Los Alamos
NATIONAL LABORATORY

Neutron scattering: an experimental perspective (with side-notes on history and philosophy)

Allen Scheie

6/25/2025

LA-UR-25-25909

Outline:

1. History of neutron scattering
2. Modern neutron scattering
3. Current uses of neutron scattering: exploring the unknown

Part 1:
History of neutron scattering

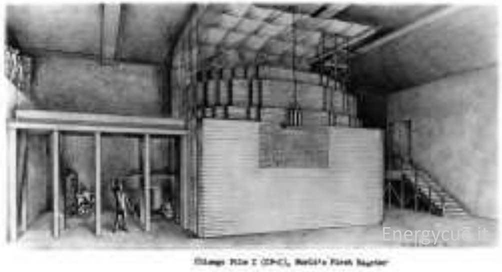
1939: USA begins secret project to research nuclear bomb

ahf.nuclearmuseum.org

Osti.gov

Nuclear reactors built to produce plutonium

Chicago pile 1 (1942)



0.5 W

Oak Ridge X-10 (1943)

1 MW

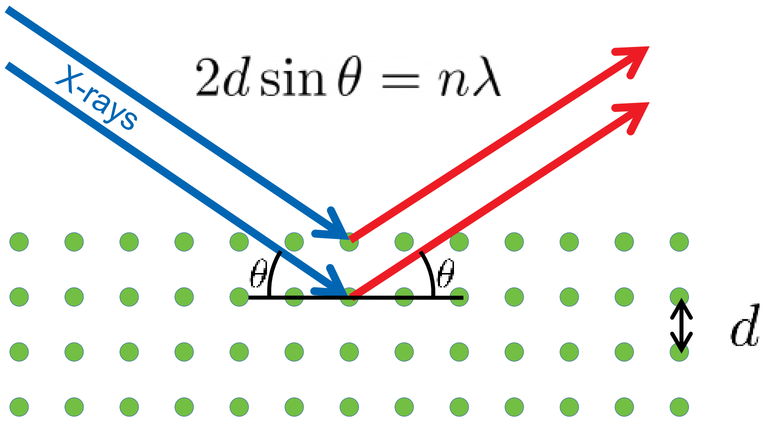
Hanford B (1944)

250 MW

After the war, X-10 kept running: what else can we do with a nuclear reactor?

One idea: crystallography

- Von Laue & Paul Ewald: X-ray diffraction on copper sulfate (1912)
  - Technique refined by William Braggs (Nobel Prize 1915), Scherrer, Debye, Hull, etc.



- Moderated neutrons have a similar wavelength to X-rays (by DeBroglie's relation)

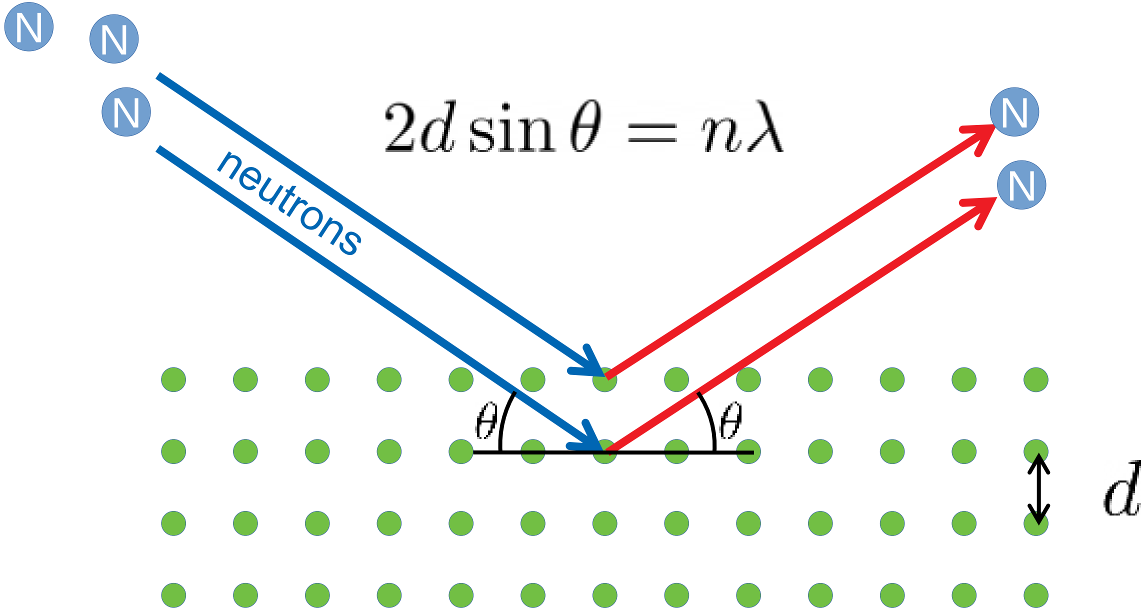
Thermal neutron wavelength:

$$\lambda = 1.8\ \text{Å}$$

Close to typical atom spacing in a crystal

Los Alamos NATIONAL LABORATORY

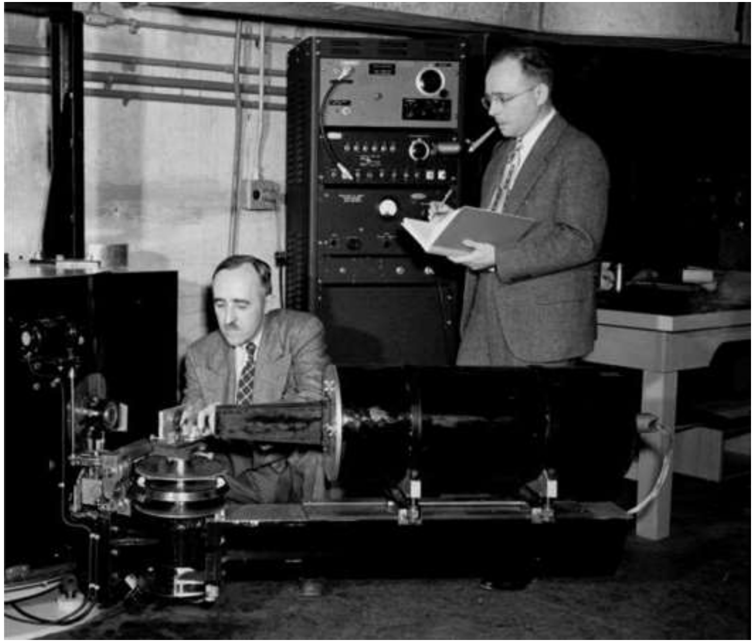
1944: Ernest Wollan measures neutron diffraction on gypsum and NaCl (table salt)



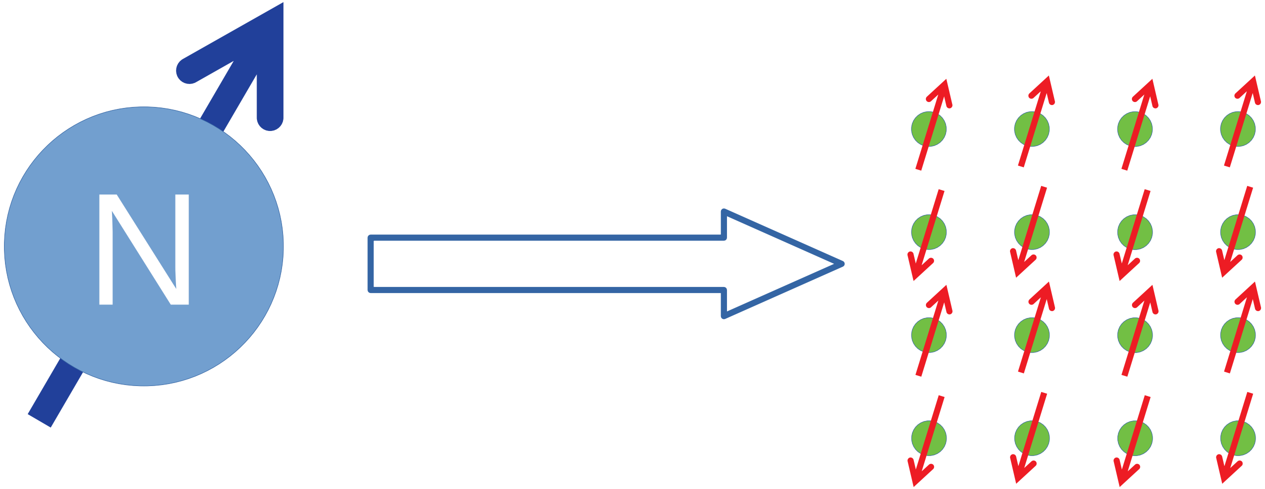
1946: Shull joins Wollan to pursue technique

X-10 site (now ORNL)

Clifford Shull and Ernest Wollan
at the X-10 graphite reactor



Neutrons are chargeless (pass through materials easily) but magnetic: so they scatter off magnetic patterns



1949: neutrons reveal antiferromagnetism in MnO

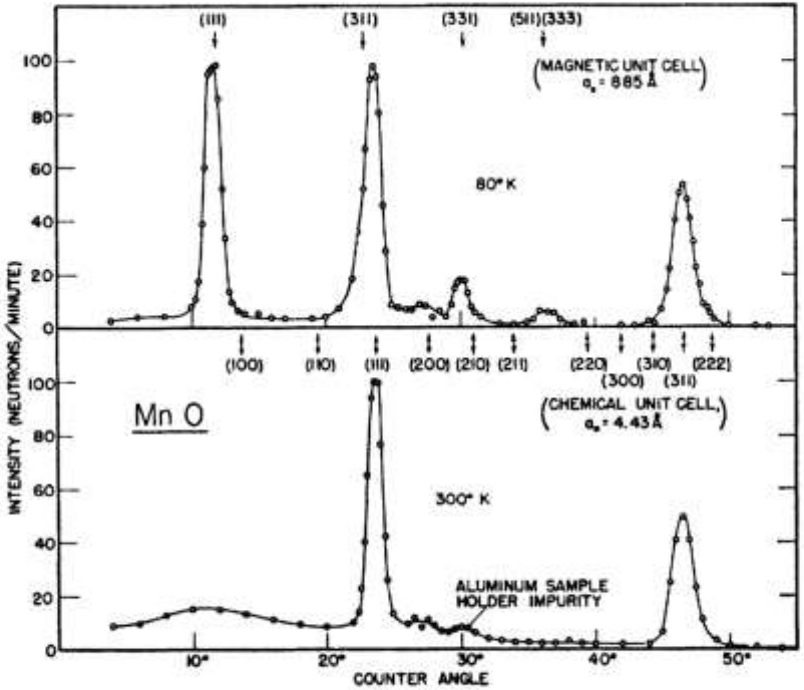
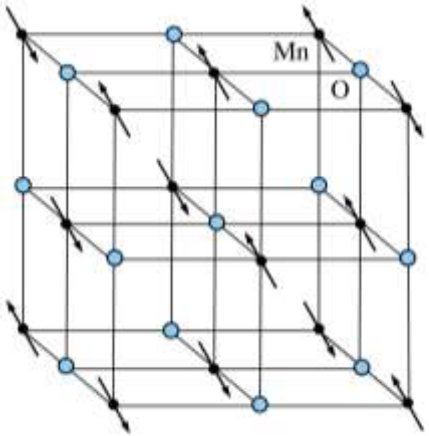


FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.



Shull & Smart, *Phys. Rev.* (1949)

Philosophical interlude:

Antiferromagnetism was first clear example of *spontaneous symmetry breaking*

- Global symmetry preserved, local symmetry broken

- Only occurs in thermodynamic limit (large system size)
  - Quantum theory predicts a superposition of two AFM states:

$$\frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\uparrow\downarrow\ldots\rangle \pm |\downarrow\uparrow\downarrow\uparrow\ldots\rangle\right)$$

  - Seems to be related to decoherence?

- Now invoked all over physics (e.g. QCD)

Back to the physics.

Neutrons vs. X-rays: neutrons have much smaller energy at the same wavelength

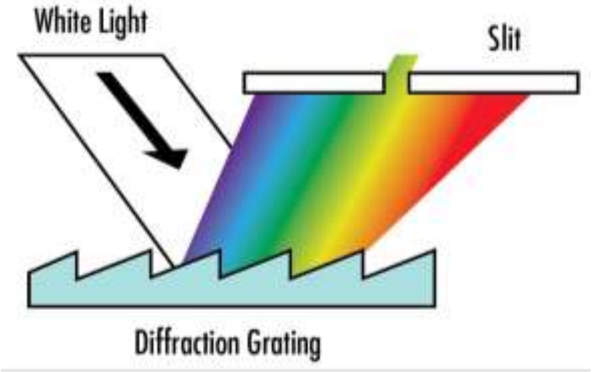
Thermal neutron energy (λ=1.8 Å):		X-ray energy (λ=1.8 Å):
25 meV	10^5 difference	6.9 keV

Can energy resolve to measure low-energy features?

Bertram Brockhouse: builds triple-axis spectrometer in 1955 at Chalk River reactor in Canada

Cins.ca

crystal diffraction acts like a diffraction grating

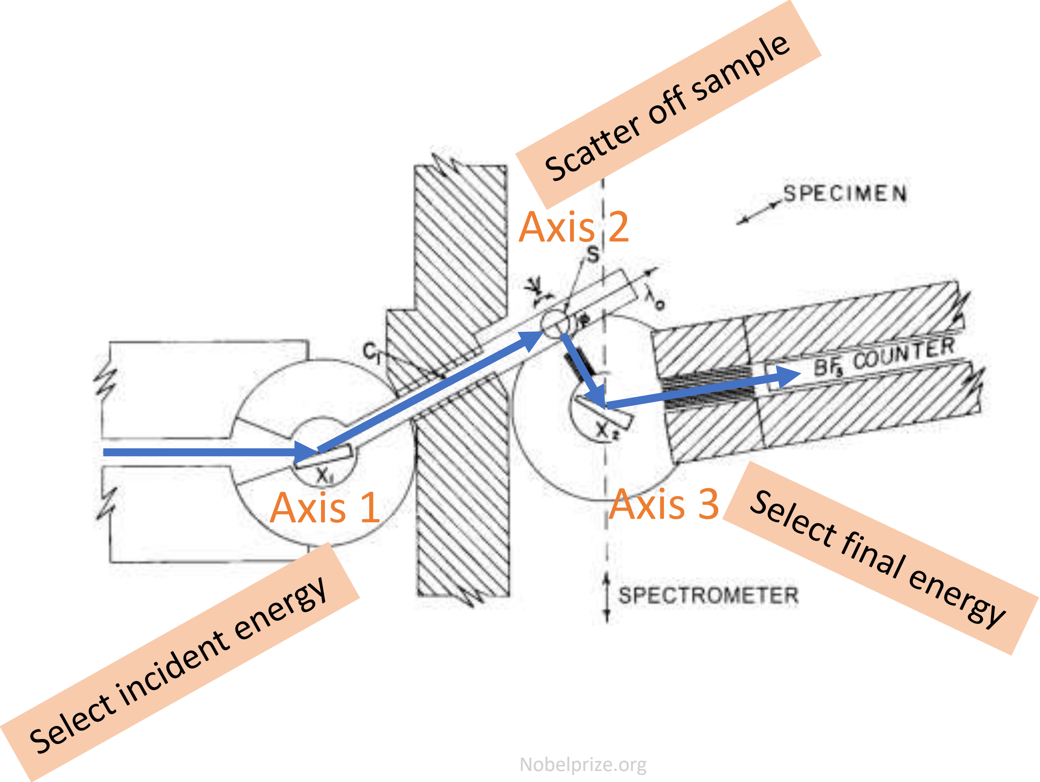


Selecting a wavelength = selecting an energy

$$\lambda = \frac{h}{mv} \qquad E = \frac{1}{2}mv^2$$

Bertram Brockhouse: builds triple-axis spectrometer in 1955 at Chalk River reactor in Canada

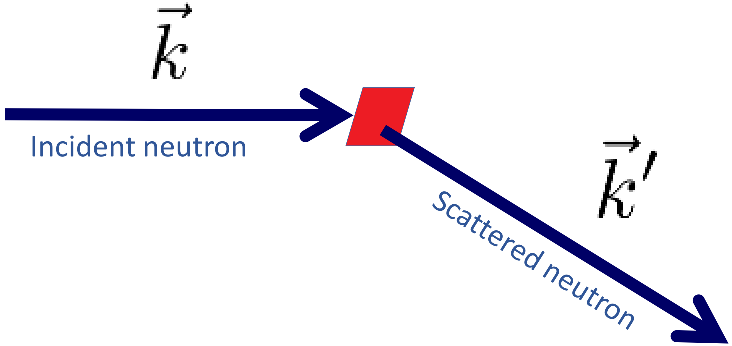
Cins.ca



Nobelprize.org

Conservation of momentum and energy:

$$\vec{k} = \frac{2\pi}{\lambda}\hat{\lambda}$$



Momentum:

$$\vec{Q} = \vec{k} - \vec{k}'$$

$$\vec{k} = \frac{m\vec{v}}{\hbar} \qquad \vec{Q} = \frac{1}{\hbar}(m\vec{v} - m\vec{v}')$$

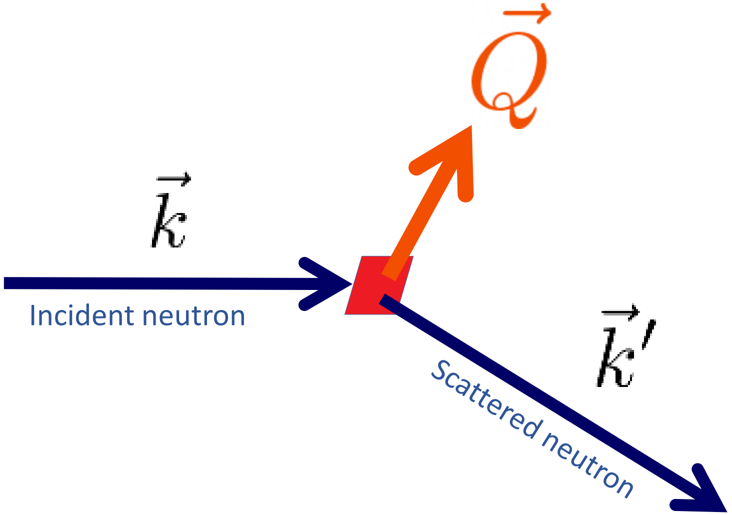
Energy:

$$\Delta E = E_1 - E_2$$

also written

$$\hbar\omega = \frac{\hbar^2}{2m}(k^2 - k'^2)$$

Energy and momentum can be deposited in the sample, and it can be measured!



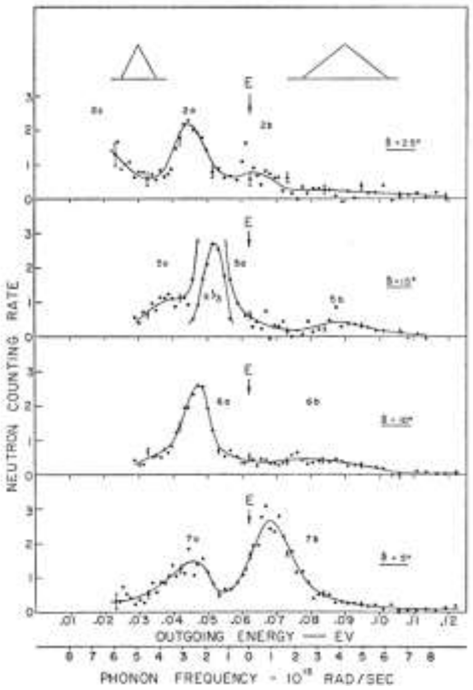
Momentum:

$$\vec{Q} = \vec{k} - \vec{k}'$$

Energy:

$$\hbar\omega = \frac{\hbar^2}{2m}(k^2 - k'^2)$$

Brockhouse measured a dispersion curve for the lattice vibrations in aluminum



Scattering of Neutrons by Phonons in an Aluminum Single Crystal

B. N. BROCKHOUSE AND A. T. STEWART

Physics Division, Atomic Energy of Canada, Limited, Chalk River, Ontario, Canada

(Received August 29, 1955)

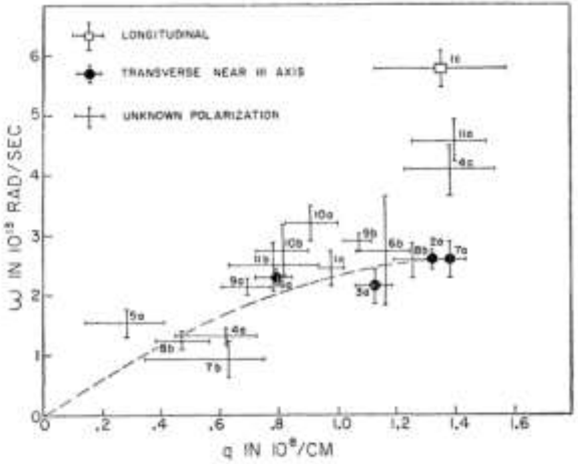
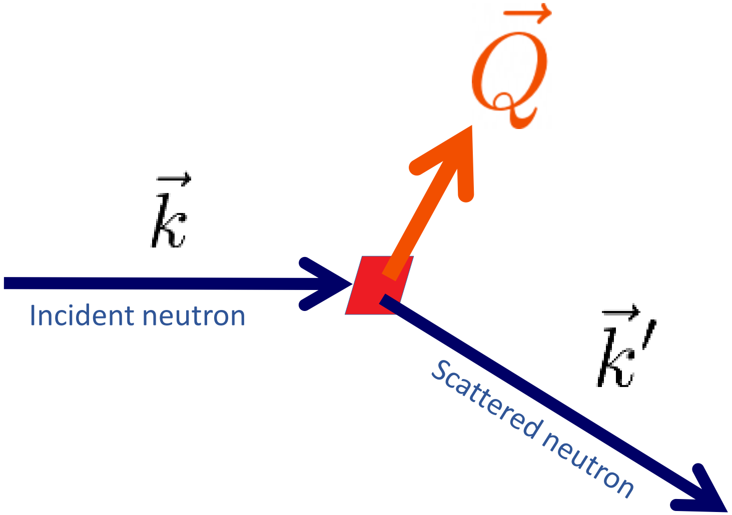
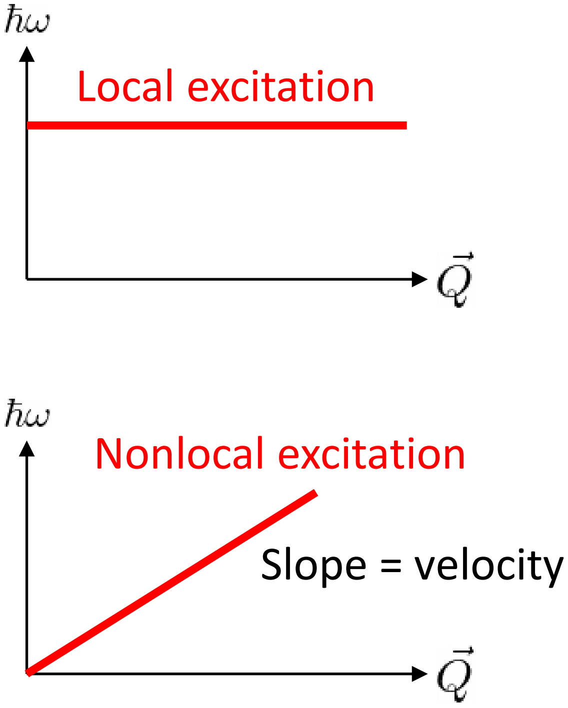


FIG. 3. Relation between ω and q for observed phonon groups, with estimated errors.

Dispersion curve: relation between energy and momentum





First direct evidence of “quasiparticles”

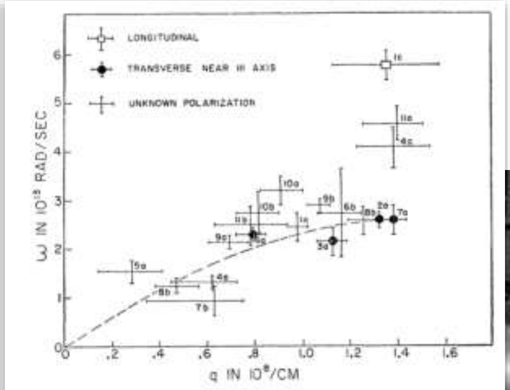
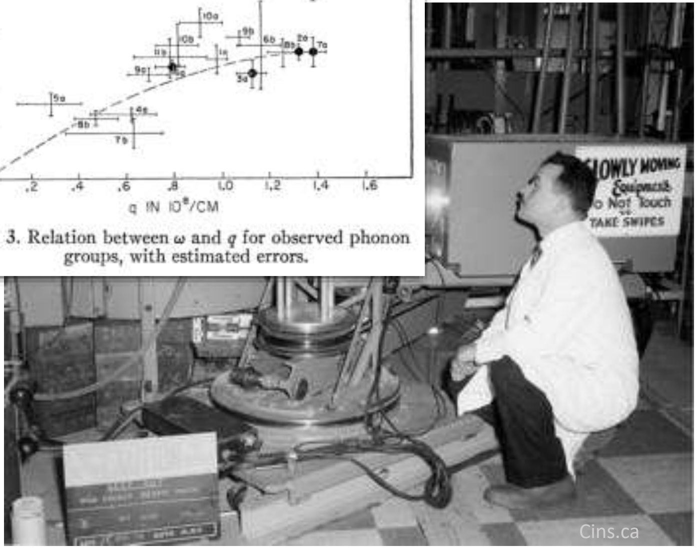


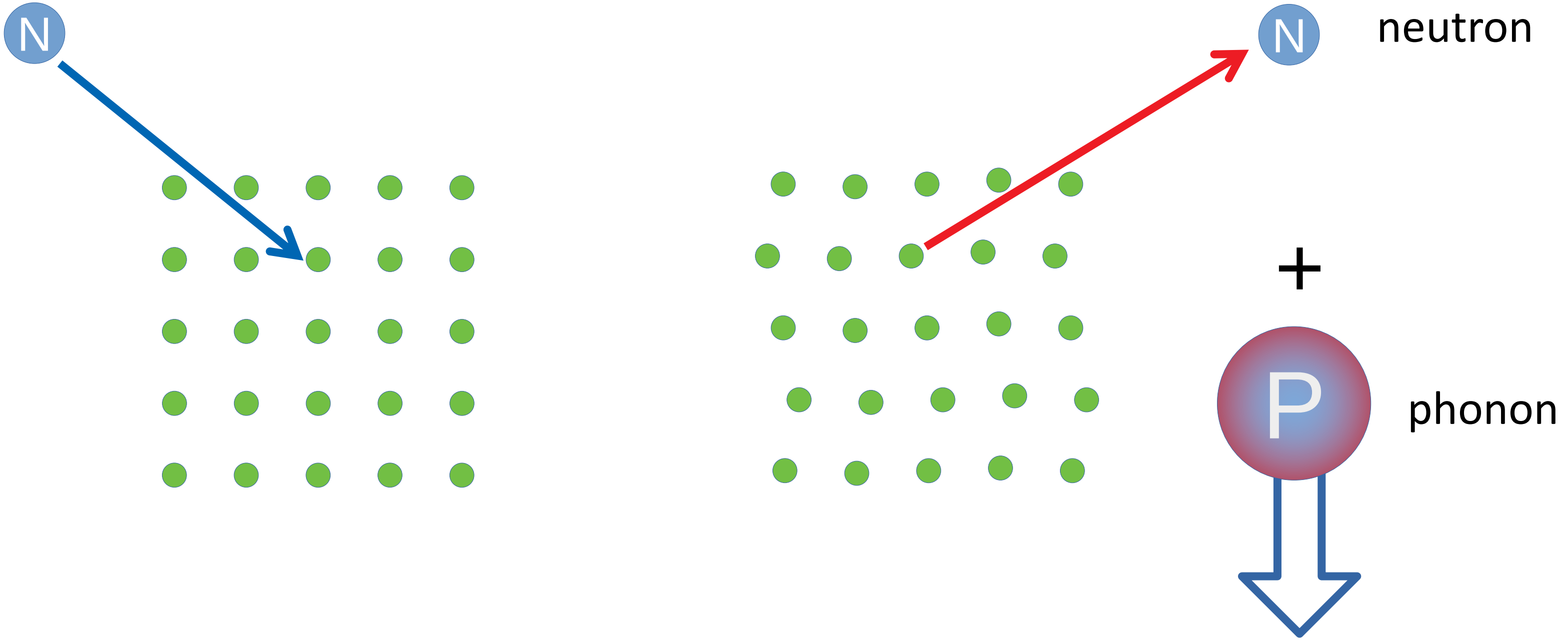
Fig. 3. Relation between ω and q for observed phonon groups, with estimated errors.



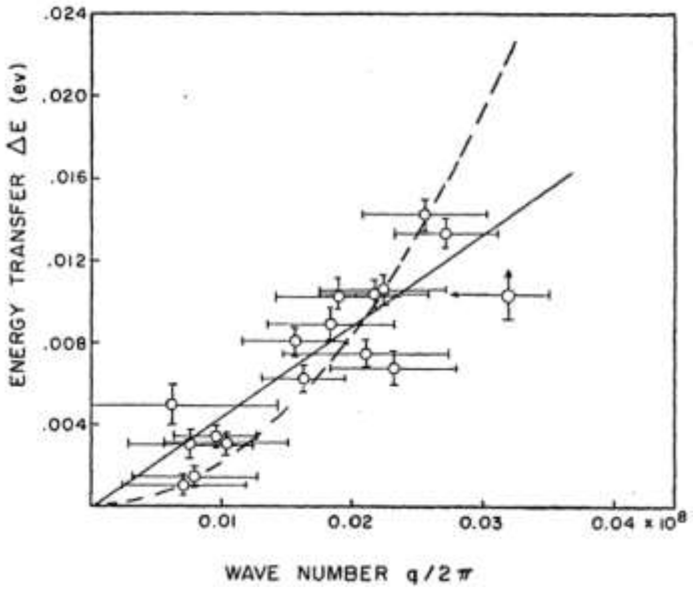
“The results permitted refutation of the simple models for interatomic forces then extant… these early results show surprisingly long-range behavior for the interatomic force system.”

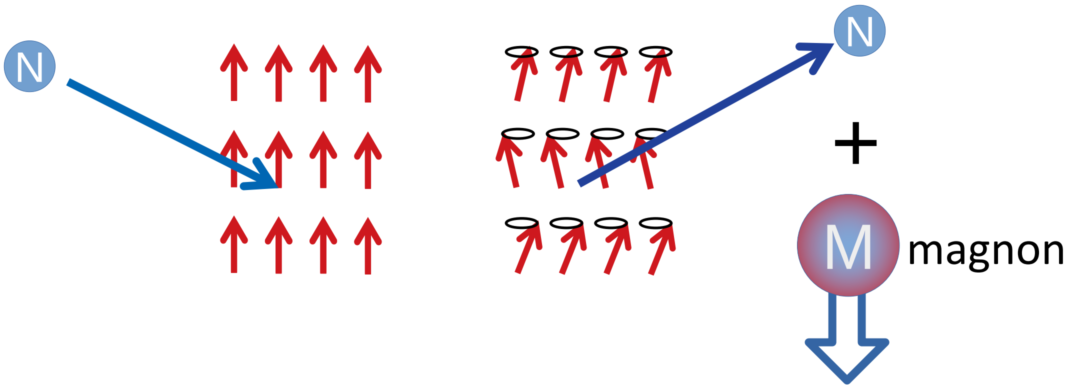
Brockhouse, Nobel Prize lecture (1995)

Phonons: collective wave of atomic vibrations, we use the mathematics of particle physics to describe it



Brockhouse also measured magnetic quasiparticles ("Magnons") in Fe_3O_4 (1957)

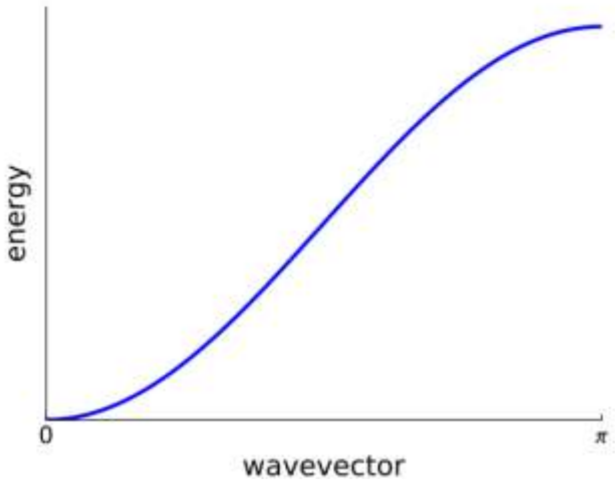




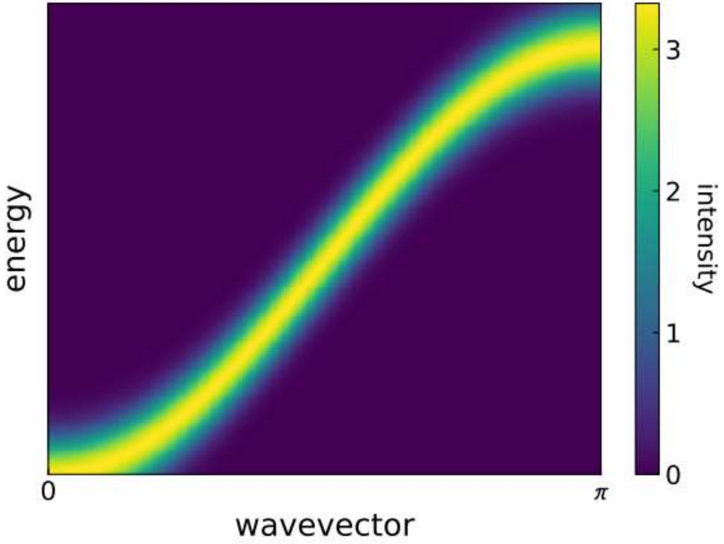
Brockhouse, Phys. Rev. **106**, 5 (1957)

Quasiparticles were originally considered a calculation tool; neutron scattering showed they are *real*

Theoretical dispersion



Measured neutron signal



Los Alamos NATIONAL LABORATORY

Philosophical interlude:

Are quasiparticles real?

- We treat these as “emergent reality”
  - Can take on properties that are forbidden for individual parts

Examples: quasiparticles, 2nd law of thermodynamics, organic life

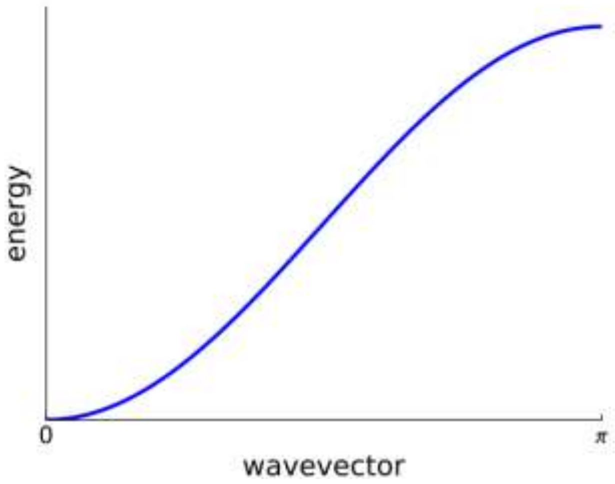
- Most of my colleagues (myself included) consider quasiparticles to be “real” objects.
- ALL our theories are incomplete approximations of underlying reality.
  - “quasiparticles” differ from “particles” in degree, not in kind.

“In quantum physics there is no logical way to distinguish a real particle from an excited state of the system that behaves like one. We therefore use the same word for both.”

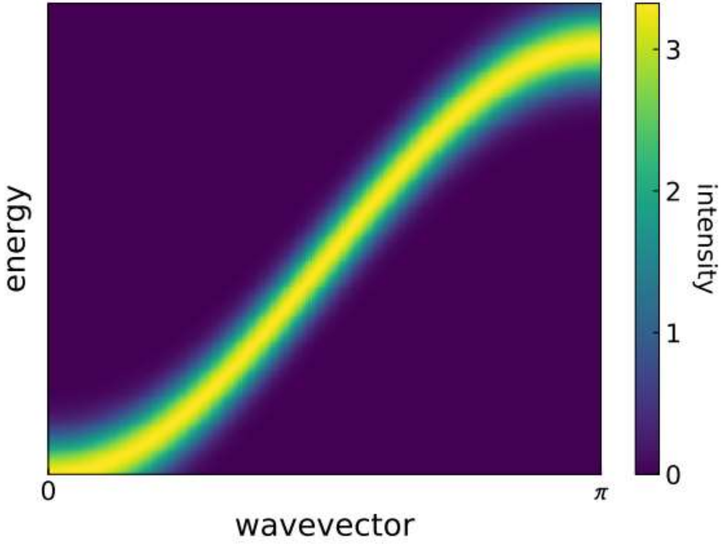
- R. Laughlin (Nobel Lecture, 1998)

Quasiparticles were originally considered a calculation tool; neutron scattering showed they are real

Theoretical dispersion



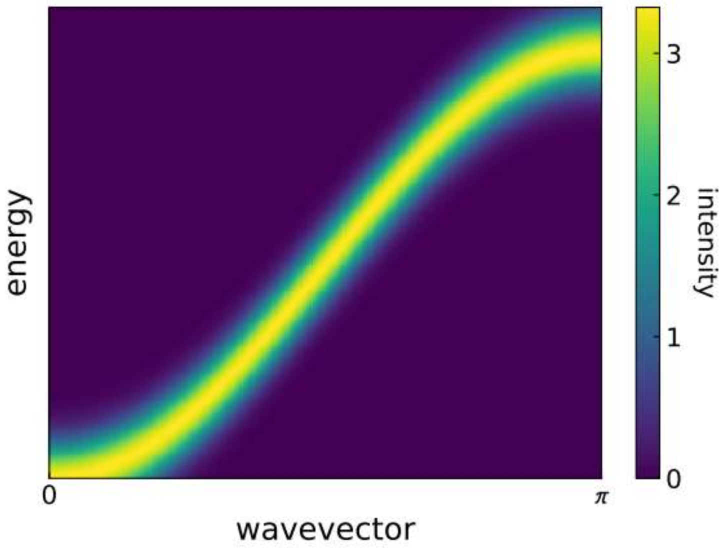
Measured neutron signal



Quasiparticles were originally considered a calculation tool; neutron scattering showed they are real

Scattered intensity is proportional to the **Fourier transform** of **spin correlation**

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \int \sum_l d^{-i\vec{Q}\cdot\vec{l}} \langle S_0(0) S_l(t) \rangle e^{-i\omega t} dt$$



Part 2:
What neutrons are used for now

Old... vs New

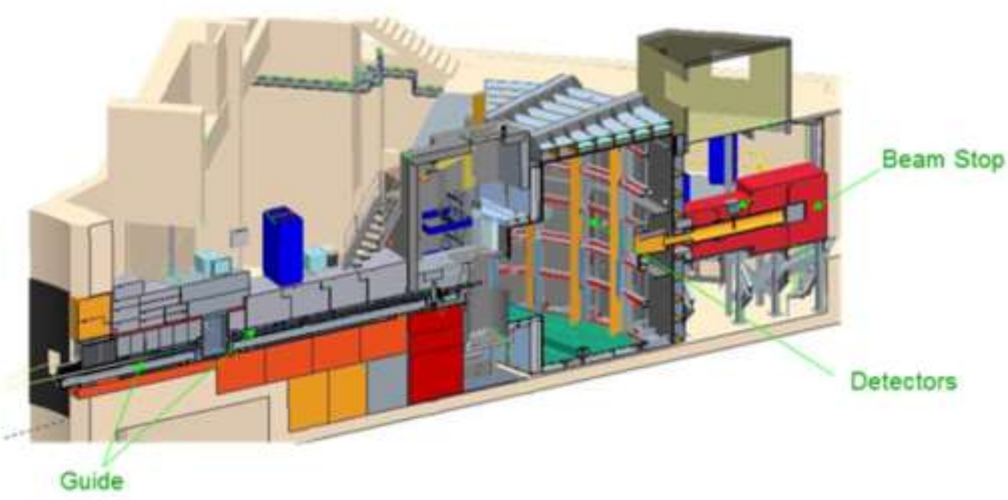
1 detector

SEQUOIA spectrometer, ORNL

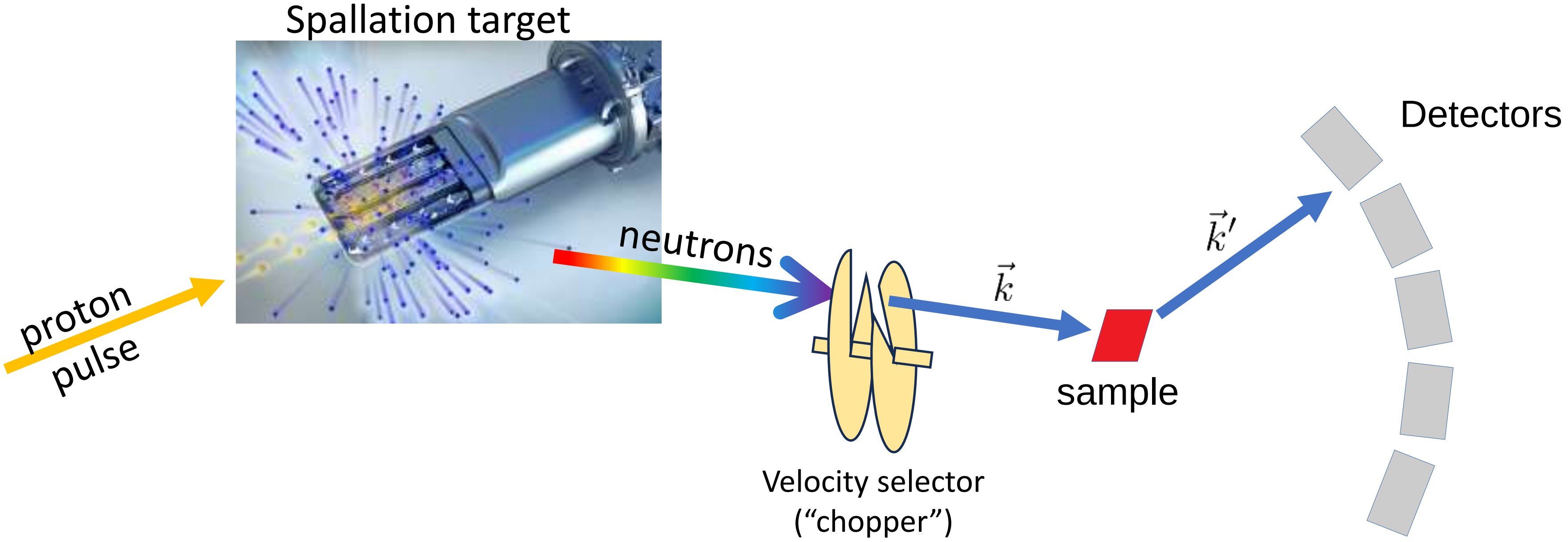
144,000 detector pixels

Time-of-flight technique: hit sample with a pulse of neutrons, and measure neutron velocity (more efficient)

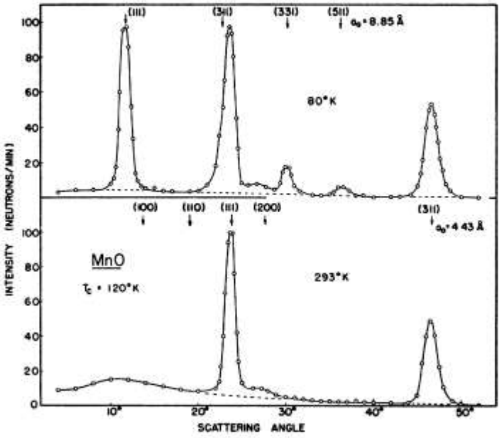
SEQUOIA spectrometer, ORNL



Time-of-flight technique: hit sample with a pulse of neutrons, and measure neutron velocity (more efficient)

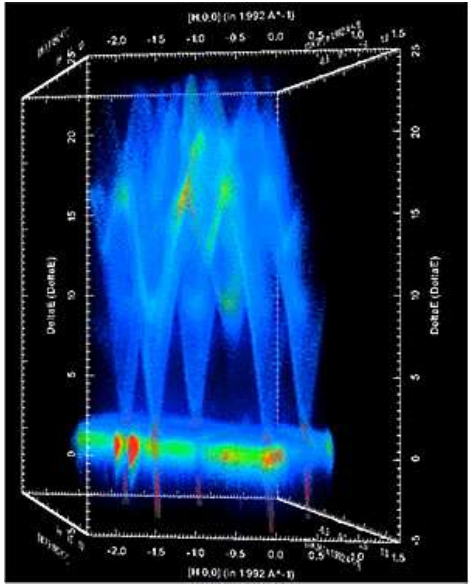


Old:
1D plots



New:
Map out four-dimensional space:

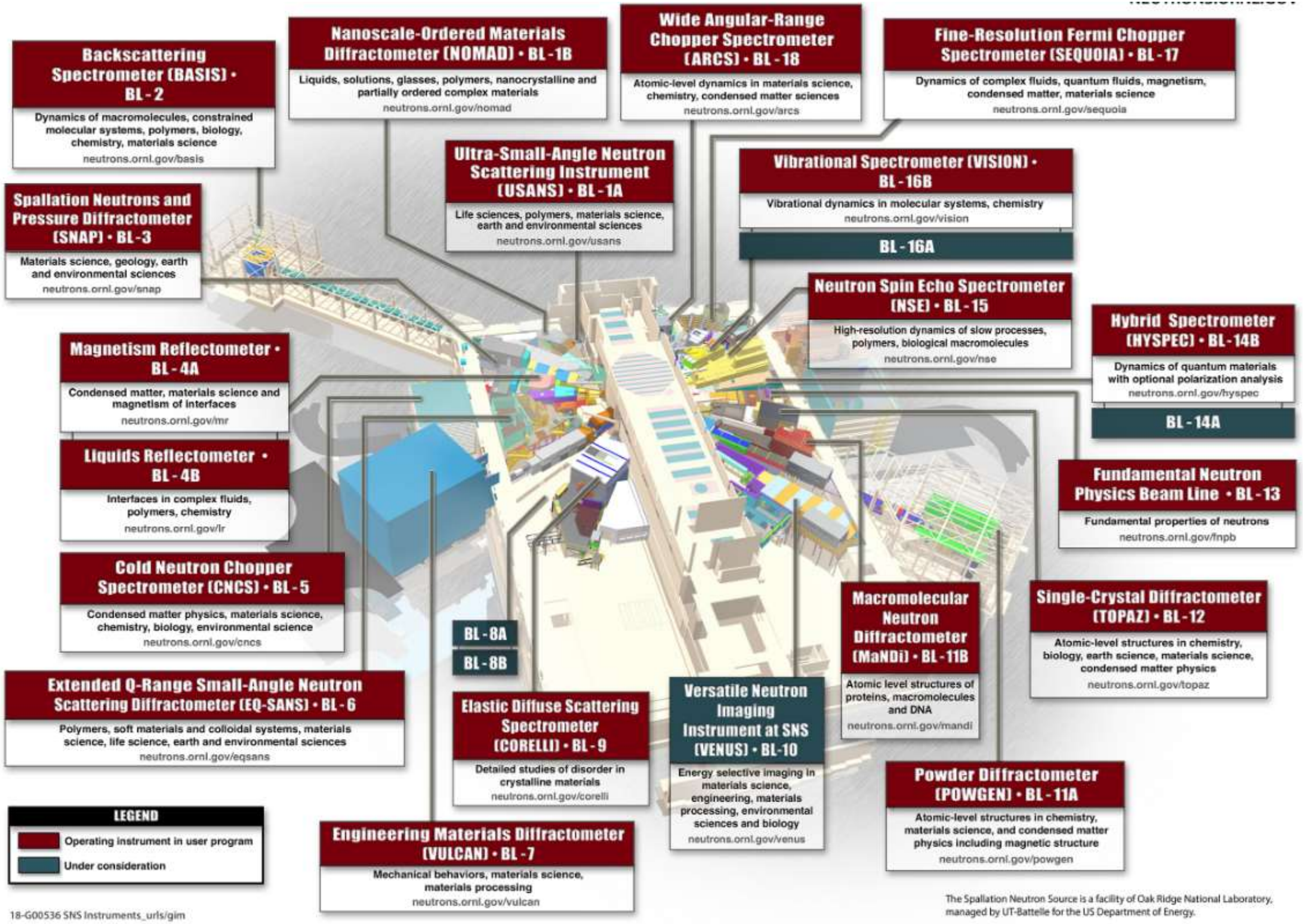
Three spatial dimensions,
Energy transfer, Intensity



Europeanspallationsource.se

Modern neutron facilities:

Neutrons.ornl.gov


Backscattering Spectrometer (BASIS) • BL-2
Dynamics of macromolecules, constrained molecular systems, polymers, biology, chemistry, materials science
neutrons.ornl.gov/basis
Nanoscale-Ordered Materials Diffractometer (NOMAD) • BL-1B
Liquids, solutions, glasses, polymers, nanocrystalline and partially ordered complex materials
neutrons.ornl.gov/nomad
Wide Angular-Range Chopper Spectrometer (ARCS) • BL-18
Atomic-level dynamics in materials science, chemistry, condensed matter sciences
neutrons.ornl.gov/arcs
Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA) • BL-17
Dynamics of complex fluids, quantum fluids, magnetism, condensed matter, materials science
neutrons.ornl.gov/sequoia
Spallation Neutrons and Pressure Diffractometer (SNAP) • BL-3
Materials science, geology, earth and environmental sciences
neutrons.ornl.gov/snap
Ultra-Small-Angle Neutron Scattering Instrument (USANS) • BL-1A
Life sciences, polymers, materials science, earth and environmental sciences
neutrons.ornl.gov/usans
Vibrational Spectrometer (VISION) • BL-16B
Vibrational dynamics in molecular systems, chemistry
neutrons.ornl.gov/vision
BL-16A
Neutron Spin Echo Spectrometer (NSE) • BL-15
High-resolution dynamics of slow processes, polymers, biological macromolecules
neutrons.ornl.gov/nse
Hybrid Spectrometer (HYSPEC) • BL-14B
Dynamics of quantum materials with optional polarization analysis
neutrons.ornl.gov/hyspec
BL-14A
Magnetism Reflectometer • BL-4A
Condensed matter, materials science and magnetism of interfaces
neutrons.ornl.gov/mr
Liquids Reflectometer • BL-4B
Interfaces in complex fluids, polymers, chemistry
neutrons.ornl.gov/lr
Fundamental Neutron Physics Beam Line • BL-13
Fundamental properties of neutrons
neutrons.ornl.gov/fnpb
Cold Neutron Chopper Spectrometer (CNCS) • BL-5
Condensed matter physics, materials science, chemistry, biology, environmental science
neutrons.ornl.gov/cncs
BL-8A
BL-8B
Macromolecular Neutron Diffractometer (MaNDi) • BL-11B
Atomic level structures of proteins, macromolecules and DNA
neutrons.ornl.gov/mandi
Single-Crystal Diffractometer (TOPAZ) • BL-12
Atomic-level structures in chemistry, biology, earth science, materials science, condensed matter physics
neutrons.ornl.gov/topaz
Extended Q-Range Small-Angle Neutron Scattering Diffractometer (EQ-SANS) • BL-6
Polymers, soft materials and colloidal systems, materials science, life science, earth and environmental sciences
neutrons.ornl.gov/eqsans
Elastic Diffuse Scattering Spectrometer (CORELLI) • BL-9
Detailed studies of disorder in crystalline materials
neutrons.ornl.gov/corelli
Versatile Neutron Imaging Instrument at SNS (VENUS) • BL-10
Energy selective imaging in materials science, engineering, materials processing, environmental sciences and biology
neutrons.ornl.gov/venus
Powder Diffractometer (POWGEN) • BL-11A
Atomic-level structures in chemistry, materials science, and condensed matter physics including magnetic structure
neutrons.ornl.gov/powgen
LEGEND
Operating instrument in user program
Under consideration
Engineering Materials Diffractometer (VULCAN) • BL-7
Mechanical behaviors, materials science, materials processing
neutrons.ornl.gov/vulcan
The Spallation Neutron Source is a facility of Oak Ridge National Laboratory, managed by UT-Battelle for the US Department of Energy.
18-G00536 SNS Instruments_urls/gim

Neutron instruments worldwide:

"Neutrons for the Nation" report, Aps.org

Neutron facilities in the US:

SNS
SPALLATION NEUTRON SOURCE

HFIR

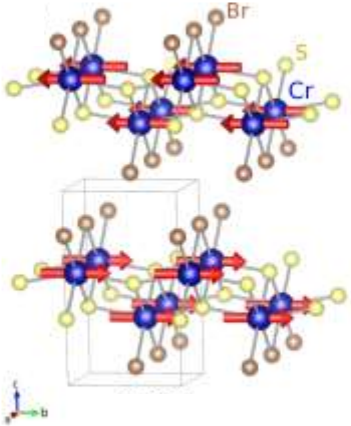
NIST Center for Neutron Research

Oak Ridge, TN

Gaithersburg, MD

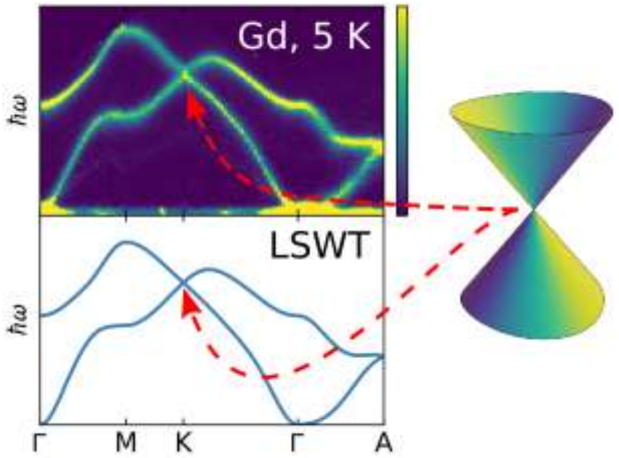
“Bread and butter” neutron experiments:

Static magnetic order



Scheie et al, *Adv. Science* (2022)

Magnetic dynamics



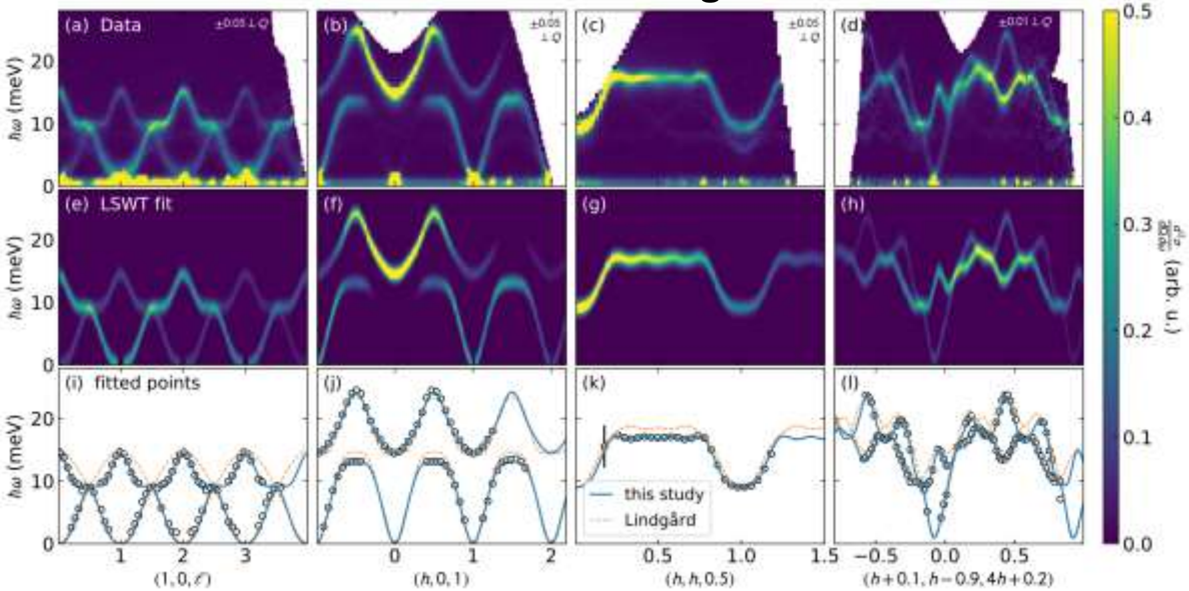
Scheie et al, *PRL* (2022)

Example: modeling and fitting the magnetic interactions of elemental gadolinium

Neutron data and fitted magnon model

data

model



Fitted exchange parameters

$J_1 =$	-138 ± 8	$J_{10} =$	-2 ± 2	$J_{19} =$	-5 ± 3
$J_2 =$	-174 ± 4	$J_{11} =$	0 ± 20	$J_{20} =$	-5.6 ± 1.3
$J_3 =$	50 ± 20	$J_{12} =$	-25 ± 6	$J_{21} =$	5 ± 9
$J_4 =$	41 ± 6	$J_{13} =$	0 ± 2	$J_{22} =$	-5 ± 7
$J_5 =$	-10 ± 20	$J_{14} =$	-8 ± 10	$J_{23} =$	29 ± 7
$J_6 =$	-14 ± 4	$J_{15} =$	4 ± 2	$J_{24} =$	-5 ± 9
$J_7 =$	-4 ± 2	$J_{16} =$	1 ± 7	$J_{25} =$	1 ± 8
$J_8 =$	0 ± 3	$J_{17} =$	4 ± 8	$J_{26} =$	7 ± 2
$J_9 =$	-10 ± 20	$J_{18} =$	10 ± 20		

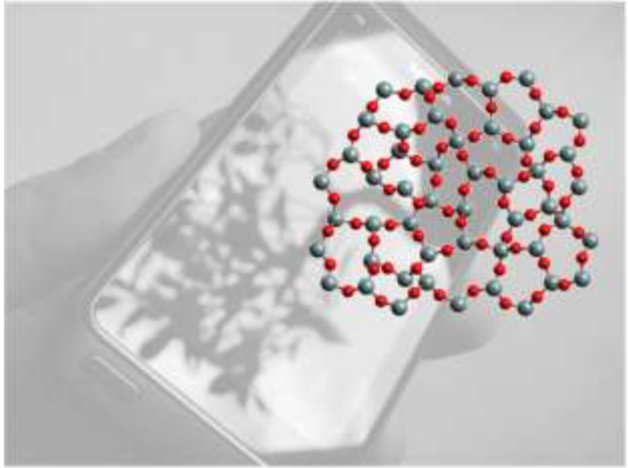
Scheie, PRB (2022)

Other “Bread and butter” neutron experiments:

Locations of hydrogen

Kneller et al, *JCB* **295**, 50 (2020)

Contrast between elements



Shi et al, *N. Comm.* **14**, 13 (2023)

Part 3
Current uses of neutron scattering: exploring the unknown

Condensed matter theorists propose exotic states of matter—we try to find them.

Quantum spin liquid (QSL): a massively entangled magnetic state potentially useful for quantum computers

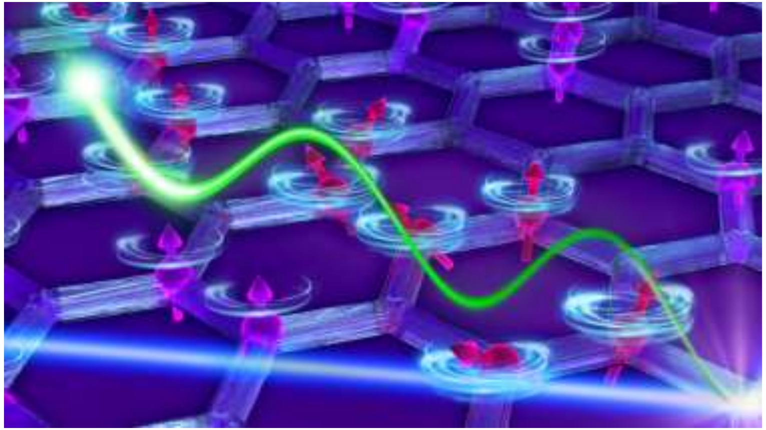


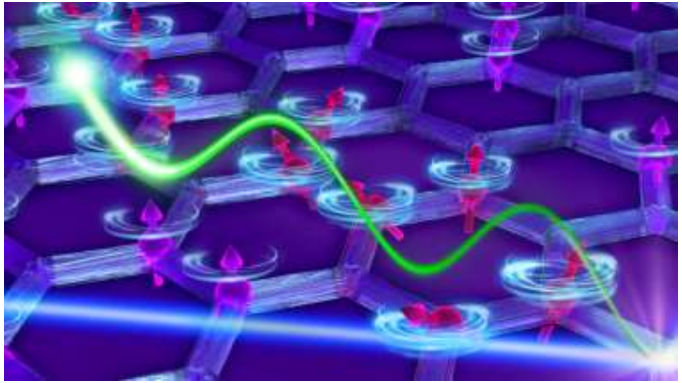
Image: neutrons.ornl.gov

Theoretically, they should exist.

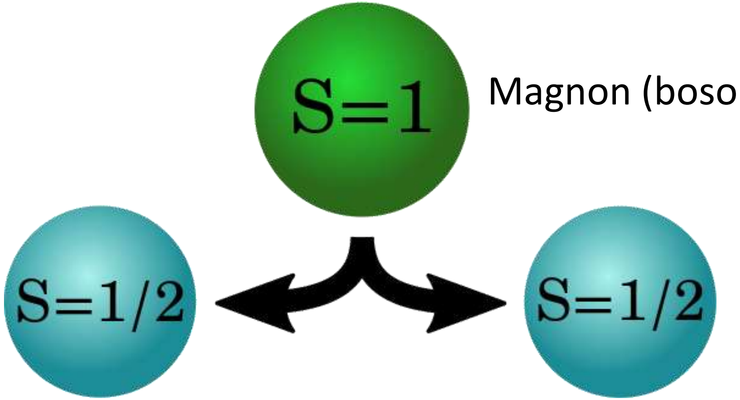
...but they are frustratingly difficult to identify **experimentally.**

Key features of QSL ground state:

(1) Long range entanglement:



(2) Fractional quasiparticles:

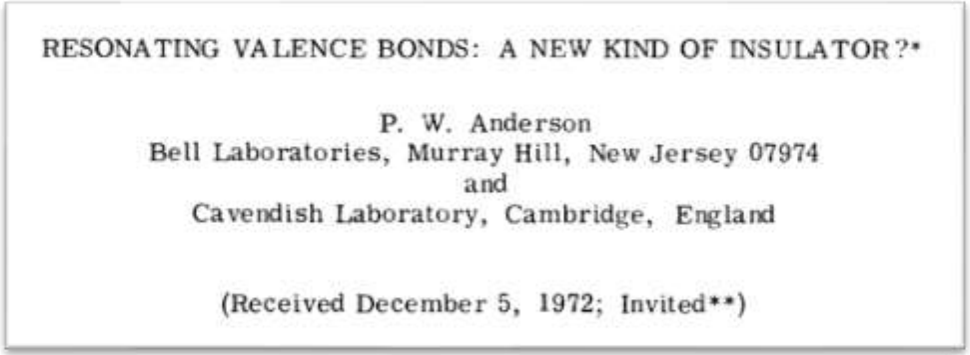


Magnon (boson)

Spinon (fermion)

Spinon (fermion)

Original idea for a quantum spin liquid (QSL): Triangular lattice Heisenberg antiferromagnet



RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR ?*

P. W. Anderson
Bell Laboratories, Murray Hill, New Jersey 07974
and
Cavendish Laboratory, Cambridge, England

(Received December 5, 1972; Invited**)

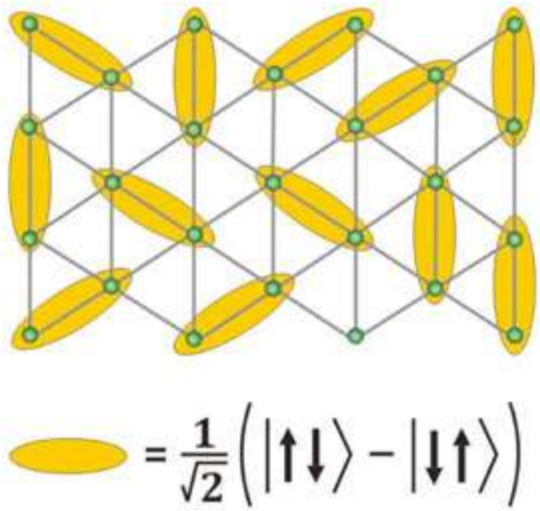
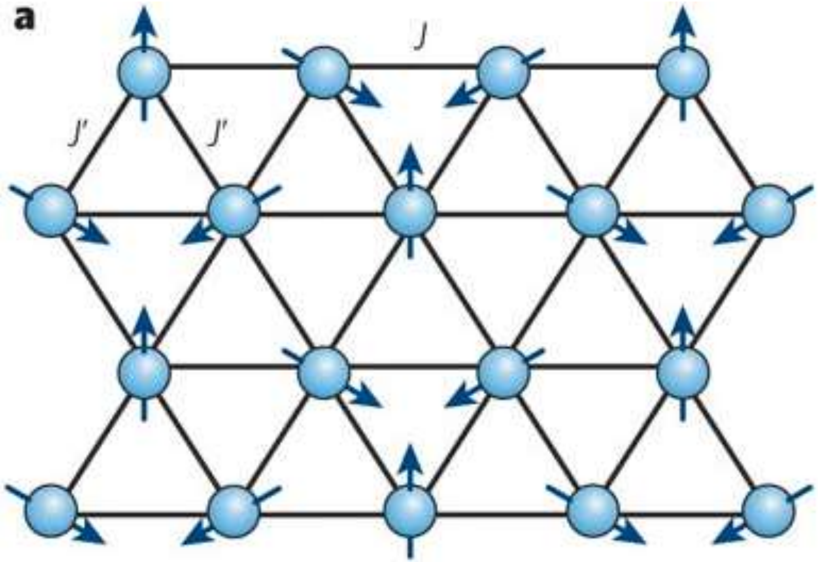


Image: Universität Augsburg, EP VI/EKM

However, subsequent calculations showed that the Heisenberg triangular lattice antiferromagnet magnetically orders at low temperatures.

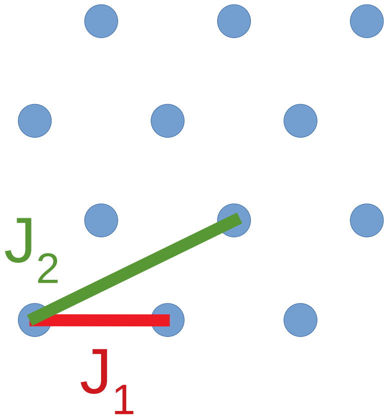
120° order

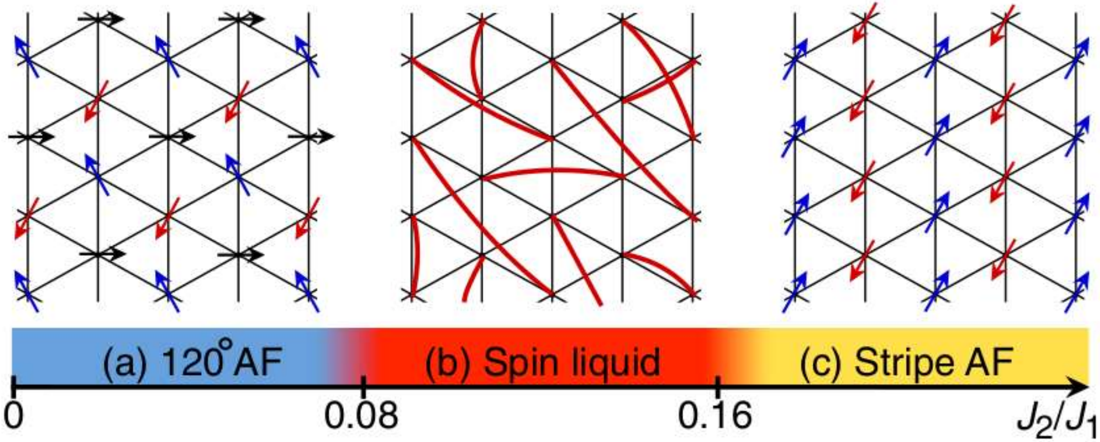
Capriotti et al, PRL (1999)
White and Chernyshev, PRL (2007)



Balents, Nature (2010)

Instead, a small second neighbor exchange J_2 ($0.06 \lesssim J_2/J_1 \lesssim 0.16$) induces a QSL phase

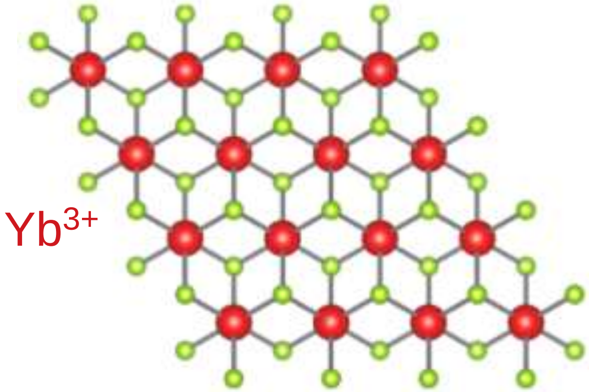


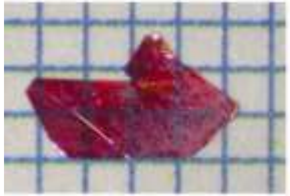


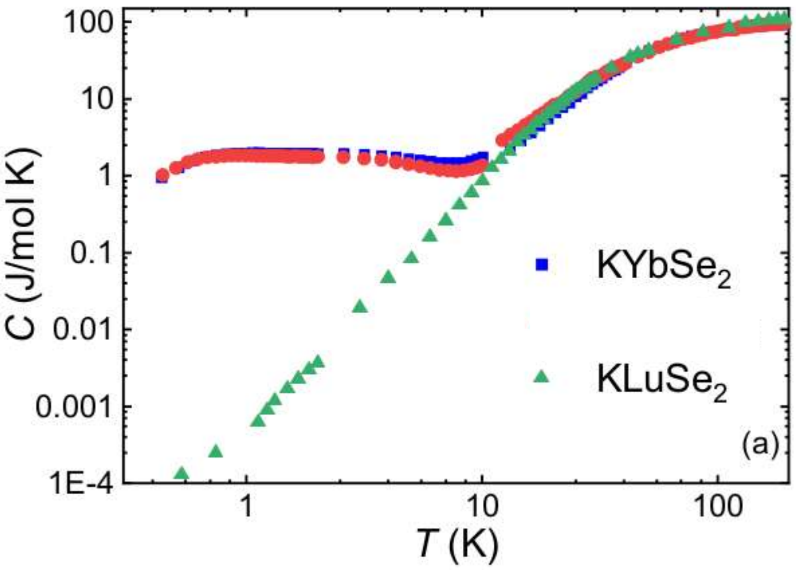
Iqbal et al, PRB (2016)
Zhu et al, PRB (2015)

The precise nature of the QSL is debated

$KYbSe_2$ showed no magnetic order down to 400 mK. Maybe a QSL state?

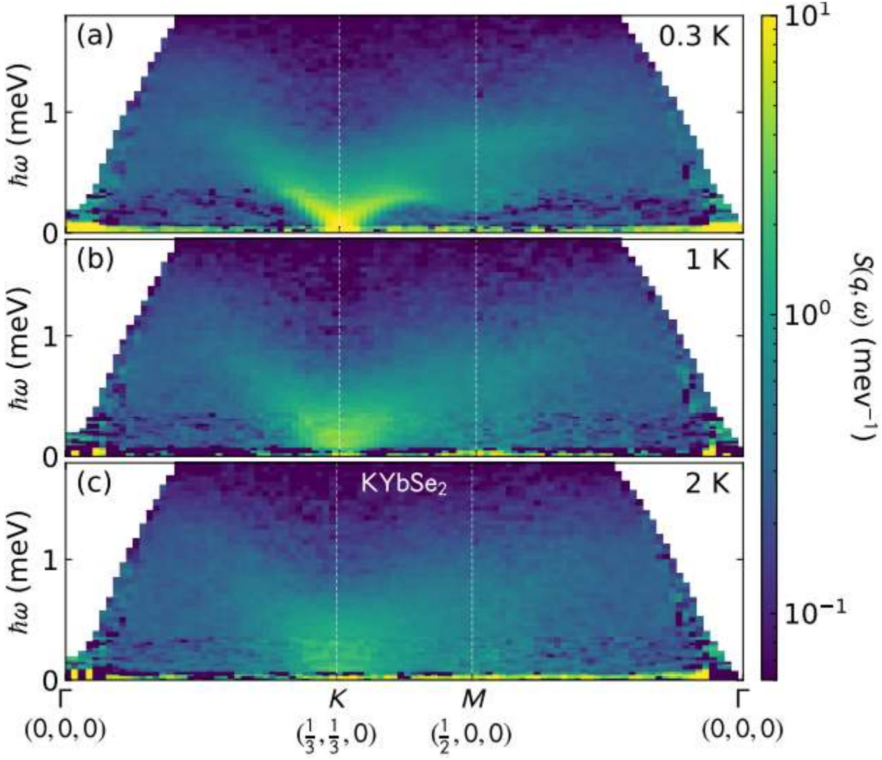




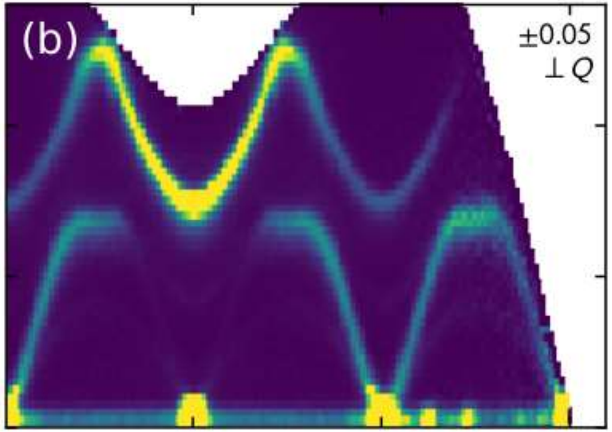


Xing, *APL Materials* (2021)

Neutron scattering experiment on $KYbSe_2$ @ CNCS



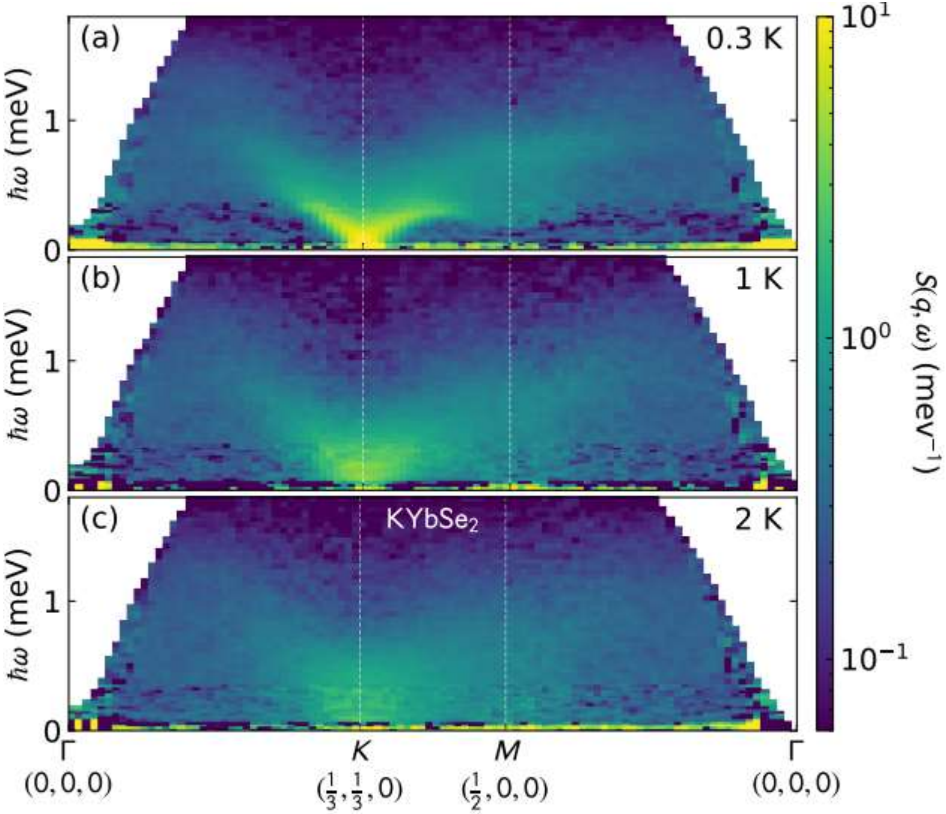
Ordinary magnetic material: well-defined modes



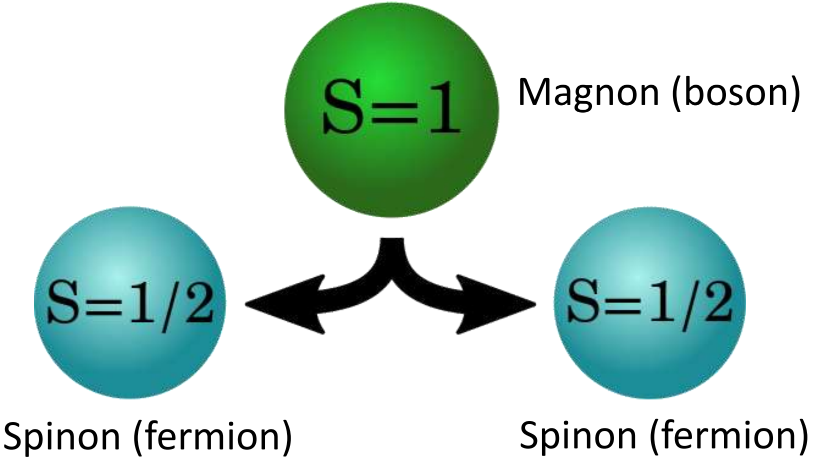
Elemental Gd, 5K

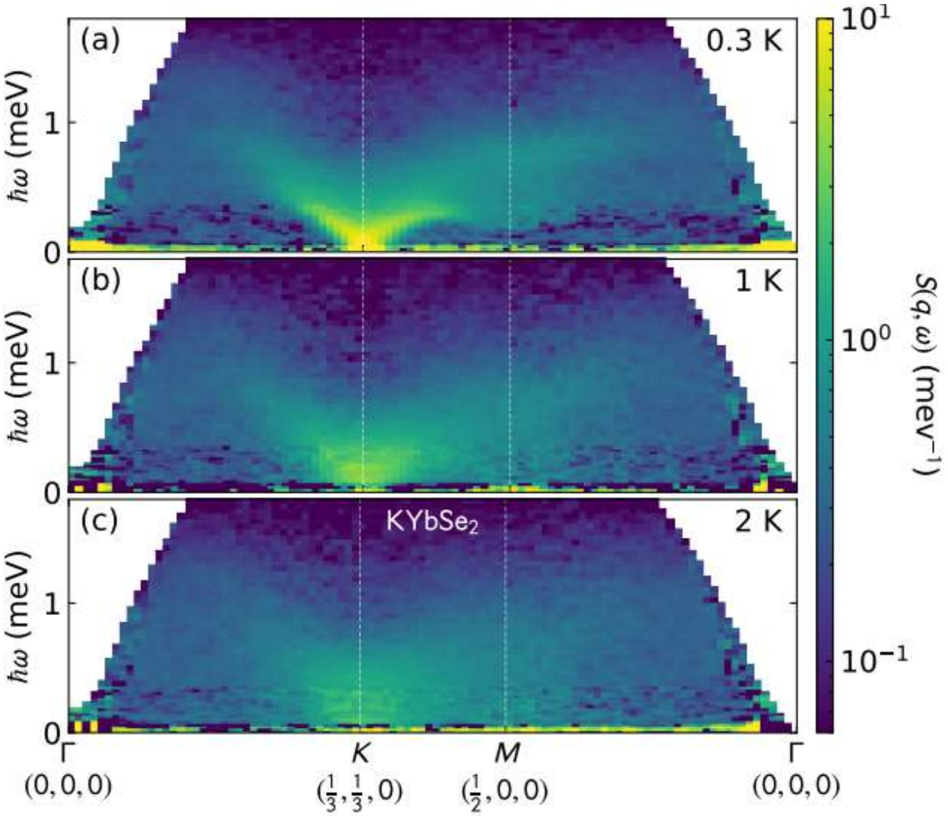
Scheie et al, PRB (2022)

$KYbSe_2$: Diffuse continuum

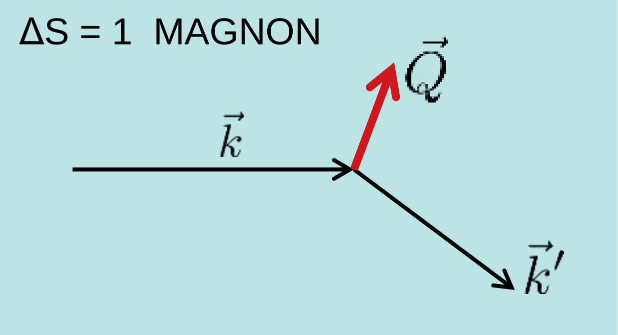


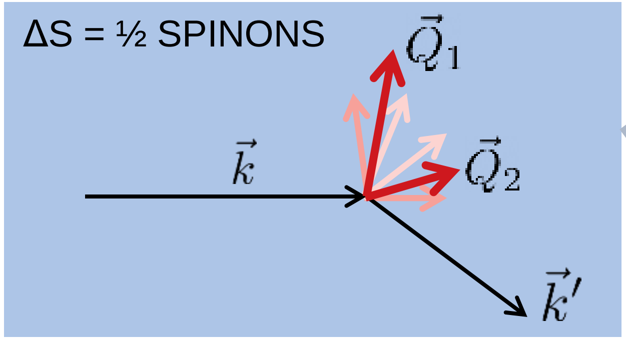
Continuum scattering can indicate fractional quasiparticles

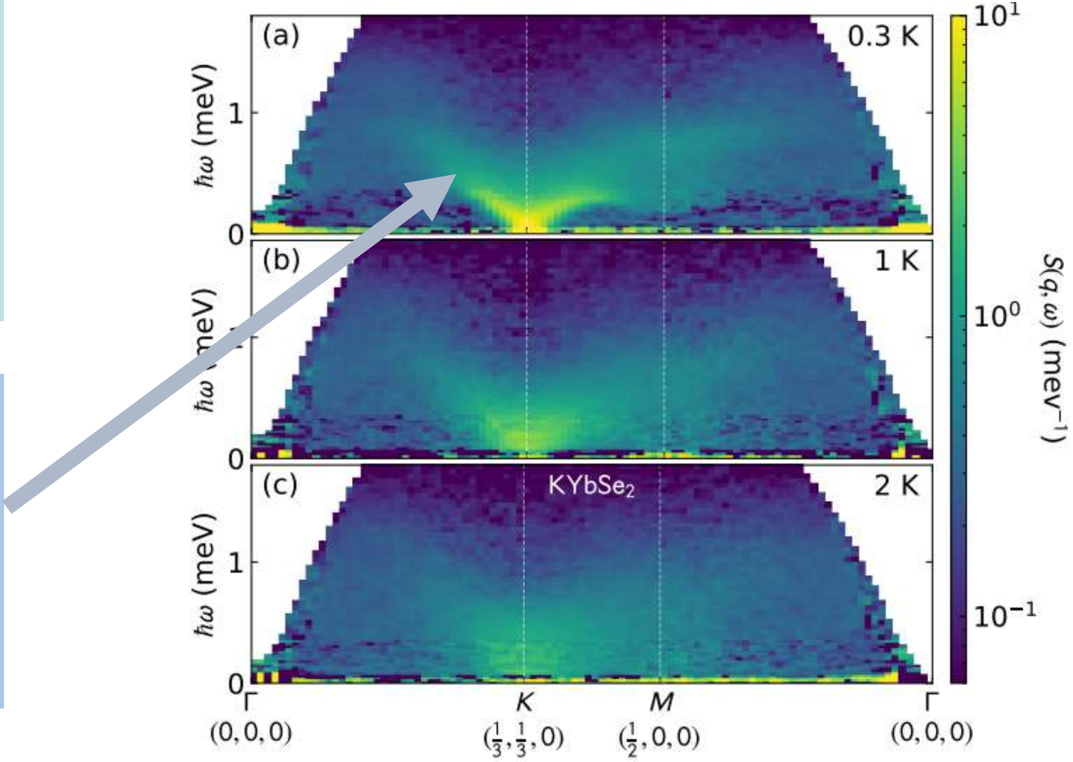




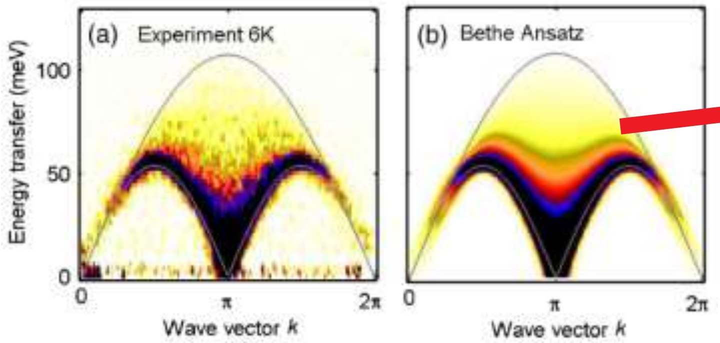
Continuum scattering can indicate fractional quasiparticles



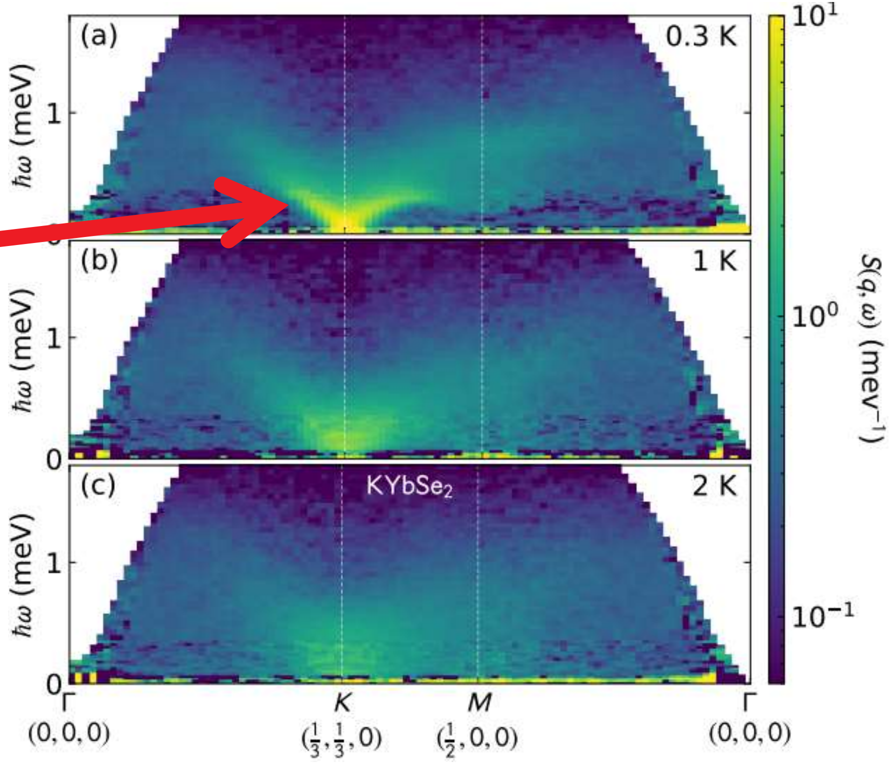




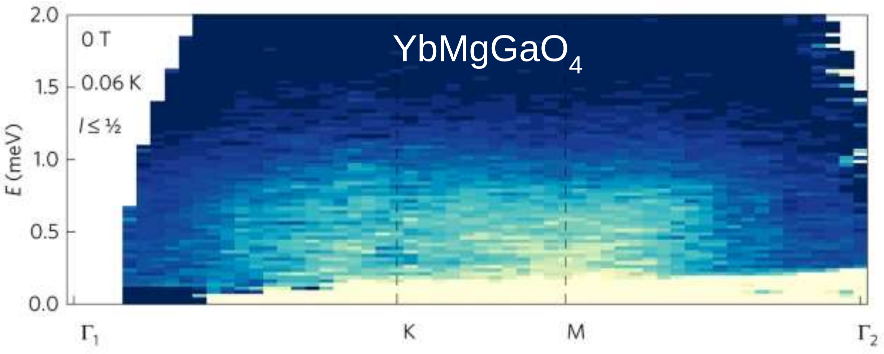
Continuum resembles the 1D spin chain spinon signal (known instance of fractionalization)…



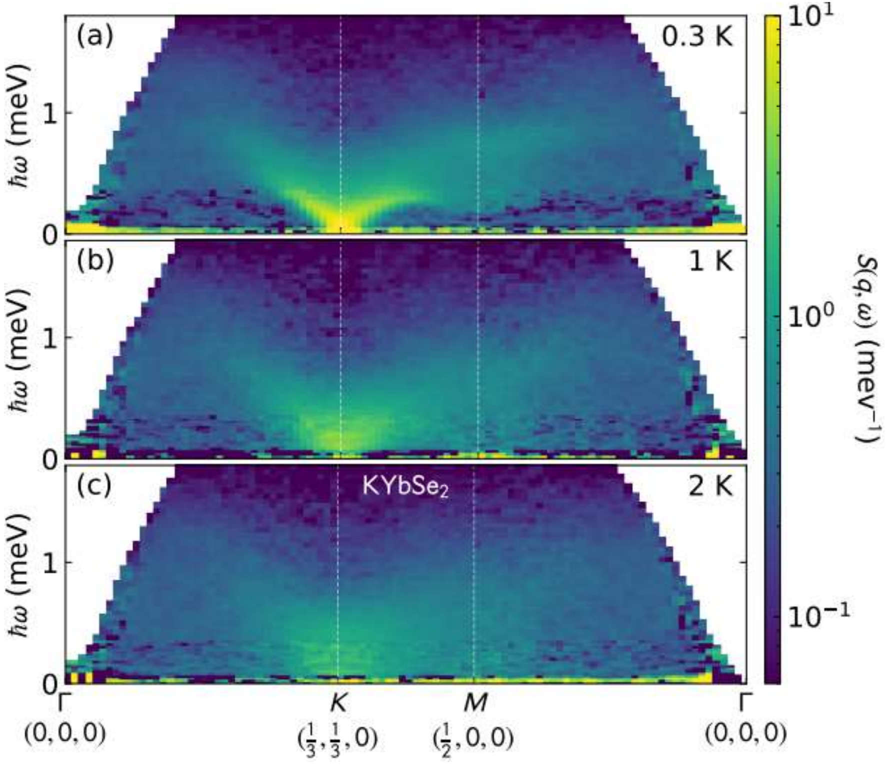
Lake et al, Phys. Rev. Lett. **111**, 137205 (2013)



But crystalline disorder also causes a diffuse continuum…



Paddison et al, N. Phys (2017)



Los Alamos NATIONAL LABORATORY

So… is $KYbSe_2$ a QSL?

We use two tools to answer this question:

A) Magnetic Hamiltonian analysis

B) Comparison to theoretical models

So… is $KYbSe_2$ a QSL?

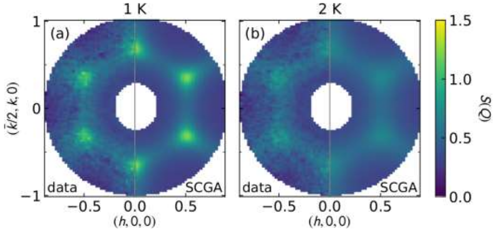
We use two tools to answer this question:

A) Magnetic Hamiltonian analysis

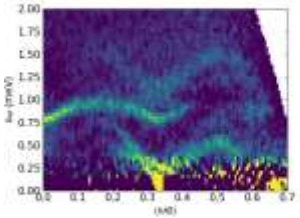
B) Comparison to theoretical models

Three methods to fit Hamiltonian:

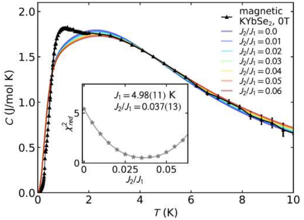
1. Paramagnetic diffuse scattering



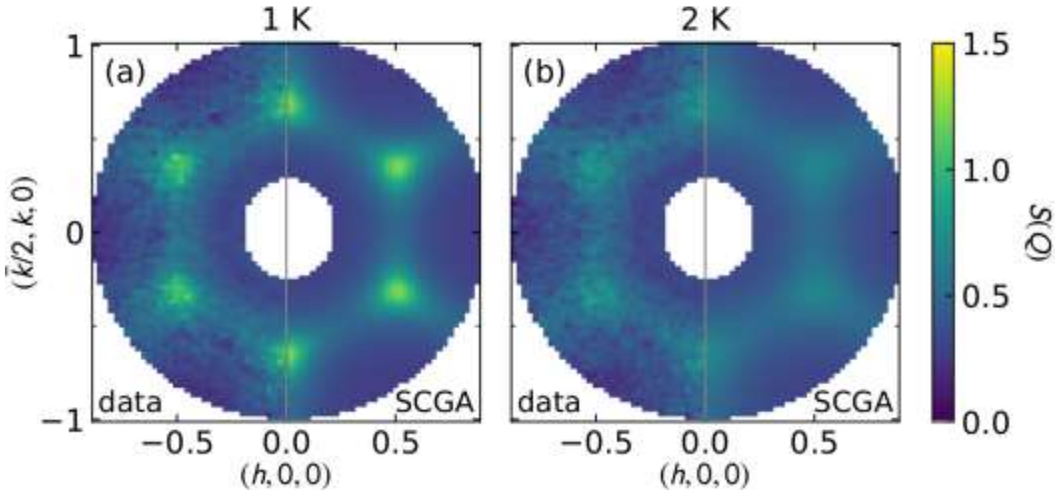
2. Nonlinear spin waves



3. Heat capacity expansion



<u>Method 1</u>: Onsager Reaction Field theory fitting the high temperature diffuse scattering



Fitted values (in K):

$$J_1 = \begin{pmatrix} 2.34(10) & 0.016(7) & 0.00(4) \\ 0.016(7) & 2.34(10) & 0.00(6) \\ 0.00(4) & 0.00(6) & 2.28(10) \end{pmatrix}$$

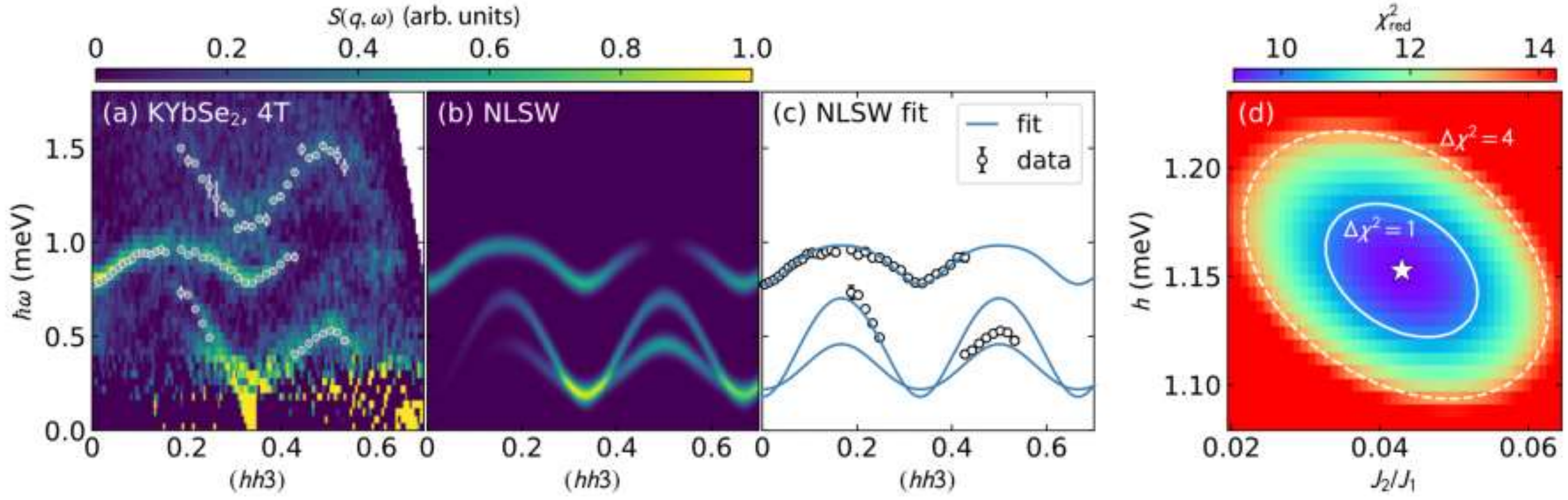
$$J_2 = 0.11(2)$$

(Classical method, so overall energy scale is wrong.)

Result: **nearly isotropic exchange, with J_2/J_1 = 0.047(7)**

For the technique, see Paddison, PRL (2020)

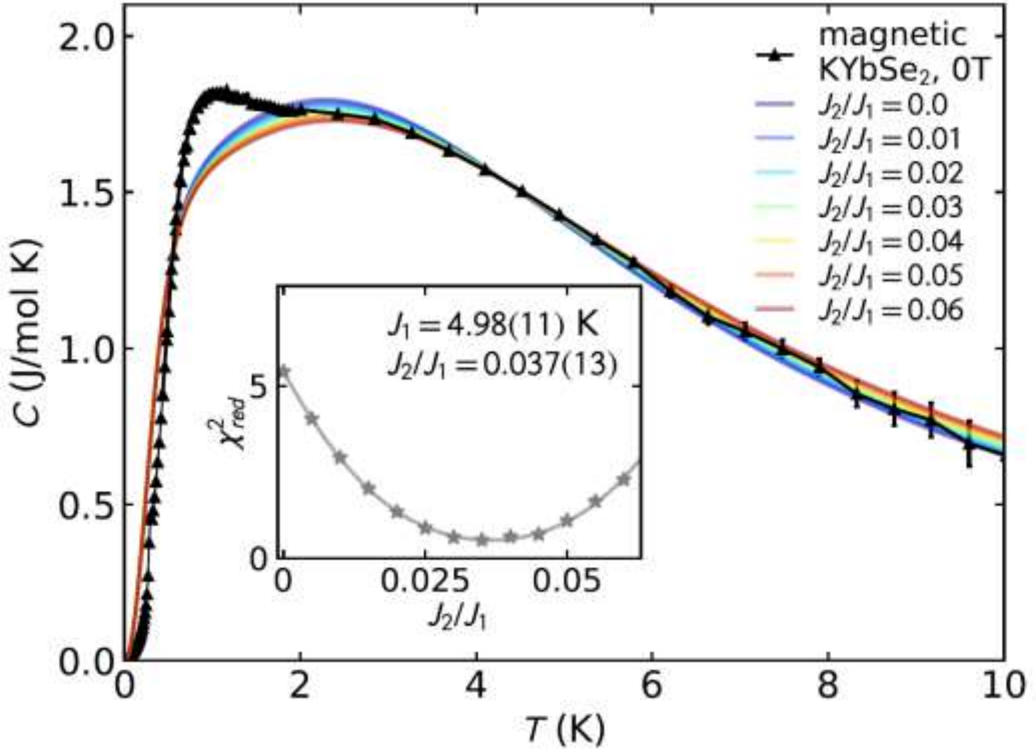
Method 2: Nonlinear spin wave fits in the 1/3 magnetization plateau



Result: J_2/J_1 = 0.043 ± 0.010

Agrees with J_2/J_1=0.047(7) from diffuse scattering fits

Method 3: Heat capacity expansion fits



Fit between 2 K and 8 K

Courtesy Laura Messio @ Sorbonne University

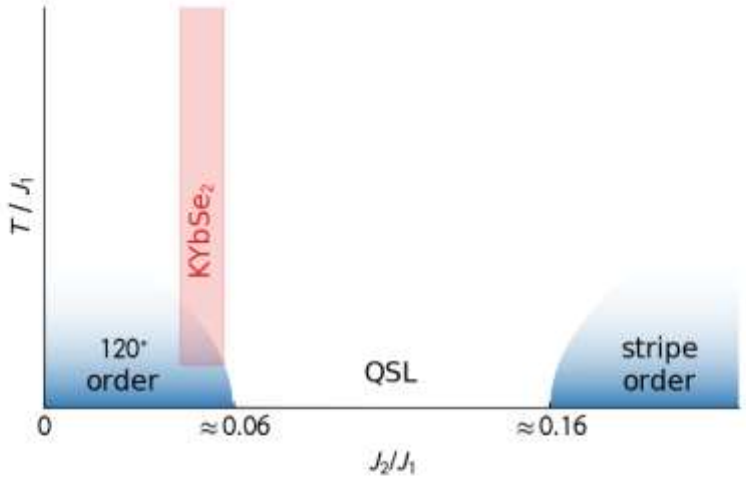
Result:

$\mathbf{J_2/J_1 = 0.037 \pm 0.013}$

For method, see Gonzalez, Bernu, Pierre, & Messio, Scipost Phys. (2022)

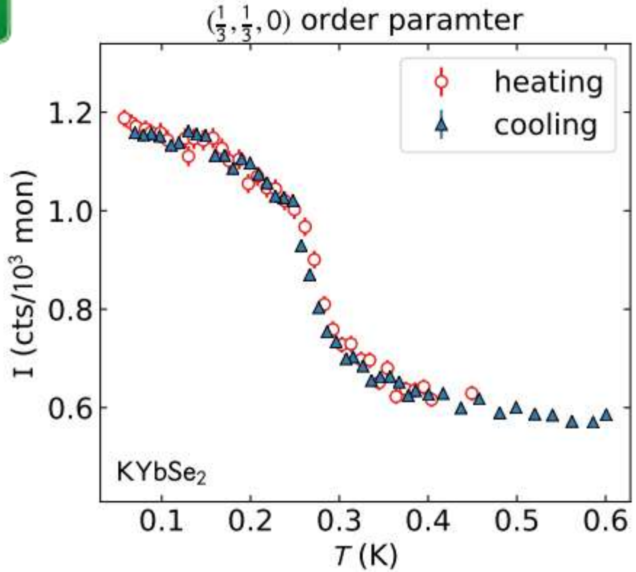
All methods agree to within uncertainty: $KYbSe_2$ is very close to the QSL phase

Theoretical technique	J_1 (meV)	J_2/J_1
Onsager reaction field	*NA*	0.047 ± 0.007
Nonlinear spin waves	0.456 ± 0.013	0.043 ± 0.010
Heat capacity	0.429 ± 0.010	0.037 ± 0.013
Weighted mean:	0.438 ± 0.008	$\boxed{0.044 \pm 0.005}$

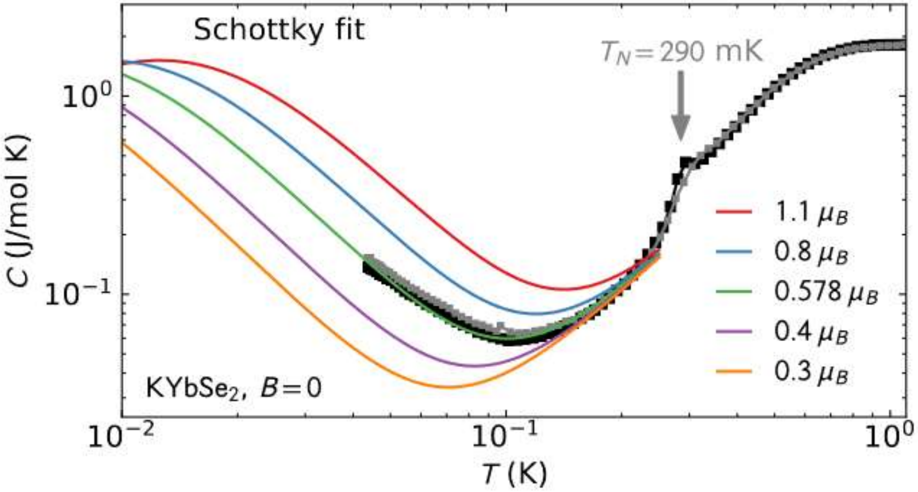


However, if our model is correct, we should see 120° magnetic order at low temperatures. This was subsequently observed:

Neutron scattering



Heat capacity



So… is $KYbSe_2$ a QSL?

We use two tools to answer this question:

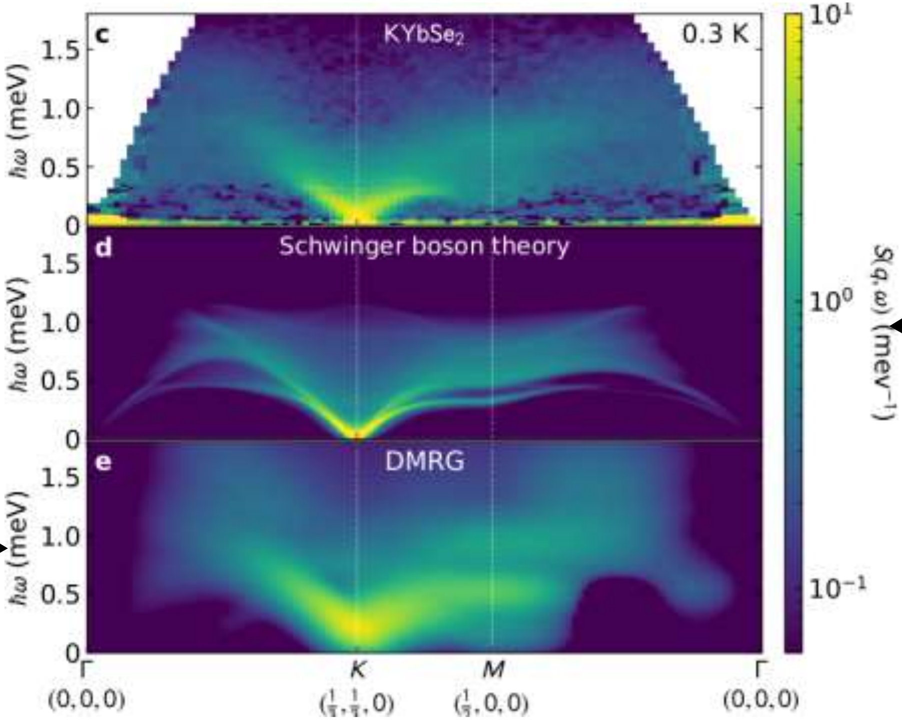
A) Magnetic Hamiltonian analysis

B) Comparison to theoretical models

Theoretical calculations for a 2D triangular lattice with the fitted Hamiltonian closely resemble our data

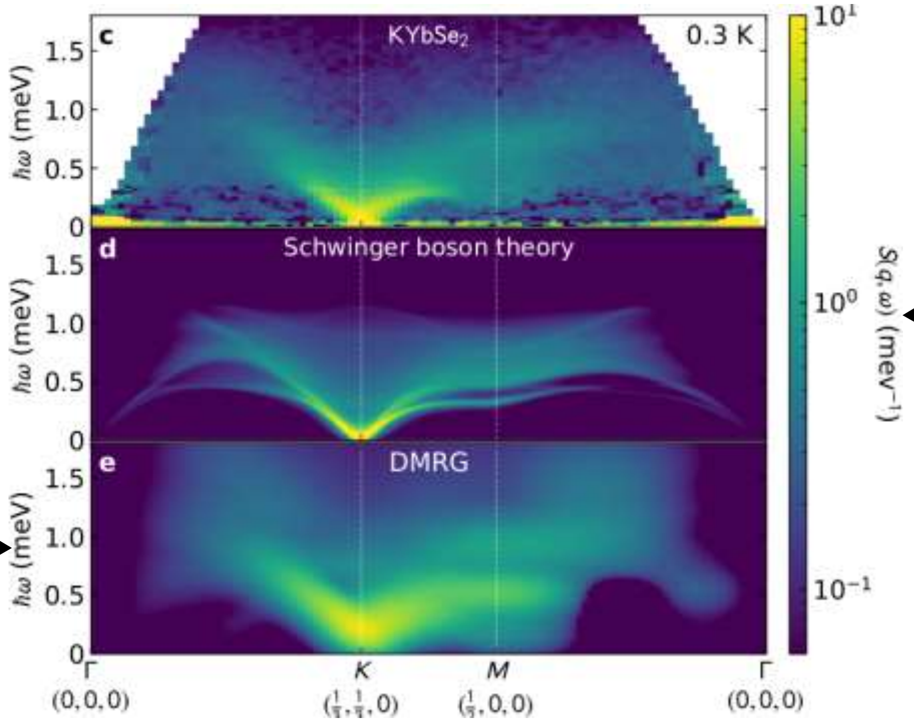
Christian Batista
U. Tennessee

Joel Moore
UC Berkeley



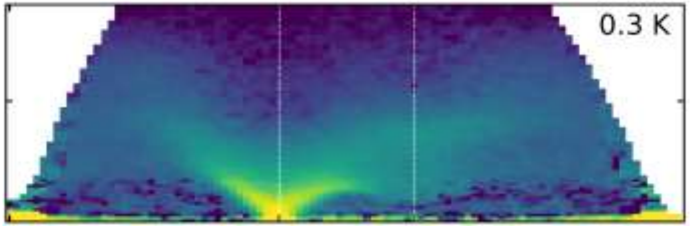
Theoretical calculations for a 2D triangular lattice with the fitted Hamiltonian closely resemble our data

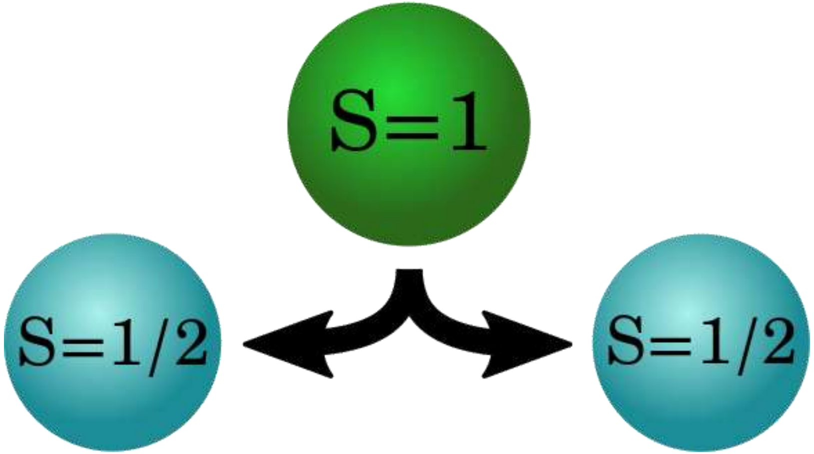
Brute force:
Includes all
quantum effects



Fractionalized
spinons with two-spinon
continuum

Although $KYbSe_2$ orders magnetically, its inelastic spectrum is dominated by fractionalized spinons

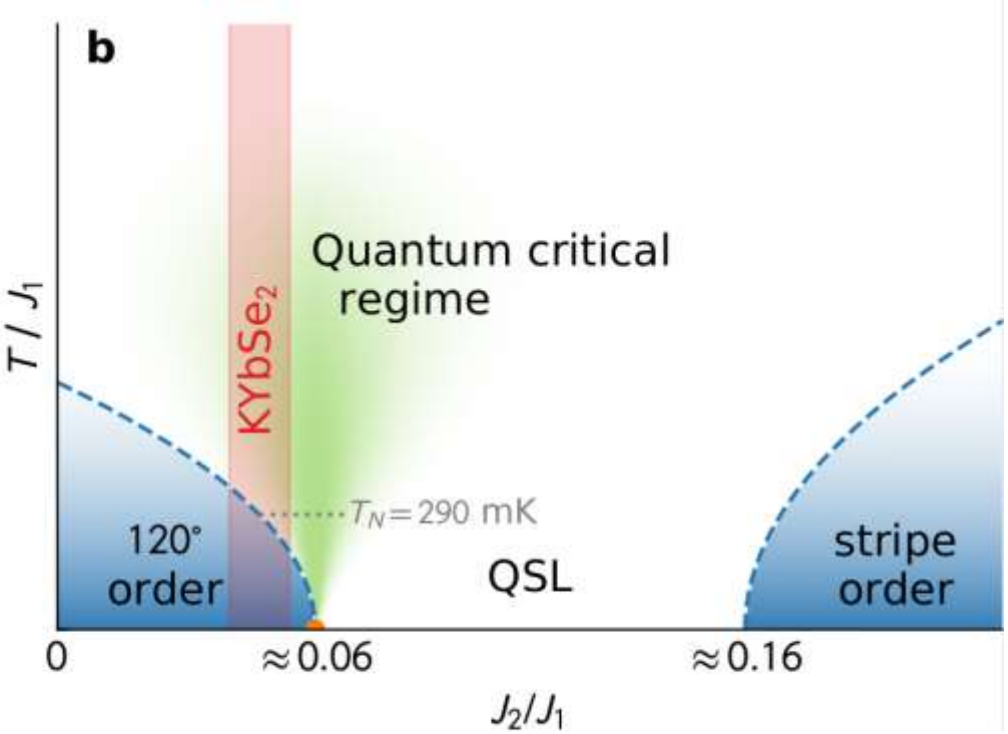




So is $KYbSe_2$ a QSL?

So is $KYbSe_2$ a QSL?

It's a <u>proximate QSL</u>



Another project: measuring quantum entanglement using neutron scattering

To really understand quantum materials, we need a probe which can “see the quantumness” in a material.

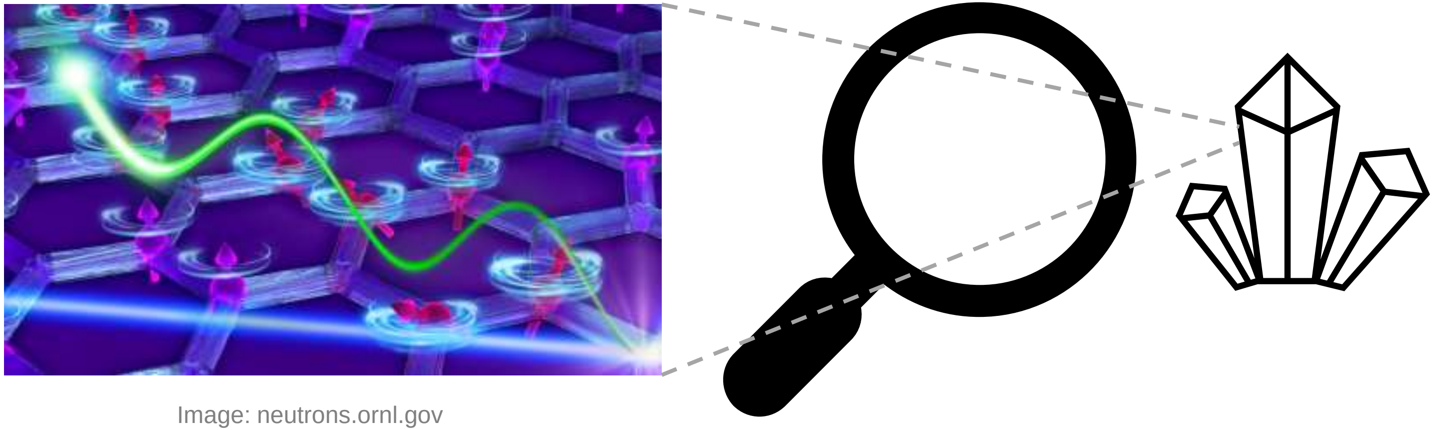
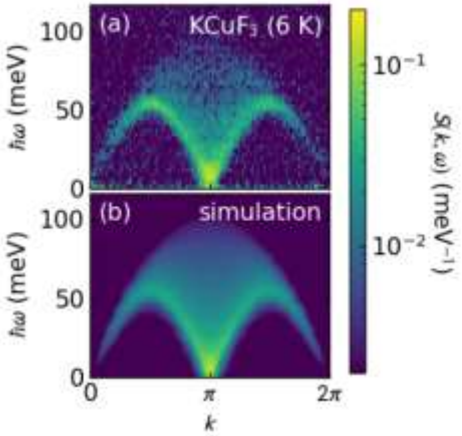


Image: neutrons.ornl.gov

entanglement witnesses

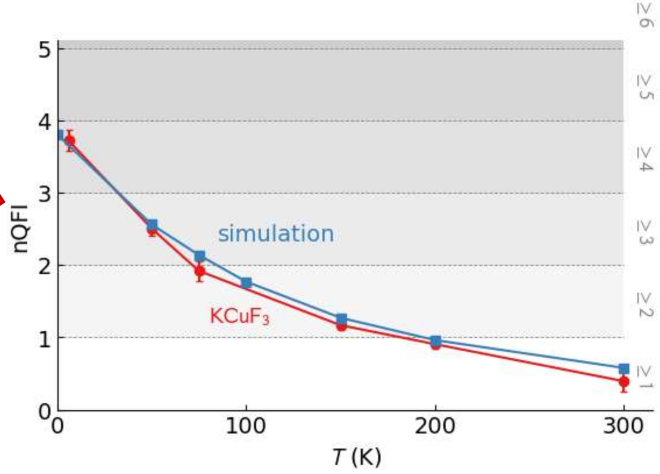
Data



Mathematical Filter
(**entanglement witness**)

$$f_Q(T) = \frac{4\hbar}{\pi}\int_0^\infty d(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\chi''(\hbar\omega, T)$$

Quantum information



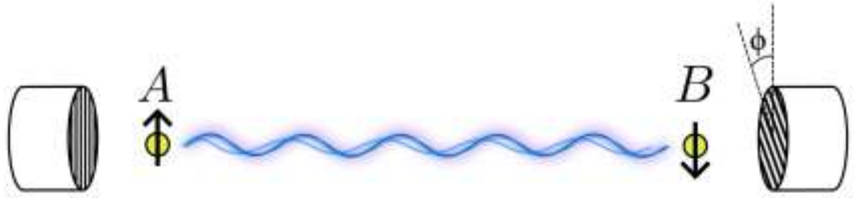
First “entanglement witness” proposed by John Stewart Bell in 1964, based on two-point (i.e., two-spin) correlation

$$|\langle S_1^a S_2^b \rangle - \langle S_1^a S_2^c \rangle| \leq 1 + \langle S_1^b S_2^c \rangle$$

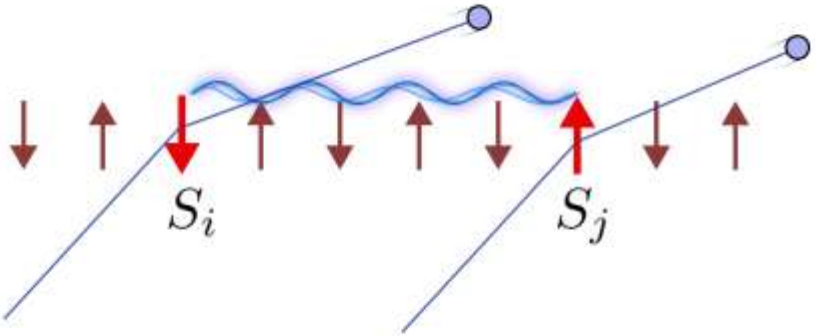
Classically, the inequality is always satisfied.
In the presence of quantum entanglement, the inequality is violated.

Wikipedia
Bell, *Rev. Mod. Phys.* (1966)

Entanglement information is encoded in two-point correlations:



Bell test $\langle A_p B_p \rangle$



Neutron scattering $\langle S_i S_j \rangle$

Entanglement information is encoded in the neutron structure factor; the trick is knowing how to extract it.

$$S(\vec{q},\omega) \propto \int e^{i\vec{q}\cdot\vec{r}} \langle \vec{S}_{i,\alpha} \vec{S}_{j,\beta} \rangle$$

We recently showed it is possible to measure quantum spin entanglement called Quantum Fisher Information (QFI) using neutron scattering

REVIEW Adv. Quantum Technol. 2024, 2400196 ADVANCED QUANTUM TECHNOLOGIES www.advquantumtech.com

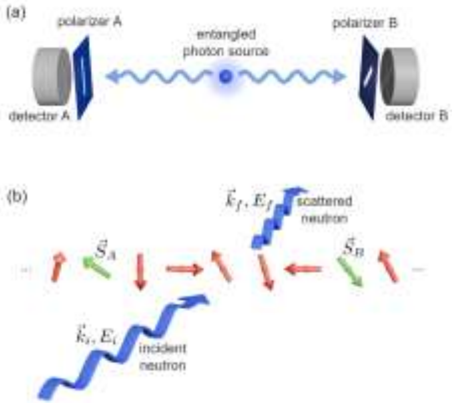
Witnessing Entanglement and Quantum Correlations in Condensed Matter: A Review

Pontus Laurell, Allen Scheie, Elbio Dagotto, and D. Alan Tennant**

ELSEVIER Materials Today Quantum Volume 5, March 2025, 100020

Tutorial: Extracting entanglement signatures from neutron spectroscopy

Allen Scheie [a], Pontus Laurell [b], Wolfgang Simeth [a], Elbio Dagotto [c, d], D. Alan Tennant [c, e]



Quantum Fisher Information:

defines how precisely an operator $\mathcal{O}$ can be measured

$$f_Q = 4\left(\langle\psi|\mathcal{O}^\dagger\mathcal{O}|\psi\rangle - \langle\psi|\mathcal{O}|\psi\rangle^2\right)$$

(pure state QFI)

By the Cramér-Rao bound, this gives a <u>lower bound on the number of entangled objects</u>:

$$f_Q[\rho] > m(\lambda_{\max} - \lambda_{\min})^2$$

m **= entanglement depth,**

i.e. (m+1) partite entanglement

Hyllus et al, Phys. Rev. A **85**, 022321 (2012)
Hauke et al, Nat. Phys. **12**, 778–782 (2016)

Quantum Fisher Information:

defines how precisely an operator $\mathcal{O}$ can be measured

$$f_Q = 4\left(\langle\psi|\mathcal{O}^\dagger\mathcal{O}|\psi\rangle - \langle\psi|\mathcal{O}|\psi\rangle^2\right)$$

(pure state QFI)

$\mathcal{O}$ **can be *anything****:

$\mathcal{O} =$ 

$f_Q = 4\big(\langle$  †  $\rangle - \langle$  $\rangle^2\big)$

***must be Hermitian**

Quantum Fisher Information:

defines how precisely an operator $\mathcal{O}$ can be measured

$$f_Q = 4\left(\langle\psi|\mathcal{O}^\dagger\mathcal{O}|\psi\rangle - \langle\psi|\mathcal{O}|\psi\rangle^2\right)$$

(pure state QFI)

For magnetic neutron scattering: $\mathcal{O} = S_\alpha(\mathbf{Q}) = \sum_j S^\alpha{}_j e^{i\mathbf{Q}\cdot\mathbf{r}_j}$

$$f_Q = 4\left(\underbrace{\langle\psi|S_\alpha^\dagger(\mathbf{Q})S_\alpha(\mathbf{Q})|\psi\rangle}_{\text{Total scattering}} - \underbrace{\langle\psi|S_\alpha(\mathbf{Q})|\psi\rangle^2}_{\text{elastic scattering}}\right)$$

This is experimentally measurable!

Hauke et al, Nat. Phys. **12**, 778–782 (2016)

Hauke et al provided a generalization to T>0:

$$f_{\mathcal{Q}} = 2\sum_{k,l}\frac{1}{Z}\frac{\left(e^{-\beta E_k} - e^{-\beta E_l}\right)^2}{e^{-\beta E_k} + e^{-\beta E_l}}|\langle k|\hat{\mathcal{O}}|l\rangle|^2$$

(thermal state QFI)

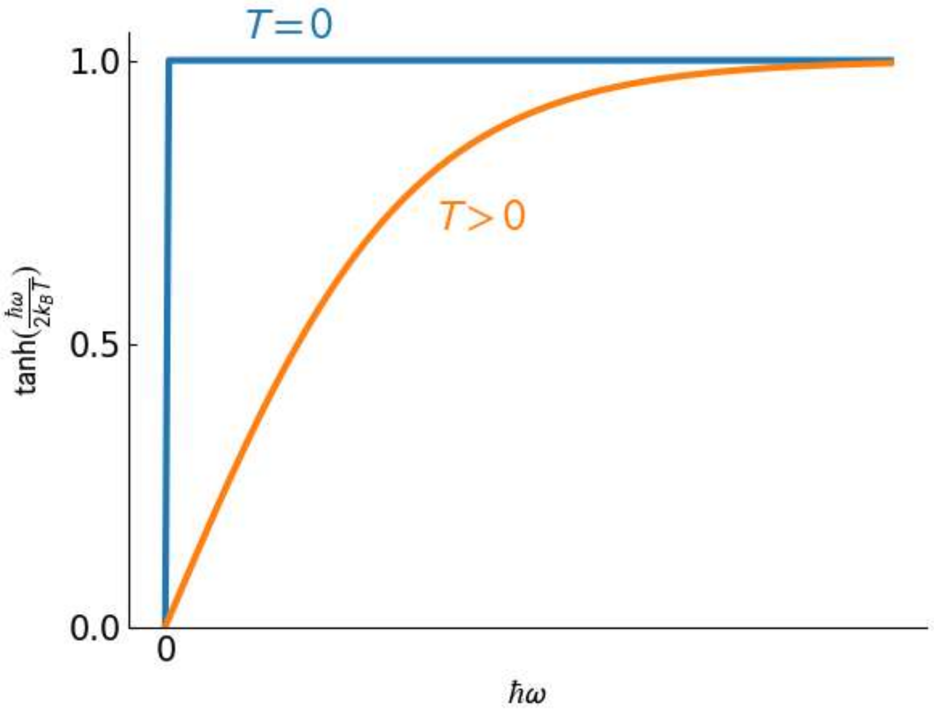
Can be related to dynamic susceptibility

$$\chi(\omega, T) = \frac{i}{\hbar}\int_0^\infty dt\, e^{i\omega t}\langle[\hat{\mathcal{O}}(t), \hat{\mathcal{O}}]\rangle$$

$$f_Q(T) = \frac{4\hbar}{\pi}\int_0^\infty \mathrm{d}(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\chi''(\hbar\omega, T)$$

Hauke et al, Nat. Phys. **12**, 778–782 (2016)

At T=0, thermal state QFI reduces to pure state QFI

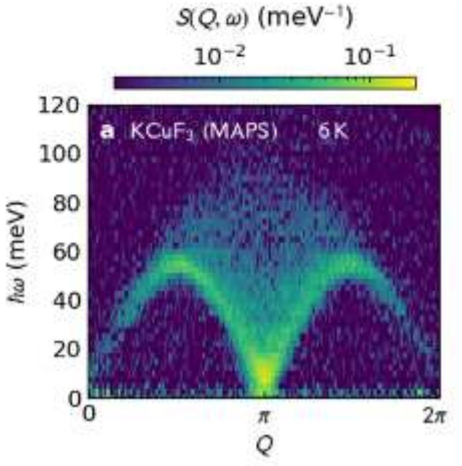


$$f_Q(T) = \frac{4\hbar}{\pi}\int_0^{\infty} \mathrm{d}\,(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\chi''(\hbar\omega, T)$$

T=0

$$f_Q = 4\left(\langle S^2\rangle - \langle S\rangle^2\right)$$

Quantum Fisher Information gives a lower bound on the number of entangled spins, and is measurable with neutron scattering



$$f_Q\left[\mathbf{Q},T\right]=4\int_0^\infty \mathrm{d}(\hbar\omega)\left[\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\left(1-e^{-\hbar\omega/k_BT}\right)S_{\alpha\alpha}(\mathbf{Q},\omega)\right]$$

By the Cramer Rao bound:

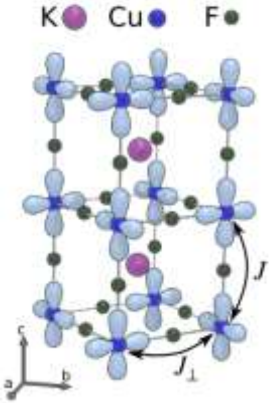
$$\mathrm{nQFI}=\frac{f_Q}{4S^2}>m$$

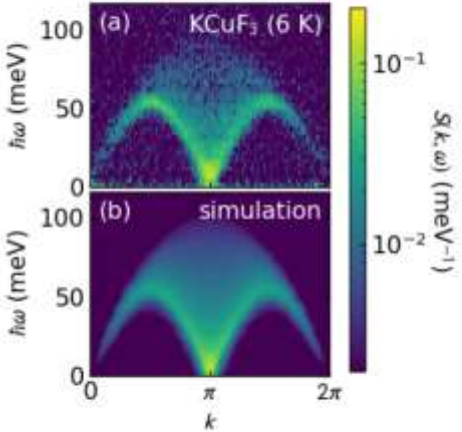
m = entanglement depth

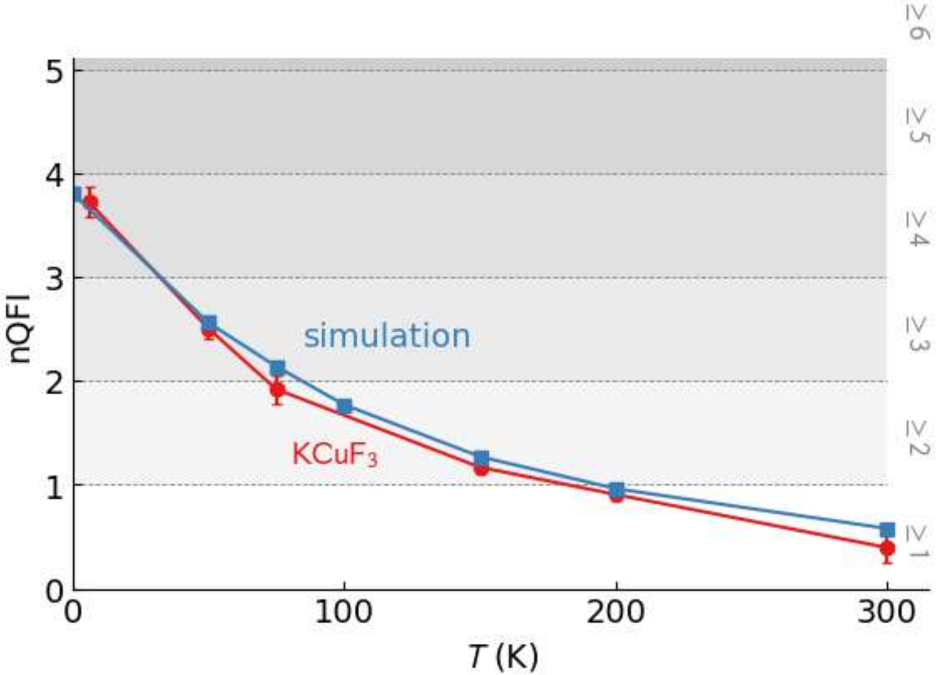
if a system has nQFI > *m*,
it is at least (*m*+1)-partite entangled.

Hauke et al, *N. Physics* (2016)

We tested QFI on 1D spin chain $KCuF_3$, witnessing ≥4 partite entanglement at low temperatures







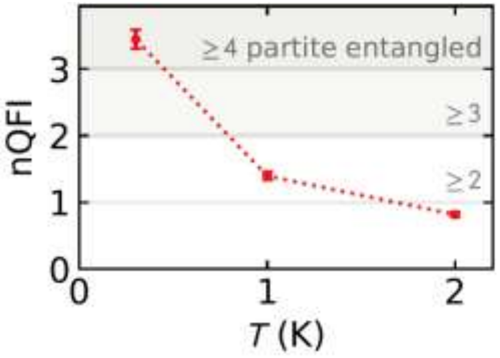
Los Alamos NATIONAL LABORATORY

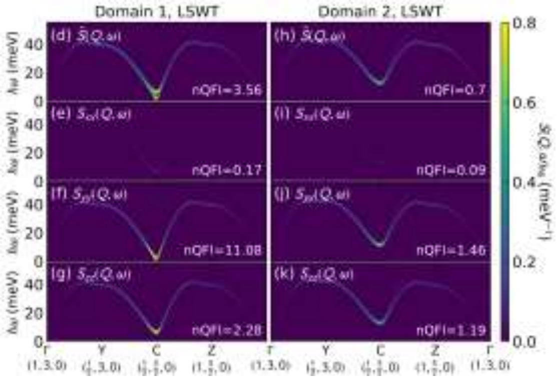
We've also measured quantum entanglement in

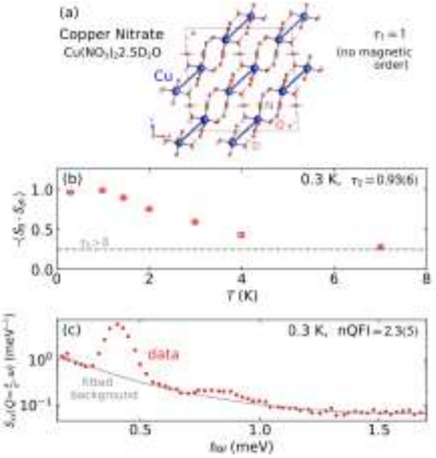
KYbSe$_2$

NiPS$_3$

Copper nitrate (dimer)







Scheie et al, *Materials Today Quantum* (2025)

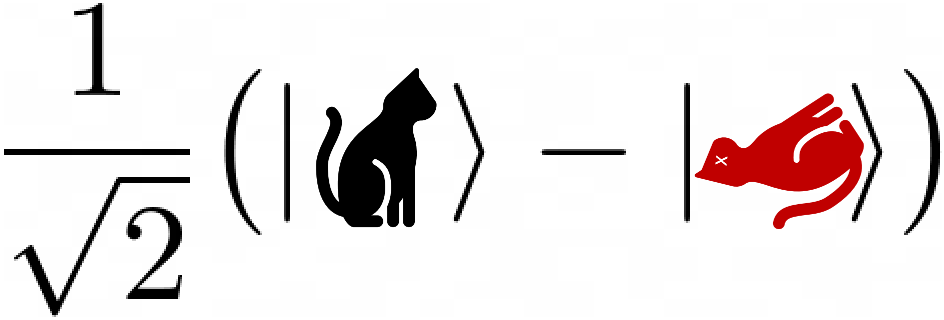
Is it crazy that integrating conventional neutron scattering tells us how many particles are entangled?

Some trivial examples demonstrate that nQFI does scale with entanglement:

$$\text{nQFI} = 4\left(\langle\psi|\mathcal{O}^\dagger\mathcal{O}|\psi\rangle - \langle\psi|\mathcal{O}|\psi\rangle^2\right)$$

wavefunction		operator	nQFI
Free spin	$\frac{1}{\sqrt{2}}(\|\uparrow\rangle + \|\downarrow\rangle)$	$\mathcal{O} = S^z$	1
Singlet	$\frac{1}{\sqrt{2}}(\|\uparrow\downarrow\rangle - \|\downarrow\uparrow\rangle)$	$\mathcal{O} = \sum_j S^z_j e^{i\pi r_j}$	2
GHZ	$\frac{1}{\sqrt{2}}(\|\uparrow\uparrow\uparrow\rangle - \|\downarrow\downarrow\downarrow\rangle)$	$\mathcal{O} = \sum_j S^z_j$	3
Cat state	$\frac{1}{\sqrt{2}}(\|\uparrow\downarrow \ldots N\rangle - \|\downarrow\uparrow \ldots N\rangle)$	$\mathcal{O} = \sum_j S^z_j e^{i\pi r_j}$	N

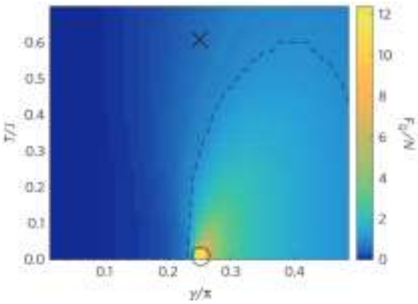
There’s a way in which the QFI measures the “cat-ness” of a ground state



Q: But what does QFI really tell us?

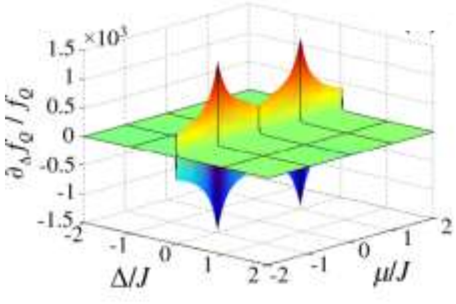
- Allows us to rule out trivial un-entangled states.
  - Doesn't necessarily rule out a magnon spectrum
- Diverging QFI seems to be related to quantum criticality

Transverse field Ising model



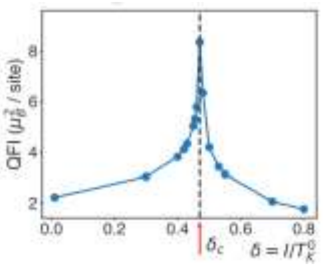
Hauke, N. Phys. (2016)

Kitaev chain model



Pezze, PRL (2017)

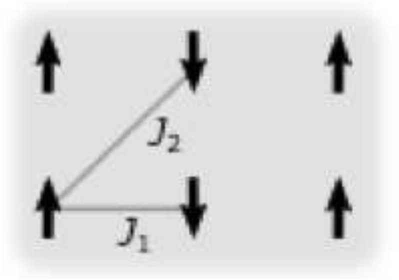
Kondo lattice model

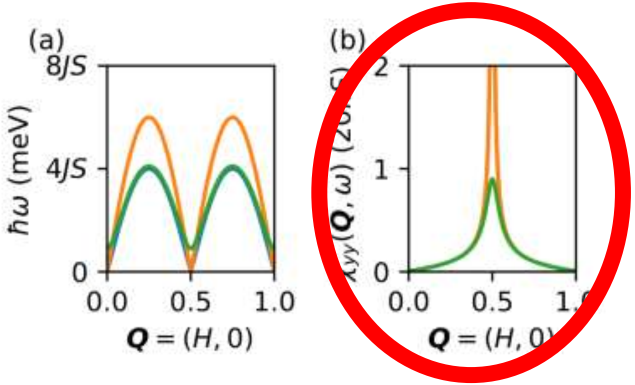


Fang, ArXiv:2402.18552 (2024)

Implication: many conventional antiferromagnets are highly entangled states!

2D Square lattice antiferromagnet: divergent inelastic intensity at ordering vector



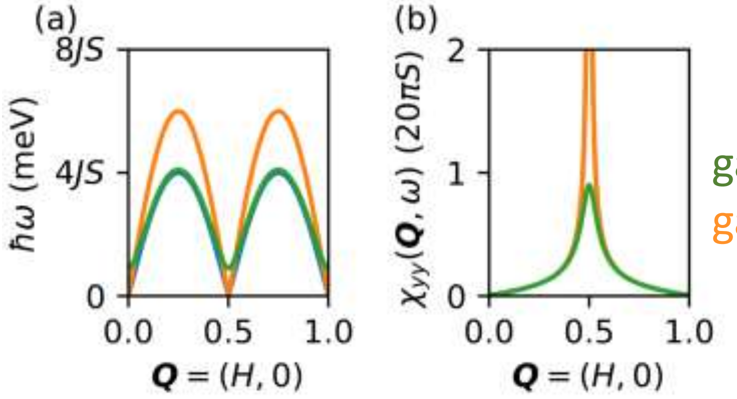


gapped
gapless

Implies divergent QFI $f_Q = 4\left(\langle\psi|S_\alpha^\dagger(\mathbf{Q})S_\alpha(\mathbf{Q})|\psi\rangle - \langle\psi|S_\alpha(\mathbf{Q})|\psi\rangle^2\right)$

What’s going on?

The Bogoliubov transform also diverges at the ordering wavevector, so the diagonal basis is dramatically rotated from the Neel basis (implying entanglement!)



gapped
gapless

$$u_{\boldsymbol{q}} = \cosh(\theta_{\boldsymbol{q}}) = \sqrt{\frac{A_{\boldsymbol{q}} + \hbar\omega_{\boldsymbol{q}}}{2\hbar\omega_{\boldsymbol{q}}}}$$

$$v_{\boldsymbol{q}} = \sinh(\theta_{\boldsymbol{q}}) = s \cdot \sqrt{\frac{A_{\boldsymbol{q}} - \hbar\omega_{\boldsymbol{q}}}{2\hbar\omega_{\boldsymbol{q}}}}$$

Forthcoming paper with Wolfgang Simeth
(LANL postdoc)

Summary of entanglement witnesses:

- We've successfully witnessed solid state entanglement using spectroscopy.
- These are *model-independent*: don't have to know or solve the underlying Hamiltonian.
- This doesn't tell us everything about the underlying quantum state, but it gives some valuable information.

Scheie, Laurell et al, PRB (2021)
Laurell, Scheie et al, PRL (2021)
Laruell et al, *Adv. Quant. Tech.* (2024)
Scheie et al, *Mat. Tod. Quant.* (2025)

But really, these are just baby steps.

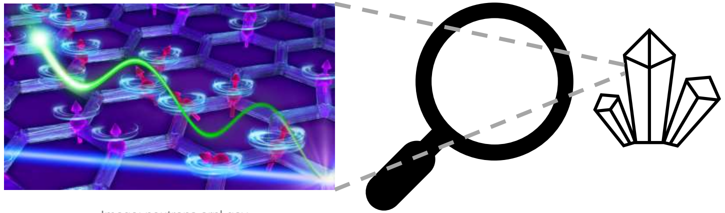


Image: neutrons.ornl.gov

Closing thoughts:

- Neutron scattering has historically revolutionized the way we think about condensed matter.

- This will continue to be the case: neutron scattering is one of the most direct ways to probe atomic scale quantum properties.

  - Quantum spin liquids
  - Many-body quantum entanglement and decoherence
  - Other states: unconventional superconductivity