Abstract: Numerical Linked Cluster (NLC) expansions can accurately compute thermal properties of quantum spin models in the thermodynamic limit in certain parameter regimes. In classical spin-ice models, where all correlations remain short-ranged down to $T=0$, these expansions can be convergent even at low $T$. However, for quantum spin-ice models, they converge only when either temperatures are not too small or there is a strong magnetic field present. To turn these studies into a spectroscopy of exchange parameters, when multiple exchange constants are relevant, is a challenge both because of the limited temperature-range of validity of effective spin-half models and difficulties in isolating magnetic properties in experiments at intermediate and high temperatures. We discuss ways in which such a spectroscopy can proceed.
Numerical Linked Cluster Spectroscopy of Quantum Spin Ice?

Rajiv Singh UC Davis
Michel Gingras Waterloo

Vindication of Yb$_2$Ti$_2$O$_7$ as a Model Exchange Quantum Spin Ice

Thermodynamic properties of Yb$_2$Ti$_2$O$_7$ pyrochlore as a function of temperature and magnetic field: Validation of a quantum spin ice exchange Hamiltonian

Are Multiphase Competition and Order by Disorder the Keys to Understanding Yb$_2$Ti$_2$O$_7$?
Summary of experiments on Quantum Spin Ice

Rare-earth pyrochlores provide enormous variety/flexibility, but that comes at the cost of various forms of disorder/soft modes. How does that affect properties? Can enhance quantum effects.

Effective quantum spin degrees of freedom originate from high spin + spin-orbit + crystal-field effects. That can lead to rather unusual dipolar-octupolar objects whose coupling to magnetic-fields and neutrons can be rather esoteric. Symmetry analysis is very elegant. But, often symmetries may be perturbed. What does it do?

Dipolar and exchange quantum spin-ice may have very different timescales and local spectral features. Quadrupolar couplings? Often spectral features set in at temperatures of order J (pinch points, monopoles, Dirac strings?)
Quantum Spin Ice defined by fractionalization --- most clearly electric and magnetic spinons and photons --- signatures in transport (thermal), neutrons, heat capacity—Less by a plateau in entropy. Can even coexist with magnetic order.

Rich phase diagram – proximity to many ordered phases, reentrance, fragmentation, many spin-liquids–pinch points may be there outside of classical spin-ice. Chemical pressure, disorder, pressure, magnetic fields can all cause transition.

Quantum spin-liquids not just quantum spin-ice should be the focus. Multimagrons versus spinons, can one relate the two pictures?

How well do we know (need to know) the exchange constants?
OUTLINE

- **Linked Cluster Methods (based on real-space Graphs)**
  Series Expansions versus NLCE
  Thermodynamic/Static (and Dynamic?) properties

- **Application to quantum spin-ice models:**
  from Hamiltonian to Thermodynamic Properties
  scope and limitations

- **Can it be used to determine exchange constants?**

  NLC Spectroscopy?
REAL SPACE GRAPH-BASED LINKED-CLUSTER METHODS

Extensive Property $P$: $(C, S, \chi)$ in thermodynamic limit ($N \to \infty$)

$$P(L)/N = \sum_c L(c) W(c)$$

$L(c)$: Lattice Constant
(in a translationally invariant system)

Weight $W(c)$ only depends on $P(c)$ on finite cluster and its subclusters

$$P(c) = \sum_{s \subseteq c} W(s)$$

$$W(c) = P(c) - \sum_{s \subseteq c} W(s)$$
High Temperature Series Expansions (HTE):

Extensive Property P: \( C, S, \chi \) can be expanded as

\[
P/N = a_0 + a_1\beta + a_2\beta^2 + \ldots
\]

Expand \( P(c) \) for finite clusters as

\[
\sum_n p_n \beta^n
\]

\[
W(c) = P(c) - \sum_{s \subset c} W(s)
\]

Summing over all graphs with up to \( l \) bonds

\[
\sum_c L(c) W(c)
\]

Remainder:

\[
R_l = P(\mathcal{L})/N - \sum_c L(c) W(c) - O(\beta^{l+1})
\]
High Field Series Expansions (HFE):

\[ H = H_0 + \lambda H_1 \]

\( H_0 \) Single-site Hamiltonian \( \lambda = 1/h \)

\[ \frac{P}{N} = b_0(\beta) + b_1(\beta)\lambda + b_2(\beta)\lambda^2 + \ldots \]

Once again the coefficients can be computed by a linked-cluster method:

\[ \frac{P(\mathcal{L})}{N} = \sum_c L(c) W(c) \]

Weights can be expanded in powers of \( \lambda \)
What if there is no small parameter?

Exact Diagonalization remains the mainstay of computational quantum many-body systems

Can we combine it with Linked Cluster methods to obtain answers in the thermodynamic limit?
NUMERICAL LINKED-CLUSTER (NLC) METHODS

\[ P(\mathcal{L})/N = \sum_c L(c) W(c) \]

Compute weight by exact diagonalization one \((T, h)\) at a time

\[ P(c) = \sum_{s \subseteq c} W(s) \quad \quad W(c) = P(c) - \sum_{s \subseteq c} W(s) \]

Formally in the thermodynamic limit

Remainder

\[ R_l = P(\mathcal{L})/N - \sum_{g}^{l} L(g) W(g) \]

It should converge when either \(\beta\) or \(1/h\) is small

\[ O(\beta^{l+1}, 1/h^{l+1}) \]
Will it converge beyond the radius of convergence?

In principle yes! It depends on length scales in problem.
Enumeration/organization of clusters:

In a power series expansion: **Maximize the number of coefficients**

High-T expansions: Weak Embeddings: Count by bonds

Low-T Expansions: Strong Embeddings: Count by sites

**FIG. 1:** All clusters with up to three bonds and their lattice constant for the square lattice.

**FIG. 2:** All clusters with up to four sites and their lattice constant for the square lattice.
Enumeration/organization of clusters:

NLC: Weak? Strong? Or, something else?

Maximize parameter regimes of convergence

Organize by larger units: Squares, Triangles, Tetrahedra

Kagome Lattice

<table>
<thead>
<tr>
<th>No. of sites</th>
<th>No. of topological clusters</th>
<th>$\sum L(c)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>3</td>
<td>2</td>
<td>14/3</td>
</tr>
<tr>
<td>4</td>
<td>2</td>
<td>12</td>
</tr>
<tr>
<td>5</td>
<td>4</td>
<td>33</td>
</tr>
<tr>
<td>6</td>
<td>7</td>
<td>$281/3$</td>
</tr>
<tr>
<td>7</td>
<td>12</td>
<td>272</td>
</tr>
<tr>
<td>8</td>
<td>22</td>
<td>805</td>
</tr>
<tr>
<td>9</td>
<td>45</td>
<td>2420</td>
</tr>
<tr>
<td>10</td>
<td>88</td>
<td>7358</td>
</tr>
<tr>
<td>11</td>
<td>183</td>
<td>22581</td>
</tr>
<tr>
<td>12</td>
<td>389</td>
<td>$209552/3$</td>
</tr>
<tr>
<td>13</td>
<td>842</td>
<td>217522</td>
</tr>
<tr>
<td>14</td>
<td>1855</td>
<td>681224</td>
</tr>
<tr>
<td>15</td>
<td>4162</td>
<td>2143905</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>No. of triangles</th>
<th>No. of topological clusters</th>
<th>$\sum L(c)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>2/3</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>4</td>
<td>2</td>
<td>14/3</td>
</tr>
<tr>
<td>5</td>
<td>2</td>
<td>12</td>
</tr>
<tr>
<td>6</td>
<td>5</td>
<td>94/3</td>
</tr>
<tr>
<td>7</td>
<td>7</td>
<td>250/3</td>
</tr>
<tr>
<td>8</td>
<td>15</td>
<td>225</td>
</tr>
</tbody>
</table>

Or simply by rectangular boxes!
Specific heat and susceptibility For KLHM (Heisenberg)

Organizing contributions by rectangular boxes simplifies counting and allows one to interface with DMRG/QMC and other methods by keeping number of graphs small.
Dynamical Properties?

Static structure factors OK. But dynamics?
Frequency moments can be calculated in a controlled manner
Then reconstruct the spectral functions/rates

\[ \int \frac{\chi''(\omega)}{\omega} \omega^n d\omega \]

Nuclear relaxation rates in the Herbertsmithite Kagome antiferromagnets ZnCu3(OH)6Cl2
Nicholas E. Sherman, Takashi Imai, Rajiv R. P. Singh
Pyrochlore Lattice: Tetrahedra based expansion

- $n = 1, L = 0.5$
- $n = 2, L = 1.0$
- $n = 3, L = 3.0$
- $n = 4$
  - ABA, $L = 3.0$
  - ABC, $L = 6.0$
  - Y-graph, $L = 2.0$
CLASSICAL SPIN ICE: ISING MODEL ON PYROCHLORITE LATTICE

NLC in terms of graphs of complete tetrahedra
NLC-1 is Pauling approximation

Range of convergence of NLC far beyond HTE
Quantum spin-ice Models

\[ H = \sum_{\langle ij \rangle} \left\{ J_{zz} S_i^z S_j^z - J_\pm (S_i^+ S_j^- + S_i^- S_j^+) + J_{z\pm} \left[ \gamma_{ij} S_i^+ S_j^+ + \gamma_{ij} S_i^- S_j^- \right] \right. \\
\left. + J_{z\mp} \left[ S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij} S_j^-) + i \leftrightarrow j \right] \right\}, \]

Exchange parameters: \( J_{zz}, J_\pm, J_{z\pm}, J_{z\mp} \)

\( g \)-factors: \( g_{zz}, g_{xy} \)

How well can we calculate thermodynamic properties?
Specific Heat:

Parameters from Neutron Scattering

Ross et al PRX

Old Data from

Inverse Problem: Data to Hamiltonian is much harder

Neutron data can be fit by multiple parameters!
FIG. 7. (Color online) (a) Experimental spectra obtained in reference 8 at $H = 2\ T$ and $T = 0.05\ mK$. (b,c,d) Examples of simulations, performed in the same temperature and field conditions, giving a good agreement with the data: (b) $(J_1, J_2, J_3, J_4) = (-0.09, -0.19, -0.26, 0.005)$; (c) $(J_1, J_2, J_3, J_4) = (0.64, -0.29, -0.26, 0.024)$, and (d): $(J_1, J_2, J_3, J_4) = (-0.02, -0.34, -0.29, 0.036)\ meV$. 
New set of exchange constants where $J_{zz}$ no longer dominates!
EXCHANGE CONSTANTS FROM THERMODYNAMIC DATA

NOT MEANT TO REPLACE THE NEUTRON SPECTRA BUT GET A QUICK AND DIRTY FIRST ESTIMATE OF EXCHANGE CONSTANTS

WHERE ONE IS IN PARAMETER SPACE?
Inverse problem: from data to Hamiltonian

**What can not work:** Fitting to coefficients of HTE is not a viable option.

Need robust properties that are not sensitive to accurate details (e.g. very high-\(T\))

Magnetic susceptibility of hyper-kagome Lattice Heisenberg Model

![Graph 1](image1)

![Graph 2](image2)

Slope related to momentt

Measured Curie-Weiss constant depends on temperature range of fit
Can we get more than one exchange constants?

Magnetic susceptibility of anisotropic triangular -Lattice Heisenberg Model

Wethone Zheng, Rajiv R. P. Singh, Ross H. McKenzie and Radu Coldea

PHYSICAL REVIEW B 71, 134422 (2005)
In an anisotropic system, the Curies-Weiss constant is some combination of g-factors and exchange constants!

\[ \Theta_{CW} = \frac{1}{2k_B(2g_{xy}^2 + g_z^2)} \left[ g_z^2 J_{zz} - 4g_{xy}^2 (J_+ + 2J_{\pm\pm}) - 8\sqrt{2} g_{xy} g_z J_{\pm\pm} \right] \]

Zero-field heat capacity depends only on exchange constants

Can it be a quick and dirty method to get first estimate of exchange parameters?
KITAEV-HEISENBERG MODEL ON HONEYCOMB LATTICE (OITMAA+RRPS)

Height of the short-range heat capacity-peak tells you relative importance of Kitaev terms!
ZERO FIELD HEAT CAPACITY

How discriminating is short-ranged peak?

Peak Value of specific heat $C_m$

Peak temperature $T_m$

Peak Shape: Width to high-$T$/Width to low $T$

Overall energy scale is certainly constrained by the peak
Zoom in near the peak: Experimental data are close but not exact
One could adjust exchange constants, but …
Peak is shallow! … Significantly more variations from sample to sample
Going beyond the peak: 
High-Temperature behavior sets in above 5K
Twice above the peak temperature (I have not seen any experimental data for YbTO)
Curie-Weiss type plot can give two parameters: Wide temperature range
Both slope and intercept related to exchange constants

$T > 4-5 \text{ K will be needed.}$

Are there experimental data at such temperatures?
It would be difficult to get 4 exchange constants from zero-field measurements
It would be very difficult to get more than two parameters from zero-field heat capacity.

How about data in a field with single crystals (or powder average?)

Now there are at least six parameter! Js and gs

Variations from sample to sample? Disorder (McQueen, Petit)?
HEAT CAPACITY IN A FIELD [110]

Euler Transforms 3rd and 4th Order. At strong fields (>1T) 2nd order is good enough

In a field (strength and orientation) the peaks are well resolved in NLC. Are variations in peak locations discriminating enough?
Field along 111

Scheie et al 2017

Broholm talk
If specific heat is measured on a grid of field values
One can also do subtraction to eliminate background.

Without trying it is difficult to say if this would be discriminating enough!
Magnetization in a Field (M/H plots)


3 Different Field Directions
[110] [100] [111]

5T, 4T, 3T, 2T, 1T, 0.5T, 0.2T

Except for low temperature and low fields NLC converges well for thermodynamic properties.
Do we really need 6 (or more) parameters? Can machine learning help?

Sloppiness and Emergent Theories in Physics, Biology, and Beyond

Mark K. Transtrum, Benjamin Machta, Kevin Brown, Bryan C. Daniels, Christopher R. Myers, James P. Sethna

(Submitted on 30 Jan 2015)

Large scale models of physical phenomena demand the development of new statistical and computational tools in order to be effective. Many such models are ‘sloppy’, i.e., exhibit behavior controlled by a relatively small number of parameter combinations. We review an information theoretic framework for analyzing sloppy models. This formalism is based on the Fisher Information Matrix, which we interpret as a Riemannian metric on a parameterized space of models. Distance in this space is a measure of how distinguishable two models are based on their predictions. Sloppy model manifolds are bounded with a hierarchy of widths and extrinsic curvatures. We show how the manifold boundary approximation can extract the simple, hidden theory from complicated sloppy models. We attribute the success of simple effective models in physics as likewise emerging from complicated processes exhibiting a low effective dimensionality. We discuss the ramifications and consequences of sloppy models for biochemistry and science more generally. We suggest that the reason our complex world is understandable is due to the same fundamental reason: simple theories of macroscopic behavior are hidden inside complicated microscopic processes.

What are the most discriminating directions in parameters space?

Can a data-analysis based approach be complimentary to theoretical studies of global phase diagram?

Theory of multiple--phase competition in pyrochlore magnets with anisotropic exchange, with application to Yb2Ti2O7, Er2Ti2O7 and Er2Sn2O7

Han Yan, Owen Benton, Ludovic D. C. Jaubert, Nic Shannon

The End