Modeling suspected spin liquids on the pyrochlore lattice $NaCaNi_2F_7$ and $MgCr_2O_4$

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11/20/2019 KITP

Spin liquids

- Exactly solvable models
  - Topological degeneracy of the ground state
  - Long-range quantum entanglement
  - Fractionalized excitations
- Material candidates and experiments
  - Transport
  - Spectroscopy
- This talk
  - Spin-1 $NaCaNi_2F_7$ and spin-3/2 $MgCr_2O_4$
  - Antiferromangetic Heisenberg model on a pyrochlore lattice
  - Neutron scattering probe of spin correlations
  - Modeling the spin dynamics

References

Article | Published: 29 October 2018

Continuum of quantum fluctuations in a three-dimensional S = 1 Heisenberg magnet

K. W. Plumb, Hitesh J. Changlani, A. Scheie, Shu Zhang, J. W. Krizan, J. A. Rodriguez-Rivera, Yiming Qiu, B. Winn, R. J. Cava & C. L. Broholm

Nature Physics **15**, 54–59 (2019) | Download Citation

arXiv.org > cond-mat > arXiv:1810.09481

Condensed Matter > Strongly Correlated Electrons

Dynamical structure factor of the three-dimensional quantum spin liquid candidate $NaCaNi_2F_7$

Shu Zhang, Hitesh J. Changlani, Kemp W. Plumb, Oleg Tchernyshyov, Roderich Moessner

(Submitted on 22 Oct 2018)

arXiv.org > cond-mat > arXiv:1810.11869

Condensed Matter > Strongly Correlated Electrons

Magnetic excitations of the classical spin liquid MgCr2O4

X. Bai, J. A. M. Paddison, E. Kapit, S. M. Koohpayeh, J.-J. Wen, S. E. Dutton, A. T. Savici, A. I. Kolesnikov, G. E. Granroth, C. L. Broholm, J. T. Chalker, M. Mourigal

(Submitted on 28 Oct 2018)

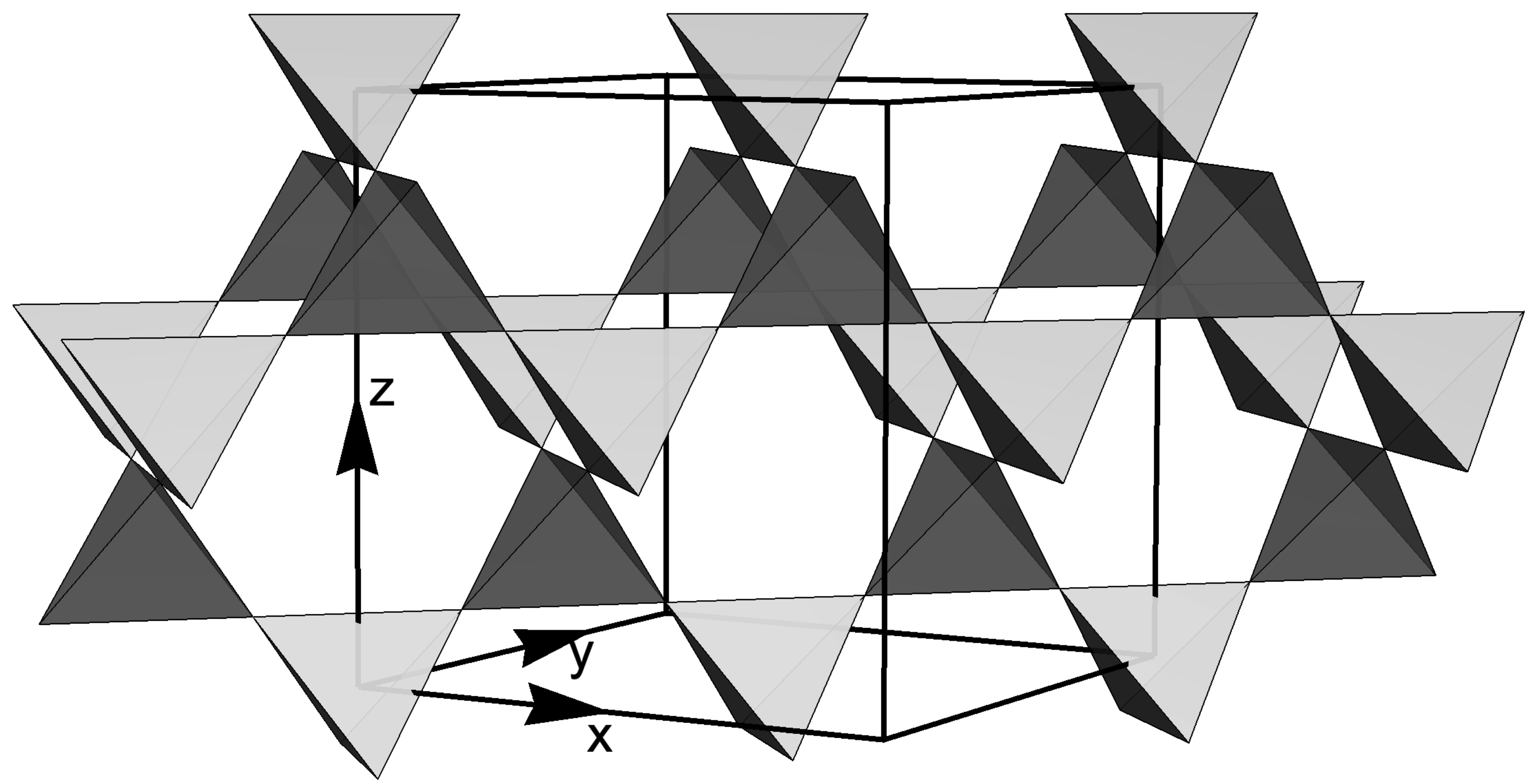
Outline

- Heisenberg pyrochlore antiferromagnet:
    - Classical model: A classical spin liquid
    - Quantum model: A quantum spin liquid?
    - Material realizations
        - $NaA'B_2F_7$ (A' = Ca, B = Ni spin-1; A' = Sr, B = Mn spin-5/2)
        - ACr_2O_4 (A = Cd; Zn; Mg spin-3/2; …)
        - …
- A general picture for spin dynamics
    - Theoretical modeling of $NaCaNi_2F_7$ and $MgCr_2O_4$
- Discussion

Heisenberg pyrochlore antiferromagnet

Pyrochlore: corner-sharing tetrahedra

Antiferromagnetic Heisenberg interaction $H = J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j, \quad J > 0$



Heisenberg pyrochlore antiferromagnet

Pyrochlore: corner-sharing tetrahedra

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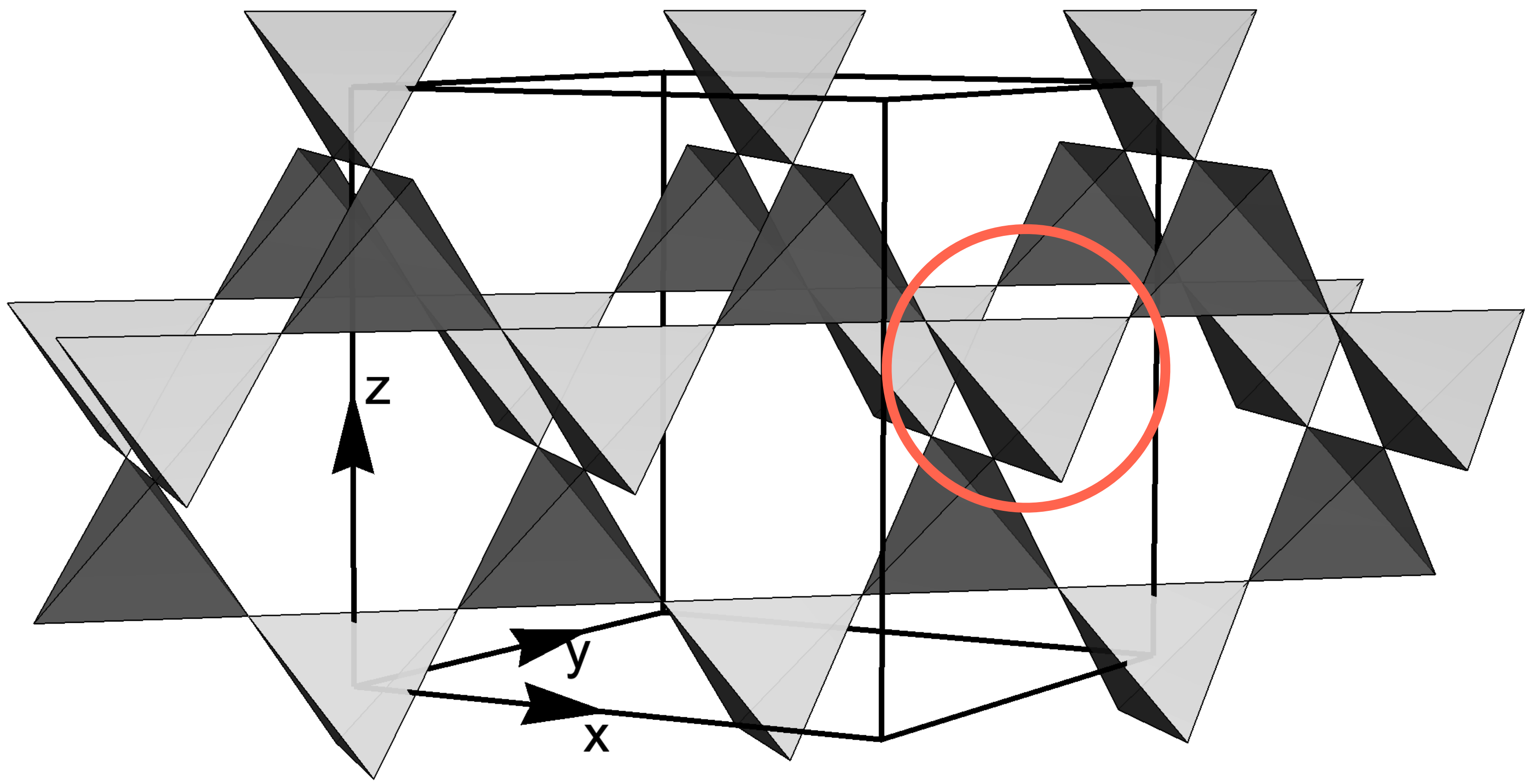


Ground state is macroscopically degenerate

$$H_{\boxtimes} = \frac{J}{2}\left(\sum_{i \in \boxtimes} \mathbf{s}_i\right)^2 + \text{const.} \implies \sum_{i \in \boxtimes} \mathbf{s}_i = 0$$

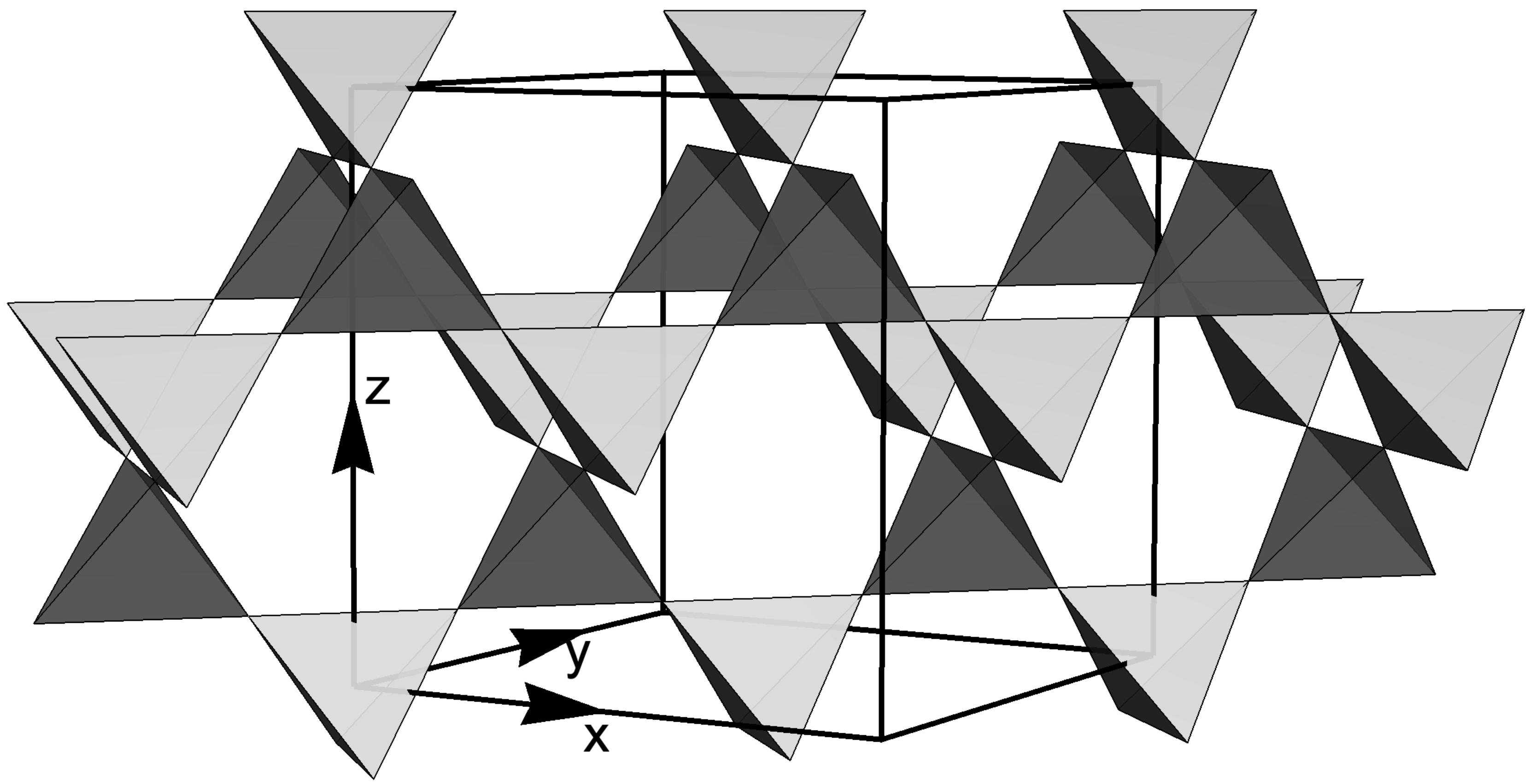
Magnetic frustration

Moessner and Chalker 1998

Heisenberg pyrochlore antiferromagnet

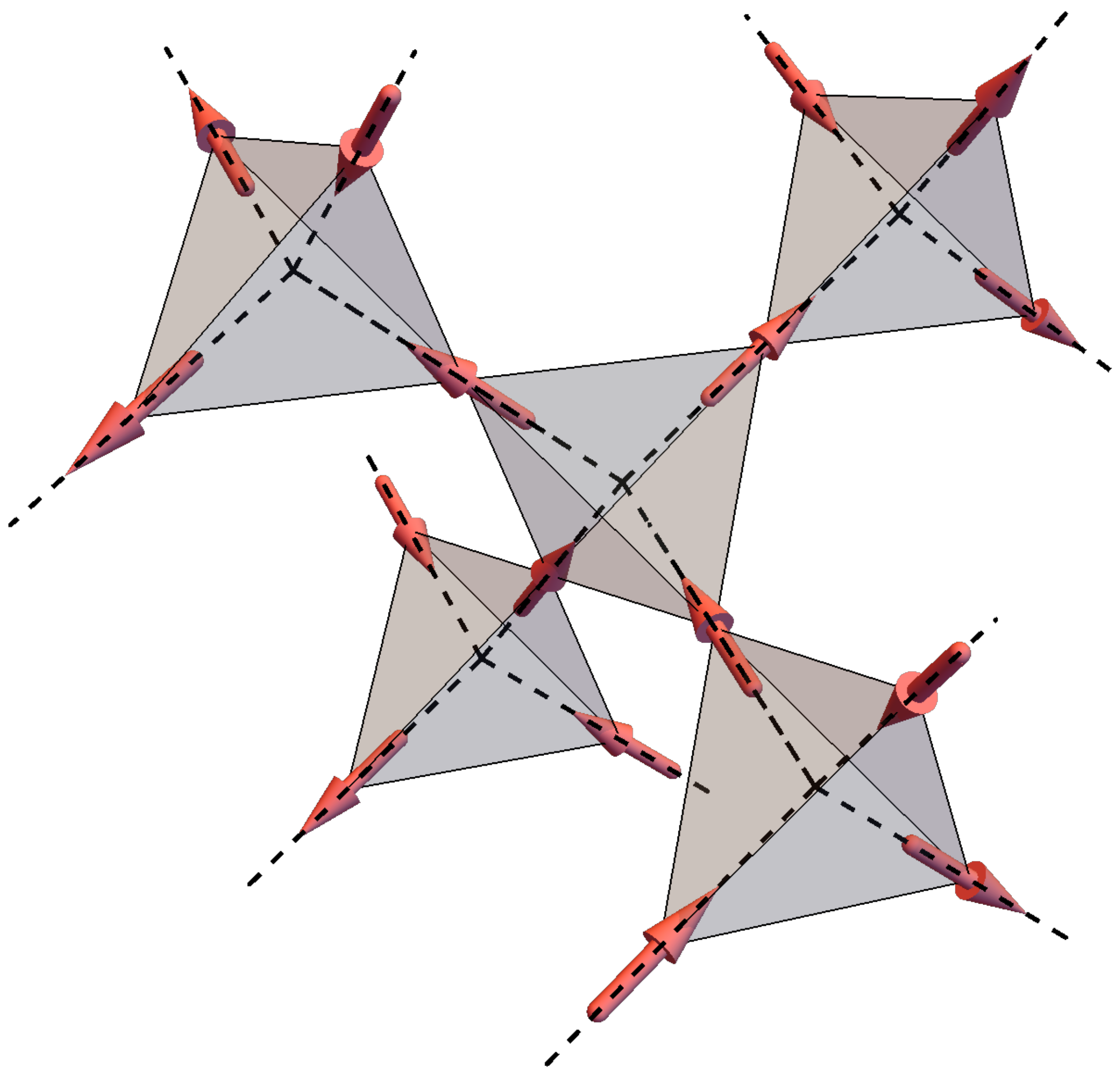
Pyrochlore: corner-sharing tetrahedra

Antiferromagnetic Heisenberg interaction $H = J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j, \quad J > 0$



Searching for the Coulomb phase…

Spin ice



$$H = J \sum_{\langle ij \rangle} \sigma_i \sigma_j, \quad J > 0$$

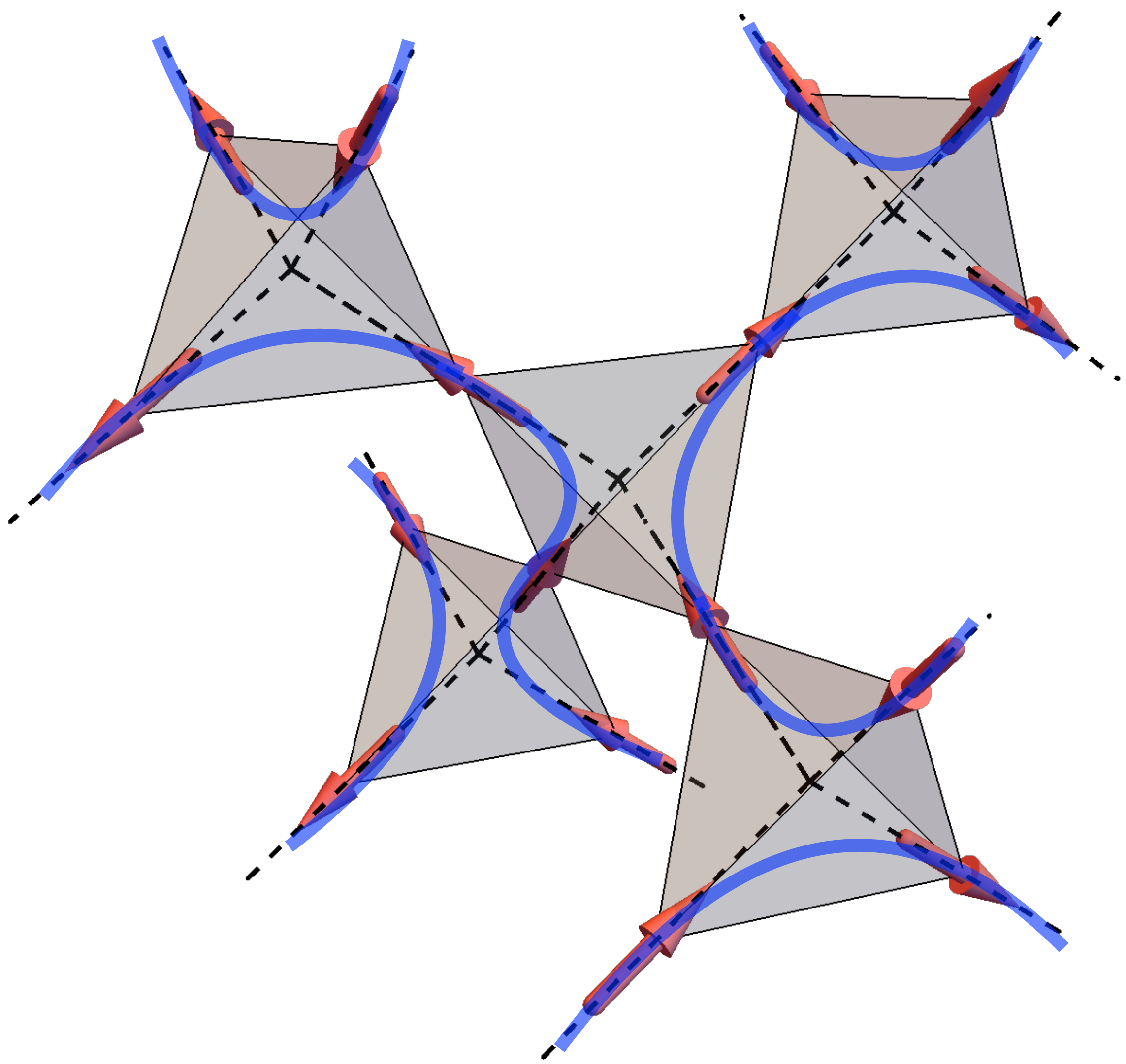
$$\sigma = \pm 1$$

$$\sum_{i \in \boxtimes} \sigma_i = 0$$

Ice rule: two in two out

Gingras and McClarty 2014

Spin ice



Divergence free magnetic field

$\nabla \cdot \mathbf{B} = 0$

Magnetostatics

$$\mathcal{Z}[\mathbf{B}(\mathbf{r})] = \int \mathcal{D}(\mathbf{B}) \exp\left[-\frac{\beta}{8\pi}\int d^3\mathbf{r}\ \mathbf{B}^2\right] \qquad \beta = 1/k_B T$$

$\nabla \cdot \mathbf{B} = 0 \qquad \mathbf{B} = \nabla \times \mathbf{A}$ Vector potential

Momentum space

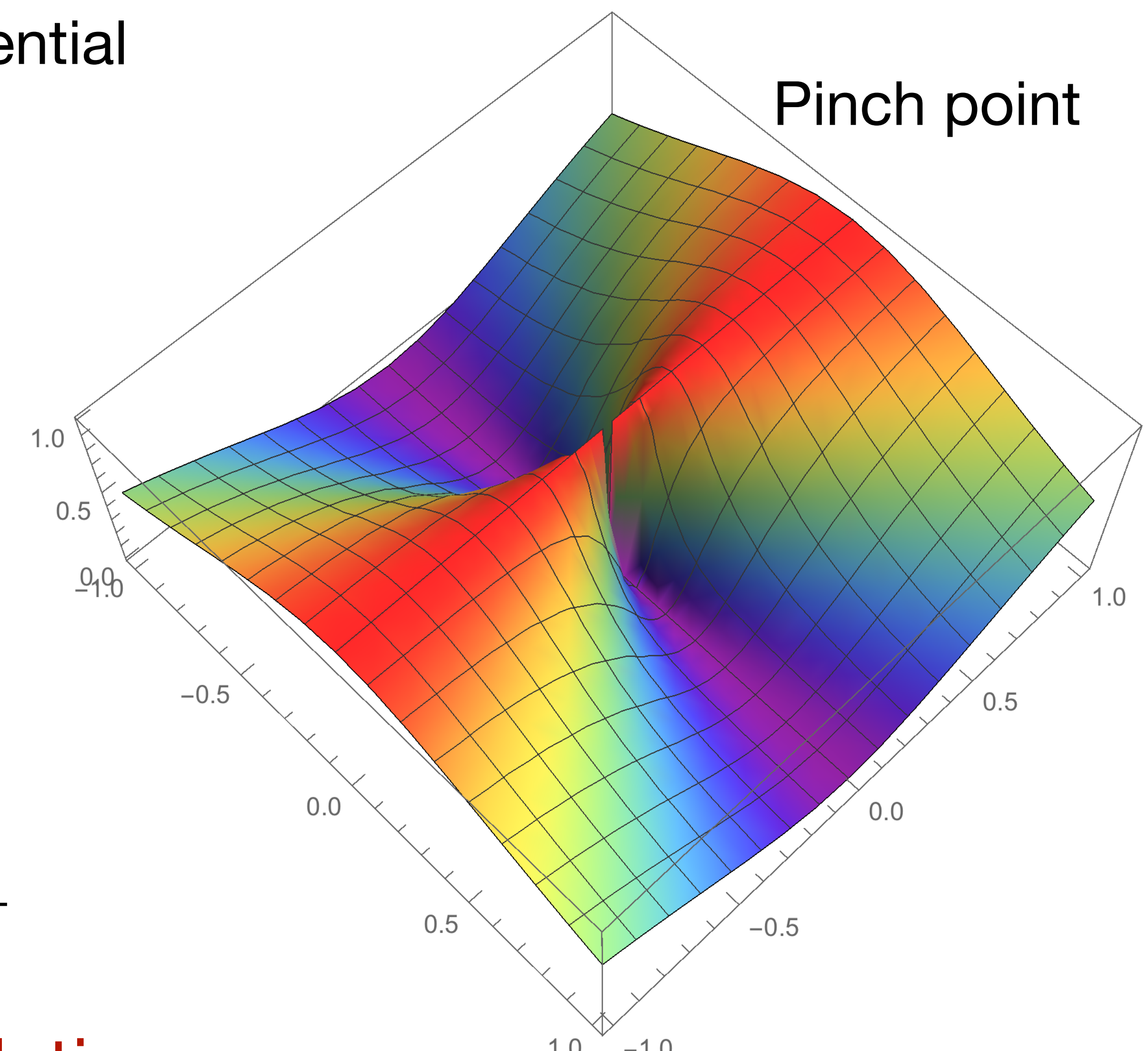
$$\langle B_\mu(\mathbf{q}) B_\nu(-\mathbf{q})\rangle \propto \delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2}$$

Magnetostatics

$$\mathcal{Z}[\mathbf{B}(\mathbf{r})] = \int \mathcal{D}(\mathbf{B}) \exp\left[-\frac{\beta}{8\pi}\int d^3\mathbf{r}\ \mathbf{B}^2\right] \qquad \beta = 1/k_B T$$

$\nabla \cdot \mathbf{B} = 0 \qquad \mathbf{B} = \nabla \times \mathbf{A}$ Vector potential

Pinch point



Momentum space

$$\langle B_\mu(\mathbf{q}) B_\nu(-\mathbf{q})\rangle \propto \delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2}$$

$$\langle B_z(\mathbf{q}) B_z(-\mathbf{q})\rangle\Big|_{q_x=0} \propto \frac{q_y^2}{q_z^2 + q_y^2}$$

Real space

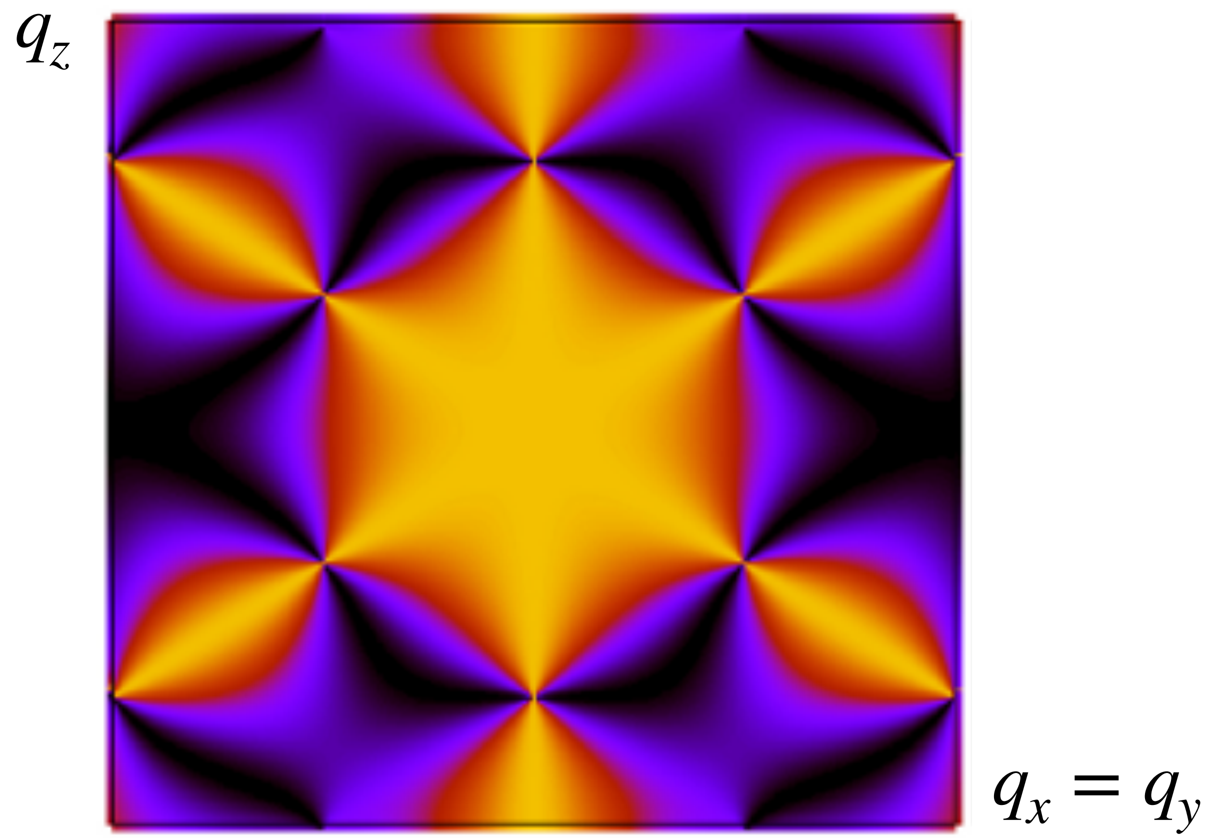
$$\langle B_\mu(\mathbf{r}) B_\nu(0)\rangle \propto \frac{3 r_\mu r_\nu - \delta_{\mu\nu} r^2}{r^5}$$

Dipolar correlation

Oleg Tchernyshyov

Coulomb phase

Ising $\sum_{i \in \boxtimes} \sigma_i = 0$



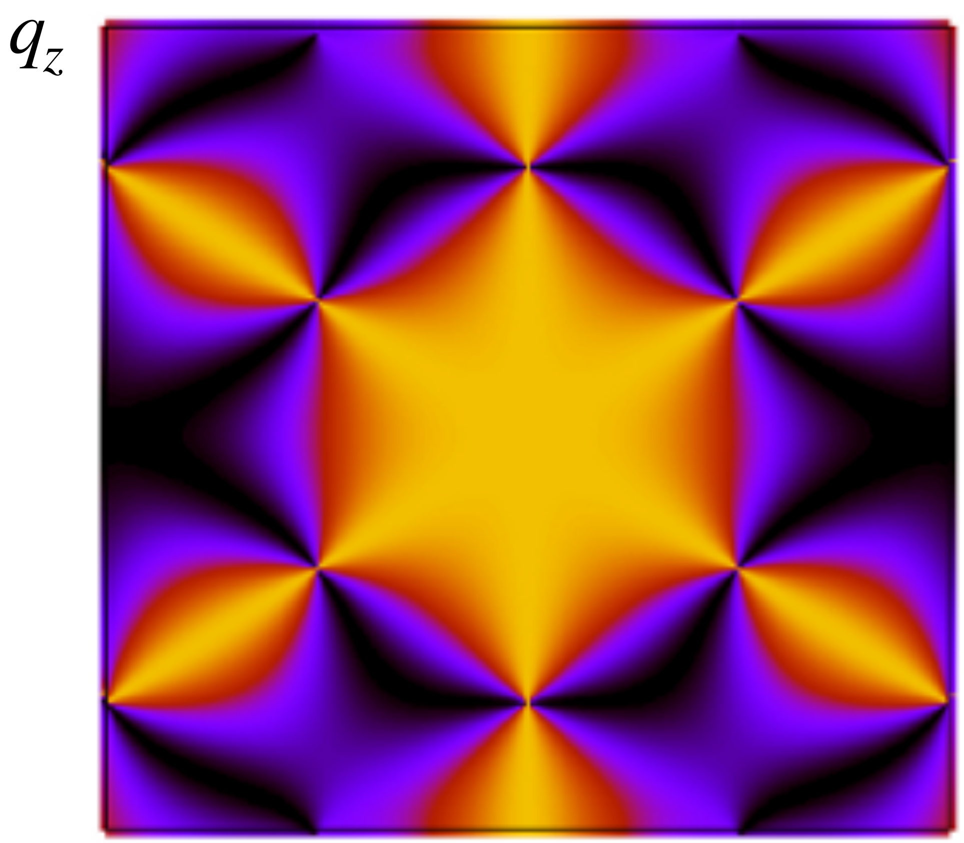
Spin correlation with power-law decay:
Disordered but strongly correlated

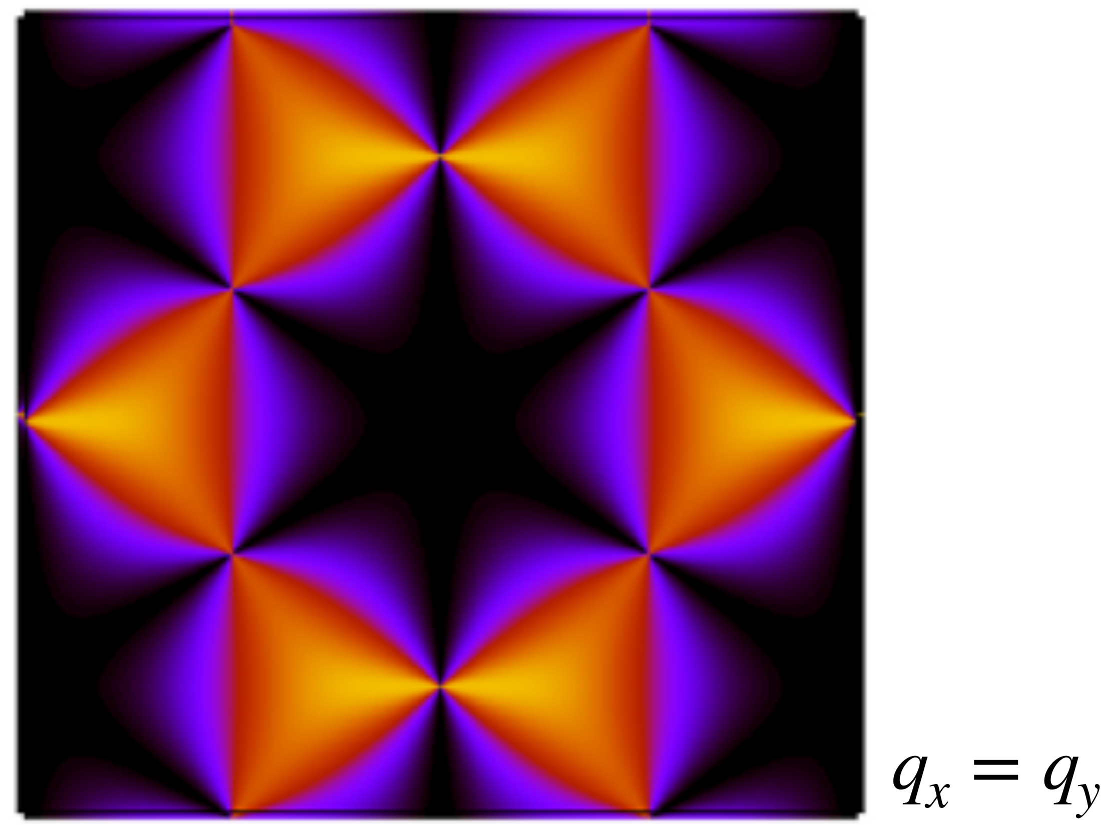
Canals and Lacroix 1998
Isakov et. al. 2004
Henley 2005

Coulomb phase

Ising $\sum_{i\in\boxtimes}\sigma_i = 0$

Heisenberg $\sum_{i\in\boxtimes}\mathbf{s}_i = 0$





Spin correlation with power-law decay:

Disordered but strongly correlated

Canals and Lacroix 1998
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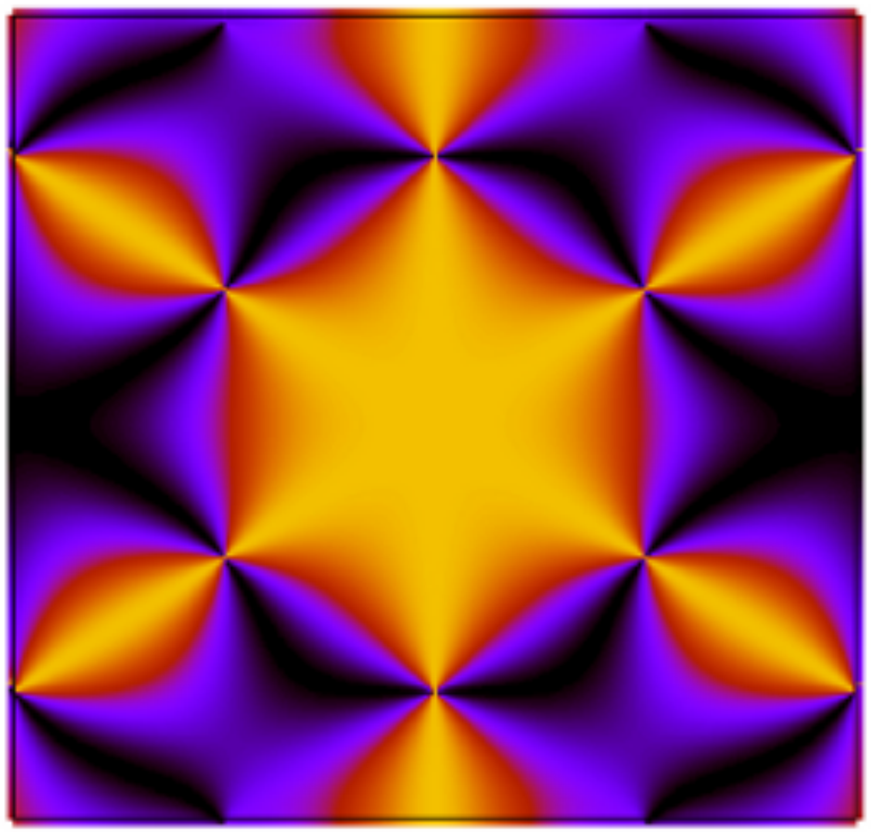
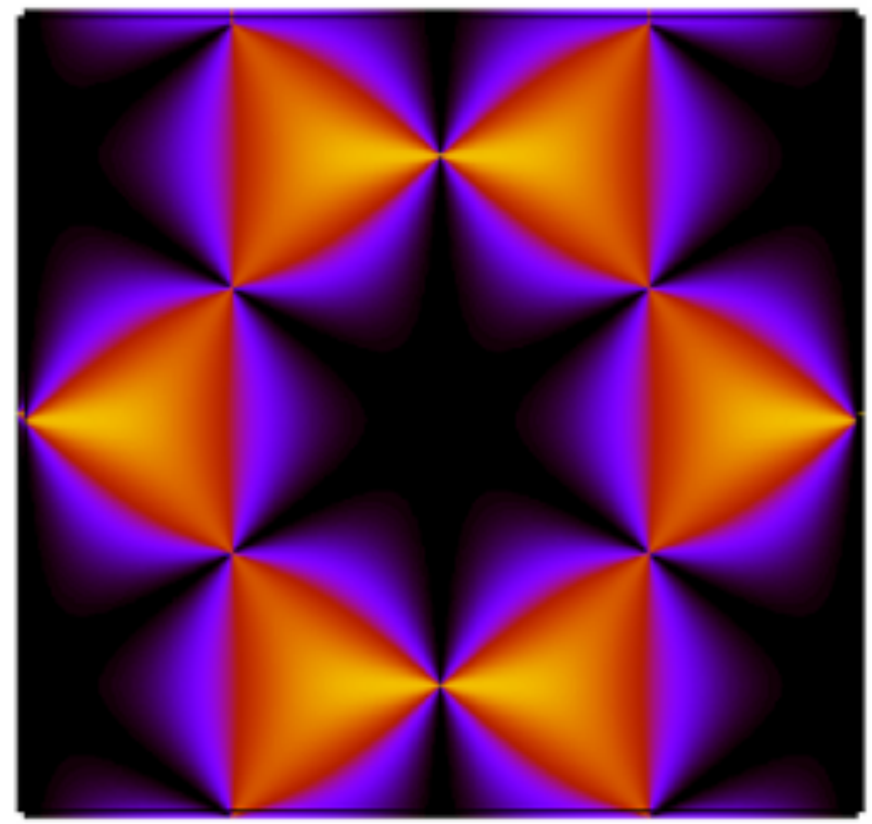
Coulomb phase

Ising $\sum_{i \in \boxtimes} \sigma_i = 0$ Heisenberg $\sum_{i \in \boxtimes} \mathbf{s}_i = 0$

q_z





$q_x = q_y$

Spin correlation with power-law decay:
Disordered but strongly correlated

Measured by neutron scattering

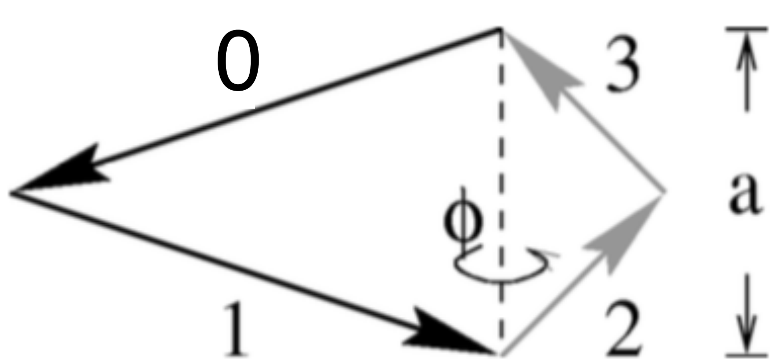
Canals and Lacroix 1998
Isakov et. al. 2004
Henley 2005

A classical spin liquid

Heisenberg pyrochlore antiferromagnet

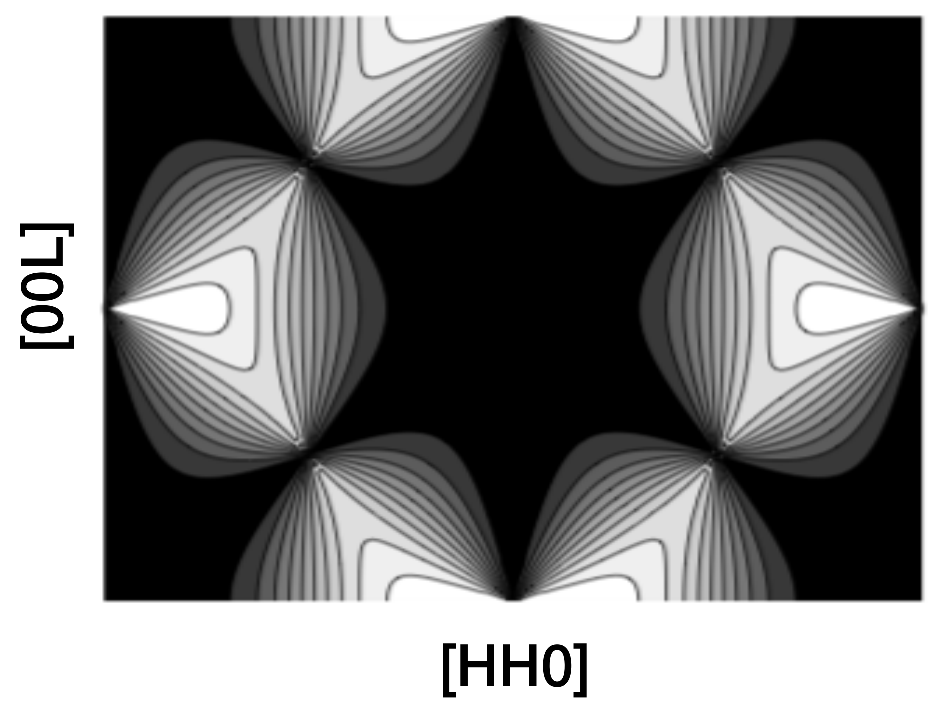
$$H = J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j, \quad J > 0$$

Macroscopic degeneracy

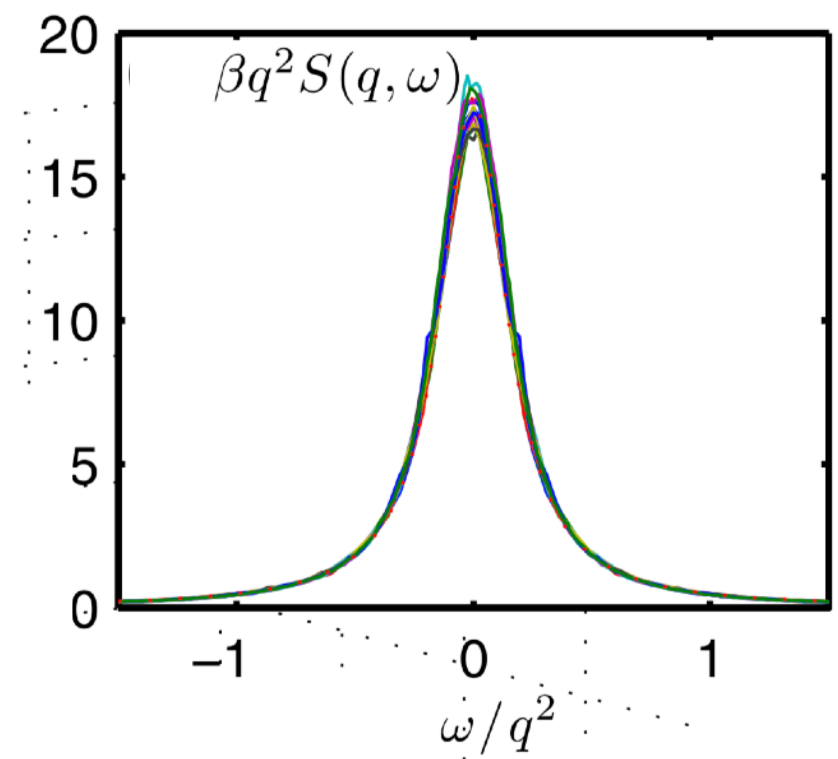


$$\sum_{i \in \boxtimes} \mathbf{S}_i = 0$$

Coulomb correlation



Spin diffusion



Moessner Chalker 1999
Isakov Greger Moessner Sondhi 2004
Conlon Chalker 2009

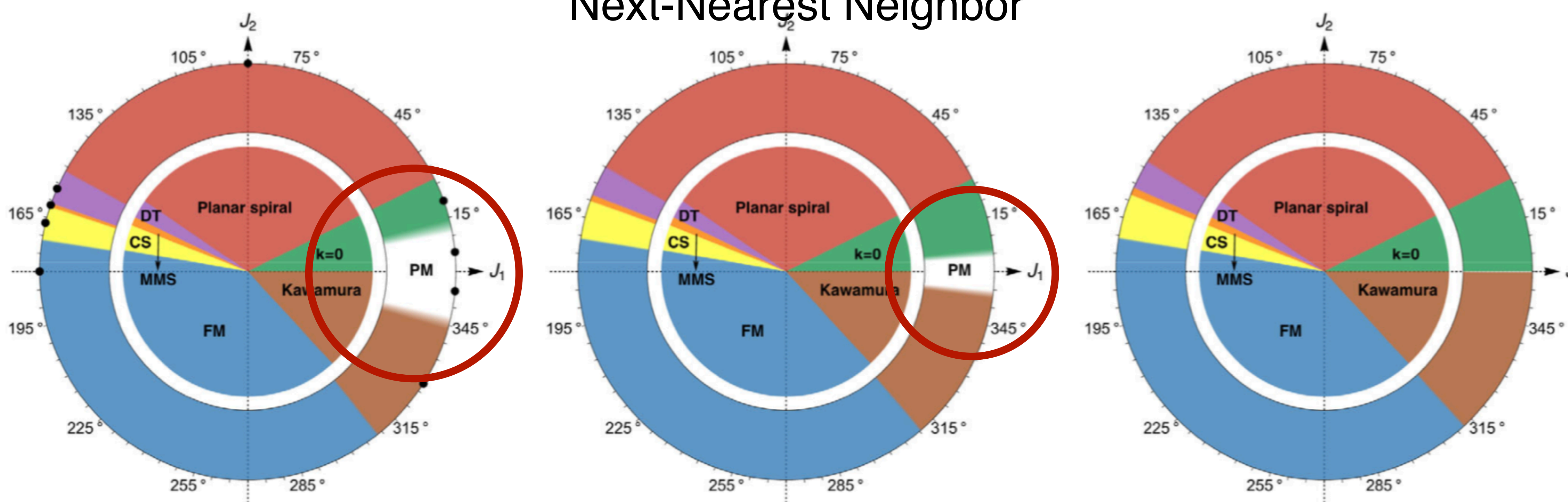
A quantum spin liquid?

Heisenberg pyrochlore antiferromagnet (quantum version)

$$H = J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j, \quad J > 0$$

S = 1/2 S = 1 S = 3/2

Next-Nearest Neighbor

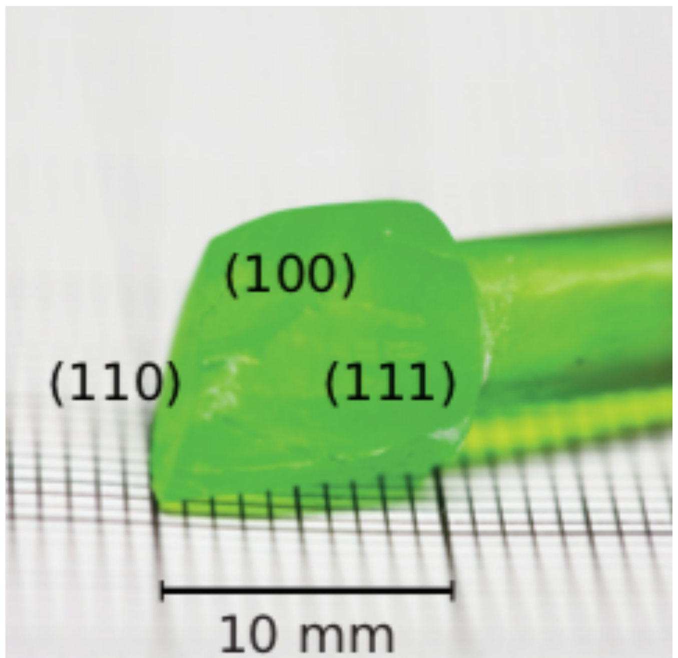


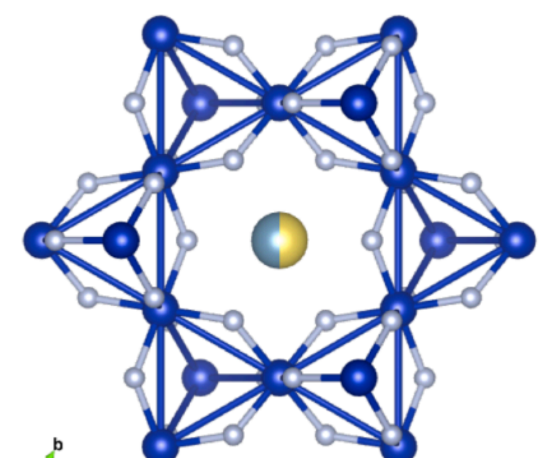
Nearest Neighbor

Iqbal et.al. 2019
PFRG

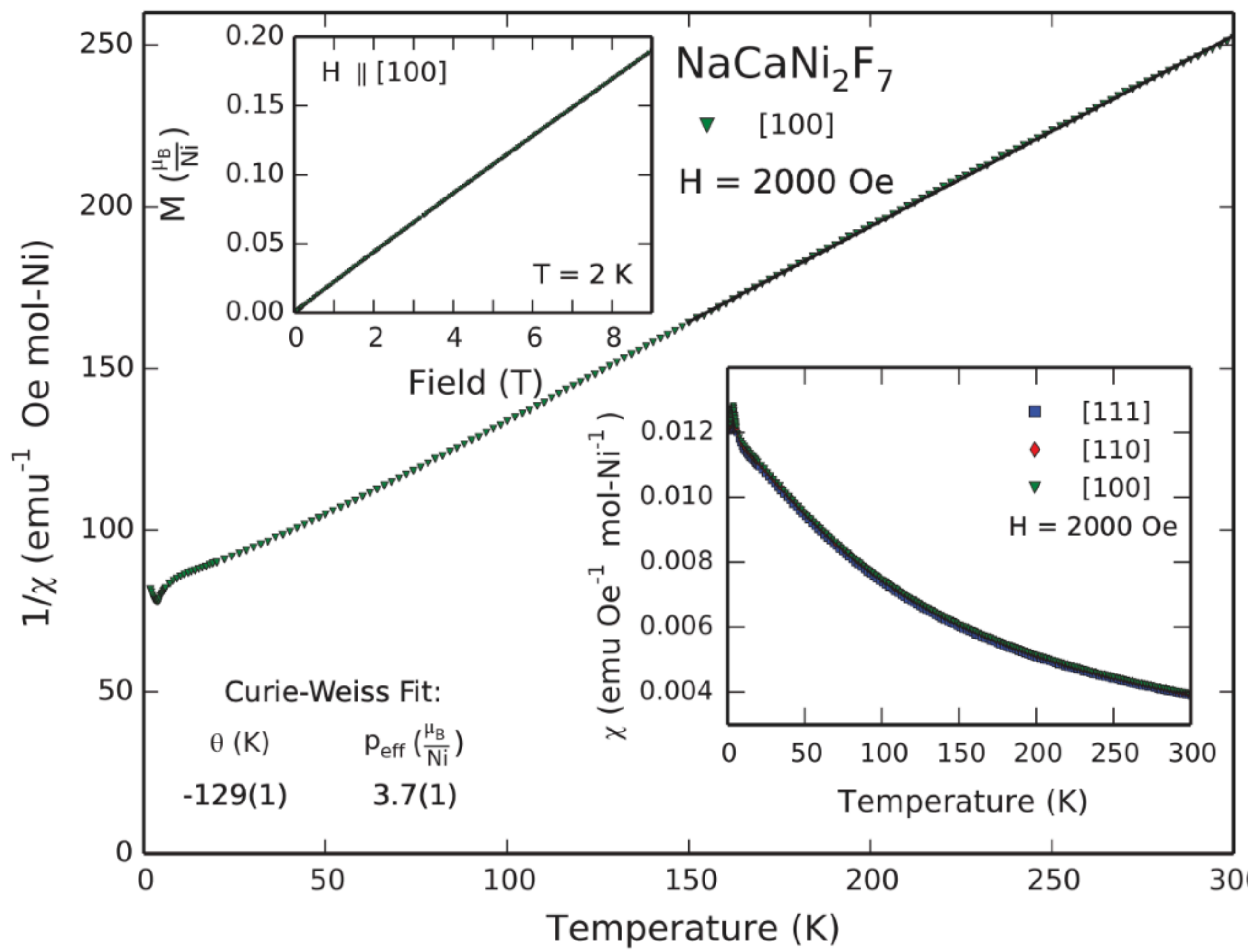
Quantum spin liquid candidate $NaCaNi_2F_7$

- Ni^{2+} on one pyrochlore lattice and Na^{1+}/Ca^{2+} on another





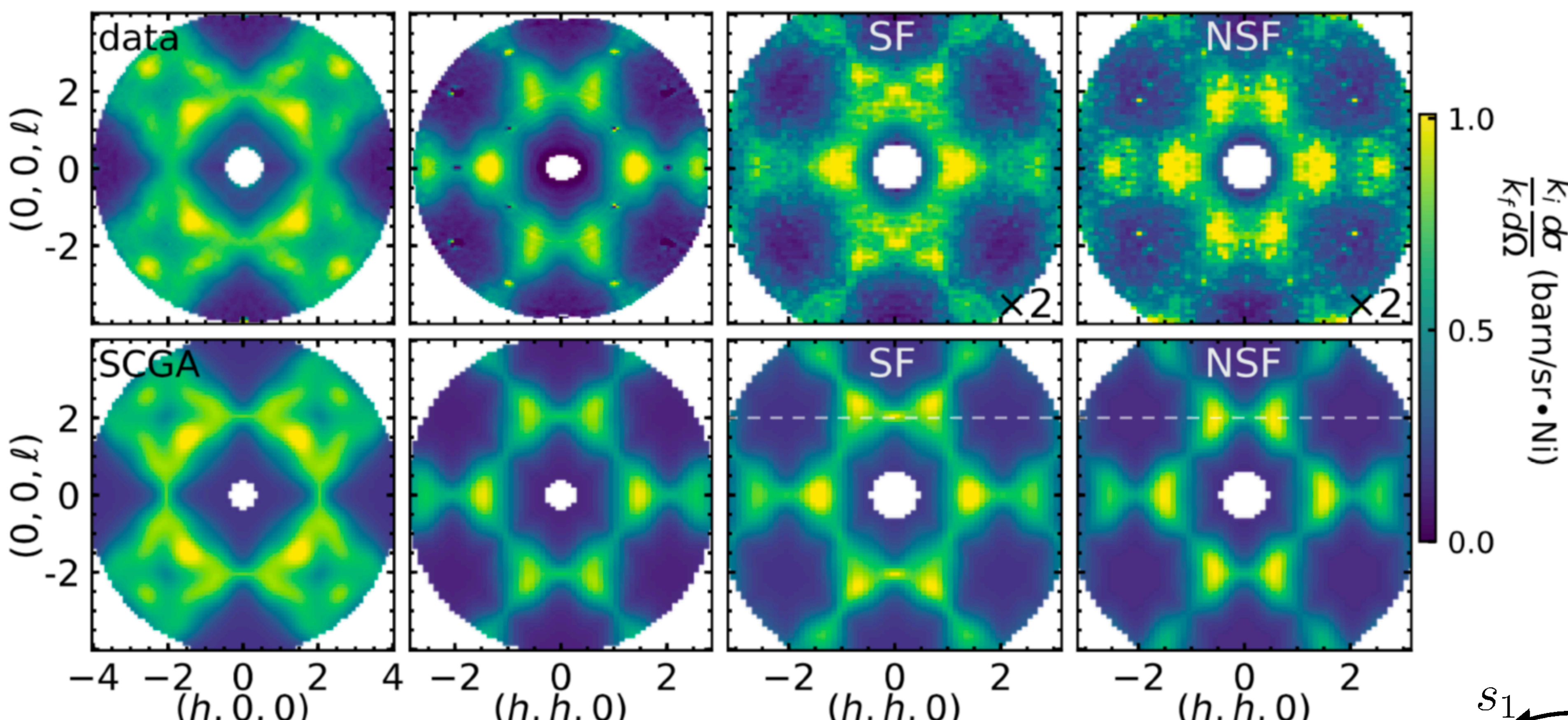
Kate Ross

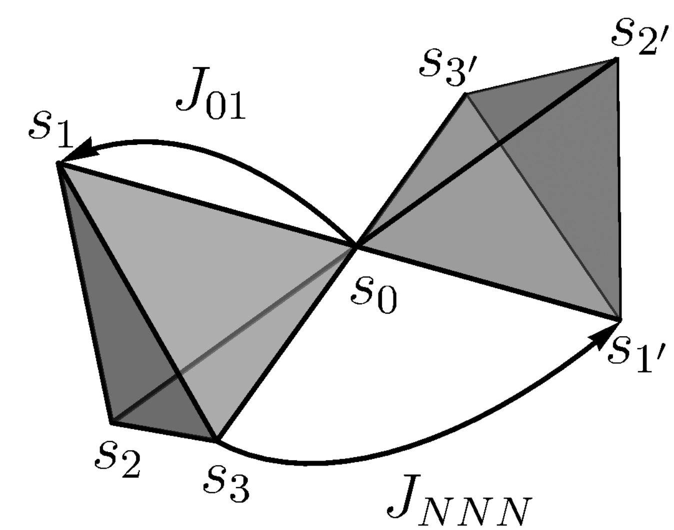


Krizan and Cava 2015

Quantum spin liquid candidate $NaCaNi_2F_7$

- Ni^{2+} on one pyrochlore lattice and Na^+/Ca^{2+} on another
- Heisenberg interaction





$$H = \frac{1}{2}\sum_{ij}\sum_{\mu\nu} J_{ij}^{\mu\nu} s_i^\mu s_j^\nu \quad J_{01} = \begin{pmatrix} J_2 & J_4 & J_4 \\ -J_4 & J_1 & J_3 \\ -J_4 & J_3 & J_1 \end{pmatrix}$$

$$J_1 = J_2 = 3.2,\ J_3 = 0.019,\ J_4 = -0.07,\ J_{NNN} = -0.025(\text{meV})$$

Plumb et. al. 2019

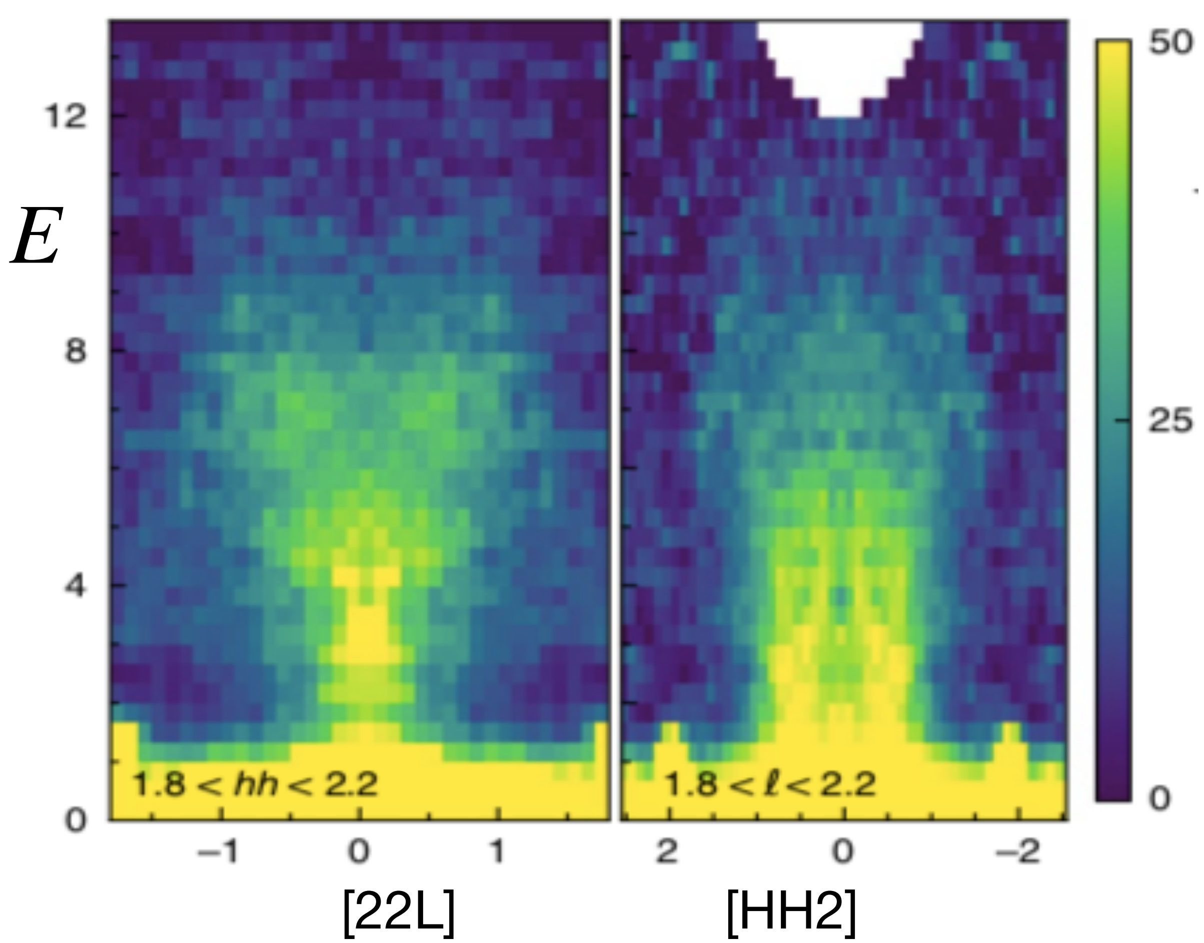
Quantum spin liquid candidate $NaCaNi_2F_7$

- Ni^{2+} on one pyrochlore lattice and Na^{+}/Ca^{2+} on another
- Heisenberg interaction
- Spin-1 system

Plumb et. al. 2019

Quantum spin liquid candidate $NaCaNi_2F_7$

- Ni^{2+} on one pyrochlore lattice and Na^+/Ca^{2+} on another
- Heisenberg interaction
- Spin-1 system
- No magnetic ordering down to 0.35K $J_1 = J_2 = 3.2\,(\mathrm{meV})$
- ~90% spectral weight from inelastic scattering at 1.8K



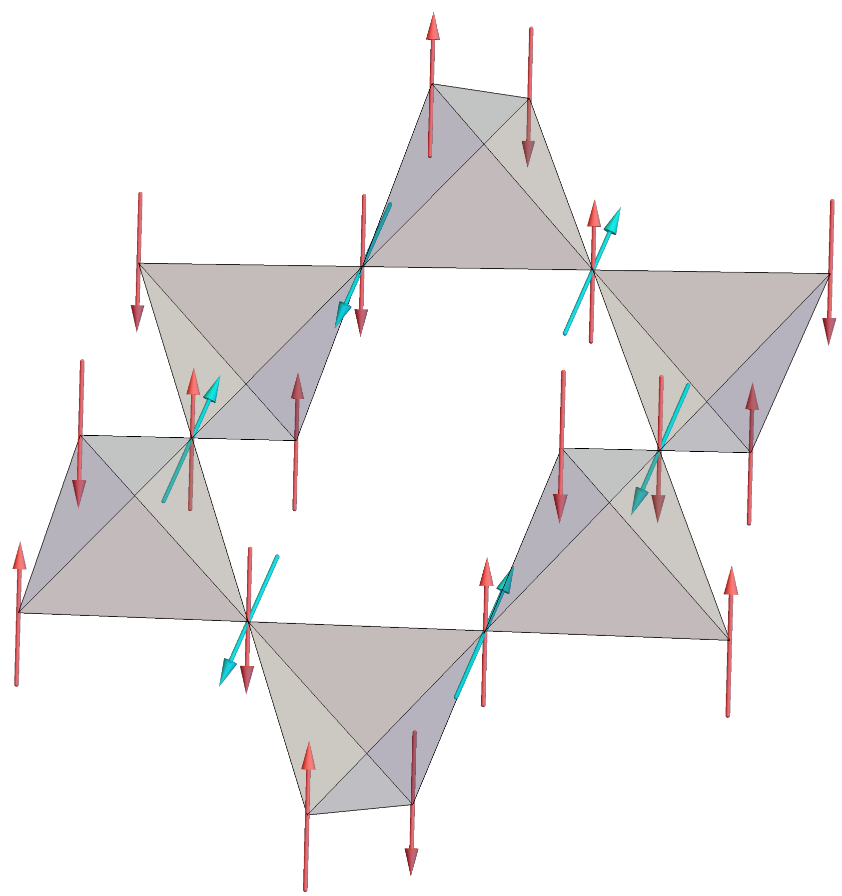
Classical expectation:
S/S(S+1) = 50%

$$\mathcal{S}(\mathbf{q},\omega) \sim \frac{1}{2\pi N}\int dt e^{-i\omega t}\sum_{i,j} e^{-i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)}\langle s_i^{\mu}(0)s_j^{\nu}(t)\rangle$$

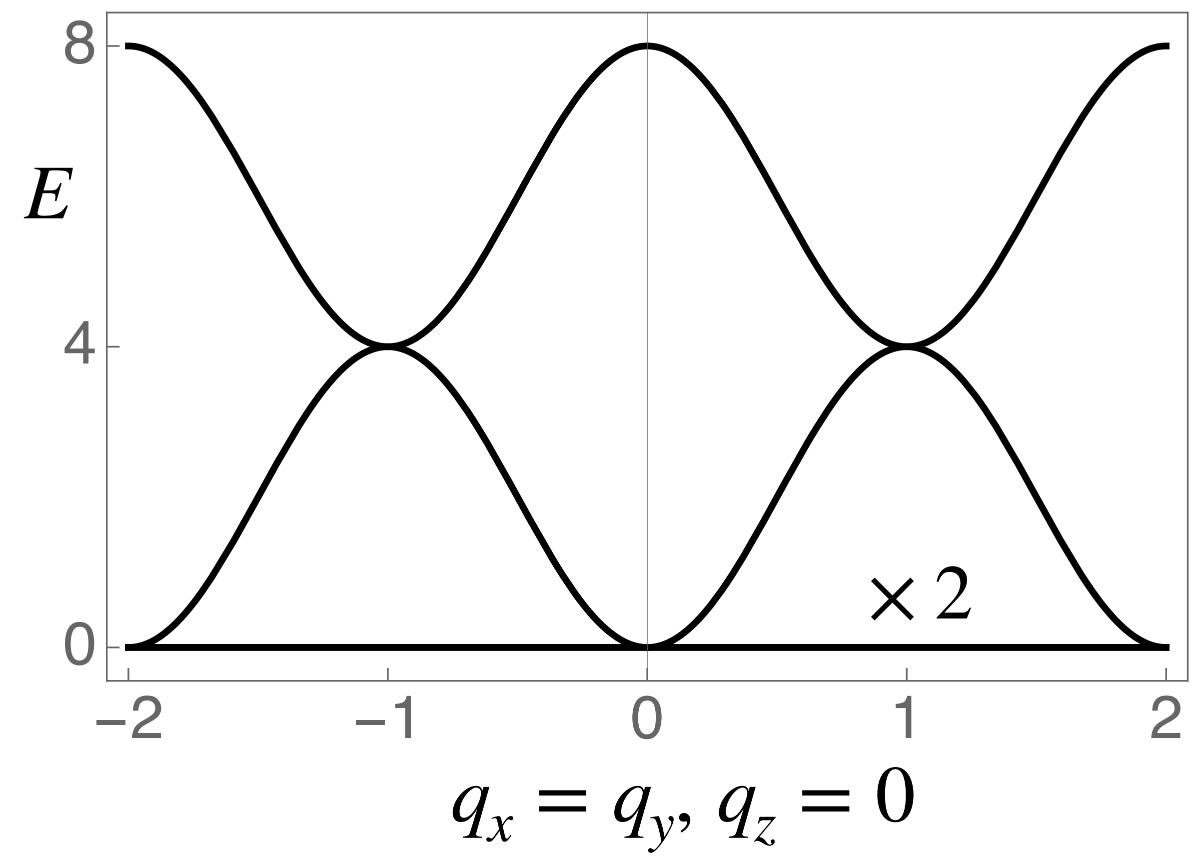
Plumb et. al. 2019

An example of spin dynamics

Ideal Heisenberg model
Collinear ground state



Single tetrahedron



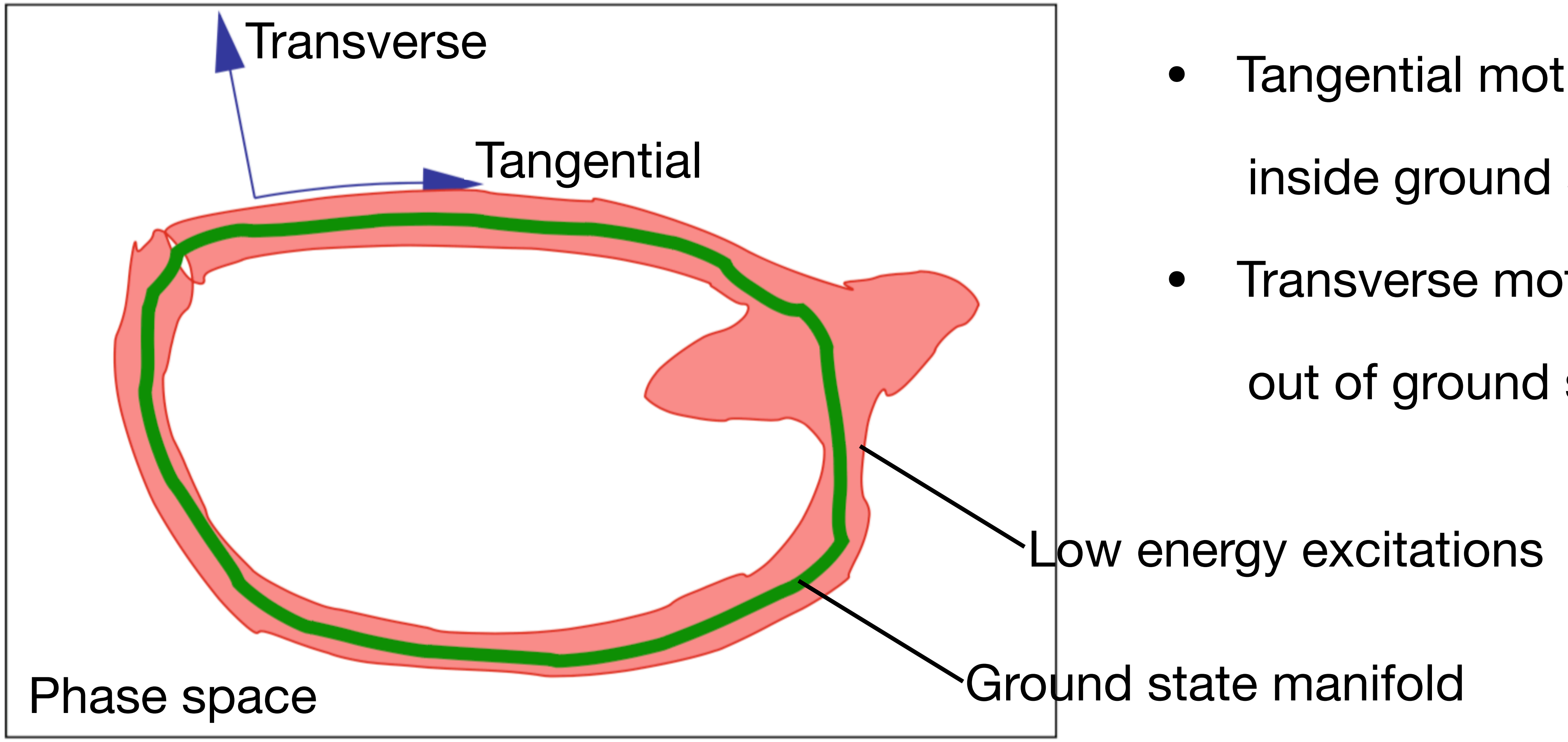
Spin wave

Canals and Lacroix 1998
Tchernyshyov, Moessner, and Sondhi 2002

Spin dynamics of $NaCaNi_2F_7$

Zhang et. al. 2019

- Ground states: many degenerate/nearly-degenerate disordered states
- Fluctuations: finite-frequency spin waves
- Dynamical picture: spin waves drive the motion between ground states



- Tangential motion:

  inside ground state manifold

- Transverse motion:

  out of ground state manifold

Dynamical models

- Stochastic Large-n model
  - Tangential motion: slow
  - Transverse motion: fast
- Brownian motion $$\frac{d\mathbf{v}}{dt} = \frac{1}{m}\mathbf{F}(t) + \boldsymbol{\xi}(t)$$

White noise

Garanin and Canals 1999
Conlon and Chalker 2009

Dynamical models

- Stochastic Large-n model
    - Tangential motion: slow
    - Transverse motion: fast
- Brownian motion $$\frac{d\mathbf{v}}{dt} = \frac{1}{m}\mathbf{F}(t) + \boldsymbol{\xi}(t)$$
- To conserve spins (angular momentum)

$$\frac{d\mathbf{s}}{dt} = -\gamma\nabla \cdot \mathbf{j}(t) + \boldsymbol{\xi}(t)$$

$$\mathbf{j}(t) = -\nabla\frac{\partial E}{\partial \mathbf{s}}$$

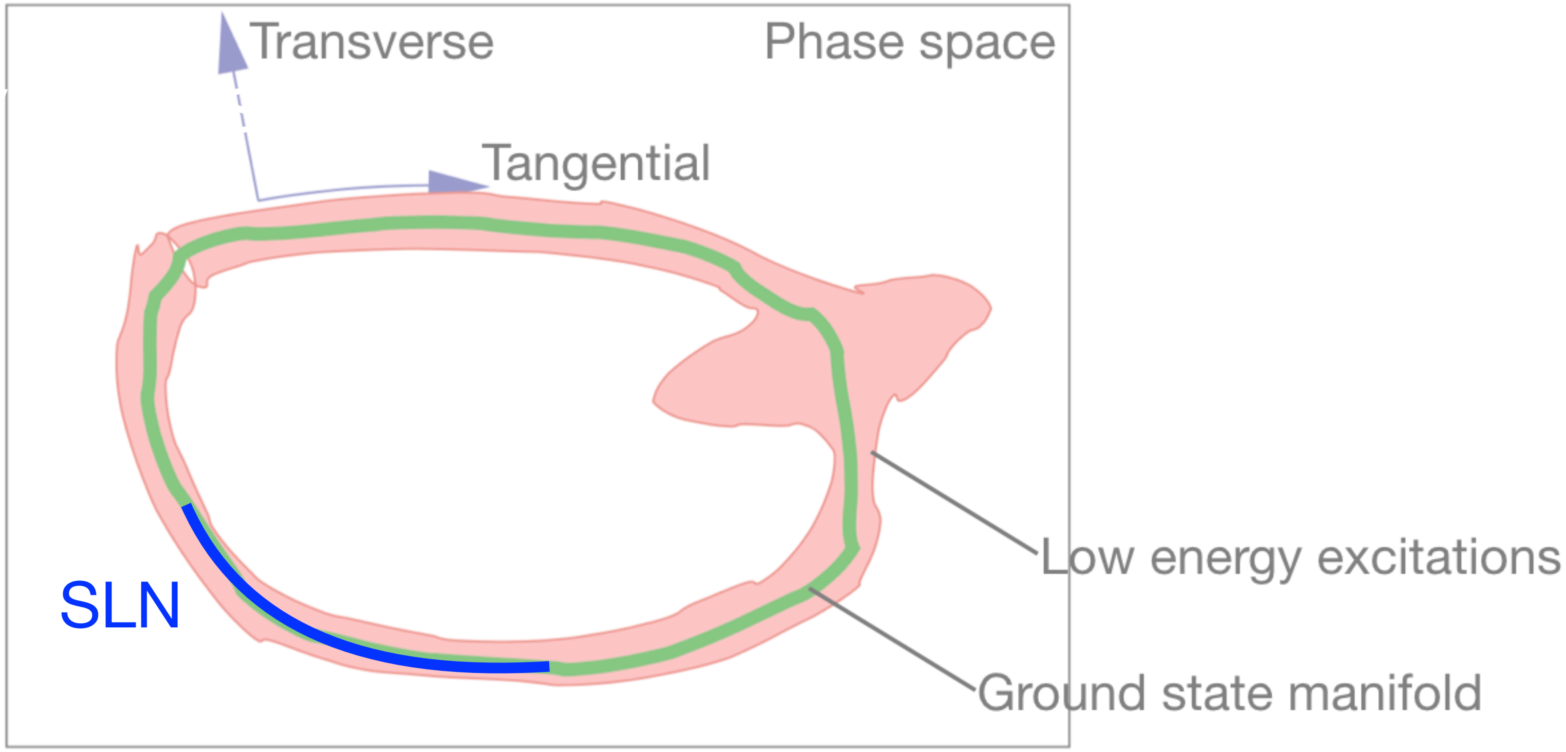
- Discrete lattice

Generalized force

Garanin and Canals 1999
Conlon and Chalker 2009

Dynamical models

- Stochastic Large-n model
- Long time scale
- Relaxation



Dynamical models

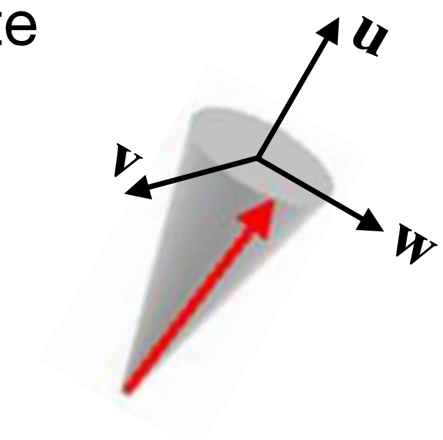
- Stochastic Large-n model
  - Long time scale
  - Relaxation

- Linear spin wave theory

$$\hbar \frac{d}{dt}\mathbf{s}_i = -\mathbf{s}_i \times \mathbf{H}_{\text{eff}}$$

Linearized for small deviations from a ground state

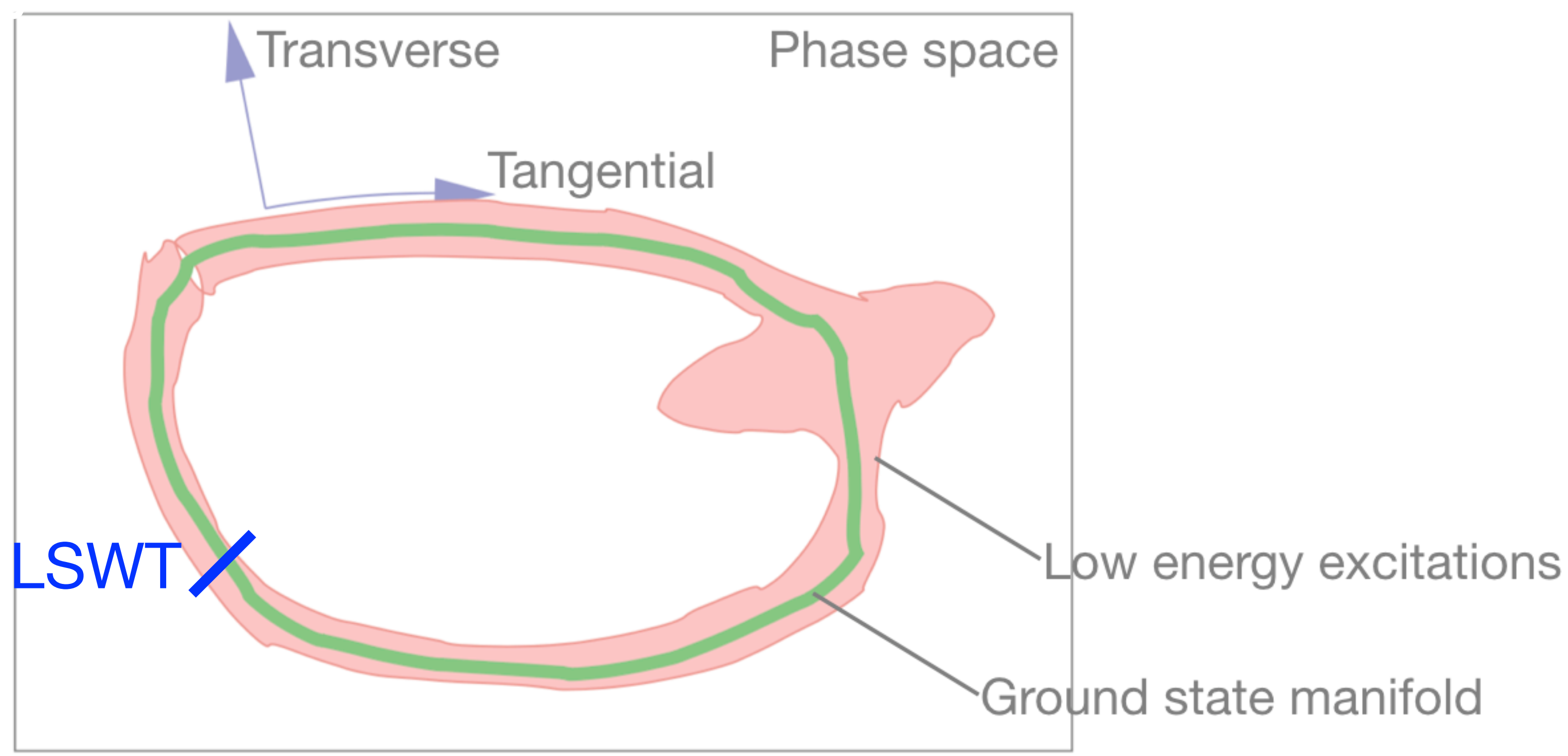
$$\begin{aligned}\mathbf{s}_i &= \sqrt{S^2 - S(x_i^2 + y_i^2)}\,\mathbf{u}_i + \sqrt{S}(x_i\mathbf{v}_i + y_i\mathbf{w}_i) \\ &\approx \left(S - \frac{x_i^2 + y_i^2}{2}\right)\mathbf{u}_i + \sqrt{S}(x_i\mathbf{v}_i + y_i\mathbf{w}_i).\end{aligned}$$



Walker and Walstedt 1980
Savary and Balents 2014

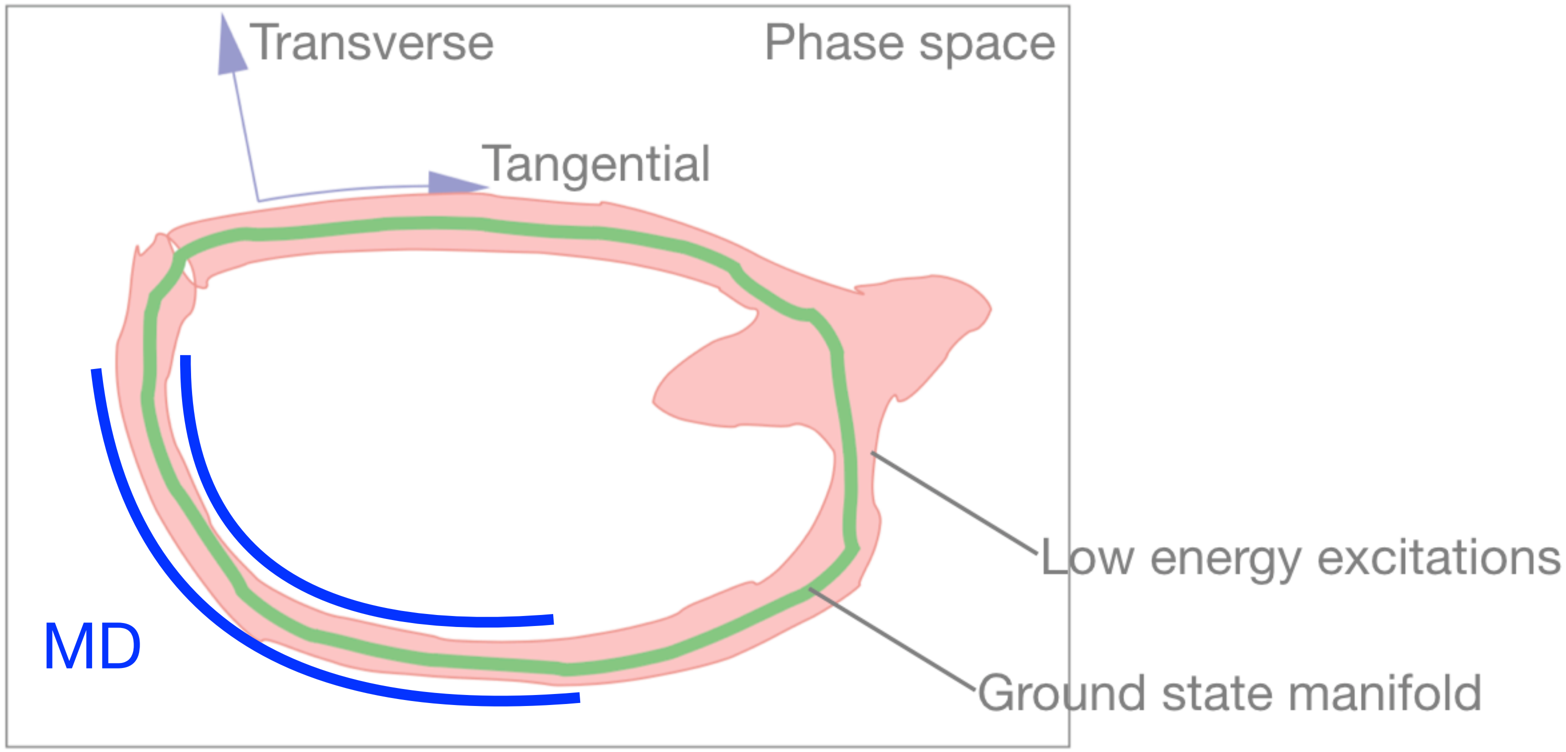
Dynamical models

- Stochastic Large-n model
  - Long time scale
  - Relaxation
- Linear spin wave theory
  - Finite-frequency excitations
  - Around one ground state



Dynamical models

- Stochastic Large-n model
  - Long time scale
  - Relaxation
- Linear spin wave theory
  - Finite-frequency excitations
  - Around one ground state
- Molecular dynamics
  - Full simulation
  - Short + long time scale



Dynamical models

- Stochastic Large-n model
  - Long time scale
  - Relaxation

- Linear spin wave theory

  Zero temperature
  - Finite-frequency excitations
  - Around one ground state

- Molecular dynamics

  Thermal excitation
  - Full simulation
  - Short + long time scale

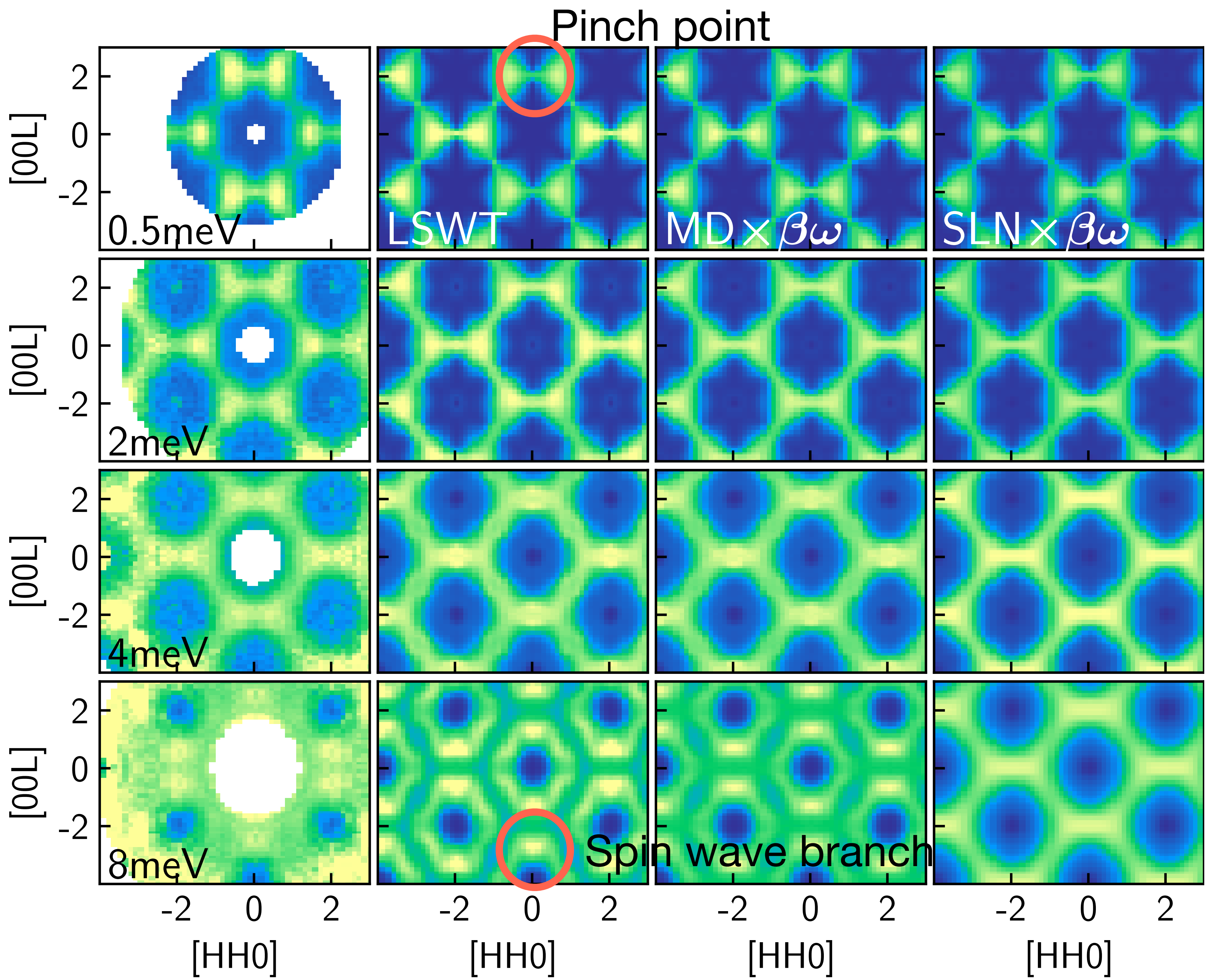
Considering quantum statistics of spin waves:

$$\beta\omega \mathcal{S}_{\rm MD}(\mathbf{q},\omega)\big|_{\rm finite\ T} = \mathcal{S}_{\rm LSWT}(\mathbf{q},\omega)\big|_{\rm T=0}$$

$$\text{for} \quad \beta\omega \gg 1$$

Zhang et. al. 2019

Theoretical modeling

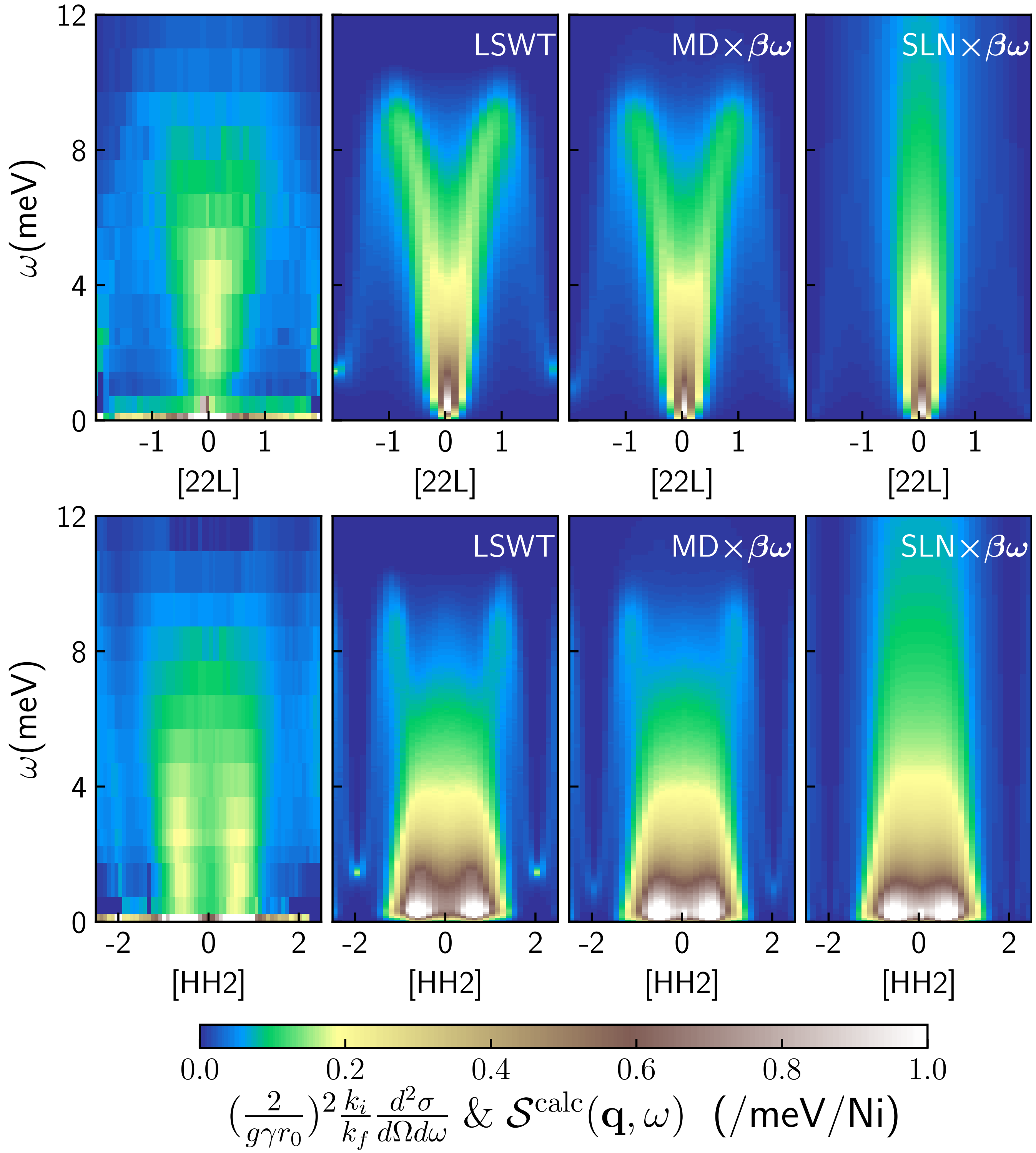


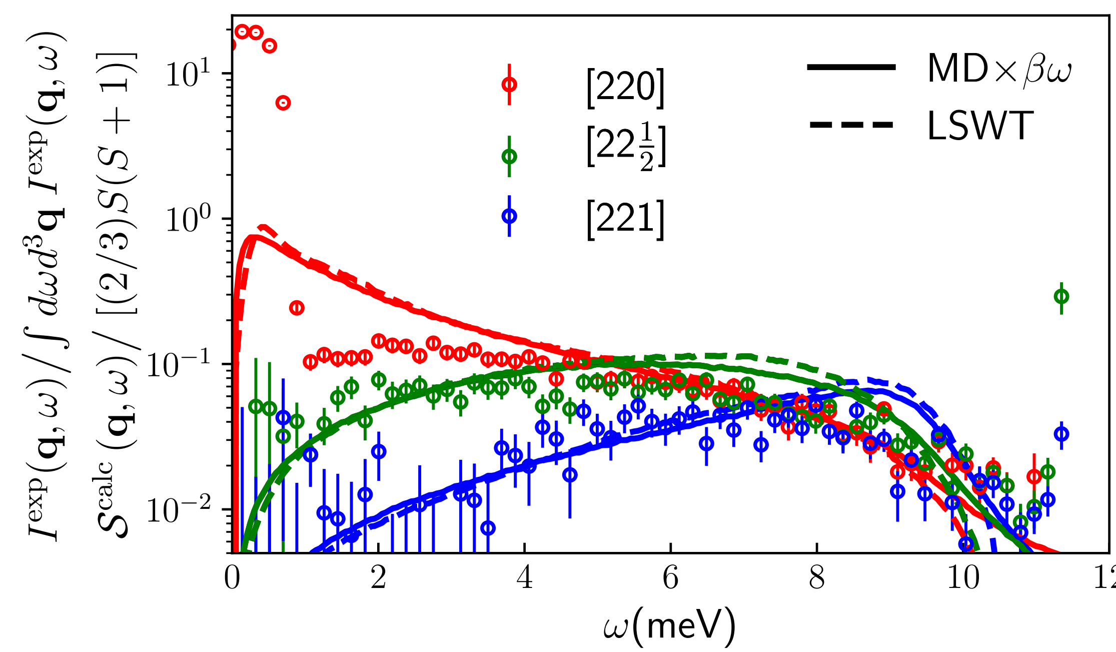
Agreement for all momenta

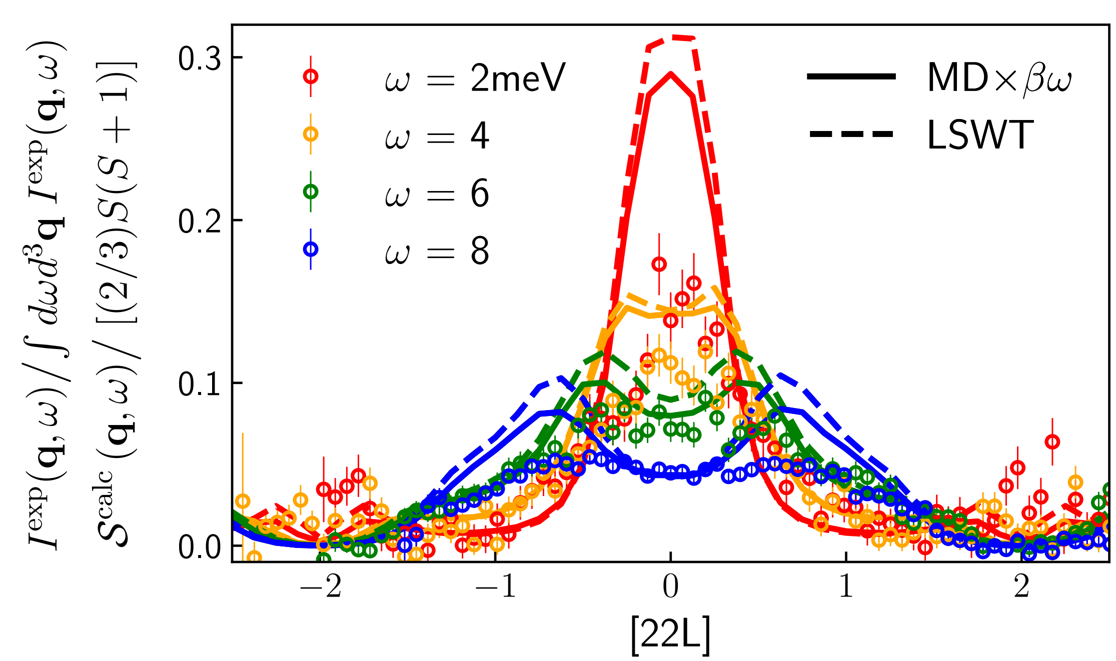
Zhang et. al. 2019
Yan, Pohle, and Shannon 2018

Theoretical modeling

Zhang et. al. 2019







Agreement between MD, LSWT and experimental data for $\gtrsim$ 2meV

Why linear spin wave theory

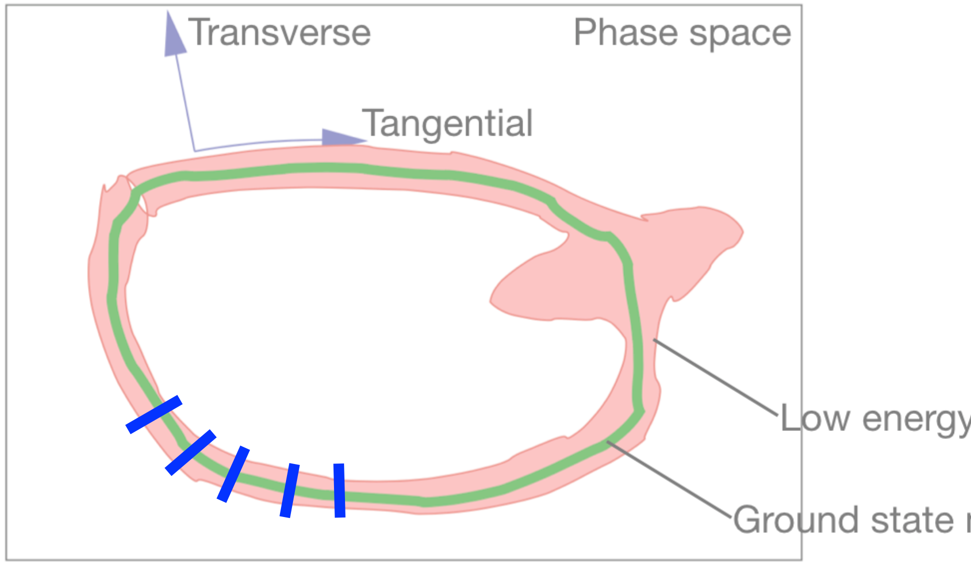
Zhang et. al. 2019

- Not considered
  - Lack of long-range order
  - Quantum renormalization
  - Thermal broadening
  - Multimagnon continuum

Why linear spin wave theory

Zhang et. al. 2019

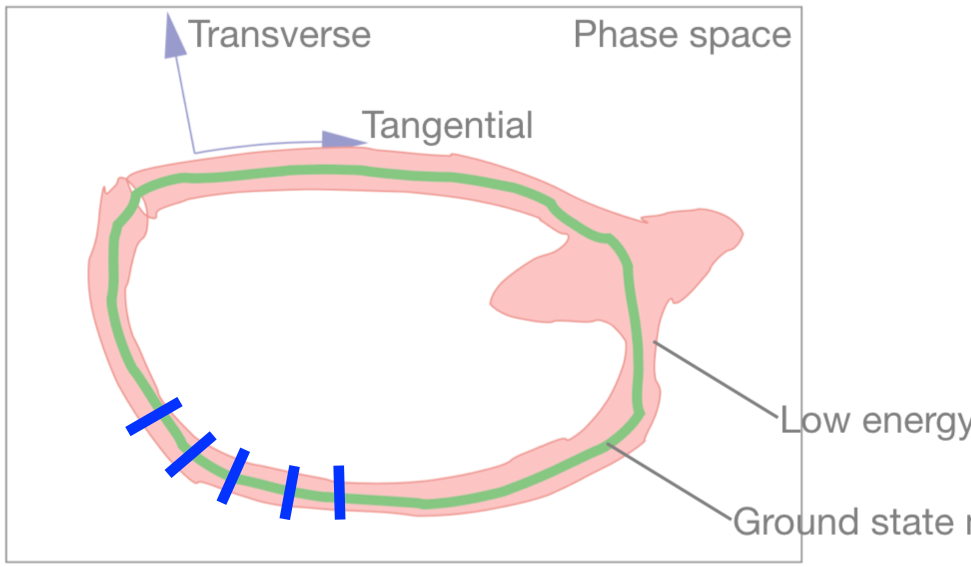
- Correct statistics
  - Fluctuations of disordered ground states
  - Averaged among similar spectra

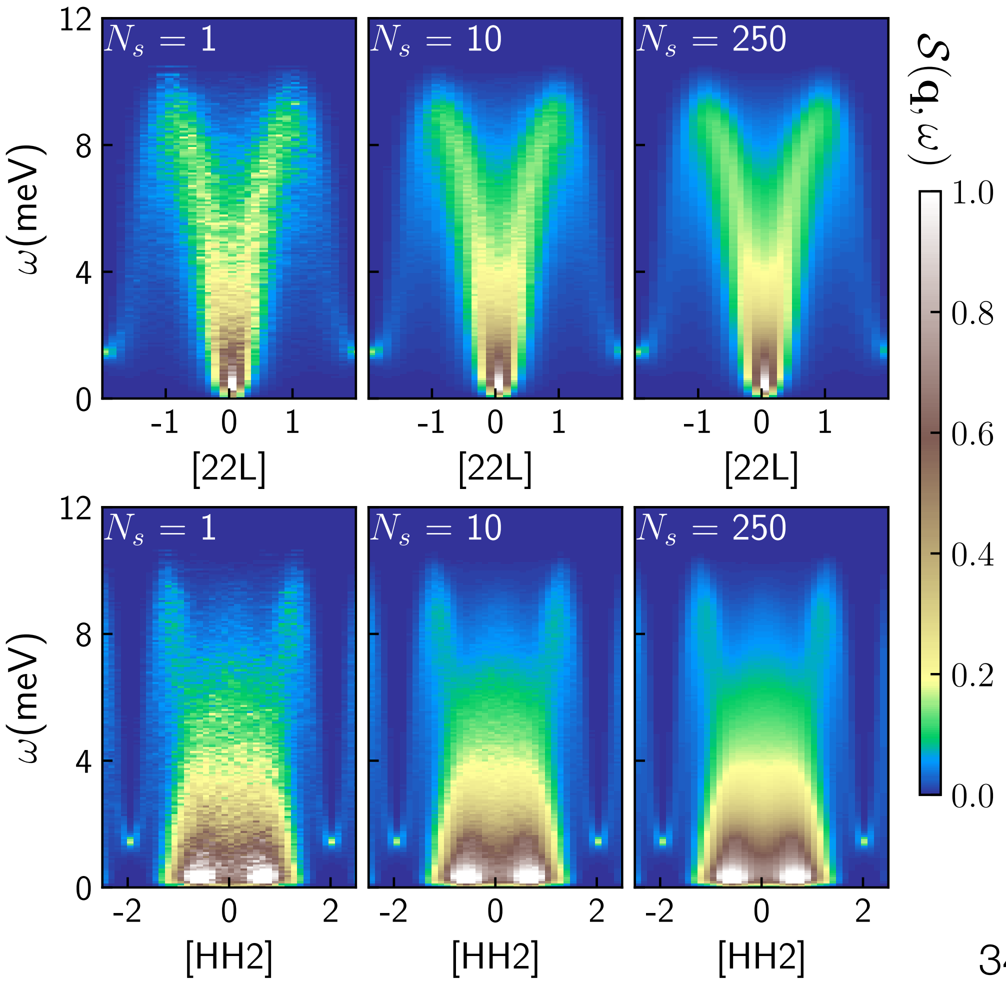


Why linear spin wave theory

Zhang et. al. 2019

- Correct statistics
  - Fluctuations of disordered ground states
  - Averaged among similar spectra
  - Without well-defined momenta

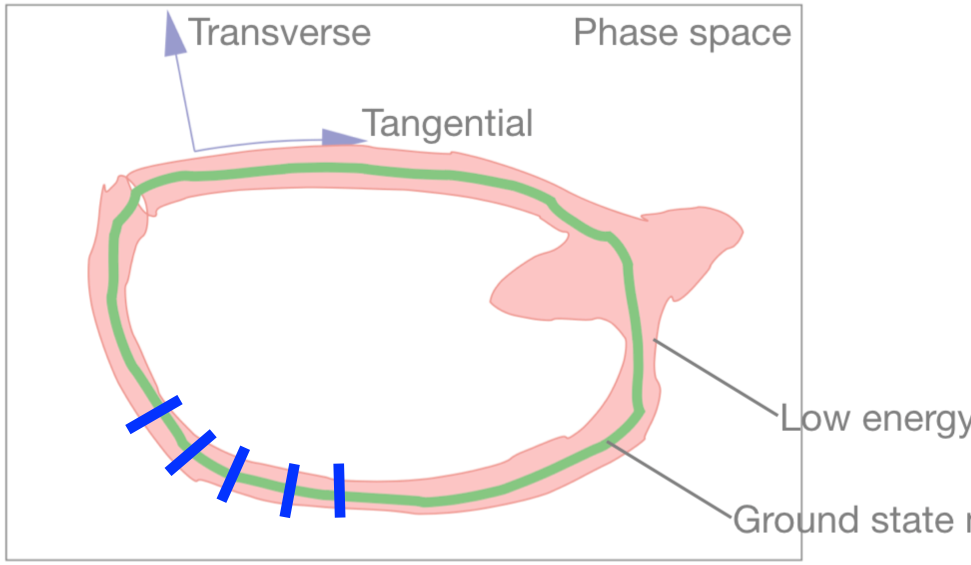


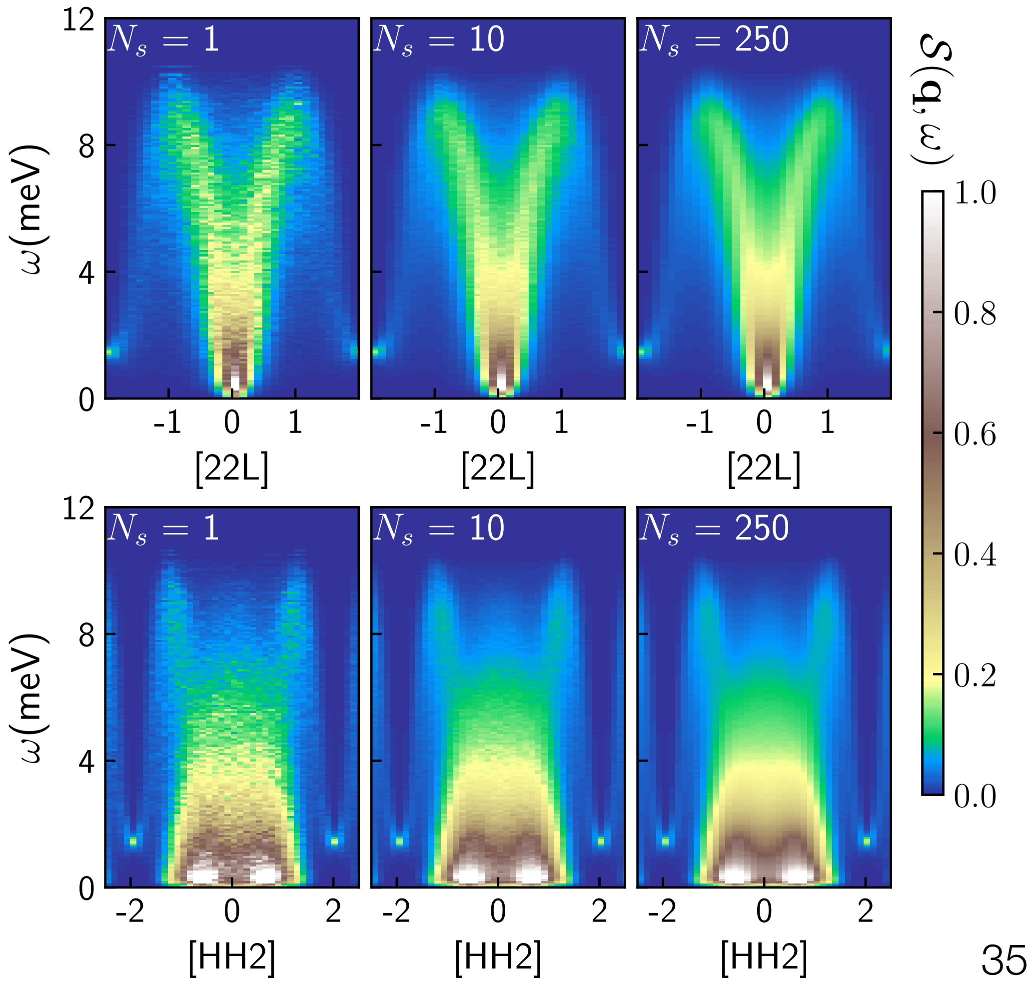


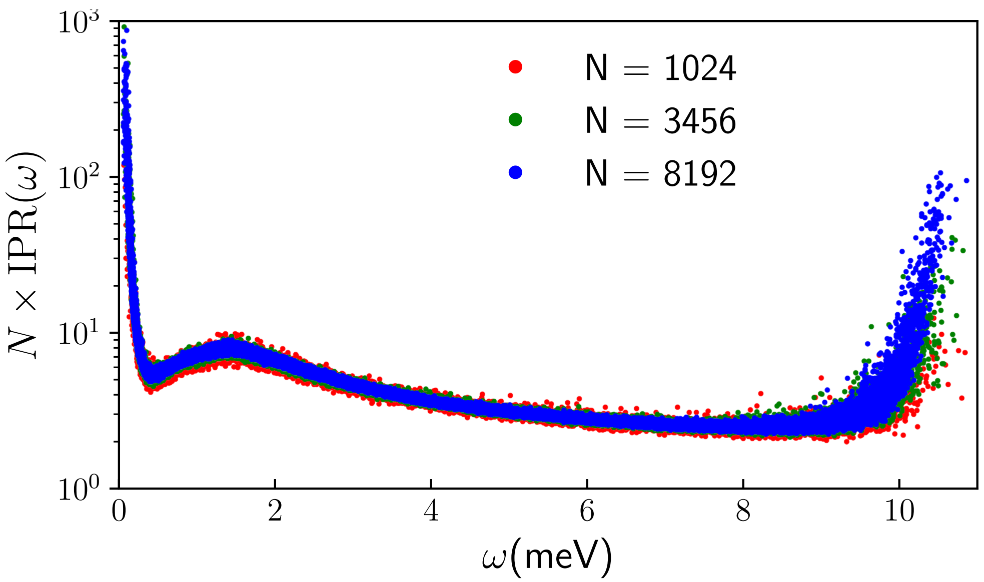
Why linear spin wave theory

Zhang et. al. 2019

- Correct statistics
  - Fluctuations of disordered ground states
  - Averaged among similar spectra
  - Without well-defined momenta
  - Delocalized



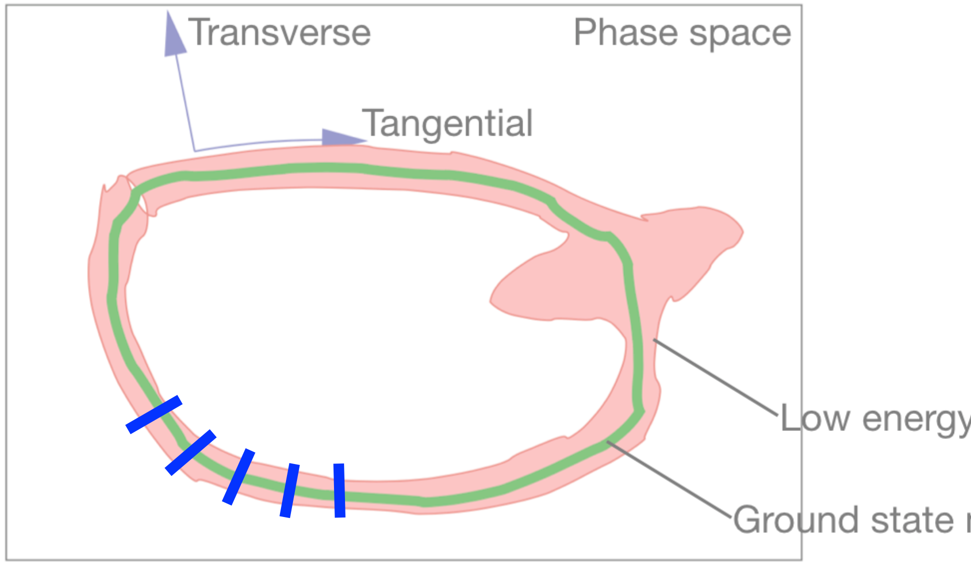


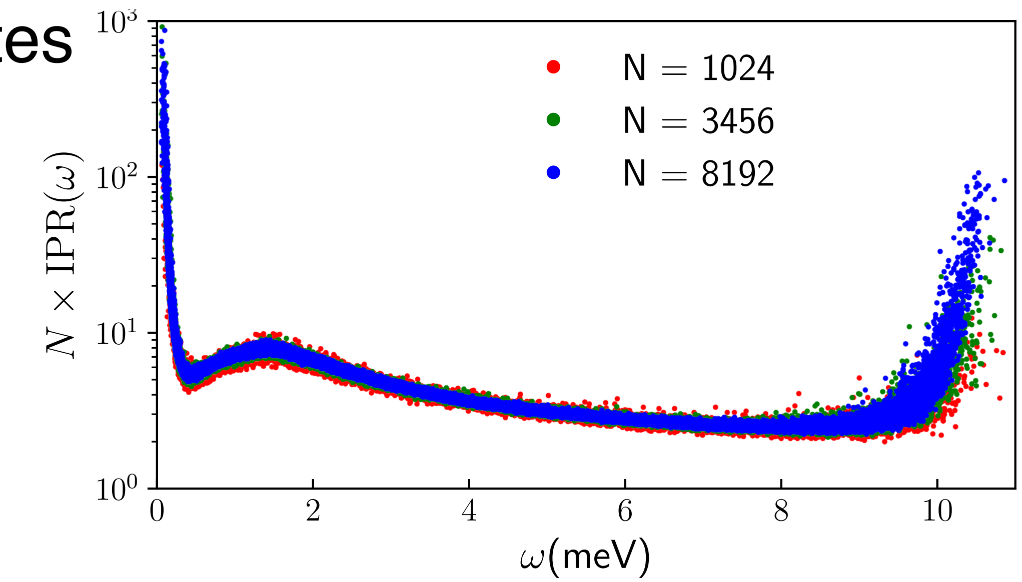


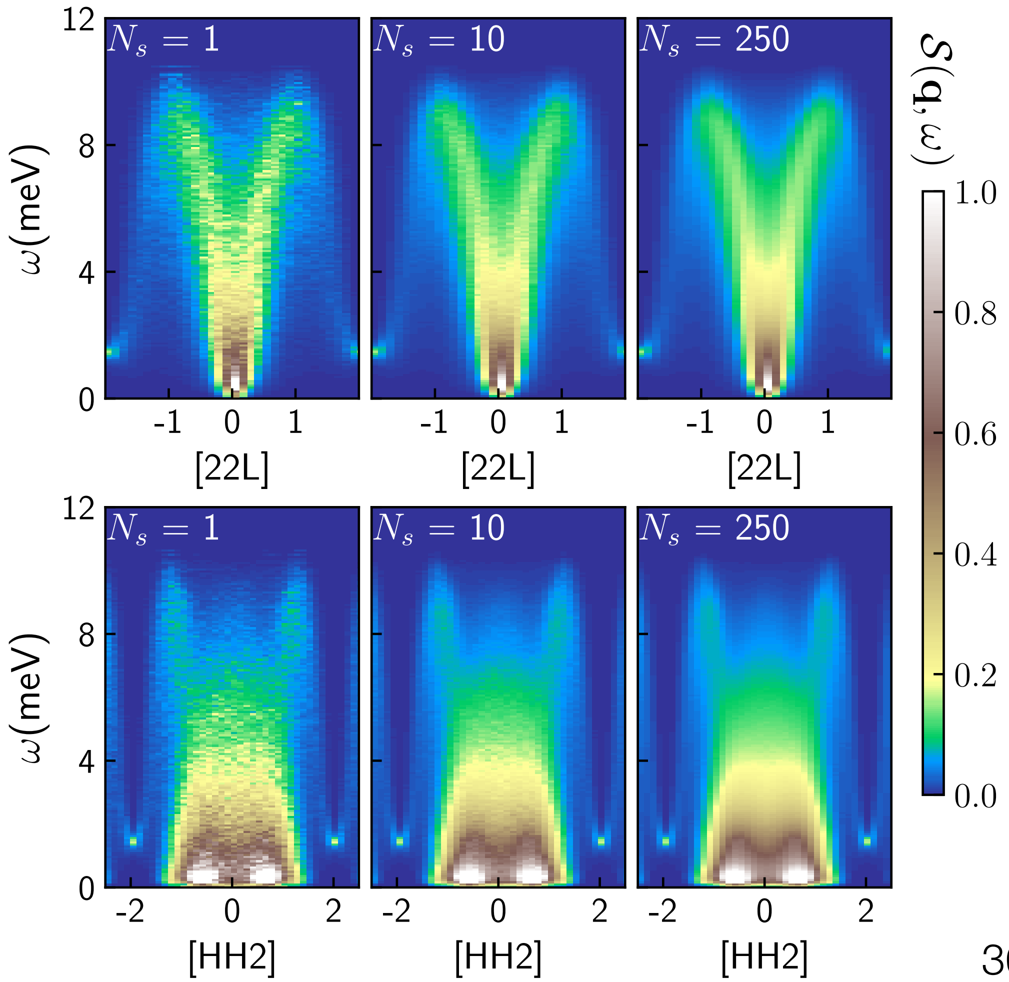
Why linear spin wave theory

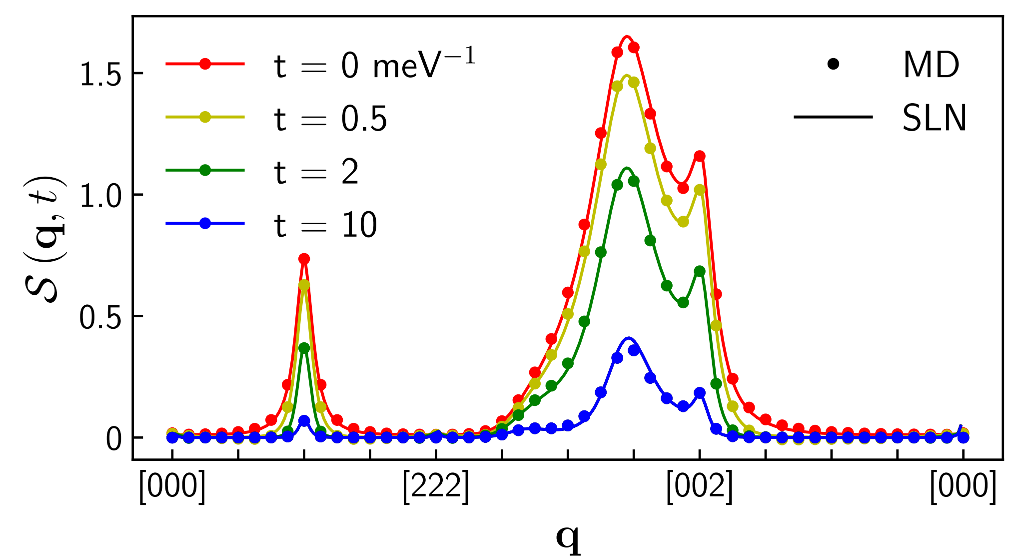
Zhang et. al. 2019

- Correct statistics
  - Fluctuations of disordered ground states
  - Averaged among similar spectra
  - Without well-defined momenta
  - Delocalized
  - Driving the motion between ground states





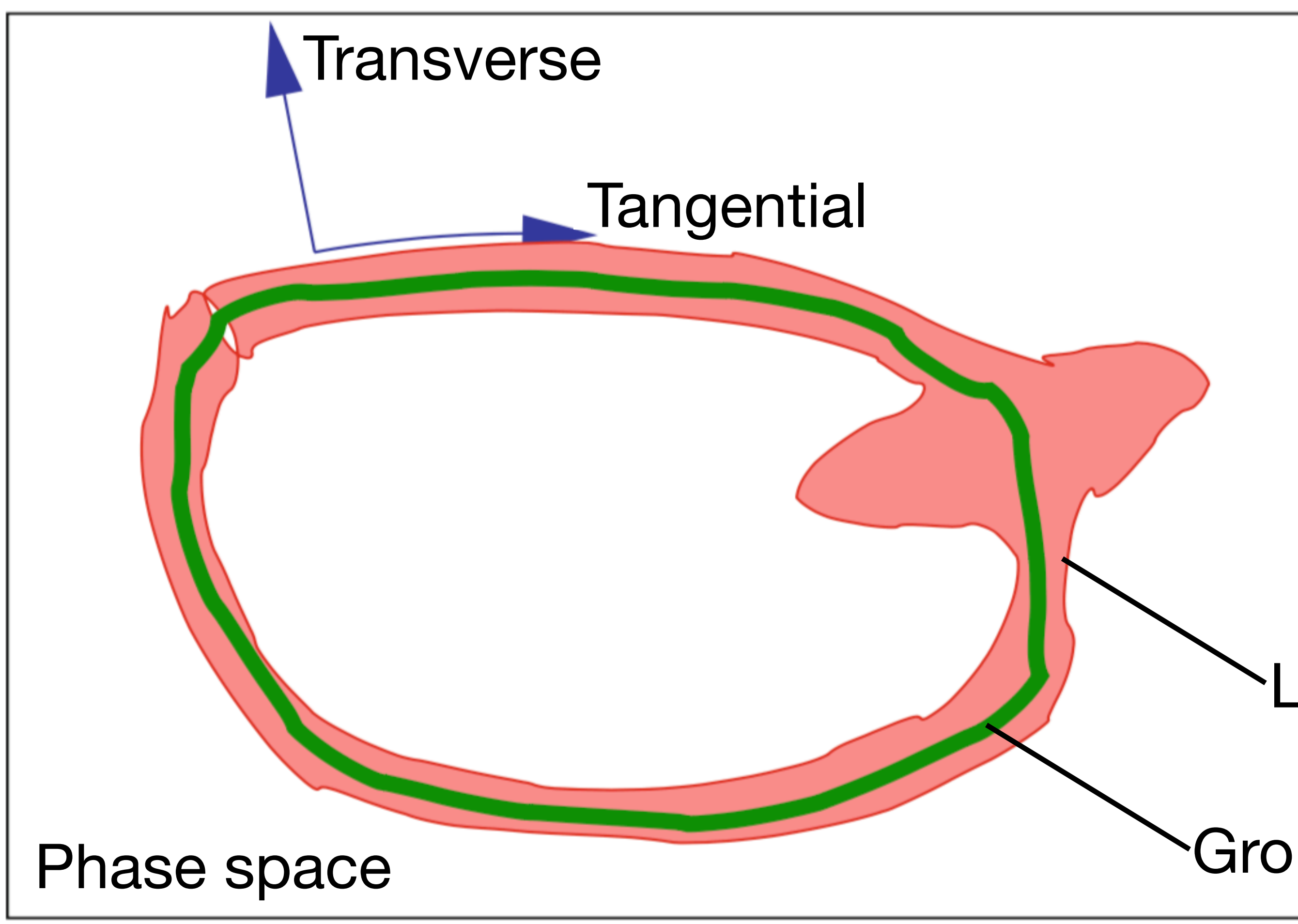




Spin dynamics

Zhang et. al. 2019

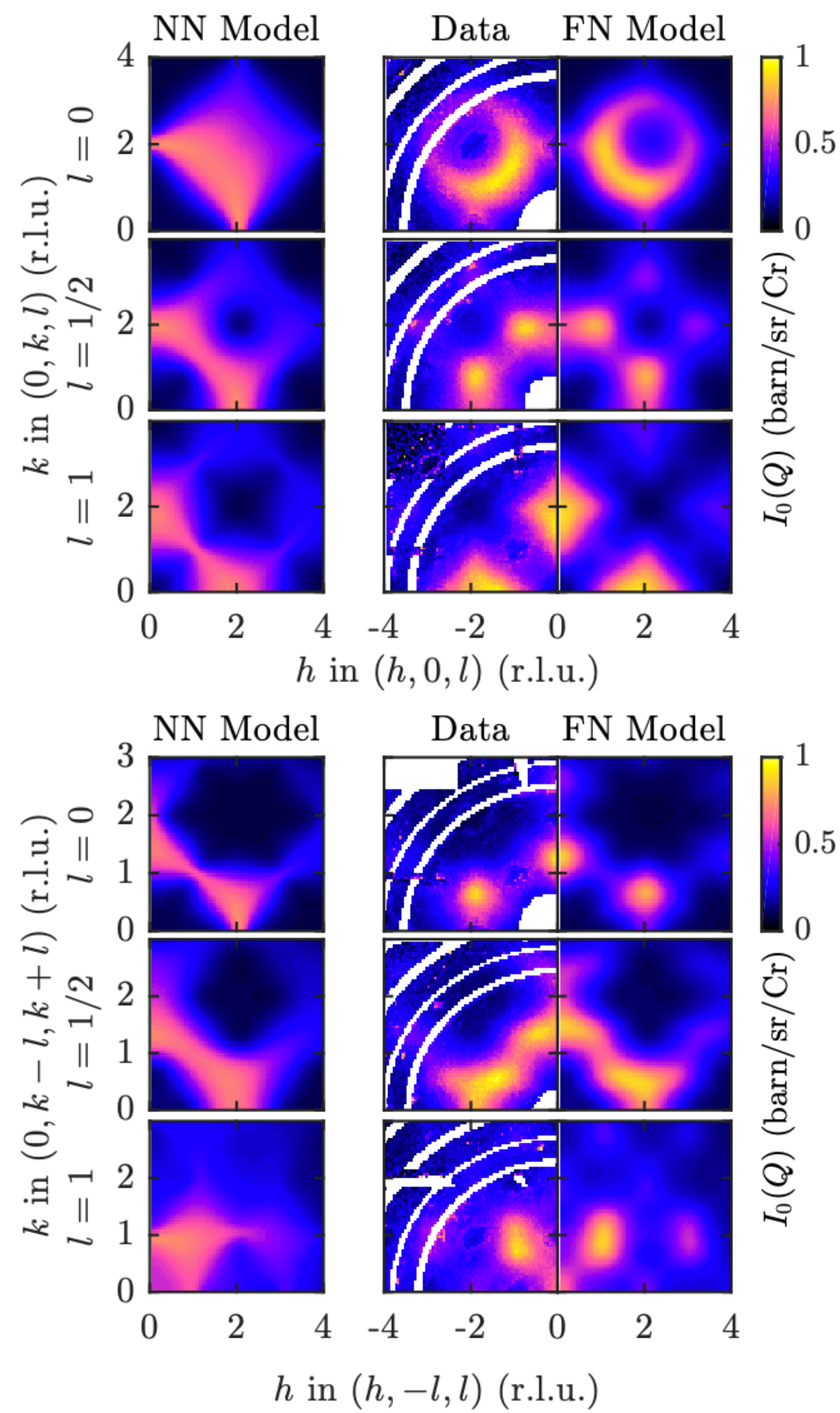
- Ground states: many degenerate/nearly-degenerate disordered states
- Fluctuations: finite-frequency spin waves
- Dynamical picture: spin waves drive the motion between ground states



For system with larger spin?

Approximate classical spin liquid $MgCr_2O_4$

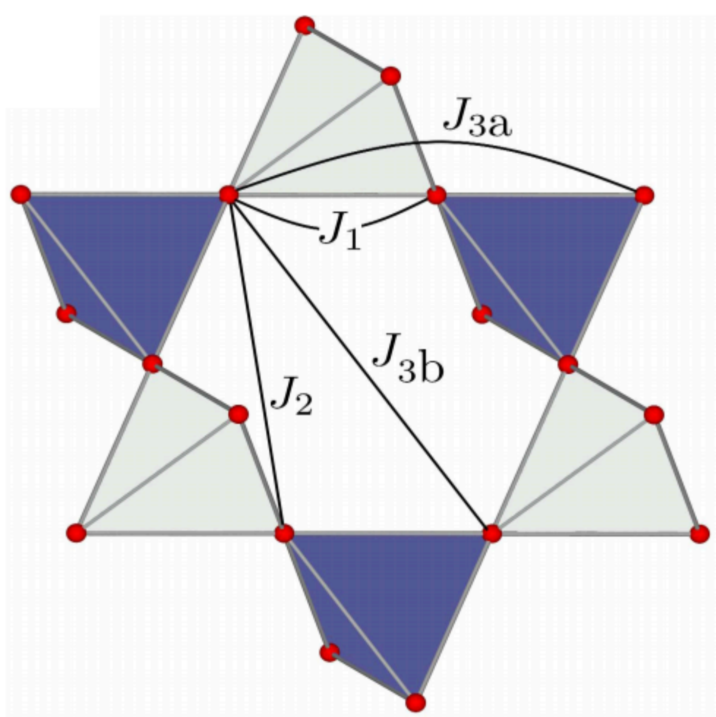
- Cr^{3+} on pyrochlore lattice
- Spin-3/2 system
- Magnetostructural transition at 13K
- Broad scattering patterns at 20K



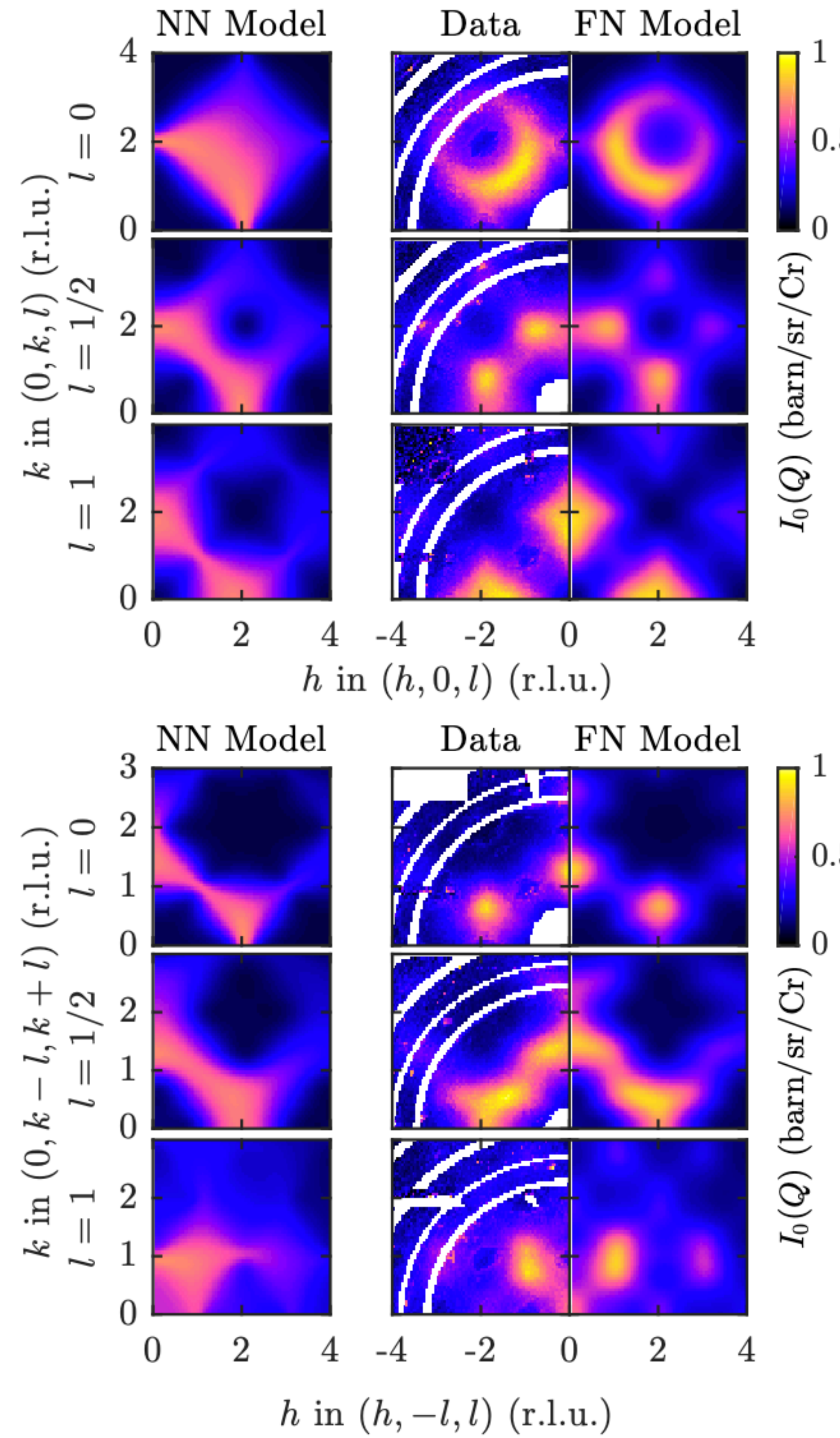
Bai et. al. 2019

Approximate classical spin liquid $MgCr_2O_4$

- Cr^{3+} on pyrochlore lattice
- Spin-3/2 system
- Magnetostructural transition at 13K
- Broad scattering patterns at 20K
- Heisenberg interaction

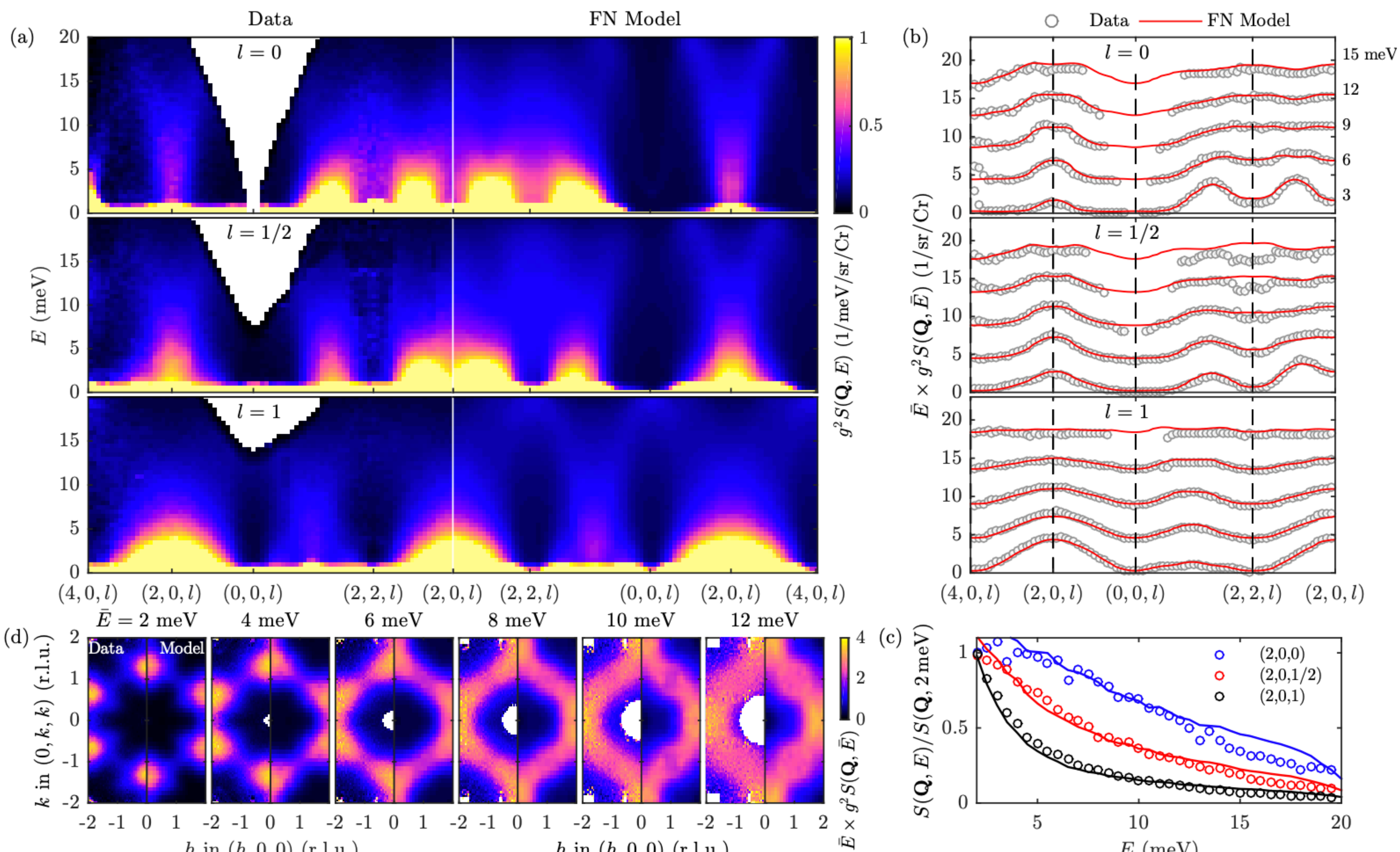


$$J_1 = 3.3,\ J_2 = 0.27,\ J_{3a} = 0.34,\ J_{3b} = 0.03(\mathrm{meV})$$



Bai et. al. 2019

Approximate classical spin liquid $MgCr_2O_4$



Bai et. al. 2019

Discussion

Linear spin wave theory as a good description for

Heisenberg pyrochlore antiferromagnets

Spin-3/2 — Spin-1 — Spin-1/2

$MgCr_2O_4$ — $NaCaNi_2F_7$ — ?

A peek into quantum spin liquids

with classical tools

Summary

- Heisenberg pyrochlore antiferromagnet
- Theoretical modeling
  - Stochastic large-n + linear spin wave theory + molecular dynamics
  - Spin-1 quantum spin liquid $NaCaNi_2F_7$
  - Spin-3/2 classical spin liquid $MgCr_2O_4$
- A general picture for spin dynamics
  - Slow & fast motion
- Quantum nature?

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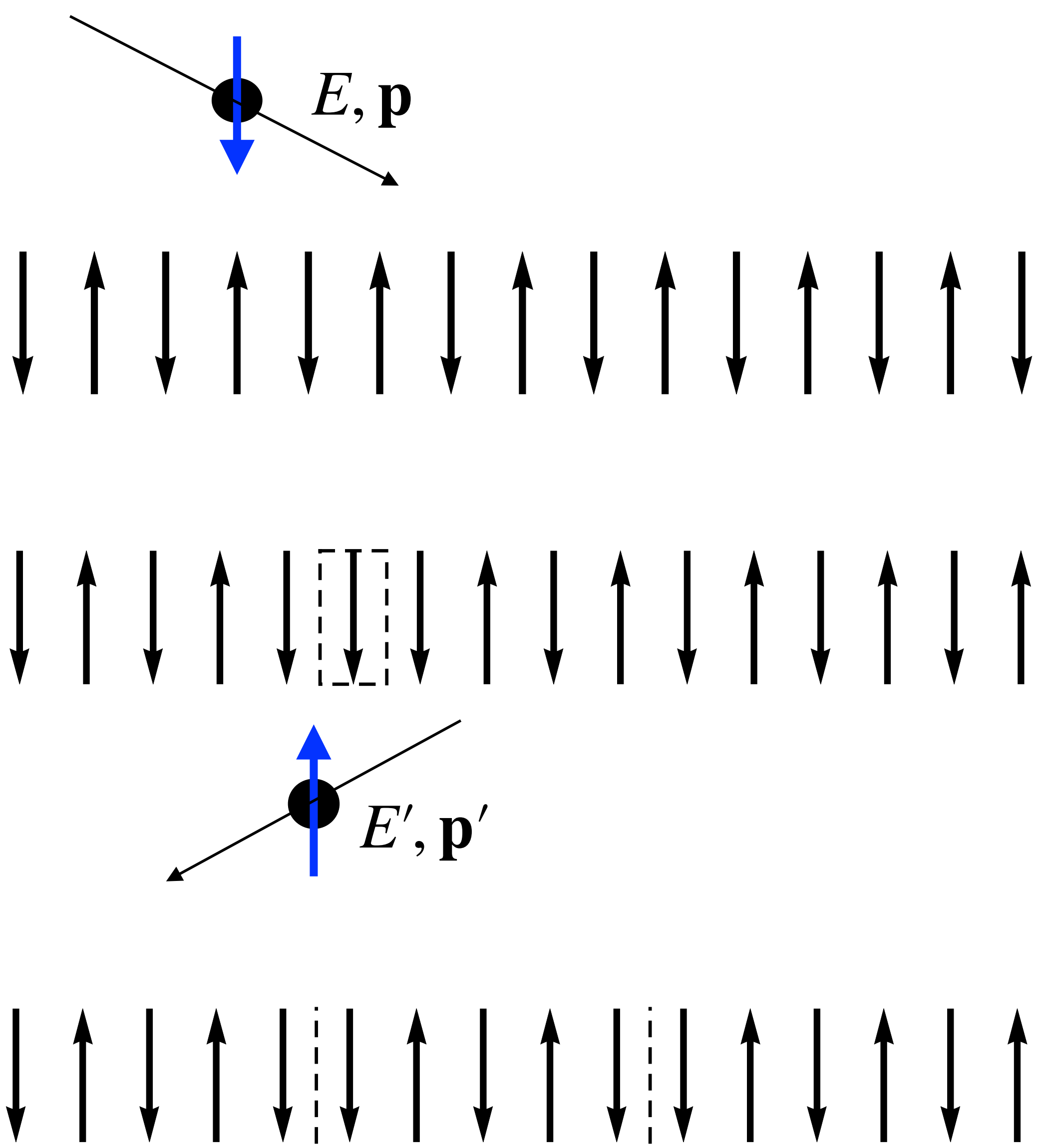
- Computation: HHPC&MARCC, Maryland

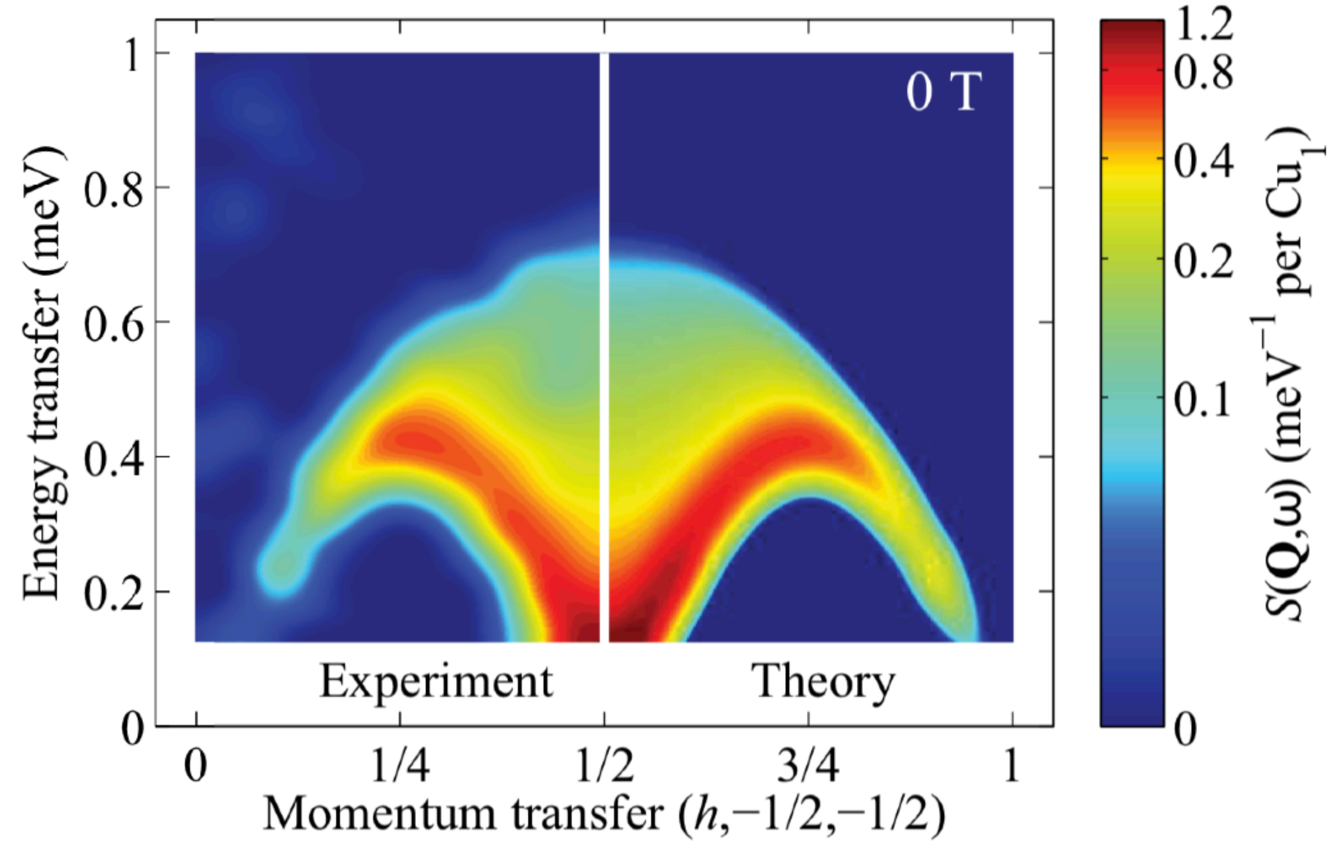
- Experiments: NIST&Oak Ridge

Fractionalized excitations



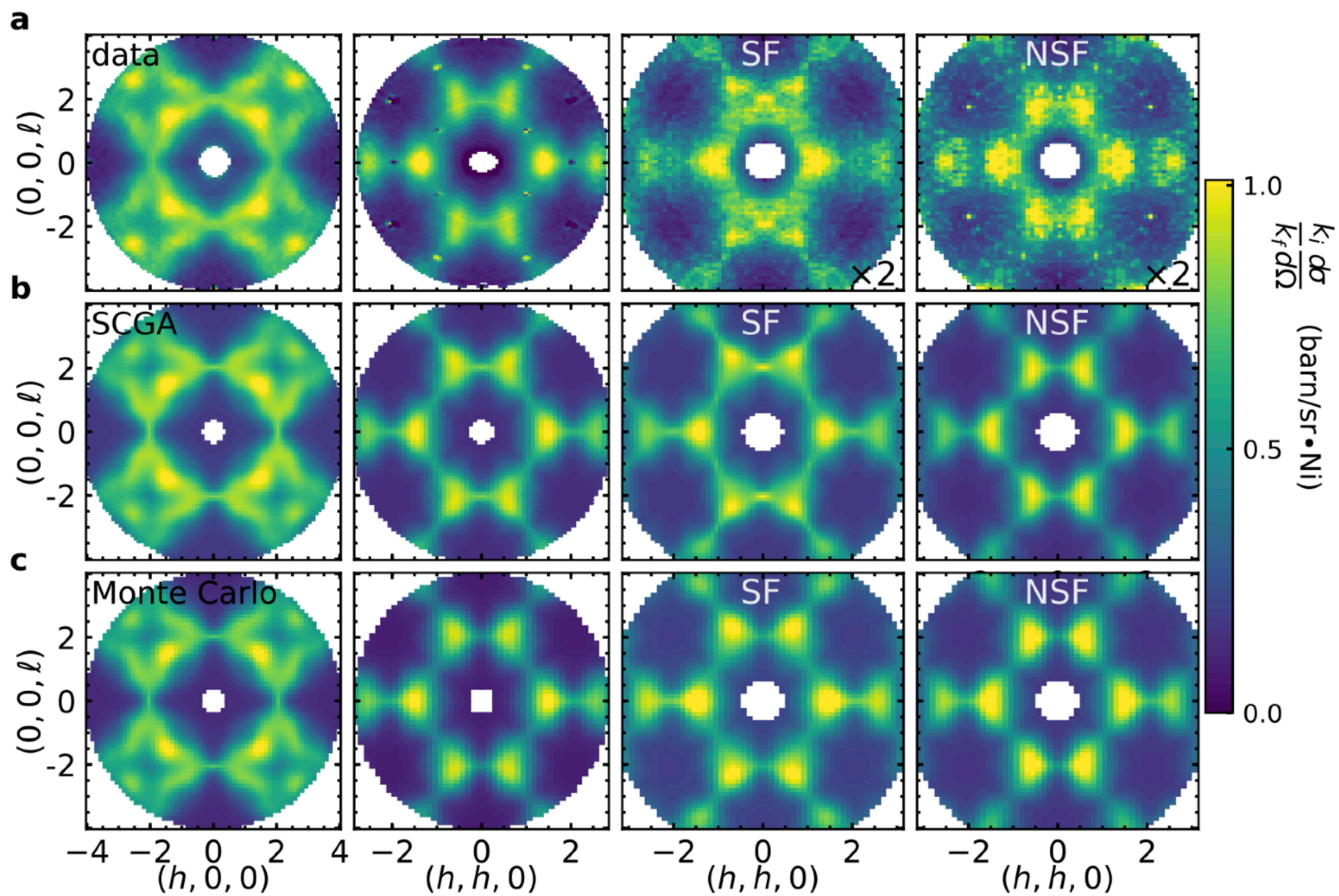
Two spinons sharing
$\Delta E = E' - E,\ \Delta \mathbf{p} = \mathbf{p}' - \mathbf{p},\ \Delta \mathcal{J} = -\hbar$

- Created in pairs
- Fractional charge

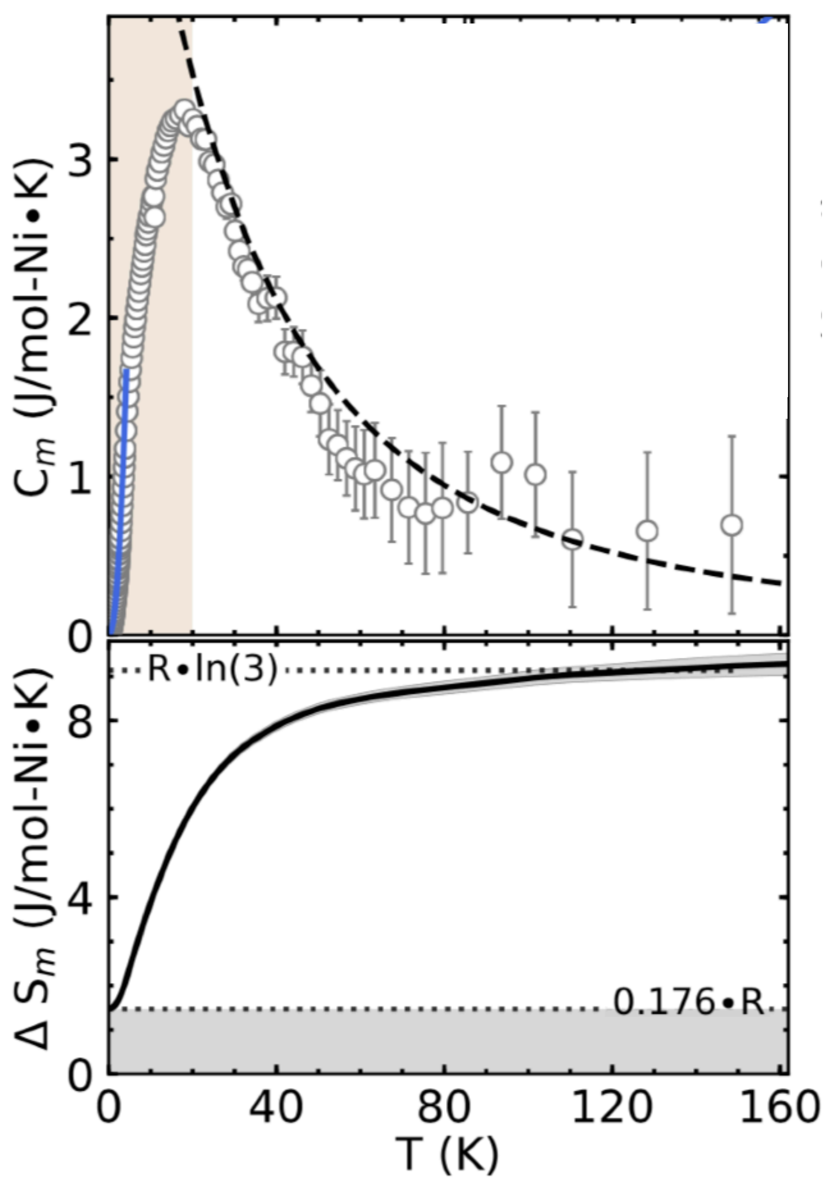


Mourigal et. al. 2013

Fit the exchange parameters



Specific heat



Ground states