Abstract: "Quantum spin ice" materials have been widely discussed in terms of an XXZ model on a pyrochlore lattice, which is accessible to quantum Monte Carlo simulation for unfrustrated interactions $J_{\pm} > 0$. Here we argue that the properties of this model may become even more interesting once it is "frustrated". Using a combination of large-scale classical Monte Carlo simulation, semi-classical molecular dynamics, symmetry analysis and analytic field theory we explore the new phases which arise for $J_{\pm} < 0$. We find that the model supports not one, but three distinct forms of spin liquid: spin ice, a U(1) spin liquid; a disguised version of the U(1) x U(1) x U(1) spin-liquid found in the Heisenberg antiferromagnet on a pyrochlore lattice; and another entirely new form of spin liquid described by a U(1) x U(1) gauge group. At low temperatures this novel spin liquid undergoes a thermodynamic phase transition into a ground state with hidden, spin-nematic order. We present explicit predictions for inelastic neutron scattering experiments carried out on the three different spin liquids [M. Taillefumier et al., arXiv:1705.00148].
Frustrating quantum spin ice: a tale of three spin liquids (and hidden order...)
“All good things come in threes.”
...life, liberty and the pursuit of happiness.
(...checksum is important)
"[...] And the LORD spake, saying, "First shalt thou take out the Holy Pin, then shalt thou count to three, no more, no less. Three shall be the number thou shalt count, and the number of the counting shall be three. Four shalt thou not count, neither count thou two, excepting that thou then proceed to three. Five is right out. Once the number three, being the third number, be reached, then lobbest thou thy Holy Hand Grenade of Antioch towards thy foe, who being naughty in My sight, shall snuff it."

Book of Armaments, Chapter 2, verses 9–21

(...checksum is important)
spin ice is a good thing...
spin ice is a good thing...
story for today

minimal model of quantum spin ice

\[ H_{\text{QSI}} = \sum_{\langle ij \rangle} J_{zz} \hat{S}_i^z \hat{S}_j^z - J_{\pm} \left( \hat{S}_i^+ \hat{S}_j^- + \hat{S}_i^- \hat{S}_j^+ \right) \]

phase diagram from quantum Monte Carlo simulation

\[(J_{\pm} > 0)\]

story for today

minimal model of quantum spin ice

$$\mathcal{H}_{\text{QSI}} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+)$$

phase diagram from quantum Monte Carlo simulation

$$(J_{\pm} > 0)$$

story for today

minimal model of quantum spin ice

\[ \mathcal{H}_{QSI} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_\pm (S_i^+ S_j^- + S_i^- S_j^+) \]

change sign of \( J_\pm \)

phase diagram from classical Monte Carlo simulation

M. Taillefumier et al., arXiv:1705.00148
story for today

minimal model of quantum spin ice
\[ \mathcal{H}_{QSI} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \]

change sign of \( J_{\pm} \)

3 spin liquids
+ hidden spin-nematic order!

phase diagram from classical Monte Carlo simulation

M. Taillefumier et al., arXiv:1705.00148
wouldn’t have happened without...

Mathieu Taillefumier  
(OIST → ETH)

Owen Benton  
(Bristol/OIST → RIKEN)

Han Yan  
(OIST)

Ludovic Jaubert  
(OIST → CNRS)
minimal model of a QSI
minimal model of a QSI

general model of nearest-neighbor exchange in local basis (Kramers ion):

\[ H_{ex} = \sum_{\langle ij \rangle} \left\{ J_{\pm \pm} S_i^z S_j^z - J_{\pm \pm} (S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm \pm} \left[ \gamma_{ij} S_i^z S_j^z + \gamma_{ij} S_i^- S_j^+ \right] + J_{\pm \pm} \left[ S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij} S_j^-) + i \leftrightarrow j \right] \} \]
minimal model of a QSI

general model of nearest-neighbor exchange in local basis (Kramers ion):

\[ \mathcal{H}_{\text{ex}} = \sum_{(ij)} \left\{ J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm\pm} \left[ \gamma_{ij} S_i^z S_j^z + \gamma_{ij}^* S_i^- S_j^+ \right] + J_{z\pm} \left[ S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j \right] \right\} \]

favors spin-ice dynamics

\((J_{zz} > 0)\)

cf. most of the talks in this workshop!

many authors (in many notations), with papers including:

what happens for \( J_\pm > 0 \) ?

QMC results taken from:
what happens for $J_\pm > 0$?

what happens for $J_{\pm} < 0$?

QMC “frustrated”, i.e. suffers from (severe) sign problems

what do we know?

1. quantum spin ice ground state perturbatively stable

what happens for $J_{\pm} < 0$?

QMC “frustrated”, i.e. suffers from (severe) sign problems

what do we know?

1. quantum spin ice ground state perturbatively stable


$$\mathcal{H}_{\text{tunneling}} = -g \sum_{\circlearrowleft} |\circlearrowright\rangle\langle\circlearrowright| + |\circlearrowleft\rangle\langle\circlearrowleft|$$

$$g = \frac{12J_{\pm}^3}{J_{zz}^2}$$
what happens for $J_{\pm} < 0$?

QMC “frustrated”, i.e. suffers from (severe) sign problems

what do we know?

1. quantum spin ice ground state perturbatively stable

   $$\mathcal{H}_{\text{tunneling}} = -g \sum_{\oplus} |\oplus\rangle\langle\oplus| + |\bigcirc\rangle\langle\bigcirc|$$

   $$g = \frac{12J_{\pm}^3}{J_{zz}^2}$$

   can swap sign using “gauge” transformation

2. spinon dispersion is modified; gauge MFT predicts broad QSI regime
can classical simulations help?

cf. QMC for \( J_\pm > 0 \)
can classical simulations help?

cf. QMC for \( J_\pm > 0 \)
can classical simulations help?

cf. QMC for $J_\pm > 0$
can classical simulations help?

$S(q)$

$S_{SF}(q)$

$S_{NSF}(q)$

$T/J_{zz}$

$10^{-1}$

$10^{-2}$

$10^{-3}$

$10^{-4}$

$T/T^*$

PM

CSI

AF$_\perp$

cf. QMC for $J_\pm > 0$

QSI

classical spin ice

$J_\pm/J_{zz}$

$0.025$

$0.050$

$0.075$

$0.100$

QSI Workshop, Perimeter Institute 08.06.17
can classical simulations help?

qualitatively identical phase diagram (apart from absence of low-T QSI regime)

quantitative differences in $T^*$, $T_N$, and critical value of $J_\pm/J_{zz}$
classical ground states?

\[ H_{ex} = \sum_{(ij)} \{ J_{\pm z} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) + J_{\pm \pm} [\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij} S_i^- S_j^-] + J_{\pm \mp} [S_i^+ \zeta_{ij} S_j^+ + \zeta_{ij} S_i^- S_j^-] \} \]

rewrite in terms of irreps of \( T_d \)

\[ \{ m_\lambda \} = \{ m_{A_2}, m_E, m_{T_1 \text{ice}}, m_{T_1 \text{planar}}, m_T \} \]

spin ice is surrounded by competing forms of easy-plane AF order

Owen Benton, PhD Thesis
H. Yan et al., arXiv:1311.3501
what do we expect at finite T?

spin ice  \[ J_{\pm}/J_{zz} \]

T=0 classical phase boundary
what do we expect at finite $T$?

$$\mathcal{H}_{QSI} = J_{zz} \sum_{\langle ij \rangle} S_i \cdot S_j$$

spin ice

$AF_{\perp}$

$T=0$ classical phase boundary

$J_{\perp}/J_{zz}$
what do we find?

M. Taillefumier et al., arXiv:1705.00148
SL 1 (spin ice)

SL 1 (spin ice)


SL 1 (spin ice)


SL 1 (spin ice)


SL 3 ("pseudo-Heisenberg AF")

\[ \mathcal{H}_{QSI} = \sum_{ij} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \]
SL 3 ("pseudo-Heisenberg AF")

\[ \mathcal{H}_\text{QSL} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_\pm (S_i^+ S_j^- + S_i^- S_j^+) \]

\[ J_\pm / J_{zz} = -0.5 \]

\[ \mathcal{H}_\text{pHAF} = J \sum_{\langle ij \rangle} S_i \cdot S_j \]

\[ S(q) \]
SL 3 ("pseudo-Heisenberg AF")

\[ \mathcal{H}_{QSI} = \sum_{ij} J_{zz} S_i^z S_j^z - J_\pm (S_i^+ S_j^- + S_i^- S_j^+) \]

\[ J_\pm / J_{zz} = -0.5 \]

\[ \mathcal{H}_{PHAF} = J \sum_{ij} S_i \cdot S_j \]

\[ S(q) \]

Heisenberg AF:

Local spin rotation

SL 3 ("pseudo-Heisenberg AF")

\[ \mathcal{H}_{\text{QSL}} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \]

\[ J_{\pm}/J_{zz} = -0.5 \]

\[ \mathcal{H}_{\text{PHAF}} = J \sum_{\langle ij \rangle} S_i \cdot S_j \]

\[ S(q) \]

N.B. HAF on pyrochlore lattice has a long history (Reimers, Berlinsky, Moessner...)

Heisenberg AF:
local spin rotation


C. Henley, Phys. Rev B 71, 014424 (2005): \( U(1) \times U(1) \times U(1) \) gauge theory
SL 2 (something completely different)
SL 2 (something completely different)

![Diagram of SL 2 (something completely different)]
where does SL 2 come from?

highly-degenerate manifold formed of combination of easy-plane states with $T_1$ and $T_2$ symmetry

O. Benton, PhD Thesis; M. Taillefumier et al., arXiv:1705.00148
where does SL 2 come from?

highly-degenerate manifold formed of combination of easy-plane states with $T_1$ and $T_2$ symmetry

![Diagram showing the relation between $J_{zz}$ and $S_{zz}$ with labels $T_1$, $T_2$, and $E$(AF$_2$).]

minimal model QSI

<table>
<thead>
<tr>
<th>$m_{T_1, planar}$</th>
<th>Definition in terms of spins within tetrahedron</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{1}{2} (S^0_0 + S^1_1 - S^2_2 - S^3_3)$</td>
<td></td>
</tr>
<tr>
<td>$\frac{1}{4} (-S^0_0 + \sqrt{3}S^0_0 + S^1_1 - \sqrt{3}S^2_2 - S^3_3 + \sqrt{3}S^4_4)$</td>
<td></td>
</tr>
<tr>
<td>$\frac{1}{4} (-S^0_0 - \sqrt{3}S^0_0 + S^1_1 + \sqrt{3}S^2_2 + S^3_3 - \sqrt{3}S^4_4)$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$m_{T_2}$</th>
<th>Definition in terms of spins within tetrahedron</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{1}{2} (-S^0_0 - S^1_1 + S^2_2 + S^3_3)$</td>
<td></td>
</tr>
<tr>
<td>$\frac{1}{4} (\sqrt{3}S^0_0 + S^1_1 - \sqrt{3}S^1_1 - S^2_2 + \sqrt{3}S^3_3 + S^4_4 - \sqrt{3}S^4_4 - S^0_0)$</td>
<td></td>
</tr>
<tr>
<td>$\frac{1}{4} (-\sqrt{3}S^0_0 + S^1_1 + \sqrt{3}S^1_1 - S^2_2 + \sqrt{3}S^3_3 - S^4_4 - \sqrt{3}S^4_4 - S^0_0)$</td>
<td></td>
</tr>
</tbody>
</table>

O. Benton, PhD Thesis; M. Taillefumier et al., arXiv:1705.00148
gauge structure?

combine $m_{T_1,\text{planar}}$ and $m_{T_2}$

into two fields satisfying zero-divergence condition

$B_1 = \frac{1}{2} (2m_{T_1,\text{planar}} - \sqrt{3}m_{T_2} - m_{T_1,\text{planar}}, \sqrt{3}m_{T_2} - m_{T_1,\text{planar}}) \quad \nabla \cdot B_1 = 0$

$B_2 = \frac{1}{2} (2m_{T_1,\text{planar}} - m_{T_2} + \sqrt{3}m_{T_1,\text{planar}}, -m_{T_2} - \sqrt{3}m_{T_1,\text{planar}}) \quad \nabla \cdot B_2 = 0$

can describe spin liquid SL 2 by introducing two U(1) gauge fields $A_1$ and $A_2$

$F_{SL2} = \frac{T}{V} \int d^3r \lambda \left[ (\nabla \times A_1)^2 + (\nabla \times A_2)^2 \right]$

$\Rightarrow$ pinch points in $B_1$ and $B_2$

M. Taillefumier et al., arXiv:1705.00148
gauge structure?

combine $\mathbf{m}_{T_1, \text{planar}}$ and $\mathbf{m}_{T_2}$ into two fields satisfying zero-divergence condition

$$\mathbf{B}_1 = \frac{1}{2} \left( 2m_{T_1, \text{planar}}^x, -\sqrt{3}m_{T_2}^y - m_{T_1, \text{planar}}^y, \sqrt{3}m_{T_1, \text{planar}}^z - m_{T_1, \text{planar}}^z \right) \quad \nabla \cdot \mathbf{B}_1 = 0$$

$$\mathbf{B}_2 = \frac{1}{2} \left( 2m_{T_1, \text{planar}}^x, m_{T_2}^y + \sqrt{3}m_{T_1, \text{planar}}^y, -m_{T_2}^z - \sqrt{3}m_{T_1, \text{planar}}^z \right) \quad \nabla \cdot \mathbf{B}_2 = 0$$

can describe spin liquid SL 2 by introducing two U(1) gauge fields $\mathbf{A}_1$ and $\mathbf{A}_2$

$$\mathcal{F}_{SL2} = \frac{T}{V} \int d^3 r \lambda \left[ (\nabla \times \mathbf{A}_1)^2 + (\nabla \times \mathbf{A}_2)^2 \right]$$

$\Rightarrow$ pinch points in $\mathbf{B}_1$ and $\mathbf{B}_2$

M. Taillefumier et al., arXiv:1705.00148
a new form of spin liquid…
a new form of spin liquid...
is that it?
staged loss of entropy

is that it?

spin correlations unchanged ⇒ hidden order

phase transition?
hidden order...

candidate for hidden order
is spin-nematic state
with director in easy-plane

\[ Q_{\perp} = \left( \frac{S_{\perp}^z - S_{\perp}^y}{2S_{\perp}^xS_{\perp}^y} \right) \]
hidden order…

candidate for hidden order is spin-nematic state with director in easy-plane

\[ Q_\perp = \left( S_{x^2} - S_{y^2} \right) / \sqrt{2} S_x S_y \]

\begin{align*}
\mathbf{m}_{T_1,\text{plan}} & = \frac{1}{2} \left( -S_0^x + S_1^x - S_2^x - S_3^x \right) \\
& \quad + \frac{1}{4} \left( -S_0^y + \sqrt{3}S_0^y + S_1^y - \sqrt{3}S_1^y - S_2^y + \sqrt{3}S_2^y + S_3^y - \sqrt{3}S_3^y \right) \\
& \quad + \frac{1}{4} \left( -S_0^z - \sqrt{3}S_0^z + S_1^z + \sqrt{3}S_1^z - S_2^z + \sqrt{3}S_2^z - S_3^z - \sqrt{3}S_3^z \right)
\end{align*}

\begin{align*}
\mathbf{m}_{T_2} & = \frac{1}{4} \left( \sqrt{3}S_0^x + S_1^x - \sqrt{3}S_1^x - S_2^x + \sqrt{3}S_2^x + S_3^x - \sqrt{3}S_3^x \right) \\
& \quad + \frac{1}{4} \left( -S_0^y - \sqrt{3}S_0^y + S_1^y + \sqrt{3}S_1^y + S_2^y - \sqrt{3}S_2^y - S_3^y - \sqrt{3}S_3^y \right) \\
& \quad + \frac{1}{4} \left( -S_0^z + \sqrt{3}S_0^z + S_1^z - \sqrt{3}S_1^z + S_2^z - \sqrt{3}S_2^z - S_3^z + \sqrt{3}S_3^z \right)
\end{align*}

\begin{tabular}{|c|c|c|}
\hline
$T_{Q_\perp}$ & SL II & SL III PM \\
\hline
\end{tabular}

\[ J_\perp / J_{zz} = -1 \]

M. Taillefumier et al., arXiv:1705.00148
what about spin excitations?
what about spin excitations?

\[ S(q) \]

Dynamical structure factor for spins: \( S(q, \omega) \)

Q-phase
what about spin excitations?

\[ S(q) \]

non-dispersing continuum

\[ \chi_Q(q, \omega) \]

 dynamical structure factor for spins: \( S(q, \omega) \)

 dynamical correlation function for quadrupoles: \( \chi_Q(q, \omega) \)
what about spin excitations?

\[ S(q) \]

\[ \chi_Q(q, \omega) \]

**Q-phase**

**non-dispersing continuum**

**linearly-dispersing Goldstone mode**

**dynamical structure factor for spins:** \( S(q, \omega) \)

**dynamical correlation function for quadrupoles:** \( \chi_Q(q, \omega) \)
what have we learned? (theory)

T / J \_z\_z

1
t2^*
T_3^*
SL II
T_Q
Q

SL III (pHAF)

PM

SL 1 (Spin Ice)

T_N

AF_\perp

J_\perp / J \_z\_z

SL 2

SL 3 (pHAF)

SL 1 (spin ice)
what have we learned? (theory)
what have we learned? (experiment)

pinch points $\not\Rightarrow$ spin ice

SL 1 (spin ice)  
SL 2 & Q  
SL 3 (pHAF)
what have we learned? (experiment)

pinch points ≠ spin ice

SL 1 (spin ice)  SL 2 & Q  SL 3 (pHAF)

non-dispersing continua may hide many secrets…
what’s left?

quantum effects !!!
the story so far...

minimal model of quantum spin ice

\[ H_{QSI} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \]

change sign of \(J_{\pm}\)

3 spin liquids
+ hidden spin-nematic order!
the story so far...

minimal model of quantum spin ice

$$\mathcal{H}_{QSI} = \sum_{\langle ij \rangle} J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+)$$

change sign of $J_{\pm}$

3 spin liquids + hidden spin-nematic order!

omne trium perfectum !!!