Winter School for Quantum Magnetism

EPFL and MPI Stuttgart

Magnetism in Strongly Correlated Systems

Vladimir Hinkov

1. Introduction – Excitations and broken symmetry
2. Spin waves in the Heisenberg model
3. Reduced dimensionality and quantum effects
4. Magnetic excitations in hole doped cuprates

1. Excitations and Broken Symmetry

a. Ordering phenomena and broken symmetry

b. Long-range order and dimensionality – the Mermin-Wagner-Berezinskii theorem

c. Collective excitations, the Goldstone theorem

Stephen Blundell, *Magnetism in condensed matter*, Oxford master series

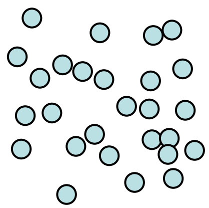
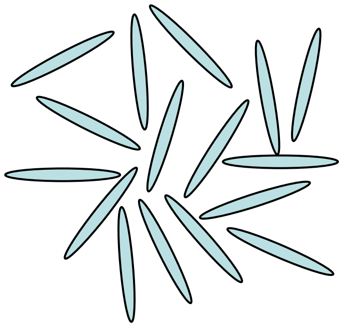
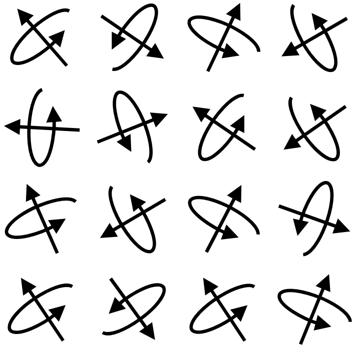
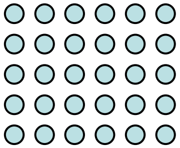
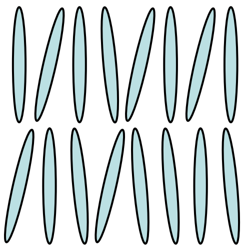
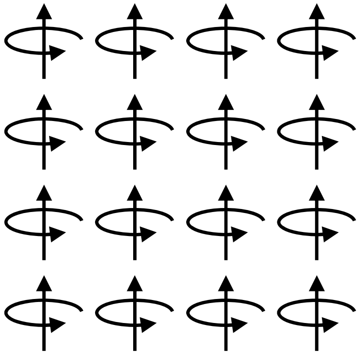
P. M. Chaikin and T. C. Lubensky, *Principles of condensed matter physics*, Cambridge UP

P. W. Anderson, *Basic notions of condensed matter physics*

F. Duan and J. Guojun, *Introduction to condensed matter physics I*, World Scientific

Ordering and broken symmetry

Hamiltonian H is invariant under symmetry transformations $\mathrm{T} \in G$

	G: translational group	G_1: rotational group G_2: translat. group		G_1: rotational group G_2: time reversal
$T > T_c$ symmetric	 liquid/gaseous	 liquid		 paramagnetic
$T < T_c$ symmetry broken	 solid	 nematic (G_1)	 smectic (G_1+G_2)	 ferromagnetic (G_1+G_2) (Heisenberg)

Phenomena associated with broken symmetry

- Symmetry is broken **spontaneously**, *H* retains full symmetry
- Symmetry does *not* change *gradually*, sharp **phase transition**, e.g. due to requirement to minimize the free energy $F = E - TS$
- State described by **order parameter** Ψ (vanishes in symmetric state): magnetization **M**, SC order parameter $|\Psi|\, e^{i\phi}$
- Attempts to restore full symmetry encounter resistance or **rigidity**: rigidity of a solid, permanent magnetization, stiffness of condensate
- Ordered state can support **collective excitations**, which weaken the order: phonons, magnons, Bogoliubov-Anderson mode
- A weak **aligning field** establishes macroscopic order and prevents decomposition in domains: magnetic field (ferromagnet), electric field (liquid crystal)

Different systems with broken symmetry

phenomenon	disordered phase	ordered phase	order parameter	collective excitations	rigidity phenomenon
crystallization	liquid/ gaseous	solid	(FT of charge density)	phonons	rigidity
ferro-magnetism	paramagnet	ferromagnet	$\mathbf{M}$	magnons	permanent magnetism
antiferro-magnetism	paramagnet	antiferro-magnet	staggered $\mathbf{M}$	AF magnons	…
nematic liquid crystal	liquid of anisotropic molecules	nematic liquid crystal	$S = \left\langle \frac{1}{2}\left(3\cos^2\theta - 1\right)\right\rangle$	director fluctuations	…
superconduc-tivity	(normal) metal	superconduc-tor	$\lvert\psi\rvert \exp(i\phi)$	Bogoliubov-Anderson mode	condensate stiffness

after Blundell

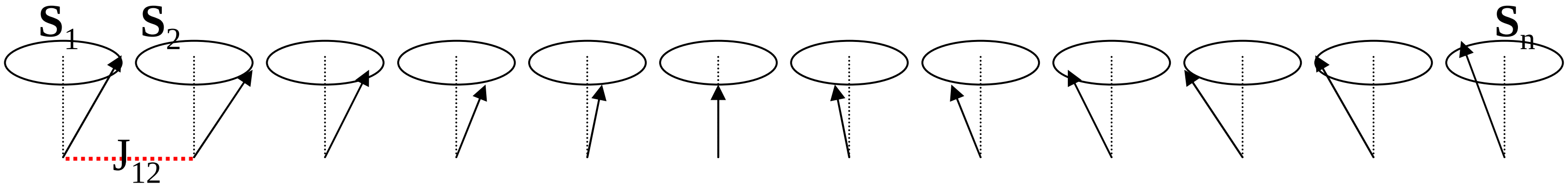
The Mermin-Wagner-Berezinskii theorem

Spontaneous symmetry breaking is a correlation effect. Are correlations always strong enough to induce symmetry breaking?

NO! It depends on the particle coordination and thus the **dimensionality of the system**:

There is no long-range order (LRO) above T = 0 in 1D and 2D for systems with continuous symmetry and short-range correlations.

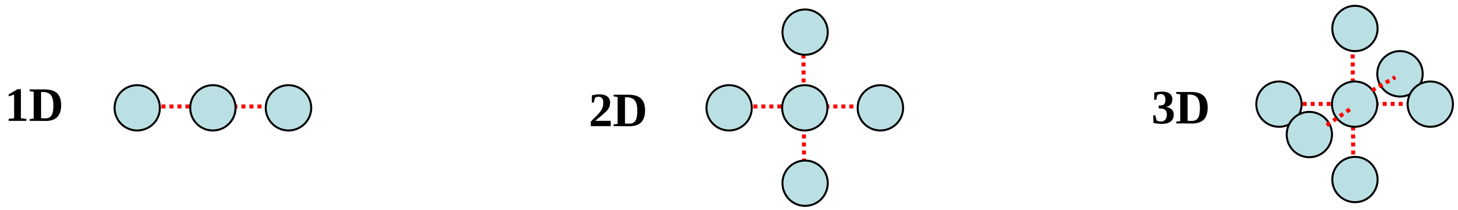
Heisenberg-ferromagnetism, -antiferromagnetism, crystallization



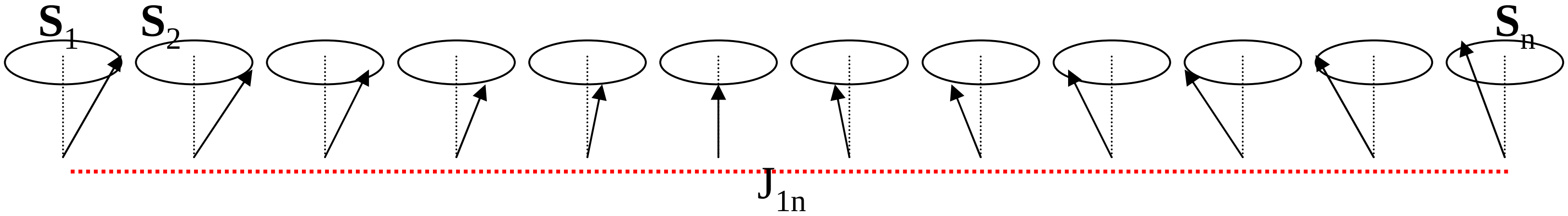
$\mathbf{S}_1$ nearly parallel to $\mathbf{S}_2$ $\Rightarrow$ energy penalty negligible. Long-λ (low-**k**) excitations are present and destroy LRO.

Conditions for the validity of the MWB-theorem

In higher dimensions, LRO is stabilized by the high coordination number of the particles ⇒ increasing mean-field like behavior



For long-range interactions even long-λ excitations cost a finite energy

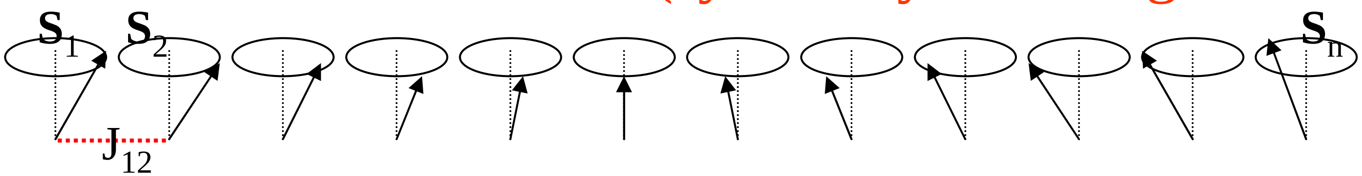


For anisotropic interactions or discrete symmetries excitations cost a finite energy

Long-range order below $T_c > 0$ in 2D Ising model

Goldstone modes

In systems with continuous broken symmetry and short-range interactions there is a gapless spectrum of collective excitations called **Goldstone modes** (symmetry-restoring modes).



Energy necessary for long-λ excitations is negligible.

FM- and AF-magnons, phonons

Range of validity:

For long-range interactions even long-λ excitations cost a finite energy

Plasmons and plasmon-like excitations in charged fluids

Anisotropic interactions or discrete symmetries

Anisotropy gaps in Heisenberg-magnets. Only defects (fully flipped spins) possible in Ising model.

2. Spin waves in the Heisenberg model

a. Describing correlated electron systems – from the Hubbard to the Heisenberg model

b. Long-range FM order, magnons

c. AF order and quantum fluctuations, AF-magnons

P. Fazekas, *Lecture notes on electron correlation and magnetism*, World Scientific

S. W. Lovesey, *Theory of neutron scattering from condensed matter*, Oxford UP, vol. 2

D. Khomskii, *Lecture Notes on advanced solid state physics*

J. M. Tranquada, *Neutron scattering studies of antiferromagnetic correlations in cuprates*, preprint available on `arXiv:cond-mat/0512115`

From the Hubbard to the Heisenberg model

$$H = -t \sum_{\langle i,j \rangle,\sigma} (c^+_{i\sigma} c_{j\sigma} + \text{herm. conj.}) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

Hubbard model

nearest neighbors → $\langle i,j \rangle,\sigma$; hopping term; interaction term

$c^+_{i\sigma}$ creates e[-] in Wannier state $\phi(\mathbf{r} - \mathbf{R}_i)$, $n_{i\sigma} = c^+_{i\sigma} c_{i\sigma}$ is the occupation number operator.

The Hubbard-*U* describes the on-site Coulomb-repulsion.

Possible states for each lattice site *i*: $|\uparrow\downarrow\rangle, |\uparrow\rangle, |\downarrow\rangle, |0\rangle$ (here spin 1/2)

$$H = -t \sum_{\langle i,j \rangle,\sigma} (\tilde{c}^+_{i\sigma} \tilde{c}_{j\sigma} + \text{herm.conj.}) + J \sum_{\langle i,j \rangle} \left(\mathbf{S}_i \mathbf{S}_j - \frac{n_i n_j}{4} \right)$$

t-*J* model

Nearest-neighbor spins interact (antiferromagnetically) via $J=4t^2/U$.

$\tilde{c}^+_{i\sigma} = (1 - n_{i,-\sigma}) c^+_{i\sigma}$ prohibits double occupancy – possible states: $|\uparrow\rangle, |\downarrow\rangle, |0\rangle$

When all lattice sites are exactly singly occupied:

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

Heisenberg model

The Heisenberg model – a low-energy effective *H*

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

Heisenberg model

$J < 0$: FM interaction; coupling to orbitals (Goodenough-Kanamouri rules), double-exchange.
$\Rightarrow$often obtained as **effective Hamiltonian**.
EuO, $La_{0.7}Pb_{0.3}MnO_3$, $YTiO_3$

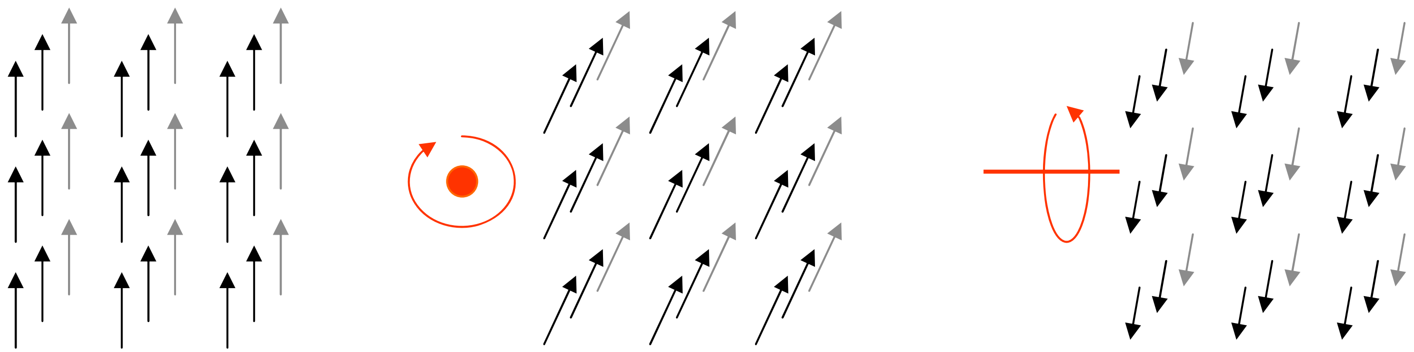
$J > 0$: AF interaction; e.g. direct exchange or super-exchange. Quantum fluctuations crucial.
MnF_2, La_2CuO_4

The Heisenberg ferromagnet, $J < 0$

Parallel spin-orientation is energetically favorable.

Ground state: $\left|\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\ldots\right\rangle_\sigma$ where σ can be in any direction:

Here the symmetry of H shows up.



$\Rightarrow$ $(2NS + 1)$-fold degeneracy (N number of sites).

Specific for the FM case: $\left|\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\ldots\right\rangle_\sigma$ is an eigenstate of H.

Quantum fluctuations not crucial.

FM magnons

Due to the degeneracy we are free to pick the quantization axis || z

$$H = J\sum_{\langle i,j\rangle}\mathbf{S}_i\mathbf{S}_j = \underbrace{J\sum_{\langle i,j\rangle}S_i^z S_j^z}_{\text{Ising term}} + \underbrace{\frac{J}{2}\sum_{\langle i,j\rangle}\left(S_i^+ S_j^- + S_i^- S_j^+\right)}_{\text{Spin-flip terms, reason for spin waves:}}$$

$$S_i^+ S_j^- \left|\uparrow\uparrow\uparrow\cdots\underset{i}{\downarrow}\cdots\underset{j}{\uparrow}\cdots\right\rangle = \left|\uparrow\uparrow\uparrow\cdots\underset{i}{\uparrow}\cdots\underset{j}{\downarrow}\cdots\right\rangle \text{ for } S = 1/2$$

Magnons are (approximately) bosonic quasiparticles involving **all spins**. For convenience, work with corresponding bosonic operators:

$$[a_i, a_j^+] = \delta_{ij},\ [a_i, a_j] = [a_i^+, a_j^+] = 0$$

Holstein-Primakoff transformation

$$S_j^+ = \sqrt{2S}\left(1 - \frac{a_i^+ a_i}{2S}\right)^{1/2} a_i, \; S_j^- = \sqrt{2S}\, a_i^+ \left(1 - \frac{a_i^+ a_i}{2S}\right)^{1/2}$$

In order to describe propagating waves, introduce

$$b_k^+, b_k \overset{FT}{\longleftrightarrow} a_i^+ a_i$$

$$b_{\mathbf{k}}^+ = \frac{1}{\sqrt{N}} \sum_{\mathbf{j}} e^{i\mathbf{kj}} a_{\mathbf{j}}^+ \text{ etc.}$$

$\Rightarrow$ a magnon is a “delocalized flipped spin”.

Each magnon reduces S_{tot} by 1, tending to restore symmetry

S_{tot} finite $\Rightarrow$ magnons are only approximately bosons.

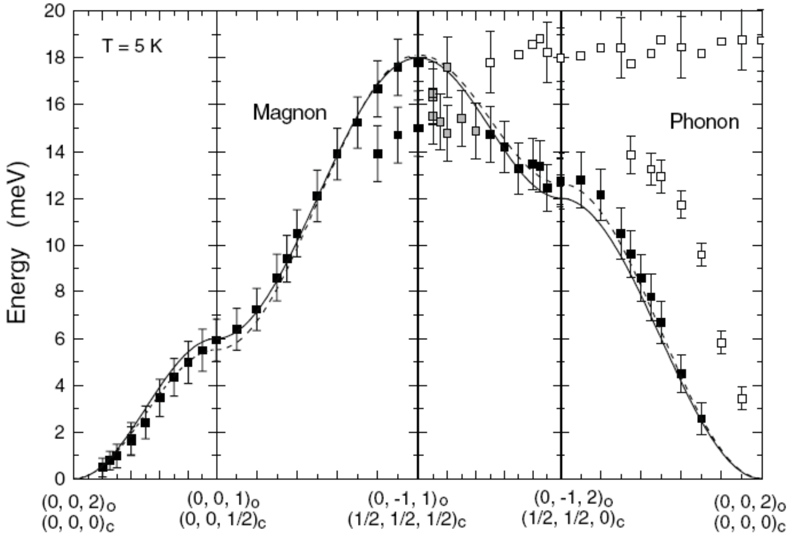
But concept is valid for few excited magnons far apart

Linearization and dispersion relation

$\Rightarrow H = \text{const.} + H_0 + H_1$ H_0 bilinear in b_k, H_1 higher order.

$$H_0 = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{+} b_{\mathbf{k}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \hat{n}_{\mathbf{k}}$$

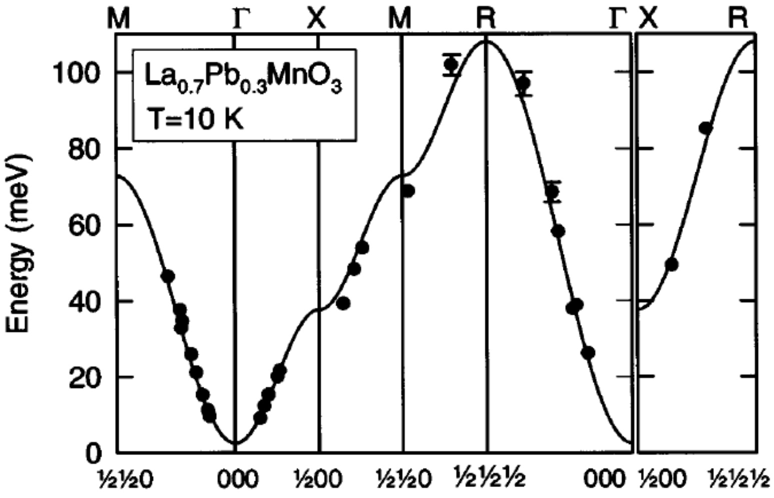
with $\omega_{\mathbf{k}} \approx S|J|(kd)^2$ in the long-wavelength limit (d lattice constant).



Magnon dispersion in $YTiO_3$:
A (nearly) gapless, isotropic FM-Heisenberg spectrum with weak AF admixtures
From C. Ulrich *et al.*, *PRL* **89**, 167202.
see also G. Khaliullin *at al.*, *PRL* **89**, 167201

ferromagnetism is stabilized by FM superexchange due to coupling to orbital degrees of freedom

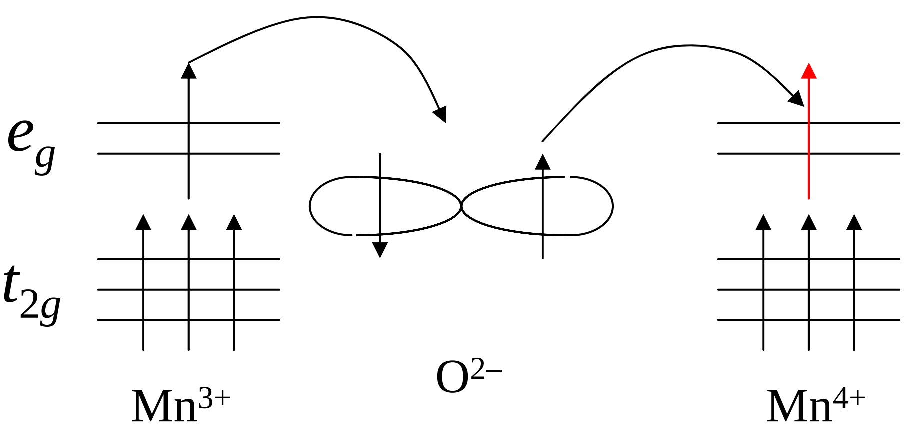
FM magnons, further example



wave-vector

Magnon dispersion in $La_{0.7}Pb_{0.3}MnO_3$ (giant magneto-resistance compound) along different high-symmetry directions

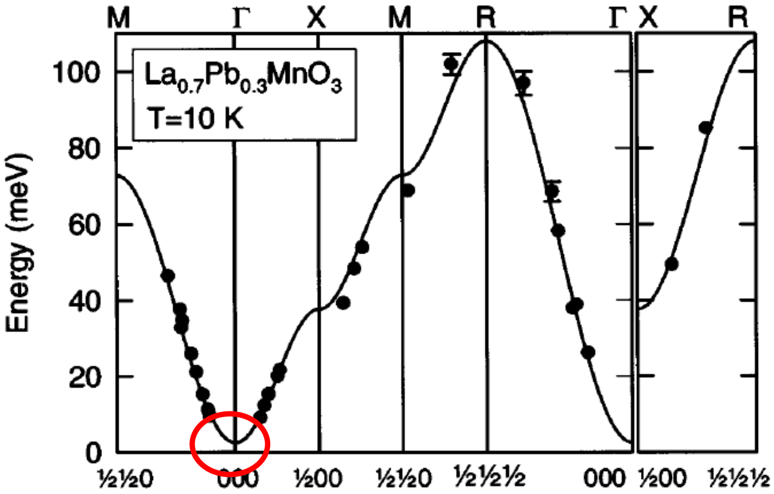
From Perring *et al.*, *PRL* **77**, 711.



Hunds rule favors FM alignment of the Mn-ions

Deviation from ideal behavior

Easy-axis anisotropy ($A > 1$):



$$H = J \sum_{\langle i,j \rangle} \left(A S_i^z S_j^z + S_i^x S_j^x + S_i^y S_j^y \right)$$

Ground-state only 2-fold degenerate:

$$\left| \uparrow\uparrow\uparrow\uparrow\uparrow\uparrow \ldots \right\rangle_z \quad \text{and} \quad \left| \downarrow\downarrow\downarrow\downarrow\downarrow\downarrow \ldots \right\rangle_z$$

Deviations from *z*-direction cost energy ⇒ gap

Long-range dipole-dipole interactions lead to a gap and sample-geometry dependence due to demagnetization effects

Non-linear effects due to H_1 introduce magnon-magnon interactions: decay due to collision and two-magnon bound states (esp. in low-D).

The Heisenberg antiferromagnet, J > 0

Energy of a bond is not minimized by $|\uparrow\downarrow\rangle$ but by a singlet $\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$

Bond energies cannot be simultaneously minimized.

Néel state $|\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\ldots\rangle$ is not a strict eigenstate.

Quantum fluctuations (excitations) of the order ~ *1/zS* are present even at $T = 0$ (z coordination number)

$\Rightarrow$ Quantum effects strongest for low S and low dimensions.

For 1D there is no long-range order even at $T = 0$. Excitation spectrum is profoundly different: spin-1/2 spinons.

For 2D and 3D bosonic spin-1 magnons are found.

AF magnons

Assume that the classical Néel state is the ground state.
Similar approach as for FM, but on the sublattices *A* and *B*
($S_{\mathbf{j}}^{z}$ = S for $\mathbf{j} \in A$, –S for $\mathbf{j} \in B$)

Holstein-Primakoff transformation and linearization lead to

$$H_0 = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(\alpha_{\mathbf{k}}^{+} \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^{+} \beta_{\mathbf{k}} \right)$$

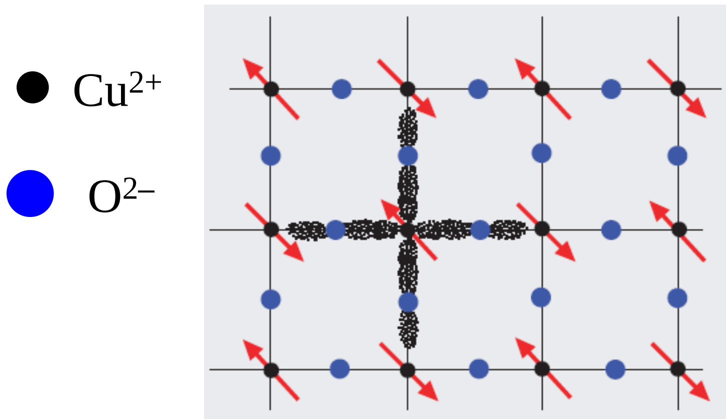
with linear dispersion for small *k*:

$$\omega_{\mathbf{k}} \approx JzSd\left|k\right|$$

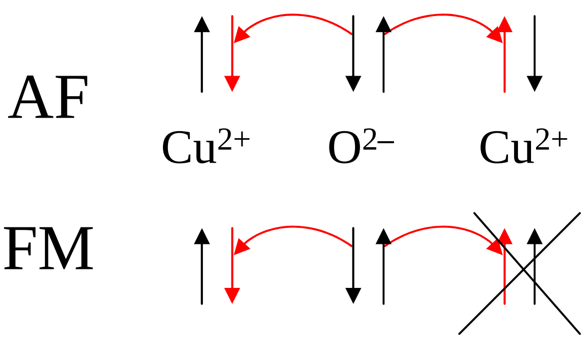
The spectrum is gapless and two-fold degenerate.

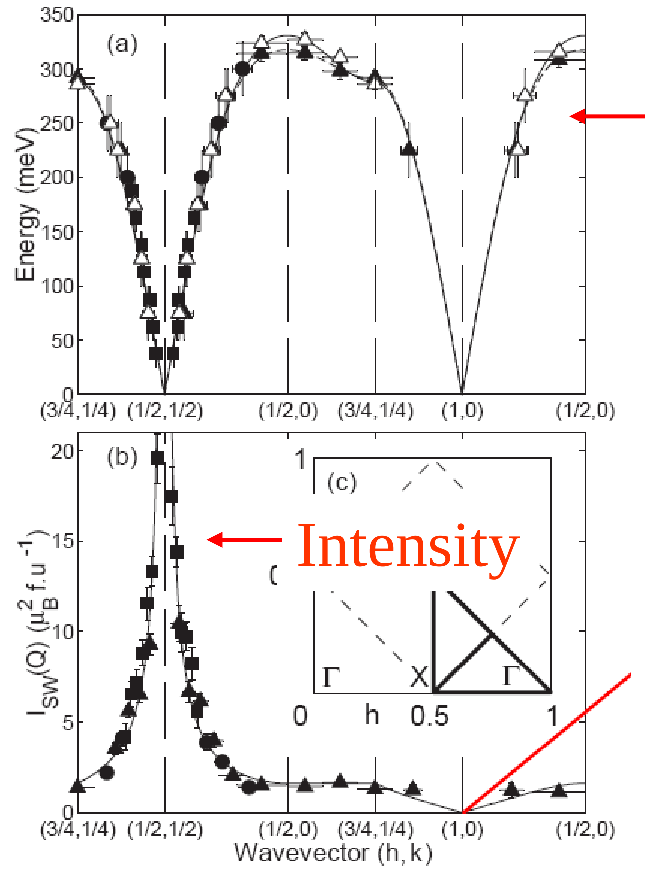
The sublattice magnetization is reduced from its classical value *NS* by $N\Delta S$ due to quantum fluctuations.
Nominally, $\Delta S \to \infty$ for 1D (no LRO)
For D=2, $\Delta S \approx 0.2$, i.e. a 40% effect for S = 1/2 (quasi-2D cuprates)

Magnons in La_2CuO_4 – in-plane dispersion



Superexchange leads to AF arrangement:



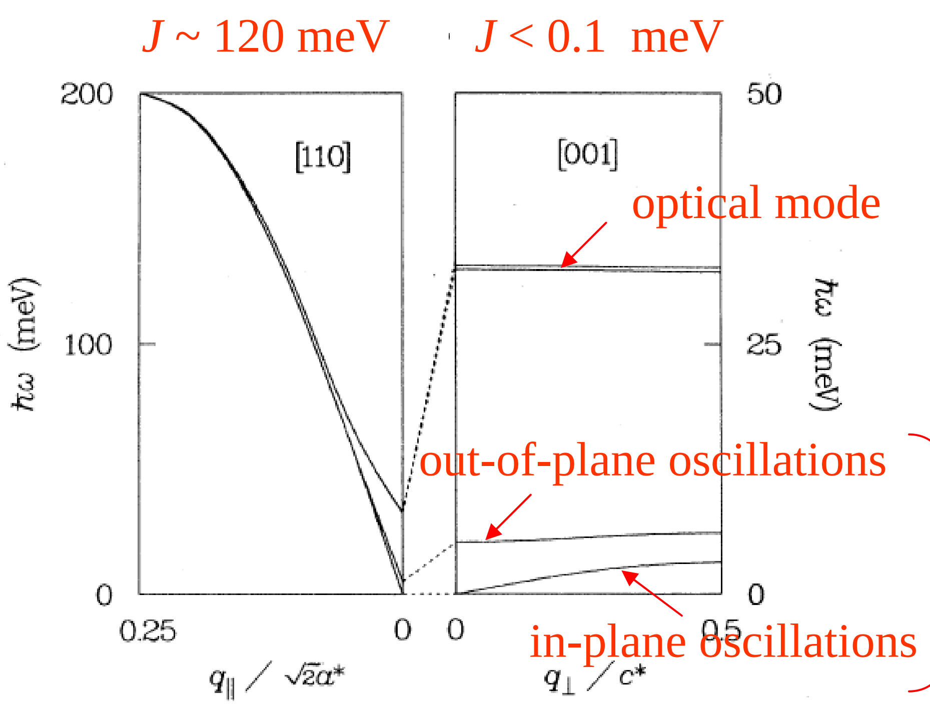


Coldea et al., *PRL* **86**, 5377.

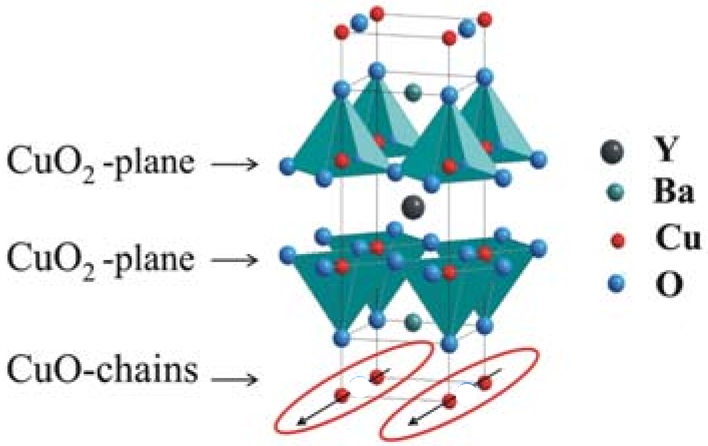
Magnetic unit cell volume is twice the nuclear unit cell volume
$\Rightarrow$ additional Bragg peaks at (1/2,1/2). Structure factor suppresses intensity around (1,0).

$YBa_2Cu_3O_6$ – optical excitations, anisotropy gaps

Two CuO_2 planes basis
⇒ Additional "optical" mode in analogy to phonons



J. M. Tranquada *et al.*, *PRB* **40**, 4503



$$H_0 = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(\alpha_{\mathbf{k}}^+ \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^+ \beta_{\mathbf{k}} \right)$$

The two-fold degeneracy can be lifted by anisotropy, e.g. in the quasi-2D antiferromagnet

3. Excitations and reduced dimensionality

a. One-dimensional magnetism, spin chains

b. Spin ladders

c. Two-dimensional magnetism, cuprates

Stephen Blundell, Magnetism in condensed matter, Oxford master series

P. Fazekas, *Lecture notes on electron correlation and magnetism*, World Scientific

J. M. Tranquada, *Neutron scattering studies of antiferromagnetic correlations in cuprates*, preprint available on `arXiv:cond-mat/0512115`

D. C. Mattis, *The theory of magnetism made simple*, World Scientific

Spin-1/2 Heisenberg chains

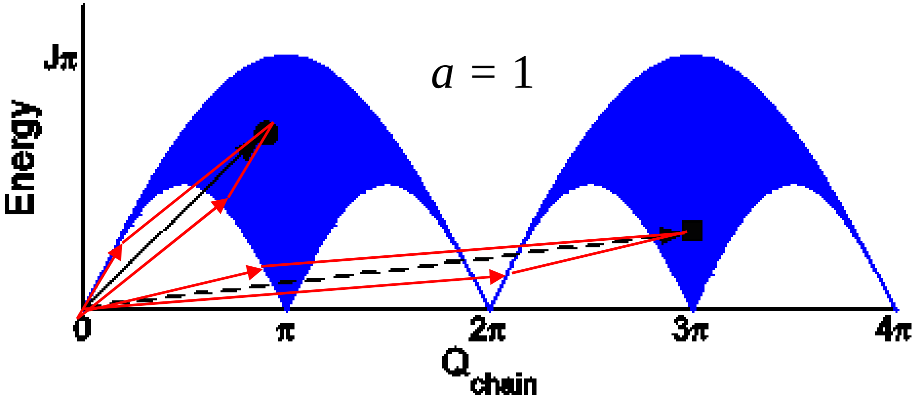
Due to quantum fluctuations, there is no LRO in AF Heisenberg chains. Excitations are spin 1/2 fermions: gapless spinons

$$\hbar\omega = \pi \left| J \sin(qa) \right|$$

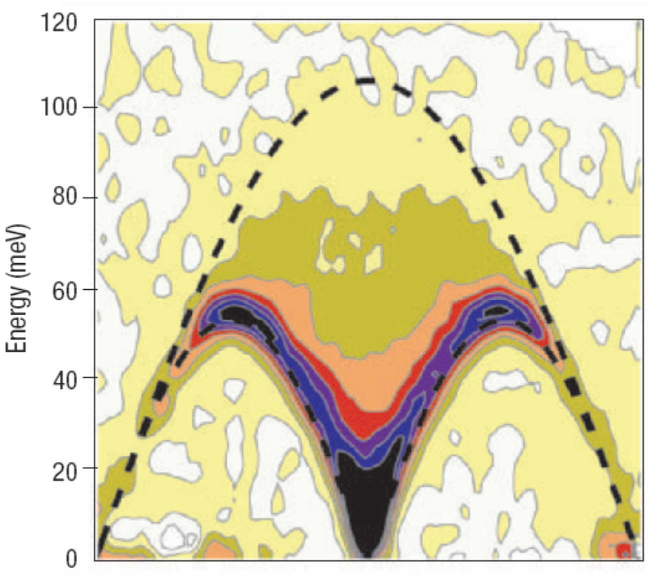
Neutron scattering changes spin by 1:

Two spinons are created/annihilated

$\Rightarrow$ continuum is obtained



after http://www.hmi.de/bereiche/SF/SFN1/themes/quantum_magnetism/index_en.html



Spinon-continuum in $KCuF_3$. Weak FM coupling between the CuF chains stabilizes LRO below 39 K. Excitations have mixed character.

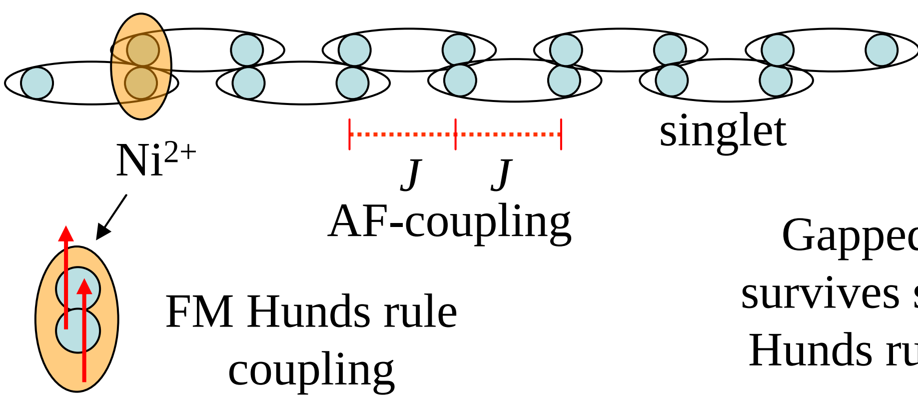
B. Lake *et al.*, *Nat. Mat.* 4, 329 (2005)

Spin-1: Haldane chains

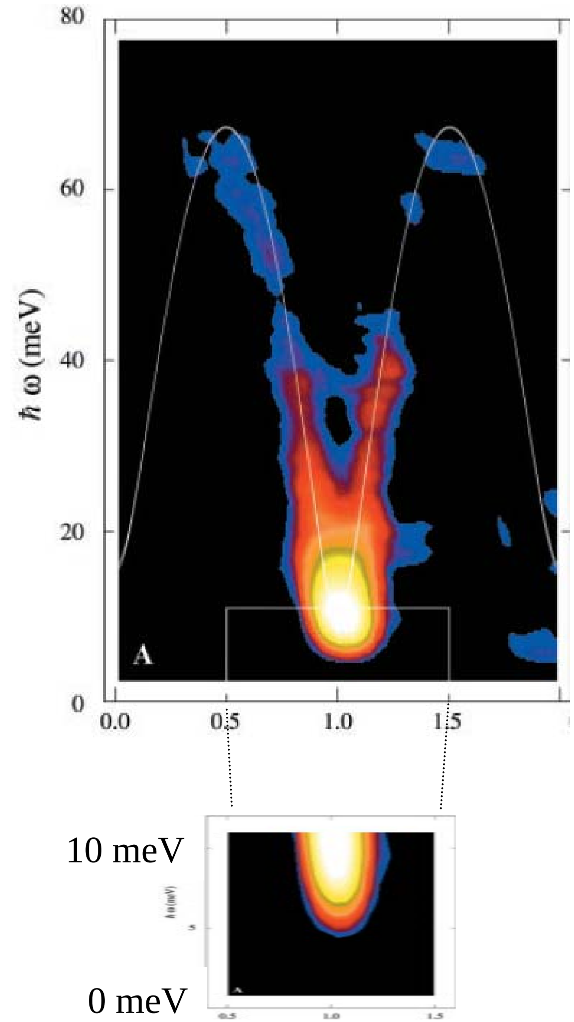
Integer-spin chains are fundamentally different, e.g. $YBaNiO_5$

Two holes on each Ni^{2+}. First neglect Hunds rule coupling.

The spin-1/2 holes can form a macroscopic singlet: valence bond solid (VBS). Spectrum consists of gapped singlet-triplet excitations.



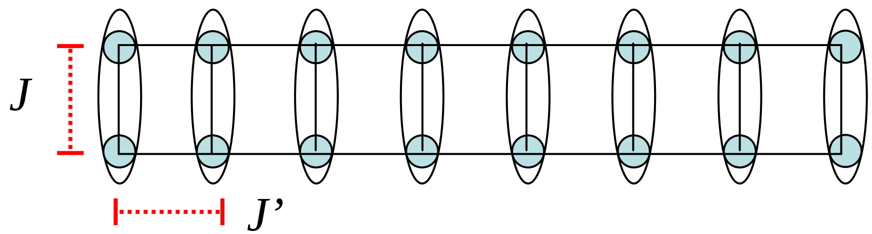
Gapped character survives switching on Hunds rule coupling.



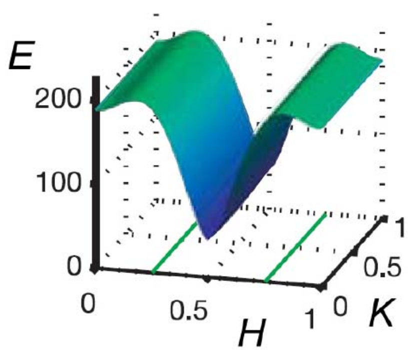
From G. Xu *et al.*, *Science* **289**, 419

Spin-1/2 ladders

The two-leg spin-1/2 ladder has a gapped excitation spectrum – easy to see in "strong rung" limit $J \gg J'$

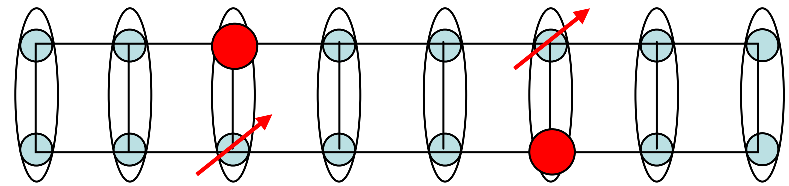


Ground state consists of singlets. Singlet-triplet excitations are gapped and disperse along the legs

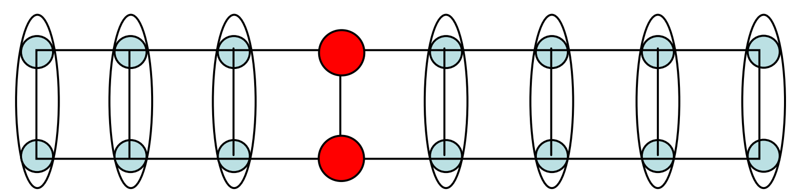


From J. M. Tranquada *et al.*, *Nature* **429**, 534

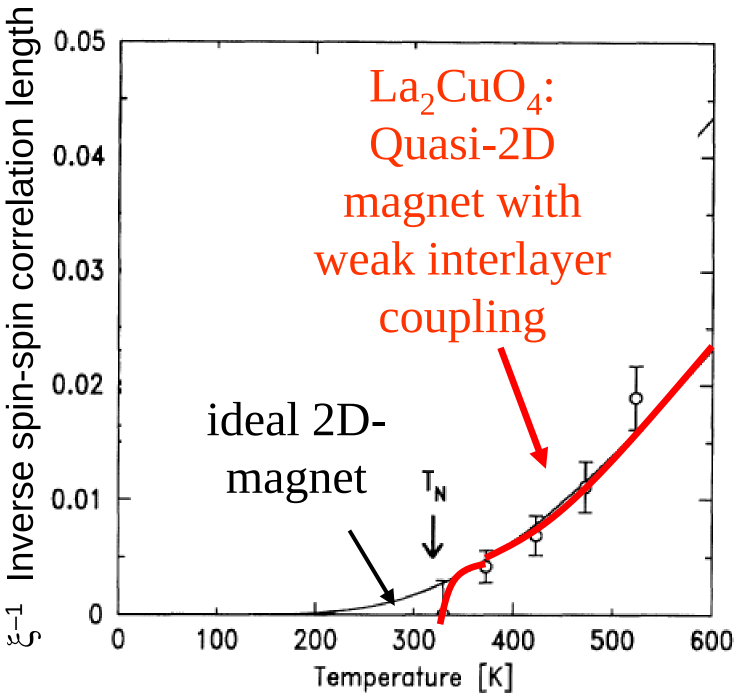
Hole doping breaks singlets



Pairing-up reduces number of "damaged" bonds – way towards superconductivity?



Quasi-2D Heisenberg magnets



After B. Keimer *et al.*, *PRL* **67**, 1930

There is no LRO above T = 0 for ideal 2D magnets (Mermin-Wagner theorem) but spin-spin correlation length diverges

$$\xi^{-1} \xrightarrow{T\to 0} \infty$$

In the presence of weak interlayer coupling, 3D LRO can be established below $T_N > 0$

$$\xi^{-1} \xrightarrow{T\to T_N} \infty$$

4. Magnetic excitations in hole doped cuprates

a. Stripe order in the CuO_2 planes

b. electronic liquid crystal phases

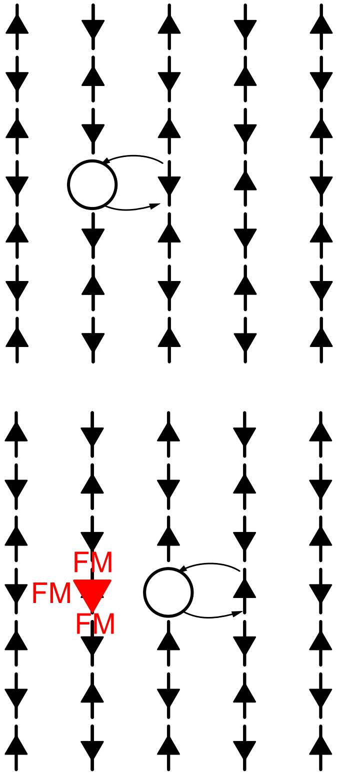
c. The hour-glass dispersion in the cuprates

J. M. Tranquada, *Neutron scattering studies of antiferromagnetic correlations in cuprates*, preprint available on `arXiv:cond-mat/0512115`

S. A. Kivelson *et al.*, *Nature* 393, 550 (1998) and *Rev. Mod. Phys.* 75, 1201 (2003)

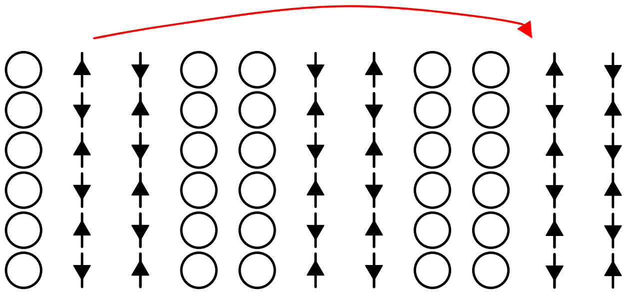
Stripe order

A single itinerant hole in the AF matrix frustrates the order

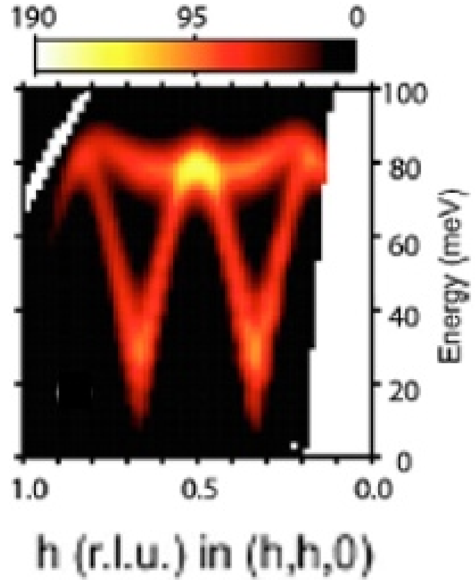


In a stripe-phase the holes segregate. Magnetic unit cell larger: "Electronic superlattice"

C$_4$-symmetry and translational symmetry broken, but arrangement commensurate with the lattice.



Spinwaves in $La_{2-1/3}Sr_{1/3}NiO_4$ with stripe order

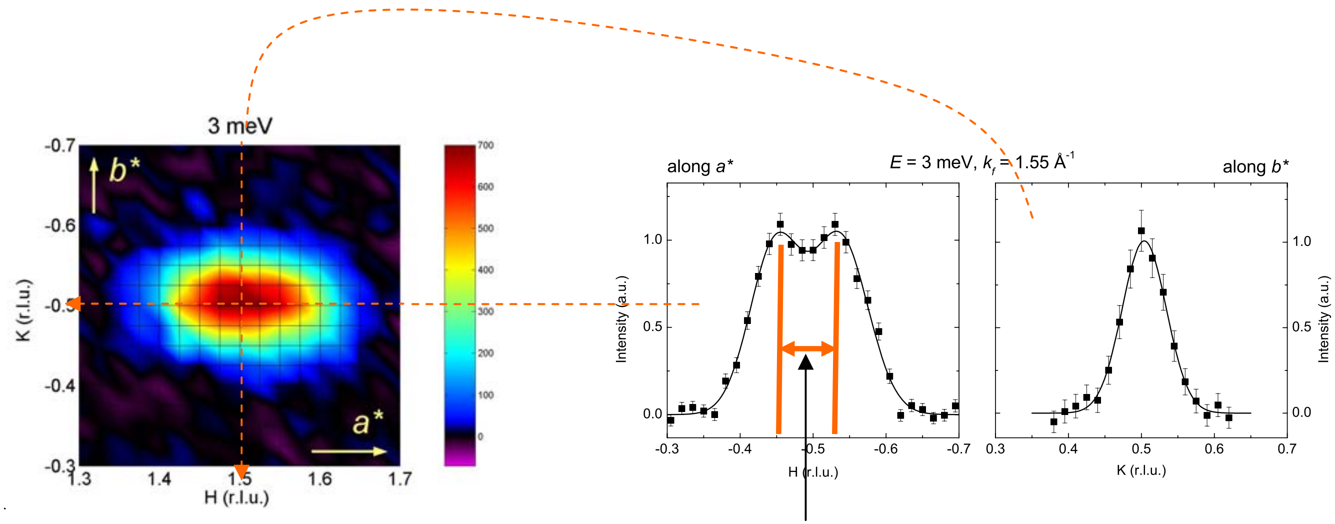


Woo et al., *PRB* 72, 064437

Other symmetry broken states

Are there states, where only the C_4-symmetry is broken?

Such states will probably show incommensurate magnetism

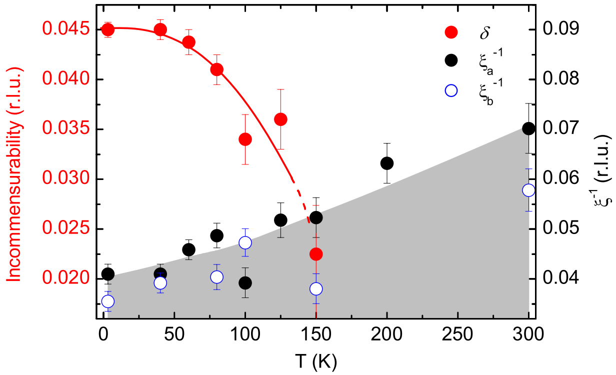


δ, incommensurability

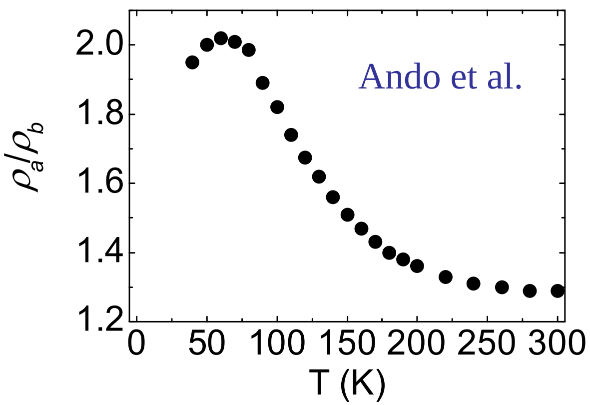
V. Hinkov *et al.*, *Science*, 10.1126/science.1152309

Possible scenarios: (i) Inhomogeneous, “stripy” arrangements with incommensurate spin modulation. (ii) Spin-spiral arrangements

Electronic liquid crystal state



3 meV



V. Hinkov *et al.*, *Science*,
10.1126/science.1152309

Spontaneously broken C_4-symmetry and onset of incommensurability around 150 K

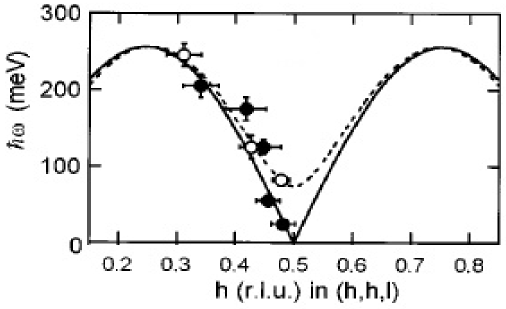
Measurements show: No static moment, no long-range order

Such electronic states of matter are called electronic liquid crystals
cf. Kivelson et al., *Nature* 393, 550 (1998)

In-plane lattice distortion ($a \neq b$) serves as a weak aligning field

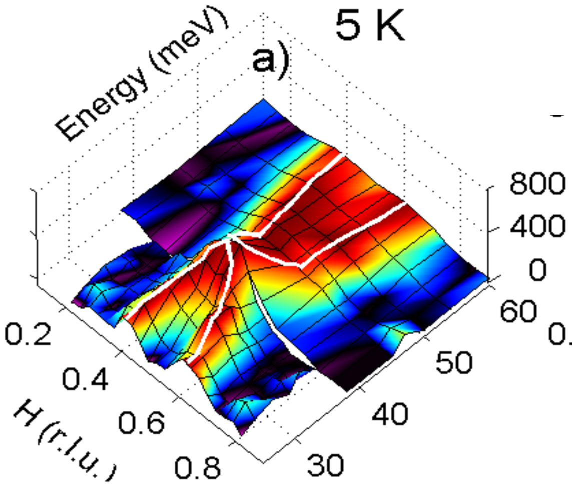
Spontaneous increase of the resistivity anisotropy ρ_a/ρ_b suggests that charge and spin dynamics are coupled

The hour-glass dispersion in hole-doped cuprates



AF Spin waves in undoped $YBa_2Cu_3O_6$

Hayden et al., *PRB* **54**, R6905

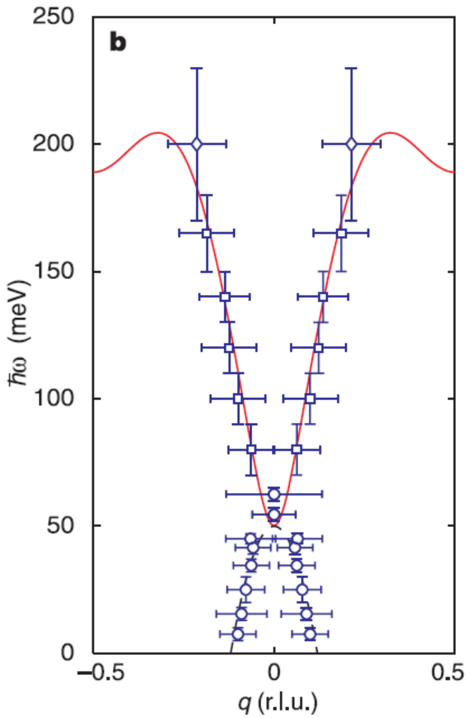


Spin excitations in $YBa_2Cu_3O_{6.6}$

Hinkov et al., *Nature Physics* **3**, 780 (2007)

See also

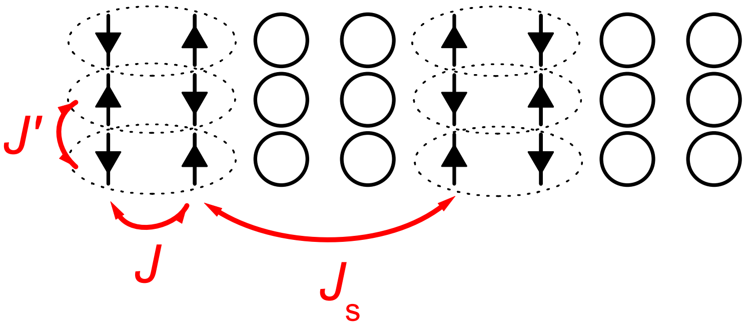
Hayden et al., *Nature* **429**, 531



Spin excitations in $La_{15/8}Ba_{1/8}CuO_4$

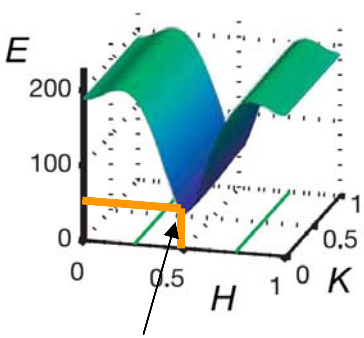
Tranquada et al., *Nature* **429**, 534

Stripes, ladders and the hour-glass dispersion



J_s intermediate:
upward- and downward-dispersion, saddlepoint at the hour-glass neck (however deviations form expt., issue not fully settled).

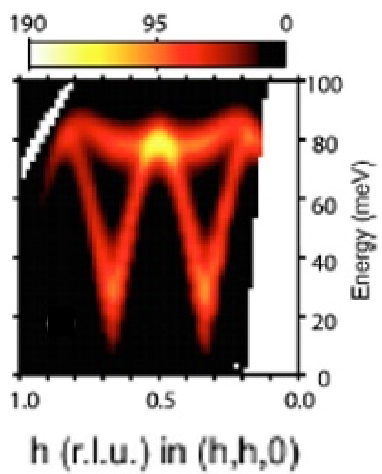
J_s negligible:
spin ladder



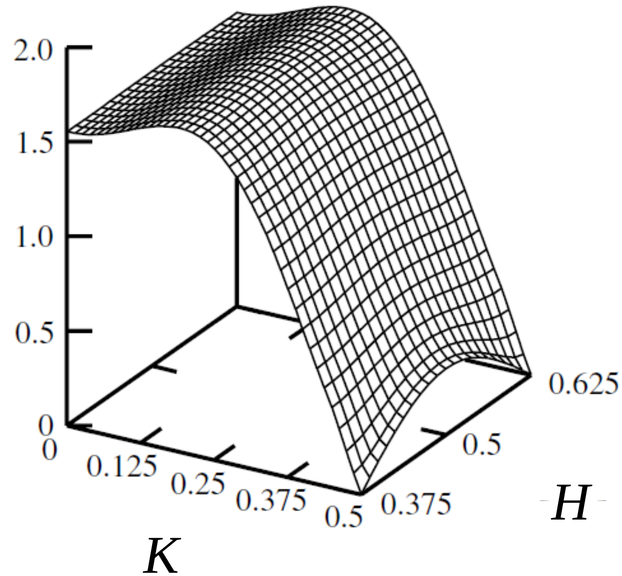
Triplet gap

From J. M. Tranquada *et al.*, *Nature* **429**, 534

J_s large:
stripe magnons



Woo et al., *PRB* 72, 064437



Uhrig et al., *PRL* **93**,267003,

see also Vojta et al., *PRL* **93**, 127002,