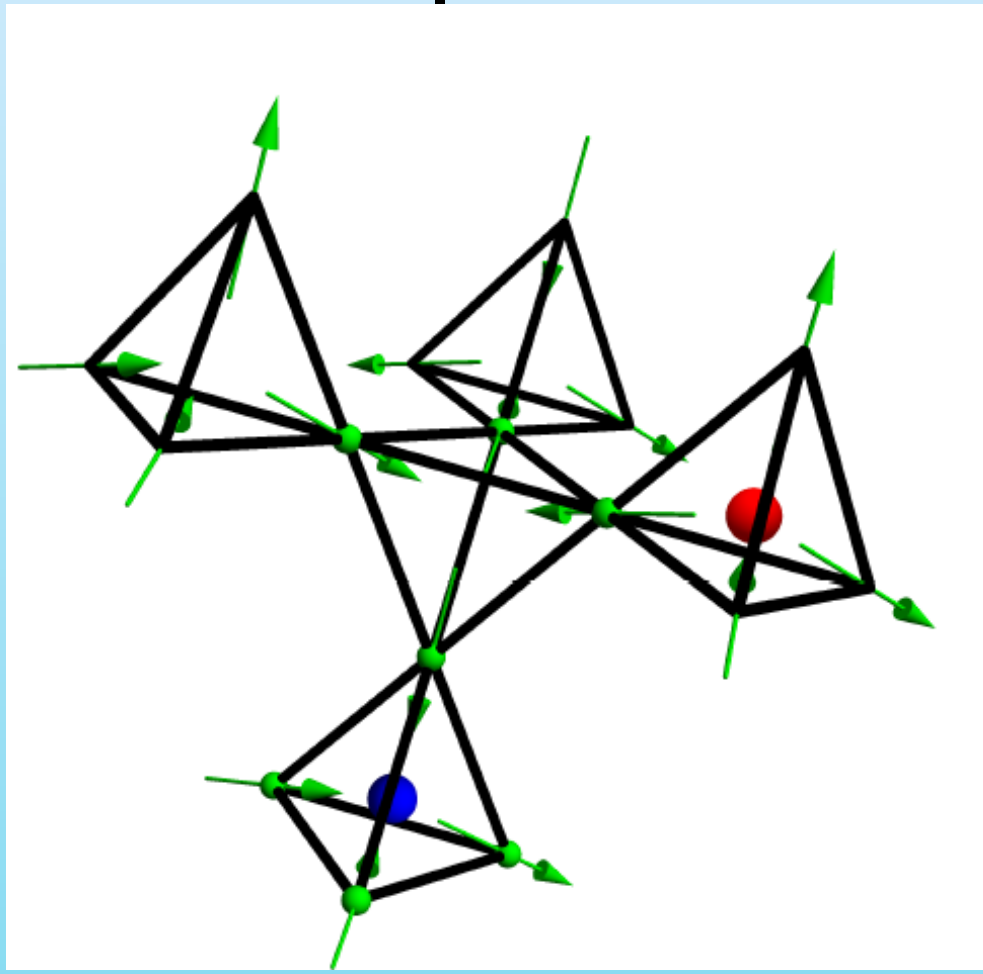
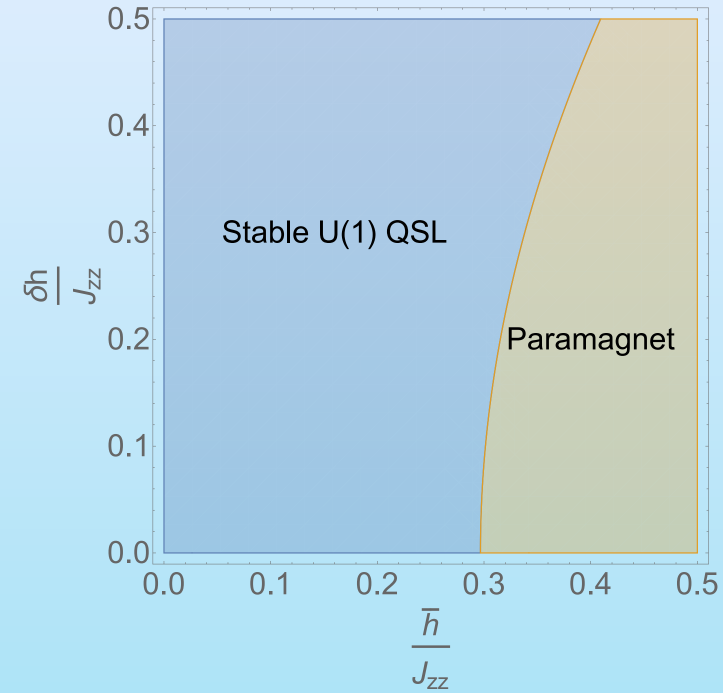
# Instabilities of the quantum spin ice state



Owen Benton
RIKEN Center for Emergent Matter Science, Wako

# Two questions for today

1) When does the U(1) QSL become unstable against spinon condensation?




2) Is there a disorder induced spin liquid in $Pr_2Zr_2O_7$ and other non-Kramers pyrochlores?

PRL 118, 087203 (2017) PHYSICAL REVIEW LETTERS week ending 24 FEBRUARY 2017

**Disorder-Induced Quantum Spin Liquid in Spin Ice Pyrochlores**

Lucile Savary[1,*] and Leon Balents[2]

PRL 118, 107206 (2017) PHYSICAL REVIEW LETTERS week ending 10 MARCH 2017

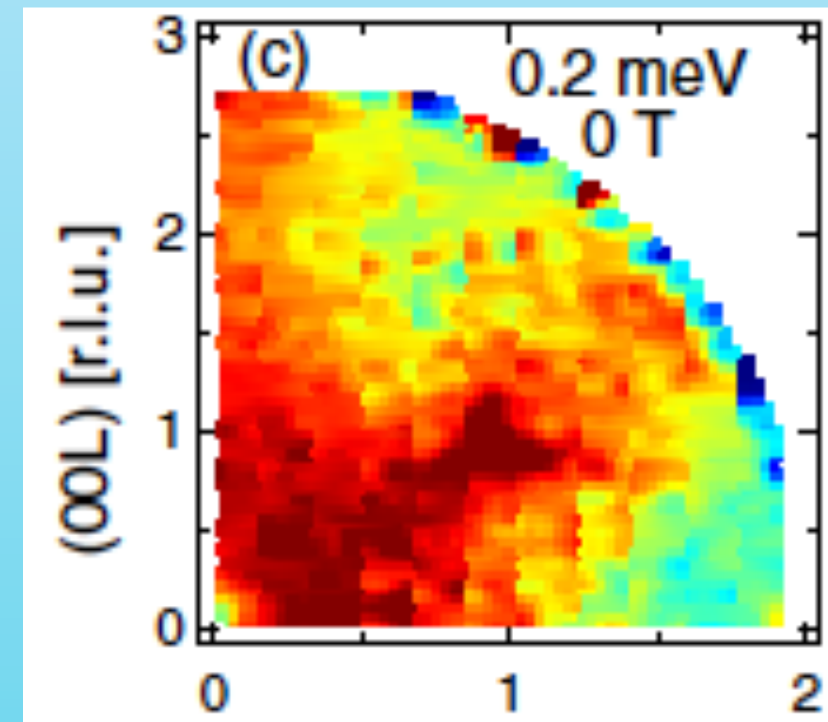
**Disordered Route to the Coulomb Quantum Spin Liquid: Random Transverse Fields on Spin Ice in $Pr_2Zr_2O_7$**

J.-J. Wen,[1,2,3] S. M. Koohpayeh,[1] K. A. Ross,[1,4] B. A. Trump,[5] T. M. McQueen,[1,5,6] K. Kimura,[7,8] S. Nakatsuji,[7,9] Y. Qiu,[4] D. M. Pajerowski,[4] J. R. D. Copley,[4] and C. L. Broholm[1,4,6]

**From quantum spin liquid to paramagnetic ground states in disordered non-Kramers pyrochlores**

Owen Benton[1]

[1] *RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama, 351-0198, Japan*



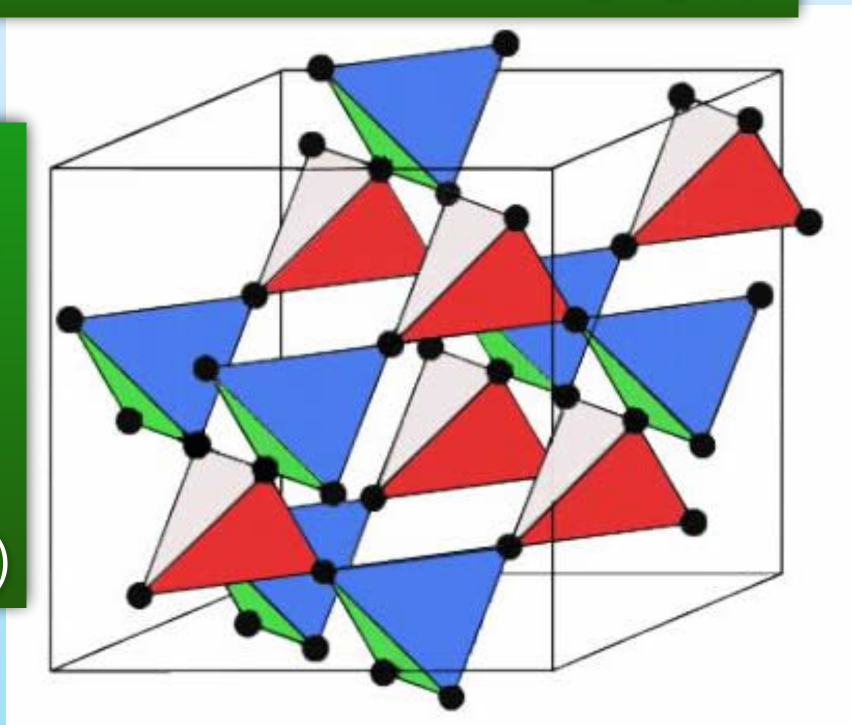

arXiv:1706.09238

# Spin Ice

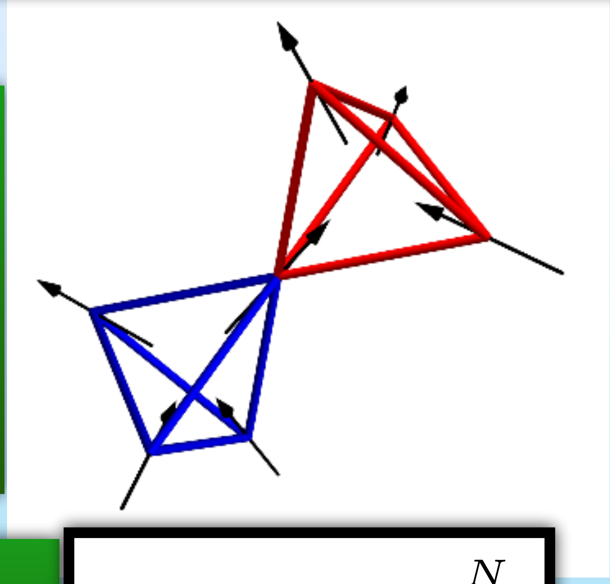
Harris et al., PRL 79, 2554 (1997)

Magnetic Rare Earth Oxides: $R_2M_2O_7$

$R^{3+}$ ions form lattice of corner-sharing tetrahedra (pyrochlore)



$R^{3+} = Ho^{3+}, Dy^{3+}$ : anisotropy forces magnetic moments to point along local <111> directions

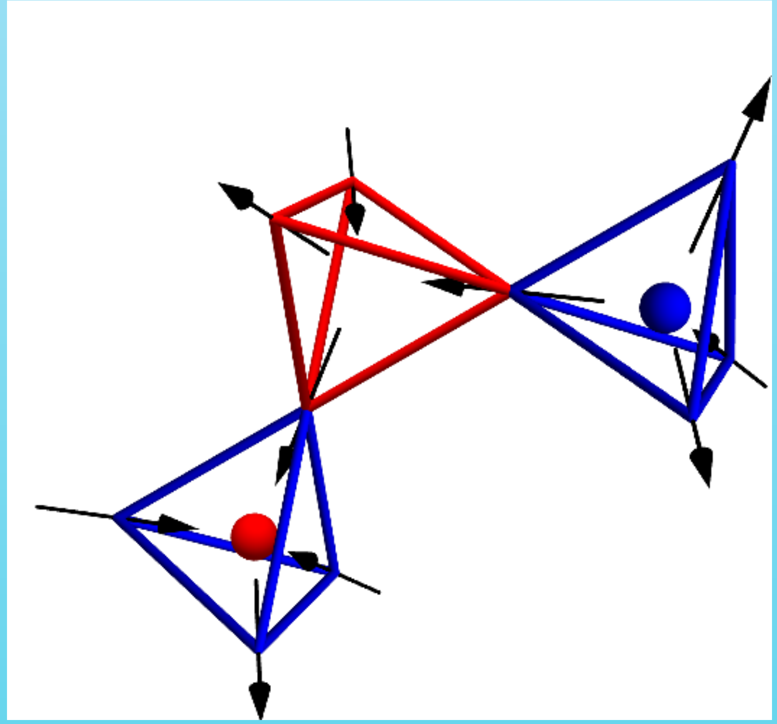


Ground states: “2 in, 2 out”

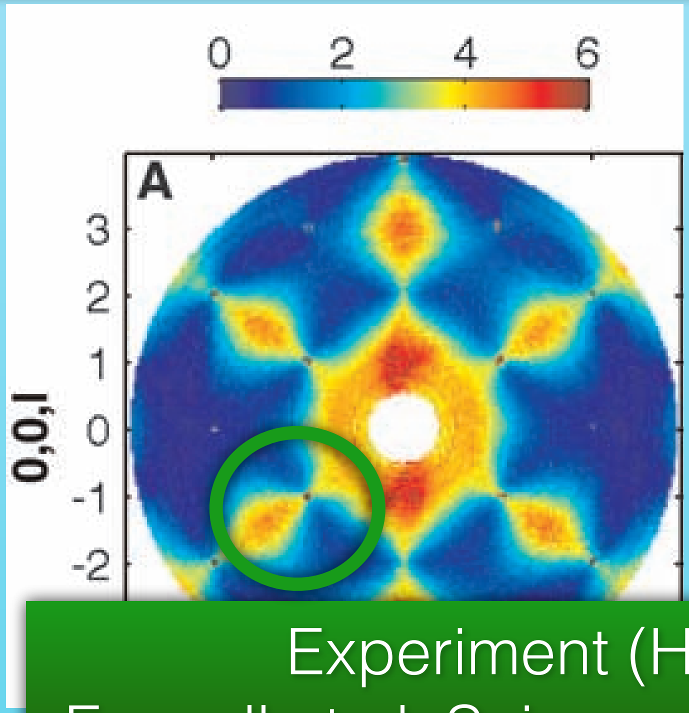
$$W \sim \left(\frac{3}{2}\right)^{\frac{N}{2}}$$

$\Longrightarrow$ large entropy at T=0!

Excitations: “magnetic monopoles”



Neutron scattering: pinch points




Experiment ($Ho_2Ti_2O_7$)
Fennell et al, Science **326**, 415 (2009)

# Quantum Spin Ice

Favours "2 in, 2 out" states

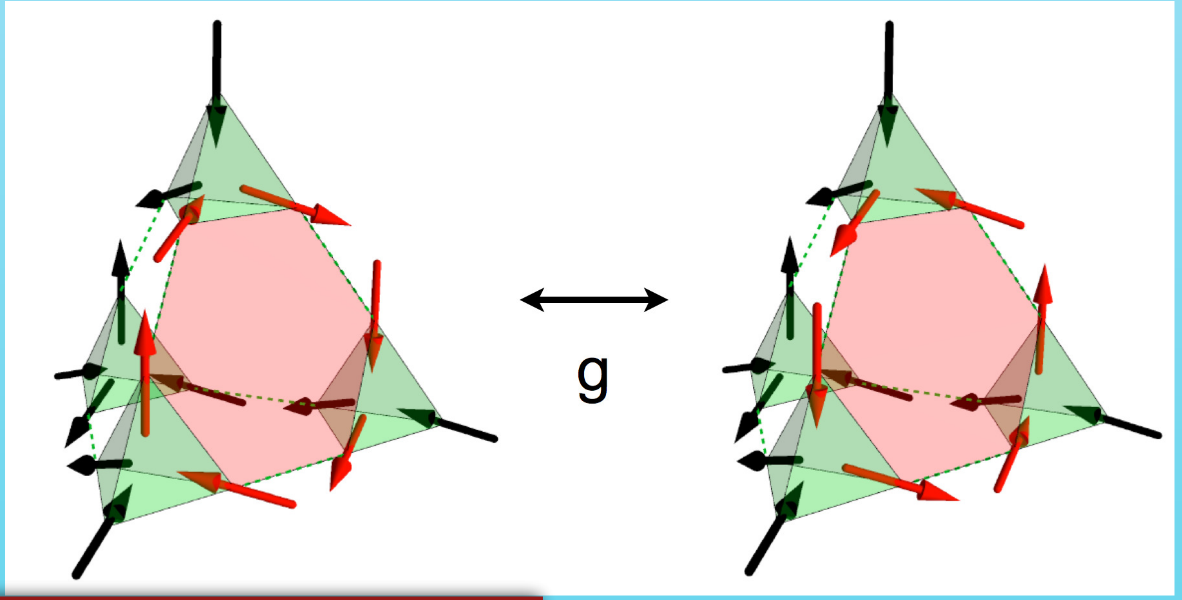
Introduces quantum fluctuations

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathsf{S}_i^z \mathsf{S}_j^z + V[\{\mathsf{S}_i^x, \mathsf{S}_j^y\}]$$

$$-J_{\pm} \sum_{\langle ij \rangle} \mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+ \qquad -h \sum_i \mathsf{S}_i^x$$

Effective Hamiltonian from perturbation theory

$$\mathcal{H}_{\text{ring}} = -g \sum_{\text{hex}} [| \circlearrowleft \rangle \langle \circlearrowright | + | \circlearrowright \rangle \langle \circlearrowleft |]$$




Hermele et al., PRB **69**, 064404 (2004)

# Quantum Spin Ice

Favours “2 in, 2 out” states

Introduces quantum fluctuations

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathsf{S}_i^z \mathsf{S}_j^z + V[\{\mathsf{S}_i^x, \mathsf{S}_j^y\}]$$

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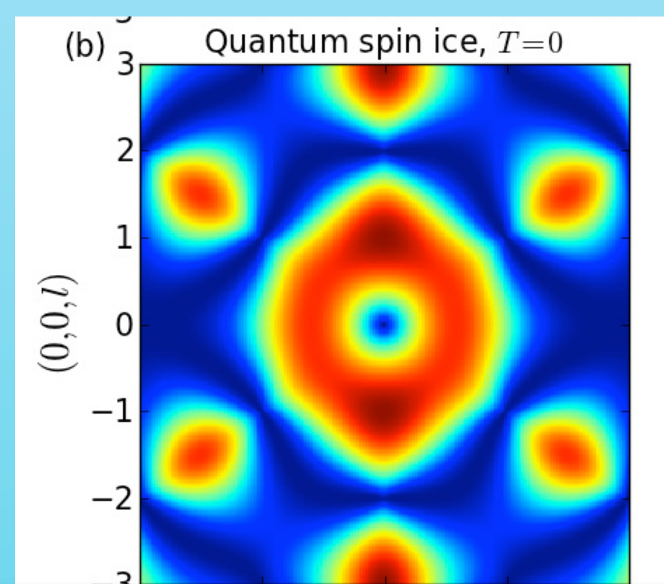
Effective Hamiltonian from perturbation theory

$$\mathcal{H}_{\mathsf{ring}} = -g \sum [| \circlearrowleft \rangle \langle \circlearrowright | + | \circlearrowright \rangle \langle \circlearrowleft |]$$

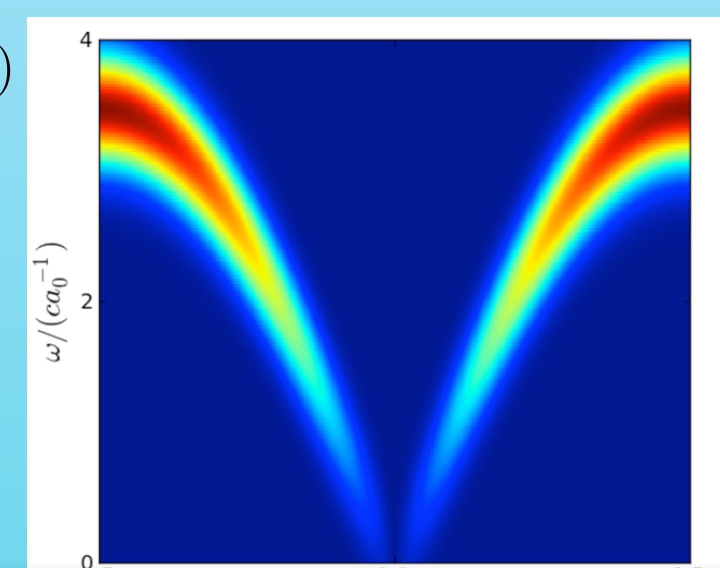
Ground state: U(1) quantum spin liquid with gapless emergent photons

$S(\mathbf{q}, t = 0)$




Pinch points disappear at T=0

$S(\mathbf{q}, \omega)$

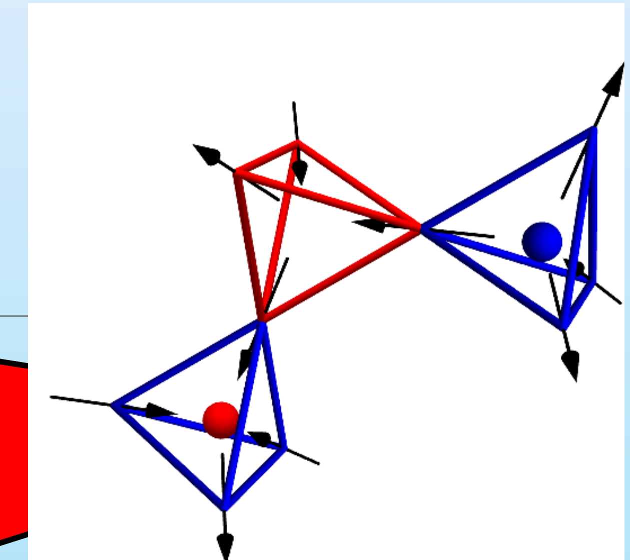


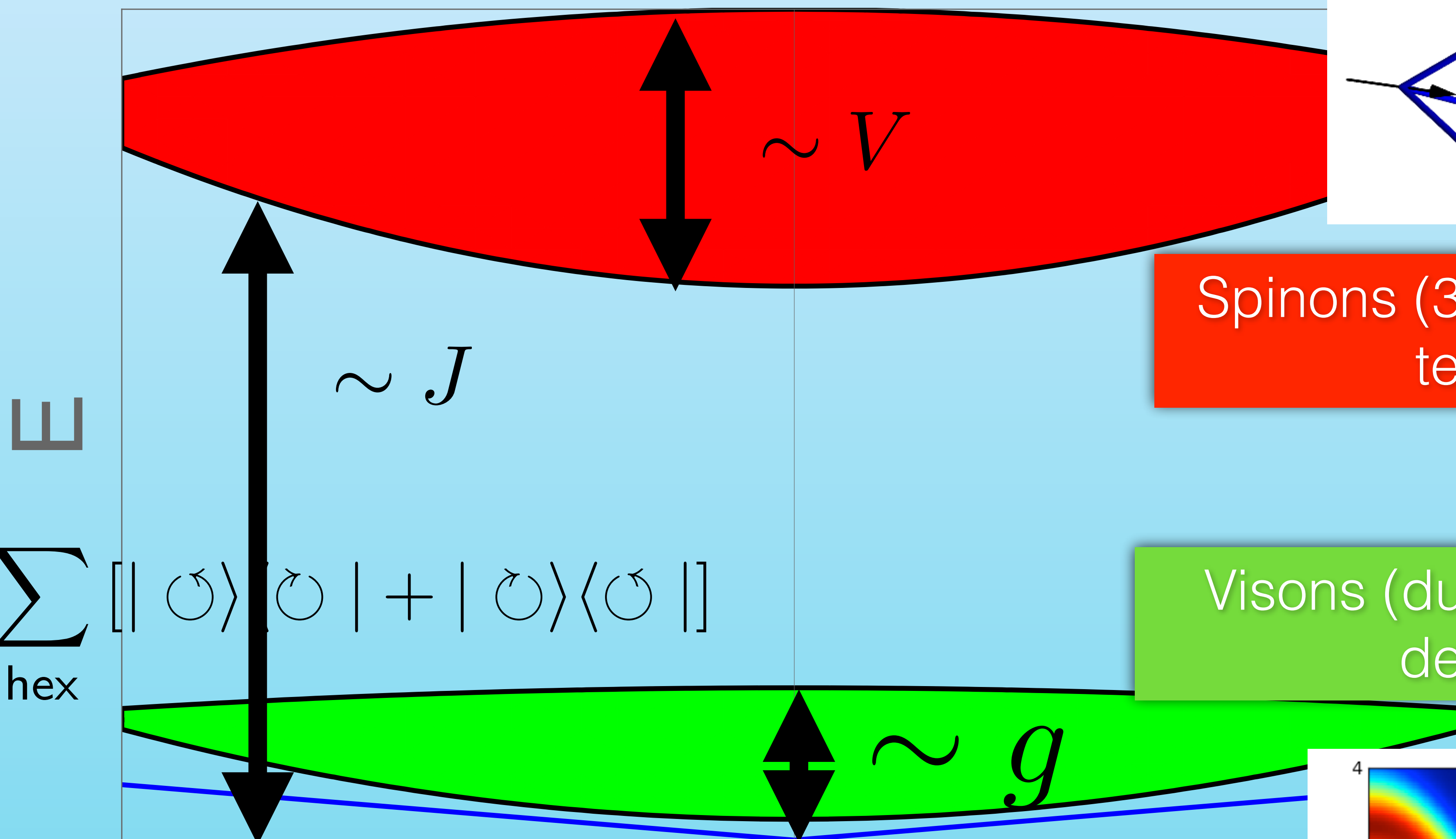

Linearly dispersing photon

Shannon et al., PRL 108, 067204 (2012); O. B. et al. PRB 86, 075154 (2012)

# Spectrum of quantum spin ice

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathsf{S}_i^z \mathsf{S}_j^z + V[\{\mathsf{S}_i^x, \mathsf{S}_j^y\}]$$



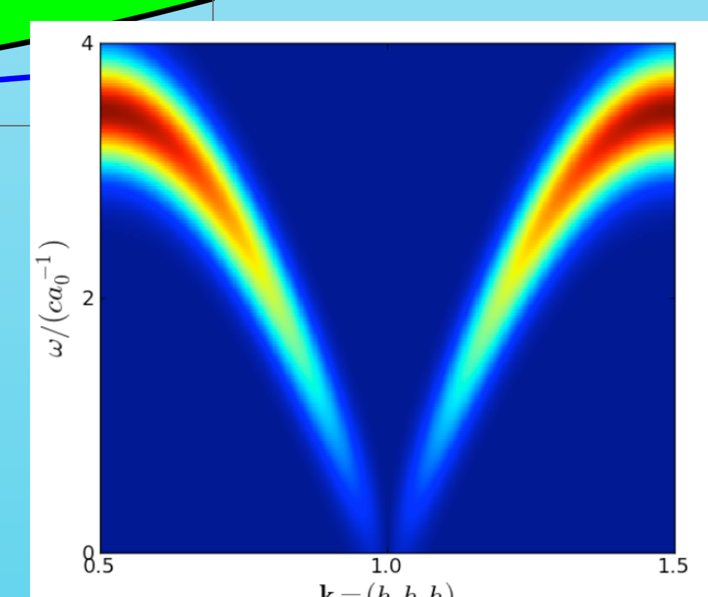



$$\mathcal{H}_{\text{ring}} = -g \sum_{\text{hex}} [|\circlearrowleft\rangle\langle\circlearrowright| + |\circlearrowright\rangle\langle\circlearrowleft|]$$

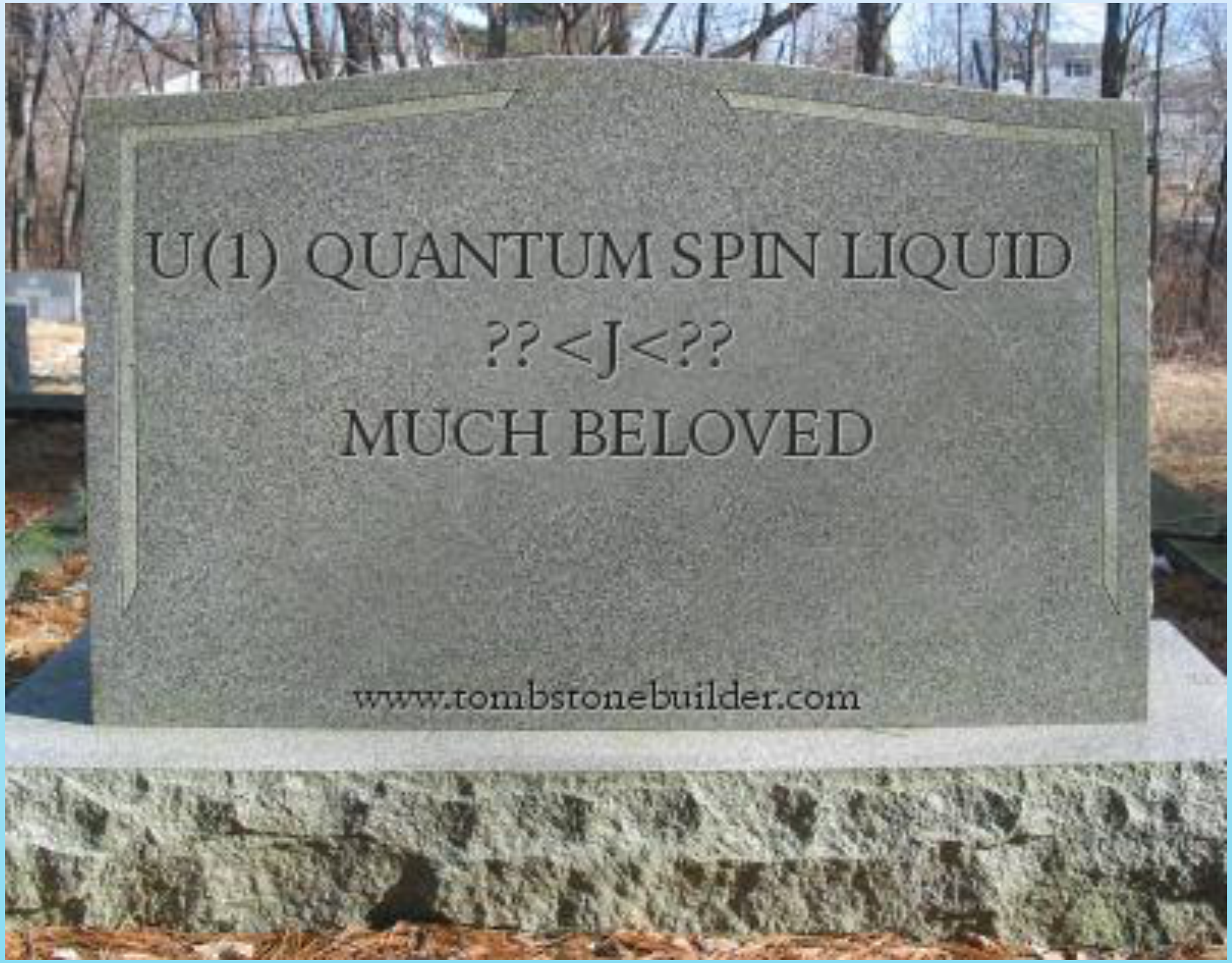
Spinons (3 in 1 out/1 in 3 out tetrahedra)

Visons (dual topological defects)

Photons
Transverse fluctuations of gauge field



# How and when does the U(1) QSL die?




How does it die?

By condensation of topological excitations

When does it die?

When the gap to these excitations closes

**Purpose of this talk:**

(1) a controlled perturbative calculation of the phase boundaries of the U(1) QSL in quantum spin ice by considering the gap to topological excitations

(2) application of this calculation to non-Kramers quantum spin ice candidates with quenched structural disorder

# Usual Approaches

(1) Gauge Mean Field Theory (gMFT)


Savary & Balents, PRL **108**, 037202 (2012)
Lee et al, PRB **86**, 104412 (2012)


$$S^{+}_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu} = \Phi^\dagger_{\mathbf{r}} \mathsf{s}^{+}_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu} \Phi_{\mathbf{r}+\mathbf{e}_\mu}$$
$$S^{-}_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu} = \Phi_{\mathbf{r}} \mathsf{s}^{-}_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu} \Phi^\dagger_{\mathbf{r}+\mathbf{e}_\mu}$$

spinon operators $\Phi^\dagger_{\mathbf{r}}, \Phi_{\mathbf{r}}$

gauge field $\mathsf{s}^{\pm}_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu} \sim e^{iA_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu}}$

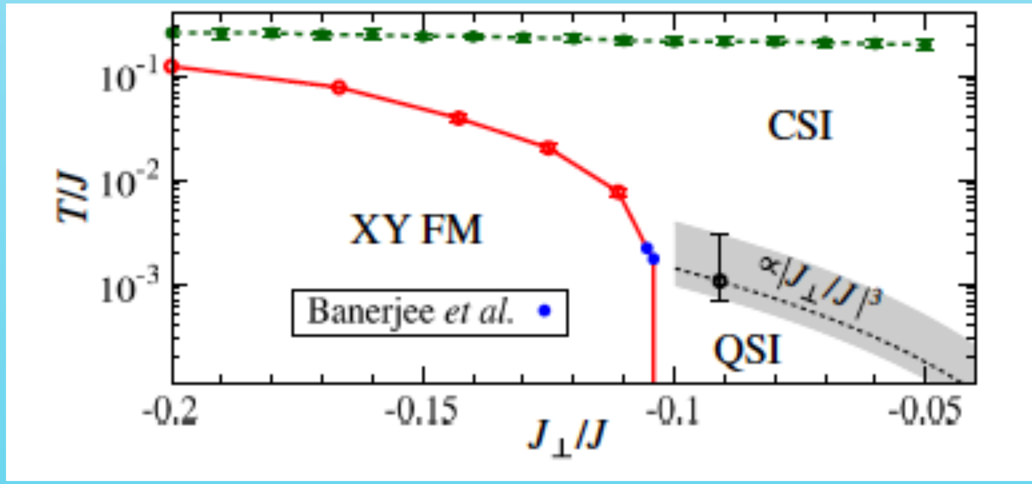
local constraints on spinon operators $\Phi^\dagger_{\mathbf{r}}\Phi_{\mathbf{r}} = 1$

Mean field decoupling gives non-interacting spinon hopping Hamiltonian + constraints enforced on average

-Uncontrolled treatment of constraints
-Tendency to overestimate QSL regime

(2) Numerics


Banerjee et al, PRL **100**, 047208 (2008)
Shannon et al, PRL **108**, 067204 (2012)
Kato & Onoda, PRL **115**, 077202 (2015)


-QMC not always possible (sign problems)
-Many other methods are limited to small system size
-Can be hard to interpret

# An alternative method

Perturbation theory in manifolds of classical monopole (spinon) states

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathsf{S}_i^z \mathsf{S}_j^z + V[\{\mathsf{S}_i^x, \mathsf{S}_j^y\}]$$

Finding lowest energy state containing M spinons in a system of $N_t$ tetrahedra

$$\rho = \frac{M}{N_t} << 1$$

Expand energy of system in terms of spinon density

$$E(\rho) = N_t \left[ \epsilon_0(J,V) + \underbrace{\epsilon_1(J,V)}\rho + \epsilon_2(J,V)\rho^2 + ... \right]$$

Calculate coefficient of linear term using perturbation theory in V

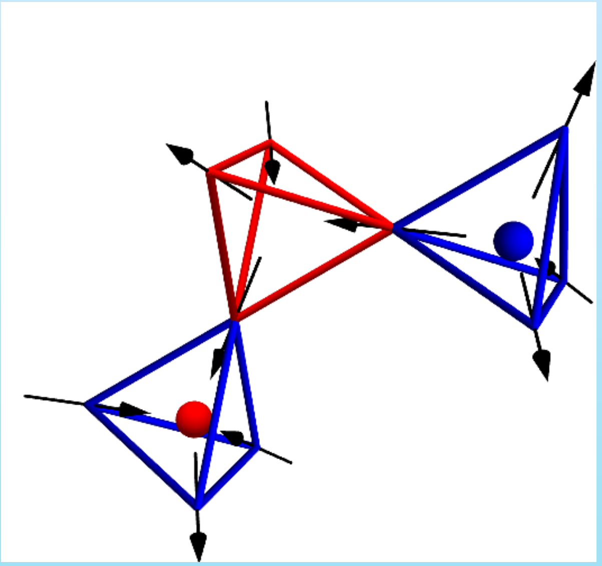
Change in sign of $\varepsilon_1$ +ve to -ve means that it becomes energetically for spinons to proliferate $\Longrightarrow$ Instability of the U(1) QSL

# Worked Example: unfrustrated XXZ model

Perturbation theory in manifolds of classical monopole (spinon) states

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

$$J_{\pm} > 0$$



State with M spinons

Classical energy (0th order PT)

$$E = E_0 + M \frac{J_{zz}}{2} = E_0 + N_t \rho \frac{J_{zz}}{2}, \quad \epsilon = \frac{J_{zz}}{2}$$

First order PT: Effective Hamiltonian

acts within set of M spinon states

$$\mathcal{H}_{\text{eff}}^{(1)} = -J_{\pm} \mathcal{P}_M \left[ \sum_{\langle ij \rangle} S_i^+ S_j^- + S_i^- S_j^+ \right] \mathcal{P}_M$$

projector onto manifold of state containing M spinons

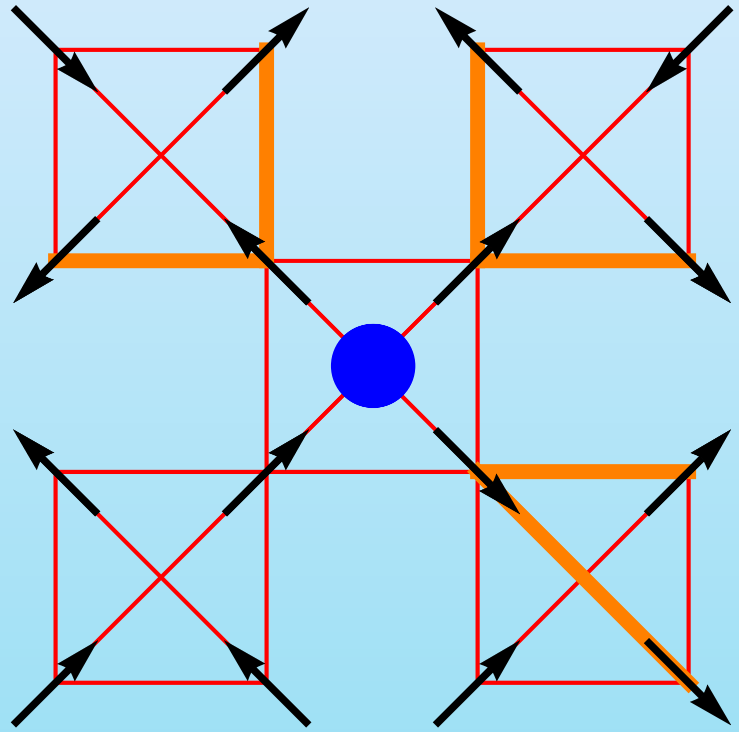
What is the ground state of the effective Hamiltonian?

# Worked Example: unfrustrated XXZ model

$$\mathcal{H}_{\text{eff}}^{(1)} = -J_{\pm}\mathcal{P}_M \left[ \sum_{\langle ij \rangle} S_i^+ S_j^- + S_i^- S_j^+ \right] \mathcal{P}_M$$

What is the ground state of the effective Hamiltonian?

As long as spinons are well separated (assume low density ρ<<1), each one has 6 neighbouring bonds that can be flipped to propagate the monopole



$\Longrightarrow$ Constant column sum of effective Hamiltonian for ρ<<1

$$\sum_{\alpha} \left( \mathcal{H}_{\text{eff}}^{(1)} \right)_{\alpha\beta} = -6MJ_{\pm}$$

$\Longrightarrow$ Ground state is equal weight sum over all M spinon states is an eigenstate of Hamiltonian

$$|\phi_0\rangle = \sqrt{\frac{1}{\mathcal{N}_M}} \sum_{|\alpha\rangle \in \{|M\rangle\}} |\alpha\rangle$$

with eigenvalue $-6MJ_{\pm}$

PRL 116, 167202 (2016) PHYSICAL REVIEW LETTERS week ending 22 APRIL 2016

**Spinon Walk in Quantum Spin Ice**

Yuan Wan,[1] Juan Carrasquilla,[1] and Roger G. Melko[1,2]

PHYSICAL REVIEW B 94, 104401 (2016)

**Free coherent spinons in quantum square ice**

Stefanos Kourtis
*Department of Physics, Princeton University, Princeton, New Jersey 08544, USA*

Claudio Castelnovo
*TCM Group, Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom*

# Worked Example: unfrustrated XXZ model

Total energy of ground state for M spinons

$$|\phi_0\rangle = \sqrt{\frac{1}{\mathcal{N}_M}} \sum_{|\alpha\rangle \in \{|M\rangle\}} |\alpha\rangle$$

$$E = E_0 + M\left(\frac{J_{zz}}{2} - 6J_\pm + \mathcal{O}(J_\pm^2)\right) = E_0 + N_t\rho\left(\frac{J_{zz}}{2} - 6J_\pm + \mathcal{O}(J_\pm^2)\right)$$

cf. $$E(\rho) = N_t\left[\epsilon_0(J,V) + \epsilon_1(J,V)\rho + \epsilon_2(J,V)\rho^2 + \ldots\right]$$

$$\Longrightarrow \quad \epsilon_1 = \frac{J_{zz}}{2} - 6J_\pm + \mathcal{O}(J_\pm^2)$$

Estimate of ground state instability to first order:

$$J_\pm^{(c)} = \frac{J_{zz}}{12} \approx 0.083 J_{zz}$$

# Worked Example: unfrustrated XXZ model

$$\epsilon_1 = \frac{J_{zz}}{2} - 6J_{\pm} + \mathcal{O}(J_{\pm}^2)$$

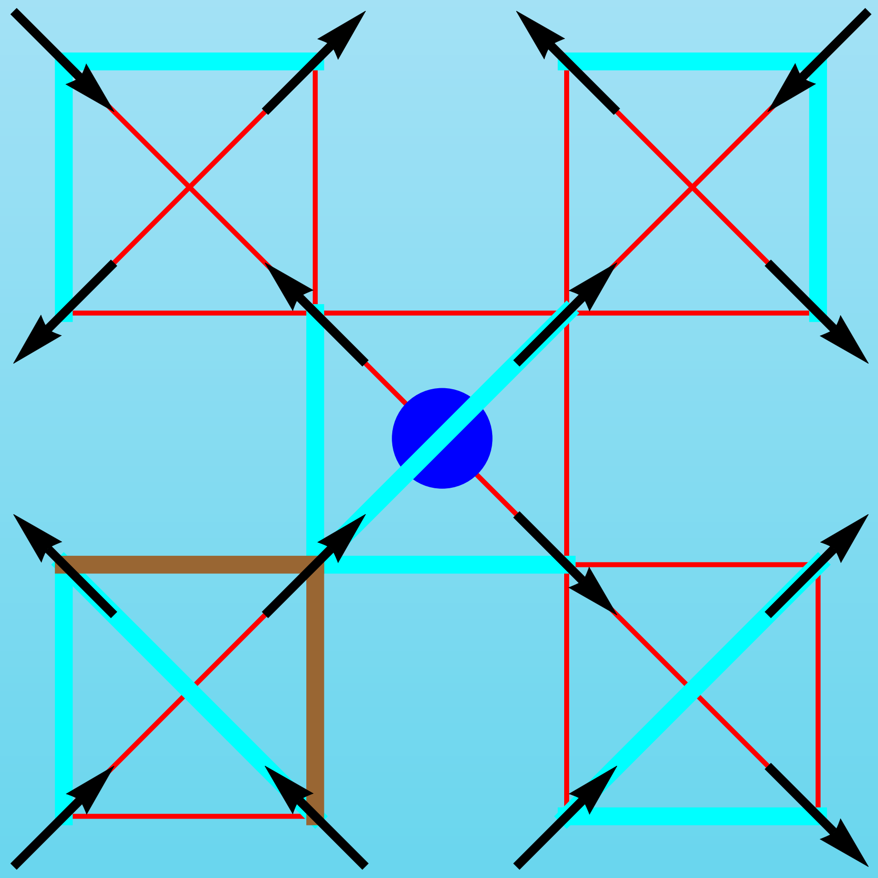
Second order corrections to energy

$$\mathcal{H}_{\text{eff}}^{(2)} = -\mathcal{P}_M V \frac{1-\mathcal{P}_M}{\mathcal{H}_0 - (E_0^{\text{cl}} + M\frac{J_{zz}}{2})} V\mathcal{P}_M$$

$$V = -J_{\pm}\sum_{\langle ij\rangle}\left[S_i^+ S_j^- + S_i^- S_j^+\right]$$

$$\mathcal{H}_0 = J_{zz}\sum_{\langle ij\rangle} S_i^z S_j^z$$

Diagonal corrections to the energy (act on same bond twice)



$$\left(\mathcal{H}_{\text{eff}}^{(2)}\right)_{\alpha\alpha} = \frac{J_{\pm}^2}{J_{zz}}(-4N_t + 8M)$$

Correction to ground state energy

Diagonal correction to energy of spinons

# Worked Example: unfrustrated XXZ model

$$\epsilon_1 = \frac{J_{zz}}{2} - 6J_\pm + \mathcal{O}(J_\pm^2)$$

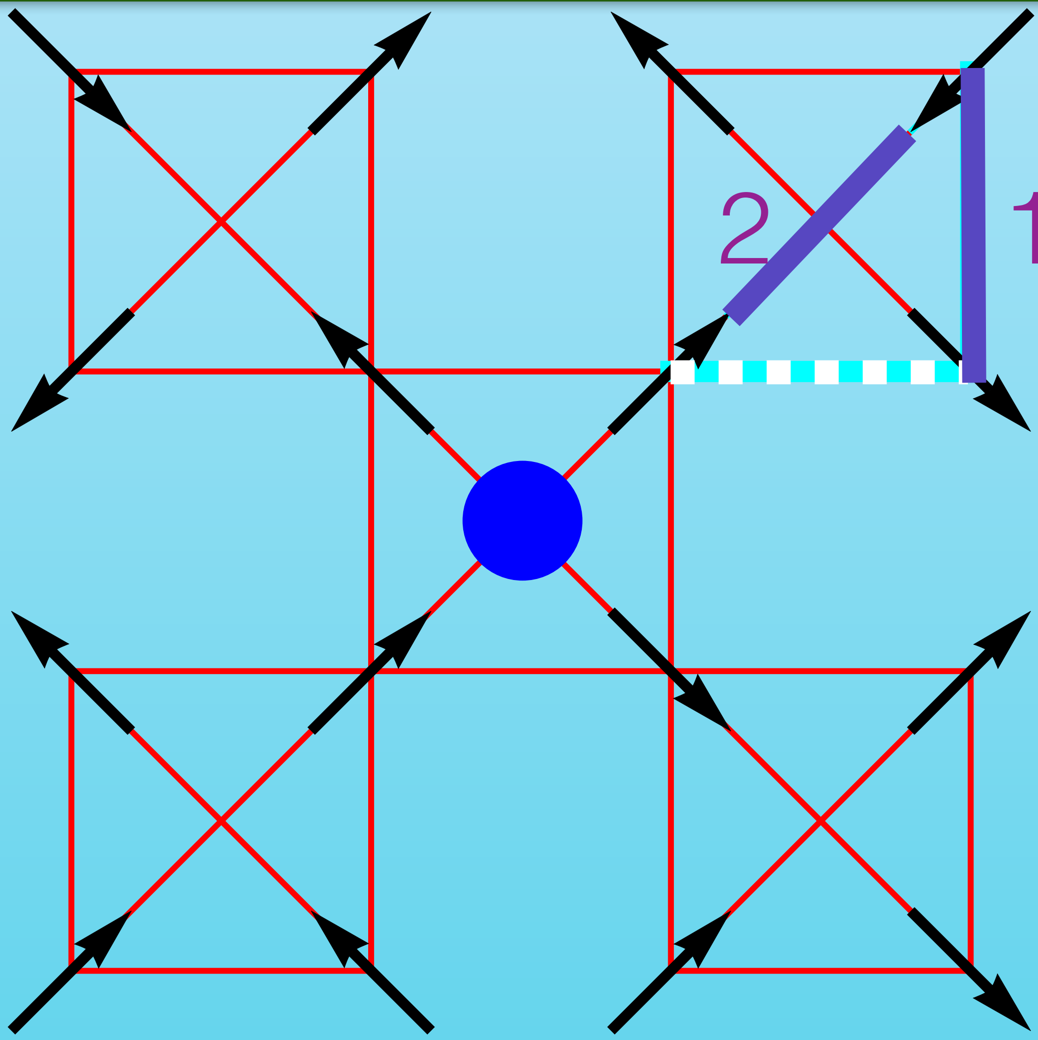
Second order corrections to energy

$$\mathcal{H}_{\text{eff}}^{(2)} = \mathcal{P}_M V \frac{1-\mathcal{P}_M}{\mathcal{H}_0} V \mathcal{P}_M$$

$$V = -J_\pm \sum_{\langle ij \rangle} \left[ S_i^+ S_j^- + S_i^- S_j^+ \right]$$

$$\mathcal{H}_0 = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z$$

Corrections to first order hopping process:




Modifies hopping matrix element of spinons

$$-J_\pm \to -J_\pm \left( 1 + \frac{J_\pm}{J_{zz}} \right)$$

# Worked Example: unfrustrated XXZ model

$$\epsilon_1 = \frac{J_{zz}}{2} - 6J_{\pm} + \mathcal{O}(J_{\pm}^2)$$

Second order corrections to energy

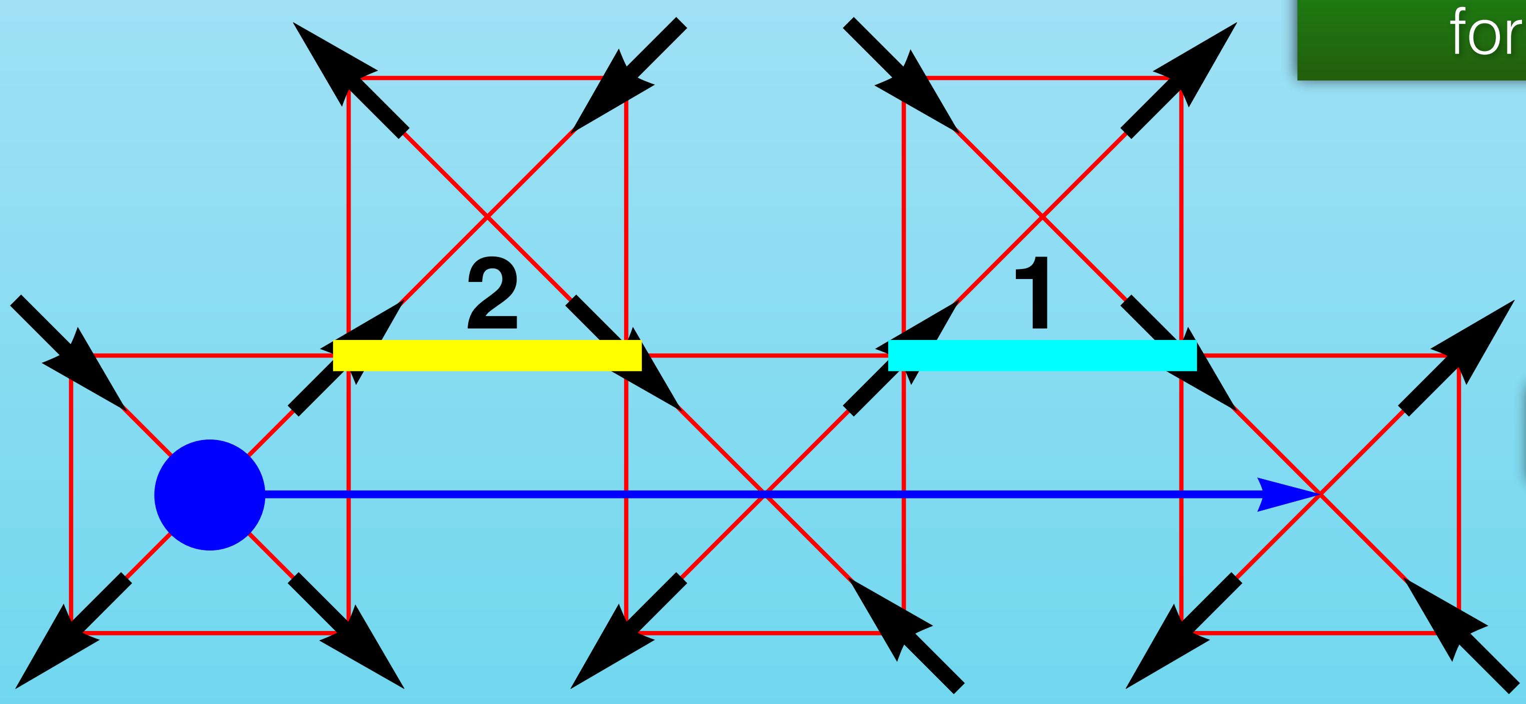
$$\mathcal{H}_{\text{eff}}^{(2)} = \mathcal{P}_M V \frac{1-\mathcal{P}_M}{\mathcal{H}_0} V \mathcal{P}_M$$

$$V = -J_{\pm} \sum_{\langle ij \rangle} [S_i^+ S_j^- + S_i^- S_j^+]$$

$$\mathcal{H}_0 = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z$$

Further neighbour hoppings

Introduces 24 further neighbour hoppings for every spinon (assuming ρ<<1)




Matrix element

$$-\frac{J_{\pm}^2}{J_{zz}}$$

# Worked Example: unfrustrated XXZ model

Equal weight superposition of all spinon states is still an eigenstate of effective Hamiltonian

$$|\phi_0\rangle = \sqrt{\frac{1}{\mathcal{N}_M}} \sum_{|\alpha\rangle \in \{|M\rangle\}} |\alpha\rangle$$

Energy:

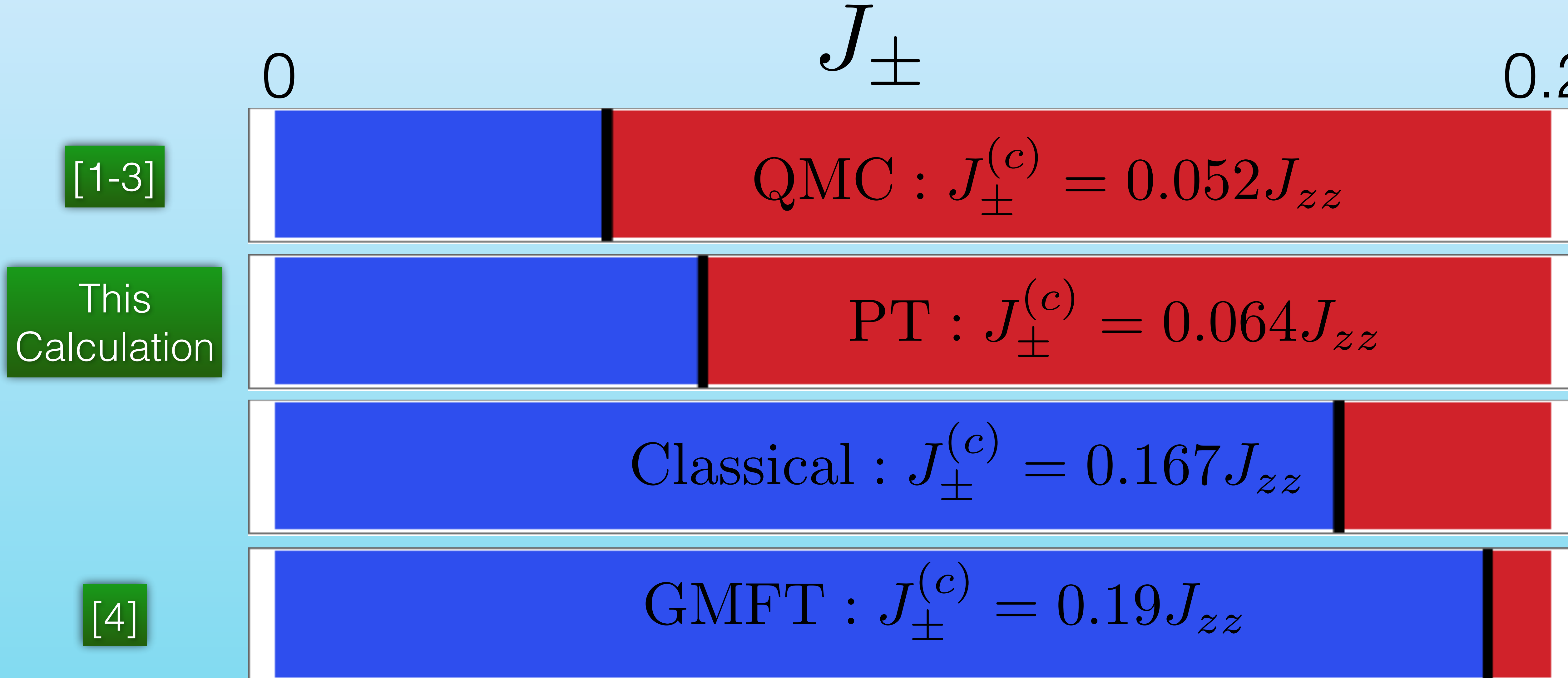
$$E(\rho) = E_0 - 4N_t \frac{J_\pm^2}{J_{zz}} + \rho N_t \left( \frac{J_{zz}}{2} - 6J_\pm - 28 \frac{J_\pm^2}{J_{zz}} \right) + \mathcal{O}(\rho^2, J_\pm^3)$$

Instability of ground state occurs when coefficient of ρ changes sign

$$J_\pm^{(c)} = \frac{1}{28} \left( \sqrt{23} - 3 \right) J_{zz} \approx 0.064 J_{zz}$$

# Comparison with other approaches

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





[1] Banerjee et al., PRL 100, 047208 (2008)
[2] Kato and Onoda, PRL 115, 077202 (2015)
[3] Lv et al., PRL 115, 037202 (2015)
[4] Savary and Balents, PRL 108, 037202 (2012)

# A more general model

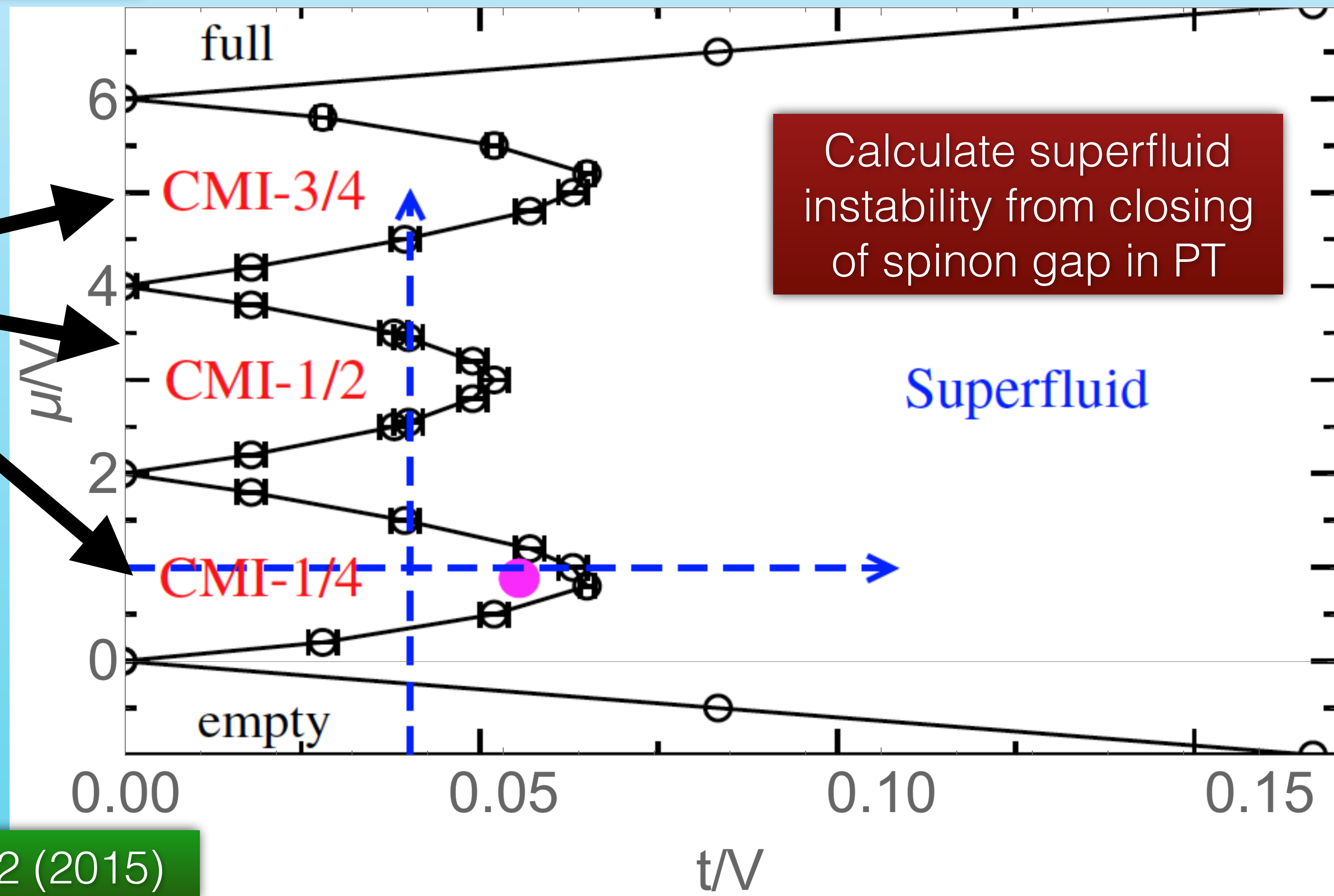
Quantum spin ice in a staggered field, coupling uniformly to $S^z$

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[ J_{zz} S_i^z S_j^z - J_{\pm} \left( S_i^+ S_j^- + S_i^- S_j^+ \right) \right] - h \sum_i S_i^z$$

Equivalently: hardcore bosons with nearest neighbour repulsion on the pyrochlore lattice

$$\mathcal{H}_b = \sum_{\langle ij \rangle} \left[ V n_i n_j - t \left( b_i^\dagger b_j + b_i b_j^\dagger \right) \right] - \mu \sum_i n_i$$

QMC: three separate Coulomb spin liquids with different filling factors




Calculate superfluid instability from closing of spinon gap in PT

Lv et al., PRL 115, 037202 (2015)

# A more general model

Quantum spin ice in a staggered field, coupling uniformly to $S^z$

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[ J_{zz} S_i^z S_j^s - J_{\pm} \left( S_i^+ S_j^- + S_i^- S_j^+ \right) \right] - h \sum_i S_i^z$$

Equivalently: hardcore bosons with nearest neighbour repulsion on the pyrochlore lattice

$$\mathcal{H}_b = \sum_{\langle ij \rangle} \left[ V n_i n_j - t \left( b_i^\dagger b_j + b_i b_j^\dagger \right) \right] - \mu \sum_i n_i$$

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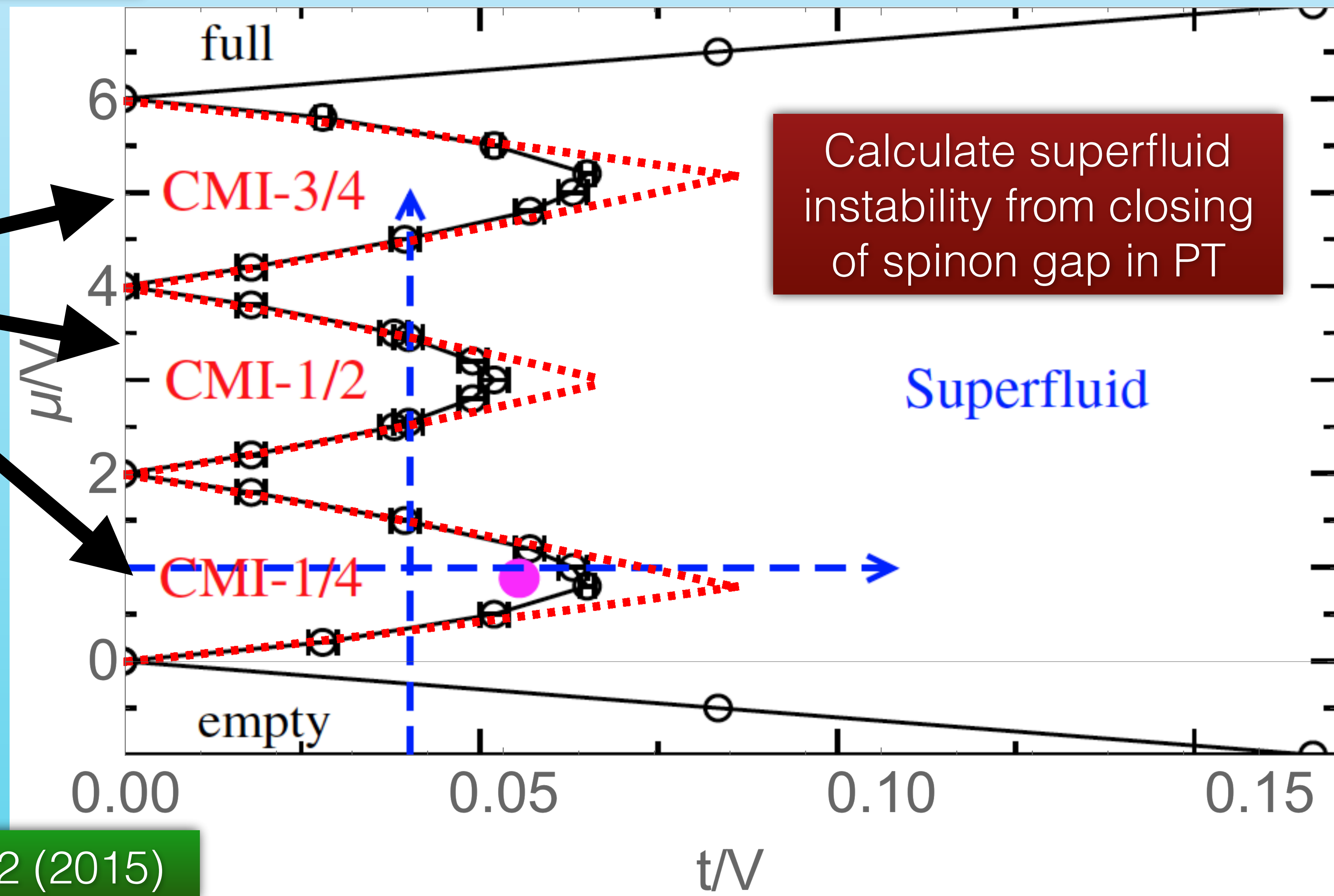
Calculate superfluid instability from closing of spinon gap in PT




Lv et al., PRL 115, 037202 (2015)

# Comparison with QMC simulations of spinon dynamics

arXiv:1707.00099

**Dynamics of topological excitations in a model quantum spin ice**

Chun-Jiong Huang,[1] Youjin Deng,[1] Yuan Wan,[2,3] and Zi Yang Meng[3,4]

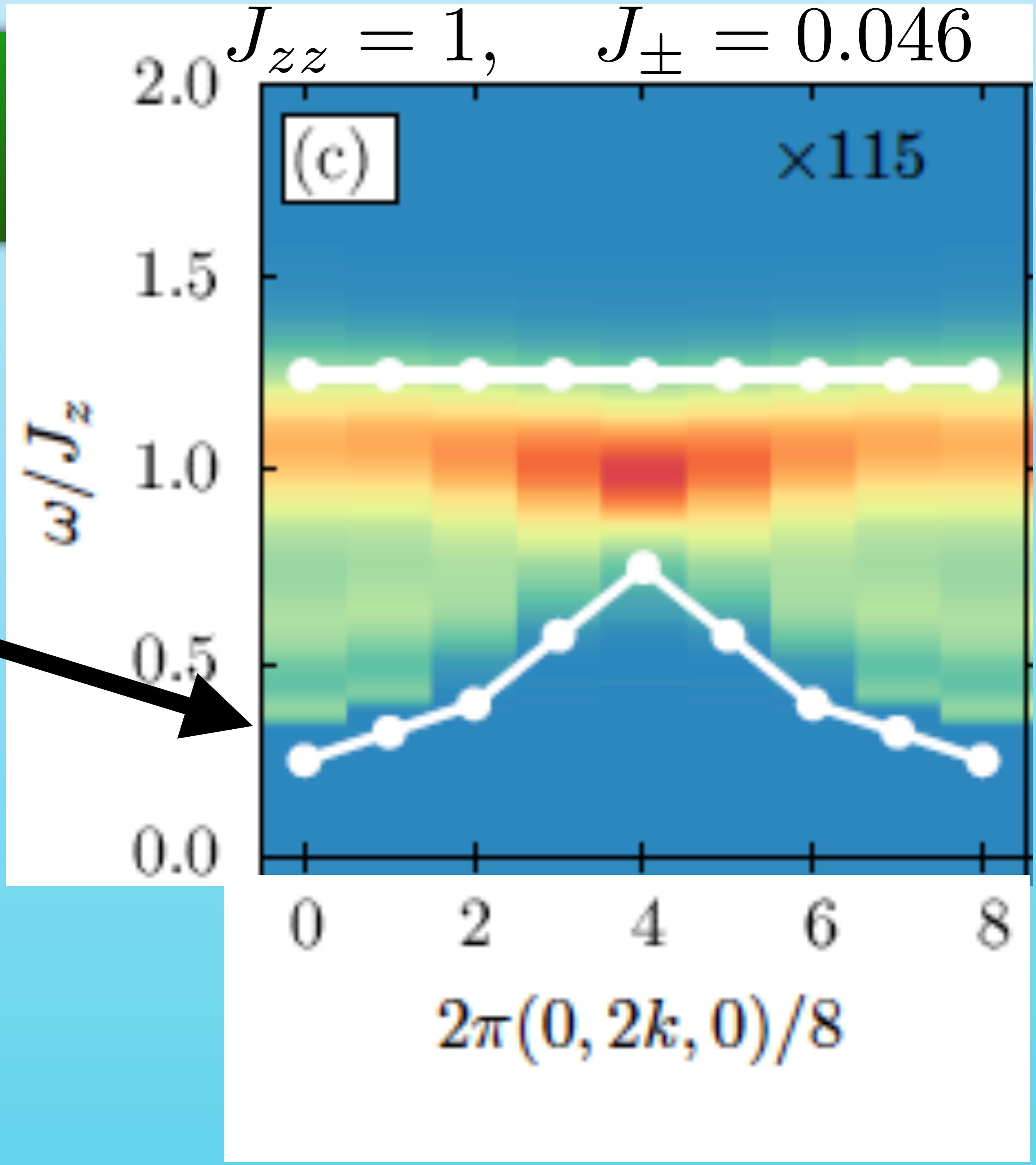
Monte Carlo simulation of two spinon continuum in dynamic structure factor

Minimum of lower continuum edge

$$2\Delta \approx 0.35 J_{zz}$$

cf. perturbation theory calculation

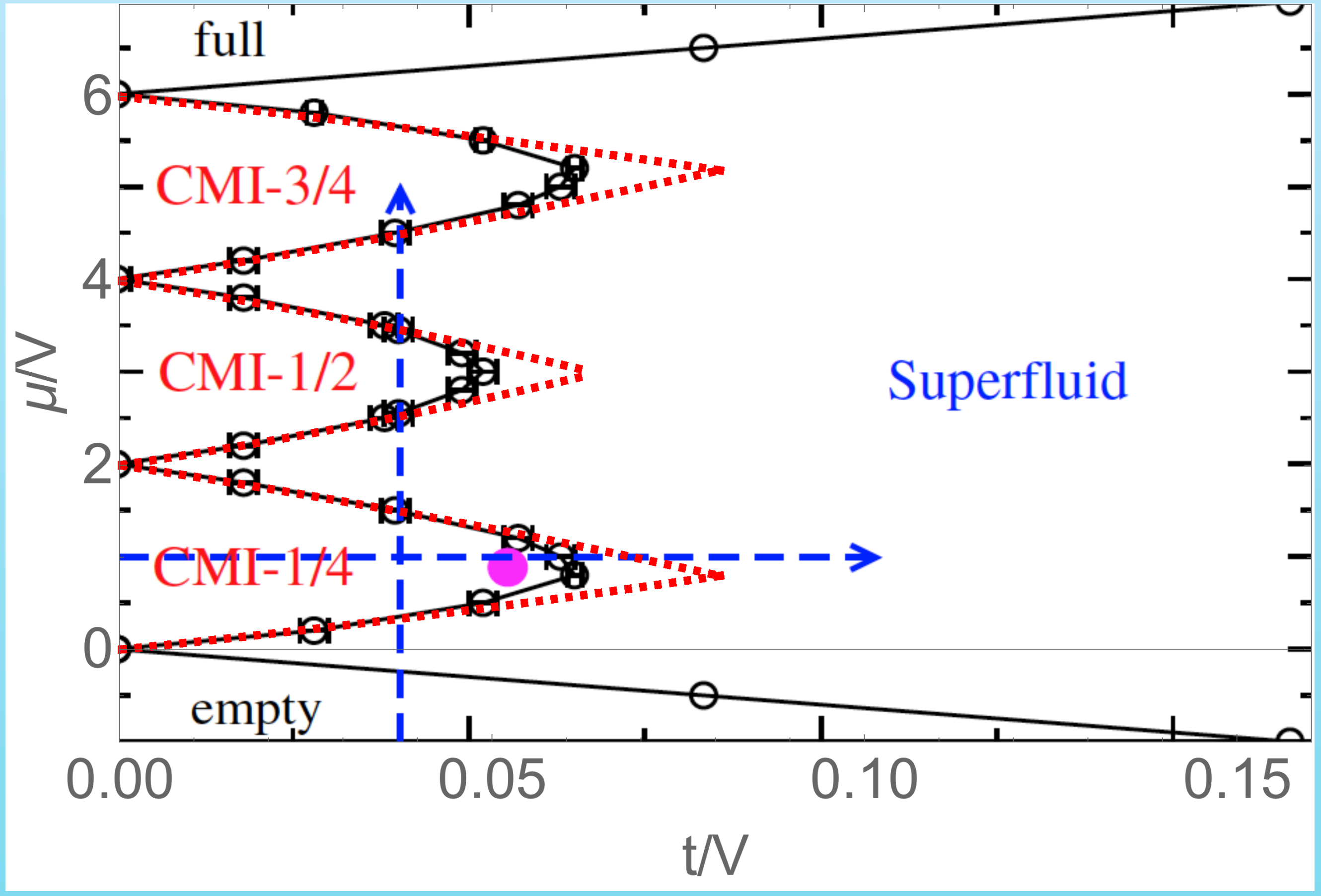
$$\approx 0.33 J_{zz}$$



# The story so far

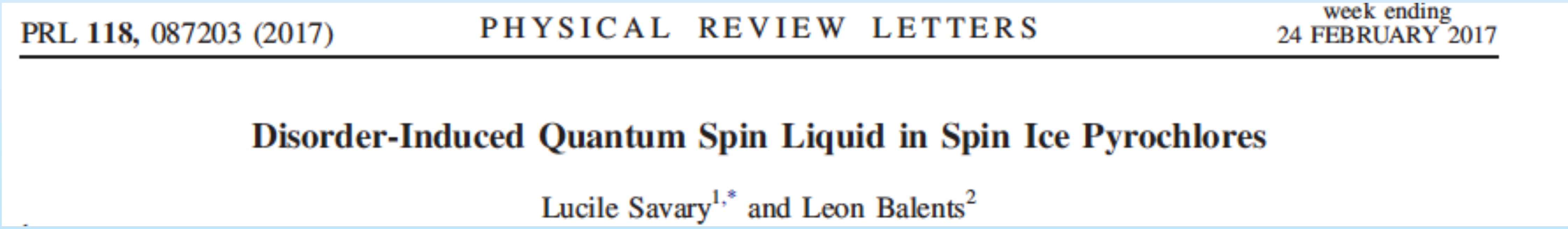
- We have illustrated a perturbation theory calculation of the point at which the U(1) QSL of quantum spin ice becomes unstable against spinon condensation

- The calculation gives excellent agreement with published QMC studies of the unfrustrated XXZ/hardcore boson model on the pyrochlore lattice




But the XXZ model is already quite well studied…. where else can we apply this?

# Random Transverse Field Ising Model


PRL 118, 087203 (2017) PHYSICAL REVIEW LETTERS week ending 24 FEBRUARY 2017

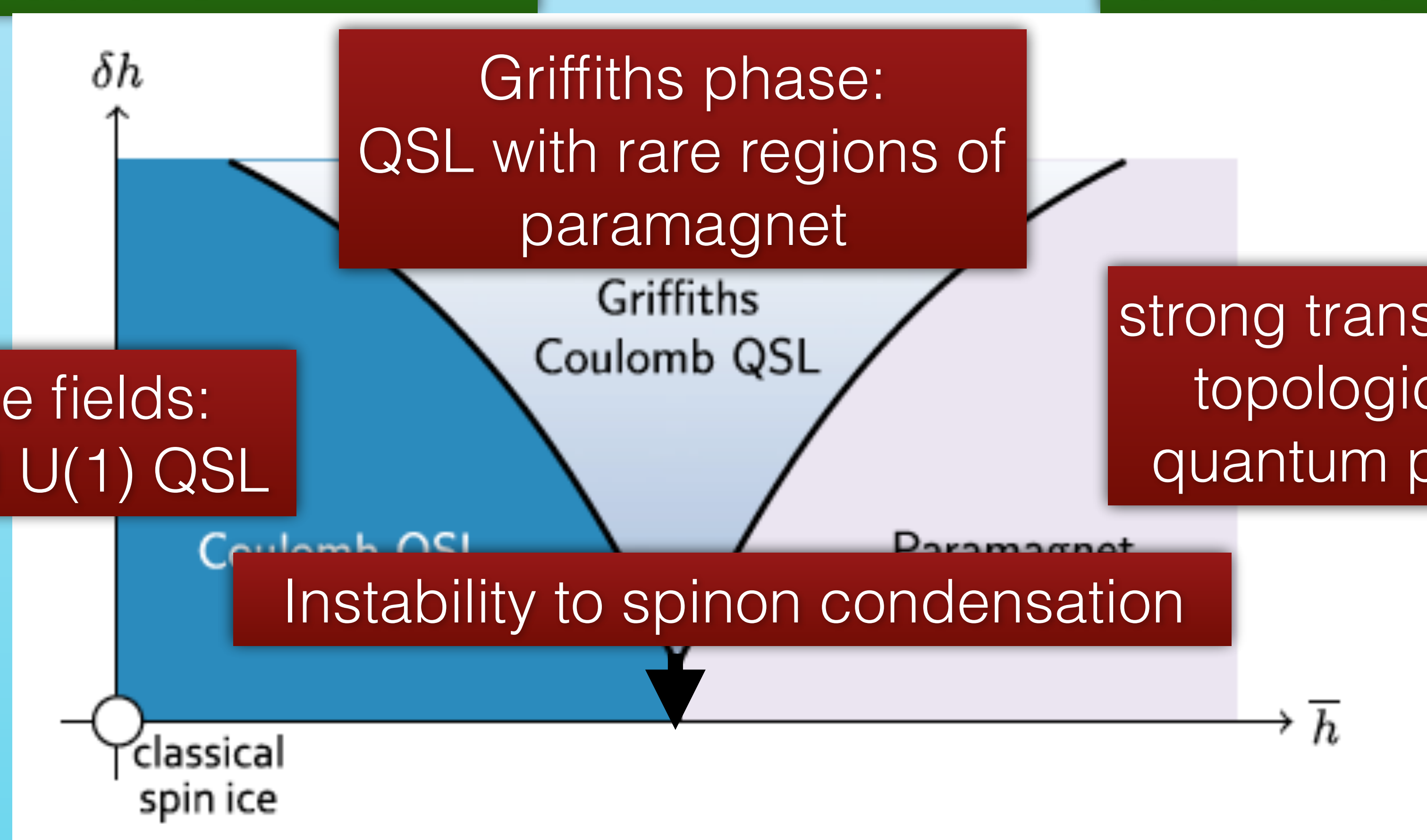
**Disorder-Induced Quantum Spin Liquid in Spin Ice Pyrochlores**

Lucile Savary[1,*] and Leon Balents[2]

$$\mathcal{H}_{\text{RTFIM}} = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z - \sum_i h_i S_i^x$$

spin ice-like exchange interactions

distribution of random fields $h_i$




Griffiths phase: QSL with rare regions of paramagnet

weak transverse fields: disorder induced U(1) QSL

strong transverse fields: topologically trivial quantum paramagnet

Instability to spinon condensation

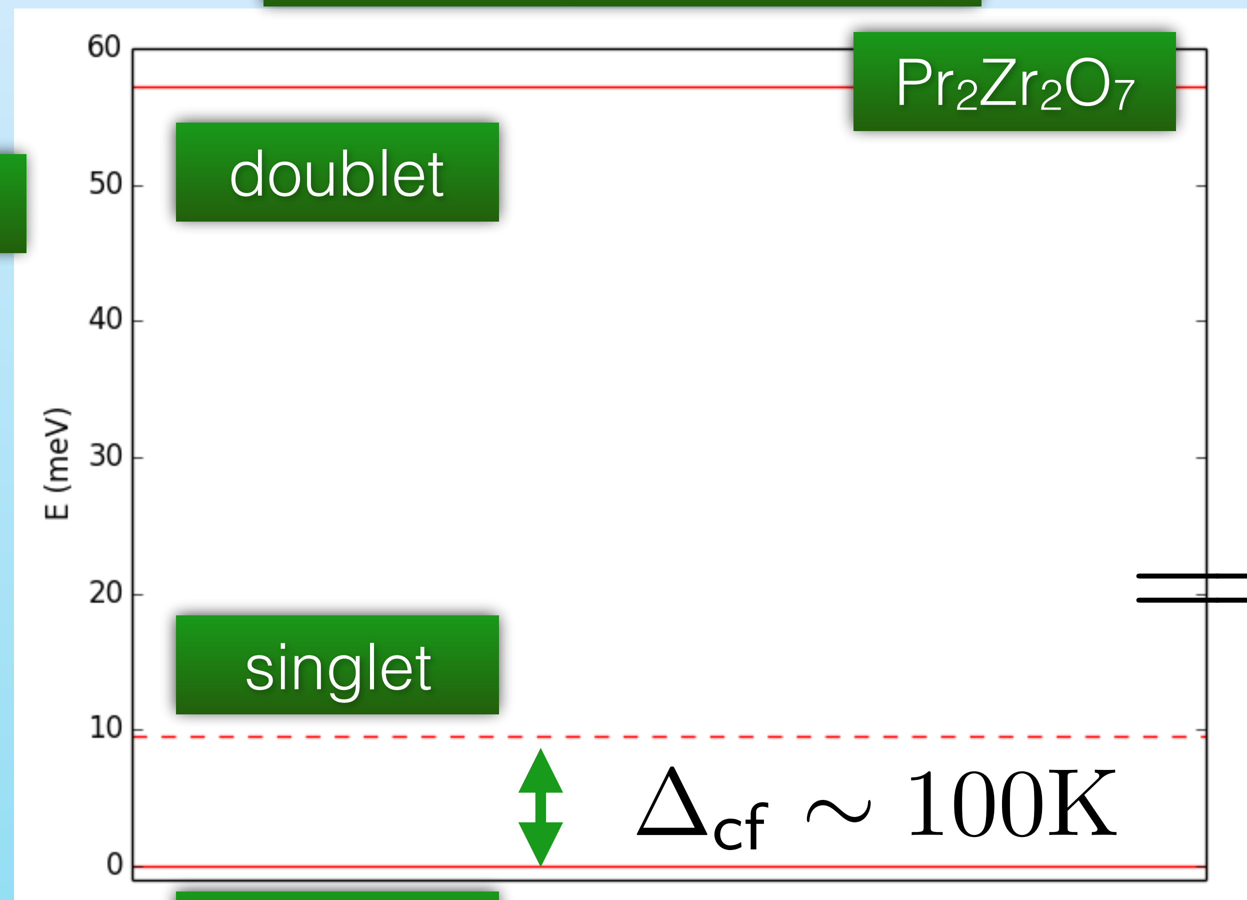
# Why is this model of interest?

Model for non-Kramers doublets in the presence of weak structural disorder

$Pr_2M_2O_7$ (M=Sn, Zr, Hf)

Kimura et al, Nat. Commun. **4** 1934, (2013)

Non-Kramers ions $Pr^{3+}$

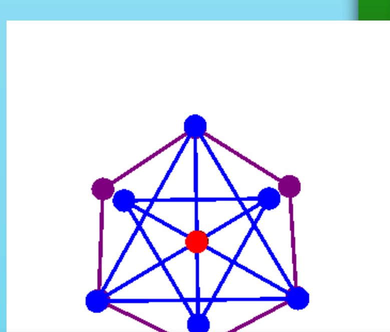



Crystal field ground state is a doublet with large gap to excited states

$\Longrightarrow$ Pseudospin-1/2 description

$$S_i^z = \uparrow, \downarrow$$

Degeneracy of ground doublet protected by lattice symmetry **not** time reversal

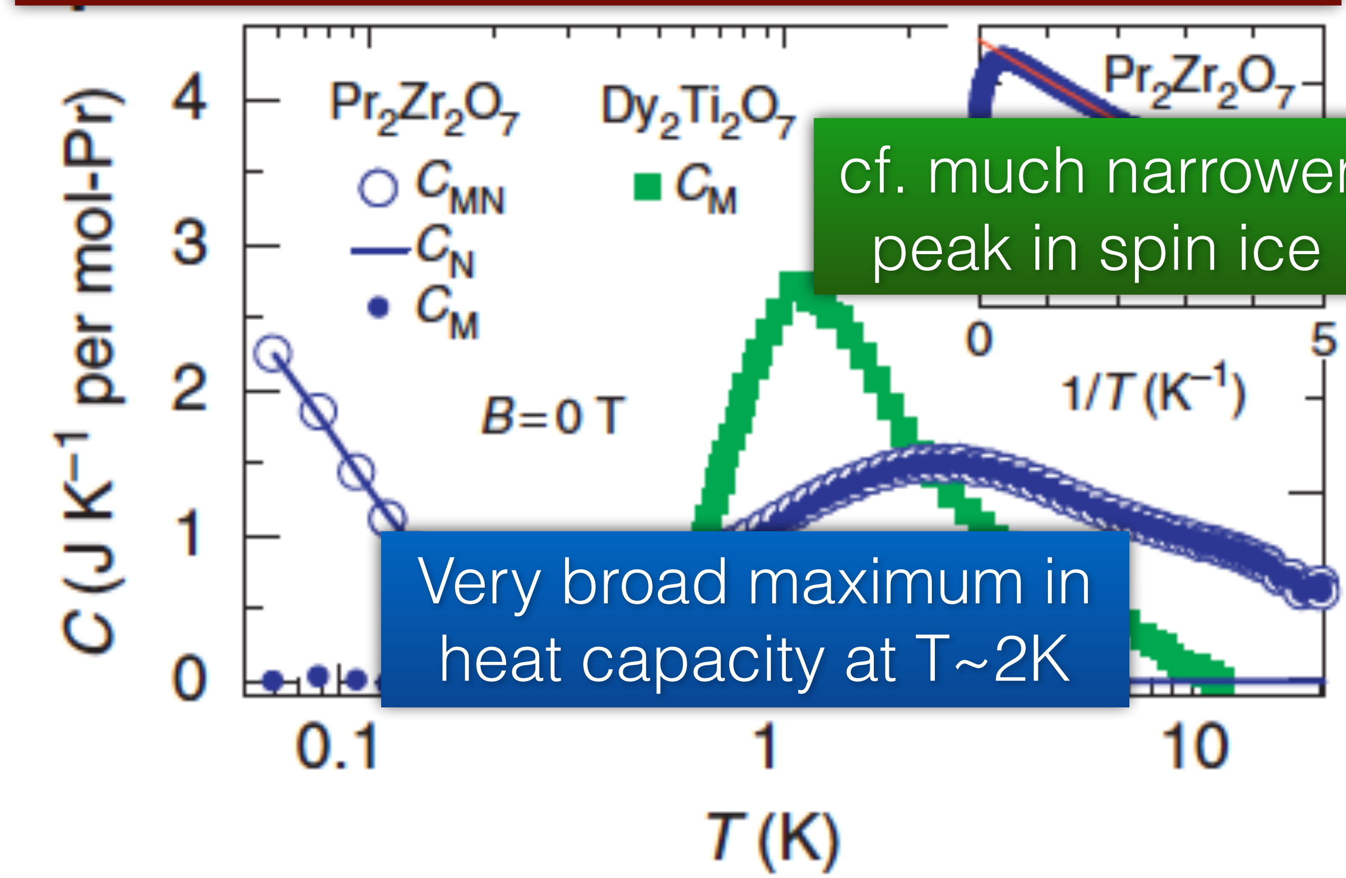


[111] view through a Pr site

Any local deviation from this trigonal symmetry lifts degeneracy of doublet $\Longrightarrow$ $-h_i S_i^x$

# $Pr_2Zr_2O_7$

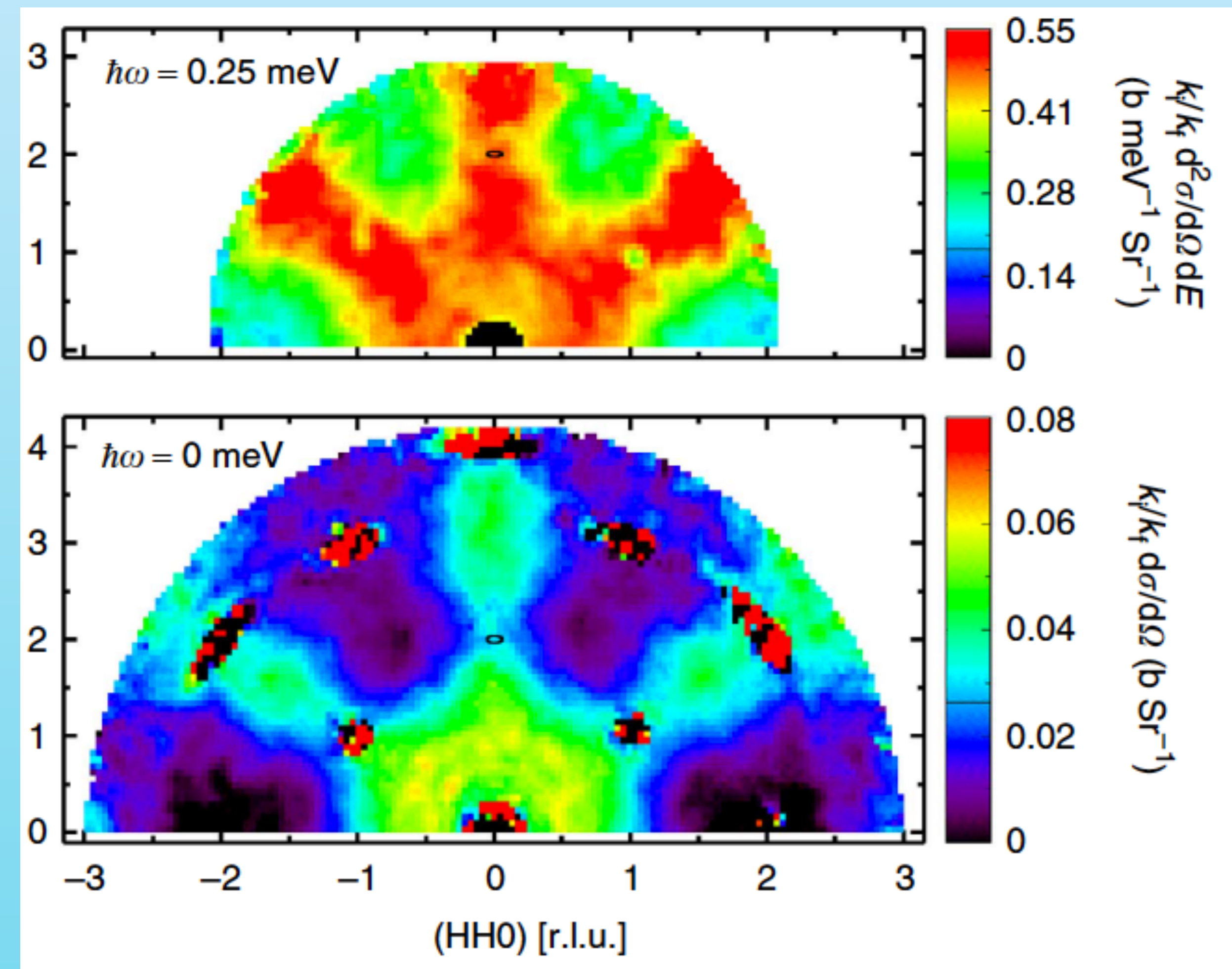
Kimura et al, Nat. Commun. **4**, 1934, (2013)



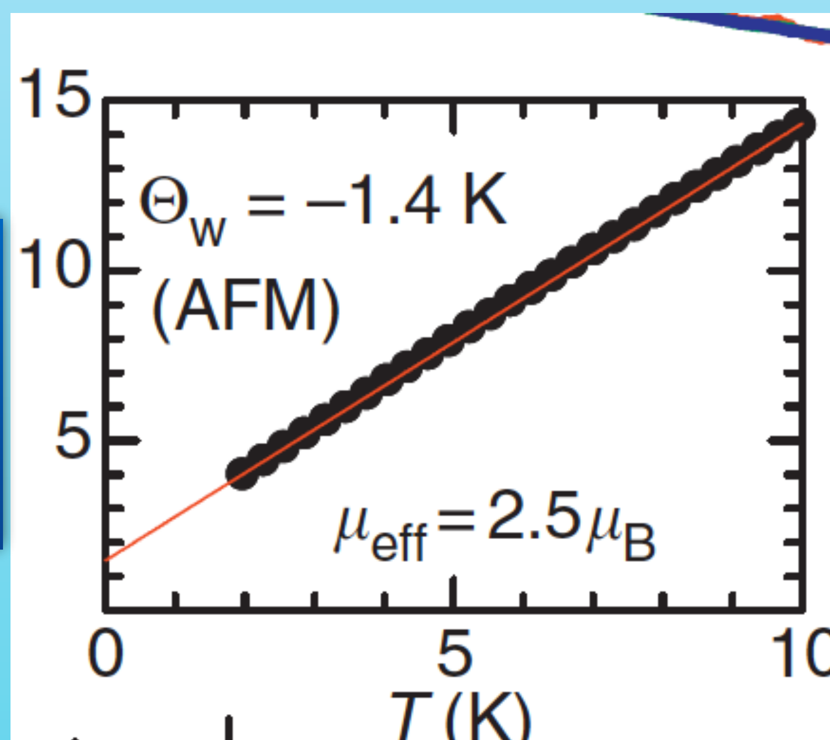

cf. much narrower peak in spin ice

Very broad maximum in heat capacity at T~2K

Spin ice like correlations with finite correlation length which increases with decreasing energy




Susceptibility shows AF Curie-Weiss temperature



# Distribution of transverse fields in $Pr_2Zr_2O_7$

PRL 118, 107206 (2017) PHYSICAL REVIEW LETTERS week ending 10 MARCH 2017

**Disordered Route to the Coulomb Quantum Spin Liquid: Random Transverse Fields on Spin Ice in $Pr_2Zr_2O_7$**

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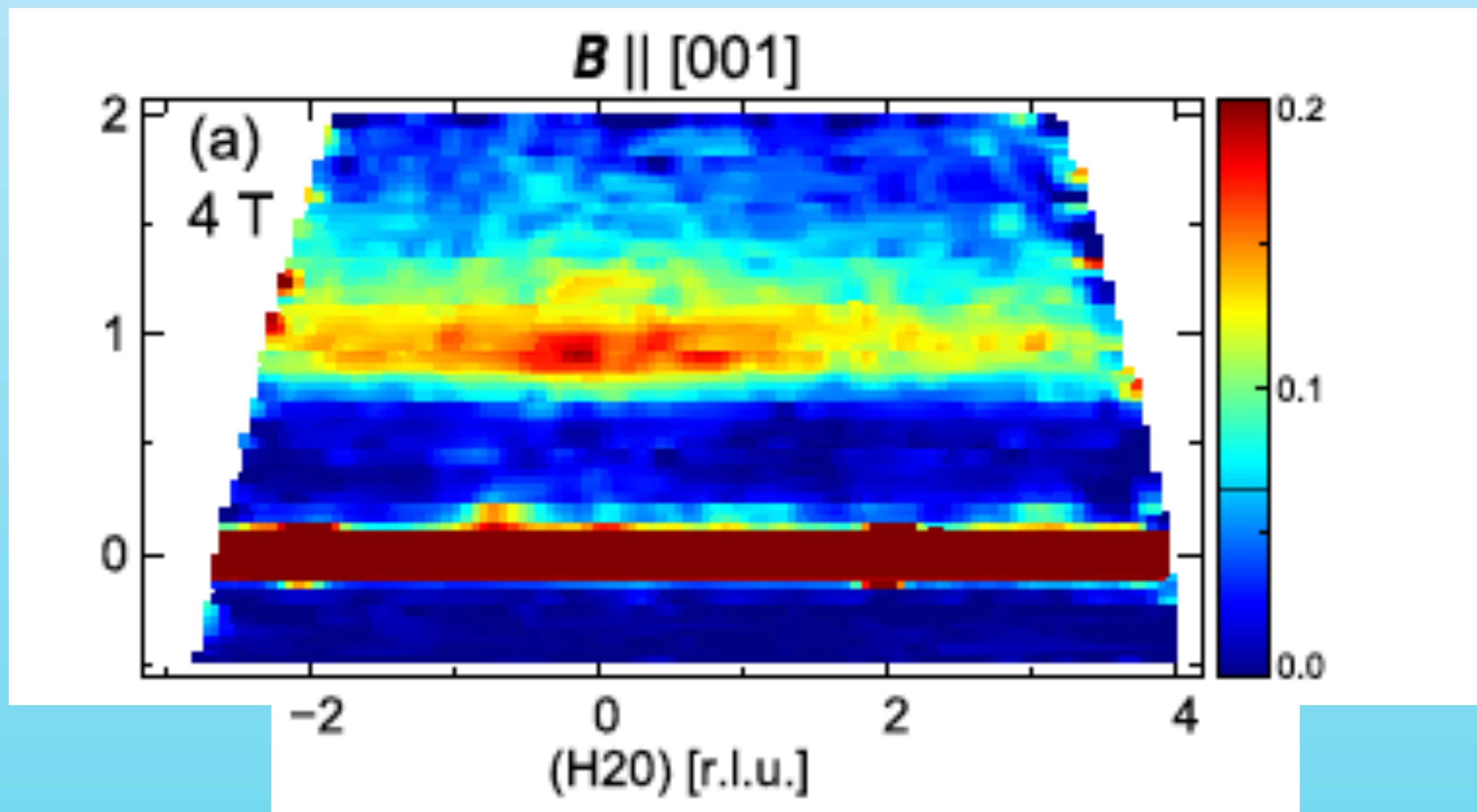
Inelastic neutron scattering in applied [100] field




Localized spin excitations with a broad distribution of gaps

# Distribution of transverse fields in $Pr_2Zr_2O_7$

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Inferring distribution of transverse fields from inelastic neutron scattering

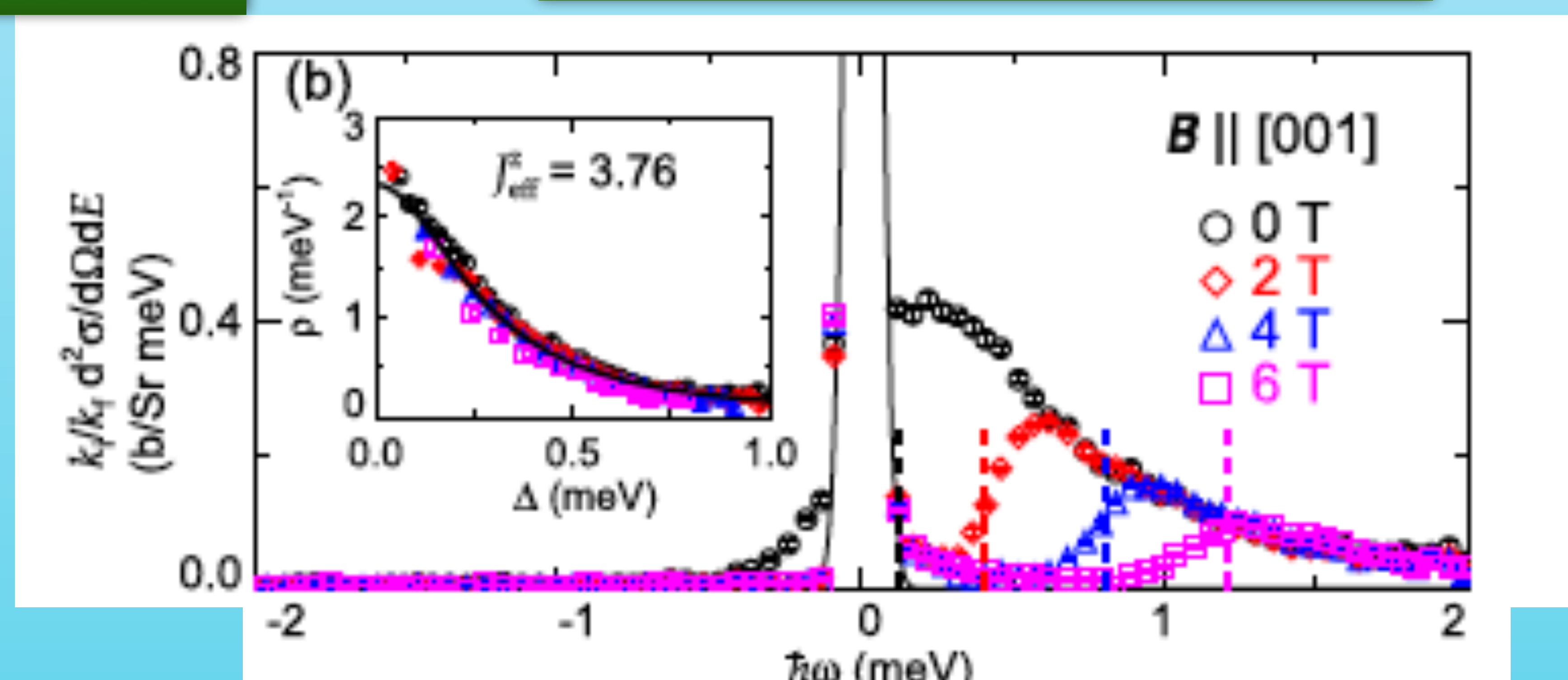
$$\mathcal{H}_{\mathrm{RTFIM+H}} = J_{\mathrm{zz}} \sum_{\langle ij \rangle} S_i^{\mathrm{z}} S_j^{\mathrm{z}} - \sum_i h_i S_i^x - g_z \mu_B \mathbf{H}_{\mathrm{ext}} \cdot \sum_i \hat{\mathbf{z}}_i S_i^{\mathrm{z}}$$

Find broad Lorentzian distribution of h

q integrated scattering intensity

$$p(h) = \frac{2\Gamma}{\pi} \frac{1}{h^2 + \Gamma^2}$$

$$\Gamma = 0.54\mathrm{meV}$$



# Is $Pr_2Zr_2O_7$ a disorder induced QSL?

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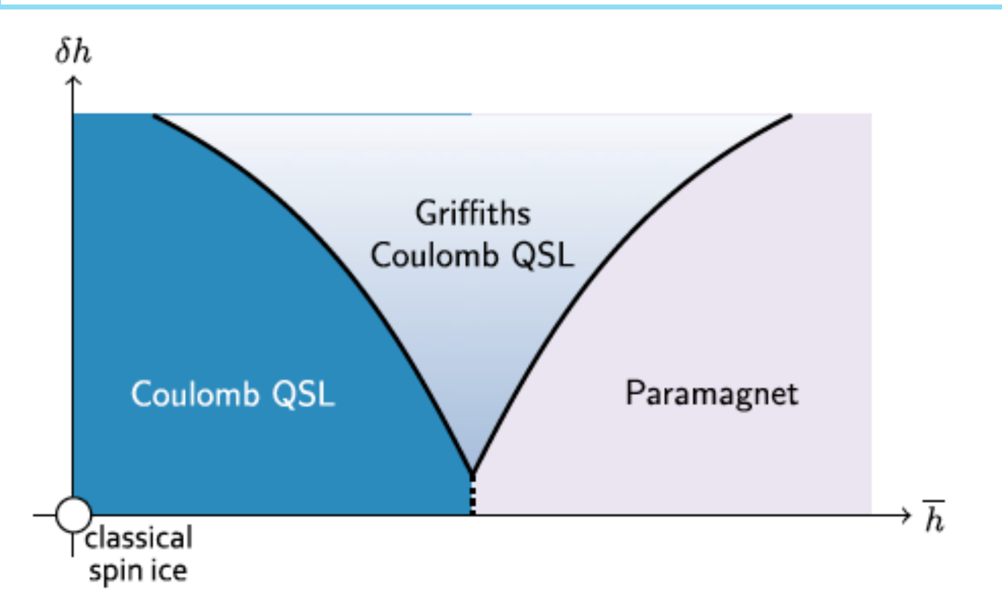
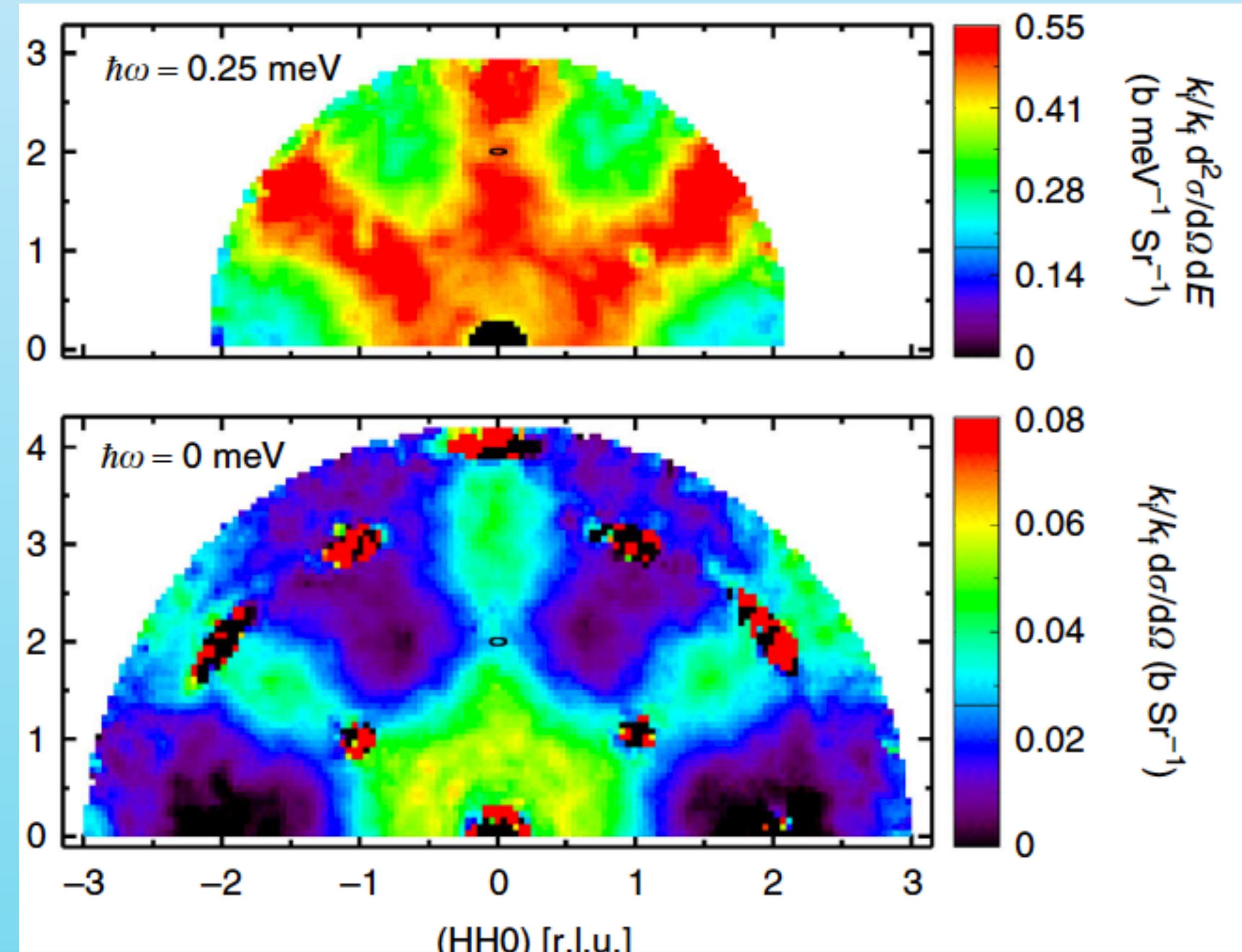
Suffice it to say that $\mathcal{H}$, which contains only the longitudinal Ising interactions, provides an acceptable account of the data.

QSI is expected to be stable to all local perturbations, despite being gapless [2]. A recent theoretical study of $\mathcal{H}$ shows random transverse fields induce quantum entanglement and two distinct QSLs [5]. The gapless nature of the spectrum [Fig. 1(b)] and the spin-ice-like correlations reported here (Fig. 3) and in previous studies [4] preclude a trivial paramagnet and point to a disorder induced QSL in PZO.

Given the importance of random transverse fields that we

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To answer this from a theory point of view we want to know two things

1) What is the stability regime of the U(1) QSL in the random transverse field Ising model on the pyrochlore lattice?

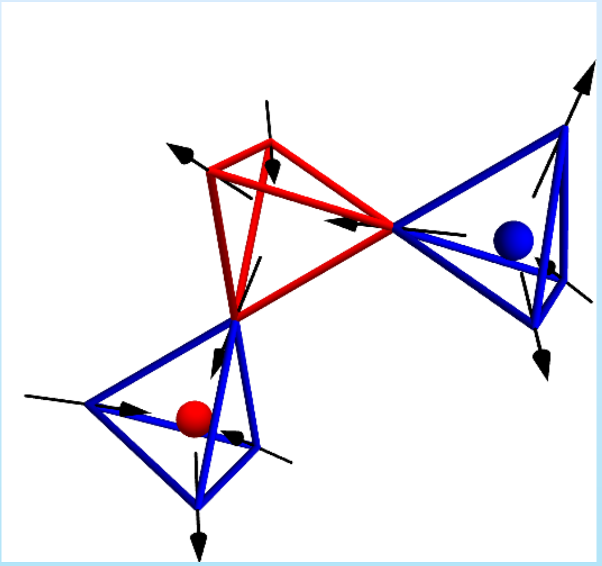
Perturbation theory calculation is well suited to answer this:
-constructed in real space not momentum space, doesn't rely on translational symmetry
-by considering state of M spinons 1<<M<<Nt have "self-averaging" of spinon environments, and obtain result which depends only on average and variance of h

2) What is the strength of interaction $J_{zz}$ and distribution of transverse field $p(h_i)$ in currently studied samples of $Pr_2Zr_2O_7$?

Can find out by comparing thermodynamic data with Numerical Linked Cluster (NLC) Calculations

# Stability criterion for U(1) QSL?

$$\mathcal{H}_{\text{RTFIM}} = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z - \sum_i h_i S_i^x$$



Consider once again a state of M spinons

$$1 << M << N_t$$

allows averaging over spinon environments

neglect spinon-spinon interactions

Effective Hamiltonian from perturbation theory

$$\mathcal{H}_{\text{eff}}^{(M)} = E_0^{\text{cl}} + M\frac{J_{zz}}{2} + \mathcal{H}_1^{(M)} + \mathcal{H}_2^{(M)}$$

$$\mathcal{H}_1^{(M)} = \mathcal{P}_M V \mathcal{P}_M$$

$$\mathcal{H}_2^{(M)} = -\mathcal{P}_M V \frac{1 - \mathcal{P}_M}{\mathcal{H}_0 - (E_0^{\text{cl}} + M\frac{J_{zz}}{2})} V \mathcal{P}_M$$

$$\mathcal{H}_0 = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z$$

$$V = -\sum_i h_i S_i^x$$

# First order perturbation theory

$$\mathcal{H}_1^{(M)} = \mathcal{P}_M V \mathcal{P}_M$$

Column sum:

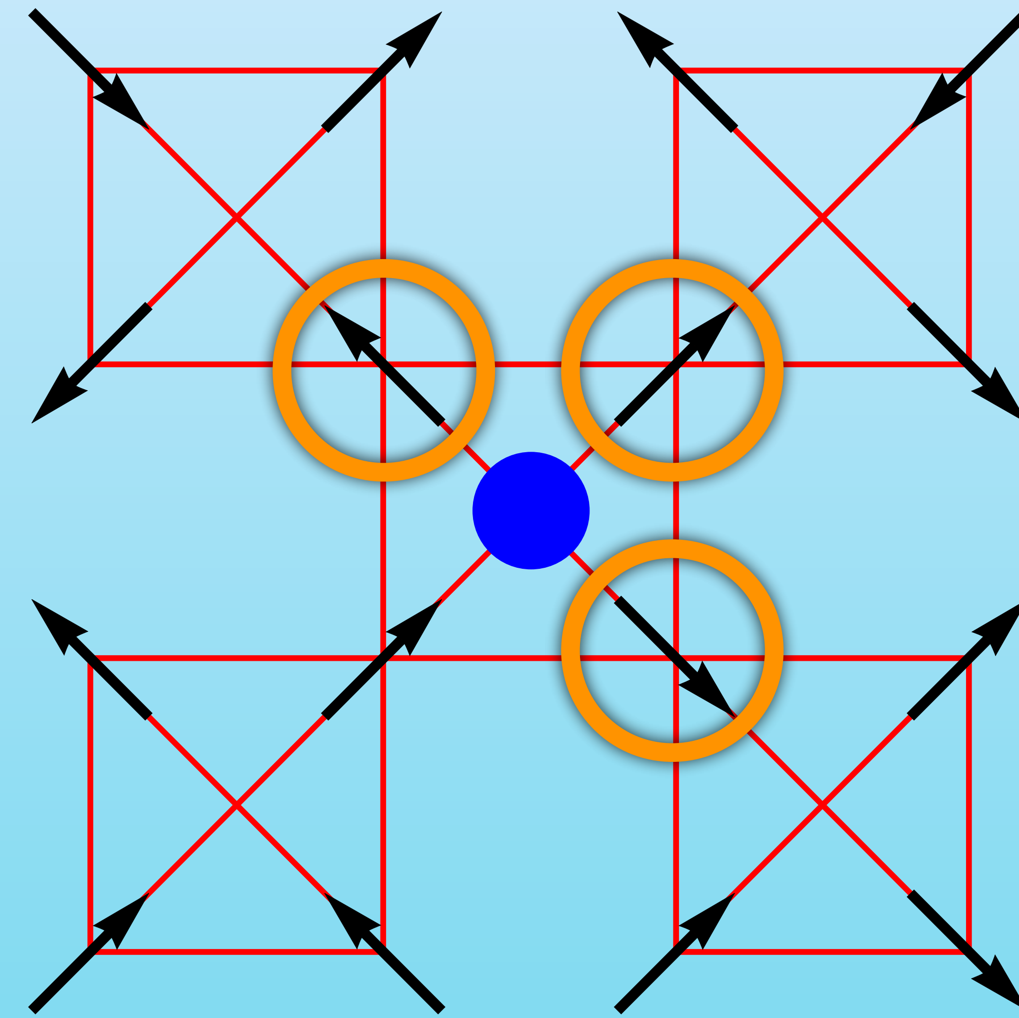
$$\sum_\alpha \left(\mathcal{H}_1^{(M)}\right)_{\alpha\beta} = -\frac{1}{2}\sum_{i\in\text{flippable}} h_i$$

average of h

For 1<<M<<$N_t$

$$= -3M\frac{\overline{h}}{2}$$

Every spinon surrounded by 3 flippable spins



Equal weight superposition of all M spinon states is a good eigenstate for 1<<M<<$N_t$

$$|\phi_M\rangle = \frac{1}{\sqrt{\mathcal{N}_M}}\sum_{|\alpha\rangle\in|\{M\}\rangle}|\alpha\rangle$$

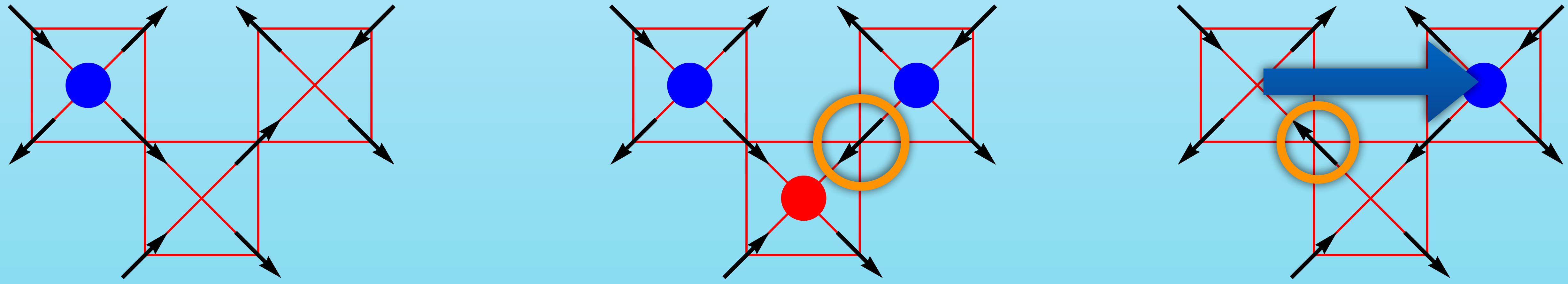
# Second order perturbation theory

$$\mathcal{H}_2^{(M)} = -\mathcal{P}_M V \frac{1-\mathcal{P}_M}{\mathcal{H}_0 - (E_0^{\text{cl}} + M\frac{J_{zz}}{2})} V \mathcal{P}_M$$

Diagonal contributions from flipping same spin twice

$$\left(\mathcal{H}_2^{(M)}\right)_{\alpha\alpha} = -\frac{N_t}{2J_{zz}}\overline{h^2} + \frac{3M}{8J_{zz}}\overline{h^2}$$

Second order spinon hopping



Possible on 6 nearby bonds for every spinon ($M << N_t$)

Matrix element $-\frac{h_k h_l}{4J}$

# Second order perturbation theory

Lowest energy state for M spinons

$$|\phi_M\rangle = \frac{1}{\sqrt{\mathcal{N}_M}} \sum_{|\alpha\rangle \in |\{M\}\rangle} |\alpha\rangle$$

$$E(M) = E_0^{\text{cl}} - \frac{N_t}{2J_{zz}}\overline{h^2} + M\left(\frac{J_{zz}}{2} - \frac{3\overline{h}}{2} + \frac{3\overline{h^2}}{8} - \frac{3\overline{h_k h_l}}{2}\right)$$

nearest neighbour correlation function of random fields

Instability determined by coefficient of M

$$\frac{J_{zz}}{2} - \frac{3\overline{h}}{2} + \frac{3\overline{h^2}}{8} - \frac{3\overline{h_k h_l}}{2} = 0$$

Take uncorrelated case

$$\overline{h_k h_l} = \overline{h}^2 \qquad \delta h = \sqrt{\overline{h^2} - \overline{h}^2}$$

$$\frac{J_{zz}}{2} - \frac{3\overline{h}}{2} + \frac{7\delta h^2 - 5\overline{h}^2}{8J} = 0$$

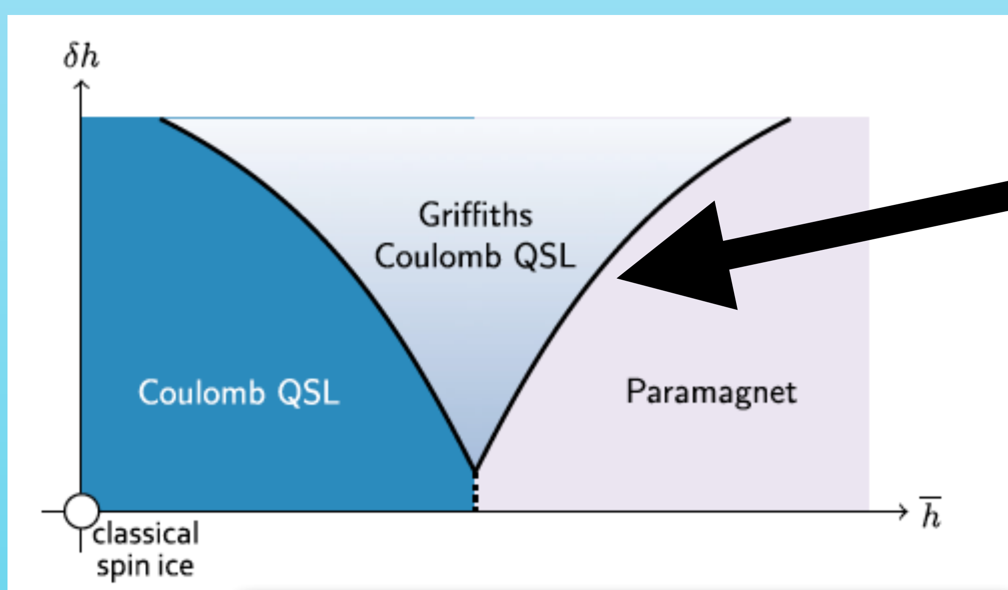
# Stability regime of U(1) QSL

**Stability criterion in terms of average and standard deviation of distribution of transverse fields**

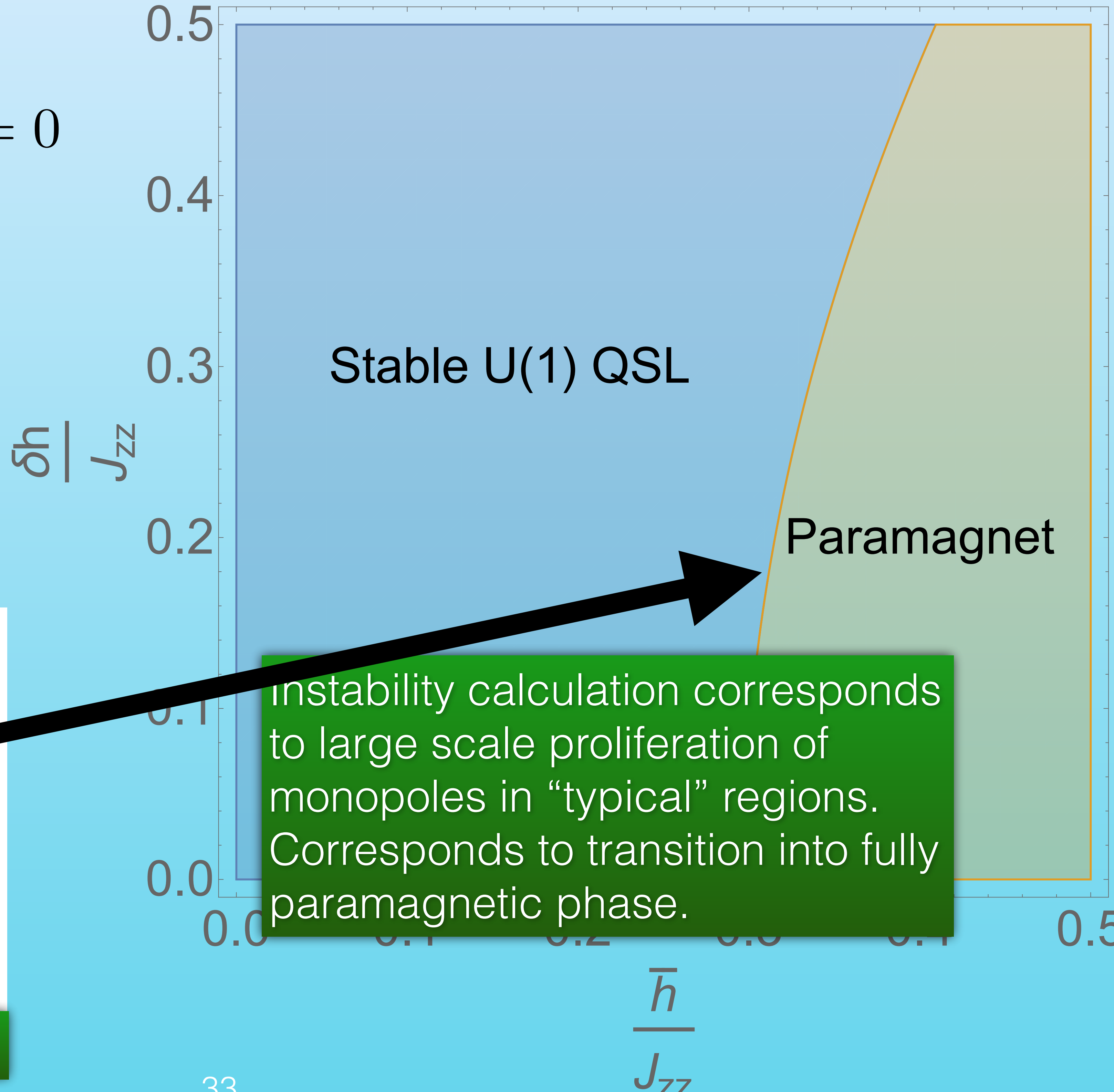
$$\frac{J_{zz}}{2} - \frac{3\overline{h}}{2} + \frac{7\delta h^2 - 5\overline{h}^2}{8J} = 0$$

$$\delta h = \sqrt{\overline{h^2} - \overline{h}^2}$$

cf. schematic Savary-Balents phase diagram




PRL **108**, 087302 (2017)




Instability calculation corresponds to large scale proliferation of monopoles in "typical" regions. Corresponds to transition into fully paramagnetic phase.

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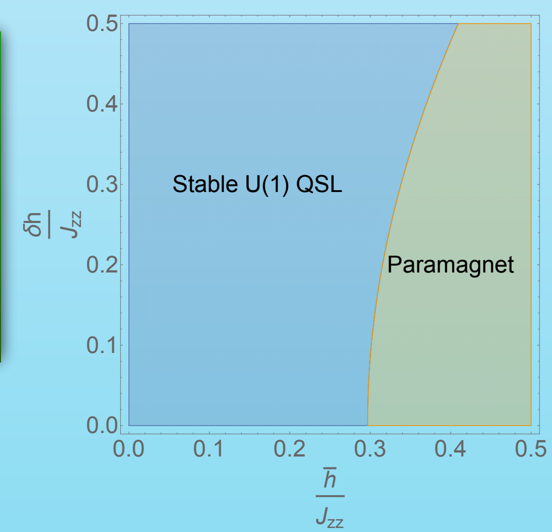



$$\frac{J_{zz}}{2} - 3\frac{\overline{h}}{2} + \frac{7\delta h^2 - 5\overline{h}^2}{8J} > 0$$

2) What is the strength of interaction $J_{zz}$ and distribution of transverse field $p(h_i)$ in currently studied samples of $Pr_2Zr_2O_7$?

Can find out by comparing thermodynamic data with Numerical Linked Cluster (NLC) Calculations

# Modelling $Pr_2Zr_2O_7$

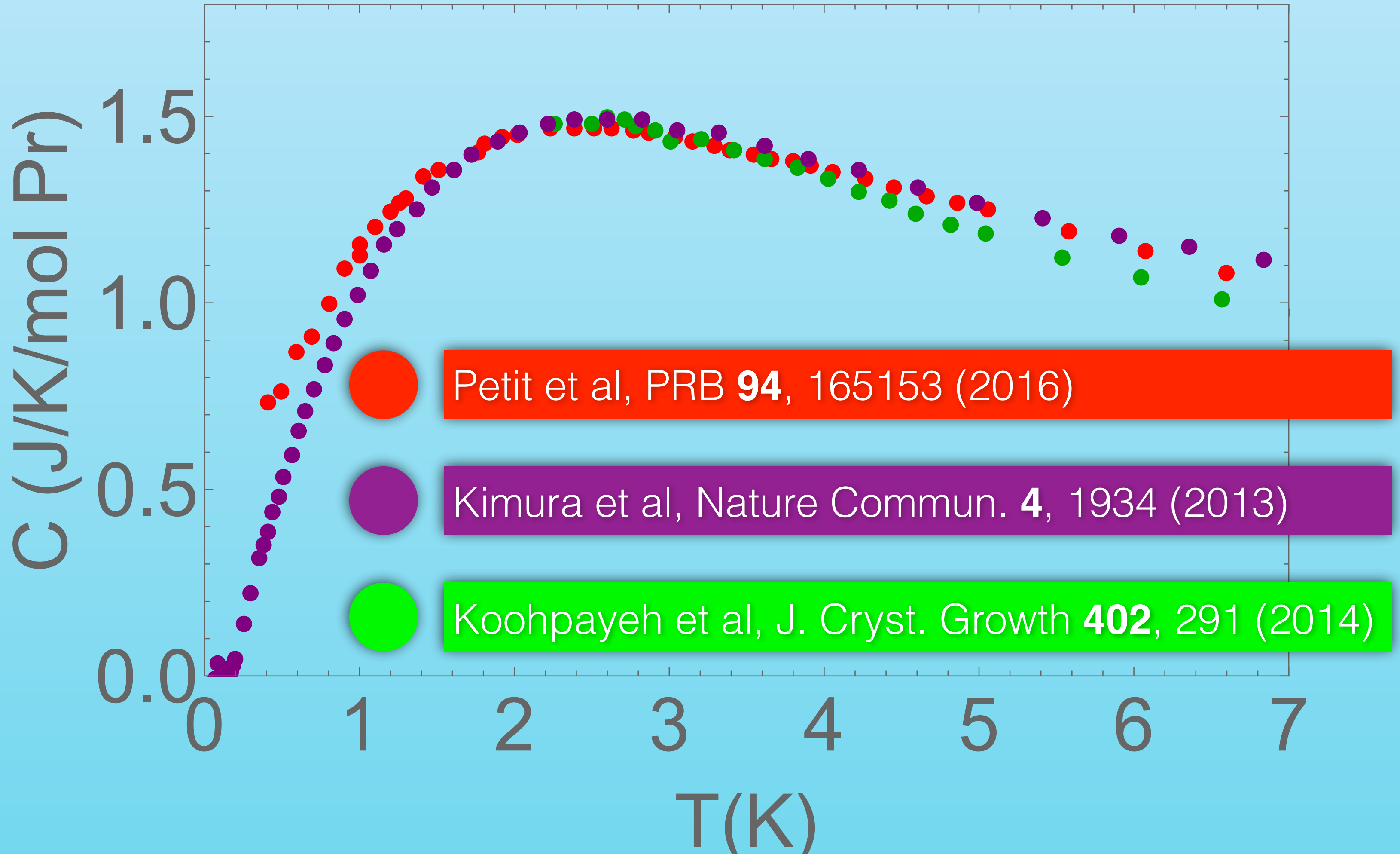
Need parameterisation of interactions

$$\mathcal{H}_{\text{RTFIM}} = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z - \sum_i h_i S_i^x$$

Strategy:
1) assume Lorentzian distribution of transverse fields

$$p(h) = \frac{2\Gamma}{\pi} \frac{1}{h^2 + \Gamma^2}$$

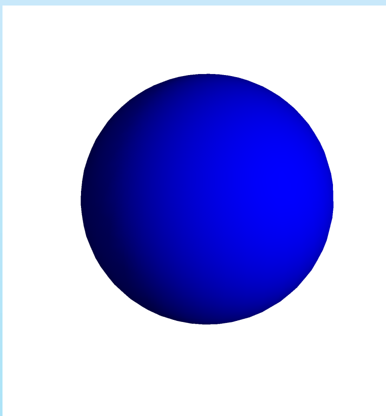
2) Fit available thermodynamic data to Numerical Linked Cluster (NLC) expansion of Hamiltonian with $J_{zz}$ and $\Gamma$ as adjustable parameters



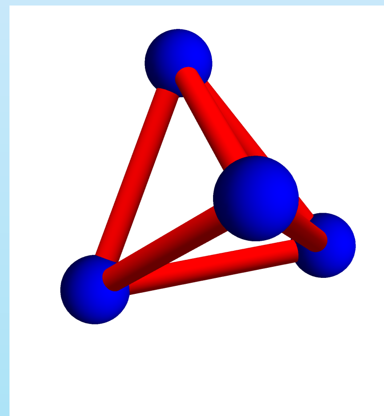

3) Compare back to high energy scattering data

# Numerical Linked Cluster (NLC) calculations

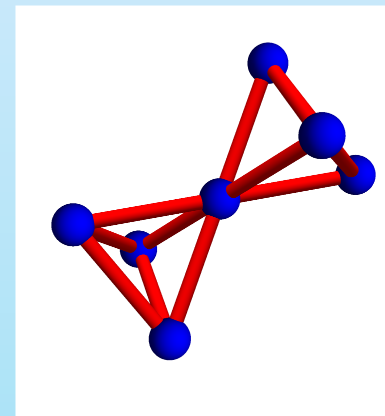
Estimating quantities in the thermodynamic limit from a series of exact diagonalizations of small clusters [1]



$c_0$



$c_1$



$c_2$

(….)

$c_n$

Estimate of extensive quantity O per site in terms of cluster multiplicities L and weights W

$$\frac{\langle \mathcal{O} \rangle}{N} = \sum_n L_n W_n; \qquad W_n = \langle O \rangle_{c_n} - \sum_{s \subset n} W_s$$

Disorder averages can be taken term by term in expansion

$$\overline{W}_n = \overline{\langle O \rangle}_{c_n} - \sum_{s \subset n} \overline{W}_s$$

[1] Rigol et al, PRL **97**, 187202 (2006); [2] Tang et al, PRB **91**, 174413 (2015)

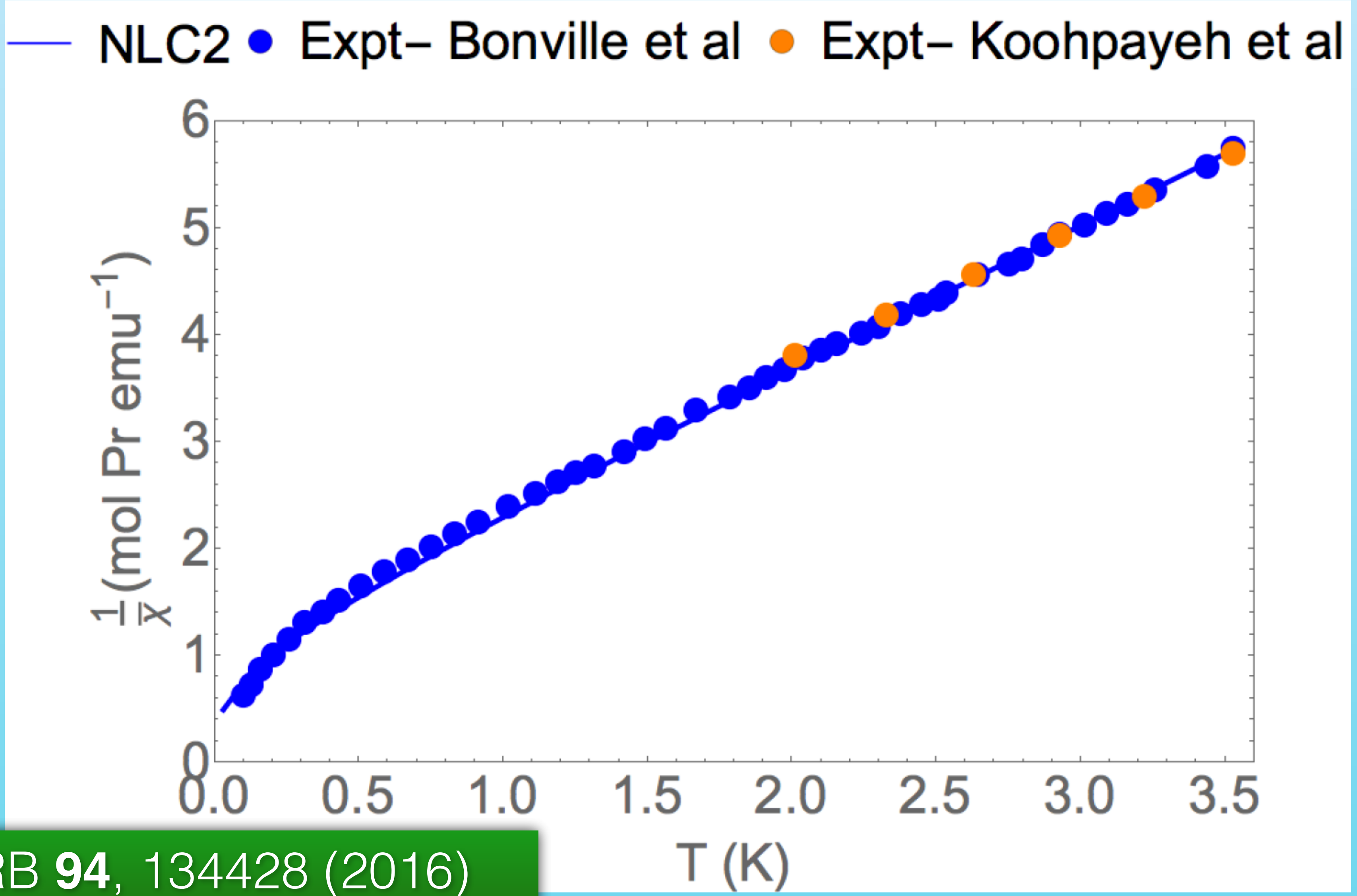
# NLC description of thermodynamics in $Pr_2Zr_2O_7$

Reasonable description of thermodynamics obtained with parameterisation:

$$\mathcal{H}_{\mathrm{RTFIM}} = J_{zz} \sum_{\langle ij \rangle} S_i^z S_j^z - \sum_i h_i S_i^x$$

$$p(h) = \frac{2\Gamma}{\pi} \frac{1}{h^2 + \Gamma^2} \qquad \Gamma = 0.38\mathrm{meV} \quad J_{zz} = 0.08\mathrm{meV} \quad g_z = 4.9$$

Inverse susceptibility




Effective $T_{CW}$<0 despite spin-ice like $J_{zz}$>0

Captures downturn at low temperature seen in experiment

Data from [1] Bonville et al, PRB **94**, 134428 (2016)
[2] Koohpayeh et al, J. Cryst. Growth **402**, 291 (2014)

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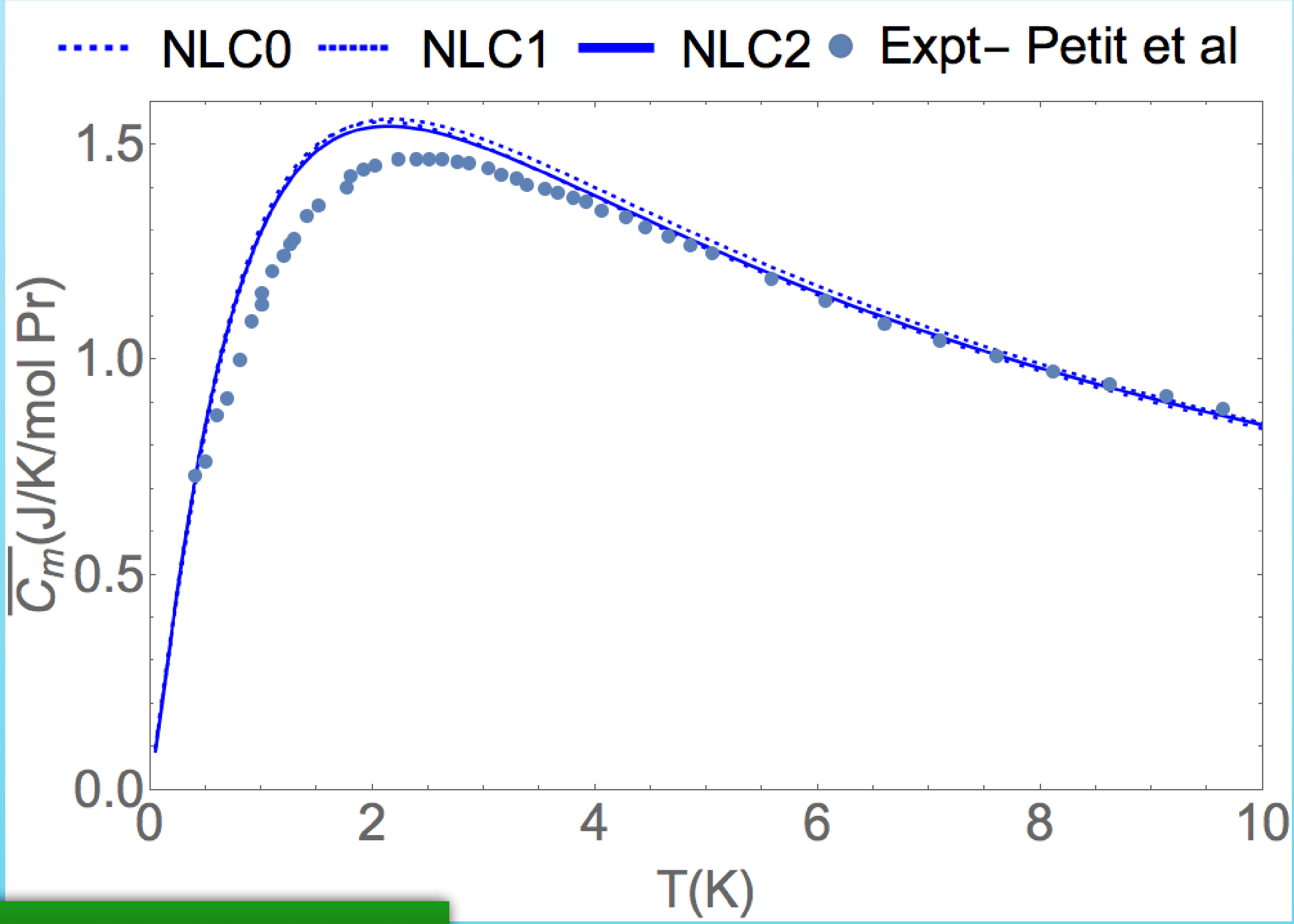
Heat capacity

Broad maximum at T~2K

Accurate descrption of high T tail of heat capacity

Overestimates height of maximum




Data from [1] Petit et al, PRB **94**, 165153 (2016)

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$$\mathcal{H}_{\mathsf{RTFIM+H}} = J_{\mathsf{zz}} \sum_{\langle ij \rangle} S_i^{\mathsf{z}} S_j^{\mathsf{z}} - \sum_i h_i S_i^x - g_z \mu_B \mathbf{H}_{\mathsf{ext}} \cdot \sum_i \hat{\mathbf{z}}_i S_i^{\mathsf{z}}$$

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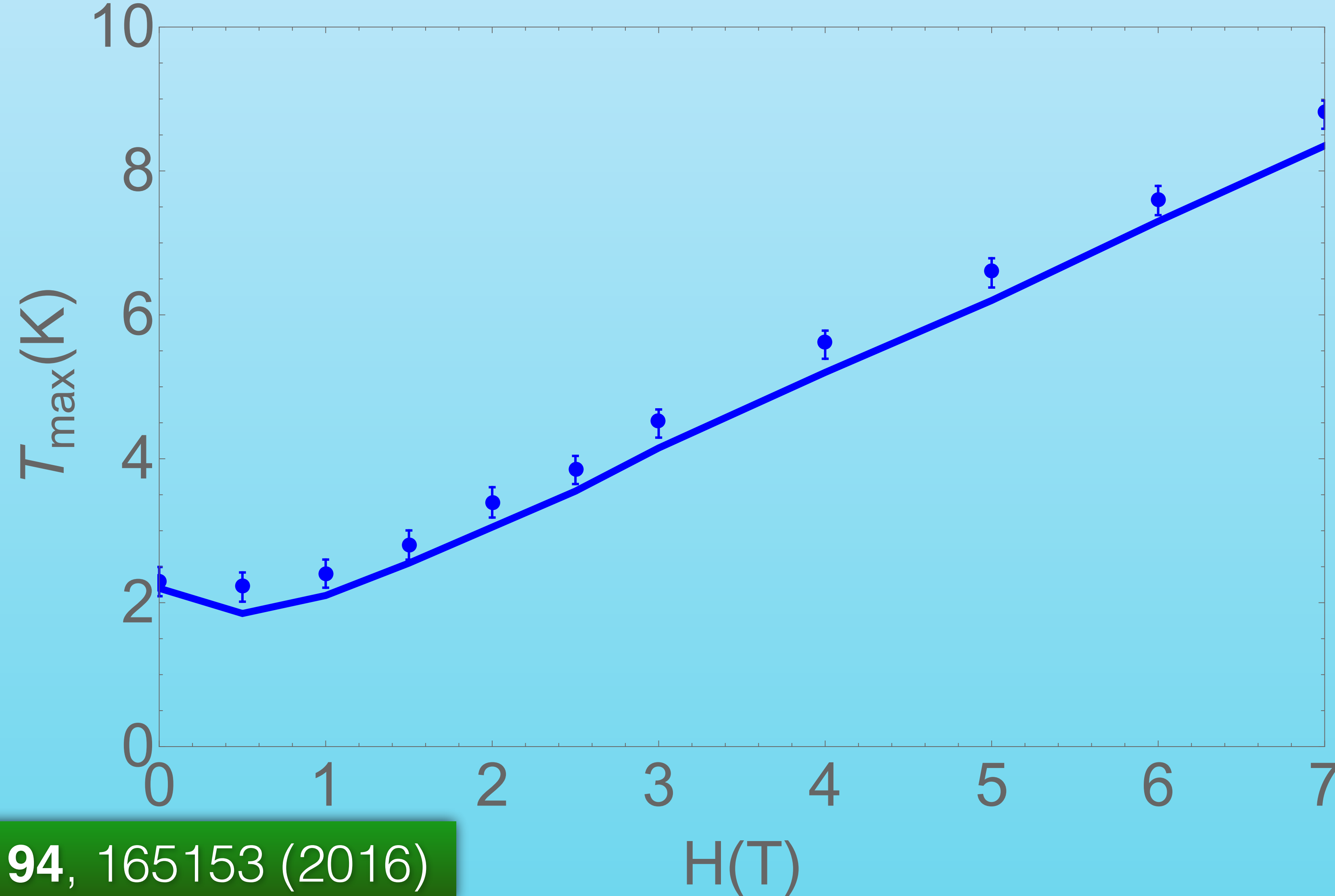



Evolution of heat capacity maximum in applied [110] field

Data from [1] Petit et al, PRB **94**, 165153 (2016)

# Comparison to scattering data in [100] field

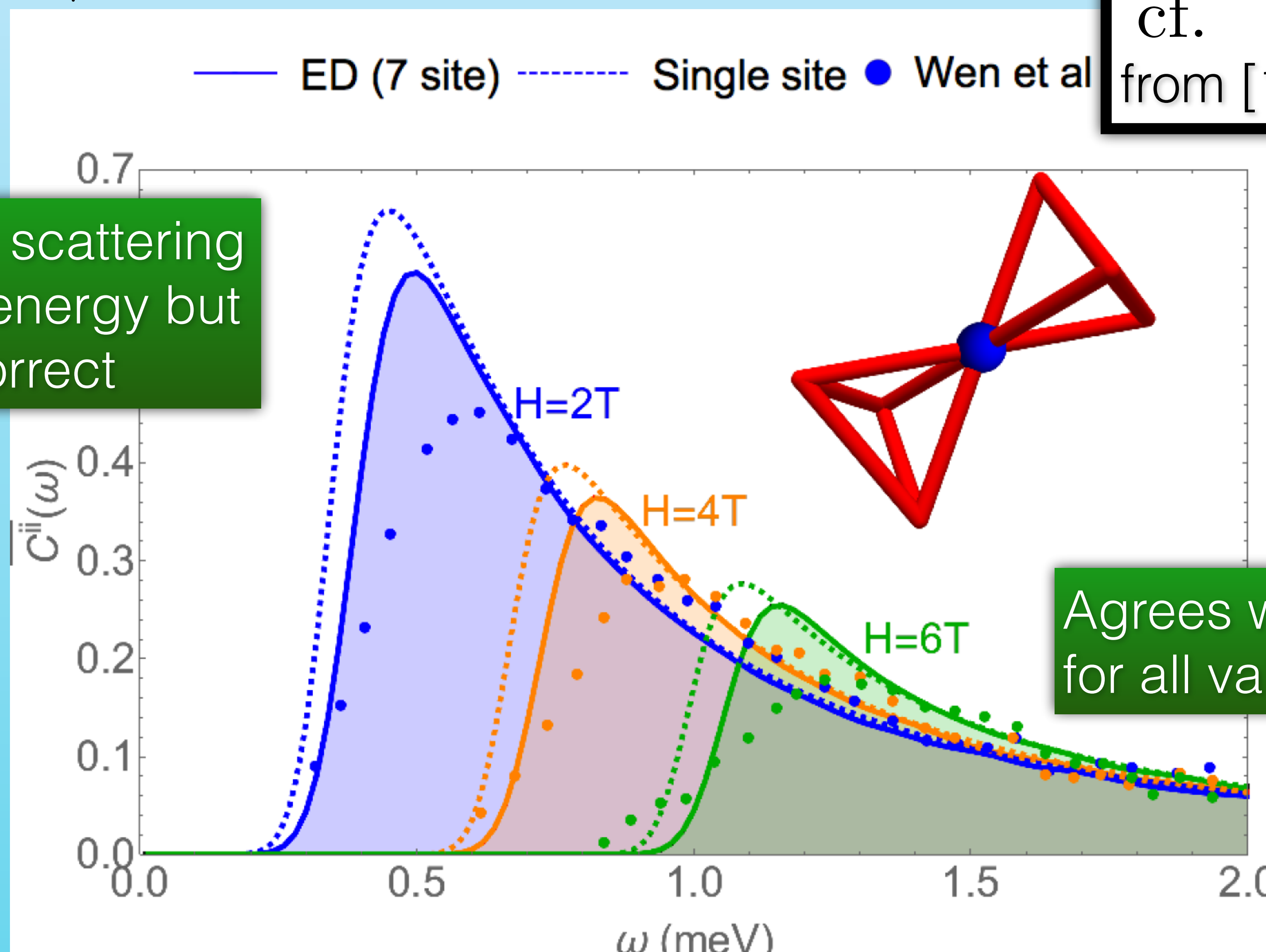
Onsite correlation function calculated for central spin of 7-site cluster in ED

$$\overline{C}_{ii}(\omega) = 4\sum_{|\alpha\rangle} \overline{|\langle 0|S_i^z|\alpha\rangle|^2\delta(\omega - E_\alpha)}$$

Compared to q-integrated scattering data from [1] Wen et al, PRL **118**, 107206 (2017)

$$p(h) = \frac{2\Gamma}{\pi}\frac{1}{h^2+\Gamma^2} \qquad \Gamma = 0.38\text{meV} \quad J_{zz} = 0.08\text{meV} \quad g_z = 4.9$$

cf. $\Gamma = 0.54\text{meV}$ from [1]



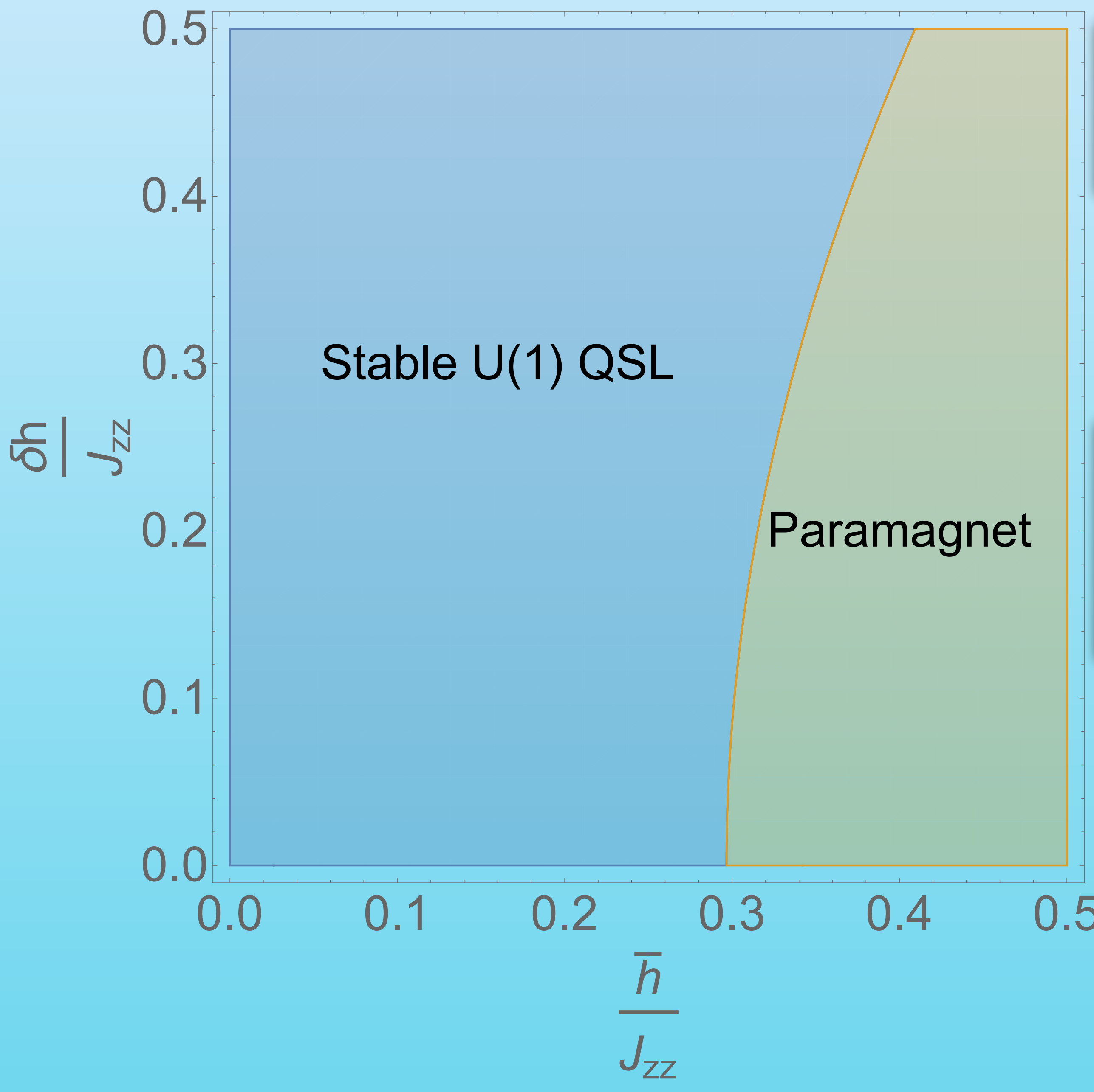

Overestimates scattering near Zeeman energy but qualitatively correct

Agrees with high enery tail for all values of applied field

# What does this model suggest about the ground state of $Pr_2Zr_2O_7$?

Wish to apply stability criterion

$$\frac{J_{zz}}{2} - 3\frac{\overline{h}}{2} + \frac{7\delta h^2 - 5\overline{h}^2}{8J} > 0$$




Problem: Lorentzian distribution lacks well defined moments $\overline{h}, \overline{h^2}$

$$p(h) = \frac{2\Gamma}{\pi}\frac{1}{h^2+\Gamma^2} \quad h \in [0, \infty]$$

Workaround: apply finite cut-off $h_{max}$ to distribution and observe trajectory in phase diagram as function of $h_{max}$

$$\Gamma = 0.38\text{meV} \quad J_{zz} = 0.08\text{meV}$$

$$p(h) = \frac{\Gamma}{\arctan\left(\frac{h_{max}}{\Gamma}\right)}\frac{1}{\Gamma^2+h^2}$$

$$h \in [0, h_{max}]$$

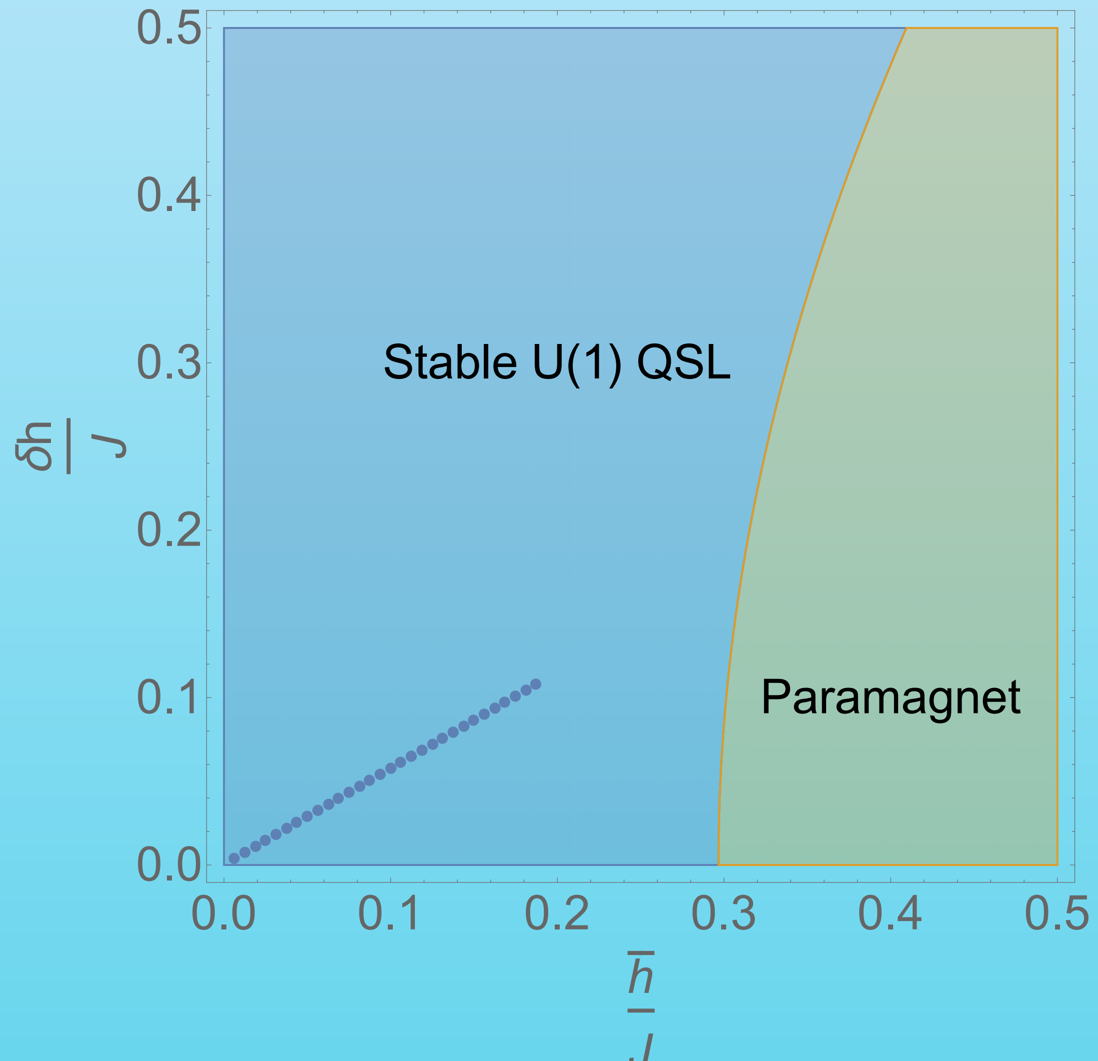
# What does this model suggest about the ground state of $Pr_2Zr_2O_7$?

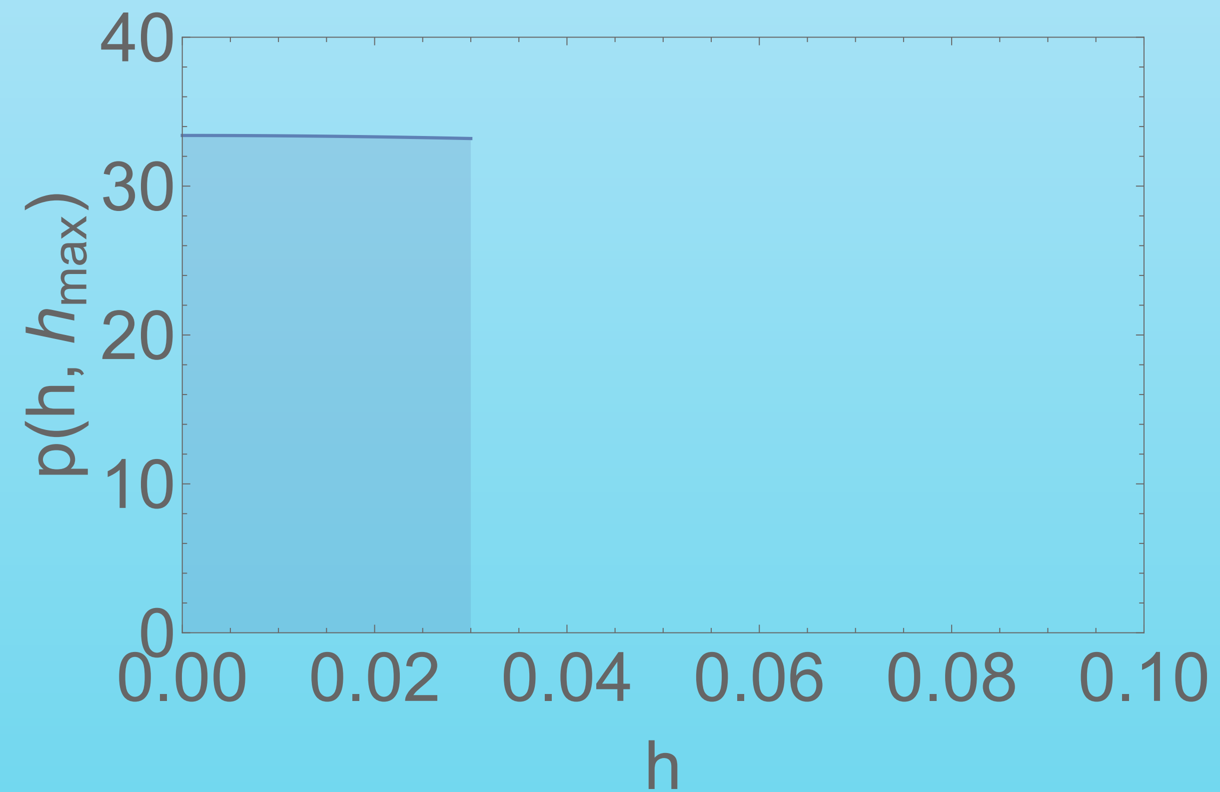
Series of distributions

$$\Gamma = 0.38\text{meV} \qquad J_{zz} = 0.08\text{meV}$$

$$p(h) = \frac{\Gamma}{\arctan\left(\frac{h_{max}}{\Gamma}\right)} \frac{1}{\Gamma^2 + h^2} \qquad h \in [0, h_{max}]$$

$$h_{max} = 0.03\text{meV}$$



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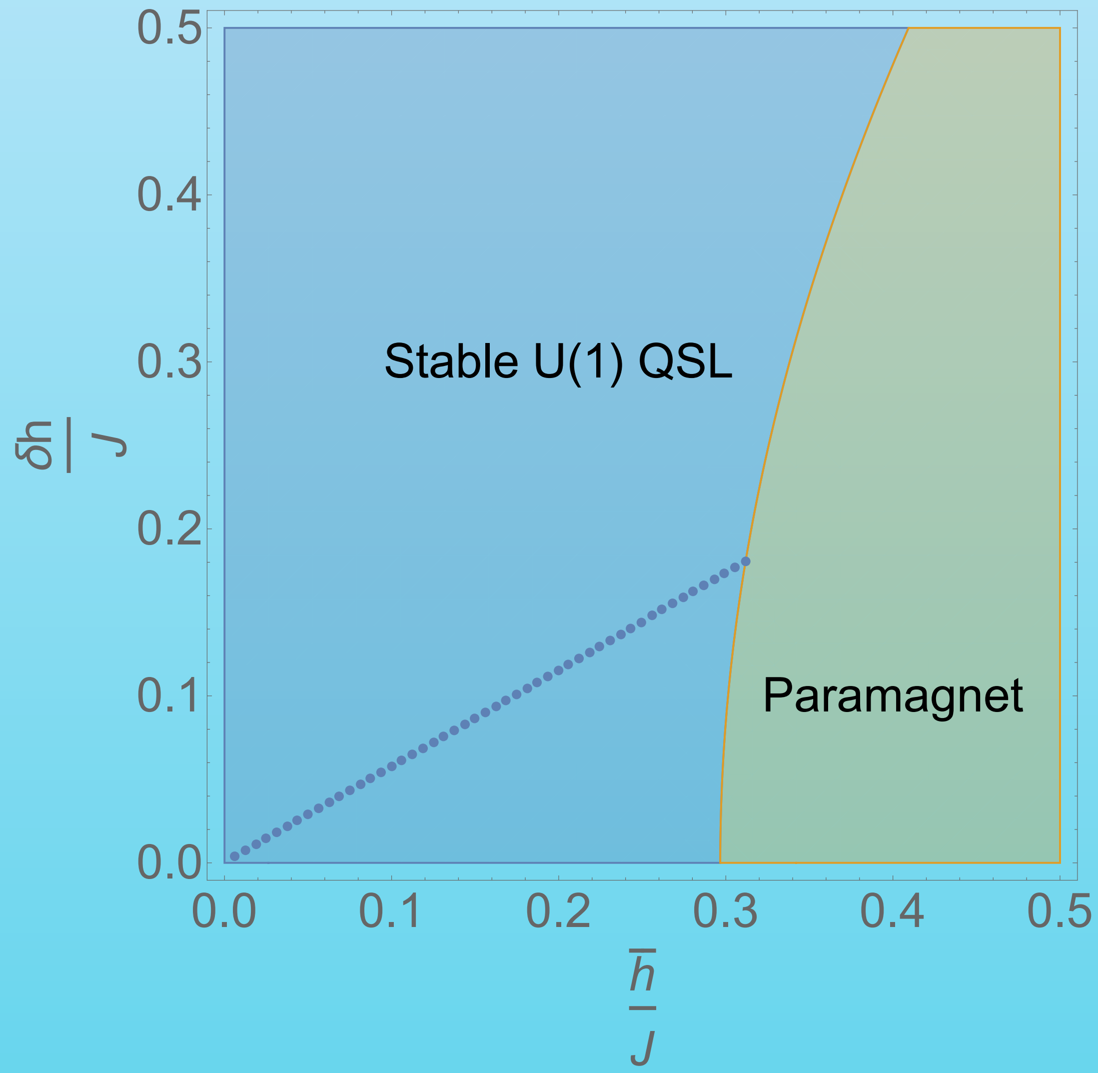
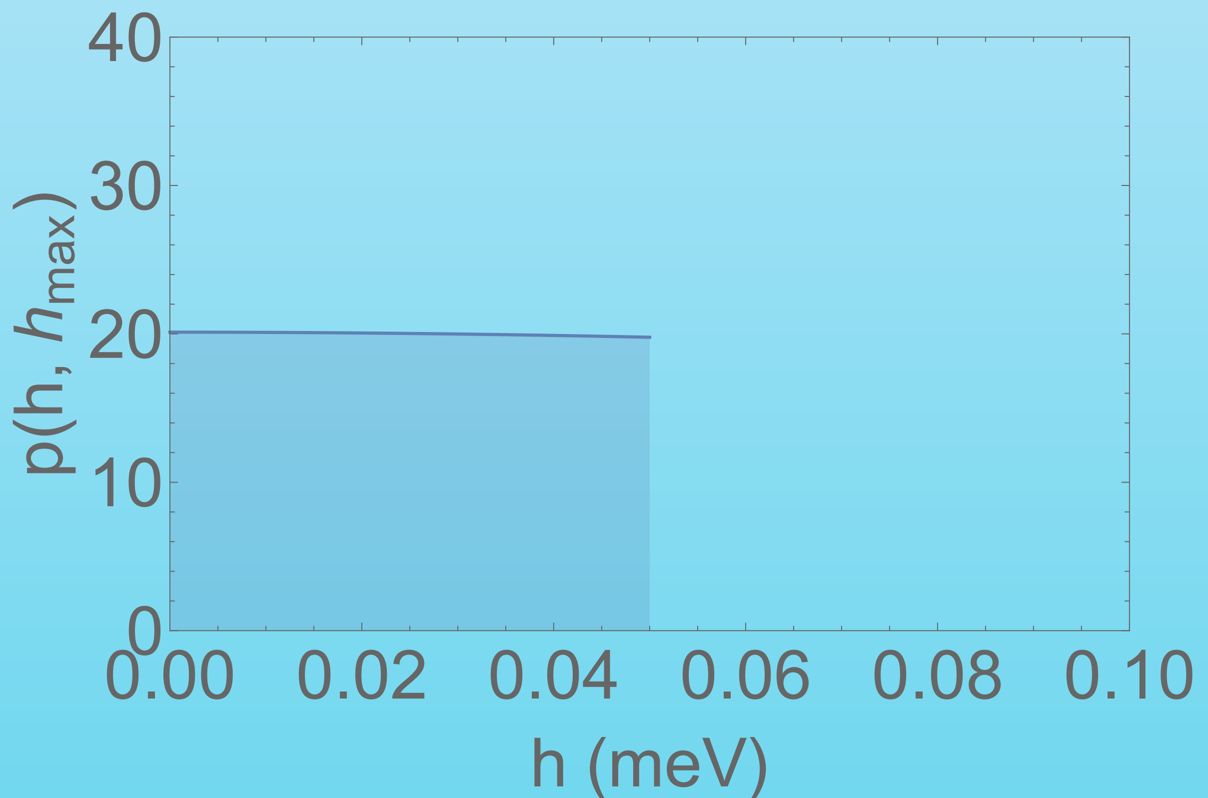
$$h_{max} = 0.05\text{meV}$$



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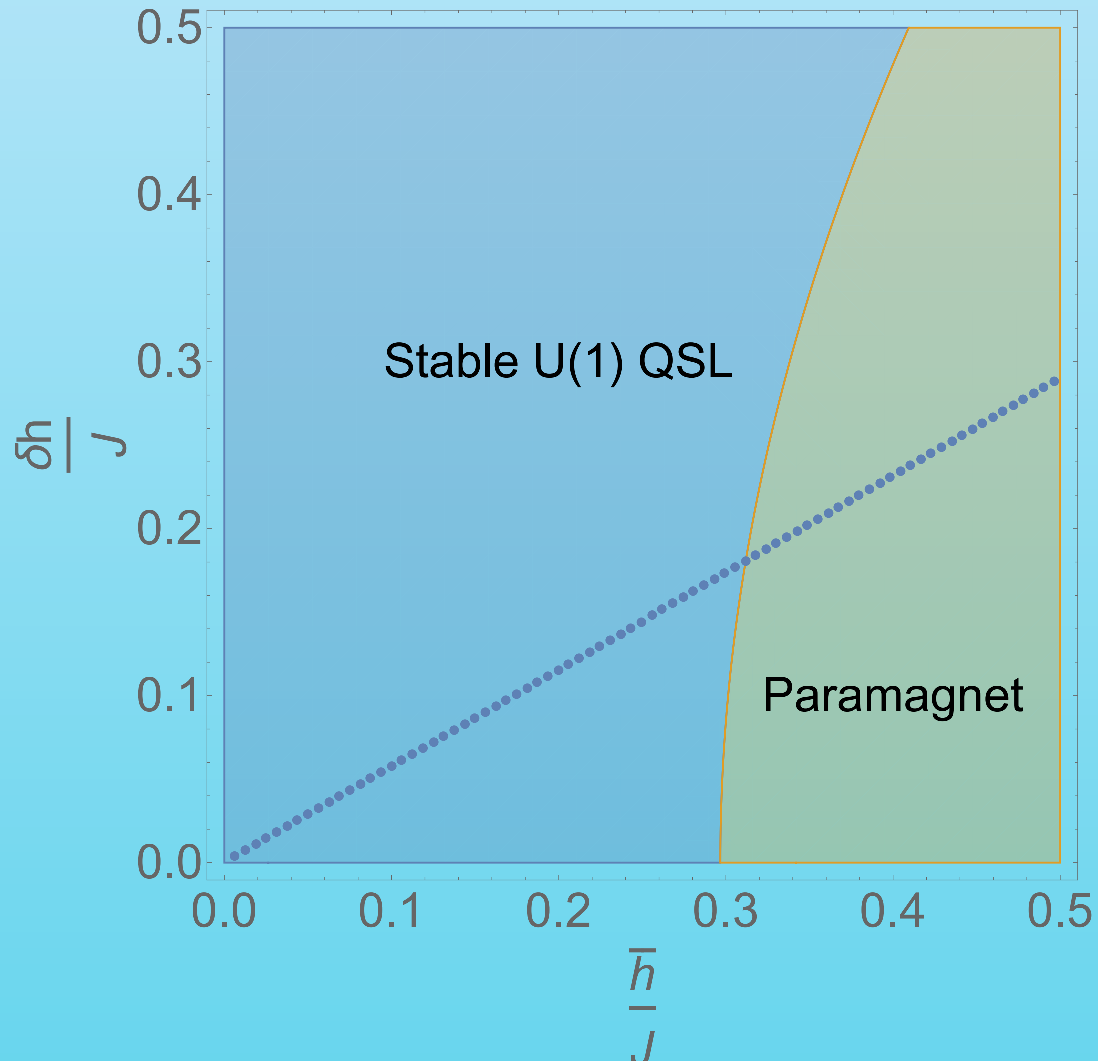
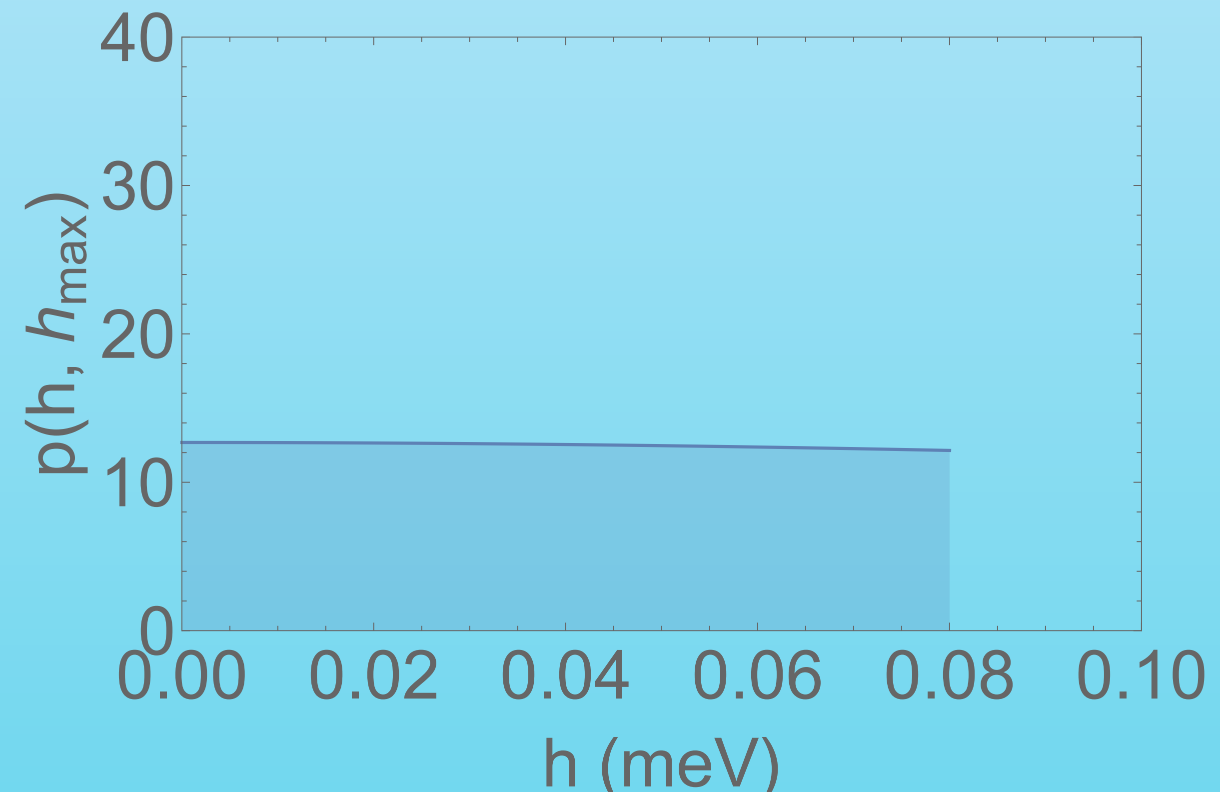
$$h_{max} = 0.08\text{meV}$$



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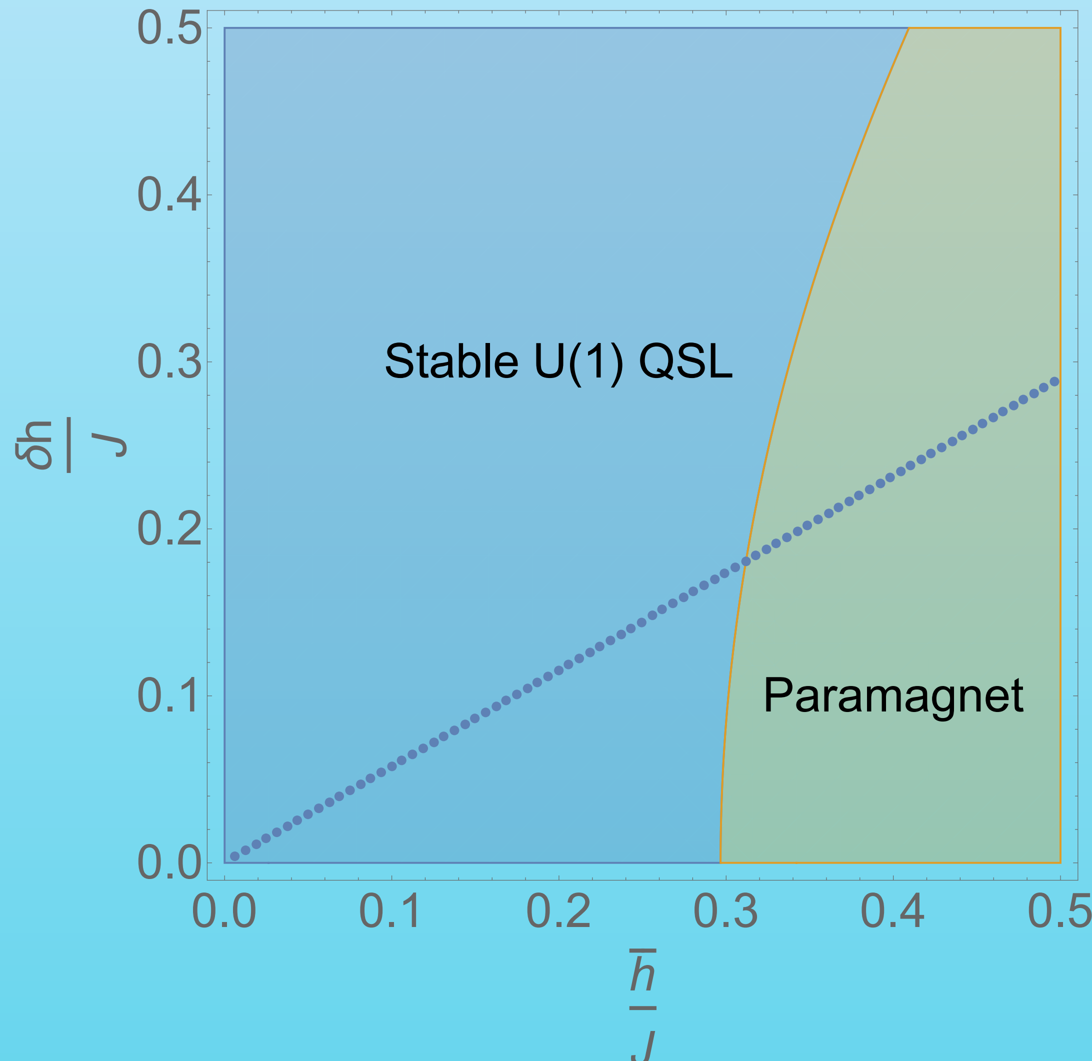
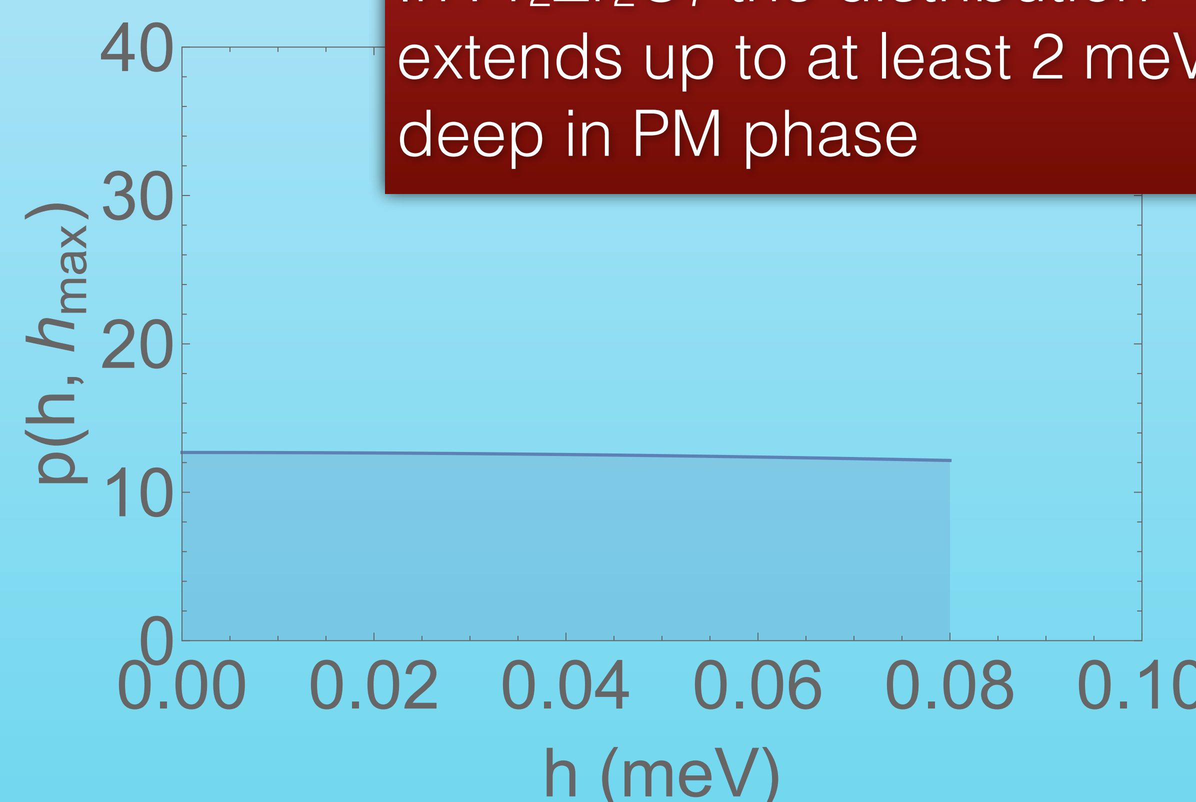
$$h_{max} = 0.08\text{meV}$$




In $Pr_2Zr_2O_7$ the distribution extends up to at least 2 meV: deep in PM phase

# Is this consistent with scattering data?

PRL 118, 107206 (2017) PHYSICAL REVIEW LETTERS week ending 10 MARCH 2017

**Disordered Route to the Coulomb Quantum Spin Liquid: Random Transverse Fields on Spin Ice in $Pr_2Zr_2O_7$**

J.-J. Wen,[1,2,3] S. M. Koohpayeh,[1] K. A. Ross,[1,4] B. A. Trump,[5] T. M. McQueen,[1,5,6] K. Kimura,[7,8] S. Nakatsuji,[7,9] Y. Qiu,[4] D. M. Pajerowski,[4] J. R. D. Copley,[4] and C. L. Broholm[1,4,6]

Suffice it to say that $\mathcal{H}$, which contains only the longitudinal Ising interactions, provides an acceptable account of the data.

QSI is expected to be stable to all local perturbations, despite being gapless [2]. A recent theoretical study of $\mathcal{H}$ shows random transverse fields induce quantum entanglement and two distinct QSLs [5]. The gapless nature of the spectrum [Fig. 1(b)] and the spin-ice-like correlations reported here (Fig. 3) and in previous studies [4] preclude a trivial paramagnet and point to a disorder induced QSL in PZO.

Given the importance of random transverse fields that we

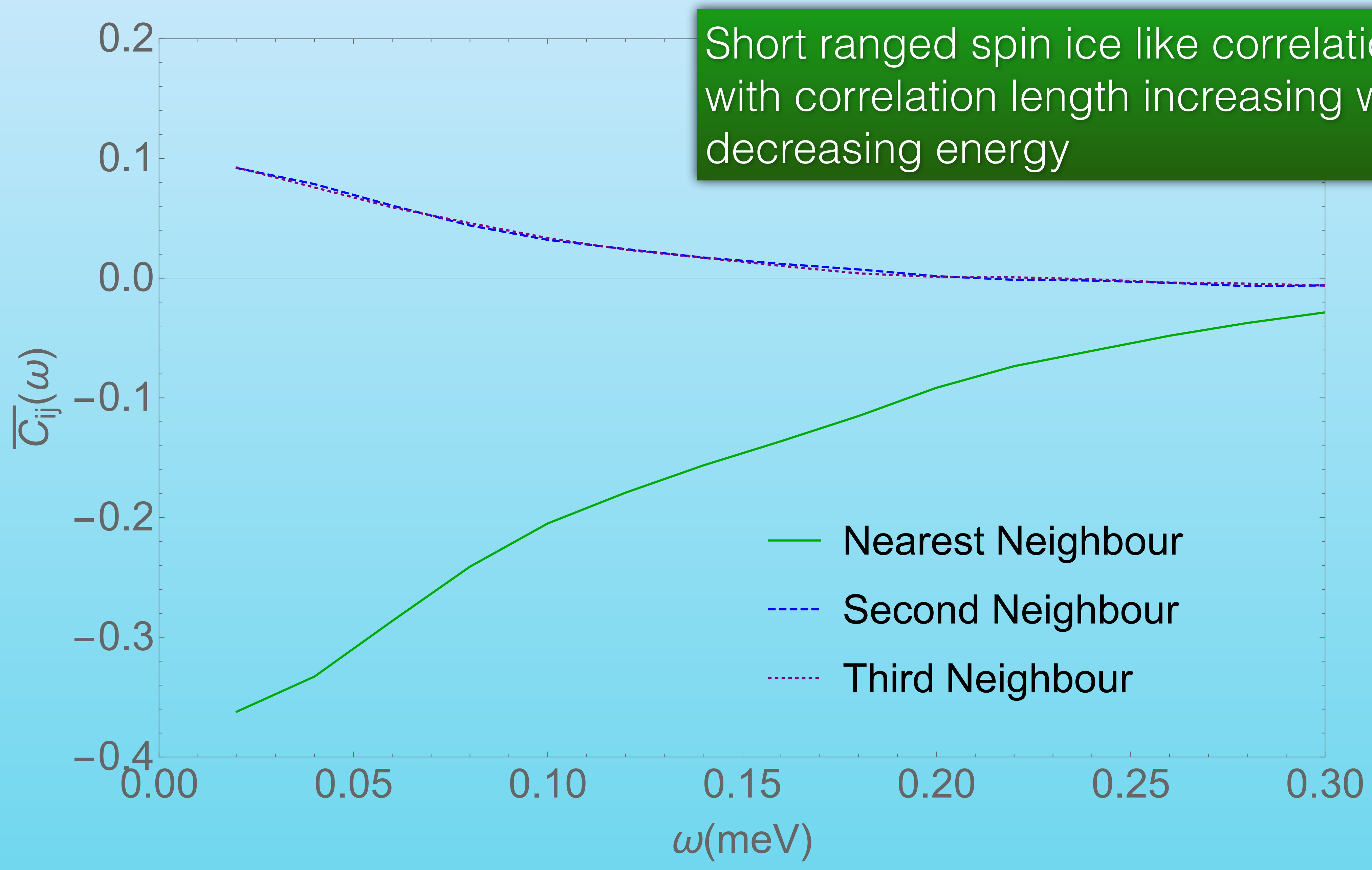
Is a paramagnetic ground state precluded by scattering data?

# Dynamics from exact diagonalization

Calculate real space correlations as a function of energy in 16-site ED within parameterised model

$$\overline{C_{ij}(\omega) = 4\sum_{|\alpha\rangle}\langle 0|S_i^z|\alpha\rangle\langle\alpha|S_j^z|0\rangle\delta(\omega - E_\alpha)}$$

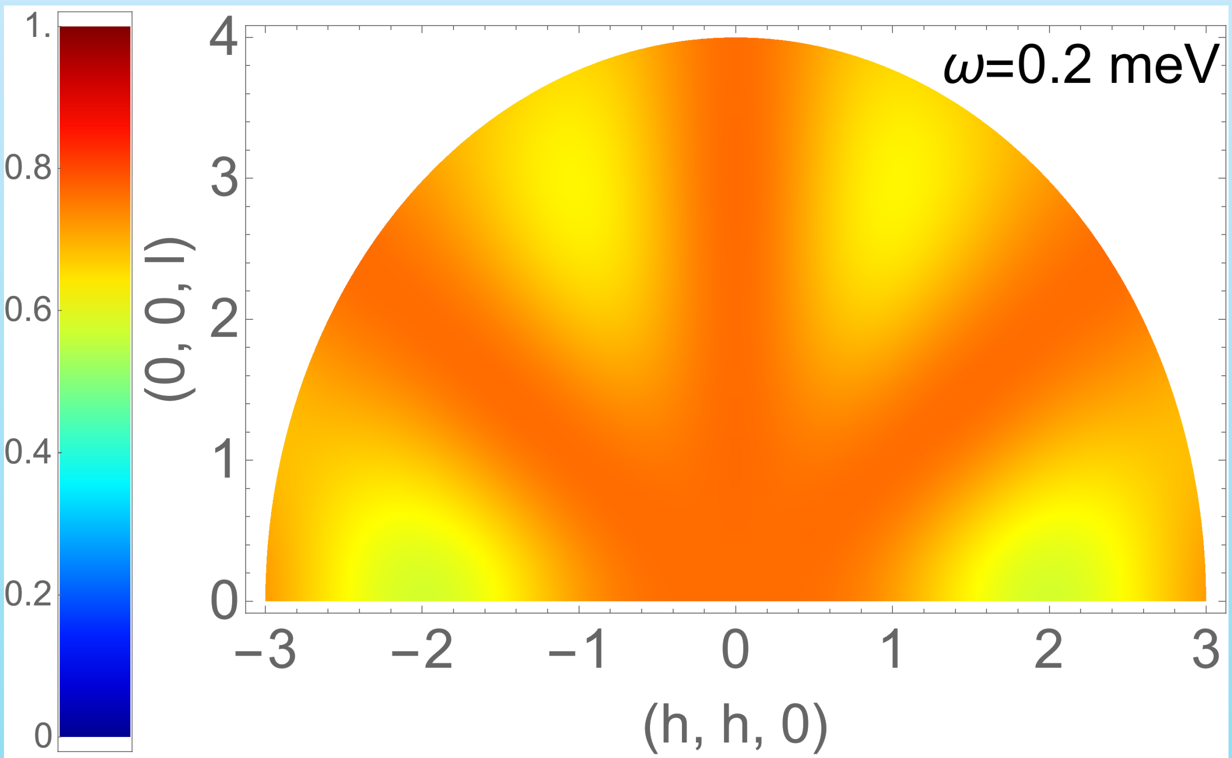
Short ranged spin ice like correlations, with correlation length increasing with decreasing energy

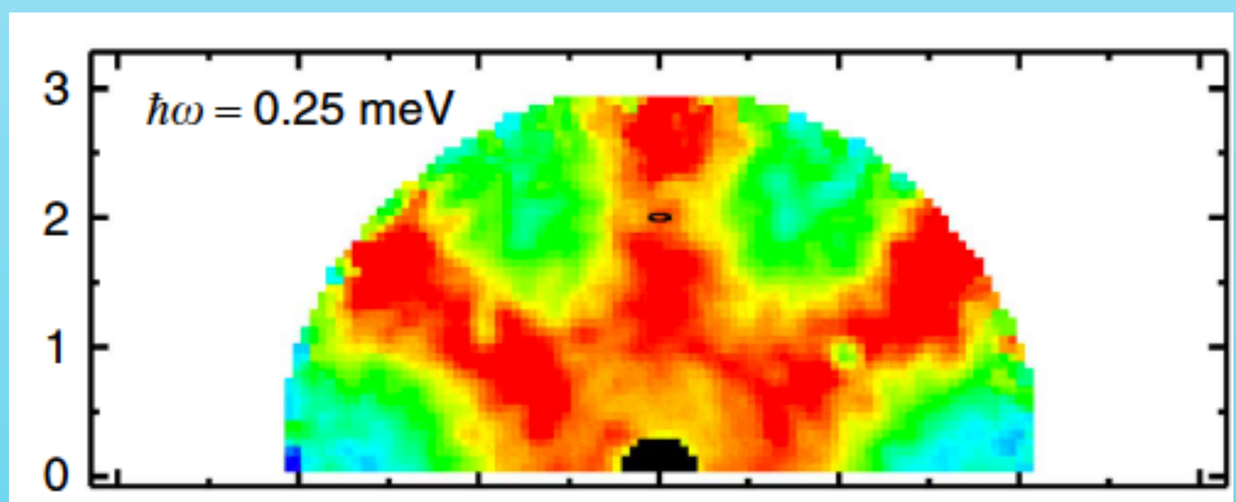


# Dynamics from exact diagonalization

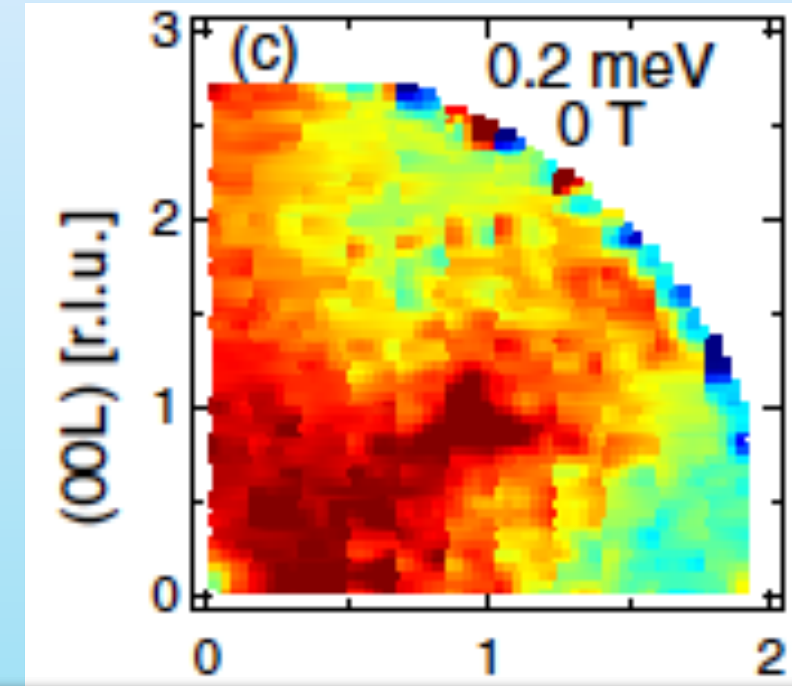
$$S(\mathbf{q},\omega) = \frac{1}{N_{\text{uc}}} \sum_{i,j} \left(\hat{\mathbf{z}}_i \cdot \hat{\mathbf{z}}_j - (\hat{\mathbf{z}}_i \cdot \hat{\mathbf{q}})(\hat{\mathbf{z}}_j \cdot \hat{\mathbf{q}})\right) \overline{C}_{ij}(\omega) e^{i\mathbf{q}\cdot(\mathbf{r}_i - \mathbf{r}_j)}$$

Broadened remnants of spin ice correlations at finite energy

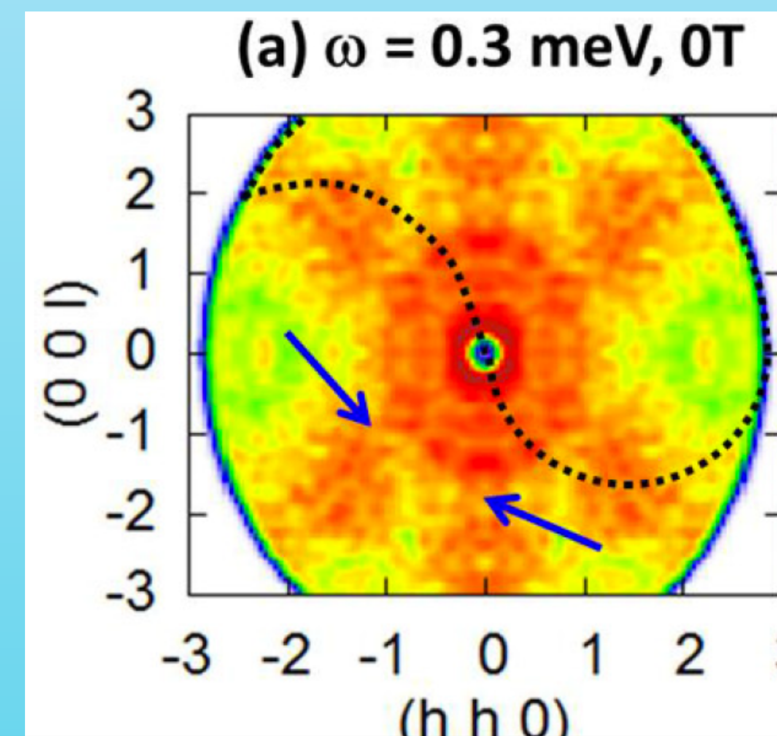



Kimura et al, Nat. Commun. **4**, 1934, (2013)



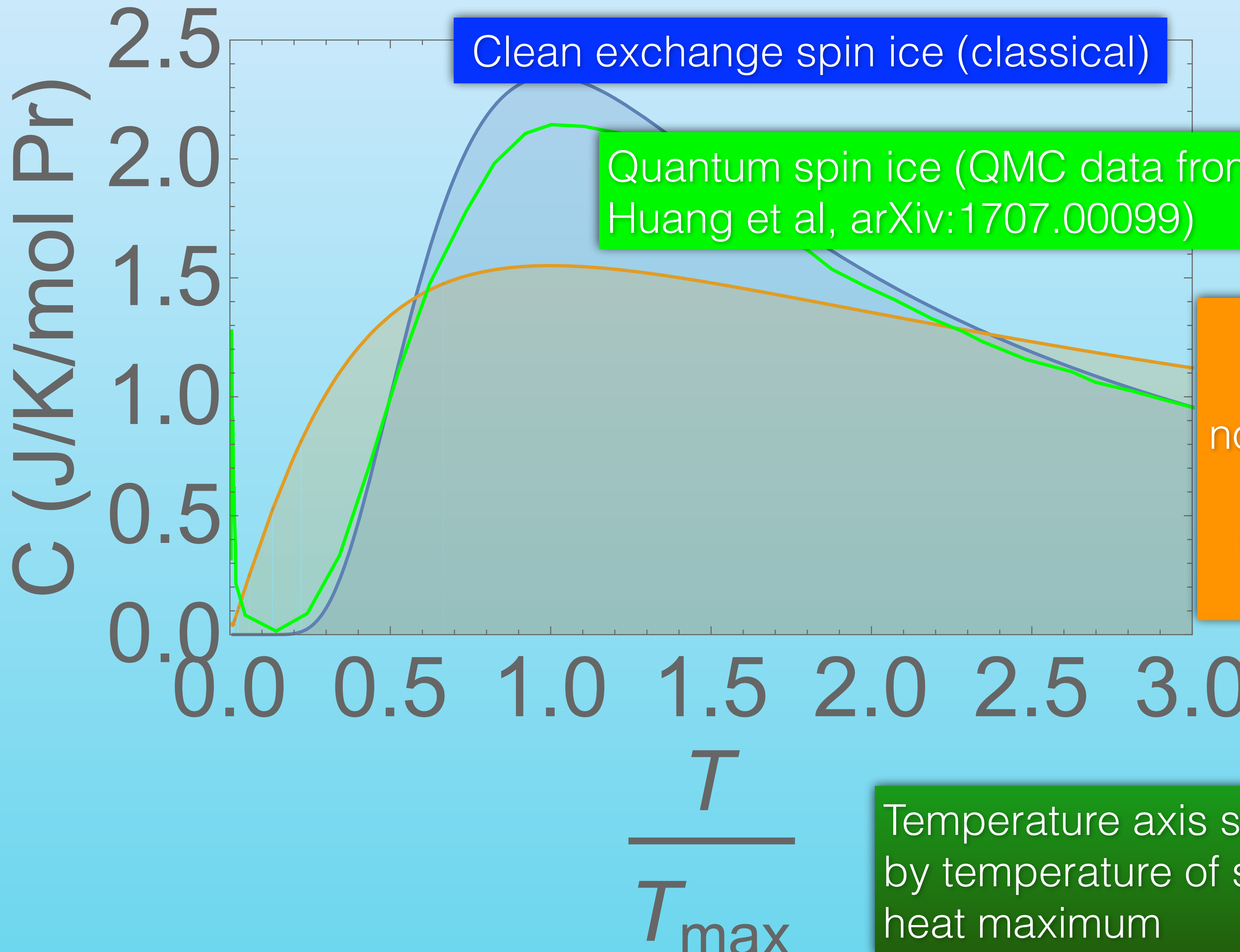

Wen et al, PRL **118**, 107206, (2017)




Petit et al, PRB **94**, 165153, (2016)

# What about other Pr pyrochlores?

Can evaluate prospects by looking at shape of heat capacity curves




Clean exchange spin ice (classical)

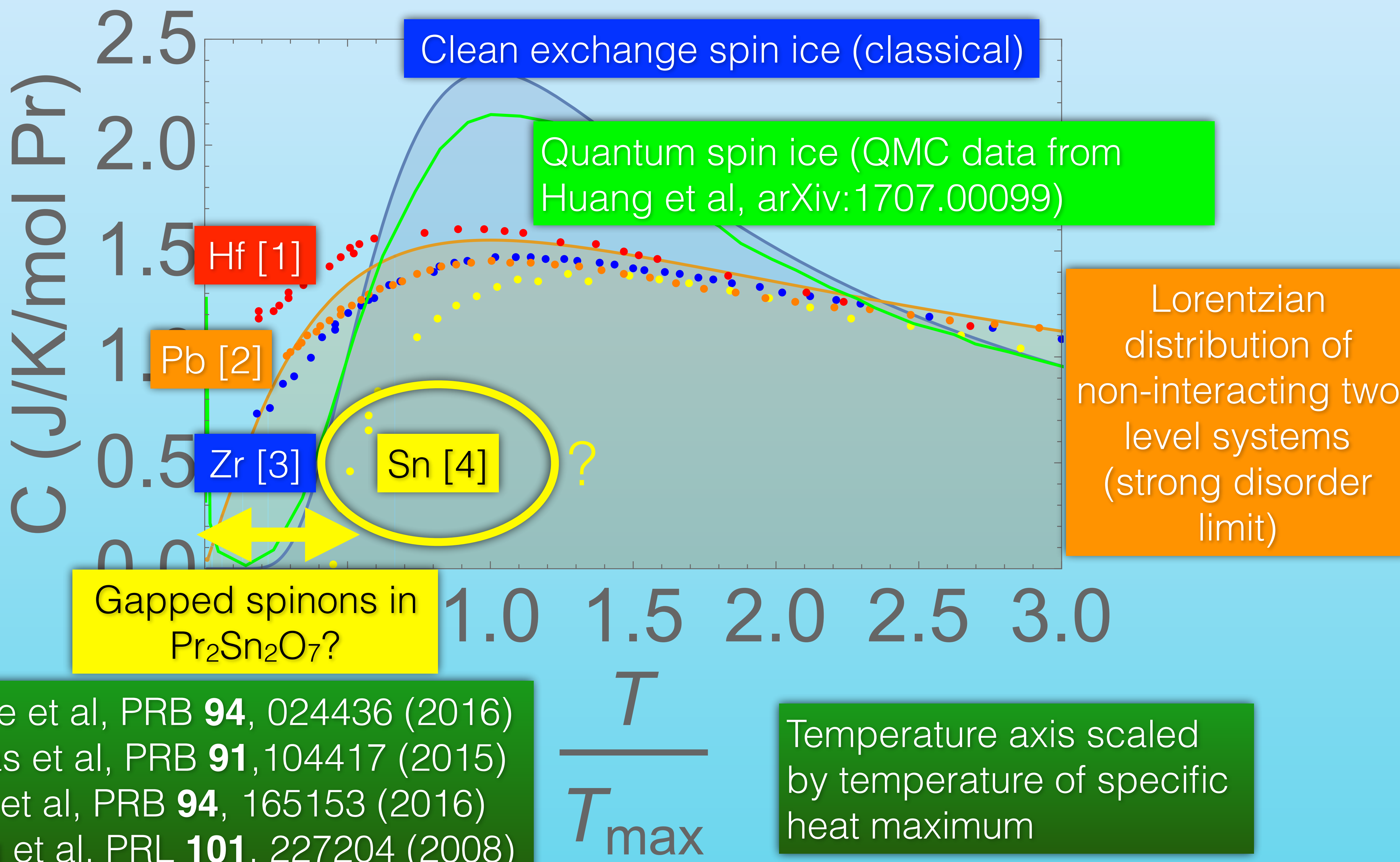
Quantum spin ice (QMC data from Huang et al, arXiv:1707.00099)

Lorentzian distribution of non-interacting two level systems (strong disorder limit)

Temperature axis scaled by temperature of specific heat maximum

# What about other Pr pyrochlores?

Can evaluate prospects by looking at shape of heat capacity curves




Temperature axis scaled by temperature of specific heat maximum

[1] Sibille et al, PRB **94**, 024436 (2016)
[2] Hallas et al, PRB **91**,104417 (2015)
[3] Petit et al, PRB **94**, 165153 (2016)
[4] Zhou et al, PRL **101**, 227204 (2008)

# Conclusions

Perturbation theory calculation of energy cost of introducing spinons is an effective means of finding ground state instability of U(1) QSL phase of quantum spin ice

Parameterising a random transverse field Ising model for $Pr_2Zr_2O_7$ leads to the conclusion that currently studied samples are deep within a topologically trivial paramagnetic phase

$Pr_2Sn_2O_7$ is a good candidate for further investigation in the light of the disorder induced QSL scenario

**From quantum spin liquid to paramagnetic ground states in disordered non-Kramers pyrochlores**

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arXiv:1706.09238

Thanks for listening!