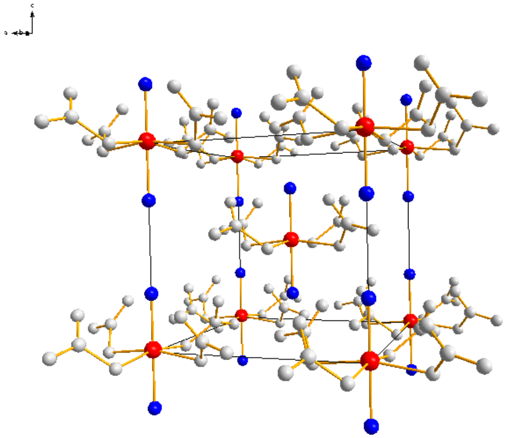
PSI Master School 2017

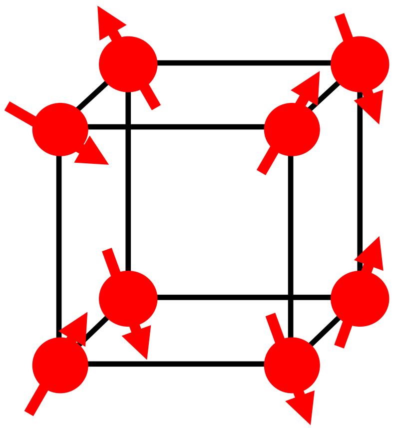
Introducing photons, neutrons and muons for materials characterization

Lecture 14: Neutron Spectroscopy and Magnetic Excitations

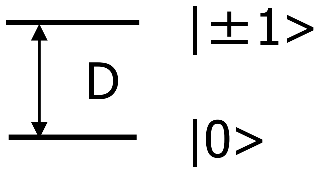
Simple example of a quantum magnet



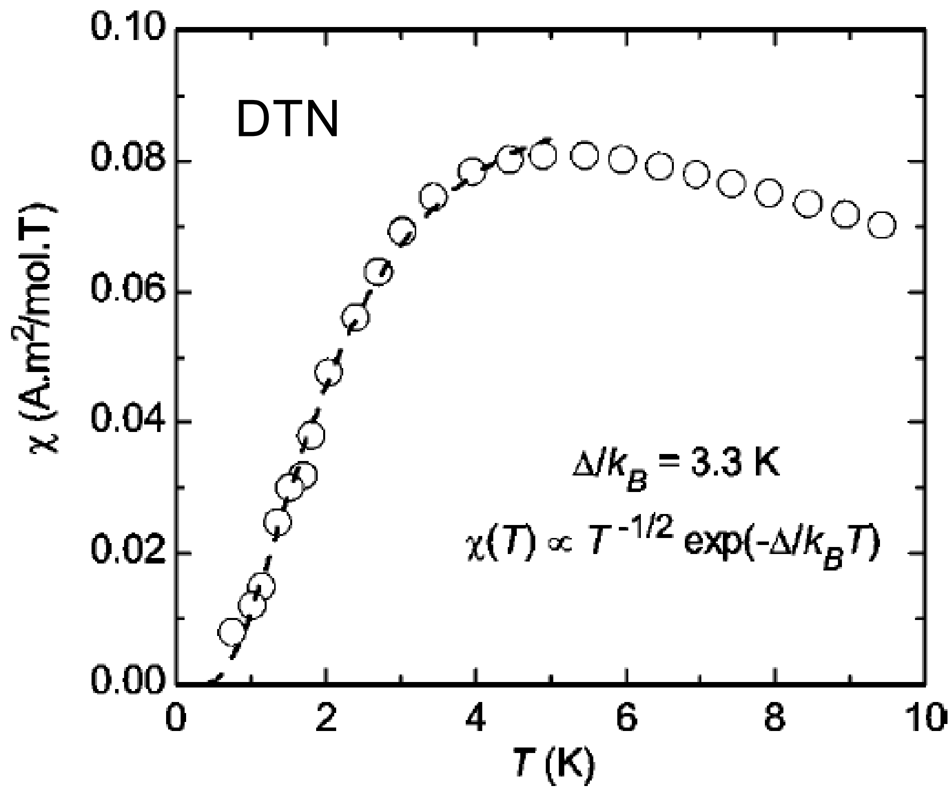
(b)



Strong single-ion anisotropy

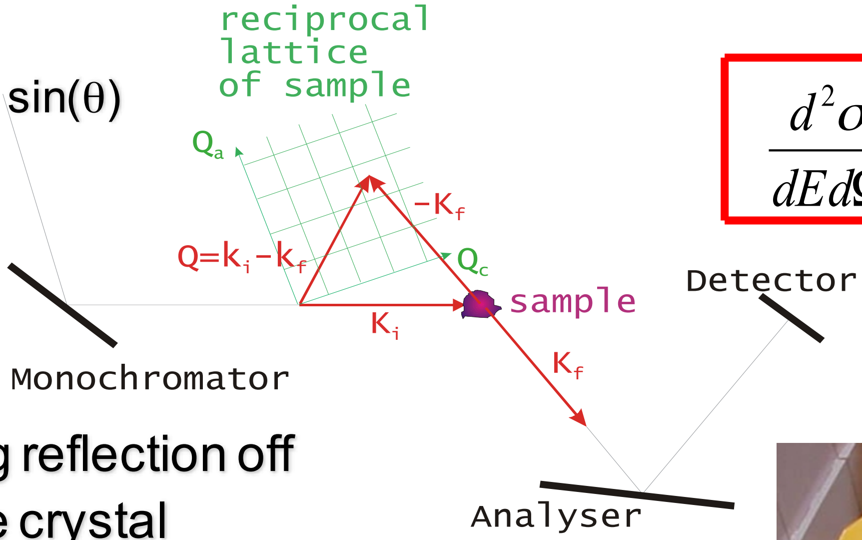


Magnetic susceptibility



A. Paduan-Filho et al, J. Appl. Phys. **95**, 7537 (2004)

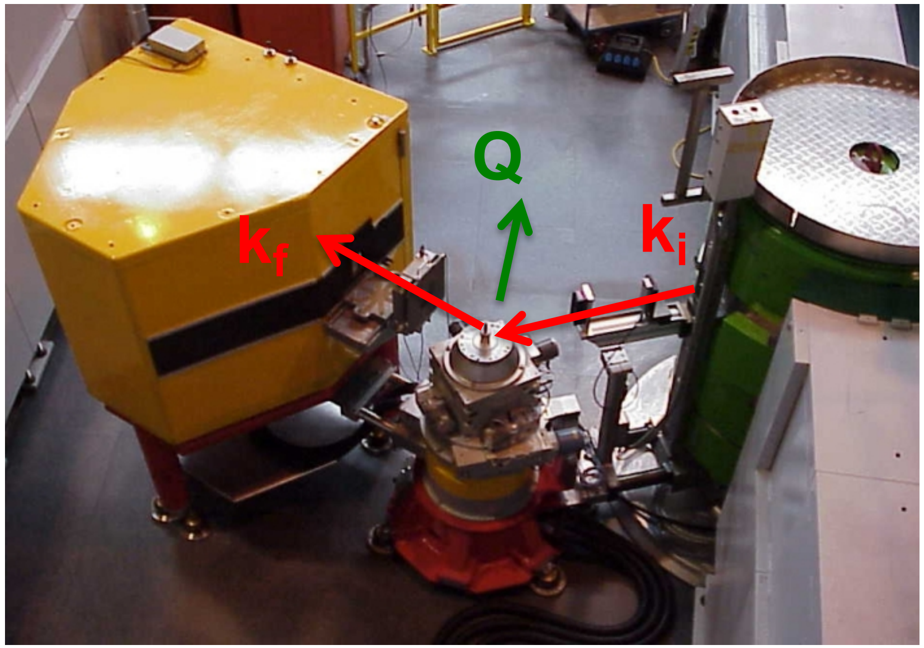
Magnetic neutron scattering



Bragg reflection off single crystal

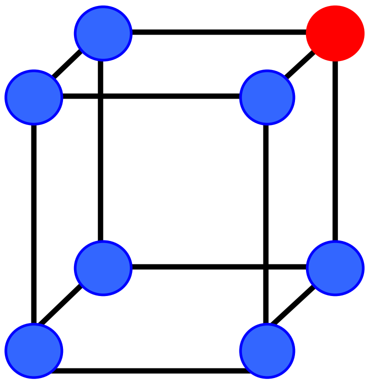
$$\frac{d^2\sigma}{dEd\Omega} \propto \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(Q,\omega)$$

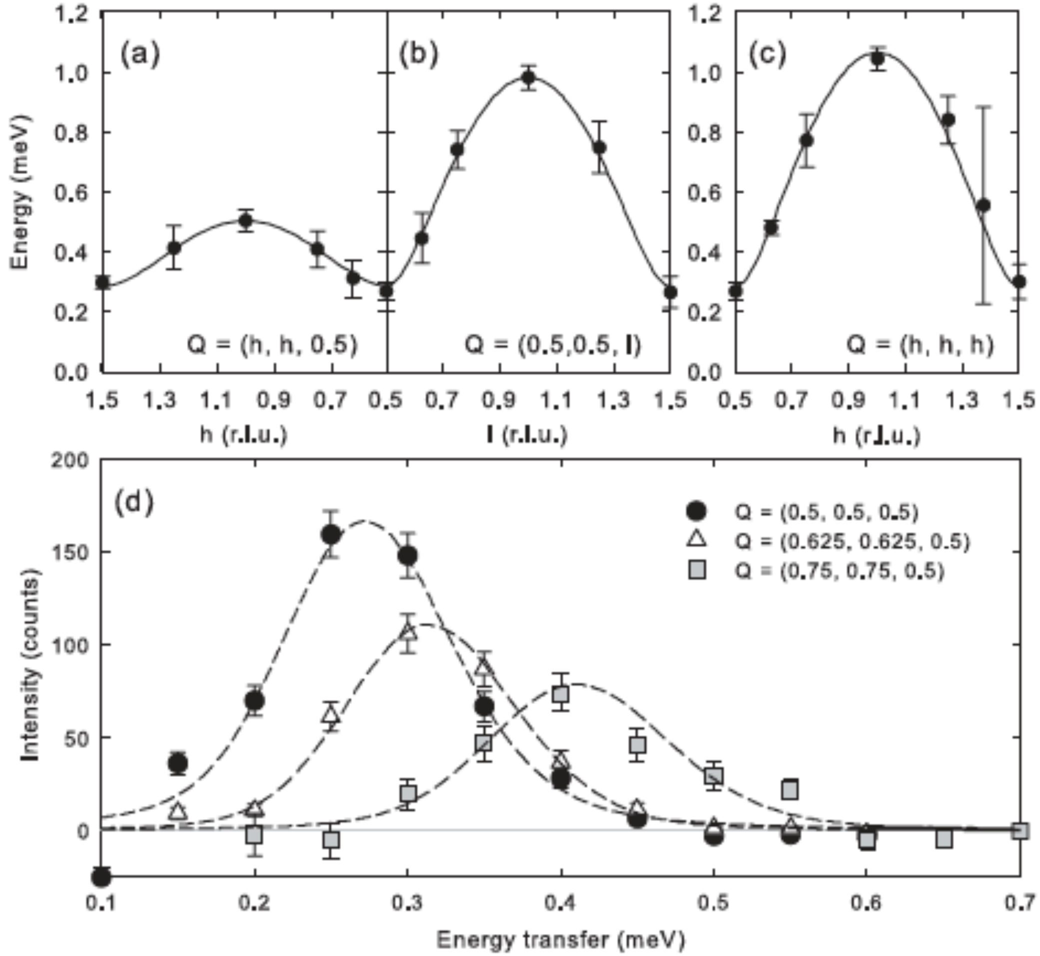
Sensitive to spin correlation functions



RITA spectrometer at PSI

Excitations of a quantum magnet





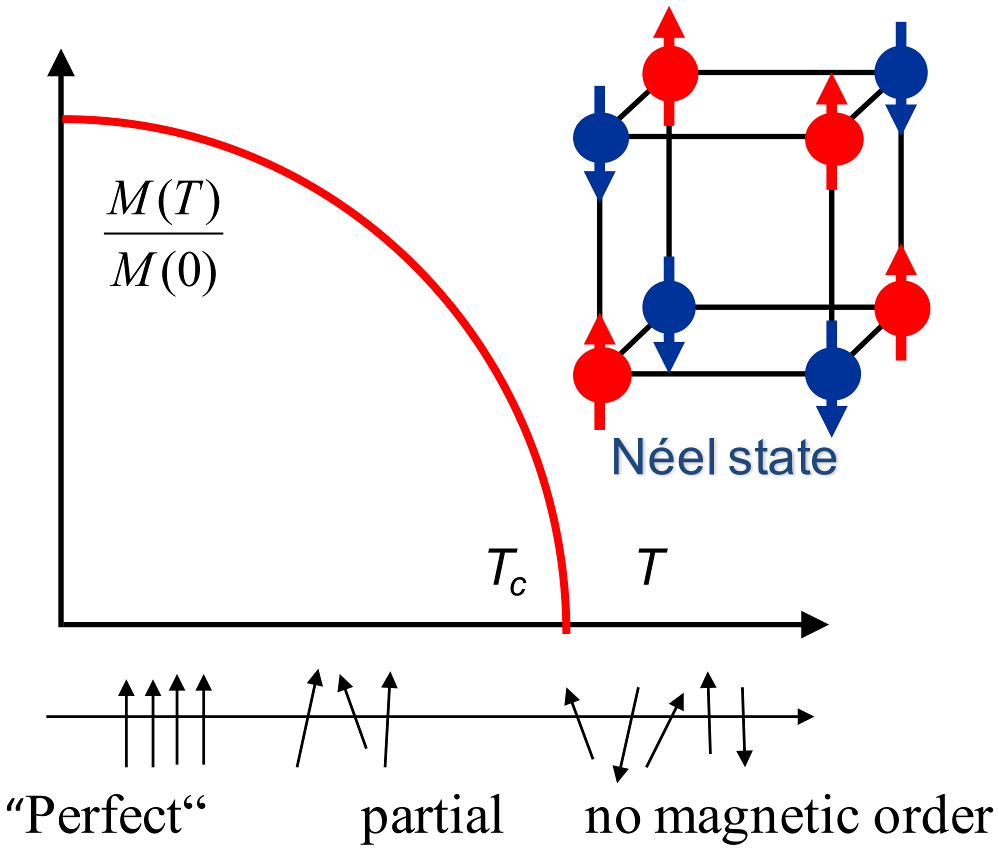
delocalization of |±1> states due to spin interactions

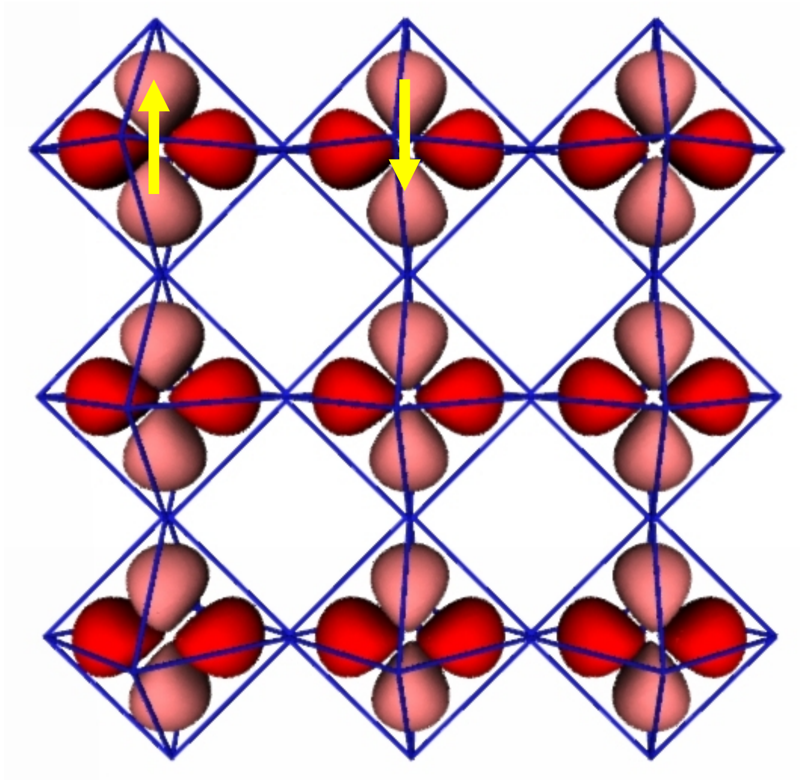
$$\omega_{\mathbf{k}\pm} = \sqrt{D^2 + 2D\sum_{\nu} J_\nu \cos k_\nu}$$

Zapf et al, Phys. Rev. Lett. **96**, 077204 (2006)

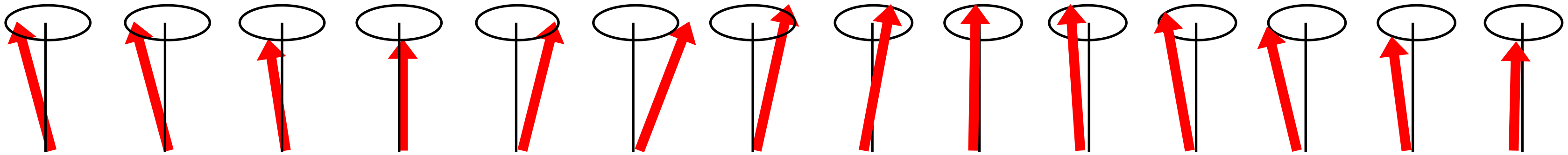
From the quantum nature of magnetism to “classical long-range order”

- T >>J: spin fluctuates between up and down, but neighbours always want to point in opposite direction
- cooperative phase transition for T < J



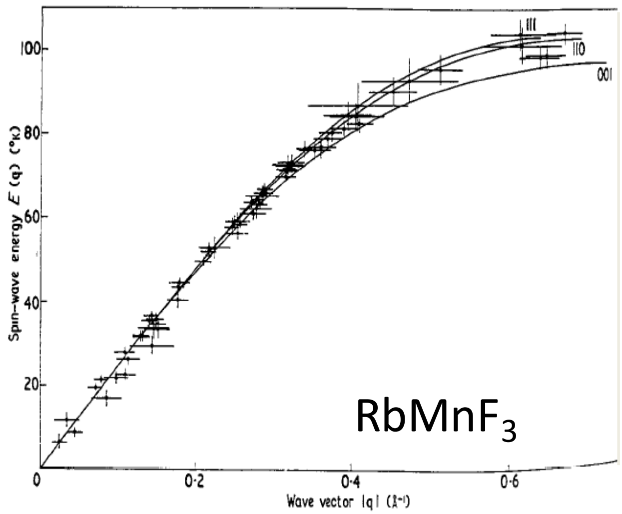


The nature of spin waves



Small deviations of the magnetic moments away from ordered direction

$$\hat{\mathcal{H}} = \sum_{l \neq l'} J_{ll'} \hat{S}_l \hat{S}_{l'}$$



Windsor and Stevenson, Proc Phys Soc London (1966)

$$\hat{S}_l^+ = \sqrt{2S}\,\hat{a}_l \qquad \hat{S}_l^- = \sqrt{2S}\,\hat{a}_l^+ \qquad \hat{S}_l^z = S - \hat{a}_l^+ \hat{a}_l$$

$$a_l = \frac{1}{\sqrt{N}} \sum_{\kappa} \exp(i\boldsymbol{\kappa} \boldsymbol{l})\, a_{\kappa} \qquad \left\{\hat{a}_{\kappa}, \hat{a}_{\kappa'}^+\right\} = \delta_{\kappa\kappa'}$$

$$\hat{H} = \text{const} + \sum_{\kappa} \hbar\omega_{\kappa} a_{\kappa}^+ a_{\kappa}$$

$$\hbar\omega_{\kappa} = 2S\left[J(\boldsymbol{\kappa}) - J(0)\right]$$

$$\hat{H} = \text{const} + \sum_{\kappa} \hbar\omega_{\kappa} b_{\kappa}^+ b_{\kappa}$$

$$\hbar\omega_{\kappa} = 2S\sqrt{\left[J(0)\right]^2 - \left[J(\boldsymbol{\kappa})\right]^2}$$

Cross-section of the inelastic magnetic neutron scattering

Polarization factor

Form factor

DW factor

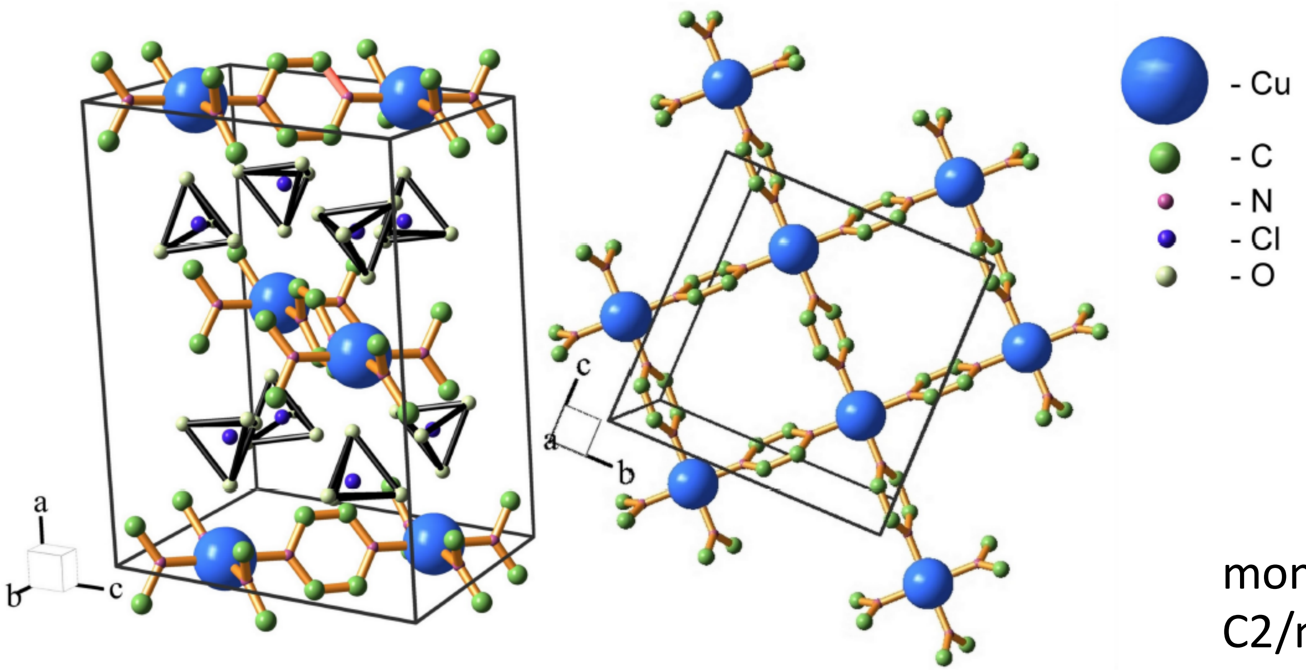
$$\left(\frac{d^2\sigma}{d\Omega\, dE'}\right) = (\gamma r_0)^2 \frac{k'}{k}\frac{(2\pi)^3}{V_0}\frac{1}{2}S\left(1+\hat{q}_z^2\right)\left|\frac{1}{2}gF(\boldsymbol{q})\right|^2 e^{-2W_{\boldsymbol{q}}}\times$$

$$\times\sum_{\boldsymbol{\tau},\boldsymbol{\kappa}}\left[\delta(\boldsymbol{\kappa}-\boldsymbol{q}-\boldsymbol{\tau})\delta(\hbar\omega_{\boldsymbol{\kappa}}-\hbar\omega)\langle\langle n_{\boldsymbol{\kappa}}+1\rangle\rangle+\delta(\boldsymbol{\kappa}+\boldsymbol{q}-\boldsymbol{\tau})\delta(\hbar\omega_{\boldsymbol{\kappa}}+\hbar\omega)\langle\langle n_{\boldsymbol{\kappa}}\rangle\rangle\right]$$

Magnon creation

Magnon annihilation

Simple Example: $Cu(pz)_2(ClO_4)_2$

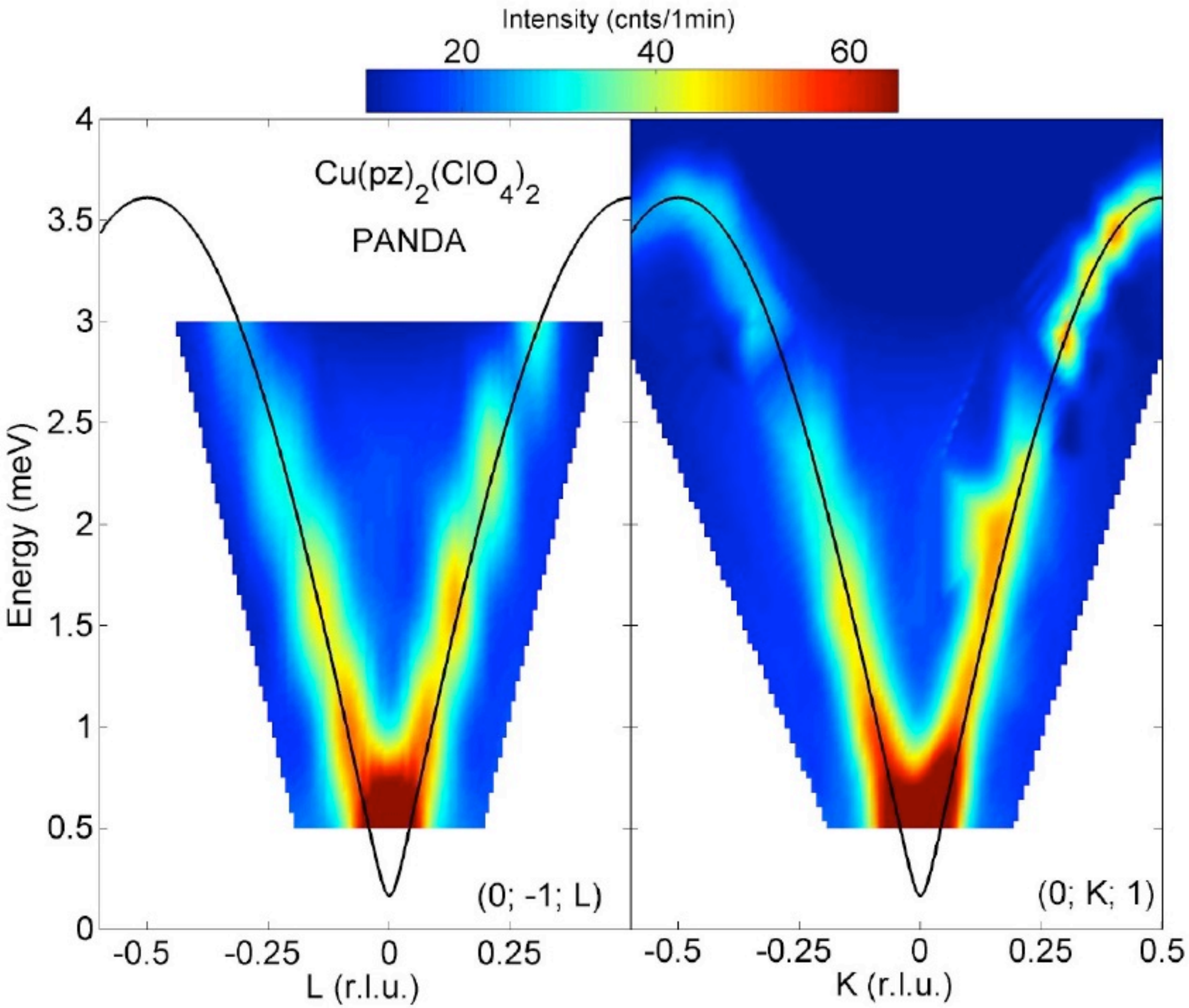


monoclinc
C2/m
β= 96.5

2_b, m_{ac}

Identical exchange interactions along bc and –bc directions
Absence of DM interactions due to symmetry

Spin-wave excitations



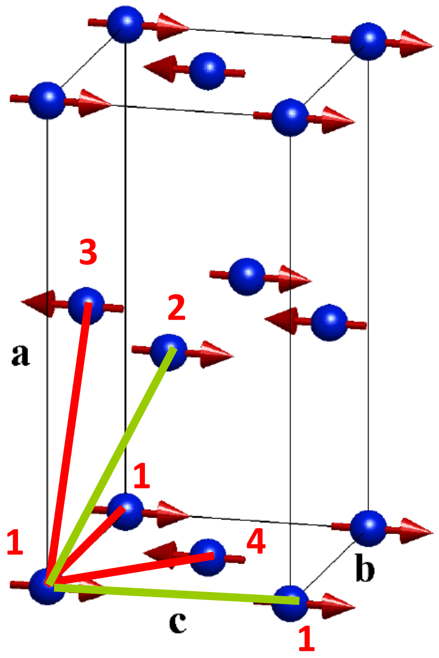
- Spin waves are best seen in reciprocal space where they occur as peaks as a function of energy

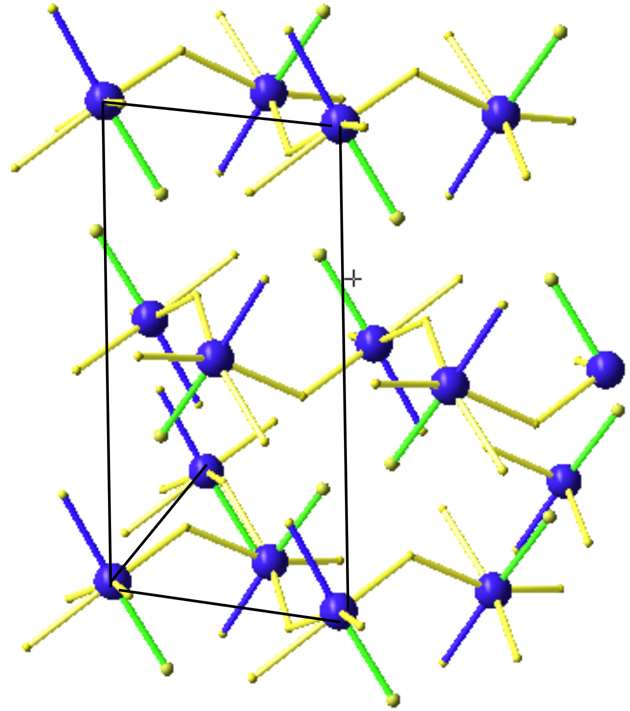
- Measured with inelastic neutron scattering instruments (types?)

N. Tsyrulin, et al, Phys. Rev. B (2009)

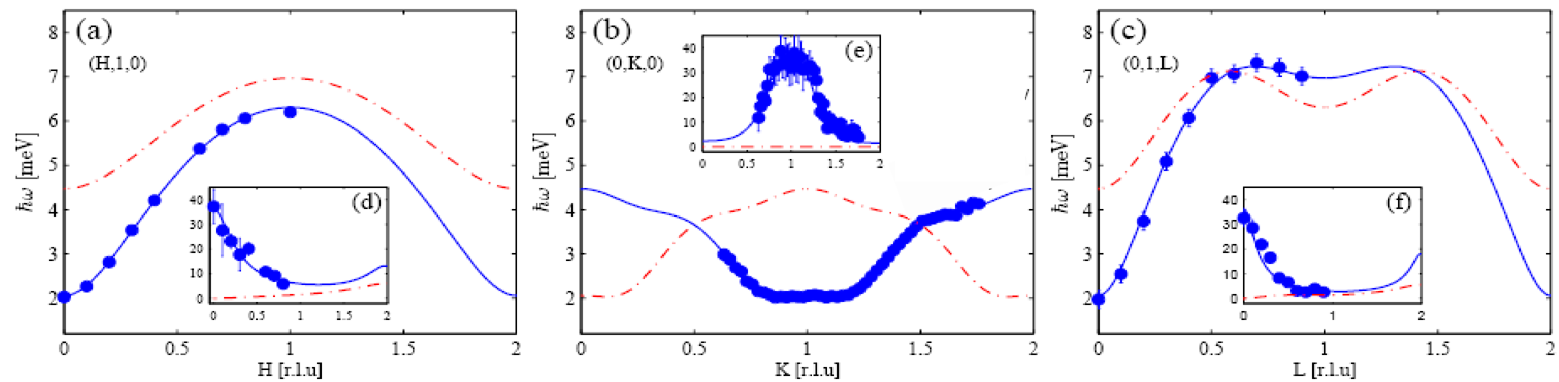
$LiNiPO_4$: exchange interactions

J_{14}=1.04meV
J_{11}=0.67meV
J_{13}=0.3meV
J_{12}=-0.11meV
J_{11}=-0.05meV
D_x=0.17meV
D_y=0.91meV
D_z=0meV

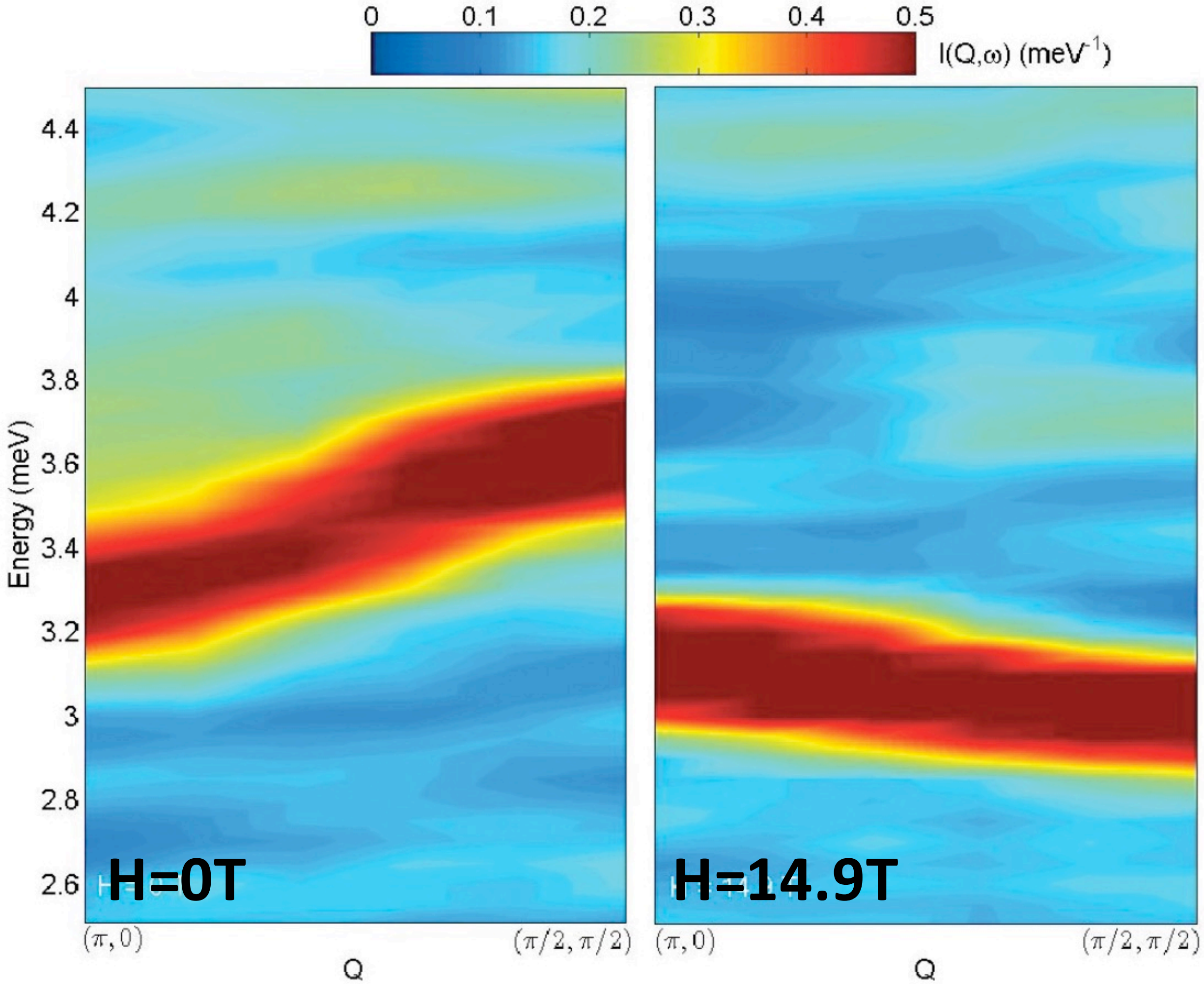




T.B.S. Jensen et al, Phys. Rev. B 79, 092413 (2009).



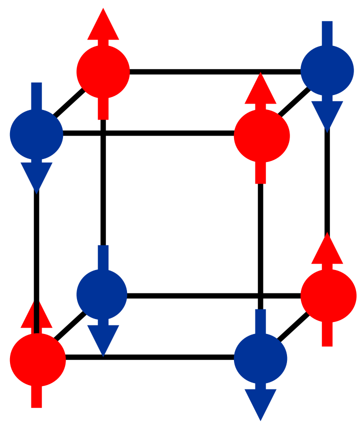
Novel excitations in $Cu(pz)_2(ClO_4)_2$



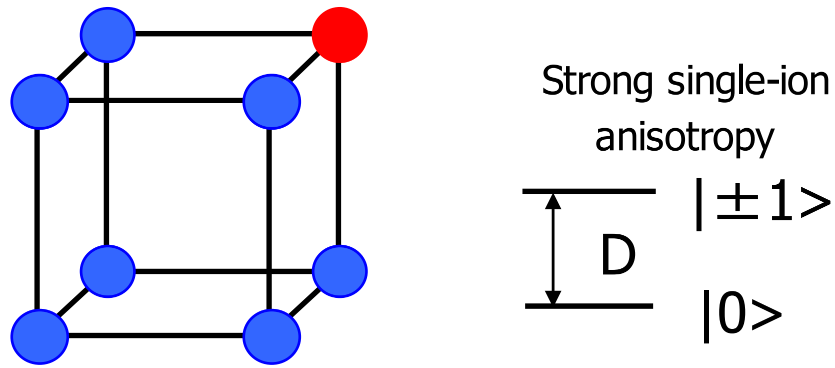
N. Tsyrulin, et al, Phys. Rev. Lett. **102**, 197201 (2009)

Quantum vs. “classical” magnetism

- Magnetism is of quantum nature
- Many materials behave rather classical, and the quantization of spin degrees of freedom is suppressed

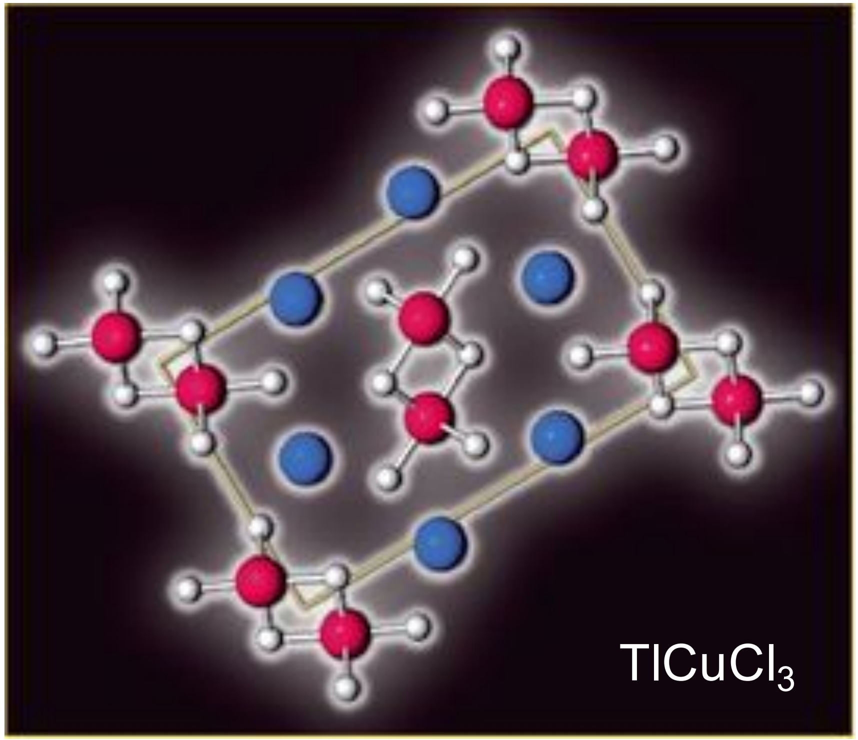


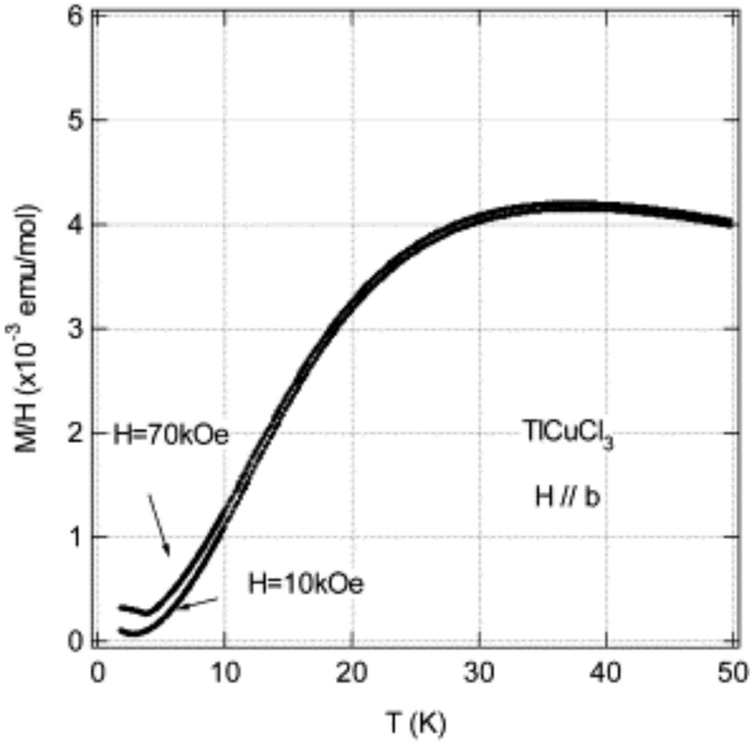
Wave-like perturbation of magnetic moments away from their ordered direction



delocalization of |±1> states due to spin interactions

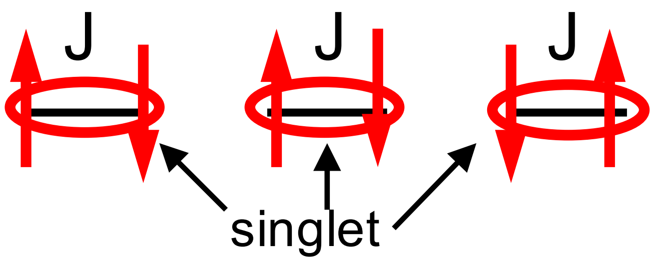
Quantum dimer material



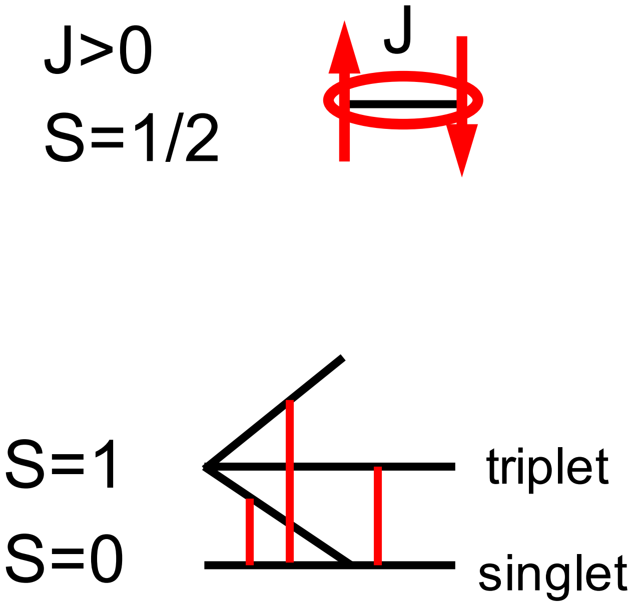


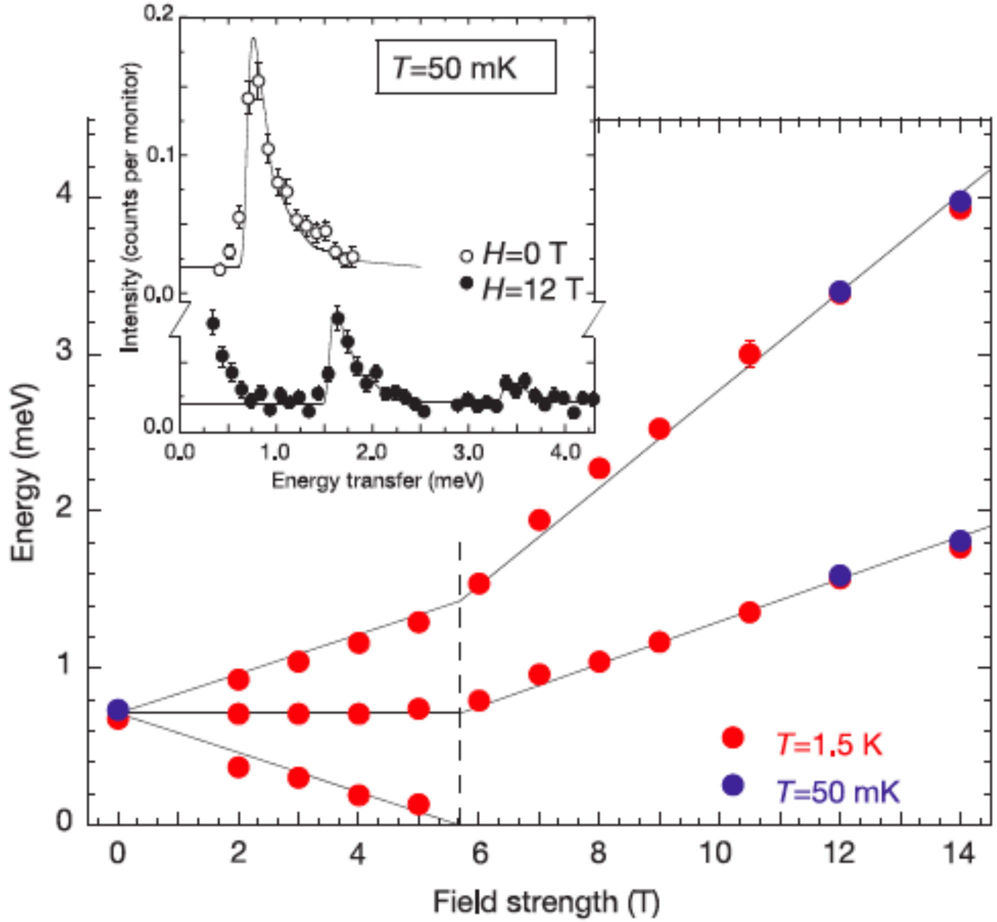
J > 0

S=1/2



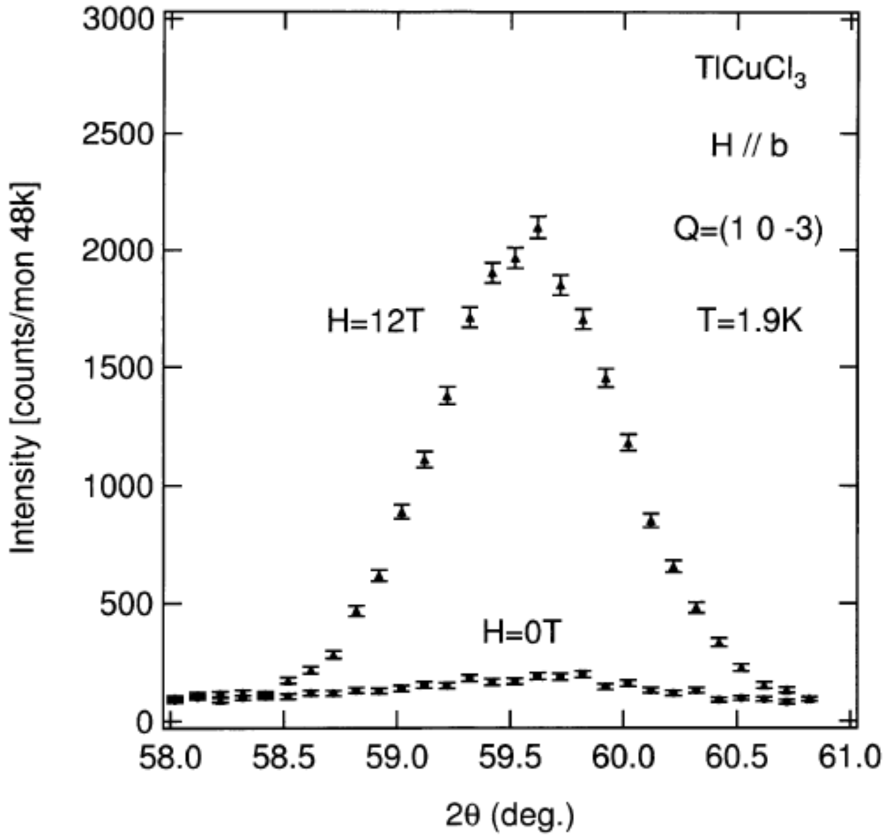
Field dependence of excitations

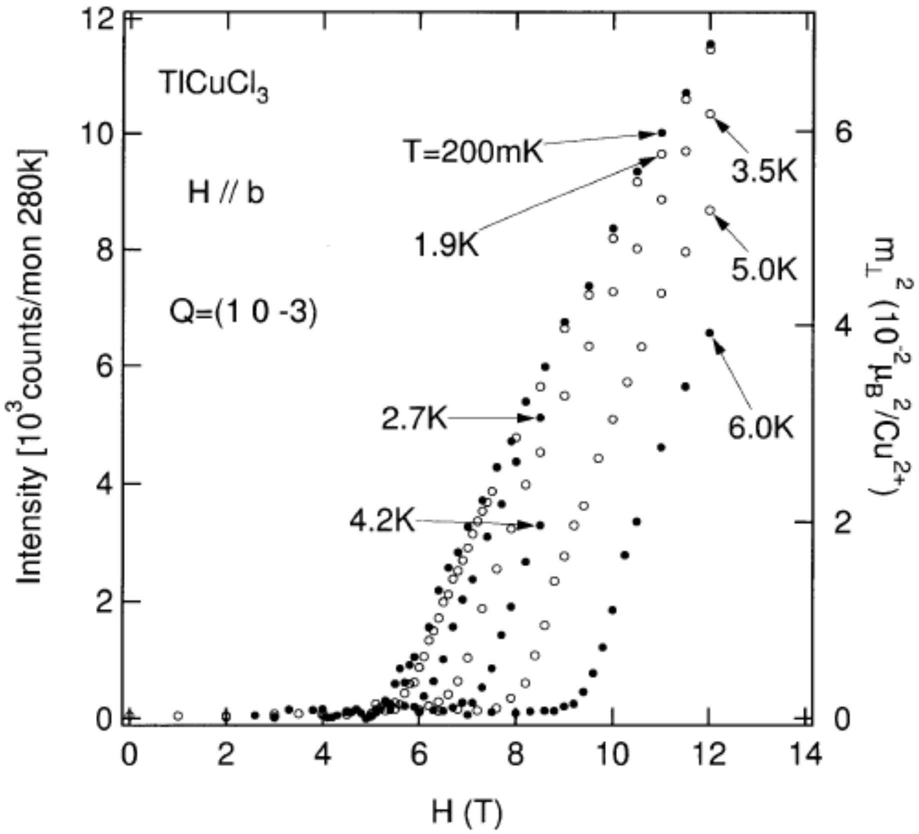




C. Rüegg et al, Nature **423**, 62 (2003).

Quantum phase transition in $TlCuCl_3$





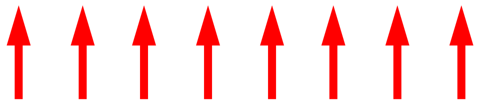
H. Tanak et al, J. Phys. Soc. Japan **4**, 939 (2001).

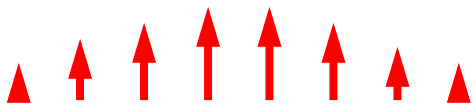
What are the properties of a material at the quantum critical point?

Tuning of the magnetic ground state

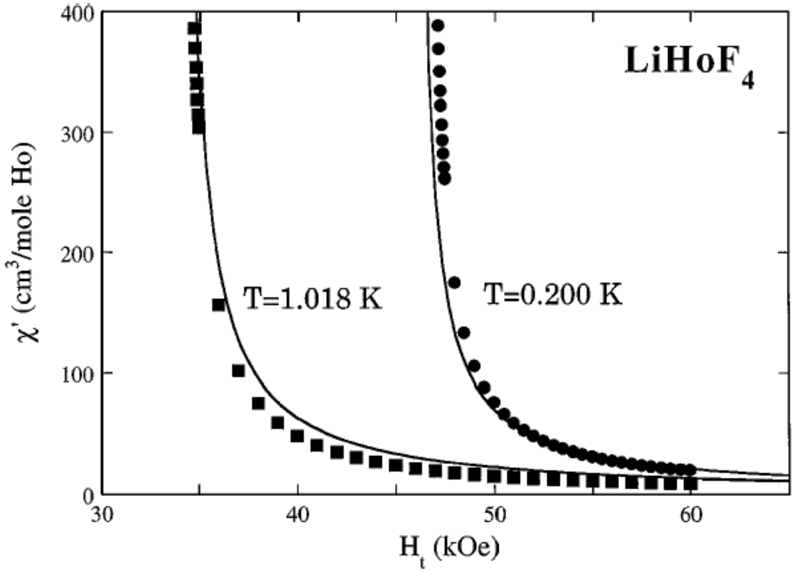
Quantum Phase Transition: $\mathcal{H} = -J\sum S_i^z S_j^z - \mu_B H^x \sum S_i^x$

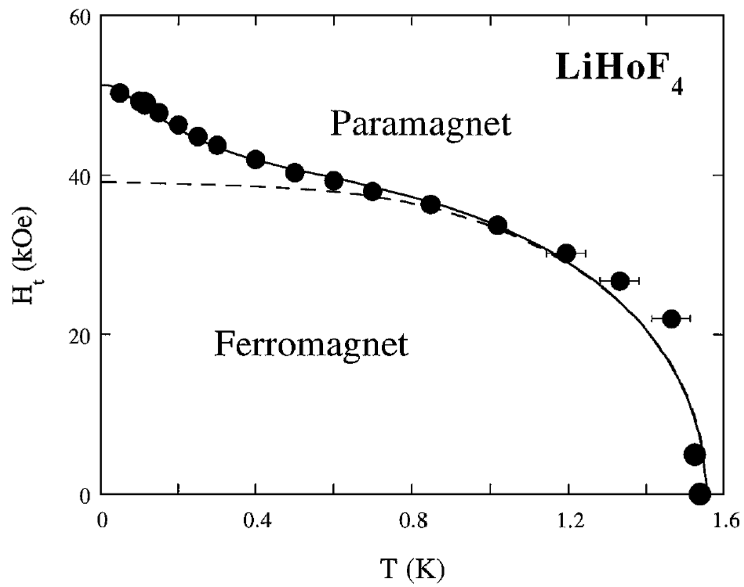
$J \gg \mu_B H^x$ — H → **QCP** — $J \ll \mu_B H^x$ — H →





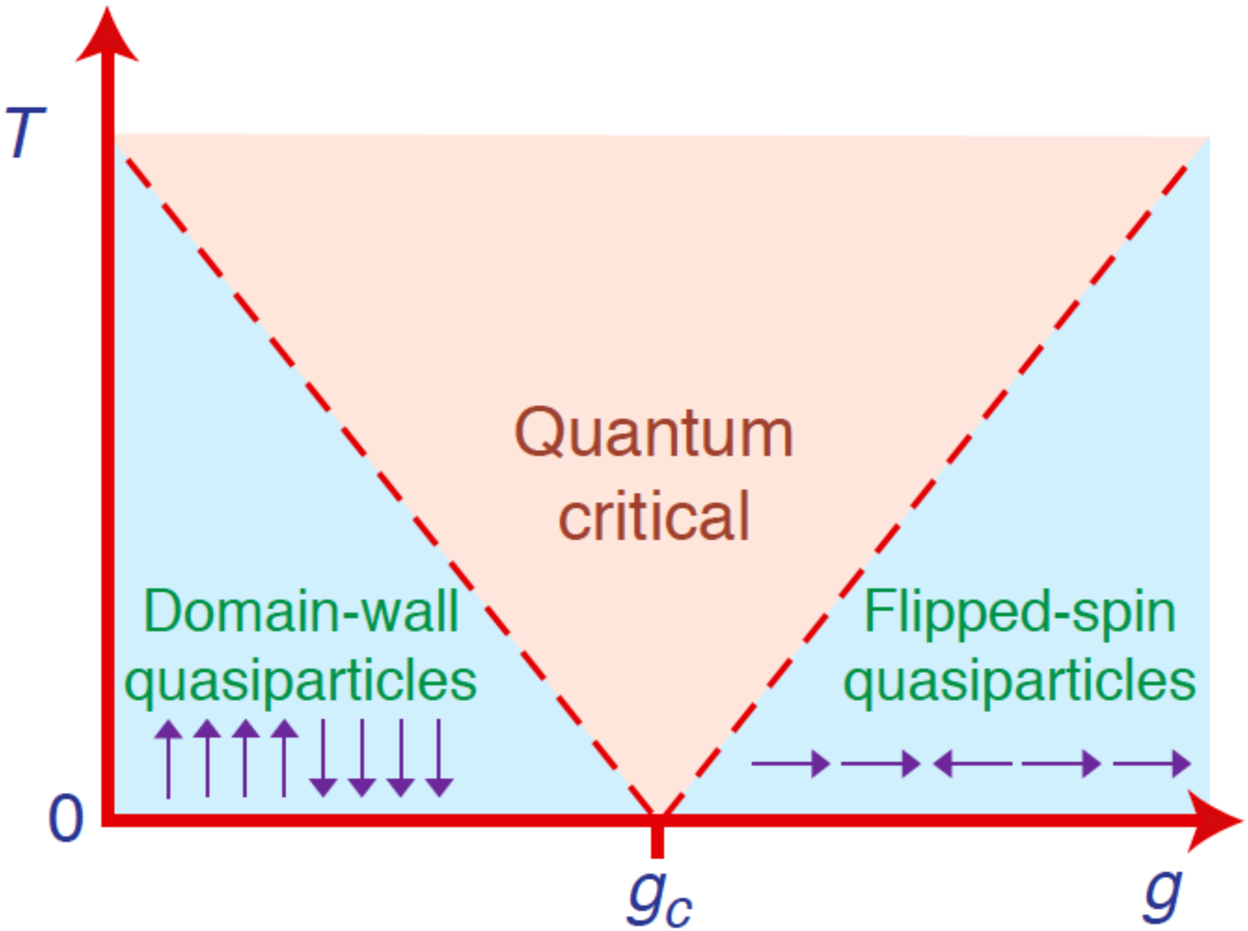
long-range S^z order — power-law decay — short-range S^z order



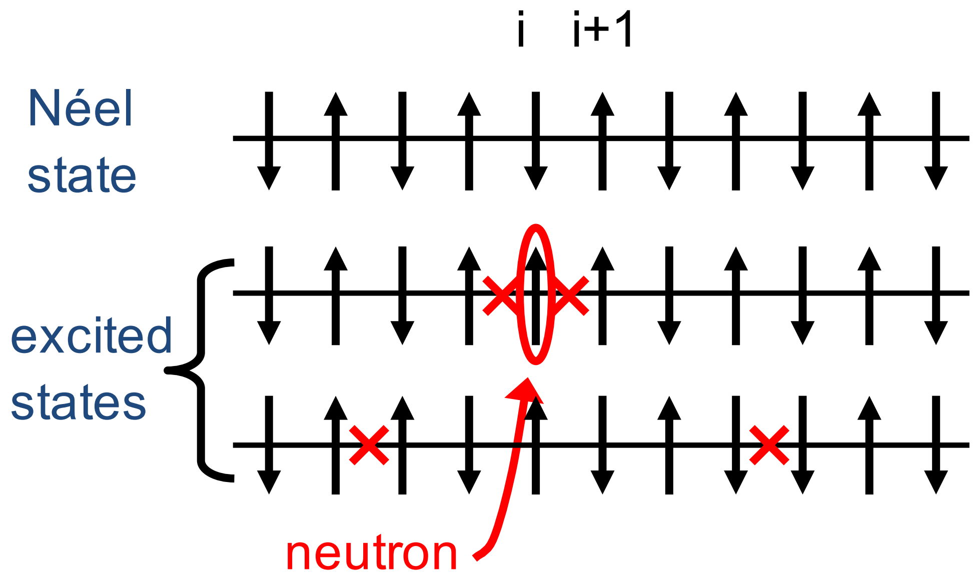


Bitko et al, Phys. Rev. Lett. 77, 940 (1996)

No energy scale at the quantum critical point apart from T



Quantum critical AF S=1/2 Heisenberg Chains

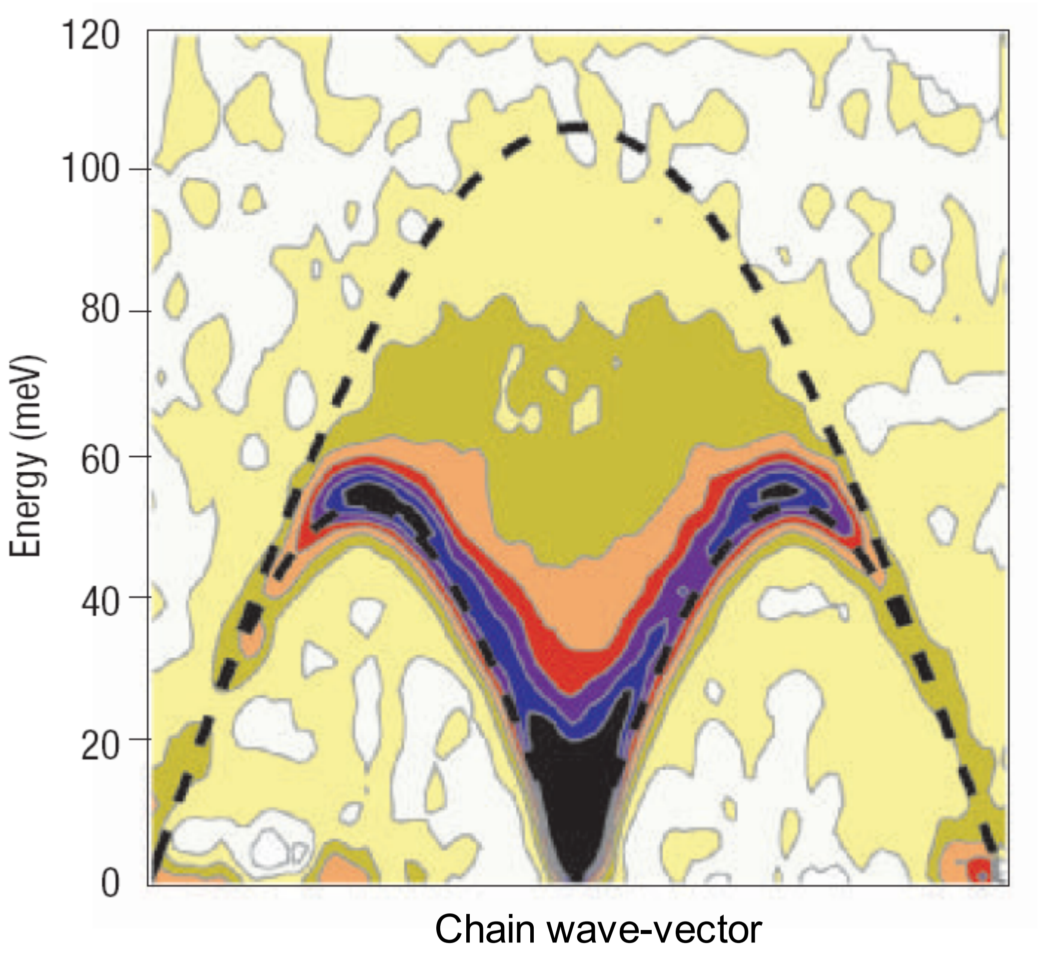


induces spin flips as an excitation

$$\mathcal{H} = J \sum S^z_i S^z_{i+1} + {}^1/_2 \left(S^+_i S^-_{i+1} + S^-_i S^+_{i+1}\right)$$

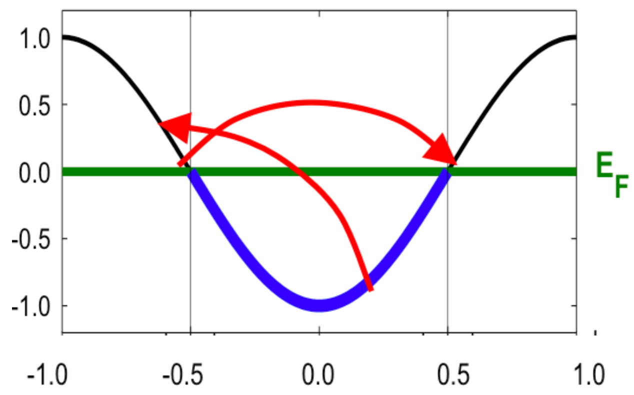
- quasi long-range order: AF correlations fall of as a power law
- new type of excitations: spinons carrying S=1/2
- pair of excitations induced by neutron scattering

AF S=1/2 Heisenberg Chains



B. Lake et al, Nature Materials

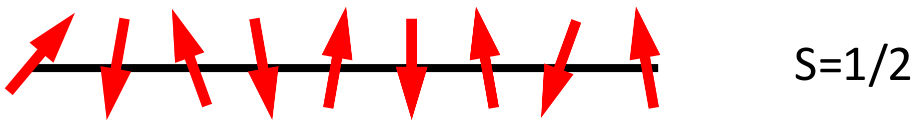
- pair of spinons are created by neutron scattering
- continuum of excitations
- can be pictures as a particle excitition

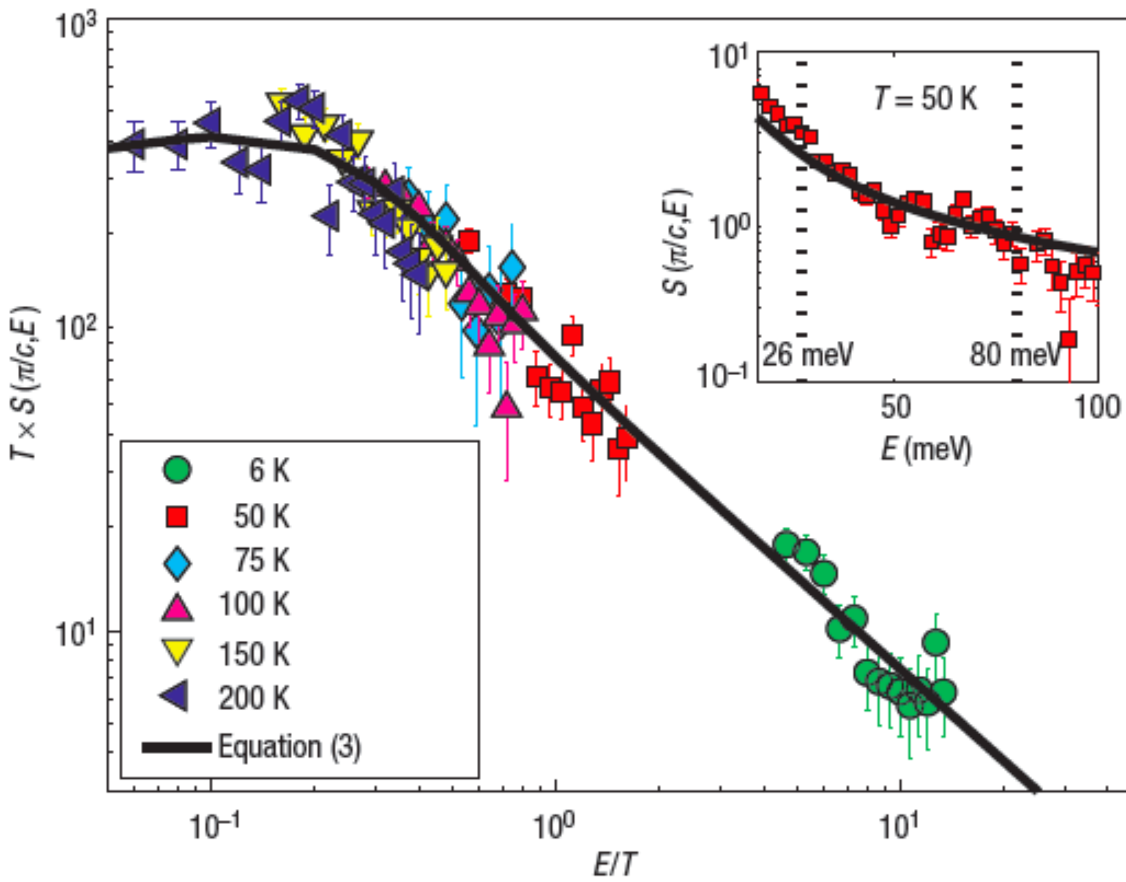


$KCuF_3$ at $T=50K>T_N$

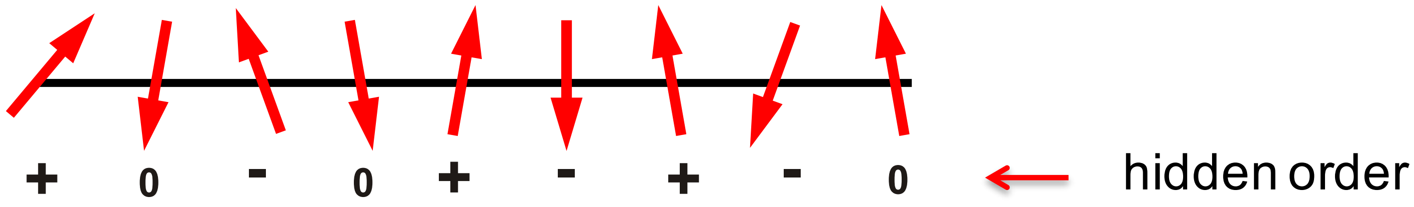
$$\mathcal{H} = J \sum S^z_i S^z_{i+1} + {}^1/_2 \left(S^+_i S^-_{i+1} + S^-_i S^+_{i+1}\right)$$

Quantum critical scaling of AF S=1/2 chain

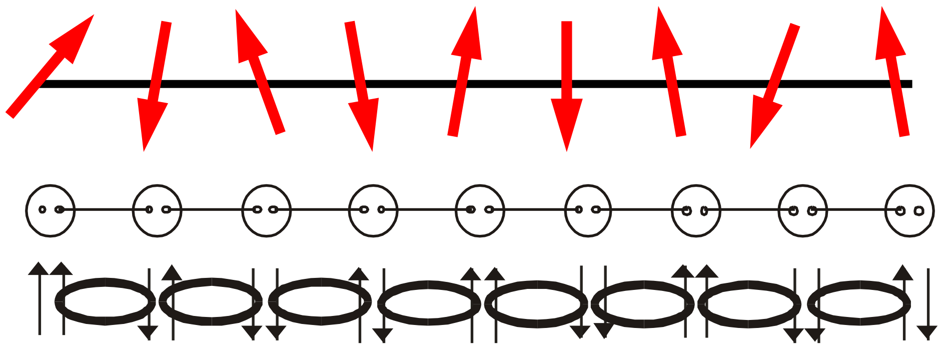




Quantum Spin liquid: antiferromagnetic S=1 chain



coupled S=1 model with string order

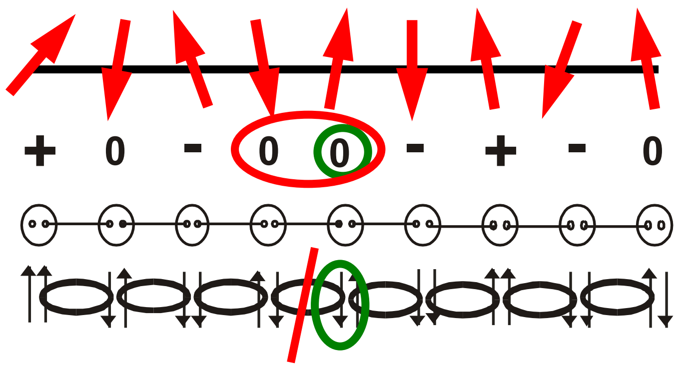


valence-bond solid model
symmetrized pair of S=1/2

F.D.M. Haldane, Phys. Lett. A **93**, 464 (1983).

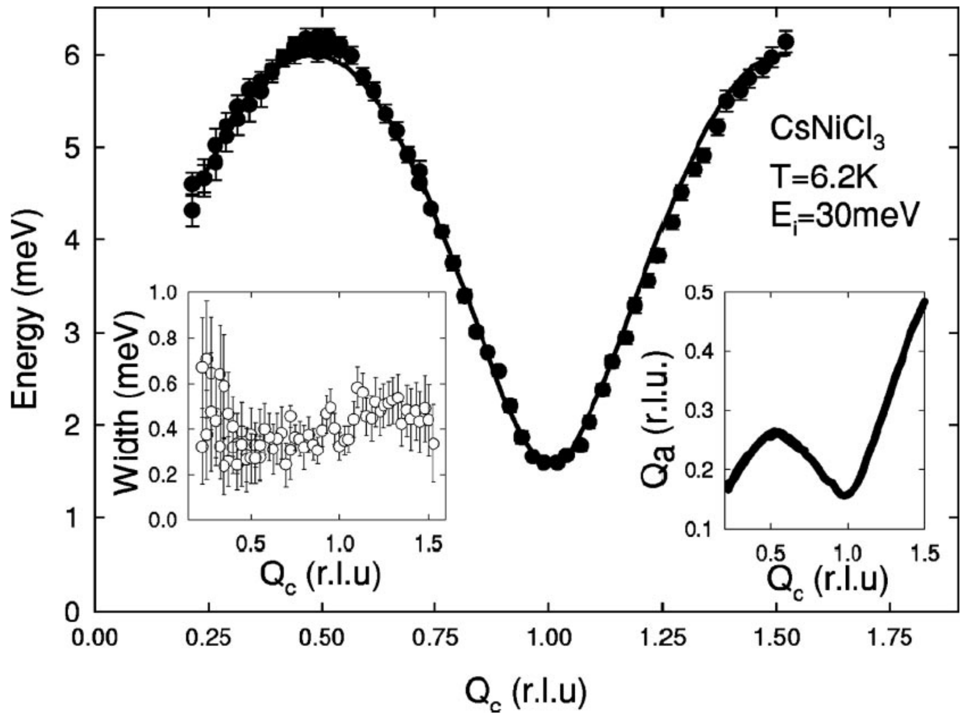
Quantum spin liquid state in AF S=1 chains

The gapped excitation spectrum is dominated by S=1 triplet excitations with hidden spin order



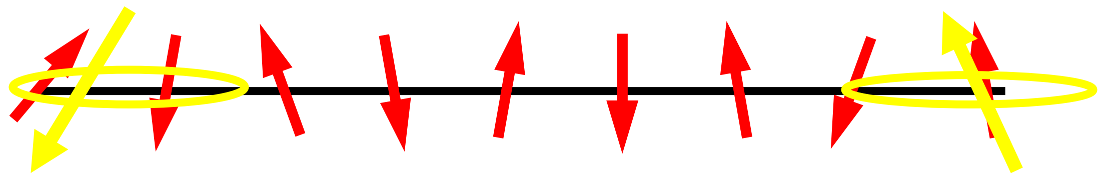
excitations are moving hidden domain walls

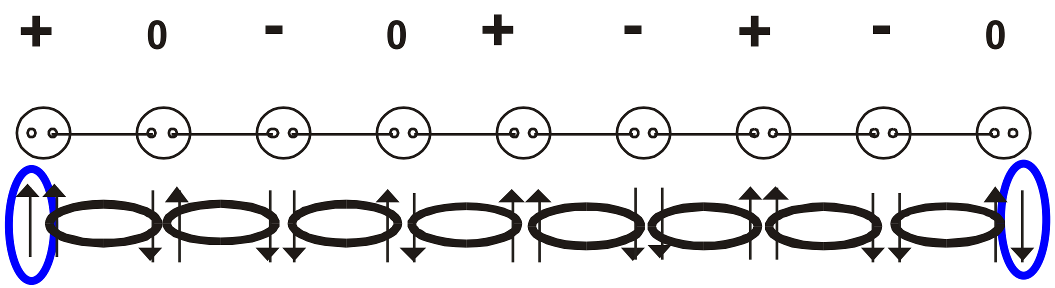
Fath & Solyom, J. Phys. Condens. Matter **5**, 8983 (1993)



M. Kenzelmann et al, Phys. Rev B **66**, 024407 (2002)

Role of defects in spin liquids?





valence-bond model predicts chain-end S=1/2

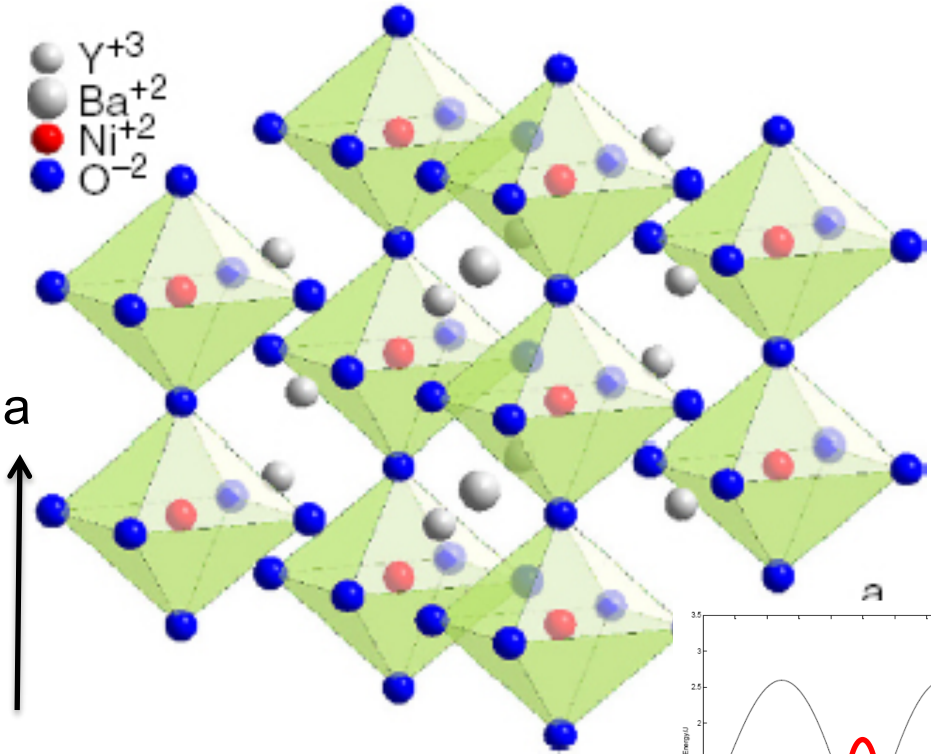
S=1/2 degree of freedoms are coupled (either ferromagnetically or antiferromagnetically)

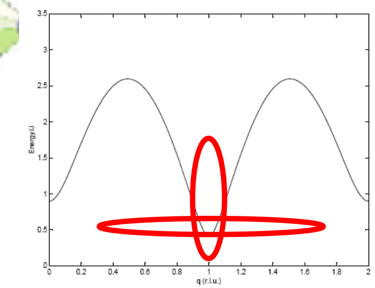
I. Affleck et al. *Phys. Rev. Lett.* **1987**

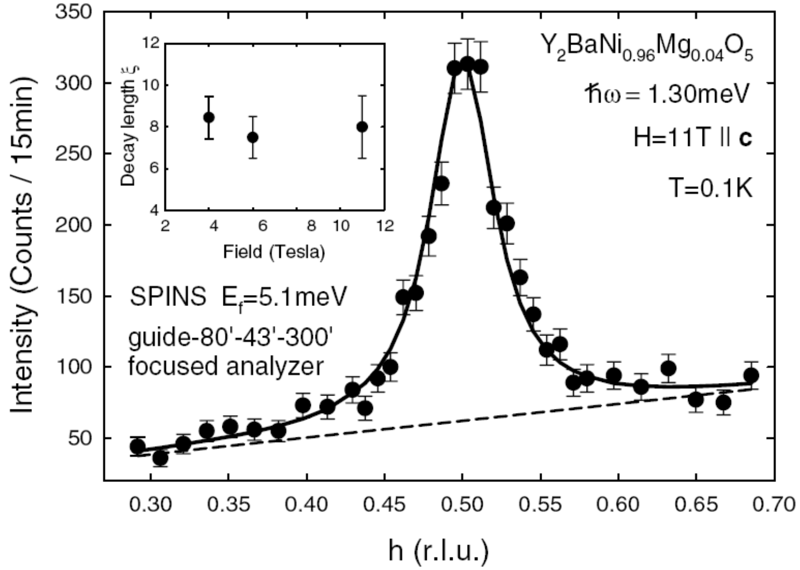
M. Hagiwara et al. *Phys. Rev. Lett.* **1990**

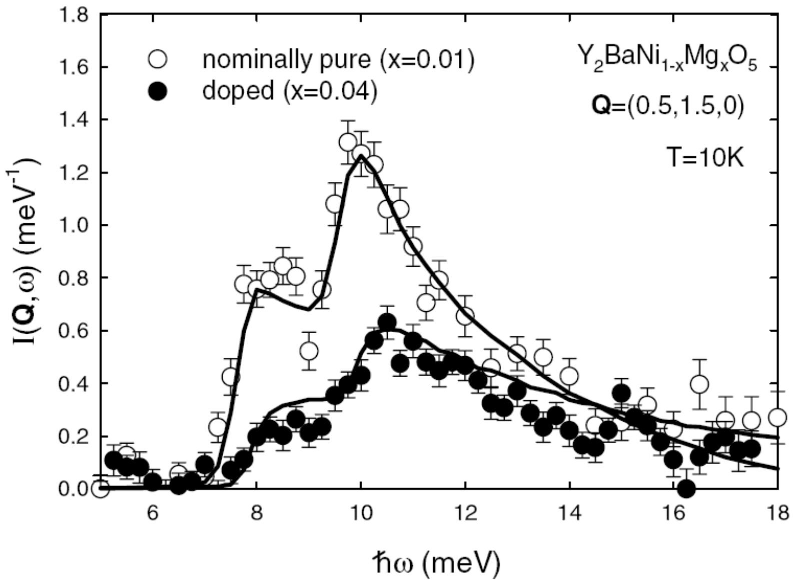
Sensitivity of a quantum spin liquid to impurities

Replace a small amount of Ni ions with non-magnetic Mg







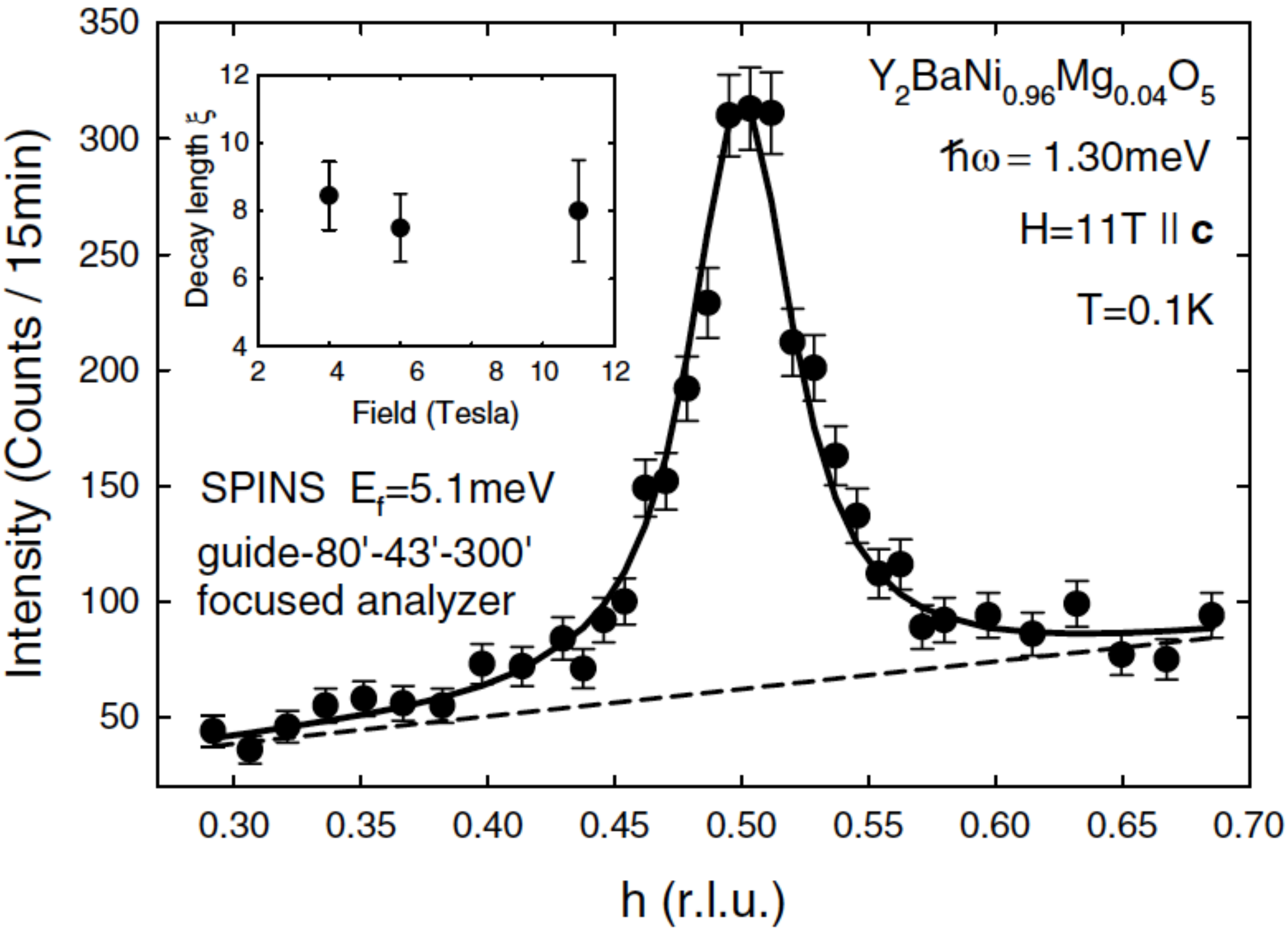


Upward renormalization of quantum gap with decreasing chain length

$\Delta \sim 1/\xi$

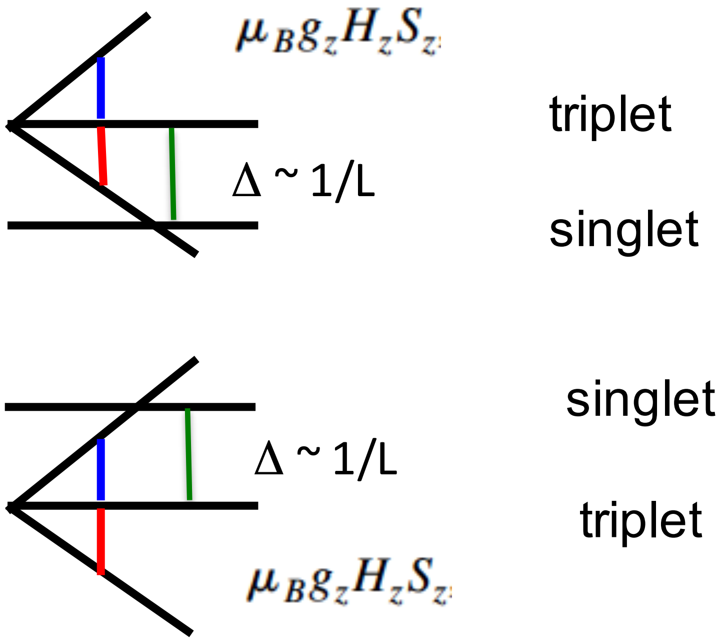
M. Kenzelmann et al. Phys. Rev. Lett. **90**, 087202 (2003)

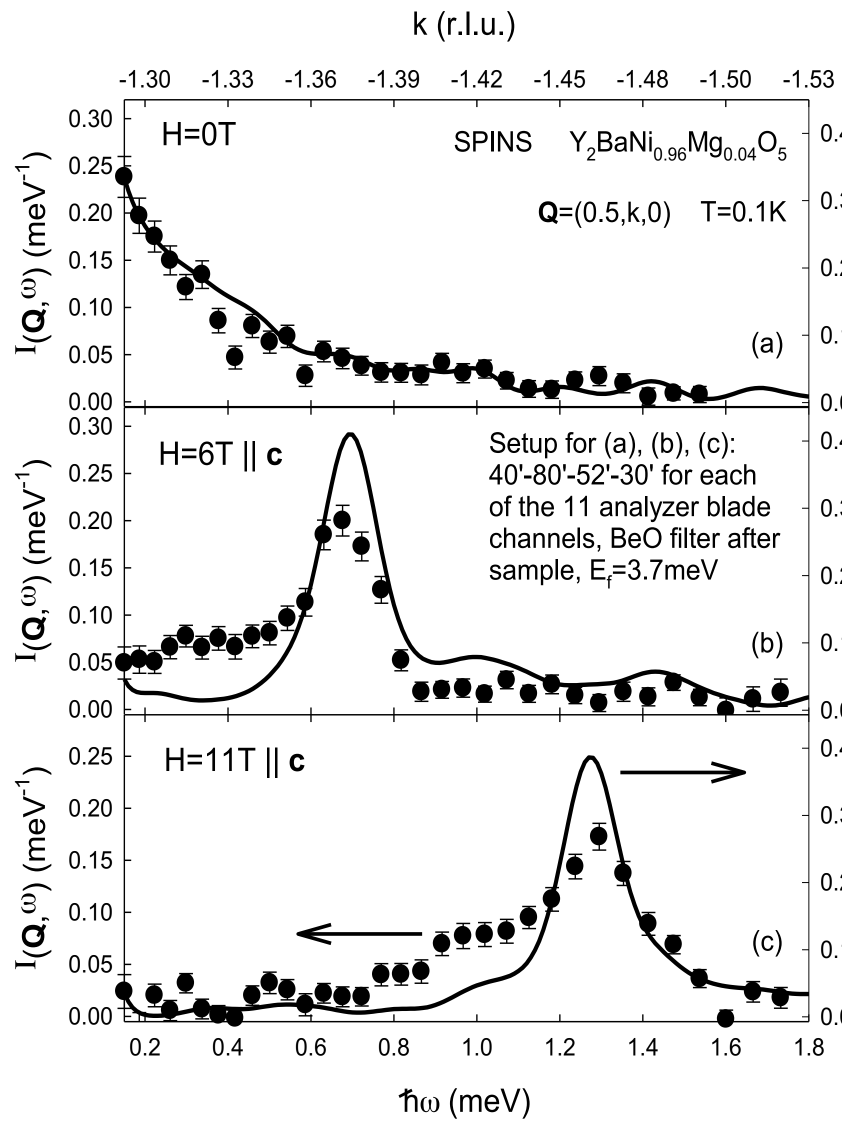
Cooperative chain-end degree of freedoms



M. Kenzelmann et al. Phys. Rev. Lett. **90**, 087202 (2003)

End-chain cooperative degrees of freedom





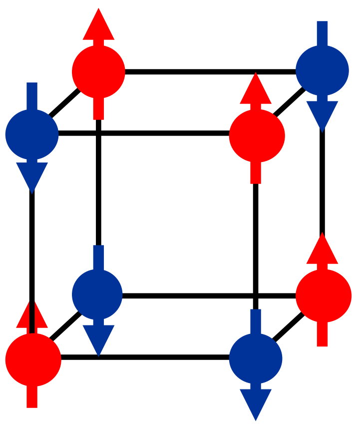
Presence of S=1/2 degrees of freedom at the end of S=1 chains in doped Y_2BaNiO_5

Quantum liquids are sensitive to impurities

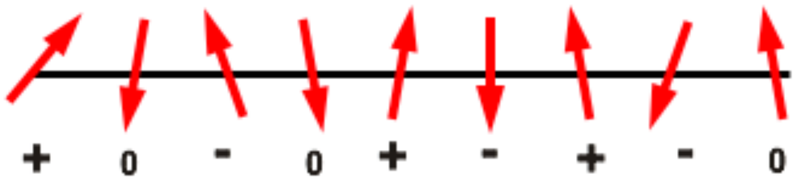
M. Kenzelmann et al. Phys. Rev. Lett. **90**, 087202 (2003)

Quantum vs. “classical” magnetism

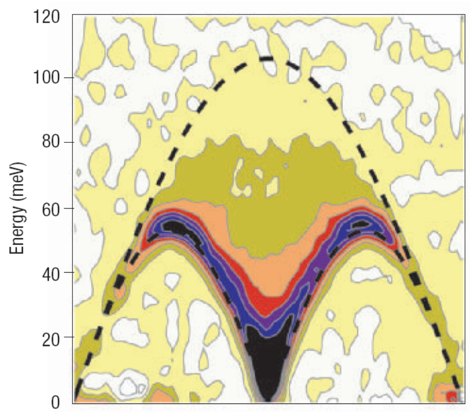
- Magnetism is of quantum nature
- Many materials behave rather classical, and the quantization of spin degrees of freedom is suppressed



Wave-like perturbation of magnetic moments away from their ordered direction



Hidden order in S=1 chains



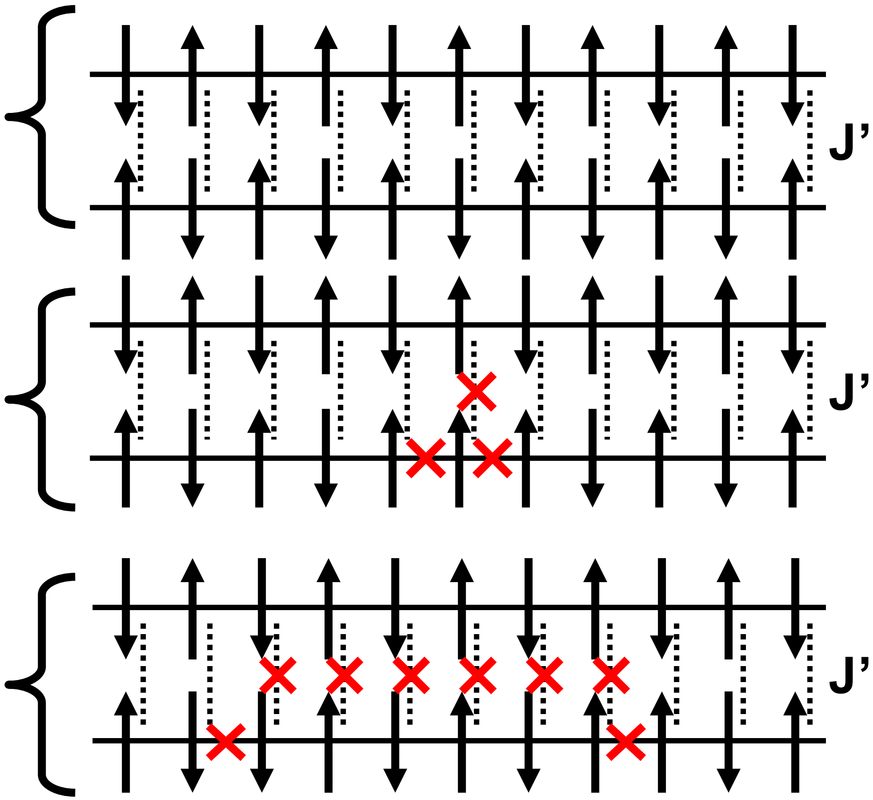
Spinons in S=1/2 chains

Spinon confinement in coupled S=1/2 Chains

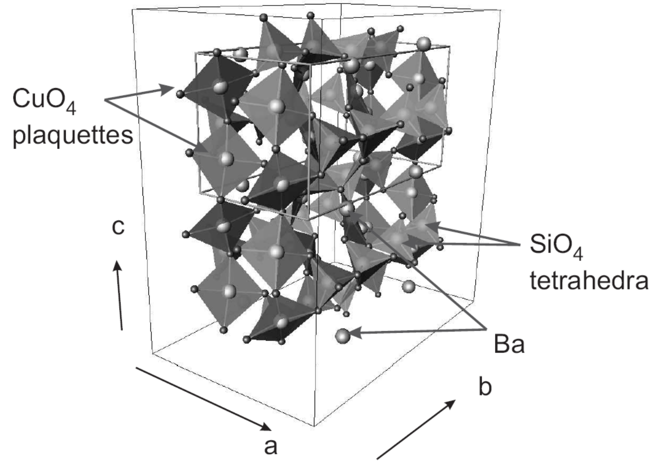
ordered Néel state

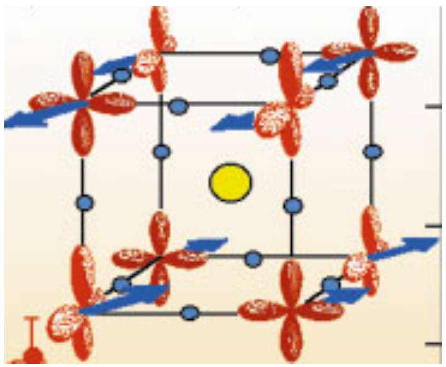
$\Delta S=1$ spin flip

Confined spinons



AF S=1/2 chain $BaCu_2Si_2O_7$

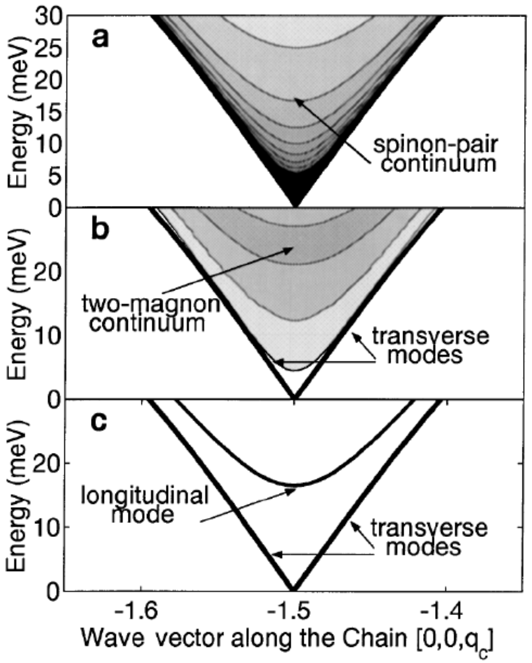


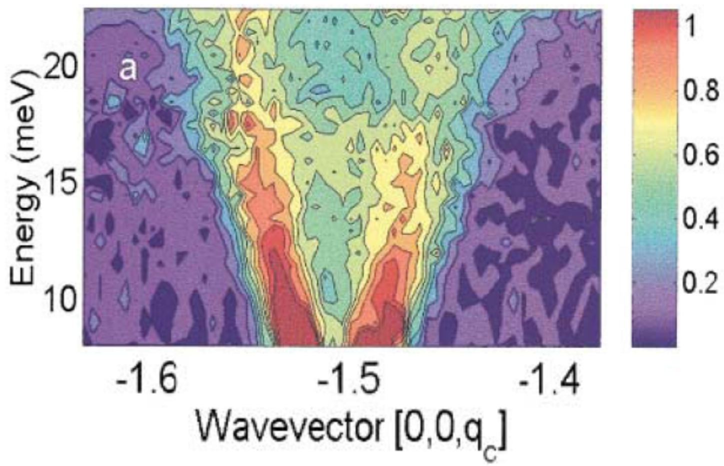


AF S=1/2 chain $KCuF_3$

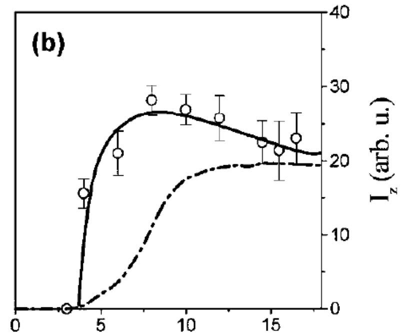
- Long-range magnetic order
- Low-energy spectrum consists of spin waves carrying spin-1
- at higher energies two-spinon continuum

Binding of spinons and longitudinal spin-wave

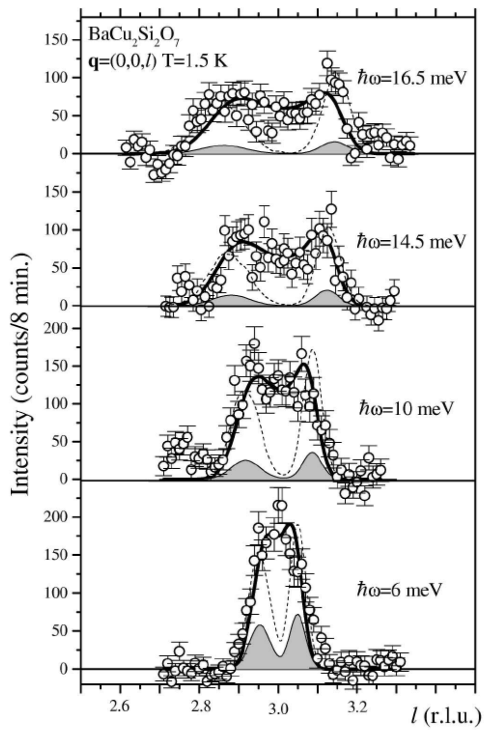




B. Lake et al, Phys. Rev. Lett. 85, 834 (2000)

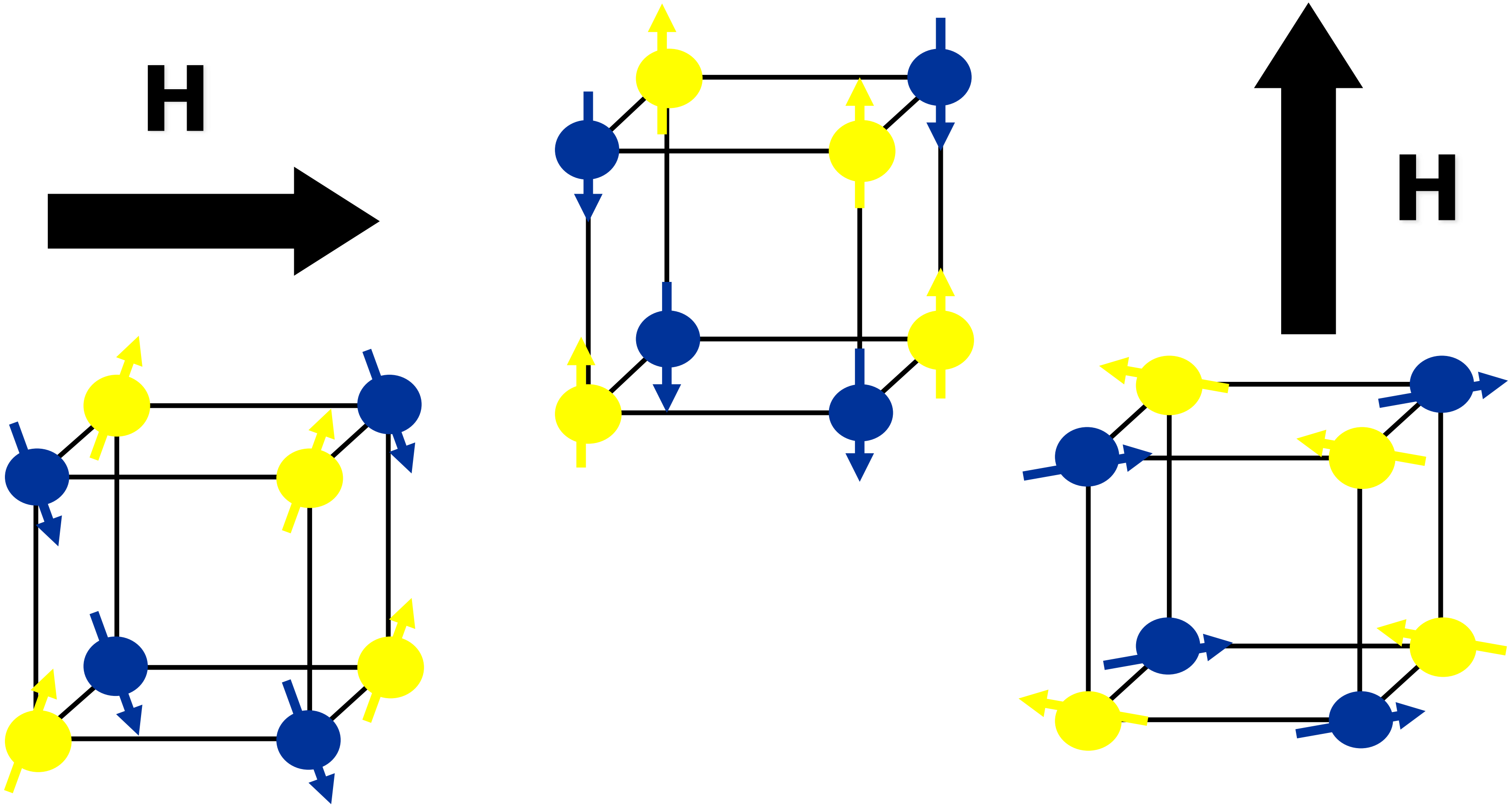


A. Zheludev et al, Phys. Rev. Lett. 85, 4801 (2000)



- Longitudinal fluctuations of ordered moment possible
- Low-energy spectrum consists of spin waves carrying spin-1
- at higher energies two-spinon continuum

Ordered Magnet in Magnetic Field



Canting of spins results in small net magnetization

Spin undergo reorientation (spin-flop transition)

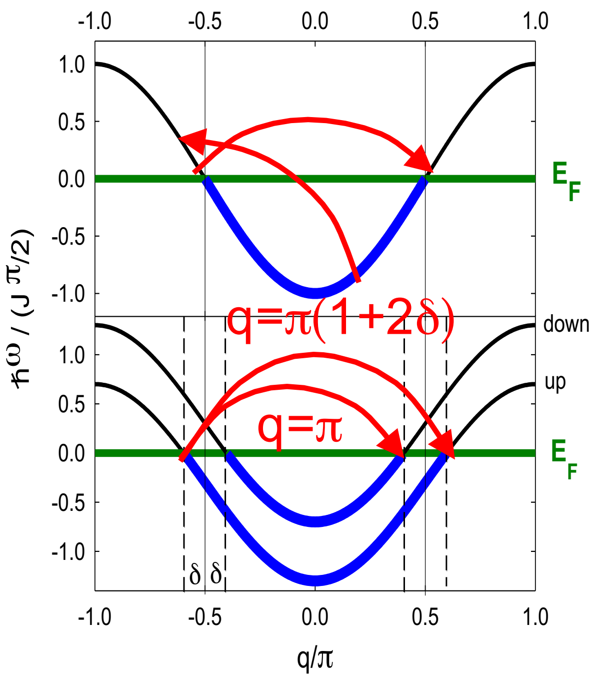
S=1/2 Chain in a Magnetic Field

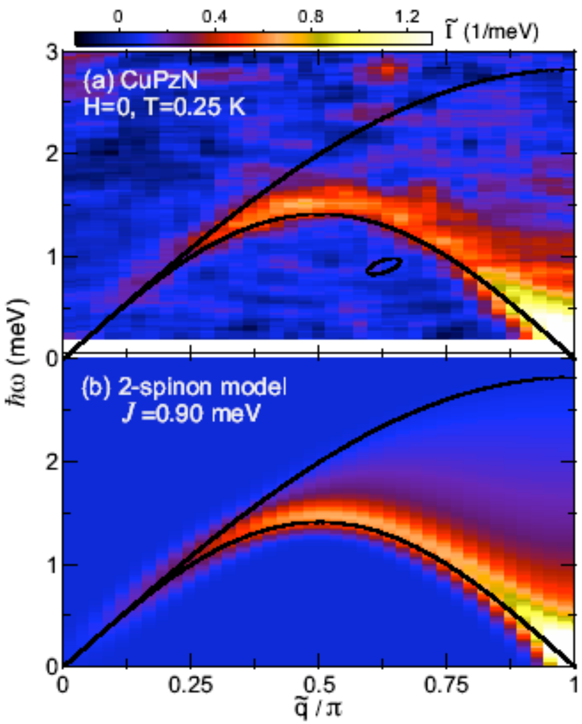
$$\mathcal{H} = \sum_i J\, \mathbf{S}_i\, \mathbf{S}_{i+1} - g_u\, \mu_B\, H\, S^z_i$$

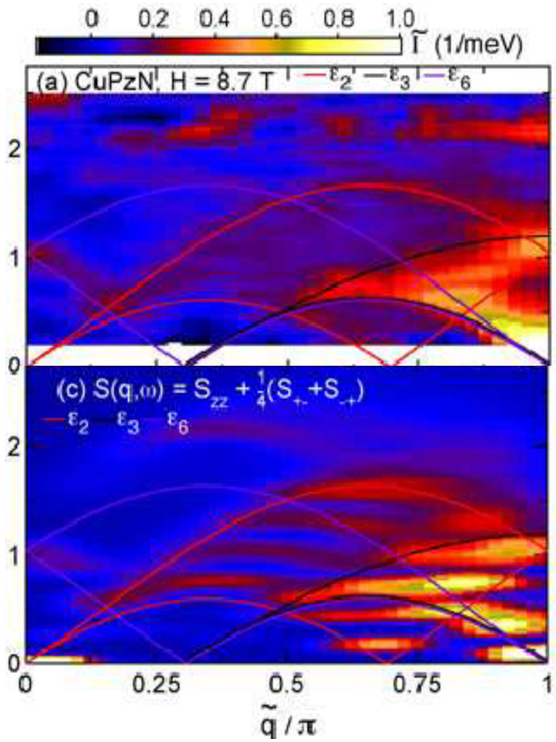
spinon excitations

incommensurate with lattice

Particle-hole excitations



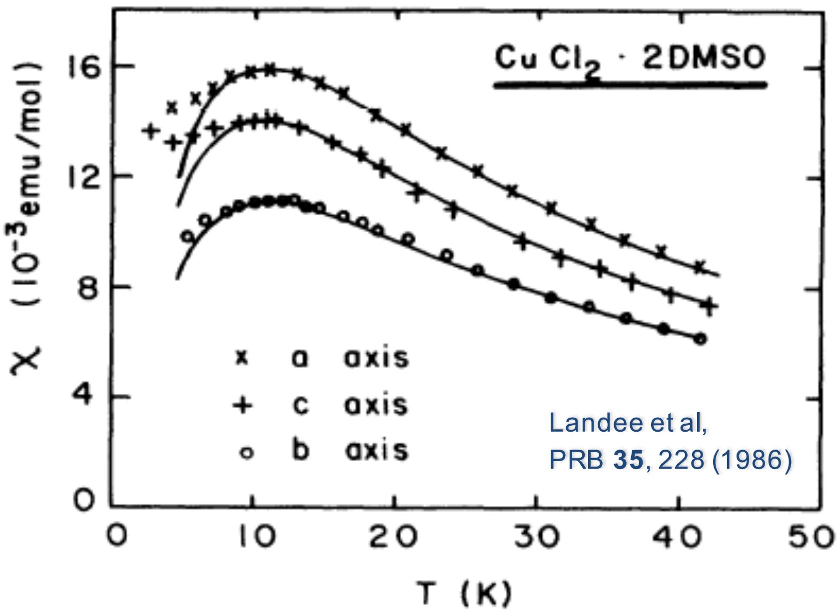




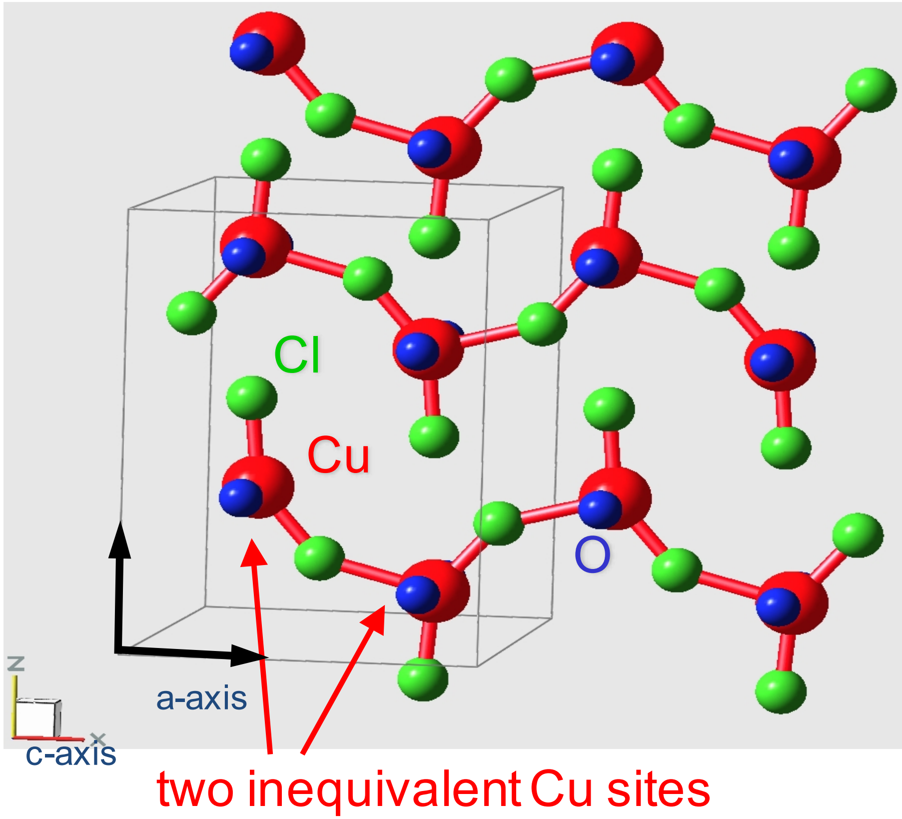
Excitation spectrum of CuPzN

M.B. Stone et al, Phys. Rev. Lett 2003

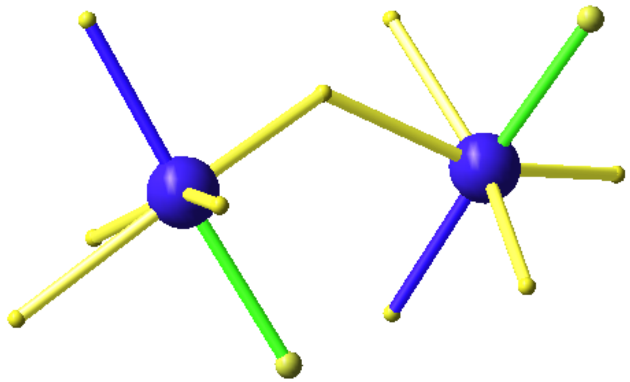
CDC S=1/2 chain



Broad maximum in magn. susceptibility



Magneto-electric quantum magnet CDC



$$D/J = 0.0102(5) \qquad g^s = 0.068(3)$$

Y. Chen et al, Phys. Rev. B **75**, 214409 (2007)

1) Staggered gyromagnetic factor

$$g = \begin{pmatrix} 2.28 & 0 & \pm g_s \\ 0 & 1.97 & 0 \\ \pm g_s & 0 & 2.12 \end{pmatrix} = g^u \pm g^s,$$

2) Staggered Dzyaloshinskii-Moriya interactions

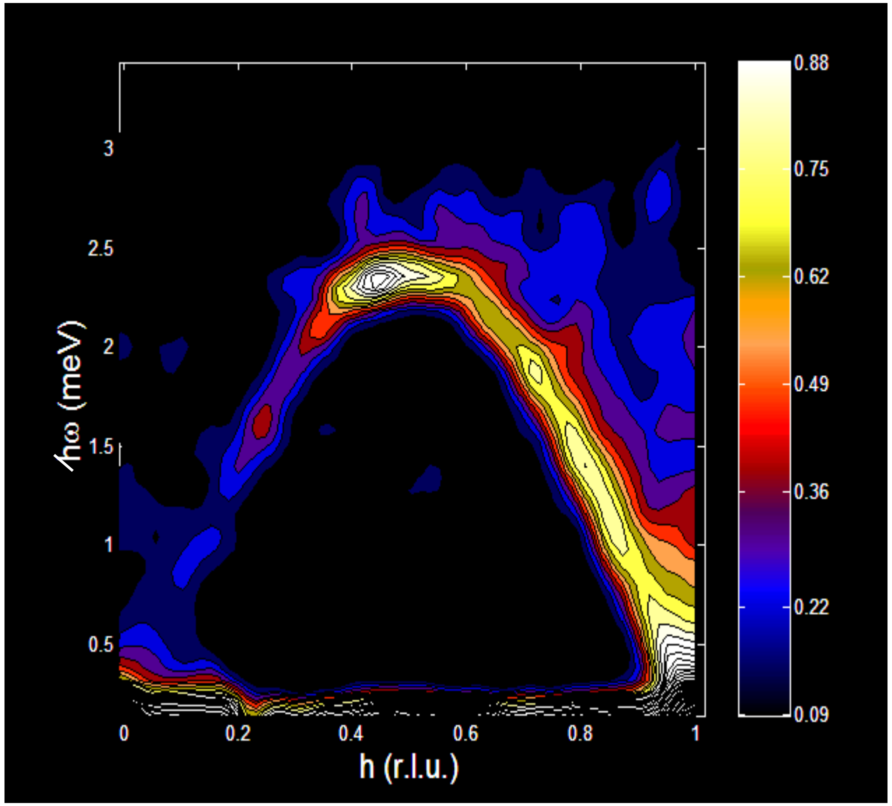
$$H_{DM} = \sum_i (-1)^i \mathbf{D} \cdot (\mathbf{S_{i-1}} \times \mathbf{S_i})$$

$$\mathbf{h} = \frac{1}{2J}\mathbf{D} \times g^u \mathbf{H} + g^s \mathbf{H},$$

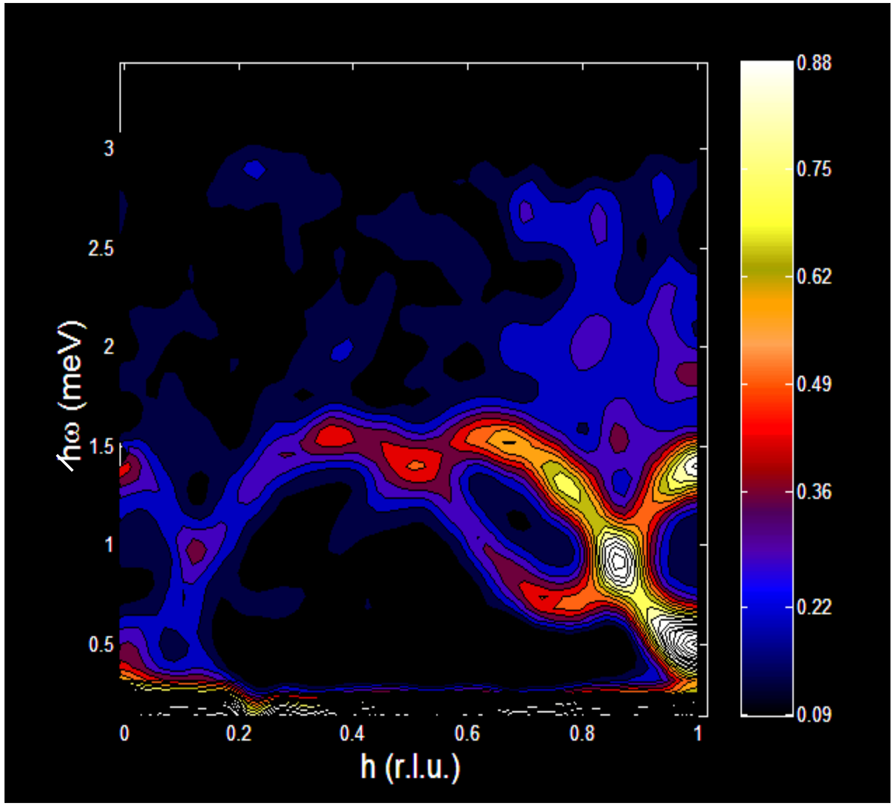
$$\mathcal{H} = J \Sigma_i \mathbf{S}_i \mathbf{S}_{i+1} - \Sigma_{j,a,b} H^a[g^u_{ab} + (-1)^j g^s_{ab}]S^b_j + \Sigma_j (-1)^j \mathbf{D}\cdot(\mathbf{S}_{j-1} \times \mathbf{S}_j)$$

S(Q,ω) measured using neutron scattering

Zero field and T=40mK



H=11T≈0.8J and T=40mK

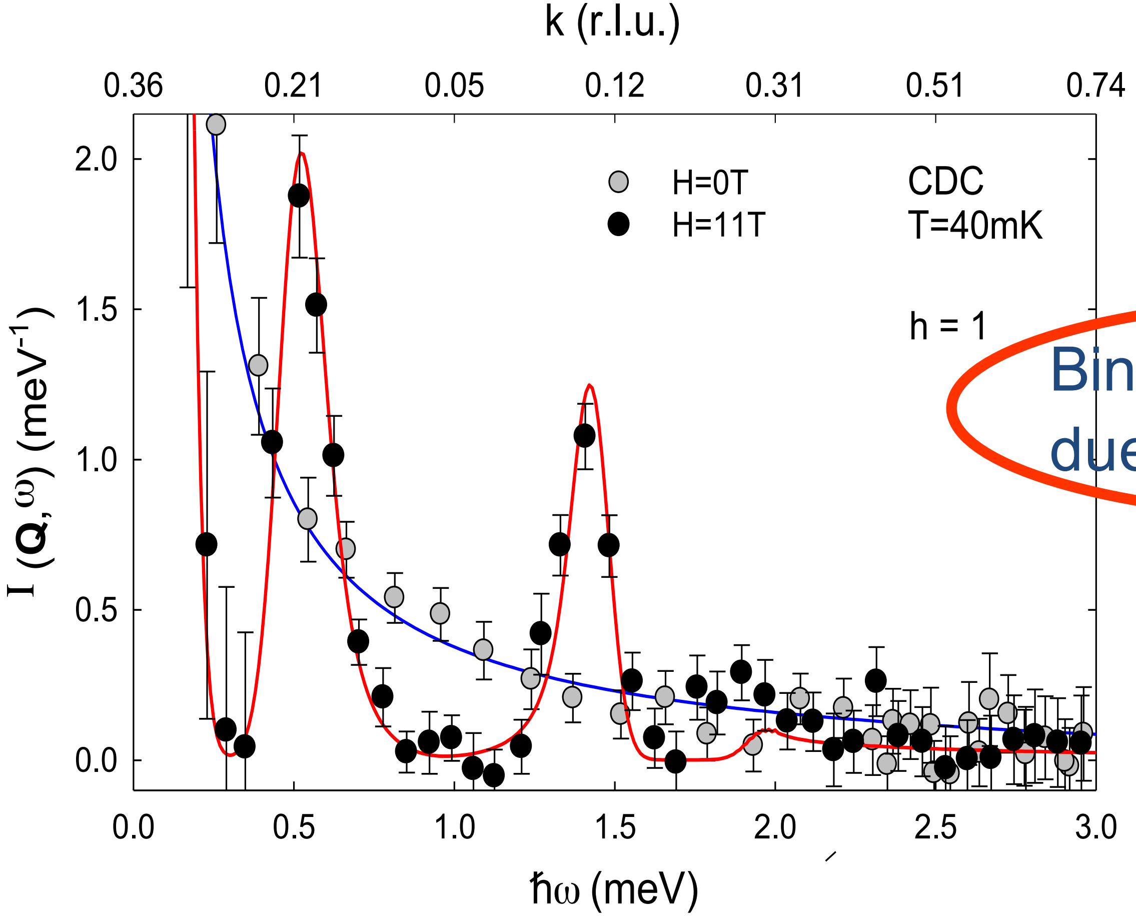


Two-spinon continuum of AF spin-1/2 Heisenberg chain with J=1.5meV
Intense lower bound $\hbar\omega = \pi/2\, J\, |\sin(\pi h)|$

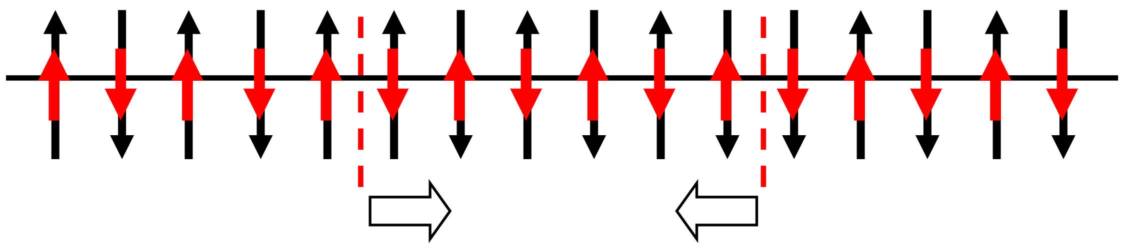
Spectrum at high field consists of well-defined excitations

M. Kenzelmann et al, Phys. Rev. Lett. **93**, 017204 (2004)

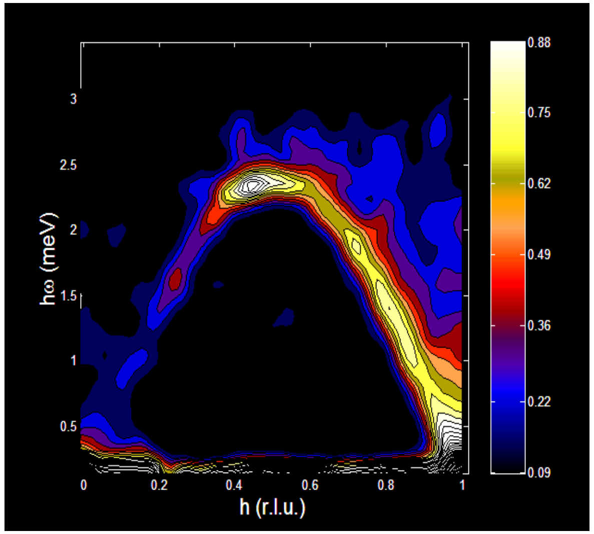
Bound spinons in staggered fields

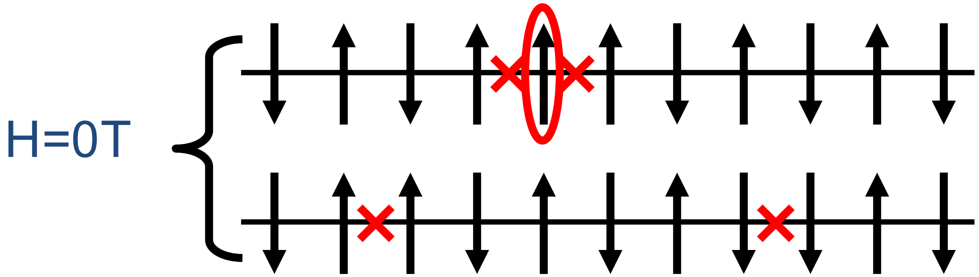


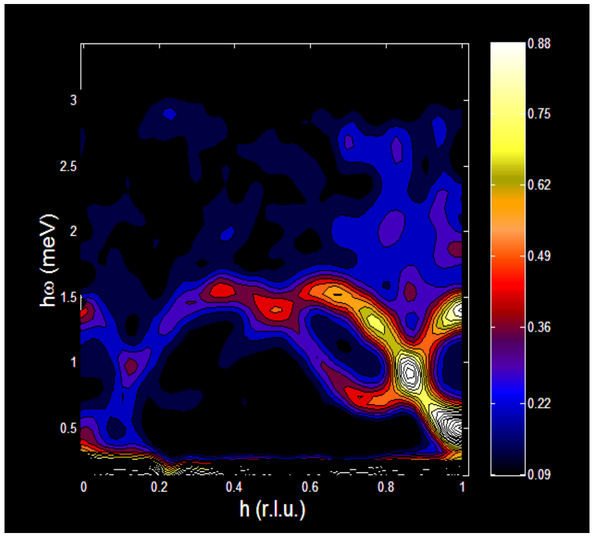
Binding of Spinons due to staggered field



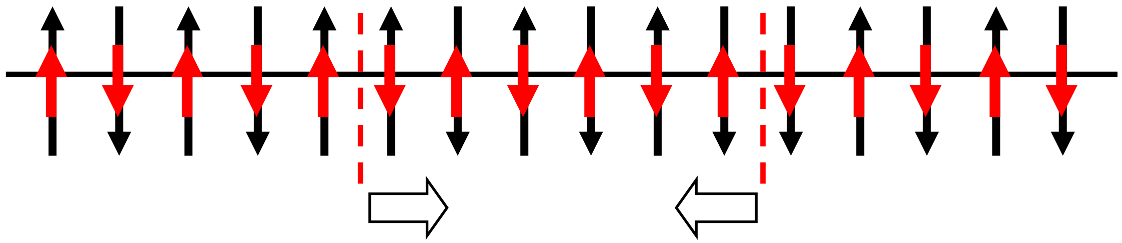
Spinons and Spinon binding in staggered fields





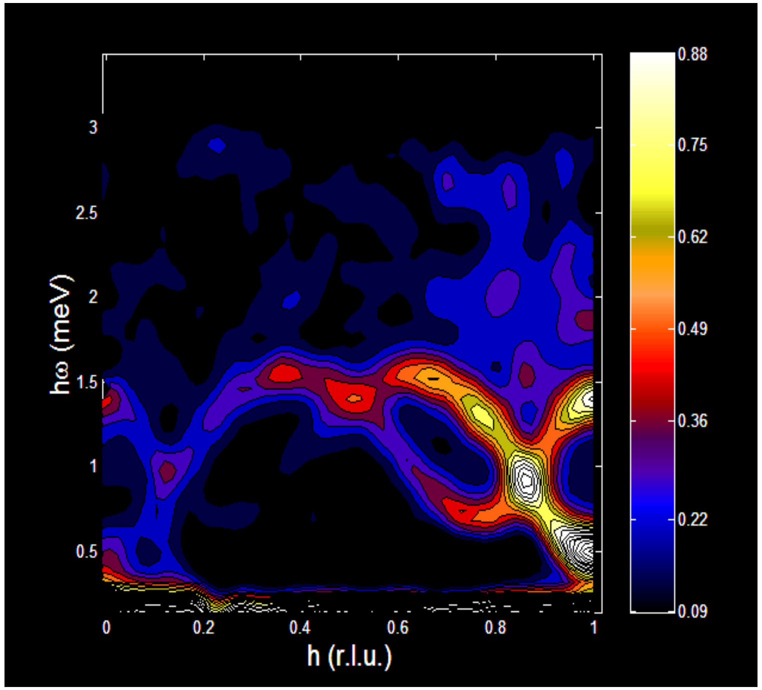


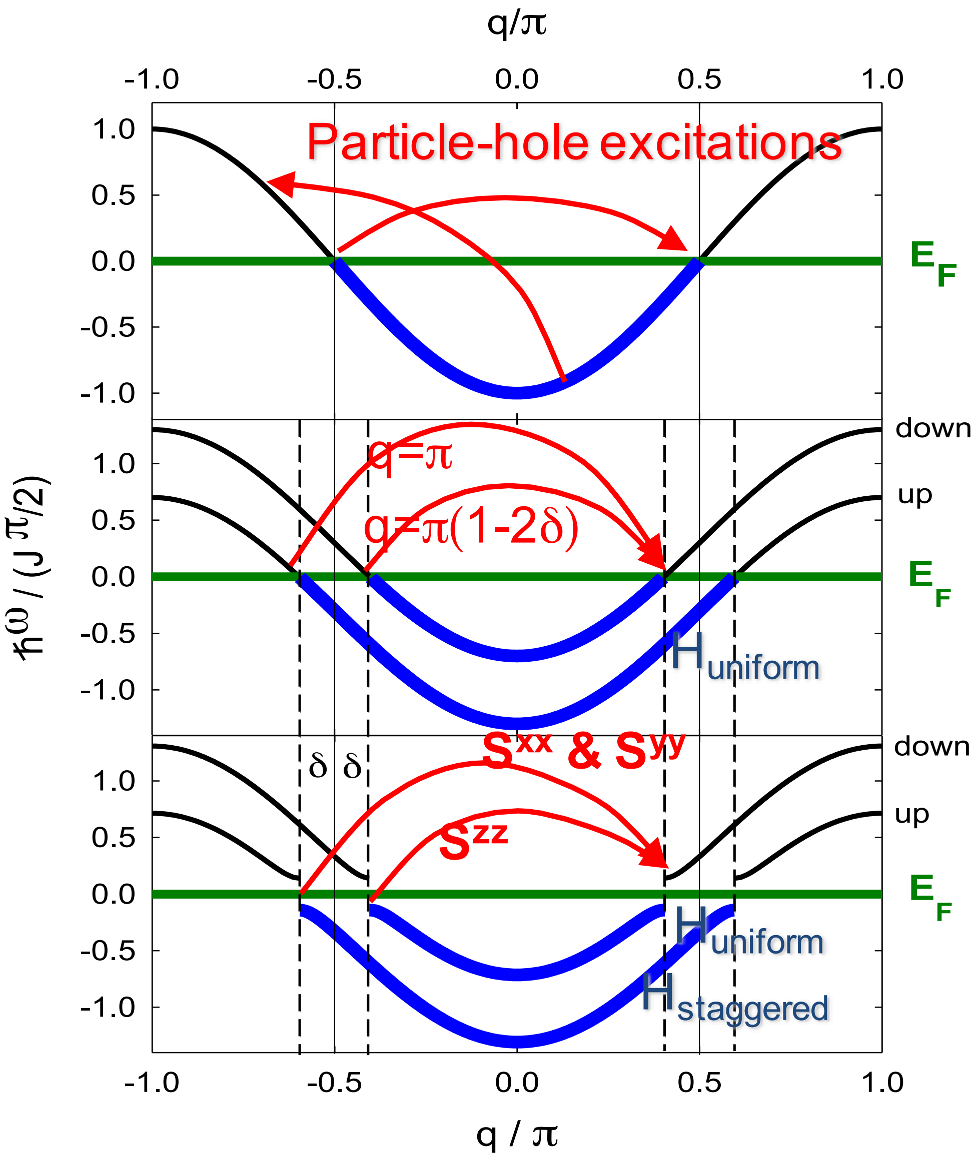
H > 0T & staggered (AF) fields



M. Kenzelmann et al, Phys. Rev. Lett. **93**, 017204 (2004)

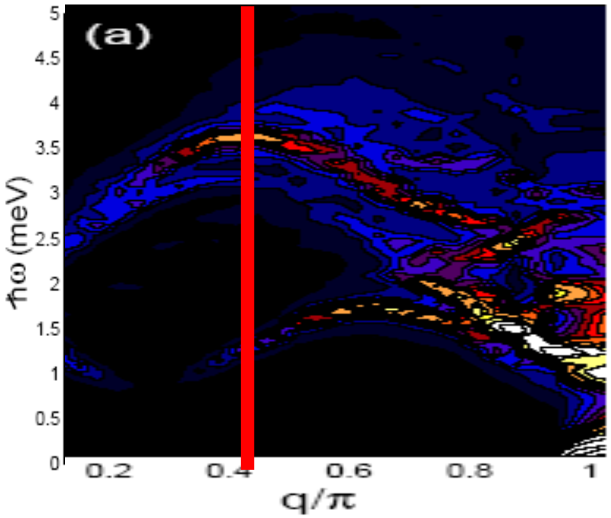
Bound spinons in the Spin fermion model

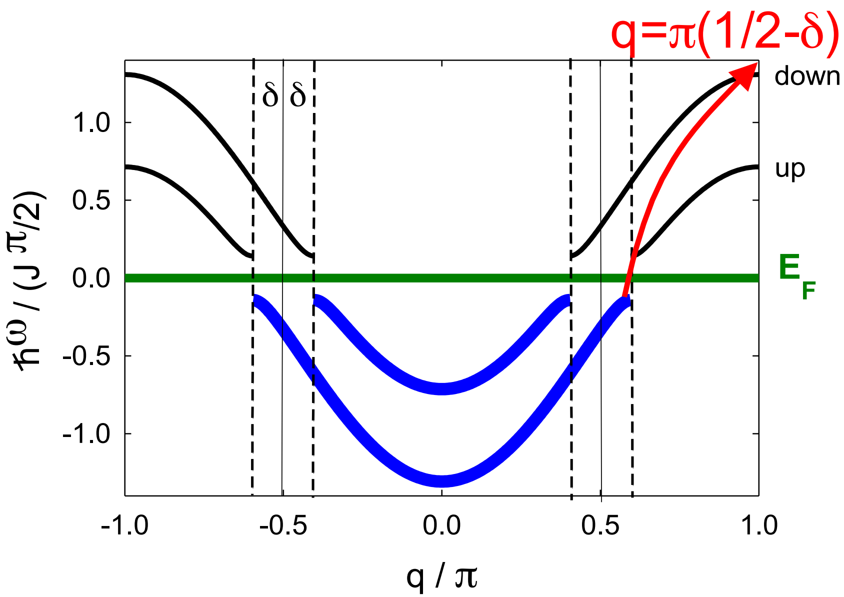




diverging density of states leads to increase of scattering at states close to Fermi surface

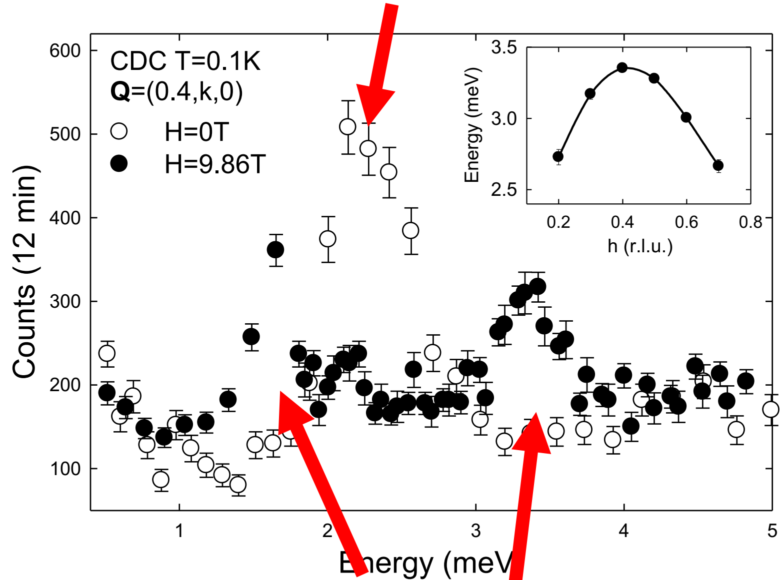
Small energy scales can have an effect at much higher energies





M. Kenzelmann et al, Phys. Rev. B **71**, 094411 (2005)

- H=0T: lower bound of two-spinon continuum



- H~10T: lower-energy branch at ~ 1.5meV

- H~10T: novel excitation at ~ 3.4meV