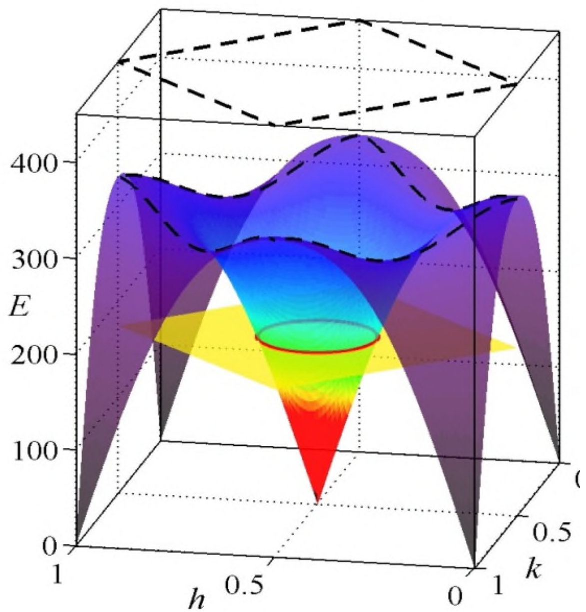
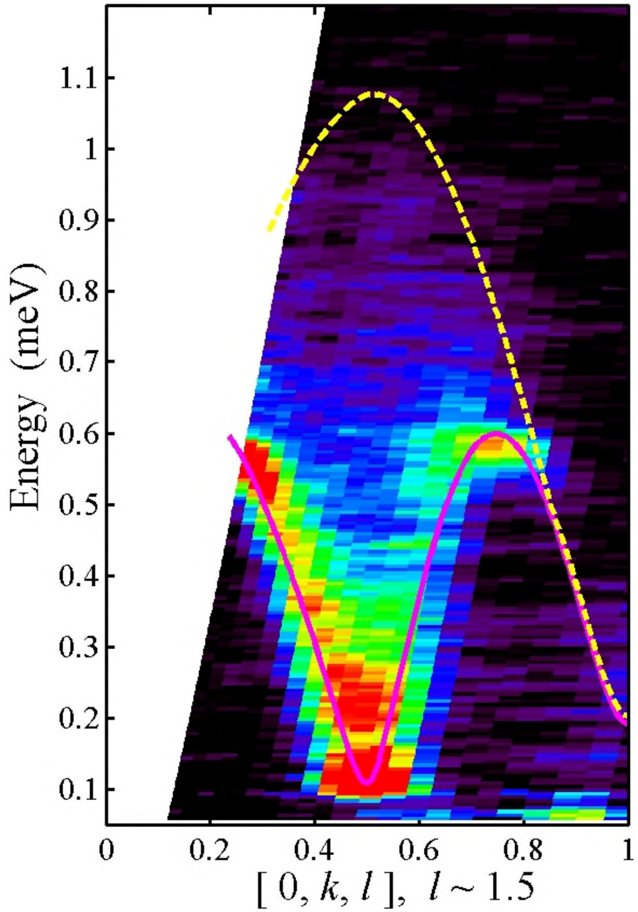
Neutron scattering as a tool to study quantum magnetism

Radu Coldea

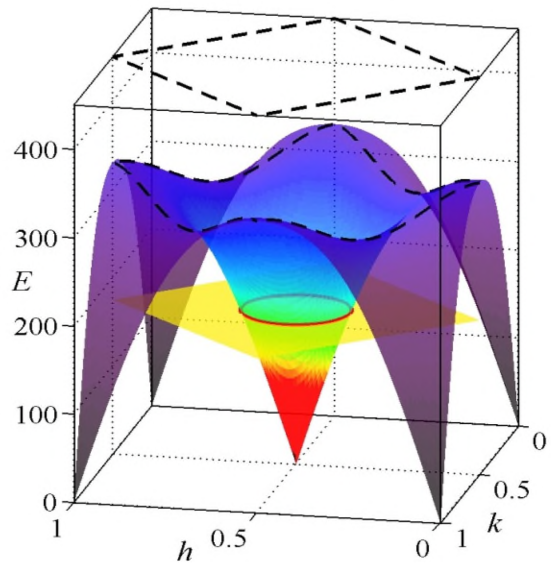
Oxford





Outline

- principles of (magnetic) neutron scattering
- spin waves in a Heisenberg ferromagnet
- spin waves in square-lattice AFM La_2CuO_4

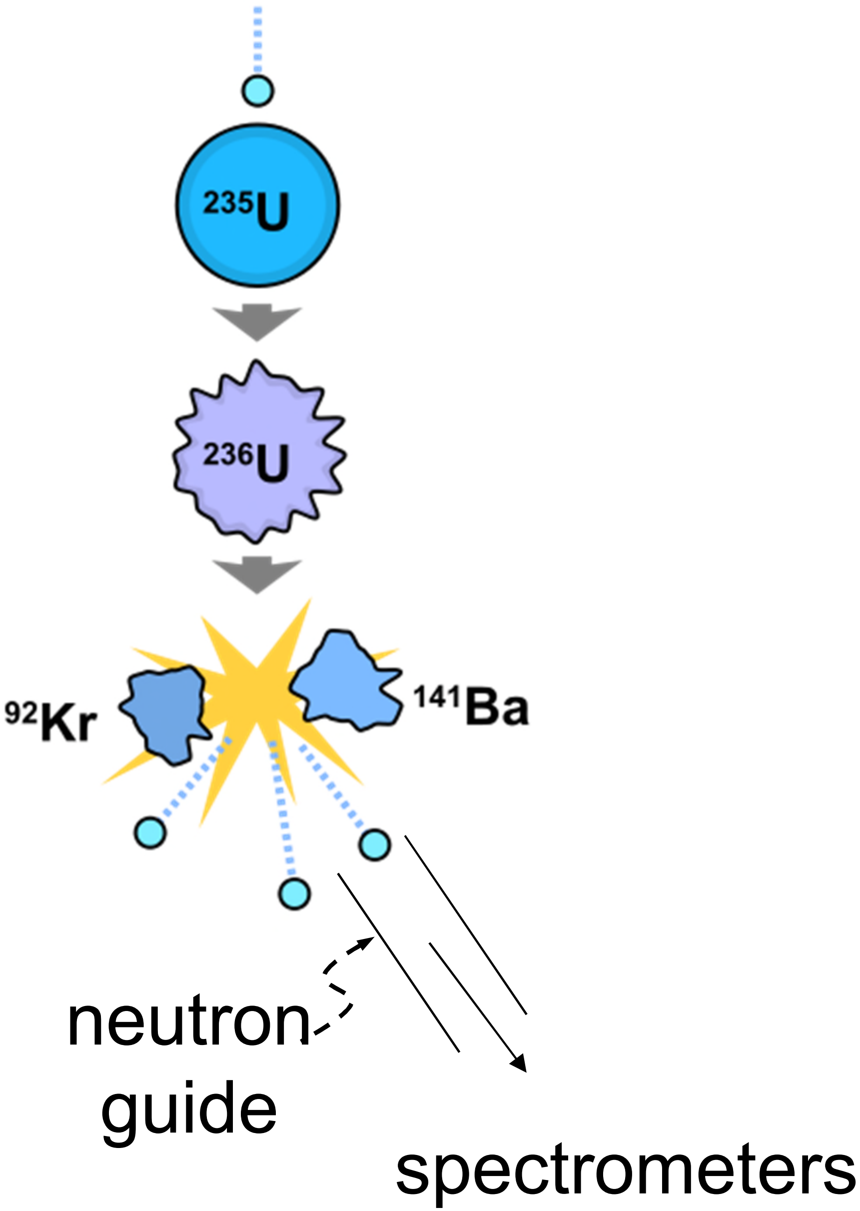


- quantum renormalization, spinons and method to determine Hamiltonian – triangular AFM $S = 1/2$ Cs_2CuCl_4

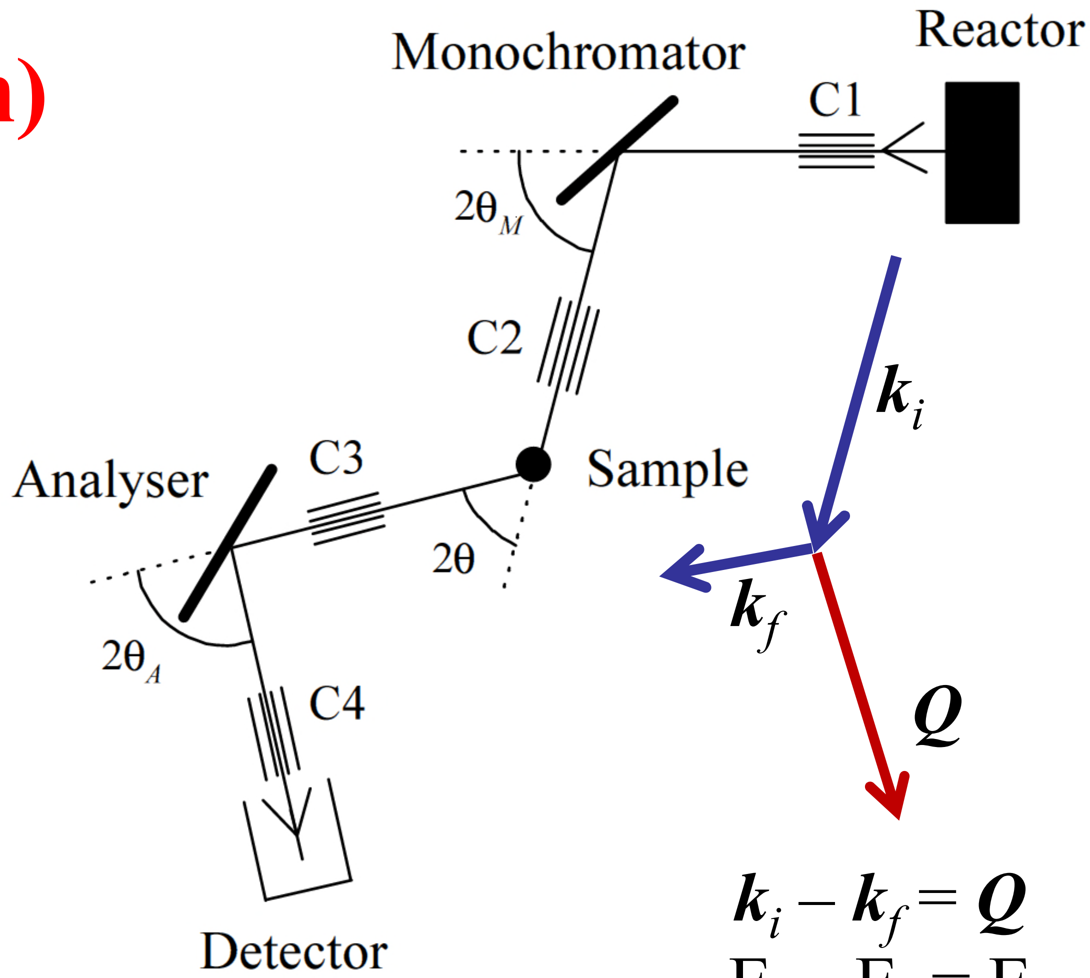
- quantum phase transition in the Ising chain $CoNb_2O_6$ in transverse field

Neutron reactors (Institute Laue Langevin)

- nuclear fission of Uranium



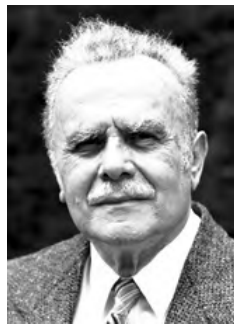
Three-axis neutron spectrometer



$$\boldsymbol{k}_i - \boldsymbol{k}_f = \boldsymbol{Q}$$

$$E_i - E_f = E$$

Clifford Shull

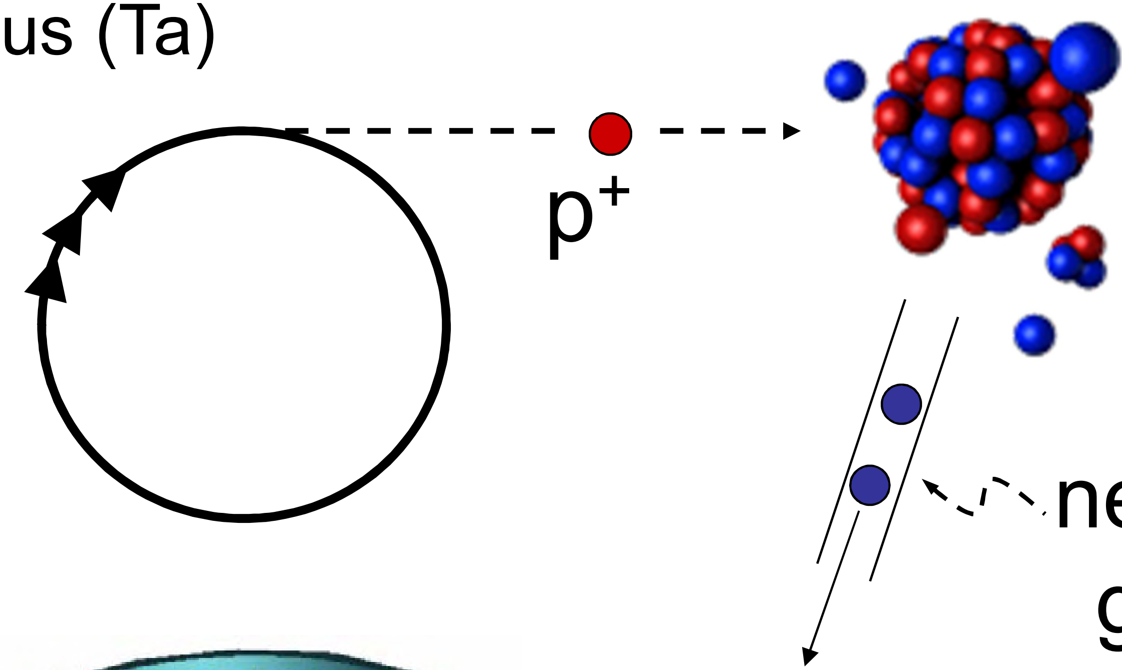


Bertram Brockhouse

Spallation neutron sources (ISIS, SNS …)

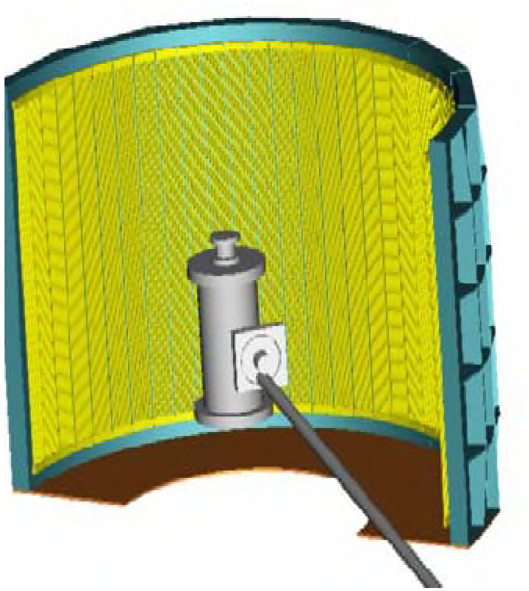
- "evaporation" when fast protons hit a heavy nucleus (Ta)

proton synchrotron accelerator ~800MeV



neutron guide

LET@ISIS

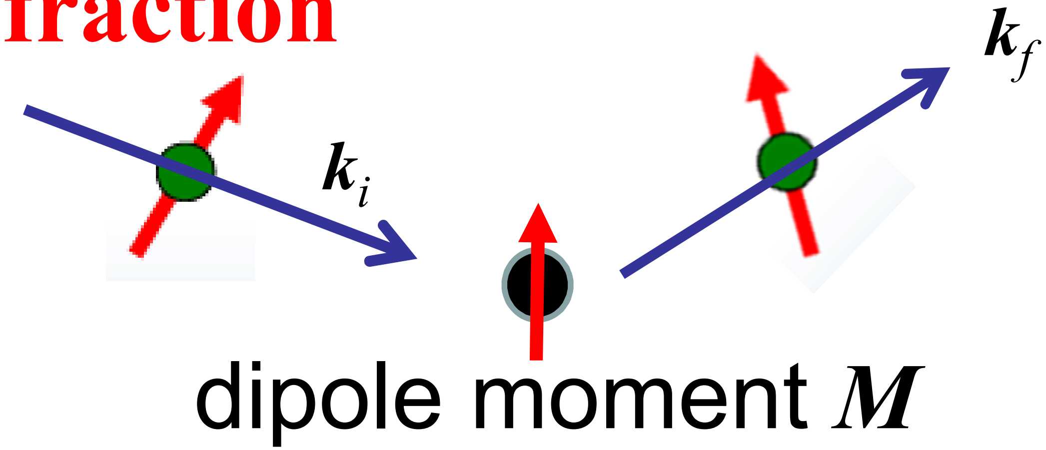


time-of-flight spectrometer

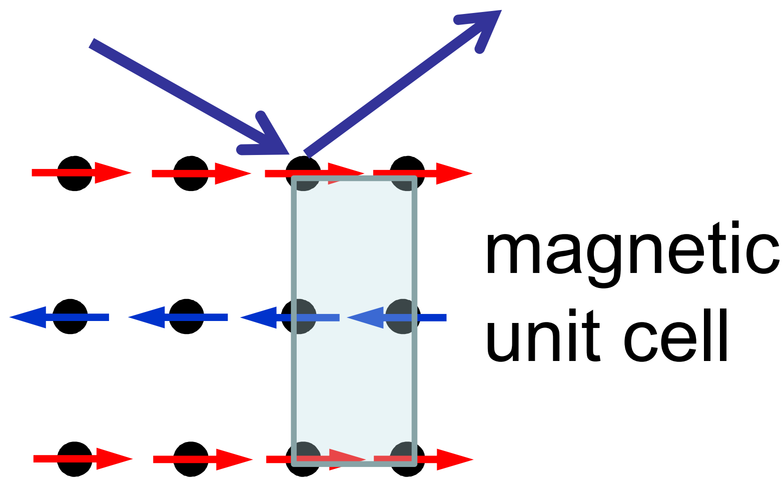
~40,000 detector elements count simultaneously (time-stamp each arriving neutron

Magnetic neutron diffraction

Neutrons have
- no charge
- spin-1/2 moment

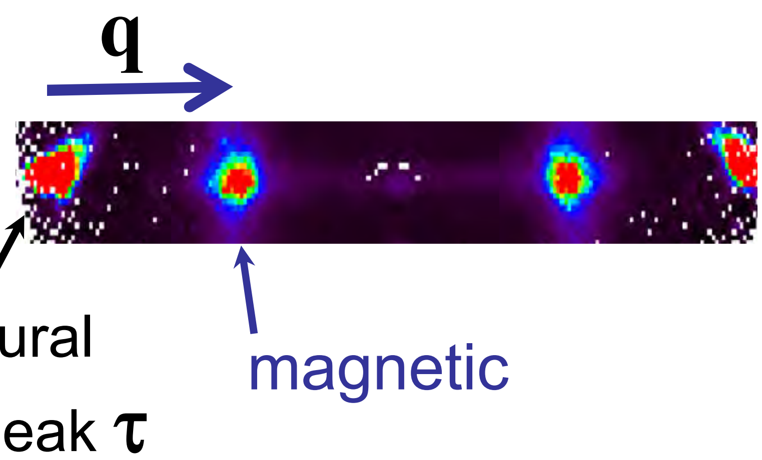


Periodic magnetic order
=> magnetic Bragg peaks τ
at $Q = \tau \pm \mathbf{q}$

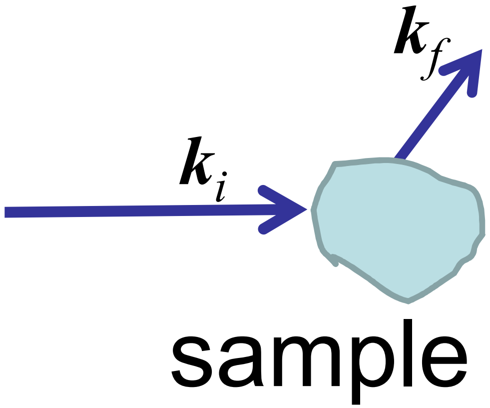


- Intensity ~ $|M_{\perp}(Q)|^2$

Fourier transform of magnetic moment density (perp to scattering wavevector Q)

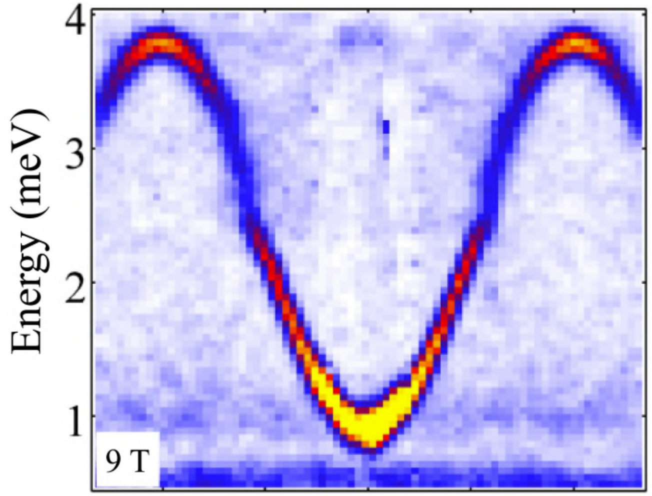


Inelastic magnetic neutron scattering



$|\lambda\rangle$ excited state

$|0\rangle$ ground state



matrix element for transition $\langle\lambda|S^{\alpha}(\boldsymbol{Q})|0\rangle$

$$S^{\alpha}(\boldsymbol{Q}) = \frac{1}{\sqrt{N}}\sum_j e^{\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{R}_j}\, S_j^{\alpha}$$

Fourier transform of magnetic moment density M^{α}

$$S^{\alpha\alpha}(\boldsymbol{Q},\omega) = \sum_{\lambda} |\langle\lambda|S^{\alpha}(\boldsymbol{Q})|0\rangle|^2\ \delta(\hbar\omega + E_0 - E_{\lambda})$$

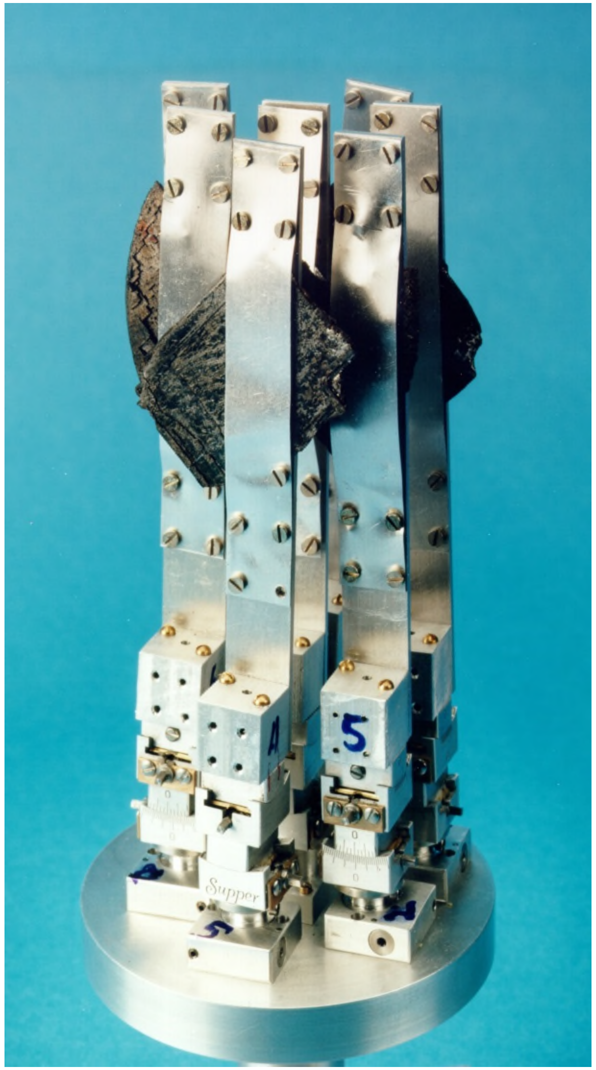
$$\frac{d^2\sigma}{d\Omega dE'} \equiv \frac{k'}{k} N r_0^2 \left|\frac{g}{2}F(\mathbf{Q})\right| e^{-2W(\mathbf{Q})} \sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{Q}_{\alpha}\hat{Q}_{\beta}\right) \boldsymbol{S}^{\alpha\beta}(\mathbf{Q},\omega)$$

Fourier transform of magnetic e- density

polarization factor

Single crystals for inelastic neutron scattering

La_2CuO_4

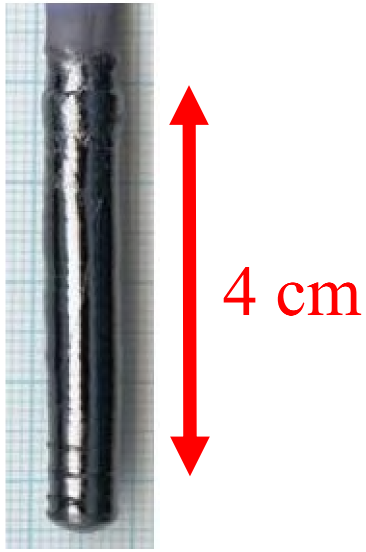


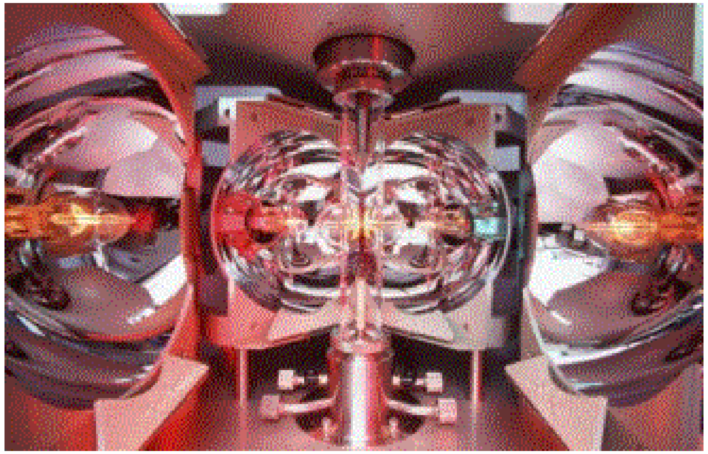
7 single-crystal mount ~50 g
(flux growth)

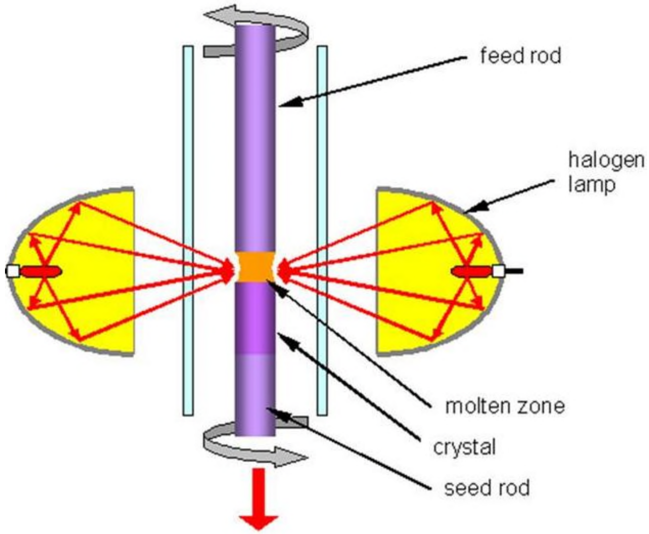
Cs_2CuCl_4
(solution growth)

2.5 cm

$CoNb_2O_6$
(mirror furnace growth)





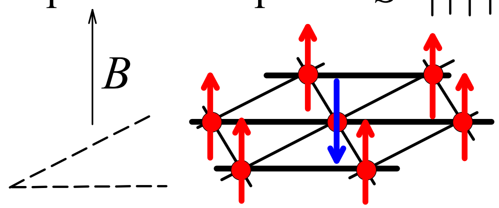


floating-zone mirror furnace

Spin waves in a Heisenberg ferromagnet

$H = -\sum_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$ if all $J_{ij} > 0$ T=0 ground state is $\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow$

- neutrons flip over one spin $S^- |\uparrow\uparrow\uparrow\uparrow\uparrow\cdots\rangle = |\uparrow\uparrow\downarrow\uparrow\uparrow\cdots\rangle$



$$H = \sum_{ij} -J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - h \sum_i S_i^z$$

$$= \sum_{ij} \frac{-J_{ij}}{2} (S_i^- S_j^+ + S_i^+ S_j^-) - \sum_{ij} J_{ij} S_i^z S_j^z - h \sum_i S_i^z$$

hopping

coherent propagation of spin-flip states (if Hamiltonian conservs S^z)

magnon energy

$$\omega(\boldsymbol{q}) = -J(\boldsymbol{q}) + J(0) + h$$

exact result

$$J(\boldsymbol{q}) = \frac{1}{2} \sum_{ij} J_{ij} \, e^{i\boldsymbol{q}\boldsymbol{r}_{ij}}$$

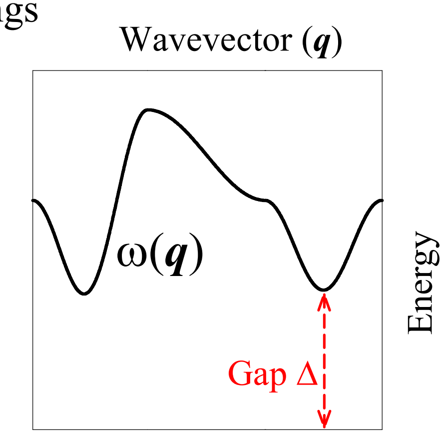
Fourier transform of magnetic couplings

Zeeman energy

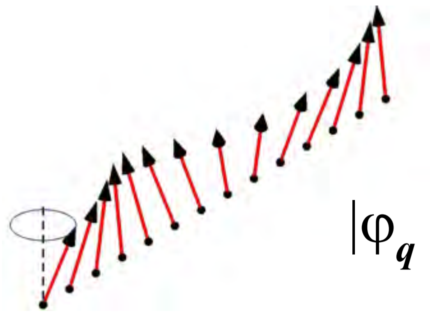
One spin flip manifold $\left\{ |\uparrow\uparrow\downarrow\uparrow\uparrow\cdots\rangle \right.$

S^-

$|\uparrow\uparrow\uparrow\uparrow\uparrow\cdots\rangle$

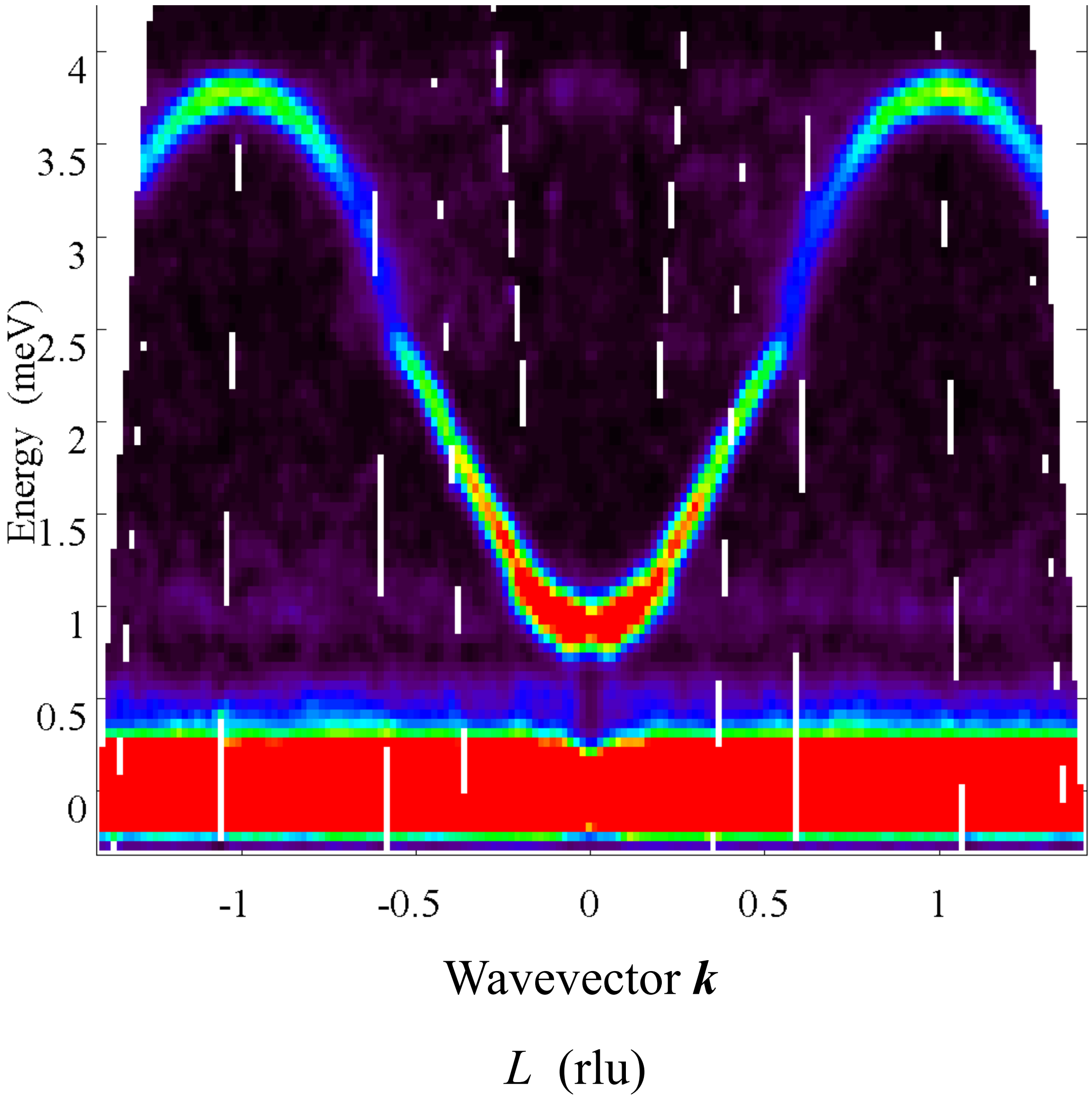


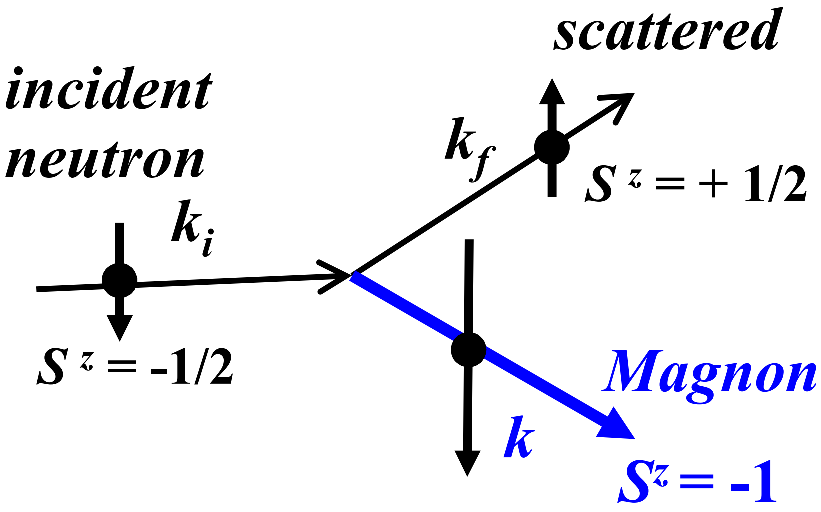
Dispersion images exchange Hamiltonian

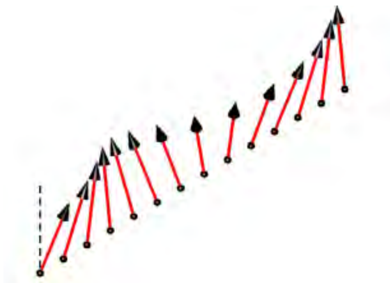


$$|\varphi_{\boldsymbol{q}}\rangle = \frac{1}{\sqrt{N}} \sum_i e^{i\boldsymbol{q}\boldsymbol{r}_i} |\downarrow_i\rangle$$

Neutron scattering by ferromagnetic magnons



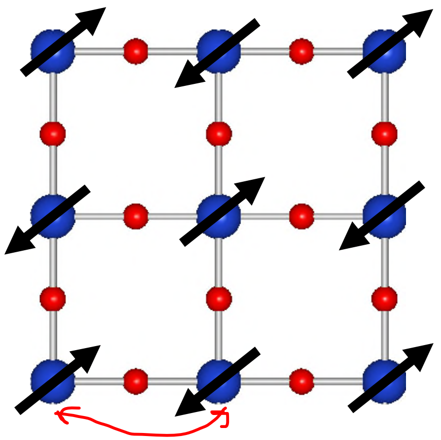




$$\boldsymbol{k}_i - \boldsymbol{k}_f = \boldsymbol{k}$$

$$E_i - E_f = \hbar\,\omega(\boldsymbol{k})$$

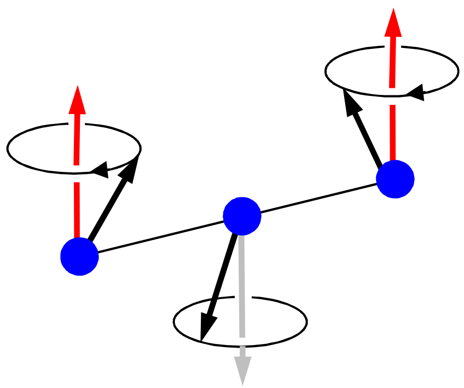
Spin waves in the square-lattice anti-ferromagnet



J antiferromagnetic

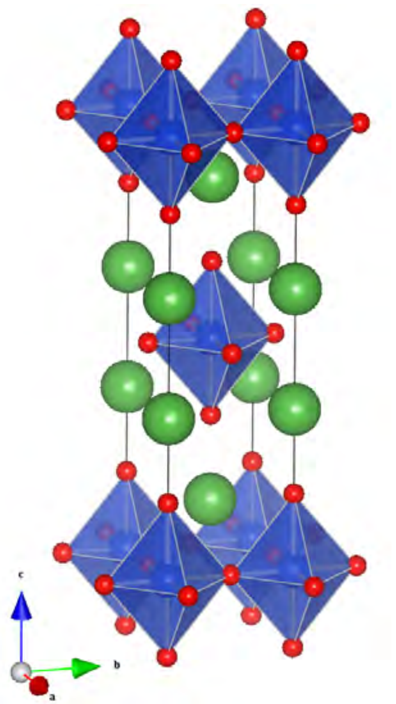
Ground state has Neel order
(<S> reduced by quantum fluctuations)

$$\boldsymbol{S}_i \cdot \boldsymbol{S}_j = S_i^z S_j^z + \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right)$$



Spin wave excitations
(approximate eigenstates)

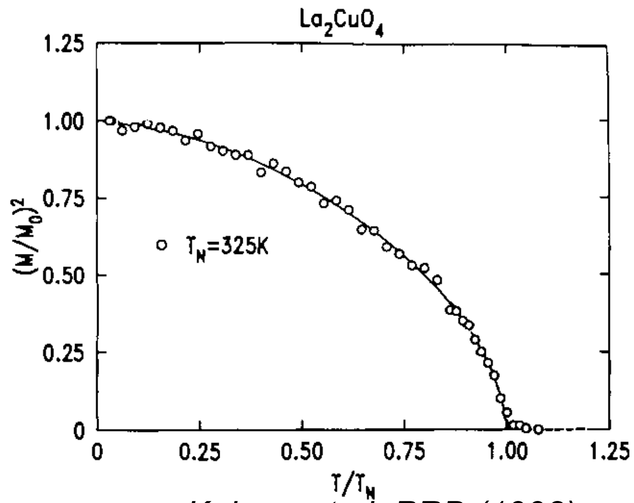
La_2CuO_4



- insulating parent of high-T_C cuprates

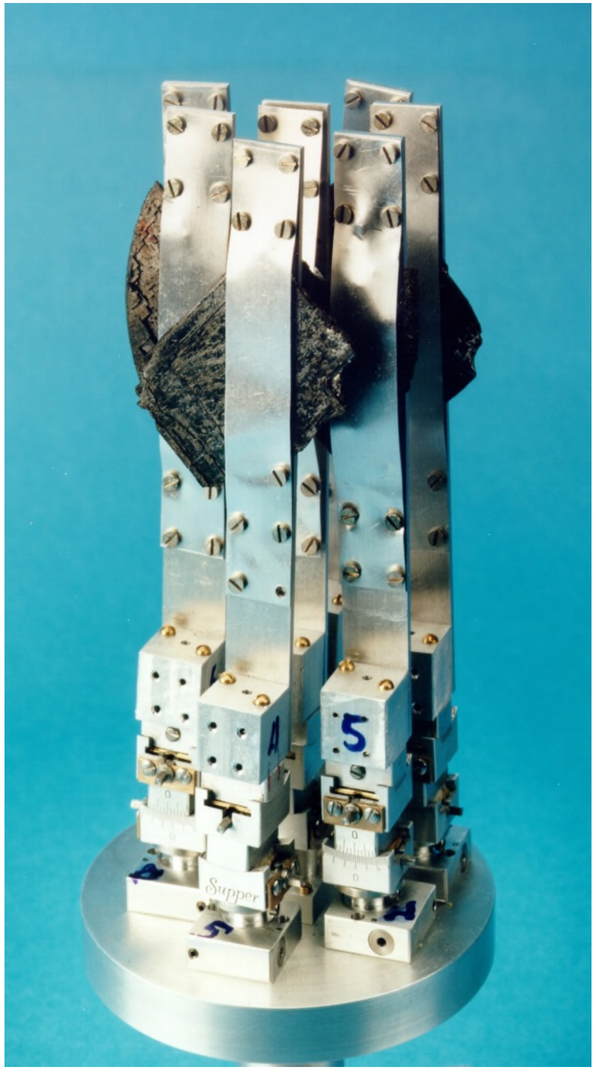
- square-lattice of CuO_2 planes, Cu^{2+} S=1/2

Magnetic Bragg peak (1/2,1/2,0)

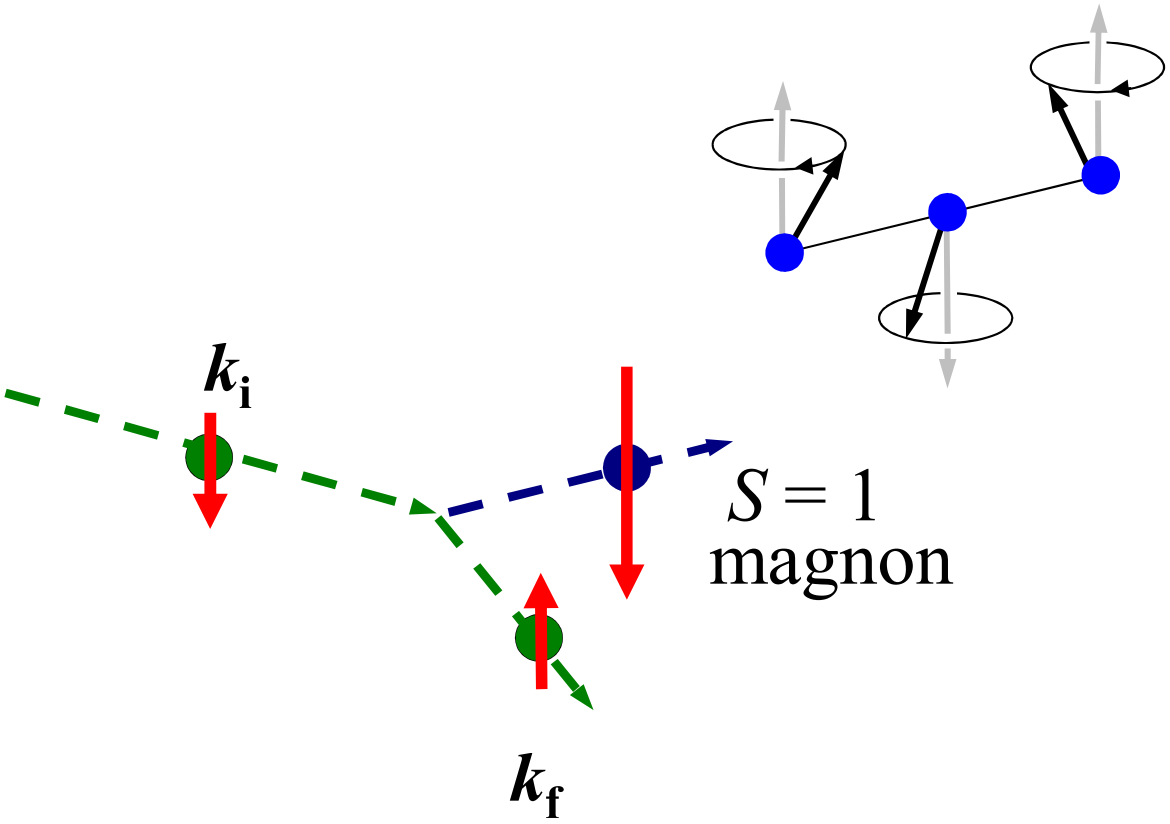


Keimer at al, PRB (1992)

Neutron scattering experiments on La_2CuO_4



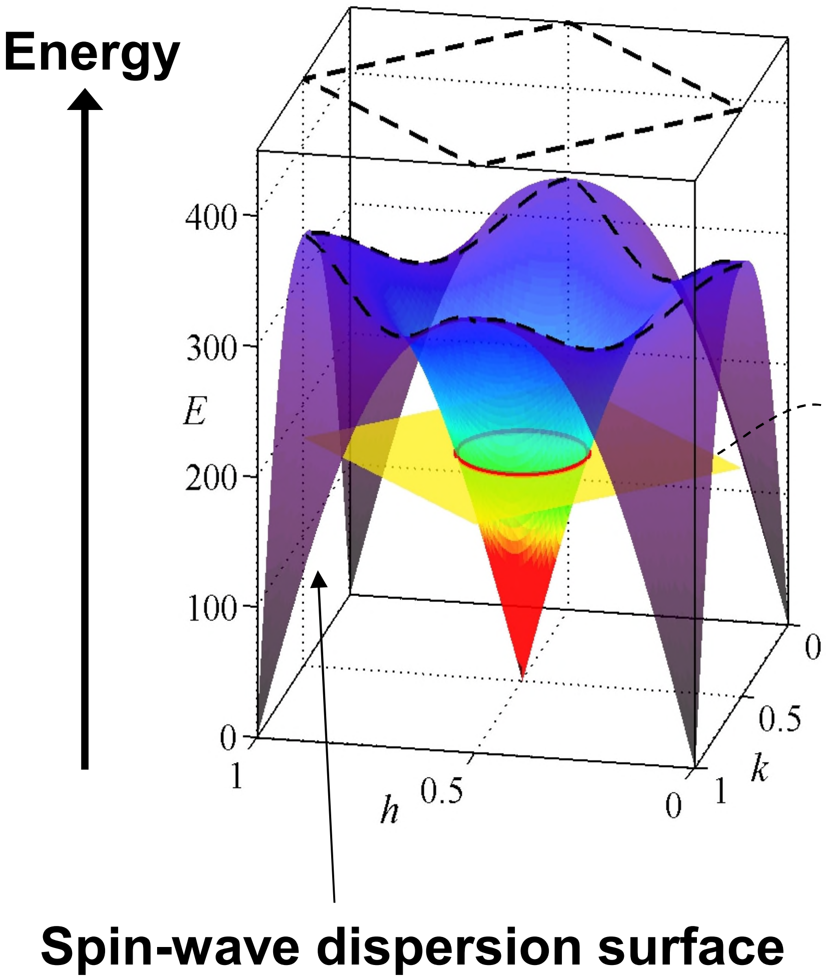
7 single crystal mount, ~50 g

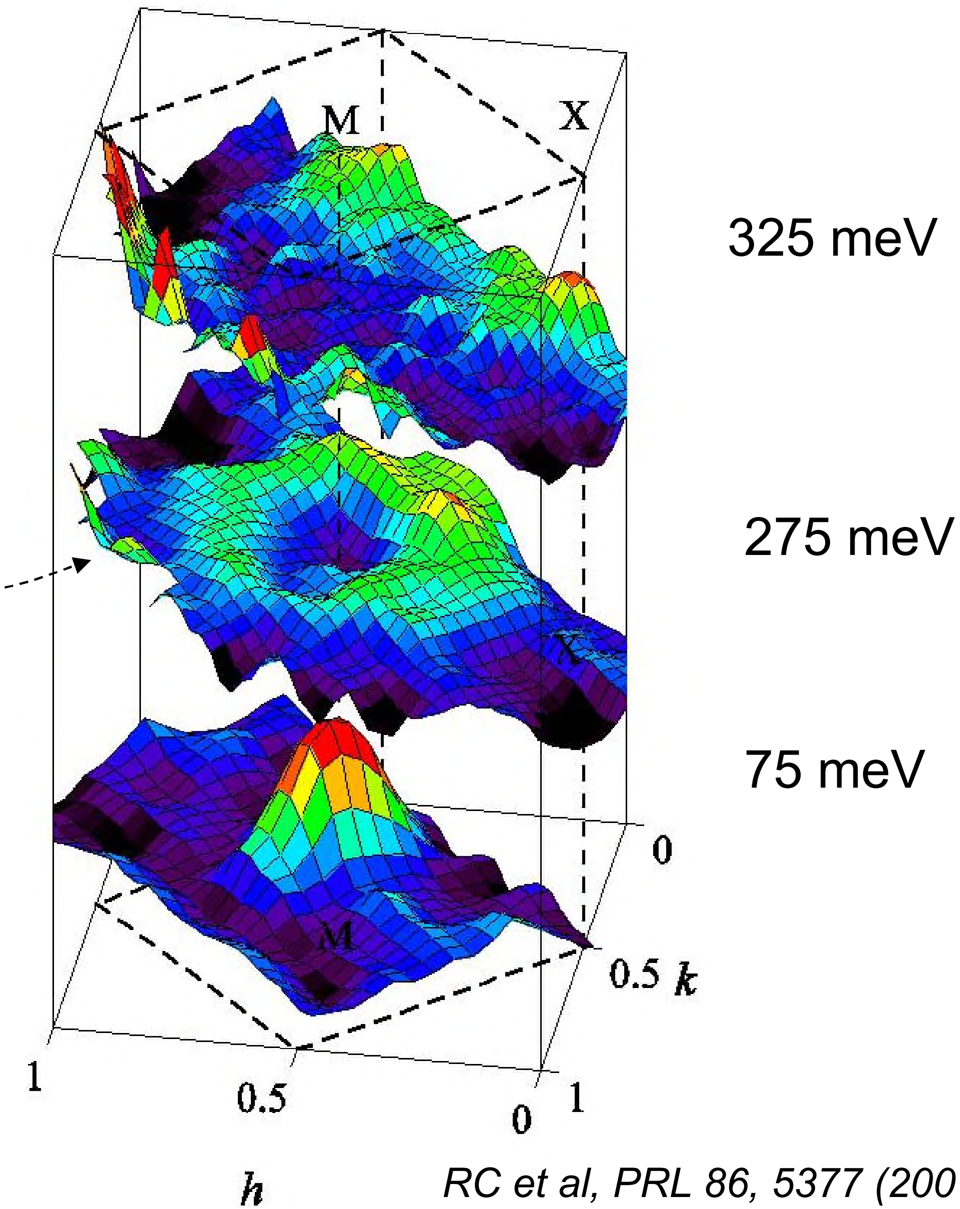


RC et al, PRL 86, 5377 (2001)

Magnetic excitations in La_2CuO_4

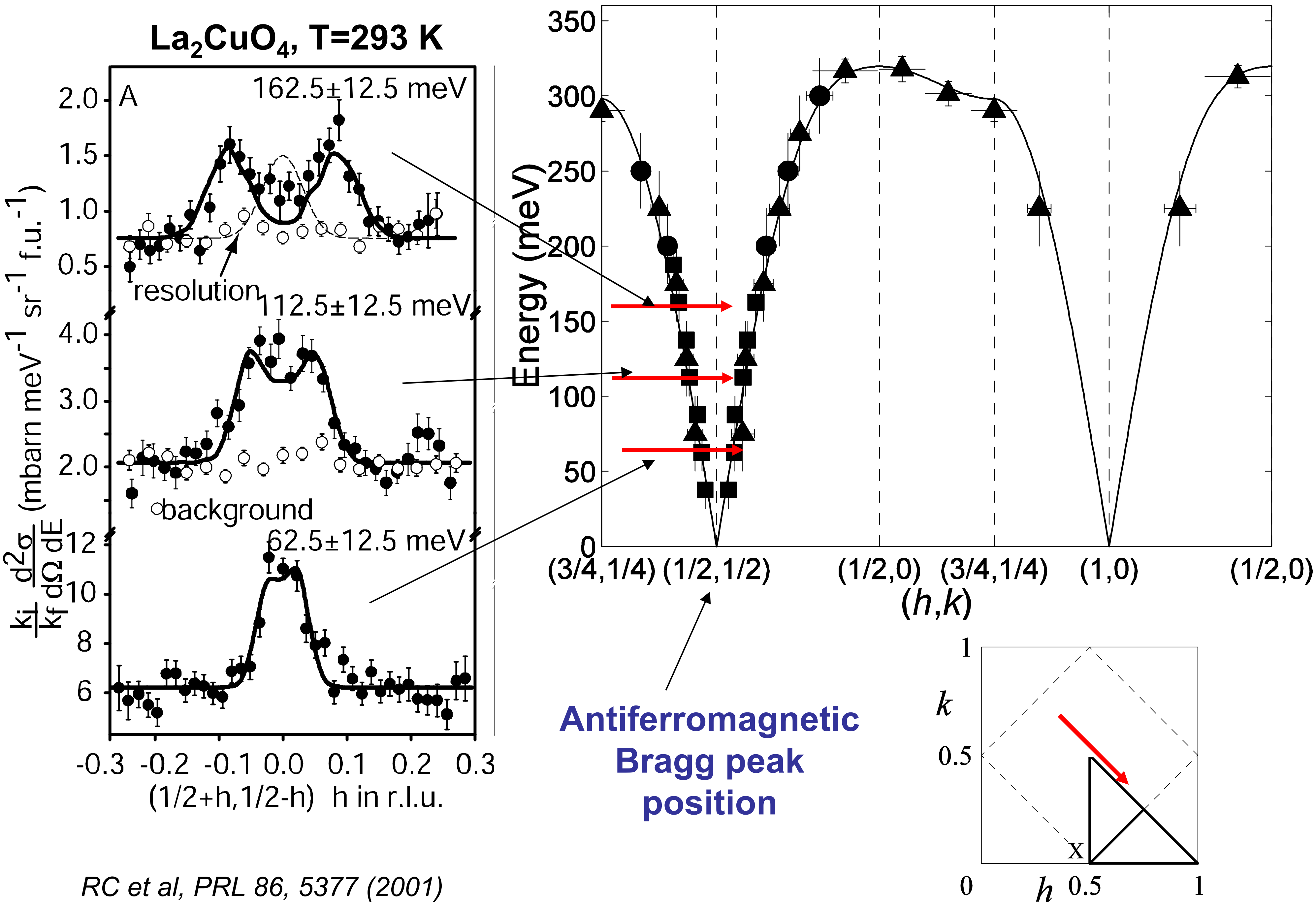
Collect maps of magnetic scattering in the whole 2D Brillouin zone (*h,k*) at increasing energies *E*





RC et al, PRL 86, 5377 (2001)

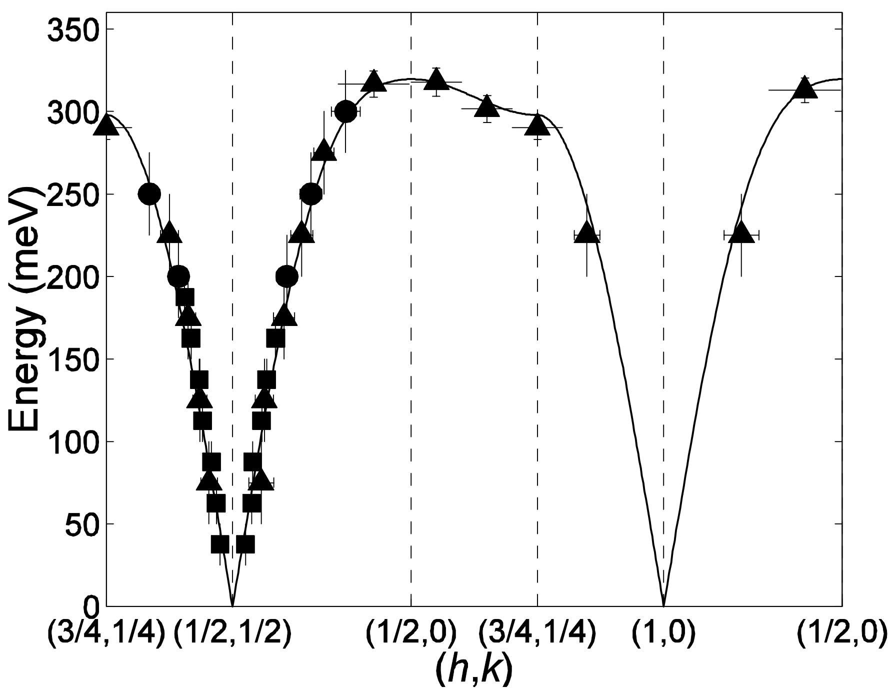
Dispersion relations

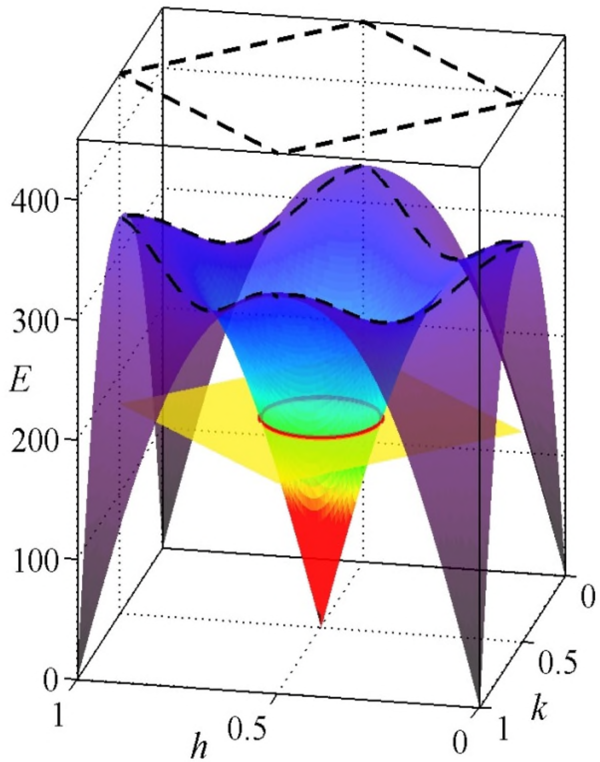


RC et al, PRL 86, 5377 (2001)

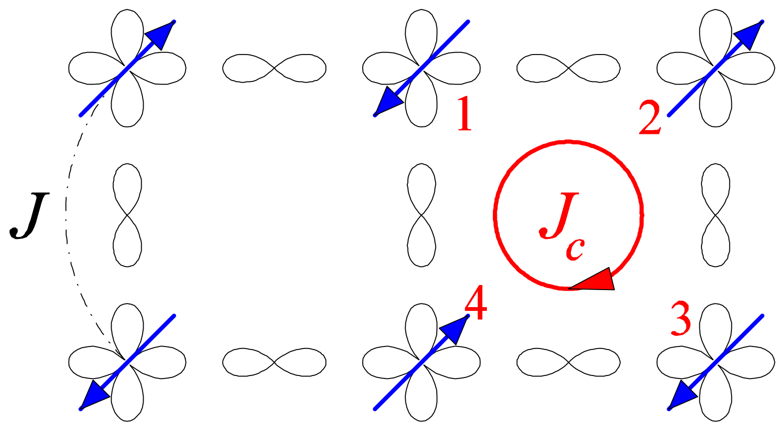
Dispersion relation and interactions

- dispersion shape is a direct fingerprint of the magnetic interactions





- "wiggle" in high-energy dispersion is evidence for a cyclic-exchange between the 4 spins on a plaquette in addition to the main exchange J



$$\mathcal{H} = J\sum_{\langle i,j\rangle} \boldsymbol{S}_i\cdot\boldsymbol{S}_j + J_c \sum_{\langle i,j,k,l\rangle} \{(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)(\boldsymbol{S}_k\cdot\boldsymbol{S}_l) + (\boldsymbol{S}_i\cdot\boldsymbol{S}_l)(\boldsymbol{S}_k\cdot\boldsymbol{S}_j) - (\boldsymbol{S}_i\cdot\boldsymbol{S}_k)(\boldsymbol{S}_j\cdot\boldsymbol{S}_l)\}, \quad (1)$$

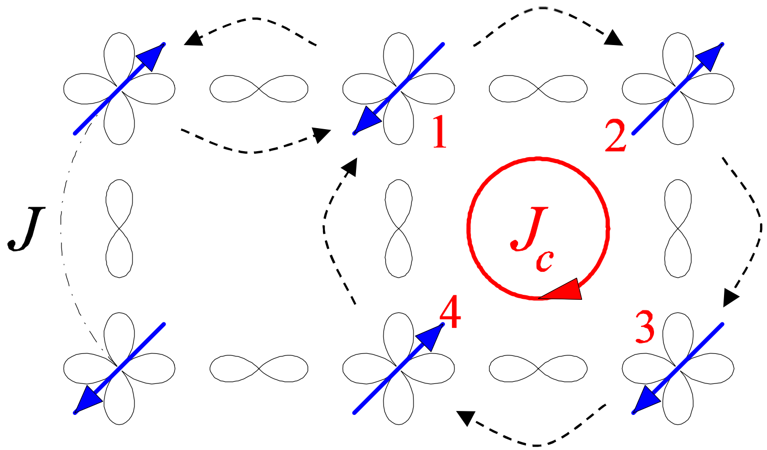
$J = 138\text{meV} \quad J_c = 38\text{meV}$

RC et al, PRL 86, 5377 (2001)

Ring exchange in the Hubbard model

Kinetic hopping

Potential energy

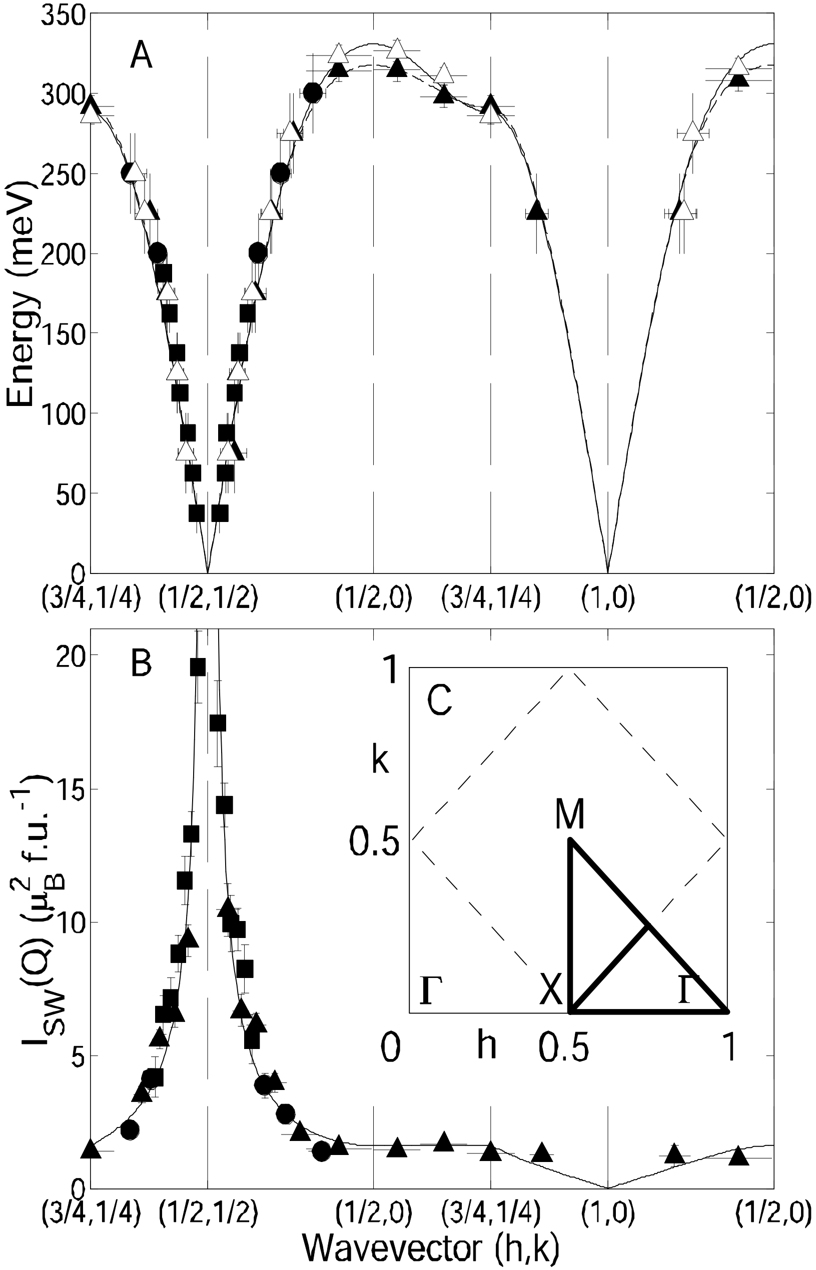


Expand up to 4 electron hops

A.H. MacDonald (1990), Takahashi (1977)

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J' \sum_{\langle i,i' \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_{i'} + J'' \sum_{\langle i,i'' \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_{i''}$$
$$+ J_c \sum_{\langle i,j,k,l \rangle} \{ (\boldsymbol{S}_i \cdot \boldsymbol{S}_j)(\boldsymbol{S}_k \cdot \boldsymbol{S}_l) + (\boldsymbol{S}_i \cdot \boldsymbol{S}_l)(\boldsymbol{S}_k \cdot \boldsymbol{S}_j)$$
$$- (\boldsymbol{S}_i \cdot \boldsymbol{S}_k)(\boldsymbol{S}_j \cdot \boldsymbol{S}_l) \} , \qquad (1)$$

Hubbard model parameters for La_2CuO_4



- dispersions and intensities well described by linear spin-wave theory

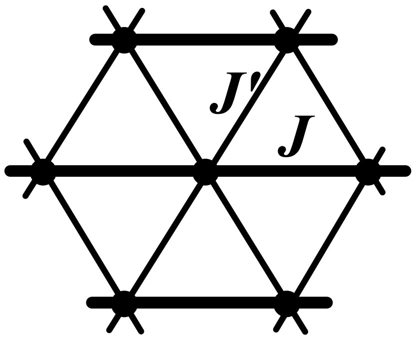
t = 0.30(2) eV, U = 2.2 (4) eV

U/t = 7.3 ± 1.3 (10 K)

Intensity renormalization factor
Zc = 0.51 +/- 0.13 (predicted 0.61)

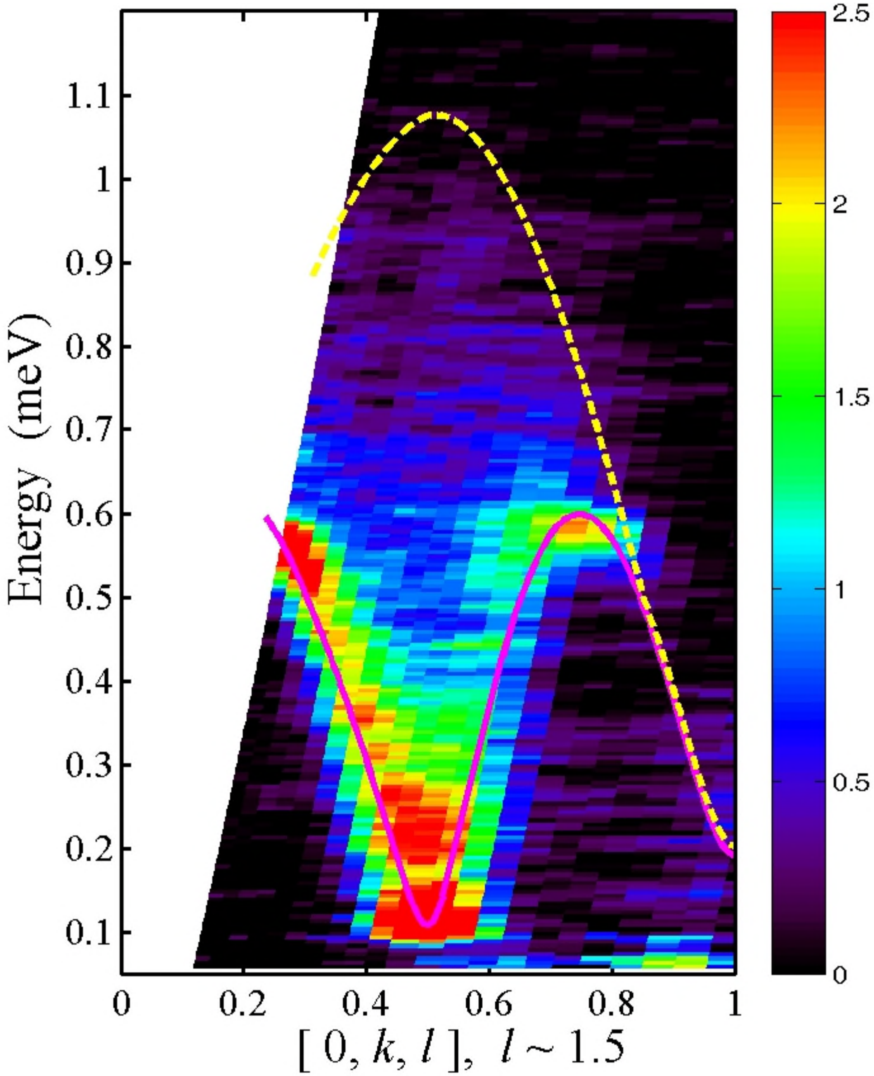
RC et al, PRL 86, 5377 (2001)

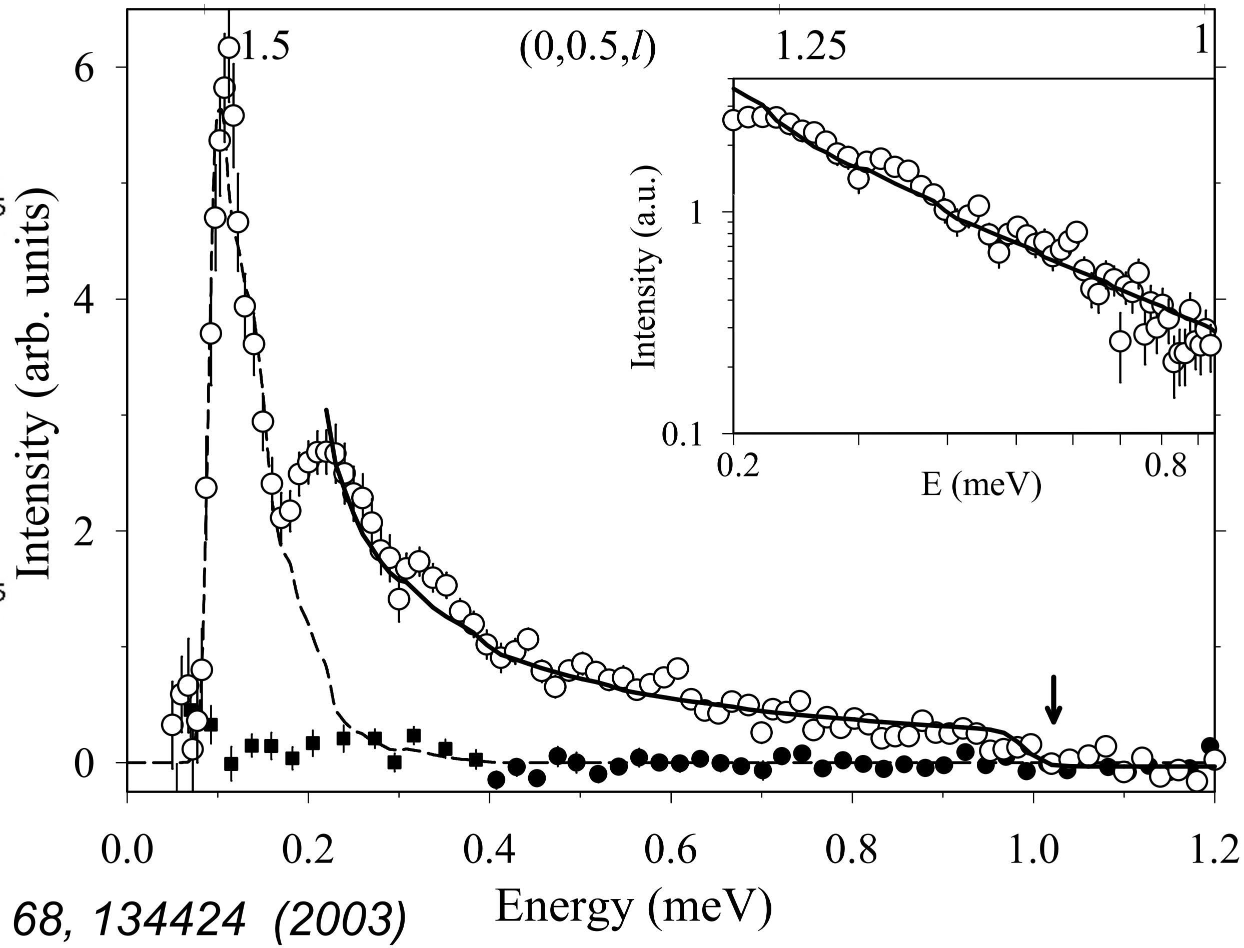
Magnetic excitations in S =1/2 triangular lattice AFM Cs_2CuCl_4



- sharp spin-wave mode only very small weight
- dominant scattering continuum with strongly-dispersive boundaries

=> Quantum fluctuations very strong, spin-wave theory inadequate

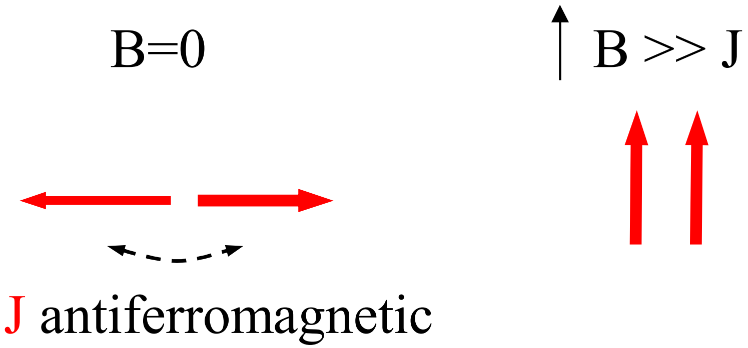


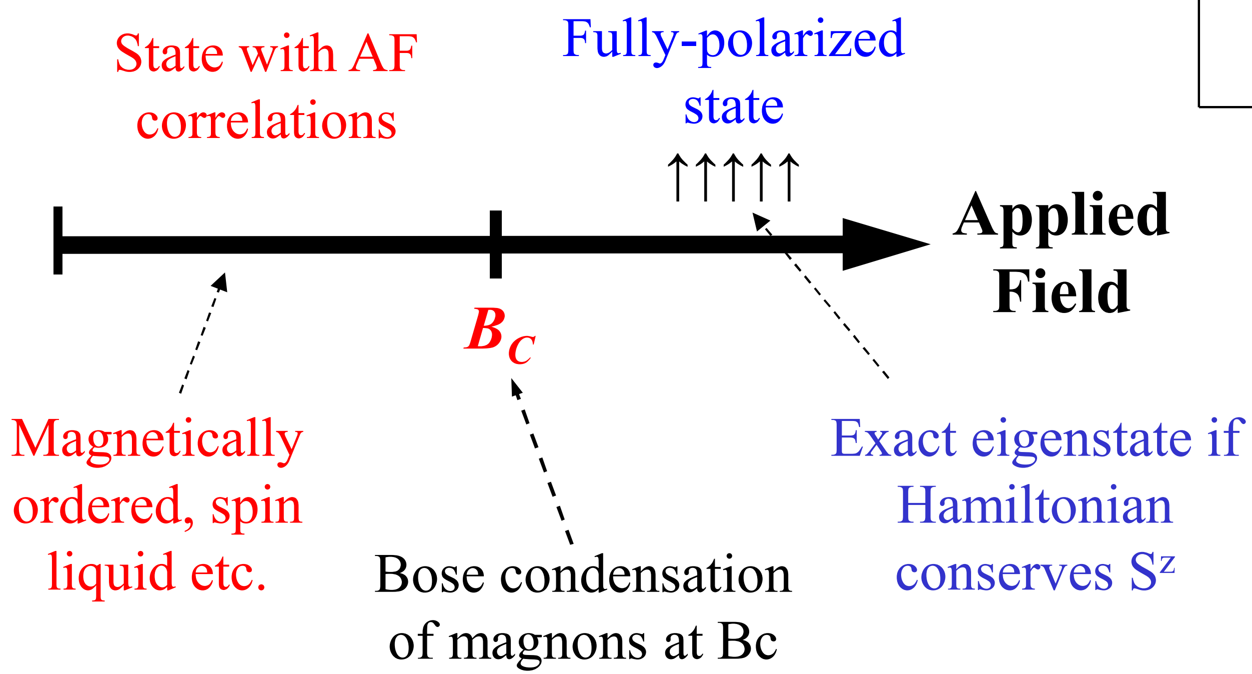


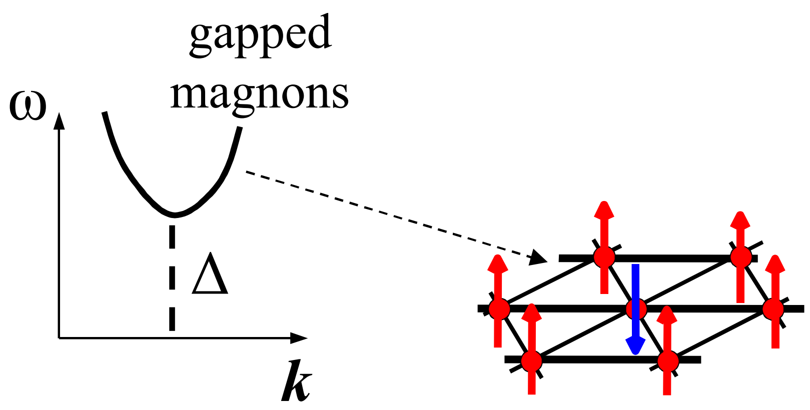
RC et al, PRB 68, 134424 (2003)

Experimental method to determine Hamiltonian via spin waves in the fully-polarized state in high field

RC et al, PRL 88,137203 (2002)





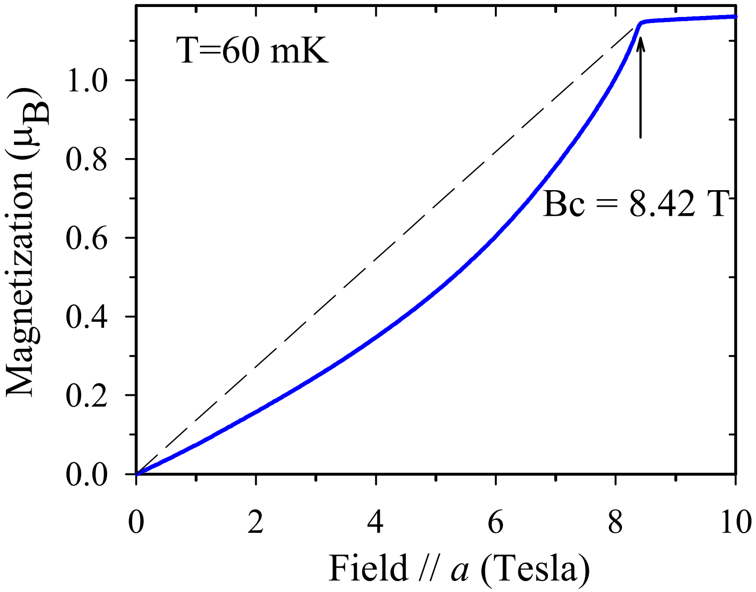


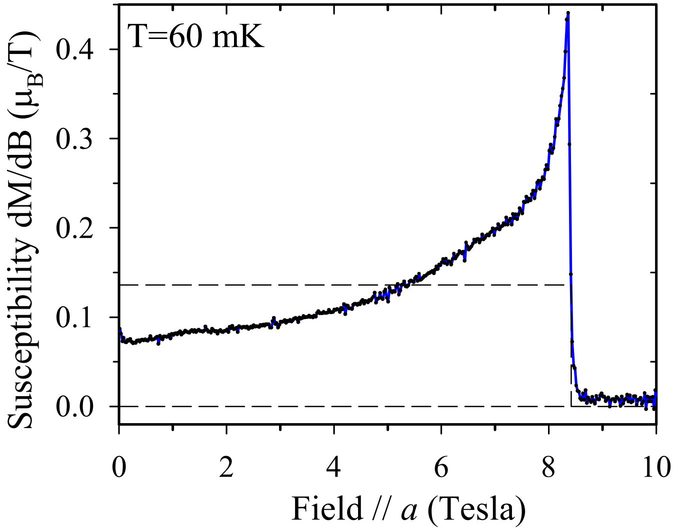
Dispersion is the Fourier transform of exchange couplings

$$\omega(\boldsymbol{k}) = J(\boldsymbol{k}) - J(0) + h$$

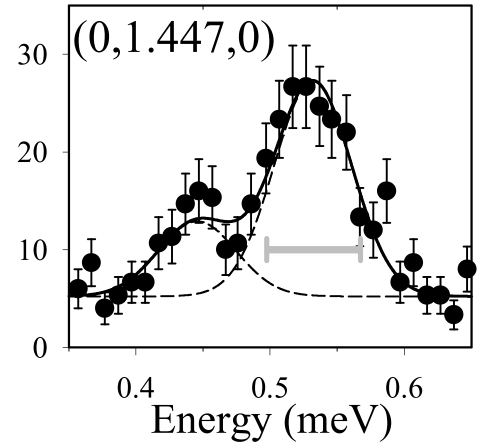
$$J(\boldsymbol{k}) = \frac{1}{2}\sum_{\mathbf{d}} J_{\mathbf{d}} \exp(i\, \boldsymbol{k} \cdot \delta)$$

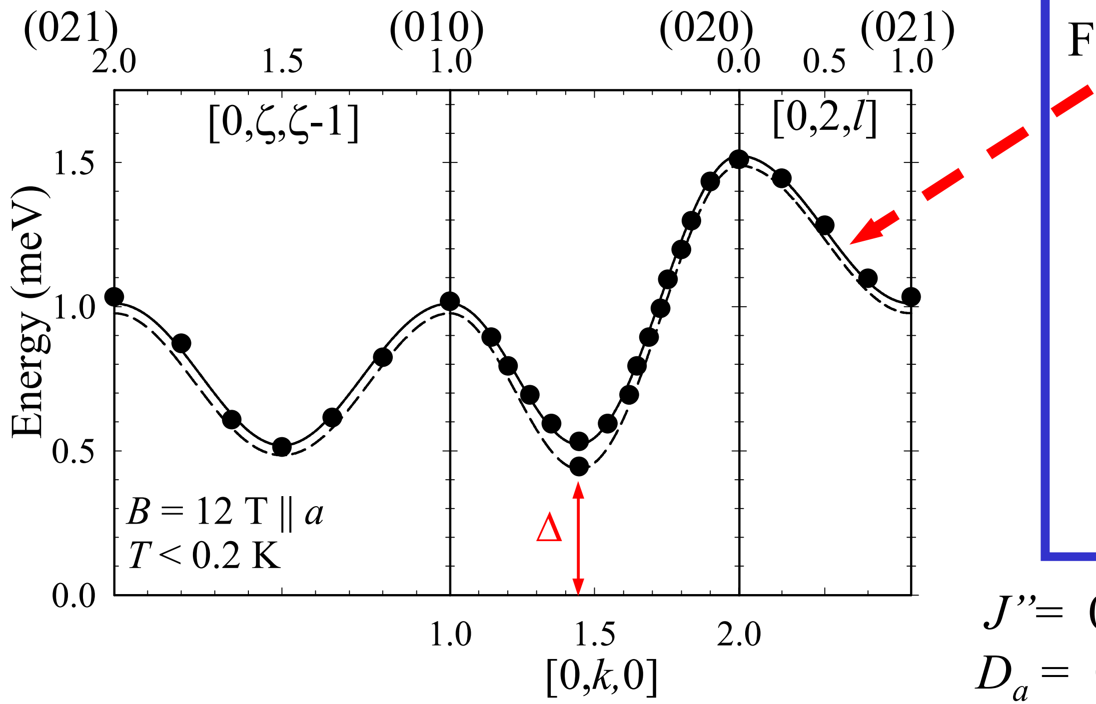
Excitations in the saturated ferromagnetic phase at B=12 T





Lineshapes show sharp magnon excitations





Fourier transform of couplings $J(\boldsymbol{q})$

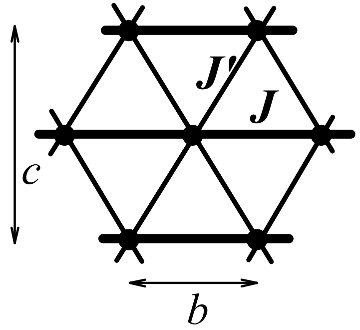
J=0.374(5) meV

J'=0.128(5) meV

$$\frac{J'}{J} \sim \frac{1}{3}$$

$\Downarrow$

2D Hamiltonian

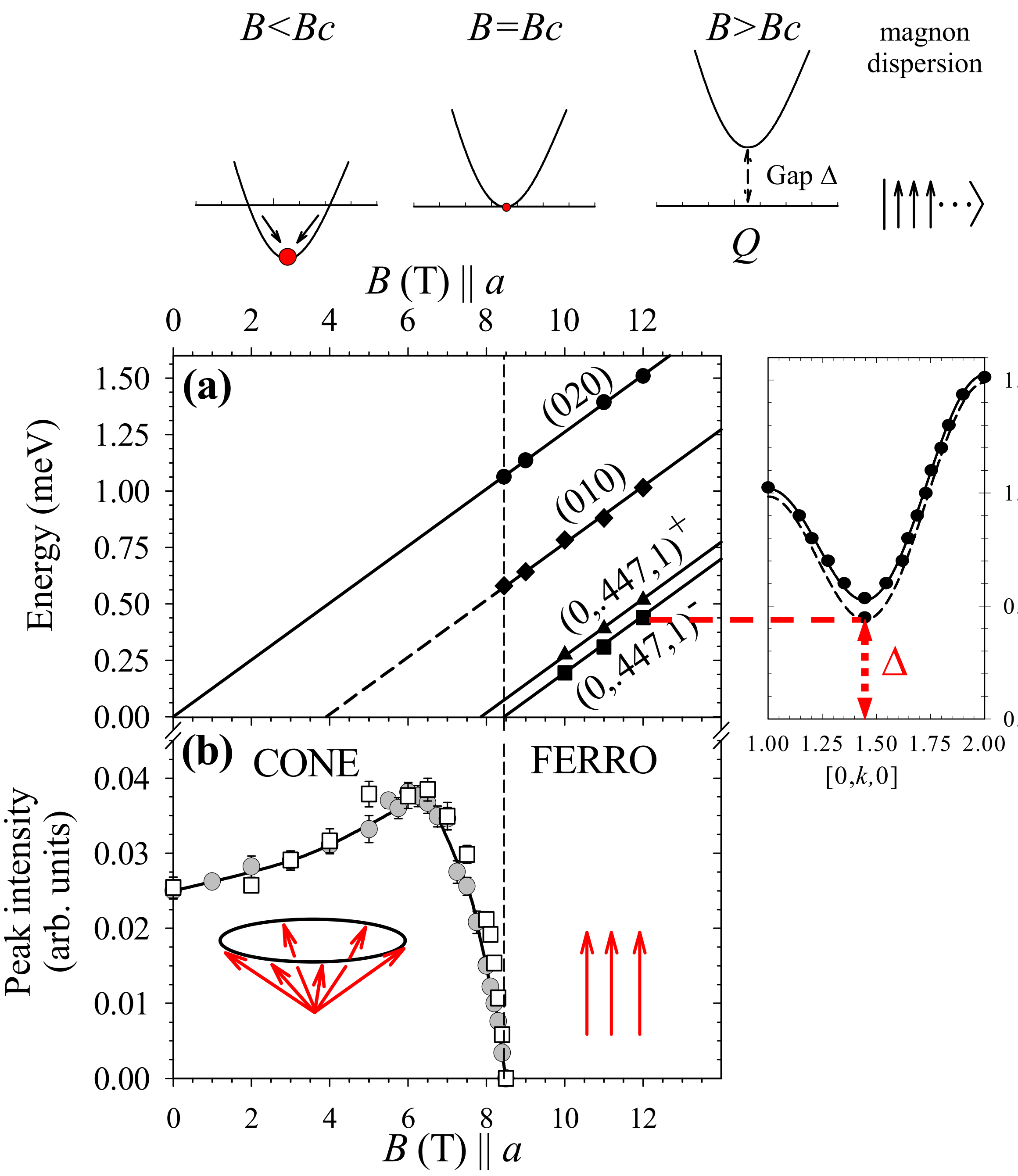


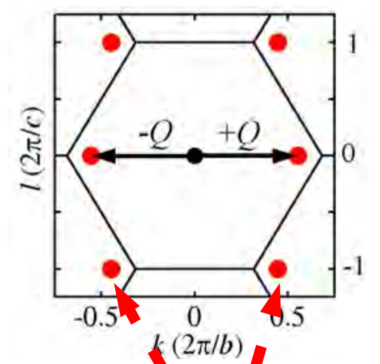
J''= 0.017(2) meV — interlayer coupling

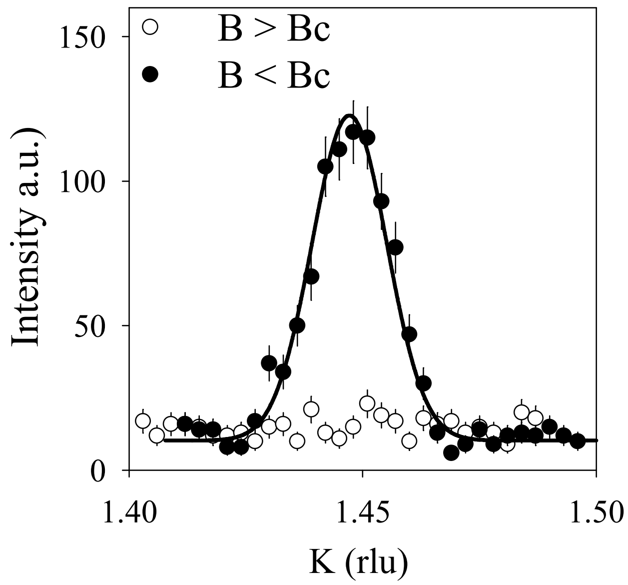
D_a = 0.020(0) meV — DM anisotropy ⊥ bc plane

RC et al, PRL 88,137203 (2002)

Magnon condensation below critical field induces transverse order

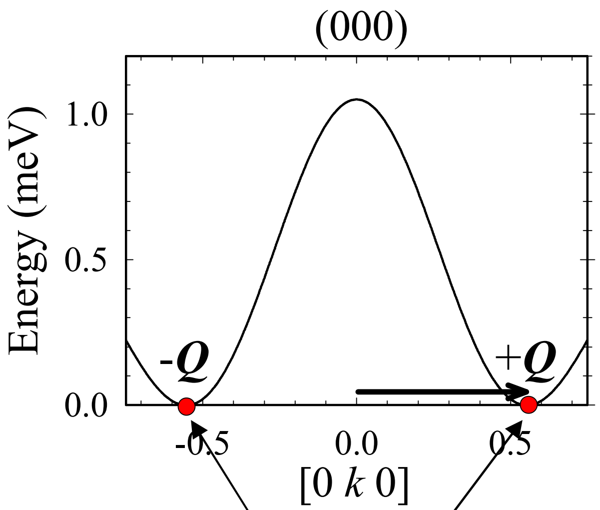




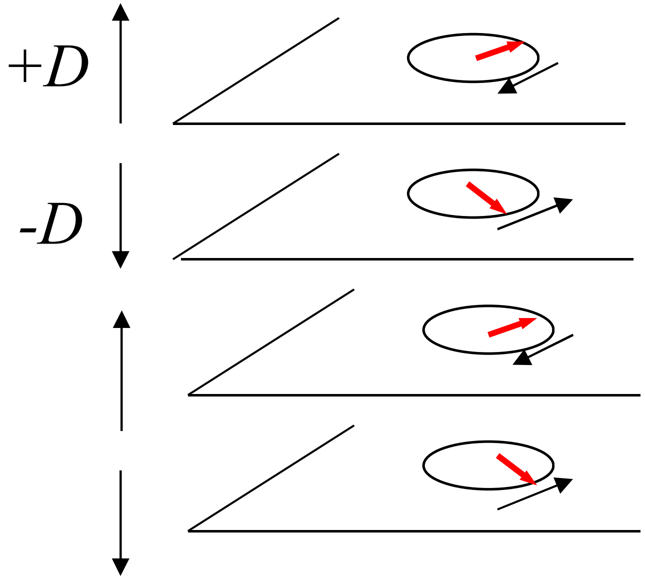


RC et al, PRL 88,137203 (2002)

Link magnetic order with magnon wavefunctions



2 condensates at +Q and -Q

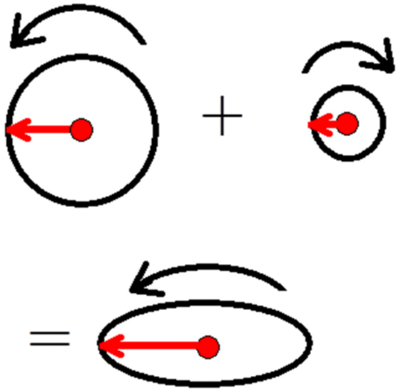


Alternating layers counter-rotate

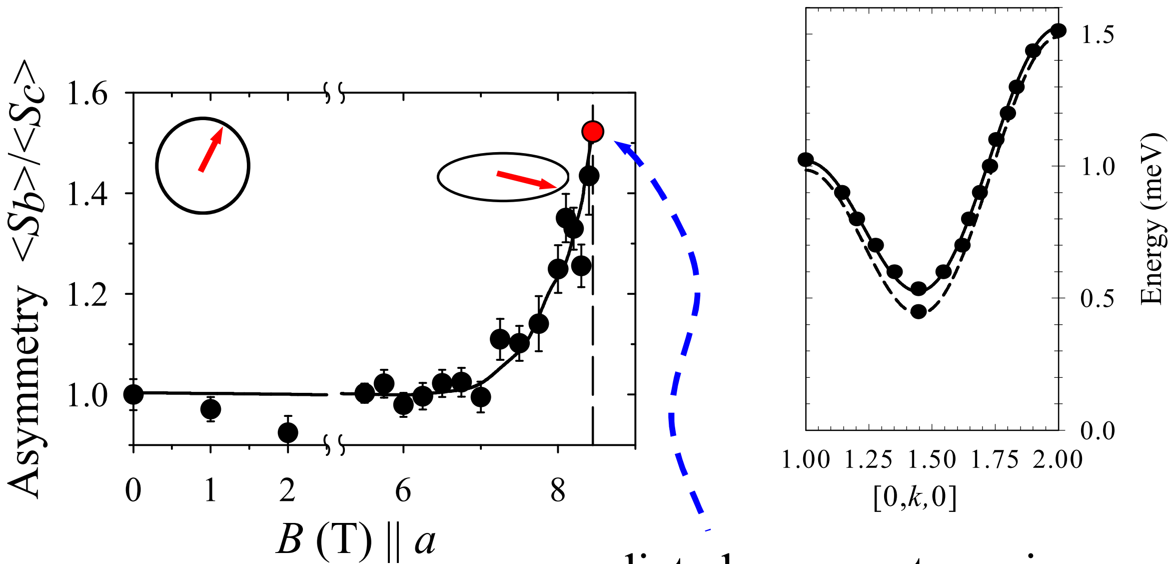
Sense selected by DM couplings

$\boldsymbol{D}\cdot(\boldsymbol{S}_i \times \boldsymbol{S}_j)$

The two condensates interact via the inter-layer couplings *J''*



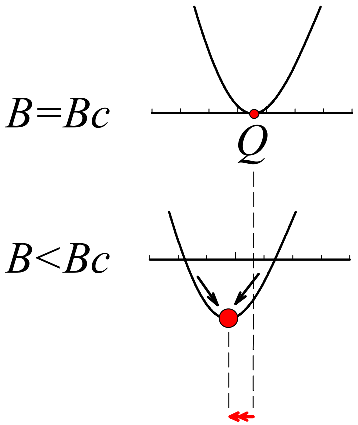
Asymmetric shape

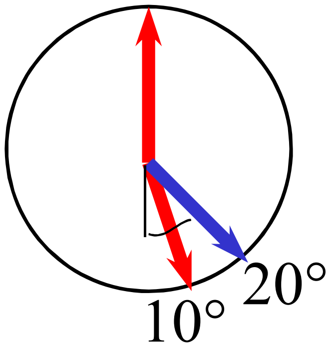


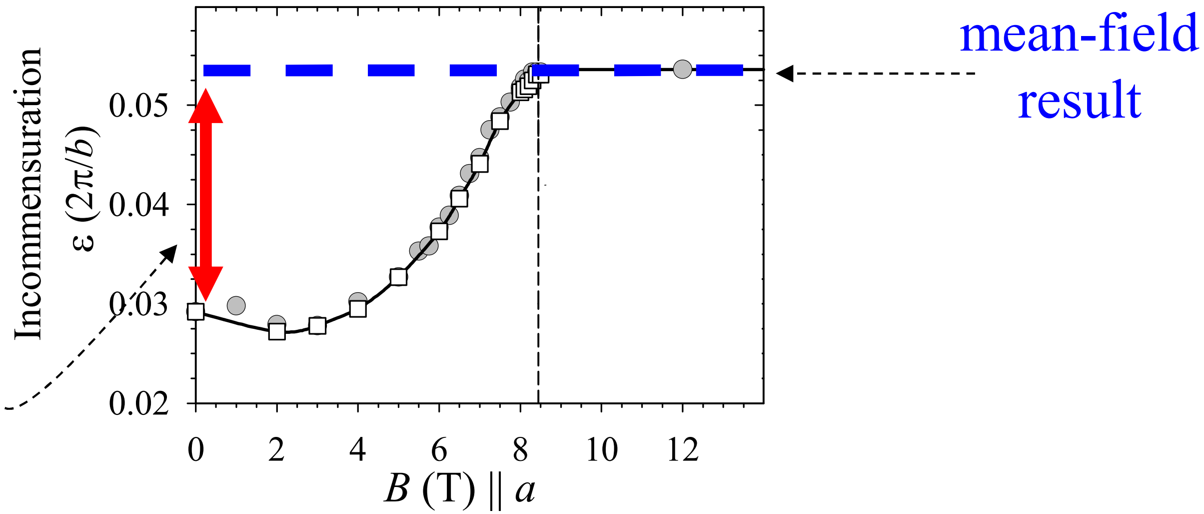
predicted asymmetry using magnon wavefunctions

RC et al, PRL 88,137203 (2002)

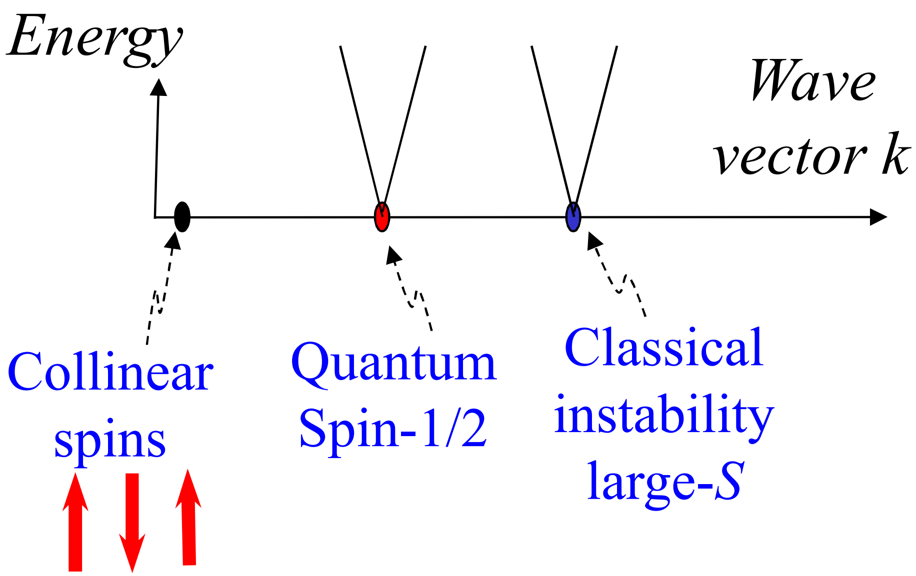
Quantum renormalization of incommensurate ordering wavevector





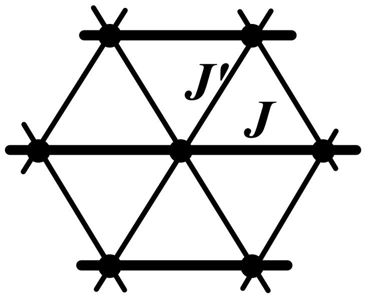


Strong quantum renormalization
$\varepsilon_0/\varepsilon_{cl}=0.56$

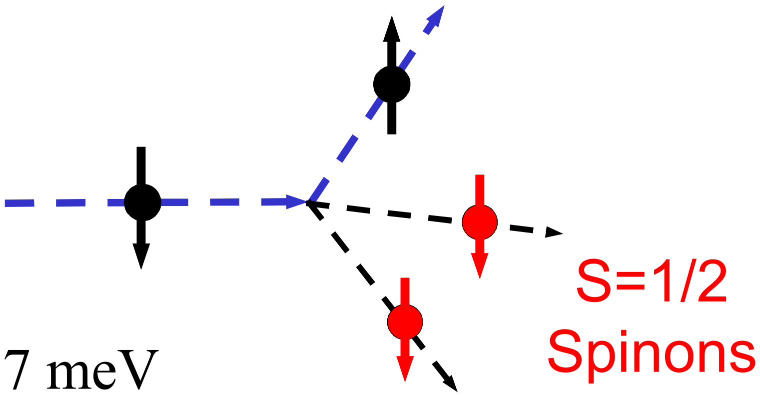


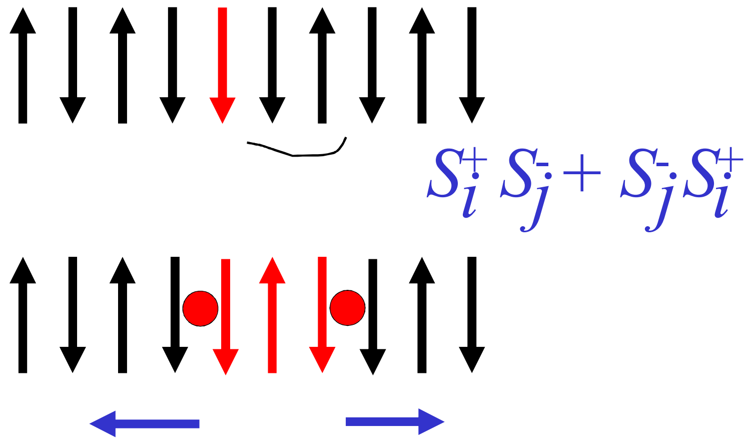
RC et al, PRL 88,137203 (2002)

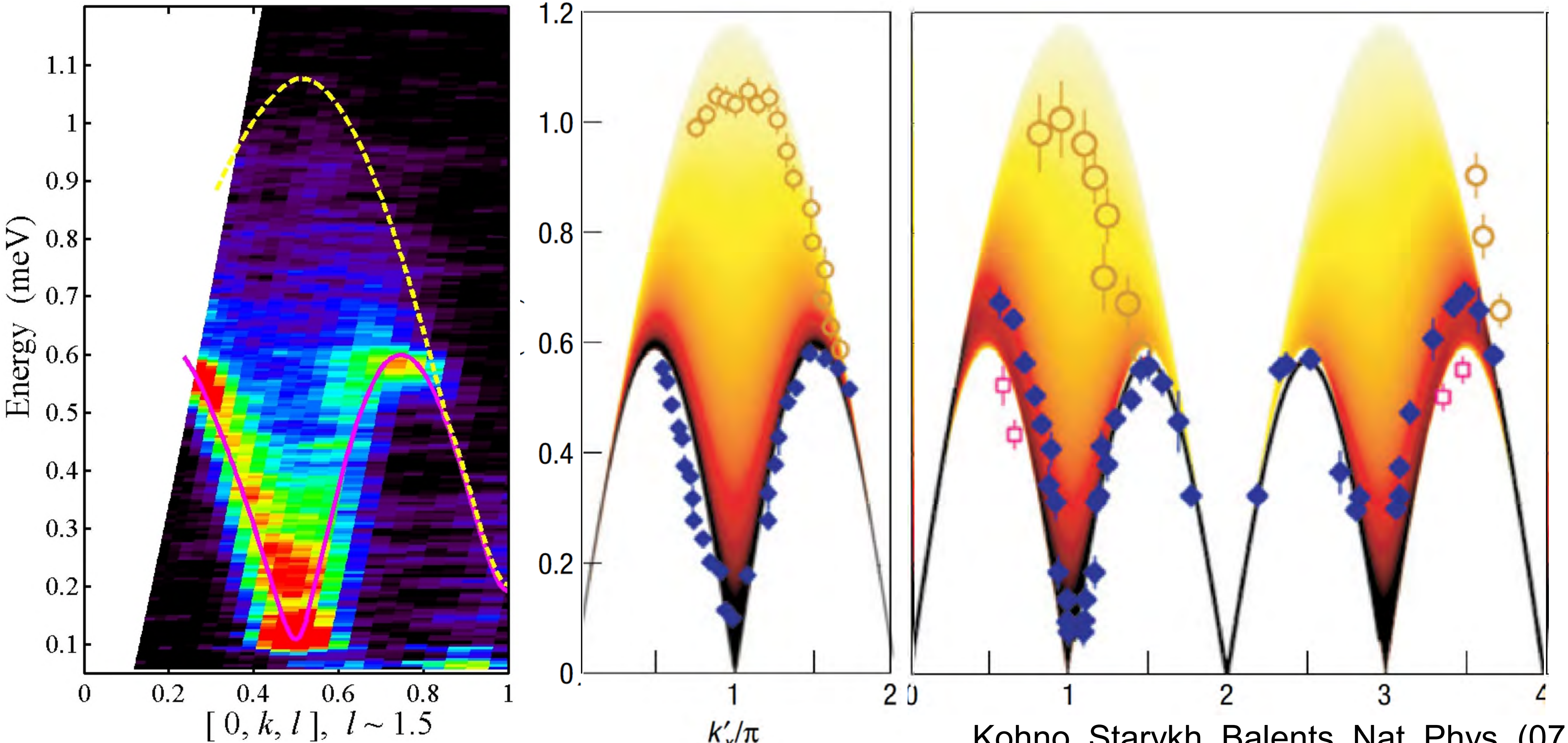
Magnetic excitations at zero field: spin-waves fractionalize into pairs of S=1/2 spinons



$J'/J \sim 1/3$, J=0.37 meV

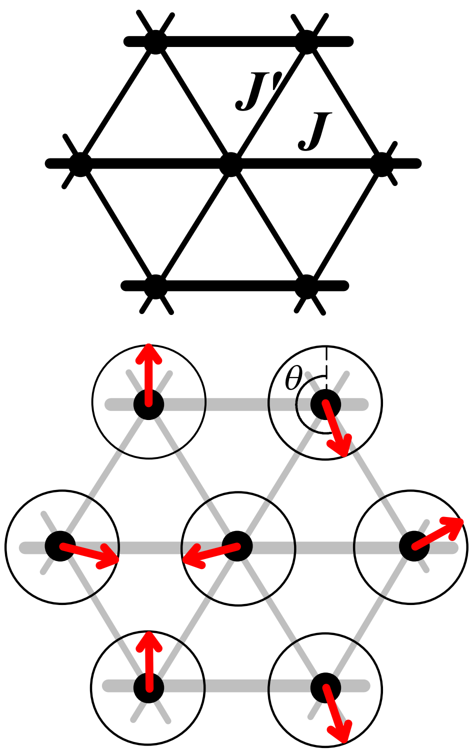


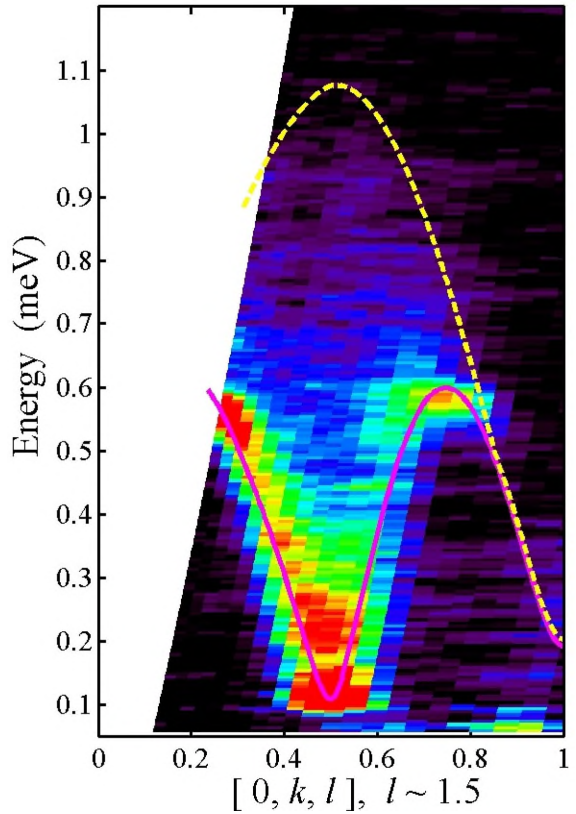


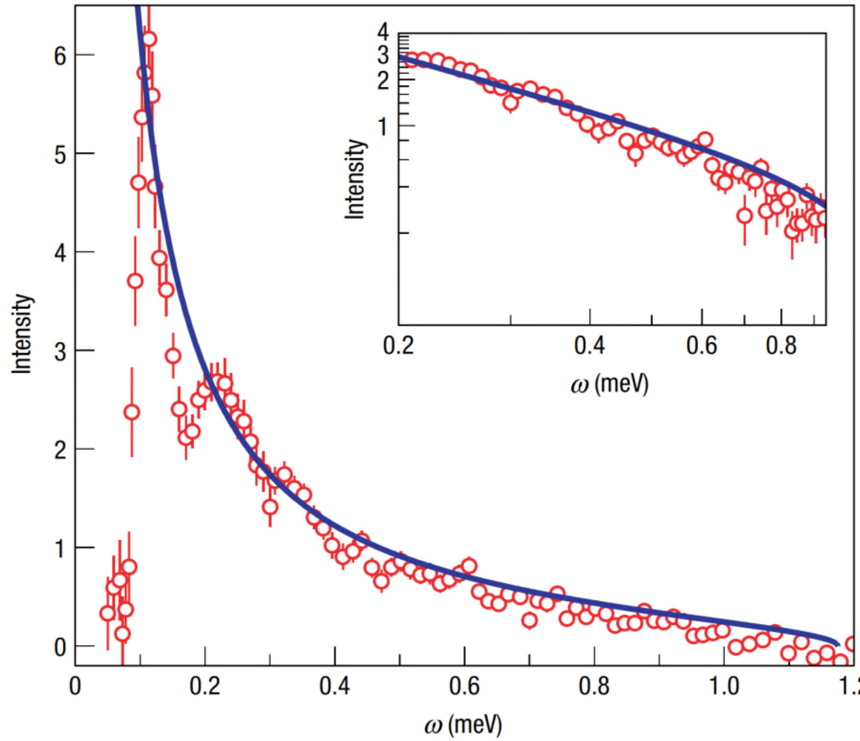


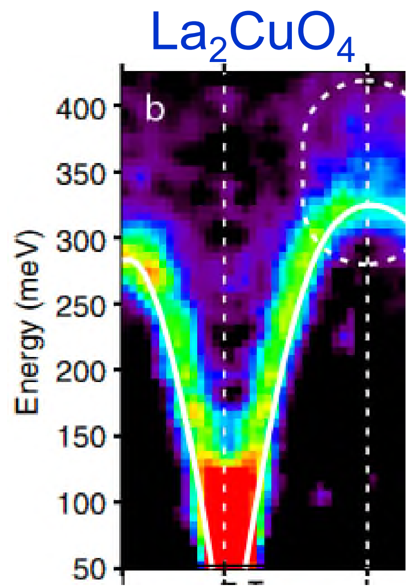
Kohno, Starykh, Balents, Nat. Phys. (07)

Summary on anisotropic triangular AFM Cs_2CuCl_4









- anisotropic triangular lattice S=1/2 AFM Cs_2CuCl_4 has spiral order coexisting with **strong quantum fluctuations**
- renormalization of *Q*-vector and zone-boundary energy measured by quenching quantum fluctuations via field and revealing "classical" behaviour
- dominant continuum scattering (spin-waves fractionalize into pairs of spinons)

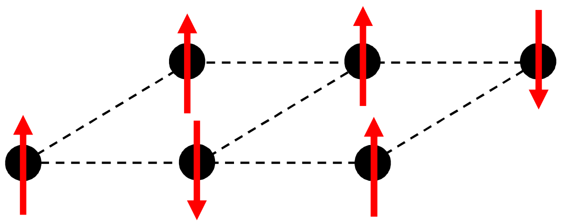
Headings, Hayden, RC …, PRL (2010)

Ising magnets and phase transitions

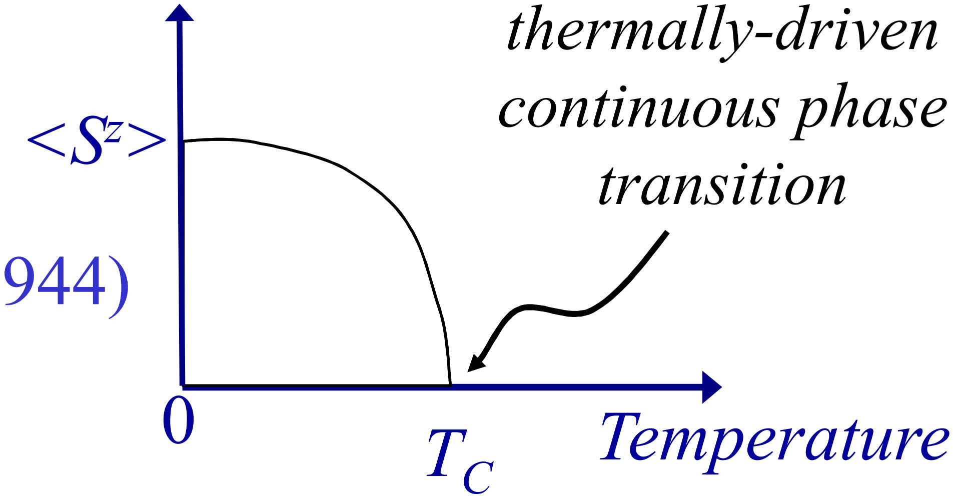
- classical Ising model

$$H = -\sum_{i,j} J\ S_i^z S_j^z$$

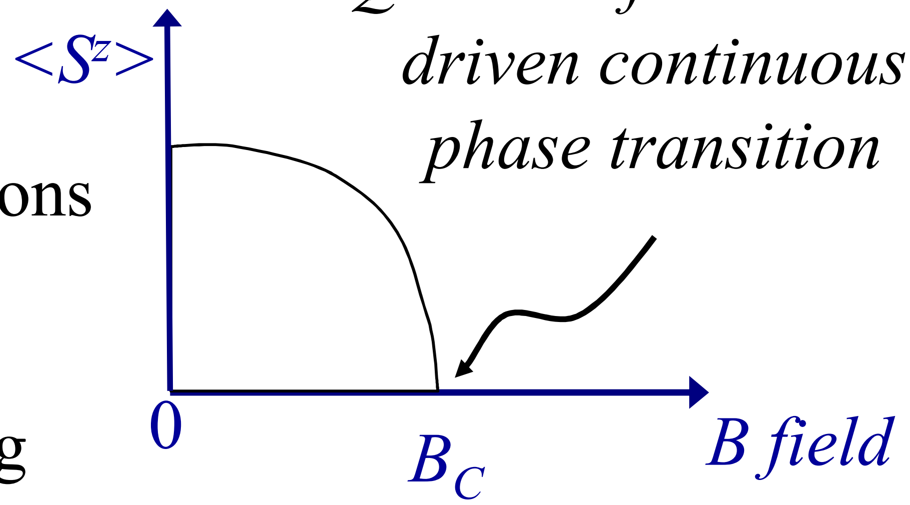
- 2D model Onsager exact solution (1944)



Classical thermally-driven continuous phase transition

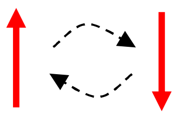


Quantum fluctuations driven continuous phase transition



add transverse field → quantum fluctuations

- $-B\,S^x$
- $-B\,(S^+ + S^-)/2$

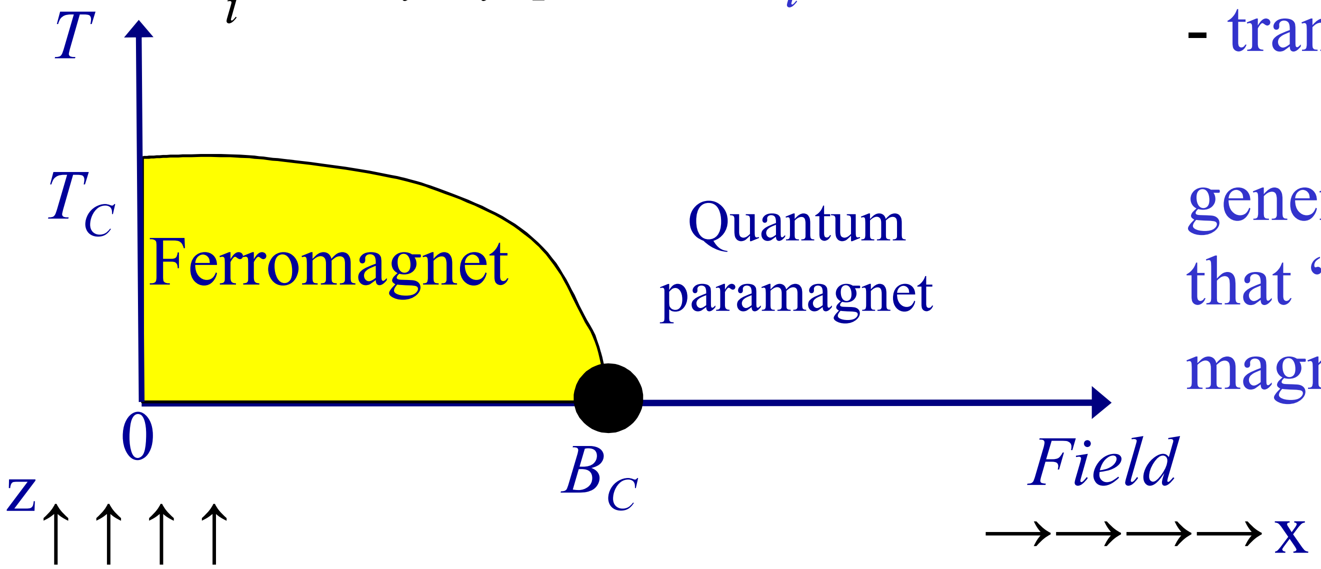


quantum tunneling

T=0 "quantum melting of order"

An Ising ferromagnet in transverse field

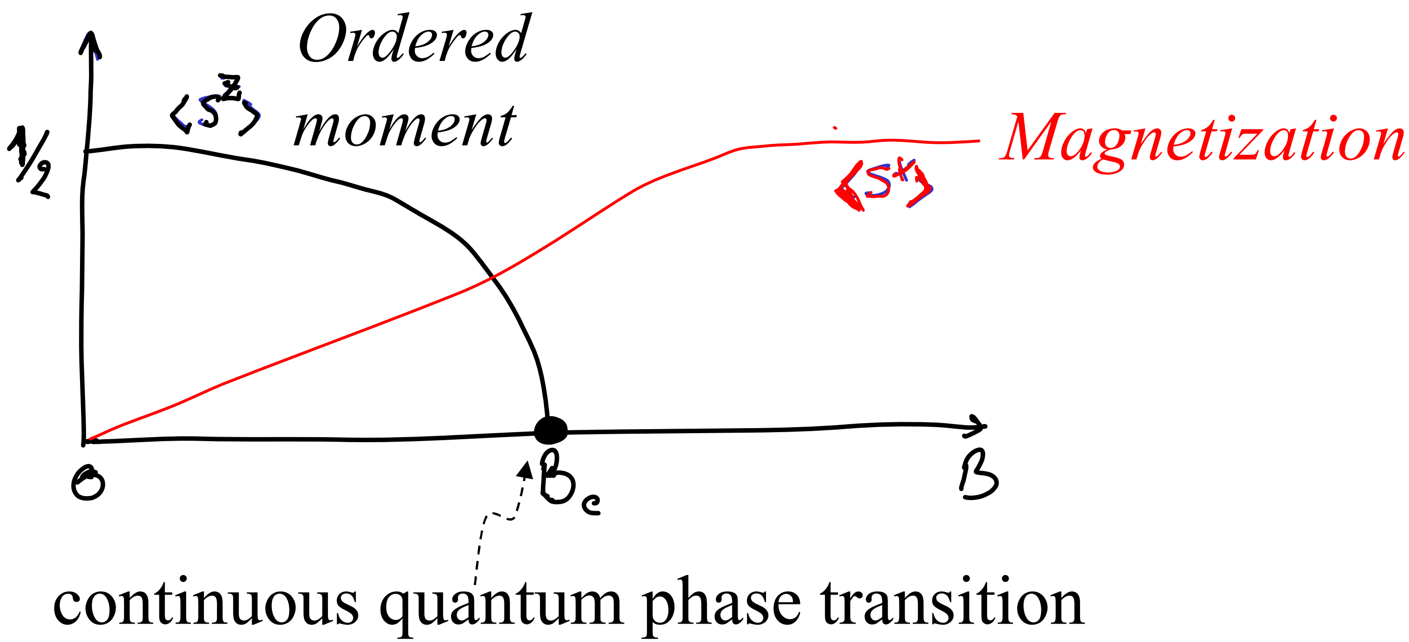
$$H = -\sum_i J\, S_i^z S_{i+1}^z \;\; - B\, S_i^x$$



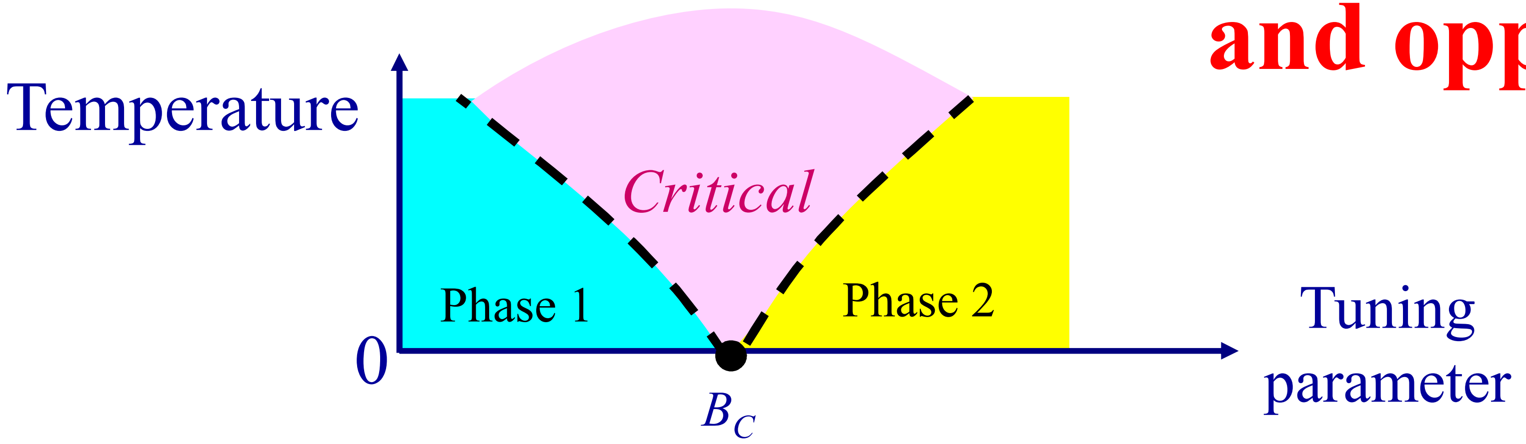
- transverse field

$- B\, S^x = - B\, (S^+ + S^-) / 2$

generates quantum fluctuations that "melt" the spontaneous magnetic order at $B_C \sim J/2$



Quantum criticality: experimental challenges and opportunities



- -what is **microscopic mechanism of transition,** can one observe the quantum fluctuations that drive transition ?

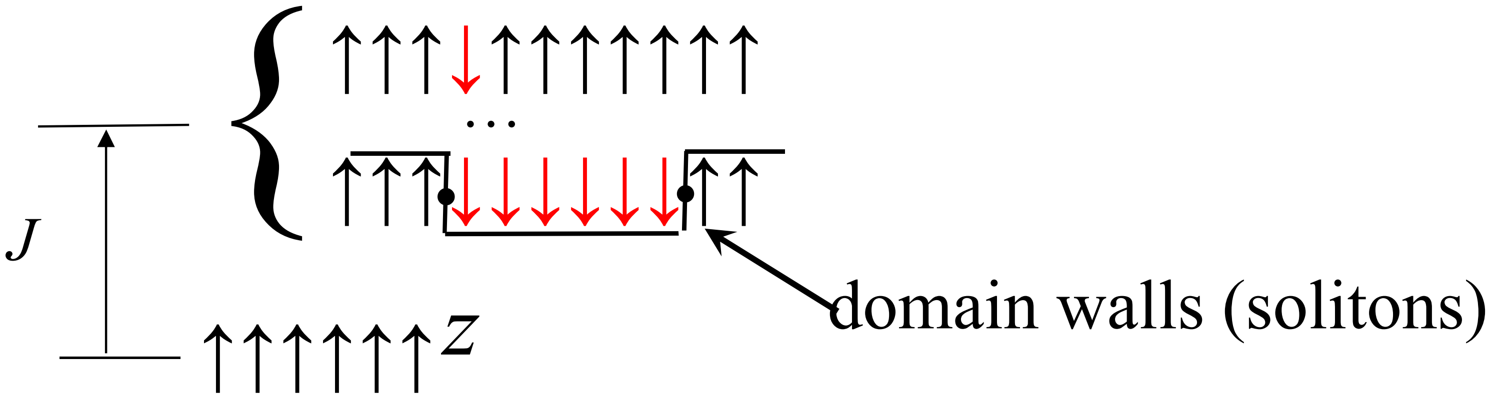
- - how **quasiparticles evolve near critical point** ?

- - what are the **fundamental symmetries** that govern physics of QCP ?

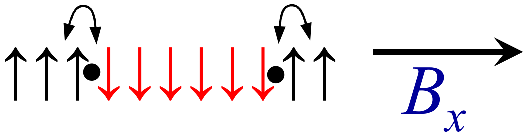
- what are **finite-T properties** (interplay of thermal and quantum fluctuations, under what conditions universal scaling ?

1D Ising chain in transverse field

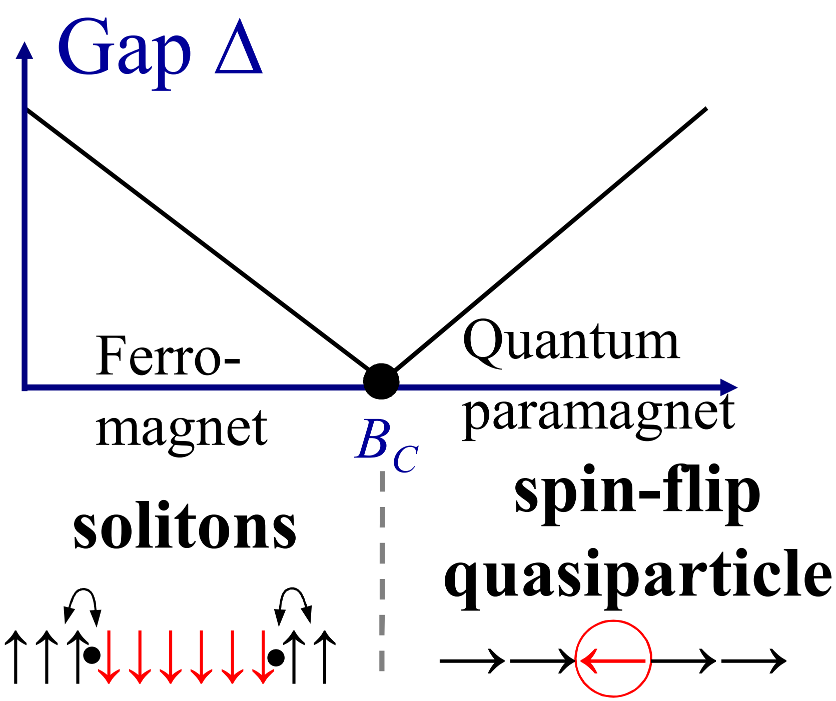
$H = -\sum_i J\, S_i^z S_{i+1}^z$ 2 ground states: ↑↑↑↑↑↑ or ↓↓↓↓↓↓



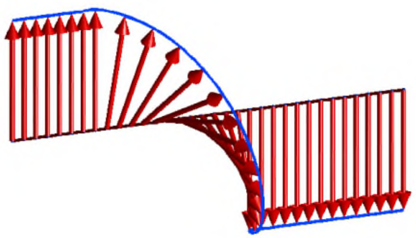
- transverse field $B\, S^x \sim -B\,(S^+ + S^-)$
flips spins



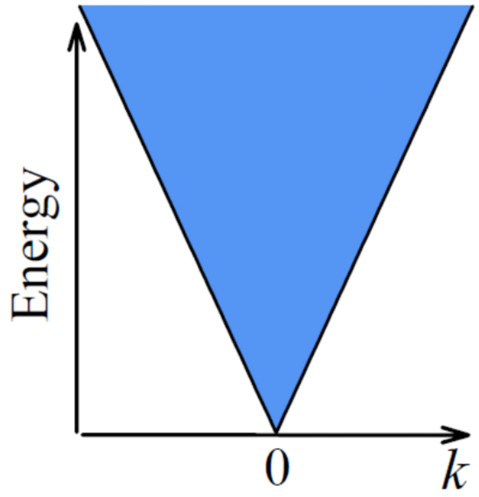
=> **propagating solitons**
(Jordan Wigner fermions)



classical soliton
(large-*S* limit)



Ising chain at criticality



gapless linear (Dirac) spectrum

$\omega = c|k|$

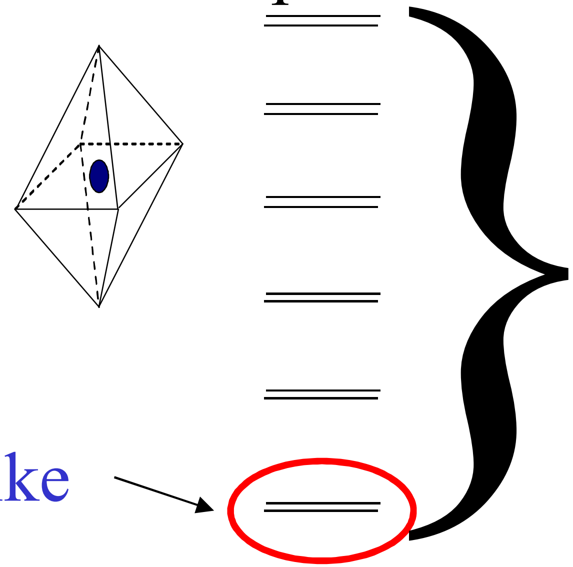
for critical solitons

- ***ω/T scaling expected, special "conformal" symmetry***
- ***different universality class from Luttinger liquids (1D Heisenberg and XY AFM chains)***

Experimental requirements

1) good 1D character to see solitons
2) low-exchange $J \sim 1$ meV to access critical field $BC \sim J/2 < 10$ T
3) strong uniaxial anisotropy (Ising character) but not perfect to still have transverse g-factor

λ L•S
Strong Crystal field + Spin Orbit



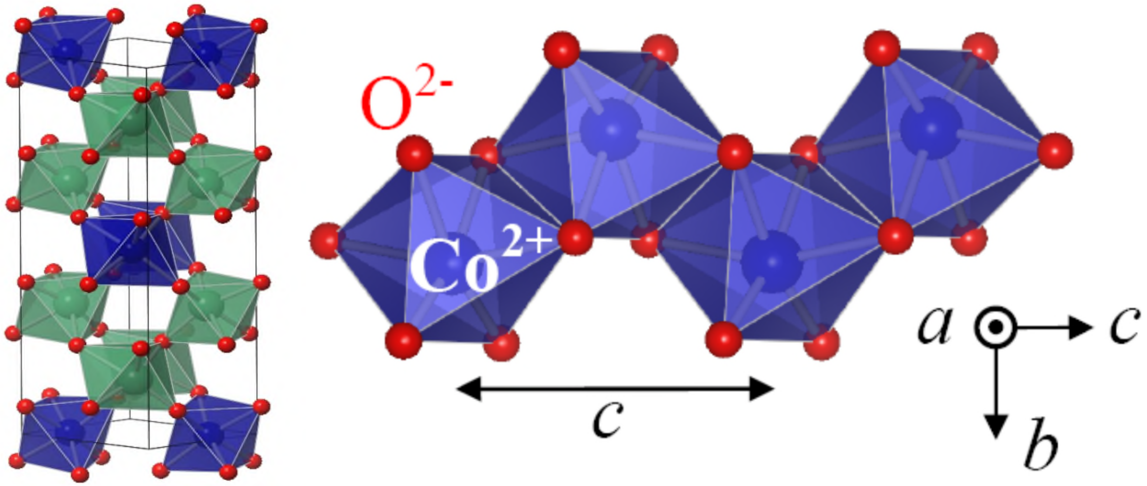
- best Ising magnets are based on Co^{2+} $3d^7$

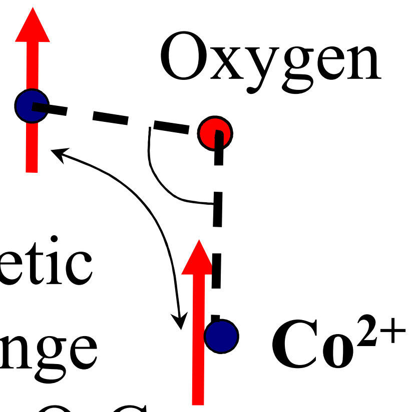
lowest Kramers doublet effective spin-1/2 Ising-like

2D Ising AF K2CoF4 (*Birgeneau '73, Cowley '84*)
1D Ising AF CsCoCl3 (*Goff '95*) *also* CsCoBr3 (*Nagler*)
$J \sim 12$ meV $BC > 50$ T not accessible

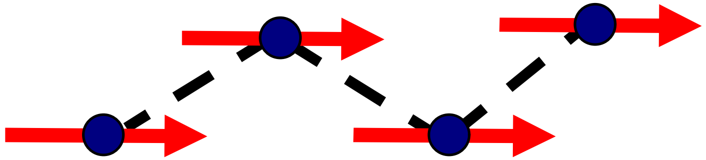
Quasi-1D Ising ferromagnet $CoNb_2O_6$

zig-zag Co^{2+} spin chain along c



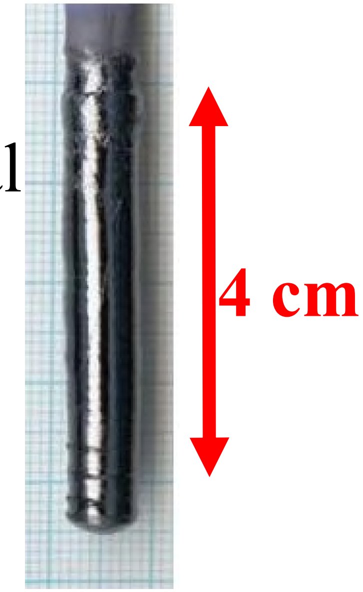


Ferromagnetic superexchange
~ 90° bond Co-O-Co
~ 20K ~ 2meV



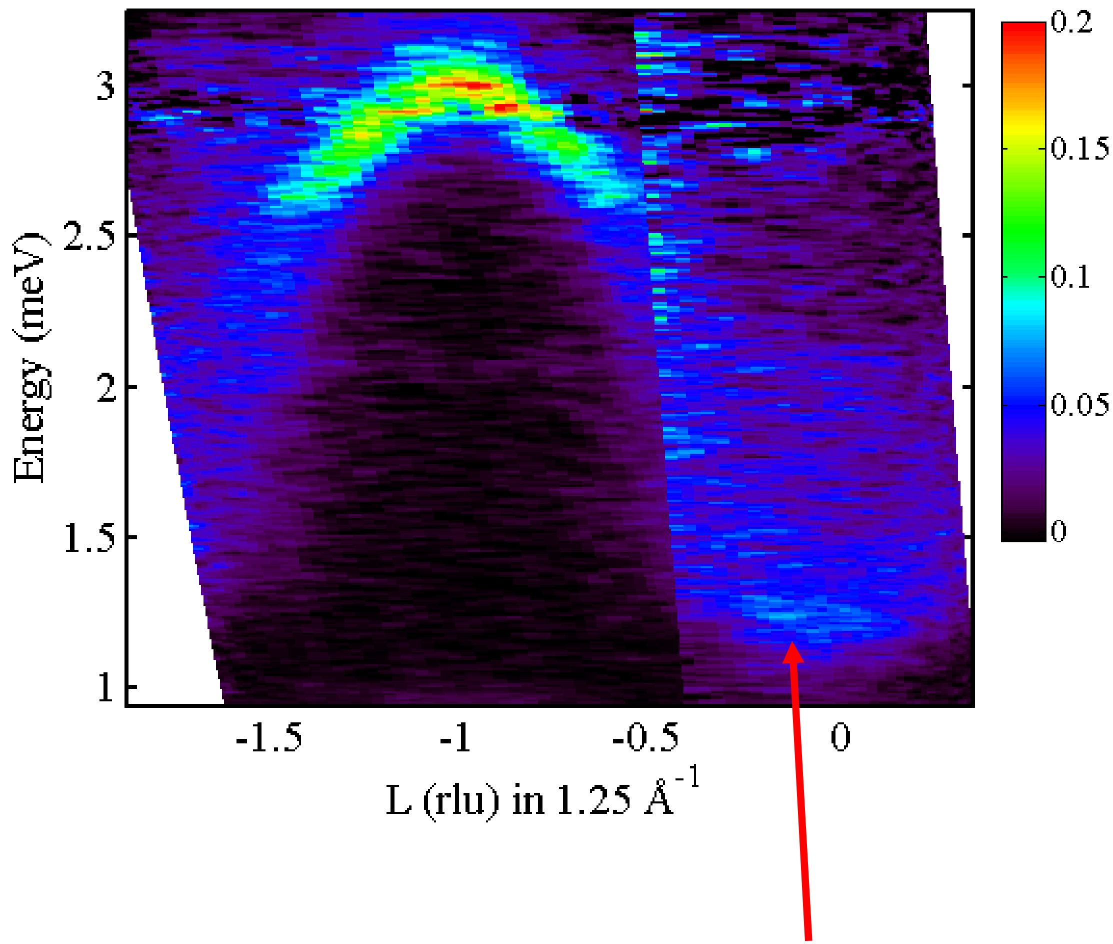
Ferromagnetic order along chain
Strong easy-axis (Ising) in *ac* plane

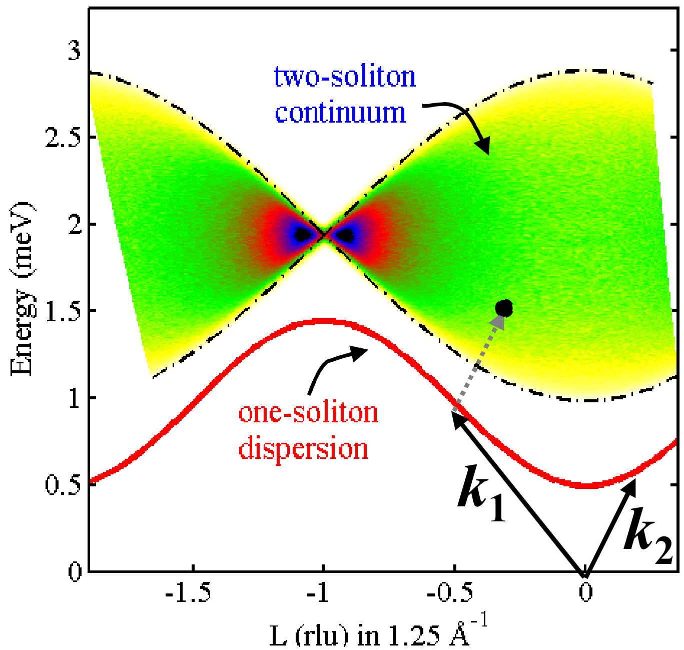
Single crystal of $CoNb_2O_6$ (Oxford image furnace)

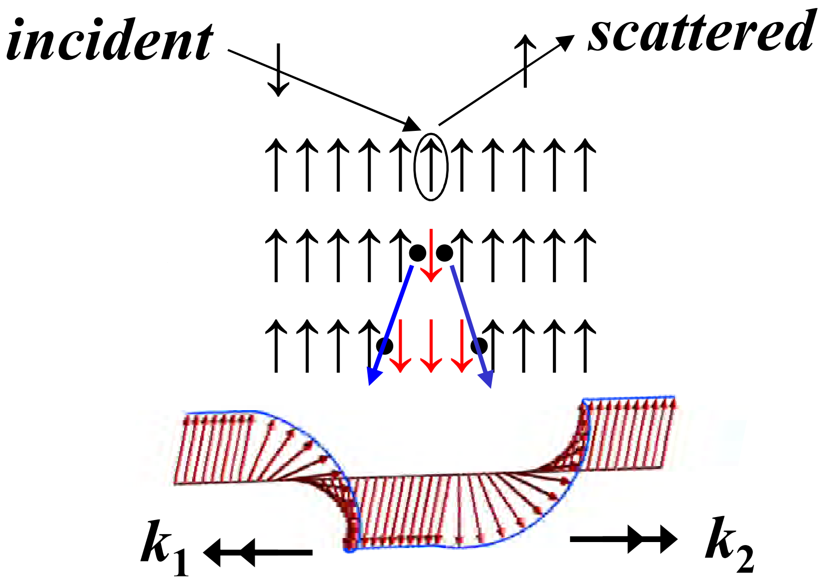


Magnetic excitations in 1D phase seen by neutron scattering

T= 5 K, 1D phase above T_N



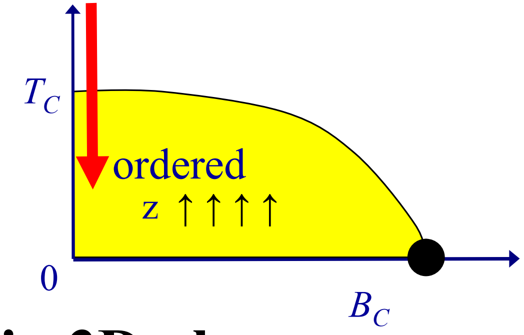




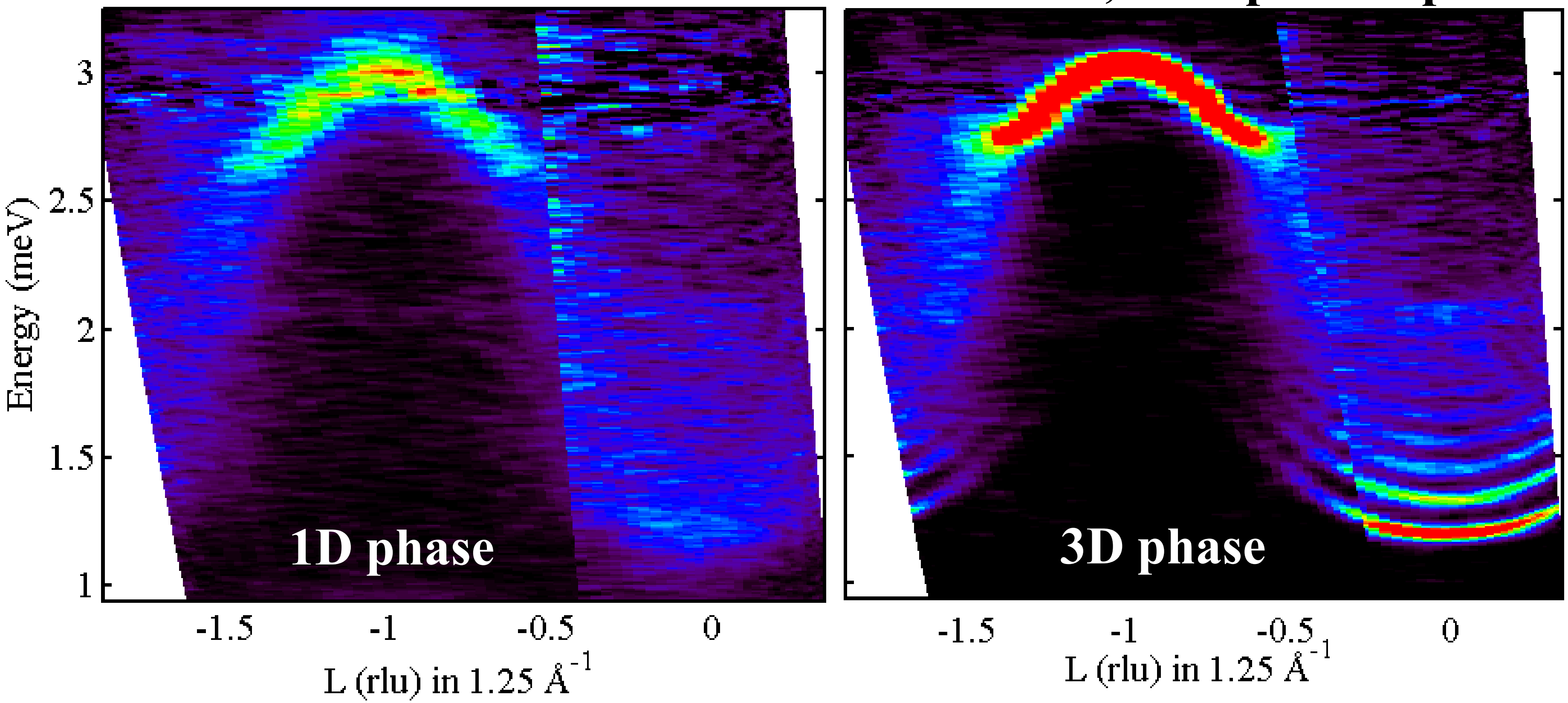
- gapped continua characteristic of 2-soliton excitations

RC et al, Science 327, 177 (2010)

Magnetic excitations in zero field



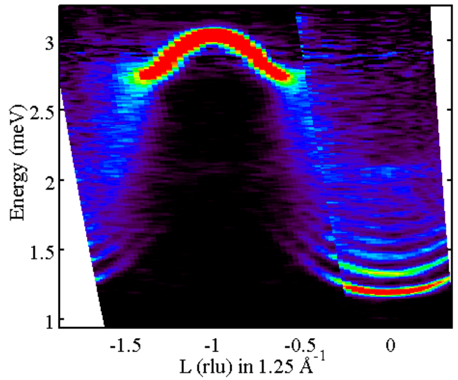
T= 5 K, 1D phase above T_N **T= 0.04 K, deep in 3D phase**



- rich structure : continuum as characteristic of 2-soliton excitations

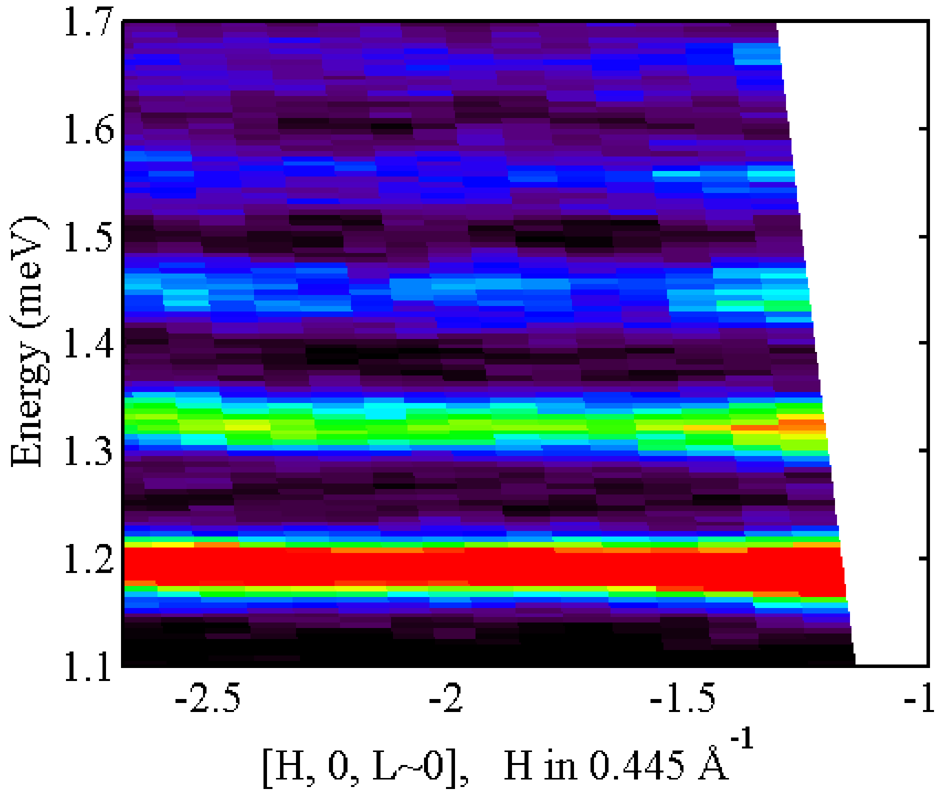
+ sharp modes (bound states)

RC et al, Science 327, 177 (2010)

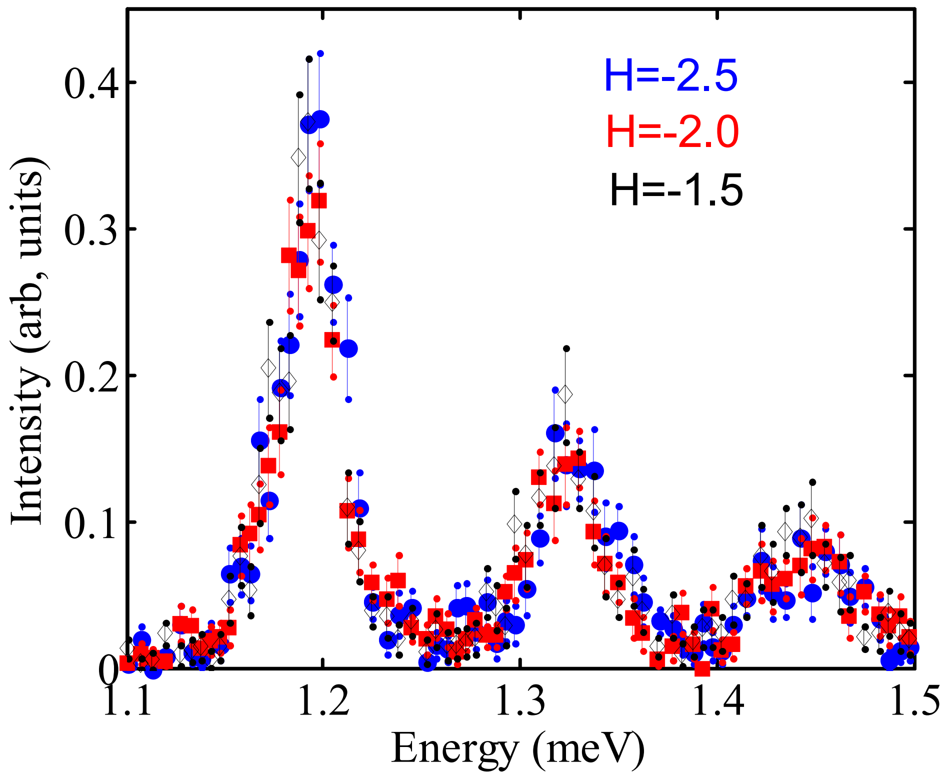


|| **chain direction**

Excitations have 1D character – no measurable dispersion ⊥ chains

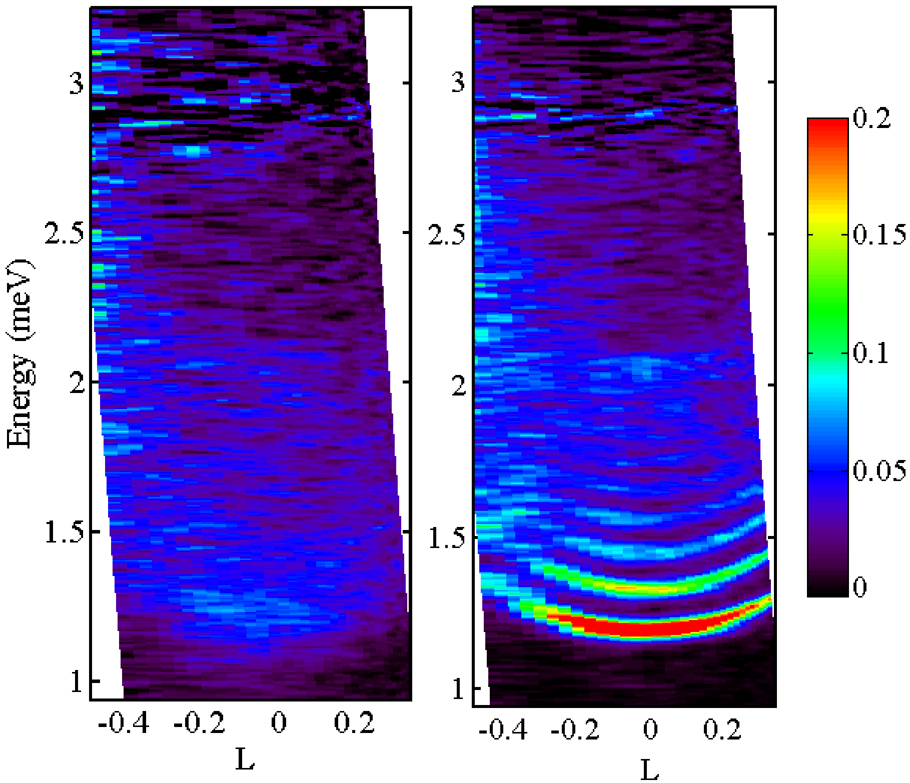


⊥ **chain direction**

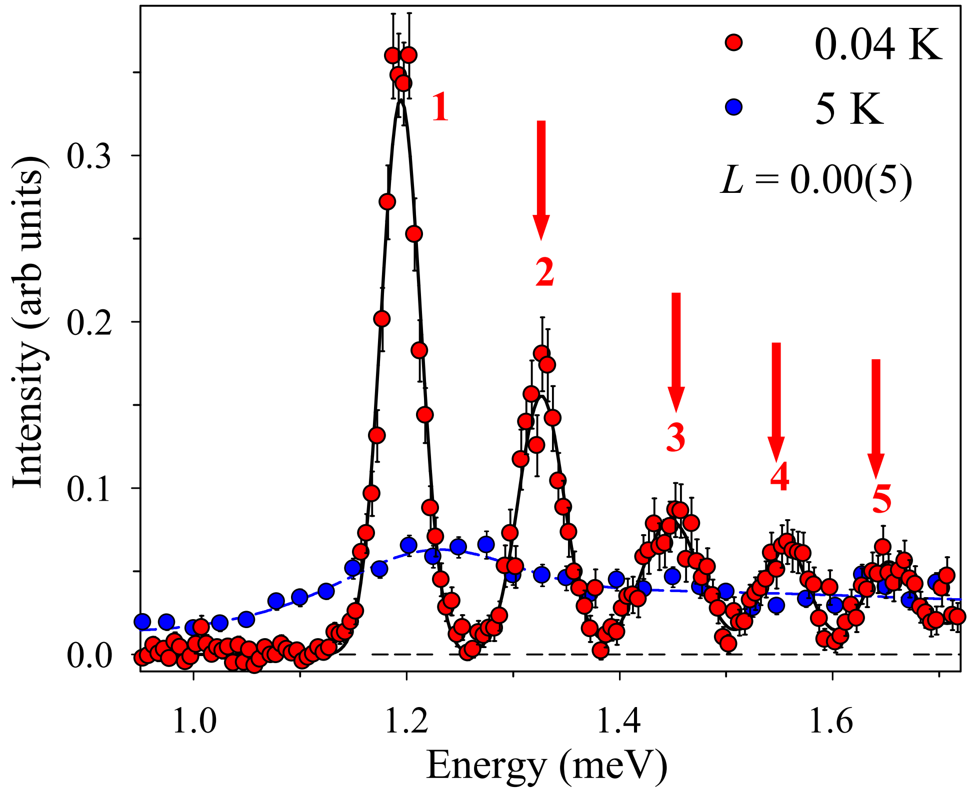


Zeeman ladder of bound states in 3D ordered phase

T = 5 K　　*T* =40 mK

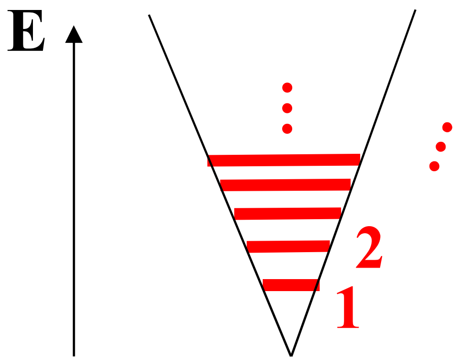


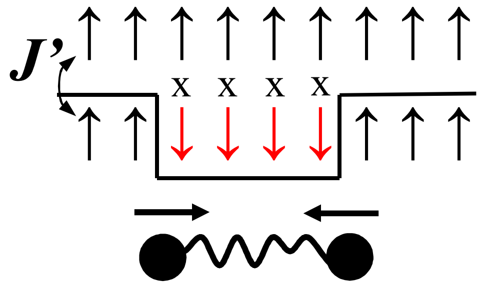
Zeeman ladder of bound states



Continuum of free 2-soliton states

Bound states in confining potential





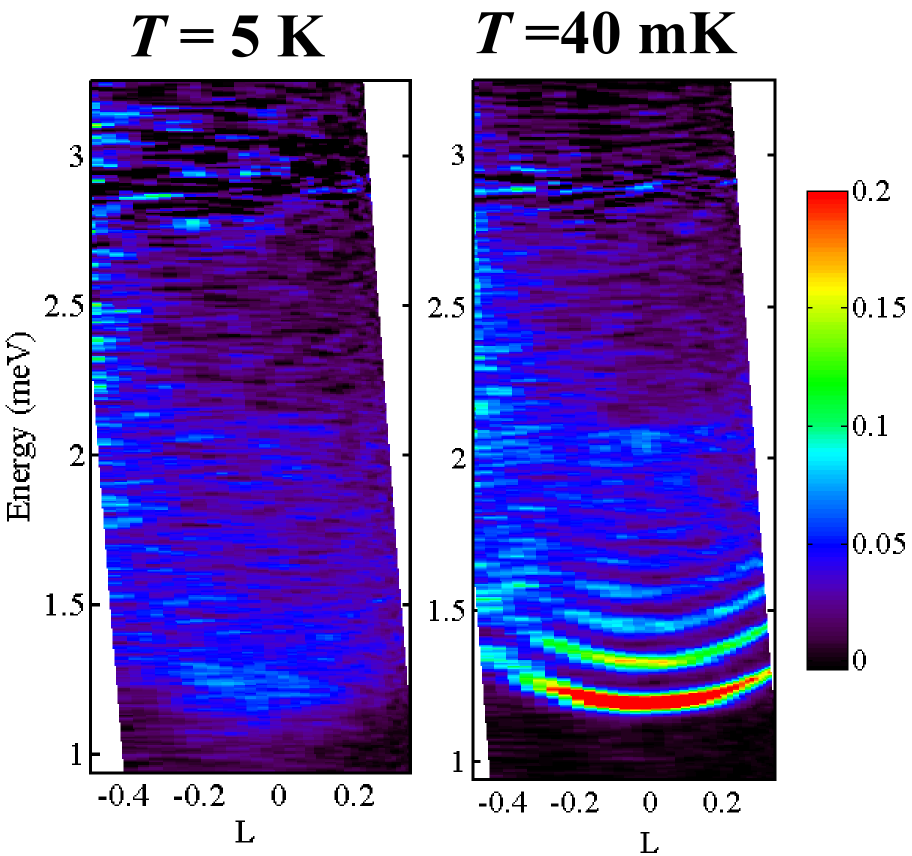
Soliton separation costs energy

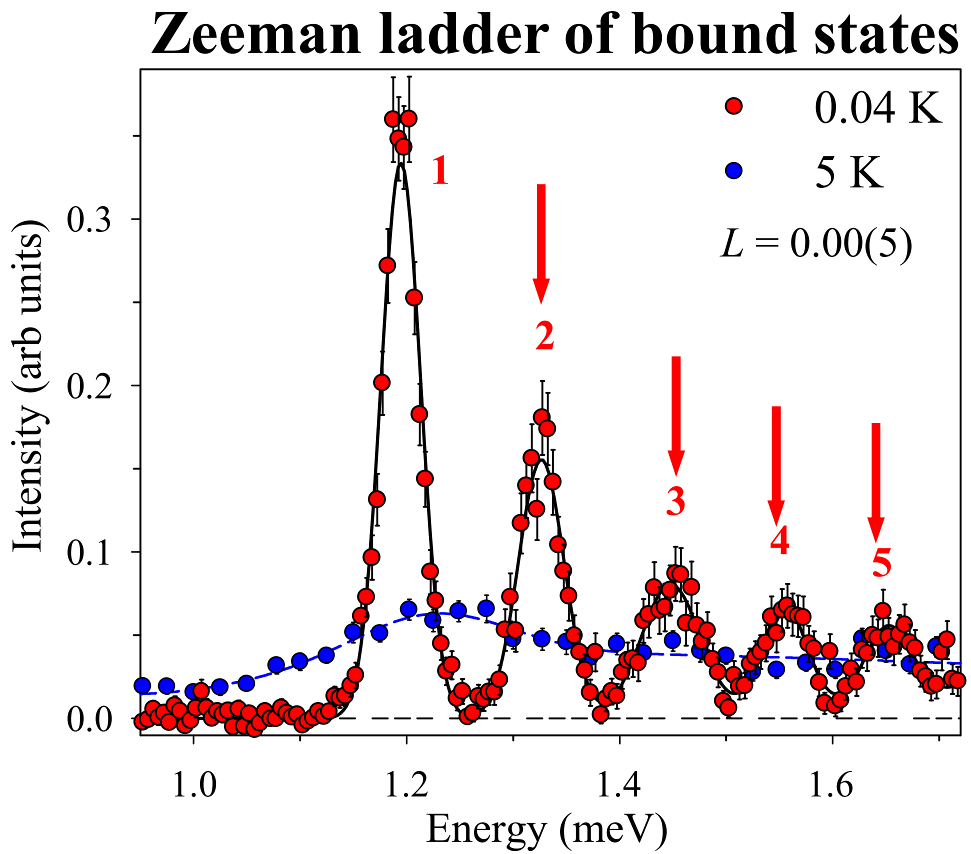
$V(x) = \lambda\, x$

$\lambda \sim J'\langle S^z\rangle$

Longitudinal mean-field $-hS^z$, $\lambda = 2\, h\langle S^z\rangle$

Zeeman ladder of bound states in 3D ordered phase





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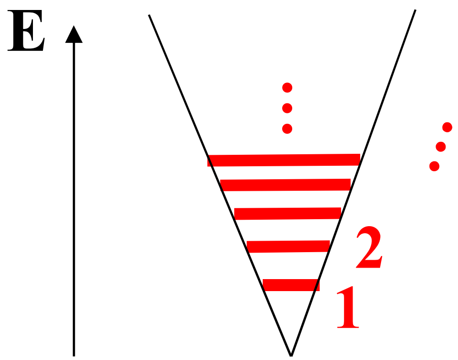

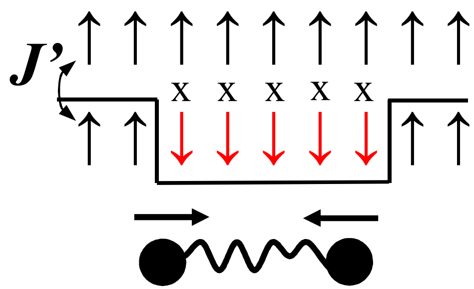

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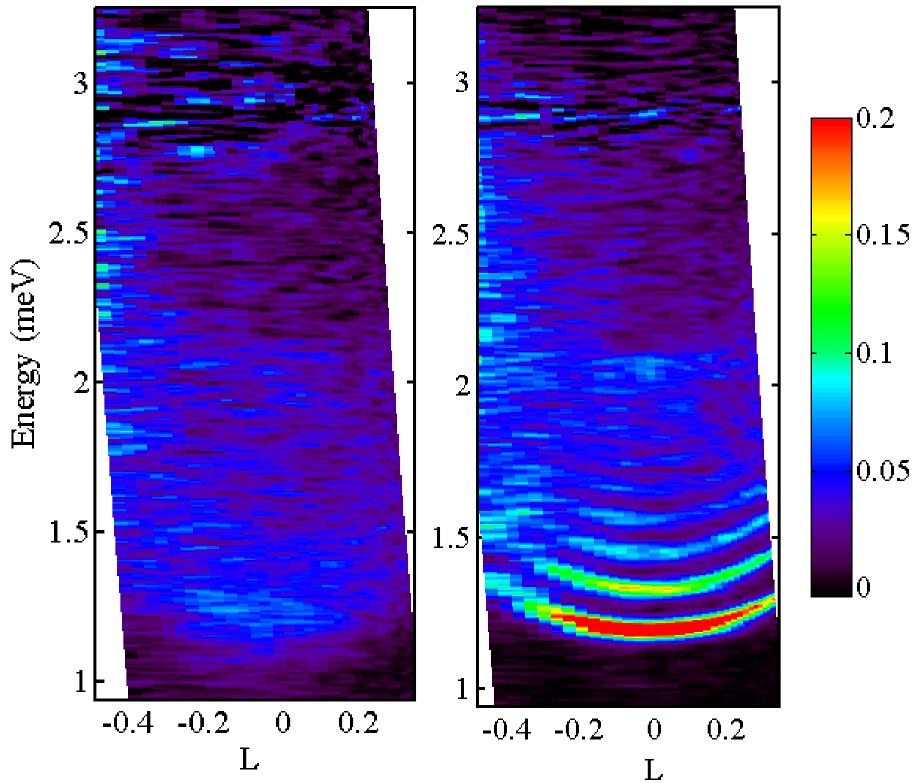
Zeeman ladder of bound states in 3D ordered phase

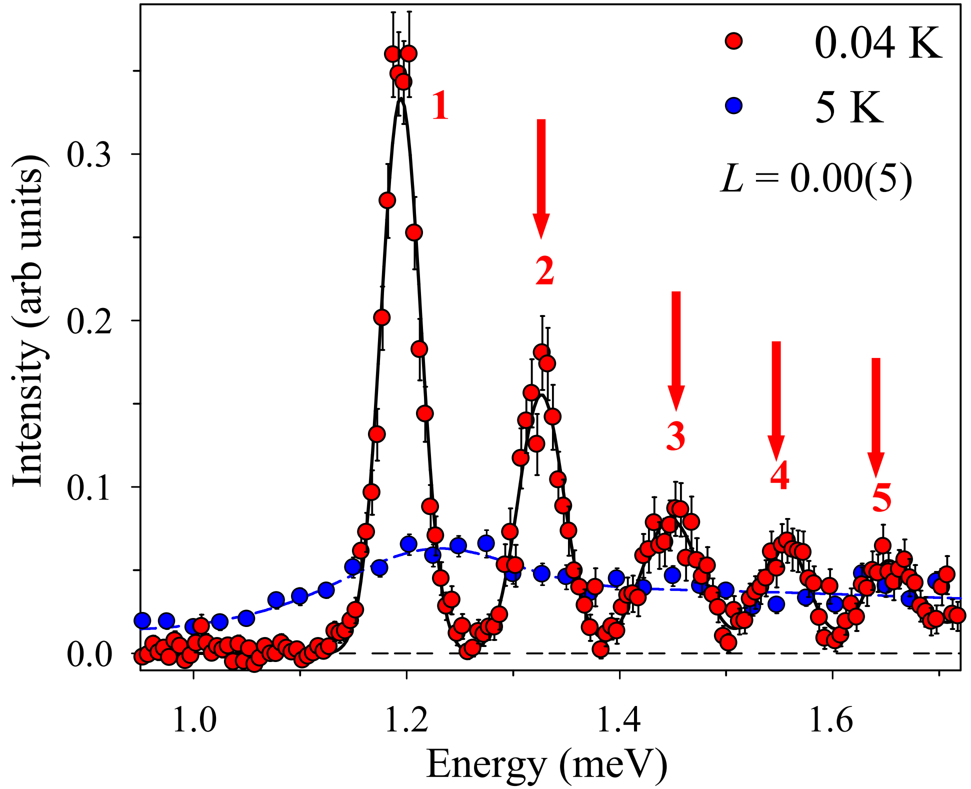
T = 5 K *T* =40 mK

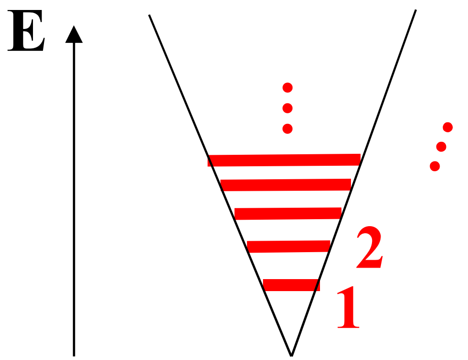


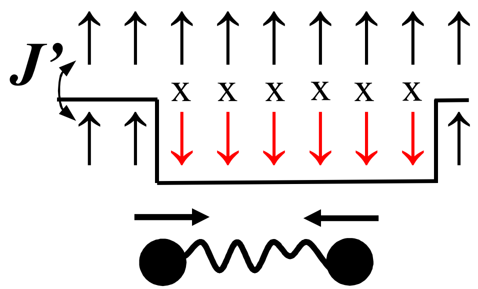
Continuum of free 2-soliton states

Bound states in confining potential

Zeeman ladder of bound states







Soliton separation costs energy

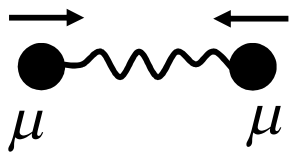
$$V(x) = \lambda x$$

$$\lambda \sim J'\langle S^z \rangle$$

Longitudinal mean-field $-hS^z$, $\lambda = 2h\langle S^z \rangle$

Soliton confinement

McCoy&Wu ('78)



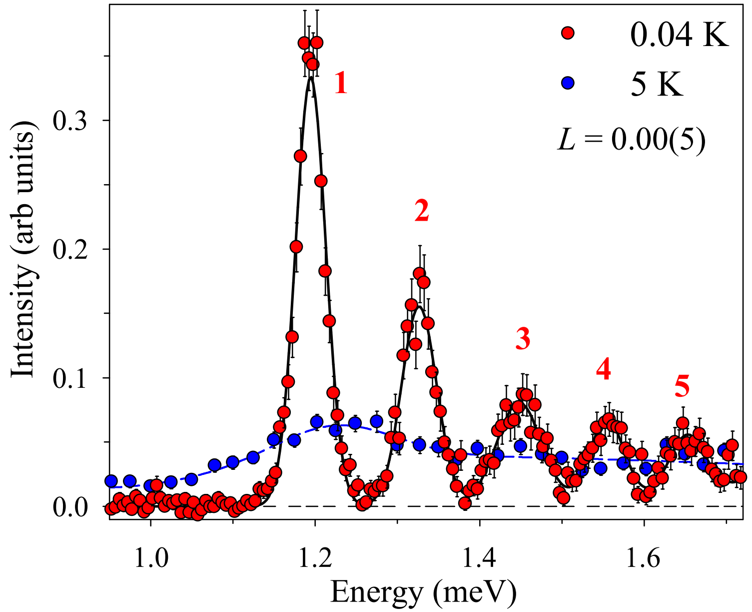
Schrödinger's equation

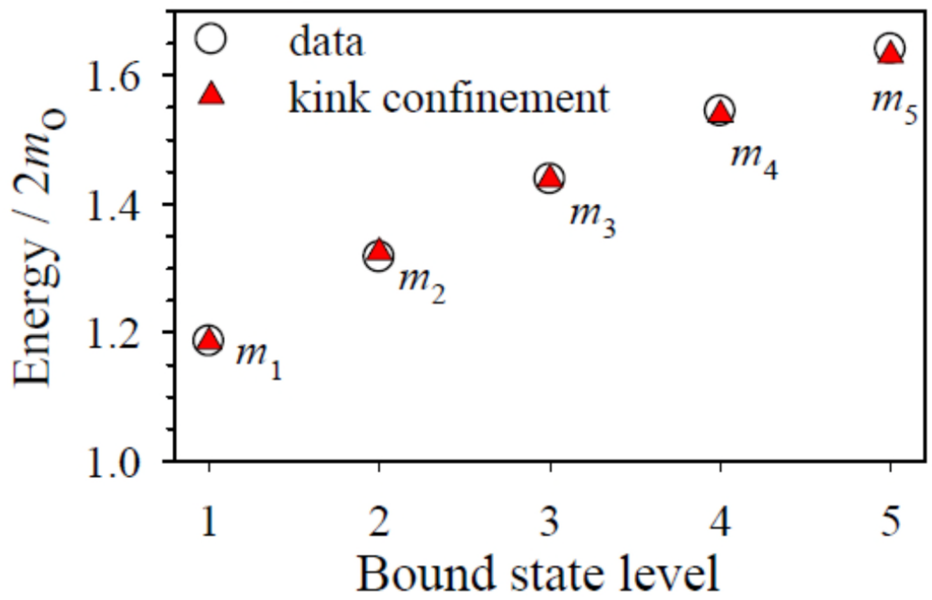
$$-\frac{\hbar^2}{\mu}\frac{d^2\varphi}{dx^2} + \lambda|x|\varphi = (m - 2m_o)\varphi$$

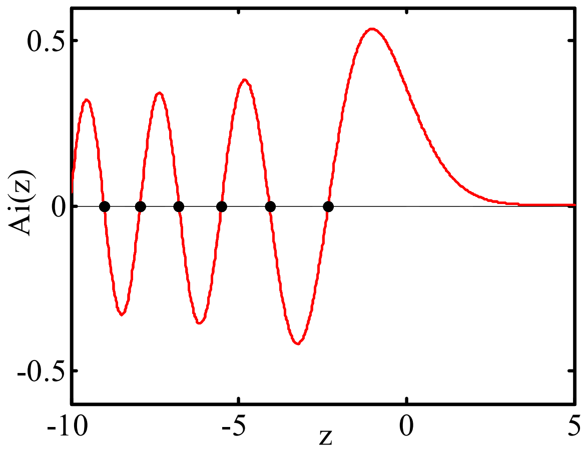
kinetic energy ***string tension***

=> bound states

$$m_j = 2m_o + z_j\lambda^{2/3}\left(\frac{\hbar^2}{\mu}\right)^{1/3}$$







$\mathrm{Ai}(-z_n) = 0$ Airy function

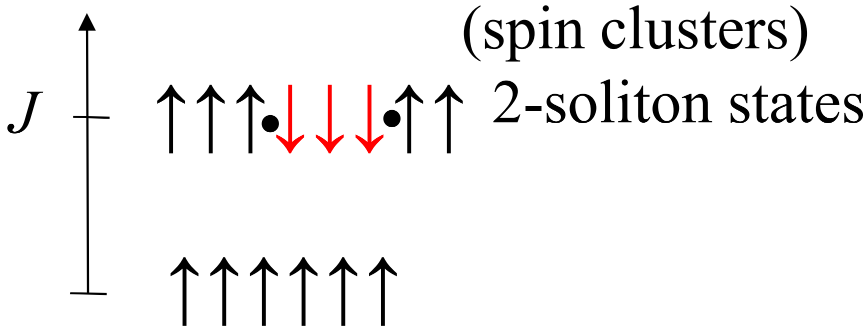
$z_n = 2.33, 4.08, 5.52, 6.78\ldots$

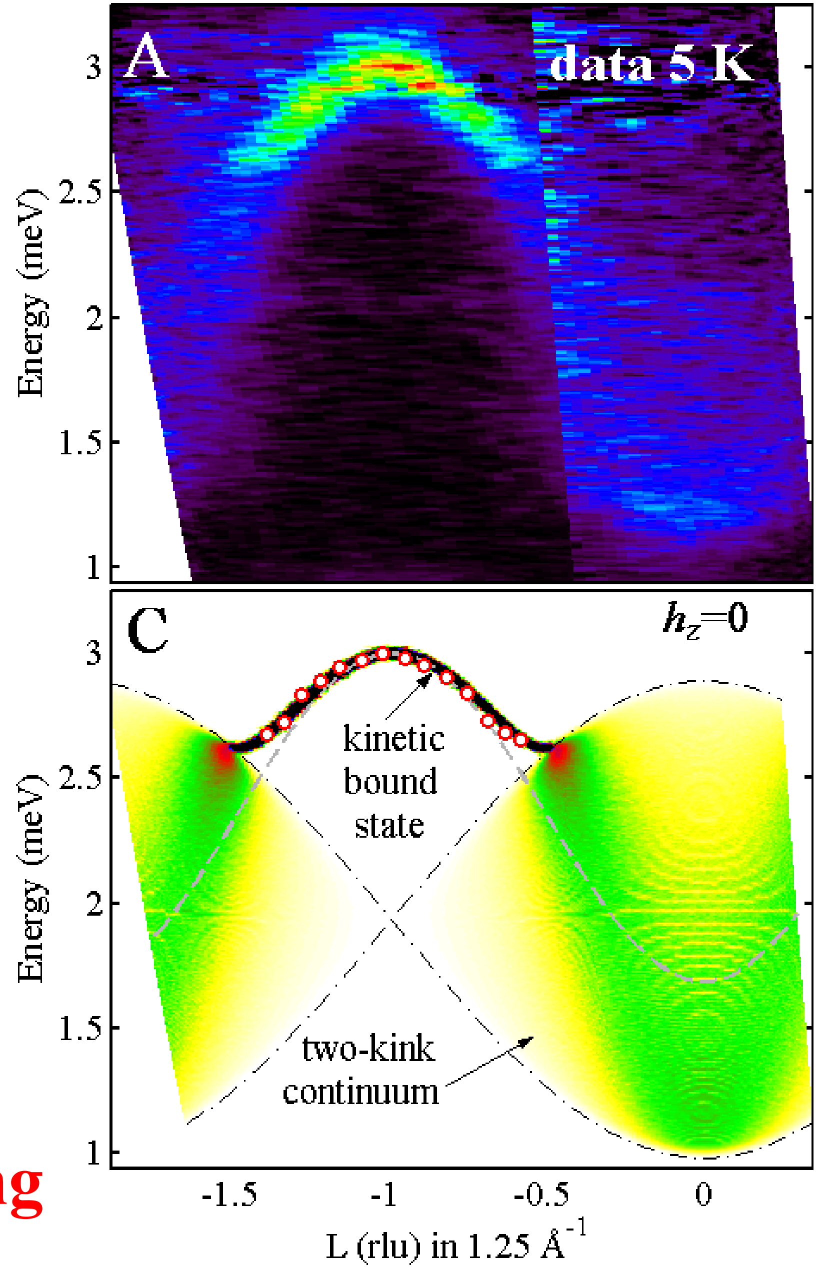
$\hbar^2/(2\mu\tilde{c}^{\,2}) = 0.23(2)$ meV

$\lambda\tilde{c} = 0.033(1)$ meV.

Phenomenological model of soliton gas

- work perturbatively around the Ising limit





$H\,|\uparrow\uparrow\uparrow\downarrow\downarrow\downarrow\downarrow\downarrow\downarrow\uparrow\uparrow\rangle$ a 2 soliton state

$= J\,|\uparrow\uparrow\uparrow\downarrow\downarrow\downarrow\downarrow\downarrow\downarrow\uparrow\uparrow\rangle$ from Ising $J\,S^z_i\,S^z_{i+1}$ **gap**

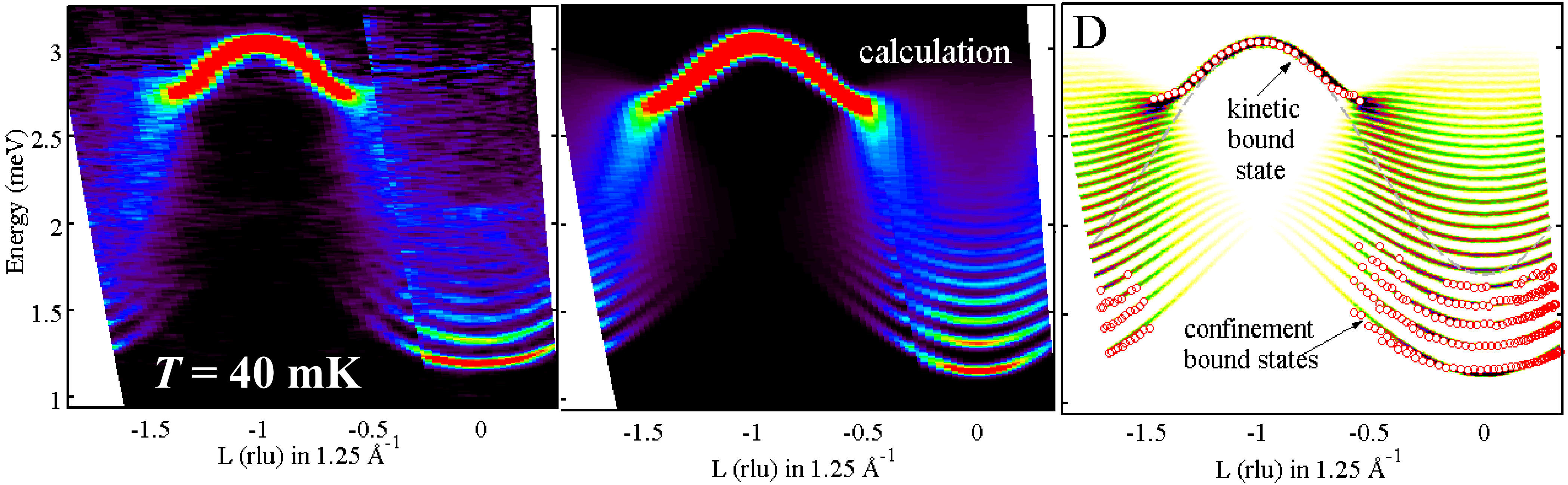
$-\alpha\,|\uparrow\uparrow\uparrow\downarrow\downarrow\downarrow\downarrow\downarrow\downarrow\downarrow\uparrow\rangle + \ldots$ **soliton hopping**

$-\beta\,\delta_{n,1}\left(|\uparrow\uparrow\underset{i+1}{\downarrow}\uparrow\uparrow\rangle + |\uparrow\uparrow\underset{i-1}{\downarrow}\uparrow\uparrow\rangle\right)$ from XY term **kinetic bound state**

$-J_{xy}\,(S^x_i\,S^x_{i+1} + S^y_i\,S^y_{i+1}) \sim S^+_i\,S^-_{i+1} + S^-_i\,S^+_{i+1}$

RC et al, Science 327, 177 (2010)

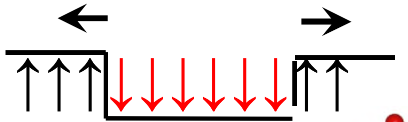
Phenomenological model of soliton gas describes full spectrum

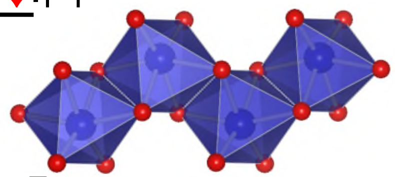


Gap: $J \sim 1.94$ meV from Ising *zz* exchange

Bandwidth $\alpha = 0.12\,J$ domain-wall hopping term

[microscopic origin S^zS^x … ?]





Kinetic bound state : transverse couplings for nn bond $S^xS^x + S^yS^y$, $J^{\perp}/J^{z} = 0.24$

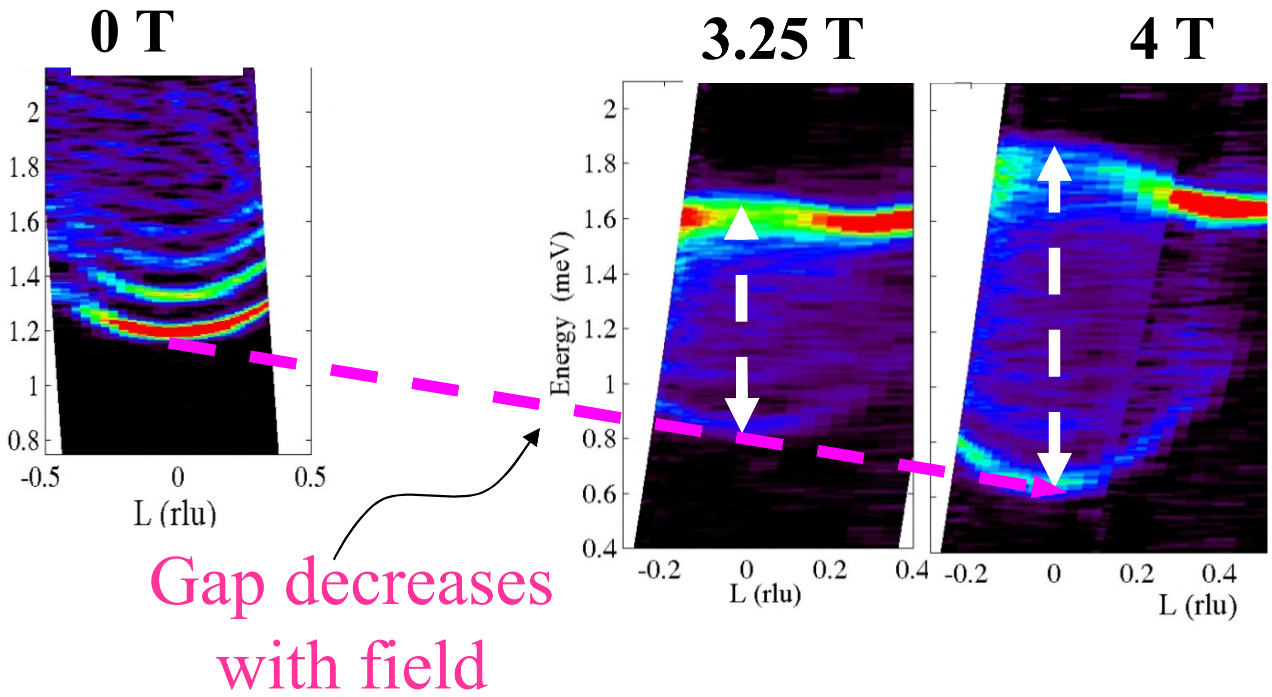
and 2-nd neighbour AFM along chain $J^{z\prime} = -0.15\,J^{z}$

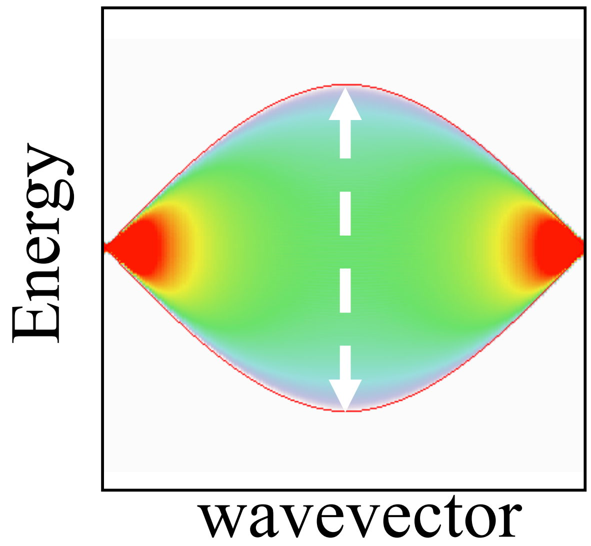
***Weak** confinement term:* $h_z \sim 0.02\,J$ longitudinal field includes interchain mean-field

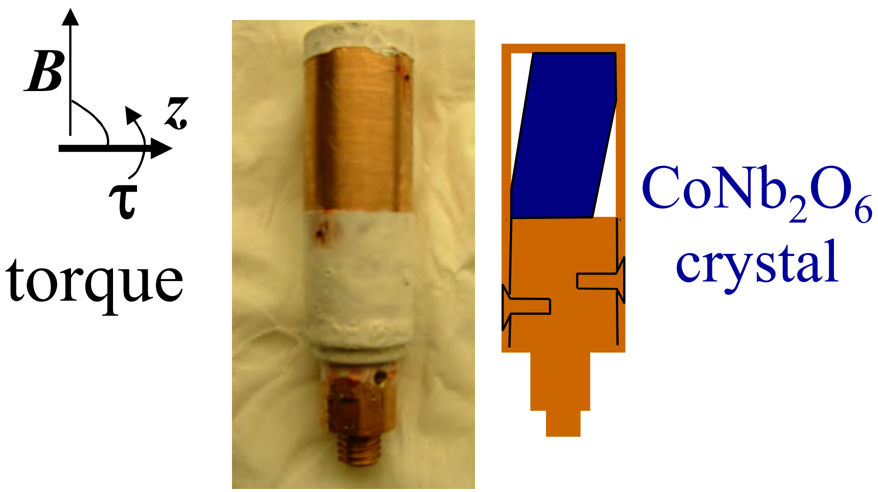
numerical calculation agrees with exact analytic solution of effective Hamiltonian

S.B. Rutkevich, J. Stat. Phys (2010)

Experiments in applied transverse field



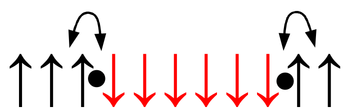




Place crystal in metallic cage to prevent movement under high torque

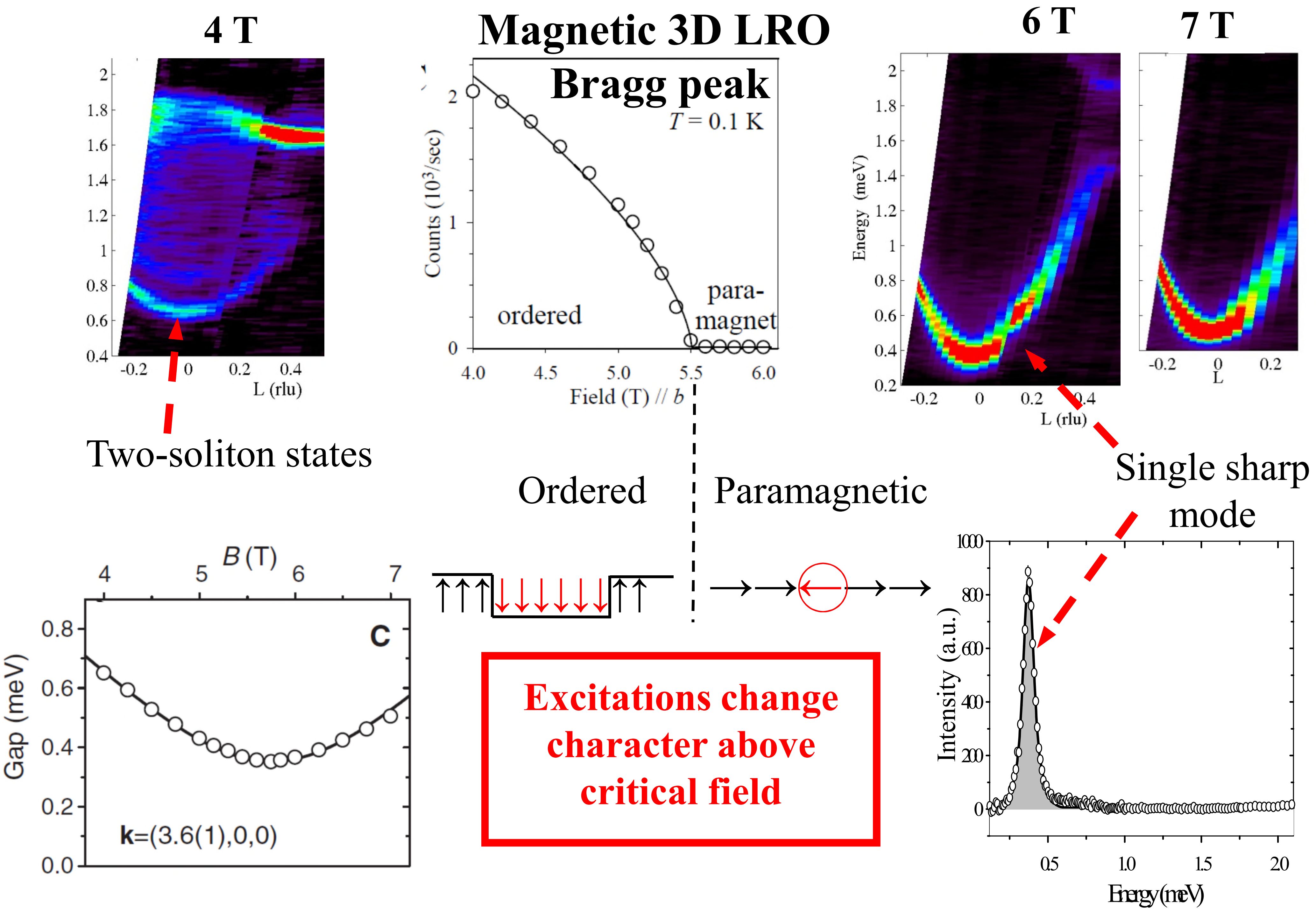
- field tunes quasiparticle dispersion

Field ~ kinetic energy

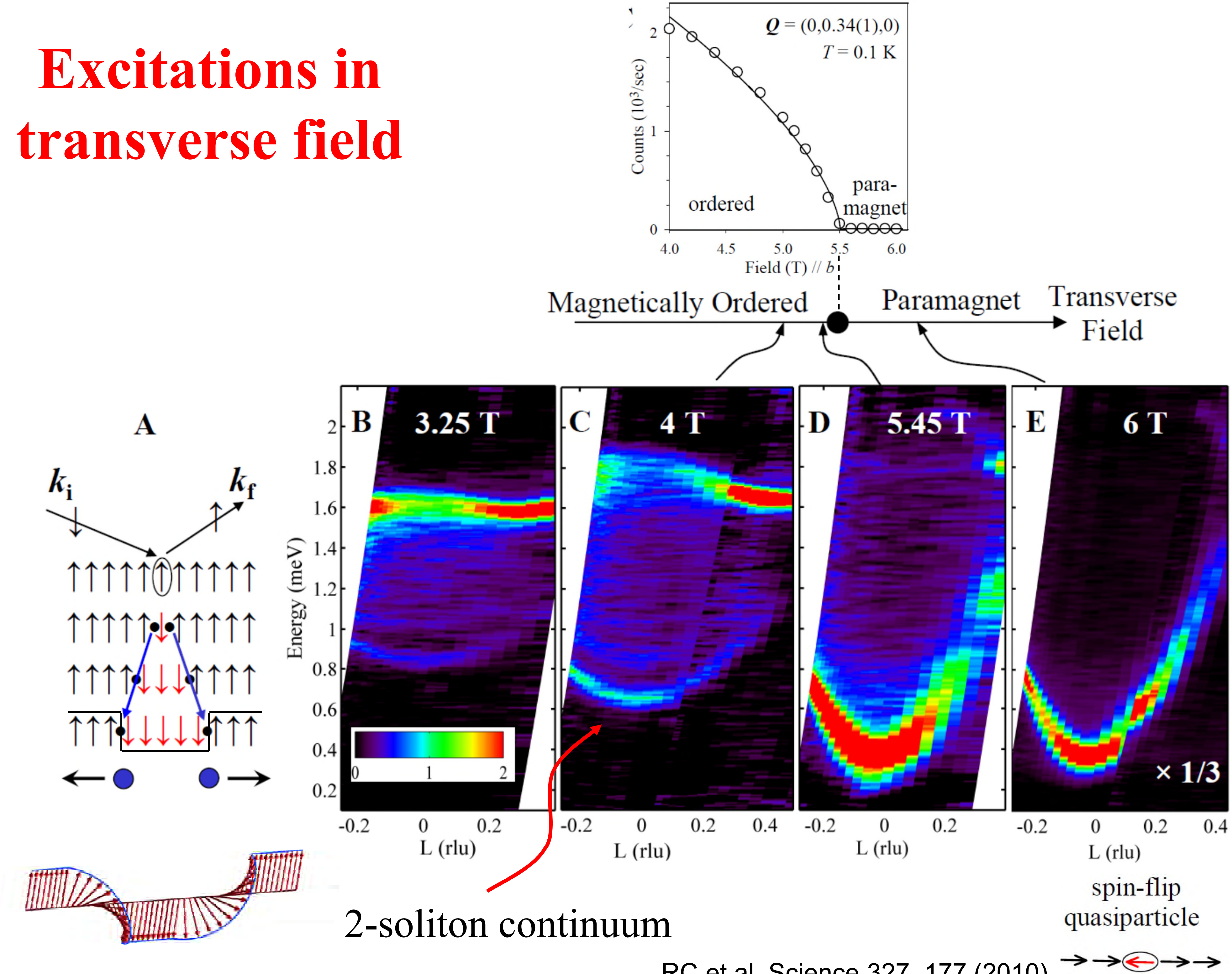


$B_x S^x = (S^+ + S^-)/2$

Excitations as a function of transverse field



Excitations in transverse field

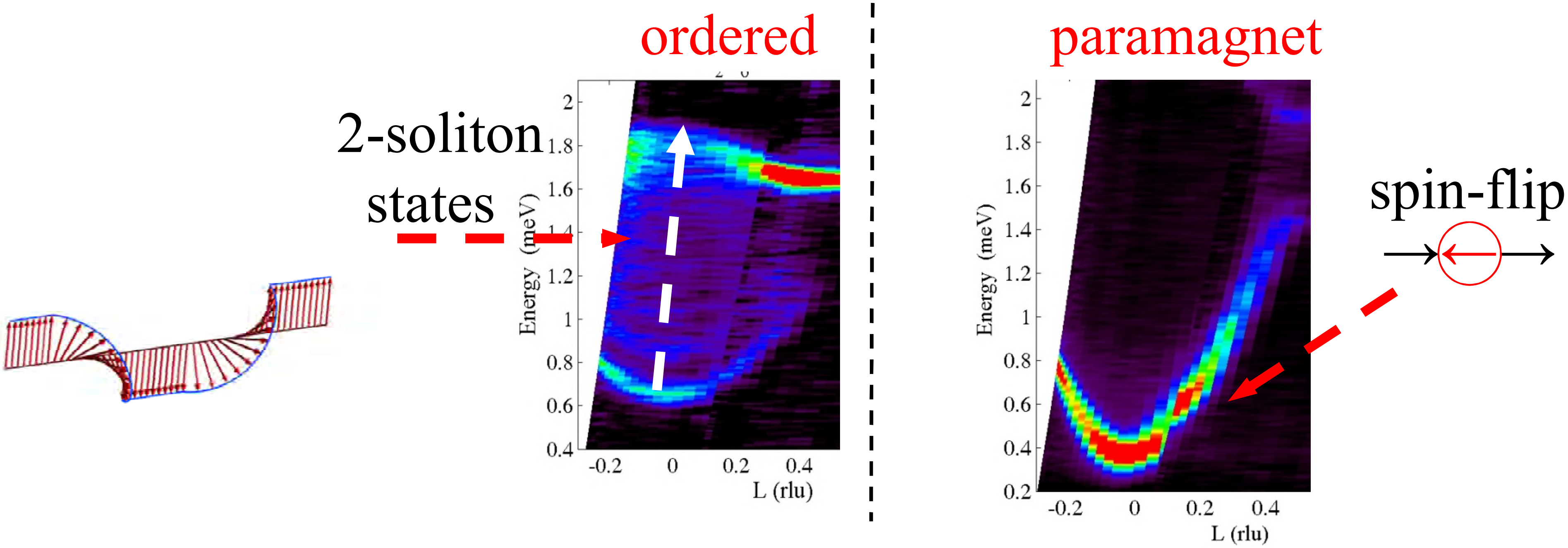


RC et al, Science 327, 177 (2010)

Summary

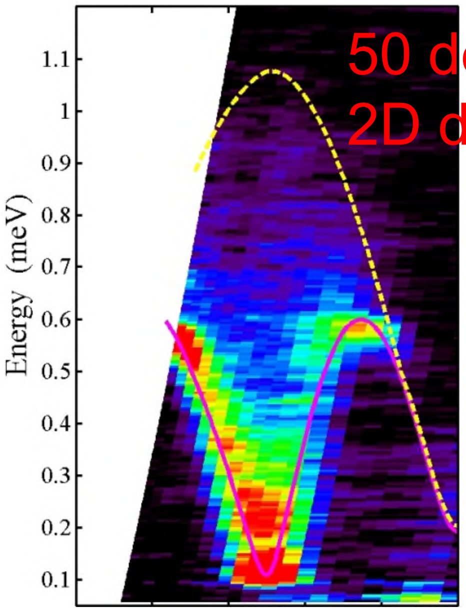
- realized experimentally field-tuned quantum phase transition in quasi 1D Ising magnet $CoNb_2O_6$

- observed transmutation of quasiparticles at critical point

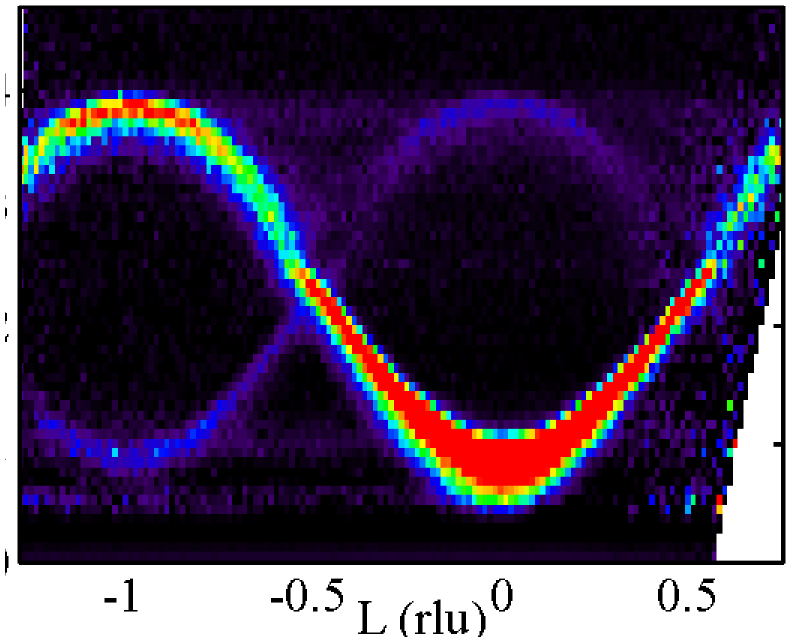


Conclusions

- neutron scattering **is a very versatile** probe of magnetic ordering and dynamics (μeV->eV, magnetic field (15 T->25 T), low T (mK)
- **quantitative:** probe dispersions of excitations through well-understood matrix element (quantitative comparison with theories)
- sample size limited, large crystals needed (advanced crystal growth)
- new neutron sources (ISIS 2nd target station, USA + JAPAN, ESS) will bring **new opportunities:** higher flux, higher resolution, wider coverage



50 detectors,
2D data set



40,000 detectors,
4D data set, 4
energies measured
simultaneously,

- complementary to resonant x-ray diffraction and inelastic (RIXS) (samples 10's of μm, resolution >30 meV, T > few K – beam heating)

Collaborators

La_2CuO_4

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