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SMR.1320 - 16

# SUMMER SCHOOL
on
LOW-DIMENSIONAL QUANTUM SYSTEMS:
Theory and Experiment
(16 - 27 JULY 2001)

PLUS

PRE-TUTORIAL SESSIONS
(11 - 13 JULY 2001)

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# USING NEUTRON SCATTERING AND MAGNETIC FIELDS TO EXPLORE NEW PHYSICS IN Cs2CuC14

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# Using Neutron Scattering and Magnetic Fields to Explore New Physics in Cs2CuCl4

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In collaboration with: (Cs2CuCl4)

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A.M. Tsvelik (BNL), Klaus Habicht, Peter Smeibidl, Michael Hoffman (HMI, Berlin).

(KCuF3)

Bella Lake, Stephen Nagler (ORNL) and Chris Frost (ISIS)

# Outline of Presentation

- 1. Introduction
- 2. 1D S=1/2 Heisenberg antiferromagnet.
- 3. Using high magnetic fields to find a spin Hamiltonian experimentally.
- 4. 2D S=1/2 Frustrated Heisenberg antiferromagnet.
- Conclusions

# 1 INTRODUCTION

AIMS:-

- WANT TO LOOK AT AND MANIPULATE STRONGLY CORRELATED QUANTUM STATES EXPERIMENTALLY.
- DEVELOP TECHNIQUES SO WE CAN FIND NEW TYPES OF BEHAVIOUR "IN THE LABORATORY" AND EXEMPLARY SYSTEMS.

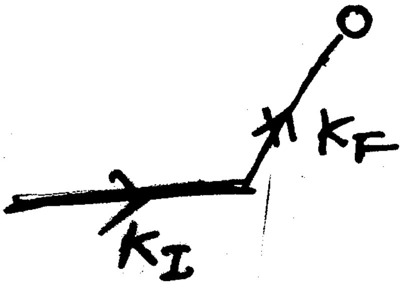
USE :-

1.1 NEUTRON TECHNOLOGY

1.2 QUANTUM MAGNETS

## 1.1 NEUTRON TECHNOLOGY

NEUTRON IS NEUTRAL $S=1/2$ PARTICLE. WEAKLY INTERACTING. SIMPLE SCATTERING MATRIX ELEMENT FOR MAGNETIC SCATTERING



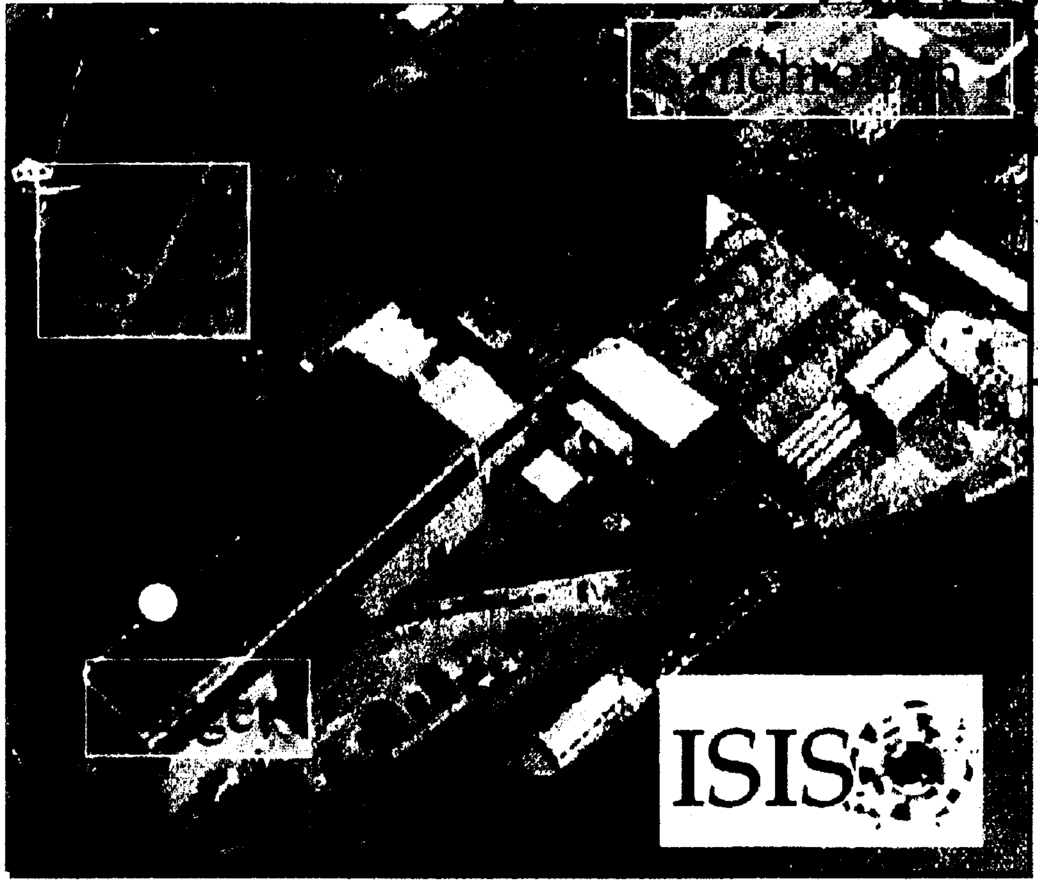
$$Q = K_I - K_F$$

$$\hbar\omega = E_I - E_F$$

$$S(Q,\omega) \propto |\langle E|S^{\alpha}_{Q}|G\rangle|^2$$

$$\propto \frac{e^{\omega/kT}}{e^{\omega/kT}-1}\,\mathrm{Im}\,\chi(Q,\omega)$$

*Protons accelerated in 800MeV Synchrotron*

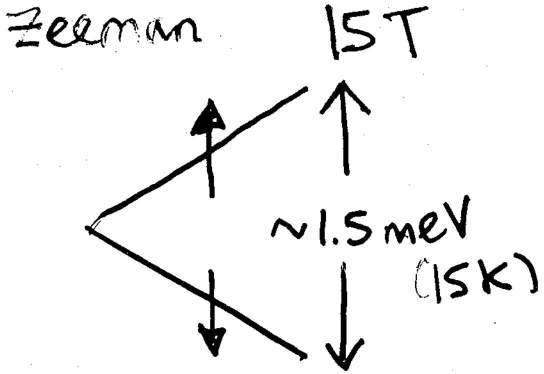


*Protons hit heavy metal nuclei in the Target which emit neutrons*

-5-

# 3.3 The neutron scattering measurements

## Inside the Reactor Building at HMI, Berlin

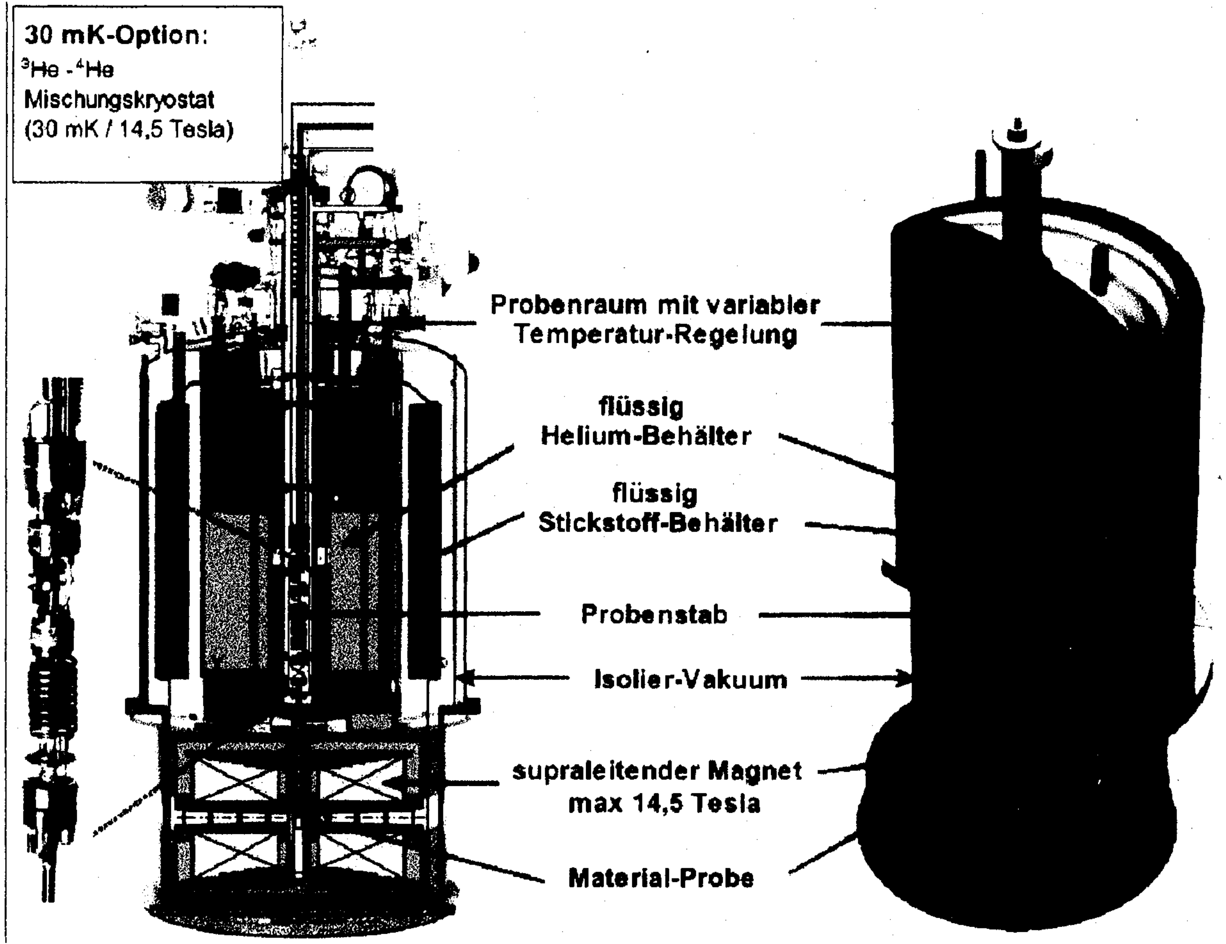


# HMI 14.5 Tesla superconducting magnet with dilution insert.

*To probe quantum effects we need extreme environments:*

30 mK is about 1/10000th Room Temp.

14 Tesla is about 1,000,000 times Earths surface field (frogs levitate at 10 Tesla!)



# 1.3 Low-Dimensional Quantum Magnets

- Quantum magnets provide examples of strongly correlated systems showing *novel ground and excited states*.
- They can be studied *in great detail experimentally*.
- Their quantum states can be manipulated by magnetic fields *in a clean way*.
- The Hamiltonians are *simple but nontrivial*.
- *Nonlinearity* is embedded in the spin commutation relations.

*Some of the seminal physics problems in the development of the field include*: -

- Haldane gaps in integer spin chains
- Spinons in half-odd-integer chains
- Spin-charge separation in doped antiferromagnets
- Hole pairing in conducting spin ladders

# 2. 1D S=1/2 Heisenberg Antiferromagnet

- 2.1 Conventional methods break down.
- 2.2 A physical picture emerges.
- 2.3 Experimental aspects:
  - 2.3.1 Spinon continuum and scaling.
  - 2.3.2 Renormalization of the energy scale.
  - 2.3.3 Filling a band of spinons.

# $S=1/2$ Heisenberg AF Chain

$$H = J \sum_I \vec{S}_I \cdot \vec{S}_{I+1} \qquad S = 1/2 \,,\; J > 0$$

Spinwaves: zero point fluctuations destroy Néel ground state.

Spin-1/2 (on site): Rotation operator $R(2\pi) = e^{i2\pi S} = -1$ for $S=1/2$. Complex phase factor in semiclassical mapping.

## Advanced Methods

1. **BETHE ANSATZ:** $T=0$ ground state $\langle S^z_r S^z_{r+d} \rangle \propto 1/d$ (algebraic decay "quasi-long-range" order).

   Excitation spectrum $\omega_{dCP} = \frac{\pi}{2} J |\sin q|$

   $\pi/2$ higher than classical spinwaves.

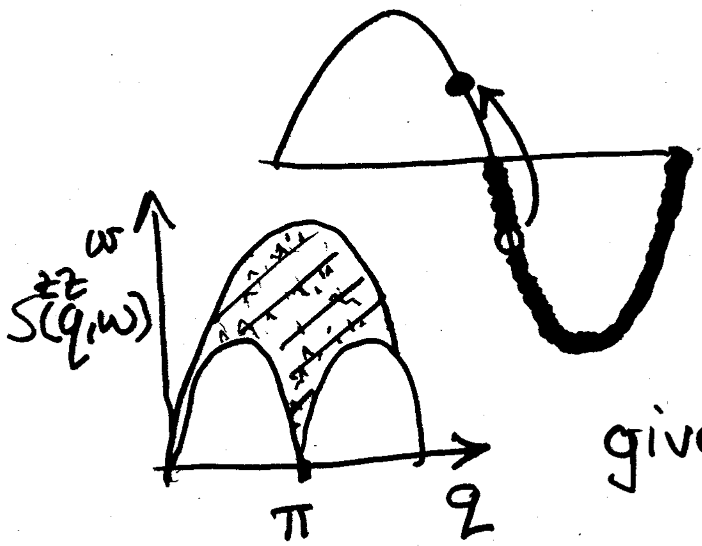
   $\boxed{\textbf{Excitations are } S=1/2 \textbf{ spinons.}}$

2. **Bosonization & CFT:** around $q=\pi$ (continuum approx.) Luttinger liquid Hamiltonian. $T$, $\omega$ & $q$ spin correlations known:

$$\boxed{\begin{gathered} S(q,\omega;T) = \frac{e^{\omega/kT}}{e^{\omega/kT}-1} \cdot \frac{A}{T} \cdot \mathrm{Im}\left[\rho\left(\frac{\omega - v|q-\pi|}{4\pi T}\right) \rho\left(\frac{\omega + v|q-\pi|}{4\pi T}\right)\right] \\ \rho(x) = \Gamma(1/4 - ix)\,\Gamma(3/4 - ix). \text{ Schulz '86.} \end{gathered}}$$

# Applied Field: Band Filling.

$B=0$ Jordan-Wigner Fermion Band.

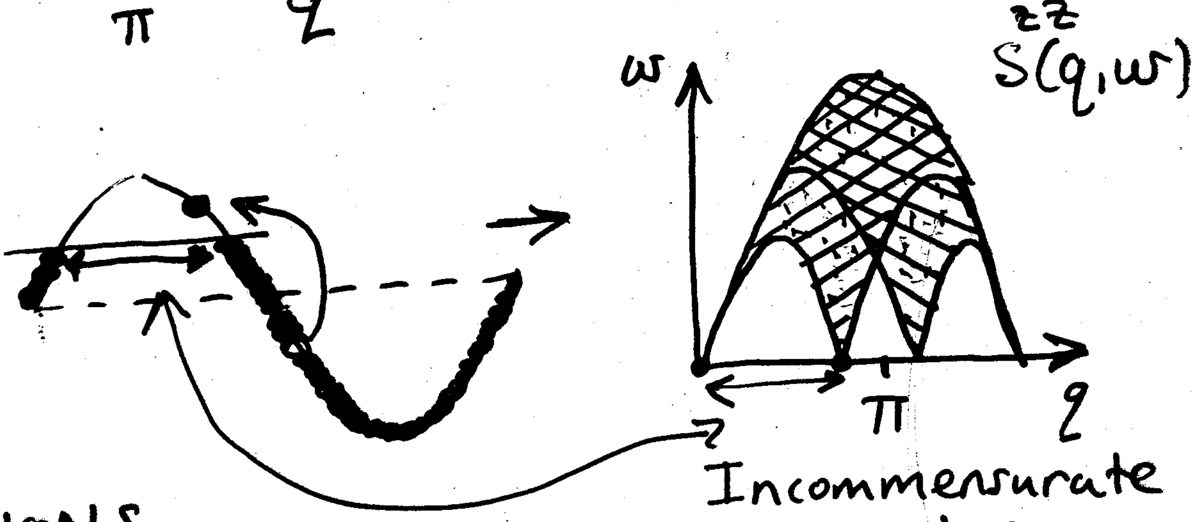


$\frac{1}{2}$ Filled

$S^zS^z$ correlations = Particle-Hole pair.

gives $S^{zz}(q,\omega)$ continuum.

$B>0$



Incommensurate wavevector.

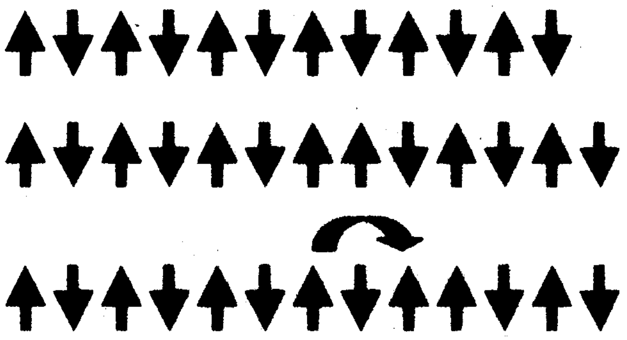
## 3. SPINONS

Some exact results are known.

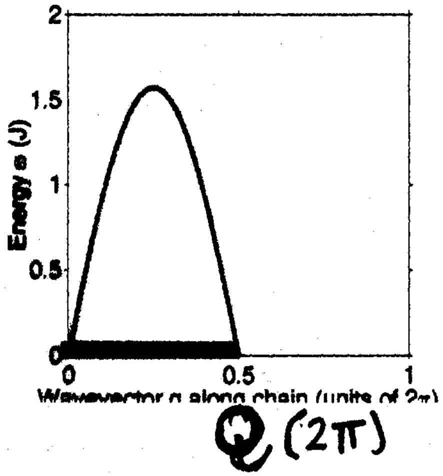
- $1/r^2$ Haldane-Shastry model exactly solved
- 2-spinon neutron scattering exact (75%)

# A physical picture

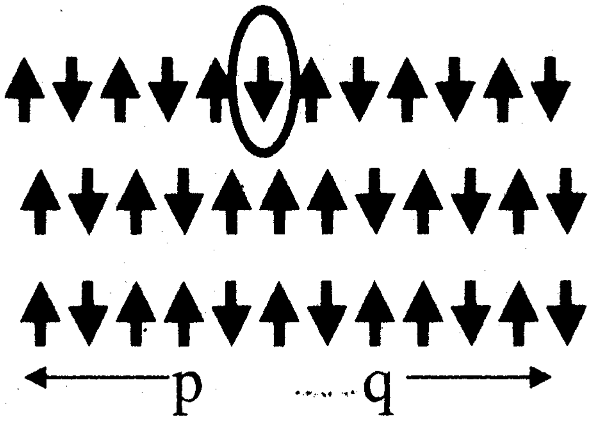
Instead of semiclassical spin-1 spinwaves a new picture of spin-1/2 spinons emerges



1. Ground state is quasi-long-range ordered
2. A spinon is a spin-1/2 inserted into the ground state. It also looks like a π twist.
3. Spinons are restricted to hopping only to every second site. Exist over ½ B.Z.



Spinon dispersion



4. Neutrons scatter by flipping over a spin creating two spinons which partition the energy and wavevector.

NB: NEUTRON CHANGES S BY 1 UNIT NOT S=1/2. MUST CREATE PAIRS OF SPINONS.

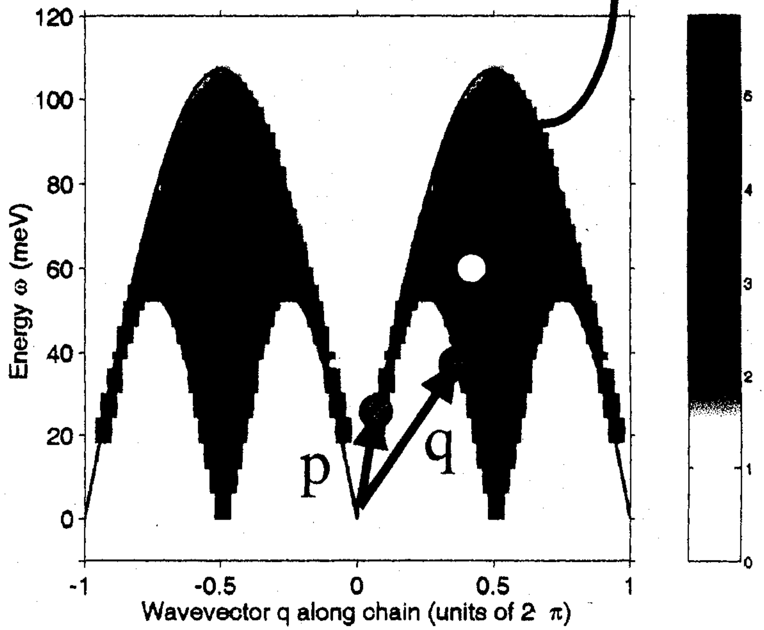
# A physical picture cont...

Two-spinon neutron scattering event.

$$S(q,\omega)=\frac{\Theta(\omega-\omega_L)\Theta(\omega_U-\omega)}{\sqrt{\omega^2-\omega^2{}_L}}$$

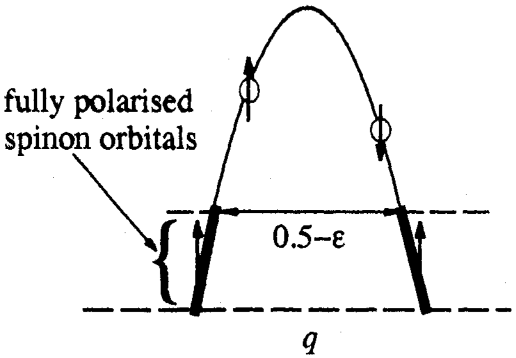
(AGREES WITH T=0 LIMIT OF BOSONIZATION)

$\omega_L$ =lower and $\omega_U$ =upper boundary



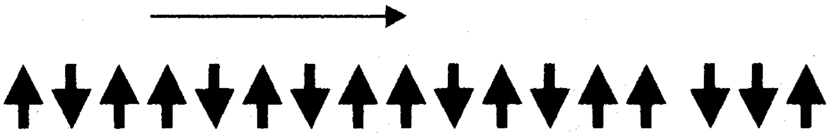
W=w(p)+w(q)
Q=p+q

Condensation of spinons in field



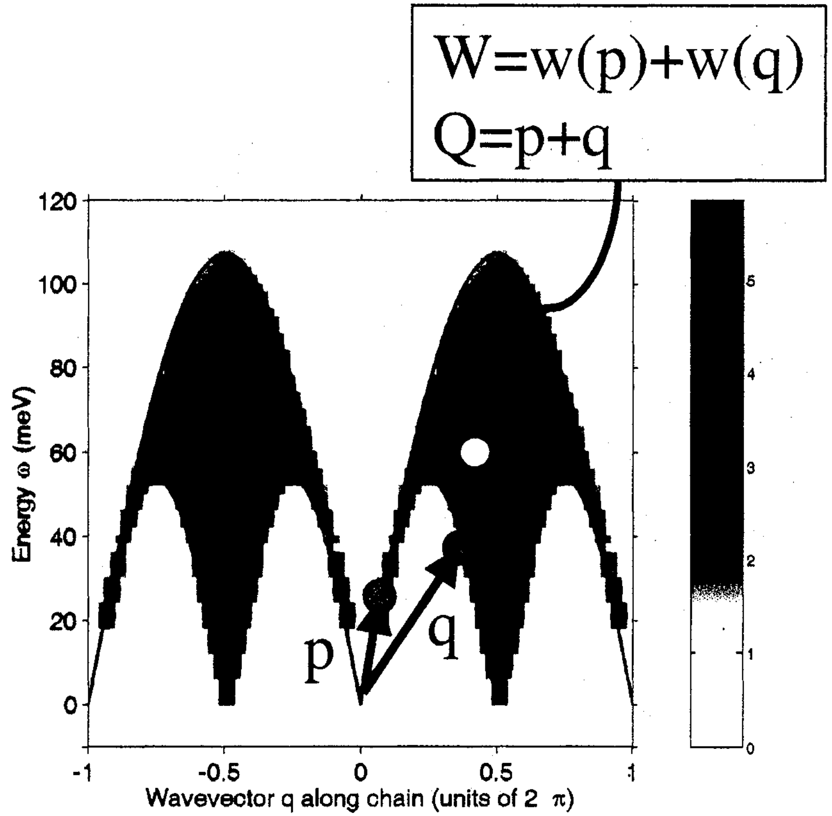
Magnetic fields polarise the spin chain. The spinons are like pockets of spin which repel each other.

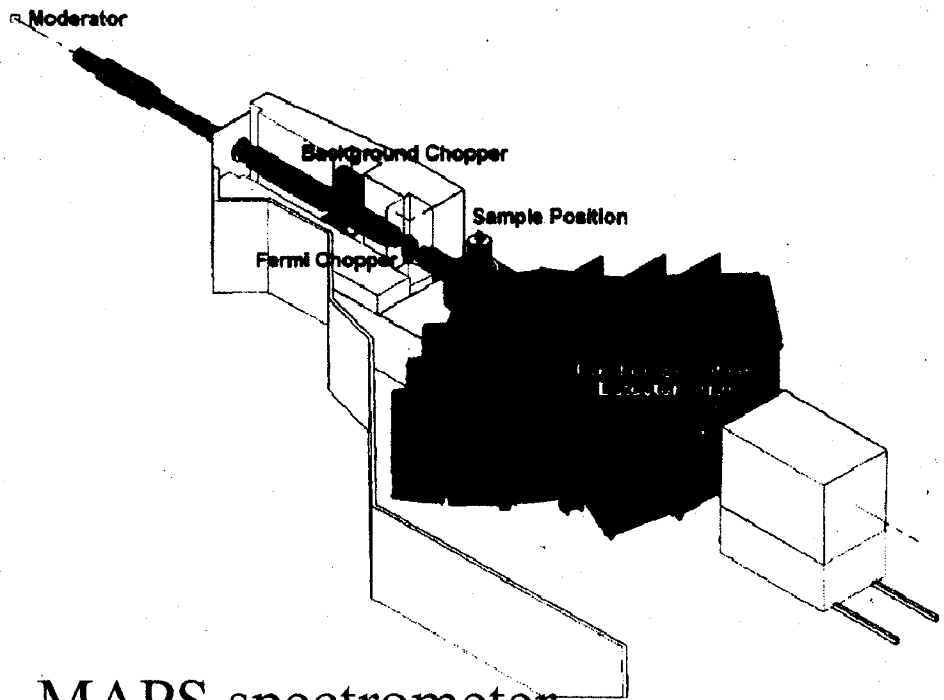
They space out evenly 1/5th of total magnetization:



# 2.3 Experimental results

Predicted neutron scattering





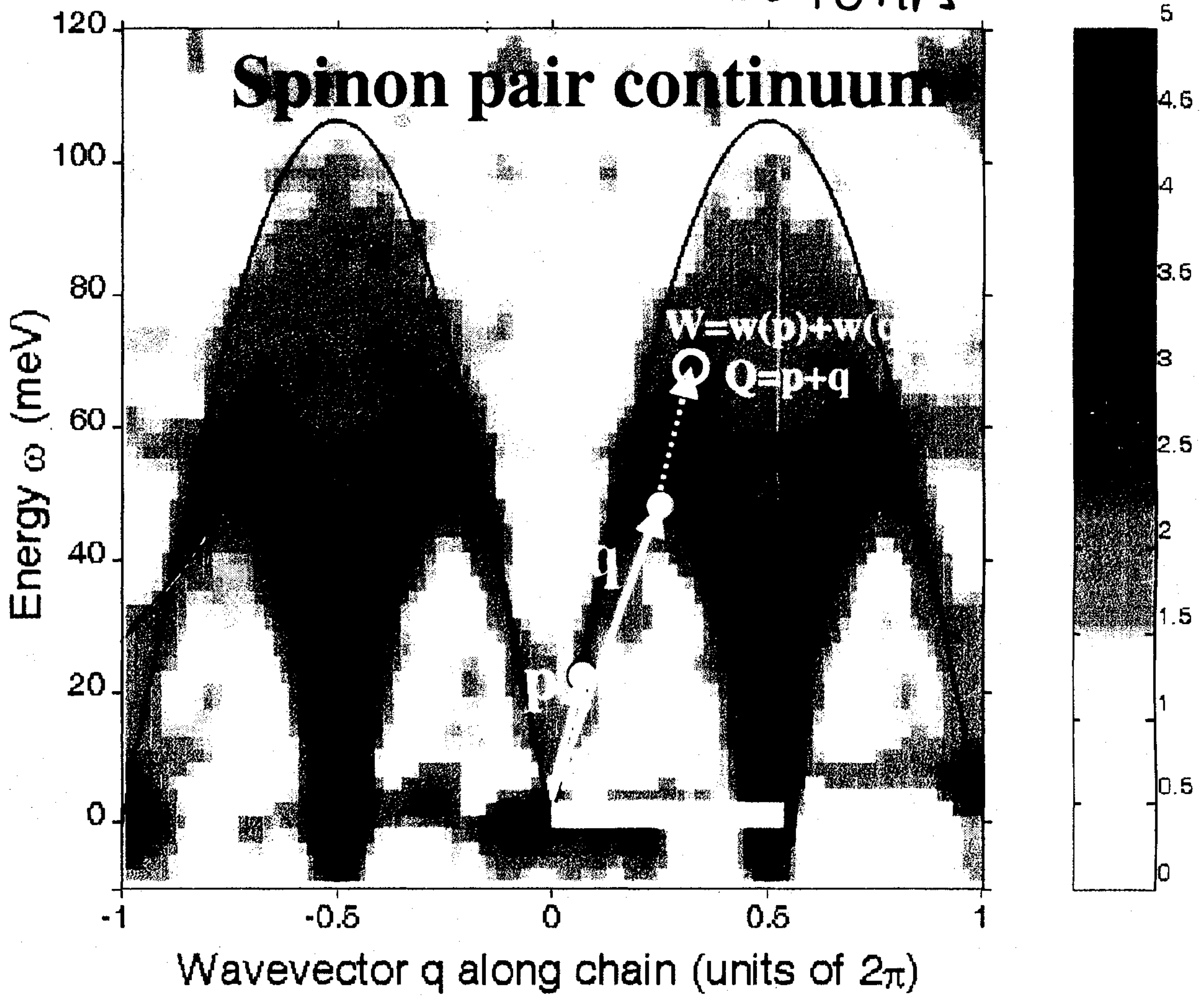
MAPS spectrometer

~40,000 detector pixels
200Mb data per run.

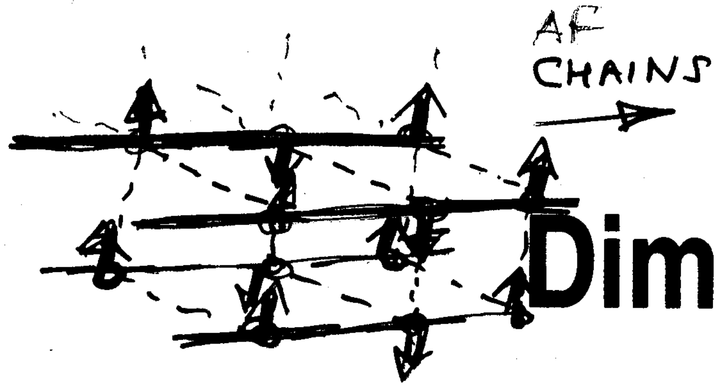
# 2.3.1 Excitation Continuum

Inelastic neutron scattering measurement of the *dynamical correlations* in the S=1/2 Heisenberg Antiferromagnetic chain KCuF3 across two Brillouin Zones. Agrees with predictions

~16hrs



MAPS 2000

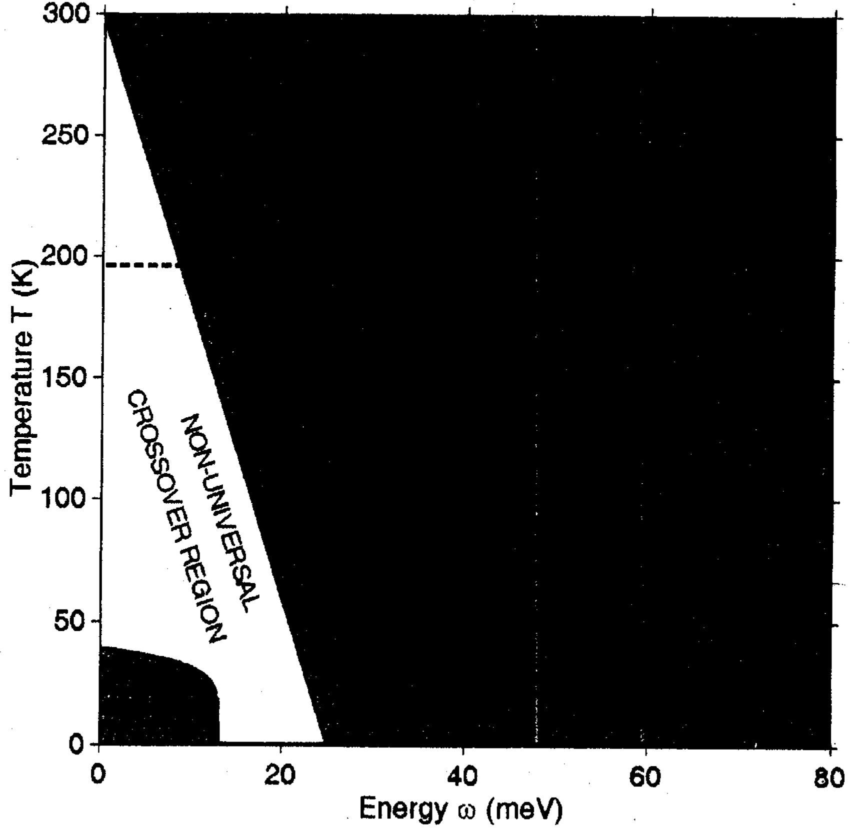


"RPA approaches" and studies of RG flows from 1D RGT to 3D RGT AGREE WITH ω,q,T correlations.

# Dimensional Crossover

In Nature we only have approximations to Heisenberg chains and the 1D field theories only apply at temperatures, energies, and wavevectors where interchain effects are not important.

Affleck, JPhysA 29, 2627 (1996)
Schulz, PRL 77, 2790 (1996)
Essler, PRB 56, 11001 (1997).



(EXPERIMENTAL CROSSOVER DIAGRAM)

Measurement of the
*1D static correlations*
in KCuF3 across a Brillouin
Zone.

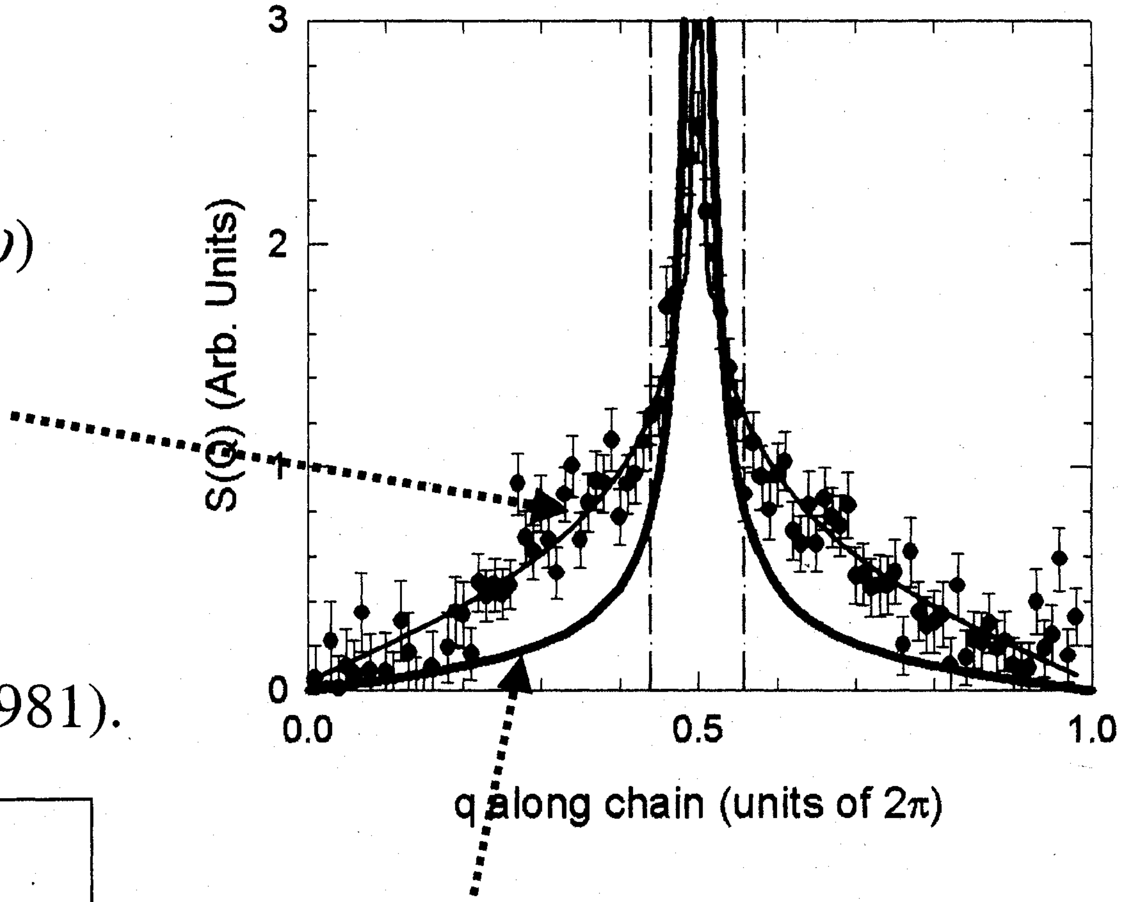
$$S(q) = S(q, t=0) = \int_{-\infty}^{\infty} d\omega S(q,\omega)$$

$$\cong \frac{A}{2\pi} \ln \frac{1+\sin(q/2)}{\cos(q/2)}, (q \geq 0).$$

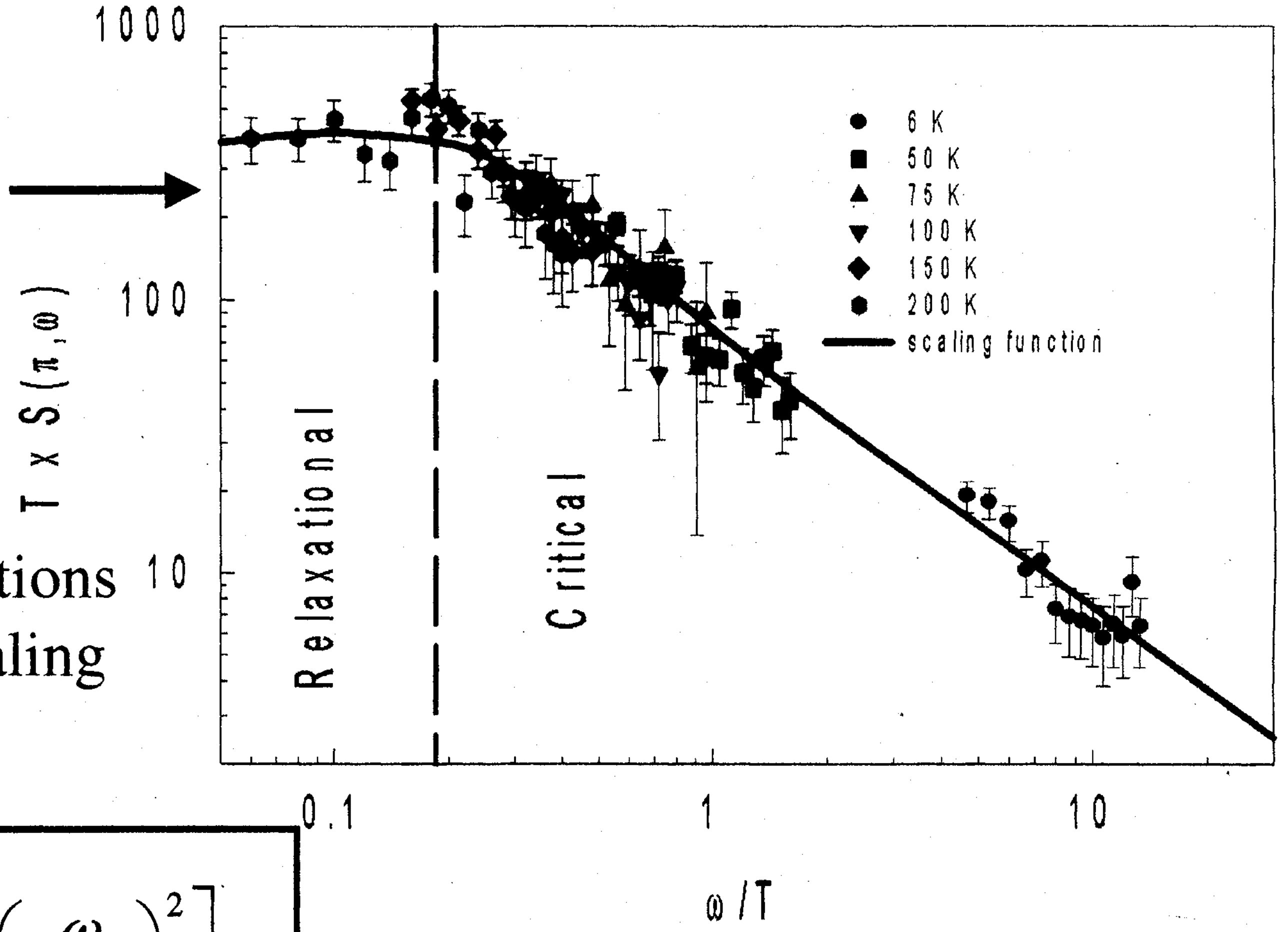
$$\therefore < S_r S_{r+d} > \propto 1/d, (q \sim \pi)$$

Muller et al PRB24, 1429 (1981).

Algebraically 1/d decaying
correlations in the ground state.



Spinwave model result



The 1D dynamical correlations around $q=\pi$ show $\omega/T$ scaling and agree with CFT.

$$S(\pi,\omega)=\frac{e^{\omega/kT}}{e^{\omega/kT}-1}\frac{A}{T}\mathrm{Im}\left[\rho\left(\frac{\omega}{4\pi T}\right)^{2}\right];$$

$$\rho(x)=\Gamma(1/4-ix)/\Gamma(3/4-ix).$$

Functional form derived by H.J. Schulz, PRB34, 6372 (1986)

# 3. Using high magnetic fields to measure the spin Hamiltonian

- 3.1 Excitations in the fully polarized state
- 3.2 The material: Cs2CuCl4
- 3.3 The measurements
- 3.4 Results

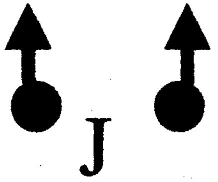
# 3.1 Excitations in the fully polarized state

(see next talk by **Radu Coldea**)

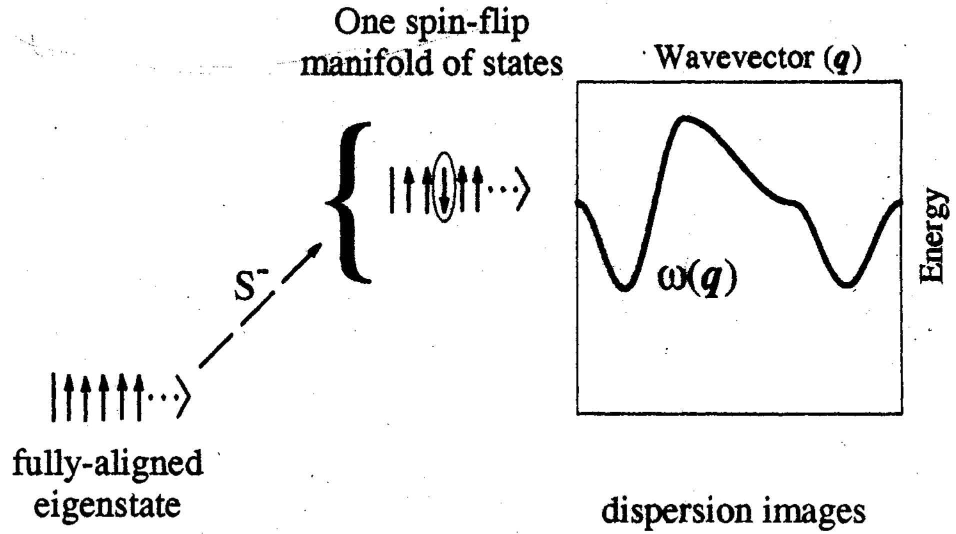
**- neutrons flip over one spin** $S^{-}|\uparrow\uparrow\uparrow\uparrow\uparrow\cdots\rangle = |\uparrow\uparrow\downarrow\uparrow\uparrow\cdots\rangle$

For antiferromagnetic couplings neighbouring spins like to point antiparallel

In a big enough magnetic field the spins become fully polarized along the field direction



The excitations in this state have remarkable properties. (True for any magnet with conserved Sz).



dispersion images exchange Hamiltonian

exact result

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

Fourier transform of magnetic couplings

Zeeman energy

$$J(\boldsymbol{q}) = \Sigma_{ij}\ J_{ij}\ e^{i\boldsymbol{q}\boldsymbol{r}_{ij}}$$

## 3.2 The material: Cs2CuCl4

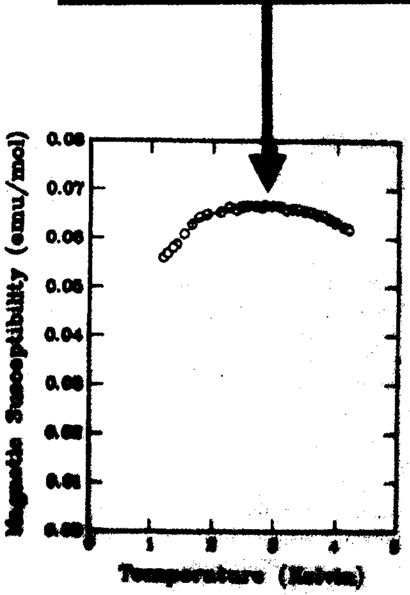
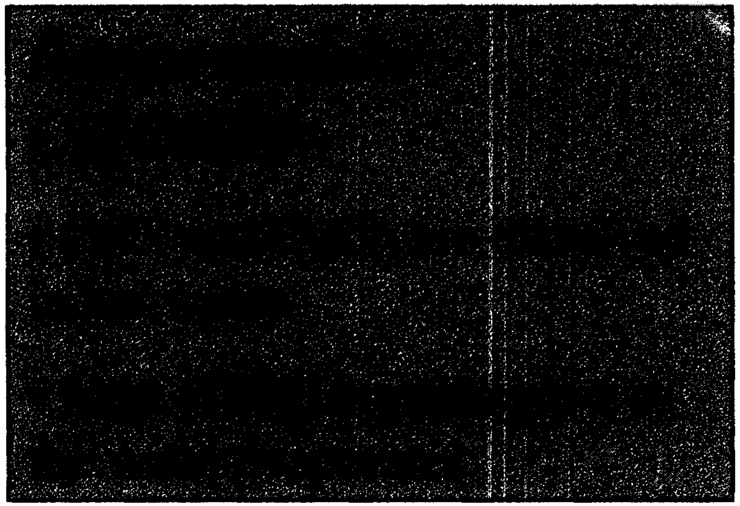


FIG. 2. Experimental susceptibility of polycrystalline $Cs_2CuCl_4$ (points). A fit of the data to the molecular-field-corrected Bonner–Fisher theory, as described in the text, is illustrated by the solid curve.

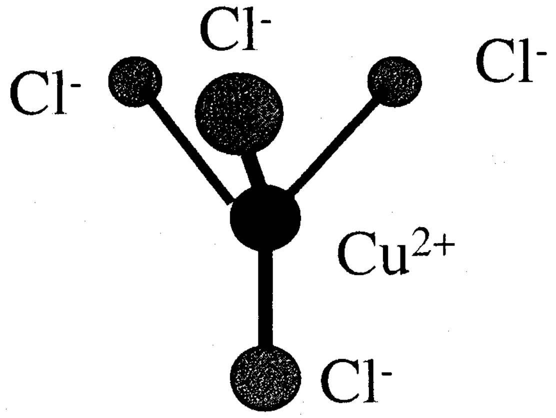
- Zeeman splitting of an S=1/2 magnetic moment is about 0.1meV for a field of 1Tesla.
- Exchange energy is about 0.3 meV.
- Expect fields of about 8 Tesla to fully polarize Cs2CuCl4.
- Energy window where cold neutrons and high-fields overlap.

**Carlin et al, J. Appl. Phys.**57,3351 (1985)

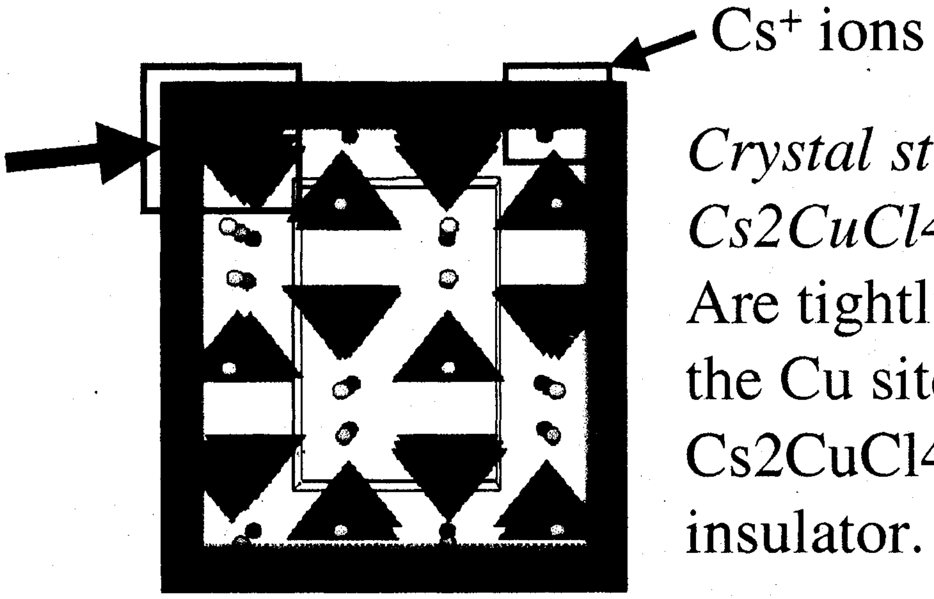


- $Cu^{2+}$ ion has $3d^9$ outer shell i.e. 1 hole.
- The orbital moment is quenched by the four $Cl^-$ ions.
- Near **isotropic spin-1/2** (within 1%)

$CuCl_4^{2-}$ tetrahedra



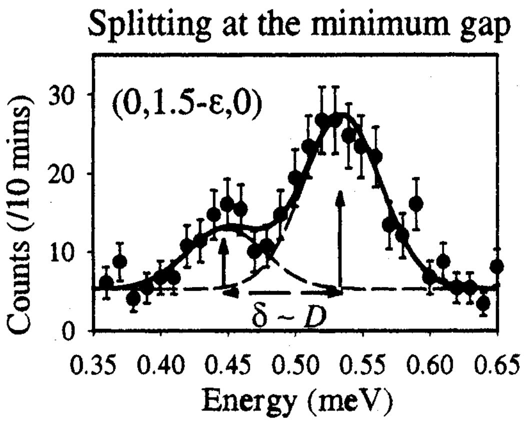
Tetrahedrally coordinated Cu ion

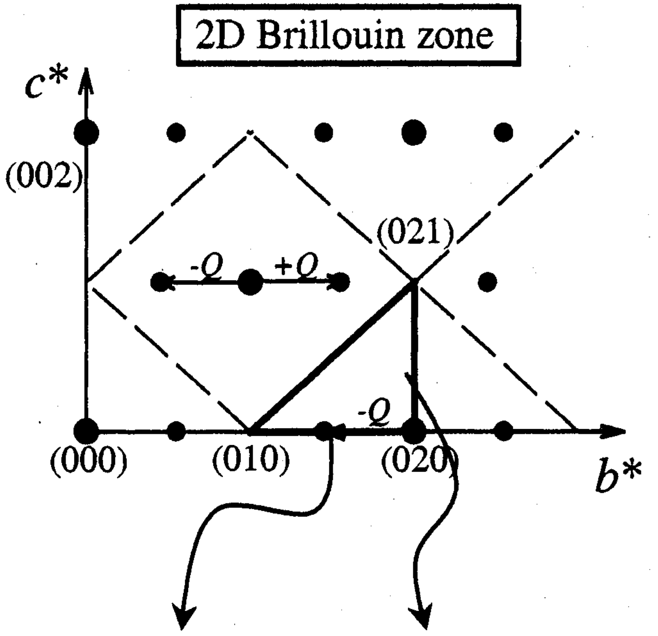


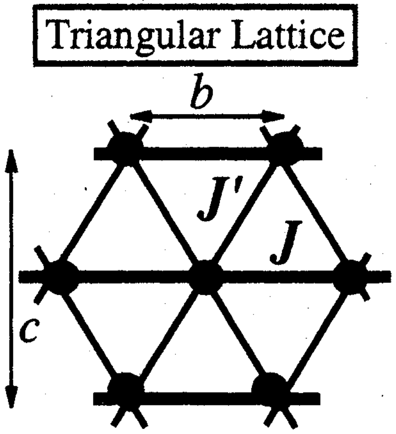
*Crystal structure of Cs2CuCl4*. The holes Are tightly bound to the Cu sites and so Cs2CuCl4 is an insulator.

# 3.4 Dispersion of the magnons in the ferromagnetic state

A typical scan in energy





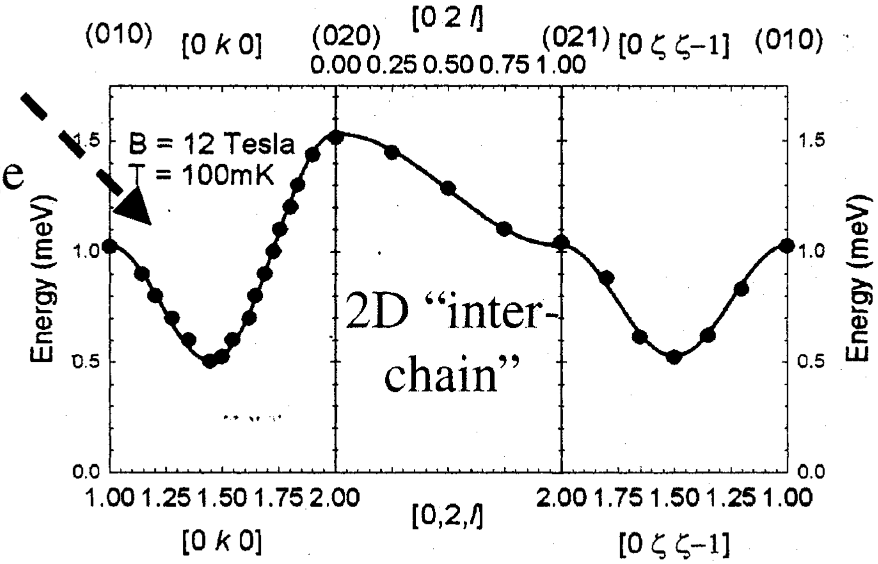


**Quasi-2D Hamiltonian**

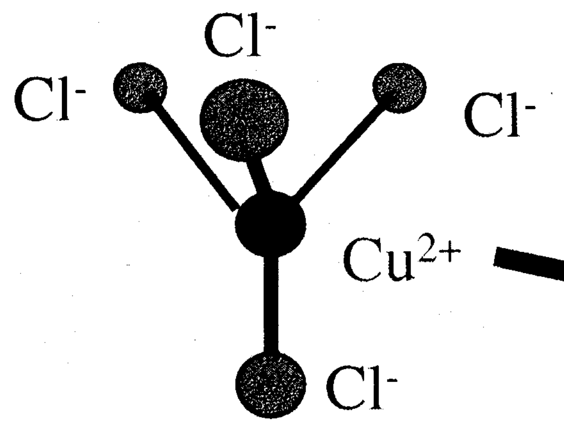
$J$=0.376 meV

$J'$=0.124 meV

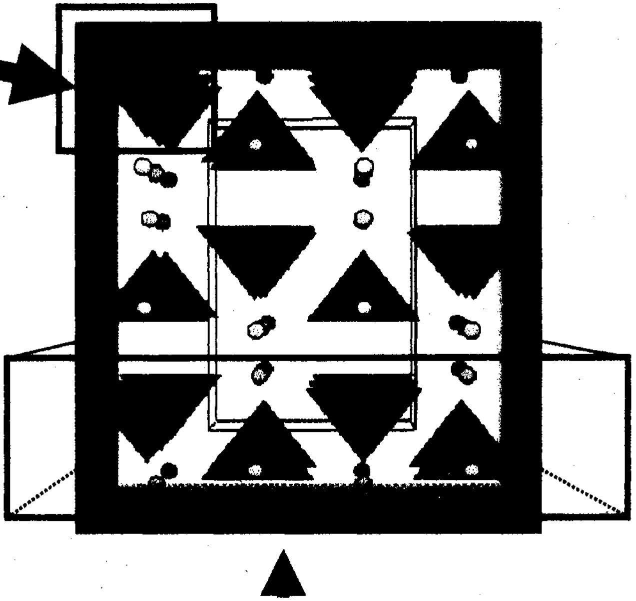
Fourier transform of exchange couplings



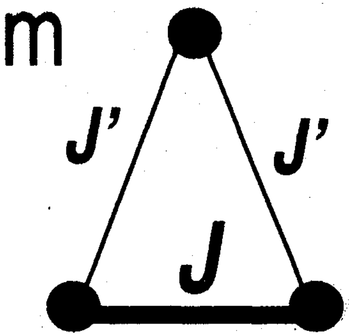
# Magnetically Cs2CuCl4 is a 2D S=1/2 quantum antiferromagnet.



Tetrahedrally coordinated Cu ion

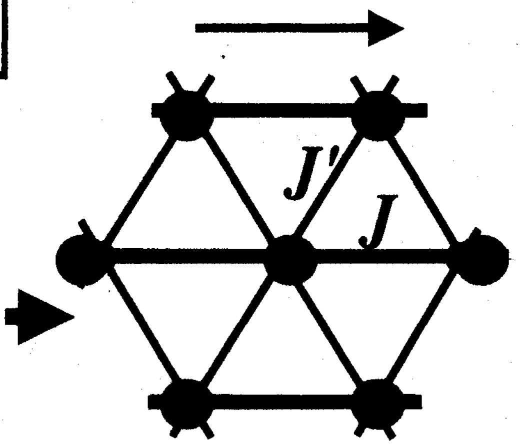


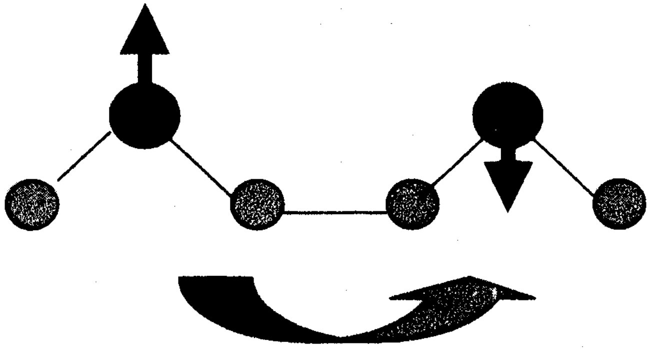
Layers of S=1/2 Coppers coupled in An isoscelese triangle geometry



Isoscelese triangle building block

"Chains"



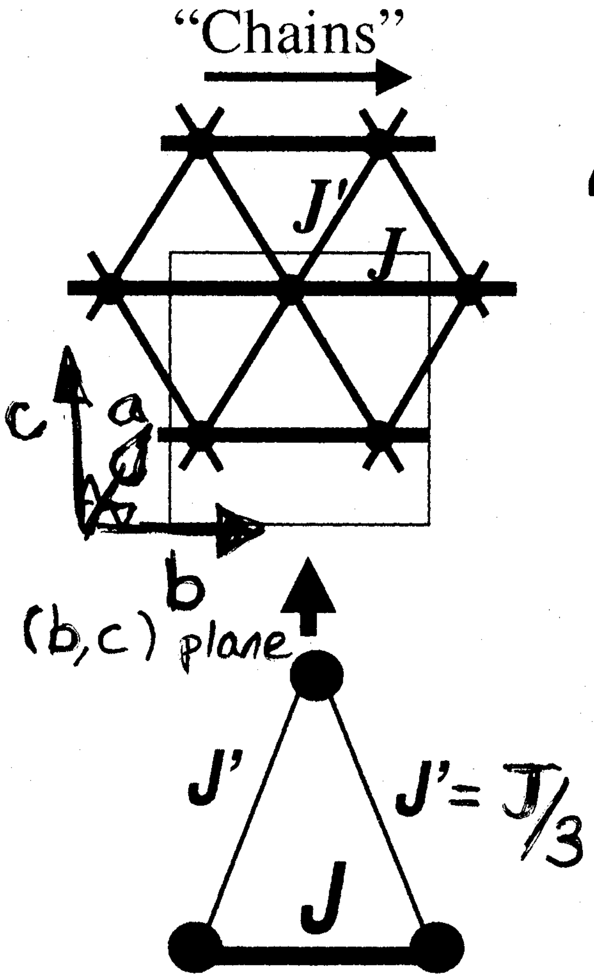


Superexchange is via two chlorine ions.

# 4. A 2D S=1/2 Frustrated Heisenberg Antiferromagnet

- 4.1 Preliminaries
- 4.2 Quantum renormalization effects
- 4.3 Excitation continua
- 4.4 Field effects
- 4.5 Conclusions

# 4.1 Preliminaries



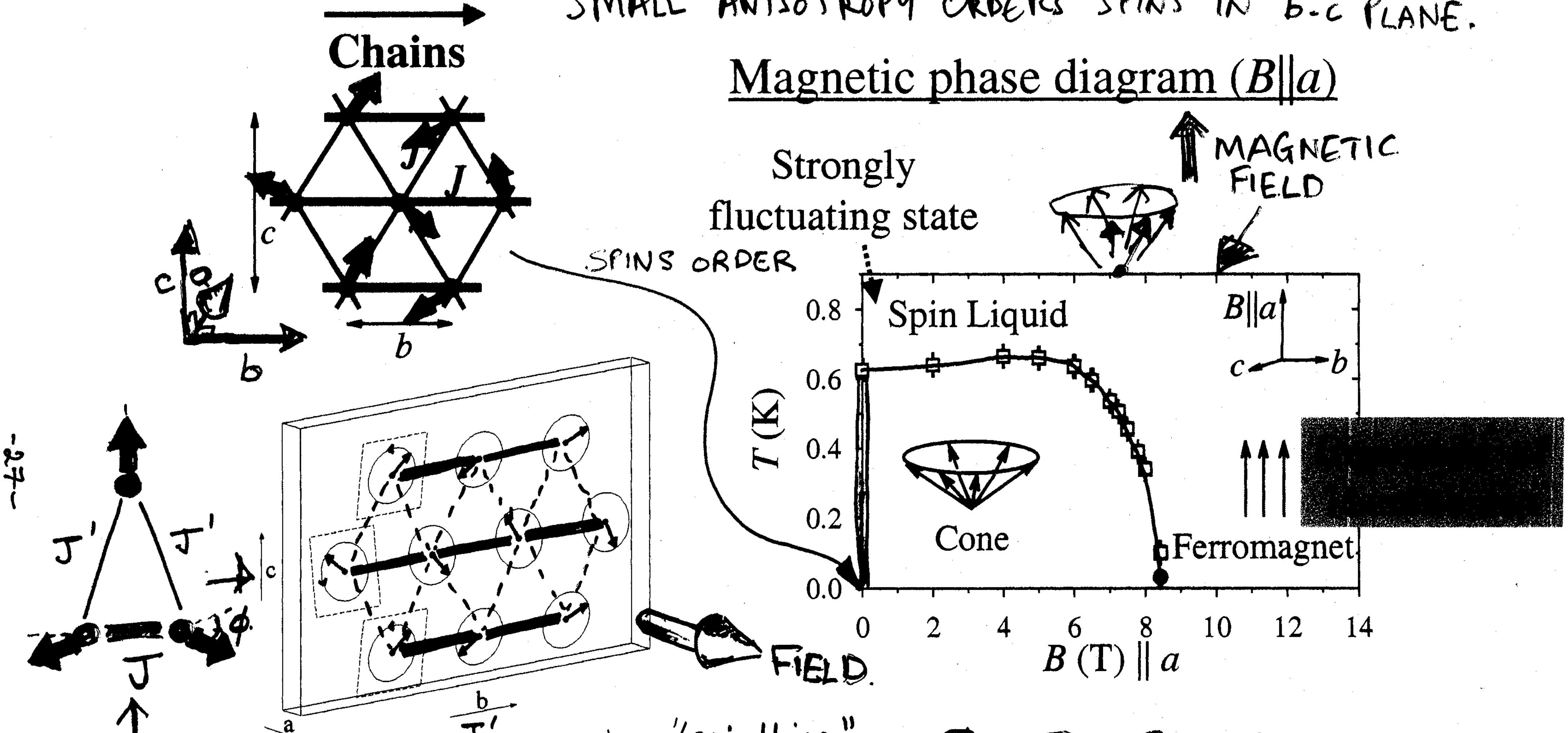
Isoscelese triangle building block

- The magnetism of Cs2CuCl4 is that of layers of spins coupled in an antiferromagnetic isosceles triangular arrangement
- This is a new Hamiltonian which is frustrated and strongly fluctuating.
- Hope is to find new physics and challenging problems for theory!

- Strongly fluctuating system. As for the S=1/2 Heisenberg chain spinwave theory not reliable!
- We don't have any exact results!
- Have the complex phase factor messing up the field theory!
- Can't use Bosonization in 2D!
- We have to use experiment to work out what's going on!
- Follow S=1/2 HAFC and look for distinctive characteristics: continua versus delta functions, renormalization effects, and the effects of a magnetic field.

SMALL ANISOTROPY ORDERS SPINS IN b-c PLANE.

## Magnetic phase diagram (B||a)



[redacted]

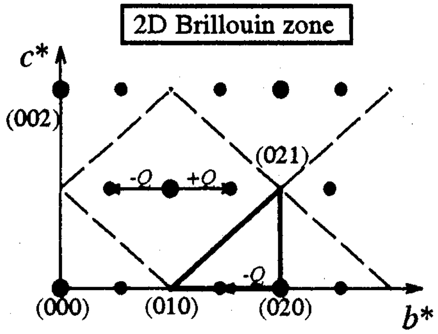
chain spins pick up $J'$ energy by "spiralling" ...

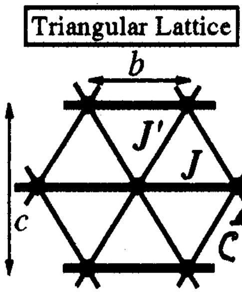
...

Spins rotate in cycloids in zero field.

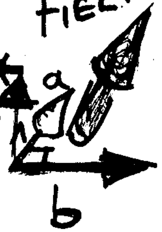
TAS 6, Risoe, Denmark

# 4.2 Quantum renormalization effects

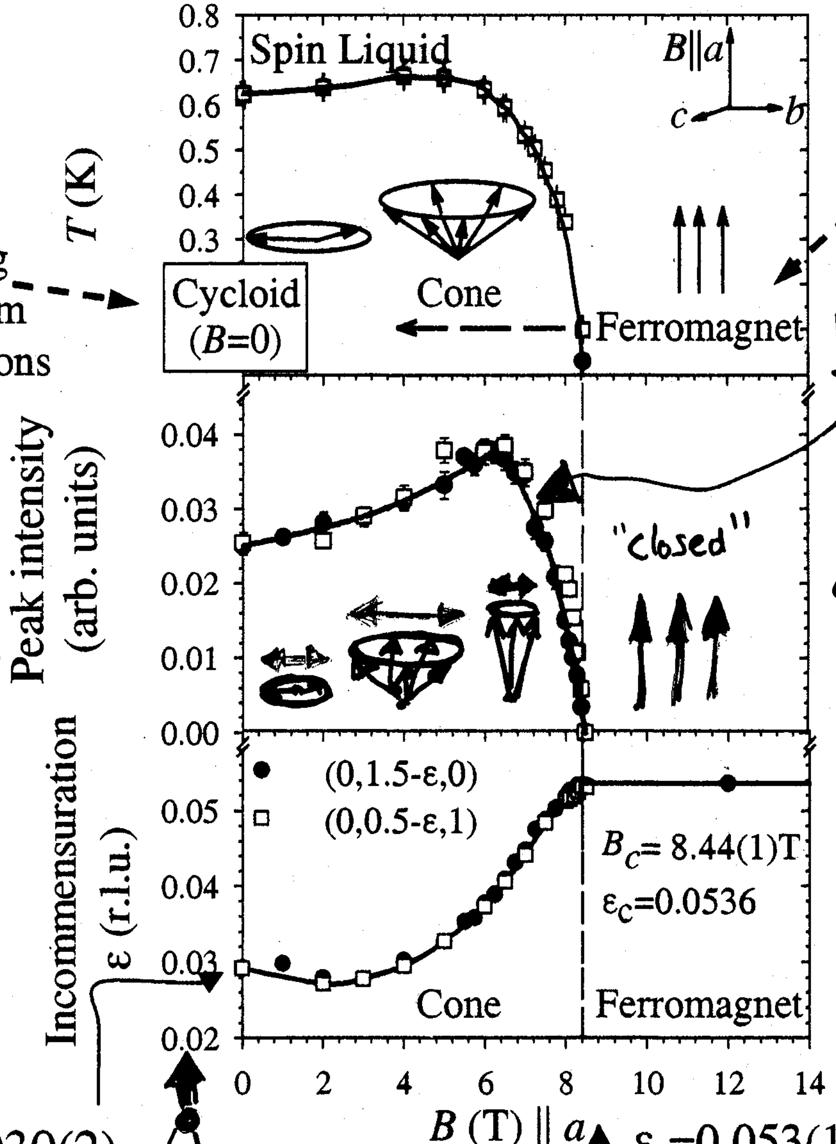




B||a FIELD

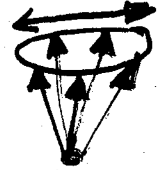


Strong quantum fluctuations



Eigenstate of Hamiltonian (no fluctuations)

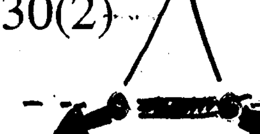
E.G. THIS BRAGG PEAK TELLS US HOW OPEN THE CONE IS



*Shift in intensity and position of Magnetic Bragg peaks tell us precisely how the structure changes with field*

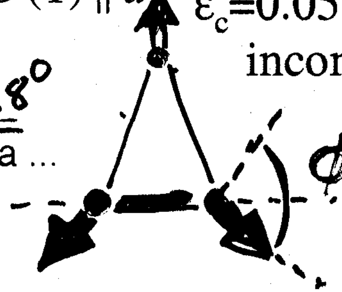
**Quantum renormalization $\varepsilon_0/\varepsilon_c$=0.56**

$\varepsilon_0$=0.030(2)



$\phi_0$=10.8°

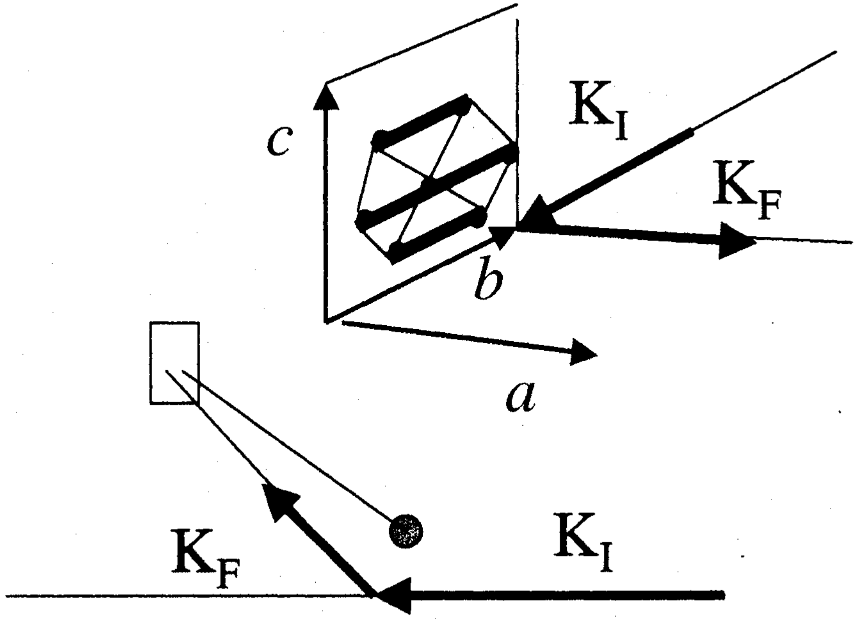
$\varepsilon_c$=0.053(1) "classical" incommensuration

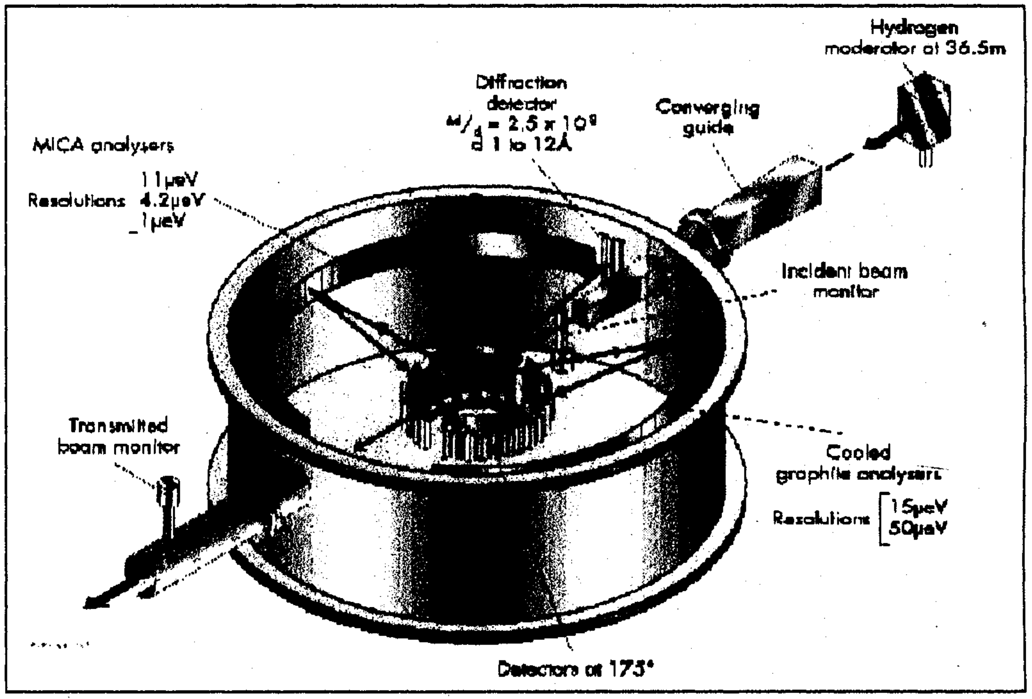


$\phi_c$ = 19.1°

# IRIS neutron spectrometer

Time-of-flight
inverted geometry.
Energy resolution 15 μeV.
Energy transfer -0.2 to
1.6 meV (variable)
Wide angular coverage
25°<2θ<158°

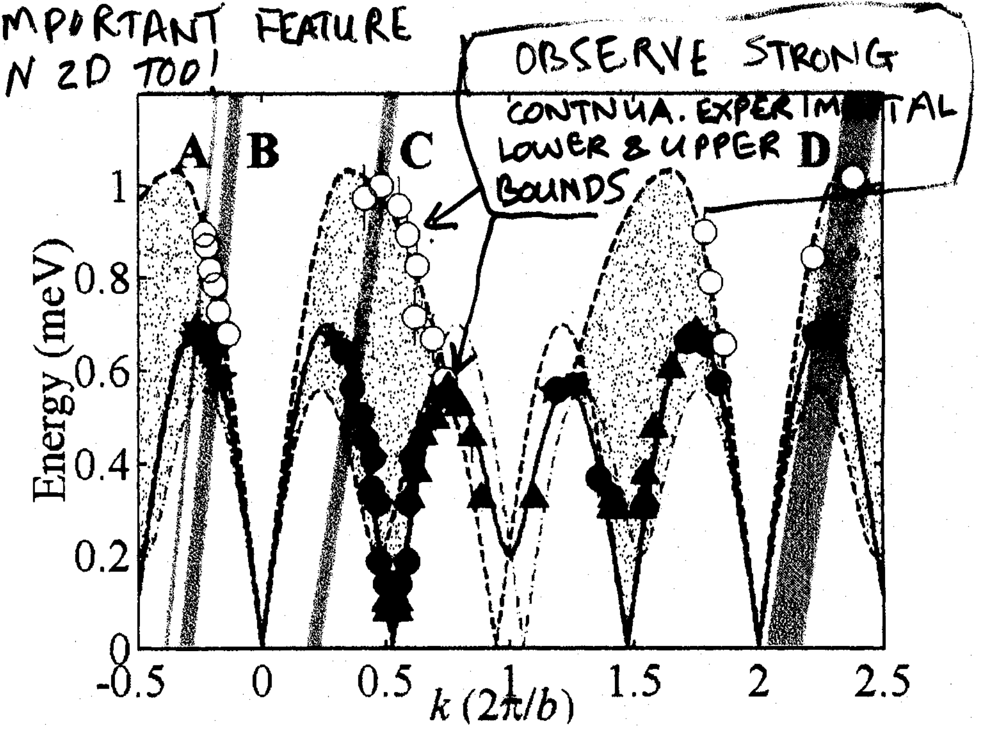




THIS WAS AN IMPORTANT FEATURE
OF 1D. SEE IT IN 2D TOO!

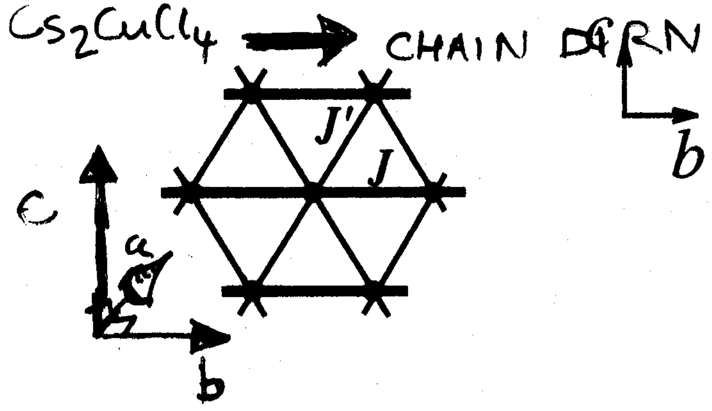
# 4.3 Excitation Continua

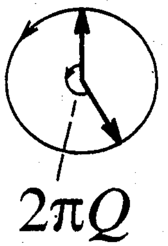
Quantum renormalization of excitation energy
$R = J/J = 1.65$
(1D S=1/2 HAFC $R = \pi/2$)



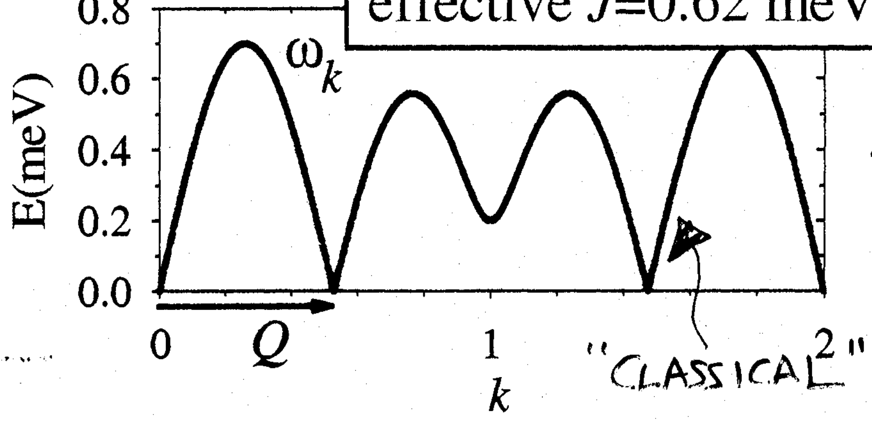
CHAIN DIRN.

## Spin-wave model





effective $J$=0.62 meV



$J$=0.37 meV

$\omega_k$ polarized out-of-plane
$\omega_{k-Q}$, $\omega_{k+Q}$ polarized in-plane

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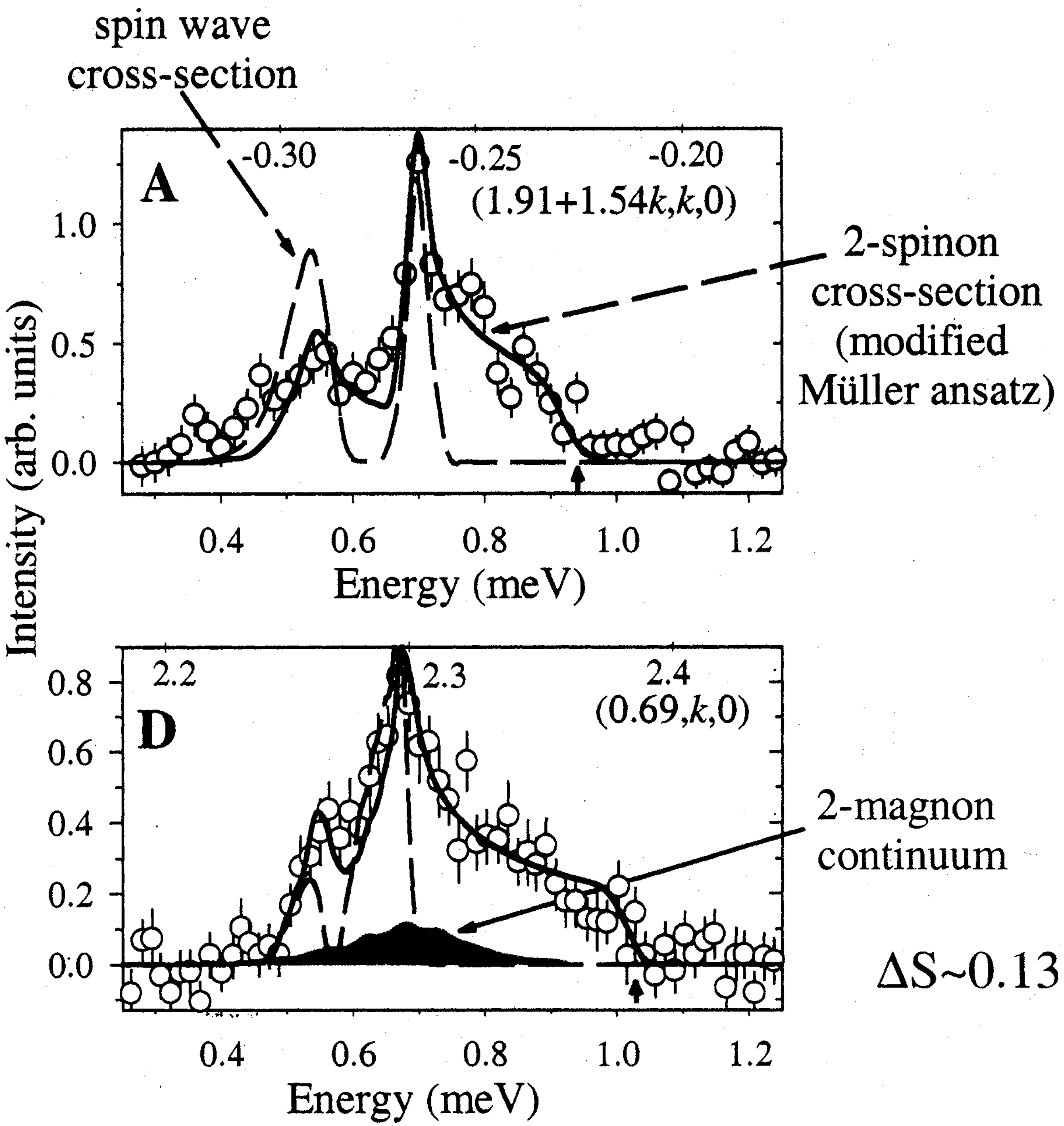
# Lineshapes compared with spinwave theory

NEUTRON PROBES $\Delta S = 0, 1$ SECTOR.

SEE NO WELL DEFINED MODES! ONLY CONTINUA! UNDERLYING EXCITATIONS DO NOT HAVE INTEGER QUANTUM NUMBER OF SPIN. NOT SPINWAVES.



spin wave cross-section

2-spinon cross-section (modified Müller ansatz)

2-magnon continuum

$\Delta S \sim 0.13$

# Model for 2-spinon cross-section

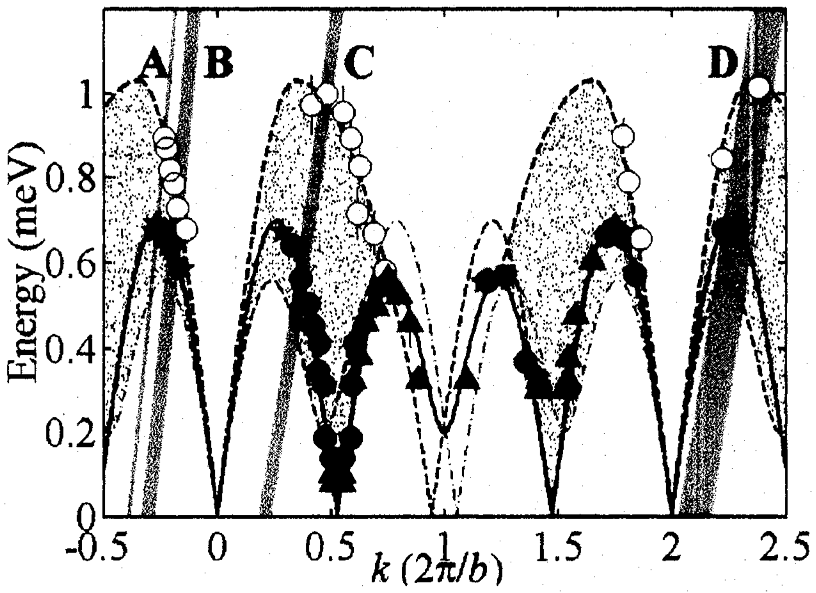
$$S(q,\omega)=\frac{\Theta(\omega-\omega_L)\Theta(\omega_U-\omega)}{\sqrt{\omega^2-\omega^2{}_L}}$$

$\omega_L$ =lower boundary
$\omega_U$ =upper boundary

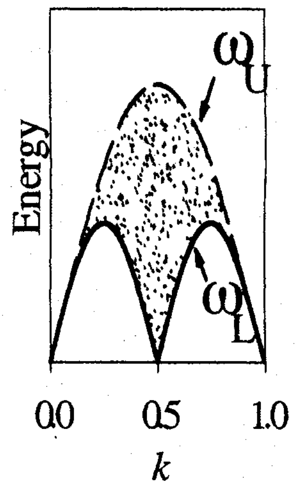
Elementary excitations are S=1/2 spinons with a dispersion relation modified at all energy scales by the 2D frustrated couplings

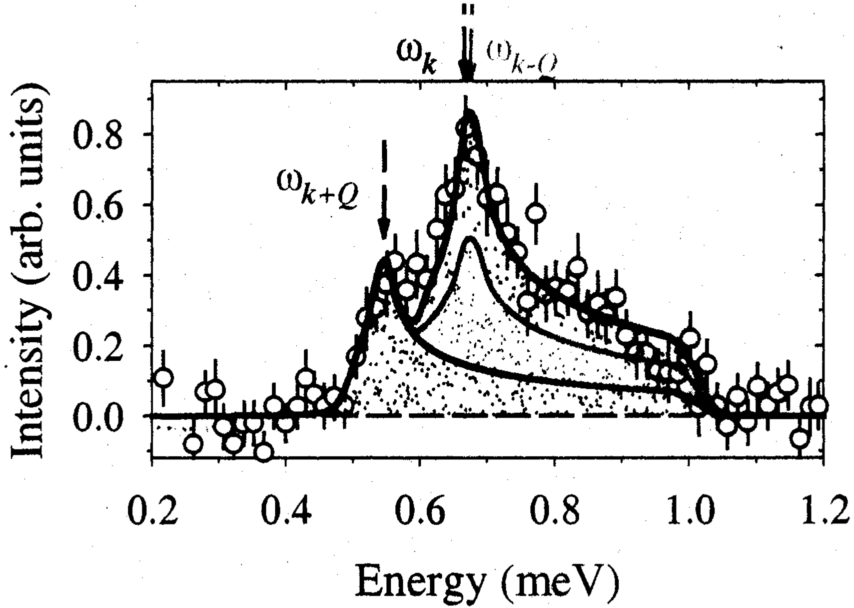
MODIFYING 2 SPINON ANSATZ FROM 1D TO 2D CASE MODELS DATA WELL THROUGHOUT B.Z.

2D quantum magnet $Cs_2CuCl_4$



1D magnet





2-spinon cross-section in $Cs_2CuCl_4$:

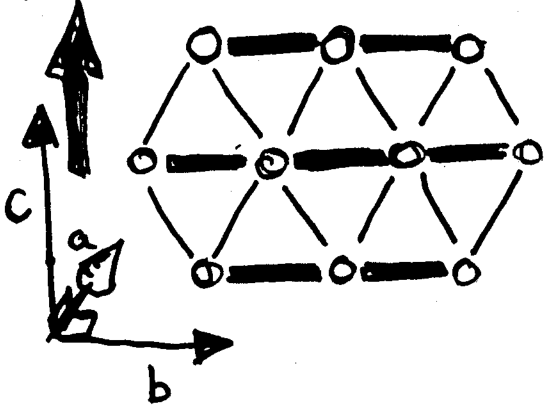
1) 3 overlapping contiuna

2) modified $\omega_U$

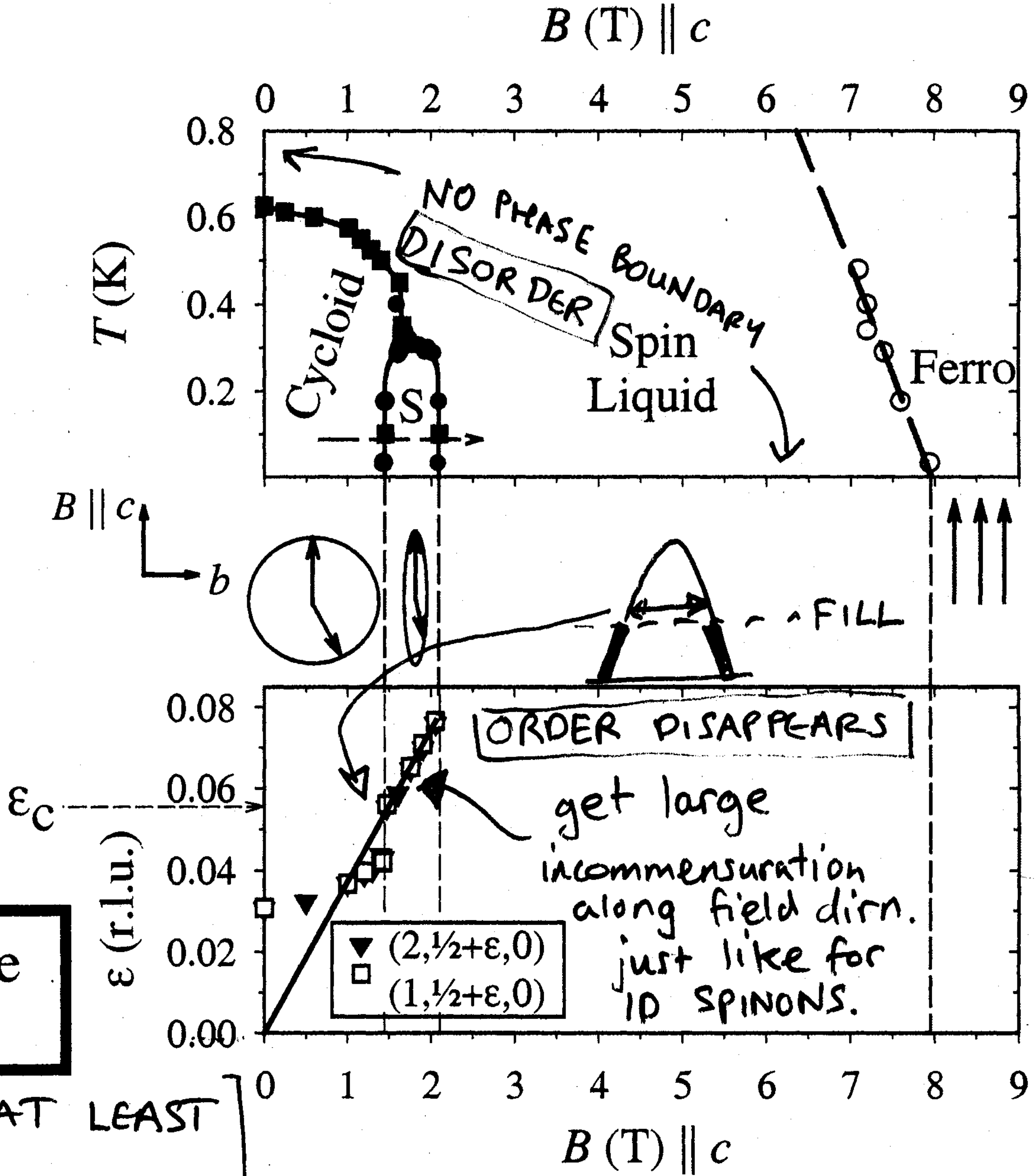
# 4.4 Field effects

Phase diagram for in-plane fields ($B||c$)

FIELD IN PLANE ALONG C



In-plane fields stabilise the spin liquid phase.



NO PHASE BOUNDARY

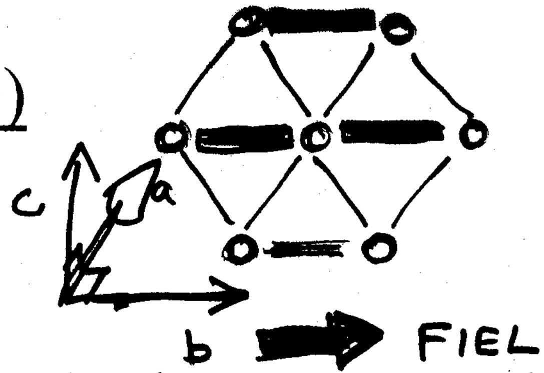
DISORDER

ORDER DISAPPEARS

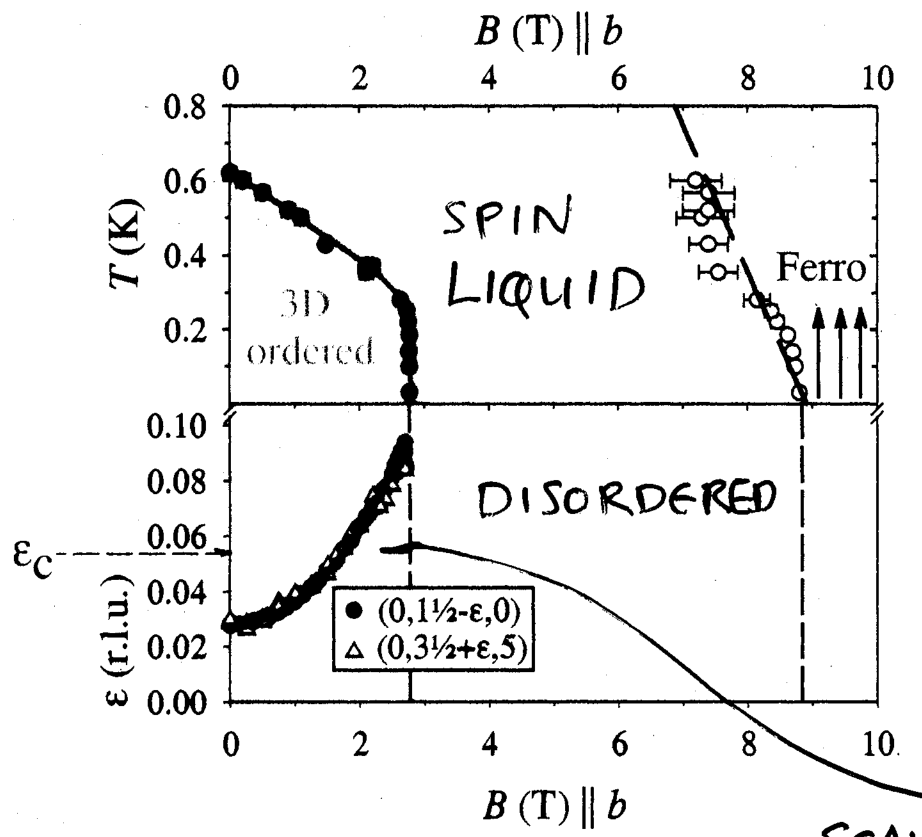
get large incommensuration along field dirn. just like for 1D SPINONS.

NO ORDER TO AT LEAST $\Theta_{CW}/300$

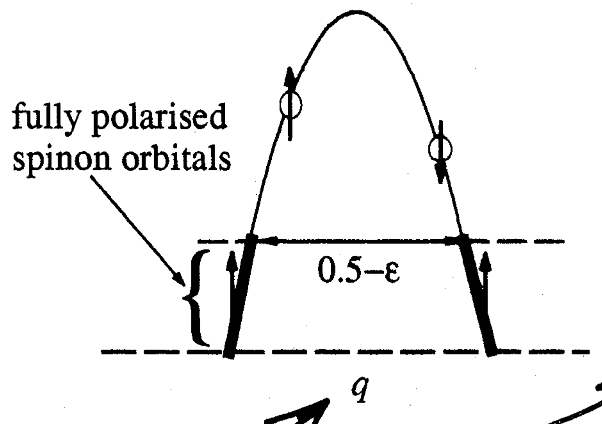
# Phase diagram for in-plane fields ($B||b$)



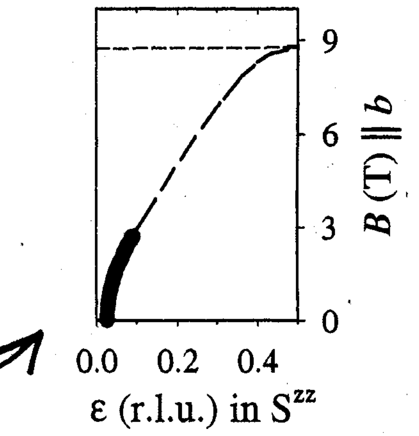
FIELD IN PLANE ALONG 'b'.



Condensation of spinons in field



$Cs_2CuCl_4$

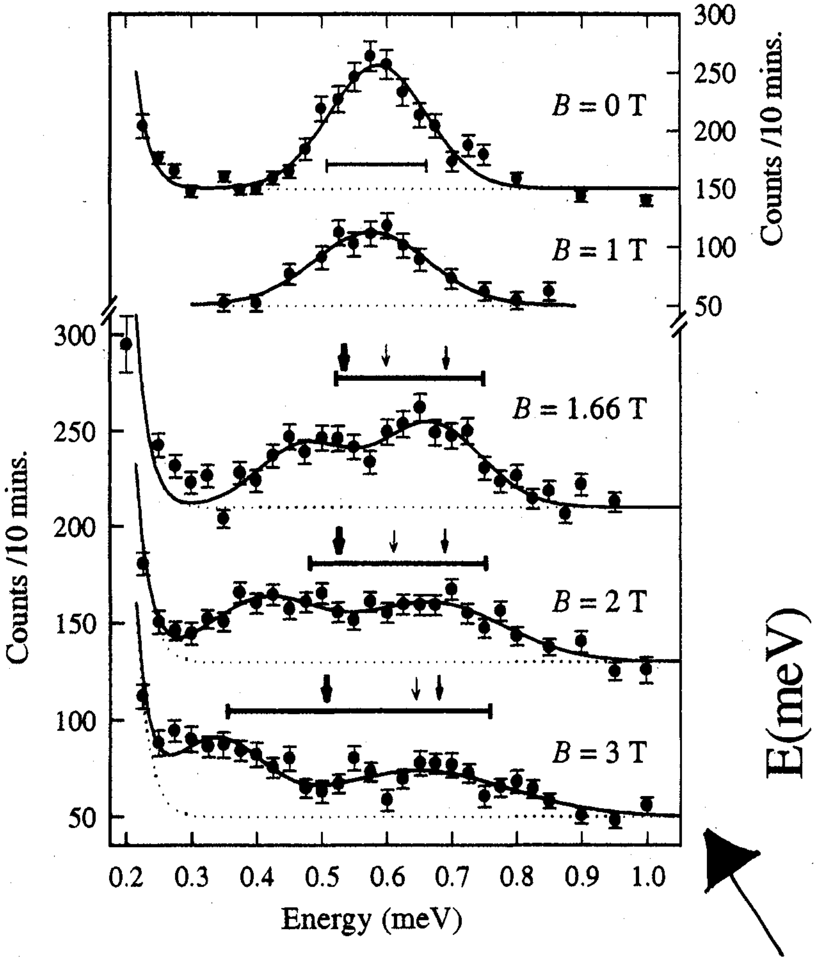


SCALE OF INCOMMENSURATION AGREES WITH FILLING.

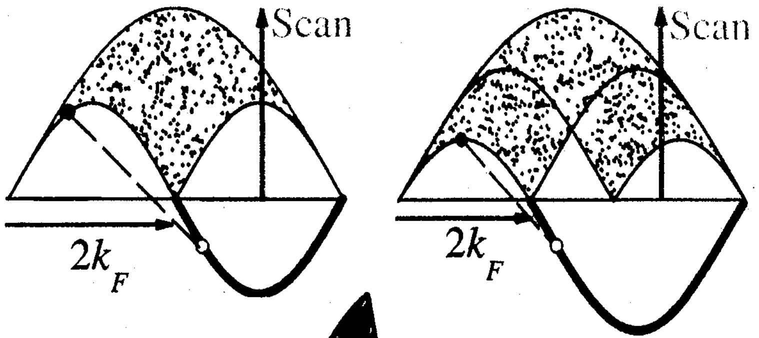
SPIN LIQUID PHASE AGAIN OBSERVED!

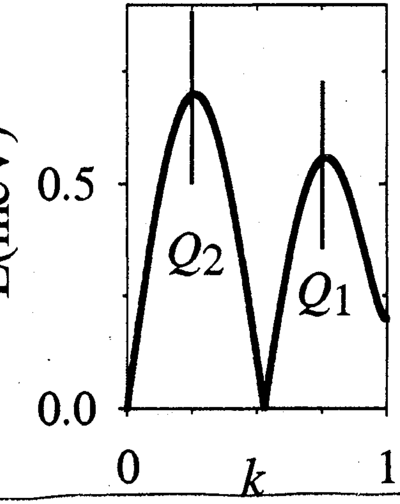
**Spinons in $Cs_2CuCl_4$ follow exclusion statistics**

# Inelastic scattering in applied field ($B||c$)



R.C. et al. ,PRL **79**, 151 (1997).





Large shifts of the inelastic scattering in applied field show that spinons condense by filling up a band of states.

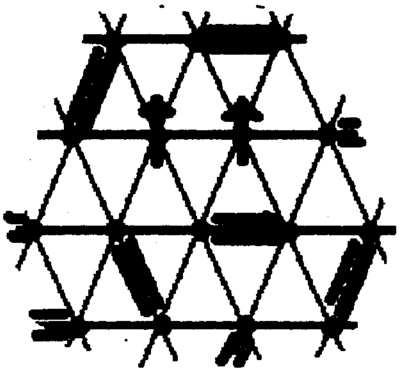
TO TEST THE FILLING OF A BAND HYPOTHESIS LOOK FOR SHIFTS IN INELASTIC CONTINUA (PARTICLE - HOLE) WITH FIELD.

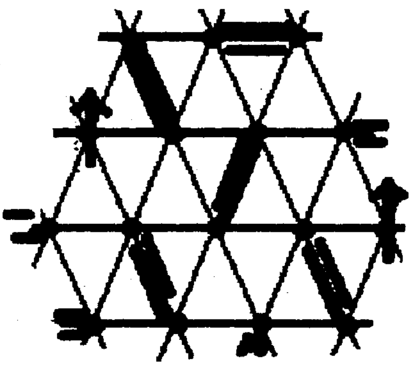
# Evidence of 2D $S=1/2$ spinons in $Cs_2CuCl_4$

1. Excitations are strongly renormalized ($R=1.65$) from classical.
2. Don't see any $S=1$ modes (magnons) only continua. ($S=1/2$ excitations).
3. Continua are ~~the~~ highly dispersive with 2D lower boundary.
4. Clear upper boundary → phase space restrictions.
5. Modified 2-spinon ansatz generalized to 2D describes data well.
6. Fields cause incommensuration effects ~~along~~ for the longitudinal spin component of the scale of filling spinon orbitals.
7. Excitation continua shift in field as expected for $S=1/2$ spinon pairs.

# Resonating Valence Bond Phenomena:-

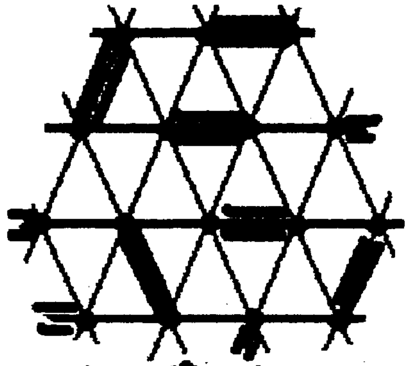
Neutron breaks RVB singlet – creates two spinons.

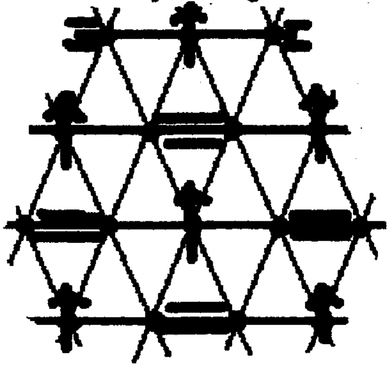




Spinons separate via a rearrangement of singlet bonds.

Zero-field RVB configuration





In-field RVB configuration at 1/3rd filling

**RVB phenomenology provides a physical picture for continuum scattering**

## SUMMARY

1. Now measure excitation continua very accurately.
2. See dimensional crossover to 1D phase in $KCuF_3$.
3. Able to find a spin Hamiltonian using high fields for first time.
4. Exploration of $S=1/2$ triangular magnet revealed: 2D excitation continua.

   Field stabilized spin liquid

   Band filling effects

   Large Quantum Renorms.

   (2D spinons).

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# Next Steps

- Need theoretical explanations of why Cs2CuCl4 behaves this way.
- Why does it show 2D continua?
- Can the quantum renormalizations be calculated?
- Can the phase diagrams be explained fully?
- Can the 1D theory tools be generalized to handle strongly fluctuating 2D systems?
- What other materials are out there?
- What does a 2D spinon look like.