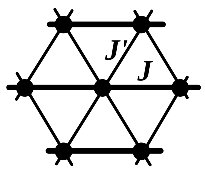
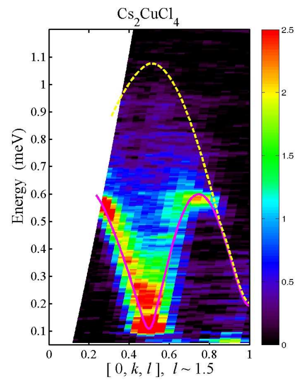
Neutron scattering from spinons in the anisotropic triangular quantum magnet Cs_2CuCl_4

Radu Coldea

University of Oxford





Collaborators:

Neutron scattering

D. Alan Tennant (St. Andrews Univ.,UK)

Alexei M. Tsvelik (Brookhaven, USA)

K. Habicht, P. Smeibidl (HMI,Berlin)

Crystal growth

Z. Tylczynski (Poland)

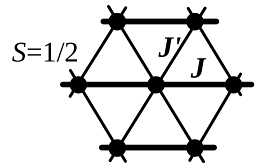
Magnetization

Yoshi Tokiwa

Philipp Gegenwart

Frank Steglich

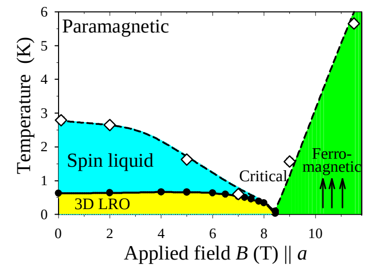
(Max Planck Institute CPfS Dresden, Germany)



Outline

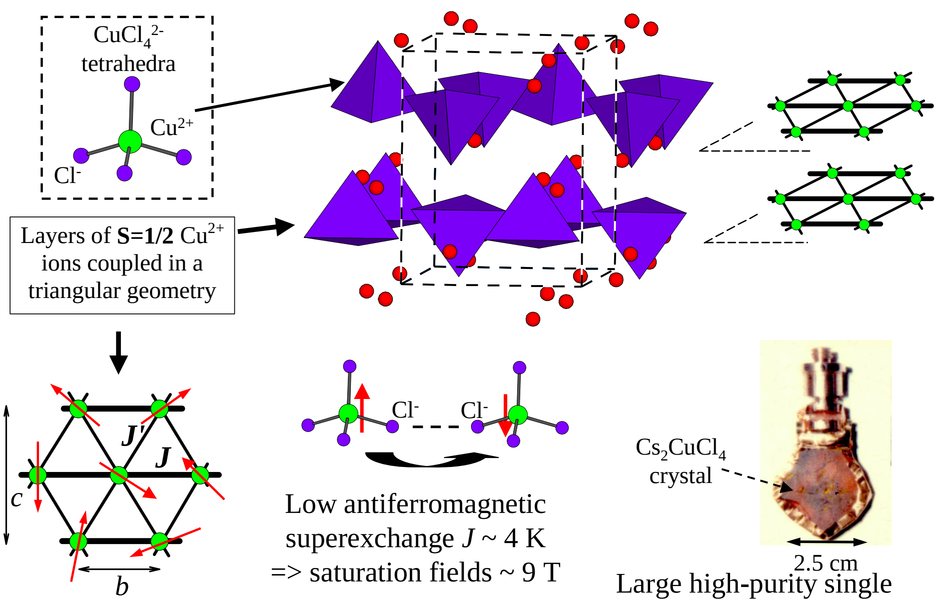
Explore 2D frustrated quantum magnet Cs_2CuCl_4 with magnetization & neutrons

1. Phase diagram
2. Measurement of Hamiltonian
3. Spin excitations
4. Summary



What are the effects of frustration and quantum fluctuations (S=1/2) in this 2D antiferromagnet ?

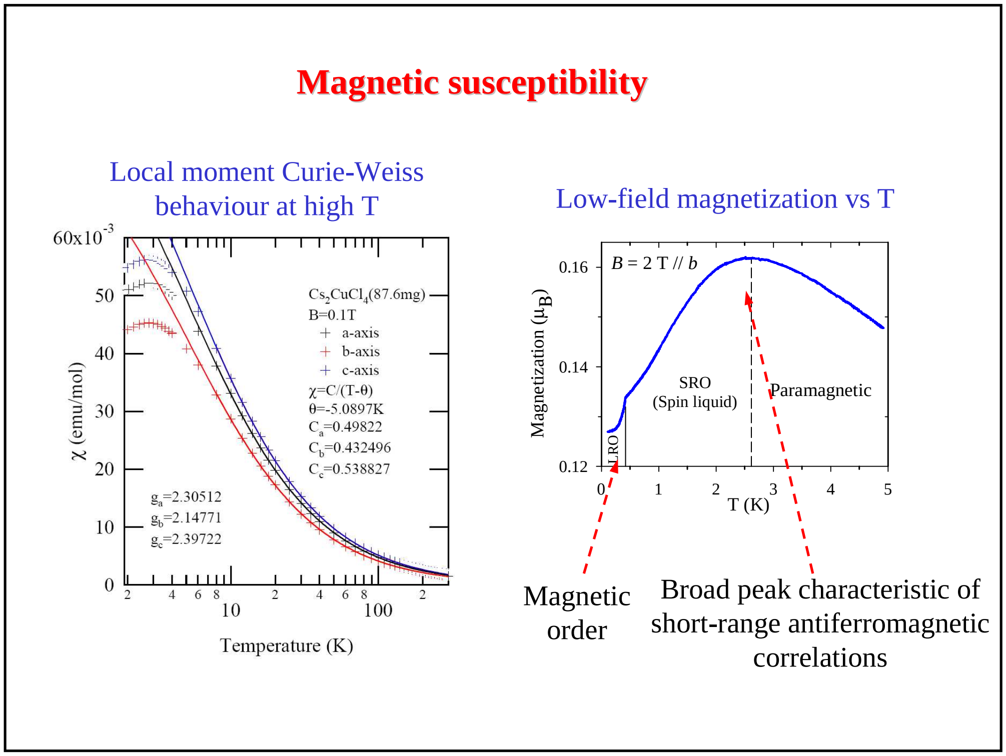
Crystal structure and magnetism of Cs_2CuCl_4

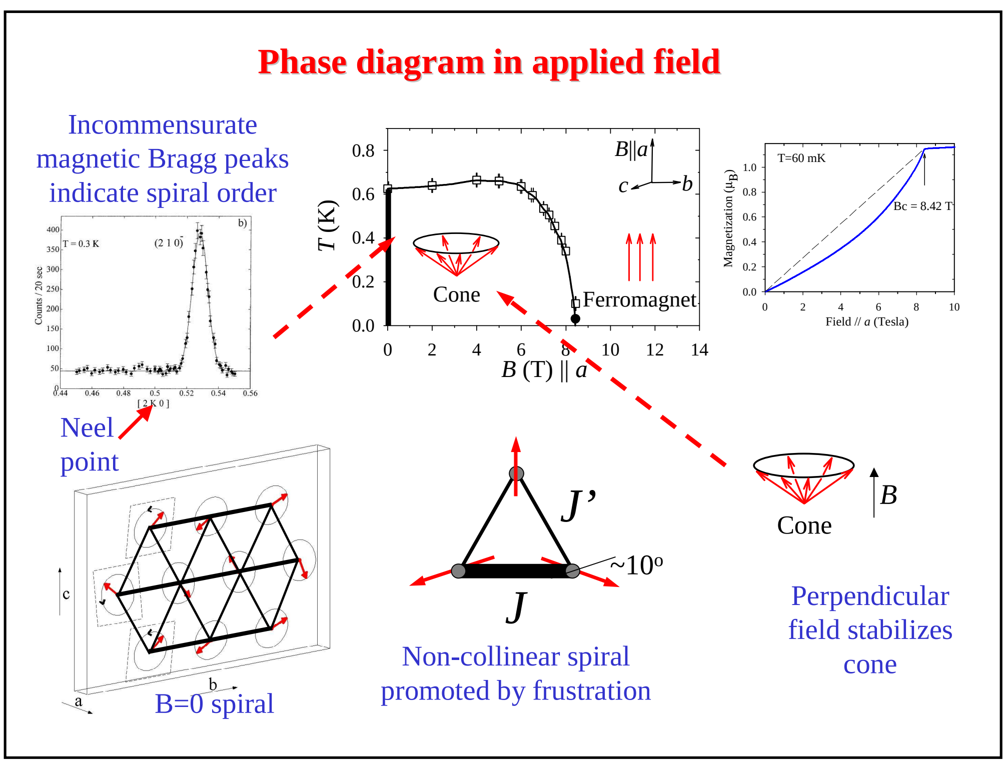


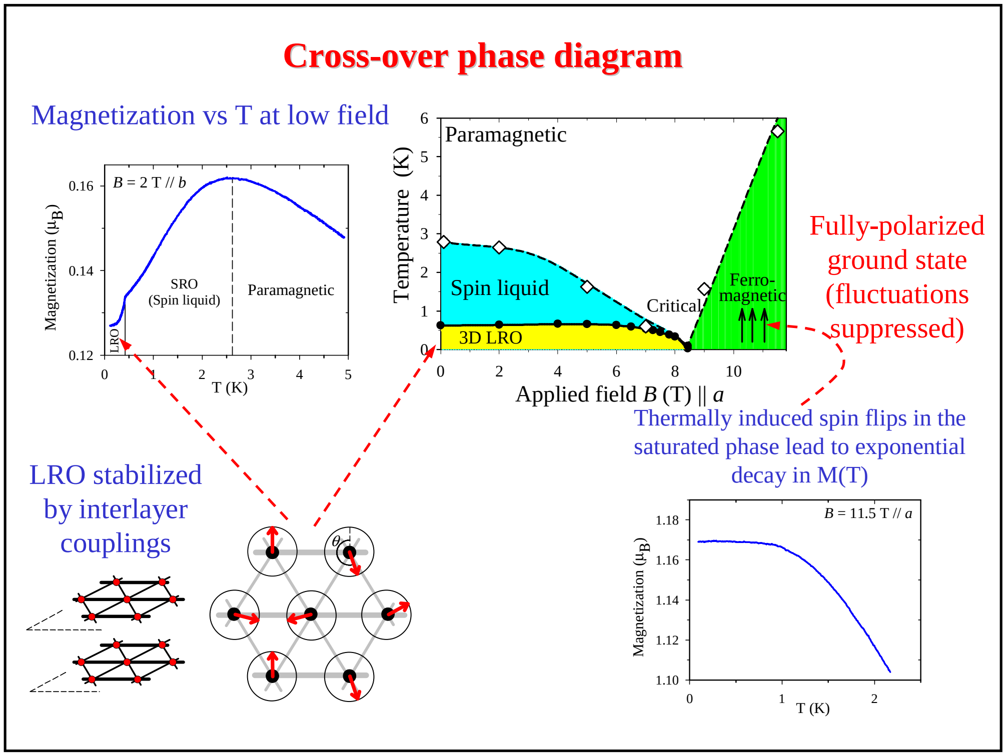
J, *J'* antiferromagnetic

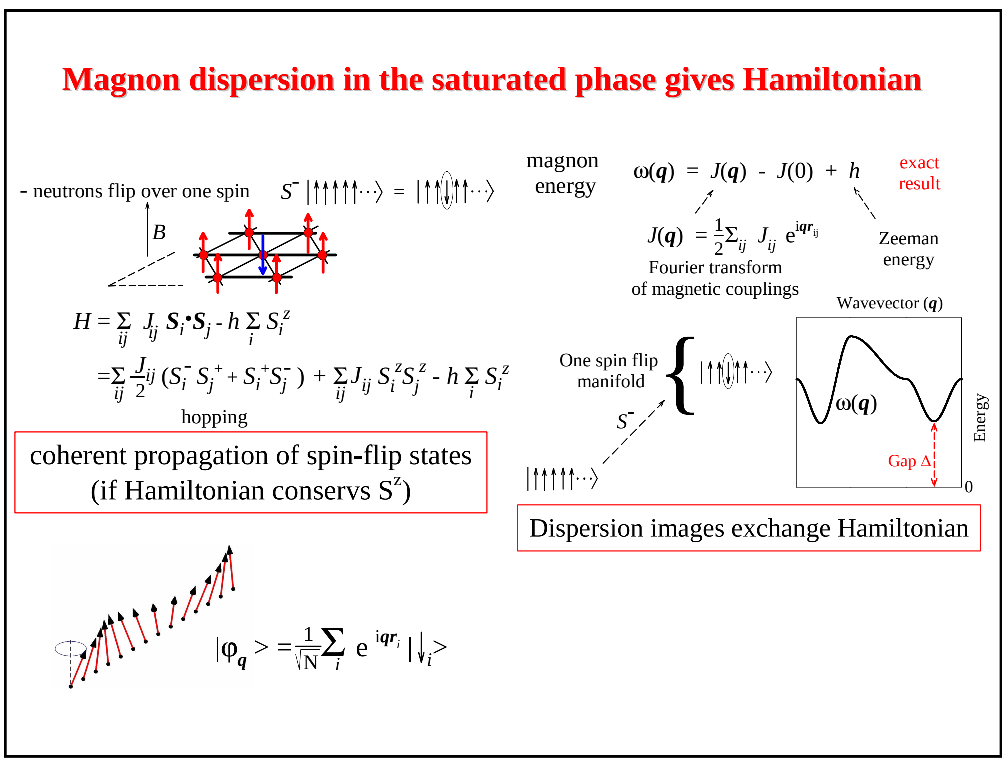
Low antiferromagnetic superexchange J ~ 4 K
=> saturation fields ~ 9 T

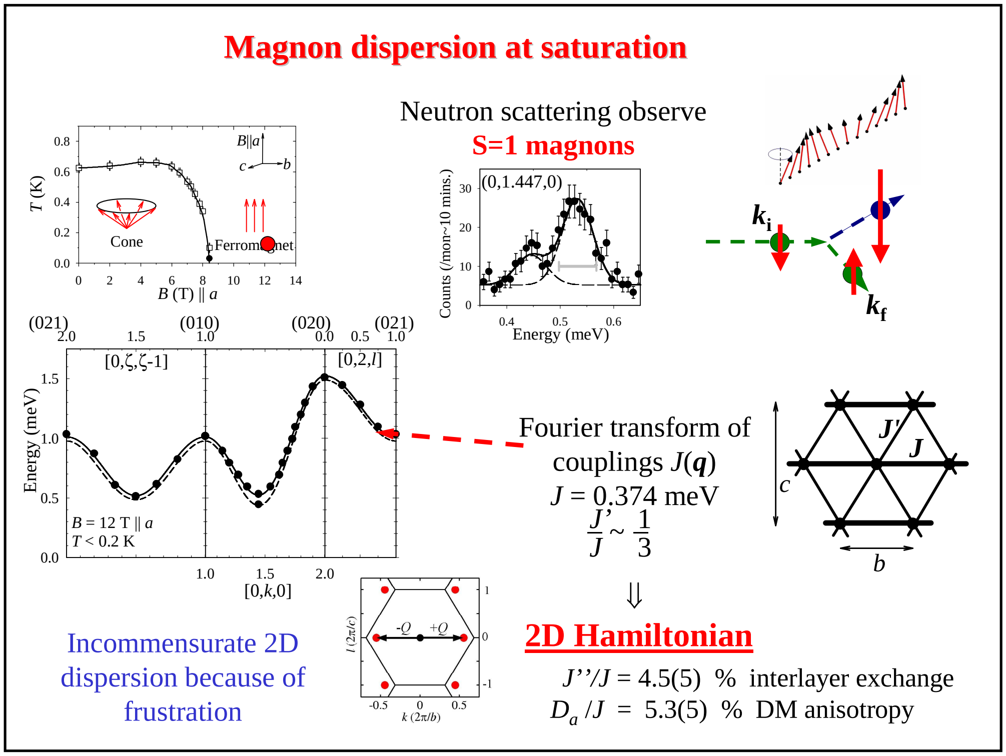
Large high-purity single crystals grow from solution


Magnetic susceptibility
Local moment Curie-Weiss behaviour at high T
Low-field magnetization vs T
60x10^-3
χ (emu/mol)
Temperature (K)
Cs2CuCl4(87.6mg)
B=0.1T
a-axis
b-axis
c-axis
χ=C/(T-θ)
θ=-5.0897K
Ca=0.49822
Cb=0.432496
Cc=0.538827
ga=2.30512
gb=2.14771
gc=2.39722
B = 2 T // b
Magnetization (μB)
T (K)
LRO
SRO
(Spin liquid)
Paramagnetic
Magnetic order
Broad peak characteristic of short-range antiferromagnetic correlations

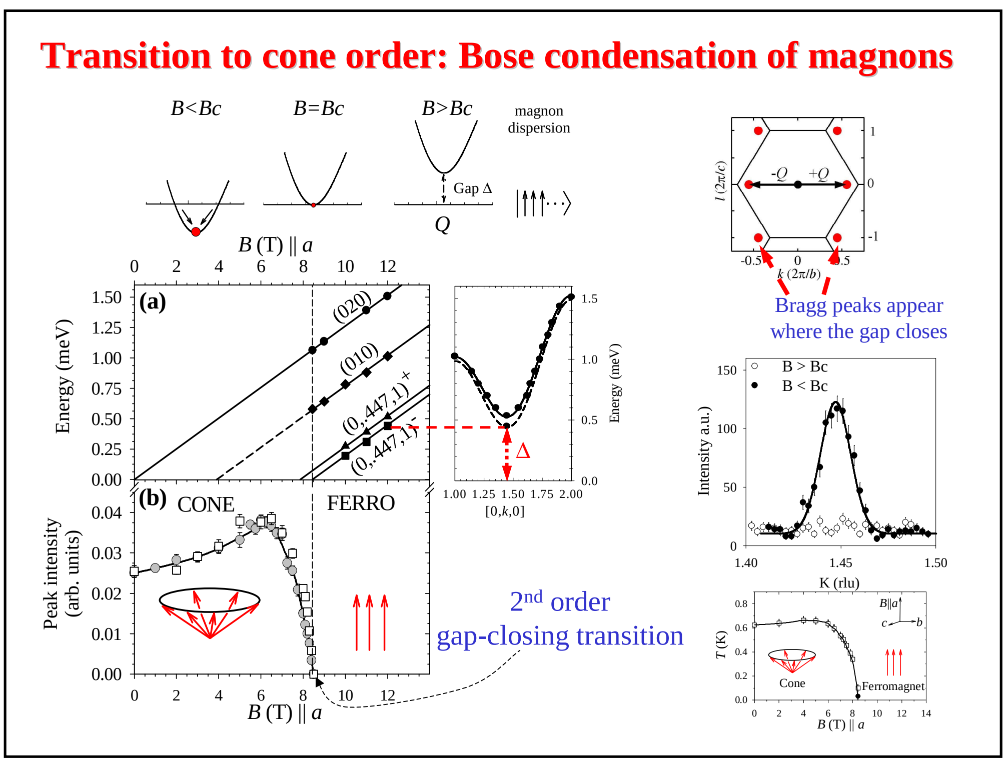

Phase diagram in applied field
Incommensurate magnetic Bragg peaks indicate spiral order
T = 0.3 K
(2 1 0)
Counts / 20 sec
[2 K 0]
Neel point
T (K)
B (T) || a
B||a
c
b
Cone
Ferromagnet
T=60 mK
Bc = 8.42 T
Magnetization (μB)
Field // a (Tesla)
J'
J
~10º
Cone
B
B=0 spiral
Non-collinear spiral promoted by frustration
Perpendicular field stabilizes cone


Cross-over phase diagram
Magnetization vs T at low field
B = 2 T // b
Magnetization (μB)
SRO (Spin liquid)
Paramagnetic
LRO
T (K)
Paramagnetic
Spin liquid
Critical
Ferro-magnetic
3D LRO
Temperature (K)
Applied field B (T) || a
Fully-polarized ground state (fluctuations suppressed)
Thermally induced spin flips in the saturated phase lead to exponential decay in M(T)
LRO stabilized by interlayer couplings
B = 11.5 T // a
Magnetization (μB)
T (K)


Magnon dispersion in the saturated phase gives Hamiltonian
- neutrons flip over one spin
$S^- |\uparrow\uparrow\uparrow\uparrow\uparrow\cdots\rangle = |\uparrow\uparrow\downarrow\uparrow\uparrow\cdots\rangle$
B
$H = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - h \sum_i S_i^z$
$= \sum_{ij} \frac{J_{ij}}{2} (S_i^- S_j^+ + S_i^+ S_j^-) + \sum_{ij} J_{ij} S_i^z S_j^z - h \sum_i S_i^z$
hopping
coherent propagation of spin-flip states (if Hamiltonian conservs S^z)
magnon energy
$\omega(\mathbf{q}) = J(\mathbf{q}) - J(0) + h$
exact result
$J(\mathbf{q}) = \frac{1}{2} \Sigma_{ij} J_{ij} e^{i\mathbf{q}\mathbf{r}_{ij}}$
Fourier transform of magnetic couplings
Zeeman energy
Wavevector (q)
One spin flip manifold
$|\uparrow\uparrow\downarrow\uparrow\uparrow\cdots\rangle$
S^-
$|\uparrow\uparrow\uparrow\uparrow\uparrow\cdots\rangle$
ω(q)
Gap Δ
Energy
0
Dispersion images exchange Hamiltonian
$|\varphi_{\mathbf{q}}\rangle = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{q}\mathbf{r}_i} |\downarrow_i\rangle$



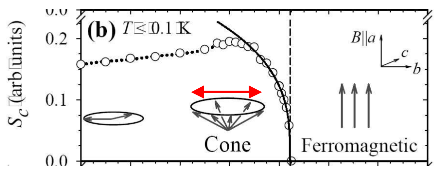
Magnon dispersion at saturation
Neutron scattering observe
S=1 magnons
Cone
Ferromagnet
(0,1.447,0)
Counts (/mon~10 mins.)
Energy (meV)
[0,ζ,ζ-1]
[0,2,l]
Energy (meV)
B = 12 T || a
T < 0.2 K
[0,k,0]
Fourier transform of
couplings J(q)
J = 0.374 meV
J'/J ~ 1/3
2D Hamiltonian
J''/J = 4.5(5) % interlayer exchange
D_a /J = 5.3(5) % DM anisotropy
Incommensurate 2D
dispersion because of
frustration



Transition to cone order: Bose condensation of magnons
B<Bc
B=Bc
B>Bc
magnon
dispersion
Gap Δ
Q
B (T) || a
Energy (meV)
(020)
(010)
CONE
FERRO
Peak intensity
(arb. units)
[0,k,0]
Bragg peaks appear
where the gap closes
B > Bc
B < Bc
Intensity a.u.)
K (rlu)
2nd order
gap-closing transition
Cone
Ferromagnet

Observe non-mean-field scaling laws

Order parameter $\langle S_\perp \rangle$:



$\langle S_\perp \rangle \sim (B_C - B)^{\beta}$

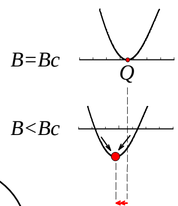
β=0.33(3)

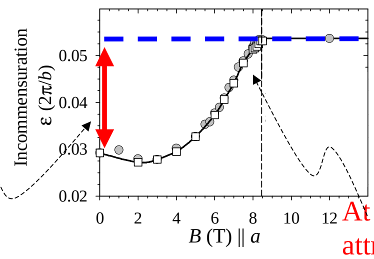
mean field (3D)

β = 0.5

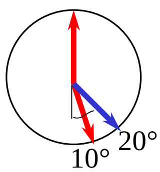
Exponent consistent with 2D BEC (3D XY universality)

Renormalization of ordering wavevector *Q*





mean-field result

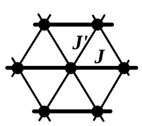


Strong quantum renormalization

$\varepsilon_0/\varepsilon_{cl}$=0.56

At high field could attribute this to magnon interactions in the condensed phase

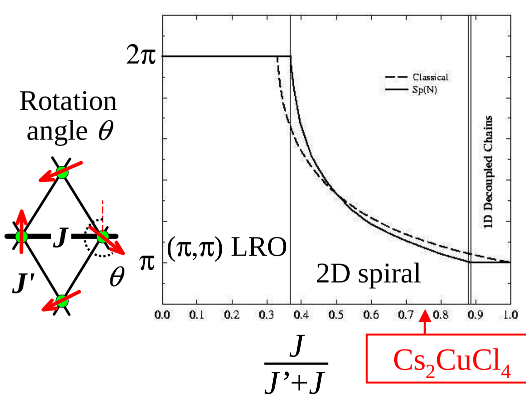
Renormalization of incommensuration by fluctuations



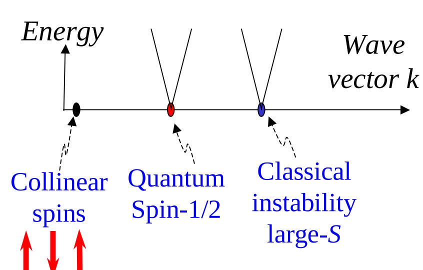
J'/J =1/3

Approximate solutions to the *S*=1/2 quantum problem give:

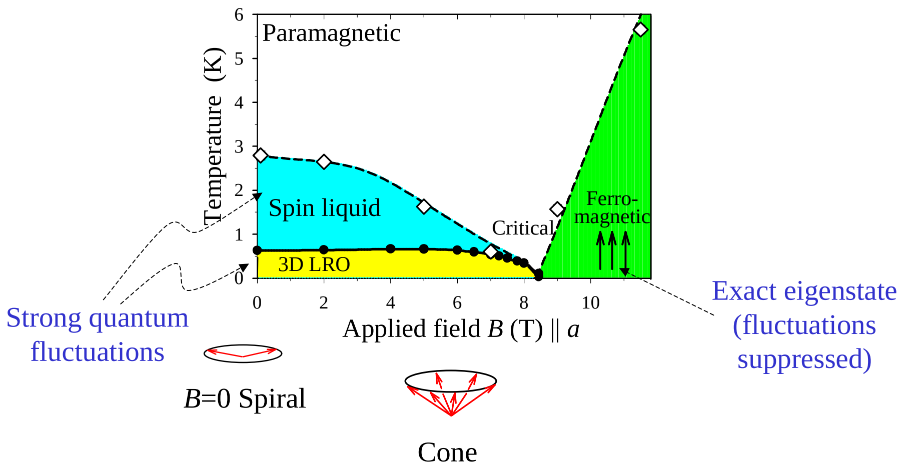
$\varepsilon_0/\varepsilon_{cl}$ = 0.43(1) Series expansions, *Weihong et al (1999)*
= 0.70(3) Large-N, *Chung et al (2001)*
= 0.56 observed



Cs_2CuCl_4

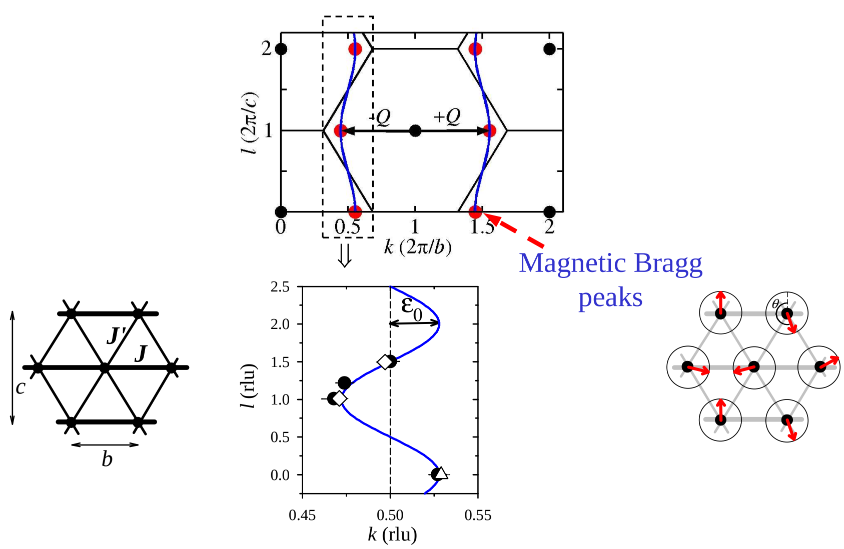


Phase diagram in applied field

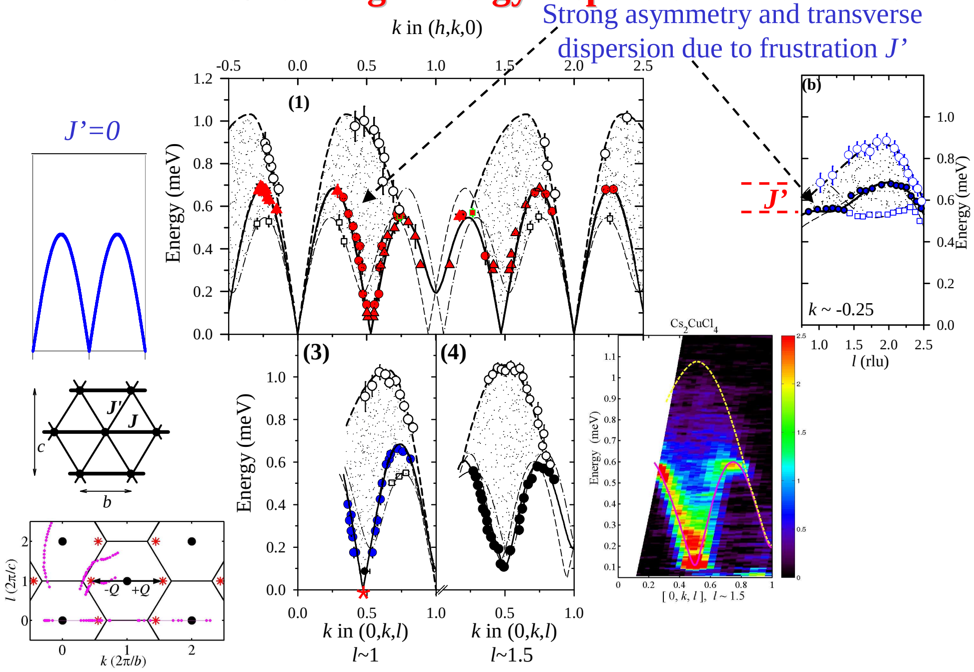


Incommensurate low-energy dispersion in zero field

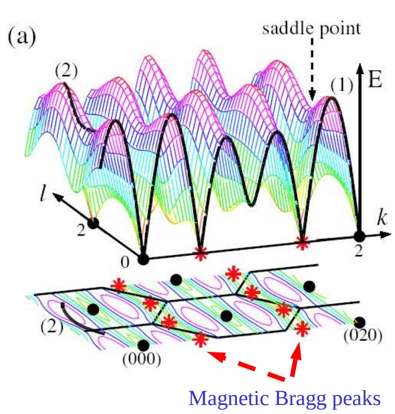
Low-energy dispersion shows
incommensurate 2D modulations



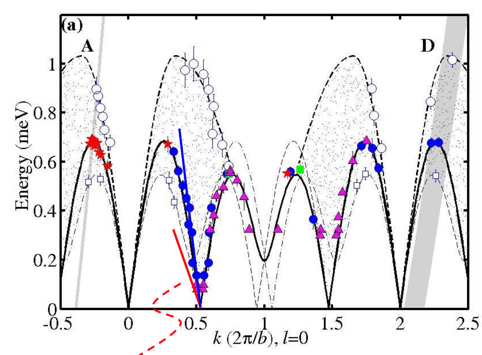
Incommensurate high-energy dispersion in zero field



Dispersion relations in zero field



2D incommenusurate modulations observed both at low and high energies



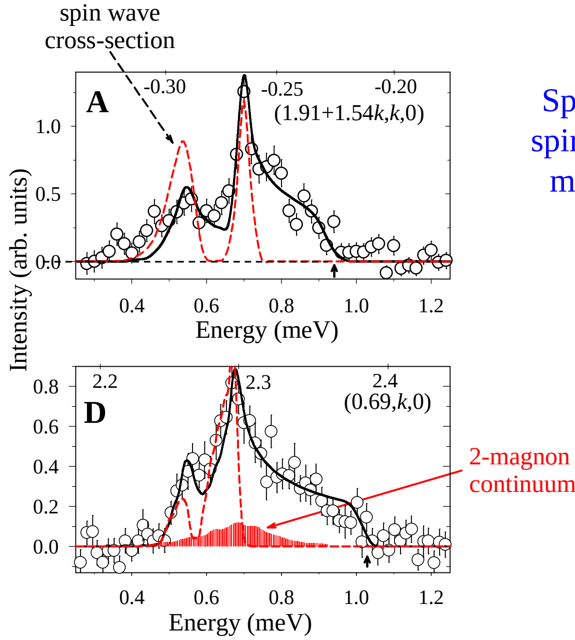
Semi-classical velocity for bare (J,J') couplings

⇒ quantum renormalization R=1.65

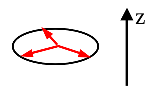
(quantum solution for

1D S=1/2 chain gives R=1.57)

Excitation lineshapes and breakdown of the spin-wave description



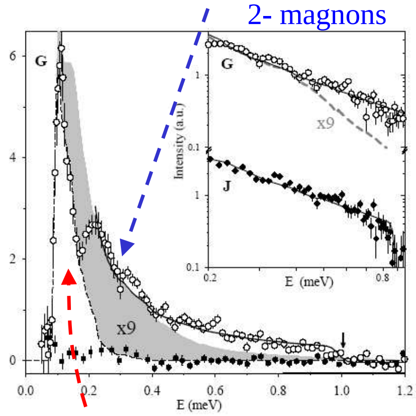
Spin-wave theory for a spiral predicts 3 magnon modes $\Delta S^z = 0, +1, -1$

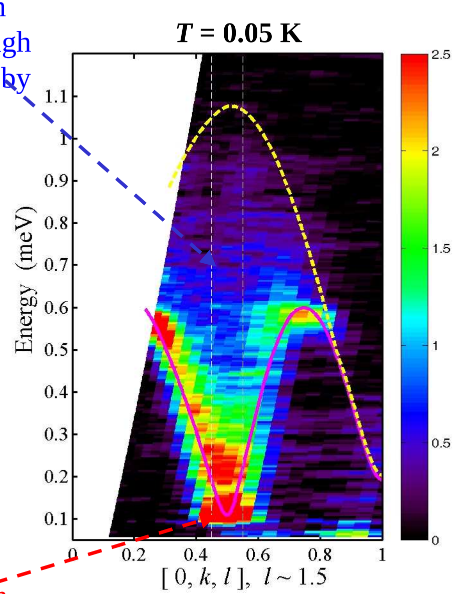


Even higher-order processes in LSWT can not explain the dominant continuum scattering

Analysis of the dominant continuum scattering

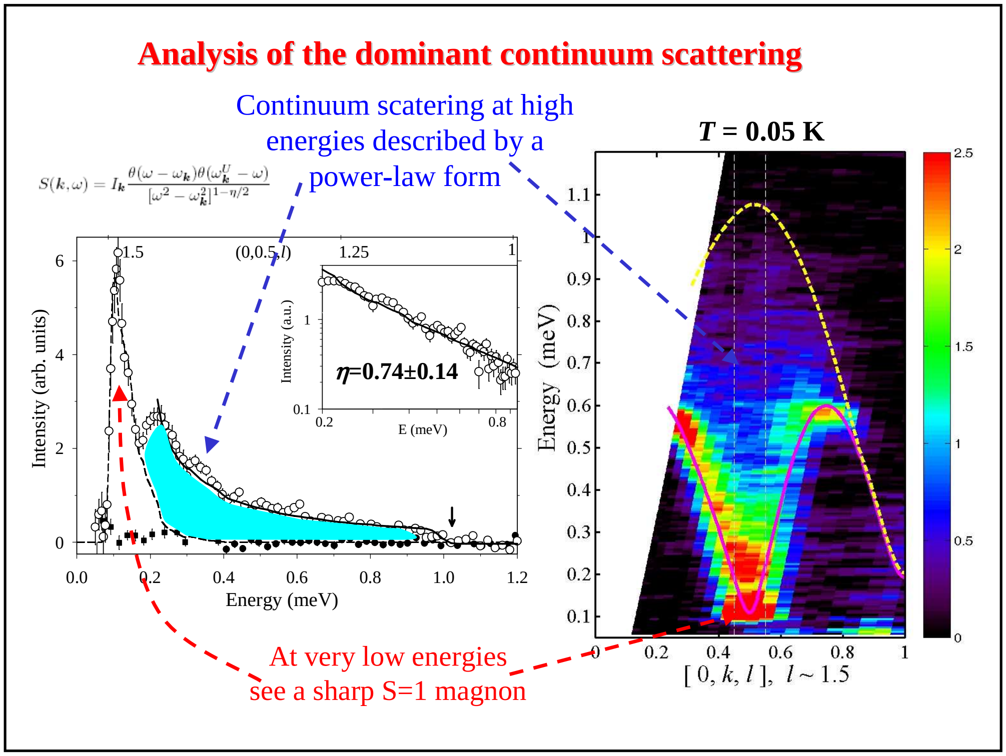
Dominant continuum scatering at medium-high energies not described by 2- magnons

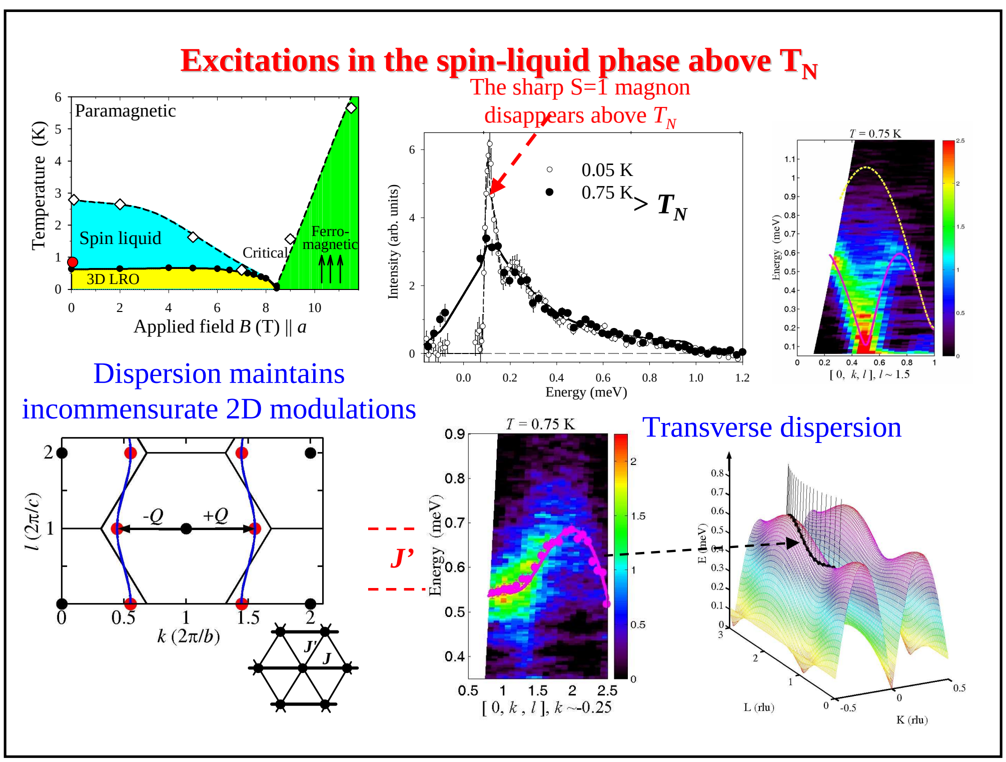




At very low energies see a sharp S=1 magnon

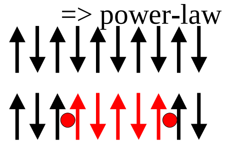
Neutron scattering from spinons in the anisotropic triangular quantum magnet Cs2CuCl4

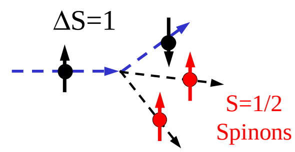


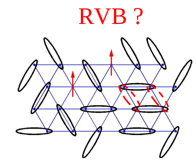


Continuum of scattering and *S*=1/2 spinons

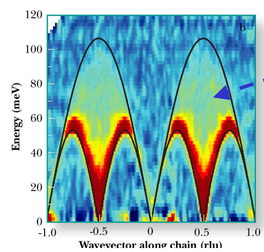
Make analogy with 1D where spin-wave description breaks down
=> power-law correlations and spin-1/2 spinons



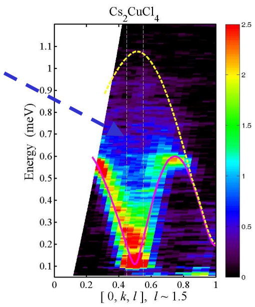


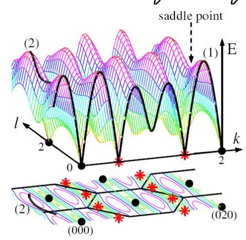


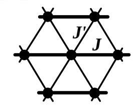
KCuF$_3$ prototypical 1D system



D.A. Tennant, B. Lake, S.E. Nagler and C.D. Frost





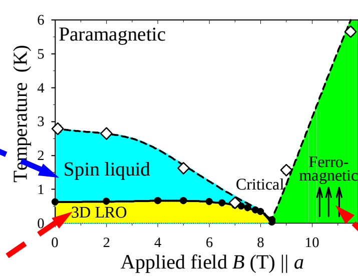


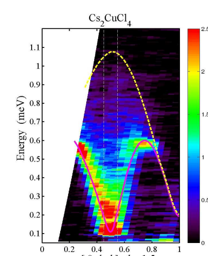
- Interchain couplings small J'/J~ 5%
- unfrustrated, commensurate
- high energies dispersion essentially 1D

- Interchain couplings **large** J'/J~ 34% (2D)
- frustrated
- incommensurate dispersion, shows strong 2D modulations at all energies

Physical picture of excitations

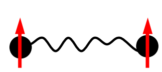
***S*=1/2 spinons at all energy scales**



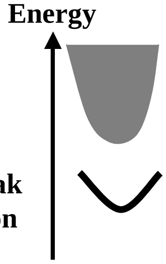


***S*=1/2 spinons at medium energies**
***S*=1 magnon at low energies**

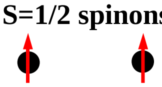
***S*=1 magnon only**



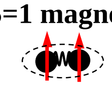
***S*=1/2 Spinons + weak attractive interaction (mean field)**

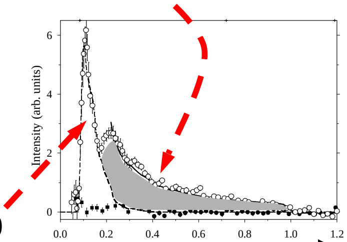


Continuum of de-confined S=1/2 spinons

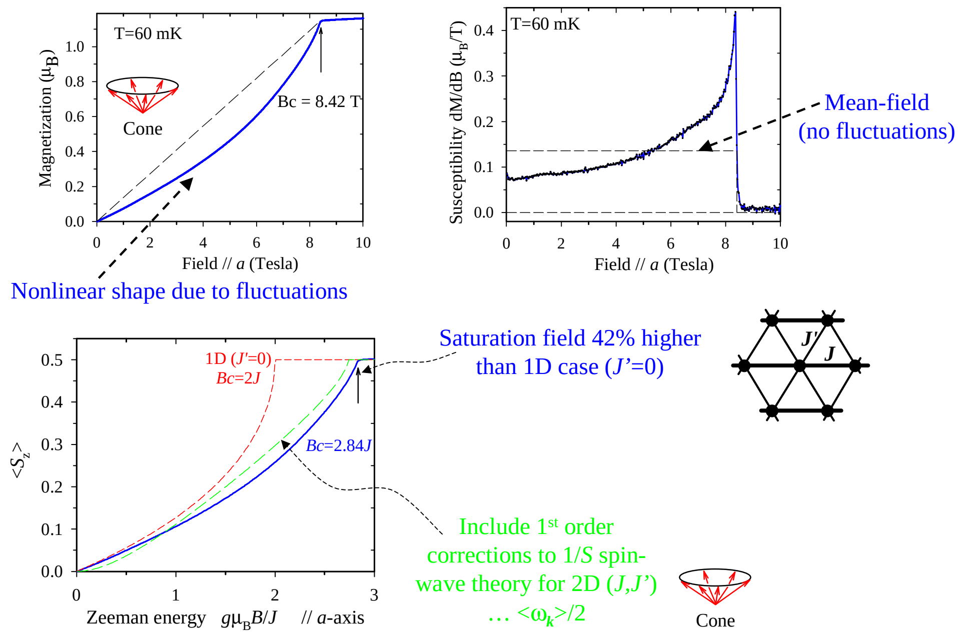


Bound state (S=1 magnon)

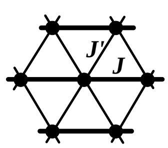


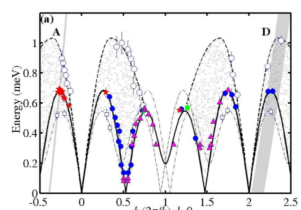


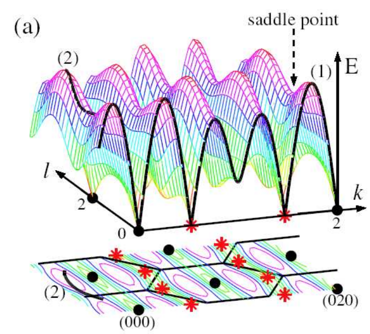
Magnetization curve in Cs_2CuCl_4



Summary & conclusions







1. Cs_2CuCl_4 is spin-1/2 HAF on anisotropic triangular lattice (2D) : $J'/J \sim 1/3$

2. See excitations continua in neutron scattering, as characteristic of deconfined S=1/2 spinons

3. Dispersion relations show 2D incommensurate modulations

4. Intensities described by a 2-spinon power-law lineshape

5. Excitations velocity renormalized from classical (R=1.65)

6. Observe large incommensuration effects in field on the scale of band filling of S=1/2 spinon orbitals