ANTIFERROMAGNET ON PYROCHLORE LATTICE IN SPHERICAL APPROXIMATION

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Abstract. We develop an $1/n$ expansion method for the systematic approximation of the solutions of classical spin systems (tri-dimensional vectors) which can describe general spin-spin interactions and phases with complex magnetization pattern. This method is applied to study the phase transitions observed in pyrochlore crystals (*e.g.* $Gd_2Ti_2O_7$) and the results have qualitative agreement with the experimental ones.

Key words: phase transition, antiferromagnetism, spherical model.

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1. INTRODUCTION

In the last decades, the experimental and theoretical study of antiferromagnets with geometric frustration showed remarkable properties, total different from the properties of other magnetic systems [1–3].

The general form of a pyrochlore system is $A_2B_2O_7$ where the A and B species are generally rare-earth or transition metal species (metallic ions), only one of them being magnetic. The typical example is the $Gd_2Ti_2O_7$ crystal. In the crystalline state, each of the metallic ions occupies a site of a lattice of corner sharing tetrahedrons. The magnetic ions are well described as classical Heisenberg spins, the most powerful interaction being the antiferromagnetic exchange interaction between nearest neighbors. For this interaction, the high degeneracy of the ground state explains how pyrochlore systems preserve a disordered state even at very low temperatures (1K). In the magnetic phase the spins are ordered in 4 sublattices with orthogonal magnetization. In an homogeneous magnetic field, the crystal has a complex phase diagram structure.

The theoretical study of pyrochlore compounds is usually limited to mean field theories [4, 5, 7]. In this paper we consider a different approach, based on the 1/n-expansion method, for the approximation of pyrochlore system. We build a spherical model, as 0-order approximation, to describe the transition to a state with non-collinear magnetization vectors. This approach allows us to obtain the following results:

- Considering only anti-ferromagnetic interactions between first order neighbors, the model doesn't has phase transition.

- Considering an additional isotropic ferromagnetic interaction between third-order neighbors, the model exhibits a phase transition to an ordered phase at low temperatures. The pure phases have different magnetization on four sublattices, corresponding to the four corners of the basic tetrahedron.
- Considering (an approximation of) the dipolar interaction, the magnetization directions in the four sublattices are correlated with the lattice geometry: the spins located in two corners of the tetrahedron are parallel with the opposite edge and have opposite orientations.

2. PYROCHLORE LATTICE

In order to define the pyrochlore lattice, we consider the vectors which give the positions of the corners of a regular tetrahedron of edge a:

$$\mathbf{e}_0 = \mathbf{0}, \mathbf{e}_1 = \frac{a}{\sqrt{2}}(\mathbf{i}_2 + \mathbf{i}_3), \mathbf{e}_2 = \frac{a}{\sqrt{2}}(\mathbf{i}_3 + \mathbf{i}_1), \mathbf{e}_3 = \frac{a}{\sqrt{2}}(\mathbf{i}_1 + \mathbf{i}_2)$$

where $\mathbf{i}_k$, $k = 1, 2, 3$ are versors of the Cartesian coordinate system. Let $\Delta = \{0, 1, 2, 3\}$ be the set of tetrahedron corners.

The vectors $\mathbf{e}_{ij} = \mathbf{e}_j - \mathbf{e}_i$, $i \neq j = 0, 1, 2, 3$ are the oriented edges of the tetrahedron ($\mathbf{e}_{0i} = \mathbf{e}_i$).

The sites of the pyrochlore lattice are given by Δ translations with linear combination of $\mathbf{e_i}$ with even integer coefficients:

$$L = \bigcup_n \tau(\sum_{i=1}^{3} 2n_i \mathbf{e}_i)\Delta, \tag{2.1}$$

where $n = (n_1, n_2, n_3) \in \mathbf{Z}^3$ and τ is the natural action of translations in $\mathbf{R}^3$ (*i.e.* $\tau(\mathbf{v})\mathbf{x} = \mathbf{v} + \mathbf{x}$).

Let $(\mathbf{r}, i)$ be the lattice sites, where $\mathbf{r} = \sum_{j=1}^{3} 2n_j \mathbf{e}_j$ gives the translation of Δ on which the site stands and $i = 0, 1, 2, 3$ gives its position in the tetrahedron.

Each site of L has 6 nodes (nearest neighbors) at distance a and it's the unique common corner of two tetrahedrons which are congruent with Δ. For example (for instance), the origin belongs to Δ and to the tetrahedron of corners

$\{0, \tau(-2\mathbf{e_1})1, \tau(-2\mathbf{e_2})2, \tau(-2\mathbf{e_3})3\}$ (which is not a translation of Δ). The nearest neighbors of $(\mathbf{r}, i)$ are $(\mathbf{r}, j)$, $j \in \Delta$, $j \neq i$ and $(\mathbf{r} - 2\mathbf{e}_{i,j}, j)$, $j \in \Delta$, $j \neq i$.

Further on, among the higher order neighbors we are interested only in the third order neighbors, placed at a distance $2a$. The lattice site $(\mathbf{r}, i)$ has 12 third-order neighbors: $(\mathbf{r} - 2\mathbf{e}_{j,j'}, i)$, $j \neq j' \in \Delta$.

3. ANTIFERROMAGNET ON THE PYROCHLORE LATTICE

We suppose that the lattice's sites are occupied by classical spins $\vec{S}_{(\mathbf{r},i)} = \left\{ S^{\alpha}_{(\mathbf{r},i)}, \alpha = 1,2,3 \right\}$, where $\vec{S}^2_{(\mathbf{r},i)} = 1$, which interact according to the formal Hamiltonian:

$$H = -\frac{1}{2} \sum_{(\mathbf{r},i)\neq(\mathbf{r}',i')} J(\mathbf{r}-\mathbf{r}')^{\alpha,\alpha'}_{i,i'} S^{\alpha}_{(\mathbf{r},i)} S^{\alpha'}_{(\mathbf{r}',i')} - \sum_{(\mathbf{r},i)} \vec{h_i} \vec{S}_{(\mathbf{r},i)} \tag{3.1}$$

Here the external magnetic field h is independent of the position of the tetrahedron $h^{\alpha}_{(\mathbf{r},i)} = h^{\alpha}_i$ and matrix J is the sum of the following matrices which describe the main types of interaction:

1. $J^{(1)}$ - isotropic antiferromagnetic exchange between nearest neighbors

$$J^{(1)}(0)^{\alpha,\alpha'}_{i.i'} = \delta_{\alpha,\alpha'}(\delta_{i,i'} - 1) = \delta_{\alpha,\alpha'}(I - 4P_0)_{i,i'} \tag{3.2}$$

where I is the identity matrix and P_0 is the orthogonal projector on the vector $(1,1,1,1)$ in $\mathbf{C}^4$;

$$J^{(1)}(2\mathbf{e}_{j,j'})^{\alpha\alpha'}_{i,i'} = \delta_{\alpha,\alpha'}\delta_{i,j}\delta_{i',j'} = \delta_{\alpha,\alpha'}(E_{jj'})_{ii'} \tag{3.3}$$

Finally,

$$J^{(1)}(\mathbf{r})^{\alpha\alpha'}_{i,i'} = 0 \tag{3.4}$$

elsewhere (*e.g.* for $|\mathbf{r}| > 2a$)

2. $J^{(2)}$ - isotropic ferromagnetic exchange between third order neighbors:

$$J^{(2)}(\mathbf{r})^{\alpha\alpha'}_{i,i'} = J'\delta_{\alpha,\alpha'}\delta_{i,i'}, J' > 0 \tag{3.5}$$

for $|\mathbf{r}| = 2a$ and zero elsewhere.

3. $J^{(3)}$ - dipolar interaction

$$J^{(3)}(\mathbf{r})^{\alpha\alpha'}_{i,i'} = \mu^2 \frac{3(\mathbf{r}+\mathbf{e}_{i,i'})^{\alpha}(\mathbf{r}+\mathbf{e}_{i,i'})^{\alpha'} - \delta_{\alpha,\alpha'}|\mathbf{r}+\mathbf{e}_{i,i'}|^2}{|\mathbf{r}+\mathbf{e}_{i,i'}|^5} \tag{3.6}$$

where μ is the magnetic dipole. The dipolar interaction is the only one which couples different components of the spins (it breaks the $O(3)$ symmetry) in a manner which depends on their position in the crystal, and is thus able to explain the connection between the directions of the sublattices magnetizations and the geometry of the lattice. On the other hand, handling it is difficult because its long (infinite) range of action. Hereafter, for simplicity, we will treat only the main term; namely, we will restrain the dipolar interaction to the nearest neighbors. On this purpose, we will use the matrix $\tilde{J}^{(3)}$ defined by

$$\tilde{J}^{(3)}(\mathbf{r})^{\alpha\alpha'}_{i,i'} = J^3(\mathbf{r})^{\alpha\alpha'}_{i,i'} \tag{3.7}$$

if $|\mathbf{r}+\mathbf{e}_{i,i'}| = a$ and zero elsewhere.

4. THE SPHERICAL VECTOR MODEL

In order to build a spherical vector model, we consider n copies of the model built in previous section with relaxed restriction on spin lenght: instead of considering the spins of each copy of length unity (1), we impose the condition that the sum of the squares of the spine lengths over the n copies should be (equal to) n. In this way, a coupling of the n copies is introduced. In the limit $n \to \infty$ the n copies are being decoupled and each copy has a Gaussian distribution. This distribution is the spherical vector model which we consider as the 0-order approximation of the initial model. Therefore, in contrast to the mean field theory, the entropic factor is supra-estimated and we expect that the two approaches would be complementary. The convergence proof of the n-vector model to the spherical model and the calculus of the corrections in $1/n$ are similar to those for the isotropic Heisenberg model [8, 9], so we will confine to describe and solve the limit model. The limit model is a Gaussian model and its covariance is obtained as a solution of the following system of equations:

$$\frac{1}{\beta}(\gamma - J)^{-1}_{\mathbf{r},j,\alpha;\mathbf{r},j,\alpha} = 1 - [(\gamma - J)^{-1}h]^2_{\mathbf{r},j,\alpha}\,. \tag{4.1}$$

Here, $\gamma = (\delta_{\alpha,\alpha'}\gamma_{\mathbf{r},j,\alpha})$ is the diagonal matrix of spherical fields and J is the matrix of interactions $J = J^{(1)} + J^{(2)} + J^{(3)}$, defined in the previous section. Magnetization at the node $(\mathbf{r}, i)$ of the lattice is the average value of the spin:

$$m_{\mathbf{r},i,\alpha} = \langle S^{\alpha}_{(\mathbf{r},i)} \rangle = [(\gamma - J)^{-1}h]_{\mathbf{r},i,\alpha} \tag{4.2}$$

On finite volume, the system (4.1) has a unique solution on the $\gamma - J > 0$ domain. In the limit of infinite volume, the solution for the finite volume converges to the solution of the infinite system, when this solution exist. The existence of the solution is ensured by conveniently choosing the magnetic field so that $\gamma - J$ stays uniform strictly positive definite. By vanishing the magnetic field to zero, we obtain the system:

$$\frac{1}{\beta}(\gamma - J)^{-1}_{\mathbf{r},j,\alpha;\mathbf{r},j,\alpha} = 1 - m^2_{\mathbf{r},j,\alpha}\,, \tag{4.3a}$$

$$(\gamma - J)m = 0. \tag{4.3b}$$

In the limit of infinite volume, considering the symmetries of the system and the uniqueness of the solution, we can prove that γ does not depend on $\mathbf{r}, j, \alpha$, therefore it is a multiple of the unit matrix. In this way, we can replace the equations (4.3a) by a single equation, which we obtain by summing over j, α and the infinite system (4.3) is reduced to a system of 13 equations for the unknowns γ and $m^{\alpha}_i, \alpha = 1, 2, 3; i = 0, 1, 2, 3$. We will rewrite the system (4.3) using the unitary operator: $l_2(L^3) \to$

$L_2(B;\mathbf{C}^{12})$ given by the Fourier transform ($B=(-\pi,\pi]^3$):

$$\hat{x}(\mathbf{q})_j^\alpha=\frac{1}{(2\pi)^3}\sum_r e^{i(\mathbf{q},\mathbf{r})}x^\alpha(\mathbf{r},j) \tag{4.4}$$

where the sum is over all the translations of Δ, *i.e.* $\mathbf{r}=\sum\limits_{i=1}^{3}2n_i\mathbf{e}_i$; $n_i\in\mathbf{Z}$. We obtain:

$$\frac{1}{\beta}\int_B tr[(\gamma-\hat{J}(\mathbf{q}))^{-1}]d\mathbf{q}=1-\sum_i\vec{m}_i^2 \tag{4.5a}$$

$$(\gamma-\hat{J}(0))\mathbf{m}=0. \tag{4.5b}$$

Here $\hat{J}(\mathbf{q}))$ is a matrix in $\mathbf{C}^3\otimes\mathbf{C}^4$ and $\mathbf{m}=(\vec{m}_0,\vec{m}_1,\vec{m}_2,\vec{m}_3,)$ are the magnetizations of the spines at the tops of the tetrahedron Δ.

We will now calculate the matrix function $\hat{J}(\mathbf{q})$ which appears in (4.5). On this purpose we denote:

$$z_j=e^{i(\mathbf{q},\mathbf{e}_{0j})/a};\mathbf{z}=(1,z_1,z_2,z_3)\in\mathbf{C}^4. \tag{4.6}$$

1. Using the equations (3.2), (3.3), (3.4) we have, for the antiferromagnetic exchange interaction:

$$\hat{J}^{(1)}(\mathbf{q})=-I_3\otimes[2(P_0+P_z)-I] \tag{4.7}$$

where I_3 is the unit matrix in the spin space (indicating the isotropy of this interaction), P_0 and P_z are the orthogonal projectors in $\mathbf{C}^4$ on the vectors $\mathbf{1}=(1,1,1,1)$ and $\mathbf{z}$, respectively. As the projectors are positive operators and the sum of positive operators is positive, equation (4.7) gives immediately the following property of the spectra of matrix $\hat{J}^{(1)}(\mathbf{q})$:

Proposition 1. *For any* $\mathbf{q}\in B,\hat{J}^{(1)}(\mathbf{q})$ *has a maximum eigenvalue* $\lambda_0=1$ *which has as corresponding eigen-vectors all the vectors* $\mathbf{x}^\alpha,\alpha=1,2,3$ *with orthogonal components (in* $\mathbf{C}^4$*) on* $\mathbf{1}$ *and on* $\mathbf{z}$.

It follows, as a consequence:

Proposition 2. *If* $J'=0$ *and* $\mu=0$*, i.e., the only interaction is the isotropic anti-ferromagnetic exchange, then the system* (4.5) *has, for any temperature, an unique solution* γ *with* $\gamma-J>0$*. In particular, the system has no phase transition.*

Actually, the trace in equation (4.5a) contains the maximum term $\gamma-\lambda_0$ which is independent of $\mathbf{q}$, hence the integral contains this term, which is divergent when $\gamma\to 1$ impling the existence of the solution. Because $\gamma-J>0$, from equation (4.5b) it follows $\mathbf{m}=0$.

2. The ferromagnetic exchange interaction between third order neighbors has

a contribution to $\hat{J}(\mathbf{q})$ with a multiple of the unit matrix:

$$\hat{J}^{(2)}(\mathbf{q}) = J' \left(\sum_{j \neq j' \in \{0,1,2,3\}} z_j \bar{z'} \right) I_3 \otimes I = J' \left(\left| \sum_{j=0}^{3} z_j \right|^2 - 4 \right) I_3 \otimes I \qquad (4.8)$$

The function $\left| \sum_{j=0}^{3} z_j \right|^2 - 4$ has a non-degenerate maximum equal to 12 at $\mathbf{q} = 0$. As a consequence the matrix $\hat{J}^{(1)}(\mathbf{q}) + \hat{J}^{(2)}(\mathbf{q})$ doesn't have the maximum eigenvalue independent of $\mathbf{q}$, and $\int_B tr[\left(\gamma - \hat{J}^{(1)}(\mathbf{q}) - \hat{J}^{(2)}(\mathbf{q})\right)^{-1}] d\mathbf{q} < \infty$. The consequence of this observation, for the considered model, is the following:

Proposition 3. *If the only interactions in the system are $J^{(1)}$ and $J^{(2)}$ (i.e. the dipolar interaction is neglected) then the model presents a phase transition at the temperature*

$$T_c = 1 / \int_B tr[\left(\gamma - \hat{J}^{(1)}(\mathbf{q}) - \hat{J}^{(2)}(\mathbf{q})\right)^{-1}] d\mathbf{q} \qquad (4.9)$$

For $T < T_c$ the pure states are characterized by $\sum_{i=0}^{3} \vec{m}_i = 0$ and $\vec{m}_0^2 = \vec{m}_1^2 = \vec{m}_2^2 = \vec{m}_3^2 \neq 0$.

Indeed, for $\mathbf{q} = 0$, $P_z = P_0$ and, in view of equation (4.7) and (4.8) we see that equation (4.5b) admits as its solutions all the vectors $\mathbf{m}$ which satisfy the last condition. The first condition results from the equivalence of the four corners of Δ and the common value of the magnetizations lengths is, accordingly to (4.5a), $\vec{m}_i^2 = (T_c - T)/T_c$.

3. The dipolar interaction $\tilde{J}^{(3)}$ has the Fourier transform

$$\hat{J}^{(3)}(\mathbf{q}) = \frac{1}{2}[J^{(3)}(0) + M(\mathbf{z}) J^{(3)}(0) M(\bar{\mathbf{z}})] \qquad (4.10)$$

where $M(\mathbf{z})$ is the diagonal matrix with the diagonal $\mathbf{z}$ and the matrix $J^{(3)}(0)$ is expressed by 4×4 matrices of size 3×3:

$$J^{(3)}(0) = \{J_{i,i'}\}_{i,i' \in \{0,1,2,3\}} \text{ with } J_{i,i'} = \mu^2 / a^3 [3P_{i,i'} - I_3], \qquad (4.11)$$

where $P_{i,i'}$ is the orthogonal projector on the vector $\vec{\mathbf{e}}_{i,i'}$. It can be verified by direct calculation that the minimum eigenvalue of $J^{(3)}(0)$ is $-\mu^2/a^3$, a degenerate eigenvalue with multiplicity 3, with the corresponding eigenvectors

$$\mathbf{v}_{ij,kl} = ((\vec{\mathbf{e}})_{ij}, -\vec{\mathbf{e}}_{ij}, \vec{\mathbf{e}}_{kl}, -\vec{\mathbf{e}}_{kl}) \qquad (4.12)$$

where $\{i,j\}, \{k,l\}$ are opposite edges of Δ. As a consequence, we can make the following characterization of the pure phases of this model, considering all the three interactions:

Proposition 4. *The spherical vector model described by the system of equations* (4.5) *presents a phase transition at the temperature*

$$T_c = 1/\int\limits_B tr[\Big(\gamma - \hat{J}^{(1)}(\mathbf{q}) - \hat{J}^{(2)}(\mathbf{q}) - \hat{J}^{(3)}(\mathbf{q})\Big)^{-1}]d\mathbf{q} \tag{4.13}$$

For $T > T_c$, there exists a unique paramagnetic phase $(\mathbf{m} = 0)$. For $T < T_c$, there exists 6 *pure phases magnetizated in* 4 *Neel sublattices so that*

$$\mathbf{m}(\mathbf{r}) = \mathbf{m}(0) = m\mathbf{v}_{ij,kl}, \tag{4.14}$$

where

$$m^2 = (T_c - T)/T_c. \tag{4.15}$$

5. CONCLUSIONS

In this paper we developed a method for systematic approximation of classical spins models with multicomponent phases. Unlike / in contrast to the usual n-vectorial model, which was successfully used to describe isotropic ferromagnets, we here consider n copies of one model of three-dimensional spins in the $n \to \infty$ limit. The convergence to a limit Gaussian model is proved by analogy to the proof for the regular n-vectorial model. We must notice that the essential point in this proof is not the system isotropy, but the n copies equivalence, *i.e.* the complete permutational symmetry. The spherical vector model that we obtain in the 0-order of the asymptotic series is able to describe models with multidimensional order parameters. Unlike the mean field approximation, the spherical approximation overrates the entropic factor, and the two are complementary descriptions.

We apply this method to study the magnetic ordering in pyrochlore crystals which presents experimental established properties due to the high grade of geometric frustration of anti-ferromagnetic interaction between the closest neighbors: very low critical temperature (1 K) in relation with the measured Curie-Weiss temperature (10 K); the existence of four sublattices with non-collinear magnetizations, etc. The model we use explain all these particularities, and highlights the role of each spin-spin interaction. As a main result, we prove that frustrated anti-ferromagnetic interaction has the tendency to suppress transition; the ferromagnetic interaction between high-order neighbors leads to transition in a state with four sublattices, but with high degenerate phases; the dipolar interaction is responsible for the selection of only six sublattices magnetization patterns, which are correlated with the lattice geometry in concordance with experimental observations.

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