Disorder and its consequences in Pr-based quantum spin ices

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Two questions for today

The theory question:

"How stable is the U(1) quantum spin liquid phase of quantum spin ice against disorder in non-Kramers pyrochlores?"

The experimental question:

"Do presently studied samples of $Pr_2Zr_2O_7$ and $Pr_2Hf_2O_7$ support a quantum spin liquid ground state?"

arXiv:1706.09238

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

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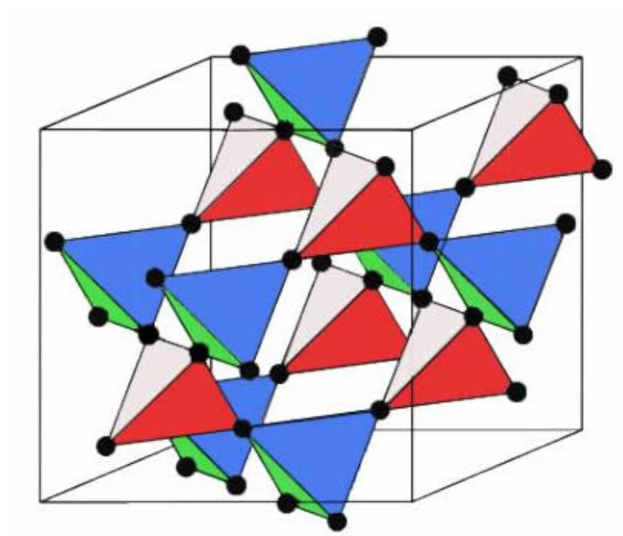
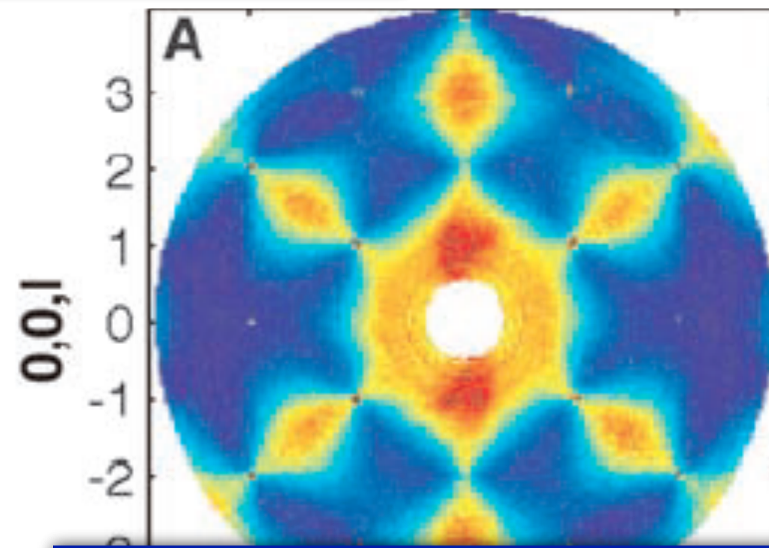
accepted for publication in Physical Review Letters

Quantum spin ice

Spin ice: $(Ho/Dy)_2Ti_2O_7$





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

"Two in, two out"

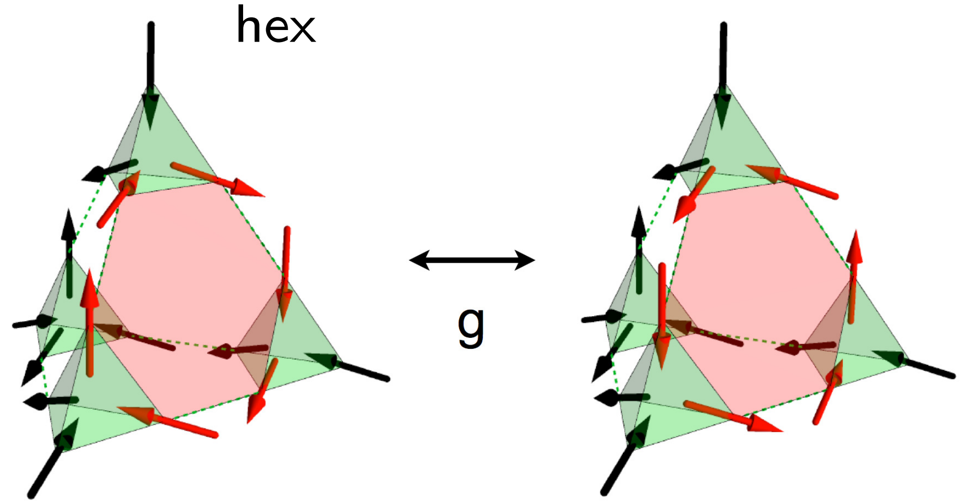
Exponentially large ground state degeneracy

Algebraic spin correlations (pinch points)

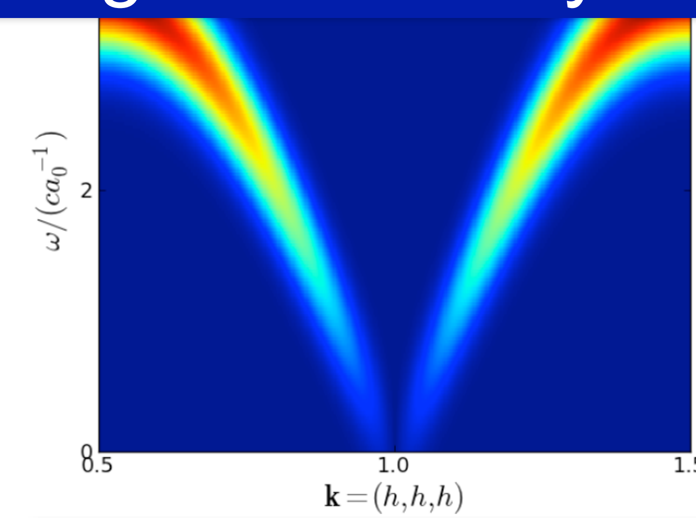
"Magnetic Monopoles"

What happens if you make spin ice "quantum"?

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



U(1) QSL with "emergent electrodynamics"



Quantum tunnelling between spin ice states

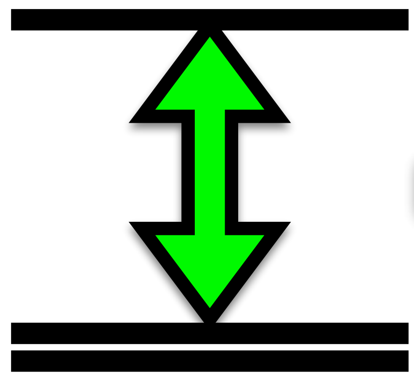
"photons"

Pr pyrochlores: $Pr_2X_2O_7$

Crystal electric field states in pyrochlores: 2J+1 multiplet

Pseudospin-1/2 representation of doublet

Ground doublet separated by large gap



$$|\uparrow\rangle,\ |\downarrow\rangle \qquad \sigma_i^z = \pm 1$$

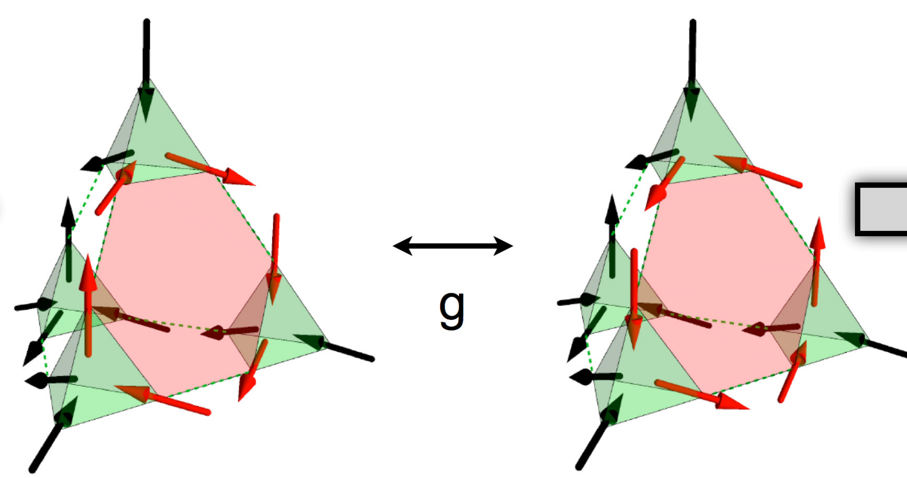
Interactions between nearest neighbours dictated by symmetry of non-Kramers doublet

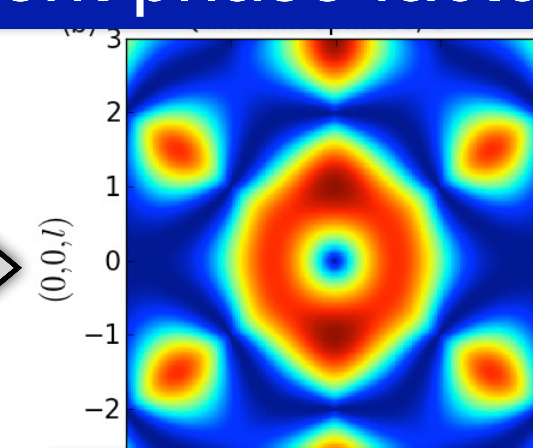
$$\mathcal{H} = \sum_{\langle ij \rangle} \left[\mathrm{J}_z \sigma_i^z \sigma_j^z - \mathrm{J}_\pm (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) + \mathrm{J}_{\pm\pm} (\gamma_{ij} \sigma_i^+ \sigma_j^+ + \gamma_{ij}^* \sigma_i^- \sigma_j^+) \right]$$

U(1) symmetric transverse exchange

Anisotropic exchange with bond-dependent phase factors

$$\mathrm{J}_z > 0,$$
$$|\mathrm{J}_\pm|, |\mathrm{J}_{\pm\pm}| \ll \mathrm{J}_z$$



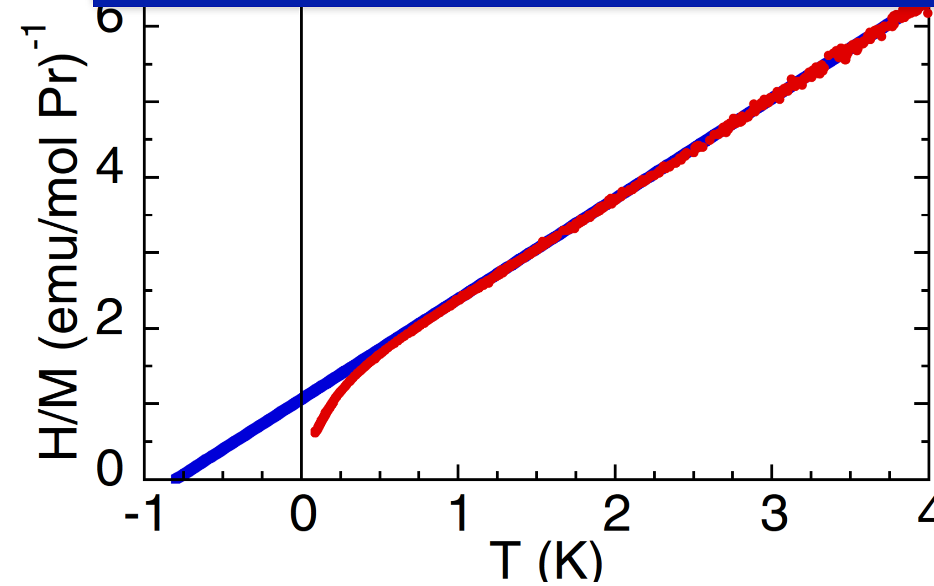


U(1) QSL?

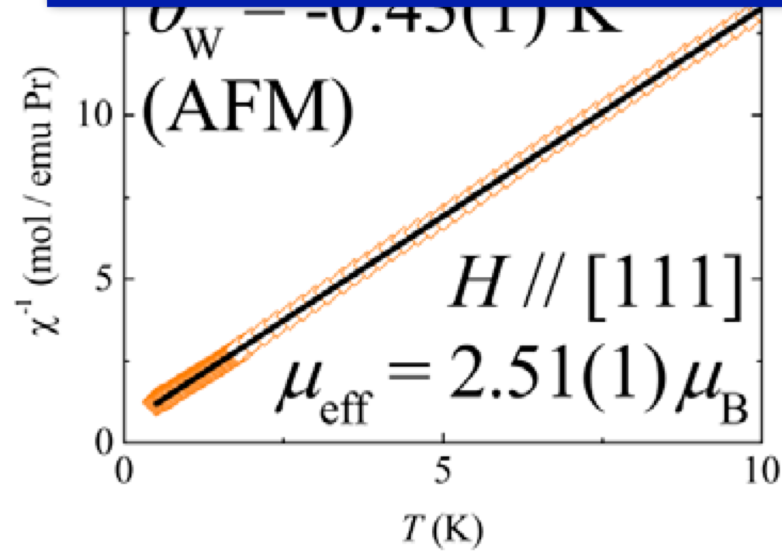
Thermodynamics of Pr pyrochlores

No signature of ordering in magnetic susceptibility

$Pr_2Zr_2O_7$
Petit et al, PRB **94**, 165153 (2016)

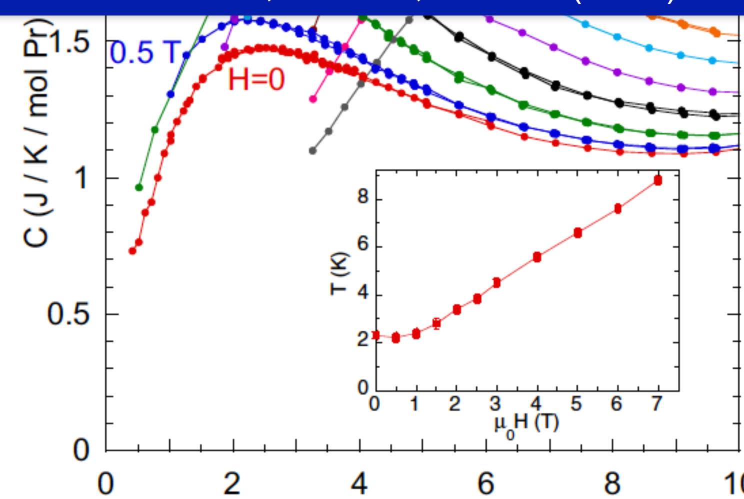


$Pr_2Hf_2O_7$
Ciomaga Hatnean et al, JPCM **29**, 075902 (2017)

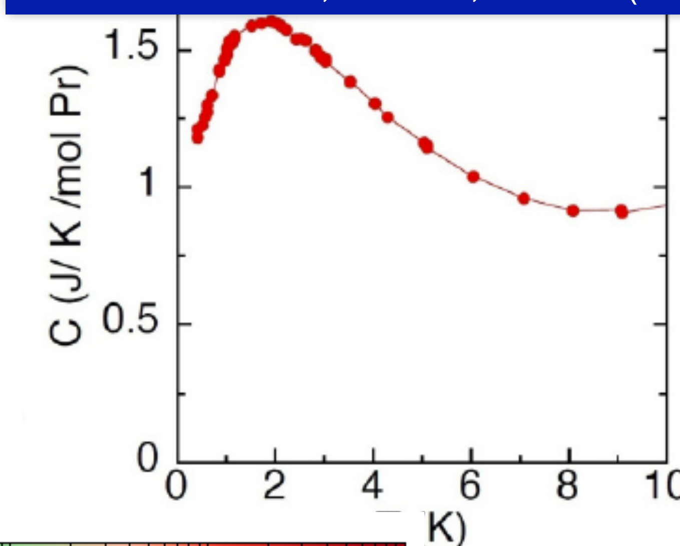


Smooth bump in specific heat

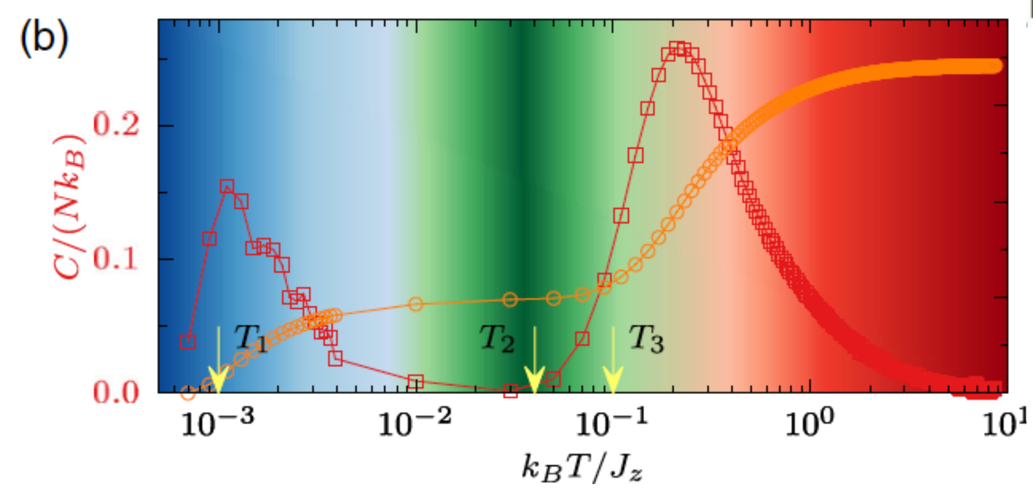
$Pr_2Zr_2O_7$
Petit et al, PRB **94**, 165153 (2016)



$Pr_2Hf_2O_7$
Sibille et al, PRB **94**, 024436 (2016)



cf. QMC for a model quantum spin ice
Huang et al, PRL **120**, 167202 (2018)



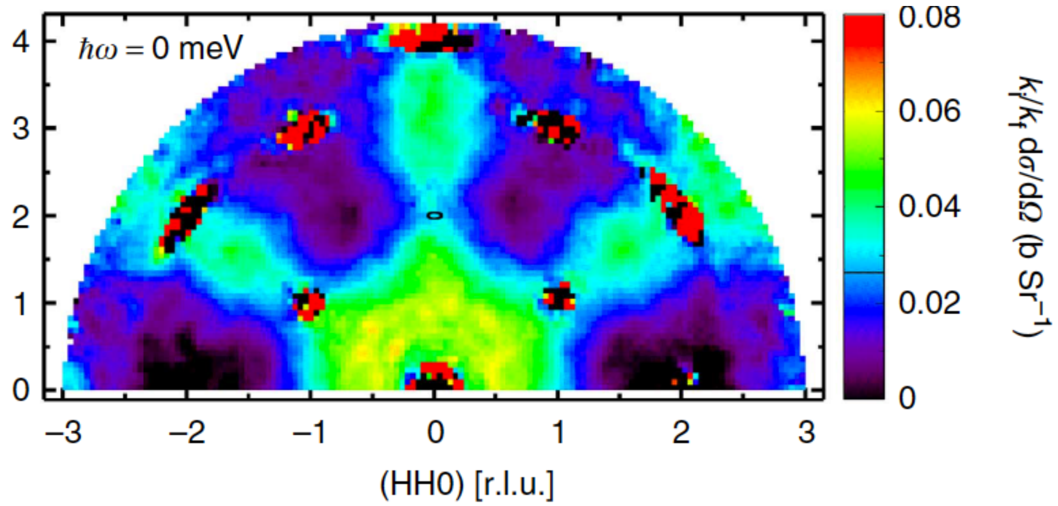
Neutron scattering

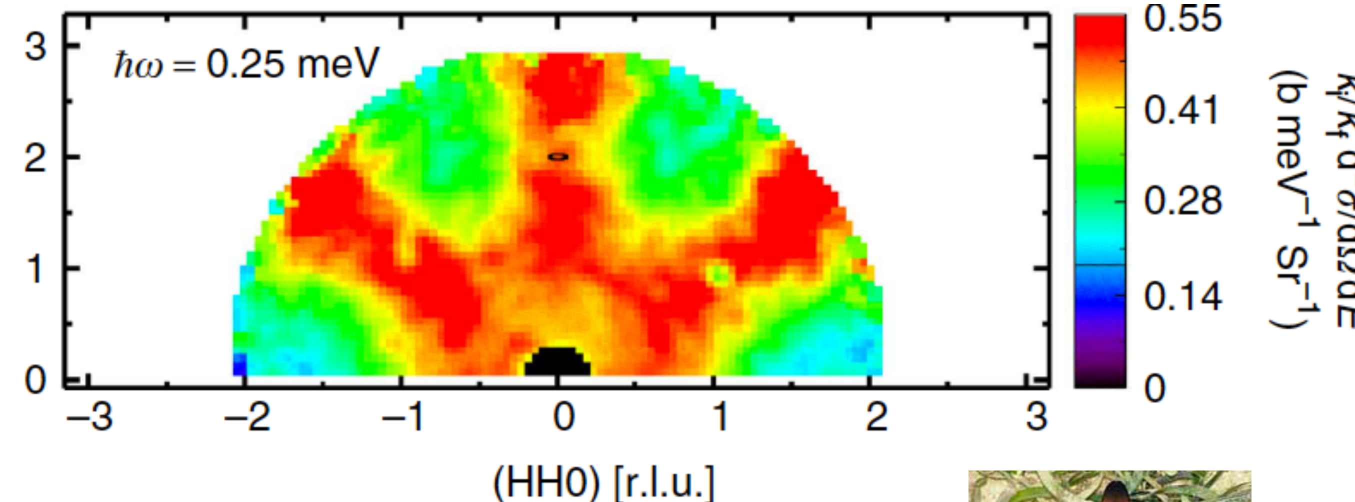
Elastic/low energy scattering

Higher energy scattering

$Pr_2Zr_2O_7$

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



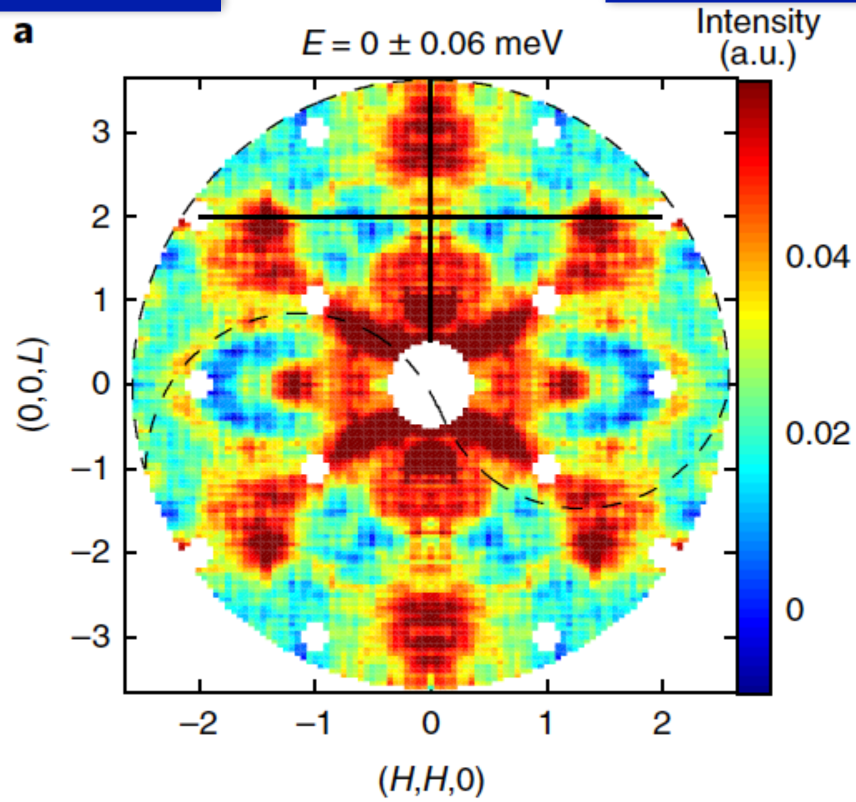


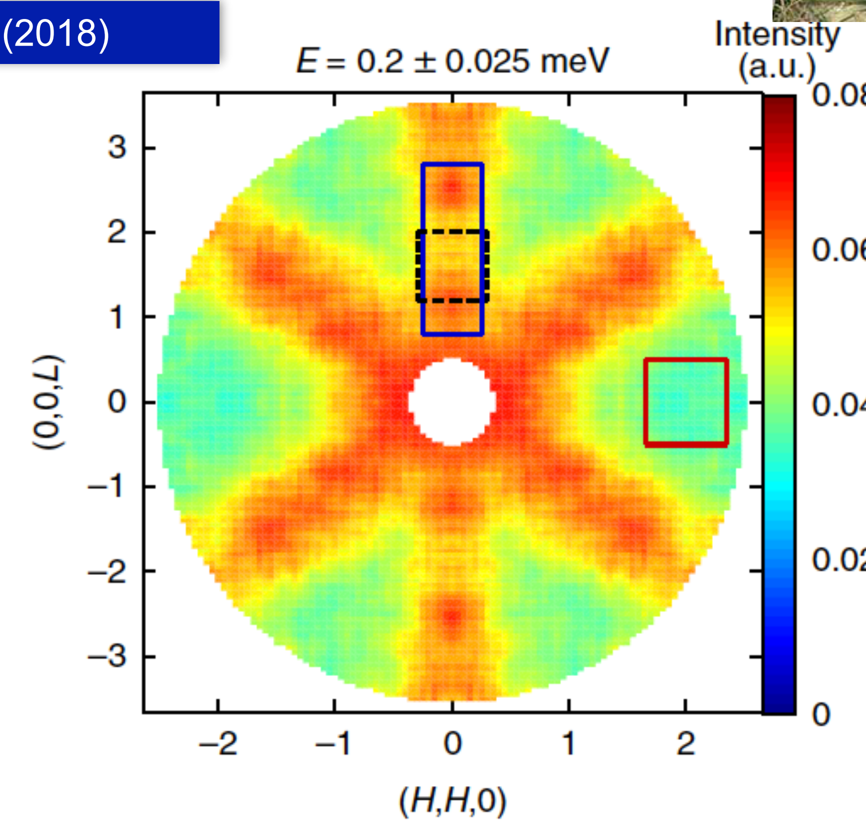
Spin-ice-like correlations

Starfish

$Pr_2Hf_2O_7$

Sibille et al, Nature Phys. **14**, 711 (2018)



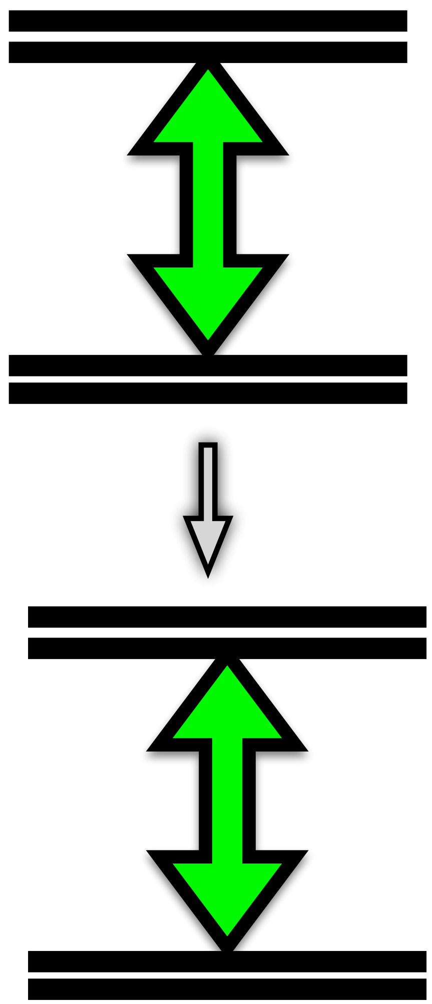


Disorder is important

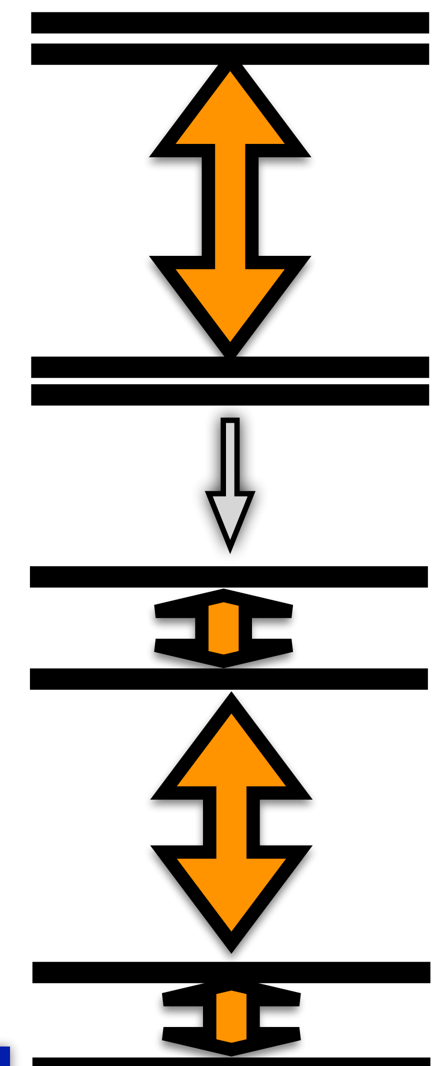
Fundamental difference between non-Kramers (integer J) and Kramers (half-integer J) ions

Kramers ions: doublets protected by Time Reversal Symmetry

non-Kramers ions: doublets not protected by time reversal, only by space group



Structural disorder: space group symmetry locally lifted



Doublets are robust

Doublets become singlets

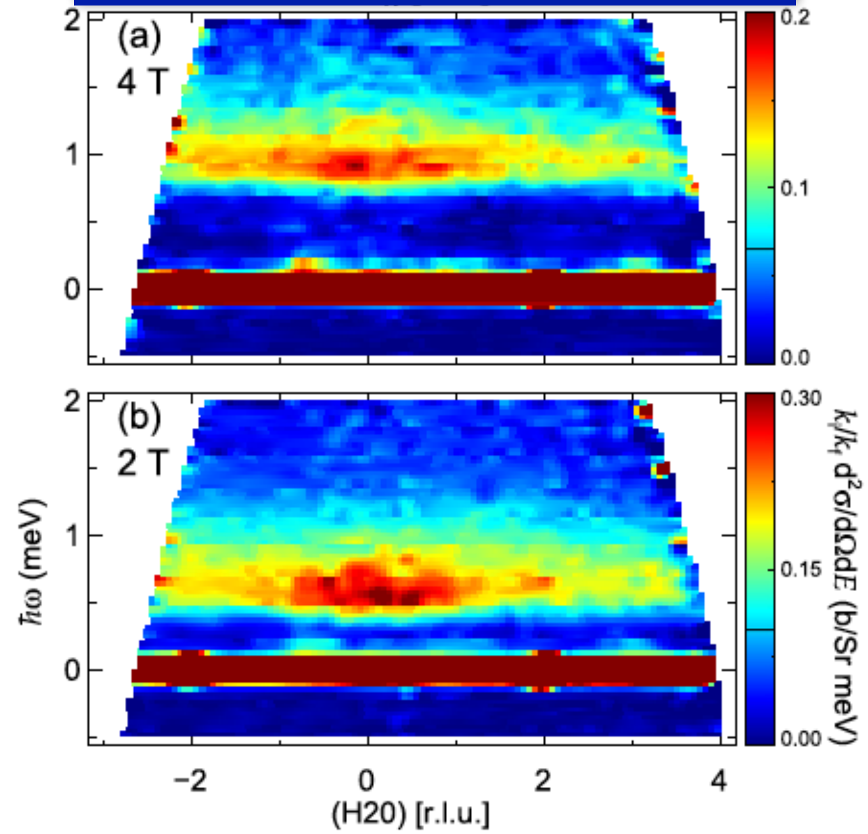
Structural disorder generates random splittings of doublet

$$\delta\mathcal{H} = -\sum_i h_i(e^{i\theta_i}\sigma_i^+ + e^{-i\theta_i}\sigma_i^-)$$

Does this matter in real materials?

$Pr_2Zr_2O_7$: inelastic scattering in a 100 field

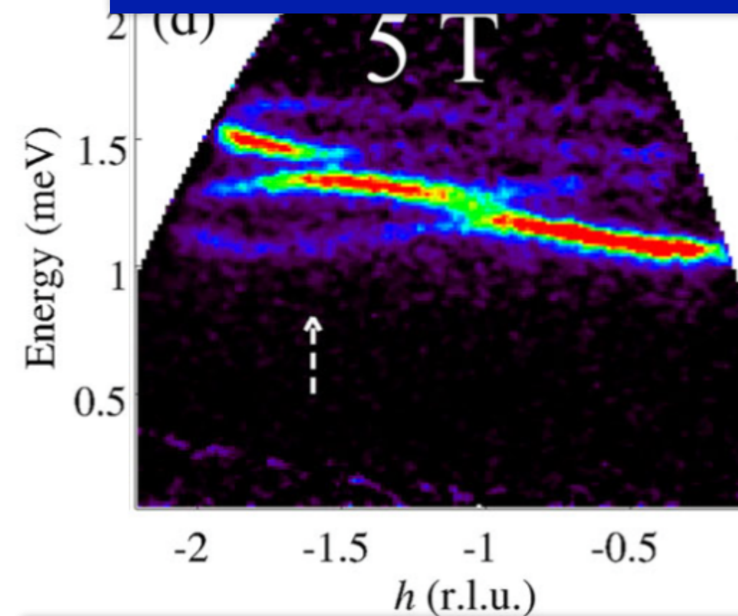
Wen et al, PRL **118**, 107206 (2017)



Broad, non-dispersing continuum that moves up in energy with increasing field

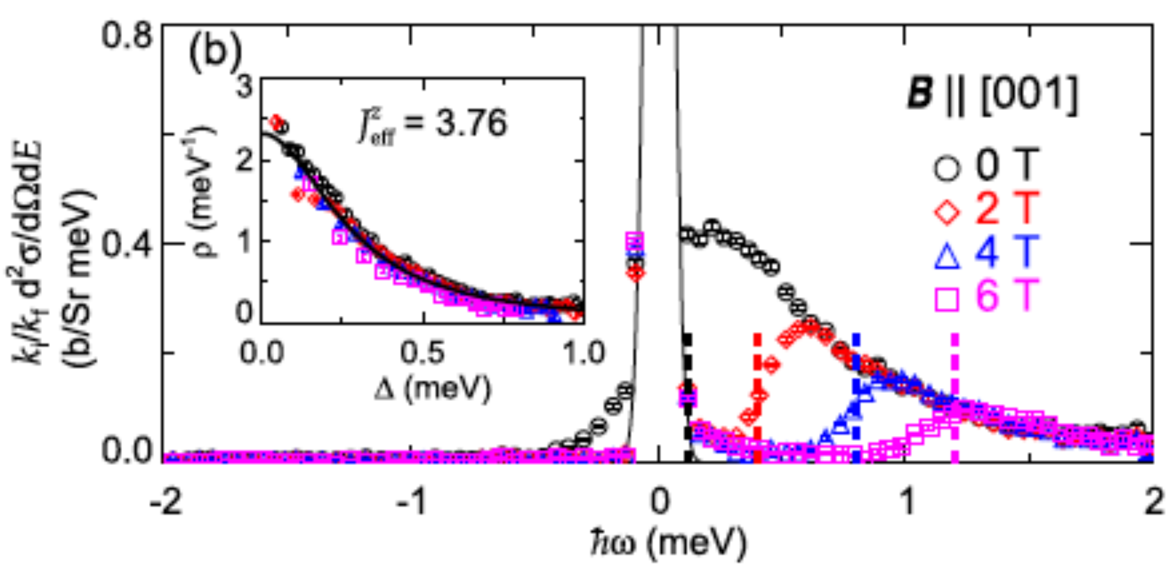
cf. $Yb_2Ti_2O_7$

Thompson et al, PRL **119**, 057203 (2017)



clearly defined dispersive bands

Distribution of excitation energies over range of **H** consistent with Lorentzian distribution of transverse fields



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

$$\Gamma = 0.27\mathrm{meV}$$

large contribution to the Hamiltonian

Disorder isn't necessarily the enemy

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

Disorder-Induced Quantum Spin Liquid in Spin Ice Pyrochlores

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[1]Department of Physics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA
[2]Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106-4030, USA

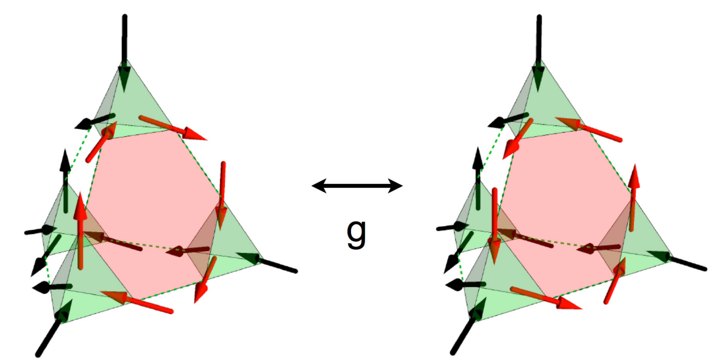
$$\mathcal{H} = J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z - \sum_i h_i(e^{i\theta_i}\sigma_i^+ + e^{-i\theta_i}\sigma_i^-)$$

local coordinate transformation

$$\mathcal{H} = J\sum_{\langle ij\rangle}\sigma_i^z\sigma_j^z - \sum_i h_i\sigma_i^x$$

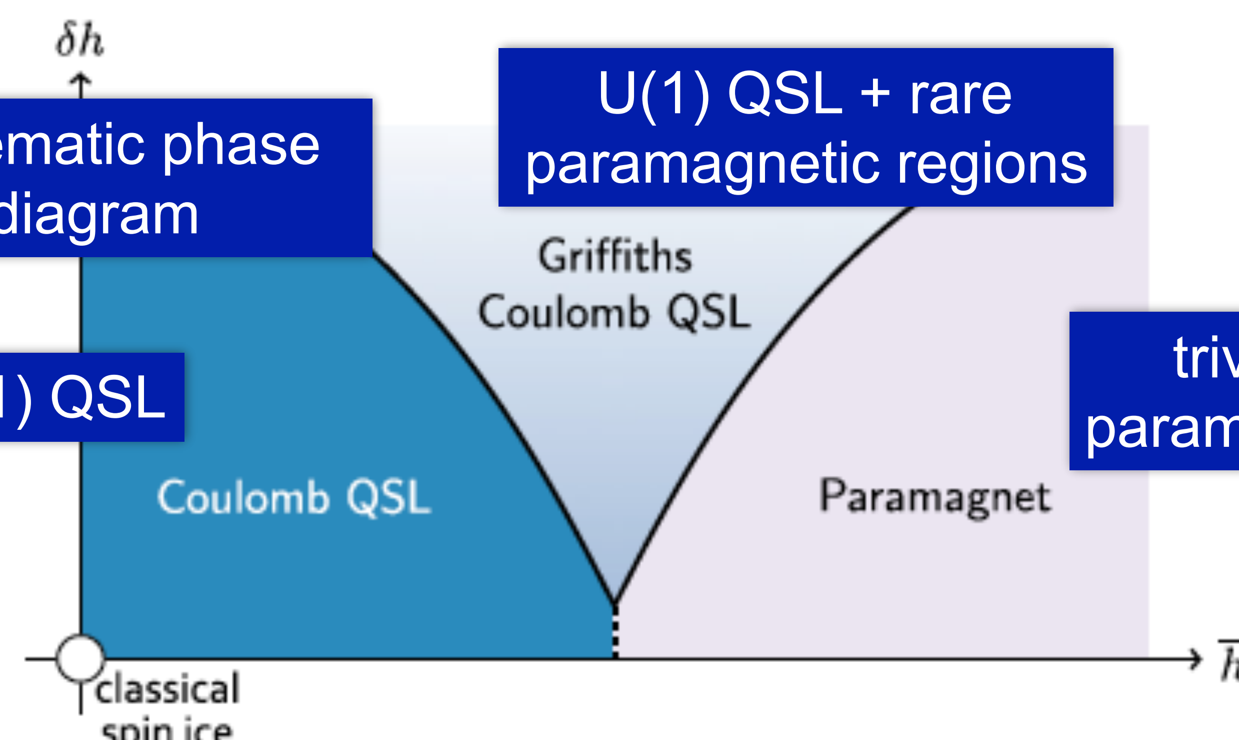
U(1) QSL is stable against disorder so phase persists over finite regime of random transverse field model phase diagram

perturbation theory: transverse fields generate ring tunnelling which stabilizes QSL



Schematic phase diagram

U(1) QSL + rare paramagnetic regions



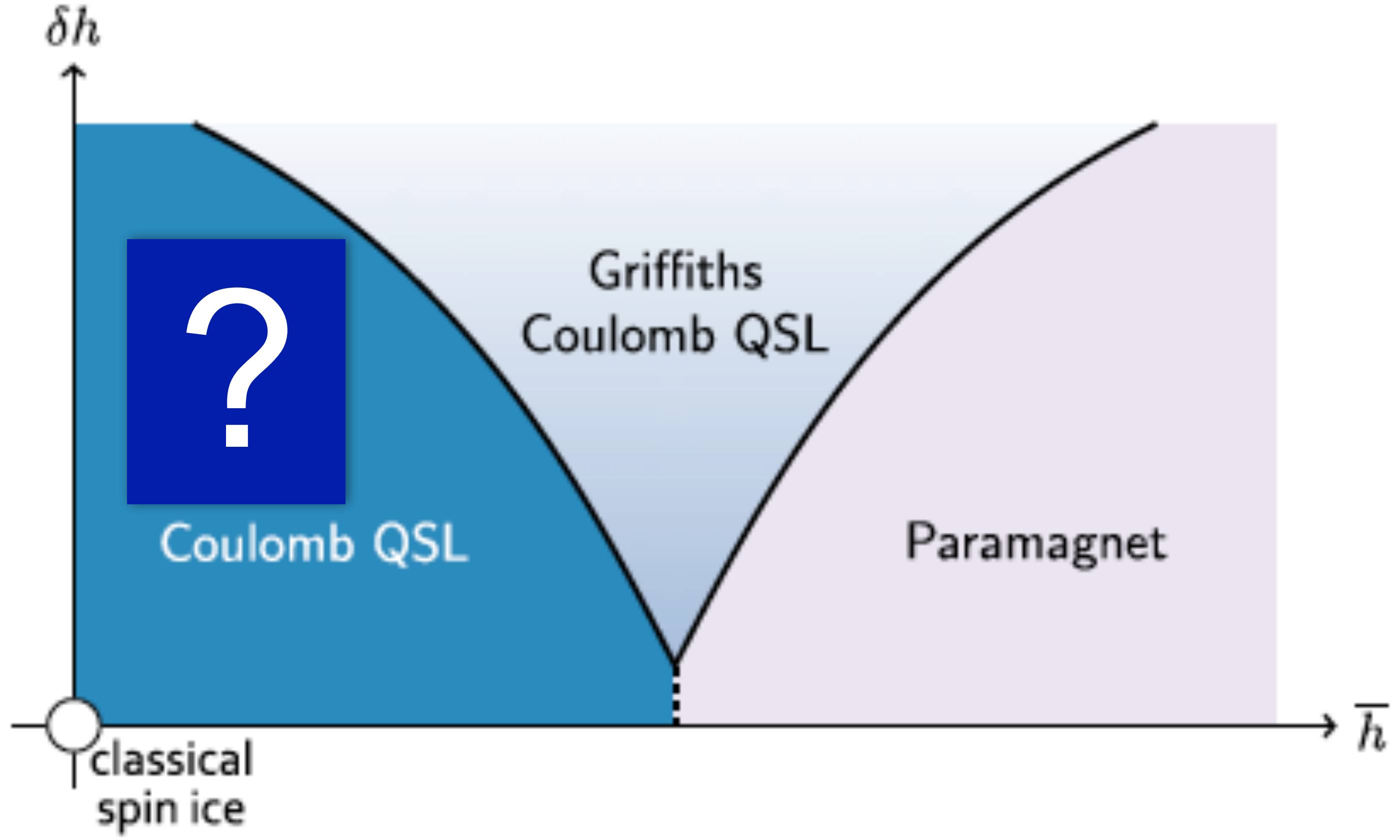
U(1) QSL

trivial paramagnet

The theory question:

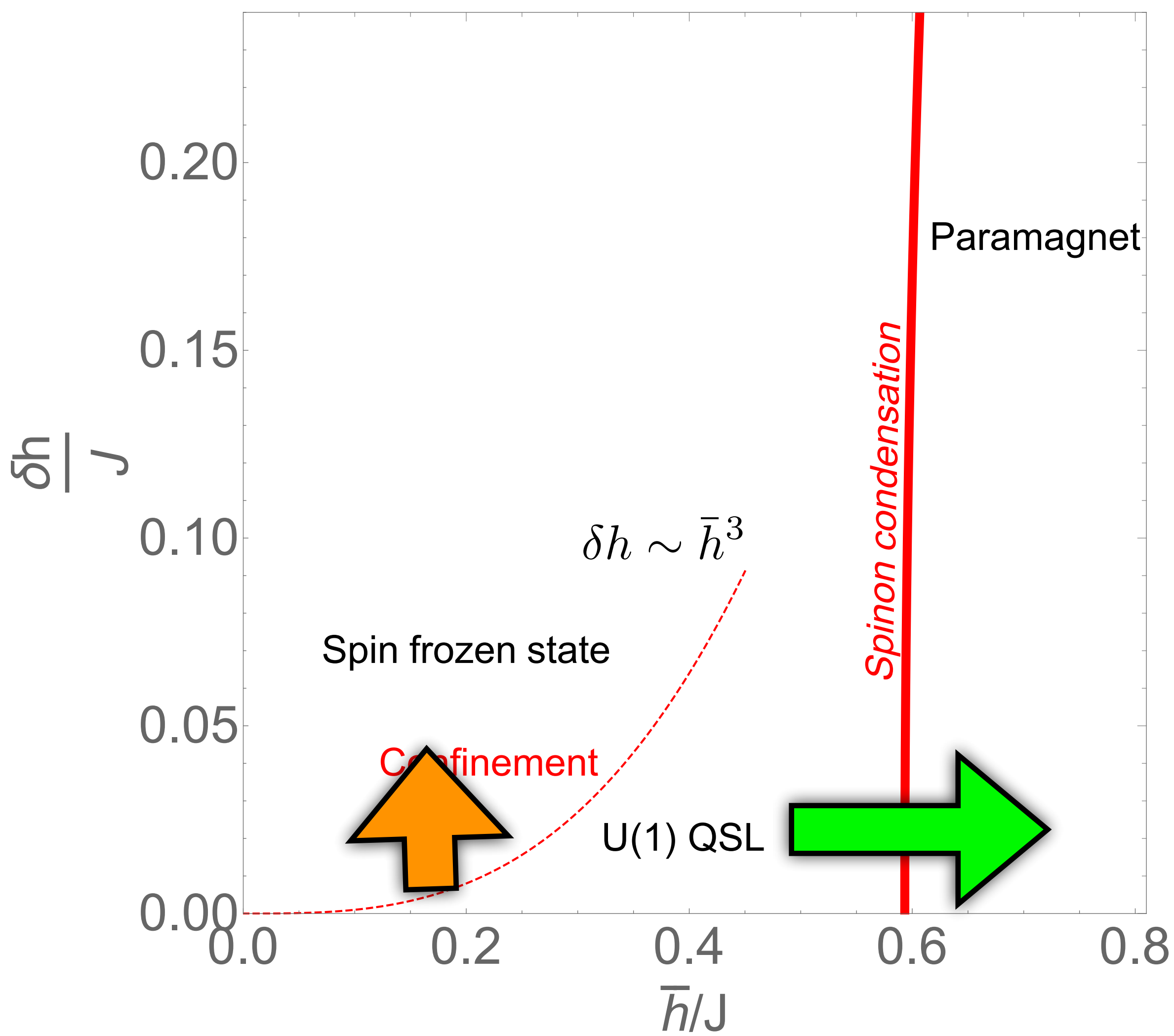
"How stable is the U(1) quantum spin liquid phase of quantum spin ice against disorder in non-Kramers pyrochlores?"

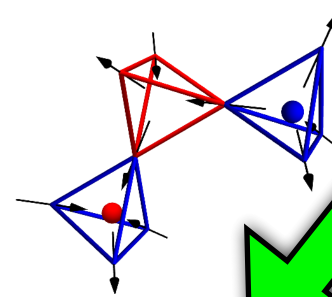
$$\mathcal{H}_{\mathrm{RTFIM}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - \sum_i h_i \sigma_i^x$$



How does the QSL die?

$$\mathcal{H}_{\mathrm{RTFIM}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - \sum_i h_i \sigma_i^x$$





Two main mechanisms

Higgs: condense spinons (spin ice monopoles)

Confinement: condense “dual” monopoles (gapped excitations within ice manifold)

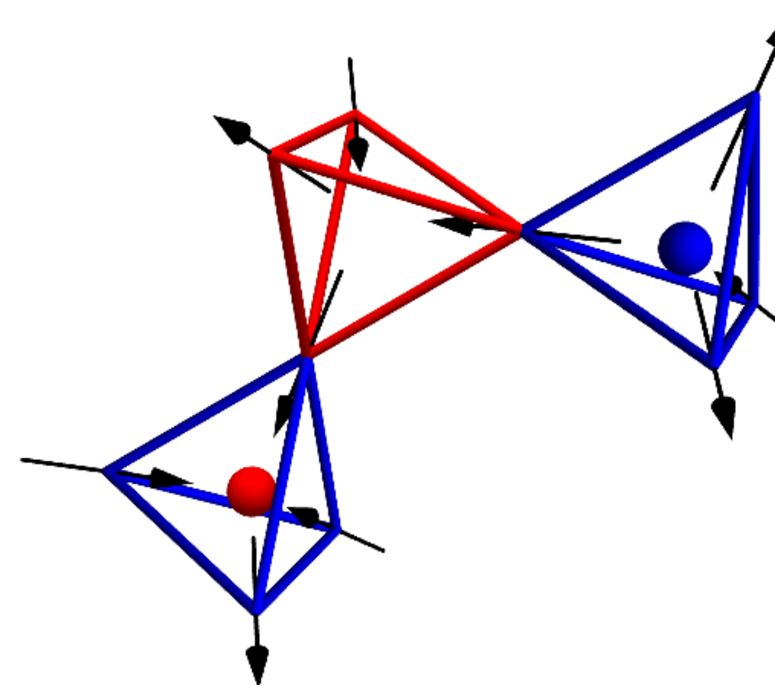
Transverse paramagnet

Frozen moment

Both instablities can be calculated in perturbation theory

Spinon perturbation theory

$$\mathcal{H}_{\mathsf{RTFIM}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - \sum_i h_i \sigma_i^x$$



Consider state of M spinons in limit h_i<<J

$$1 \ll M \ll N$$

allows averaging over spinon environments

allows neglect of spinon-spinon interactions

Degenerate set of states with M spinons has classical energy

$$E(M) = E_0^{\mathsf{cl}} + 2MJ$$

Treat transverse field terms in degenerate perturbation theory

$$\mathcal{H}_{\mathsf{eff}}^{(M)} = E_0^{\mathsf{cl}} + 2MJ + \mathcal{H}_1^{(M)} + \mathcal{H}_2^{(M)}$$

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

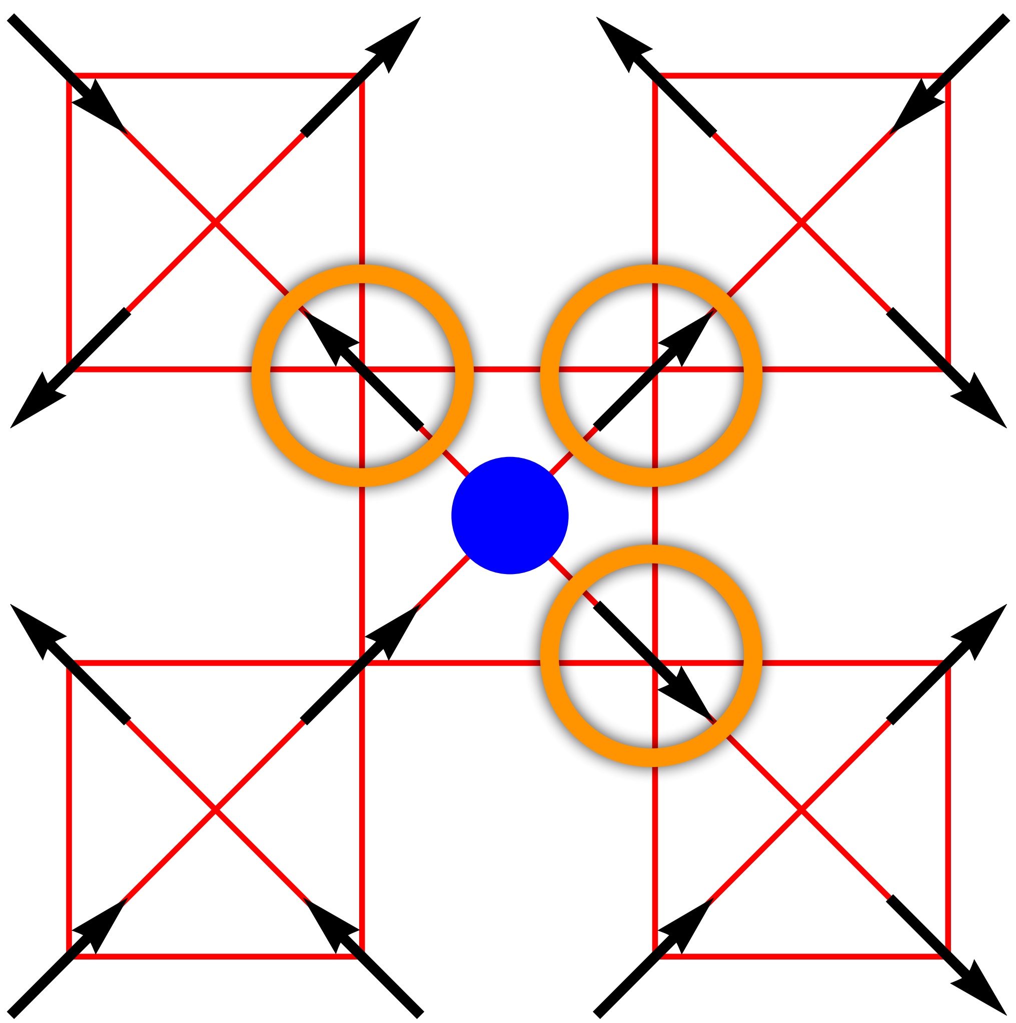
$$\mathcal{H}_2^{(M)} = -\mathcal{P}_M V \frac{1 - \mathcal{P}_M}{\mathcal{H}_0 - (E_0^{\mathsf{cl}} + 2MJ)} V \mathcal{P}_M$$

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

First order perturbation theory

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

Each spinon surrounded by 3 fliipable spins



Column sum:

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

for 1<<M<<N: $= -3M\bar{h}$

independent of β!

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

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

Energy of spinon states

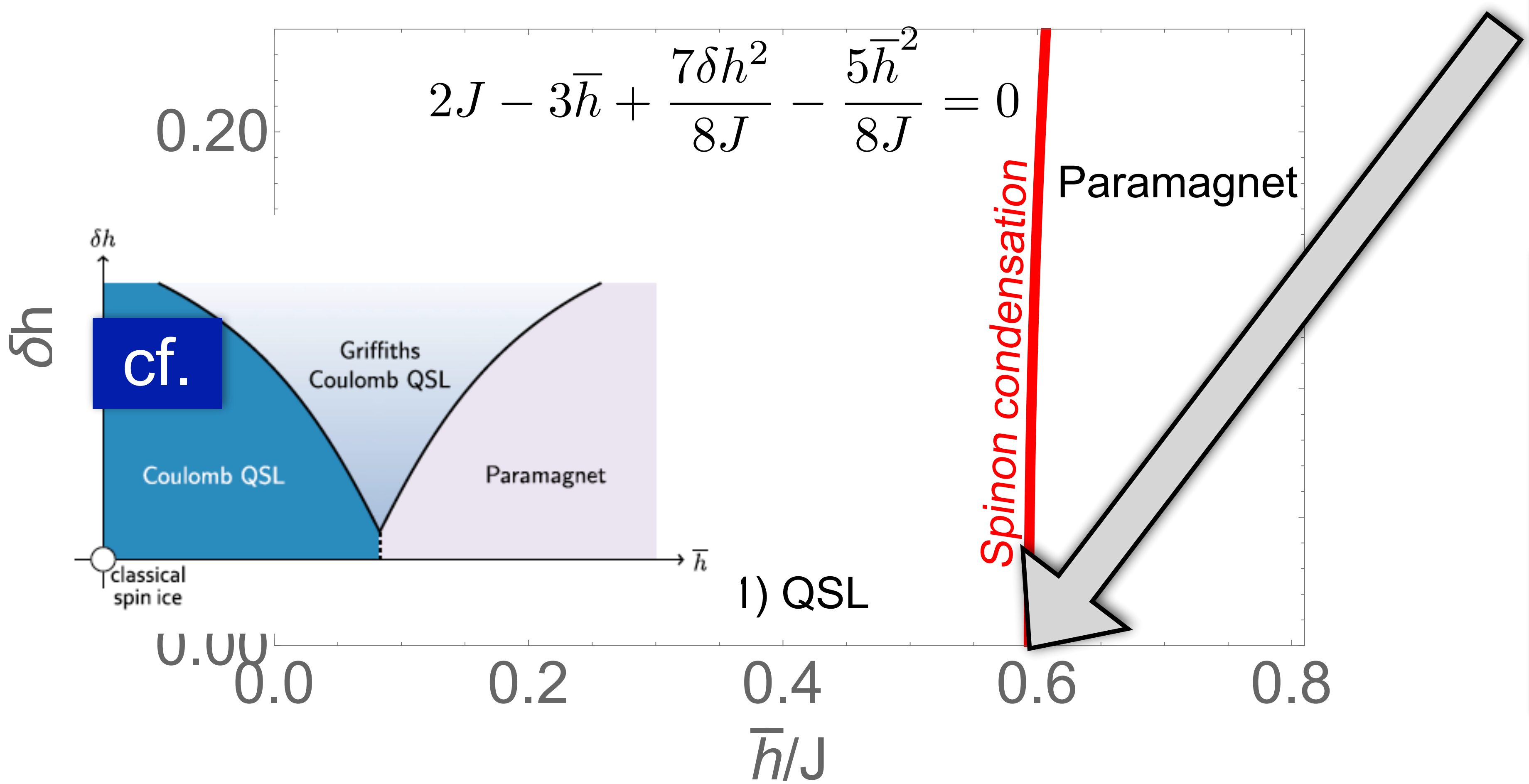
Lowest energy for M spinons (1st order PT)

$$E(M) \approx E_0^{\text{cl}} + M(2J - 3\bar{h})$$

Second order correction

$$E(M) \approx E_0^{\text{cl}} - \frac{N\overline{h^2}}{2J} + M\left(2J - 3\bar{h} + \frac{7\delta h^2 - 5\bar{h}^2}{8J}\right)$$

Instability when coefficient of M becomes negative



Uniform limit (δh=0)

$$h_c \approx 0.593J$$

cf. h_c=0.602J found from high field expansion [Röchner et al, PRB **94**, 201111(R), (2016)]

Instability occurs in “typical” regions- may be preceeded by Griffiths phase

Confinement

Confinement comes from the condensation of dual monopoles which are excitations within the ice manifold

Degenerate perturbation theory amongst classical spin ice ground states with non-uniform transverse field

Fourth order perturbation renormalizes Ising interaction

$$\mathcal{H} = \sum_{\langle ij \rangle} J'_{ij} \sigma_i^z \sigma_j^z \qquad J'_{ij} = J + \frac{h_i^2 h_j^2}{48 J^3}$$

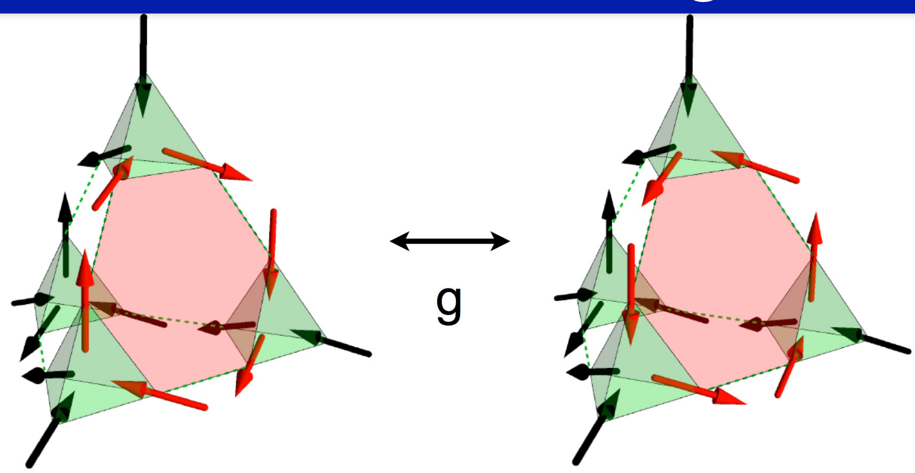
Creates energetic preference to put AFM bonds between pairs of sites with large values of h

Breaks ice degeneracy and favours realization dependent frozen spin state

Competition between spin freezing and spin liquid

Take limit $\delta h \ll \overline{h} \ll J$ weak "nearly uniform", transverse field

At δh=0 fourth order term is a constant so leading non-trivial term is 6th order ring exchange which stabilizes QSL



$$g = -\frac{63h^6}{256J^5}$$

Small δh: renormalized Ising interaction splits ice manifold by amount ε

$$J'_{ij} = J + \frac{h_i^2 h_j^2}{48J^3}$$

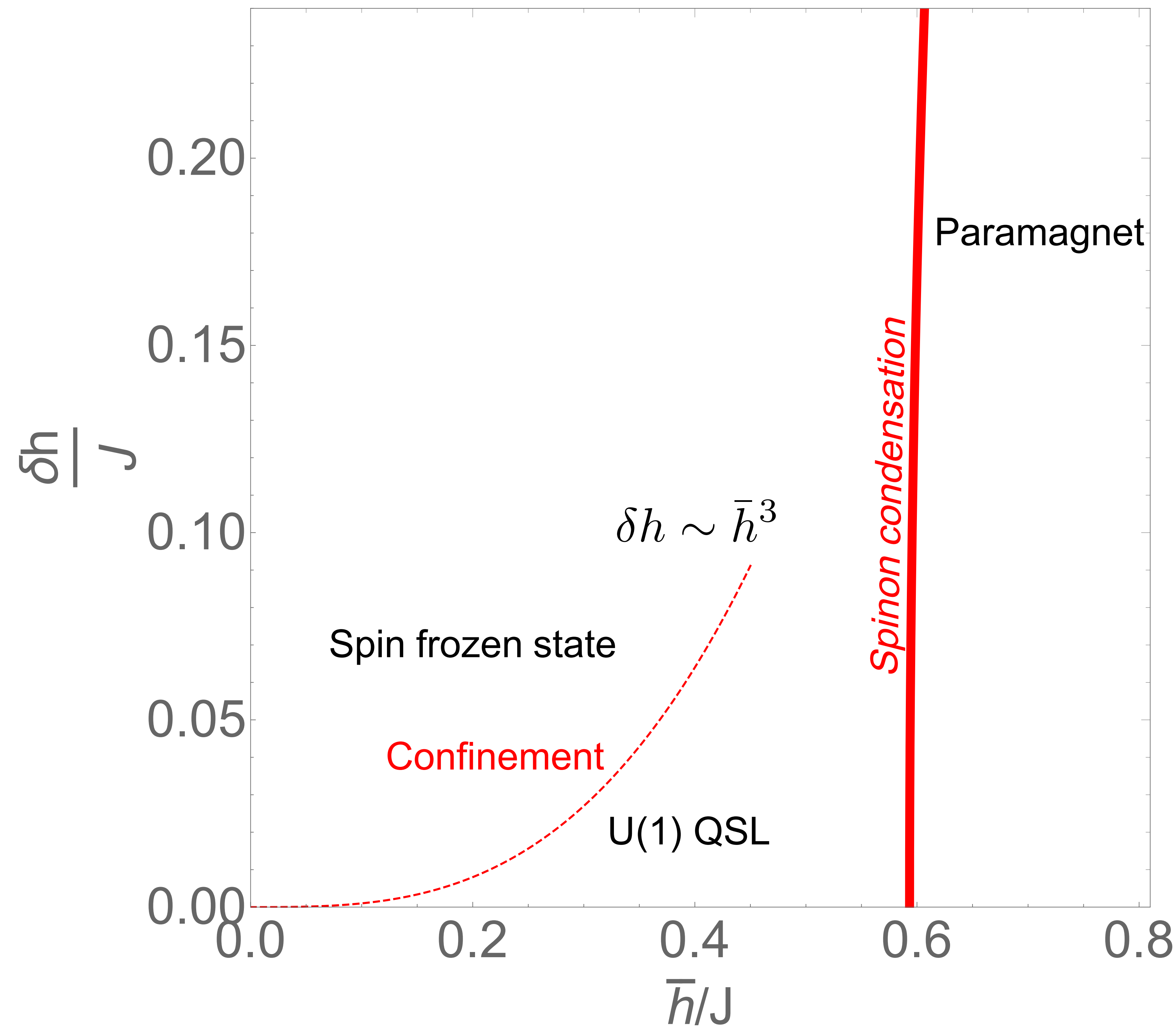
$$\epsilon \sim \overline{h}^3 \delta h$$

Ground state decided by competition between ε and g

Estimate phase transition

$$\epsilon \sim g \implies \overline{h}^3 \delta h \sim \overline{h}^6 \implies \delta h \sim \overline{h}^3$$

Instabilities of the U(1) QSL



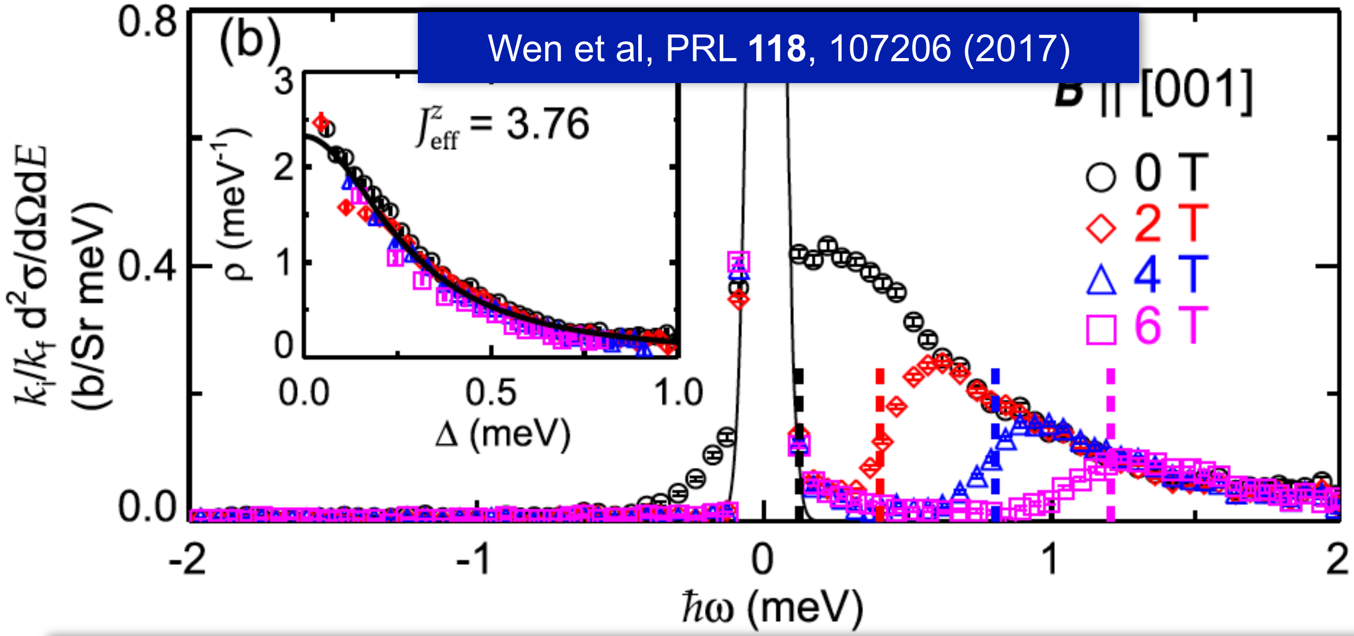
The experimental question:

"Do presently studied samples of $Pr_2Zr_2O_7$ support a quantum spin liquid ground state?"

Need to know the interaction strength and disorder distribution

Parameterising a model for $Pr_2Zr_2O_7$

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



Inelastic neutron scattering in field: inferred Lorentzian distribution of transverse fields

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

fits find width parameter

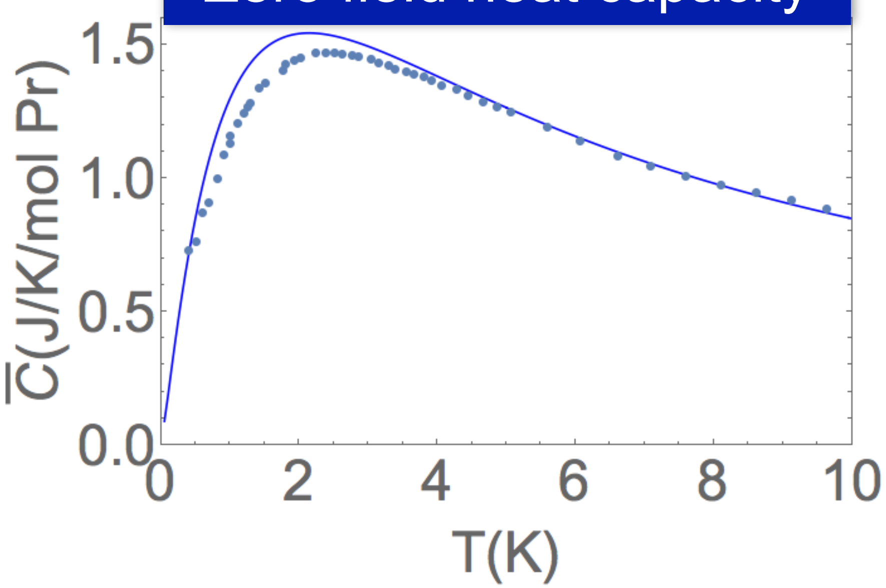
$$\Gamma = 0.27\mathrm{meV}$$

Alternative approach: fit to heat capacity and susceptibility using disorder averaged Numerical Linked Cluster expansion

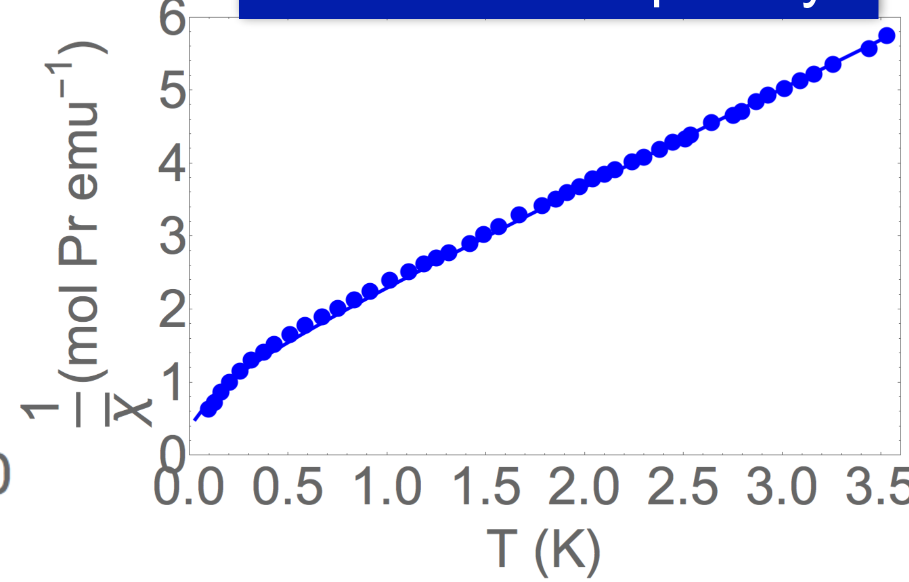
Continue to assume Lorentzian distribution. Adjustable parameters are J, Γ and effective moment μ_{eff}

Find: J=0.02 meV; Γ=0.2 meV; μ_{eff}=2.45 μ_B

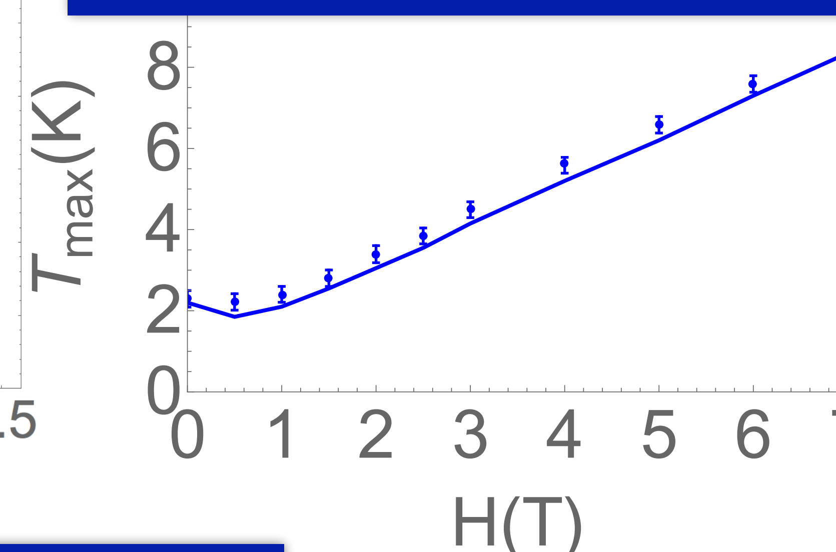
Zero field heat capacity



Inverse susceptibility

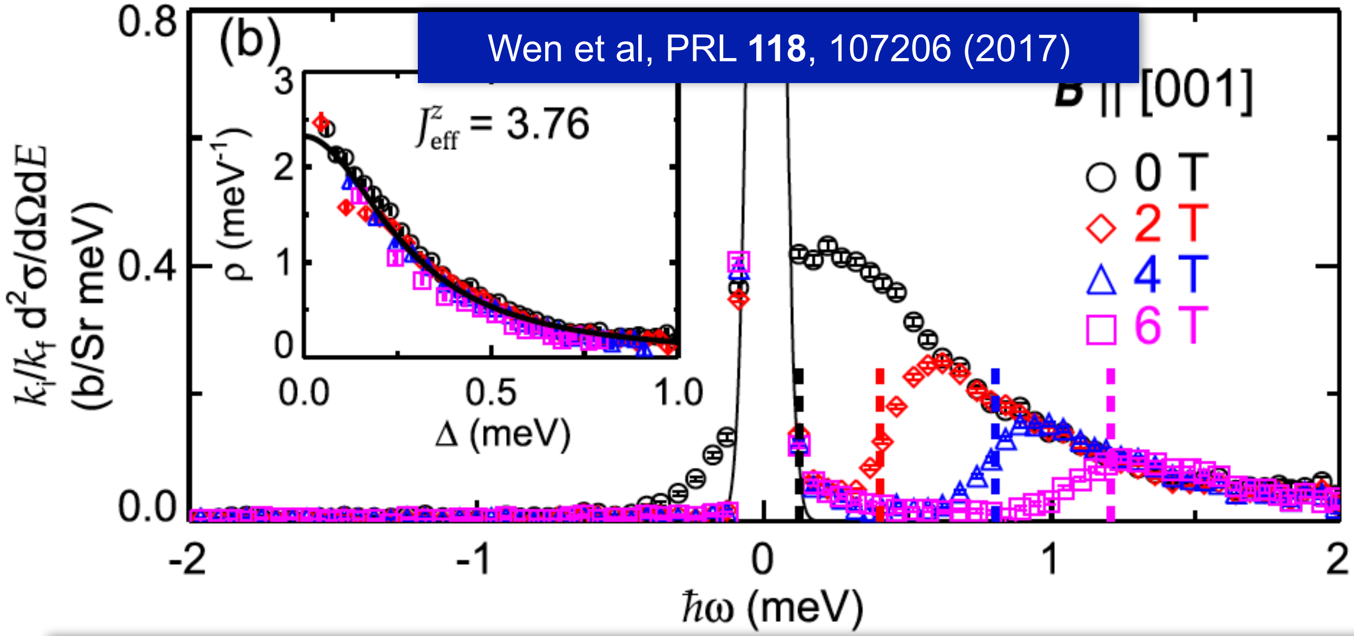


Temperature of heat capacity maximum in 110 field



Data from: Petit et al, PRB **94**, 165153 (2016)

Parameterising a model for $Pr_2Zr_2O_7$



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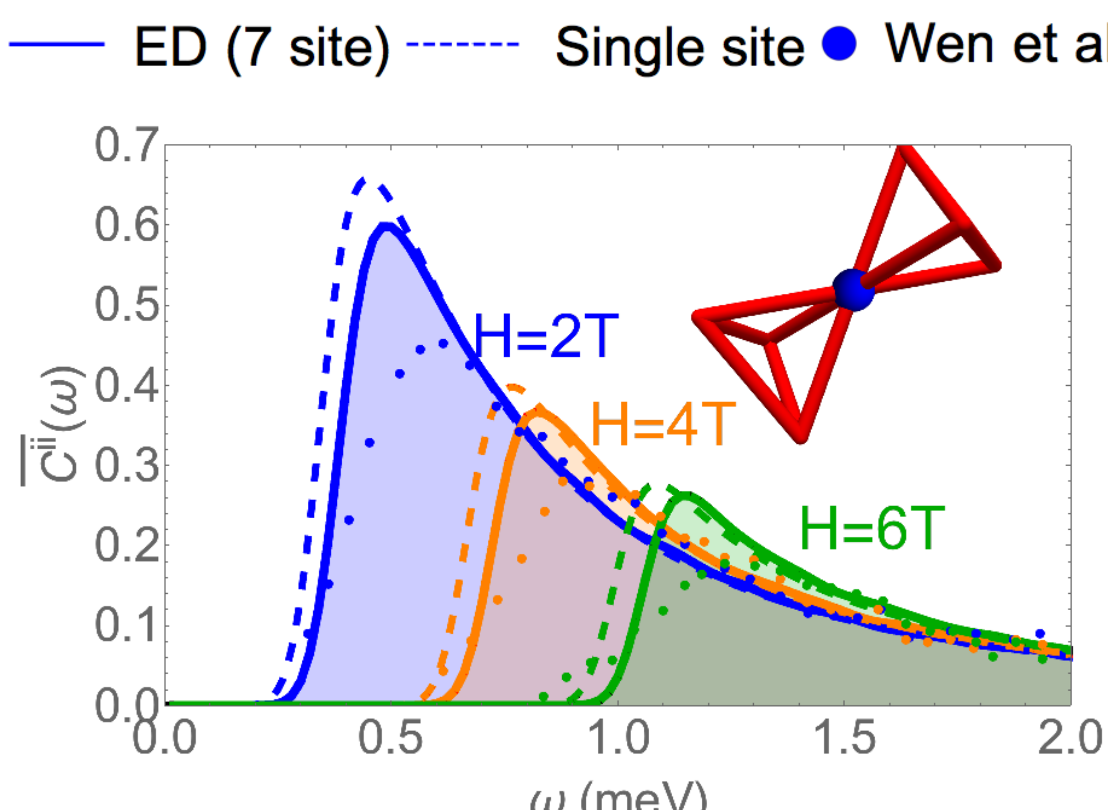
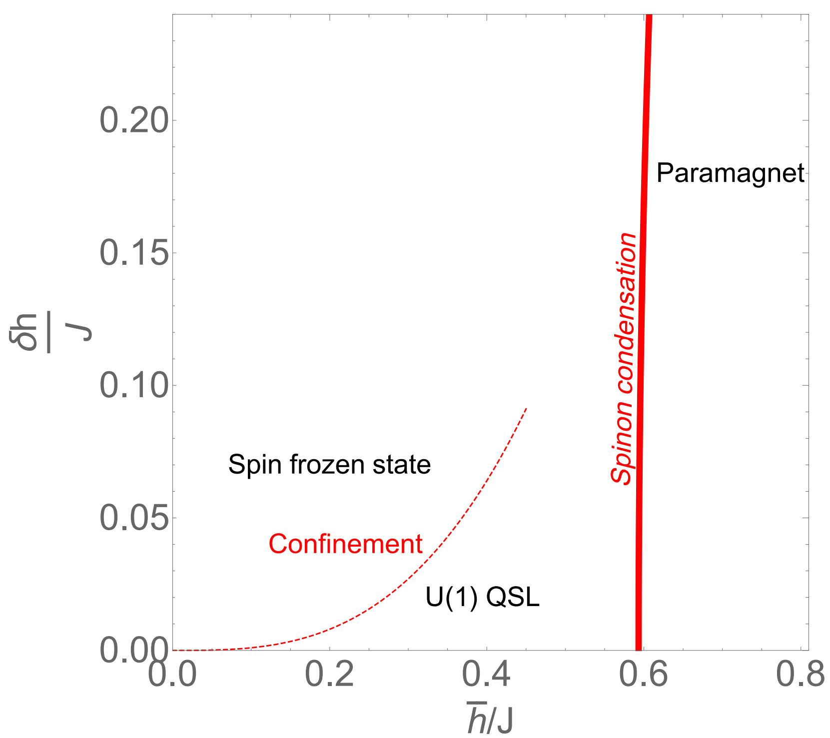
Fit to thermodynamics of Petit et al's sample still gives reasonable agreement with neutron scattering data in field from Wen et al

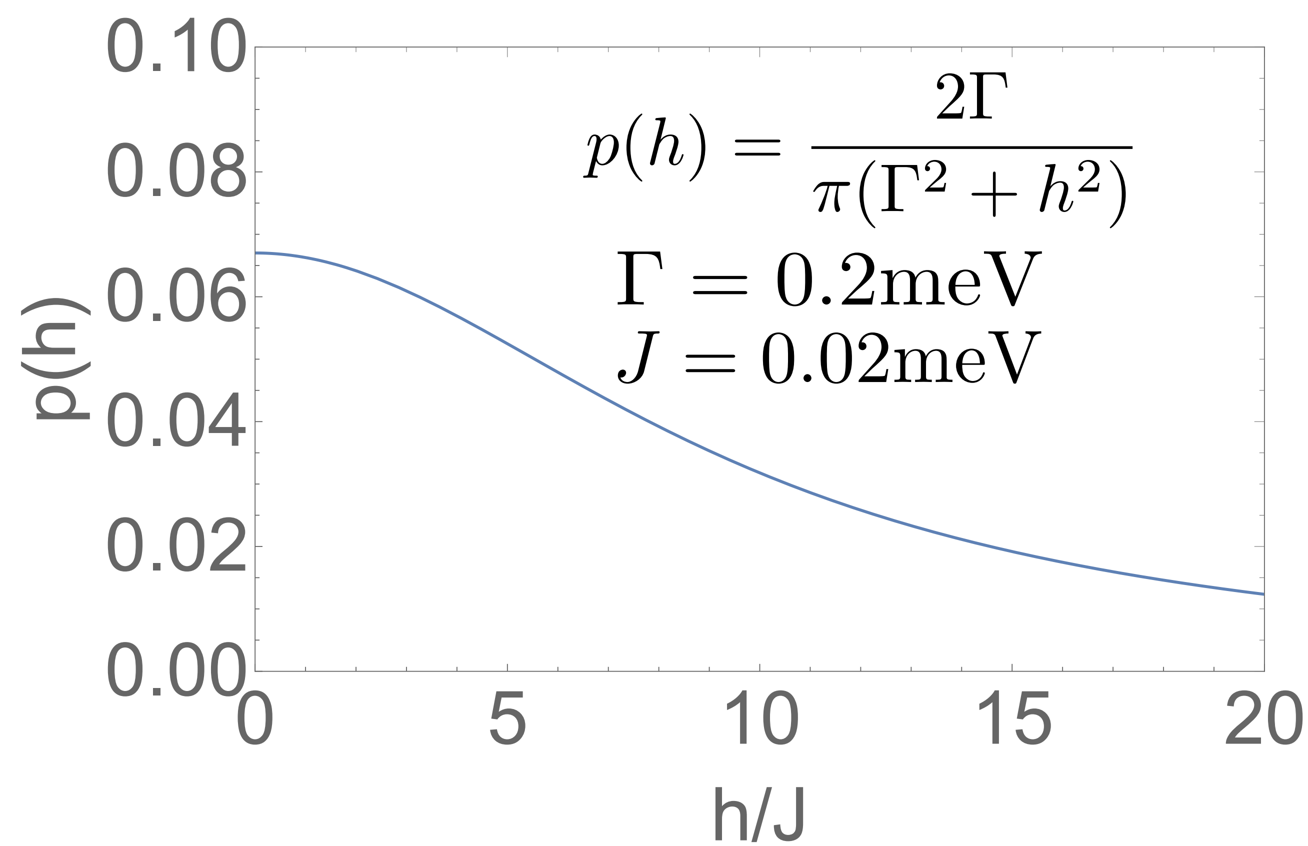


What does that mean for the ground state?

h=0.6 J is enough to close spinon gap

Typical values of h are ~ 10 J !





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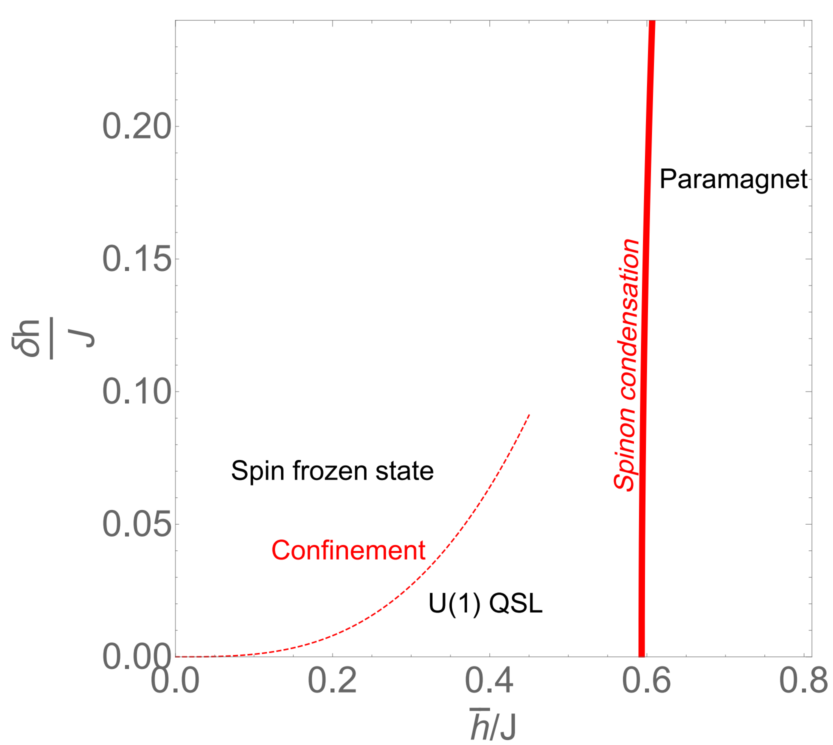
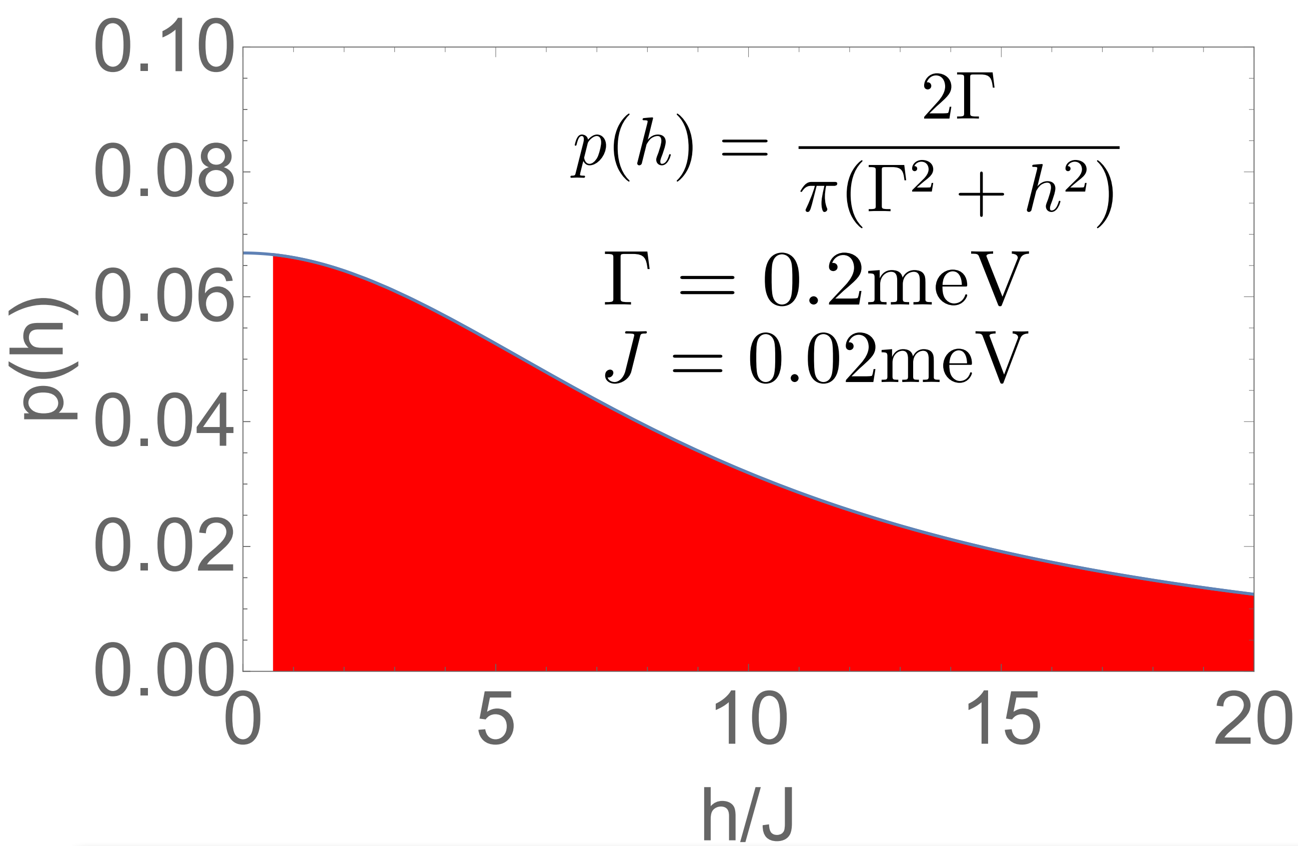




h_i>0.6 J on ~96% of sites

suggests system deep in paramagnetic phase

ED/NLC calculation of transverse polarisation:

$$\overline{\langle \sigma^x \rangle} \approx 0.98$$

pseudospins are strongly polarized by transverse fields

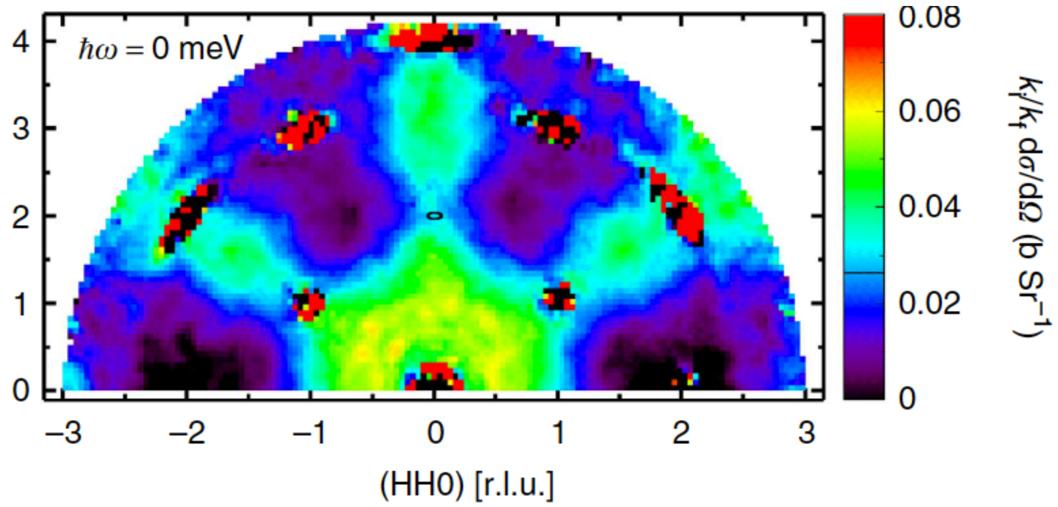
What about the spin correlations?

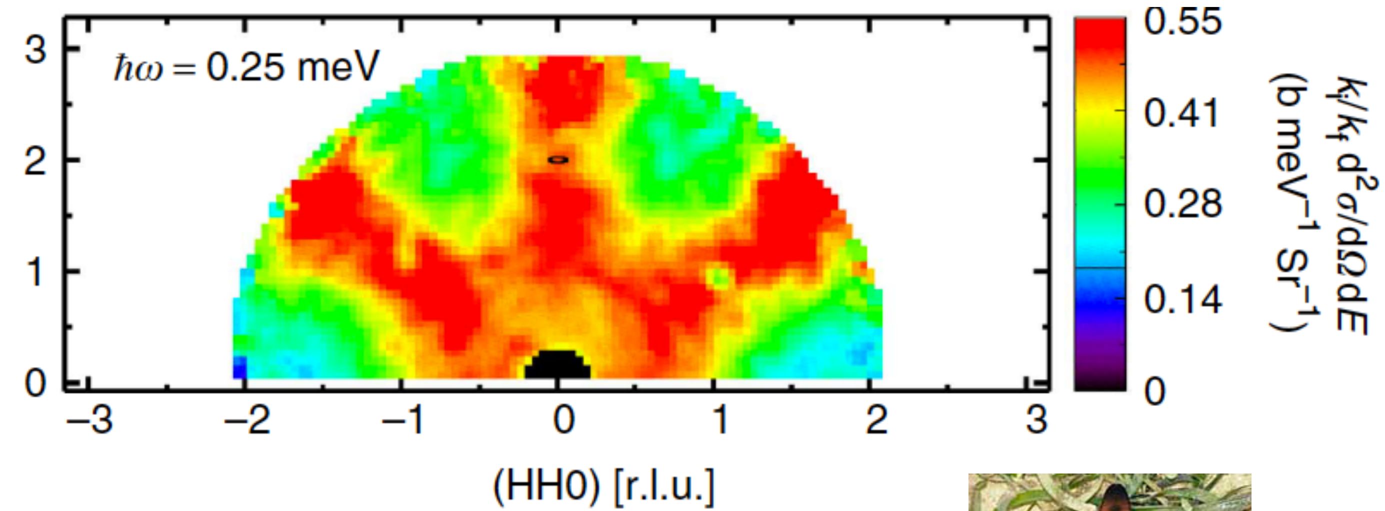
Elastic/low energy scattering

Higher energy scattering

$Pr_2Zr_2O_7$

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





Spin-ice-like correlations

Starfish

can these observations be consistent with a paramagnetic ground state?

What about the spin correlations?

Elastic/low energy scattering

Higher energy scattering

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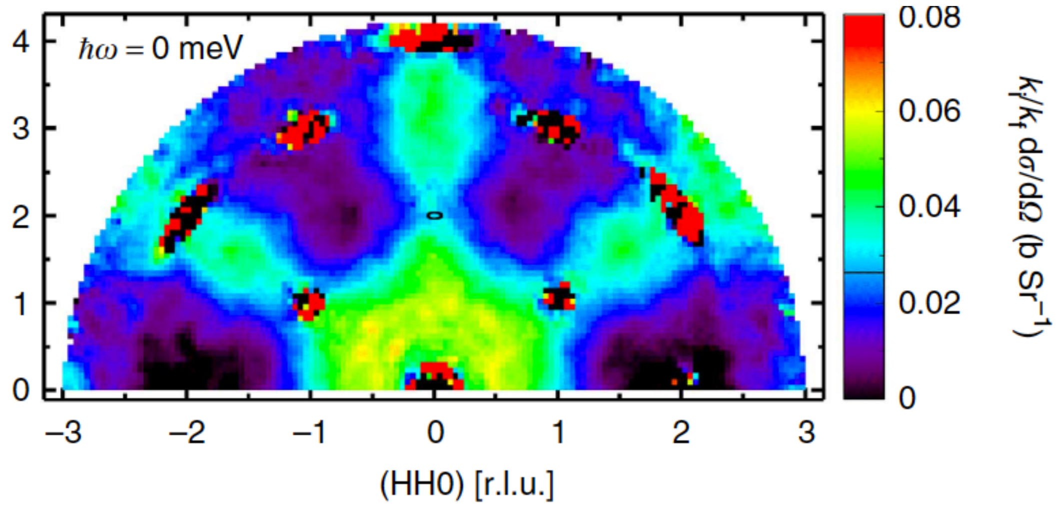
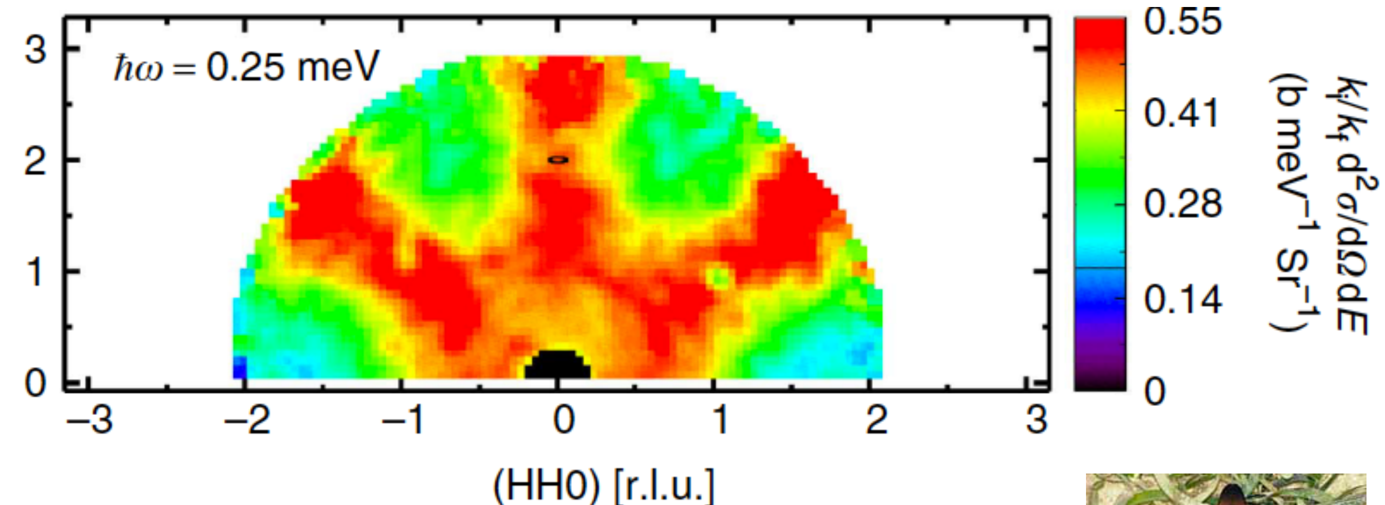
Kimura et al, Nature Commun. **4**, 1934 (2013)

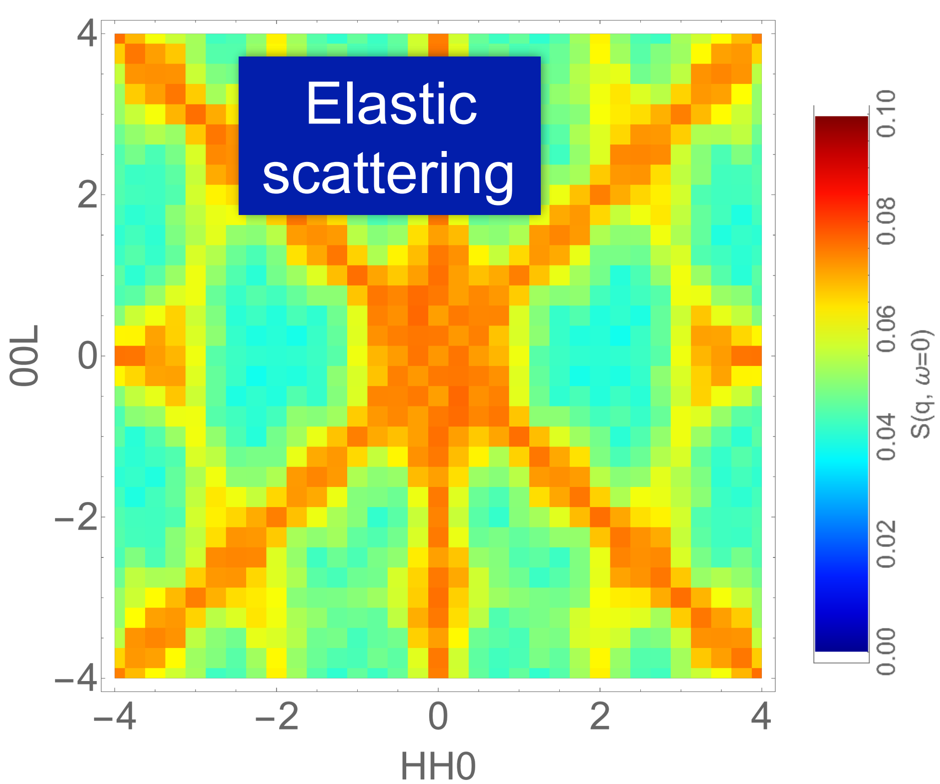




Spin-ice-like correlations

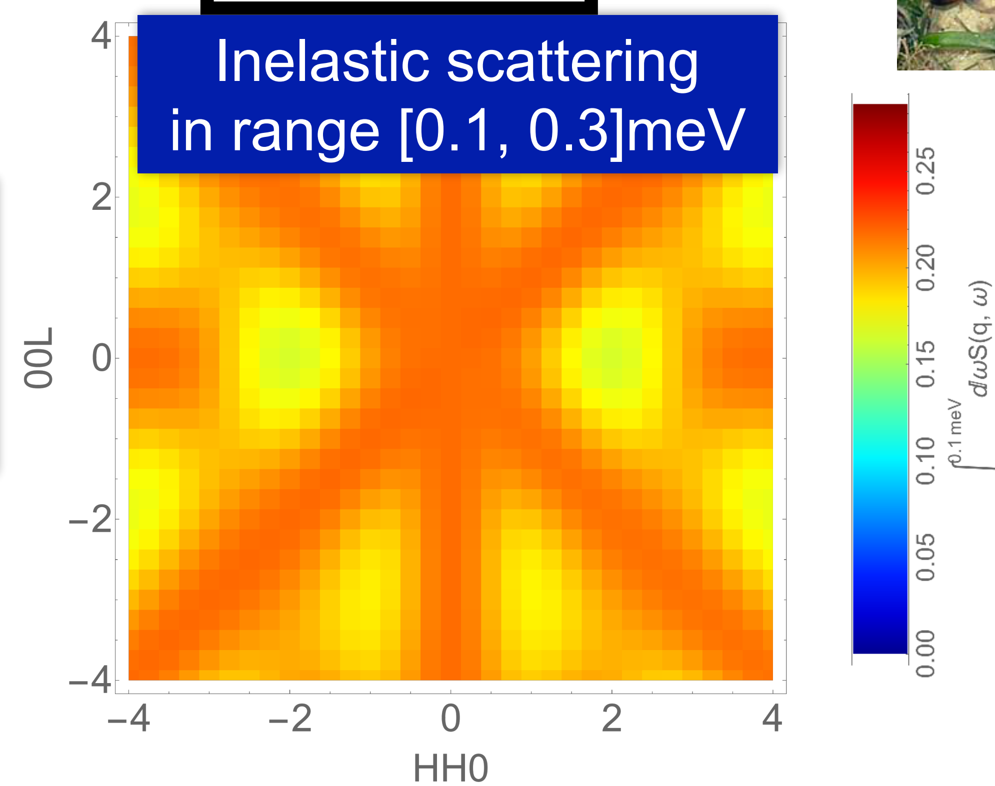
Starfish

Elastic scattering



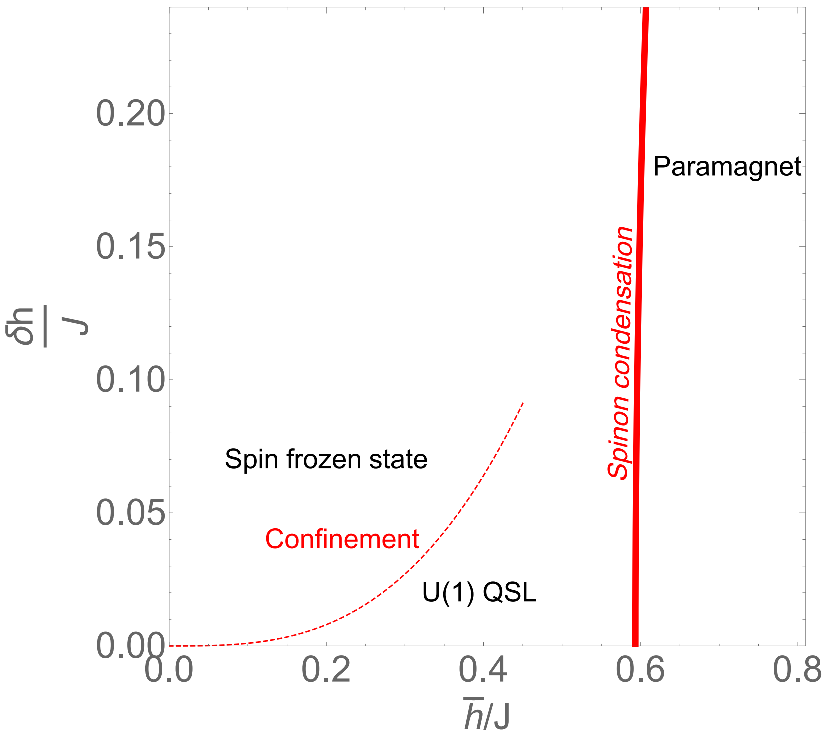
Calculations assuming paramagnetic ground state

Inelastic scattering in range [0.1, 0.3]meV



N=1024 sites, average over 360 realizations of disorder

Summary



Perturbative theory of the instabilities of the U(1) QSL phase in non-Kramers quantum spin ice with disorder

Parameterisation of model suggests current samples of $Pr_2Zr_2O_7$ are deep in the paramagnetic phase

Present zero-field inelastic neutron scattering data is consistent with paramagnetic ground state

Thank you for listening!