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Magnetic and Thermal Properties of an Antiferromagnetic Decorated Ising Lattice in Finite Magnetic Fields

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We discuss in detail the magnetic and thermal properties of a certain kind of plane decorated Ising lattice; that is, its interesting spin-ordering characteristics, such as the temperature and magnetic field dependence of the magnetization, the susceptibility and the heat capacity are investigated. These characteristics may be classified according to the ratio between two kinds of exchange coupling parameters. The system shows metamagnetism in certain cases. The characteristic magnetic field at which a logarithmic singularity takes place in the susceptibility as well as the heat capacity is obtained as a function of temperature. In some cases two chracteristic fields are found below a certain temperature, above which only the one characteristic field survives up to a higher temperature. A number of typical curves which represent the relations of the magnetization, the magnetic susceptibility and the heat capacity with the magnetic field at a fixed temperature and with temperature at a fixed strength of magnetic field are investigated.

§ 1. Introduction

The decorated lattice was originally introduced by Syozi[1] in the case of the honeycomb lattice. By applying the decoration transformation on the one hand and by applying the star-triangle transformation on the other to that decorated lattice, he found a relation between the transition temperatures of the honeycomb and the kagomé lattices. By making use of the already-known transition temperature of the honeycomb lattice in that relation he found the transition temperature of the kagomé lattice. Naya[2] has extended this approach to include the effects of an external magnetic field and was able to obtain an expression for the spontaneous magnetization of the kagomé lattice by making use of that of the honeycomb lattice.

Syozi and Nakano[3] have made use of the decorated square lattice and other decorated lattices as a model of a ferrimagnet for which an exact calculation can be made. This model has typical features of the ferrimagnet in the temperature dependence of its spontaneous magnetization. Fisher[4] has also investigated the decorated square lattice but he has designed it to be a model of an antiferromagnet for which the magnetization can be calculated exactly even in the presence of a finite magnetic field. He has discussed the magnetic property of his model, which he calls a super-exchange antiferromagnet.

Syozi and Miyazima[5] have made use of the decorated Ising lattice to investigate the problem of dilute ferromagnetism, where the spin variable on every decorating site takes the values not only plus and minus unity but also zero, corresponding to the occupations of that site by non-magnetic as well as magnetic atoms. A similar design has been utilized by Hattori and others[6] in discussing the ferrimagnetism of a decorated square lattice where all matrix lattice sites and half of the decorating lattice sites are occupied by a sort of Ising spin and the remaining lattice sites by another sort of Ising spin. This model is thought to correspond to the ferrite with the inverse spinel structure and is in contrast with Syozi and Nakano's model[3] which corresponds to the ferrite with the normal spinel structure.

Nakano[7] has extended the exchange coupling to every pair of next-nearest neighbouring sites in Syozi and Nakano's model of ferrimagnetism and has shown that the spin ordering in such a model does not monotonously decay with increase of temperature. He has demonstrated that certain three-dimensional decorated lattices, e.g. simple cubic and body-centred cubic lattices, have three transition temperatures. Syozi[8] has found that the partly- or semi-decorated square lattice can have three transition temperatures. After him Nakano[9] has shown that the partly-decorated triangular lattice can have three transition temperatures as well and that in general the square and the triangular decorated lattices with anisotropic exchange couplings can have three transition temperatures. Miyazima[10] has shown that the condition for the appearance of three transition temperatures is more easily satisfied by increasing the number of decoration sites on every bond in the lattice. Syozi and Nakano[11] have shown that three transition temperatures can exist in the semidecorated hempleaf lattice and a certain super-decorated triangular lattice, in which the phases separated by these three transition temperatures are ferro-, para-, ferro- and para-magnetic ones in the order of successively rising of temperatures in contrast with the ferro-, para-, antiferro- and paramagnetic phases in those models which we have hitherto mentioned.

In this article we modify the decorated lattice model in accordance with Fisher's super-antiferromagnet so that we can calculate exactly the partition function of the system even in the presence of a finite magnetic field. This may be distinguished from Fisher's model by the existence of an exchange interaction acting between every pair of nearest-neighbouring spins of the matrix lattice. By means of such a modification, all decorated lattices, mentioned above, are transformed into the corresponding antiferromagnet lattices where the ordered phase is always antiferromagnetic. In particular the exchange coupling parameters may be appropriately chosen so that the lattice has three transition temperatures; the phases separated by these temperatures are antiferro-, para-, antiferro- and para-magnetic ones in the order of increasing temperature.

In § 2 the model is explained. We confine ourselves to investigating the decorated square Ising lattice which has only a single transition temperature; the general features of the argument are applicable to other appropriate decorated lattices. In this system we suggest that the spin ordering does not decay monotonously with increasing temperature although the long range ordering never appears above a single transition temperature. The system shows certain complicated behaviours when exposed to an external magnetic field. We can calculate the partition function exactly in the presence of a finite applied field and with its use obtain expressions for the magnetization, the susceptibility and the heat capacity of the system. These calculations are given in § 2.

As shown in § 2, the partition function of the decorated lattice can be reduced to that of the matrix lattice which is obtained by removing every decorating site. The parameter x of the effective exchange coupling in the matrix lattice is investigated as a function of the magnetic field and temperature in § 3. A certain kind of singularity occurs in the magnetic and thermal properties of the system at a particular magnetic field strength, called the critical field, which is dependent on the temperature. Conversely, the singularity occurs at a critical temperature which is dependent on the magnetic field. By solving the equation either $x=x_C$ or $x=-x_C$, one can determine the critical field H_C as a function of temperature T as well as the critical temperature T_C as a function of the magnetic field H, where x_C denotes the critical value of x which is characteristic for the matrix lattice. That is, one can get $H_C=H_C(T)$ from the equation $x(T, H_C)=\pm x_C$.

Thus there can exist two kinds of critical fields and two kinds of critical temperatures, corresponding to plus and minus x_C in the above equations. Accordingly, we denote the critical fields and temperatures by H_+, H_-, T_+ and T_- when we need to distinguish between them. We show the critical fields as functions of temperature schematically in § 3.

In § 4 we investigate qualitatively the field dependences of the magnetization, the susceptibility and the heat capacity at constant temperature. Under certain conditions, a metamagnetic transition either of a single step or of double steps takes place in the relation of magnetization with magnetic field. The susceptibility and the heat capacity show logarithmic singularities at the critical fields. We also discuss the temperature dependences of these quantities fixing the magnetic field constant and see that the susceptibility and the heat capacity diverge logarithmically at the critical temperature.

We calculate numerically, in § 5, the magnetization, the susceptibility and the heat capacity as functions of the magnetic field and temperature for a few typical cases of the exchange coupling ratio and show rather precise curves relating these quantities to the magnetic field and temperature. In some cases the system exhibits a metamagnetic transition of one stage or of two stages.

§ 2. The partition function, the magnetization, the susceptibility and the heat capacity

The Ising lattice, an antiferromagnetic square lattice investigated in this article, is shown in Fig. 1. White circles, each of which is occupied by a sort of Ising spin (called a white spin for brevity), compose a square lattice. This plane lattice is decorated at the midpoint of every bond-line by a black circle which is occupied by a spin (called a black spin) different from the white spin. The black and white spins are assumed to be magnetic, moment β and non-magnetic respectively.

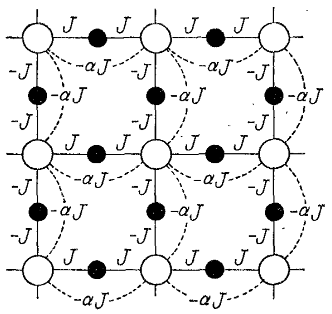


Fig. 1. Antiferromagnet of decorated lattice. $J>0$, ●: magnetic spin, ○: non-magnetic spin.

We assume the exchange coupling to be either $+J$ or $-J(J>0)$ between every pair of black and white spins which are nearest-neighbouring to each other and $-\alpha J$ between every pair of nearest-neighbouring white spins, as shown in Fig. 1. In order to make the description definite, we restrict the following discussion exclusively to the case of the particular decorated square lattice, shown in Fig. 1, where the plus and minus signs in $\pm J$ are for the transverse and longitudinal bonds, respectively.

The Hamiltonian H of this system exposed to an external magnetic field H can be written with the use of the Ising spin variables μ_i and ν_k respectively for the white and black spins:

$$H=-J\sum_{(ik):\circ-\bullet}\mu_i\nu_k+J\sum_{(ik):\,\circ\atop\bullet}\mu_i\nu_k+\alpha J\sum_{(ij):\circ-\circ}\mu_i\mu_j-\beta H\sum_{k:\bullet}\nu_k\,, \tag{1}$$

where the first and second summations are taken over the pairs of nearest-neighbouring white and black spins on the transverse and longitudinal bonds respectively, the third summation is taken over every pair of nearest-neighbouring white spins, and the last summation represents the magnetic energy of all black (magnetic) spins with magnetic moment β due to the applied magnetic field H.

The partition function Z of the system can be expressed as

$$Z=\sum_{\{\mu\}}\sum_{\{\nu\}}\exp\left(-\frac{H}{kT}\right)=A^{2N}Z_0(x)\,, \tag{2}$$

where $\sum_{\{\mu\}}$ and $\sum_{\{\nu\}}$ represent the summations over all spin variables $\mu_i, \cdots$ and $\nu_k, \cdots$ respectively for the white and black spins, plus and minus the unity, and N denotes the total number of lattice sites of the square lattice. $Z_0(x)$ denotes the partition function of this matrix lattice. The parameters A and x are defined in terms of the parameters,

$$J' \equiv \frac{J}{kT} \quad \text{and} \quad \eta \equiv \frac{\beta H}{J}, \tag{3}$$

and they are given by

$$A = 2[\cosh\{(2+\eta)J'\}\cosh\{(2-\eta)J'\}\cosh^2(\eta J')]^{1/4},$$
$$x = \frac{1}{4}\ln\left[\frac{\cosh\{(2+\eta)J'\}\cosh\{(2-\eta)J'\}}{\cosh^2(\eta J')}\right] - \alpha J'. \tag{4}$$

The partition function $Z_0(x)$ in (2) is written as

$$Z_0(x) = \sum_{\mu_1=\pm 1} \cdots \sum_{\mu_N=\pm 1} \exp[x \sum_{(ij)} \mu_i \mu_j], \tag{5}$$

where $\sum_{(ij)}$ in the exponent means the summation over all pairs of the nearest-neighbouring sites of the square lattice.

The magnetization of the system can be derived from the formula

$$M = \frac{\beta}{J'}\frac{\partial}{\partial \eta}\ln Z. \tag{6}$$

By making use of (2), (3) and (4), one obtains from (6)

$$M = \frac{2N\beta}{J'}\left[\frac{\partial}{\partial\eta}\ln A + \frac{\partial x}{\partial \eta}U(x)\right] \tag{7}$$
$$= \frac{N\beta}{2}[\tanh(2J'+\eta J') - \tanh(2J'-\eta J') + 2\tanh(\eta J')$$
$$+ U(x)\{\tanh(2J'+\eta J') - \tanh(2J'-\eta J') - 2\tanh(\eta J')\}],$$

where $U(x)$ denotes the pair correlation of the nearest-neighbouring spins in the square lattice and is given by

$$U(x) = \frac{1}{2N}\frac{\partial}{\partial x}\ln Z_0(x). \tag{8}$$

One can obtain the magnetic susceptibility by differentiating (7) with respect to the magnetic field; viz.

$$\chi = \frac{\beta}{J}\frac{\partial M}{\partial \eta} = \frac{N\beta^2}{2kT}[\mathrm{sech}^2(2J'+\eta J') + \mathrm{sech}^2(2J'-\eta J')$$
$$+ 2\,\mathrm{sech}^2(\eta J') + U\{\mathrm{sech}^2(2J'+\eta J') + \mathrm{sech}^2(2J'-\eta J') - 2\,\mathrm{sech}^2(\eta J')\}$$
$$+ \frac{1}{4}\frac{\partial U}{\partial x}\{\tanh(2J'+\eta J') - \tanh(2J'-\eta J') - 2\tanh(\eta J')\}^2]. \tag{9}$$

The enthalpy U of the system can be calculated from the formula $U = -J \times \partial \ln Z/\partial J'$. The heat capacity at a constant field is then obtained as

$$C = \partial U/\partial T = (NkJ'^2/2)[(2+\eta)^2\,\mathrm{sech}^2(2J'+\eta J') + (2-\eta)^2$$
$$\times\,\mathrm{sech}^2(2J'-\eta J') + 2\eta^2\mathrm{sech}^2(\eta J') + U\{(2+\eta)^2\,\mathrm{sech}^2(2J'+\eta J')$$

$$+ (2-\eta)^2 \operatorname{sech}^2(2J'-\eta J') - 2\eta^2 \operatorname{sech}^2(\eta J')\} + (1/4)\,(\partial U/\partial x) \tag{10}$$
$$\{(2+\eta)\tanh(2J'+\eta J') + (2-\eta)\tanh(2J'-\eta J') - 2\eta\tanh(\eta J') - 4\alpha\}^2].$$

§ 3. Preliminary investigation and the critical magnetic fields

We investigate the effective exchange coupling x; that is, its relation with temperature and the magnetic field or with the parameters J' and η defined in (3). By differentiating x in (4) in respect of J' one gets

$$\partial x/\partial J' = x_0' - \alpha\,,$$

where

$$x_0' = (1/4)\,[(2+\eta)\tanh(2J'+\eta J') + (2-\eta)\tanh(2J'-\eta J') - 2\eta\tanh(\eta J')].$$

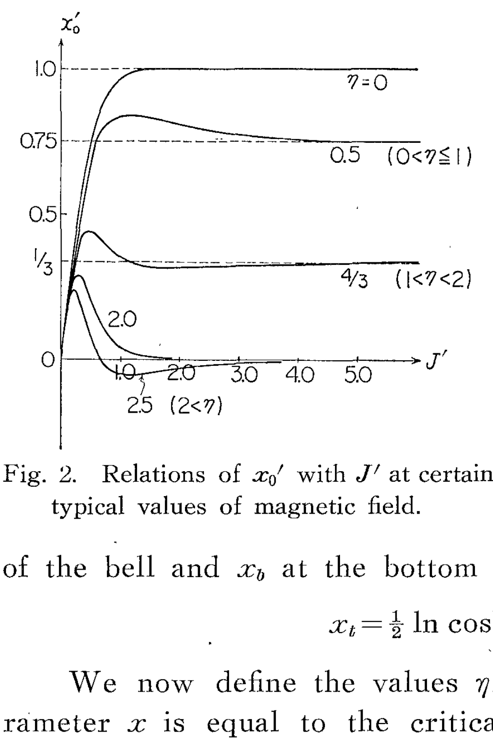


Fig. 2. Relations of x_0' with J' at certain typical values of magnetic field.

The curve which represents the relation between x_0' and J' is shown in Fig. 2. On the basis of this figure the x versus J' curves are shown schematically in Fig. 3 and they are classified according to the value of the parameter η. The maximum and minimum values of x_0' in Fig. 2, if they exist, are denoted by α_0 and α_1, respectively, in Fig. 3.

The parameter x is an even function of the magnetic field or of the parameter η at any given temperature and is represented by the bell-like curve shown in Fig. 4, where the value x_t of x at the top of the bell and x_b at the bottom are given respectively by

$$x_t = \tfrac{1}{2}\ln\cosh(2J') - \alpha J',\quad x_b = -\alpha J'. \tag{11}$$

We now define the values η_+ and η_- for which the effective coupling parameter x is equal to the critical value x_C of the square lattice and to $-x_C$ respectively. Ferromagnetic and antiferromagnetic orders exist in the square Ising lattice if the parameter x in (4) is more than x_C and less than $-x_C$ respectively, where x_C equals[12] $(1/2)\ln(\sqrt{2}+1) = 0.4407$. The partition function Z has a singularity at the values η_+ and η_-; $\partial^2 Z/\partial T^2$, $\partial^2 Z/\partial T\partial H$ and $\partial^2 Z/\partial H^2$ show logarithmic singularities at these points. In other words, the derivative dU/dx of the pair correlation given by (8), to which the singularities of the second derivatives of the partition function correspond, diverges logarithmically[12] at $\eta = \eta_+$ and η_-.

One can say that the present system has two critical fields at most at certain

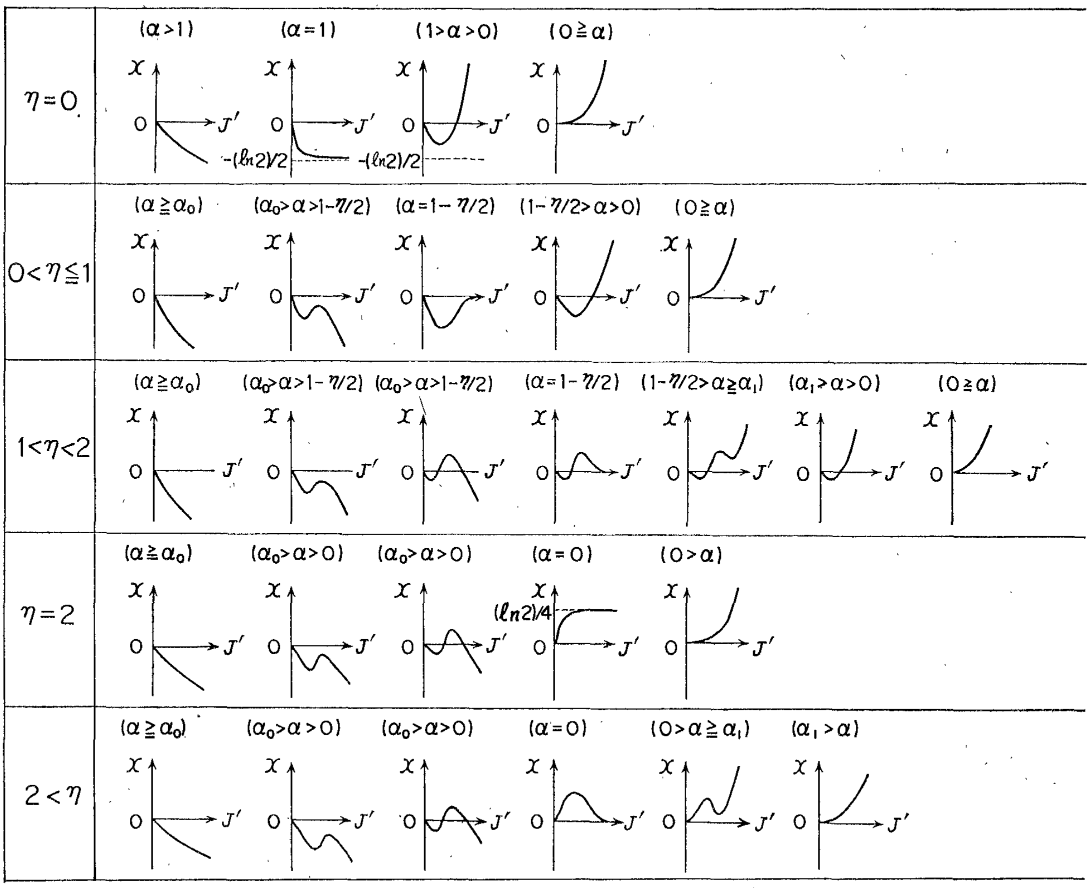


Fig. 3. Relations of the effective exchange coupling x with the reciprocal J' of temperature in the cases of different coupling ratios and different field strengths.

temperatures; namely the critical fields H_+ and H_- which are equal to $\eta_+ J/\beta$ and $\eta_- J/\beta$, respectively. We may denote these critical values by H_C and η_C, irrespective of whether $x=x_C$ or $x=-x_C$. From the x versus η curve shown in Fig. 4, it is apparent whether there exist both, either or none of η_+ and η_- at a given temperature and a given exchange coupling ratio α.

The critical field H_+ exists in the temperature region, $x_b<x_C<x_t$, where x_b and x_t are functions of temperatures given by (11) and x_C is the critical value charasteristic for the lattice. Another critical field H_- exists for $x_b<-x_C<x_t$. H_+ and H_- shrink to zero at the temperatures T_{+0} and T_{-0} respectively, where the equations $x_t=x_C$ and $x_t=-x_C$ are satisfied respectively. On the other hand, H_+ and H_- diverge to the infinity at the temperatures $T_{+\infty}$ and $T_{-\infty}$ where x_b equals x_C and $-x_C$ respectively. One can conclude that both H_+ and H_- exist so far as $x_b<-x_C<x_C<x_t$, only H_+ exists for $-x_C<x_b<x_C<x_t$, only H_- exists for $x_b<-x_C<x_t<x_C$ and neither H_+ nor H_- exist otherwise; namely in the case that either $x_t<-x_C$, $x_C<x_b$ or $-x_C<x_b<x_t<x_C$.

In order to investigate the temperature dependence of the critical field H_C, it is useful to picture those curves ①, ②, ③ and ④ on the $\alpha-J'$ plane as shown

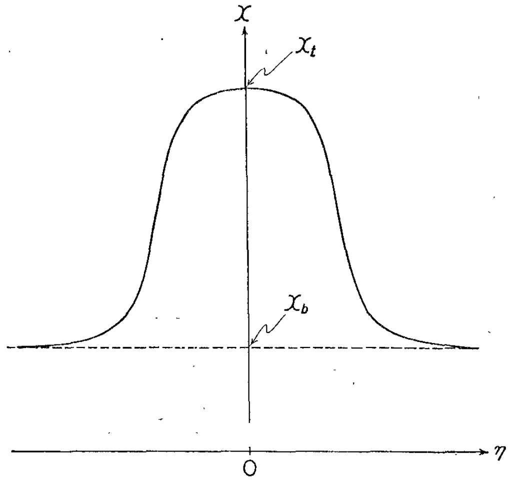


Fig. 4. Relation of the effective exchange coupling parameter x with the field parameter η.

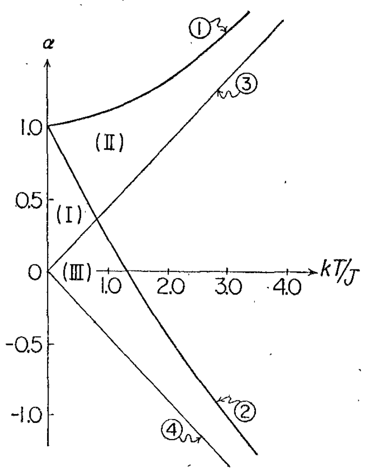


Fig. 5. Critical field map on the α-J' plane. curves ①: $H_-=0$, ②: $H_+=0$, ③: $H_-=\infty$, ④: $H_+=\infty$; domains (I): both H_+ and H_- exist. (II): H_- exists, (III): H_+ exists. At the intersection of ② with ③ $kT/J=0.817$ and $\alpha=0.360$.

in Fig. 5 which represent the relations $x_t=-x_C$, $x_t=x_C$, $x_b=-x_C$ and $x_b=x_C$ respectively. $H_-=0$ on the curve ①, $H_+=0$ on ②, $H_-=\infty$ on ③ and $H_+=\infty$ on ④. There exist two critical fields H_+ and H_- in the region (I) in Fig. 5, only H_- in (II), only H_+ in (III) and none in any other regions.

The critical field-temperature curve is drawn as shown in Fig. 6 and is considered in Appendix 1 for some exchange-coupling ratios, α.

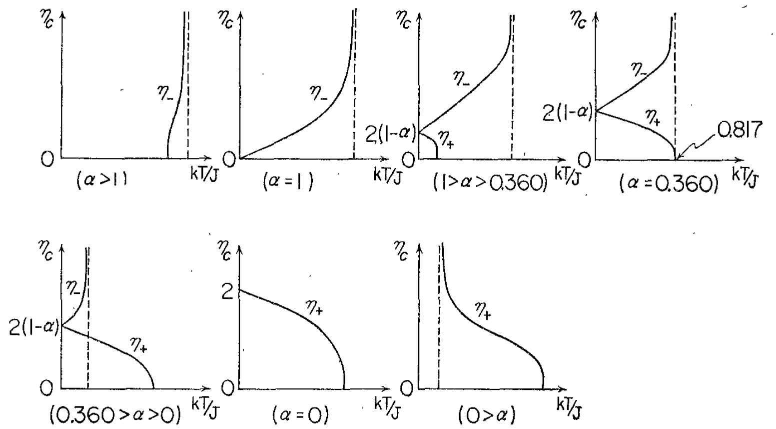


Fig. 6. Relations of the critical field with temperature in various cases of the coupling ratio α.

§ 4. Temperature and field dependences of the magnetization, the susceptibility and the heat capacity

We first discuss the limiting constant temperature properties of the magnetization M, the susceptibility χ and the heat capacity C in the limits of vanishing and increasing of magnetic field. It is seen from (7) that M is proportional to the field in the limit of vanishing field,

$$M \simeq (N\beta^2/kT)[2-(1+U_{H=0})\tanh^2(2J')]H, \tag{12}$$

where $U_{H=0}$ denotes the spin-pair correlation (8) with $\eta=H=0$ substituted in the parameter x given by (4). In another extreme, $H=\infty$, of the field, one obtains

$$M=2N\beta. \tag{13}$$

The limiting properties of the susceptibility χ and the heat capacity C are seen from (9) and (10). In the limit of vanishing field one obtains

$$\begin{aligned}\chi &\simeq \frac{N\beta^2}{kT}\left[2-(1+U_{H=0})\tanh^2(2J')+\frac{1}{2}\left(\frac{\partial U}{\partial x}\right)_{H=0}\tanh^2(2J')\left(\frac{\beta H}{kT}\right)^2\right],\\ C &\simeq 4NkJ'^2\left[(1+U_{H=0})\operatorname{sech}^2(2J')+\left(\frac{\partial U}{\partial x}\right)_{H=0}\frac{\{\tanh(2J')-\alpha\}^2}{2}\right].\end{aligned} \tag{14}$$

In the limit of extremely strong field,

$$\chi \simeq 0$$

and

$$C \simeq 2Nk\alpha^2J'^2\left(\frac{\partial U}{\partial x}\right)_{H=\infty}=2Nk\alpha^2J'^2\left(\frac{\partial U}{\partial x}\right)_{x=-\alpha J'}=\text{finite constant}. \tag{15}$$

The singularities of these quantities in the vicinity of the critical field are discussed in Appendix 2 and are shown in Table I.

Table I. Singularities of the magnetization, the susceptibility and the heat capacity in the vicinity of the critical field H_C (=either H_+ or H_-).

H_C	M	χ	C
non-zero	$-(H-H_C)\ln\|H-H_C\|$	$-\ln\|H-H_C\|$	$-\ln\|H-H_C\|$
zero	$-H^3\ln H$	$-H^2\ln H$	$-\ln H$

On the basis of (12), (13), (14), (15) and Table I as well as the investigation of the critical field in the preceding section, one can picture the relations of the magnetization, the susceptibility and the heat capacity with the magnetic field by fixing the temperature constant, as shown in Fig. 7, where several curves with different characteristics appear according to the values of the temperature T and the exchange coupling ratio α.

We next discuss the limiting properties of M, χ and C in the limits of falling and rising of temperature at a constant magnetic field. It is seen from (7) that the magnetization tends to zero in the limit of infinitely high temperature, $J'=0$, and in the low temperature limit, $J'=\infty$, to a certain value which depends upon the strength of magnetic field as well as the exchange coupling ratio α

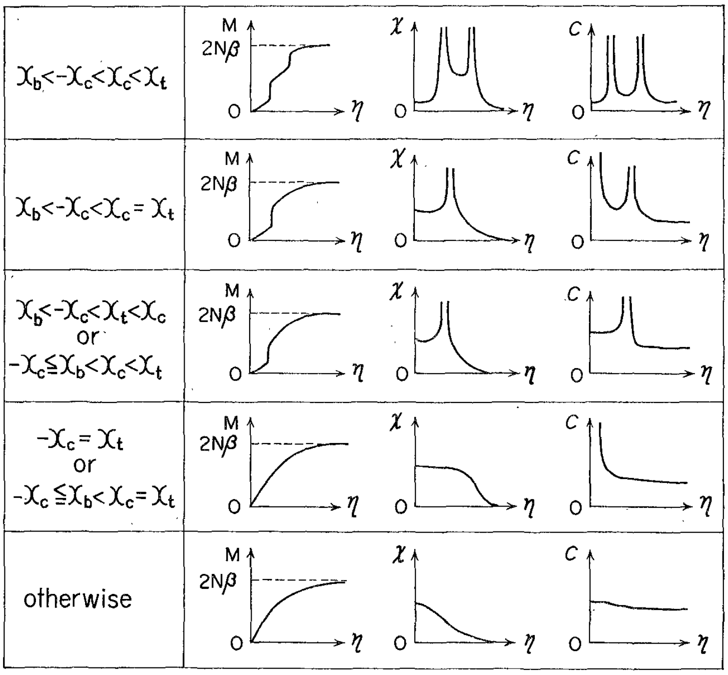


Fig. 7. Relations of the mgnetization M, the susceptibility χ and the heat capacity C with the field parameter η at various temperatures and coupling ratios.

and which is given in the following. In the case $\eta>2$, M is equal to $2N\beta$. When $\eta=2$, M equals $N(3-U_{J'=\infty})\beta/2$ which reduces to $2N\beta$, $N\beta[3-U(x=4^{-1}\ln 2)]/2$ and $N\beta$ according to whether $\alpha>0$, $\alpha=0$ or $\alpha<0$. When $2>\eta>0$, M tends to $N\beta(1-U_{J'=\infty})$ which reduces to $2N\beta$, $N\beta$ and zero according to whether α is larger than, equal to, or smaller than $1-(\eta/2)$. In the vicinity of the vanishing temperature, M varies proportionally to $\exp(-\tau/T)$ in which τ is a constant and the first as well as the higher derivatives of M vanish.

One can see from (9) that χ tends to $2N\beta^2/kT$ in the high temperature limit and tends to those different values in the low temprature limit, depending on the values of the parameters η and α; that is, when $\eta>2$ $\chi\rightarrow 0$, when $\eta=2$ $\chi\rightarrow(N\beta^2/2kT)[1+U+(1/4)\partial U/\partial x]_{J'=\infty}$, which reduces to zero, $(N\beta^2/kT)[1+U+(1/4)\partial U/\partial x]_{x=(\ln 2)/4}$ and $N\beta^2/kT$ in the cases $\alpha>0$, $\alpha=0$ and $\alpha<0$ respectively, when $2>\eta>0$ $\chi\rightarrow(N\beta^2/2kT)(\partial U/\partial x)_{J'=\infty}$, which reduces to zero, $N\beta^2/2kT$ and zero in the cases of α larger than, equal to and smaller than $1-(\eta/2)$ respectively, and when $\eta=0$ $\chi\rightarrow(N\beta^2/kT)[1-U]_{J'=\infty}$, which reduces to $2N\beta^2/kT$, $(N\beta^2/kT)[1+U]_{x=(\ln 2)/2}$ and zero in the cases $\alpha>1$, $\alpha=1$ and $\alpha<1$ respectively.

It is seen from (10) that the heat capacity C tends to zero in both high and low temperature limits. It behaves like $\exp(-\gamma/T)$ at low temperatures and like $1/T^2$ at high temperatures.

The singularities of M, χ and C in the vicinity of the critical temperature are discussed in Appendix II and are shown in Table II below. On the basis of the limiting properties of the quantities as mentioned above and of the singularities shown in Table II, one can picture the curves which represent the relations of the magnetization, the susceptibility and the heat capacity with temperature at any strength of magnetic field as are shown in Fig. 8. They are given in a

Table II. Singularities of the magnetization, the susceptibility and the heat capacity in the vicinity of the critical temperature T_C(=either T_+ or T_-).

at the field strength	in the vicinity of the critical temperature	magnetization M	susceptibility χ	heat capacity C
$H \neq 0$	T_+	$-(T-T_+)\ln\|T-T_+\|$	$-\ln\|T-T_+\|$	$-\ln\|T-T_+\|$
	T_-	$(T-T_-)\ln\|T-T_-\|$	$-\ln\|T-T_-\|$	$-\ln\|T-T_-\|$
$H=0$	T_+	zero	$-(T-T_+)\ln\|T-T_+\|$	$-\ln\|T-T_+\|$
	T_-	zero	$(T-T_-)\ln\|T-T_-\|$	$-\ln\|T-T_-\|$

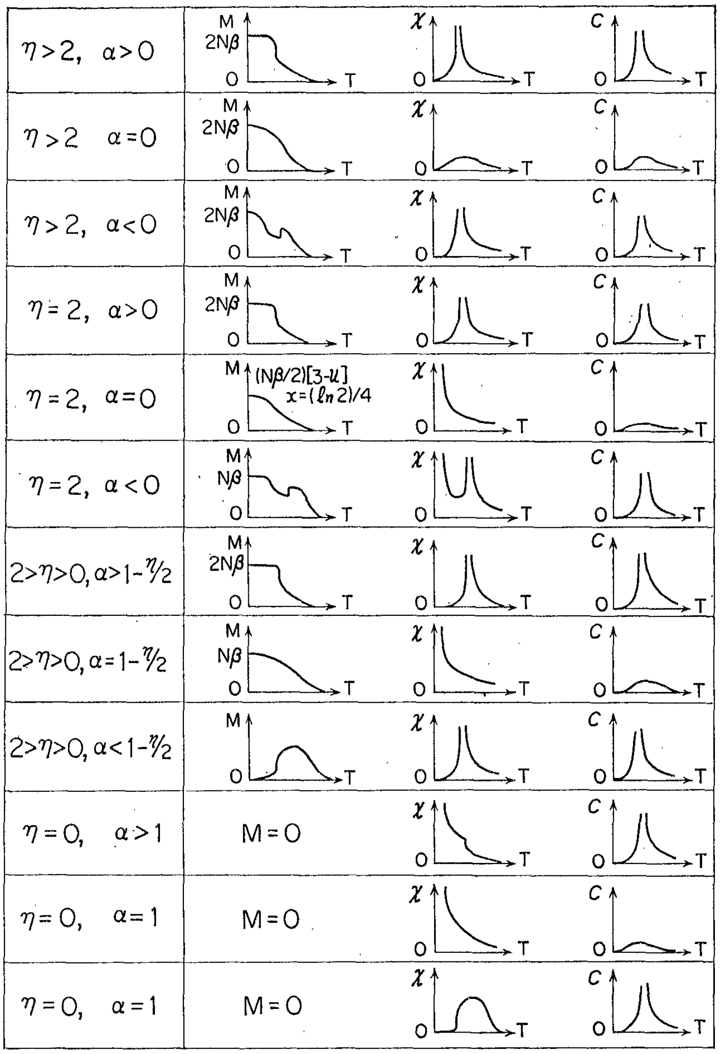


Fig. 8. Relations of the magnetization M, the susceptibility χ and the heat capacity C with temperature at various field strengths and coupling ratios.

certain number of typical cases which are specified by the values of magnetic field and coupling parameters and are characteristically distinguished from one another.

§ 5. Detailed investigation of certain typical systems and general discussion

In the preceding section we have investigated the temperature and field dependences of the magnetization, the susceptibility and the heat capacity, not quantitatively but rather qualitatively. In this section we calculate these quantities numerically as functions of the magnetic field and temperature for several typical values of the coupling ratio α. We choose $\alpha=1.04$, 0.6, 0.2 and -0.2 in discussing the field dependences of the quantities and as $\alpha=0.6$ and -0.2 in discussing their temperature dependences. By considering Figs. 5, 6, 7 and 8, we see that these ratios are suitable.

First we show the field dependences of the magnetization, the susceptibility and the heat capacity at different temperatures in Figs. 9, 10, 11 and 12 corresponding to the cases $\alpha=1.04$, 0.6, 0.2 and -0.2 respectively. We next show the temperature dependences of these quantities at different field strengths in Figs. 13 and 14 for $\alpha=0.6$ and -0.2 respectively.

Many of the curves possess characteristics of what is called metamagnetism. For example one can recognize sharp rises in some of the magnetization-field curves which indicate a certain kind of transition from antiferro- to ferro-magnetic configurations, as were observed by many anthors, e.g. in $FeCl_2$,[13] $FeCo_3$,[14] $FeBr_2$,[15] rare-earth copper compounds,[16] $HgCr_2S_4$ spinel[17] and so on.

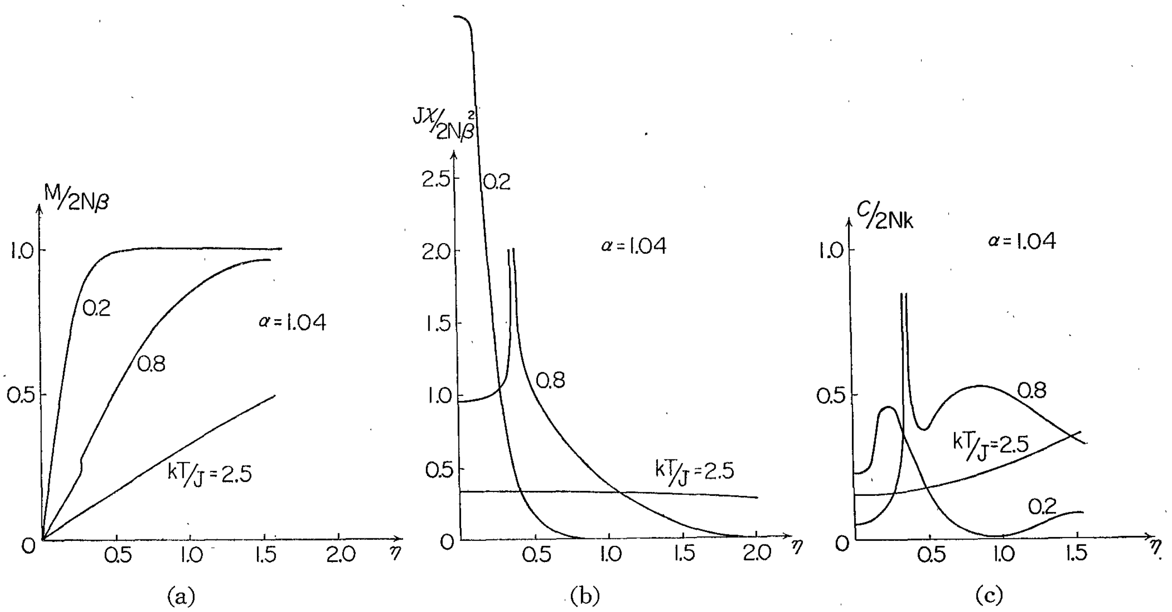
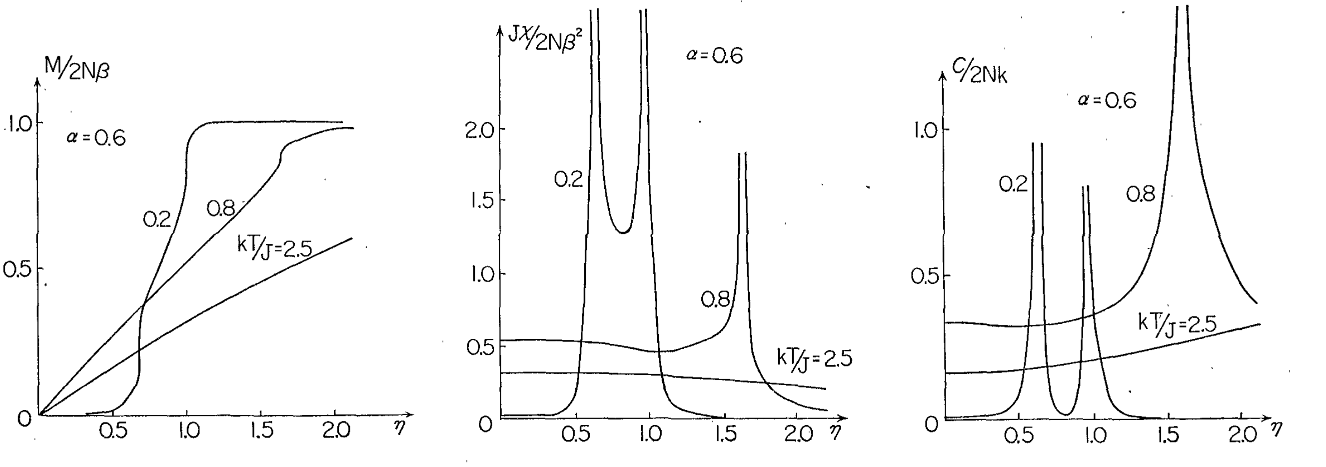
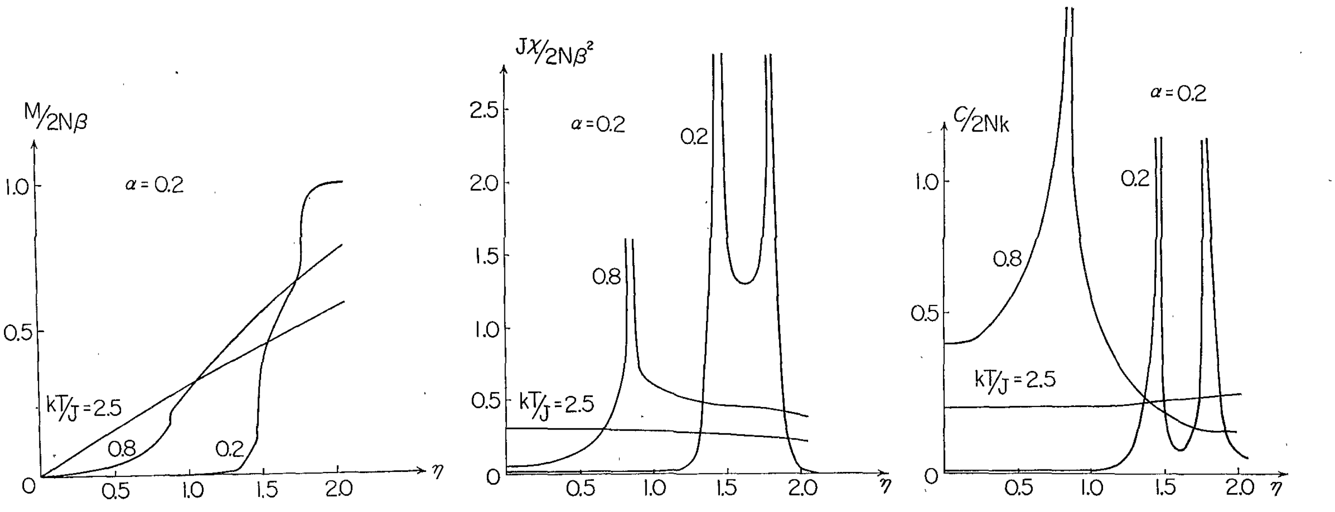


Fig. 9. Relations of the magnetization M, the susceptibility χ and the heat capacity C with the field parameter η in the case $\alpha=1.04$.



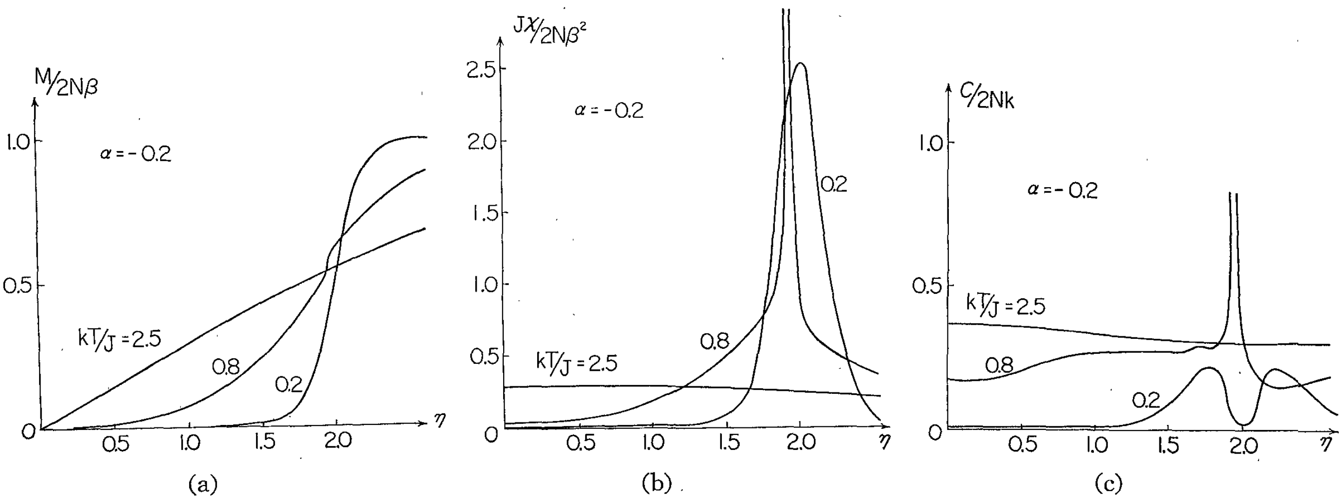
(a) (b) (c)

Fig. 10. Relations of the magnetization M, the susceptibility χ and the heat capacity C with the field parameter η in the case $\alpha=0.6$.



(a) (b) (c)

Fig. 11. Relations of the magnetization M, the susceptibiltiy χ and the heat capactiy C with the field parameter η in the case $\alpha=0.2$.



(a) (b) (c)

Fig. 12. Relations of the magnetization M, the susceptibility χ and the heat capacity C with the field parameter η in the case $\alpha=-0.2$.

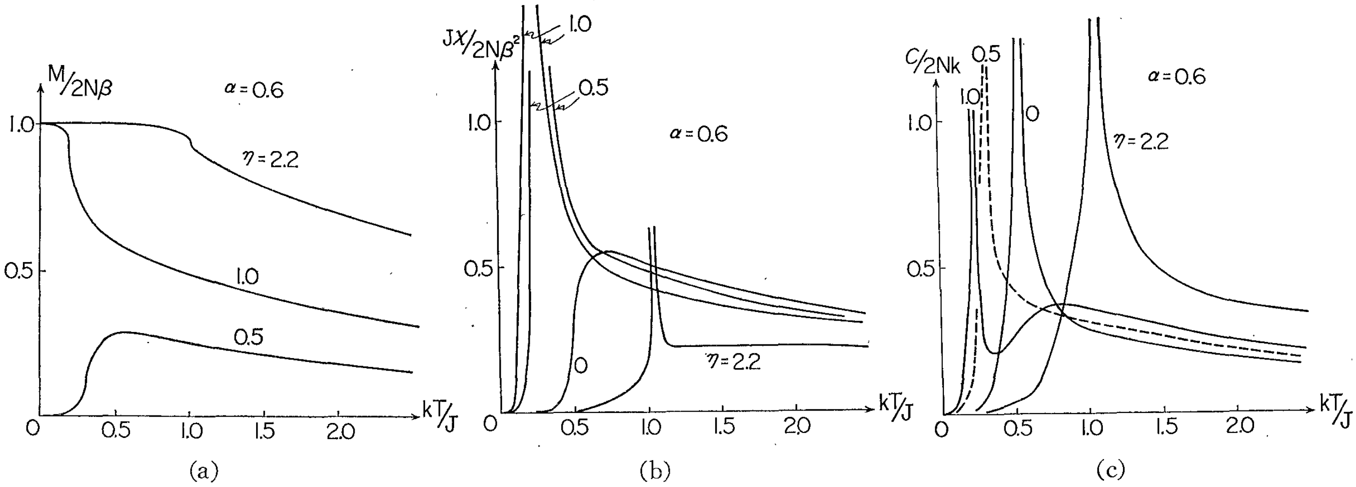


Fig. 13. Relations of the magnetization M, the susceptibility χ and the heat capacity C with temperature in the case $\alpha=0.6$.

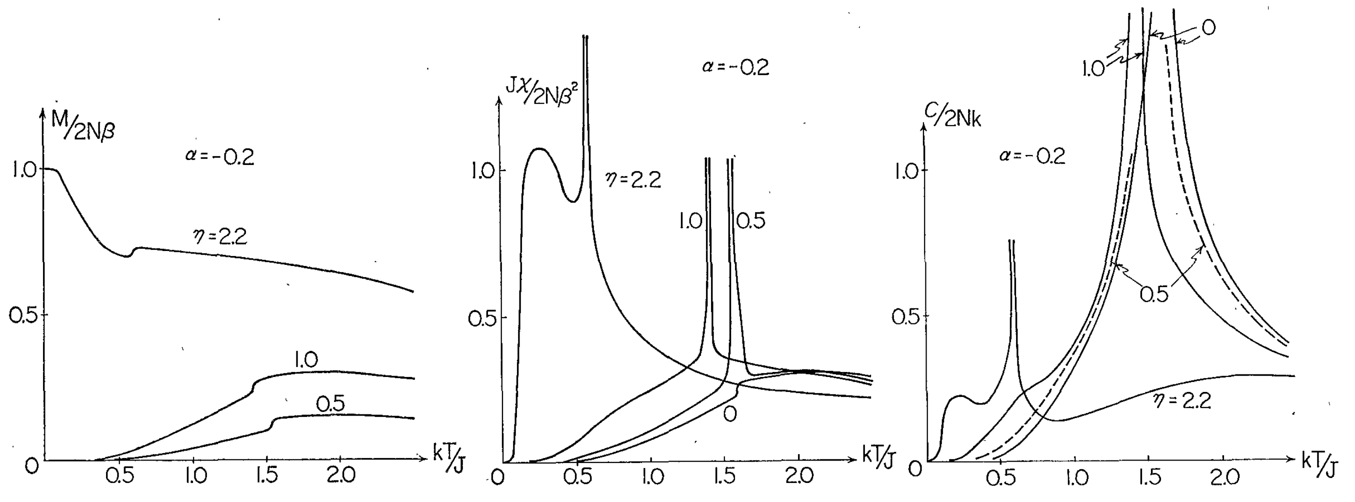


Fig. 14. Relations of the magnetization M, the susceptibility χ and the heat capacity C with temperature in the case $\alpha=-0.2$.

Baltzer et al.[17] have discussed the magnetism of chromium chalcogenide spinels including the compound $HgCr_2S_4$ which shows metamagnetism, on the basis of the Hamiltonian of the system of spins which are distributed on the B sites in the spinel structure and couple not only with the nearest-neighbour spins but also with the second-neighbour spins. They have investigated the dependence of the magnetic character of the system on the exchange coupling ratio between the first and second neighbour pairs. Such a kind of investigation has been made recently also by Adachi and the present authors[18] in connection with the experimental research by Adachi et al.[19] on the series of compound $Co(S_xSe_{1-x})_2$ of various fraction x and they have made a numerical calculation in order to picture the magnetization-field curves at different temperatures, where one can see how the system changes from a metamagnetic to paramagnetic states with rising of temperature.

From the point of view how the system changes from metamagnetic to paramagnetic states with rising of temperature, the present calculation is quite similar to the above-mentioned works by Baltzer et al. and by Adachi and the present authors.

One can find certain two steps of sharp rising in some of the magnetization-field curves in the present paper, viz. in Figs. 10(a) and 11(a) both at the temperature $0.2J/k$. Such a magnetic behaviour has been found by Kobayashi and Haseda,[20] Narath and Barham[21] in the compound $CoCl_2 \cdot 2H_2O$ and by Narath[22] in the compounds $CoBr_2 \cdot 2H_2O$ and $FeCl_2 \cdot 2H_2O$. These compounds are constituted of a bundle of linear chains and isomorphous with one another. From the energetical point of view Oguchi and Takano,[23] Narath[24] and Kanamori[25] have shown the doubly-stepwise magnetization process can occur in certain Ising spin systems which may be the models of the above-mentioned compounds. Yamada and Kanamori[26] have extended the discussion to finite temperatures by making use of the Weiss approximation. The present model can be regarded as a simplest system which is shown, on the basis of an exact calculation, to have the same sort of magnetization properties.

6. Conclusion

We have investigated the magnetic and thermal properties of the decorated Ising lattice, especially that of the square lattice, which is designed to allow the exact calculation according to Fisher.[4] If we take such a decorated Ising lattice that it possesses three transition temperatures, we shall find a more interesting character in the magnetic and thermal properties of the system. The investigation of such a system will be reported in the near future.

Appendices

1. *On the critical field as a function of temperature*

One can see from the expression for x in (4) in the main text that x tends, in the limit $J'=\infty$ (viz. absolute zero of temperature), to $-\alpha J'$ if $\eta>2$, to $-\alpha J'+(\ln 2)/4$ if $\eta=2$, to $[1-\alpha-(\eta/2)]J'$ if $2>\eta>0$ and to $(1-\alpha)J'-(\ln 2)/2$ if $\eta=0$. The solution $\eta=\eta_C$ of the equation $x=\pm x_C$ in the limit of zero temperature is thus found as $\eta_C=\eta_\pm=2(1-\alpha)\mp 2(x_C/J')=2(1-\alpha)\mp 2(kx_C/J)T$.

In the vicinity of the Néel temperature T_N (the critical temperature at vanishing field), η_C as well as $T-T_N$ are so small that one can expand $x(T, \eta_C)$ in a power series of these two small parameters. By solving the equation $x(T, \eta_C)=\pm x_C$ in which the above expansion is substituted for $x(T, \eta_C)$, we get

$$T-T_N=\frac{J}{4k}\left[1-\mathrm{sech}^2\left(\frac{2J}{kT_N}\right)\right]\left[\alpha-\tanh\left(\frac{2J}{kT_N}\right)\right]^{-1}\eta_C^2, \qquad \text{(A1)}$$

where we have made use of the facts that x is given by (4) in the text and that $x=x_C$, $(\partial x/\partial \eta)=0$ and $(\partial^2 x/\partial \eta \partial T)=0$ when $T=T_N$ and $\eta=0$. Then one can see that $T-T_N$ is written as $-c\eta_C^2$ and $+c\eta_C^2$ on the curves of the critical field $H_C=H_+$ and $H_C=H_-$ respectively, where c is a positive constant. This is due to the fact that the α on the curve ① in Fig. 5 as a function of T is larger than $\tanh(2J/kT)$ and that on the curve ② in that figure is smaller than $\tanh(2J/kT)$.

2. *Singularities of the magnetization and other quantities in the critical region*

It is already known[12] that the behaviour of the spin-pair correlation $U(x)$ given by (8) in the vicinity of the critical value $\pm x_C$ is expressed as

$$U(x)-U(\pm x_C)\sim-(x\mp x_C)\ln|x\mp x_C|. \tag{A2}$$

If one regards x as a function of the magnetic field with temperature fixed, one can write $x\mp x_C=x(H)-x(H_C)\simeq(\partial x/\partial H)_{H=H_\pm}(H-H_\pm)$ so far as the critical field $H_C=H_+$ or H_- is unequal to zero. By substituting this relation into (A2) one gets

$$U(x)-U(\pm x_C)\sim-(\partial x/\partial H)_{H=H_\pm}(H-H_\pm)\ln|H-H_\pm|\sim(H-H_\pm)\ln|H-H_\pm|. \tag{A3}$$

In the case that the critical field is equal to zero, $x-x_C\simeq(\partial^2 x/\partial H^2)_{H=0}H^2/2$ because $(\partial x/\partial H)_{H=0}=0$ as seen from (4) or Fig. 4. In this case (A2) can be rewritten as

$$U(x)-U(\pm x_C)\sim-(\partial^2 x/\partial H^2)_{H=0}H^2\ln H\sim H^2\ln H. \tag{A4}$$

We next consider x as a function of temperature by fixing the magnetic field constant. Then in the vicinity of the critical field, $x(T)-x(T_C)\simeq(\partial x/\partial T)_{T=T_C}\times(T-T_C)$, where $T_C=T_+$ or T_- should satisfy $x(T_\pm)=\pm x_C$ and is found as a function of the magnetic field. It is noted that $(\partial x/\partial T)_{T=T_C}$ never vanishes; that is, $(\partial x/\partial T)_{T=T_+}<0$ and $(\partial x/\partial T)_{T=T_-}>0$. Therefore we have $x-x_C=x(T)-x(T_\pm)\sim\mp(T-T_\pm)$ and can rewrite (A2) as

$$U(x)-U(\pm x_C)\sim\pm(T-T_\pm)\ln|T-T_\pm|. \tag{A5}$$

The singularities of the magnetization M, the magnetic susceptibility χ and the heat capacity C given by (7), (9) and (10) respectively are involved in the factors $U(x)$ and $\partial U(x)/\partial x$ which appear in these quantities; that is,

$$\begin{aligned}&M\sim(\partial x/\partial\eta)\,U(x),\\&\chi\sim(\partial x/\partial\eta)^2\partial U(x)/\partial x+(\partial^2 x/\partial\eta^2)\,U(x),\\&C\sim(\partial x/\partial J')^2\partial U(x)/\partial x.\end{aligned} \tag{A6}$$

By substituting (A3), (A4) and (A5), one can see the singularities of M, χ and C in the vicinity of the critical field and in the vicinity of the critical temperature. The results are shown in Tables I and II in the main text.

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