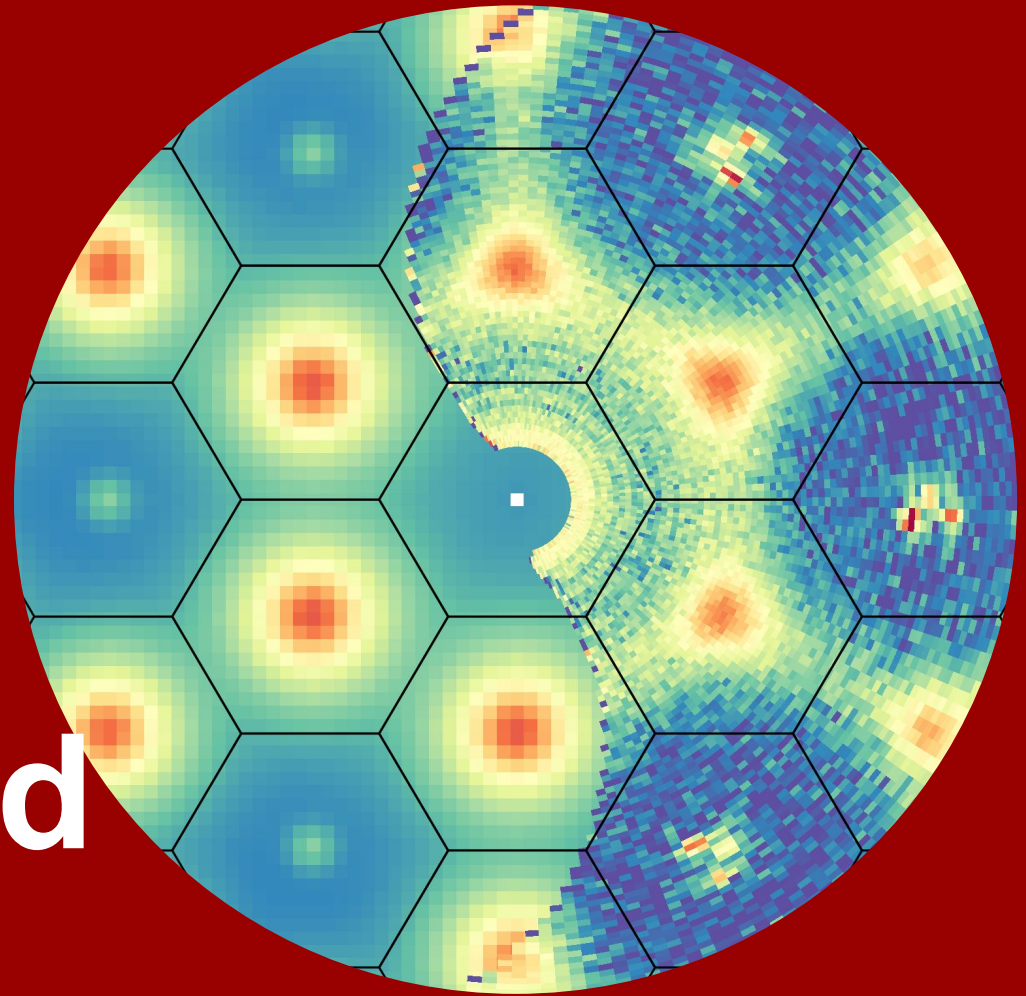
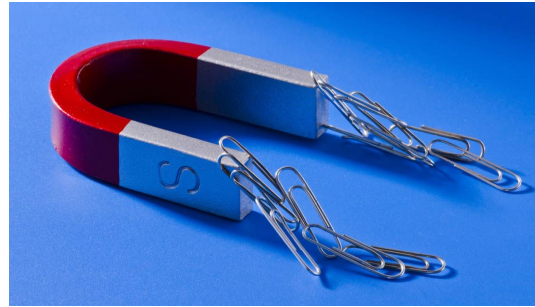


Sofie Janas  
Postdoc

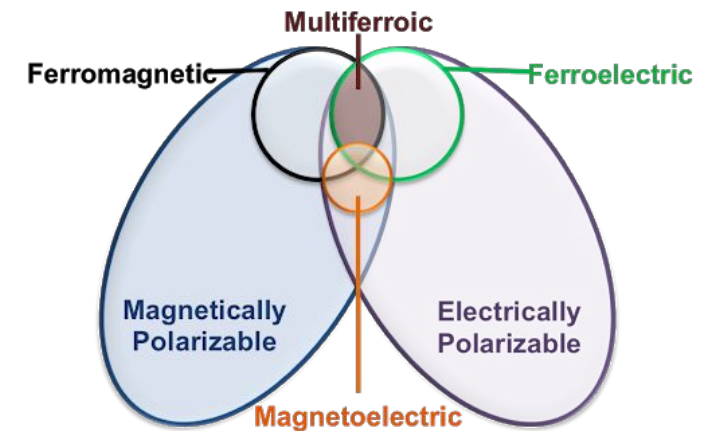
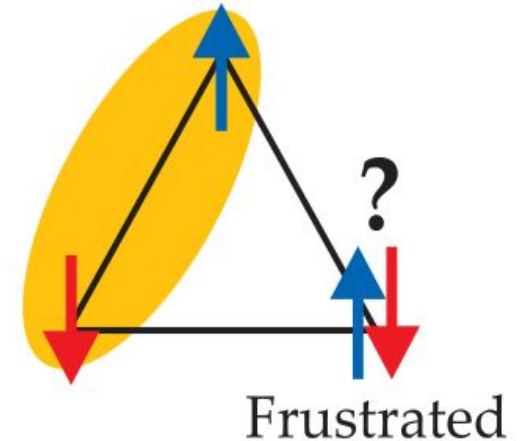
# Classical spin liquid phase in frustrated magnet $\text{YMnO}_3$



# My background



- PhD in Physics from Niels Bohr Institute
  - Supervisor: Kim Lefmann
  - Neutron spectroscopy
  - Frustrated magnets: triangular and kagome geometries
- Now: Postdoc at Technical University of Denmark
  - With Rasmus Toft-Petersen (BIFROST lead)
  - Project: magnetoelectric materials & time-resolved neutron diffraction

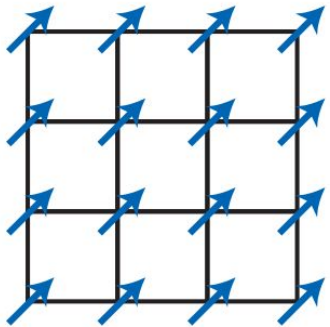


# Geometrically frustrated magnets: The Triangular lattice Ising magnet

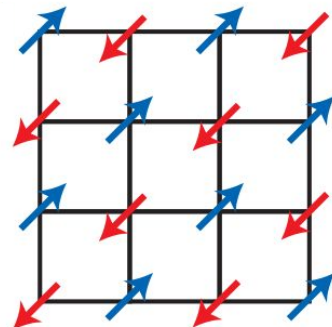
$$\mathcal{H} = -J \sum_{ij} S_i^z S_j^z$$

Frustration suppresses magnetic ordering

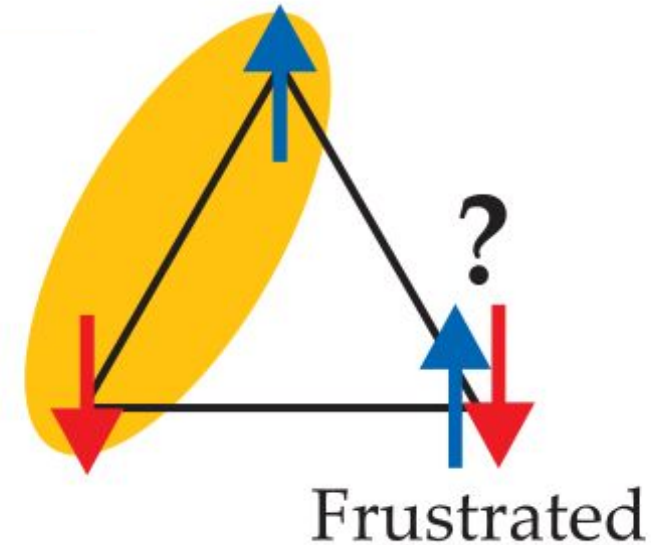
Ferromagnet



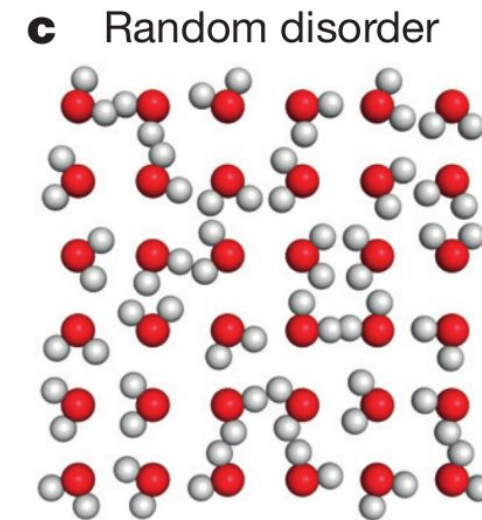
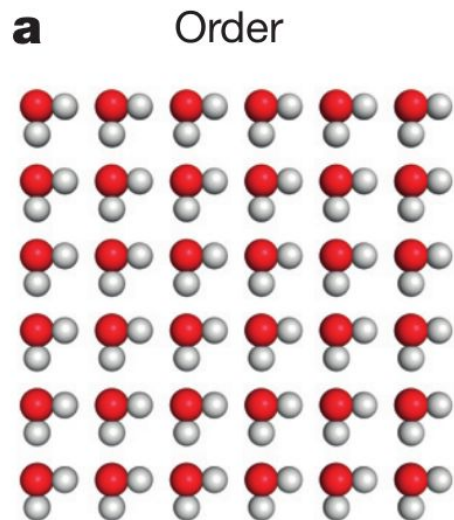
Antiferromagnet



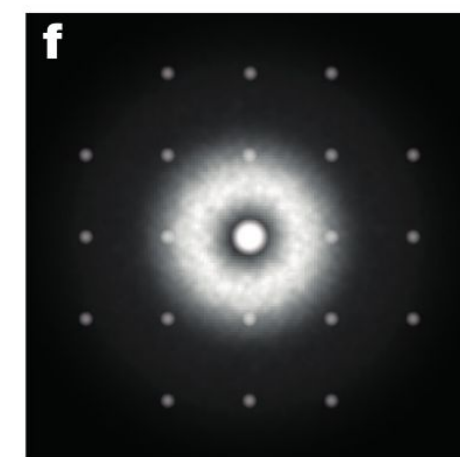
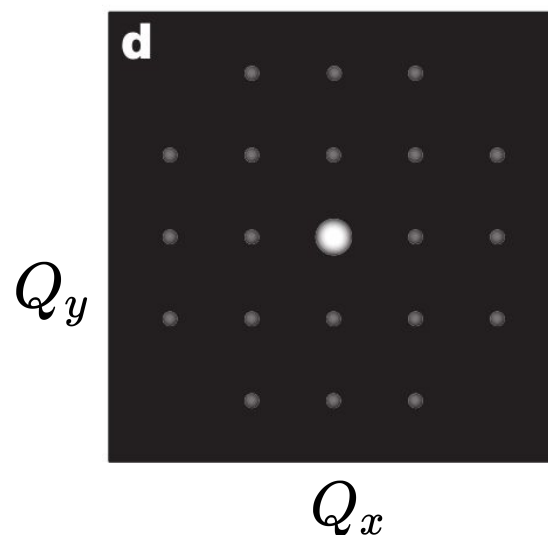
Competing interactions give degenerate ground states



# Correlated disorder in square ice



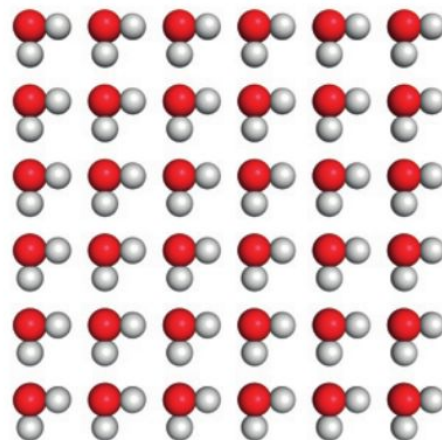
Reciprocal space,  $1/\text{\AA}$



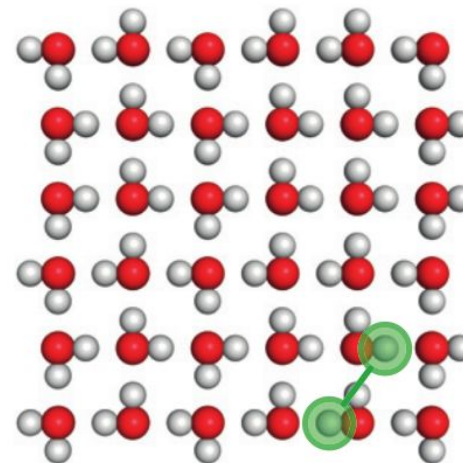
# Correlated disorder in square ice



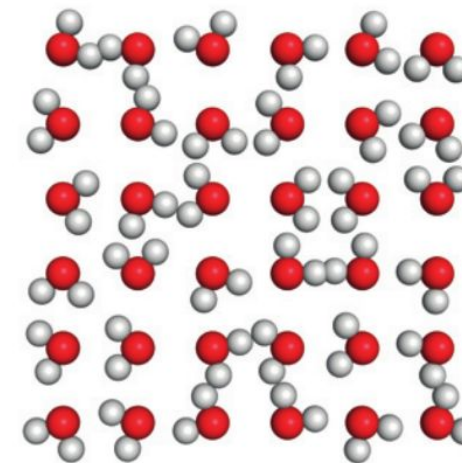
**a** Order



**b** Correlated disorder

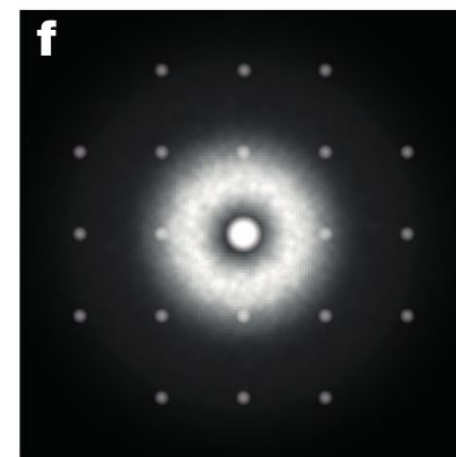
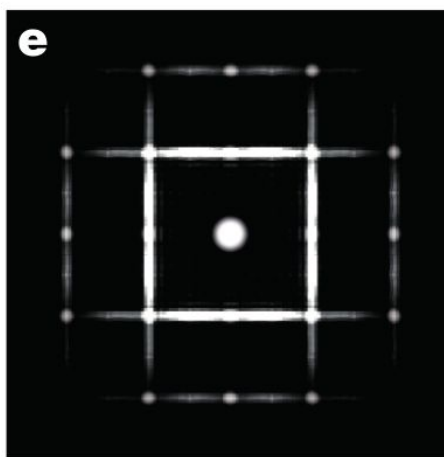
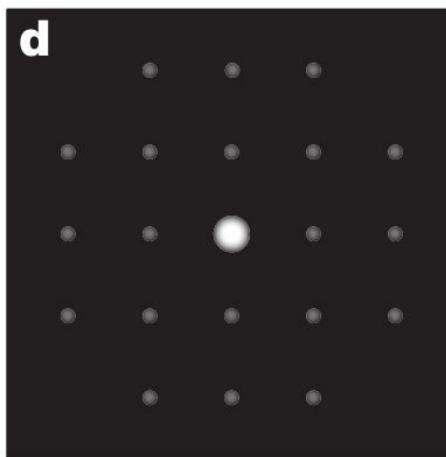


**c** Random disorder



Reciprocal space,  $1/\text{\AA}$

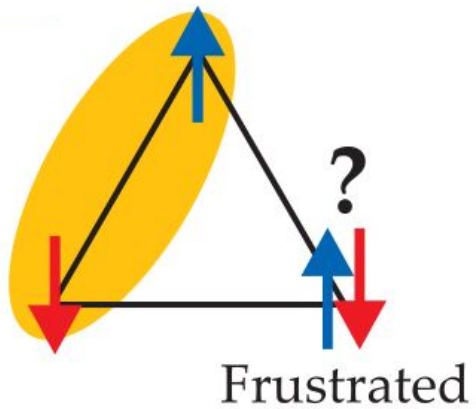
$Q_y$



$Q_x$

# Geometrically frustrated magnets: The Triangular lattice Ising magnet

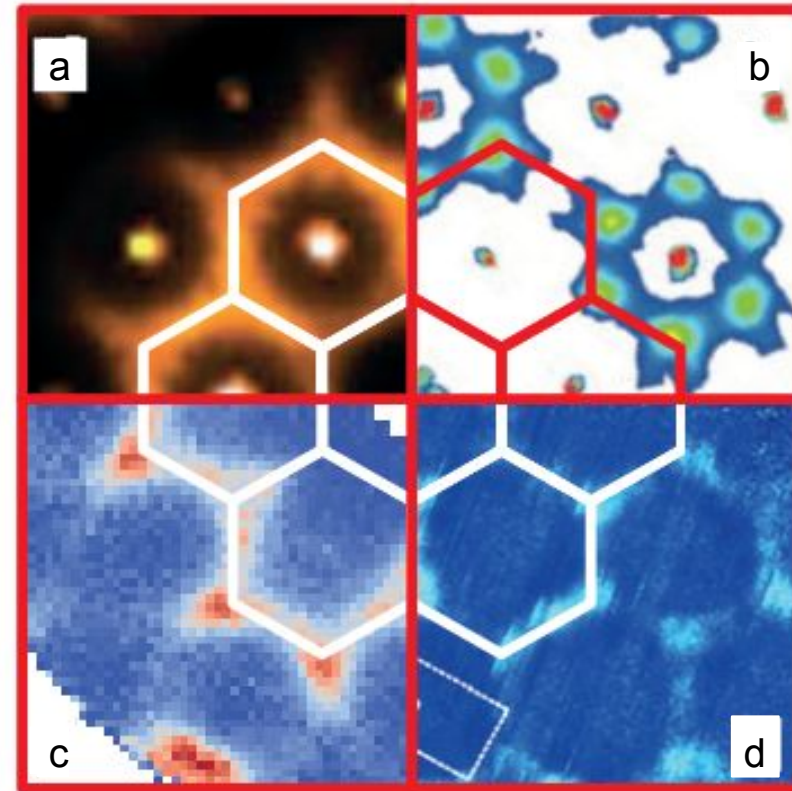
$$\mathcal{H} = -J \sum_{ij} S_i^z S_j^z$$



Packing of  
Gag proteins

$Q_y$

Spin structure  
in  $\beta$ -Mn



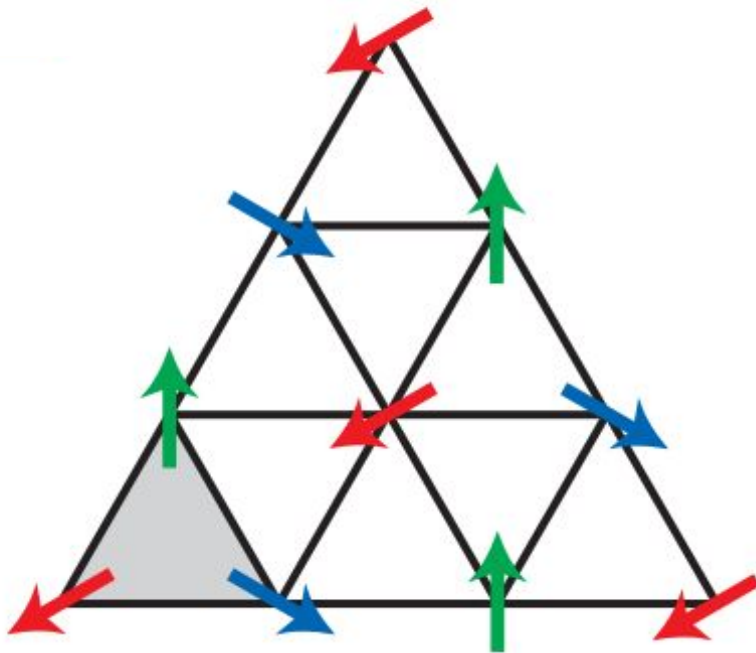
Orbital orientation  
in frustrated  
magnet  
 $\text{Ba}_3\text{CuSb}_2\text{O}_9$

Electronic  
structure in  
graphene

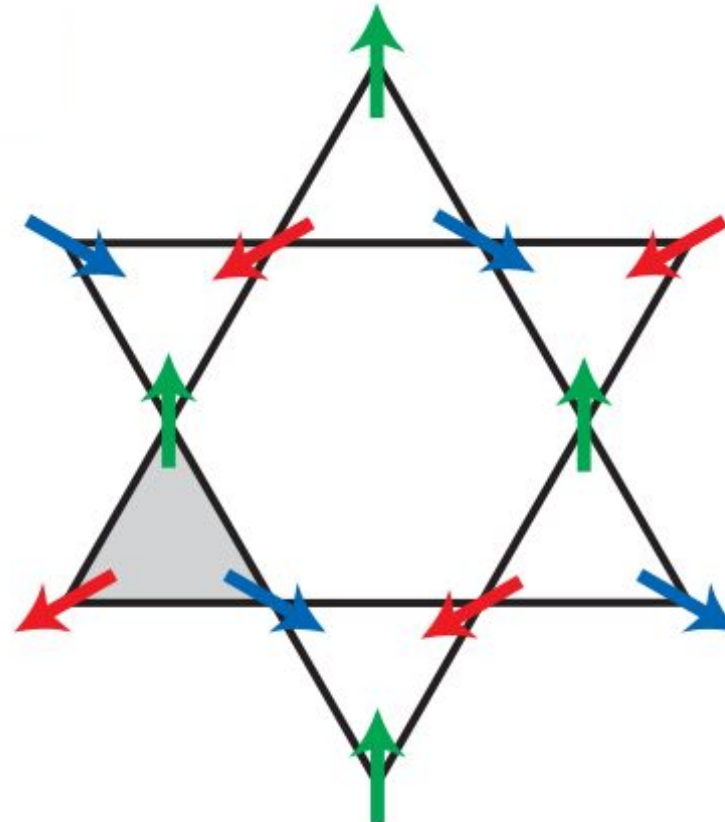
$Q_x$

# Geometrically frustrated magnets: The Heisenberg magnet

$$\mathcal{H} = -J \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$



Triangular lattice



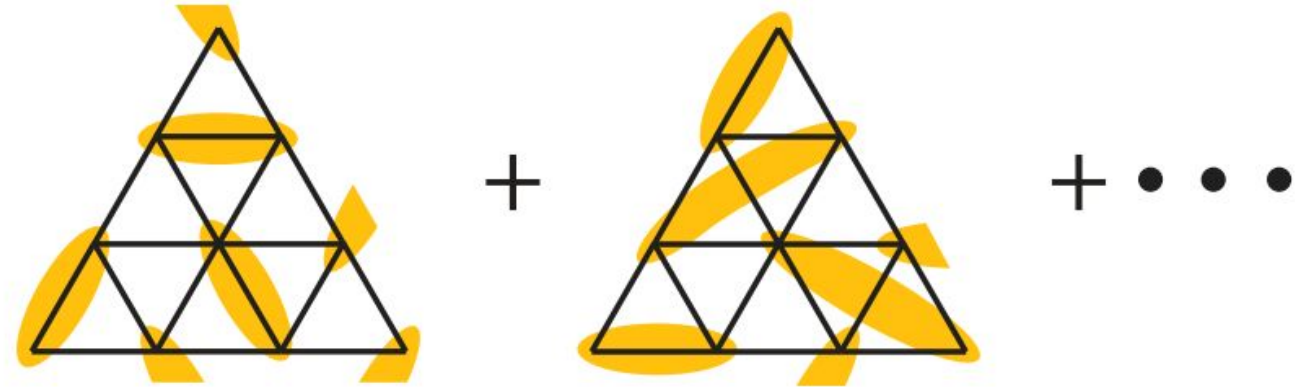
Kagome lattice

Imai, T., & Lee, Y. S. (2016). *Physics Today*, 69(8), 30–36.

# Quantum spin liquids

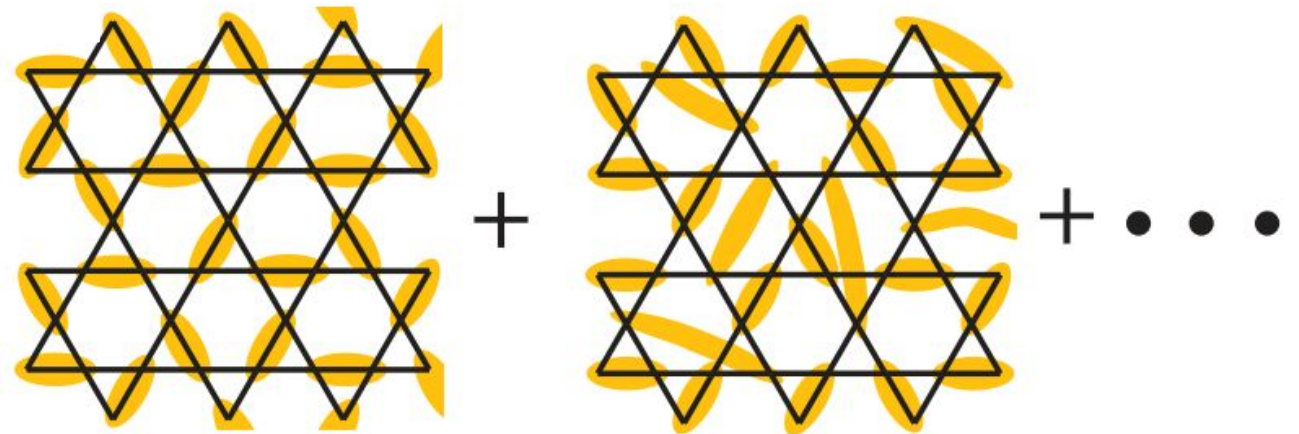
Properties:

- No magnetic order, even at  $T = 0$
- Liquid-like fluctuations at all  $T$
- Long range entanglement
- Fractionalized excitations



Quantum spin:  $S = \frac{1}{2}$

→ Spin singlets

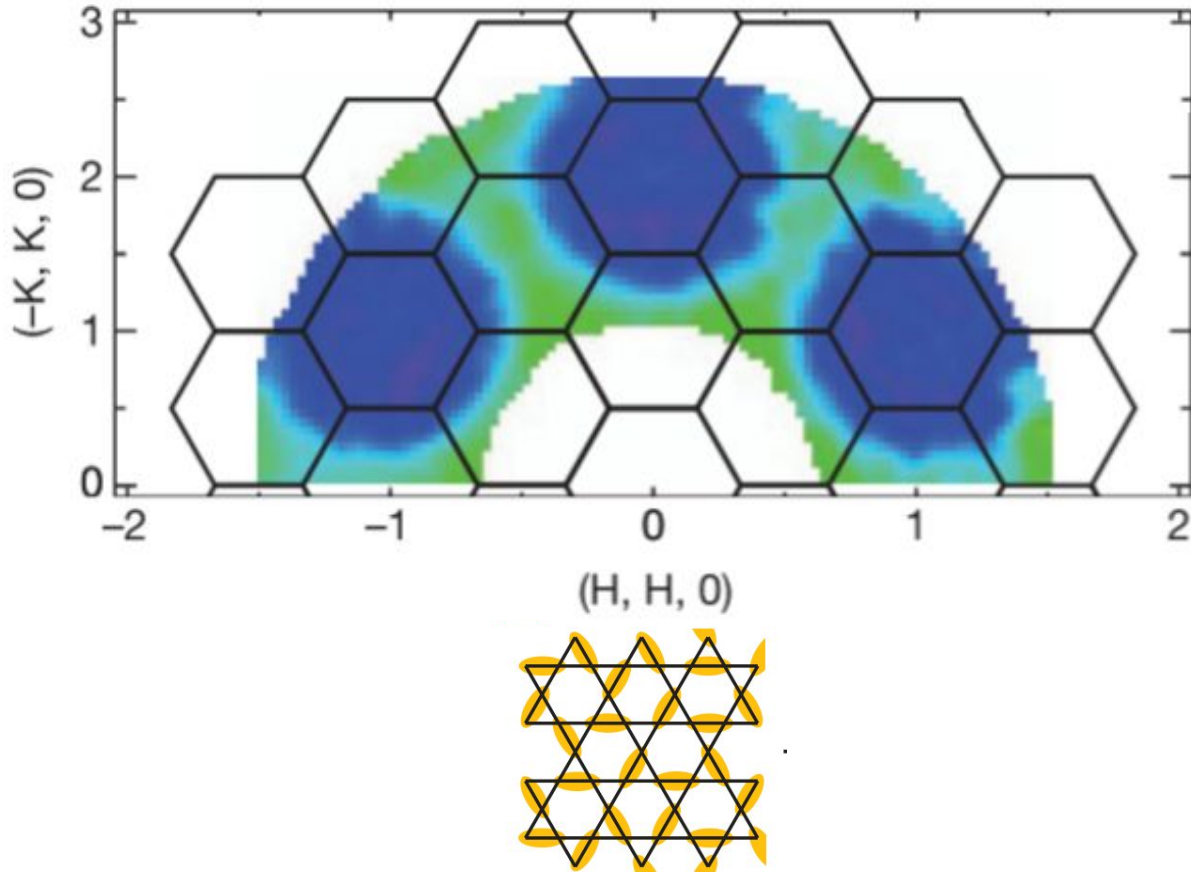




# Quantum spin liquids

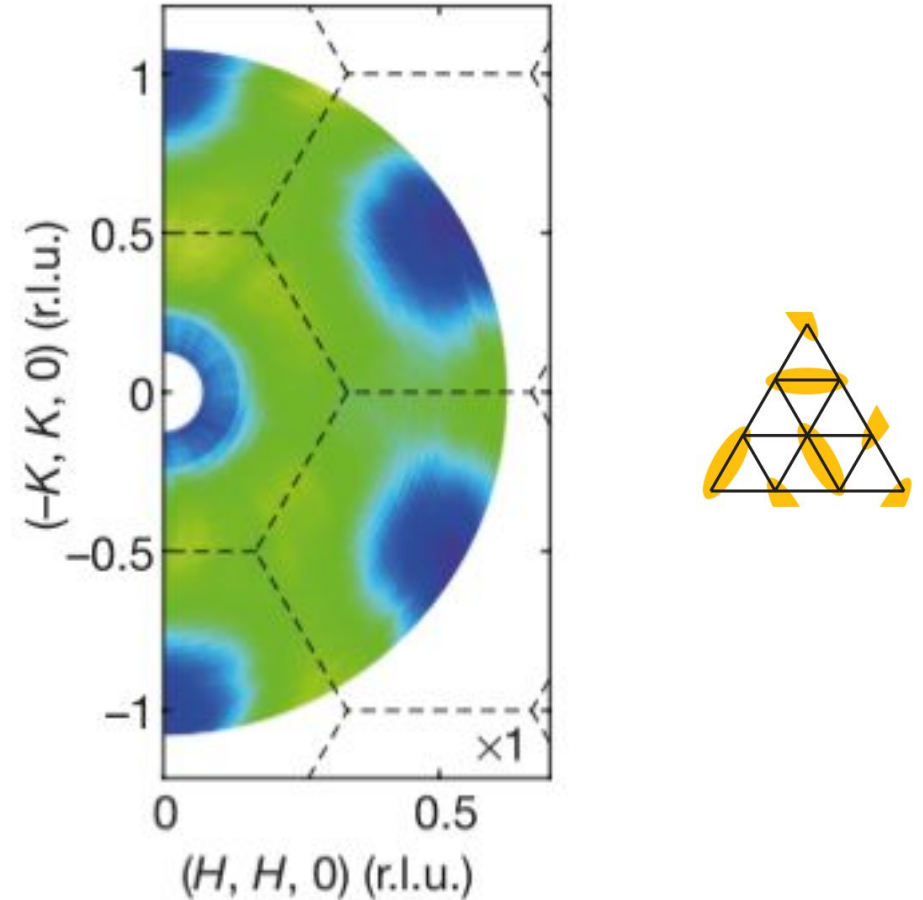
Herbertsmithite: spin- $\frac{1}{2}$  kagome

$\hbar\omega = 1 - 9 \text{ meV}$ ,  $T = 1.5 \text{ K}$



$\text{YbMgGaO}_4$  : spin- $\frac{1}{2}$  Triangular lattice

$\hbar\omega = 0.6 \text{ meV}$ ,  $T = 70 \text{ mK}$



Imai, T., & Lee, Y. S. (2016). *Physics Today*, 69(8), 30–36.  
 Shen, Y., et al (2016). *Nature*, 540(7634), 559–562.  
 Han, T. H., et al (2012). *Nature*, 492(7429), 406–410.

# Classical spin liquids

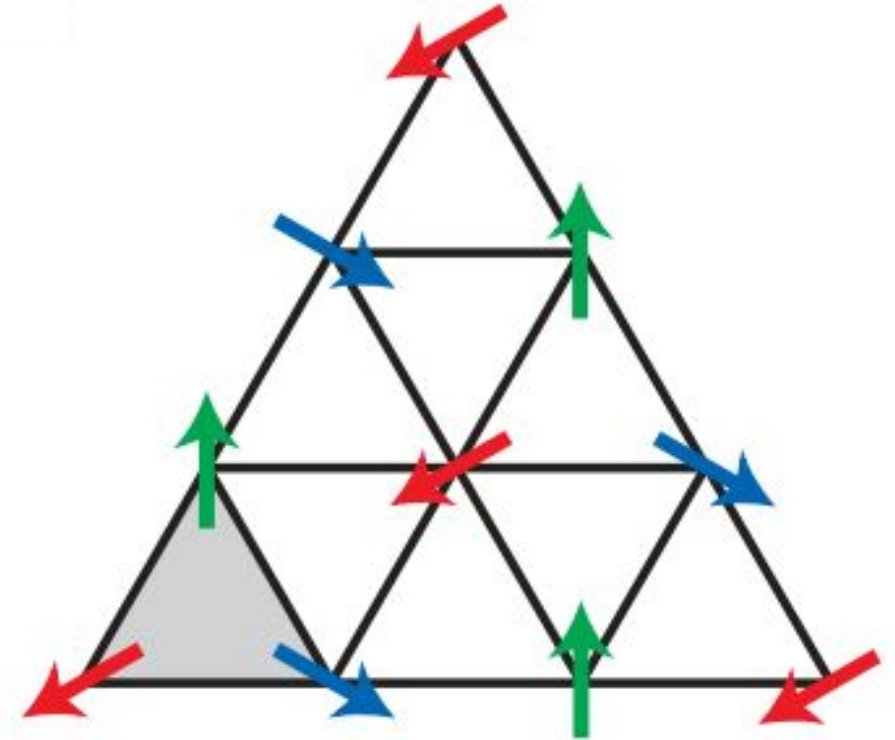
- Frustrated magnets
- Classical spin ( $S > 1/2$ )
- Finite ordering temperature

Above  $T_N$ :

- Broad excitations: short-ranged and finite lifetimes
- Low-energy excitations

However:

Definition not agreed upon!



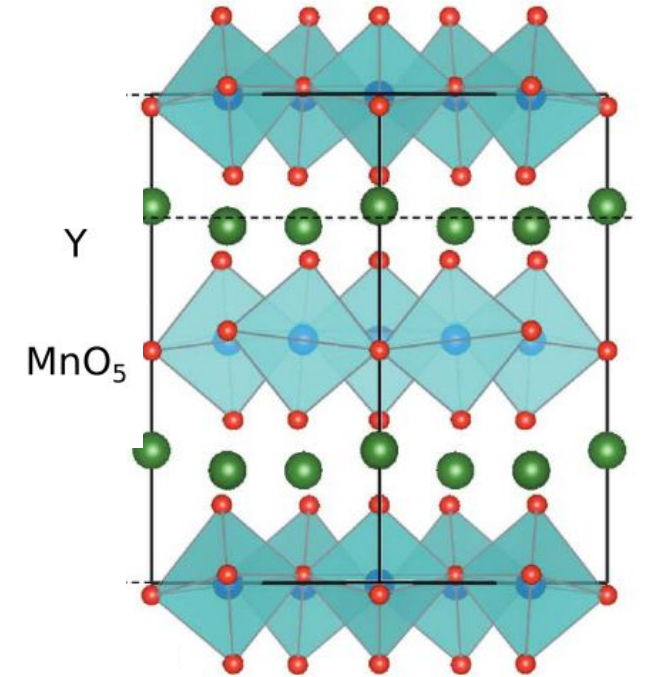
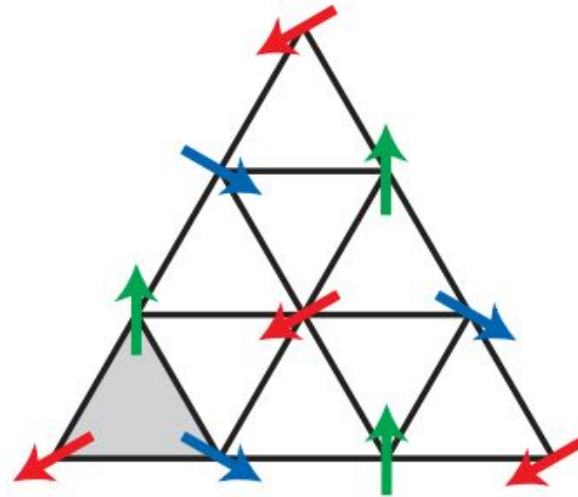
# Classical spin liquid phase in $\text{YMnO}_3$

# Hexagonal $\text{YMnO}_3$

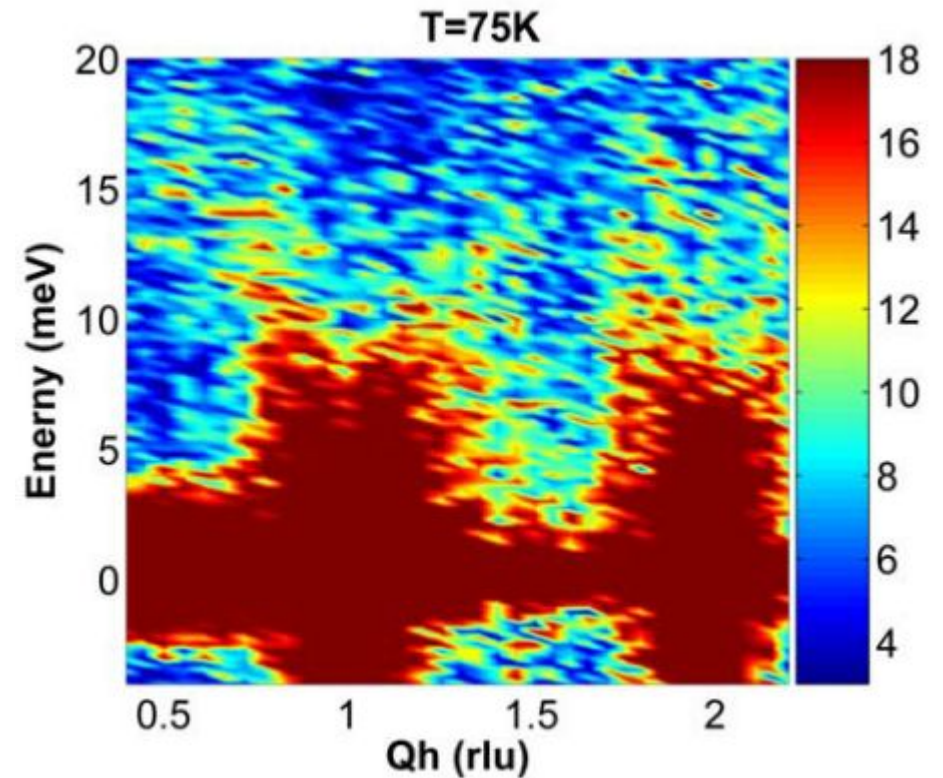
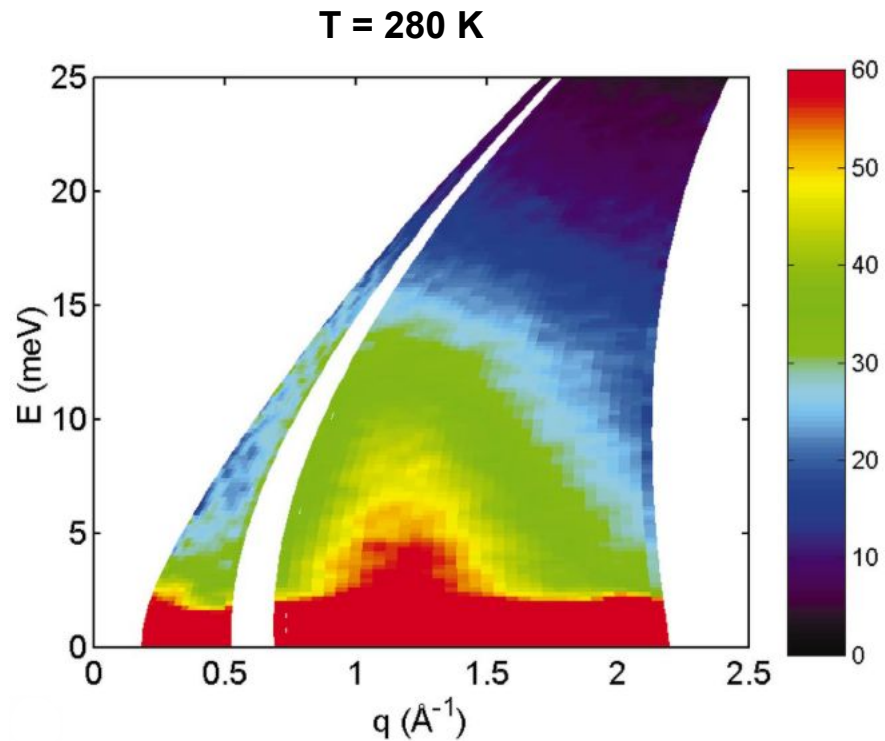
Frustrated, triangular lattice Heisenberg AFM  
Magnetic  $\text{Mn}^{3+}$  :  $S = 2$

Ferroelectric transition:  $T_C = 1259 \text{ K} \approx 1000 \text{ }^\circ\text{C}$

Antiferromagnetic ordering:  $T_N = 72 \text{ K} \approx -200 \text{ }^\circ\text{C}$



# Diffuse scattering in $\text{YMnO}_3$



Park, J., et al (2003). *Physical Review B*, 68, 104426.

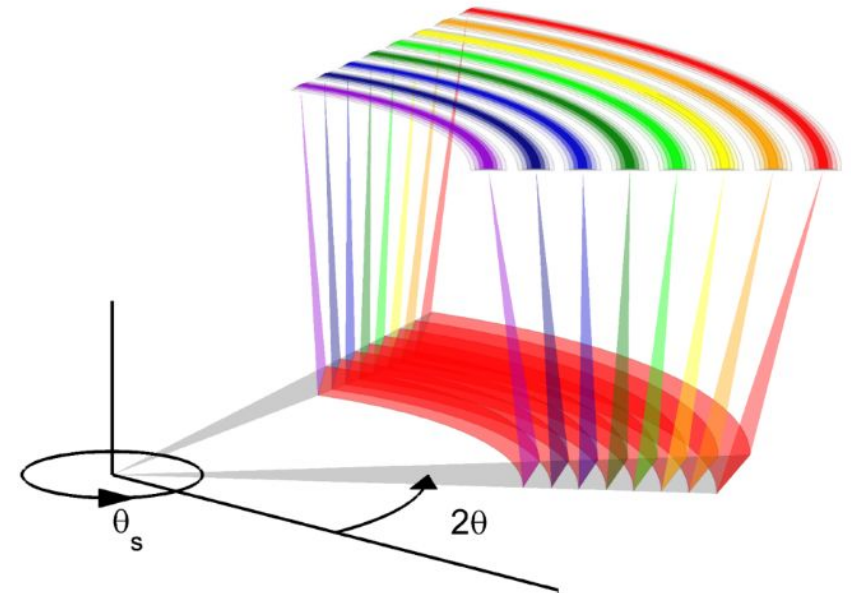
Demmel, F., & Chatterji, T. (2007). *Physical Review B*, 76(21), 212402.

# Experimental

CAMEA - multiplexing spectrometer, newly commissioned  
Paul Scherrer Institute, Switzerland



Similar to BIFROST @ ESS:  
Multiple analysers & detectors

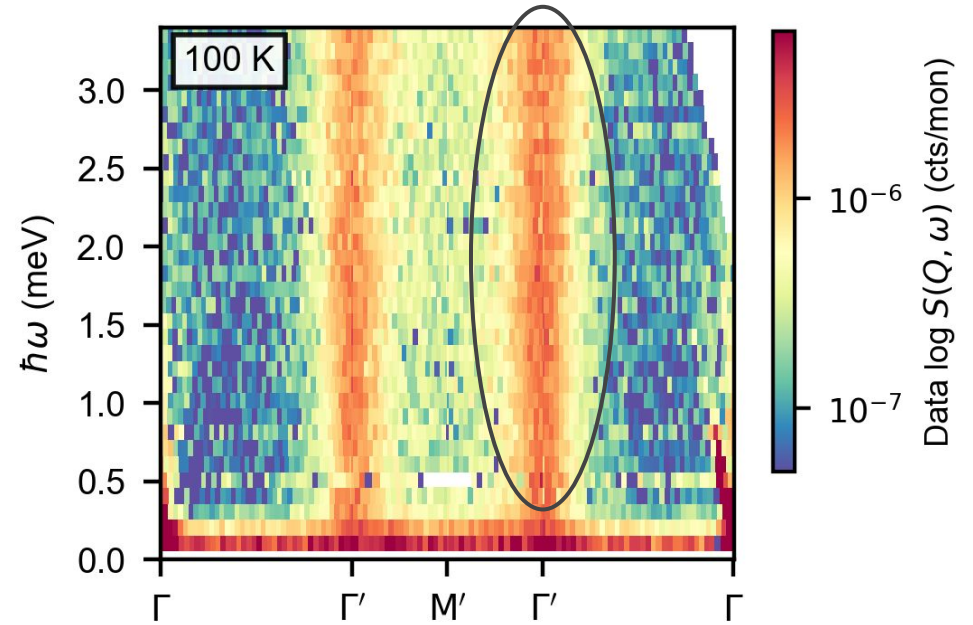
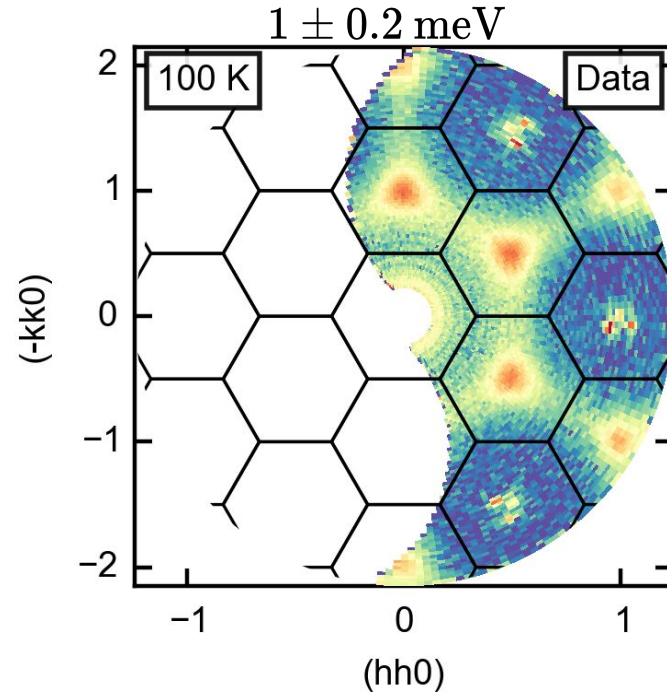


# Dynamic, diffuse scattering

$$T_N = 72 \text{ K}$$

$$\theta_{CW} = -550 \text{ K}$$

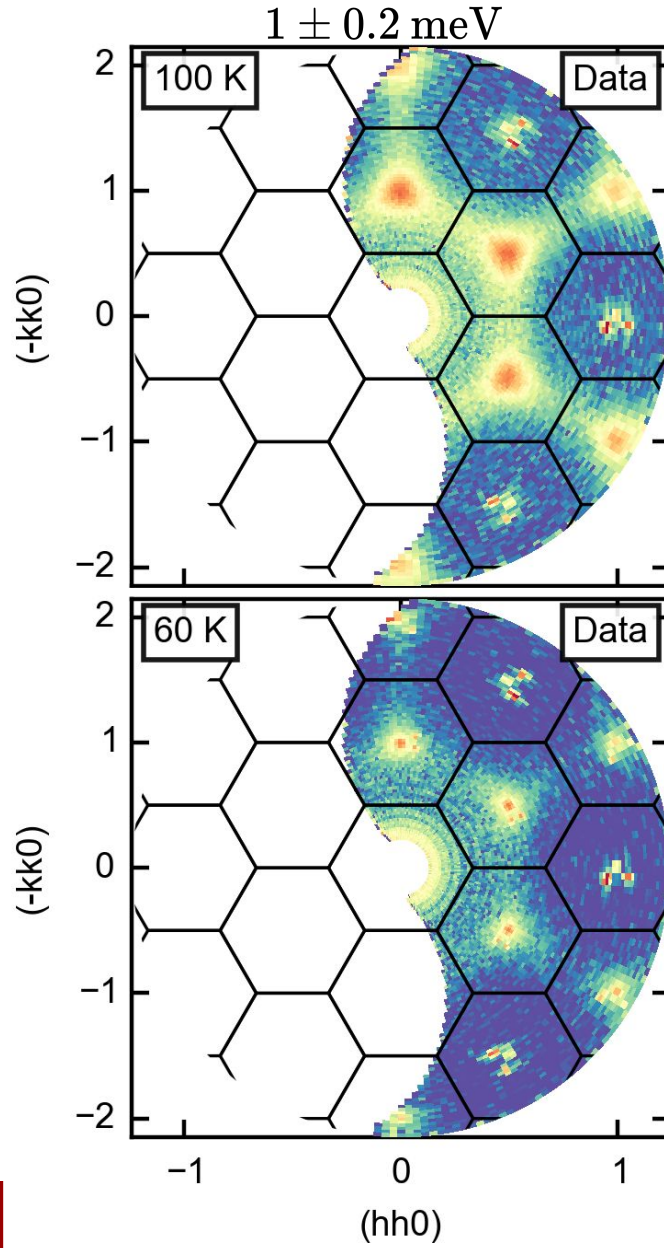
$$T > T_N$$



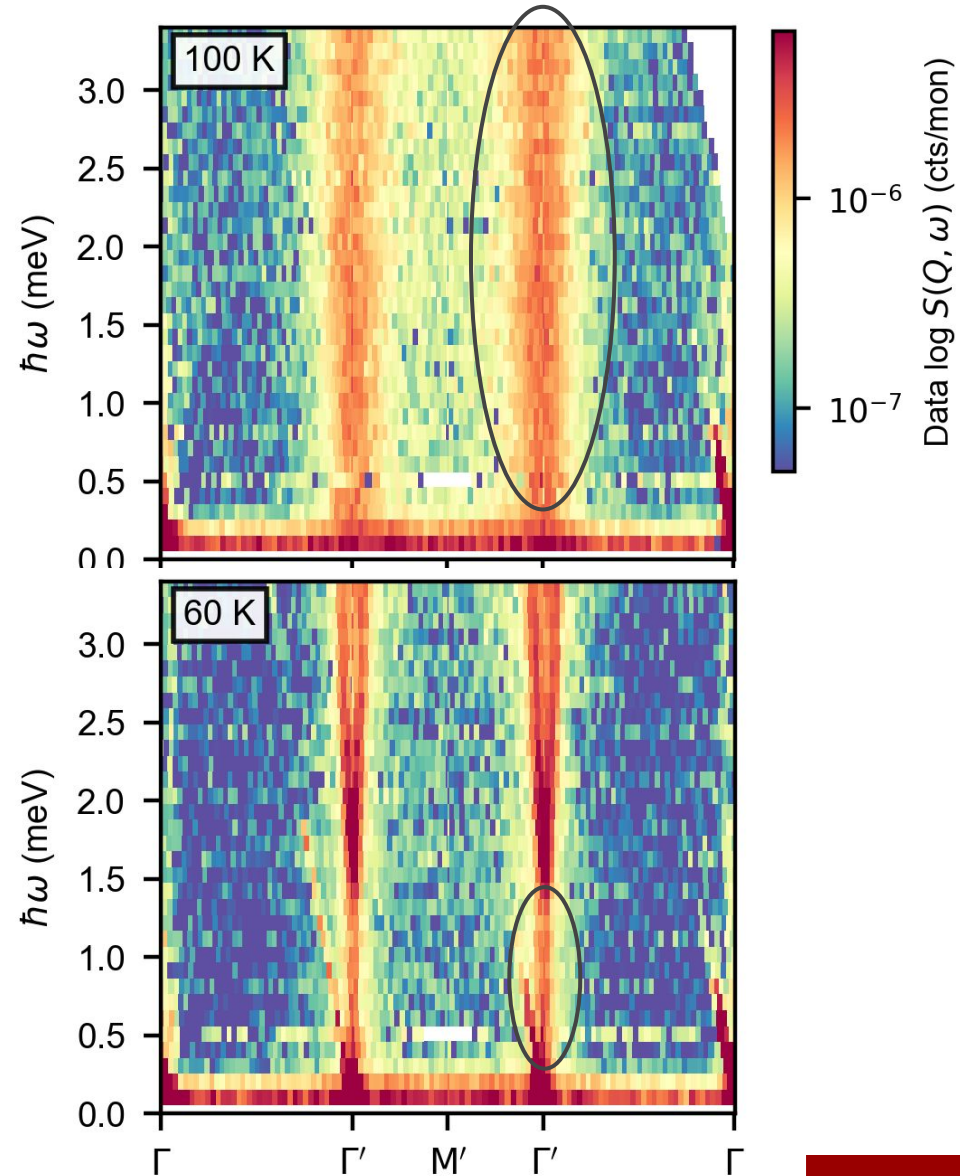
# Dynamic, diffuse scattering

$T_N = 72\text{ K}$

$T > T_N$



$T < T_N$



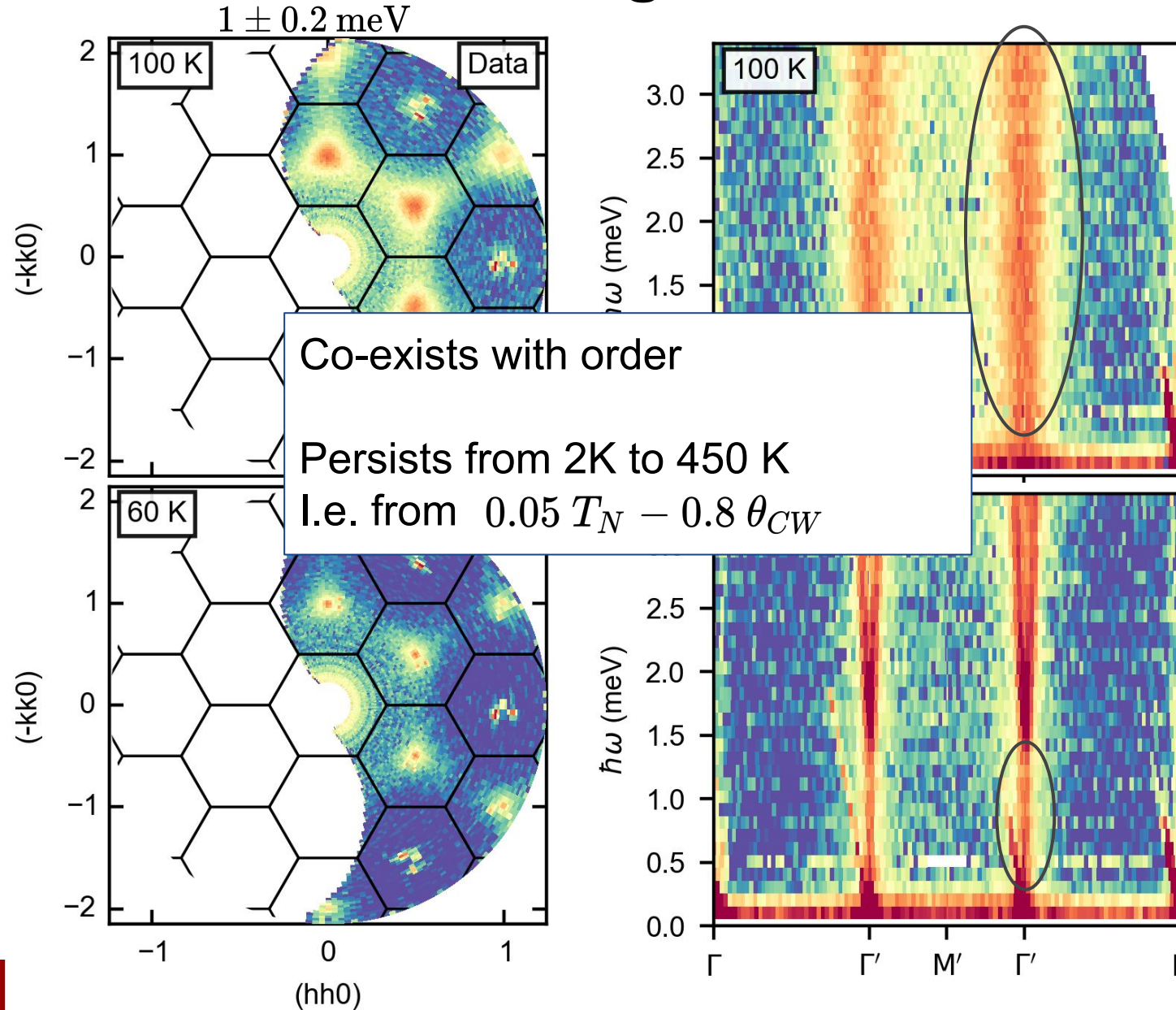


# Dynamic, diffuse scattering

$$T_N = 72 \text{ K}$$

$$\theta_{CW} = -550 \text{ K}$$

$$T > T_N$$



$$T < T_N$$

# Critical scattering near phase transitions

Fluctuations on all length and time scales

→ Diffuse scattering:  
Short-ranged correlations  
Quasielastic profile

Critical spin-spin correlation

$$\langle S_0 S_l \rangle \propto \exp(-|\mathbf{r}_{0l}|/\xi)$$



# Critical scattering near phase transitions

Fluctuations on all length and time scales

→ Diffuse scattering:  
Short-ranged correlations  
Quasielastic profile

Critical spin-spin correlation

$$\langle S_0 S_l \rangle \propto \exp(-|\mathbf{r}_{0l}|/\xi)$$

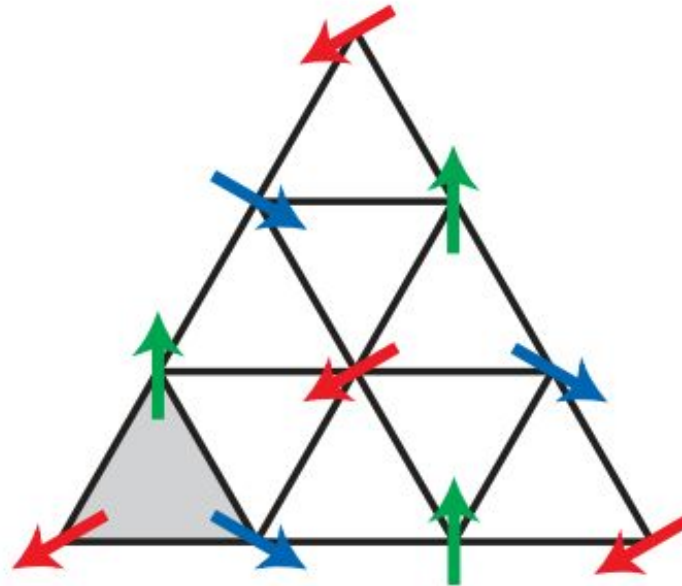
Correlation length:  
Measurable quantity!



# Modelling: structure factor of spin clusters

$$S(\mathbf{Q}) = f(Q)^2 \sum_{l=0}^N e^{i\mathbf{Q}\cdot\mathbf{r}_l} \sum_{\substack{\{\alpha,\beta\} \\ \in\{x,y,z\}}} (\delta_{\alpha,\beta} - \hat{Q}_\alpha \hat{Q}_\beta) \langle S_0^\alpha S_l^\beta \rangle_{\text{Rot}}$$

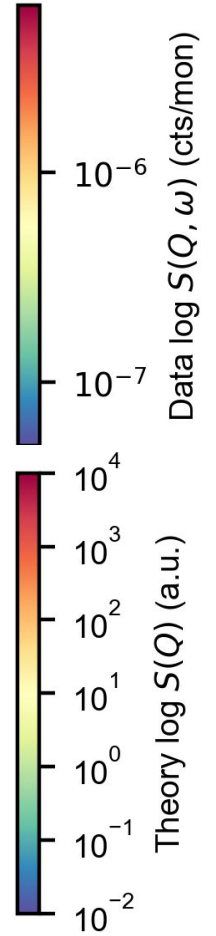
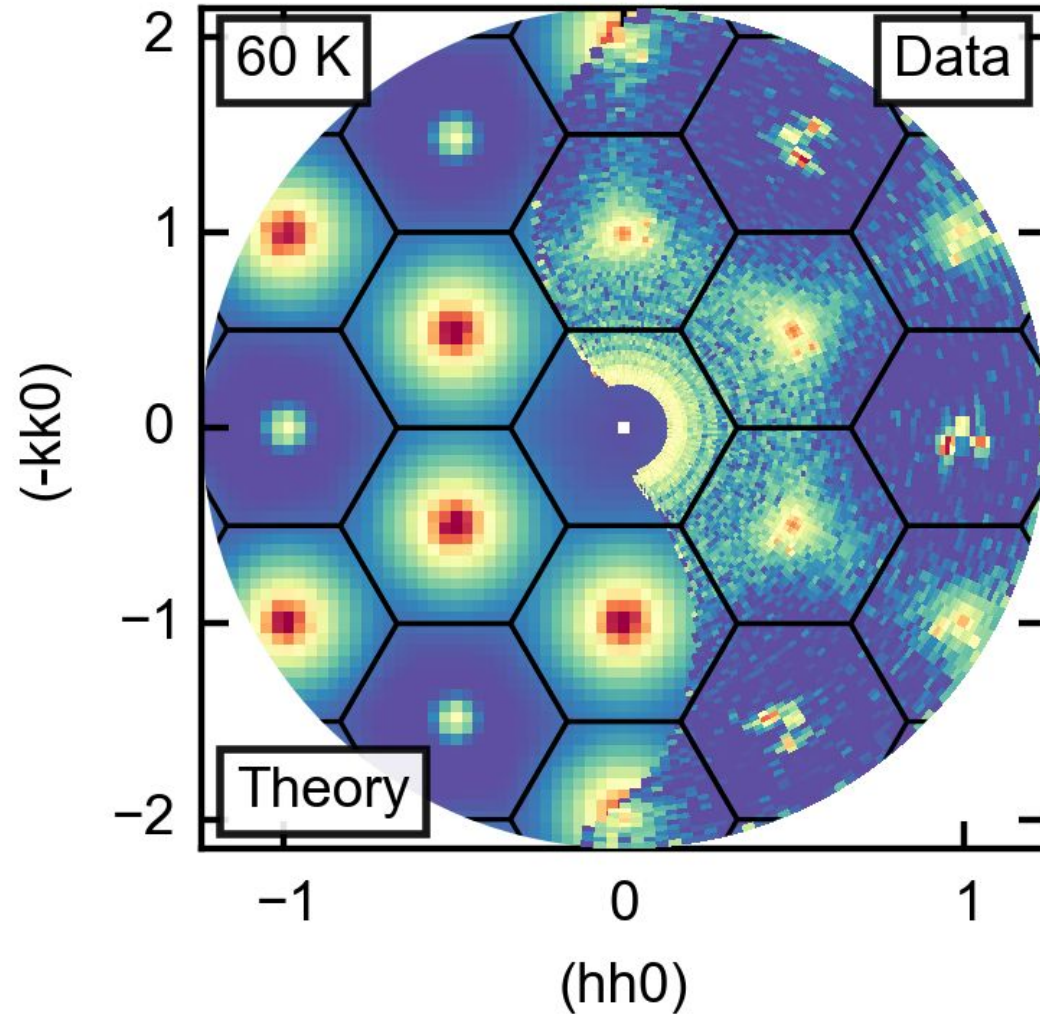
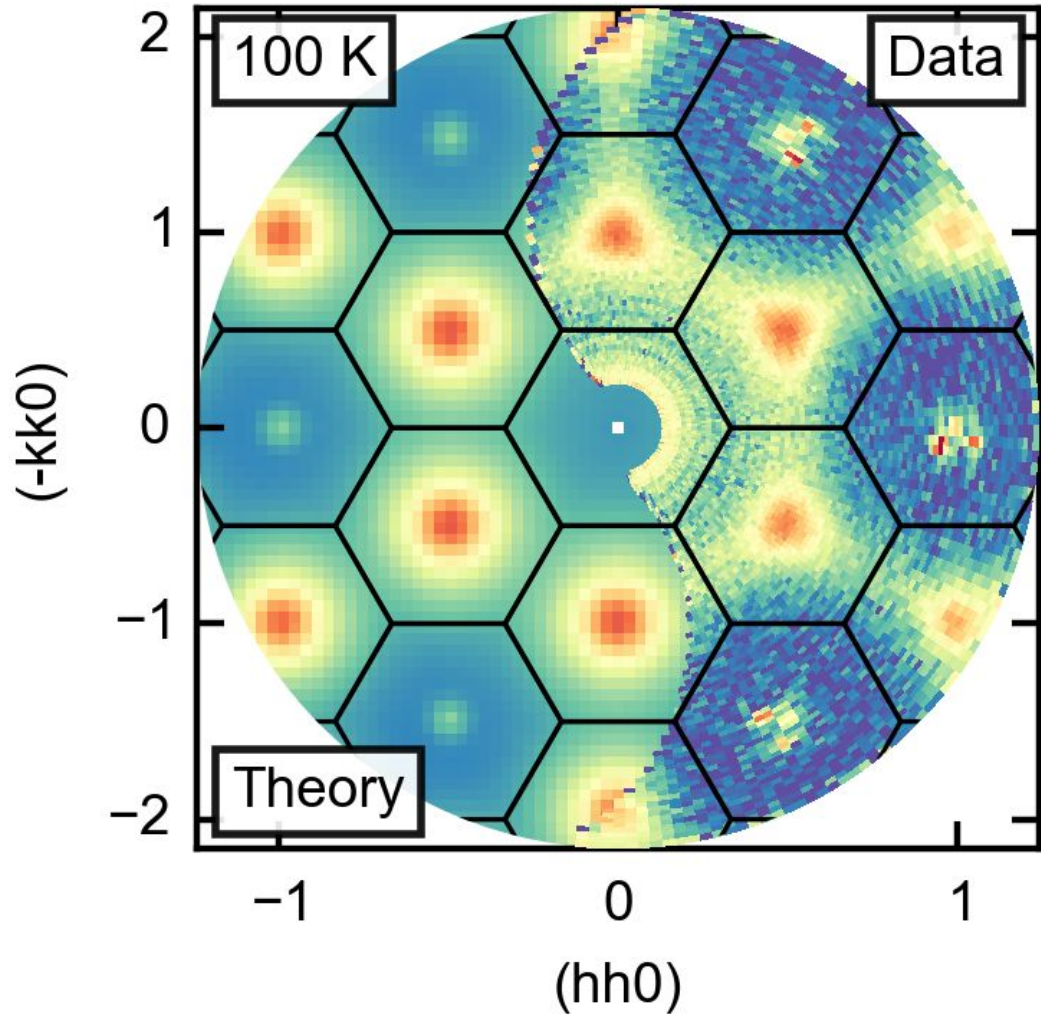
$$\langle S_0 S_l \rangle \propto \exp(-|\mathbf{r}_{0l}|/\xi)$$



# Dynamic, diffuse scattering

$T > T_N$

$T < T_N$



**Classical Spin Liquid or Extended Critical Range in  $h$ -YMnO<sub>3</sub>?**

Sofie Janas<sup>1,2</sup>, Jakob Lass<sup>1,3</sup>, Ana-Elena Țuțueanu<sup>1,4</sup>, Morten L. Haubro<sup>1</sup>, Christof Niedermayer<sup>3</sup>, Uwe Stuhr<sup>3</sup>,  
Guangyong Xu<sup>5</sup>, Dharmalingam Prabhakaran<sup>6</sup>, Pascale P. Deen<sup>7,1</sup>, Sonja Holm-Dahlin<sup>1,8</sup> and Kim Lefmann<sup>1,\*</sup>

<sup>1</sup>*Nanoscience Center, Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen Ø, Denmark*

<sup>2</sup>*Institute of Physics, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*

<sup>3</sup>*Laboratory for Neutron Scattering, Paul Scherrer Institute (PSI), 5232 Villigen, Switzerland*

<sup>4</sup>*Institut Laue-Langevin (ILL), Grenoble Cedex 9 38042, France*

<sup>5</sup>*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA*

<sup>6</sup>*Clarendon Laboratory, Department of Physics, University of Oxford, Oxford OX1 3PU, United Kingdom*

<sup>7</sup>*European Spallation Source ESS ERIC, Box 176, SE-221 00 Lund, Sweden*

<sup>8</sup>*ISIS Facility Rutherford Appleton Laboratory Chilton, Didcot, OX 11 0QX, United Kingdom*



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Neutron spectroscopy on the classical triangular-lattice frustrated antiferromagnet  $h$ -YMnO<sub>3</sub> reveals diffuse, gapless magnetic excitations present both far below and above the ordering temperature. The correlation length of the excitations increases as the temperature approaches zero, bearing a strong resemblance to critical scattering. We model the dynamics in the ordered and correlated disordered phase as critical spin correlations in a two-dimensional magnetic state. We propose that our findings may provide a general framework to understand features often attributed to classical spin liquids.

DOI: 10.1103/PhysRevLett.126.107203

Link:

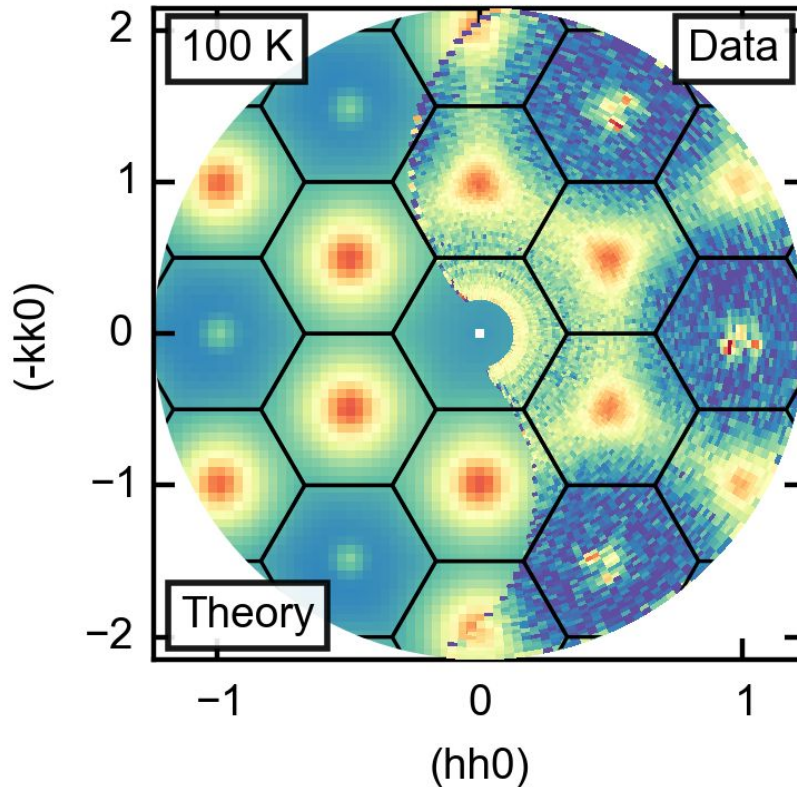
<https://doi.org/10.1103/PhysRevLett.126.107203>

# So what are classical spin liquids really?

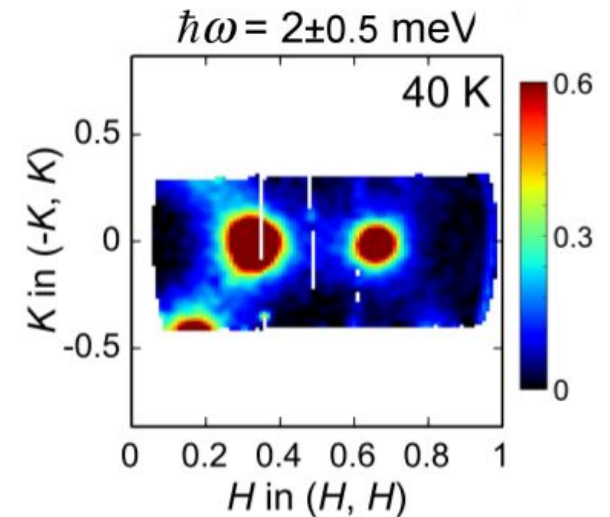
# Classical spin liquids and critical scattering

Model: Critical scattering in vastly extended critical region due to frustration

YMnO<sub>3</sub>  
S = 2  
Triangular lattice

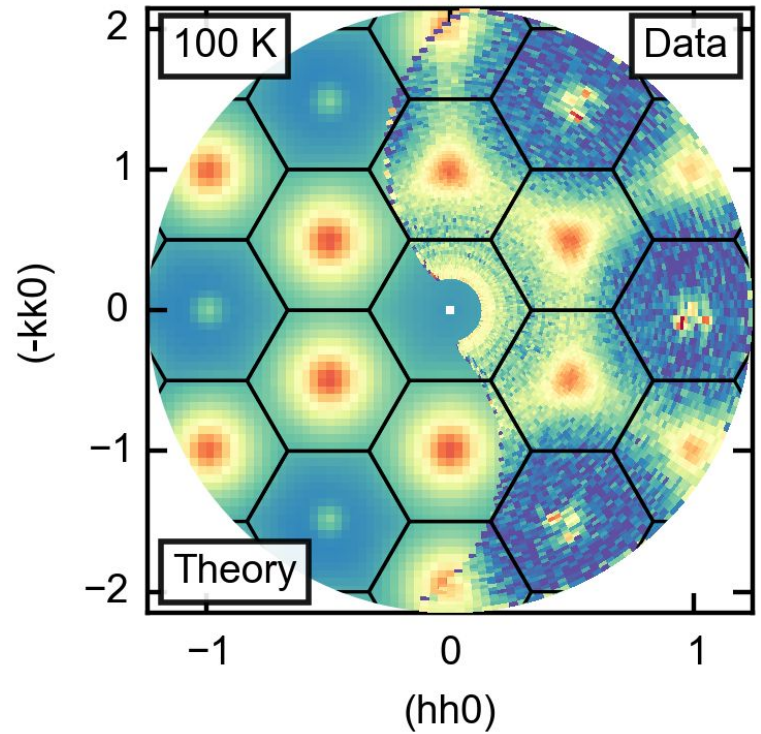


CuCrO<sub>2</sub>:  
S = 3/2  
Triangular lattice



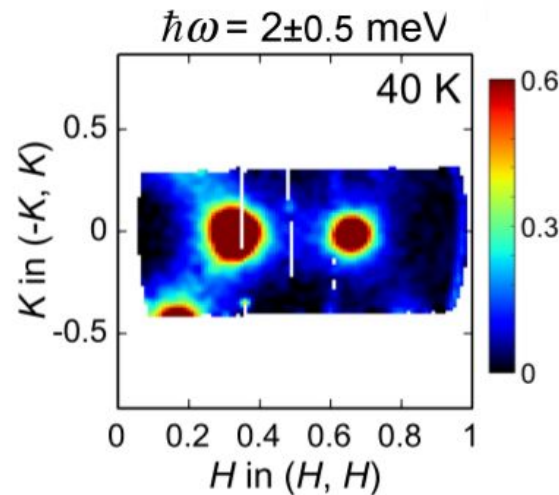


# Telling classical and quantum spin liquids apart

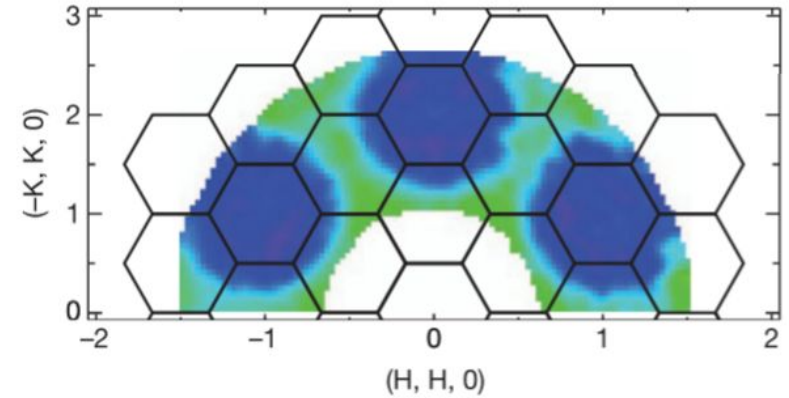


YMnO<sub>3</sub>  
 $S = 2$   
 Triangular lattice

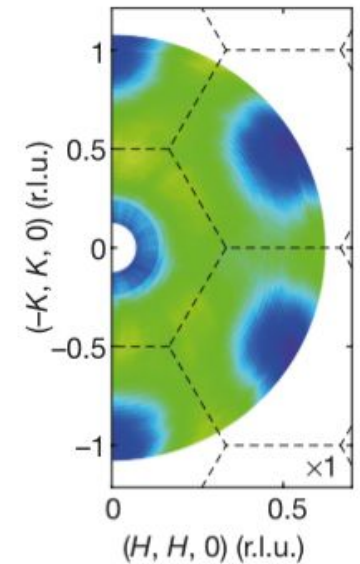
CuCrO<sub>2</sub>:  
 $S = 3/2$   
 Triangular lattice

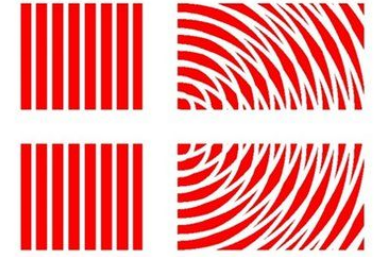


Herbertsmithite:  $S = 1/2$ , kagome lattice



YbMgGaO<sub>4</sub> :  
 $S = 1/2$   
 Triangular lattice





**DanScatt**

# Thanks!

PhD Supervisor  
Kim Lefmann



Collaborator  
Pascale P. Deen



PAUL SCHERRER INSTITUT

