Review

Phase transition in spin systems with various types of fluctuations

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Abstract: Various types ordering processes in systems with large fluctuation are overviewed. Generally, the so-called order–disorder phase transition takes place in competition between the interaction causing the system be ordered and the entropy causing a random disturbance. Nature of the phase transition strongly depends on the type of fluctuation which is determined by the structure of the order parameter of the system. As to the critical property of phase transitions, the concept "universality of the critical phenomena" is well established. However, we still find variety of features of ordering processes. In this article, we study effects of various mechanisms which bring large fluctuation in the system, *e.g.*, continuous symmetry of the spin in low dimensions, contradictions among interactions (frustration), randomness of the lattice, quantum fluctuations, and a long range interaction in off-lattice systems.

Keywords: phase transition, critical phenomena, frustration, quantum effect, randomness, long-range interacting system

1. Introduction

The phase transition is one of the most significant phenomena in the nature.[1] The most familiar phase transition is the boiling phenomenon of the water. The state of the water changes at the boiling temperature (≃100°C), *i.e.*, from the liquid to the gas. The microscopic description of the system does not change at all at this point. Namely, the interaction among molecules is given by a normal non-singular form which does not depend on the temperature. However, the macroscopic property shows the singular dependence on the temperature. This dependence was first explained by the van der Waals equation of state. This equation takes the interaction between molecules into the equation of state of the ideal gas (Boyle–Charles law). Thus, it became clear that the interaction is important for the phase transition. But it was still mystery that the singular behavior of phase transition can be explained by the statistical mechanics. In the canonical ensemble, all the thermodynamic properties are derived from the partition function which consists of analytic functions of the temperature, *i.e.* $e^{-E_i/k_{\rm B}T}$, where E_i is an energy of a state i, T is the temperature, and $k_{\rm B}$ is the Boltzmann constant.

This singular property was one of the main topics of the statistical physics in the early twenty century.[2] L. Onsager finally succeeded to obtain the exact form of the free energy in the thermodynamic limit, and showed a divergence of the specific heat.[3] This work first demonstrated the intrinsic difference between the so-called microscopic state and the macroscopic state.

Since then, natures of phase transitions have been extensively studied, and various properties have been clarified. One of the most significant properties is the universality of the criticality. This indicates that the critical property of systems only depends on the so-called relevant characteristics, such as the spatial dimensionality of the system, symmetry of the order parameter, and the range of interaction. The concept of the universality has been supported by the idea of the renormalization group.[4] However, in the two dimensions, it has turned out that infinite different types of critical properties exist in the studies on the exactly solvable models,[5),6] and also

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on the conformal field theory.[7] There the concept of "relevance" of quantities became rather unclear.

In three dimensions, so far we knew little on the variety of the criticality. At present, the relevant quantities in three dimensions are only the dimensionality of spin and the range of interaction. Most models which show the second-order phase transitions in the two dimensions with different critical exponents exhibit the first order phase transition. For example, the case of the three-state Potts model in three dimension was very marginal,[8),9)] but it has turned out to have a weak first-order transition.[10)] Thus, the approach to classify the second order phase transition is not very powerful in three dimensions. There may be various unknown types of ordered states in three dimensions, and a new concept for classification would be necessary.

The phase transition is understood as competition between the interaction which causes the order and the thermal fluctuation (the entropy) which causes random configurations. Beside this fundamental mechanism of phase transition, ordering processes show various aspects depending on the structure of the order parameter of the system. In this paper, we study several examples of ordering processes in systems with some mechanisms for large fluctuations.

There are various types of models, such as spin models with Ising, XY and Heisenberg spins, Potts model, and the clock model, *etc.* They are expressed in the following form

$$\mathcal{H} = -\sum_{\langle ij\rangle} J_{ij}X_iX_j - H{\sum}' X_i, \qquad [1]$$

where X_i represents a local quantities such as the spin at the position of i, and H is an external field conjugate to the order parameter. Here $\langle ij\rangle$ denotes the interacting pairs and J_{ij} is the coupling constant for the pair, and ${\sum}'$ denotes the sum for the order parameter $M = {\sum}' X_i$. We assume that the local quantities are on lattice points which are fixed except in the last section.

Critical property is studied by investigating the order parameter

$$\langle M\rangle = \frac{\mathrm{Tr}Me^{-\beta\mathcal{H}}}{\mathrm{Tr}e^{-\beta\mathcal{H}}}, \qquad [2]$$

and its fluctuation

$$\langle M^2\rangle - \langle M\rangle^2 = \frac{\mathrm{Tr}M^2e^{-\beta\mathcal{H}}}{\mathrm{Tr}e^{-\beta\mathcal{H}}} - \langle M\rangle^2, \qquad [3]$$

which corresponds to the response to the conjugate field H, *i.e.*, $\chi = dM/dH$. Type of the critical property is characterized by the singularity at a critical point T_C. For example, the susceptibility diverges as

$$\chi = \frac{\langle M^2\rangle - \langle M\rangle^2}{k_\mathrm{B}T} \propto |T - T_\mathrm{C}|^{-\gamma}, \qquad [4]$$

and the spontaneous order parameter m_S appears as

$$m_\mathrm{S} = \lim_{H\to 0}\lim_{N\to\infty}\frac{\langle M\rangle}{N} \propto |T_\mathrm{C} - T|^{\beta}, \qquad [5]$$

which represents the symmetry breaking of the system. The specific heat C shows a singular dependence as

$$C = \frac{\langle \mathcal{H}^2\rangle - \langle \mathcal{H}\rangle^2}{k_\mathrm{B}T^2} \propto |T - T_\mathrm{C}|^{-\alpha}. \qquad [6]$$

Here, the exponents α, β and γ are called "critical exponents".

As an example, we show the critical properties of the two-dimensional Ising model on the square lattice which is the most well-known and a prototype system for the phase transition:

$$\mathcal{H}_\mathrm{Ising} = -J\sum_{\langle ij\rangle}\sigma_i\sigma_j - H\sum_i \sigma_i, \qquad [7]$$

where $\sigma = \pm 1$ and $\langle ij\rangle$ denotes all the nearest neighbor pairs on the lattice. In Fig. 1, we depict the specific heat obtained by Onsager[3)] by the thin curves. There, we see the divergence at the critical point $k_\mathrm{B}T_\mathrm{C}/J = (2/\ln(1+\sqrt{2}) \simeq 2.269\cdots$.

In the figure, we also show the temperature dependence of the spontaneous magnetization obtained by C. N. Yang[11)]

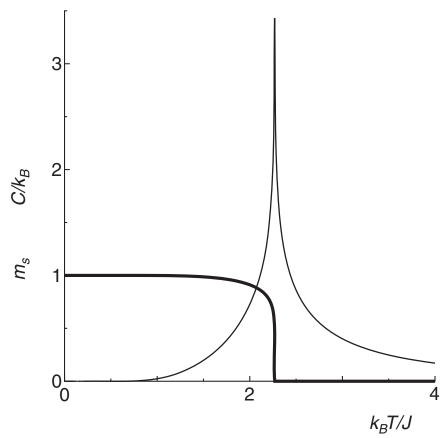

Fig. 1. Temperature dependences of the specific heat (thin line) and of the spontaneous magnetization of the ferromagnetic two-dimensional Ising model [7] with $H = 0$.

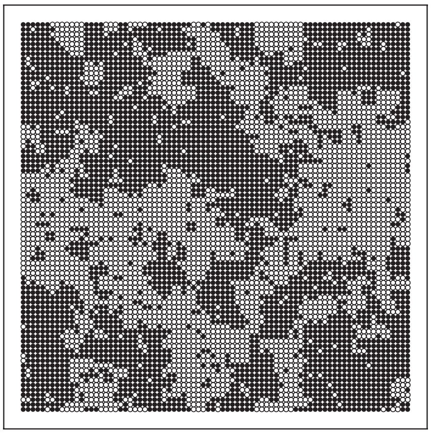

Fig. 2. A snapshot of equilibrium spin configuration obtained by a Monte Carlo method near the critical point $T = 2.3/J$ and $H = 0$ (*cf.* $T_{\mathrm{C}}/J \sim 2.269\cdots$). The solid and open circles denote $\sigma_i = 1$ and $\sigma_i = -1$, respectively.

$$m_s = \left(1 - \frac{1}{\sinh^4 2K}\right)^{1/8}. \qquad [8]$$

The free energy of the Ising model has been obtained by various methods,[3),5),12)–14)] and a new field of the so-called "exactly solvable model" has been developed.[15)]

In Fig. 2, we plot a typical spin configuration near the critical temperature where we find large domains. The linear dimension of the domain corresponds to the correlation length ξ

$$\langle S_i S_j \rangle \sim \frac{1}{r^{d-2+\eta}} e^{-r_{ij}/\xi} \quad \text{for large } r_{ij}, \qquad [9]$$

where r_{ij} is the distance between the sites i and j, and η is the exponent so-called anomalous dimension. The divergence of the correlation length is expressed as

$$\xi \sim |T - T_{\mathrm{C}}|^{-\nu} \qquad [10]$$

with $\nu = 1$. It is known that the exponent η is $1/4$ for the present model. At the critical point, fluctuation of the order parameter becomes large which results in the divergence of χ. Although the susceptibility has not yet been obtained analytically, it has been established that it diverges with the exponent $\gamma = 7/4$ of Eq. [4]. This divergence of χ corresponds to the correlation length ξ. The exponent ν is related to γ and η as

$$\gamma = (2 - \eta)\nu. \qquad [11]$$

This is an example of the scaling relations among the critical exponents.[16)]

For the Ising ferromagnet, the ordered state is a simple ferromagnetic state and ordering process is rather straightforward. However, when the system has additional sources of fluctuation, the phase transition shows various interesting aspects of the ordering. In this paper, we study characteristics of phase transitions of systems in which the fluctuation is enhanced by various reasons, such as the continuous symmetry of the order parameter which causes the so-called infrared divergence of fluctuation in low dimensions ($d = 1$ and 2), contradictions among the interactions (frustration), randomness of the lattice, and quantum fluctuation. We also study the effect of the long range interaction induced by an elastic interaction in off-lattice systems.

2. XY model (a continuous spin symmetry)

In the Ising model, the boundary of the two phases is given by a domain wall. On the other hand, when the spin has a continuous symmetry, *e.g.*, the XY model with $\boldsymbol{S}_i = (\cos\theta_i, \sin\theta_i)$:

$$\mathcal{H}_{\mathrm{XY}} = -J \sum_{\langle ij \rangle} \cos(\theta_i - \theta_j), \qquad [12]$$

the direction of the spin can change smoothly, and the domain wall is not formed. Thus, the fluctuation easily occurs, and it causes the so-called infrared divergence of the fluctuation, and the long range order can not appear in two dimensions.[17)] Although the long range order does not exist in the two-dimensional XY model, it has been known that the system exhibits a peculiar critical phenomenon which is called "Kosterlitz–Thouless transition".[18)]

At low temperatures, this system can be approximated by a harmonic system, $\mathcal{H} \simeq J/2 \sum_{\langle ij \rangle} (\theta_i - \theta_j)^2$, where the periodicity of the angle θ is not relevant. The spin correlation function decays by a power law[19)]

$$\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle \sim r_{ij}^{-\eta}, \qquad [13]$$

with a temperature dependent exponent $\eta = k_{\mathrm{B}}T/4J$ in the harmonic approximation. In contrast, the correlation function decays exponentially at high temperatures, where the periodicity of the potential $\cos(\theta_i - \theta_j)$ is relevant. This difference is explained by using the picture of vortex-pair association. The vortex is characterized by the vortex number n defined by

$$\oint d\theta = 2n\pi. \qquad [14]$$

Because n can be any integer, this type of vortex is called Z vortex. But vortices with large n cause high energies, and thus the vortices with $n = \pm 1$ are important. We call them '±' vortex, respectively. The vortex represents a topological defect in configurations of the system with O(1) symmetry representing the periodicity of the angle.

Appearance of single vortex indicates the relevance of the periodicity, and thus it is a symbol of the high temperature phase. On the other hand, in the low temperature phase the interaction is so strong that configurations reflecting the periodicity of the angle cannot appear. Thus, at low temperatures, the periodicity of the angle is irrelevant. This fact is expressed by the absence of single vortex, which means essentially no vortex. However, at finite temperatures, thermal fluctuation may still cause the appearance of them in a form of pair of '±' vortices which is localized in the space. This change from the free single vortices to the bounded pair vortices is called "vortex association" at the Kosterlitz–Thouless transition. In equilibrium configurations of the XY model, it is hard to identify vortices clearly. At high temperatures, configurations are too much disturbed to identify the vortices, and at low temperatures, the probability for well-recognize vortex pair is very low.[20] In Fig. 3, we depict a typical configuration with vortices, where the '±' vortices are shown by open and solid circles, respectively. It should be noted that this is a transient configuration from a random configuration to an aligned one at a low temperature.

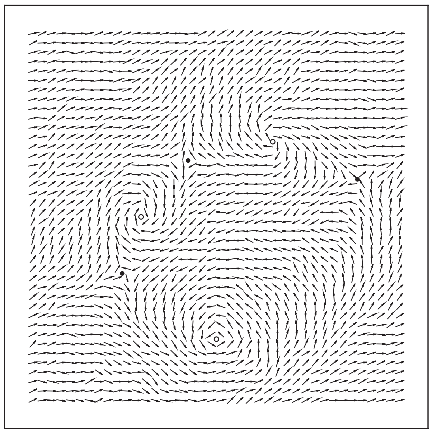

Fig. 3. A vortex configuration of the two-dimensional XY model [12] in a transient process of ordering. The open circles denote the '+' vortices and the closed circles '−' vortices.

The model [12] is transformed to the so-called solid-on-solid (SOS) model by the dual transformation.[21)–23)] Using the following identity relation

$$Z = \mathrm{Tr} e^{K\sum_{\langle ij\rangle}\cos(\theta_i-\theta_j)} = \int_0^{2\pi} d\theta_i \prod_{\langle ij\rangle}\sum_{s_{ij}=-\infty}^{\infty} e^{is_{ij}(\theta_i-\theta_j)}\int_0^{2\pi}\frac{d\phi}{2\pi}e^{-is_{ij}\phi+K\cos(\phi)}, \quad [15]$$

and integrating over θ_i, the partition function is given by

$$Z = \prod_{\langle ij\rangle}\sum_{s_{ij}=-\infty}^{\infty}\left(\prod_k \delta_{\sum_{m:\text{ around } k} s_{km},0}\right) I_{s_{ij}}(K), \quad [16]$$

where

$$I_s(K) = \int_0^{2\pi}\frac{d\phi}{2\pi}e^{-is\phi+K\cos(\phi)} \simeq \frac{1}{\sqrt{2\pi K}}e^{-s^2/2K} \quad [17]$$

for large K. If we introduce the dual variables

$$s_{km} = h_k - h_m, \quad [18]$$

then the condition $\sum_{m:\text{ around } k} s_{km} = 0$ is automatically satisfied. Here, we have to be careful to assign the suffix k and m. Thus, the partition function is expressed by the integer variable $\{h_i\}$

$$Z = \sum_{h_i=-\infty}^{\infty}\prod_{\langle ij\rangle} I_{h_i-h_j}(K) \simeq \left(\frac{1}{2\pi K}\right)^{zN/2}\sum_{h_i=\infty}^{\infty} e^{-\frac{1}{2K}\sum_{\langle ij\rangle}(h_i-h_j)^2}, \quad [19]$$

where z is the number of the nearest neighbor sites. This partition function can be interpreted as that a system of the integer variable $\{h_i\}$:

$$\mathcal{H}_{\mathrm{SOS}} = \frac{1}{2}\sum_{\langle ij\rangle} J(h_i - h_j)^2 \quad [20]$$

at the temperature J/kT. Regarding h_i as the height of solid, this model is called the solid-on-solid (SOS) model. It is used to study the crystal growth where h_i denotes the height of the surface at the position i.[24] Thus, we find that the planar model and the SOS model are in the dual relation.[23] The SOS model is used to study phase transitions of the surface structures such as the roughening transition[25] and facet transition.[26]

Instead of using the planar model, if we use the Villain model $\mathcal{H}_V$[21]

$$e^{-\beta\mathcal{H}_{\mathrm{V}}} = \sum_{m=-\infty}^{\infty} e^{\frac{1}{2}\sum_{\langle ij\rangle} K(\theta_i-\theta_j-2m\pi)^2}, \quad [21]$$

we have exactly

$$Z = \sum_{h_i=-\infty}^{\infty} e^{-\frac{1}{2K}\sum_{\langle ij\rangle}(h_i-h_j)^2}. \qquad [22]$$

The model is further transformed by using an identity expression (Poisson sum):

$$Z = \sum_{h_i=-\infty}^{\infty} \int_{-\infty}^{\infty} d\phi_i e^{-\frac{1}{2K}\sum_{\langle jk\rangle}(\phi_j-\phi_k)^2+\sum_j 2\pi i h_j \phi_j}. \qquad [23]$$

If we perform the Gauss integration over ϕ, we find that only the case of $\sum_j h_j = 0$ is relevant, and there the partition function has the form

$$Z = Z_0 \sum_{h_i} e^{-a\sum_i h_i^2 - 2\pi K \sum_{i\neq j} h_i h_j \ln(r_{ij})}, \qquad [24]$$

where Z_0 and a are constants independent of $\{h_i\}$. This model is a neutral two-dimensional Coulomb system of particles with the charges $\{h_i\}$.[27)]

3. Frustration I: Ising spin systems

In the models so far studied, the interaction tends to cause a perfect alignment of the spins. So, all the interactions work cooperatively, and how the system is ordered is well defined. However, if there are contradictions among the interactions, the ordering process becomes not trivial, and various interesting ordering phenomena occur. Although phase transitions in the frustrated or competing interactions have been studied for a long time, the concept of frustration was introduced by Toulouse[28)] in the study of the spin glass.

In this section, we study the case of frustrated Ising spin systems in regular lattices.[29)] As a typical example, let us consider spins interacting antiferromagnetically on a triangle lattice (Fig. 4(a)). On the triangle, many degenerate ground states exist, that is, six states give the ground state for the antiferromagnetic case while two states (all up or down) for the ferromagnetic case. If we consider a larger lattice, number of the ground states in the antiferromagnetic case increases while that of the ferromagnetic model remains two. It has been known by exact calculations[30)] that this antiferromagnetic Ising model does not have a critical point at finite temperatures, and the ground state has a macroscopic degeneracy W, and thus the system has nonzero entropy at $T = 0$:

$$S(T=0) = k_B \ln W \simeq 0.338 N k_B, \qquad [25]$$

where N is the number of the lattice sites. Similar properties have been obtained in the square version of the fully frustrated model (Villain model[31)] (Fig. 4(b))). It is known that the spin correlation

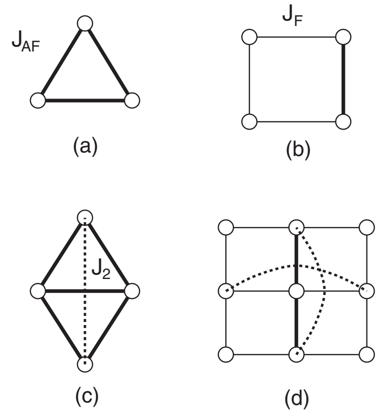

Fig. 4. Fully frustrated lattices. The bold lines denote antiferromagnetic bonds and the thin lines ferromagnetic bonds. (a) Antiferromagnetic triangular lattice. (b) The Villain lattice (Fully frustrated square lattice. (c) The antiferromagnetic model on the triangular lattice with a next nearest neighbor interaction (dotted line). (d) The Villain model with next nearest neighbor interactions (dotted lines).

function of this type of models decays in a power law[32),33)]

$$\langle \sigma_i \sigma_j \rangle \sim r_{ij}^{-1/2}, \qquad [26]$$

which means that the critical temperature is 0 in these models.

In the followings, we study phase transitions in this type of highly frustrated systems, for which some additional interaction is necessary to extract some ordered configurations from the highly degenerate configurations.

3.1. Phase transitions induced by next-nearest neighbor interactions in the fully-frustrated Ising model. Although no phase transition takes place in the fully frustrated Ising models, successive phase transitions were observed in experiments ($CsCoCl_3$).[34)] In order to explain these phase transitions, Mekata introduced next-nearest neighbor (nnn) interactions (Fig. 4(c)), and found successive phase transitions by a mean-field theory. In particular, he found a phase in which a part of spins remain disorder due to the frustration and he called this phase "partially disordered (PD) phase". (Fig. 5(a)). At a lower temperature, the remaining spins order and form a kind of ferrimagnetic phase. Because of the new feature of the successive phase transitions, detailed investigations on this type of models have been done.

First, models in two dimensions were studied. The present author proposed an exactly solvable model with a next nearest neighbor (nnn) interaction in the fully frustrated square lattice (Fig. 4(d)). It was pointed out that the model without the nnn interaction (the original Villain model) is transformed into an exactly solvable symmetric eight vertex model,[5),33)] and it was shown that the spin correlation function in the ground state shows the power law decay [26]. Following the above mentioned Mekata's idea, we expect that, by setting the nnn interaction, the sublattice order is enhanced and the model exhibits a phase transition. The model with the nnn interaction only on one of the sublattices can still be transformed into the eight vertex model as well as the Villain model. It was shown that the model with the nnn interaction has a finite critical temperature as a function the strength (say J_2) of the nnn interaction.[35)] The critical temperature decreases with J_2 and vanishes at $J_2 = 0$. Moreover, it was shown that the critical exponent of the specific heat α (Eq. [6]) of this system changes continuously with J_2 reflecting the peculiar property of the eight vertex model.[5)] If the frustration is strong, *i.e.*, for small J_2, the critical exponent α has a negative value with a large absolute value, which means a smooth change of the specific heat although it is still singular. This feature is natural and suggestive for phase transitions of frustrated models. But the feature of phase transition is rather different from that of the original Mekata problem.

More direct studies of the antiferromagnet on the triangular lattice of the Ising spin (AFTI) with nnn interactions were performed. Reflecting the symmetry of the ground state, the model was mapped to the six-state clock model by making use of the relation depicted in Fig. 5(b).[36)–39)] In two dimensions, the six-state clock model,

$$\mathcal{H} = -J\cos(\theta_i - \theta_j), \quad \theta_i = \frac{\pi}{3}n, n = 0, \ldots 5, \qquad [27]$$

is known to have two KT-type phase transitions which are dual to each other, although the systems with $n \leq 4$ have a unique self-dual transition. The intermediate temperature phase is a massless phase in which the correlation function decays in a power law.[40)] Thus, one may consider that the PD phase can be understood in an analogy to this intermediate KT phase.

To study properties in three dimensions, the standard three dimensional six-state clock model [27] was also studied. It turned out that the nature of fluctuation is different from that of the two dimensions. It was shown that the model has no intermediate temperature phase though it shows a large intermediate crossover temperature region.[41),42)]

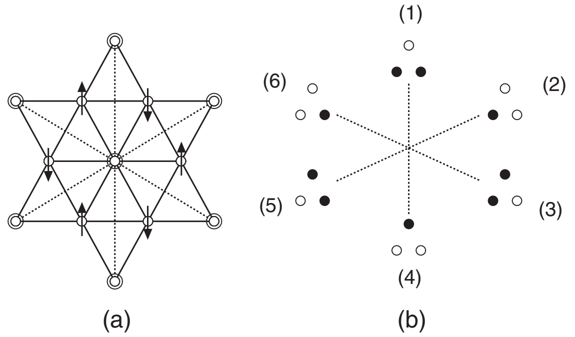

Fig. 5. (a) The partially disordered (PD) state in the antiferromagnetic Ising model with a next nearest neighbor interaction (Mekata model). The circles denote the disordered sites. (b) Correspondence between the six-fold ground state of the Mekata model and the six-state clock model.

Thus, the PD phase was regarded as the intermediate temperature region with a large fluctuation.

But, finally it was discovered that the generalized six-state clock model has a new type of ordered structure. Although the antiferromagnets on the triangular lattice (AFT) has the six-fold symmetry (Fig. 5(b)), the structure of energy levels for the states is not necessarily given by the standard six-state clock model [27], but it may have a generalized form

$$\mathcal{H} = \varepsilon_1\delta_{n_i,n_j\pm1} + \varepsilon_2\delta_{n_i,n_j\pm2} + \varepsilon_3\delta_{n_i,n_j\pm3}. \qquad [28]$$

In the case $\varepsilon_1 = \varepsilon_2 = \varepsilon_3 > 0$, the model corresponds to the six-state Potts model, which exhibits a first-order phase transition.[9)]

It was discovered that, in the case $\varepsilon_1 \ll \varepsilon_2 \simeq \varepsilon_3$, the model has a new type of intermediate-temperature phase in which two neighboring states mix microscopically.[43)] In Fig. 6, we depict a snap shot of the intermediate phase. There we set different boundary conditions in the left and right sides. At the left boundary, the states are restricted to the states 1 and 2 (left) and at the right the states 2 and 3 (right). There are a mixed phase of the states 1 and 2 in the left and that of 2 and 3 in the right. Between them, we find a sharp domain boundary which indicates the stability of the mixed states.

This state really possesses the property of the PD phase proposed by Mekata,[44)] because, if we average two neighboring structures in Fig. 5, we have a disordered site in one of the three sites. Thus, finally it was proved that there exists an ordered state corresponding to the Mekata's PD phase in three dimensions. This type of phase with mixed states is not known so far, and gives a new type of ordered state.

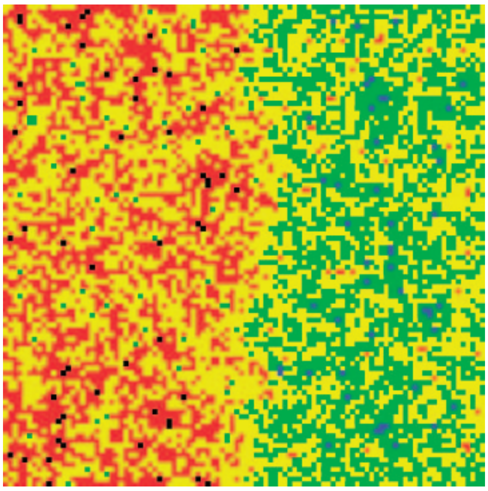

Fig. 6. Domain structure of mixed states of the generalized six-state clock model [28] with $\varepsilon_1 = 0.1J$ and $\varepsilon_2 = \varepsilon_3 = J$. The six states ($n = 1, \sim 6$) are plotted by red, yellow, green, blue, gray, and black squares, respectively. The left part is a mixed phase of $n = 1$ and 2, and the right part is a mixed phase of $n = 2$ and 3. The lattice is a cubic lattice with $L = 100$ and $T = 0.4J$. An intersection of the lattice is depicted.

In three dimensions, it has been pointed out a rich structure of the ordered states, so-called "devil's stair case" or "devil's flower", appears in the ferromagnetic Ising model with a nearest neighbor interaction in one direction (ANNNI model).[45] These observations indicate that various unknown rich structures of ordered states exist in higher dimensions for the future studies.

3.2. Reentrant phase transition. Another interesting property of the frustrated systems is the non-monotonic ordering process due to the peculiar distribution of degeneracy. The non-monotonicity is understood by the idea of the decorated bond.[46),47)] A typical example of the decorated bond is depicted in Fig. 7. There, two spins, σ_1 and σ_2, which we call the system spin, are connected by a direct bond (J_0) and two paths with the spins s_i ($i = 1$ and 2) which we call the decoration spin. The Hamiltonian of the decorated bond is given by

$$\mathcal{H}_{\text{decoratedbond}} = J_0\sigma_1\sigma_2 - J_1(\sigma_1 + \sigma_2)(s_1 + s_2). \quad [29]$$

The effective interaction $K(T_{\text{eff}})$ between the system spins is given by

$$Z_0 e^{-K(T)_{\text{eff}}\sigma_1\sigma_2} = \mathrm{Tr}_{s_1,s_2} e^{-\mathcal{H}_{\text{decoratedbond}}/k_{\mathrm{B}}T}, \quad [30]$$

where Z_0 is a factor independent of σ_i. At high temperatures, because of the entropy effect, the contribution to the effective interaction through the decoration spins is weak (*i.e.*, $\tanh^{-1}(\tanh^2 \beta J_1) \propto (\beta J_1)^2$). On the other hand, at low temperatures, the effective coupling constant is approximately given by the sum of interactions of all the three paths, *i.e.*, $K(T) \simeq (2J_1 - J_0)/k_{\mathrm{B}}T$.

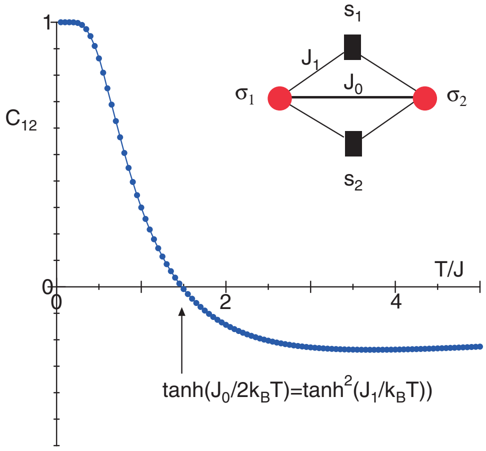

Fig. 7. Temperature dependence of the correlation function $\langle\sigma_1\sigma_2\rangle = \tanh(K_{\text{eff}}(T))$ of the decorated bond [29]. Inset shows the bond structure, where the circles denote the system spins σ_1 and σ_2, and the squares the decoration spins s_1 and s_2.

Here, we consider a competing case, *e.g.*, $J_0 = 2J$ and $J_1 = 1.5J$ with a unit of energy J. Because the direct path is antiferromagnetic ($J_0 > 0$), the effective interaction is antiferromagnetic at high temperatures $K(T) < 0$. At low temperatures, it is ferromagnetic because $2J_1 > J_0$. In Fig. 7, temperature dependence of the correlation function $C_{12} = \langle\sigma_1\sigma_2\rangle = \tanh K_{\text{eff}}(T)$ is plotted. The effective coupling disappears at the temperature T_0 at which $\tanh(J_0/2k_{\mathrm{B}}T_0) = \tanh^2(J_1/k_{\mathrm{B}}T_0)$.

We can also provide more complicated types of reentrant phase transitions in exactly solvable two-dimensional Ising models.[48)] This reentrant behavior can give a mechanism of temperature dependent configuration of ordered states in random systems, where interesting phenomena such as memory and rejuvenation take place.[49)] It is also known that this type of decoration causes a kind of screening effect on the spin state at each site and stabilize it, and thus the spin dynamics becomes very slow.[50)]

4. Frustration II: Continuous spin systems

4.1. XY model on the triangular lattice. So far we studied the frustrated Ising model where the frustration causes degeneracy of the ground state. However, in the case of continuous spin systems, the degeneracy of the ground state can be resolved in a non-collinear spin structure. The continuous degree of freedom allows the system to have a spiral structure.[51),52)] The structure of the spiral state is obtained by the Fourier transformation of the interaction

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j = \sum_{\boldsymbol{k}} J(\boldsymbol{k}) \boldsymbol{S}_{\boldsymbol{k}} \cdot \boldsymbol{S}_{-\boldsymbol{k}}. \quad [31]$$

The minimum point of $J(\boldsymbol{k})$ gives the period of the helical structure. For an antiferromagnet on the triangular lattice, the unit cell consists of three spins (three sublattices), and $J(\boldsymbol{k})$ is given by

$$J(\boldsymbol{k}) = J\left(\cos(k_x) + \cos\left(\frac{k_x}{2} + \frac{\sqrt{3}k_y}{2}\right) + \cos\left(\frac{k_x}{2} - \frac{\sqrt{3}k_y}{2}\right)\right), \quad [32]$$

and it has minima at two points $(k_x, k_y) = \pm(\frac{2\pi}{3}, \frac{2\pi}{3\sqrt{3}})$ which correspond to the degree of the chirality (see Eq. [33]), while it has a maximum at $(k_x, k_y) = (0, 0)$ which corresponds to the ground state in the ferromagnetic case.

The ground state of the antiferromagnetic XY model on a triangle is given by the configurations depicted in Fig. 8. The ground state has no more macroscopic degeneracy as the case of Ising model. There remains a non-trivial two-fold degeneracy as depicted in Figs. 8(a) and (b) which are called ±120° structure, respectively.[53),54)] Thus, the symmetry of the ground state is given by $\mathrm{Z}_2 \times \mathrm{S}_1$, *i.e.*, the two-fold degeneracy and the trivial degeneracy of the rotation around the *z*-axis. Amplitude of the local ±120° structure is described by the quantity "chirality"[54)]

$$\kappa = \frac{2}{3\sqrt{3}} (\boldsymbol{S}_i \times \boldsymbol{S}_j + \boldsymbol{S}_j \times \boldsymbol{S}_k + \boldsymbol{S}_k \times \boldsymbol{S}_i). \quad [33]$$

This is a scalar variable for the XY spin system, and it causes an Ising-like critical property. The rotational degree of freedom gives a Kosterlitz–Thouless transition of the spin order. Therefore, we expect two phase transitions in this system.[54)] There were long discussions on the problem which phase transition takes place at higher temperature. Nowadays, it is almost settled that the chirality orders first (at a higher temperature) as discussed in the first paper of this problem.[55)]

4.2. Heisenberg model on the triangular lattice. In the isotropic Heisenberg spin model, the symmetry of the ground state configuration is more complicated. The ground state is given by a three-dimensional chiral vector and this structure is characterized by the three-dimensional rotation group SO(3) or the projective space P_3.[56)] This structure has a new type of vortex (Z_2 vortex) as the point defect (the homotopy group classification $\pi_1(\mathrm{P}_3) = \mathrm{Z}_2$[57)]). The phase transition of this system was discussed by using the Wilson loop in an analogy of the quark-confinement problem in the lattice gauge theory.[58)]

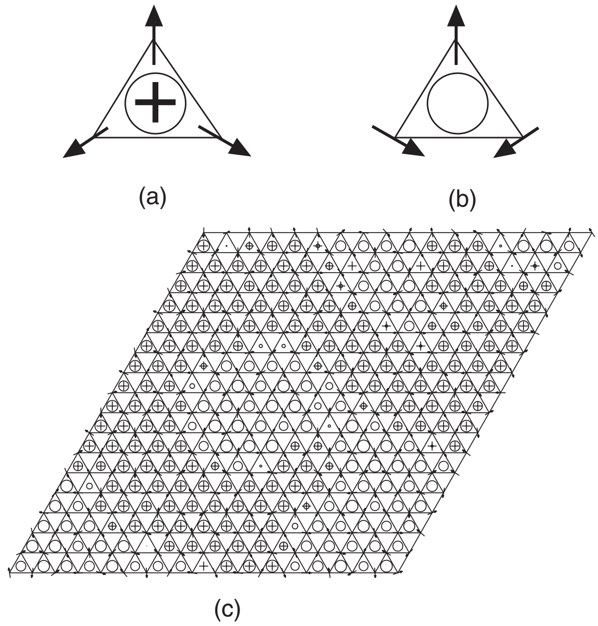

Fig. 8. Ground state spin configurations of antiferromagnetic XY model (a) +120° structure, and (b) −120° structure (c) A domain structure of the chiralities. Open circles denote triangles with '−' chirality, and open circles with '+' denote those of '+' chirality (Eq. [33]). The radius of the circle is proportional to the amplitude of the chirality. This is a snap shot during a relaxation from random configuration to a low temperature ($T = 0.1J$).

In three dimensions, the symmetry P_3 is expected to give a new universality class (chiral universality),[59)] and various new aspects were pointed out. However, it turned out that the system exhibits a first-order phase transition in three dimensions at least for the XY and Heisenberg models.[60)] It is still a challenging topic to look for a phase transition of the chiral universality class in some parameter regions of three dimensional models.

Next, we discuss anisotropic Heisenberg models on the triangular lattice

$$\mathcal{H} = J\sum_{\langle ij \rangle}(S_i^x S_j^x + S_i^y S_j^y + A S_i^z S_j^z) - H \sum_i S_i^z, \quad [34]$$

where A denotes the anisotropy. In the ferromagnetic model, even a weak Ising anisotropy will bring the system to be ordered in the easy axis (along the *z*

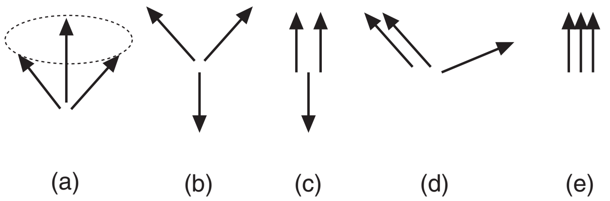

Fig. 9. Spin configurations of solutions of Eq. [35] for the antiferromagnetic anisotropic Heisenberg model in the magnetic field H [34]: (a) Umbrella structure, for $A=1$, $(\theta_1,\phi_1)=(\mathrm{acos}(h/3),0)$, $(\theta_2,\phi_2)=(\mathrm{acos}(h/3),2\pi/3)$, $(\theta_3,\phi_3)=(\mathrm{acos}(h/3),4\pi/3)$. (b) distorted 120° structure ($0\le h\le 1$), for $A=1$, $(\theta_1,\phi_1)=(\pi,0)$ $(\theta_2,\phi_2)=(\mathrm{acos}((h+1)/2),0)$, $(\theta_3,\phi_3)=(\mathrm{acos}((h+1)/2),\pi)$. (c) collinear up-up-down structure. (d) v-shape structure ($1\le h\le 3$), for $A=1$, $(\theta_1,\phi_1)=(\mathrm{acos}(h^2+3)/4h,0)$, $(\theta_2,\phi_2)=(\mathrm{acos}(h^2+3)/4h),0)$, $(\theta_3,\phi_3)=(\mathrm{acos}(h^2-3/2h),\pi)$. (e) collinear ferromagnetic structure. Here $h=H/3J$.

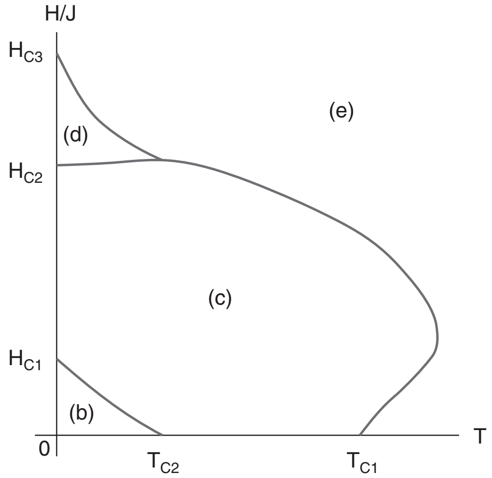

Fig. 10. A schematic phase diagram of the Ising-like Heisenberg model in the coordinate (T,H), where $H_{\rm C1}=3J$, $H_{\rm C2}=\frac{2A-1\sqrt{4A^2+4A-7}}{2}J$, and $H_{\rm C1}=(6A+3)J$.

axis), and the system shows a phase transition of the Ising universality with the order parameter of the Z_2 symmetry. However, because of the frustration, the order parameter has a distorted 120° structure, and nature of the phase transition is different from the simple Ising case.

There are several locally stable structures of the spins on a triangle in the field as depicted in Fig. 9. The stable configurations in the ground state are obtained by

$$\begin{cases} \cos\theta_1(\sin\theta_2+\sin\theta_3) \\ \quad = A\sin\theta_1\left(\cos\theta_2+\cos\theta_3-\dfrac{H}{3J}\right), \\ \cos\theta_2(\sin\theta_3+\sin\theta_1) \\ \quad = A\sin\theta_2\left(\cos\theta_3+\cos\theta_1-\dfrac{H}{3J}\right), \\ \cos\theta_3(\sin\theta_1+\sin\theta_2) \\ \quad = A\sin\theta_3\left(\cos\theta_1+\cos\theta_2-\dfrac{H}{3J}\right). \end{cases} \qquad [35]$$

In the case of zero magnetic field, the system shows successive phase transitions when we change the temperature. At a high temperature $T_{\rm C1}$, the z component is ordered, and then at a low temperature $T_{\rm C2}$ the transverse component is ordered to form the distorted 120° structure[61] as shown in Fig. 10 at $H=0$. The former phase transition belongs to the universality classes of the six-state clock model, and the latter to that of the XY model. Both of them are of the Kosterlitz–Thouless type in two dimensions. In three dimensions, the phase transitions are of normal second order of the three dimensional XY universality class.[62] This mechanism of successive phase transition gives an alternate scenario to that of Mekata model for the successive phase transitions of frustrated Ising-like models in the triangular lattice. Indeed successive phase transitions of this type were also found in experiments (VCl_2,[63] *etc.*).

In the magnetic field, this model gives complicated phases.[64] The phase diagram in the field is depicted in Fig. 10. This type of phase diagram has been also observed in experiments.[65]

In the case of $A=1$, all the configurations which satisfy

$$\cos\theta_1+\cos\theta_1+\cos\theta_1=\frac{H}{3} \qquad [36]$$

are degenerate. At finite temperatures, however, the degeneracy is resolved by the entropy effect.[66] Generally speaking, the collinear structure is entropically more favorable than the coplanar structure, and the coplanar structure is more favorable than the non-collinear structure (umbrella structure: Fig. 9(a)). The entropy effect is estimated by a harmonic analysis of the model:

$$\begin{aligned}\mathcal{H}(\theta_1,\theta_2,\theta_3,\phi_1,\phi_2,\phi_3) &= \mathcal{H}(\theta_1^0,\theta_2^0,\theta_3^0,\phi_1^0,\phi_2^0,\phi_3^0) \\ &\quad+\frac{\partial\mathcal{H}}{\partial\theta_1}\delta\theta_1+\cdots+\frac{\partial\mathcal{H}}{\partial\sin\theta_3^0\phi_3}\sin\theta_3^0\delta\phi_3+{}^t\boldsymbol{x}\hat{A}\boldsymbol{x}, \end{aligned} \qquad [37]$$

where $\theta_i=\theta_i^0+\delta\theta_i$, $\phi_i=\phi_i^0+\delta\phi_i$, ($i=1$, 2, and 3), and

$$\boldsymbol{x}=(\delta\theta_1,\delta\theta_2,\delta\theta_3,\sin\theta_1^0\delta\phi_1,\sin\theta_2^0\delta\phi_2,\sin\theta_3^0\delta\phi_3), \qquad [38]$$

and $\hat{A}$ is a $6N\times 6N$ matrix. The free energy F in the harmonic approximation for a given configuration $(\theta_1^0,\theta_2^0,\theta_3^0,\phi_1^0,\phi_2^0,\phi_3^0)$ is given by

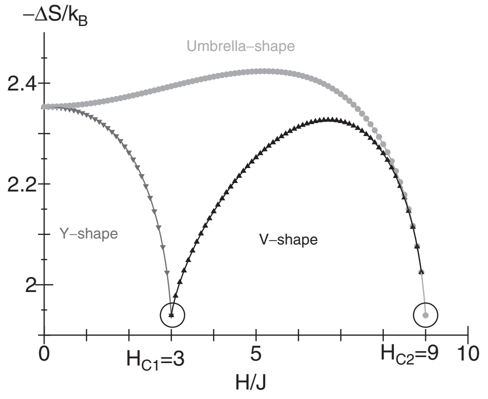

Fig. 11. The ground state entropy [40] for $A = 1$ for the umbrella, Y-shape and V-shape structures. The big circles show entropy for the collinear phases, *i.e.*, the up-up-down and ferromagnetic structures.

$$F/\beta = E_0 + N\ln(\pi\beta) + \frac{1}{2}\left(\ln\prod_i{}'\lambda_i\right), \qquad [39]$$

where λ_i is the i-th (non-zero) eigenvalue of $\hat{A}$. The zero eigenvalues of $\hat{A}$ denote some continuous degeneracies in the ground state, and they are removed in the product $\prod'$. For example, the model has U(1) symmetry, and thus the uniform rotation around the z axis gives the zero eigenvalue, and also the model of $A = 1$ has the degeneracy [36]. Moreover, it has been pointed out that the model with $A > 1$ has a non-trivial degeneracy that there is a ground state for all the values of θ_1^0. The zero modes for θ and ϕ give π and 2π to the product, respectively. We define the ground state entropy for the configuration by

$$\Delta S = -\frac{k_{\mathrm{B}}}{2}\left(\ln\prod_i{}'\lambda_i\right). \qquad [40]$$

We plot $-\Delta S/k_{\mathrm{B}}$ for various configurations as a function of H in Fig. 11.[67]

Because of the entropy effect, at finite temperatures, phases of the Y-shape structure (Fig. 9(b)), and of the up-up-down structure (Fig. 9(c)), and of the V-shape structure (Fig. 9(d)) appear.[66] In systems with a weak XY anisotropy, the umbrella structure is energetically favorable. However, the thermal fluctuation still causes the structures of Figs. 9(b)–(d), and we have a very complicated phase diagram in the coordinate (T,H) as depicted in Fig. 12 where we used a single-ion type anisotropy model instead of the anisotropic coupling model [34]

$$\mathcal{H} = -J\sum_{\langle ij\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + D\sum_i(S_i^z)^2. \qquad [41]$$

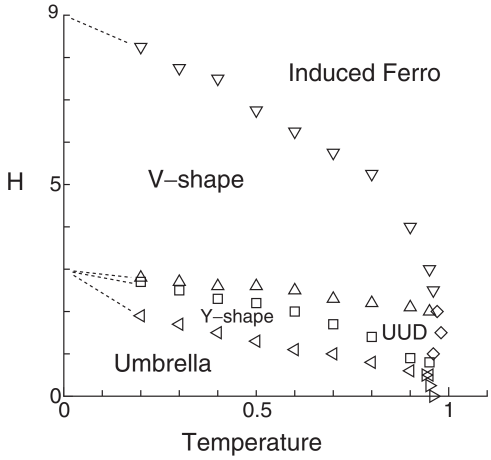

Fig. 12. A schematic phase diagram of a weakly XY-like Heisenberg model in the field. $D = 0.01$.

In the figure, we adopted a very small value of $D = 0.01$. If we use a larger value such as $D = 0.1$, the Y-shape structure disappears, while the V shape structure still remains.[68]

4.3. Antiferromagnet on the Kagome lattice. Finally, let us refer to the case of the Kagome lattice. This model has the so-called corner-sharing structure and effect of the frustration is more significant than that in the edge-sharing lattices such as the triangular lattice. The Ising antiferromagnet in this lattice has no phase transition and the correlation function decays exponentially even at $T = 0$.[69] Because of the high degeneracy, even with the continuous spins, the XY and Heisenberg models do not have a phase transition, either.[70]

However, in the Ising-like Heisenberg model, the spins have to choose the same configuration from those given in Fig. 9 at all the triangles. For example, in a zero or weak field, they have the distorted 120° structure. There are still two possible configurations at zero field as depicted in Figs. 13(a) and (b). The system must choose one of them. This degree of freedom gives a two-fold degeneracy of the ground state, and it causes a phase transition of the Ising universality class at a finite temperature.[71] Indeed the structures of Figs. 13(a) and (b) have nonzero magnetizations $m_0 = \mp(A-1)/(A+1)$, respectively. Therefore, the phase transition causes an appearance of the spontaneous magnetization. Below the critical temperature the two-fold degeneracy is broken, but there still remains macroscopic degeneracy. The number of degeneracy is the same as that of the Ising Kagome antiferromagnet in the magnetic field.[72] An example of the degenerate ground states

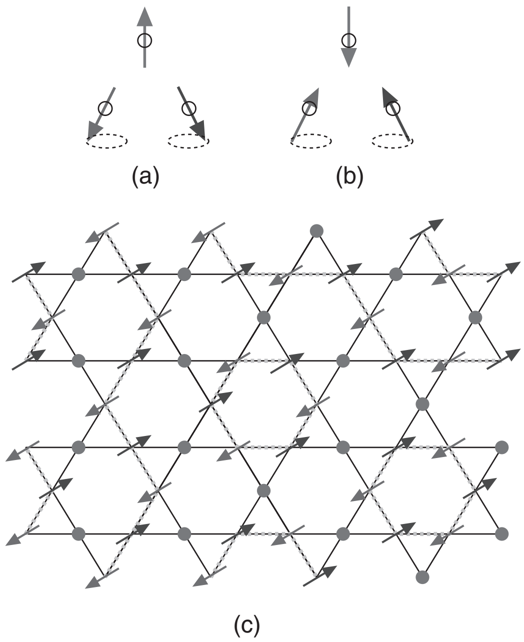

Fig. 13. (a) Distorted chiral structure with a negative magnetization, (b) distorted chiral structure with a positive magnetization, and (c) a snap shot of the ground state spin configuration of antiferromagnetic Ising-like Heisenberg model on the Kagome lattice. The dots denote the spins directing the z direction. The arrows denote the xy components of the spins. The dotted lines denote the weather-vane lines.

is depicted in Fig. 13(c). There is no ferrimagnetic structure, and the spontaneous magnetization appears uniformly over the lattice. Thus, we may regard this phase transition as a ferromagnetic phase transition, and we called this "exotic ferromagnetic transition".

Configurations of the dotted lines connecting the slanting spins characterize the degenerated ground state configurations, and we call the line "weather-vane line". In a weather-vane line, the slanting spins are parallel and each weather-vane loop contributes $k_{\rm B}\ln 2\pi$ to the entropy. Therefore, a configuration with more weather-vane loops is more favorable. The minimum length of the weather-vane loop is six. Thus, we expect that a state with maximum number of the weather-vane loop gives the thermodynamic ground state, in which we have a ferrimagnetic long range order of the z component of spins. Existence of the long range order in the ground state due to the entropy effect has been discussed also for the Heisenberg model.[70)]

Similar induction of long range order in the ground state of fully frustrated model was also pointed out in the Ising antiferromagnet of $S > 1/2$ in the triangular lattice.[73)] These are examples of the "order by disorder"[74)] The relaxation process of this entropy-induced selection of configuration was also investigated.[75)]

In three dimensions, the unit cell of frustration is a tetrahedron, and various interesting phenomena have been found in pyrochlore or spinel lattices, such as the spin ice phenomena.[76)]

5. Effect of randomness

Spatial randomness of interactions also causes various peculiar features in the ordering processes. Generally the randomness smears out the critical phenomena.[77)–80)] In particular, if the specific heat diverges with the exponent $\alpha > 0$ in the original pure model, the exponents is renormalized to $\alpha_X = -\alpha/(1-\alpha)$[77)] in the model with randomness. It was also pointed out that the Ising model on the square lattice with randomly distributed vertical coupling constants keeping the horizontal coupling uniform, the specific heat has the essential singularity, *i.e.*, the all the derivatives of the free energy are continuous.[78)]

Whether a system with randomly distributed interaction can have a phase transition or not is a very interesting problem. Effects of the random field on the phase transition were studied, where the reduction of lower critical dimensions for Ising models was discussed.[81),82)]

To study the phase transition for randomly quenched interaction, we have to average the free energy,

$$F_{\rm quenchrandom} = -k_{\rm B}T\overline{\ln Z_{\rm fixedconfiguration}} \qquad [42]$$

not the partition function

$$F_{\rm annealrandom} = -k_{\rm B}T\ln \overline{Z_{\rm fixedconfiguration}}. \qquad [43]$$

For the former average, we need to calculate the free energy for each fixed random configuration, which is generally difficult, except for some special cases such as the so-called Nishimori line.[83)] To avoid this difficulty, the so-called replica trick has been introduced.[84),85)]

$$F_{\rm quenchrandom} = \lim_{n\to 0}\frac{\overline{Z^n_{\rm fixedconfiguration}}-1}{n}. \qquad [44]$$

In particular, properties of spin glass have been studied in these decades extensively.[82),84),85)] The combination of the randomness and frustration causes a very complicated ordering process. Experimentally, various interesting properties, such as memory effect, rejuvenation phenomena, etc., have been also pointed out.[86)]

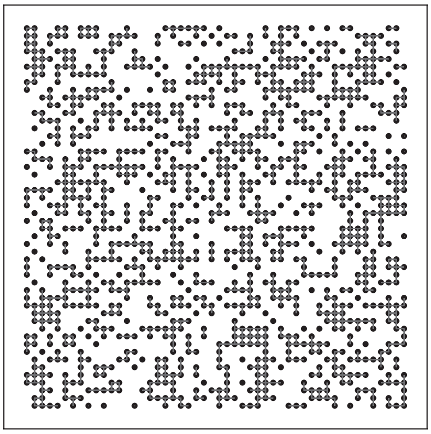

Fig. 14. A diluted configuration with site occupation probability $p = 0.5$ which is below the critical probability of the percolation.

If the system has a well-defined ground state, the system can be regarded as a generalized antiferromagnetic state. But, the spin glass shows several properties inherent to the randomly frustrated system, such as the divergence of the non-linear susceptibility.[87),88)] The analysis of the reprica trick was extended to the case of replica symmetry broken (Parisi) solution which gives a new picture of ordered state of the random system.[89)] As an alternate picture, an idea of extended antiferromagnetic state with weak stiffness has been discussed.[90)] Moreover, a scenario of chirality ordering for the spin glass has been also proposed.[91)] Generally, the frustration among the interaction causes a distribution of strength of the correlation.[92),93)] We may define domains in which spins strongly interact. The structure of the domains plays important role in the dynamics. Large clusters relax slowly, which causes slow a relaxation of the autocorrelation function of spins in average. Although extensive studies for the nature of the spin glass have been done, here we will not discuss on them.

In the diluted ferromagnet, the domains are well defined. When dilution probability p exceeds the critical percolation concentration $p_{\rm C}$,[94)] the lattice is separated into finite domains with probability one. In Fig. 14, we depict a snap shot of a lattice of site dilution with $p = 0.5$ which is above $p_{\rm C}$ for the square lattice with nearest neighbor interaction. We find finite clusters on it. In this case, we expect paramagnetic behavior. However, it has been pointed out that randomly diluted ferromagnets have a non-analytic free energy below the critical temperature of the non-diluted system. Although the probability for large clusters is very small, there is a non-vanishing probability to find arbitrarily large clusters for any p, which causes the non-analytic effect on the free energy. While it has little effect on the equilibrium properties, it has significant effect on the dynamics. For example, the autocorrelation function of spin is affected by the very long relaxation time of the large clusters at the temperature below the critical temperature of the pure system ($p = 0$), and shows a slow relaxation than the simple exponential decay. This type of slow relaxation is called dynamical effect of the Griffiths singularity.[95),96)]

6. Quantum effect

In this section, we discuss effects of the quantum fluctuation on the phase transition. Generally, phase transitions of magnetic systems take place as a macroscopic change of a classical order parameter such as the magnetization, and there the quantum effect is irrelevant at finite temperatures. This type of order parameter is called "Diagonal Long Range Order (DLRO)". On the other hand, for the superfluidity and superconductivity, the quantum effect is essential. The latter type is called "Off-Diagonal Long Range Order (ODLRO)".

Usually, quantum fluctuation tends to destroy the classical ordered state (DLRO). The ground state order–disorder transition in the transverse Ising model[97)]

$$\mathcal{H} = -J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z - \Gamma\sum_i \sigma_i^x \qquad [45]$$

is the most typical example of the quantum phase transition. When Γ exceeds a critical value $\Gamma_{\rm C}$, the ground state becomes disordered, and it is called "quantum disordered state". The effect of quantum fluctuation is taken into account by the Suzuki–Trotter expression of the partition function which is a path-integral representation of the canonical weight.[98)] This formula was proposed for the quantum Monte Carlo method.[99)] The ground state property of a d-dimensional quantum system is expressed by the partition function

$$Z = \lim_{\beta\to\infty} {\rm Tr} e^{-\beta\mathcal{H}}. \qquad [46]$$

This can be expressed by a path-integral or Suzuki–Trotter formula in a $(d+1)$-dimensional space,

$$\begin{aligned} Z = {\rm Tr}e^{-\beta\mathcal{H}} &\simeq {\rm Tr}_{d+1}\left(e^{\frac{\beta}{n}J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z}e^{\frac{\beta}{n}\Gamma\sum_i\sigma_i^x}\right)^n \\ &= \sum_{\{\sigma_i^m=\pm1\}}\langle\{\sigma_i^1\}|e^{\frac{\beta}{n}J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z}e^{\frac{\beta}{n}\Gamma\sum_i\sigma_i^x}|\{\sigma_i^2\}\rangle\langle\{\sigma_i^2\}|\cdots \\ &\quad\cdots e^{\frac{\beta}{n}J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z}e^{\frac{\beta}{n}\Gamma\sum_i\sigma_i^x}|\{\sigma_i^n\}\rangle, \end{aligned} \qquad [47]$$

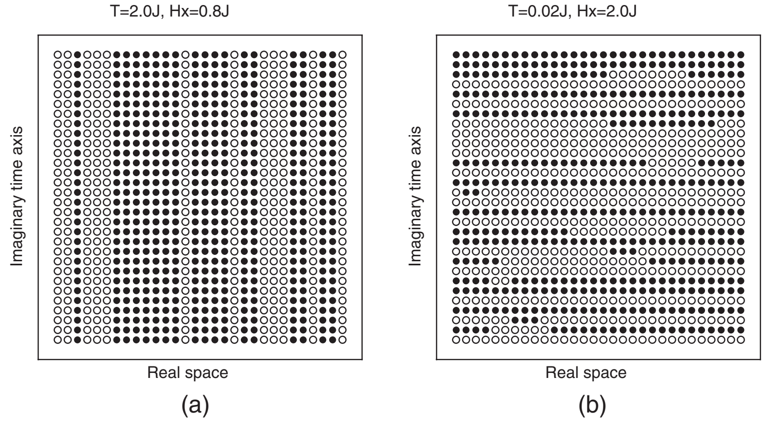

Fig. 15. Configurations of the quantum Monte Carlo simulation for the transverse Ising model. (a) Thermal fluctuation, and (b) Quantum fluctuation.

where Tr_{d+1} denotes the sum all over the spin states including those inserted as a complete set of states $\{\sum_i |\{\sigma_i^m\}\rangle\langle\{\sigma_i^m\}| \quad (m = 1, \ldots n)\}$, and we call n "Suzuki–Trotter number". If we regard the direction of n as a new spatial direction, the model [45] is transformed into a $(d+1)$-dimensional Ising model with the coupling constants:

$$\beta\mathcal{H} = -\frac{\beta J}{n}\sum_{\langle ij\rangle}\sigma_{i,k}\sigma_{j,k} - K_2\sum_k \sigma_{i,k}\sigma_{i,k+1} \quad [48]$$

where

$$K_2 = -\frac{1}{2}\ln\left(\tanh\frac{\beta\Gamma}{n}\right). \quad [49]$$

Thus, the critical property of a d-dimensional quantum system is related to that of a $(d+1)$-dimensional classical system at finite temperature.[98] In Fig. 15, we depict configurations in the $(d+1)$-dimensional system for an one dimensional transverse Ising model. The left and right panels show typical configurations for the thermal fluctuation and the quantum fluctuation, respectively. The thermal fluctuation gives disorder in the real space while the quantum fluctuation gives disorder in the imaginary time space.

In the random systems such as the diluted system, the randomness exists in the real space but not along the imaginary time axis. Thus, the clusters in the $(d+1)$-dimensional spaces has a rod type shape and they are not isotropic random clusters. Because of this fact, it has been known that the Griffiths singularity in random systems has serious effects on static properties in the ground state. Indeed, the magnetic susceptibility diverges in the diluted transverse Ising model in some regions of quantum disorder phase, which is called "quantum Griffiths singularity".[100),101)]

In the above systems, the quantum fluctuation reduces the order, but in some cases the quantum fluctuation induces an order. In a model of $S=1$ with a tunneling effect between the state of $S=\pm 1$

$$\mathcal{H} = -J\sum_{\langle ij\rangle} S_i^z S_j^z + D\sum_i (S_i^z)^2 + \Gamma\sum_i ((S_i^+)^2 + (S_i^-)^2), \quad [50]$$

where $S_i^{\pm}$ denote the operations to change the magnetization by one, we find the spontaneous magnetization appears in a region where no magnetization exists in the classical case ($\Gamma = 0$).[102)]

Recently, various peculiar ground states of quantum spin systems have been found in low dimensions such as the Haldane state.[103)] In those systems, spatial configuration of the singlet pairs plays an important role.[104)] A transition between different types of ground states gives the quantum phase transition.[105),106)] It is also known that, in quantum systems, inhomogeneity of the lattice shape affects on the spin configuration in the ground state significantly, *e.g.* the edge effect.[107)] Therefore, randomness of lattice shapes such as the dilution causes various non intuitive properties.[108)–110)] The ground state phase transition has been studied extensively,[111)] but we will not go in details.

It is also an interesting problem to study the similarity of the quantum and thermal fluctuations. As we see in the section 4.2, the fluctuation has important effects on the magnetization process of the antiferromagnetic XXZ model in the triangular lattice. Classically, the umbrella structure is the lowest energy configuration for easy-plane (XY like) systems, and the magnetization process $M(H)$ is linear until its saturated value at $T = 0$. However, existence of a 1/3-plataux was observed in the experiment on an easy-plane (XY-like) magnet $CsCuCl_3$.[112] This observation implies an existence of some easy-axis (Ising like) anisotropy. Because the temperature is low, we do not expect the thermal fluctuation, and the thermal-fluctuation induced metamagnetic structure which was explained in the section 4.2 is not applicable. However, Nikuni and Shiba pointed out that the quantum fluctuation takes the role to induce the 1/3 plateau by calculating the energy gain of the zero-temperature fluctuation in a spin wave theory.[113] This problem was examined by direct numerical studies of the ground state wave-function (PWFRG[114] and DMRG[115]).[116] There, a metamagnetic process similar to the classical one was revealed, and a phase diagram in the coordinate (A,H) was obtained[116),117] where A is the ratio of the exchange energy of the xy component and that of the z-component (see Eq. [34]).

As to the ODLRO, their cooperative properties have been studied as the ordering of phase of the wave function, because the ODLRO is regarded as the problem of phase coherence of the macroscopic wave function. Thus, phase transitions of the superconductivity were often analyzed by the XY model.[118] Recently, the nature of ODLRO has been studied more directly by using the quantum Monte Carlo method, and a transition between the Mott state and superfluidity,[119] and also coexistence of the solid state and superfluidity (supersolid)[120] have been clarified.

7. Phase transitions in the spin-crossover type models

7.1. Spin-crossover transition. So far, we studied orderings of the direction of spins on lattices. However, the spin itself on each lattice site may also change as a function of parameters. One of typical examples is the spin-crossover transition,[121] where the spin of an atom changes between the high spin (HS) state and the low spin (LS) state depending on the ligand field as depicted in Fig. 16. When the ligand field is weak, the spins of electrons align due to the Hund law and the total spin of the atom S is large. On the other hand when the ligand field is strong the electrons occupy the low energy states and S is small.

Here, we consider cases in which the energy of the LS state is low by $2D$, while the degeneracy of the

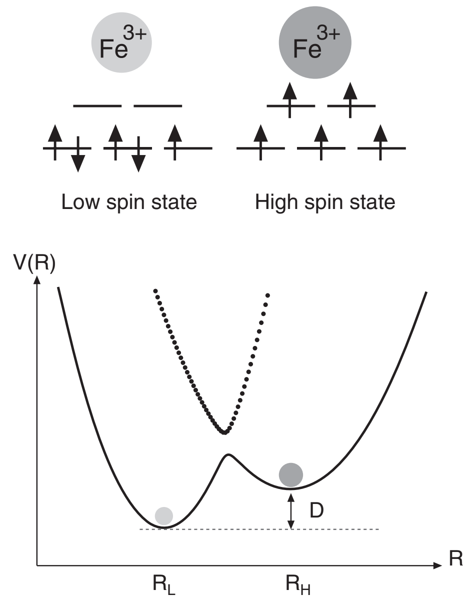

Fig. 16. The High spin (HS) and low spin (LS) states, and a schematic energy structure as a function of the radius R of the molecule.

HS state (g_+) is larger than that of the LS state (g_-). We denote the HS and LS states by $\sigma = 1$ and -1, respectively. The partition function of a molecule is given by

$$Z_1 = \sum_{\sigma=\pm 1} g_\sigma e^{-\beta D \sigma} = g_+ e^{-\beta D} + g_- e^{\beta D}. \qquad [51]$$

This can be rewritten as

$$Z_1 = \sqrt{\frac{g_+}{g_-}} \sum_{\sigma=\pm 1} e^{-\beta(D - \frac{1}{2}k_B T \ln g)\sigma}, \qquad [52]$$

where $g = g_+/g_-$. This partition function can be regarded as that of a model with a temperature dependent field

$$H = -D + \frac{1}{2} k_B T \ln g. \qquad [53]$$

The importance of the interaction among molecules for the SC transition was pointed out in the observation of the specific heat,[122] and actually the transition often takes place as a discontinuous transition. The variety of transitions, *e.g.*, the smooth change and discontinuous change can be attributed to relations among parameters. If we include an interactions among spin states $\mathcal{H}_{\rm int}$, the Hamiltonian of the system is given by

$$\mathcal{H} = \mathcal{H}_{\rm int}(\{\sigma_i\}) + \sum_i \left(D - \frac{1}{2} k_B T \ln(g) \right) \sigma_i. \qquad [54]$$

The simplest choice of the interaction is the ferromagnetic Ising model[123] $\mathcal{H}_{\rm int} = -J\sum_{\langle ij\rangle}\sigma_i\sigma_j$. For this model, we can obtain the phase diagram of the SC system by making use of that of the ferromagnetic systems with the temperature dependent magnetic field [53].

The free energy of the ferromagnetic Ising model

$$\mathcal{H}_{\rm int} = -J\sum_{\langle ij\rangle}\sigma_i\sigma_j - H\sum_i \sigma_i. \qquad [55]$$

in the mean-field theory is given by

$$F(m,T,H) = \frac{1}{2}zJm^2 - k_{\rm B}T\ln(2\cosh(\beta zJm + \beta H)), \qquad [56]$$

where $m = \langle\sigma_i\rangle$. The spinodal point is given by

$$\frac{\partial F(m,T,H)}{\partial m} = 0 \quad \text{and} \quad \frac{\partial^2 F(m,T,H)}{\partial m^2} = 0, \qquad [57]$$

and we have the spinodal field as

$$H_{\rm SP} = \pm zJ\sqrt{1-\frac{1}{\beta zJ}} \mp \frac{1}{2\beta}\ln\left(\frac{1+\sqrt{1-\frac{1}{\beta zJ}}}{1-\sqrt{1-\frac{1}{\beta zJ}}}\right). \qquad [58]$$

Let the critical temperature of the system be $T_{\rm C} = zJ$, and then the phase diagram in the coordinate (T,H) is schematically given by Fig. 17 with the lines indicating spinodal field. Within the spinodal fields [58], there is a metastable state with the magnetization opposite to the field H. In the present case, the spinodal field at $T = 0$ is zJ.

It has turned out that there are various ordering patterns of the HS fraction[124),125)]

$$f_{\rm HS}(T) = (m(T,H)+1)/2. \qquad [59]$$

In this phase diagram, temperature dependence of the model [54] is given by the straight lines (I–V) with the slope given by the relation [53]. For large D, the line of [53] crosses the line of $H = 0$ at a higher temperature than $T_{\rm C}$ (along the line I). In this case the change of $m(T,H)$ along this line is smooth. That is, the change of $f_{\rm HS}(T)$ for large D is smooth.

On the other hand, for small D, the line crosses the line of $H = 0$ at a temperature below $T_{\rm C}$ (the lines III–V). There, $f_{\rm HS}(T)$ changes discontinuously at $H = 0$, and $f_{\rm HS}(T)$ shows a thermal hysteresis between the lines of the spinodal fields. The relation [53] indicates that the thermal hysteresis takes place for

$$D < D_{\rm C1} = \frac{1}{2}k_{\rm B}T_{\rm C}\ln g. \qquad [60]$$

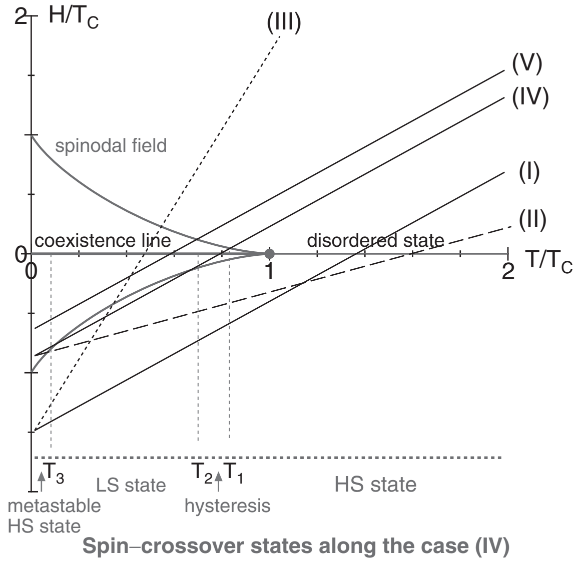

Fig. 17. Phase diagram of the ferromagnetic system in the coordinate (T,H) with the spinodal field [58]. The temperature dependences of the SC models [54] for various cases (I)–(V) are plotted (see the text). Phases along the line (IV) are indicated blow the figure, where T_1 and T_2 denote the temperatures of the upper limit and lower limit of the thermal hysteresis, respectively, and T_3 denotes the temperature below which the metastable HS state appears.

If $D > zJ$ and $D < D_{\rm C1}$, the line [53] crosses the spinodal lines as plotted by the dotted line (III).

If $D < zJ$ and $D < D_{\rm C1}$, beside the thermal hysteresis between T_1 and T_2, the line [53] again crosses the spinodal line [58] at a low temperature T_3 (the line IV), because the slop of the spinodal line [58] is infinite at $T = 0$. Below this crossing point ($T < T_3$), the HS state is metastable. This low temperature metastable HS state may explain the long life time of the LIESST (light-induced excited spin state trapping) state.[124)] Existence of such metastable state has been confirmed experimentally in a charge transfer material.[126)]

If $D < zJ$ and $D > D_{\rm C1}$, although there is no thermal hysteresis, the low temperature metastable HS state exists (along the line II). The critical value of D for the low temperature metastable state is

$$D_{\rm C2} = zJ. \qquad [61]$$

If the line [53] stays inside the metastable region (the line V), the HS state remains metastable at all the temperatures. The critical value of D for this case is given by

$$D_{\rm C3} = zJ\tanh\left(\frac{1}{2}\ln g\right). \qquad [62]$$

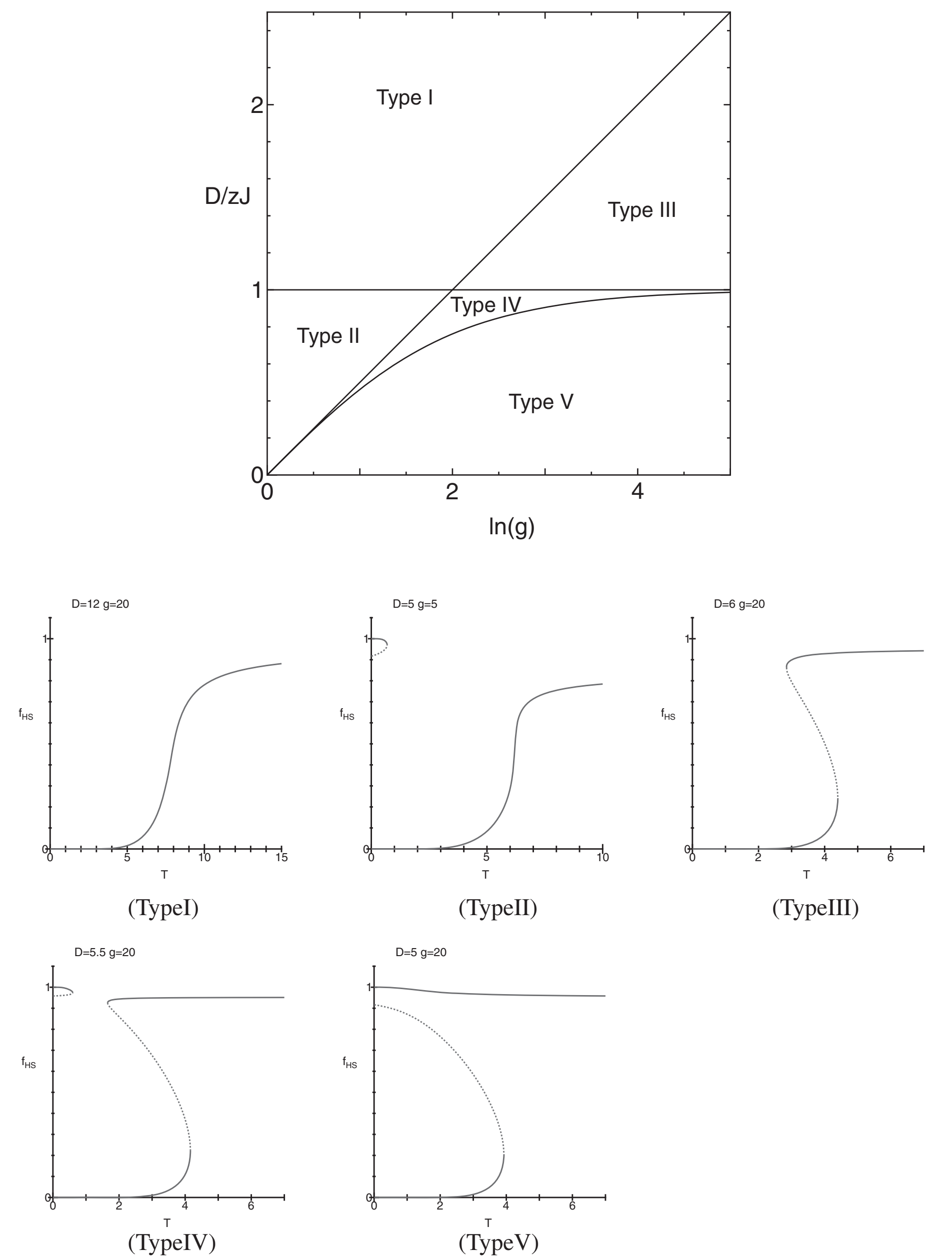

Fig. 18. Phase diagram of the ordering patterns $f_{\rm HS}(T)$ in the coordinate $(D/zJ, \ln g)$. Ordering patterns for various values of D: (Type I) smooth change for $D > D_{\rm C}$, (Type II) smooth change and a metastable branch at low temperatures for $D > D_{\rm C1}$ and $D < D_{\rm C2}$, (Type III) discontinuous change for $D_{\rm C1} > D > D_{\rm C2}$, (Type IV) discontinuous change and a metastable branch at low temperatures for $D_{\rm C1} > D$ and $D < D_{\rm C2}$, and (Type V) HS is always stable or metastable for $D < D_{\rm C3}$.

In Fig. 18, types of ordering processes corresponding the lines (I–V), and the phase diagram in the coordinate $(\ln g, D/zJ)$ are depicted.[124),125)] We find that the type IV appears for all the sequences in the parameter space. We call this feature "generic sequence of the temperature dependence of ordering $f_{\rm HS}(T)$". We found this type of sequence also appears in the change of other parameters such as the pressure and stiffness of the elastic constant.

This type of description can be applied to many systems where the energy and the degeneracy of local degree of freedom compete, *e.g.* the charge transfer (CT) materials,[127)] and we expect the present generic sequence will be found in all the such materials.

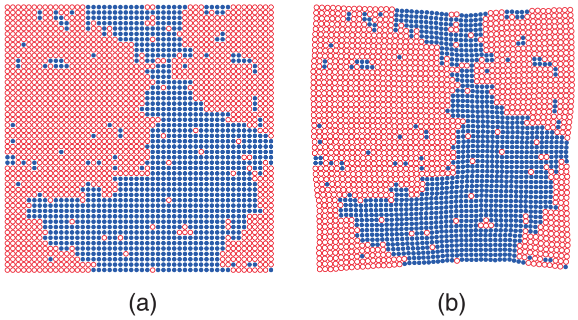

Fig. 19. Deformation due to the size difference of molecules (a) A typical configuration near the critical point in the ferromagnetic Ising model $R_H = R_L$. (b) The lattice deformation in the elastic model ($R_H > R_L$) with the same spin configuration.

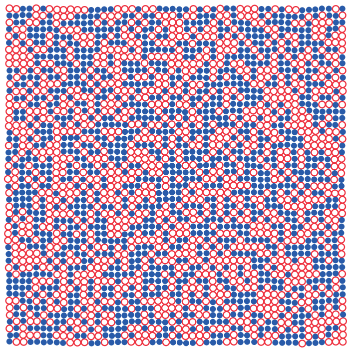

Fig. 20. A typical configuration near the critical point. $T = 0.2 \simeq T_C$, $D = 0.6$, $k = 40$, $g = 20$, $R_{HS} = 1.1R_{LS}$.

7.2. Mean-field phase transition due to the elastic interaction. So far, we studied the spin-crossover phenomena in the analogy of ferromagnetic model on a lattice. However, a significant feature of the SC system is the volume change between the HS and LS states. As we depicted in Fig. 19, the volume change causes a distortion of the lattice. Therefore, in this case, an elastic energy due to the deformation of the lattice must be taken into account.[128] The elastic interaction favors non-deformed structure. In the previous subsection, we introduced a nearest neighbor ferromagnetic interaction Eq. [55] between the molecules to induce the cooperative behavior. However, it is expected that even in systems without the short range interaction, the elastic interaction can induce a cooperative effect. Thus we studied a model with only an elastic interaction between the molecules, *e.g.*,

$$V = \frac{k_1}{2}\sum_{\langle i,j\rangle}[r_{ij} - (R_i + R_j)]^2 + \frac{k_2}{2}\sum_{\langle\langle i,j\rangle\rangle}[r_{ij} - \sqrt{2}(R_i + R_j)]^2, \qquad [63]$$

where r_{ij} is the distance between the ith and jth sites, and R_i and R_j are the molecular radii, R_{HS} and R_{LS} for HS and LS, respectively. In the simulation we adopted the ratio $R_{HS}/R_{LS} = 1.1$ and the elastic constants $k_1/k_2 = 10$ with $k_1 = 50$.

This scenario of phase transition due to the elastic interaction has been confirmed in both studies of molecular dynamics method[129] and Monte Carlo method.[130] Because the elastic interaction causes an effective long range interaction among the spin states, characteristics of the critical phenomena of the model change from that of the short range ferromagnetic Ising model. It turned out that the model exhibits critical behavior of the universality class of the mean-field model.[131]

Realization of the mean-field universality class has been discussed for the models in higher ($D > 4$) dimensions or models with a long range exchange model.[132] In contrast, the SC model consists of a normal short range elastic model in three dimensional materials, and thus we expect this mean-field universality class will be found in a wide range of real materials.

As a consequence of the mean-field criticality, we found that the model does not show configurations with large domains even near the critical point[131] as depicted in Fig. 20. That is, the model does not show divergence of the correlation length of the order parameter. The model exhibits a symmetry breaking at the critical point keeping the uniformity of the configuration. This fact indicates that the model will not show the critical opalescence which is a symbol of the phase transition of the short range models.

At the end points of the hysteresis loop, the spinodal phenomenon takes place.[133] We expect that at the points the nucleation processes take place and inhomogeneous configurations appear, which is an essential feature of the relaxation from the metastable state in short-range interaction models. However, in the present model, the configuration is kept uniform. Moreover, the spinodal phenomenon occurs as a true critical change in the present elastic model, while it is a crossover in the short range model due to the local nucleation processes. A similar threshold singular behavior occurs in the photo-excitation process from the LS states to a photo-induced HS states (LIESST), and the critical properties of the switching have been studied.[134]

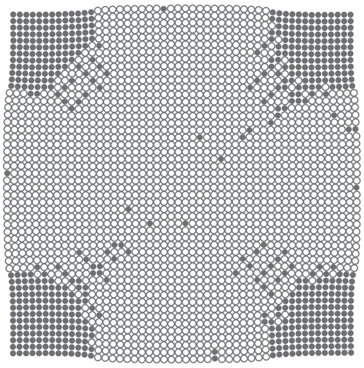

Fig. 21. A configuration in the relaxation from the complete HS state to a LS state in an open boundary system. $T = 0.01$, $D = 0.5$, $g = 20$, $k = 40$, $R_{\mathrm{HS}} = 1.1R_{\mathrm{LS}}$.

Although the elastic interaction causes an effective long range interaction, it is not the infinite range model (Husimi–Temperley model), and thus the boundary condition has a serious effects. If we study the system in a free boundary condition,[135] the relaxation begins from the corners and macroscopic domain structure appears as depicted in Fig. 21. The effect of boundary condition in the effective long range interaction will be an interesting problem in the future.

8. Summary and discussion

We have overviewed natures of phase transitions of systems with large fluctuation. We saw various types of ordering processes reflecting structure of the order parameters. The fluctuations played an important role not only to destroy the ordered state but also to choose an ordered state and also to create a new type of ordered state both in classical and quantum systems. We have found some examples of peculiar orders, and it would be also an interesting problem to study the ordered states inherent to the high dimensions where more than one order can percolate and be in ordered state. Moreover, in the last section we studied the off-lattice model. The structural phase transition is a challenging topic in this direction.

In this paper we mainly studied static properties of phase transitions. Phase transitions also show various types of relaxation processes. The dynamics is also an important characteristic of the phase transition, which will be reviewed elsewhere.

We hope the resent overview would help the further studies of phase transition.

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Profile

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