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Kyoto University
Frustrated quantum three-spins coupled with vibration modes:
Quantum chaos in the context of dynamical Jahn-Teller problem

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(Dated: January 29, 2005)

The triangular Heisenberg antiferromagnets play an important role in our understanding the resonating valence bond (RVB) state, in which the scalar chirality for three spins $S_1 \cdot (S_2 \times S_3)$ is expected to have a nonzero expectation value[1-4]. This subject has been a focus of recent experimental activities. We investigate a triangular cluster model of the Heisenberg antiferromagnet in which quantum spins are coupled with lattice vibrations, for the purpose to see magnetic properties of its high-lying states in relation to a typical dynamical Jahn-Teller system. The spin-lattice interaction is introduced by expanding the exchange interaction with respect to deviation of lattice displacements from equilibrium. We shall address the issue: the present model becomes equivalent to that of the well-known vibronic problem for $E_g \otimes e_g$ Jahn-Teller system.

Let us consider the quantum spin system where three spins of spin=1/2 are localized at lattice sites 1, 2 and 3 on triangle. The coupling between neighboring spins are expressed by the antiferromagnetic exchange interactions $J_A$, $J_B$ and $J_C$ as shown in Fig.1.

The corresponding Heisenberg Hamiltonian is

$$\mathcal{H} = J_A S_1 \cdot S_2 + J_B S_2 \cdot S_3 + J_C S_3 \cdot S_1. \quad (1)$$

We concentrate our attention on the spin state where $x$ component of the total spin satisfies $s_1x + s_2x + s_3x = 1/2$. Therefore, these bases are expressed explicitly as $| \uparrow \uparrow \uparrow \rangle, | \uparrow \downarrow \downarrow \rangle, | \downarrow \uparrow \downarrow \rangle, | \downarrow \downarrow \uparrow \rangle$, where arrows denote $s_jx$ for site $j$.

By using these bases, we obtain the Hamiltonian matrix,

$$\mathcal{H} = \begin{pmatrix}
| \uparrow \uparrow \uparrow \rangle & | \uparrow \downarrow \downarrow \rangle & | \downarrow \uparrow \downarrow \rangle & | \downarrow \downarrow \uparrow \rangle \\
-J_A + J_C & 2J_A & 2J_C & -J_A - J_B + J_C \\
2J_A & -J_A + J_B & 2J_C & 2J_B \\
2J_C & 2J_B & -J_A - J_B - J_C & 2J_A
\end{pmatrix}. \quad (2)$$

Next we introduce the interaction between the spins and lattice vibrations, noting the dependence of $J_A$, $J_B$ and $J_C$ on distances between spin sites. As for the lattice vibration, we employ the normal modes for the triangle; The normal $e_g$ modes, $Q_1$ and $Q_2$ which are degenerate are given in Fig.2. The remaining $a_{1g}$ mode(the breathing mode) has a much higher strain energy and is ignored hereafter. (There are other global degrees of freedom related to translation of the center of mass and to rotation around the axis perpendicular to the triangular plane. They however have nothing to do with lattice vibrations and are also ignored.) Then the spin-lattice interaction
FIG. 2: The normal modes $Q_1$ and $Q_2$ in the triangle.

is obtained as a result of the the expansion of $J_A$, $J_B$ and $J_C$ linearly in the $e_g$ modes as follows:

$$
J_A = J \cdot \left[ 1 + \frac{\alpha}{2} (Q_1 - \sqrt{3} Q_2) \right]
$$

$$
J_B = J \cdot \left[ 1 - \alpha Q_1 \right]
$$

$$
J_C = J \cdot \left[ 1 + \frac{\alpha}{2} (Q_1 + \sqrt{3} Q_2) \right]
$$

where $\alpha$ is the coupling constant.

Concerning the spin system on the other hand, we introduce the following bases introduced by Nakamura and Bishop for the triangular spin plaquet[5-7]:

$$
|k = 0\rangle = \frac{1}{\sqrt{3}} \left( |1 1 1\rangle + \frac{2}{\sqrt{3}} |1 1 1\rangle + |1 1 1\rangle \right)
$$

$$
|k = \frac{2\pi}{3}\rangle = \frac{1}{\sqrt{3}} \left( |1 1 1\rangle + e^{\frac{2\pi i}{3}} |1 1 1\rangle + e^{-\frac{2\pi i}{3}} |1 1 1\rangle \right)
$$

$$
|k = -\frac{2\pi}{3}\rangle = \frac{1}{\sqrt{3}} \left( |1 1 1\rangle + e^{-\frac{2\pi i}{3}} |1 1 1\rangle + e^{\frac{2\pi i}{3}} |1 1 1\rangle \right).
$$

These bases reflect clockwise and anticlockwise rotations of a spin configuration on the plane of the triangle. The wave numbers $k = 0, \pm 2\pi/3$ correspond to phase factors in Bloch’s theorem for the system with discrete rotational symmetry. From a viewpoint of the ligand-field theory, the construction of the bases (4) from $|1 1 1\rangle$, $|1 1 1\rangle$ and $|1 1 1\rangle$ is regarded as a formation of $E_g$ and $A$ representations in $D_{3d}$ symmetry from the triply-degenerate $T_{2g}$ ones in $O_h$ symmetry. By using this new bases, the Hamiltonian matrix (2) can be transformed to

$$
\mathcal{H}/\left(-\frac{3}{4} \hbar^2 J\right) = 
\begin{pmatrix}
1 & 1 & 0 \\
0 & 0 & -1 \\
\alpha(-Q_1 - iQ_2) & \alpha(-Q_1 + iQ_2) & -1
\end{pmatrix}
$$

$$
|k = 0\rangle |k = \frac{2\pi}{3}\rangle |k = -\frac{2\pi}{3}\rangle
$$

From Eq.(5) we find that the $k = 0$ manifold is completely separated from other manifolds, i.e., $\mathcal{H} = \mathcal{H}_{k=0} \otimes \mathcal{H}_{k=\pm 2\pi/3}$. $\mathcal{H}_{k=0}$ and $\mathcal{H}_{k=\pm 2\pi/3}$ correspond to $A$ and $E_g$ representation, respectively. The interaction Hamiltonian $\mathcal{H}_{k=\pm 2\pi/3}$ can result in a pair of adiabatic energy surfaces, which together with the harmonic term ($\propto Q_1^2 + Q_2^2$), forms the Mexican hat potential. In fact, by applying the unitary transformation:

$$
U = -\frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i \\ -1 & i \end{pmatrix},
$$

we obtain

$$
\tilde{\mathcal{H}}_{k=\pm 2\pi/3} = U^{-1} \mathcal{H} U
$$

$$
= \frac{3}{4} \hbar^2 J I - \frac{3\alpha}{4} \hbar^2 J \left( Q_1 + Q_2 \right) + Q_2 - Q_1).
$$

This expression accords with the electron-lattice interaction part of the vibronic Hamiltonian for the Jahn-Teller system $E_g \otimes e_g$,

$$
\mathcal{H}_{JT} = \frac{1}{2} \omega^2 (Q_1^2 + Q_2^2) + \alpha \left( \frac{Q_1}{Q_2} + \frac{Q_2}{-Q_1} \right).
$$

Thus, we would like to emphasize that the present system for quantum spins on the triangle coupled with doubly-degenerate vibrational $e_g$ modes is equivalent to $E_g \otimes e_g$ vibronic system intensively investigated in the context of the dynamical Jahn-Teller problem.

Before proceeding to the argument on the chiral order parameter of the spin system, we shall recall the definition of the electronic orbital angular momentum in the dynamical Jahn-Teller system. For the quantal Hamilto-
where the \( \psi_{n,m} \)'s are the eigenfunctions of the isotropic two-dimensional harmonic oscillator (\( n \) and \( m \) are radial and azimuthal quantum numbers, respectively), and \( \phi_+ \) and \( \phi_- \) are degenerate electronic states. The expansion (9) was found by rewriting \( H_{JT} \) of (8) into a suitable form with the use of \( \phi_\pm \) [9]. In the context of the spin-lattice system under consideration, the block matrix \( H_{k=\pm2\pi/3} \) in (5) already takes such a suitable form with the use of Nakamura-Bishop's bases \( |k=\pm2\pi/3\rangle \), and the whole wave function takes the same form as (9).

In the vibronic state \( \Psi_{p,1/2} \) in the dynamical Jahn-Teller system, the expectation value of the electronic orbital angular momentum \( \ell_z \) is given as

\[
\langle \ell_z \rangle_p = \langle \Psi_{p,1/2} | \ell_z | \Psi_{p,1/2} \rangle = \sum_{n=1}^{\infty} |a_{n,p}|^2 (-1)^{n-1/2} \Xi_{\ell} \quad (p = 1, 2, \ldots) \tag{10}
\]

Here, \( \Xi_{\ell} \) is the expectation value of \( \ell_z \) in the electronic states \( \phi_+ \) and \( \phi_- \):

\[
\Xi_{\ell} = \langle \phi_+ | \ell_z | \phi_+ \rangle = \langle \phi_- | \ell_z | \phi_- \rangle.
\]

The emergence of an outstanding regular oscillation of \( \langle \ell_z \rangle_p \) as a function of energy(\( p \)) was pointed out three decades ago [10], and has received a renewed attention recently in the context of nonlinear dynamics [9].

Now let's come back to the argument of the characteristic operator for the frustrated quantum spin system. With use of the bases (4), we evaluate the expectation values for chiral order parameter

\[
\hat{\chi} = \mathbf{S}_1 \cdot (\mathbf{S}_2 \times \mathbf{S}_3).
\]

which characterizes the degree of frustration of the triangular antiferromagnet [4]. The expectation values of \( \hat{\chi} \) in each of \( k = \pm2\pi/3 \) states (4) are

\[
\langle k | 2\pi/3 \hat{\chi} | k = 2\pi/3 \rangle = -\sqrt{3}/4 \equiv \Xi_{\chi}
\]

\[
\langle k = -2\pi/3 | 2\pi/3 \hat{\chi} | k = -2\pi/3 \rangle = \sqrt{3}/4 \equiv \Xi_{\chi}.
\]

(The value \( k = 0 | \hat{\chi} | k = 0 \) = 0 is now irrelevant since \( k = 0 \) is coupled only with the higher frequency \( \omega_{1g} \) mode.) Thus, the states \( |k = \pm2\pi/3 \rangle \) and chiral order parameter \( \hat{\chi} \) in the spin-lattice system correspond to states |\( \phi_k \rangle \) and \( \ell_z \) in the dynamical Jahn-Teller system, respectively. Taking the eigenstates similar to (9), the value \( \langle \hat{\chi} \rangle_p \) in the \( p \)-th eigenstate is given by

\[
\langle \hat{\chi} \rangle_p = \sum_{n=1}^{\infty} |a_{n,p}|^2 (-1)^n \Xi_{\chi}.
\]

This means that the behavior of \( \langle \hat{\chi} \rangle_p \) can be revealed by applying the analysis of \( \langle \ell_z \rangle_p \) in (10). In fact, the expectation value \( \langle \hat{\chi} \rangle_p \) in (14) shows regular oscillation with increasing the energy(see Fig.3(a)), just as in the case of \( \langle \ell_z \rangle_p \) in the dynamical Jahn-Teller system [10]. This is a consequence of the integrable system which includes no anharmonic term. If chiral order parameter of one triangular lattice is observed, we can propose chiral order parameter as a new precursor of quantum chaos.

Finally we note a role of the anharmonic term involved in the triangular three particle system. Let us introduce Toda-lattice potential [11]

\[
U(x) = \frac{c}{d} e^{-dx} + \frac{c}{d} x,
\]

where \( x \) is the deviation of inter-particle distance from the equilibrium lattice constant. \( c \) and \( d \) are constant with the condition \( cd > 0 \). The total lattice potential is a sum of \( U(x) \) with \( x \) the three kind of deviations for three segments of the regular triangle. In the limit \( d \ll 1 \) under the constraint \( cd = \) constant, we obtain the following expansion in \( x \):

\[
U(x) = \frac{c}{d} (1 - dx + \frac{d^2}{2!} x^2 - \frac{d^3}{3!} x^3 + \ldots) + \frac{cd}{2} x^2 - \frac{cd^2}{6} x^3 + \ldots
\]

Suppressing a high-frequency \( a_{1g} \) mode and noting the symmetry of the \( e_g \) modes in Fig.2, the bilinear term in (16) leads to the 2-d harmonic oscillator potential. On the other hand, the cubic term in (16) leads to the trigonal(anharmonic) potential

\[
V_A = V_A(Q_1, Q_2) = \frac{\gamma}{3} (Q_1^3 - 3Q_1Q_2^2)
\]

with \( \gamma = cd^2/2 \) in terms of normal \( e_g \) modes \( Q_1 \) and \( Q_2 \). Equation (17) is just the Hénon-Heiles potential [17] and the resultant semiclassical dynamics (quantal spin + classical lattice vibrations) can show a chaotic behavior. Then, in the fully quantized system \( \langle \hat{\chi} \rangle \) has the largest
value at low energies and shows a rapidly decaying irregular oscillation with respect to energy by increasing the anharmonicity (chaoticity) \( \gamma \) (see Figs.3(b) and 3(c)).

In real triangular antiferromagnets like NaTiO\(_2\) or LiNiO\(_2\), the ground-state degeneracy due to the intrinsic frustration is serious. To remove such degeneracy, quantum spins are expected to be coupled with lattice vibrations. These extended lattices should correspond to the cooperative Jahn-Teller system where individual Jahn-Teller clusters are mutually correlated. As shown in Fig.3, in the case of coupling with lattice-vibrational modes a chiral order parameter for a three-spins cluster takes the largest value in low energies. Therefore this novel order parameter will keep to play a vital role in quantifying the ground-state frustration in extended triangular lattices coupled with harmonic or anharmonic phonons.

In conclusion the frustrated quantum spin system on a triangle coupled with lattice vibrations is equivalent to \( E_g \otimes e_g \) Jahn-Teller system. The chiral order parameter \( \chi \) should signify a quantum chaos (or quantum irregularity) induced by the interaction between quantum spins and anharmonic (or harmonic) lattice vibrations, and the energy dependence of \( \langle \chi \rangle \) that quantifies the spin frustration shows the transition from regular to irregular oscillations by increasing anharmonicity. We hope the present work will stimulate further experimental activities on the chiral order in frustrated triangular antiferromagnets.

This talk is based on the joint work with Y. Natsume, A. Terai and K. Nakamura[13]

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