Quantum Magnetism
- Neutrons in the Quasi-particle Zoo

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Outline

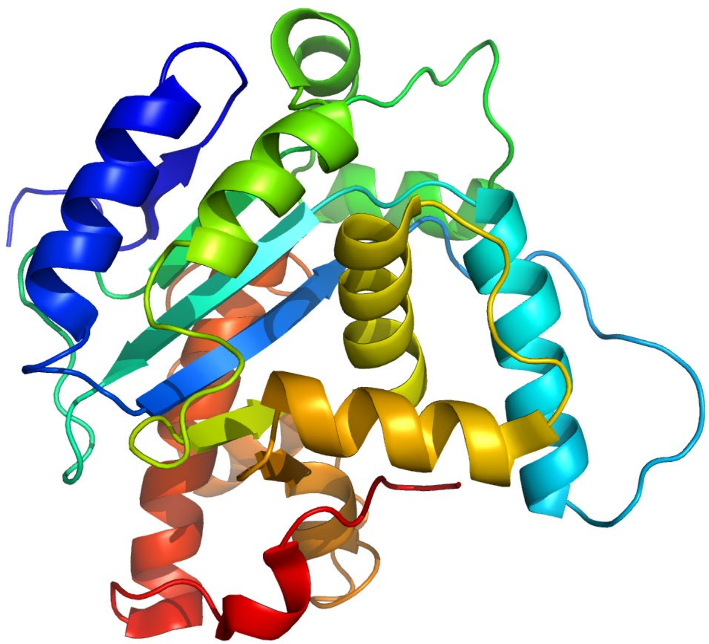
- Quantum Magnetism
  - Arena for many-body physics and novel electronic materials
  - Models – Materials – Measurements

- 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

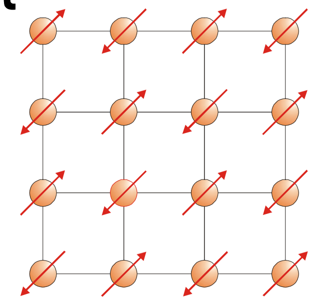
Complexity of many-body systems

- Structure of a protein



- Pop2p-subunit Jonstrup et al (2007)
- Mega-Dalton:
  ~1'000'000 atoms
  ~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.

$$= -2\left|\begin{smallmatrix}\uparrow&\downarrow\\\downarrow&\uparrow\end{smallmatrix}\right\rangle - 2\left|\begin{smallmatrix}\downarrow&\uparrow\\\uparrow&\downarrow\end{smallmatrix}\right\rangle + \left|\begin{smallmatrix}\uparrow&\downarrow\\\uparrow&\downarrow\end{smallmatrix}\right\rangle + \left|\begin{smallmatrix}\downarrow&\uparrow\\\downarrow&\uparrow\end{smallmatrix}\right\rangle + \left|\begin{smallmatrix}\uparrow&\uparrow\\\downarrow&\downarrow\end{smallmatrix}\right\rangle + \left|\begin{smallmatrix}\downarrow&\downarrow\\\uparrow&\uparrow\end{smallmatrix}\right\rangle$$

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: $S = 0, 1/2, 1, 3/2, \ldots\ldots \infty$

$S = 1/2$ → $+1/2$ ↑, $-1/2$ ↓

- 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

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

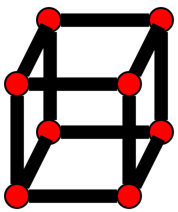
  e.g. two spins 'entangled'

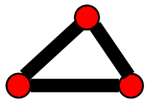
  this is why $\propto 2^N$, not $\propto N$

Quantum Magnetism – materials as quantum simulators

1) Model and Materials

Spin, interactions
dimension
frustration





2) Theoretical methods

analytic approximations
numerical simulations

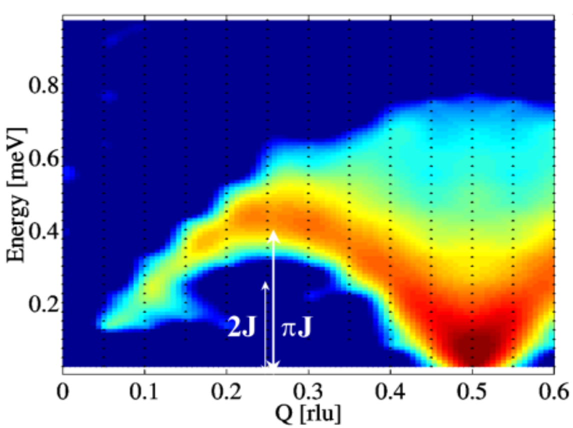
The LEGO of many body physics

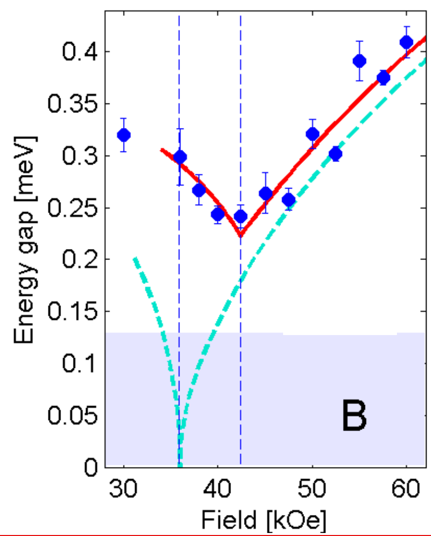
3) Experimental tools:

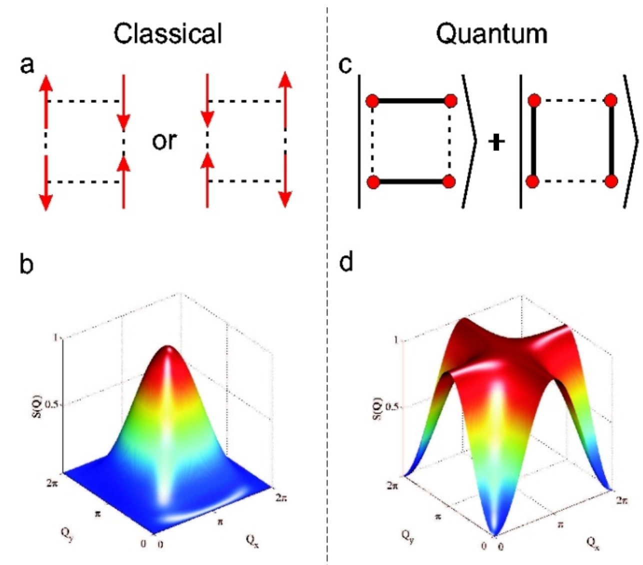
Bulk probes
Neutron scattering

Phenomena:

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

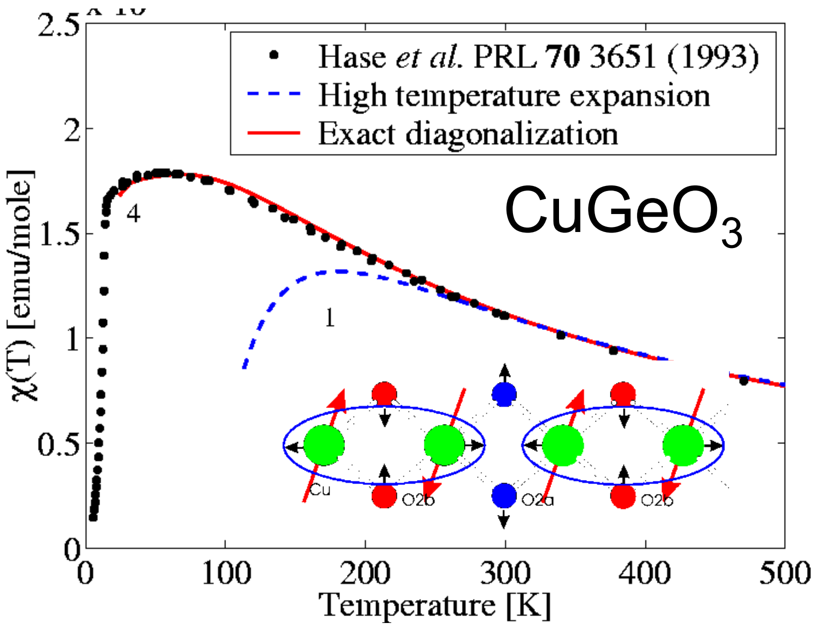




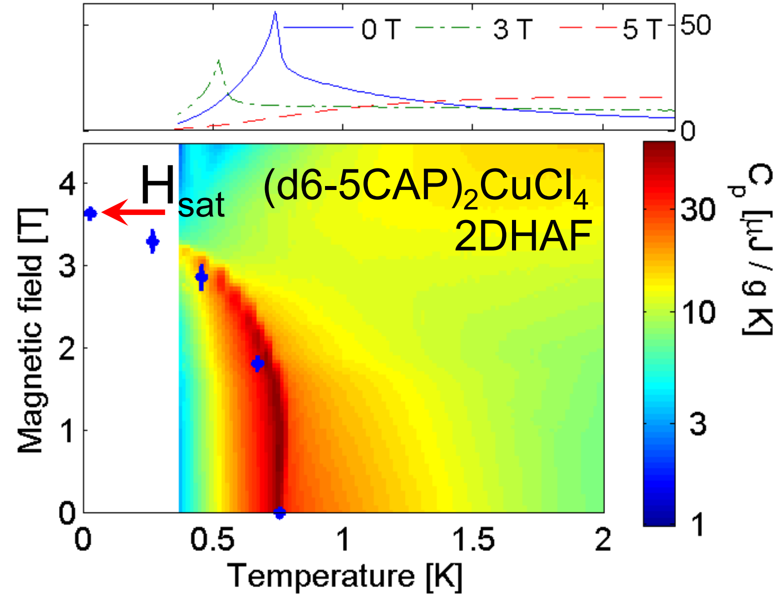


Magnetic measurements

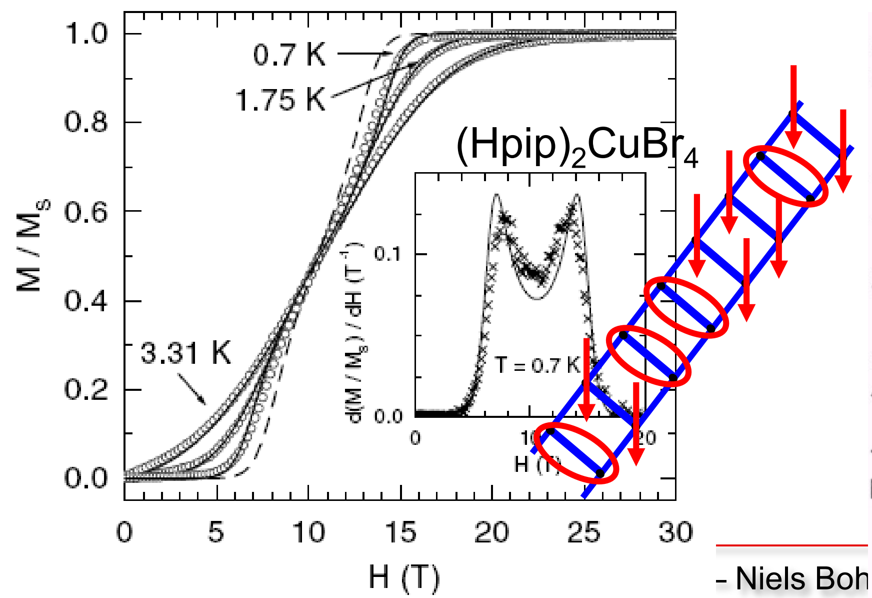
Susceptibility



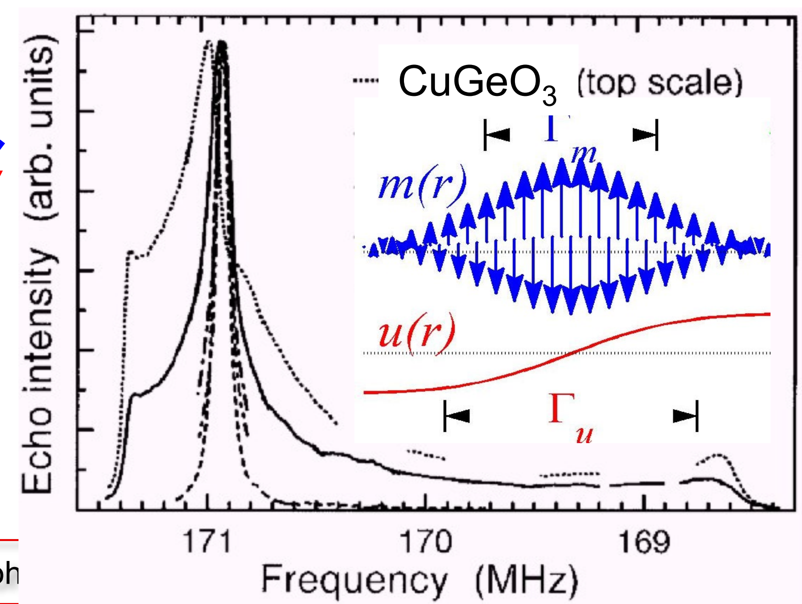
Specific heat



Magnetization



NMR, μSR etc.



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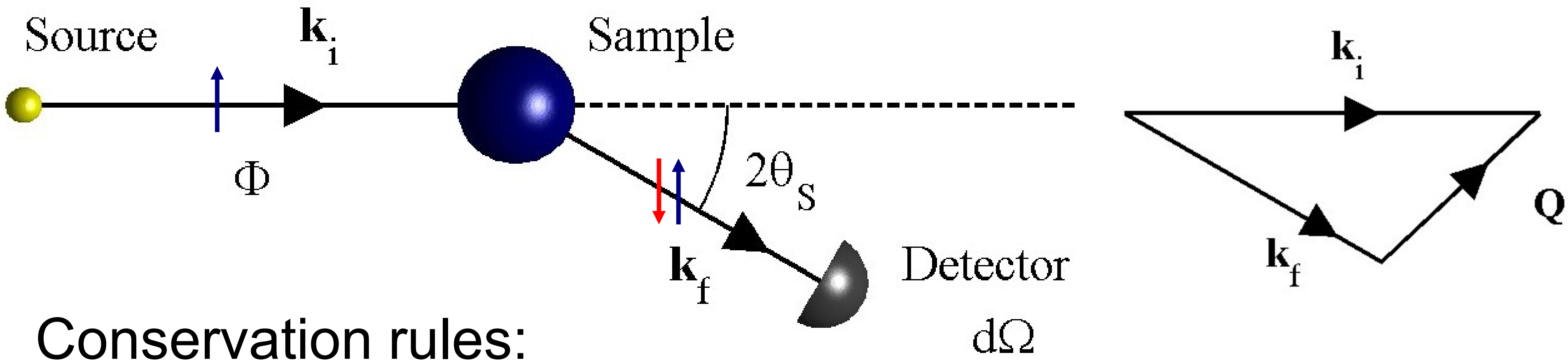
A unique tool: Neutron scattering

scattering and conservation rules

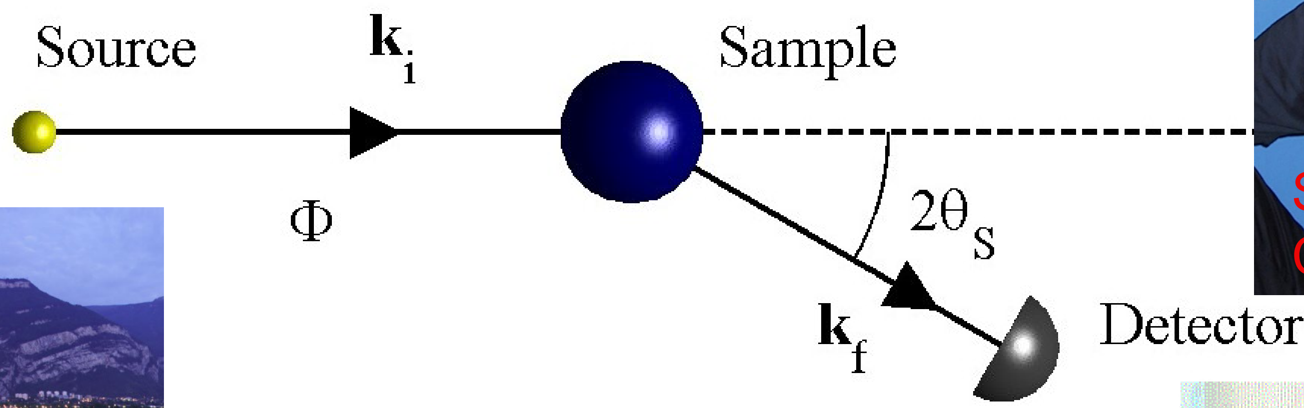


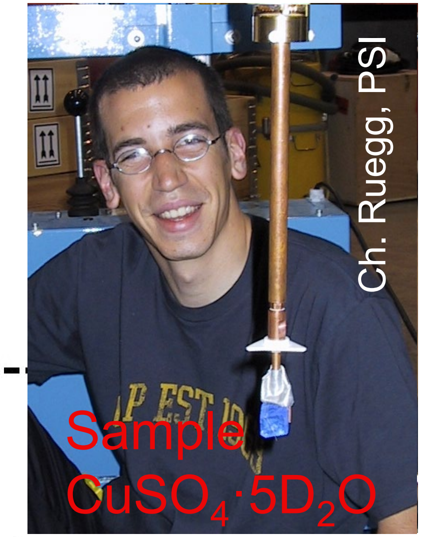
Conservation rules:

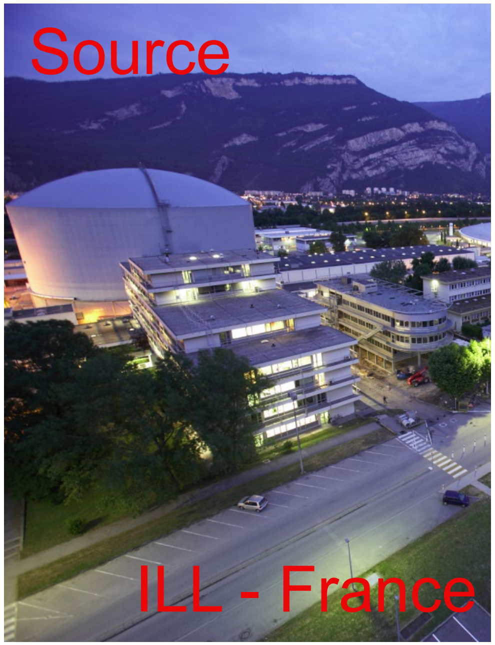
	Sample		Neutron	
Momentum	$\hbar\mathbf{Q}$	=	$\hbar\mathbf{k}_i - \hbar\mathbf{k}_f$	
Energy	$\hbar\omega$	=	$E_i - E_f$	$= \hbar(k_i^2 - k_f^2)/2m_n$
Spin	ΔS	=	$\sigma_i - \sigma_f$	

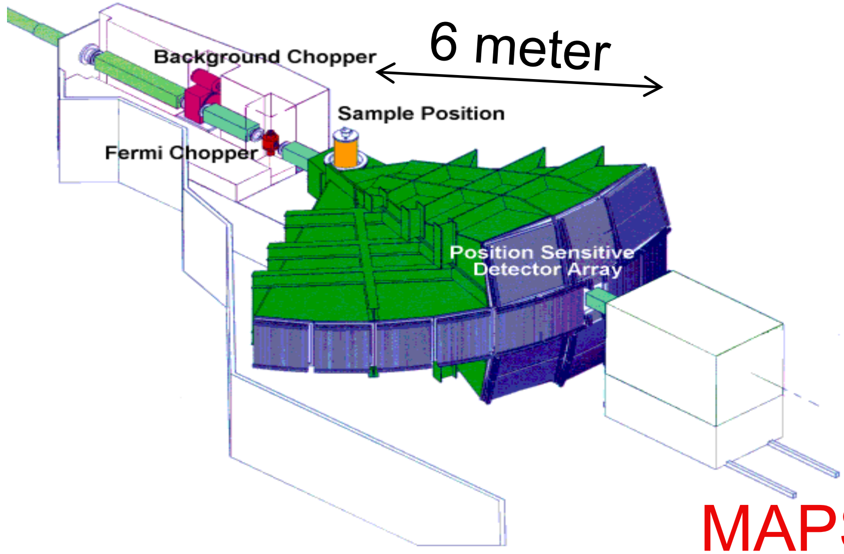
$\Rightarrow$ We can control and measure these quantities !

Large scale instruments and facilities









MAPS $16m^2$ detector bank

Neutron scattering – an intense future

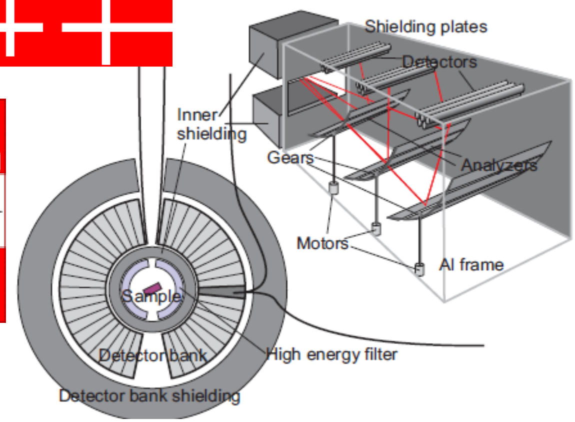
- 1^{st} generation facilities:
  - General purpose research reactors
- 2^{nd} generation facilities:
  - Dedicated to neutron scattering:
  - ILL, France, FRM2 Munich, SINQ CH, ISIS, UK etc.
- 3^{rd} generation facilities:
  - SNS, US 1.4b$, commissioned 2006
  - J-Parc, Japan 150b¥, commissioned 2008
  - ESS, Sweden 2b€, start 2013, commission 2023
  - 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), Lund

Denmark is co-host nation
Switzerland contributes 3.4%
⇒ CH-DK collaboration on instruments

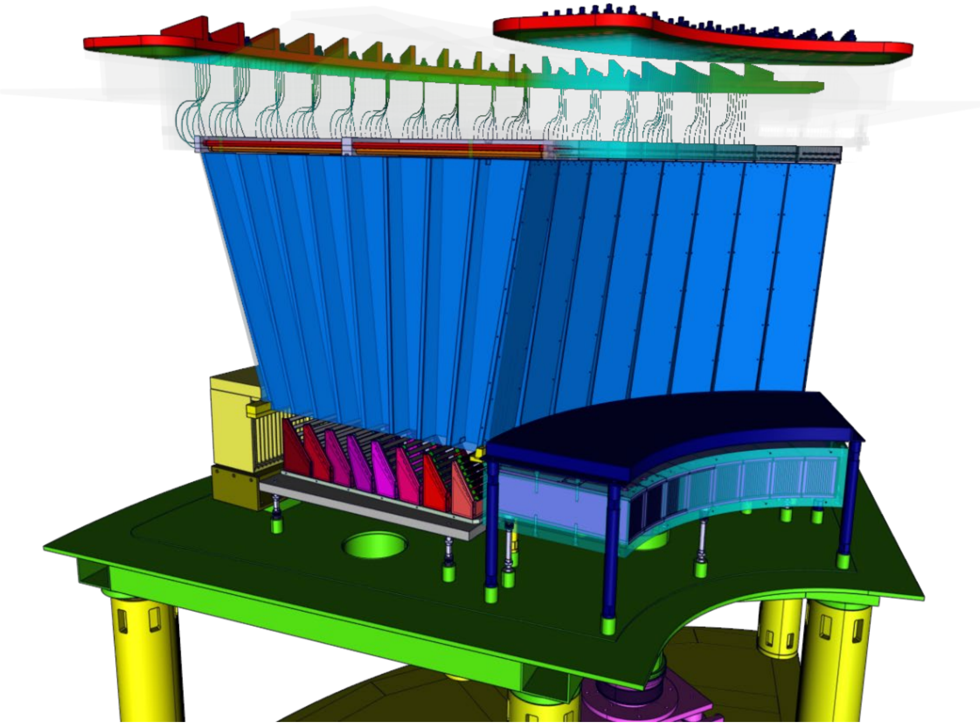
⇒ BiFrost: 10^2 - 10^4 over current best

ESS + SSDN SGN



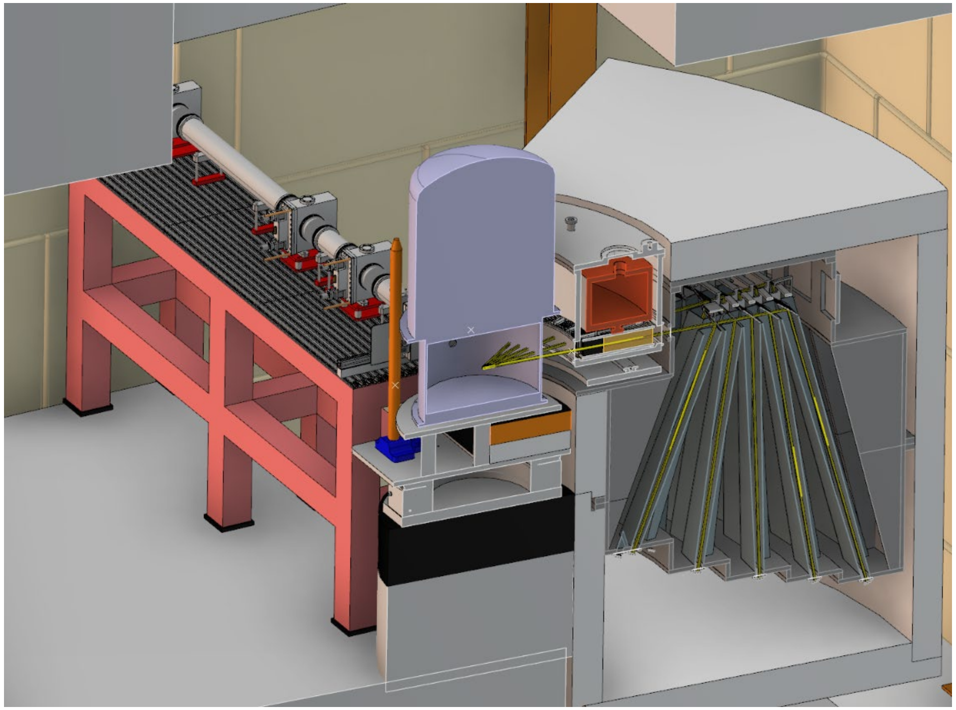
CAMEA

- Multi-TAS
- At Swiss Spallation Neutron Source
- End 2018: first measurements
- First of a new instrument type



BiFrost

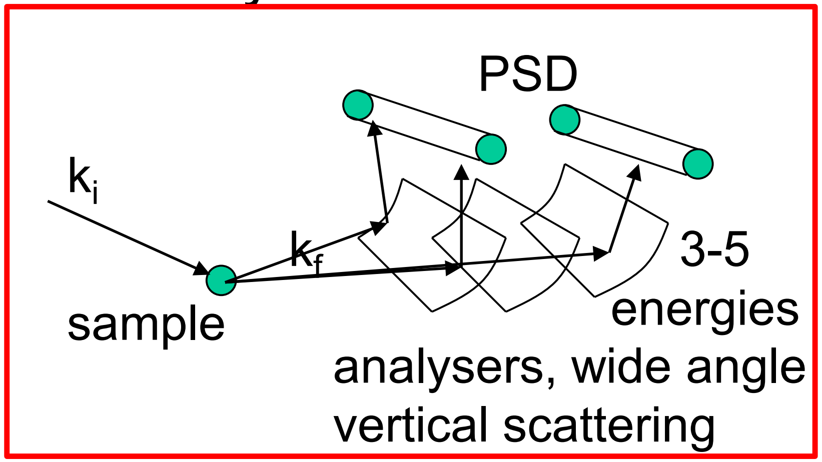
- Indirect-TOF
- Scheduled among 1st instruments at ESS
- 202? first neutrons
- Spectroscopy from 1mm3 samples
- Continuous parametric studies



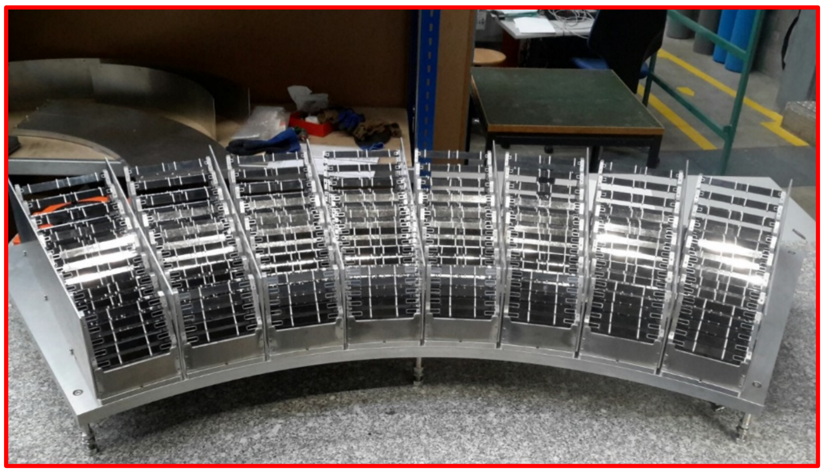
CAMEA - Continuous Angle Multiple Energy Analysis

Measure 100 times more neutrons

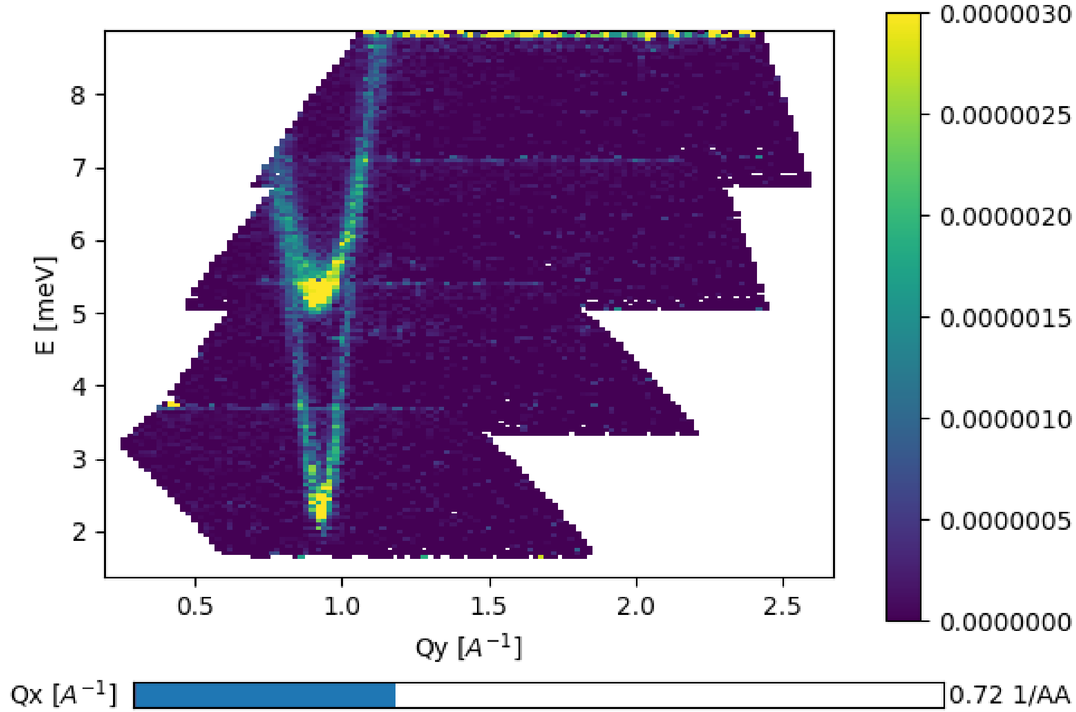
~2002 my first sketch



2018 first instrument

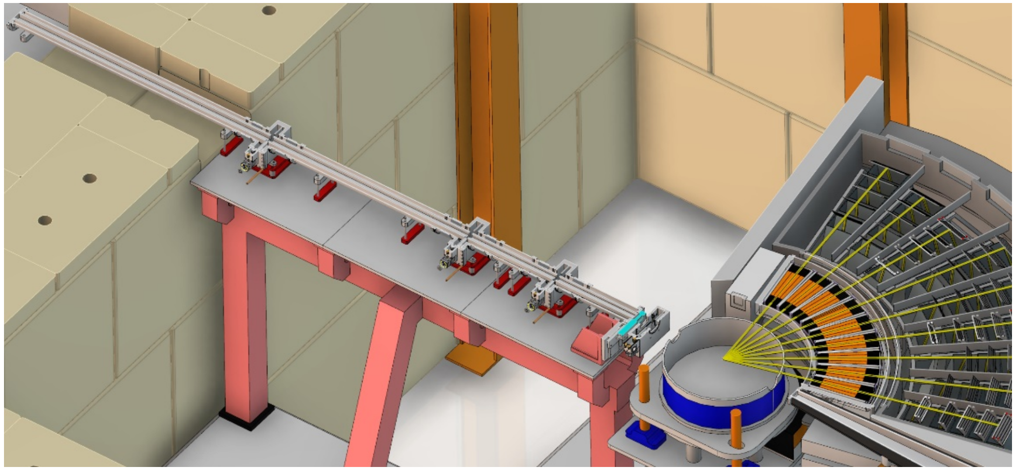


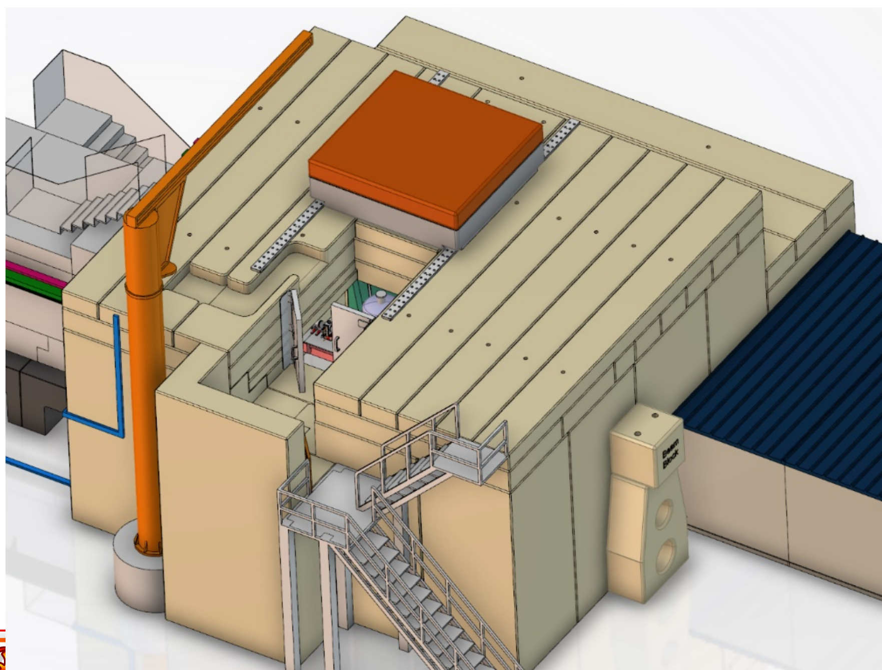
First measurements last week :



DK Team members: Kim Lefmann, Jakob Lass, Jonas O Birk, Mads Bertelsen, Martin Olsen, Rebekka Frøystad og Asla Husgard

BiFrost @ ESS





Quick Facts	
Sector	West
Beam Port	W04
Class	Spectrometry
Commissioning/Operation	2022/2023
Moderator	Cold
Length	162 m
2θ-Range [deg.]	7-135
Analyzer energies [meV]	2.7, 3.2, 3.8, 4.4 and 5.0
2θ-Coverage (2 settings)	90 degrees
ΔE-Range	-3 – 55 meV
Divergence range (FWHM)	0.4 – 1.5 deg.
2θ resolution	0.7 – 1.2 deg.
High flux mode [2.3 – 4.0 Å]	
Flux [n/s/cm^2]	$1 \cdot 10^{10}$
Elastic line resolution @ E_f = 5.0 meV	190 μeV
Resolution at ΔE = 5 meV [E_f = 5 meV]	450 μeV
High resolution mode [2.3 – 4.0 Å]	
Flux [n/s/cm^2]	$6 \cdot 10^{8}$
Elastic line resolution @ E_f = 5.0 meV (prismatic)	50 μeV
Resolution at ΔE = 5 meV [E_f = 3.8 meV] (prismatic)	50 μeV

Neutron scattering cross-section – the power of simplicity

Fermi's golden rule for transition probability

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

From initial state ***i*** to final state ***f*** of neutron $\mathbf{k}$ and sample λ

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 $\Rightarrow$ gen. susceptibility

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

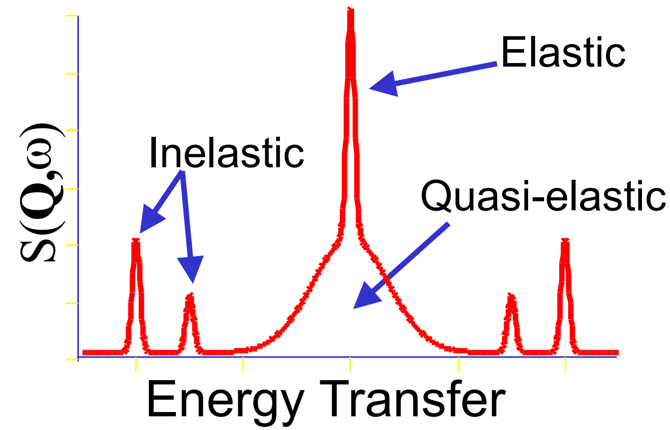
intrinsic dynamics $\Leftrightarrow$ response to perturbation

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

Inelastic magnetic scattering: Lets take the scenic route…

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 ?

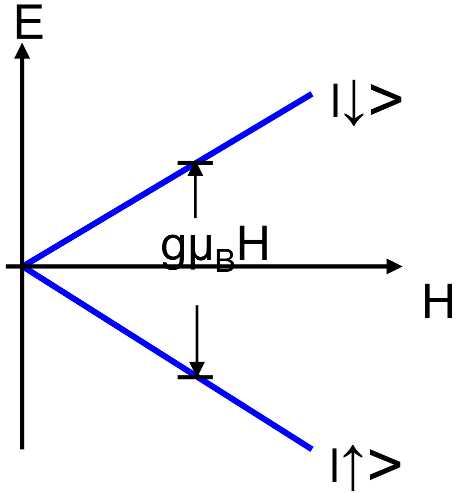
Between long range ordered states

… and spin liquids

paramagnetic spins S=1/2

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

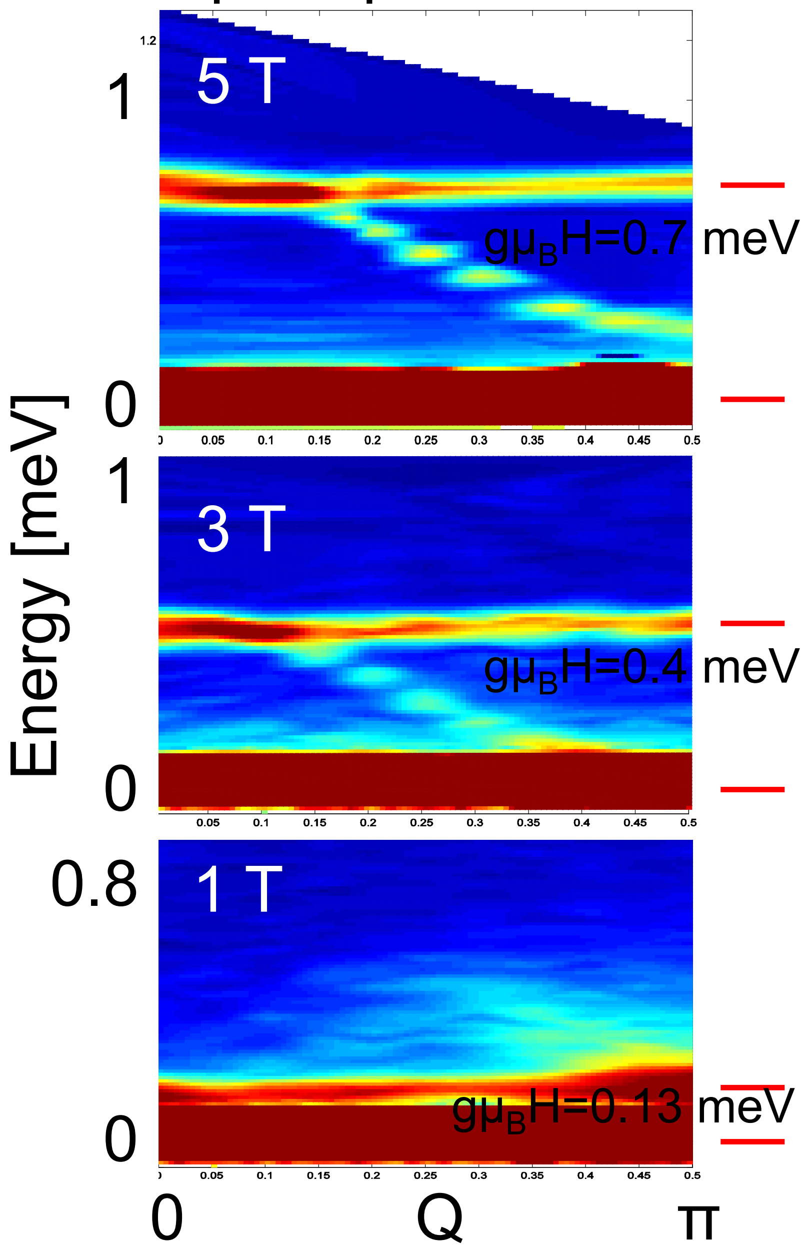
$CuSO_4 \cdot 5D_2O$



- Local excitation

  ⇒ no Q-dependence

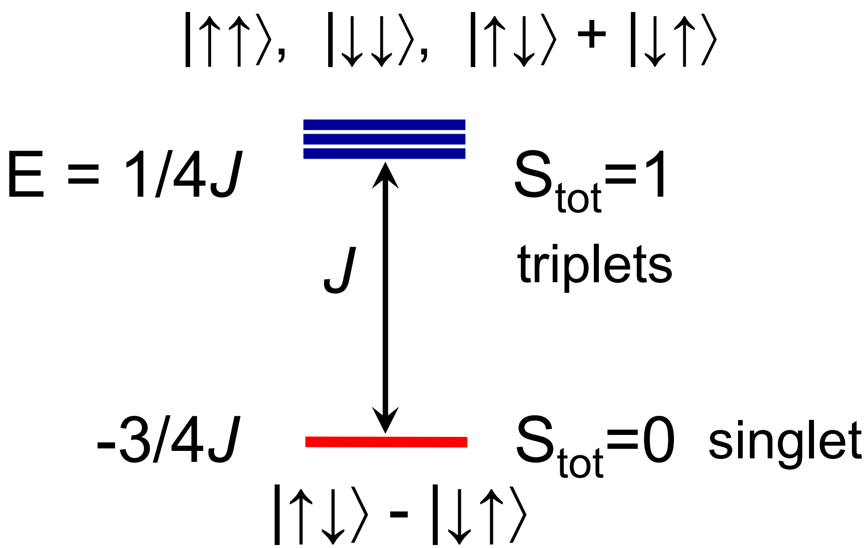
Spin-flip excitation



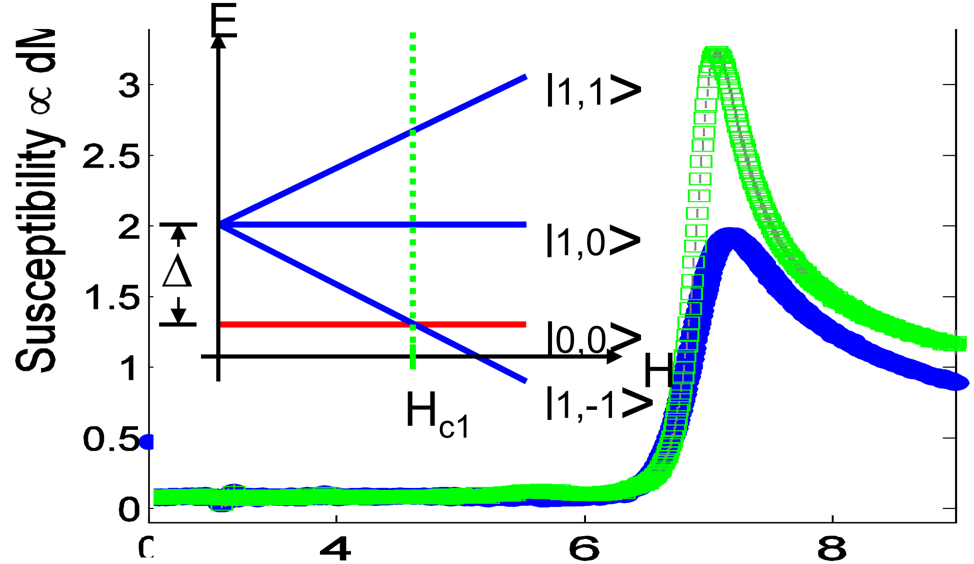
Take two – the spin pair

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

Antiferromagnetic: $J > 0$

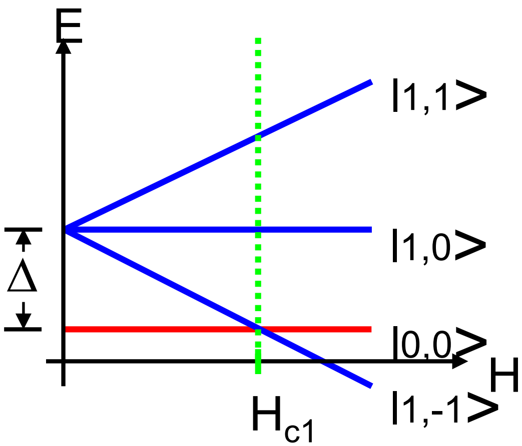


No magnetization or susceptibility up to critical field



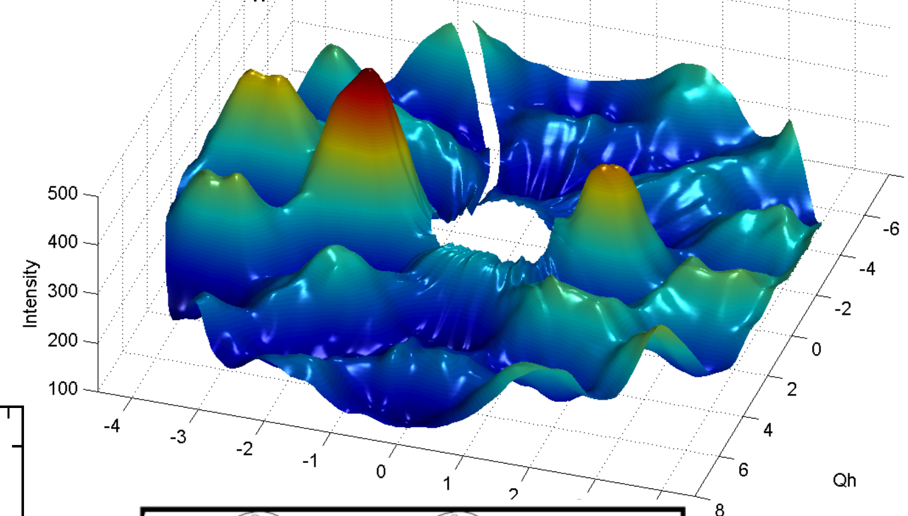
Singlet ground state: $\langle S^z{}_1 \rangle = \langle S^z{}_2 \rangle = 0$

Take two – the spin pair

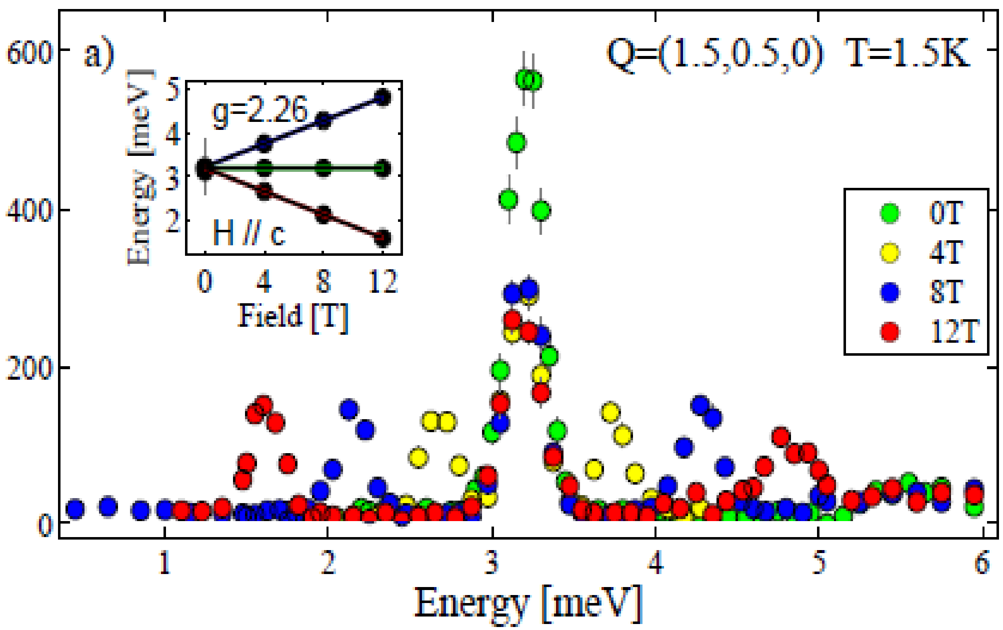


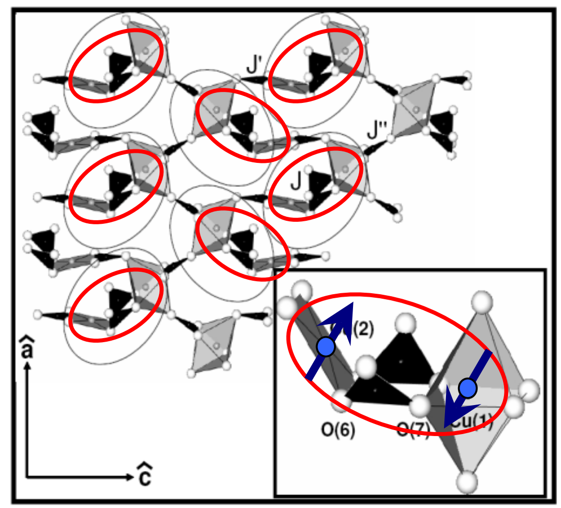
Structure factor along pairs:

$S(q) \sim \cos(d_{||}\cdot q) + \cos(d_{\perp}\cdot q)$



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





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

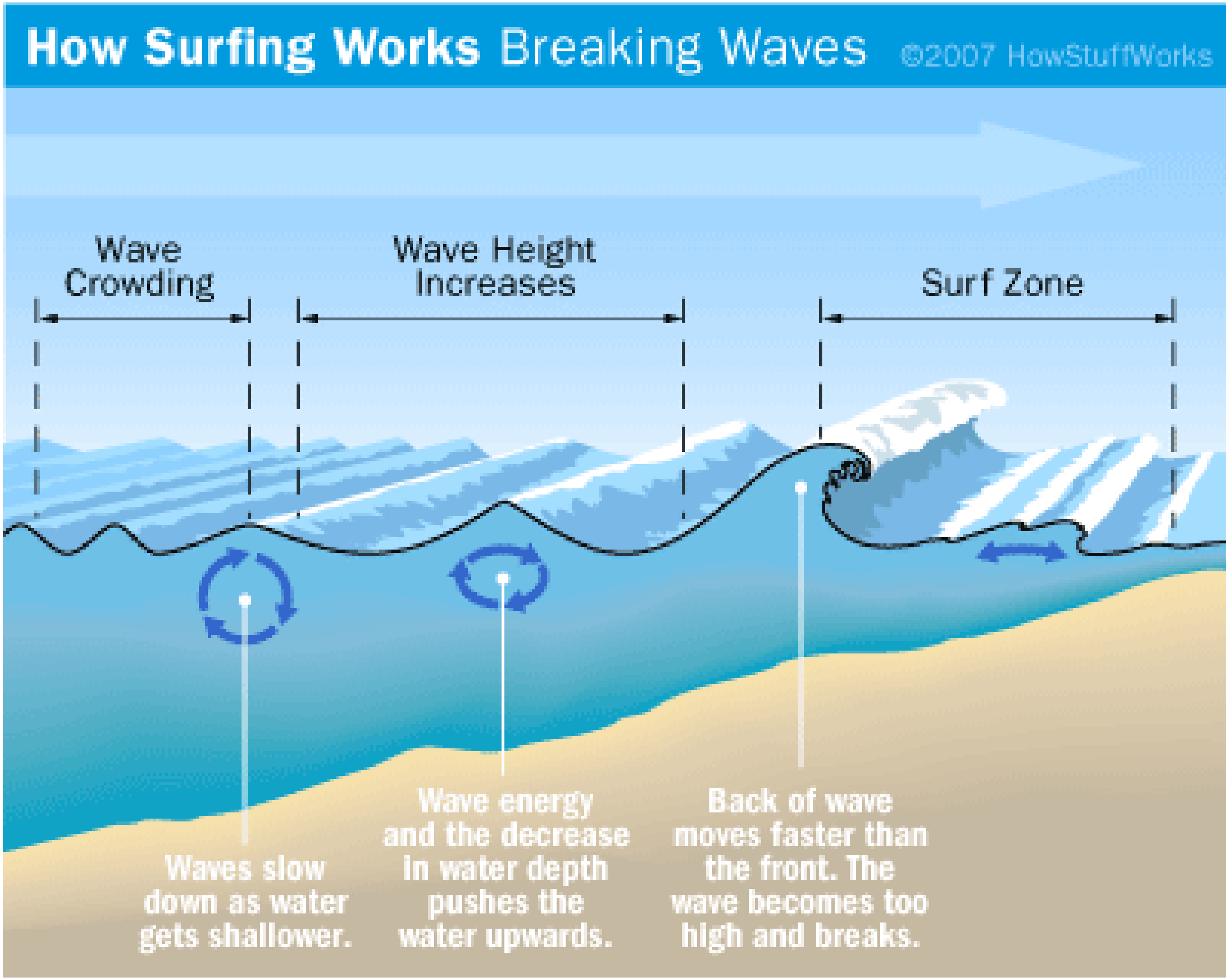
The Heisenberg model

- Seem innocently simple. 1 and 2 spins were trivial.

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

- for extended systems, do we understand it well enough ?

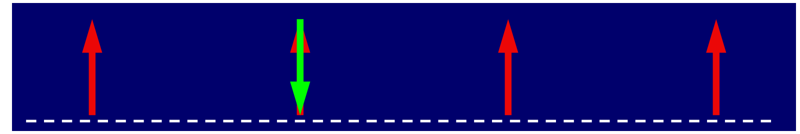
When do we understand ? What is a theory ?



- Model
- Solution
- Picture

- Waves are collective excitations of water
- Energy travels far while water only moves locally
- “Quasi-particles” are collective excitations in the atomic limit

Ferromagnets are easy, exact solution:



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

⇑ 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

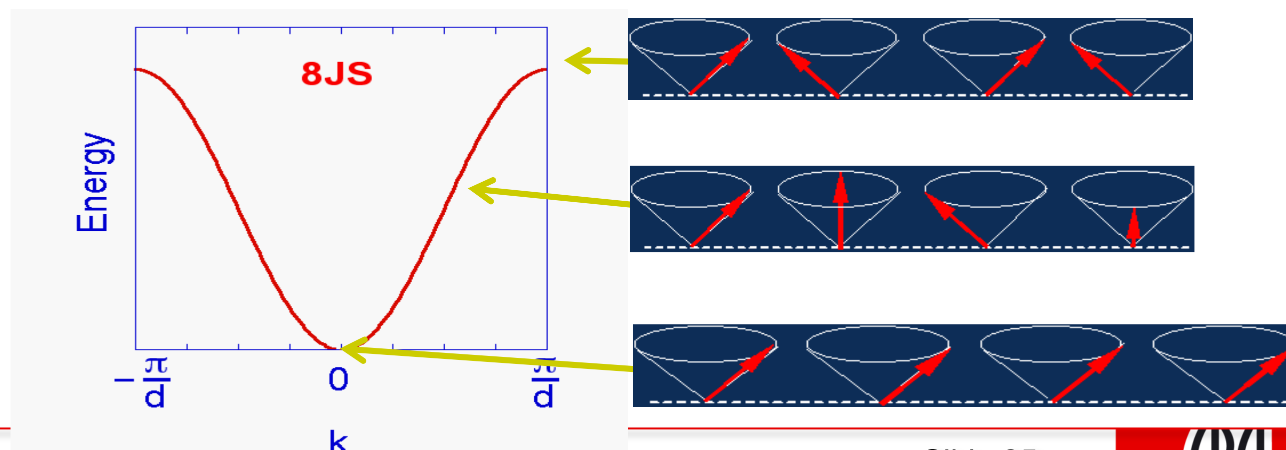
Periodic linear combination: $|k\rangle = N^{-1/2}\Sigma_r e^{ikr}|r\rangle$ — plane wave

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

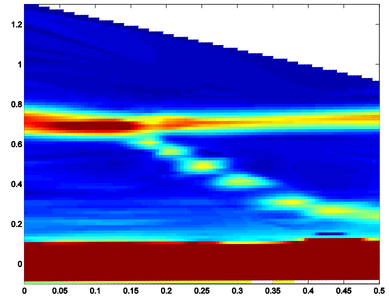
Time evolution: $|k(t)\rangle = e^{iHt}|k\rangle = e^{iE_k t}|k\rangle$ — sliding wave

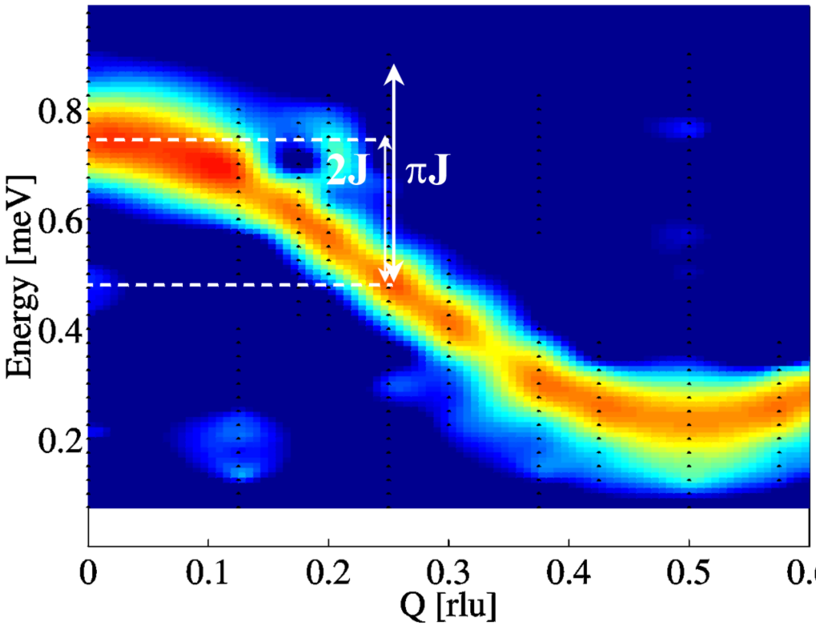
Dispersion:
relation between
time- and space-
modulation period

Same result in classical
calculation ⇒ precession:



Spin waves in a “ferromagnet”





$CuSO_4 \cdot 5D_2O$

=

$Cu_2(SO_4)_2 \cdot 10D_2O$

=

1 Cu S=1/2 uncoupled

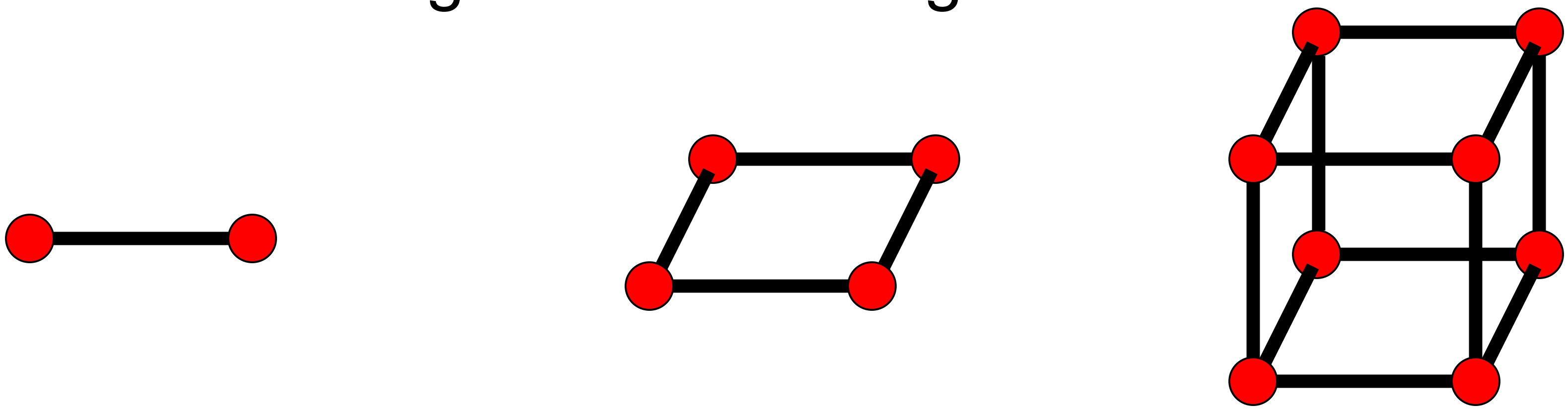
1 Cu S=1/2 chain

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

Actually it is an antiferromagnet polarized by 5T field

Quantum 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

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

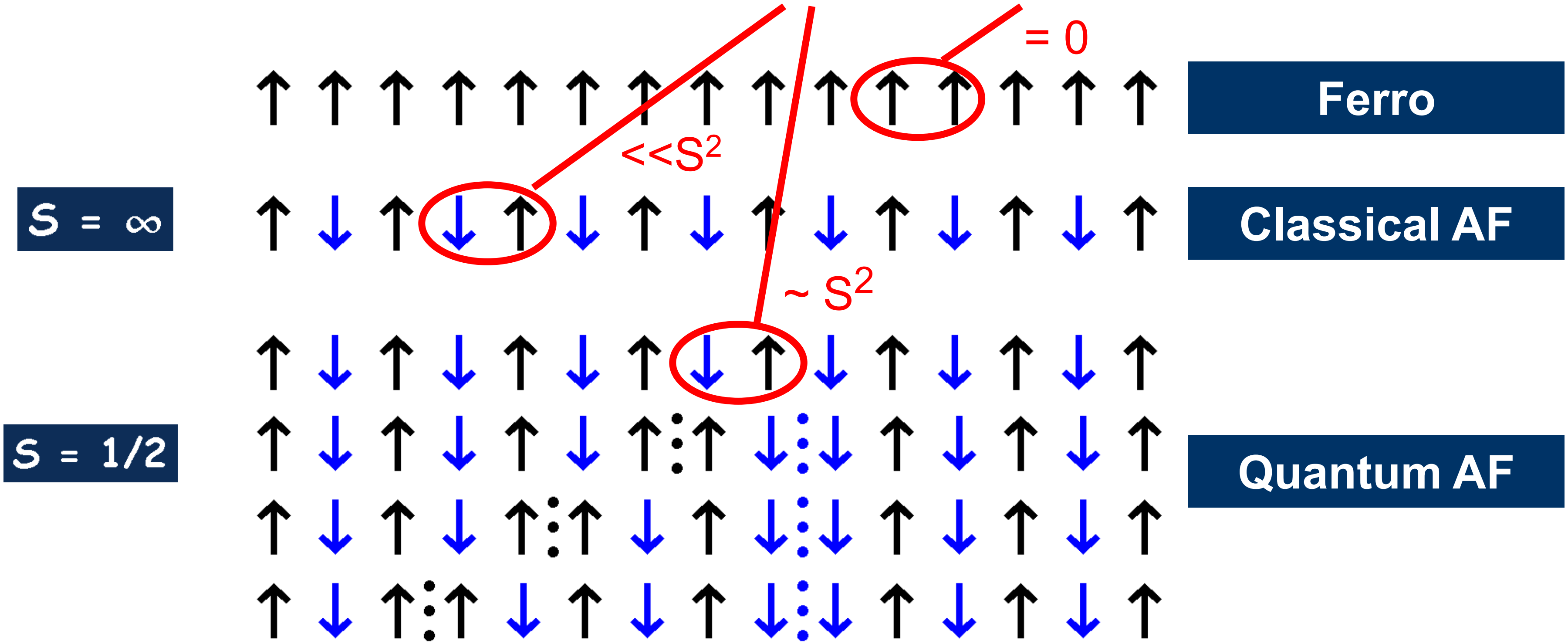
- Analytic solution by Bethe in 1931: 'domain wall quantum soup'

Outline

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

antiferromagnetic spin chain

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

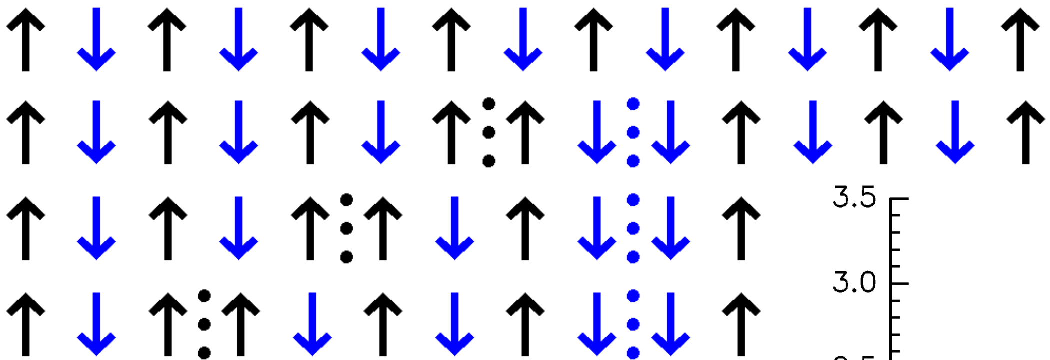


Ground state (Bethe 1931) – a soup of domain walls

Spinon excitations

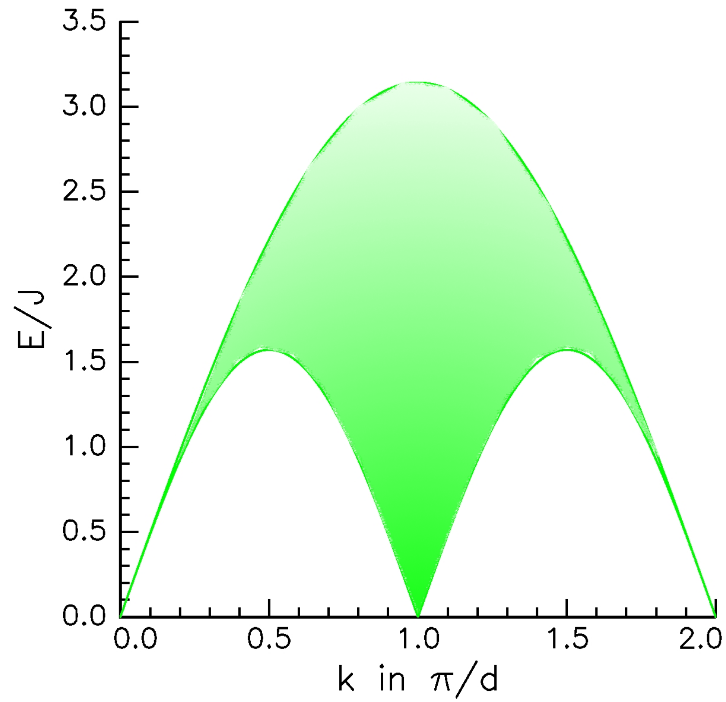
Elementary excitations:

- "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 ⇒



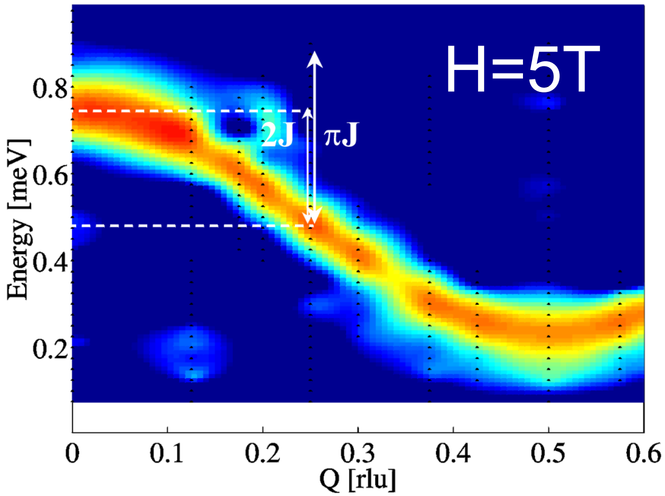
The *antiferromagnetic* spin chain

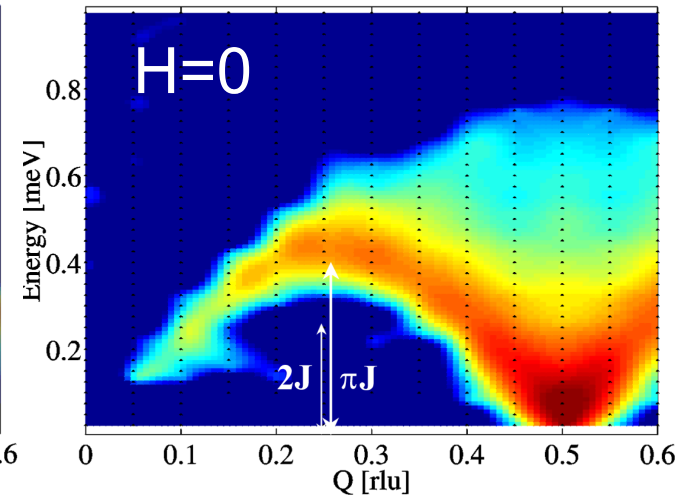
FM: ordered ground state (in 5T mag. field)

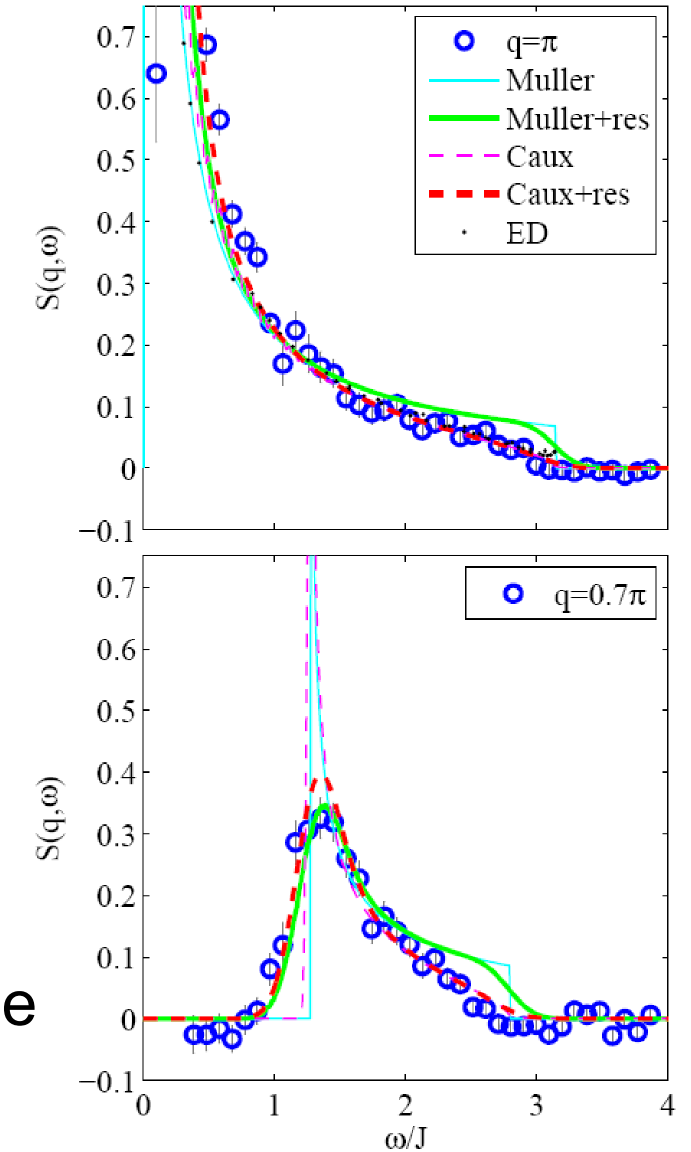
- semiclassical spin-wave excitations

AFM: quantum disordered ground state

- Staggered and singlet correlations
- Spinon excitations







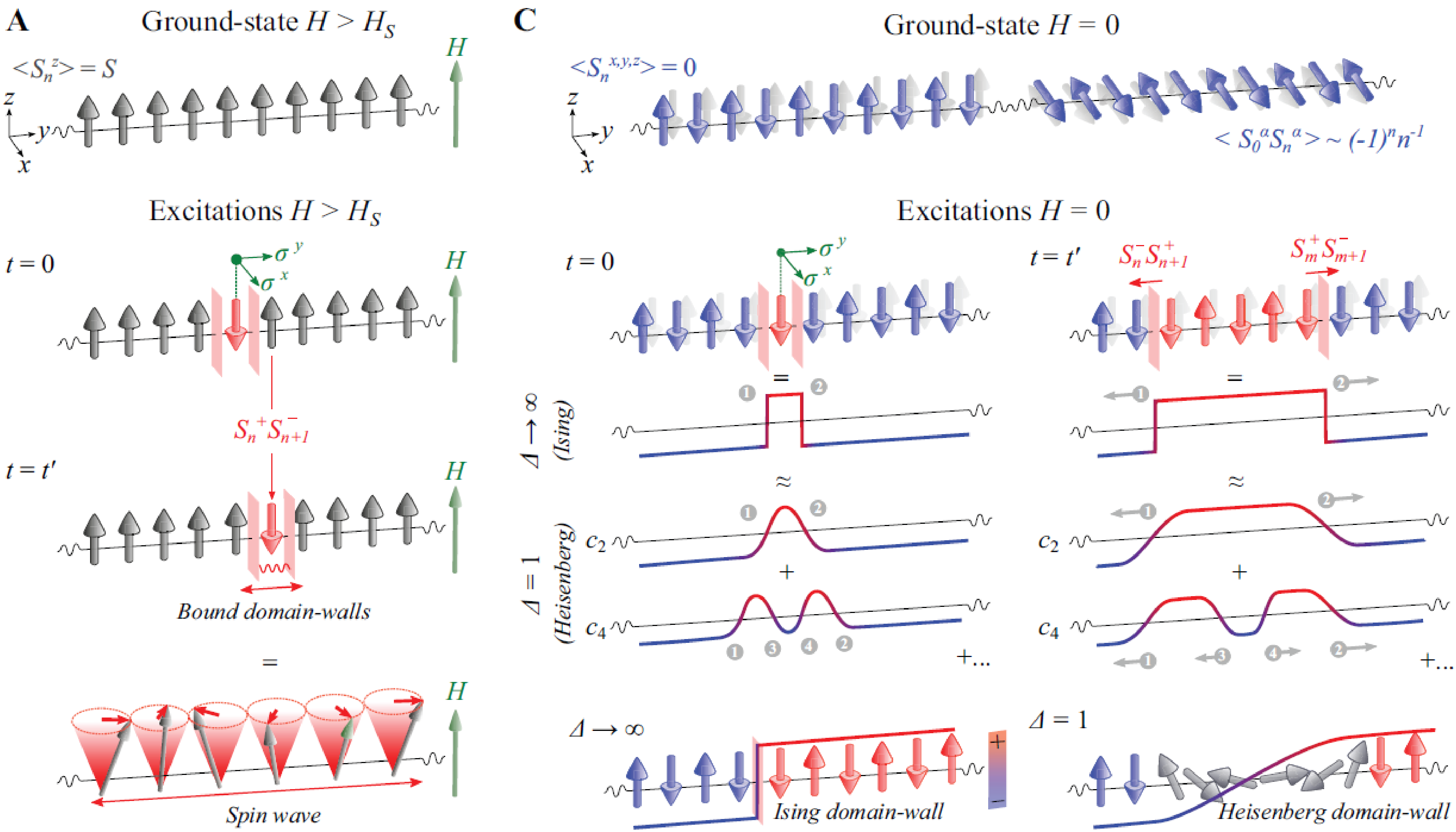
– Agebraic Bethe ansatz for inelastic lineshape

– Beyond Müller-conjecture ⇒

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

Spinons – our cartoon for excitations in 1D spin chain

Spin waves Spinons: 2- and 4 spinon states ?



Detecting 4-spinon states?

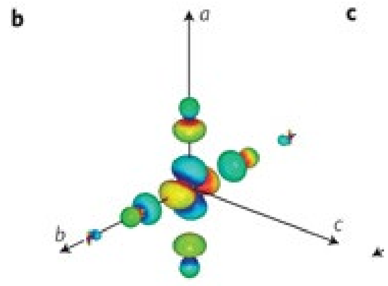
Neutrons see spinon continuum

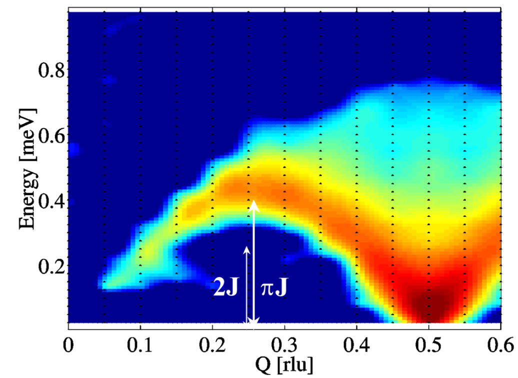
But, 2- and 4-spinon almost identical line-shap

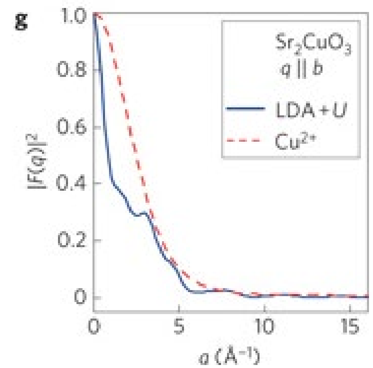
Only way to distinguish is absolute amplitude

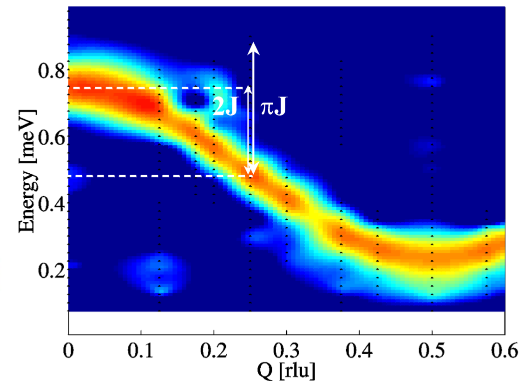
Previous attempts uncertainty in form factor

Trick: Normalise to ferromagnetic spin-waves









Intensity = instrument-stuff * cross-section

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

cross-section

pre factor

dipole factor

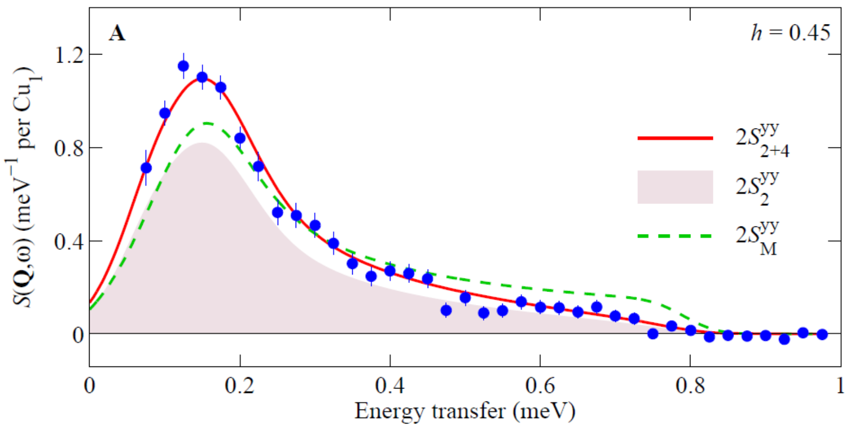
magnetic form factor

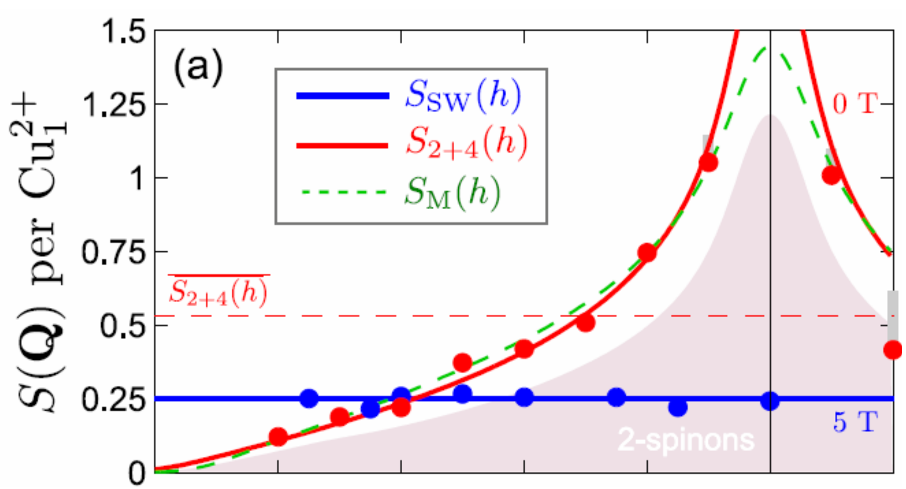
Fourier transform

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 73% 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)

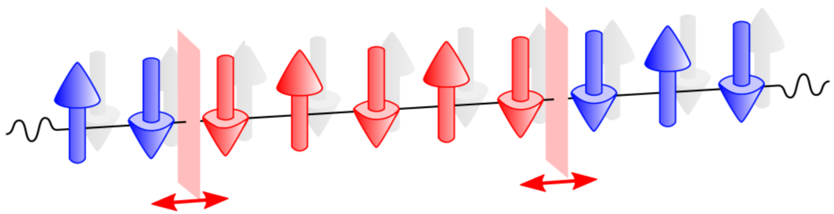
- Possible combinatorial arguments?

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

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

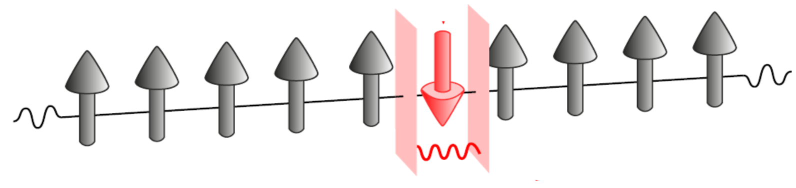
Intermediate fields – a teaser

- ❑ $H = 0$ (Spinon vacuum)



2- + 4- spinon

- ❑ $H > H_s$ (Magnon vacuum)



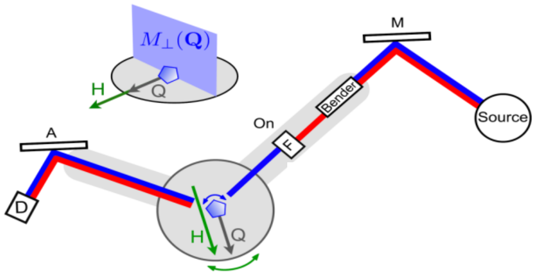
1- magnon

- ❑ $0 < H < Hs$ (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 $\Rightarrow$ Several new quasi-particles observed



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

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

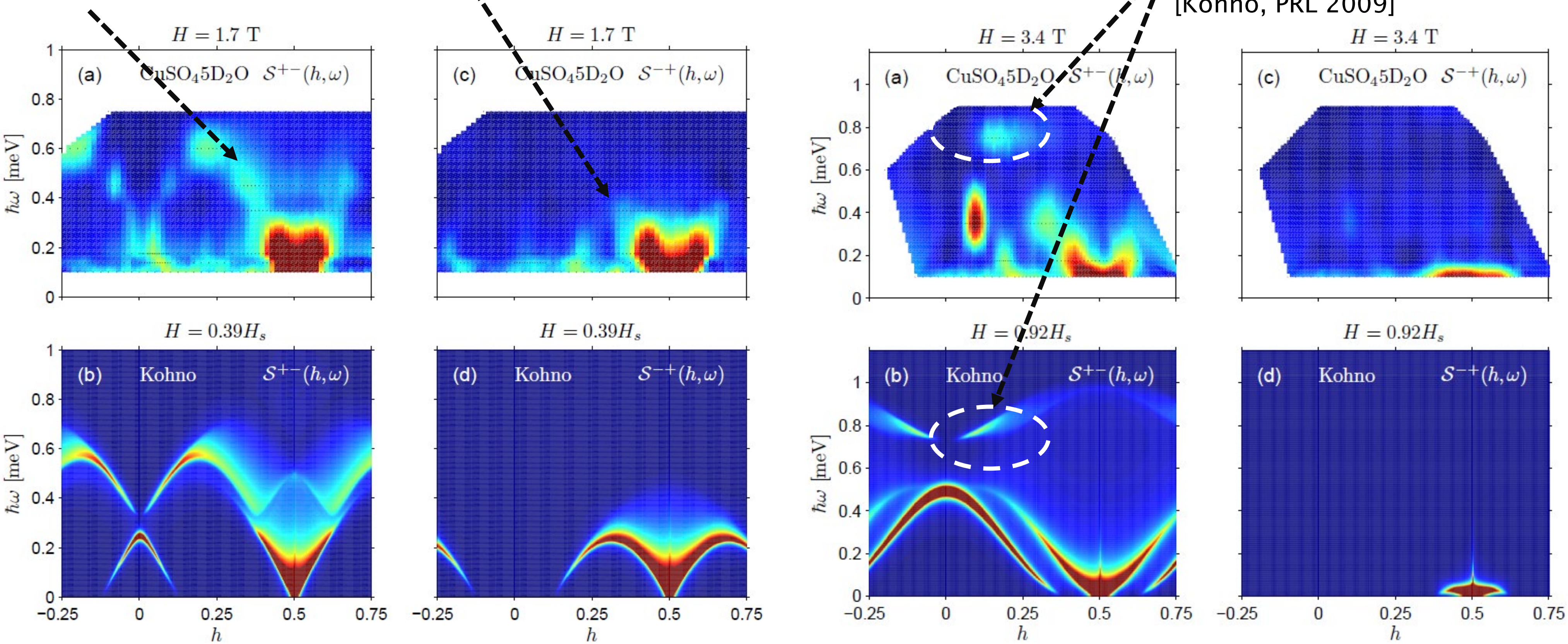
- more expt planned
- picture of these new excitations ?

σ_2 (2-string) ?

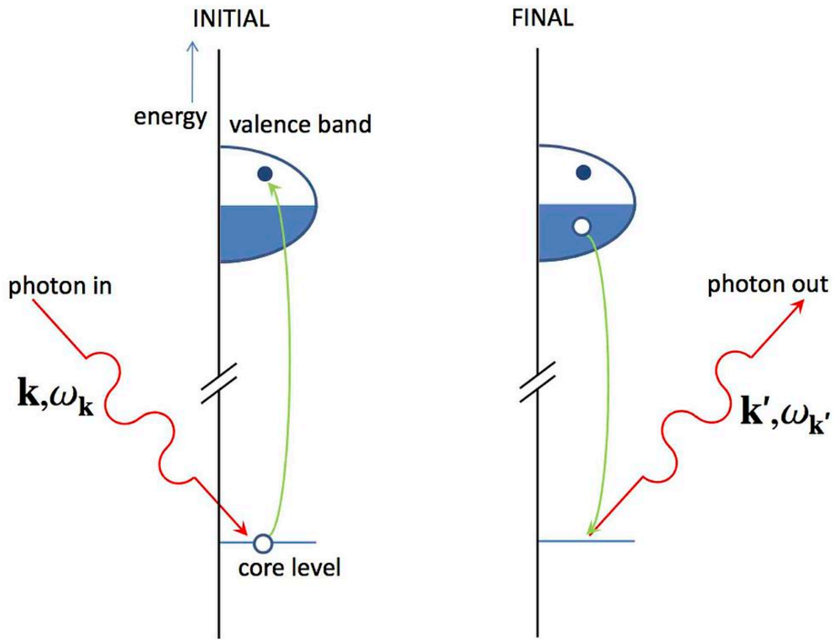
[Kohno, PRL 2009]

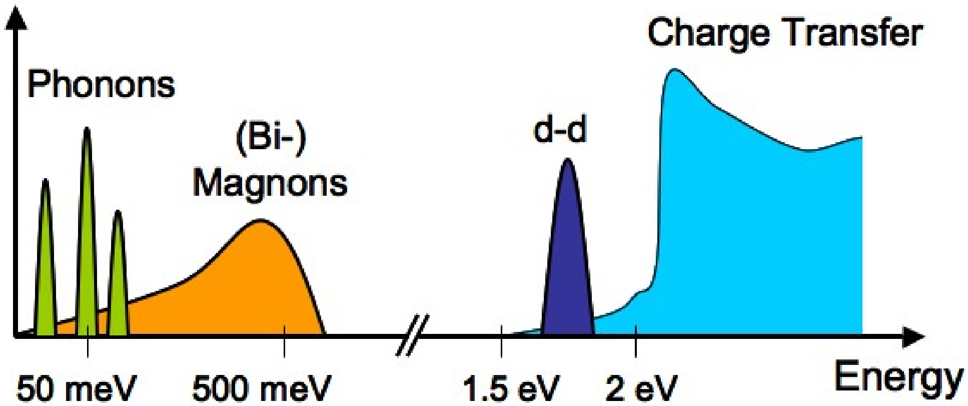
$\psi\psi^*$ in $\mathcal{S}^{+-}$

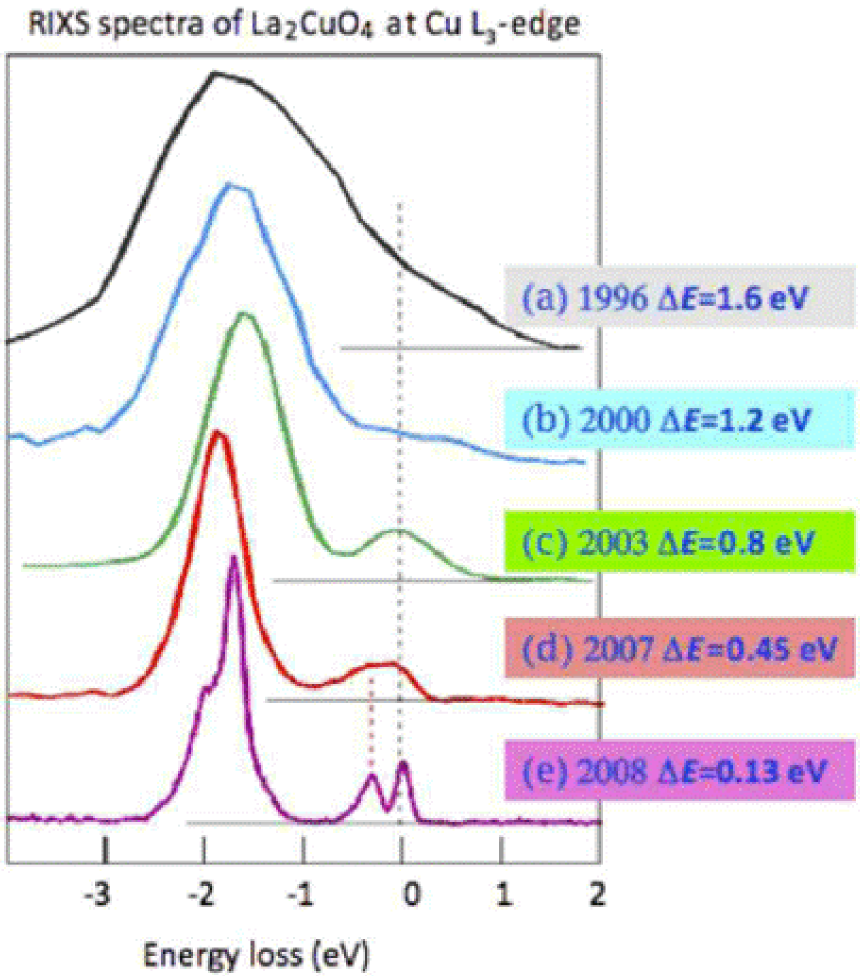
$\psi\psi$ in $\mathcal{S}^{-+}$



Resonant Inelastic X-ray scattering





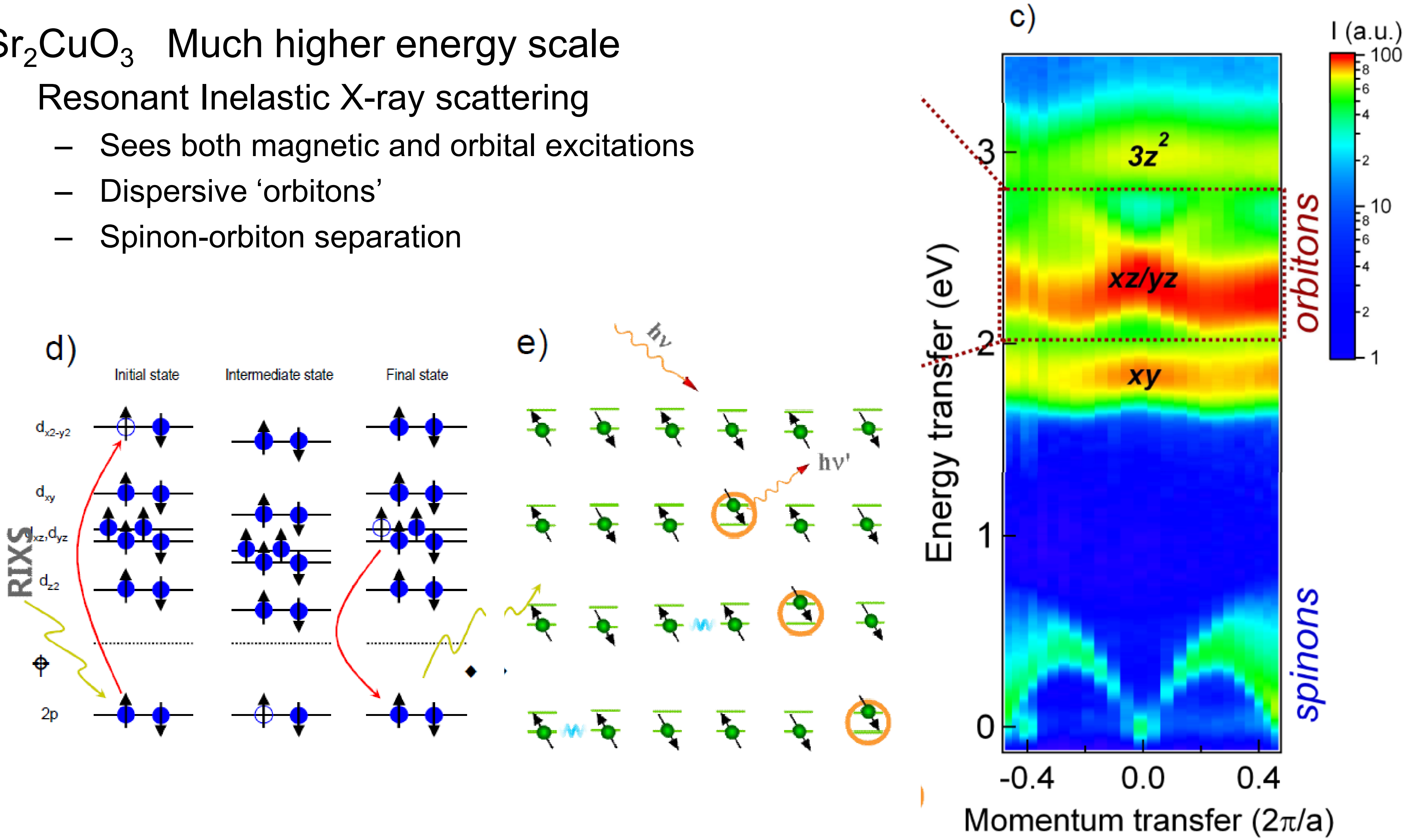


New measure of magnetic excitations

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

Electronic states of matter:

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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
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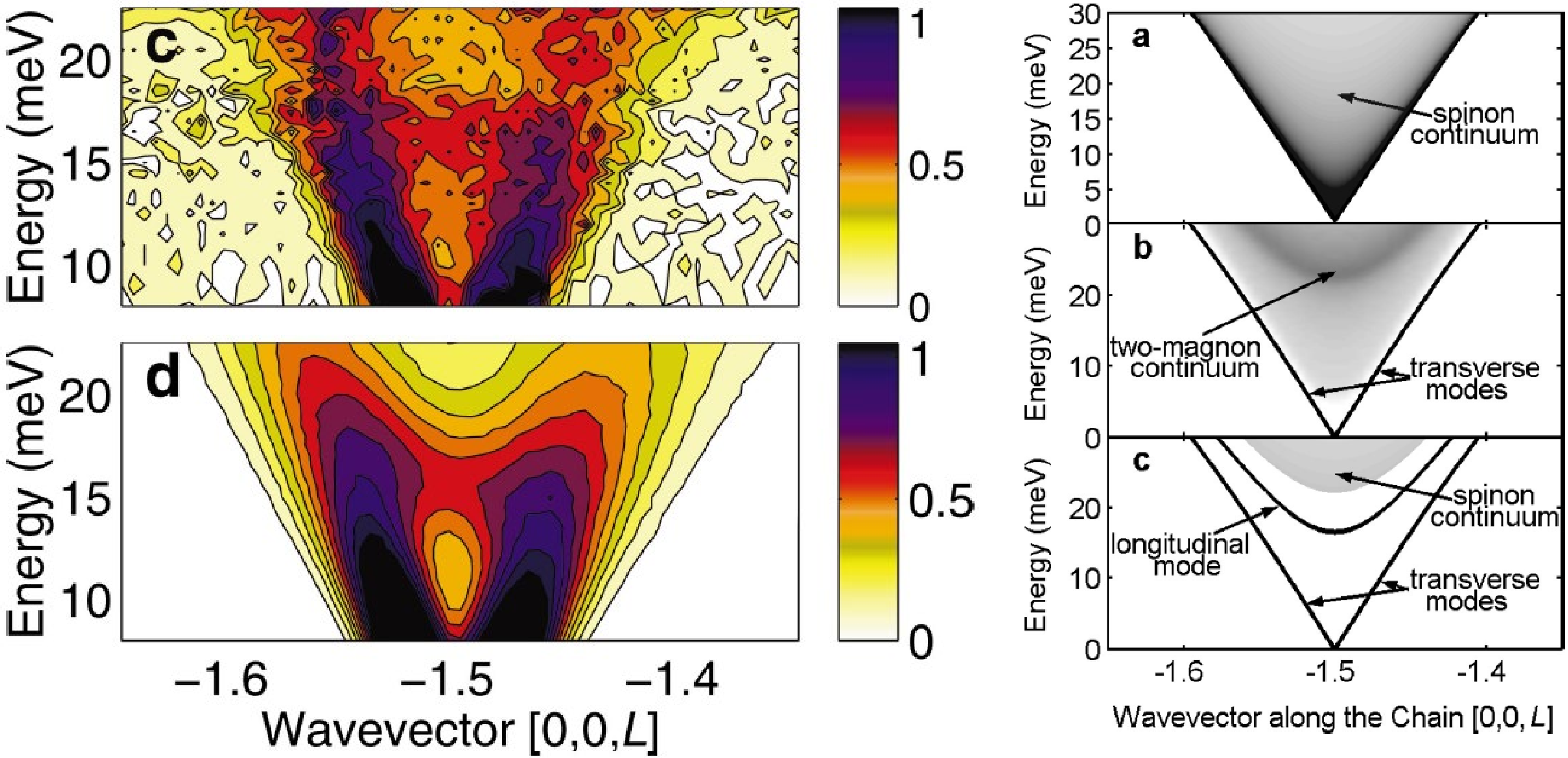
} collective quantum states

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)

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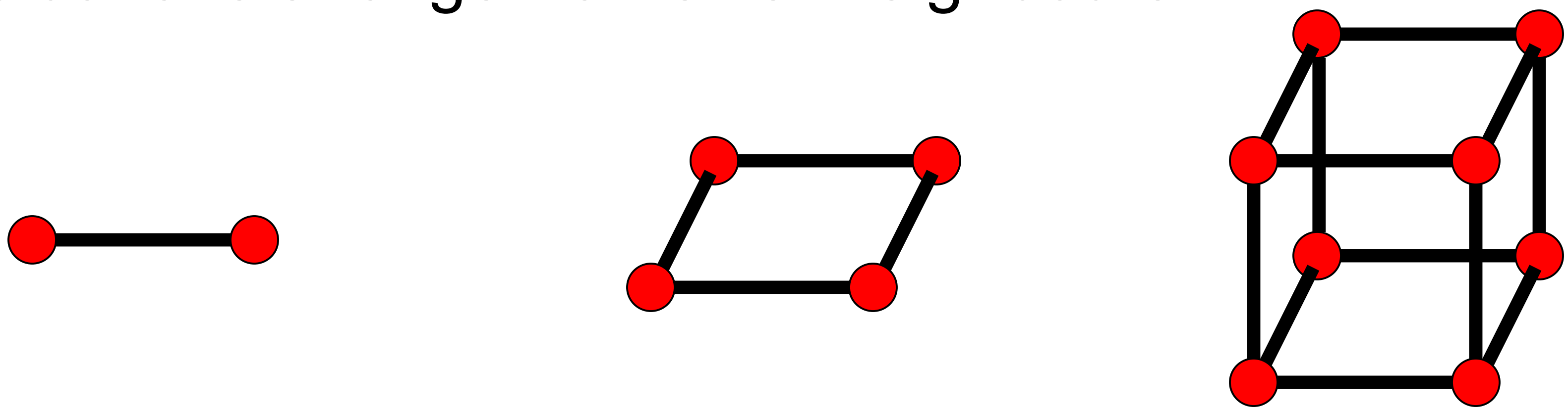
The 2D borderline

Fluctuations stronger for fewer neighbours



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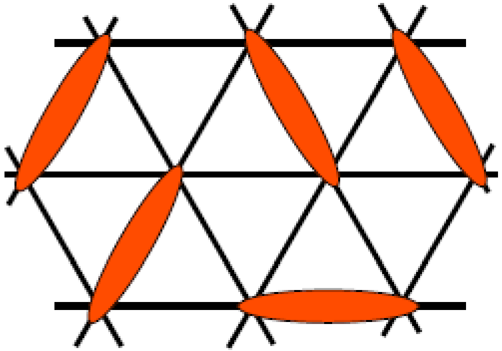
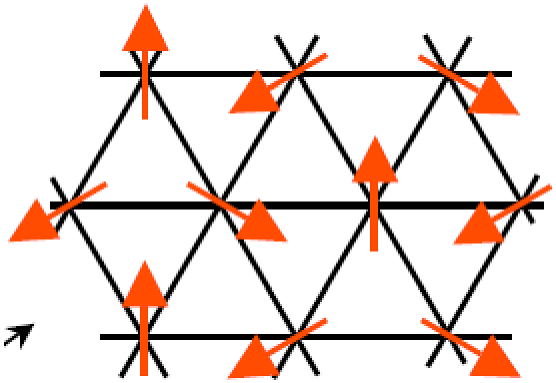
3D: Ground state long range ordered, very weak Q-effects

Valence Bonds and Anderson

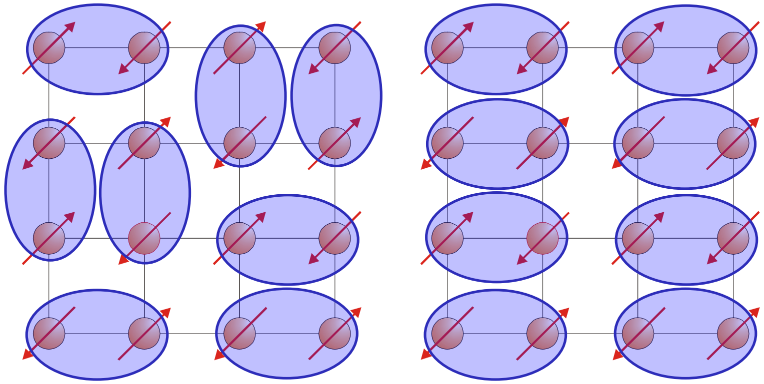
- 1973: Anderson suggests RVB on triangular lattice

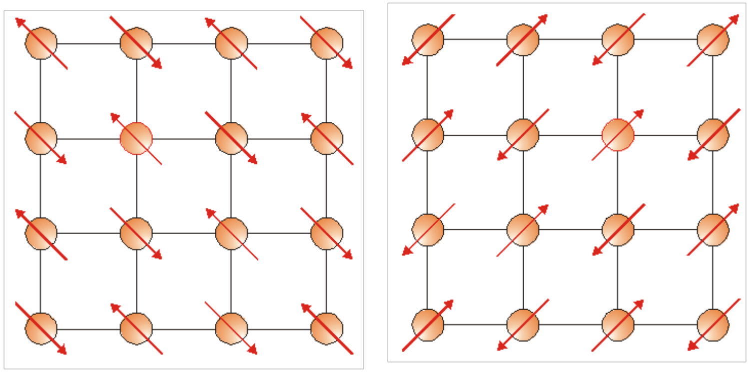




But - actually long range order

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





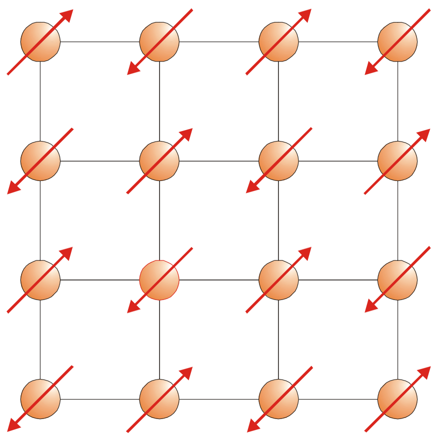
But - actually long range Neel order

Quantum Magnetism in Flatland

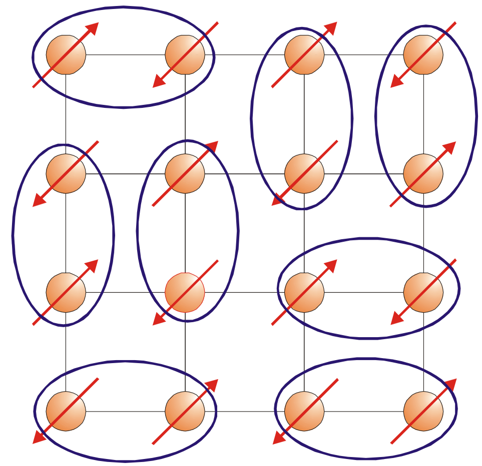
2D Heisenberg antiferromagnet on a square lattice

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

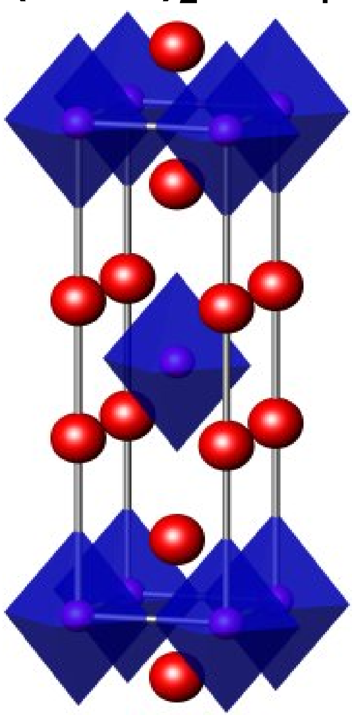
- Are there other types of 'correlations' ?
  – Resonating valence bonds (RVB)

} Investigate excitations with neutron scattering

Physical realisations

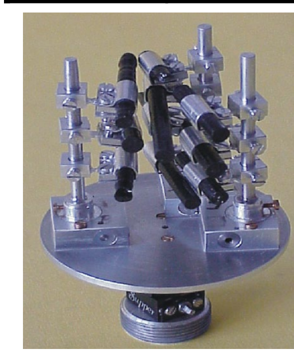
- Representation of model: No/small extra terms, anisotropy gaps etc.
- Energy scale: Zone boundary, resolution, temperature, field H_s

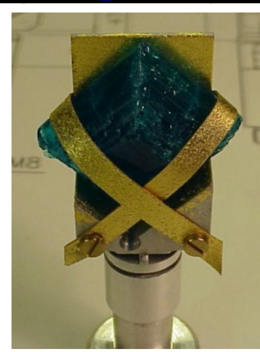
$(La,Sr)_2CuO_4$



CuO_2 planes

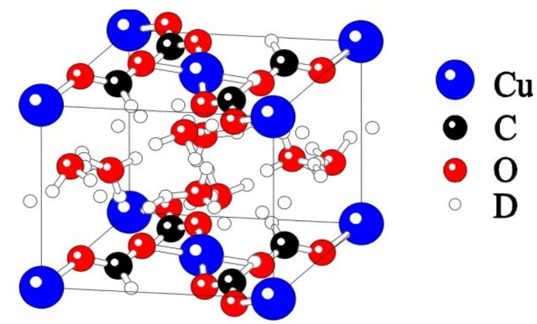
	La_2CuO_4	CFTD	CAPCuBr	CAPCuCl
J [K]	1500	73.3	8.5	1.2
J'/J	5×10^{-5}	4×10^{-5}	~ 0.1	~ 0.1
T_N [K]	325	16.4	5.1	0.64
H_S [T]	4500	220	24	3.4

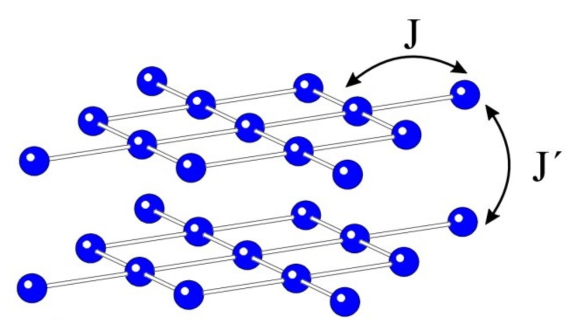




+ CuPzClO Tsyrulin & Kenzelmann

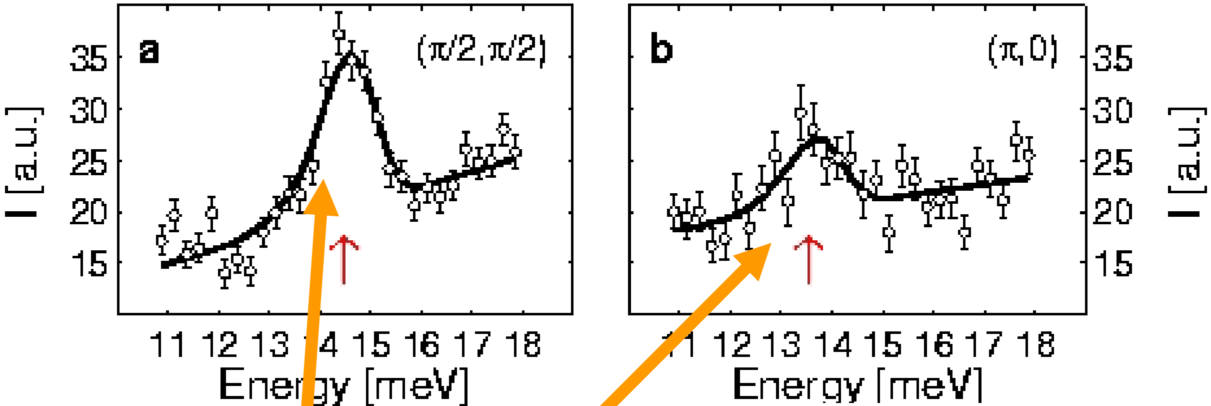
Copper Formate Tetra-Deuterate





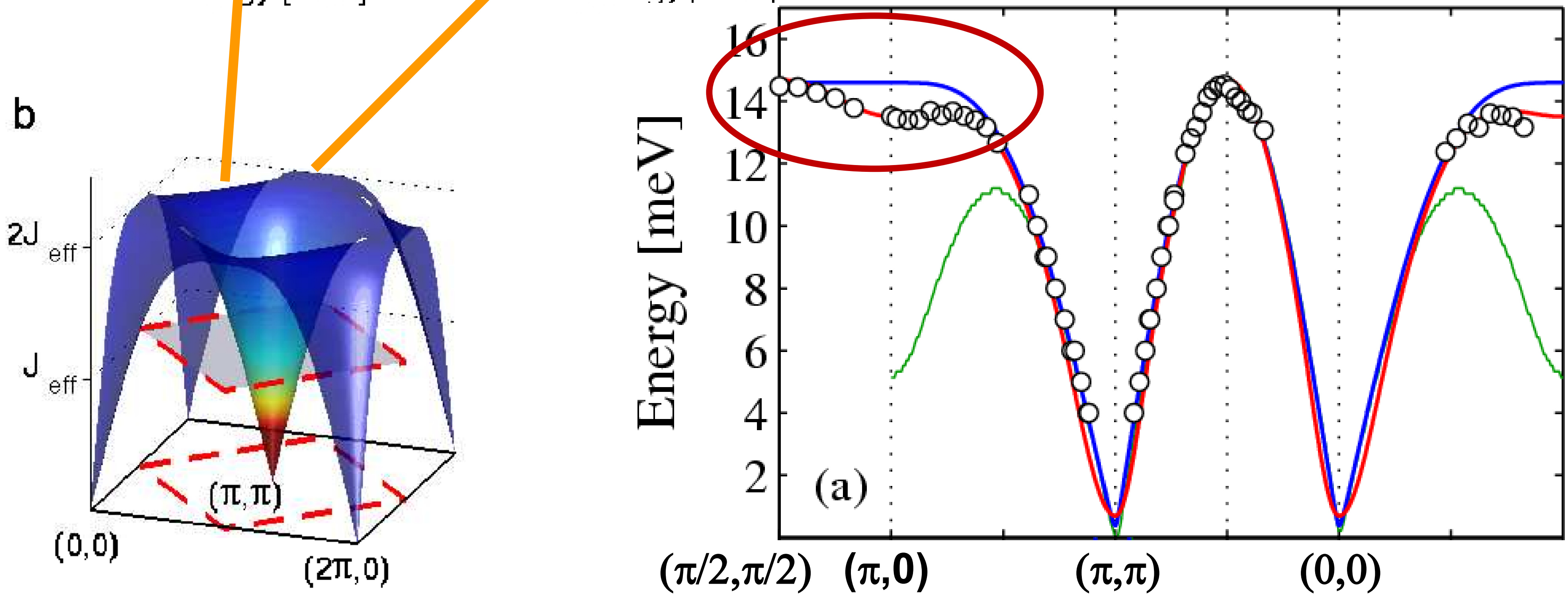
$Cu(DCO_2)_2 \cdot 4D_2O$

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

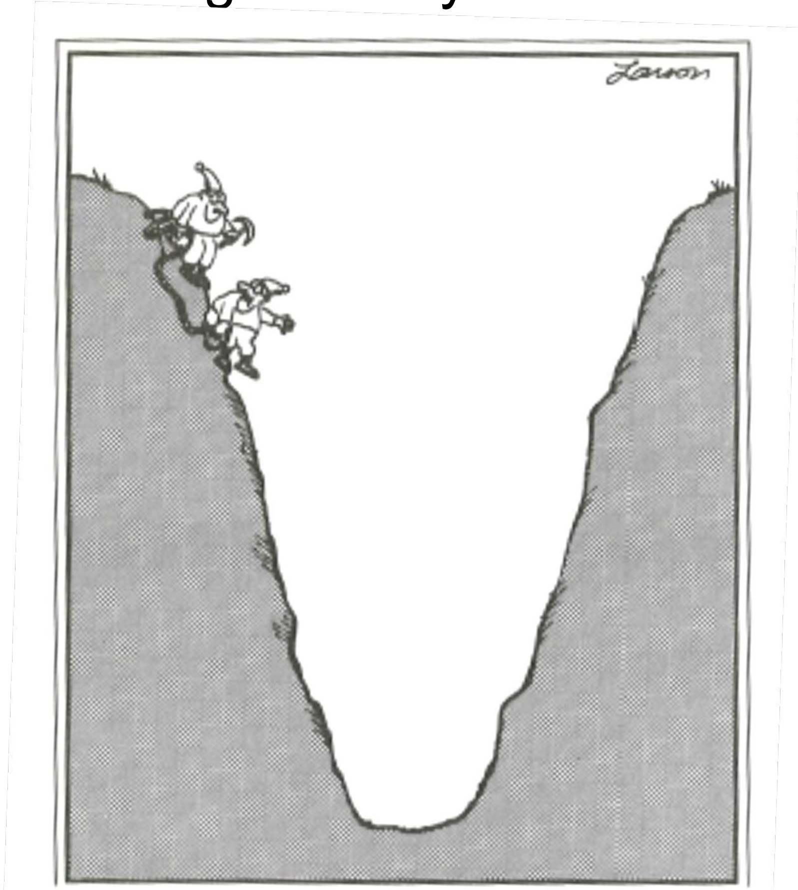


Magnon intensities

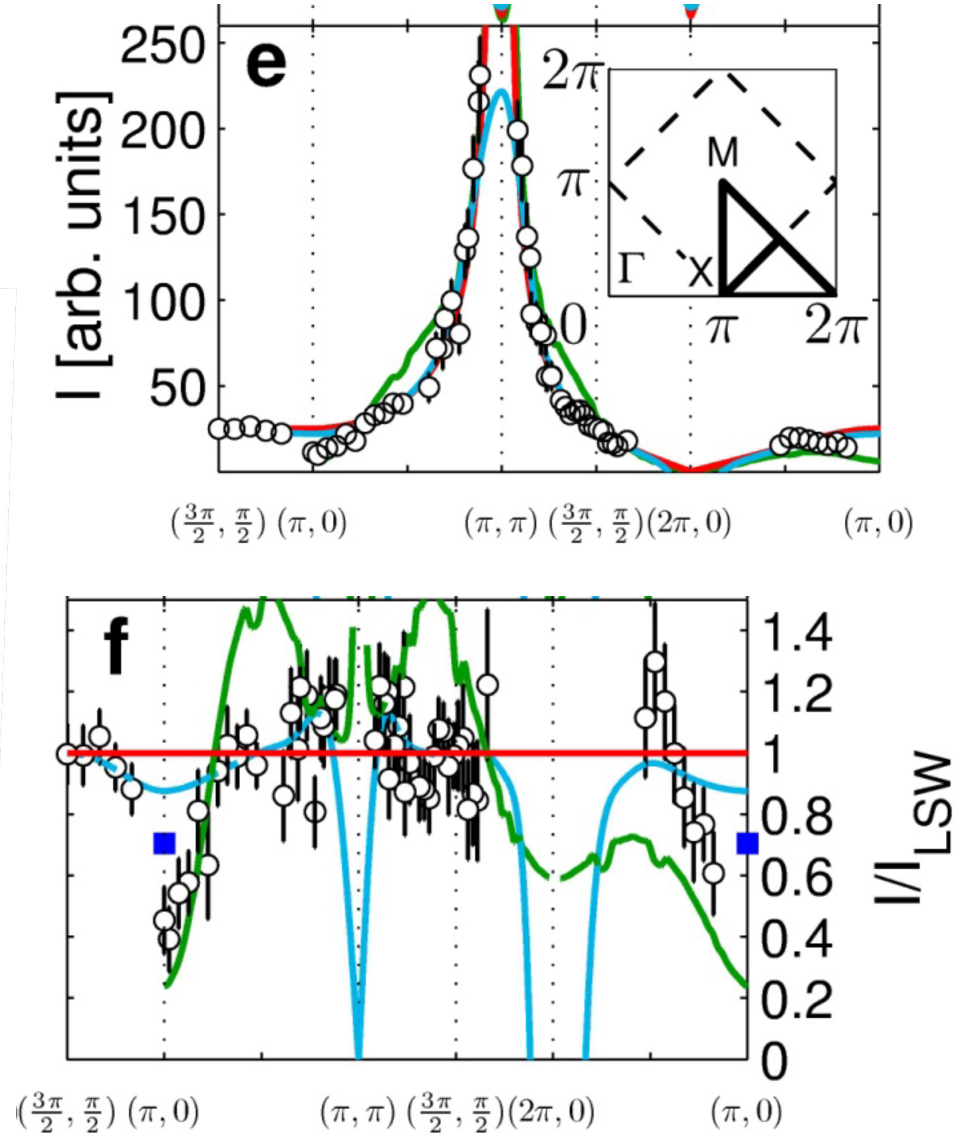
Giant 50% intensity effect at $(\pi,0)$

Remember SW already 51% reduced

$\Rightarrow$ A tale of missing intensity !

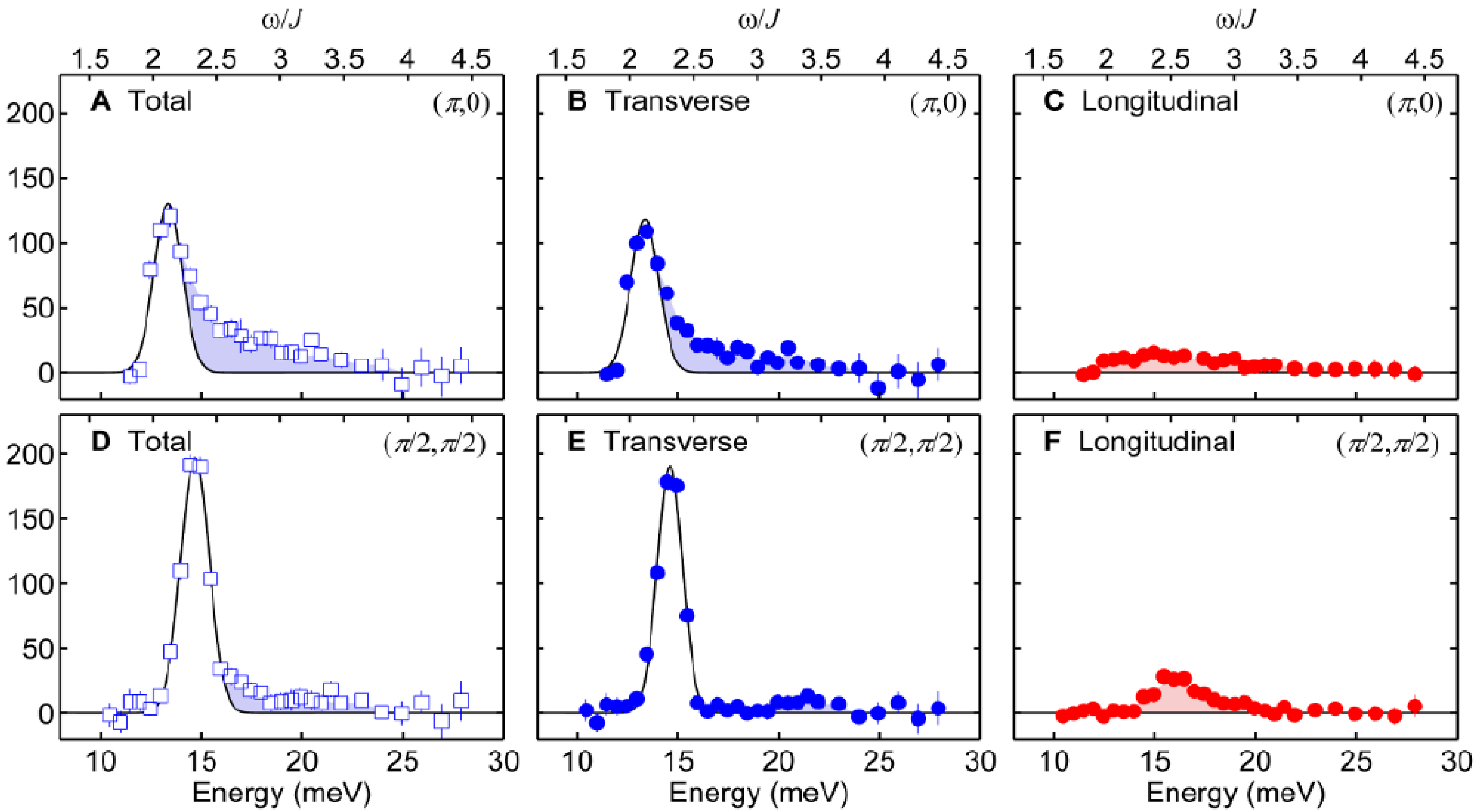


"Because it's not there."



Christensen PNAS **104** 15264 (2007)

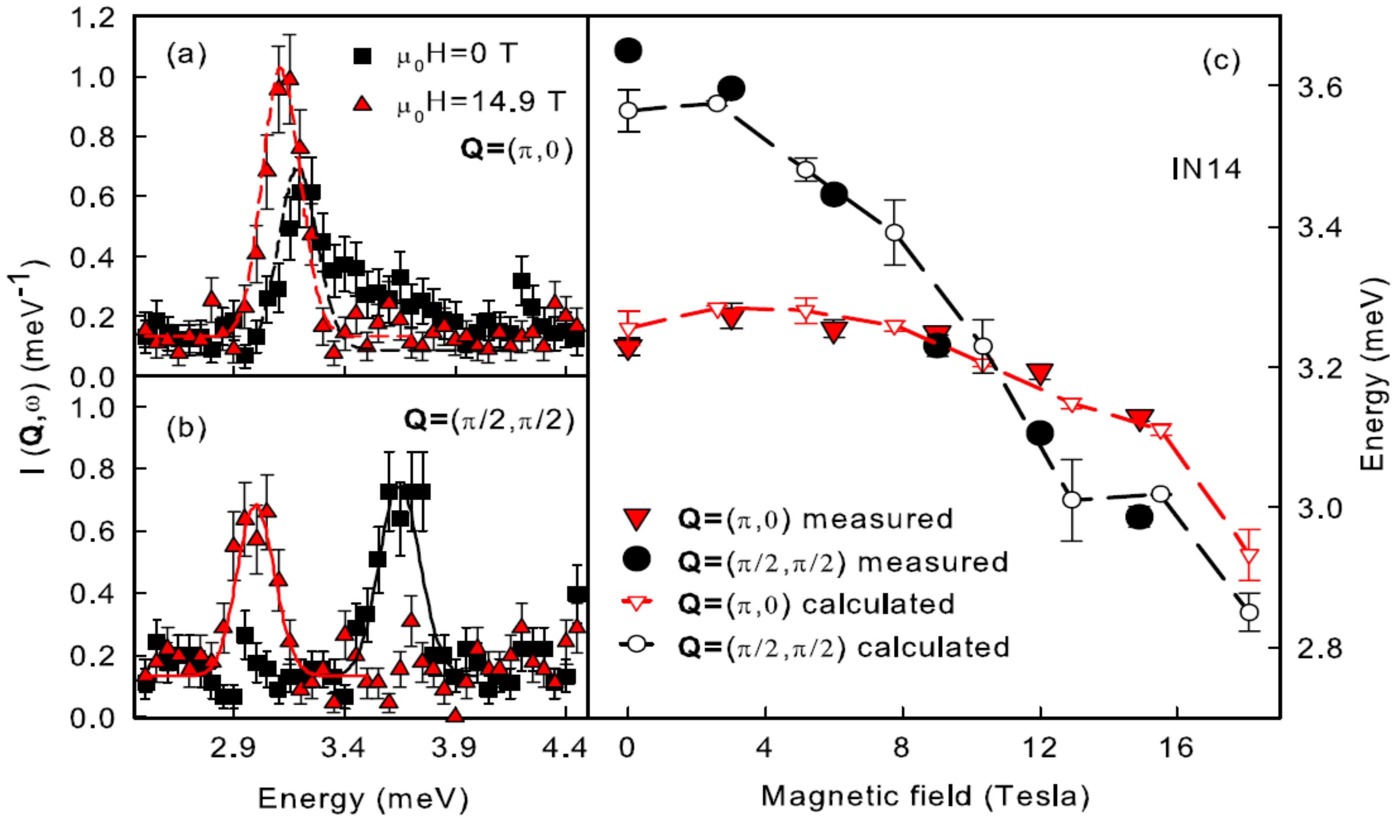
Polarised neutrons: Line-shapes at the Zone Boundary



Both longitudinal and transverse continuum

Same phenomenon in all S=1/2 square lattice AFMs

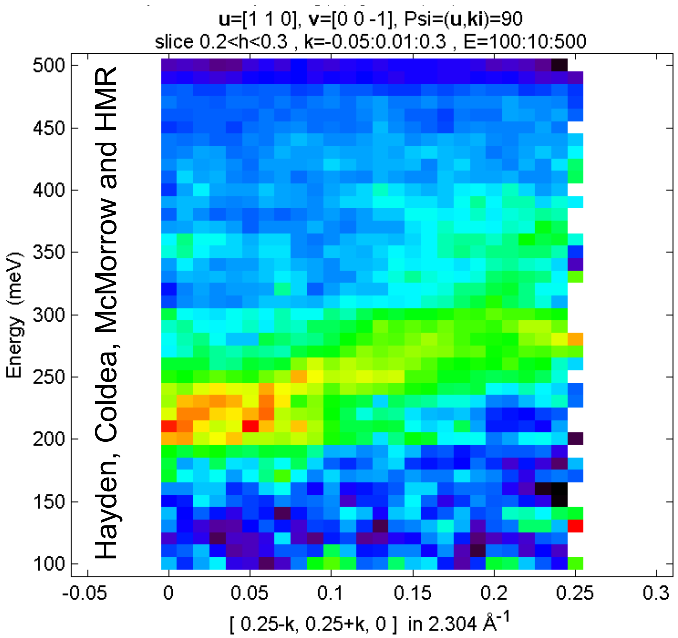
- $Cu(pz)_2(ClO_4)_2$ ZB with diagonal J_{nnn}



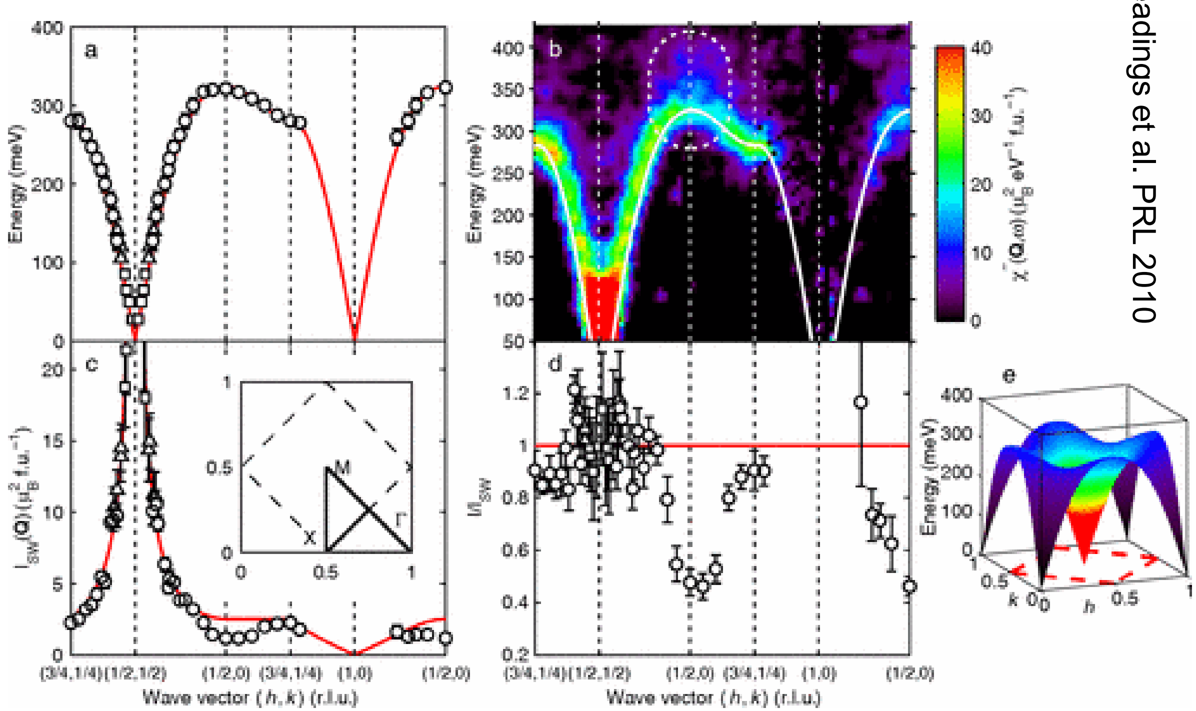
N. Tsyrulin… A. Schneidewind, P. Link…M. Kenzelmann, Phys. Rev. B **81**, 134409 (2010); Phys. Rev. Lett. **102**, 197201 (2009)

quantum anomaly also in cuprates !

$YBa_2Cu_3O_{6.1}$



La_2CuO_4

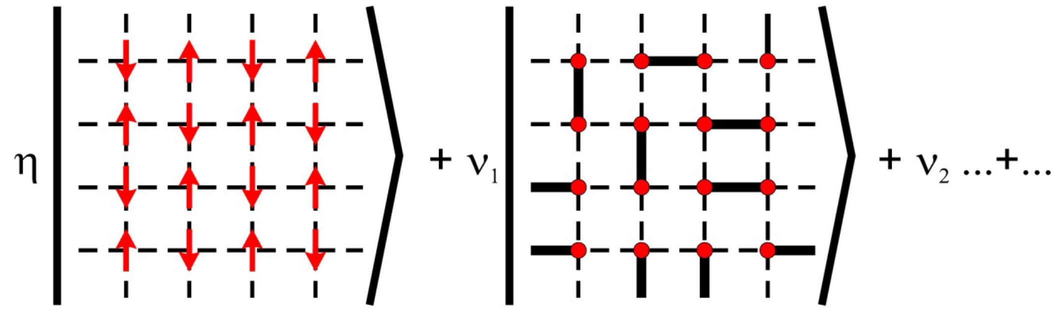


Headings et al. PRL 2010

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

simple experimentalist's picture:

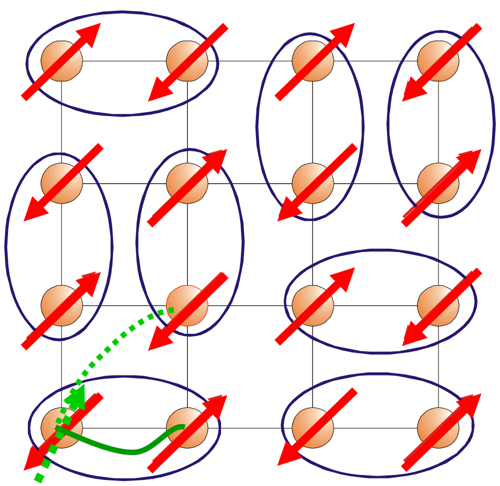
The missing 40% Neel order partly resides in n.n. singlet correlations



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

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

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



Bond energies:

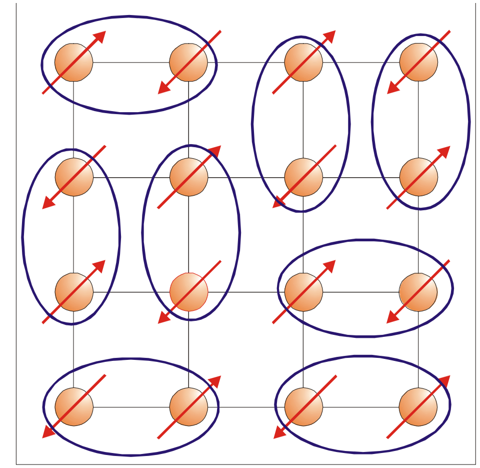
- Classical spins $E_b = -JS^2 = -0.25J$
- Best estimates $E_b \approx -0.34J$

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 !

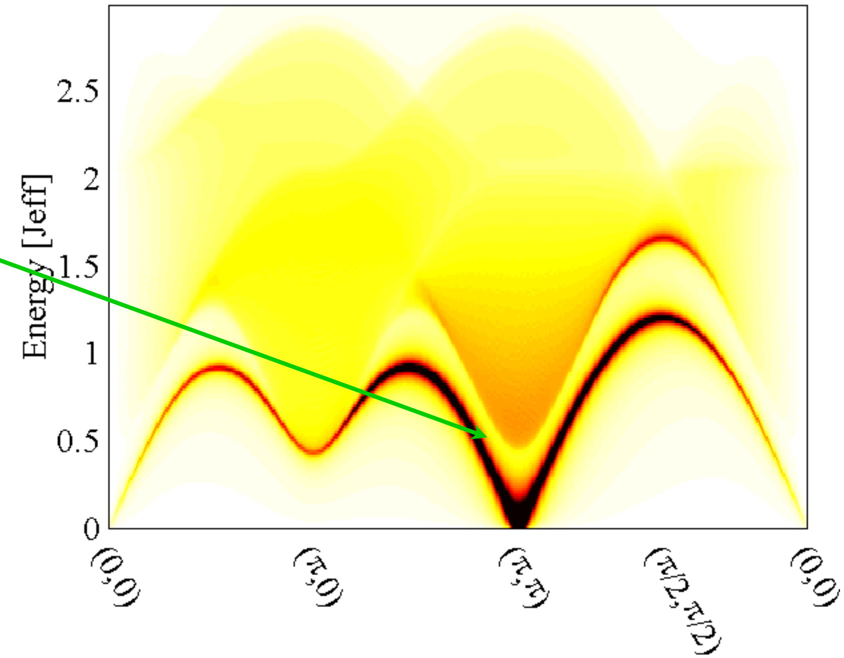
Insert magnetization by hand ⇒ π-flux state

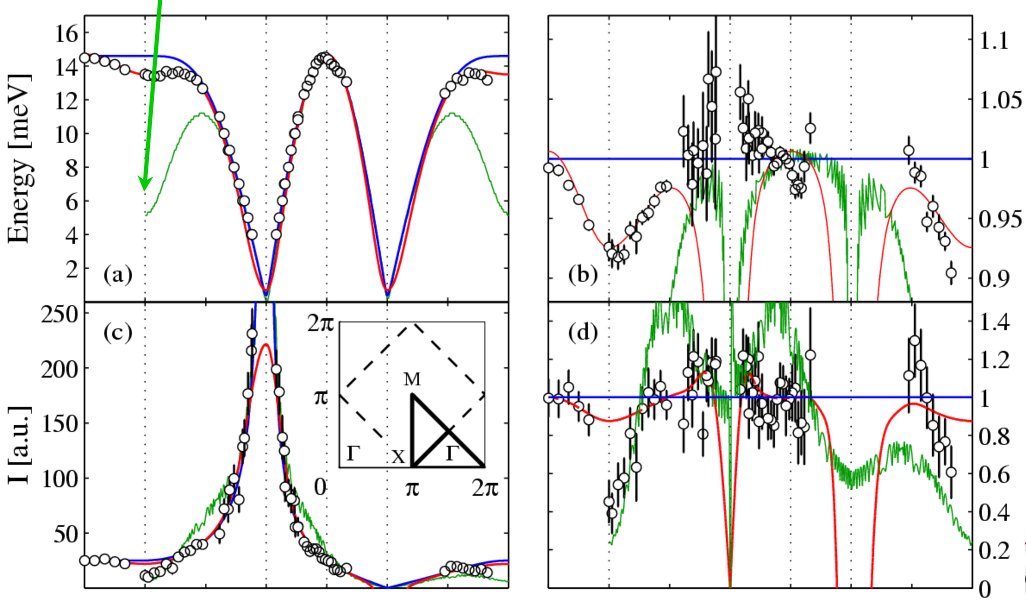
right trend, but too much:

ZB dispersion at (π,0)

but, Gap at (π,π)

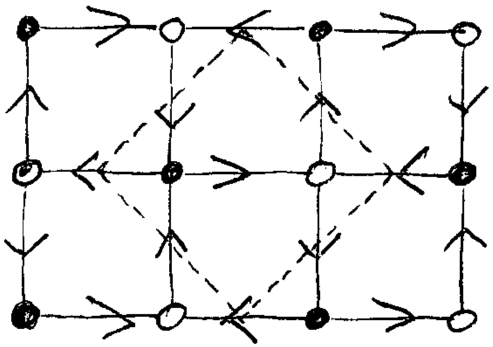
Need better theory





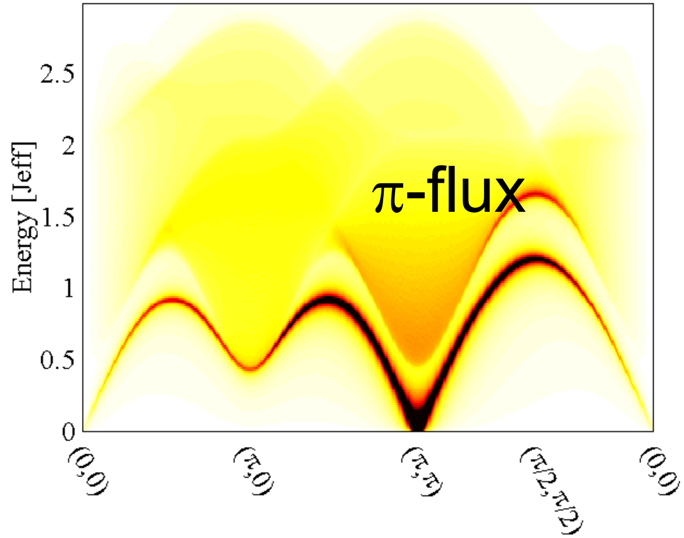
Anderson Science **235** 1196 (1987)
Hsu PRB **41** 11379 (1990); Ho, Ogota, Muthumukar & Anderson PRL (2001),
Syljuasen *et al.* PRL **88** 207207 (2002)

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)

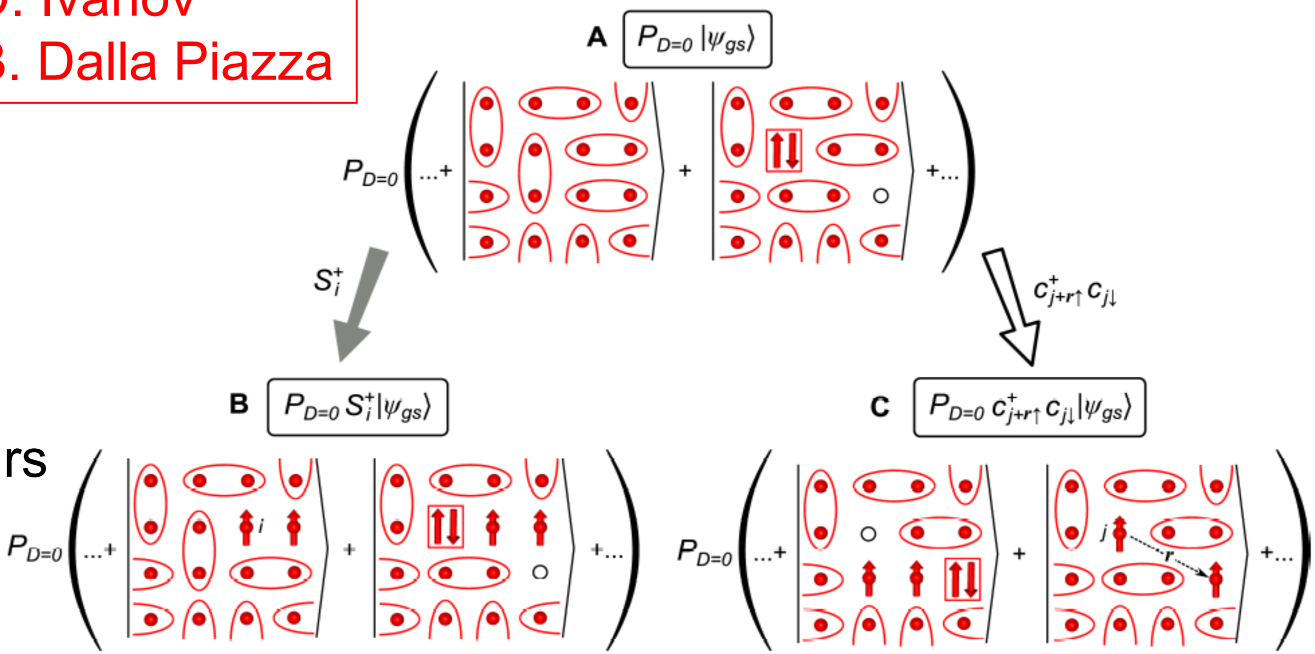


Work in Fermionic space

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

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

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

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

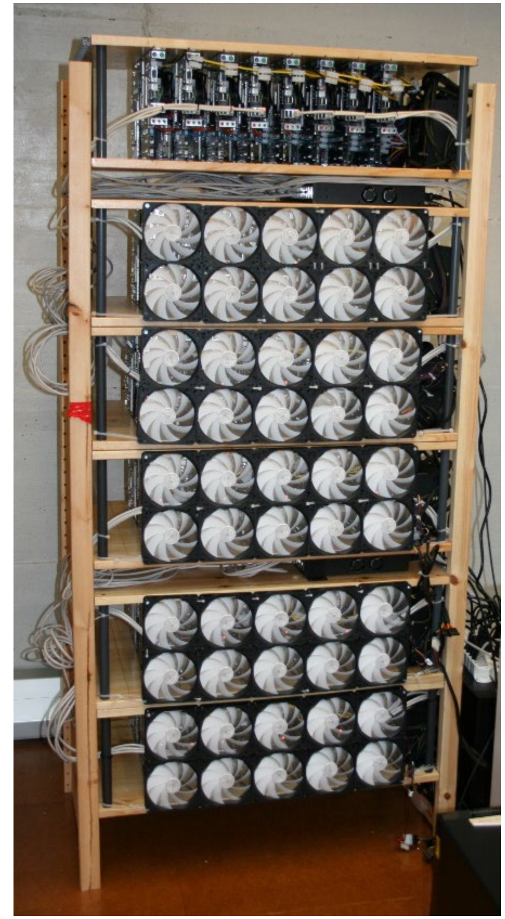
7m CPU hours later ….

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
Open for collaborations

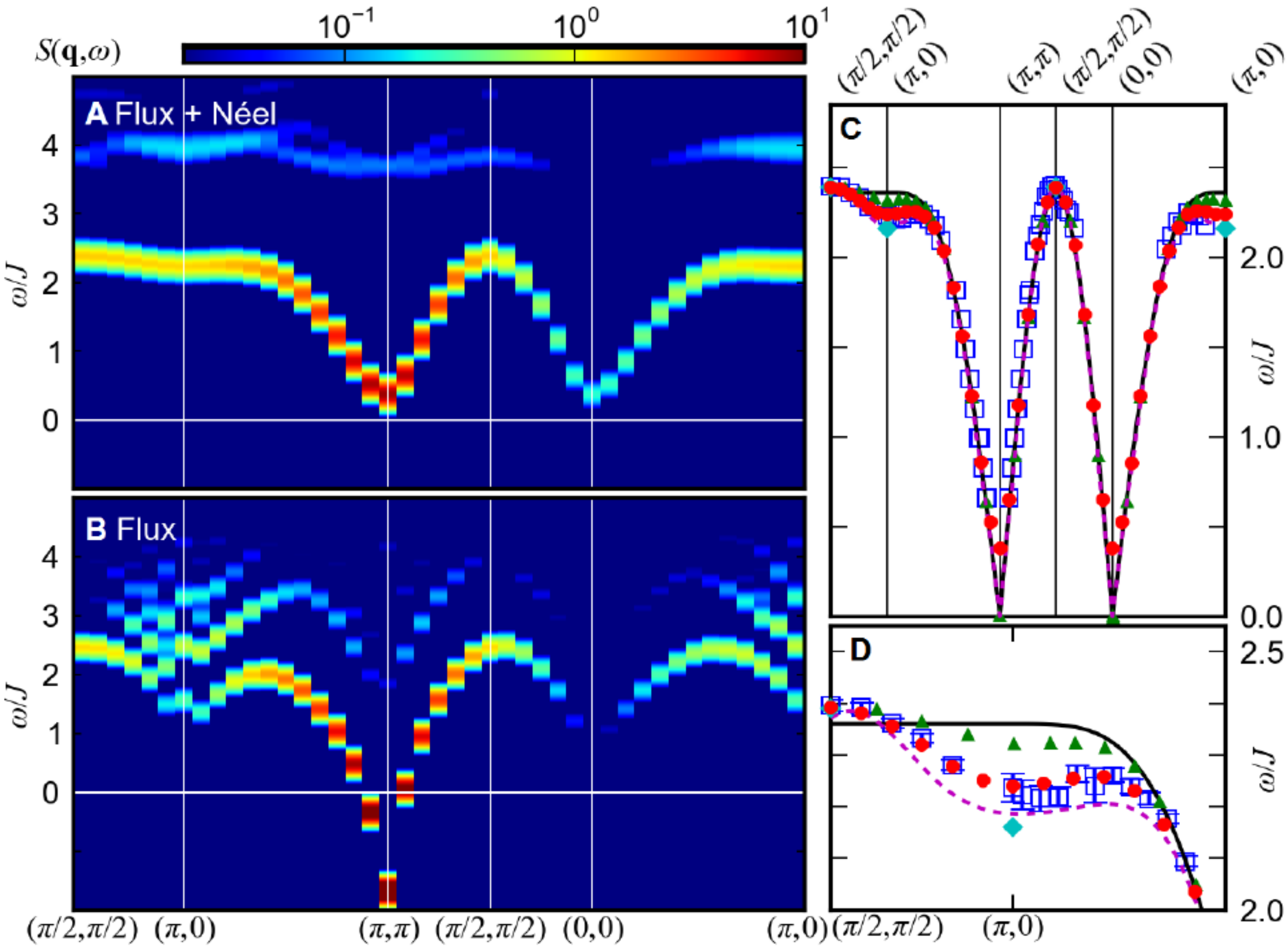
Not perfect, but captures the features:

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-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

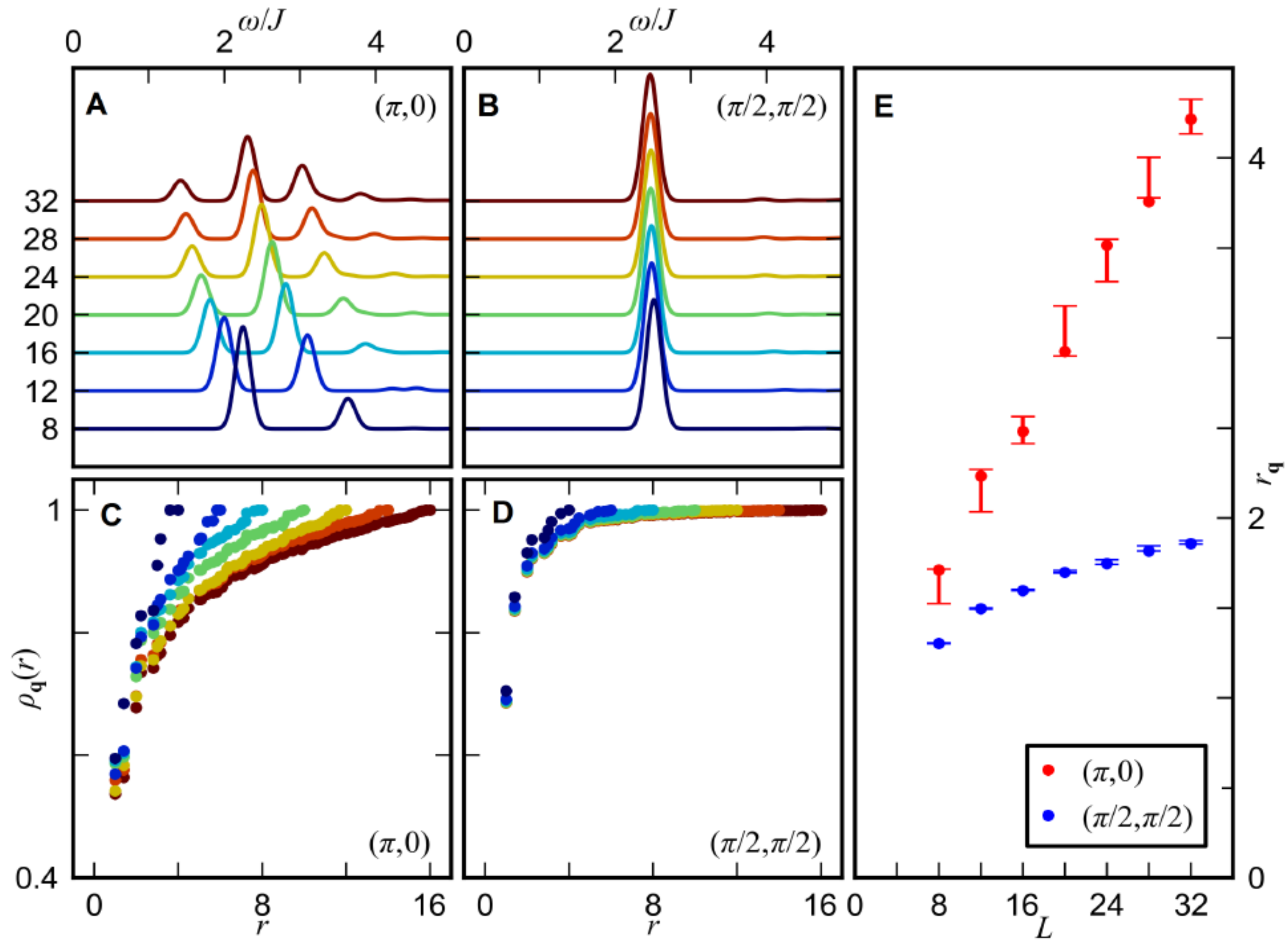
$$\tilde{\rho}_{\mathbf{q}}(\mathbf{r}) = \sum_n |_t\langle \mathbf{q},\mathbf{r}|\mathbf{q},n\rangle_{tt}\langle \mathbf{q},n|\mathbf{q},0\rangle_t|^2$$

(π/2, π/2) converge and has short spinon separation

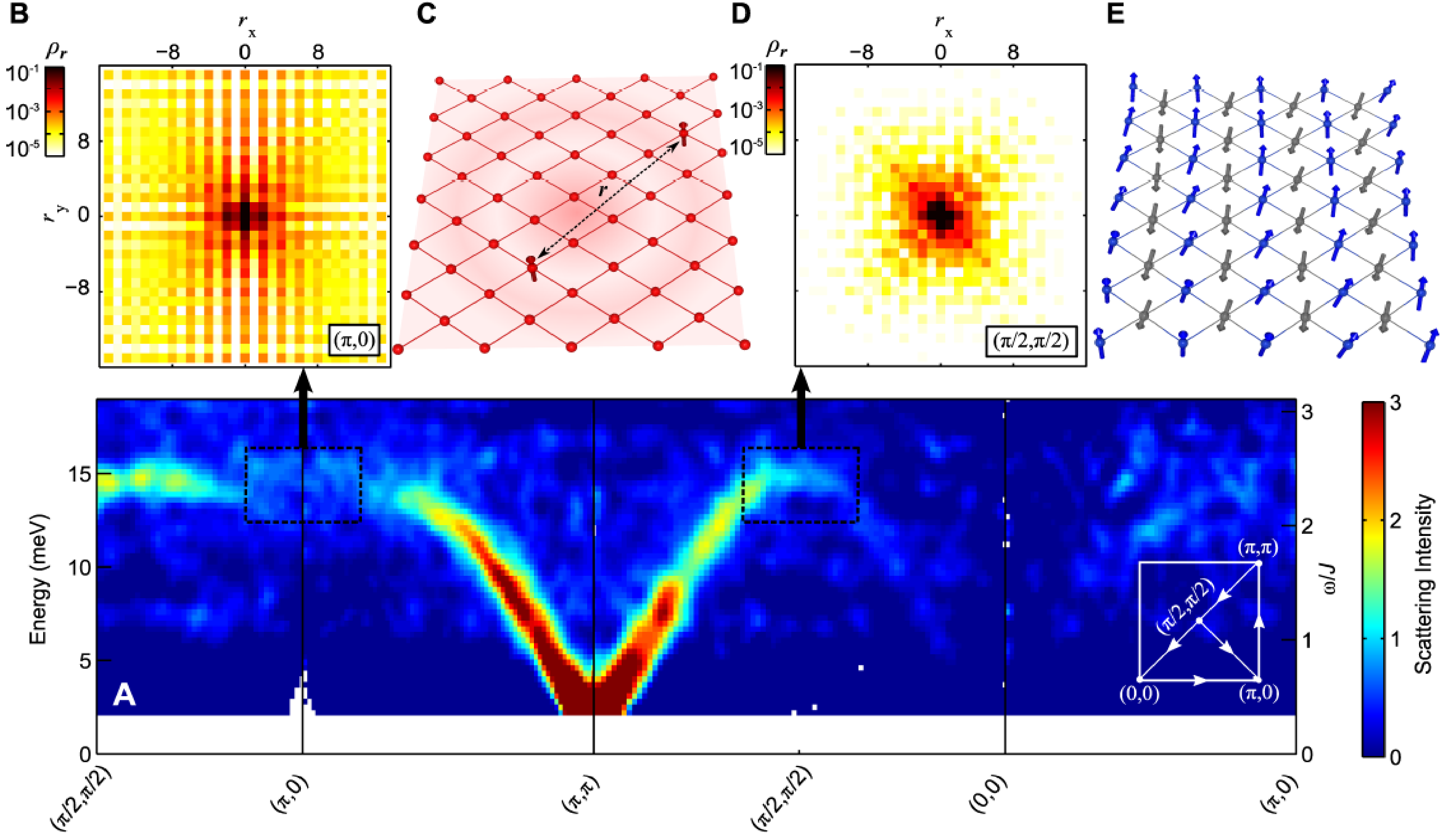
⇒ spin-waves

(π, 0) grow linear with system size

⇒ spinons deconfine !

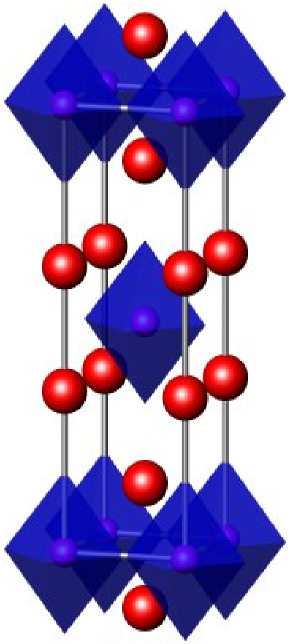


Spinons in 2D square lattice !



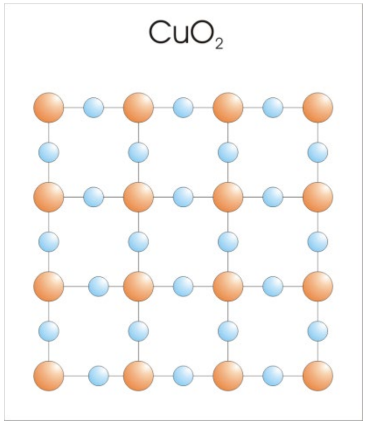
B. Dalla Piazza, M. Mourigal, D. Ivanov et al. Nat. Phys. **11**, 62 (2014)

RVB in 2D magnet – *so what* ?



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

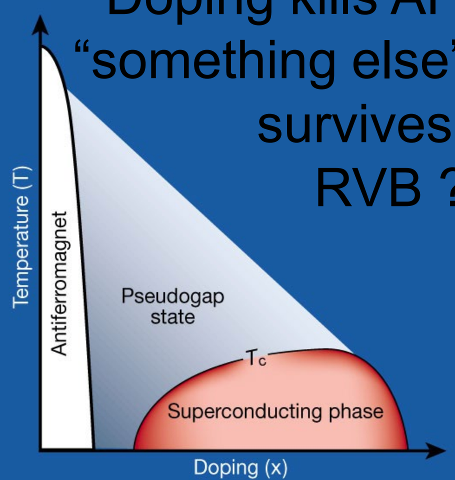
J. Phys.: Condens. Matter **16** (2004) R755–R769 PII: S0953-8984(04)80644-1

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]

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



Is ZB anomaly the smoking gun of RVB ?

Conclusion

- Quantum magnets as quantum simulators for exotic ground states and excitations
  - Order, quantum spin-liquid, valence bond states etc
  - Spin-flips, triplons, spin-waves, spinons,
- Neutron spectroscopy allow detailed testing / guiding of theory
- 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
  - Coexistence of magnetic order and quantum fluctuations
  - Fractional excitations can exist in un-frustrated 2D models
  - Implications: role in high-Tc superconductivity?