Theory of One-Dimensional Random Mixture of Ising Spins

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A general theory to obtain the magnetic properties of the one-dimensional random mixture of plural kinds of magnetic atoms (Ising spins) in the finite magnetic field is formulated in terms of distribution functions which give the probabilities of finding powers of average values of spins situated on the end site of the chain. The distribution functions are determined from the simultaneous functional equation of order \( n \), where \( n \) is the number of the kind of magnetic atoms. Using this theory, we study the magnetic properties of the following systems: (1) The regular system composed of only one kind of magnetic atoms, (2) the simple random system composed of one kind of magnetic atoms and non-magnetic atoms, (3) the random binary mixture composed of two kinds of magnetic atoms of \( S=1/2 \), (4) the random binary mixture of two kinds of magnetic atoms of \( S=1/2 \) and \( S=1 \). An Ising chain with random exchange integrals is also formulated in terms of a distribution function, and the magnetic properties are investigated when \( S=1/2 \). It is shown that the magnetization curves of those mixtures at low temperatures have various steps when the antiferromagnetic elements are included, and the susceptibilities at \( H=0 \) and \( T \to 0 \) diverge when the ferromagnetic elements are included even if the magnetization vanishes when \( T=0 \) and \( H \to 0 \). Those are discussed in detail from the random nature of the mixtures.

§ 1. Introduction and summary

The random magnetic system which is composed of some kinds of magnetic atoms and non-magnetic atoms has been a subject of interest in past years.\(^1\)\(^-\)\(^10\) In previous papers, the one-dimensional simple random Ising system of spin \( S \) which is composed of one kind of magnetic atoms and non-magnetic atoms,\(^11\) and the one-dimensional random Ising mixture of plural kinds of magnetic atoms of spin \( 1/2 \) in vanishing magnetic field\(^13\)\(^,\)\(^14\) were studied. In this paper, a general method for studying the one-dimensional random Ising mixture of plural kinds of magnetic atoms in the finite magnetic field is given and is applied to some systems to obtain some exact aspects of random systems.

In § 2, a method of the distribution function for the problem of the one-dimensional random mixture is developed with an example of a random binary mixture of two kinds of magnetic atoms of \( S=1/2 \), and the magnetic properties of the mixture are investigated. In § 2-1, the mathematical formulation for the random binary mixture is given in terms of distribution functions \( g_\alpha(x) \) which gives the probability of finding the average value \( x \) of the spin of \( \alpha \) atom situated on the end site of the chain. The energy and the magnetization are expressed by using the distribution functions. Following a similar procedure...
of obtaining the matrix method for the regular Ising chain, the simultaneous functional equation for the distribution functions is obtained.

Special cases are examined in §§ 2-2 and 2-3. In § 2-2, we consider the regular system composed of one kind of magnetic atoms. The functional equation is exactly solved. The distribution function is described by $\delta$ function, which means that the average value of the spin on the end site approaches some stationary value. The energy and the magnetization in the finite magnetic field are analytically obtained and they are completely in agreement with the well-known results. In § 2-3, we consider a simple random system. The solution of the equation is described as a superposition of $\delta$ functions. The energy and the magnetization of this system are given in terms of power series of concentration of magnetic atoms, whose coefficients are obtained from recurrence relations. These are also in agreement with previous results.

In § 2-4, a procedure of the low field expansion is given. The energy and the magnetization of the mixture are expressed in terms of power series of the magnetic field and the configurational averages of powers of average values $y_a^{(\alpha)}(=\int x^\alpha g_a(x)dx)$. Using the functional equations, we obtain the simultaneous linear equation for $y_a^{(\alpha)}$ whose coefficients are obtained from recurrence relations. The first two terms of the power series of the energy and the magnetization are calculated. The first terms of them are in agreement with the previous results.

The behavior of the magnetization process is a subject of interest in the random mixture. It was shown that the antiferromagnetic magnetization of the simple random system has three steps when the temperature is low. In § 2-5, we numerically study the magnetization processes of the random binary mixture. The magnetization curve has plural steps when the antiferromagnetic elements are included. These are discussed in detail from the random nature of the mixture.

The formulation is extended to the system composed of $n$ kinds of magnetic atoms with arbitrary spins in § 3. For each kind of atoms of spin $S$, a $2S$-dimensional distribution function $g_a(x_1, x_2, \cdots, x_{2n})$ is introduced. The simultaneous functional equation of order $n$ for the distribution functions is also obtained. The energy and the magnetization are expressed by using the distribution functions. The susceptibility of a random binary mixture of two kinds of atoms $S=1/2$ and $S=1$ is investigated as an example.

The problem of the random exchange integrals is also studied in § 4. Since the individualities of the magnetic atoms are lost in this case, it is enough for us to define one distribution function $g(x_1, x_2, \cdots, x_{2n})$. A similar functional equation can be obtained. The magnetic properties of this system are investigated when $S=1/2$. The method of low field expansion is given and the first two terms of the energy and the magnetization are obtained. We numerically calculate the magnetization of the binary mixture of two kinds of exchange integrals in various cases. The magnetization curves also have several steps when the antiferro-
magnetic elements are included. Finally, it is to be noted that the idea of the distribution function for the problem of the random mixture is essentially important not only in the one-dimensional mixture but also in the two- or three-dimensional mixture. It is the point of this idea that it enables us to take into account the effects caused by the random arrangements of magnetic atoms. Using this idea, we can improve the approximation theories such as the molecular field approximation and the Bethe approximation. In fact, we can easily show that the formulation for the one-dimensional lattice obtained by the improved Bethe approximation is completely in agreement with the exact one obtained in this paper. This corresponds to the fact that the Bethe approximation for the regular system gives exact results when the system is one-dimensional. These improvements are especially important when the effective fields which act on the spins on the lattice sites are not small as in the cases that the system is in the ordered state and that the external magnetic field is finite. These will be reported in future.

§ 2. Random mixture of two kinds of magnetic atoms of $S=1/2$

In this section, a method of the distribution function for the problem of the one-dimensional random mixture of plural kinds of Ising spins is developed using a simple random mixture composed of two kinds of magnetic atoms of $S=1/2$ as an example. The method will be generalized in § 3, and will be applied to the problem of the random exchange integrals in § 4.

2.1) Mathematical formulation

We consider a linear chain of $N$ lattice sites where two kinds of magnetic atoms A and B are randomly frozen on the sites, which are named from 1 to $N$ along the chain from left to right. Ising spins $\sigma_i = \pm 1$ are associated with both A and B atoms. The magnetic moments of A and B atoms are denoted by $m_A$ and $m_B$, the exchange integrals between them by $J_{AA}$, $J_{BB}$ and $J_{AB}$ ($=J_{BA}$), and concentrations of them by $p_A$ and $p_B$ ($p_A+p_B=1$).

The average value of the spin of $\alpha$ atom on $i$-th site, which is sandwiched between $\beta$ atom on $(i-1)$-th site and $\gamma$ atom on $(i+1)$-th site, is given by

$$\langle \sigma_i \rangle = \frac{\sum \sigma e^{-S/kT}}{\sum e^{-S/kT}},$$

where $\mathcal{H}$ is the Hamiltonian of the chain and $\alpha, \beta, \gamma = A$ or $B$. It is easily shown that the average value is rewritten as

$$\sigma_\alpha(x_\beta; x_\gamma) = \langle \sigma_i \rangle$$

$$= \frac{\tanh C_\alpha + x_\beta \tanh K_{\beta\alpha} + x_\gamma \tanh K_{\gamma \alpha} + x_\beta x_\gamma \tanh K_{\beta\gamma} \tanh K_{\gamma\beta} \tanh K_{\alpha\beta} \tanh K_{\alpha\gamma} \tanh C_\alpha}{1 + (x_\beta \tanh K_{\beta\alpha} + x_\gamma \tanh K_{\gamma\alpha}) \tanh C_\alpha + x_\beta x_\gamma \tanh K_{\beta\gamma} \tanh K_{\gamma\beta} \tanh K_{\alpha\beta} \tanh K_{\alpha\gamma}},$$

(2.2)
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where $K_{a\beta} = J_{a\beta}/2kT$, $C_a = m_aH/kT$ and $H$ is the magnetic field. The $x_\beta$ and $x_r$ are the average values of the spins of $\beta$ atom on $(i-1)$-th site and $\gamma$ atom on $(i+1)$-th site when the $\alpha$ atom on $i$-th site is removed. This means that these values are the average values of the spins on the end sites of the chains with given configurations of atoms. The exchange energy between $\alpha$ and $\beta$ atoms is similarly written as

$$\varepsilon = -\frac{1}{2} J_{a\beta} \frac{(1 + x_\alpha x_\beta) e^{K_{a\beta}} - (1 - x_\alpha x_\beta) e^{-K_{a\beta}}}{(1 + x_\alpha x_\beta) e^{K_{a\beta}} + (1 - x_\alpha x_\beta) e^{-K_{a\beta}}}.$$  \hfill (2.3)

Next we consider the $\alpha$ atom on the end site named 1st site which neighbors with $\beta$ atom on the 2nd site. The average value of the spin of $\alpha$ atom, $x_\alpha$, is given as

$$x_\alpha = \frac{\tanh C_a + x_\beta \tanh K_{a\beta}}{1 + x_\beta \tanh K_{a\beta} \tanh C_a}.$$  \hfill (2.4)

Equation (2.4) is rewritten as

$$x_\beta = \frac{x_\alpha - \tanh C_a}{\tanh K_{a\beta} (1 - x_\alpha \tanh C_a)}.$$  \hfill (2.5)

This shows that the average value $x_\beta$ is also related to $x_\alpha$ by Eq. (2.5) irrespective of other information of the chain. Then we define the function

$$f_{a\beta}(x) = \frac{x - \tanh C_a}{\tanh K_{a\beta} (1 - x \tanh C_a)}.$$  \hfill (2.6)

This function gives the average value of the spin of $\beta$ atom on the end site of a chain which gives the average value $x$ for the spin of additional $\alpha$ atom on the next site.

Now we consider the random mixture where $x_\alpha$ are random variables corresponding to the random arrangement of atoms. Then we introduce distribution functions $g_A(x)$ and $g_B(x)$ which give the probabilities of finding the average value $x$ for the spin of A atoms and B atoms, respectively. The distribution functions are normalized as

$$\int_{-1}^{1} g_A(x) dx = 1, \quad \alpha = A, B.$$  \hfill (2.7)

Using these distribution functions, we have the energy and the magnetization per lattice site as follows:

$$\varepsilon = \sum_\alpha \sum_\beta p_\alpha p_\beta \int \int \varepsilon(x_\alpha; x_\beta) g_\alpha(x_\alpha) g_\beta(x_\beta) dx_\alpha dx_\beta - \langle m\sigma \rangle H,$$  \hfill (2.8)

$$\langle m\sigma \rangle = \sum_\alpha \sum_\beta \sum_\gamma m_\alpha p_\alpha p_\beta p_\gamma \int \int \sigma_\alpha(x_\beta; x_\gamma) g_\beta(x_\beta) g_\gamma(x_\gamma) dx_\beta dx_\gamma.$$  \hfill (2.9)

To determine the distribution functions, we consider an average value of the spin on the end site of an infinitely large chain where concentrations of A...
and B atoms are given by \( p_A \) and \( p_B \), respectively. The probabilities of finding A and B atoms on the end site named the 1st site are given by \( p_A \) and \( p_B \), and the probabilities of finding the average value of the spin in the interval \((x, x + \Delta x)\) are given by \( g_A(x) \Delta x \) and \( g_B(x) \Delta x \), respectively. Next, add an \( \alpha \) atom to the next site named the 0-th site. The probability of finding the average value of the spin of the atom in the interval \((x, x + \Delta x)\) is given in two ways; (1) \( g_\alpha(x) \Delta x \), because the chain is assumed to be infinitely large and (2) \( \sum_\beta p_\beta g_\beta(f^{\alpha\beta}(x)) (d/dx)f^{\alpha\beta}(x) \Delta x \), because the interval \((x, x + \Delta x)\) at 0-th site with \( \alpha \) atom is projected from the interval \((f^{\alpha\beta}(x), f^{\alpha\beta}(x) + (d/dx)f^{\alpha\beta}(x) \Delta x)\) at 1st site with \( \beta \) atom. Then we have

\[
g_\alpha(x) \Delta x = \sum_\beta p_\beta g_\beta(f^{\alpha\beta}(x)) \frac{d}{dx} f^{\alpha\beta}(x) \Delta x. \tag{2.10}
\]

Thus we obtain a simultaneous functional equation

\[
g_\alpha(x) = \sum_\beta p_\beta \frac{d}{dx} f^{\alpha\beta}(x) g_\beta(f^{\alpha\beta}(x)), \quad \alpha = A, B. \tag{2.11}
\]

If we define the function \( G_\alpha(x) \) as

\[
G_\alpha(x) = \int_{-\infty}^{x} g_\alpha(x') dx'. \tag{2.12}
\]

with

\[
\begin{align*}
G_\alpha(-1) &= 0, \\
G_\alpha(1) &= 1.
\end{align*} \tag{2.13}
\]

we have

\[
G_\alpha(x) = \sum_\beta p_\beta G_\beta(f^{\alpha\beta}(x)), \quad \alpha = A, B. \tag{2.14}
\]

These functions give the ratios of finding the average value of the spin on the end site in the region \((-1, x)\).

Hence, we can obtain the energy and the magnetization by using Eqs. (2.6), (2.8), (2.9) and (2.11) or (2.14) with Eq. (2.12). We study the magnetic properties of this binary mixture in the next sections.

2.2) Regular system

We first consider the regular system where \( p_A = 1 \). Equation (2.14) is

\[
G_A(x) = G_A(f^{AA}(x)) \tag{2.15}
\]

with

\[
\begin{align*}
G_A(-1) &= 0, \\
G_A(1) &= 1.
\end{align*} \tag{2.16}
\]
We consider the graph shown in Fig. 1. The \( x_0 \) is the root of equation

\[ f^{AA}(x) = x. \]  

Since the function \( f^{AA}(x) \) is a monotonically increasing function of \( x \) and Eq. (2.17) has only one root, the solution of Eq. (2.15) is given by

\[ G_A(x) = \theta(x - x_0), \]  

where \( \theta(x) \) is a step function defined by

\[ \theta(x) = \begin{cases} 1 & \text{for } x > 0, \\ 0 & \text{for } x < 0. \end{cases} \]  

Hence we obtain

\[ g_A(x) = \delta(x - x_0). \]  

This means that in the regular system the average value of the end spin has a stationary value \( x_0 \). Inserting (2.20) into Eqs. (2.8) and (2.9), we obtain

\[ \varepsilon = -\frac{1}{2} J^{AA} \left( (1 + x_0^2) e^{K^{AA}} - (1 - x_0^2) e^{-K^{AA}} \right) - \langle m\sigma \rangle H \]  

and

\[ \langle m\sigma \rangle = m_A \tanh C_A + x_0 \tanh K^{AA} + x_0^2 \tanh^2 K^{AA} \tanh C_A \]

These reproduce the well-known results for the regular system.\(^{14}\)

### 2.3) Simple random system

Next, consider the simple random system where \( B \) atoms are non-magnetic, i.e., \( C_B = K_{BB} = K_{AB} = 0 \). The average value of the spin of \( A \) atom at the end site which neighbors with \( B \) atom is simply given by \( \tanh C_A \) without use of Eq. (2.4). Then, Eq. (2.11) is modified as

\[ \varepsilon' = p_A \frac{d}{dx} f^{AA}(x) g_A(f^{AA}(x)) + p_B \delta(x - \tanh C_A) \]  

or Eq. (2.14) as

\[ G_A(x) = p_A G_A(f^{AA}(x)) + p_B \theta(x - \tanh C_A). \]  

The solution of Eq. (2.24) is

\[ G_A(x) = p_B \sum_{i=1}^{\infty} p_A^{-i} \theta(x - x_i), \]  

where
\[ x_i = \tanh C_A, \]
\[ x_{i+1} = f^{AA}(x_i), \quad i \geq 1, \]  
\[ \text{(2.26)} \]

where \( f^{ab}(x) \) is the inverse function of \( f^{ab}(x) \). Then we obtain

\[ g_A(x) = p_B \sum_{i=1}^{\infty} p_{A,i}^{-1} \delta(x - x_i). \]  
\[ \text{(2.27)} \]

Inserting (2.27) into Eqs. (2.8) and (2.9), we obtain

\[ \langle m\sigma \rangle = m_A p_B (\sum_{i=1}^{\infty} \sum_{j=1}^{\infty} p_{A,i} f_{A,j}^{+1} + 1 + x_i x_j) \tanh C_A + x_i x_j \tanh C_A = \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} p_{A,i} f_{A,j}^{+1} \]  
\[ \frac{\tanh C_A + x_i x_j \tanh C_A + x_i x_j \tanh C_A}{1 + x_i x_j \tanh C_A + x_i x_j \tanh C_A}, \]  
\[ \text{(2.28)} \]

where \( x_0 = 0 \), and

\[ \varepsilon = -\frac{1}{2} J_{AA} p_B (\sum_{i=1}^{\infty} \sum_{j=1}^{\infty} p_{A,i} f_{A,j}^{+1} \tanh C_A + x_i x_j \tanh C_A + x_i x_j \tanh C_A) - \langle m\sigma \rangle H. \]  
\[ \text{(2.29)} \]

These are in agreement with the previous exact results.  

2.4) The case \( H \sim 0 \)

In this section the low field expansion is given. Since the thermal average \( x_a \) is of order \( \tanh C_A \), we can expand Eqs. (2.8) and (2.9) as

\[ \langle m\sigma \rangle = \sum_a \sum_{j=1}^{\infty} m_a p_a p_B (\sum_{i=1}^{\infty} \sum_{j=1}^{\infty} (-1)^j \left( \begin{array}{c} i \\ j \end{array} \right) \tanh C_A + x_i x_j \tanh C_A) \]
\[ \times \{ \tanh a(y \tanh K_{ab})^{(l-j-k)} (y \tanh K_{ab})^{(l-k)} + (y \tanh K_{ab})^{(l-j+k+1)} \tanh a(y \tanh K_{ab})^{(l-j-k)} (y \tanh K_{ab})^{(l-k+1)} \tanh a(y \tanh K_{ab})^{(l-j+k+1)} (y \tanh K_{ab})^{(l-k+1)} \} \]  
\[ \text{(2.30)} \]

and

\[ \varepsilon = -\frac{1}{2} \sum_a \sum_{j=1}^{\infty} J_{ab} p_a p_B (\tanh K_{ab} - 2 \cosech 2K_{ab}) \]
\[ \times \sum_{i=1}^{\infty} (-1)^j g_a^{(i)} g_b^{(i)} \tanh K_{ab} - \langle m\sigma \rangle H, \]  
\[ \text{(2.31)} \]

where

\[ (a y)^{\delta} = \int (a x)^y g_a(x) \, dx = a^y g_a^{(0)}. \]  
\[ \text{(2.32)} \]

The configurational average \( y_a^{(0)} \) are obtained in the following way. From Eq. (2.11), we have

\[ y_a^{(n)} = \sum_p p_p \int x^n \frac{d}{dx} f^{ab}(x) g_b(f^{ab}(x)) \, dx \]
\[ = \sum_p p_p \int \{ f^{ab}(x) \}^n g_b(x) \, dx. \]  
\[ \text{(2.33)} \]
Next, define $a_{\alpha,\beta,t}$ by

$$\int \{ f^{(\alpha\beta)}(x) \} \gamma^a(x) dx = \sum_{t=0}^n a_{\alpha,\beta,t} y^{(t)}_\alpha. \quad (2.34)$$

Then we have a simultaneous linear equation

$$y^{(n)}_\alpha = \sum_{\beta} \sum_t p_{\beta} a_{\alpha,\beta,t} y^{(t)}_\beta. \quad (2.35)$$

The coefficients $a_{\alpha,\beta,t}$ are determined from recurrence relations. Consider

$$\int \{ f^{(\alpha\beta)}(x) \}^{n+1} g^\beta(x) dx = \sum_{t=0}^n a_{\alpha,\beta,t} y^{(t)}_\beta = \int \{ f^{(\alpha\beta)}(x) \} \sum_{t=0}^n a_{\alpha,\beta,t} x^t g^\beta(x) dx$$

$$= \sum_{t=0}^n \sum_{k=0}^n (-1)^k \tanh^k C_a \tanh^k K_{a\beta} \{ \tanh C_a a_{\alpha,\beta,t-k} + \tanh K_{a\beta} a_{\alpha,\beta,t-k-1} \} y^{(t+k)}_\beta. \quad (2.36)$$

Then we have

$$a_{\alpha,\beta,t}^{n+1} = \sum_{k=0}^{t} (-1)^k \tanh^k C_a \tanh^k K_{a\beta} \{ \tanh C_a a_{\alpha,\beta,t-k} + \tanh K_{a\beta} a_{\alpha,\beta,t-k-1} \},$$

$$a_{\alpha,\beta,t}^n = 0 \quad \text{for} \quad i \leq -1. \quad (2.37)$$

Equation (2.37) is a recurrence formula for $a_{\alpha,\beta,t}$ with initial terms

$$a_{\alpha,\beta,t}^0 = \delta_{t,1}, \quad a_{\alpha,\beta,0} = \tanh C_a,$$

$$a_{\alpha,\beta,t}^1 = (-1)^{t-1} \tanh^{t-1} C_a \tanh^t K_{a\beta} (1 - \tanh^2 C_a), \quad t \geq 1. \quad (2.38)$$

Thus, we can obtain the low field expansions for the magnetization and the energy of the random mixture.

The first two terms of them are given by

$$\mathcal{E} = -\frac{1}{2} \sum_{\alpha} \sum_{\beta} J_{a\beta} p_{\alpha} p_{a\beta} t_{a\beta} - \left\{ \frac{1}{2} \sum_{\alpha} \sum_{\beta} J_{a\beta} p_{\alpha} p_{a\beta} (1 - t_{a\beta}^2) y^{(1)}_a y^{(1)}_\beta \right\} + \langle m H (\tanh C + 2 \mathcal{F}) \rangle \quad (2.39)$$

and

$$\langle m \sigma \rangle = \langle m (\tanh C + 2 \mathcal{F}) \rangle - 2 \langle m \mathcal{F} \rangle \tanh^2 C \quad + \langle m (\Phi + \mathcal{F}^2) \tanh C \rangle + \langle m \Phi \mathcal{F} \rangle \}, \quad (2.40)$$

where

$$\langle A \rangle = \sum_{\alpha} p_{\alpha} A(\alpha), \quad t_{a\beta} = \tanh K_{a\beta} \quad (2.41)$$

and

$$\mathcal{F}_a = \sum_{\beta} p_{\beta} y^{(1)}_a t_{a\beta}, \quad \Phi_a = \sum_{\beta} p_{\beta} y^{(1)}_a t_{a\beta}^2 \quad (2.42)$$

$y^{(1)}_a$ and $y^{(2)}_a$ are given in the Appendix. The first terms of Eqs. (2.39) and (2.40)
are in agreement with the previous results.\textsuperscript{10,11}

2-5) Magnetization processes

The magnetization process of the random system is of great interest. It was shown that the magnetization per magnetic atom ($\langle m_0 \rangle / \rho$) of the ferromagnetic simple random magnetic system increases with concentration of magnetic atoms $\rho$, and the magnetization curve of the antiferromagnetic simple random magnetic system has three steps when the temperature is sufficiently low.\textsuperscript{11} These are due to the behavior of the magnetic chains in the random system.

In this section, we investigate the magnetization processes of the random binary mixtures by using Eqs. (2.9) and (2.11). The simultaneous functional equation is numerically solved (see Fig. 2). Using these results, we can numerically obtain the magnetization and the energies of the random mixtures. The magnetization curves are shown in Figs. 3~5. It is interesting to observe that these curves have also various steps when the temperature is sufficiently low. To explain these magnetization processes, we consider the following four cases assuming $m_A = m_B$.

(1) The case $J_{AA}, J_{BB}, J_{AB} > 0$. The magnetization at a sufficiently low tempera-

![Image of distribution functions](https://example.com/distribution.jpg)

Fig. 2. The distribution functions $g_A(x)$ and $g_B(x)$, when $J_{AA} = 2J$, $J_{BB} = J$, $J_{AB} = 0.5J$ ($J < 0$), $m_A = m_B$, $\rho_A = 0.6$, $\rho_B = 0.4$, and $kT/|J| = 0.3$.

![Image of magnetization curves](https://example.com/magnetization.jpg)

Fig. 3. The magnetization of the binary mixture of $J_{AA} = 2J$, $J_{BB} = J$, $J_{AB} = 0.5J$ ($J < 0$), $m_A = m_B$, $\rho_A = 0.6$, and $\rho_B = 0.4$. 

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ture saturates with infinitesimal magnetic field. The magnetization process of this mixture at finite temperatures resembles that of the regular ferromagnetic system with an effective exchange integral and an effective magnetic moment.

(2) The case $J_{AA}, J_{BB}, J_{AB} < 0$ (see Figs. 3 and 4). The ground state of this mixture is antiferromagnetic when $H \sim 0$. Antiparallel spins turn over when the magnetic field increases. Now, we consider an antiparallel spin of $\alpha$ atom sandwiched between two parallel spins of $\beta$ and $\gamma$ atoms. The probability of finding this clusters is $p_{\alpha}p_{\beta}p_{\gamma}/2$. If the antiparallel spin turns over, the energy increases as much as
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\[ \Delta E = |J_{ab}| + |J_{ar}| - 2m_a H. \]  

(2.43)

Then we find a step at

\[ H = \frac{1}{2m_a} (|J_{ab}| + |J_{ar}|). \]  

(2.44)

The height of the step is \( p_a p_b p_r \). The susceptibility decreases when the temperature tends to zero, and the magnetization curve has in general six steps when the temperature is sufficiently low.

(3) The case \( J_{AA}, J_{BB} > 0 \) and \( J_{AB} < 0 \) (see Fig. 5). The ground state is one where all spins of A atoms are aligned in parallel to the magnetic field and all spins of B atoms are aligned in antiparallel, when \( H \sim 0 (p_A \geq p_B) \). Since the local concentration of each kind of magnetic atoms is random, the response of the magnetization for the magnetic field is very sensitive when \( H \sim 0 \). The part where almost all lattice sites are occupied by B atoms turns over as soon as the magnetic field is applied. (The critical field is given by \( H \sim |J_{ab}| / (2N_B - N) \), where \( N \) is the number of the lattice sites of the part and \( N_B \) is the number of B atoms.) Then the magnetization per lattice site of this mixture is given as \( \langle m \sigma \rangle = m (p_A - p_B) \) when \( H = 0 \), and it increases abruptly with \( H \) when \( H \sim 0 \). The susceptibility diverges even if \( p_A = p_B \) when the temperature tends to zero. On the other hand we find remarkable steps in the magnetization curve in the high field region, which is due to the flips of several spins.

(4) The case \( J_{AA}, J_{BB} < 0 \) (see Fig. 5). The ground state is also decided. The magnetization vanishes when \( H = 0 \). Since the local magnetization of this mixture is also at random, the magnetization increases abruptly with \( H \) when \( H \sim 0 \). Then the susceptibility diverges when the temperature tends to zero. Remarkable steps found in the magnetization curves in the high field region are also due to the behavior of spins.

We can also discuss the magnetization processes in other cases. Interesting properties have been found in the mixtures of ferromagnetic and antiferromagnetic elements. These properties will be also found in an Ising chain with random exchange integrals (see § 4).

§ 3. Generalization of the formulation

In this section we consider a random mixture composed of \( n \) kinds of magnetic atoms of spin \( S \). Since the average value of the spin and the correlation of spins on any lattice sites are expressed in terms of average values of powers of end spins \( \langle (S_i^j)^{\gamma_i} \rangle, i = 1, 2, \cdots, 2S \rangle \) the magnetization and the energy of this mixture can also be obtained by using distribution functions having \( 2S \) variables \( x_1 (\equiv \langle (S_i^j)^{\gamma_i} \rangle) \).

Now, we consider \( \beta \) atom on the end site, the average values of which are expressed by \( (x_1^\prime, x_2^\prime, \cdots, x_{2s^\prime}) \). Consider the addition of \( \alpha \) atom on the next lattice site. The average values \( (x_1, x_2, \cdots, x_{2s}) \) of that \( \alpha \) atom and \( (x_1^\prime, x_2^\prime, \cdots, \)
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The relations

\[ x_i = f_i(x_1, x_2, \ldots, x_n), \quad i = 1, 2, \ldots, n \]  

(3.1)

Next, consider the volume element \( \Delta v = \Delta x_1 \Delta x_2 \cdots \Delta x_n \) on the point \( (x_1, x_2, \ldots, x_n) \). The volume element is projected from the volume element

\[ \Delta v' = \left| \frac{\partial (f_{1}^{\alpha}, f_{2}^{\alpha}, \ldots, f_{n}^{\alpha})}{\partial (x_1, x_2, \ldots, x_n)} \right| \Delta v \]  

(3.2)

on the point \( (x_1', x_2', \ldots, x_n') \). Then we have a simultaneous functional equation

\[ g_\alpha(x_1, x_2, \ldots, x_n) = \sum_{\beta=1}^{n} p_\beta \left| \frac{\partial (f_{1}^{\alpha}, f_{2}^{\alpha}, \ldots, f_{n}^{\alpha})}{\partial (x_1, x_2, \ldots, x_n)} \right| g_\beta(f_{1}^{\alpha}, f_{2}^{\alpha}, \ldots, f_{n}^{\alpha}), \]

\[ \alpha = 1, 2, \cdots, n, \]  

(3.3)

where \( g_\alpha(x_1, x_2, \ldots, x_n) \) is the distribution function for \( \alpha \) atoms and is normalized as

\[ \int g_\alpha(x_1, x_2, \ldots, x_n) \, dv = 1. \]  

(3.4)

It we define

\[ G_\alpha(x_1, x_2, \ldots, x_n) = \int_{x_1}^{x_2} \int_{x_2}^{x_3} \cdots \int_{x_n}^{x_1} g_\alpha(x_1', x_2', \ldots, x_n') \, dv', \]  

(3.5)

we have

\[ G_\alpha(x_1, x_2, \ldots, x_n) = \sum_{\beta=1}^{n} p_\beta G_\beta(f_{1}^{\beta}, f_{2}^{\beta}, \ldots, f_{n}^{\beta}). \]  

(3.6)

The magnetic quantities \( A \) are given by

\[ A/N = \sum_\alpha \sum_\beta p_\alpha p_\beta \int \int A(x_1, x_2, \ldots, x_n; x_1', x_2', \ldots, x_n') \]

\[ \times g_\alpha(x_1, x_2, \ldots, x_n) g_\beta(x_1', x_2', \ldots, x_n') \, dv \, dv'. \]  

(3.7)

It is not difficult to extend our formulation to the random mixture composed of different kinds of spins. In those systems, the \( 2S \)-dimensional distribution function is introduced for each kind of magnetic atoms of spin \( S \). The distribution functions are also determined from the simultaneous functional equation including \( \partial \) functions and integrals.

For example, the simultaneous functional equation for a random binary mixture of \( A \) atoms of \( S = 1 \) and \( B \) atoms of \( S = 1/2 \) is given as follows:

\[ g_A(x_1, x_2) = p_A \left| \frac{\partial (f_{1}^{A}, f_{2}^{A})}{\partial (x_1, x_2)} \right| g_A(f_{1}^{A}, f_{2}^{A}) + p_B \frac{\partial f_{1}^{AB}}{\partial x_1} \delta(x_2 - a(x_1)) g_B(f_{1}^{AB}), \]  

(3.8)

\[ g_B(x) = p_A \lim_{\Delta x \to 0} \frac{1}{\Delta x} \int_{x'} g_A(x_1, x_1') \, dx_1' \, dx_2' + p_B \frac{\partial f_{2}^{AB}}{\partial x} g_B(f_{2}^{AB}), \]  

(3.9)

where \( \Delta s \) is the area closed between curves \( x = f_{2}^{A}(x_1', x_2') \) and \( x + \Delta x = f_{2}^{B}(x_1', x_2') \), and \( a(x) \) is the function determined from \( f_{1}^{AB}(x_1) = f_{2}^{AB}(x_2) (= x') \),
namely,

$$a(x) = \tilde{f}_1^{AB}(f_1^{AB}(x)).$$

(3.10)

It should be remarked here that Eqs. (3.8) and (3.9) are analytically treated when \( H = 0 \), and the energy and the susceptibility in vanishing magnetic field are obtained. The specific heat of this mixture has two or three maxima when the difference of exchange integrals is considerably large, and in general the susceptibility \( \chi \) diverges when the temperature tends to zero even if all exchange integrals were antiferromagnetic (see Fig. 6). These are also due to the randomness of distribution of magnetic atoms.

Fig. 6. The susceptibilities of binary mixtures of \( S = 1 \) and \( S = 1/2 \), when \( J_{AA} = J_{BB} = J_{AB} = J < 0 \), \( m_A = m_B \) and \( \rho_A + \rho_B = 1 \).

§ 4. The problem of random exchange integrals

In this section we consider an Ising chain of spin \( S \) with random exchange integrals. Since the kind of magnetic atoms is the same, the variables \( (x_1, x_2, \ldots, x_{2n}) \) and \( (x_1', x_2', \ldots, x_{2n}') \) are connected as follows:

$$x_1' = f_1^\alpha(x_1, x_2, \ldots, x_{2n}),$$

(4.1)

$$x_1 = f_1^\alpha(x_1', x_2', \ldots, x_{2n}'),$$

(4.2)

where the superscripts \( \alpha = 1, 2, \ldots, n \) indicate the kind of exchange integrals, and \( n \) is the number of them. In a way similar to § 3, we consider the volume element \( \Delta v' = \Delta x_1 \Delta x_2 \cdots \Delta x_{2n} \). The element is projected from the volume element

$$\Delta v = \frac{\partial (f_1^\alpha, f_2^\alpha, \ldots, f_{2n}^\alpha)}{\partial (x_1, x_2, \ldots, x_{2n})} \Delta v'$$

(4.3)

by exchange integral \( J_\alpha \). Then we also obtain functional equation

$$g(x_1, x_2, \ldots, x_{2n}) = \sum_{\alpha=1}^{n} \rho_\alpha \frac{\partial (f_1^\alpha, f_2^\alpha, \ldots, f_{2n}^\alpha)}{\partial (x_1, x_2, \ldots, x_{2n})} g(f_1^\alpha, f_2^\alpha, \ldots, f_{2n}^\alpha).$$

(4.4)

The magnetization and the energy are given by

$$\langle m_\sigma \rangle = m \sum_{\alpha} \sum_{\beta} \rho_\alpha \rho_\beta \int \int \sigma^{a\beta}(x_1^a, x_2^a, \ldots, x_{2n}^a; x_1^\beta, x_2^\beta, \ldots, x_{2n}^\beta)$$
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\[ \times g(x_1^a, x_2^a, \ldots, x_n^a)g(x_1^b, x_2^b, \ldots, x_n^b) dv_1 dv_2, \quad (4\cdot5) \]

and

\[ \varepsilon = \sum_a p_a \int \int \epsilon^a(x_1^a, x_2^a, \ldots, x_n^a; x_i^a, x_i^b, \ldots, x_i^b) \]

\[ \times g(x_1^a, x_2^a, \ldots, x_n^a)g(x_1^b, x_2^b, \ldots, x_n^b) dv_1 dv_2 - \langle m\sigma \rangle H. \quad (4\cdot6) \]

In this paper, we consider the case \( S = 1/2 \). The functions \( f^a(x) \) and \( \tilde{f}^a(x) \) are given by

\[ f^a(x) = \frac{x - \tanh C}{\tanh K_a (1 - x \tanh C)}, \quad (4\cdot7) \]

\[ \tilde{f}^a(x) = \frac{\tanh C + x \tanh K_a}{1 + x \tanh K_a \tanh C}, \quad (4\cdot8) \]

where \( C = mH/kT \) and \( K_a = J_a/2kT \). We first give the procedure of the low field expansion. The magnetization and the energy are expanded as

\[ \langle m\sigma \rangle = m \sum_a \sum_b p_a p_b \int \tan C + x_a \tanh K_a + x_b \tanh K_b + x_a x_b \tanh K_a \tanh K_b \]

\[ \times g(x_a) g(x_b) dx_a dx_b \]

\[ = m \sum_a \sum_b p_a p_b \sum_i \sum_j (-1)^i \left( \frac{i}{j} \right) \tanh i C \{ \tanh C \{ y \tanh K_a \}^{(i-j+1)} \{ y \tanh K_b \}^{(j-i-1)} + (y \tanh K_a)^{(i-j+1)} \{ y \tanh K_b \}^{(j-i-1)} \} \]

\[ + (y \tanh K_a)^{(i-j+1)} \{ y \tanh K_b \}^{(j-1)} \tanh C \} \quad (4\cdot9) \]

and

\[ \varepsilon = -\frac{1}{2} \sum_a J_a p_a \int \left( 1 + x_b x_r \right) e^{K_a} - \left( 1 - x_b x_r \right) e^{-K_a} \]

\[ \left( 1 + x_a x_r \right) e^{K_a} + \left( 1 - x_a x_r \right) e^{-K_a} g(x_r) g(x_r) dx_\beta dx_r - \langle m\sigma \rangle H \]

\[ = -\frac{1}{2} \sum_a J_a p_a \left[ \tanh K_a - 2 \cosech \tanh K_a \sum_i (-1)^i \left( \frac{i}{j} \right) \{ y \tanh K_a \}^{(i)} \right] - \langle m\sigma \rangle H, \quad (4\cdot10) \]

where

\[ (a y)^{()f} = \int (a x)^f g(x) dx = a^f y^{()f}. \quad (4\cdot11) \]

The average values are given from the simultaneous linear equation

\[ y^{()} = \sum_{f=0}^{\infty} y^{()} [ \sum_{a=1}^{n} p_a a^a_{i,f} ], \quad i = 1, 2, \ldots. \quad (4\cdot12) \]

The coefficients \( a^a_{i,f} \) are given from recurrence relations

\[ a^a_{i+1,f} = \sum_{l=0}^{i} (-1)^i \tanh C \tan h K_a \{ \tanh C a^a_{i,f-1} + \tanh K_a a^a_{i-1,f-i} \} \quad (4\cdot13) \]

with initial terms

\[ a^a_{i,0} = 1, \quad i = 1, 2, \ldots. \]
\[ a^n_j = \delta_{n,j}, \quad a^n_0 = \tanh C, \quad a^n_j = (-1)^{j-1} \tanh^j K \tanh^{j-1} C (1 - \tanh^2 C) \quad \text{for} \quad j \geq 1. \] (4.14)

The first two terms of \( y^{(t)} \) are obtained as

\[ y^{(t)} = \frac{\tanh C}{1 - \langle \tanh K \rangle} - \frac{\langle \tanh K \rangle + \langle \tanh^2 K \rangle}{(1 - \langle \tanh K \rangle)^2 (1 - \langle \tanh^2 K \rangle)} \tanh^2 C + \cdots \] (4.15)

and

\[ y^{(t)} = \frac{1 + \langle \tanh K \rangle}{(1 - \langle \tanh K \rangle) (1 - \langle \tanh^2 K \rangle)} \tanh^2 C + \cdots, \] (4.16)

where

\[ \langle A^n \rangle = \sum_a p_a A^n (\alpha). \] (4.17)

Then using Eqs. (4.9), (4.10), (4.15) and (4.16), we obtain

\[ \varepsilon = -\frac{1}{2} \langle J \tanh K \rangle - \frac{1}{2} \frac{\langle J (1 - \tanh^2 K) \rangle \tanh^2 C - m_H (1 + \langle \tanh K \rangle)}{1 - \langle \tanh K \rangle} \tanh C + \cdots \] (4.18)

and

\[ \langle m \sigma \rangle / m = \frac{1 + \langle \tanh K \rangle}{1 - \langle \tanh K \rangle} \tanh C - 2 \frac{\langle J (1 - \tanh^2 K) \rangle (\langle \tanh K \rangle + \langle \tanh^2 K \rangle)}{(1 - \langle \tanh K \rangle)^2 (1 - \langle \tanh^2 K \rangle)} \tanh^2 C + \cdots. \] (4.19)

The first terms of Eqs. (4.18) and (4.19) are in agreement with the previous results.\(^{12,13}\)

The functional equation is numerically solved. The magnetization curves of the binary mixture of two kinds of exchange integrals are shown in Figs. 7.

![Fig. 7. The magnetization of the random binary bond problem of \( J_A = 2J \), \( J_B = J \) (<0) and \( p_A = p_B = 0.5 \).](image_url)
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and 8. These have also some steps at low temperatures. To make them clear, we discuss the magnetization process at $T=0$.

First we consider the random binary mixture of two kinds of antiferromagnetic exchange integrals (see Fig. 7). The ground state of this mixture is completely antiferromagnetic when the magnetic field is low. The antiparallel spin turns over at

$$H = \frac{1}{2m} (|J_a| + |J_B|), \quad (4.20)$$

where $J_a$ and $J_B$ are exchange integrals linked with the spin. Then in general we find three steps in the magnetization curves, and the heights of those steps are given by $p_a p_B$. The susceptibility at a finite temperature

Fig. 8. The magnetization of the random binary bond problems of $J_A=J$, $J_B=-J$ ($J>0$), $kT/J=0.2$ and $p_A+p_B=1$.

Fig. 9. The magnetization of various binary systems whose concentrations of exchange bonds are the same. The solid lines indicate those of $p_2=0.5$ and $p_{-2}=0.5$, and the dotted lines those of $p_2=0.2$ and $p_{-2}=0.8$, when $kT/J=0.2$. B indicates the bond problem, and $\circ$, $\circ'$, $\circ$ and $\circ$ indicate the random binary systems of $J_{AA}=J_{BB}=J$ and $J_{AB}=-J$, $J_{AA}=J_{BB}=-J$ and $J_{AB}=J$, $J_{AA}=J_{AB}=J$, and $J_{BB}=J$, respectively.
converges when the temperature tends to zero (see Eq. (4.19)).

Next, consider the binary mixture of ferromagnetic and antiferromagnetic exchange integrals (see Fig. 8). The magnetization vanishes when $H \to 0$. However, owing to the randomness of local magnetization, the response of the magnetization to magnetic field is very strong near $H = 0$. Then the susceptibility diverges at $H = 0$. The remarkable steps in the high field region is due to the flips of spins.

Finally, we mention the relevances between the problem of the random magnetic system and the problem of the random exchange integrals. It was shown that the essential differences are found between susceptibilities of these two systems when the ferromagnetic elements and the antiferromagnetic elements are mixed.\textsuperscript{12,19} The difference is also found in the magnetization processes. In Fig. 9, we show the magnetization curves of both systems whose concentrations of bonds are the same. These differences are due to the correlations of the distribution of magnetic bonds through the magnetic atoms on the lattice sites.

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**Appendix**

The $y_a^{(1)}$ and $y_a^{(2)}$ in §2-4, are given by

$$y_a^{(1)} = y_a^0 + y_a',$$

$$y_b^{(1)} = y_b^0 + y_b',$$

$$y_a^{(2)} = \frac{1}{D_2} \{ F_A (1 - p_B t_{BB}^2) + F_B p_B t_{AB}^2 \},$$

$$y_b^{(2)} = \frac{1}{D_2} \{ F_B (1 - p_A t_{AA}^2) + F_A p_A t_{AB}^2 \},$$

where

$$F_A = \frac{1}{D_1} \left[ \tanh^2 C_A \{(1 - p_B t_{BB}) (1 + p_A t_{AA}) + p_A p_B t_{AB}^2 \} + 2p_B t_{AB} \tanh C_A \tanh C_B \right],$$

$$F_B = \frac{1}{D_1} \left[ \tanh^2 C_B \{(1 - p_A t_{AA}) (1 + p_B t_{BB}) + p_A p_B t_{AB}^2 \} + 2p_A t_{AB} \tanh C_A \tanh C_B \right],$$

$$y_a^0 = \frac{1}{D_1} \{ \tanh C_A (1 - p_B t_{BB}) + p_B t_{AB} \tanh C_B \},$$

$$y_b^0 = \frac{1}{D_1} \{ \tanh C_B (1 - p_A t_{AA}) + p_A t_{AB} \tanh C_A \},$$

$$y_a' = -\frac{1}{D_1} \{ Q_A (1 - p_B t_{BB}) + p_B t_{AB} Q_B \},$$

$$y_b' = \frac{1}{D_1} \{ Q_B (1 - p_A t_{AA}) + p_A t_{AB} Q_A \}.$$
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\[ y_b' = -\frac{1}{D_1} \{ Q_B (1 - p_{AA}) + p_{AB} Q_A \}, \]

\[ Q_A = \tanh C_A \{ p_{AA}^A y_A^A + p_{AB}^B y_B^B \} + \tanh^4 C_A \{ p_{AA}^A y_A^A + p_{AB}^B y_B^B \}, \]

\[ Q_B = \tanh C_B \{ p_{BB}^B y_B^B + p_{AB}^B y_B^B \} + \tanh^4 C_B \{ p_{BB}^B y_B^B + p_{AB}^B y_B^B \}, \]

\[ D_1 = (1 - p_{AA}) (1 - p_{BB}) - p_{A} p_{B}^2, \]

\[ D_2 = (1 - p_{AA}) (1 - p_{BB}) - p_{A} p_{B}^2. \]

References

14) H. A. Kramers and G. H. Wannier, Phys. Rev. 60 (1941), 252, 263.

Note added in proof:

An Ising chain of \( S = \frac{1}{2} \) with random magnetic moments was studied by Lehman et al. and Vedenov et al. in connection with the explanation of the phenomena of the melting of DNA. [See G. W. Lehman and J. P. McCague, J. Chem. Phys. 49 (1968), 3170; A. A. Vedenov and A. M. Dykhne, Zh. Eksp. i Teor. Fiz. 55 (1968), 357.] In this case the problem becomes easy, because the functions \( f_{\pm,\beta}(x) \) are independent of \( \beta \). Then, it is enough for us to introduce an averaged distribution function defined by \( \bar{g}(x) = \sum_{\alpha} p_{\alpha} g_{\alpha}(x) \). The distribution function is determined from a similar functional equation as Eq. (4.4), and the magnetic properties are described by using the function.