Impurities and finite temperature effects in a one-dimensional spin-1 antiferromagnet

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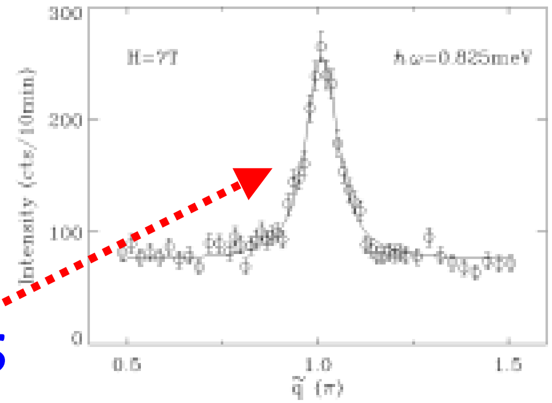
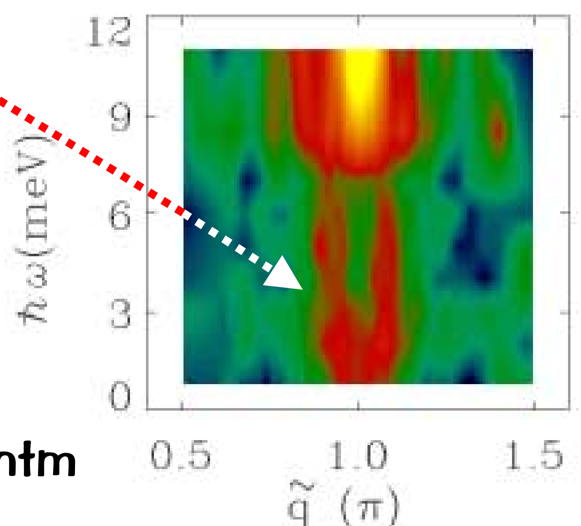
- Coherent excitations in Y_2BaNiO_5
- Loss of coherence for T>0
- Chain-end spins in $Y_2BaNi_{1-x}Mg_xO_5$
- AFM droplets in $Y_{2-x}Ca_xBaNiO_5$
- Conclusion





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Talk at http://www.pha.jhu.edu/~broholm/nhmfl/index.htm

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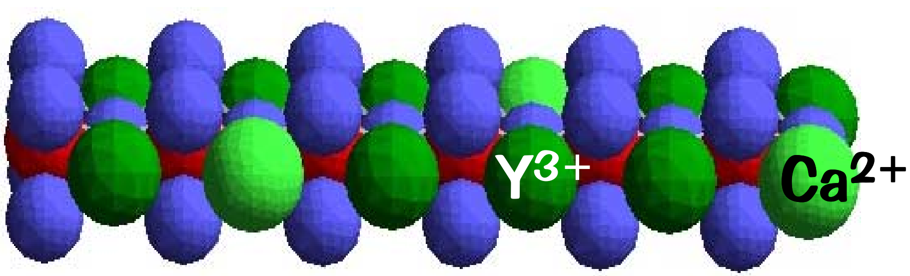
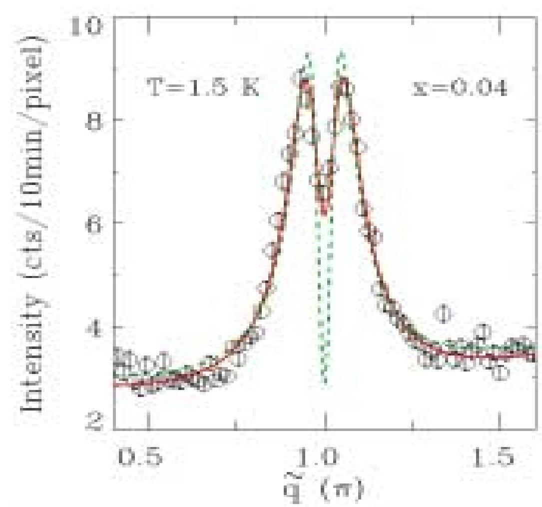
Why study quantum magnets ?

- Coherent many body wave functions are fascinating and useful
  - Laser beams
  - Superconductivity
  - Fractional Quantum Hall effect
  - Bose Condensation
  - Quantum magnets without static order at T=0
- Each phenomenon provides different experimental info about macroscopic quantum coherence
- Only in quantum magnets are dynamic correlations directly accessible (through neutron scattering)

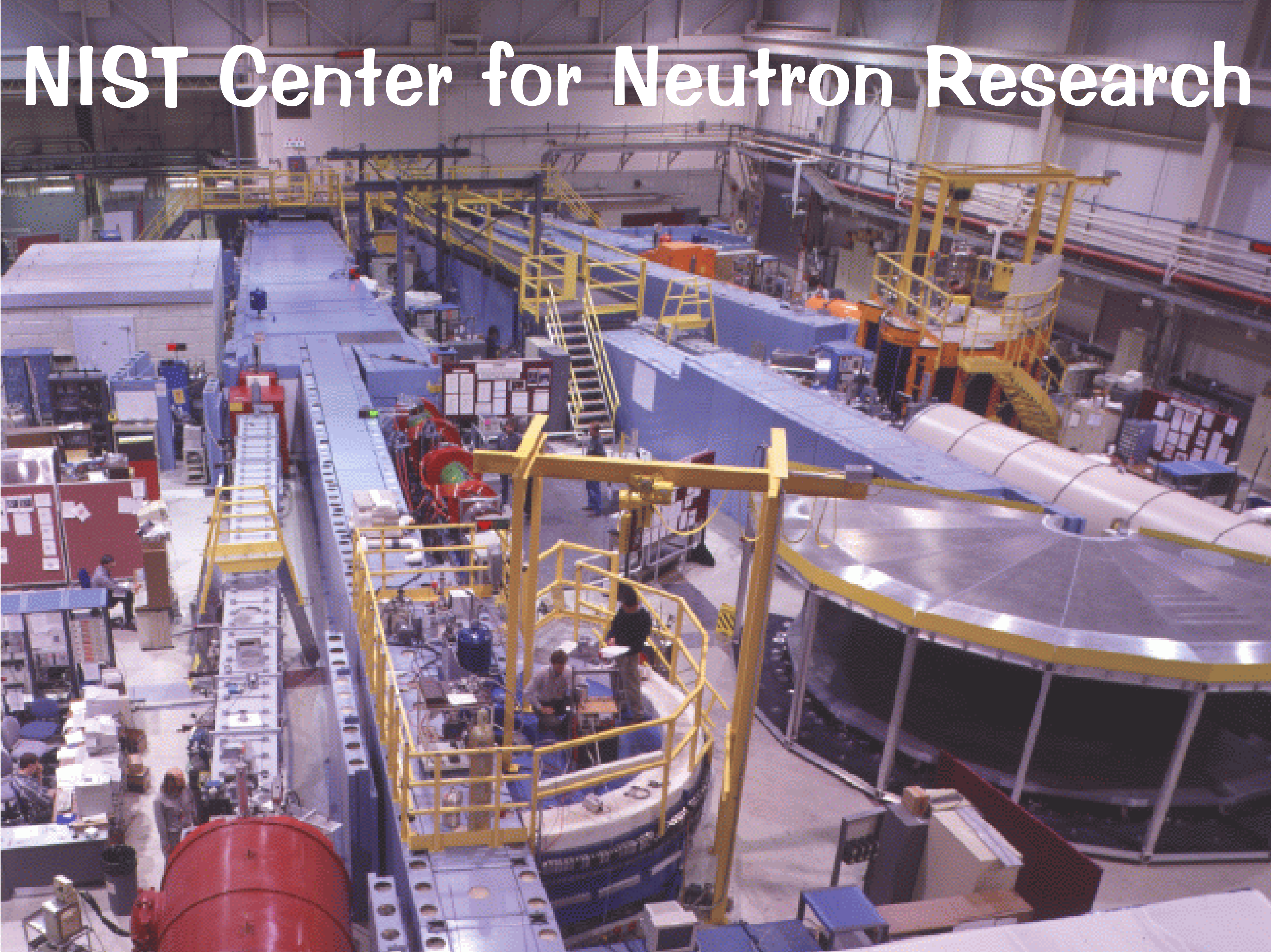
Why study impurities in quantum magnets ?

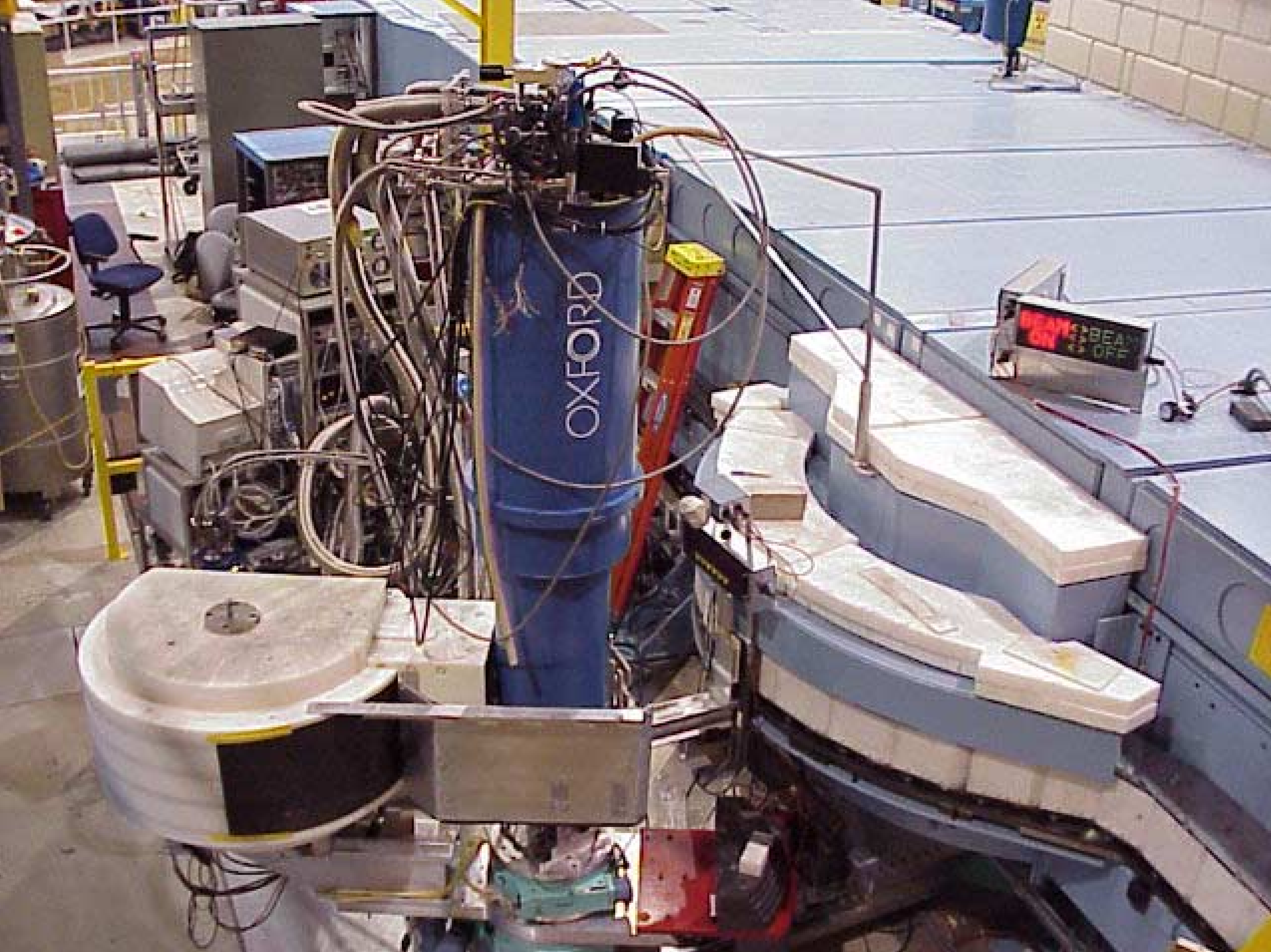
- Impurities are inevitable or even necessary to produce coherence
- Probing the response to impurities reveals the building blocks of a macroscopic quantum state.
- Impurities in quantum magnets can be explored at the microscopic level.





NIST Center for Neutron Research

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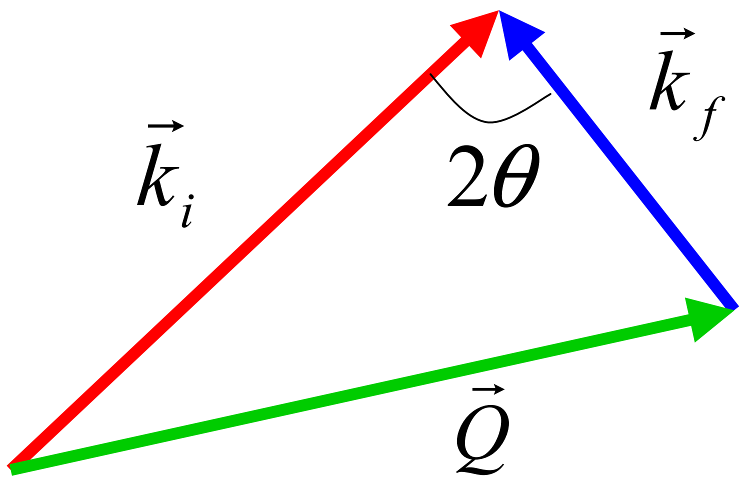
Magnetic Neutron Scattering



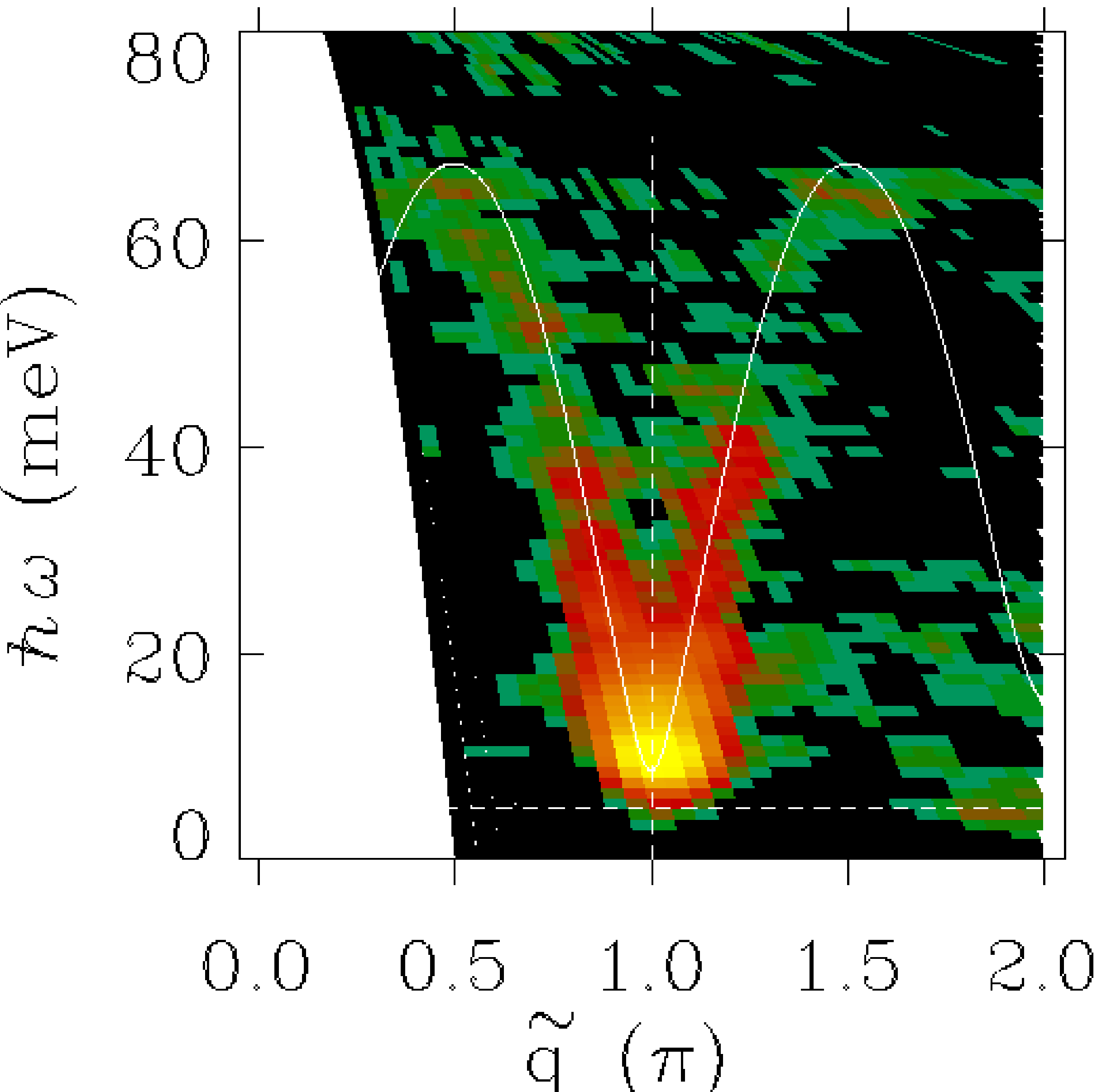
$$\vec{Q} = \vec{k}_i - \vec{k}_f$$

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

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

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

Low T excitations in pure spin-1 AFM chain



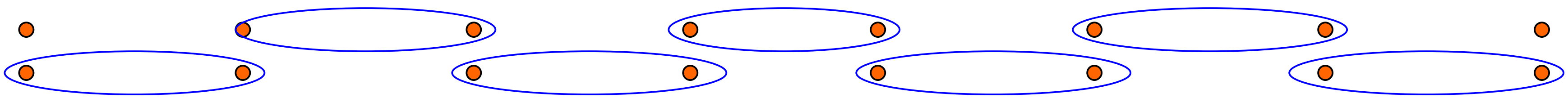
Y_2BaNiO_5 T=10 K
MARI chain ⊥ k_i

Points of interest:

- Haldane gap Δ=8 meV
- Coherent mode
- $S(q,\omega)$->0 for Q->2nπ

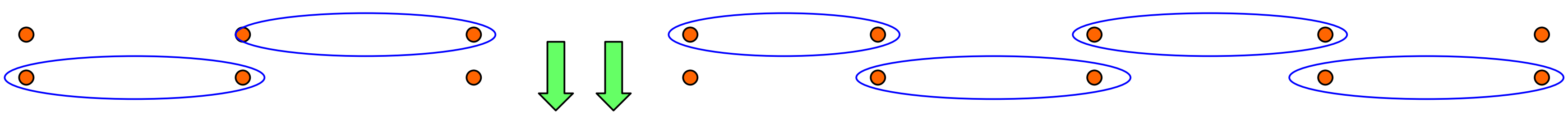
Why does spin-1 AFM have a spin gap?

- Magnets with 2S=nz have a nearest neighbor singlet covering.



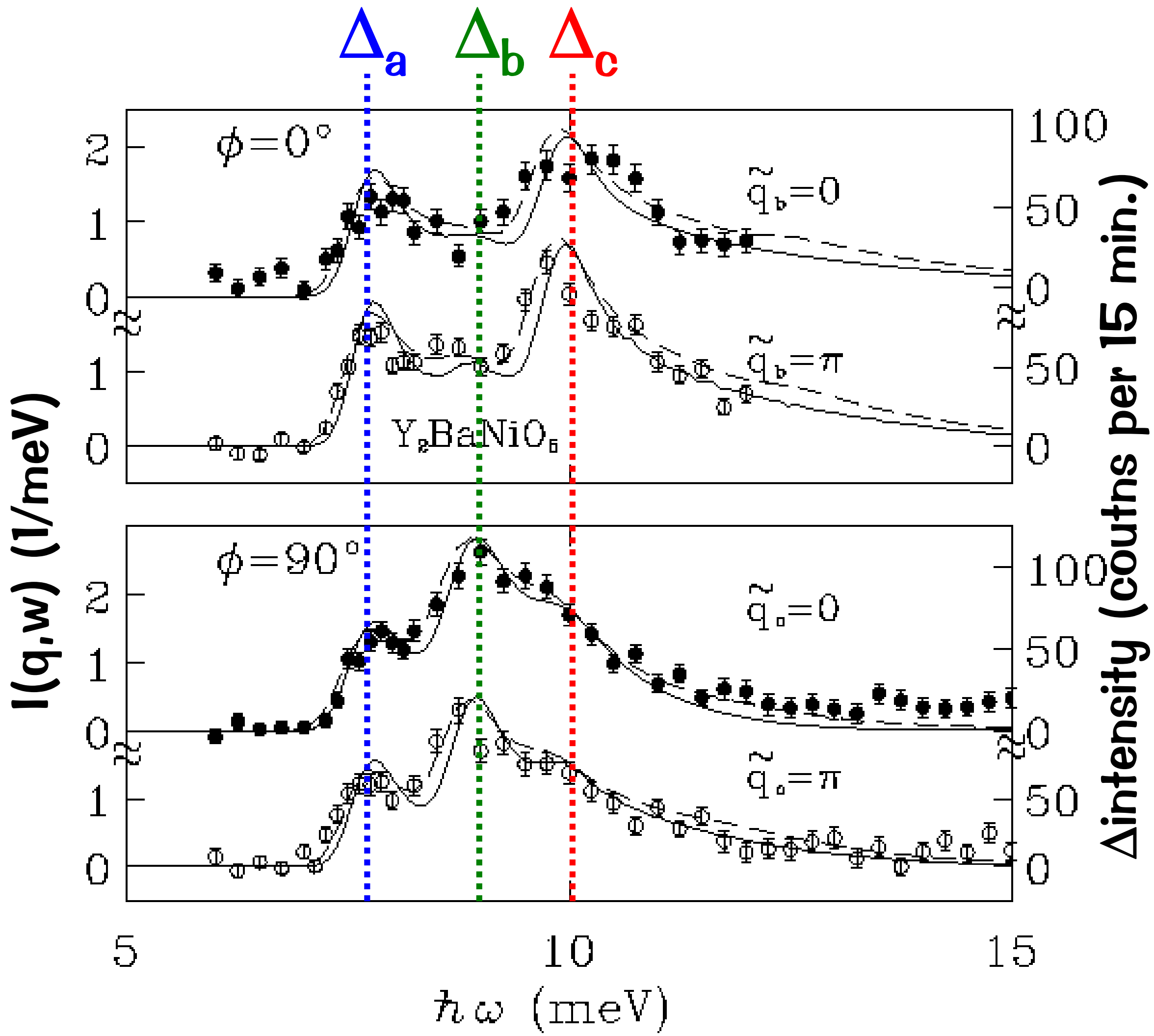
- For integer spin chains (S=n, z=2) this state is "close to" the ground state

- Excited states are propagating bond triplets separated from the ground state by an energy gap $\Delta \approx J$.



Haldane PRL 1983
Affleck, Kennedy, Lieb, and Tasaki PRL 1987

Probing anisotropy and inter-chain coupling in Y_2BaNiO_5



Maintaining $Q_{\|} = \frac{\pi}{a}$, we

- Derive polarization by rotating $\vec{Q}$ about chain
- Look for inter-chain coupling by varying $Q_{\perp}$

Weak anisotropy:

$$\delta\Delta/\overline{\Delta} \approx 10\%$$

Highly one dimensional

$$J'/J \leq 5 \cdot 10^{-4}$$

Sum rules and the single mode approximation

The dynamic spin correlation function obeys sum-rules:

$$\hbar\int d\omega\, S(\vec{q},\omega) = \frac{1}{N}\sum_{\vec{d}\vec{d}'} < \mathrm{S}_{\vec{d}}\mathrm{S}_{\vec{d}'} > e^{i\vec{q}\cdot(\vec{d}-\vec{d}')} \equiv S(\vec{q})$$

$$\hbar^2\int \omega\;\, d\omega\, S(\vec{q},\omega) = -\frac{2}{3}\frac{1}{N}\sum_{\vec{d}\vec{d}} J_{\vec{d}\vec{d}} < \vec{\mathrm{S}}_{\vec{d}}\cdot\vec{\mathrm{S}}_{\vec{d}'} > \left(1-\cos\vec{q}\cdot\left(\vec{d}-\vec{d}'\right)\right)$$

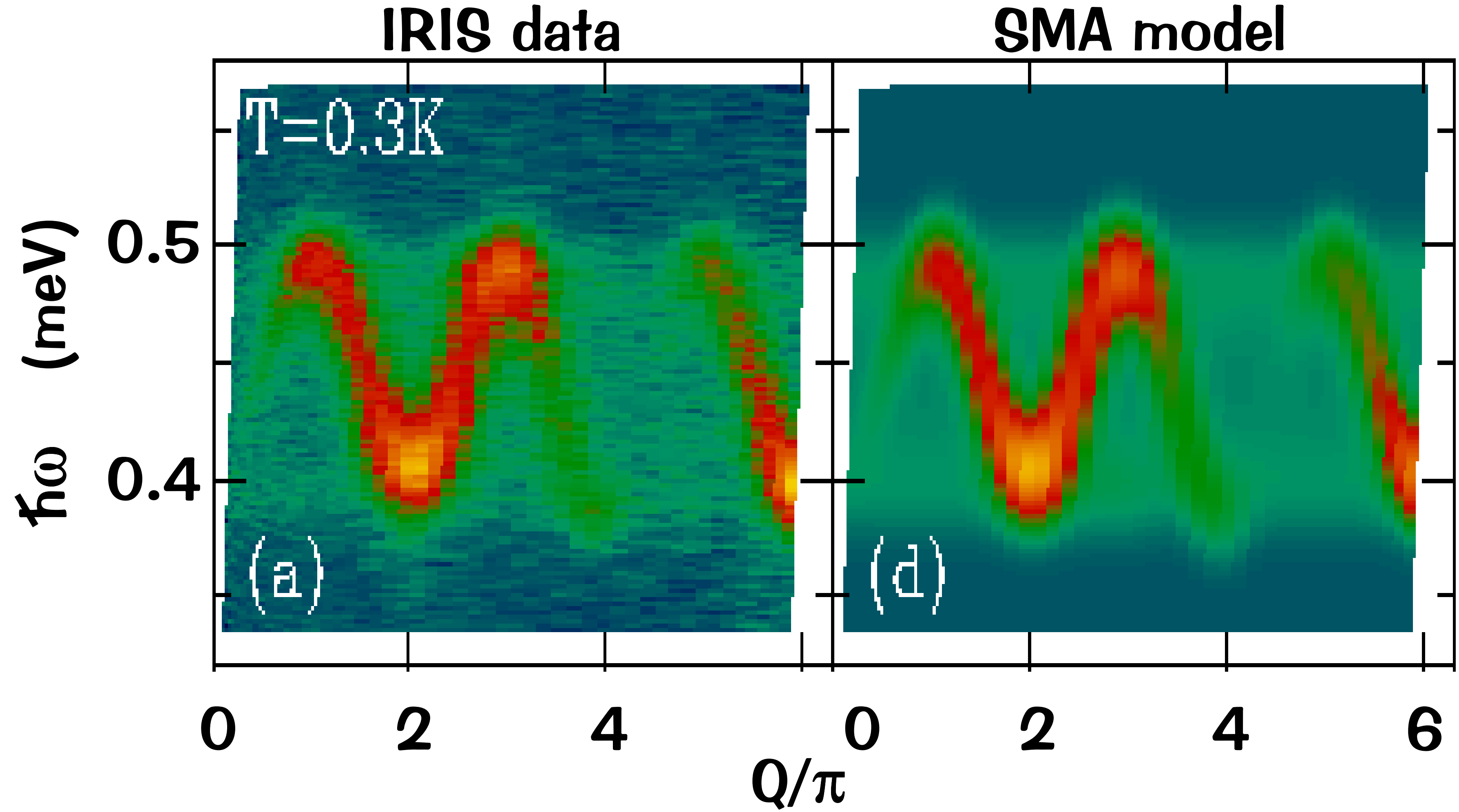
When a coherent mode dominates the spectrum:

$$S(\vec{q},\omega) \approx S(\vec{q})\delta\left(\hbar\omega - \varepsilon(\vec{q})\right)$$

Then sum-rules link S(q) and ε(q)

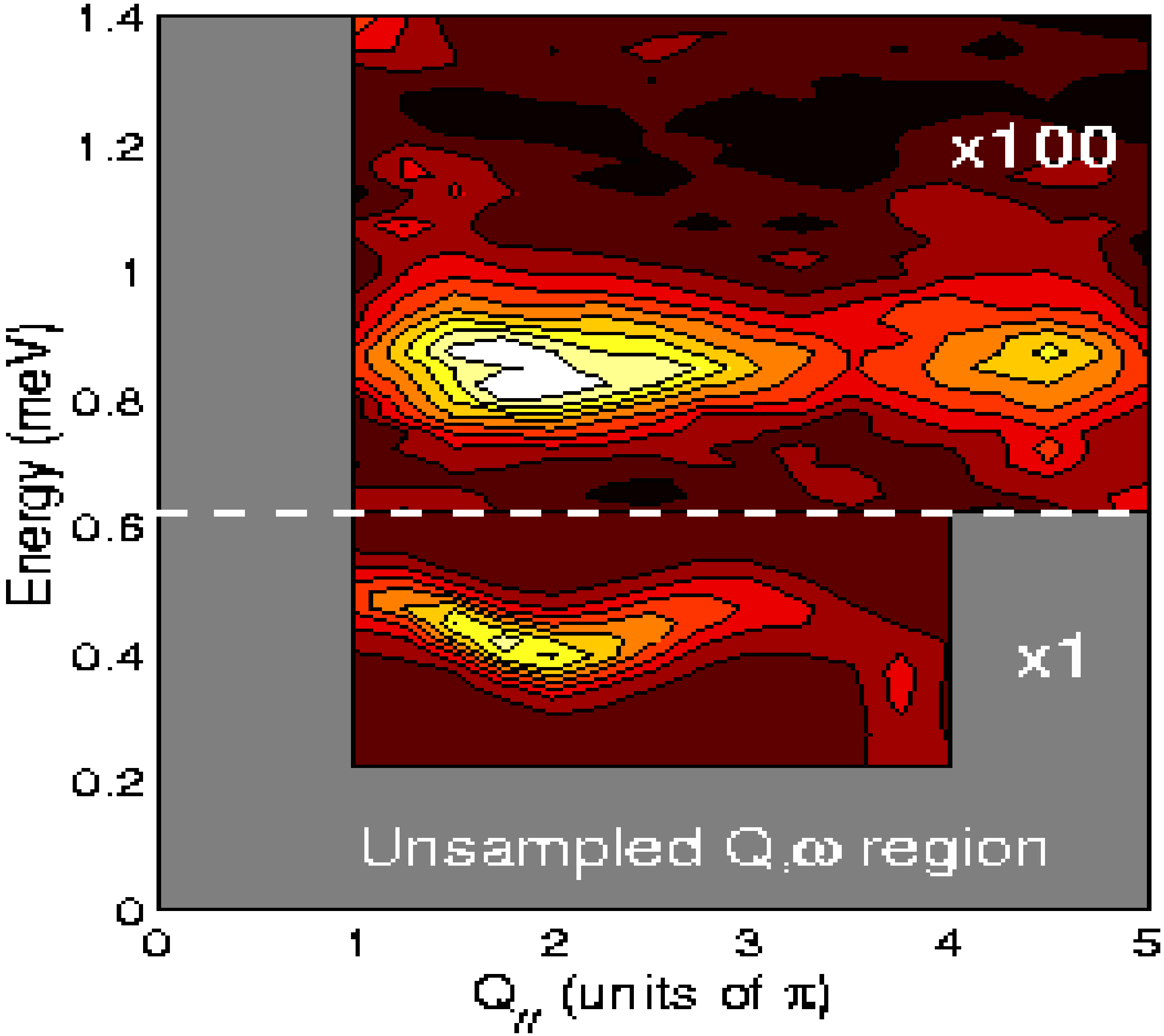
$$S(\vec{q}) \approx \frac{\hbar^2\int \omega\;\, d\omega\, S(\vec{q},\omega)}{\varepsilon\,(\vec{q})} = -\frac{2}{3}\frac{\frac{1}{N}\sum_{\vec{d}\vec{d}} J_{\vec{d}\vec{d}} < \vec{\mathrm{S}}_{\vec{d}}\cdot\vec{\mathrm{S}}_{\vec{d}'} > \left(1-\cos\vec{q}\cdot\left(\vec{d}-\vec{d}'\right)\right)}{\varepsilon\,(\vec{q})}$$

Propagating triplet in alternating spin-1/2 chain $Cu(NO_3)_2 \cdot 2.5D_2O$



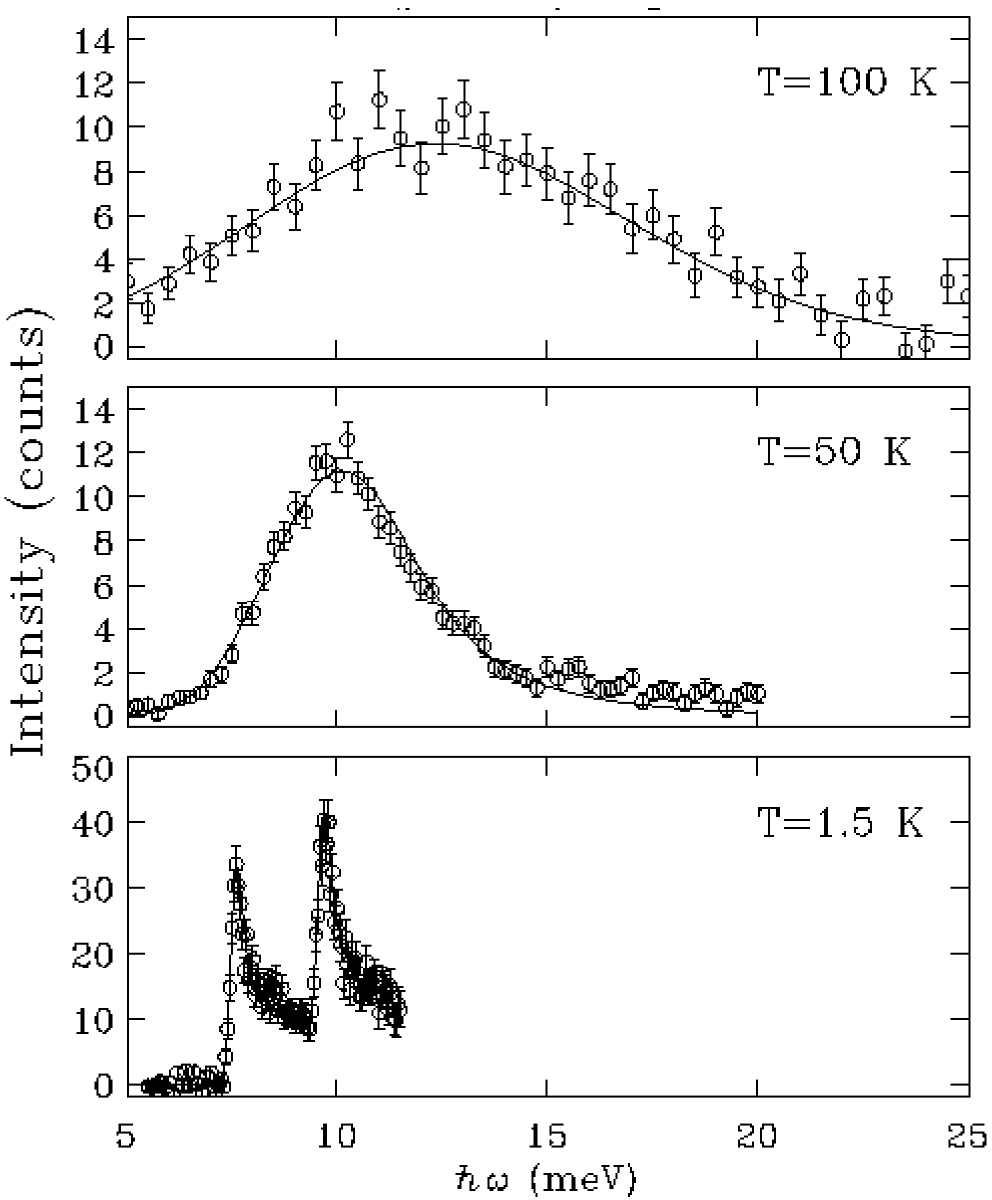
The "incommensurate" size of spin dimers yields different periods for dispersion relation and structure factor. An effect captured by the SMA.

Two magnon excitations in an alternating spin chain



Tennant, Nagler, Xu, Broholm, and Reich.

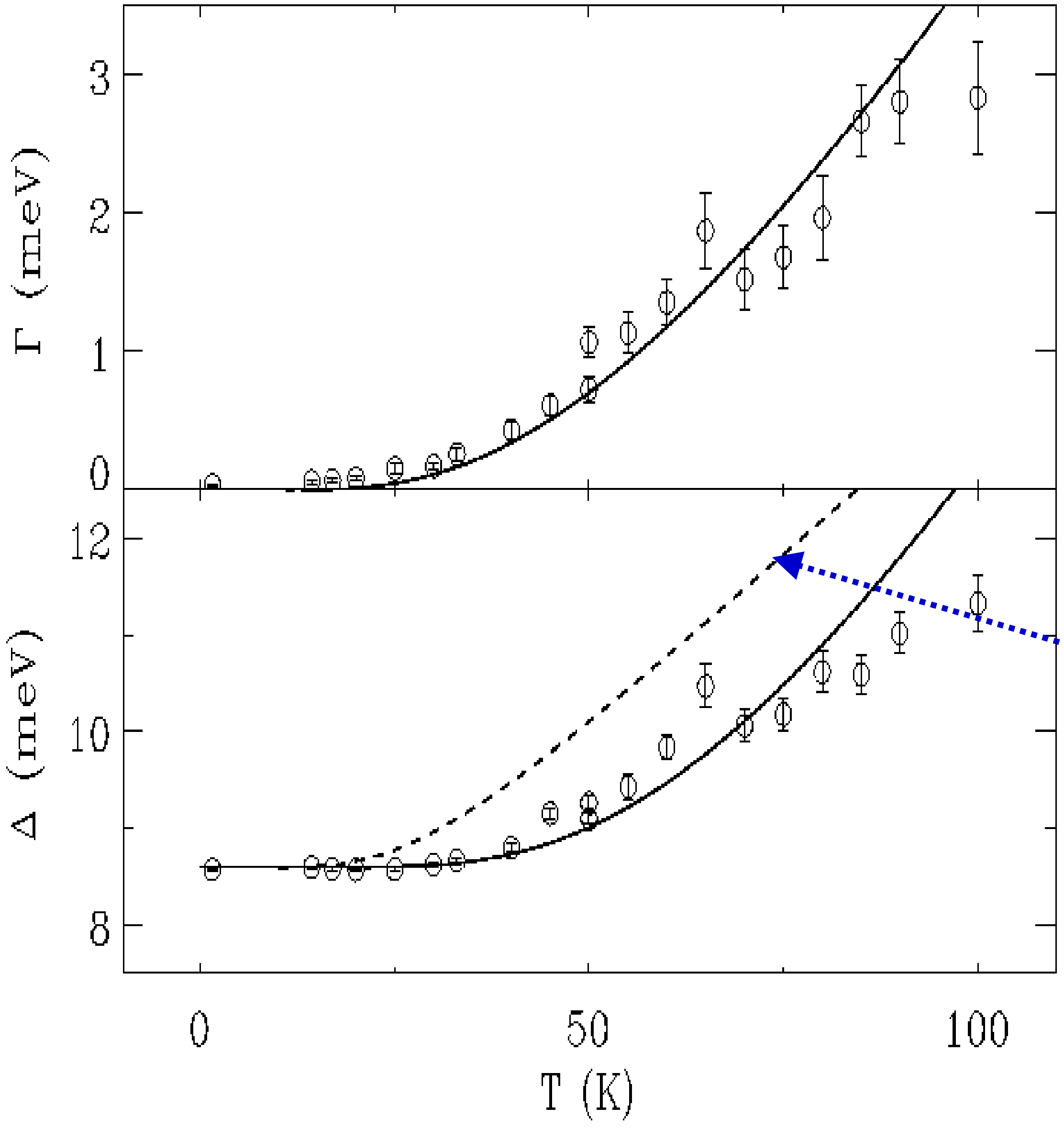
Haldane mode in Y_2BaNiO_5 at finite T



Effects of heating:

- **Line-width increases**
- **Effective Δ increases**

T-dependence of relaxation rate and "resonance" energy



Parameter free comparison:

- Semi-classical theory of triplet scattering by Damle and Sachdev

$$\Gamma(T) = \frac{3k_B T}{\sqrt{\pi}} \exp\left(-\frac{\Delta_0}{k_B T}\right)$$

- Quantum non linear σ model

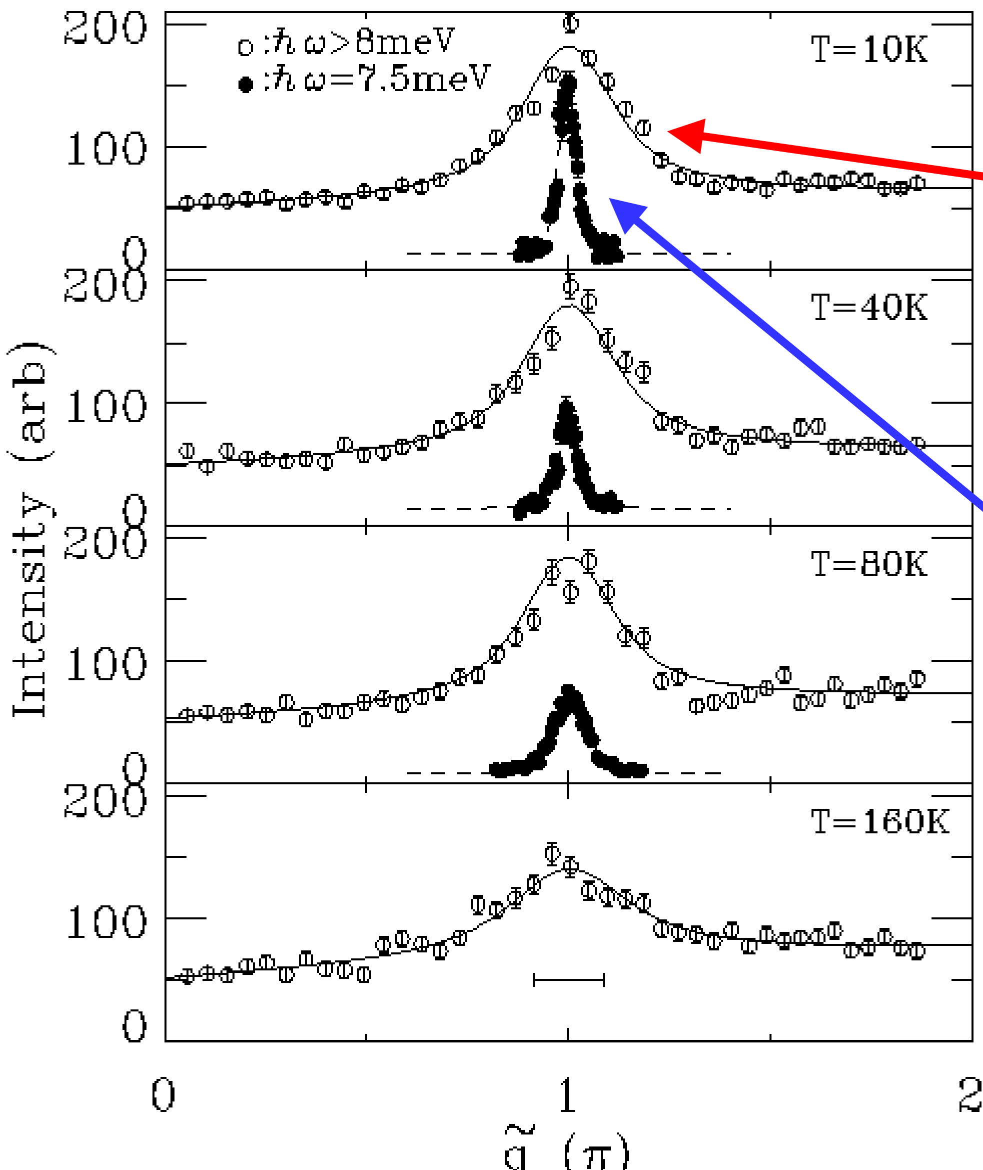
$$\Delta(T) = \Delta_0 + \sqrt{2\pi \Delta_0 k_B T} \exp\left(-\frac{\Delta_0}{k_B T}\right)$$

Derived from

$$\Delta(T) = \frac{c_0}{\xi(T)}$$

Neglecting T-dependence of spin wave velocity c_0

Q-scans versus T:
energy resolved and energy integrated

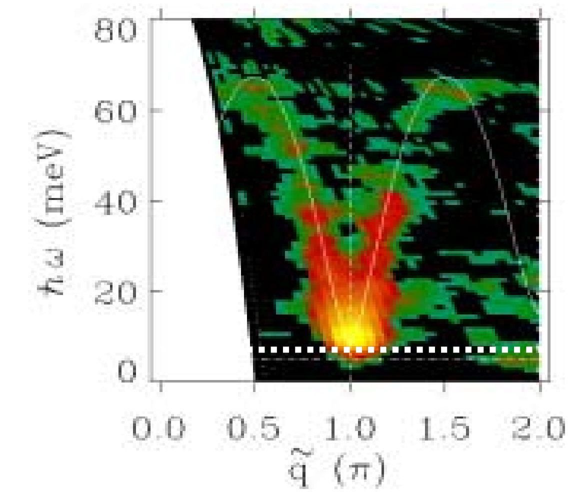


$\hbar\omega \geq \Delta$

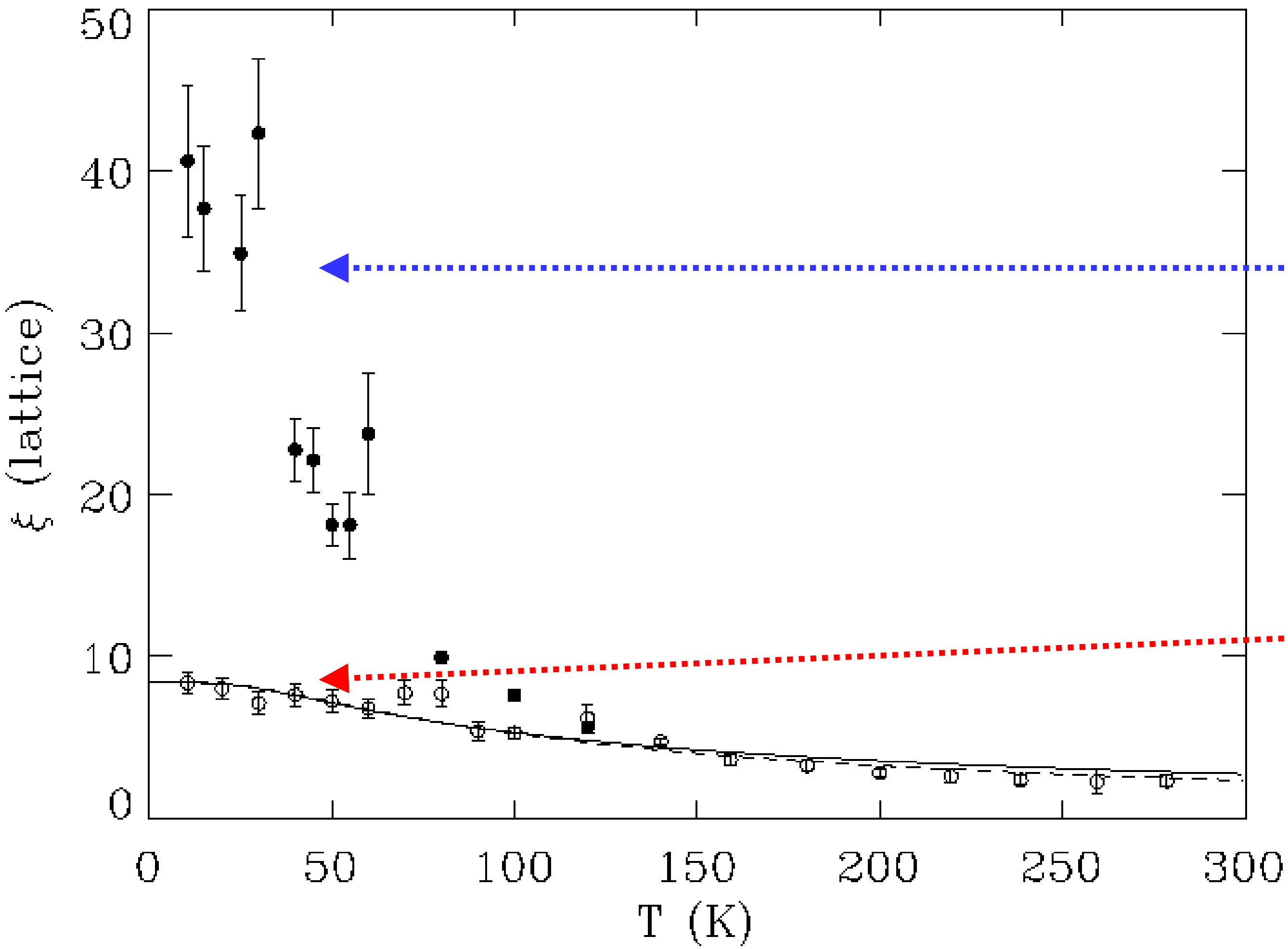
Probing equal time correlation length

$\hbar\omega = \Delta$

Probing spatial coherence of Haldane mode



Coherence and correlation lengths versus T



Coherence length exceeds correlation length for $k_B T < \Delta$ becoming very long as T $\rightarrow$ 0

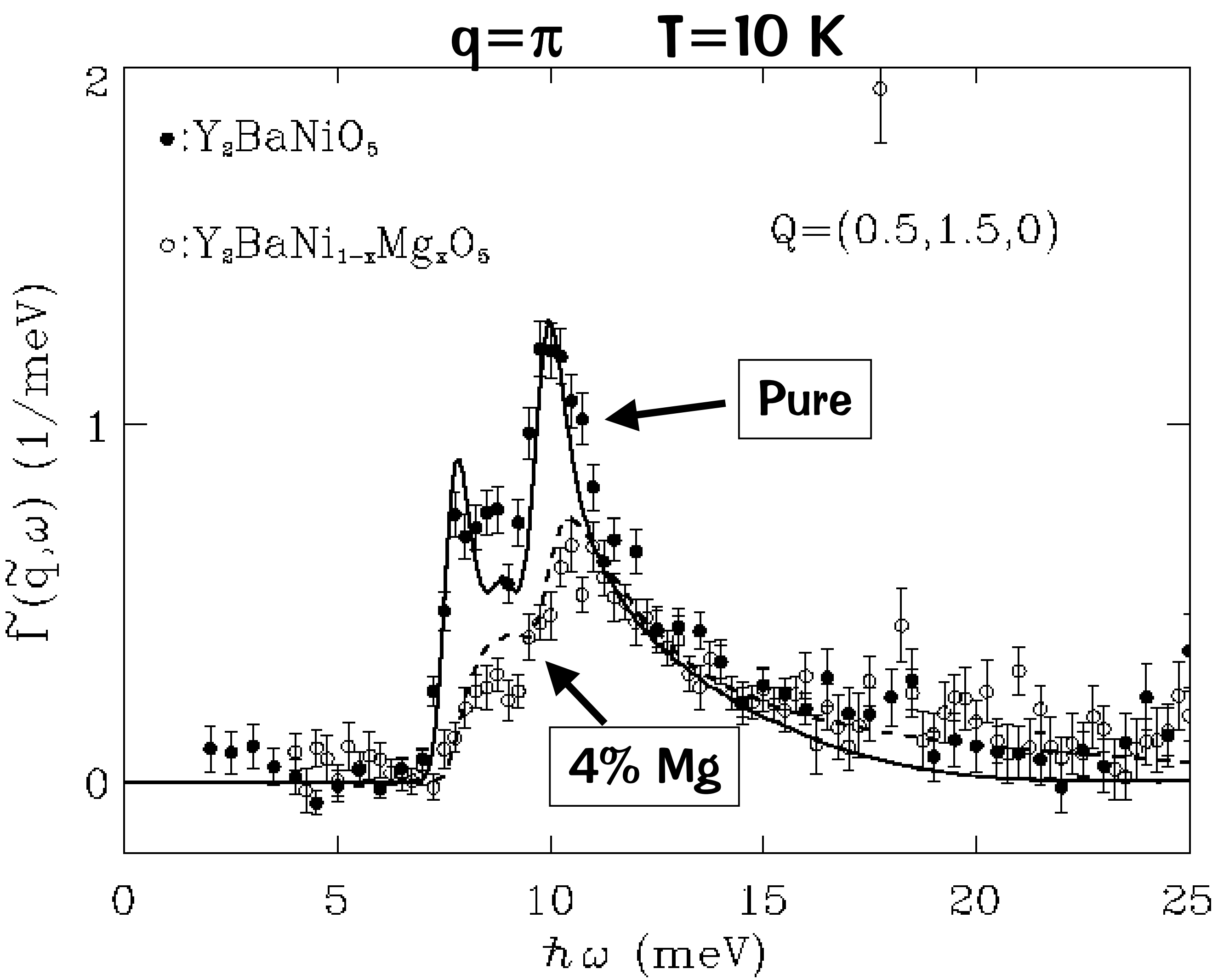
Equal-time correlation length saturates at $\xi = 8$.

(Solid line from Quantum non linear σ model)

Properties of pure Y_2BaNiO_5

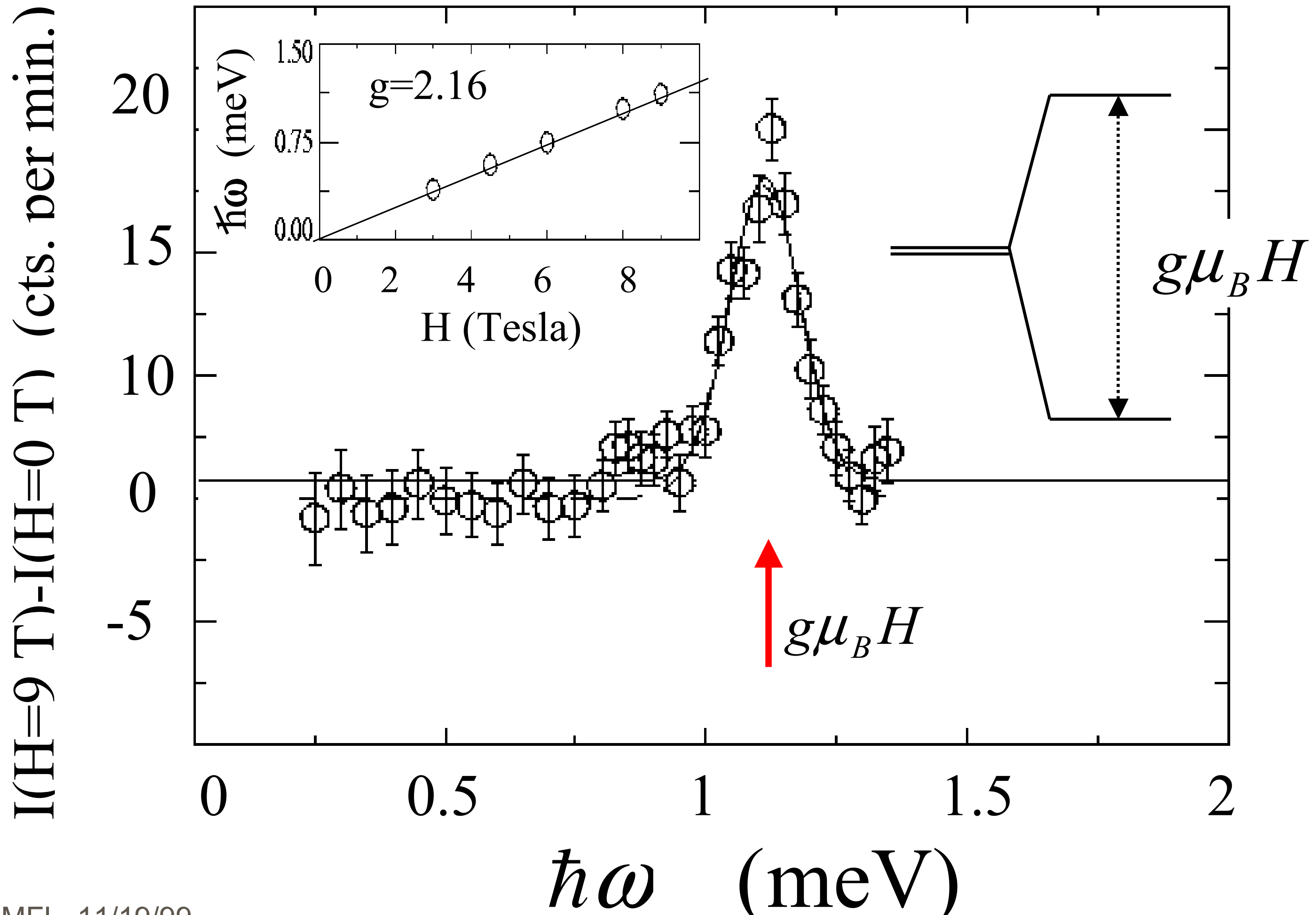
- Anisotropy split Haldane gap:
  Δ_a=7.5 meV, Δ_b=8.6 meV, Δ_c=9.5 meV
- No inter-chain coupling detected: $|J'/J|<5\cdot10^{-4}$
- Coherent mode described by SMA for $T<<\Delta/k_B$
- Activated relaxation rate of $q=\pi$ mode is described by semi-classical theory of interacting triplet wave packets.
- Activated increase in resonance energy is significantly less than predicted by Qnlσ-model
- Coherence length exceeds correlation length for $T<\Delta/k_B$ and exceeds 40 lattice spacings for $k_BT/\Delta=0.1$

Effects of finite chain length on Haldane mode



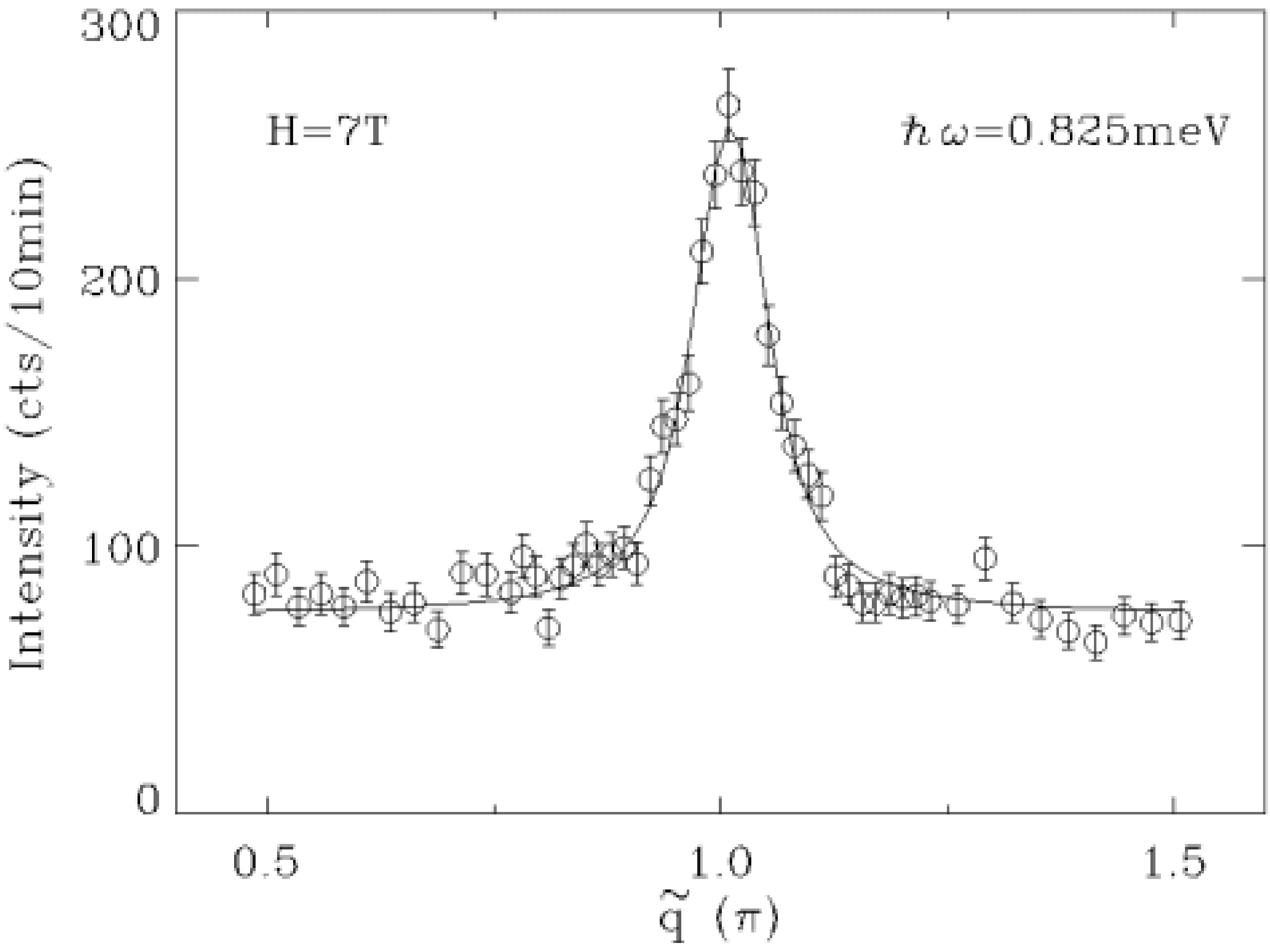
- Mode shifts towards J as in numerical work on finite length chains
- Peak Broadens because of chain length distribution

Zeeman resonance of chain-end spins



Form factor of chain-end spins

$Y_2BaNi_{1-x}Mg_xO_5$ x=4% $\hbar\omega = g\mu_B H$



Q-dependence reveals that resonating object is AFM.

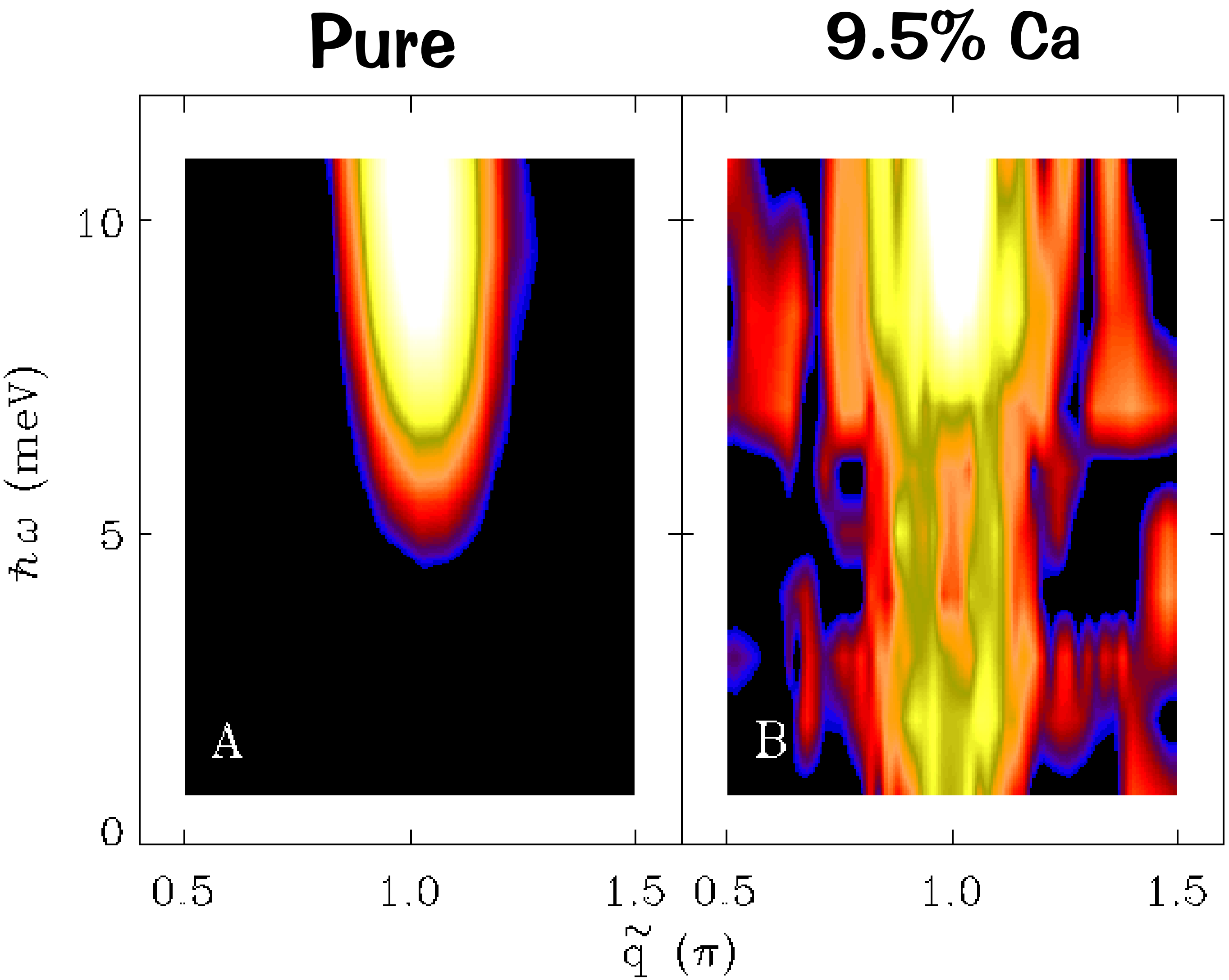
The peak resembles S(Q) for pure system.

Chain end spin carry AFM spin polarization of length ξ back into chain

Vacancy doping a Haldane spin chain

- $q=\pi$ mode shifts towards J
- $q=\pi$ mode broadens due to random chain length distribution
- Applied field induces Zeeman resonance below Haldane gap
- Resonating chain end spins have AFM form factor resembling S(q) for pure system.

New excitations in Ca-doped Y_2BaNiO_5



$Y_{2-x}Ca_xBaNiO_5$:

- Ca-doping creates states below the gap
- sub-gap states have doubly peaked structure factor

Why a double ridge below the gap in $Y_{2-x}Ca_xBaNiO_5$?

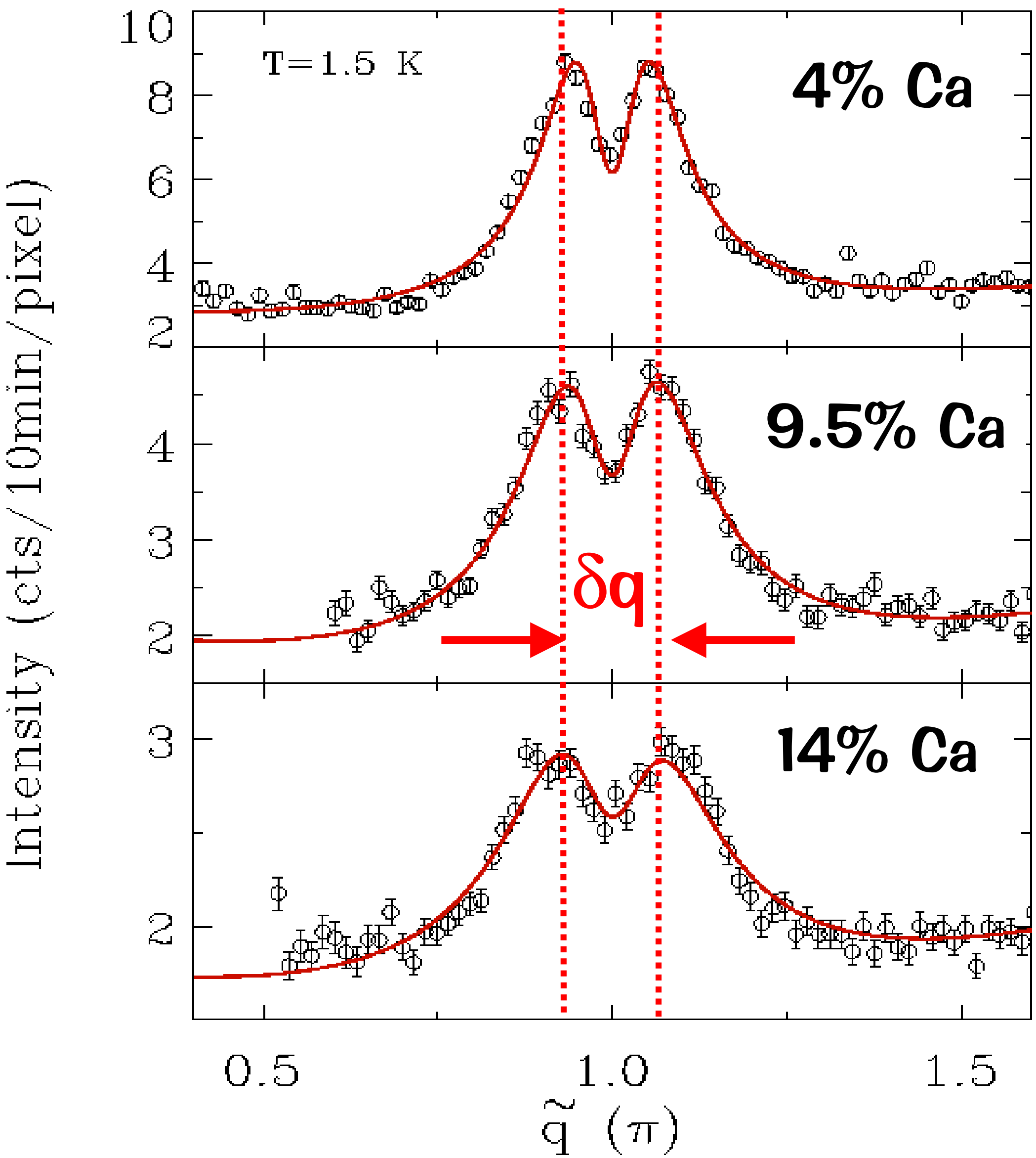
- Charge ordering yields incommensurate spin order
- Quasi-particle Quasi-hole pair excitations in a one dimensional hole liquid

$$\delta q \propto x$$

- Anomalous form factor for independent spin degrees of freedom associated with each donated hole

δq is single impurity prop. Indep. of x

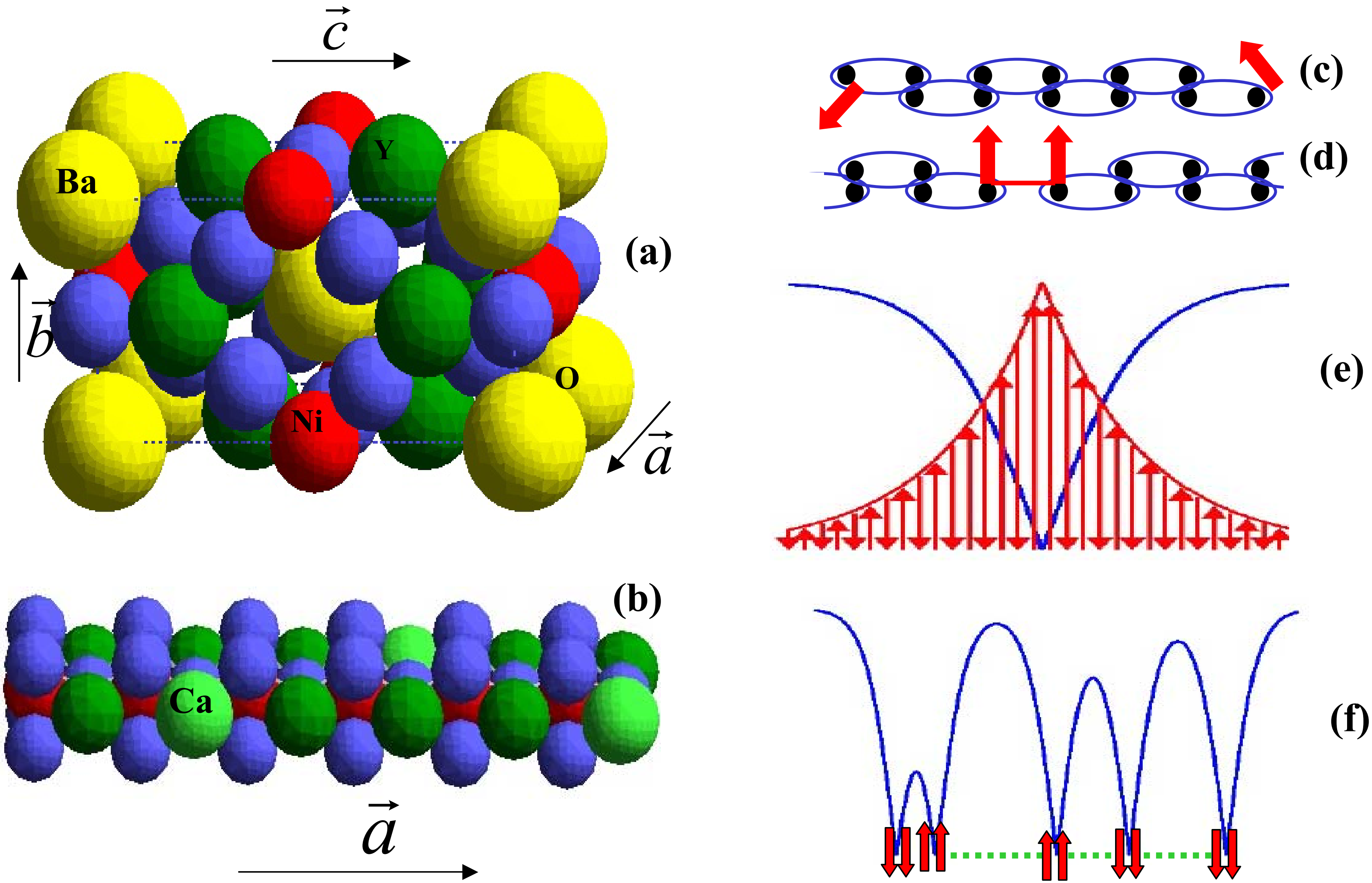
Does δq vary with calcium concentration?



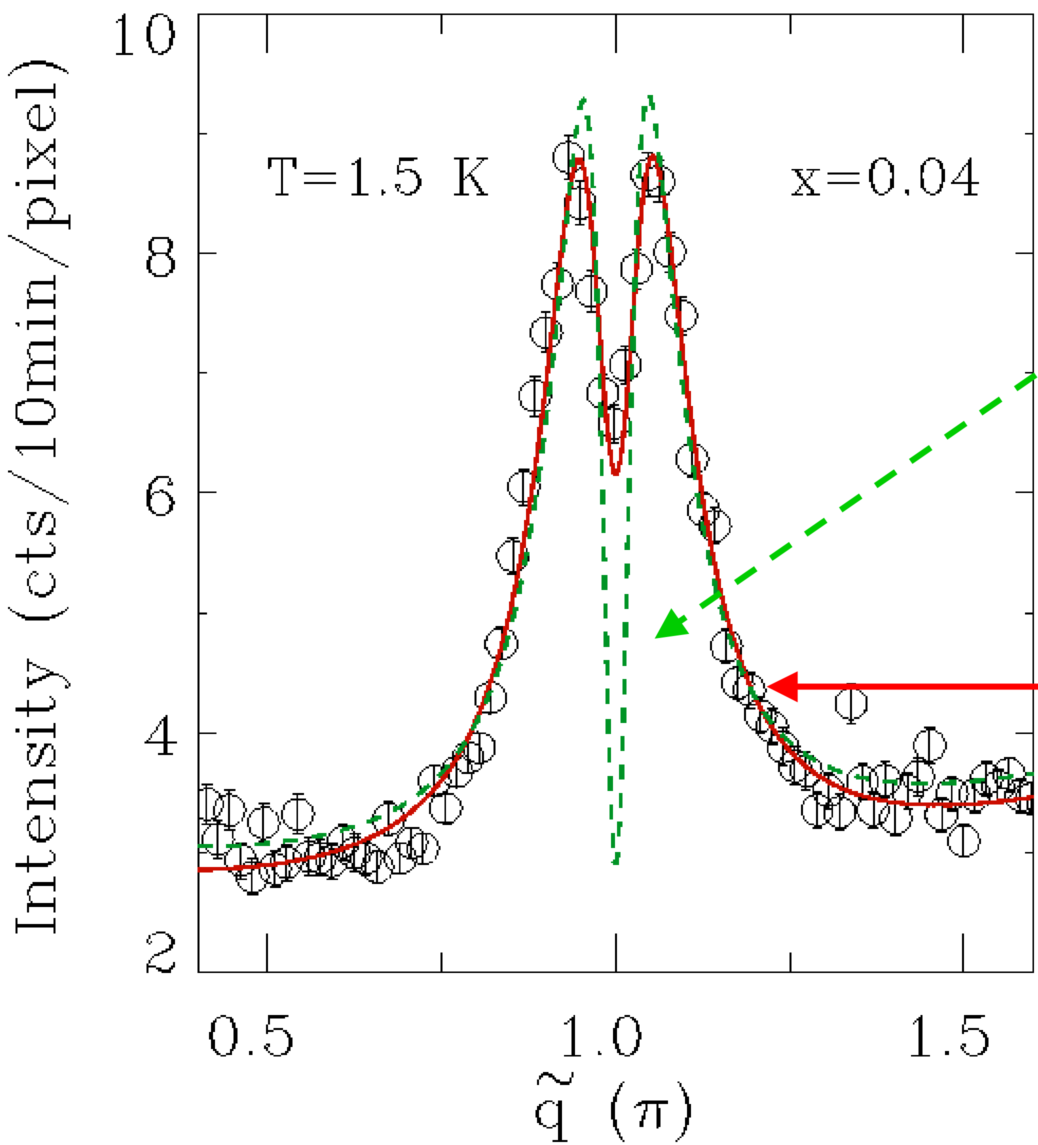
δq is independent of $x \in [0.04 \; ; 0.14]$

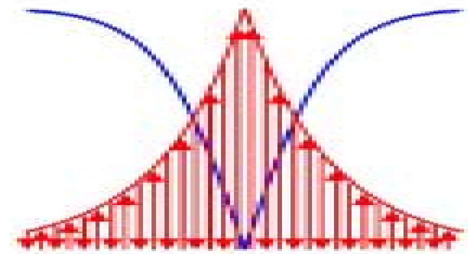
Double peak is predominantly a single impurity effect

Bond Impurities in a spin-1 chain: $Y_{2-x}Ca_xBaNiO_5$



Form-factor for FM-coupled chain-end spins



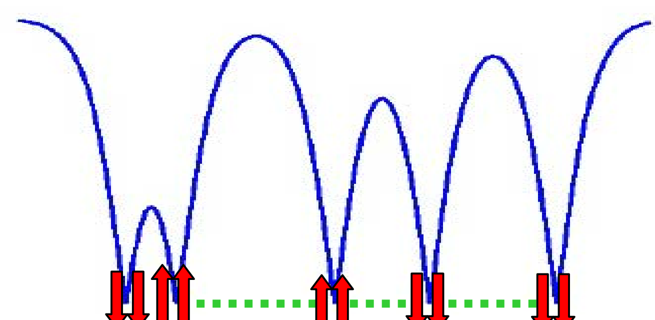


A symmetric AFM droplet

$$S(q)=2\,\mathrm{Re}\left\{M_{\infty}(q)e^{iq/2}\right\}$$

Ensemble of independent randomly truncated AFM droplets

$$S(q)=\sum_{ll'}P_{ll'}\left|M_{l}(q)e^{iq/2}+M^{*}_{l'}(q)e^{-iq/2}\right|^{2}$$



Calcium doping Y_2BaNiO_5

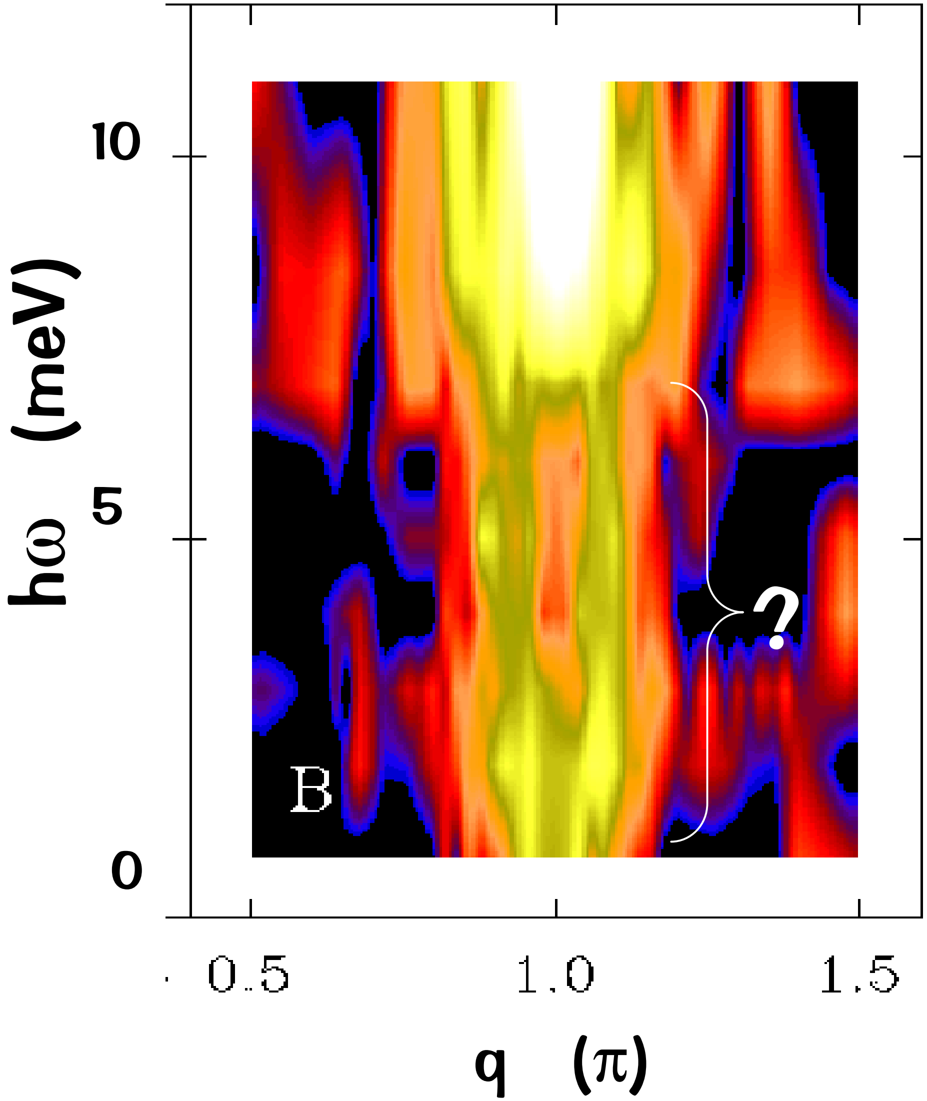
Experimental facts:

- Ca doping creates sub-gap excitations with doubly peaked structure factor and bandwidth $\geq \Delta$
- The structure factor is insensitive to concentration and temperature for $0.095<x<0.14$ and $T<100$ K

Analysis:

- Ca^{2+} creates FM impurity bonds which nucleate AFM droplets with doubly peaked structure factor
- AFM droplets interact through intervening chain forming disordered random bond 1D magnet

What sets energy scale for sub gap scattering ?



Possibilities:

- Residual spin interactions through Haldane state. A Random bond AFM.
- Hole motion induces additional interaction between static AFM droplets
- AFM droplets move with holes: scattering from a Luttinger liquid of holes.

How to distinguish:

- Neutron scattering in an applied field
- Transport measurements
- Theory

Broader Conclusions:

- Dilute impurities in the Haldane spin chain create sub-gap composite spin degrees of freedom.
- Composite spins have an AFM wave function that extends into the bulk over distances of order the Haldane length.
- Neutron scattering can detect the structure of composite impurity spins in quantum magnets when the corresponding states exist at energies where the bulk magnetic density of states vanishes.