Continuous magnetic spectra in absence of quasiparticle fractionalization

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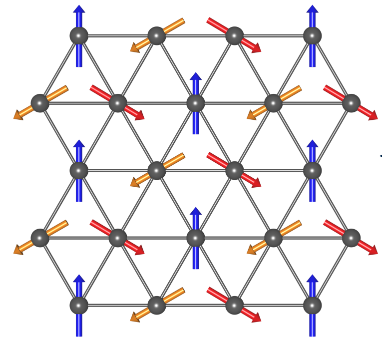
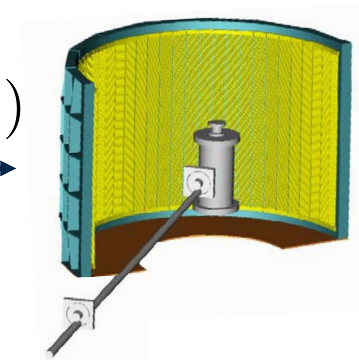
TOPOQUANT
KITP, Santa Barbara
October 14, 2019

Outline

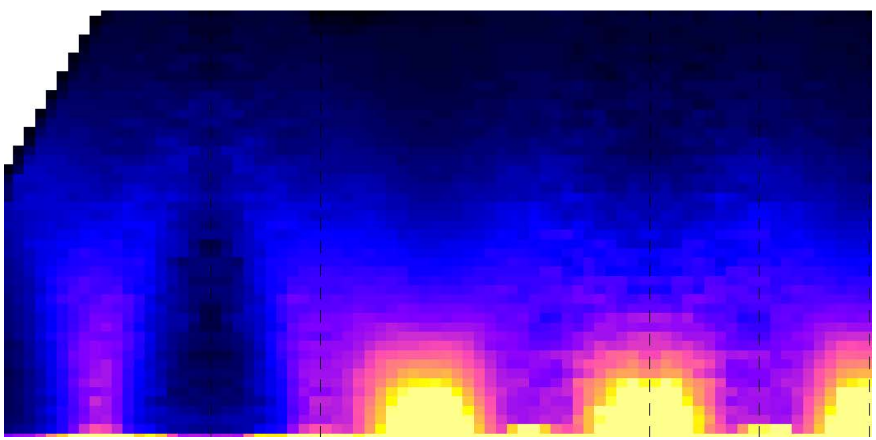
1. Introduction and neutron scattering warm-up



$$S^{\alpha\beta}(\mathbf{Q},\omega)$$

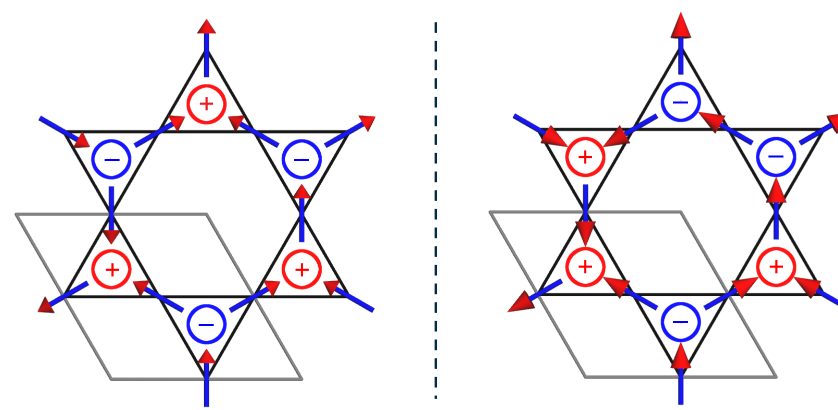


2. Nature of excitations in the classical pyrochlore Heisenberg AFM $MgCr_2O_4$



Bai *et al.* Phys. Rev. Lett. **122**, 097201 (2019)

2. Kagome spin-ice physics in the tripod compounds $\mathit{Ln}_3Mg_2Sb_3O_{14}$



Paddison *et al.*, Nat. Commun. **7**, 13842 (2016)
Dun *et al.*, arXiv:1806.04081 (2019) + in prep.

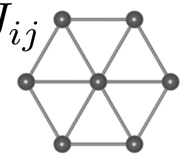
Magnetic quantum matter in Mott insulators

- The foundations of quantum magnetism:

$$\mathcal{H} = \sum_{(ij)} \hat{\mathbf{S}}_i J_{ij} \hat{\mathbf{S}}_j =$$

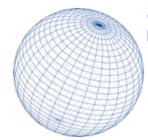
magnetic system

J_{ij}



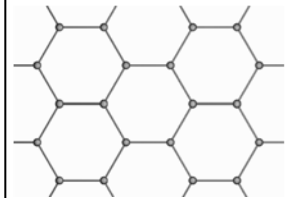
lattice-space

$\otimes$

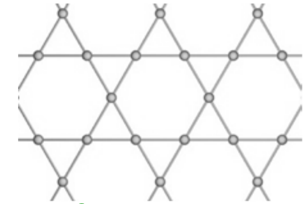


$\hat{\mathbf{S}}$

spin-space



bi-partite



frustrated

lattice geometry

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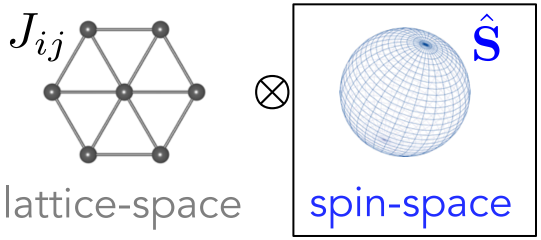

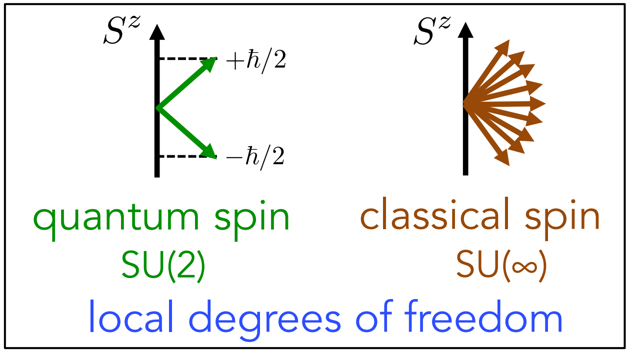

Magnetic quantum matter in Mott insulators

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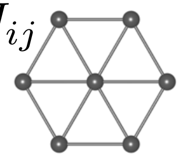
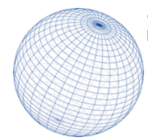
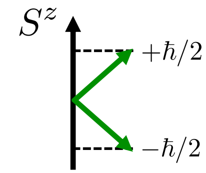


lattice-space

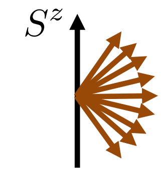
$\otimes$



spin-space

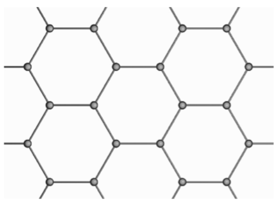


quantum spin
SU(2)



classical spin
SU(∞)

local degrees of freedom



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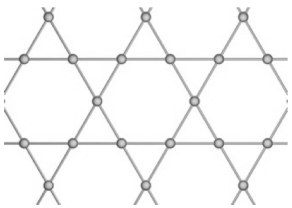
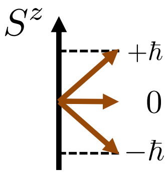


frustrated

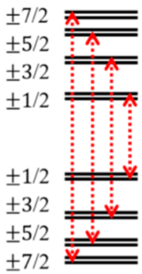
lattice geometry



SU(3) spin

Low-lying crystal-field

Nuclear hyperfine



larger on-site Hilbert space

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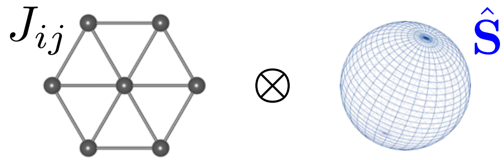
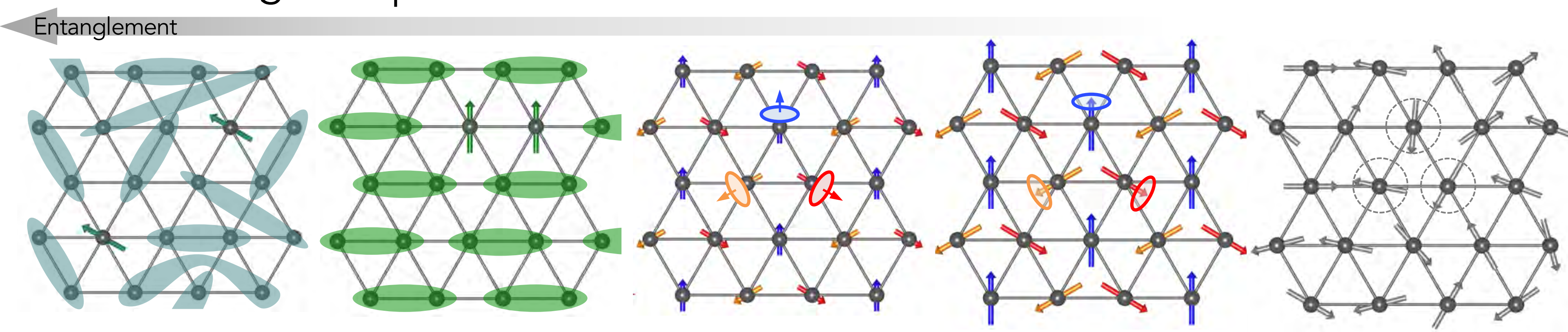


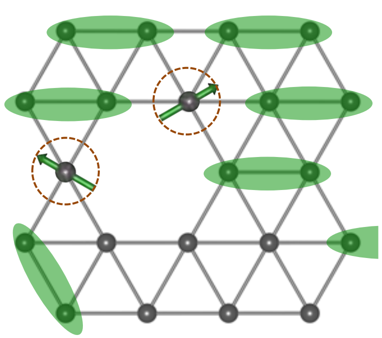
magnetic system — lattice-space — spin-space

- Clean magnetic phases and their excitations:

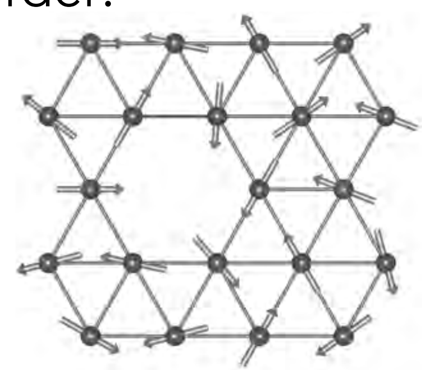


Quantum spin liquid | Quantum paramagnet | Fluctuating order | Ordered magnet | Classical paramagnet

- Extra challenge from chemical heterogeneities/disorder:



Valence Bond Glass



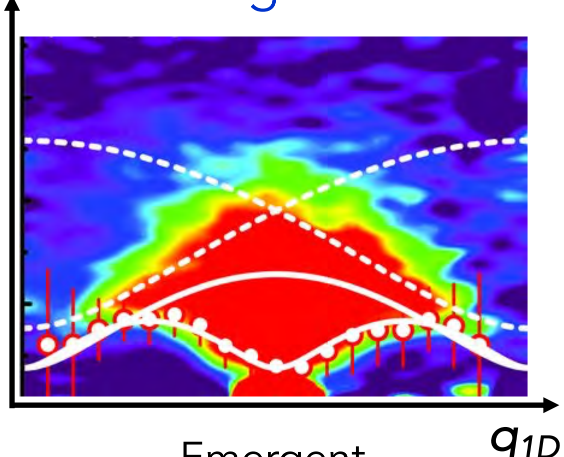
Spin Glass

The quest for a quantum spin-liquid

- Experimentalist view:

today

1D quantum magnets

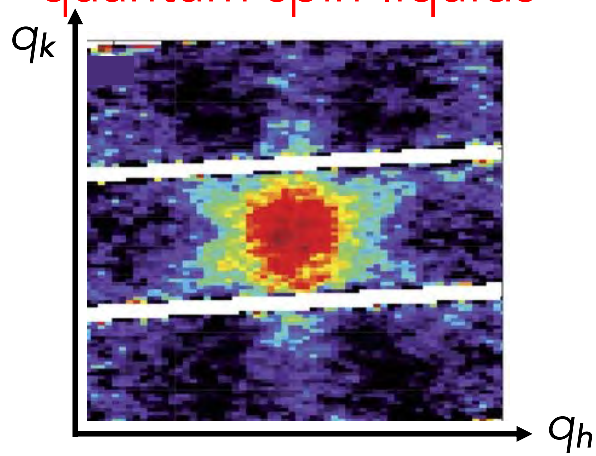


Emergent antiferromagnetic chains in metallic Yb_2Pt_2Pb

Wu *et al.*, Science **352**, 1206 ('16)

Continuous in energy

2D and 3D quantum spin-liquids



Kitaev honeycomb

Banerjee *et al.*, Science **356**, 1055 ('17)

Heisenberg kagome

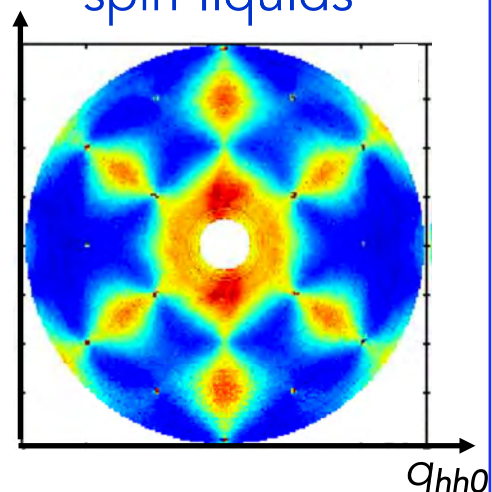
Han *et al.*, Nature **492**, 406 ('12)

Distorted bilayer kagome

Balz *et al.*, Nature Physics **12**, 942 ('16)

Continuous response in both momentum & energy

3D classical spin-liquids



Spin-ice pyrochlore in $Ho_2Ti_2O_7$

Fennell *et al.*, Science, 326, 415 ('09).

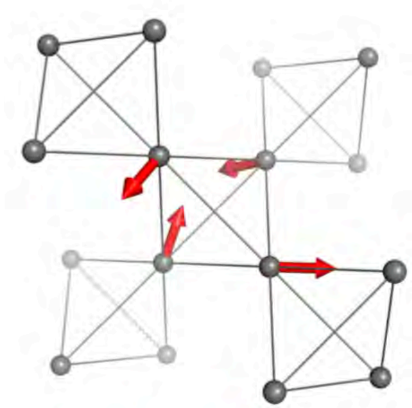
Continuous in momentum

The quest for a quantum spin-liquid

❑ Experimentalist view:

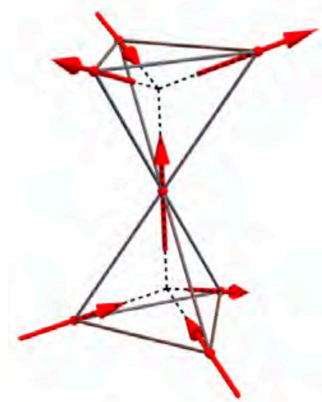
today

$MgCr_2O_4$



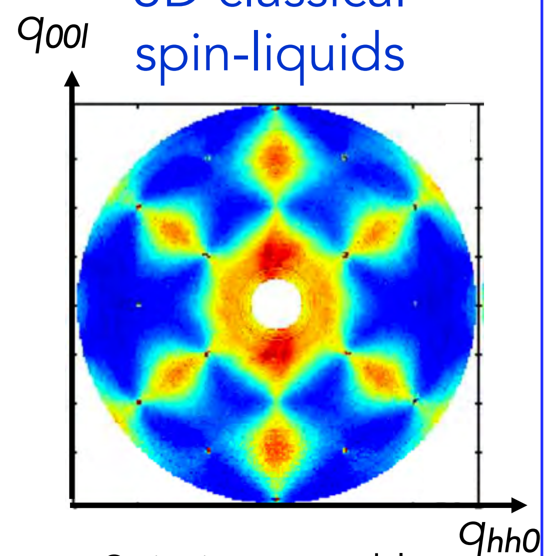
Heisenberg

$Ln_3Mg_2Sb_3O_{14}$



Ising/spin-ice

3D classical spin-liquids



Spin-ice pyrochlore in $Ho_2Ti_2O_7$

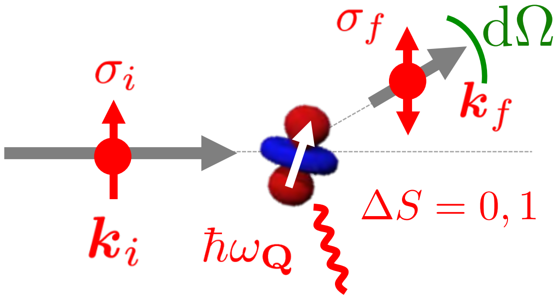
Fennell *et al.*, Science, 326, 415 ('09).

❑ General question:

In general, a continuous excitation spectrum is not enough to conclude on fractional excitations. So how to discover a quantum spin-liquid?

Technique: Neutron Scattering

- Magnetic Scattering: ideal but weak thus requires "large" samples



$$\left.\frac{d^2\sigma}{dE_f d\Omega}\right|_{\mathrm{mag}} = \frac{k_f}{k_i} r_0^2 \, |gf(\mathbf{Q})|^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) \, \mathcal{S}^{\alpha\beta}(\mathbf{Q}, \omega)$$

measured cross-section; magnetic form-factor; dipole factor; dynamic structure-factor

$$\mathbf{Q} = \boldsymbol{k}_i - \boldsymbol{k}_f$$

$$\hbar\omega = \frac{\hbar^2(\boldsymbol{k}_i^2 - \boldsymbol{k}_f^2)}{2m_n}$$

$$\Delta S = \sigma_i - \sigma_f$$

Conservation laws

Fourier transform

$$\mathcal{S}^{\alpha\beta}(\mathbf{Q}, \omega) = \frac{1}{2\pi\hbar} \int dt e^{-i\omega t} \frac{1}{N} \sum_{\mathbf{r}\mathbf{r}'} e^{-i\mathbf{Q}\cdot(\mathbf{r}-\mathbf{r}')} \langle S^\alpha(\mathbf{r}, t) S^\beta(\mathbf{r}', 0) \rangle$$

dynamic s.f.; dynamic pair correlations

$$\mathcal{S}^{\alpha\beta}(\mathbf{Q}) = \int \mathcal{S}^{\alpha\beta}(\mathbf{Q}, \omega) d\omega = \frac{1}{N} \sum_{\mathbf{r}\mathbf{r}'} e^{-i\mathbf{Q}.(\mathbf{r}-\mathbf{r}')} \langle S^\alpha(\mathbf{r}) S^\beta(\mathbf{r}') \rangle$$

instantaneous s.f.; instantaneous pair correlations

Technique: Neutron Scattering

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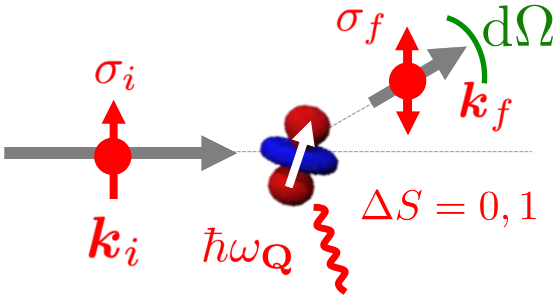

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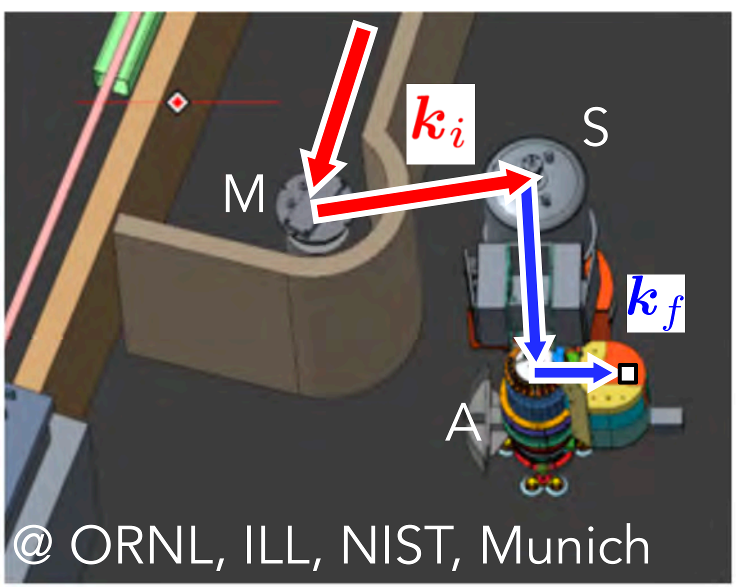
measured cross-section | magnetic form-factor | dipole factor | dynamic structure-factor

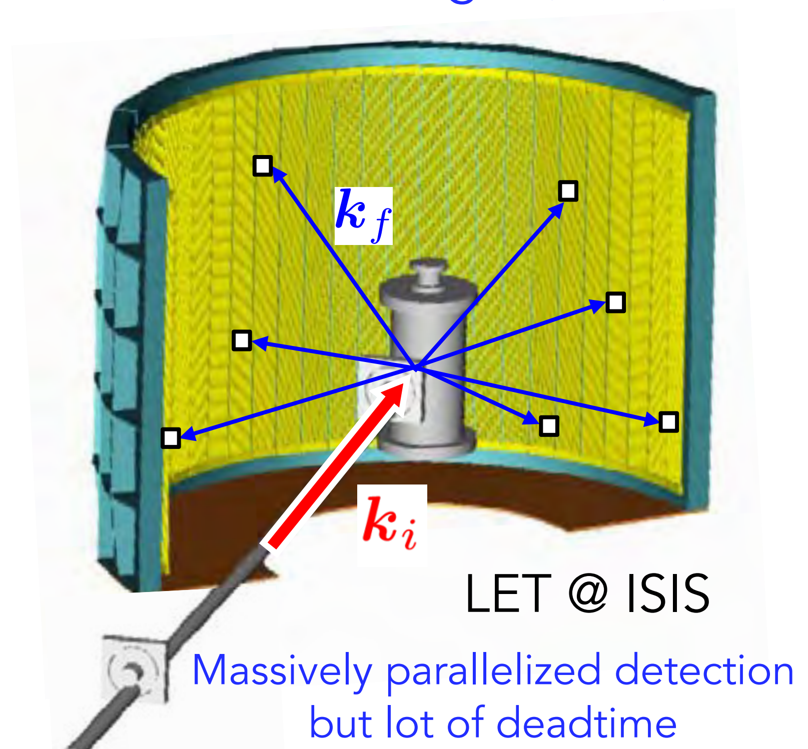
- Instrumentation: designed to detect a broad Q-E response

Triple-axis (TAS)



Point by point detection

Time-of-flight (TOF)



Massively parallelized detection but lot of deadtime

Technique: Neutron Scattering

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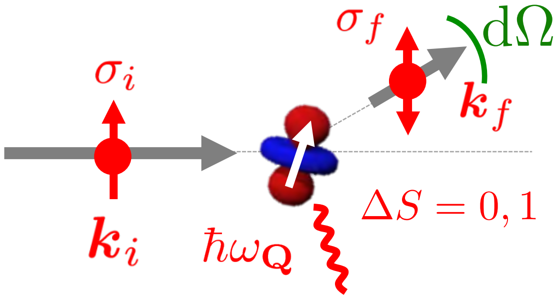

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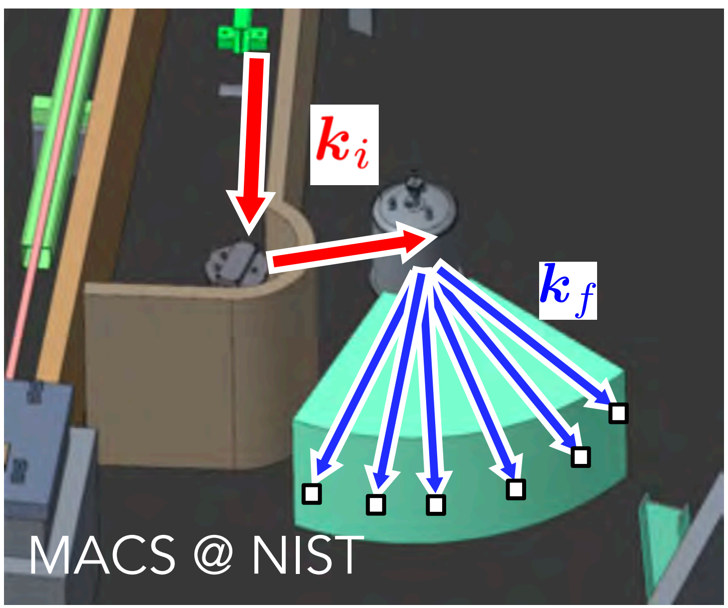
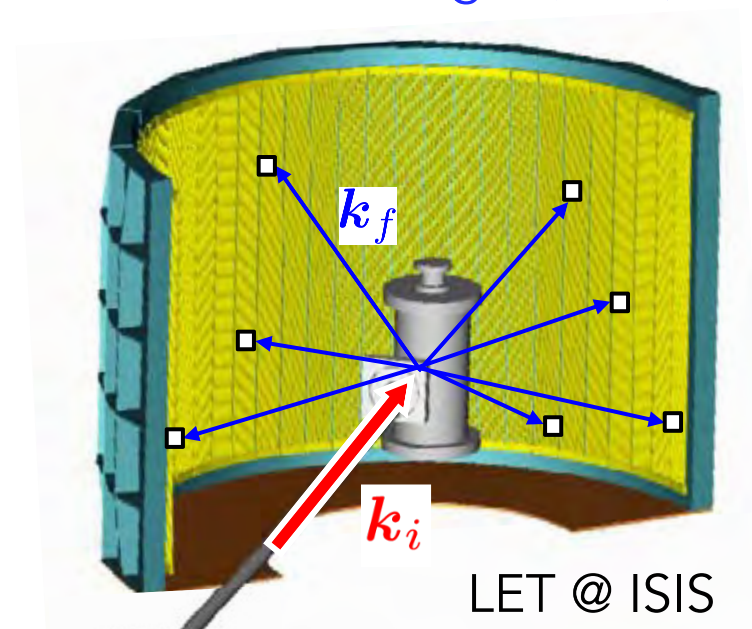
Multiplexed triple-axis (TAS)



Plane by plane detection
Focusing optics

Time-of-flight (TOF)



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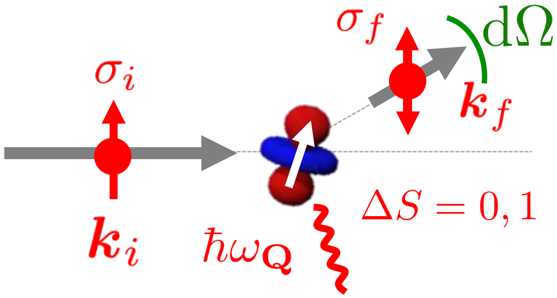

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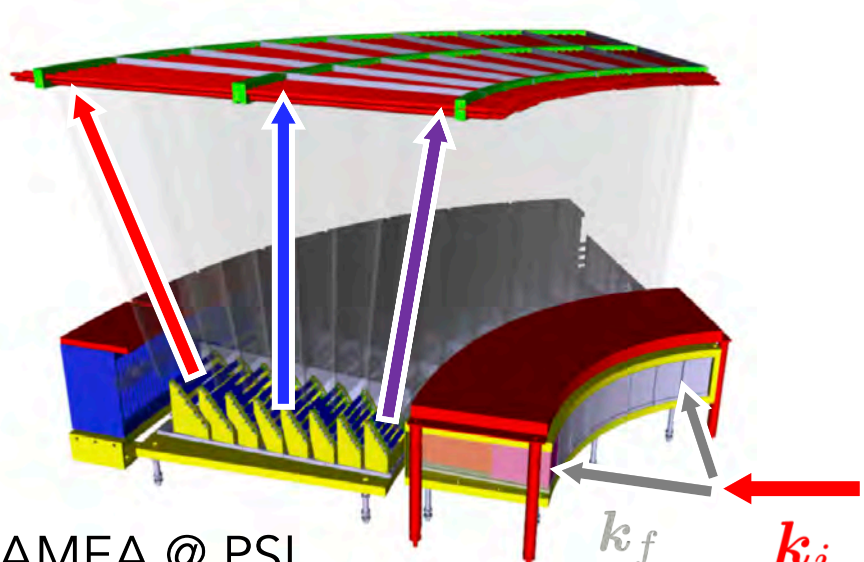
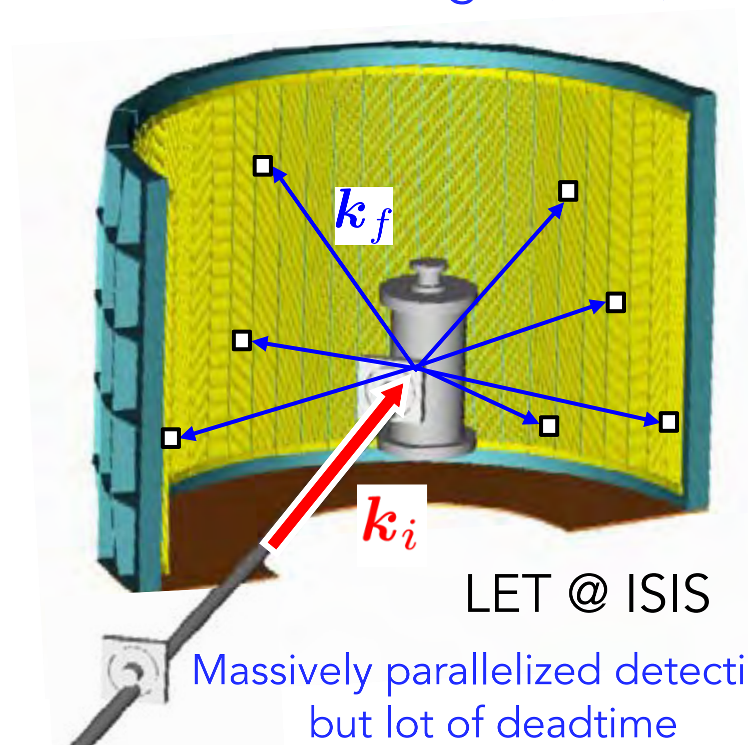


CAMEA @ PSI

Prismatic detection
Focusing optics

Time-of-flight (TOF)



LET @ ISIS

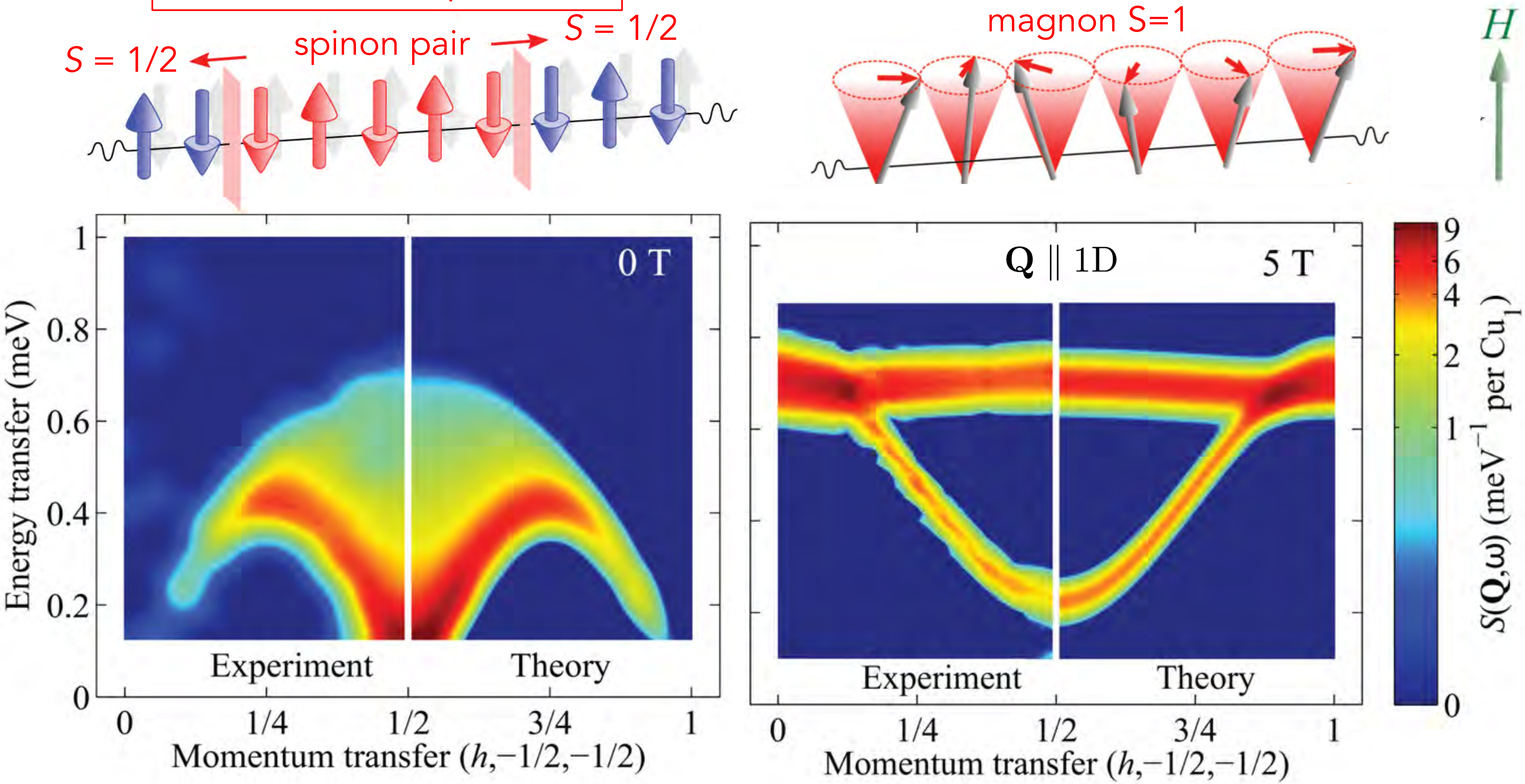
Massively parallelized detection
but lot of deadtime

Spin liquid *vs* Ordered magnet – What to expect

- 1D chains in $CuSO_4.5D_2O$ (H_s=3.4T)

Entangled ground-state
Fractional excitations
Continuous spectra

Ordered ground-state
Spin-wave excitations
Sharp spectra

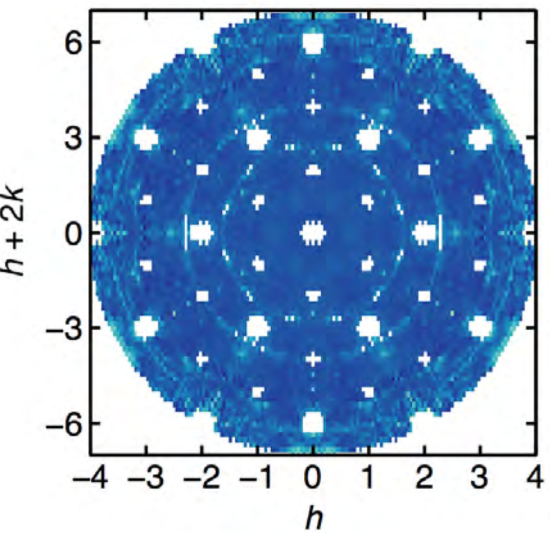


Mourigal, Enderle, Rønnow, Caux, Nature Physics **9**, 435-441 (2013)

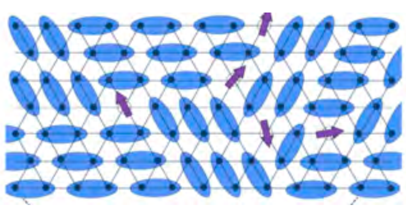
Spin liquid *vs* Random singlets – The unexpected

- ❑ Rare-earth triangular in Yb(MgGa)O_4 (H_s~5.0T)

Continuous spectra in both 0T and 7.8T

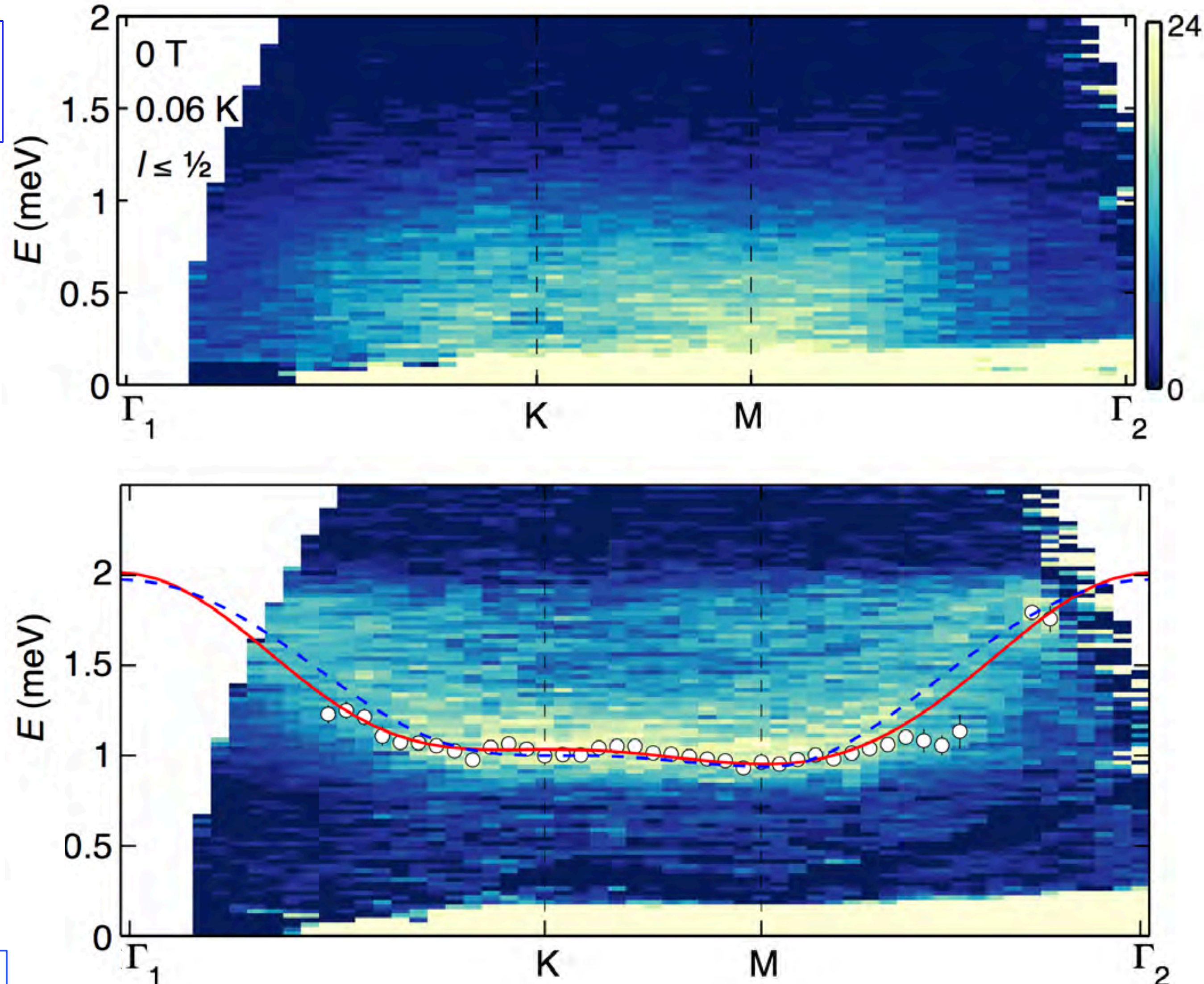


Structural Diffuse Scattering



Kimchi, Senthil PRX'18
Tsirlin, Gegenwart PRL'19

Correlated disorder
Random valence bonds?



Paddison, Dun, Daum, Zhou, Mourigal, Nature Physics **13**, 117 (2017)

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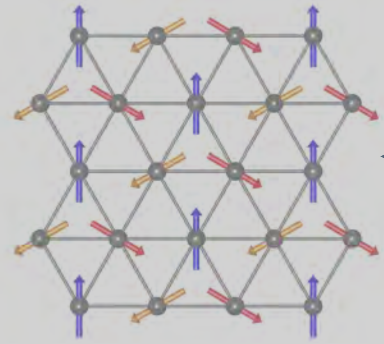
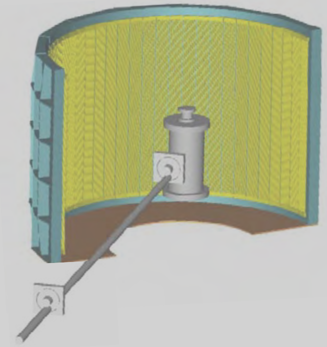


$S^{\alpha\beta}(\mathbf{Q}, \omega)$



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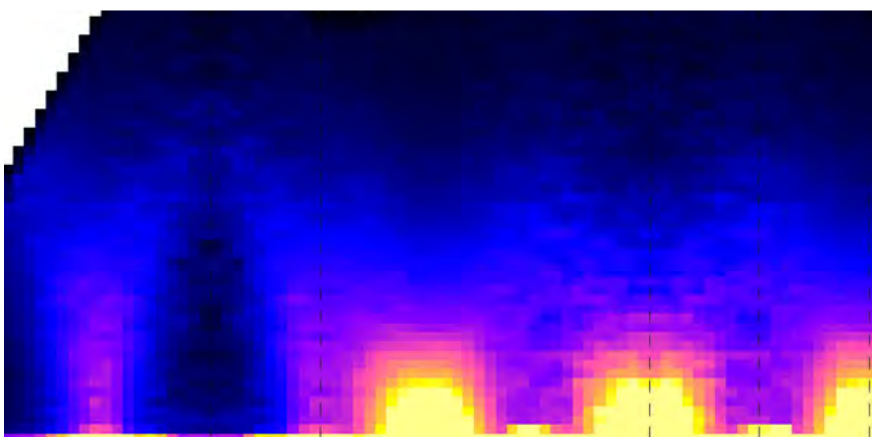
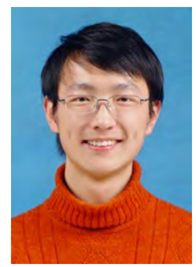


Bai *et al.* Phys. Rev. Lett. **122**, 097201 (2019)



Xiaojian Bai

Seyed Koohpayeh

Eliot Kapit

John Chalker

Siân Dutton

Joe Paddison

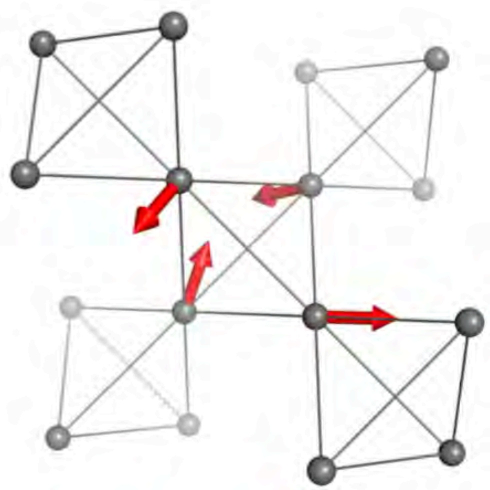
Jiajia Wen

Collin Broholm

Garrett Granroth (ORNL), Ovi Garlea (ORNL), Andrei Savici (ORNL), Sasha Kolesnikov (ORNL)

Large-S pyrochlore Heisenberg antiferro.

❑ A paradigmatic classical spin-liquid model:



Large spins
(n→∞ components)

spins are under-constrained!

$$H = J \sum_{<i,j>} \mathbf{S}_i \cdot \mathbf{S}_j \equiv \frac{J}{2} \sum_{\alpha} |\mathbf{L}_\alpha|^2 + \text{cte} \qquad \mathbf{L} = \sum_{i=1}^{4} \mathbf{S}_i$$

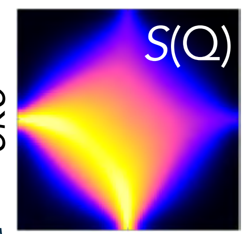
nearest-neighbor interaction

rewrite as sum over tetrahedra

spin liquid — paramagnet

0 — θ_W — T

extensive degeneracy



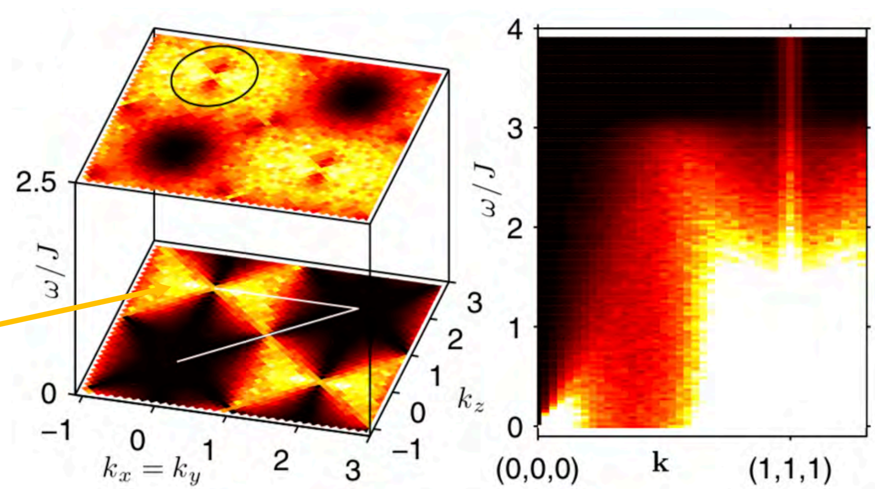
Correlations decay algebraically in real space such that sharp pinch-points appear in reciprocal space. An example of a Coulomb Phase.

Anderson 1956, Villain 1979, Reimers 1992, Moessner & Chalker 1998, Henley 2011

❑ Dynamics is very rich:

molecular dynamics $T\sim J/500$

zero-energy (quasi-elastic) spectral weight



Relaxational

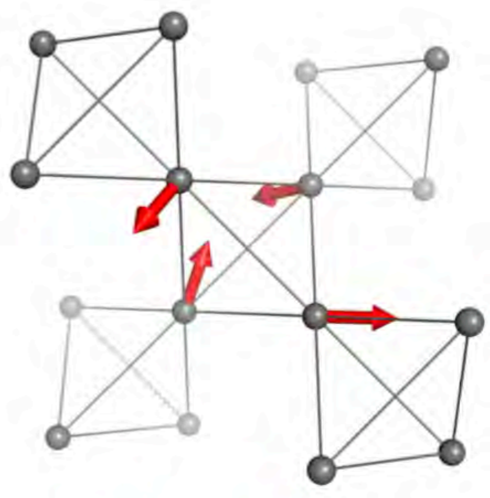
Precessional

Diffusive

Conlon and Chalker PRL 2009

Large-*S* pyrochlore Heisenberg antiferro.

- A paradigmatic classical spin-liquid model:



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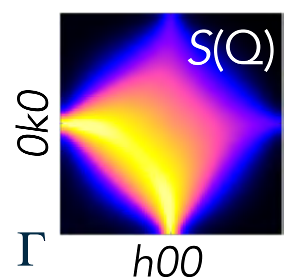
0 — θ_W — *T*

extensive degeneracy

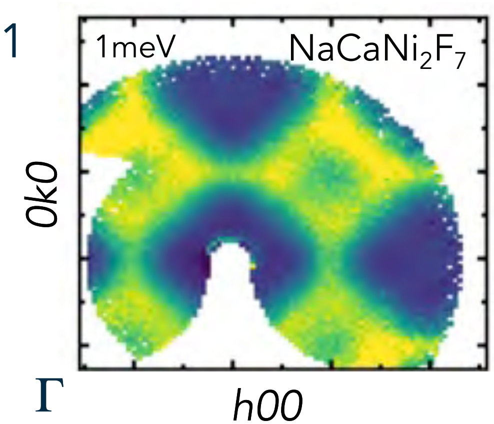


Correlations decay algebraically in real space such that sharp pinch-points appear in reciprocal space. An example of a Coulomb Phase.

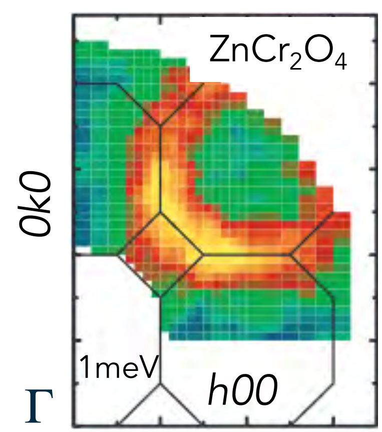
Anderson 1956, Villain 1979, Reimers 1992, Moessner & Chalker 1998, Henley 2011

- Approximated e.g. by Na*A'B*$_2$F$_7$ pyrochlore fluorides and cubic spinels *AB*$_2$O$_4$

S=1

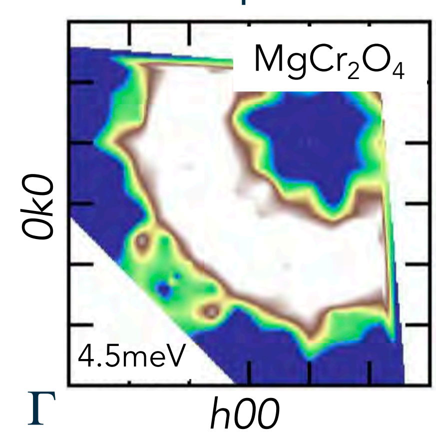


Plumb *et al.* Nature Physics 2019



S.H. Lee *et al.* Nature 2002

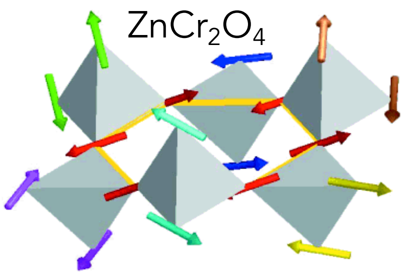
S=3/2



Tomiyasu *et al.*, PRL 2008

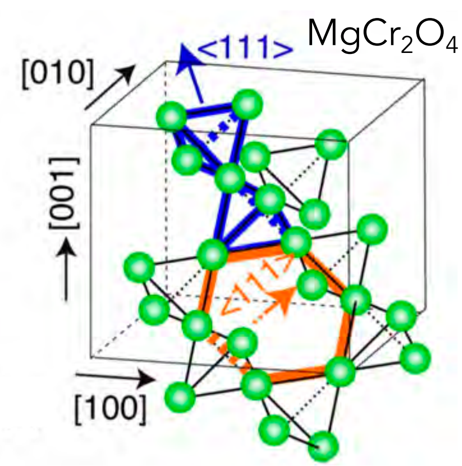
Loop Scattering in Spinels

- Absent pinch-points & excitations in ordered phase: emergent spin clusters?



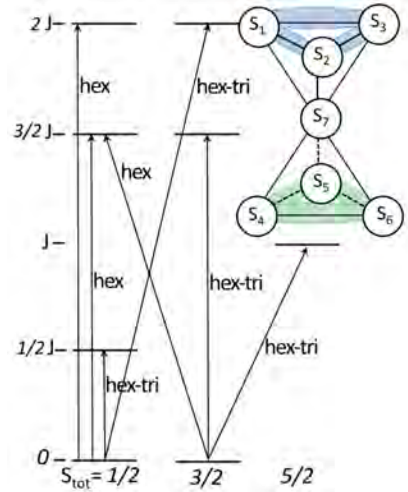
Hexamers

Tchernyshyov 2002
S.H. Lee *et al.* Nature 2002



Molecular spin resonances

Tomiyasu PRL 2008
Tomiyasu PRL 2012

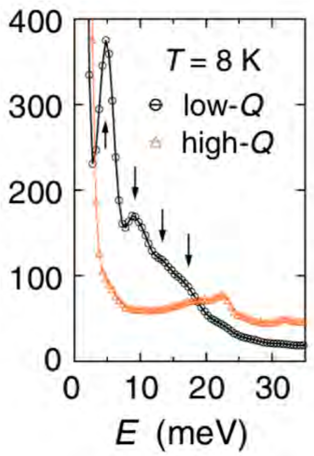


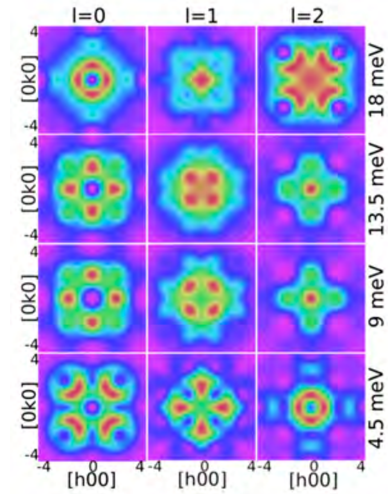
Heptatmers

Gao PRB 2018
Haraldsen PRB 2018

Loop inflation

Motivation: different clusters to explain magnetic resonances in the ordered phase and their momentum dependence



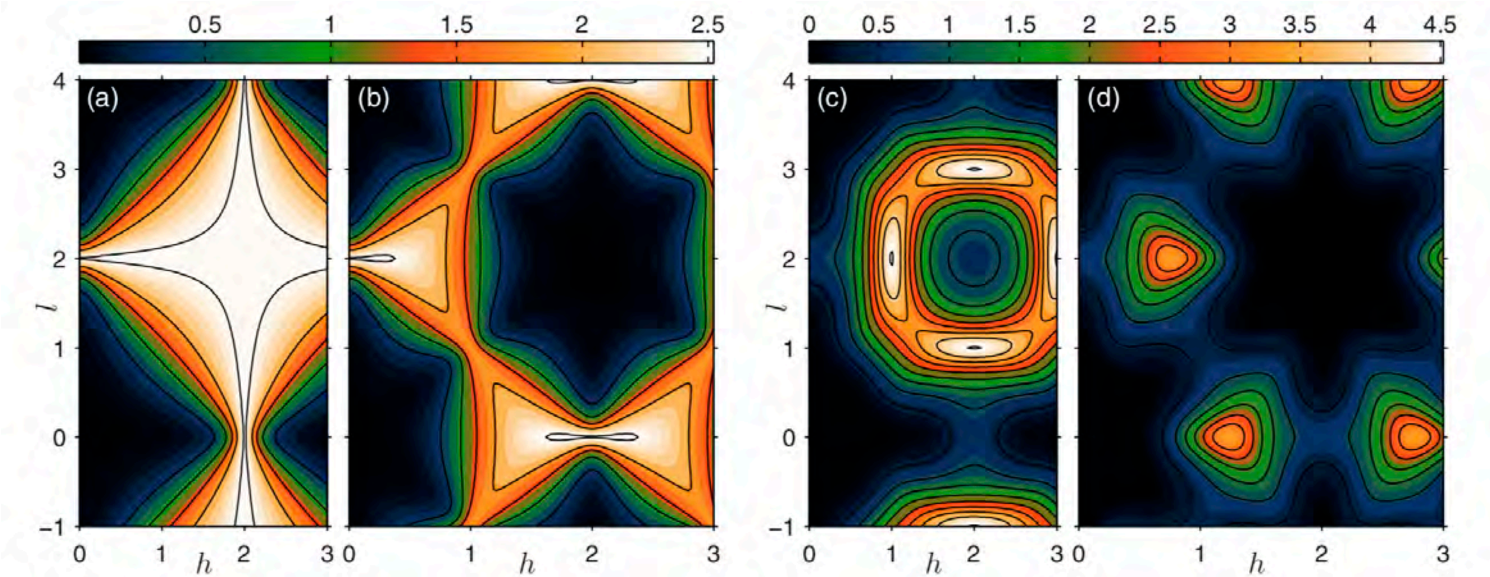


What is the origin of the loop scattering?

- Theoretical work by Conlon & Chalker attributing it to further-neighbor ex.

Nearest Neighbor Model

Further Neighbor Model



Conlon and Chalker, PRB 2010

What is the origin of the loop scattering?

- Theoretical work by Conlon & Chalker attributing it to further-neighbor ex.
- $MgCr_2O_4$: grew and co-aligned ~14 grams of single-crystals

Koohpayeh, Wen *et al.*
J. Cryst. Growth 2011

Dutton *et al.*
PRB 2010

What is the origin of the loop scattering?

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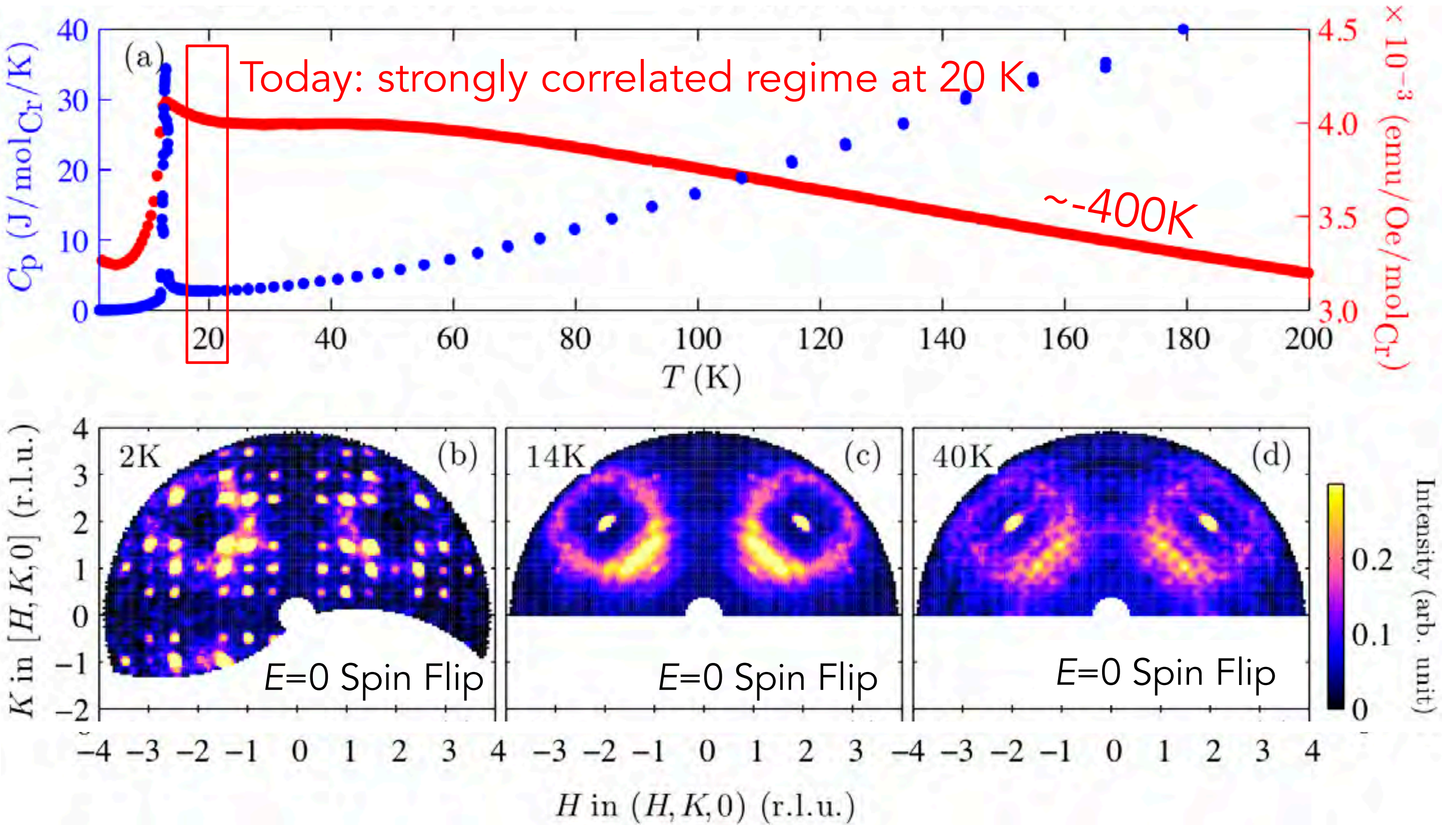

Polarized Elastic Scattering

magneto-structural ordering

coexistence of broad and sharp features

highly structured diffuse scattering

What is the origin of the loop scattering?

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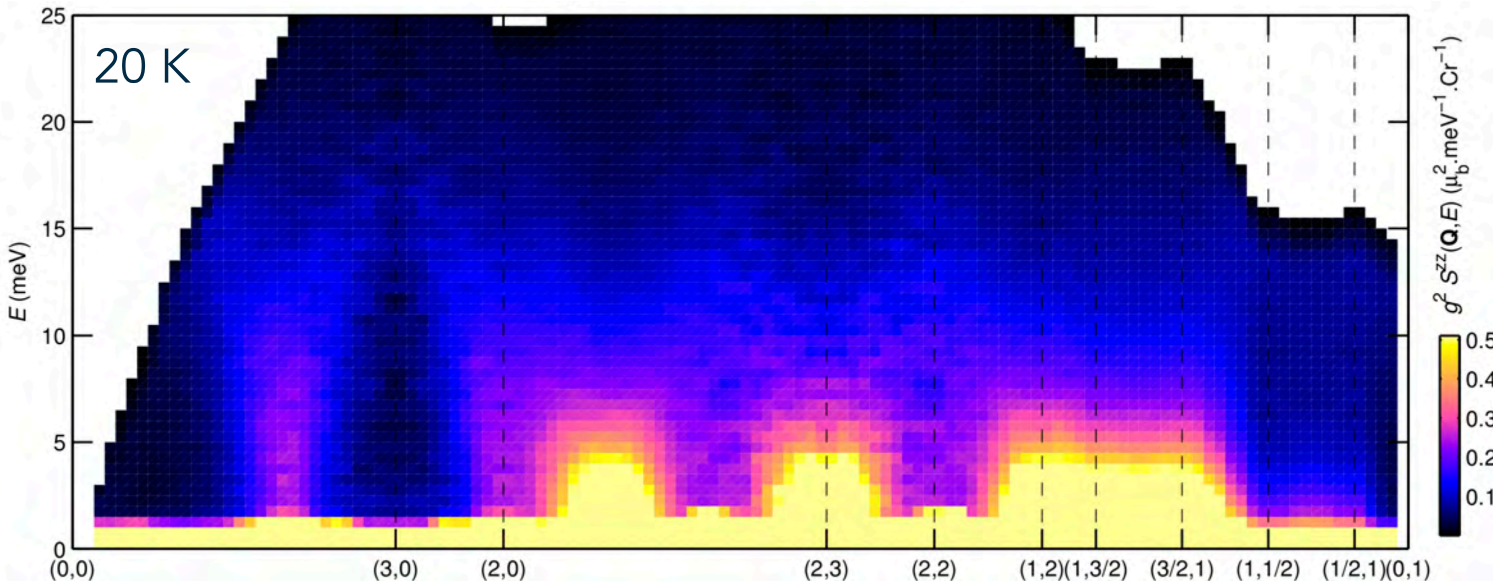

Q = (H,K,0)

Complete map of the inelastic spectrum

All measurements are performed at 20 K which is 5% of the Weiss constant

How to analyze such a large and broad "4D" dataset?

Analysis of energy-integrated data

- For a spin-space isotropic magnet with Heisenberg exchange interactions:

Structure Factor

$$\mathcal{S}(\mathbf{Q}) = \int_{-\infty}^{+\infty} \mathrm{d}E\, \mathcal{S}(\mathbf{Q}, E)$$

$$= \frac{2}{3N} \sum_{ij} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \cos(\mathbf{Q} \cdot \mathbf{r}_{ij})$$

First Moment

Hohenberg 1974
Broholm 2001

$$\mathcal{K}(\mathbf{Q}) = \int_{-\infty}^{+\infty} \mathrm{d}E\, E\, \mathcal{S}(\mathbf{Q}, E)$$

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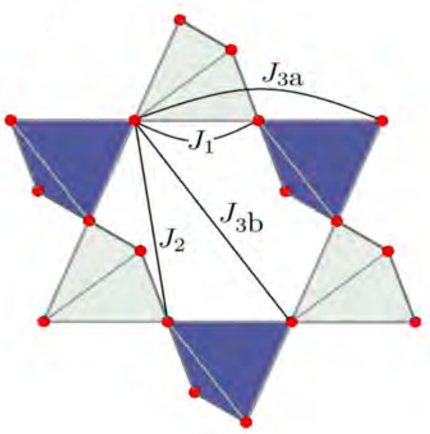
Fit then divide Fourier coefficients

- Further-neighbor interactions on the pyrochlore lattice:



$J3_a$ and $J3_b$ span the same length

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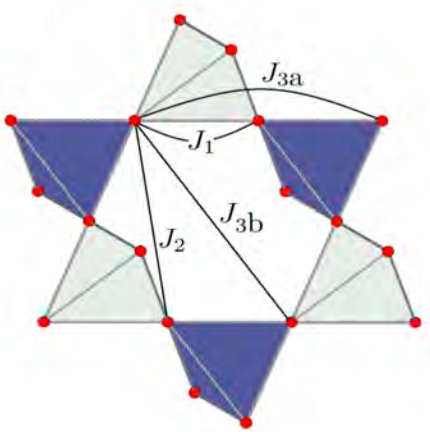
Fit then divide Fourier coefficients

- Further-neighbor interactions on the pyrochlore lattice: "microscopic" theory

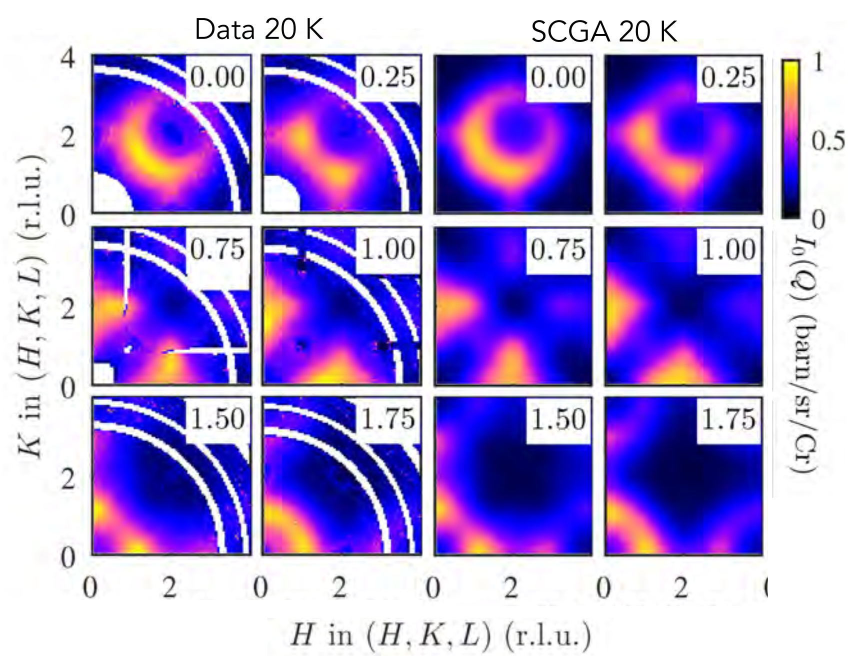


Self Consistent Gaussian Approx.

Soft spin constraint + Sum rule

Calculates $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_T$ given J_{ij}

Global Fit



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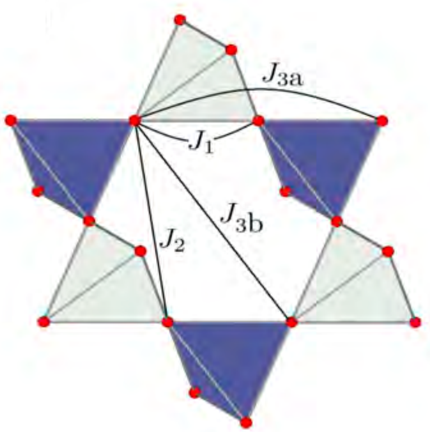

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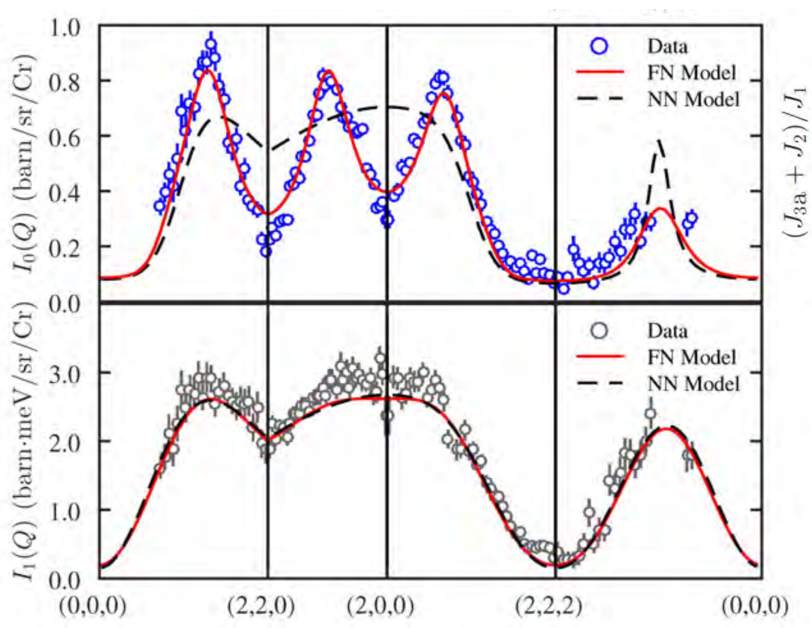

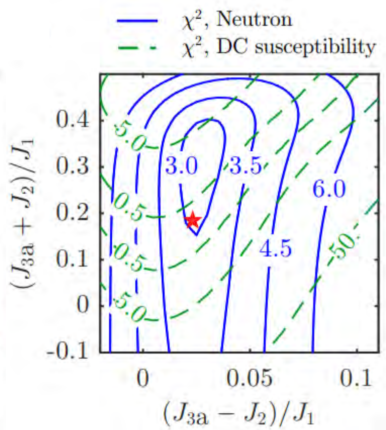

J_1 = 3.28(9) meV
J_2 = 0.081 J_1
J_{3a} = 0.105 J_1
J_{3b} = 0.009 J_1
T = 20 K

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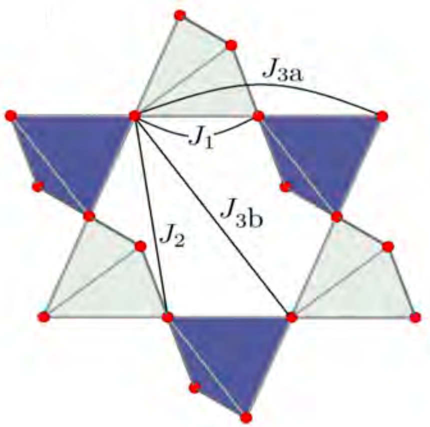

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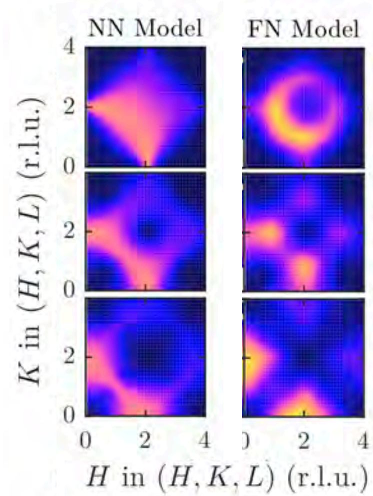
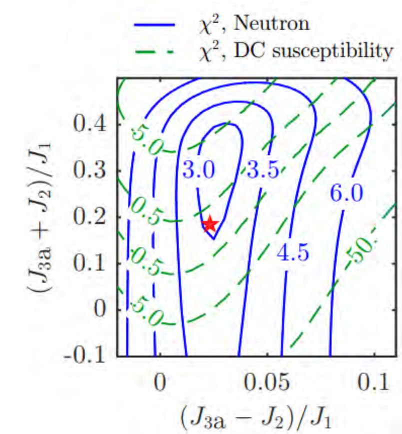


FN interactions destroy the pinch point and produce the ring-like scattering pattern



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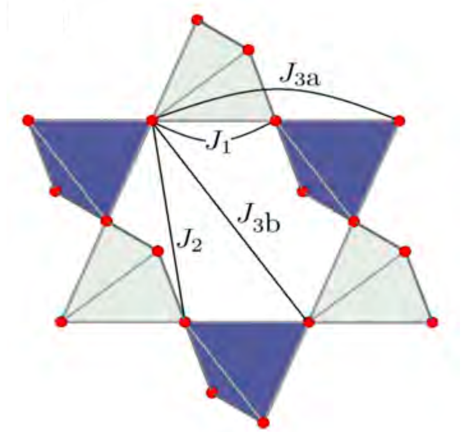

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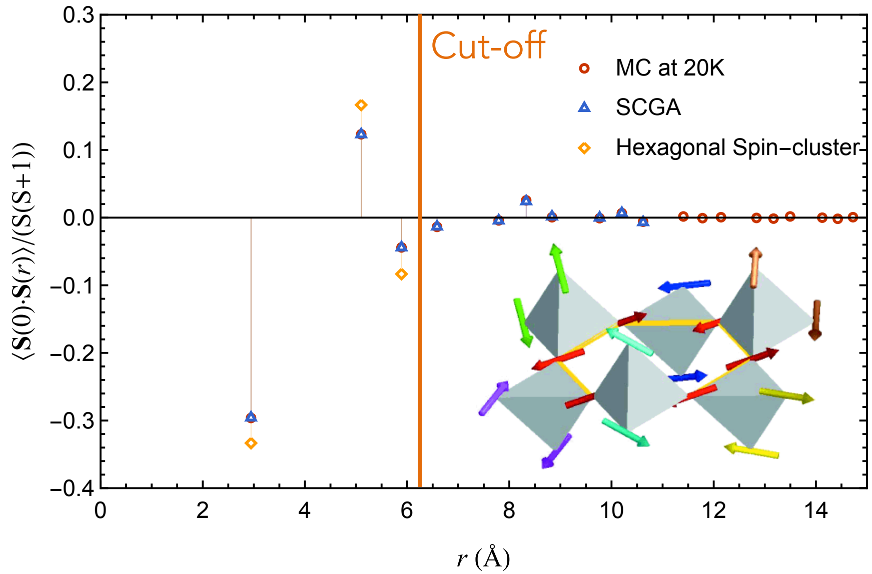

Hexagonal spin-cluster produces similar spin-correlations up to third neighbor

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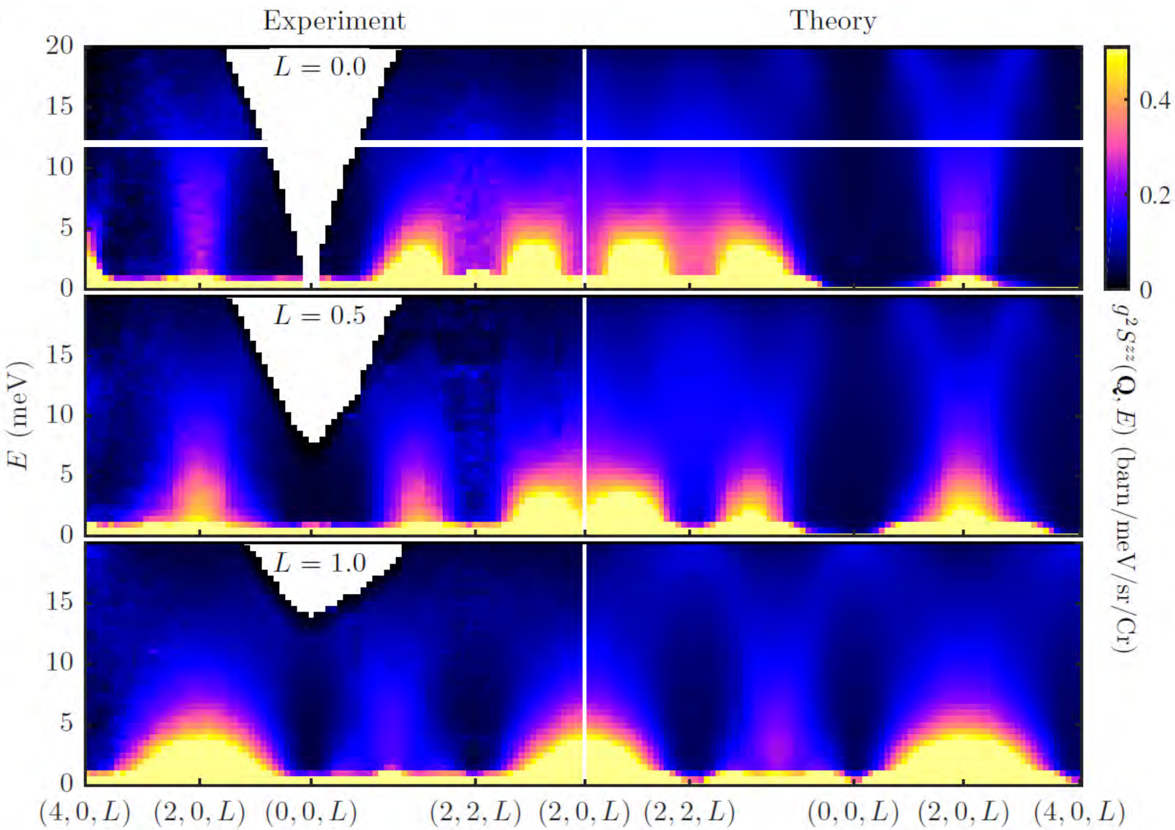
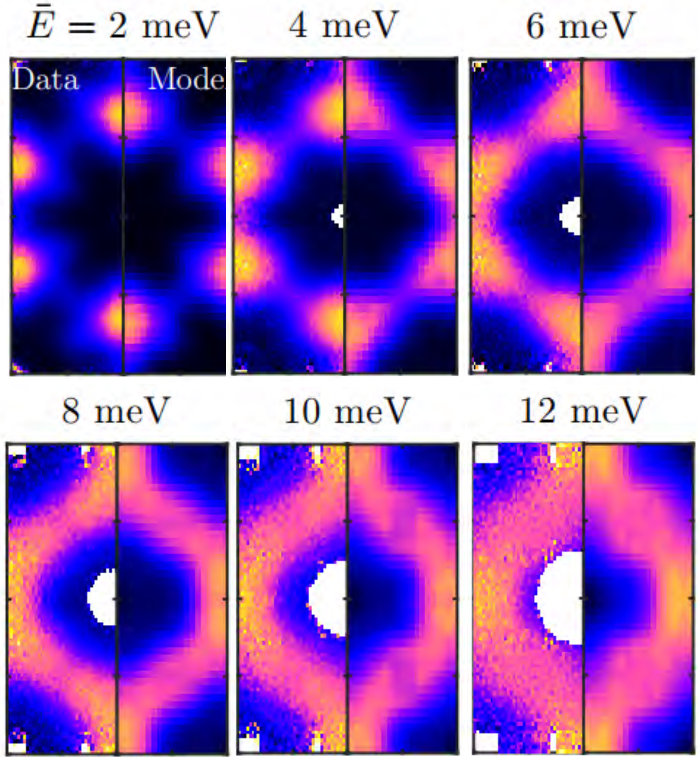
T = 20 K

Back to the energy-resolved data

- Can this FN Heisenberg model be used to model the excitations? Yes!





Our modeling strategy

Kapit, Chalker

Monte-Carlo cool down to 20 K for 6x6x6 super cell

Calculate harmonic fluctuations for resulting configuration

Discard small fraction of imaginary modes, average samples

See also

Molecular Dynamics vs Spin-Wave

Zhang, Changlani, Tchernyshyov, Moessner PRL **122**, 167203 (2019)

Back to the energy-resolved data

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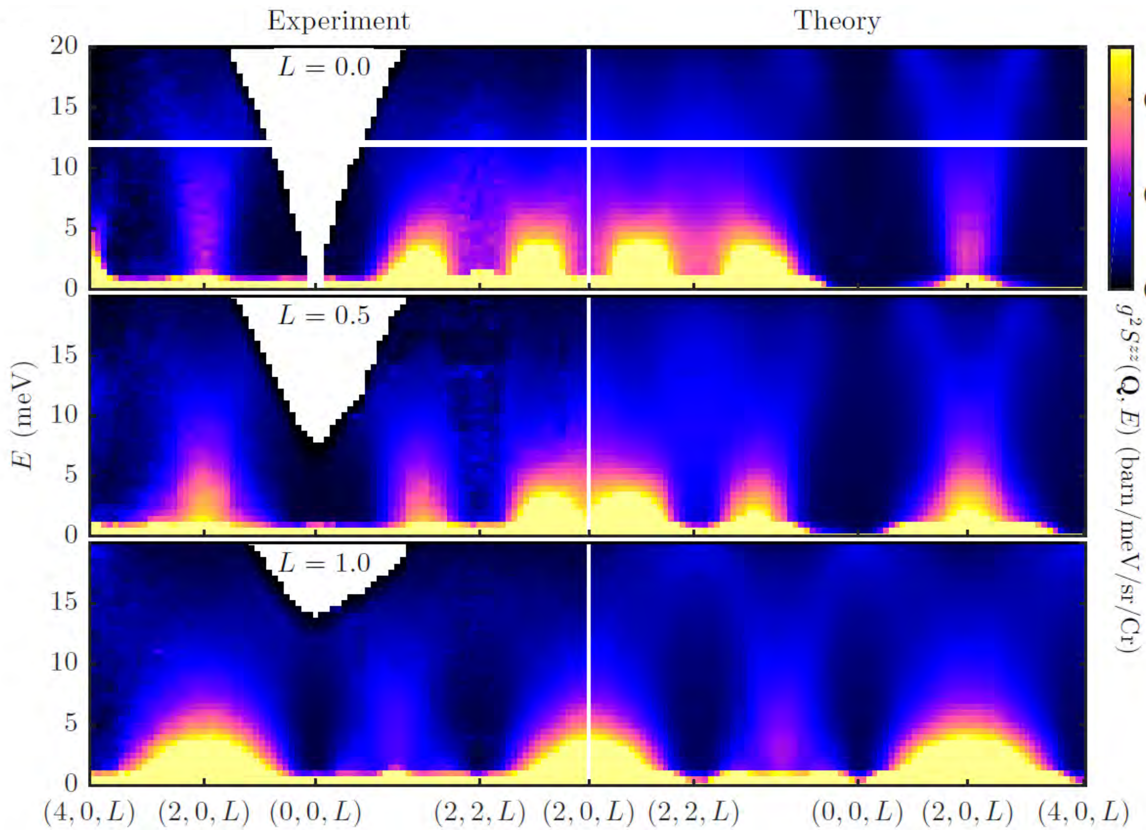
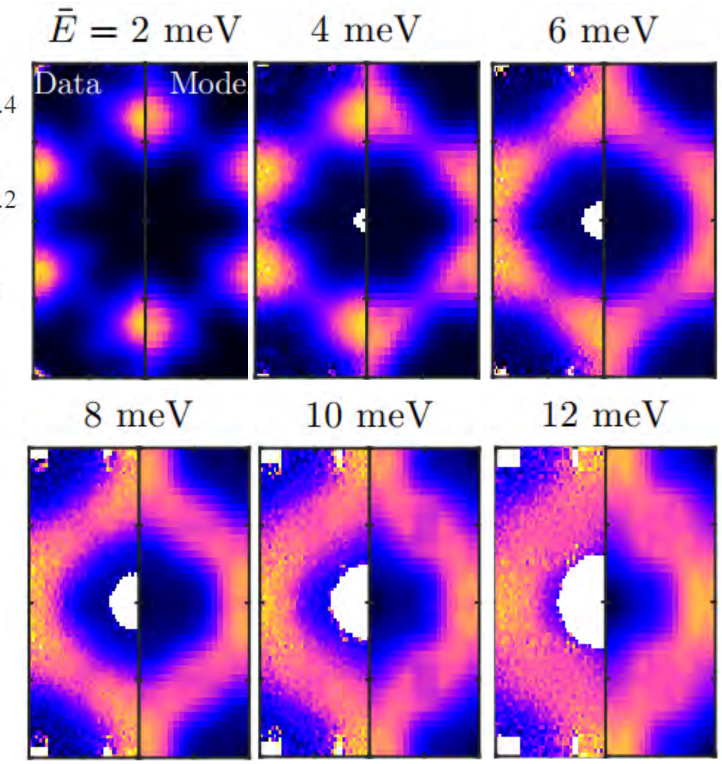




Dynamics is broad because spins ride a disordered background

To first order: not because of fractionalization, not because of magnon decay

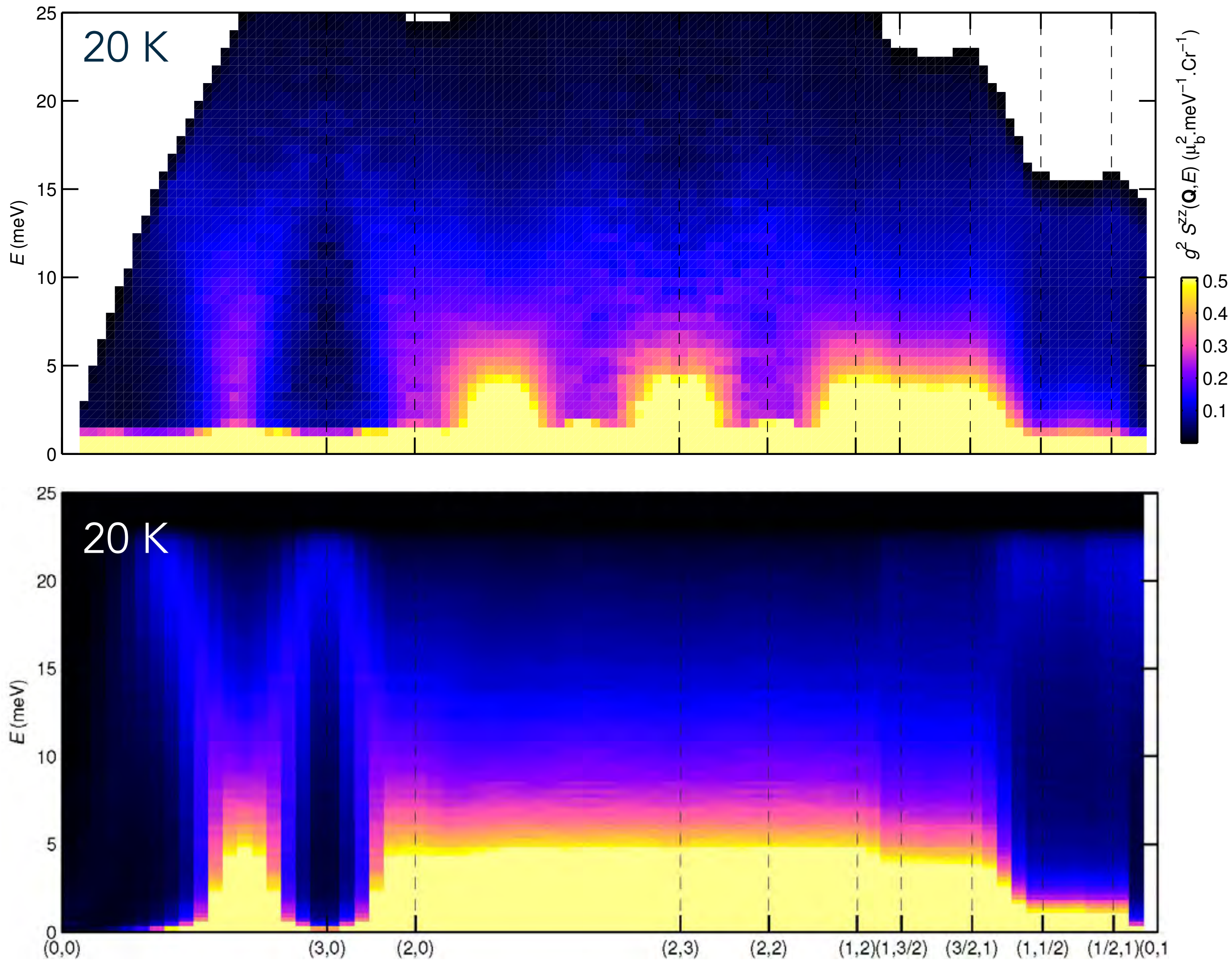
Time-scales of harmonic spin precessions and ground-state reconfigurations vastly differ

Final Remarks on $MgCr_2O_4$

- (1) Further neighbor interactions clearly modify the dynamics

Data

J_1 only

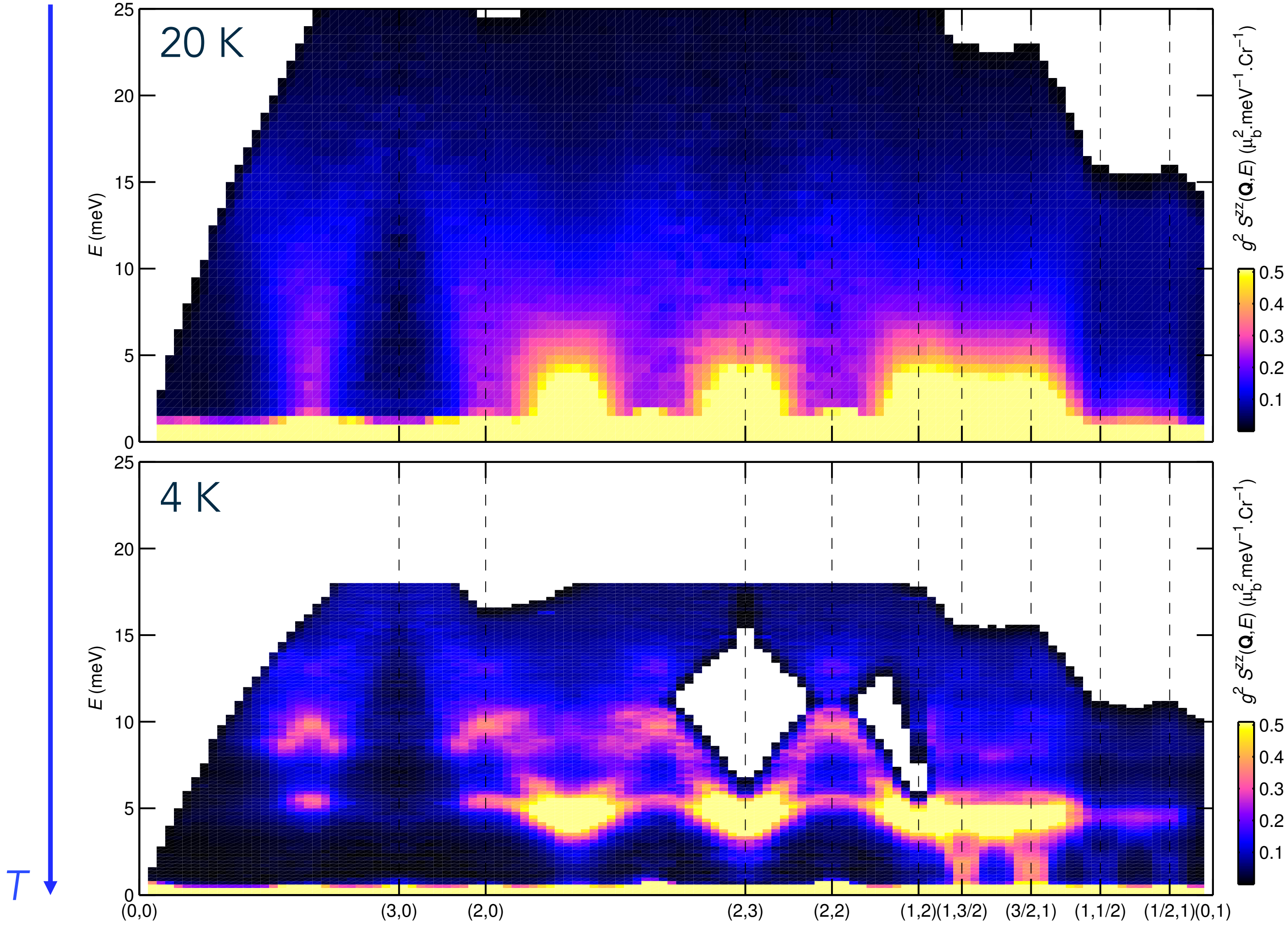


Clear mode *vs* wall of scattering

A continuum does not necessarily mean fractionalization

Final Remarks on $MgCr_2O_4$

- (2) Excitations become sharp below the magneto-structural transition



See also Gao PRB 2018

Final Remarks on $MgCr_2O_4$

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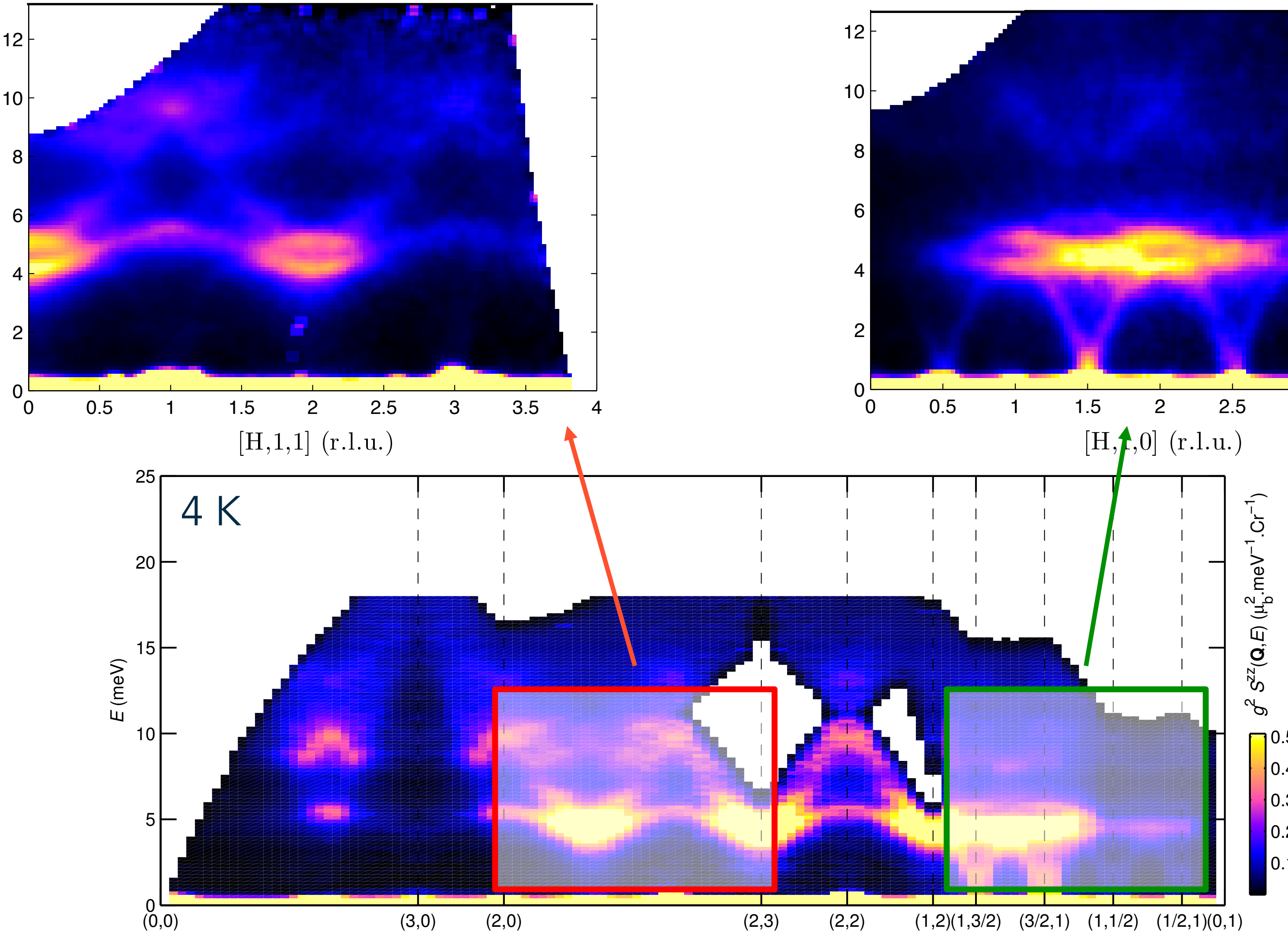


Conclusion $MgCr_2O_4$

- Loop scattering in chromium spinels: presence of further neighbor exchanges can explain all features of the paramagnetic data.

- Excitations are broad primarily because the spins ride a spatially disordered background in a strongly correlated spin configuration.

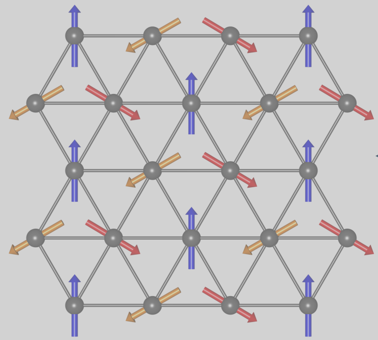
- Modeling works because in a classical spin-liquid there is a separation of time-scales between fast harmonic fluctuations and slow exploration of the ground-state manifold.

- Can we explain the structure and excitations below 13K?

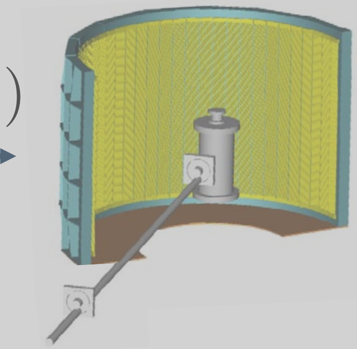
- Classical vs quantum spin-liquids: how does the spectrum evolves with lowering S?

Outline

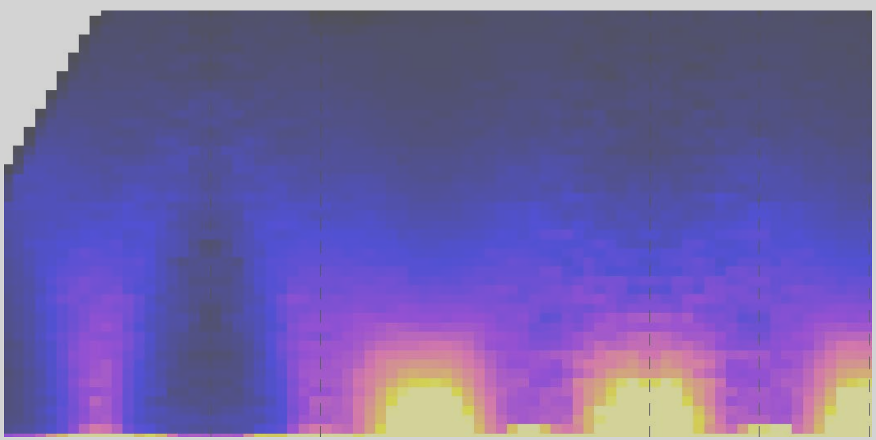
1. Introduction and neutron scattering warm-up



$S^{\alpha\beta}(\mathbf{Q},\omega)$

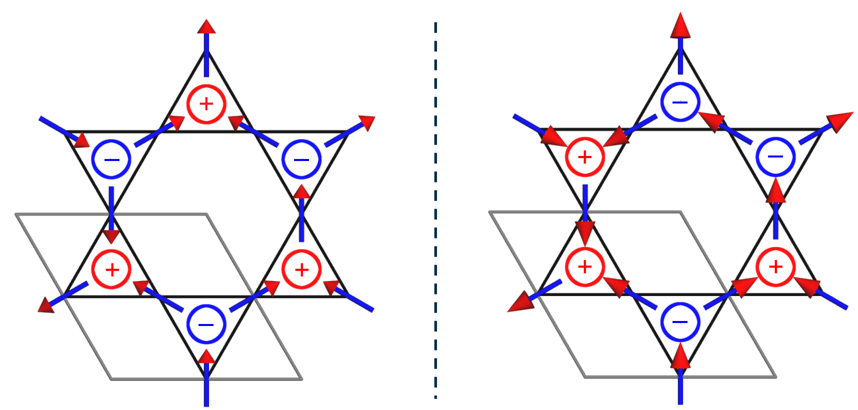


2. Nature of excitations in the classical pyrochlore Heisenberg AFM $MgCr_2O_4$



Bai *et al.* Phys. Rev. Lett. **122**, 097201 (2019)

3. Kagome spin-ice physics in the tripod compounds $Ln_3Mg_2Sb_3O_{14}$



Paddison *et al.*, Nat. Commun. **7**, 13842 (2016)

Dun *et al.*, arXiv:1806.04081 (2019) + in prep.

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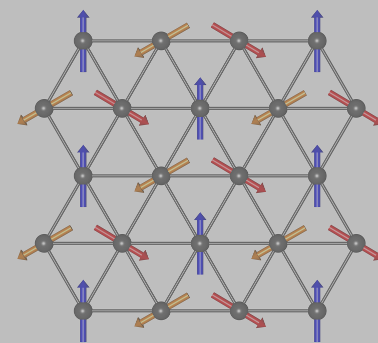

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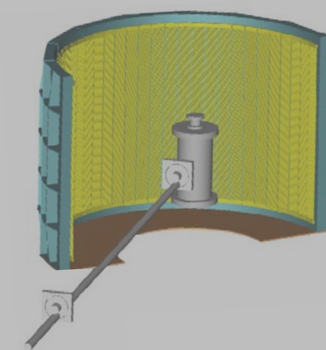
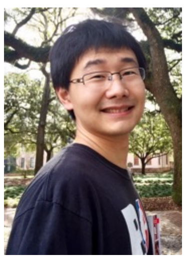
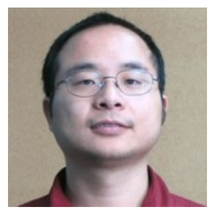




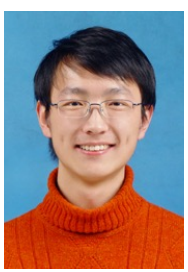
Zhiling Dun

Joe Paddison

Siân Dutton



Haidong Zhou



Xiaojian Bai

Emily Hollingworth

Claudio Castelnovo

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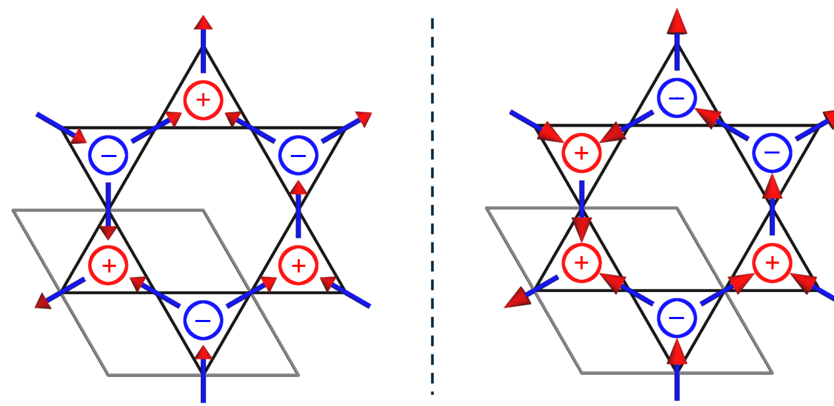

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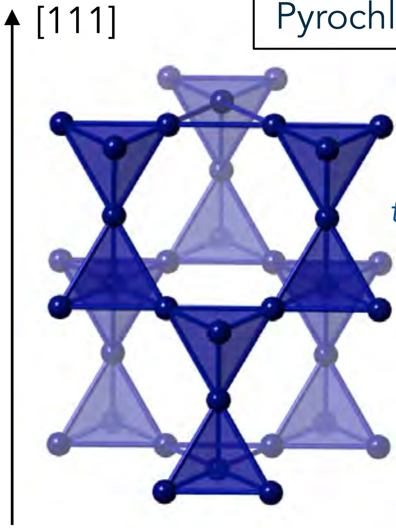
Dun *et al.*, arXiv:1806.04081 (2019) + in prep.

Matt Stone (ORNL), Clarina de la Cruz (ORNL), Matt Tucker (ORNL/ISIS), Franz Demmel (ISIS), Nick Butch (NIST)

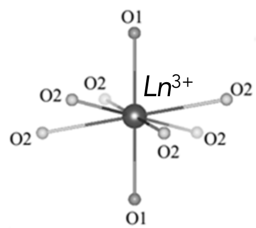
From pyrochlore spin-ice to kagome spin-ice

- ❑ Discovery of “tripod” systems:

Pyrochlore $Ln_2Ti_2O_7$

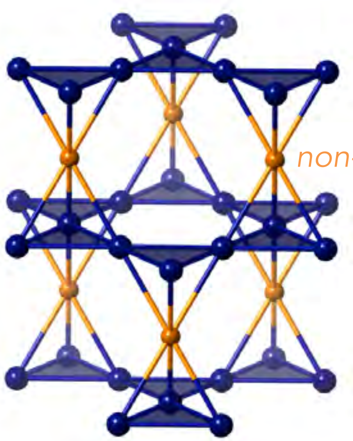


Space Group *Fd–3m*

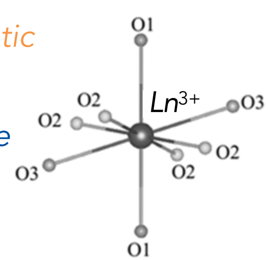


Point Group D_{3d}

Tripod Kagome $Ln_3Mg_2Sb_3O_{14}$



Space Group *R-3m*



Point Group C_{2h}

site mixing possible, only powders

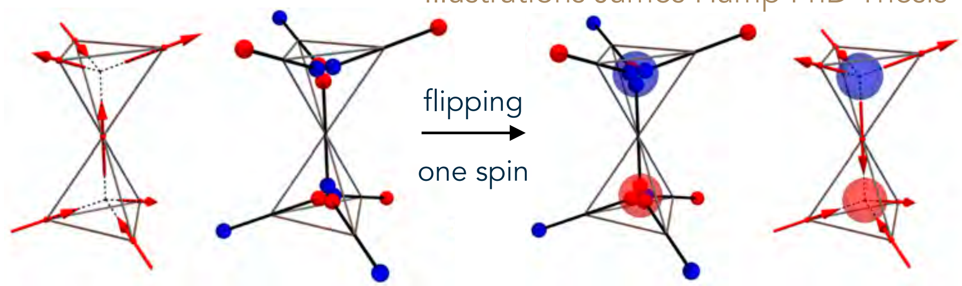
Dun *et al.*, *PRL* **116**, 157201 (2016); *PRB* **95**, 104439 (2017).
Paddison, Dutton *et al.*, *Nat. Comm.* **7**, 13842 (2016)

- ❑ Spin-ice anisotropy is maintained

Dy Ho

- ❑ Magnetic charges and monopoles

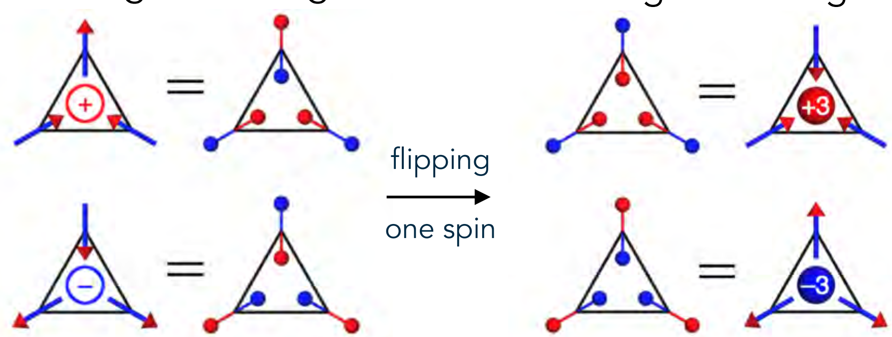
Illustrations James Hamp PhD Thesis



Monopole pairs interact via Coulomb’s law

+/-1 magnetic charges

+/-3 magnetic charges



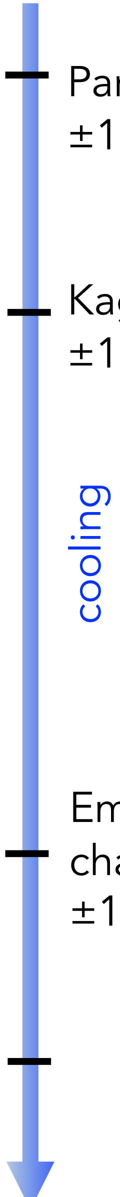
System is dense in interacting magnetic charges

Castelnovo, Moessner, and Sondhi, *Nature* **451**, 42 (2008)
Moller & Moessner, PRB **80**, 140409 (2009)

The canonical behavior of $Dy_3Mg_2Sb_3O_{14}$

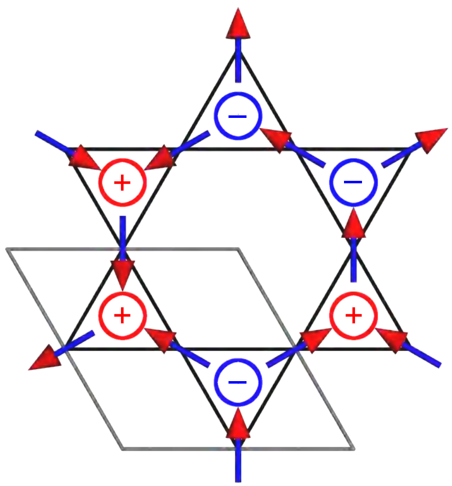
- Predictions from classical MC:

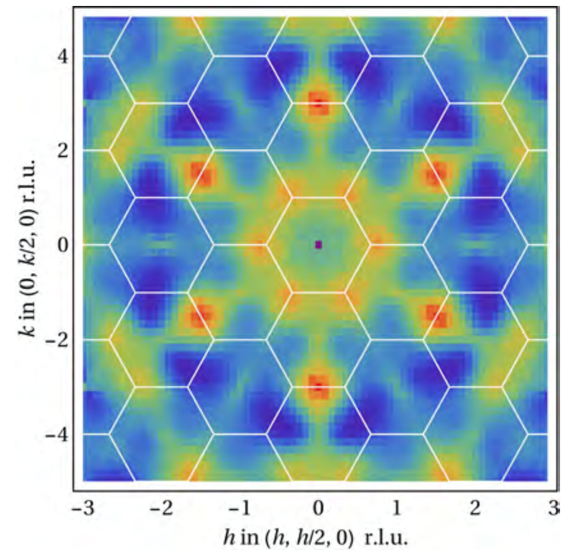
spins in plane, no disorder



Paramagnet
±1 or ±3

Kagome spin ice
±1





Emergent-
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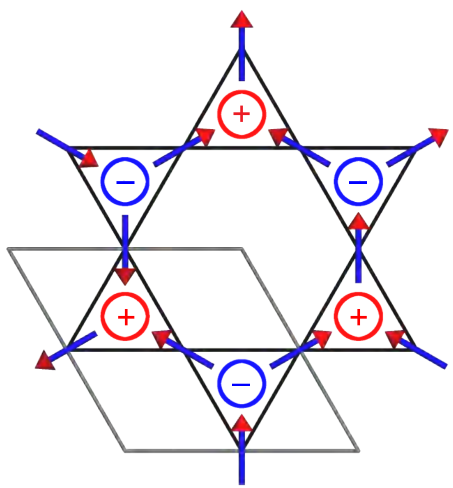
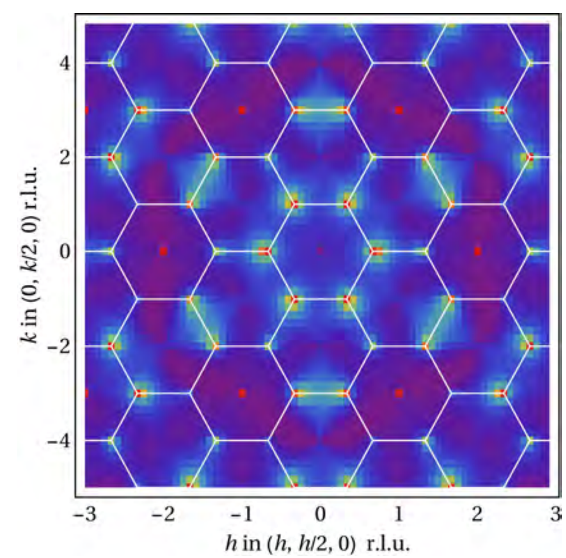
±1





Chern, Mellado & Tchernyshyov, *PRL* 106, 207202 (2011)

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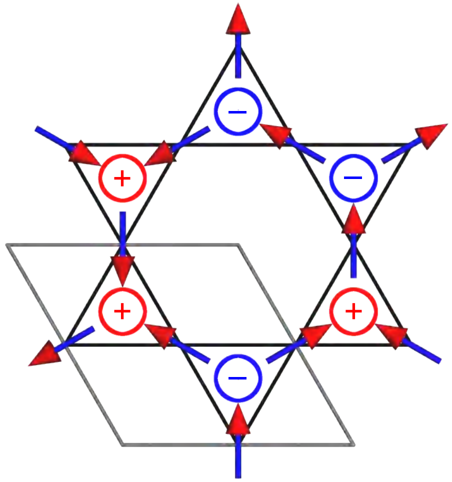
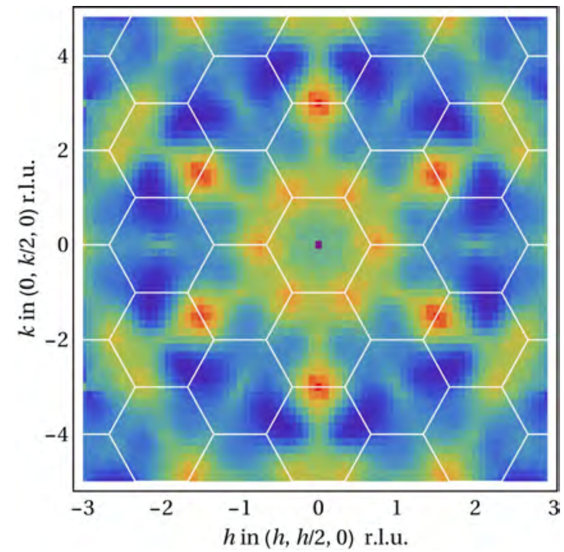
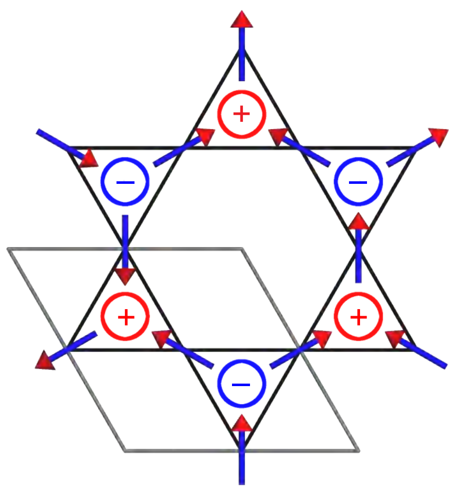
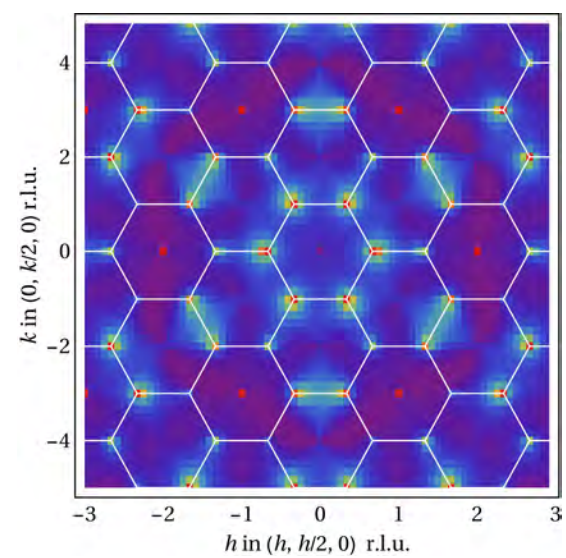
- Emergent-charge order ±1
- Spin order

cooling









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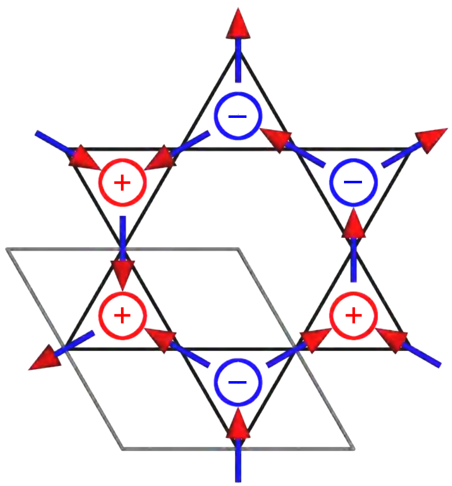

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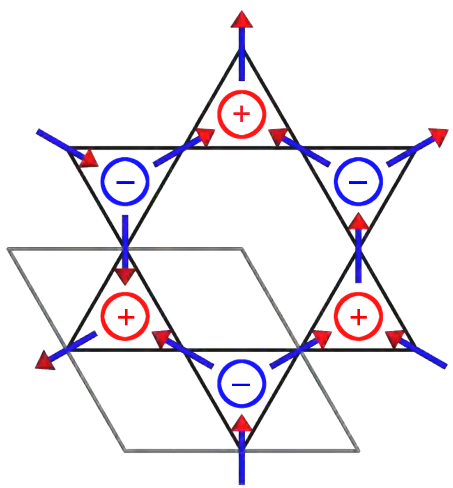
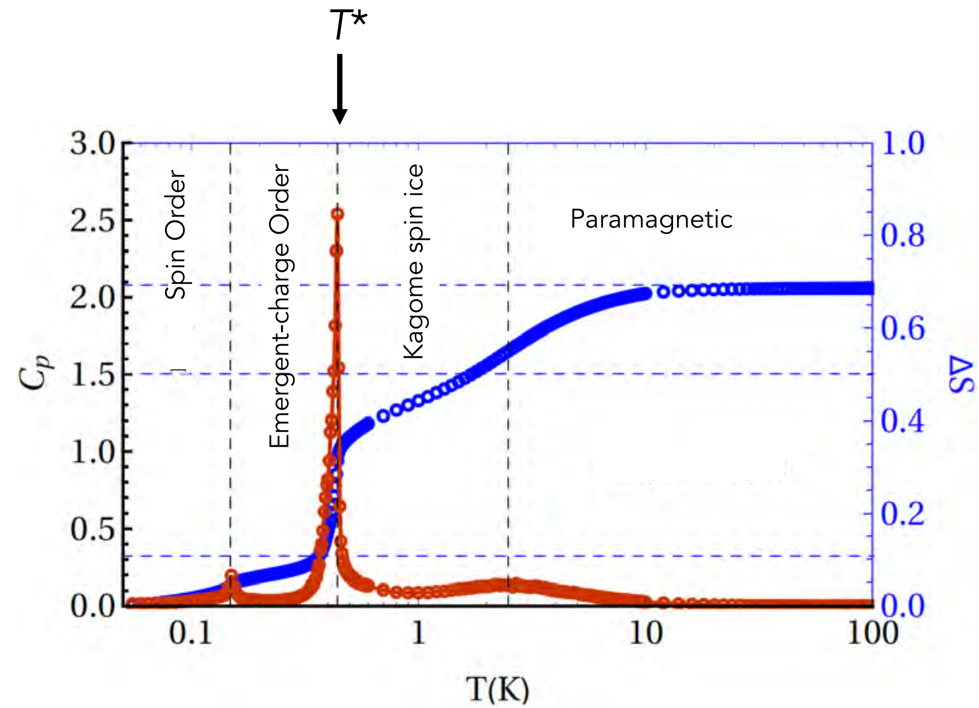


Spin order



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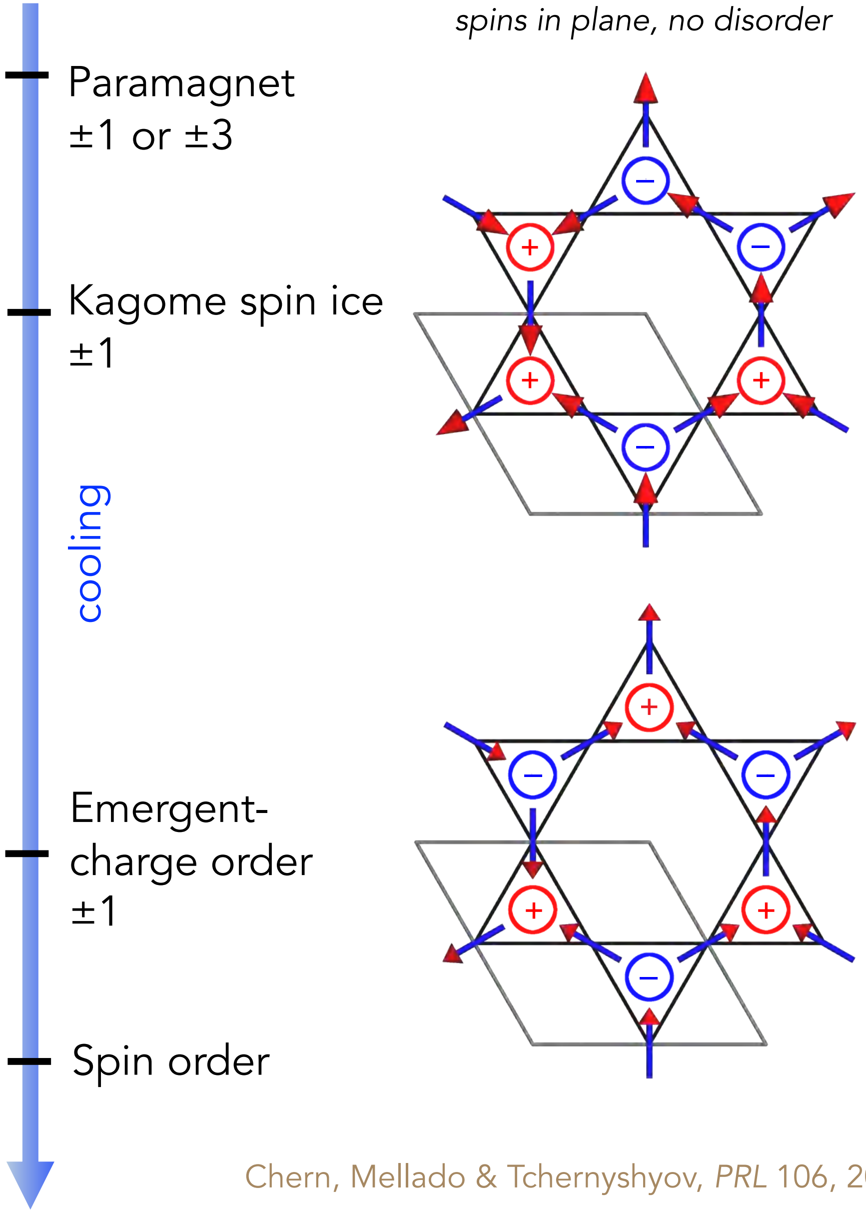
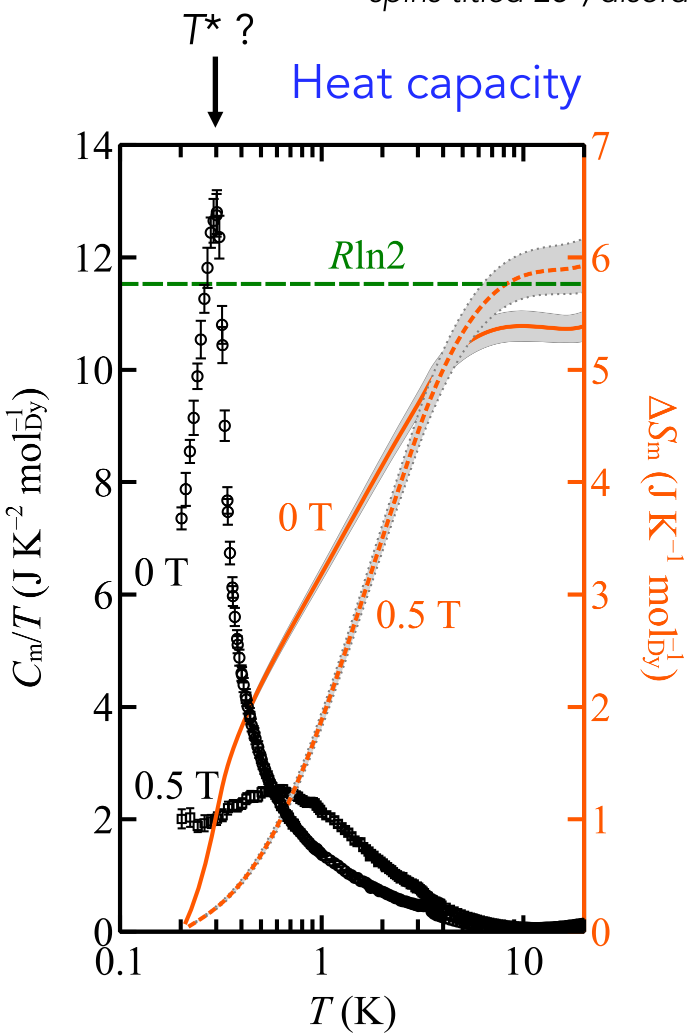


❑ Experiments on powder samples:

spins titled 26°, disorder 6%



Chern, Mellado & Tchernyshyov, *PRL* 106, 207202 (2011) Paddison *et al.*, *Nature Commun.* **7**, 13842 (2016)

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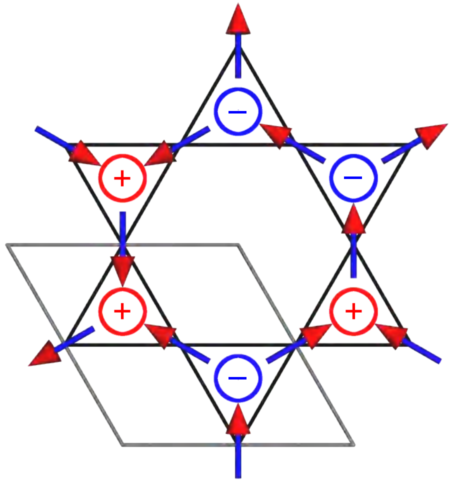

cooling

Emergent-
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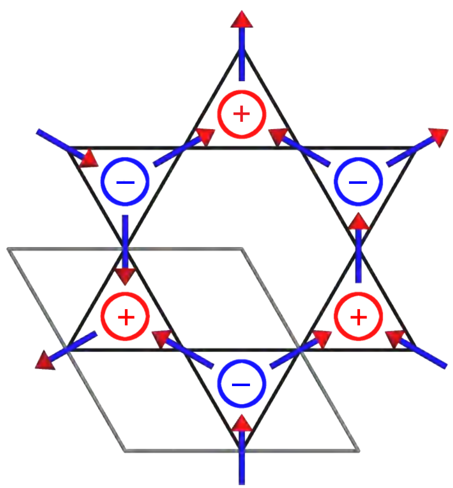

Spin order

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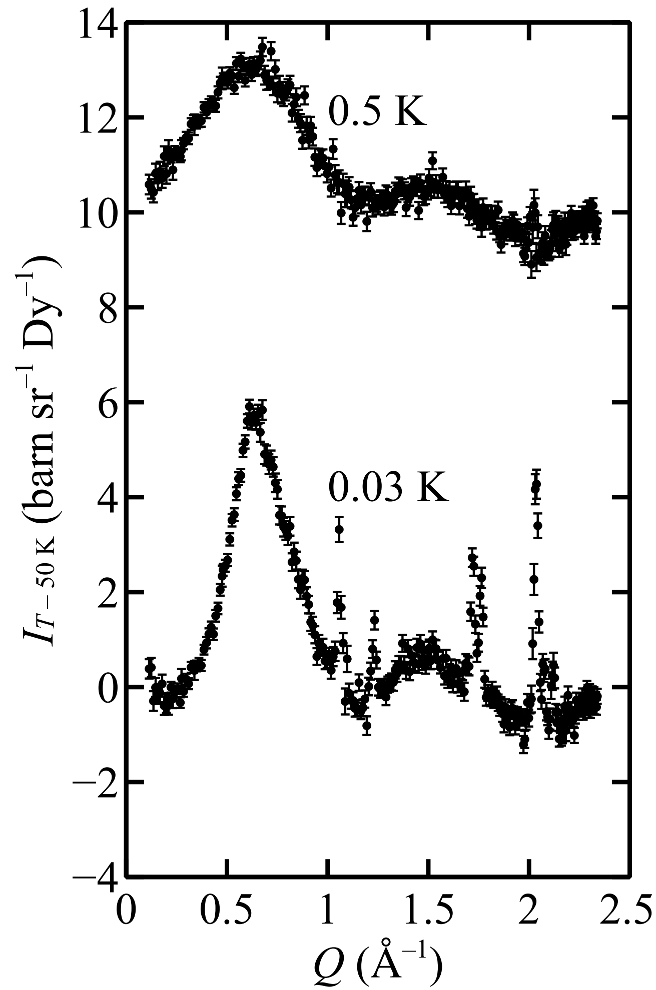
Magnetic neutron scattering



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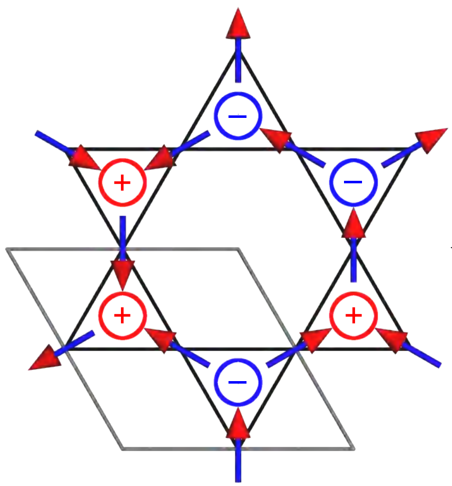

cooling

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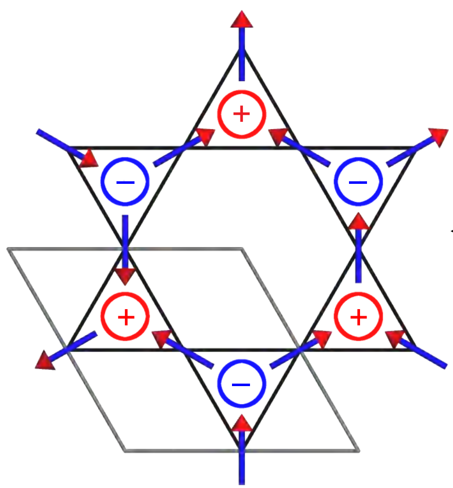

Spin order

?

?

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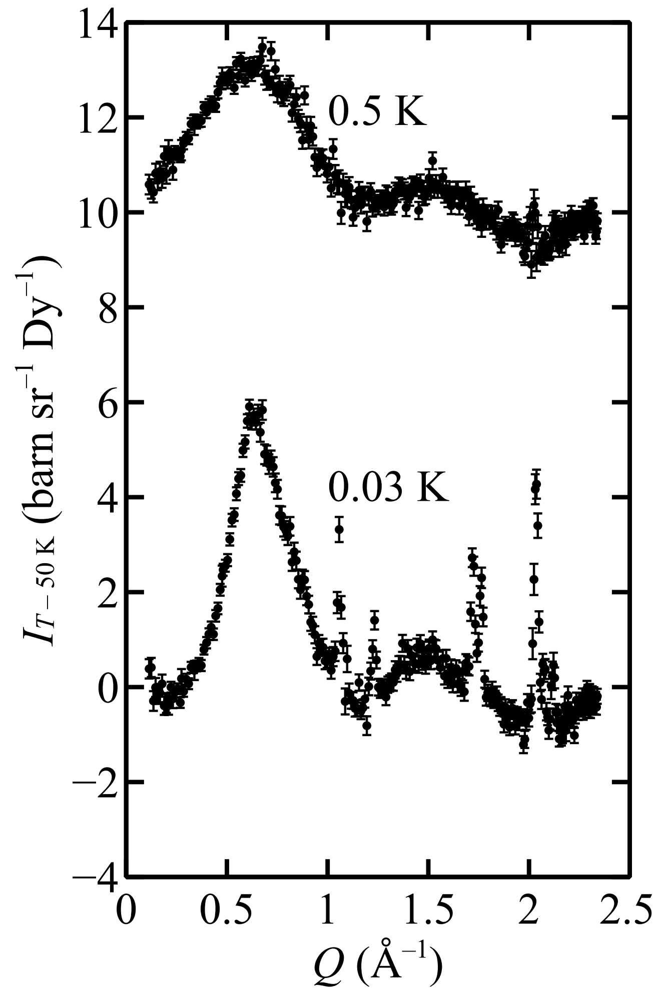

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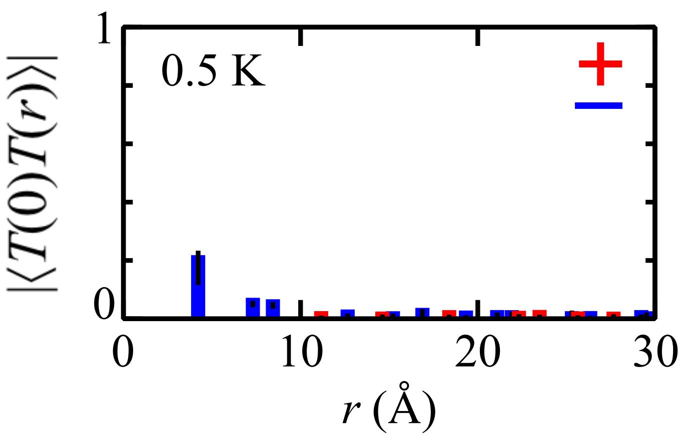
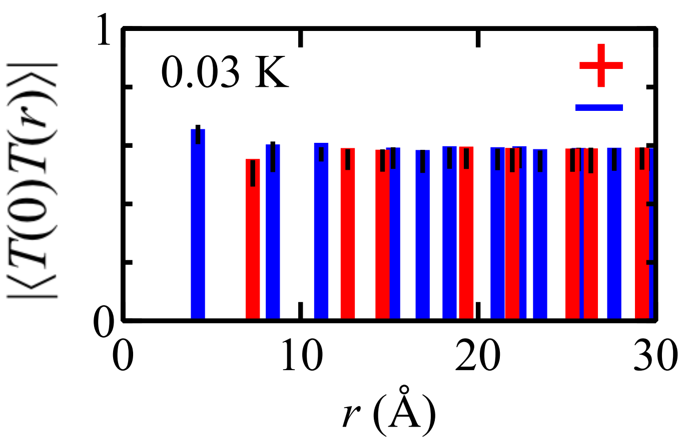
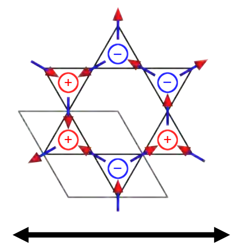
The canonical behavior of $Dy_3Mg_2Sb_3O_{14}$

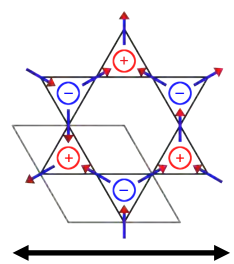
❑ Reverse Monte Carlo fits:

Charge-charge correlations









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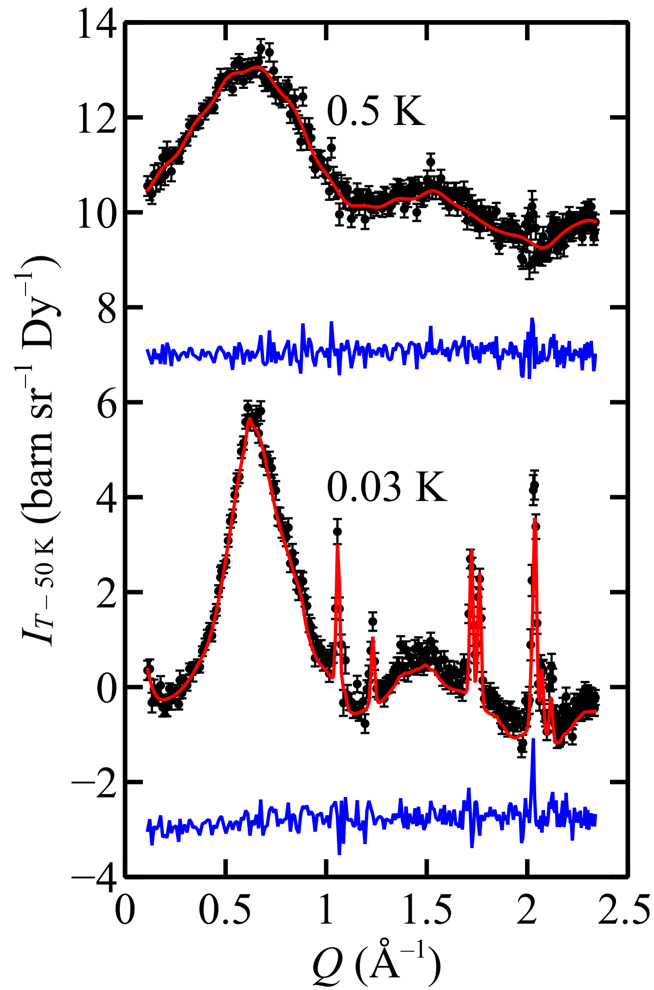
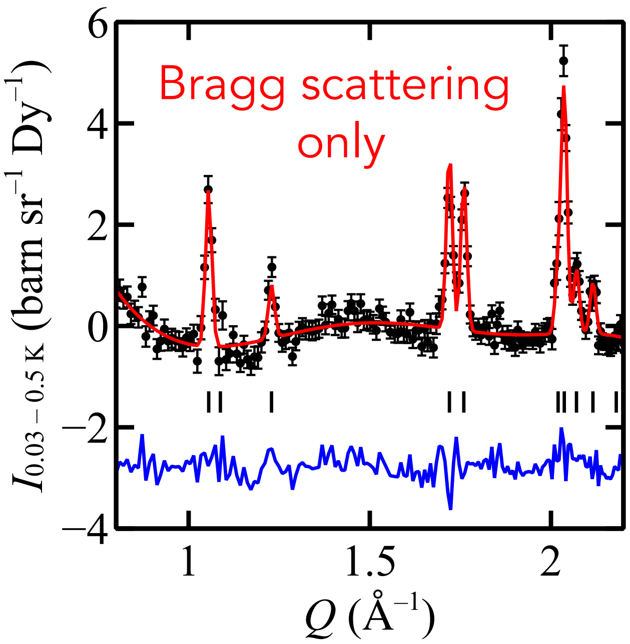
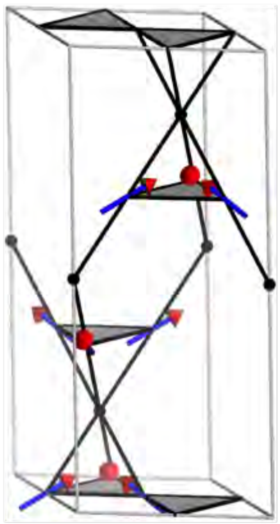


Blech & Averbach, *Physics* **1**, 31 (1964)
Paddison & Goodwin, *PRL* **108**, 017204 (2012)
Paddison *et al.*, *Nature Commun.* **7**, 13842 (2016)

The canonical behavior of $Dy_3Mg_2Sb_3O_{14}$

- Coexistence of Bragg and diffuse scattering



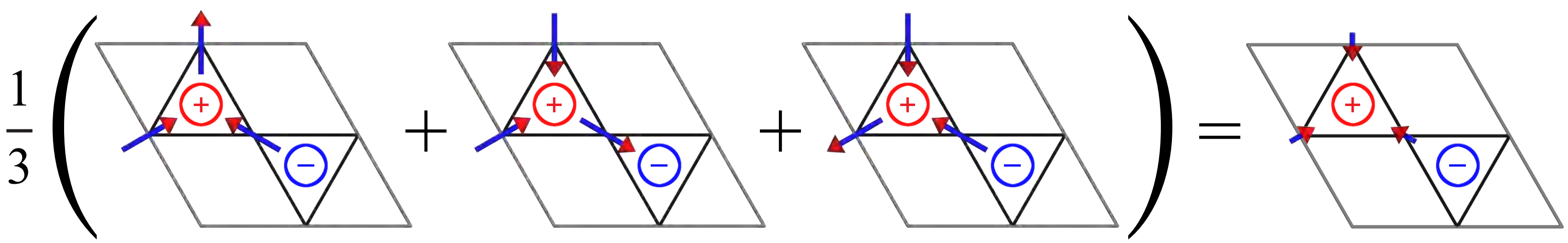


k = 0 average magnetic structure

All-In All-Out (AIAO) state with 2.8 μ_B

Only ≃1/3 of the total moment is found in the Bragg peaks

- "Spin fragmentation"



Brooks-Bartlett *et al.*, *PRX* **4**, 011007 (2014)
Canals *et al.*, *Nature Commun.* **7**, 11446 (2016)
Petit *et al.*, *Nature Physics* **12** 746-750 (2016)

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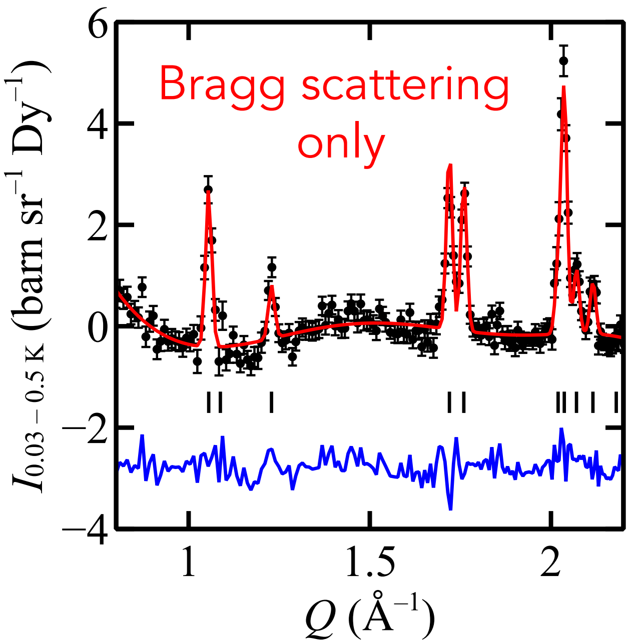

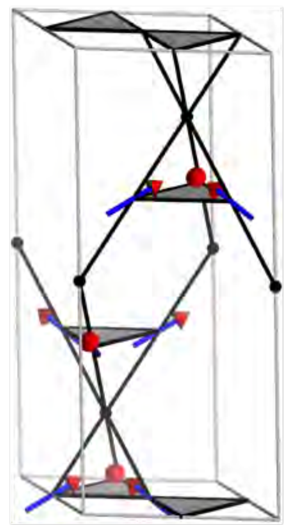

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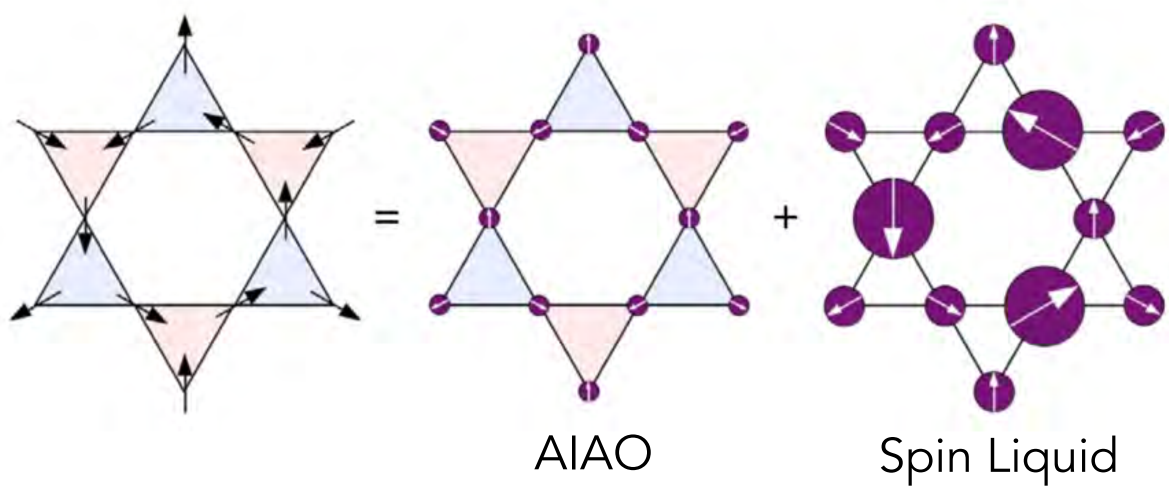

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Thank you for your attention!