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

PLUS

PRE-TUTORIAL SESSIONS (11 - 13 JULY 2001)

NEUTRON SCATTERING AND LOW DIMENSIONAL ANTIFERROMAGNETS

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These are preliminary lecture notes, intended only for distribution to participants

Neutron Scattering

and

Low Dimensional Antiferromagnets

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Oak Ridge National Laboratory

Outline:

1. Magnetic Neutron scattering
   Experimental methods, Cross sections, correlations, excitations

2. Quasi-one dimensional antiferromagnets
   Ising-like chain, Heisenberg AF chain
   (NENP, $CsCoX_3$, CPC, $KCuF_3$)

3. Interacting chains
   Longitudinal modes in ordered S=1/2 quasi-1D HAF

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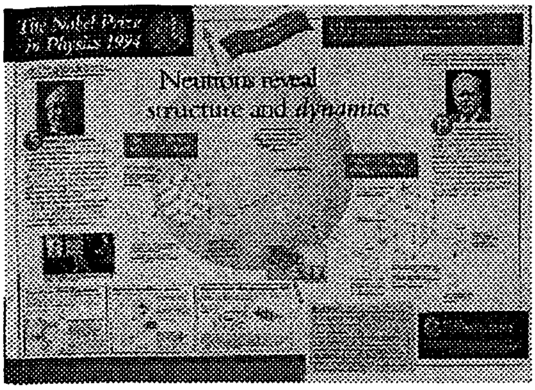

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Elementary properties of the neutron

Energy	$E = \frac{\hbar^2 k^2}{2m}$	$E(\text{meV}) = \frac{81.8}{[\lambda(\text{Å})]^2}$
Wave vector	$k = \frac{2\pi}{\lambda}$	$k(\text{Å}^{-1}) = 0.695\sqrt{E(\text{meV})}$
Neutron magnetic moment		$\mu = -1.91\mu_N \boldsymbol{\sigma}$

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Neutrons as a probe of condensed Matter

Compared to x-rays:
- similar wavelength: structure of materials
- weaker interactions: bulk probe
- nuclear scattering: sensitive to both light and heavy elements
- magnetic moment: sensitive to magnetic structure
- low (meV) energy: collective excitations - phonons, magnons

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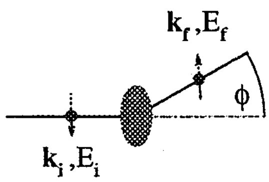
Neutron Scattering



Momentum transfer $\vec{Q} = \vec{k}_i - \vec{k}_f$

Energy transfer $\omega = E_i - E_f$

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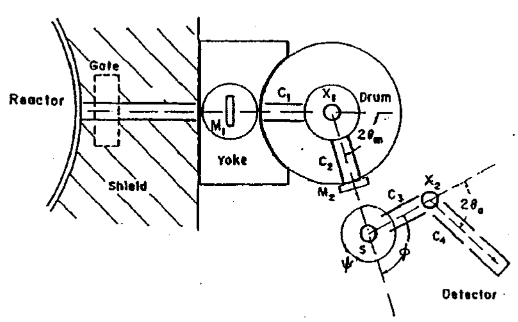
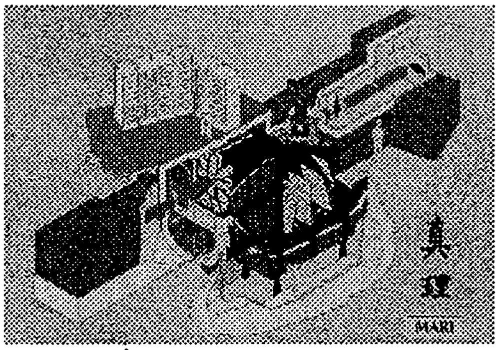
Triple Axis Spectrometer



Triple Axis Spectrometer

MARI chopper spectrometer (ISIS)



Cross-section for magnetic scattering intensity of an *unpolarized* neutron beam:

$$I^{mag}(Q,\omega) \propto |f(Q)|^2 \sum_{\alpha,\beta} \left(\delta^{\alpha\beta} - \hat{Q}^{\alpha}\hat{Q}^{\beta}\right) S^{\alpha\beta}(Q,\omega)$$

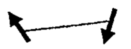
magnetic form factor

components of spin $\alpha,\beta = x,y,z.$

unit vector in direction of Q

magnetic structure factor

Magnetic Structure Factor:

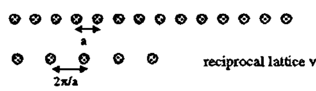


$$S^{\alpha\beta}(Q,\omega) = \int \left\langle m^{\alpha}(0,0) m^{\beta}(\mathbf{r},t) \right\rangle e^{i(Q\cdot\mathbf{r}-\omega t)} d\mathbf{r}dt$$

$$m^{\alpha} = L^{\alpha} + 2S^{\alpha}$$

For magnons usually $S(Q,\omega) \propto \delta(\omega-\omega_Q)$

Scattering from magnetically ordered crystals



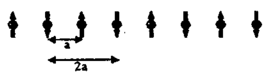
crystal in real space

reciprocal lattice vectors $G_n = n(2\pi/a)$

Scattering measurements show Bragg peaks at G_{hkl}

Ferromagnetic crystals have extra scattering at the same G's.

Antiferromagnets show new, magnetic reciprocal lattice vectors.



Antiferromagnetic crystal in real space

Antiferromagnet in reciprocal space

nuclear Bragg peaks

magnetic Bragg peaks

Neutrons and Antiferromagnetic Structure

C.G. Shull, W.A. Strauser and E.O. Wollan, Phys. Rev. 83, 333 (1951).

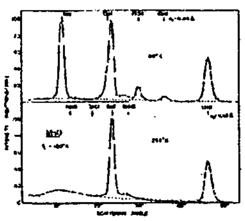


Fig. 4. Neutron diffraction patterns for MnO taken at liquid nitrogen and room temperatures. The patterns have been corrected for the various forms of extraneous, diffuse scattering mentioned in the text. Four extra antiferromagnetic reflections are to be noticed in the low temperature pattern.

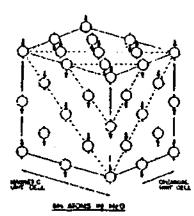
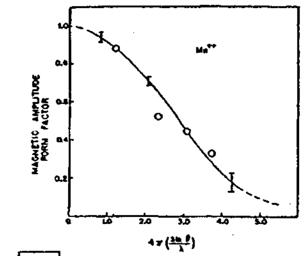


Fig. 5. Antiferromagnetic structure existing in MnO below its Curie temperature of 120°K. The magnetic unit cell has twice the linear dimensions of the chemical unit cell. Only Mn ions are shown in the diagram.

Example of form factor (Shull et al. Phys. Rev. **83**, 333 (1951))



Magnetic amplitude form factor for Mn++ ions. The curve is that obtained from paramagnetic diffuse scattering with estimated error as shown. The points represent values of the form factor obtained from the low temperature antiferromagnetic reflections of MnO.

The intensity of a magnetic bragg peak is proportional to the square of the sublattice magnetization.

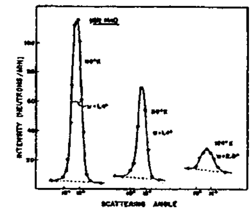
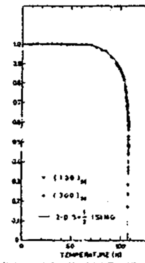


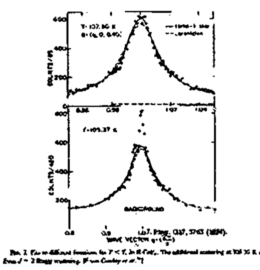
FIG. 6. The (111) antiferromagnetic reflection of MnO as obtained at various temperatures. A noticeable broadening occurs in the vicinity of the Curie temperature.

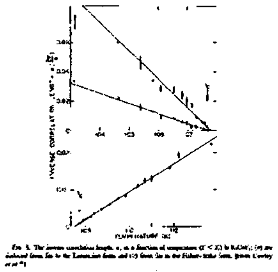


The magnetic correlations in the disordered state give information about short range order

$\langle S^{\alpha}(0)S^{\alpha}(r)\rangle \sim \exp(-\kappa r)$ $\quad S^{\alpha\alpha}(G+q) \sim (q^2+\kappa^2)^{-1}$

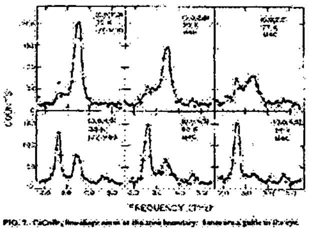
The size of an ordered region is characterized by a correlation length $\xi=\kappa^{-1}$





Inelastic neutron scattering – magnetic or non-magnetic scattering??

	phonon	magnon
type of scattering	nuclear	magnetic
Q dependence of intensity (low T)	$\lvert e\cdot Q\rvert^2$ increases with Q)	$\lvert f(Q)\rvert^2$ (decreases with Q)
T dependence of intensity	$(n\omega+1)$ (increases with T)	disappears for $T > T_C$
polarization	usually non-spin flip	usually spin flip



PRB 27, 1784 (1983).

Further Reading:

Introduction to the Theory of Thermal Neutron Scattering, G.L. Squires, 1978, Cambridge University Press, NY, ISBN 0-521-21884-5.

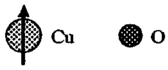
Theory of Neutron Scattering from Condensed Matter, S.W. Lovesey, 1984, Clarendon Press, Oxford, ISBN 0-19-852015-8 (vol. 1), 0-19-852017-4 (vol. 2) - volume 2 is on magnetic scattering.

Magnetic Critical Scattering, M.F. Collins, 1989, Oxford University Press, NY, ISBN 0-19-504600-5.

Methods of Experimental Physics, Volume 23, Neutron Scattering, K. Skold and D.L. Price, ed., 1987, Academic Press, NY, ISBN 0-12-475965-3 (part A), 0-12-475969-6 (part B), 0-12-475968-8 (part C) - chapters 19 and 20 (part C) are specialized to magnetic scattering

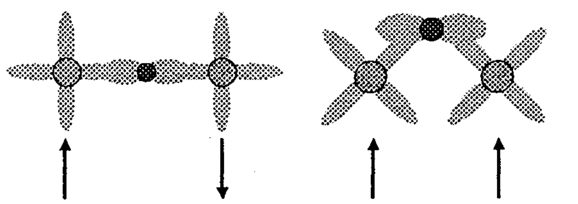
Magnetic Exchange interactions: $JS_1 \cdot S_2$

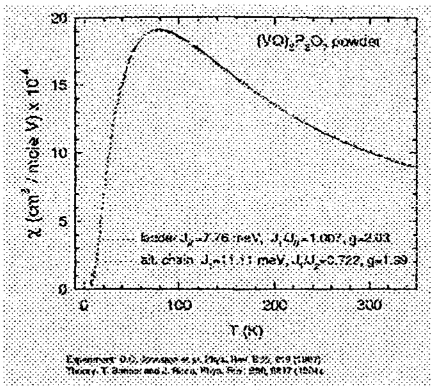
Example:



J > 0, strong antiferromagnetic

J < 0, weak ferromagnetic



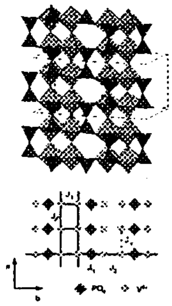


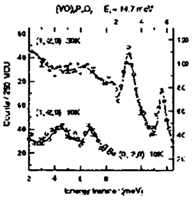
Inelastic Scattering - magnons in $(VO)_2P_2O_7$

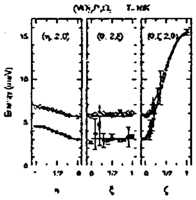
Effective Hamiltonian:

$$\hat{H} = \sum_{i,j} J_{ij} \vec{S}(r_i) \cdot \vec{S}(r_j)$$

Determine J_{ij} from $\omega(Q)$







PRL 78, 4998 (1997) I PRL 79, 745 (1997)

Simple Example: Antiferromagnetic Spin Dimer, S=1/2

Hamiltonian: $JS_1 \cdot S_2$

↑ ← R → ↓

$|\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle, \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$

↑ J ↓

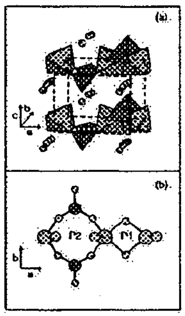
$\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$

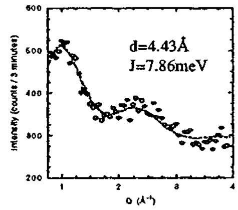
$$S^{\alpha\alpha}(\vec{Q},\omega) = \sin^2\left(\frac{\vec{Q}\cdot\vec{R}}{2}\right)\delta(\omega - J)$$

(for T=0)

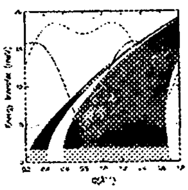
example simple spin dimer material $VODPO_4 \cdot 1/2D_2O$ (powder)

$$S^{\alpha\alpha}(Q,\omega) = \left(1 - \frac{\sin(QR)}{QR}\right)\delta(\omega - J)$$

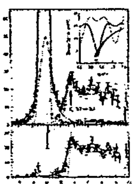


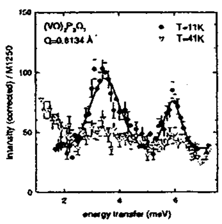


See D.A. Tennant et al., PRL 78, 4998 (1997)



$(VO)_2P_2O_7$ powder
R. Eccleston et al.
PRL 73, 2626 (1994)
ISIS - HET

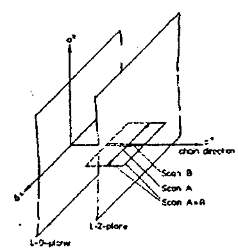




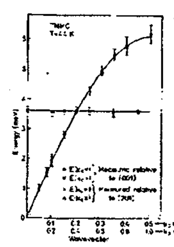
A.W. Garrett et al.
PRB 55, 3631 (1997)
HFIR - HB1A

Caution: Beware of data on powders, and of pretty color pictures!!

- Quasi - 1D systems have sheets in reciprocal space
- excitations vary only along directions parallel to chains



TMMC S=5/2 HAC



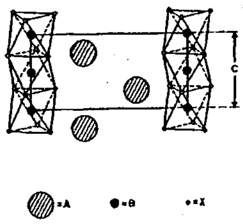
M.T. Hutchings et al. PRB 5, 1999 (1972)

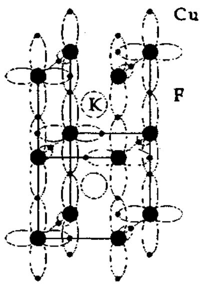
Examples of quasi-one dimensional magnetic materials

hexagonal ABX_3 salts

e.g. $RbCoCl_3$, $CsMnBr_3$, etc.

almost cubic: $KCuF_3$





Real world effective Hamiltonians:

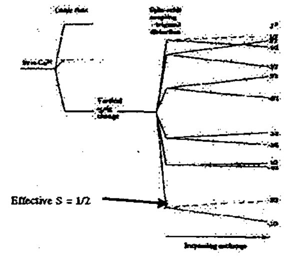


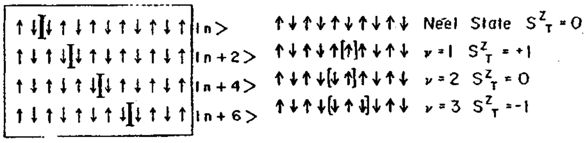
FIG. 1. Schematic diagram of the crystal-field levels of the Co^{2+} ion in $CsCoCl_3$ (Ref. 19). Spin-orbit coupling gives rise to a mixing of levels with the same $J'=L'+T'$.

Co^{2+} is S=3/2, with effective L=1 in lowest cubic manifold. Projecting S•S into lowest lying Kramer's doublet gives an effective spin Hamiltonian with S=1/2, and anisotropic (nearly Ising) exchange.

$CsCoCl_3$ and $CsCoBr_3$ are examples of nearly Ising (Ising-Like) antiferromagnetic chains with S = 1/2

Ising Antiferromagnetic Chain

$$\hat{H} = 2J\sum_r S_r^z \cdot S_{r+1}^z$$



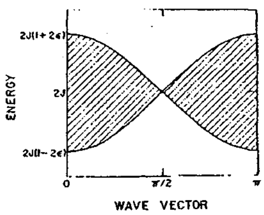
localized excitation with ω = 2J

$S^{+}(Q,\omega) \sim \delta(\omega-2J)$

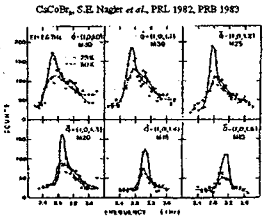
Ising-like antiferromagnetic chain

$$\hat{H} = 2J\sum_r \left\{ S_r^z \cdot S_{r+1}^z + \varepsilon\left[S_r^x \cdot S_{r+1}^x + S_r^y \cdot S_{r+1}^y \right] \right\}$$

Examples: $RCoX_3$, R=Cs,Rb X=Cl,Br ε ~ 0.1



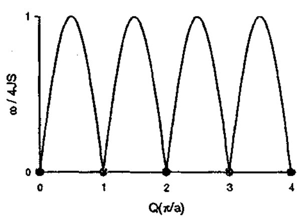
$CsCoBr_3$, S.E. Nagler *et al.*, PRL 1982, PRB 1983



Heisenberg Antiferromagnetic Chain

$$\hat{H} = 2J\sum_r \vec{S}_r \cdot \vec{S}_{r+1}$$

- ground state has $S_T = 0$
- LSW: ω = 4JS|sinQ|
- S=1/2: ω = πJ|sinQ|



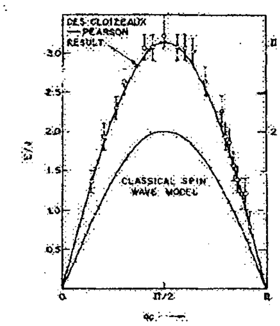


FIG. 3. Dispersion of the excitations in CPC at T = 1.3 K with energy in units of J = 13.4 K.

Historically: Main quantum effect was thought to be a renormalization of the spin wave dispersion by a factor of π/2.

$CuCl_2 \cdot 2N(C_5D_5)$ or CPC

S=1/2 chain

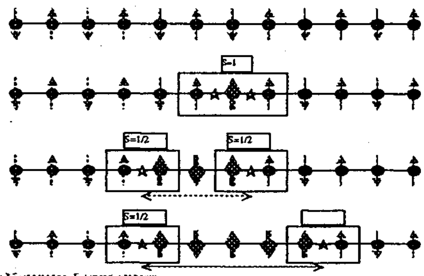
Y. Endoh *et al.* PRL 32, 170 (1974).

Quick facts about the antiferromagnetic Heisenberg chain:

- Ground state is a singlet
- The natural excitations are "spinons" and carry spin 1/2 relative to the ground state unlike "magnons" or "spin-waves" which have spin 1 relative to the ground state
- spinons are created only in pairs (or even numbers)
- physically observed states have S_T=1
- for S=1,2,3, ... spinons are bound - result is an energy gap (Haldane gap)
- for S=1/2, 3/2, 5/2, ... spinons are "free" - S(Q,ω) has a continuum

One way to picture a spinon pair:

recall: $$S_r^x S_{r+1}^x + S_r^y S_{r+1}^y = \frac{1}{2}\left(S_r^+ S_{r+1}^- + S_r^- S_{r+1}^+\right)$$



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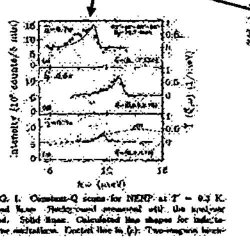
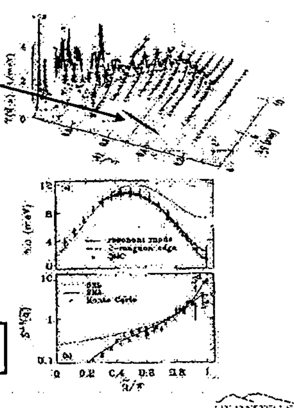
Caution: Experimental resolution affects lineshapes!!





Magnons in S=1 material NENP

S. Ma *et al.* PRL 69, 3571 (1992)

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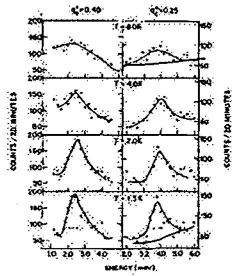
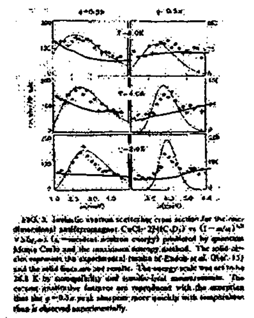


FIG. 2. Variation with temperature of scans at q_c = 0.40 and 0.25. Solid lines, guides for the eye. The arrows at T = 1.3 K give the instrumental width.

$CuCl_2 \cdot 2N(C_5D_5)$ or CPC – S=1/2 chain

Y. Endoh *et al.* PRL 32, 170 (1974), I. U. Heilman *et al.* PRB 18, 3530 (1978)



Caution: Know the difference between signal and background!!

J. Deisz *et al.* PRB 42, 4869 (1990).

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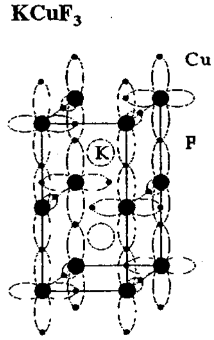
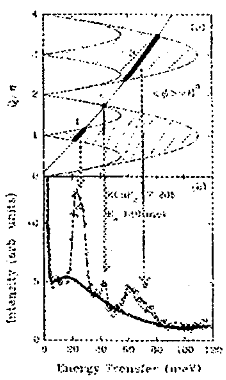
$KCuF_3$



S.E. Nagler *et al.*, PRB 44, 12361 (1991).



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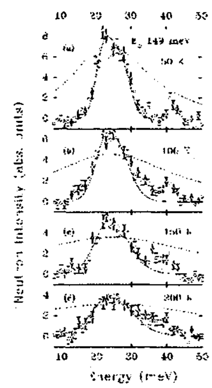

- The temperature dependence of the scattering is in excellent agreement with predictions of field theory (H.J. Schulz, PRB 34, 6372, (1986)).
- Classical theory (H.H. Kretzen *et al.*, Z. Phys. 271, 269 (1974)) underestimates the linewidth at T=0 and overestimates the thermal broadening.

$KCuF_3$, D.A. Tennant *et al.*, PRL 70, 4003 (1993).

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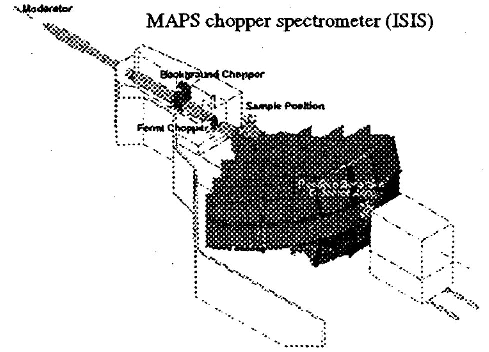
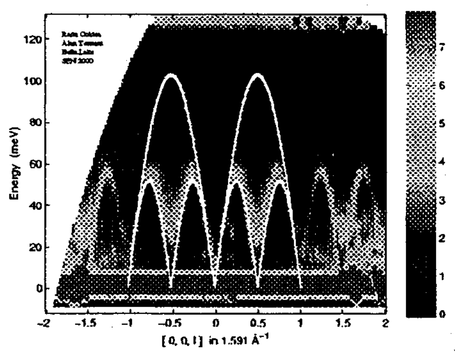
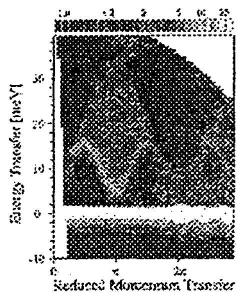
MAPS chopper spectrometer (ISIS)



$KCuF_3$ on MAPS $E_o = 150$ meV T = 6K



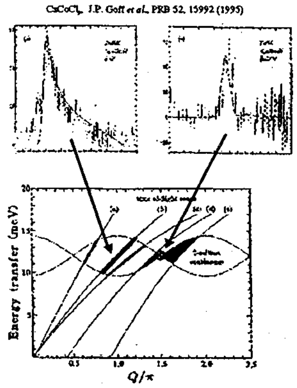


The spectrum of a weakly alternating chain is overall very similar to that of a pure Heisenberg chain.

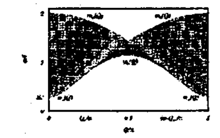
e.g. $CuGeO_3$ (spin Peierls)

M. Arai et al., PRL 77, 3649-3652 (1996)

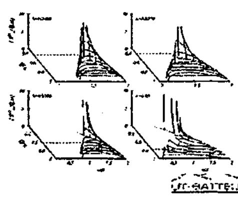
$CsCoCl_3$ J.P. Goff et al., PRB 52, 15992 (1995)



Modified spinon states can also account for the spectrum of the Ising-like AF.



A.H. Bougourzi et al., PRB 57, 11429 (1998)



Pure 1D S=1/2 chain is relatively well understood.

- "free" spinon pair (even multiplet) excitations
- continuum of S=1 states – triply degenerate

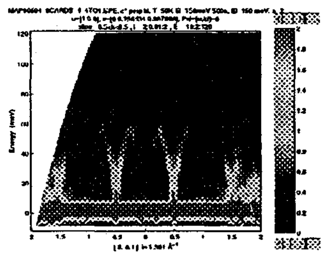
Ordered 3 dimensional systems also well understood.

- doubly degenerate magnons with poles δ(ω–ω(q)) in $S^T(Q,\omega)$ (transverse)
- Goldstone modes when system has broken continuous symmetry
- two-magnon scattering appears in $S^L(Q,\omega)$ (longitudinal)

What about "quasi –1D": coupled chains in 3 dimensions ??

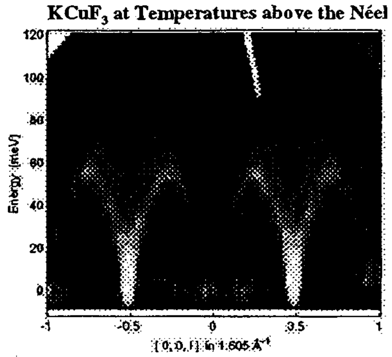
- Large energy scale dynamics must look 1D like
- Low energy scales must show Goldstone modes

$KCuF_3$ at Temperatures above the Néel Temperature



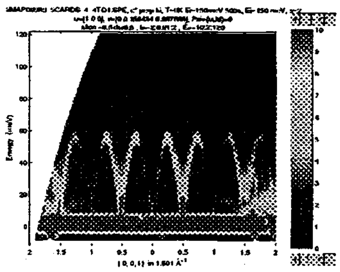
- The 1D characteristics of $KCuF_3$ should dominate at temperatures above the Néel temperature where there is no long-range order.
- There is a continuum of excitation
- The scattering weight is concentrated around the lower edge of the continuum but does not appear to be well-defined.

$KCuF_3$ at Temperatures above the Néel Temperature



The non-magnetic background has been subtracted

$KCuF_3$ at Temperatures below the Néel Temperature



- 3D characteristics should emerge at temperatures below the Néel temperature where $KCuF_3$ develops long-range antiferromagnetic order.
- We still see a continuum!
- The scattering at the lower edge of the continuum is sharpened.

Hamiltonian: $\hat{H} = J\sum_{n,\vec{r}}\left(\vec{S}_{n,\vec{r}}\cdot\vec{S}_{n+1,\vec{r}}\right) + J_\perp\sum_{n,\vec{r},\vec{\delta}}\left(\vec{S}_{n,\vec{r}}\cdot\vec{S}_{n+1,\vec{r}+\vec{\delta}}\right)$

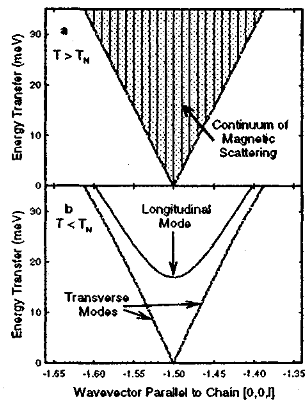
Bosonization (field theory) + RPA predicts:

- non-zero staggered magnetization m_0

Poles in S(Q,ω) near ordering wavevector comprising:

- gapless transverse modes (spin waves / Goldstone modes)
- novel gapped longitudinal mode (zero point fluctuations)

Free spinon continuum resumes at high energies



The extra longitudinal mode violates the common "folklore" concerning excitations in spin systems. It is a quantum effect.

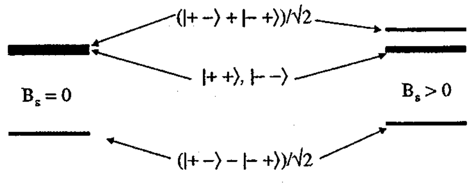
From A. Bunker and D.P. Landau, *Longitudinal Magnetic Excitations in Classical Spin Systems*, PRL 85, 2601 (2000):

"...for all classical Heisenberg models the longitudinal propagative excitations are entirely multiple spin wave in nature."

How do we understand this intuitively??

Consider S=1/2 spin dimer with staggered field:

$$JS_1\cdot S_2 + B_s(S_1^z - S_2^z)$$



Hamiltonian: $\hat{H} = J\sum_{n,\vec{r}}\left(\vec{S}_{n,\vec{r}}\cdot\vec{S}_{n+1,\vec{r}}\right) + J_{\perp}\sum_{n,\vec{r},\vec{a}}\left(\vec{S}_{n,\vec{r}}\cdot\vec{S}_{n+1,\vec{r}+\vec{a}}\right)$

Bosonization (field theory) + RPA leads to:

- staggered magnetization $m_0 \approx 1.02\left(\frac{|J_{\perp}|}{J}\right)^{1/2}$
- Mass gap $M = \omega(\pi,0,\pi) = 6.2\,|J_{\perp}|$

This gives poles in $S(Q,\omega)$ near $Q_z=\pi$ ($q=Q_z-\pi$)

- Transverse $\omega^2 = \left(\frac{\pi J}{2}\right)^2 q^2 + M^2\left(1-\frac{\cos k_x + \cos k_y}{2}\right)$
- Longitudinal $\omega^2 = \left(\frac{\pi J}{2}\right)^2 q^2 + M^2\left(3-\frac{\gamma}{2}\left(\cos k_x + \cos k_y\right)\right)$

$\gamma = 0.49$

Continuum resumes for energies $\omega \geq 2M$

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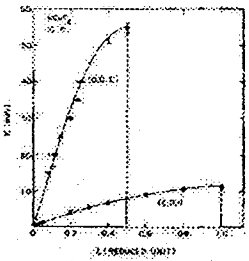

S.K. Satija et al, PRB 21, 2001 (1980)

Dispersion of magnetic excitations in $KCuF_3$ showed that the ratio $J_a/J_c \approx 0.01$.

We also have $M \approx 10$ meV

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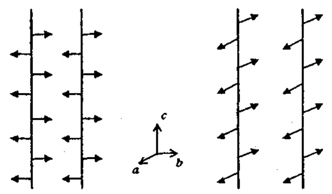
$KCuF_3$

chains in *c* direction

- intra-chain coupling is antiferromagnetic
- inter-chain coupling is ferromagnetic

ordered structure below $T_N \sim 38K$

- moments lie in *a-b* plane - anisotropy from DM interaction



b domain *a* domain

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Magnetic scattering intensity for unpolarized neutrons:

$$I \sim \sum_{\alpha}\left(1-\left(\hat{Q}^{\alpha}\right)^2\right)S^{\alpha\alpha}(Q,\omega)$$

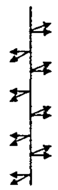
Zero Field: equal populations of *a* and *b* domains

for Q making an angle ϕ with the *c* (chain) direction:

- $I \sim [1-\cos^2\phi]S^{cc} + [1-\sin^2\phi](S^{aa}+S^{bb})$

S^{cc} is transverse (T), S^{aa} and S^{bb} are ½ T, ½ longitudinal (L)

- $I \sim I^T + (\cos^2\phi)I^L$



For Q along the chain direction:

- $I \sim I^T + I^L$ (equal contributions)

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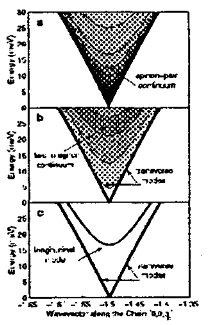

(a) Expected continuum scattering in disordered (one dimensional) phase.

(b) Predicted scattering in ordered phase from multi spin wave theory. The longitudinal scattering has a broad peak near 23 meV.

(c) Predicted excitations in the ordered phase from RPA/field theory (H. Schulz, PRL 1996, F. Essler, A. Tsvelik and G. Delfino, PRB 1997)

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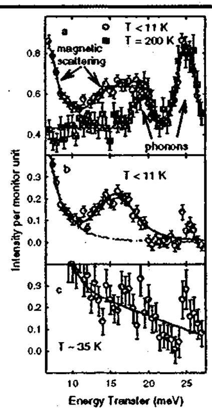

constant Q = (0,0,1.5)

- 200K: weak magnetic scattering may persist and phonon modes are evident
- 35K: the magnetic scattering decays monotonically with energy transfer
- 11K: magnetic scattering has broad decaying contribution plus a lump of scattering near 16 meV. This mode is significantly broader than resolution.

B. Lake, D.A. Tennant and S.E. Nagler, Phys. Rev. Lett 85, 832 (2000).

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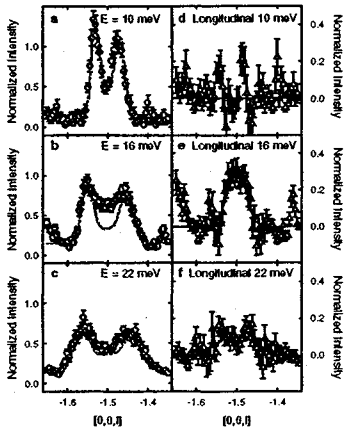
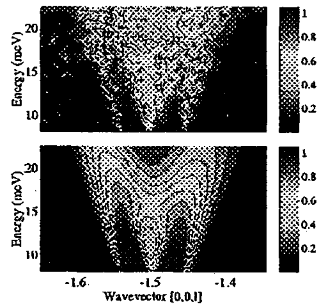
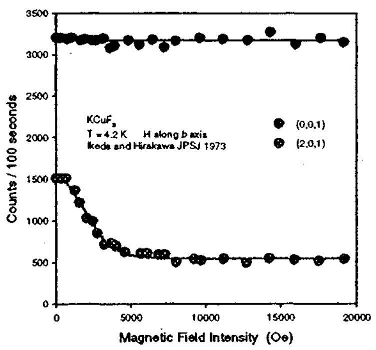


•Constant E scans show that the extra scattering is not an artifact of resolution.

•The red lines are a convolution of the transverse cross-section with instrumental resolution.



Low temperature data from a large set of scans. The upper panel shows the data, lower panel is a simulated intensity distribution including the instrumental resolution



Note: reciprocal lattice definition differs by factor of two from our convention

Applied field along the b axis of magnitude 0.5 T causes the spins to align along the a axis.

Magnetic field along *b* direction:

spin flop phase with moments directed along *a*

•S^{aa} is L S^{bb} and S^{cc} are T

Unpolarized neutrons, Q along *c*, measure $S^{aa} + S^{bb}$

•$I = I^T + I^L$

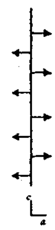
Polarized neutrons with axis of neutron polarization *b*:

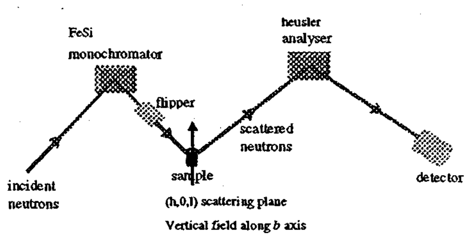
•S^{aa} is spin-flip, S^{bb} is non-spin-flip

Therefore, for Q along *c*:

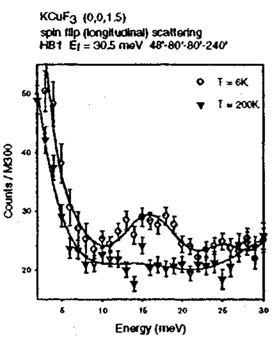
•Longitudinal magnetic scattering is spin-flip

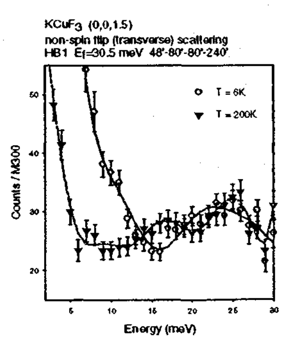
•Transverse magnetic scattering is non-spin-flip





Cartoon of HB1 spectrometer at ORNL





Some Conclusions:

➤Quasi – 1D antiferromagnets exist in nature.

➤The main theoretical predictions for Heisenberg AF chains (Haldane gap, free spinons, etc.) are confirmed by inelastic neutron scattering experiments on model materials.

➤In the 3D ordered state of a quasi-1D HAFC:

- The low-energy transverse modes are sharp spin-waves. (Goldstone modes).
- The longitudinal response contains a gapped mode, possibly broadened by collisions with spin waves.
- Polarized neutron experiments confirm the nature of the fluctuations.

Acknowledgement:

A word of thanks to my 1D collaborators over the years:

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