Time evolution of fluctuations in the path-probability method. I

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The path-probability method (PPM) is applied for the first time to the time development of the fluctuation from the most probable path, where the kinetic Ising ferromagnetism in the pair approximation is treated as an example. The evolution equations of the variance as well as the average motion of order parameters are derived and compared critically with the results obtained by the master-equation method (MEM). In the pair approximation, the PPM results agree completely with those of the MEM. This agreement is to be compared with the disagreement in the triangle approximation presented in Wada *et al.* (the following paper).

I. INTRODUCTION

The path-probability method[1] (PPM) of irreversible statistical mechanics has been applied successfully to a variety of diffusion[2] and phase transition kinetics.[3–5] The PPM has been developed as a natural extension to the time domain from the cluster-variation method (CVM) (Ref. 6) in equilibrium statistical mechanics. In the CVM, the free energy function is constructed in terms of state variables which characterize a state of the system. Minimization of the free energy function with respect to these variables gives us the free energy and the state variables in thermodynamical equilibrium. On the other hand, in the PPM, the path-probability function (PPF) is constructed in terms of a set of path variables which denote the change of the state variables in time Δt. Then the equations for describing the evolution of the state variables are derived from the most probable path corresponding to a maximum of the PPF with respect to these path variables. In addition to the most probable path, because of its variational nature, the PPF is expected to contain information with respect to the fluctuation from the most probable path. Nevertheless, in the framework of the PPM, only the most probable path has been studied so far and no work has been done on the time evolution of fluctuation except for one simple example.[1]

The derivation of the transition probability by a systematic construction of the PPF and its evaluation based on the variation principle in a similar fashion to those of the free energy function of the CVM in dealing with irreversible phenomena are strong points of the PPM. However, the applicability of the PPM is also closely connected to this formalism and its success depends largely on the type of irreversible phenomena. For example, the PPM is most suitable in following the time dependence of the state variables (macrovariables) such as the time evolution of order[3,4] and of magnetization, but is less success-

ful in dealing with transport phenomena where the time correlation of individual particles rather than the assembly of particles is followed.[7–9] In order to find a way to extend the applicability of the PPM, it would be most fruitful if the process of deriving the evolution equations can be compared with other respectable methods such as the master-equation method (MEM) adopted in irreversible statistical mechanics.[10] Because of this, we took an example of Ising ferromagnetism which has been proved to be suitable for the treatment by the PPM and compared the procedures with those of the MEM as the first step toward this goal. At the same time, we demonstrate how the information on fluctuation can be derived by the PPM.

Recently, starting with the master equation, Kubo *et al.*[11] developed a general theory dealing with the evolution of macrovariables, assuming that those variables followed the Markovian process. With the use of this theory, which we will call the MEM specifically, Saito and Kubo[12] investigated the kinetic Ising model in the pair approximation. They derived the evolution equations for the average and the fluctuation of order parameters of the system. Therefore, the derivation of the evolution equations by the PPM can be critically compared with those by the MEM.

In Sec. II, the most probable path is derived from the PPM. Section III presents the PPF expression for the fluctuation from the most probable path. Section IV shows that the PPM leads to the Fokker-Planck equation for the fluctuation of order parameters, and then to the evolution equations of the fluctuation which are identical to those derived from the MEM reviewed in the Appendix.

In the following paper (hereafter referred to as II), we show the extension of this treatment to a higher degree of approximation (triangle approximation). As is shown in the present paper (paper I), the PPM and the MEM agree

completely and this seems to show the equivalence of the PPM and the MEM. Nevertheless, in the triangle approximation, these two methods show a minor but a definite difference. The derivations in this paper are given in a way to make the understanding of this difference clear.

II. THE MOST PROBABLE PATH IN THE PPM

Let us consider a homogeneous ferromagnetic Ising system composed of N spins on a lattice. The Hamiltonian of the system is as follows:

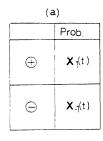$$\mathscr{H} = -J \sum_{\langle n,n' \rangle} \sigma_n \sigma_{n'} - \mu_0 H \sum_n \sigma_n , \quad (2.1)$$

where σ_n $(= \pm 1)$ is the Ising spin on the nth site, J is the exchange integral, $\langle n,n' \rangle$ represents a nearest-neighbor pair, and $\mu_0 H$ is the Zeeman energy. In the PPM, we define two kinds of variables, to be called the state and the path variables, specified at a time instant t. The state variables in the pair approximation are the fractionals shown in Fig. 1. We use subscripts $i = 1$ and -1 for up and down spins along an external magnetic field, respectively. The probability of appearance of an i spin at a lattice site at time t is denoted by $x_i(t)$ and that of a nearest-neighbor pair having a configuration (i,j) by $y_{ij}(t)$. The x and y variables are connected by the geometrical relations

$$x_i(t) = \sum_{j=\pm 1} y_{ij}(t) = \sum_{j=\pm 1} y_{ji}(t) \quad (\text{for } i = \pm 1) ,$$

$$x_1(t) + x_{-1}(t) = 1 . \quad (2.2)$$

Along with the x and y variables, we introduce the variables to specify the macroscopic state of the homogeneous system at time t, namely, the magnetization $m_1(t)$ per site and the short-range order parameter $m_2(t)$, which are defined as

$$m_1(t) = \sum_{i=\pm 1} i x_i(t) ,$$

$$m_2(t) = \frac{z}{2} \sum_{i,j=\pm 1} ij y_{ij}(t) , \quad (2.3)$$

FIG. 1. State variables $x_i(t)$ and $y_{ij}(t)$ for the homogeneous Ising model. These indicate the probabilities of configurations at time t for a point and a pair. The symbol β indicates the weight factor.

FIG. 2. Path variables $X_{i,i'}(t,t+\Delta t)$ and $Y_{ij,i'j'}(t,t+\Delta t)$ for the Ising model in the pair approximation. Their arguments are omitted here. The "Abbr." column shows abbreviated notations for path variables, and the β column shows the weight factors.

where z is the coordination number of the lattice.

The path variables are defined in Fig. 2 in which $X_{i,i'}(t,t+\Delta t)$ describes the fraction of spins which is i at t and will change to i' at $t+\Delta t$, and $Y_{ij,i'j'}(t,t+\Delta t)$ is the fraction of pairs which is (i,j) at time t and will change to (i',j') at $t+\Delta t$. We define Δt small enough so that at most one of the second subscripts i' and j' is different from the first subscripts i and j in the time span of Δt. For later convenience we introduce abbreviated notations $X(i)$ and $Y_s(i)$ as in Fig. 2. Note that these variables in which a spin makes a flip in Δt are taken to be of the order $O(\Delta t)$. Since the path variables are the joint probability connecting the configurations at t and $t+\Delta t$, we can project (by forming linear combinations) the path variables X and Y to t and to $t+\Delta t$ to obtain the state variables $x_i(t), y_{ij}(t)$ and $x_{i'}(t+\Delta t), y_{i'j'}(t+\Delta t)$, respectively. Using the projections to t and $t+\Delta t$, we can write the change of the state variables defined by $\Delta f(t) \equiv f(t+\Delta t) - f(t)$ as follows:

$$\Delta x_i(t) = X_{-i,i} - X_{i,-i} ,$$

$$\Delta y_{ij}(t) = (Y_{-ij,ij} + Y_{i-j,ij})$$

$$\qquad - (Y_{ij,-ij} + Y_{ij,i-j}) \quad (i,j = \pm 1) . \quad (2.4)$$

In using the PPM, we ask how the system changes in Δt when the state at t is given. In the present problem, since

$y_{ij}(t)$ are given, we can choose $Y_s(i)$ ($i = \pm 1$, $s = 1,2$) as the independent variables as is seen from the relations

$$Y_{ii,ii} = y_{ii}(t) - 2Y_1(-i) ,$$

$$Y_{ij,ij} = y_{ij}(t) - [Y_2(i) + Y_2(-i)] \quad (i,j = \pm 1) . \qquad (2.5)$$

In the PPM of irreversible statistical mechanics, we start with the basic variation function in the same sense as the free energy function is the basic variation function in the CVM of equilibrium statistical mechanics. The basic variation function of the PPM is the PPF, and represents the probability that the path specified by the path variables occurs.[1] The PPF is made of three factors. The first one corresponds to the probability of occurrence of unit kinetic process (for which there is no counter part in the free energy function in the CVM):

$$\mathscr{P}_1 = \prod_{i=\pm 1} (\theta \Delta t)^{X_{i,-i}} (1 - \theta \Delta t)^{X_{i,i}} , \qquad (2.6)$$

where θ is a spin-flip rate per unit time. The second factor depends on the energy of activation and is a generalization of the Boltzmann factor. With the use of the ener-

gy change in Δt of the system

$$\Delta E = \sum_{i=\pm 1} \{ 2JzN[Y_1(i) - Y_2(i)] + 2\mu_0 HNiX(i) \} , \qquad (2.7)$$

the second factor is taken to be

$$\mathscr{P}_2 = \exp(-\Delta E / 2k_B T) . \qquad (2.8)$$

The third factor is the combinatorial factor corresponding to the entropy in the equilibrium case. The state variables in the combinatorial factor of the CVM are replaced by the path variables in the corresponding approximation

$$\mathscr{P}_3 = \left[\frac{P_{\text{point}}}{P_{\text{pair}}} \right]^{z/2} \left[\frac{P_{\text{point}}}{N!} \right]^{z/2-1} ,$$

$$P_{\text{point}} = \prod_{i,j} [(NX_{i,j})!] , \qquad (2.9)$$

$$P_{\text{pair}} = \prod_{(i,j),(i',j')} [(NY_{ij,i'j'})!] .$$

The final form of the PPF, $\mathscr{P}(t, t + \Delta t)$, is written in the logarithmic form as

$$\frac{1}{N} \ln \mathscr{P}(t, t + \Delta t) = (z-1) \sum_{i,j} \mathscr{L}(X_{i,j}) - \frac{z}{2} \sum_{(i,j),(k,l)} \mathscr{L}(Y_{ij,kl})$$

$$+ \sum_{i=\pm 1} \{ X_{i,-i} \ln(\theta \Delta t) + X_{i,i} \ln(1 - \theta \Delta t) - zK[Y_1(i) - Y_2(i)] - LiX(i) \} , \qquad (2.10)$$

where $K = J/k_B T$, $L = \mu_0 H/k_B T$, and $\mathscr{L}(x) = x \ln x - x$. We obtain the most probable path variables by differentiating $\mathscr{P}(t, t + \Delta t)$ with respect to the independent path variables $Y_s(i)$. The most probable values of the one spin-flip pair variables are then derived as

$$\hat{Y}_1(i) = \theta \Delta t \, Y_{ii}(t) e^{-K-iL} [\lambda(i)]^{z-1} ,$$

$$\hat{Y}_2(i) = \theta \Delta t \, Y_{i-i}(t) e^{K-iL} [\lambda(i)]^{z-1} \qquad (2.11a)$$

and those for a single spin flip become

$$\hat{X}_{i,-i}(t) = \theta \Delta t \, x_i(t) e^{-iL} [\lambda(i)]^z \quad (i = \pm 1) , \qquad (2.11b)$$

where the caret indicates the most probable path and $\lambda(i)$ is a function representing the effect of environment defined by

$$\lambda(i) = \sum_{j=\pm 1} Y_{ij}(t) e^{-ijK} / x_i(t) \quad (i = \pm 1) . \qquad (2.12)$$

The evolution equations in Δt for the most probable values of the order parameters are written from Eqs. (2.3) and (2.4) as

$$\Delta \bar{m}_1(t) = 2(\hat{X}_{-1,1} - \hat{X}_{1,-1}) \equiv R_1(t) \Delta t ,$$

$$\Delta \bar{m}_2(t) = 2z \sum_{i=\pm 1} [\hat{Y}_2(i) - \hat{Y}_1(i)] \equiv R_2(t) \Delta t , \qquad (2.13)$$

where $R_1(t)$ and $R_2(t)$ are defined for later purposes. The bar on $m_i(t)$'s indicates the most probable path value. Thus in the limit of $\Delta t \to 0$, the evolution equations for the order parameters become

$$\frac{d\bar{m}_i(t)}{dt} = R_i(t) \quad (i = \pm 1) . \qquad (2.14)$$

Note that from the following definition of $\bar{G}(t)$,

$$\bar{G}(t) = \sum_{i=\pm 1} \theta x_i(t) e^{-iL} \lambda^z(i) , \qquad (2.15)$$

we can derive the relations

$$R_1(t) = 2 \frac{\partial \bar{G}(t)}{\partial L}, \quad R_2(t) = 2 \frac{\partial \bar{G}(t)}{\partial K} . \qquad (2.16)$$

These equations for the most probable path are exactly the same as those derived by the MEM as will be shown in the Appendix.

III. THE FLUCTUATION DISTRIBUTION FUNCTION

In order to apply the system size expansion developed by van Kampen[13] to the PPF, we separate out the fluctuating quantity from the most probable path and write

$$Y_s(i) = \hat{Y}_s(i) + \sqrt{\epsilon} \xi_s(i) \quad (i = \pm 1, \, s = 1,2) , \qquad (3.1)$$

where $\epsilon \equiv 1/N$ is the smallness parameter of the system, and $\xi_s(i)$ is the deviation variable. The PPF is expanded around the most probable path up to the second order of $\xi_s(i)$ as

$$\frac{1}{N}\ln\mathscr{P}(t,t+\Delta t)=(z-1)\sum_i\mathscr{L}(x_i(t))-\frac{z}{2}\sum_{i,j}\mathscr{L}(y_{ij}(t))+(z-1)$$

$$+\frac{1}{N}\sum_{i=\pm1}\left[\frac{z-1}{2}\frac{[\xi_1(i)+\xi_2(i)]^2}{\hat{Y}_1(i)+\hat{Y}_2(i)}-\frac{z}{2}\left[\frac{[\xi_1(i)]^2}{\hat{Y}_1(i)}+\frac{[\xi_2(i)]^2}{\hat{Y}_2(i)}\right]\right].$$

(3.2)

The PPF $\mathscr{P}(t,t+\Delta t)$ gives a probability distribution function of fluctuation from the most probable path. It is a product of two factors except for a multiplicative constant factor:

$$\mathscr{P}(t,t+\Delta t)\propto\prod_{i=\pm1}F(\xi_1(i),\xi_2(i)),$$ (3.3)

with

$$F(\xi_1(i),\xi_2(i))$$

$$=C\exp\left[-\tfrac{1}{2}\sum_{s,s'=1,2}\xi_s(i)[D(i)]_{ss'}^{-1}\xi_{s'}(i)\right],$$ (3.4)

where C is a normalization constant and the matrix in Eq. (3.4) is given by

$$[D(i)]^{-1}=\begin{vmatrix}\dfrac{z}{\hat{Y}_1(i)}-\dfrac{z-1}{\hat{X}(i)} & -\dfrac{z-1}{\hat{X}(i)}\\[2mm]-\dfrac{z-1}{\hat{X}(i)} & \dfrac{z}{\hat{Y}_2(i)}-\dfrac{z-1}{\hat{X}(i)}\end{vmatrix}.$$ (3.5)

The inverse matrix of that in Eq. (3.5) is then given by

$$D(i)=\frac{\hat{Y}_1(i)\hat{Y}_2(i)}{z}\begin{vmatrix}\dfrac{z}{\hat{Y}_2(i)}-\dfrac{z-1}{\hat{X}(i)} & \dfrac{z-1}{\hat{X}(i)}\\[2mm]\dfrac{z-1}{\hat{X}(i)} & \dfrac{z}{\hat{Y}_1(i)}-\dfrac{z-1}{\hat{X}(i)}\end{vmatrix}.$$

(3.6)

From the general theory of Gaussian distribution,[14] the matrix (3.6) is also a correlation matrix defined by

$$D(i)=\begin{bmatrix}\langle\xi_1(i)\xi_1(i)\rangle & \langle\xi_1(i)\xi_2(i)\rangle\\\langle\xi_2(i)\xi_1(i)\rangle & \langle\xi_2(i)\xi_2(i)\rangle\end{bmatrix},$$ (3.7)

where $\langle\ \rangle$ denotes the average over the distribution function $F(\xi_1(i),\xi_2(i))$. By noting the relations

$$\Delta m_1(t)-\Delta\bar{m}_1(t)=-2\epsilon^{1/2}\sum_i i[\xi_1(i)+\xi_2(i)],$$

$$\Delta m_2(t)-\Delta\bar{m}_2(t)=-2z\epsilon^{1/2}\sum_i[\xi_1(i)-\xi_2(i)],$$ (3.8)

and by using the variance matrix R whose elements are defined as

$$\epsilon R_{ss'}(t)\Delta t$$

$$=\langle[\Delta m_s(t)-\Delta\bar{m}_s(t)][\Delta m_{s'}(t)-\Delta\bar{m}_{s'}(t)]\rangle$$

$$(s,s'=1,2),$$ (3.9)

we have the probability distribution function of the fluctuation from the most probable path in Δt as

$$\psi(\Delta\mathbf{m};\mathbf{m},t)=C'\exp\left[-\frac{1}{2\epsilon\Delta t}[\Delta\mathbf{m}-\Delta\bar{\mathbf{m}}(t)]\right.$$

$$\left.\times[R(t)]^{-1}[\Delta\mathbf{m}-\Delta\bar{\mathbf{m}}(t)]^T\right],$$

(3.10)

where C' is a normalization constant, and $\mathbf{m}$ is defined by $\mathbf{m}=(m_1,m_2)$ and its transposed vector by $\mathbf{m}^T$. The explicit values of $R_{s,s'}(t)$ are given by

$$R_{11}(t)\Delta t=4[\hat{X}(1)+\hat{X}(2)],$$

$$R_{12}(t)\Delta t=4z\sum_{i=\pm1}i[\hat{Y}_1(i)-\hat{Y}_2(i)]=R_{21}(t)\Delta t,$$ (3.11)

$$R_{22}(t)\Delta t$$

$$=4z^2\sum_{i=\pm1}\hat{X}(i)\left[1+(z-1)\left[\frac{\hat{Y}_1(i)-\hat{Y}_2(i)}{\hat{X}(i)}\right]^2\right].$$

It should be noted that we can show the relation

$$4\begin{vmatrix}\dfrac{\partial^2\bar{G}(t)}{\partial L^2} & \dfrac{\partial^2\bar{G}(t)}{\partial L\partial K}\\[2mm]\dfrac{\partial^2\bar{G}(t)}{\partial K\partial L} & \dfrac{\partial^2\bar{G}(t)}{\partial K^2}\end{vmatrix}=\begin{bmatrix}R_{11}(t) & R_{12}(t)\\R_{21}(t) & R_{22}(t)\end{bmatrix}.$$ (3.12)

IV. THE EVOLUTION OF FLUCTUATION

Since the PPF $\mathscr{P}(t,t+\Delta t)$ essentially represents the transition probability of the entire system from the state $\{y_{ij}(t)\}$ to $\{y_{i'j'}(t+\Delta t)\}$ in Δt, we can consider $\psi(\Delta\mathbf{m};\mathbf{m}t)$ as the transition probability that the system changes from the state specified by $\mathbf{m}$ at t to the state $\mathbf{m}+\Delta\mathbf{m}$ in Δt. In the Markovian process, the probability distribution $W(\mathbf{m},t+\Delta t)$ at time $t+\Delta t$ is connected to $W(\mathbf{m},t)$ at t through the above transition probability[15] by

$$W(\mathbf{m},t+\Delta t)=\int d(\Delta\mathbf{m})\psi(\Delta\mathbf{m};\mathbf{m}-\Delta\mathbf{m},t)W(\mathbf{m}-\Delta\mathbf{m},t).$$

(4.1)

By making use of the mathematical relation that

$$\psi(\Delta\mathbf{m};\mathbf{m}-\Delta\mathbf{m},t)W(\mathbf{m}-\Delta\mathbf{m},t)$$

$$=\exp(-\nabla\cdot\Delta\mathbf{m})\psi(\Delta\mathbf{m};\mathbf{m},t)W(\mathbf{m},t),$$ (4.2)

we expand both sides of Eq. (4.1) with respect to Δt and $\Delta \mathbf{m}$ to obtain, in the limit of $\Delta t \to 0$,

$$\frac{\partial W}{\partial t}(\mathbf{m},t) = -\sum_{s=1}^{2} \frac{\partial}{\partial m_s}[R_s(t)W(\mathbf{m},t)]$$
$$+ \frac{1}{2}\epsilon \sum_{s,s'=1}^{2} \frac{\partial^2}{\partial m_s \partial m_{s'}}[R_{ss'}(t)W(\mathbf{m},t)] \ . \quad (4.3)$$

This is the master equation for the order parameters $\mathbf{m}$ correct up to $O(\epsilon)$. Note that the above equation is correct enough to derive the evolution equations for the fluctuation of the order parameters.[14]

In order to obtain the Fokker-Planck equation for the fluctuation, we again separate the variables into two parts,

$$m_s = \overline{m}_s(t) + \epsilon^{1/2}\eta_s \quad (s=1,2) \ , \quad (4.4)$$

where $\overline{m}_1(t)$ and $\overline{m}_2(t)$ follow the evolution equations Eq. (2.15). By changing independent variables from $(\mathbf{m},t)$ to $(\boldsymbol{\eta},t)$ with the definition

$$W(\mathbf{m},t) = Q(\boldsymbol{\eta},t) \ , \quad (4.5)$$

and by substituting Eq. (4.4) into (4.3), we obtain, in the limit of $\epsilon \to 0$,

$$\frac{\partial Q(\boldsymbol{\eta},t)}{\partial t} = -\sum_s \frac{\partial}{\partial \eta_s}\left[\sum_{s'} \frac{\partial R_s(t)}{\partial \overline{m}_{s'}(t)}\eta_{s'} \cdot Q(\boldsymbol{\eta},t)\right]$$
$$+ \frac{1}{2}\sum_{s,s'} \frac{\partial^2}{\partial \eta_s \partial \eta_{s'}}[R_{ss'}(t)Q(\boldsymbol{\eta},t)] \ , \quad (4.6)$$

where s runs over 1 and 2, and $R_s(t)$ and $R_{ss'}(t)$ are independent of η_s and functions of $\overline{m}_s(t)$ only. This is the Fokker-Planck equation governing the fluctuations of the order parameters. Using this equation, we immediately obtain equations for evolution of fluctuations:

$$\frac{d\langle\langle\eta_s(t)\rangle\rangle}{dt} = \sum_{s'} \frac{\partial R_s(t)}{\partial \overline{m}_{s'}(t)}\langle\langle\eta_{s'}(t)\rangle\rangle \quad (s=1,2) \quad (4.7)$$

and

$$\frac{d\langle\langle\eta_s(t)\eta_{s'}(t)\rangle\rangle}{dt} = \sum_{k=1}^{2}\left[\frac{\partial R_s(t)}{\partial \overline{m}_k(t)}\langle\langle\eta_{s'}(t)\eta_k(t)\rangle\rangle + \frac{\partial R_{s'}(t)}{\partial \overline{m}_k(t)}\langle\langle\eta_s(t)\eta_k(t)\rangle\rangle\right] + R_{ss'}(t) \quad (s,s'=1,2) \ , \quad (4.8)$$

where $\langle\langle\ \rangle\rangle$ is now carried out over η_s with respect to $Q(\boldsymbol{\eta},t)$. These equations are exactly the same as those derived from the MEM, which are given in the Appendix and are thus also identical with those of Saito and Kubo.[12]

V. DISCUSSION

The PPF is a variational function with respect to the path variables, which specify the change of state in Δt. From the nature of the PPF, it contains information not only on the most probable path but also on the fluctuation away from it. In this paper, the equation for the fluctuation is derived by the application of the system size expansion method[14] to the PPF. Since the PPF essentially represents the transition probability of the entire system connecting the state at t and the state at $t + \Delta t$, the development of the system is determined through the PPF on the basis of the assumption of the Markovian process. The differential form of evolution of the system gives the master equation. The Fokker-Planck equation for the fluctuation of the order parameters is also derived in the MEM with the application of the system size expansion to the master equation. It is shown that this Fokker-Planck equation is identical with that obtained by the MEM. Thus the PPM gives the same evolution equations for the average and for the fluctuation of the order parameters as those obtained by the MEM in the pair approximation. This result is particularly significant because it has been considered that it is difficult to derive the evolution equations for the fluctuations by the PPM and because the results indicate that both PPM and the MEM are equivalent.[10] However, as is shown in the following paper,[16] the results of both the methods are not identical in the triangle approximation, and this poses an interesting question whether the disagreements between both methods in the triangle approximation is intrinsic or not.

ACKNOWLEDGMENTS

APPENDIX:
THE MEM IN THE PAIR APPROXIMATION

Although the kinetic Ising model by the MEM has been treated in the pair approximation,[12,17,18] we deal with it here again for the purpose of closely comparing it with the PPM.

The master equation for the total magnetization M_1 and the short-range order parameter M_2 are given by

$$\frac{\partial P}{\partial t}(\mathbf{M},t) = -\int W(\mathbf{M}\to\mathbf{M}';t)P(\mathbf{M},t)\,d\mathbf{M}'$$
$$+ \int W(\mathbf{M}'\to\mathbf{M};t)P(\mathbf{M}',t)\,d\mathbf{M}' \ , \quad (A1)$$

where $\mathbf{M} = (M_1,M_2)$, and $W(\mathbf{M}\to\mathbf{M}';t)$ is the transition probability per unit time at t from the state $\mathbf{M}$ to $\mathbf{M}'$. The variables M_1 and M_2 are related to m_1 and m_2 defined in Eq. (2.3) by the definition $M_s = Nm_s$. When a spin with the state i changes its direction, the total magnetization changes by an amount $\Delta M_1(i) = -2i$ and the total short-range order by $\Delta M_2(i,\{j_s\}) = -2i(j_1+j_2+\cdots+j_z)$, because $\{j_s = \pm 1, \ s=1,\ldots,z\}$ specifies the states of nearest-neighboring spins of the center spin i. We postulate that spin flips occur at each

lattice point independently. Then the transition probability can be written as

$$W(\mathbf{M} \rightarrow \mathbf{M} + \Delta \mathbf{M}; t) = N w(m; \Delta \mathbf{M}(i, \{j_s\}), t) . \quad \text{(A2)}$$

According to the coarse-graining method of the microscopic master equation by van Baal,[18] the transition probability w per spin is made of two parts. The first factor is the probability $p_{z+1}(i, j_1, j_2, \ldots, j_z, t)$ for the spin cluster, and the second factor is made of the energy change of the system in contact with the heat reservoir. The first factor needs a transformation. The probability that a center spin and its nearest neighboring spins take a configuration $(i, j_1, j_2, \ldots, j_z)$ can be written as

$$p_{z+1}(i, j_1, j_2, \ldots, j_z, t) = x_i(t) \prod_{s=1}^{z} \frac{y_{ij_s}(t)}{x_i(t)} . \quad \text{(A3)}$$

This relation is, however, not prescribed in the MEM as in the PPM, and the justification of its use is only that it looks reasonable. We can also interpret this transformation as based on the superposition approximation in the CVM.[19] Therefore, this is a key step in the comparison of the MEM with the PPM. For the second factor, we look at the energy change. When a center spin changes its direction, the effect due to the heat reservoir is assumed to be

$$\exp\left[-\frac{\Delta E(i, \{j_s\})}{2 k_B T} \right] = \exp\left[-i \left[K \sum_{s=1}^{z} j_s + L \right] \right] , \quad \text{(A4)}$$

where $\Delta E(i, \{j_s\})$ is an energy increase of the $z + 1$ cluster associated with the center spin flip. The transition probability w is then written as the product of Eqs. (A3) and (A4) as

$$w(\mathbf{m}; \Delta \mathbf{M}(i, \{j_s\}), t)$$

$$= \theta x_i(t) \exp(-Li) \prod_{s=1}^{z} \frac{y_{ij_s}(t) \exp(-K i j_s)}{x_i(t)} . \quad \text{(A5)}$$

By substituting Eq. (A5) into (A1), we obtain the master equation in the pair approximation

$$\frac{1}{N} \frac{\partial P}{\partial t}(\mathbf{m}, t) = -\text{tr}[w(\mathbf{m}; \Delta \mathbf{M}(i, \{j_s\}), t) P(\mathbf{m}, t)]$$

$$+ \text{tr}[w(\mathbf{m} - \epsilon \Delta \mathbf{M}(i, \{j_s\}); \Delta \mathbf{M}(i, \{j_s\}), t)$$

$$\times P(\mathbf{m} - \epsilon \Delta \mathbf{M}(i, \{j_s\}), t)] , \quad \text{(A6)}$$

where $\text{tr} = \sum_{i = \pm 1} \sum_{j_1, j_2, \ldots, j_z}$ and $P(\mathbf{M}, t)$ is rewritten in terms of $P(\mathbf{m}, t)$. The Kramers-Moyal expansion is obtained from Eq. (A6) as

$$\frac{\partial P}{\partial t}(\mathbf{m}, t) = \sum_{\substack{l_1 = 0 \\ l_1 + l_2 \neq 0}}^{\infty} \sum_{l_2 = 0}^{\infty} (-1)^{l_1 + l_2} \frac{\epsilon^{l_1 + l_2 - 1}}{l_1! l_2!} \frac{\partial^{l_1}}{\partial m_1^{l_1}} \frac{\partial^{l_2}}{\partial m_2^{l_2}} C_{l_1 l_2}(\mathbf{m}, t) P(\mathbf{m}, t) , \quad \text{(A7)}$$

where

$$C_{l_1 l_2}(\mathbf{m}, t) = \text{tr}[[\Delta M_1(i)]^{l_1} [\Delta M_2(i, \{j_s\})]^{l_2}$$

$$\times w(\mathbf{m}; \Delta \mathbf{M}(i, \{j_s\}), t)]$$

$$= \left[2 \frac{\partial}{\partial L} \right]^{l_1} \left[2 \frac{\partial}{\partial K} \right]^{l_2}$$

$$\times \text{tr}[w(\mathbf{m}; \Delta \mathbf{M}(i, \{j_s\}), t)] . \quad \text{(A8)}$$

The last equation is derived by the difference of the energy factor Eq. (A5). However, by noting that the following relation holds:

$$\text{tr}[w(\mathbf{m}; \Delta \mathbf{M}(i, \{j_s\}), t)] = \overline{G}(t) , \quad \text{(A9)}$$

we find that Eq. (A7) leads to

$$\frac{\partial P}{\partial t}(\mathbf{m}, t) = -\sum_{s=1}^{2} \frac{\partial}{\partial m_s}[R_s(t) P(\mathbf{m}, t)]$$

$$+ \frac{\epsilon}{2} \sum_{s, s' = 1}^{2} \frac{\partial^2}{\partial m_s \partial m_{s'}}[R_{ss'}(t) P(\mathbf{m}, t)] + O(\epsilon^2) . \quad \text{(A10)}$$

Up to $O(\epsilon)$, the above master equation is completely identical with Eq. (4.3) derived by the PPM. It is easily shown that when the system size expansion is applied to Eq. (A10) or (A7), the evolution equations for the average and for the fluctuation of order parameters are identical with those obtained in Sec. IV.

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