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A NEW CLUSTER APPROXIMATION TO THE ISING
AND HEISENBERG SPIN SYSTEMS

— DILUTION EFFECT AND ROLE OF THE SECOND-
NEIGHBOR INTERACTION —

by Hiroshi MANO

February 1976

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Abstract

In Part I a new cluster approximation is proposed. The present approximation bases upon a cluster expansion of the expression of the magnetization and an approximate reduction of the density matrix. The density matrix of a cluster is obtained by introducing stochastic variables and the fluctuation of the effective field is taken into account. The approximation can be improved systematically by expanding the size of clusters to be considered and it gives satisfactory results for the whole range of the concentration of magnetic atoms.

The 1-, 2- and 3- site approximations are applied to the dilute Ising and Heisenberg spin systems with the nearest-neighbor interaction. The critical concentration, Curie temperature, susceptibility and spontaneous magnetization are calculated. The 2- site and 3- site approximations give the results reflecting more details of the geometry of the lattice than the coordination number. With decreasing concentration of magnetic atoms the Curie temperature for the Heisenberg model decreases more rapidly than that for the Ising model. The value of the critical concentration for the Heisenberg model is, however, quite close to that for the Ising model. The value for the three-dimensional lattices is in agreement with the value obtained in the site percolation problem.

In Part II the Ising spin system with the first- and second-neighbor interactions is investigated in the 1- site

and 2- site approximations. For the pure ferromagnetic systems the Curie temperature is calculated. The dependence of the Curie temperature on the ratio of the interactions ($R = J_2/J_1$) agrees well with the series expansion results of Dalton and Wood. For the square lattice the Curie temperature decreases with decreasing R to vanish at the boundary where the ferromagnetic ground state disappears. When R is negative, the specific heat has a peak below T_c in addition to the peak at $T = T_c$.

In the diluted case the critical concentration, Curie temperature, susceptibility, spontaneous magnetization, pair correlation function and specific heat are calculated. The critical concentration depends only on the range of the interactions when R is positive. The interesting phenomena are found when the second-neighbor interaction is small and ferromagnetic. The Curie temperature has a trail near the critical concentration. In the low temperature region near $k_B T \sim J_2$ the spontaneous magnetization shows apparent decrease and the specific heat has a peak corresponding to the rapid decrease of the pair-correlation of the second-neighboring spins.

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Part I

A New Cluster Approximation to the Ising and Heisenberg
Magnets

§ 1. Introduction

The problem of the dilute magnet, i.e. a mixture of magnetic and non-magnetic atoms on a given lattice, has received considerable attention in magnetism. Such a system is usually treated in two ideal limits; one is the quenched limit in which atoms are frozen randomly in their positions, and the other is the annealed limit in which the distribution of atoms is determined as to minimize the free energy of the system. The distinction between these two limits was discussed by Brout.¹⁾ In this paper we will consider the ferromagnetic Ising and Heisenberg models of $S = 1/2$ in the quenched limit.

As the concentration of the magnetic atoms p decreases, the Curie temperature lowers and finally vanishes at the critical concentration p_c . It is almost self-evident that a system does not show ferromagnetic behaviors for p less than the critical concentration p_s for the site percolation problem,²⁾ since there is no infinite network of adjacent magnetic atoms. It is, however, not evident whether a system starts to exhibit ferromagnetic behaviors or not as soon as p exceeds p_s . Various approximate methods have been applied to the problem and the value of p_c and the dependence of the Curie temperature on p have been calculated. Many authors^{3)~6)} have concluded on the basis of series expansion in powers of p that p_c for the Ising model is equal to p_s . On the other hand, whether p_c for the Heisenberg model coincides with p_c for the Ising model or not

seems to be an unsettled problem. The results obtained by the series expansion in powers of $p^{3/6}$ and in inverse powers of the temperature^{7),8)} show the concentration dependence of the Curie temperature for the Heisenberg model is strikingly different from that for the Ising model. Elliott, Heap, Morgan and Rushbrooke⁹⁾ argued that p_c was a geometrical property of the lattice, being independent of the models. Some doubt, however, has been cast on this argument. For example, Morgan and Rushbrooke⁴⁾ pointed out that in the proof given by Elliott et. al. a uniform convergence of the series expansion of the susceptibility in powers of p was tacitly assumed near $T=0$. Also Murray¹⁰⁾ obtained a significantly higher value of p_c than the corresponding value for the Ising model, applying the spin wave theory to the dilute Heisenberg model.

The problem has been discussed also by use of the cluster approximations of various types by Sato, Arrott and Kikuchi,¹¹⁾ Mamada and Takano¹²⁾ and Matsudaira¹³⁾ for the Ising model and by Oguchi and Obokata¹⁴⁾ for the Ising and Heisenberg models. The results obtained by these methods, except Matsudaira's¹³⁾ method, depend on the coordination number z only, independent of the further details of the geometry of the lattice, and the values of p_c for the Ising model are considerably small compared with the percolation probabilities. We think that these results should be meaningful in the case of the Bethe lattice.

It is the purpose of this paper to present a new cluster approximation which reflects the geometry of the lattice in

more detail and is applicable to the Heisenberg model as well as the Ising model; moreover we can improve the approximation systematically by expanding the size of the cluster from 1 site to 2, 3, ... sites. The idea of the effective field has been used in the various small cluster type approximations, for example, in the molecular field, Bethe or constant-coupling approximation. In these methods, the effective field is always assumed to be a constant. As was discussed by Mamada and Takano¹²⁾, it is necessary to take into account the fluctuation of the effective field. Mamada and Takano considered a single spin cluster and gave the probability distribution of the effective field acting on the spin as the direct products of one-spin density matrices. They obtained satisfactory results in spite of the fact that the approximation is a single site one, i.e. the size of the cluster was the same as in the molecular field approximation. We introduce pseudo-spins which are stochastic variables taking the values $1/2$ and $-1/2$ with self-consistently determined probabilities to represent the fluctuation of the effective field. When we reduce the density matrix of the total system to that of the cluster of a given size, we replace spins not included in the cluster by the pseudo-spins. The probability distribution of the pseudo-spin is determined by the condition that its average value is equal to the average value of the magnetization per spin (divided by $g\mu_B$). In order to derive the self-consistent equation, we expand the expression of the magnetization in cluster series and terminate it at the n -th term which consists

of n and less than n site cluster contributions; n , which is taken to be 1, 2, 3 \dots , defines the degree of the approximation. Our 1-site approximation coincides with the Mamada-Takano approximation. There is, however, no obvious way of extending their approximation to many site clusters, whereas the present discussion will yield a systematic way of deriving many site approximation. When $p=1$, our cluster expansion resembles Strieb, Callen and Horwitz's¹⁵⁾ cluster expansion.

We give the formalism in §2. In §3 we present the results of the calculation for the Curie temperature, the critical concentration, the spontaneous magnetization and the susceptibility for the Ising and Heisenberg models together with discussions. Calculations are carried out up to the 3-site approximation for the typical lattices; linear chain, square (sq.), planer triangular (p.t.), simple cubic (s.c.), body centered cubic (b.c.c.) and face centered cubic (f.c.c.) lattices.

§ 2 Formalism

In the present paper we consider the Heisenberg and Ising models of $S=1/2$ in which only the nearest neighbor ferromagnetic interaction $J > 0$ exists, although there is no limitation for the range of the interaction in applying our method. We denote the spacial distribution of atoms in the quenched dilute system composed of magnetic and non-magnetic atoms by a set of the occupation numbers $\{\alpha\} = \{\alpha_1, \alpha_2, \dots, \alpha_N\}$, where

the occupation number α_i is 1 if a magnetic atom occupies the i -th site and 0 if a non-magnetic atom occupies the site; N is the total number of the sites in the system. We assume the perfect randomness when we take average over the configuration of atoms.

The Hamiltonian of the system for a spacial configuration denoted by $\{\alpha\}$ is given by

$$H^{\{\alpha\}} = -2J \sum_{(i,j)} \alpha_i \alpha_j \vec{S}_i \cdot \vec{S}_j - H \sum_i \alpha_i S_i^z \quad (1)$$

$$H^{\{\alpha\}} = -2J \sum_{(i,j)} \alpha_i \alpha_j S_i^z S_j^z - H \sum_i \alpha_i S_i^z \quad (2)$$

in the Heisenberg and Ising models, respectively. In both cases the first summation is taken over all pairs of nearest neighboring sites and the second over all sites. H is the external magnetic field which is assumed to be in the negative z -direction and measured in the unit of $g\mu_B = 1$. Though we describe our formalism mainly in terms of the Heisenberg spins in the following, the equations derived in this section can easily be adapted to the Ising model.

(i) Approximate reduction of density matrix

In principle all the properties of the system can be calculated by use of the density matrix

$$\rho^{\{\alpha\}} = \exp[-\beta H^{\{\alpha\}}] / \text{Tr} \exp[-\beta H^{\{\alpha\}}] , \quad \beta = 1/k_B T \quad (3)$$

which, for example, determines the magnetization (divided by $g\mu_B$) to be

$$M^{\{\alpha\}} = \text{Tr}[(\sum_i \alpha_i S_i^Z) \cdot \rho^{\{\alpha\}}] \quad (4)$$

Expanding the r.h.s. of Eq.(4) in a cluster series, we develop a systematic self-consistent approximation in the following subsections. To that purpose we discuss here our method of an approximate reduction of the density matrix given by Eq.(3) to the density matrix of a cluster of sites. In the conventional small cluster approximation so far proposed one obtains the density matrix for a given cluster by replacing in the Hamiltonian those spin operators which do not belong to the cluster by constant vectors such that

$$S_i^X = S_i^Y = 0 \text{ and } S_i^Z = \bar{S} \quad (5)$$

The constant \bar{S} plays the role of an effective field acting on the spins in a given cluster and is determined in a self-consistent way. As was discussed by Mamada and Takano,¹²⁾ however, it is more reasonable to assume that the effective field will take various values instead of being a constant even in the thermal equilibrium. Thus we propose the following approximation in the present paper. We replace the spin operator S_i^Z not belonging to a given cluster by a stochastic variable \bar{S}_i (we call it 'pseudo-spin') which takes values 1/2 and -1/2 with self-consistently determined probabilities. In this approximation the fluctuation of the spin operators S_i^X and S_i^Y are not considered, so that the approximation is better for the Ising model than for the Heisenberg model.

We define the approximate density matrix $\rho_{\{l\}}^{\{\alpha\}}$ of a cluster

of ℓ sites under consideration in the following way. Replacing the spins not belonging to the cluster by the pseudo-spins in the Hamiltonian, we obtain the effective Hamiltonian $\mathcal{K}_{\{\ell\}}^{\{\alpha\}}$ in which the spins $\{\vec{S}_i\}_{\ell} = \{\vec{S}_1, \dots, \vec{S}_m\}$ existing in the cluster appear as operators, where m is less than or equal to ℓ . Defining $\text{Tr}\{\ell\}$ as the trace over the spin operators $\{\vec{S}_i\}_{\ell}$, we express $\rho_{\{\ell\}}^{\{\alpha\}}$ as,

$$\rho_{\{\ell\}}^{\{\alpha\}}(\{\vec{S}_i\}_{\ell}) = \langle \rho_{\{\ell\}}^{\{\alpha\}}(\{\vec{S}_i\}_{\ell}) \rangle_{\bar{S}} \quad (6)$$

where

$$\rho_{\{\ell\}, \bar{S}}^{\{\alpha\}}(\{\vec{S}_i\}_{\ell}) = \exp[-\beta \mathcal{K}_{\{\ell\}}^{\{\alpha\}}] / \text{Tr}_{\{\ell\}} \exp[-\beta \mathcal{K}_{\{\ell\}}^{\{\alpha\}}] \quad (7)$$

and where $\langle \dots \rangle_{\bar{S}}$ is the average over the stochastic variables \bar{S}_i in the effective Hamiltonian $\mathcal{K}_{\{\ell\}}^{\{\alpha\}}$. Noting that the variables \bar{S}_i which are not the nearest neighbors of the cluster drop out in Eq.(7), $\langle \dots \rangle_{\bar{S}}$ is written as

$$\langle \dots \rangle_{\bar{S}} = \int \dots P(\{\bar{S}_i\}_{nn}) d\{\bar{S}_i\}_{nn} \quad (8)$$

where $\{\bar{S}_i\}_{nn}$ represents the set of the pseudo-spins in the n.n. sites of the cluster and $P(\{\bar{S}_i\}_{nn})$ is the probability for a given set of values of $\{\bar{S}_i\}_{nn}$.

In the present approximation we treat \bar{S}_i 's as variables independent of one another to obtain

$$P(\{\bar{S}_i\}_{nn}) = \prod_{i \in nn} P_1(\bar{S}_i) \quad (9)$$

with the probability distribution function of a single pseudo-spin $P_1(\bar{S}_i)$. Then the average value of a pseudo-spin should be

equal to the average of the z-component of a spin operator $\langle S^Z \rangle_c$, where $\langle \dots \rangle_c$ means the average over atomic configurations $\{\alpha\}$ as well as the thermal average. With this assumption and the fact that the pseudo-spin takes the discrete values $1/2$ and $-1/2$ we determine the probability distribution $P_1(\bar{S}_1)$ as

$$P_1(\bar{S}_1) = \delta(\bar{S}_1 - \frac{1}{2}) \cdot (1 + X)/2 + \delta(\bar{S}_1 + \frac{1}{2}) \cdot (1 - X)/2 \quad (10)$$

where $X = 2\langle S^Z \rangle_c$.

(ii) Cluster expansion of the magnetization

In order to derive a self-consistent equation for $\langle S^Z \rangle_c$ in the next subsection, we expand $M^{\{\alpha\}}$ defined by Eq.(4) in a series of cluster averages by use of the approximate density matrix $\rho_{\{\ell\}}^{\{\alpha\}}$ defined above. We can prove that

$$M^{\{\alpha\}} = \sum_{\ell=1}^N M_{\ell}^{\{\alpha\}} \quad (11)$$

with

$$M_{\ell}^{\{\alpha\}} = \sum_{\{\ell\}} \sum_{k=1}^{\ell} \sum_{\{k\}} (-1)^{\ell-k} \text{Tr}_{\{k\}} [(\sum_{i \in \{k\}} \alpha_i S_i^Z) \cdot \rho_{\{k\}}^{\{\alpha\}}] \quad (12)$$

where the first sum over $\{\ell\}$ is taken over all possible clusters of ℓ sites and the third sum is taken over all subclusters of k sites belonging to a given $\{\ell\}$ with $\rho_{\{k\}}^{\{\alpha\}}$ representing the approximate density matrix for a subcluster $\{k\}$. Eq.(11) is exact as far as the sum is taken up to $\ell = N$. We will give a brief proof in the Appendix A.

For $M_{\ell}^{\{\alpha\}}$ defined by Eq.(12), the following theorems are easily proved.

(a) If a cluster $\{\ell\}$ is unlinked, i.e. composed of at least two subclusters $\{\ell_1\}$ and $\{\ell_2\}$ which are not connected by the interaction to each other, it makes no contribution to $M_{\ell}^{\{\alpha\}}$.

(b) If α_i of any one ^{of the site} belonging to $\{\ell\}$ is equal to zero, i.e. the site is not occupied by a spin, the cluster $\{\ell\}$ makes no contribution to $M_{\ell}^{\{\alpha\}}$.

The theorem (a) can be proved by noting that the traces on spin operators in the sites belonging to subclusters $\{\ell_1\}$ and $\{\ell_2\}$ can be taken separately in our approximate density matrices appearing in the r.h.s. of Eq.(12), and the theorem (b) by noting that if a subcluster $\{k\}$ appearing in Eq.(12) contains the i -th site, then $\rho_{\{k\}}^{\{\alpha\}}$ is equal to $\rho_{\{k-1\}}^{\{\alpha\}}$. Details of the proofs will be given in the Appendix B.

By use of these theorems we conclude that only linked clusters of ℓ spins are to be taken into account in the sum over $\{\ell\}$ in $M_{\ell}^{\{\alpha\}}$. Thus $M_{\ell}^{\{\alpha\}}$ is of order p^{ℓ} in powers of the concentration of magnetic atoms p , resulting in a rapid convergence in the cluster expansion when p is small. As will be discussed in §3, the convergence of the expansion is also good for p near 1. When $\bar{p}=1$, i.e. all α_i 's are equal to 1, our expansion coincides formally with the expression for the magnetization given by Strieb, Callen and Horwitz,¹⁵⁾ though their definition of density matrices for clusters is different.

(iii) Self-consistent approximation

With a given concentration of magnetic atoms p , we carry out the configurational average under the assumption that α_i 's are independent of each other, in other words, we assume the perfect randomness of atomic distribution. Then the probabilities of $\alpha = 1$ and $\alpha = 0$ are given by p and $q = 1-p$, respectively. The configurational average of the total spins, $M^c \equiv \langle M^{\{\alpha\}} \rangle_c$ is related to the average value of each spin by

$$\langle S^z \rangle_c = M^c / pN = X/2 \quad (13)$$

On the other hand, we can calculate $\langle M^{\{\alpha\}} \rangle_c$ approximately with a given value of X if we terminate the cluster expansion given by Eq.(11) at a certain value of $\ell = \ell_c$. Thus Eq.(13) can be regarded as the self-consistent equation to determine X . We shall show in the following that this procedure yields successively improved approximations if we increase ℓ_c .

(iii-1) One-site approximation

Keeping only the first term $M_1^{\{\alpha\}}$ in the r.h.s. of Eq.(11) and taking the configurational average, we get the lowest order approximation as

$$\begin{aligned} X/2 = \langle S^z \rangle_c &= \langle M_1^{\{\alpha\}} \rangle / pN \\ &= \langle \text{Tr}_1 [S_1^z \rho_1^{\{\alpha\}}(S_1^z)] \rangle_{c_i} \\ &= \langle \langle \text{Tr}_1 [S_1^z \rho_{1,s}^{\{\alpha\}}(S_1^z)] \rangle_s \rangle_{c_i} \end{aligned} \quad (14)$$

where $\langle \dots \rangle_{c_1}$ represents the configurational average on condition that $\alpha_1 = 1$. The density matrix which appears in Eq.(14) has the form

$$\rho_{1, \bar{s}}^{\{\alpha\}}(S_1^Z) = \exp[-\beta \mathcal{H}_1^{\{\alpha\}}] / \text{Tr}_1 \exp[-\beta \mathcal{H}_1^{\{\alpha\}}] \quad (15)$$

with

$$\mathcal{H}_1^{\{\alpha\}} = - (2J \sum_{\nu} \alpha_{\nu} \bar{S}_{\nu} + H) S_1^Z \quad (16)$$

where the sum over ν is taken over all the n.n. sites of the site 1. (cf. Fig.1(a-1) and (b-1)). After a short calculation we get the equation

$$\text{Tr}_1 [S_1^Z \rho_{1, \bar{s}}^{\{\alpha\}}(S_1^Z)] = \frac{1}{2} \tanh \left[\frac{\beta}{2} (2J \sum_{\nu} \alpha_{\nu} \bar{S}_{\nu} + H) \right] \quad (17)$$

We derive now explicitly the self-consistent equation to determine X . Suppose we have n magnetic atoms at the z nearest neighbors of the site 1. The probability of such a configuration is given by

$$Q_n^z = \binom{z}{n} p^n q^{z-n} \quad (18)$$

Since we neglect the correlation among the pseudo-spins \bar{S}_{ν} , the probability of the pseudo-spin configurations where k out of n pseudo-spins take the value $-1/2$ and the rest have the value $1/2$ is

$$D_k^n = \binom{n}{k} \left(\frac{1+X}{2} \right)^{n-k} \left(\frac{1-X}{2} \right)^k \quad (19)$$

For this pseudo-spin configuration $\sum_{\nu} \alpha_{\nu} \bar{S}_{\nu}$ takes the value

$$\sum_{\nu} \alpha_{\nu} \bar{S}_{\nu} = -\frac{1}{2} \cdot k + \frac{1}{2} \cdot (n-k) = (n-2k)/2 \quad (20)$$

The average over the atomic and pseudo-spin configurations, $\langle \langle \dots \rangle_{\bar{S}} \rangle_{c_1}$, in Eq.(14) can be written by use of Eqs.(18),(19) and (20). together with Eq.(17) as

$$X = \sum_{n=0}^z Q_n^z \sum_{k=0}^n D_k^n \tanh \left[\frac{\beta}{2} \{ J(n-2k) + H \} \right] \quad (21)$$

Note that D_k^n depends on X. In the present 1-site approximation there is no difference between the Heisenberg and Ising models. This equation is identical with that given by Mamada and Takano. Thus we conclude that our 1-site approximation corresponds to the approximation proposed by them.

(iii-2) Two-site approximation

Terminating the expansion given by Eq.(11) at $M_2^{\{\alpha\}}$, we obtain the self-consistent equation in the 2-site approximation. There are $\frac{1}{2} p^2 zN$ n.n. pairs of magnetic atoms, so that the equation is

$$\begin{aligned} X/2 &= \langle M_1^{\{\alpha\}} + M_2^{\{\alpha\}} \rangle_c / pN \\ &= \langle \text{Tr}_1 [S_1^z \cdot \rho_1^{\{\alpha\}}(S_1^z)] \rangle_{c_1} \\ &\quad + \frac{pz}{2} \cdot \langle \text{Tr}_{12} [(S_1^z + S_2^z) \cdot \rho_{12}^{\{\alpha\}}(\vec{S}_1, \vec{S}_2) - \text{Tr}_1 [S_1^z \cdot \rho_1^{\{\alpha\}}(S_1^z)] \\ &\quad \quad \quad - \text{Tr}_2 [S_2^z \cdot \rho_2^{\{\alpha\}}(S_2^z)] \rangle_{c_{12}} \quad (22) \end{aligned}$$

where the sites 1 and 2 are a pair of the nearest neighbors and $\langle \dots \rangle_{c_{12}}$ represents the configurational average on condition that $\alpha_1 = \alpha_2 = 1$; $\rho_{12}^{\{\alpha\}}$ is the approximate density matrix of the

cluster consisting of the sites 1 and 2. The terms containing the 1-site density matrix can be calculated in the same way as in the subsection (iii-1), giving a little care to the configurational average $\langle \dots \rangle_{c_{12}}$. The term containing the 2-site density matrix can be expressed as

$$\langle \text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12}^{\{\alpha\}}] \rangle_{c_{12}} = \langle \langle \text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12, \bar{s}}^{\{\alpha\}}] \rangle_{\bar{s}} \rangle_{c_{12}} \quad (23)$$

It is convenient for the calculation to use the equation

$$\text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12, \bar{s}}^{\{\alpha\}}] = \frac{\partial}{\partial (\beta H)} [\ln \{ \text{Tr}_{12} \exp(-\beta \mathcal{H}_{12}^{\{\alpha\}}) \}] \quad (24)$$

with

$$\mathcal{H}_{12}^{\{\alpha\}} = -2J \cdot \{ \vec{S}_1 \cdot \vec{S}_2 + \sum'_{v(1)} \alpha_{v(1)} \bar{S}_{v(1)} \cdot S_1^Z + \sum'_{v(2)} \alpha_{v(2)} \bar{S}_{v(2)} \cdot S_2^Z \} - H \cdot (S_1^Z + S_2^Z) \quad (25)$$

where the sums over $v(1)$ and $v(2)$ are taken over the nearest neighbours of the sites 1 and 2, respectively, the sites 1 and 2 being excluded. (cf. Fig. 1(a-2) and (b-2)) the eigenvalues of the effective Hamiltonian $\mathcal{H}_{12}^{\{\alpha\}}$ are

$$\begin{aligned} \lambda_1 &= -\frac{J}{2} (1 + h_1 + h_2) - H \\ \lambda_2 &= -\frac{J}{2} (1 - h_1 - h_2) + H \\ \lambda_{3,4} &= -\frac{J}{2} \left(-1 \pm \sqrt{(h_1 - h_2)^2 + 4} \right) \end{aligned} \quad (26)$$

$$\text{where } h_1 = 2 \sum'_{v(1)} \alpha_{v(1)} \bar{S}_{v(1)}, \quad h_2 = 2 \sum'_{v(2)} \alpha_{v(2)} \bar{S}_{v(2)} \quad (27)$$

using these eigen values, we obtain

$$\text{Tr}_{12}[(S_1^z + S_2^z) \rho_{12, \mathcal{S}}^{\{\alpha\}}] = \frac{\{\exp(-\beta\lambda_1) - \exp(-\beta\lambda_2)\}}{\sum_{i=1}^4 \exp(-\beta\lambda_i)}$$

$$\equiv m_2[h_1, h_2; H] \quad (28)$$

In the case of the close packed lattices (for example the triangular and fcc lattices), the site 1 and 2 have common nearest neighbors ; z_c , the number of these sites, is 2 for the triangular lattice and 4 for the fcc lattice, whereas for the loose packed lattices (for example the square, sc and bcc lattices) z_c is equal to zero. This fact makes the average over the atomic and pseudo-spin configurations somewhat complicated, since we have to distinguish the nearest neighbors shared by the sites 1 and 2 from the nearest neighbors not shared by them. After some manipulation we get the self-consistent equation as

$$X = \sum_{n=0}^z Q_n^z \sum_{k=0}^n D_k^n \tanh\left[\frac{\beta}{2} \{J(n-2k) + H\}\right]$$

$$+ pz \cdot \left(\sum_{n_1=0}^{z-z_c-1} Q_{n_1}^{z-z_c-1} \sum_{n_2=0}^{z-z_c-1} Q_{n_2}^{z-z_c-1} \sum_{n_{12}=0}^{z_c} Q_{n_{12}}^{z_c} \right.$$

$$\times \sum_{k_1=0}^{n_1} D_{k_1}^{n_1} \sum_{k_2=0}^{n_2} D_{k_2}^{n_2} \sum_{k_{12}=0}^{n_{12}} D_{k_{12}}^{n_{12}}$$

$$\times m_2[h_1, h_2; H] \left. - \sum_{n=0}^{z-1} Q_n^{z-1} \sum_{k=0}^{n+1} D_k^{n+1} \tanh\left[\frac{\beta}{2} \{J(n+1-2k) + H\}\right] \right) \quad (29)$$

where n_1+n_{12} and n_2+n_{12} are equal to the numbers of magnetic atoms which are at the nearest neighbors of the site 1 and 2, respectively; and n_{12} is the number of magnetic atoms which are at the nearest neighbors shared by the site 1 and 2. For the Ising model, replacing the term $\vec{S}_1 \cdot \vec{S}_2$ in Eq.(25) by $S_1^Z S_2^Z$, we can obtain a similar equation to Eq.(29). We note that the geometry of the lattice is reflected in the self-consistent equation through the number z_c as well as z , whereas in the 2-spin cluster approximation so far proposed only the number z enters in the equation. Thus we obtain different results in the present approximation for the sc and triangular lattices which have the same z ($=6$).

(iii-3) Three-site approximation

The next approximation is obtained by including the term $M_3^{\{\alpha\}}$ which corresponds to the contribution of the 3-site clusters. There are several types of distinct clusters consisting of three linked sites; they are the triangle, V-linked and straight-line clusters. The triangle clusters (as shown in Fig.1(b-5)) appear in the case of the closed packed lattices and their contributions to $M_3^{\{\alpha\}}$ is

$$\begin{aligned} & \sum_{(ijk)} \{ \text{Tr}_{ijk} [(\alpha_i S_i^Z + \alpha_j S_j^Z + \alpha_k S_k^Z) \rho_{ijk}^{\{\alpha\}}] - \text{Tr}_{ij} [(\alpha_i S_i^Z + \alpha_j S_j^Z) \rho_{ij}^{\{\alpha\}}] \\ & - \text{Tr}_{jk} [(\alpha_j S_j^Z + \alpha_k S_k^Z) \rho_{jk}^{\{\alpha\}}] - \text{Tr}_{ki} [(\alpha_k S_k^Z + \alpha_i S_i^Z) \rho_{ki}^{\{\alpha\}}] \\ & + \text{Tr}_i [\alpha_i S_i^Z \rho_i^{\{\alpha\}}] + \text{Tr}_j [\alpha_j S_j^Z \rho_j^{\{\alpha\}}] + \text{Tr}_k [\alpha_k S_k^Z \rho_k^{\{\alpha\}}] \} \end{aligned}$$

(30)

where the sites i, j and k are the nearest neighbors of one another, and the sum is taken over all the triangle clusters; $\rho_{ijk}^{\{\alpha\}}$ is the approximate density matrix of the cluster consisting of the sites i, j and k . The contribution of the V-linked clusters (as shown in Fig.1(a-4) and (b-4)) and the straight-line clusters (as shown in Fig.1(a-3) and (b-3)) to $M_3^{\{\alpha\}}$ is

$$\sum_{(i,j,k)} \text{Tr}_{ijk} [(\alpha_i S_i^Z + \alpha_j S_j^Z + \alpha_k S_k^Z) \rho_{ijk}^{\{\alpha\}}] - \text{Tr}_{ij} [(\alpha_i S_i^Z + \alpha_j S_j^Z) \rho_{ij}^{\{\alpha\}}] - \text{Tr}_{jk} [(\alpha_j S_j^Z + \alpha_k S_k^Z) \rho_{jk}^{\{\alpha\}}] + \text{Tr}_j [\alpha_j S_j^Z \rho_j^{\{\alpha\}}] \quad (31)$$

where the sites i and k are the nearest neighbors of the site j but not of each other, and the sum is taken over all the V-linked and straight-line clusters. In the case of the bcc and fcc lattices there are two types of the V-linked clusters which have the different angles between two links. Since the method of obtaining the self-consistent equation is the same as in the cases of the 1- and 2- site approximations, we omit further discussions.

(iv) Magnetic susceptibility, Curie temperature and critical concentration

The magnetic susceptibility χ can be obtained by differentiating $X = 2 \langle S^Z \rangle_c$ with respect to the external magnetic field as

$$\chi / \chi_0 = \frac{1}{\beta} \frac{d X}{d H} \quad (32)$$

where $\chi_0 = \frac{1}{4} (g\mu_B)^2 \beta \cdot pN$. In order to investigate the derivative in Eq.(32) we note that the self-consistent equation which determines X is of the form as

$$X = f(p, \beta, X, H) \quad (33)$$

Differentiating both sides of Eq.(33) with respect to H , we get the magnetic susceptibility as

$$\chi/\chi_0 = \frac{1}{\beta} \frac{dX}{dH} = \frac{1}{\beta} \cdot \frac{\partial f}{\partial H} / \left(1 - \frac{\partial f}{\partial X}\right) \quad (34)$$

The paramagnetic susceptibility at zero external field obtained by putting $X=0$ and $H=0$ in Eq.(34).

The Curie temperature $k_B T_c = \beta_c^{-1}$ is defined as the temperature at which the paramagnetic susceptibility diverges. Since the derivative $\partial f/\partial H$ in the r.h.s. of Eq.(34) does not diverge at any temperature, T_c for a given concentration p is determined by the equation

$$0 = 1 - \left. \frac{\partial f}{\partial X} \right|_{\beta = \beta_c, X=0, H=0} \quad (35)$$

The critical concentration p_c is defined as the concentration at which the Curie temperature becomes zero, putting $\beta_c = \infty$ in Eq.(35), the equation which determines p_c is given by

$$0 = 1 - \left. \frac{\partial f}{\partial X} \right|_{p=p_c, \beta_c = \infty, X=0, H=0} \quad (36)$$

§ 3 Results and Discussions

We calculate the Curie Temperature, critical concentration, spontaneous magnetization and paramagnetic susceptibility for the Ising and Heisenberg models, using the equations obtained in § 2. The numerical calculations are carried out for the linear chain, square, triangular, s.c., b.c.c. and f.c.c. lattices in the 1-site, 2-site and 3-site approximations.

(i) Dense limit ($p=1$)

We first consider the system in which there are no non-magnetic atoms in order to investigate the accuracy of our successive approximations in the dense limit. In this limit the system corresponds to the usual pure ferromagnet.

The numerical values of the Curie Temperature $k_B T_c / J$ for the Ising and Heisenberg models are listed in Table 1 and 2, respectively, together with the values calculated by other methods. For the linear chain ($z=2$), our successive approximations yield always the results $T_c=0$. In the 1-site approximation, which is identical with the Mamada-Takano approximation and gives no difference between the Ising and Heisenberg models, the values of T_c are already better than those obtained in the molecular field approximation, but they depend on the coordination number z only as in the Bethe and constant coupling approximations.

It is known that the critical properties depend on more details of the geometry of the lattice than the coordination number; in particular, the closed loops consisting of links of interacting pairs which appear in the non-Bethe lattices play

an important role in determining the critical properties. The primitive closed loop is a triangular loop for the closed packed lattices and is a square loop for the loose packed lattices. In a general cluster approximation it appears that we need to consider at least 3-site or 4-site clusters in order to take the effects of the triangular or square loops into account. In our approximation, however, we can consider these effects by smaller clusters, since spins outside a given cluster are not replaced by constants but by the pseudo-spins. For example, consider a cluster of two sites which are the nearest neighbors of each other. In the case of the closed packed lattices there appears the closed loop which connects two spins in the cluster and one pseudo-spin at the common nearest neighbor that these spins have. This loop approximately represents the triangular loop since the value of the pseudo-spin, which is $1/2$ or $-1/2$, affects simultaneously both of spins. For the loose packed lattices the closed loop which connects three spins and one pseudo-spin is considered in the calculation of the 3-site density matrix and it approximately represents the square loop. Consequently our 2-site and 3-site approximations exhibit reasonable dependence on the geometry of the lattice. For example, the value of T_c for the triangular lattice becomes smaller than that for the s.c. lattice which has the same coordination number $z (=6)$.

For the Ising model, the ratio of the value of T_c in the 3-site approximation to the value obtained exactly or by the

high temperature expansion method¹⁶⁾ is about 1.07 for the three dimensional s.c., b.c.c. and f.c.c. lattices and about 1.21 for the two dimensional square and triangular lattices. The ratios depend only on the dimension of the lattice. Thus we may conclude that our 3-site approximation has almost the same accuracy for the closed packed lattices as for the loose packed lattices. We also carried out the calculation for the Bethe lattices which have no closed loops and obtained the values of T_c quite close to Bethe's values in the 3-site approximation.

For the Heisenberg model, the quantum effect of the spin operators plays an important role in determining the critical properties as well as the effect of the closed loops. The quantum effect is considered for the spin operators in the clusters of two and three sites. Going from the 1-site to the 2- and 3-site approximations, the values of T_c decrease rapidly and they are less than the values for the corresponding Ising model. We find only one value for the Curie temperature. The ratio of the value in the 3-site approximation to the value obtained by the high temperature expansion method¹⁷⁾ is about 1.22 for the s.c. lattice and about 1.20 for the b.c.c. and f.c.c. lattices. The fact that these ratios are larger than those for the Ising model is attributed to the assumption of the Ising spin like behavior of the pseudo-spins. The result of our 3-site approximation is close to that of Strieb, Callen and Horwitz's 3-spin cluster approximation.

The spontaneous magnetization reaches its absolute saturation value, that is, $X = 2\langle S^Z \rangle$ reaches 1, as T decreases to zero for the Heisenberg model as well as the Ising model, contrasting with the constant coupling and Strieb, Callen and Horwitz's 3-spin cluster approximations where the magnetization is less than the absolute saturation value at $T = 0$. In Fig.2 we show the temperature dependence of $X = 2\langle S^Z \rangle$ of the Ising and Heisenberg models for the square lattice together with the exact solution by Yang¹⁸⁾ and molecular field result. Comparing our results for the Ising model with Yang's, we can see that our approximations are getting better successively from the 1-site to the 3-site one.

The high temperature susceptibility can be expressed in the form as

$$\chi = \frac{1}{4} (g\mu_B)^2 \beta N \sum_{n=0}^{\infty} A_n (\beta J)^n \quad (37)$$

Comparing the coefficients A_n with the exact values, the results of the 1-, 2- and 3-site approximations are correct to order β^2 , β^3 and β^4 , respectively for the Heisenberg model. In the case of the Ising model, all the 1-, 2- and 3- site results are correct to order β^4 for the loosepacked lattices. For the close packed lattices the 3-site results is also correct to β^4 but the 1- and 2- site ones are to β^3 .

(ii) Dilution effects

In this subsection we consider the diluted case. Since the presence of non-magnetic atoms cuts off the interactions, the Curie temperature and the spontaneous magnetization decrease. The closed loops discussed in the former subsection also play

an important role in the dilution problem. For example, consider a cluster of two sites which are a pair of the nearest neighbors. When there is a non-magnetic atom in the nearest neighbor of the cluster, the number of the interactions that are cut off by this atom is two if it is at one of the common nearest neighbors of the two sites whereas it is only one for other cases. Thus our 2- and 3-site approximations show actually that the effect of dilution depends on more details of the geometry of the lattice than the coordination number. In the following we first consider the Ising model and then consider the Heisenberg model. We have not carried out the calculation of the 3-site approximation for the f.c.c. lattice yet, since we cannot afford to spend the required computer time.

(a) Results for the Ising model

We show the concentration dependence of the Curie temperature, $T_c(p)$ in Figs.3 and 4. Fig.3 shows the results of the 1-, 2- and 3-site approximations for the triangular and s.c. lattices which have the same coordination number. Fig.4 shows the 3-site results for the square, triangular, s.c. and bcc lattices and the 2-site result for the f.c.c. lattice. As can be seen from the figures, the curves are close to straight lines for p near 1. The values of $d[T_c(p)/T_c(p=1)]/dp$ at $p=1$ are greater than 1 and change little between the 2-site and 3-site approximation, about 1.33, 1.27, 1.14 and 1.10 for the square, triangular, s.c. and b.c.c. lattices, respectively, in the 3-site approximation and about 1.09 for the f.c.c. lattice

in the 2-site approximation. The linear dependence persists down to $p \sim 0.8$ and then $dT_c(p)/dp$ starts to increase monotonically and becomes infinite as p reaches p_c . The shape of the curve does not change appreciably except for the terminal values; p_c and $T_c(p)$, when the approximation is elevated successively from the 1-site to 3-site one.

The numerical values of p_c are listed in Table 3 with the values estimated by other methods. In the 3-site approximation the values for the s.c. and b.c.c. lattices are in agreement with the critical concentration p_s estimated in the percolation problem, though the values for the two dimensional lattices are smaller than those of the percolation problem. From the behaviors of p_c (as can be seen from the Table 3), it is reasonable to expect that the value of p_c for the f.c.c. lattice would be in agreement with or at least close to the value of p_s , if the calculation for the 3-site approximation is carried out.

At present the only case where the exact solution is available in the quenched dilute system is the linear chain of the Ising spins.¹⁹⁾ Comparing the 3-site result for the paramagnetic susceptibility with the exact one, the difference is quite small. In the dilute limit ($p \sim 0$), the susceptibility obeys the Curie law.

In the dilute system there appear isolated spins and clusters, so that the value of $X = 2\langle S^z \rangle_c$ is less than 1 even at $T = 0$, when there is no external field. In our approximation the effect of the isolated clusters is considered in a self-

consistent way. When $p < 1$, $X=1$ is no longer the solution of the self-consistent equation which determines X even at $T=0$, since the clusters which have no magnetic atoms in their nearest neighbours appear with finite probabilities. Then the ferromagnetic order of the spins which interact with the pseudospins is affected, since the probability with which the pseudospin takes the value $-1/2$ becomes finite for $X < 1$. The concentration dependence of X at $T=0$ is shown in Fig.5. As the temperature increases, X decrease monotonically from the value at $T=0$.

If we investigate the thermal average of a spin, it takes various values depending on its environment. It is interesting to calculate the value $\langle S^z \rangle^{(n)}$; the averaged value of a spin on condition that the spin has n magnetic atoms in its nearest neighbors. The cluster expansion formula given by Eq.(11) can be transformed into the form as

$$M^{\{\alpha\}} = \sum_{i=1}^N \alpha_i \langle S_i^z \rangle^{\{\alpha\}} \quad (38)$$

$$\text{with } \langle S_i^z \rangle^{\{\alpha\}} = \text{Tr}_i [S_i^z \rho_i^{\{\alpha\}}] + \sum_j \{ \text{Tr}_{ij} [S_i^z \rho_{ij}^{\{\alpha\}}] - \text{Tr}_i [S_i^z \rho_i^{\{\alpha\}}] \} \\ + \dots \quad (39)$$

where the sum over j in the second term is taken over the nearest neighbors of the site i . Eq.(39) represents the cluster expansion for the thermal average of the spin operator S_i^z . Taking the configurational average of $\langle S_i^z \rangle^{\{\alpha\}}$ on condition that n magnetic atoms exist in the nearest neighbors of the spin, we obtain the value $\langle S^z \rangle^{(n)}$. Terminating the expansion given by Eq.(39) at the term which corresponds to the term $M_\rho^{\{\alpha\}}$ of Eq.(11), we can calculate the value $\langle S^z \rangle^{(n)}$ by use of the self-consistently

determined value X in the l -site approximation. It is obvious that $X^{(n)} \equiv 2 \langle S^Z \rangle^{(n)}$ satisfies the following equation.

$$X = \sum_{n=0}^Z Q_n^Z X^{(n)} \quad (40)$$

where Q_n^Z is given by Eq.(18). This procedure is rather trivial for the 1-site approximation.

We show the temperature dependence of X and $X^{(n)}$ obtained in the 3-site approximation for the square lattice in Fig.6. Of course, $X^{(0)}$ is always zero, since it means the value for an isolated spin. As can be seen from the figure, the shape of the $X^{(n)} - T$ curve depends on the number n . Especially the behavior of $X^{(1)}$ is of interest. $X^{(1)}/2$ is the averaged value of a spin which has only one magnetic atom in its nearest neighbors. For p near 1, the behavior of $X^{(1)}$ is approximately expressed as

$$X^{(1)} = X \cdot \tanh (\beta J/2) \quad (41)$$

As p decreases, however, the value of $X^{(1)}$ becomes smaller than that given by the r.h.s. of Eq.(41) even at $T=0$.

b) Results for the Heisenberg model

First we consider the critical concentration. The numerical values of p_c are listed in Table 3. In the 3-site approximation the values are much the same as the corresponding values for the Ising model, though there is some difference in the case of the triangular lattice. This result seems to support the prediction by Elliott et.al.⁹⁾ that p_c is a geometrical property of the

lattice and is the same in the Ising and Heisenberg models. However, the behavior of $T_c(p)$ for the Heisenberg model is quite different from that for the Ising model. We show the concentration dependence of $T_c(p)$ in Figs. 4 and 7. Fig. 4 shows the results obtained by the 3-site approximation (for the f.c.c. lattice we show the results of the 2-site approximation) together with those for the Ising model. Fig. 7 shows the results of the 1-, 2- and 3-site approximations for the triangular and s.c. lattices. For p near 1, $T_c(p)$ decreases linearly and the values of $d[T_c(p)/T_c(p=1)]/dp$ at $p=1$ are about 1.90, 1.69, 1.35 and 1.24 for the square, triangular, s.c. and b.c.c. lattices, respectively, in the 3-site approximation. If we compare the results for the Heisenberg and Ising models, the following inequality relation is realized for p near 1.

$$\frac{d}{dp} \left[\frac{T_c(p)}{T_c(p=1)} \right]_{[H]} > \frac{d}{dp} \left[\frac{T_c(p)}{T_c(p=1)} \right]_{[I]} \quad (42)$$

This indicates that the ferromagnetic state of the Heisenberg model is more readily destroyed by non-magnetic atoms than that of the Ising model.

As p decreases from 1, $dT_c(p)/dp$ increases monotonically in the high concentration region as in the Ising model. If this behavior persists down to the low concentration region, $T_c(p)$ should become zero at the concentration which is appreciably greater than the value of p_c we obtained. In the 3-site approximation, however, there appears a region of p where $d^2T_c(p)/dp^2$ becomes positive and the $T_c(p)$ curve has a "trail"

for p near p_c . Heap⁶⁾ and Elliott⁵⁾ obtained the similar behavior of $T_c(p)$ based on the concentration expansion of the susceptibility. However, we cannot conclude definitely that this is what happens actually in the Heisenberg model, because the shape of the $T_c(p)$ curve changes considerably as we go from the 2-site to the 3-site approximation; we cannot deny the possibility that the "trail" of the $T_c(p)$ curve disappears if we go beyond the 3-site approximation. It is noted that in 2- and 3-site our approximations there appears the anti-Curie temperature for p very near p_c , although we cannot show it in the figures because the region of p where the anti-Curie temperature appears is quite narrow.

The value of X at $T=0$ decreases monotonically as p decreases. In the 3-site approximation the X - p curves are nearly identical with those for the Ising model except for the case of the triangular lattice. As the temperature increases from zero, X decreases monotonically in the case of the high concentration. In the case of smaller p , however, there appears a region of p and T where X undergoes a small but finite increase as T increases. Such a increase of $X=2\langle S^z \rangle$ with increasing T appears also in the case of the constant coupling and Strieb, Callen and Horwitz's 3-spin approximations. This region becomes narrower with respect to both p and T in the 3-site approximation than in the 2-site one. This behavior of X corresponds to the appearance of the anti-Curie temperature.

§ 4 Summary

We have presented a new approximation to treat the dilute Ising and Heisenberg ferromagnets. In this approximation the value of $X = 2\langle S^Z \rangle_c$ is self-consistently determined. In order to derive the self-consistent equation for X , we use the cluster expansion formula given by Eq.(11) and the approximately reduced density matrix for a given cluster. The reduced density matrix is obtained by replacing the spin operators not included in the cluster by stochastic variables (the pseudospins) whose probability distribution is given as a function X .

The approximation can be improved systematically by expanding the size of the clusters to be considered. In the 2-site and 3-site approximations we have obtained satisfactory results for the Curie temperature, critical concentration, spontaneous magnetization and magnetic susceptibility, compared with those obtained by other methods. The results exhibit good dependence on more details of the geometry of the lattice than the coordination number. For the three dimensional lattices the values of p_c obtained in the 3-site approximation are in agreement with those of the site percolation problem and are nearly same for the Ising and Heisenberg models. The behavior of $T_c(p)$ for the Heisenberg model, however, is quite different from that for the Ising model; $T_c(p)$ for the Heisenberg model decreases more rapidly than for the Ising model as p decreases, and the $T_c(p)$ curve has a "trail" for p near p_c . It is noted that the averaged value of a spin with a specified environment can be

calculated without difficulties in our approximation.

Although we do not show the results, the two spin correlation function and the specific heat can be calculated by use of the 2-site and 3-site density matrices obtained in § 2.

It is interesting to investigate the system in which the next nearest neighbor interaction exists as well as the nearest neighbor interaction, or the quasi one and two dimensional systems with the weak interactions between chains or planes. Our approximation can be applied without difficulties to these systems. We will report the results in part II.

Appendix A. Proof of the cluster expansion formula of Eqs.(11)
and (12)

First we substitute Eq.(12) in the r.h.s. of Eq.(11). The r.h.s. of Eq.(11) is written as

$$\sum_{\ell=1}^N M_{\ell} = \sum_{\ell=1}^N \sum_{\{\ell\}k=1}^{\ell} (-1)^{\ell-k} \sum_{\{k\}} \cdot m_{\{k\}} \quad (\text{A -1})$$

with

$$m_{\{k\}} = \text{Tr}_{\{k\}} [(\sum_{i \in \{k\}} \alpha_i S_i^z) \cdot \rho_{\{k\}}] \quad (\text{A -2})$$

where we omit the symbol $\{\alpha\}$ for brevity. The double sum

$$\sum_{\ell=1}^N \sum_{k=1}^{\ell} () \text{ is equivalent to } \sum_{k=1}^N \sum_{\ell=k}^N (), \text{ so that}$$

$$\sum_{\ell=1}^N M_{\ell} = \sum_{k=1}^N \sum_{\{k\}} m_{\{k\}} \cdot \sum_{\ell=k}^N (-1)^{\ell-k} \sum_{\{\ell\}} \quad (\text{A -3})$$

It must be noted that the last sum over $\{\ell\}$ is taken over the clusters of ℓ sites which contain the cluster $\{k\}$ ($k \leq \ell$) as a subcluster; the cluster $\{\ell\}$ is composed of the k -sites belonging to the cluster $\{k\}$ and $(\ell-k)$ sites which are chosen from $(N-k)$ sites not belonging to $\{k\}$. Thus the last two summations in Eq.(A-2) with a given $\{k\}$ is calculated

$$\sum_{\ell=k}^N (-1)^{\ell-k} \sum_{\{\ell\}} = \sum_{\ell=k}^N (-1)^{\ell-k} \binom{N-k}{\ell-k} = \sum_{\ell=0}^{N-k} (-1)^{\ell} \binom{N-k}{\ell} \quad (\text{A -4})$$

Eq.(A -4) is the binomial expansion of $(1-x)^{N-k}$ with $x=1$; that is, it is equal to zero except for the case $k=N$, in which case it is unity. Thus

$$\begin{aligned}
\sum_{\ell=1}^N M_{\ell} &= \sum_{k=1}^N \sum_{\{k\}} m_{\{k\}} \cdot \delta_{kN} = m_{\{N\}} \\
&= \text{Tr}_{\{N\}} \left[\left(\sum_{i=1}^N \alpha_i S_i^z \right) \rho_{\{N\}} \right]
\end{aligned}
\tag{A -5}$$

This equation is equal to the exact expression given by Eq.(4). It is noted that Kubo²⁰⁾ and Strieb, Callen and Horwitz¹⁵⁾ proved the expansion theorem of the density matrix or of the free energy in the similar fashion as ours.

Appendix B Proof of the theorem (a)

The contribution of a cluster $\{\ell\}$ to M_ℓ given by Eq.(12) is written as

$$M_{\{\ell\}} = \sum_{k=1}^{\ell} \sum_{\{k\}} (-1)^{\ell-k} \cdot m_{\{k\}} \quad (\text{B -1})$$

where $\{k\}$ is the subcluster belonging to $\{\ell\}$ and $m_{\{k\}}$ is defined by Eq.(A-2). The symbol $\{\alpha\}$ is omitted. If the cluster $\{\ell\}$ is composed of two subclusters $\{m\}$ and $\{n\}$ which are not linked to each other, the traces on the spin operators in the sites belonging $\{m\}$ and $\{n\}$ can be taken separately in our approximate density matrices; that is,

$$\text{Tr}_{\{n\}} \rho_{\{\ell\}} = \rho_{\{m\}}, \quad \text{Tr}_{\{m\}} \rho_{\{\ell\}} = \rho_{\{n\}} \quad (\text{B -2})$$

If $\{k\}$ is divided into two subclusters, one is $\{kn\}$, the sites of which belong to $\{n\}$ and the other is $\{km\}$, the sites of which belong to $\{m\}$, $m_{\{k\}}$ is divided into two parts

$$m_{\{k\}} = m_{\{kn\}} + m_{\{km\}} \quad (\text{B -3})$$

when we define $m_{\{0\}}$ ($\{0\}$ is the cluster which contains no sites) as to be zero, Eq.(B-3) ^{also} holds for ^{those} clusters, all the sites of which belong to $\{m\}$ or $\{n\}$; that is, $kn=0$ or $km=0$. By use of Eq.(B-3), $M_{\{\ell\}}$ is written as

$$M_{\{\ell\}} = \sum_{k=1}^{\ell} \sum_{\{k\}} (-1)^{\ell-k} \cdot m_{\{kn\}} + \sum_{k=1}^{\ell} \sum_{\{k\}} (-1)^{\ell-k} \cdot m_{\{km\}} \quad (\text{B -4})$$

We first prove that the first term of the r.h.s. of Eq.(B -4) is equal to zero. Dividing the sum over {k} into the sums over {kn} and {km},

$$\sum_{k=1}^{\ell} \sum_{\{k\}} (-1)^{\ell-k} \cdot m_{\{kn\}} = \sum_{kn=1}^n \sum_{\{kn\}} (-1)^{\ell-kn} \cdot m_{\{kn\}} \cdot \sum_{km=0}^{\ell-n} (-1)^{-km} \cdot \sum_{\{km\}} \quad (\text{B -5})$$

$\sum_{\{km\}}$ is the number of ways of choosing km sites out of $(\ell-n)$ sites, so that

$$\begin{aligned} \sum_{km=0}^{\ell-n} (-1)^{-km} \sum_{\{km\}} &= \sum_{km=0}^{\ell-n} (-1)^{km} \binom{\ell-n}{km} \\ &= 0 \quad (\text{since } m = \ell-n \geq 1) \quad (\text{B -6}) \end{aligned}$$

In the same way we can prove that the second term of the r.h.s. of Eq.(B -4) is equal to zero, since $n \geq 1$.

Proof of the theorem (b)

When a site i belongs to the cluster $\{\ell\}$, the summation over $\{k\}$ in the r.h.s. of Eq.(B-1) can be written as

$$M_{\{\ell\}} = \sum_{k'=1}^{\ell-1} \sum_{\{k'\}} [(-1)^{\ell-k'} \cdot m_{\{k'\}} + (-1)^{\ell-k'-1} \cdot m_{\{k'+i\}}] + (-1)^{\ell-1} \cdot m_{\{i\}} \quad (\text{B -7})$$

where the sum over $\{k'\}$ is taken over all subclusters which do not contain the site i . $\{k'+i\}$ means the cluster of $(k'+1)$ sites made of the site i and the cluster $\{k'\}$ and $\{i\}$ means the cluster which contains the site i only. If the occupation number α_i of the site i is equal to zero, $m_{\{i\}} = 0$ and $m_{\{k'+i\}} = m_{\{k'\}}$, since the density matrix $\rho_{\{k'+i\}}$ is equal to $\rho_{\{k'\}}$. Thus

$$M_{\{\ell\}} = \sum_{k'=1}^{\ell-1} \sum_{\{k'\}} [(-1)^{\ell-k'} \cdot m_{\{k'\}} + (-1)^{\ell-k'-1} \cdot m_{\{k'\}}] = 0 \quad (\text{B -8})$$

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Table 1 Numerical values of $k_B T_c / J$ at $p=1$ for the Ising model

	sq.	p.t.	sc	bcc	fcc
Bethe	1.443	2.466	2.466	3.476	5.485
Matsudaira	1.311	2.136	2.366	3.332	
High temperature expansion*)	1.135	1.820	2.255	3.177	4.897
Our 1-site (Mamada-Takano)	1.545	2.537	2.537	3.530	5.522
Our 2-site	1.466	2.307	2.479	3.484	5.327
Our 3-site	1.367	2.203	2.410	3.382	5.228

*) For the sq. and pt. lattices the values are exact.

Table 2 Numerical values of $k_B T_c / J$ at $p=1$ for the Heisenberg model
 $S=1/2$

	sq.	p.t.	sc	bcc	fcc
Constant coupling	none	1.821	1.821	2.884	4.932
SCH's 3-spin cluster	1.144	1.596	2.082	3.048	4.650
Hith temperature expansion			1.68 ±0.01	2.53 ±0.015	4.02 ±0.02
Our 2-site	1.177	1.979	2.197	3.211	5.044
Our 3-site	1.001	1.735	2.062	3.026	4.811

Table 3 Numerical values of P_c for the Ising [I] and Heisenberg [H] models

	sq.	p.t.	sc	bcc	fcc
Matsudaira [I]	0.484	0.338	0.310	0.234	
Spin Wave ^{*)} [H]			>0.44	> 0.39	>0.345
EHMR ^{**)}	0.48	0.36	0.28	0.22	0.18
Site percolation	0.59 ±0.01	0.5 (exact)	0.307 ±0.010	0.243 ±0.010	0.199 ±0.008
Our 1-site (Mamada-Takano)	0.428	0.293	0.293	0.223	0.150
Our 2-site [I]	0.431	0.377	0.283	0.211	0.180
[H]	0.472	0.401	0.312	0.233	0.194
Our 3-site [I]	0.488	0.410	0.301	0.235	
[H]	0.485	0.440	0.306	0.233	

^{*)} Murray¹⁰⁾

^{**)} Elliott, Heap, Morgan and Rushbrooke⁹⁾

Figure Captions

- Fig. 1 Figures of the distinct clusters which contains one, two and three sites. White circles represent the sites in the clusters and black circles represent the nearest neighbouring sites. (a), (b), the clusters which appear in the square and triangular lattices, respectively.
- Fig. 2 Temperature dependence of $X = 2\langle S^Z \rangle$ at $p = 1$ for the square lattice. Temperature is normalized by the Curie temperature. Full lines: the 1-, 2- and 3-site approximation for the Ising model. Dot dashed line: the 3-site approximation for the Heisenberg model. Dashed line: Exact solution by Yang. Dotted line: the molecular field approximation.
- Fig. 3 Concentration dependence of the normalized Curie temperature $T_c(p)/T_c(p=1)$ for the s.c. and triangular lattices, results of the Ising model. Dotted line: the 1-site approximations. Full lines: the 2- and 3-site approximation for the s.c. lattice. Dashed lines: the 2- and 3-site approximations for the triangular lattice.
- Fig. 4 Concentration dependence of the Curie temperature for the square (sq.), triangular (pt), s.c. and b.c.c. lattices (results of the 3-site approximation) and for the f.c.c. lattice (results of the 2-site approximation). Full lines: the Ising model. Dashed lines: the Heisenberg model.
- Fig. 5 Concentration dependence of $X = 2\langle S^Z \rangle_c$ at $T = 0$ for the square, s.c. and b.c.c. lattices. The curves are obtained in the 3-site approximation for the Ising model.

Fig. 6 Temperature dependence of $X = 2 \langle S^z \rangle_c$ and $X^{(N)}$ for $p = 1, 0.9, 0.7, 0.55$ and 0.5 for the square lattice. The curves are obtained in the 3-site approximation for the Ising model. Full lines, dotted lines, dashed lines, double dot dashed lines and dot dashed lines represent $X, X^{(1)}, X^{(2)}, X^{(3)}$ and $X^{(4)}$, respectively. For $p = 0.5$, the curve for $X^{(3)}$ is not shown since it is almost the same as that for $X^{(2)}$.

Fig. 7 Concentration dependence of the normalized Curie temperature $T_c(p)/T_c(p=1)$ for the s.c. and triangular lattices, results of the Heisenberg model. Dotted line: the 1-site approximation. Full lines: the 2- and 3-site approximations for the sc lattice. Dashed lines: the 2- and 3-site approximations for the triangular lattice.

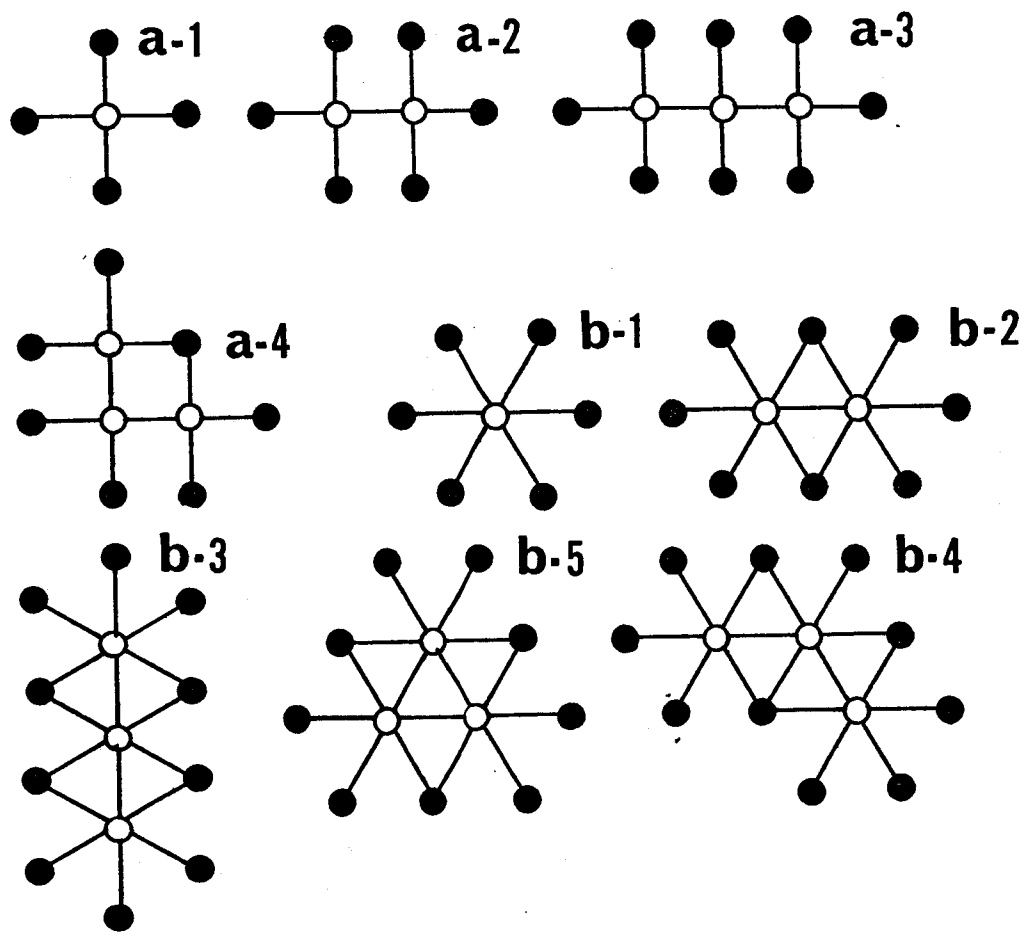
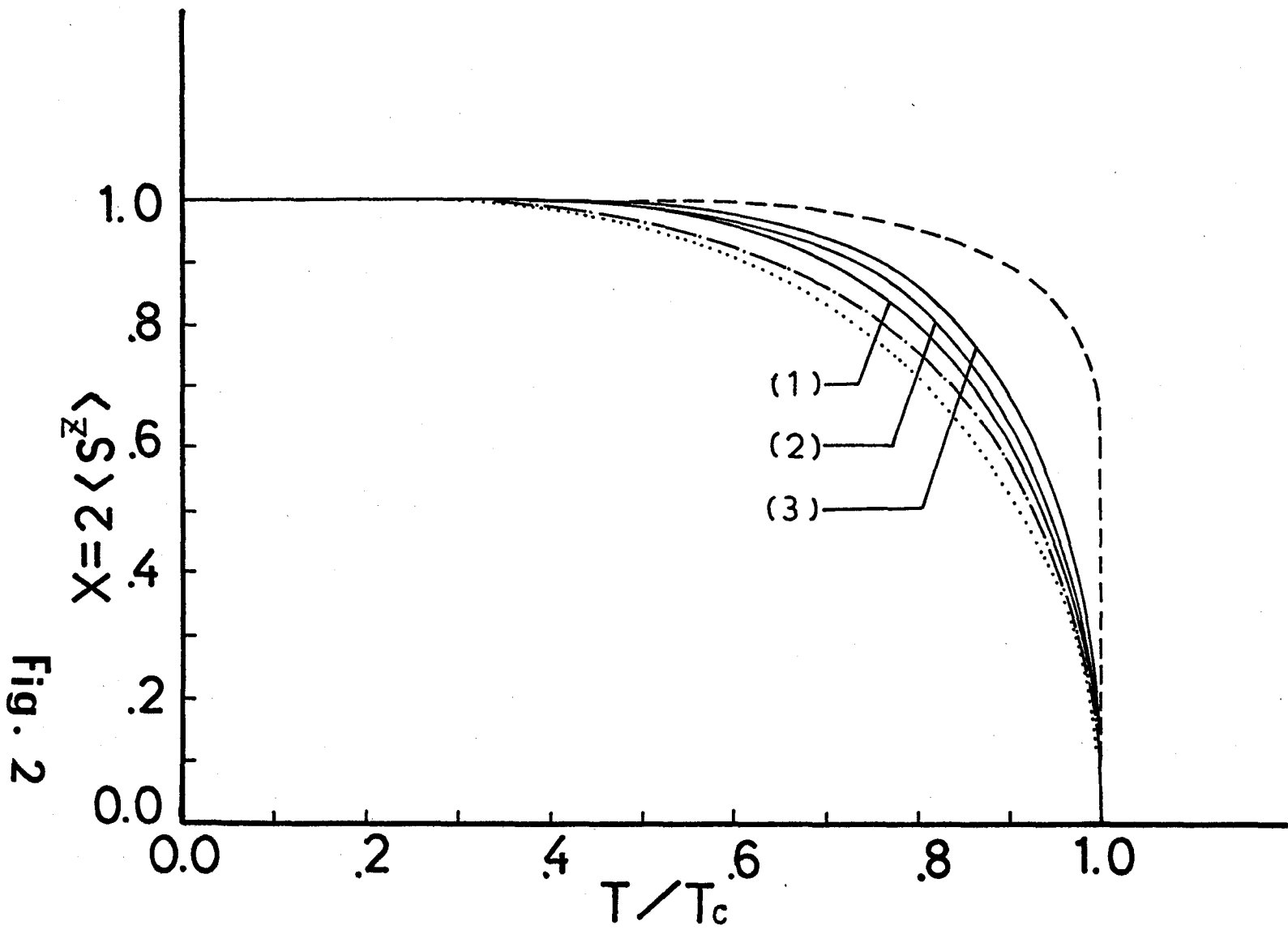


Fig. 1



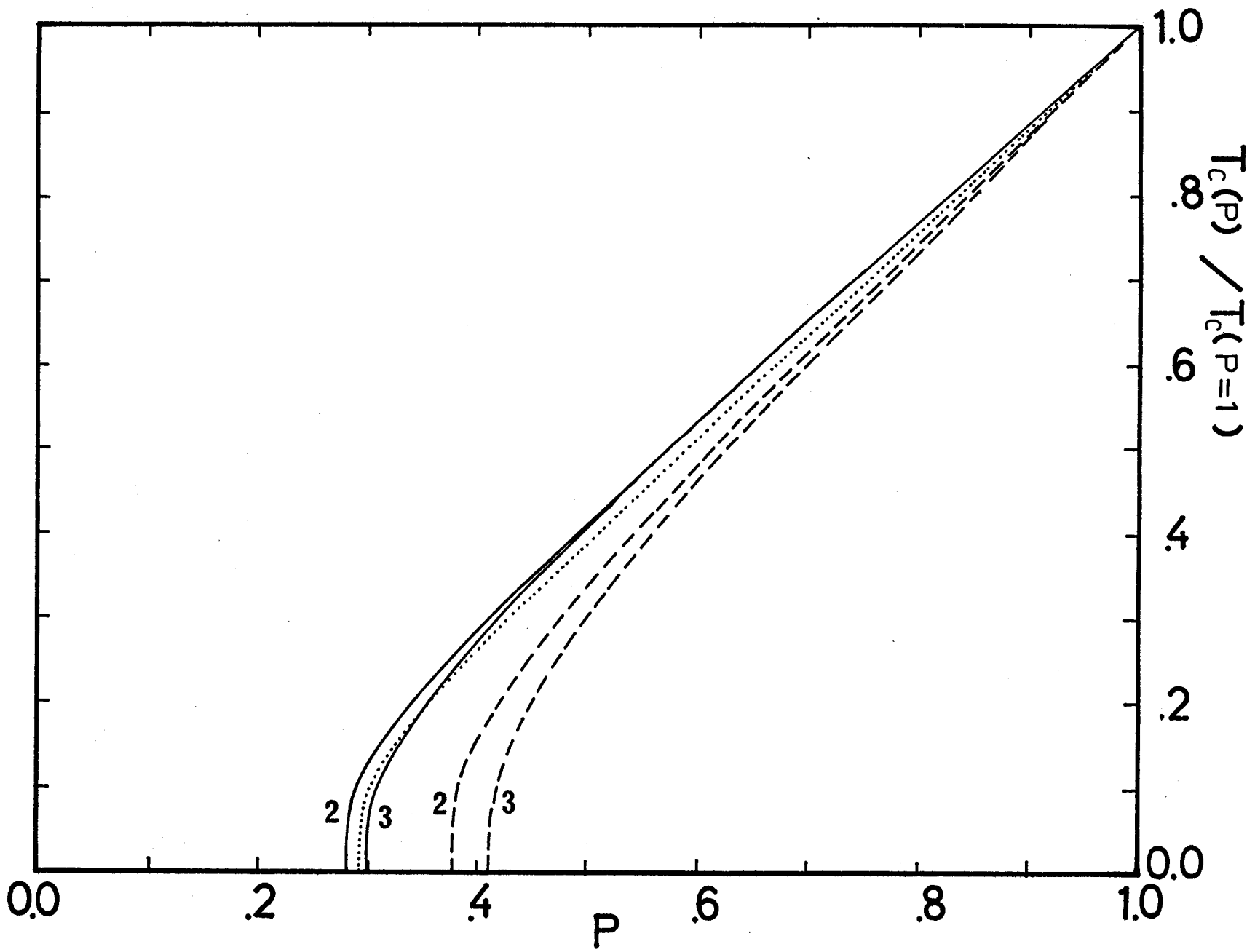


Fig. 3

Fig. 4

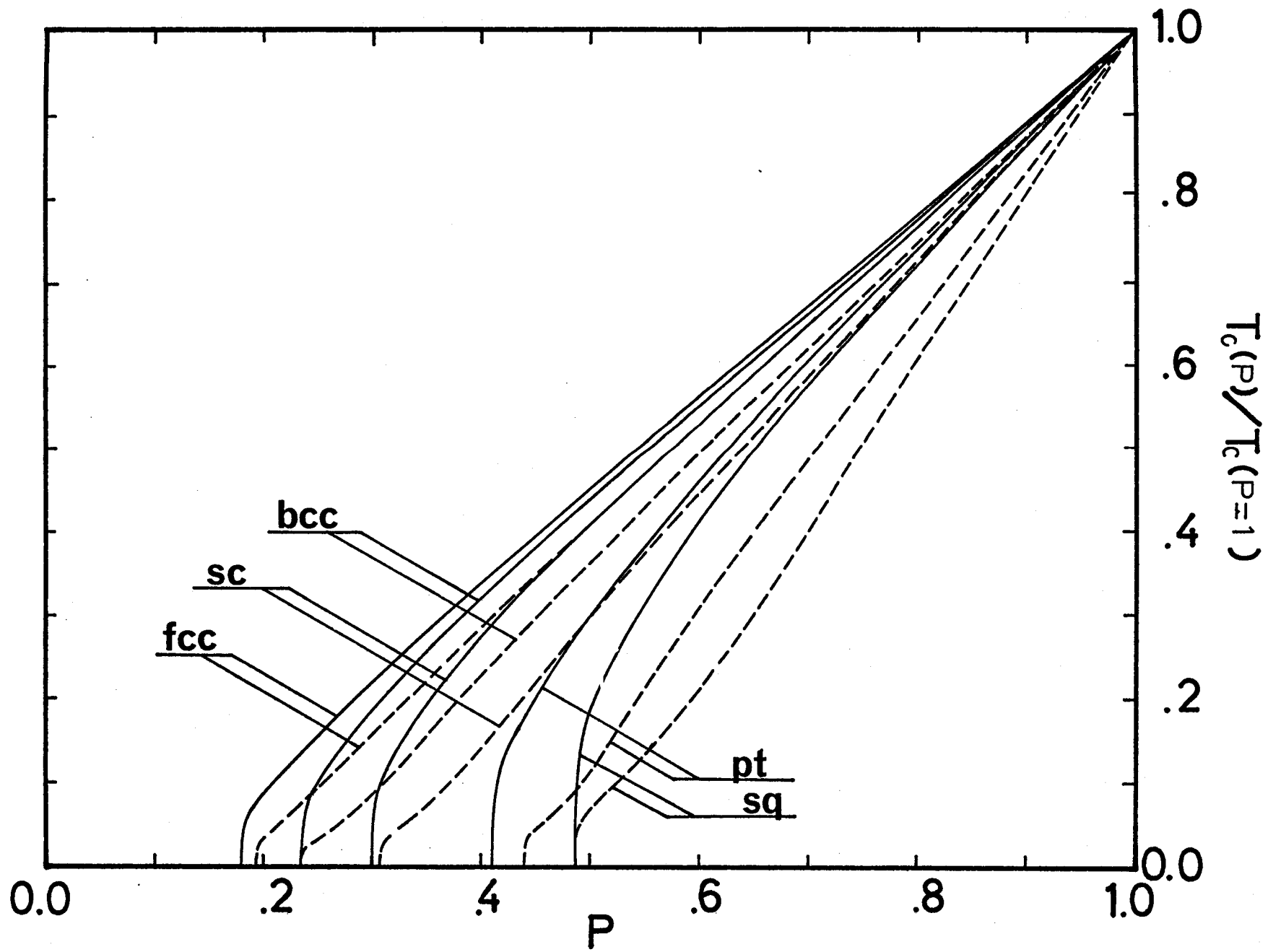
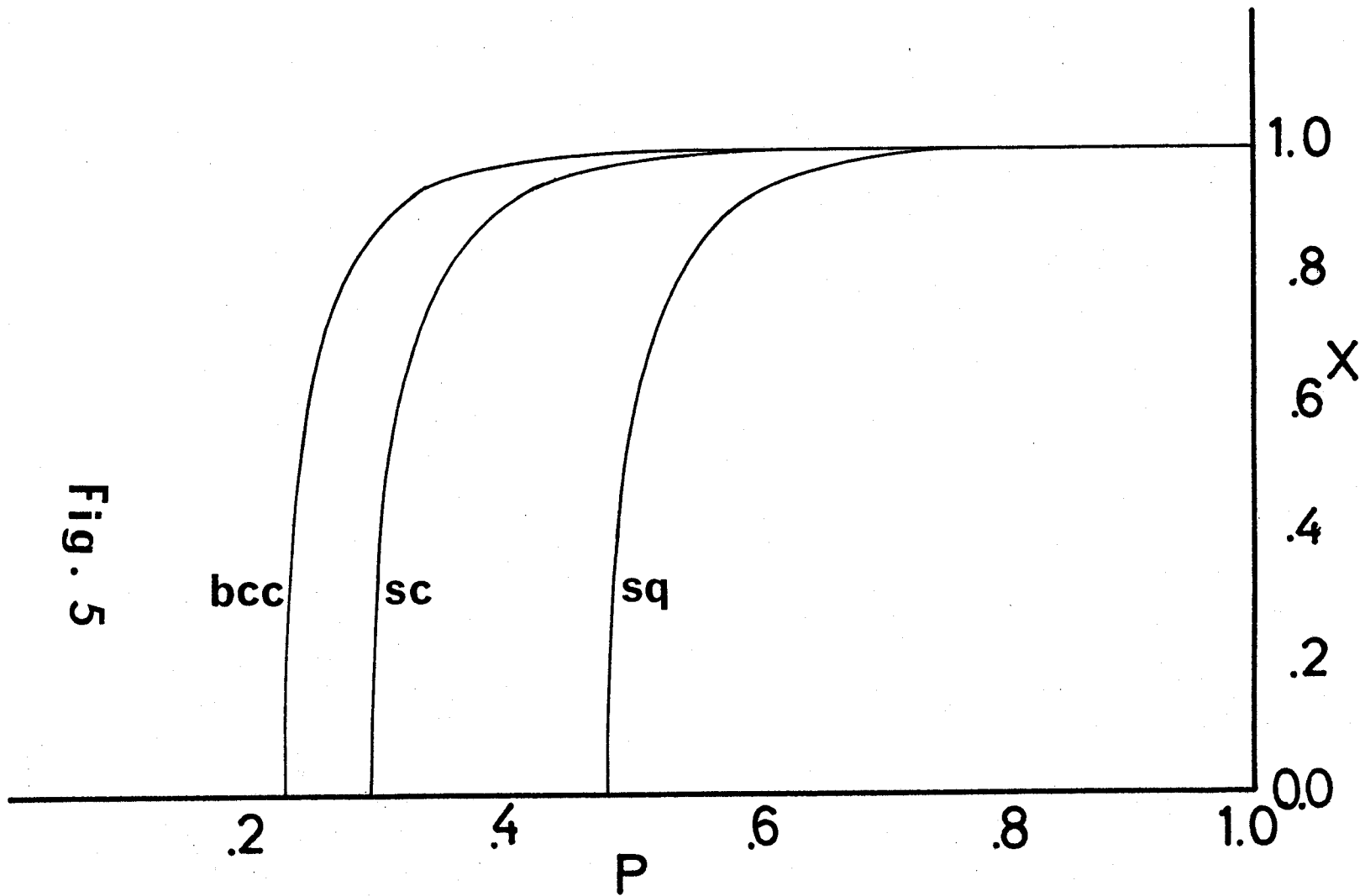


Fig. 5



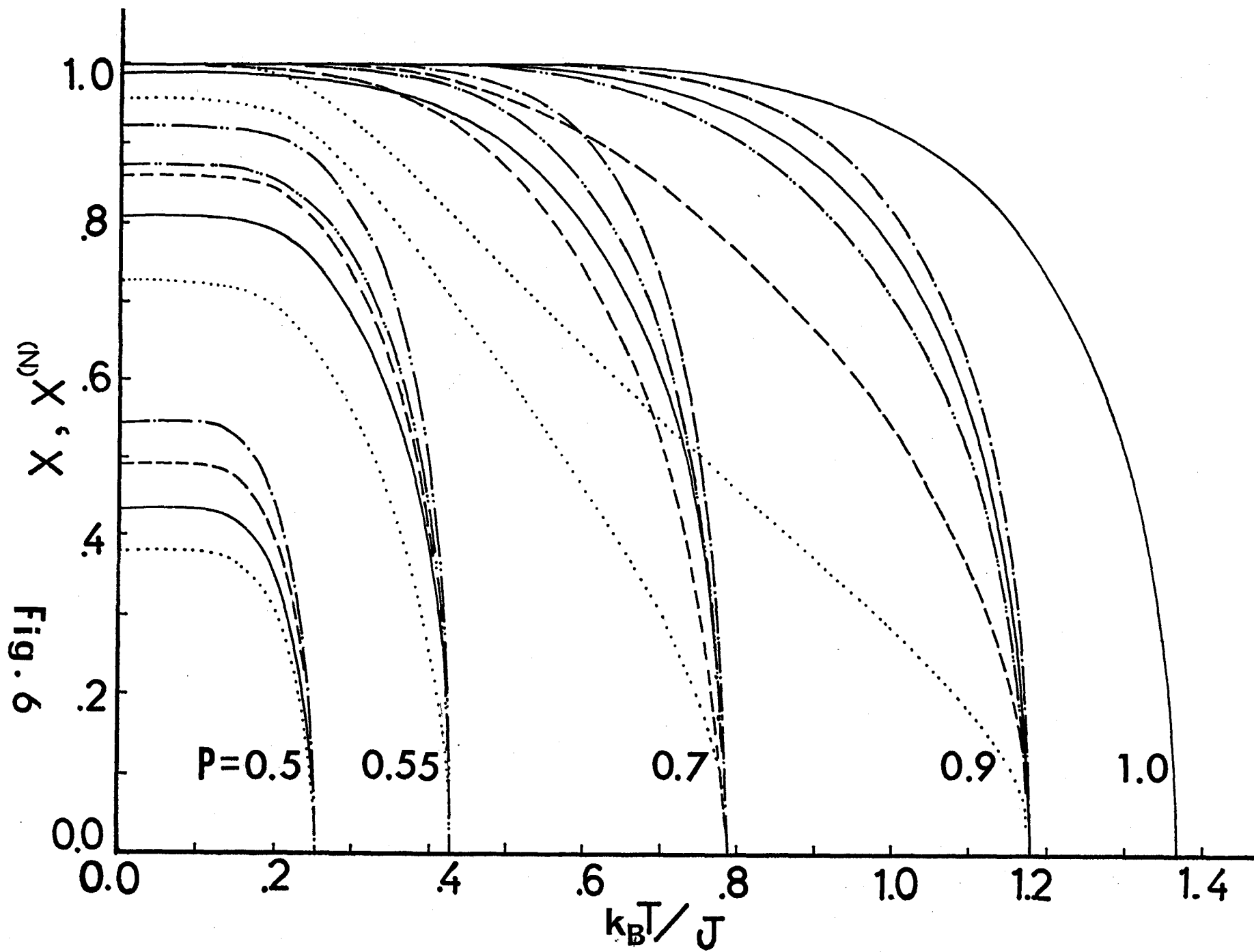


Fig. 6

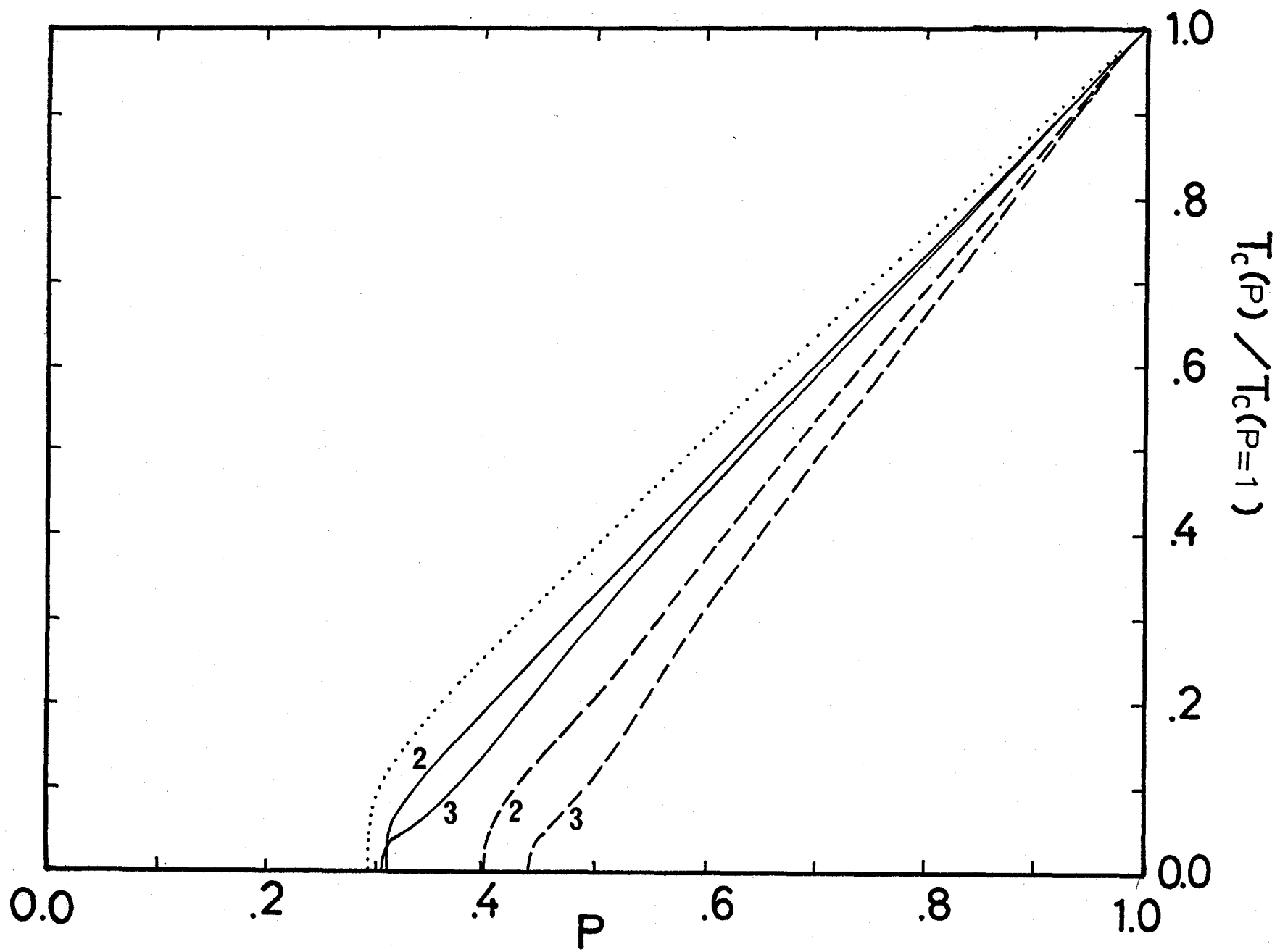


Fig. 7

Part II

Application of a Cluster Approximation to Ferromagnets
with the First- and Second-Neighbor Interactions

§ 1 Introduction

In the preceding part (hereafter referred to as [I]), we proposed a new cluster approximation which can be improved systematically. This approximation is applicable to the dilute magnetic system (a random mixture of magnetic and non-magnetic atoms) as well as to the pure system. In [I] we have investigated the dilute Ising and Heisenberg magnets with the nearest-neighbor interaction in this approximation. In the present part we shall investigate the pure and dilute Ising ferromagnets including the second-neighbor interaction by extending the cluster approximation to this case.

Our approximation is based on a cluster expansion of the magnetization and an approximate reduction of the density matrix. We introduce a "pseudo-spin" which is a stochastic variable taking the values $1/2$ and $-1/2$ with self-consistently determined probabilities. When we reduce the density matrix of the total system to that of a small cluster, we replace spins not included in the cluster by the pseudo-spins. The density matrix of the cluster is obtained by averaging over all possible configurations of pseudo-spins. The probability distribution of a pseudo-spin is approximately determined by the condition that its average value is equal to the magnetization per spin (divided by $g\mu_B$). On the other hand we expand the expression of the magnetization into a cluster series and obtain the self-consistent equation which determines the magnetization per spin. The 1-site approximation is identical

with that given by Mamada and Takano.¹⁾ The approximation is improved systematically by expanding the size of clusters to be considered. The 2-site approximation already reflects the geometry of the lattice in more detail than the coordination number.

In § 2 we shall derive the self-consistent equation in the 1-site and 2-site approximation. The equations for the susceptibility, Curie temperature, critical concentration, pair correlation function and specific heat is also given.

Investigation of the Ising model with the first- and second-neighbor interactions is of interest not only theoretically but also for a better description of real magnetic systems. In the present paper, we study the case where the first-neighbor interaction J_1 is positive, i.e. ferromagnetic and the second-neighbor interaction takes various values. If J_2 is positive, the ferromagnetic long range order is reinforced and the Curie temperature increases. On the other hand, if J_2 is negative, the Curie temperature decreases, and below some critical value of $R=J_2/J_1$ the model does not show ferromagnetic ordering. The model, however, is not solved exactly even for the two-dimensional lattices since the interactions' cross each other (so-called "crossed-bond" problem). In recent years several works have been done on this problem. Dalton and Wood²⁾ studied the two- and three-dimensional Ising models by the method of power series expansion for the partition function and thermodynamic functions. They determined critical

indices and calculated the dependence of the Curie temperature on the value of R for $R > 0$. Kawakani and Osawa,³⁾ Gibberd⁴⁾ and Fan and Wu⁵⁾ calculated the Curie temperature for the square lattice. They write the partition function as the vacuum-to-vacuum expectation value of fermion operators. Then using the anticommutation relation of the fermion operators, the partition function is written as the expectation value of the time ordered exponentials in which a quartic term of fermion operators appears in addition to the quadratic terms. They ignore the quartic term and calculate the Curie temperature approximately. If $J_2 = 0$, their result is exact. For $R > 0$ their result agrees well with the result of Dalton and Wood. Fan and Wu obtained the result that the Curie temperature decreases smoothly as R decreases and becomes zero at $R = -1/2$.

Takase⁶⁾ also studied the Ising model of the square lattice in the approximation which is identical with our 1-site approximation. His approximation, however, is based on a different principle from ours. He has calculated the Curie temperature and spontaneous magnetization. On the basis of a free energy expression, he concluded that the first-order phase transition from ferromagnetic to paramagnetic phase occurs for range $-0.238 > R > -0.5$. As will be discussed in § 3, this conclusion which contradicts with that of the present theory is hard to believe.

In § 3 we shall investigate the thermodynamic properties of the Ising model with the first- and second-neighbor inter-

actions. The dependence of the Curie temperature on $R=J_2/J_1$ is calculated for the square, triangular, s.c., b.c.c. and f.c.c. lattices. For the square lattice the spontaneous magnetization, pair correlation function and specific heat are calculated, which show reasonable dependences on temperature and the ratio of the interaction constants, R . The Curie temperature decreases smoothly with decreasing R to vanish at $R=-1/2$ in agreement with Faw and Wu's result.

In § 4 we shall investigate the random mixture of magnetic and non-magnetic atoms. We assume the perfect randomness of the atomic distribution. The presence of non-magnetic atoms results in the decrease of the Curie temperature. Below the critical concentration of magnetic atoms the system does not show any phase transition. As was discussed in [I], the critical concentration would correspond to the formation of a infinite network of adjacent spins. The value of the critical concentration and concentration dependence of the Curie temperature have been calculated by many authors for the case where only the nearest-neighbor interaction exists. The relevant references are found in [I].

The role of the farther neighbor interaction has been qualitatively discussed by Brout.⁷⁾ The second-neighbor interaction, even if it is small, is expected to play an important role at least at low temperatures. Especially it has been discussed by Elliott, Heap, Mogan and Rushbrook⁸⁾ that the critical concentration depends on the range of interactions

and not on the strength. Idogaki and Uryu⁹⁾ have calculated the concentration dependence of the Curie temperature for various values of $R=J_2/J_1 > 0$ by the method of concentration expansion. They obtained the result that the critical concentration does not depend on the strength of J_2 and the Curie temperature vs. concentration curve has a "trail" in the region of concentration near the critical one when R is small.

We shall calculate the critical concentration, Curie temperature, paramagnetic susceptibility, spontaneous magnetization, pair correlation function and specific heat in our 2-site approximation. The calculation is carried out for the square lattice including the second-neighbor interaction. We shall find the remarkable role of the weak second-neighbor interaction in the low temperature region. The calculation for the s.c., b.c.c. and f.c.c. lattices are carried out in our 1-site approximation.

§ 2 Formalism

§ 2-1 The cluster approximation

In this section we extend the cluster approximation proposed in [I] to the case where the second-neighbor interaction also exists. We consider the Ising model of $S = 1/2$ with the ferromagnetic first-neighbor interaction $J_1 > 0$ and the second-neighbor interaction J_2 ; the sign and magnitude of J_2 is assumed to be such that the long range order, if it appears, is ferromagnetic. The spacial configuration of magnetic and non-magnetic atoms is denoted by a set of occupation numbers, $\{\alpha\} = \{\alpha_1, \alpha_2, \dots, \alpha_N\}$, where the occupation number α_i is equal to 1 if a magnetic atom occupies the site i and 0 if a non-magnetic atom occupies the site. For a given atomic configuration $\{\alpha\}$, the Hamiltonian of the system is written as

$$H^{\{\alpha\}} = -2J_1 \sum^{(1)} \alpha_i \alpha_j S_i^Z S_j^Z - 2J_2 \sum^{(2)} \alpha_i \alpha_k S_i^Z S_k^Z - g\mu_B H \sum \alpha_i S_i^Z \quad (2 - 1)$$

where H is the external magnetic field and the sums $\sum^{(1)}$ and $\sum^{(2)}$ are taken over all the first- and second-neighbors, respectively, and the last sum is over all sites.

In our cluster approximation, the average value of a spin, $X = 2\langle S^Z \rangle_c$, where $\langle \dots \rangle_c$ means the average over the atomic configuration as well as the thermal average, is determined self-consistently. The procedure to derive the self-consistent equation is the same as that discussed in [I]. We briefly summarize, without proof, the procedure in the following.

First we define the approximate density matrix $\rho_{\{\ell\}}$ of a cluster $\{\ell\}$, consisting of ℓ sites. (We will omit the symbol $\{\alpha\}$ hereafter when there is no confusion.) We replace the spins S_i^Z not belonging to the cluster by stochastic variables \bar{S}_i (pseudo-spins) which take the values $1/2$ and $-1/2$ with the probabilities $(1+X)/2$ and $(1-X)/2$, respectively. In this approximation we assume that the pseudo-spins are independent of one another and the average value of a pseudo-spin is equal to the average value of a spin, $\langle S^Z \rangle_c = X/2$. For a given configuration of the pseudo-spins, the density matrix $\rho_{\{\ell\}, \bar{S}}$ of the cluster $\{\ell\}$ is expressed in terms of the effective Hamiltonian $\mathcal{H}_{\{\ell\}, \bar{S}}$ thus derived as

$$\rho_{\{\ell\}, \bar{S}} = \exp[-\beta \mathcal{H}_{\{\ell\}, \bar{S}}] / \text{Tr}_{\{\ell\}} \exp[-\beta \mathcal{H}_{\{\ell\}, \bar{S}}] \quad (2 - 2)$$

where $\beta = 1/k_B T$ and $\text{Tr}_{\{\ell\}}$ means the trace taken over the spins which belong to the cluster. Taking the average over all possible configuration of the pseudo-spins, the density matrix $\rho_{\{\ell\}}$ is calculated as

$$\rho_{\{\ell\}} = \langle \rho_{\{\ell\}, \bar{S}} \rangle_{\bar{S}} \quad (2 - 3)$$

Noting that the pseudo-spins that are not at the first- and second-neighbors of the cluster $\{\ell\}$ drop out, we only consider the pseudo-spins that are at the first- or second-neighbors of the cluster when we take the average $\langle \dots \rangle_{\bar{S}}$.

Next we expand the magnetization of the system (divided by $g\mu_B$) which is given by

$$M^{\{\alpha\}} = \text{Tr} [(\sum \alpha_i S_i^Z) \cdot \rho_N] \quad (2 - 4)$$

into the cluster series, where ρ_N is the density matrix of the total system. The following cluster expansion formula is proved.

$$M^{\{\alpha\}} = \sum_{\ell=1}^N M_{\ell}^{\{\alpha\}} \quad (2 - 5)$$

with

$$M_{\ell}^{\{\alpha\}} = \sum_{\{\ell\}} \sum_{k=1}^{\ell} (-1)^{\ell-k} \sum_{\{k\}} \cdot \text{Tr}_{\{k\}} [(\sum_{i \in \{k\}} \alpha_i S_i^Z) \cdot \rho_{\{k\}}] \quad (2 - 6)$$

where the sum over $\{\ell\}$ is taken over all possible clusters of ℓ sites and the sum over $\{k\}$ is taken over all subclusters of k sites belonging to a given $\{\ell\}$. By use of the theorems proved in the appendix of [I], the clusters $\{\ell\}$ which give non zero contribution to M_{ℓ} are the linked clusters of ℓ spins, that is, all occupation numbers α_i 's of the sites belonging to $\{\ell\}$ are equal to 1 and all the spins are connected by the interactions.

When the concentration of magnetic atoms is equal to p , there are pN spins in the system and the probabilities of $\alpha=1$ and $\alpha=0$ are given by p and $q=1-p$, respectively. (We assume the perfect randomness of atomic configuration). Then taking

the configurational average of Eq.(2-5), the value of X is given by

$$X/2 = \langle S^z \rangle_c = \langle M^{\{\alpha\}} \rangle_c / pN \quad (2 - 7)$$

$\langle M^{\{\alpha\}} \rangle_c$ can be calculated approximately with a given value of X if we terminate the cluster expansion at a certain value of ℓ , so that Eq.(2-7) is the selfconsistent equation which determines X. Successive approximations are made by taking progressively larger value of ℓ at which the cluster expansion is terminated.

§ 2 - 2 1 - site approximation

Keeping only the first term in the cluster expansion given by Eq.(2-5) and taking the configurational average with a given concentration p , we get the lowest order approximation as

$$\begin{aligned} X/2 &= \langle M_1^{\{\alpha\}} \rangle_c / pN \\ &= \langle \text{Tr}_1 [S_1^Z \cdot \rho_1] \rangle_{c_1} \end{aligned} \quad (2 - 8)$$

where the ρ_1 is the approximate density ^(matrix) of the site 1 occupied by the spin S_1^Z and $\langle \dots \rangle_{c_1}$ represents the configurational average on condition that $\alpha_1 = 1$. After a short calculation, the r.h.s. of Eq.(2-8) can be written as

$$\langle \text{Tr}_1 [S_1^Z \cdot \rho_1] \rangle_{c_1} = \langle \langle \frac{1}{2} \tanh[K_1 \cdot 2 \sum^{(1)} \alpha_\nu \bar{S}_\nu + K_2 \cdot 2 \sum^{(2)} \alpha_\mu \bar{S}_\mu + L] \rangle_{\bar{S}} \rangle_{c_1} \quad (2 - 9)$$

where $K_1 = \beta J_1/2$, $K_2 = \beta J_2/2$ and $L = \beta g \mu_B H/2$, and where $\sum^{(1)}$ and $\sum^{(2)}$ mean the sums over the first- and second- neighbors of the site 1, respectively. The average over the atomic and pseudo-spin configurations, $\langle \dots \rangle_{\bar{S}}$ can be carried out in the similar way as was discussed in [I] by use of the following definitions.

$$Q_n^Z = \binom{Z}{n} p^n q^{Z-n} \quad (2 - 10)$$

and

$$D_k^n = \binom{n}{k} \left(\frac{1+X}{2}\right)^{n-k} \left(\frac{1-X}{2}\right)^k \quad (2 - 11)$$

Let z_1 and z_2 be the numbers of the first- and second-neighbours, the self-consistent equation which determines X is given by

$$X = \sum_{\nu=0}^{z_1} Q_{\nu}^{z_1} \sum_{\mu=0}^{z_2} Q_{\mu}^{z_2} \sum_{k\nu=0}^{\nu} D_{k\nu}^{\nu} \sum_{k\mu=0}^{\mu} D_{k\mu}^{\mu} \cdot \tanh[K_1(\nu-2k\nu)+K_2(\mu-2k\mu)+L]$$

(2 - 12)

where ν and μ are the numbers of magnetic atoms which occupy the first- and second-neighbors, respectively, and where $k\nu$ and $k\mu$ are the numbers of pseudo-spins which take the value $-1/2$ out of ν and μ pseudo-spins, respectively. When $K_2=0$, that is, $J_2=0$, Eq.(2-12) becomes identical with Eq.(21) obtained in [I].

§ 2 - 3 2 - site approximation

The 2-site approximation is obtained by terminating the cluster expansion at $M_2^{\{\alpha\}}$. There are $p^2 z_1 N/2$ first-neighboring spins and $p^2 z_2 N/2$ second-neighboring spins, so that the self-consistent equation is given by

$$\begin{aligned}
 X/2 &= \langle M_1^{\{\alpha\}} + M_2^{\{\alpha\}} \rangle_c / pN \\
 &= \langle \text{Tr}_1 [S_1^Z \cdot \rho_1] \rangle_{c_1} \\
 &+ \frac{pz_1}{2} \langle \text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12}] - \text{Tr}_1 [S_1^Z \rho_1] - \text{Tr}_2 [S_2^Z \rho_2] \rangle_{c_{12}} \\
 &+ \frac{pz_2}{2} \langle \text{Tr}_{13} [(S_1^Z + S_3^Z) \rho_{13}] - \text{Tr}_1 [S_1^Z \rho_1] - \text{Tr}_3 [S_3^Z \rho_3] \rangle_{c_{13}}
 \end{aligned}
 \tag{2 - 13}$$

where ρ_{12} and ρ_{13} are the approximate density matrices of the first-neighboring spins S_1^Z and S_2^Z and that of the second-neighboring spins S_1^Z and S_3^Z , respectively, and where $\langle \dots \rangle_{c_{12}}$ represents the configurational average on condition that $\alpha_1 = \alpha_2 = 1$ and similarly for $\langle \dots \rangle_{c_{13}}$. The terms containing the 1-site density matrix, ρ_1 , ρ_2 , or ρ_3 , can be calculated in the same way as in the subsection (2-2).

For a given values of pseudo-spins, the density matrix $\rho_{12, \bar{S}}$ of the first-neighboring spins is expressed by use of Eq.(2-2) with the effective Hamiltonian given by

$$\begin{aligned}
H_{12, \bar{S}} = & -2J_1 \cdot S_1^Z S_2^Z - (J_1 h_1^{(1)} + J_2 h_1^{(2)}) \cdot S_1^Z \\
& - (J_1 h_2^{(1)} + J_2 h_2^{(2)}) \cdot S_2^Z - g\mu_B H \cdot (S_1^Z + S_2^Z) \quad (2 - 14)
\end{aligned}$$

where

$$\begin{aligned}
h_1^{(1)} = & 2 \sum'_{\nu(1)} \alpha_{\nu(1)} \bar{S}_{\nu(1)} & h_1^{(2)} = & 2 \sum'_{\mu(1)} \alpha_{\mu(1)} \bar{S}_{\mu(1)} \\
h_2^{(1)} = & 2 \sum'_{\nu(2)} \alpha_{\nu(2)} \bar{S}_{\nu(2)} & h_2^{(2)} = & 2 \sum'_{\mu(2)} \alpha_{\mu(2)} \bar{S}_{\mu(2)}
\end{aligned} \quad (2 - 15)$$

where the sums over $\nu(1)$ and $\nu(2)$ are taken over the first-neighbors of the sites 1 and 2, respectively, the sites 1 and 2 being excluded, and where the sums over $\mu(1)$ and $\mu(2)$ are over the second-neighbors of the sites 1 and 2, respectively. Then the term which contains ρ_{12} is written as

$$\begin{aligned}
\langle \text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12}] \rangle_{c_{12}} &= \langle \langle \text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12, \bar{S}}] \rangle_{\bar{S}} \rangle_{c_{12}} \\
&= \langle \langle m_{J1}(h_1^{(1)}, h_1^{(2)}, h_2^{(1)}, h_2^{(2)}; g\mu_B H) \rangle_{\bar{S}} \rangle_{c_{12}}
\end{aligned} \quad (2 - 16)$$

m_{J1} defined by Eq.(2-16) can be calculated without difficulty by use of the eigenvalues of the effective Hamiltonian $H_{12, \bar{S}}$. When we take the averages, $\langle \langle \dots \rangle_{\bar{S}} \rangle_{c_{12}}$, we have to classify the neighboring sites of the sites 1 and 2 according to the lattice geometry into several types. In the case of the square

lattice (see Fig.1(1)), for example, the sites 4, 7 and 10 are the first-neighboring sites of the site 1, and the sites 4 and 10 are also the second-neighboring sites of the site 2 whereas the site 7 is not. Then the number and values of pseudo-spins at the sites 4 and 7 affect the values of $h_1^{(1)}$ and $h_2^{(2)}$ simultaneously, but those of the pseudo-spin at site 7 affects only the value of $h_1^{(1)}$. Such a situation always appears in the case of the non-Bethe lattice, since there exist triangular closed loops of the first- and second-neighbor interactions. If we abbreviate the configurational average of magnetic atoms by $\langle Q(J_1) \rangle$ and that of pseudo-spins by $\langle D(J_1) \rangle$,

$$\begin{aligned} \langle \text{Tr}_{12} [(S_1^Z + S_2^Z) \rho_{12}] \rangle_{c_{12}} \\ = \langle Q(J_1) \rangle \langle D(J_1) \rangle \cdot m_{J_1}(h_1^{(1)}, h_1^{(2)}, h_2^{(1)}, h_2^{(2)}, g\mu_B H) \quad (2 - 17) \end{aligned}$$

Using the similar notation for the term which contains ρ_{13} , we can write down the self-consistent equation as

$$\begin{aligned} X = & \sum_{\nu=0}^{Z_1} Q_{\nu}^{Z_1} \sum_{\mu=0}^{Z_2} Q_{\mu}^{Z_2} \cdot D_1(\nu, \mu) \\ & + pz_1 \left[\langle Q(J_1) \rangle \langle D(J_1) \rangle \cdot m_{J_1} - \sum_{\nu=0}^{Z_1-1} Q_{\nu}^{Z_1-1} \sum_{\mu=0}^{Z_2} Q_{\mu}^{Z_2} \cdot D_1(\nu+1, \mu) \right] \\ & + pz_2 \left[\langle Q(J_2) \rangle \langle D(J_2) \rangle \cdot m_{J_2} - \sum_{\nu=0}^{Z_1} Q_{\nu}^{Z_1} \sum_{\mu=0}^{Z_2-1} Q_{\mu}^{Z_2-1} \cdot D_1(\nu, \mu+1) \right] \end{aligned} \quad (2 - 18)$$

where

$$D_1(v, \mu) = \sum_{kv=0}^v D_{kv}^v \sum_{k\mu=0}^v D_{k\mu}^\mu \tanh[K_1(v-2kv) + K_2(\mu-2k\mu) + L]$$

(2 - 19)

The explicit expressions of $\{Q(J_1)\}$, $\{D(J_1)\}$, $\{Q(J_2)\}$ and $\{D(J_2)\}$ and values of $h_1^{(1)}$, $h_1^{(2)}$, etc. are given in the appendix for the square lattice. When $J_2=0$, the terms corresponding the second-neighboring pair cancel out, and Eq.(2-19) becomes identical with Eq. (29) obtained in [I].

§ 2 - 4 Susceptibility, Curie temperature and critical concentration

The self-consistent equations derived in the previous subsections are of the form as

$$X = f(X, p, K_1, K_2, L) \quad (2 - 20)$$

Since the equations for the susceptibility χ , the Curie temperature T_c and the critical concentration p_c are derived in the same way as in [I], we show the results without discussions. The magnetic susceptibility is given by

$$\chi/\chi_0 = \frac{\partial f}{\partial L} / \left(1 - \frac{\partial f}{\partial X}\right) \quad (2 - 21)$$

where $\chi_0 = \frac{1}{4}(g\mu_B)^2\beta \cdot pN$. The equation which determines T_c is

$$0 = 1 - \frac{\partial f}{\partial X} \Big|_{K_1=K_{c1}, K_2=K_{c2}, X=L=0} \quad (2 - 22)$$

where $K_{c1} = J_1 / 2k_B T_c$ and $K_{c2} = J_2 / 2k_B T_c$. The equation which determines p_c is

$$0 = 1 - \frac{\partial f}{\partial X} \Big|_{p=p_c, K_1=|K_2|=\infty, X=L=0} \quad (2 - 23)$$

§ 2 - 5 Spin pair correlation function and specific heat

The correlation functions of the first-neighboring spins S_1^Z and S_2^Z and of the second-neighboring spins S_1^Z and S_3^Z are defined as

$$\xi_{J1} \equiv 4 \cdot \langle S_1^Z S_2^Z \rangle_c = 4 \langle \text{Tr}[S_1^Z S_2^Z \rho_N^{\{\alpha\}}] \rangle_{c_{12}} \quad (2 - 24)$$

$$\xi_{J2} \equiv 4 \cdot \langle S_1^Z S_3^Z \rangle_c = 4 \langle \text{Tr}[S_1^Z S_3^Z \rho_N^{\{\alpha\}}] \rangle_{c_{13}} \quad (2 - 25)$$

where $\rho_N^{\{\alpha\}}$ is the exact density matrix of the system with a given atomic configuration $\{\alpha\}$. In this subsection we give the equations which determine the correlation functions by use of the approximate 1-site and 2-site density matrices obtained in the previous subsections. Before we show the calculation, it is noted that there are various ways to calculate ξ_{J1} and ξ_{J2} and we cannot determine the equations for ξ_{J1} and ξ_{J2} uniquely in the framework of our cluster approximation. This is because the correlation functions are not considered explicitly in the derivation of the self-consistent equation which determines X and there is no evident principle for the calculation of ξ_{J1} and ξ_{J2} . In the following, we calculate them by use of the 1-site density matrix in the 1-site approximation and by use of the 2-site density matrices in the 2-site approximation. It is however, possible to calculate ξ_{J1} and ξ_{J2} by expanding Eqs.(2-24) and (2-25) into a cluster series like Eq.(2-5) and terminating it at the corresponding size of

clusters to that of the self-consistent equation for X. We do not go into more details of such a method here, since the following method seems to yield the most reasonable result.

In the 1-site approximation we approximate ξ_{J1} as the correlation between the spin S_1^Z and the pseudo-spin \bar{S}_2 , that is,

$$\xi_{J1} = 4 \cdot \langle \langle \text{Tr}_1 [S_1^Z \bar{S}_2 \rho_{1, \bar{S}}] \rangle_{\bar{S}} \rangle_{c_{12}} \quad (2 - 26)$$

where the density matrix $\rho_{1, \bar{S}}$ also contains the pseudo-spin \bar{S}_2 . After a short calculation, Eq.(2-27) is written as

$$\begin{aligned} \xi_{J1} = & \sum_{\nu=0}^{Z_1-1} Q_{\nu}^{Z_1-1} \sum_{\mu=0}^{Z_2} Q_{\mu}^{Z_2} \sum_{k\nu=0}^{\nu} D_{k\nu}^{\nu} \sum_{k\mu=0}^{\mu} D_{k\mu}^{\mu} \\ & \times \left\{ \frac{1+X}{2} \cdot \tanh[K_1 + K_1(\nu - 2k\nu) + K_2(\mu - 2k\mu)] \right. \\ & \left. - \frac{1-X}{2} \cdot \tanh[-K_1 + K_1(\nu - 2k\nu) + K_2(\mu - 2k\mu)] \right\} \quad (2 - 27) \end{aligned}$$

This approximation is satisfactory in spite of the fact that is a single site one, since ξ_{J1} thus obtained does not vanish even if $T > T_c$ (where $X=0$). Moreover it becomes $\tanh K_1$ in the dilute limit ($p \rightarrow 0$). ξ_{J2} is calculated in the similar way. If $J_2=0$, ξ_{J2} is equal to X^2 which is the same result as that of the molecular field approximation.

In the 2-site approximation, ξ_{J_1} is given by

$$\begin{aligned} \xi_{J_1} &= 4 \langle \text{Tr}_{12} [S_1^Z S_2^Z \rho_{12}] \rangle_{c_{12}} \\ &= \sum Q(J_1) D(J_1) \cdot G_1(h_1^{(1)}, h_1^{(2)}, h_2^{(1)}, h_2^{(2)}) \quad (2 - 28) \end{aligned}$$

where

$$G_1(h_1^{(1)}, h_1^{(2)}, h_2^{(1)}, h_2^{(2)}) = 4 \text{Tr}_{12} [S_1^Z S_2^Z \rho_{12, \bar{S}}] \quad (2 - 29)$$

and where the same symbols are used as those employed in § 2-3. The value of ξ_{J_1} is calculated with the self-consistently determined value of X . In the dilute limit Eq.(2-28) becomes $\xi_{J_1} = \tanh K_1 \cdot \xi_{J_2}$ is calculated in the similar way by use of the density matrix ρ_{13} which is that of the second-neighboring spins S_1^Z and S_3^Z . It is noted that, even if $J_2=0$, ξ_{J_2} is not equal to X^2 since there exist the common first-neighbors which the spins 1 and 3 share (for example, the sites 7 and 10 in Fig.1(2)).

Finally the magnetic contribution to the specific heat per spin is given by

$$C_m = - \frac{p}{4} (z_1 J_1 \frac{d}{dT} \xi_{J_1} + z_2 J_2 \frac{d}{dT} \xi_{J_2}) \quad (2 - 30)$$

§ 3 Pure ferromagnet

In this section we shall consider pure ferromagnets with the first- and second-neighbor interactions; all the sites of the system are occupied by magnetic atoms. The equations for the system are given by putting $P=1$ in the equations obtained in § 2, and they are solved numerically. It is noted that our 1-site approximation is identical with that of Mamada and Takano when $J_2=0$. When the second-neighbor interaction is included there appear interactions crossing one another. Thus the problem is not solved exactly even for the two-dimensional Ising model (so-called "crossed bond" problem).

Before discussing the results we shall investigate some exactly soluble cases with anisotropic interactions in order to examine the accuracy of our approximation in § 3-1. In § 3-2 the results of our approximation for the square lattice are presented and discussed in detail. In § 3-3 the results for the triangular, s.c., b.c.c. and f.c.c. lattices are discussed.

§ 3 - 1 The accuracy of the approximation

The two-dimensional Ising model with anisotropic interactions can be solved exactly if there are no crossed bonds. In order to investigate the accuracy of our approximation we apply our approximation to such a system and compare the results with the exact solution. We consider the following two lattices. One is the well-known rectangular lattice with the interactions J_1 and J_2 along the vertical and horizontal lines, respectively. The other is the anisotropic triangular lattice with the interaction J_1 along two directions and J_2 along the other (see Fig.2).

First we consider the rectangular lattice. By varying J_2 while leaving $J_1 (>0)$ constant we go continuously from the square lattice ($J_2=J_1$) to the linear chain ($J_2=0$). The Curie temperature $T_c(R)$ depends on the value of $R=J_2/J_1$. The values of $T_c(1)$ (the square lattice) obtained in the 1- and 2-site approximation are shown in Table 1 with the exact value. The value of the 2-site approximation is better than that of the 1-site one, but it is still higher than the exact value. The dependence of the Curie temperature on R , however, is quite good. When $1 > R > 0.1$, the value of $T_c(R)/T_c(1)$ is in agreement with the exact one to within 1% in the 2-site approximation and to within 5% in the 1-site approximation. The agreement becomes worse as R decreases from 0.1, but the value of $T_c(0)$ is equal to zero as it is for the exact case.

The exact solution for the anisotropic triangular lattice

was recently discussed by Eggarter.¹⁰⁾ This system is interesting. When $J_1=J_2 > 0$, we get the triangular lattice. When $J_2=0$, we get the square lattice. When $J_2=-J_1$, the system is equivalent with the triangular antiferromagnetic Ising model and it does not show any phase transition. The values of $T_c(l)$ are shown in Table 1. We show in Fig. 2 the dependence of $T_c(R)/T_c(0)$ on R obtained in the 1- and 2-site approximations with the exact result. As can be seen from the figure, the 2-site approximation gives good dependence of $T_c(R)$ on R for $1 > R \gtrsim -0.6$. The 1-site approximation is good for $R > 0$. In both approximations $T_c(-1) = 0$ and there is no Curie temperature for $R < -1$.

From the above discussions we can conclude that the 2-site approximation shows the good dependence on $R = J_2/J_1$ except the neighborhood where the Curie temperature becomes zero. The 1-site approximation is also good for the positive R .

§ 3 - 2 Results for the square lattice

In this section we shall investigate the square Ising ferromagnet with the first- and second-neighbor interactions. As was discussed in Introduction this system has been studied by many authors. Some exact results are obtained. Kanamori¹¹⁾ has determined the ground state of the system rigorously. His conclusion is as follows. For $R > -1/2$ i.e. $J_1 > 0, J_2 > -J_1/2$, the system is in the ferromagnetic state. For $R < -1/2$ i.e. $J_1 > 0, J_2 < -J_1/2$, the system is in the antiferromagnetic state. Its structure is that all spins in a row have the same signs and the spins in the adjacent rows have the opposite signs. On the boundary $R = -1/2$ the ground state of the system is degenerate with a macroscopic degree of degeneracy and there would be no preferred state at low temperatures. Fan and Wu⁵⁾ concluded that the transition temperature vanishes at $R = -1/2$. This phenomenon is quite alike with the vanishing of the transition temperature for the antiferromagnetic triangular Ising model.

We first consider the Curie temperature. In Fig. 3 the dependence of $T_c(R)/T_c(0)$ on $R = J_2/J_1$ obtained in the 1- and 2-site approximations is shown with the results of the series expansion by Dalton and Wood.²⁾ The absolute values of $T_c(0)$ and $T_c(1)$ are listed in Table 1. For positive R the Curie temperature vs. R curve of the 2-site approximation is very near to the straight line although it shows a slight curvature. The curve of the 1-site approximation shows a larger curvature than

that of the 2-site one. As Dalton and Wood discussed, the variation of the Curie temperature is represented to a good approximation by

$$T_c(R) = T_c(0) \{1 + \gamma R\} \quad \text{for } 0 < R < 1 \quad (3 - 1)$$

The value of $\gamma = 1.35$ reproduces the result of Dalton and Wood to within 2%. In the 2-site approximation the value of $\gamma = 1.30$ reproduces the result in the same accuracy. As the value of R decreases from 0, the Curie temperature obtained in the 2-site approximation smoothly decreases and vanishes at the exact value of $R = -1/2$. Fan and Wu⁵⁾ obtained the similar behavior of the Curie temperature in the free-fermion approximation. This result is reasonable since the exact Curie temperature for the anisotropic triangular lattice discussed in § 3-1 decreases continuously for negative R and vanishes at $R = -1$ where the system becomes equivalent with the antiferromagnetic triangular lattice. On the other hand, in the 1-site approximation we have two solutions for $0 > R > -0.31$ and no solution for $R < -0.31$. The similar behavior was obtained in the Bethe approximation and the free-fermion approximation derived by Kawakami and Osawa³⁾ and Gibberd⁴⁾.

In order to investigate the properties of the system for $T < T_c$ we calculated the spontaneous magnetization, pair correlation functions and specific heat in the 2-site approximation. For $R > 0$ the behavior of the spontaneous magnetization and specific heat is similar to that for $R = 0$. $X = 2 \langle S^z \rangle$ decreases

monotonically as the temperature increases and the value of d^2X/dT^2 is always negative. The specific heat increases monotonically as the temperature increases and has the gap at $T=T_c$. The shape of the C_m vs. T curve is similar to that obtained in the Bethe approximation. For $R < 0$, however, the behavior of the system is rather complicated. We show the temperature dependence of X , pair correlation function ξ_{J_1} and ξ_{J_2} and the specific heat for various values of negative R in Fig. 4, 5 and 6, respectively.

As the value of R decreases, the shape of the X vs. T curve changes continuously. There appears below T_c the region of T where the X decrease quite rapidly. For $R < -0.38$ we have three non-trivial solutions for X . If a system shows the first-order phase transition, we often encounter such a behavior of the order parameter. So it is interesting to investigate the behavior of the free energy. In general the free energy minimum determines the value of the order parameter. Thus it is reasonable to assume that the free energy $G(X)$ satisfies the following equation if there is no external field.

$$\frac{\partial G(X)}{\partial(X)} = X - f(X) \quad (3 - 2)$$

where $f(X)$ is given by Eq.(2-20). Then integrating this equation, we can obtain the free energy by

$$G(X) - G(0) = \int_0^X [X - f(X)] dx \quad (3 - 3)$$

The function $G(0)$ is a function of temperature and we cannot determine its absolute value in this discussion. The free energy thus obtained gives one minimum together with one maximum at $X=0$ when $R \gtrsim -0.38$ and $T < T_c$. For $-0.5 < R \lesssim -0.4$ there appears the region of T where two minima and one maximum together with the maximum at $X=0$. From the behavior of the free energy calculated for various temperatures we expect the first-order phase transition. The temperature where the first-order phase transition occurs are always lower than the Curie temperature obtained by Eq.(2-22) and are denoted by the dashed line in Fig. 4. Then the Curie temperature vs. R curve shown in Fig. 3 has its meaning as the second-order phase transition even when the first-order phase transition also occurs. When $R = -0.5$, the solution of $X=0$ has always the lowest energy.

The results of the pair correlation function for the first-neighboring spins, ξ_{J_1} and for the second-neighboring spins, ξ_{J_2} are shown in Fig. 5. As can be seen from the figure, ξ_{J_2} decreases more rapidly than ξ_{J_1} for small positive R and $R \leq 0$. The value of ξ_{J_2} is positive near the Curie temperature even if R is negative. As the temperature increases from T_c , ξ_{J_2} for negative R decreases and becomes negative. Such behavior is reasonable since there remains the ferromagnetic short range order near the Curie temperature. The specific heat C_m exhibits the interesting behavior for negative R as can be seen from Fig. 6. The C_m v.s. T curve for $R = -0.2$ has a slight shoulder below the peak corresponding to the phase tran-

sition. As the value of R decreases from -0.2 , this shoulder grows up to a peak which is much more conspicuous than that at $T=T_c$.

The rather curious behavior of the square lattice is qualitatively accounted for as follows. For $R > -0.5$ the ground state is ferromagnetic and all spins have positive (or negative) signs. We shall consider the energy required to flip some positive spins. It requires the energy, $4J_1 - 4|J_2|$, to flip a single spin. The energies required to flip one more spin are $2J_1 - 4|J_2|$ and $4J_1 - 2|J_2|$ at one of first-neighboring sites and a second-neighboring sites, respectively. Thus it is preferable to flip the first-neighboring spin. Consider the case where a pair of spins at the sites 1 and 2 in Fig. 1(1) have negative signs and they forms an isolated island in the sea of positive spins. In Fig.1(1) the energies required to flip one more spin at the sites 5, 6 and 8 are $E_5 = 2J_1 - 2|J_2|$, $E_6 = 4J_1 - 2J_2$ and $E_8 = 2J_1 - 4|J_2|$, respectively. The energy E_8 is the smallest if $|J_2|$ is finite. When $|R|$ is small, the difference between E_5 and E_8 is small and the excitation spectrum is similar to that for $R=0$. Thus for small $|R|$ the thermodynamic behavior is similar to that for $R=0$. On the other hand, when $|R|$ becomes large, the difference between E_5 and E_8 increases and E_8 becomes small. At $R=-0.5$, E_8 is equal to zero. In this case it is quite preferable to flip the spin at the site 8 and the negative spin cluster tend to

spread out into a chain. Crudely speaking, as temperature reaches the value $k_B T^* \sim E_g$, the negative spin clusters rapidly spread out into a chain. This fact results in the rapid decrease of the spontaneous magnetization and a peak of the specific heat around this temperature. As can be seen from figures, a crude estimate $k_B T^*/J_1 \sim 2 - 4|R|$ gives a reasonable value for $R < -0.3$.

Finally we shall give some comments on the 1-site approximation. Takase ⁶⁾ has studied the square lattice in the approximation which is identical with our 1-site one. His derivation of the approximation is based upon the different principle from ours. He calculated the spontaneous magnetization and the free energy which is defined in the same way as ours. He concluded that for the range $-0.238 > R \gtrsim -0.5$ the first-order phase transition occurs instead of the second-order phase transition. This conclusion contradicts our results. We think it is not appropriate to apply the 1-site approximation to the case for negative R . The reasons are (1) the closed loops consisting of the links of the interacting spins are not considered and (2) the short range correlation are not considered appropriately. When we calculate ξ_{J_2} by the method discussed in § 2-5, the value of ξ_{J_2} becomes negative at $T=T_c$ even for the very small $|R|$. For $R \geq 0$, however, the 1-site approximation gives reasonable results.

§ 3 - 3 Results for other lattices

We calculate the Curie temperature for various value of R and various lattice in the 2-site approximation. The calculation is carried out for the triangular, s.c., b.c.c. and f.c.c. lattices. The values of $T_c(0)$ and $T_c(1)$ are listed in Table 1. As was discussed in [I], our approximation is more accurate for the three-dimensional lattices than for the two-dimensional lattices. The dependence of $T_c(R)/T_c(0)$ on R is shown in Fig.7. For positive R every curve is very near to the straight lines although it shows a slight curvature. For this region of R the 1-site approximation gives the similar results.

The ground states of these lattices have been determined rigorously by Kanamori¹¹⁾ and Kaburagi and Kanamori¹²⁾. The condition that the system is in the ferromagnetic state is as follows; $R > -1/2$ for the triangular lattice; $R > -1/4$ for the s.c. lattice; $R > -2/3$ for the b.c.c. lattice; $R > -1$ for the f.c.c. lattice. The boundaries R^* where the ferromagnetic ground state disappear are denoted in Fig. 7 by the vertical dashed lines. The Curie temperature for the triangular lattice vanishes at the exact boundary i.e. $R^* = -1/2$. For the three dimensional lattices, however, the Curie temperature remains finite below the exact boundaries. The Curie temperatures for the b.c.c. and f.c.c. lattices exhibit complicated behavior for $R < R^*$. As was discussed in § 3-1 our two-site approximation has poor reliance for R near the boundary where the ferromagnetic ground state disappear. Thus in order to investigate the region of R near R^*

more correctly we have to take into consideration three-site clusters or much larger clusters than three sites.

§ 4 Dilute ferromagnet

In the present section we investigate the effects of non-magnetic atoms on the thermodynamic properties of the Ising model. As is well known the presence of non-magnetic atoms results in the decrease of the Curie temperature and below a critical concentration p_c of magnetic atoms the system does not show any phase transition. We calculate the critical concentration, Curie temperature, paramagnetic susceptibility, spontaneous magnetization, pair correlation functions and specific heat. The calculation is carried out for various values of $R = J_2/J_1$.

In § 4-1 we investigate the thermodynamic properties in our 2-site approximation. As was discussed in the previous paper [1] the geometry of the lattice is taken into consideration in more detail than the coordination number. The calculation is carried out for the square lattice. For other lattices we have not carried out the calculations in the 2-site approximation except the critical concentrations for $R=0$ and 1 , since it requires enormous computer time. In §4-2 we show that our 1-site approximation gives qualitatively similar results to those of the 2-site approximation. For the s.c., b.c.c. and f.c.c. lattices the calculation is carried out in the 1-site approximation.

§ 4 - 1 Results for the square lattice— 2-site approximation

(a) Critical concentration

Numerical value of the critical concentration p_c is calculated by Eq.(2-23). We first investigate the dependence of p_c on the strength of the second-neighbor interaction. In Table 2 we show the numerical values of p_c obtained for various values of $R = J_2/J_1$. For $R > 0$ the value of p_c decreases from that for $R=0$. This decrease occurs discontinuously as R increases from zero. The value of p_c is equal to 0.3108 and independent of the value of R for $0 < R < 0.2$. For $0.2 < R < 1$ the value shows a slight dependence on R . The changes of p_c occur discontinuously when the values of R reach some rational numbers. We think, however, such a dependence of p_c on R (> 0) is due to the approximation and the value of p_c should be independent of the value of R for $R > 0$. The reason is as follows. It is self-evident that the system does not show any phase transition if there exist only spin clusters of finite sizes. As was discussed in [1], the critical concentration of the ferromagnetic Ising model should be correspond to the appearance of the infinite network of adjacent spins. To form the network of adjacent spins it does matter whether the interaction exists or not and the strength of the interaction does not have any concern. Namely the critical concentration is a geometrical property of the lattice and its value depends on the range of interactions and not on the strength. This fact has been discussed by Elliott, Heap, Morgan and Rushbrook⁸⁾ based on the concentration expansion of the suscep-

tibility. Idogaki and Uryu⁹⁾ also obtained the results that the value of p_c is independent of the value of R by the concentration expansion method.

The numerical values of the critical concentration at $R=0$ and 1 are also calculated for the triangular, s.c., b.c.c. and f.c.c. lattices in the 2-site approximation. They are listed in Table 3 with the results obtained by Idogaki and Uryu and the critical concentrations obtained in the site-percolation problem.¹³⁾ For the sake of comparison the values of $2/z_1$ and $2/(z_1+z_2)$ are also listed. Comparing our results with those of the percolation, the values show good dependence on the geometry of the lattices although they are somewhat smaller than those of the percolation. For the three-dimensional lattices the difference is about 10% for both $R=0$ and $R=1$. The simplest estimation of the critical concentration, $2/z_1$ gives satisfactory results for $R=0$, but the estimation $2/(z_1+z_2)$ for the case of $R=1$ is not good. For $R<0$ we obtained the critical concentration which increases as R decreases for the square lattice. As will be discussed in the following, these values have little reliance.

(b) Curie temperature and paramagnetic susceptibility

The Curie temperature is calculated by Eq.(2-22). It depends on values of p and R . We calculate the Curie temperature for the square lattice in the 2-site approximation. The concentration dependence of the normalized Curie temperature for various values of R is shown in Fig.8(a). In Fig.8(b) we show the magnification of a low temperature part. For p near 1, the Curie temperature decreases linearly as p decreases. The slope of the $T_c(R, p)$ vs. p curve monotonically increases as the value of R decreases. The numerical values of $d[T_c(R, p)/T_c(R, 1)]/dp$ at $p=1$ are 1.167, 1.203, 1.259 and 1.285 for $R=1.0, 0.5, 0.1$ and 0.0 , respectively. They are 1.330, 1.429, 1.664 and 2.513 for $R=-0.1, -0.2, -0.3$ and -0.4 , respectively. This result shows us that the positive second-neighbor interaction relieves the influence of non-magnetic atoms.

As the concentration decreases, the Curie temperature decreases monotonically. For $R=0$ and $R \geq 1/2$, the value of $dT_c(R, p)/dp$ increases monotonically and the Curie temperature drops abruptly at $p=p_c$. For the small value of $R (>0)$, the Curie temperature vs. p curve shows a considerable decrease near the critical concentration for $R=0$ and then the curve has a "trail". The region of temperature where the curve shows the trail becomes lower as the value of R decreases. The second-neighbor interaction is indispensable for occurrence of the phase transition in the region of concentration where the Curie temperature has the trail. This behavior of the Curie temperature

has been predicted by Brout. Idogaki and Uryu obtained the same results.

For negative R , the Curie temperature shows complicated behavior in the low temperature region. For $R=-0.05$ and $0.46 > p > 0.456$ for example there are three transition points. As temperature decreases, the system shows a phase transition at the highest Curie temperature and becomes ferromagnetic. Then it becomes paramagnetic at the next Curie temperature. At the lowest Curie temperature the system becomes ferromagnetic again. For $0.456 > p > 0.451$, the system shows two phase transition. The phenomenon like this is obtained exactly by Kasai and Syozi for the annealed Ising model of a mixture of ferromagnetic and antiferromagnetic bonds. In the present problem, however, we cannot conclude that this is what happens actually. The reason is as follows. In our approximation we assume that the ordered state, if it appears, is the ferromagnetic one. In other words we assume that the system is described by only one order parameter X . On the other hand, when p is less than 1, there exist clusters of spins which are connected to their neighboring clusters only by the second-neighbor interactions. If J_2 is negative, the spins in such clusters take opposite signs from the signs of their neighboring clusters at low temperatures. Thus the ground state should not be the simple ferromagnetic one and our approximation becomes worse in the low temperature region. In the high temperature region, however, our approximation is reasonable since the strong ferromagnetic interaction J_1 is dominant in this region.

The paramagnetic susceptibility are calculated for various values of R and p . In Figs. 9 and 10 we show the inverse susceptibility per spin for $R=0.0$ and -0.1 . For $R>0$ the behavior of the susceptibility is similar to that for $R=0.0$. In the high temperature region, the inverse susceptibility curve becomes a straight line and obeys the Curie-Weiss law. In the dilute limit ($p \rightarrow 0$), it obeys the Curie law. As temperature approaches the Curie temperature the inverse susceptibility curve deviates from the straight line. This deviation becomes large as p approaches p_c . The curve for $p=0.4$ in Fig. 9 shows appreciable deviation from the straight line in the low temperature region. ^(In Fig. 10) the curve for $p=0.4$ shows little temperature dependence for $k_B T/J_1 \lesssim 0.3$

(c) Spontaneous magnetization and specific heat

In this section we investigate the effect of the weak second-neighbor interaction $J_2 > 0$ on the spontaneous magnetization, pair correlation function and specific heat.

The spontaneous magnetization at $T=0$ decreases slowly as p decreases from 1 and it falls to zero rapidly at $p=p_c$. We calculate the temperature dependence of $X = 2 \langle S^z \rangle_c$ for various values of p . In the case of $R=0.0$, as the temperature increases, X decreases quite slowly in the low temperature region and the decrease becomes rapid for T near T_c . In the case of small $R(>0)$, the X vs. temperature curve shows an appreciable decrease

in the low temperature region for some values of p . The result for $R=0.05$ is shown in Fig.11. The decrease of X in the low temperature region occurs near $k_B T \sim J_2$ and is conspicuous for $0.6 > p > 0.4$ and for small values of R . This behavior of X is explained as follows. When p is less than 1, there appears a spin cluster which is connected to its neighboring spins only by the second-neighbor interaction. The magnetization of this cluster starts to decrease rapidly as the value of $k_B T$ approaches J_2 . The probability of finding such clusters increases as p decreases. Thus the behavior of X under consideration becomes conspicuous for $p \lesssim 0.6$. On the other hand if p is near the critical concentration, the second-neighbor interaction plays an essential role on the formation of the long range order. Then as the temperature approaches to $k_B T \sim J_2$, X decreases directly to zero. This fact is easily examined in our approximation by calculating the value of $X^{(n,m)}$, where $X^{(n,m)}$ is defined as the averaged value of a spin on condition that the spin has n and m magnetic atoms in its first- and second-neighbors, respectively. The equation for $X^{(n,m)}$ is obtained without difficulties. (cf. Eq.(38), (39) and (40) in [I]).

The specific heat of the system is calculated by Eq.(2-30). We first consider the paramagnetic state which appears for $T > T_c$. When $p < p_c$, the value of T_c is equal to zero. The paramagnetic state gives the specific heat due to spin orientation which is of the Schotlky type. For $R=0.0$, the curve has one maximum. For $R=0.05$ and 0.1 , the curve has two maxima, one of which

corresponds to the second-neighbor interaction. The position of the maximum corresponding to J_1 is $k_B T/J_1 \sim 0.5$ and shows little dependence on R and p . The position of the maximum corresponding to J_2 shows dependence on R and it is about $k_B T/J_1 \sim 0.025$ for $R=0.05$ and 0.05 for $R=0.1$.

The temperature dependence of the specific heat for $R=0.0$ is shown in Fig.12. The specific heat has a peak at $T=T_c$ for every concentration. In the region of temperature $0 < T < T_c(R,p)$, the specific heat increases monotonically as the temperature increases. The appearance of the Schottky-type maximum in the curve depends on whether the Curie temperature lies above or below the maximum. For $k_B T_c/J_1 \lesssim 0.5$, we can see the Schottky-type maximum. The curve has one or two peaks depending on whether the Curie temperature lies above or below the Schottky-type maximum.

For $R=0.05$ and 0.1 , the specific heat curves are more complicated. They are shown in Figs. 13 and 14. When p is near 1, the curve is in the similar shape as the case of $R=0.0$. As p approaches to 0.6, the specific heat in the low temperature region increases. In the case of $R=0.05$ the specific heat has one more peak in the low temperature region for $0.4 < p \lesssim 0.5$ together with the peak at $T=T_c$. For $p \lesssim 0.4$ the specific heat has only one peak below T_c . The new peak is also seen in the case of $R=0.1$ but it is less notable than that for $R=0.05$. This new peak has its maximum value about at $T \sim T_c \cdot z_2 J_2 / z_1 J_1$. The position of the maximum shows a slight concentration dependence

and it lowers as p decreases. The peak of the specific heat curve in the low temperature region corresponds to a rapid decrease of the spin pair correlation of the second-neighboring spins. This fact is ascertained by calculating the pair correlation function ξ_{J_1} and ξ_{J_2} . The result shows ξ_{J_2} start to decrease rapidly near the temperature corresponding the peak of the specific heat curve, whereas ξ_{J_1} shows only a slight decrease.

§ 4 - 2 Results for three-dimensional lattices— 1-site approximation

In this subsection we investigate the properties of the s.c., b.c.c. and f.c.c. lattices in our 1-site approximation. As was discussed in [I], the 1-site approximation gives qualitatively good results for the Ising model with the nearest-neighbor interaction only. In this case the approximation is identical with that given by Mamada and Takano. In order to examine the accuracy of the 1-site approximation for the case including the second-neighbor interaction, we compare the results for the square lattice with those obtained in the 2-site approximation.

The 1-site approximation reproduces characteristic properties of the system for $R > 0$. The Curie temperature shows a trail in the low temperature region when R is small. The deviation of the inverse susceptibility from the Curie-Weiss law near the Curie temperature becomes large, as p approaches p_c . The specific heat shows one more maximum in the low temperature region together with the peak at $T = T_c$. The specific heat of the paramagnetic state has two Schottky-type maximum for $R > 0$. The numerical values of the critical concentration for various values of R is shown in Table 2. For $0 < R < 1/3$, the value $p = 0.2452$ is independent of the value of R . The decrease of p_c occurs discontinuously at $R = 1/3, 1/2$ and $2/3$, although it is quite small. We think that this decrease does not have any meaning actually. It is due to the approximation. For $R < 0$, the 1-site approxi-

mation does not give reasonable results even if $p=1$; it is discussed in §2. From the results discussed above we conclude that our 1-site approximation gives fairly good description of the dilute Ising model for $R \geq 0$. It is noted that Matsudaira et. al.¹⁴⁾ has calculated the critical concentration and Curie temperature for the square lattice in the approximation identical with our 1-site approximation. His approximation, however, is based on the different principle from ours.

We have calculated the critical concentration, Curie temperature, paramagnetic susceptibility, spontaneous magnetization and specific heat for the s.c., b.c.c. and f.c.c. lattices. Numerical values of p_c for $R=0$ and 1 are listed in Table 3. They are considerably small in the cases of $R=1$ and of the f.c.c. lattice. They do not show the dependence on the geometry of lattices in more detail than the coordination number z_1 and z_2 . The reason is that the effect of the closed loop consisting of links of the interacting spin pairs is not taken into consideration at all in the 1-site approximation. The behavior of the Curie temperature is, however, reasonable. The "trail of the Curie temperature for p near critical concentration appears for every lattice when R is small. As an example, we show the results for the s.c. lattice in Fig. 15. In these lattices we obtain the results, which are similar to those for the square lattice, for the susceptibility, temperature dependence of the spontaneous magnetization and specific heat. The rapid decrease of $X = 2 \langle S^z \rangle_c$ and a peak of the specific heat corres-

ponding to the rapid decrease of the correlation between second-
neighboring spin pairs are remarkable when the value of R is
small. For a fixed value of R , these behavior is more remarkable
for the f.c.c. lattice ($z_2/z_1 = 6/12$) than the b.c.c. ($z_2/z_1 = 6/8$)
and s.c. ($z_2/z_1 = 12/6$) lattices.

§ 5 Concluding Remarks

We have investigated the thermodynamic properties of the pure and dilute Ising spin system including the second-neighbor interaction in our cluster approximation.

In § 3 the pure system (including no non-magnetic atoms) has investigated. The Curie temperature calculated in the 2-site approximation shows reasonable dependence on $R = J_2/J_1$. For $R > 0$ the dependence of the normalized Curie temperature, $T_c(R)/T_c(R=0)$ on R agrees well with the series-expansion result of Dalton and Wood. For $R < 0$ the Curie temperature decreases smoothly with decreasing R . It vanishes at the boundary where the ground state ceases to be ferromagnetic in the cases of the square and triangular lattices, whereas it remains finite at the boundary for the three-dimensional lattices. We have discussed the thermodynamic properties of the square lattice in detail. For $-0.3 < R < -0.38$, there appears the region of temperature near $k_B T \sim 2J_1 - 4|J_2|$ where the spontaneous magnetization shows a rapid decrease and the specific heat has a peak or a shoulder in addition to a peak at $T = T_c$. For $-0.5 < R < -0.4$, the system shows the first-order phase transition at the lower temperature than T_c .

In § 4 we have investigated the role of the second-neighbor interaction in the dilute case. Main results are as follows. The critical concentration depends only on the range of the interactions if $J_2 > 0$. A slight dependence of P_c on R obtained in our theory may be due to the approximation. When R is

positive and small, the second-neighbor interaction plays an essential role in the low temperature region and we find interesting phenomena. The Curie temperature vs. concentration curve has a trail near the critical concentration. In the region of temperature near $k_B T \sim J_2$, the spontaneous magnetization and pair-correlation function of the second-neighboring spins show an apparant decrease and correspondingly the specific heat has a peak. These phenomena are remarkable when the concentration is around the critical concentration for the case of $R=0$.

Our approximation can be applicable to the Heisenberg spin system with the second-neighbor interaction without difficulties. If we carry out the calculation, we would obtain results that are different from those for the Ising spin system in some respects. We expect, however, a trail of the Curie temperature and a peak of the specific heat due to the weak second-neighbor interaction will be found in the Heisenberg spin system. The reason is that these phenomena are the results of the appearance of the spin clusters which interact to the neighboring spins mainly with the second-neighbor interactions.

Appendix Explicit expressions of $\{Q(J_1)\}[D(J_1) \cdot m_{J_1}]$ and $\{Q(J_2)\}[D(J_2) \cdot m_{J_2}]$

We first give the expression of $\{Q(J_1)\}[D(J_1) \cdot m_{J_1}]$ for the square lattice. The cluster of the first-neighboring spins S_1^Z and S_2^Z is shown in Fig.1(1) and it has ten neighbors to be considered. When we take the configurational averages of magnetic atoms and pseudo-spins, it is convenient to classify the ten neighbors into six sets of sites, a,b,c,d,e and f in the following way

$$\begin{aligned} a &= \{ 7 \} & b &= \{ 3, 9 \} & c &= \{ 4, 10 \} \\ d &= \{ 5, 11 \} & e &= \{ 6, 12 \} & f &= \{ 8 \} \end{aligned} \quad (A - 1)$$

where the numerals represent the neighboring sites of the cluster shown in Fig.1(1).

Letting a number v_i ($i=a, \dots, f$) be the number of magnetic atoms which occupy the set of sites i , each configuration of magnetic atoms can be denoted by a set, $\{v\} = \{v_a, v_b, v_c, v_d, v_e, v_f\}$. The number v_i takes the values 0 and 1 if $i=a$ or f and it takes the values 0, 1 and 2 if $i=b, c, d$ or e . The probability of the configuration $\{v_i\}$ is written as

$$Q_{\{v\}} = Q_{va}^1 Q_{vb}^2 Q_{vc}^2 Q_{vd}^2 Q_{ve}^2 Q_{vf}^1 \quad (A - 2)$$

where we use the notation defined by Eq.(2-10).

With a given configuration of atoms $\{v\}$, each configuration of the pseudo-spins can be denoted by a set, $\{k\} = \{k_a, k_b, k_c, k_d, k_e, k_f\}$

The number k_i ($i=a, \dots, f$) is the number of pseudo-spins which take the value $-1/2$ out of v_i pseudo-spins. The probability of the configuration $\{k\}$ is written as

$$D_{\{v\},\{k\}} = D_{ka}^{va} D_{kb}^{vb} D_{kc}^{vc} D_{kd}^{vd} D_{ke}^{ve} D_{kf}^{vf} \quad (\text{A} - 3)$$

where we use the notation defined by Eq.(2-11).

For each configuration denoted by $\{v\}$ and $\{k\}$, the values of $h_1^{(1)}$, $h_1^{(2)}$, $h_2^{(1)}$ and $h_2^{(2)}$ given by Eq.(2-15) are as follows.

$$\begin{aligned} h_1^{(1)} &= 2 \sum_{v(1)}' \alpha_{v(1)} \bar{S}_{v(1)} \\ &= 2 \{ \alpha_7 \bar{S}_7 + (\alpha_4 \bar{S}_4 + \alpha_{10} \bar{S}_{10}) \} \\ &= (v_a - 2k_a) + (v_c - 2k_c) \end{aligned} \quad (\text{A} - 4)$$

similarly

$$h_1^{(2)} = (v_b - 2k_b) + (v_d - 2k_d) \quad (\text{A} - 5)$$

$$h_2^{(1)} = (v_d - 2k_d) + (v_f - 2k_f) \quad (\text{A} - 6)$$

$$h_2^{(2)} = (v_c - 2k_c) + (v_e - 2k_e) \quad (\text{A} - 7)$$

Thus summing over all the possible configurations $\{v\}$ and $\{k\}$. we obtain the following expression.

$$\begin{aligned} & \sum Q(J_1) \sum D(J_1) \cdot m_{J_1} \\ &= \sum_{\{v\}} Q_{\{v\}} \cdot \sum_{\{k\}} D_{\{v\},\{k\}} \\ & \times m_{J_1} [v_a + v_c - 2(k_a + k_c), v_b + v_d - 2(k_b + k_d), \\ & \quad v_d + v_f - 2(k_d + k_f), v_c + v_e - 2(k_c + k_e)]; g\mu_B H] \end{aligned} \quad (\text{A} - 8)$$

In the similar way we can obtain the expression of $\{Q(J_2)\}D(J_2) \cdot m_{J_2}$ for the cluster of the second-neighboring spins S_1^Z and S_3^Z shown in Fig.1(2).

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Table 1. Numerical values of $k_B T_c / J_1$
for $R=0$ ($J_2=0$) and $R=1$ ($J_1=J_2$)

	1-site	2-site	Dalton & Wood	Bethe
[For $R=0$]				
square	1.545	1.466	1.132	1.443
triangle	2.537	2.307	1.823	2.466
s.c.	2.537	2.479	2.267	2.466
b.c.c.	3.530	3.484	3.179	3.476
f.c.c.	5.522	5.327	4.897	5.485
[For $R=1$]				
square	3.530	3.299	2.629	3.476
triangle	5.522	5.285	4.386	5.485
s.c.	8.516	8.312	7.750	8.490
b.c.c.	6.520	6.312	5.815	6.487
f.c.c.	8.516	8.312	7.748	8.490

Table 2 Numerical values of p_c for the square lattice
with various values of $R = J_2/J_1$

$R = J_2/J_1$	2-site	1-site
1.0	0.2854	0.2225
0.9	0.3058	0.2225
0.8	0.3058	0.2225
0.7	0.3064	0.2225
0.6	0.3100	0.2228
0.5	0.3152	0.2310
0.4	0.3155	0.2406
0.3	0.3114	0.2452
0.2	0.3108	0.2452
0.1	0.3108	0.2452
0.0	0.4309	0.4284

Table 3 Numerical values of p_c for $R=0$ ($J_2=0$)
and $R=1$ ($J_1=J_2$)

	1-site	2-site	Idogaki and Uryu	2/z	percolation
[For $R=0$]					
square	0.428	0.431	0.49	0.5	0.59
triangle	0.293	0.377		0.333	0.5 (exact)
s.c.	0.293	0.283	0.26	0.333	0.307
b.c.c.	0.223	0.211	0.22	0.25	0.243
f.c.c.	0.150	0.180		0.167	0.199
[For $R=1$]					
square	0.223	0.285	0.25	0.25	0.410
triangle	0.150	0.193		0.167	0.295
s.c.	0.101	0.122	0.10	0.111	0.137
b.c.c.	0.129	0.151	0.13	0.143	0.175
f.c.c.	0.101	0.122		0.111	0.136

Figure Captions

- Fig. 1 Figures of the 2-site clusters for the square lattice. (1) is for the first-neighboring pair and (2) is for the second-neighboring pair. White circles represent the sites in clusters and black circles represent the neighboring sites. The first- and second-neighbor interactions are represented by the straight and dashed lines, respectively.
- Fig. 2 Dependence of the normalized Curie temperature $T_c(R)/T_c(0)$ on R for the anisotropic triangular lattice. Full lines denoted by 1 and 2 are obtained in the 1- and 2-site approximations. Dashed line is the exact solution. The lattice structure is also shown.
- Fig. 3 Dependence of the normalized Curie temperature $T_c(R)/T_c(0)$ on R for the square lattice obtained in the 1- and 2-site approximations.
- Fig. 4 Temperature dependence of $X = 2\langle S^z \rangle$ for several values of negative R . Result of the 2-site approximation for the square lattice. Dashed line shows the position where the first-order phase transition occurs for each R .

- Fig. 5 Temperature dependence of spin pair correlation functions, ξ_{J1} and ξ_{J2} for several values of R. Result of the 2-site approximation for the square lattice.
- Fig. 6 Temperature dependence of specific heat for several values of R obtained in the 2-site approximation. Result for the square lattice.
- Fig. 7 Dependence of the normalized Curie temperature $T_c(R)/T_c(0)$ on R for the triangular, s.c, b.c.c. and f.c.c. lattices obtained in the 2-site approximation. Each dashed line shows the rigorously determined boundary where the ferromagnetic ground state disappears.
- Fig. 8(a) Dependence of the normalized Curie temperature $T_c(R,p)/T_c(R,1)$ on the concentration p for various values of R. Results of the 2-site approximation for the square lattice.
- Fig. 8(b) Magnification of a part of the low temperature region.
- Fig. 9 Inverse paramagnetic susceptibility for several values of p in the case of R=0. Result of the 2-site approximation for the square lattice.
- Fig.10 Inverse paramagnetic susceptibility for several values of p in the case of R=-0.1. Result of the 2-site approximation for the square lattice.

- Fig. 11 Temperature dependence of $X = 2\langle S^z \rangle_c$ for several values of p . Result of the 2-site approximation for the square lattice. The value of R is fixed to 0.05.
- Fig. 12 Temperature dependence of specific heat for several values of p in the case of $R=0$. Result of the 2-site approximation for the square lattice.
- Fig. 13 Temperature dependence of specific heat for several values of p in the case of $R=0.05$. Result of the 2-site approximation for the square lattice.
- Fig. 14 Temperature dependence of specific heat for several values of p in the case of $R=0.1$. Result of the 2-site approximation for the square lattice.
- Fig. 15 Dependence of the normalized Curie temperature $T_c(R,p)/T_c(R,1)$ on the concentration p for several values of R . Result of the 1-site approximation for the s.c. lattice.

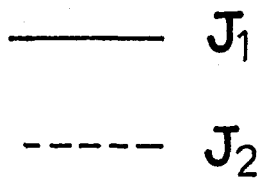
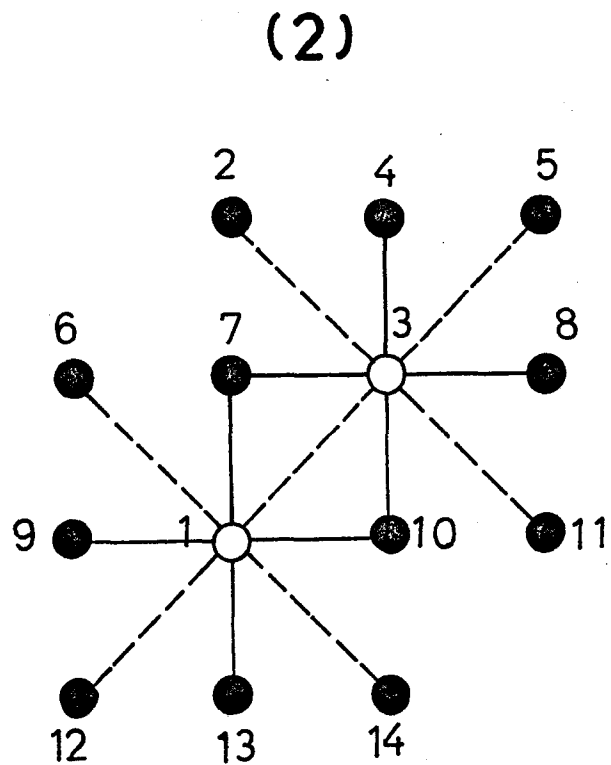
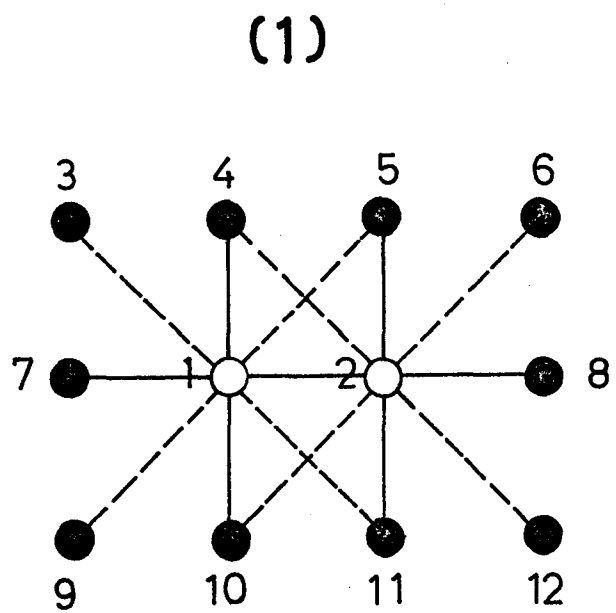
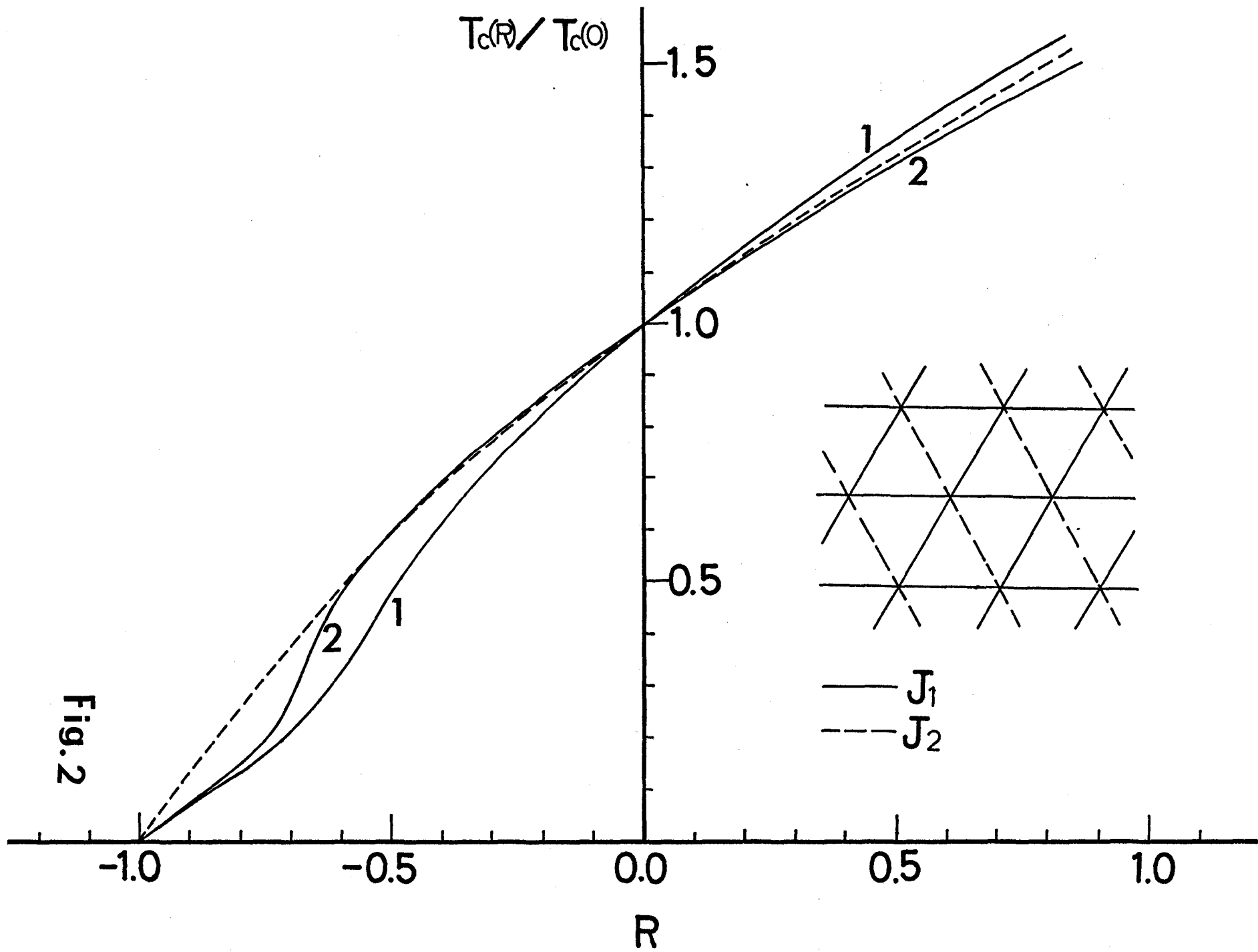


Fig. 1



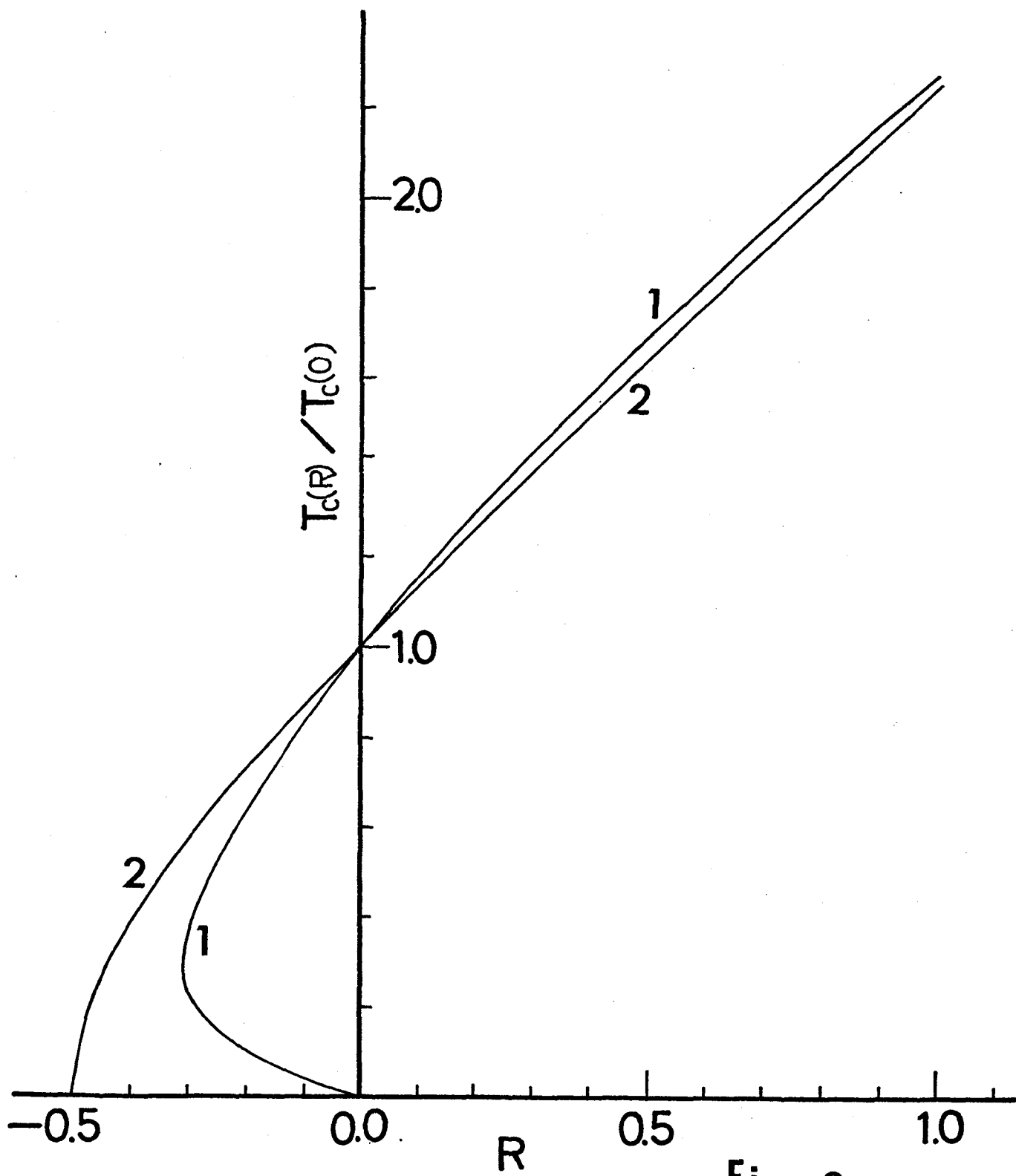


Fig. 3

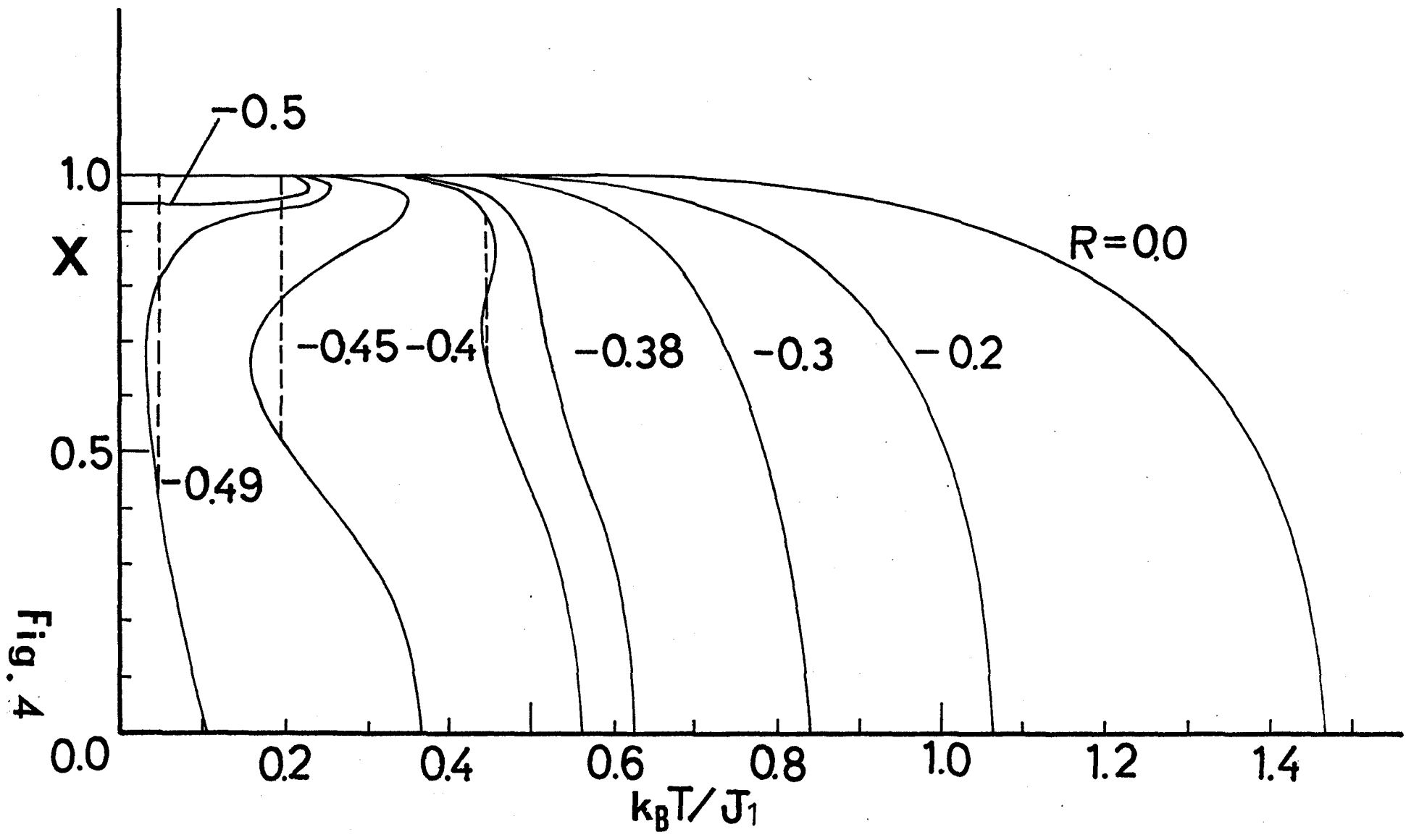


Fig. 4

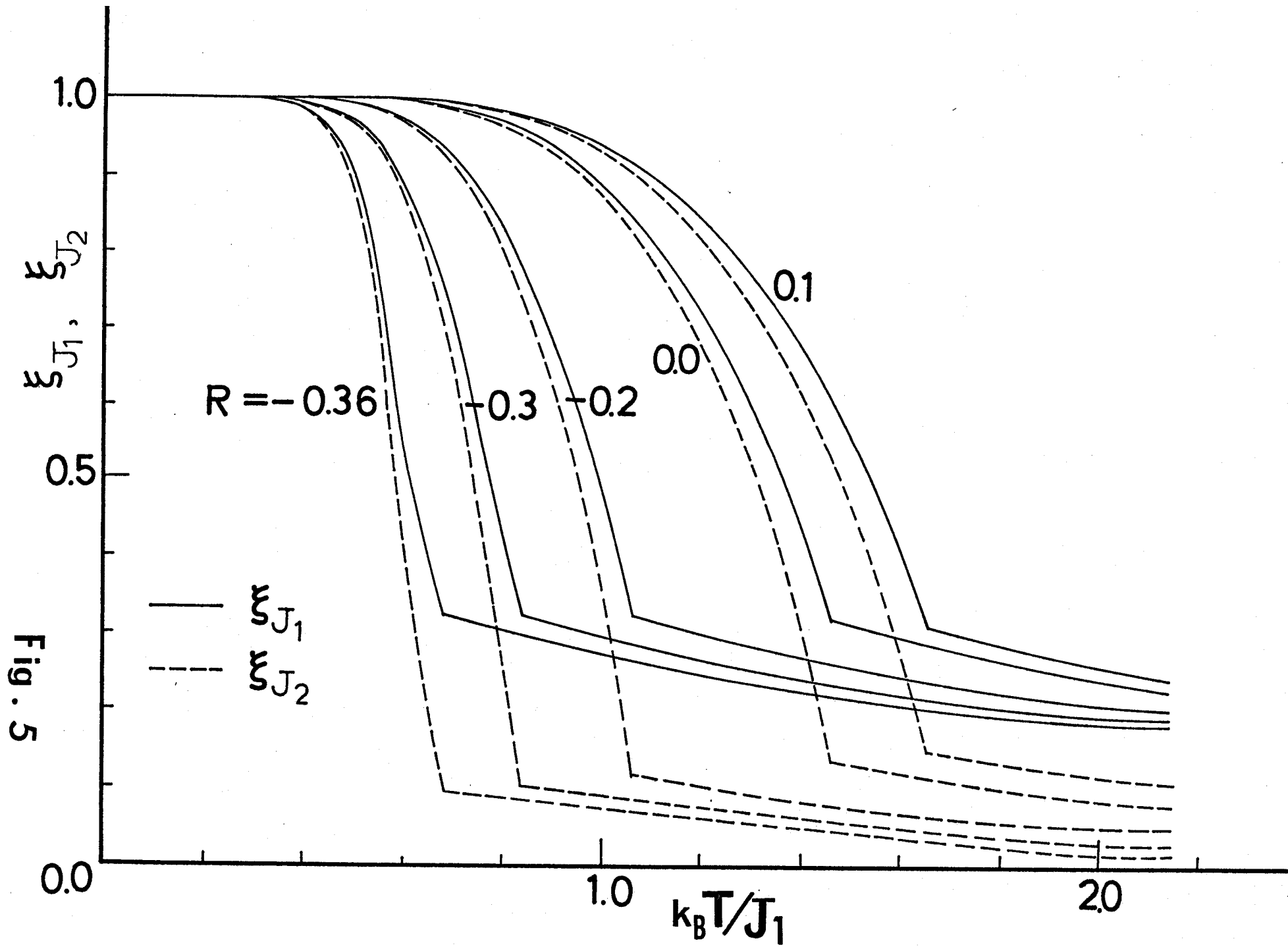


Fig. 5

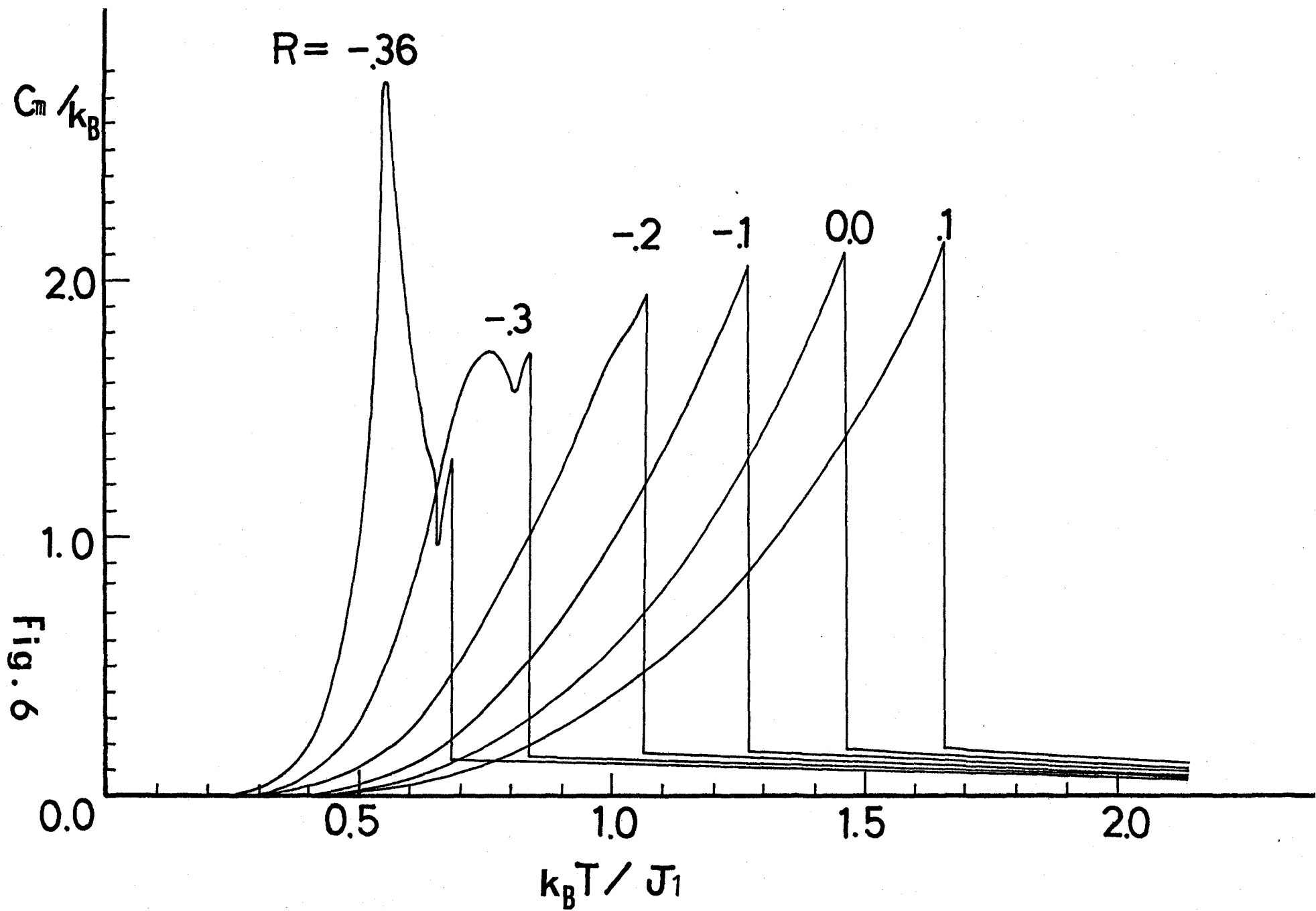
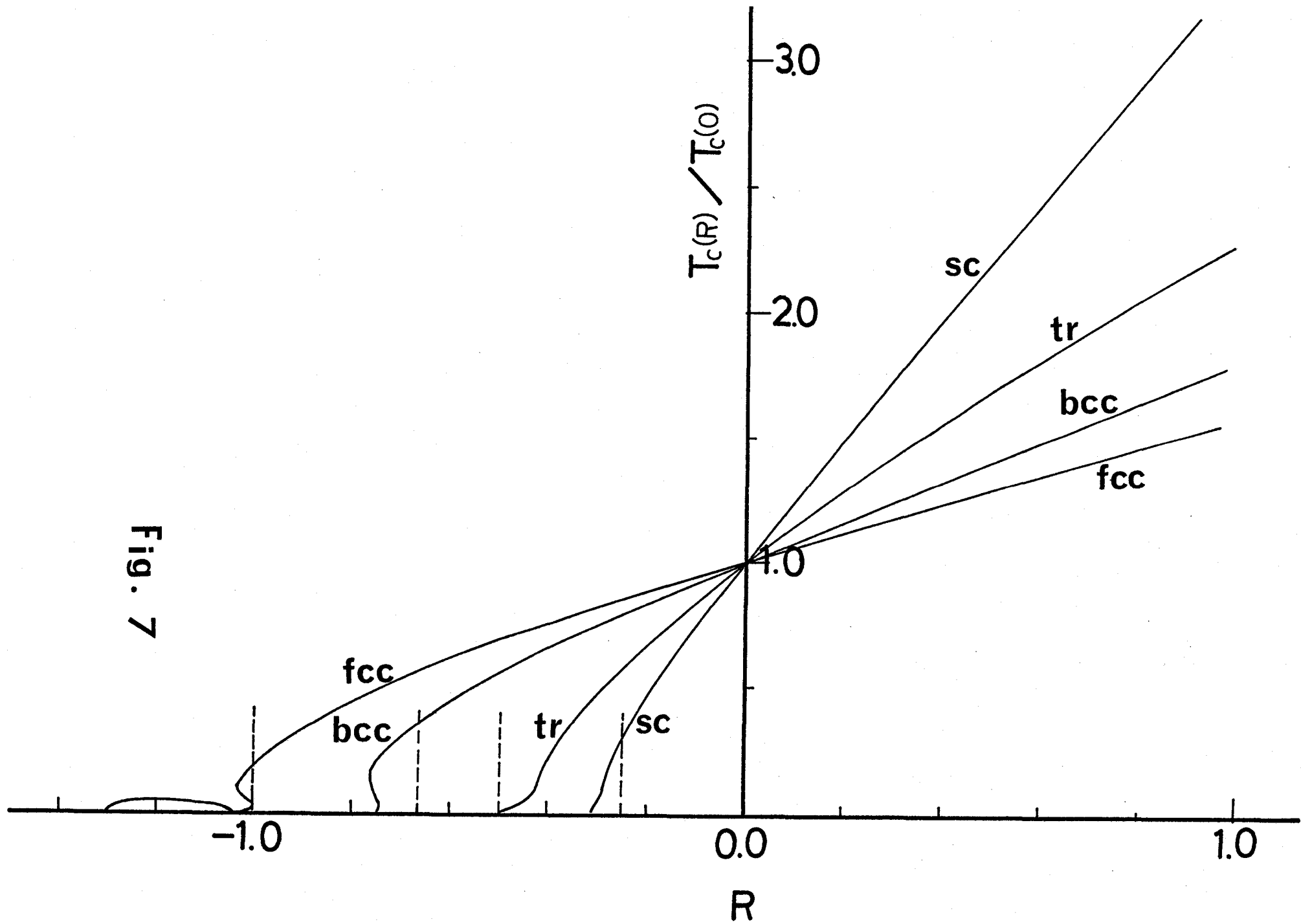


Fig. 6

Fig. 7



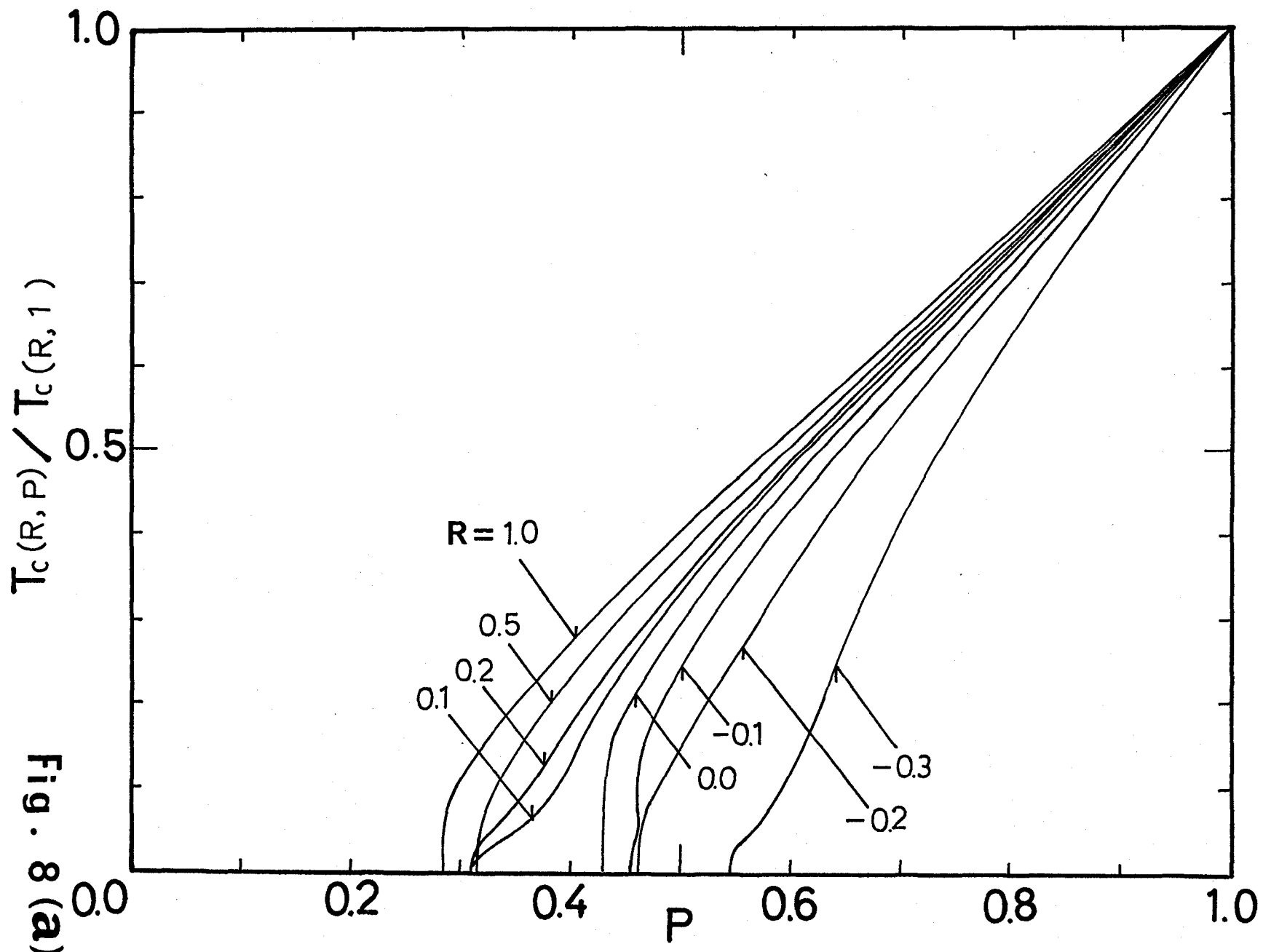


Fig. 8 (a)

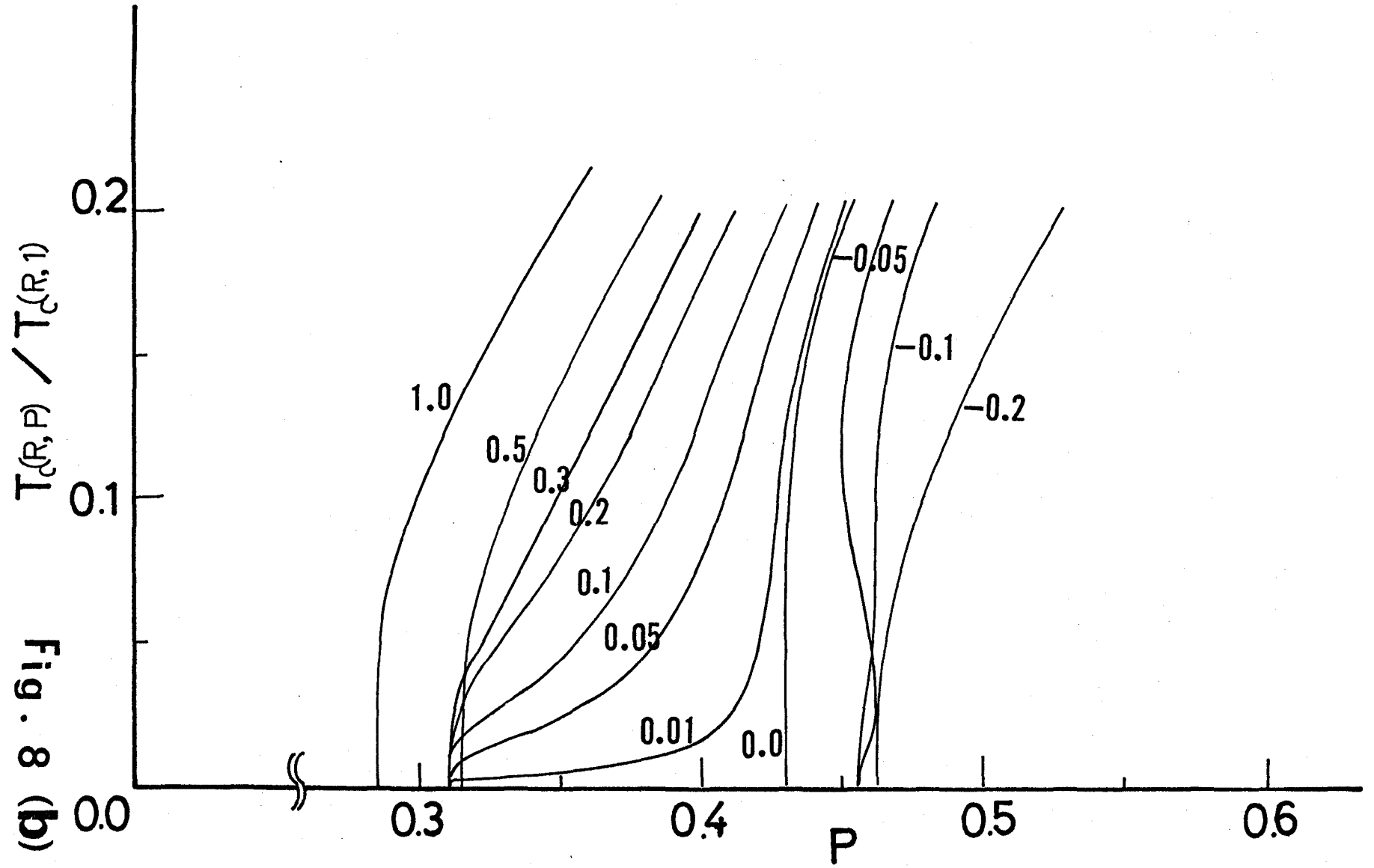


Fig. 8 (b)

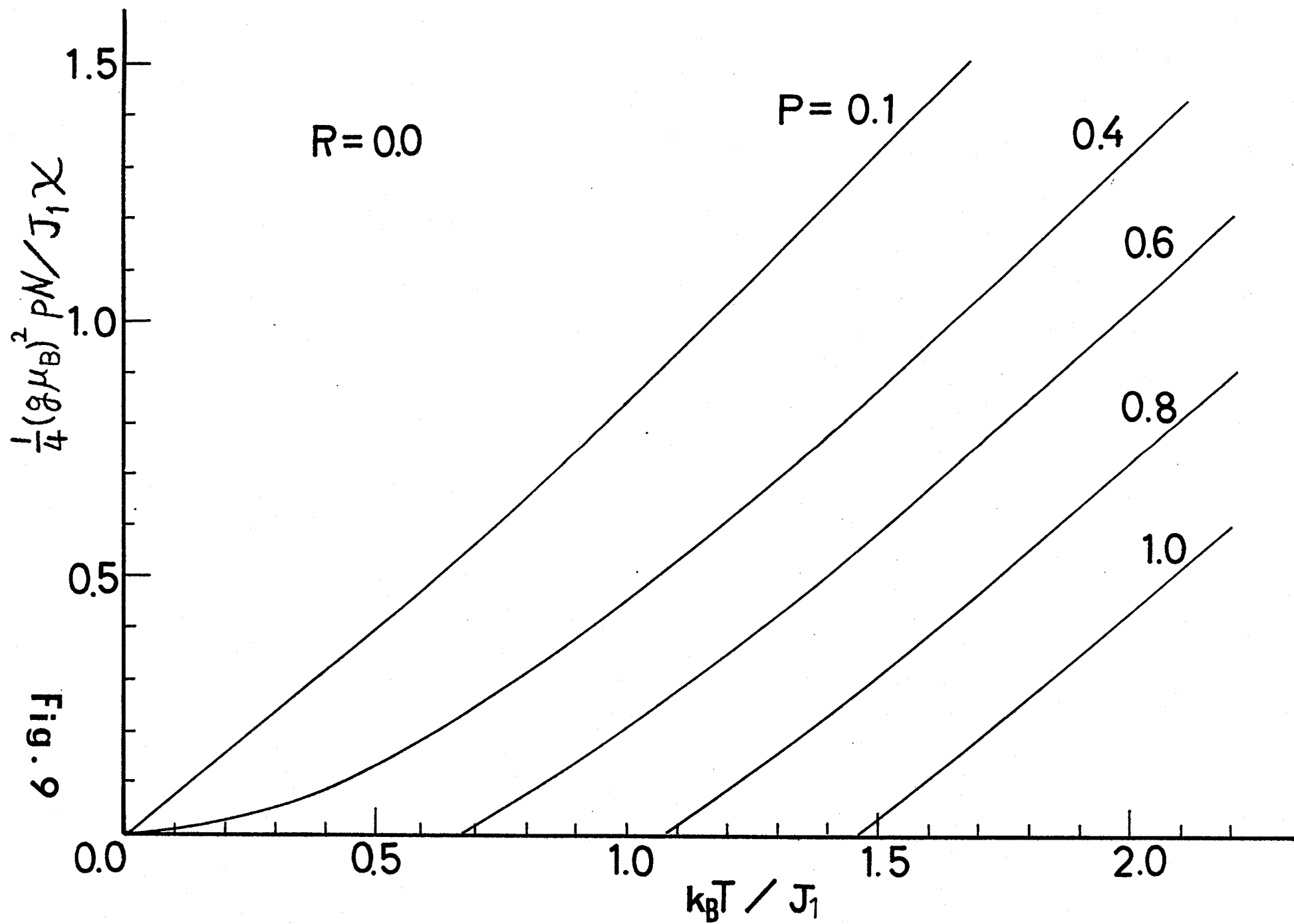


Fig. 9

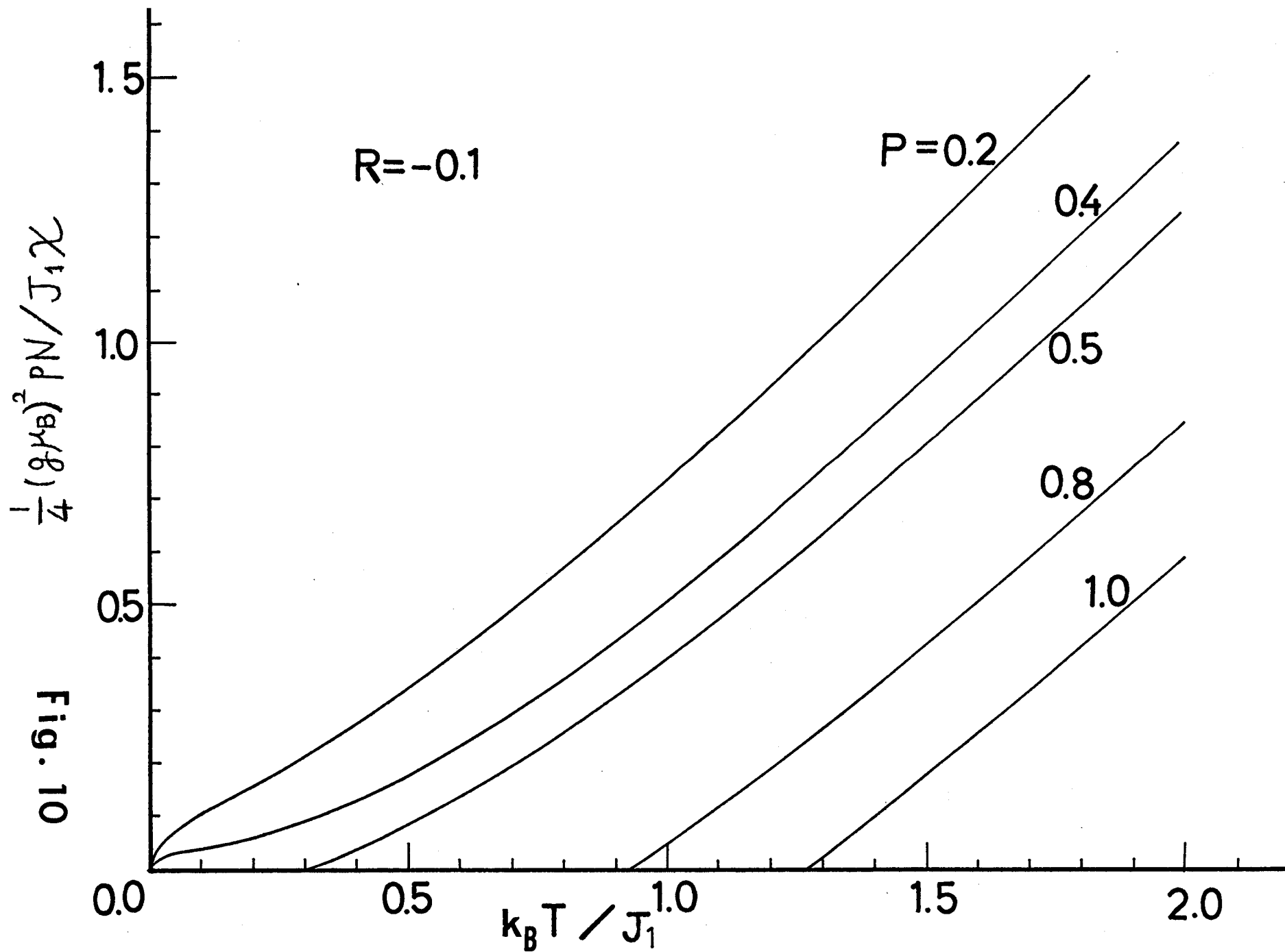


Fig. 10

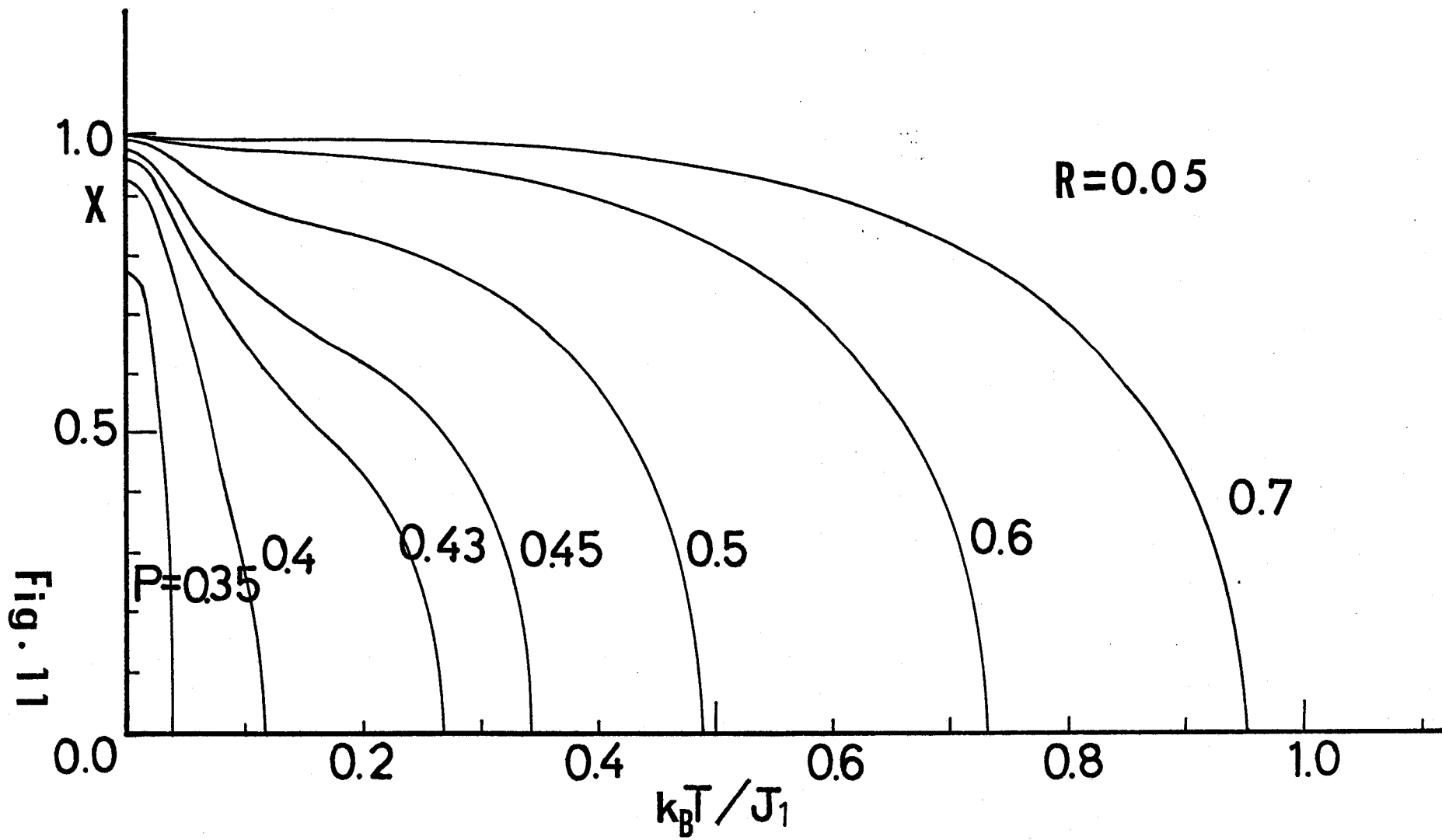


Fig. 11

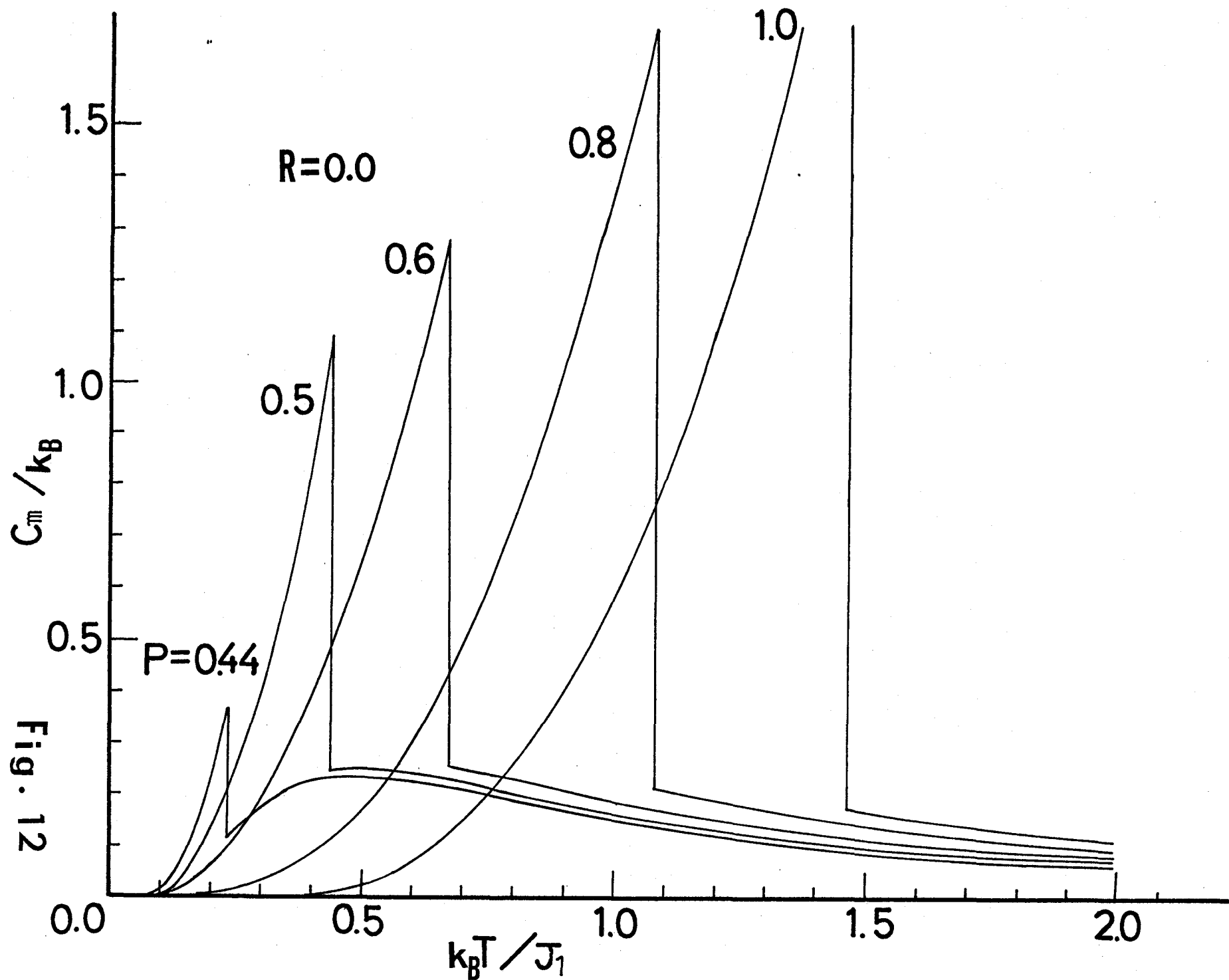


Fig. 12

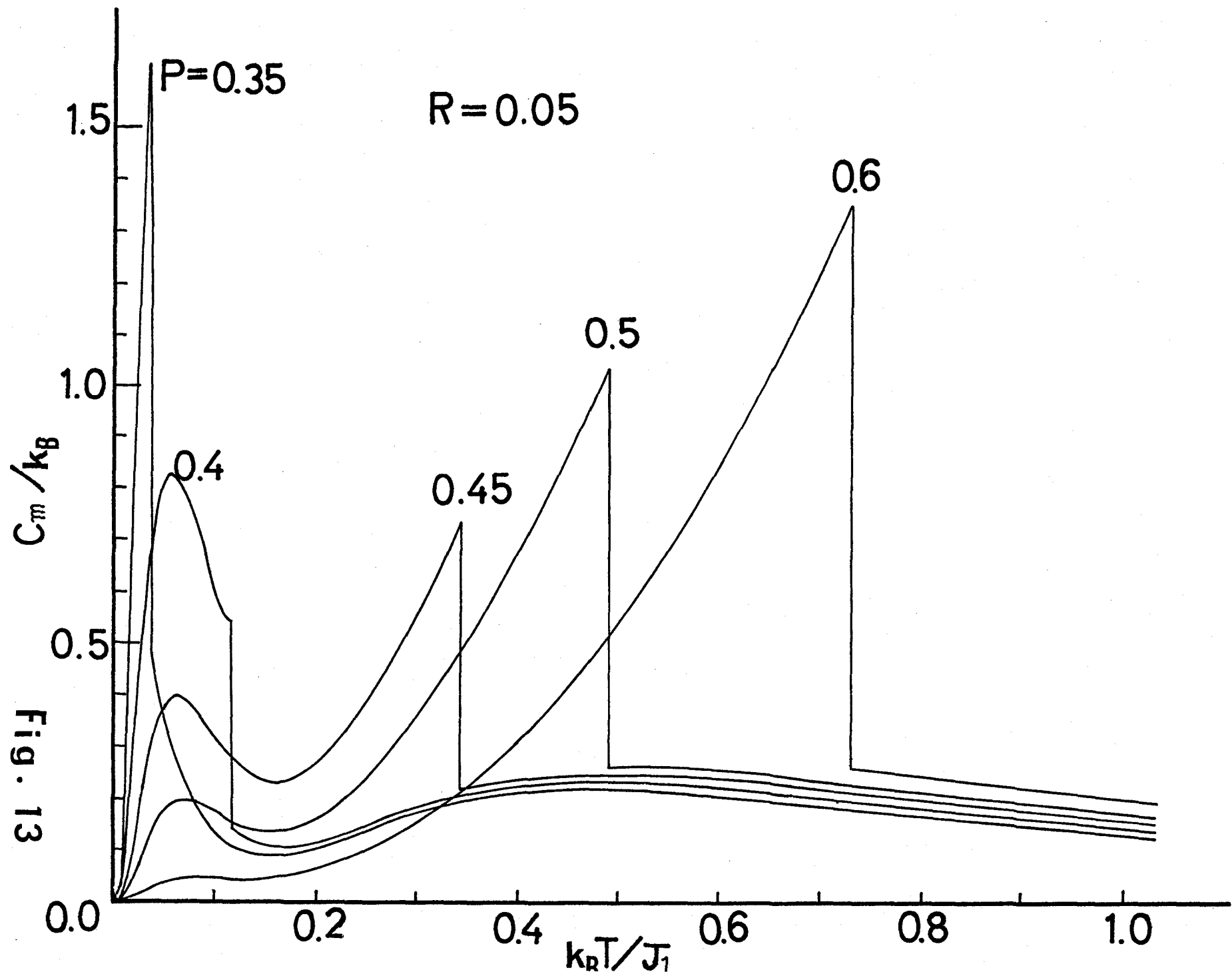


Fig. 13

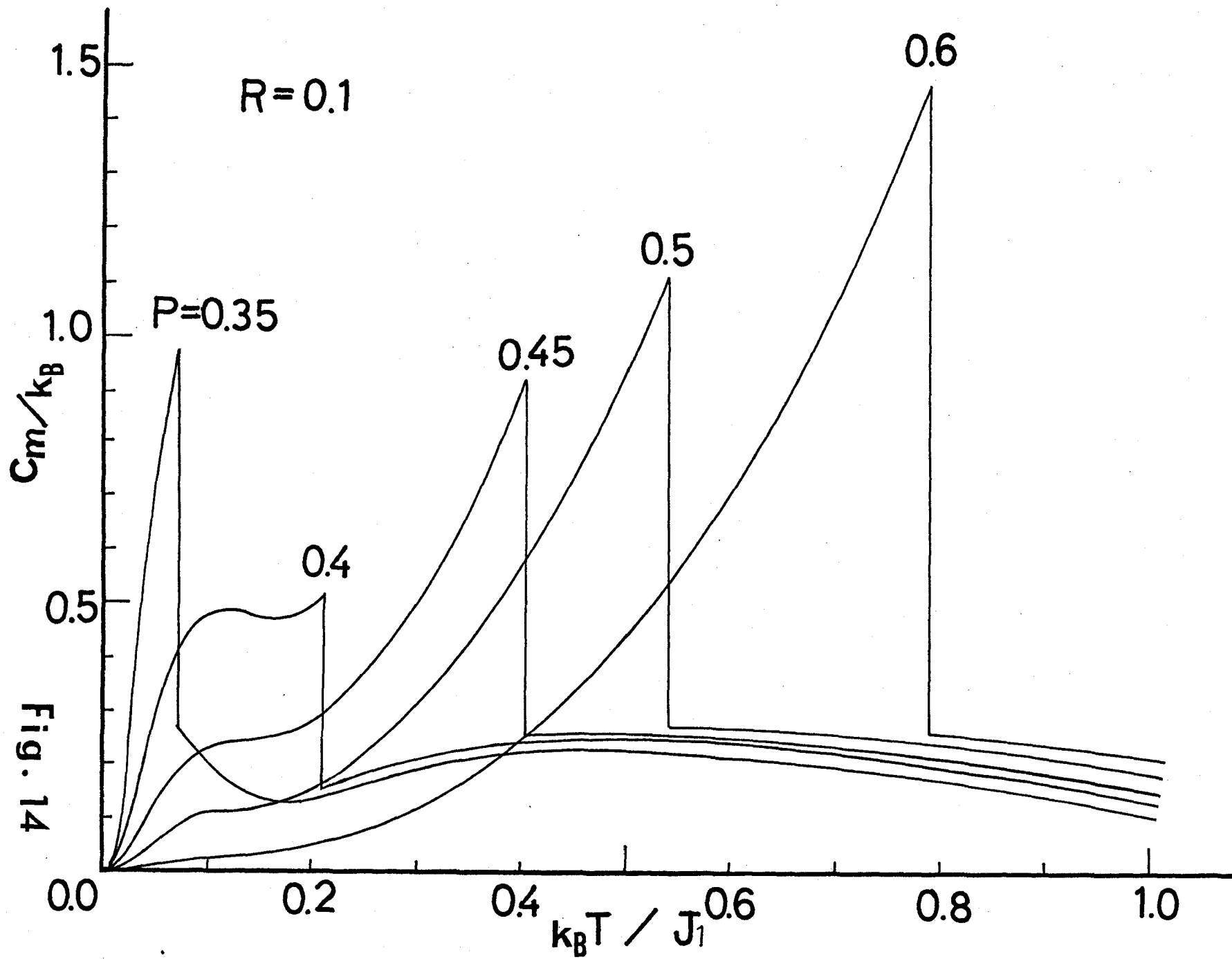


Fig. 14

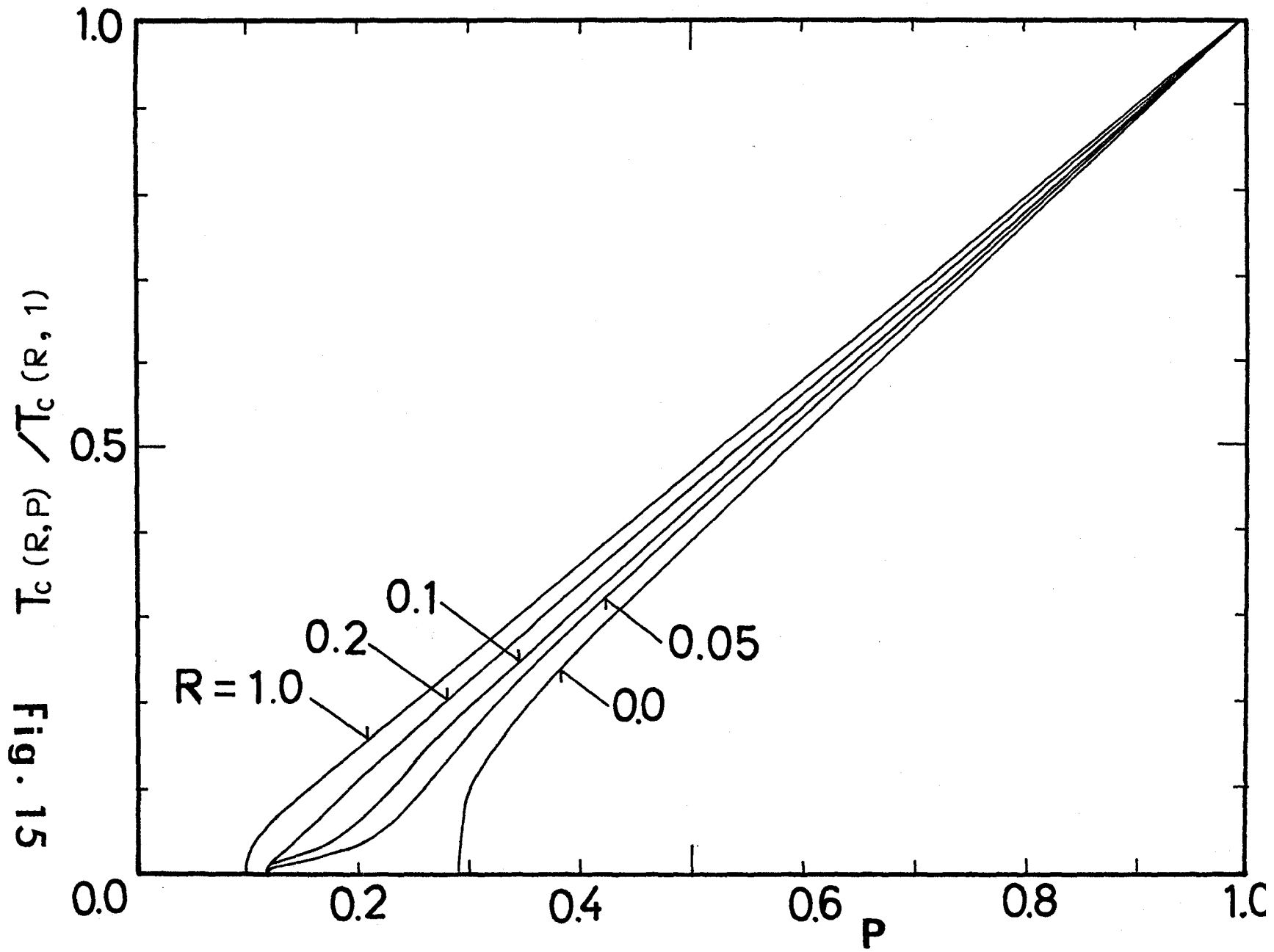


Fig. 15