The quantum spin quadrumer

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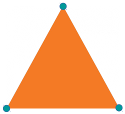
Introduction

A fundamental motif in frustrated magnetism: the ‘fully coupled’ cluster of N spins with every spin coupled to every other spin

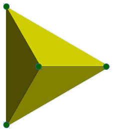
$$H_N = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j \sim J(\sum_{i=1}^{N} \mathbf{S}_i)^2$$

Natural molecular geometries:

N=2
dimer

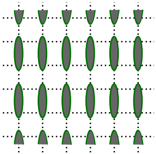


N=3
trimer
(triangle)

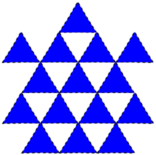


N=4
quadrumer
(tetrahedron)

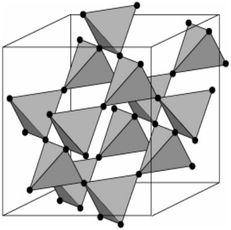
- $N = 2 \Rightarrow$ dimerized systems, square antiferromagnet



- $N = 3 \Rightarrow$ Triangular and Kagome antiferromagnets



- $N = 4 \implies$ pyrochlore, checkerboard and square $J_1 - J_2 - J_3$ model



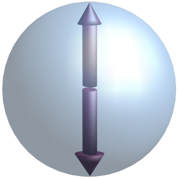
- $N = 2, 3$ are well studied. 'Semiclassical' descriptions exist and have proved fruitful.
- Result: a semiclassical theory for a single N=4 cluster
  - Non-trivial extension from $N = 2, 3$
  - Example of dynamics on a 'non-manifold'
  - Elegant physical description in terms of emergent fields
  - Starting point for semiclassical field theories for pyrochlore and other systems

Review of $N = 2$ case

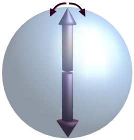
Cluster Hamiltonian: $H_2 = J(\mathbf{S}_1 + \mathbf{S}_2)^2$.

- Total configuration space is $S^2 \otimes S^2$
- Classical ground state condition: $(\mathbf{S}_1 + \mathbf{S}_2) = 0$.
- Degrees of freedom of the system: 4
- Number of constraints in ground state*: 2
- Number of independent parameters needed to specify a ground state: 2 $\Rightarrow$ two-dimensional ground state space

- Classical ground states: spins pointing towards antipodal points

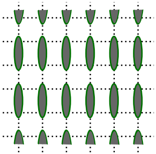


- 'Classical ground state space': S^2, specifies the first spin
- Such states are *manifold*: 2D tangent space at every point, i.e., every ground state has two 'soft fluctuations'



- Low energy dynamics of cluster:
    - described by a unit vector order parameter
    - maps to particle constrained to move on the surface of a sphere $\rightarrow$ describes semiclassical and quantum limits

Example: low energy effective theory for a square antiferromagnet[1]



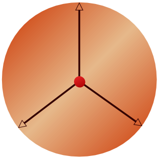
$$\mathcal{L} = \frac{1}{2J}\left[c(\nabla\hat{n}(\mathbf{x}))^2 + c^{-1}(\partial_\tau\hat{n}(\mathbf{x}))^2\right]$$

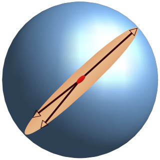
[1] Haldane PRL 1983, T. Dombre and N. Read, PRB 1988

Review of $N = 3$

Cluster Hamiltonian: $H_3 = J(\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3)^2$

- Configuration space: $S^2 \otimes S^2 \otimes S^2$
- Classical ground state condition: $\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 = 0$
- Degrees of freedom: 6
- Number of constraints 3
- Number of independent parameters needed to specify a ground state: 3
- Ground states: Planar 120° states, all such states can be achieved by a global rotation on a reference ground state[1]



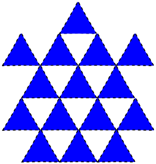


[1] H. Kawamura and S. Miyashita, JPSJ 1984

- Ground state space: $SO(3)$
- Manifold: 3D tangent space $\implies$ each ground state has three soft fluctuations (rotations about three fixed axes)
- Low energy dynamics of cluster:
  - described by a $SO(3)$ matrix order parameter
  - maps to a rigid body $\rightarrow$ describes semiclassical and quantum limits

Example: low energy effective theory for the triangular antiferromagnet[2]

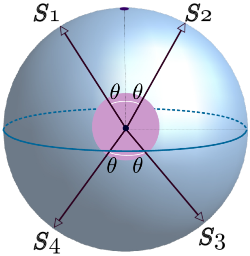
$$\mathcal{L} = -\frac{g^{-1}}{2}\Big[c^{-1}\,Tr(R^{-1}\partial_\tau R)^2 + c\ Tr\big[P\{(R^{-1}\partial_x R)^2 + (R^{-1}\partial_y R)^2\}\big]\Big]$$

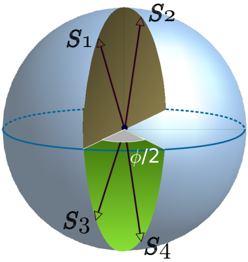


[2] T. Dombre and N. Read, Phys.Rev. B 1989

Cluster Hamiltonian : $H_4 = J(\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4)^2$

- Configuration space: $S^2 \otimes S^2 \otimes S^2 \otimes S^2$, classical ground state space: $\{S^2 \otimes S^2 \otimes S^2 \otimes S^2 | \sum_{i=1}^4 \mathbf{S}_i = 0\}$.
- Degrees of freedom: 8
- Number of constraints in a ground state: 3
- Naive expectation: ground state space is a 5D manifold!
- Generic ground state described by 5 parameters
  - Two parameters θ and ϕ to fix relative angles
  - An $SO(3)$ matrix (with 3 parameters) for an overall rotation





$$\hat{n}_1 = \sin\theta\left(\cos\frac{\phi}{4}\,\hat{x} + \sin\frac{\phi}{4}\,\hat{y}\right) + \cos\theta\,\hat{z}$$

$$\hat{n}_2 = \sin\theta\left(-\cos\frac{\phi}{4}\,\hat{x} - \sin\frac{\phi}{4}\,\hat{y}\right) + \cos\theta\,\hat{z}$$

$$\hat{n}_3 = \sin\theta\left(-\cos\frac{\phi}{4}\,\hat{x} + \sin\frac{\phi}{4}\,\hat{y}\right) - \cos\theta\,\hat{z}$$

$$\hat{n}_4 = \sin\theta\left(\cos\frac{\phi}{4}\,\hat{x} - \sin\frac{\phi}{4}\,\hat{y}\right) - \cos\theta\,\hat{z}$$

Here $\theta \in [0, \pi]$ and $\phi \in [0, 2\pi]$

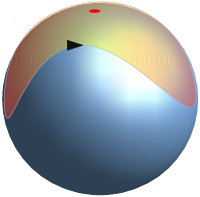
- (θ, ϕ) fix the relative angles between spins
- Their allowed ranges* resemble that of a unit vector $\implies$ 'emergent' unit vector order parameter
- Naive expectation: ground state space is $SO(3) \otimes S^2$.
- Large-S path integral description of the quadrumer $\Rightarrow$ starting point for constructing field theories for systems containing $N = 4$ cluster as motif

Path integral for a spin system

- Partition function: $\mathcal{Z} = \int_{\Omega_j(0)=\Omega_j(\beta)} \prod_j \mathcal{D}\Omega_j(\tau) e^{-\mathcal{S}}$

$$\mathcal{S} = \int_0^\beta d\tau \Big[\underbrace{iS \sum_j \vec{A}(\hat{\Omega}_j) \cdot \partial_\tau \hat{\Omega}_j}_{\text{Berry phase}} + \underbrace{H(S\hat{\Omega}_1, S\hat{\Omega}_2, S\hat{\Omega}_3 \cdots)}_{\text{energy}} \Big]$$

- Here $\vec{A}$ is defined by: $\vec{\nabla} \times \vec{A}(\Omega_i) = \hat{\Omega}_i$
- Berry phase is a geometric quantity given by iS times the area covered by $\hat{\Omega}(\tau)$ on the surface of the unit sphere



[3] e.g., A. Auerbach, Interacting electrons and quantum magnetism

Semiclassical action for quadrumer

- Parametrization: $S_j = S\hat{\Omega}_j \approx SR(\hat{n}_j + M_j \frac{\vec{L}}{S})$
  where $M_j^{\alpha\beta} = \delta^{\alpha\beta} - n_j^{\alpha} n_j^{\beta}$
- $\vec{L}$ is a vector with three independent components which induces magnetization: increases energy and lifts the system out of the ground state space
- Degrees of freedom counting (8 in total): R has three, $\hat{n}_j$'s have two (θ and ϕ) and $\vec{L}$ has three degrees of freedom
- Semi classical low energy limit: $S \gg 1$, $\vec{L} \sim \mathcal{O}(1)$; system remains in a very low energy region
- All spins are normalized to S to $\mathcal{O}(S^0)$
- Magnetization: $\sum_{j=1}^{4} \vec{S}_j = R\left(M\vec{L}\right)$, where $M = \sum_j M_j$
- Magnetization, being the sum of spins, is an angular momentum variable

Berry phase

The Berry phase: $S_B = \int_0^\beta d\tau \, i \sum_j \vec{A}(\hat{\Omega}_j) \cdot \partial_\tau \vec{S}_j$ can be shown to be

$$\int_0^\beta d\tau \left[iS \sum_{j=1}^{4} \vec{A}(R\hat{n}_j) \cdot \partial_\tau (R\hat{n}_j) + 4i\vec{L} \cdot \vec{U} - i\vec{V} \cdot RM\vec{L} \right]$$

where $\vec{U} = \frac{1}{4} \sum_i \partial_\tau \hat{n}_i \times \hat{n}_i$ and $V_\beta = -\frac{1}{2} \epsilon_{\beta\sigma'\delta} \{(\partial_\tau R) R^{-1}\}^{\sigma'\delta}$

- The first term in S_B, remarkably, simplifies to $\int_0^\beta d\tau \left(-iS \cos\theta \dot{\phi} \right)$, due to $\sum_j \hat{n}_j = 0$
- U is, in fact, zero. This simplification arises due to the symmetric parametrization of $\hat{n}_i$'s in terms of (θ, ϕ)
- $\vec{V}$ has the form of the angular velocity of a rigid body
- Finally, $S_B = \int_0^\beta d\tau \left(-iS \cos\theta \dot{\phi} - iRM\vec{L} \cdot \vec{V} \right)$

Energy

The cluster Hamiltonian is
$$H_4 = J(\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4)^2$$
$$= J(M\vec{L})^2$$

(θ, ϕ) appears in the Hamiltonian as a part of the matrix M

Partition function in terms of new variables

$$\mathcal{Z} = \int \left(\prod_{\tau} J(\theta_\tau, \phi_\tau, \alpha_\tau, \beta_\tau, \gamma_\tau, \vec{L}_\tau)\, d\theta_\tau\, d\phi_\tau\, d\alpha_\tau\, d\beta_\tau\, d\gamma_\tau\, d\vec{L}_\tau \right) e^{-\mathcal{S}},$$

Where $\mathcal{S} = \int_0^\beta d\tau \left(\underbrace{-iS\cos\theta\dot{\phi} - iRM\vec{L}\cdot\vec{V}}_{\text{Berry phase}} + \underbrace{J(M\vec{L})^2}_{\text{energy}} \right)$

- τ refers to imaginary time slices
- (α, β, γ) parametrizes the rotation matrix: (α, β) specify an axis while γ specifies the angle of rotation
- $J(\theta_\tau, \phi_\tau, \alpha_\tau, \beta_\tau, \gamma_\tau, \vec{L}_\tau)$, Jacobian of the transformation, turns out to be proportional to $\frac{1}{4\pi^2}\sin^2(\frac{\gamma_\tau}{2})\sin\alpha_\tau\, Det(M)\sin\theta_\tau$ to $\mathcal{O}(S^0)$.

The path integral measure

- The measure comes out to be
  $\frac{1}{4\pi^2}\left\{\sin^2(\frac{\gamma_\tau}{2})\sin\alpha_\tau\, d\alpha_\tau\, d\beta_\tau\, d\gamma_\tau\right\}\{\sin\theta_\tau\, d\theta_\tau\, d\phi_\tau\}\left\{Det(M)\, d\vec{L}_\tau\right\}$
  - Emergent unit vector measure $d\hat{\Omega}_\tau = \sin\theta_\tau\, d\theta_\tau\, d\phi_\tau$, an area element on the surface of a unit sphere
  - $SO(3)$ group invariant measure: $dR_\tau := \frac{1}{4\pi^2}\sin^2(\frac{\gamma_\tau}{2})\sin\alpha_\tau\, d\alpha_\tau\, d\beta_\tau\, d\gamma_\tau$, volume element in $SO(3)$ space
- Redefine $\vec{L}$: $\vec{L'}_\tau = R_\tau M_\tau \vec{L}_\tau$
  - Measure: $d\vec{L'}_\tau = Det(R_\tau M_\tau)\, d\vec{L}_\tau$
    $= Det(M_\tau)\, d\vec{L}_\tau$
- We can denote the measure as $dR_\tau\, d\hat{\Omega}_\tau\, d\vec{L'}_\tau$
- Partition function:
  $\mathcal{Z} = \int \prod_\tau dR_\tau\, d\hat{\Omega}_\tau\, d\vec{L'}_\tau\, e^{-\mathcal{S}} = \int \mathcal{D}\hat{\Omega}(\theta,\phi)\,\mathcal{D}\vec{L'}\,\mathcal{D}R\, e^{-\mathcal{S}}$

  where $\mathcal{S} = \int_0^\beta d\tau\,\left(-iS\cos\theta\dot{\phi} - i\vec{L'}\cdot\vec{V} + JL'^2\right)$.

The partition function:

$$\mathcal{Z} = \left(\int \mathcal{D}\hat{\Omega}(\theta,\phi)\, e^{\int_0^\beta d\tau\, iS\cos\theta\dot{\phi}} \right) \times \left(\int \mathcal{D}\vec{L}'\, \mathcal{D}R\, e^{-\int_0^\beta d\tau \left(-i\vec{L}'\cdot\vec{V} + JL'^2\right)} \right)$$
$$= \mathcal{Z}_1 \times \mathcal{Z}_2.$$

- $\mathcal{Z}_1$ is the partition function for an 'emergent' free spin-S spin
- $\mathcal{Z}_2$ is the partition function for a spherical top rigid rotor of moment of inertia $\frac{1}{2J}$
  - $\vec{L}'$ (net mangetization) is angular momentum
  - R is angular 'position'
- Result: The quadrumer, in semi-classical low-energy limit, decouples into an emergent free spin-S spin and a spherical top[4] rigid rotor!

[4] Spherical top: rigid body with three principal moments of inertia equal

Comparison with a conventional quantum analysis

Hamiltonian: $H_4 = J(S_1 + S_2 + S_3 + S_4)^2$

- Energy eigenvalues: $Jj(j+1)\hbar^2$, with total spin quantum number $j = 0, 1, \cdots, 4S$
- Semi-classical approach $\implies$ A free spin and a rigid rotor
  From the form of the path integral, we infer the Hamiltonian

$$H = H_{\text{free spin}} + H_{\text{rigid rotor}}$$

  As $H_{\text{free spin}}$ is zero, it does not contribute to energy, nevertheless it increases the degeneracy of each rotor state by a factor $(2S+1)$

  Rotor eigenvalues: $Jj(j+1)\hbar^2$ with $j = 0, 1, \cdots, \infty$

  $\implies$ eigenvalues match exactly (for low energies)

Degeneracy of energy levels

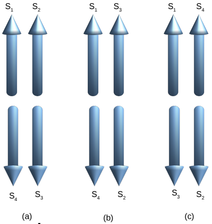
State	Energy	degeneracy: usual quantum approach	degeneracy: semi-classical approach
Ground state	0	$(2S+1)$	$(2S+1)$
First excited	$2J\hbar^2$	$3^2(2S+1)-9$	$3^2(2S+1)$
Second excited	$6J\hbar^2$	$5^2(2S+1)-45$	$5^2(2S+1)$
$\vdots$	$\vdots$	$\vdots$	$\vdots$

Comparing the degeneracy, both approaches agree to $\mathcal{O}(S)$!
However, there is a small $\mathcal{O}(S^0)$ discrepancy in the degeneracy

What causes this discrepancy?

Non-manifold character of the ground space

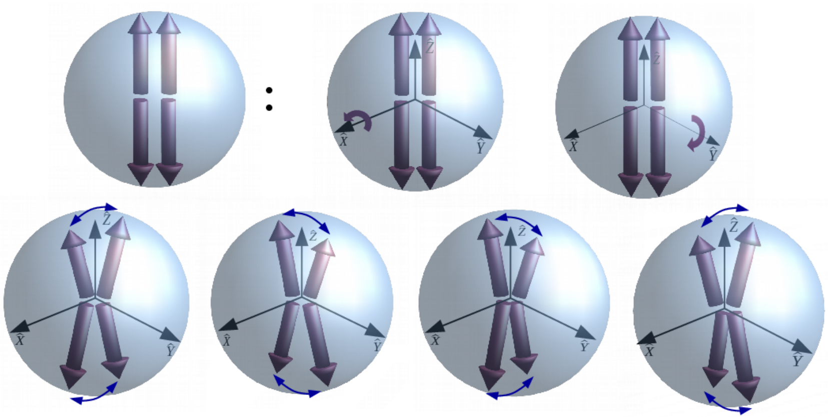
- Mapping into free spin and rigid rotor fails when the matrix M becomes singular
- $\vec{L'}_\tau = R_\tau M_\tau \vec{L}_\tau$: if $det(M_\tau) = 0$, we don't have three independent components of angular momentum $\vec{L'} \implies$ not a rigid rotor
- This occurs whenever the ground state is collinear! There are precisely three collinear states (upto global rotations)



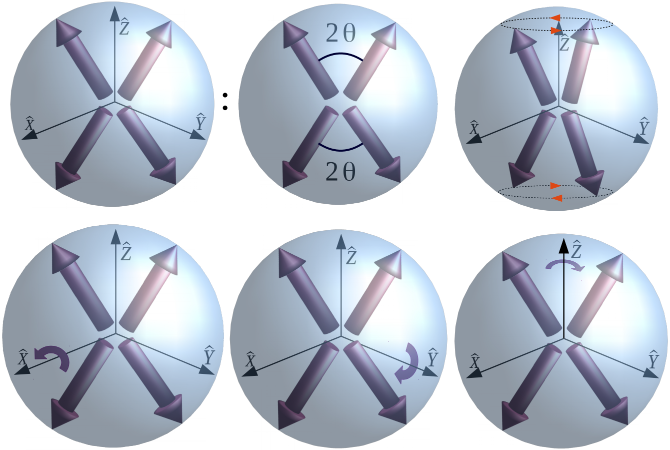
- This can be traced back to our parametrization; one component of $\vec{L}$ becomes redundant at a collinear ground state

Soft fluctuations about collinear state

- Is the ground state space just $SO(3) \otimes S^2$ or something more?
- To understand the nature of the ground state space, we look at 'soft' fluctuations about different ground states



Soft fluctuations around a coplanar state



- About collinear states, we have 6 soft fluctuations
- About any non-collinear state, we only have 5
- If we exclude collinear states, the space is a 5D manifold (by implicit function theorem)
- A crude illustration:

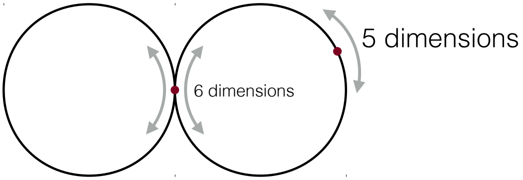


Figure: 'figure of 8'

- Upon including collinear states, it is no more a 5D manifold
- Collinear states form a subset of measure zero (3 states modulo rotations) $\implies$ neglecting collinear states is a 'good' approximation $\implies$ mapping of quadrumer into a free spin and a rigid rotor successfully describes the system

Thermodynamics

- From Semi-classical picture, the partition function is $\mathcal{Z} = \sum_j (2S+1)(2j+1)^2 e^{-j(j+1)\beta g \hbar^2}$
  - Factor of $(2S+1)$ comes from free spin
- At $T = 0$, entropy is $k_B \log(2S+1)$, determined by the ground state degeneracy.
- Low temperature expansion gives
  - Entropy: S = $k_B \left[\log(2S+1) + \log(1 + 9e^{-2\beta g \hbar^2}) + \frac{18 g\beta\hbar^2}{e^{2g\beta\hbar^2}+9} + \cdots \right]$.
  - Specific heat: $C_v = 36 k_B (g\beta\hbar^2) \frac{e^{2\beta g \hbar^2}}{(9+e^{2\beta g\hbar^2})^2}$.

In presence of small magnetic field

- Application of a small magnetic field changes the Hamiltonian by $-B\hat{L'}_z$, where L'_z is z component of the magnetization.
- Partition function changes to $\mathcal{Z} = \sum_j (2S+1)(2j+1)e^{-j(j+1)\beta g\hbar^2} \left(\sum_{m=-j}^{j} e^{m\beta B\hbar}\right)$, where the eigenvalues of L'_z is $m\hbar$.
- Low temperature susceptibility:

  $\chi = \frac{1}{\beta}\partial_B^2 log\,\mathcal{Z}|_{B\to 0} = \frac{6\beta\hbar^2}{e^{2\beta g\hbar^2}+9}$.

Experimentally realized materials

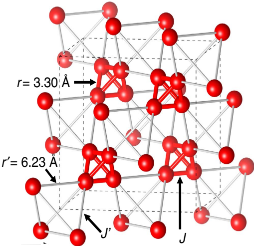


Figure: $Ba_3Yb_2Zn_5O_{11}$: red balls are Ybs with pseudospin-1/2 which form near-isolated tetrahedra. Park et al, Nat. Comm. 2016

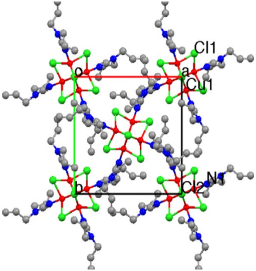


Figure: $Cu_4OCl_6daca_4$: magnetic building block is a tetrahedron of 4 Cu atoms (red balls) with spin-1/2. Zaharko et al , PRB 2008

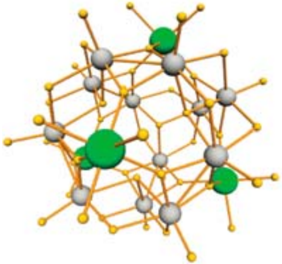


Figure: Ni_4Mo_{12}: tetrahedron formed by Ni atoms (green balls) with spin-1. J. Nehrkorn et al, EPJB 2010

- Deviations from Heisenberg model: DM interactions, phonon modes, Ising anisotropy, etc. – may couple the spin and the rotor fields

Future directions

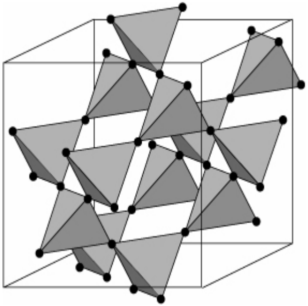


Figure: Pyrochlore lattice, e.g., $Mn_2Sb_2O_7$.

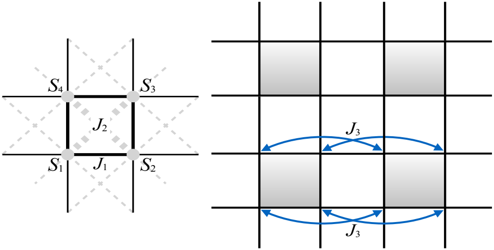


Figure: Square $J_1 - J_2 - J_3$ model, Danu et al, PRB 2016.

Several insights about these models can emerge from their low energy field theories. Our single cluster semiclassical description serves as a starting point.

Thank You

Our state space

$$S \otimes S \otimes S \otimes S = (0 \oplus 1 \oplus 2 \oplus 3 \oplus \cdots \oplus 2S) \otimes (0 \oplus 1 \oplus 2 \oplus 3 \oplus \cdots \oplus 2S).$$

0 comes from $(0 \otimes 0), (1 \otimes 1), (2 \otimes 2), \cdots (2S \otimes 2S)$, occurs $2S + 1$ times.

1 comes from

$$(0 \otimes 1)$$
$$(1 \otimes 0),(1 \otimes 1) \text{ and } (1 \otimes 2)$$
$$(2 \otimes 1),(2 \otimes 2) \text{ and } (2 \otimes 3)$$
$$(3 \otimes 2),(3 \otimes 3) \text{ and } (3 \otimes 4)$$
$$\vdots$$
$$(2S - 1 \otimes 2S - 2),(2S - 1 \otimes 2S - 1) \text{ and } (2S - 1 \otimes 2S)$$
$$(2S \otimes 2S - 1) \text{ and } (2S \otimes 2S).$$

1 occurs $3(2S - 1) + 3 = 3(2S + 1) - 3$. The degeneracy is $3^2(2S + 1) - 9$.

Spin wave analysis around Néel state

Consider $R = e^{i\pi_i T_i}$ consider Néel order along X axis, After doing spin wave analysis around this state we would get the action

$$\mathcal{S} = \int_0^\beta d\tau \left(iS\delta\theta\delta\dot{\phi} + 4L_y \cdot \dot{\pi}_y + 4L_z \cdot \dot{\pi}_z + 16g(L_y^2 + L_z^2) \right)$$