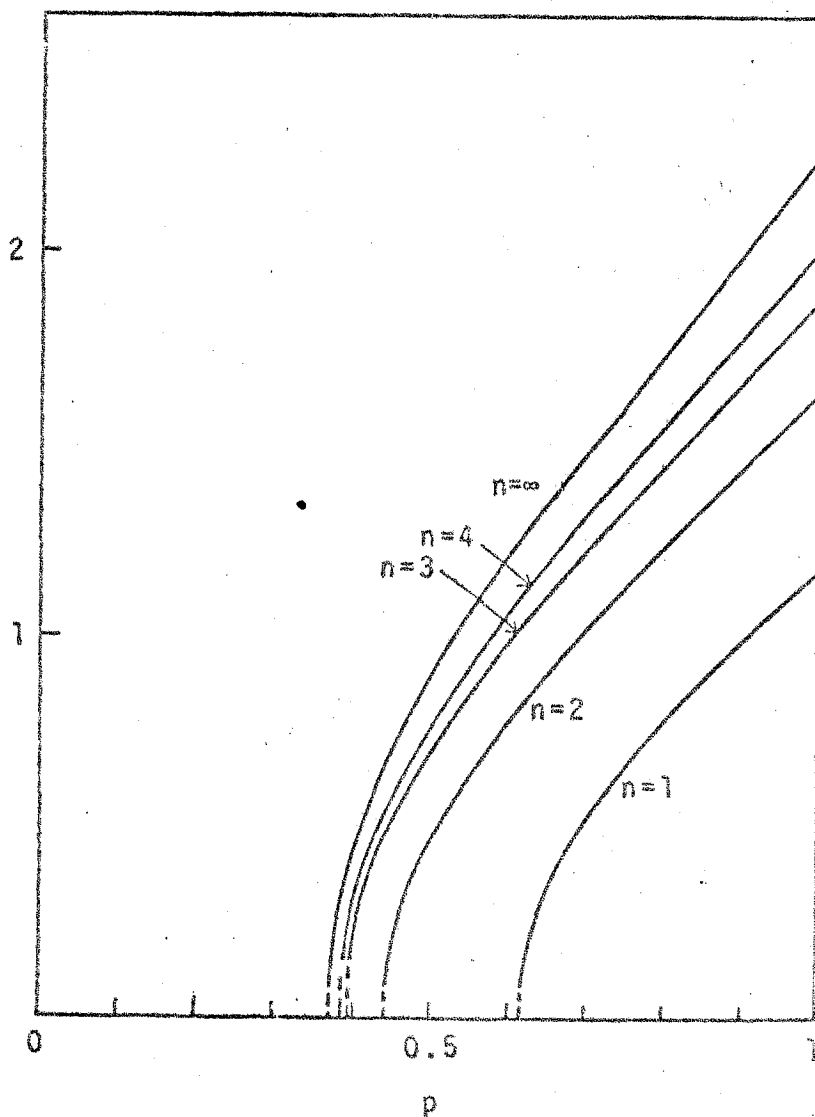


Fig.2-6 The critical exponent γ of the susceptibility for the quenched site model is represented against the concentration of magnetic atoms.

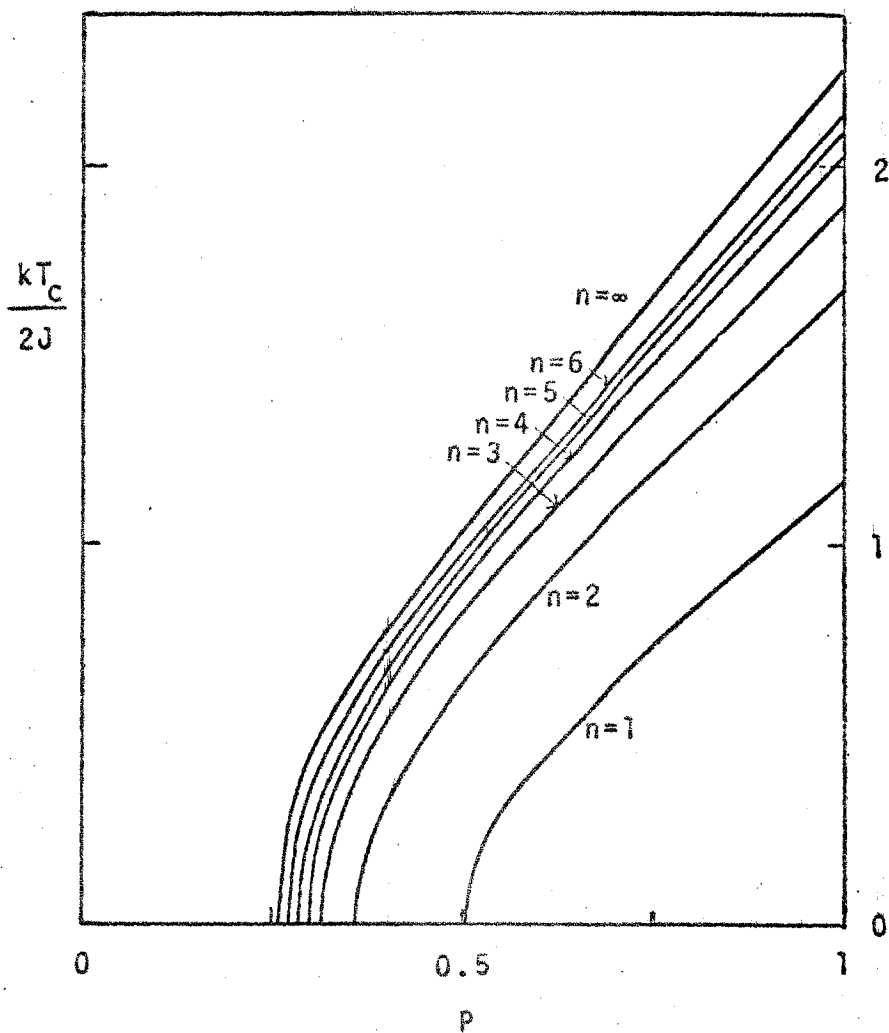
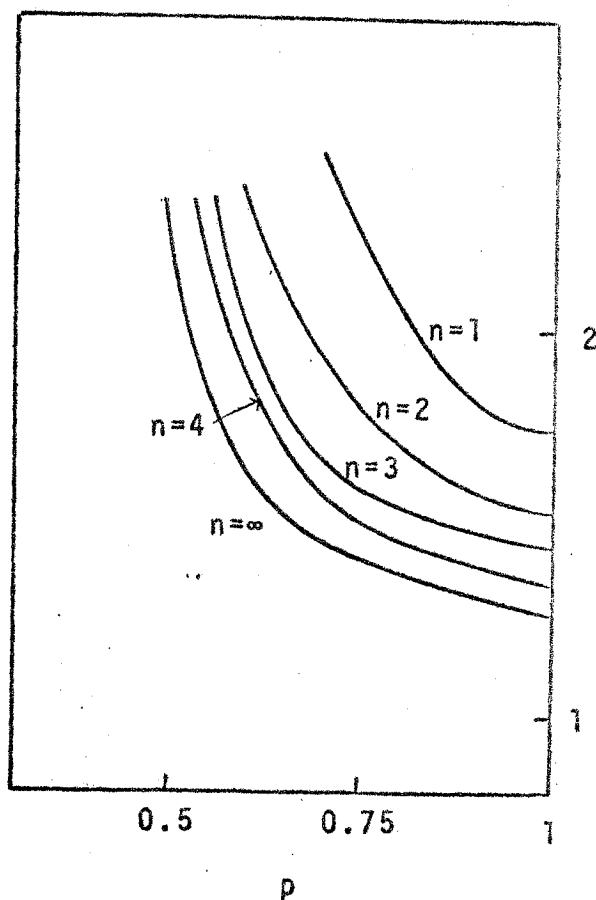
 γ 

Fig.2-7 The critical temperature obtained from the high temperature series expansion for the quenched bond model is represented.

those of the Syozi model, but cannot deny the existence of the above discontinuity because the number of terms in series is small.

In the case of the quenched bond model, the critical temperature is represented in Fig.2-7, where the general trend is the same as in the quenched site model. It is natural that the initial gradients and the critical concentrations for the n-layer lattices are smaller than those for the corresponding quenched site models respectively.

§ 2-3 Size-effect on the critical temperature

In order to investigate the size-effect on the critical temperature, v_c is plotted as a function of $n^{-\lambda}$ ($\lambda = 1.25$) in Fig. 2-8. This analysis has been carried out for the Ising model by Allan and for the Heisenberg model by Ritchie and Fisher.²⁴⁾ They obtained $\lambda = 1.27$ and $\lambda = 1.1$, respectively. It is for the first time that the size-effect in the dilute ferromagnet is analyzed and the dependence of λ on p is found as follows, $\lambda = 1.25$ for $p = 1$, $\lambda = 1.44$ for $p = 0.8$ and $\lambda = 1.75$ for $p = 0.7$. It is difficult to estimate a relation between λ and p from these few data.

Similar analysis of size-effect has been given by Abe,²⁵⁾ who proposed n-layer Ising lattice with intra-layer exchange coupling J and inter-layer one ζJ . The critical temperatures are calculated by the perturbation theory as follows

$$T_c(\zeta) - T_c(\zeta = 0) = A \zeta^{1/\gamma}, \quad (2-23)$$

where A is a constant. Ishikawa and Oguchi²⁶⁾ have investigated the same problem in more general models by the high temperature series expansion of susceptibility.

There are some magnetic materials which show one or two-dimensional orderings. But in the lowest temperature they show three-dimensional properties. Therefore the above analyses are important to investigate magnetic properties, in particular behaviors of the critical exponents γ in the intermediate temperature.

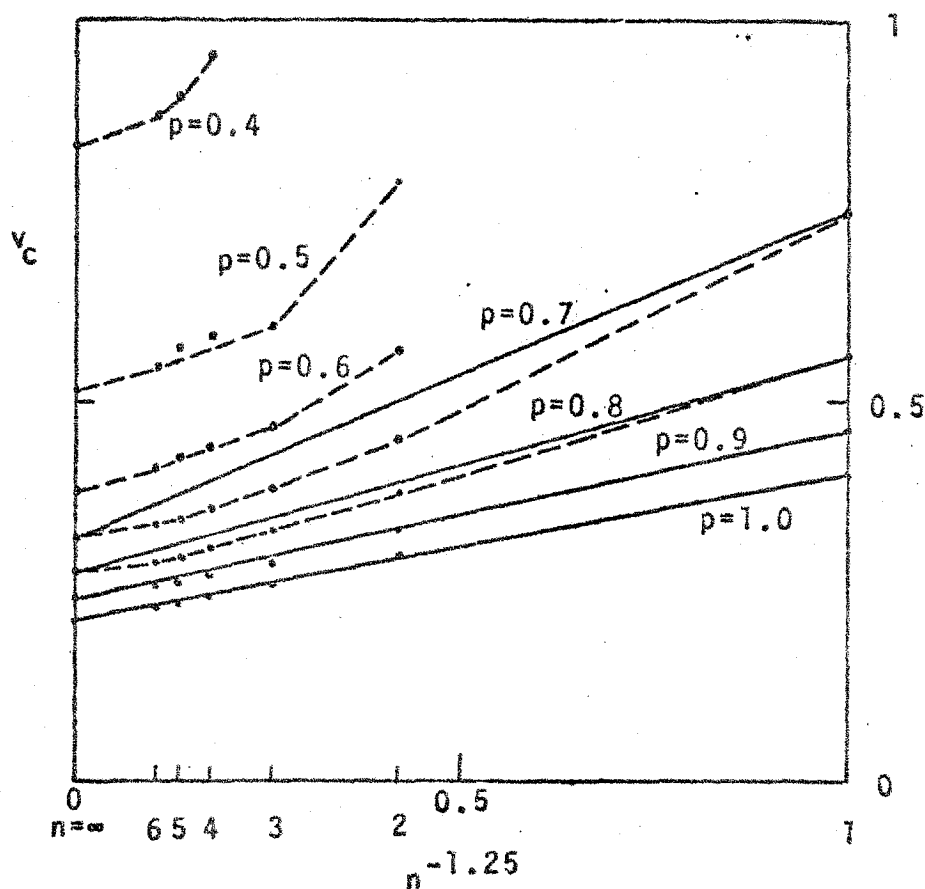


Fig. 2-8 The critical values $v_c = \tanh J/kT_c$ versus $n^{-1.25}$ are plotted. The solid lines connect the two points at $n = 1$ and $n = \infty$. The dashed lines connect the calculated values from the high temperature expansion.

Chapter III Magnetic properties in solid solutions

In this chapter we discuss the magnetic properties of solid solutions which consist of two components of magnetic substances. Annealed systems are discussed in § 3-1 exactly and quenched systems are in § 3-2 by the high temperature approximation. Extension to systems with many components is straightforwardly possible.

§ 3-1 Extended Syozi model

The model for the dilute ferromagnet discussed in § 2-1 can be extended to a following model for solid solutions with random arrangements of two kinds of exchange integrals, which is shown in Fig.3-1. The variables $\mu_i (= \pm 1)$ and $\mu_j (= \pm 1)$ represent the two states of the up and the down Ising spins at i - and j -th sites. The variables $\sigma_{ij} (= 0, 1)$ and $\sigma'_{ij} (= 0, 1)$ are introduced to represent the absence and the presence of a bond with first kind of exchange integral J and the second kind of exchange integral $J' (= \alpha J)$ between i - and j -th sites respectively.

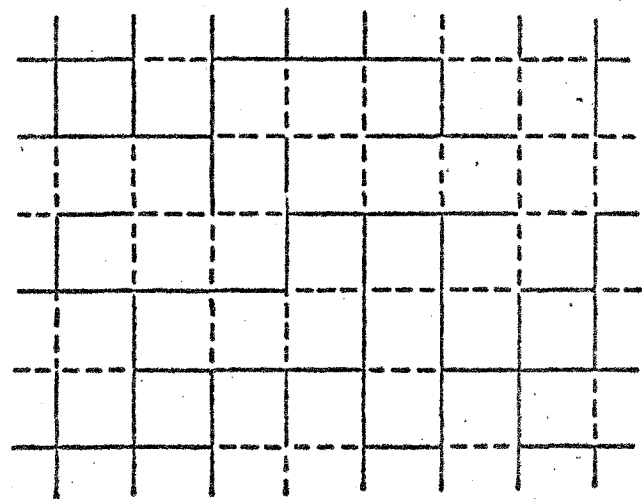


Fig.3-1 One of the bond configurations of the bond model for the magnetic solid solutions is given. The solid bond means the existence of an exchange interaction J between a pair of spins. The dashed bond means the existence of another exchange interaction J' .

The Hamiltonian can be expressed as

$$H = -J \sum_{\langle ij \rangle} \mu_i (\sigma_{ij} + \alpha \sigma'_{ij}) \mu_j . \quad (3-1)$$

The grand canonical partition function is given by introducing ξ and ξ' as follows,

$$\begin{aligned} \hat{Z} = \sum_{\mu} \sum_{\sigma} \sum_{\sigma'} \exp \left\{ \sum_{ij} L \mu_i (\sigma_{ij} + \alpha \sigma'_{ij}) \mu_j \right. \\ \left. + \xi \sigma_{ij} + \xi' \sigma'_{ij} \right\} . \end{aligned} \quad (3-2)$$

By applying the extended iteration transformation,

$$\hat{Z} = A^{zN/2} Z_0(K) , \quad (3-3)$$

where

$$A^2 = (e^L + \xi + e^{\alpha L} + \xi') (e^{-L} + \xi + e^{-\alpha L} + \xi') \quad (3-4)$$

$$e^{2K} = (e^L + \xi + e^{\alpha L} + \xi') / (e^{-L} + \xi + e^{-\alpha L} + \xi') . \quad (3-5)$$

In the similar way as in § 2-1, the average concentration of bonds with first kind of exchange integral is given by

$$p = \frac{1}{2} \frac{\sinh(\alpha L - K)}{e^{L+K} \sinh(\alpha L - L)} \left\{ e^{2L}(1 + \xi) + e^{2K}(1 - \xi) \right\} . \quad (3-6)$$

Other bonds of which concentration is given by $p' = 1 - p$ have the second kind of exchange integral J' . Here if we utilize the critical values for the square lattice obtained by Onsager

$$e^{-2K_c} = \sqrt{2} \mp 1, \quad \xi_c = \pm \frac{\sqrt{2}}{2}, \quad (3-7)$$

we can calculate the critical temperature by (3-6). In (3-7) the critical values with the upper signs are used for the ferromagnetic phase transition and those with the lower signs are used for the antiferromagnetic phase transition. Fig.3-2 represents the critical temperatures of solid solutions with two kinds of antiferromagnetic exchange integral, that is, $J < 0$ and $J' (= \alpha J) < 0$ for several values of α . The curve in the case of $\alpha = 0$ is equivalent to that of the dilute ferromagnet discussed in § 2-1. Fig. 3-3 represents the critical temperature in the case of $J > 0$ and $\alpha = -1$. In this case the ferromagnetic ordered state appears for $\frac{1+\xi_c}{2} < p \leq 1$ and the antiferromagnetic one does for $0 < p < \frac{1-\xi_c}{2}$, where $\xi_c = \sqrt{2}/2$. For $\frac{1-\xi_c}{2} < p < \frac{1-\xi_c}{2}$,

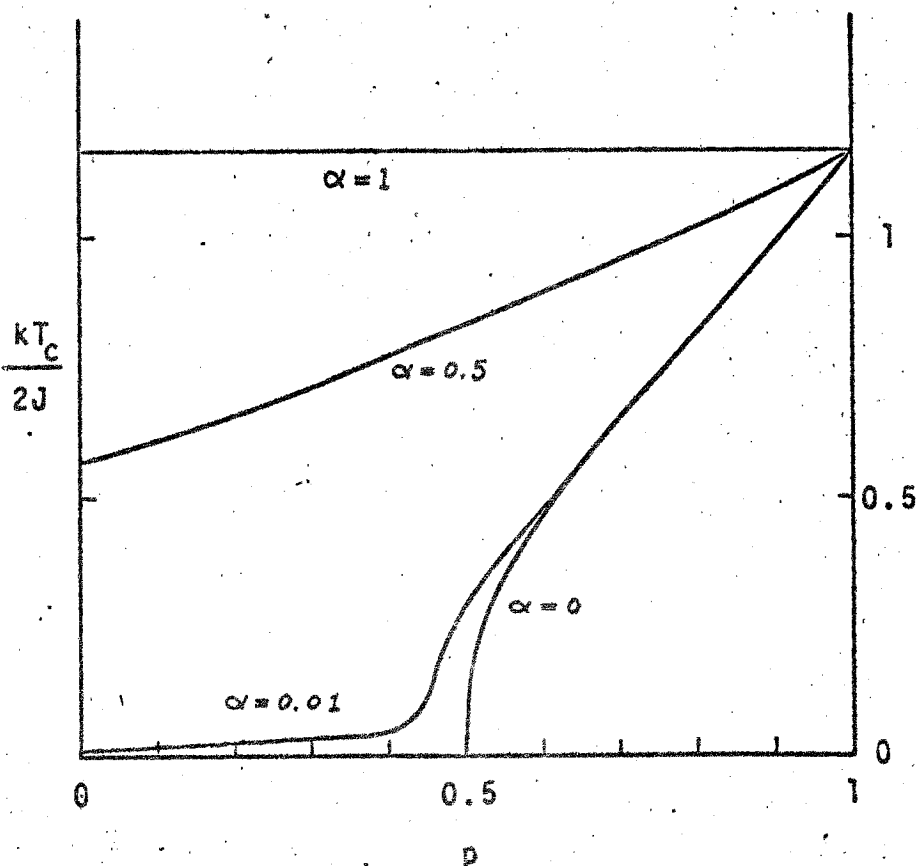


Fig.3-2 The critical temperatures of the annealed model for the magnetic solid solutions are plotted versus the concentration p of bonds.

here is no phase transition.

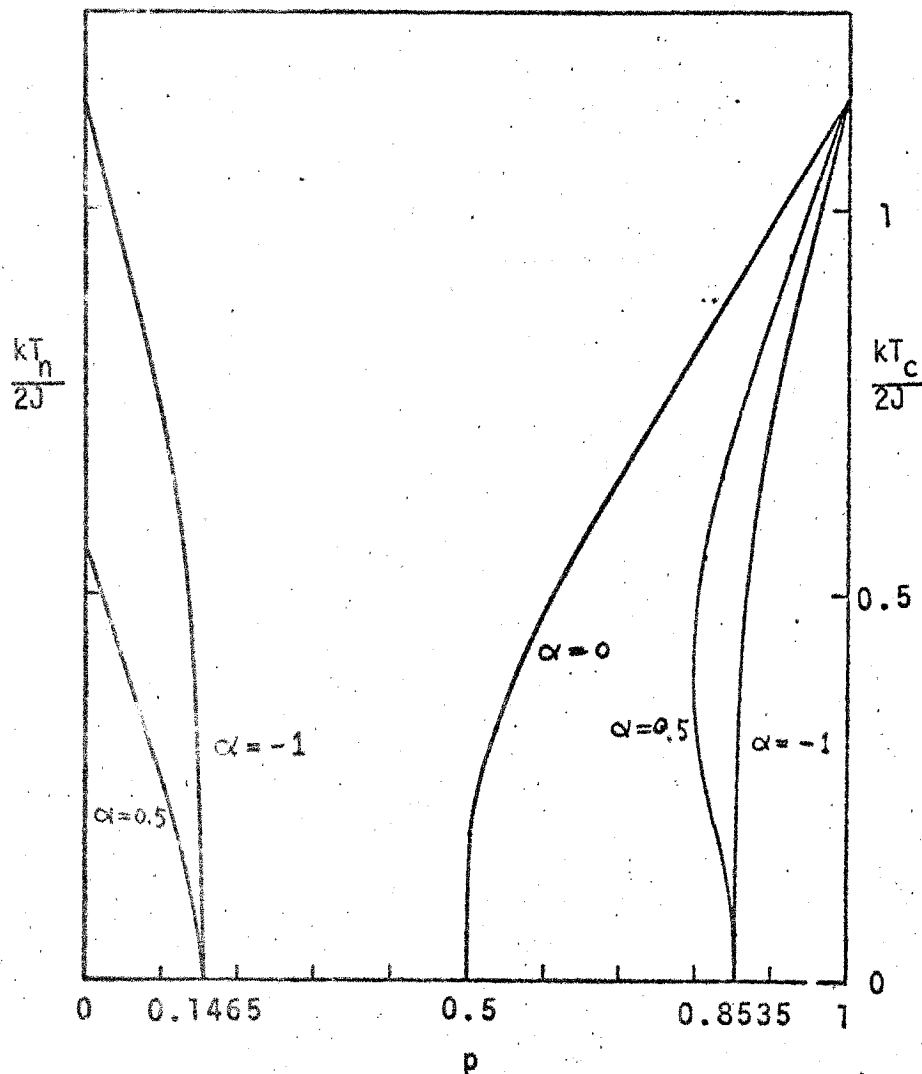


Fig. 3-3 The critical temperatures of the annealed model of the magnetic solid solution are plotted versus the concentration p of bonds with the exchange integral J . Here J is assumed to be positive and J' is negative T_c and T_N are the Curie temperature and the Neel temperature respectively.

The specific heat at the constant concentration p is

$$\begin{aligned}
 C = kL^2 & \left[\frac{d\xi}{dK} \frac{dK}{dL} + (\alpha-1) \left\{ \left(\frac{dK}{dL} - 1 \right) \cosh(K-L) \right\} \sinh(\alpha L-K) + \right. \\
 & \left. \xi \cosh(K-\alpha L) \right\} + \sinh(K-L) \left\{ \left(\alpha - \frac{dK}{dL} \right) \cosh(\alpha L-K) + \frac{d\xi}{dK} \frac{dK}{dL} \right. \\
 & \left. \cosh(K-\alpha L) + \xi \sinh(K-\alpha L) \left(\frac{dK}{dL} - \alpha \right) \right\} \right] / \sinh(\alpha L-L) \\
 & - (\alpha-1)^2 \sinh(K-L) \left\{ \sinh(\alpha L-K) + \xi \cosh(K-\alpha L) \right\} \cosh(\alpha L-L) / \\
 & \sinh^2(\alpha L-L) \Bigg\}, \tag{3-8}
 \end{aligned}$$

where

$$\begin{aligned}
 \frac{dK}{dL} = & \left[p(1-\alpha) \cosh(\alpha L-L) + \alpha \cosh(\alpha L-K) \right\} \cosh(L-K) + \xi \sinh(L-K) \Bigg\} \\
 & + \sinh(\alpha L-K) \left\{ \sinh(L-K) + \xi \cosh(L-K) \right\} \Bigg] / \left[\cosh(\alpha L-K) \left\{ \cosh(L-K) \right. \right. \\
 & \left. \left. + \xi \sinh(L-K) \right\} + \sinh(\alpha L-K) \left\{ \sinh(L-K) - \frac{d\xi}{dK} \sinh(L-K) + \xi \cosh(L-K) \right\} \right] \tag{3-9}
 \end{aligned}$$

This is shown in Fig.3-4. In this model, the specific heat at the critical temperature remains finite as far as $p \neq 0$ or 1 . The same renormalization in the critical exponents as the dilute ferromagnet can be easily understood from (3-9).

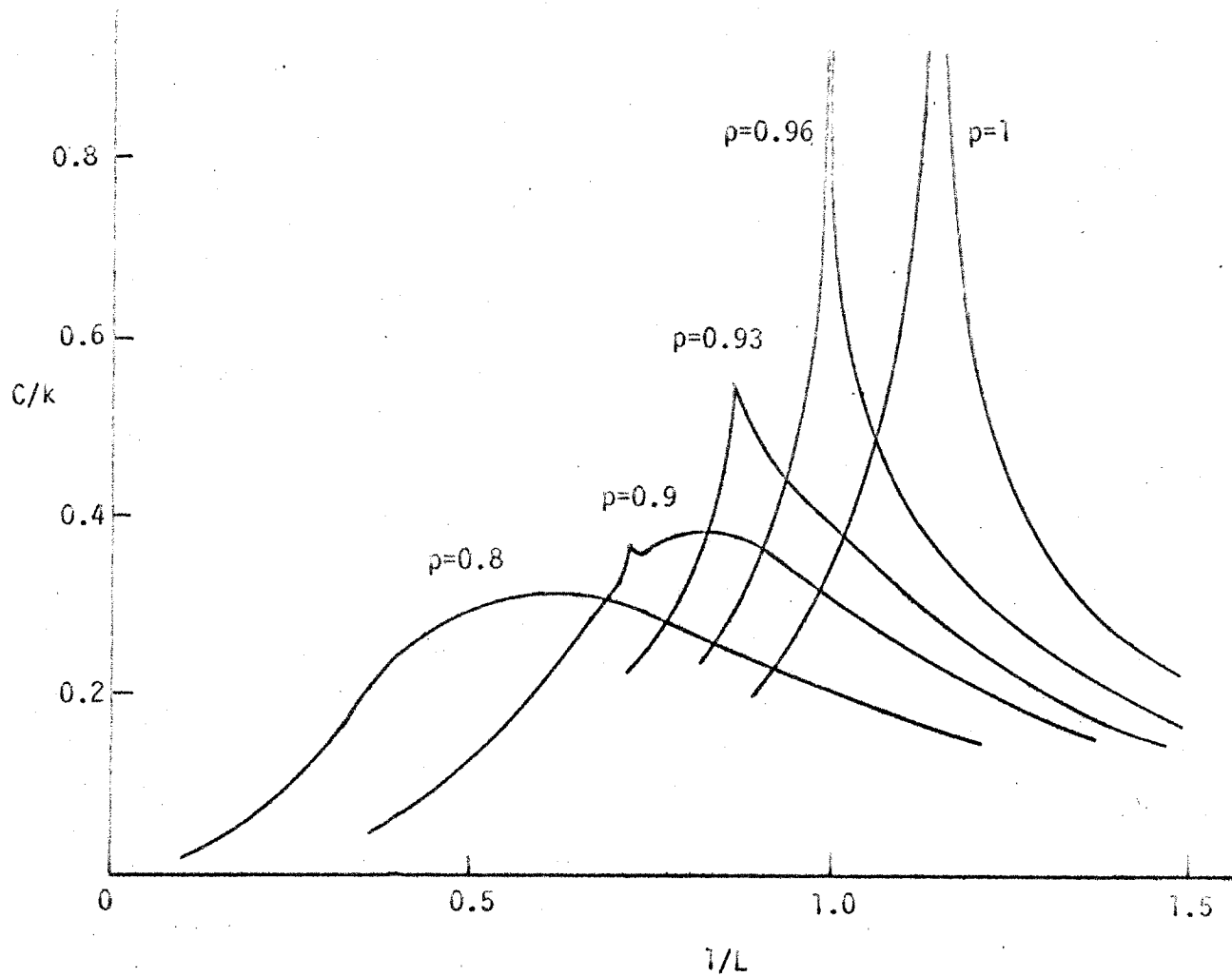


Fig. 3-4 The specific heats of the annealed model for the magnetic solid solutions are given.

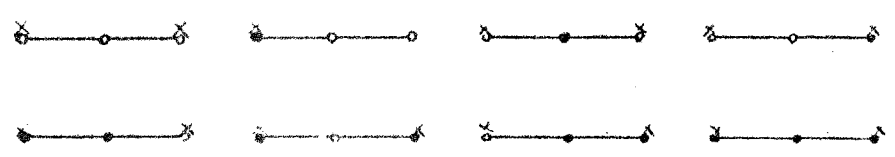
§3-2 A quenched model for magnetic solid solutions

A quenched model for magnetic solid solutions which consist of two component is considered. In a quenched site model, one of two kinds of atoms A or B can occupy a lattice site. Therefore three kinds of exchange integrals J_{AA} , J_{BB} and J_{AB} , which are exchange integrals between a pair of atoms A, a pair of atoms B and a pair of atoms A and B respectively, must be introduced. J_{AB} is not always the average of J_{AA} and J_{BB} , but it can take the larger or smaller value than J_{AA} and J_{BB} . In these cases it is interesting to investigate the behavior of critical temperature as the concentration of atoms A changes. On the other hand we introduce only two kinds of exchange integrals J_A and J_B in a quenched bond model.

The zero field susceptibility in the quenched site model can be expanded as discussed in §2-2. The number of magnetic graphs is same as before for the every order. However the weight for each magnetic graph is different from the dilute ferromagnetic model. As an example, we consider a magnetic graph such as



Two kinds of atoms A and B occupy the three sites, so we have 2^3 different configurations such as



, where o and \bullet mean an A atom and a B atom respectively, and each graph has the following weight v_{AA}^2 , $v_{AA}v_{AB}$, v_{AB}^2 , $v_{AA}v_{AB}$, $v_{AB}v_{BB}$, v_{AB}^2 , $v_{AB}v_{BB}$ and v_{BB}^2 , where $v_{AA} = \tanh J_{AA}/kT$, $v_{AB} = \tanh J_{AB}/kT$ and $v_{BB} = \tanh J_{BB}/kT$. Thus $12p^3v^2$ in Table 1 is replaced by $12x$

$\{v_{AA}^2 p^3 + (2v_{AA}v_{AB} + v_{AB}^2)p^2q + (2v_{BB}v_{AB} + v_{AB}^2)pq^2 + v_{BB}^2q^3\}$. Other coefficients also must be replaced in the similar way. Expanding $\tanh J_{AA}/kT$, $\tanh J_{BB}/kT$ and $\tanh J_{AB}/kT$ in a power series of $1/kT$, we obtain

$$(kT/Nm^2) \chi_0 = 1 + \sum_{r=1}^{\infty} b_r(a_1, a_2, a_3)(1/kT)^r, \quad (3-10)$$

where $a_1 = J_{AA}$, $a_2 = J_{BB}$ and $a_3 = J_{AB}$ for the sake of simplicity. The coefficients b_r are given in Appendix A.

Applying the ratio method to these series, we obtain the critical temperatures which are represented in Fig's 3-5(a), 3-5(b) and 3-5(c) for the cases of $(a_1 = 1, a_2 = 0.5, a_3 = 1.5)$, $(a_1 = 1, a_2 = 0.5, a_3 = 0.75)$ and $(a_1 = 1, a_2 = 0.5, a_3 = 0.25)$ for n -layer Ising lattices ($n = 1, 2, \dots, 6$ and ∞) respectively. The critical temperature shows the peak in the first case ($a_1 = 1, a_2 = 0.5, a_3 = 1.5$) and has the minimum point in the third case ($a_1 = 1, a_2 = 0.5, a_3 = 0.25$) as expected. In the second case ($a_1 = 1, a_2 = 0.5, a_3 = 0.75$) the critical temperature decreases monotonically from a critical temperature at $p = 1$ to that at $p = 0$. The variations of phase transition temperature similar to the first case are found in the solid solution of MnF_2 and FeF_2 ²⁷⁾.

The critical exponents γ are represented in Fig. 3-6. The calculated value in the three-dimensional lattice is constant as expected. Those values in other lattices show small fluctuations, but it should be concluded to be constant. Further the values for the n -layer lattice with finite n should be the value of the two-dimensional lattice.

The susceptibility for the quenched bond model is given by

$$(kT/Nm^2) \chi_0 = 1 + \sum_{r=1}^{\infty} b'_r(a_1, a_2)(1/kT)^r, \quad (3-11)$$

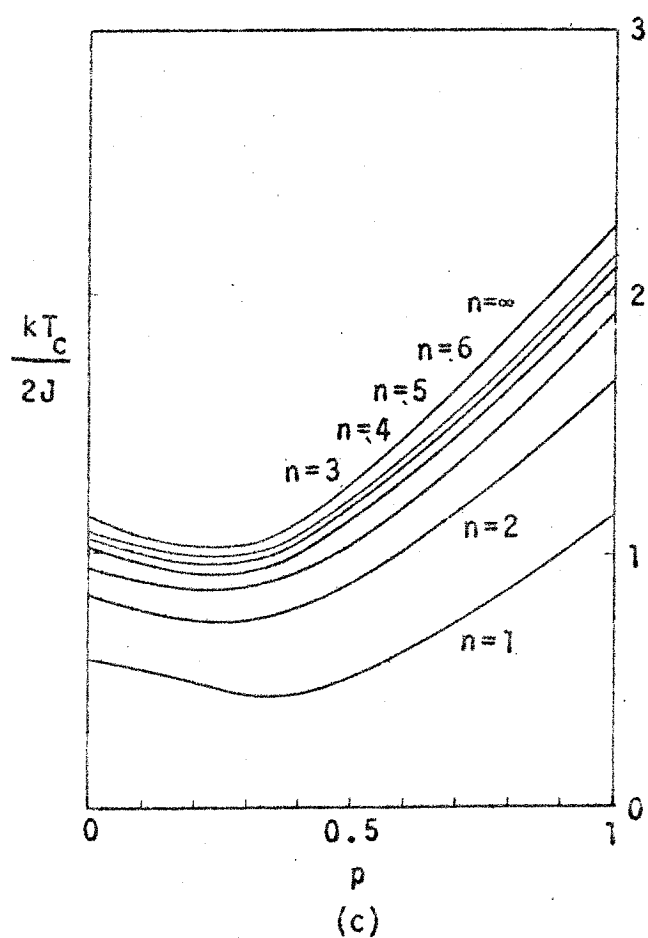
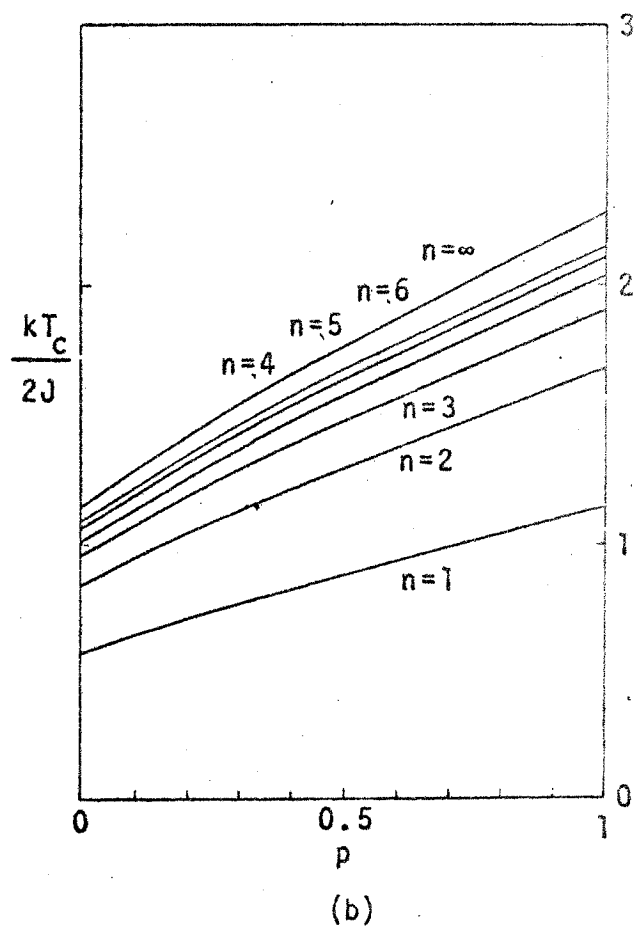
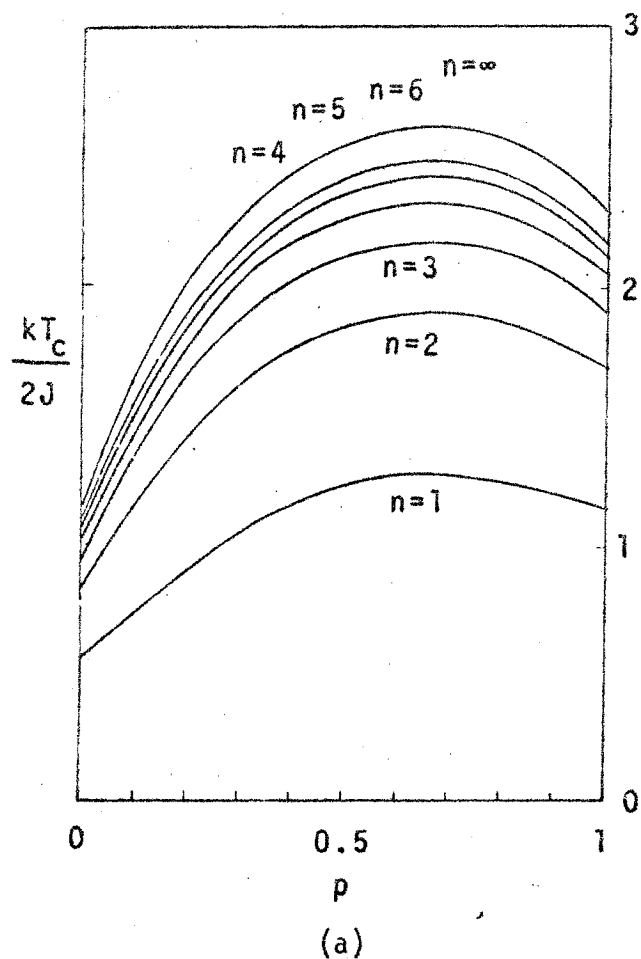


Fig.3-5 The critical temperature of the quenched site model for n -layer Ising lattice is given against the concentration of magnetic atoms. (a) is for $(a_1=1, a_2=0.5, a_3=1.5)$, (b) for $(a_1=1, a_2=0.5, a_3=0.75)$, and (c) for $(a_1=1, a_2=0.5, a_3=0.25)$.

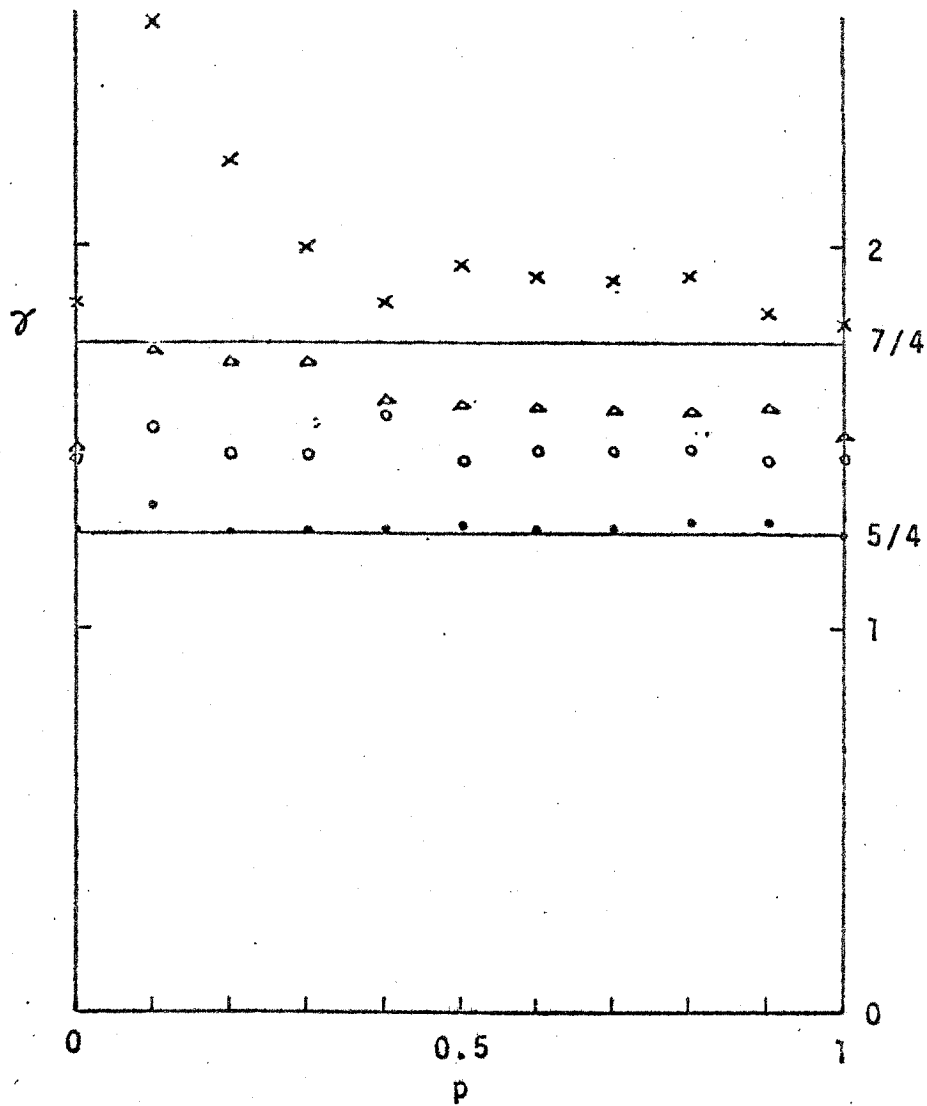


Fig.3-6 The critical exponents γ of n -layer Ising lattices treated as the quenched site model are shown in the case of $a_1 = 1$, $a_2 = 0.5$ and $a_3 = 1.5$. \times , Δ , \circ and \bullet are for $n = 1, 2, 3$ and ∞ respectively.

where the coefficients b'_p are given in Appendix B. The critical temperatures of the various cases as in the quenched bond model are represented by Fig.3-7. The critical exponents are shown in Fig.3-8.

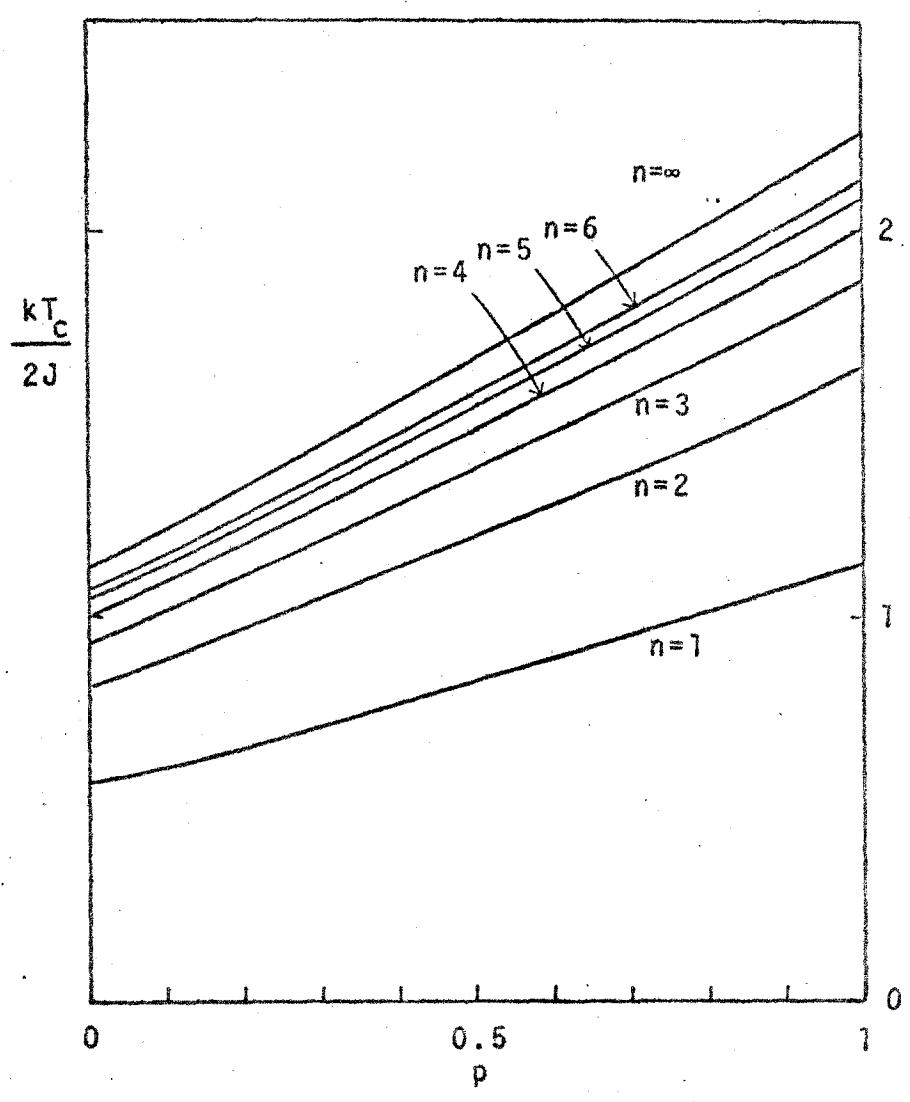


Fig. 3-7 The critical temperatures of the quenched bond model for the magnetic solid solutions are given in the case of $a_1 = 1$ and $a_2 = 0.5$. n is the number of layers of the square lattices.

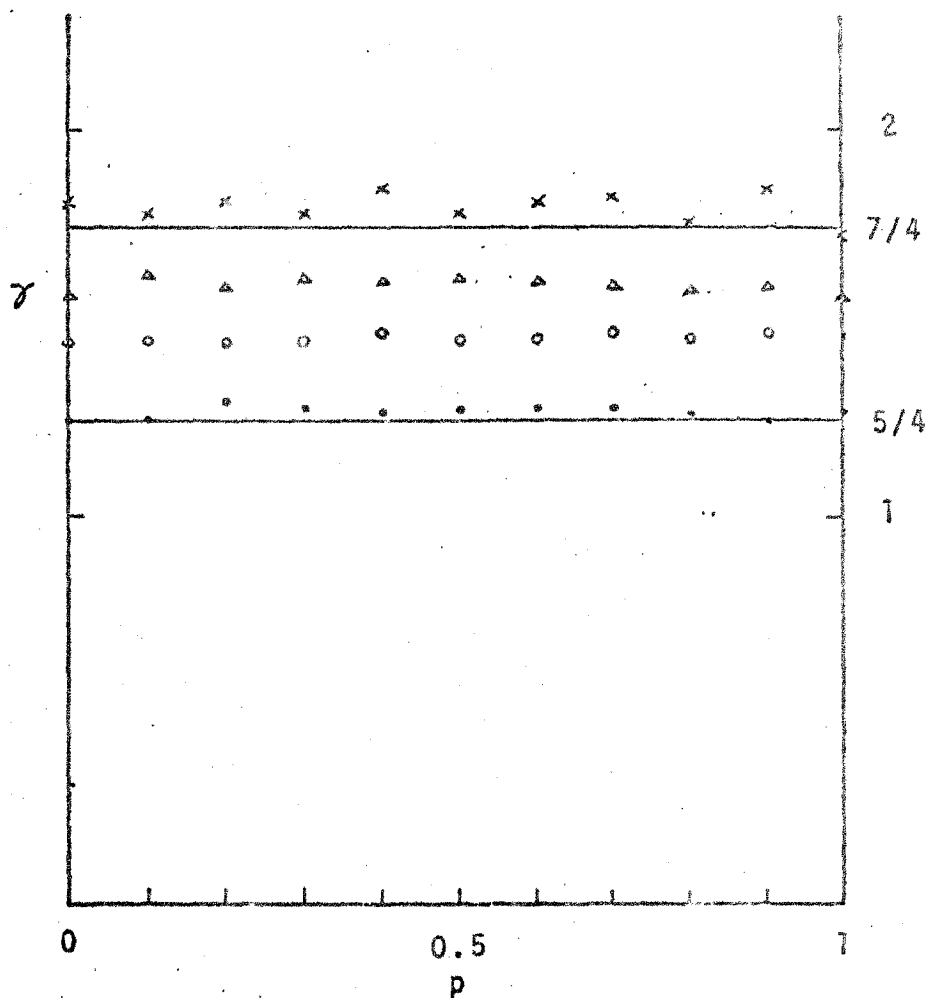


Fig. 3-8 The critical exponents γ of n -layer Ising lattices treated as the quenched bond model are shown in the case of $a_1 = 1$ and $a_2 = 0.5$. \times , Δ , O and \bullet are for $n = 1, 2, 3$ and ∞ respectively.

§ 3-3 Size-effect on the critical temperature

When the dimensionality changes from two- to three-dimensional lattices, the behavior of critical temperature is discussed. In Fig.3-9, the shifts of critical values v_c are represented against $n^{-1.25}$. The calculated critical values v_c show good fit with lines connecting the two critical values for the two- and three-dimensional lattices.

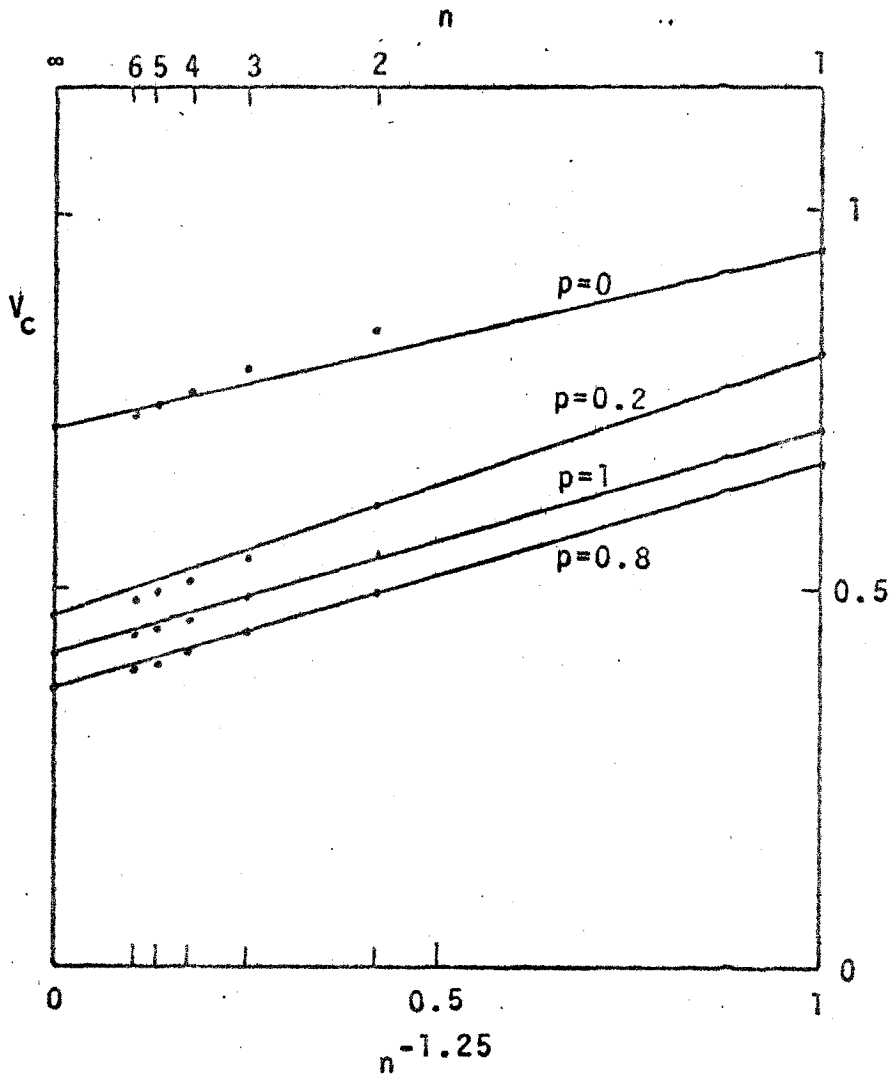


Fig. 3-9 The critical values $v_c = \tanh J/kT_c$ versus $n^{-1.25}$ are plotted.

Chapter IV Critical exponents and some rigorous results in random systems

The dimensionality of systems, the interaction range and the type of interaction affect the critical exponents which characterize the singularities of the specific heat, the susceptibility, the correlation length and so on. Now we discuss the critical exponents in the random ferromagnet, especially the dilute ferromagnets already discussed in chapter II and the magnetic solid solutions which appeared in chapter III.

Some theorems concerning to the dilute ferromagnet (the percolation problem) which have been proved by Griffiths, Lebowitz, Fisher and so on are briefly explained. The existence of critical concentration has been proved by Griffiths and the critical concentration and exponents have been actually calculated by Syozi in the annealed bond model for the dilute ferromagnet.

{ 4-1 Renormalization of the critical exponents

Essam and Garelick²⁸⁾ have discussed this problem in the Syozi model with the magnetic field and obtained the following results for α' , β' and γ' , which are the critical exponents for the specific heat, the spontaneous magnetization and the susceptibility respectively,

$$\alpha' = \alpha/(1-\alpha), \quad \beta' = \beta/(1-\alpha), \quad \gamma' = \gamma/(1-\alpha), \quad p_c < p < 1, \quad (4-1)$$

where α , β and γ are the critical exponents for the original Ising model. Rapaport²⁹⁾ also has discussed the same problem in the annealed bond model modified by Kano and Miyazima⁸⁾ and obtained the same results as above. This is explained briefly as follows.

In the specific heat, essential part is $\frac{\partial \epsilon}{\partial K}$, which in the three-dimensional Ising model has the form in the vicinity of the critical point K_c ,

$$\frac{\partial \xi}{\partial K} = A_0 \left(1 - \frac{K}{K_c}\right)^{-\alpha} + A_1 + \dots, \quad K \leq K_c, \quad (4-2)$$

where A_0 and A_1 are constant.

Integrating (2-14) with respect to K , and the result is found to be

$$\frac{T}{T_c} - 1 \simeq \phi_1 \left(1 - \frac{K}{K_c}\right) + (1-p) \phi_2 \left(1 - \frac{K}{K_c}\right)^{1-\alpha} + \dots, \quad (4-3)$$

neglecting the term of higher order. ϕ_1 and ϕ_2 can be shown to be positive when $p_c < p < 1$. Thus we obtain

$$1 - \frac{K}{K_c} = \left\{ (1-p) \phi_2 \right\}^{-1/(1-\alpha)} \left(\frac{T}{T_c} - 1 \right)^{1/(1-\alpha)}, \quad (4-4)$$

and using (4-2) and (4-4) we get

$$C_p \simeq A_1 - B_2 \left(\frac{T}{T_c} - 1 \right)^{\alpha/(1-\alpha)} \quad \text{for } T > T_c. \quad (4-5)$$

When the leading term of susceptibility has a form,

$$\chi_0 \simeq C_0 \left(1 - \frac{K}{K_c}\right)^{-\gamma}, \quad (4-6)$$

we obtain for the dilute ferromagnet by (4-4),

$$\chi_0 \simeq C_0 \left\{ (1-p) \phi_2 \right\}^{\gamma/(1-\alpha)} \left(\frac{T}{T_c} - 1 \right)^{-\gamma/(1-\alpha)}. \quad (4-7)$$

The renormalized exponent for the spontaneous magnetization can be obtained in the similar way as above. It must be noted that each renormalized exponent is constant for $p_c < p < 1$ and has a gap at $p = 1$. As shown in Fig.4-1, Rapaport has calculated the critical exponents γ for the susceptibility in the annealed bond and the quenched bond models by means of the high temperature series expansion. The critical exponent γ increases gradually as p decreases and about at $p = 0.2$ it is close to the expected value of $10/7$. It appears that they continue to increase. This strange behavior obtained by studying

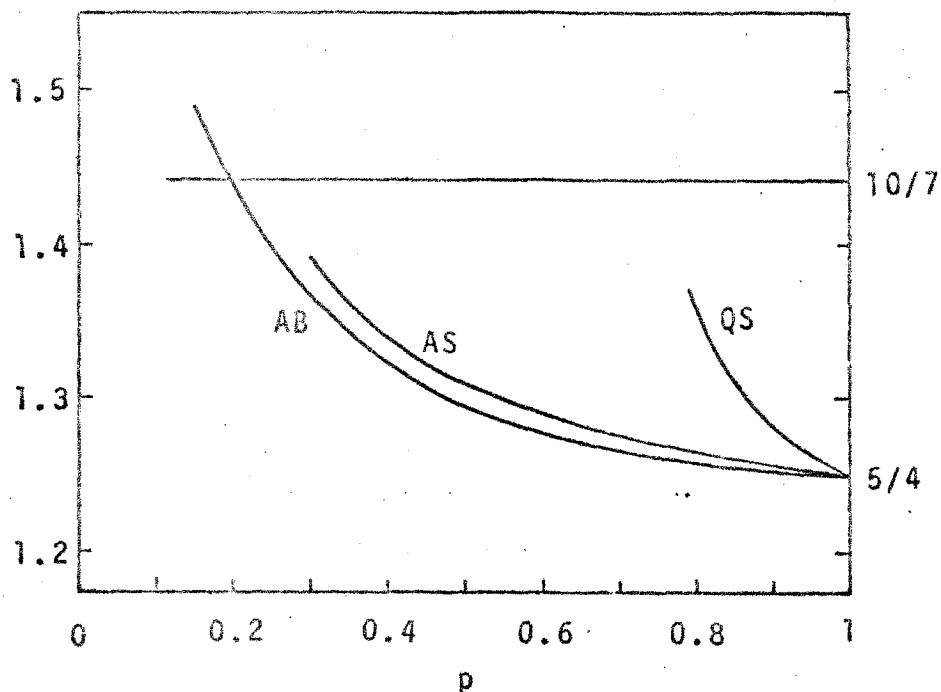


Fig. 4-1 An analysis of the critical exponent γ of the Syozi model by the high temperature approximation are represented for the fcc lattice. AB, AS and QS mean the annealed bond, the annealed site and the quenched site model respectively.

the series expansion can be understood as the confluent singularity. If the series $F(z) = \sum_{n=0}^{\infty} d_n z^n$ has the following behavior near the closest real singularity z_c , such as

$$F(z) \sim e_1 \left(1 - \frac{z}{z_c}\right)^{-\mu_1} + e_2 \left(1 - \frac{z}{z_c}\right)^{-\mu_2} \quad \text{for } z \lesssim z_c, \quad (4-8)$$

where $\Delta = \mu_1 - \mu_2$ is small positive and the coefficients e_1 and e_2 are of similar magnitude, the ratios $r_n = d_n/d_{n-1}$ will be expected to have the form

$$r_n \sim z_c^{-1} \left(\frac{n}{n-1}\right) \frac{1 + g n^{-\Delta}}{1 + g (n-1)^{-\Delta}} \quad \text{for } n \gg 1, \quad (4-9)$$

where $g = e_2 \Gamma(\mu_1)/e_1 \Gamma(\mu_2)$ and $\Gamma(x)$ is a function of x .

From (4-9) ,

$$\begin{aligned} r_n &\sim z_c^{-1} \left(1 + \frac{\mu_1 - 1}{n} - \frac{\Delta g}{n g + n^4} \right) + O(n^{-2}) \\ &\sim z_c^{-1} \left(1 + \frac{\bar{\mu}_1 - 1}{n} \right) + O(n^{-2}) , \end{aligned} \quad (4-10)$$

where $\bar{\mu}_1 = \mu_1 - \Delta g/(g+1)$. Thus a new exponent $\bar{\mu}_1$ arises out of the interference of the term of (4-8) . Therefore it is difficult to estimate the critical exponents by the ratio method in the dilute ferromagnet such as the annealed bond model because of the above reason.

As seen in § 2-2, the critical exponent γ estimated by the series for the quenched model increases monotonically as p decreases. If

Δ is assumed to decrease when p decreases, we cannot find the true critical exponent . We cannot decide if the present calculation reveals the true behavior of the critical exponent.

{ 4-2 Critical exponents of dilute ferromagnet with four-spin interaction

In this section we give another exactly soluble eight-vertex model and discuss the critical exponents of diluted eight-vertex model. The eight-vertex model which was solved by Baxter³⁰⁾, includes the Ising model, the Slater model for ferroelectricity³¹⁾, P-model for³²⁾ antiferroelectricity and most of other exactly soluble model as special cases.

The eight vertex model has an arrow on each bond of the square lattice. An arrow takes two states, up and down direction on the vertical bond and right and left on the horizontal bond. Even numbers of entering arrows at each vertex are permitted. There are the following eight kinds of configurations of arrows with energies ϵ_1 , ϵ_2 , ϵ_3 and ϵ_4 as shown in Fig.4-2. The free energy is

$$-kTf = -kT\epsilon_3 + 2 \sum_{n=1}^{\infty} \frac{\sinh^2[(\tau-\lambda)n]}{n \sinh(2n\tau) \cosh(n\lambda)} \{ \cosh(n\lambda) - \cosh(n\alpha) \}, \quad (4-11)$$

where $\tau = \pi K_1/K_1'$, $\lambda = \pi \zeta/K_1'$, $\alpha = \pi V/K_1'$, and $\ell' = (1-\ell)^{-1/2}$. ℓ , ζ and V are determined from ϵ_1 , ϵ_2 , ϵ_3 and ϵ_4 . (see ref. 30)

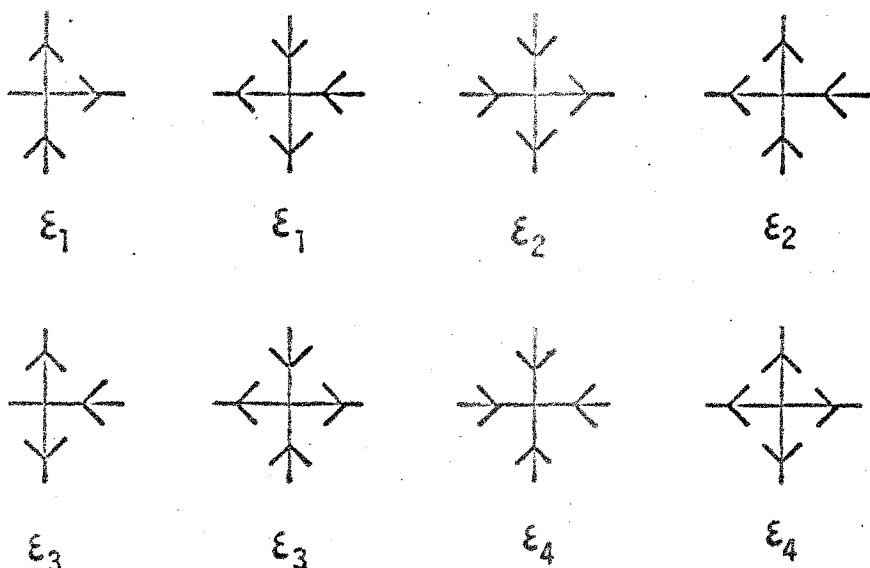


Fig.4-2 Allowed configurations of arrows about a vertex with energy assignment.

Kadanoff and Wegner²³⁾ showed that the eight-vertex model is equivalent to a two-planar Ising model interacting with a four-spin interaction, as shown in Fig.4-3, whose Hamiltonian is given by

$$H = - \sum_{j,k} (J \sigma_{jk} \sigma_{j+1,k+1} + J \sigma_{j+1,k} \sigma_{jk+1} + \lambda \sigma_{jk} \sigma_{j+1,k+1} \sigma_{j+1,k} \sigma_{jk+1}). \quad (4-12)$$

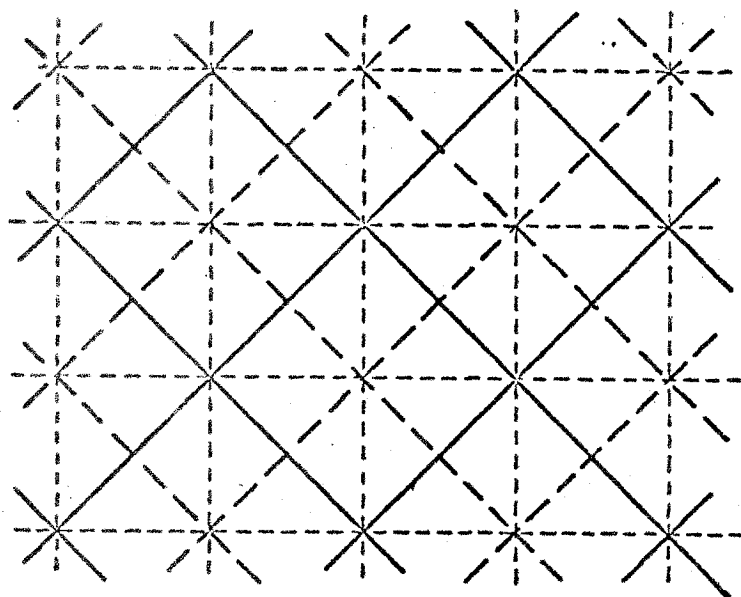


Fig. 4-3 A two-planar Ising lattice which is equivalent to the eight-vertex model. The two square lattices with the ordinary exchange interaction are represented by the solid and the dashed lines. A unit cell of the dotted square lattice represents the four-spin interaction.

The critical temperature of this system is determined by

$$\exp(2\Lambda) \sinh(2K) = 1, \quad (4-13)$$

where

$$K = J/kT, \quad \Lambda = \lambda/kT.$$

The critical exponents are calculated in the weak coupling limit.

$$\gamma/\gamma_0 = \beta/\beta_0 = \nu/\nu_0 = 1 - 4\Lambda/\pi kT_c, \quad (4-14)$$

where γ_0 , β_0 and ν_0 are the exponents of the susceptibility, the spontaneous magnetization and correlation length for the square lattice.

Now we investigate two cases of dilution problem in this two-planar lattice with four-spin interaction; in the first case only ordinary spin interactions are diluted and in the second case units of four-spin interactions are diluted. As following to {2-1 and {3-1 we introduce variables $\nu_{j,k}$ which take 1 and 0, corresponding to whether an interaction unit exists or not. The partition functions of two models are

$$\begin{aligned} \Xi_1 = \sum_{\sigma, \nu} \left[\exp \left\{ \sum_{j,k} (K \sigma_{j,k} \sigma_{j+1,k+1} \nu_{jk} + K \sigma_{j+1,k} \sigma_{j,k+1} \nu_{j+1,k} \right. \right. \\ \left. \left. + \xi_1 \nu_{jk} + \xi_1 \nu_{j+1,k} + \Lambda \sigma_{j,k} \sigma_{j+1,k+1} \sigma_{j+1,k} \sigma_{j,k+1}) \right\} \right], \end{aligned} \quad (4-15)$$

$$\begin{aligned} \Xi_2 = \sum_{\sigma, \nu} \left[\exp \left\{ \sum_{j,k} (K \sigma_{j,k} \sigma_{j+1,k+1} + K \sigma_{j+1,k} \sigma_{j,k+1} \right. \right. \\ \left. \left. + \xi_2 \nu_{jk} + \Lambda \sigma_{j,k} \sigma_{j+1,k+1} \sigma_{j+1,k} \sigma_{j,k+1} \nu_{jk}) \right\} \right], \end{aligned} \quad (4-16)$$

where ξ_1 and ξ_2 are chemical potentials which are divided by temperature. Applying the iteration transformation,

$$\Xi_1 = \sum_{\{\sigma\}} \prod_{j,k} \{ A_1 \exp(K' \sigma_{jk} \sigma_{j+1k+1} + K' \sigma_{j+1k} \sigma_{jk+1} + \Lambda \sigma_{jk} \sigma_{j+1k+1} \sigma_{j+1k} \sigma_{jk+1}) \}, \quad (4-17)$$

$$\Xi_2 = \sum_{\{\sigma\}} \prod_{j,k} \{ A_2 \exp(K \sigma_{jk} \sigma_{j+1k+1} + K \sigma_{j+1k} \sigma_{jk+1} + \Lambda' \sigma_{jk} \sigma_{j+1k+1} \sigma_{j+1k} \sigma_{jk+1}) \}, \quad (4-18)$$

where

$$A_1^2 = \{ \exp(K + \xi_1) + 1 \} \{ \exp(-K + \xi_1) + 1 \}, \quad (4-19)$$

$$\exp(2K') = \{ \exp(K + \xi_1) + 1 \} / \{ \exp(-K + \xi_1) + 1 \}, \quad (4-20)$$

$$A_2^2 = \{ \exp(\Lambda + \xi_2) + 1 \} \{ \exp(-\Lambda + \xi_2) + 1 \}, \quad (4-21)$$

$$\exp(2\Lambda') = \{ \exp(\Lambda + \xi_2) + 1 \} / \{ \exp(-\Lambda + \xi_2) + 1 \}. \quad (4-22)$$

In the same way as before, the critical temperature at p is calculated from a set of equations for the first case,

$$\exp(2\Lambda) \sinh 2K' = 1, \quad (4-23)$$

$$\exp(2K_c) = \frac{2p + \{ \exp(2K') - 1 \} \{ 1 - \xi_2(K', \Lambda) \}}{2p - \{ 1 - \exp(-2K') \} \{ 1 + \xi_2(K', \Lambda) \}}, \quad (4-24)$$

where ξ_2 is the pair correlation in the two-planar Ising lattice with the four-spin interaction. For the second case it is given by

$$\exp(2\Lambda') \sinh 2K = 1, \quad (4-25)$$

$$\exp(2\lambda/kT_c) = \frac{2p + \{ \exp(2\Lambda') - 1 \} \{ 1 - \xi_4(K, \Lambda') \}}{2p - \{ 1 - \exp(-2\Lambda') \} \{ 1 + \xi_4(K, \Lambda') \}}, \quad (4-26)$$

where ξ_4 is the four-spin correlation.

For the variation of critical exponent ν we have

$$\nu(p, \lambda) = \nu(0,0) \left\{ 1 + 4 \lambda / \pi k T_c(p,0) \right\} \quad \text{for the first case,} \quad (4-27)$$

$$\nu(p, \lambda) = \nu(0,0) \left\{ 1 - 4 \Lambda'(p, \lambda) / \pi \right\} \quad \text{for the second case.} \quad (4-28)$$

Other exponents β , ν are renormalized in the similar way. Here it is noted that the above exponent ν increases or decreases with decrease of the concentration p according as Λ is positive or negative, since the transition temperature T_c decreases monotonically as p decreases.

McCoy and Wu³⁴⁾ proposed an exactly solvable quenched bond model in which the horizontal exchange energies are equal and all the vertical interaction $J(j)$ between j -th and $(j+1)$ st row are equal but $J(j)$ is allowed to depend on j . $J(j)$ are treated as independent random variables and each is described by the same temperature independent probability density $P(J)$. The specific heat is shown to have a singularity at a certain T_c , at which it is non-analytic but infinitely differentiable.

§ 4-3 Critical exponents in the percolation problem

Kasteleyn and Fortuin³⁵⁾ proved the following analogies between the percolation problem and the Ising model,

pair connectedness \longleftrightarrow pair correlation function,

mean number of clusters \longleftrightarrow free energy,

percolation Probability $P(p) \longleftrightarrow$ magnetization

mean size of finite cluster $S(p) \longleftrightarrow$ zero field susceptibility

Extending this analogies, they get that the fluctuation of mean number of cluster $K(p)$ corresponds to the specific heat. When $p \rightarrow p_c^+$, $K(p)$, $P(p)$ and $S(p)$ behave as

$$\begin{aligned} K(p) &\sim (p - p_c)^{-\alpha'_p} \\ P(p) &\sim (p - p_c)^{\beta_p} \\ S(p) &\sim (p - p_c)^{-\gamma'_p} \end{aligned} \quad (4-29)$$

And the critical exponents α'_p , β_p and γ'_p satisfy the inequality

$$\alpha'_p + 2\beta_p + \gamma'_p \geq 2, \quad (4-30)$$

which is first derived by Rushbrooke³⁶⁾ for the critical exponents for the ferromagnet.

§4-4 Rigorous theorems on the dilute ferromagnet

Although we have no exact solution of the site problem even in the two-dimensional square lattice, following exact results for this random model have been proved by Griffiths and Lebowitz.¹⁰⁾

(1) If $|J_{ij}(r_{ij})| \leq c/r_{ij}^{d+\epsilon}$; $c, \epsilon > 0$, the limiting free energy per site

$$\lim_{|V| \rightarrow \infty} |V|^{-1} \sum_{V' \subseteq V} \pi(V') F(V, G) \quad (4-31)$$

exists for a sequence of d -dimensional lattices of sufficiently regular shape and is a continuous function of the concentration p with the usual convexity properties. Here r_{ij} is the distance between sites i and j , $\pi(V')$ is the probability that the vertices V' are occupied and $V-V'$ are unoccupied. If $H(V', G)$ is the Hamiltonian, the free energy is given by

$$-\beta F(V', G) = \log \text{tr} e^{-\beta H(V', G)} \quad (4-32)$$

(2) For a ferromagnetic Ising interaction of finite range the limiting free energy is analytic in p and H_0 for $H_0 \neq 0$ and $0 \leq p \leq 1$.

(3) For the nearest neighbor ferromagnetic Ising interaction on a plane square or simple cubic lattice, the spontaneous magnetization exists at sufficiently high concentration and low temperatures. They obtained about 0.985 for the critical concentration for the plane square lattice. This estimation is not good. Our estimation of the critical concentration for the plane square lattice by the high temperature series expansion is about $1/2$.

(4) The existence and analyticity of the correlation functions for $H_0 \neq 0$ and $0 \leq p \leq 1$ are proved.

(5) The spontaneous magnetization is monotonically increasing function of p , $1/T$ and H . Thus $T_c(p)$ is also a monotonically increasing function of p .

(6) For nearest neighbor interactions, the concentration p_0 at which the spontaneous magnetization appears at $T = 0$ is greater than or equal to the critical concentration p_c for the site problem. The critical temperature for $\alpha > 1$ as shown in Fig.3-3 shows the strange behavior and the Ising lattice has the spontaneous magnetization in the intermediate temperature. These Ising lattices, including the Ising models with the many phase transition temperatures, may have any different ordering in the paramagnetic region at the lower temperature and may have any correspondence each other.

Chapter V Concluding Remarks

§ 5-1 Conclusions

The annealed bond models for the dilute ferromagnet and the magnetic solid solution are exactly solved and the expression of several thermodynamic quantities has been obtained, and the quenched site and the quenched bond models are discussed by the high temperature series expansion. The critical temperature versus concentration curves have similar character between these models. As the non-magnetic impurities are substituted in, the critical temperature begins to decrease linearly depending on p . The gradient of critical temperature near $p = 1$ depends on the number of the nearest neighbor sites in the site model. The larger is the number of the nearest neighbor sites, the steeper is the gradient. Just above p_c the behavior of the critical temperature is expressed by $-\{\log(p - p_c)\}^{-1}$ in the case of the annealed bond model. This property is also found in the case of the quenched model. In general the gradient in the bond model is smaller than that in the site model.

An inequality for the critical concentrations $p_c^{(B)} \leq p_c^{(S)}$, which is proved in the next section, is also satisfied with the present bond and site models. The critical exponents in the annealed model of the dilute ferromagnet are renormalized but constant as seen already. On the other hand the critical exponent in the quenched model increases with the decrease of p calculated from the high temperature series of the zero field susceptibility. In order to establish this increasing exponent in the quenched model for the dilute ferromagnet it is necessary much longer series of susceptibility. In the case of the magnetic solid solution we obtain the constant exponent in both the annealed and the quenched models.

The size-effect on the critical phenomena is discussed and a simple relation between the shift of critical temperature and critical concentration from three-dimensional values and the number of layers $\varepsilon \sim n^{-\lambda}$ is established. Ritchie and Fisher conjectured that $\lambda \sim 1/\nu$ where ν is the critical exponent of coherent length. Here we obtain the p-dependent λ . This is an evidence of the p-dependent critical exponents, judging from the scaling law.

It is also shown that a pile of some layers of two-dimensional Ising lattice which does not show any spontaneous magnetization for $p < p_c$ has a possibility of having the spontaneous magnetization.

§ 5-2 The site problem and the bond problem

The percolation problem introduced in §1-3 is formulated in term of the linear graph theory and the critical probabilities (which is equivalent to the critical concentration in the dilute ferromagnet) in the site and bond problems are discussed.

Let's consider a general linear graph $G = (V, E)$ with vertex set V and edge set E . In the site problem the vertices of the graph are the possible locations of a particle and an edge $[i, j] \in E$ is said to be occupied if both its vertices are occupied by a particle. Two particles belong to the same cluster if there is a chain of occupied edges connecting the vertices which they occupy. The subset $V' \subseteq V$ consisting of all the occupied vertices defines a section graph $G' = (V', E')$ where E' consists of all edges of E with both vertices in V' . If $A(V', G)$ is a function of the state of the system, its mean value is

$$\langle A; G \rangle = \sum_{V' \subseteq V} \pi(V') A(V', G) \quad (5-1)$$

where $\pi(V')$ is the probability that the vertices V' are occupied and $V - V'$ are unoccupied. The probability that vertex i is occupied is given by

$$p_i = \langle \nu_i; G \rangle \quad (5-2)$$

where

$$\nu_i(V') = \begin{cases} 1 & \text{for } i \in V' \\ 0 & \text{otherwise} \end{cases} \quad (5-3)$$

if the vertices are occupied independently.

In the case of the bond problem the edges which are in one of two states (open or closed for the water) define a partial graph $G' = (V', E')$ where E' is the open edges in E . In the similar way mean value is defined as

$$\langle A; G \rangle = \sum_{E' \in E} \pi(E') A(E', G), \quad (5-4)$$

where $\pi(E')$ is the probability that the edges E' are open and $E - E'$ are closed.

The probability that the edge $[i, j]$ is open is given by

$$p_{ij} = \langle v_{ij}; G \rangle, \quad (5-5)$$

where

$$v_{ij} = \begin{cases} 1 & \text{for } [i, j] \in E' \\ 0 & \text{otherwise} \end{cases} \quad (5-6)$$

The critical probability p_c is defined by

$$p_c = \sup_{P(p)=0} p \quad (5-7)$$

where $P(p)$ is the percolation probability which is defined in the following paragraph.

Now it is assumed that there is at least one infinitely long, self-avoiding walk beginning at every vertex. In the following part, we add the superscript B and S to the quantities of bond- and site problems respectively. And let $S_n(i)$ be the total number of n -stepped self-avoiding walks beginning from the vertex i . The percolation probability in the bond problem is defined by

$$P^{(B)}(i, p) = \lim_{n \rightarrow \infty} P_n^{(B)}(i, p) \quad (5-8)$$

where $P_n^{(B)}(i, p)$ is the probability that at least one of the $S_n(i)$ is open. Similar definition is also given in the case of the site problem.

Property 1. The critical probability in the site problem is larger than that in the bond problem, that is,

$$p_c^{(S)} \geq p_c^{(B)} \quad (5-9)$$

It can be shown that for any system and for all i

$$P_n^{(S)}(i,p) \leq P_n^{(B)}(i,p) \quad (5-10)$$

and hence

$$P^{(S)}(i,p) \leq P^{(B)}(i,p) \quad (5-11)$$

therefore the above results is obtained.

Property 2. A lower bound for $p_c^{(B)}$ is given by γ^{-1} , where

$$\log \gamma = \sup_i \lim_{n \rightarrow \infty} \sup n^{-1} \log S_n(i) \quad (5-12)$$

If $p(j,n)$ is the probability that exactly j of the walks $S_n(i)$ are open then

$$P_n(i,p) = \sum_{j \geq 1} p(j,n) \leq \sum j p(j,n) = S_n(i) p^n \quad (5-13)$$

$S_n(i) p^n$ is the expected number of open n stepped walks from i .

When $p = p_c$

$$\lim_{n \rightarrow \infty} S_n(i) p_c^n = 0 \quad (5-14)$$

thus the lower bound is given as follows

$$p_c^{(B)} \geq \gamma^{-1} \quad (5-15)$$

Property 3. If L is obtained from L^+ by removal of edges we have for bond and site problems

$$P_{n(L^+)}(i,p) \geq P_{n(L)}(i,p) \quad (5-16)$$

Thus

$$p_c(L^+) \leq p_c(L) \quad (5-17)$$

Fisher has used (5-17) to show that p_c does not decrease in going from the triangular, to the plane square, to the honeycomb lattice and also in going from the face-centered cubic to the body-centered cubic to the simple cubic and finally to the diamond lattice.

Property 4. Bond-to-site transformation

Any bond problem can be transformed into an equivalent site problem on a different graph called the covering graph.

The vertices of the covering graph G^c of a graph G are the middle points of edges of G and the edges of G^c consist of the lines connecting the pairs of edges which are adjacent in G . If G is a system L , and L^c is the covering system then

$$p_c^{(S)}(L^c) = p_c^{(B)}(L) \quad (5-18)$$

For example the covering lattice of the square lattice is given in Fig. 5-1

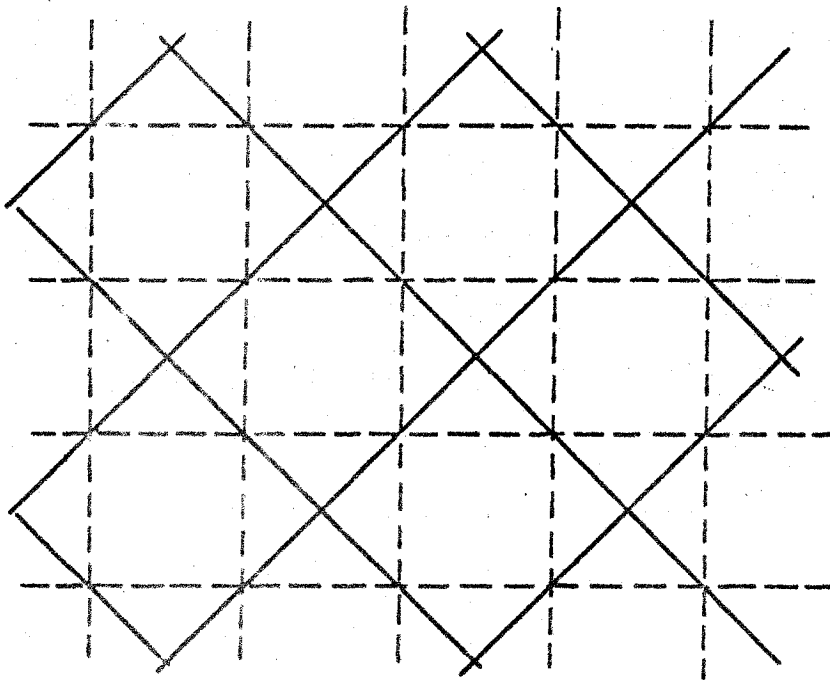


Fig. 5-1 An example of the site-bond transformation in the percolation problem, where the dashed line means the site percolation corresponding to the bond percolation problem (written by the solid line).

{5-3 Related problems

In this chapter we discuss some problems which are interesting in this field of critical phenomena and should be discussed in the future.

1. On the dependence of critical exponents in the dilute ferromagnet upon the concentration p . We have the exact analysis on the critical exponents in the annealed bond model for the dilute ferromagnet, but there is no such exact theory on the critical exponent in the quenched site model for the dilute ferromagnet. As an approach to this problem we can consider to calculate much longer series of high temperature expansion of susceptibility, and to apply Kadanoff-Weger and Suzuki's perturbational method to the dilute ferromagnet. The renormalization group theory may give us informations about critical exponents.

2. On the magnetic solid solutions which consist of ferromagnetic and antiferromagnetic substances. We discussed the ferromagnetic solid solutions in § 3-2. If we consider magnetic solid solutions which consist of ferromagnetic and antiferromagnetic substances, it might be interesting to calculate the critical exponents. For the critical exponent γ of susceptibility is $7/4$ in the two-dimensional ferromagnetic lattice and that is 0 (logarithmic singularity) in the two-dimensional antiferromagnetic lattice. Changes from $7/4$ to 0 in γ would be found in this system.

3. Relations between the magnetic solid solution and the percolation problem

The equivalence of the dilute ferromagnet and the percolation problem is discussed by many authors. In a similar way there may

exist some correspondences between the magnetic solid solution and the percolation problem. Adding an antiferromagnetic impurity to the magnetic solid solution has the possibility to affect spin arrangement in longer ranges as contrasted with local effects in the dilute ferromagnet.

4. Random magnetic system with different magnitudes of spin.
5. Magnetic solid solution with different types of spin-spin interaction.

For example we have $\text{Ni}_x\text{Co}_{1-x}\text{Cl}_2 \cdot 2\text{H}_2\text{O}$, where $\text{NiCl}_2 \cdot 2\text{H}_2\text{O}$ has the exchange interaction as the X-Y model and $\text{CoCl}_2 \cdot 2\text{H}_2\text{O}$ has that as the Ising model. In this case it is interesting to calculate the critical exponent as well as the critical temperature.

Acknowledgement

The author is indebted to Professor I. Syozi throughout accomplishing this work

Appendix A

$$b_1 = a_1(1) (a_1 p^2 + 2a_3 pq + a_2 q^2) ,$$

$$b_2 = a_2(1) \{ a_1^2 p^3 + (2a_1 a_3 + a_3^2) p^2 q + (a_1 \leftrightarrow a_2) pq^2 + a_2^2 q^3 \} ,$$

$$b_3 = a_3(1) \{ a_1^3 p^4 + 2(a_1^2 a_3 + a_1 a_3^2) + (2a_1 a_2 a_3 + 2a_3^3 + a_1 a_3^2 + a_2 a_3^2) p^2 q^2 + 2(a_1 \leftrightarrow a_2) pq^3 + a_2^3 q^4 \} - \frac{1}{3} a_1(1) (a_1^3 p^2 + 2a_3^3 pq + a_2^3 q^2) ,$$

$$b_4 = a_4(1) \{ a_1^4 p^5 + (2a_1^3 a_3 + 3a_1^2 a_3^2) p^4 q + (2a_1^2 a_2 a_3 + 4a_1 a_3^3 + a_1^2 a_3^2 + 2a_1 a_2 a_3^2 + a_3^4) p^3 q^2 + (a_1 \leftrightarrow a_2) p^2 q^3 + (a_1 \leftrightarrow a_2) pq^4 + a_2^4 q^5 \} - \frac{2}{3} a_2(1) \{ a_1^4 p^3 + (a_1^3 a_3 + a_1 a_3^3 + a_3^4) p^2 q + (a_1 \leftrightarrow a_2) pq^2 + a_2^4 q^3 \} ,$$

$$b_5 = a_5(1) \{ a_1^5 p^6 + 2(a_1^4 a_3 + 2a_1^3 a_3^2) p^5 q + (2a_1^3 a_2 a_3 + 7a_1^2 a_3^3 + 3a_1^2 a_2 a_3^2 + 3a_1 a_3^4) p^4 q^2 + 2(a_1^2 a_2^2 a_3 + a_1^2 a_2 a_3^2 + a_1 a_2^2 a_3^2 + 4a_1 a_2 a_3^3 + a_1 a_3^4 + a_2 a_3^4 + a_3^5) p^3 q^3 + (a_1 \leftrightarrow a_2) p^2 q^4 + (a_1 \leftrightarrow a_2) pq^5 + a_2^5 q^6 \} - a_5(1) \{ a_1^5 p^4 + 2(a_1^3 a_3^2) p^3 q + (a_1^2 a_2 a_3^2 + a_1 a_2^2 a_3^3 + 2a_1 a_2 a_3^3 + 2a_3^5) p^2 q^2 + 2(a_1 \leftrightarrow a_2) pq^3 + a_2^5 q^4 + \frac{2}{15} a_1(1) a_1^5 p^2 + 2a_3^5 pq + a_2^5 q^2 \} - \frac{1}{3} a_3(1) \{ 3a_1^5 p^4 + 2(a_1^2 a_3^2 + 2a_1^4 a_3 + 2a_3^4 + a_1^3 a_3^2) p^3 q + (2a_1^3 a_2 a_3 + 2a_1 a_2^3 a_3 + 2a_1 a_2 a_3^3 + 6a_3^5 + a_1^3 a_3^2 + a_2^3 a_3^2 + 2a_1 a_3^4 + 2a_2 a_3^4) p^2 q^2 + 2(a_1 \leftrightarrow a_2) pq^3 + 3a_2^5 q^4 \} ,$$

$$b_6 = a_6(1) \{ a_1^6 p^7 + (2a_1^5 a_3 + 5a_1^4 a_3^2) p^6 q + (2a_1^4 a_2 a_3 + 4a_1^3 a_2 a_3^2 +$$

$$\begin{aligned}
& 8a_1^3a_3^3 + 6a_1^2a_3^4 + a_1^4a_3^2)p^5q^2 + (2a_1^3a_2^2a_3 + 3a_1^2a_2^2a_3^2 + 12a_1^2a_2a_3^3 + \\
& 6a_1a_2^4a_3 + 6a_1a_3^5 + a_3^6 + 2a_1^3a_2a_3^2 + 3a_1^2a_3^4)p^4q^3 + (a_1 \leftrightarrow a_2)p^3q^4 \\
& + (a_1 \leftrightarrow a_2)p^2q^5 + (a_1 \leftrightarrow a_2)pq^6 + a_2^6q^7) - a_6^2(1) \{ a_1^6p^5 + (a_1^5a_3 + \\
& 2a_1^4a_3^2 + a_1^3a_3^3 + a_1^2a_3^4)p^4q + (2a_1^2a_2a_3^3 + 2a_1^3a_3^3 + a_1^2a_3^4 + a_1a_2^2a_3^3 \\
& a_3^6 + a_1a_2a_3^4 + a_1a_3^5 + a_1^3a_2a_3^2)p^3q^2 + (a_1 \leftrightarrow a_2)p^2q^3 + (a_1 \leftrightarrow a_2) \\
& xpq^4 + a_2^6q^5 \} - a_6^3(1) \{ a_1^6p^4 + (a_1^4a_3^2 + a_1^2a_3^4 + 2a_1^3a_3^3)p^3q + 2(a_1a_2^2a_3^3 \\
& + a_1^2a_2a_3^3 + a_3^6)p^2q^2 + (a_1 \leftrightarrow a_2)pq^3 + (a_2^6q^4) + a_2(1) \left[\frac{17}{45}a_1^6p^3 + \right. \\
& \left. \left\{ \frac{4}{15}(a_1^5a_3 + a_1a_3^5) + \frac{1}{9}a_1^3a_3^3 + \frac{17}{45}a_3^6 \right\} p^2q + \{ a_1 \leftrightarrow a_2 \} pq^2 + \frac{17}{45}a_2^6q^3 \right] - \\
& a_4(1) \left[\frac{4}{3}a_1^6p^5 + \left\{ \frac{2}{3}a_1^3a_3^3 + 2(a_1^5a_3 + a_1^4a_3^2 + a_1^2a_3^4) \right\} p^4q + \left\{ \frac{2}{3}(a_1^2a_2a_3^3 \right. \right. \\
& \left. \left. + a_1^2a_2^3a_3 + a_1a_2^3a_3^2 + a_1^3a_2a_3^2 + a_1^4a_3^2 + a_1^2a_3^4) + \frac{4}{3}(a_1^4a_2a_3 + a_1a_2^4a_3 \right. \right. \\
& \left. \left. + a_1^3a_3^3 + a_3^6) + 4a_1a_3^5 \right\} p^3q^2 + \{ a_1 \leftrightarrow a_2 \} p^2q^3 + \{ a_1 \leftrightarrow a_2 \} pq^4 + \frac{4}{3}a_2^6q^5 \right] \\
b_7 = & a_7^1(1) \left[a_1^7p^8 + \{ 2(a_1^6a_3 + 3a_1^5a_3^2) \} p^7q + \{ 2a_1^5a_2a_3 + 5(a_1^4a_2a_3^2 + 2a_1^4a_3^3 \right. \\
& \left. + 2a_1^3a_3^4) + a_1^5a_3 \} p^6q^2 + 2 \{ a_1^4a_2^2a_3 + a_1^4a_2a_3^2 + 2(a_1^3a_2^2a_3^2 + a_1^2a_2a_3^3 \right. \\
& \left. + a_1a_3^6 + a_1^3a_3^4) + 6(a_1^3a_2a_3^3 + a_1^2a_2a_3^4 + a_1^2a_3^5) \} p^5q^3 + \{ 2(a_1a_2^3a_3^3 \right. \\
& \left. a_3^7) + 3(a_1^2a_2^3a_3^2 + a_2a_3^6 + a_1^3a_2a_3^2) + 9(a_1a_2^2a_3^4 + a_1^2a_2a_3^4) + \right. \\
& \left. 18(a_1^2a_2^2a_3^3 + a_1a_2a_3^5) \} p^4q^4 + \{ a_1 \leftrightarrow a_2 \} p^3q^5 + \{ a_1 \leftrightarrow a_2 \} p^2q^6 + \{ a_1 \leftrightarrow a_2 \} \times \\
& \left. pq^7 + a_2^8q^8 \right] - a_7^2(1) \left[a_1^7p^6 + (a_1^6a_3 + 3a_1^5a_3^2 + a_1^3a_3^4 + a_1^4a_3^3)p^5q + \right. \\
& \left. \{ a_1^5a_2a_3 + a_1^2a_2^2a_3^3 + 3(a_1^2a_3^5 + a_1^3a_3^4) + a_1a_3^6 + 2(a_1^2a_2a_3^4 + a_1^4a_3^3) - \right. \\
& \left. a_1^3a_2a_3^3 + a_1^4a_2a_3^2 \} p^4q^2 + \{ 2(a_1^2a_2^2a_3^3 + a_1a_2a_3^5 + a_3^7) + 3(a_1a_2^3a_3^3 + \right. \\
& \left. a_1^2a_2a_3^4 + a_1a_2^2a_3^4 + a_1^3a_2a_3^3) + a_2a_3^6 + a_1a_3^6 \} p^3q^3 + \{ a_1 \leftrightarrow a_2 \} p^2q^4 + \right.
\end{aligned}$$

$$\begin{aligned}
& (a_1 \leftrightarrow a_2) p q^5 + a_2^7 q^6 \Big] - a_7^{2b}(1) \Big\{ a_1^7 p^6 + 2(a_1^5 a_3^2 + a_1^2 a_3^4 + a_1^4 a_3^2) p^5 q + \\
& \{ 4(a_1^3 a_2 a_3^3 + a_1^4 a_3^3) + a_1 a_2^2 a_3^4 + a_1^4 a_2 a_3^2 + 2(a_1^2 a_3^5 + a_1 a_3^6) + a_1^5 a_3^2 \} \times \\
& p^4 q^2 + 2 \{ a_1 a_2^3 a_3^3 + a_1^2 a_2^2 a_3^3 + a_1^3 a_2 a_3^3 + a_1^2 a_2 a_3^4 + a_1 a_2^2 a_3^4 + \\
& a_1^3 a_3^4 + a_2^3 a_3^4 + 2a_1 a_2 a_3^5 + a_3^7 \} p^3 q^3 + \{ a_1 \leftrightarrow a_2 \} p^2 q^4 + \{ a_1 \leftrightarrow a_2 \} p q^5 \\
& + a_2^7 q^6 \Big] - a_7^{2c}(1) \Big\{ a_1^7 p^6 + 2(a_1^4 a_3^3 + 2a_1^5 a_3^2) p^5 q + \{ 2(a_1^3 a_2 a_3^3 + \\
& a_1^4 a_2 a_3^2) + a_1^3 a_2^2 a_3^2 + a_1^4 a_2 a_3^2 + 6a_1^2 a_3^5 + 3a_1^3 a_2^4 \} p^4 q^2 + 2(a_1^2 a_2^2 a_3^3 + \\
& a_1^3 a_2^2 a_3^2 + a_1^2 a_3^3 a_3^2 + 4a_1 a_2 a_3^5 + a_1^2 a_2 a_3^4 + a_1 a_2^2 a_3^4 + a_3^7) p^3 q^3 + \\
& \{ a_1 \leftrightarrow a_2 \} p^2 q^4 + (a_1 \leftrightarrow a_2) p q^5 + a_2^7 q^6 \Big] - a_7^{2d}(1) \Big\{ a_1^7 p^6 + 2(a_1^4 a_3^3 \\
& + 2a_1^5 a_3^2) p^5 q + \{ 2a_1^4 a_2 a_3^2 + 4(a_1^3 a_2 a_3^3 + a_1 a_3^5 + a_1^3 a_3^4) + a_1^2 a_2 a_3^4 \} \times \\
& p^4 q^2 + 2 a_3^7 + 3a_1^2 a_2^2 a_3^3 + 2(a_1^2 a_2 a_3^4 + a_1 a_2 a_3^5 + a_1 a_2 a_3^4) p^3 q^3 + \\
& \{ a_1 - a_2 \} p^2 q^4 + (a_1 \leftrightarrow a_2) p q^5 + a_2^7 q^6 \Big] - a_7^3(1) \Big\{ a_1^7 p^5 + (a_1^6 a_3^4 + \\
& 2a_1^3 a_3^4 + a_1^4 a_3^3 + a_1^5 a_3^2) p^4 q + (2a_1^3 a_2 a_3^3 + a_1 a_2^2 a_3^4 + a_1^2 a_2^2 a_3^3 + \\
& a_1^2 a_3^5 + a_3^7 + a_1 a_3^6 + a_1^3 a_3^4 + a_1^2 a_2 a_3^4 + a_1^4 a_3^3) p^3 q^2 + (a_1 \leftrightarrow a_2) p^2 q^3 \\
& + (a_1 \leftrightarrow a_2) p q^4 + a_2^7 q^5 \Big\} - a_7^4(1) \Big\{ a_1^7 p^4 + 2(a_1^4 a_3^3 + a_1^3 a_3^4) p^3 q + \{ \\
& 2(a_1^2 a_2^2 a_3^3 + a_3^7) + a_1 a_2^2 a_3^4 + a_1^2 a_2 a_3^4 \} p^2 q^2 + (a_1 \leftrightarrow a_2) p q^3 + a_2^7 q^5 \Big\} \\
& - a_1(1)(a_1^7 p^2 + 2a_3^7 p q + a_2^7 q^2) - a_3(1) \Big\{ \frac{11}{15} a_1^7 p^4 + 2 \Big\{ \frac{17}{45} (a_1 a_3^6 + \\
& a_1^6 a_3) + \frac{2}{9} (a_1^3 a_3^4 + a_1^4 a_3^3) + \frac{2}{15} (a_1^5 a_3^2 + a_1^2 a_3^5) \Big\} p^3 q + \Big\{ \frac{4}{15} (a_1 a_2^5 a_3 \\
& + a_1^5 a_2 a_3 + a_1 a_2 a_3^5 + a_1 a_3^6 +
\end{aligned}$$





$$\begin{aligned}
& a_2 a_3^6) + \frac{2}{9}(a_1^3 a_2^3 a_3 + a_1 a_2^3 a_3^3 + a_1^3 a_2 a_3^3 + a_2^3 a_3^4 + a_1^3 a_3^4) + \frac{2}{15}(a_1^5 a_3^2 + \\
& a_2^5 a_3^2) + \frac{1}{9}(a_1 a_3^6 + a_2 a_3^6)\} p^2 q^2 + \{a_1 \leftrightarrow a_2\} p q^3 + \frac{11}{15} a_2^7 q^4 \} - a_5^1(1) \{ \\
& \frac{5}{3} a_1^7 p^6 + \{ \frac{8}{3}(a_1^6 a_3 + a_1^3 a_3^4) + \frac{2}{3} a_1^4 a_3^3 + 4 a_1^5 a_3^2 \} p^5 q + \{ \frac{2}{3}(a_1^3 a_2 a_3^3 + \\
& a_1^3 a_2^3 a_3 + a_1^3 a_3^4) + a_1^2 a_2^3 a_3^2 + a_1^3 a_3^4 + a_1^5 a_3^2 + 2(a_1^5 a_2 a_3 + a_1^2 a_2 a_3^4 + \\
& a_1^4 a_2 a_3^2) + 4(a_1^4 a_3^3 + a_1 a_3^6) \} p^4 q^2 + \{ \frac{2}{3}(a_1^3 a_2^2 a_3^2 + a_1^2 a_2^2 a_3^3 + a_1^2 a_2^3 a_3^2 + \\
& a_1^3 a_3^4 + a_2^3 a_3^4 + a_3^7) + \frac{4}{3}(a_1^2 a_2 a_3^4 + a_1 a_2^2 a_3^4 + a_1 a_2^4 a_3^2 + a_1^4 a_2 a_3^2 + \\
& a_1^2 a_2^4 a_3 + a_1^4 a_2 a_3^2) + \frac{8}{3}(a_1 a_2^3 a_3^3 + a_1^3 a_2 a_3^3 + a_1 a_3^6 + a_2 a_3^6 + a_3^7) + \\
& 8 a_1 a_2 a_3^5 \} p^3 q^3 + \{a_1 - a_2\} p^2 q^4 + \{a_1 - a_2\} p q^5 + \frac{5}{3} a_2^7 q^6 \} - a_5^2(1) \{ \\
& \frac{5}{3} a_1^7 p^4 + \{ \frac{4}{3}(a_1^3 a_3^4 + a_1^4 a_3^3) + 2(a_1^5 a_3^2 + a_1^2 a_3^5) \} p^3 q + \{ \frac{1}{3}(a_1^2 a_2^3 a_3^2 + \\
& a_1^3 a_2^2 a_3^2) + \frac{2}{3}(a_1 a_2^2 a_3^4 + a_1^2 a_2 a_3^4 + a_1 a_2^3 a_3^3 + a_1^3 a_2 a_3^3 + a_1 a_2^4 a_3^2 + \\
& a_1^4 a_2 a_3^2) + 2 a_1 a_2 a_3^5 + \frac{10}{3} a_3^7 \} p^2 q^2 + \{a_1 \leftrightarrow a_2\} p q^3 + \frac{5}{3} a_2^7 q^4 \} ,
\end{aligned}$$

where a_7^{2a} , a_7^{2b} , a_7^{2c} and a_7^{2d} are the partial coefficients of a_7^2

which are given in Table 1, corresponding to the four magnetic graphes. They all consist of the six vertexes, and given in Table 3.

The coefficients $(a_1 \leftrightarrow a_2)$ of $p^m q^n$ are obtained by interchanging a_1 and a_2 in the coefficients of $p^n q^m$.

Table 3. The partial coefficients of a_7^2 are given.

| a_r | $n = 1$ | $n = 2$ | $n = 3$ | $n = 4$ | $n = 5$ | $n = 6$ | $n = \infty$ |
|--|---------|---------|------------------|---------|---------|------------------|--------------|
| a_7^{2a}  | 96 | 384 | $565\frac{1}{3}$ | 664 | 723.2 | $762\frac{2}{3}$ | 960 |
| a_7^{2b}  | 32 | 144 | $221\frac{1}{3}$ | 262 | 286.4 | $302\frac{2}{3}$ | 384 |
| a_7^{2c}  | 24 | 132 | 176 | 198 | 211.2 | 220 | 264 |
| a_7^{2d}  | 8 | 36 | 48 | 54 | 57.6 | 60 | 72 |

Appendix B

$$b'_1 = a_1(1) \bar{a}$$

$$b'_2 = a_2(1) \bar{a}^2$$

$$b'_3 = a_3(1) \bar{a}^3 - \frac{1}{3} a_1(1) \bar{a}^3$$

$$b'_4 = a_4(1) \bar{a}^4 - \frac{2}{3} a_2(1) \bar{a} \bar{a}^3$$

$$b'_5 = a_5^1(1) \bar{a}^5 - a_5^2(1) \bar{a}^3 \bar{a}^2 + \frac{2}{15} a_1(1) \bar{a}^5 - a_3(1) \bar{a}^2 \bar{a}^3$$

$$b'_6 = a_6^1(1) \bar{a}^6 - a_6^2(1) \bar{a}^4 \bar{a}^2 - a_6^3(1) \bar{a}^2 \bar{a}^2^2 + \frac{1}{9} a_2(1) \bar{a}^3^2 + \frac{4}{15} a_2(1) \bar{a} \bar{a}^5 - \frac{4}{3} a_4(1) \bar{a}^3 \bar{a}^3$$

$$b'_7 = \{a_7^1(1) - a_7^{2d}(1)\} \bar{a}^7 - \{a_7^{2a}(1) + a_7^{2b}(1) + a_7^{2c}(1)\} \bar{a}^5 \bar{a}^2 - a_7^3(1) \bar{a}^3 \bar{a}^2^2 - a_7^4(1) \bar{a} \bar{a}^2^3 - \frac{17}{315} a_1(1) \bar{a}^7 + \frac{1}{3} a_3(1) \bar{a} \bar{a}^3^2 + \frac{2}{5} a_3(1) \bar{a}^2 \bar{a}^5 - \frac{5}{3} a_5^1(1) \bar{a}^4 \bar{a}^3 + a_5^2(1) \bar{a}^2 \bar{a}^2 \bar{a}^3 + \frac{2}{3} a_5^2(1) \bar{a}^3 \bar{a}^4 ,$$

where

$$\bar{a} = a_1 p + a_2 q$$

$$\bar{a}^2 = a_1^2 p + a_2^2 q$$

$$\bar{a}^3 = a_1^3 p + a_2^3 q$$

$$\bar{a}^4 = a_1^4 p + a_2^4 q$$

$$\bar{a}^5 = a_1^5 p + a_2^5 q$$

and

$$\bar{a}^7 = a_1^7 p + a_2^7 q .$$

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