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# Nonlinearity in Cooperative Systems—Dynamical Bethe–Ising Model

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The dynamics of the short-range order as well as the long-range order in the nonlinear cooperative system is investigated specifically for a kinetic Ising model in the Bethe approximation. The phenomena of critical slowing down near the transition temperature $T_c$ and anomalous fluctuation below $T_c$ are directly related to the instability of the long-range order. The dynamics of the short-range order is essentially a fast mode and is noncritical. However, through the nonlinear coupling the short-range order is also influenced by the critical behavior of the long-range order.

**KEY WORDS**: Kinetic Ising model; quasi-chemical approximation; critical slowing down; anomalous fluctuations far from equilibrium; coupling between order parameters.

## 1. INTRODUCTION

One of the most fascinating and outstanding problems in statistical physics concerns nonlinear phenomena in cooperative systems. The nonlinear nature of the phase transition as an equilibrium process can be seen even in the simplest molecular field approximation,[1] although the real subtleties of the phenomena can be understood only by more elaborate theories such as developed in recent years.[2]

In nonequilibrium, the nonlinearity of cooperativity is manifested in various sorts of dynamical properties.[3–6] Relaxation of order parameters is generally nonlinear, except for small deviations from equilibrium. The nonlinearity is even more pronounced as the system approaches a critical condition. In the very neighborhood of the critical point, even a very small deviation from equilibrium relaxes nonlinearly. Nonlinear relaxation is generally associated with nonlinear response and with anomalous fluctuations. This

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aspect of cooperative phenomena has been treated in a simple way in a previous paper,[4] which will be referred to as KMK.[4] There the Weiss–Ising kinetic model[6] was discussed as a specific example.

In this work we take up the same model as a typical example of cooperative systems and treat it in a better approximation which corresponds to the Bethe–Peierls approximation,[7] or the quasi-chemical approximation,[8] well known in statistical thermodynamics. In the spirit of this approximation, we explicitly consider only a few macrovariables to describe the state of ordering of the cooperative system; namely a macroscopic state is defined by the long-range order and the short-range order parameters, which correspond to the magnetization density and the energy density.[7–9] This is of course an essential limitation of the approximation, which prevents us from any approach to the real singularities appearing at the critical point in equilibrium and nonequilibrium properties. Accepting this sacrifice, however, we can look into the global nature of nonlinear dynamics of a cooperative system in which different order parameters couple to each other through the intrinsic internal interactions. The long-range order, or the magnetization, is directly related to the phase transition, whereas the short-range order is only indirectly related to it. However, generally there should be a complex interplay of their dynamics.

Some years ago Kikuchi[10] treated this problem, concentrating his attention only on the averaged motion of the order parameters. Here we shall extend his treatment to include fluctuations of the order parameters. Taking a somewhat different view from that of Kikuchi, we use the formulations as developed by KMK, which give the evolution equations for the averaged motion and the fluctuation of the order parameters. From these equations, we can easily see how the equilibrium states are determined and how the order parameters are coupled to each other in nonequilibrium states even in far-from-equilibrium conditions.

In Section 2, we describe the model and derive the evolution equations assuming single-spin flips as the underlying basic microscopic mechanism. Equilibrium properties are briefly treated in Section 3. In Section 4, the averaged motion and the fluctuations are investigated under some typical conditions above and below the transition temperature. In the appendix an application to a binary alloy system is briefly discussed in order to compare the present approach to that of Kikuchi.

## 2. EVOLUTION EQUATIONS

We first briefly summarize the main points of KMK relevant to the present work. Extensive macrovariables of a macrosystem are denoted by the set $(X_1, X_2,...) \equiv \{X_i\}$. They are assumed to make a Markovian process to be described by the master equation,

$$(d/dt)P(\{X_i\}, t) = -\sum_{\{r_i\}} W(\{X_i\}; \{r_i\})P(\{X_i\}, t) + \sum_{\{r_i\}} W(\{X_i - r_i\}; \{r_i\})P(\{X_i - r_i\}, t) \tag{1}$$

where $P(\{X_i\}, t)$ is the probability of finding the variables $\{X_i\}$ at these values at time $t$, and $W(\{X_i\}, \{r_i\})$ is the transition probability from the state $\{X_i\}$ to $\{X_i + r_i\}$ with the jumps $\{r_i\}$. Each of the macrovariables is of the order of the size, or the number $N$, of elements in the system, whereas elementary jumps $\{r_i\}$ are only of the order of unity. Since any elementary process occurring at any one of the constituent elements of the whole system induces a transition of the variables $\{X_i\}$, the transition probability $W$ should be proportional to the size $N$, so that we assume

$$W(\{X_i\}; \{r_i\}) = Nw(\{x_i\}; \{r_i\}) \tag{2}$$

where we define the densities

$$x_i = X_i/N \tag{3}$$

corresponding to the extensive variables $\{X_i\}$. The moments of the normalized transition probability are defined by

$$C_{m_1 m_2 \cdots}(\{x_i\}) = \sum_{r_1}\sum_{r_2}\cdots r_1^{m_1} r_2^{m_2}\cdots w(\{x_i\}; \{r_i\}) \tag{4}$$

Then the evolution of the averages

$$\bar{x}_i(t) = \langle x_i \rangle_t \tag{5}$$

and their fluctuations

$$N^{-1}\sigma_{ij}(t) = \langle [x_i - \bar{x}_i(t)][x_j - \bar{x}_j(t)] \rangle_t \tag{6}$$

can be shown to be governed, up to the leading order of $N^{-1}$, by[5]

$$\frac{d}{dt}\bar{x}_i(t) = C_{1_i}(\{\bar{x}_l\}) \tag{7}$$

$$\frac{d}{dt}\sigma_{ij}(t) = \sum_k \frac{\partial C_{1_i}(\{\bar{x}_l\})}{\partial \bar{x}_k}\sigma_{kj} + \sum_k \sigma_{ik}\frac{\partial C_{1_j}}{\partial \bar{x}_k} + C_{1_i 1_j} \tag{8}$$

Now we consider an Ising spin system composed of $N$ spins on a lattice.[6] The number of up spins is denoted by $N_+$, that of down spins by $N_-$, and their difference by $X$; namely

$$N_+ + N_- = N, \qquad N_+ - N_- = X \tag{9}$$

The Ising spins are interacting with the nearest neighbor interaction $J$, which is assumed to be ferromagnetic. The total number of up–up spin pairs is denoted by $N_{++}$, that of down–down pairs by $N_{--}$, and that of up–down pairs by $Q$. These numbers are related by

$$N_{++} + N_{--} + Q = zN/2 \tag{10}$$

$$N_{++} + \tfrac{1}{2}Q = zN_{+}/2 \tag{11}$$

$$N_{--} + \tfrac{1}{2}Q = zN_{-}/2 \tag{12}$$

where $z$ is the coordination number. Thus the total energy of the spin system is given by

$$\begin{aligned} E &= -J(N_{++} + N_{--} - Q) - g\mu_B H(N_{+} - N_{-}) \\ &= -(z/2)NJ + 2JQ - g\mu_B HX \end{aligned} \tag{13}$$

where the first term is the exchange energy and the second term is the Zeeman energy in the presence of an external field $H$. We choose $X$ and $Q$ as the macrovariables to define a macrostate of the spin system; $X$ is essentially the total magnetization and $Q$ is the exchange energy. The latter is also the total area (in three dimensions) or the total length (in two dimensions) of the boundaries between domains or clusters of up spins and down spins. The corresponding densities will be denoted by $x$ and $q$; namely

$$x = X/N, \qquad q = Q/N. \tag{14}$$

The spins are in contact with a heat reservoir, which induces spontaneous flips of spins.[11] If an up spin surrounded by $k$ up spins and $z - k$ down spins is flipped down by thermal agitations, $X$ decreases by two and $Q$ by $2k - z$. The probability per unit time for occurrence of such changes of $X$ and $Q$ is assumed to be

$$W_{+}(X, Q \rightarrow X - 2, Q + 2k - z) = \frac{1}{\tau}\bar{N}_{+}(k)\exp[K(z - 2k) - \mu] \tag{15}$$

Here $\tau$ is a proper rate constant of spin flips determined by the temperature. The exponential factor takes care of the energy change associated with this type of spin flip, $K$ and $\mu$ being

$$K = J/k_B T, \qquad \mu = g\mu_B H/k_B T \tag{16}$$

The second factor $\bar{N}_{+}(k)$ is the average number of up spins with this particular local configuration of their nearest neighbors. Our approximation consists in assuming this to be given by

$$\begin{aligned} \bar{N}_{+}(k) &= N_{+}\binom{N_{++}}{k}\binom{\frac{1}{2}Q}{z-k}\Big/\binom{\frac{1}{2}zN_{+}}{z} \\ &= N_{+}\binom{z}{k}\frac{N_{++}^{k}(Q/2)^{z-k}}{(zN_{+}/2)^{z}} \end{aligned} \tag{17}$$

This means that in a macrostate defined by $X$ and $Q$, an up spin finds $k$ up and $z - k$ down spins as its neighbors with the probability in random distribution of spin pairs under the restriction of the given number of different types of spin pairs on the whole lattice. This assumption is not rigorous, but is an approximation which is equivalent to the Bethe–Peierls method. In fact, in a somewhat different context, the same idea was used by Takagi in his reformulation of the Bethe–Peierls method.[12]

Similarly, for flips of down spins with $k$ down spin neighbors, we assume the transition probability

$$W_-(X, Q \to X + 2, Q + 2k - z) = \frac{1}{\tau} \bar{N}_-(k) \exp[K(z - 2k) + \mu] \tag{18}$$

with

$$\bar{N}_-(k) = N_- \binom{N_{--}}{k} \binom{\frac{1}{2}Q}{z - k} \Big/ \binom{\frac{1}{2}zN_-}{z} \tag{19}$$

Since the variables $X$ and $Q$ are extensive, these transition probabilities have the form (2) when rewritten in terms of the densities $x$ and $q$. So we have

$$w_+(x, q; -2, 2k - z) = n_+ \binom{z}{k} \frac{n_{++}^k (q/2)^{z-k}}{(\frac{1}{2}zn_+)^z} e^{K(z-2k)-\mu} \tag{20}$$

$$w_-(x, q; 2, 2k - z) = n_- \binom{z}{k} \frac{n_{--}^k (q/2)^{z-k}}{(\frac{1}{2}zn_-)^z} e^{K(z-2k)+\mu} \tag{21}$$

where we put

$$n_{++} = N_{++}/N, \qquad n_{--} = N_{--}/N$$

It is now an easy task to write the evolution equations. For this purpose we define

$$C_+ = \sum_k w_+(x, q; -2, 2k - z) = n_+ \left( \frac{n_{++}e^{-K} + \frac{1}{2}qe^K}{\frac{1}{2}zn_+} \right)^z e^{-\mu} \tag{22}$$

$$C_- = \sum_k w_-(x, q; 2, 2k - z) = n_- \left( \frac{n_{--}e^{-K} + \frac{1}{2}qe^K}{\frac{1}{2}zn_-} \right)^z e^{\mu} \tag{23}$$

which are used as the generating functions of the moments, namely

$$C_{m,m'} = \left(2 \frac{\partial}{\partial\mu}\right)^m \left(-\frac{\partial}{\partial K}\right)^{m'} (C_+ + C_-)$$

Equation (7) now reads

$$dx/dt = (2/\tau)(C_- - C_+) \tag{24}$$

and

$$\frac{dq}{dt} = \frac{z}{\tau} \left[ C_+ \left( \frac{n_{++}e^{-K} - \frac{1}{2}qe^K}{n_{++}e^{-K} + \frac{1}{2}qe^K} \right) + C_- \left( \frac{n_{--}e^{-K} - \frac{1}{2}qe^K}{n_{--}e^{-K} + \frac{1}{2}qe^K} \right) \right] \tag{25}$$

where $x$, $q$, and other density variables stand for the corresponding averages.

By the relations (9)–(12), the above equations can be solved for $x$ and $q$. Equation (8) for fluctuations reads

$$\frac{d}{dt}\sigma_{xx} = 2b_{xx}\sigma_{xx} + 2b_{xq}\sigma_{xq} + \frac{4}{\tau}(C_+ + C_-) \tag{26}$$

$$\begin{aligned}\frac{d}{dt}\sigma_{xq} &= b_{qx}\sigma_{xx} + (b_{xx} + b_{qq})\sigma_{xq} + b_{xq}\sigma_{qq} \\ &\quad + \frac{2z}{\tau}\left[-C_+\left(\frac{n_{++}e^{-K} - \frac{1}{2}qe^{K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\right) + C_-\left(\frac{n_{--}e^{-K} - \frac{1}{2}qe^{K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\right)\right]\end{aligned} \tag{27}$$

$$\begin{aligned}\frac{d}{dt}\sigma_{qq} &= 2b_{qx}\sigma_{xq} + 2b_{qq}\sigma_{qq} \\ &\quad + \frac{2z}{\tau}\left\{C_+\left[(z-1)\left(\frac{n_{++}e^{-K} - \frac{1}{2}qe^{K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\right)^2 + 1\right]\right. \\ &\quad \left. + C_-\left[(z-1)\left(\frac{n_{--}e^{-K} - \frac{1}{2}qe^{K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\right)^2 + 1\right]\right\}\end{aligned} \tag{28}$$

where

$$\begin{aligned}\tau b_{xx} &= C_+\left(\frac{z-1}{n_+} - \frac{z^2}{2}\frac{e^{-K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\right) \\ &\quad + C_-\left(\frac{z-1}{n_-} - \frac{z^2}{2}\frac{e^{-K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\right) \\ \tau b_{xq} &= -z(e^{K} - e^{-K})\left(C_+\frac{1}{n_{++}e^{-K} + \frac{1}{2}qe^{K}} - C_-\frac{1}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\right) \\ \tau b_{qx} &= \frac{z}{2}C_+\left[(z-1)\frac{n_{++}e^{-K} - \frac{1}{2}qe^{K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\left(\frac{z}{2}\frac{e^{-K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}} - \frac{1}{n_+}\right)\right. \\ &\quad \left. + \frac{z}{2}\frac{e^{-K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\right] \\ &\quad - \frac{z}{2}C_-\left[(z-1)\frac{n_{--}e^{-K} - \frac{1}{2}qe^{K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\left(\frac{z}{2}\frac{e^{-K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}} - \frac{1}{n_-}\right)\right. \\ &\quad \left. + \frac{z}{2}\frac{e^{-K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\right] \\ \tau b_{qq} &= \frac{z}{2}C_+\frac{1}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\left[(z-1)(e^{K} - e^{-K})\frac{n_{++}e^{-K} - \frac{1}{2}qe^{K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}}\right. \\ &\quad \left. - (e^{K} + e^{-K})\right] + \frac{z}{2}C_-\frac{1}{n_{--}e^{-K} + \frac{1}{2}qe^{K}} \\ &\quad \times\left[(z-1)(e^{K} - e^{-K})\frac{n_{--}e^{-K} - \frac{1}{2}qe^{K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}} - (e^{K} + e^{-K})\right]\end{aligned} \tag{29}$$

Equations (26)–(28) are linear, inhomogeneous equations to determine the evolution of fluctuations, in which the coefficients are functions of time as determined by Eqs. (24)–(25).

## 3. EQUILIBRIUM PROPERTIES

Equilibrium is determined by equating the right-hand sides of Eqs. (24)–(28) to zero. From Eq. (24) we have

$$C_+ = C_- \tag{30}$$

Using this, we get from Eq. (25)

$$q^2/n_{++}n_{--} = 4e^{-4K} \tag{31}$$

which is the quasi-chemical condition of Fowler and Guggenheim.[8] Equation (30) is then rewritten as

$$(n_-/n_+)^{z-1} = e^{2\mu}(n_{--}/n_{++})^{z/2} \tag{32}$$

Equilibrium values of $x$ and $q$ are thus calculated from Eqs. (31) and (32) with the use of the identities corresponding to (9)–(12). This procedure is in fact equivalent to the Bethe–Peierls approximation. In particular, the critical point is found to be

$$k_B T_c = 2J/\log[z/(z-2)] \tag{33}$$

in the absence of external magnetic fields.

The magnetization vanishes above the critical temperature if there is no magnetic field; then the exchange energy density becomes

$$q_e = \frac{z}{2}\frac{1}{1+e^{2K}} \tag{34}$$

Below the critical temperature there will be a spontaneous magnetization. The solution of Eqs. (31) and (32) is expressed as

$$x_e = \tanh\frac{z}{2}\phi$$

$$q_e = \frac{z}{2}\frac{\tanh[(z/2)\phi]}{e^{2K}\sinh[(z-1)\phi + (2\mu/z)]} \tag{35}$$

in terms of the parameter $\phi$ defined by the equation

$$e^{2K} = \frac{\sinh[(z/2)\phi]}{\sinh\{[(z/2)-1]\phi + (2\mu/z)\}} \tag{36}$$

When the magnetic field is smaller than the coercive field, that is to say,

$$|\mu| \leqslant \mu_c$$

Eq. (36) has three real solutions for $\phi$, which correspond to the stable, the unstable, and the metastable states. The marginal value $\mu_c$ is defined by the conditions

$$\left(\frac{z}{2}-1\right)\coth\left[\left(\frac{z}{2}-1\right)\phi_c+\frac{2\mu_c}{z}\right]=\frac{z}{2}\tanh\frac{z}{2}\phi_c$$

and

$$e^{2K}=\frac{\sinh[(z/2)\phi_c]}{\sinh\{[(z/2)-1]\phi_c+(2\mu_c/z)\}} \tag{37}$$

When the magnetic field exceeds $\mu_c$ in magnitude, there is only one stable solution.

Fluctuations are determined by Eqs. (26)–(28). They are related to thermodynamic responses, namely

$$\begin{aligned}\sigma_{xx}&=(\partial x/\partial\mu)_K\\ \sigma_{xq}&=-\tfrac{1}{2}(\partial x/\partial K)_\mu=(\partial q/\partial\mu)_K\\ \sigma_{qq}&=-\tfrac{1}{2}(\partial q/\partial K)_\mu\end{aligned} \tag{38}$$

In the paramagnetic phase, the fluctuations are given simply by

$$\begin{aligned}\sigma_{xx}&=\frac{2}{z}\frac{1}{e^{-2K}-e^{-2K_c}}\\ \sigma_{xq}&=0\\ \sigma_{qq}&=\frac{z}{8}\frac{1}{\cosh^2 K}\end{aligned} \tag{39}$$

In the ferromagnetic phase, they are expressed in terms of the parameter $\phi$ as

$$\sigma_{xx}=\frac{2\,\mathrm{sech}^2[(z/2)\phi]\coth\{[(z/2)-1]\phi+(2\mu/z)\}}{z\coth[(z/2)\phi]-(z-2)\coth\{[(z/2)-1]\phi+(2\mu/z)\}}$$

$$\sigma_{xq}=-\frac{z\,\mathrm{sech}^2[(z/2)\phi]}{z\coth[(z/2)\phi]-(z-2)\coth\{[(z/2)-1]\phi+(2\mu/z)\}}$$

and

$$\sigma_{qq}=\frac{z\tanh[(z/2)\phi]}{2e^{2K}\sinh[(z-1)\phi+(2\mu/z)]}$$
$$\times\left[1-\frac{z\,\mathrm{sech}^2[(z/2)\phi]-2(z-1)\tanh[(z/2)\phi]\coth[(z-1)\phi+(2\mu/z)]}{z\coth[(z/2)\phi]-(z-2)\coth\{[(z/2)-1]\phi+(2\mu/z)\}}\right] \tag{40}$$

## 4. DYNAMICS FAR FROM EQUILIBRIUM

In this section, we discuss the averaged motion and fluctuations under some typical conditions above and below the transition temperature. First the paramagnetic phase is considered, where no instabilities exist. Then we treat the ferromagnetic phase, in which anomalous fluctuations occur due to the existence of an unstable state. The internal coupling between the long-range order and the short-range order is investigated in the nonlinear dynamics of the cooperative system. Finally this coupling is examined in detail in the case where a magnetic field is applied in the direction opposite to the spontaneous magnetization.

### 4.1. Paramagnetic Phase Without an External Field ($T > T_c, H = 0$)

We first consider the relaxation process of magnetization above $T_c$; initially magnetization is set at a finite value and then released to relax. The nonlinear dynamics of the averaged values and the fluctuations are calculated numerically by Eqs. (24)–(28) and they are shown typically in Fig. 1. The initial state is chosen to be far from equilibrium, namely a perfectly ordered state where $x = 1$, $q = 0$, and $\sigma_{xx} = \sigma_{xq} = \sigma_{qq} = 0$.

In order to make the nonlinear effect clear, we first make the linearization approximation around the equilibrium state, given by Eqs. (34) and (39). The linearized equations of motion are given by

$$\frac{d}{dt}\,\delta x = -\frac{1}{\tau_s}\,\delta x \tag{41}$$

$$\frac{d}{dt}\,\delta q = -\frac{1}{\tau_f}\,\delta q \tag{42}$$

$$\frac{d}{dt}\,\delta\sigma_{xx} = -\frac{2}{\tau_s}\,\delta\sigma_{xx} + \frac{1}{\tau}\frac{32}{z}\frac{e^{-K}\sinh K}{(\cosh K)^{z-2}}\frac{1}{e^{-2K} - e^{-2K_c}}\,\delta q \tag{43}$$

$$\frac{d}{dt}\,\delta\sigma_{xq} = -\left(\frac{1}{\tau_s} + \frac{1}{\tau_f}\right)\delta\sigma_{xq} - \frac{e^{-2K}}{2\tau} \times \frac{(z-1)^2\sinh^3 K - (z+1)\cosh K\sinh^2 K - 2\sinh K}{(\cosh K)^{z+1}(e^{-2K} - e^{-2K_c})}\,\delta x \tag{44}$$

$$\frac{d}{dt}\,\delta\sigma_{qq} = -\frac{2}{\tau_f}\,\delta\sigma_{qq} - \frac{4(z-2)}{\tau}\frac{\sinh K}{(\cosh K)^{z-1}}\,\delta q \tag{45}$$

where

$$\frac{1}{\tau_s} = \frac{1}{\tau}\frac{z(e^{-2K} - e^{-2K_c})}{(\cosh K)^z} \tag{46}$$

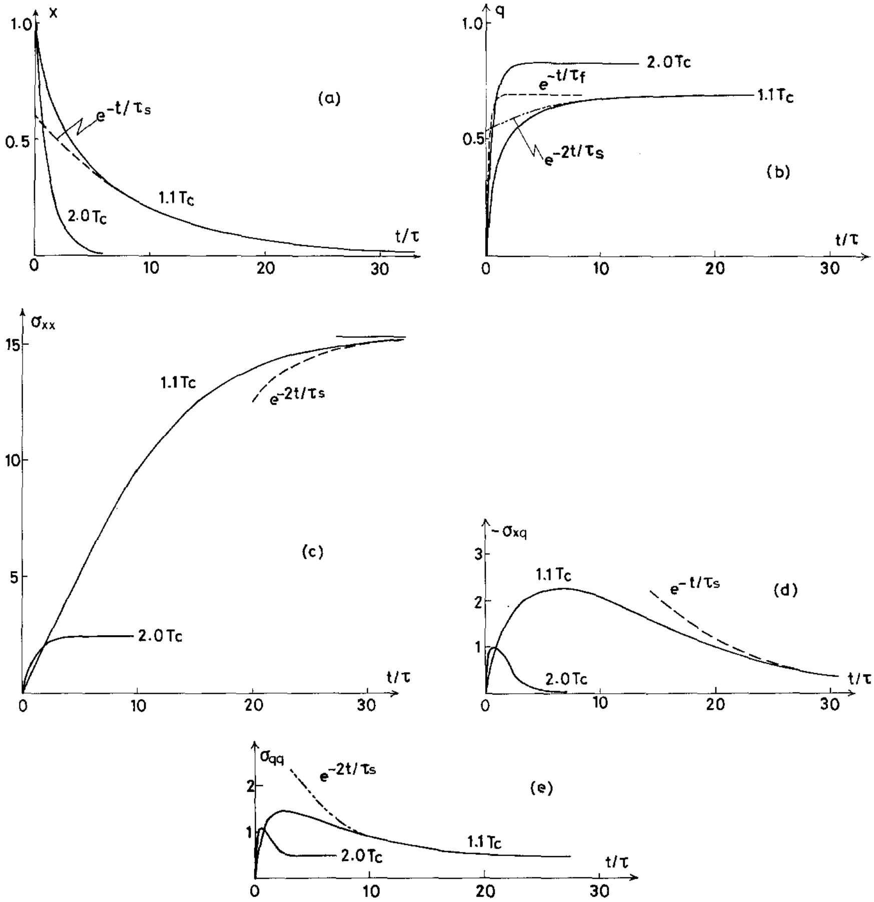


Fig. 1. The relaxation of averaged values $x$ and $q$ and fluctuations $\sigma_{xx}$, $\sigma_{xq}$, and $\sigma_{qq}$ at $T = 2.0\ T_c$ and $1.1\ T_c$. Here $z = 4$ and $k_B T_c = 2J/\log 2$. The dashed lines represent the exponential behavior determined by Eqs. (41)–(45) at $T = 1.1\ T_c$. Now $1/\tau_s$ and $1/\tau_f$ are 0.11 and 3.63, respectively. The dot-dot-dashed lines represent the nonlinear equation (50).

and

$$\frac{1}{\tau_f} = \frac{1}{\tau}\frac{4}{(\cosh K)^{z-2}} \tag{47}$$

Exponential relaxations according to the linear approximation are shown at $T = 1.1T_c$ by dashed lines in Fig. 1. In Figs. 1a and 1c, there appears critical slowing down in $x$ and $\sigma_{xx}$, which can be represented well by the linearized treatment with a relaxation time $\tau_s$. Initial rapid relaxation in Fig. 1a represents the deviation from the linear approximation and is due to the nonlinear effect.[13]

In the linearized scheme, there appears additionally a noncritical short relaxation time $\tau_f$ corresponding to the short-range order. This short relaxation may be attributed to the fast relaxation process of the small clusters, which are easily perturbed by the surrounding reservoir. These two time scales, $\tau_s$ and $\tau_f$, are already found in the computer simulation of the Ising system,[14] where the spin correlation between nearest neighbors is found to relax more rapidly than the magnetization does.

In the evolution of $q$, $\sigma_{xq}$, and $\sigma_{qq}$, however, there is actually found the phenomenon of critical slowing down (Figs. 1b, 1d, and 1e). Evidently the linear approximation is insufficient for the short-range order near the transition temperature. The short-range order follows adiabatically the slow motion of the long-range order through the nonlinear coupling.

We have to supplement the motion (42) with the nonlinear term to get

$$\frac{d}{dt}\,\delta q = -\frac{1}{\tau_f}\,\delta q - A\,\delta x^2 \tag{48}$$

where

$$A = \frac{z(z-1)}{\tau}\,\frac{\sinh K}{e^{2K}(\cosh K)^{z-1}} \tag{49}$$

Near the transition temperature, where $\tau_f \ll \tau_s$, $q(t)$ is adiabatically determined by $x(t)$ as

$$q(t) = \frac{z}{2}\,\frac{1}{1+e^{2K}}\left[1 - \frac{z-1}{2}\,(1 - e^{-2K})x^2(t)\right] \tag{50}$$

and relaxes slowly with the relaxation time $\tau_s/2$, exhibiting a critical long tail. This tail is also induced in $\sigma_{qq}$, and both are shown in Figs. 1b and 1c, by the dot-dot-dashed lines. The relation (50) between the long-range order and the short-range order is slightly different from the molecular field relation $q = (z/4)(1 - x^2)$. The solution (50) is nothing but the adiabatic solution ($\dot{q} = 0$) of Eq. (25) expanded in powers of $x$. The critical dynamic behavior of the short-range order is ascribed to the adiabatic nonlinear coupling with the long-range order.

### 4.2. Ferromagnetic Phase Without an External Field ($T < T_c, H = 0$)

Next we consider the growth of magnetization below $T_c$. If the system is initially kept at a temperature higher than $T_c$ and is then suddenly cooled below $T_c$, it will stay in a state with zero magnetization, because that state is stationary though unstable. In order for the system to reach the symmetry-broken state with a spontaneous magnetization, we make the system start from a state with some small magnetization. The dynamics far from equilibrium is shown in Fig. 2, where the initial state is taken to be $x = 0.0001$, $q = 0$, and $\sigma_{xx} = \sigma_{xq} = \sigma_{qq} = 0$.

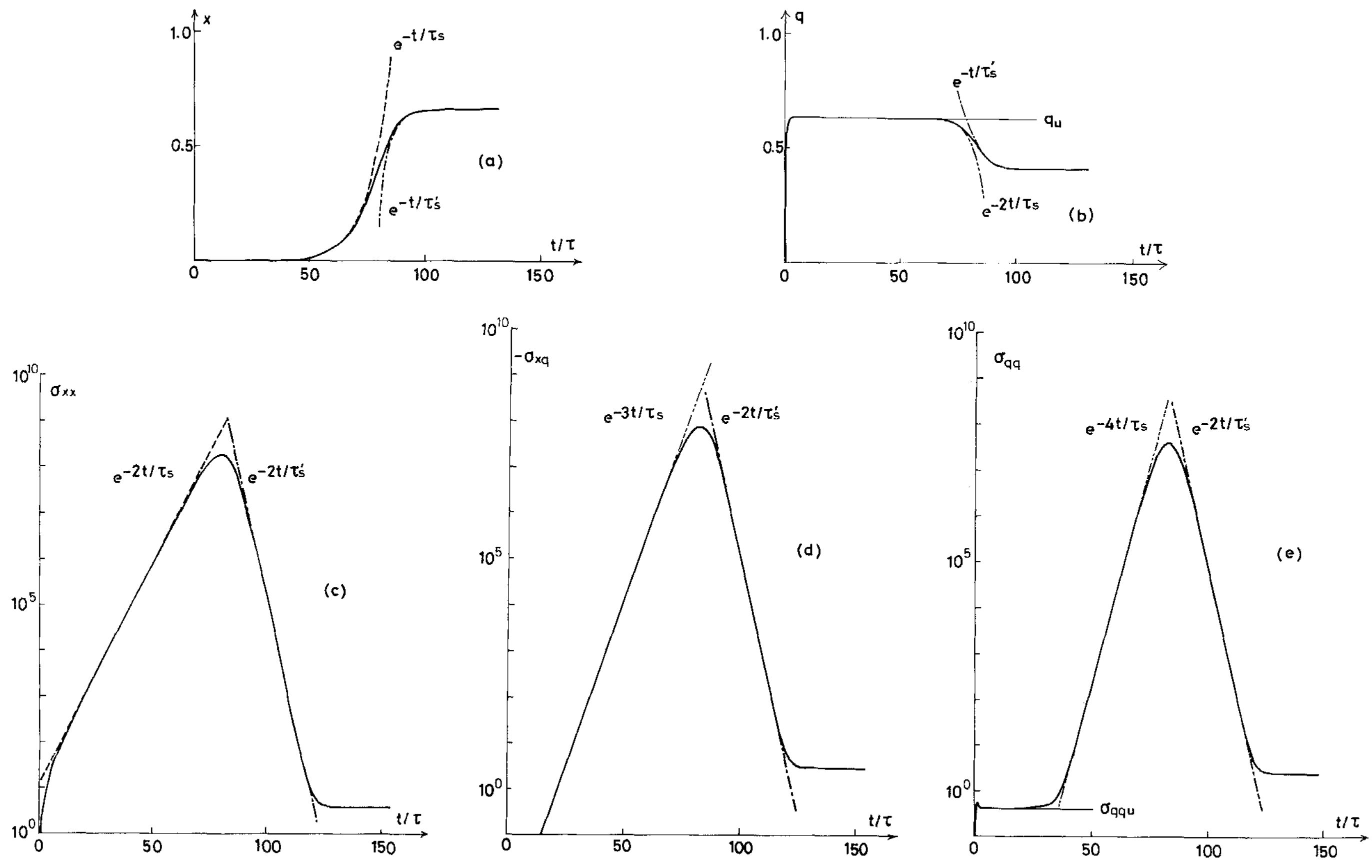


Fig. 2. Averaged motions $x$ and $q$ and fluctuations $\sigma_{xx}$, $\sigma_{xq}$, and $\sigma_{qq}$ at $T = 0.9\ T_c$. The dashed lines represent the exponential behavior given by Eqs. (41)–(45) around the unstable point, where $1/\tau_s = -0.11$. The dot-dashed lines represent the exponential behavior given by Eqs. (52), (53), and (59) around the stable equilibrium point, where $1/\tau_s' = 0.25$. The dot-dot-dashed lines represent the adiabatic behavior given by Eqs. (50), (56), and (57).

Around the unstable stationary state, given by Eqs. (34) and (39), the evolution of the system is very interesting. Due to the instability of the system, $\tau_s$ is now negative and $x$ increases exponentially. Meanwhile, $q$ relaxes rapidly to its unstable value $q_u$ given by Eq. (34), as is shown in Fig. 2b. Thereafter, $q$ is driven adiabatically away from $q_u$ through the nonlinear coupling (50) with $x$. This adiabatic motion of $q$ is represented by dot-dot-dashed line in Fig. 2b. As $x$ grows further, the nonlinearity in the evolution of $x$ becomes important, and the system relaxes to its stable equilibrium state, given by Eqs. (35) and (40).

Near the equilibrium state without an external field, $x$ and $q$ are no longer good linear modes. Eigenmodes are given, up to the order of $\phi^2$ near the transition temperature, by

$$\begin{aligned} y_s &= [1 - O(\phi^2)]x + \phi q \\ y_f &= \tfrac{1}{4}z(z-2)\phi x + [1 - O(\phi^2)]y \end{aligned} \tag{51}$$

Deviations of the averaged values from those at equilibrium obey equations in the linearized from

$$\begin{aligned} (d/dt)\delta y_s &= -(1/\tau_s')\,\delta y_s \\ (d/dt)\delta y_f &= -(1/\tau_f')\,\delta y_f \end{aligned} \tag{52}$$

where

$$\frac{1}{\tau_s'} = \frac{1}{\tau}\frac{z^{z/2}(z-2)^{1+(z/2)}}{3(z-1)^{z-1}}\,\phi^2 \tag{53}$$

represents the critical slow relaxation time, and

$$\frac{1}{\tau_f'} = \frac{4}{\tau}\left[\frac{z(z-2)}{(z-1)^2}\right]^{(z/2)-1}\left(1 + \frac{z^2 - 2z + 5}{24}\,\phi^2\right)$$

According to this linear approximation, both $x$ and $q$ relax critically and slowly with relaxation time $\tau_s'$, as are shown by the dot-dashed lines in Figs. 2a and 2b.

Looking at the fluctuations, we can find their anomalous enhancements. Since the initial value $\sigma_{xx}(t = 0)$ ($\geqslant 0$) is far from the unstable stationary value (39), which is negative, the fluctuation of the long-range order $\sigma_{xx}(t)$ grows large and steadily, proportional to $\exp(-2t/\tau_s)$. This causes an "anomalous fluctuation" around the unstable state. This anomalous enhancement corresponds to the critical divergence of the susceptibility at equilibrium associated with the emergence of the order or cooperativity in the system.

At equilibrium the fluctuation of the short-range order $\sigma_{qq}$ or specific heat does not diverge but only has a jump at the transition point. On the other hand, dynamically an anomalous enhancement is found even in $\sigma_{qq}$.

This anomaly is induced from the nonlinear coupling with the long-range order. Since $\sigma_{xx}(t)$ increases anomalously, we can not neglect the nonlinearity in the evolution. We have to supplement the motion (44) and (45) with the nonlinear terms

$$\frac{d}{dt}\,\delta\sigma_{xq} = -\left(\frac{1}{\tau_s}+\frac{1}{\tau_f}\right)\delta\sigma_{xq} - 2A\,\delta\sigma_{xx}\,\delta x \tag{54}$$

$$\frac{d}{dt}\,\delta\sigma_{qq} = -\frac{2}{\tau_f}\,\delta\sigma_{qq} - 4A\,\delta\sigma_{xq}\,\delta x \tag{55}$$

Near the transition temperature, where $\tau_s \gg \tau_f$, fluctuations follow adiabatically the variation of $x$ and $\sigma_{xx}$. These adiabatic fluctuations determined by Eqs. (54) and (55) are given by

$$\sigma_{xq}(t) \sim -\tfrac{1}{2}z(z-1)e^{-2K}(\tanh K)\sigma_{xx}(t)x(t) \tag{56}$$

and

$$\sigma_{qq}(t) \sim \tfrac{1}{4}z^2(z-1)^2e^{-4K}(\tanh^2 K)\sigma_{xx}(t)x(t)^2 \tag{57}$$

These are nothing but the approximate solutions of Eqs. (27) and (28) in the adiabatic case, $\dot\sigma_{xq} = 0$ and $\dot\sigma_{qq} = 0$, neglecting the slow ($b_{xx}$) and less anomalous terms,

$$\sigma_{xq} = -(b_{qx}/b_{qq})\sigma_{xx}, \qquad \sigma_{qq} = -(b_{qx}/b_{qq})\sigma_{xq} \tag{58}$$

As $x$ increases slowly ($\propto e^{-t/\tau_s}$) and the fluctuation $\sigma_{xx}$ increases anomalously in proportion with $e^{-2t/\tau_s}$, the fluctuations in the short-range order are enhanced as $\sigma_{xq} \sim e^{-3t/\tau_s}$ and $\sigma_{qq} \sim e^{-4t/\tau_s}$, which agree with the enhancements shown by dot-dot-dashed curves in Figs. 2d and 2e.

Intermediately the mode recombination sets in through the nonlinear effect, and finally fluctuations approach the equilibrium values. This final relaxation process is well approximated by the linearized treatment around the equilibrium state (40):

$$\begin{aligned} d(\delta\sigma_{ss})/dt &= -2(\tau_s')^{-1}\,\delta\sigma_{ss} + U_1\,\delta y_s + U_2\,\delta y_f \\ d(\delta\sigma_{sf})/dt &= -[(\tau_s')^{-1} + (\tau_f')^{-1}]\,\delta\sigma_{sf} + U_3\,\delta y_s + U_4\,\delta y_f \\ d(\delta\sigma_{ff})/dt &= -2(\tau_f')^{-1}\,\delta\sigma_{ff} + U_5\,\delta y_s + U_6\,\delta y_f \end{aligned} \tag{59}$$

where the $U$'s are the time-independent and temperature-dependent coefficients, their explicit forms being complicated and not so important. The linear approximation (59) is shown by the dot-dashed lines in Figs. 2c–2e, and fits well with the numerical calculations.

### 4.3. Ferromagnetic Phase with an External Magnetic Field ($T < T_c, H = 0$)

In order to see the adiabatic character of the motion of the short-range order and the nonlinear coupling between the long-range order and the short-range order more clearly, we apply a magnetic field to reverse the magnetization through the value zero. Initially the system is kept in an equilibrium state at some temperature below $T_c$ without an external field. Then we apply a magnetic field $H$ or $\mu$, larger than the marginal field $H_c$ or $\mu_c$, in the direction opposite to the initial magnetization.[15] Examples of the computed temporal evolution of the system are shown in Fig. 3.

In the averaged motion of $x$, we find a flat region before $x$ turns down. The system tries to remain in the "metastable"[15] state. The system, however, is actually unstable, and accordingly the fluctuation $\sigma_{xx}$ increases anomalously through this "latent" period.

If we assume that the short-range order follows adiabatically the motion of the long-range order, we can expand the adiabatic relation $\dot{q} = 0$, given by Eq. (25), with nonzero magnetic field in powers of $x$ and we get

$$q(t) = \frac{z}{2}\frac{1}{1 + e^{2K}}\{1 - (\tanh\mu)x - \frac{1 - e^{-2K}}{2}[(z - 1) - z\tanh^2\mu]x^2 + \cdots\} \tag{60}$$

which agrees completely with the temporal behaviors of $q$, shown in Fig. 3b. For comparison, we calculated the short-range order given by the molecular field scheme, $q = (z/4)(1 - x^2)$, which is shown by the thin line in Fig. 3b.

The fluctuations $\sigma_{xq}$ and $\sigma_{qq}$ increase through the adiabatic nonlinear coupling with $\sigma_{xx}$. Concentrating on the anomalous behavior determined by $\sigma_{xx}$, we obtain adiabatic approximations for $\sigma_{xq}$ and $\sigma_{qq}$ with the magnetic field, which are expanded in powers of $x$ as follows

$$\sigma_{xq} = -\Big[\frac{z}{4}\frac{e^{-K}}{\cosh K}\tanh\mu + \frac{z(z - 1)}{2}e^{-2K}(\tanh K)(\operatorname{sech}^2\mu + \tfrac{1}{2}\tanh^2\mu)x + \cdots\Big]\sigma_{xx} \tag{61}$$

$$\sigma_{qq} = \sigma_{xq}^2/\sigma_{xx}$$

This adiabatic behavior is shown by dot-dot-dashed lines in Figs. 3d and 3e, and they fit very well with exact calculations: As $x$ crosses zero and reverses its sign, $\sigma_{qq}$ becomes small and $\sigma_{xq}$ reverses its sign. Since the crossing occurs very rapidly, the adiabatic approximation becomes poor in this region; in fact deviations of the adiabatic curves from the exact ones are noticeable there. As a whole, however, the adiabatic nature of the short-range order is obvious.

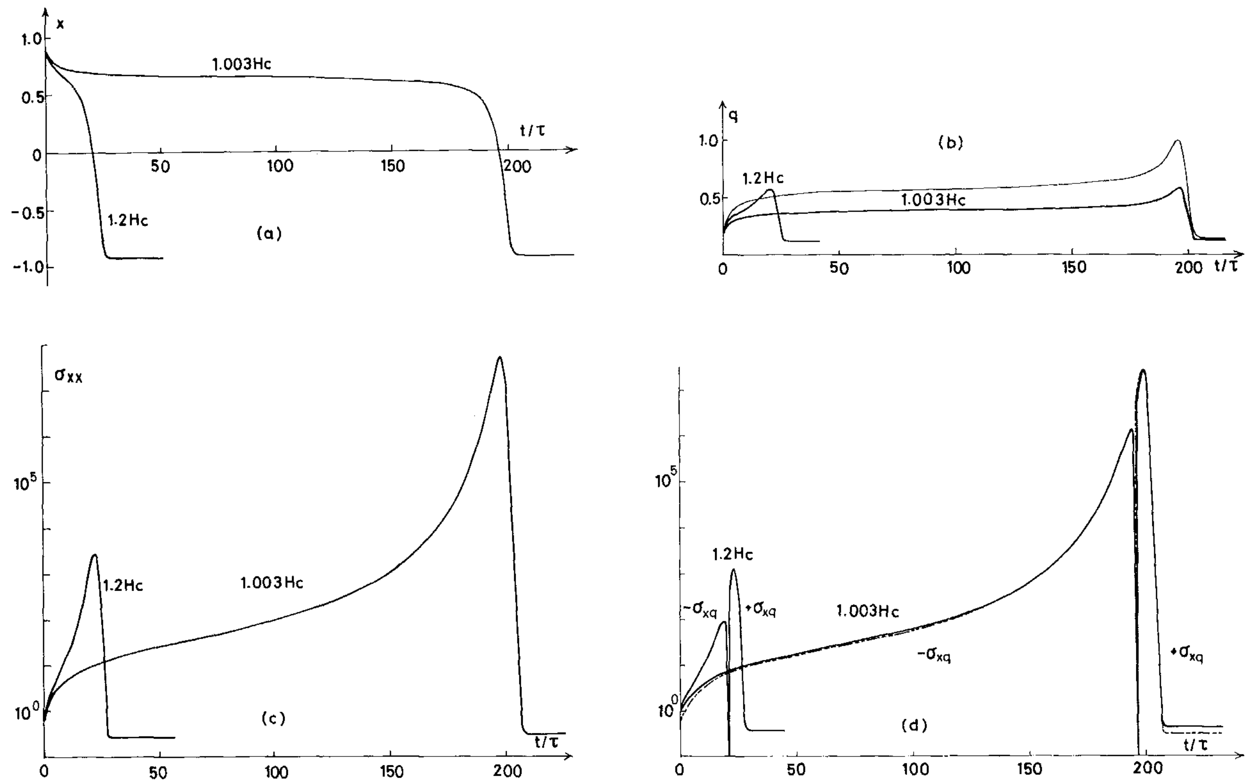


Fig. 3. Averaged motions $x$ and $q$ and fluctuations $\sigma_{xx}$, $\sigma_{xq}$, and $\sigma_{qq}$ at $T = 0.76\ T_c$, when the magnetic field $H = 1.003\ H_c$ or $1.2\ H_c$ is applied after $t = 0$. Here $H_c(T) = 0.0996 k_B T / g\mu_B$. The dot-dot-dashed lines represent the adiabatic motion given by Eqs. (51). The thin line represents the short-range order given by $q = (z/4)(1 - x^2)$.

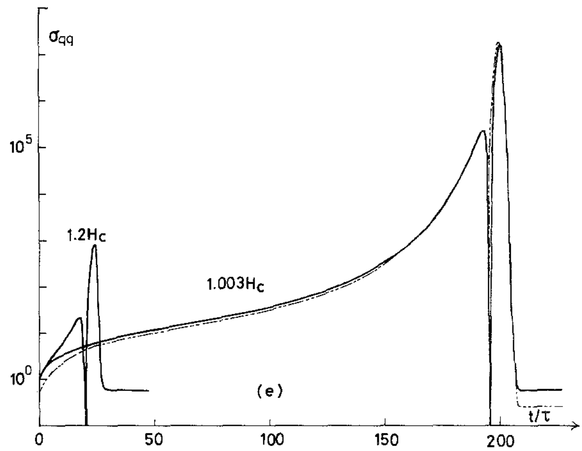


Fig. 3. Continued.

## 5. CONCLUSIONS

We have investigated the dynamical behavior of the kinetic Ising system taking into account the short-range order $q$ as well as the long-range order $x$. The short-range order is not directly related to the phase transition or the cooperative change of state, so that it is characterized by a fast relaxation time which has no critical slowing down. As the result of nonlinear coupling between the long-range and short-range order parameters, the dynamics of the latter is affected by the former. Such nonlinear coupling was here treated by a set of evolution equations and was illustrated by examples computed from these equations. It was shown that the short-range order parameter almost follows the slow and critical motion of $x$: $q$ is induced into its critical nature through the nonlinear coupling with $x$.

This adiabatic nonlinear coupling is clearly seen also in the fluctuations. The fluctuation of $x$ exhibits an anomalous enhancement, indicating the instability of the system. This enhancement in $\sigma_{xx}$ adiabatically causes the fluctuations $\sigma_{xq}$ and $\sigma_{qq}$ to increase. This enhancement is seen even in the "metastable" region, when an external magnetic field is applied. These features found for the specific model of Ising spins should be quite general for a wide class of cooperative systems.

## APPENDIX

In this appendix, we give a simple derivation of the basic equations for the order–disorder transition of $A$–$B$ alloys. Those for the most probable values were already derived by Kikuchi. We derive the evolution equations for fluctuations as well.

**Table I. Definition of Spin Variables in Terms of Atomic Configurations, Their Numbers, and Associated Chemical Potential**

| Spin | Configuration on sublattice | | Number | Chemical potential |
|---|---|---|---|---|
| | $a$ | $b$ | | |
| + | $A$ | — | $N_A{}^a = N_+/2$ | $\mu_A{}^a = \mu_+$ |
| + | — | $B$ | $N_B{}^b = N_+/2$ | $\mu_B{}^b = \mu_+$ |
| − | $B$ | — | $N_B{}^a = N_-/2$ | $\mu_B{}^a = \mu_-$ |
| − | — | $A$ | $N_A{}^b = N_-/2$ | $\mu_A{}^b = \mu_-$ |

The lattice, consisting of $a$ and $b$ sublattices, is occupied by an $A$ or a $B$ atom at each site. When we consider 50–50 alloys, there are equal numbers of $A$ atoms on $a$ sublattices, $N_A{}^a$, and $B$ atoms on $b$ sublattices, $N_B{}^b$; we identify these atoms as up spins. The $A$ atoms on $b$ sublattices and $B$ atoms on $a$ sublattices can also be identified as down spins (see Table I). Similarly we represent the nearest-neighboring atomic pairs in terms of spins as shown in Table II. The numbers $N$ follow the same relations as Eqs. (10)–(12).

A chemical potential for each configuration and pair interaction energies are tabulated in Tables I and II, respectively. Then the total energy of the system is given by

$$E = -J(N_{++} + N_{--} - Q) - g\mu_{\mathrm{B}}HX + \text{const}$$

where

$$J = \tfrac{1}{2}(\epsilon_{AA} + \epsilon_{BB} - 2\epsilon_{AB}) > 0$$

and

$$g\mu_{\mathrm{B}}H = \tfrac{1}{2}(\mu_+ - \mu_-)$$

An atom interchanges its site with one of its nearest neighbors by thermal fluctuation. In spins terms, these transition processes are tabulated in Table III. Taking the configurations surrounding the pair into account, we can

**Table II. Definition of Pair Numbers and Interaction Energies**

| Spin pair | Configuration on sublattice | | Number | Interaction energy |
|---|---|---|---|---|
| | $a$ | $b$ | | |
| + − | $A$ | $A$ | $N_{AA}^{ab} = Q/2$ | $\epsilon_{AA}$ |
| − + | $B$ | $B$ | $N_{BB}^{ab} = Q/2$ | $\epsilon_{BB}$ |
| + + | $A$ | $B$ | $N_{AB}^{ab} = N_{++}$ | $\epsilon_{AB}$ |
| − − | $B$ | $A$ | $N_{BA}^{ab} = N_{--}$ | $\epsilon_{BA} = \epsilon_{AB}$ |

**Table III. Exchange Processes in Terms of Spin Variables**

| | | Configuration on | | | | |
|---|---|---|---|---|---|---|
| | | sublattice | | | sublattice | |
| Process | Spin | $a$ | $b$ | | $a$ | $b$ |
| I | $+ + \to - -$ | $A$ | $B$ | $\to$ | $B$ | $A$ |
| II | $- - \to + +$ | $B$ | $A$ | $\to$ | $A$ | $B$ |

assume the transition probabilities for each process. Process I occurs with an exchange probability

$$W_{++}(X, Q \to X - 4, Q - 2z + 2k + 2p + 2)$$
$$= \frac{1}{\tau} N_{++}\left[\binom{N_{++}}{k}\binom{\frac{1}{2}Q}{z - k - 1}\Big/\binom{\frac{1}{2}zN_{+}}{z - 1}\right]$$
$$\times \left[\binom{N_{++}}{p}\binom{\frac{1}{2}Q}{z - p - 1}\Big/\binom{\frac{1}{2}zN_{+}}{z - 1}\right] e^{-K(2k+2p+2-2z)-2\mu}$$

where $p + 1$ and $k + 1$ are the numbers of up spins surrounding the initial up spins. For process II, the exchange probability $W_{--}$ is given by

$$W_{--}(X, Q \to X + 4, Q + 2z - 2k - 2p - 2)$$
$$= \frac{1}{\tau} N_{--}\left[\binom{N_{--}}{z - k - 1}\binom{\frac{1}{2}Q}{k}\Big/\binom{\frac{1}{2}zN_{-}}{z - 1}\right]$$
$$\times \left[\binom{N_{--}}{z - p - 1}\binom{\frac{1}{2}Q}{p}\Big/\binom{\frac{1}{2}zN_{-}}{z - 1}\right] e^{K(2k+2p+2-2z)+2\mu}$$

where $p$ and $k$ are the numbers of up spins around the initial down spins.

We can employ again the method of KMK in this case. Using the same notation as in Sections 2–4, we obtain the averaged motions as well as their fluctuations:

$$\frac{dx}{dt} = -\frac{4}{\tau}(\eta_{++} - \eta_{--}) \equiv C(x) \tag{A.1}$$

$$\frac{dq}{dt} = \frac{2(z-1)}{\tau}\left(\eta_{++}\frac{n_{++}e^{-K} - \frac{1}{2}qe^{K}}{n_{++}e^{-K} + \frac{1}{2}qe^{K}} + \eta_{--}\frac{n_{--}e^{-K} - \frac{1}{2}qe^{K}}{n_{--}e^{-K} + \frac{1}{2}qe^{K}}\right) \equiv C(q) \tag{A.2}$$

$$\frac{d}{dt}\sigma_{xx} = 2\frac{\partial C(x)}{\partial x}\sigma_{xx} + 2\frac{\partial C(x)}{\partial q}\sigma_{xq} + 2\frac{\partial C(x)}{\partial \mu} \tag{A.3}$$

$$\frac{d}{dt}\sigma_{xq} = \frac{\partial C(q)}{\partial x}\sigma_{xx} + \left(\frac{\partial C(x)}{\partial x} + \frac{\partial C(q)}{\partial q}\right)\sigma_{xq} + \frac{\partial C(x)}{\partial q}\sigma_{qq} - \frac{\partial C(x)}{\partial K} \tag{A.4}$$

and

$$\frac{d}{dt}\sigma_{qq} = 2\frac{\partial C(q)}{\partial x}\sigma_{xq} + 2\frac{\partial C(q)}{\partial q}\sigma_{qq} - \frac{\partial C(q)}{\partial K} \tag{A.5}$$

where we have put

$$\eta_{++} = n_{++}\left(\frac{n_{++}e^{-K} + \frac{1}{2}qe^{K}}{\frac{1}{2}zn_{+}}\right)^{2(z-1)} e^{-2\mu}$$

and

$$\eta_{--} = n_{--}\left(\frac{n_{--}e^{-K} + \frac{1}{2}qe^{K}}{\frac{1}{2}zn_{-}}\right)^{2(z-1)} e^{2\mu}$$

Equations (A.1) and (A.2) for the averaged values coincide with those derived by Kikuchi. Further, we have also derived a set of evolution equations (A.3)–(A.5) for fluctuations.

The equilibrium properties are the same as those in Section 3. By linearizing the evolution equations around this equilibrium state, we obtain evolution of the same form as in Sections 4.1 and 4.2. Only the relaxation times are modified somewhat by a constant factor:

$$\tau_s^{-1} = \tau^{-1}z^2e^K(\cosh K)^{1-2z}(e^{-2K} - e^{-2K_c})$$

and

$$\tau_f^{-1} = \tau^{-1}2(z-1)e^K(\cosh K)^{3-2z}$$

in the paramagnetic phase ($T > T_c$), and

$$\frac{1}{\tau_s'} = \frac{1}{\tau}\frac{z^{z+1}(z-2)^z}{3(z-1)^{2(z-1)}}\phi^2$$

and

$$\frac{1}{\tau_f'} = \frac{1}{\tau}\frac{2(z-2)^{z-2}z^{z-1}}{(z-1)^{2(z-2)}}$$

in the ferromagnetic phase near the transition temperature ($T < T_c$ and $\phi \sim 0$). The temporal behavior far from equilibrium is similar to the results shown in Figs. 1–3.

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