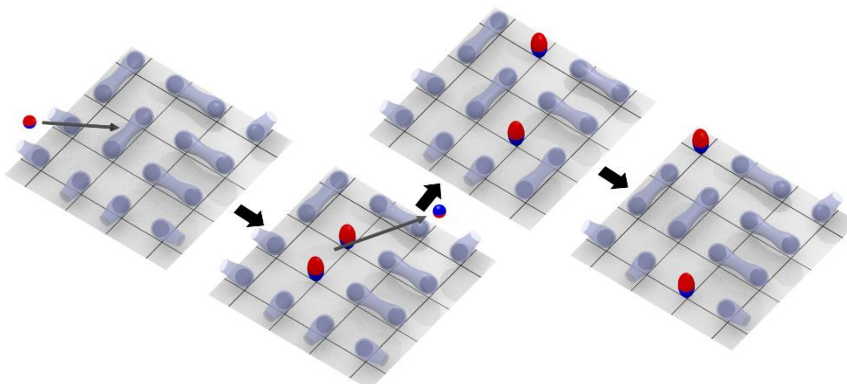
# Quantum Magnetism
# - Neutrons in the Quasi-particle Zoo



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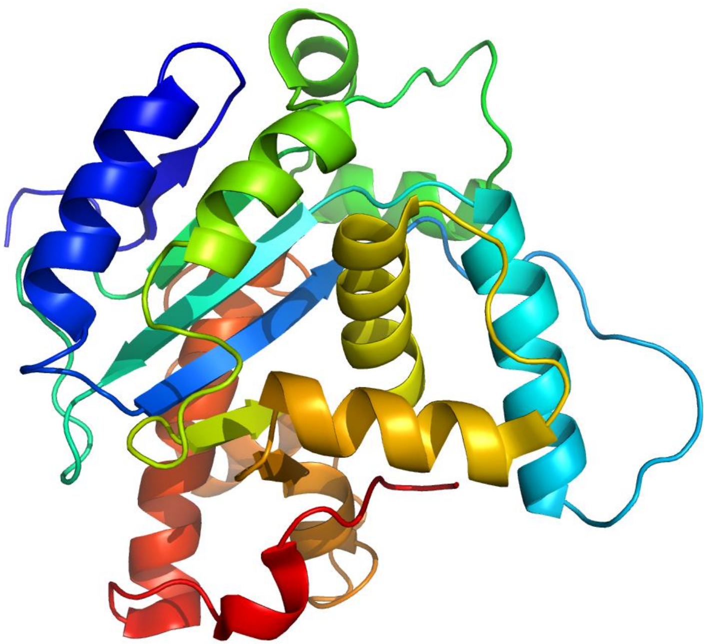
# Outline

- Quantum Magnetism
  - Arena for many-body physics and highly correlated materials
  - Models-Materials-Measurements
- Neutron scattering
  - Basics, uniqueness, and a bright future
  - The quasi-particle zoo
- Selected examples
  - Multi-spinons in one-dimensional chains
  - Spin-wave anomaly and quest for pairing in 2D

# Complexity of many-body systems

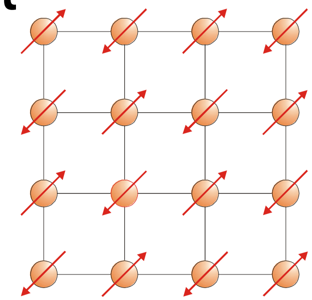
- Structure of a protein



- Pop2p-subunit Jonstrup et al (2007)
- Mega-Dalton:
  ~1’000’000 atoms (5 colors?)
  ~3’000’000 numbers needed
  to describe the structure

Ground state of a magnet

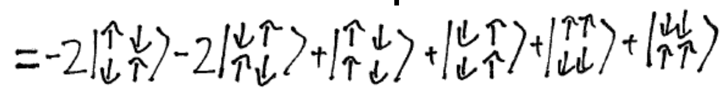
$$\mathcal{H} = J \sum \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$



1 spin: trivial

2 spins: singlet state $|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$

4 spins: back-of-the-envelope calc.



16 spins: 10 seconds on computer (4GB)

40 spins: World record: 1’099’511’627’776
coefficients needed to describe a state

**Classical: 3N   Quantum: $2^N$**

$10^{23}$ spins:
- 1D: analytic solution (Bethe 1931)
- 2D: antiferromagnet (Néel 1932) or fluctuating singlets? (Anderson 1973,1987)

$10^{23}$ ±some electrons: High-$T_c$ superconductivity
– THE enigma of modern solid state physics

# Spin – the *drosophila* of quantum physics

Spin: an atomic scale magnetic moment

- Quantization: S=0, 1/2, 1, 3/2,.....∞

S = 1/2 → +1/2 ↑ ; - 1/2 ↓

- Superposition: $|\psi\rangle = \alpha|\uparrow\rangle + \beta|\downarrow\rangle$

  likelihood of up: $\rho(\uparrow) = |\langle\uparrow|\psi\rangle|^2 = \alpha^2$

- Quantum fluctuations

  average moment $\langle S^z\rangle = 0$

  imagine that spins fluctuate in 'imaginary time'

- Quantum correlations

  $|\psi\rangle = ( |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle ) /\sqrt{2}$

*e.g.* two spins 'entangled'

this is why $\propto 2^N$, not $\propto N$

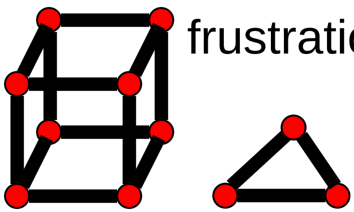
# Quantum Magnetism – an arena for quantum phenomena

## 1) Model and Materials

Spin, interactions

dimension

frustration



The LEGO of many body physics

## 3) Experimental tools:

Bulk probes

Neutron scattering

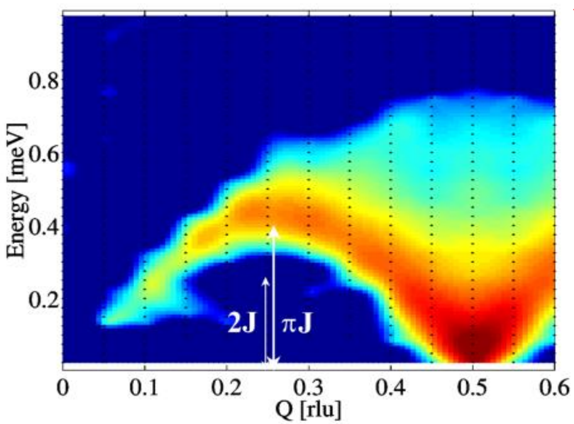
## Phenomena:

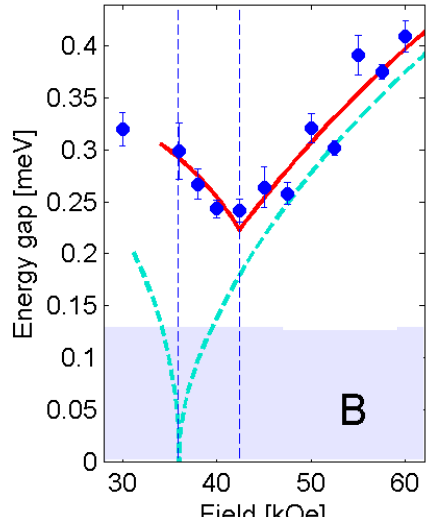
Order, phase transitions,
quantum fluctuations,
collective excitations,
entanglement …

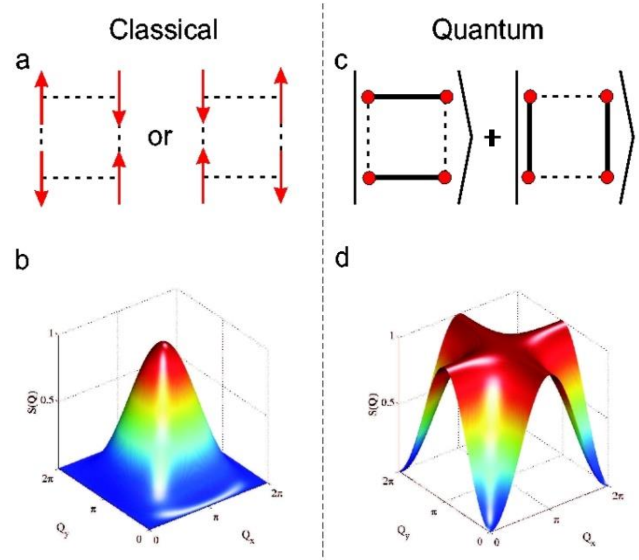
## 2) Theoretical methods

analytic approximations

numerical simulations

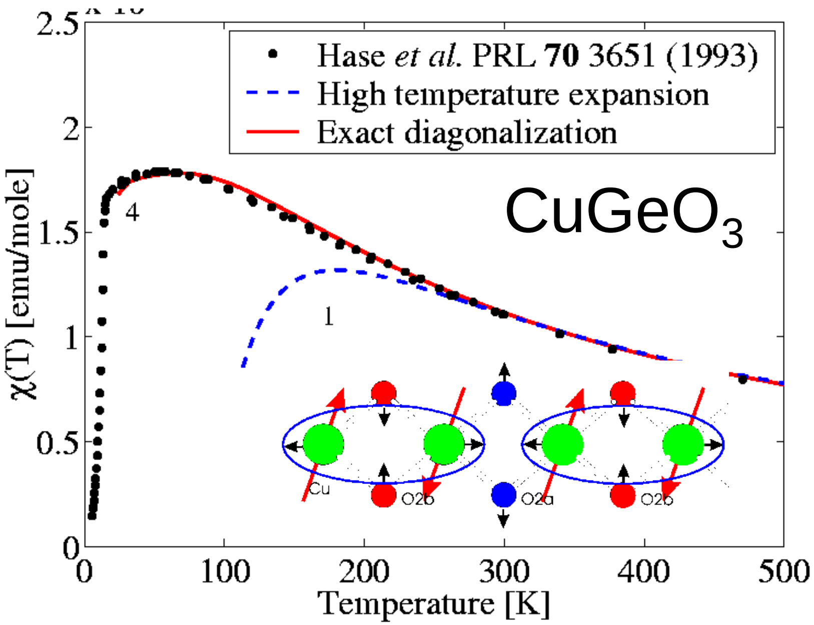


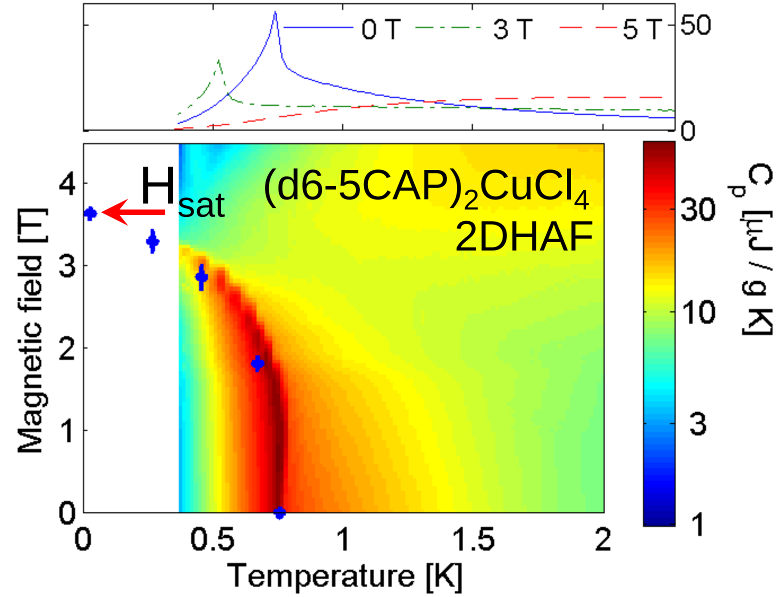



# Magnetic measurements

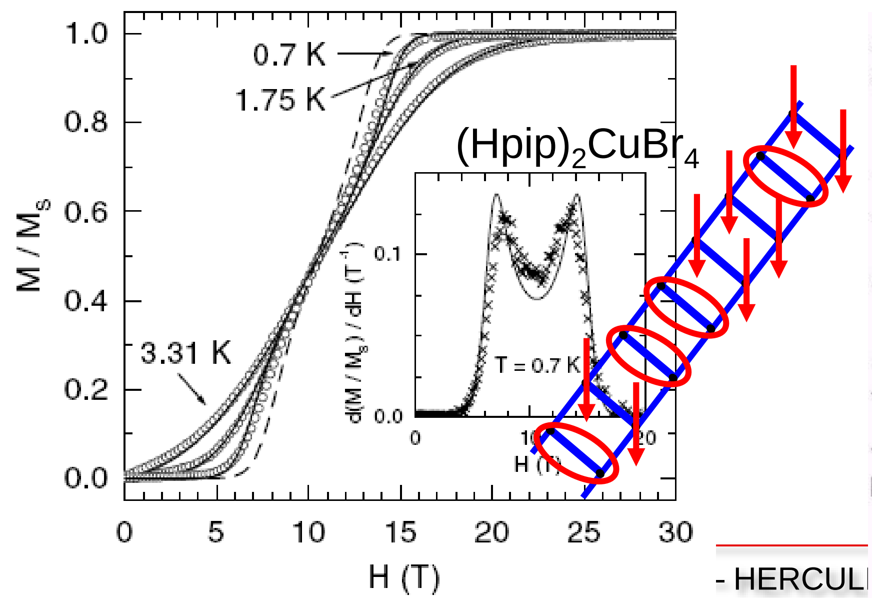
Susceptibility



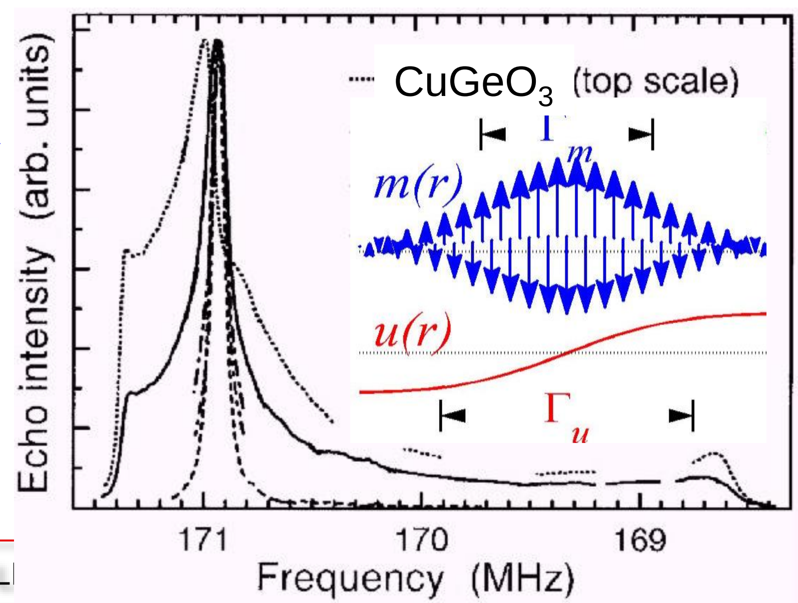

Specific heat




Magnetization

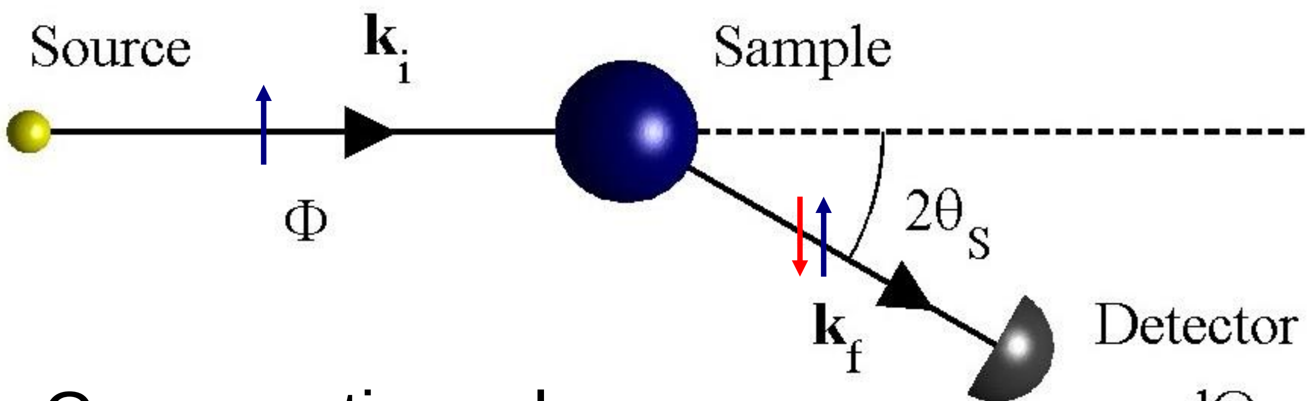


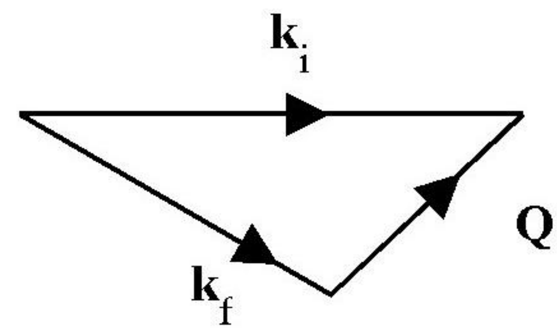

NMR, µSR etc.



# A unique tool: Neutron scattering

scattering and conservation rules



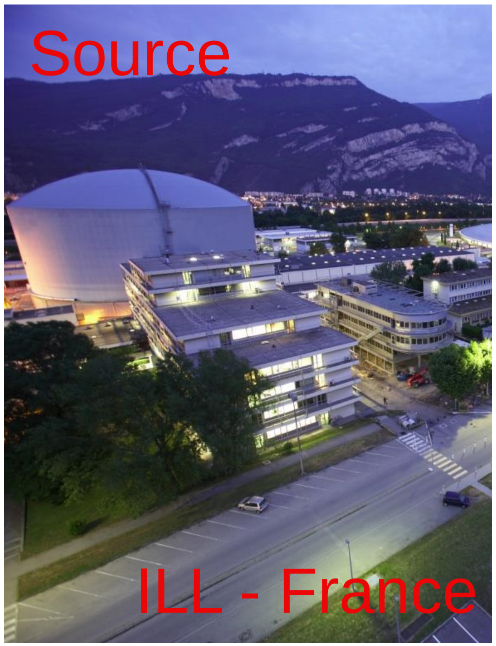



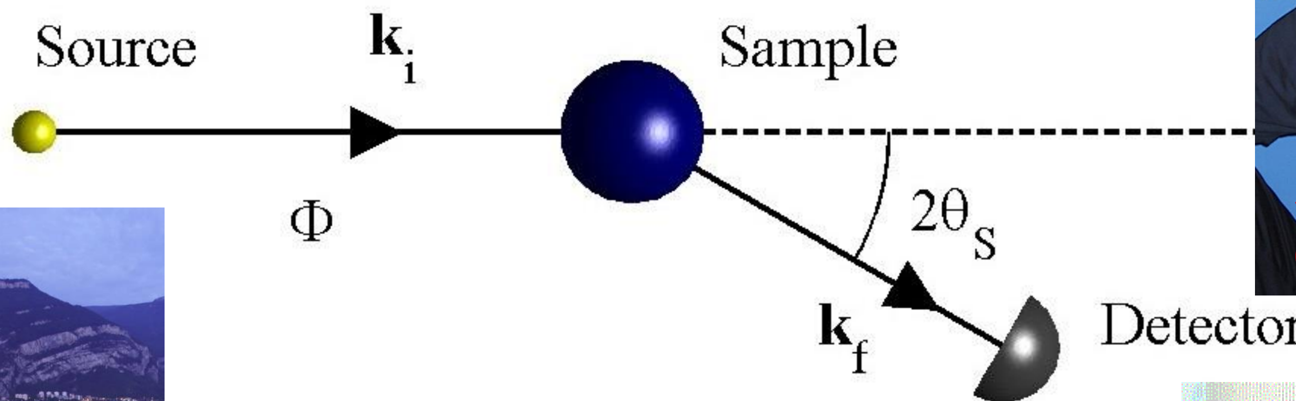

Conservation rules:

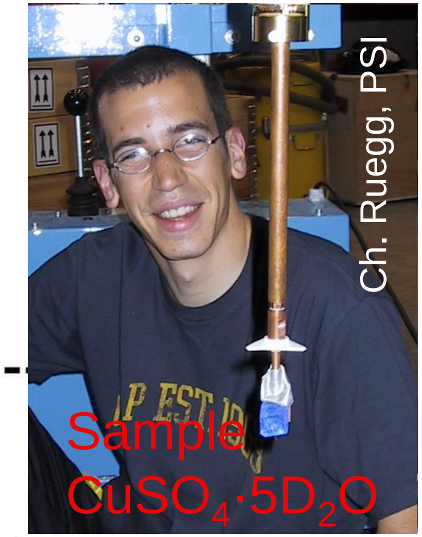
| | Sample | | Neutron | | |
|---|---|---|---|---|---|
| Momentum | $\hbar\mathbf{Q}$ | = | $\hbar\mathbf{k}_i - \hbar\mathbf{k}_f$ | | |
| Energy | $\hbar\omega$ | = | $E_i - E_f$ | = | $\hbar(k_i^2 - k_f^2)/2m_n$ |
| Spin | $\Delta S$ | = | $\sigma_i - \sigma_f$ | | |

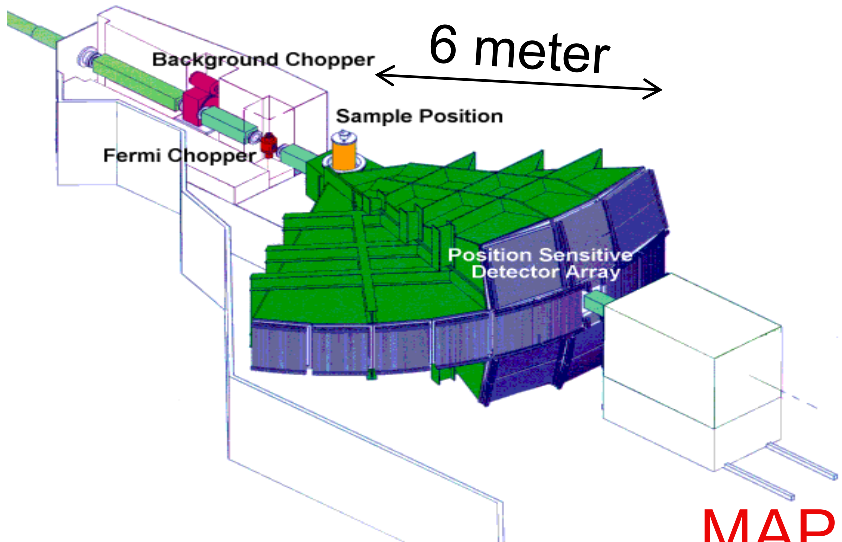
$\Rightarrow$ We can control and measure these quantities !

# Large scale instruments and facilities




MAPS 16m$^2$ detector bank

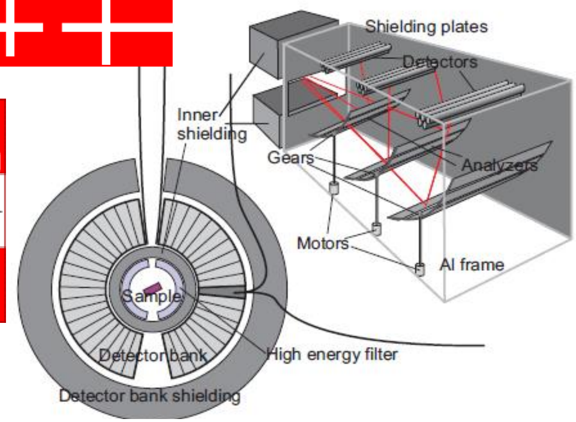
# Neutron scattering – an intense future

- $1^{st}$ generation facilities:
  - General purpose research reactors
- $2^{nd}$ generation facilities:
  - Dedicated to neutron scattering:
  - ILL, France, FRM2 Munich, SINQ CH, ISIS, UK etc.
- $3^{rd}$ generation facilities:
  - SNS, US 1.4b$, commission 2006
  - J-Parc, Japan 150b¥, commission 2008
  - ESS, Sweden 1.4b€, start 2013, commission 2019
  - China Spallation, start 2011*, commission 2018
- $2^{nd}$ to $3^{rd}$ generation gains of 10-1000 times !
  - Faster experiments, smaller samples, better data
  - Time resolved physics, new fields of science
  - New instrument concepts

European Spallation Source (ESS)
- All eyes are on Lund – congratulations !

Switzerland will contribute 3-4%
⇒ CH-DK collaboration on 5 instrument design workpackages
⇒ CAMEA: $10^2$ -$10^4$ over current instr.

ESS + ✚






# Neutron scattering cross-section – the power of simplicity

From initial state ***i*** to final state ***f*** of neutron **k** and sample λ

$$\left(\frac{d^2\sigma}{d\Omega\, dE_f}\right)_{\lambda_i\to\lambda_f} = \frac{k_f}{k_i}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \underline{|\langle \mathbf{k}_f\lambda_f\,|V|\,\mathbf{k}_i\lambda_i\rangle|^2}\;\delta(E_{\lambda_i}-E_{\lambda_f}+\hbar\omega)$$

Neutrons treated as plane waves:

$$|\mathbf{ks}_n> = V^{-1/2}\exp(i\mathbf{k}\cdot\mathbf{r}_n)\,|\mathbf{s}_n>$$

Energy conservation ⇒ integral rep.:

$$\delta(\hbar\omega+E_i-E_f) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} e^{i(\hbar\omega+E_i-E_f)t/\hbar}dt$$

Fourier transform in

- space/momentum
- time/energy

# Magnetic neutron scattering

$$|\langle \mathbf{k}_f \lambda_f \,|V|\, \mathbf{k}_i \lambda_i \rangle|^2$$

## Dipole interaction – electron spin and orbit moment

$$V_{\mathrm{mag}}(\mathbf{r}) = -\frac{\mu_0}{4\pi} 2\gamma \mu_N \mu_B \boldsymbol{\sigma}_n \cdot \left( \nabla \times \left( \frac{\mathbf{s} \times \hat{\mathbf{R}}}{|\mathbf{R}|^2} \right) + \frac{1}{\hbar} \frac{\mathbf{p} \times \hat{\mathbf{R}}}{|\mathbf{R}|^2} \right)$$

$$\left( \frac{d^2\sigma}{d\Omega dE_f} \right)_{\mathrm{mag}} = \frac{(\gamma r_0)^2}{2\pi\hbar} \frac{k_f}{k_i} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) |gF_R(Q)|^2 \sum_{RR'} \int dt e^{iQ(R-R')-i\omega t} \langle S_R^\alpha(0) S_{R'}^\beta(t) \rangle$$

cross-section

pre factor

dipole factor

magnetic form factor

Fourier transform

spin-spin correlation function

# Dynamic structure factor

Spin-spin correlation function

$$S^{\alpha\beta}(Q,\omega) = \frac{1}{2\pi}\sum_{RR'}\int dt e^{iQ(R-R')-i\omega t}\langle S_R^\alpha(0)S_{R'}^\beta(t)\rangle$$

Dynamic structure factor

Theory !

$$\left(\frac{d^2\sigma}{d\Omega\, dE_f}\right)_{\mathrm{mag}} = \frac{1}{\hbar}\frac{k_f}{k_i}p^2\exp(-2W)\sum_{\alpha\beta}(\delta_{\alpha\beta}-\hat{\mathbf{Q}}_\alpha\hat{\mathbf{Q}}_\beta)S^{\alpha\beta}(\mathbf{Q},\omega)$$

Fluctuation dissipation theorem ⇒ gen. susceptibility

$$S(\mathbf{Q},\omega) = [\,n(\omega)+1\,]\,\chi''(\mathbf{Q},\omega) = \frac{\chi''(\mathbf{Q},\omega)}{1-\exp(-\hbar\omega/k_BT)}$$

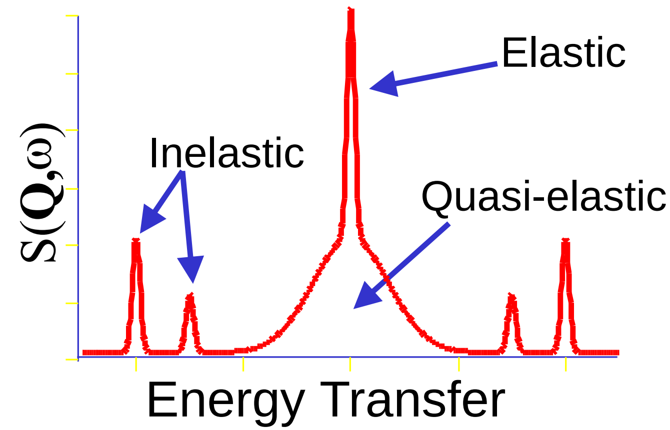
intrinsic dynamics ⇔ response to perturbation

# Structure factors – time and energy

- Dynamic structure factor: inelastic

$$S(\mathbf{Q},\omega) \propto \int_{-\infty}^{\infty} dt e^{-i\omega(t-t')} \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t') \rangle$$

  – periodic: $\sin(\omega_0 t) \Rightarrow$ peak: $\delta(\omega_0-\omega)$
  decay: $\exp(-t/\tau) \Rightarrow$ Lorentzian: $1/(1+\omega^2\tau^2)$




- Static structure factor: elastic

$$S(\mathbf{Q},\omega=0) \propto \int_{-\infty}^{\infty} dt \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t') \rangle \simeq \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(\infty) \rangle$$

  – Bragg peaks at $\omega = 0$

- Instantaneous structure factor - integrate over energy

$$S(\mathbf{Q}) = \int d\omega S(\mathbf{Q},\omega) \propto \int_{-\infty}^{\infty} dt \delta(t-t') \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t') \rangle = \langle S_{\boldsymbol{r}'}(t) S_{\boldsymbol{r}}(t) \rangle$$

  – Finite time/length scale of correlations

# Remark: instantaneous correlations

$$\langle S_{\mathbf{r}'}(t) S_{\mathbf{r}}(t) \rangle \propto e^{-|r-r'|/\xi}$$

$$\Downarrow$$

$$S(\mathbf{Q}) \propto \frac{1}{1+Q^2\xi^2}$$

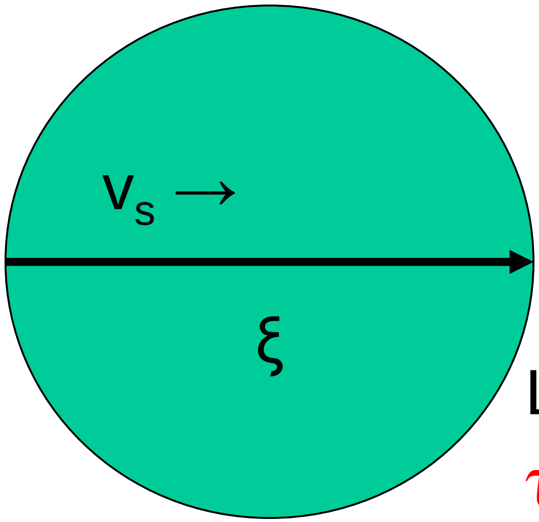
Width $\Rightarrow$ Correlation length $\xi$

**Softening**

$v_s \propto Z_c$

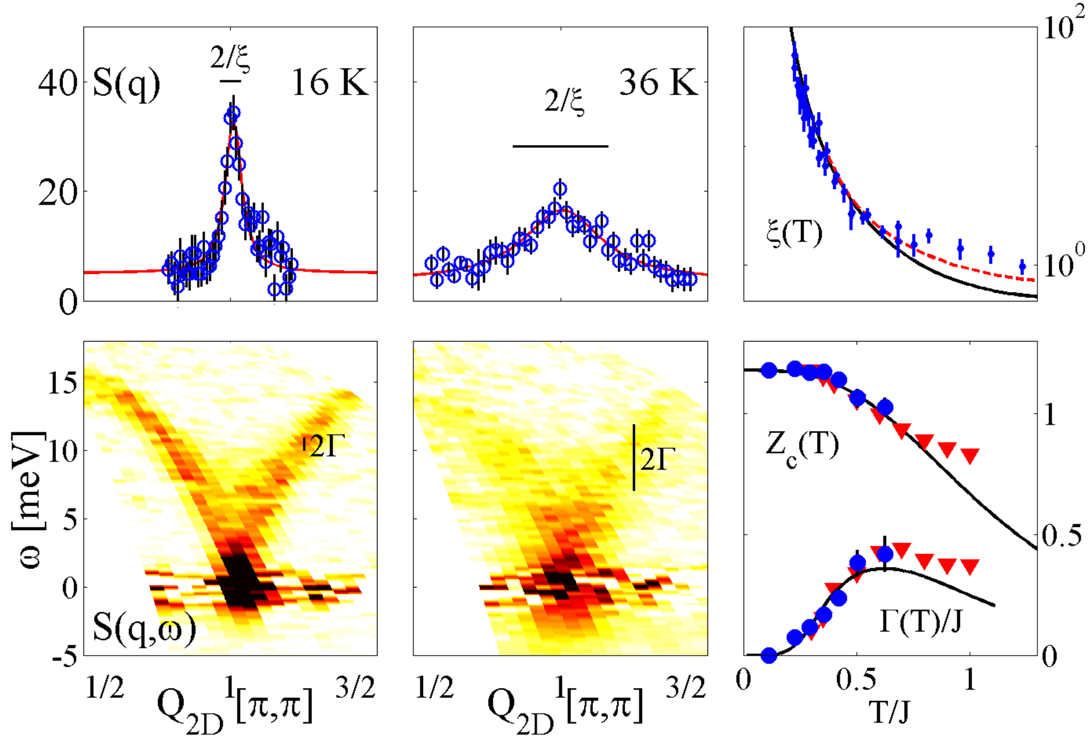
**Damping**

$\Gamma = v_s / \xi$




**Life-time**

$\tau = \xi / v_s = 1/\Gamma$




Correlations and fluctuations on the 2D square lattice Heisenberg antiferromagnet:

J. Mag. Mag. Mat. 236, **4** (2001)

PRL **82**, 3152 (1999); **87**, 037202 (2001)

# Inelastic magnetic scattering: Lets take the scenic route…

Between long range ordered states

Selected examples

– the zoo :

- Spin-flip, singlet-triplet, dispersive triplets
- 1D spin chain
  – spinons vs spin waves
- 2D HAF zone boundary anomaly
  – as instability of spin waves ?
  – the smoking gun of RVB ?

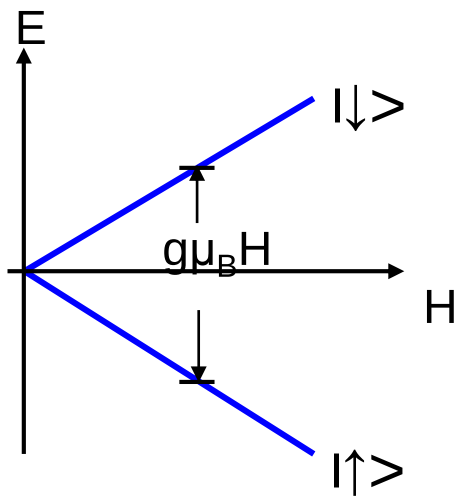
Aim:

- Show the many types of quasiparticles
- Show quantitativeness of neutron scattering

… and spin liquids

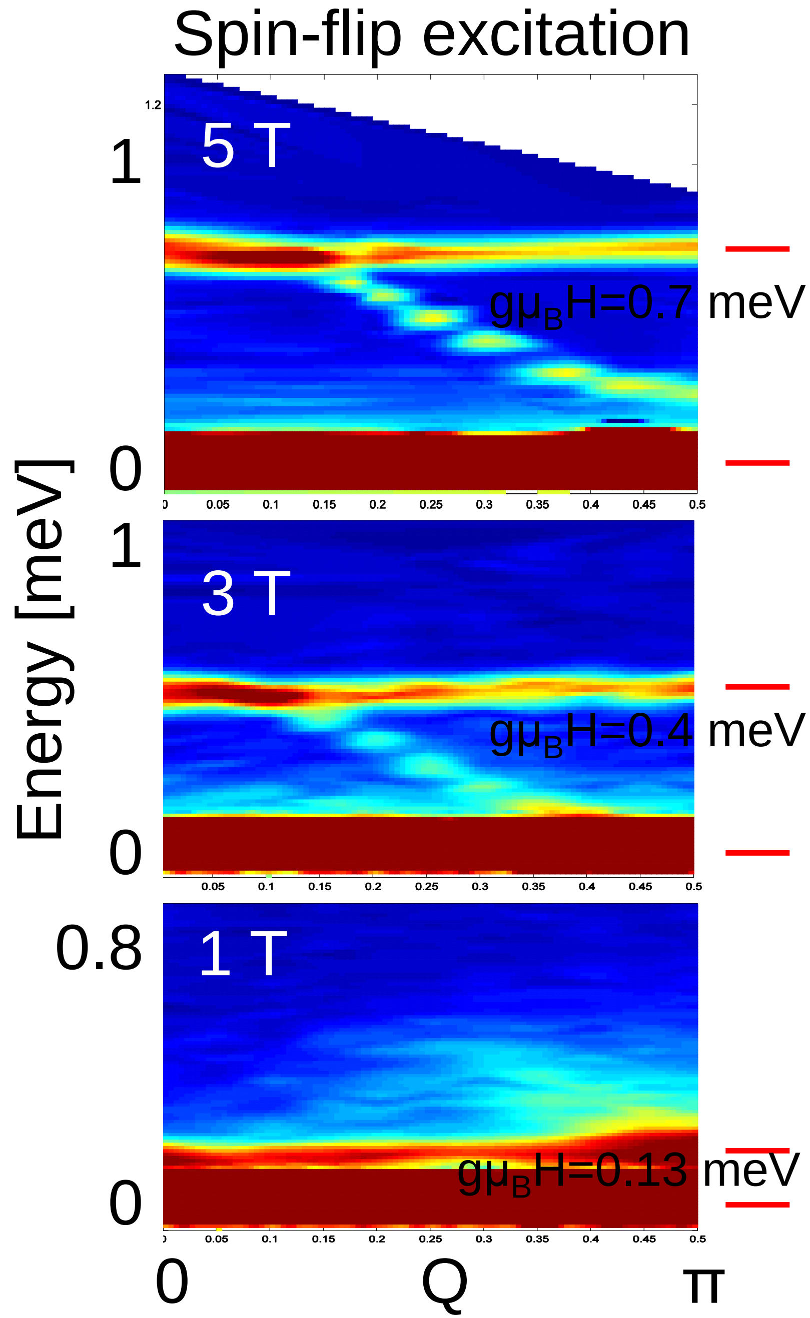
# paramagnetic spins S=1/2

- Two states |↑⟩, |↓⟩, can be magnetized
- Zeemann-split energy of the levels
- A gap for transitions



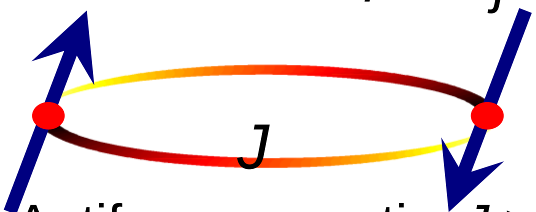

- Local excitation

  ⇒ no Q-dependence



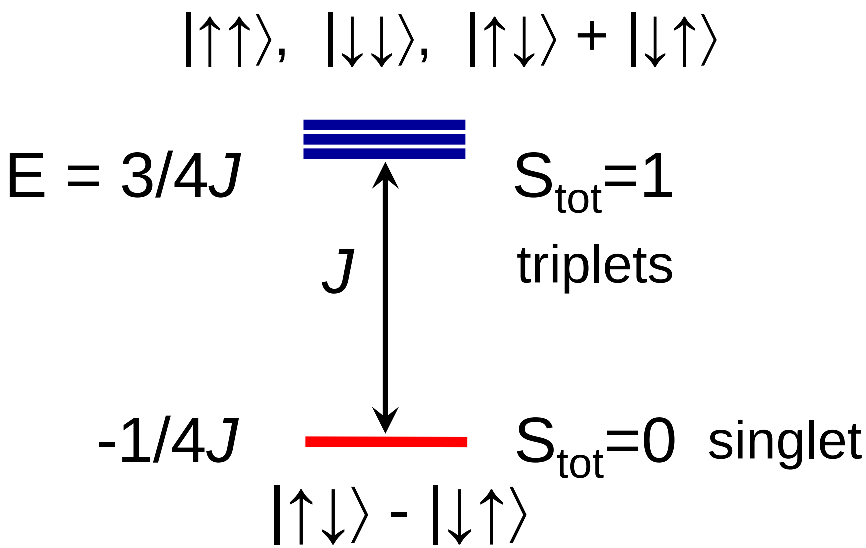
# Take two – the spin pair

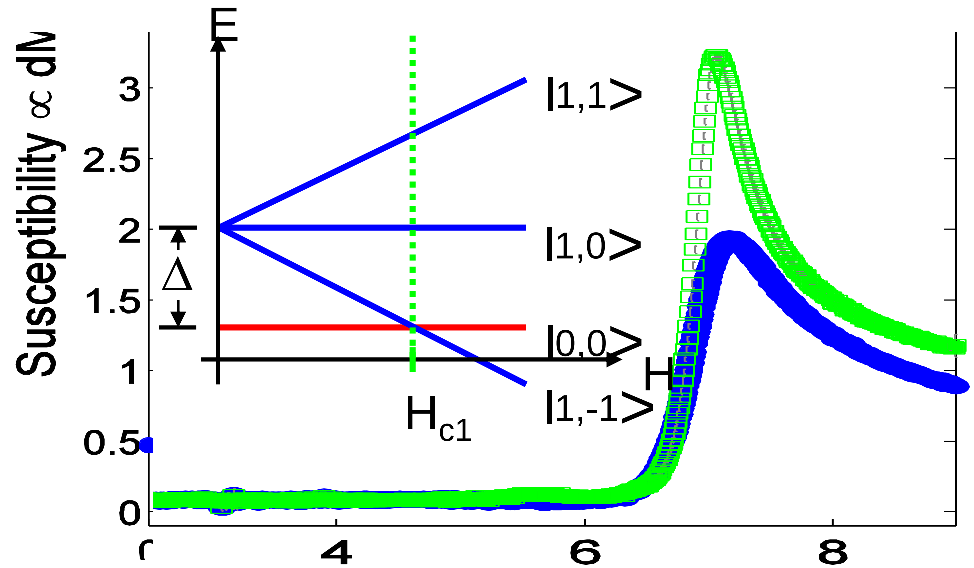
$$\mathcal{H} = J \sum \mathbf{S}_i \cdot \mathbf{S}_j$$




Antiferromagnetic: $J > 0$

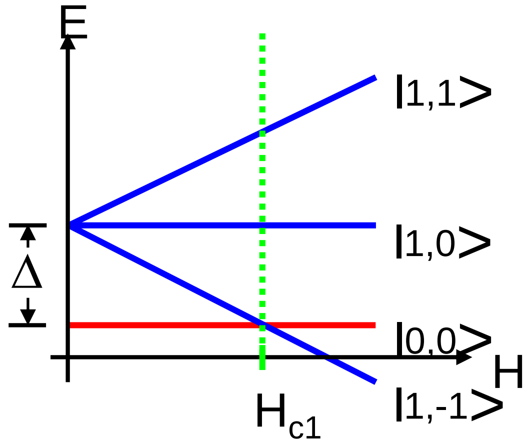
No magnetization or susceptibility up to critical field

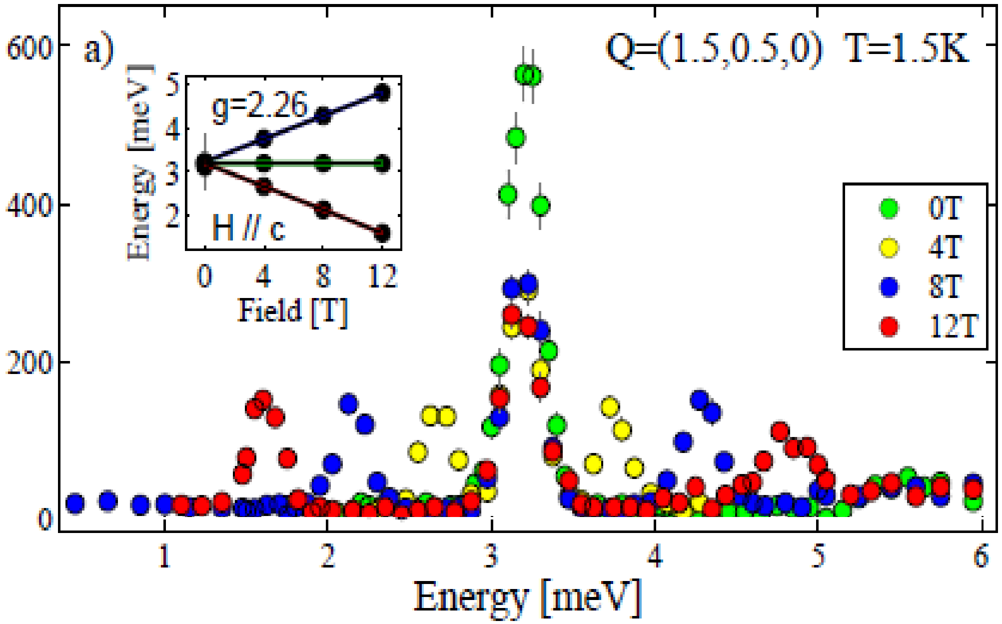


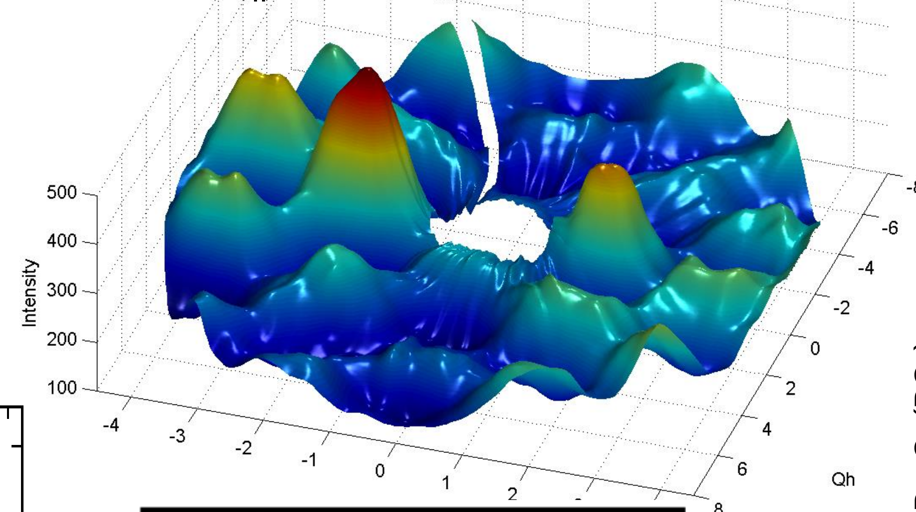

Singlet ground state: $\langle S^z{}_1 \rangle = \langle S^z{}_2 \rangle = 0$

# Singlet-Triplet excitations

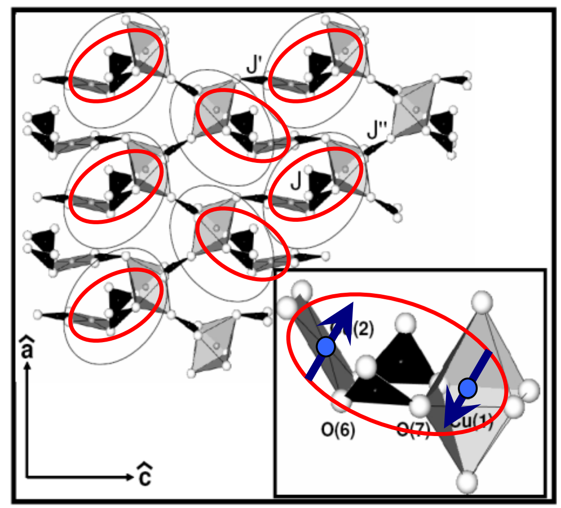



Structure factor along pairs:
$S(q) \sim \cos(d_{||} \cdot q) + \cos(d_{\perp} \cdot q)$

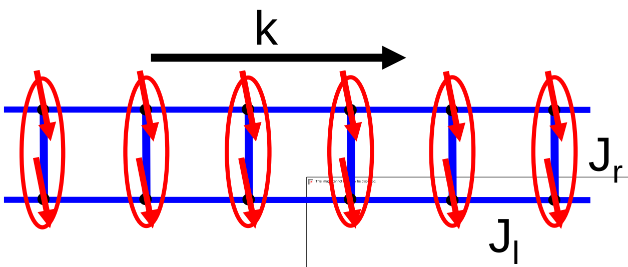


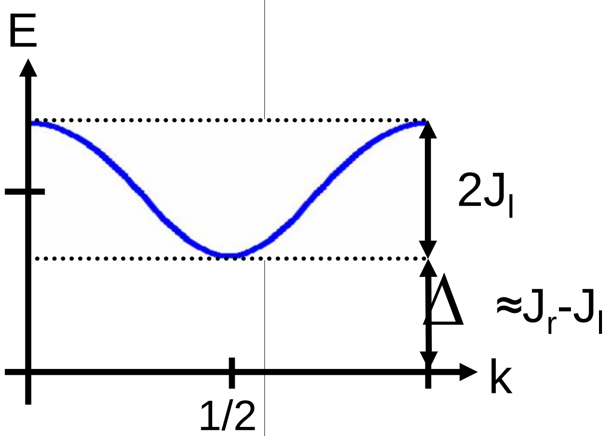

$Ba_2Cu(BO_3)_2$
Rüegg, HMR, Demmel *et al.*




$SrCu_2(BO_3)_2$
Zayed, Rüegg, HMR *et al.*

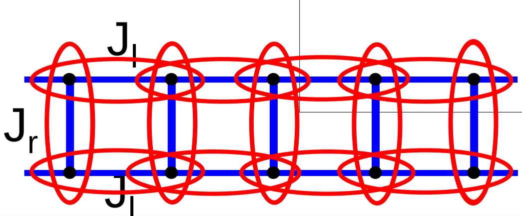
# The spin-ladder – array of spin pairs




$0\ T < H < H_{c1} = \Delta$

spin liquid phase




Perturbation from isolated rungs:

Ground state ≈ product of singlets

Excited states are triplets $t^{\pm}(r)$, $t^{0}(r)$

Leg coupling $J_l$ makes triplets move

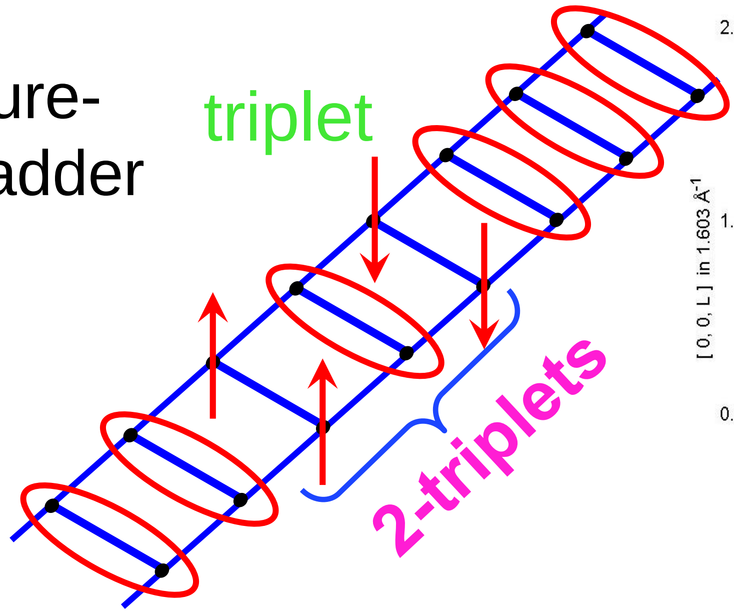
Create Bloch-waves of triplets t(k)

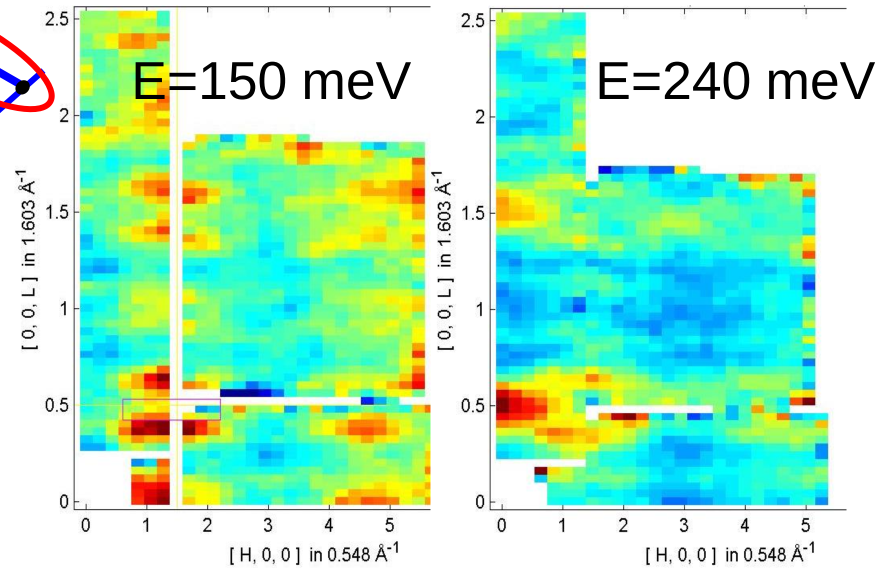
Dispersion $E_k = J_r + J_l \cos kd$

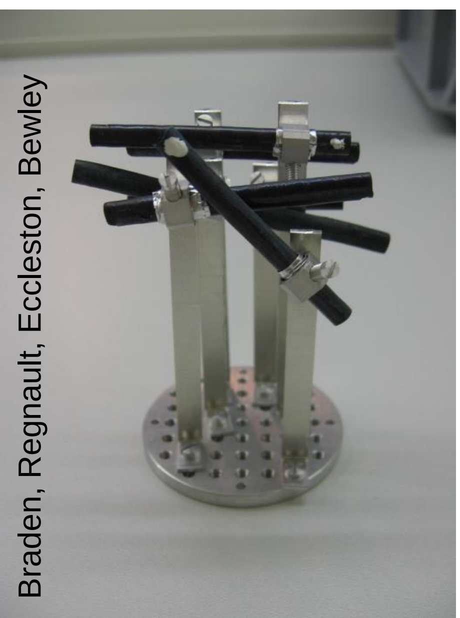
Real ground state has singlet fluctuations – renormalisation of above result

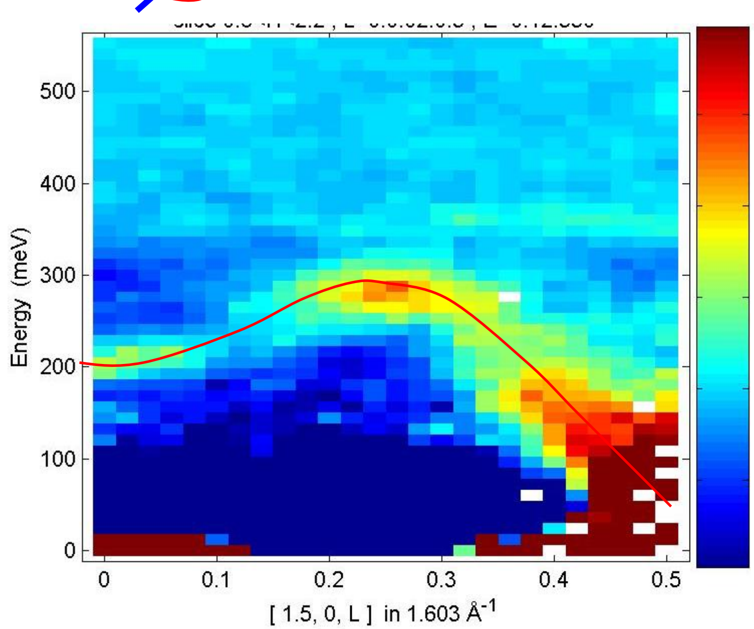
# 1- and 2-triplet dispersion in $Sr_{14}Cu_{24}O_{41}$

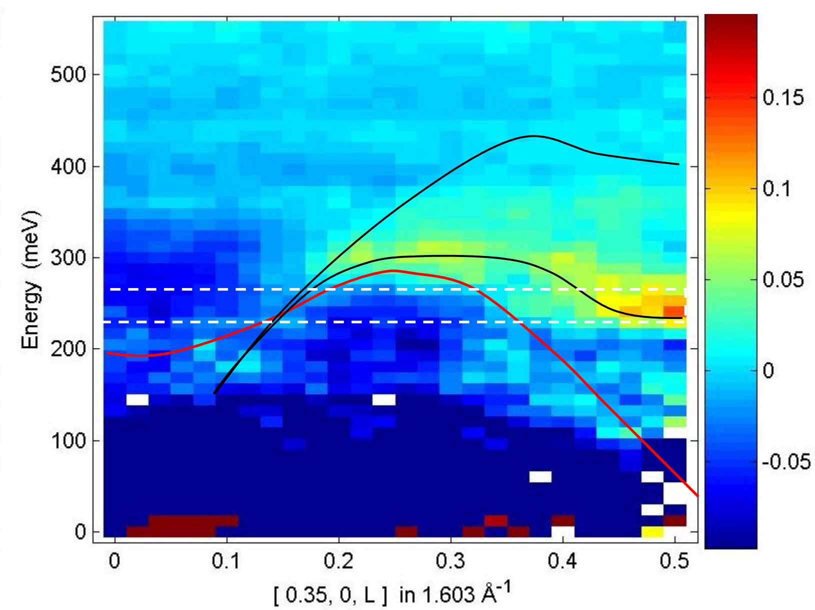
Different structure-factor perp to ladder



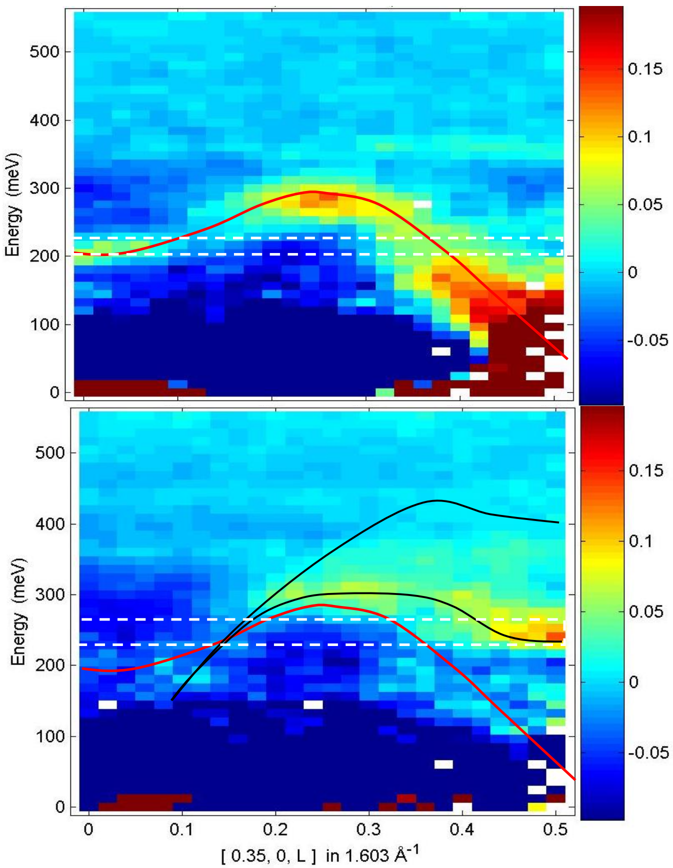
# Spin ladders

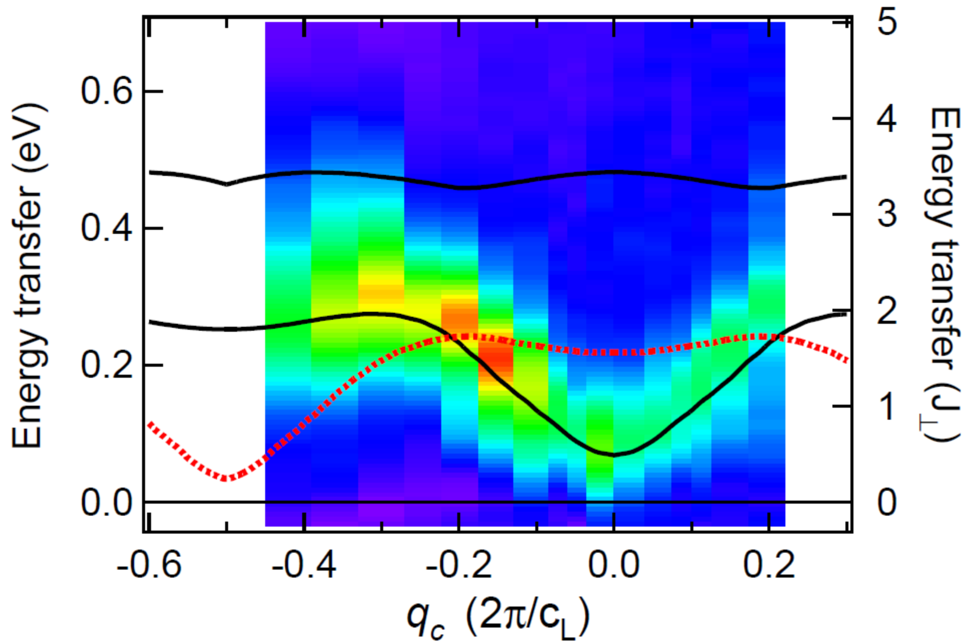
- $Sr14Cu24O41$: Cuprate ladders, 1- and 2-triplons

⇐Neutrons

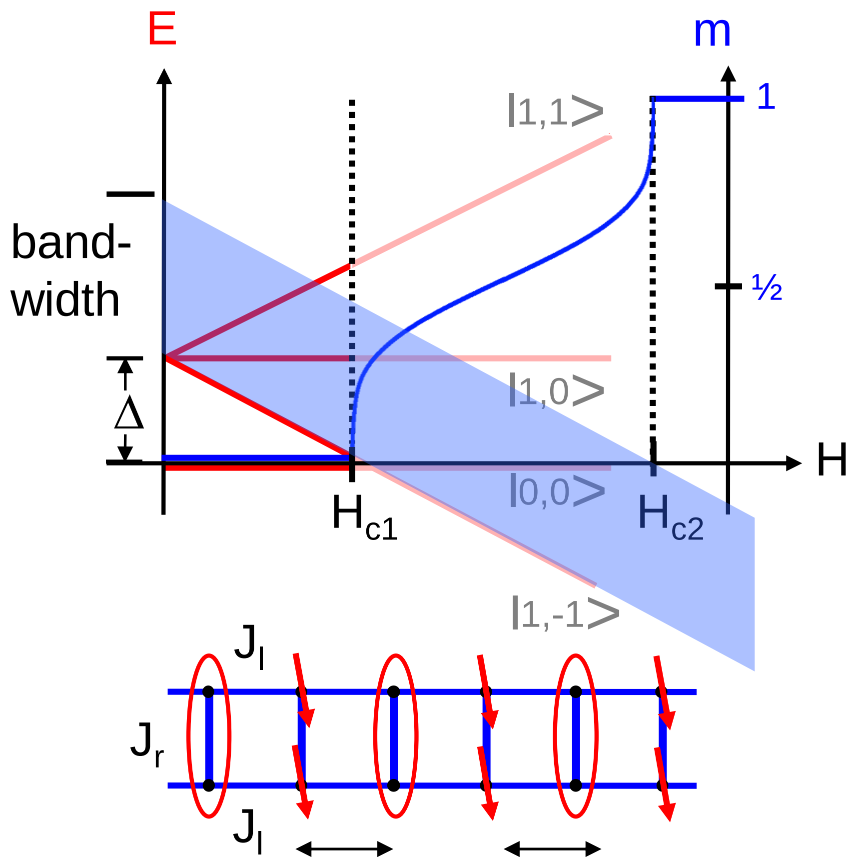
Resonant Inelastic X-ray Scattering

⇓ RIXS



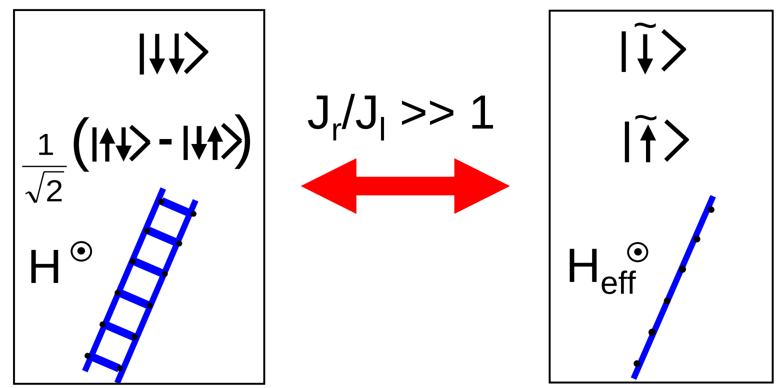



# Dynamics Luttinger-Liquid




$H_{c1} < H < H_{c2}$ : Luttinger liquid

## Ladder → Chain mapping




F. Mila, Eur. Phys. J. B 6 (1998)

no 3D long-range order
→unique 1D dynamics

Search Ch Ruegg et al.
for refs on beautiful ladder expts

# Quasi-particle zoo in one-dimension

## Electronic states of matter:

Metal / Semiconductor / Insulator } Single particle picture

Superconductors: Cooper-pairs, Majorana fermions } Correlated electron states
fractional Quantum Hall effect: fractional charges

## Magnetic states and excitations:

Magnetic order } semiclassical
spin-wave magnon excitations } single particle picture

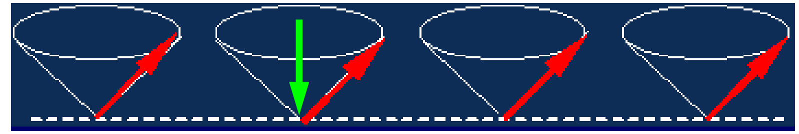
Quantum 'disordered' states (quantum spin liquids)

- Multi-magnon excitations } collective quantum states
- Fractionalized excitations

Possibly simplest example: 1D Heisenberg chain

- Analytic solution by Bethe in 1931: 'domain wall quantum soup'

# Ferromagnets are simple (classical)



$H=-\sum_{rr'} J_{rr'} S_r \cdot S_{r'} = -J \sum_{<r,r'=r+d>} S^z_r S^z_{r'} + \frac{1}{2}(S^+_r S^-_{r'} + S^-_r S^+_{r'})$

⇑ nearest neighbour ⇑

Ordered ground state, all spin up: $H|g\rangle = E_g|g\rangle$, $E_g=-zNS^2J$

Single spin flip not eigenstate: $|r\rangle = (2S)^{-1/2} S^-_r|g\rangle$, $S^-_r S^+_{r'}|r\rangle = 2S|r'\rangle$

$H|r\rangle=(-zNS^2J+2zSJ)|r\rangle - 2SJ\sum_d |r+d\rangle$ — flipped spin moves to neighbors

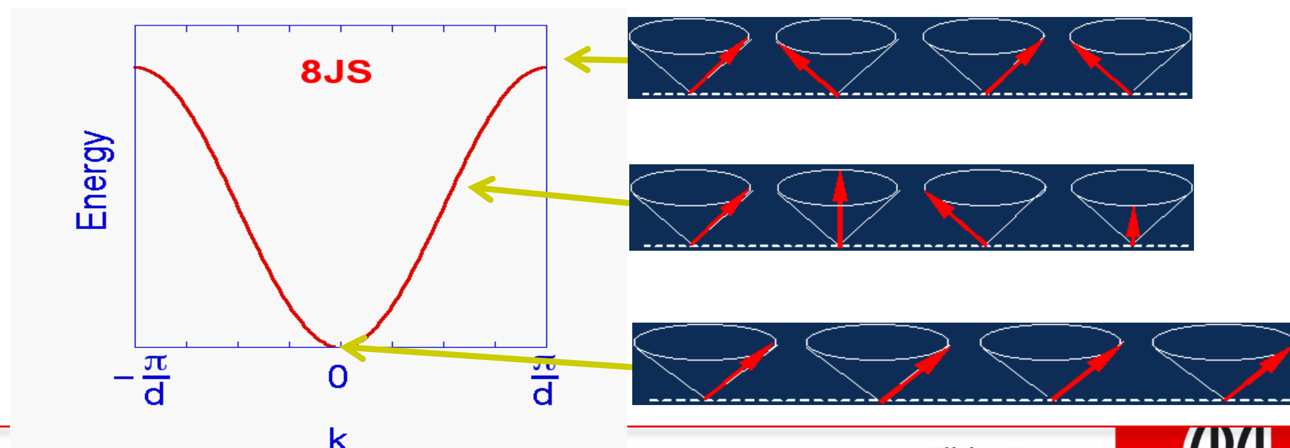
Periodic linear combination: $|k\rangle = N^{-1/2}\Sigma_r e^{ikr}|r\rangle$ — plane wave

Is eigenstate: $H|k\rangle = E_g+E_k|k\rangle$, $E_k=SJ\Sigma_d 1-e^{ikd}$ — dispersion = 2SJ (1-cos(kd)) in 1D

Time evolution: $|k(t)\rangle = e^{iHt}|k\rangle = e^{iE_k t}|k\rangle$ — sliding wave

Dispersion:
relation between
time- and space-
modulation period

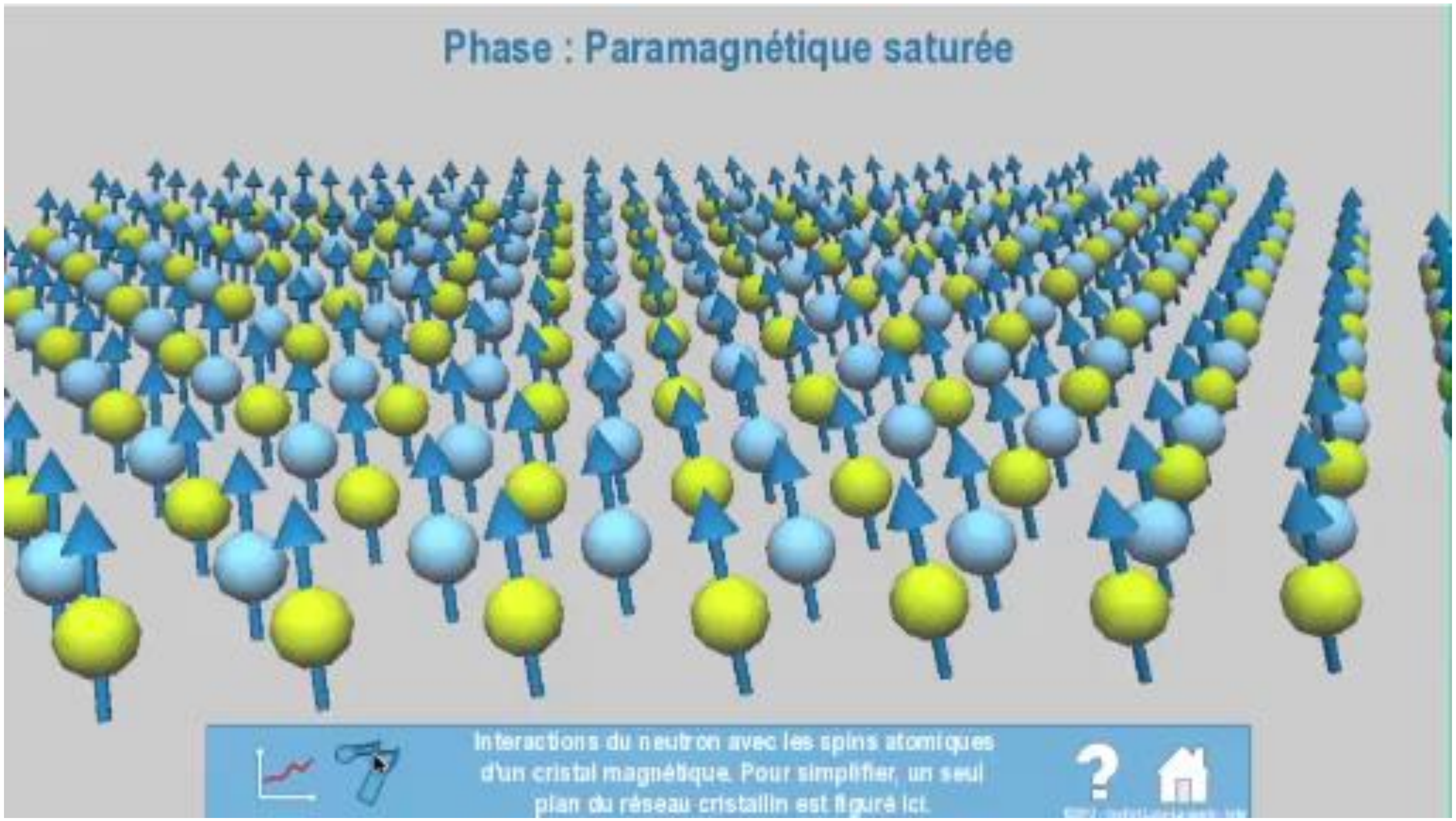
Same result in classical
calculation ⇒ precession:



# Ferromagnetic model is simple:

Solution: Spin waves $\Rightarrow$ sharp dispersion

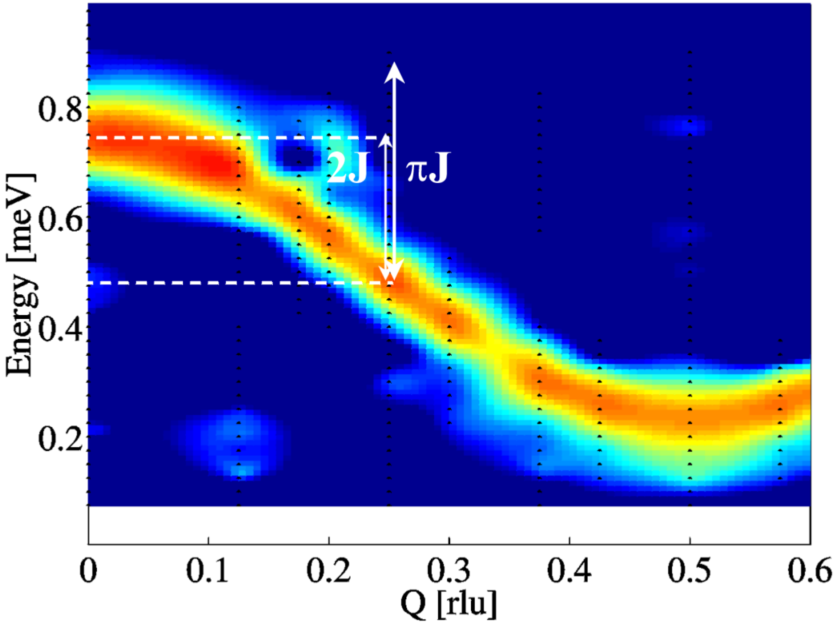
Picture: $\Downarrow$ easy cartoon




https://www.ill.eu/?id=11644

# Spin waves in a "ferromagnet"

$CuSO_4 \cdot 5D_2O$

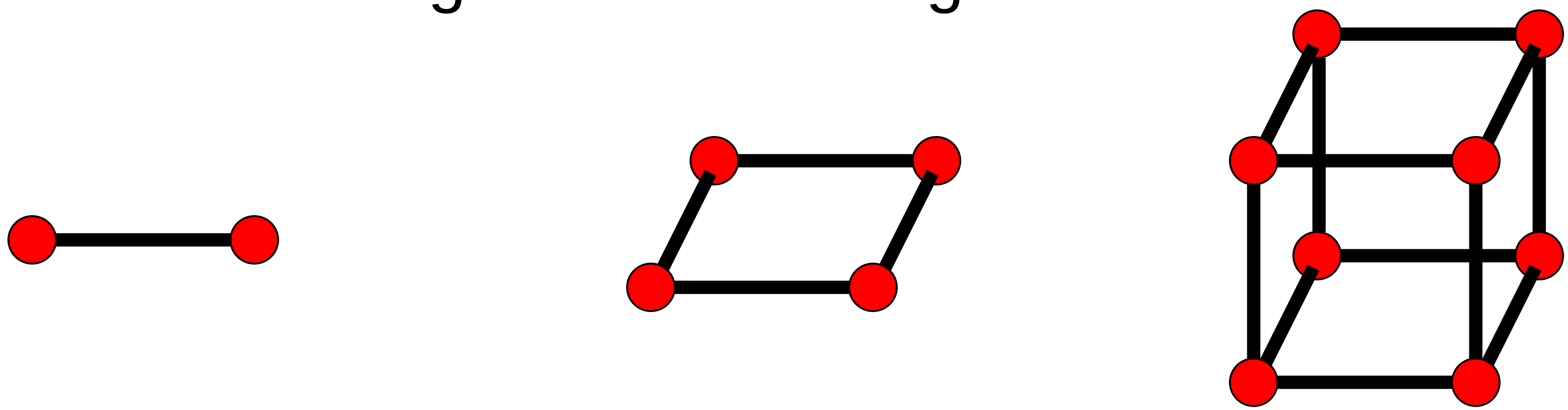



dispersion = 2SJ (1-cos(kd))

Actually it is an antiferromagnet polarized by 5T field

# Antiferromagnets are tricky

Fluctuations stronger for fewer neighbours



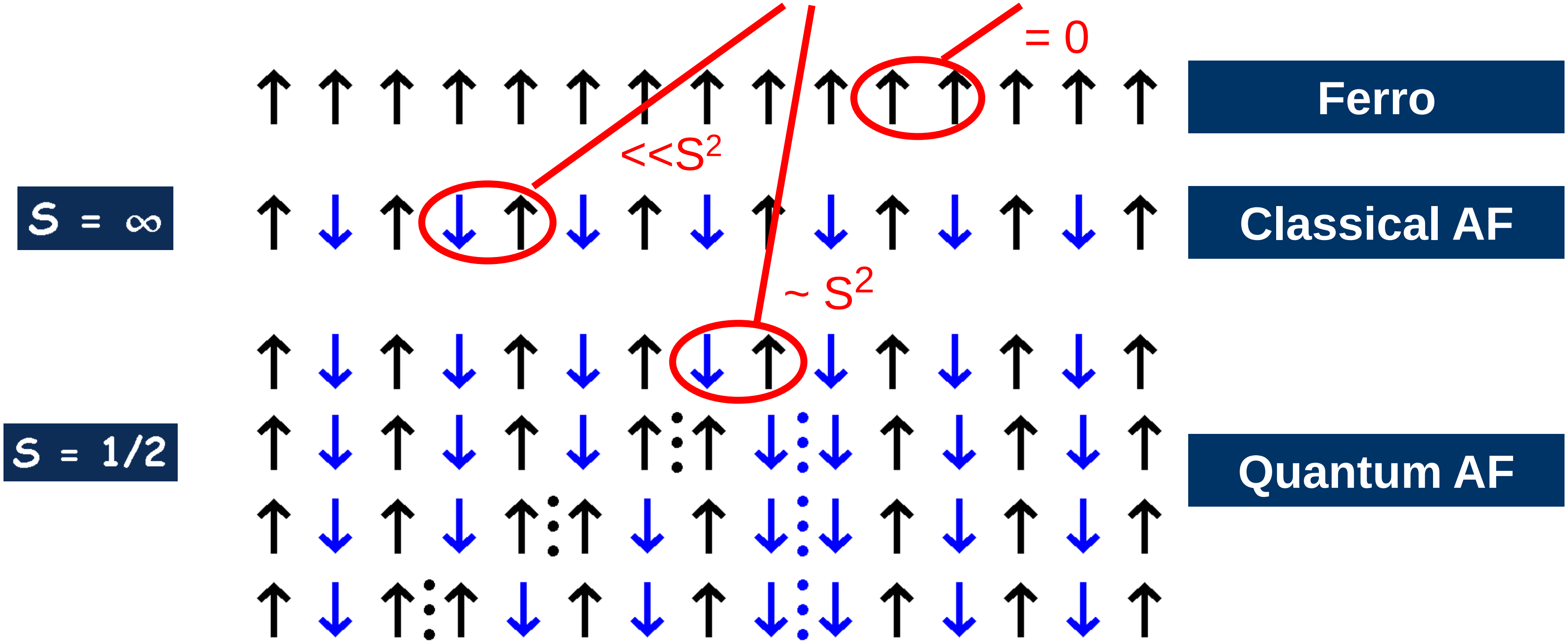
1D: Ground state ‘quantum disordered’ spin liquid of S=1/2 spinons. Bethe ansatz ‘solves’ the model

2D: Ground state ordered at T=0 <S> = 60% of 1/2 (although not rigorously proven).

3D: Ground state long range ordered, weak quantum-effects

# antiferromagnetic spin chain

$$\mathcal{H} = J \sum S^z_n S^z_{n+1} + \frac{1}{2}\left(S^+_n S^-_{n+1} + S^-_n S^+_{n+1}\right)$$

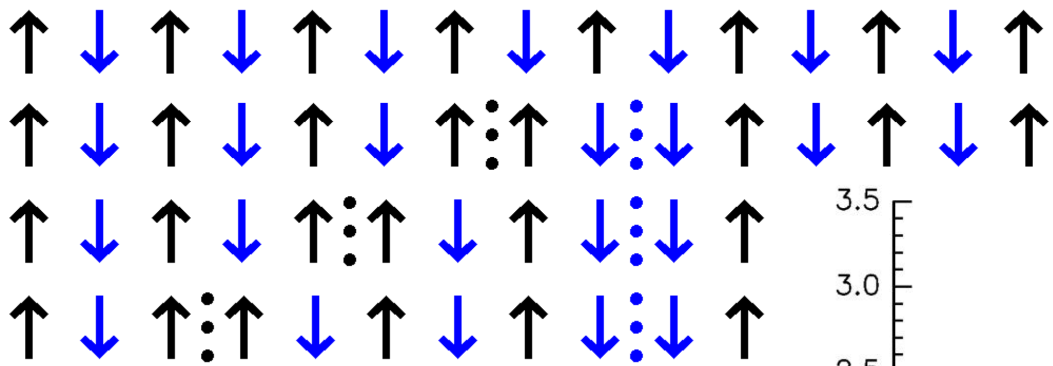



**Ground state (Bethe 1931) – a soup of domain walls**

# Spinon excitations

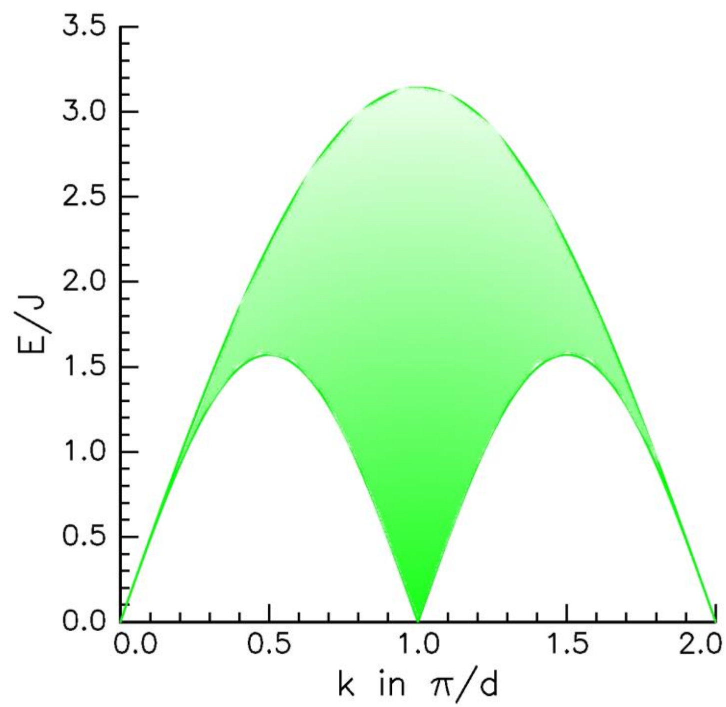
Elementary excitations:

- "Spinons": spin S = ½ domain walls with respect to local AF 'order'
- Need 2 spinons to form S=1 excitation we can see with neutrons



| | | | | |
|---|---|---|---|---|
| Energy: | $E(q) =$ | $E(k_1)$ | $+$ | $E(k_2)$ |
| Momentum: | $q =$ | $k_1$ | $+$ | $k_2$ |
| Spin: | $S =$ | ½ | ± | ½ |

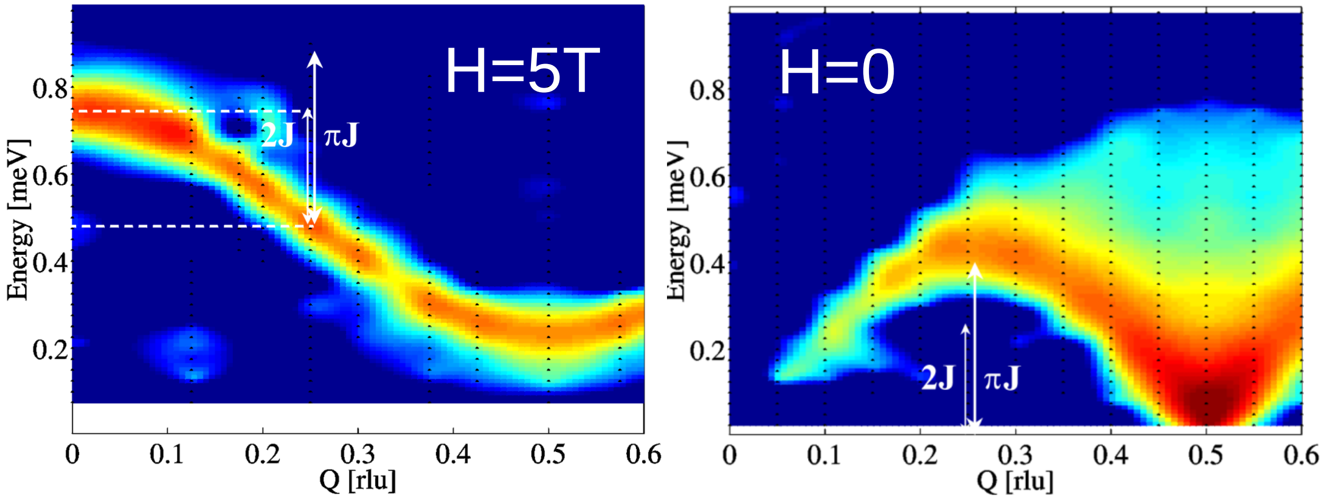
Continuum of scattering ⇒

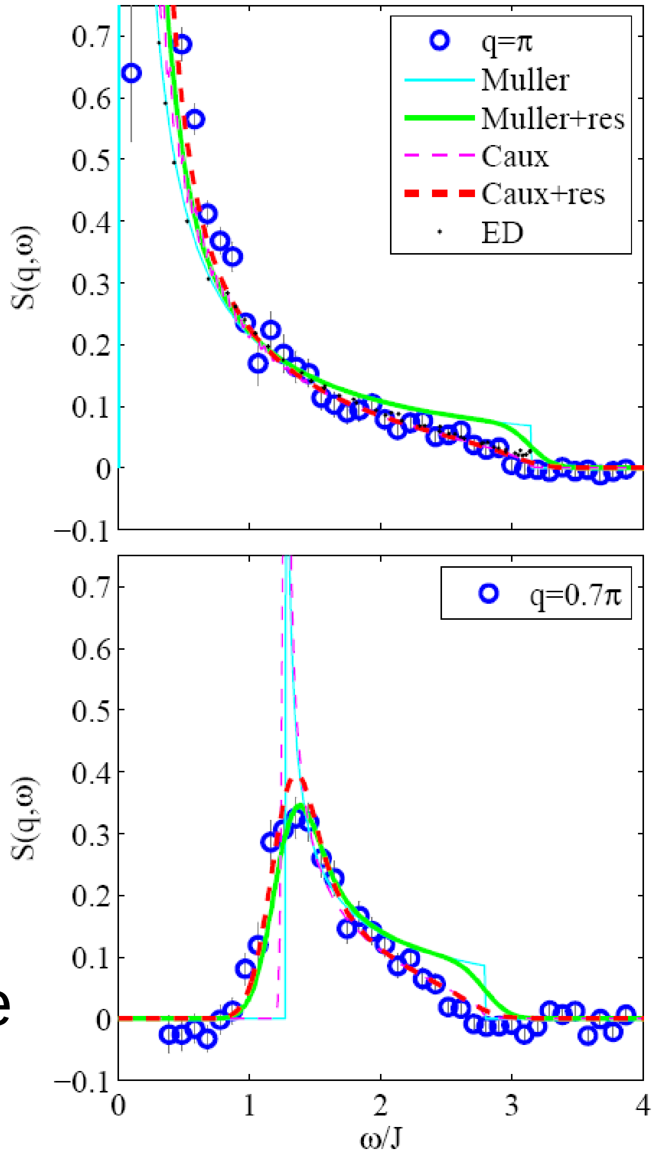


# The *antiferromagnetic* spin chain

FM: ordered ground state (in 5T mag. field)
- semiclassical spin-wave excitations

AFM: quantum disordered ground state
- Staggered and singlet correlations
- Spinon excitations



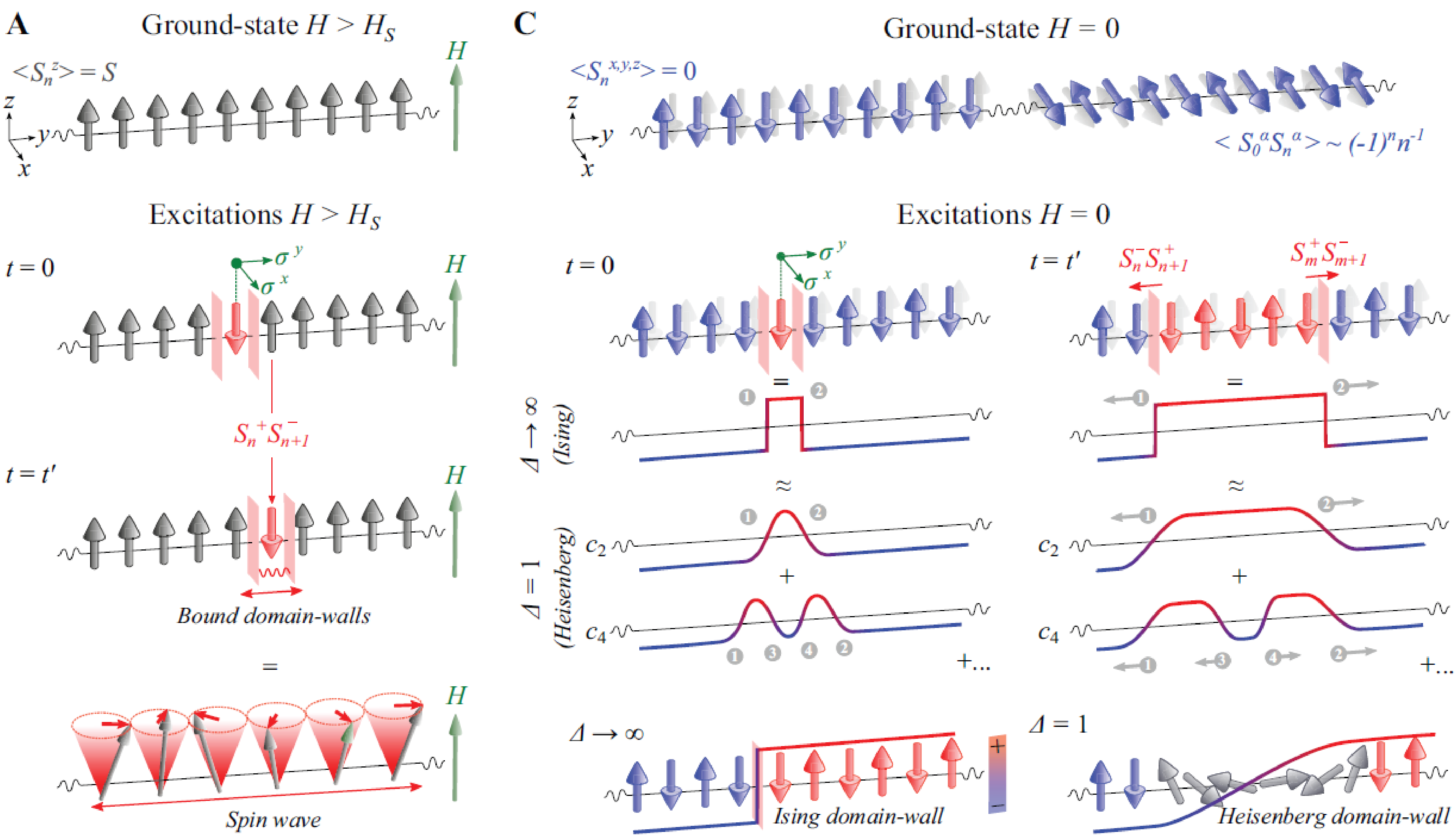

– Agebraic Bethe ansatz for inelastic lineshape

– Beyond Müller-conjecture ⇒

Mourigal, Enderle, HMR, Caux

# Spinons – our cartoon for excitations in 1D spin chain

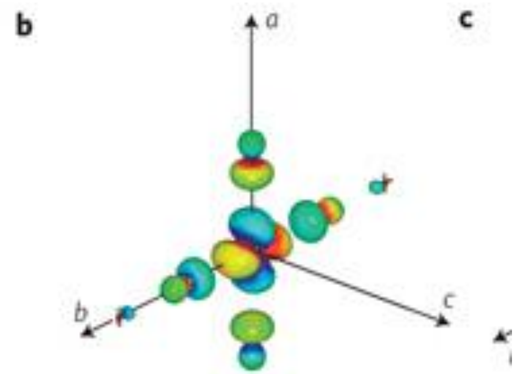
## Spin waves — Spinons: 2- and 4 spinon states ?

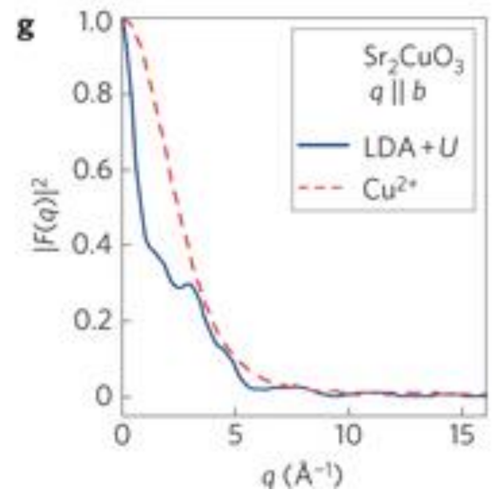


# Detecting 4-spinon states?

- Neutrons see spinon continuum
- But, 2- and 4-spinon continuum almost identical line-shape
- Only way to distinguish is absolute amplitude
- Previous attempts, covalency etc.

- Trick: Normalise to ferromagnetic spin-waves



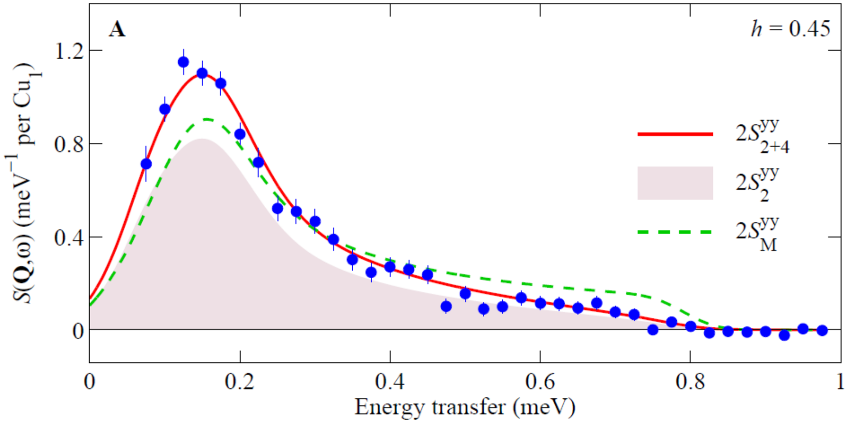

Intensity = instrument-stuff * cross-section

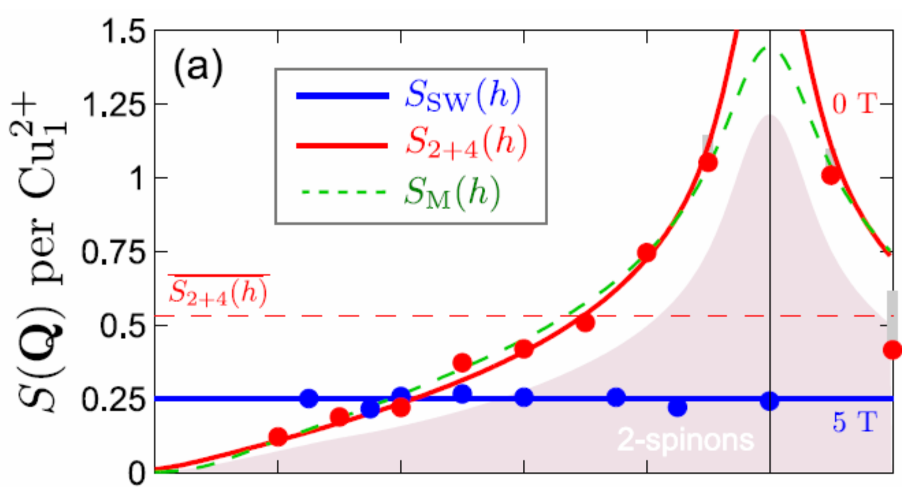
$$\left(\frac{d^2\sigma}{d\Omega dE_f}\right)_{\rm mag} = \frac{(\gamma r_0)^2}{2\pi\hbar}\frac{k_f}{k_i}\sum_{\alpha\beta}(\delta_{\alpha\beta}-\hat{Q}_\alpha\hat{Q}_\beta)|gF_R(Q)|^2\sum_{RR'}\int dt e^{iQ(R-R')-i\omega t}\langle S_R^\alpha(0)S_{R'}^\beta(t)\rangle$$

cross-section | pre factor | dipole factor | magnetic form factor | Fourier transform | spin-spin correlation function

# 4- spinon states:

- 2-spinons 72.9%, 4-spinons 25+-1%, 6-spinon ?




- Normalising to FM intensity, we account for 99% of the sum rule
- Comparing to Caux et al, this corresponds to 74% 2-spinon
- Physical picture ⇒ dominant states have one "dispersing" spinon and n-1 around zero energy

(in a string of Bethe numbers – a bit complicated)

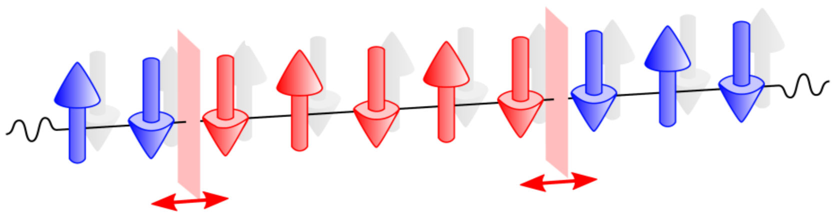
- Possible combinatorial arguments?

  Interestingly: 2^(n/2)/(n-1)!
  ⇒ [73.1%, 24.4%, 2.4%, 0.1% …

Mourigal *et al.* Nat Phys **9**, 435 (2013)

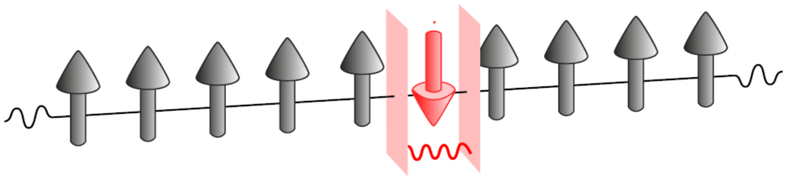
# Intermediate fields – a teaser

- ❑ H = 0 (Spinon vacuum)



2- + 4- spinon

- ❑ H > $H_s$ (Magnon vacuum)



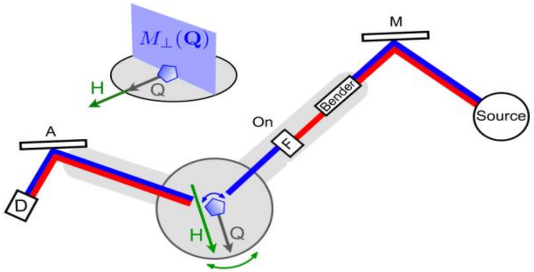
1- magnon

- ❑ 0< H < Hs (finite spinon population) $\mathcal{S}^{+-} \neq \mathcal{S}^{-+}$

**What are the excitations in intermediate field ?**

- ❑ Psinons **ψ** and anti-psinons **ψ*** [Karbach *et al.*, PRB 1997]
- ❑ + « String solutions » [Caux *et al.*, PRL 2005; Kohno, PRL 2009]

# Polarised neutron scattering ⇒ Several new quasi-particles observed




$$\sigma_{x0} \propto \mathcal{S}^{-+} + \sigma_{\rm si}$$

$$\sigma_{\bar{x}0} \propto \mathcal{S}^{+-} + \sigma_{\rm si}$$

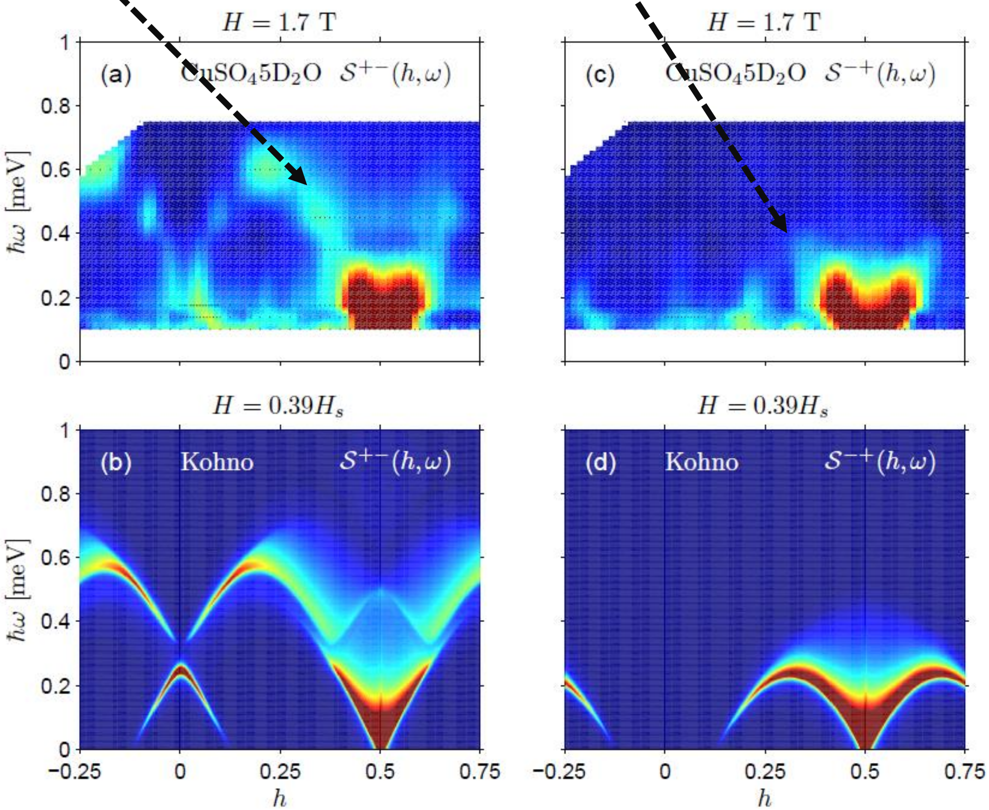
- more expt planned
- picture of these new excitations ?

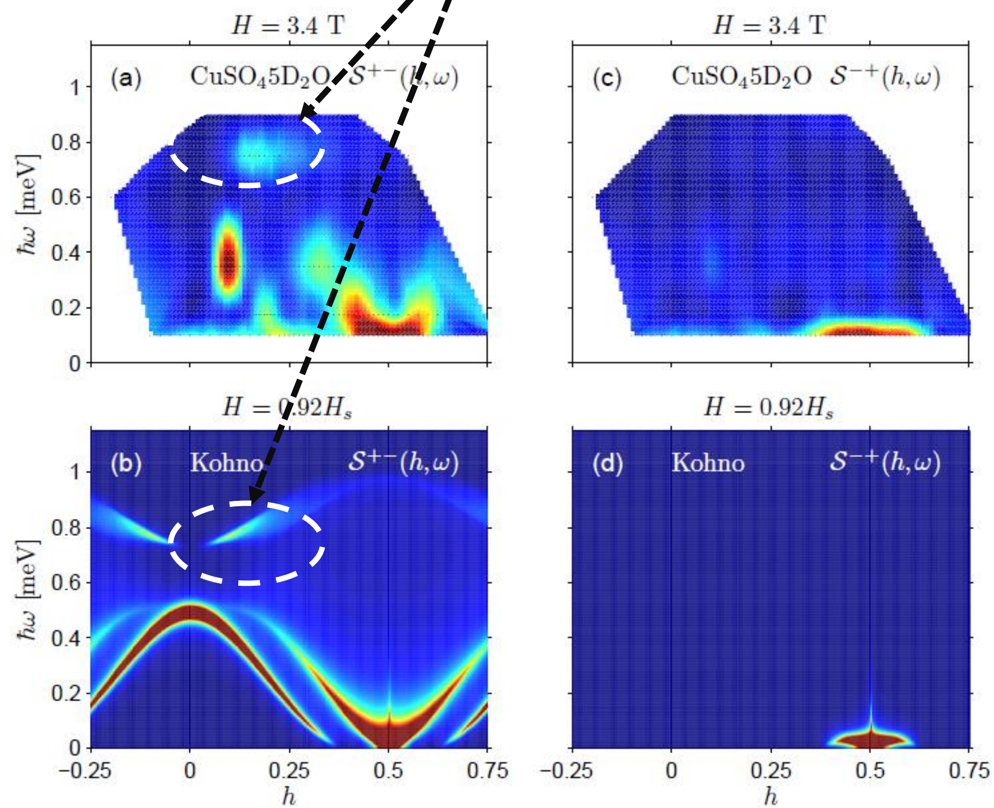
$\sigma_2$ (2-string) ?

[Kohno, PRL 2009]

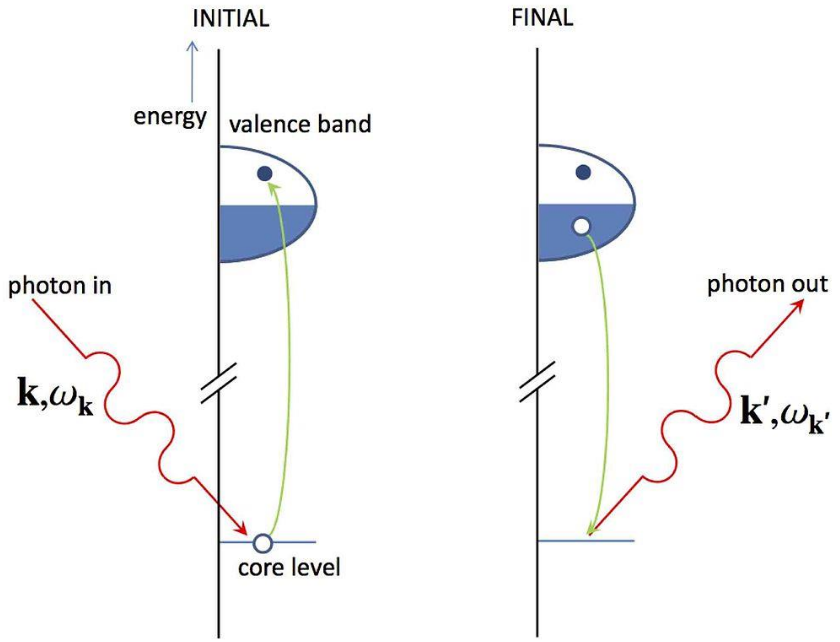
$\psi\psi^*$ in $\mathcal{S}^{+-}$

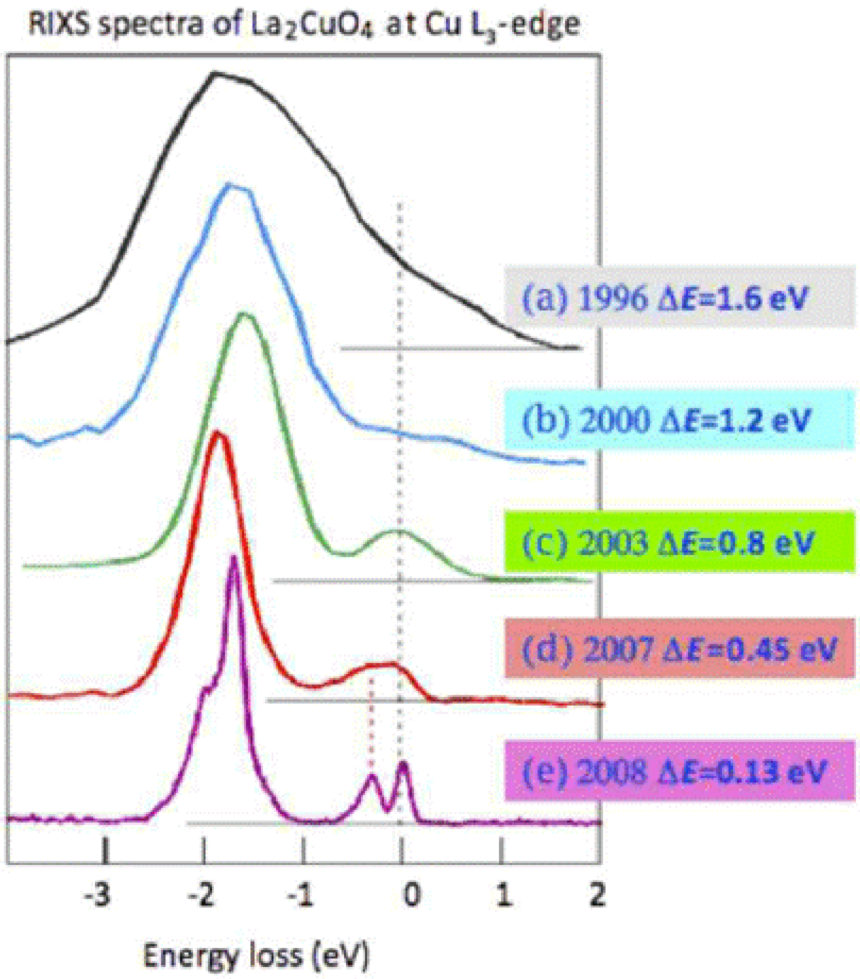
$\psi\psi$ in $\mathcal{S}^{-+}$

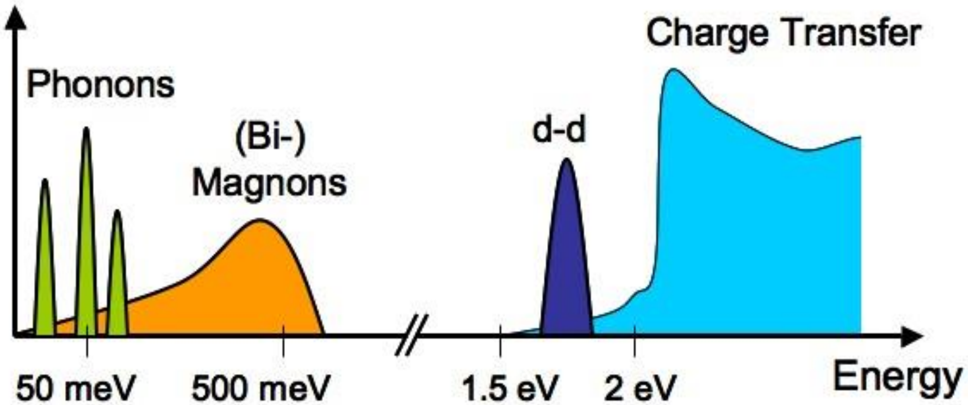


# Resonant Inelastic X-ray scattering



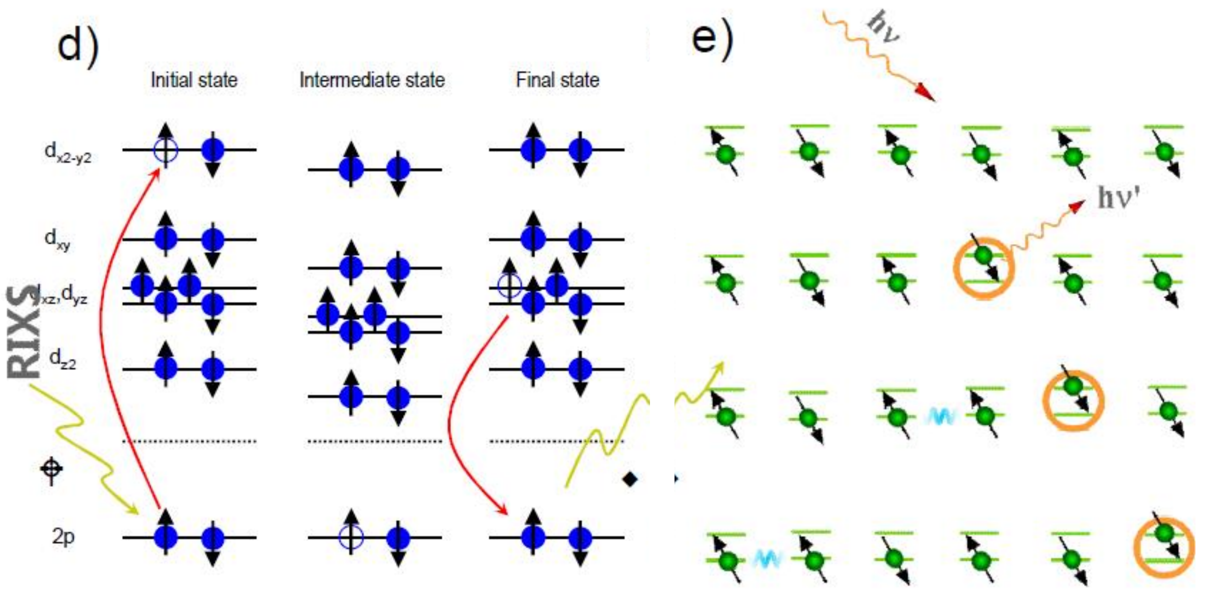



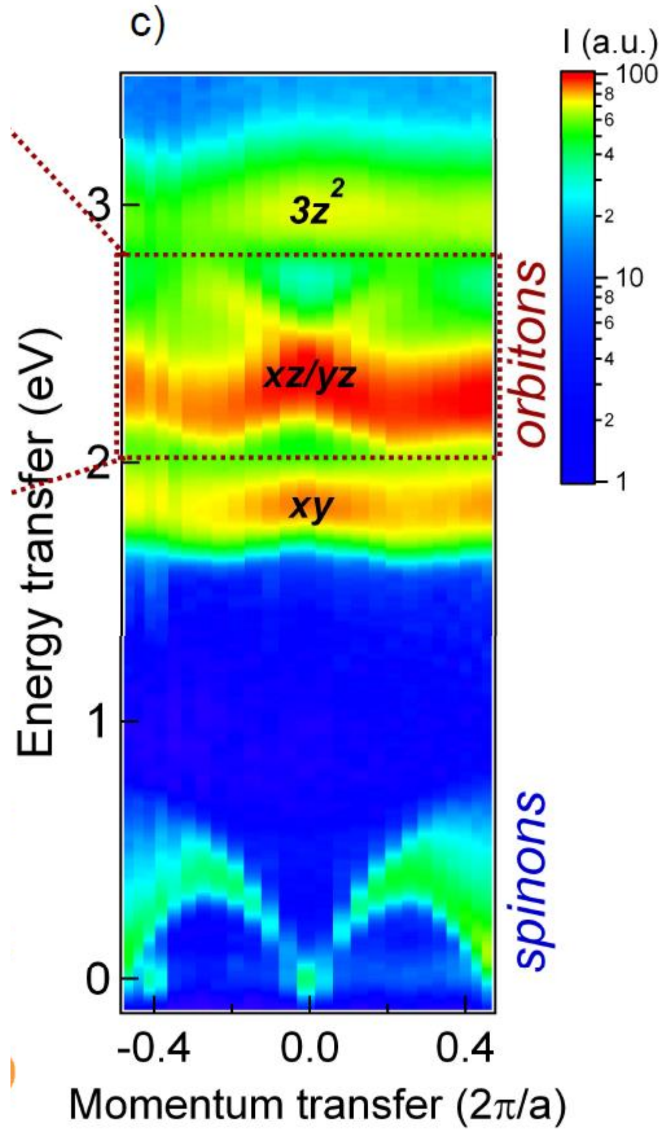

New measure of magnetic excitations

# RIXS and new correlation functions

$Sr_2CuO_3$ Much higher energy scale

- Resonant Inelastic X-ray scattering
  - Sees both magnetic and orbital excitations
  - Dispersive ‘orbitons’
  - Spinon-orbiton separation




J. Schlappa *et al.*, Nature **485**, 82 (2012)

# Quasi-particle zoo in one-dimension

## Electronic states of matter:

Metal / Semiconductor / Insulator } Single particle picture

Superconductors: Cooper-pairs
fractional Quantum Hall effect: fractional charges } Correlated electron states

## Magnetic states and excitations:

Magnetic order
spin-wave magnon excitations } semiclassical single particle picture

Quantum ‘disordered’ states (quantum spin liquids)

- Multi-magnon excitations
- Fractionalized excitations

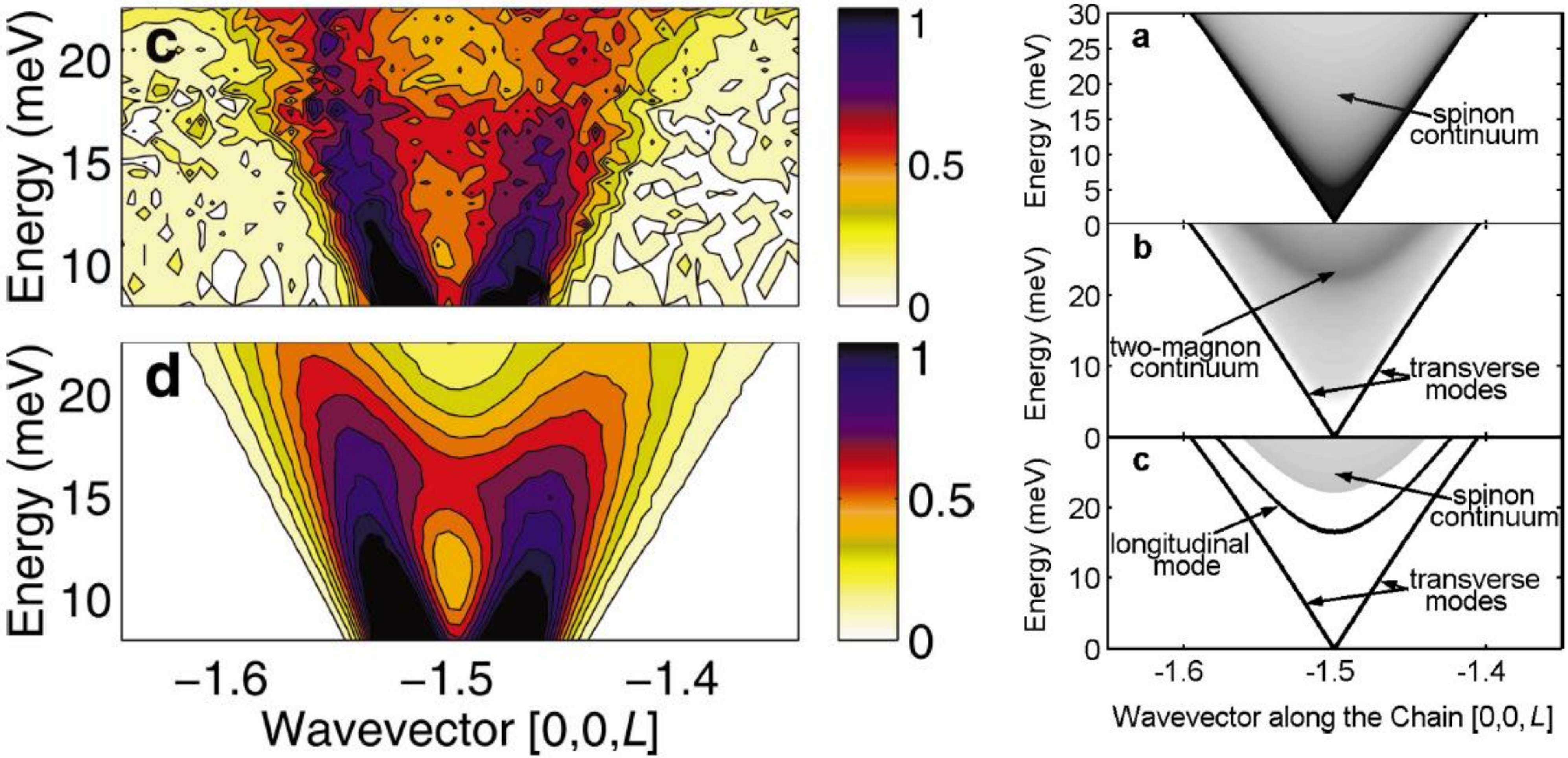
} collective quantum states

Possibly simplest example: 1D Heisenberg chain

- Analytic solution by Bethe in 1931: ‘domain wall quantum soup’

# Quantum heritage in ordered state

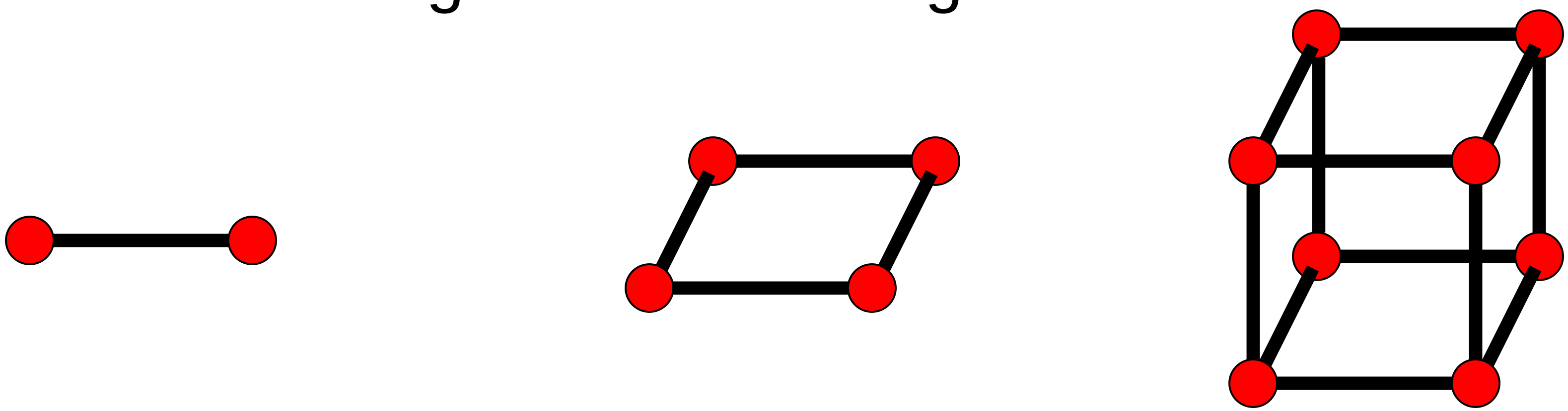
Can we have both 'classical' and 'quantum excitations?




Longitudinal mode below Tn in AFM chain: $KCuF_3$ Lake, Tenant et al. PRB **71** 134412 (2005)

# The 2D borderline

Fluctuations stronger for fewer neighbours



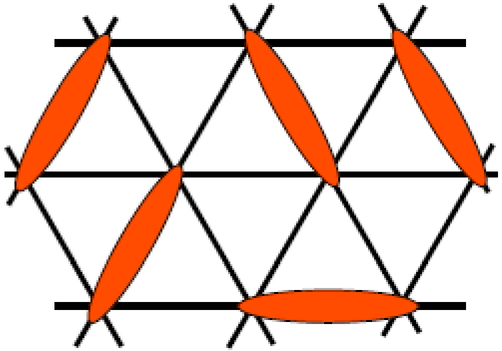
1D: Ground state ‘quantum disordered’ spin liquid of S=1/2 spinons. Bethe ansatz ‘solves’ the model

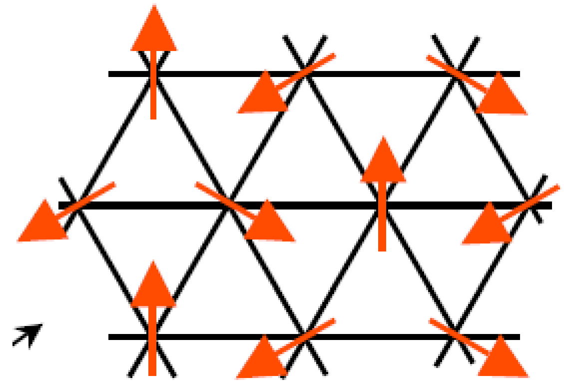
2D: Ground state ordered at T=0 <S> = 60% of 1/2 (although not rigorously proven).

3D: Ground state long range ordered, very weak Q-effects

# Valence Bonds and Anderson

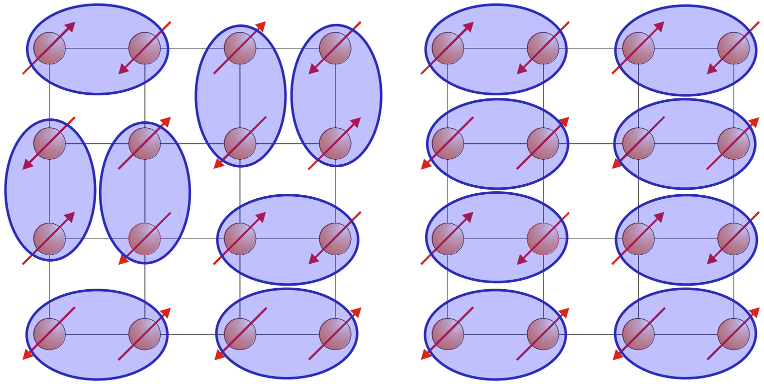
- 1973: Anderson suggests RVB on triangular lattice

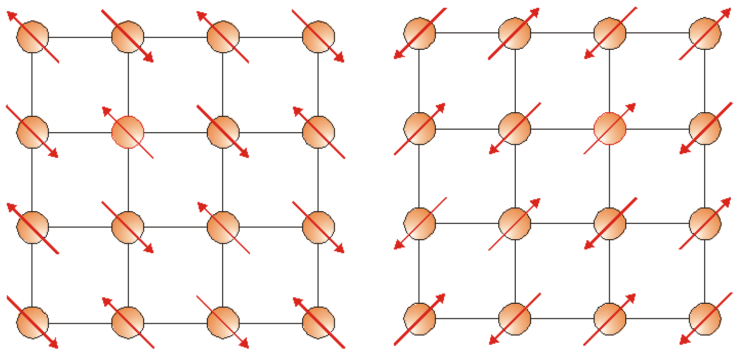




But - actually long range order

- 1987: Anderson suggests RVB on square lattice
  (as precursor and glue for High-Tc Superconductivity)





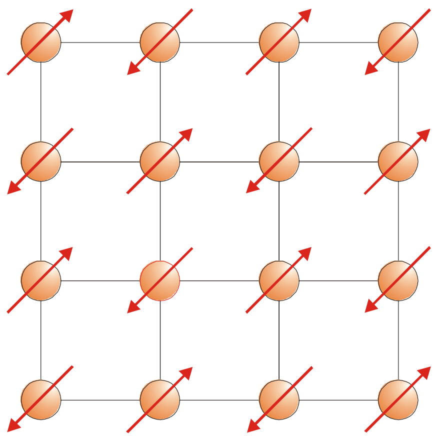
But - actually long range Neel order

# Quantum Magnetism in Flatland

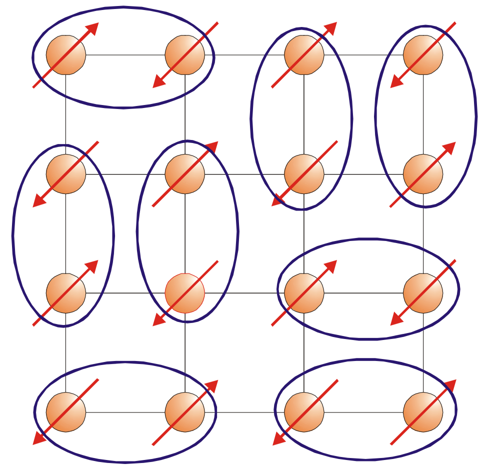
## 2D Heisenberg antiferromagnet on a square lattice

**Louis Neel :**
**Long-range**
**‘Néel’ Order**

$\langle S \rangle = 1/2$



**v. s.**



**Phil Anderson:**
**Spin-liquid**
**Resonating**
**Valence Bond**
**(RVB)**

$\langle S \rangle = 0$

2D: ordered, but only 60% of full moment, and only at T=0

$\Downarrow$ Spin-waves

$\Uparrow$ Quantum fluctuations

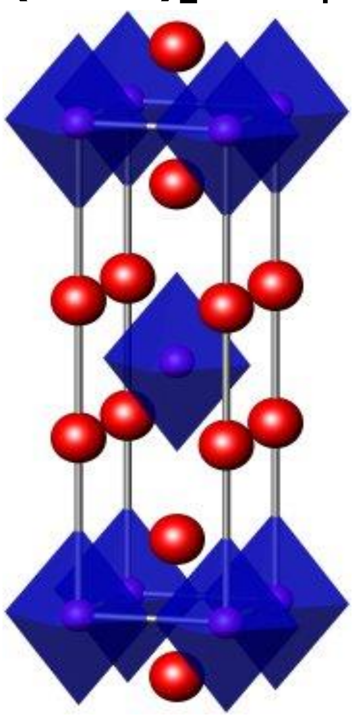
- Are there other types of ‘correlations’ ?
  - Resonating valence bonds (RVB)

} Investigate excitations with neutron scattering

# Physical realisations

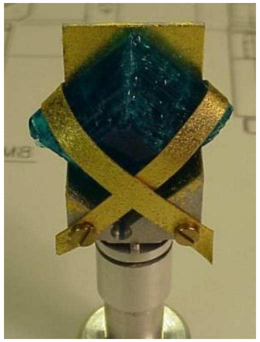
- Representation of model: No/small extra terms, anisotropy gaps etc.
- Energy scale: Zone boundary, resolution, temperature, field $H_s$

**$(La,Sr)_2CuO_4$**



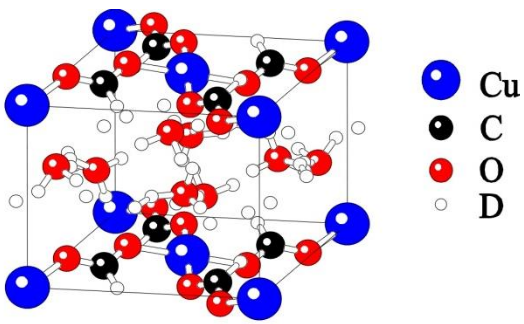
**$CuO_2$ planes**

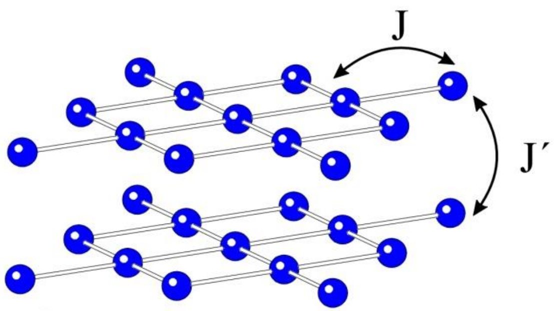
| | $La_2CuO_4$ | CFTD | CAPCuBr | CAPCuCl |
|---|---|---|---|---|
| J [K] | 1500 | 73.3 | 8.5 | 1.2 |
| J'/J | $5 \times 10^{-5}$ | $4 \times 10^{-5}$ | ~ 0.1 | ~ 0.1 |
| $T_N$ [K] | 325 | 16.4 | 5.1 | 0.64 |
| $H_S$ [T] | 4500 | 220 | 24 | 3.4 |



+ CuPzClO Tsyrulin & Kenzelmann

**Copper Formate Tetra-Deuterate**



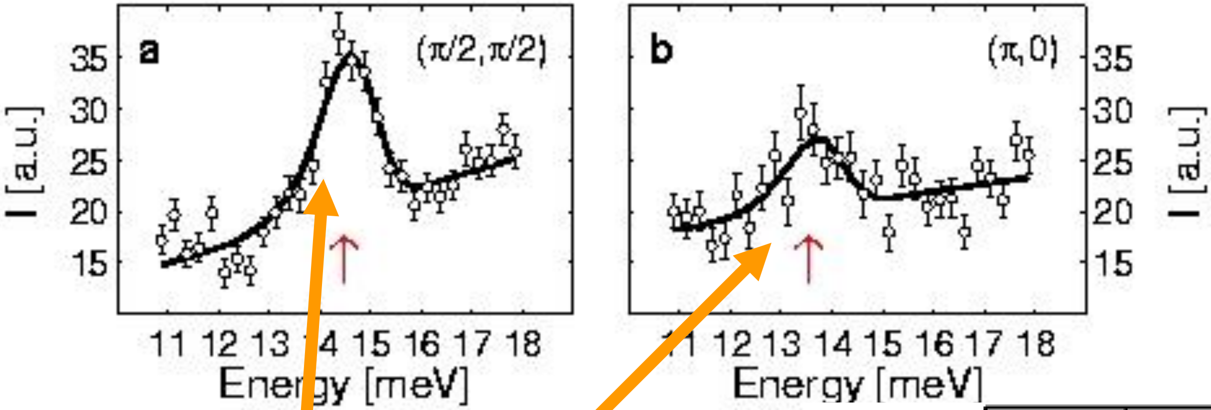



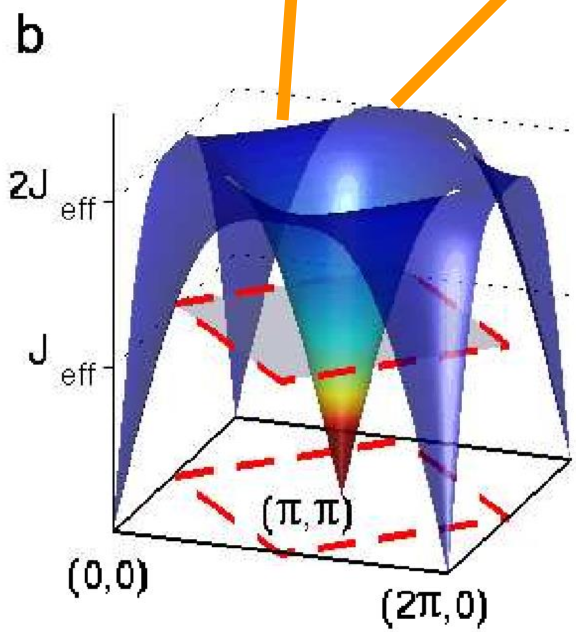

**$Cu(DCO_2)_2 \cdot 4D_2O$**

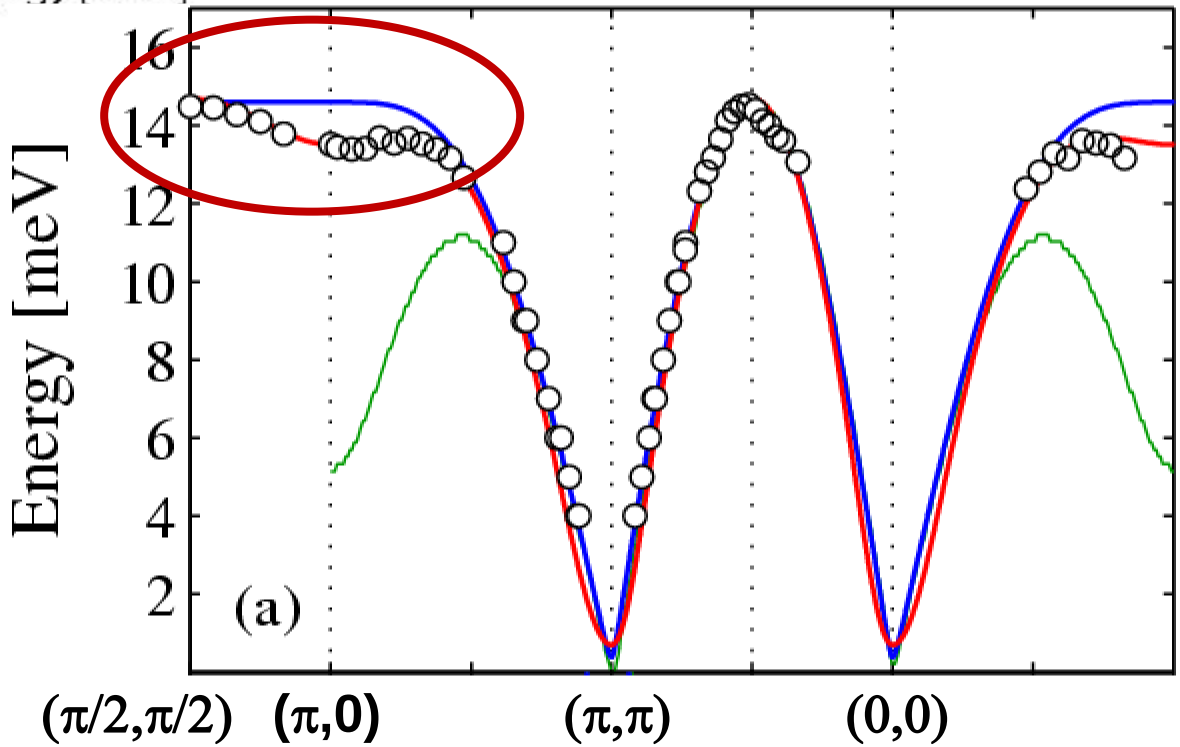
# 2D ordered ⇒ spin-waves – problem solved ?
# Surprise: zone boundary anomaly!

Zone boundary dispersion:
7 ± 1 % lower energy at (π,0) than (π/2, π/2)

A quantum effect



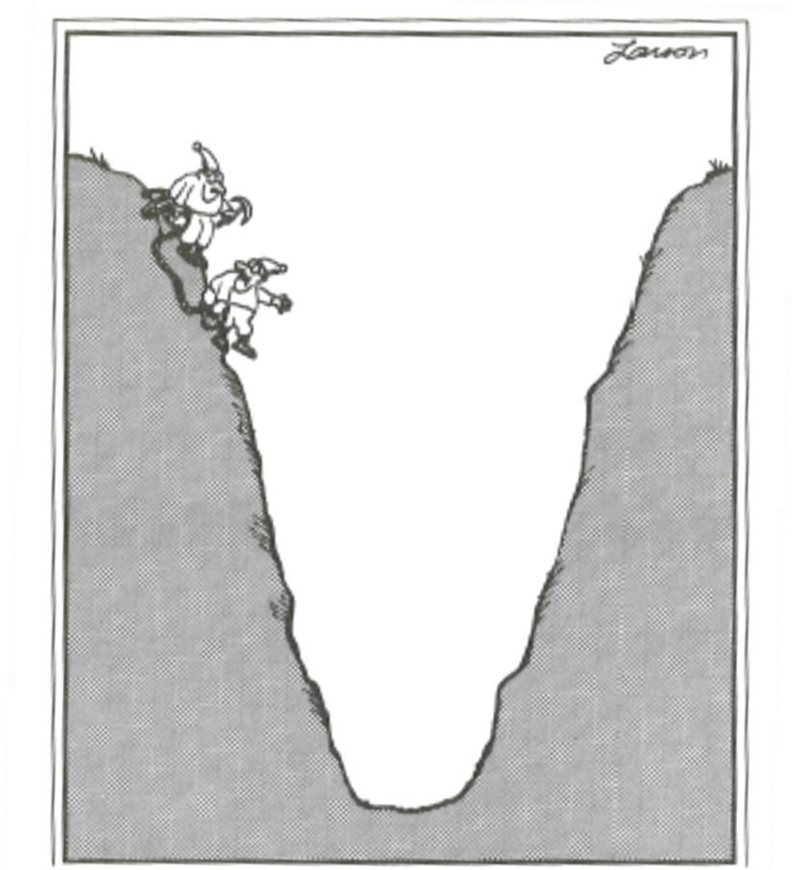



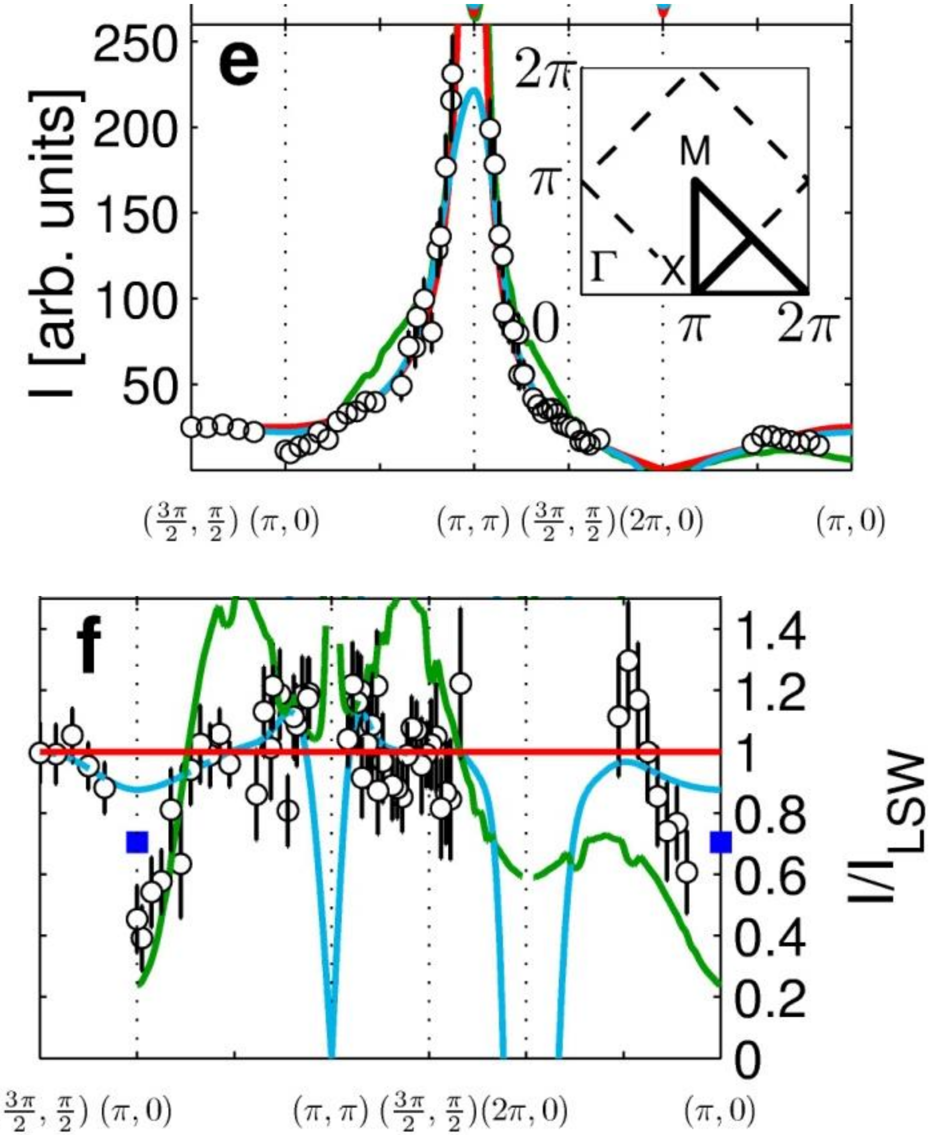
# Magnon intensities

Giant 50% intensity effect at (π,0)

Remember SW already 51% reduced

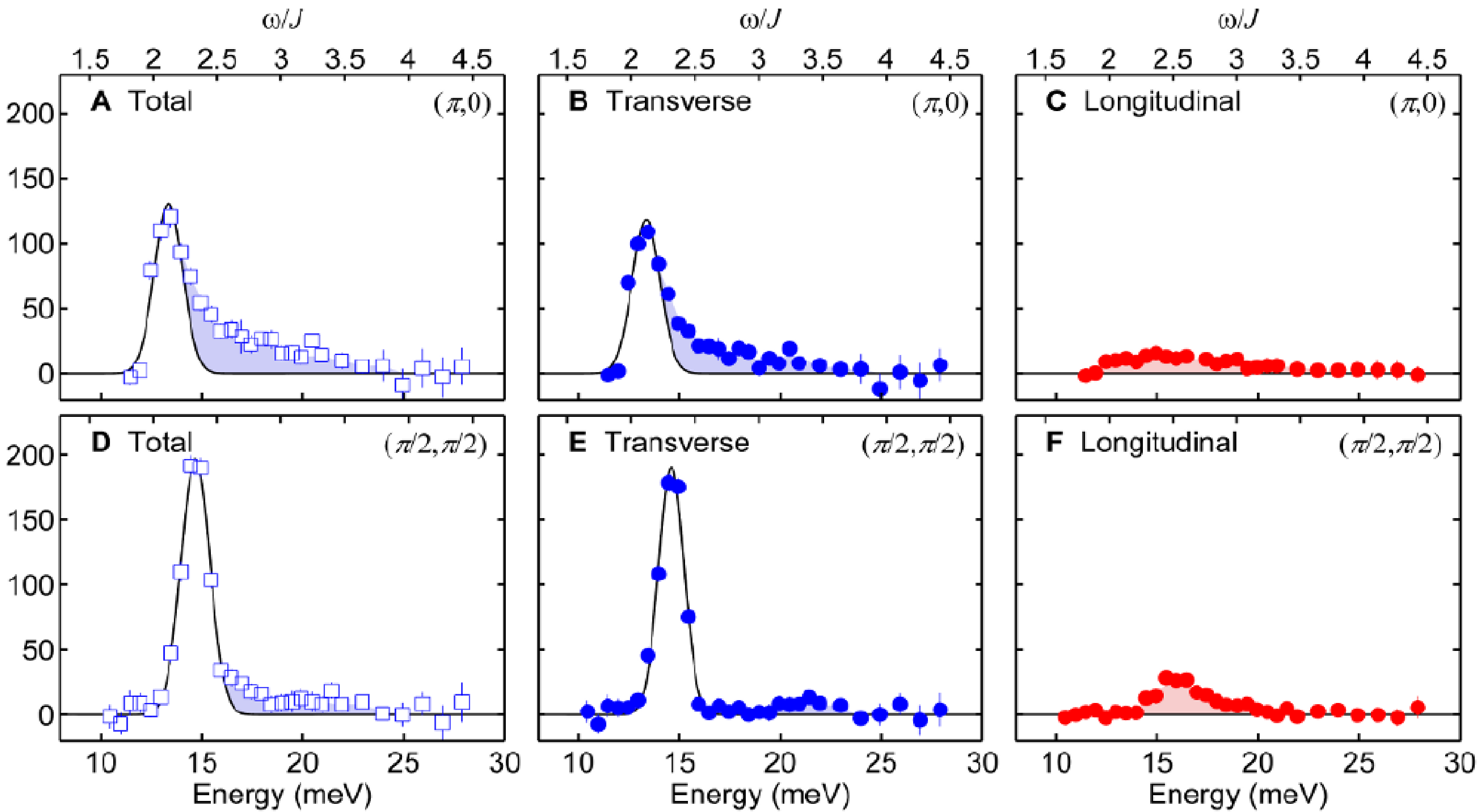
⇒ A tale of missing intensity !




Christensen PNAS **104** 15264 (2007)

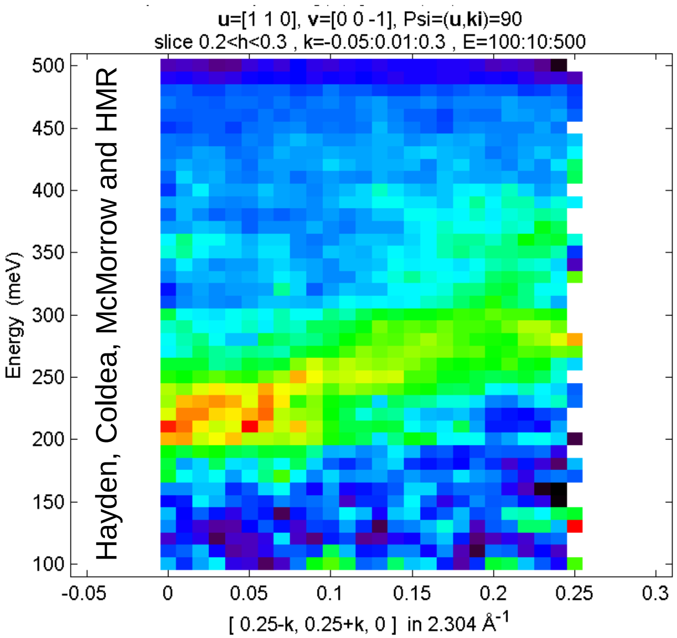
# Polarised neutrons: Line-shapes at the Zone Boundary



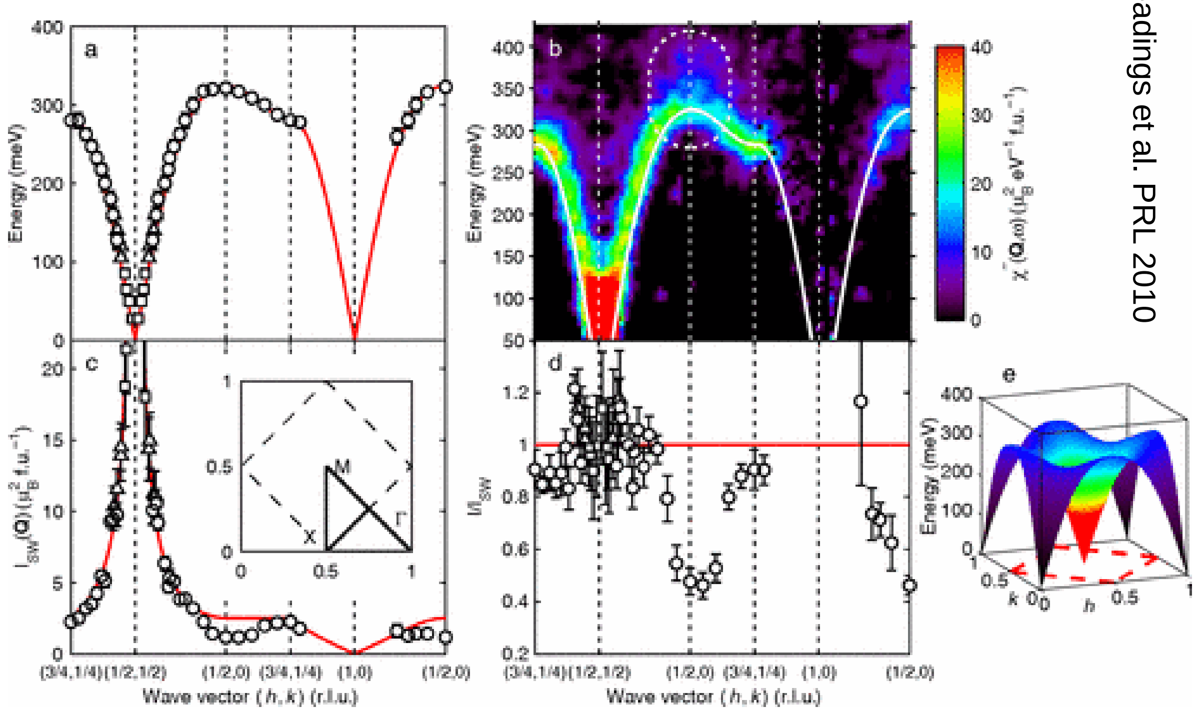

Both longitudinal and transverse continuum

# quantum anomaly also in cuprates !

## $YBa_2Cu_3O_{6.1}$



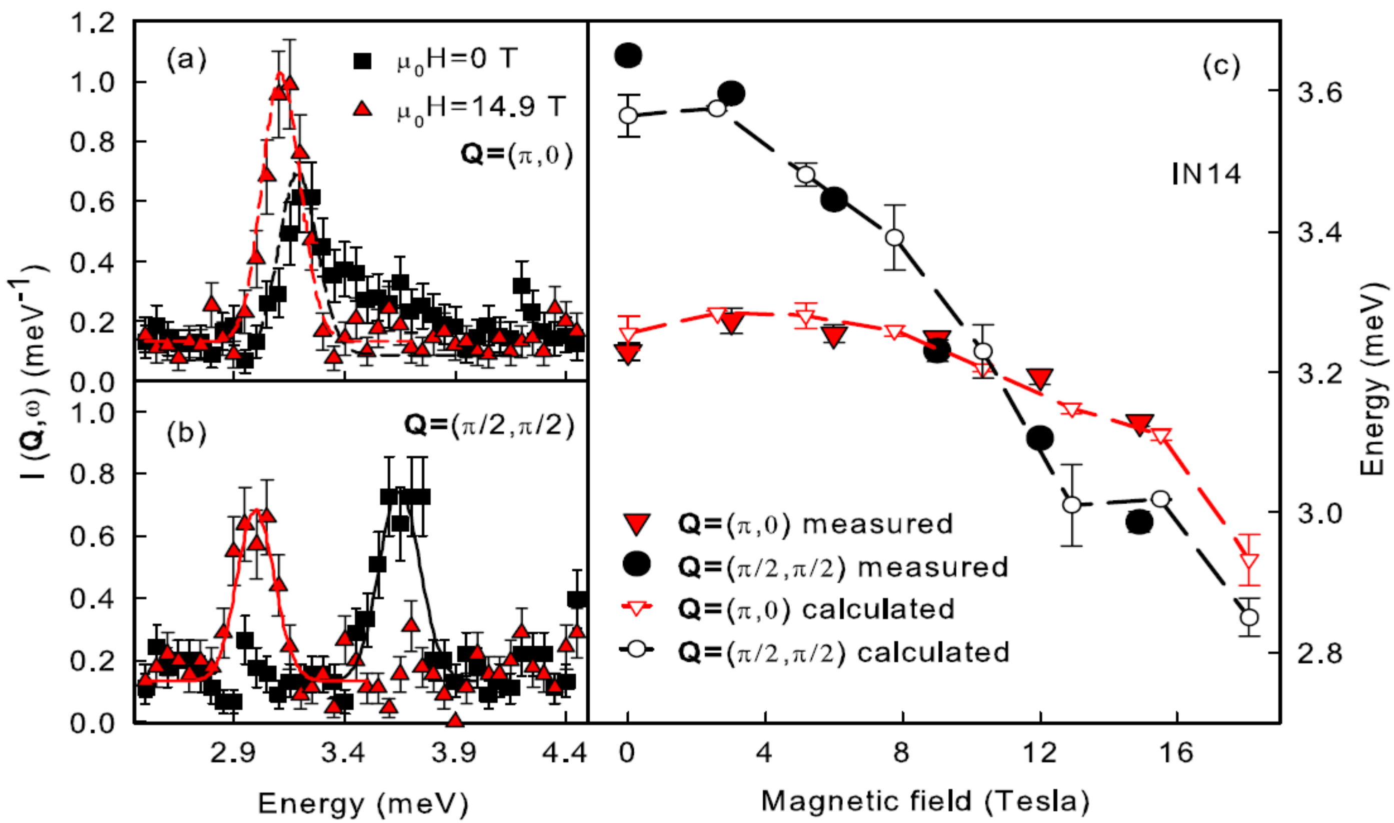

## La2CuO4




Headings et al. PRL 2010

Cuprates have different ZB dispersion due to further neighbor exchange interactions – also known as Hubbard heritage

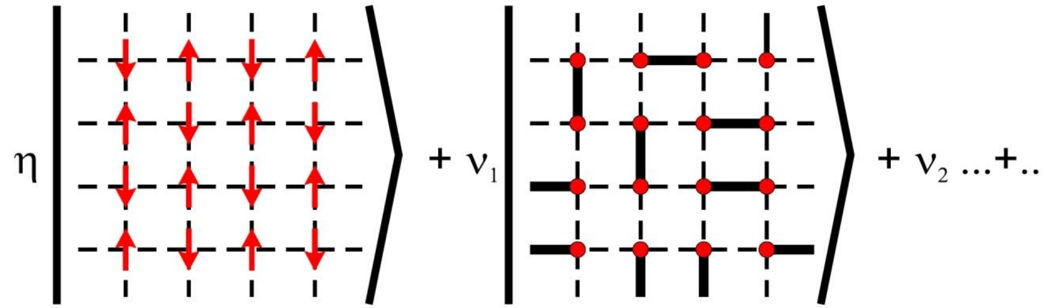
# $Cu(pz)_2(ClO_4)_2$ ZB with diagonal $J_{nnn}$




N. Tsyrulin… A. Schneidewind, P. Link…M. Kenzelmann, Phys. Rev. B **81**, 134409 (2010); Phys. Rev. Lett. **102**, 197201 (2009)

# simple experimentalist's picture:

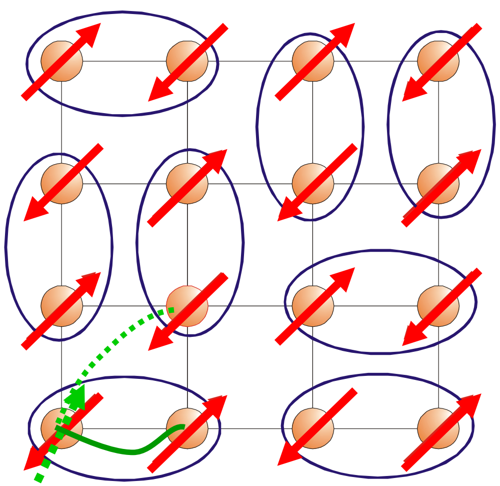
The missing 40% Neel order partly resides in n.n. singlet correlations




Consider the plaquette: 4 spins ⇒ $2^4 = 16$ states – ground state is RVB

$$|\text{8}_{2}^{1}\rangle + |0_1 0_2\rangle = \left(|\uparrow\downarrow\rangle_1 - |\downarrow\uparrow\rangle_1\right)\times\left(|\uparrow\downarrow\rangle_2 - |\downarrow\uparrow\rangle_2\right) + \left(|\substack{\uparrow\\ \downarrow}\rangle_1 - |\substack{\downarrow\\ \uparrow}\rangle_1\right)\times\left(|\substack{\uparrow\\ \downarrow}\rangle_2 - |\substack{\downarrow\\ \uparrow}\rangle_2\right)$$

Hypothesis: ZB effect because superposed on Neel order there are VB correlations
Along ($\pi$,0) n.n. sinlget correlations impede propagating spin waves



Bond energies:

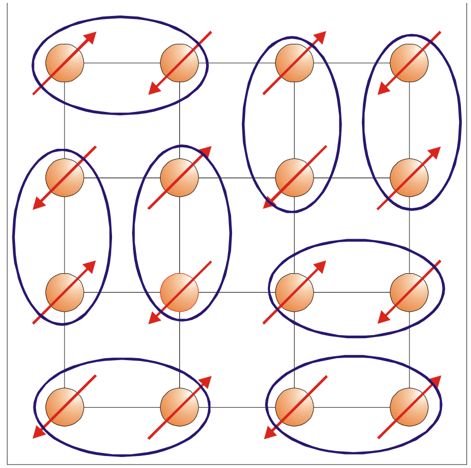
- Classical spins $E_b=-JS^2=-0.25J$
- Best estimates $E_b \approx -0.34J$

Dimers:

- $E_{triplet}$ =+0.25J
- $E_{singlet}$ =-0.75J
- Average for uncorrelated bonds = 0

Need a theory to support or discard this postulate!

# RVB + Neel ?



Starting from RVB?  <S>=0 !

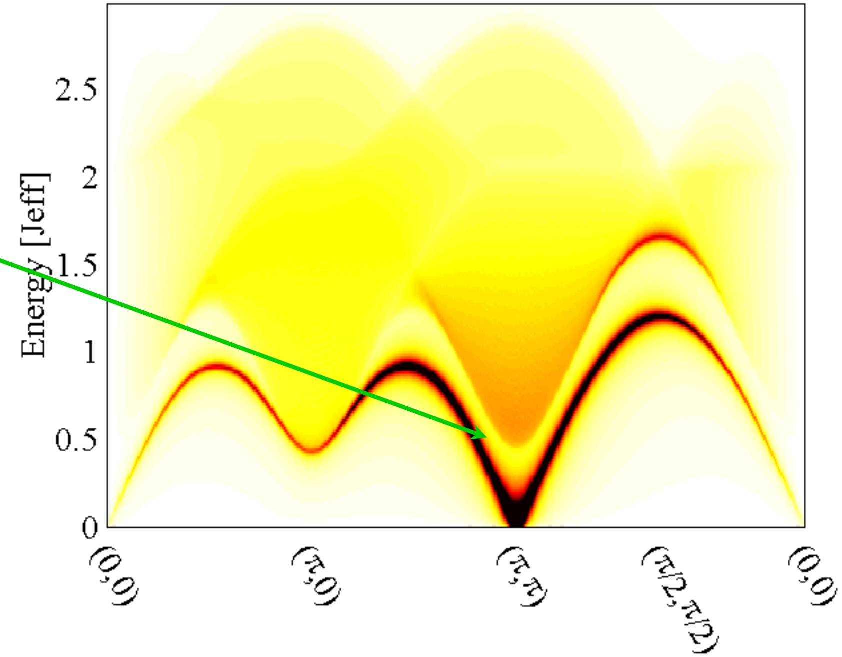
Insert magnetization by hand ⇒ π-flux state

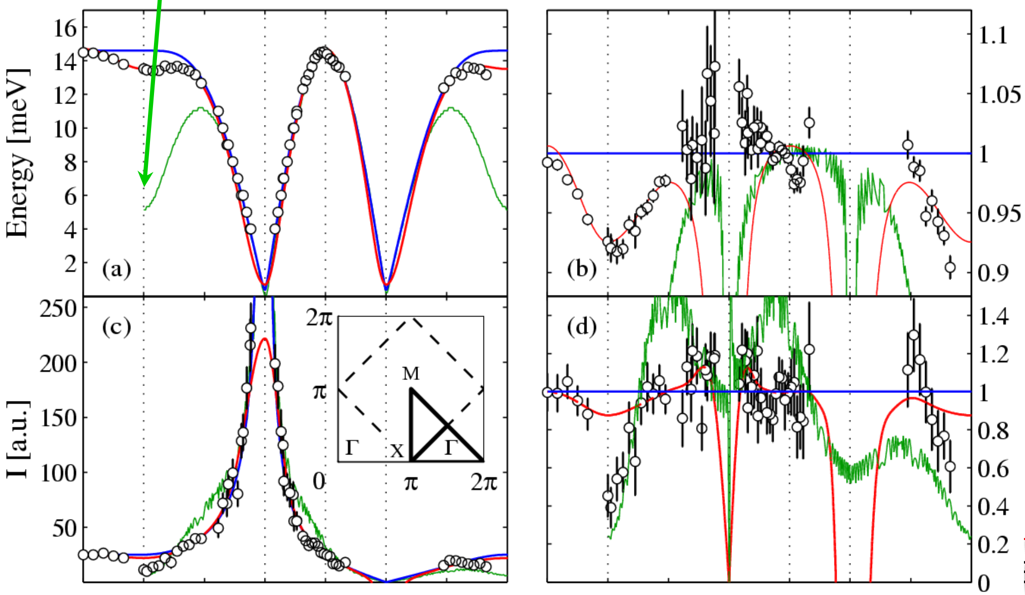
right trend, but too much:

ZB dispersion at (π,0)

but, Gap at (π,π)

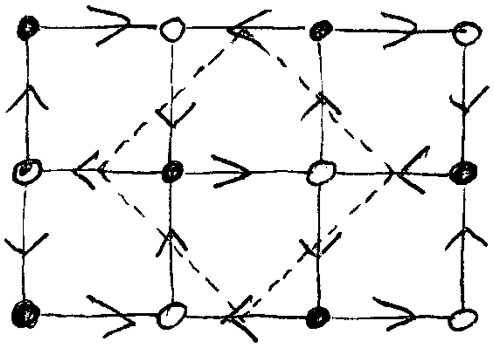
Need better theory



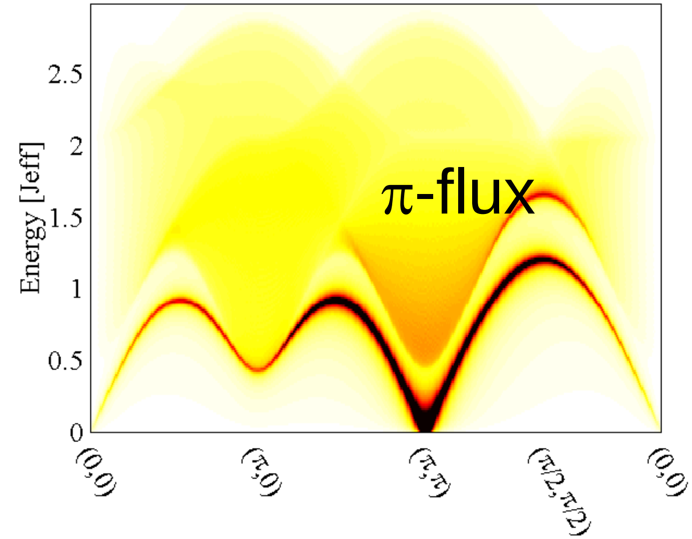

Anderson Science **235** 1196 (1987)
Hsu PRB **41** 11379 (1990); Ho, Ogota, Muthumukar & Anderson PRL (2001),
Syljuasen *et al.* PRL **88** 207207 (2002)

# Staggered flux phases



- RVB-like theory

Anderson Science **235** 1196 (1987)
Hsu PRB **41** 11379 (1990); Ho, Ogota, Muthumukar & Anderson PRL (2001),
Syljuasen *et al.* PRL **88** 207207 (2002)




Work in Fermionic space

$$\mathcal{H} = J\sum_{\langle i,j\rangle} \mathbf{S}_i\cdot\mathbf{S}_j$$
$$= -\frac{J}{2}\left(\sum_{\langle i,j\rangle,\sigma,\sigma'} c^\dagger_{i\sigma}c_{j\sigma}c^\dagger_{j\sigma'}c_{i\sigma'} - \frac{1}{2}\right)$$

D. Ivanov
B. Dalla Piazza

Allow Neel + staggered flux (SF ≅ RVB)

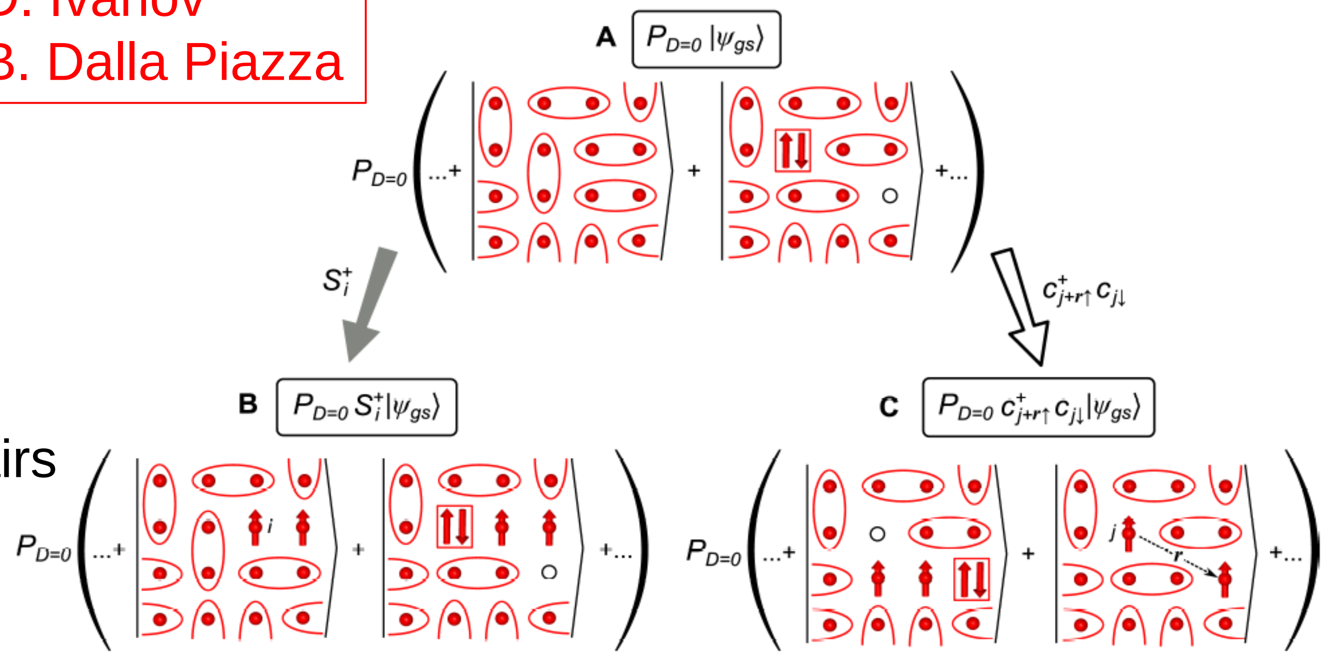
Project our double occupancy

$$|\mathrm{SF}+\mathrm{N}\rangle = P_{D=0}|\psi_{\mathrm{GS}}\rangle$$

Excitations as particle-hole pairs

$$|\mathbf{q},n\rangle_t = \sum_{\mathbf{k}\in\mathrm{MBZ}} \phi^n_{\mathbf{kq}}|\mathbf{k},\mathbf{q}\rangle_t$$

$$|\mathbf{k},\mathbf{q}\rangle_t = P_{D=0}\gamma^\dagger_{\mathbf{k}\uparrow+}\gamma_{\mathbf{k}-\mathbf{q}\downarrow-}|\psi_{\mathrm{SF}(+\mathrm{N})}\rangle$$



# 7m CPU hours later ….

## Monte Rosa at
## Swiss National Supercomputing Center

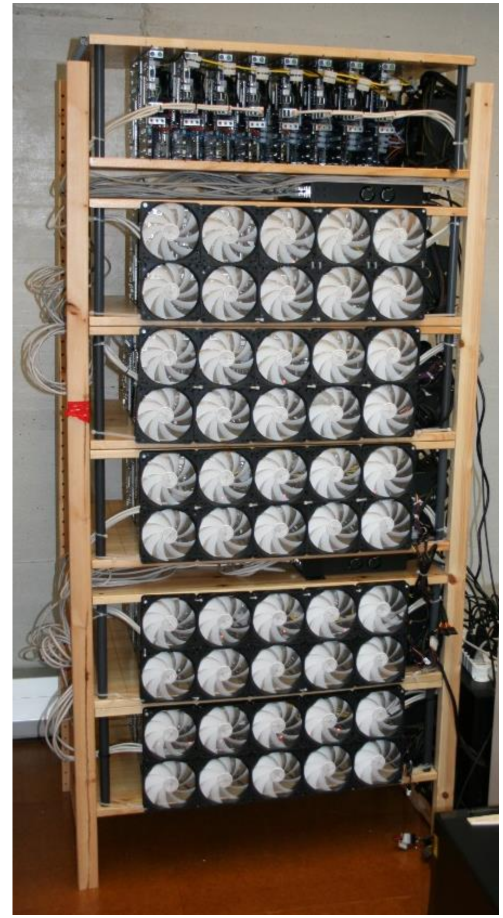

Significance of the proposed research (Please, explain how the proposed work compares and extends the existing body of research and identify weaknesses, if any)

The case for further studies of the Heisenberg model is not strong. The scientific questions have mostly been answered around 1990. Although this might be a good student project, I do not think it is cutting edge research; the model is probably too simplified to explain superconductivity.

Soundness of research methods and tools (Please comment on strengths and weaknesses of the proposed research scheme and its shortcomings, if any)

Rather than doing VMC, this research should be done with exact methods (since there is no sign problem here). Using variational methods, one always wonders how much bias there will be. I would say it is not worth the investment in human and computer time. See for example Phys. Rev. B 40, 2737 (1989), citations, later references, and recent work of Sandvig on

## Quantum Wolf Cluster at LQM



**Key figures:**
96 nodes, 384 CPUs
9.6 Tflops, 4.8 kW
312 CHF/ node
Plan x2 / year
Open for collaborations

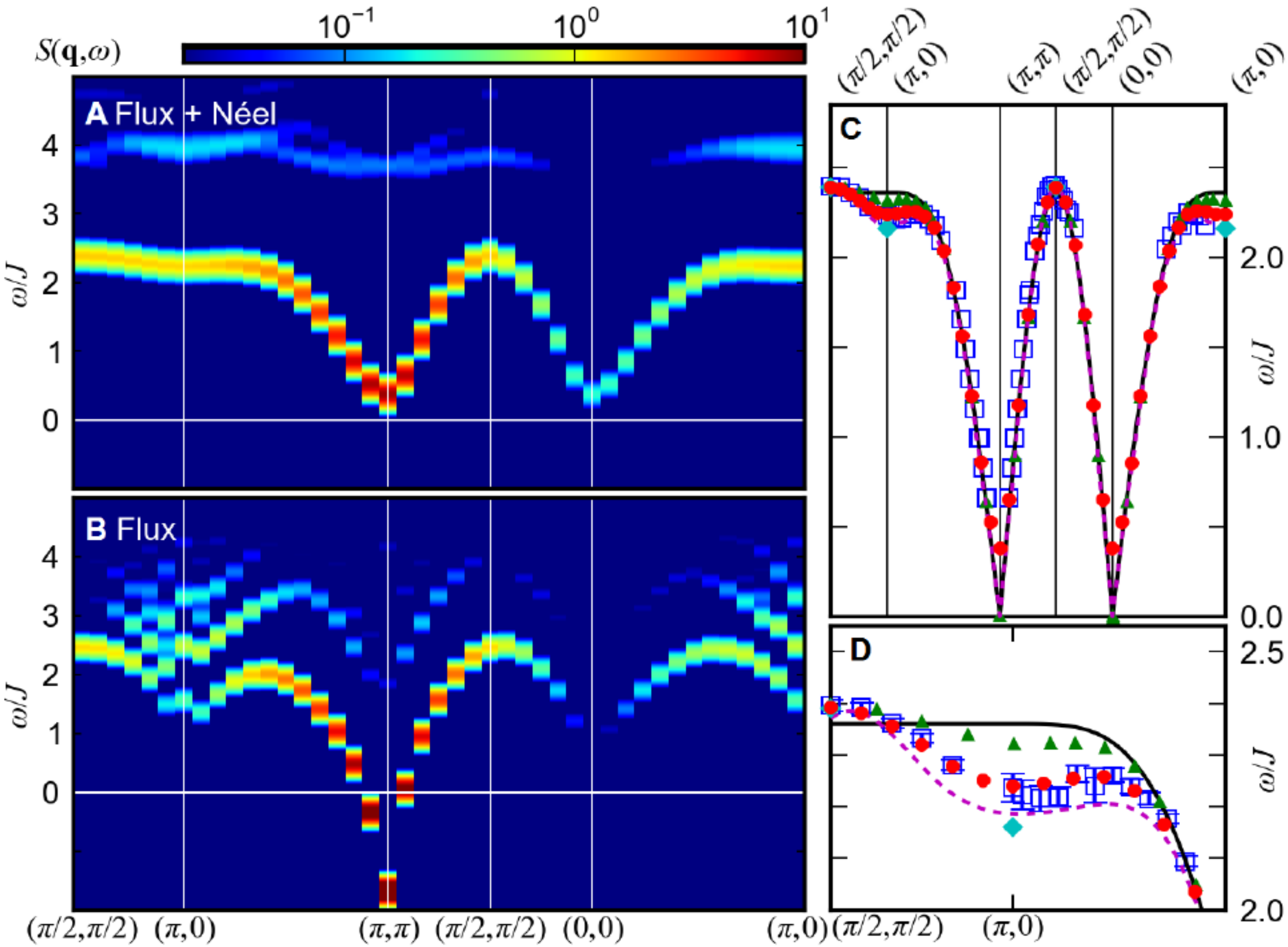
# Not perfect, but best description so far:

Spinon description recovers spin wave dispersion for most Q

Best match of ZB dispersion.
Beats 3rd order SWT

Con: must switch off Neel to get continuum

Pro: when do, we get continuum around (π,0) as in experiment



# Measure spinon separation

Define separated spinon state

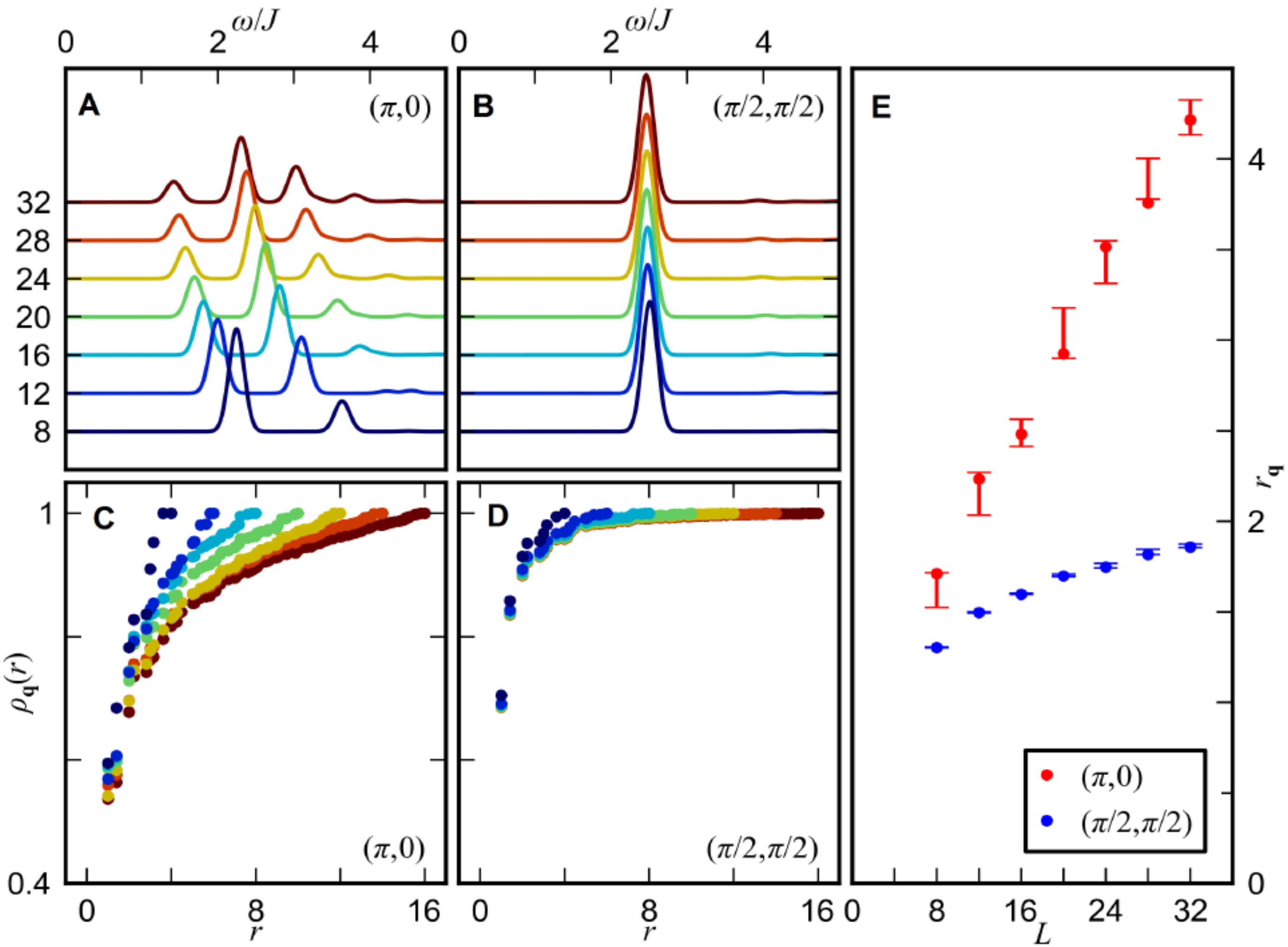
$$|\mathbf{q},\mathbf{r}\rangle_t = \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} c^\dagger_{\mathbf{R}+\mathbf{r}\uparrow} c_{\mathbf{R}\downarrow} |\psi_{\mathrm{SF}}\rangle$$

Calculate overlap with our excitations

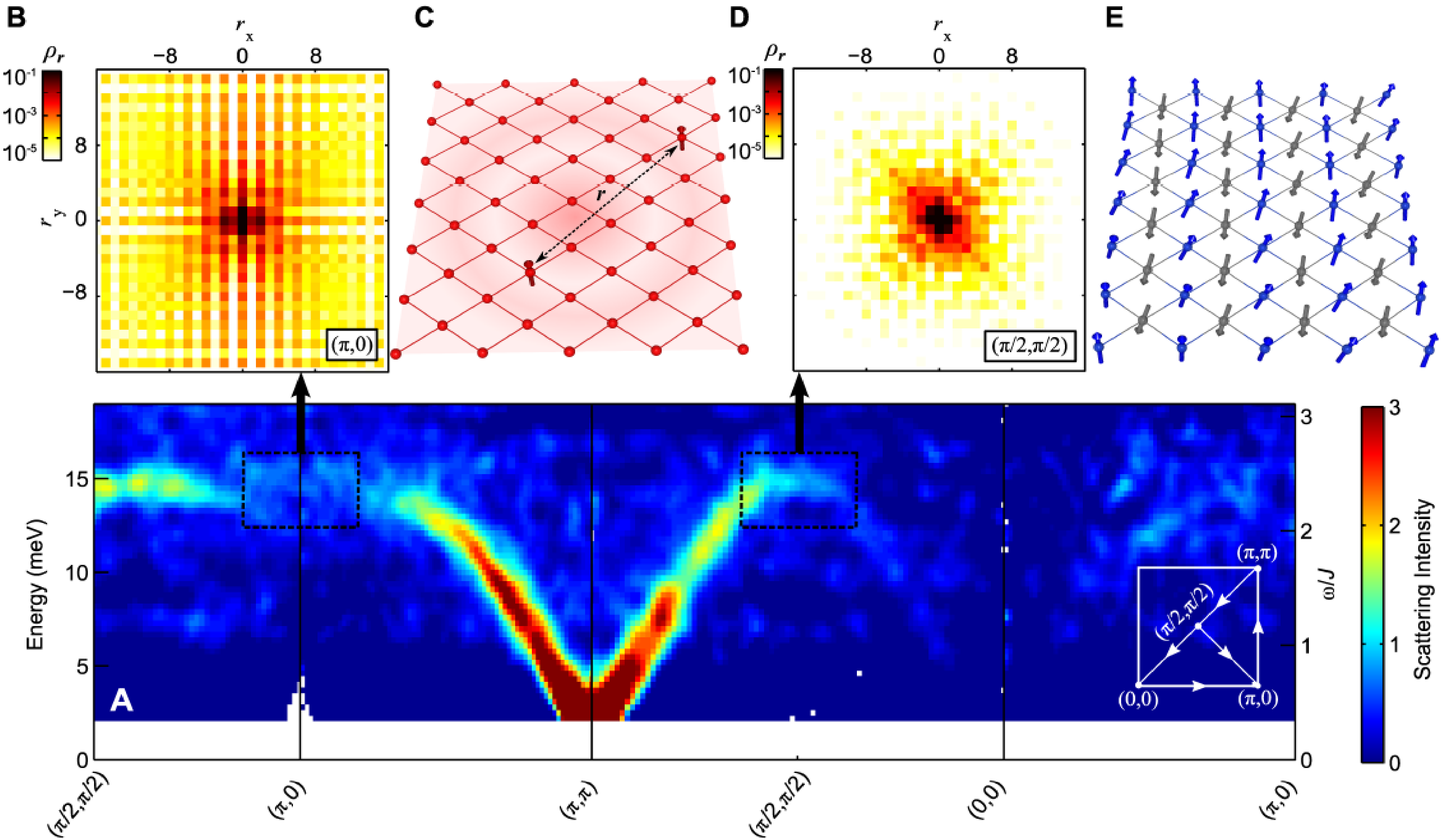
$$\tilde{\rho}_{\mathbf{q}}(\mathbf{r}) = \sum_n |_t\langle \mathbf{q},\mathbf{r}|\mathbf{q},n\rangle_{tt}\langle \mathbf{q},n|\mathbf{q},0\rangle_t|^2$$

(π/2, π/2) converge and has short spinon separation
⇒ spin-waves

(π, 0) grow linear with system size
⇒ spinons deconfine !

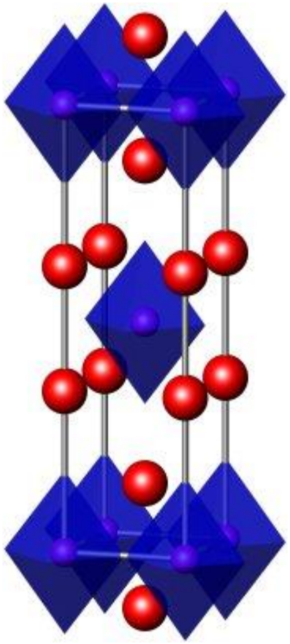


# Spinons in 2D square lattice !



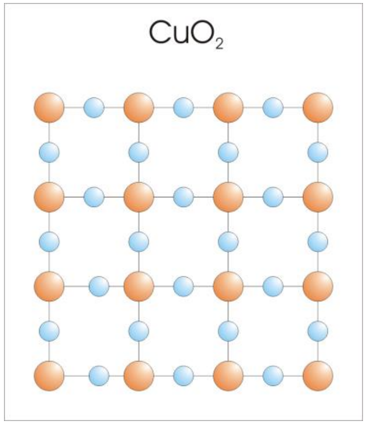

B. Dalla Piazza, M. Mourigal, D. Ivanov et al. Nat. Phys. **11**, 62 (2014)

# RVB in 2D magnet – *dakara nani* ?



## Cuprate superconductors

**Bednorz and Müller (1986)**



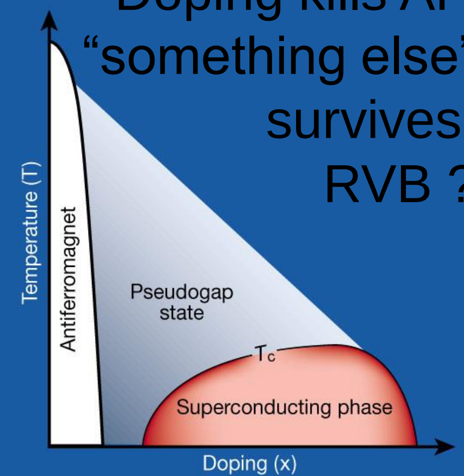

THE RESONATING VALENCE BOND STATE IN LA2CUO4 AND SUPERCONDUCTIVITY
ANDERSON PW
SCIENCE
235 (4793): 1196-1198 MAR 6 1987

**Language:** English **Cited References:** 27 **Times Cited:** 3823

1987 Rather Vague B…

2009 Quantitative efforts

Doping kills AF,
"something else"
survives,
RVB ?




J. Phys.: Condens. Matter **16** (2004) R755–R769 PII: S0953-8984(04)80644-1

TOPICAL REVIEW

**The physics behind high-temperature superconducting cuprates: the 'plain vanilla' version of RVB**

P W Anderson[1], P A Lee[2], M Randeria[3], T M Rice[4], N Trivedi[3] and F C Zhang[5,6]

Is ZB anomaly the smoking gun of RVB ?

# Conclusion

- Quantum magnets allow studying exotic ground states and correspondingly exotic excitations

- Comparison theory experiment (especially neutron scattering)
  - Spin-flips, triplons, spin-waves, spinons,

- 1D S=1/2 antiferromagnetic chain host fractional spinons
  - we can quantify 2-spinon and 4-spinon excitations

- 2D S=1/2 square lattice HAF is so simple we should understand it
  - Fractional excitations can exist in un-frustrated 2D models
  - Implications:
    - How high-energy spinons evolve upon doping
    - Need better theories for quantum fluctuations in ordered systems
    - Spin-charge separation in 2D ?

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Felix Groitl
Elahi Shaik
A. Omrani (now Berkeley)
M. Mourigal (building group at Georgia Tech: postdoc positions open: Solid-state chemist and/or neutron-X-ray expertise)

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Denmark: NB Christensen, Lefmann

Enderle, Harrison ILL France
McMorrow, Aeppli LCN & UCL
Caux, Amsterdam, Kiefer Germany
Tokura, Isobe, Hiroi, Masuda Japan