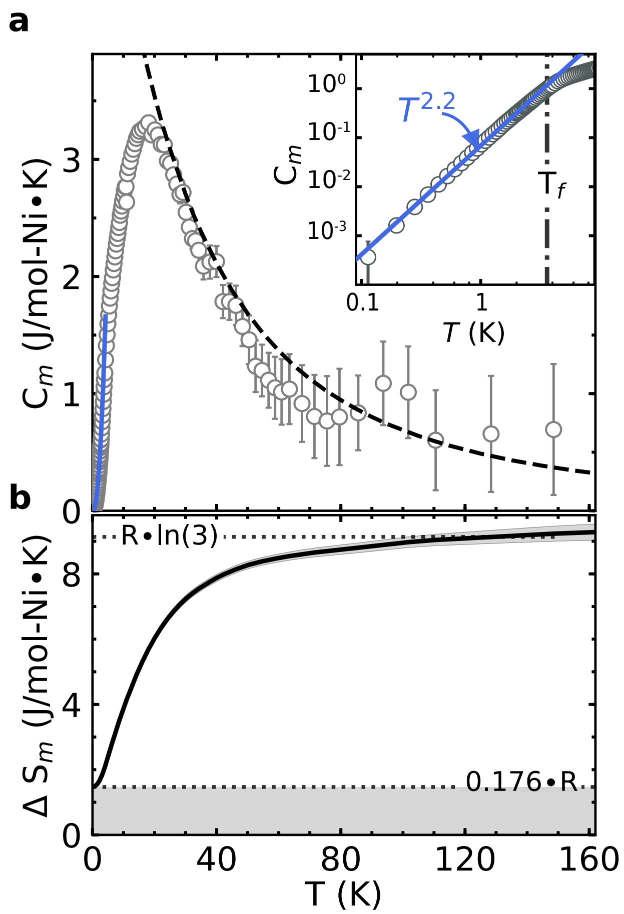
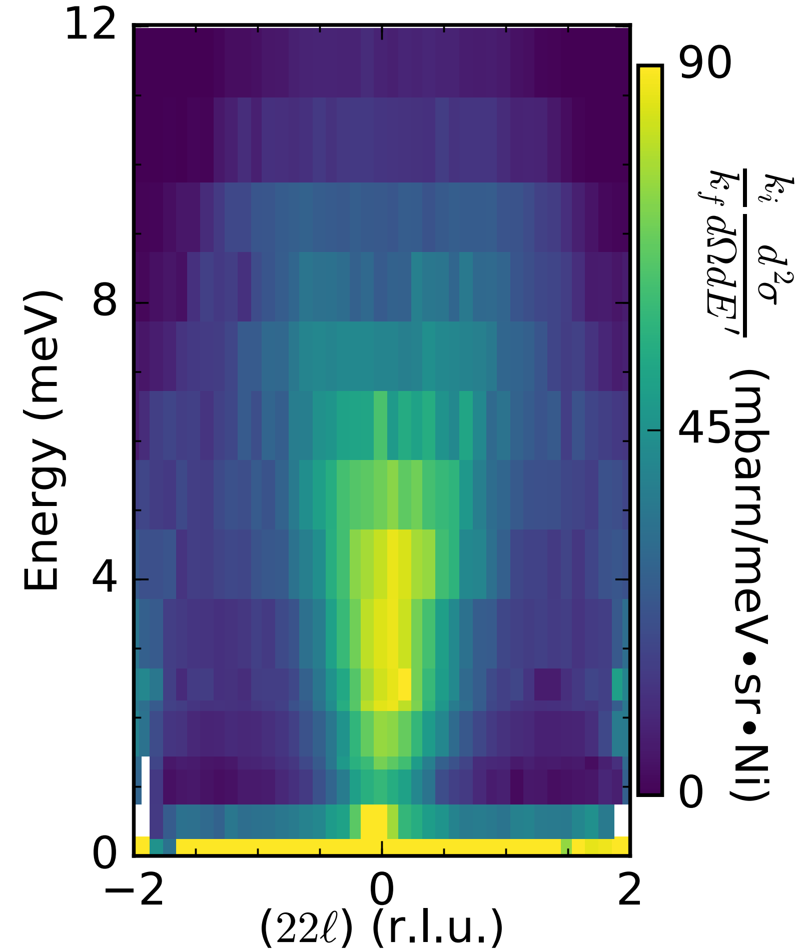
Continuum of Magnetic Excitations in $NaCaNi_2F_7$

Kemp Plumb

HFM, July 2018





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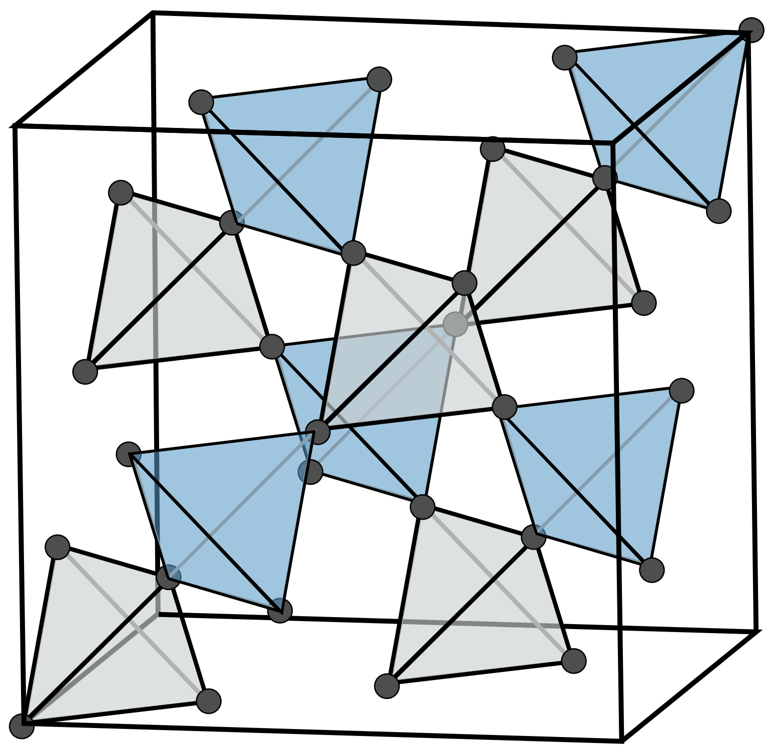
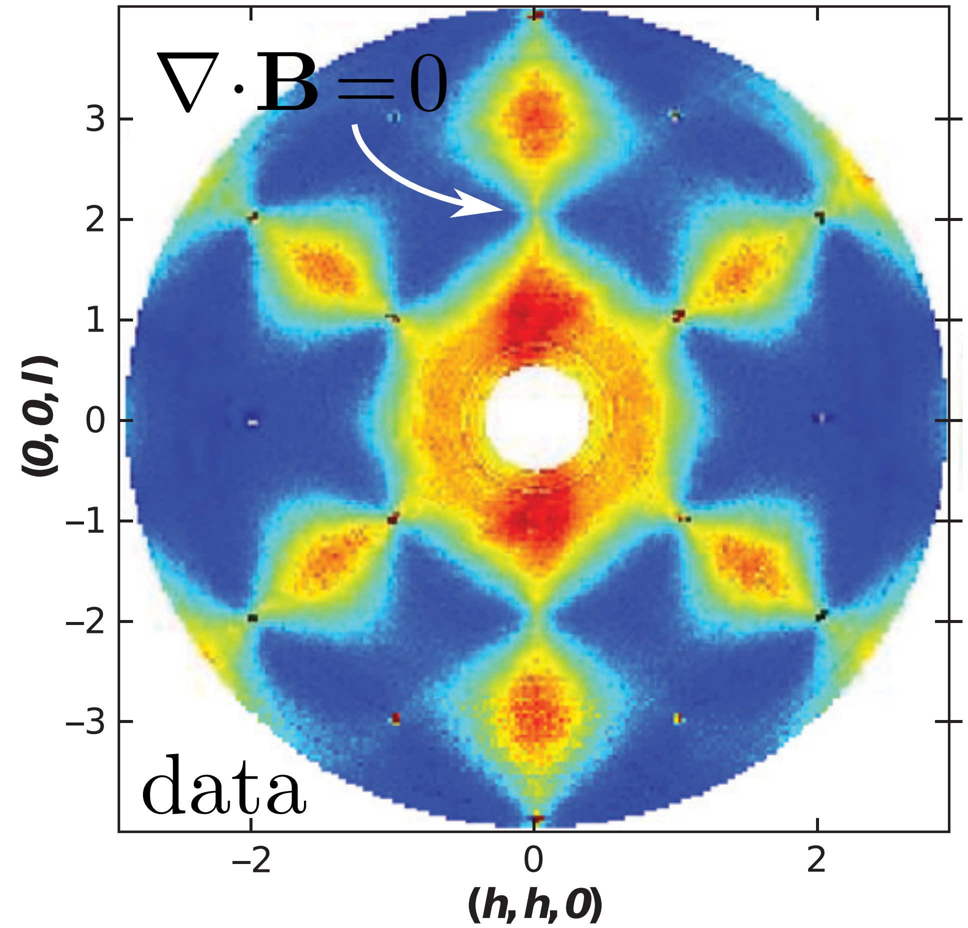
The pyrochlore lattice

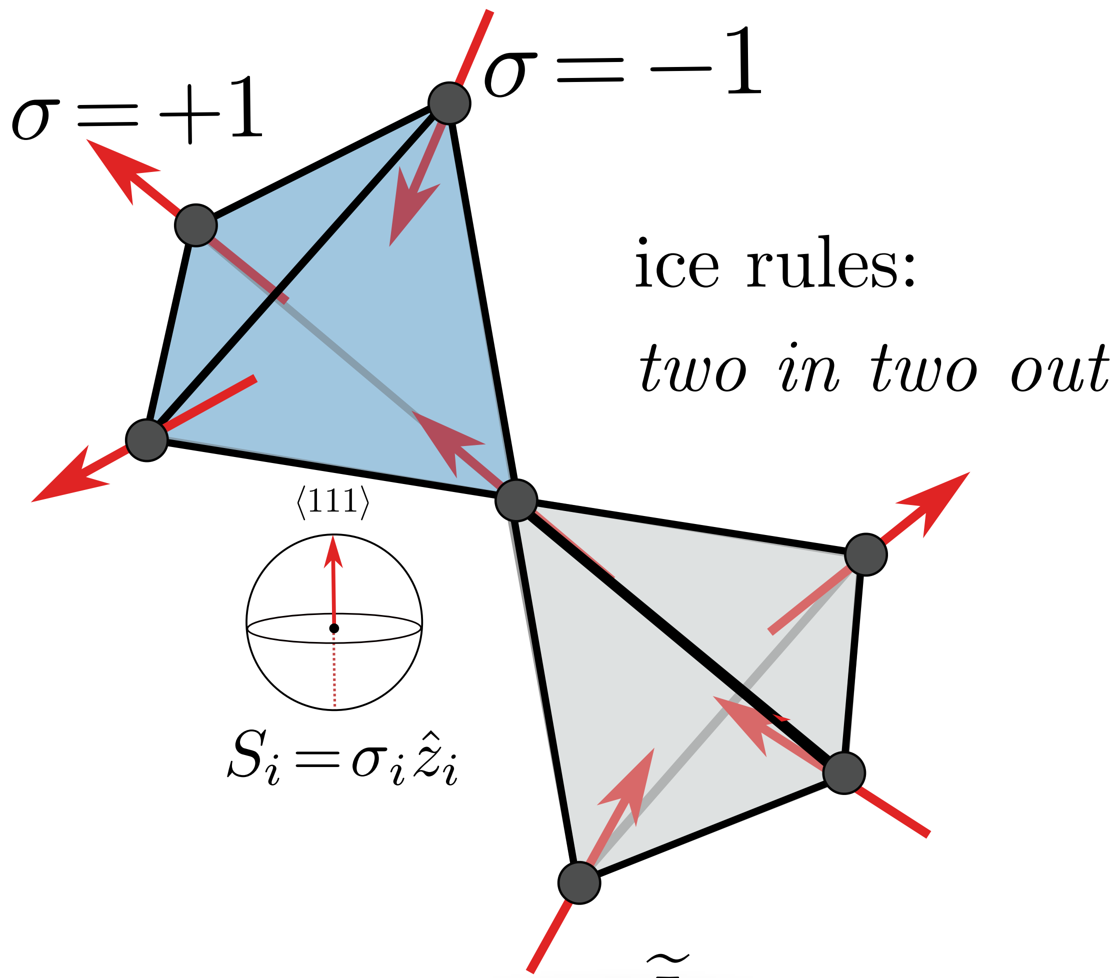


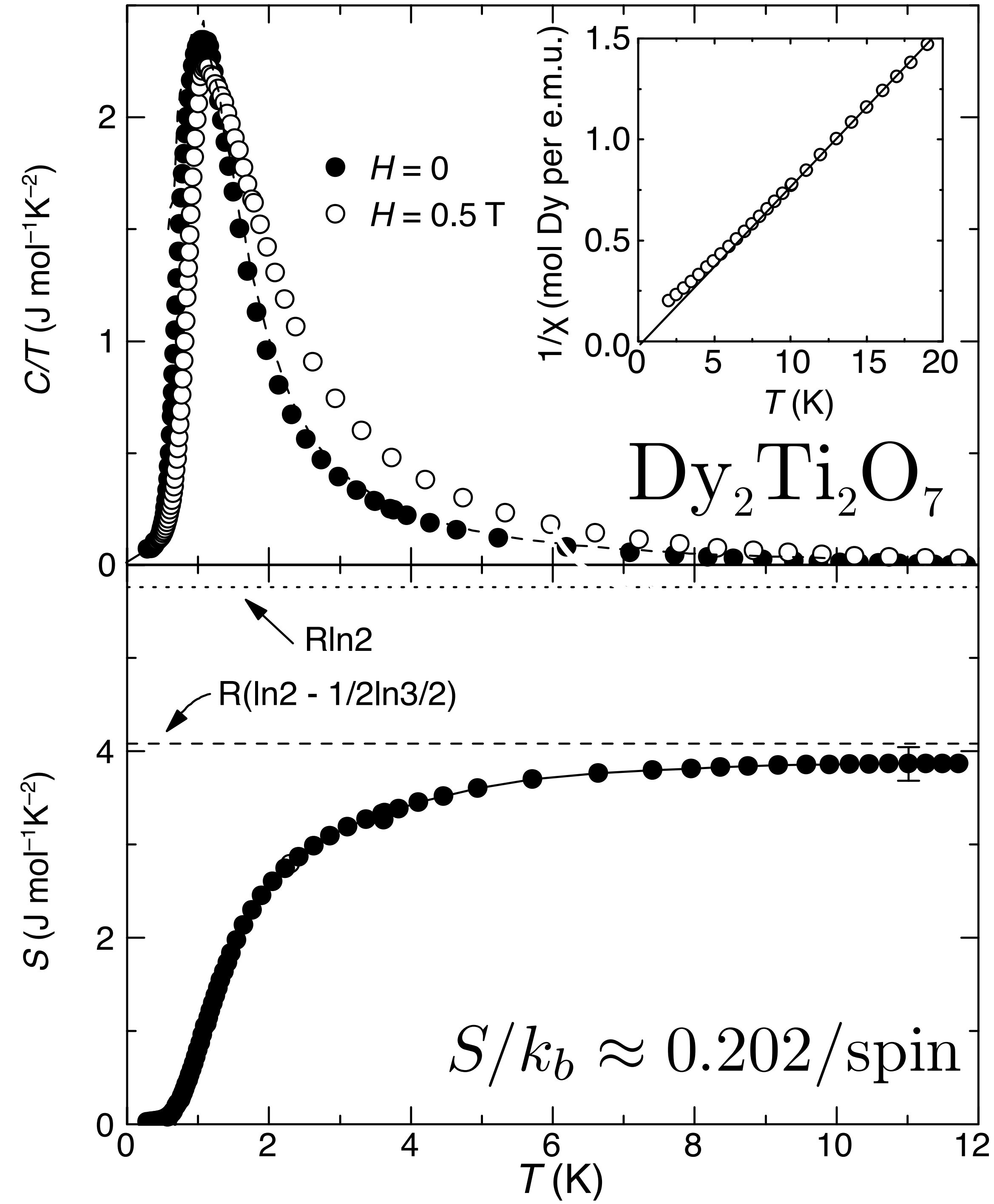


- A paradigm for magnetic frustration: degenerate low energy states subject to local constraints, leads to ....
  - Spin ice: Ising ferromagnet.
  - Order-by-disorder: xy models.
  - Spin liquid: Heisenberg antiferromagnet. Quantum?

†Fennel *et al.* Science (2009)

Coulomb Phase: Spin-ice





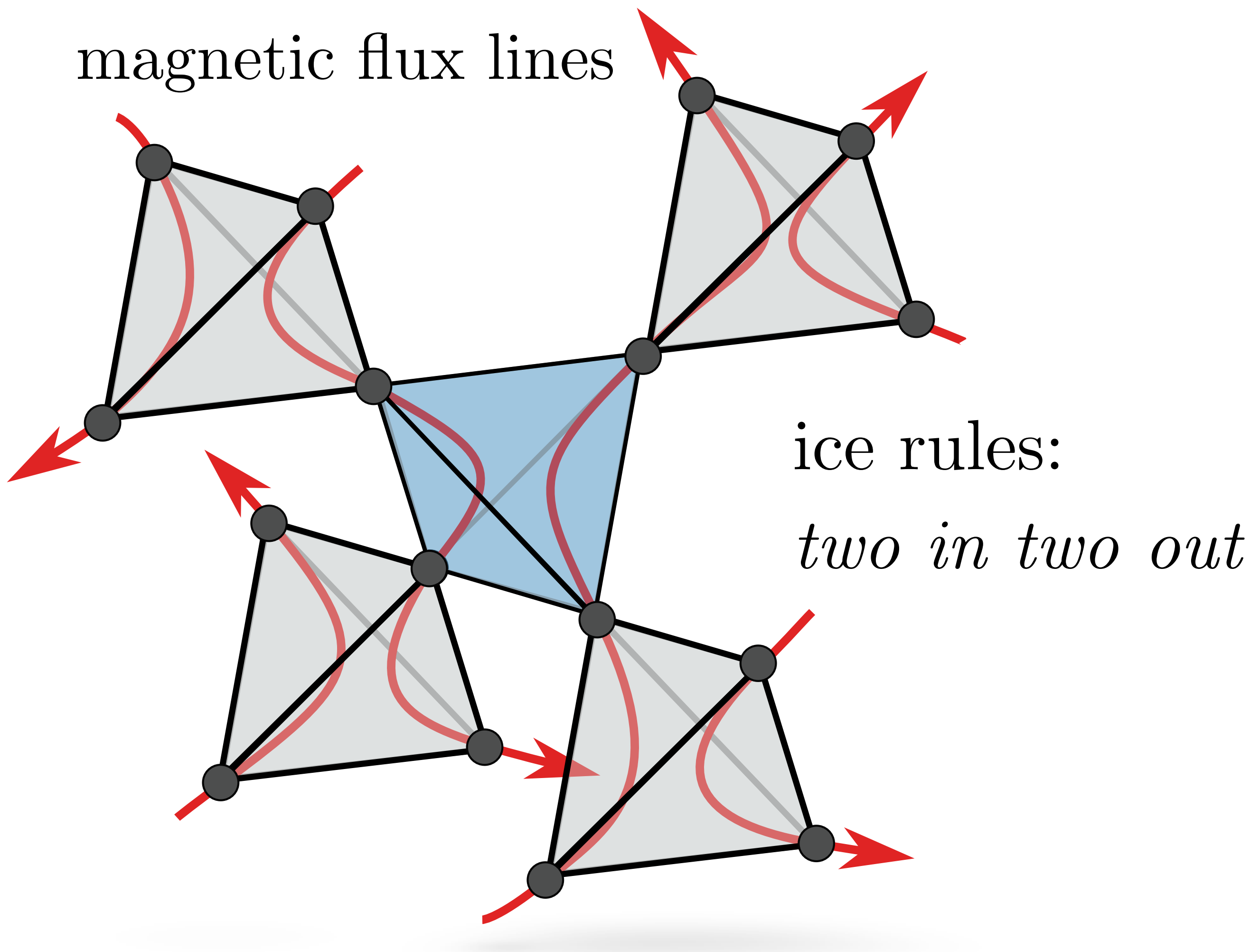
$$H = \widetilde{J} \sum_{\langle ij \rangle} \sigma_i \sigma_j = \frac{\widetilde{J}}{2} \sum_{\triangle} \sigma_{\triangle}^2 + \text{cnst.}$$

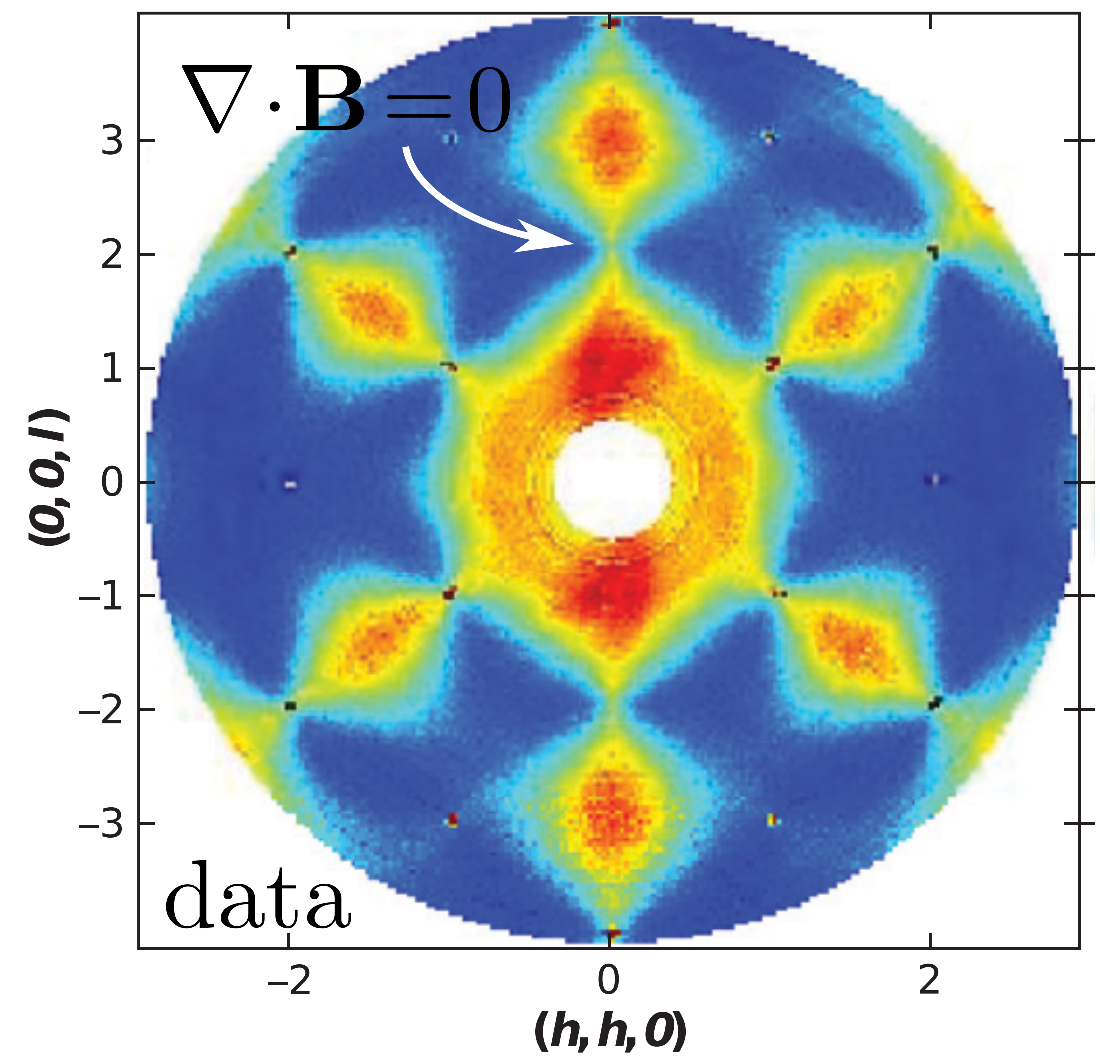
$$\sigma_{\triangle} = \sum_{i \in \triangle} \sigma_i$$

- Lowest energy states have: $\sigma_{\triangle} = 0 \rightarrow$ ice rules.
- Macroscopic degeneracy, $W = (3/2)^{(N/2)}$ states with $\sigma_{\triangle} = 0$.

†Ramirez *et al.* Nature (1999)

Coulomb Phase: Spin-ice

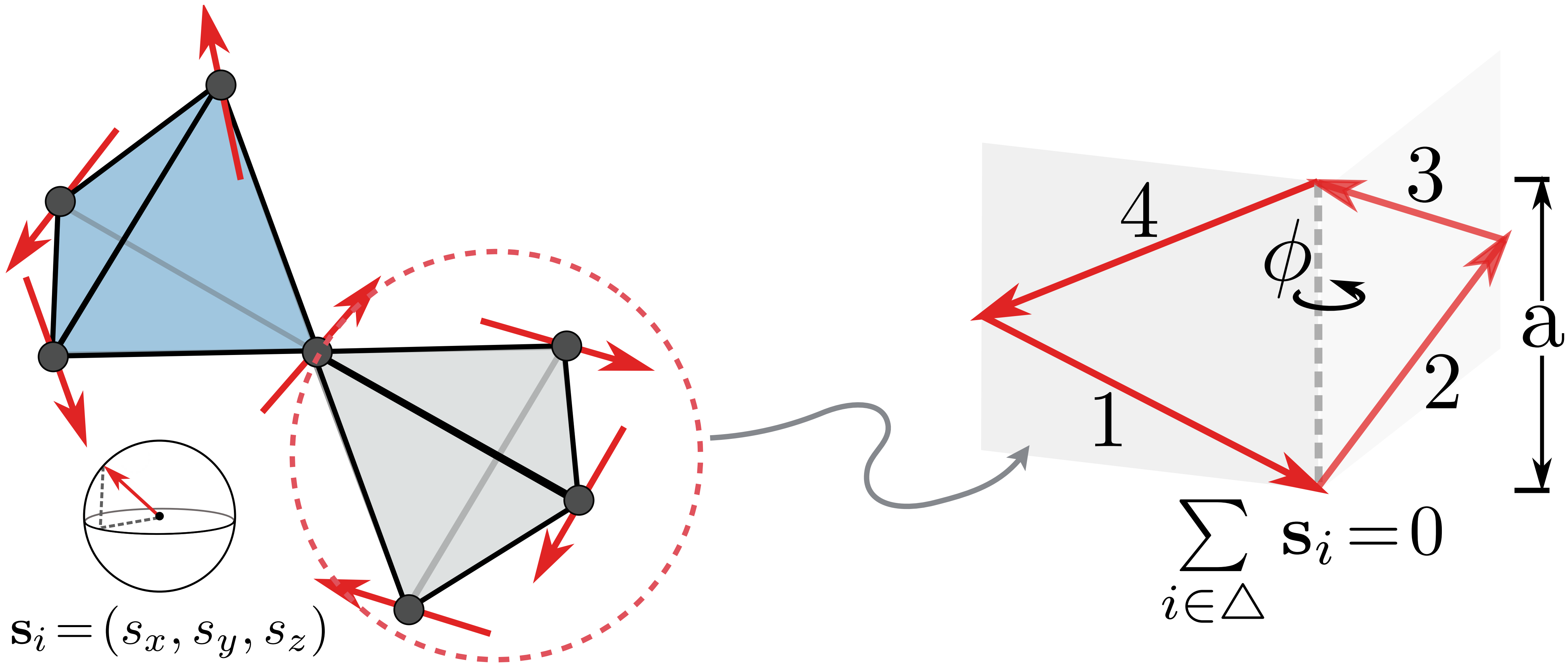




- Highly degenerate, like a liquid, but correlated.
- Imagine spins defining flux lines, then $\nabla\cdot\mathbf{B}=0$.
- Consequence is power law correlations: $\langle\mathbf{B}(0)\mathbf{B}(\mathbf{r})\rangle \propto \dfrac{1}{r^3}$.
- Spins are frozen in time.

†Fennel *et al.* Science (2009)

Coulomb phase: continuous spins



$$H = J \sum_{\langle ij \rangle} \mathbf{s_i} \cdot \mathbf{s_j} = \frac{J}{2} \sum_{\triangle} |\mathbf{L}_\triangle|^2 + \text{cnst.} \qquad L_\triangle = \sum_{i \in \triangle} \mathbf{s}_i$$

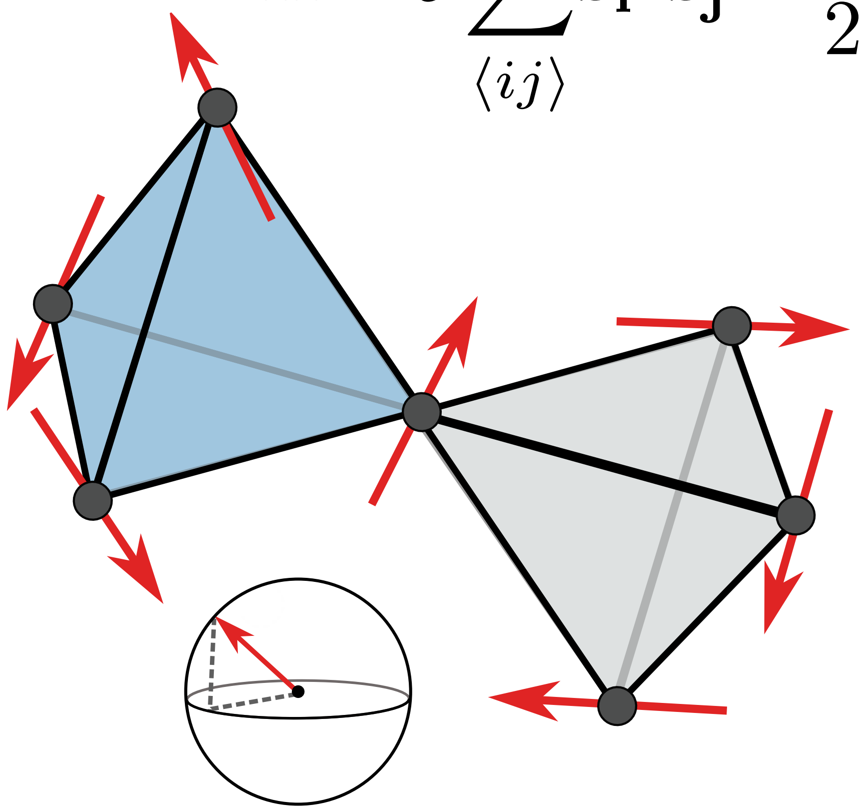
- In lattice: Total degrees of freedom - constraints $\propto$ N.
- No internal energy barriers, spins are dynamic.

†Moessner and Chalker, PRB (1998)

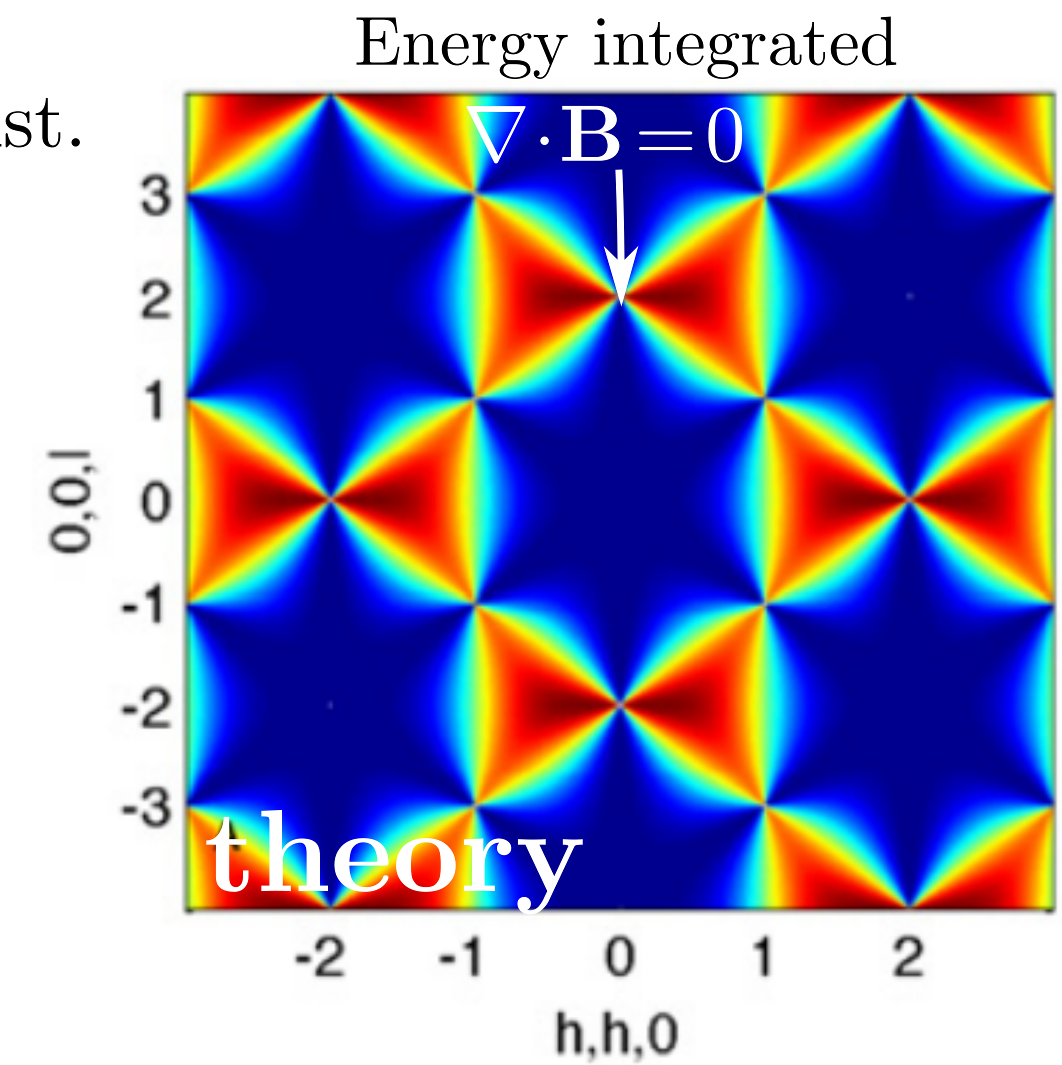
Coulomb Phase: continuous Spins

$$H = J \sum_{\langle ij \rangle} \mathbf{s_i} \cdot \mathbf{s_j} = \frac{J}{2} \sum_{\triangle} |\mathbf{L}_{\triangle}|^2 + \text{cnst.}$$

$$L_{\triangle} = \sum_{i \in \triangle} \mathbf{s}_i$$



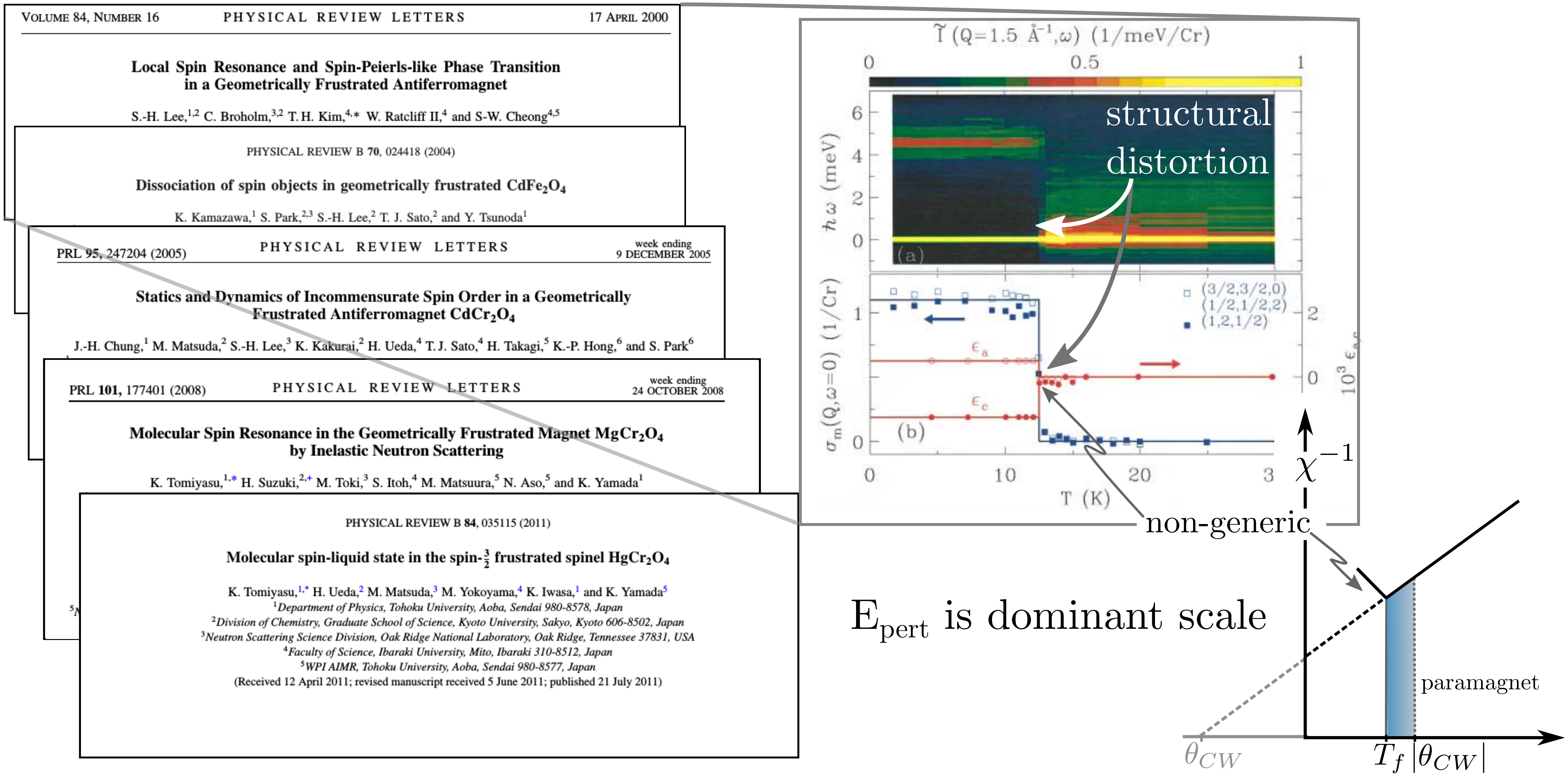
$\mathbf{s}_i = (s_x, s_y, s_z)$



- Can map spin configurations to solenoidal field.
- $\nabla \cdot \mathbf{B} = 0$ implies dipolar correlations $\propto 1/r^3$.
- A (classical) U(1) spin-liquid in three dimensions.
- Unsolved problem in quantum limit.

†Henley, PRB (2005)
Villain, Z. Phys. B. (1979)

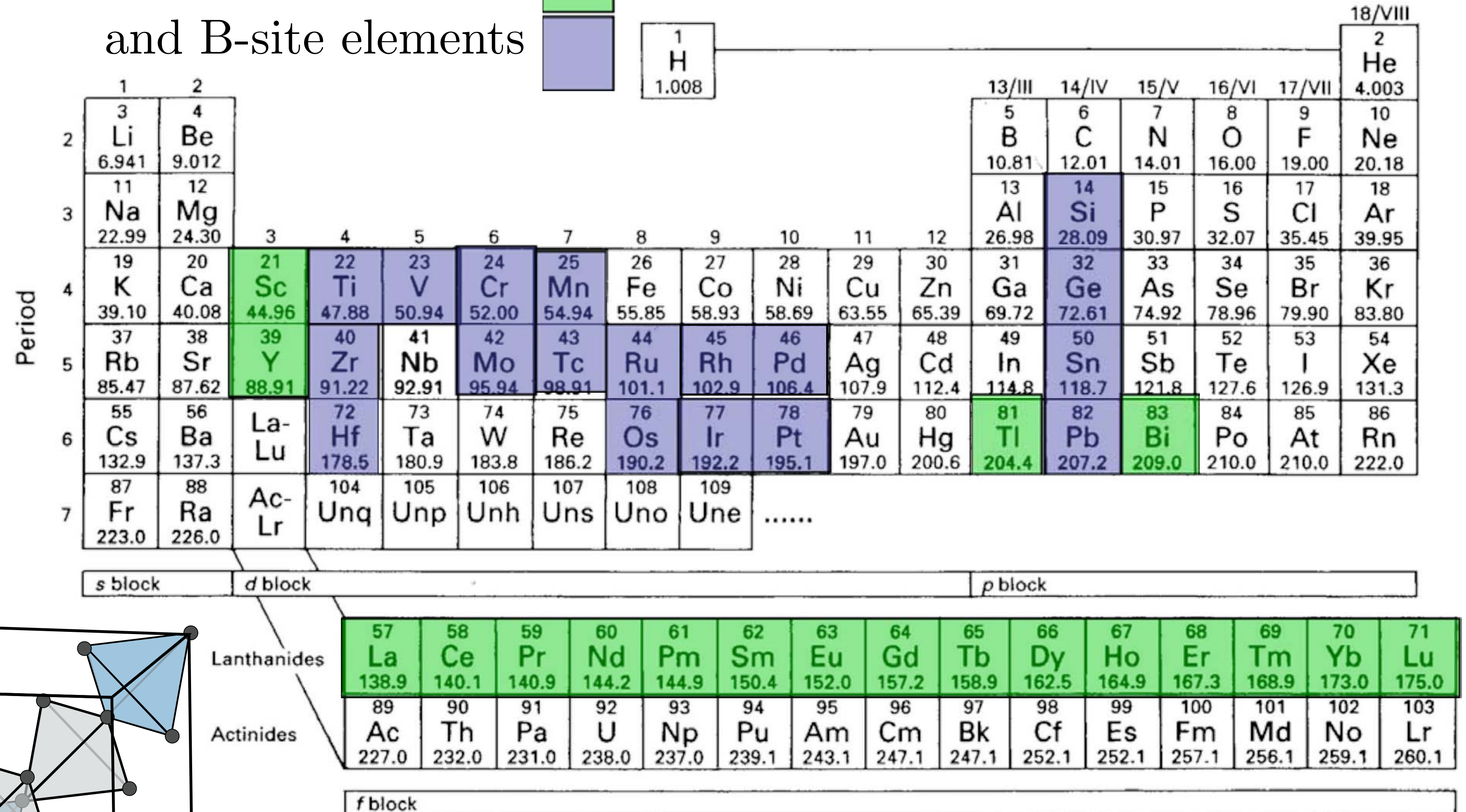
Experiments on Heisenberg pyrochlores

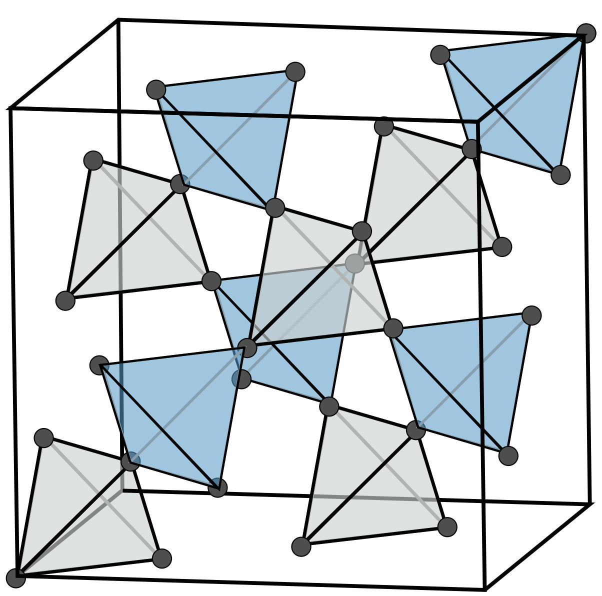


- B-sites of spinel lattice AB_2O_4 have pyrochlore structure.
- In all of these compounds, lattice deformations impact magnetism.
- Perturbations are always there can we access regime where $E_p < k_b T < E_J$.

Why do we need more pyrochlores?

Possible A-site elements
and B-site elements





$A_2B_2O_7^{2-}$

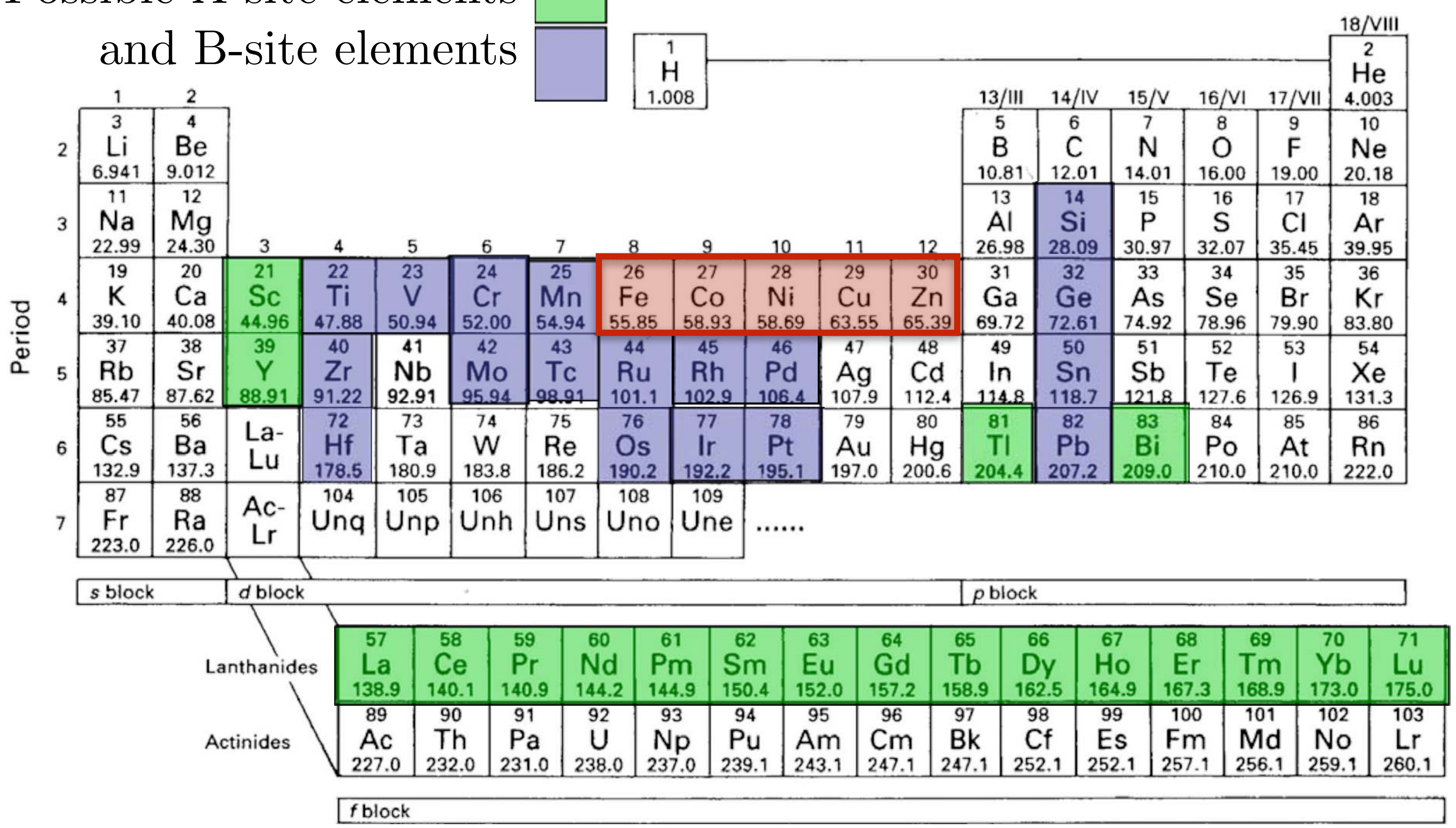
- Stable pyrochlores can be synthesized from many elements.
- Small exchange energy scale from f-electrons.

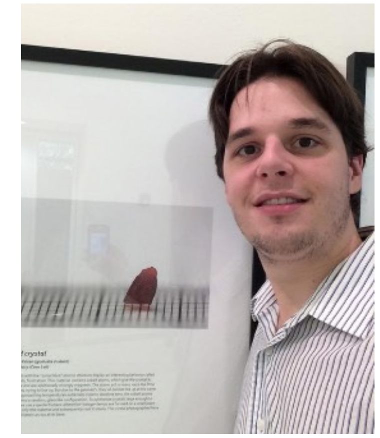
†Gardner *et al.*, Rev. Mod. Phys (2010)

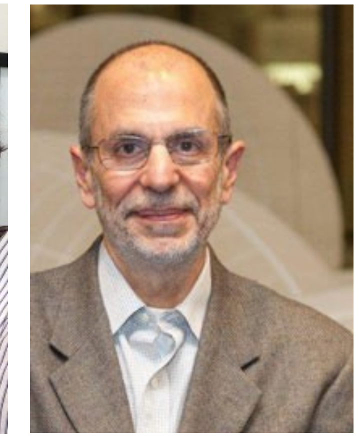
WHY $NaCaNi_2F_7$?

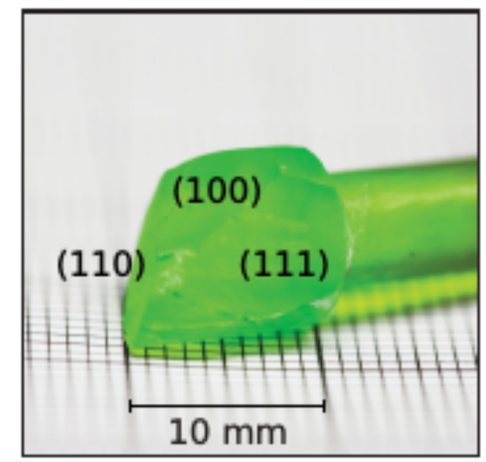
$$A_2(B)_2^{2+}(F)_7^{1-}$$

Possible A-site elements
and B-site elements





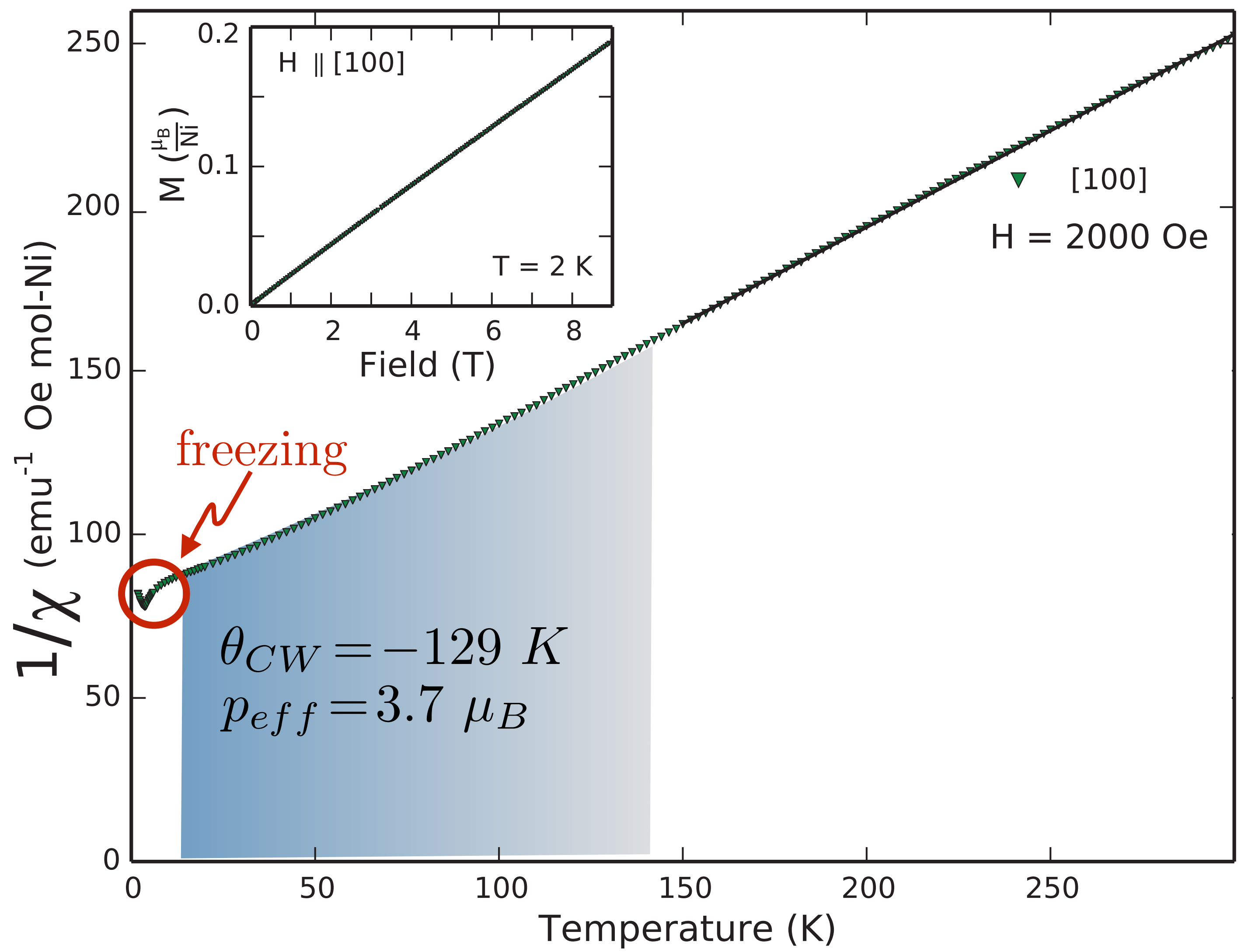


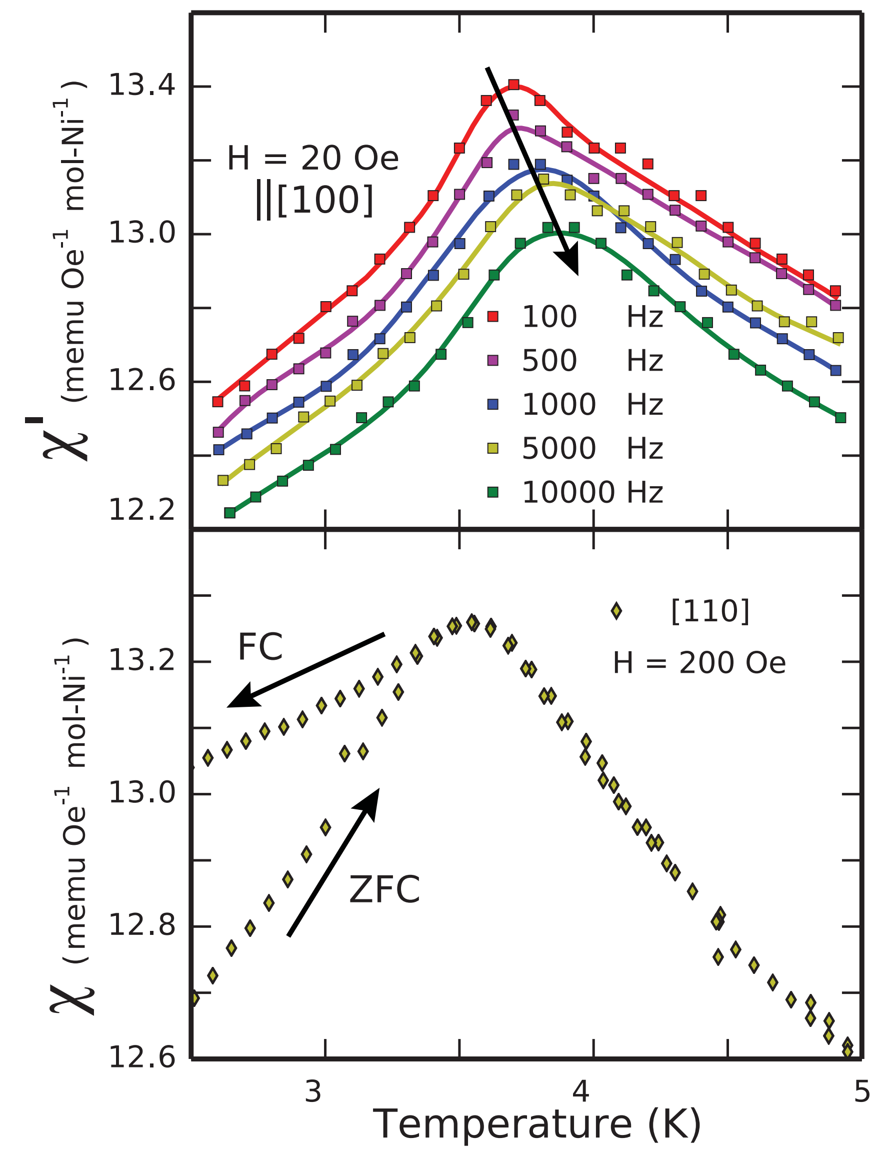


- Stronger magnetic exchange for 3d transition metals.
- Low oxidation states of 3d TM's rules out oxides.
- Charge balance requires +1.5 on A-site ⟶ mix Na^{1+} and Ca^{2+}.

†Krizan and Cava, PRB (2015)

FLOURIDE PYROCHLORES: $NaCaNi_2F_7$

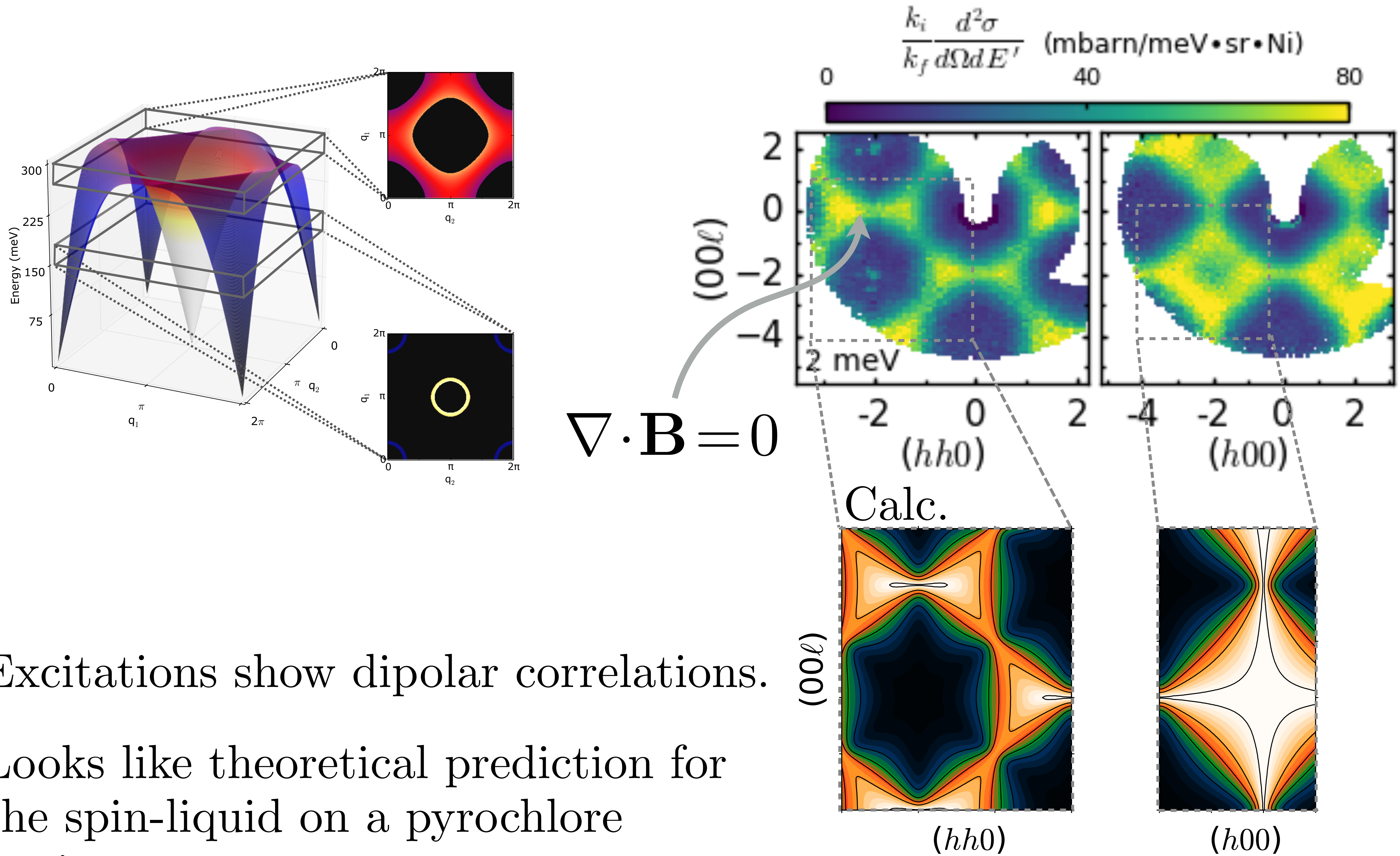




- Magnetic interactions are strong: Θ_{CW} = -129 K.
- $Ni^{2+} \longrightarrow$ S=1, in the quantum limit.
- Spin freezing at T_f = 3.5 K. Signatures of a spin glass.
- Estimate bond disorder from T_f: $\delta J \approx 0.2$ meV.

†Krizan and Cava, PRB (2015)

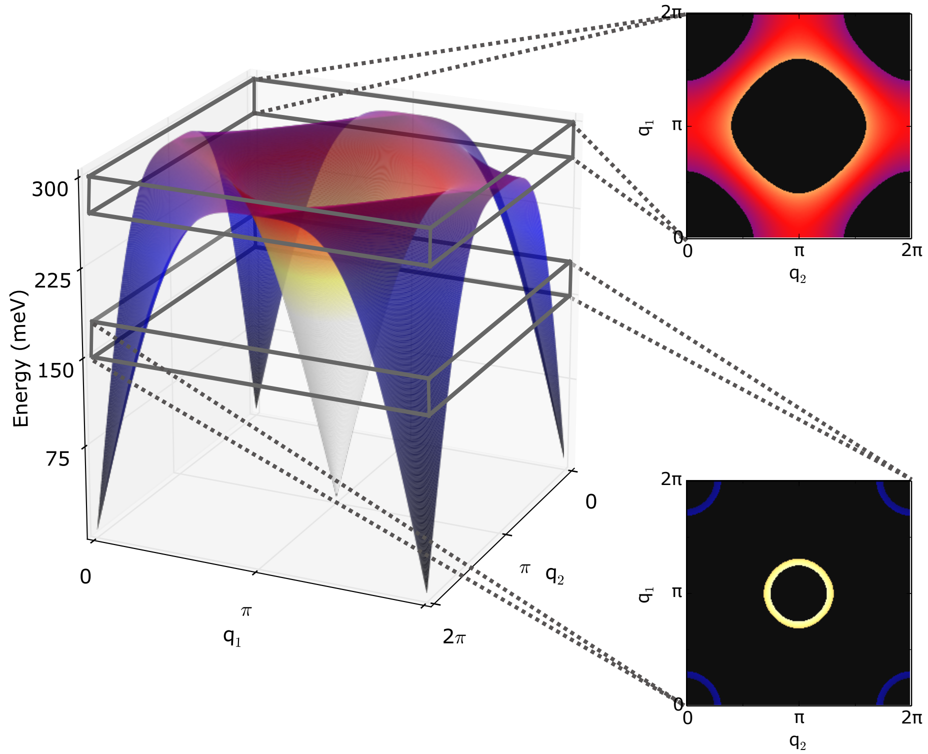
Magnetic excitations in $NaCaNi_2F_7$

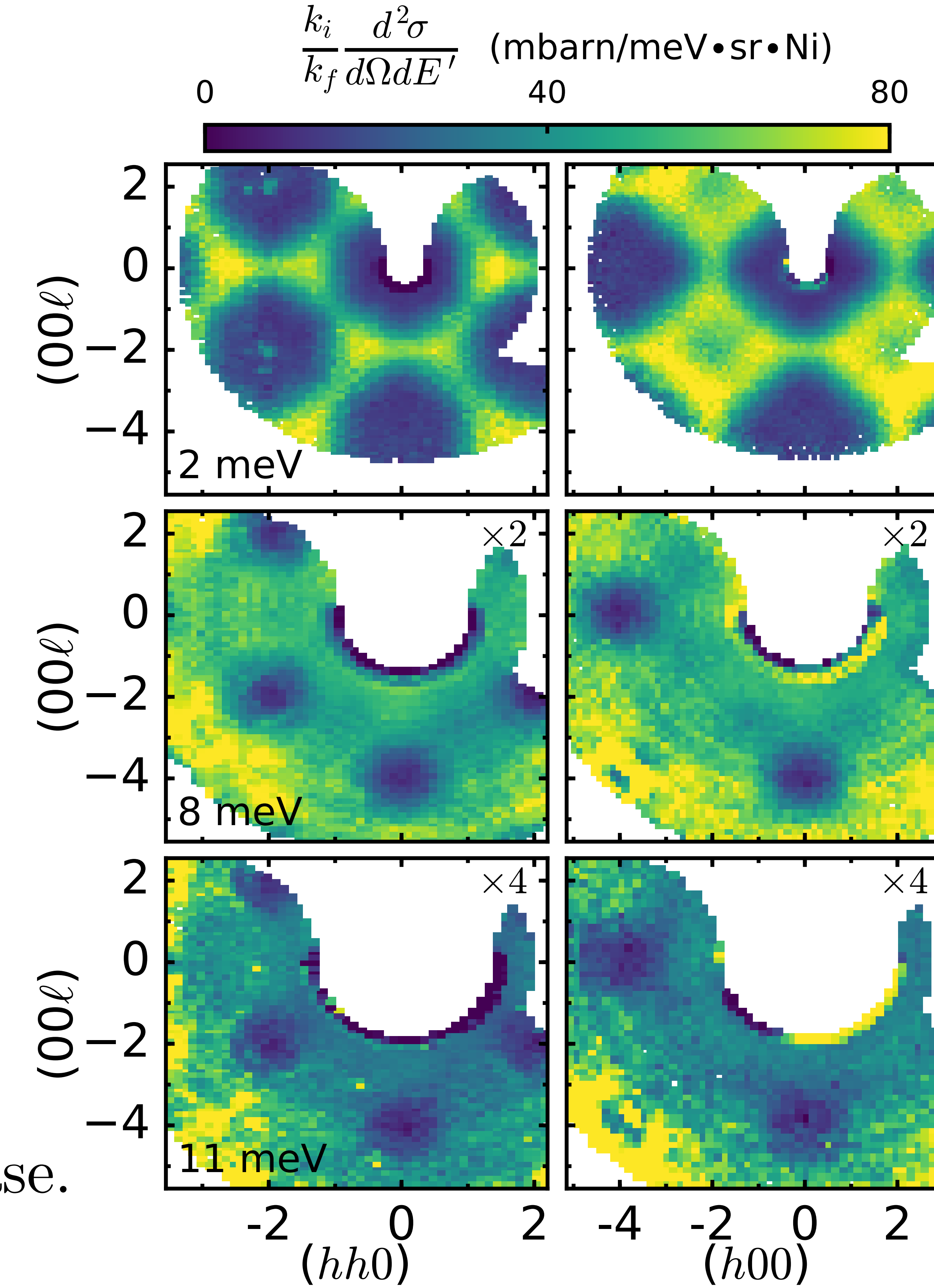


$$\nabla \cdot \mathbf{B} = 0$$

- Excitations show dipolar correlations.
- Looks like theoretical prediction for the spin-liquid on a pyrochlore lattice.

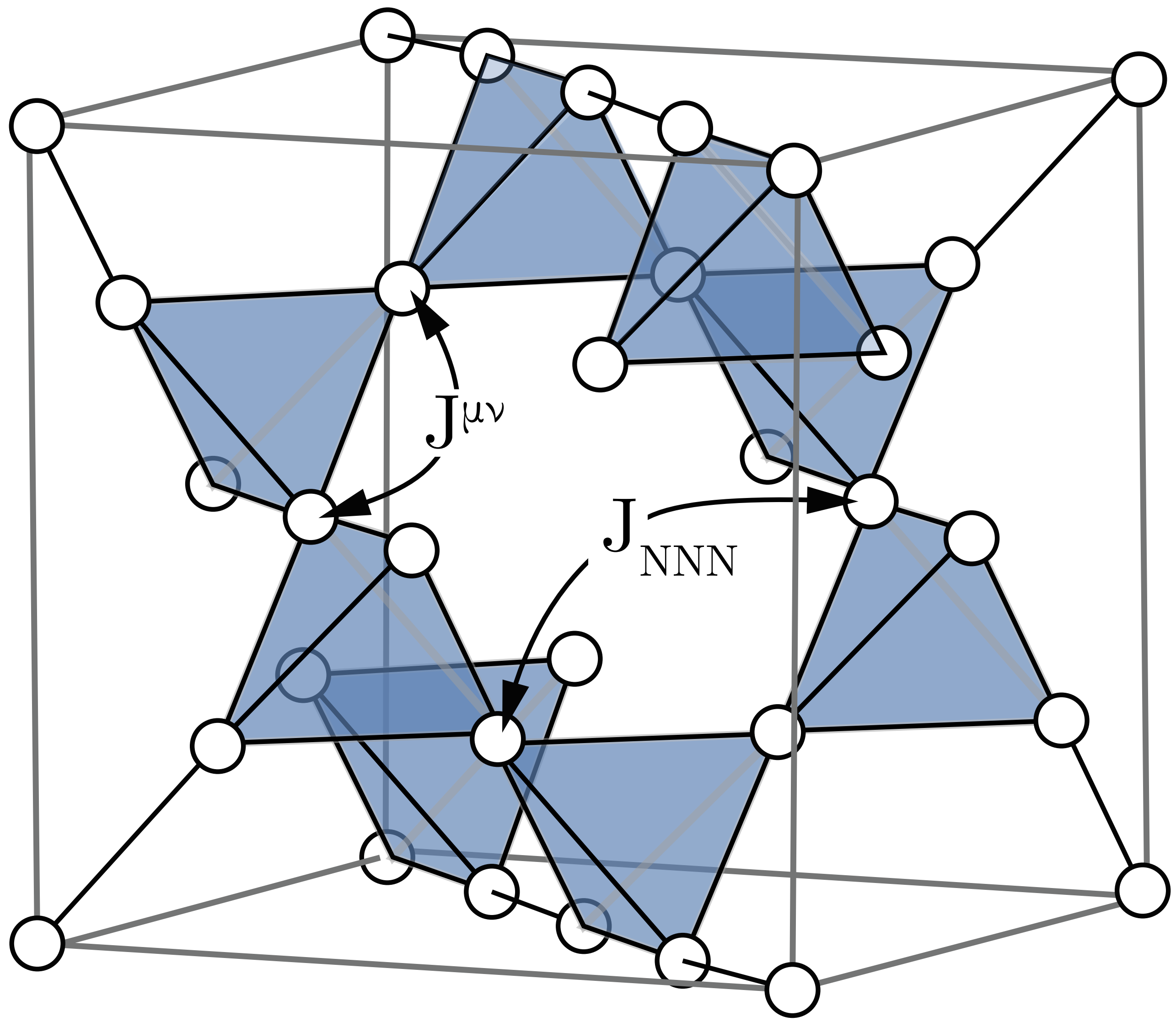
Magnetic excitations in $NaCaNi_2F_7$





- No LRO and no spin waves.
- Dispersion perpendicular to bow-ties.
- Excitations of the Coulomb phase.

Determining the magnetic interactions



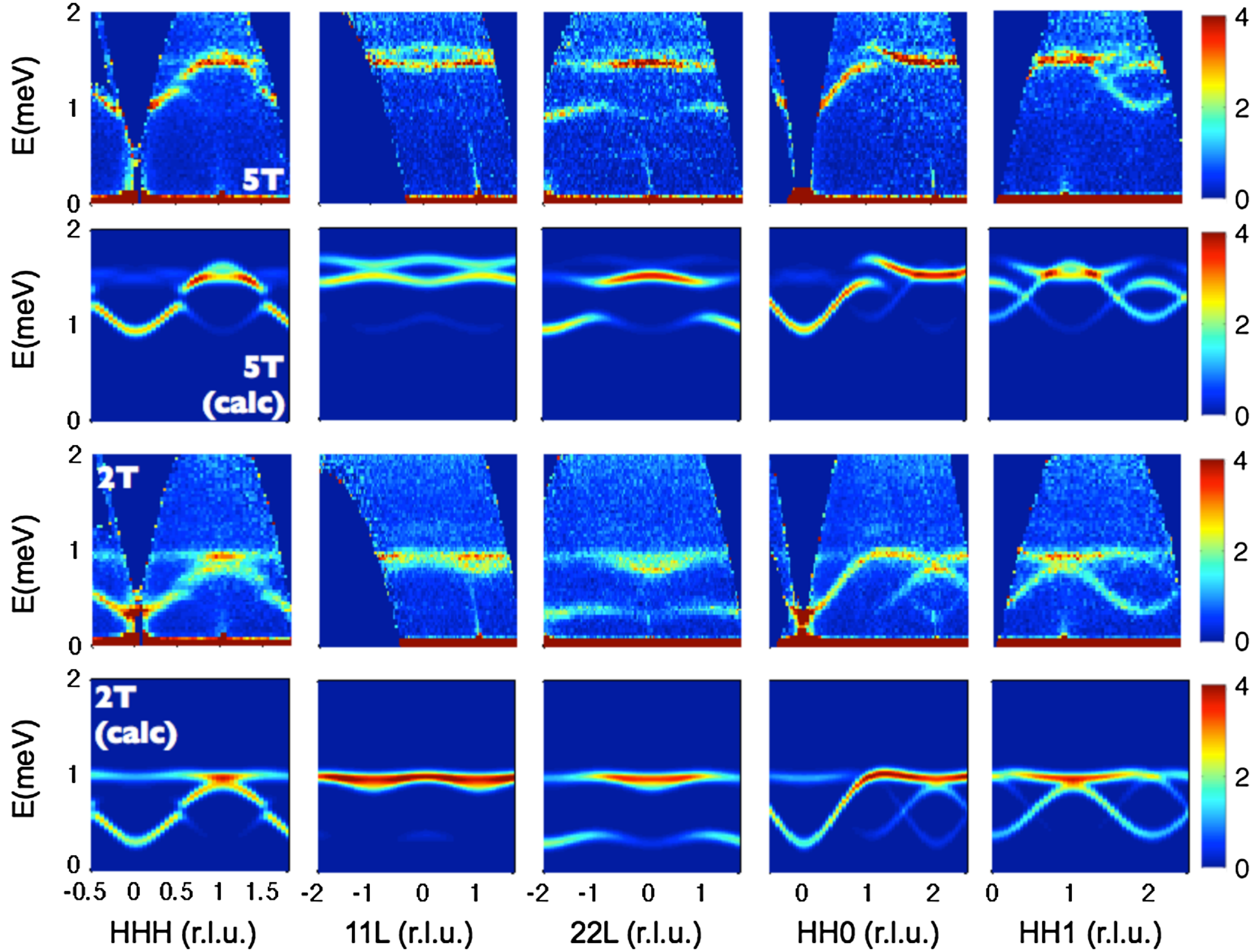
$$H = \sum \mathbf{S_i} \cdot \mathbf{J} \cdot \mathbf{S_j}$$

$$\mathbf{J} = \begin{pmatrix} J_2 & J_4 & J_4 \\ -J_4 & J_1 & J_3 \\ -J_4 & J_3 & J_1 \end{pmatrix}$$

- Must consider symmetry allowed exchange matrix.
- Further neighbor exchange also possible.

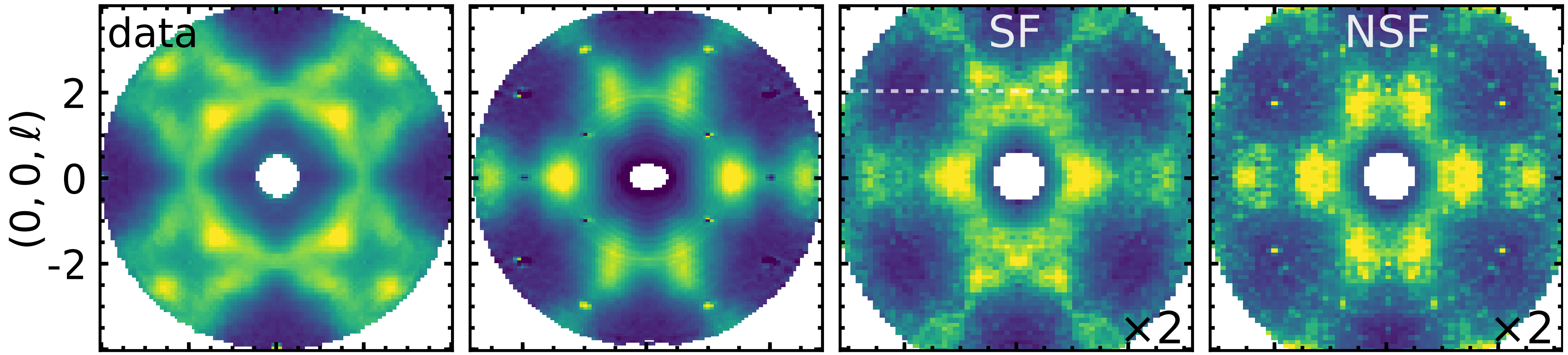
Determining the magnetic interactions

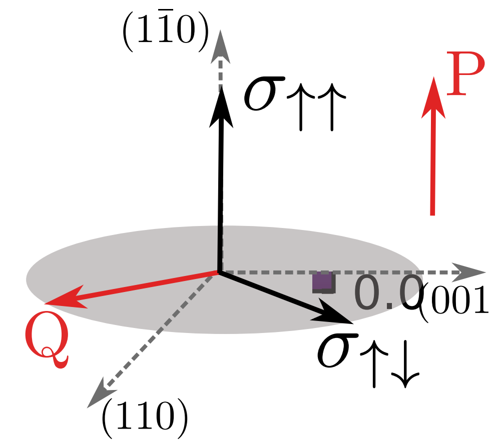
$$H = \sum \mathbf{S_i} \cdot \mathbf{J} \cdot \mathbf{S_j} \qquad \mathbf{J} = \begin{pmatrix} J_2 & J_4 & J_4 \\ -J_4 & J_1 & J_3 \\ -J_4 & J_3 & J_1 \end{pmatrix}$$



†Ross *et al.*, PRX (2011)

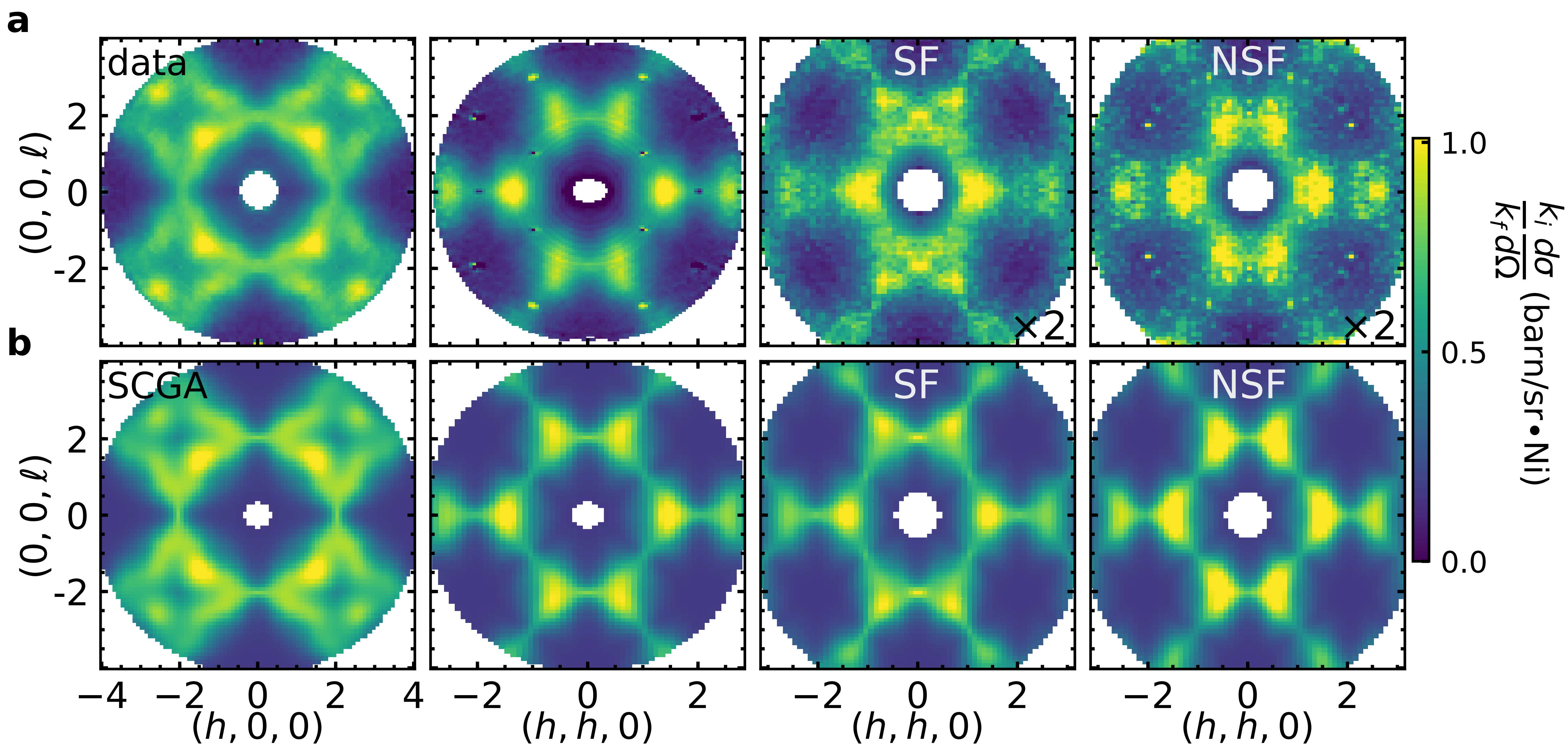
Determining the magnetic interactions





- Measure equal time correlation function S(Q).
- Resolve spin components with polarized neutrons, large anisotropies are ruled out.
- Refine interactions using Gaussian approximation to calculate S(Q).

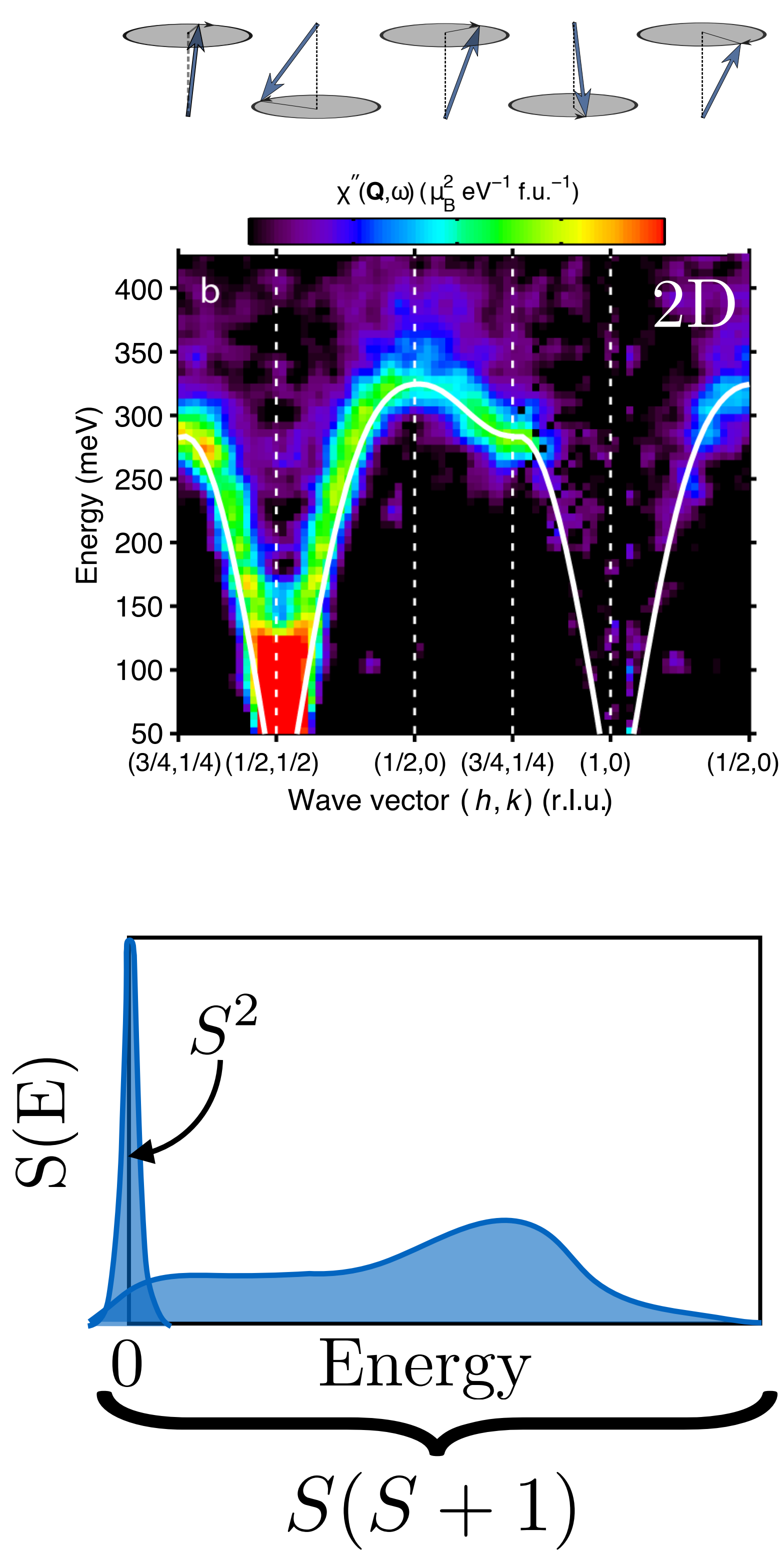
Determining the magnetic interactions



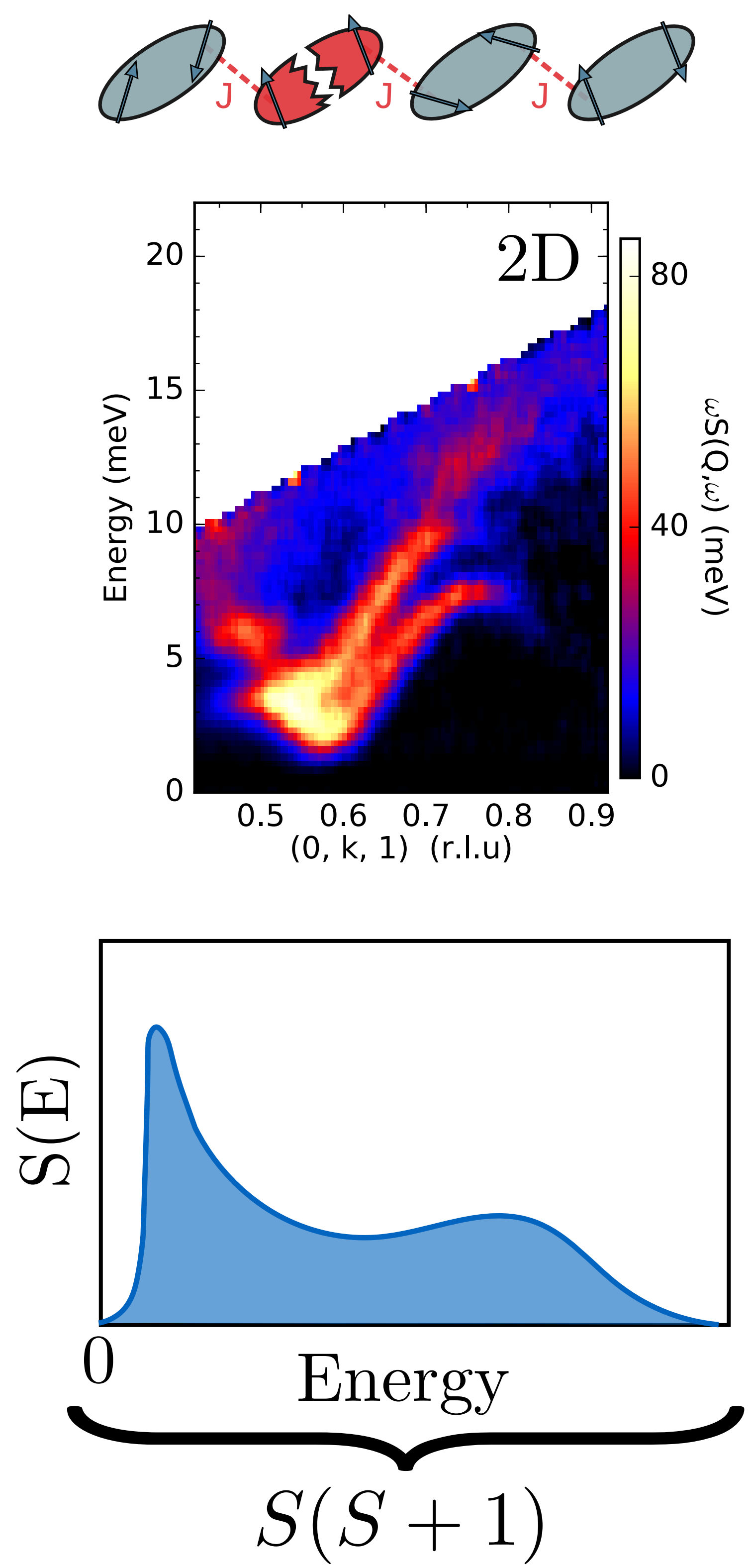
- Global refinement of Gaussian approximation to S(Q).
- Nearest neighbor: $J_1 = J_2 = 3.2(1)$ meV, $J_3 = 0.019(3)$ meV, $J_4 = -0.070(4)$ meV.
- Next nearest neighbor: $J_{NNN} = -0.025(5)$ meV.

Collective Excitations in Magnets

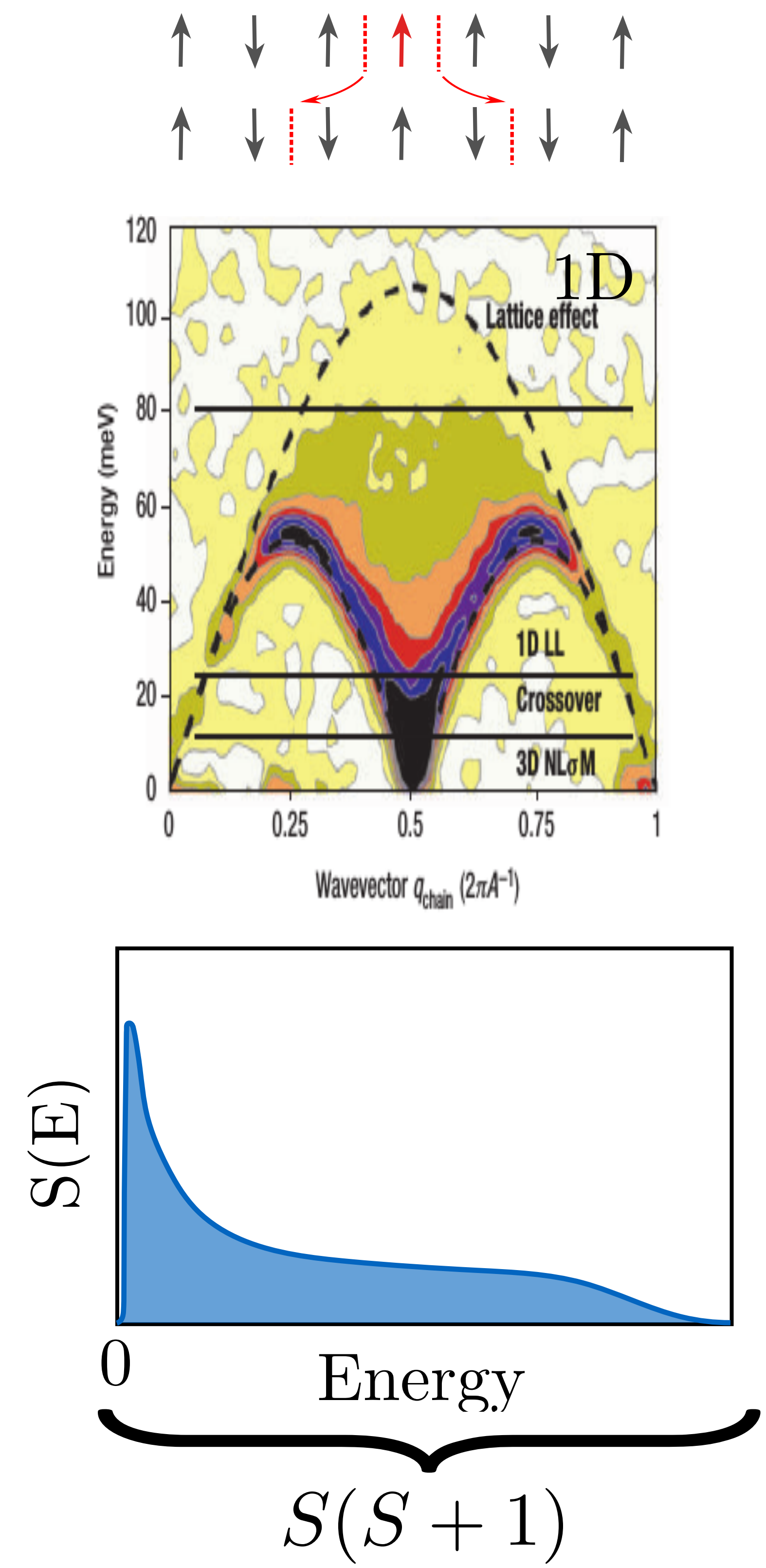
magnetic order



valence bond order

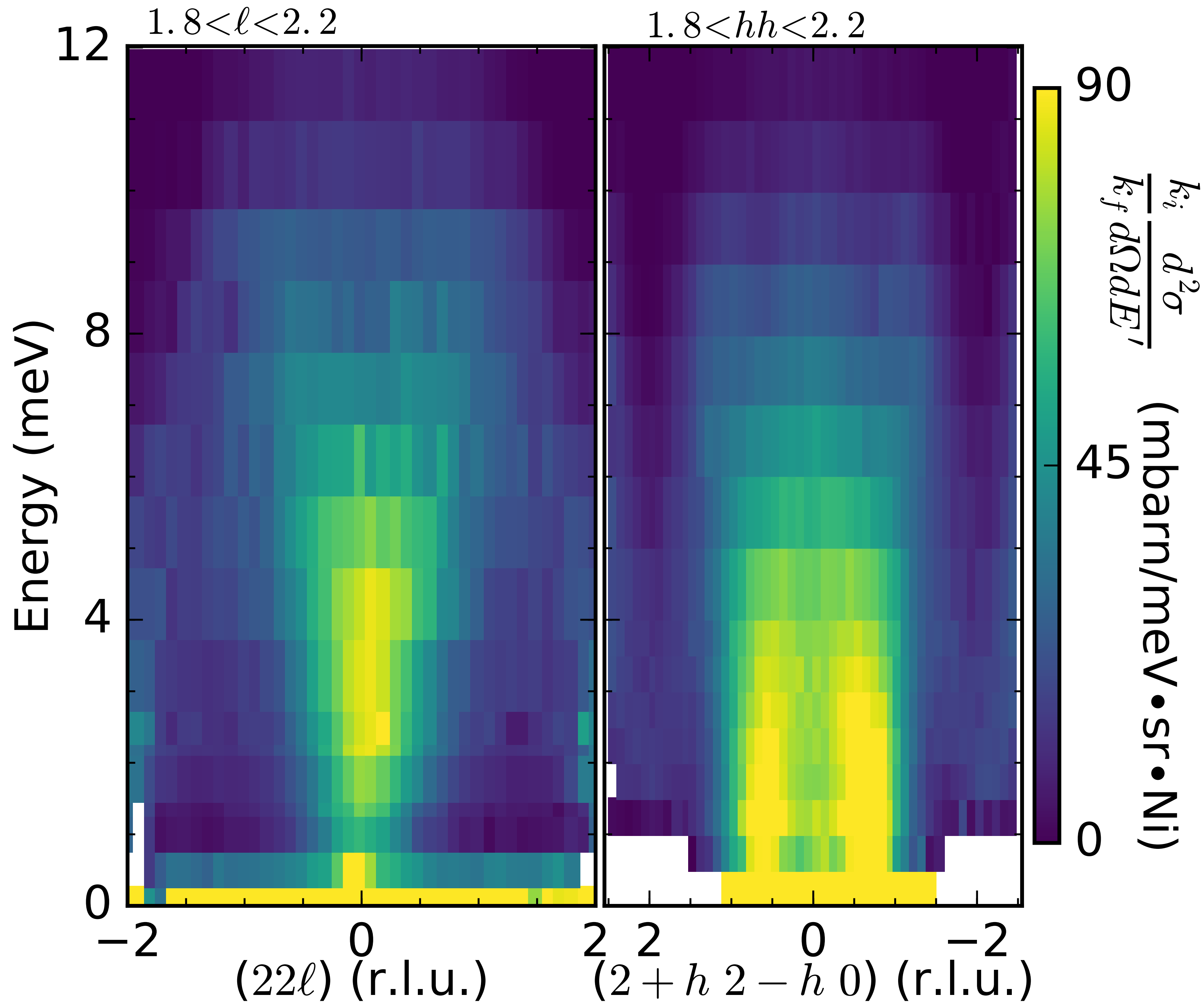


fractionalization



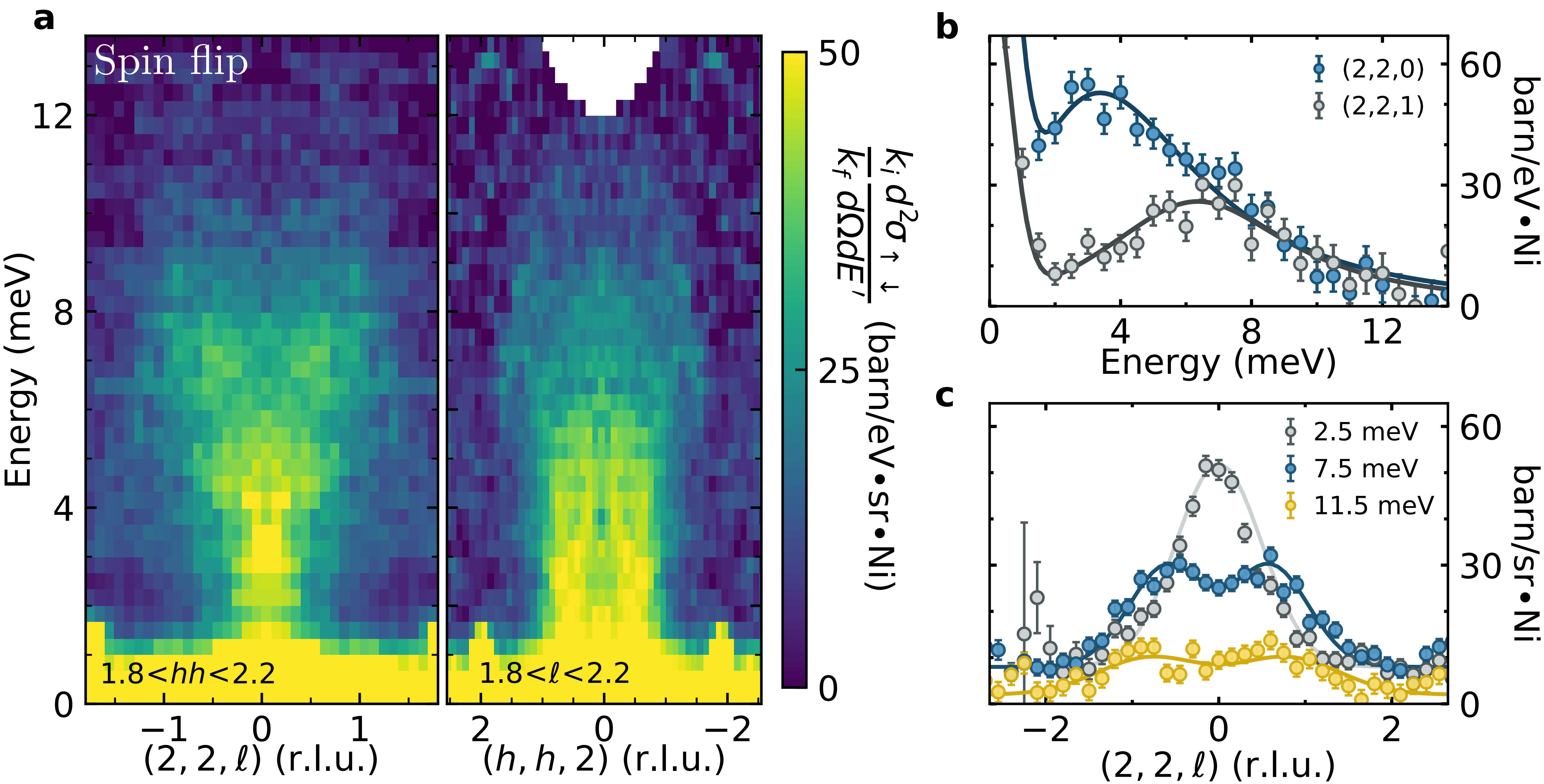
†Headings *et al.* PRL (2010), Plumb *et al.*, N. Phys. (2016), Lake *et al.* N. Matter. (2005)

Excitations from a Coulomb phase



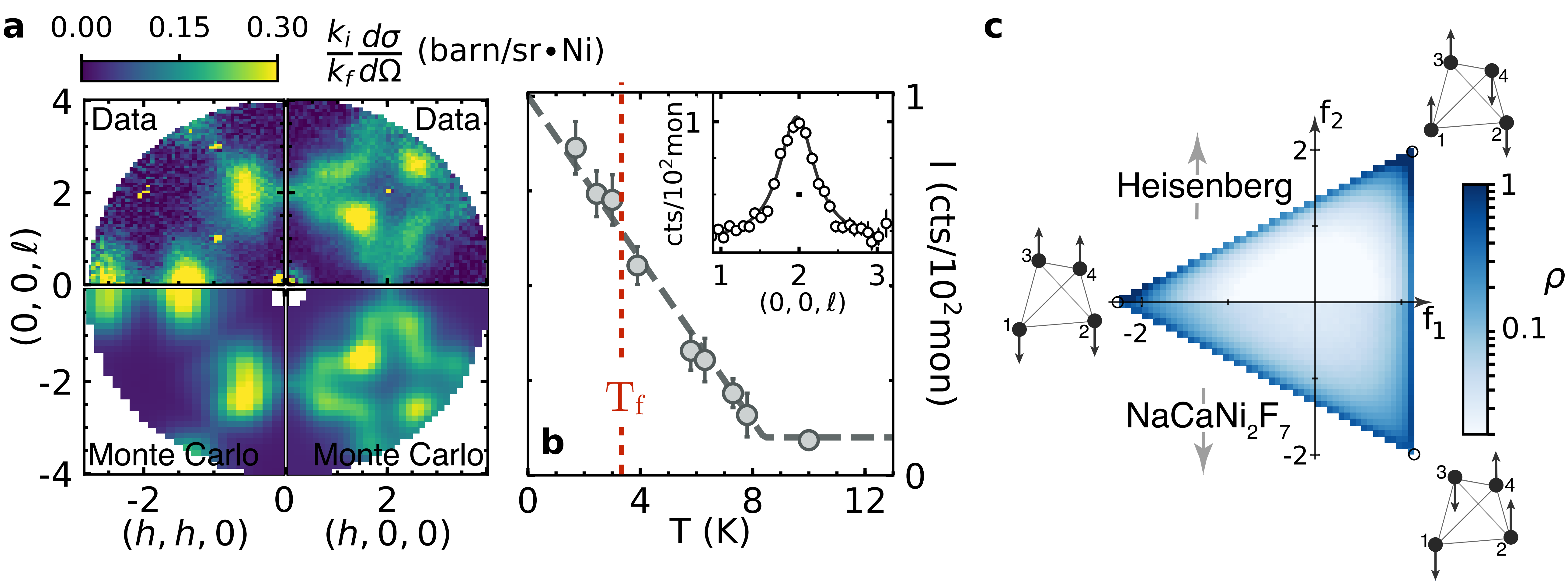
- Continuum of excitations in a three-dimensional magnet.
- Total signal is $S\,(S+1)$and 90% of the spectral weight is inelastic (>0.4 meV).

Excitations from a Coulomb phase



- No resonant modes, but well defined structure.
- Spectral weight build up at 3.5 meV≈J_1.

Unconventional Spin freezing

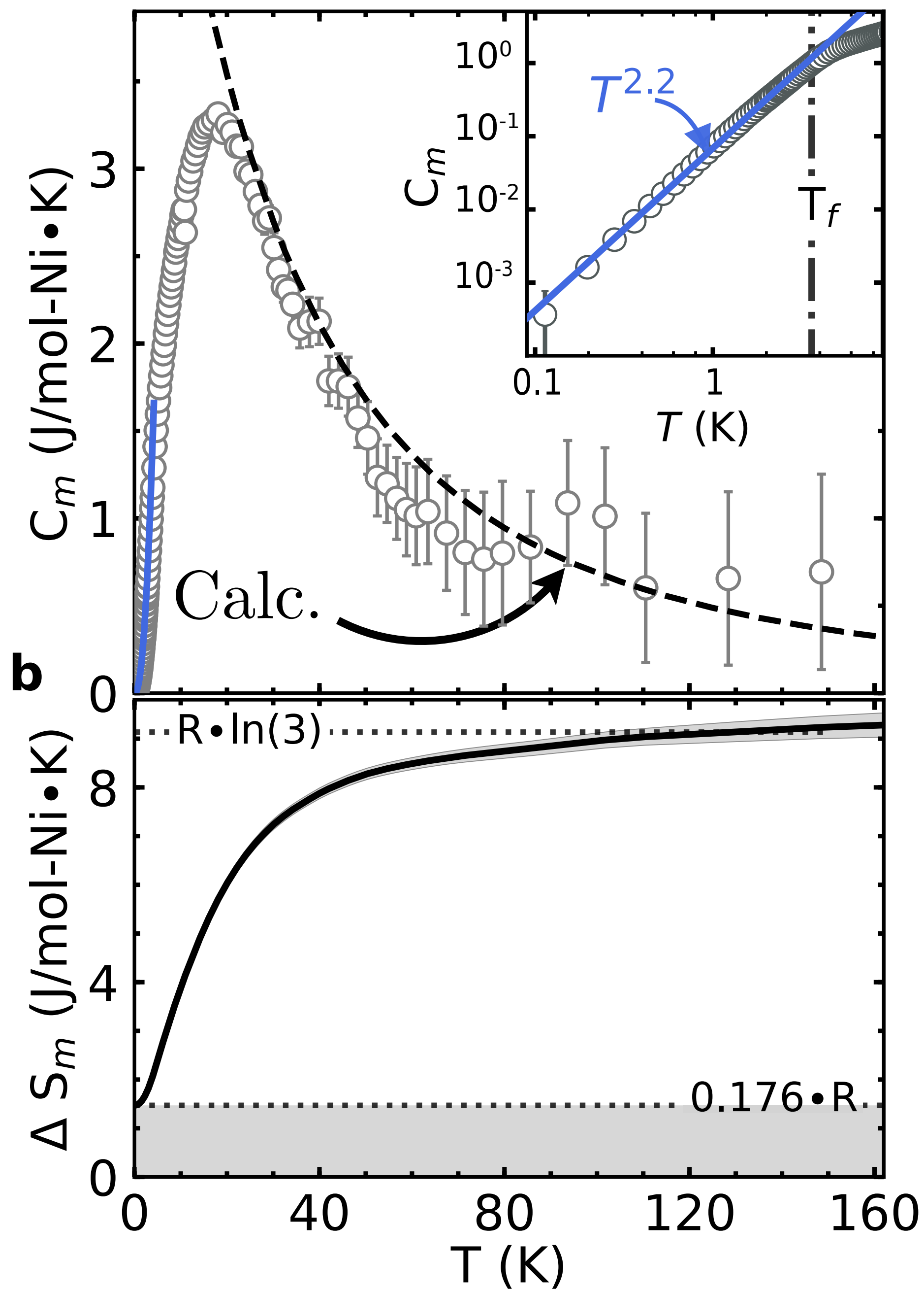


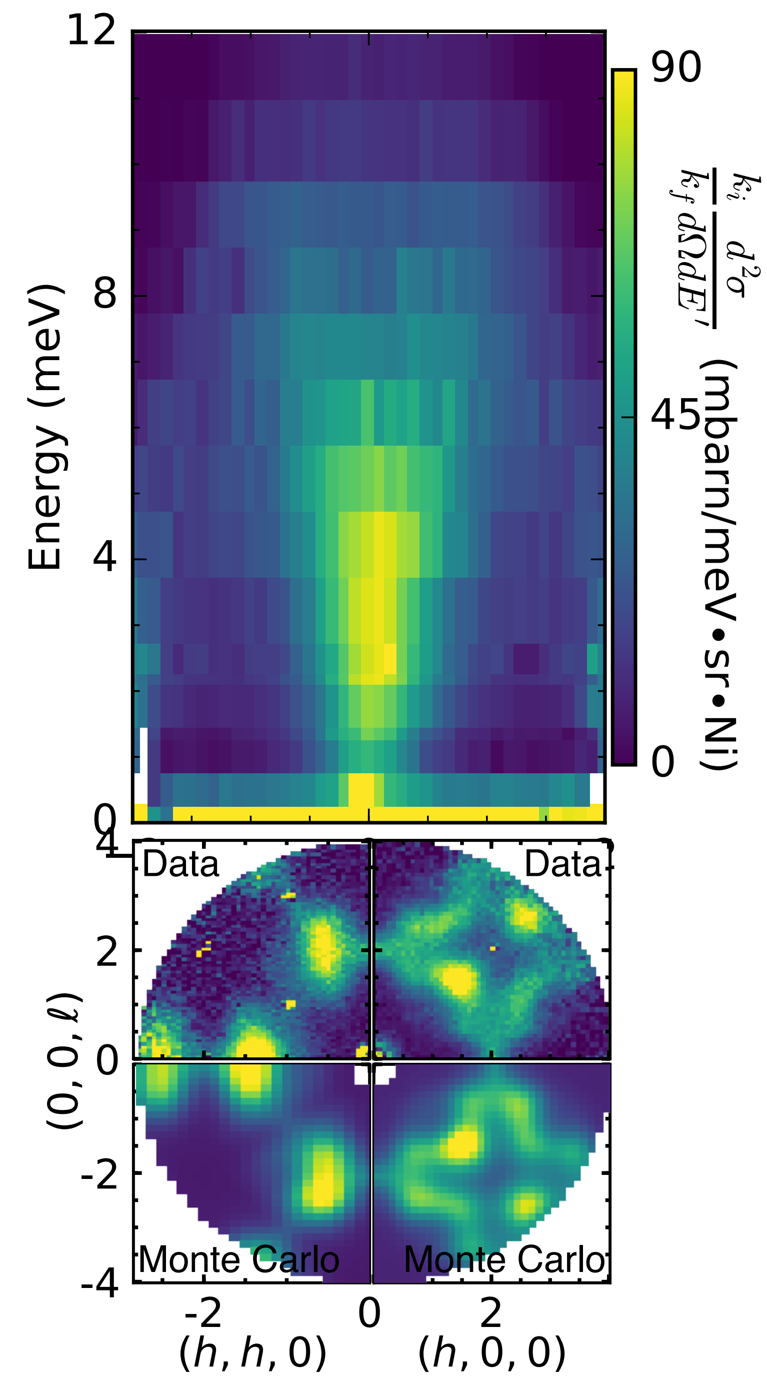
- Unconventional frozen ground state.

$$\langle m_{elastic}\rangle^2/(g^2 S(S+1)) = 10\%$$

- Study local structure with numerics and knowledge of Hamiltonian.
- Frozen moment configurations have approximately $\mathbf{S}_{\rm tot} = \sum_{i\in\Delta} \mathbf{S}_i = 0$.

The overall picture

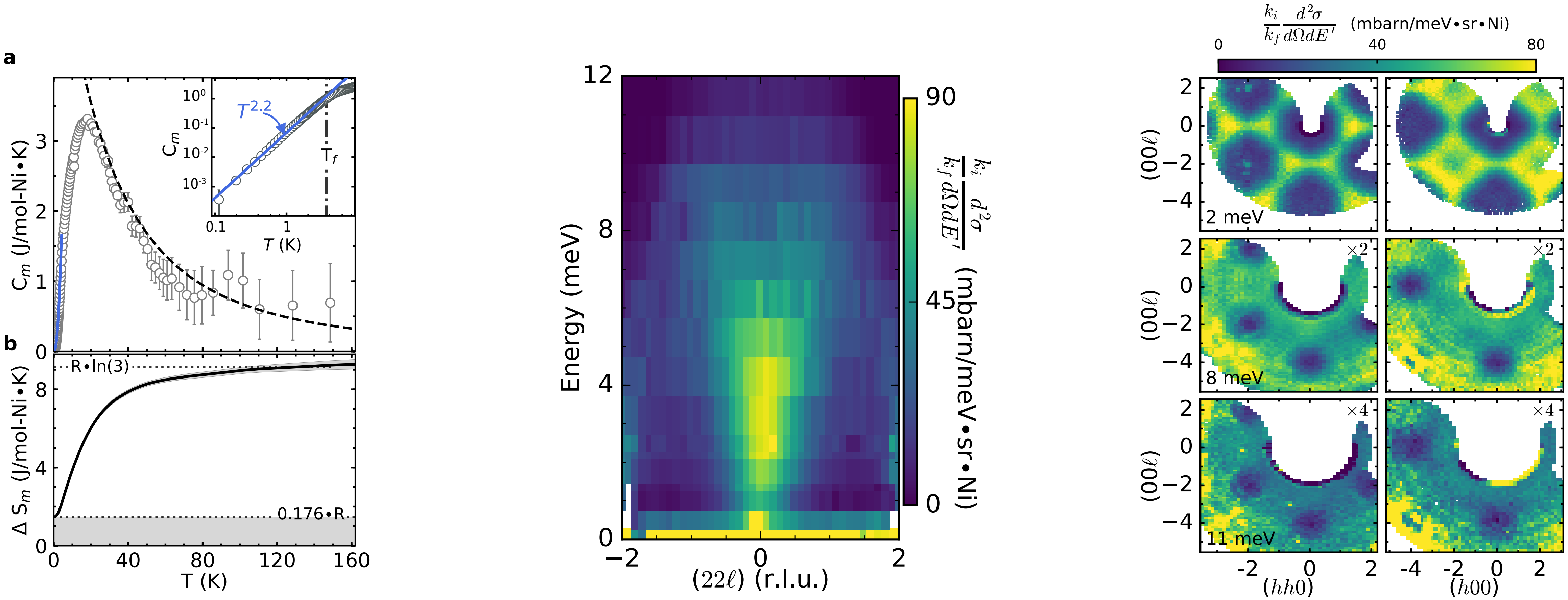




- Classical spin liquid above 20 K. Villain's cooperative paramagnet.
- Residual entropy is 0.176/spin $\longrightarrow$ broken ergodicity, disorder?

$$\Delta S/R\ln(3) \approx \langle m_{elastic}\rangle^2/(g^2 S(S+1))$$

Summary



- Role of "quantum" $\rightarrow$ 90% of spectral weight inelastic.
- Existence of quaisparticles? Fractionalization?
- Nature of the frozen state? Kinetically arrested spin liquid?

Thank You!