Resolving a Magnetic Quandary

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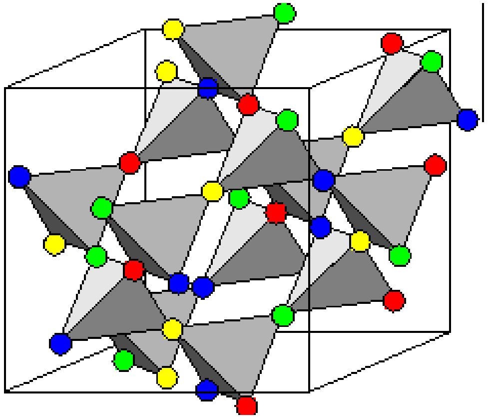

- Introduction
  - Magneto-elastic transitions
  - Magnetic frustration
- Order by coupling to lattice or charge
  - Spin Peierls like transition in $ZnCr_2O_4$
  - Resolving frustration in $Y_2Mo_2O_7$
  - orbital fluctuations and magnetism in $(V_{1-x}Cr_x)_2O_3$
- Conclusions

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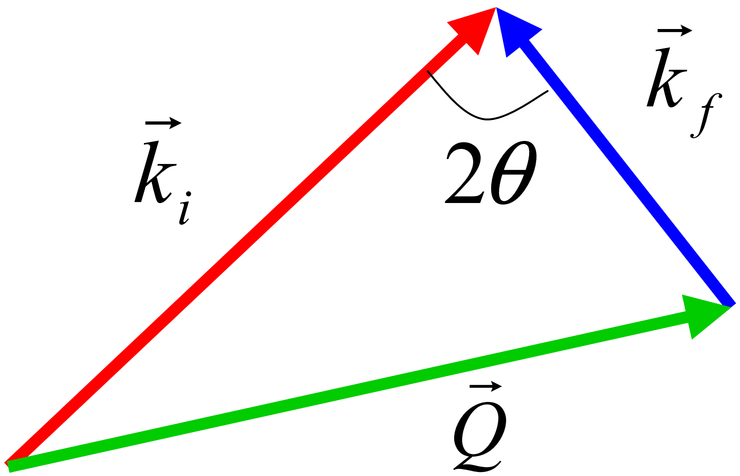
Magnetic Neutron Scattering



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

$$\hbar\omega = E_i - E_f$$

The scattering cross section is proportional to the Fourier transformed dynamic spin correlation function

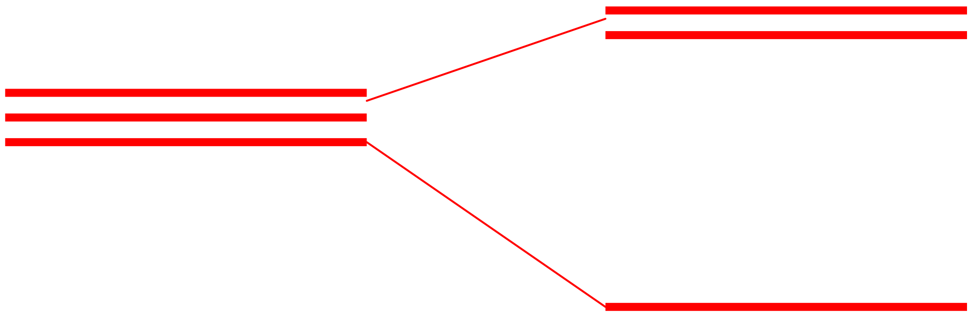
$$\mathbf{S}^{\alpha\beta}(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, e^{-i\omega t}\frac{1}{N}\sum_{\vec{R}\vec{R}'} e^{i\vec{Q}\bullet(\vec{R}-\vec{R}')} < \mathrm{S}^{\alpha}_{\vec{\mathrm{R}}}(t)\mathrm{S}^{\beta}_{\vec{R}'}(0) >$$

Fluctuation dissipation theorem:

$$\chi''(Q,\omega) = (g\mu_B)^2\pi\left(1 - e^{-\beta\hbar\omega}\right)\mathbf{S}(Q,\omega)$$

Jahn-Teller Theorem

Any molecule or complex ion in an electronically degenerate state will be unstable relative to a configuration of lower symmetry in which the degeneracy is absent



Is exchange *constant* ?

$$\mathscr{H} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

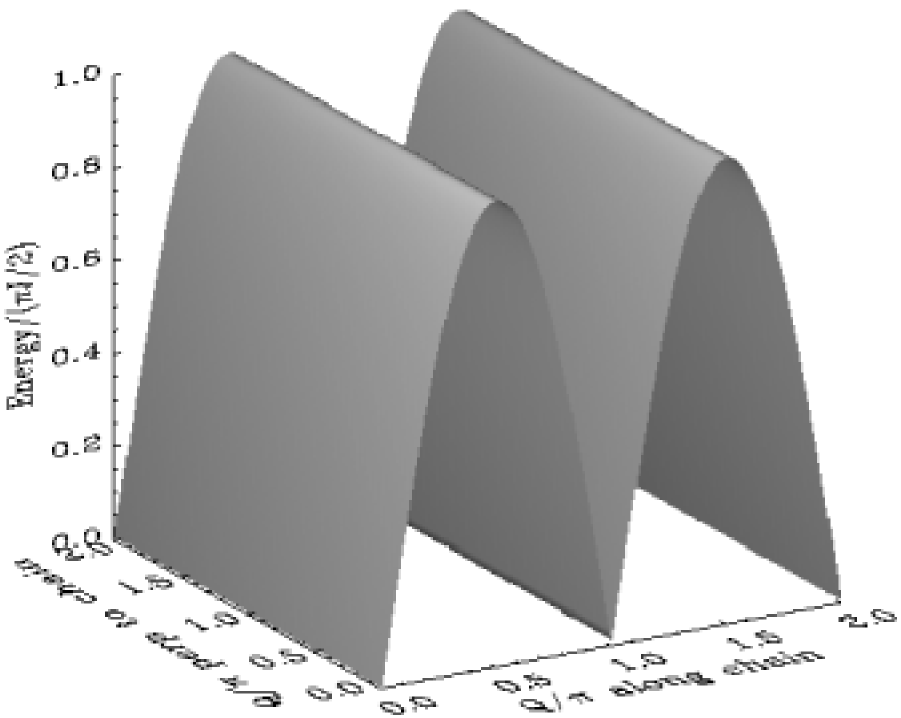
☞ J_{ij} is controlled by higher energy physics that we like to consider irrelevant at low energies

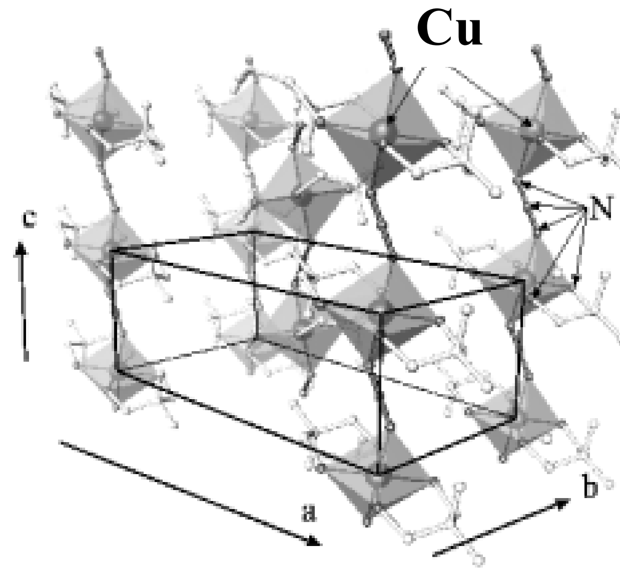
- atomic spacing
- Orbital overlap
- Orbital occupancy
- localized or itinerant electronic states

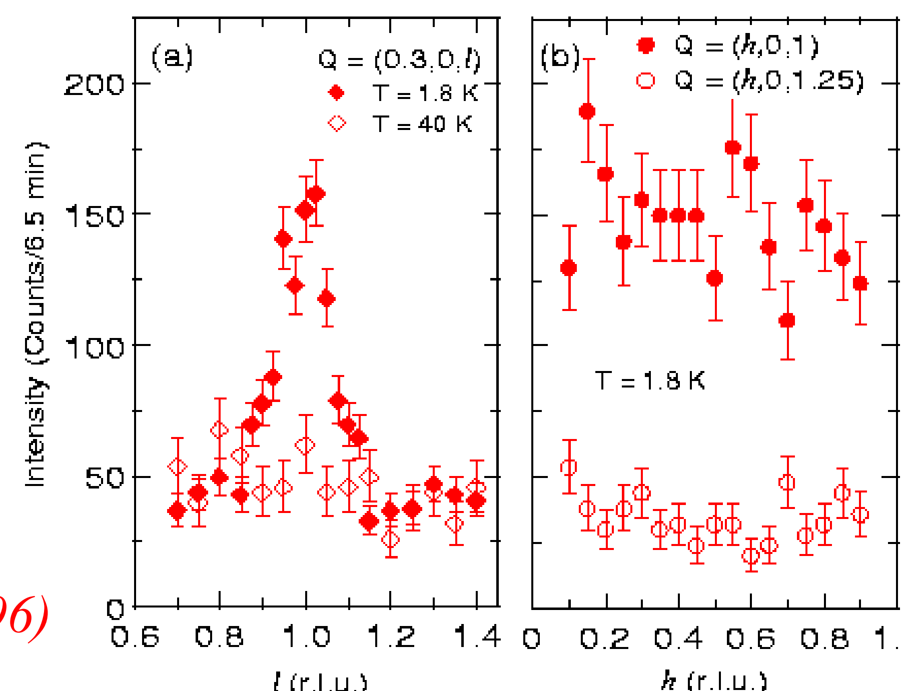
☞ These degrees of freedom can become relevant if $\mathscr{H}$ produces "degenerate" state

☞ The result can be intricate interplay between spin charge and lattice sectors

D<3 magnets can have extensive soft modes

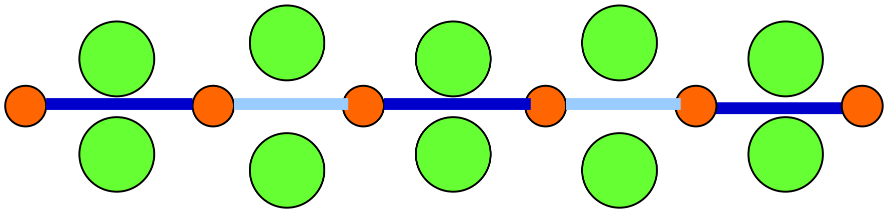


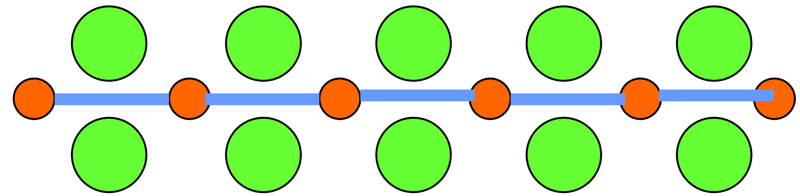




Copper Benzoate Dender et al. (1996)

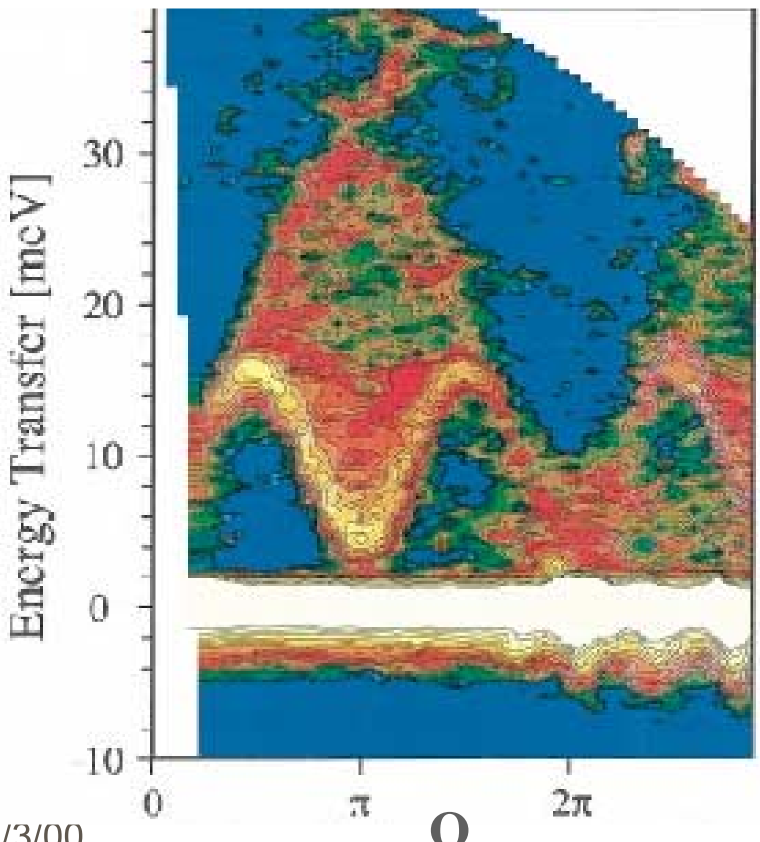
Spin Peierls Transition

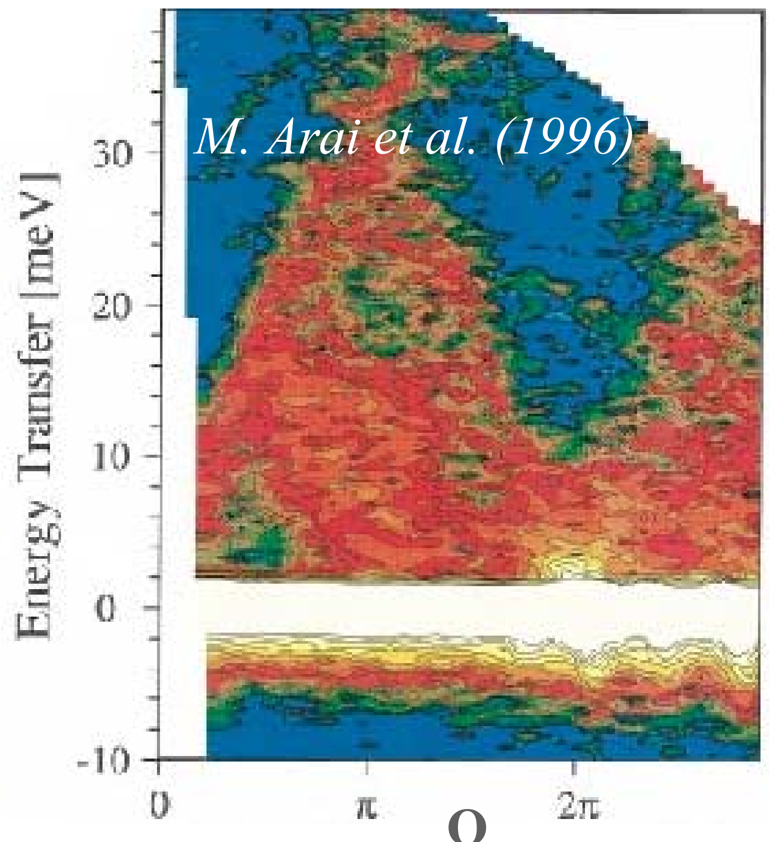




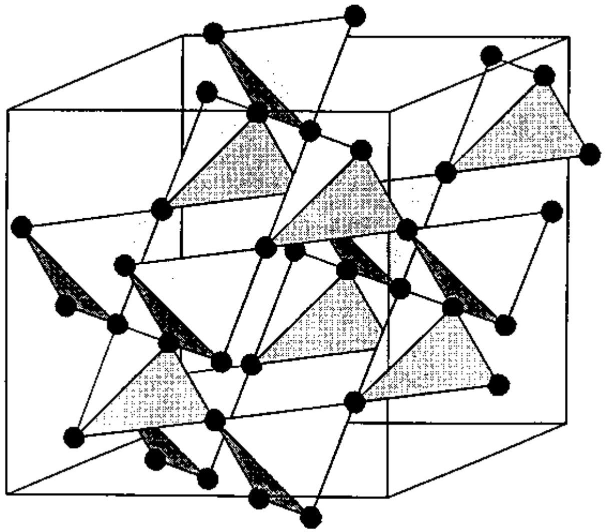
$$\mathscr{H} = -\sum_l \left(J\mathbf{S}_{2l} \cdot \mathbf{S}_{2l+1} + J'\mathbf{S}_{2l} \cdot \mathbf{S}_{2l-1} \right)$$

$$\mathscr{H} = -\sum_l J\mathbf{S}_l \cdot \mathbf{S}_{l+1}$$





Spins with AFM interactions on corner-sharing tetrahedra



What is special about this lattice?

- Low coordination number
- Triangular motif
- Infinite set of mean field ground states
- with zero net spin on all tetrahedra
- No barriers between mean field ground states
- Q-space degeneracy for spin waves

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
Anisotropic	$S=\infty$	Neel order	MC sim.	Bramwell, Gingras, Reimers J. Appl. Phys. '94

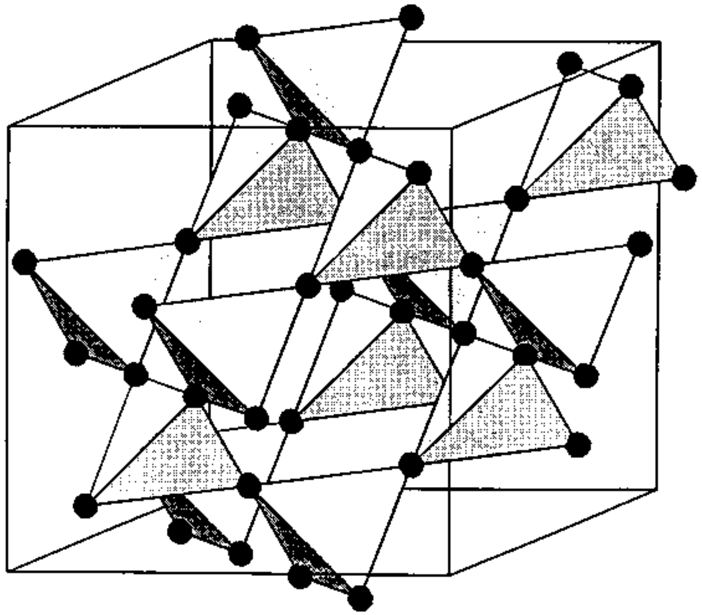
spins on corner sharing tetrahedra

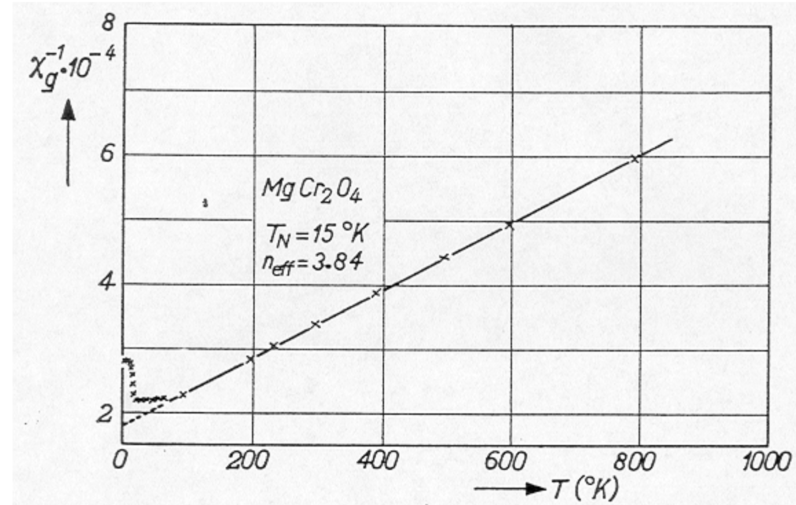
B-spinel / Pyrochlore

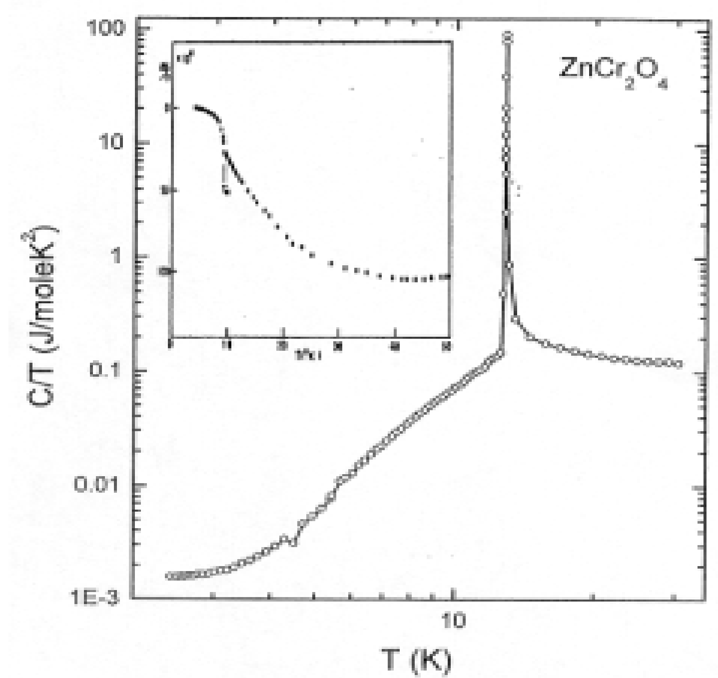
Material	spin type	spin value	Θ_{CW} (K)	T_c (K)	Low T phase	Ref.
MgV_2O_4	isotrop.	1	-750	45	LRO	Baltzer et al '66
ZnV_2O_4	isotrop.	1	-600	40	LRO	Ueda et al '97
$CdCr_2O_4$	isotrop.	3/2	-83	9	LRO	Baltzer et al '66
$MgCr_2O_4$	isotrop.	3/2	-350	15	LRO	Blasse and Fast '63
$ZnCr_2O_4$	isotrop.	3/2	-392	12.5	LRO	S.-H. Lee et al '99
FeF_3	isotrop.	5/2	-230	20	LRO	Ferey et al. '86
$Y_2Mo_2O_7$	isotrop.	1	-200	22.5	spin glass	Gingras et al. '97
$Y_2Mn_2O_7$	isotrop.	3/2		17	spin glass	Reimers et al '91
$Tb_2Mo_2O_7$	anisotr.	6 and 1		25	spin glass	Greedan et al '91
$Gd_2Ti_2O_7$	isotrop.	7/2	-10	1	LRO	Radu et al '99
$Er_2Ti_2O_7$	anisotr.		-25	1.25	LRO	Ramirez et al '99
$Tb_2Ti_2O_7$	anisotr.		-19		spin liquid?	Gardner et al '99
$Yb_2Ti_2O_7$	anisotr.		0	0.21	LRO	Ramirez et al '99
$Dy_2Ti_2O_7$	Ising	7.5 → 1/2	0.5	1.2	spin ice	Ramirez et al '99
$Ho_2Ti_2O_7$	Ising	8 → 1/2	1.9		spin ice	Harris et al "97

Subjects of this talk

$ZnCr_2O_4$: Corner sharing tetrahedra

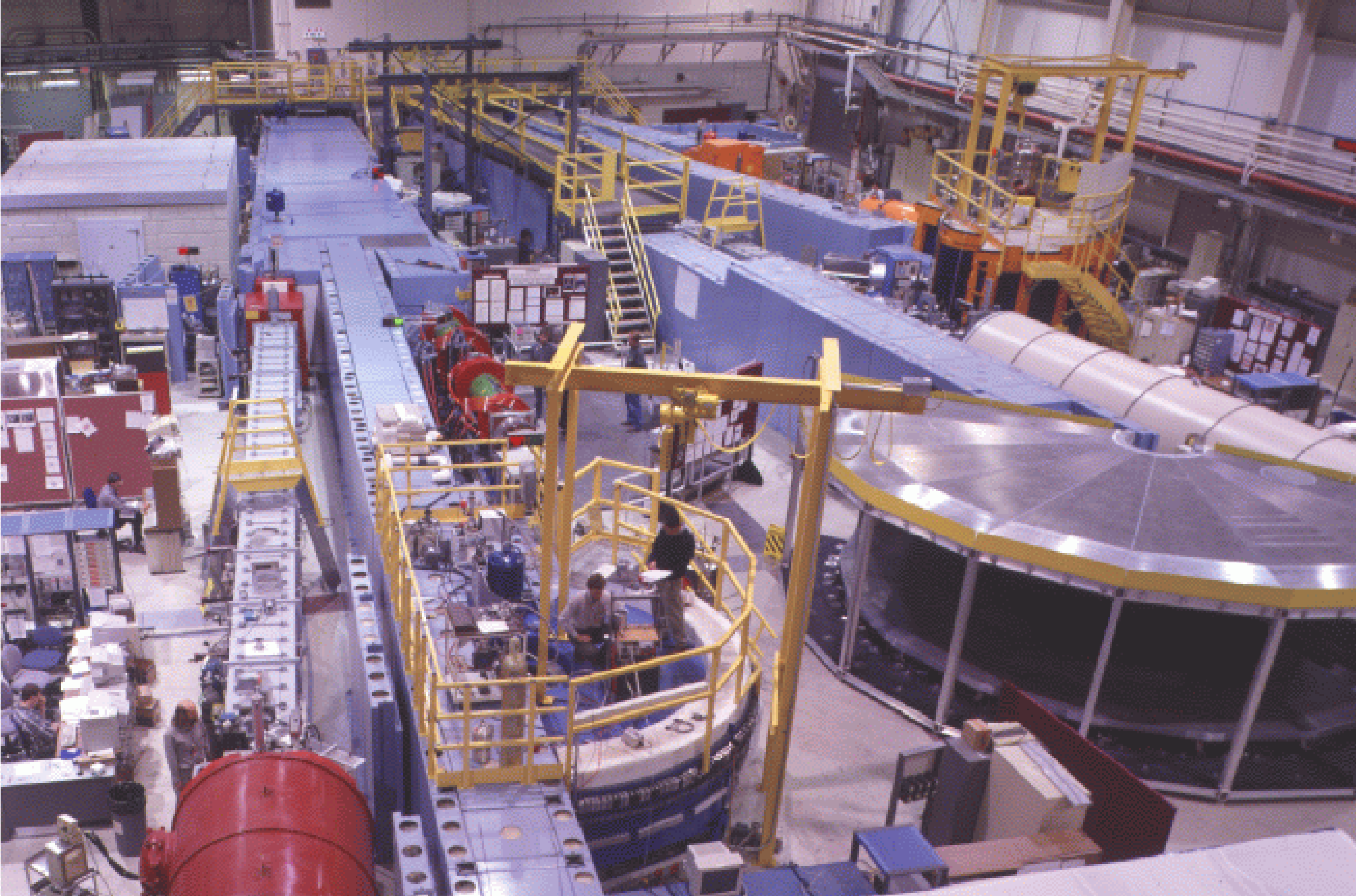


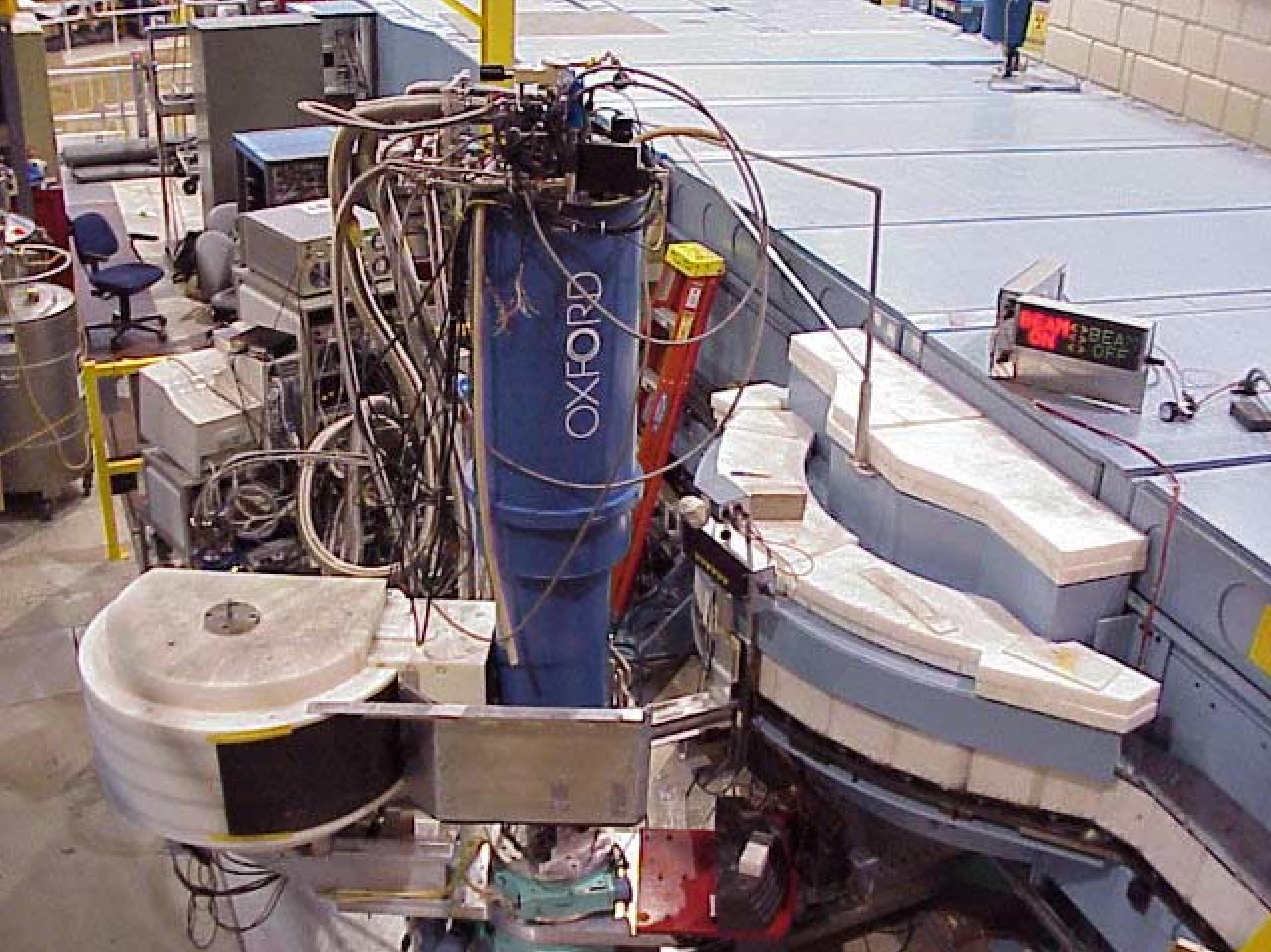




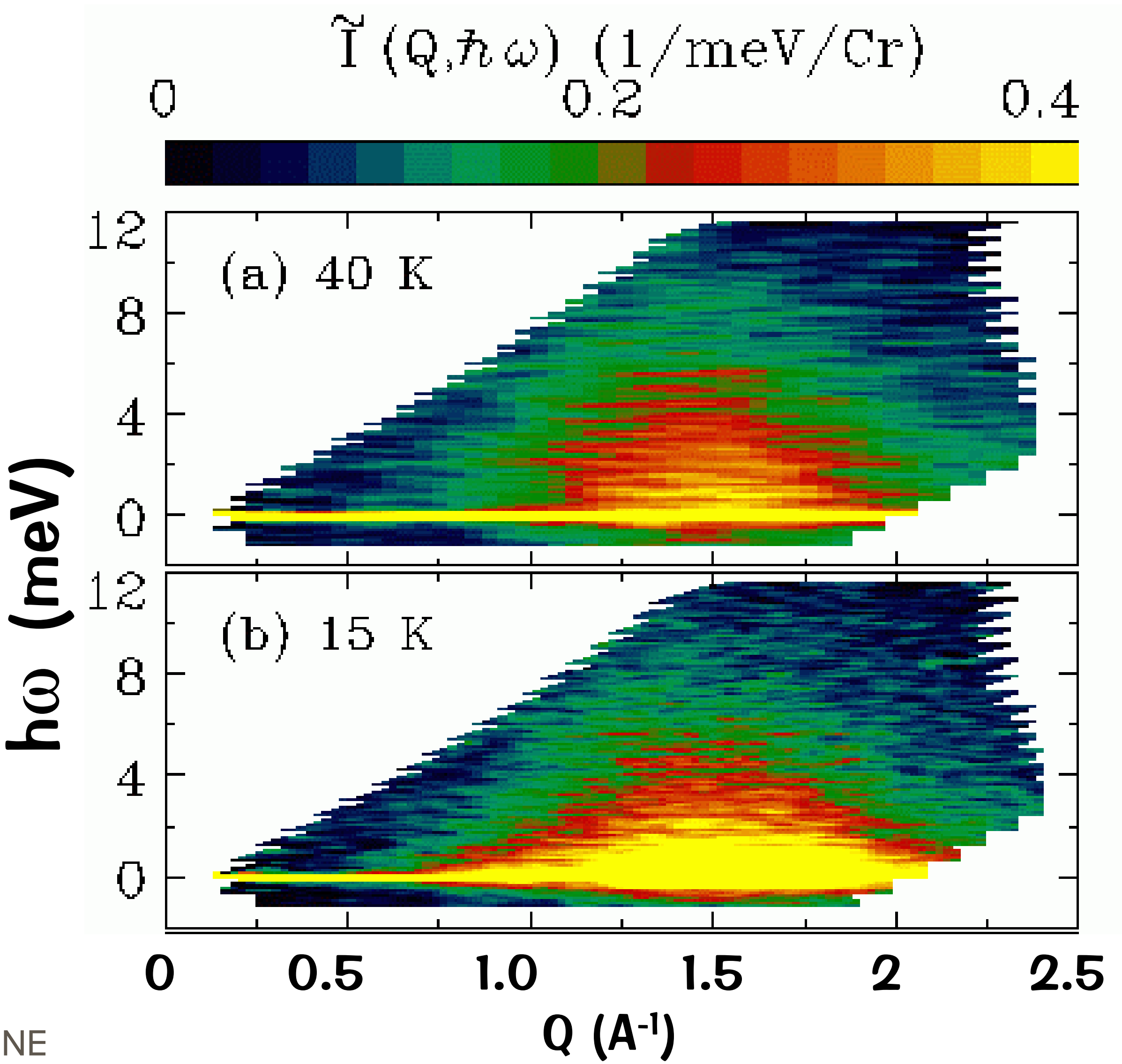
Material	Θ_{CW}	T_N	Θ_{CW}/T_N
MgV_2O_4	-750	45	17
ZnV_2O_4	-600	40	15
$CdCr_2O_4$	-83	9	9
$MgCr_2O_4$	-350	15	23
$ZnCr_2O_4$	-392	12.5	31

NIST Center for Neutron Research




OXFORD
BEAM OFF

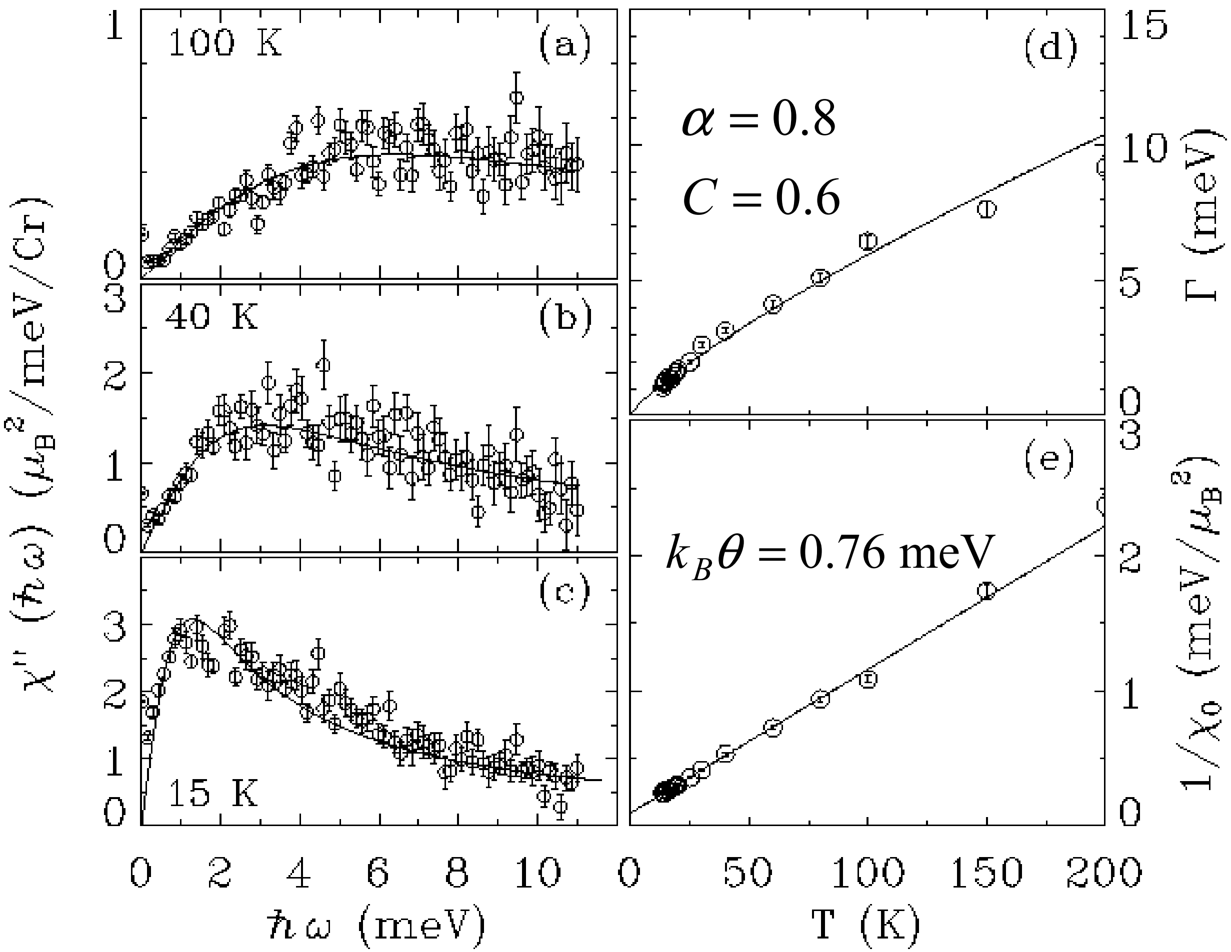
Signs of frustration: short range order for $T<|\Theta_{CW}|$



<u>Points of interest:</u>

- $2\pi/Qr_0=1.4$
  => nn. AFM correlations
- No scattering at low Q
  => satisfied tetrahedra
- Relaxation rate of order k_BT
  => quantum critical

Approaching quantum criticality



Lorentzian relaxation spectrum:

$$\chi''(Q,\omega) = \frac{\chi_Q \Gamma_Q \omega}{\omega^2 + \Gamma_Q^2}$$

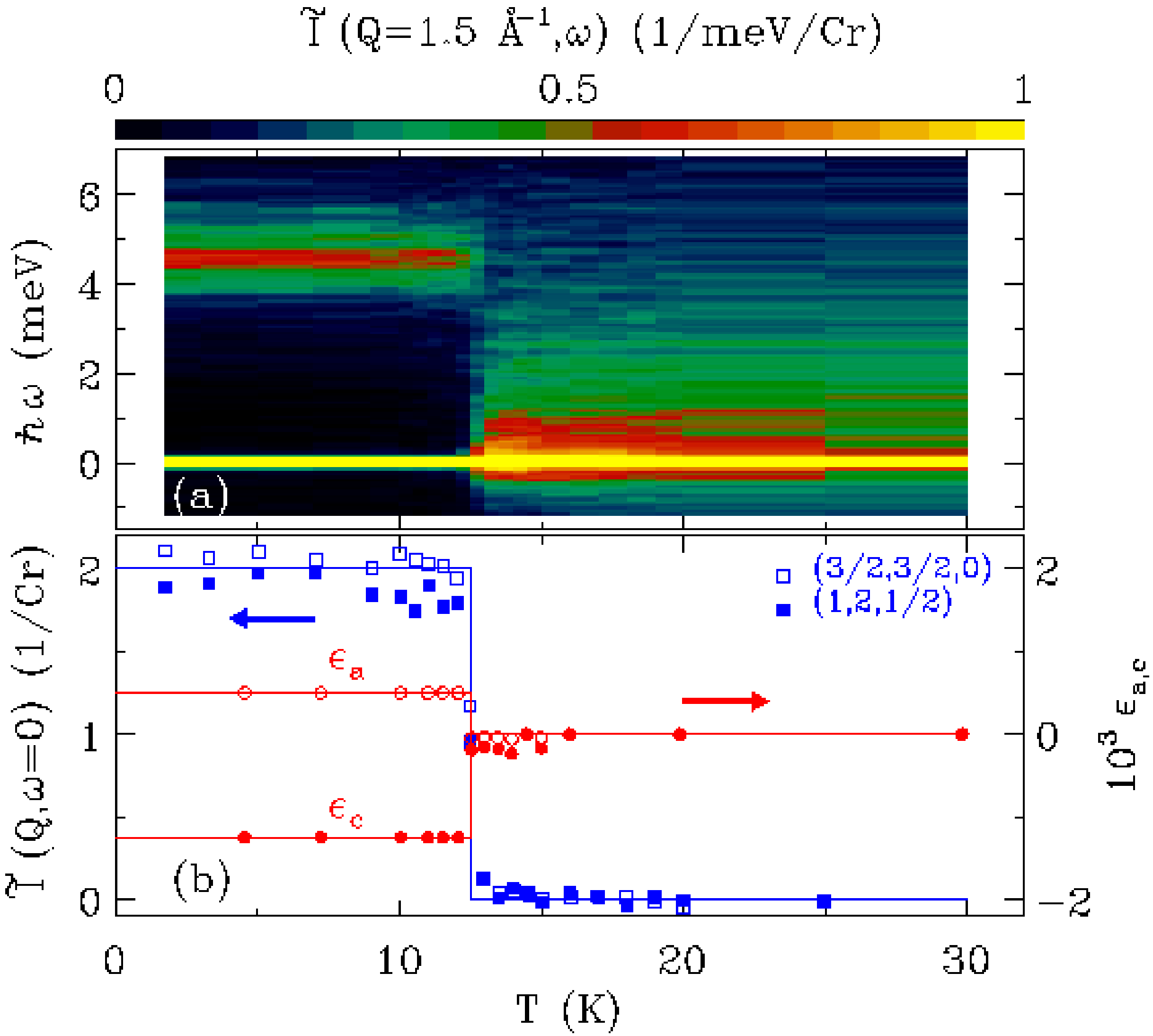
Near Quantum Critical spin system:

$$\Gamma_Q(T) = C\, k_B T \left(\frac{T}{\theta}\right)^{\alpha-1}$$

$$\chi_Q(T) = \frac{\mu_Q^2}{3k_B\theta}\frac{1}{1+T/\theta}$$

No indication of finite T cross over or phase transition in cubic phase

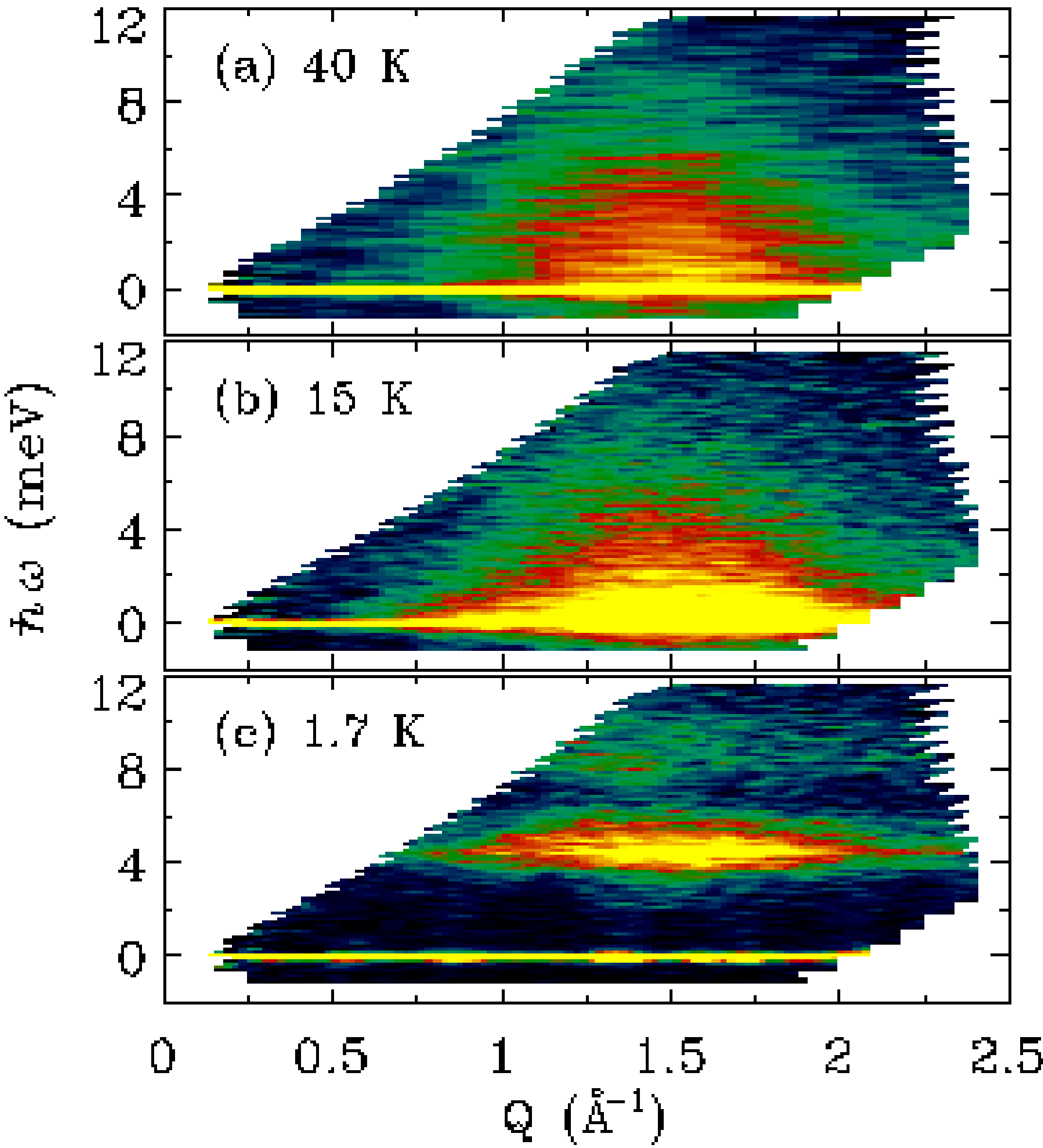
First order phase transition in $ZnCr_2O_4$



Dynamics:
- Low energy PM. Fluctuations form resonance

Statics:
- Staggered magnetization
- tetragonal distortion

Local spin resonance in ordered phase

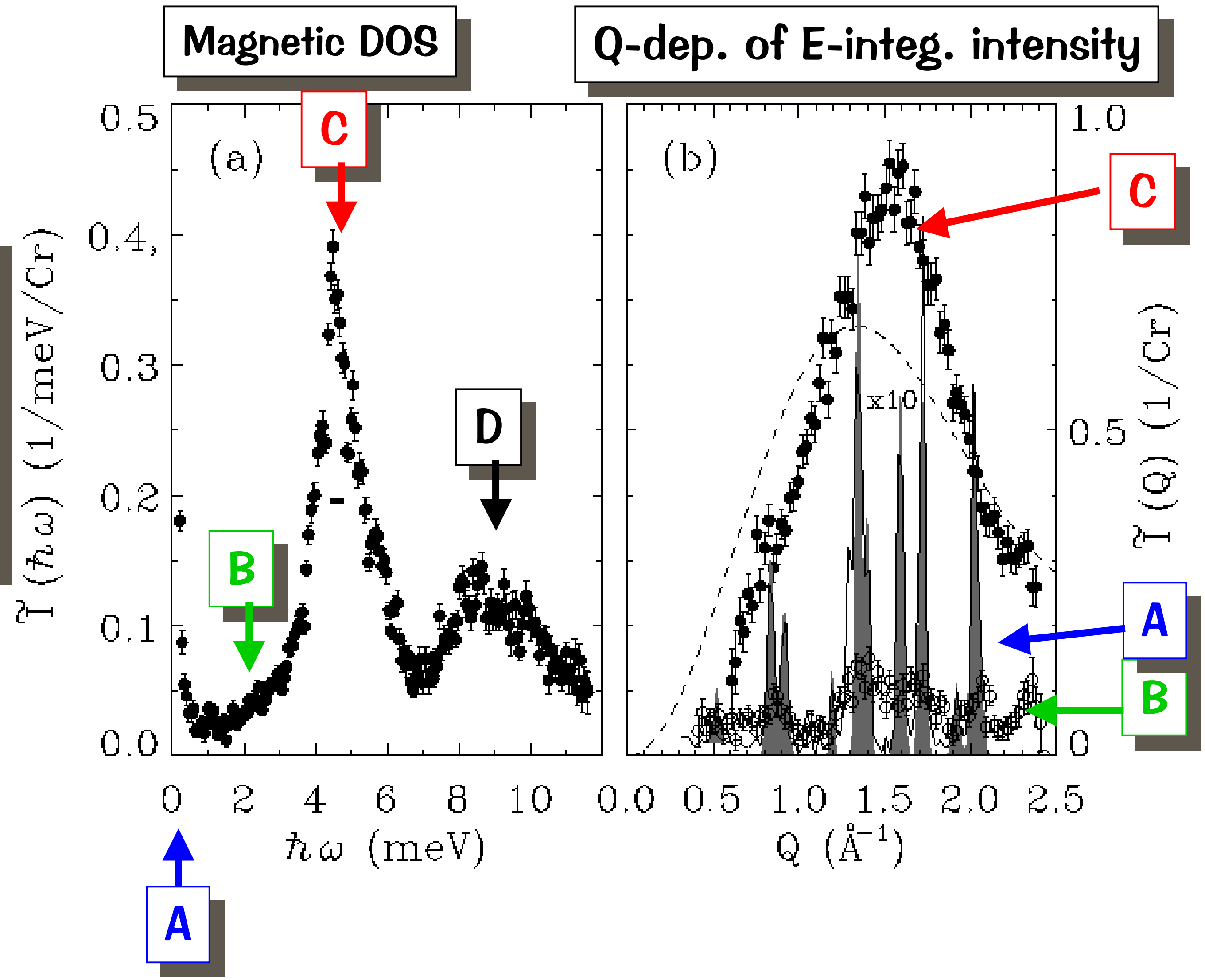


Paramagnetic fluctuations in frustrated AFM

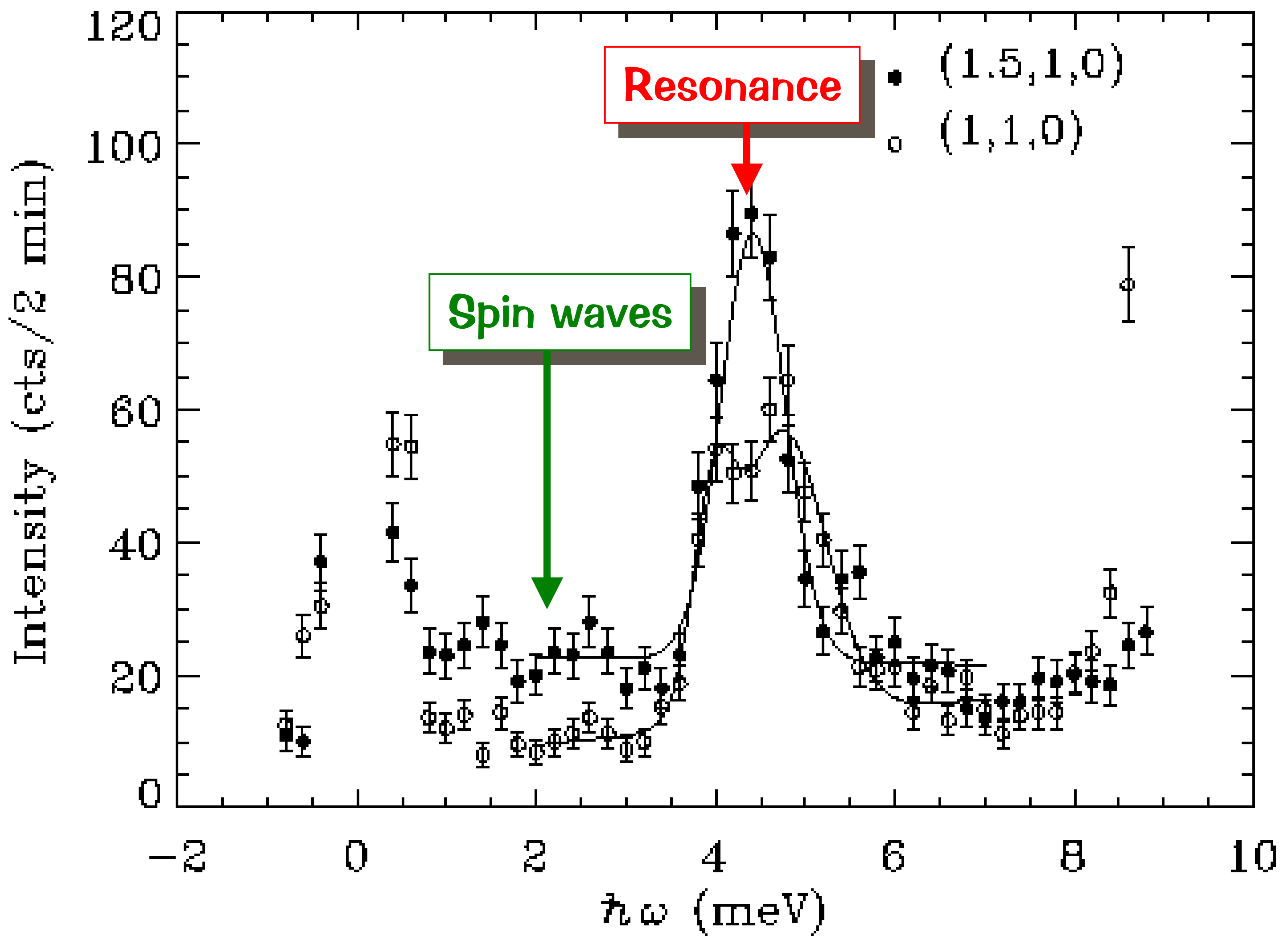
Local spin resonance in magneto-elastic LRO phase

Low T excitations in $ZnCr_2O_4$:

A: Bragg peaks
B: Spin waves
C: Resonance
D: Upper band

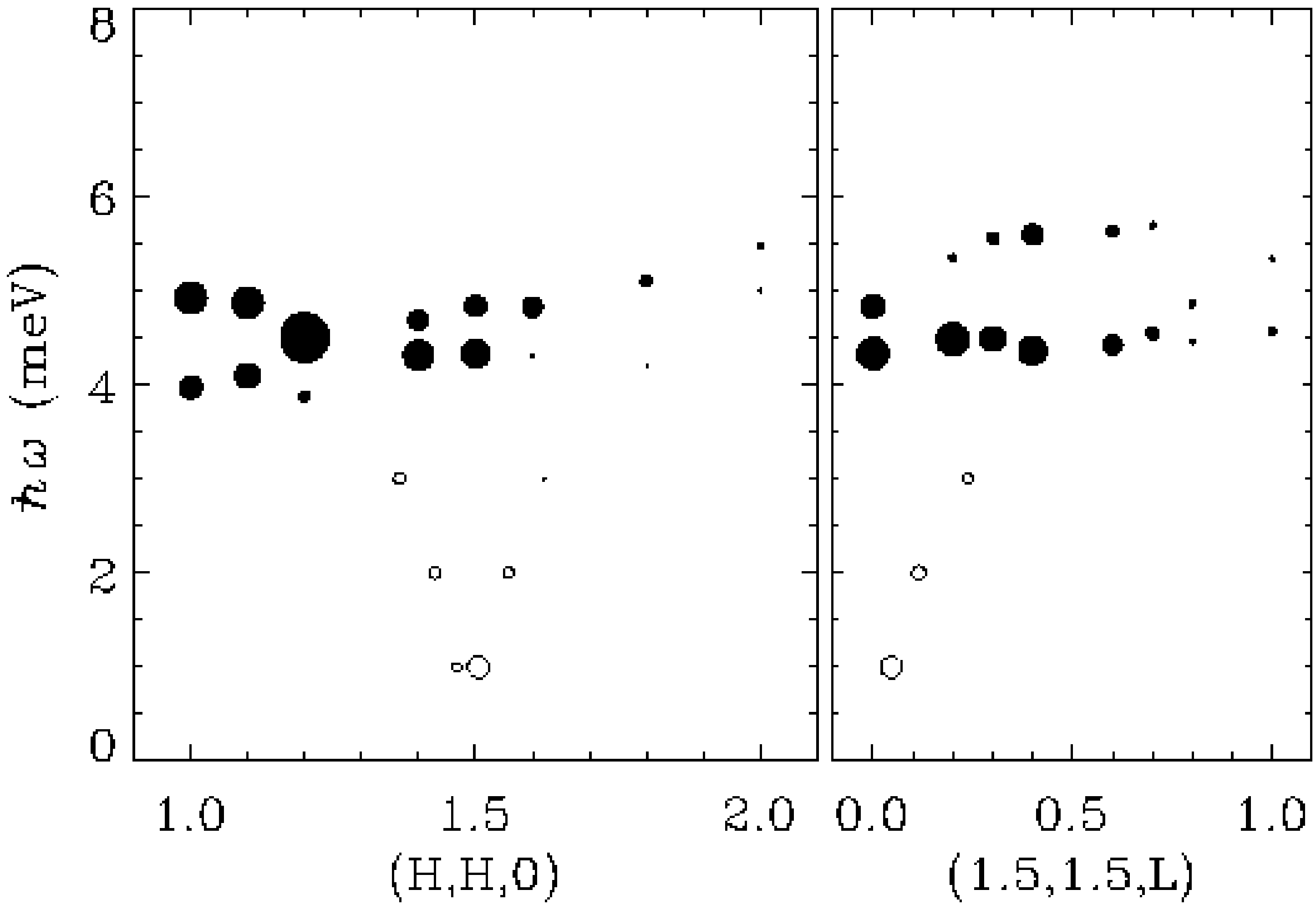


Spectra at specific $\vec{Q}$



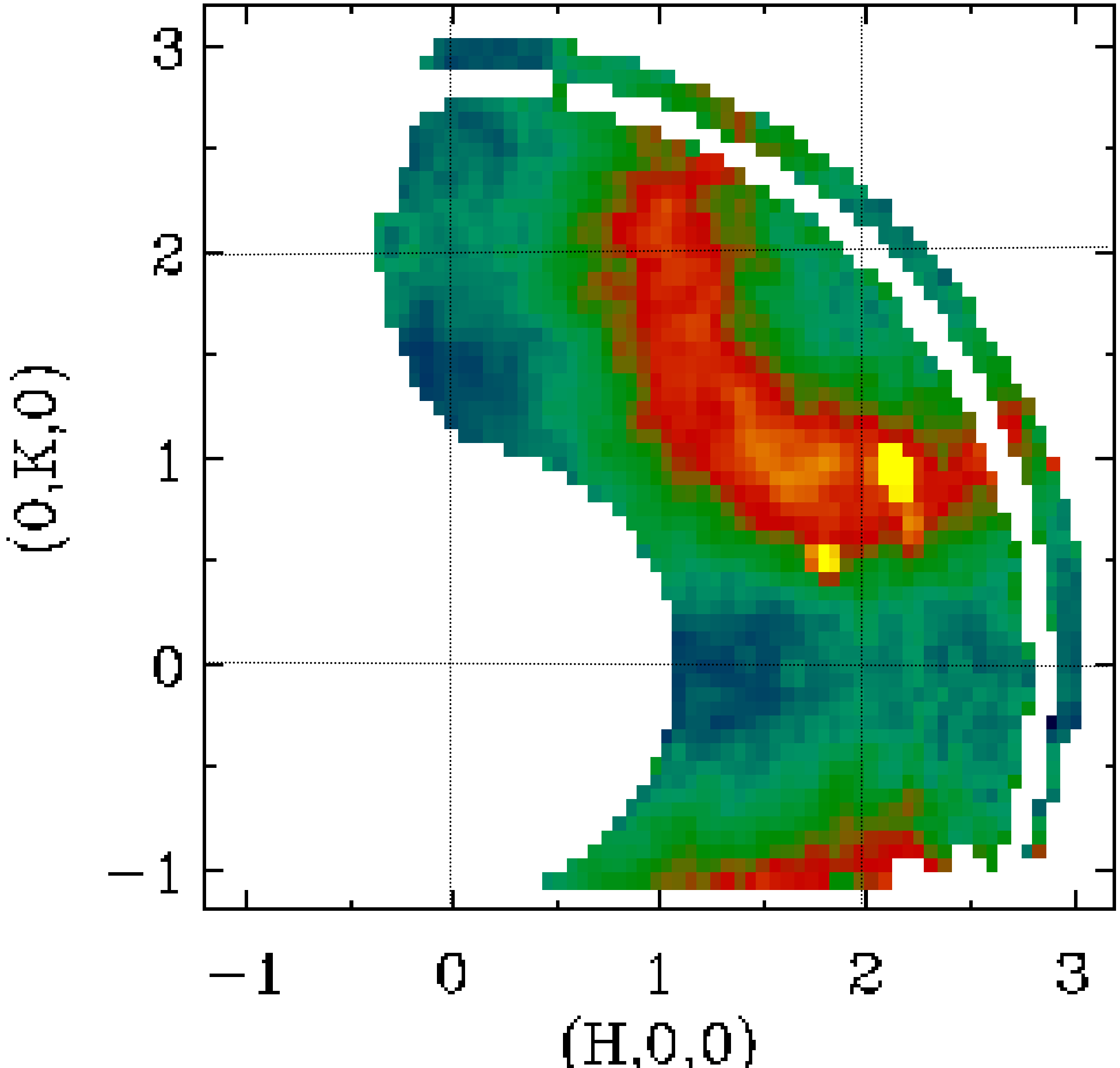
Dispersion relation for resonance

$ZnCr_2O_4$ single crystals T=1.5 K



Structure factor for resonance

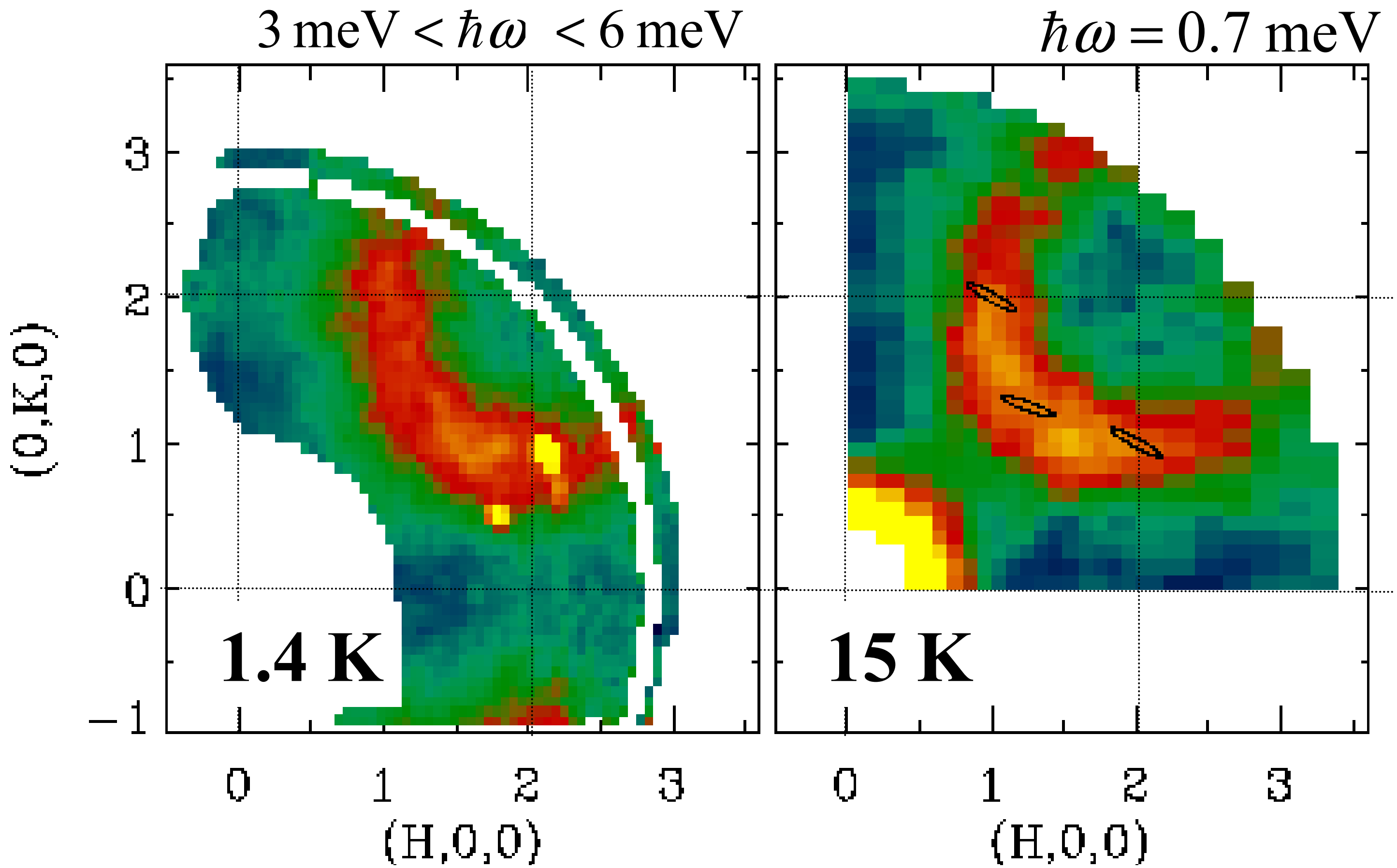
$ZnCr_2O_4$ T=1.4 K $3\,\text{meV} < \hbar\omega < 6\,\text{meV}$



Extended sharp structures in reciprocal space

Fluctuations satisfy local constraints

Comparing resonance and PM fluctuations

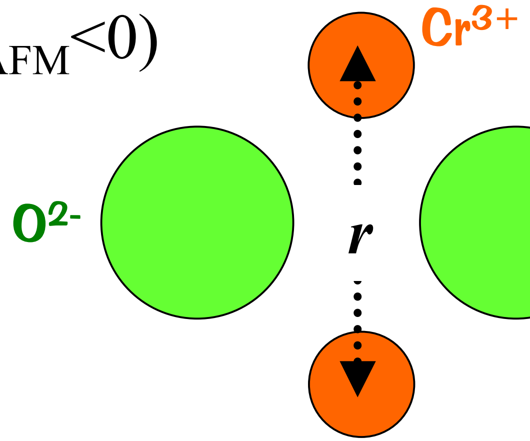


- Paramagnetic fluctuations and resonance satisfy same local constraints.
- Transition pushes low energy fluctuations into a resonance

How can this frustrated system order?

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

$(J_{AFM}<0)$

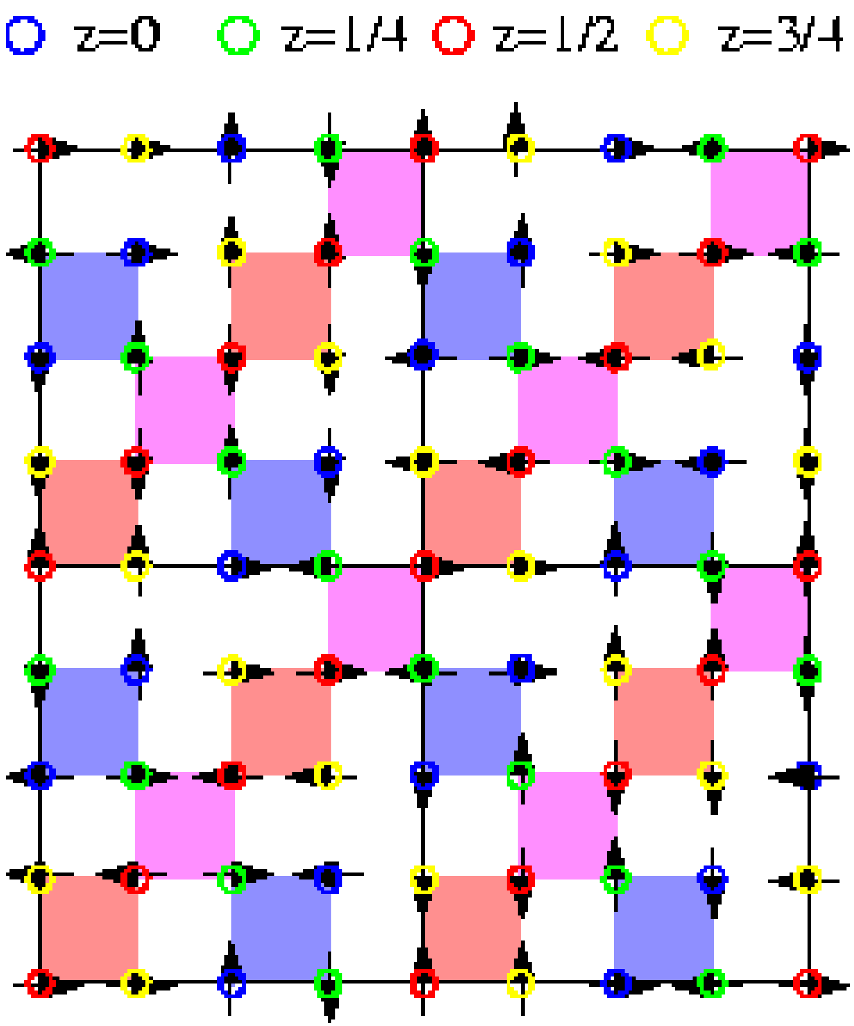


From series of Cr-compounds: $\frac{dJ}{dr} \approx 40 \text{ meV}/\mathring{A}$

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

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

Magnetic order in $ZnCr_2O_4$



-View along tetragonal c-axis

- tetrahedra have zero net moment
  ➡ this is a mean field ground state for cubic $ZnCr_2O_4$

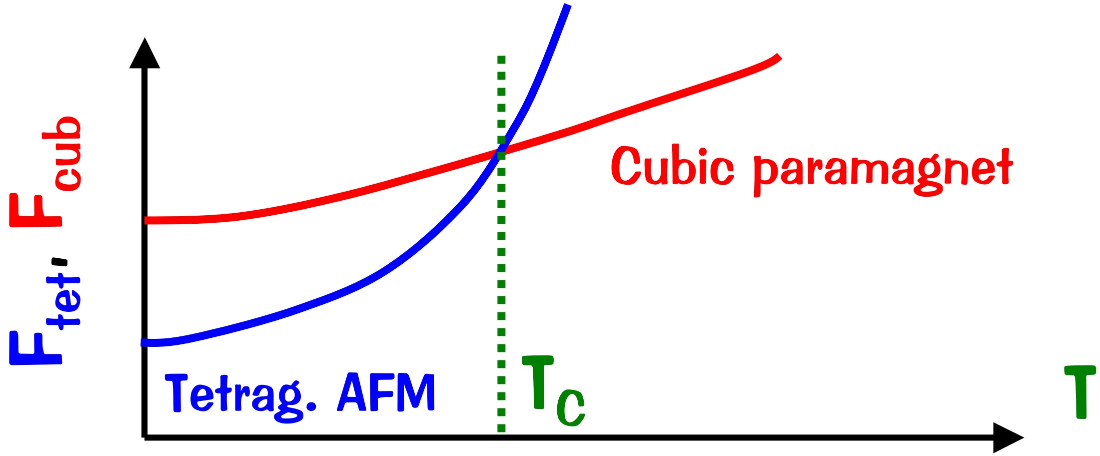
- Tetragonal distortion lowers energy of this state compared to other mean field ground states:

$$\Delta\langle H_S\rangle_{MF} = \frac{1}{2}\left(5\Delta J_{\parallel} - \Delta J_{\perp}\right) = -0.07\ \text{meV}$$

- In a strongly correlated magnet this shift may yield

$$k_B T_{Nt} >> \Delta\langle H_S\rangle_{MF}$$

Analysis of spin and lattice energies at T_C



From first moment sum-rule

$$\Delta\langle \mathcal{H}_s \rangle = -\tfrac{3}{2}\frac{\hbar^2 \int_0^\infty \omega\left(1-e^{\beta\hbar\omega}\right)\Delta S(Q\omega)}{1-\sin Qr_0 \big/ Qr_0} = -0.40(7)\ \mathrm{meV/Cr}$$

Based on scattering data above and below T_C and assuming that nearest neighbor exchange dominates

Change in lattice energy at T_C

Free energy of the two phases coincide at T_C

$$0 = \Delta F = \Delta\langle \mathcal{H}_s \rangle + \Delta\langle \mathcal{H}_l \rangle - T_c \Delta S$$

From this we derive increase in lattice energy at transition

$$\begin{array}{lcrl} & T_C \Delta S & = & -0.16 \quad \text{meV/Cr} \\ - & \Delta\langle \mathcal{H}_s \rangle & = & -0.40(7)\ \text{meV/Cr} \\ \hline & \Delta\langle \mathcal{H}_l \rangle & = & 0.24(7)\ \text{meV/Cr} \end{array}$$

Compare to tetragonal strain energy

$$\Delta\langle \mathcal{H}_l \rangle\big|_t = v_0\left(\tfrac{1}{2}c_{11}\left(\varepsilon_c^2 + 2\varepsilon_a^2\right) + c_{12}\left(\varepsilon_a^2 + 2\varepsilon_a\varepsilon_c\right)\right) = 0.026\ \text{meV/Cr}$$

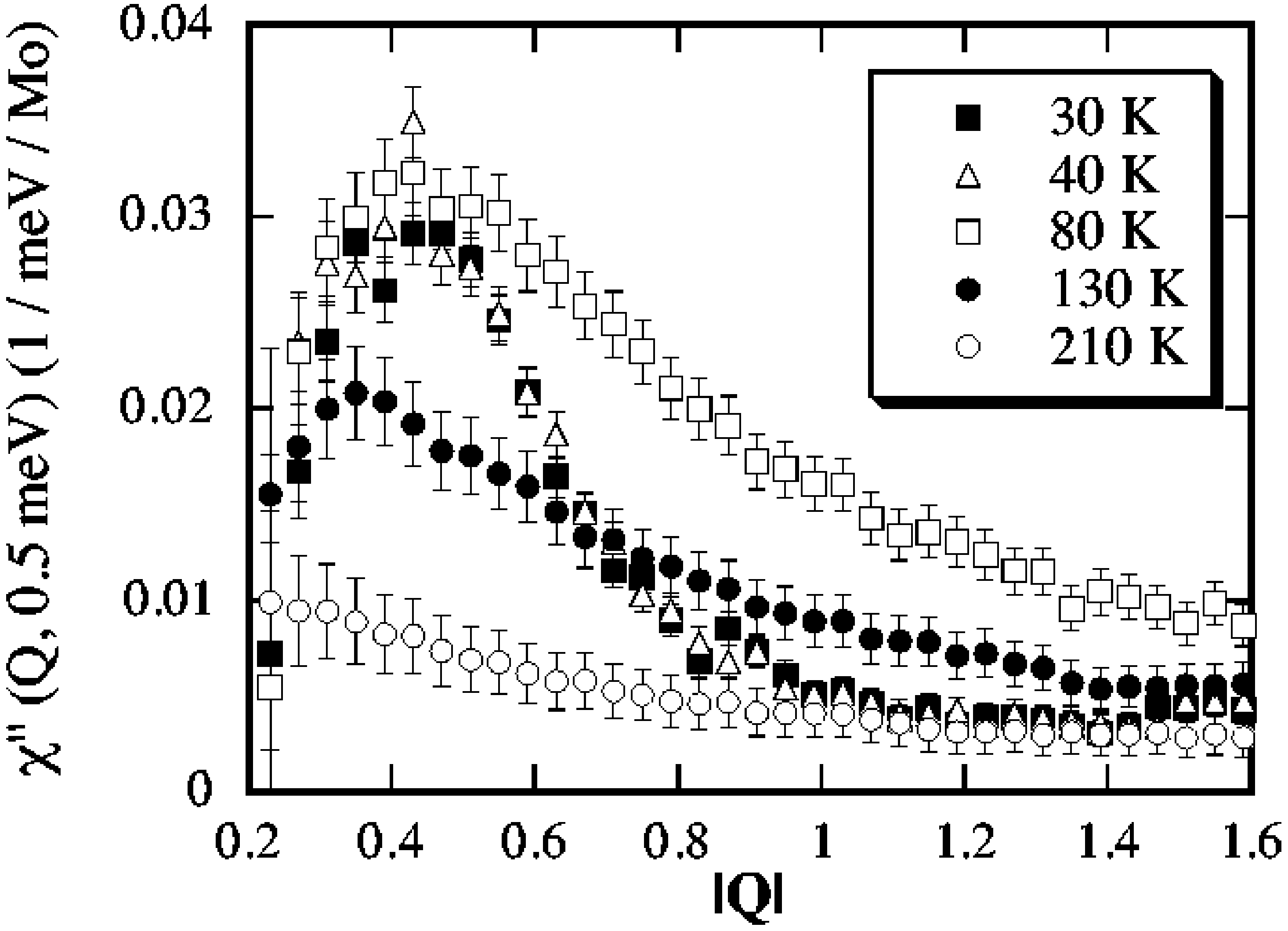
Discrepancy calls for additional lattice changes at T_C

Analogies with Spin Peirls transition?

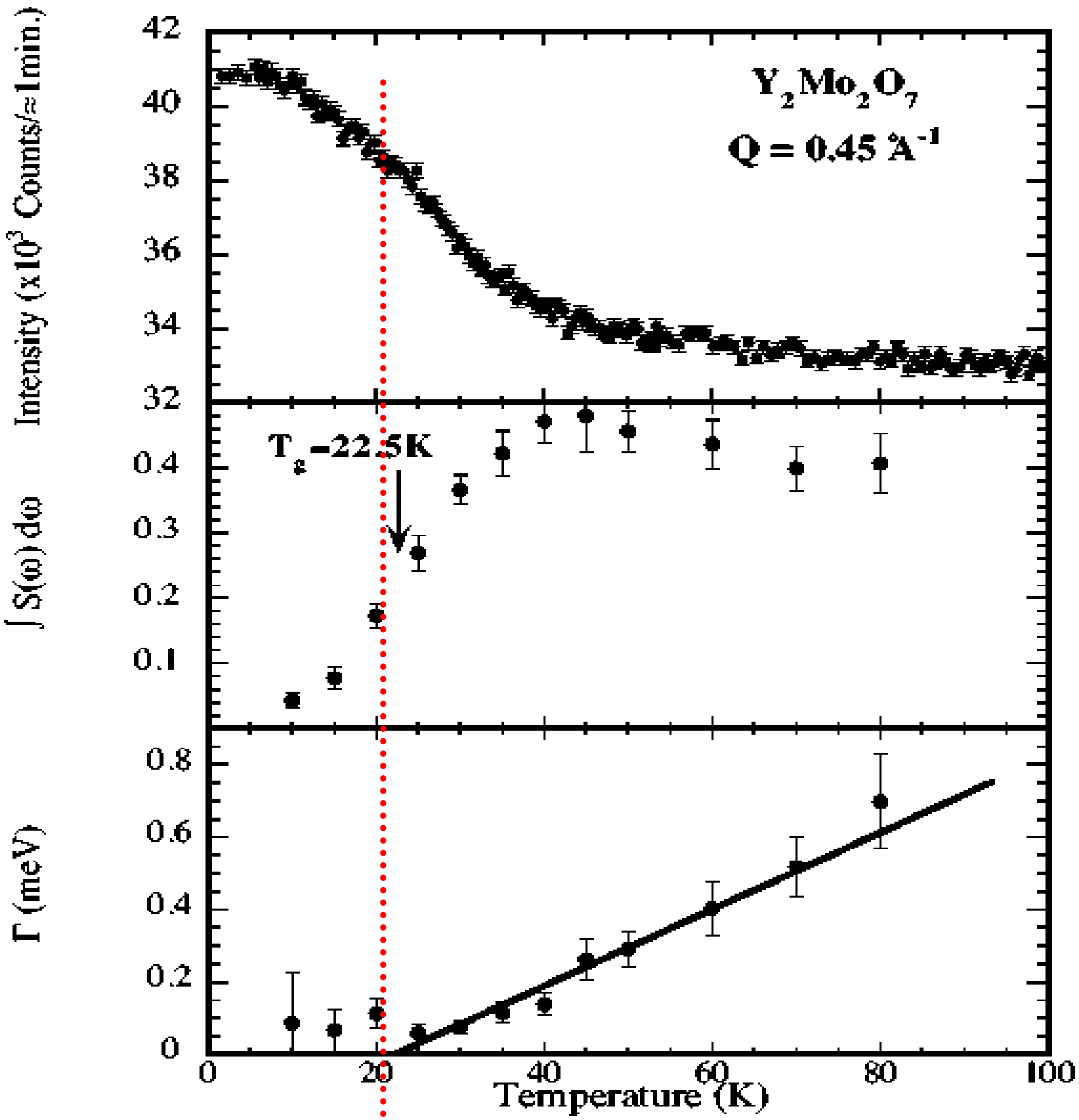
	Spin-Peirls	$ZnCr_2O_4$
Quantum critical above T_c	yes	yes
Order suppressed to $\|T/\Theta_{cw}\|<<1$ due to	low D	frustra-tion
Change of lattice symmetry at T_c enables lower energy spin state	yes	yes
Low energy magnetic spectral weight is pushed into resonance	yes	yes
Order of phase transition	second	first
Low T phase	isolated singlet	Neel LRO
$T_c \Delta S$ is significant energy scale	no	yes

There are similarities as well as important distinctions!

Short range correlations in $Y_2Mo_2O_7$



Spin-glass transition in $Y_2Mo_2O_7$



Elastic scattering intensity:

- Development of spin correlations static on the 50 ps time-scale of the experiment.

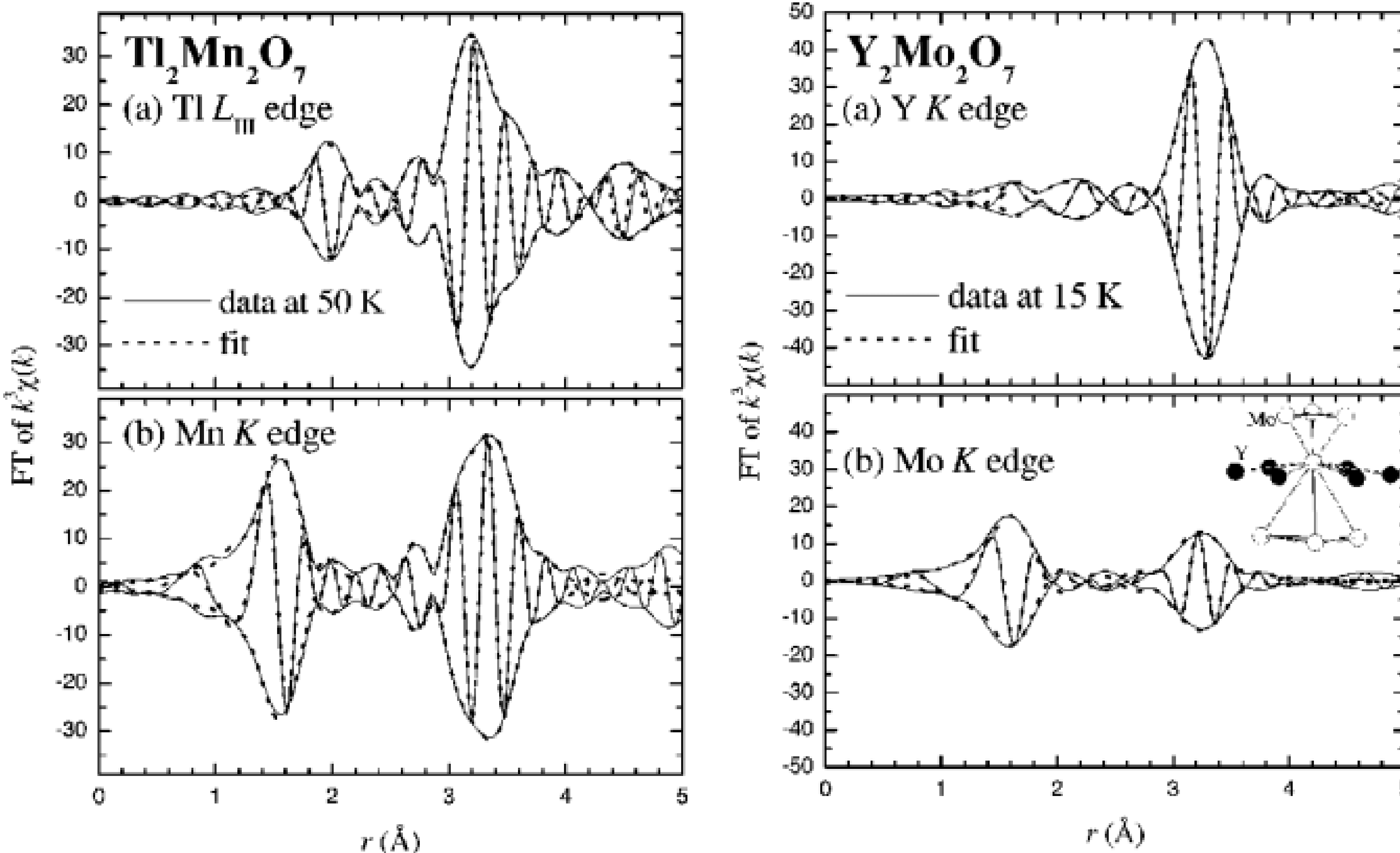
Inelastic scattering intensity:

- Inelastic scattering decreases as spins cease to fluctuate.

Spin relaxation rate:

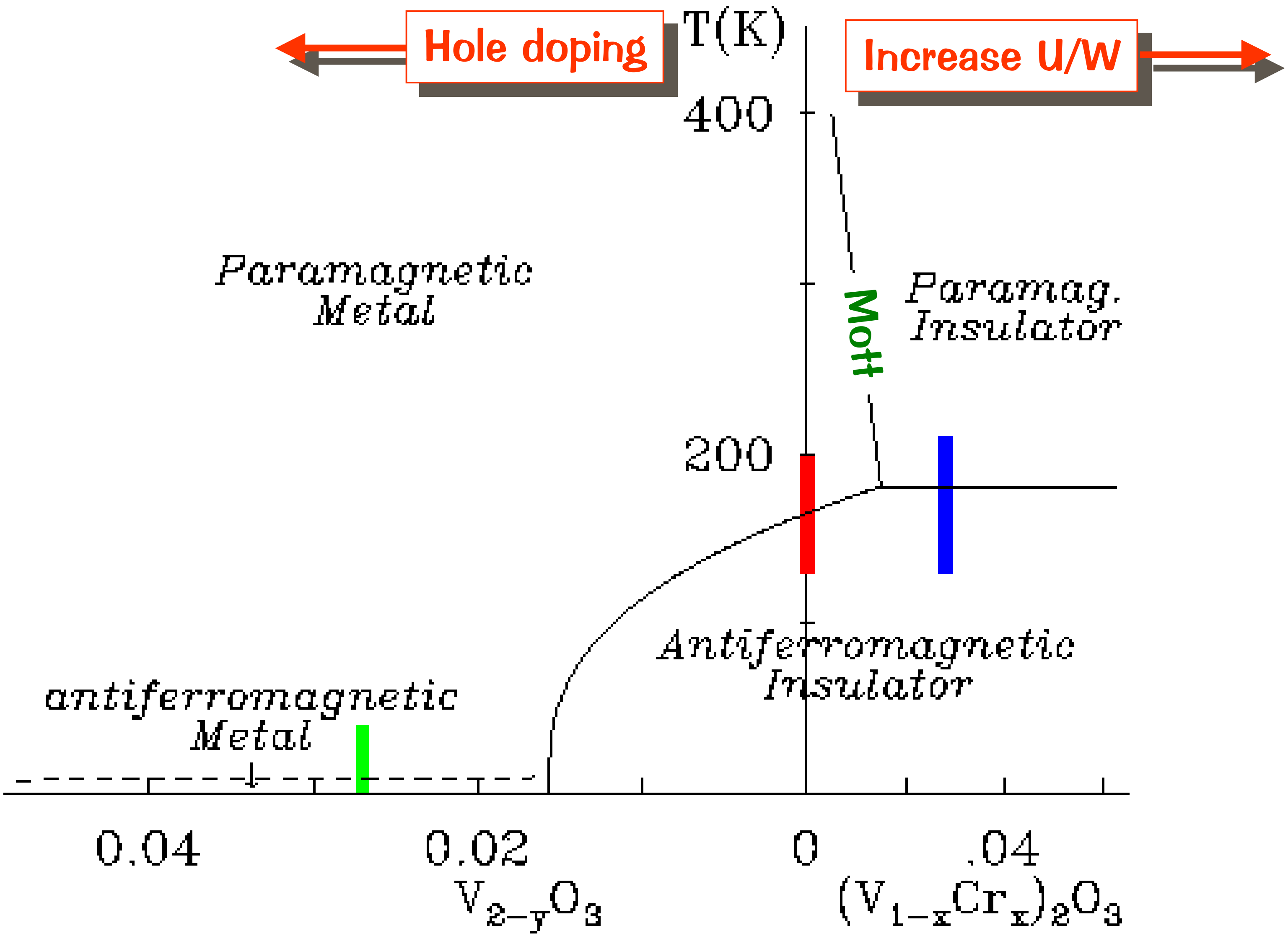
- $\Gamma(T)$ decreases linearly with T and extrapolates to T_g=23 K derived from AC-susceptibility

EXAFS Evidence for disorder in $Y_2Mo_2O_7$



Booth et al (2000)

Metal Insulator transition in V_2O_3



McWhan et al. (1973); Y. Ueda et al. (1980)

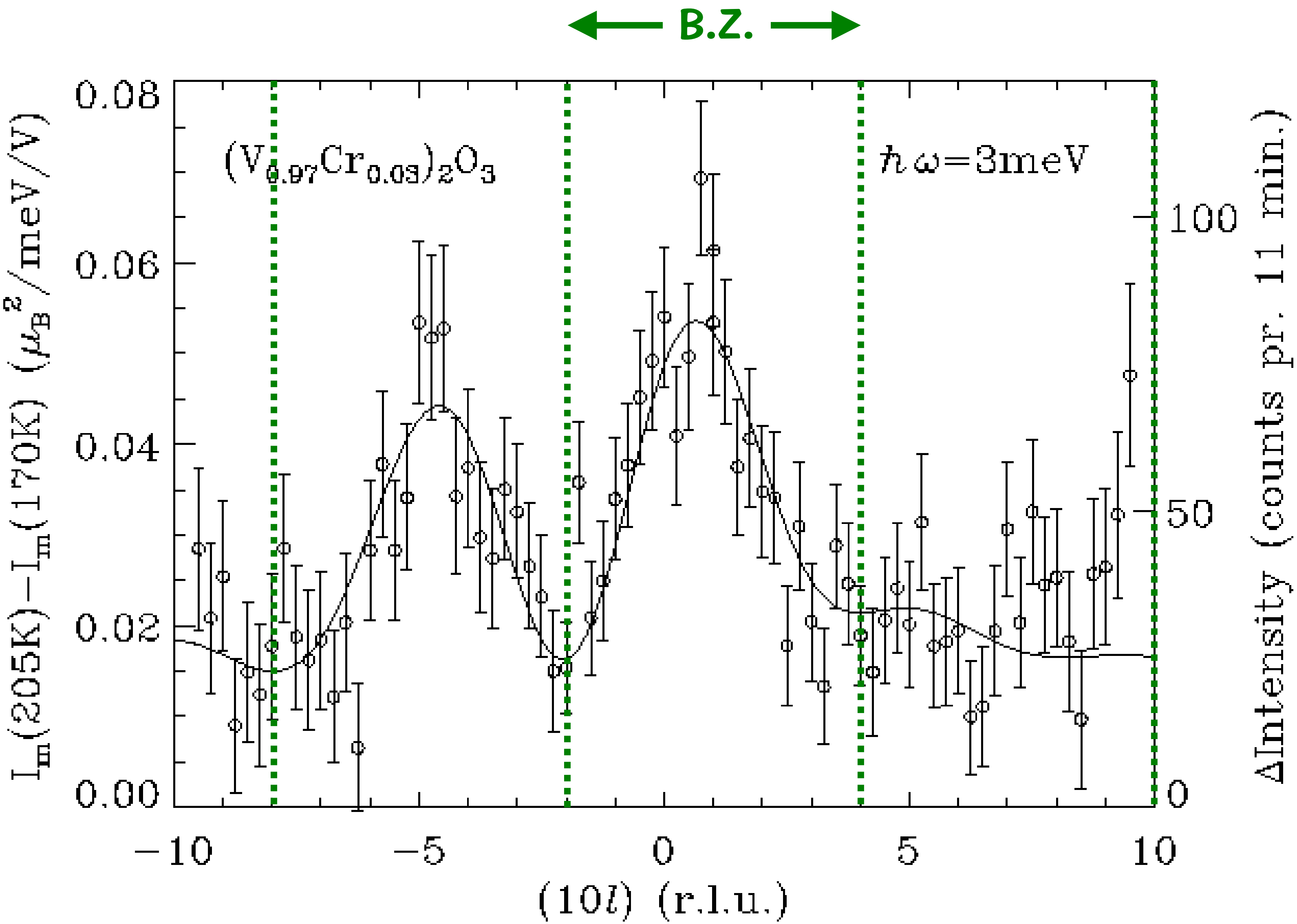
The World according to Bill Gates

Mott, Sir Nevill Francis

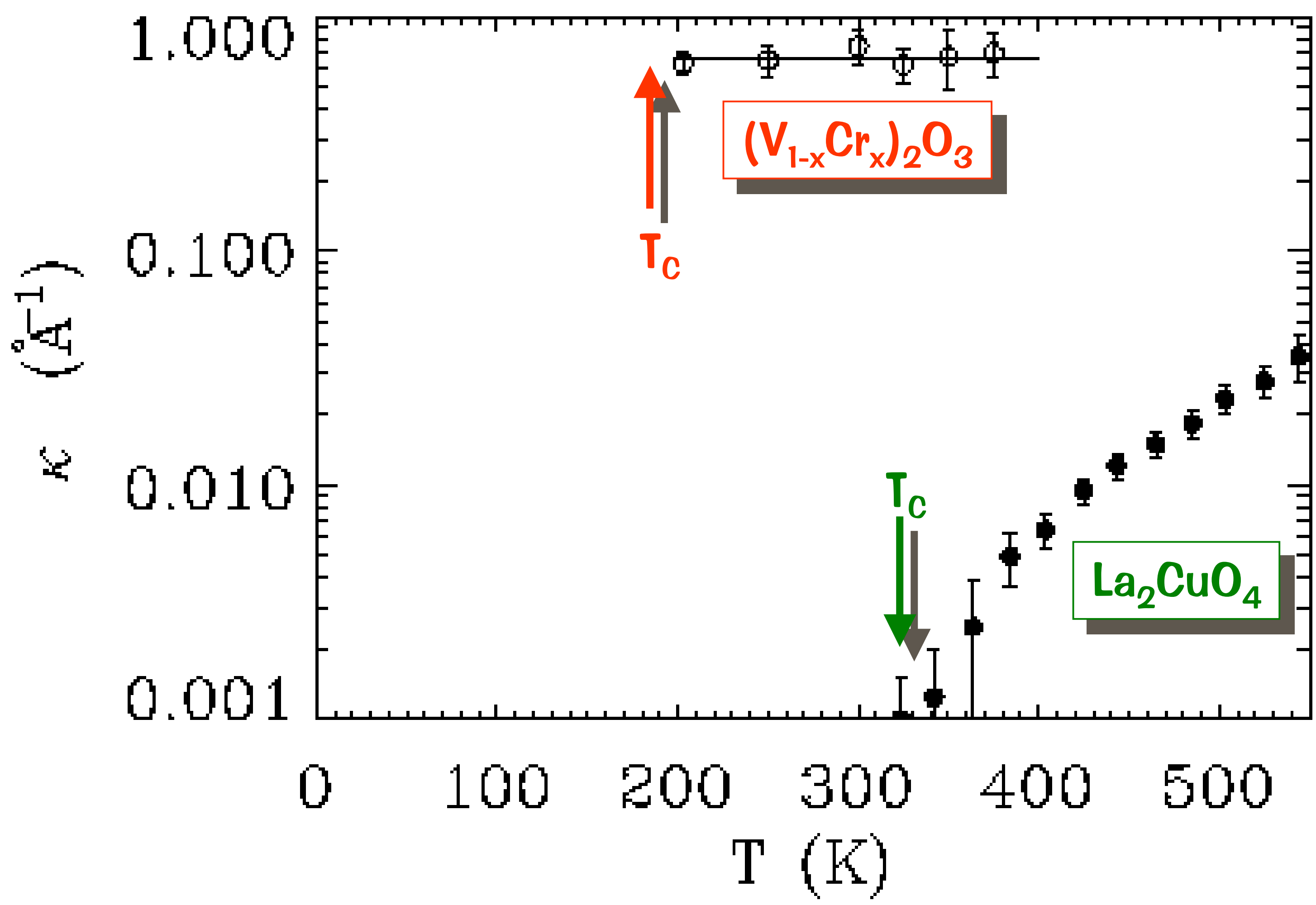
Mott (m⇩t), Sir **Nevill Francis**
Born 1905
British physicist. He shared a 1977 Nobel Prize **for developments in computer memory.**

- from Microsoft Bookshelf Basics circulation >50,000,000?
on every Windows 98 based PC with Office

Short Range order in Paramagnetic Insulator



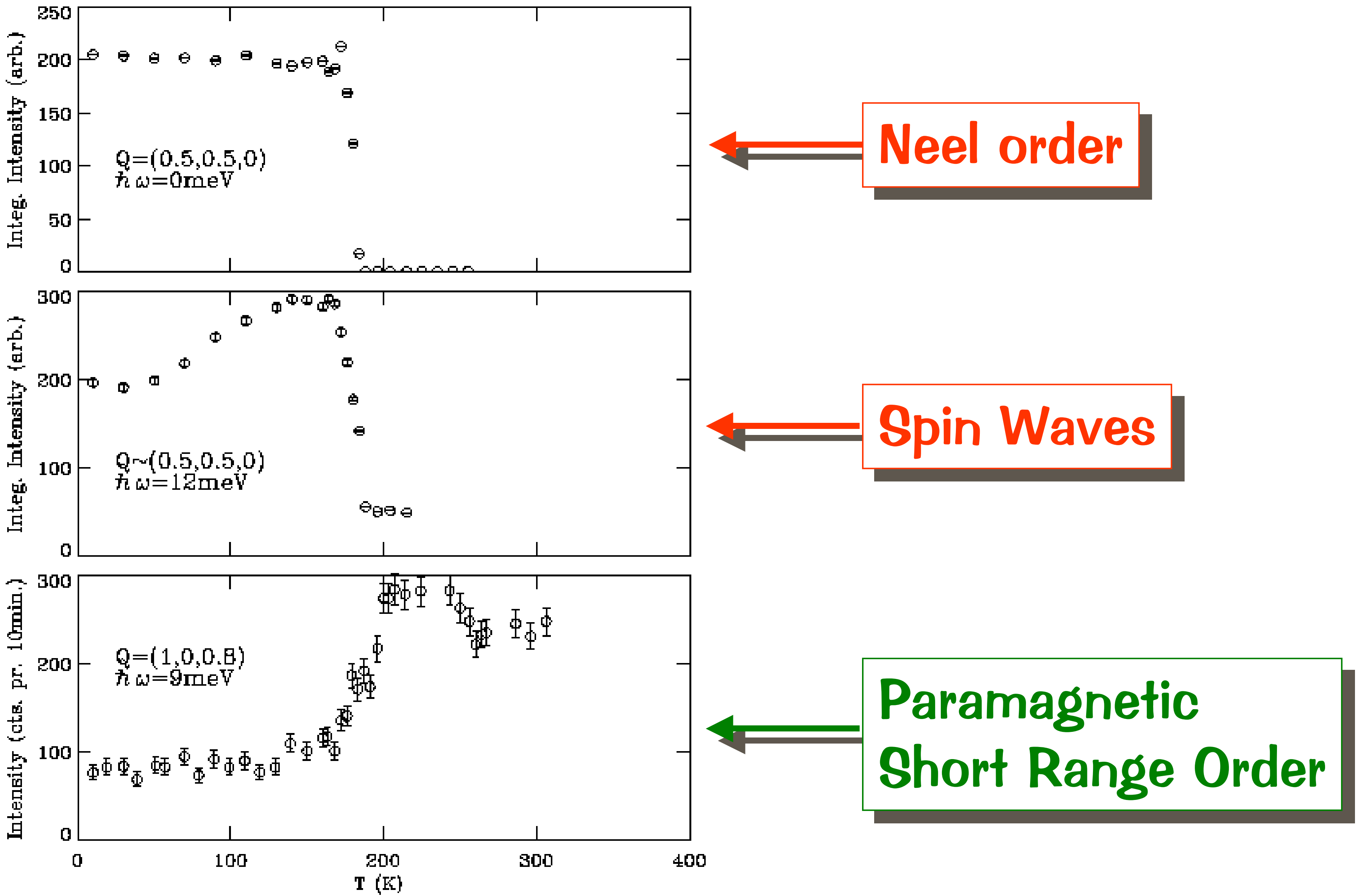
Correlations in $(V_{1-x}Cr_x)_2O_3$ and La_2CuO_4



Bao et al.

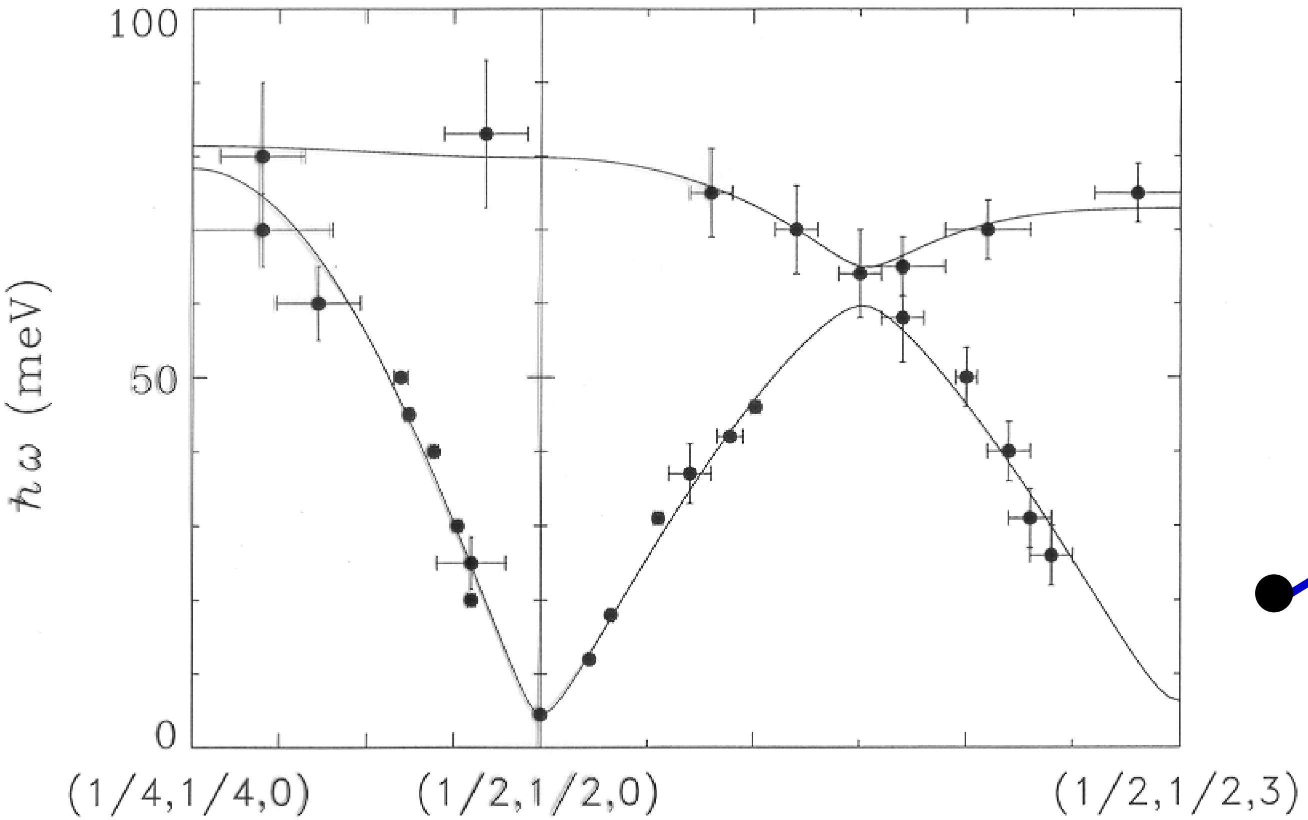
Keimer et al.

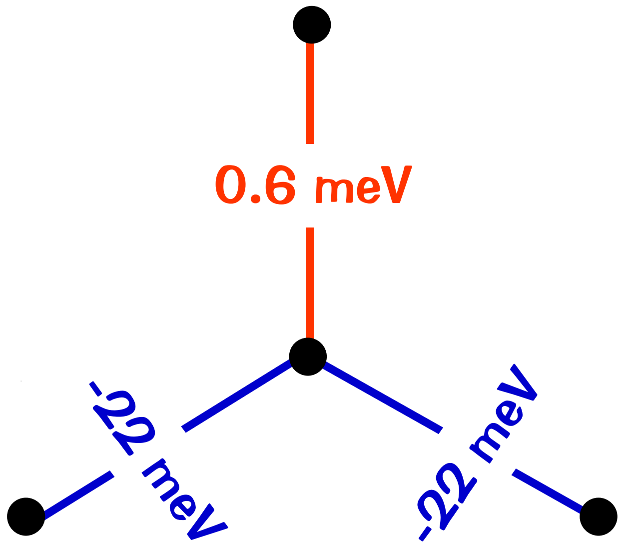
First order phase transition in $(V_{1-x}Cr_x)_2O_3$



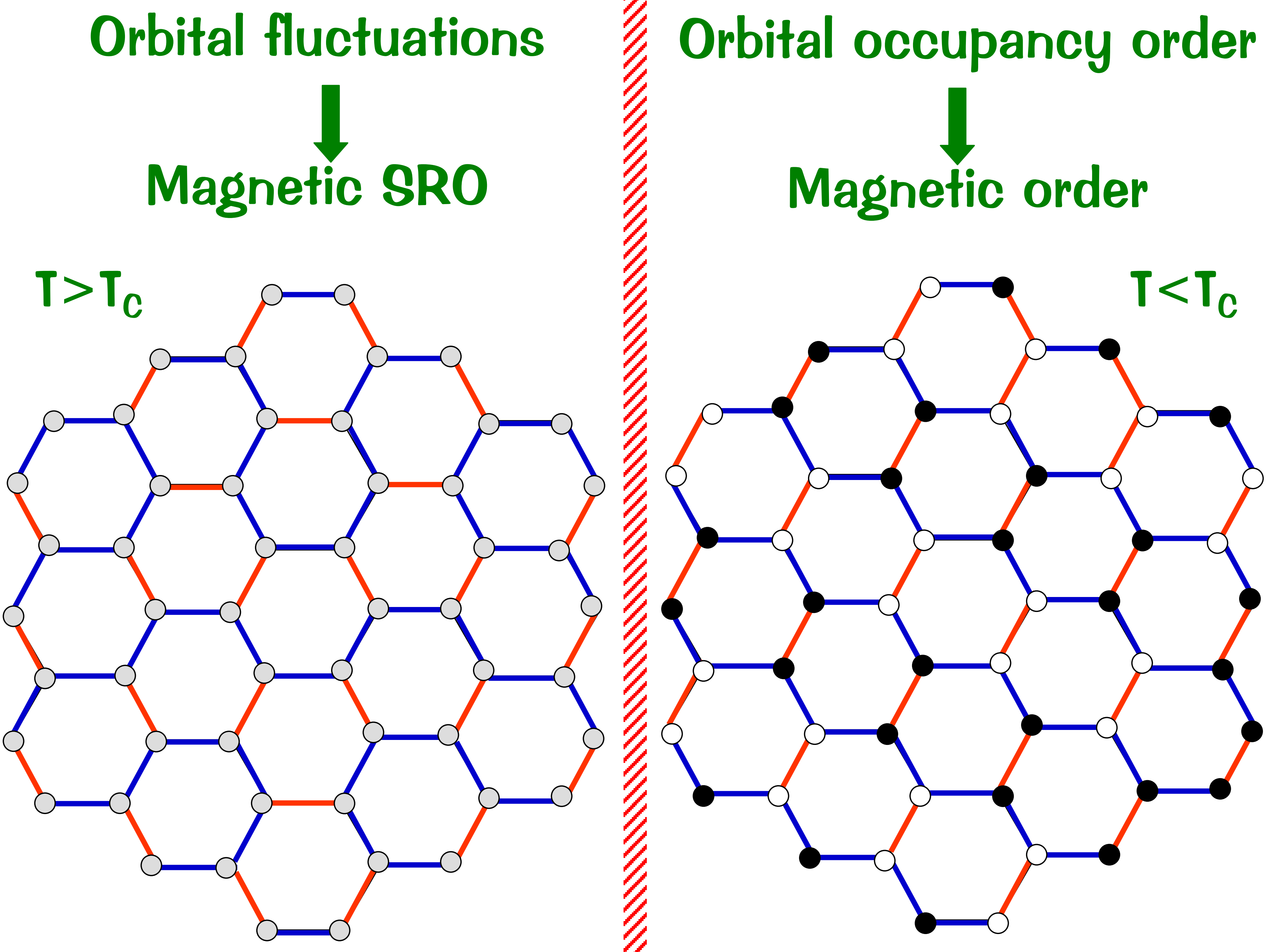
NEC

Spin wave dispersion ➡ Exchange constants



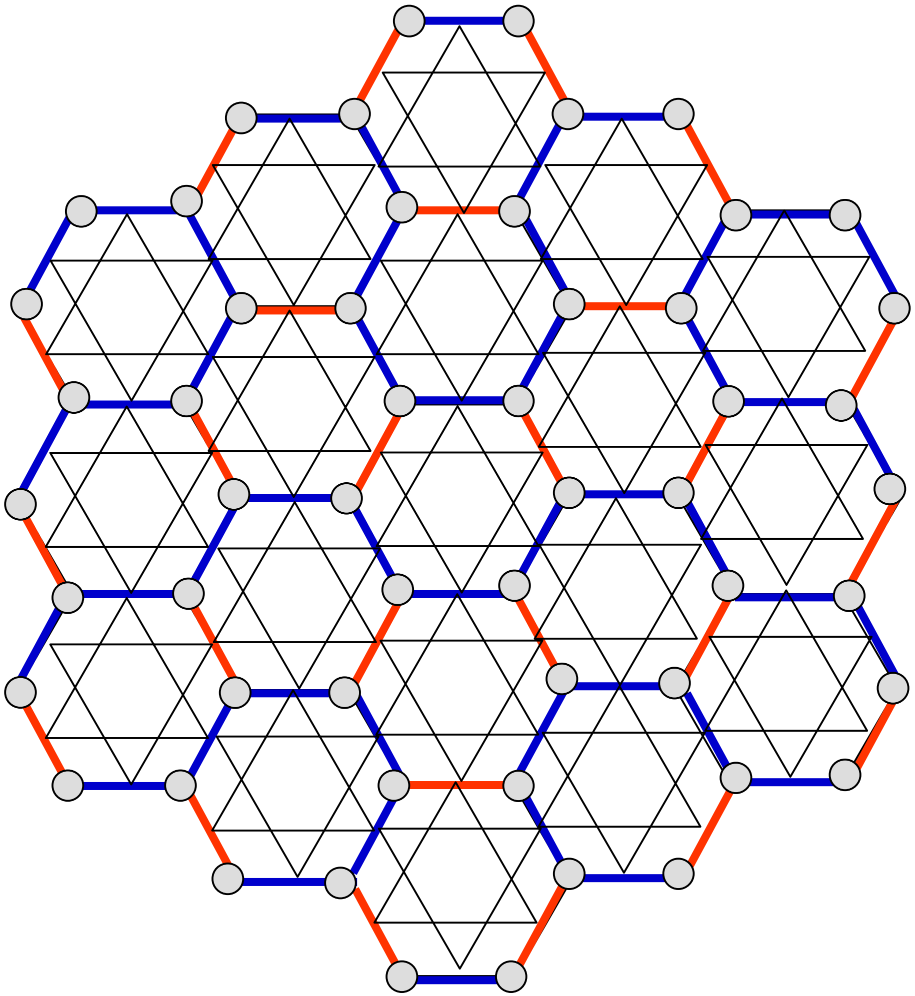


Bao et al. Unpublished (2000)



Orbital fluctuations
Magnetic SRO
T>Tc
Orbital occupancy order
Magnetic order
T<Tc

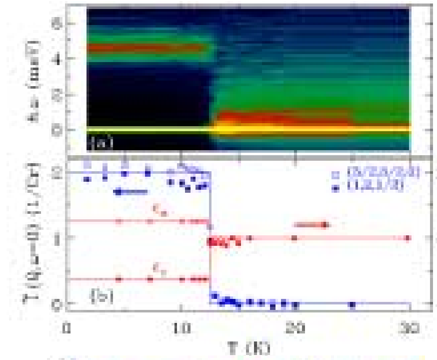
Why orbital fluctuations at low T ?



An interesting possibility:

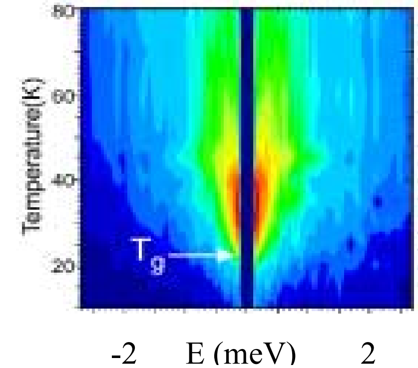
- Bonds occupy kagome' lattice
- Ising model on kagome' lattice has no phase transition whence the low T_C
- Orbital occupational order occurs to lower energy of spin system

Conclusions



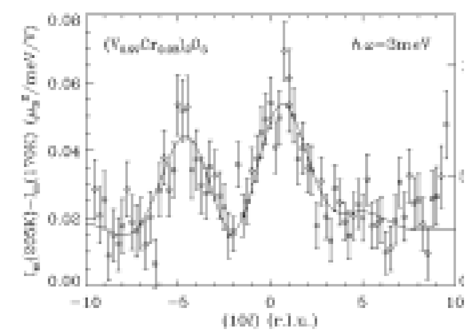
$ZnCr_2O_4$

- Quantum critical fluctuations in PM phase
- strain relieves frustration, enables Neel order



$Y_2Mo_2O_7$

- Short range order at all temperatures $T<|\Theta_{CW}|$
- spin freezing due to quenched lattice disorder ?



$(V_{1-x}Cr_x)_2O_3$

- MIT visible due to orbital occupational frustration
- Short range order due to orbital fluctuations
- Orbital occupational order enables spin order

Is there a Jahn-Teller like theorem for quantum critical magnets?