The quantum spin quadrumer: In how many ways can four vectors add to zero?

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Un éléphant qui se balançait
Sur une toile, toile, toile… toile d'araignée
Et qui trouvait ce jeu tellement amusant
Que bientôt vint un deuxième éléphant

Deux éléphants qui se balançaient
Sur une toile, toile, toile… toile d'araignée
Et qui trouvaient ce jeu tellement amusant
Que bientôt vint un troisième éléphant

Trois éléphants qui se balançaient
Sur une toile, toile, toile… toile d'araignée
Et qui trouvaient ce jeu tellement amusant
Que bientôt vint un quatrième éléphant

Quatre éléphants qui se balançaient
Sur une toile, toile, toile… toile d'araignée
Et qui trouvaient ce jeu tellement amusant
Que tout à coup…

Boum, Badaboum !

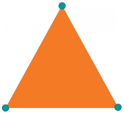
Introduction

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

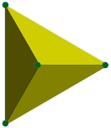
$$H_N = J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j \sim J(\sum_{i=1}^{N} \mathbf{S}_i)^2$$

Natural molecular geometries:

N=2
dimer

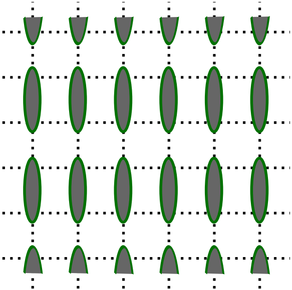


N=3
trimer
(triangle)

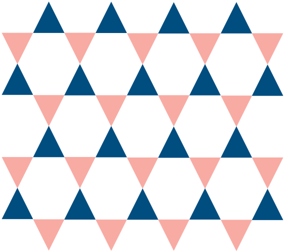


N=4
quadrumer
(tetrahedron)

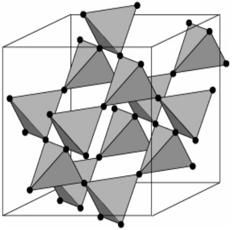
- $N = 2 \Rightarrow$ dimerized systems, square antiferromagnet



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



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



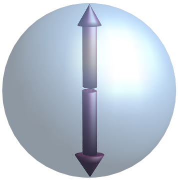
- $N = 2, 3$ are well studied. 'Semiclassical' descriptions exist and have proved fruitful.
- Result: a semiclassical theory for a single N=4 cluster
  - Non-trivial extension from $N = 2, 3$
  - Example of dynamics on a 'non-manifold'
  - Elegant physical description in terms of emergent fields
  -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

Review of $N = 2$ case

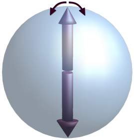
Cluster Hamiltonian: $H_2 = J(\mathbf{S}_1 + \mathbf{S}_2)^2$.

- 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

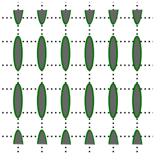


- 'Classical ground state space': S^2, specifies the first spin
- Such states are *manifold*: 2D tangent space at every point, i.e., every ground state has two 'soft fluctuations'



- 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]

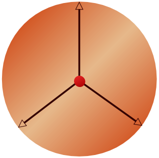


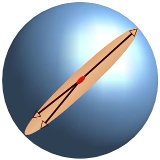
$$\mathcal{L} = \frac{1}{2J}\left[c(\nabla\hat{n}(\mathbf{x}))^2 + c^{-1}(\partial_\tau\hat{n}(\mathbf{x}))^2\right]$$

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

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

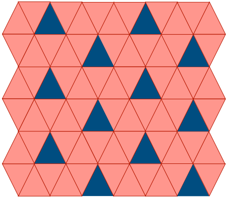




- Ground state space: $SO(3)$
- Manifold: 3D tangent space $\implies$ each ground state has three soft fluctuations (rotations about three fixed axes)
- Low energy dynamics of cluster:
  - described by a $SO(3)$ matrix order parameter
  - maps to a rigid body $\rightarrow$ describes semiclassical and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

Example: low energy effective theory for the triangular antiferromagnet

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

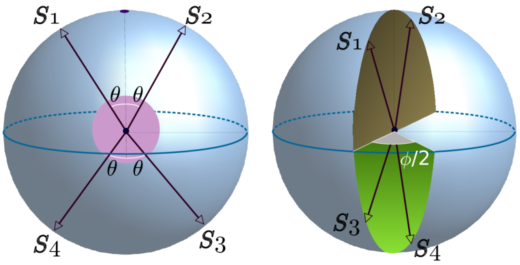


[2]T. Dombre and N. Read, Phys.Rev. B 1989

$N = 4$ cluster

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



$$
\begin{aligned}
\hat{n}_1 &= \sin\theta \left(\cos\frac{\phi}{4}\,\hat{x} + \sin\frac{\phi}{4}\,\hat{y} \right) + \cos\theta\,\hat{z} \\
\hat{n}_2 &= \sin\theta \left(-\cos\frac{\phi}{4}\,\hat{x} - \sin\frac{\phi}{4}\,\hat{y} \right) + \cos\theta\,\hat{z} \\
\hat{n}_3 &= \sin\theta \left(-\cos\frac{\phi}{4}\,\hat{x} + \sin\frac{\phi}{4}\,\hat{y} \right) - \cos\theta\,\hat{z} \\
\hat{n}_4 &= \sin\theta \left(\cos\frac{\phi}{4}\,\hat{x} - \sin\frac{\phi}{4}\,\hat{y} \right) - \cos\theta\,\hat{z}
\end{aligned}
$$

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

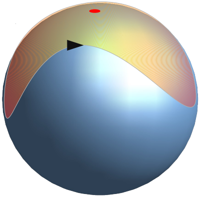
- (θ, ϕ) fix the relative angles between spins
- Their allowed ranges* resemble that of a unit vector $\implies$ 'emergent' unit vector order parameter
- Naive expectation: ground state space is $SO(3) \otimes S^2$.
- Large-S path integral description of the quadrumer $\Rightarrow$ starting point for 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, $S_B = \int_0^\beta d\tau \left(-iS \cos\theta \dot{\phi} - iRM\vec{L} \cdot \vec{V} \right)$

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

(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

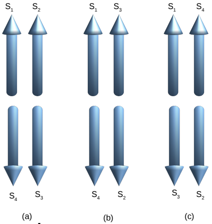
State	Energy	degeneracy: usual quantum approach	degeneracy: semi-classical approach
Ground state	0	$(2S+1)$	$(2S+1)$
First excited	$2J\hbar^2$	$3^2(2S+1)-9$	$3^2(2S+1)$
Second excited	$6J\hbar^2$	$5^2(2S+1)-45$	$5^2(2S+1)$
$\vdots$	$\vdots$	$\vdots$	$\vdots$

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

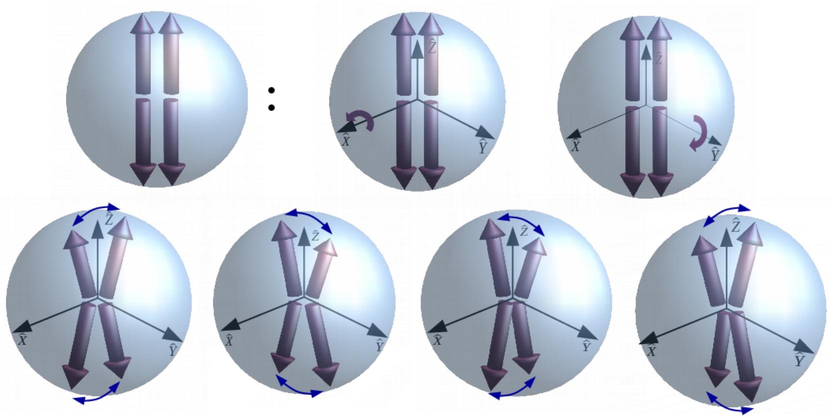
- Mapping into free spin and rigid rotor fails when the matrix M becomes singular
- $\vec{L'}_\tau = R_\tau M_\tau \vec{L}_\tau$: if $det(M_\tau) = 0$, we don't have three independent components of angular momentum $\vec{L'} \implies$ not a rigid rotor
- This occurs whenever the ground state is collinear!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



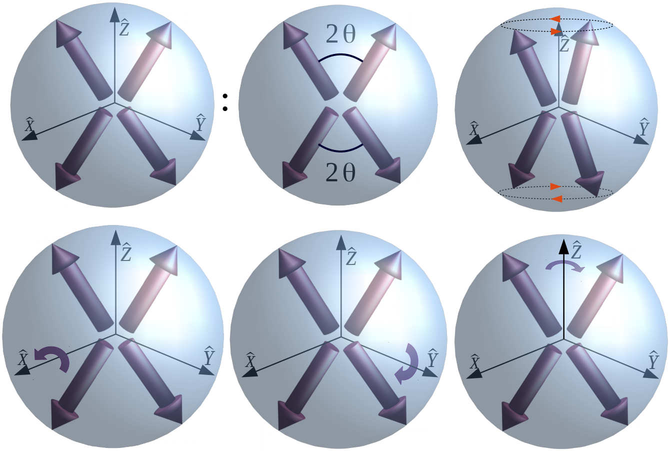
- This can be traced back to our parametrization; one component of $\vec{L}$ becomes redundant at a collinear ground state

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:

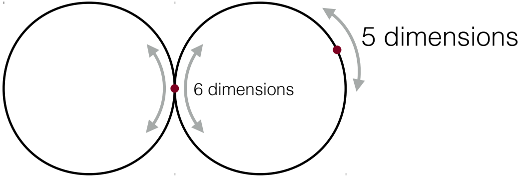


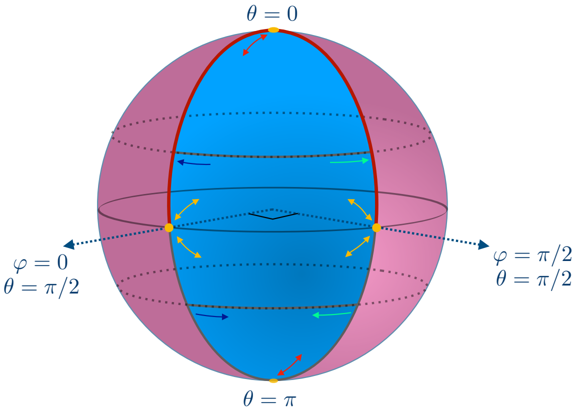
Figure: 'figure of 8'

- 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

Orbifold-like structure of ground state space

Two identification rules that will help us achieve this:

- $(\theta, \varphi + \pi) \equiv (\theta, \varphi)$ under R_z^π
- $(\pi - \theta, \pi - \varphi) \equiv (\theta, \varphi)$ under R_y^π



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

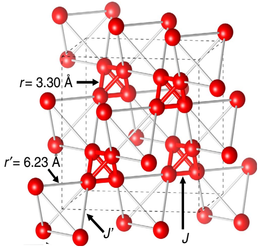


Figure: $Ba_3Yb_2Zn_5O_{11}$: red balls are Ybs with pseudospin-1/2 which form near-isolated tetrahedra. Park et al, Nat. Comm. 2016

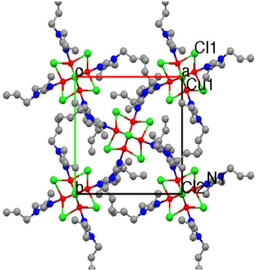


Figure: $Cu_4OCl_6daca_4$: magnetic building block is a tetrahedron of 4 Cu atoms (red balls) with spin-1/2. Zaharko et al , PRB 2008

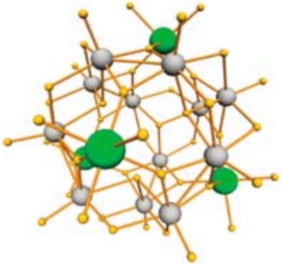


Figure: Ni_4Mo_{12}: tetrahedron formed by Ni atoms (green balls) with spin-1. J. Nehrkorn et al, EPJB 2010

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

Future directions

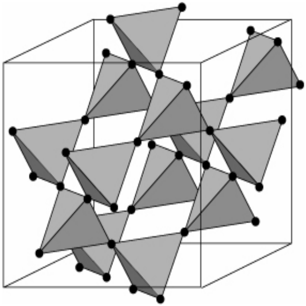


Figure: Pyrochlore lattice, e.g., $Mn_2Sb_2O_7$.

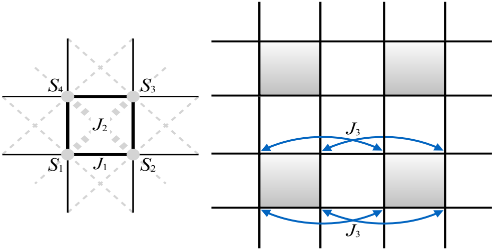


Figure: Square $J_1 - J_2 - J_3$ model, Danu et al, PRB 2016.

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