Partial order in a frustrated Potts model

Ryo Igarashi$^{1,2}$ and Masao Ogata$^3$

$^1$CCSE, Japan Atomic Energy Agency, Hyashi-Ueno, Taito-ku, Tokyo 110-0015, Japan
$^2$CREST(JST), Honcho, Kawaguchi, Saitama 433-0012, Japan
$^3$Department of Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

(Dated: July 22, 2009)

We investigate a 4-state ferromagnetic Potts model with a special type of geometrical frustration on a three dimensional diamond lattice by means of Wang-Landau Monte Carlo simulation motivated by a peculiar structural phase transition found in $\beta$-pyrochlore oxide KOs$_2$O$_6$. We find that this model undergoes unconventional first-order phase transition; half of the spins in the system order in a two dimensional hexagonal-sheet-like structure, while the remaining half stay disordered. The ordered sheets and the disordered sheets stack one after another. We obtain a fairly large residual entropy at $T = 0$ which originates from the disordered sheets.

PACS numbers: 75.10.Hk, 05.50.+q, 05.10.Ln, 02.70.Uu

I. INTRODUCTION

Generally speaking, frustrated systems have some constraints that forbid simultaneous minimization of all the interaction energies. Therefore, frustration usually suppresses phase transitions to long-range orders, and, as a result, leads to very rich low temperature phases. Moreover, a frustrated system may become a spin-liquid phase or sometimes exhibit phase transition to a partially ordered state. In this paper, we study a 4-state ferromagnetic Potts model with a special type of geometrical frustration on a three-dimensional diamond lattice. This model is proposed as a simplified model Hamiltonian for magnetic Potts model with a special type of geometrical frustration.

Let us briefly summarize experimental results of $\beta$-pyrochlore superconductors, AOs$_2$O$_6$ (A is one of K, Rb or Cs)\cite{ref1, ref2}, from which the particular model studied in this paper is derived. We mainly focus on the anomalous phase transition other than superconducting transition in KOs$_2$O$_6$. The pyrochlore structure in general is of type $\beta$-pyrochlore KOs$_2$O$_6$. Although the obtained phase transition in this model does not explain that in KOs$_2$O$_6$, we find that it has several interesting properties as a model with geometrical frustration.

We study a 4-state ferromagnetic Potts model with a special type of geometrical frustration on a three-dimensional diamond lattice. This model is proposed as a simplified model Hamiltonian for magnetic Potts model with a special type of geometrical frustration.

Let us briefly summarize experimental results of $\beta$-pyrochlore superconductors, AOs$_2$O$_6$ (A is one of K, Rb or Cs)\cite{ref1, ref2}, from which the particular model studied in this paper is derived. We mainly focus on the anomalous phase transition other than superconducting transition in KOs$_2$O$_6$. The pyrochlore structure in general is of type $\beta$-pyrochlore KOs$_2$O$_6$. Although the obtained phase transition in this model does not explain that in KOs$_2$O$_6$, we find that it has several interesting properties as a model with geometrical frustration.

II. MODEL

Having the peculiar first-order phase transition of KOs$_2$O$_6$ described above in mind, we study a simple classical model with special kind of geometrical frustration, which was proposed by Kums\'es \textit{et al.} using the density functional calculation\cite{ref3, ref4}. They investigated that the on-site potential of the K atoms along with K-K bond direction is heavily unharmonic and very flat near the symmetry center, which is formed by osmium and oxygen surrounding the K ion is over-sized.

The cages formed by OsO$_6$ octahedra in the $\beta$-pyrochlore structure have 4 holes towards nearest-neighbor K atoms because the 16c sites are empty. Therefore these four holes in the cages are not only the origin of anisotropic potential of a K atom but also the origin...
Interaction can be neglected. The flat potential and the caging will play the role of metallic screening which represents the $sp^3$-like structure which point to the nearest-neighbor K atoms. We assign these states as the four states of the classical Potts model. Note that we have one-to-one correspondence between the Potts variable 1 to 4 and the direction in the diamond lattice.

We concentrate on the inter-site Coulomb couplings represented in the lowest 4 states. Although there are $4^4$ matrix elements, $\langle \alpha|\beta|\alpha'|\beta' \rangle$, the largest contribution comes from the diagonal matrix elements, $\langle \alpha|\beta|\alpha |\beta \rangle$. These matrix elements are estimated to be

$$\langle \alpha|\beta|W|\alpha|\beta \rangle = -2J_1\delta_{\alpha\beta} + 2J_2\delta_{\alpha R}\delta_{\beta R} - J_3(\delta_{\alpha R} + \delta_{\beta R}) \tag{1}$$

$$(J_1 = 162K, J_2 = 371K, J_3 = 301K)$$

where $\alpha$ and $\beta$ denotes the direction of the bond, $\delta_{\alpha\beta}$ is the Kronecker delta and $R$ is a so-called “bond number”, which represents the 4 type of bond directions in the diamond lattice structure and takes the value of 1 to 4. For example, $R = 1$ means that the direction of the bond is $\langle 111 \rangle$, and similarly the basis state with $\alpha = 1$ represents one of the four wavefunctions extending in the $\langle 111 \rangle$ direction. 4 types of typical configurations are shown in Fig. 2. The first term in the right-hand-side of eq. (1) represents the energy gain of $2J_1$ when K atoms on the both ends of a bond are in the same direction $\alpha = \beta$. For example, the configuration A and B in Fig. 2 corresponds to this case. The second term in the right-hand-side of eq. (1) means that there is an energy loss of $2J_2$ when the bond direction ($R$) and the directions of the states $|\alpha \rangle$ and $|\beta \rangle$ at the both ends of the bonds are all in a straight line. The configuration A in Fig. 2 also corresponds to this case. The third term in the right-hand-side of eq. (1) represents the energy gain of $J_3$ when the wavefunction of one side is parallel to the bond direction. This corresponds to the configuration C in Fig. 2. As a result, configuration A in Fig. 2 has an energy, $-2J_1 + 2J_2$, configuration B, $-2J_1$, configuration C, $-J_3$, and configuration D, zero. We can neglect the $J_3$ terms because these terms corresponds to the constant value after summation of all bonds.

The off-diagonal terms, such as $\langle \alpha|\beta|W|\alpha|\gamma \rangle$, is of order smaller than $J_1$, $J_2$ and $J_3$. Therefore we neglect the off-diagonalized terms in the following. Furthermore, we assume that the effects of the excited states higher than the lowest four states are negligible. Finally, the effective Hamiltonian for the inter-site couplings of K atoms of rather strong Coulomb interaction between K atoms. The K atoms in the $\beta$-pyroclore KO$_2$O$_6$ form the diamond lattice structure. Thus the coupling between the nearest-neighbor K atoms is essentially of Coulomb origin and repulsive. Actually, as shown in the approximate length scale of the cage and the flat potential region of the K atoms in Fig. 1, the distance between cages is very small.

For discussing K atoms quantum mechanically, it is useful to use the states $|\alpha \rangle$ ($\alpha = 1, 2, 3, 4$) as the basis which represents the $sp^3$-like wavefunctions made out of 1$s$ and 2$p$ states, each located towards the holes of the cages. The cages will play the roll of metallic screening and thus we assume that the further long-range interaction can be neglected. The flat potential and the holes in cages lessen the energy difference between the singlet and triplet states and increases the energy difference to the higher energy states. Therefore we focus on the lowest four states and neglect the energy difference between the lowest singlet and triplet states. In this case we can assume the four-fold degenerate basis states, $|\alpha \rangle$ ($\alpha = 1, 2, 3, 4$), for each K atom in every cage. These states can be represented in the $sp^3$-like structure which point to the nearest-neighbor K atoms. We assign these states as the four states of the classical Potts model. Note that we have one-to-one correspondence between the Potts variable 1 to 4 and the direction in the diamond lattice.
The three dimensional 4-state ferromagnetic Potts model undergoes the first order transition. In order to handle the possible first order phase transition within the Monte Carlo simulation framework, we choose the Wang-Landau algorithm. This method enables us to calculate directly the density of states (DOS), \( g(E) \), and also allows us to efficiently sample the ground state. This algorithm is very effective for studying first-order phase transitions. The various thermodynamical quantities are also obtained very accurately even near the first-order phase transition temperature. Furthermore, this method can give estimates of the accurate ground state structure, the ground state energy and residual entropy, all of which are not obtained before.

This algorithm works as follows. Since the density of states, \( g(E) \), is \textit{a priori} not known at the very beginning of the simulation, we first simply set \( g(E) = 1 \) for all possible energies \( E \). Then we continue to update \( g(E) \) until it converges to a reasonable functional form and the energy histogram \( h(E) \) becomes flat. We construct a Markov chain of microscopic configuration \( \{ \mu \} \) using the local update scheme. We accept the new configuration using the transition probability

\[
p(\mu_1 \rightarrow \mu_2) = \min \left( \frac{g(E_{\mu_2})}{g(E_{\mu_1})}, 1 \right),
\]

where \( E_{\mu_1} \) and \( E_{\mu_2} \) are the energy of the system in a specific configuration \( \mu_1 \) and \( \mu_2 \), respectively. The calculated density of states \( g(E) \) and the energy histogram \( h(E) \) are updated regardless of the acceptance of the new configuration as

\[
\ln g(E_\mu) \rightarrow \ln g(E_\mu) + \ln f_i,
\]

\[
h(E_\mu) \rightarrow h(E_\mu) + 1,
\]

where \( f_i \) is a modification factor of the \( i \)th step of the Wang-Landau algorithm, as defined below. At first, \( f_1 \) is chosen as \( f_1 = e \). After some iteration, we then check the “flatness” of the obtained energy histogram \( h(E) \) by using a criteria that the minimum value of \( h(E) \) is not less than 80% of the average of the histogram, i.e.,

\[
h(E)|_{\text{min}} \geq 0.8 \times \langle h(E) \rangle.
\]

When the “flatness” of the histogram is reached, go to the \((i+1)\)th step of the Wang-Landau algorithm, by changing the modification factor as

\[
\ln f_{i+1} = a \ln f_i,
\]

\[
(0 < a < 1)
\]

where we choose \( a = 0.5 \), and also reset the energy histogram. We repeat until the \( i = 16 \) Wang-Landau step. The choice of \( i = 16 \) and \( a = 0.5 \) gives a reasonable convergence of \( g(E) \) for the 4-state Potts model.

We carry out the Monte Carlo simulations with different seeds for pseudo random number generator and calculated the average and variance of each quantity for \( J_2 = \infty \) and \( J_2 = 10 \). We use \( 8^3 = 512 \) unit cells, each consists of 8 atoms and therefore the total number is 4096.

### IV. RESULTS

Figure 4 shows the results of energy density for the case of \( J_2 = \infty \). We can see a clear kink at \( T \sim 0.44 \) which indicates a first-order phase transition of the model. We also find that the ground state energy density is equal to

\[
\text{energy density}
\]

\[
\text{ground state energy density}
\]

\[
\text{residual entropy}
\]

\[
\text{ground state temperature}
\]
to $-1$ per site within the statistical error. If $J_2 = 0$, the ground state energy density should be exactly $-2$, because the model is reduced to the ordinary 4-state ferromagnetic Potts model with $2N$ bonds in the diamond lattice. This result indicates that the ground state of the present model is different from the simple ferromagnetic ground state due to the frustration induced by $J_2$ term.

In order to confirm the first-order phase transition, we study the weight factor, $g(E) \exp(-\beta E)$, as a function of the energy density $E$. Figure 5 shows the obtained weight factor of $J_2 = 10$ and $T = 0.435$ case. The double-peak structure is a clear evidence of the first-order phase transition. We confirmed that this double-peak structure disappears when the temperature is only slightly off the transition temperature, for example, at $T = 0.45$ or $T = 0.42$.

A snapshot of the ground state is shown in Fig. 6. We can see that half of the wavefunctions $|\alpha\rangle$ represented by Potts variables order in a two-dimensional hexagonal sheet-like structure, in which half sites are located slightly above the sheet and the remaining half slightly below. This sheet-like plane is perpendicular to the majority rattling direction. The sandwiched sheets between the directions of the majority wavefunction remain disordered consisting of the other three wavefunctions. This ground state structure is a very new and peculiar ground state structure induced by an interesting frustration interaction in the present model.

Let us now discuss the residual entropy density originating from the disordered sheets. The obtained entropy density as a function of $T$ in the case with $J_2 = 10$ is shown in Fig. 7. We find that the entropy density in the $T \rightarrow \infty$ limit converges to 1.092(6) when we set the ground-state entropy to be zero. The correct entropy density in the $T \rightarrow \infty$ limit should become

$$\langle S \rangle (T \rightarrow \infty) = \log 4,$$

since every site has 4 degrees of freedom in the present model. Therefore the residual entropy density in the present model is calculated as

$$\langle S_0 \rangle = \log 4 - 1.092 = 0.294.$$

This residual entropy density must come from the degenerate ground state of the disordered sheets as shown in Fig. 8.
In the following, we consider the residual entropy in detail. Two examples of the ground state configurations are shown in Fig. 8. Here the bonds with “bond number” 4 are perpendicular to the hexagonal sheet, and it is assumed that all the sites in the nearest-neighbor sheet have Potts variable 4. Therefore, the Potts variable 4 is forbidden in the sheets shown in Fig. 8. Therefore, if the Potts variables in the disordered sheets with \( N/2 \) sites are completely random, the total number of possible configurations is \( 3^{\frac{N}{2}} \) and the corresponding entropy density is

\[
\langle S'_0 \rangle = \frac{1}{N} \log 3^{\frac{N}{2}} = 0.5493. \tag{10}
\]

However, the residual entropy density in eq. (9) is much smaller than that calculated in eq. (10). The origin of this difference comes from the constraint inside the disordered sheet.

Actually, a bond direction and the two states at the both ends of the bond cannot be all parallel inside the disordered sheet. Even under the constraint, there are many possible configurations in the ground state. The upper figure and the lower figure of Fig. 8 look very different but they have exactly the same energy. The upper figure has a site-number unbalance. The \( \frac{N}{2} \) sites are filled with Potts variable 1 whereas only \( \frac{N}{2} \) sites are filled with Potts variable 2 and 3. In the lower figure, the sites are almost equally filled with the Potts variables 1, 2 and 3. Note that the number of the spins in the sheet shown in Fig. 8 is not divisible by 3 but there is no difficulty of filling Potts variables almost equally.

The effect of the constraint inside the sheets can be taken into account approximately as follows. Let us start from the completely random configurations which consists of \( 3^{\frac{N}{2}} \) states. If we focus on a single bond in the hexagonal sheet, we notice that the forbidden states for that bond are included. Since the configurations are chosen completely randomly, the probability of appearance of this forbidden state is \( \frac{1}{9} \). Here \( 3 \times 3 = 9 \) represents the total number of the states at the both ends of the corresponding bond. Therefore, for each bond, \( \frac{1}{9} \) configurations should be discarded. As a result, the total number of the allowed configurations can be estimated as

\[
3^{\frac{N}{2}} \left( 1 - \frac{1}{9} \right)^{\frac{N}{2}} = \frac{8^{\frac{N}{2}}}{3^N}, \tag{11}
\]

where \( \frac{3}{4}N \) is the number of bonds in the sheet. The resulting entropy density is given by

\[
S_0 = \log \left( \frac{8^{\frac{N}{2}}}{3^N} \right) = \log(1.586) = 0.461, \tag{12}
\]

which is closer to the numerically obtained residual entropy density, 0.294, than the completely random value 0.5493.

**FIG. 7:** Entropy density calculations of the extended Potts model with \( J_2 = 10 \) are shown. These results are of \( 8^3 \) unit cells and consists of 4096 sites and the calculation up to the temperature \( T = 3 \) is shown. The jump of the Entropy density is observed at \( T \approx 0.44 \) which corresponds to the first order phase transition temperature. The inset shows the calculation up to the temperature \( T = 30 \). The line denotes the value of 1.092.

**FIG. 8:** (Color Online) Two examples of the ground state configurations in a disordered sheet are shown. Both configurations satisfy periodic boundary condition. The yellow, green and blue ellipse denote Potts variable 1, 2 and 3, respectively.
V. DISCUSSION AND CONCLUSIONS

Let us compare the present results with experiments of KO$_2$O$_6$. Despite the various peculiar properties of the present model, it apparently shows directional symmetry breaking, which is not observed in KO$_2$O$_6$. The transition temperature of the model, 0.44$J_1$ $\sim$ 80K, is of order high compared with the experimentally obtained first order transition temperature of $T_p = 7.5$K. Thus, the simplified model proposed by Kuněs et al. does not explain the rattling transition in KO$_2$O$_6$. A more sophisticated model Hamiltonian will be necessary. Recently, Hattori and Tsunetsugu\cite{29} proposed another model for this rattling transition of KO$_2$O$_6$. They argued the rattling transition by introducing a fifth state in addition to the four states discussed above. Although they succeeded to explain the first-order phase transition without symmetry breaking, the physical origin of the fifth state is not clear. Moreover, a recent experiments shows that the lattice expands below $T_p$, which contradicts their prediction. Thus the rattling transition in KO$_2$O$_6$ remains an open question.

Although the present model is not applied to the transition in KO$_2$O$_6$, we found that this model has very peculiar features. The obtained phase transition is of first order, which is verified from the double peak structure of the weight factor shown in Fig. 5. The ground state spin configuration of this model shows that half of the spins in the system are ordered and form a hexagonal-sheet-like structure. The overall possible structures of the ground state snapshot configuration are illustrated in Fig. 6 and Fig. 8. The remaining half of the spins are distributed disordered even in the low temperature limit $T \to 0$, which gives the residual entropy density of 0.294(6). These results indicate a new kind of interesting ground state which will be worth further studying as a result of new type of frustration. We use the ALPS library for calculation.\cite{30}

Acknowledgments

We are grateful to Dr. Todo (Department of Applied Physics, University of Tokyo) for allowing to use the part of the ALPS paralib library. This work is supported in part by Global COE Program “the Physical Sciences Frontier”, MEXT, Japan. The computation in this work has in part been done using the facilities of the Supercomputer Center, Institute for Solid State Physics, University of Tokyo and the Next Generation Super Computing Project, Nanoscience Program, MEXT, Japan.
25. J. Schoenes, A.-M. Racu, K. Doll, Z. Bukowski, and J. Karpinski, Phys. Rev. B 77, 134515 (2008).
26. J. Kuneš and W. E. Pickett, Phys. Status Solidi A 203, 2962 (2006).
27. J. Kuneš and W. E. Pickett, Phys. Rev. B 74, 094302 (2006).
28. J. Kuneš and W. E. Pickett, Physica B 378, 898 (2006).
29. F. Y. Wu, Rev. Mod. Phys. 54, 235 (1982).
30. F. Wang and D. P. Landau, Phys. Rev. E 64, 056101 (pages 16) (2001).
31. F. Wang and D. P. Landau, Phys. Rev. Lett. 86, 2050 (2001).
32. D. P. Landau and F. Wang, Comp. Phys. Comm. 147, 674 (2002).
33. D. P. Landau, S.-H. Tsai, and M. Exler, Amer. J. Phys. 72, 1294 (2004).
34. M. Udagawa, M. Ogata, and Z. Hiroi, J. Phys. Soc. Jpn. 71, 2365 (2002).
35. K. Hattori and H. Tsunetsugu, J. Phys. Soc. Jpn. 78, 013603 (2009).
36. A. F. Albuquerque, F. Alet, P. Corboz, P. Dayal, A. Feiguin, S. Fuchs, L. Gamper, E. Gull, S. Gürler, A. Honecker, et al., (ALPS Collaboration), J. Magn. Magn. Mater. 310, 1187 (2007).