Magnetic Raman scattering in quasi-one-dimensional antiferromagnets

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&

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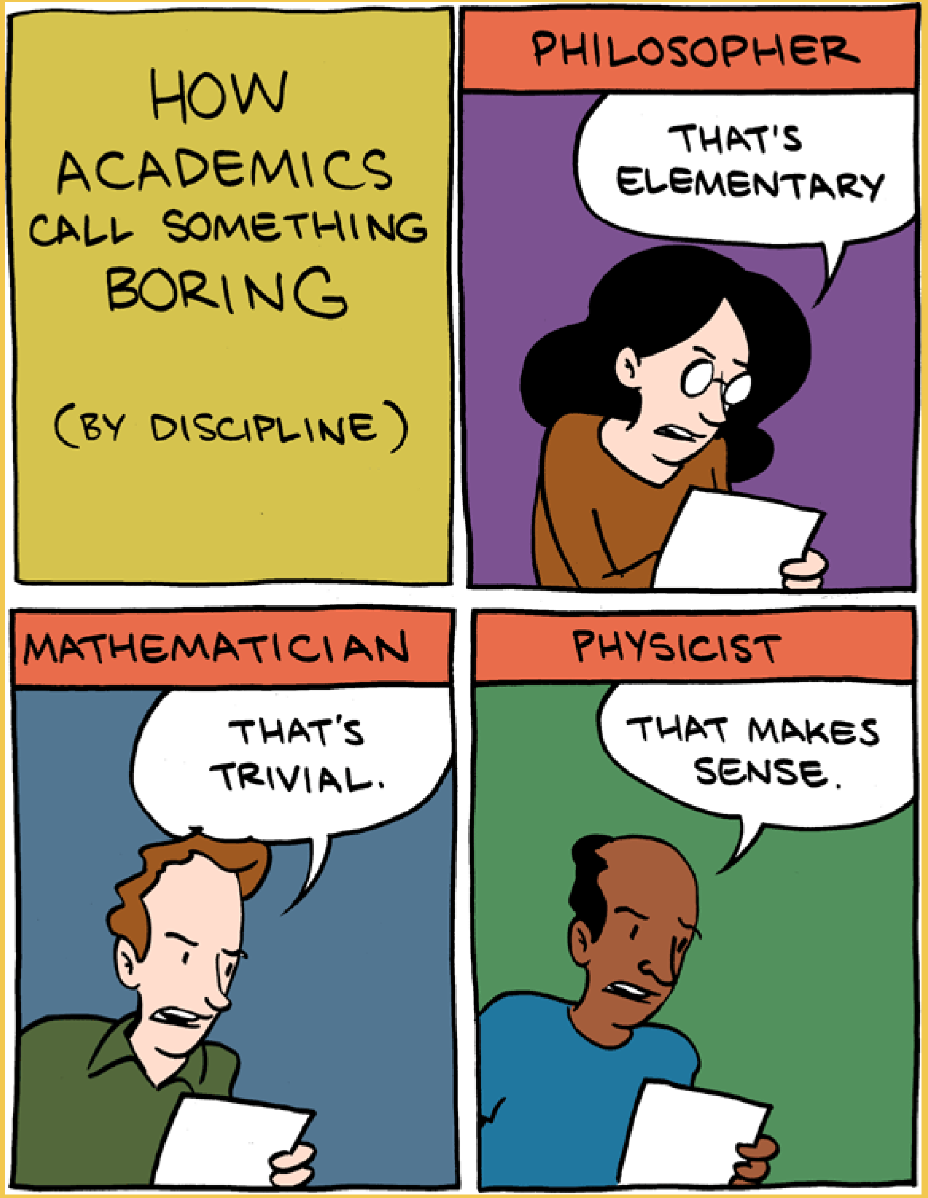
Quantum spin liquids

- 1D spin liquids → topologically trivial
  - No long range entanglement
- 2D spin liquids → exotic
  - (Potential) topological order



Zach Weinersmith
www.smbc-comics.com/comic/2011-10-23

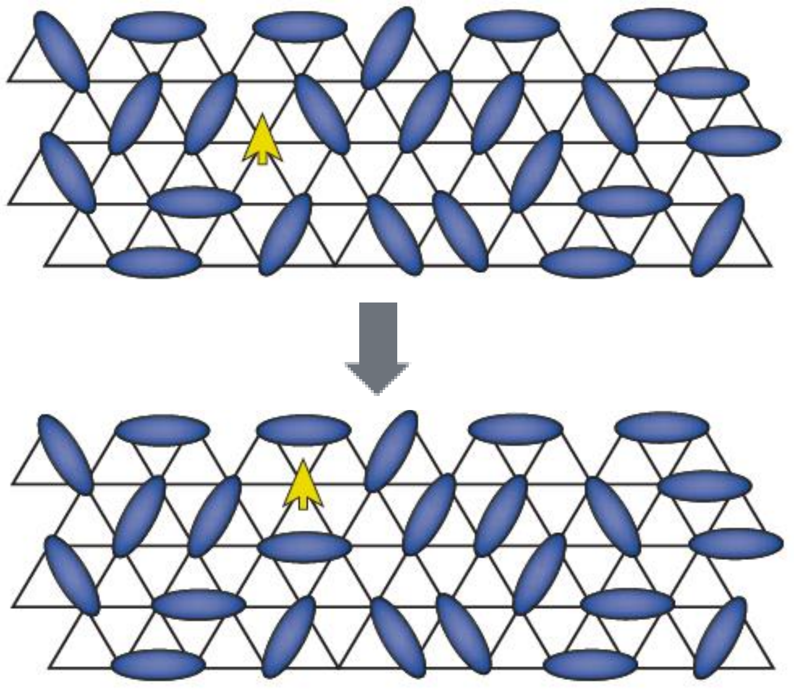
Fractional quasiparticles: The spinons

- Spinons - charge neutral, spin=1/2 quasiparticles

- Carry a **fraction** of an electron's quantum numbers

- Occur in:
  - 2D and 3D quantum spin liquids

Spinons in a 'Resonating Valence Bond' state

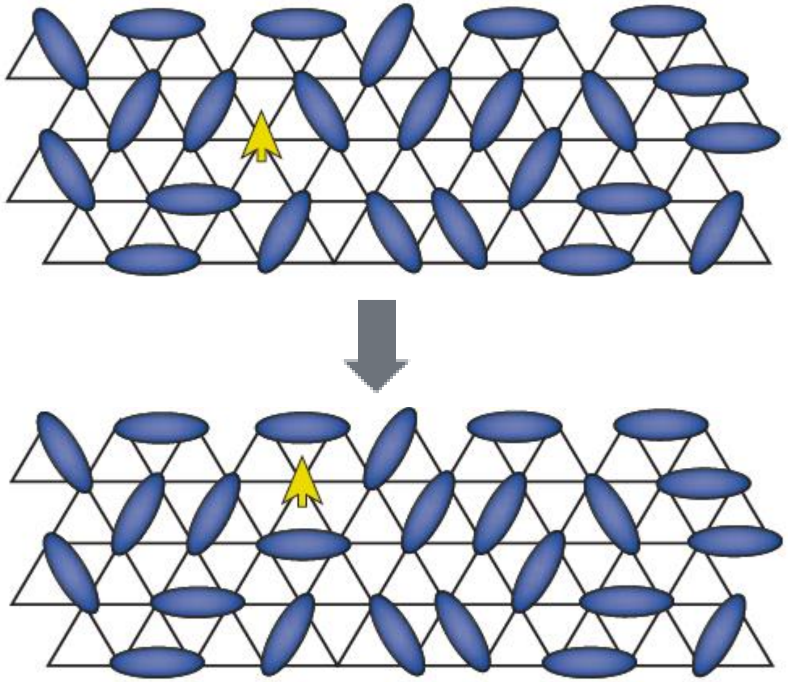


Balents (2010) *Nature*

Fractional quasiparticles: The spinons

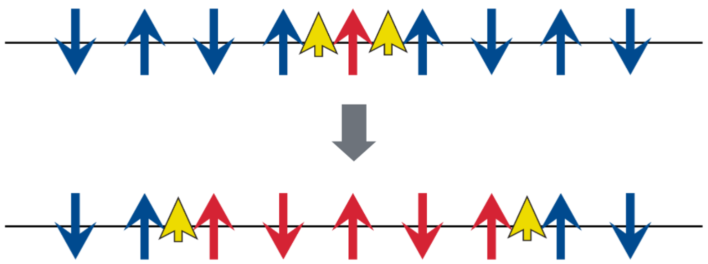
- Spinons - charge neutral, spin=1/2 quasiparticles
- Carry a **fraction** of an electron’s quantum numbers
- Occur in:
  - 2D and 3D quantum spin liquids
  - 1D antiferromagnetic spin=1/2 Heisenberg chain

Spinons in a ‘Resonating Valence Bond’ state

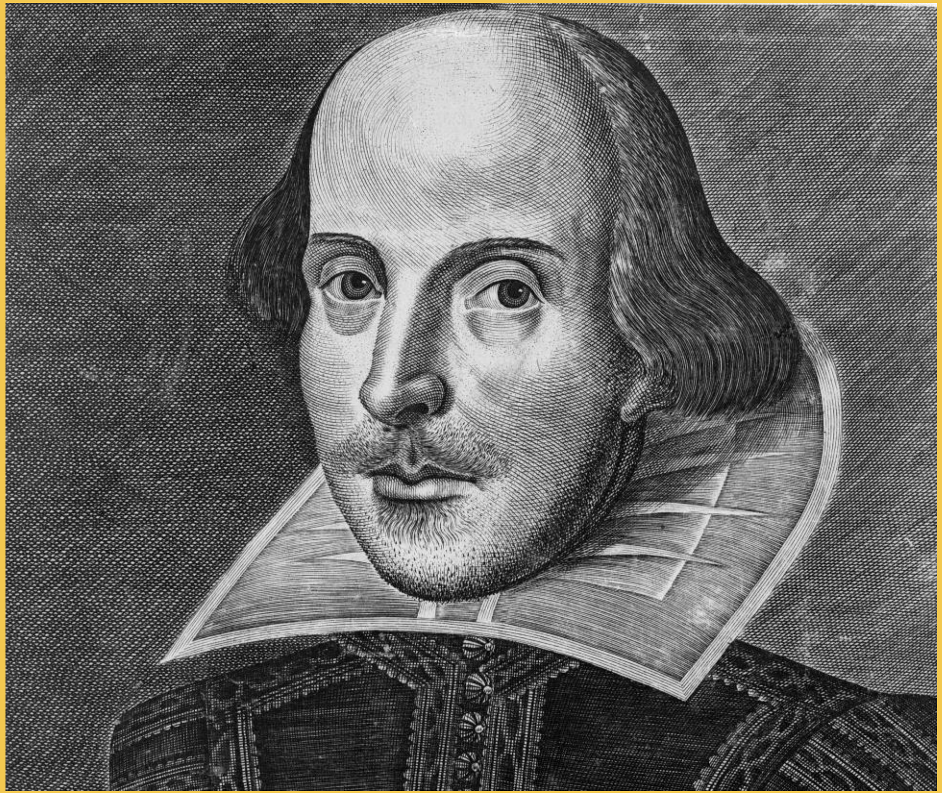


Balents (2010) *Nature*

Spinons in the Heisenberg chain



2D or not 2D?



William Shakespeare:
source of a terrible pun

Hans Bethe:
pioneer of 1D quantum magnetism

The 1D Heisenberg antiferromagnet

Heisenberg Hamiltonian

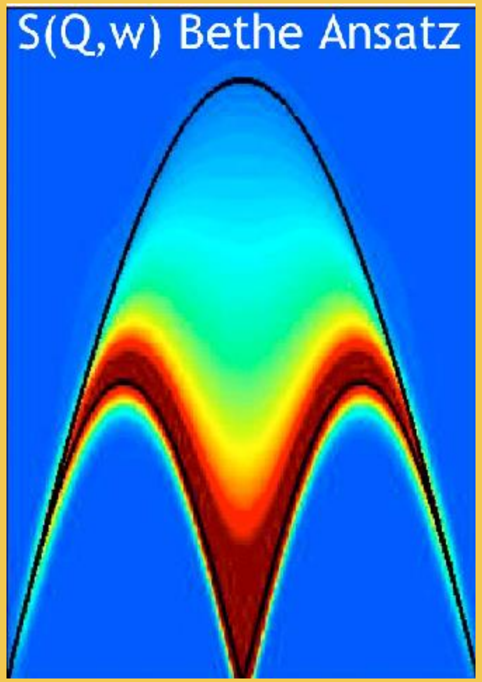
$$\hat{H} = J \sum_{i=1}^{N} \mathbf{S}_i \cdot \mathbf{S}_{i+1}$$

2-spinon dispersion

$$\omega = \pi J |\sin(q/2)\cos(p/2)|$$

- 1D, spin=1/2, Heisenberg Hamiltonian is diagonalizable
- Solutions given by the Bethe ansatz (e.g. the two-spinon dispersion)

The 2-spinon dynamic structure factor



Dynamic structure factor

$$S(Q,\omega) \equiv \int_{-\infty}^{+\infty} \frac{dt}{2\pi} e^{i\omega t} \langle S^z_{-Q}(t) S^z_Q(0) \rangle$$

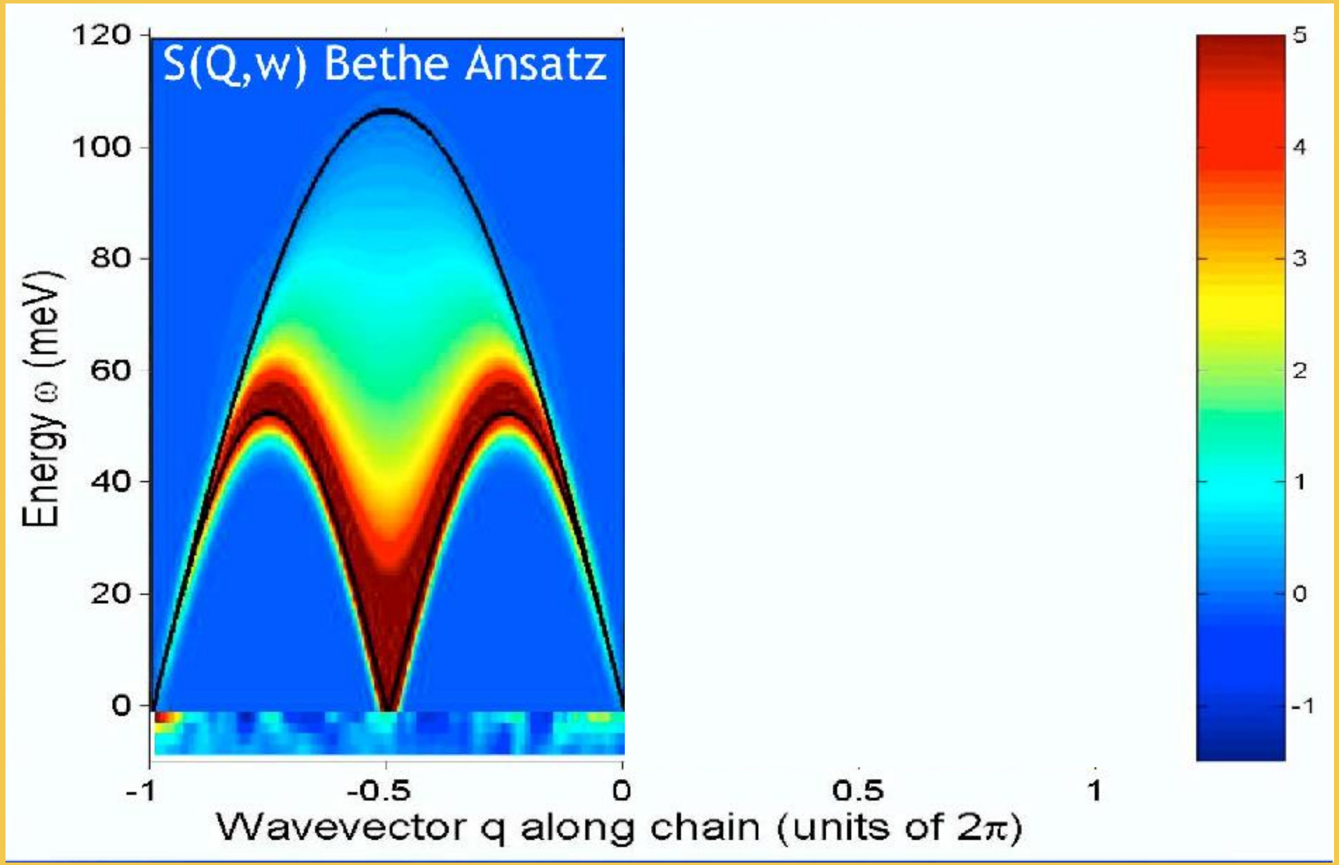
2-spinon dynamic structure factor

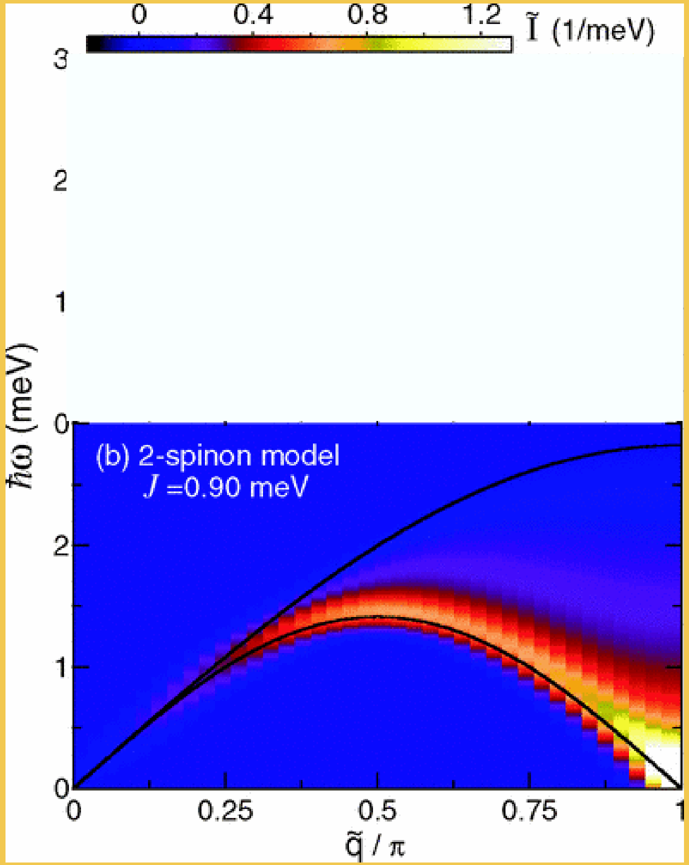
$$S^{2S}(Q,\omega) = \frac{1}{2} \frac{e^{-I(Q,\omega)}}{2\pi} D(Q,\omega)$$

$I(Q,\omega)$: transition function $D(Q,\omega)$: density of states

Lake *et al* (2013) *Phys. Rev. Lett.*

Neutron Scattering

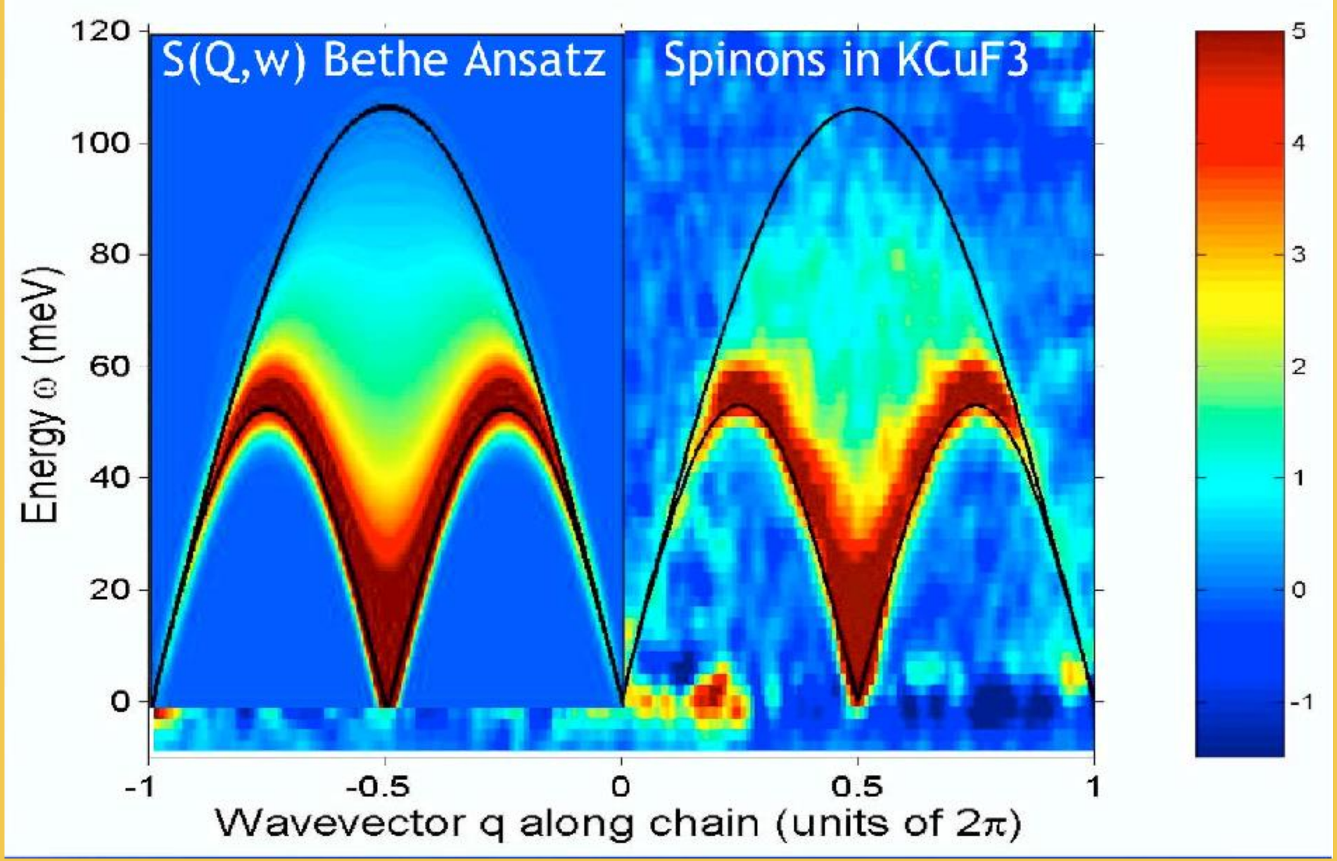




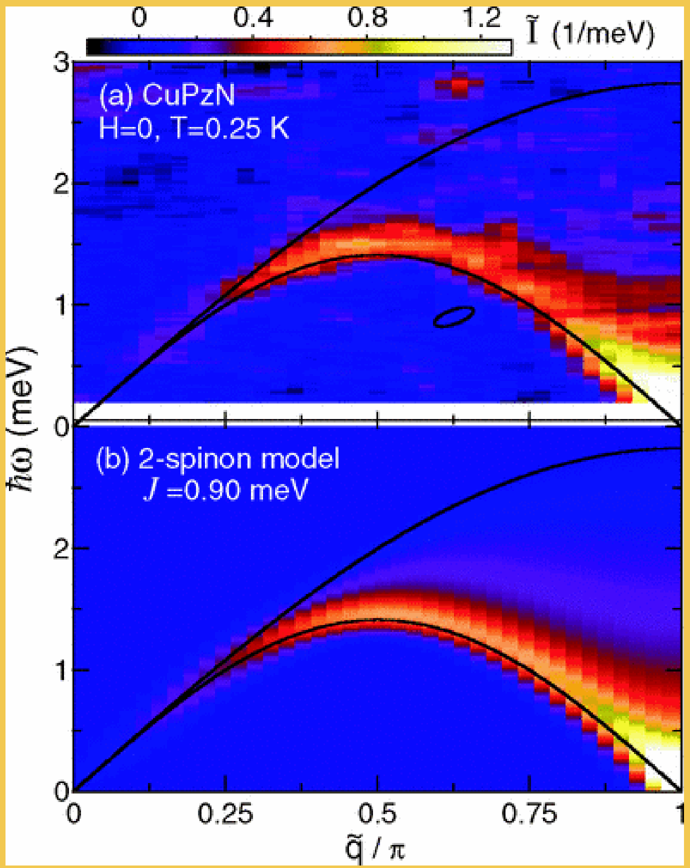
Lake *et al* (2013) *Phys. Rev. Lett.*

Stone *et al* (2003) *Phys. Rev. Lett.*

Neutron Scattering



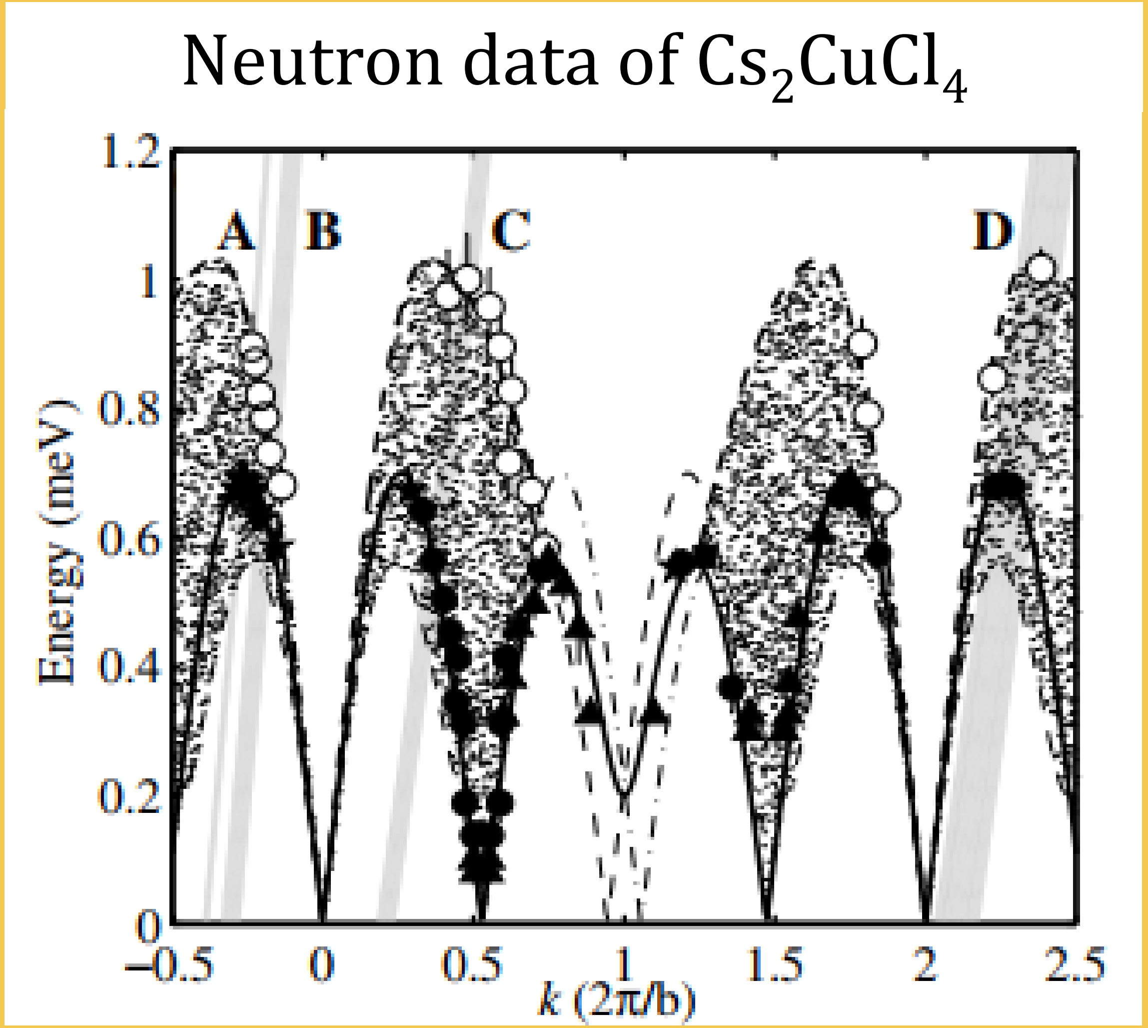
Potassium copper fluoride



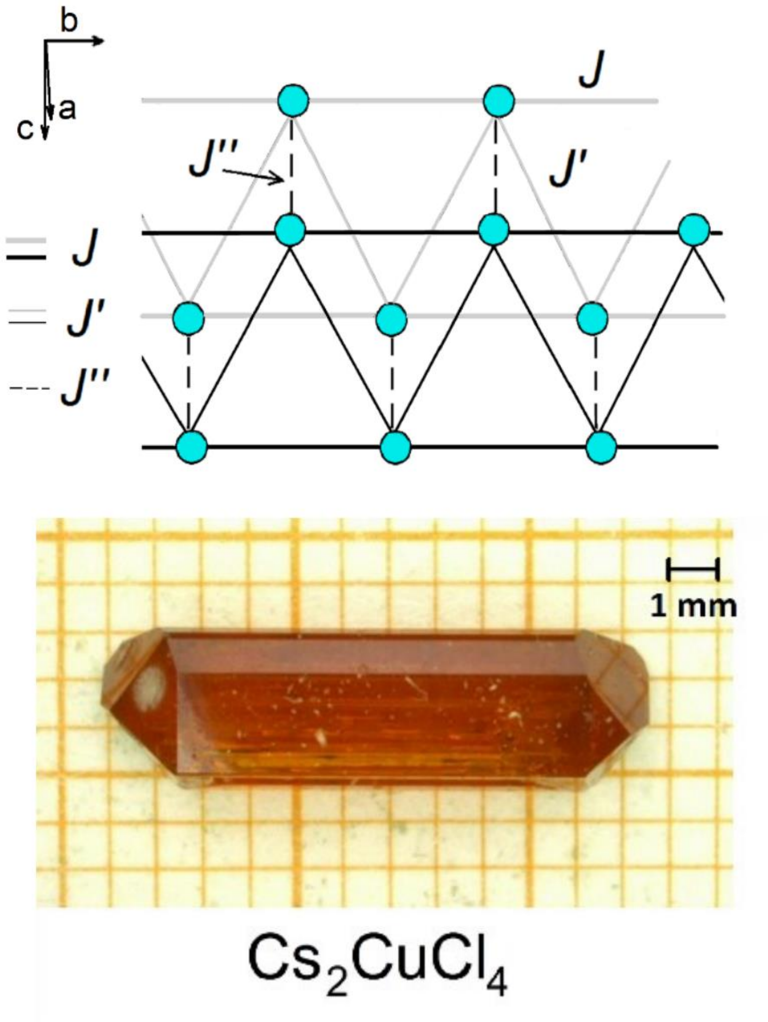
Copper pyrazine

Lake *et al* (2013) *Phys. Rev. Lett.* Tennant *et al* (1995) *Phys. Rev.* ***B*** Stone *et al* (2003) *Phys. Rev. Lett.*

Cs_2CuCl_4



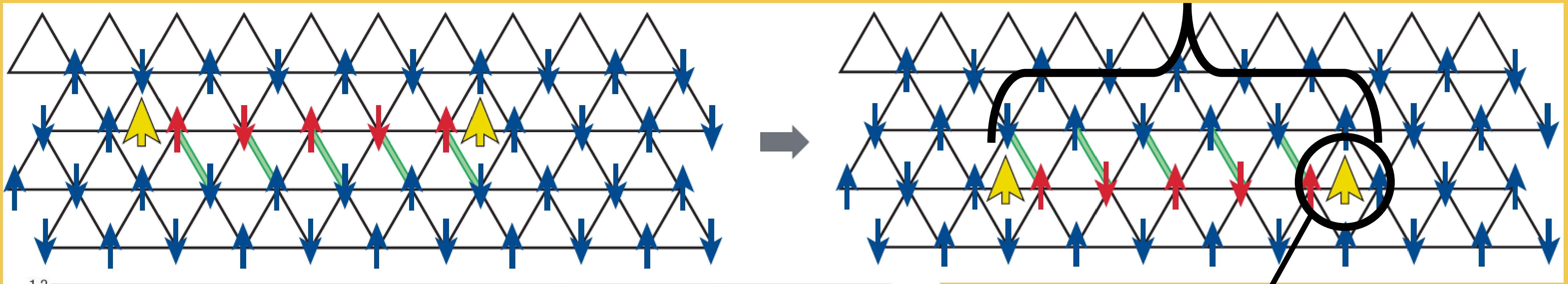
Coldea *et al* (2001) *Phys. Rev. Lett.*



van Well *et al* (2018) *Annalen der Physik*

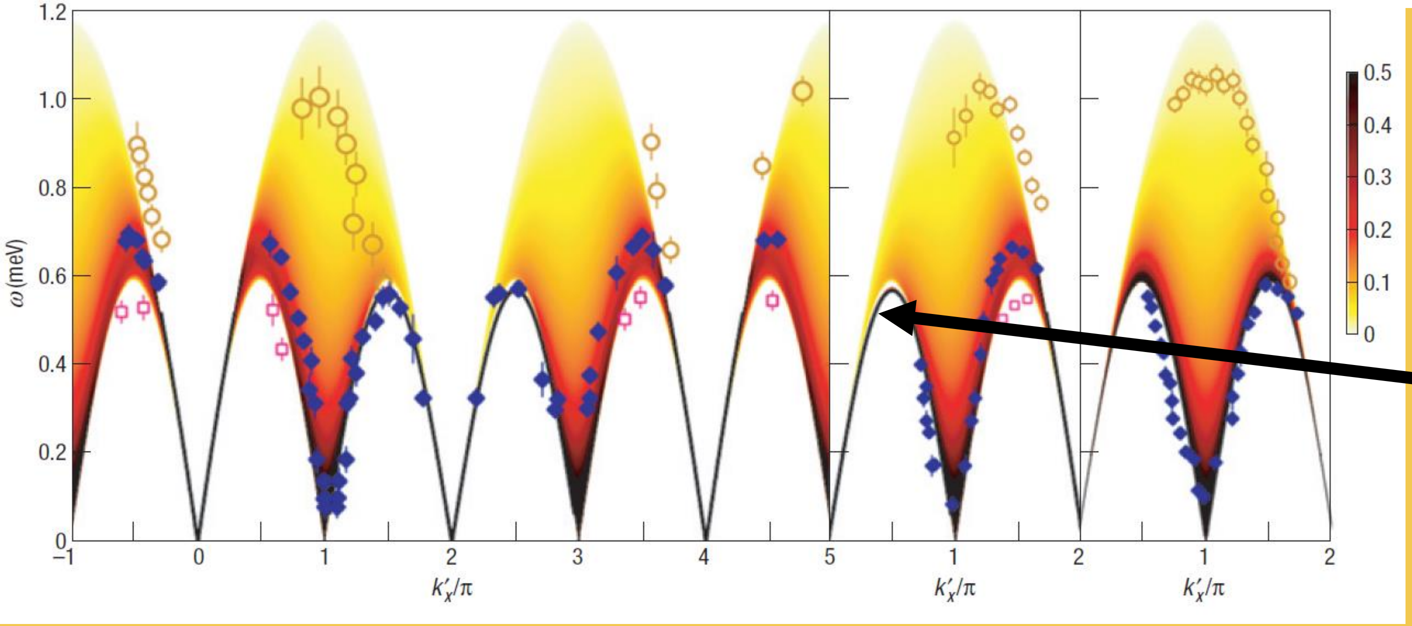
What are triplons?

Triplon, spin=1



Balents (2010) *Nature*

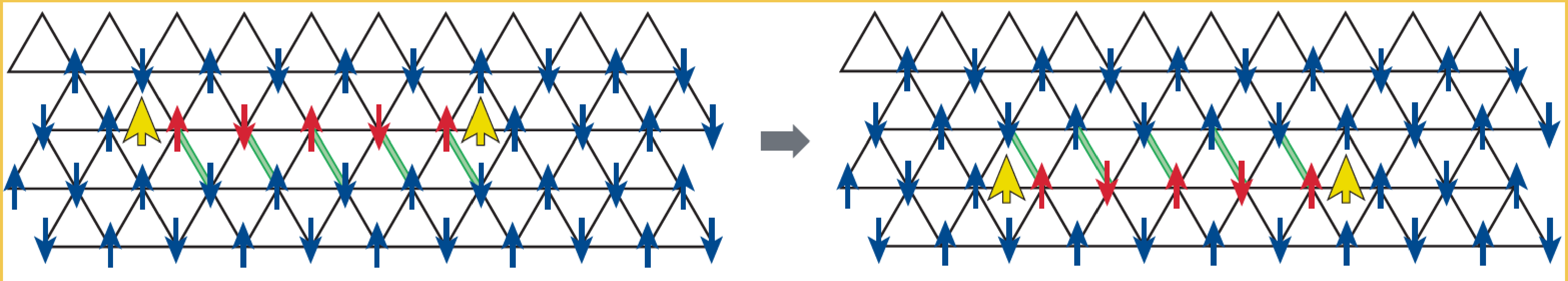
Spinon, spin=1/2



Sharp, black line is the triplon peak

Kohno *et al* (2007) *Nat. Phys.*

What are triplons?



Balents (2010) *Nature*

- **1D spinons can manifest as 2D ‘triplons’**
- **Kohno, Starykh & Balents showed Cs_2CuCl_4 is not a spin liquid**
- **The line between 1D and 2D is blurry**

A ‘small’ problem

- Small sample sizes are insufficient for neutron scattering
- Samples need to be ~cm^3
- Problem for A_2IrO_3 (A = Li, Na)
- Problem for κ-$(BEDT\text{-}TTF)_2\,Cu_2(CN)_3$

Crystal data of $(BEDT\text{-}TTF)_4[Cu(NCS)_4]$

Crystal data	
Chemical formula	$(C_{10}H_8S_8)_2[Cu(CNS)_4]\cdot 2C_{10}H_8S_8$
M_r	1834.43
Crystal system, space group	Monoclinic, $P2_1/c$
Temperature (K)	100
a, *b*, *c* (Å)	16.9036 (17), 21.004 (2), 9.6205 (9)
β (°)	103.071 (3)
V ($Å^3$)	3327.1 (6)
Z	2
Radiation type	Mo *K*α
μ (mm^{-1})	1.50
Crystal size (mm)	0.18 × 0.16 × 0.02

Faulmann *et al* (2018) Acta Cryst. Sec. **E**

Crystal size (mm) 0.18 × 0.16 × 0.02

A ‘small’ problem: solutions

1. Grow a bigger crystal

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2. Mosaic the small crystals into a big one

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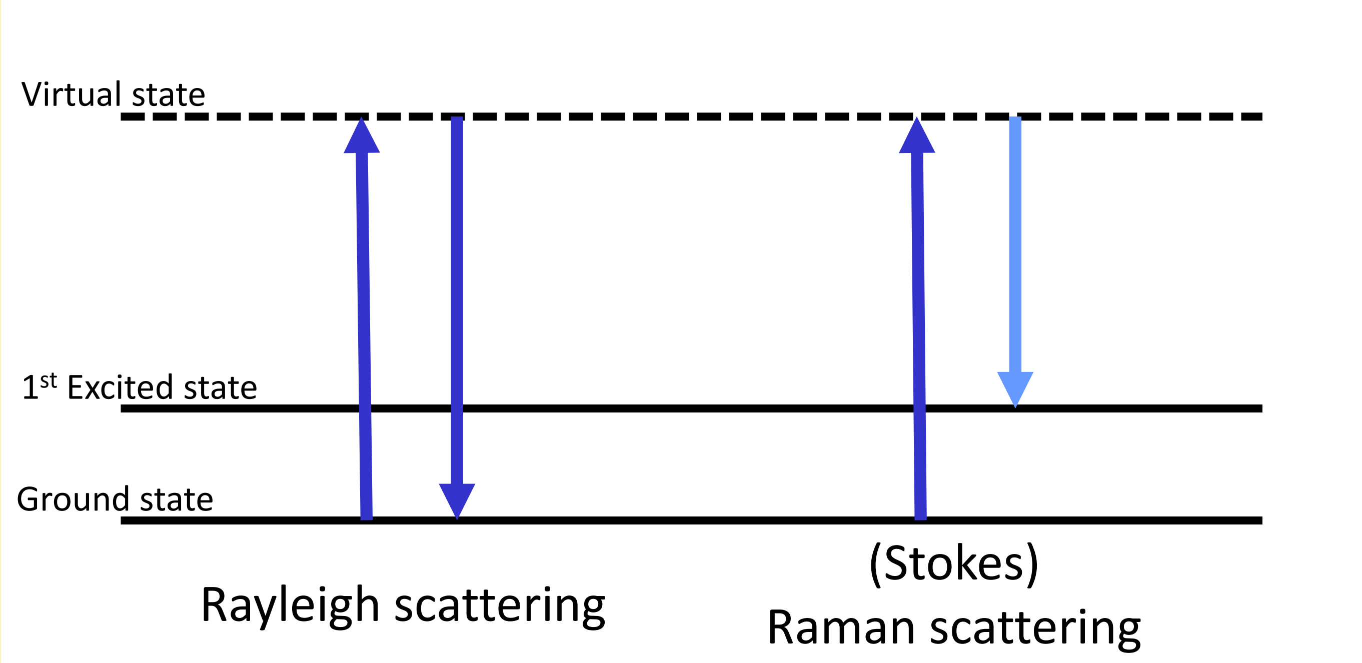
3. Tighter neutron beams, higher neutron flux

A ‘small’ problem: solutions

1. Grow a bigger crystal
2. Mosaic the small crystals into a big one
3. Tighter neutron beams, higher neutron flux
4. **Use another method**

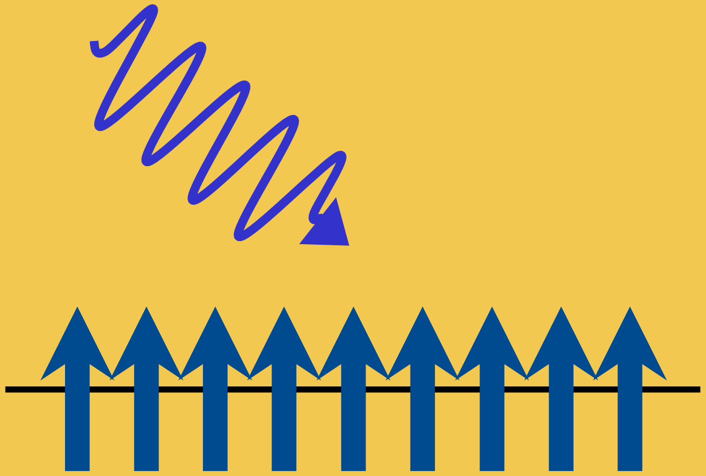
Magnetic Raman scattering

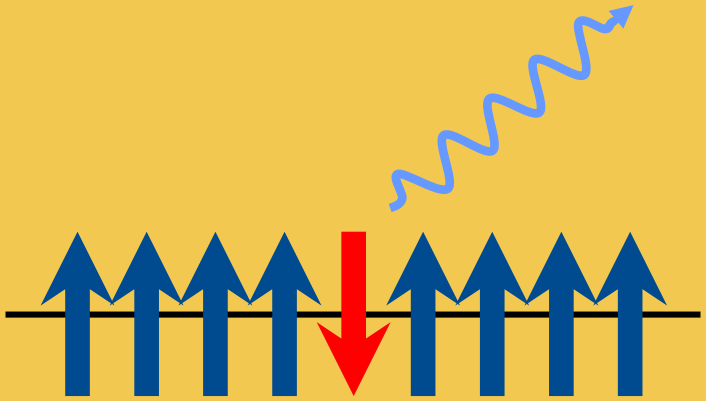


Rodney Loudon | Paul A. Fleury

Demonstrated the mechanism of inelastic light scattering from magnetic excitations in ferromagnets and antiferromagnets

R. Loudon, P. A. Fleury (1968)
DOI: 10.1103/PhysRev.166.514





Magnetic Raman scattering

Property	Inelastic neutron scattering	Raman scattering
q-resolution	over an entire BZ	$q \approx 0$ for first-order SP; entire BZ for higher-order SP
Energy range	a few μeV -20 eV	1 meV–1 eV
Energy resolution	5%–10% of incident neutron energies	<0.2 meV
Typical acquisition time per spectrum	(1) 3–5 h for powders (2) a few days for $\mathcal{S}_{\text{tot}}$ in single crystals	1–10 min
Sample volume	Several grams	μm-sized bulk and thin films
Magnetic field range	0–15 T	0–45 T
Selection rules	$\Delta S_z = \pm 1$	$\Delta S_z = 0$
Assignment of phonon and magnetic excitations	q dependence	Temperature and polarization dependence

Wulferding *et al* (2019) *J. Phys. Condens. Matter*

Magnetic Raman scattering

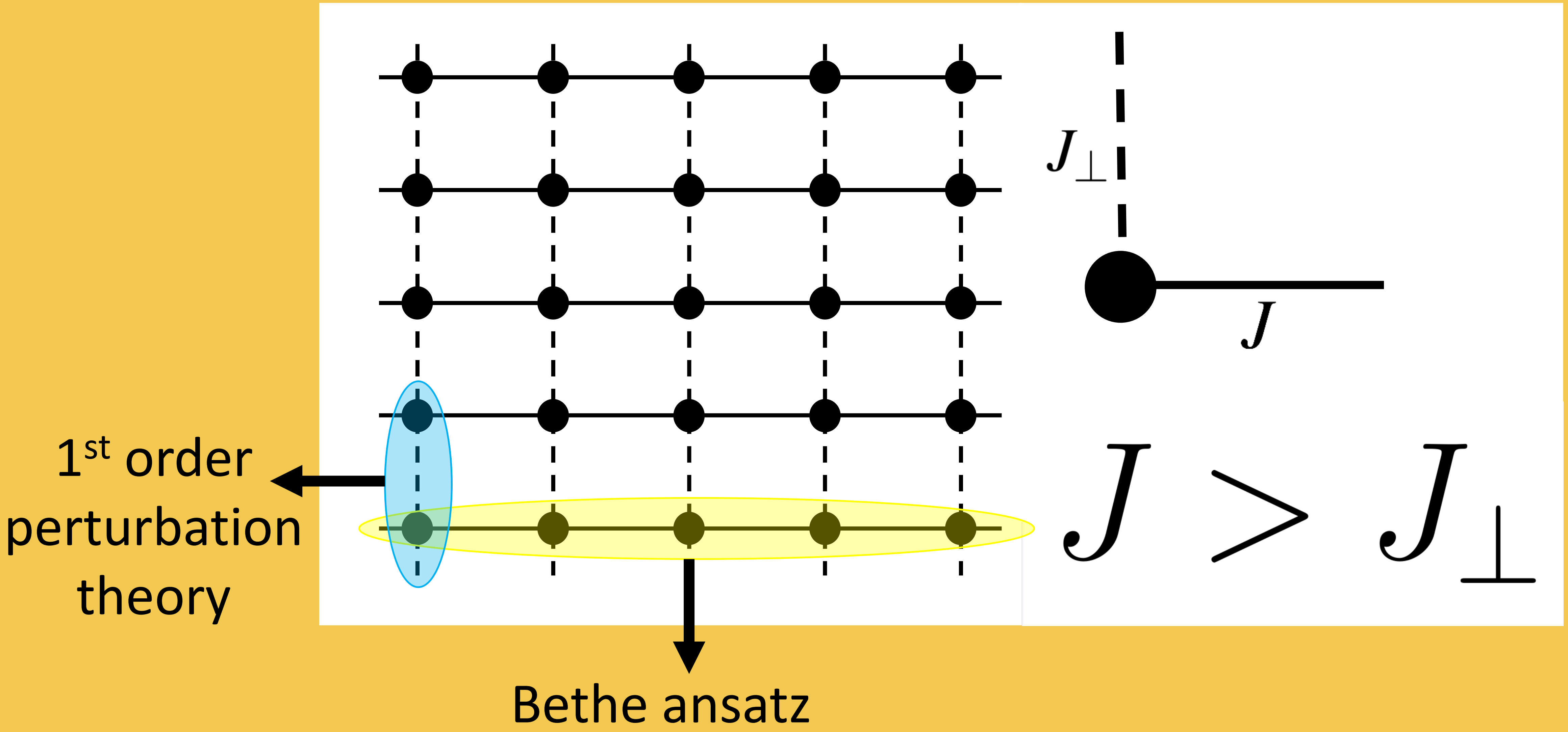
Property	Inelastic neutron scattering	Raman scattering
Spectroscopic quantity	$S(\mathbf{k},\omega)$	$I(\omega) \propto \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \hat{R}(t)\hat{R} \rangle$
Sample volume	Several grams	μm-sized bulk and thin films

The exchange scattering (two-magnon) Raman operator → $\hat{R} = \sum_{i,j,\hat{\delta}} (\hat{\varepsilon}_{\text{in}} \cdot \hat{\delta})(\hat{\varepsilon}_{\text{out}} \cdot \hat{\delta}) J_{\hat{\delta}} \vec{S}_{i,j} \cdot \vec{S}_{i+\delta_i,j+\delta_j}$

Wulferding *et al* (2019) *J. Phys. Condens. Matter*

What does a quasi-one-dimensional Raman spectra look like?

Model: Rectangular lattice



Solution classes

Structure factor of the rectangular lattice

$$S(\mathbf{k},\omega) = \underbrace{S^{\mathrm{S}}(\mathbf{k},\omega)}_{\text{Spinon}} + \overbrace{S^{\mathrm{T}}(\mathbf{k},\omega)}^{\text{Triplon}}$$

Solution classes

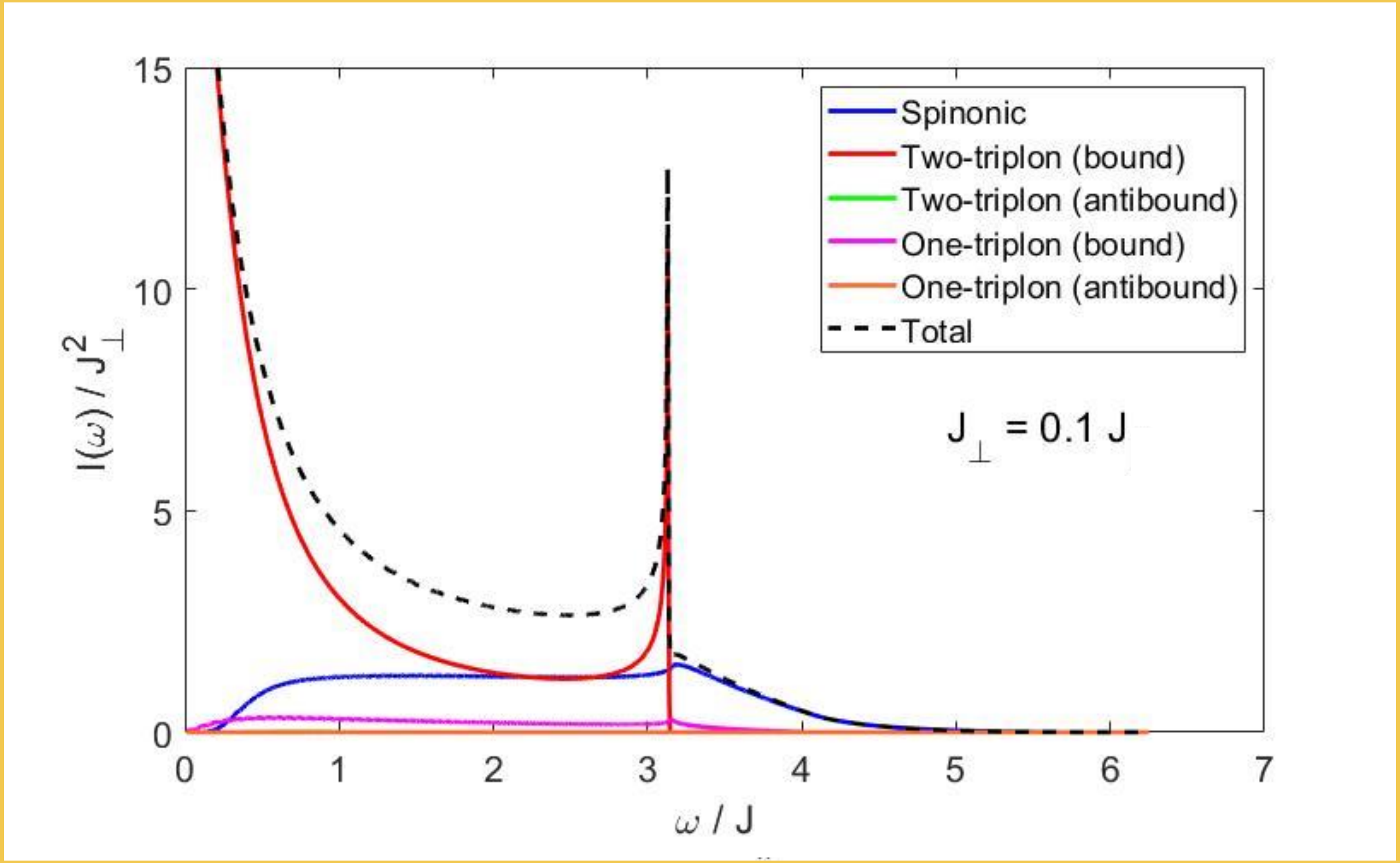
Structure factor of the rectangular lattice

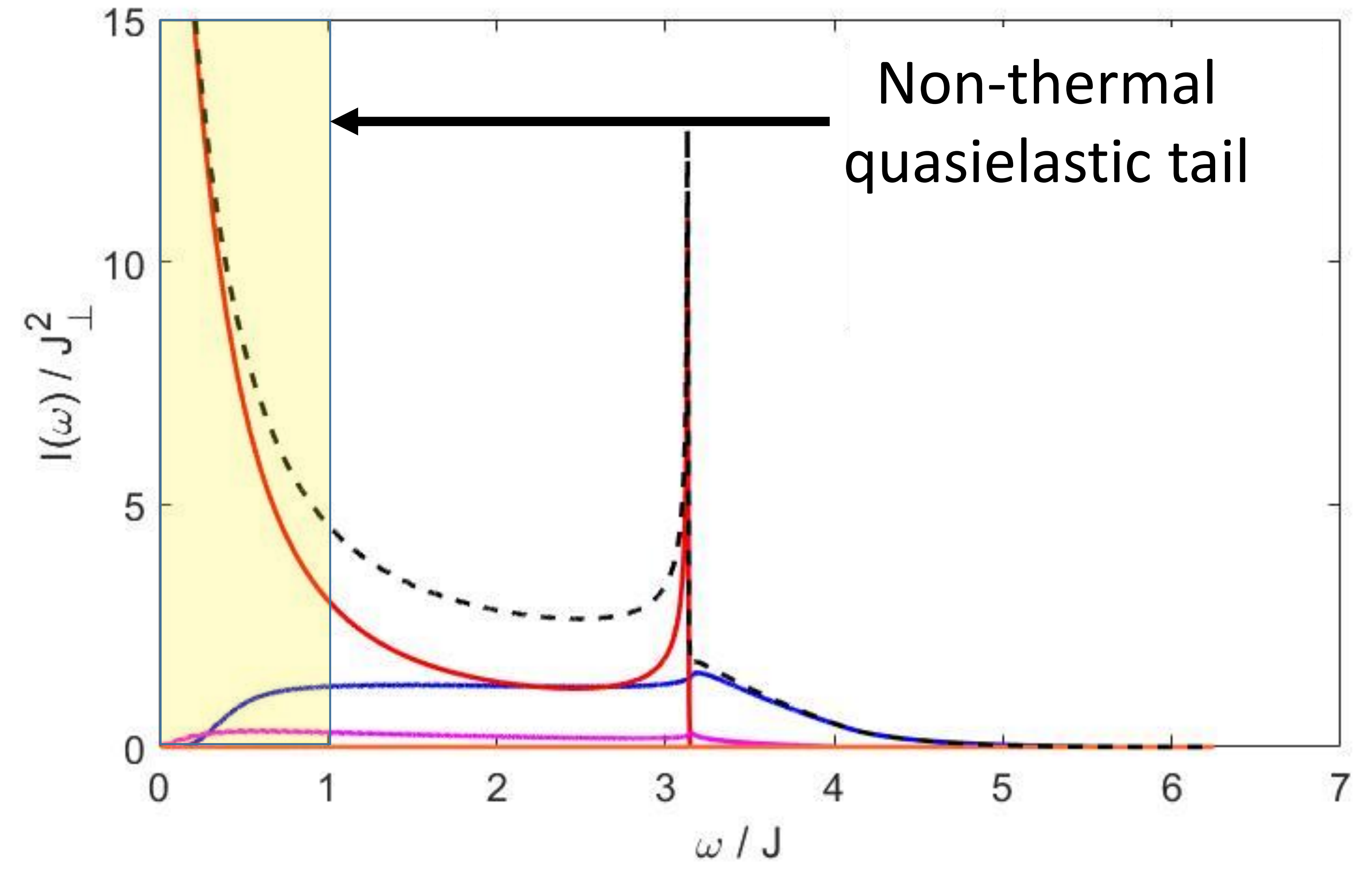
$$S(\mathbf{k},\omega) = \underbrace{S^{\mathrm{S}}(\mathbf{k},\omega)}_{\text{Spinon}} + \overbrace{S^{\mathrm{T}}(\mathbf{k},\omega)}^{\text{Triplon}}$$

Raman intensity of the rectangular lattice

$$I(\omega) = \underbrace{I^{\mathrm{S}}(\omega)}_{\text{Spinonic}} + \overbrace{I^{1\mathrm{T}}(\omega)}^{\text{One triplon}} + \underbrace{I^{2\mathrm{T}}(\omega)}_{\text{Two triplon}}$$

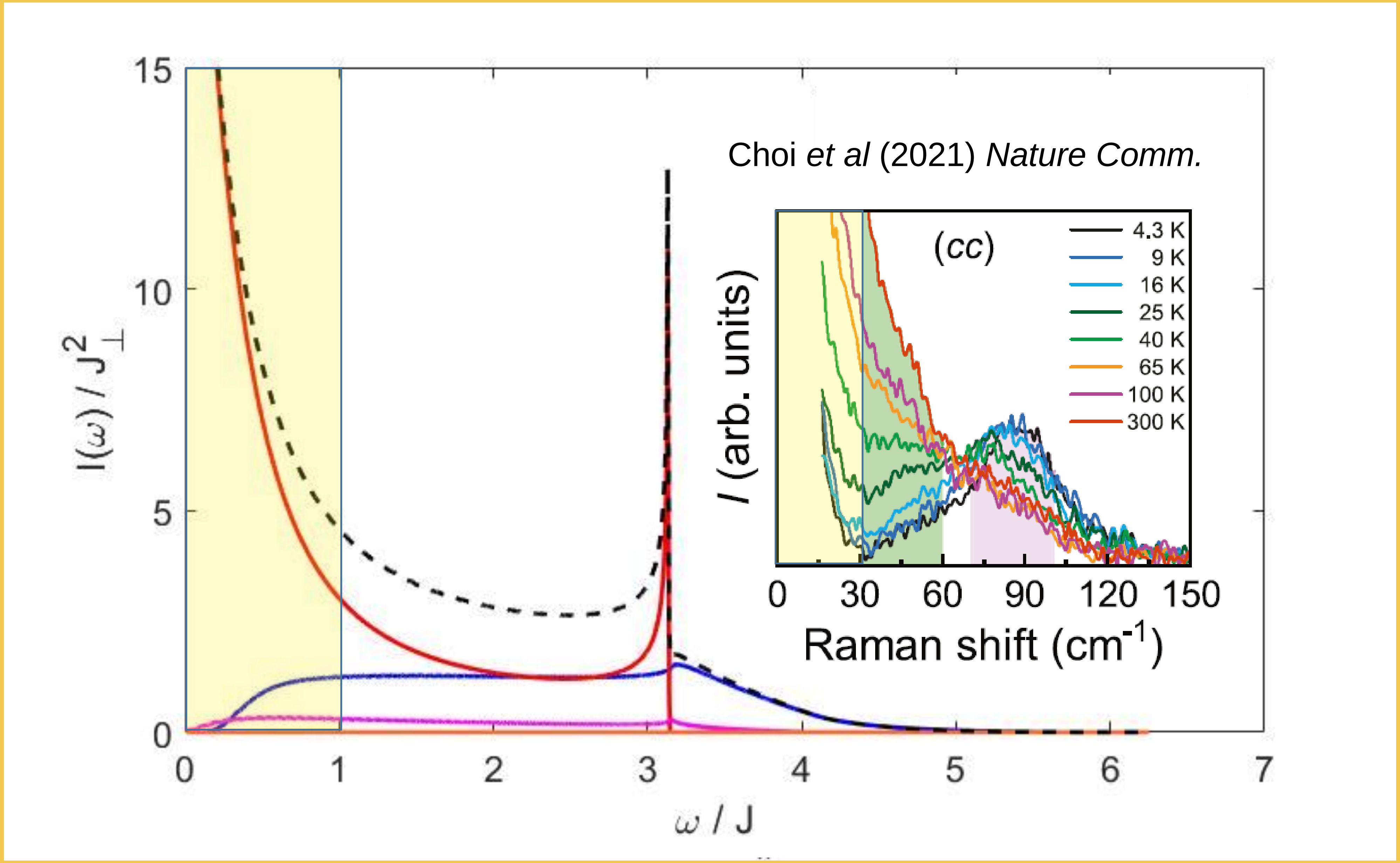
Raman intensity



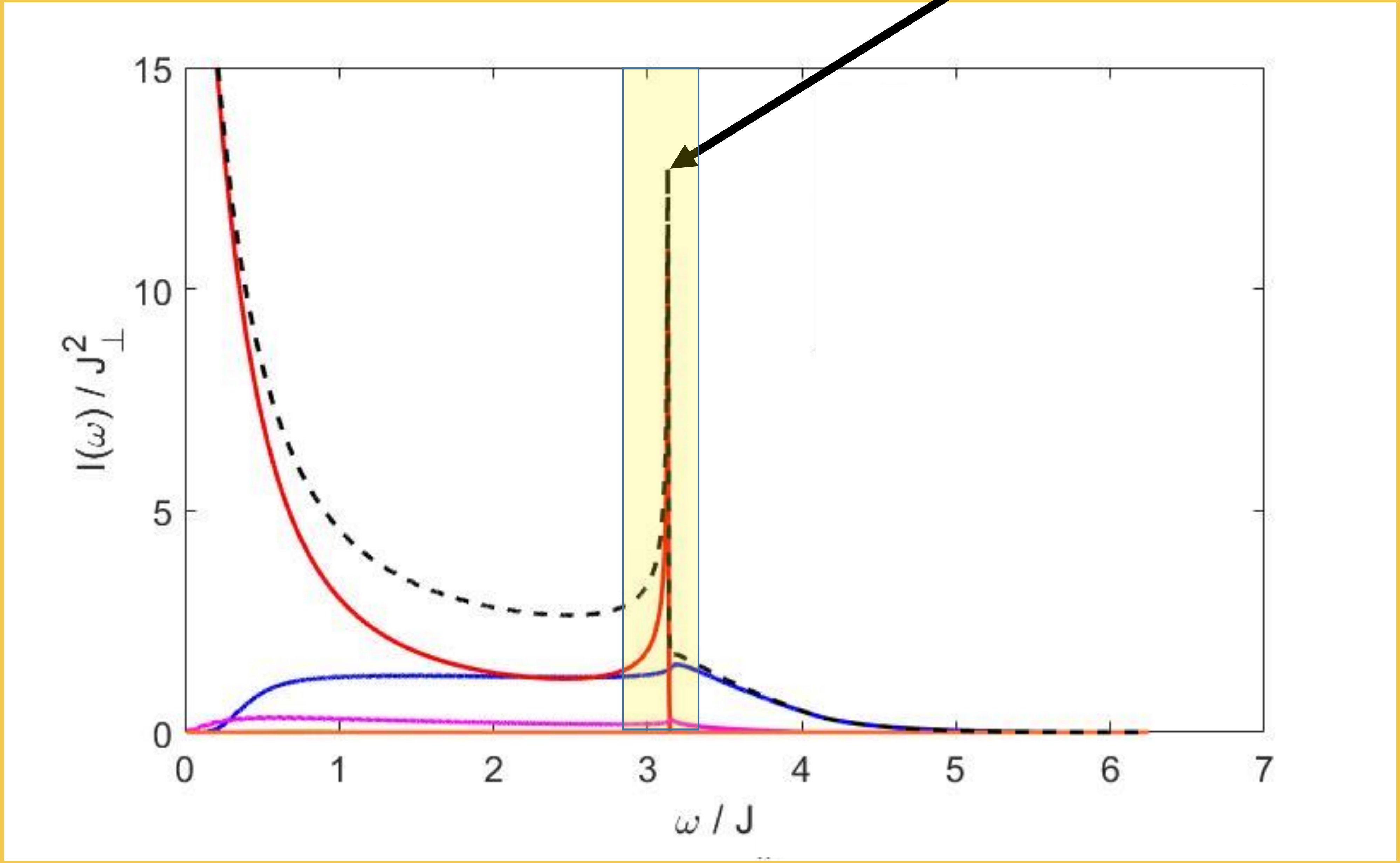


Non-thermal
quasielastic tail
I(ω) / J⊥²
ω / J
15
10
5
0
0
1
2
3
4
5
6
7

$Ca_3ReO_5Cl_2$ J ~ $30 cm^{-1}$



(πJ peak) van-Hove singularity



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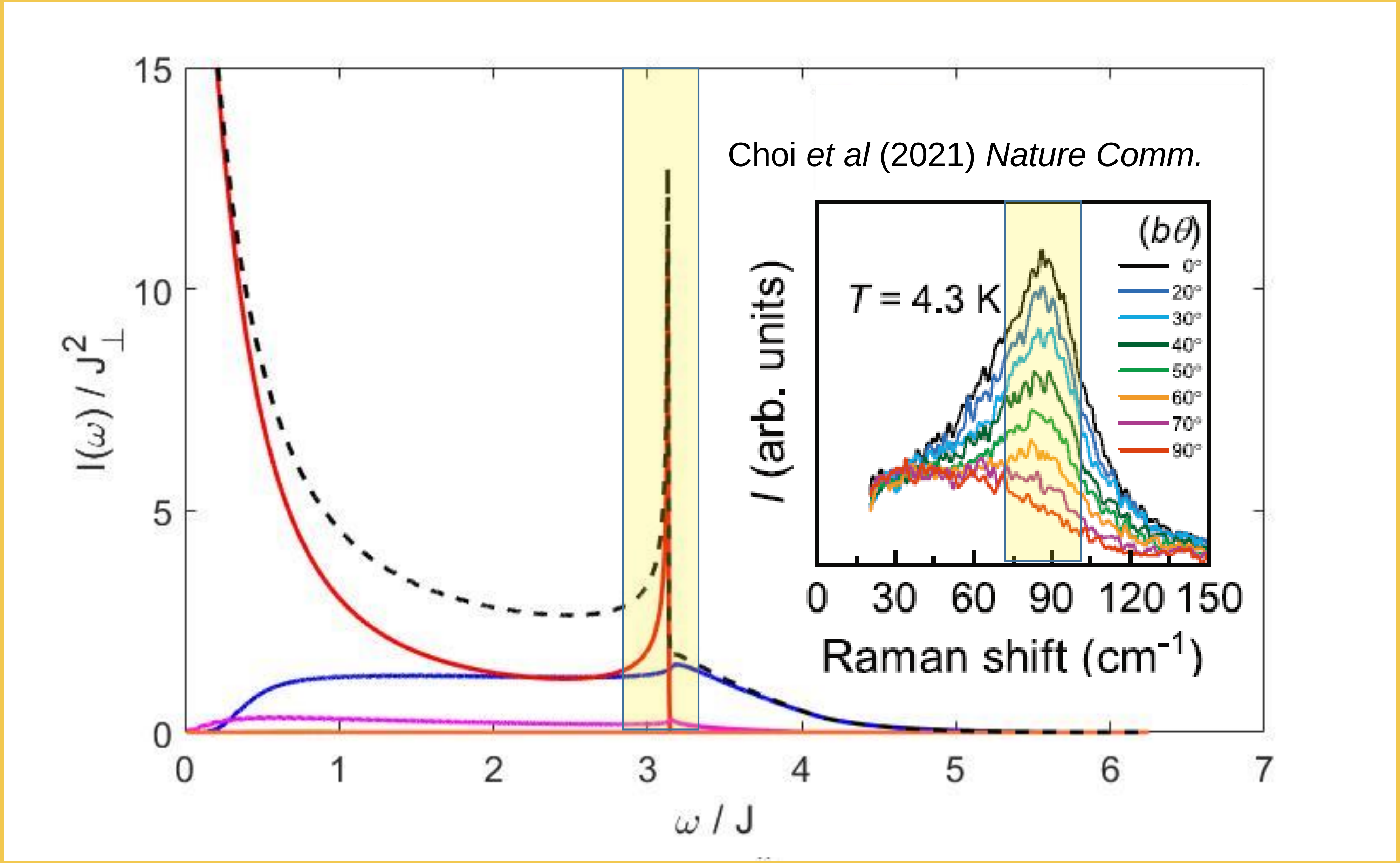

Summary

1. **Classifying dimensionality of spin liquids is important**
2. **Raman scattering is a potential stand-in for neutron scattering**
3. **1D Raman signatures are the low-T quasielastic tail and the πJ peak**