Emergent Excitations in Geometrically Frustrated Magnets

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- Issues on geometrical frustrated insulating and metallic magnets
  - What is the nature of the spin liquid phase?
  - Any novel phase transition?
  - What is the effect of GF in the metallic systems?
- Neutron scattering studies on insulating and metallic spinels AB_2O_4
  - $ZnCr_2O_4$ and $Li_{1-x}Zn_xV_2O_4$
- Future studies…

Collaborators:

Experiments:

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S. Park (NIST)
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Theory:

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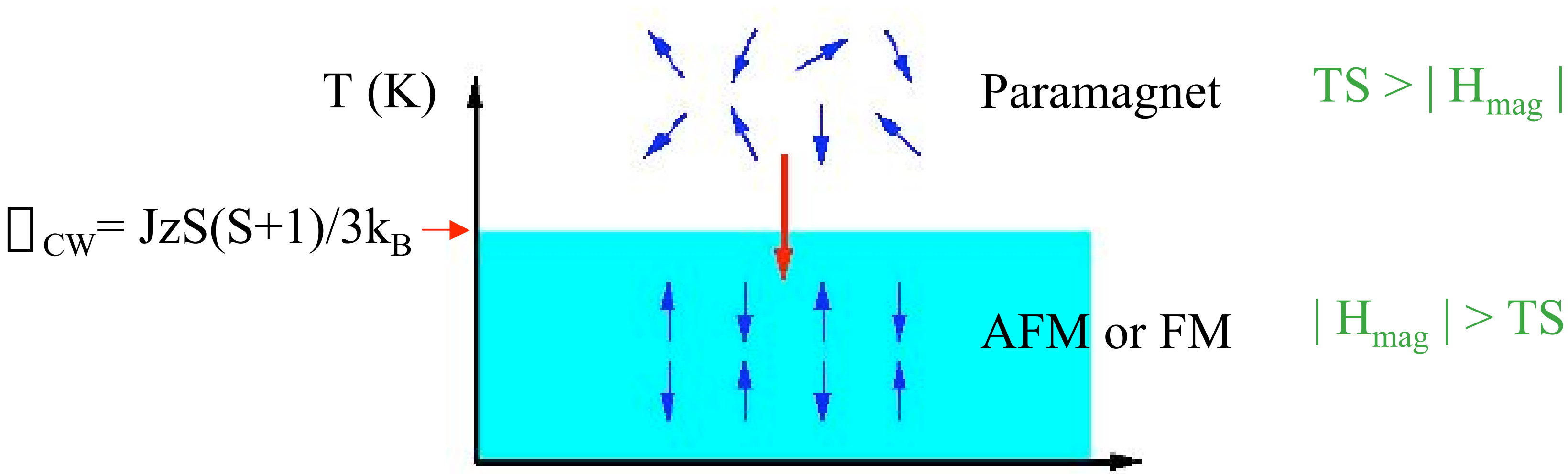
Phase Diagram for an Ordinary Magnet

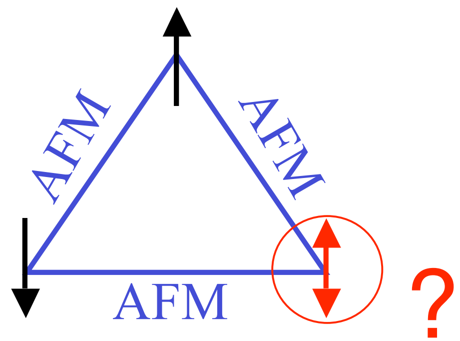
$$F = H_{mag} - TS = -\Sigma J S_i S_j - TS$$



Ground State : Long range (anti)ferromagnetically ordered state.
Low Energy Excitations : linear spin waves around the ordered state.

Geometrical Frustration

A simplest example: a Triangle of three antiferromagnetic Ising spins

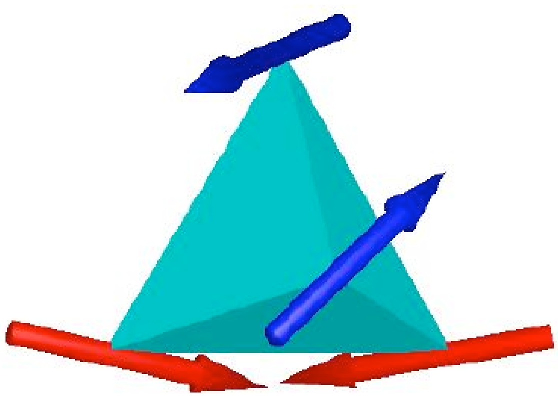


E=-|J|

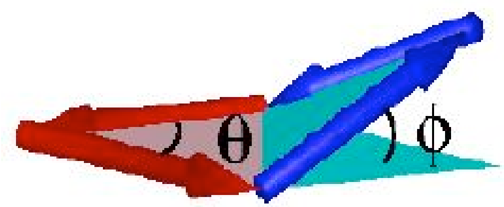
$$H = -J \sum \mathbf{S_i} \cdot \mathbf{S_j}$$

All exchange interactions can not be satisfied.

A tetrahedron with four Heisenberg spins



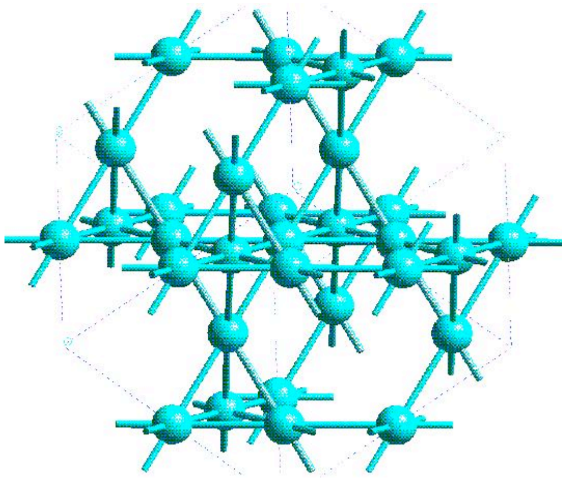
$\sum \mathbf{S_i} = \mathbf{0}$



Zero energy modes
in the ground state manifold

Geometrical frustration leads to a large degeneracy in the ground state

Theory of spins with AFM interactions on corner-sharing tetrahedra



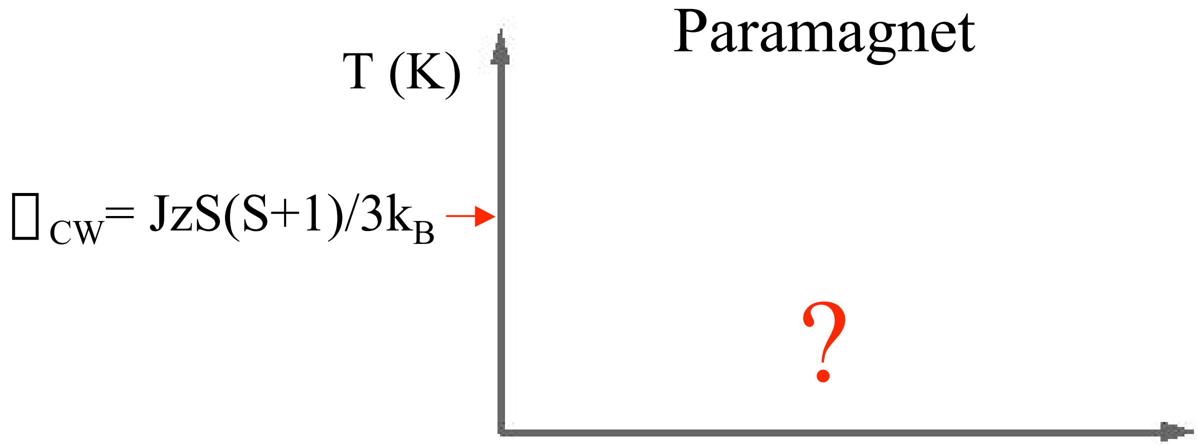
The simplest and most frustrated case is a system with isotropic uniform nearest neighbor interactions only:

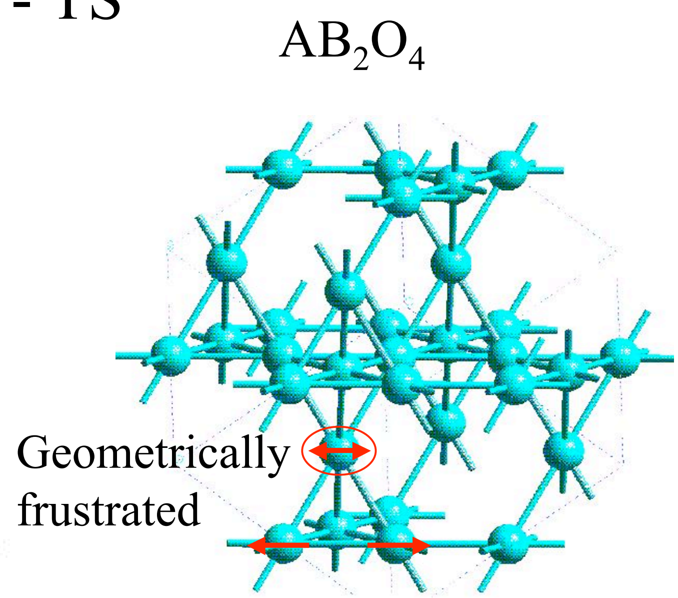
$$H = -J \sum \mathbf{S}_i \cdot \mathbf{S}_j$$

SPIN TYPE	SPIN Value	LOW T PHASE	METHOD	REFERENCE
Isotropic	S=1/2	**Spin Liquid**	Exact Diag.	Canals and Lacroix PRL '98
Isotropic	$S=\infty$	**Spin Liquid**	MC sim.	Reimers PRB '92 Moessner, Chalker PRL '98

Phase Diagram for GF Insulating Magnets

$$F = H_{mag} - TS = -\sum J\, S_i\, S_j - TS$$





- **Nature of the Spin Liquid State**

  How are the fluctuating spins in the SL phase correlated with each other?

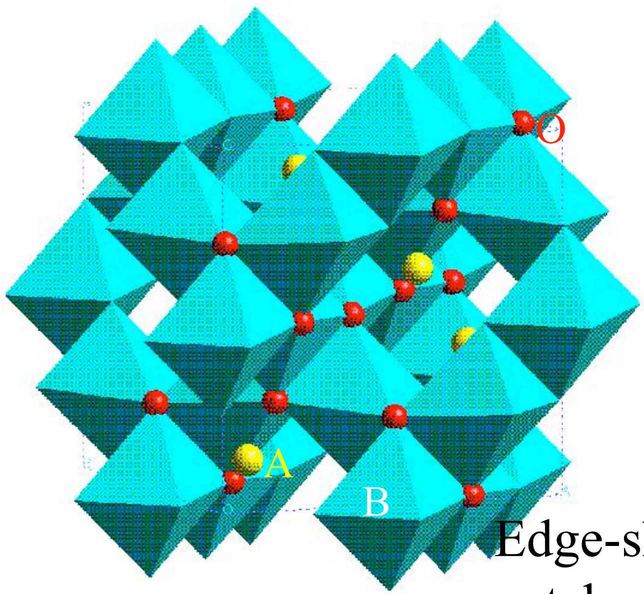
  Zero energy mode?

- **Novel Phase Transition?**

  How does nature respond to the ground state degeneracy?

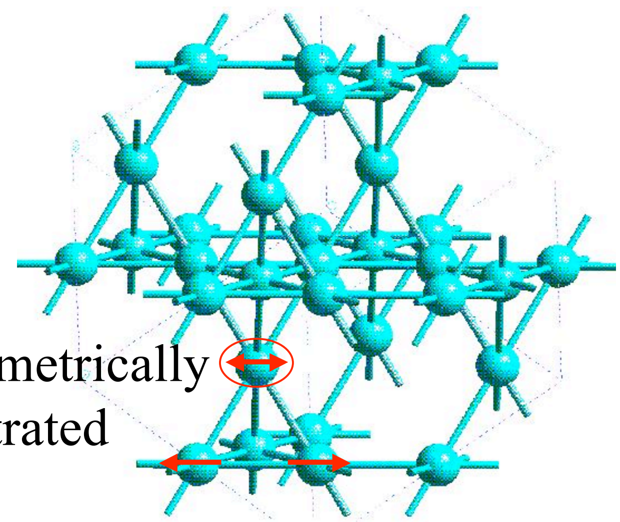
Why $ZnCr_2O_4$?

Space group Fd$\bar{3}$m



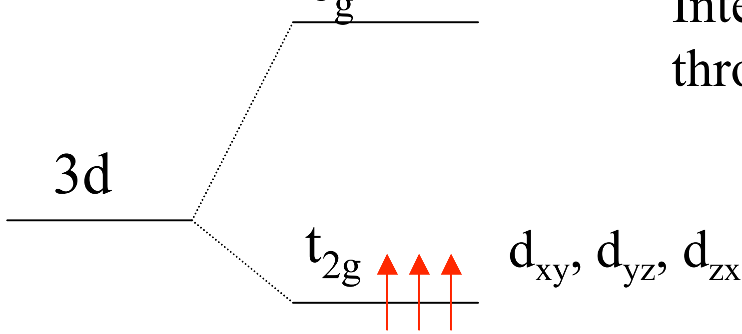
Edge-sharing octahedra

Lattice of B sites
: Corner-sharing tetrahedra



Geometrically frustrated

Cr^{3+} ($3d^3$)

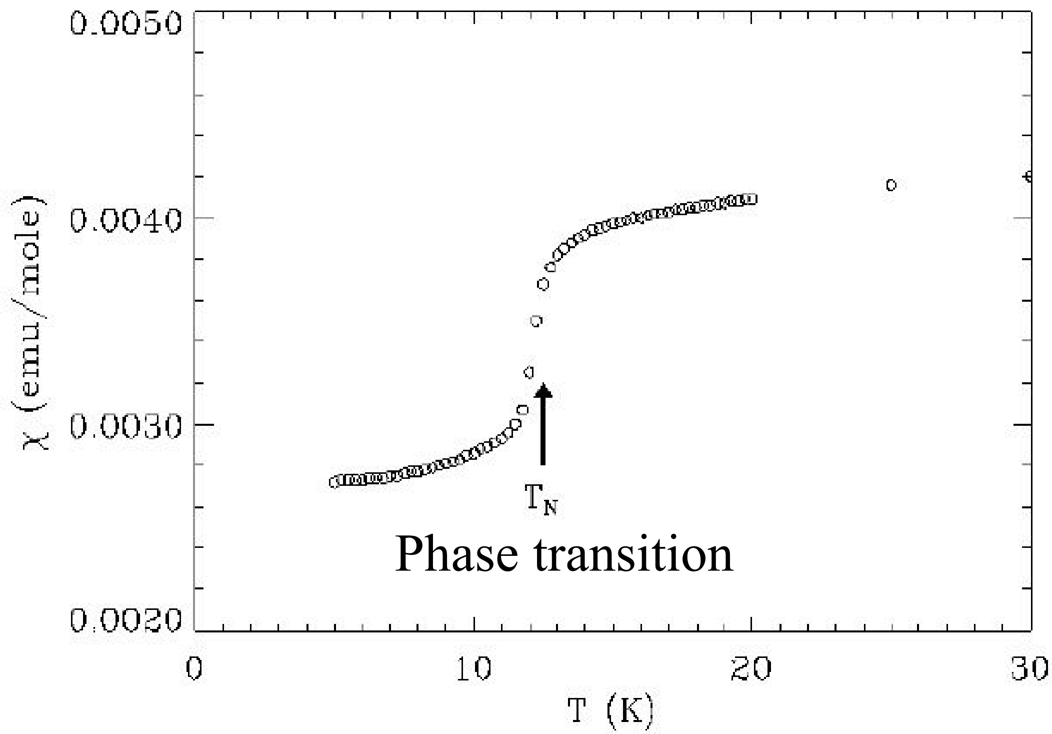


Interaction between nearest neighbor Cr^{3+} ions through direct overlap of t_{2g} orbitals is dominant.

Best candidate for a system with

$$H = -J \sum_{nn} \mathbf{S}_i \cdot \mathbf{S}_j$$

Magnetic Phase Transition in $ZnCr_2O_4$



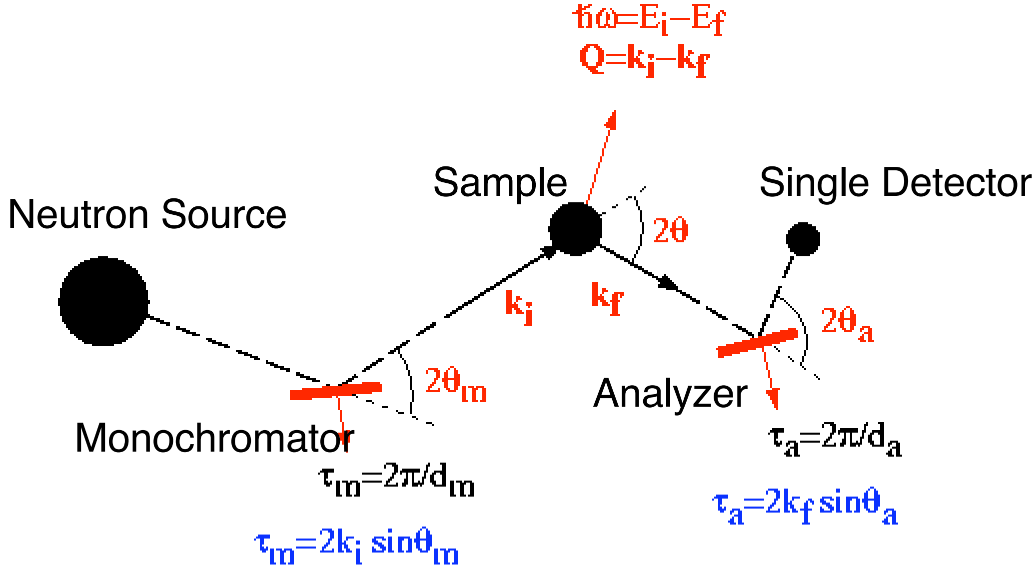
Θ_{CW} = -390 K

T_N = 12.5 K

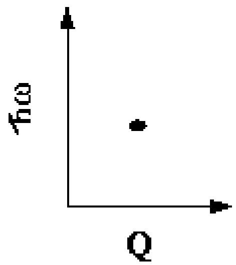
What is nature of the phase transition ?

SPINS : Cold Neutron Triple-Axis Spectrometer

Conventional Triple-Axis Spectroscopy (TAS)



A single point at a time



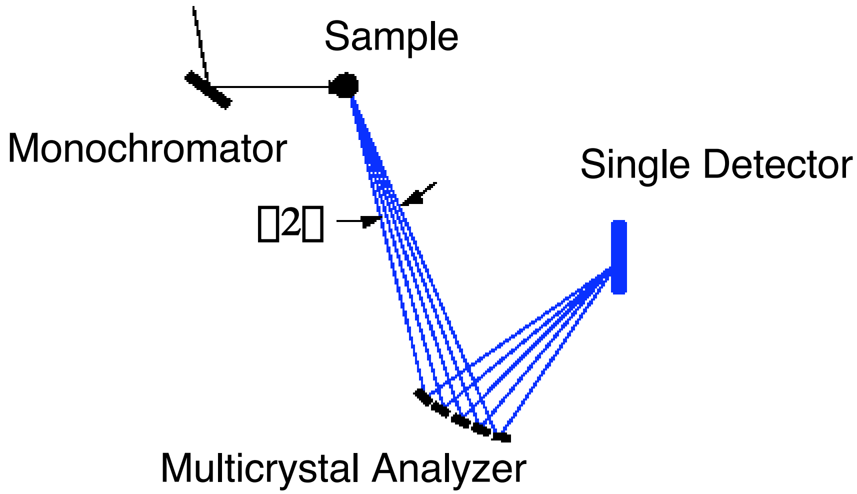
TAS is ideally suited for probing small regions of phase space

Shortcoming: Low data collection rate

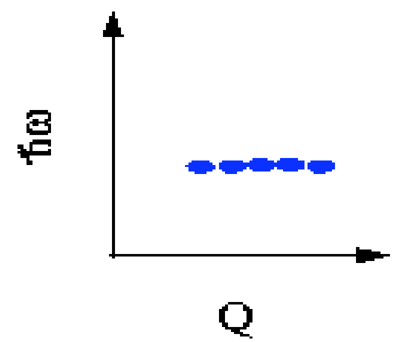
Improvement

Multicrystal analyzer and position-sensitive detector

Horizontally Focusing (HF) Analyzer Mode



Relaxed Q-resolution

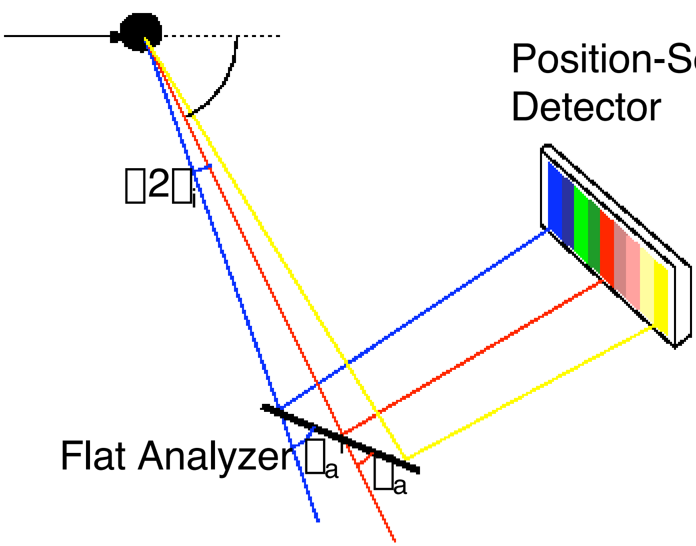


L = distance from sample to HF analyzer
w_a = total width of HF analyzer

$\Delta 2\theta = w_a \sin\theta_a / L$ ~ 9 degree for E_f=5 meV at SPINS

Useful for studying systems with short-range correlations

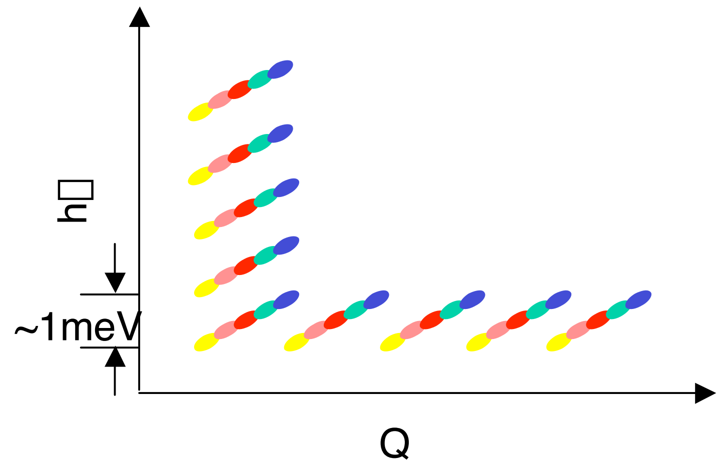
Multiplexing Detection System for TAS

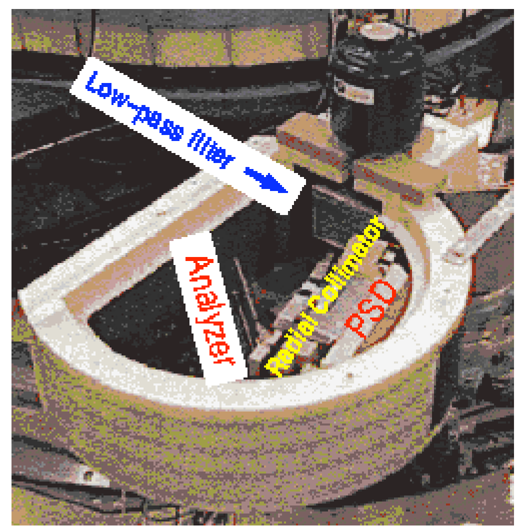


$\square_a^i = \square_a + \square 2\square_i = \square_a$ - atan(x sin$\square_a$/(L+xcos$\square_a$))

$k_f^i = \square_a / 2\sin\square_a^i$

$\mathbf{Q_i = k_i - k_f^i}$

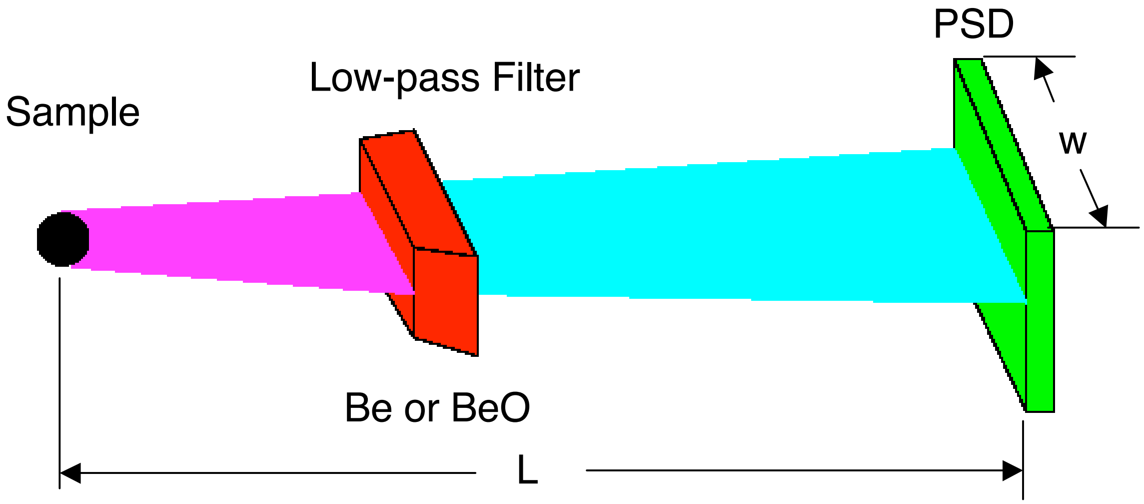




Probes scattering events at different energy and momentum transfers simultaneously

Survey (h□-Q) space by changing the incident energy and scattering angle

Position-Sensitive Detector in Two-Axis Mode

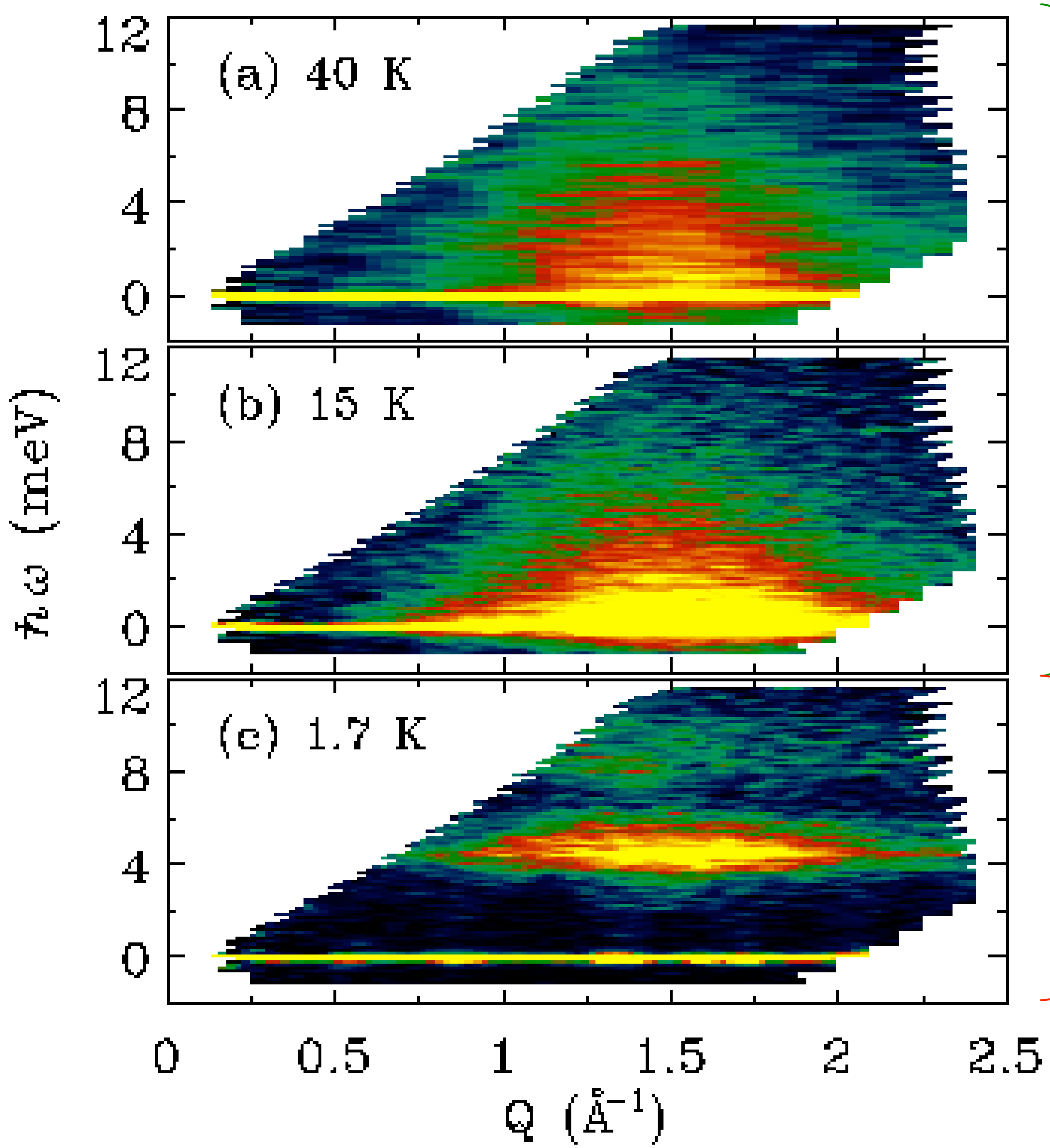


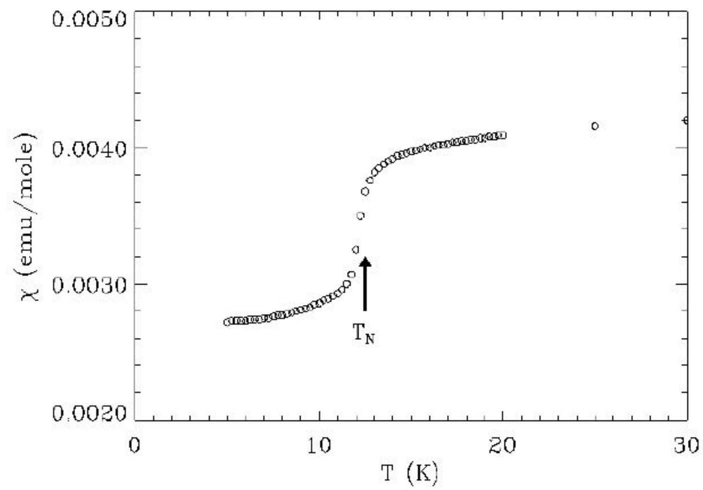
The filter passes only those neutrons with $0 < E_f < E^{filter}_{cutoff}$

The PSD measures $\int_{E_i - E_{cutoff}}^{E_i} S(Q,\omega)\, d\omega$

Large angular acceptance = w / L ~ 11^{o} for SPINS

Neutron Scattering from $ZnCr_2O_4$





Cooperative Paramagnetic fluctuations in frustrated AFM

Local spin resonance in magneto-elastic LRO phase

SHL et al., PRL **84**, 3718 (2000)

Nature of the Spin Liquid State of GF magnets

Emergent Excitations

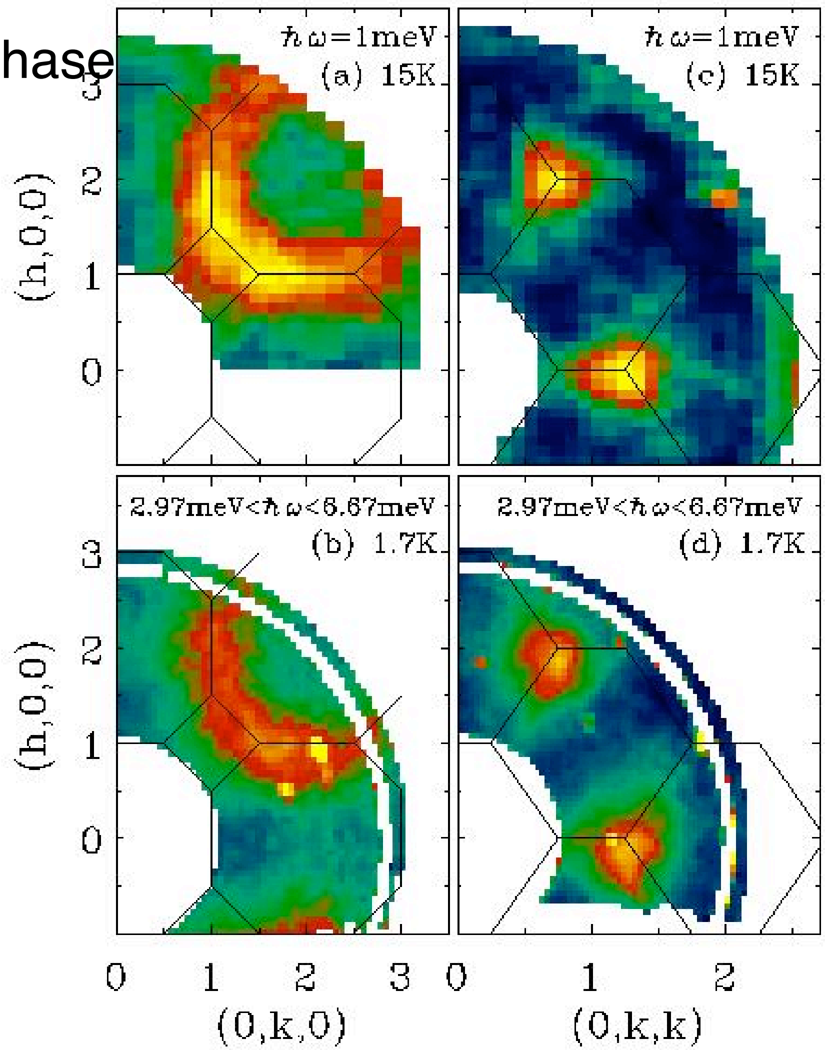
Structure Factor of Spin Fluctuations in $ZnCr_2O_4$

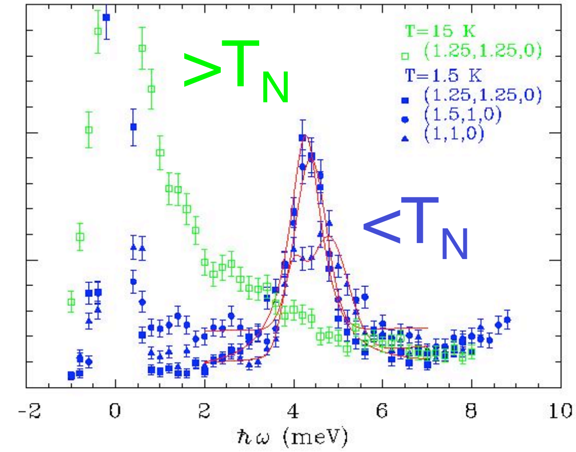
Spin liquid phase

$T > T_N$

Neel phase

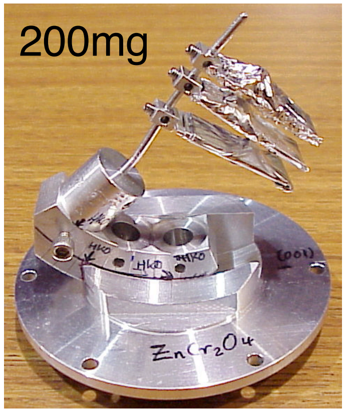
$T < T_N$





Identical S(Q)

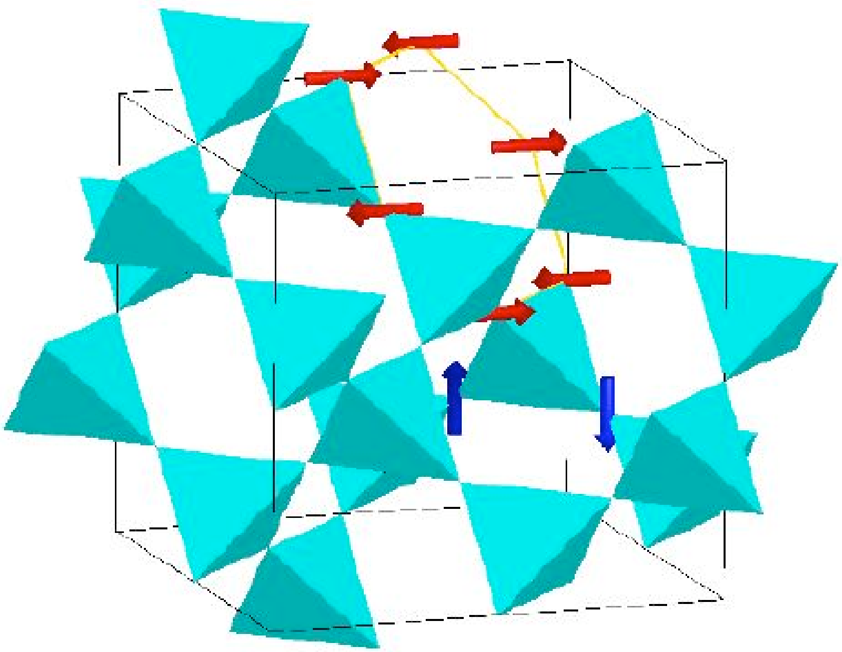
$ZnCr_2O_4$ single crystals

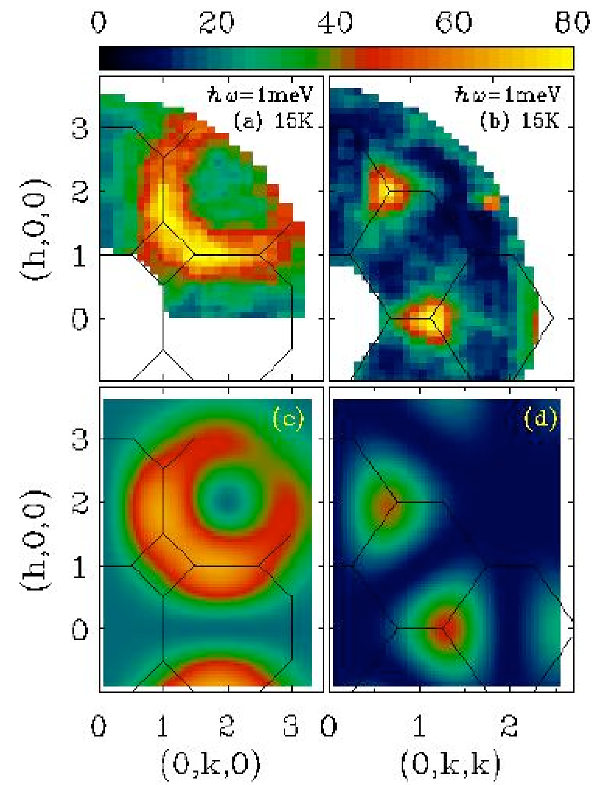


Extended Brillouin zone boundary scattering

Emergent Excitations in $ZnCr_2O_4$

SHL et al., Nature **418**, 856 (2002)





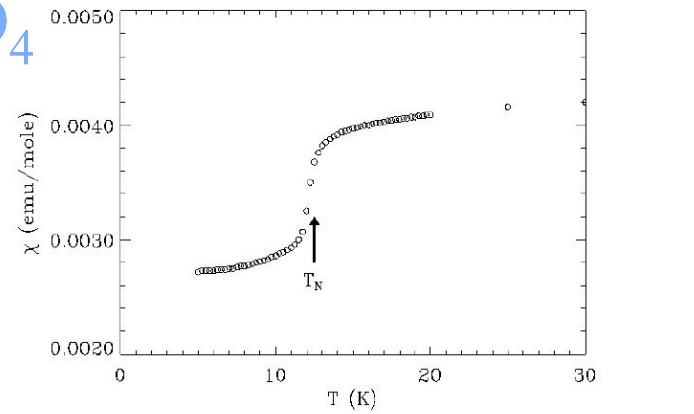
Hexagonal antiferromagnetic loops !

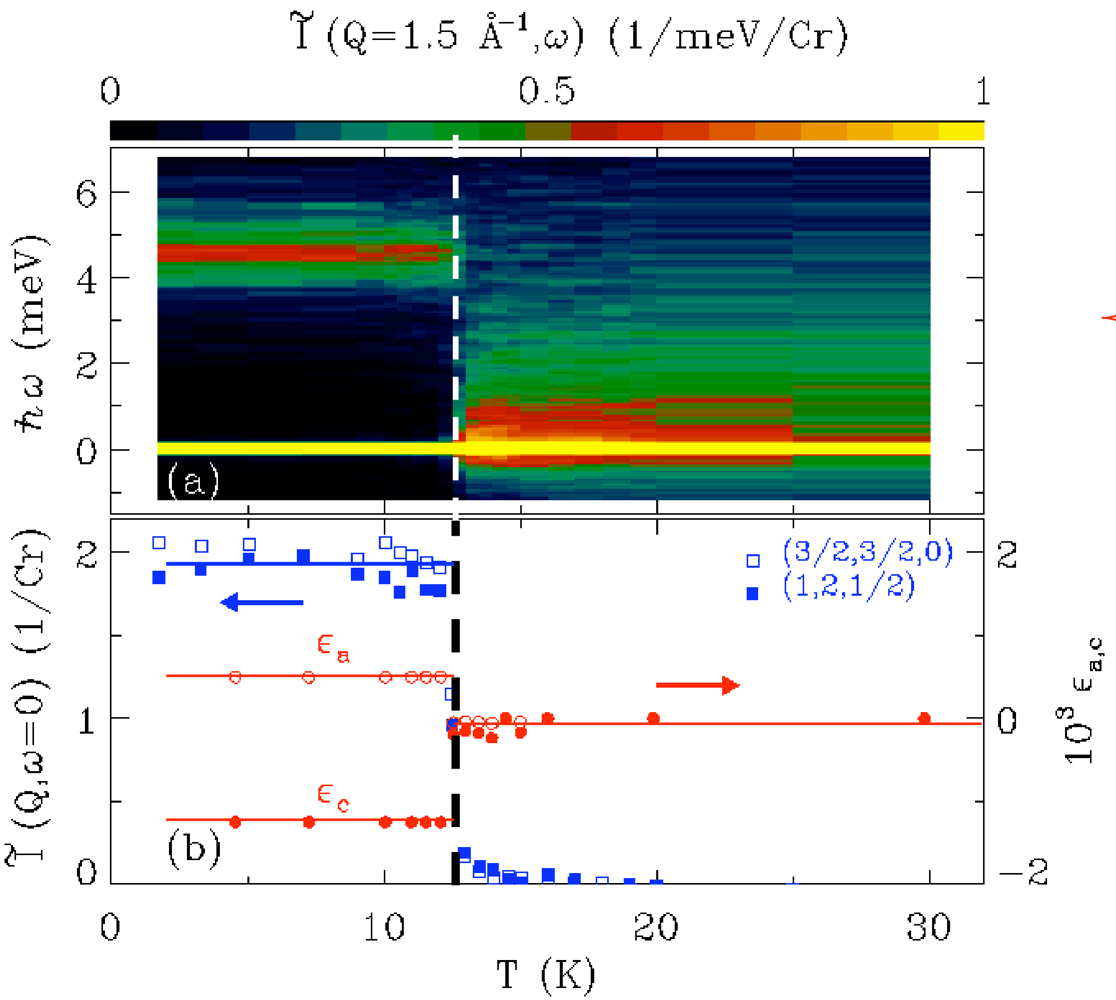
How does nature respond to the ground state degeneracy?

Spin-Peierls-like phase transition

Magneto-elastic phase transition in $ZnCr_2O_4$

How does nature respond to the ground state degeneracy?





Dynamics:

- Low energy paramagnetic fluctuations move to a resonance at 4.5 meV

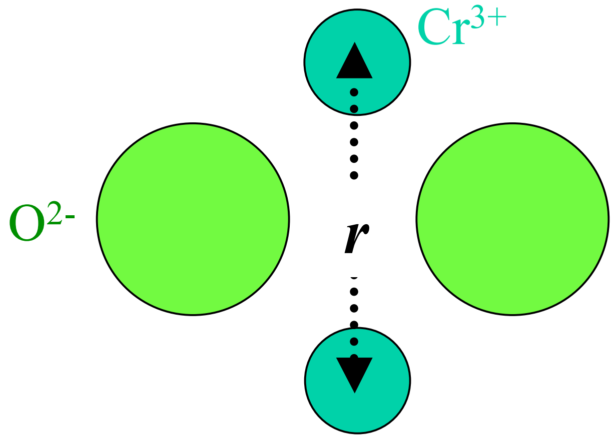
Statics:

- Staggered magnetization
- tetragonal lattice distortion

SHL et al., PRL **84**, 3718 (2000)

Why does tetragonal strain encourage Neel order ?

Edge sharing n-n exchange in $ZnCr_2O_4$ depends strongly on Cr-Cr distance, ***r*** :



From series of Cr-compounds: $\frac{dJ}{dr} \approx 40 \text{ meV}/\text{Å}$

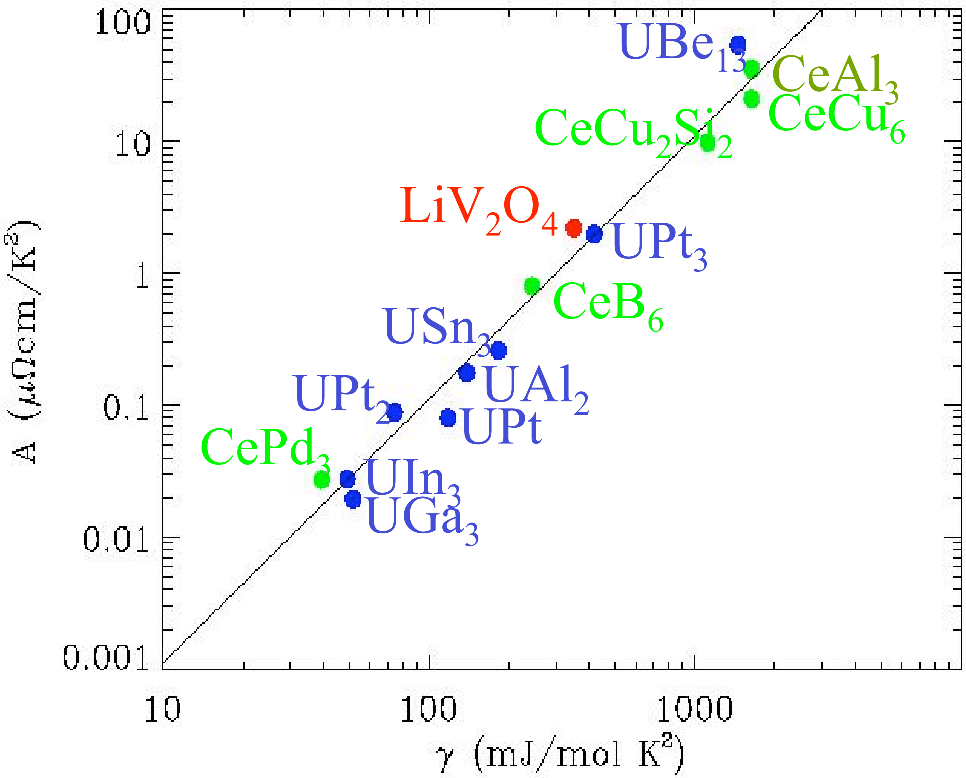
$$\begin{cases} \Delta J_{\parallel} = \frac{\delta_a + \delta_c}{2} r_0 \frac{dJ}{dr} = -0.04 \text{ meV} \\ \Delta J_{\perp} = \delta_a r_0 \frac{dJ}{dr} = 0.06 \text{ meV} \end{cases}$$

The effect for a single tetrahedron is to make 4 bonds more AFM and two bonds are less AFM. This relieves frustration!

What is the effect of geometrical frustration in a metallic system?

LiV_2O_4: d-Electron Heavy Fermion

S. Kondo et al., PRL (1998)



Bulk measurement data from LiV_2O_4 at low T exhibit Fermi liquid behaviors

$C_v \sim \gamma T$

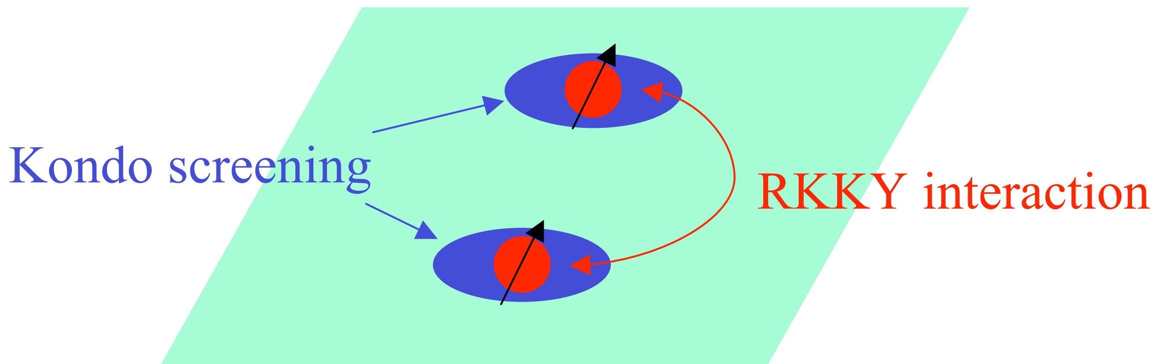
$\rho = \rho_0 + AT^2$

$\chi =$ const

LiV_2O_4 with d-electrons is as heavy as UPt_3 !

Heavy Fermion

Heavy fermion behaviors with a heavy mass m ~ 100-1000 m_e are usually found in **Ce- or U-based compounds** that have **two different types of electrons**: (1) localized f-electrons and (2) conduction (s,p)-electrons.

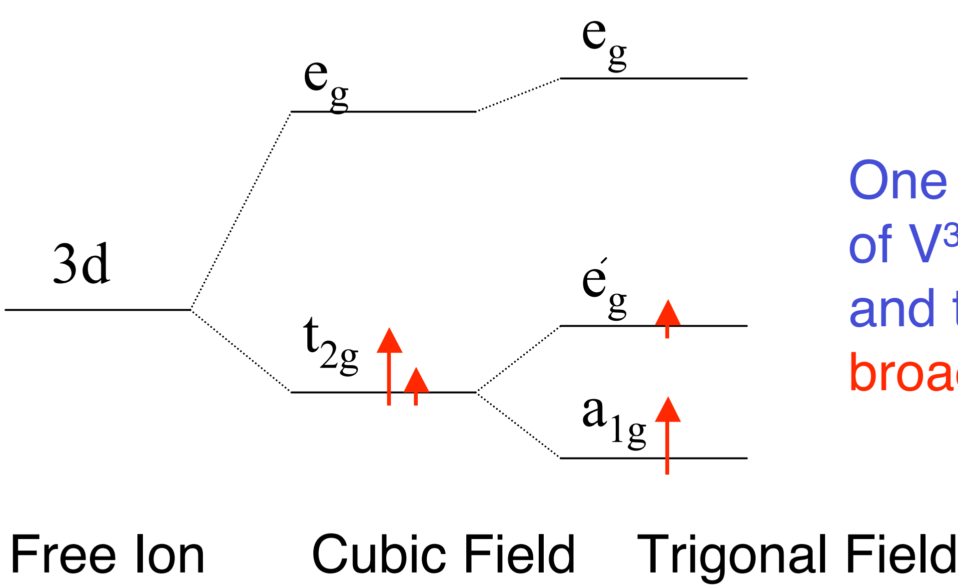


Issues : What is the mechanism for the HF behaviors in LiV_2O_4?
Why is LiV_2O_4 as heavy as UPt_3 ?
Is the HF state of LiV_2O_4 different from those in other Ce- and U-based HF compounds?

Mechanism for HF behavior in LiV_2O_4

1. Kondo screening

V.I. Anisimov et al., PRL 83, 364 (1999)

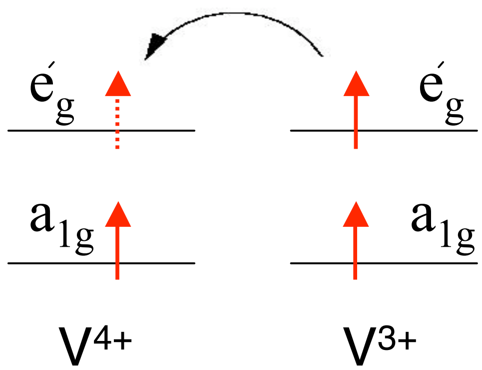


One electron of the $d^{1.5}$ configuration of $V^{3.5+}$ ion is in the localized a_{1g} orbital and the rest partially fills the relatively broad conduction e'_g band

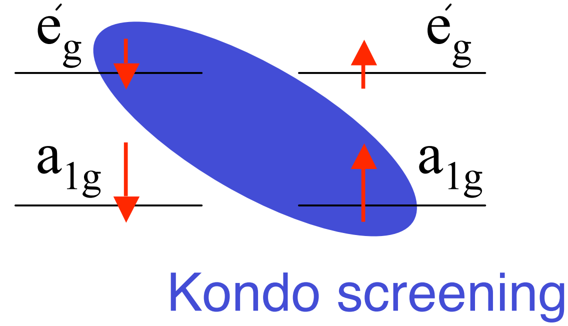
A Kondo screening possible !?!

Two competing interactions between neighboring local V moments:

FM double exchange interaction



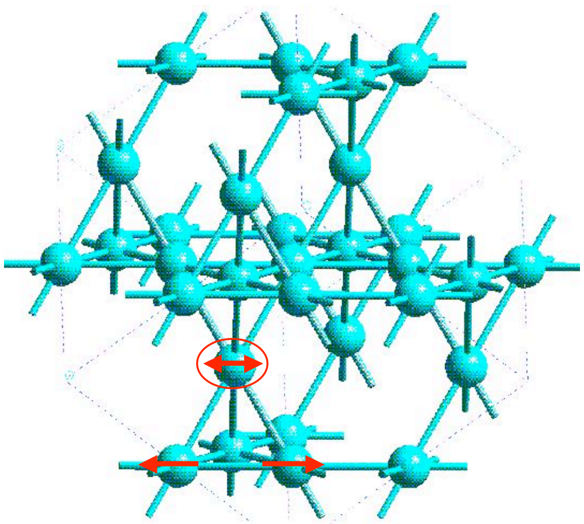
Kondo-induced **AFM** interaction



Mechanism for HF behavior in LiV_2O_4

2. Geometrical Frustration

V. Eyert et al., Europhy. Letters, 46, 762 (1999)

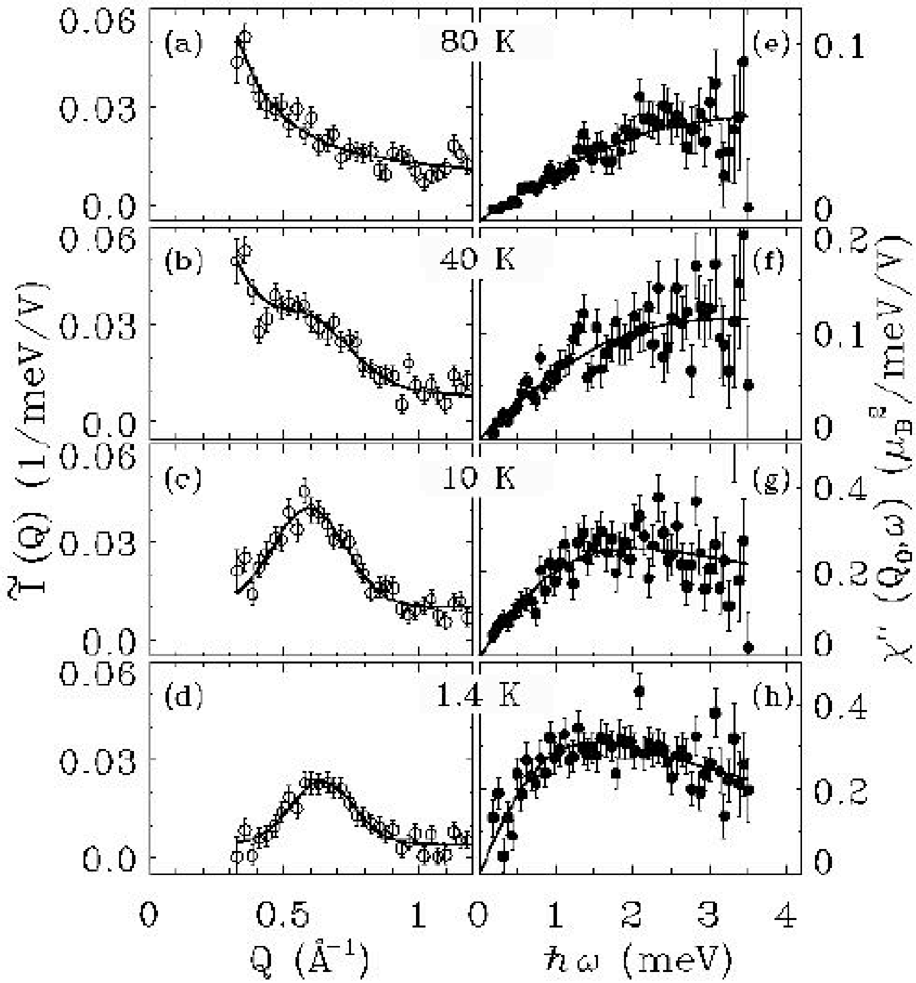


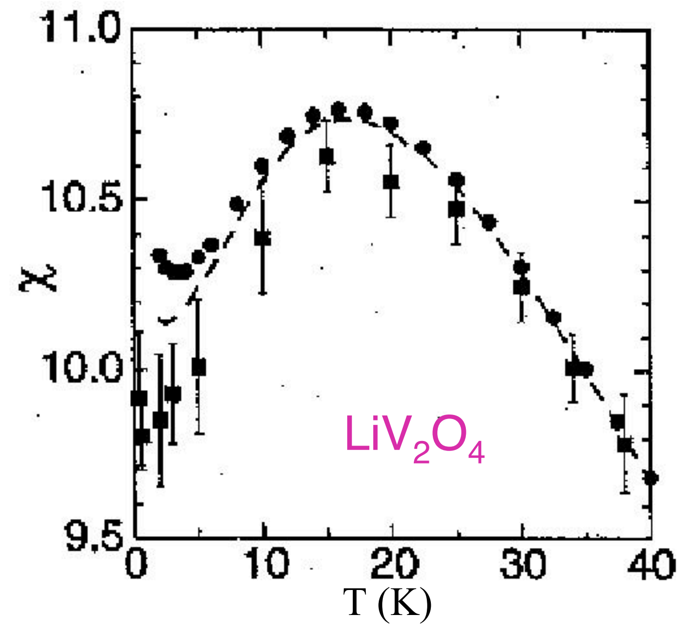
V moments interact antiferromagnetically
in the frustrating lattice.

C_v = dE/dT = d(TS)/dT ~ S (Entropy)

Geometrical frustration → Strong spin fluctuations
→ Enhancement of specific heat (HF behavior)

LiV_2O_4 $(3d^{1.5})$:
Dynamic Spin Correlations

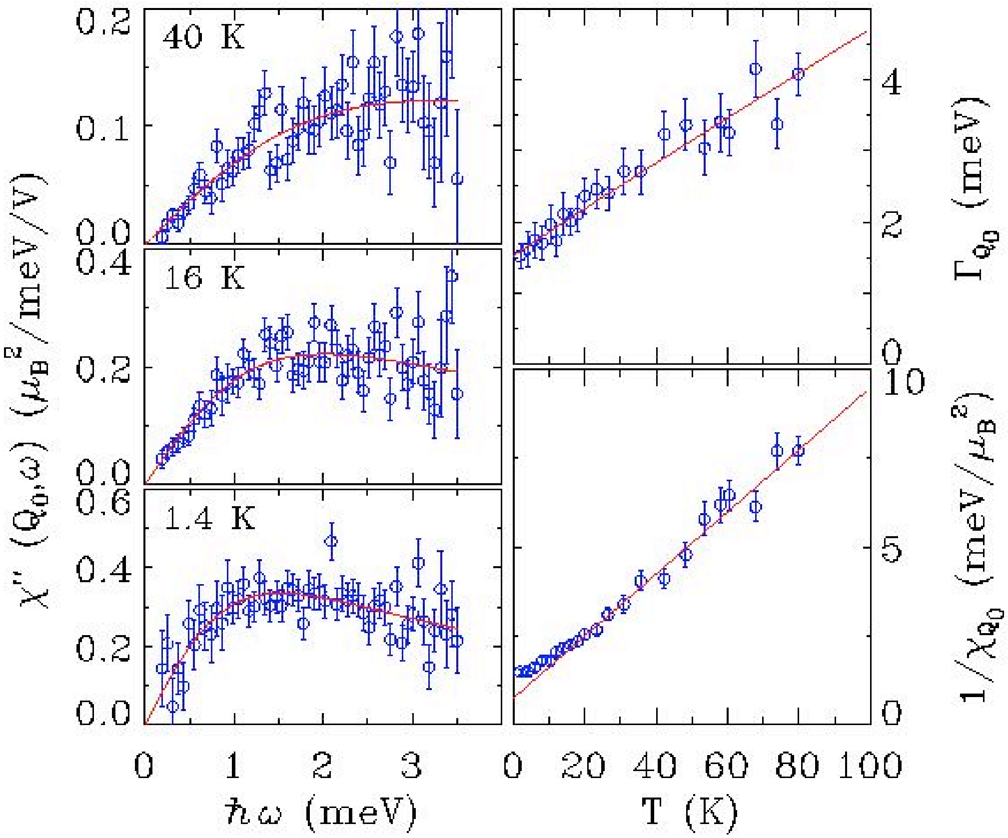




SHL et al., PRL (2001)

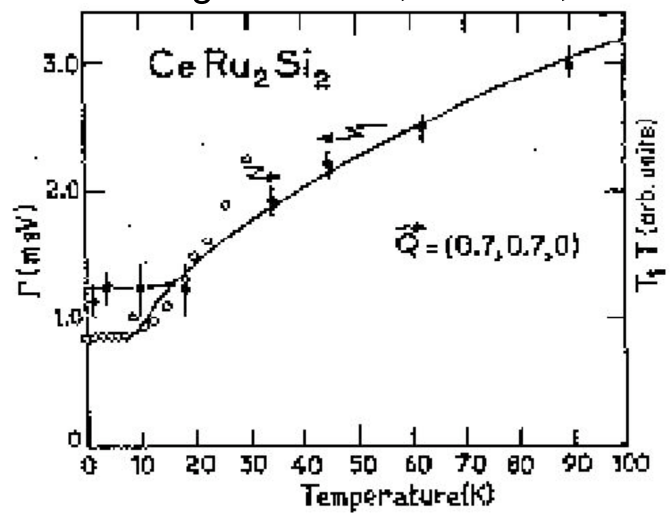
Dynamic spin correlations become AFM as LiV_2O_4 enters the heavy fermion phase

Dynamic Magnetic Correlations in LiV_2O_4



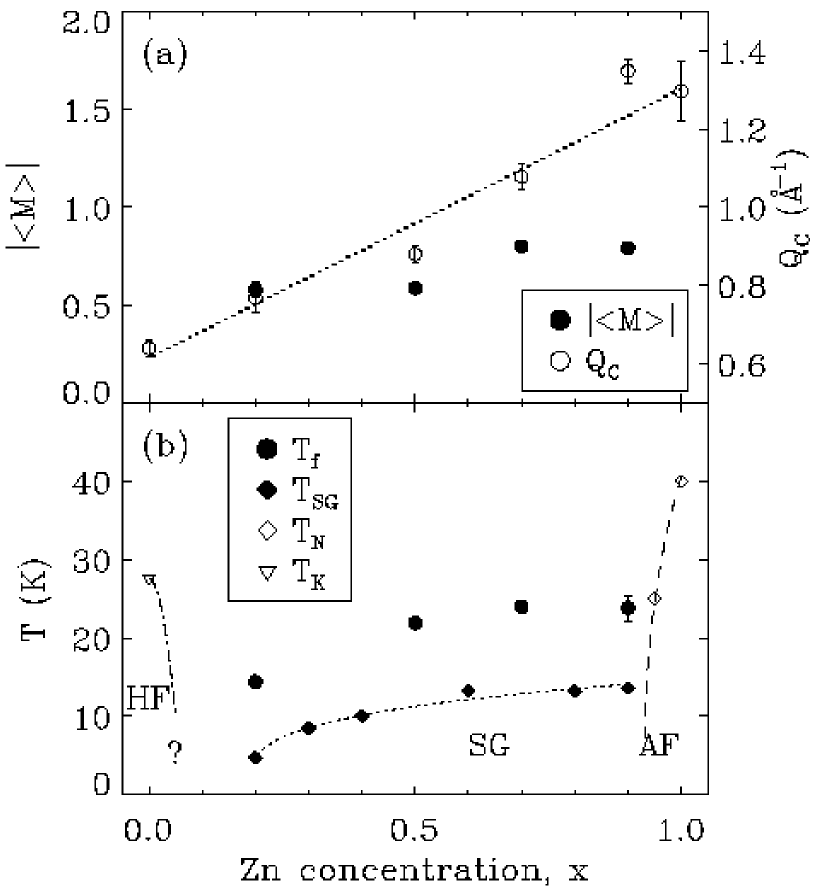
$\square(T)=\square_0+k_BT^{\square}$
with $\square_0$=1.5 meV=T_K (Kondo T)~T_c
and $\square$=0.96(2)

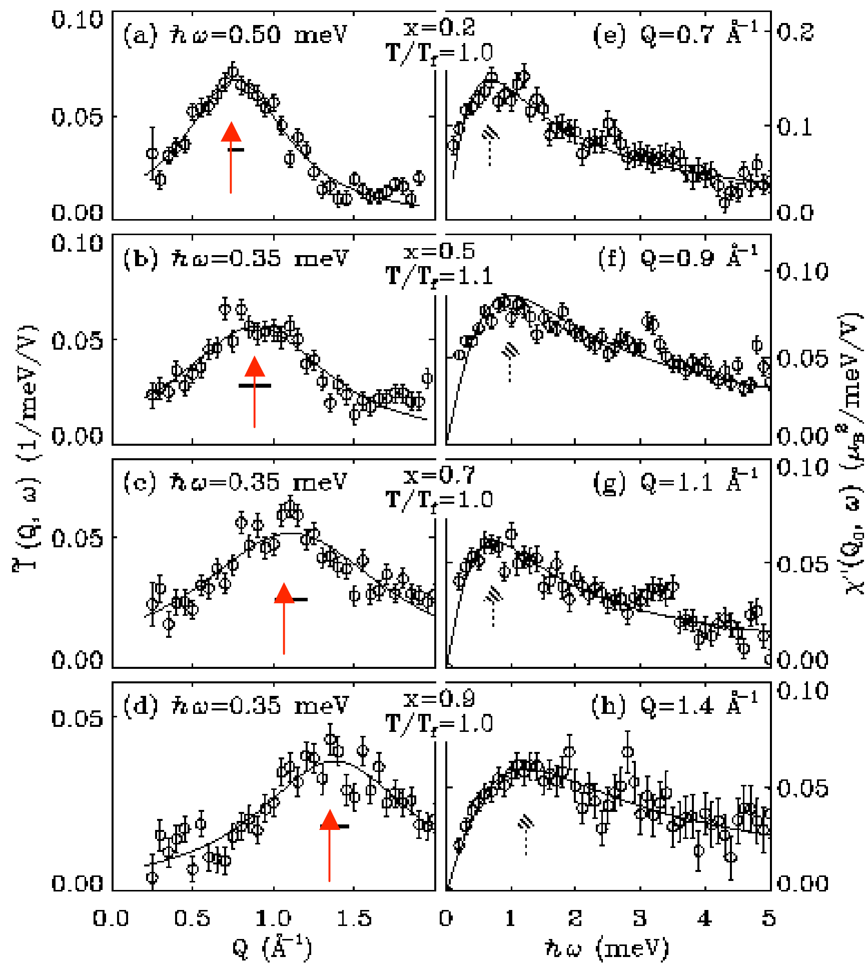
L.P. Regnault et al., PRB 38, 4481 (1988)



Linear □ (T) is a common feature of geometrically frustrated antiferromagnets.

Evolution of magnetic correlations in $Li_{1-x}Zn_xV_2O_4$





S. Park et al., unpublished (2003)

Summary

$ZnCr_2O_4$

1. Ground state degeneracy
   → Spectral weight down-shift to $E < \square_{CW}$
2. Weak connectivity of spin correlations due to triangular motif
   → Emergence of local spin clusters with zero energy modes

$Li_{1-x}Zn_xV_2O_4$

3. Relation between the metallic properties and magnetism is still in question. The clue to the answer might be in understanding why unlike insulating magnets the characteristic wave vector for magnetic correlations changes gradually as the Zn concentration

Elastic and Inelastic diffuse scattering on single crystals is indispensable to identify the fundamental spin degrees of freedom in GF magnets