Special emphasis was given the dependence of transition temperature on laminar period. The dependence was found to be in qualitative, but not quantitative, agreement with proximity-effect theory developed for superimposed films.

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Diffusion Constants near the Critical Point for Time-Dependent Ising Models. I*

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The diffusion constant of spins in ferromagnets or of molecules in binary mixtures near the critical point is discussed employing a time-dependent Ising model in which spin interactions are replaced by certain temperature-dependent transition probabilities of spin exchange. The spin diffusion constant is calculated with the single approximation of replacing a reduced spin distribution function by its value for local equilibrium with a given inhomogeneous spin density. The behavior of the diffusion constant near the critical point is dominated by a factor χ^{-1}, where χ is the magnetic susceptibility. This problem is also studied with the use of the Bethe lattice. The effects of surrounding spins on the transition probability for spin exchange are found to be essential for obtaining the critical slowing-down near the critical point. In view of this, Kociński's calculation of the spin diffusion constant is critically discussed.

1. INTRODUCTION

THE behavior of various transport coefficients near the critical point is one of the most interesting but least understood problems in statistical physics today, and a large body of experimental work is appearing without proper theoretical understanding.[1] Among these phenomena, the problem of spin diffusion in ferromagnets near the Curie point has received more theoretical treatment than others because of the apparent simplicity of the problem.[2–7] In the present paper we shall also be concerned with this problem. This problem can be translated into the problem of moleulcar diffusion in binary mixtures if we ignore the quantum nature of the Heisenberg system.[8] The spin density and the external magnetic field correspond to the concentration and the chemical potential, respectively. In this paper we shall use the terminology of the spin system.

We shall now briefly discuss the present status of this problem. The most common theoretical argument goes somewhat like this.[2,4,9] Let us consider an isolated spin system and divide it into small but macroscopic cells. Let the fluctuation of the magnetization of the jth cell be M_j. Then the excess entropy associated with this fluctuation is written as

$$\Delta S = -k_B/2 \sum_{jl} a_{jl} M_j M_l \tag{1.1}$$

$$= -k_B/2n \sum_{\mathbf{q}} \lambda_{\mathbf{q}} M_{\mathbf{q}} M_{\mathbf{q}}^*, \tag{1.2}$$

where n is the total number of cells and k_B the Boltzmann constant, and we have introduced the following Fourier transforms:

$$\lambda_{\mathbf{q}} \equiv \sum_l a_{jl} \exp[i\mathbf{q}\cdot(\mathbf{r}_j - \mathbf{r}_l)], \tag{1.3}$$

$$M_{\mathbf{q}} \equiv \sum_l M_l \exp(-i\mathbf{q}\cdot\mathbf{r}_l), \tag{1.4}$$

where $\mathbf{r}_j$ denotes the position vector of the jth cell. Since the probability of occurrence of the fluctuation $M_{\mathbf{q}}$ is proportional to $\exp(\Delta S/k_B)$, we have

$$\lambda_{\mathbf{q}}^{-1} = n^{-1}\langle M_{\mathbf{q}} M_{\mathbf{q}}^*\rangle = k_B T \chi_{\mathbf{q}}, \tag{1.5}$$

where $\chi_{\mathbf{q}}$ is the wave-vector-dependent magnetic sususceptibility and T the temperature. Thermodynamics of irreversible processes then gives the relaxation rate

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* A portion of this work was supported by the National Science Foundation.

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[2] L. Van Hove, Phys. Rev. **95**, 249 (1954); **95**, 1374 (1954); P. G. de Gennes, Comm. Energie At. (France) Rappt. No. 925, 1959; (unpublished); P. G. de Gennes and J. Villain, J. Phys. Chem. Solids **13**, 10 (1960); P. G. de Gennes, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. III.

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[6] J. Kocínski, Acta Phys. Polon. **24**, 273 (1963).

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[8] T. L. Hill, *Statistical Mechanics* (McGraw-Hill Book Company, Inc., New York, 1956).

[9] P. Debye, Phys. Rev. Letters **14**, 783 (1965).

of M_{q} as

$$\dot{M}_{\mathrm{q}}=L_{\mathrm{q}}\partial\Delta S/\partial M_{\mathrm{q}}^{*}=-\gamma_{\mathrm{q}}M_{\mathrm{q}}, \tag{1.6}$$

where L_{q} is Onsager's kinetic coefficient and the rate constant γ_{q} is, using (1.2) and (1.5),

$$\gamma_{\mathrm{q}}=(L_{\mathrm{q}}/nT)\chi_{\mathrm{q}}^{-1}. \tag{1.7}$$

When the total magnetization is conserved, for small q we have

$$L_{\mathrm{q}}\cong q^{2}L, \tag{1.8}$$

and χ_{q} reduces to a uniform susceptibility χ_0. Thus,

$$\gamma_{\mathrm{q}}=Dq^{2}, \tag{1.9}$$

with the diffusion constant D given by

$$D\equiv(L/nT)\chi_0^{-1}. \tag{1.10}$$

Now, L is *assumed* to be insensitive to the temperature variation near the critical point and the anomalous temperature dependence of D is attributed to that of χ_0. Thus the diffusion constant vanishes at the critical point because of the infinite susceptibility there. In other words, the diffusion constant vanishes at the critical point because the thermodynamic driving force which causes the diffusion vanishes.

Although the above argument is a quite general and plausible one, we cannot give any justification for the assumption that L is finite at the critical point without going into the microscopic meaning of L. Following the pioneering works of de Gennes,[2] such a microscopic study of L has been started for the Heisenberg spin system by Mori and the present author[3] employing a correlation-function expression for the spin diffusion constant. They found that L is expressed in terms of a time correlation function of torques acting on each spin. Since the static pair correlations of torques contain only short-range spin correlations, they argued that L would not contain any long-range spin correlations which can give rise to an anomalous temperature dependence of L. No rigorous justification for this, however, was given. Bennett and Martin[5] discussed this problem by means of Green's functions. However, they reached no definite conclusion concerning the behavior of D near the Curie point. Kociński[6] studied the problem using the constant-coupling approximation, and obtained a nonvanishing diffusion constant at the Curie point at variance with the phenomenological argument mentioned earlier. Recently, Mori[7] developed a very general and powerful method for calculating time correlation functions in the form of continued-fraction expansions which are expressed in terms of various static correlations, and applied the technique to the present problem. Although his theory may turn out to give the first-order approximations in some sense to the anomalies in various transport coefficients, the question of the convergence of his expansion, especially near the critical point, remains unanswered.

On the other hand, the Ising spin system in which certain transition probabilities for spin flips are allowed is used by Kikuchi[10] to calculate the spin diffusion constant. He introduced the method of path probability in which a certain variational principle is used to determine the most probable path for the states of the entire system. His result turned out to be equivalent to the Bethe approximation and his diffusion constant vanishes at the Curie point.

In the present paper, we shall discuss this problem using a well-defined mathematical model for spin diffusion which is the Ising spin system in which spin interactions are replaced by a set of certain transition probabilities for spin exchange, as described in Sec. 2. The loss of generality for choosing a particular model is compensated by the simplicity of the treatment and the clarity of the approximation introduced. Furthermore, the present treatment gives some insight into the nature of the approximations which give the diffusion constant which vanishes at the critical point. In Sec. 3 we calculate the diffusion constant by expressing the spin current in terms of the reduced distribution function for a pair of nearest-neighbor spins and their nearest neighbors. Under a single approximation of replacing the reduced distribution function by its value at local equilibrium with a given spin-density gradient, we obtain a diffusion constant whose behavior near the Curie point is dominated by the inverse of the magnetic susceptibility in agreement with earlier theories. In Sec. 5 we present a slightly different treatment for a special system (Bethe lattice), whereby we critically discuss the calculation of Kociński.

Although the model employed is a rather special one, we believe, in view of the enormous difficulties of treating real systems, that it is certainly worthwhile to study the simplest possible microscopic models for which something definite can be said.

2. KINETIC MODEL

The system we shall consider is an array of N coupled spins. The coupling between spins is represented by a set of transition probabilities of spin exchange rather than by the conventional exchange interactions. The transition probabilities are so chosen as to give the same equilibrium spin distribution function as the conventional Ising system. In this respect, our model is quite similar to Glauber's,[11] but is different from his in that the total spin is conserved at each transition and we do not restrict the problem to one dimension.

Restricting ourselves to the lattices which do not have the so-called nearest-neighbor triangles, the transition probability for the isothermal spin-exchange transition in which the pair of spins σ_1 and σ_2 on the nearest neighboring sites 1 and 2 exchange each other,

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[10] R. Kikuchi, Ann. Phys. (N.Y.) 10, 127 (1960); 11, 306 (1960).

[11] R. J. Glauber, J. Math. Phys. 4, 294 (1963).

which satisfies the requirements of the preceding paragraph, is of the form

$$W_{12}(\{\sigma\}_{(12)})=\tfrac{1}{2}\alpha\prod_j(1+\gamma\sigma_2\sigma_j)\prod_l(1+\gamma\sigma_1\sigma_l)\,,\qquad(2.1)$$

where σ_j denotes the spin on the jth site and takes the values of $+1$ and -1 corresponding to up and down spins; α does not depend on σ's, γ is a function of temperature defined below. The product of j (or l) is over the nearest-neighboring lattice sites of the site 1 (or 2) excluding the site 2 (or 1). We use a symbol (ij) where i and j are nearset neighbors to designate a set of sites consisting of the sites i, j and their nearest neighbors surrounding them. A symbol $[ij]$ is used in a similar way, but excluding the sites i and j. Thus $\{\sigma\}_{(12)}$ is a set of σ's referring to the sites belonging to (12). γ is determined by the following condition of detailed balancing as mentioned earlier:

$$\frac{W_{12}(\sigma_1\sigma_2\{\sigma\}_{[12]})}{W_{12}(\sigma_2\sigma_1\{\sigma\}_{[12]})}=\frac{p_{(12)}{}^{e}(\sigma_2\sigma_1\{\sigma\}_{[12]})}{p_{(12)}{}^{e}(\sigma_1\sigma_2\{\sigma\}_{[12]})}\,,\qquad(2.2)$$

where $p_{(12)}{}^{e}$ is the equilibrium distribution function of spins on the sites (12) and has a form

$$p_{(12)}{}^{e}(\{\sigma\}_{(12)})=\zeta^{-1}\exp(\mathcal{K}_0+\mathcal{K}')\,,\qquad(2.3)$$

with

$$\mathcal{K}_0\equiv K(\sigma_1\sigma_2+\sigma_1\sum\sigma_j+\sigma_2\sum\sigma_l)\,,\qquad(2.4)$$

$$\zeta\equiv\sum_{\{\sigma\}_{(12)}}\exp(\mathcal{K}_0+\mathcal{K}')\,.\qquad(2.5)$$

$\mathcal{K}'$ does not contain σ_1 and σ_2, and the sum $\sum\sigma_j(\sum\sigma_l)$ is over the nearest-neighboring spins of $\sigma_1(\sigma_2)$ excluding $\sigma_2(\sigma_1)$. K is related to the magnitude of interaction between nearest-neighbor spins J of the usual Ising Hamiltonian by $K=J/k_BT$. Equation (2.3) can be easily verified using the definition of the reduced distribution function $p_{(12)}$;

$$p_{(12)}(\{\sigma\}_{(12)})=\mathrm{Tr}'p_N(\{\sigma\}_N)\,,\qquad(2.6)$$

where p_N is the normalized distribution function of the whole system, and its equilibrium value $p_N{}^{e}$ is given by

$$p_N{}^{e}(\{\sigma\}_N)=Z^{-1}\exp\mathcal{K}\,,\qquad(2.7)$$

with

$$\mathcal{K}\equiv\sum_{<jl>}K\sigma_j\sigma_l\,,\qquad(2.8)$$

and the sum is over all the nearest-neighboring pairs of spins, and Z is the partition function. Tr$'$ means to take the trace over all the spin states with fixed $\{\sigma\}_{(12)}$. Thus the right-hand side of (2.2) becomes

$$\frac{\exp K(\sigma_2\sum\sigma_j+\sigma_1\sum\sigma_l)}{\exp K(\sigma_1\sum\sigma_j+\sigma_2\sum\sigma_l)}=\frac{\prod_j[\cosh K+\sigma_2\sigma_j\sinh K]\prod_l[\cosh K+\sigma_1\sigma_l\sinh K]}{\prod_j[\cosh K+\sigma_1\sigma_j\sinh K]\prod_l[\cosh K+\sigma_2\sigma_l\sinh K]}\,.\qquad(2.9)$$

Comparison of this with (2.1) and (2.2) shows that the following choice of γ satisfies our requirement

$$\gamma=\tanh K\,.\qquad(2.10)$$

The transition described by (2.1) is meaningful only when $\sigma_2=-\sigma_1$, but we shall use (2.1) even for $\sigma_2=\sigma_1$ because it does not affect the kinetic equation we shall derive in the following due to cancellation.

In this model W is the transition probability for a spin exchange when the surrounding spin configuration is fixed. The effects of this surrounding spin state on W give rise to cooperative irreversible change. Thus α is regarded as the transition probability in the absence of spin correlation. Therefore, it is quite reasonable to assume that α is a slowly varying function of temperature near the critical point, and in particular we shall assume for definiteness that α is a constant. For Glauber's model the physical content of α is studied by Heims[12] and α is related to a certain correlation function of "lattice variables" under some assumptions. A similar analysis may be made for our α, which is however, beyond the scope of this paper.

Given the transition probabilities of elementary spin-exchange processes, the most general kinetic equation which describes the temporal development of the system is of course the master equation. Here we have assumed that the off-diagonal elements of the density matrix can be ignored. Denoting the normalized N spin distribution function at time t by $p_N(\{\sigma\}_N;\,t)$, the master equation is written as

$$(d/dt)p_N\{\sigma\}_N;t=-\sum_{<jl>}W_{jl}(\sigma_j\sigma_l\{\sigma\}_{[jl]})p_N(\{\sigma\}_N;t)$$
$$+\sum_{<jl>}W_{jl}(\sigma_l\sigma_j\{\sigma\}_{[jl]})p_N(\{\sigma\}_N{}^{jl};t)\,,\qquad(2.11)$$

where $\{\sigma\}_N{}^{jl}$ is the spin configuration in which σ_j and σ_l are interchanged in the configuration $\{\sigma\}_N$. $\langle jl\rangle$ means the pair of nearest-neighbor spins. We cannot in general solve the maser equation (2.11). Only for the case without spin correlation ($\gamma=0$) have we been able to use (2.11) to obtain the spin diffusion equation (Appendix). However, for the calculation of the diffusion constant, we do not require full knowledge of p_N, but only certain reduced distribution functions. Thus, in the following, we shall use a method which involves only the reduced distribution functions to calculate the diffusion constant.

3. DIFFUSION CONSTANT

We start the calculation of the diffusion constant by deriving an expression for the spin flux in the presence of

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[12] S. P. Heims, Phys. Rev. 138, A587 (1965).

a spin-density gradient along the x direction. For this purpose, we take a simple cubic lattice and consider a pair of adjacent spins σ_1 and σ_2 on the x axis and their nearest-neighboring spins $\sigma_3, \sigma_4, \cdots \sigma_{12}$ (Fig. 1). Then, the spin current density at time t in the positive x direction along the bond connecting 1 and 2, $J_x(t)$, is immediately written down as

$$J_x(t)=a^{-2} \sum_{\{\sigma\}(12)} (\sigma_1-\sigma_2)W_{12}(\{\sigma\}_{(12)}) \times p_{(12)}(\{\sigma\}_{(12)};t), \quad (3.1)$$

where a is the lattice constant and the factor $(\sigma_1-\sigma_2)$ takes care of the fact that when $\sigma_1=-\sigma_2=1$ (or -1) two units of spin flow in the positive (or negative) x direction and when $\sigma_1=\sigma_2$ there is no net spin flow during the transition. $p_{(12)}$ is the reduced distribution function for $\{\sigma\}_{(12)}$, and is related to the distribution function of the whole system $p_N(\{\sigma\}_N;\, t)$ by (2.6).

Since we know the transition probablilty W_{12}, (2.1), the main problem here is to find $p_{(12)}$ in the presence of a spin-density gradient. The most rigorous approach would be to solve the master equation (2.11) for an appropriate stationary state of constant spin flow. This has not been done in general in the presence of spin correlation γ. Thus we have to introduce a fundamental approximation which has been often used in a similar situation,[6,13] namely, we approximate $p_{(12)}(\{\sigma\}_{(12)};\, t)$ by its value in the local equilibrium state consistent with the given spin density. This is supposed to be a good approximation when we are dealing with a small deviation from equilibrium, although there appears to be no rigorous justification. Physically, our approximation amounts to calculating the initial spin flux at the moment when the constraint which has kept the system in local equilibrium is removed. Thus, rigorously speaking, we are actually calculating the "initial diffusion constant," although in the following we shall refer to it simply as the diffusion constant keeping this situation in mind.

Now, the local equilibrium $p_{(12)}$ is related by (2.6) to the local equilibrium $p_N(\{\sigma\}_N)$ which, for a small deviation from equilibrium, can be written as

$$p_N(\{\sigma\}_N)=p_N{}^e(\{\sigma\}_N)(1+\textstyle\sum_i h_i\sigma_i'), \quad (3.2)$$

where

$$\sigma_i'\equiv\sigma_i-\langle\sigma_i\rangle_0, \quad (3.3)$$

with the angular bracket $\langle\cdots\rangle_0$ denoting the average over the equilibrium state. h_j is a fictitious external magnetic field acting upon the spin on the jth site to produce a given inhomogeneous average magnetization $m_j=\langle\sigma_j'\rangle$. In other words,

$$m_j=\textstyle\sum_l \phi_{jl}h_l, \quad (3.4)$$

where

$$\phi_{jl}\equiv\langle\sigma_j'\sigma_l'\rangle_0. \quad (3.5)$$

Using (2.6), (2.7), and (3.2), the local equilibrium $p_{(12)}$ is obtained in the following form:

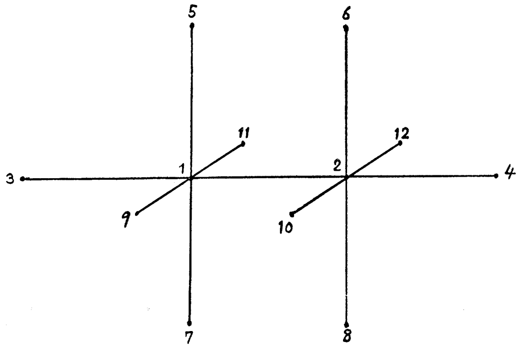

FIG. 1. Lattice sites in the simple cubic lattice.

$$p_{(12)}(\{\sigma\}_{(12)})=p_{(12)}{}^e(\{\sigma\}_{(12)})(1+h_1\sigma_1'+h_2\sigma_2'+\Delta), \quad (3.6)$$

where $p_{(12)}{}^e$ is given by (2.3) and Δ involves only $\sigma_3, \sigma_4, \cdots, \sigma_{12}$. This is a consequence of the fact that by fixing the spins $\sigma_3, \sigma_4, \cdots, \sigma_{12}$, the spins σ_1 and σ_2 are shielded from spins outside (12). Equation (3.6) is to be substituted into (3.1) to evaluate the spin current density.

In calculating the spin current density, it is convenient to re-express (2.1) as

$$W_{12}(\{\sigma\}_{(12)})=\tfrac{1}{2}\alpha(\cosh K)^{-2(z-1)} \times\exp(\mathcal{K}_0{}^{12}-K\sigma_1\sigma_2), \quad (3.7)$$

where $\mathcal{K}_0{}^{12}$ is obtained from $\mathcal{K}_0$, (2.4) by interchanging σ_1 and σ_2. Since in the spin current density only the case $\sigma_1=-\sigma_2$ contributes, we consider only this case. Then, clearly

$$\mathcal{K}_0{}^{12}-K\sigma_1\sigma_2=-(\mathcal{K}_0-K\sigma_1\sigma_2), \quad (3.8)$$

and we obtain

$$W_{(12)}(\{\sigma\}_{(12)})e^{\mathcal{K}_0}=\tfrac{1}{2}\alpha e^{-K}/(\cosh K)^{2(z-1)}. \quad (3.9)$$

The fact that this quantity is a constant number is closely related to the requirement of detailed balancing, (2.2). By the fact that both $\mathcal{K}'$ and Δ do not contain σ_1 and σ_2, the Δ term drops out after the summation over spin variables in (3.1). Thus, we finally obtain the following expression for the spin current density:

$$J=C(h_1-h_2), \quad (3.10)$$

where

$$C\equiv\frac{2\alpha}{a^2\zeta}\frac{e^{-K}}{(\cosh K)^{2(z-1)}}\sum_{\{\sigma\}[12]} e^{\mathcal{K}'}. \quad (3.11)$$

It should be noted that the current is proportional to the difference of the *external* fictitious fields h's, which do not contain any molecular field coming from surrounding spins.

When the spin density is slowly varying in space, we have

$$h_j\cong\chi^{-1}m_j, \quad (3.12)$$

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[13] H. Reiss, J. Chem. Phys. 40, 1783 (1964).

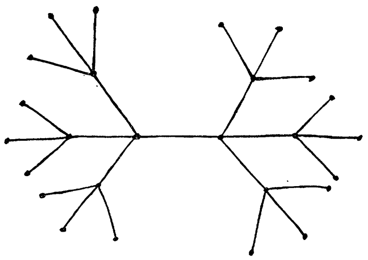

FIG. 2. A Bethe lattice of coordination number 4.

where χ is the dimensionless differential magnetic susceptibility, and

$$h_1-h_2=-\chi^{-1}a^4(\partial/\partial x)m(\mathbf{r}), \qquad (3.13)$$

where we have introduced the continuous spin density $m(\mathbf{r})$ per unit volume. Equation (3.10), then reduces to

$$J=-D(\partial/\partial x)m(\mathbf{r}), \qquad (3.14)$$

with the diffusion constant

$$D\equiv Ca^4/\chi. \qquad (3.15)$$

Let us now discuss the behavior of the diffusion constant near the Curie point. First consider the quantity

$$\zeta/4 \sum_{\{\sigma\}[12]} e^{\mathcal{K}'}=\sum_{\{\sigma\}(12)} e^{\mathcal{K}_0+\mathcal{K}'}\Big/\sum_{\{\sigma\}(12)} e^{\mathcal{K}'},$$

which can be regarded as an average of $e^{\mathcal{K}_0}$ with the weight function $e^{\mathcal{K}'}$. Since the magnitude of $\mathcal{K}_0$, (2.4), is limited except at the zero temperature, this quantity cannot be zero or infinite at any finite temperature. Other factors in C are well behaved. Therefore, the temperature dependence of the diffusion constant near the Curie point is determined mostly by the magnetic susceptibility χ in (3.15), and in particular, the diffusion constant vanishes like χ^{-1} at the Curie point. This is in complete agreement with the conclusion of various arguments discussed in Sec. 1 except that of Kociński. It should be noted that this result is obtained with the local-equilibrium approximation only besides the specific choice of the model. No other statistical approximation has been used. In fact the greatest advantage of this method is the complete separation, under the local equilibrium approximation, of the dynamics of the problem from the complicated statistics of the Ising spin problem.

4. BETHE LATTICE

In this section we shall present a slightly different treatment which does not use the magnetic susceptibility explicitly. This method is more closely related to Kociński's treatment than the previous one and thus facilitates the discussion of his theory. Since the method is only approximate in general lattices corresponding to the Bethe approximation, we shall consider the special lattice called the Bethe lattice in which the Bethe approximation becomes exact.[14,15] As an illustration a Bethe lattice in which each spin has four nearest neighbors is shown in Fig. 2.

In order to adapt a Bethe lattice in which each spin has z nearest neighbors to the problem of spin diffusion, we arrange a Bethe lattice in the following special way in three-dimensional space (Fig. 3). First, choose a chain of nearest-neighboring spins out of the Bethe lattice and arrange them on the x axis (spins $\cdots$, σ_3, σ_1, σ_2, σ_4, $\cdots$ in Fig. 3). Next set up a plane parallel to the yz plane intersecting at every spin on the x axis, and arrange the remaining spins of the Bethe lattice on these planes. Thus, each spin on the x axis has $z-2$ nearest neighbors on the plane (sheet) because two are already on the x axis. A typical spin arrangement on a sheet is illustrated in Fig. 4 for the case of $z=4$.

Let us now suppose that the spin-density gradient exists only in the x direction and consider the spin current along the x axis. The magnetization density is uniform in each sheet and there is no net spin flow within the sheets at least in the local equilibrium approximation. The spin current along the bond connecting 1 and 2 takes the same form as (3.1) where we drop a^{-2}:

$$J_x(t)=\sum_{\{\sigma\}(12)}(\sigma_1-\sigma_2)W_{12}(\{\sigma\}_{(12)})p_{(12)}(\{\sigma\}_{(12)};t). \qquad (4.1)$$

By the same manipulations as in Sec. 2, (4.1) reduces to the same form as (3.10) and (3.11) except that a^2 is missing:

$$J=C(h_1-h_2), \qquad (4.2)$$

An explicit expression of C is given below in (4.10). h_1 and h_2 can be calculated from the knowledge of m_j's and $p_{(12)}(\{\sigma\}_{(12)};t)$, which takes the following simple form for the Bethe lattice:

$$p_{(12)}(\{\sigma\}_{(12)})=p_{(12)}{}^e(\{\sigma\}_{(12)})$$

$$\times\Big[1+h_1\sigma_1+h_2\sigma_2+\hat{h}_3\sigma_3+\hat{h}_4\sigma_4+\sum_{j=1}^{z-2}(\hat{h}_{1j}\sigma_{1j}+\hat{h}_{2j}\sigma_{2j})\Big], \qquad (4.3)$$

where we have denoted the nearest-neighboring sites of the sites 1 and 2 in the sheets intersecting sites 1, and 2, respectively, by $1j$ and $2j$ ($j=1, 2, \cdots, z-2$), respec-

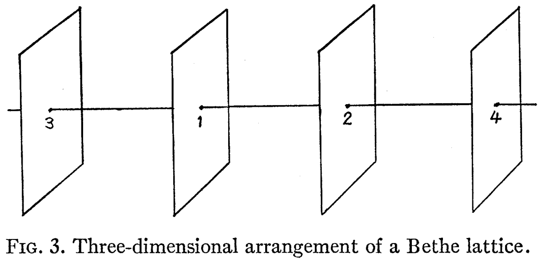

FIG. 3. Three-dimensional arrangement of a Bethe lattice.

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[14] C. Domb, Advan. Phys. 9, 149, 245 (1960).

[15] M. E. Fisher, Physica 28, 172 (1962).

tively, and $\hat{h}$'s in general differ from h's because they include the effects of spins outside the cluster (1,2). In the following discussion, we only consider the region above the Curie temperature. The h's as well as $\hat{h}$'s are determined by relations somewhat analogous to (3.4):

$$m_j = h_1\phi_{1,j} + h_2\phi_{2,j} + \hat{h}_3\phi_{3,j} + \hat{h}_4\phi_{4,j} + \sum_{l=1}^{z-2} (\hat{h}_{1l}\phi_{1l,j} + \hat{h}_{2l}\phi_{2l,j}) . \qquad (4.4)$$

This equation is considerably simplified by assuming a uniform spin-density gradient in the x direction with the zero spin density at the midpoint of the sites 1 and 2 and by noting that if the sites j and l are separated by n bonds, we have[14,15]

$$\phi_{j,l} = \gamma^n . \qquad (4.5)$$

Therefore, by symmetry, we can assume that

$$\begin{aligned} &m_2 = m_{2j} = -m_1 = -m_{1j} \equiv \Delta\tfrac{1}{2}m ,\\ &m_4 = -m_3 = \tfrac{3}{2}\Delta m ,\\ &h_2 = -h_4 , \quad \hat{h}_4 = -\hat{h}_3 , \qquad (4.6)\\ &\hat{h}_{2j} = -\hat{h}_{1i} = \text{independent of } j . \end{aligned}$$

Thus, (4.4) reduces to the following set of equations:

$$\begin{aligned} m_1 &= -\tfrac{1}{2}\Delta m = (1-\gamma)h_1 + (z-2)\gamma(1-\gamma)\hat{h}_{11} + \gamma(1-\gamma)\hat{h}_3 ,\\ m_{11} &= -\tfrac{1}{2}\Delta m = \gamma(1-\gamma)h_1 + [1+(z-3)\gamma^2 - (z-2)\gamma^3]\hat{h}_{11}\\ &\qquad + \gamma^2(1-\gamma)\hat{h}_3 , \qquad (4.7)\\ m_3 &= -\tfrac{3}{2}\Delta m = \gamma(1-\gamma)h_1 + (z-2)\gamma^2(1-\gamma)\hat{h}_{11}\\ &\qquad + (1-\gamma^3)\hat{h}_3 . \end{aligned}$$

This can be solved to yield

$$h_1 = -h_2 = -[1-(z-1)\gamma]\Delta m/2(1+\gamma) . \qquad (4.8)$$

Substituting this into (4.2), it reduces to

$$J = -D'\Delta m ,$$

with the "diffusion constant" D' given by

$$D' \equiv [1-(z-1)\gamma]C/(1+\gamma) . \qquad (4.9)$$

Because $\mathcal{K}'=0$ for the Bethe lattice above the Curie point, we can give an explicit expression for C,

$$C = \tfrac{1}{2}\alpha e^{-K} (\cosh K)^{4z-3} . \qquad (4.10)$$

Since the Curie point for the Bethe lattice is determined by[14,15]

$$1-(z-1)\gamma_c = 0 , \qquad (4.11)$$

the diffusion constant again vanishes like χ^{-1} at the Curie point.

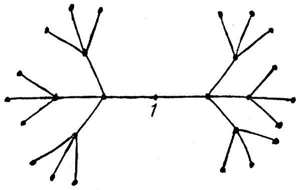

FIG. 4. Lattice sites in a sheet of Fig. 3.

Let us now discuss Kociński's treatment of spin diffusion. Although he treats the Heisenberg spin system rather than the stochastic Ising spin system, his treatment of statistics can be adapted to the present problem. He considers only the pair of spins (our σ_1 and σ_2) and neglects the dependence of the transition probability W_{12} on the spin configuration $\{\sigma\}_{[12]}$. Thus, instead of (4.1) he starts essentially from,

$$J_x(t) = \sum_{\sigma_1\sigma_2} W_{12}(\sigma_1-\sigma_2)p_{12}(\sigma_1\sigma_2;t) , \qquad (4.12)$$

where p_{12} is the reduced distribution function of the spin pair σ_1 and σ_2. The local-equilibrium approximation, which is also implicit in his treatment, gives for p_{12},

$$p_{12}(\sigma_1\sigma_2;\, t) = p_{12}{}^e(\sigma_1\sigma_2)[1+\hat{h}_1\sigma_1+\hat{h}_2\sigma_2] , \qquad (4.13)$$

where $\hat{h}_1$ and $\hat{h}_2$ are not the same as the external fictitious fields, but contain the effects of outside spins, and are determined by

$$-\tfrac{1}{2}\Delta m = m_1 = (1-\gamma)\hat{h}_1 , \qquad (4.14)$$

where we have used the facts that $m_2=-m_1$ and $\hat{h}_2=-\hat{h}_1$ by symmetry. In the same way as we have done in our exact treatment, (4.12), (4.13), and (4.14) lead to the following diffusion constant D':

$$D' = W_{12} . \qquad (4.15)$$

This result indicates that the effects of nearest neighbors of the pair σ_1 and σ_2 on the transion probability for the spin exchange $\sigma_1 \leftrightarrow \sigma_2$ are essential in the problem of spin diffusion near the critical point. Thus Kociński's result that the spin diffusion constant of the Heisenberg spin system does not vanish at the Curie point seems to be due to the crudeness of his statistical treatment, and the agreement of his theoretical values with the recent experiments[16] appears to be fortuitious.

5. CONCLUDING REMARKS

In preceding sections, we have presented two calculations of the spin diffusion constant for time-dependent Ising models. In both cases, the spin diffusion constant vanishes like χ^{-1} near the Curie point. This is a consequence of the fact that the thermodynamic driving force which causes the spin current between the adjacent lattice sites 1 and 2 is proportional to the difference between the *external* fictitious magnetic fields

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[16] B. Jacrot *et al.*, in *Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963); L. Passel *et al.*, J. Appl. Phys. **35**, 933 (1964); Phys. Rev. **139**, A1866 (1965).

on these sites which maintain the given spin-density gradient. Namely, these fields are connected to the average magnetization by (3.12), whereby introducing the magnetic susceptibility χ. This gives rise to the factor χ^{-1} in the diffusion constant. This, in turn, is a consequence of the fact that in our treatment, by fixing the spin states on the nearest-neighboring sites of 1 and 2, $\{\sigma\}_{[12]}$, the spins σ_1 and σ_2 are "protected" from outside spins. This sort of "protection" or "shielding" gives rise to the so-called critical slowing-down which causes the diffusion constant to vanish at the critical point.

Although the present treatment cannot be applied to the Heisenberg spin system, any approximate treatment which is expected to be valid for this system must give the correct result when applied to our simpler models. Thus, in Sec. 4 we have discussed Kociński's treatment as an example of an approximation in which the spins σ_1 and σ_2 are not "shielded" from the outside of (12) and fails to give the critical slowing-down near the Curie point.

In the present paper we have been concerned only with the diffusion process, whereas the master equation certainly allows more general types of processes.[17] However, if we restrict ourselves to the very slow processes which take place over very large spatial regions, the diffusion equation should normally give the good description of the process because in our model the only constant of motion is the total spin. In the immediate vicinity of the critical point, however, the simple diffusion law may not be valid if the range of spatial correlations of critical spin-density fluctuation exceeds the scale of inhomogeneous disturbances.[18] Thus the diffusion constant near the critical point is meaningful only if the scale of inhomogeneity of spin density is sufficiently larger than the range of critical fluctuation of spin density.

In a subsequent paper we shall examine the local equilibrium approximation employed in this paper, and shall show that the approximate diffusion constant obtained in this paper provides a rigorous upper bound to the true diffusion constant, thus establishing the existence of the critical slowing-down. The method put forward in this paper can be applied to other irreversible cooperative phenomena, as we shall demonstrate in subsequent papers.

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APPENDIX

Here we shall discuss the master equation (2.11) in the absence of spin correlation, $\gamma=0$, which is written as

$$(d/dt)p_N(\{\sigma\}_N)=-\sum_{<jl>} w_{jl}p_N(\{\sigma\}_N)+\sum_{<jl>} w_{jl}p_N(\{\sigma\}_N{}^{jl}). \quad \text{(A1)}$$

Using the definition of the average value of spin σ_j,

$$m_j=\sum_{\{\sigma\}_N} \sigma_j p_N(\{\sigma\}_N).$$

(Here we assume that the average spin vanishes in equilibrium.), we obtain

$$\sum_{\{\sigma\}_N} \sigma_j p_N(\{\sigma\}_N{}^{jl})=\sum_{\{\sigma\}_N} \sigma_l p_N(\{\sigma\}_N)=m_l.$$

Therefore, multiplying (A1) by σ_j and summing over all the spin states, we obtain

$$(d/dt)m_j=-\sum_l w_{jl}m_j+\sum_l w_{jl}m_l. \quad \text{(A2)}$$

By substituting the expansion

$$m_l=m_j+(\mathbf{r}_l-\mathbf{r}_j)\cdot\nabla_j m_j+\tfrac{1}{2}(\mathbf{r}_l-\mathbf{r}_j)(\mathbf{r}_l-\mathbf{r}_j):\nabla_j\nabla_j m_j+\cdots$$

into (A2) and noting that $w_{jl}=w_{lj}$, we have, for a simple cubic lattice,

$$(d/dt)m_j=D\nabla_j{}^2m_j+\cdots, \quad \text{(A3)}$$

where the diffusion constant D is given by

$$D=\tfrac{1}{6}\sum_l(\mathbf{r}_l-\mathbf{r}_j)^2w_{jl}. \quad \text{(A4)}$$

Using the definition of w_{jl} which is given by (2.1) where we set $\gamma=0$, this reduces to

$$D=\tfrac{1}{2}\alpha a^2. \quad \text{(A5)}$$

This agrees with the result of Sec. 3 when $\gamma=K=0$, as one can immediately check.

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[17] S. P. Heims (private communication).

[18] R. Mountain, J. Res. Natl. Bur. Std. (U.S.) **69A**, 523 (1965).