Neutron scattering study on static and dynamic spin structures in quasicrystalline magnets

a seminor at UVa
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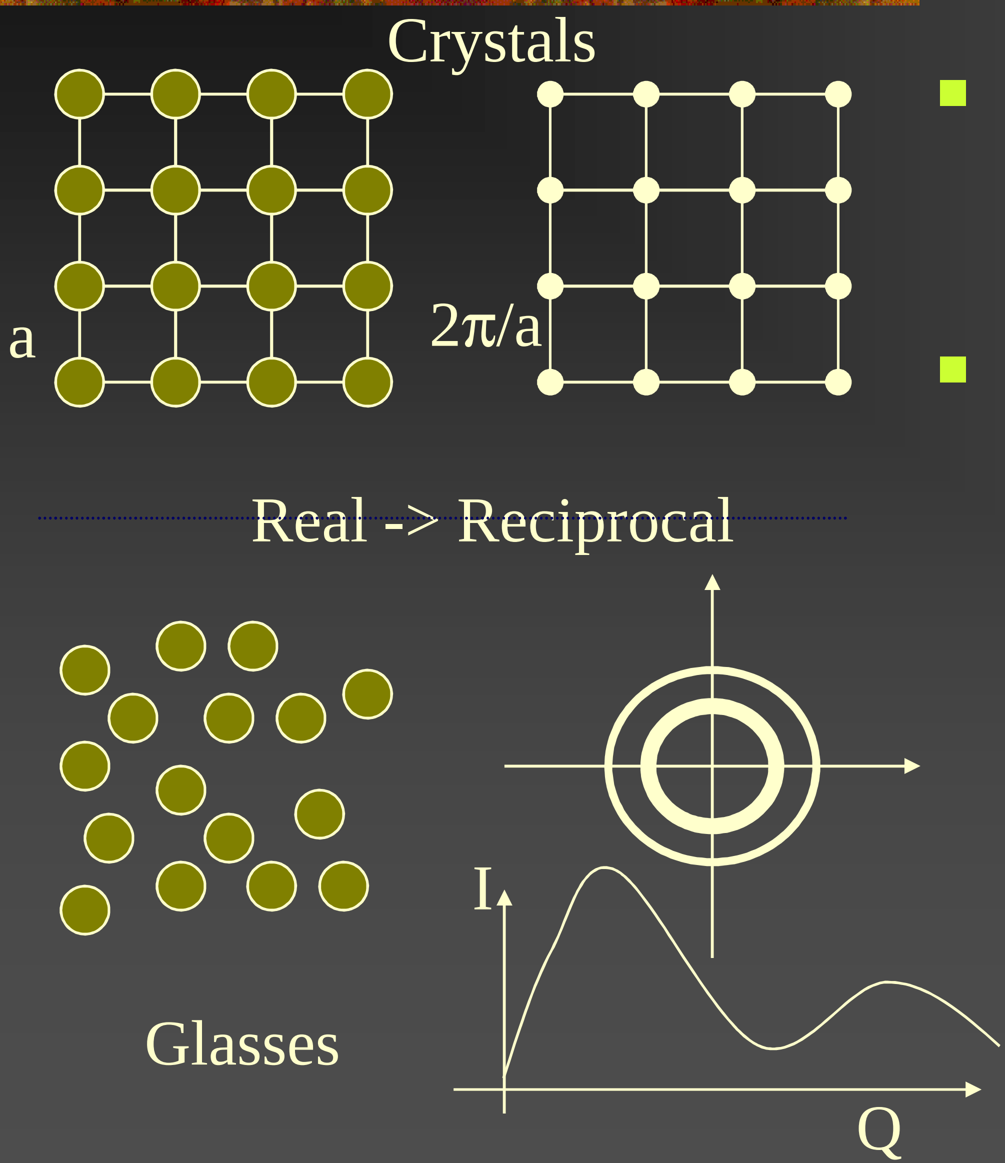
Outline

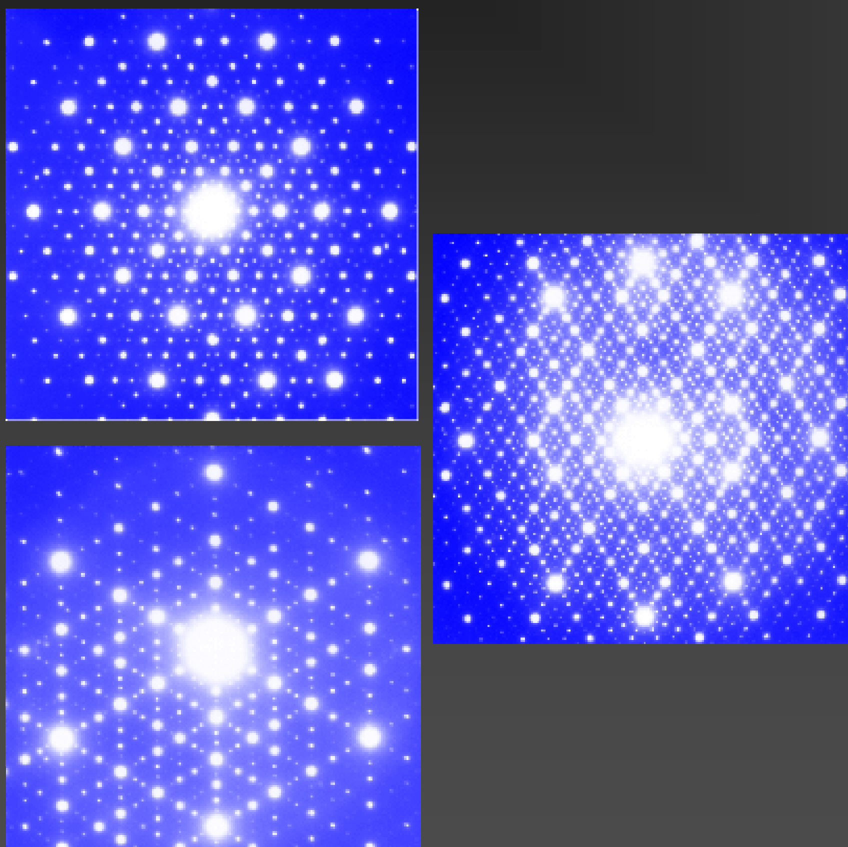
- Introduction: Magnetic quasicrystals
  - Towfold motivation of our study
    - Magnetic quasicrystals as spin systems; magnetic ordering and collective excitations
    - Magnetic quasicrystals as spins in abnormal metal; fluctuations of spins embedded in quasiperiodic electronic system
- Static spin structure study by neutron elastic scattering
  - Unique short-range spin ordering
- Spin dynamics study by neutron inelastic scattering
  - Zn-Mg-Tb: spin correlations, relaxations and dynamics in the spin freezing state with considerable short-range spin correlations
  - Zn-Mg-Ho: quasielastic (relaxational) spin fluctuations with anomalous temperature dependence; possibly related to the single-site spin fluctuations in the quasiperiodic electronic system.
- Conclusions

Crystals and glasses --long known two forms of solids--

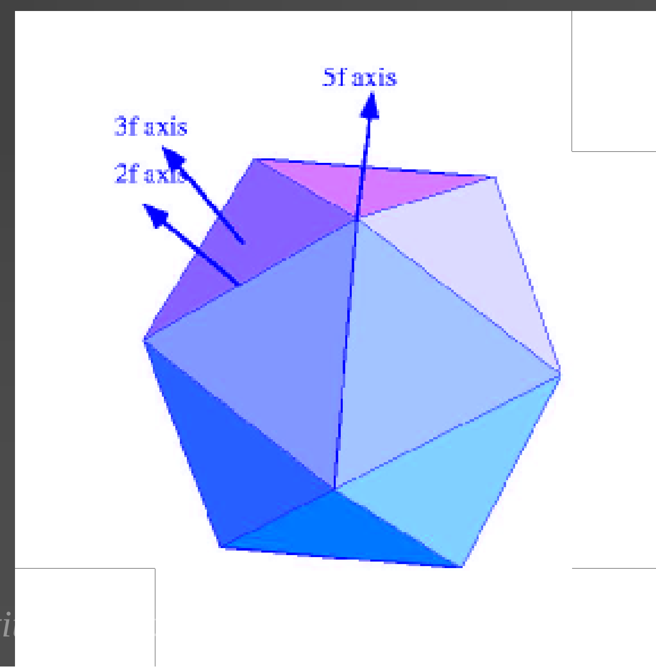


- Crystal: translation of a unit cell
  - Sharp Bragg reflections
- Glasses: random arrangement of atoms
  - Circular distribution of diffraction with broad width
  - Broad peaks in 1D diffraction pattern

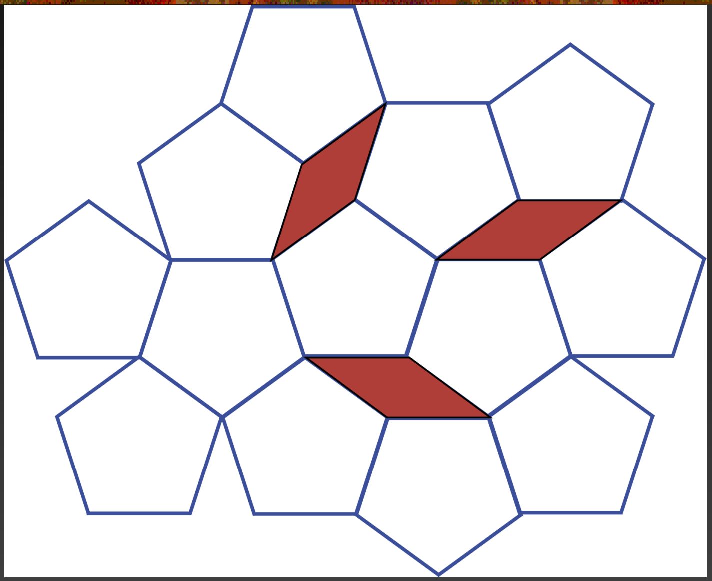
What is quasicrystal?

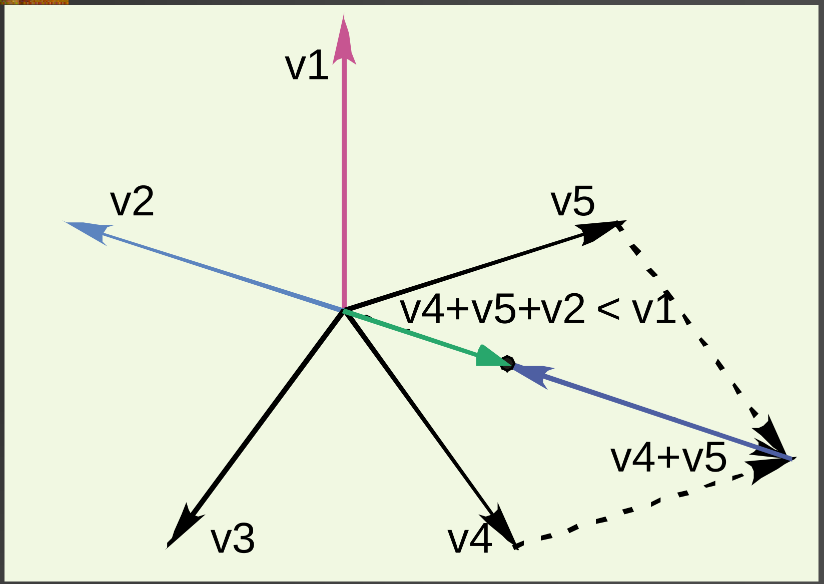


- Bragg reflections with icosahedral symmetry (Shechtman et al. PRL 53(1984) 1951)
- First found in splat-cooled Al-Mn alloy
- Thermodynamically stable QCs
- Inconsistent with spatial periodicity



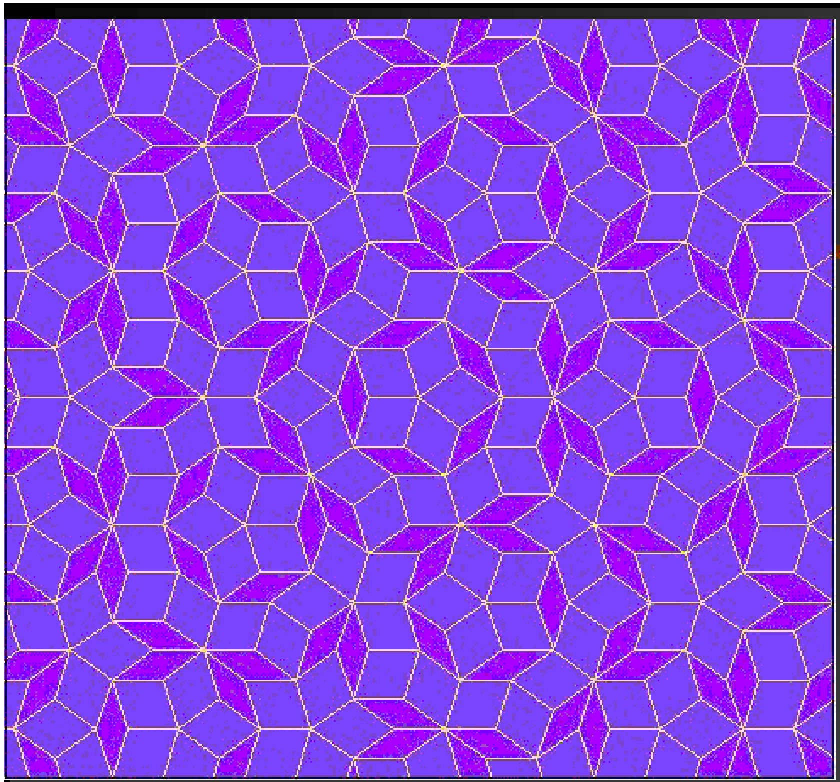
A revolution in crystallography

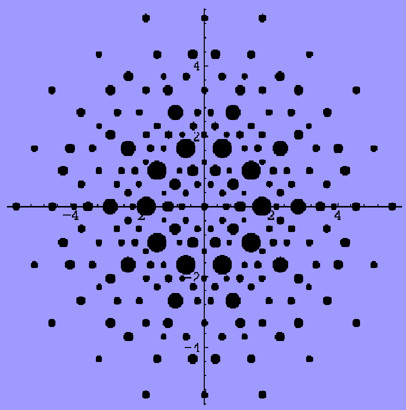




- Assume the shortest lattice vector as v1…
- 5f rotation gives rise to 5 vectors v2, …, v5
- Then for example v4+v5+v2 becomes shorter then v1!

In 1991, the International Union of Crystallography decided to redefine the term "crystal" to mean any solid having an essentially discrete diffraction diagram.



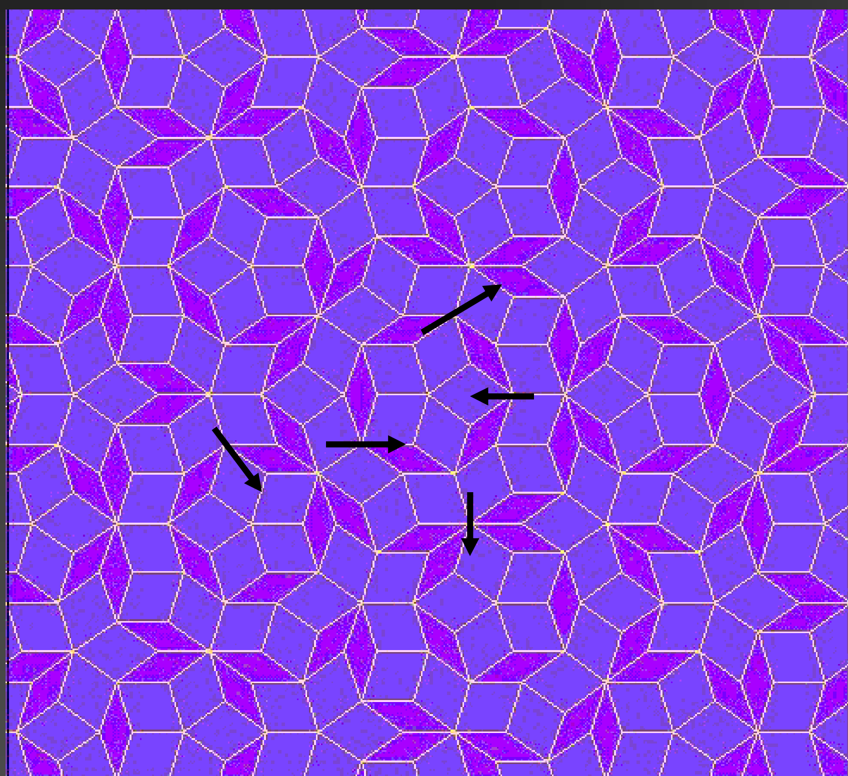


2D Penrose lattice and its diffraction pattern

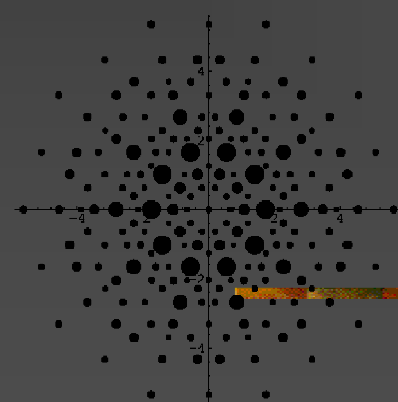
Quasicrystals

- Highly ordered structure without periodicity
  - delta-function-type Bragg reflections appearing aperiodically in reciprocal lattice space
  - 1D Fibonacci chain
  - 2D Penrose Lattice
  - 3D Icosahedral QC
- Magnetic quasicrystals: Spins occupy certain sites in the quasiperiodic lattice

Magnetic ordering in quasiperiodic lattice

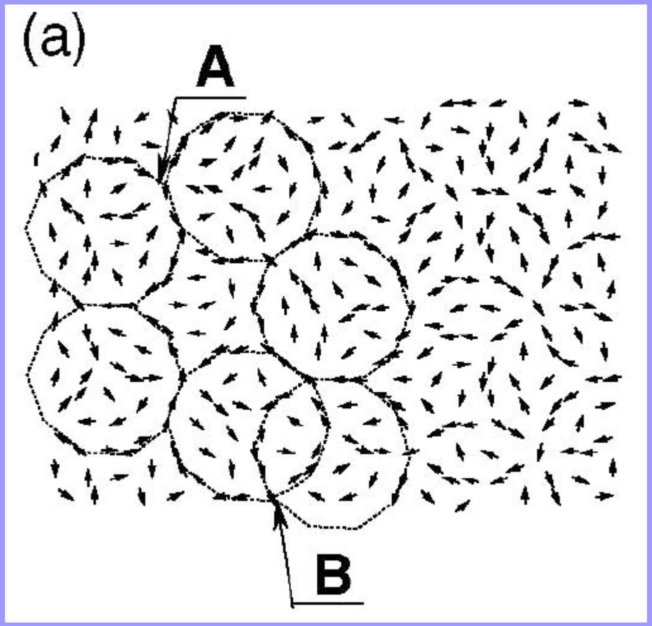


- Magnetic quasicrystal: spins in quasiperiodic structure
- Fundamental question: what ordering and excitations of spins can be seen in magnetic quasicrystal?



Theoretical background and our motivation (1): Magnetic quasicrystals as quasiperiodic spin systems

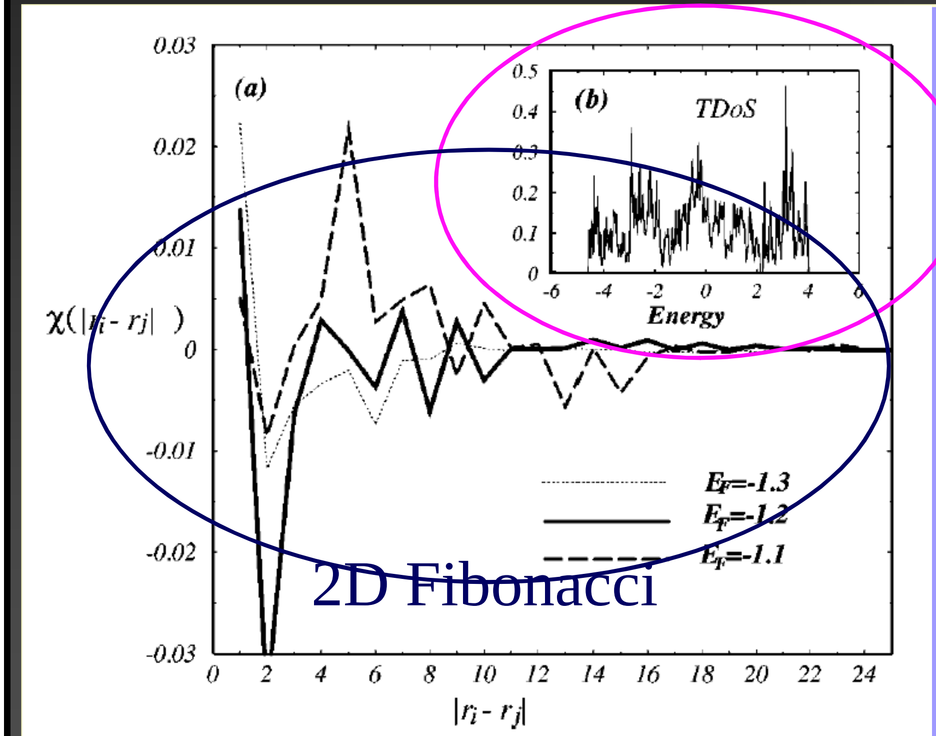
- Quasiperiodic spin systems; a fundamental issue since the discovery of quasicrystals
  - Achiam et al. PRB 33 (1986) 6460
    - 1D Fibonacci chain with Ising spins
  - Tsunetsugu et al. PRB 36 (1987) 5493
  - …
  - Vedmedenko et al. PRL 90 (2003) 137203
    - 2D Penrose lattice with Heisenberg spins and dipolar interactions
  - Wessel et al. PRL 90 (2003) 177205
    - Quantum Heisenberg spins on octagonal lattice
  - Vedmedenko et al. PRL 93(2004) 076407
    - 2D octagonal tiling with Ising, xy, Heisenberg spins
  - Not applicable to be compared with experiments
    - No 3D calculations
    - Spin dynamics calculations

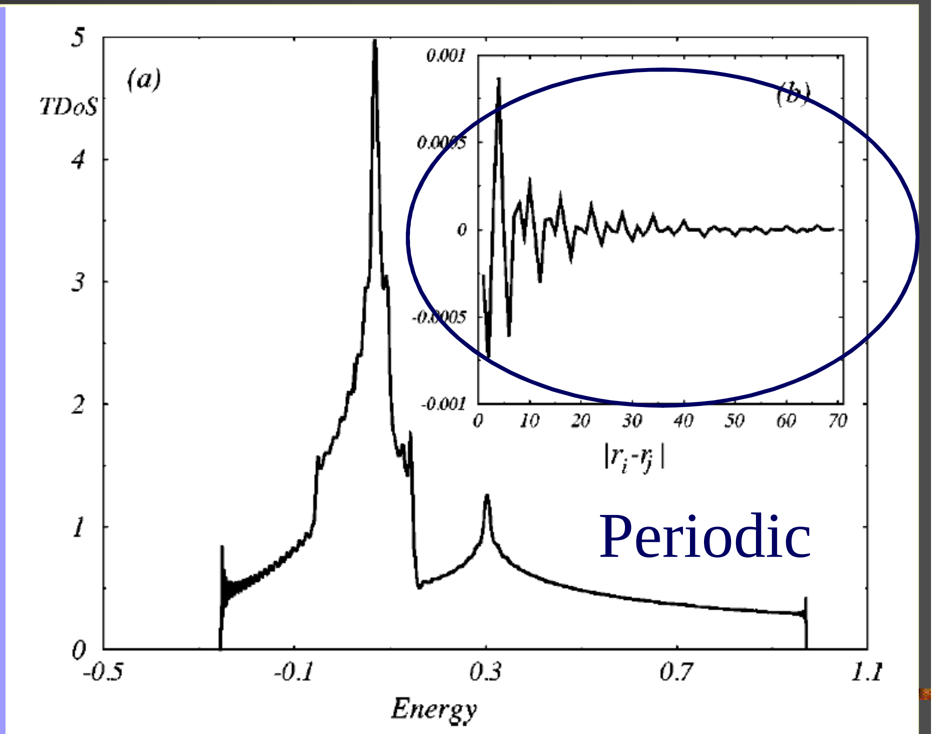


Example of non-trivial spin ordering; Spins in decagon chains with disorder inside (Vedmedenko et al.)

Theoretical background and our motivation (2):
Magnetic quasicrystals as spins in quasiperiodic electronic systems

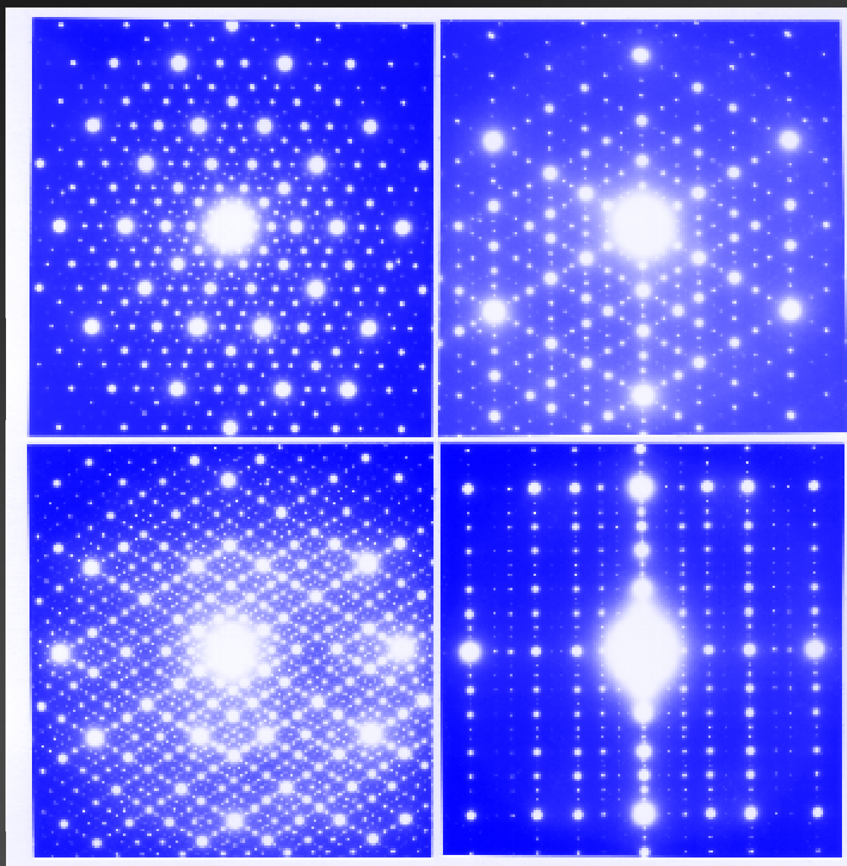
- Electronic state in quasipeiodic lattice is supposed to be "***critical***" at least in 1D (and possibly 2D, 3D)
- How magnetic moments are scattered by such anomalous *conduction* electrons?





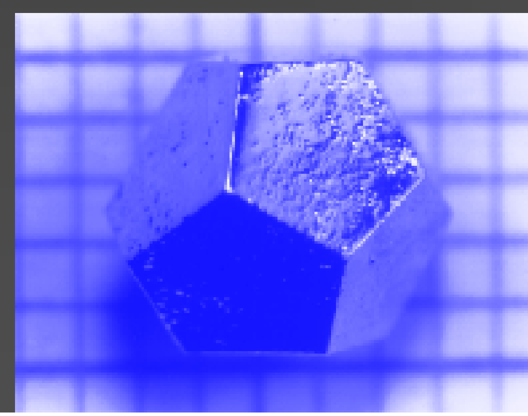
Electronic DOS and RKKY-type intersite interactions in 2D Fibonacci lattice and periodic lattice Roche et al. PRB60(1999)322

Rare-earth based quasicrystals



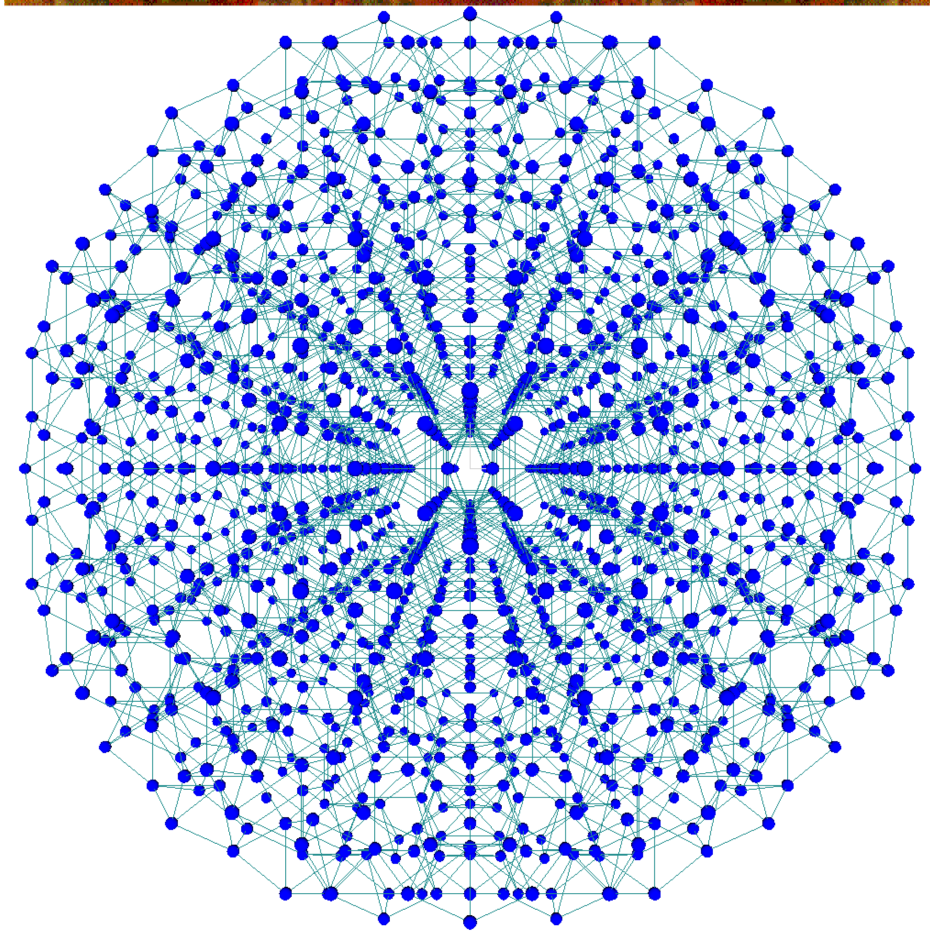
Tsai et al. Private communication

- Magnetic quasicrystals:
  - f-Zn-Mg-RE (Luo, Tsai 1994)
  - p-Cd-Mg-RE (Guo 2000)
  - p-Zn-Mg-RE (Sterzel 2000)
  - p-Zn-Fe-Sc (Kashimoto 2004)
- Icosahedral symmetry (3D QC)
- Thermodynamically stable (single quasicrystal growth)
- Present report RE=Tb and Ho



Fisher et al PRB 59(1999)308

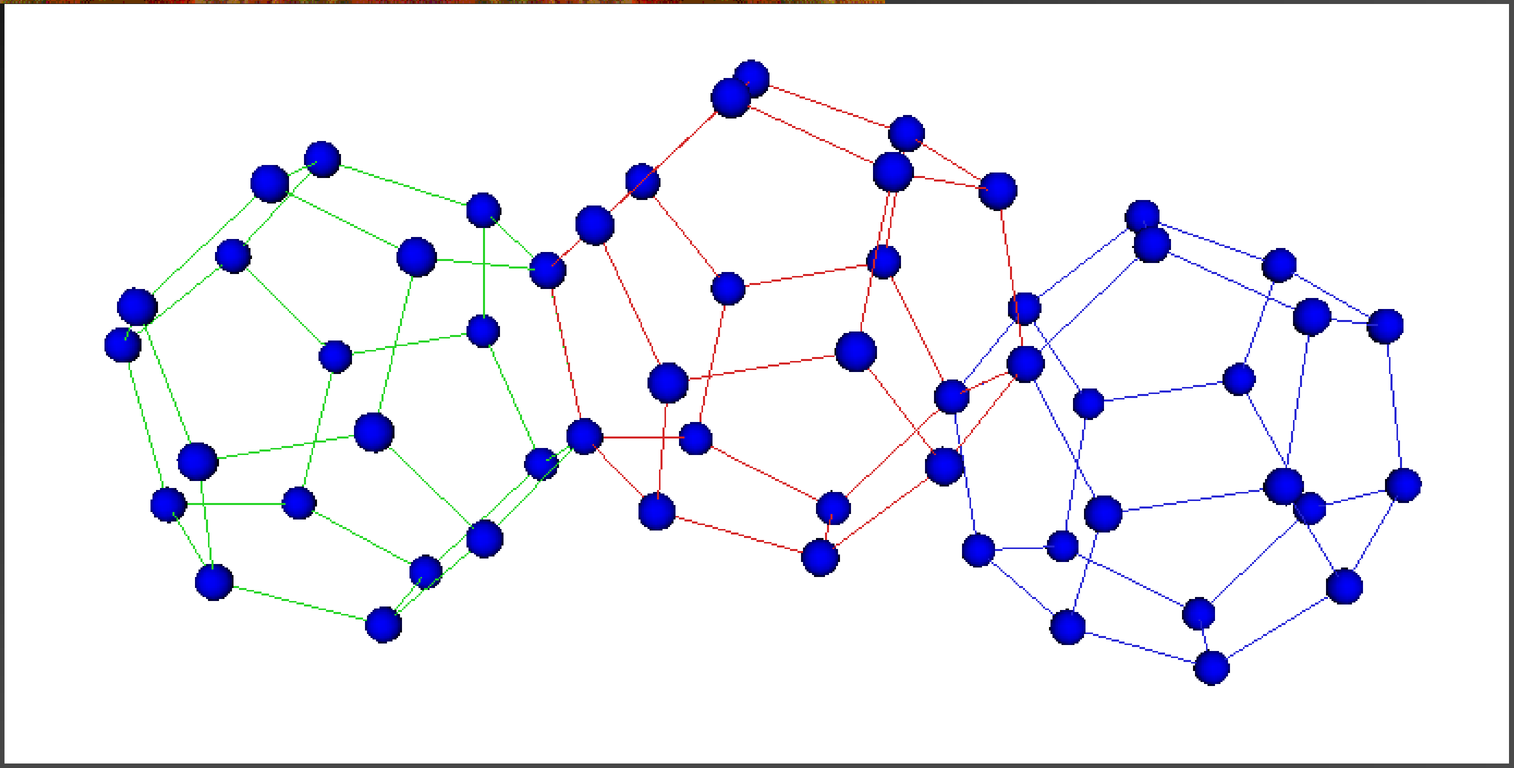
Structure of f-Zn-Mg-RE quasicrystals



RE sites in $R < 40$Å around the origin

- **Low density elimination method** applied to higher dimensional crystallography
  - (Takakura et al. PRL 86 (2001) 236)
- **Synchrotron x-ray powder diffraction** with spherical 6D occupation domains
  - (Ishimasa et al. J. Non-cryst. Solids 334&335 (2004) 167)
- RE site in 6D lattice:
  - (3/4,1/4,1/4,1/4,1/4,1/4)
  - r = 7.419Å

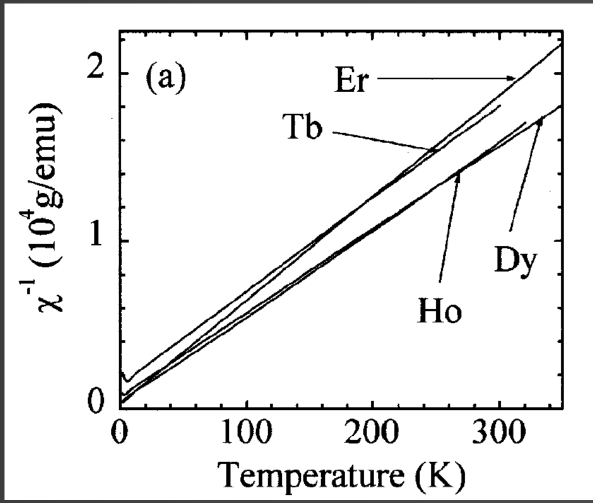
Structure of f-Zn-Mg-RE quasicrystals (II)



- Approximate network of RE sites: Highly frustrated for AF interactions
  - Edge sharing dodecahedrons (edge length = 5.5Å)
  - Extra atoms (66% participating to dodecahedron network)
    - Partly due to the spherical occupation domains

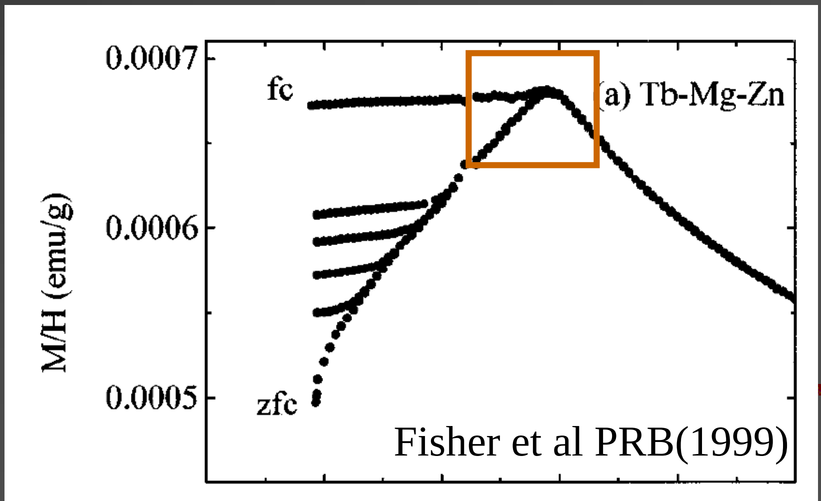
Bulk magnetic properties of f-Zn-Mg-RE

- RE: stable localized moment (Tb ~ 10μ_B)
- Macroscopically spin-glass-like freezing is seen (Tf ~ 5.8K for Zn-Mg-Tb)
- What we can learn from neutron scattering data?



Summary of magetic parameters obtained for the f-Zn-Mg-R (Fisher *et al.*, 1999) and p-Cd-Mg-R (Sato *et al.*, 2001*a*) quasicrystals. $\mu_{\rm eff}$, $\mu_{R^{3+}}$, Θ, $T_{\rm f1}$ and $T_{\rm f2}$ are the effective moment, calculated moment for free R^{3+} ions, Weiss temperature, upper freezing temperature, and lower freezing temperature (if exists).

System	$\mu_{\rm eff}(\mu_B)$	$\mu_{R^{3+}}(\mu_B)$	Θ (K)	$T_{\rm f1}$ (K)	$T_{\rm f2}$ (K)
Zn-Mg-Tb	9.91	9.72	-26.3	5.80	-
Zn-Mg-Dy	10.5	10.63	-14.8	3.60	-
Zn-Mg-Ho	10.4	10.60	-7.8	1.95	-
Zn-Mg-Er	9.49	9.59	-5.1	1.30	-
Cd-Mg-Gd	7.90	7.94	-37	13.0	4.8
Cd-Mg-Tb	10.03	9.72	-23	12.5	5.6
Cd-Mg-Dy	10.67	10.63	-14	7.4	3.8
Cd-Mg-Ho	10.42	10.60	-7	12.5	5.0
Cd-Mg-Er	9.71	9.59	-6	4.4	-
Cd-Mg-Tm	7.08	7.57	-2	-	-



Fisher et al PRB(1999)

Experimental technique; neutron scattering

- We have utilized various neutron scattering spectrometers specialized for various energy ranges
  - Inverted geometry TOF spectrometers (dE~0.2meV)
  - Triple-axis spectrometers (dE ~ 1meV)
  - Exact backscattering spectrometers (dE~1ueV)

Elastic experiment using single quasicrystals; f-ZnMgHo

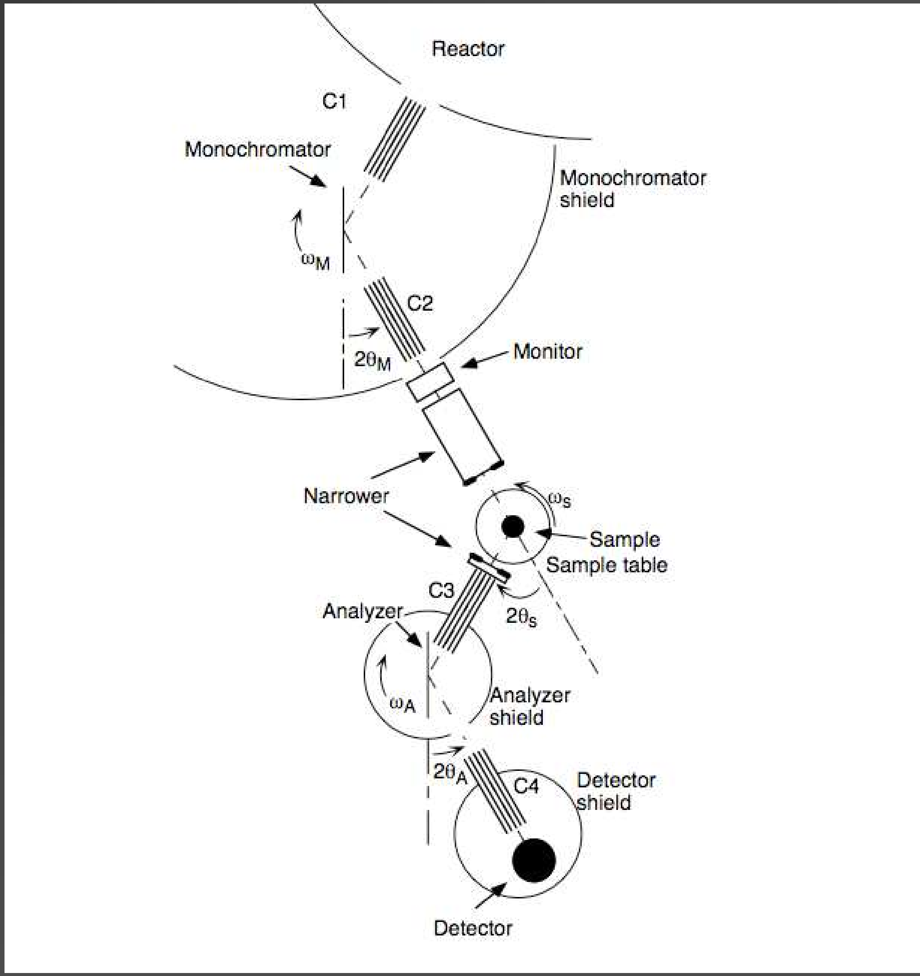
- Q: Is there any special spin ordering related to the quasicrystalline structure?

Triple-axis spectrometer

- Standard technique to measure elastic as well asa inelastic scattering with the continuous neutron source, such as the reactor.

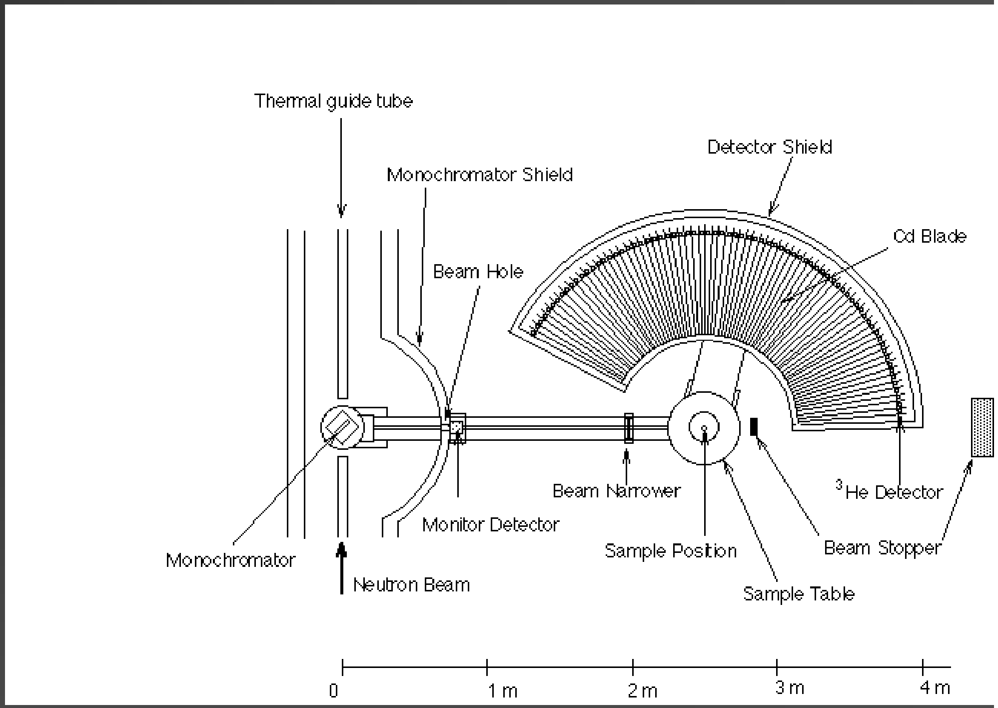
QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

4G: GPTAS at JRR-3



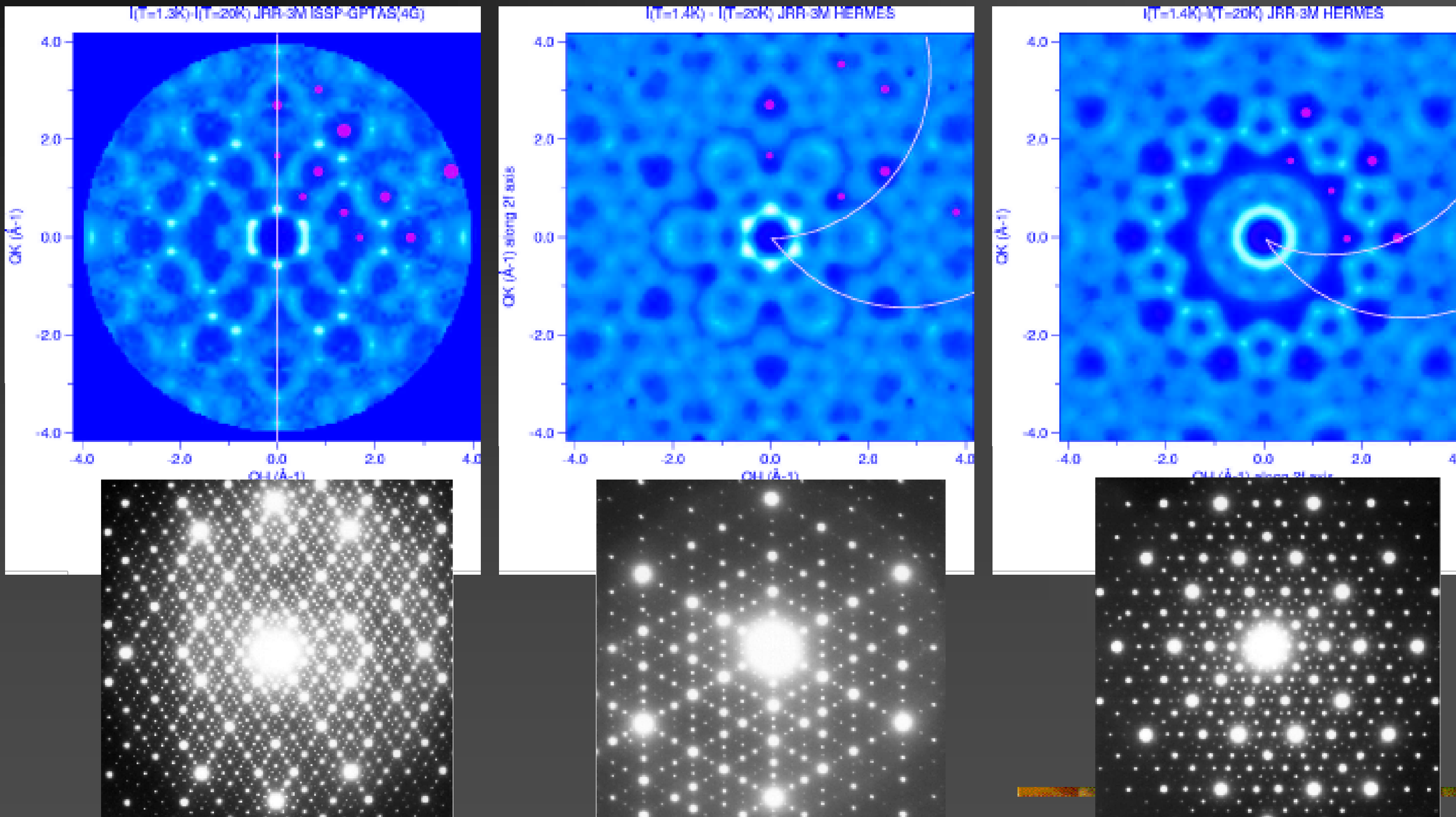
Powder diffractometer

- Specialized for elastic scattering
- Multi-detectors for efficient measurements
- We use powder diffractometer for single crystal S(Q) mapping.



HERMES at JRR-3

Single quasicrystal Zn-Mg-Ho



- Significant short-range spin correlations
- Antiferromagnetic (Satellite from nuclear Bragg reflections)
- Icosahedral symmetry in magnetic diffuse scattering

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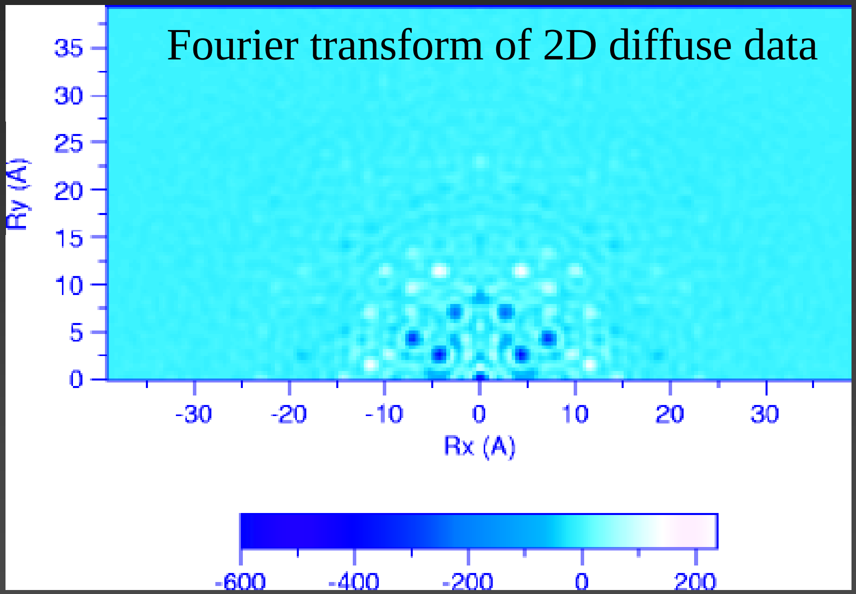
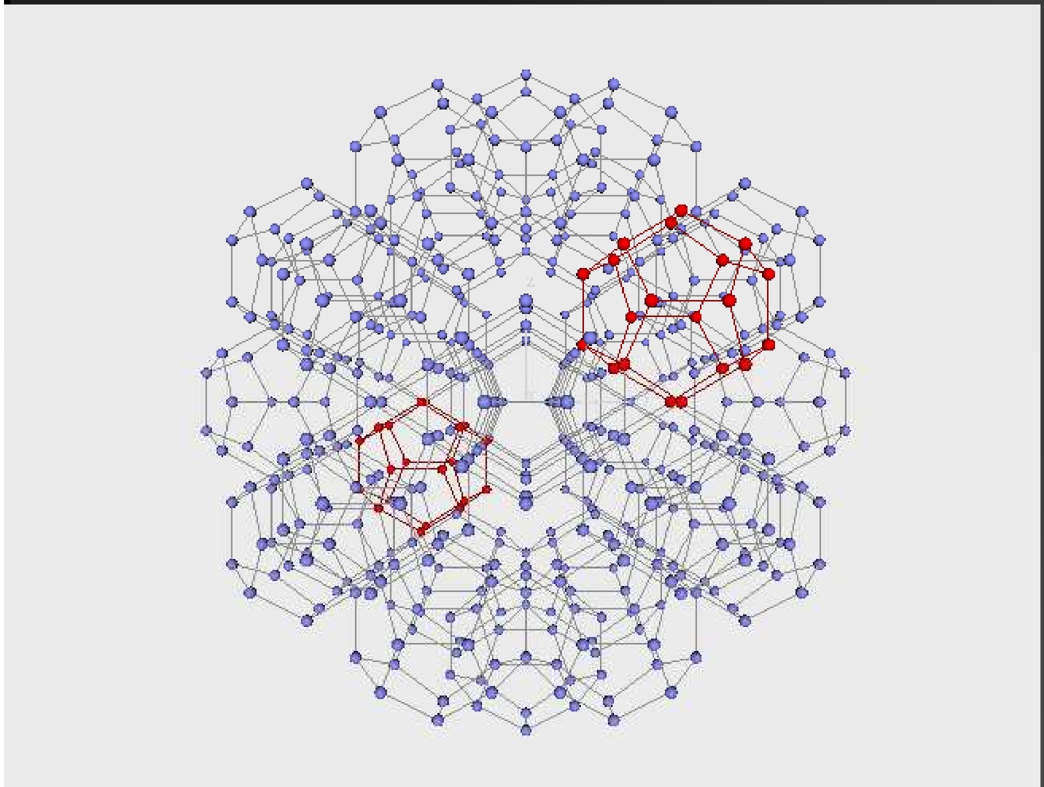
Fourier transform of diffuse scattering



- 2f-plane projected spin Patterson map
- ξ ~ 10Å (HWHM)
- Antiferromagnetic

Looking for a cluster



- RE structure of Zn-Mg-Ho QC
  - Spherical model
  - RE: (3/4,1/4,1/4,1/4,1/4,1/4)
  - R < 7.419Å
- Dodecahedral network

Dodecahedron
(Cluster atom/all atom) ~ 66%
Clusters are edge-shared
KEY CLUSTER!

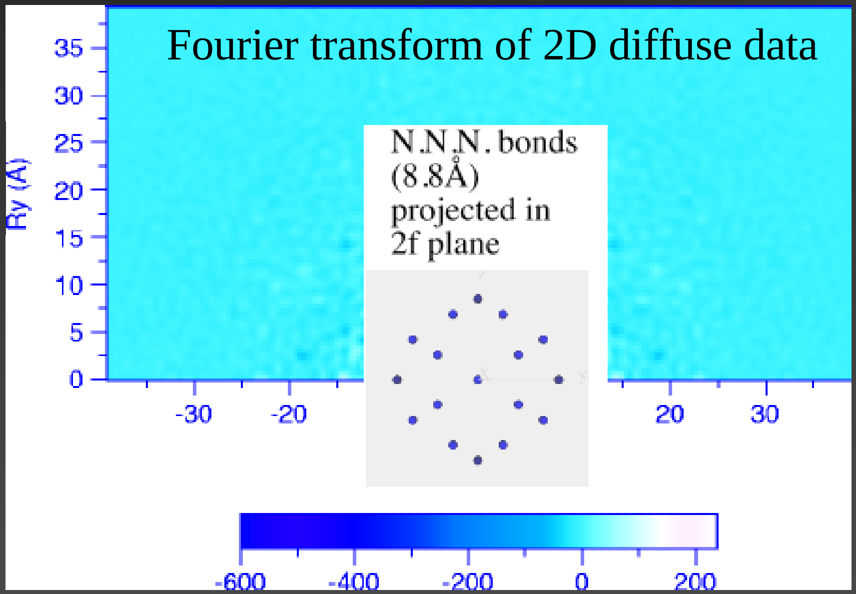
Understanding the diffuse scattering

- Static short-range order and strongly localized but possibly collective inelastic mode
- Glassy behavior in macroscopic properties

- Suggestive of randomly frozen spin clusters
  - Size: 10Å
  - Antiferromagnetic

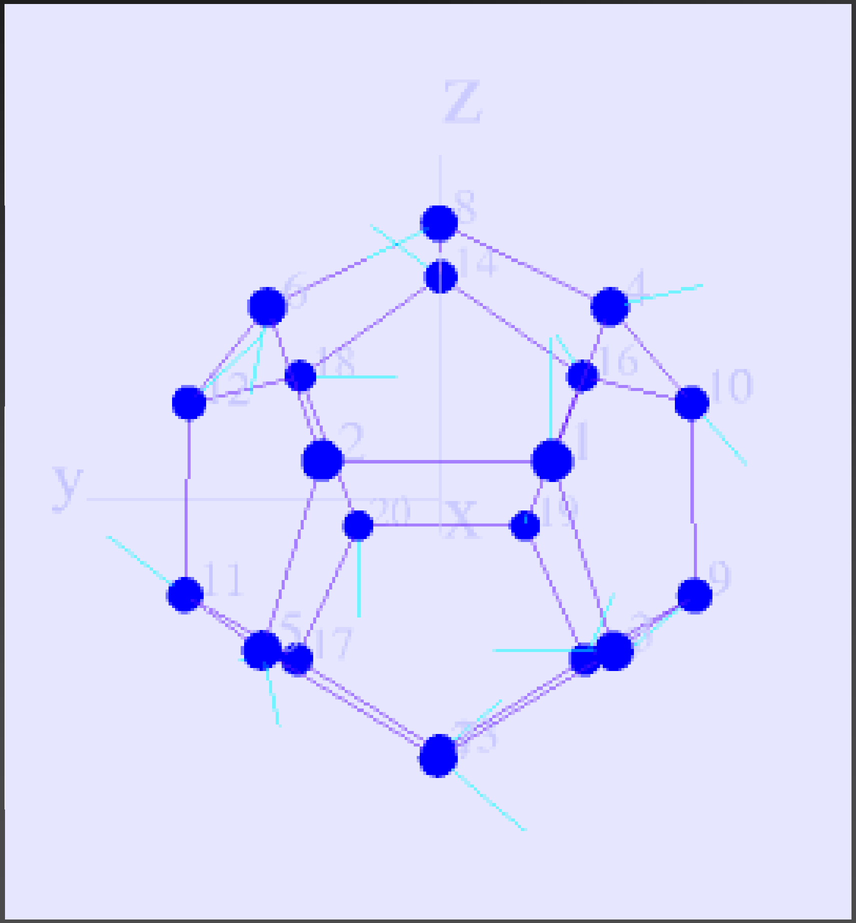
- We try to find a possible MODEL SPIN CLUSTER in the Zn-Mg-RE structure model, which can explain the observed diffuse scattering patterns

Interactions suggested



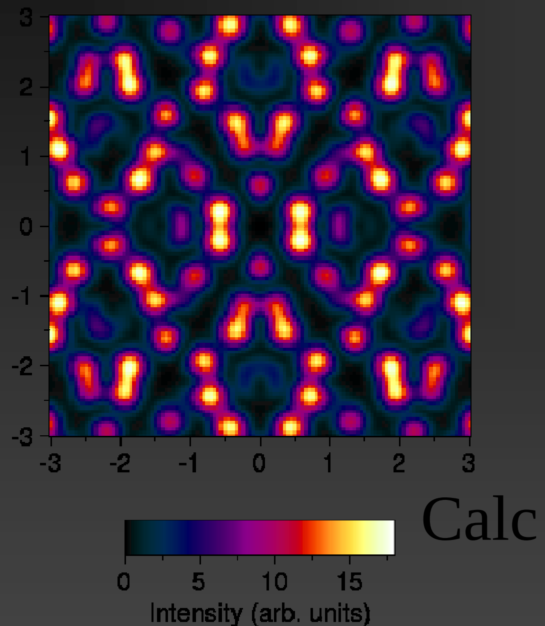
- 2f-plane projected spin Patterson map
- Consistent with the dodecahedral spin cluster if
  - N.N. bonds are ferromagnetic
  - N.N.N bonds are antiferromagnetic and substantially strong.

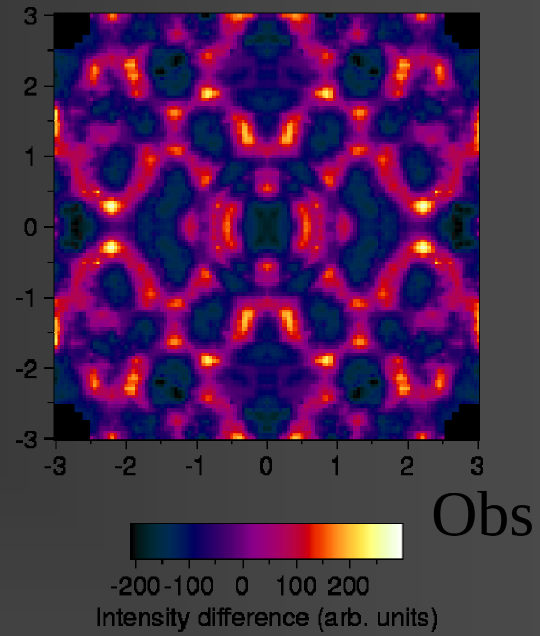
Dodecahedral spin cluster model



- Dodecahedron
- N.N.N antiferromagnetic interactions
- Heisenberg (isotropic) spins
- $H = \Sigma_{ij}\ S_i.S_j$
- Ground state spin configuration is determined by numerical minimization (Axenovich and Luban PRB 63 (2001) 100407)

Verification of the dodecahedral model





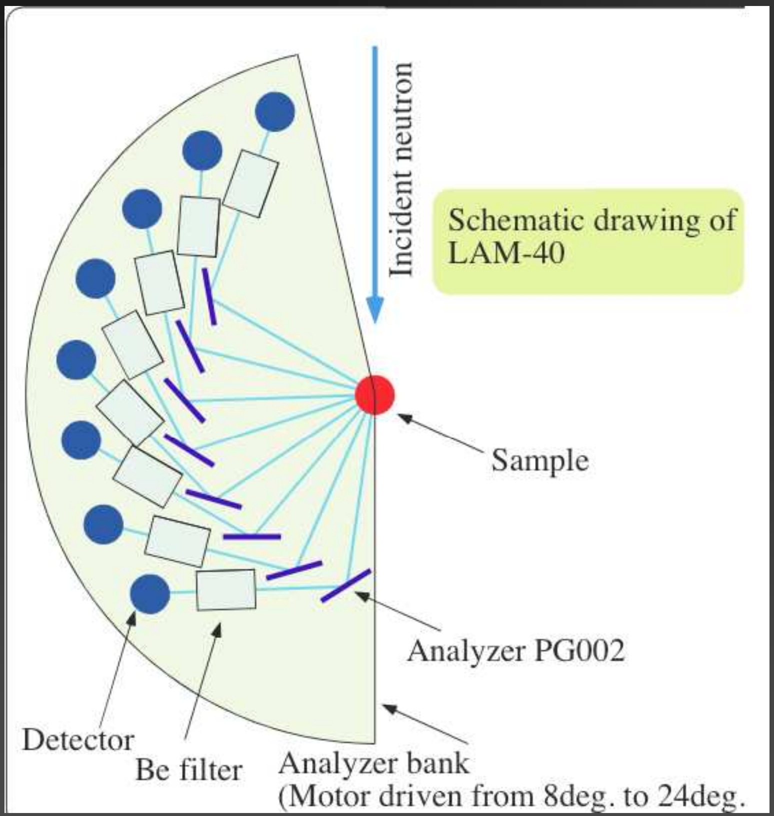
- Elastic diffuse scattering is well reproduced by the very simple single dodecahedral cluster model

- Proper statistical average is taken.

How about spin excitations?

- Q: Does the dodecahedral spin cluster model explain the spin excitation spectrum???

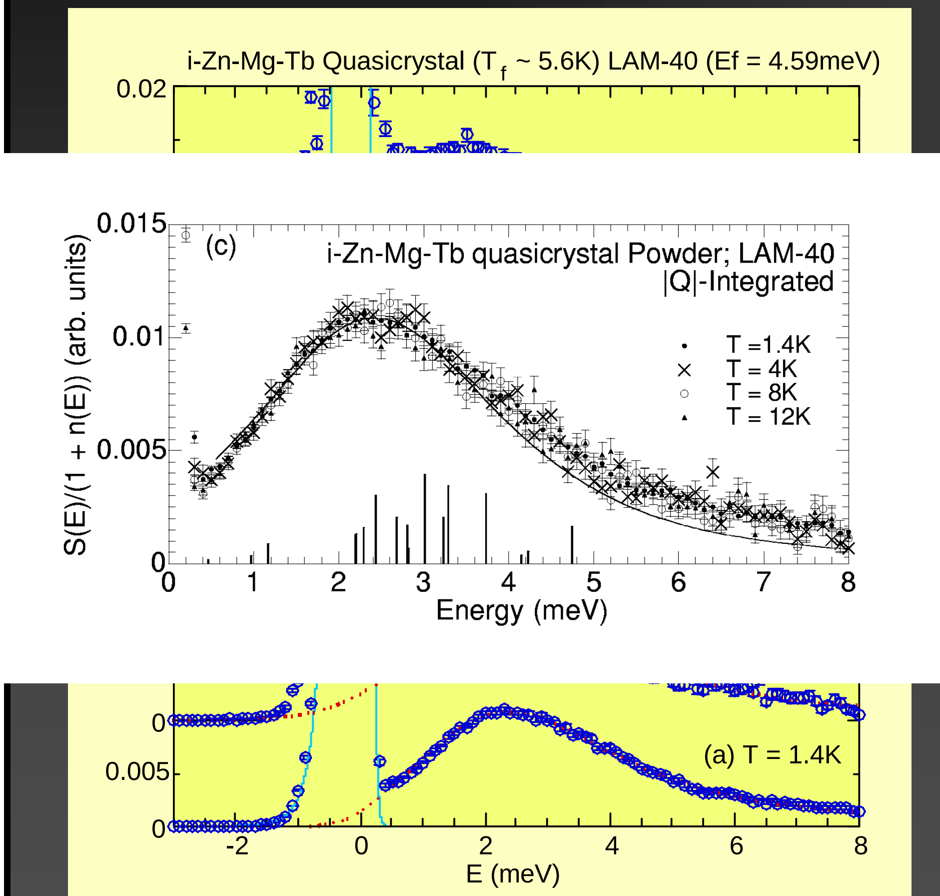
Conventional inverted geometry TOF specctrometer



LAM-40 at KENS

- Pulsed white neutrons incident on sample
- Outgoing neutrons are energy-analyzed by the crystal analyzer
- Energy resolution will be roughly detemined by the pulse width (time uncertainty) and analyzer energy width (energy uncertainty)
- For Ef~1.49meV (PG002 with θ = 40deg), dE ~ 200μeV
- Energy resolution may be increased using better resolution analyzer (i.e. larger q) and short pulse width or longer flight path

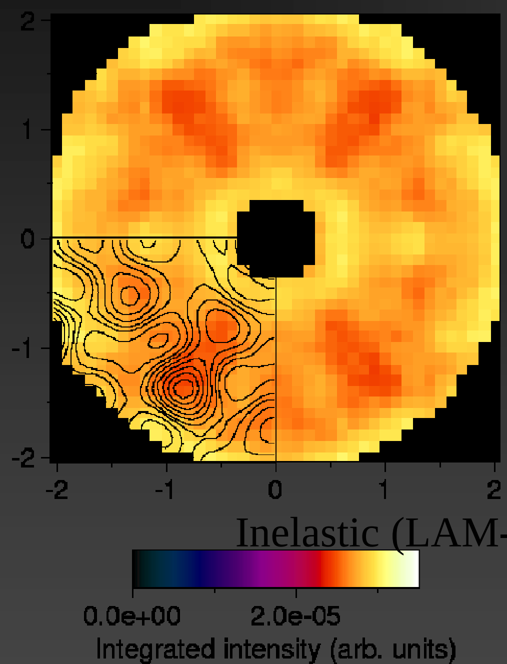
f-ZnMgTb quasicrystal
Inelastic excitation spectrum



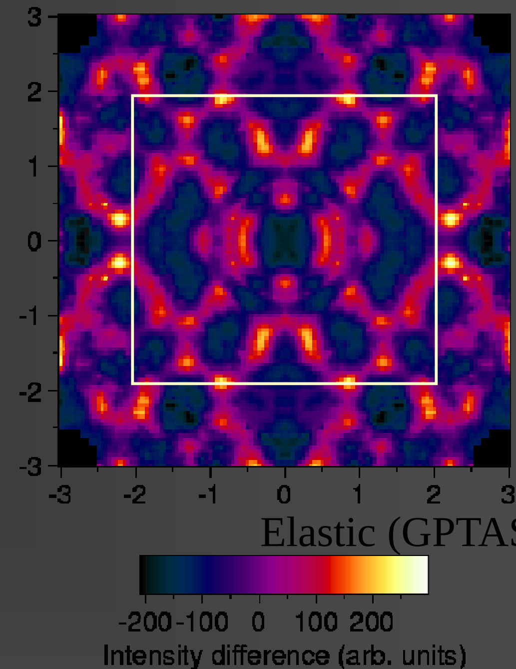
- Broad Inelastic peak at 2.5meV
- Scaled by Bose temperature factor, $(1 - \exp(-\hbar\omega/k_B T))^{-1}$ below T<12K
- Note:

  $S(hw) \sim (1-\exp(-\hbar\omega/k_B T))^{-1} \, \mathrm{Im}\chi(\hbar\omega)$
- $\mathrm{Im}\chi(\hbar\omega)$ does not change significantly
- for $\hbar\omega$ > 4meV, $S(\hbar\omega)$ is almost T-independent (even at higher T>20K)
- Not a crystalline electric field splitting level transition (CEF: Boltzmann)

f-ZnMgTb quasicrystal
Q-dependence by single-QC exp



Inelastic (LAM-40)

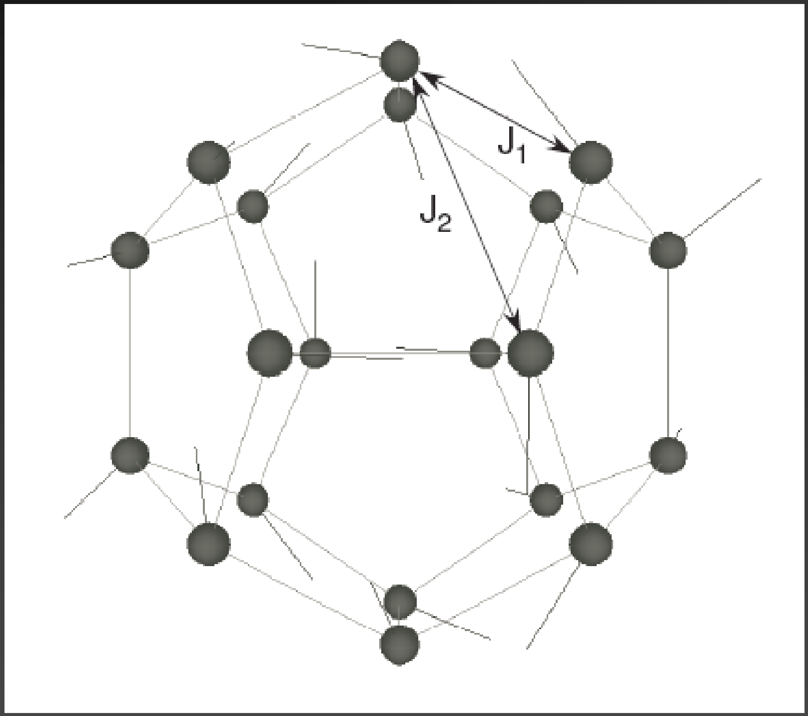


Elastic (GPTAS)

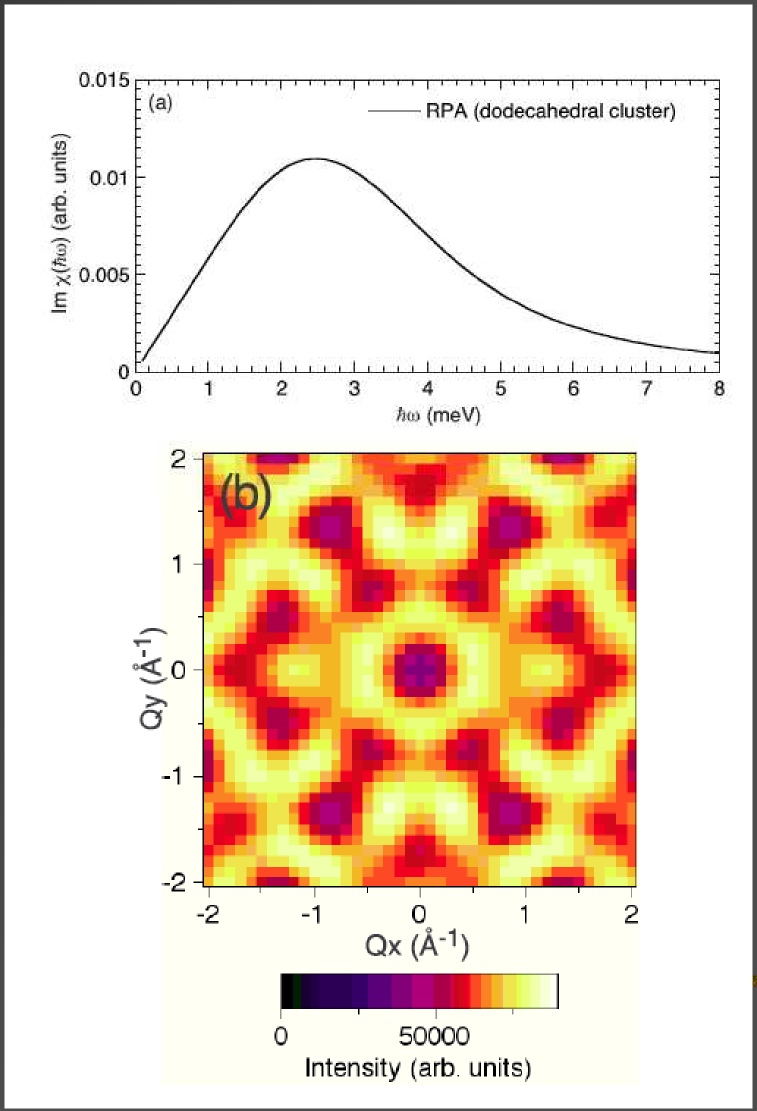
Zn-Mg-Tb

- Inelastic scattering intensity (1 < E < 4meV)
  - Intensity is similar to elastic diffuse scattering (magnetic structure factor)
- Spin-wave-type modes localized in the short-range ordered region.

RPA calculation for dodecahedral spin cluster model



$$\mathcal{H} = -\sum_{<\mathrm{n.n.n.}>} J_{ij} \hat{\boldsymbol{J}}_i \cdot \hat{\boldsymbol{J}}_j,$$



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Where is the “Freezing”?

- Q: We understand that the dodecahedral spin cluster explains the elastic and inelastic spectrum. But in which energy range the macroscopically observed “FREEZING” can be observed?

Neutron scattering technique
Exact backscattering spectrometer

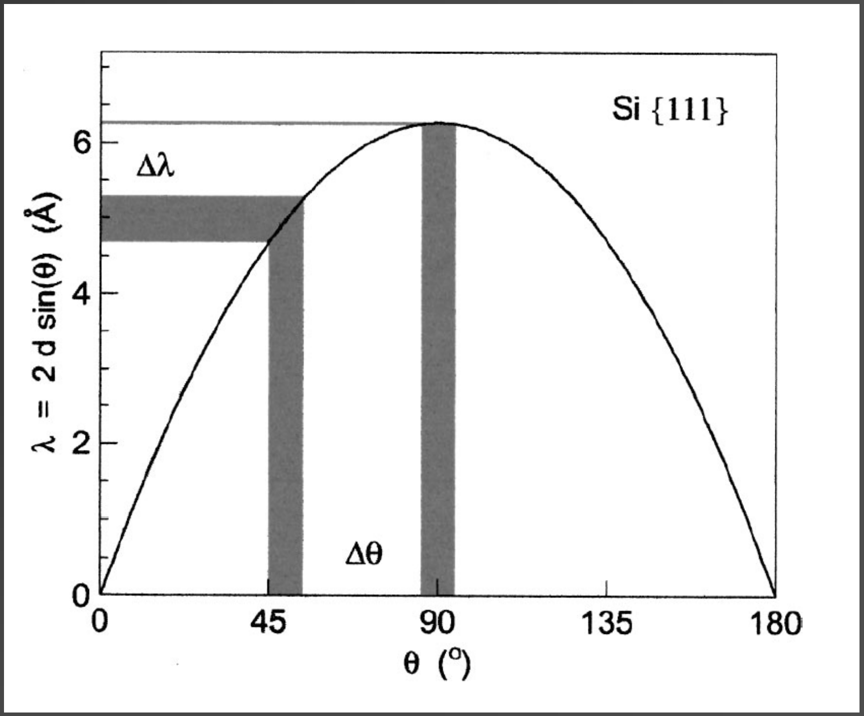
- Exact backscattering provides **1μeV resolution!**
  - -> ~250Mhz
- The principle:
  - Bragg law

$$\lambda = 2d\sin(\theta).$$

  - Differentiate it!

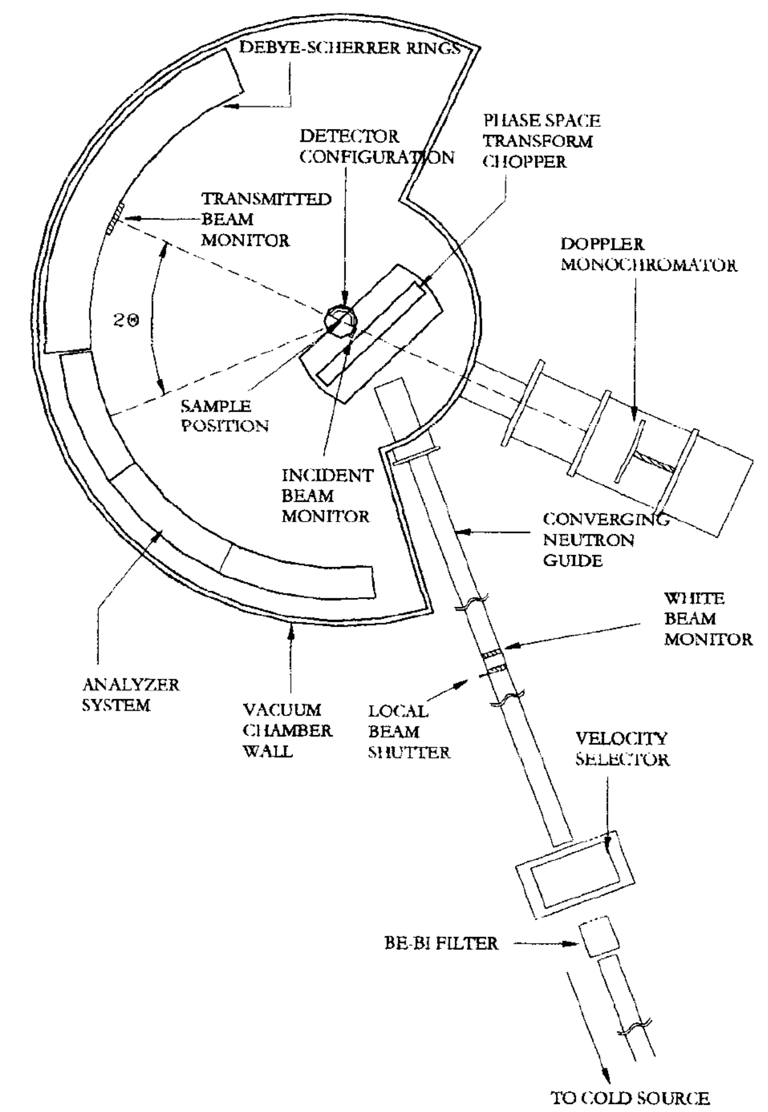
$$\mathrm{d}\lambda = 2d\cos(\theta)\mathrm{d}\theta$$

  - In practice we have finite d*d* due to dynamical scattering even for perfect crystal.



Neutron scattering technique
Exact backscattering spectrometer

- NIST HFBS spectrometer
- Monochromator and analyzer: Si(111), unpolished
- Ei = 2.08meV (~630m/s)
- dE = 0.8 to 0.9μeV
- Doppler driven monochromator: -30μeV < E < 30μeV
- 16 detectors spanning $0.25<Q< 1.75Å^{-1}$



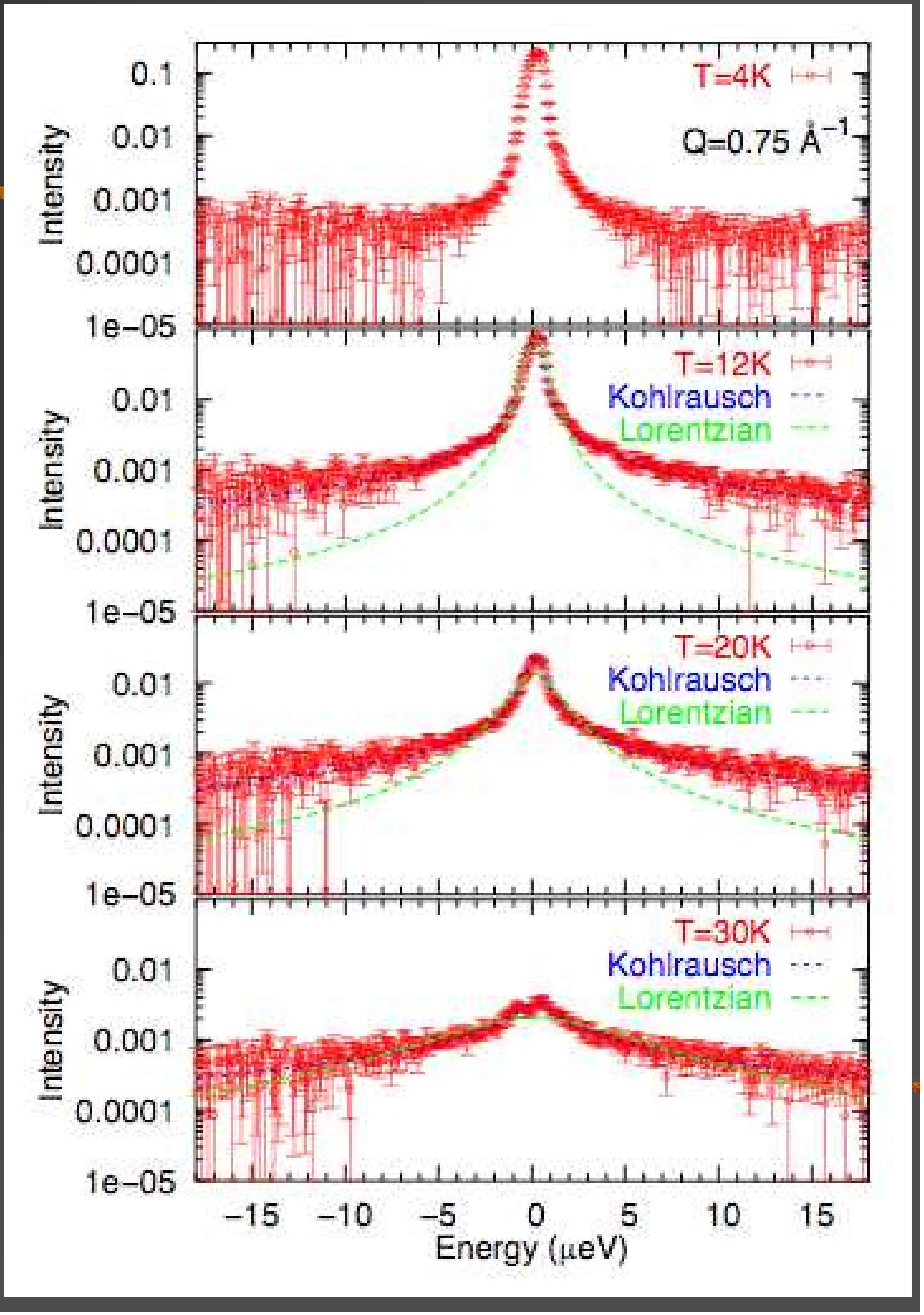
Inelastic excitation spectra

- T < Tf (~5.6K)
  - Resolution limited
  - Very small tail???
- T>Tf
  - No Lorentzian!
  - Time Fourier transform of Kohlrausch or stretched exponential function.

$$\phi(t) = \exp\left[-(t/\tau)^{\beta}\right],$$

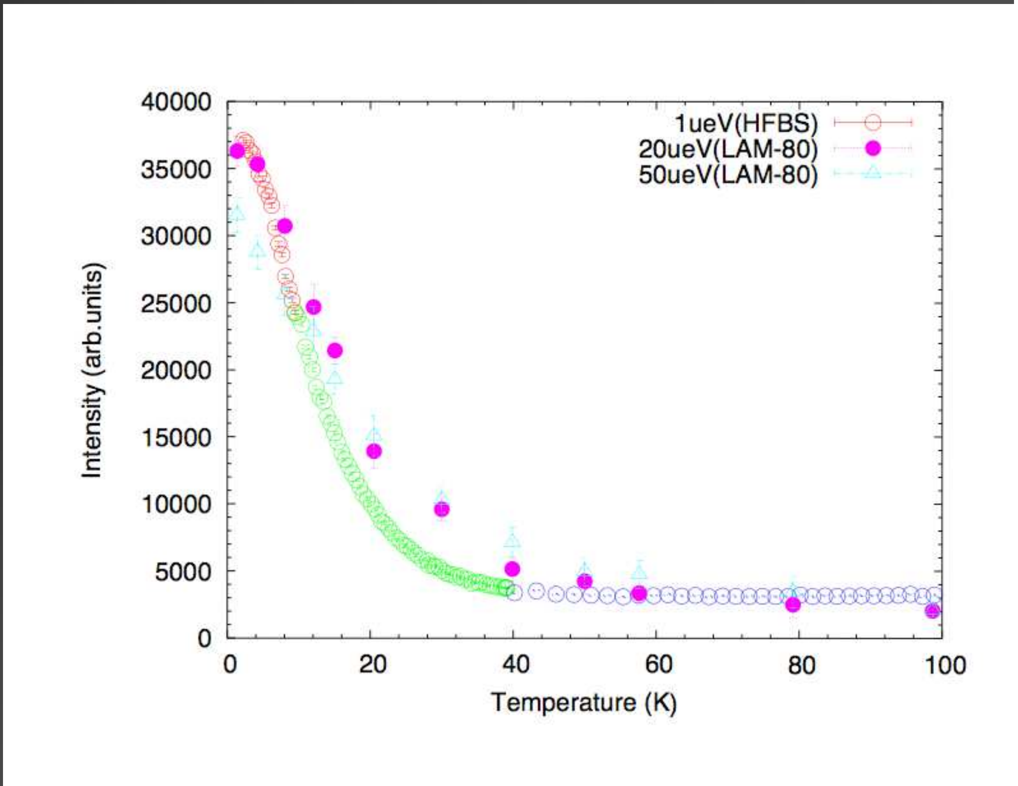
  - This indicates a certain distribution of relaxation time

$$\phi(t) = \int_0^{\infty} f(\Gamma) e^{-\Gamma t} d\Gamma$$



f-ZnMgTb quasicrystal
Spin freezing observed by neutrons

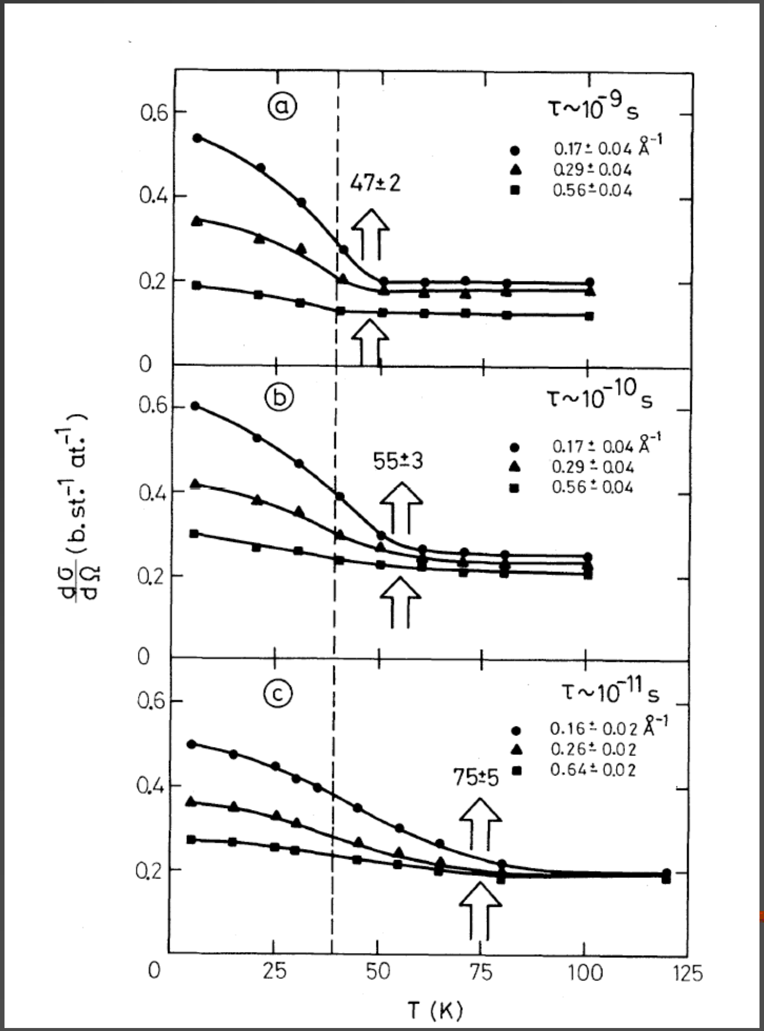
- Temperature dependence of the elastic scattering measured with three different instrumental resolutions, 1μeV(HFBS), 20μeV and 50μeV(LAM-80)
- With coarse energy resolutions, apparent increase of the intensity starts at higher temperatures
- With 1μeV probe (~250MHz~4x10^{-9}sec), spins are already static at very high temperatures as T* ~ 30K!



- Note: Tf = 5.8K
  - **T* ~ 5Tf!!!**

Typical spin-glass system

- Cu-Mn spin-glass:
  - Tf ~40K for 8% Mn.
- Neutron "elastic" signal with different resolutions, 1.5μeV, 25μeV and 230μeV.
  (Murani and Heidemann, PRL 41 (1978) 1402)



Summary for the ZnMgTb system

- Spin-glass-like behavior
  - Significant static short-range order at the lowest temperature
  - Resolution limited quasielastic peak below T_f
- Deviation from standard spin-glass
  - Broad inelastic peak of the collective excitations in the SRO clusters
  - Well above T_f (up to $5T_f$), SRO is formed and very slow
    - Existence of rigid spin clusters at $T >> T_f$
  - Spin relaxation is stretched exponential type even significantly higher temperature than T_f
    - Distribution of relaxation times at $T >> T_f$

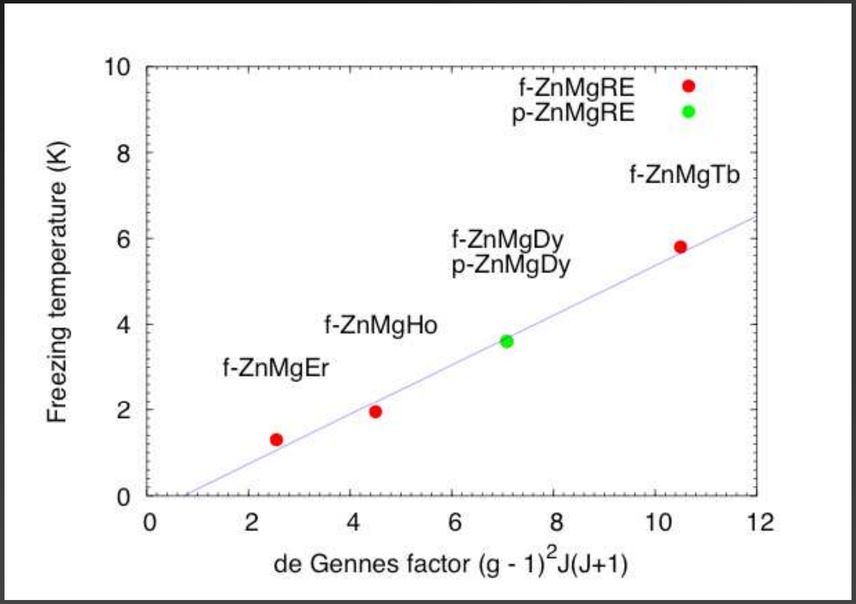
Cluster freezing | Rigid clusters with relxation time distribution | Paramagnetic

$T = 0K$ — $T_f = 5.8K$ — $\sim 30K$ — Temperature

How about other magnetic quasicrystals?

- We mostly understand the behavior of f-Zn-Mg-Tb quasicrystals. But others are the same?

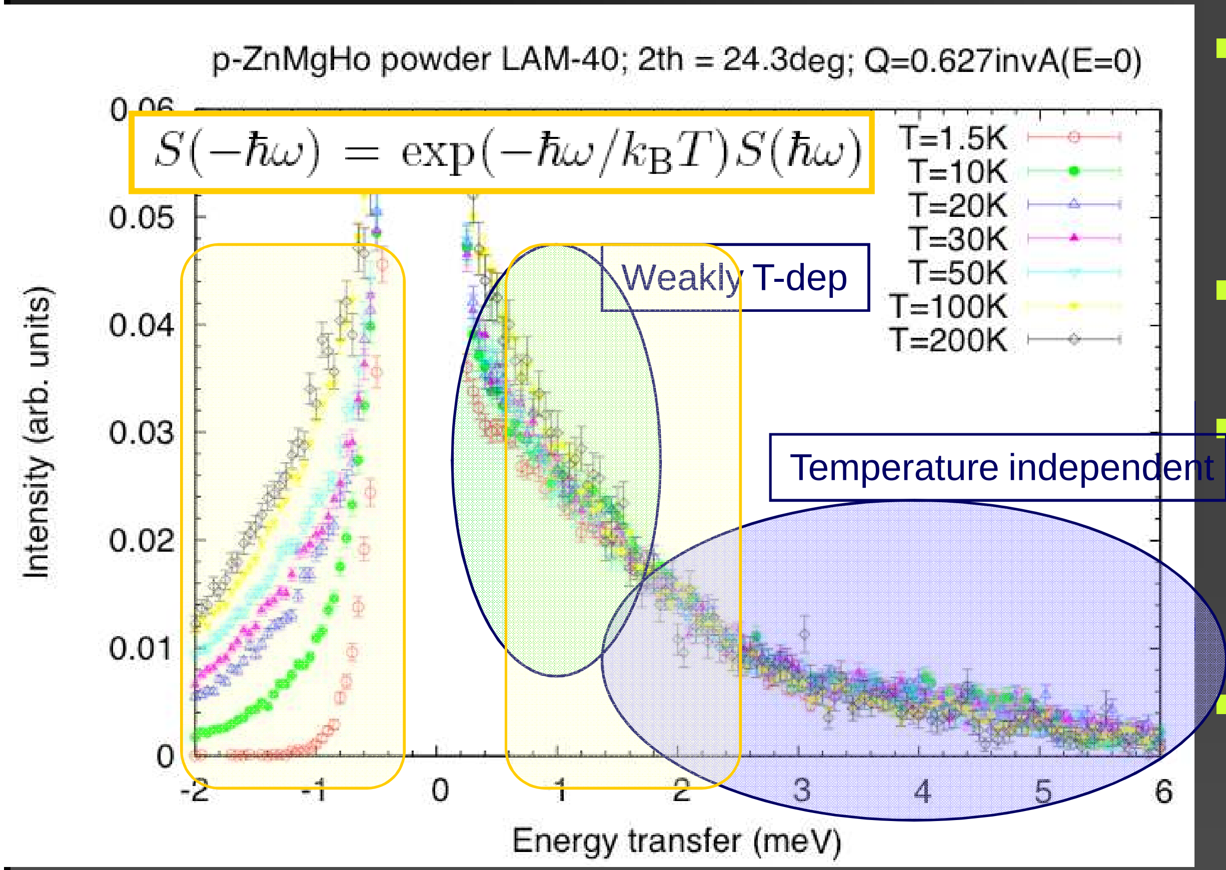
Other RE systems



- Tf decreases by changing RE from Tb (5.8K) to Er (1.35K)
- This change is scaled by the de Gennes factor
- No change in conduction electron-4f interaction strength but only S (of J) decreases.

- No qualitative difference expected...

f-ZnMgHo and p-ZnMgHo; unconventional $S(Q,\hbar\omega)$



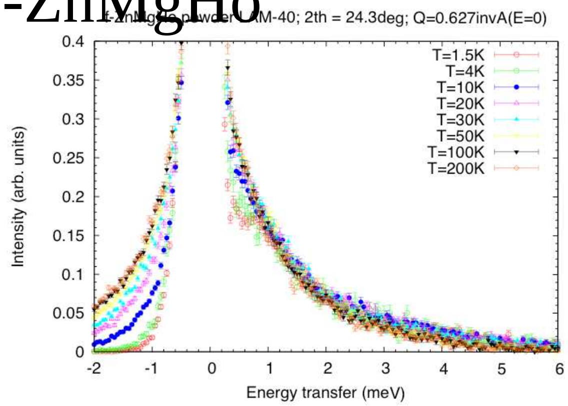
- Very weak temperature dependence in very wide temperature range 1.5K < T< 200K
- hw > 1.5meV
  - T-independent
- hw < 1.5meV
  - very weakly dependent
  - 200K ~ 17meV
- Detailed balance confirmed!

Q-independent: localized fluctuations

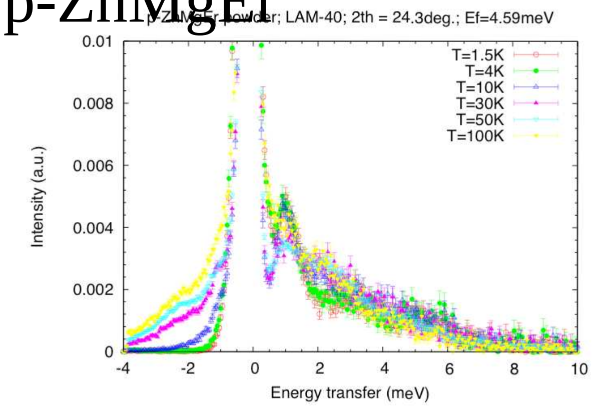
ZnMgHo is very different from others!

$S(hw) \sim (1-\exp(-\hbar\omega/k_B T))^{-1}\, \mathrm{Im}\chi(\hbar\omega)$

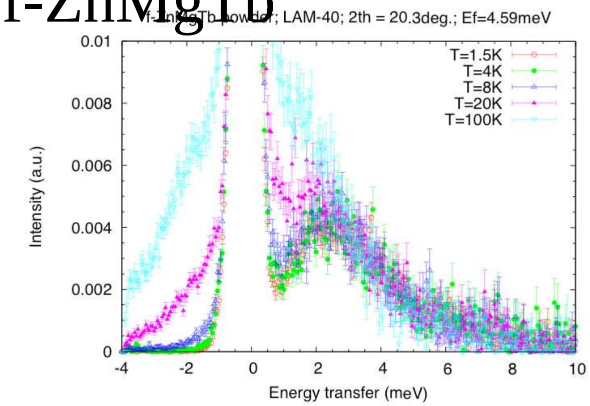
f-ZnMgHo



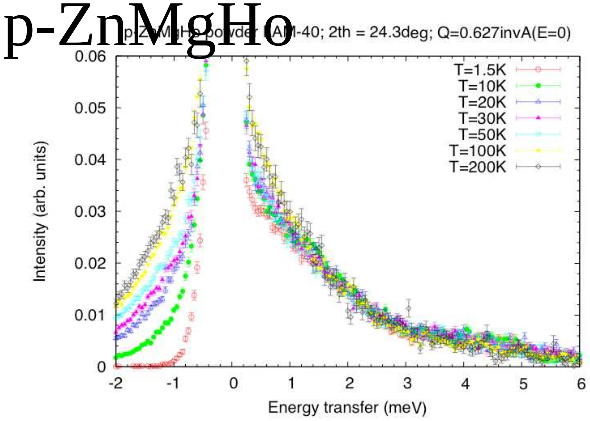
p-ZnMgEr



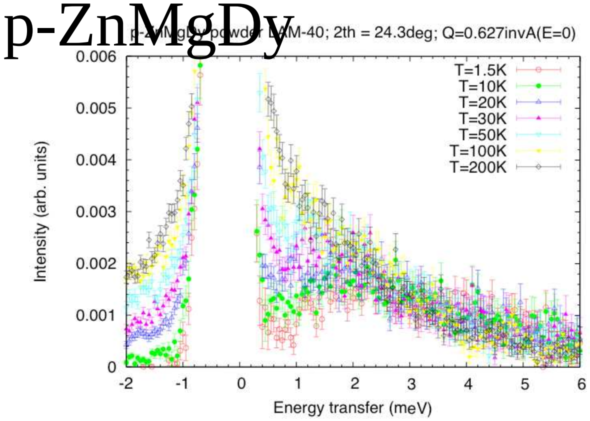
f-ZnMgTb



p-ZnMgHo

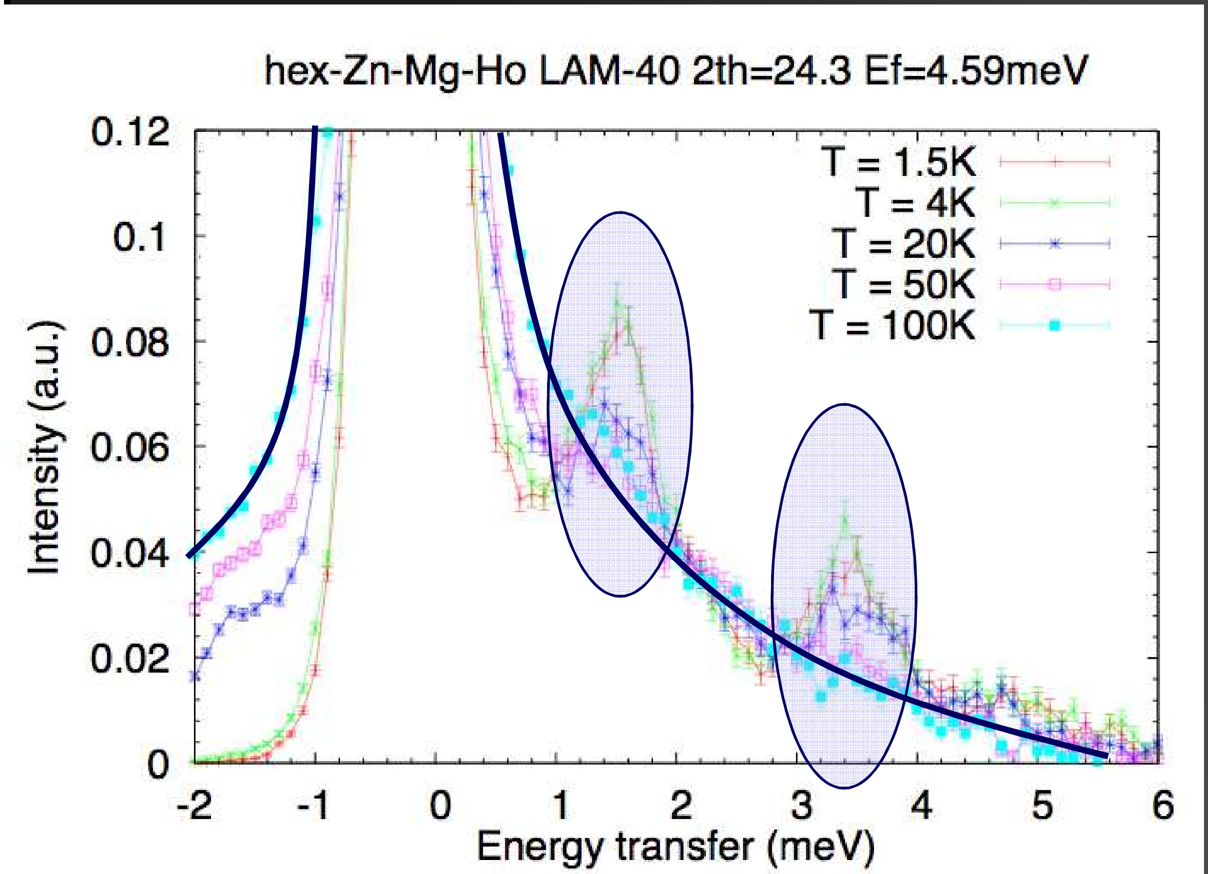


p-ZnMgDy



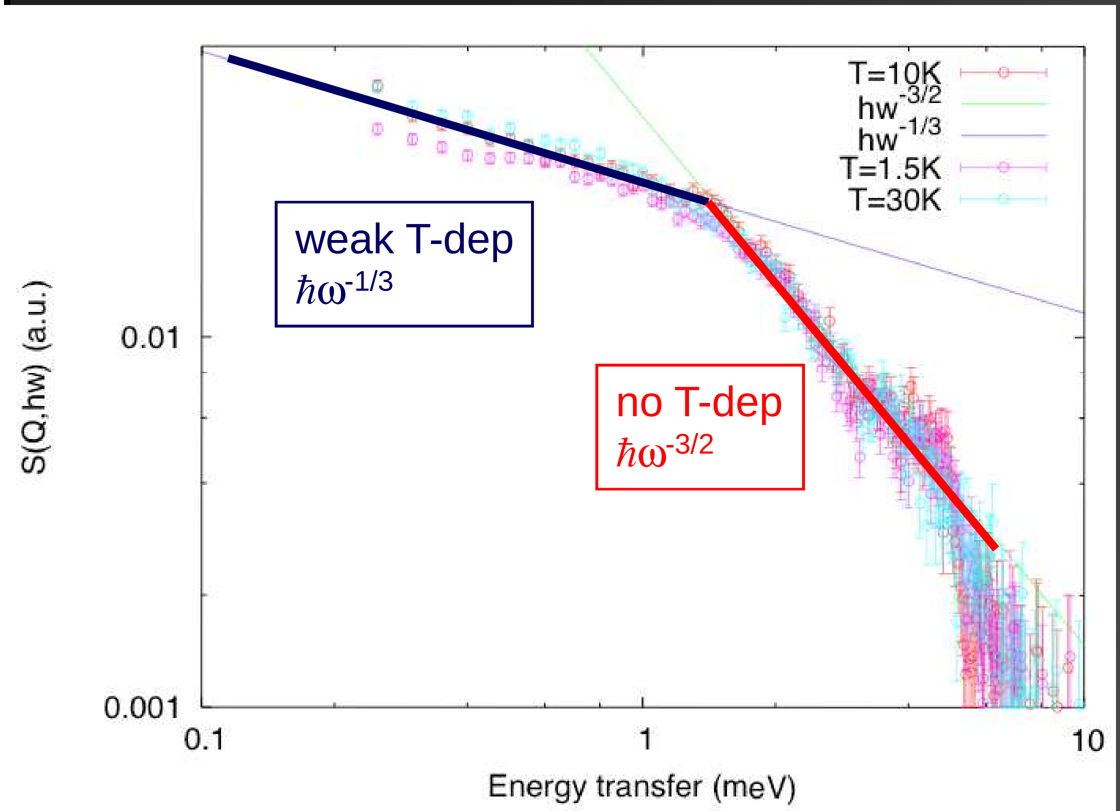
- Ho system only shows temperature independent $S(Q,\hbar\omega)$

hex-Zn-Mg-Ho; CEF peaks; very normal



- At high temperatures:
  - quasielastic
- T < 20K
  - Two CEF peaks
- Local environment around Ho is well defined
- CEF level analysis is in progress

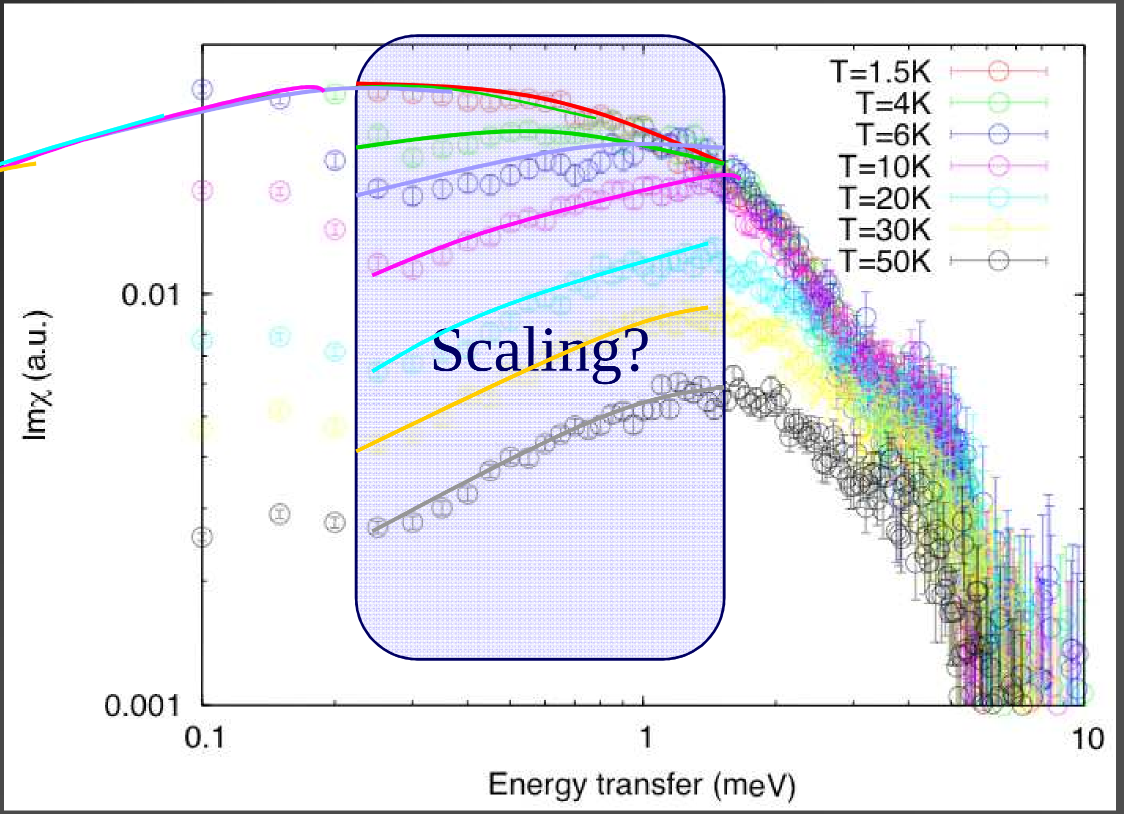
Temperature independent $S(Q,\hbar\omega)$



- Log plot suggests two energy regions:
- $\hbar\omega < 1.5$meV
  - Weak T-dependence
  - $\hbar\omega^{-1/3}$
- $\hbar\omega > 1.5$meV
  - No T-dependence
  - $\hbar\omega^{-3/2}$

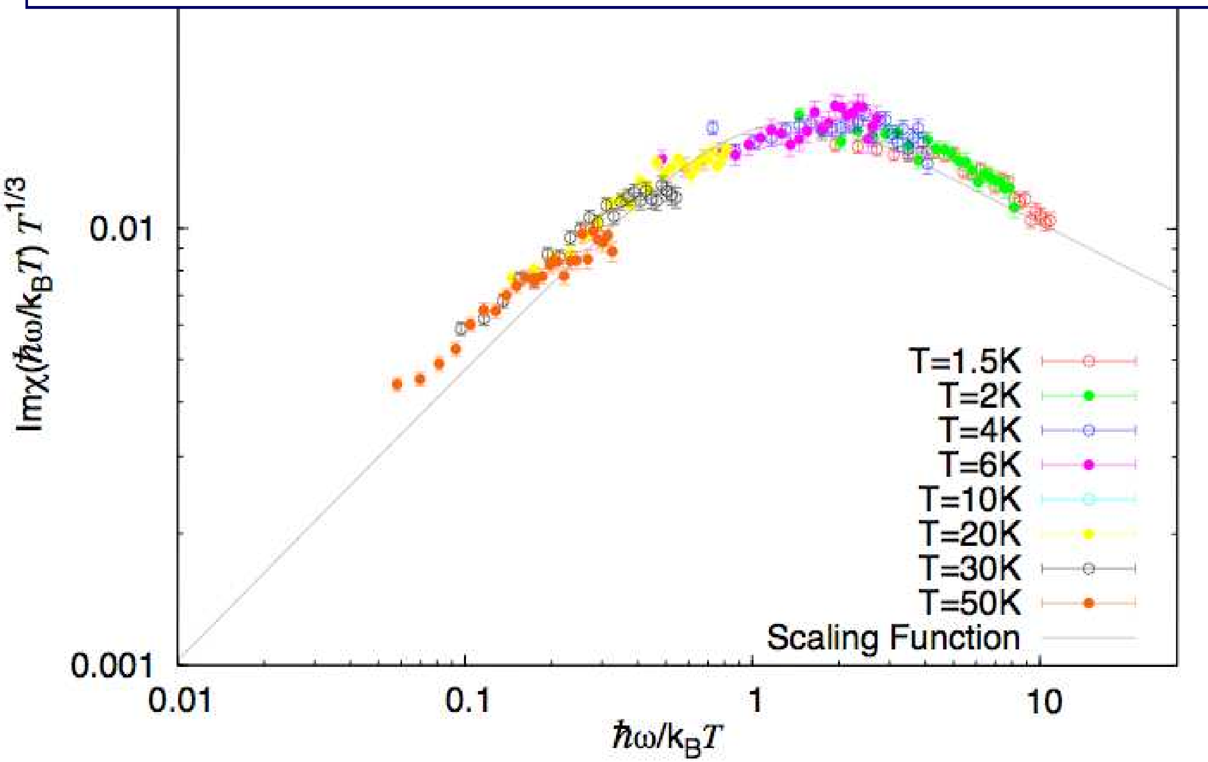
Imaginary part of generalized susceptibility Imχ($\hbar\omega$)

- Imχ($\hbar\omega$) is definitely T-dependent
  - c.f. f-ZnMgTb
- Scaling?



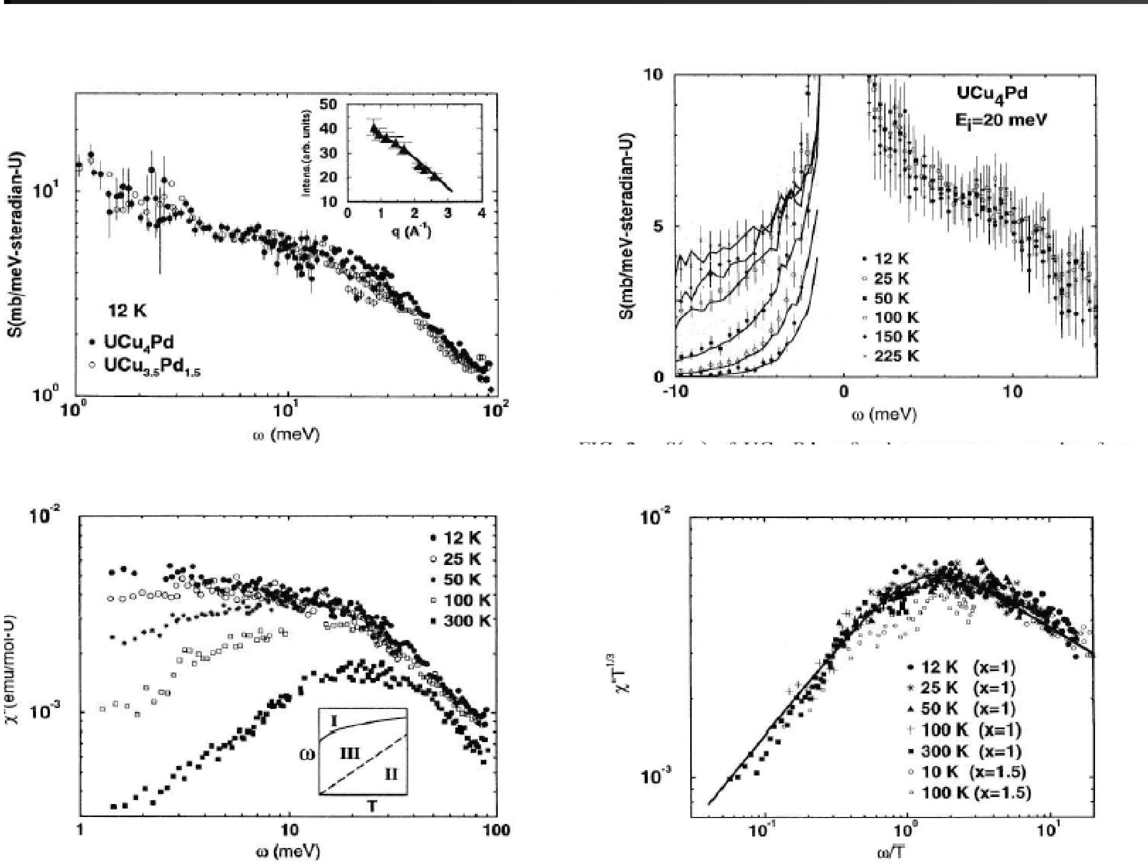
Dynamical scaling with $T_c = 0$ K

$$\mathrm{Im}\chi(\hbar\omega)(k_\mathrm{B}T)^{1/3} \propto (\hbar\omega/k_\mathrm{B}T)^{-1/3}\tanh(\alpha\hbar\omega/k_\mathrm{B}T)$$



- The scaling holds
- Note: no Tc
- Tc = 0 scaling

Systems with similar (identical) behavior



$UCu_{5-x}Pd_x$ Aronson *et al.*, PRL 75(1995)725

- The temperature independent S(Q,hw) is unusual
- Such S(Q,hw) is often observed in non-Fermi-liquid compounds:
- $UCu_{5-x}Pd_x$
  - Aronson *et al.*, PRL 75(1995)725.
- $Ce(Rh_{0.8}Pd_{0.2})Sb$
  - Park et al., JPCM 14 (2002) 3865.
- $Ce(Ru_{1-x}Fe_x)_2Ge_2$ (x=0.76)
  - Montfrooji et al., PRL 91 (2003) 087202.
- $Sc_{1-x}U_xPd_3$ (x=0.35)
  - Wilson et al. PRL 94 (2005) 056402.

But why similar? some clues?

- T=0 phase transition appears where T_f or T_N disappears
  - Note: Tf = 1.9K for ZnMgHo quasicrystals

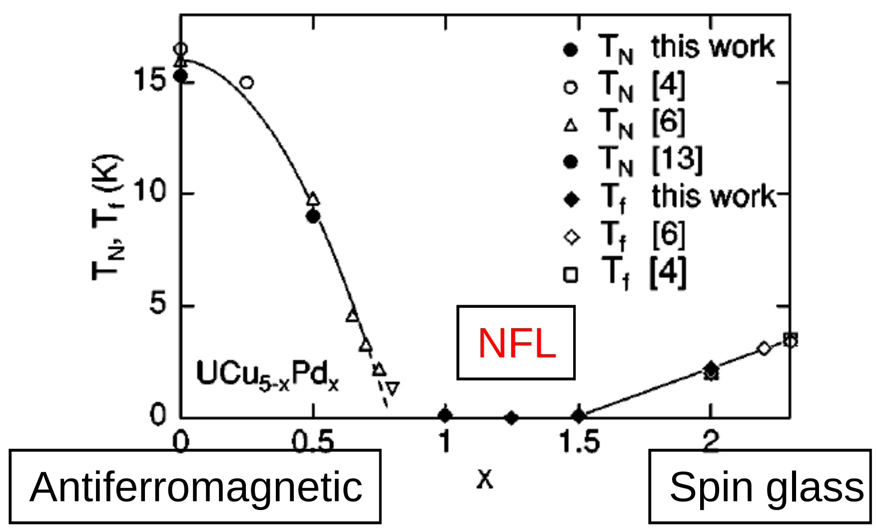


FIG. 1. Magnetic phase diagram T vs x of $UCu_{5-x}Pd_x$. T_N indicates the Nèel temperature, T_f the spin-glass freezing temperature.

- Fermi liquid
  - Strong electron correlations result in formation of quasiparticles with renormalized mass following Fermi statistics
- Non-Fermi Liquid
  - Quasiparticles no longer behave normal Fermi liquid because of magnetic instability
- QC electrons may have dense anomalies in DOS; NFL?
  - Structural origin

Summary for the ZnMgHo system non-conventional $S(Q, \hbar\omega)$

- Inelastic spectrum is temperature independent up to 200K!!!
- There are two energy regions
  - $\hbar\omega > 1.5$meV: $S(\hbar\omega) \sim (\hbar\omega)^{-3/2}$
  - $\hbar\omega < 1.5$meV: $S(\hbar\omega) \sim (\hbar\omega)^{-1/3}$
- For the lower energy regions, $\hbar\omega$ /T scaling can be seen
  - $\mathrm{Im}\chi(\hbar\omega, T)T^{1/3} \sim (\hbar\omega /T)^{-1/3} Z(\hbar\omega /T)$
- Similar to several NFL systems
  - $UCu_{5-x}Pd_x$ (x = 1,1.5): Aronson et al. PRL 75 (1995) 725
  - $Ce(Rh_{0.8}Pd_{0.2})Sb$: Park et al. JPCM 14 (2002)3865
  - $Ce(Ru_{1-x}Fe_x)_2Ge_2$ (x = 0.76): Montfrooij et al, PRL 91(2003)087202
  - $Sc_{1-x}U_xPd_3$ (x = 0.35): Wilson et al. PRL 94(2005)056402

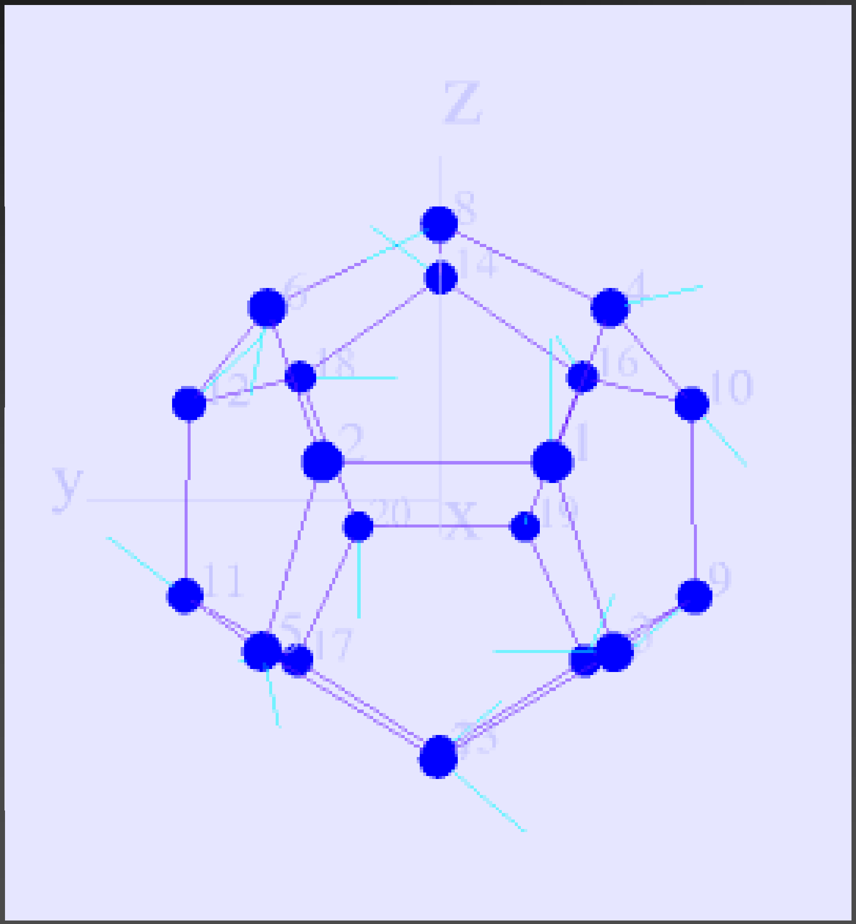
Conclusions

- There are two qualitatively different spin excitations in ZnMgRE quasicrystalline magnets
  - Normal T-dependent $S(Q, \hbar\omega)$: ZnMgTb
  - Unusual T-independent $S(Q, \hbar\omega)$: ZnMgHo
- T-dependent $S(Q, \hbar\omega)$ can be understood more or less as a sort of spin-glasses; several deviations from standard spin-glasses are found, though.
- T-independent $S(Q, \hbar\omega)$ is very unusual and thus may be interesting for further study
- Qualitative similarity for the NFL system is suggested

Acknowledgemens

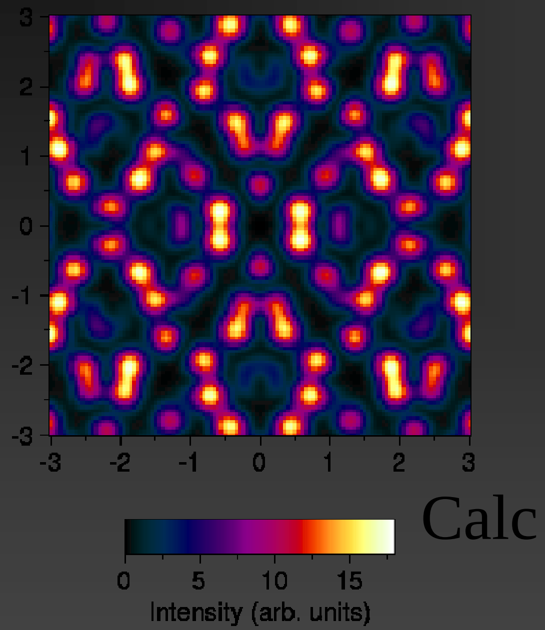
- LAM experiments at KENS, KEK
  - Dr. A. P. Tsai (Tohoku)
  - Dr. H. Takakura (Osaka)
  - Dr. K. Shibata (JAERI)
- HFBS experiments at NCNR, NIST
  - Dr. Z. Chowdhuri (NIST)
  - Dr. R. Dimeo (NIST)
  - Dr. D. Neumann (NIST)
- Reactor-based experiments at JAERI, Tokai
  - Dr. K. Ohoyama (Tohoku)

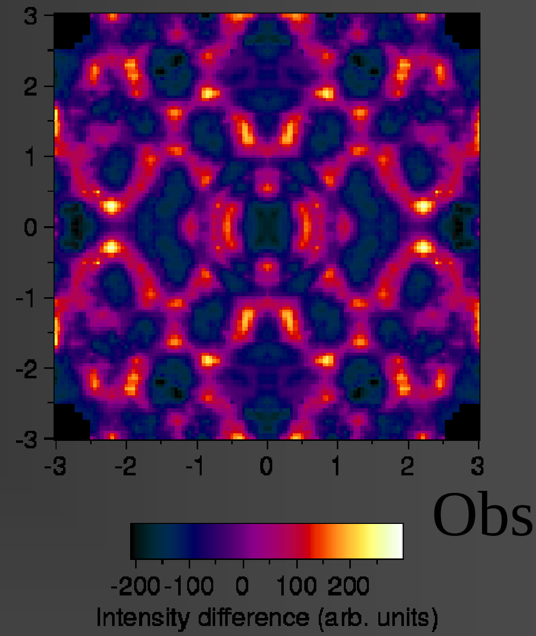
Dodecahedral spin cluster model



- Dodecahedron
- N.N.N antiferromagnetic interactions
- Heisenberg (isotropic) spins
- $H = \Sigma_{ij}\, S_i . S_j$
- Ground state spin configuration is determined by numerical minimization (Axenovich and Luban PRB 63 (2001) 100407)

Verification of the dodecahedral model



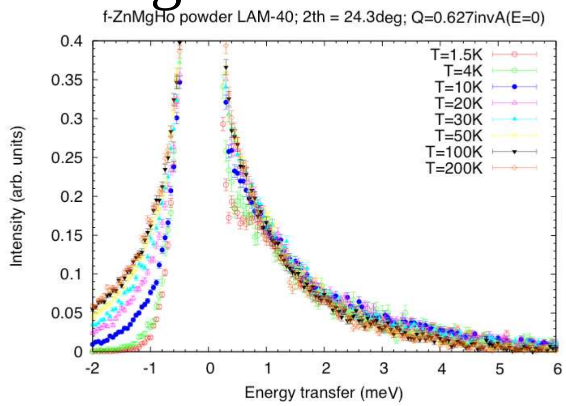


- Elastic diffuse scattering is well reproduced by the very simple single dodecahedral cluster model
- Proper statistical average is taken.

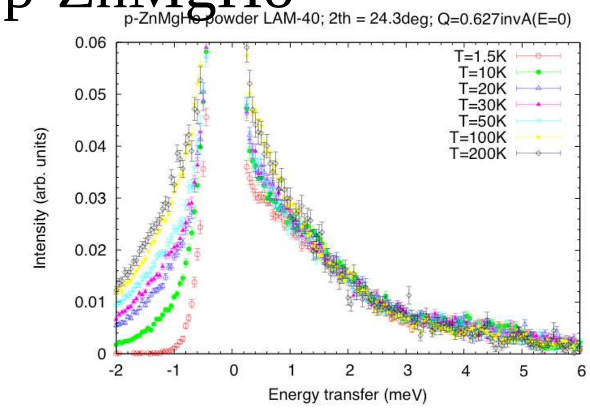
Spin dynamics; inelastic spectra for various QCs

Temperature independent S(Q,w)!

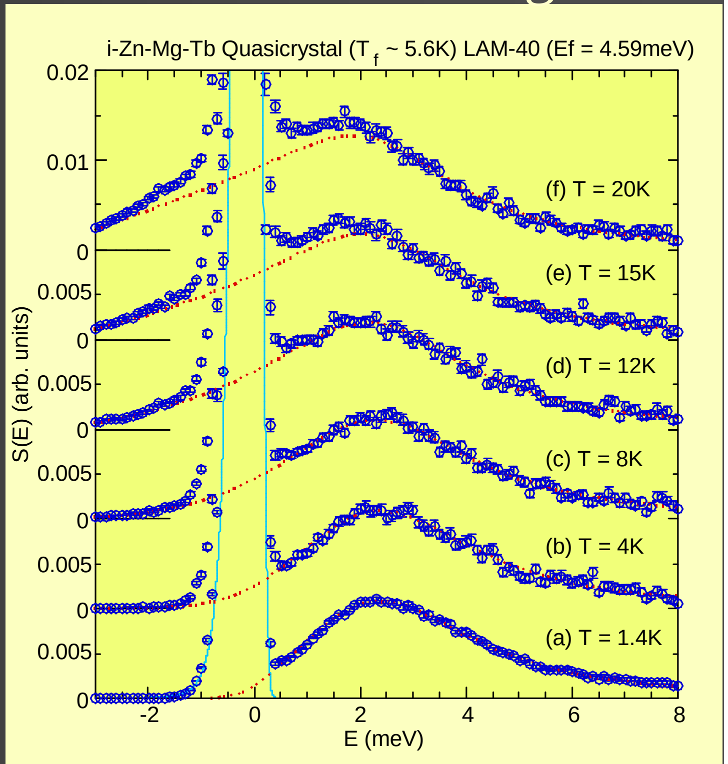
f-ZnMgHo



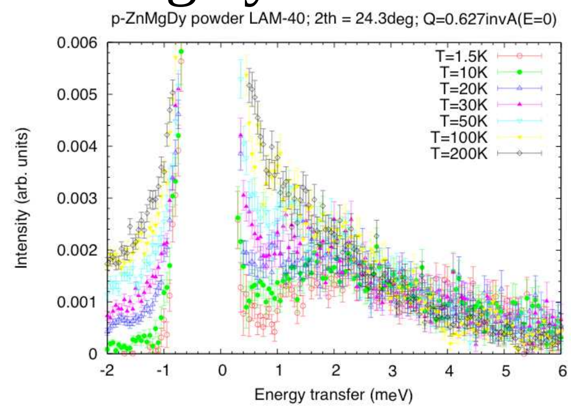
p-ZnMgHo



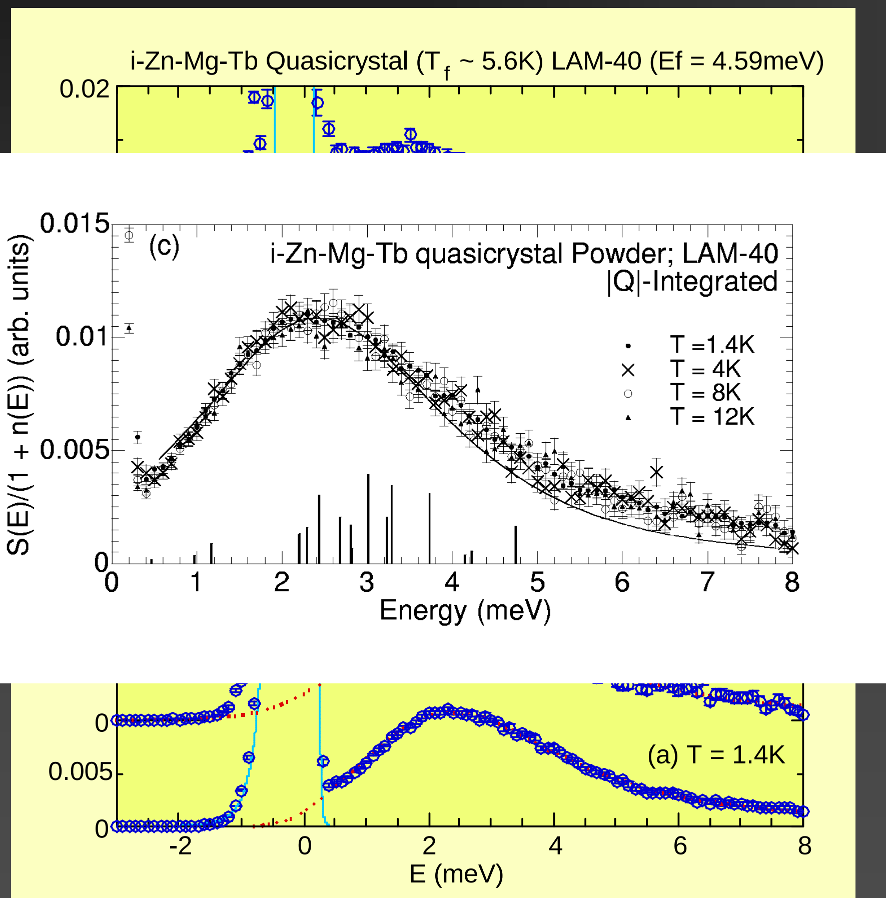
f-ZnMgTb



p-ZnMgDy



Temperature dependence

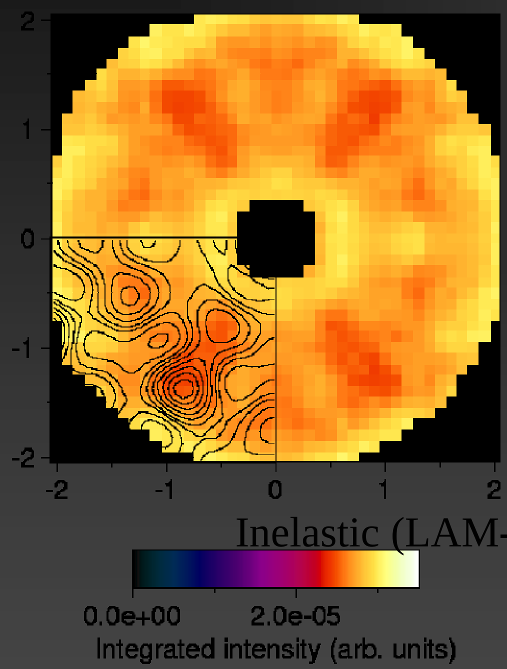


- Intensity of the inelastic peak increases at higher temperature.
- Scaled by Bose temperature factor, $(1 - \exp(-E/k_B T))^{-1}$.
- Spectral shape does not change significantly.

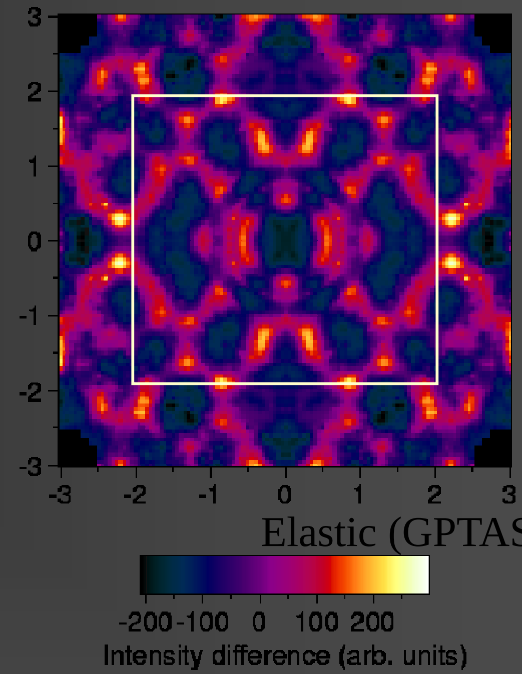
- Not a crystalline electric field splitting level transition (CEF: Boltzmann)
- but Boson
- Similar to “Boson peak” in glasses

Single QC inelastic experiment

Zn-Mg-Tb



Inelastic (LAM-40)



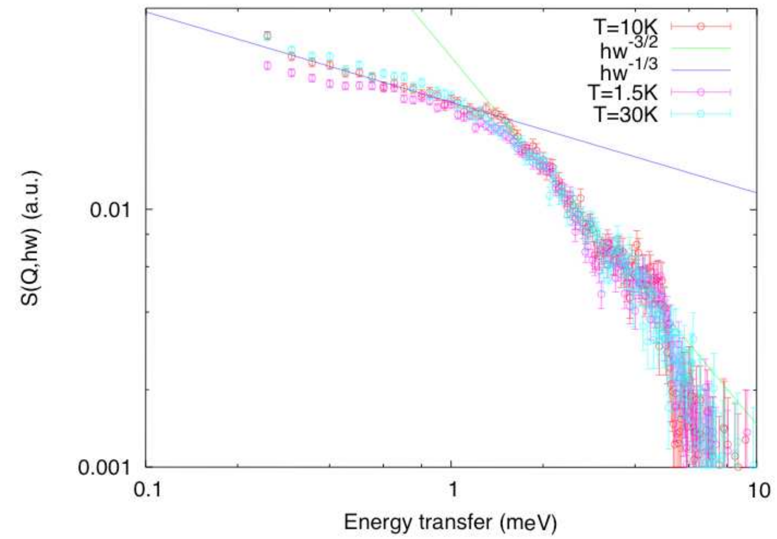
Elastic (GPTAS)

- Inelastic scattering intensity (1 < E < 4meV)
  - Intensity is similar to elastic diffuse scattering (magnetic structure factor)
- Spin-wave-type modes localized in the short-range ordered region?

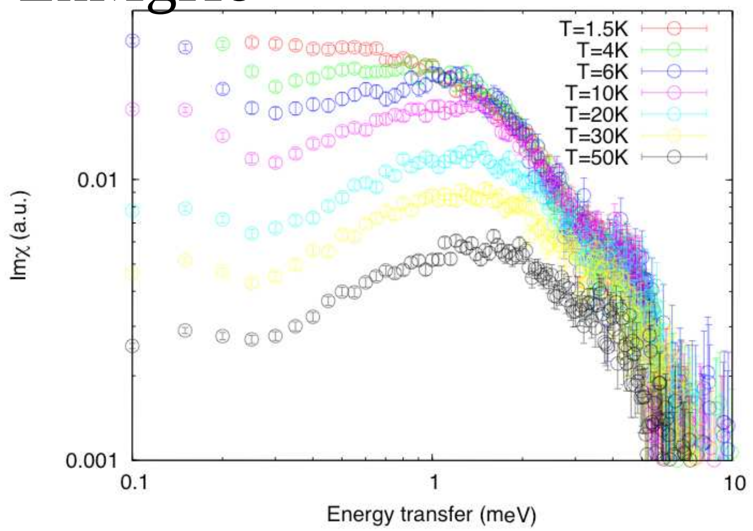
Temperature independent S(Q,ω)

- Two energy regions for different power
  - $\omega^{-3/2}$ for $\hbar\omega > 1.5$meV
  - $\omega^{-1/3}$ for $\hbar\omega < 1.5$meV

p-ZnMgHo



p-ZnMgHo



$$\mathrm{Im}\chi(\hbar\omega, T) \propto S(Q, \hbar\omega)[n(\hbar\omega) + 1]^{-1}$$

Dynamical scaling with Tc = 0K!

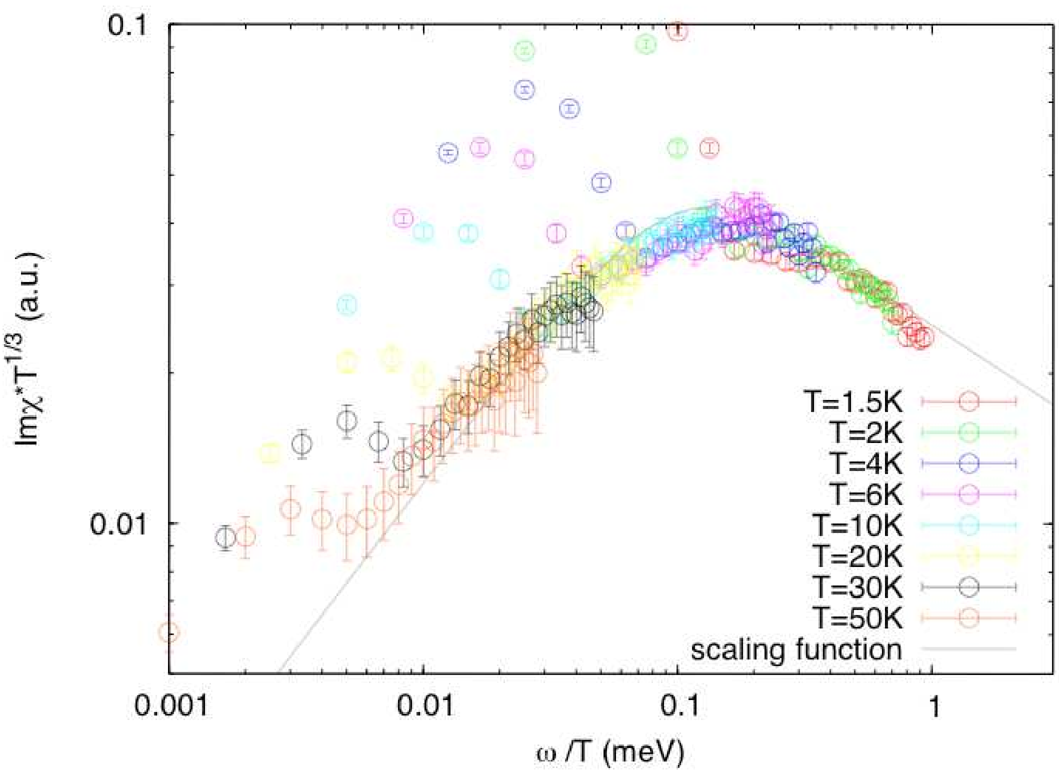
- Imχ for $\hbar\omega$ < 1.5meV can be scaled to single function

$$\mathrm{Im}\chi(\hbar\omega, T)T^{1/3} \propto (\hbar\omega/T)^{-1/3}\mathsf{Z}(\hbar\omega/T)$$

$$\mathsf{Z}(\hbar\omega/T) \sim [n(\hbar\omega)+1]^{-1}$$

$$\mathsf{Z}(\hbar\omega/T) \sim \tanh(10\hbar\omega/T)$$

◆E/T scaling for spin fluctuations
◆T=0 phase transition?



Quantum criticality in UCuPd

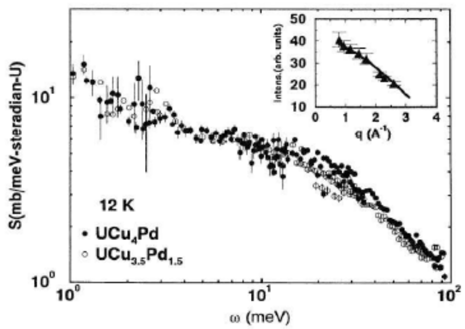


FIG. 1. The magnetic scattering intensities $S(\omega)$ for UCu_4Pd (filled circles) and $UCu_{3.5}Pd_{1.5}$ (open circles) at 12 K. Inset: $S(\omega)$ integrated over ω, as a function of wave-vector magnitude q. Solid line is the magnetic form factor for U^{3+}/U^{4+} ions [13].

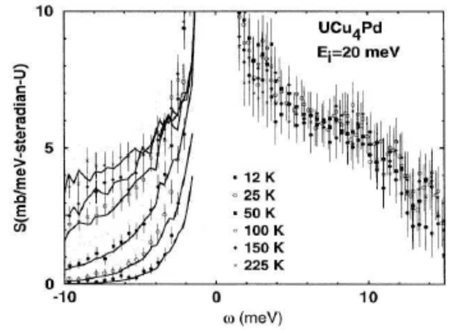


FIG. 2. $S(\omega)$ of UCu_4Pd at fixed temperatures ranging from 12 to 225 K. The incident energy is 20 meV. Solid lines for energy gain are calculated from the energy loss part of the neutron spectrum by $S(\omega > 0)\exp(-\omega/T) = S(\omega < 0)$, demonstrating that the magnetic scattering obeys detailed balance.

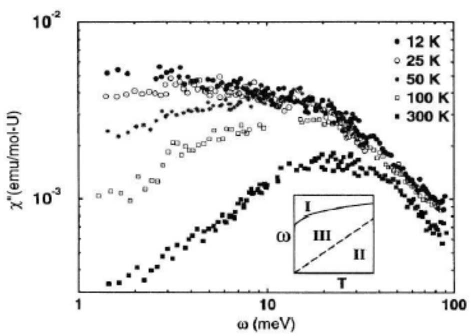


FIG. 3. $\chi''(\omega)$ for UCu_4Pd at constant temperatures ranging from 12 to 300 K. Inset: three qualitatively different regimes are observed: I ($\omega \geq \omega^*$): $\chi''(\omega, T) \sim (\omega/T_K)/[1 + (\omega/T_K)^2]$, $T_K \sim 8$ meV II ($\omega \ll T$): $\chi''(\omega, T) \sim \omega/T^{1\pm1/3}$ III ($\omega \gg T$): $\chi''(\omega, T) \sim \omega^{-1/3}$. Solid line: $\omega^*(T)$, dashed line: $\omega = T$.

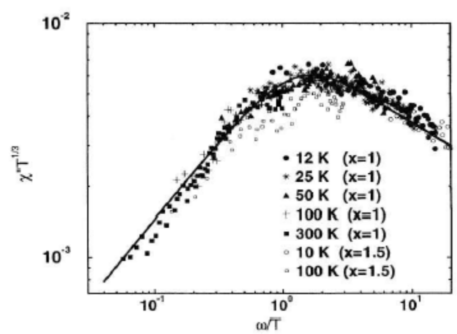


FIG. 4. $\chi''(\omega, T)T^{1/3}$ has almost identical universal scaling properties for both compounds $UCu_{5-x}Pd_x$ ($x = 1, 1.5$). Data with energy transfers $\omega \geq 25$ meV are not included in this plot. Solid line: $\chi''(\omega, T)T^{1/3} \sim (T/\omega)^{1/3}\tanh(\omega/1.2T)$.

- Exactly the same inelastic response was observed in non-Fermi-liquid UCuPd (Aronson et al PRL 75(1995)725).

Phase diagrams

- T=0 phase transition appears where T_f or T_N disappears.

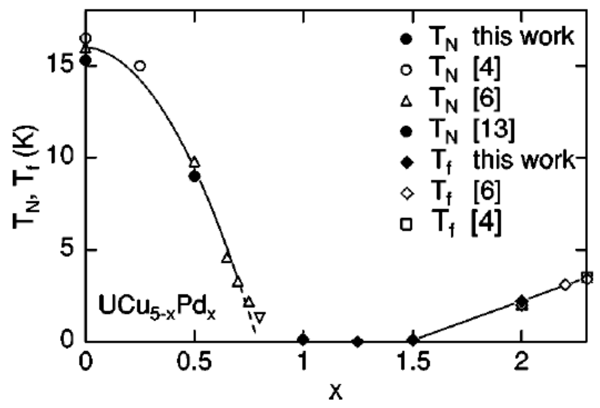
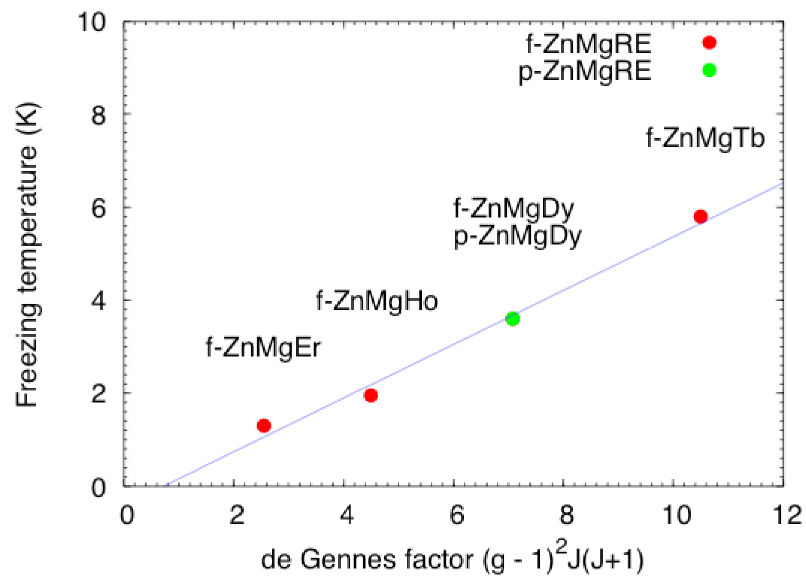


FIG. 1. Magnetic phase diagram T vs x of $UCu_{5-x}Pd_x$. T_N indicates the Nèel temperature, T_f the spin-glass freezing temperature.



Conclusions

- Magnetic quasicrystals all shows diffuse scattering indicating short-range nature of magnetic correlations.
- Diffuse scattering pattern in i-Zn-Mg-RE can be well reproduced by single dodecahedral spin object.

- Spin excitations are categorized in two cases:
  - Usual T-dependent $S(Q,\omega)$,
  - Unusual T-independent $S(Q,\omega)$.

- Possible relation to T=0K phase transition (QCP).

Magnetic quasicrystals

- 3d electron systems
  - i-Al-Mn, i-Al-Si-Mn (rapidly quenched metastable)
  - i-Al-Mn-Ge, i-Al-Pd-Mn, i-Al-Cu-Mn (weak moment)
  - i-Zn-TM-Sc (well defined 3d moment for TM = Fe)
- 4f electron systems
  - i-Zn-Mg-RE, i-Cd-Mg-RE (localized 4f moment)

- Present study:
  - i-Zn-Mg-RE (RE = Tb and Ho)