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Distortion of frustrated spin cluster caused by quantum fluctuation due to magneto-elastic coupling

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Abstract
We study the effects of magneto-elastic coupling on frustrated spin states of the antiferromagnetic Heisenberg model on a regular triangle and tetrahedron made of spin $1/2$ or 1. Displacement of spin is considered as a quantum-mechanical variable. Distortion of clusters occurs through quantum fluctuation of normal modes caused by magneto-elastic coupling. The nonmagnetic phase transitions of vanadium and chromium spinels are discussed.

1. Introduction

Reduction of degeneracy of the ground state for frustrated spins on the pyrochlore lattice by lattice distortion has been discussed by several authors for classical vector spin [1–4] and for quantum spin [5, 6]. Characteristics of the vibronic state have also been studied [7]. In these works, the displacement of spin was considered to be classical. Recently, the quantum-mechanical phonon has been considered [8]. Then the distortion occurs through the quantum fluctuation of normal modes [9, 10].

In this paper, we study magneto-elastic coupling in frustrated spin states of the antiferromagnetic (AF) Heisenberg Hamiltonian on a regular triangle and tetrahedron made of spin 1/2 or 1, giving attention to the quantum fluctuation of normal modes. We discuss the structural phase transition of spinels [11, 12] on the basis of the tetragonal distortion caused by the quantum fluctuation.

2. Magneto-elastic coupling

The Heisenberg Hamiltonian for spins on a regular triangle and tetrahedron,

$$\mathcal{H}_0 = -2J_0 \sum\nolimits_{\ell<\ell'} \boldsymbol{s}_\ell \cdot \boldsymbol{s}_{\ell'}, \tag{1}$$

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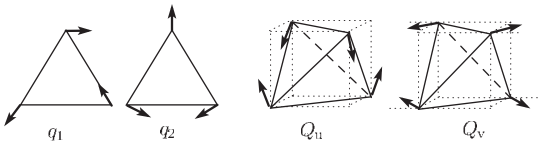

Figure 1. Displacement by doublet $E'(D_{3h})$ and $E(T_d)$ modes.

is invariant under the operations of D_{3h} and T_d groups, respectively. When the exchange parameter depends on the distance between spins, the spin states are perturbed by distortion.

Normal modes of the triangle are classified into the singlet A_1' (q_A, its normal coordinate) and doublet $E'(q_1, q_2)$ modes. Normal modes of the tetrahedron are classified into A_1 (Q_A), T_2 (Q_1, Q_2, Q_3) and E (Q_u, Q_v) modes. The singlet A_1' and A_1 modes are the breathing ones. The doublet E' and E modes are illustrated in figure 1.

The normal modes couple with the corresponding bases of the same representation made from linear combinations of $\boldsymbol{s}_\ell \cdot \boldsymbol{s}_{\ell'}$. The A_1' and E' representation for the D_{3h} group are given by

$$A_1': \quad f_A = \sum\nolimits_{\ell<\ell'} \boldsymbol{s}_\ell \cdot \boldsymbol{s}_{\ell'}/\sqrt{3}, \tag{2}$$

$$E': \quad f_1 = (\boldsymbol{s}_1 \cdot \boldsymbol{s}_2 - \boldsymbol{s}_3 \cdot \boldsymbol{s}_1)/\sqrt{2}, \qquad f_2 = (\boldsymbol{s}_1 \cdot \boldsymbol{s}_2 - 2\boldsymbol{s}_2 \cdot \boldsymbol{s}_3 + \boldsymbol{s}_3 \cdot \boldsymbol{s}_1)/\sqrt{6}. \tag{3}$$

The A_1, T_2 and E representations for the T_d group are given by

$$A_1: \quad F_A = \sum\nolimits_{\ell<\ell'} \boldsymbol{s}_\ell \cdot \boldsymbol{s}_{\ell'}/\sqrt{6}, \tag{4}$$

$$T_2: \quad \begin{cases} F_1 = (\boldsymbol{s}_1 \cdot \boldsymbol{s}_4 - \boldsymbol{s}_2 \cdot \boldsymbol{s}_3)/\sqrt{2}, \qquad F_2 = (\boldsymbol{s}_1 \cdot \boldsymbol{s}_3 - \boldsymbol{s}_2 \cdot \boldsymbol{s}_4)/\sqrt{2}, \\ F_3 = (\boldsymbol{s}_1 \cdot \boldsymbol{s}_2 - \boldsymbol{s}_3 \cdot \boldsymbol{s}_4)/\sqrt{2}, \end{cases} \tag{5}$$

$$E: \quad \begin{cases} F_u = [(\boldsymbol{s}_1 + \boldsymbol{s}_2) \cdot (\boldsymbol{s}_3 + \boldsymbol{s}_4) - 2(\boldsymbol{s}_1 \cdot \boldsymbol{s}_2 + \boldsymbol{s}_3 \cdot \boldsymbol{s}_4)]/2\sqrt{3}, \\ F_v = (\boldsymbol{s}_1 - \boldsymbol{s}_2) \cdot (\boldsymbol{s}_3 - \boldsymbol{s}_4)/2. \end{cases} \tag{6}$$

The perturbation Hamiltonian for the triangle is

$$\mathcal{H}' = \frac{1}{2m}({p_A}^2 + {p_1}^2 + {p_2}^2) + \frac{m{\omega_{A'}}^2}{2}{q_A}^2 + \frac{m{\omega_{E'}}^2}{2}({q_1}^2 + {q_2}^2) - 2[J'_{A'} q_A f_A + J'_{E'}(q_1 f_1 + q_2 f_2)], \tag{7}$$

where p_α is the momentum conjugate to q_α. The coupling constants J'_α are $J'_{A'} = \sqrt{3}J'$ and $J'_{E'} = \sqrt{3/2}J'$, where $J' = \partial J/\partial a$ and $a = |\boldsymbol{R}^0_\ell - \boldsymbol{R}^0_{\ell'}|$. For the tetrahedron,

$$\mathcal{H}' = \frac{1}{2m}({P_A}^2 + {P_1}^2 + {P_2}^2 + {P_3}^3 + {P_u}^2 + {P_v}^2) + \frac{m}{2}\left[{\omega_A}^2{Q_A}^2 + {\omega_T}^2({Q_1}^2 + {Q_2}^2 + {Q_3}^2) + {\omega_E}^2({Q_u}^2 + {Q_v}^2)\right] - 2\left[J'_A Q_A F_A + J'_T(Q_1F_1 + Q_2F_2 + Q_3F_3) + J'_E(Q_uF_u + Q_vF_v)\right], \tag{8}$$

where the J'_α are $J'_A = 2J'$, $J'_T = \sqrt{2}J'$ and $J'_E = J'$.

3. Spin states

The unperturbed Hamiltonian is rewritten as

$$\mathcal{H}_0 = -J_0\left[S(S+1) - \sum\nolimits_\ell s(s+1)\right], \tag{9}$$

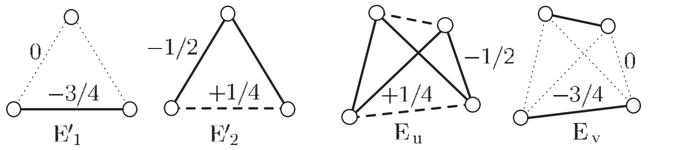

Figure 2. Spin correlations in the $E'(D_{3h})$ and $E(T_d)$ spin states.

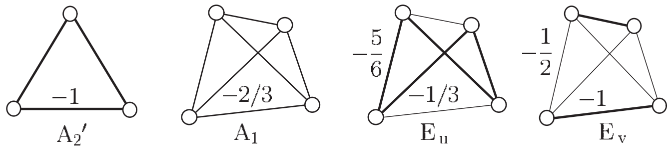

Figure 3. Spin correlations in the AF ground spin states for spin-1 clusters.

where S is spin quantum number of $\boldsymbol{S}(=\sum_\ell \boldsymbol{s}_\ell)$ and $s = 1/2$ or 1. The eigen-energy is specified by S and the order of degeneracy is known through the additive process of $\boldsymbol{s}_\ell$.

Spin 1/2. For the triangle, the AF ground state is doubly degenerate and is given as

$$\mathcal{H}_0|\mathrm{E}'i, S_z\rangle = (3/2)J_0|\mathrm{E}'i, S_z\rangle, \qquad i = 1, 2 \quad \text{for } S = 1/2. \tag{10}$$

The state vectors give the E′ representation for D_{3h} group. We put aside the Kramers degeneracy. In the subspace of $|\mathrm{E}'i, S_z\rangle$,

$$f_\mathrm{A} = -(\sqrt{3}/4)\sigma_1, \qquad f_1 = (\sqrt{6}/4)\sigma_x, \qquad f_2 = (\sqrt{6}/4)\sigma_z, \tag{11}$$

where σ_1 is the unit matrix, and σ_x and σ_z the Pauli matrices.

For the tetrahedron, the AF ground state is doubly degenerate and is given as

$$\mathcal{H}_0|\mathrm{E}\eta\rangle = 3J_0|\mathrm{E}\eta\rangle, \qquad \eta = \mathrm{u, v}, \quad \text{for } S = 0. \tag{12}$$

The state vectors give the E representation for T_d group. In the subspace of $|\mathrm{E}\eta\rangle$,

$$F_\mathrm{A} = -(\sqrt{6}/4)\sigma_1, \qquad F_\mathrm{u} = -(\sqrt{3}/2)\sigma_z, \qquad F_\mathrm{v} = -(\sqrt{3}/2)\sigma_x, \tag{13}$$

and $F_\tau = 0$ for $\tau = 1, 2, 3$. Note that the perturbation of T_2 symmetry is irrelevant because F_τ is vanishing and the product representation $T_2 \times E$ does not contain the E representation. The spin correlations in the ground states are shown in figure 2.

Spin 1. For the triangle, the AF ground state energy is $6J_0$ with $S = 0$ without degeneracy. The eigenvector $|\mathrm{A}_2'\rangle$ gives the A_2' representation for the D_{3h} group.

For the tetrahedron, the ground state is triply degenerate and is given as

$$\mathcal{H}_0|\mathrm{A}_1\rangle = 8J_0|\mathrm{A}_1\rangle, \qquad \mathcal{H}_0|\mathrm{E}\eta\rangle = 8J_0|\mathrm{E}\eta\rangle, \qquad \eta = \mathrm{u, v}, \quad \text{for } S = 0. \tag{14}$$

The eigenvectors give the A_1 and E representations for the T_d group. In the three-dimensional subspace of $\{|\mathrm{A}_1\rangle, |\mathrm{Eu}\rangle, |\mathrm{Ev}\rangle\}$,

$$F_\mathrm{A} = \frac{-2\sqrt{2}}{\sqrt{3}}\begin{pmatrix}1&0&0\\0&0&0\\0&0&0\end{pmatrix}, \qquad F_\mathrm{u} = \frac{-1}{\sqrt{3}}\begin{pmatrix}0&2\sqrt{5}&0\\2\sqrt{5}&1&0\\0&0&-1\end{pmatrix},$$
$$F_\mathrm{v} = \frac{1}{\sqrt{3}}\begin{pmatrix}0&0&-2\sqrt{5}\\0&0&1\\-2\sqrt{5}&1&0\end{pmatrix}. \tag{15}$$

The spin correlations in the ground states are shown in figure 3.

4. Distortion and fluctuation

By making use of the creation and annihilation operators, $b_\alpha^\dagger$ and b_α, for α mode, we rewrite $\mathcal{H}'$ and equations (7) and (8) as

$$\mathcal{H}' = \sum_\alpha \hbar\omega_\alpha(b_\alpha^\dagger b_\alpha + 1/2) - \sqrt{\hbar/2m\omega_\alpha}\, J'_\alpha(b_\alpha + b_\alpha^\dagger) f_\alpha, \tag{16}$$

where f_α denotes f_α for the triangle and F_α for the tetrahedron. Introducing modified operators

$$\tilde{b}_\alpha = b_\alpha - \sqrt{2/m\hbar\omega_\alpha^3}\, J'_\alpha f_\alpha, \qquad \tilde{b}_\alpha^\dagger = b_\alpha^\dagger - \sqrt{2/m\hbar\omega_\alpha^3}\, J'_\alpha f_\alpha, \tag{17}$$

we have

$$\mathcal{H}' = \sum_\alpha \hbar\omega_\alpha(\tilde{b}_\alpha^\dagger \tilde{b}_\alpha + 1/2) - (2/m{\omega_\alpha}^2) {J'_\alpha}^2 f_\alpha^2. \tag{18}$$

The commutators $[\tilde{b}_\alpha, \tilde{b}_\alpha^\dagger] = 1$ and $[\tilde{b}_\alpha, \tilde{b}_\alpha] = [\tilde{b}_\alpha^\dagger, \tilde{b}_\alpha^\dagger] = 0$ are boson-like although the $\tilde{b}_\alpha$ for E′ (D_{3h}) and E (T_d) modes with different α are not commutable because the commutators of the f_α between different α for E′ and E representations depend on the chirality of spins. The behaviour of the chiral parameter in the excited state has been studied by Yamasaki *et al* [7]. The complication of excitation due to the uncommutability arises in two or more modified phonon states with different modes. The first excited state is a single modified phonon state. So the ground state with respect to the modified phonon is simply obtained by

$$\tilde{b}_\alpha |\cdots\rangle_0 = 0. \tag{19}$$

Then, in the subspace of $|\cdots\rangle_0$,

$$q_\alpha \text{ (or } Q_\alpha) = \sqrt{\hbar/2m\omega_\alpha}\,(b_\alpha^\dagger + b_\alpha) = (2J'_\alpha/m{\omega_\alpha}^2) f_\alpha \tag{20}$$

and

$$\mathcal{H}' = \sum_\alpha - (2/m{\omega_\alpha}^2) J'^2_\alpha f_\alpha^2 + \hbar\omega_\alpha/2. \tag{21}$$

The lowest energy is obtained by the greatest eigenvalue of f_α^2.

Spin 1/2. In the subspace of E′ (D_{3h}) or E (T_d) spin states, the f_α^2 or F_α^2 are proportional to the unit matrix as seen from equations (11) and (13). Then, the changes in energy are

$$\begin{aligned} \delta\mathrm{E}' &= -3J'^2_{\mathrm{A}'}/8m\omega^2_{\mathrm{A}'} - 3J'^2_{\mathrm{E}'}/2m\omega^2_{\mathrm{E}'} \quad \text{(triangle)}, \\ \delta\mathrm{E}' &= -3J'^2_{\mathrm{A}}/4m\omega^2_{\mathrm{A}} - 3J'^2_{\mathrm{E}}/m\omega^2_{\mathrm{E}} \quad \text{(tetrahedron)}. \end{aligned} \tag{22}$$

The degeneracy is not lifted and the contribution of the E′ and E modes are twice that by the static model [5, 9].

In the subspace of the modified ground spin states,

$$q_{\mathrm{A}} = -\sqrt{3}/2(J'_{\mathrm{A}}/m{\omega_{\mathrm{A}}}^2)\sigma_1, \qquad (q_1, q_2) = \sqrt{3/2}J'_{\mathrm{E}'}/m\omega^2_{\mathrm{E}'}(\sigma_x, \sigma_z), \tag{23}$$

for the triangle and

$$Q_{\mathrm{A}} = -\sqrt{3/2}(J'_{\mathrm{A}}/m{\omega_{\mathrm{A}}}^2)\sigma_1, \qquad (Q_{\mathrm{u}},\ Q_{\mathrm{v}}) = -\sqrt{3}J'_{\mathrm{E}}/m{\omega_{\mathrm{E}}}^2(\sigma_z, \sigma_x) \tag{24}$$

for the tetrahedron by equations (17) and (19).

The apparent distortions are given by expectation values:

$$\langle \mathrm{E}'i|q_1|\mathrm{E}'i\rangle_0 = 0, \qquad \langle \mathrm{E}'i|q_{\mathrm{A}}|\mathrm{E}'i\rangle_0 = -\sqrt{3}/2J'_{\mathrm{A}'}/m{\omega_{\mathrm{A}'}}^2, \tag{25}$$

$$\langle \mathrm{E}'1|q_2|\mathrm{E}'1\rangle_0 = -\langle \mathrm{E}'2|q_2|\mathrm{E}'2\rangle_0 = \sqrt{3/2}J'_{\mathrm{E}'}/m\omega^2_{\mathrm{E}'}, \tag{26}$$

and

$$\langle \mathrm{Eu}|Q_{\mathrm{u}}|\mathrm{Eu}\rangle_0 = -\langle \mathrm{Ev}|Q_{\mathrm{u}}|\mathrm{Ev}\rangle_0 = -\sqrt{3}J'_{\mathrm{E}}/m{\omega_{\mathrm{E}}}^2, \tag{27}$$

$$\langle \mathrm{E}\eta|Q_{\mathrm{v}}|\mathrm{E}\eta\rangle_0 = 0, \qquad \langle \mathrm{E}\eta|Q_{\mathrm{A}}|\mathrm{E}\eta\rangle_0 = -\sqrt{3/2}J'_{\mathrm{A}}/m{\omega_{\mathrm{A}}}^2. \tag{28}$$

The expectation values of q_1 and Q_v are vanishing, i.e., the distortions due to these modes are smeared out by quantum fluctuation; then distortions due to q_2 and Q_u occur. The fluctuations are estimated as

$$\begin{aligned} \langle \mathrm{E}'i|{q_1}^2|\mathrm{E}'i\rangle_0 &= (3/2)J_{\mathrm{E}'}'^2/m^2\omega_{\mathrm{E}'}^4 + \hbar/2m\omega_{\mathrm{E}'}, \\ \langle \mathrm{E}\eta|{Q_v}^2|\mathrm{E}\eta\rangle_0 &= 3J_{\mathrm{E}}'^2/m^2{\omega_{\mathrm{E}}}^4 + \hbar/2m\omega_{\mathrm{E}}, \end{aligned} \tag{29}$$

of which the first terms on the right-hand side are equal to $\langle \mathrm{E}'\eta|q_2|\mathrm{E}'\eta\rangle_0{}^2$ and $\langle \mathrm{E}\eta|Q_u|\mathrm{E}\eta\rangle_0{}^2$, respectively. Although the distortion of q_1 and Q_v modes are smeared out, they have the same elastic energy as the q_2 and Q_u modes, respectively.

Spin 1. There is no degeneracy for the ground spin state of the triangle of spin 1. Moreover the symmetry of the ground spin state is A_2', which corresponds to the uniform rotation mode, so there is no coupling.

The lowest energy for the tetrahedron is calculated from equation (21) using the greatest eigenvalues of

$${F_A}^2 = \frac{8}{3}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \qquad {F_u}^2 + {F_v}^2 = \frac{1}{3}\begin{pmatrix} 40 & 4\sqrt{5} & 0 \\ 4\sqrt{5} & 22 & 0 \\ 0 & 0 & 22 \end{pmatrix}. \tag{30}$$

In the triply degenerate ground spin states, $\{|\mathrm{Ev}\rangle_0\}$ is separated from $\{|\mathrm{A}_1\rangle_0, |\mathrm{Eu}\rangle_0\}$. The eigenvalues of $\mathcal{H}'$ in $\{|\mathrm{A}_1\rangle_0, |\mathrm{Eu}\rangle_0\}$ are higher or lower than the value for the unperturbed state or that for $|\mathrm{Ev}\rangle_0$. Then, in contrast to the spin-1/2 cases, the degeneracy is lifted.

The expectation values of Q_α for each spin state are calculated from F_α by equation (20) as follows:

$$\langle \mathrm{A}_1|Q_A|\mathrm{A}_1\rangle_0 = -(4\sqrt{2}/3m{\omega_{\mathrm{A}_1}}^2)J_{\mathrm{A}_1}', \qquad \langle \mathrm{E}\eta|Q_A|\mathrm{E}\eta\rangle_0 = 0 \tag{31}$$

$$\begin{aligned} &\langle \mathrm{A}_1|Q_u|\mathrm{A}_1\rangle_0 = 0, \\ &\langle \mathrm{Eu}|Q_u|\mathrm{Eu}\rangle_0 = -\langle \mathrm{Ev}|Q_u|\mathrm{Ev}\rangle_0 = -(2/\sqrt{3}m{\omega_{\mathrm{E}}}^2)J_{\mathrm{E}}', \end{aligned} \tag{32}$$

and $\langle Q_v\rangle$ vanishes for any states.

The expectation values of ${Q_\alpha}^2$ are, apart from the zero-point motion,

$$\begin{aligned} &\langle \mathrm{A}_1|{Q_A}^2|\mathrm{A}_1\rangle_0 = \langle \mathrm{A}_1|Q_A|\mathrm{A}_1\rangle_0{}^2, \\ &\langle \mathrm{E}\eta|{Q_A}^2|\mathrm{E}\eta\rangle_0 = 0, \qquad \langle \mathrm{A}_1|Q_\eta|\mathrm{A}_1\rangle_0 = 0, \\ &\langle \mathrm{Ev}|{Q_u}^2|\mathrm{Ev}\rangle_0 = \langle \mathrm{Eu}|{Q_v}^2|\mathrm{Eu}\rangle_0 = \langle \mathrm{Eu}|Q_u|\mathrm{Eu}\rangle_0{}^2, \end{aligned} \tag{33}$$

which are not fluctuating, and

$$\langle \mathrm{E}\eta|{Q_\eta}^2|\mathrm{E}\eta\rangle_0 = (28/m^2{\omega_{\mathrm{E}}}^4)J_{\mathrm{E}}'^2. \tag{34}$$

The Q_η mode is strongly fluctuating by mixing $|\mathrm{A}_1\rangle_0$ with $|\mathrm{Eu}\rangle_0$.

5. Conclusion and discussion

For spin 1/2, the triangle and tetrahedron give very similar results to each other. Magneto-elastic coupling mixes the doublet spin state with the doublet phonon of the same symmetry. Then the degeneracy of the ground state is not lifted in spite of distortion. By the spin-1/2 tetramer model for spin-1 vanadium spinel [5], the tetragonal distortion at the non-magnetic phase transition is interpreted as a result of hidden ordering of $|\mathrm{Eu}\rangle_0$ and $|\mathrm{Ev}\rangle_0$ because the sign of distortion depends on the spin states.

The ground spin state of spin 1 for the tetrahedron is triply degenerate and classified into A_1 and E states. Magneto-elastic coupling hybridizes $|\mathrm{A}_1\rangle_0$ with $|\mathrm{Ev}\rangle_0$ and lifts the degeneracy and also brings about significant fluctuation of Q_η. Then the hidden ordering in the case of spin 1/2 may not appear as observed in spin-3/2 chromium spinel [12].

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