Review

Phase transition in spin systems with various types of fluctuations

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(Communicated by Jun Kondo, M.J.A.)

Abstract: Various types of 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., \( e^{-E_i/k_B T} \), where \( E_i \) is an energy of a state \( i \), \( T \) is the temperature, and \( k_B \) 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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doi: 10.2183/pjab.86.643
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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 parameter and the thermal fluctuation (the entropy) which causes random configurations. Beside this fundamental mechanisms 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

$$\mathcal{H} = -\sum_{\langle ij \rangle} J_{ij} X_i X_j - H \sum_{i} X_i, \quad [1]$$

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 $\sum'_{i}$ denotes the sum for the order parameter $M = \sum_{i} X_i$. 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

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

and its fluctuation

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

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

$$\chi = \frac{\langle M^2 \rangle - \langle M \rangle^2}{k_B T} \propto |T - T_C|^{-\gamma}, \quad [4]$$

and the spontaneous order parameter $m$ appears as

$$m \propto |T - T_C|^{-\beta}, \quad [5]$$

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

$$C = \frac{\langle \mathcal{H}^2 \rangle - \langle \mathcal{H} \rangle^2}{k_B T^2} \propto |T - T_C|^{-\alpha}. \quad [6]$$

Here, the exponents $\alpha$, $\beta$ and $\gamma$ 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:

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

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 Onsager3) by the thin curves. There, we see the divergence at the critical point $k_B T_C/J = (2/\ln(1 + \sqrt{2}) \simeq 2.269\ldots$.

In the figure, we also show the temperature dependence of the spontaneous magnetization obtained by C. N. Yang11)

![Fig. 1. Temperature dependences of the specific heat (thin line) and of the spontaneous magnetization of the ferromagnetic two-dimensional Ising model [7] with $H = 0$.](image-url)
This is an example of the scaling relations among the critical exponents. The divergence of the correlation length is expressed as

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

The free energy of the Ising model has been obtained by various methods, and a new field of the so-called “exactly solvable model” has been developed. 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 \(\xi\)

$$\langle S_i S_j \rangle \sim \frac{1}{\nu^{2+\eta/\gamma}} e^{-r_{ij}/\xi}$$ \[9\]

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

$$\xi \sim |T - T_c|^{-\nu}$$ \[10\]

with \(\nu = 1\). It is known that the exponent \(\gamma\) is 1/4 for the present model. At the critical point, fluctuation of the order parameter becomes large which results in the divergence of \(\chi\). 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 \(\chi\) corresponds to the correlation length \(\xi\). The exponent \(\nu\) is related to \(\gamma\) and \(\eta\) as

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

This is an example of the scaling relations among the critical exponents.

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 \(S_i = (\cos \theta_i, \sin \theta_i)\):

$$H_{XY} = -J \sum_{\langle ij \rangle} \cos(\theta_i - \theta_j),$$ \[12\]

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. 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.”

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

$$\langle S_i \cdot S_j \rangle \sim r_{ij}^{-\eta},$$ \[13\]

with a temperature dependent exponent \(\eta = k_B T/4J\) 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

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

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 from a random configuration to an aligned one at a low temperature.

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

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

then the condition \(\sum_{m, \text{around } k} s_{kn} = 0\) 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 \(\{h_i\}\):

\[
Z = \sum_{h_i = -\infty}^{\infty} \prod_{\langle ij \rangle} I_{h_i-h_j}(K)
\]

and integrating over $\theta_i$, the partition function is given by

\[
Z = \prod_{\langle ij \rangle} \sum_{h_j = -\infty}^{\infty} \left( \prod_h \delta_{\sum_{m, \text{around } k} s_{km}, h} \right) I_{s_{ij}}(K),
\]

where

\[
I_{s_{ij}}(K) = \int_{0}^{2\pi} \frac{d\phi}{2\pi} e^{-i\phi + K\cos(\phi)} \simeq \frac{1}{\sqrt{2\pi K}} e^{-s^2/2K}
\]

for large $K$. If we introduce the dual variables

\[
s_{kn} = h_k - h_m,
\]

then the condition \(\sum_{m, \text{around } k} s_{kn} = 0\) 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 \(\{h_i\}\):

\[
Z = \sum_{h_i = -\infty}^{\infty} \prod_{\langle ij \rangle} I_{h_i-h_j}(K)
\]

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

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

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

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 \(\mathcal{H}_V\)\(^{21)}\):

\[
e^{-\beta \mathcal{H}_V} = \sum_{m = -\infty}^{\infty} e^{\frac{1}{4} \sum_{i} K(\theta_i - \theta_{i+2\pi})^2}, \quad [21]
\]

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

\[ Z = \sum_{h_i} \int_{-\infty}^{\infty} d\phi_i e^{-\frac{1}{2} \sum_i \phi_i^2} \prod_{i<j} \phi_i \phi_j \phi_i \phi_j. \]  

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

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

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 \( \{h_i\} \).  

### 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 \( T = 0 \):

\[ S(T = 0) = k_B \ln W \approx 0.338 N k_B, \]  

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 function of this type of models decays in a power law\(^{32,33} \)

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

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, 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 of the strength (say $J_2$) of the nnn interaction. 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 $\alpha$ (Eq. [6]) of this system changes continuously with $J_2$ reflecting the peculiar order of the eight vertex model. If the frustration is strong, i.e., for small $J_2$, the critical exponent $\alpha$ 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). In two dimensions, the six-state clock model,

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

[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. 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.

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

$$\mathcal{H} = \varepsilon_1 \delta_{n,n_1 \pm 1} + \varepsilon_2 \delta_{n,n_1 \pm 2} + \varepsilon_3 \delta_{n,n_1 \pm 3}.$$

[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.

It was discovered that, in the case $\varepsilon_1 \ll \varepsilon_2 \approx \varepsilon_3$, the model has a new type of intermediate-temperature phase in which two neighboring states mix microscopically. 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 [41] 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.
contribution to the e
decoration spins is weak (\(Z\)). The e
rich structure of the ordered states, so-called
staircaseFig. 6. Domain structure of mixed states of the generalized six-
ferromagnetic Ising model with a nearest neighbor interaction in one direction (ANNNI model).45) These
observations indicate that various unknown rich
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, \(\sigma_1\) and \(\sigma_2\), which we call the
system spin, are connected by a direct bond (\(J_0\)) and
two paths with the spins \(s_i\) \((i = 1, 2)\) which we
call the decoration spin. The Hamiltonian of the
decorated bond is given by

\[
\mathcal{H}_{\text{decorated bond}} = J_0 \sigma_1 \sigma_2 - J_1 (\sigma_1 + \sigma_2) (s_1 + s_2). \tag{29}
\]

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

\[
Z_0 e^{-K(T)_{\text{eff}} \sigma \cdot \sigma} = \text{Tr}_{\sigma_1, \sigma_2} e^{-\mathcal{H}_{\text{decorated bond}}/k_B T}, \tag{30}
\]

where \(Z_0\) is a factor independent of \(\sigma\). 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.,
\(K(T) \approx (2J_1 - J_0)/k_B T\).

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
vanishes at the temperature \(T_0\) at which
\(\tanh(J_0/2k_BT) = \tanh^2(J_1/k_BT)\).

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.\textsuperscript{(51),(52)} The structure of the spiral state is obtained by the Fourier transformation of the interaction

\[\mathcal{H} = \sum_{(ij)} J_{ij} S_i \cdot S_j = \sum_k J(k) S_k \cdot S_{-k}. \quad [31]\]

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

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

and it has minima at two points \((k_x, k_y) = \pm (\frac{2\pi}{3}, \sqrt{3})\) 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 \(\pm 120^\circ\) structure, respectively.\textsuperscript{(51),(52)} Thus, the symmetry of the ground state is given by \(\mathbb{Z}_2 \times S_1\), i.e., the two-fold degeneracy and the trivial degeneracy of the rotation around the \(z\)-axis. Amplitude of the local \(\pm 120^\circ\) structure is described by the quantity "chirality"\textsuperscript{(54)}

\[ \kappa = \frac{2}{3\sqrt{3}} (S_i \times S_j + S_j \times S_k + S_k \times S_l). \quad [33]\]

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.\textsuperscript{(51)} 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.\textsuperscript{(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 \(P_3\).\textsuperscript{(56)} This structure has a new type of vortex \((\mathbb{Z}_2\) vortex\) as the point defect (the homotopy group classification \(\pi_1(P_3) = \mathbb{Z}_2\)). 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.\textsuperscript{(58)}

In three dimensions, the symmetry \(P_3\) is expected to give a new universality class (chiral universality),\textsuperscript{(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.\textsuperscript{(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

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

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 \(z\)
the temperature. At a high temperature, the order parameter has a distorted 120° structure, and Ising universality with the order parameter of the $Z_2$ axis), and the system shows a phase transition of the latter to that of the XY model. Both of them are of normal three dimensions, the phase transitions are of normal harmonic approximation for a given configuration $(\theta_1^0, \theta_2^0, \phi_1^0, \phi_2^0, \phi_3^0)$ is given by

$$\mathcal{H}(\theta_1, \theta_2, \theta_3, \phi_1, \phi_2, \phi_3) = \mathcal{H}(\theta_1^0, \theta_2^0, \theta_3^0, \phi_1^0, \phi_2^0, \phi_3^0) + \frac{\partial \mathcal{H}}{\partial \sin \theta_3^0} \sin \theta_3^0 \delta \phi_3 + \mathbf{\hat{A}} \mathbf{\hat{x}},$$

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

$$\mathbf{\hat{A}} = (\delta \theta_1, \delta \theta_2, \delta \theta_3, \sin \theta_1^0 \delta \phi_1, \sin \theta_2^0 \delta \phi_2, \sin \theta_3^0 \delta \phi_3),$$

and $\mathbf{x}$ is a $6 \times 6$ 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

$$F = \frac{1}{2} \mathbf{x} \mathbf{\hat{A}} \mathbf{x}.$$

Fig. 9. Spin configurations of solutions of Eq. [35] for the antiferromagnetic anisotropic Heisenberg model in the magnetic field $H$. (a) Umbrella structure, for $A = 1$, ($\theta_1, \phi_1) = (\cos(h/3), 0), (\theta_2, \phi_2) = (\cos(2h/3), 2\pi/3), (\theta_3, \phi_3) = (\cos(h/3), 4\pi/3)$, (b) distorted 120° structure ($0 \leq h \leq 1$), for $A = 1$, ($\theta_1, \phi_1) = (\pi, 0) (\theta_2, \phi_2) = (\cos((h + 1)/2), 0), (\theta_3, \phi_3) = (\cos((h + 1)/2), \pi)$, (c) collinear up-down structure, (d) v-shape structure ($1 \leq h \leq 3$), for $A = 1$, ($\theta_1, \phi_1) = (\cos(h^2 + 3)/4h, 0), (\theta_2, \phi_2) = (\cos(h^2 + 3)/4h, 0), (\theta_3, \phi_3) = (\cos(h^2 - 3/2h), \pi)$, (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 $H_{C1} = 3J$, $H_{C2} = \frac{3\sqrt{3} + 3\sqrt{3}}{4} J$, and $H_{C3} = (6A + 3)J$.
where $\lambda_i$ 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 $Q_0$. 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. 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 $\phi$. The zero modes for $\theta$ and $\phi$ give $\pi$ and $2\pi$ to the product, respectively. We define the ground state entropy for the configuration by

$$\Delta S = -\frac{k_B}{2} \left( \ln \prod_i \lambda_i \right).$$

We plot $-\Delta S/k_B$ 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.\(^{34}\)

$$H = -J \sum_{\langle i,j \rangle} S_i \cdot S_j + D \sum_i (S_i^z)^2.$$ \(^{41}\)

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 $m_0 = \pi(A - 1)/(A + 1)$, 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
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 $k_B \ln 2\pi$ 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.\(^\text{70}\)

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.\(^\text{73}\) These are examples of the "order by disorder"\(^\text{24}\). The relaxation process of this entropy-induced selection of configuration was also investigated.\(^\text{75}\)

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.\(^\text{76}\)

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.\(^\text{77-80}\) In particular, if the specific heat diverges with the exponent $\alpha > 0$ in the original pure model, the exponents is renormalized to $\alpha_X = -\alpha/(1 - \alpha)$ 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.\(^\text{78}\)

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.\(^\text{81,82}\)

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

$$F_{\text{quench random}} = -k_B T \ln Z_{\text{fixed configuration}}$$

not the partition function

$$F_{\text{anneal random}} = -k_B T \ln Z_{\text{fixed configuration}}$$

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.\(^\text{83}\) To avoid this difficulty, the so-called replica trick has been introduced.\(^\text{84,85}\)

$$F_{\text{quench random}} = \lim_{n \to 0} \frac{Z_n^{\text{fixed configuration}} - 1}{n}$$

In particular, properties of spin glass have been studied in these decades extensively.\(^\text{82,84,85}\) 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.\(^\text{86}\)
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 properties. The analysis of the replica trick was extended to the case of replica symmetry broken (Parisi) solution which gives a new picture of ordered state of the random system. As an alternate picture, an idea of extended antiferromagnetic state of the random system has been proposed. Moreover, a scenario of chirality ordering for the spin glass has been discussed. Generally, the frustration among the interaction causes a distribution of strength of the correlation. 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 \( p_c \), the lattice is separated into finite domains with probability one. In Fig. 14, we depict a snapshot of a lattice of site occupation probability \( p = 0.5 \) which is below the critical probability of the percolation.

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

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

is the most typical example of the quantum phase transition. When \( \Gamma \) exceeds a critical value \( \Gamma_C \), 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. This formula was proposed for the quantum Monte Carlo method. The ground state property of a \( d \)-dimensional quantum system is expressed by the partition function

\[
Z = \lim_{\beta \to \infty} \text{Tr} e^{-\beta \mathcal{H}}.
\]

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

\[
Z = \text{Tr} e^{-\beta \mathcal{H}} \approx \text{Tr}_{d+1} \left( e^{-J \sum_{\langle ij \rangle} \sigma_i \sigma_j - \Gamma \sum_i \sigma_i^z} \right)^n
\]

\[
= \sum_{\{\sigma_i^z\}_{i=1}^{\text{even}}} \sum_{\{\sigma_i^z\}_{i=1}^{\text{odd}}} \cdots \sum_{\{\sigma_i^z\}_{i=1}^{\text{even}}} \sum_{\{\sigma_i^z\}_{i=1}^{\text{odd}}} \cdot \cdot \cdot e^{2J \sum_{\langle ij \rangle} \sigma_i \sigma_j} \cdot e^{2\Gamma \sum_i \sigma_i^z} \cdot \cdot \cdot,
\]

\[
= \sum_{\{\sigma_i^z\}_{i=1}^{\text{even}}} \sum_{\{\sigma_i^z\}_{i=1}^{\text{odd}}} \cdots \sum_{\{\sigma_i^z\}_{i=1}^{\text{even}}} \sum_{\{\sigma_i^z\}_{i=1}^{\text{odd}}} \cdot \cdot \cdot e^{2J \sum_{\langle ij \rangle} \sigma_i \sigma_j} \cdot e^{2\Gamma \sum_i \sigma_i^z} \cdot \cdot \cdot,
\]
where $\text{Tr}_{d-1}$ denotes the sum all over the spin states including those inserted as a complete set of states $\{\sum_{m=1}^{n} |\sigma^m_i\rangle \langle \sigma^m_i| \} \ (m = 1, \ldots, n)$, 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:

$$H = J \sum_{(ij)} S^z_i S^z_j + D \sum_i (S^+_i)^2 + \Gamma \sum_i ((S^+_i)^2 + (S^-_i)^2), \quad [50]$$

where $S^\pm_i$ 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

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Fig. 15. Configurations of the quantum Monte Carlo simulation for the transverse Ising model. (a) Thermal fluctuation, and (b) Quantum fluctuation.
lowest energy con
figuration 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$_3$.\textsuperscript{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.\textsuperscript{113} This problem was examined by direct numerical studies of the ground state wave-function (PWFRG\textsuperscript{114}) and DMRG\textsuperscript{115}).\textsuperscript{116} There, a metamagnetic process similar to the classical one was revealed, and a phase diagram in the coordinate $(A, H)$ was obtained\textsuperscript{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\textsuperscript{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,\textsuperscript{119} and also coexistence of the solid state and superfluidity (supersolid)\textsuperscript{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,\textsuperscript{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 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

$$Z_1 = \sum_{\sigma = \pm 1} g_\sigma e^{-\beta D\sigma} = g_+ e^{-\beta D} + g_- e^{2D}. \tag{51}$$

This can be rewritten as

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

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

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

The importance of the interaction among molecules for the SC transition was pointed out in the observation of the specific heat,\textsuperscript{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 interaction among spin states $\mathcal{H}_{\text{int}}$, the Hamiltonian of the system is given by

$$\mathcal{H} = \mathcal{H}_{\text{int}}(\{\sigma_i\}) + \sum_i \left( D - \frac{1}{2} k_B T \ln(g) \right) \sigma_i. \tag{54}$$
The simplest choice of the interaction is the ferromagnetic Ising model\(^{123}\) \(H_{\text{int}} = -J \sum_{\langle i,j \rangle} \sigma_i \sigma_j\),

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
\[
F(m, T, H) = \frac{1}{2} z J m^2 - k_B T \ln(2 \cosh(\beta z J m + \beta H)),
\]

where \(m = \langle \sigma_i \rangle\). The spinodal point is given by
\[
\frac{\partial F(m, T, H)}{\partial m} = 0 \quad \text{and} \quad \frac{\partial^2 F(m, T, H)}{\partial m^2} = 0,
\]
and we have the spinodal field as
\[
H_{\text{SP}} = \pm z J \sqrt{1 - \frac{1}{4 \beta^2} \ln \left(1 + \frac{1 - \frac{1}{4 \beta^2}}{1 - \frac{1}{4 \beta^2}}\right)}.
\]

Let the critical temperature of the system be \(T_C = z J\), 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 \(z J\).

It has turned out that there are various ordering patterns of the HS fraction\(^{124,125}\)
\[
f_{\text{HS}}(T) = \left(\frac{m(T, H)}{H} + 1\right)/2.
\]

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 \(T_C\) (along the line I). In this case the change of \(m(T, H)\) along this line is smooth. That is, the change of \(f_{\text{HS}}(T)\) for large \(D\) is smooth.

On the other hand, for small \(D\), the line crosses the line of \(H = 0\) at a temperature below \(T_C\) (the lines III–V). There, \(f_{\text{HS}}(T)\) changes discontinuously at \(H = 0\), and \(f_{\text{HS}}(T)\) shows a thermal hysteresis between the lines of the spinodal fields. The relation \([53]\) indicates that the thermal hysteresis takes place for
\[
D < D_{C1} = \frac{1}{2} k_B T_C \ln g.
\]

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

If \(D < z J\) and \(D < D_{C1}\), 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 slope 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 \(D < z J\) and \(D > D_{C1}\), 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
\[
D_{C2} = z J.
\]

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
\[
D_{C3} = z J \tanh \left(\frac{1}{2} \ln g\right).
\]
In Fig. 18, types of ordering processes corresponding the lines (I–V), and the phase diagram in the coordinate \((D/zJ, \ln g)\) 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 \(f_{HS}(T)\)." 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. 18. Phase diagram of the ordering patterns \(f_{HS}(T)\) in the coordinate \((D/zJ, \ln g)\). Ordering patterns for various values of \(D\): (Type I) smooth change for \(D > D_{C_1}\); (Type II) smooth change and a metastable branch at low temperatures for \(D > D_{C_1}\) and \(D < D_{C_2}\); (Type III) discontinuous change for \(D_{C_1} > D > D_{C_2}\); (Type IV) discontinuous change and a metastable branch at low temperatures for \(D_{C_1} > D > D_{C_2}\) and (Type V) HS is always stable or metastable for \(D < D_{C_2}\).
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.,

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

where \(r_{ij}\) is the distance between the \(i\)th and \(j\)th 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.\(^{134}\)
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, 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.

Acknowledgments

The author would like to thank all the collaborators. He also thank the Academy for this opportunity to summarize my works on phase transitions.

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(Received Feb. 4, 2010; accepted May 25, 2010)
Profile

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