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Spin dynamics in the hyperkagome compound $Gd_3Ga_5O_{12}$

Pascale Deen

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[4] CNRS - Institut Néel, Grenoble, France.
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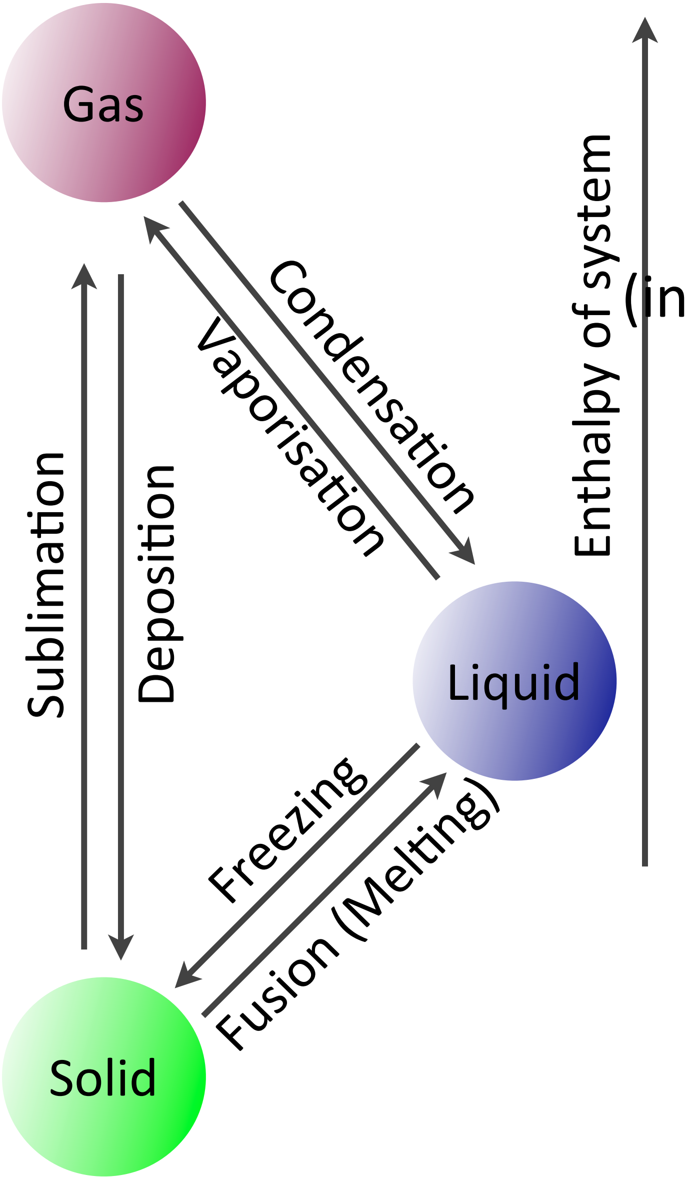
Outline

- Phase transitions, frustration and dynamics
- Pyrochlores versus Kagome compounds
- 3D Kagome $Gd_3Ga_5O_{12}$:
    - a background
    - Inelastic neutron scattering
- What does it mean.

Phase transitions



Gas

Liquid

Solid

Condensation

Vaporisation

Sublimation

Deposition

Freezing

Fusion (Melting)

Enthalpy of system

Universality

Liquids = Magnetism

$(T - Tc)^{\alpha}$

Liquid-gas critical point
(independent of composition)

FM phase transitions
uniaxial magnets

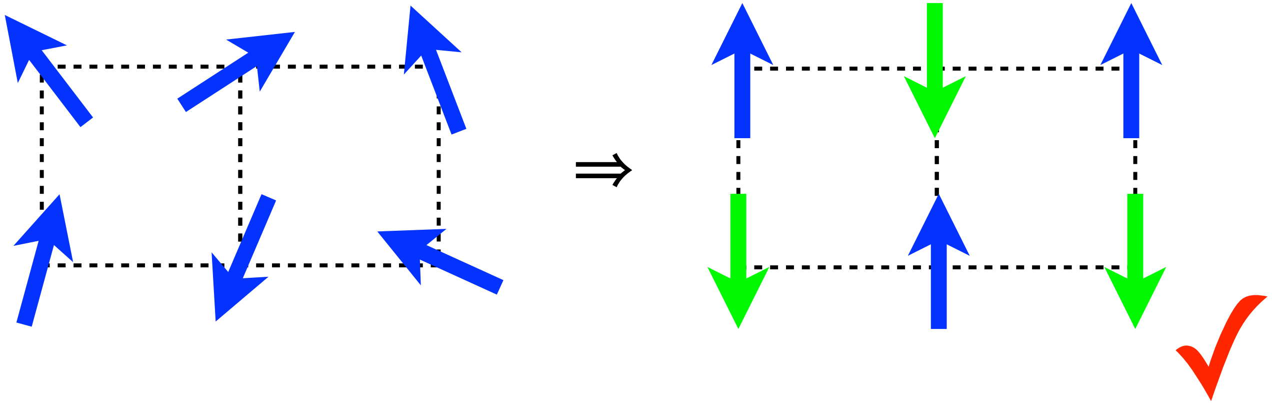
Same universality class

Universal thermodynamic properties of a system near a phase transition depend only on a small number of features, such as dimensionality and symmetry, and are insensitive to the underlying microscopic properties of the system.

Magnetic phase transitions

As T ⇒ 0 , Divergent correlation lengths

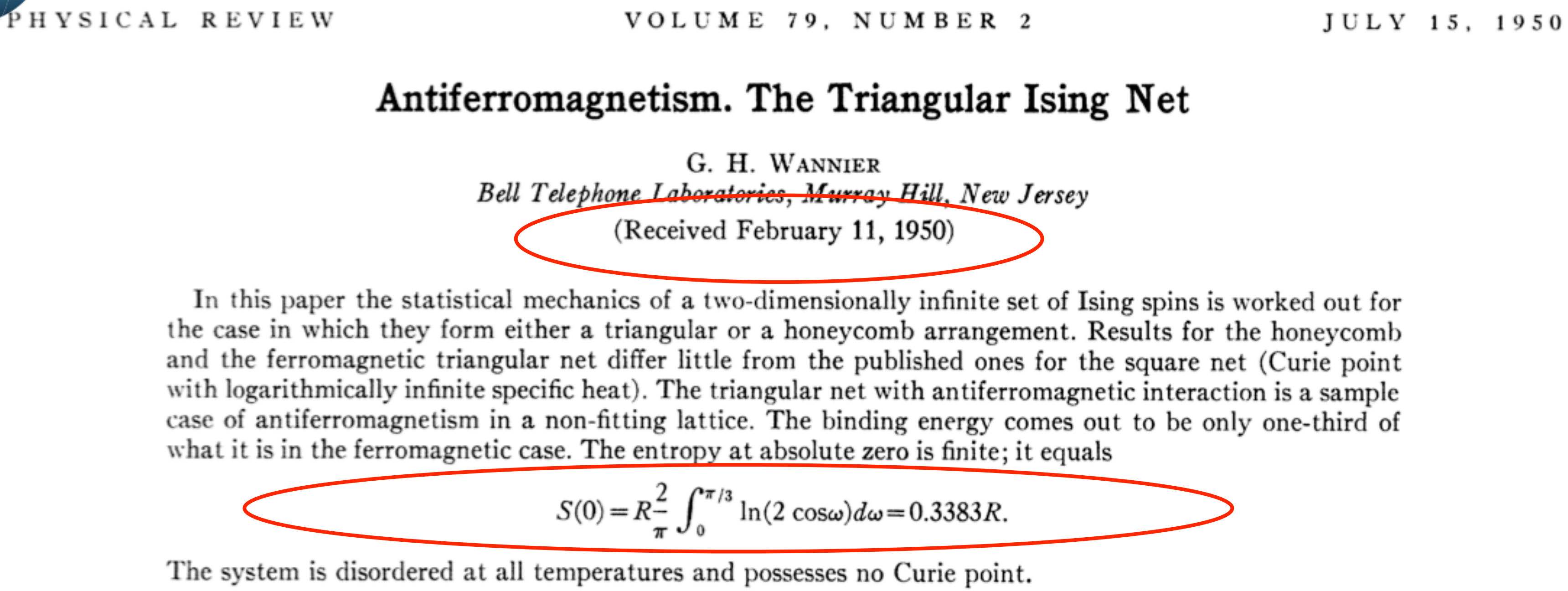
Phase transition to long range order



Minimise all interactions to achieve a ground state

$$\sigma_1 + \sigma_2 + \sigma_3 + \ldots\ldots = 0$$

Frustration ⇒ zero point entropy ⇒ degeneracy



PHYSICAL REVIEW VOLUME 79, NUMBER 2 JULY 15, 1950

Antiferromagnetism. The Triangular Ising Net

G. H. WANNIER
Bell Telephone Laboratories, Murray Hill, New Jersey

(Received February 11, 1950)

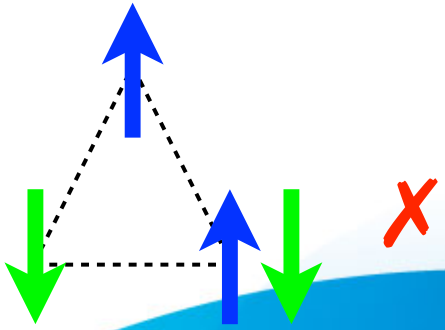
In this paper the statistical mechanics of a two-dimensionally infinite set of Ising spins is worked out for the case in which they form either a triangular or a honeycomb arrangement. Results for the honeycomb and the ferromagnetic triangular net differ little from the published ones for the square net (Curie point with logarithmically infinite specific heat). The triangular net with antiferromagnetic interaction is a sample case of antiferromagnetism in a non-fitting lattice. The binding energy comes out to be only one-third of what it is in the ferromagnetic case. The entropy at absolute zero is finite; it equals

$$S(0)=R\frac{2}{\pi}\int_0^{\pi/3}\ln(2\cos\omega)d\omega=0.3383R.$$

The system is disordered at all temperatures and possesses no Curie point.

Geometric Frustration

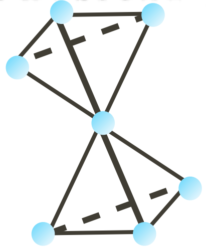
AF interactions J > 0
Ising spins
($\sigma_i = \pm 1$)



Entropy = 0.34 kb /spin at T= 0 K.

Frustration ⇒ zero point entropy ⇒ degeneracy

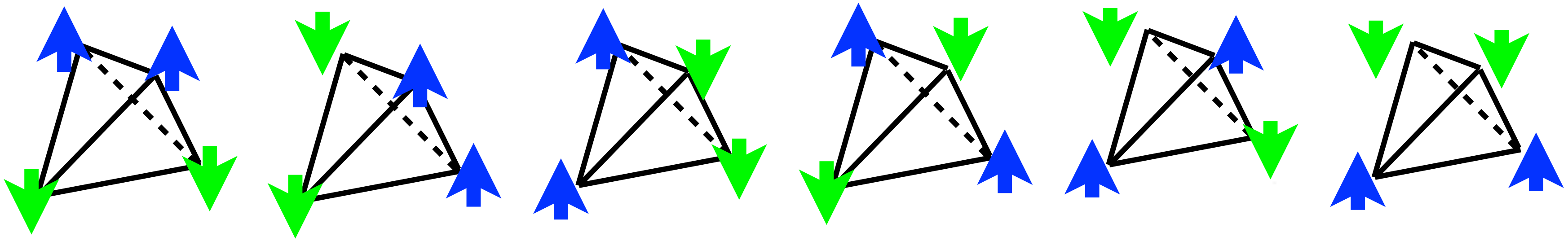
PHYSICAL REVIEW VOLUME 102, NUMBER 4 MAY 15, 1956



Ordering and Antiferromagnetism in Ferrites

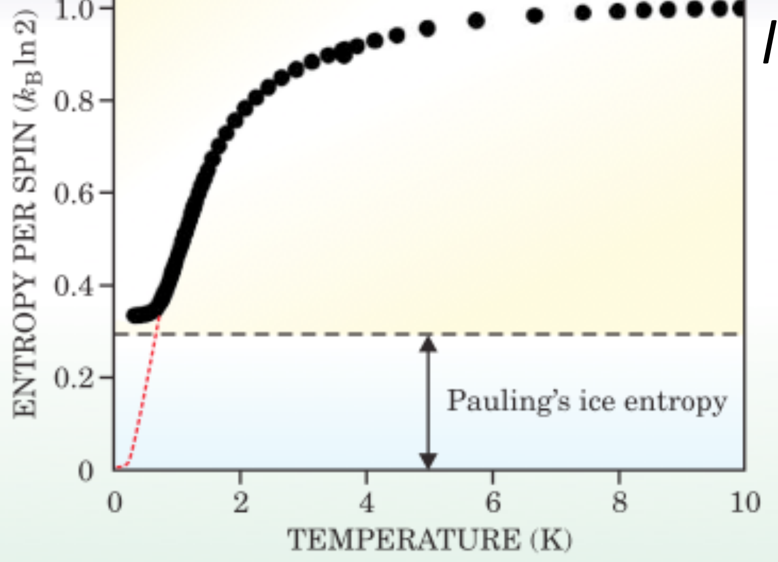
P. W. ANDERSON
Bell Telephone Laboratories, Murray Hill, New Jersey

(Received January 9, 1956)



A. P. Ramirez et al., Nature 399, 333 (1999).

Moessner et al. Physics Today 2006.



Nearest neighbour exchange cannot lead to long range order but there is short range order with finite zero point entropy

Manifold of connected ground states

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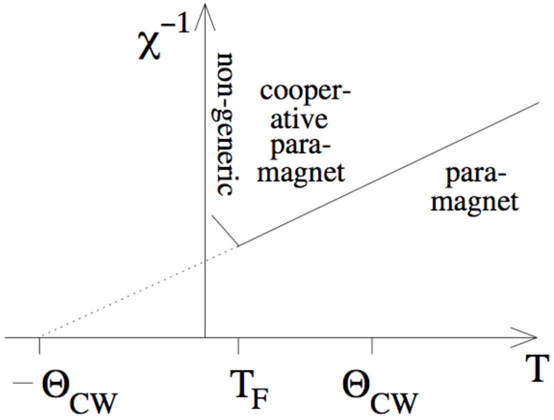
Frustration ⇒ zero point entropy ⇒ degeneracy

Signatures

- No order down to lowest temperatures
  T << J(Near neighbour) ~ θ_{CW}

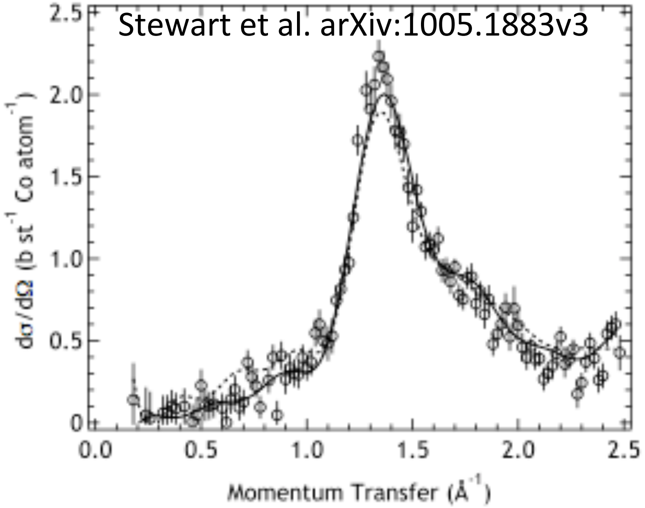
- Zero point entropy. Degenerate ground state

‘Susceptibility fingerprint’ of frustration



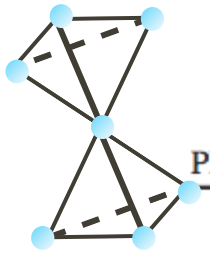
- Short range correlations in space

  Neutron scattering
  $\Delta Q = 1/\xi$


Stewart et al. arXiv:1005.1883v3

Dynamic behaviour?

Dynamic behaviour: Pyrochlores



SICAL REVIEW B VOLUME 58, NUMBER 18 1 NOVEMBER 1998-II

Low-temperature properties of classical geometrically frustrated antiferromagnets

R. Moessner and J. T. Chalker

PRL **102,** 237206 (2009) PHYSICAL REVIEW LETTERS week ending 12 JUNE 2009

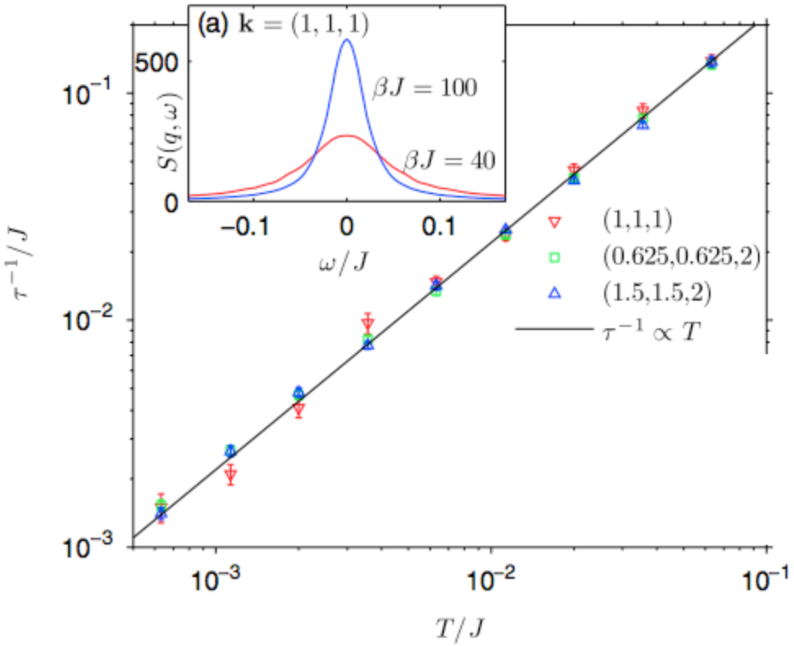
Spin Dynamics in Pyrochlore Heisenberg Antiferromagnets

P. H. Conlon* and J. T. Chalker

- Time dependence of spin-spin autocorrelation fuction.

$$\langle \mathbf{S}_\mathbf{r}(0)\mathbf{S}_\mathbf{r}(t)\rangle = \exp(-cTt),$$

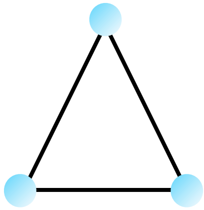
- FT = Lorentzian scattering in frequency space with ΔE ~T . Depends on T not J.
- Spin correlations relax at a rate independent of the wave vector and proportional to the temperature.



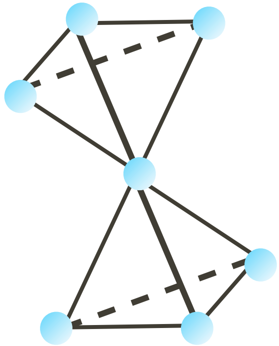
Frustration ⇒ zero point entropy ⇒ degeneracy

S > 1/2 = not quantum

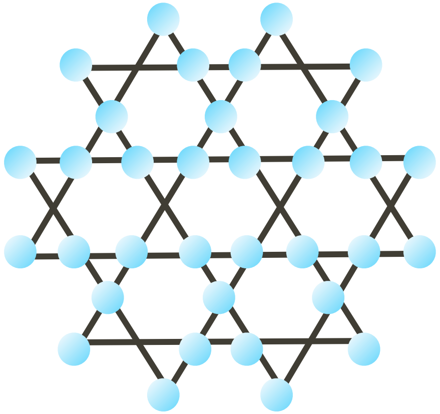
Triangle



Pyrochlore



Kagome



Spin liquid, spin ice

How does it compare?

Magnetic analogue of ice and liquid.

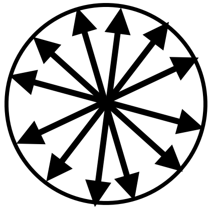
Dynamically: Dynamic gapless excitations (degeneracy) to 0 K.

Universal dynamic behaviour?

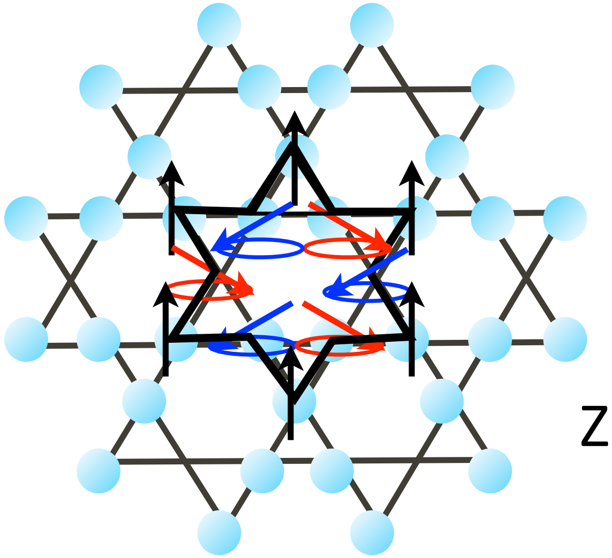
Kagome lattice

Prototype of geometrical frustration

Juxtaposition of theory and experiment



NN Heisenberg AF : Infinite degenerate number of spin configurations



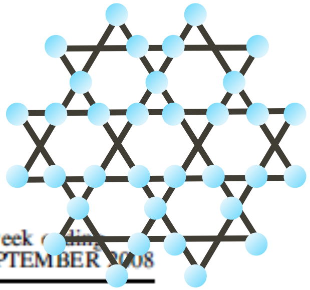
Constraint of AF Spins on a Kagome lattice constraint =120°

Zero energy modes (3 spin, 6 spin, xx spin)

Non-dispersive and costs no energy.
Rotations of finite number of spins:
No correlations in space.

Hall mark of Kagome dynamics.

Theory

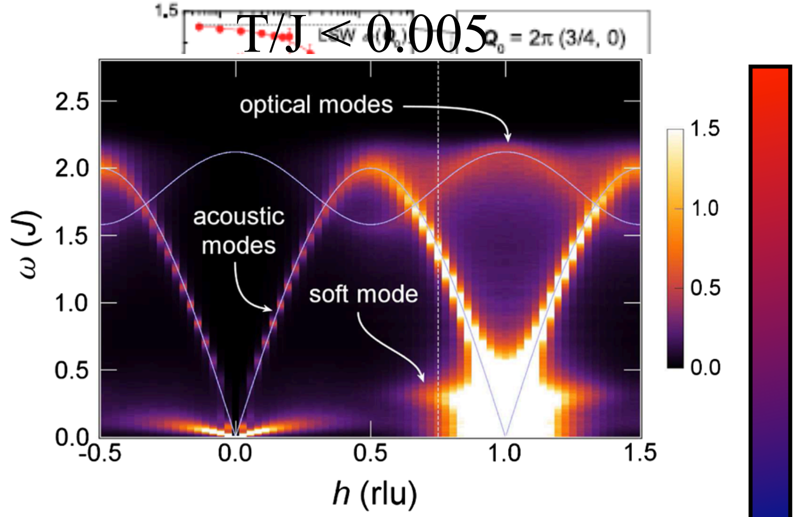


PRL **101,** 117207 (2008) PHYSICAL REVIEW LETTERS week ending 12 SEPTEMBER 2008

Propagation and Ghosts in the Classical Kagome Antiferromagnet

J. Robert,[1] B. Canals,[2] V. Simonet,[2] and R. Ballou[2]

Nearest neighbour interactions only



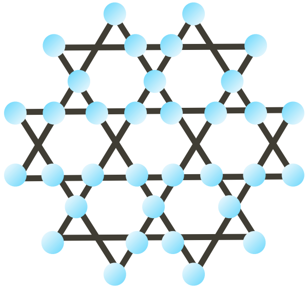
T/J < 0.005

FIG. 2 (color online). Temperature weighted scattering function vs energy at different temperatures for $\mathbf{Q}_0 = 2\pi(3/4, 0)$. Inset: position of soft and acoustic modes vs temperature; error bars obtained from several fitting processes.

T/J_{NN}	Expectation
> 0.2	QE signal only
0.2-0.01	Broad excitation
< 0.01	Splits into acoustic and soft mode
< 0.005	+ Optical mode

Sufficient temporal and spatial stiffness gives rise to soft modes. They fall to zero energy as T ➔ 0 K.

Dynamics to date



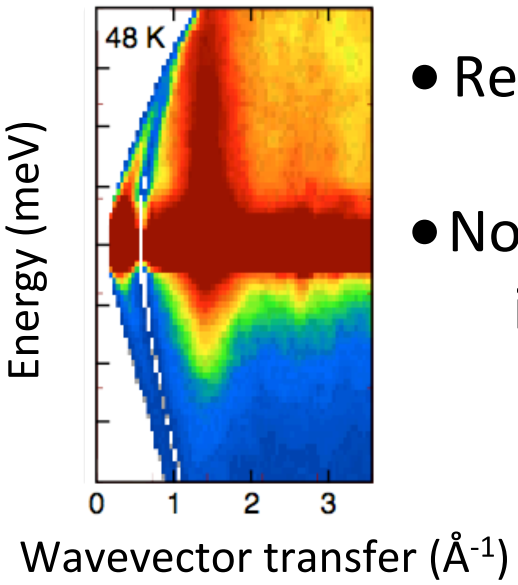
Classical Heisenberg AF on a Kagome lattice
Gapless excitations extend far out in energy
Spatial dependence: SRO

$Y_{0.5}Ca_{0.5}BaCo_4O_7$:

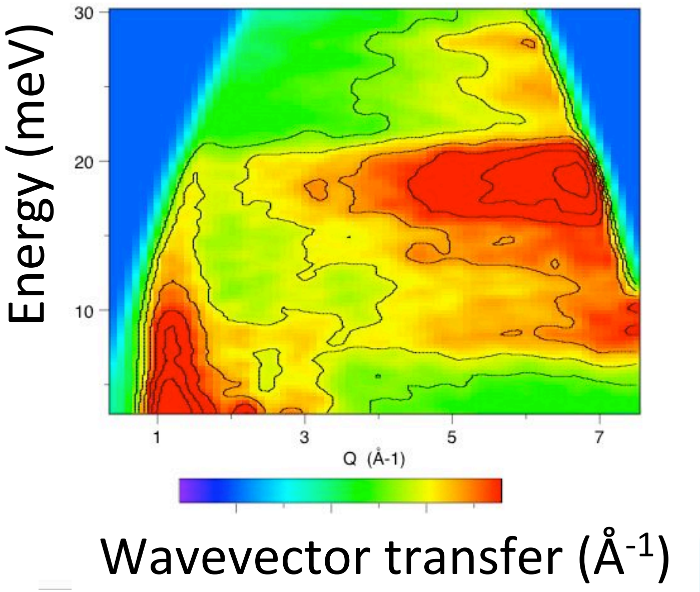
J. R. Stewart et al. arXiv:1005.1883v3

Deuterium Jarosite :

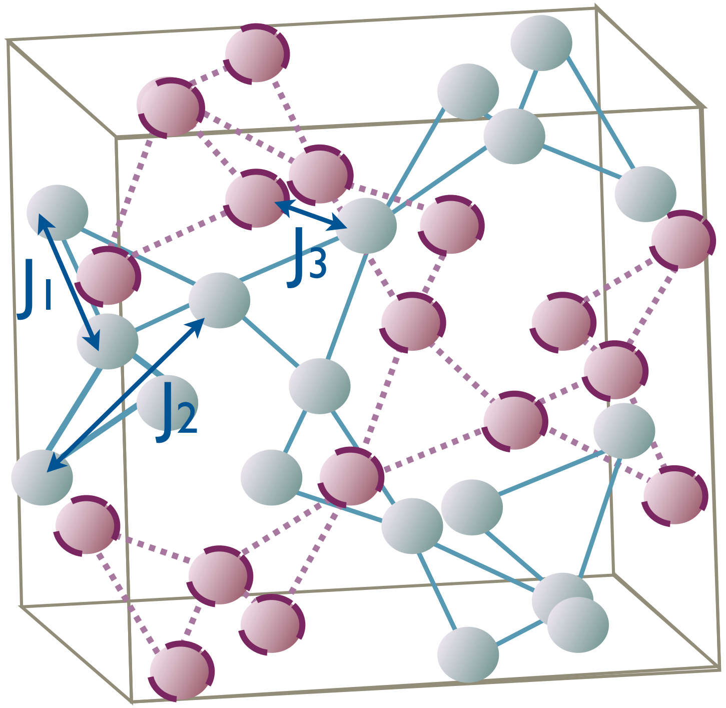
B. Fåk, *et al.*
Europhysics Letters (2008) 81, 17006



- Remains fluctuation down to lowest temperatures.
- No gap in the excitations - no internal energy barrier



Spin Dynamics in the Hyperkagome (3D Kagome) $Gd_3Ga_5O_{12}$



- Each RE spin has 4 NN
- NN form lattice of equilateral triangles.
- 2 Interpenetrating corner sublattices, spin do NOT lie in same plane.
- No inversion symmetry.

Bulk measurements

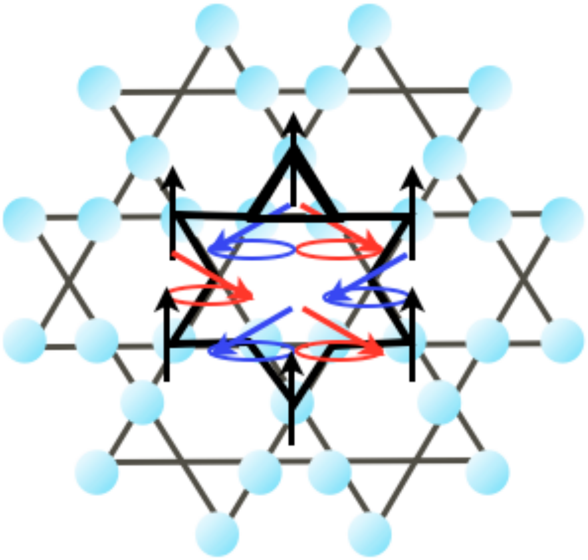
Onn et al. Phys.Rev. (1966), 156, 663.

- J_1 = NN exchange energy ~ 1.68K, J_2 & J_3 = J1/1000
- Dipole energy ~ 0.7 K - anisotropic
- Curie Weiss ~ -2.8K
- No long range order down to 25 mK.

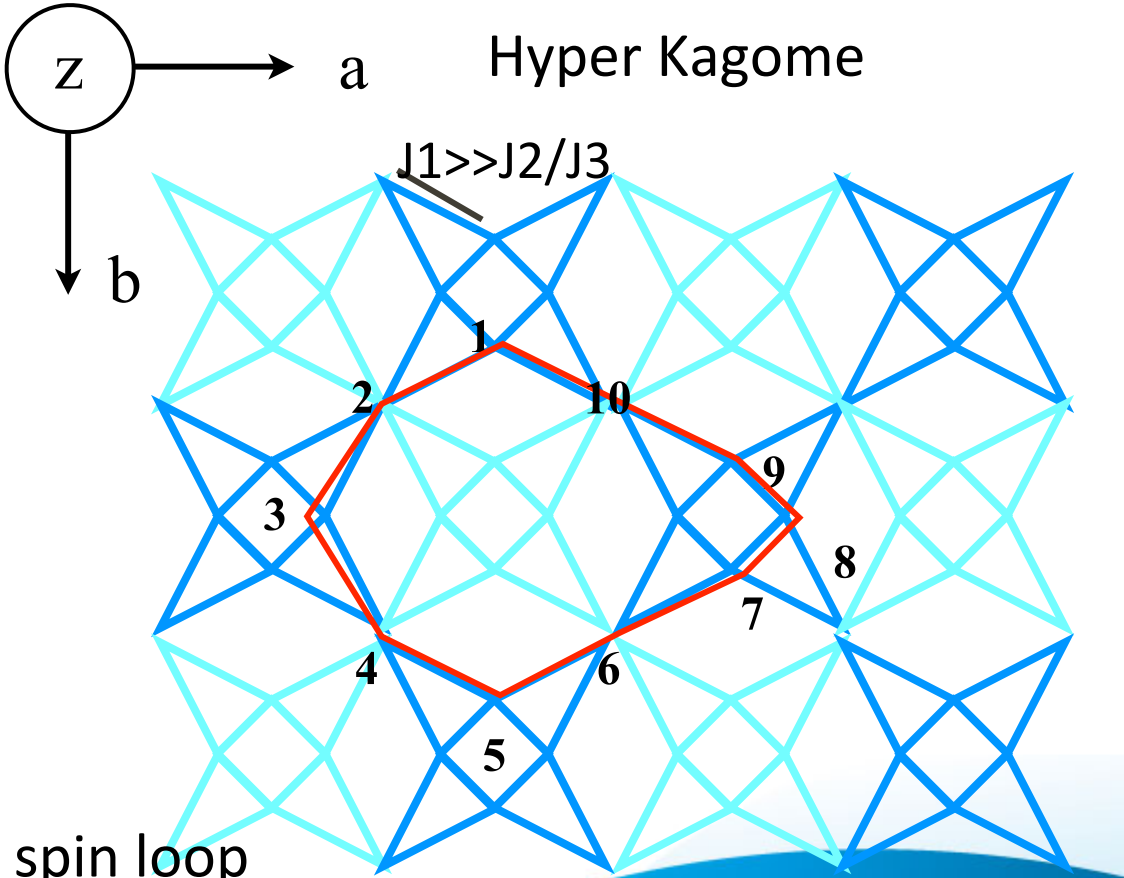
Gd$_3$Ga$_5$O$_{12}$

Soft modes

Kagome



Hyper Kagome



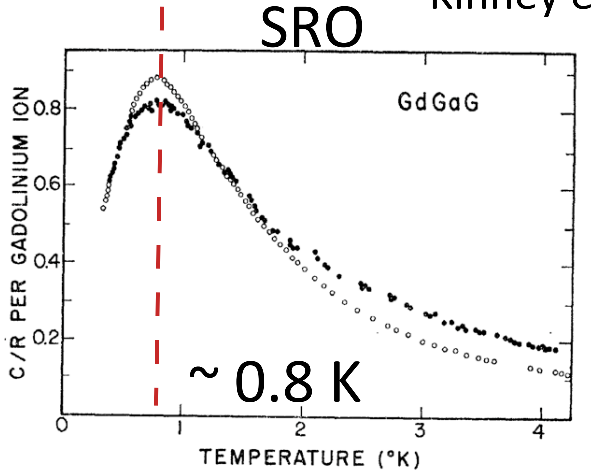
3 spin loop

10 spin loops

xx spin loop

$Gd_3Ga_5O_{12}$

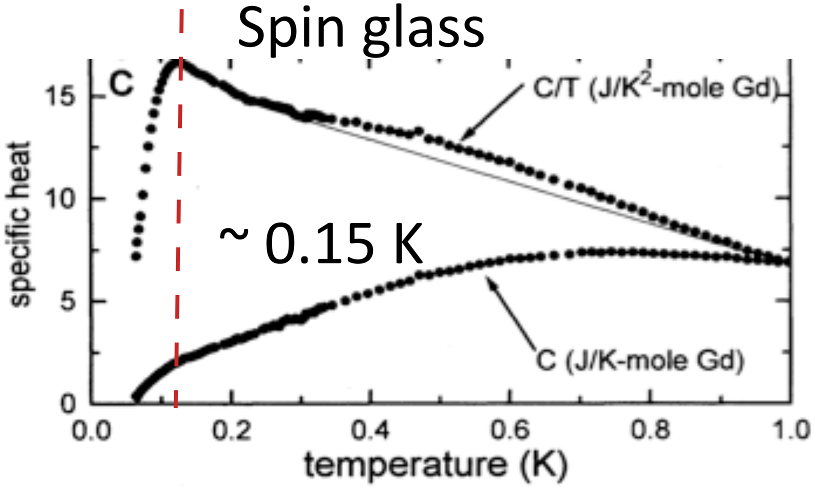
Schiffer et al. Phys. Rev. Lett (1994),73, 2500
Schiffer et al. Phys. Rev. Lett. (1995), 74, 22380
Kinney et al. J. App. Physics (1979), 50, 2115.



SRO

~ 0.8 K

FIG. 5. Specific heat of two samples of GdGaG. ● Sintered sample, ○ cluster of single crystals.



Spin glass

~ 0.15 K

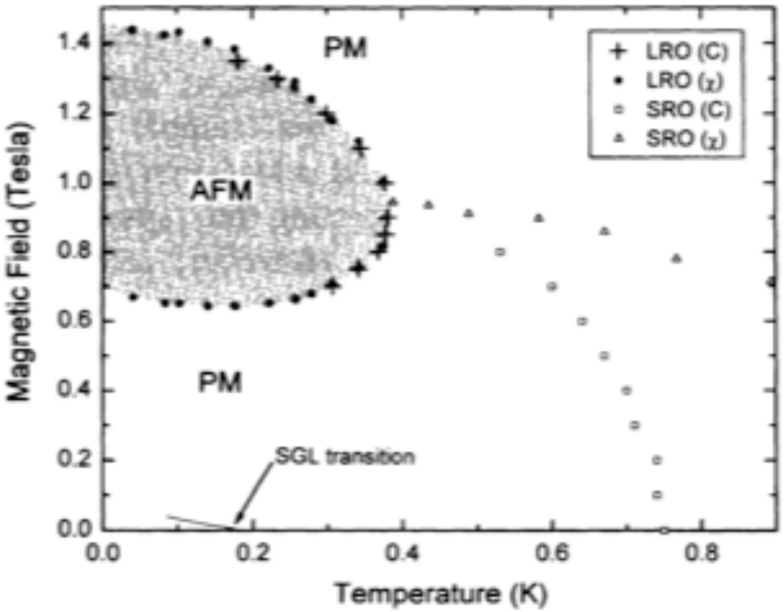
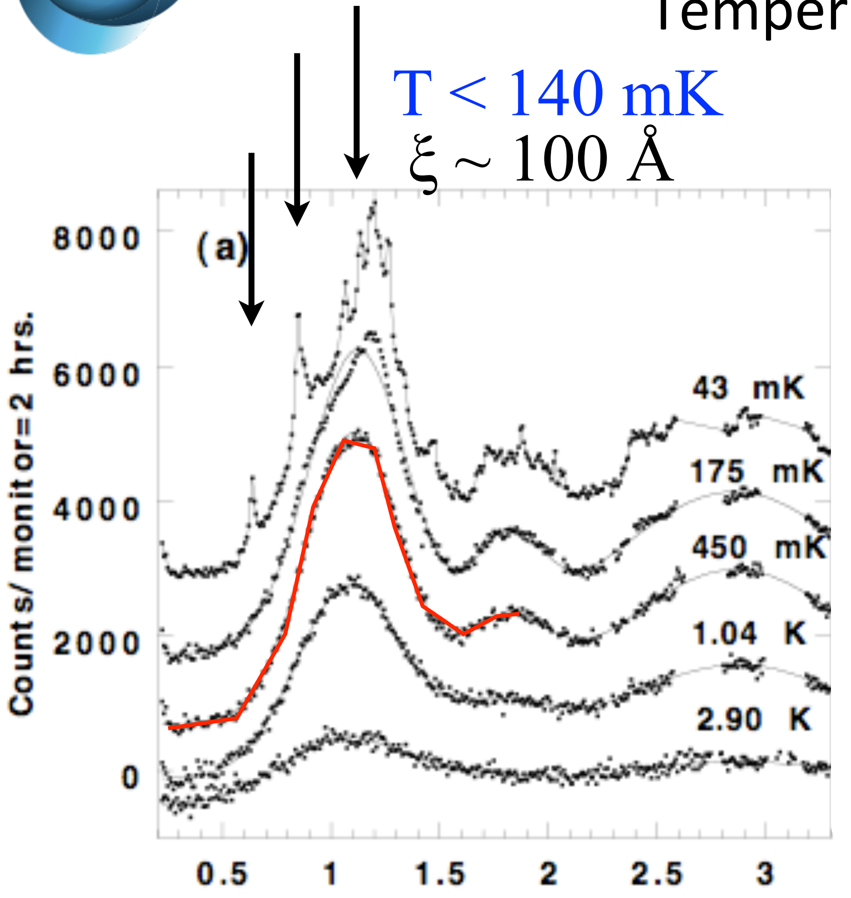


FIG. 2. The magnetic phase diagram of GGG. The long range order peaks which define the antiferromagnetic (AFM) phase boundary and the short range order peaks in the paramagnetic (PM) phase are shown. The line of maxima in χ vs T corresponding to the spin-glass-like freezing transition is also shown.

Neutron scattering

Temperature dependence

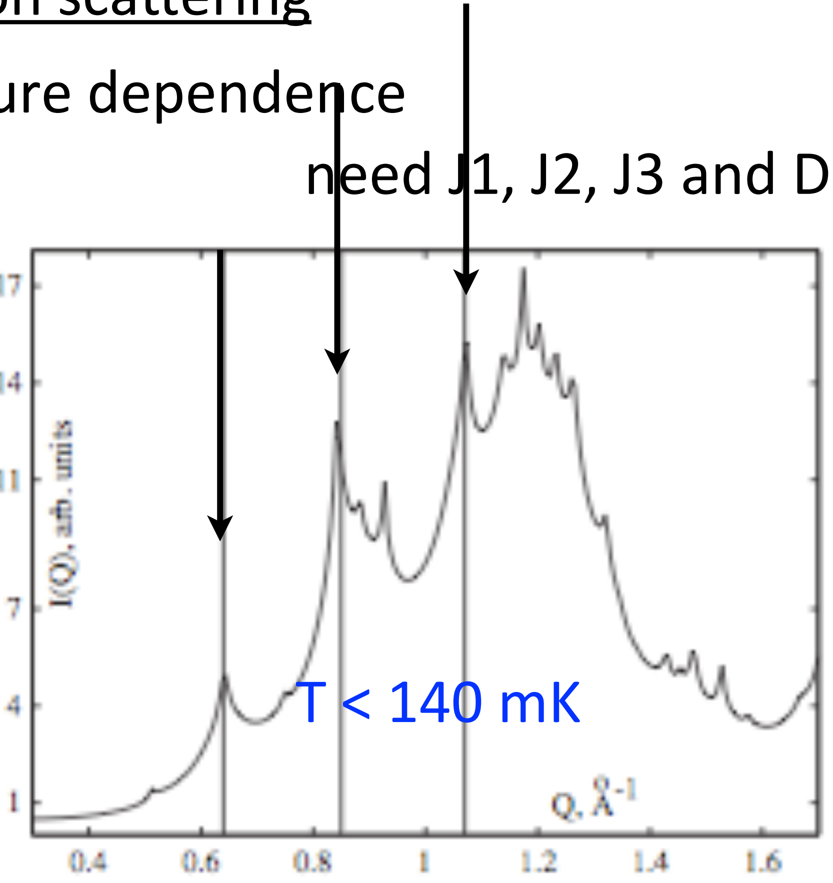


T < 140 mK

$\xi \sim 100$ Å

SRO AF order

Q, (Å^{-1})



need J1, J2, J3 and D

T < 140 mK

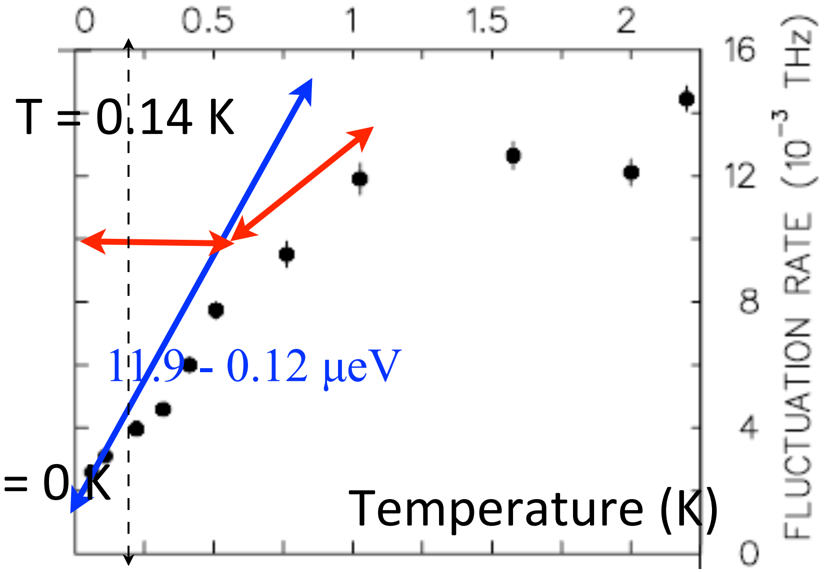
We show that it is crucial to treat accurately the <u>*long-range*</u> *nature of the magnetic dipolar interactions to allow for a determination of the small exchange energy scales involved in the selection of the experimental ordering wave vector.*

Petrenko et al. Phys. Rev. Lett. (80), 4570

T . Yavors'kii et al. Phys. Rev. Lett (97), 267203, 2006

Dynamic behaviour - spin relaxation

Pyrochlore or Kagome?



3 µeV

0.9 µeV

Finite dynamics at T = 0 K

µSR: Absence of LRO (indirect study)

Dunsiger et al. Phys. Rev. Lett. 85, 3504 (2004)

Marshall J. Phys. Cond Matt. 14, L157 (2002)

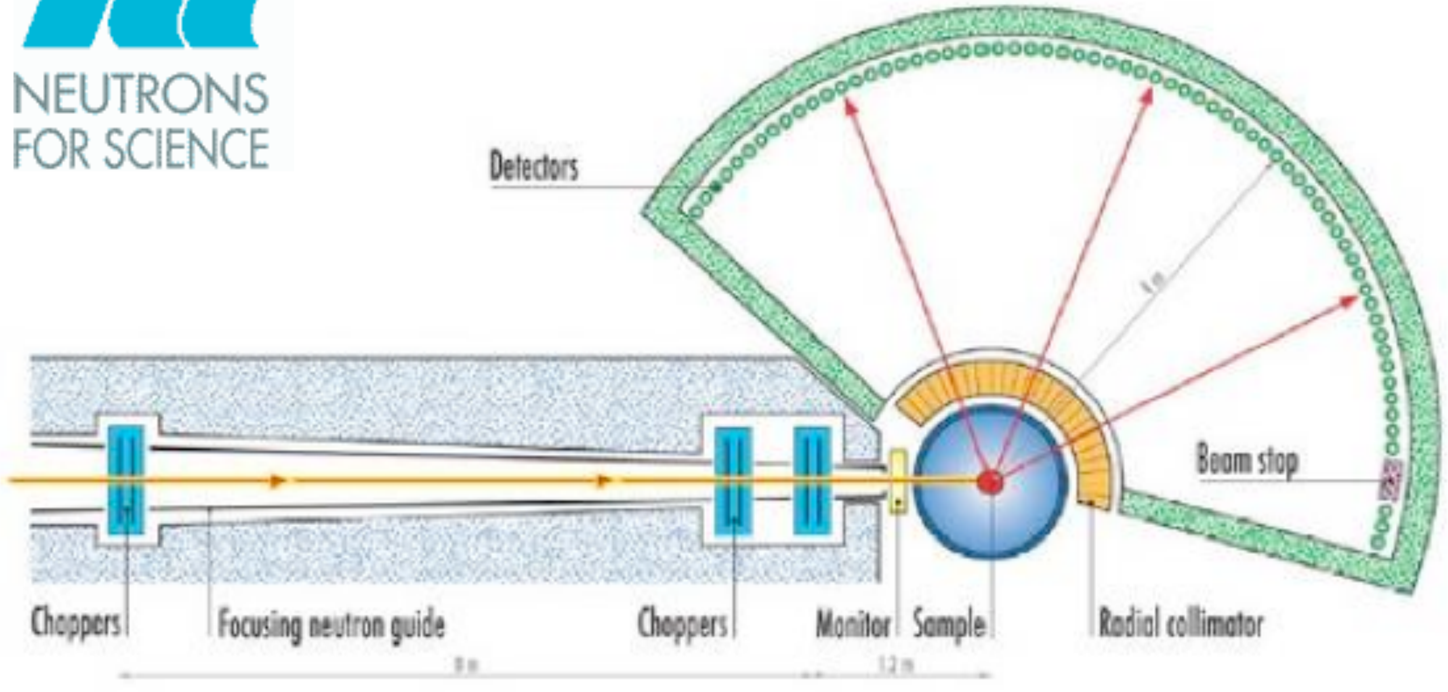
Mossbauer: Linear decrease (11.9 - 0.12 µeV) (direct study) planar confinement Bonnville Phys. Rev. Lett. 92, 167202 (2004)

Magnetic hole burning: S. Gosh Phys. Rev. Lett. 101, 157205 (2008)

T < 0.14 K protected spin clusters (10 ring) around impurity centers - pico eV dynamics !

IN5 - Cold neutron TOF spectrometer

Gain Q and E dependence

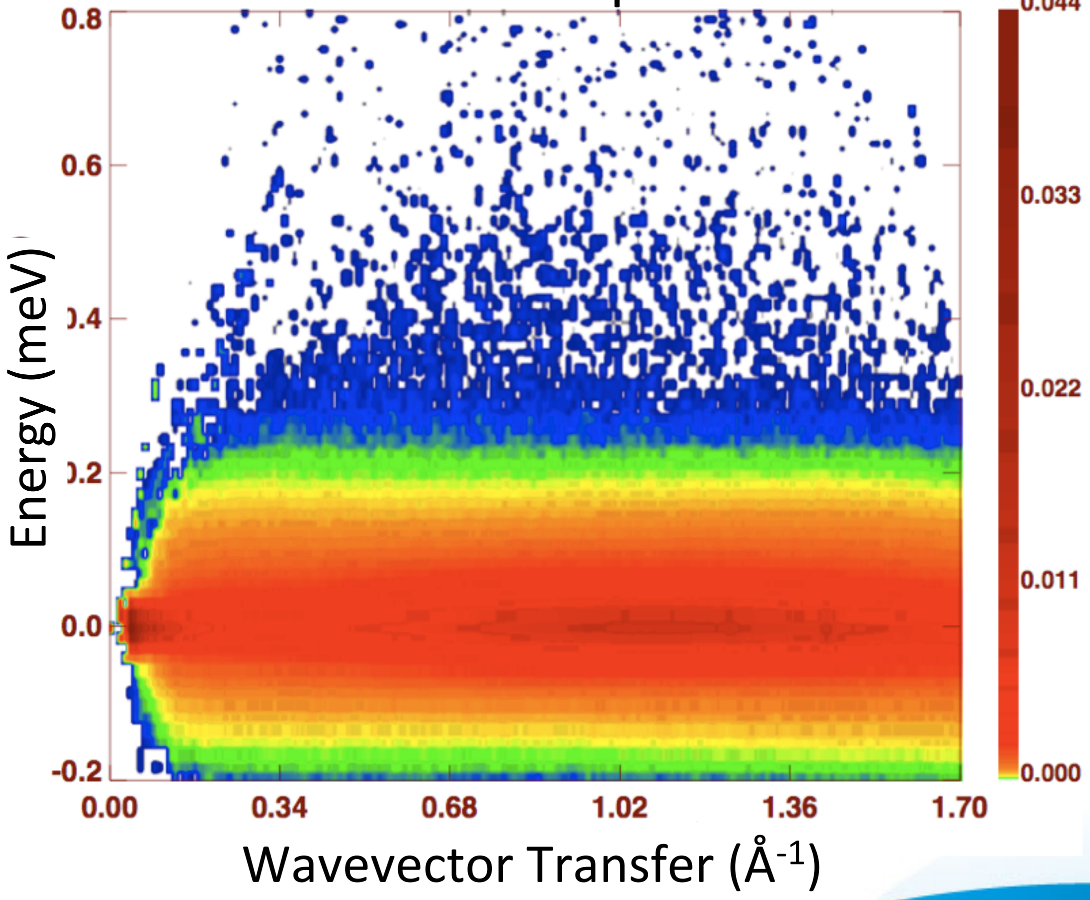


Inelastic neutron scattering (IN5) (ΔE = 50 μeV)

2 K

Powder - Isotope enriched.

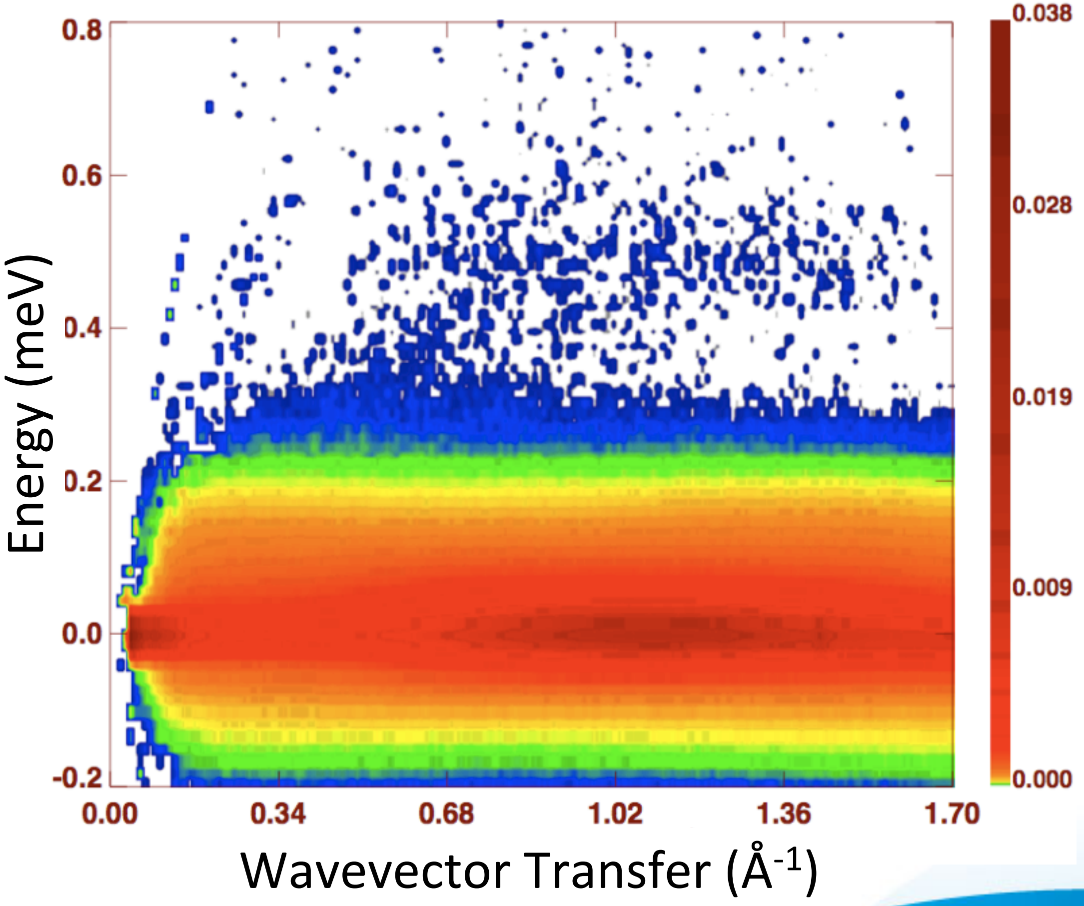
FT of spin spin autocorrelation.



Inelastic neutron scattering (IN5) (ΔE = 50 μeV)

1.2 K

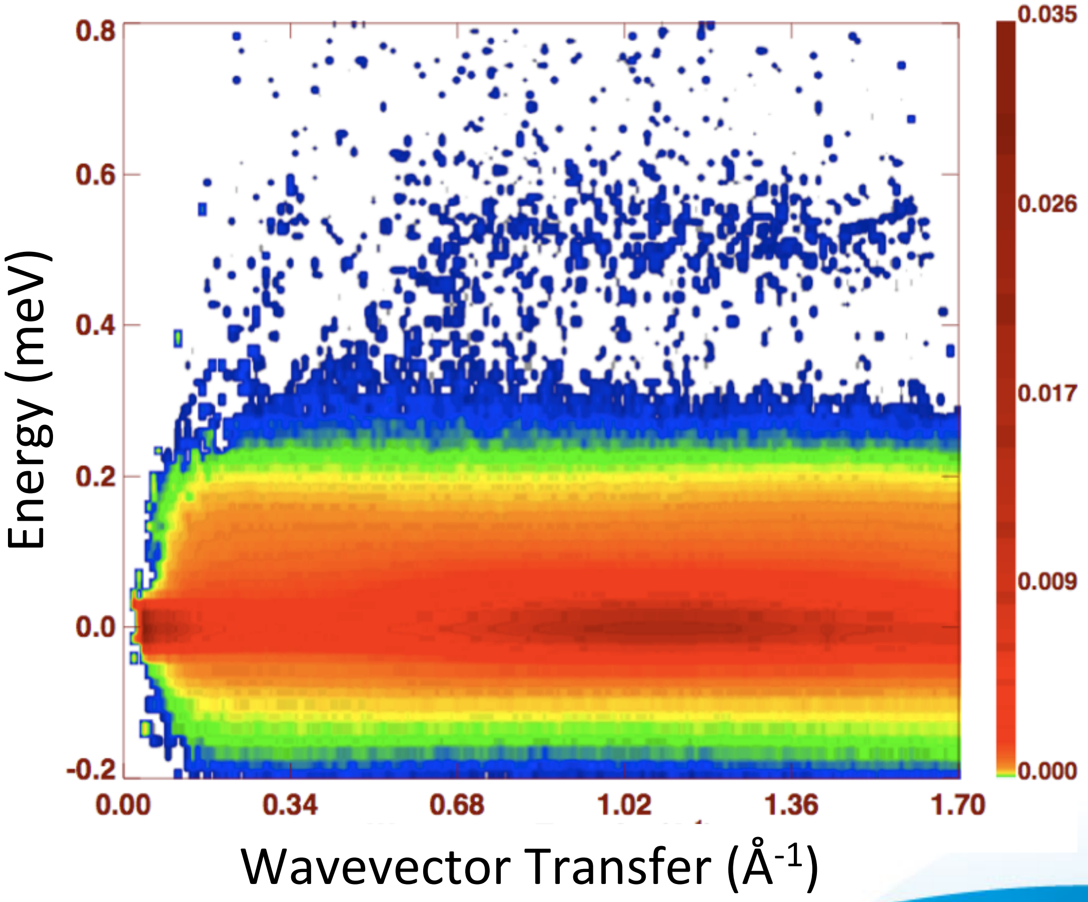
FT of spin spin autocorrelation.



Inelastic neutron scattering (IN5) (ΔE = 50 μeV)

0.9 K

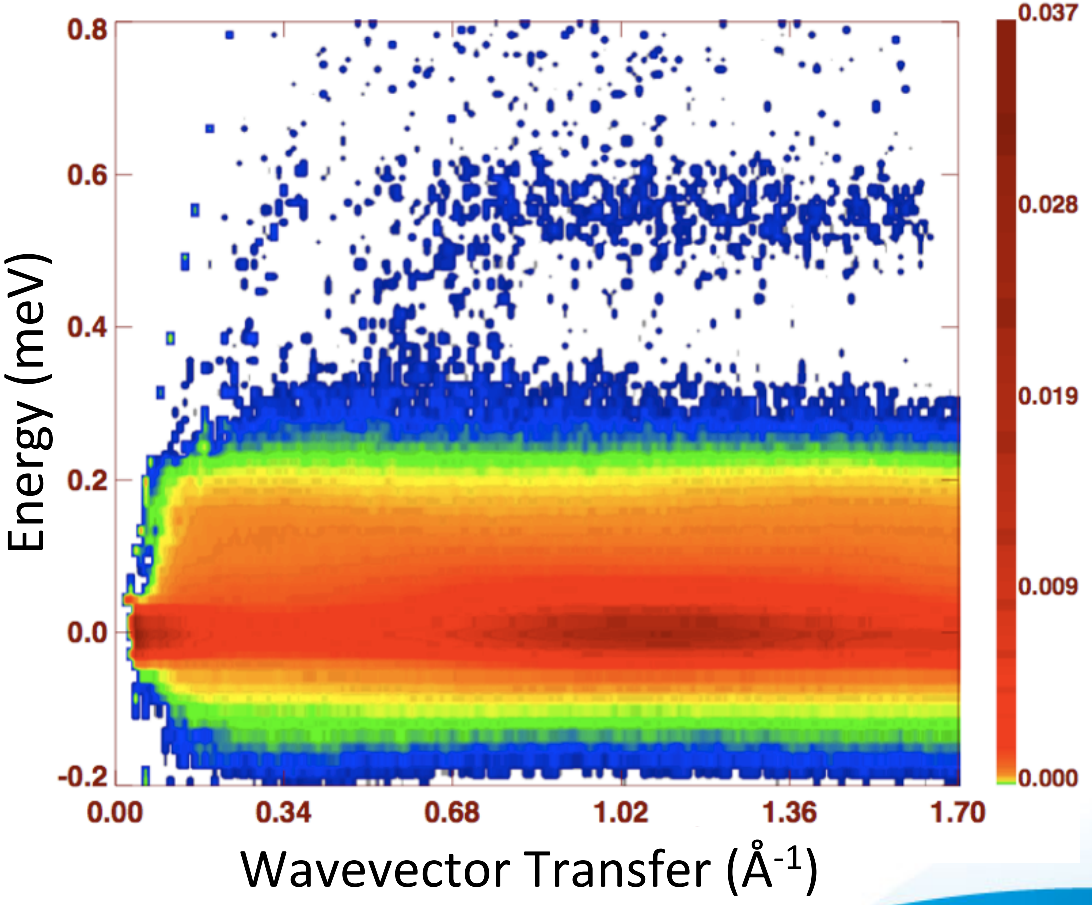
FT of spin spin autocorrelation.



Inelastic neutron scattering (IN5) (ΔE = 50 μeV)

0.6 K

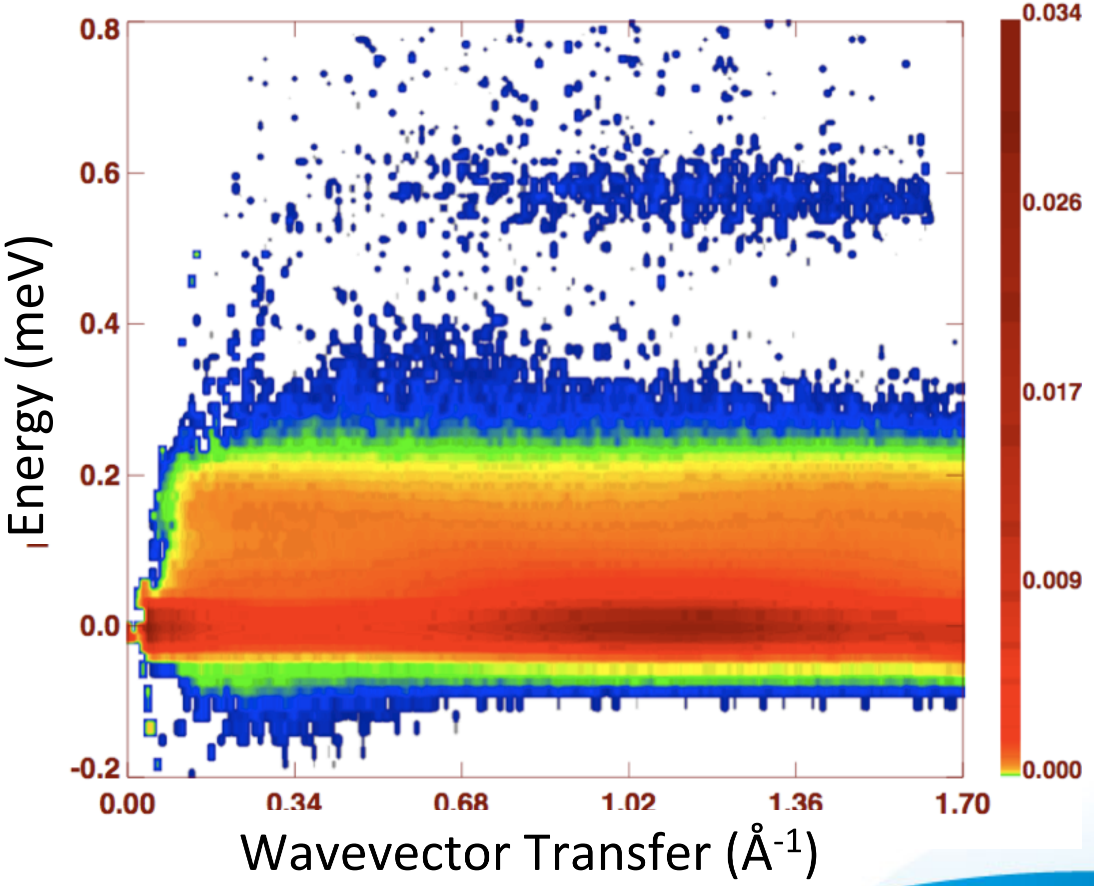
FT of spin spin autocorrelation.



Inelastic neutron scattering (IN5) (ΔE = 50 μeV)

0.25 K

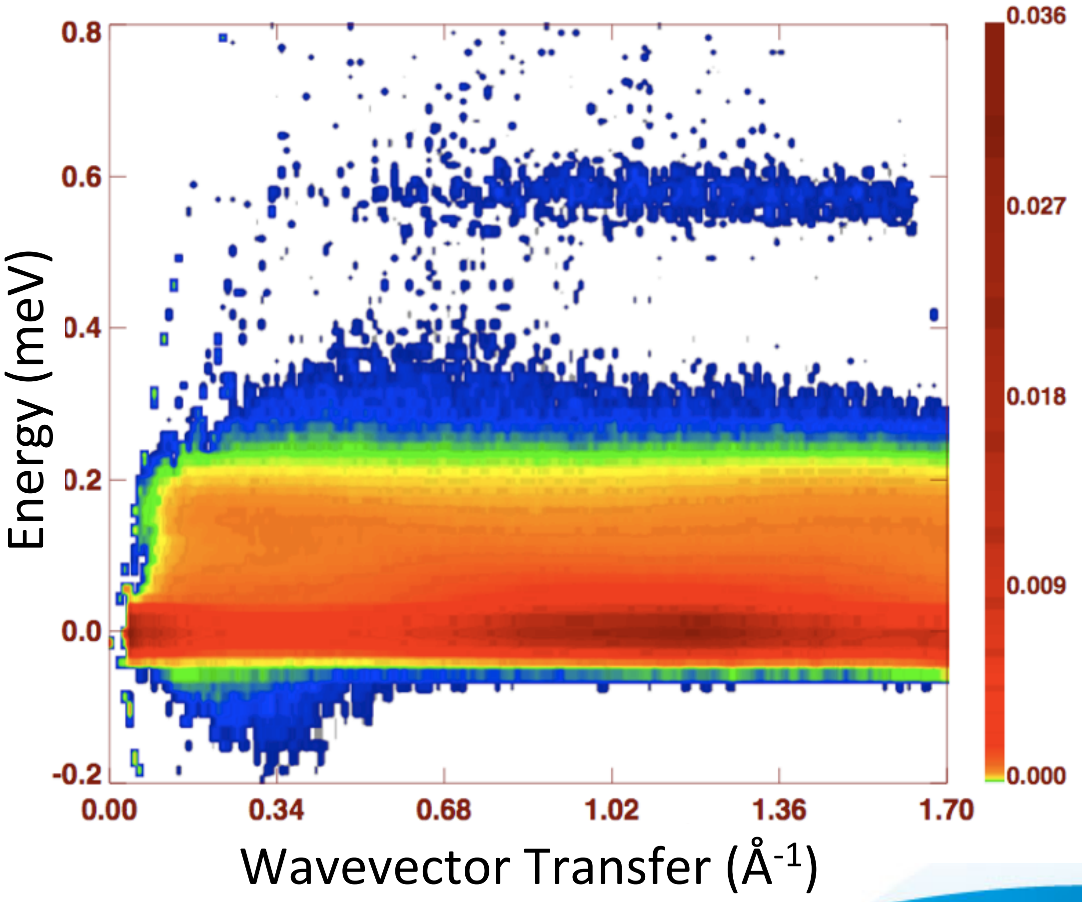
FT of spin spin autocorrelation.



Inelastic neutron scattering (IN5) (ΔE = 50 μeV)

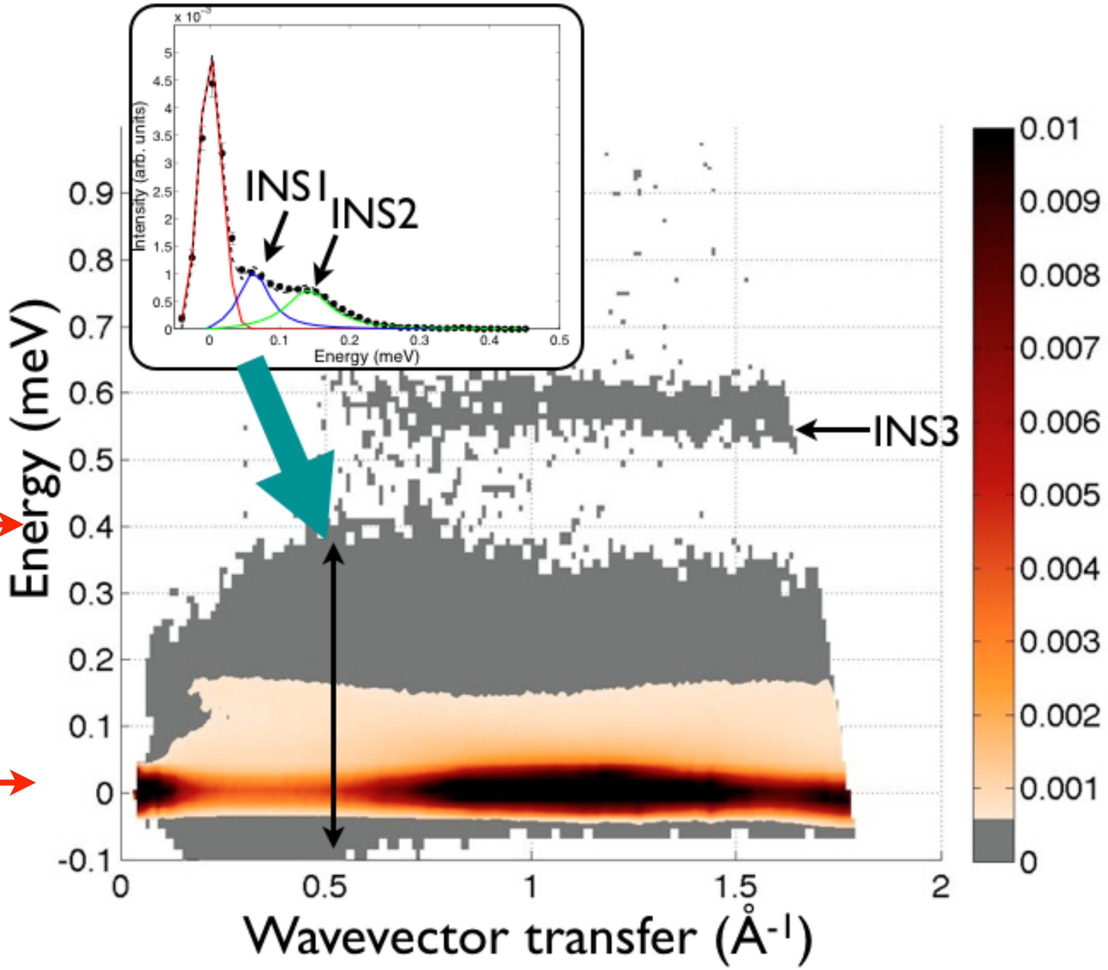
0.06 K (Below 0.14 K transition)

FT of spin spin autocorrelation.



1st inelastic neutron scattering data on a frustrated 3D Kagome compound.

After some analysis



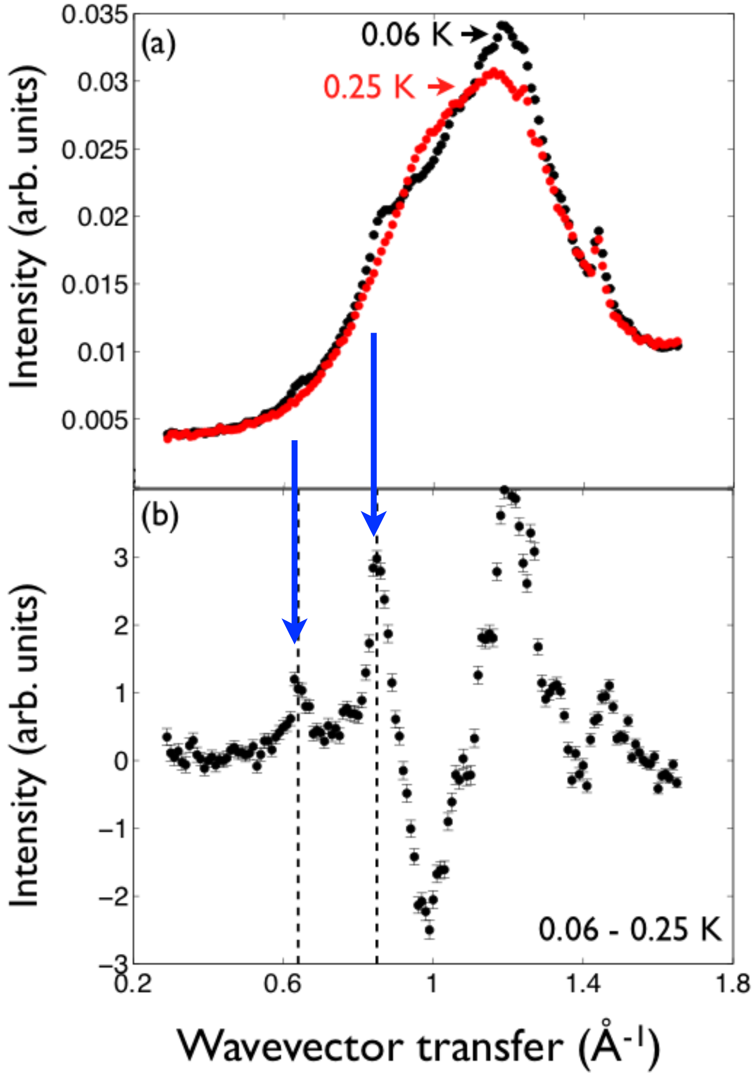
Inelastic

Elastic

3 - gapped non-dispersive excitations

INS1 = 0.04, INS2 = 0.14 and INS3 = 0.58 meV

Elastic scattering

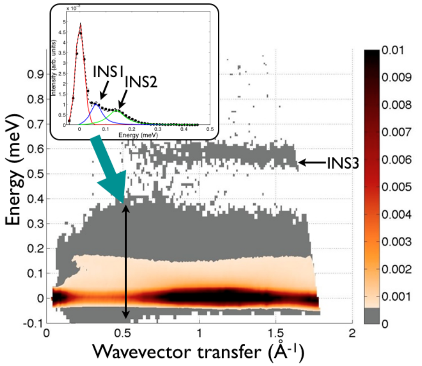


82 % of the total scattering intensity

Incommensurate order
(T < 0.14 K) within elastic line
Frozen order is indeed frozen (within resolution)
No spin wave excitations.

Inelastic scattering

Non-dispersive:
crystal fields?
Local vibrational excitations?

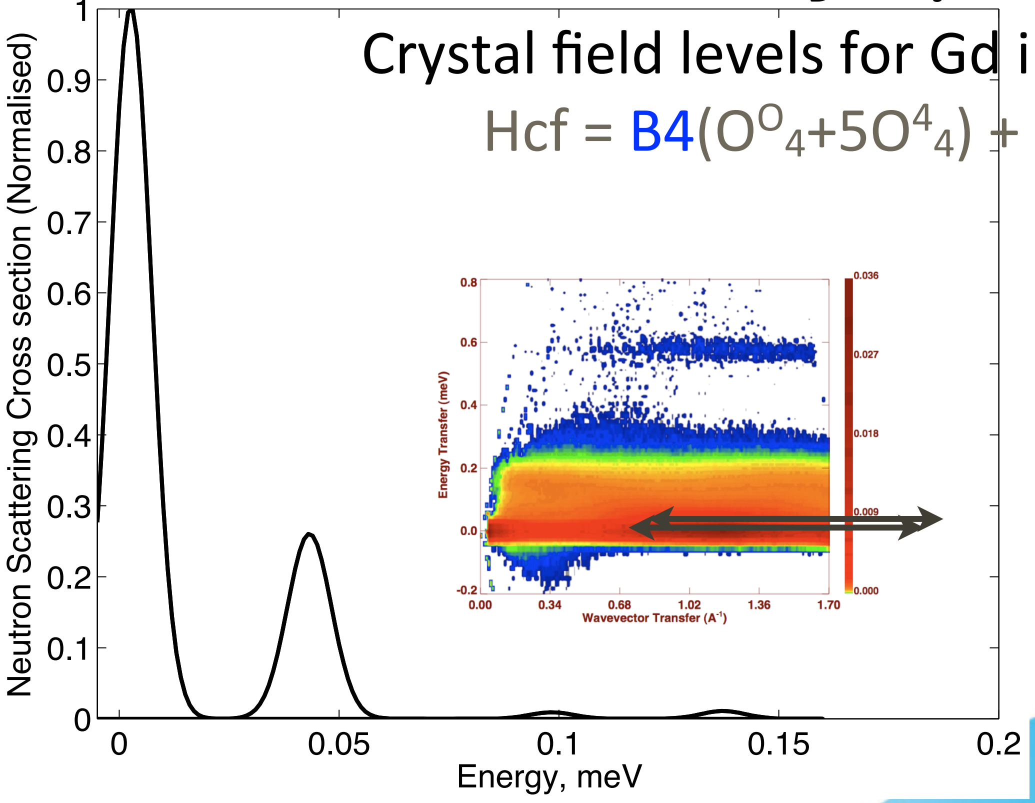


Newman et al. J. Phys. C. (8),1862, 1975

$$H = H_D + H_J + H_{CF} + H_Z$$

Crystal field levels for Gd in garnet structure.

$$Hcf = B4(O^0_4+5O^4_4) + B6(O^0_6-21O^4_6)$$



Cannot be reconciled with bulk measurements

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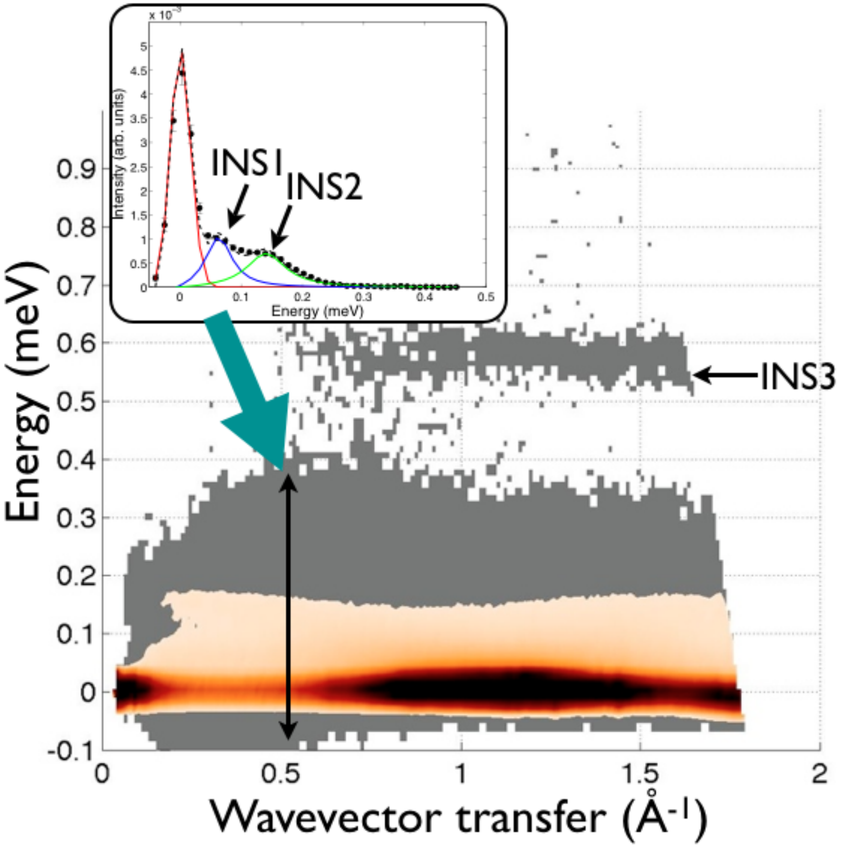
Data Analysis



Inelastic →

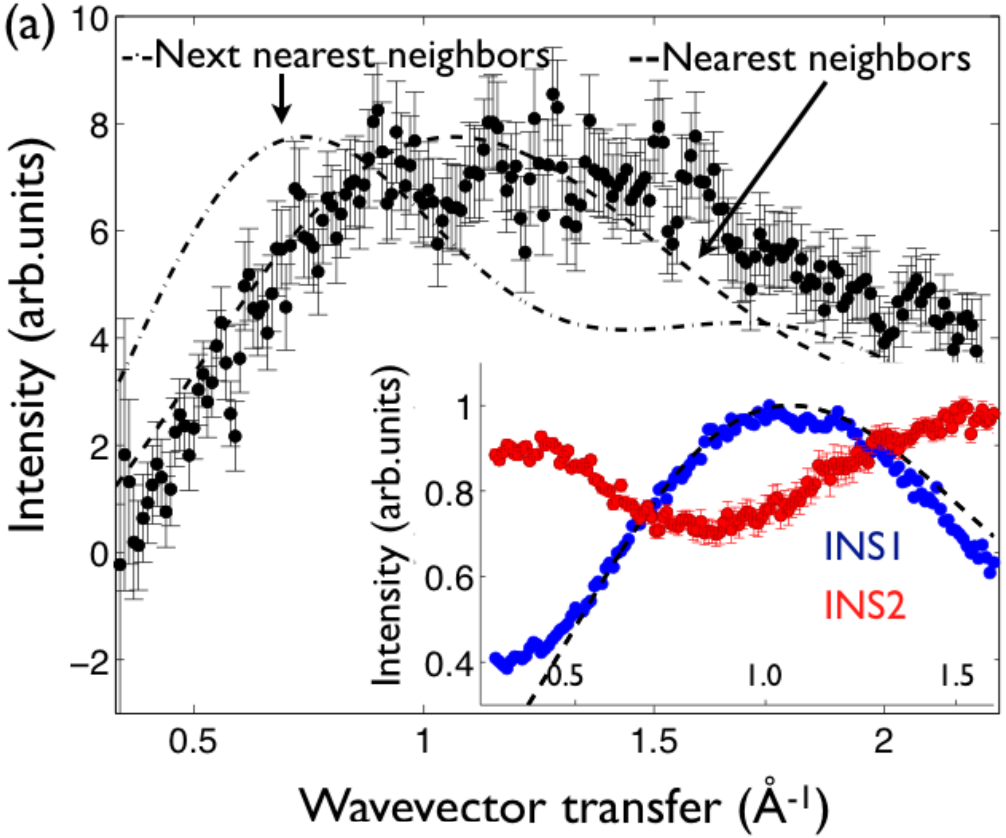
Elastic →

Q-dependence - integrate over inelastic line at each position of Q.

- Q - dependence 60 mK.
- Temperature dependence.

Inelastic scattering 60 mK - possible origin

AF short range correlations



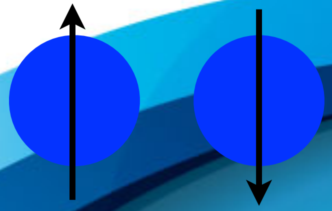
INS1 ✓

INS2 ✗

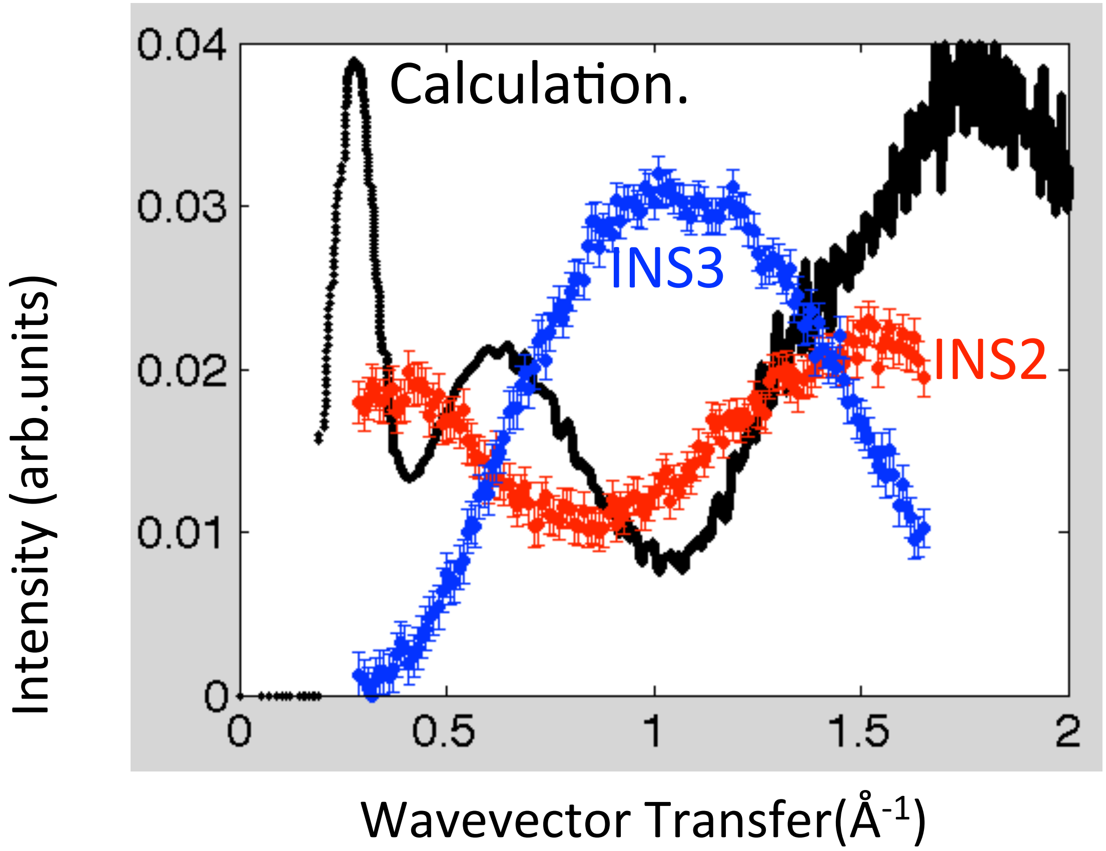
INS3 ✓

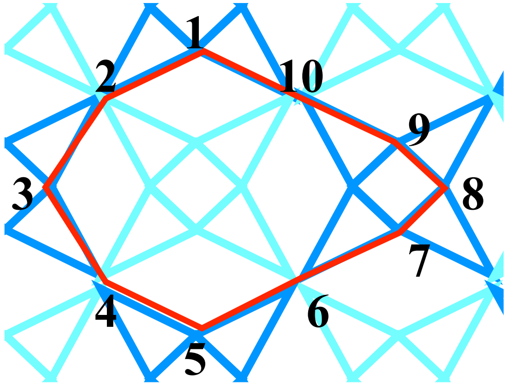
$$\frac{d^2\sigma}{d\Omega d\omega} \propto A(T) F^2(Q) \left[1 - \frac{\sin(Qd)}{Qd}\right]$$

$J_{NN} \sim 1.68$ K



Inelastic scattering 60 mK - 10 spin cluster?





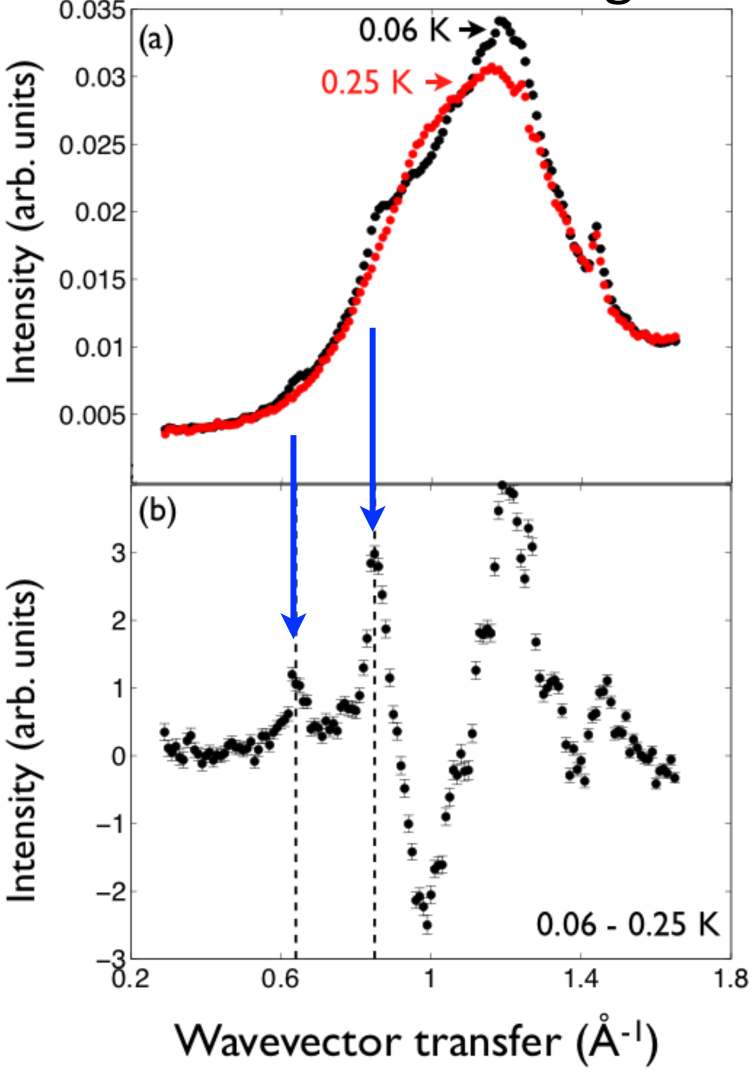
INS3:Not a 10 spin cluster

INS2: close to 10 spin cluster

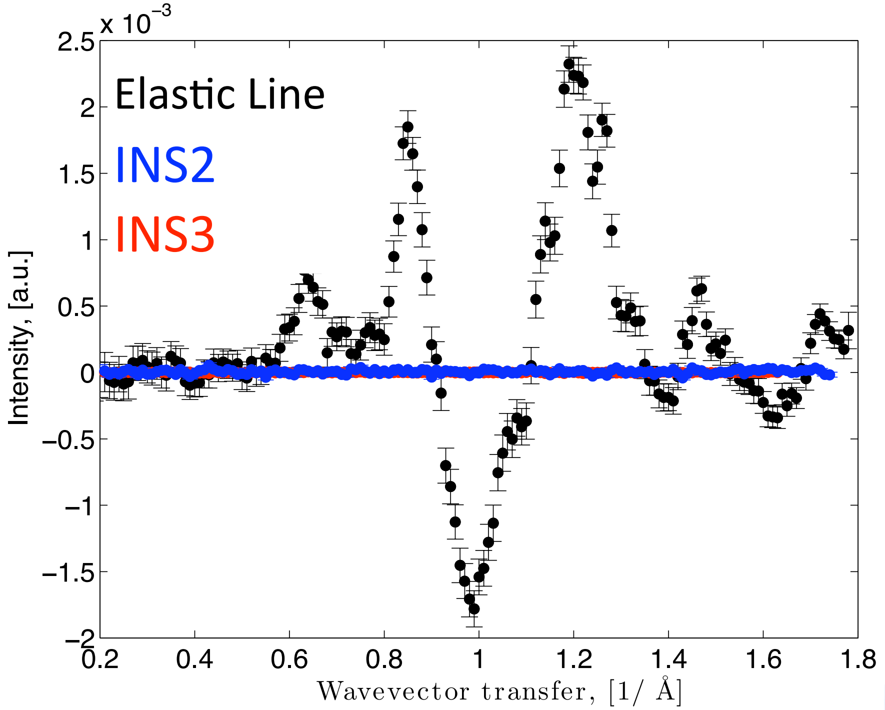
Inelastic scattering :Temperature dependence

60 mK - 250 mK (Across 0.14 K transition)

Elastic scattering



Inelastic scattering



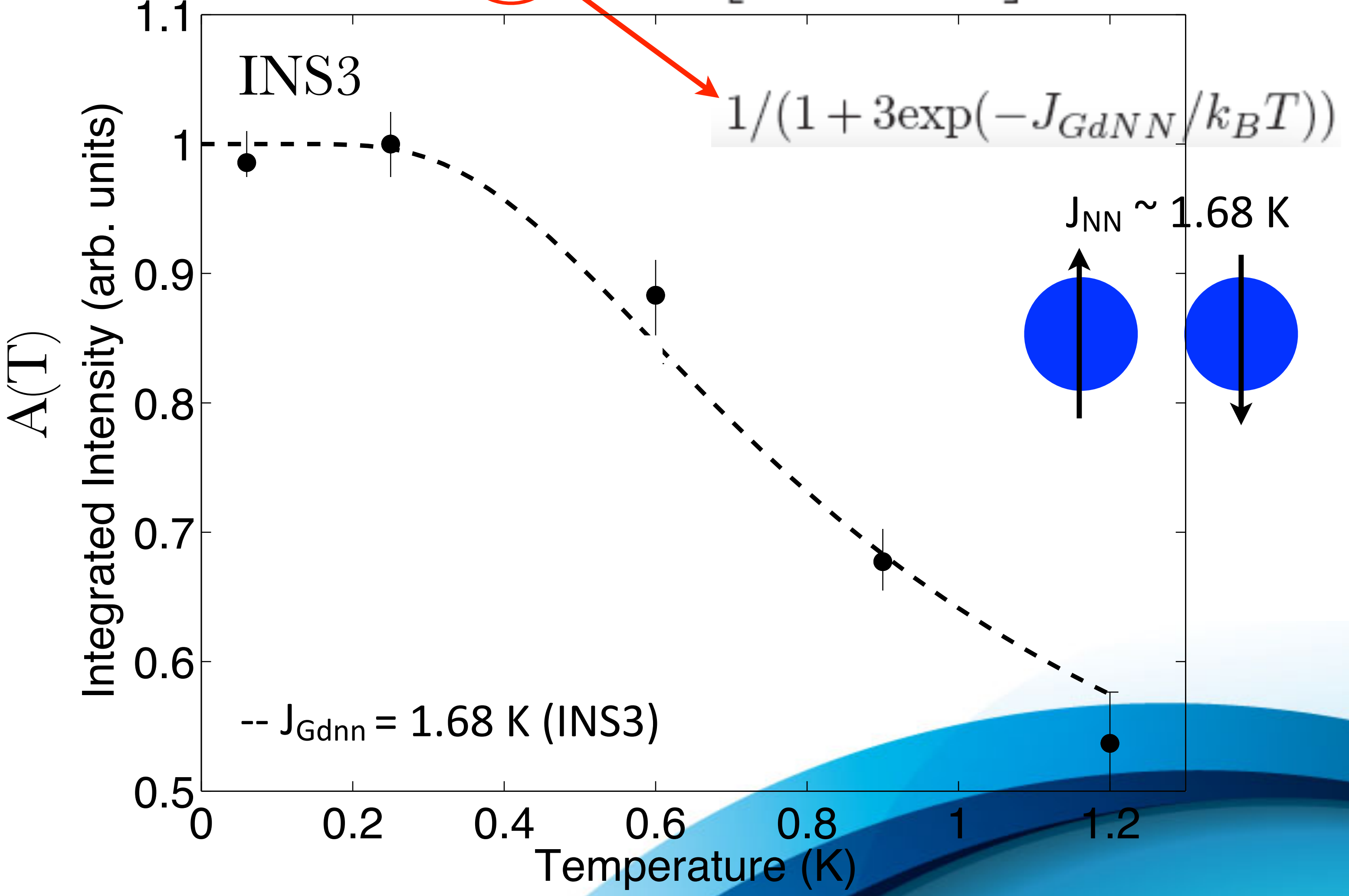
Excitations not affected by longer range order

Inelastic scattering :Temperature dependence

(Thermal distribution of singlet to triplet excitations)

$$\frac{d^2\sigma}{d\Omega d\omega} \propto A(T) F^2(Q) \left[1 - \frac{\sin(Qd)}{Qd}\right]$$

$1/(1+3\exp(-J_{GdNN}/k_BT))$

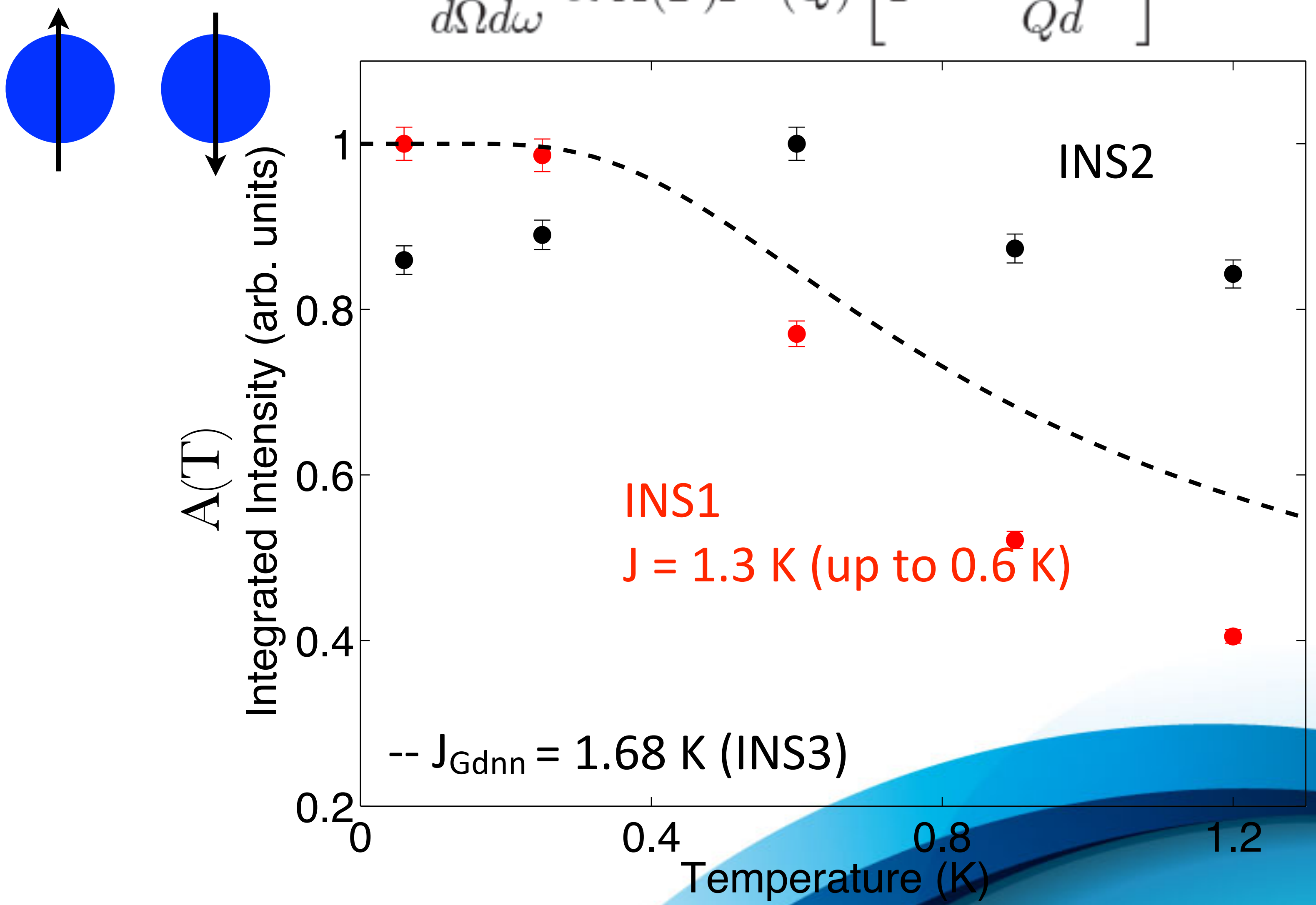


Inelastic scattering :Temperature dependence

(Thermal distribution of singlet to triplet excitations)

J_{NN} ~ 1.68 K

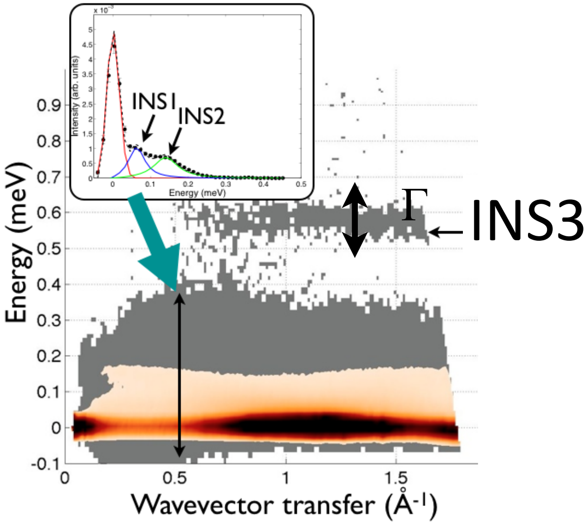
$$\frac{d^2\sigma}{d\Omega d\omega} \propto A(T)F^2(Q)\left[1 - \frac{\sin(Qd)}{Qd}\right]$$



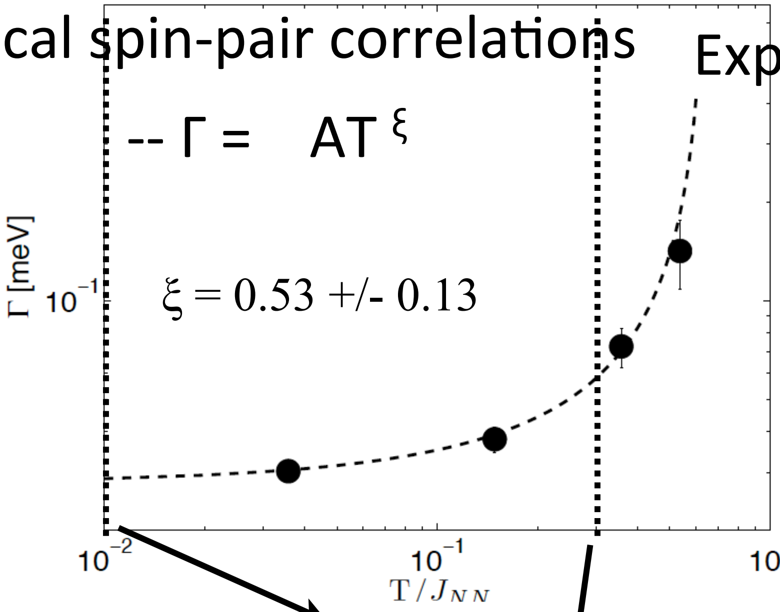
Inelastic scattering :Temperature dependence

INS3 Inelastic Width

Dynamical spin-pair correlations



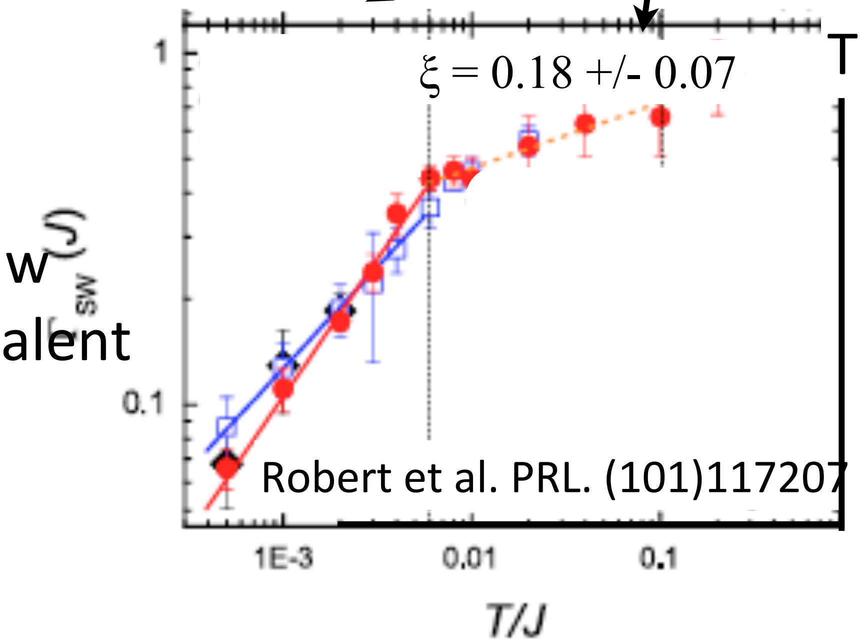
Experimental



-- $\Gamma = AT^{\xi}$

$\xi = 0.53 +/- 0.13$

Theory



$\xi = 0.18 +/- 0.07$

Robert et al. PRL. (101)117207

Scattering follows algebraic law

$\Gamma = AT^{\xi}$ but exponent not equivalent

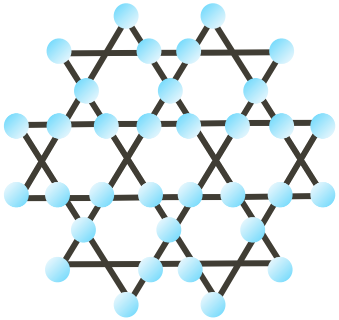
Robert only calculated
J Near neighbour

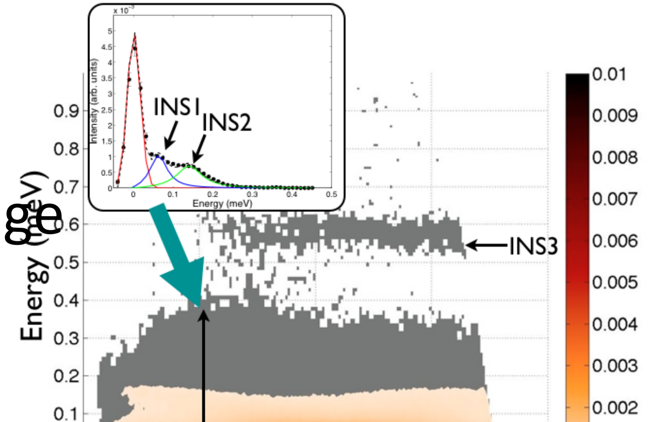
Further Theory for Kagome

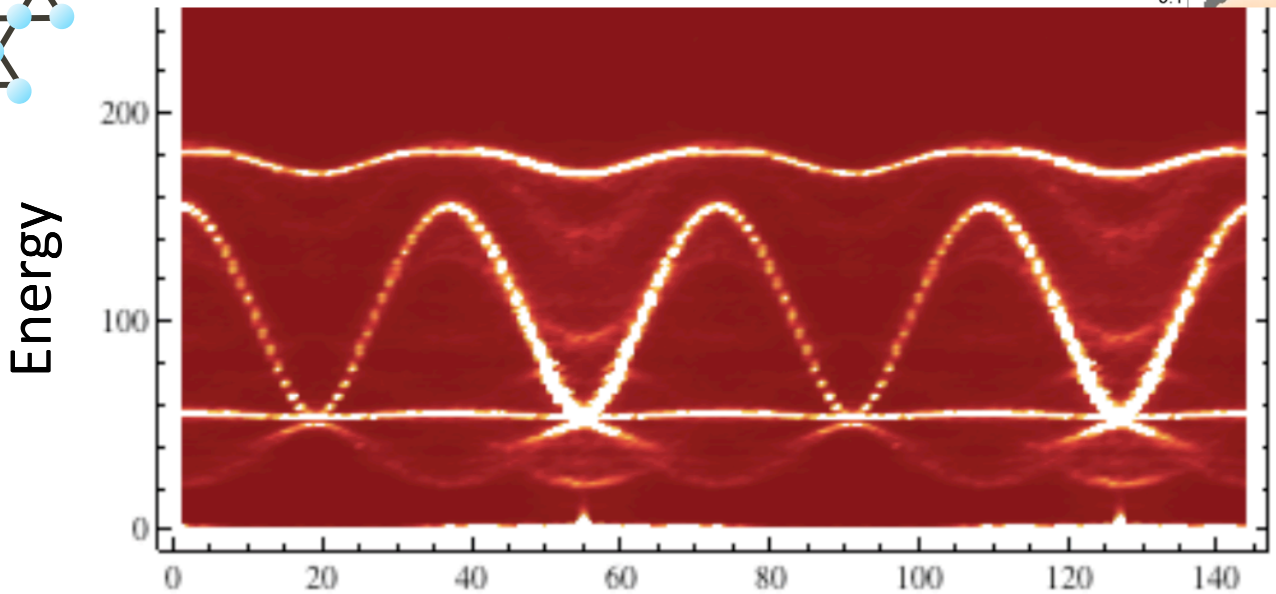
B. Canals

Near neighbour and dipole exchange interactions only

Are these soft modes?

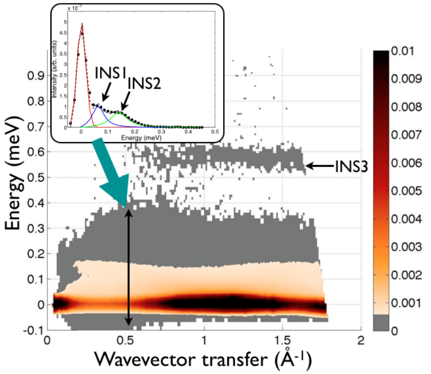






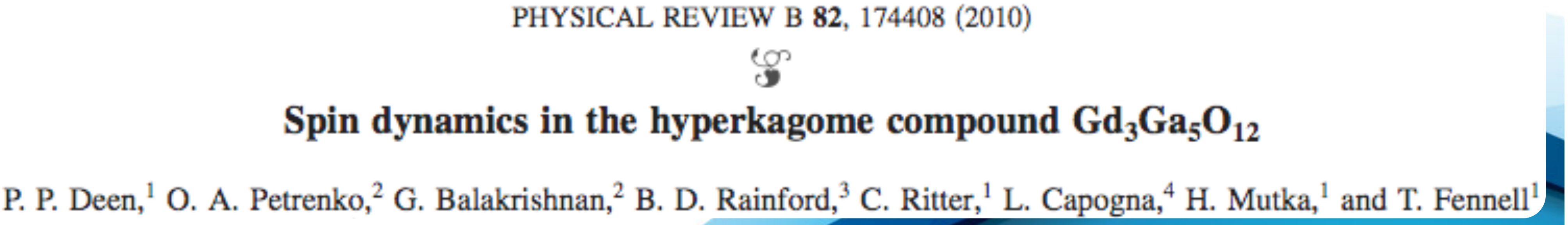
- Always 3 clear modes
- Lowest always flat
- Second always dispersive
- Appears that J_1 only drives model into a coplanar gapped modes (+D = gapped)
- Role of temperature is not important.

Some Conclusions



- No continuum of excitations in 3D Kagome.
- Soft modes exist in a zero field disordered state
- Data consistent with AF short range exchange (3 spin)
- Excitations affected by Jnn - SRO.
- Long range order does not perturb excitations.

- Are these soft modes lifted by anisotropy?
- Must include Anisotropic dipole interactions.
- No spin waves?



PHYSICAL REVIEW B **82**, 174408 (2010)

Spin dynamics in the hyperkagome compound $Gd_3Ga_5O_{12}$

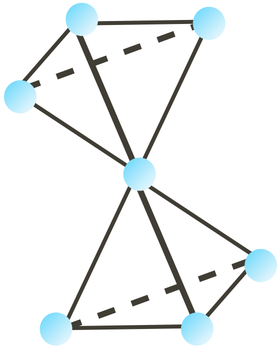
P. P. Deen,[1] O. A. Petrenko,[2] G. Balakrishnan,[2] B. D. Rainford,[3] C. Ritter,[1] L. Capogna,[4] H. Mutka,[1] and T. Fennell[1]

Frustration ⇒ zero point entropy ⇒ degeneracy

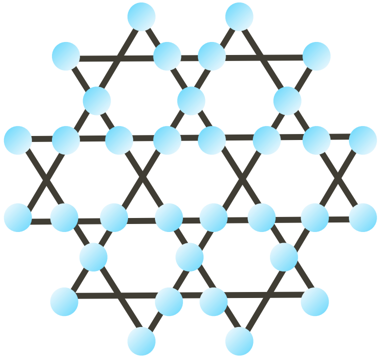
Universal dynamic behaviour?

It would appear not.

Pyrochlore



Kagome/3D Kagome



Dynamic gapless excitations to 0 K.

ΔE ~T .

Depends on T not J.

3 Gapped excitations

Algebraic relation for ΔE .

Weak temperature dependence

Dynamic range - meV - picoeV

Thank you.

Spectroscopy into the future

The study of dynamics in condensed matter with inelastic neutron scattering provides one of the most exacting tests of the understanding of the microscopic origin of the material properties, particularly when combined with powerful computer modelling techniques now being pioneered.

LET (ISIS) Website

Scientific & Sociatal Impact

Magnetism

Electron -phonon coupling
Quantum phase transition
New classes of quantum criticality
Frustrated magnetism
Electron, magnetic and SC order
Strongly electron correlations
Giant/Collosal Magnetoresistance
Molecular magnetism

Chemical Physics

Confinement
Collective behaviour in liquids and glasses
Hydrogen storage
Molecular dynamics of living cells
Targeted drug delivery

Spectroscopy into the future

A few examples

Weak continuum scattering

Polarisation analysis

Variable resolution

Theory groups

Data analysis

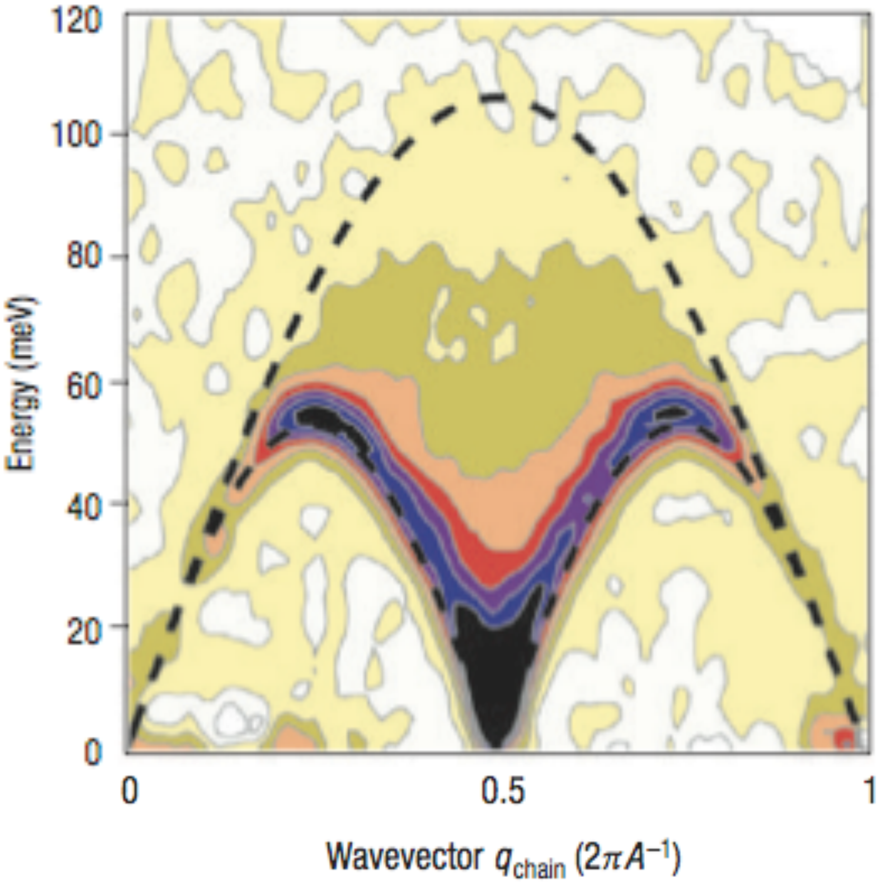
Sample characterisation
(In-situ and out)

(I) Large areas of S(Q,ω)

Coming from the magnetism angle

Total S(Q,ω) $KCuF_3$

B.Lake et al. Nature Materials (2005)



Weakly coupled 1D spin chains
-- predicted multi-spinon continuum.

model for carbon nanotube

Dynamic behaviour close to quantum criticality

Scattering extends over a broad region of S(Q,ω).
Variable resolution.

Valuable and complex physics sits within the "background".

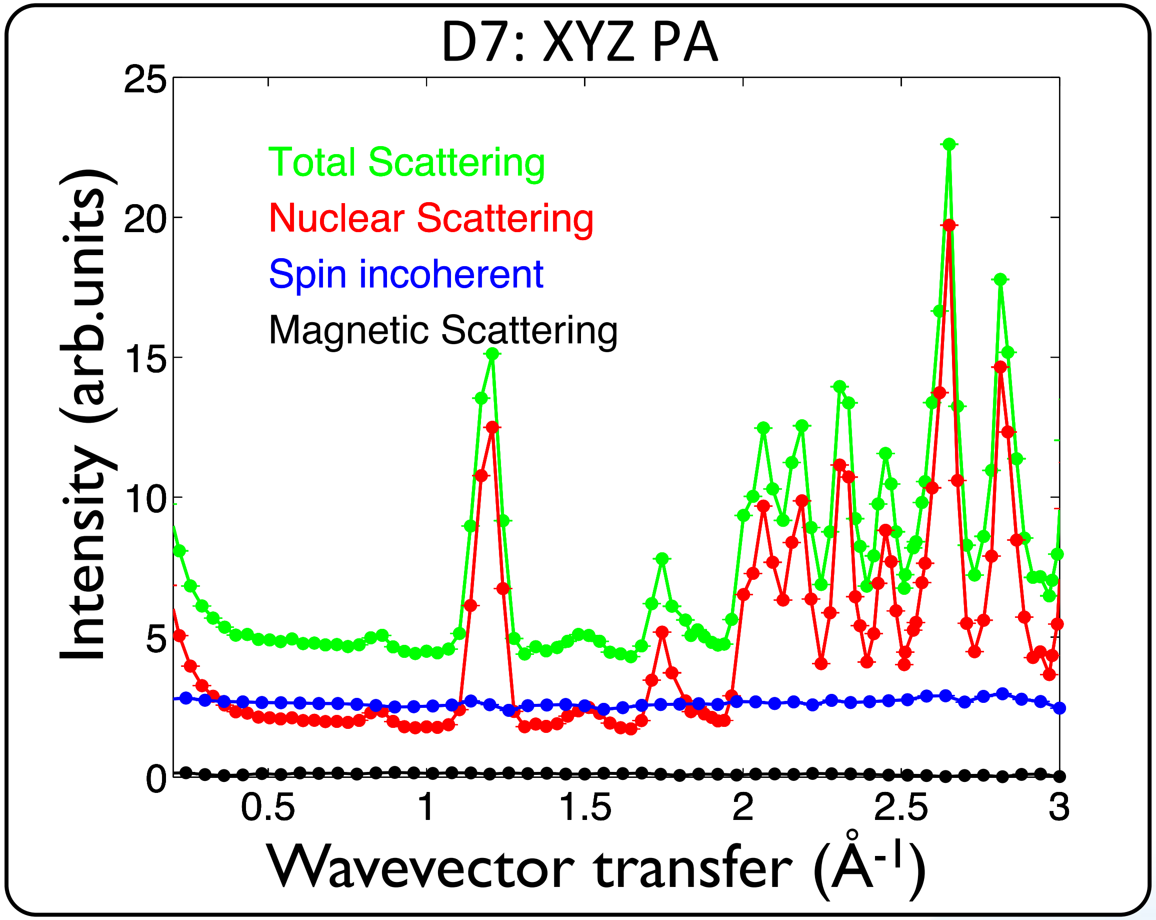
(II) PA

EUROPEAN
SPALLATION
SOURCE

Pair Correlations, Short Range Order and Dispersive Excitations in the Quasi-Kagome Quantum Magnet Volborthite

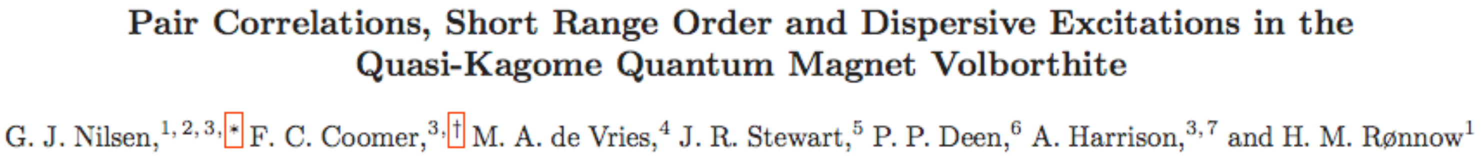
G. J. Nilsen,[1, 2, 3, *] F. C. Coomer,[3, †] M. A. de Vries,[4] J. R. Stewart,[5] P. P. Deen,[6] A. Harrison,[3, 7] and H. M. Rønnow[1]

arXiv:1001.2462



S = 1/2 Kagome Heisenberg antiferromagnet
(True quantum spin liquid?)

(II) PA



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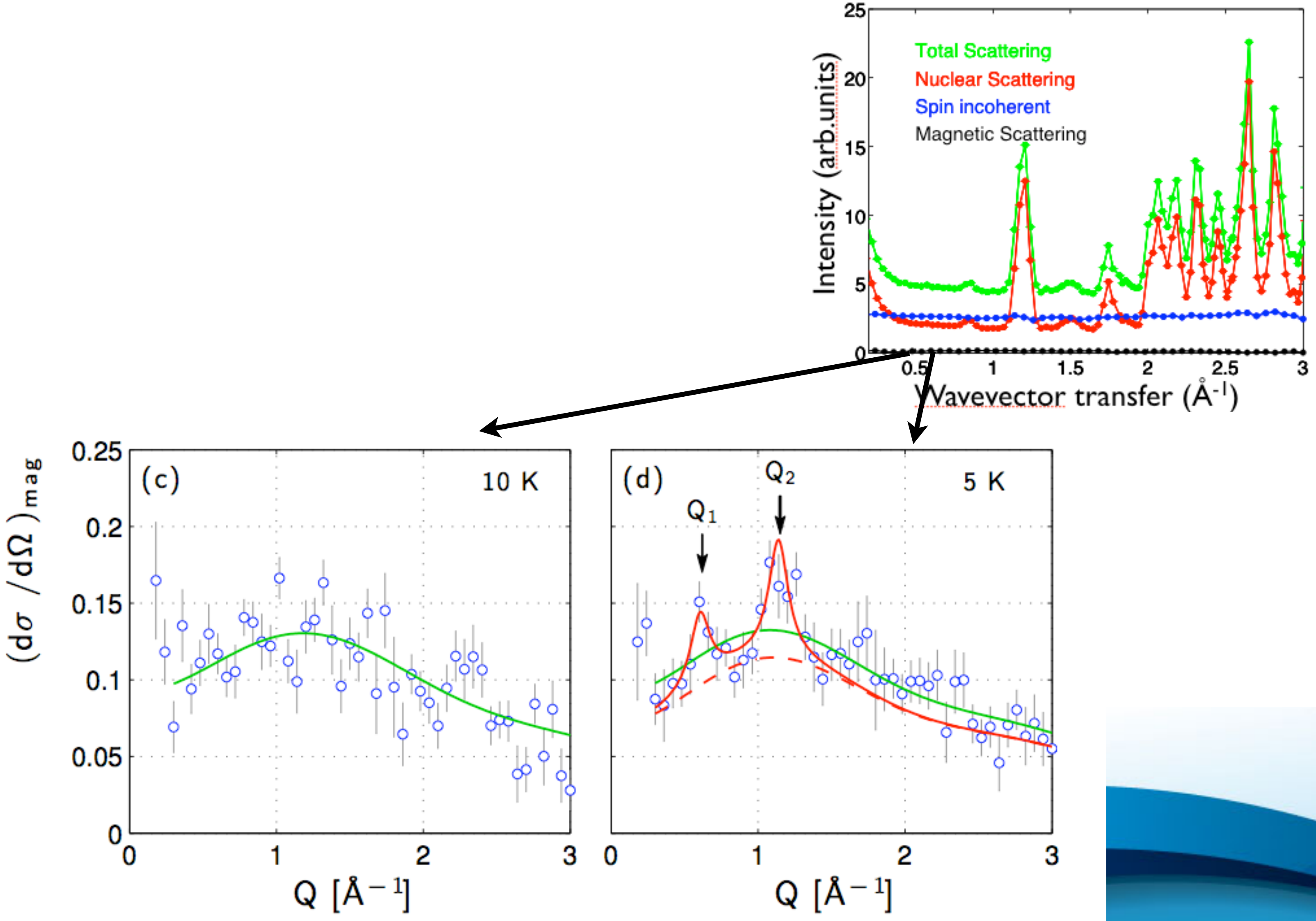
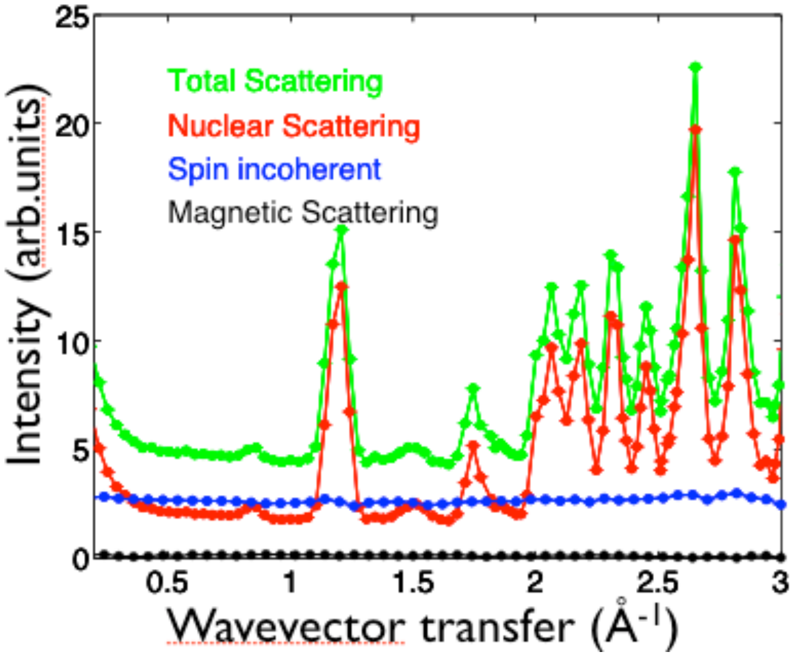


(II) PA

Polarisation analysis

Polarisation analysis is imperative in the study of magnetic order.



The unambiguous observation of magnetic continuum scattering is highly nontrivial,because of the difficulty of isolating weak magnetic continua from background and phonon contributions.

Definite proof that additional scattering is magnetic can only come from polarisation analysis.

N. B. Christensen et al. PNAS, vol. 104, p. 15264, 2007.

<u>However</u> other communities are less enthusiastic!

Loss of flux is not appreciated

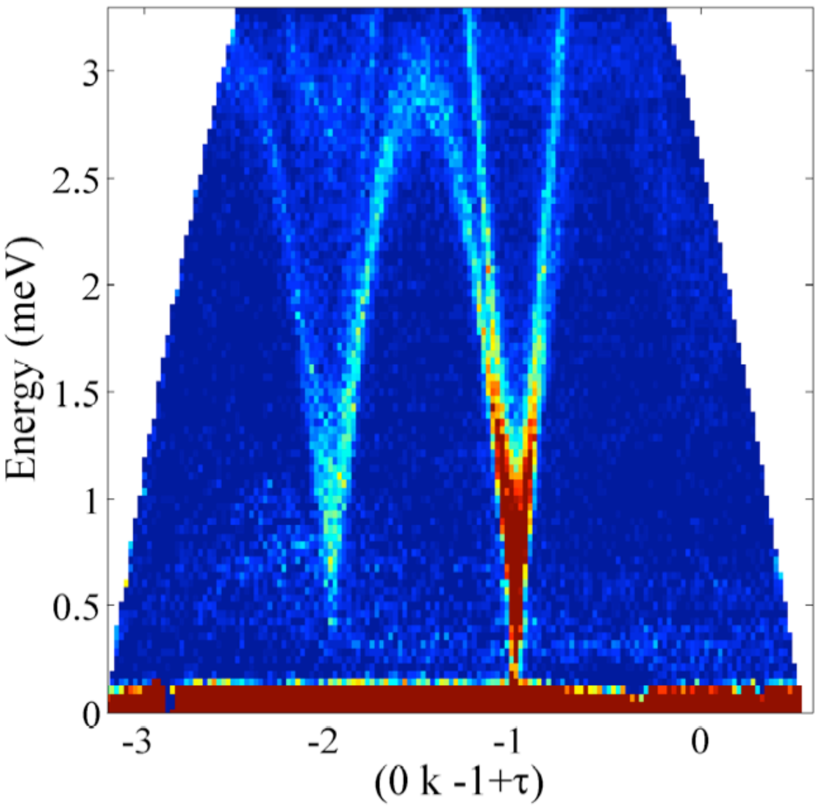
(II)(b) PA

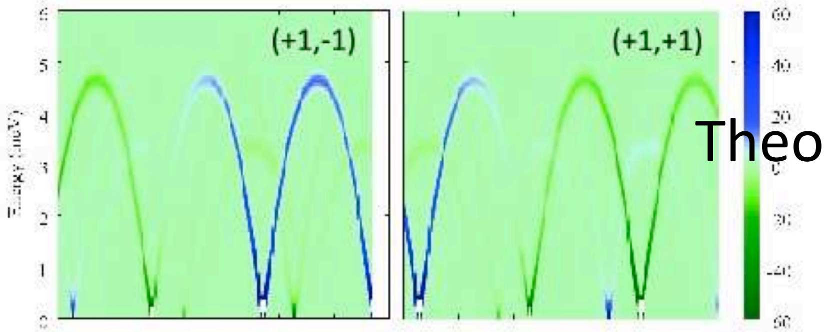
When magnetic chirality gets excited: spin waves in $Ba_3NbFe_3Si_2O_{14}$

M. Loire,[1] V. Simonet,[1] S. Petit,[2] K. Marty,[1,3] P. Bordet,[1] P. Lejay,[1] J. Ollivier,[4] M. Enderle,[4] P. Steffens,[4] E. Ressouche,[5] A. Zorko,[6] and R. Ballou[1,*]

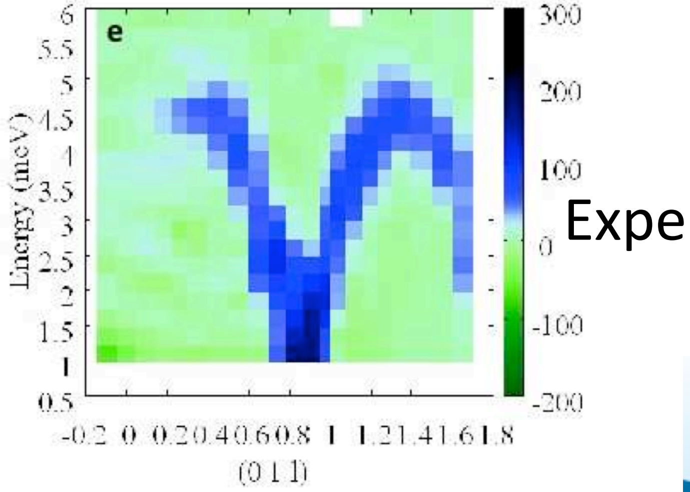
IN20 (CRYOPAD) - PA

IN5 (ILL)





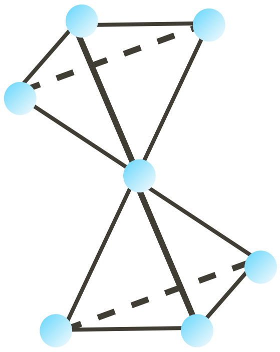
Theory



Experiment

http://horace.isis.rl.ac.uk/

(III)Variable resolution



Science

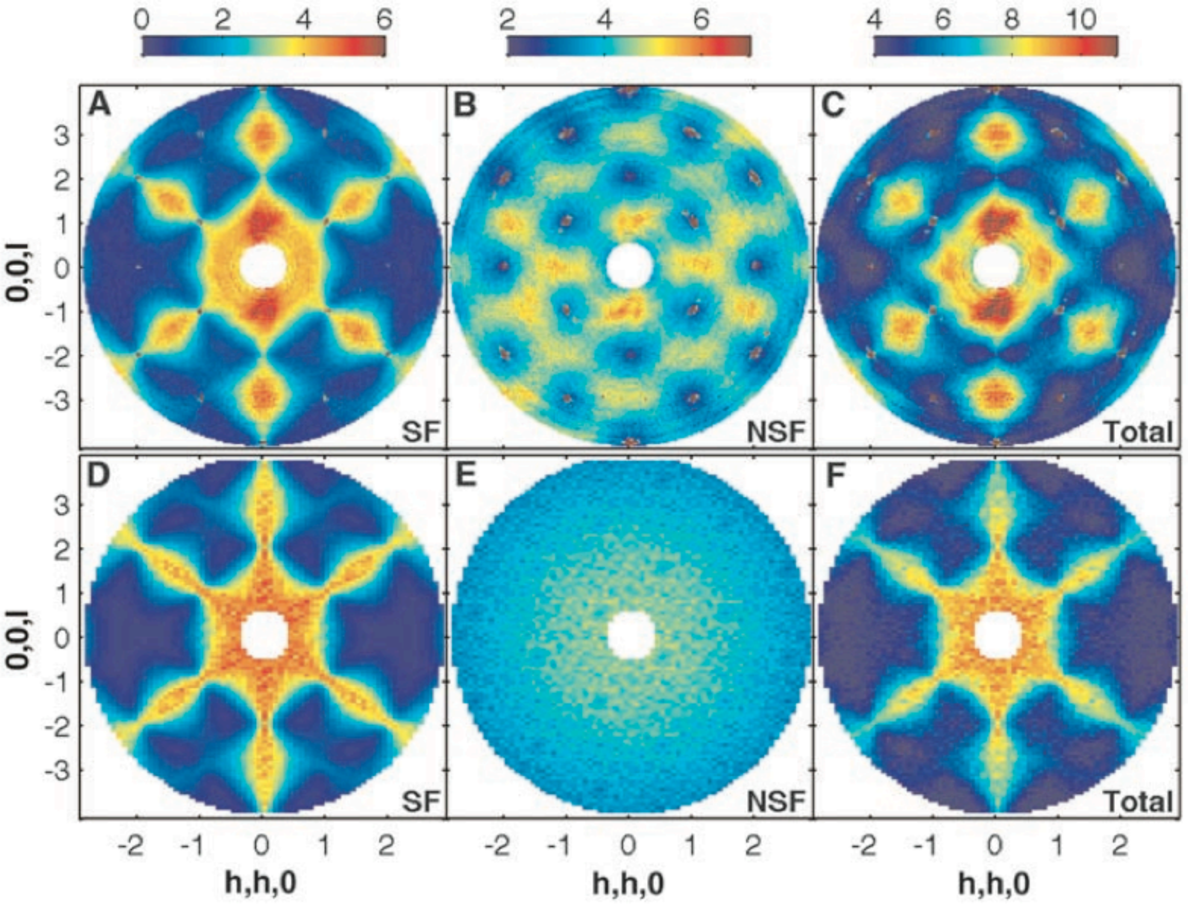
Magnetic Coulomb Phase in the Spin Ice $Ho_2Ti_2O_7$

T. Fennell[1,*], P. P. Deen[1], A. R. Wildes[1], K. Schmalzl[2], D. Prabhakaran[3], A. T. Boothroyd[3], R. J. Aldus[4],
D. F. McMorrow[4] and S. T. Bramwell[4]

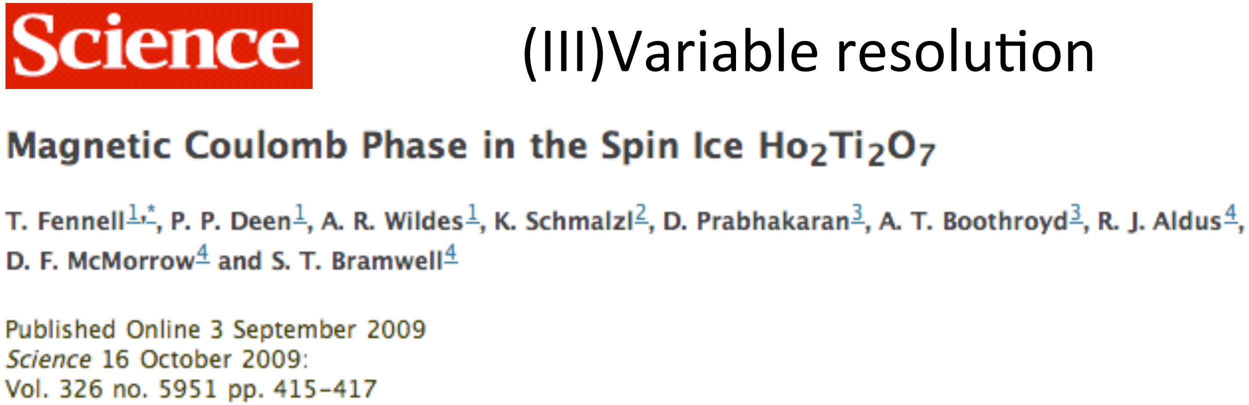
Published Online 3 September 2009
Science 16 October 2009:
Vol. 326 no. 5951 pp. 415-417

Experiment

Theory



(III)Variable resolution



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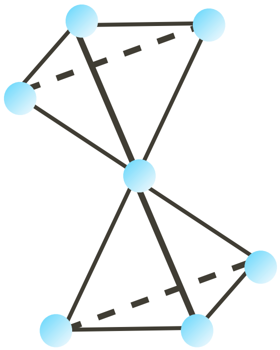
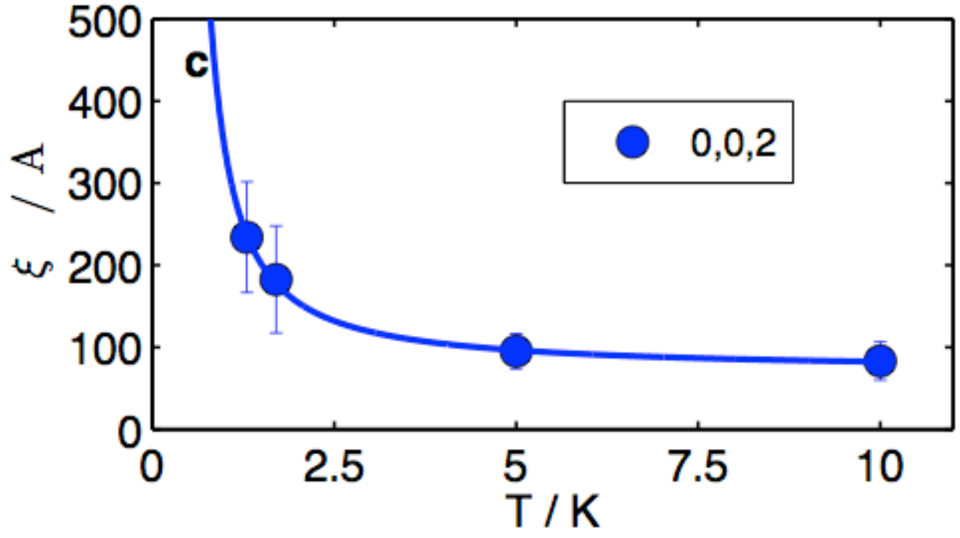


TRIPLE-AXIS IN12



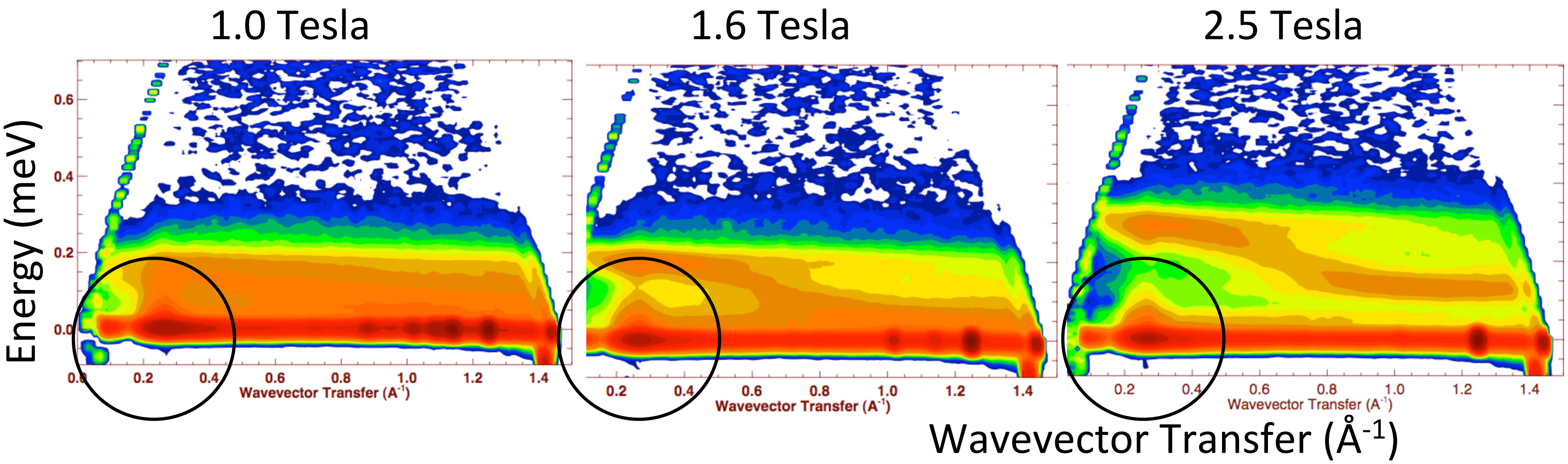
Required the resolution of a triple axis machine to measure the diverging correlation lengths but requires large S(Q, ω) for whole image.

— Describes the formation of single and double ice rules defects.
= magnetic monopoles

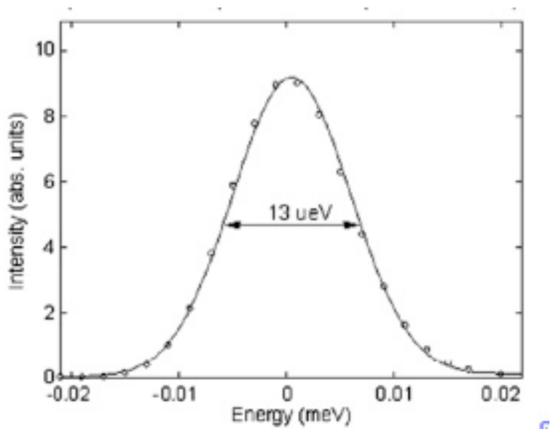
Again broad region of S(Q,ω)
but
requires variable resolution

Extras: Background & resolution : making life easy for the user

(a)Background issues: Cryogenic vacuum - limit Al windows.



(b)Resolution function



Ikeda-Carpenter versus Gaussian, Gaussian please.

- Intense neutron spectra Cold (10 microeV - 30 meV), Thermal (10 - 100 meV) (bi-spectral?)
- Tunable resolution (Gaussian..)
- Excellent PSD coverage including low angles
- Fully integrated (but optional) polarisation analysis (XYZ, CRYOPAD)
- Very low background - high S/N
- Possibility to work with advanced sample environments
- Small samples - focussing optics (Pressure)
- Sample characterisation (in-situ and out)
- Theory groups:

Nice images are being produced but we are unable to model the systems - theory groups are not a luxury but will be a necessity!

Thanks again.