Australian Government

ANSTO

Triple Axis Spectrometers

SIKA & TAIPAN

Dr. Kirrily Rule

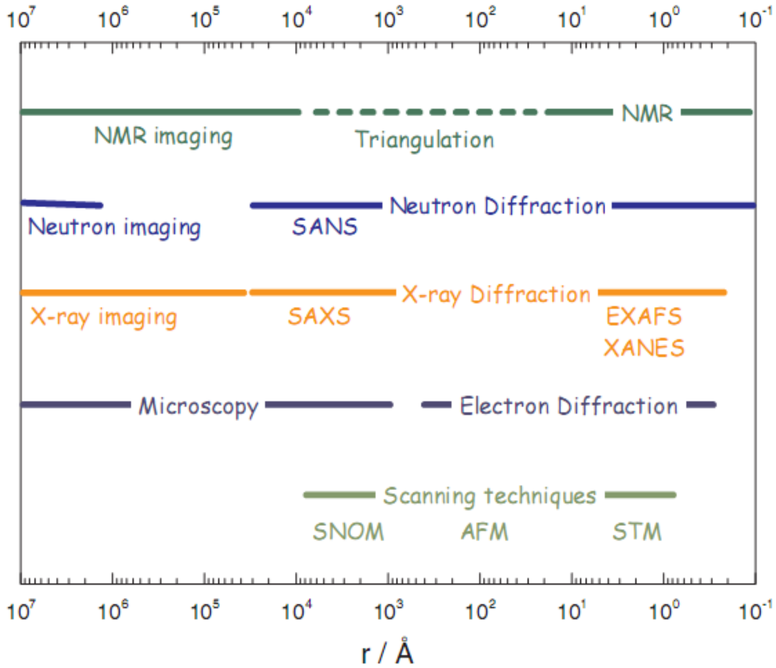
Australian Centre for Neutron scattering

Science. Ingenuity. Sustainability.

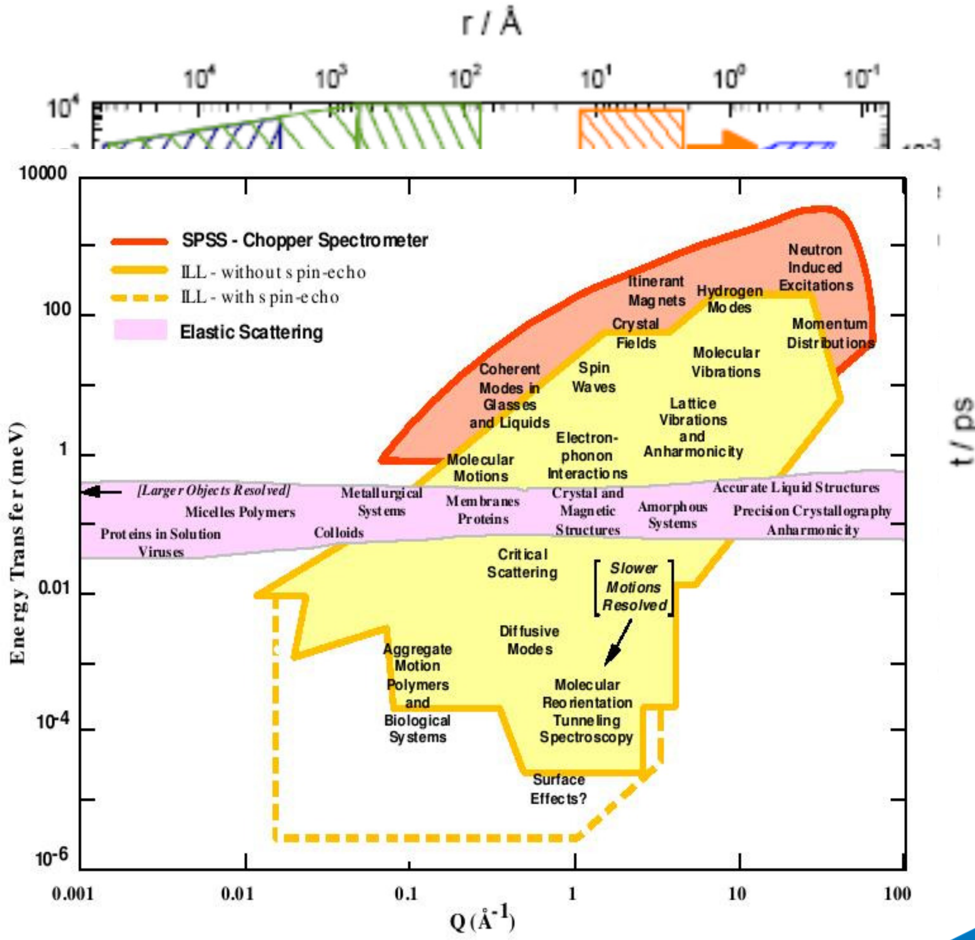
Outline

- Introduction to the Triple Axis Spectrometers, TAIPAN and SIKA
- Some of the basics: Inelastic scattering vs Elastic scattering
- Triple Axis Spectrometers
  - How do they work?
  - What can they tell us
  - Configuration requirements
  - Components
- Recent scientific examples

Scope of Neutrons



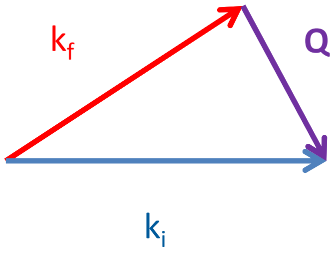
- Wavelength
- Energy
- Strong Interaction
- Highly penetrating
- Magnetic moment
- Non-destructive



Lander, G. H. and Emery, V. J. (1985). *Nucl. Instrum. Methods B* 12, 525.

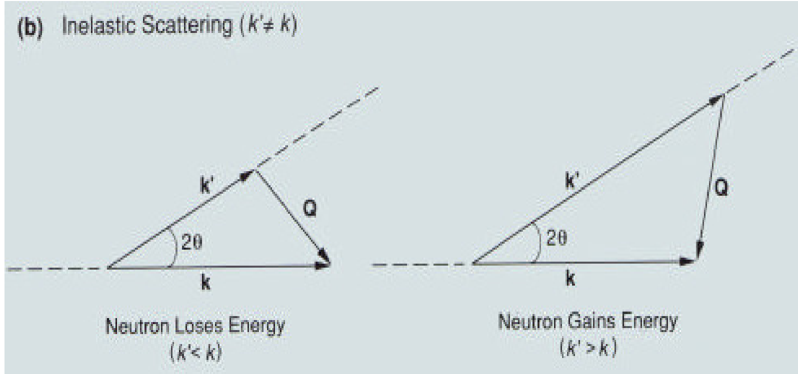
Inelastic scattering: conservation

Scattering triangle



$$k_i - k_f = Q$$

$$\frac{\hbar^2 {k_i}^2}{2m} - \frac{\hbar^2 {k_f}^2}{2m} = \Delta E$$



Differential Inelastic scattering cross-section:

\# neutrons scattered/sec into solid angle dΩ and dE

Matrix element for moving between initial and final states

Scattering potential is either:

- very short range strong nuclear force
- Dipole-dipole coupling with unpaired electrons

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\left(\frac{m}{2\pi\hbar^2}\right)^2 \sum_{\lambda_i, s_i} P\lambda_i P s_i \sum_{\lambda_f, s_f} \left|\langle k_f s_f \lambda_f | \hat{V} | k_i s_i \lambda_i \rangle\right|^2 \delta\left(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega\right)$$

Probability of being in the initial state and final state

Energy conservation term for the energy transfer ħω

Introduction to the theory of thermal neutron scattering. G.L. Squires

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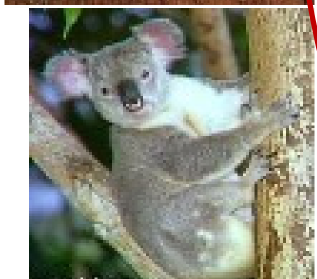
The OPAL zoo

Echidna
high resolution powder

Wombat
high intensity powder

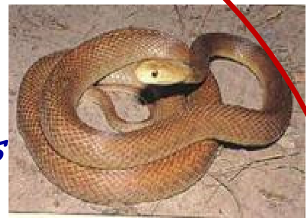
Kowari
residual stress

Koala
single crystals



Joey
Alignment diffractometer

Taipan
thermal triple axis

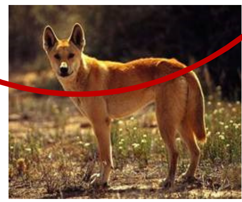


Pelican
Cold time-of-flight

Sika
cold triple axis

Emu
backscattering

Dingo
radiography



Quokka
Pinhole SANS

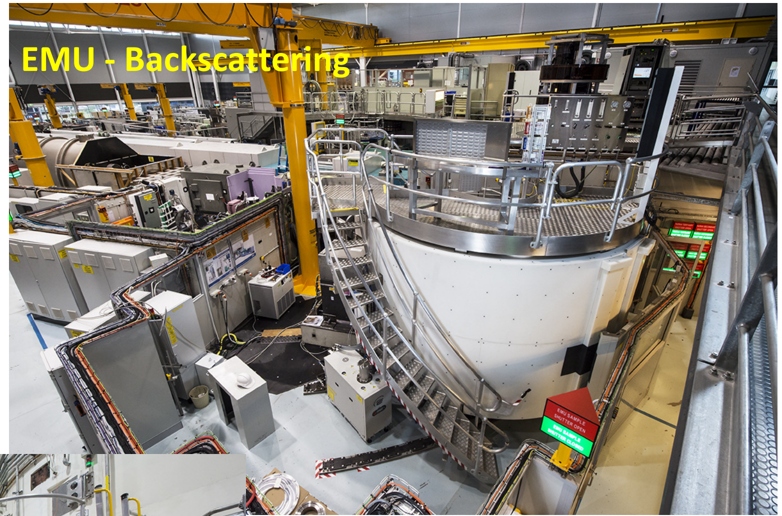
Platypus
reflectometry

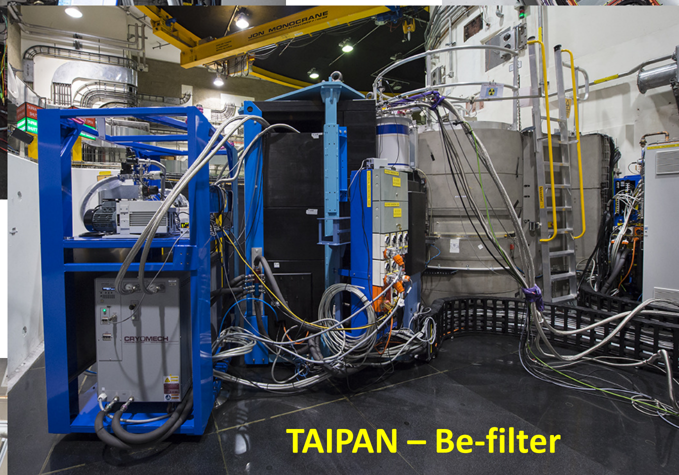
Kookaburra
ultra-SANS

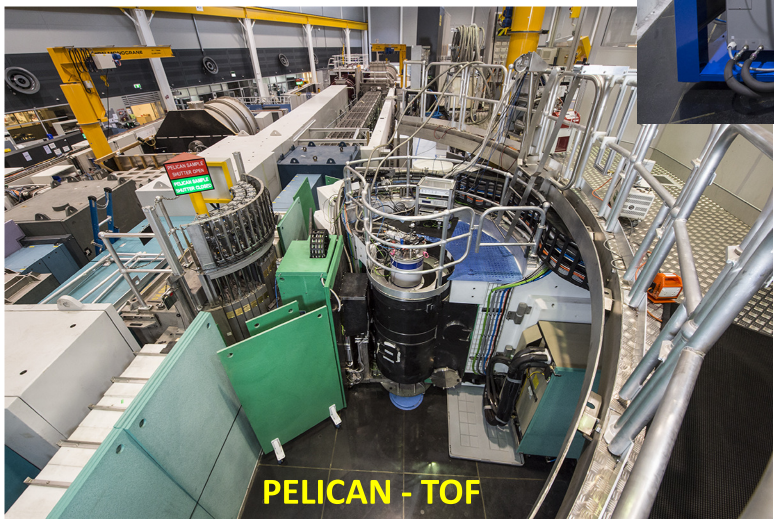
Bilby
2nd pinhole SANS

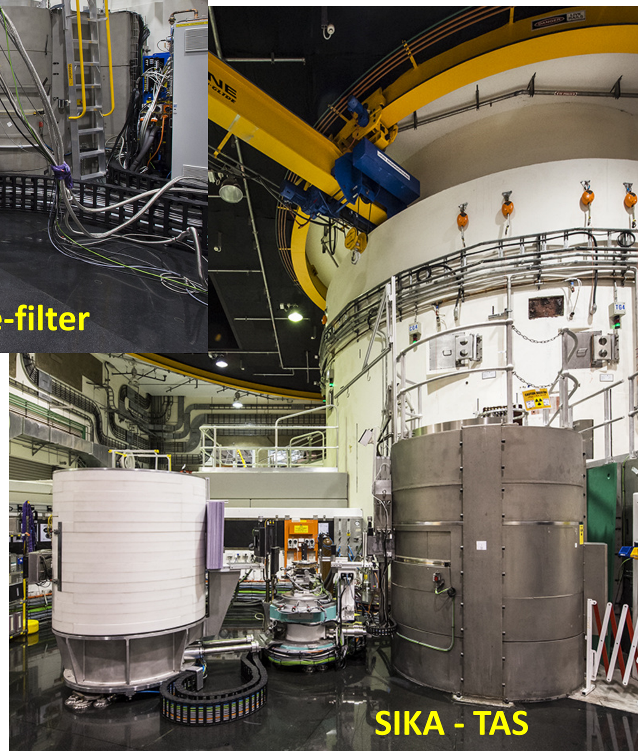
Spatz
reflectometry

Inelastic suite:

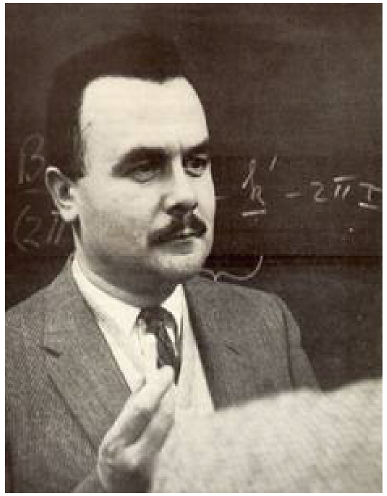


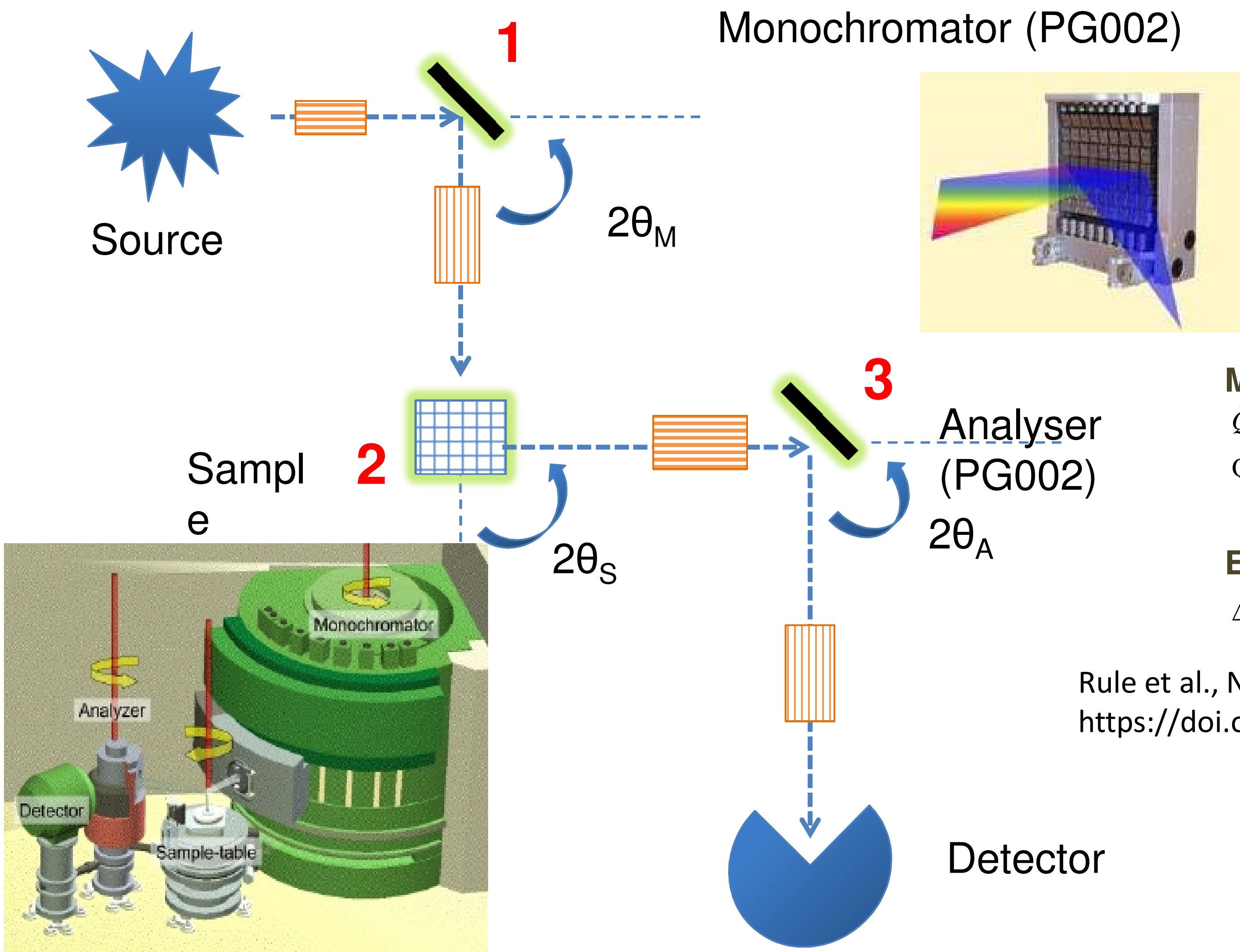






Triple Axis Spectrometer: TAIPAN and SIKA





Momentum Transfer

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

$$Q^2 = k_i^2 + k_f^2 - 2k_ik_f \cos\phi$$

Energy Transfer

$$\Delta E = E_i - E_f = \frac{\hbar^2}{2m}\left(\vec{k}_i^{\,2} - \vec{k}_f^{\,2}\right)$$

Rule et al., NIMA. **90** (2018) 140-149
https://doi.org/10.1016/j.nima.2018.05.056

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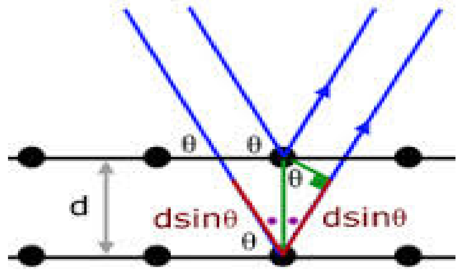
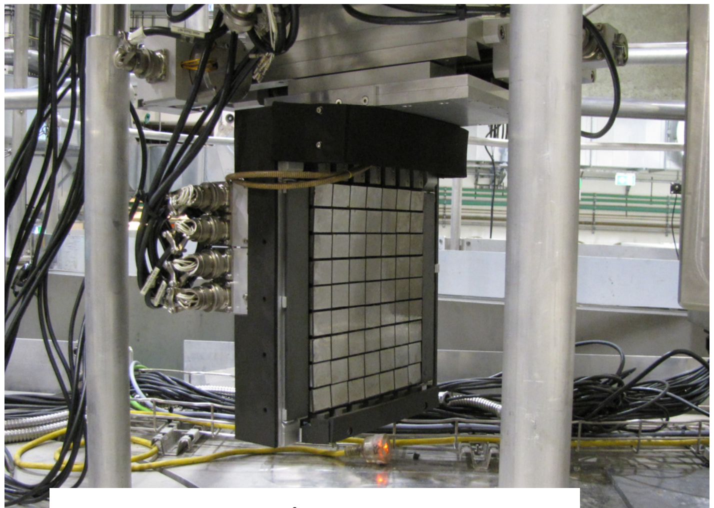
Monochromator and Analyser

- A monochromatic beam is achieved by Bragg scattering from the crystal (PG002)

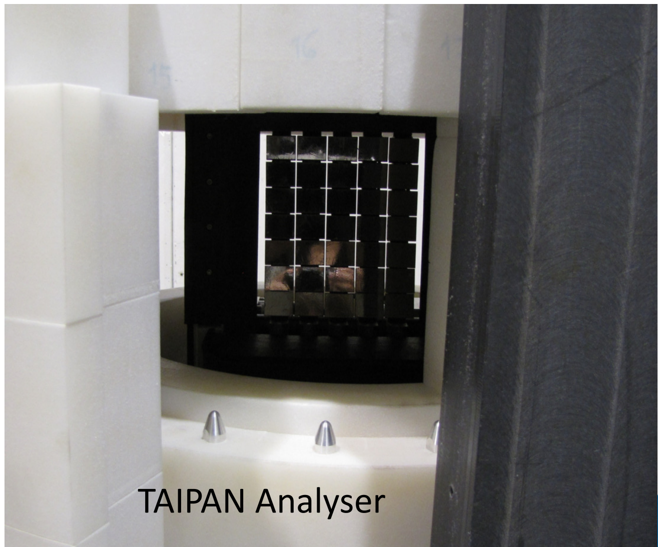
$$2d \sin\theta = n\lambda \text{ where } n=1,2,3$$



- Focusing is achieved by tilting the blades to give a concave surface to the incoming beam.



TAIPAN monochromator

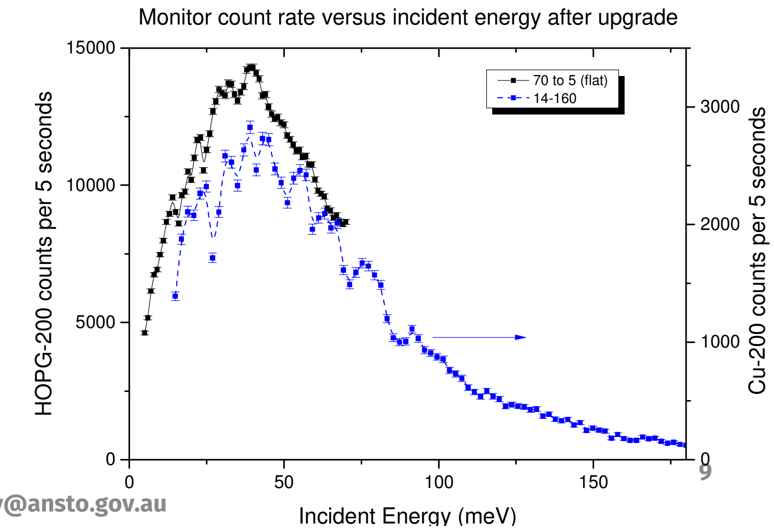


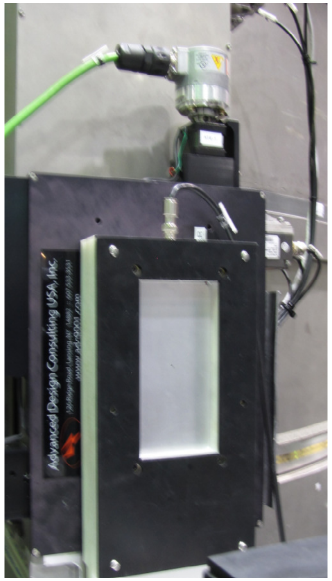
TAIPAN Analyser

Monitor and detector

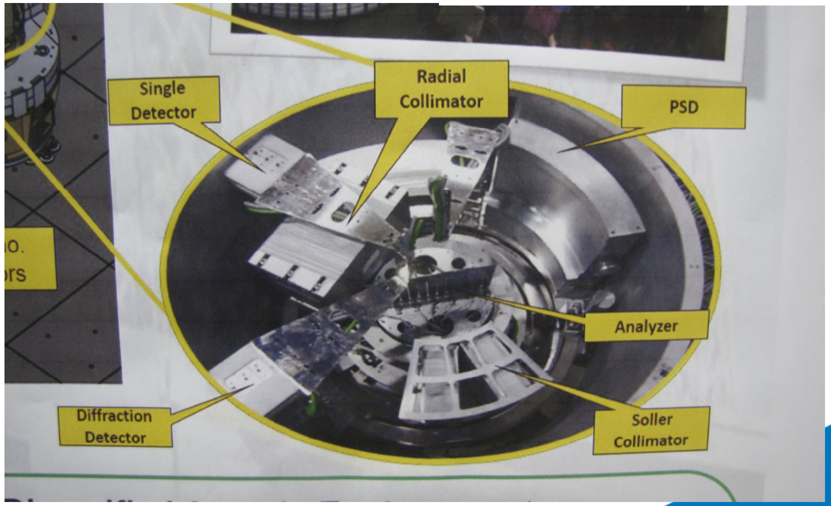
- Low efficiency beam monitor located before the sample is used to determine the number of neutrons that reach your sample. This means that higher flux energies are quicker than the lower flux energies.

- ^{3}He detector tubes used for detectors
  - Typically a single cylinder detector
  - SIKA has additional detectors



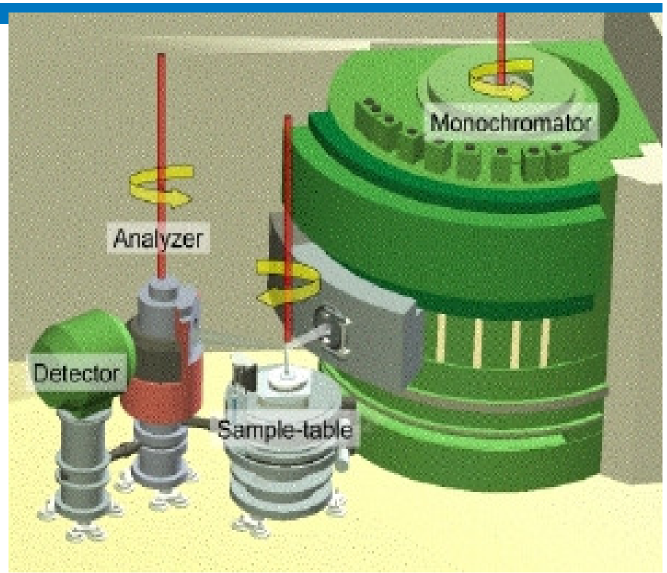


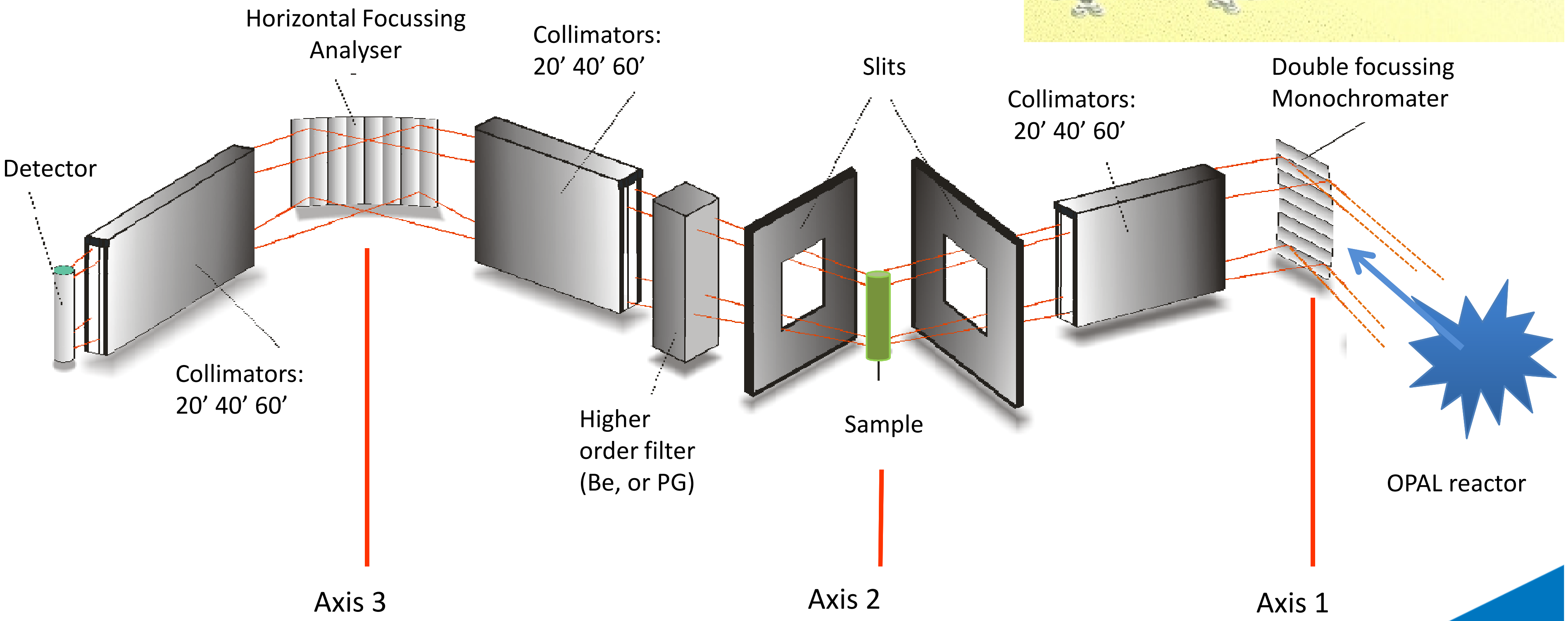
SIKA beam monitor



SIKA analyser/detector drum

Triple Axis Spectrometer





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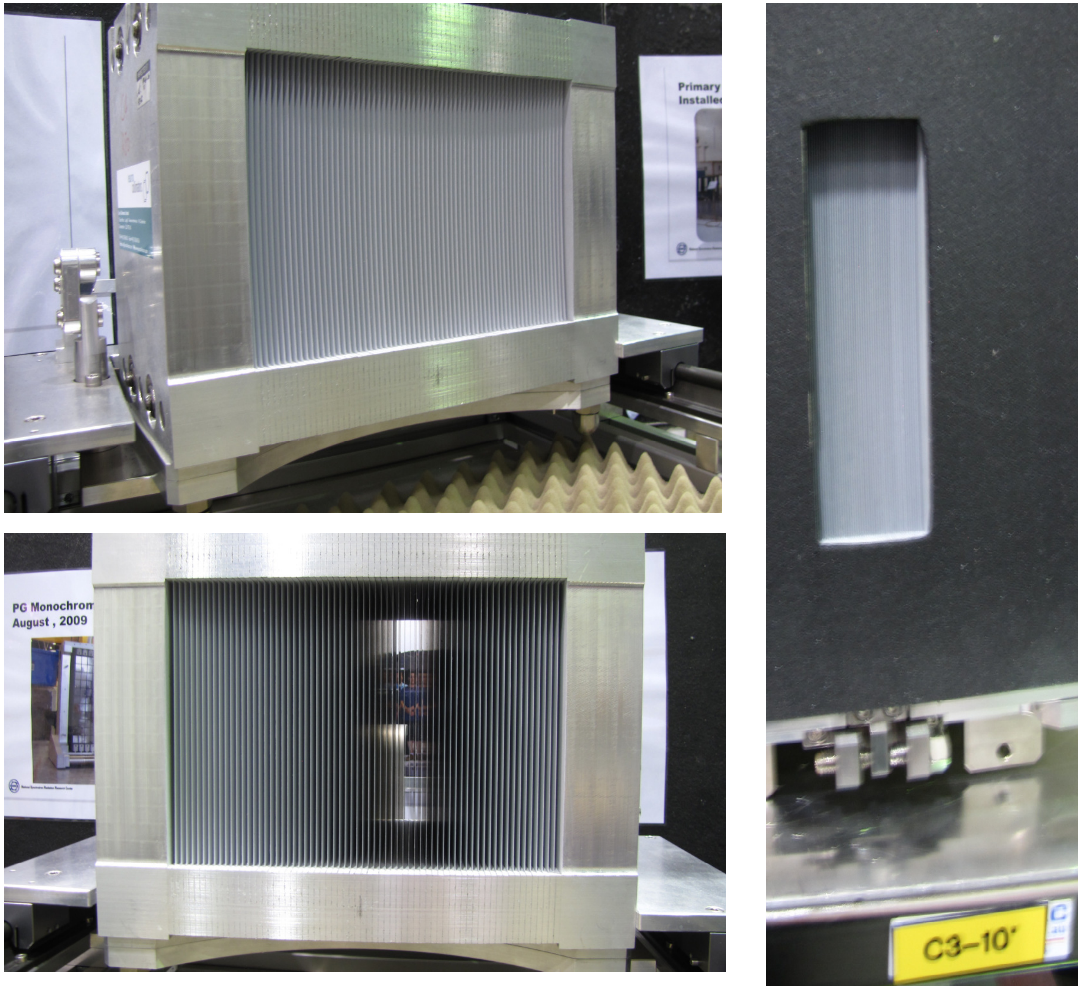
Collimators

- Collimators aim to restrict the divergence of the beam by channelling the beam through a series of tight long slits. The sides of the slits are covered with an absorbing material such as gadolinium oxide .
- Collimators will reduce the intensity of the signal and so a compromise is made between high resolution and intensity.



Suite of SIKA collimators

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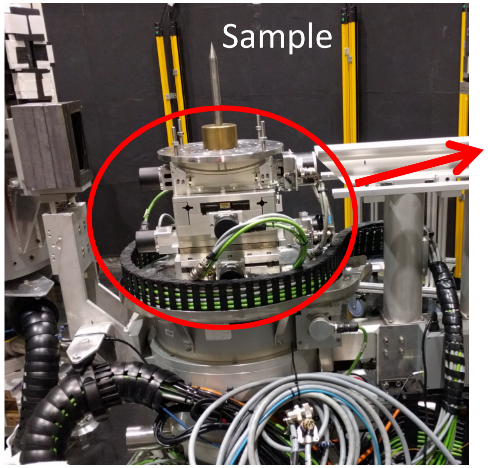
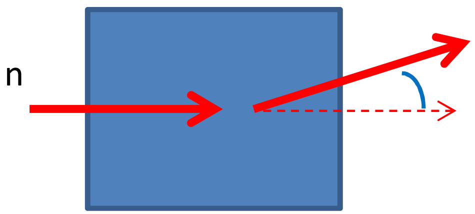
Goniometers

- TAS instruments typically operate with in-plane scattering.
- If the sample is not well aligned within this scattering plane (<1°) then we need to tilt the sample
- Start by aligning two perpendicular vectors from your sample to the goniometer axes

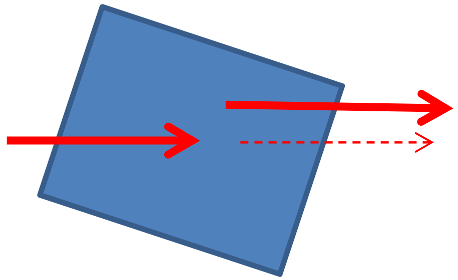


TAIPAN goniometer

- Mutually perpendicular axes



Neutrons scattered out of plane wont be detected



Tilt the sample so that the scattered beam is within the scattering plane

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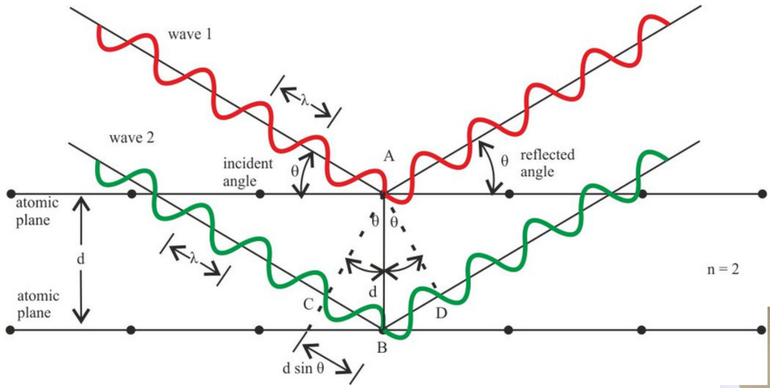
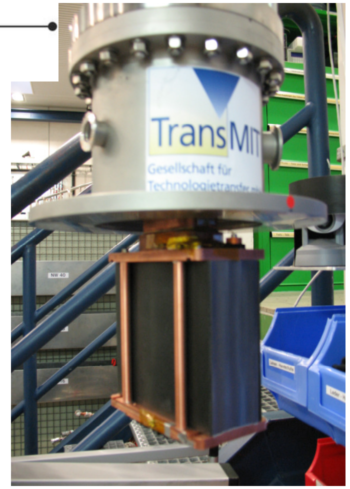
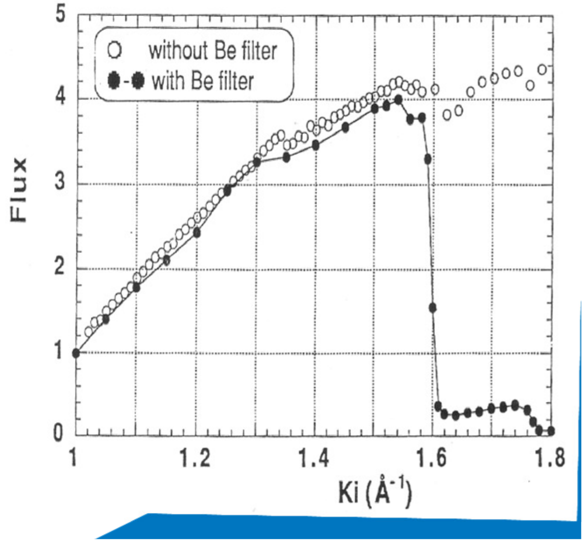
Filters

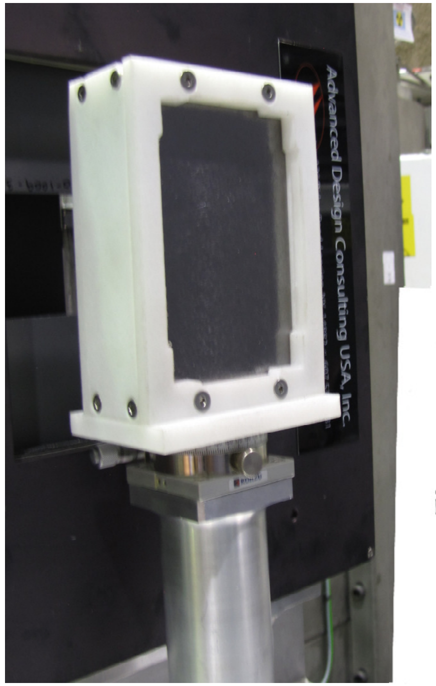
- Be-filter cuts everything above k=1.55 Å-1, but must be cooled below 80K for best efficiency
- Pyrolytic Graphite Filter – has special energies where λ intensity is maximised while λ/2 and λ/3 intensities are minimised.

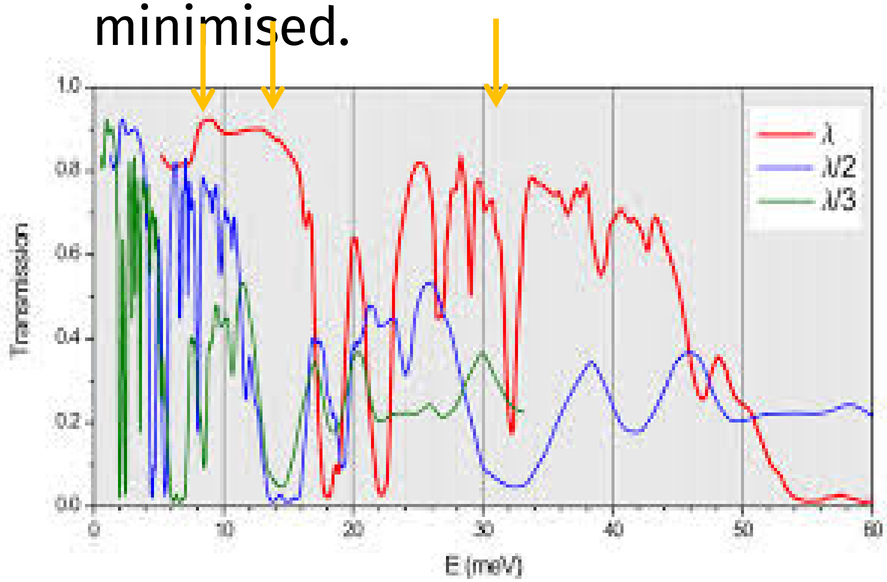




Be-filter and transmission curve



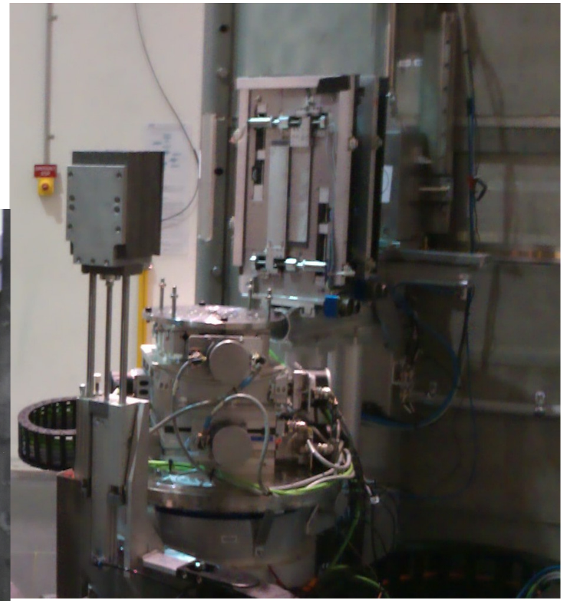




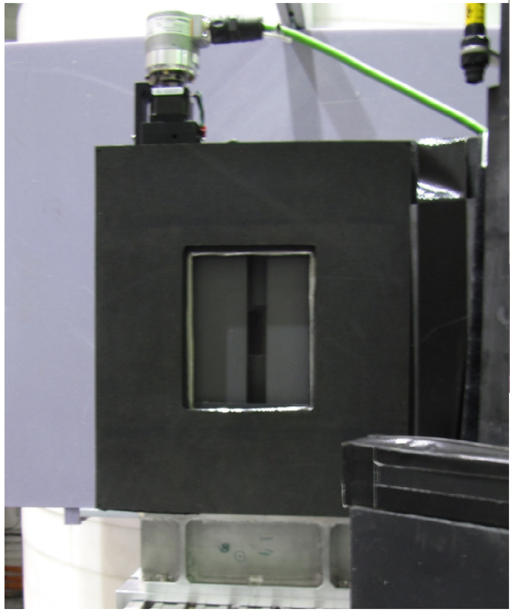
Graphite filter and transmission curve

Apertures

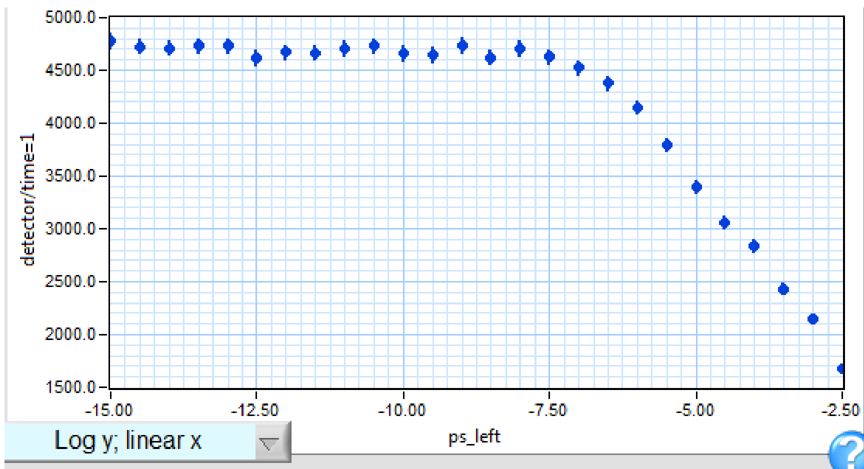
- We use slits to reduce the background neutrons from being scattered by the sample or the sample environment or from entering the detector.
- Careful setting of slits before and after the sample table can improve the signal to noise ratio considerably.
- Slits can be scanned on a Bragg peak



TAIPAN slits



SIKA slits



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TAIPAN

Three different modes:

•Inelastic scattering

- To measure excitations and dynamics in materials
- Energy range:
  - ~5-70meV (PG), ~30-200meV (Cu)
- Energy resolution:
  - ~ 1 meV (PG)

•Inelastic scattering as the Be-filter spectrometer

•Elastic scattering

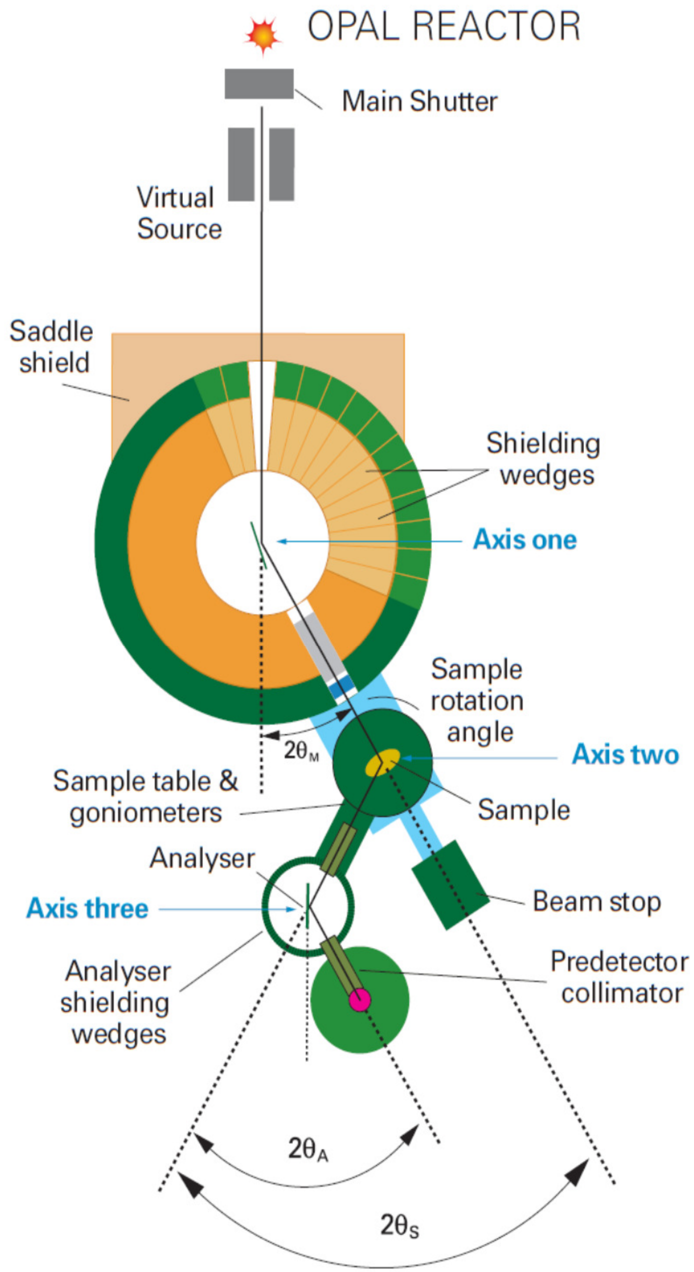
- To measure diffraction when a high signal to noise ratio is required
- Wavelength range:
  - 1-4 Å (PG) 0.6-2.5 Å (Cu)

Types of samples:

•Single crystals

•Powder

•Multi-crystal arrays including thin films



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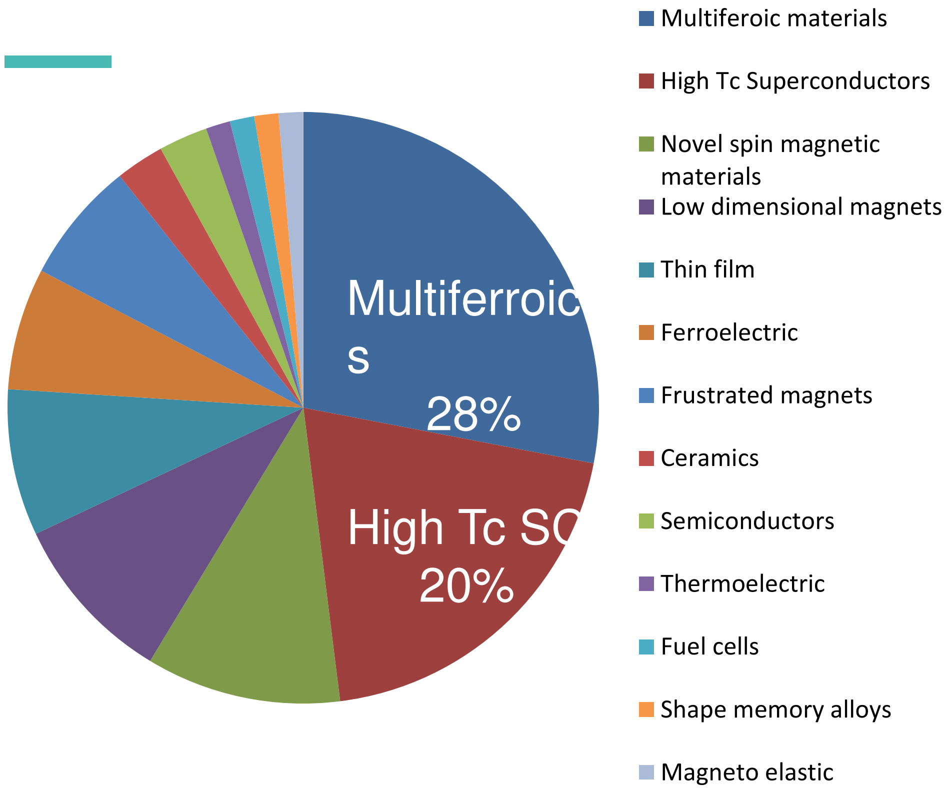
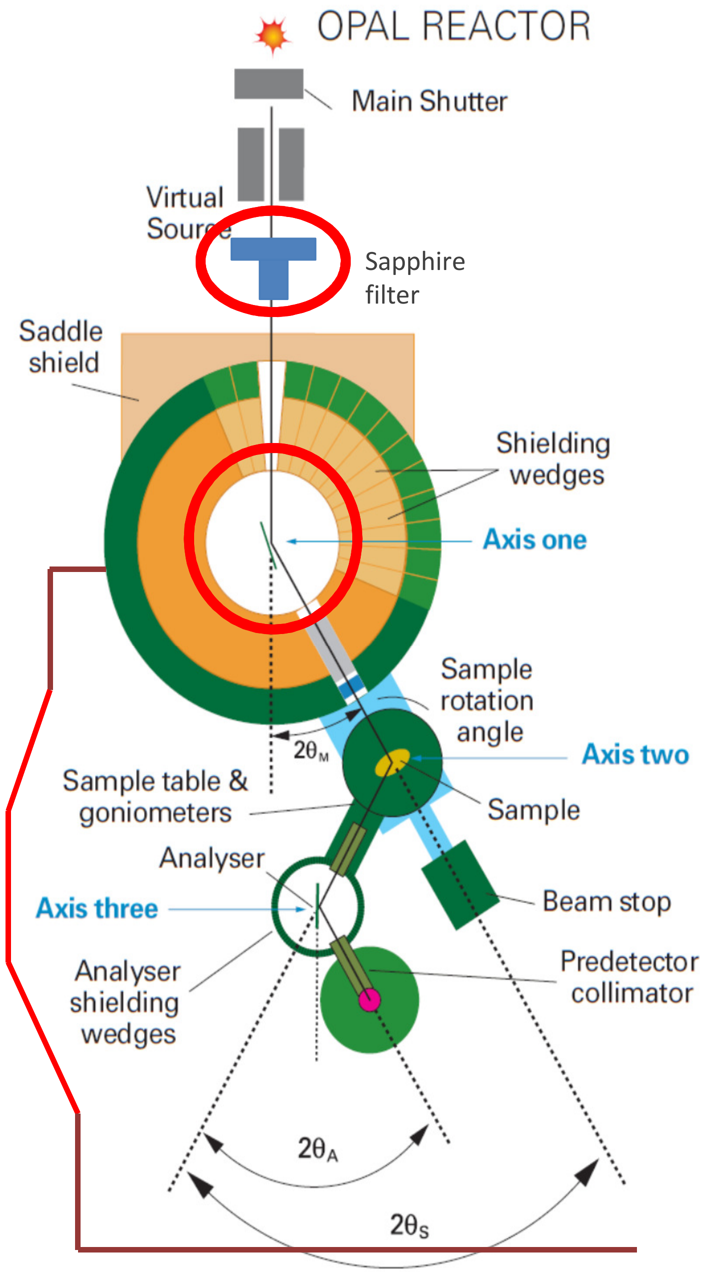
TAIPAN - TAS





- Highly demanded instrument at OPAL
- Subscription rate: 2.8
- Potential for high impact science in high impact journals (Science, Nature etc)
- Fundamental research into the structure and dynamics of materials

SIKA

The differences between SIKA and TAIPAN:

- Energy range:
  - ~2.4 - 27meV (PG)
- Energy resolution:
  - ~0.05 meV
- Wavelength range:
  - 1.6 – 5.4 Å (PG)

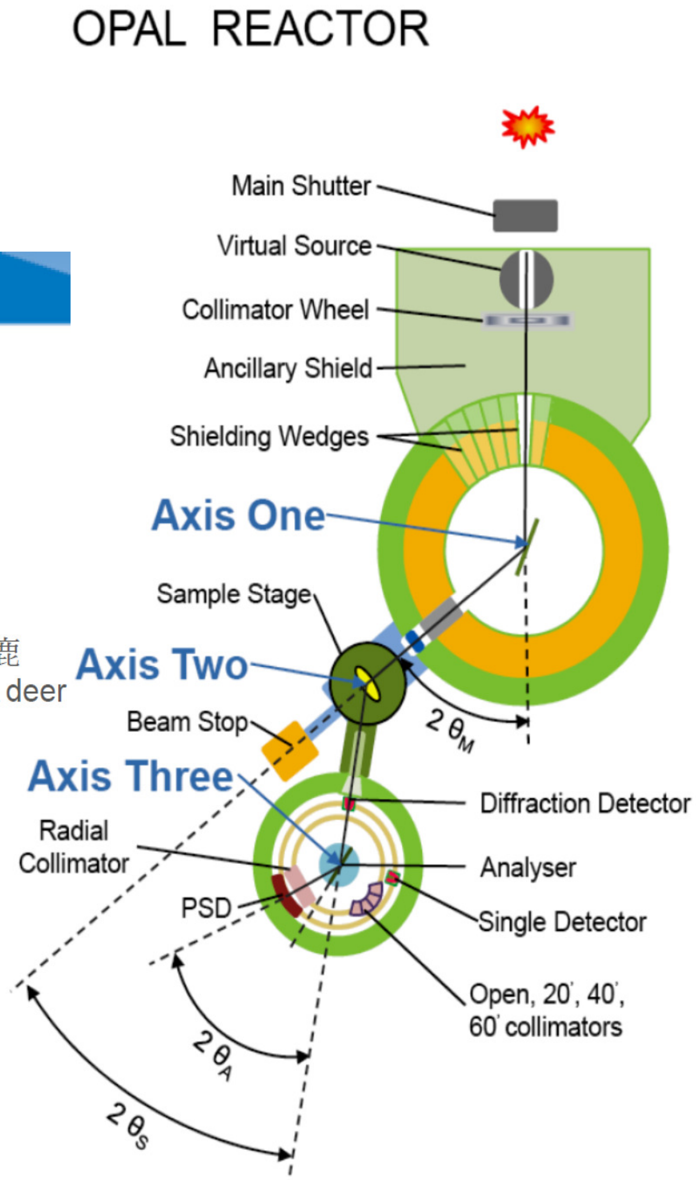
梅花鹿
SIKA deer

Three different detectors:

- Diffraction detector
- Single detector
- Position sensitive detector

Polarisation analysis:

- He-3 spin polarisation system
- Can differentiate between spin-flip and non-spin-flip scattering to reveal magnetic features and novel magnetic structures.



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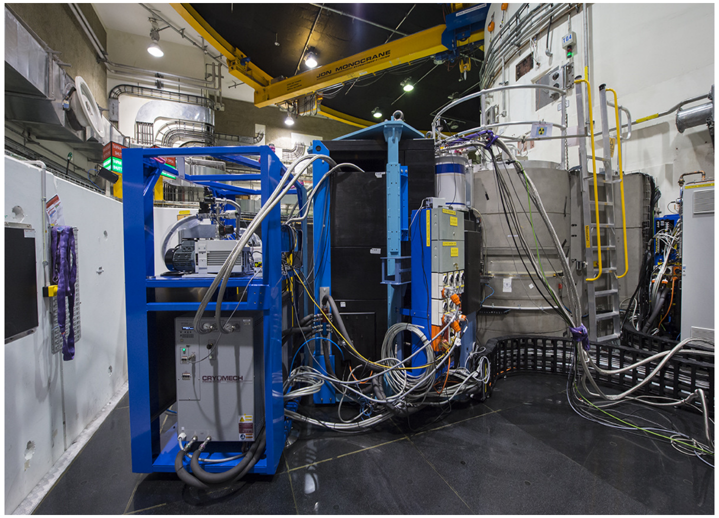
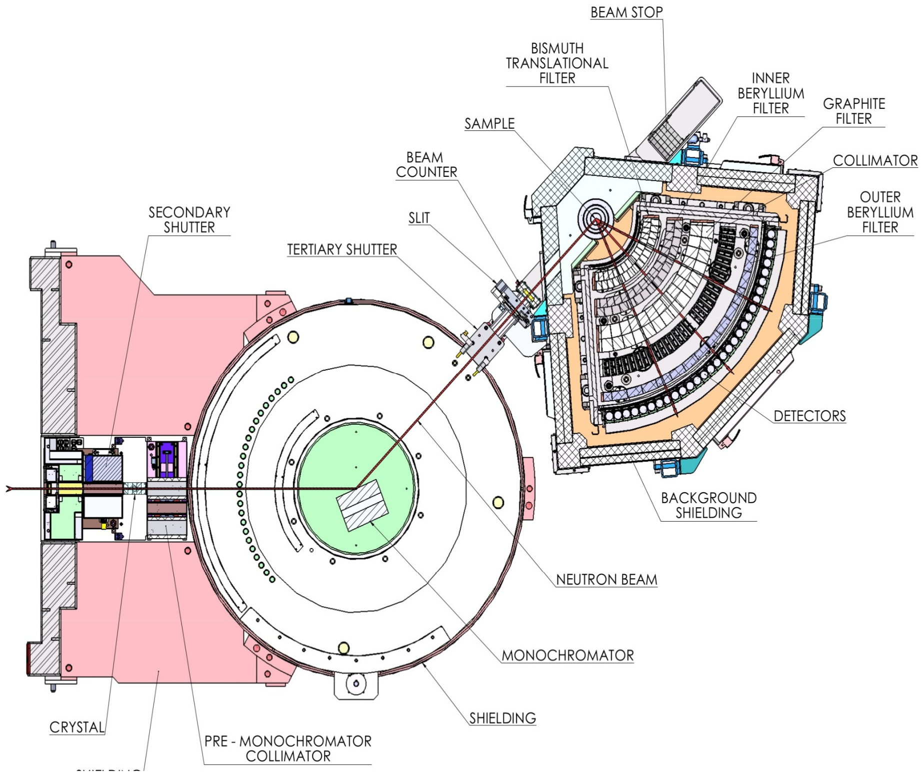
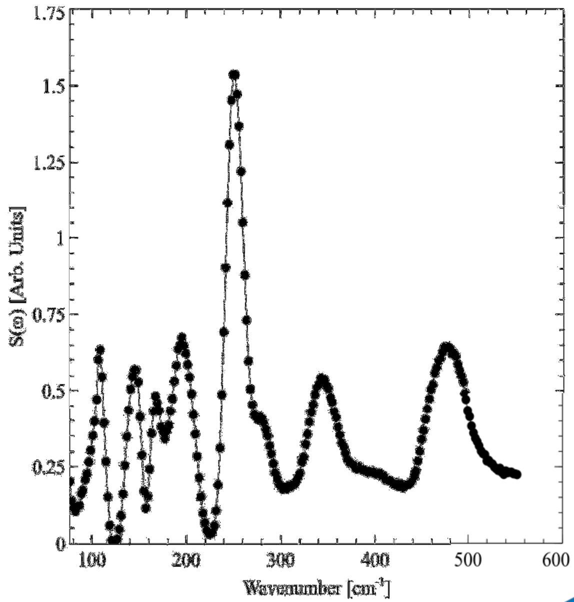
TAIPAN – Be-filter



- Lattice and molecular excitations in complex materials in the form of phonon density of states
- Molecular vibrations as “fingerprints” of surroundings
    - Future energy storage (hydrides)
    - Oil and chemical industry catalysts (zeolites)
    - Nano-crystalline materials for industry
    - Coal studies
    - Nuclear Fuels





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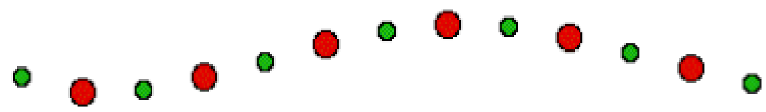
Nuclear (lattice) excitation

Commonly studied excitations

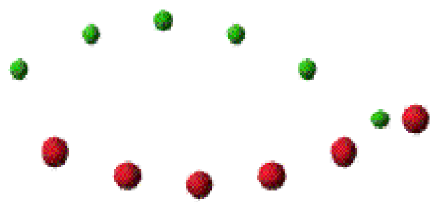
- Phonons
- Vibrations in molecules
- Diffusion
- Collective modes in glasses and liquids



Acoustic phonon

Excitations can tell us about

- Interatomic potentials & bonding
- Phase transitions & critical phenomena (soft modes)
- Fluid dynamics
- Momentum distributions & Superfluids (eg. He)
- Interactions (eg. electron-phonon coupling)



Optical phonon

Inelastic coherent scattering measures *correlated* motions of atoms
Inelastic incoherent scattering measures *self-correlations* e.g. diffusion

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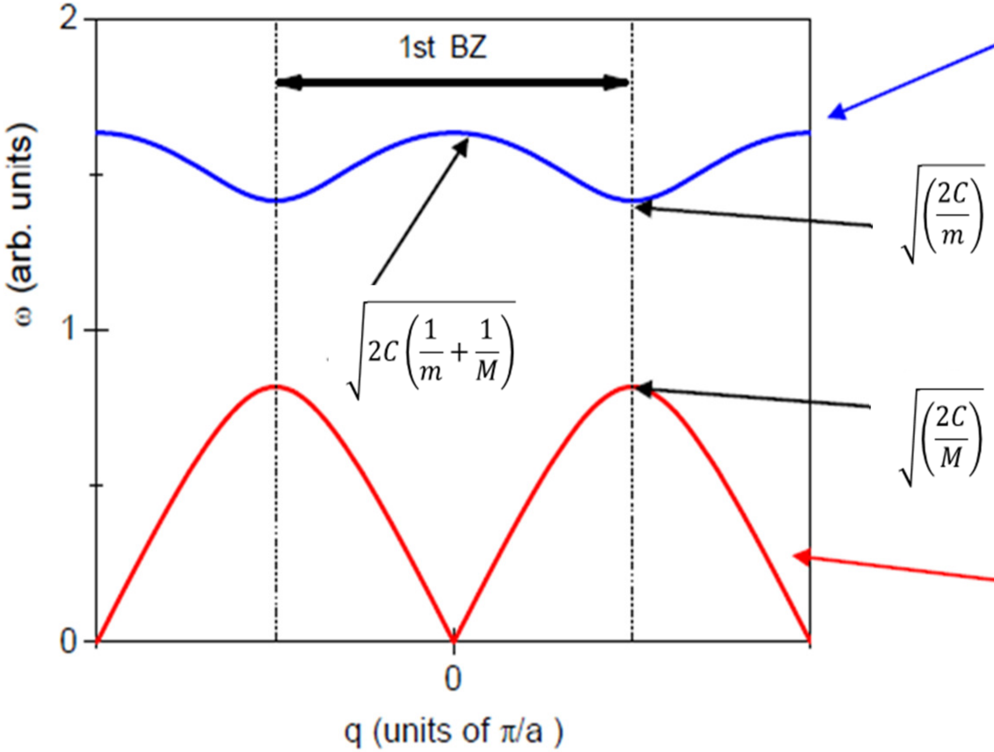
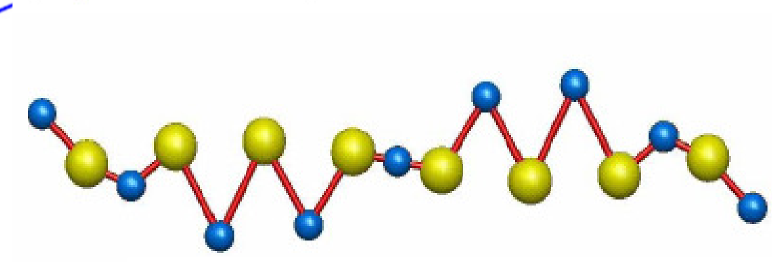
Optical vs Acoustic Phonons

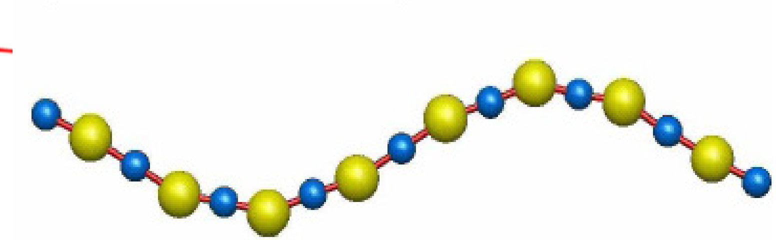


Optic mode



$$\omega_{\pm}^2 = K\left(\frac{1}{m_1}+\frac{1}{m_2}\right) \pm K\sqrt{\left(\frac{1}{m_1}+\frac{1}{m_2}\right)^2 - \frac{4\sin^2(ka/2)}{m_1 m_2}}$$

Acoustic mode



- with N unit cells and n atoms per unit cell there are 3xn branches
- 3 are acoustic, all the rest are optic (3n-3)
- for each branch there are N possible ***q***-values
- the total degrees of freedom is 3xNxn

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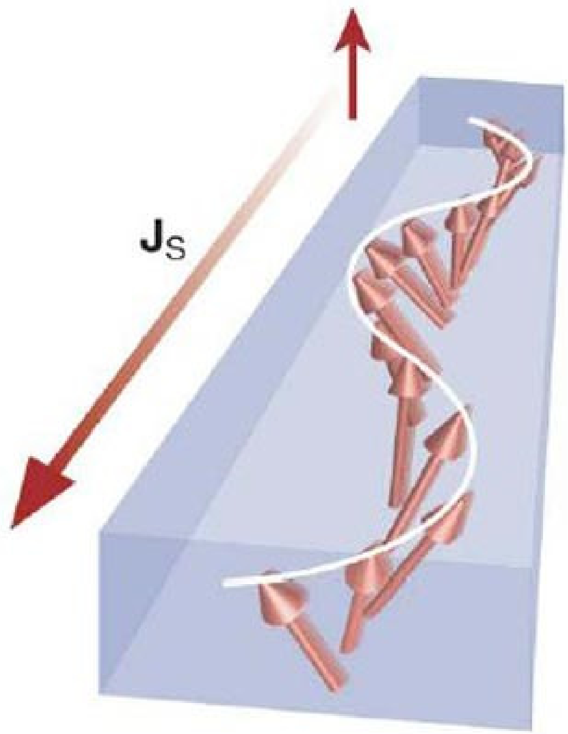
Spin (magnetic) excitations

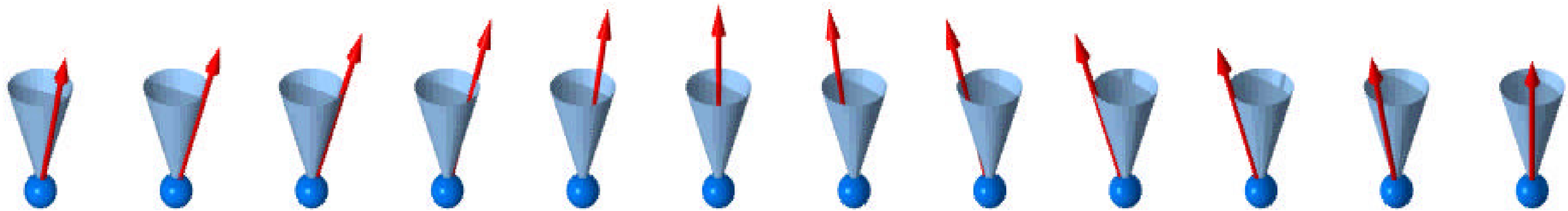
Spin excitations

- Spin waves in ordered magnets
- Paramagnetic and quantum spin fluctuations
- Crystal-field and spin-orbit excitations

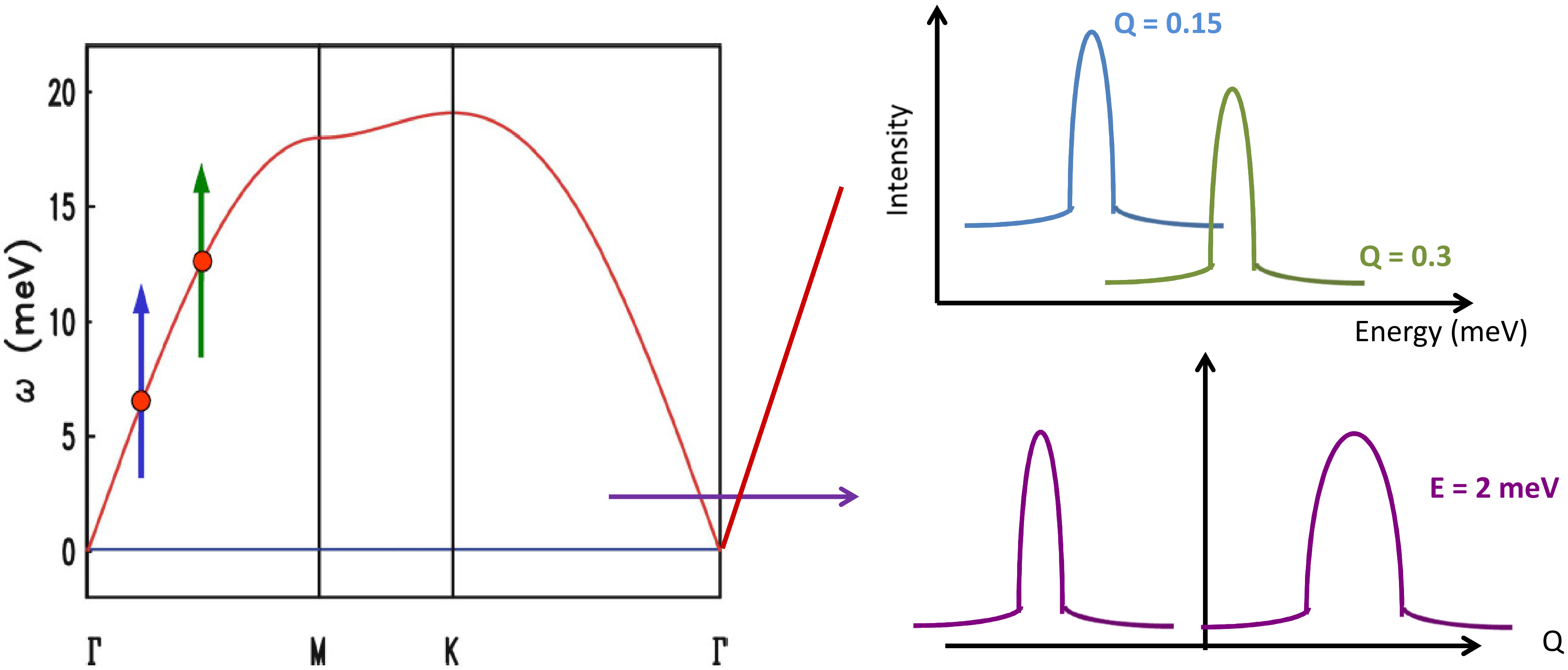


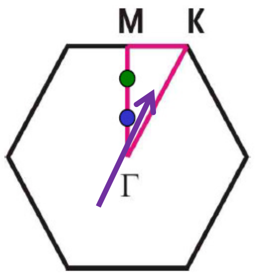
Magnetic inelastic scattering can tell us about

- Exchange interactions
- Single-ion and exchange anisotropy (determine Hamiltonian)
- Phase transitions and critical phenomena
- Quantum critical scaling of magnetic fluctuations
- Other electronic energy scales (eg CF and SO)
- Interactions (eg spin-phonon coupling)



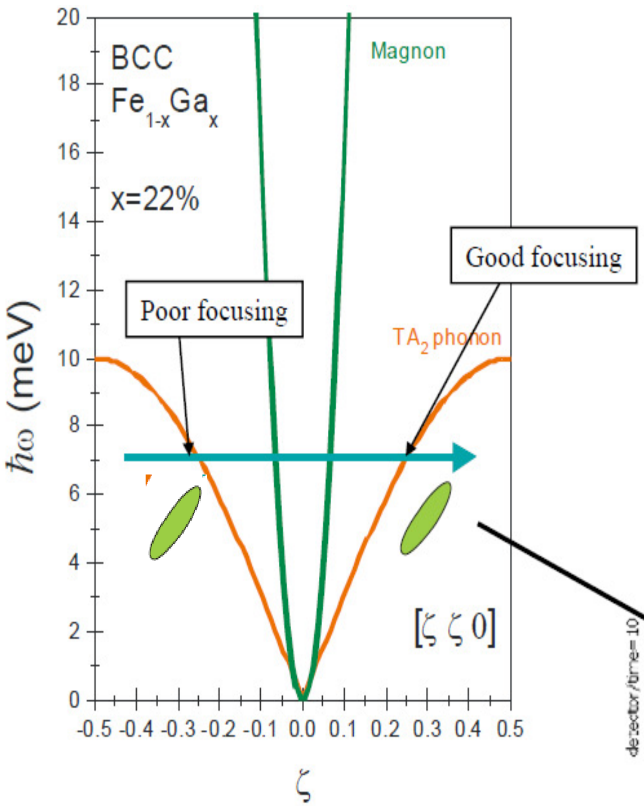
Using neutrons to measure a dispersion

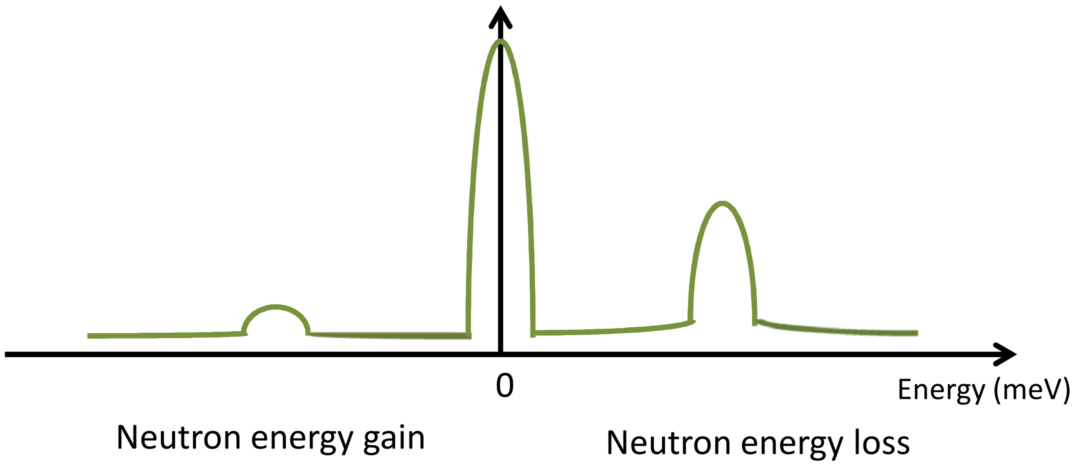


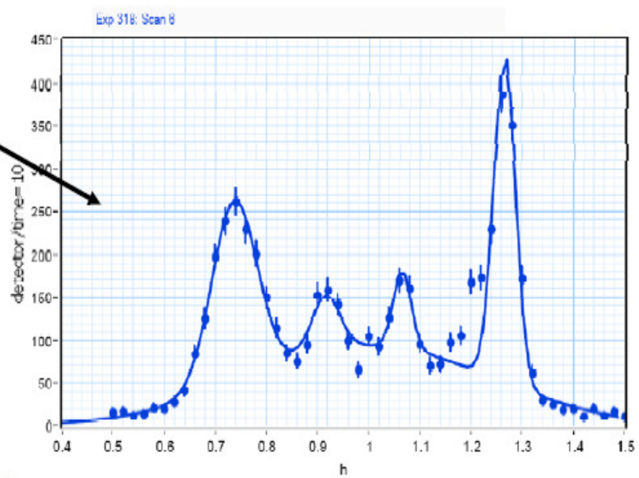


- Γ point = Brillouin zone centre (from which the excitation emerges)
- M and K are different symmetry directions
- Either constant Q, E-scans, or constant E, Q-scans
- Choose fixed Ei or fixed Ef

Focusing and Detailed balance







Detailed balance:

- Neutrons can gain energy (by stopping an excitation)
- Neutrons can lose energy (by exciting a thermally populated vibration)

- Focusing: matching 3D slope of resolution ellipse to dispersion hypersurface

A. Zheludev. ETH, Zurich
http://www.neutron.ethz.ch/people/zhelud

$$S(-\boldsymbol{Q},-\omega) = e^{-\frac{\hbar\omega}{k_B T}} S(+\boldsymbol{Q},+\omega)$$

Resolution focusing vs defocusing

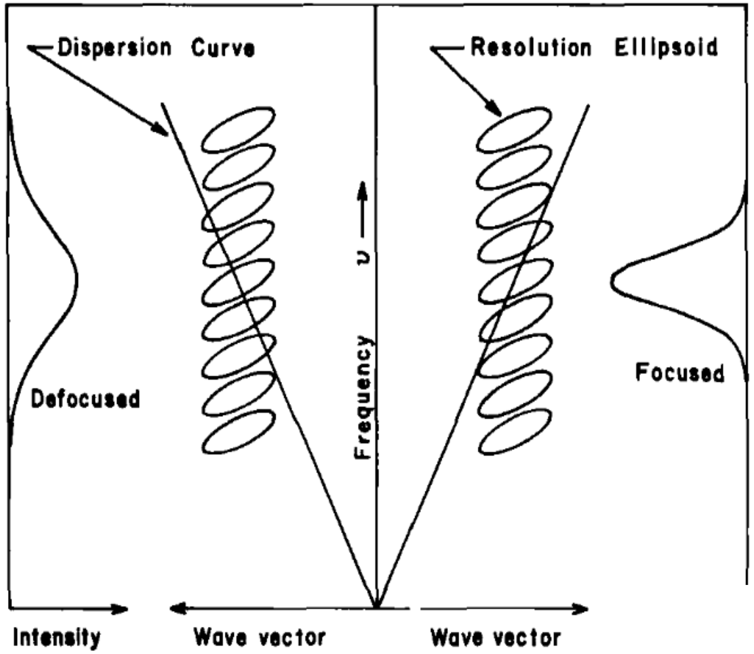


FIG. 11. Schematic representation of focusing effects. (Adapte

- When scanning a feature, the resolution of the features change with respect to the instrument configuration.
- Choose the focused configuration for the more accurate peak determination
- (Area under the curve will be the same)

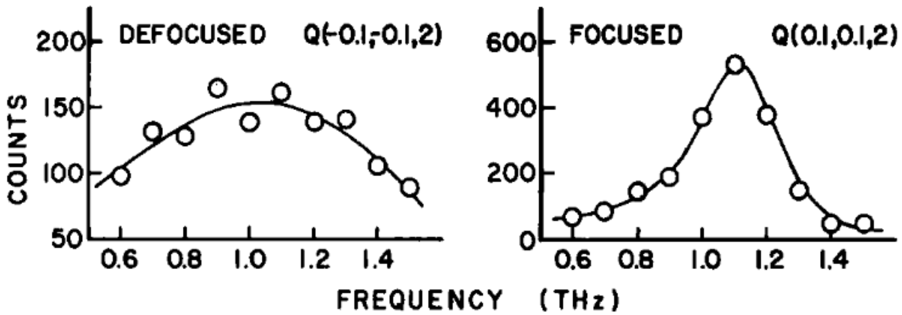
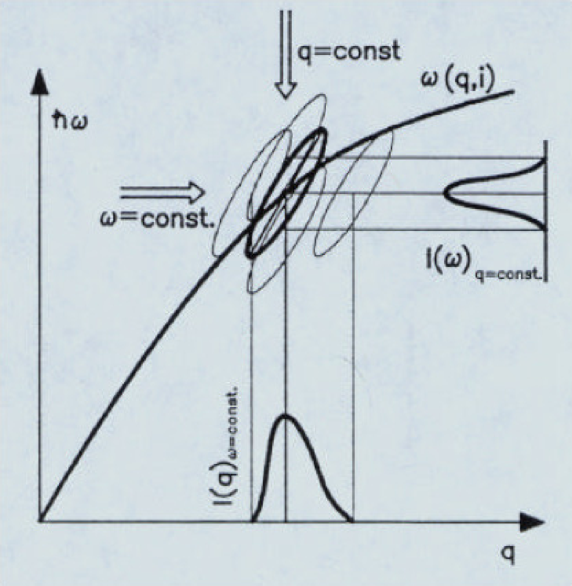


FIG. 12. The neutron groups corresponding to a T[110] low-lying acoustic phonon in Cu with **q** taken, respectively, in the clockwise and counterclockwise sense from the [002] reciprocal lattice point.

Simple rules of intensity

- Phonons have high intensity at large |Q| values and when **Q** is parallel to the mode vibration
- Magnons have high intensity at low |Q| values when **Q** perpendicular to the magnetic moment. This is due to the magnetic form factor which drops quickly with increasing **Q**



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Advantages of TAS

- By optimising all parameters (collimators, slits, Ef, focussing etc) you can focus on the point in reciprocal space that is most interesting – so you can follow dispersion curves along high symmetry directions with optimised resolution and signal to noise.
- The types of scans can be modified and optimised for either sharp (constant E scans) or flat (constant Q) modes.
- By optimising the signal to noise ratio we can measure even very small samples and observe weak scattering.
- Parametric studies (ie evolution with temperature) are relatively quick
- Polarisation analysis is possible to separate magnetic scattering (spin-flip scattering) and phonon signals which do not flip the spin of the neutrons.

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Disadvantages of TAS

- "Needle in a haystack" method
- Technique is slow to cover a large region of energy transfer and reciprocal space – poor for mapping
- 'Spurions' can arise from higher order reflections off the monochromators and analysers.
- Measurements probe very specific sections of energy transfer and reciprocal space and so unexpected excitations can be missed.

- Dangerous energies

- normal:

$$\boldsymbol{q} = \boldsymbol{k}_i - \boldsymbol{k}_f, \quad \hbar\omega = E_i - E_f$$

- spurious:

$$\boldsymbol{q} = 2\boldsymbol{k}_i - \boldsymbol{k}_f, \quad \hbar\omega = 4E_i - E_f$$

- spurious:

$$\boldsymbol{q} = 3\boldsymbol{k}_i - 2\boldsymbol{k}_f, \quad \hbar\omega = 9E_i - 4E_f$$

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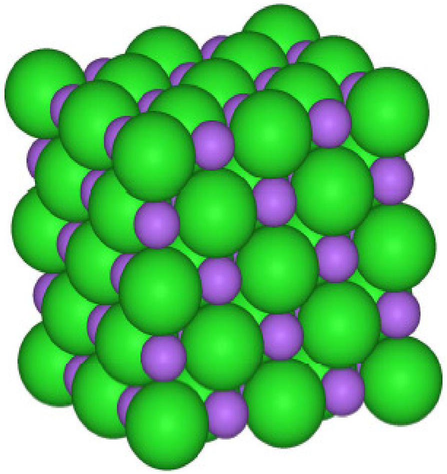
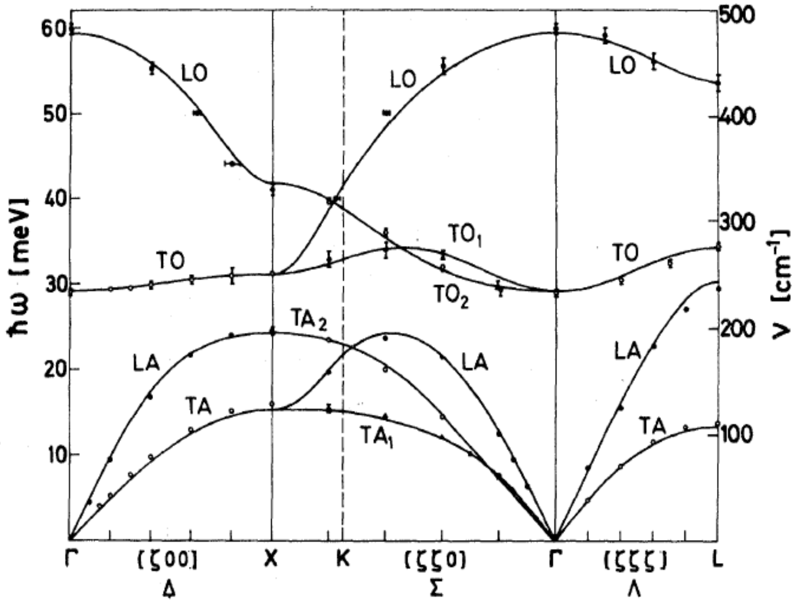
Example:



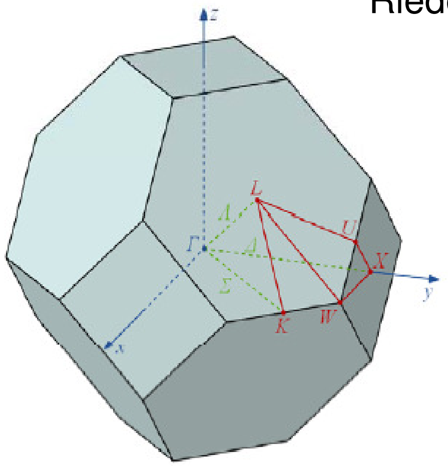
SrO

NaCl type structure
2 atoms per unit cell
-> 6 modes



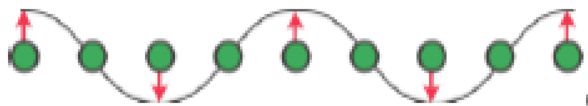
Rieder et al., PRB **12** (1975) 3374

FIG. 1. Phonon-dispersion relations of SrO at 300 °K. Experimental results with estimated errors are given as triangles and circles. The best-fit curves obtained with model I (see Table II) are shown as solid lines.



Longitudinal scan, q || ε

Transverse scan, **q** ⊥ ε



Spin dynamics of $Co_4Nb_2O_9$

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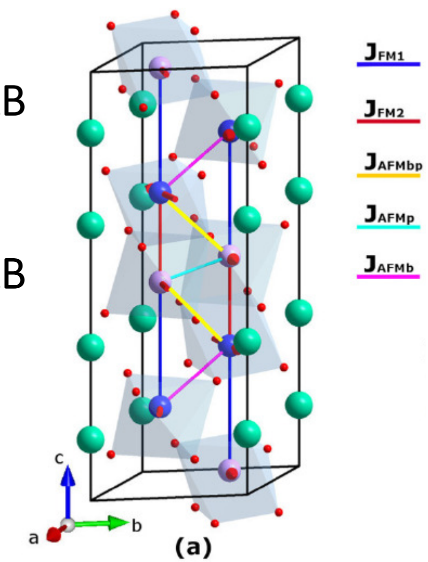
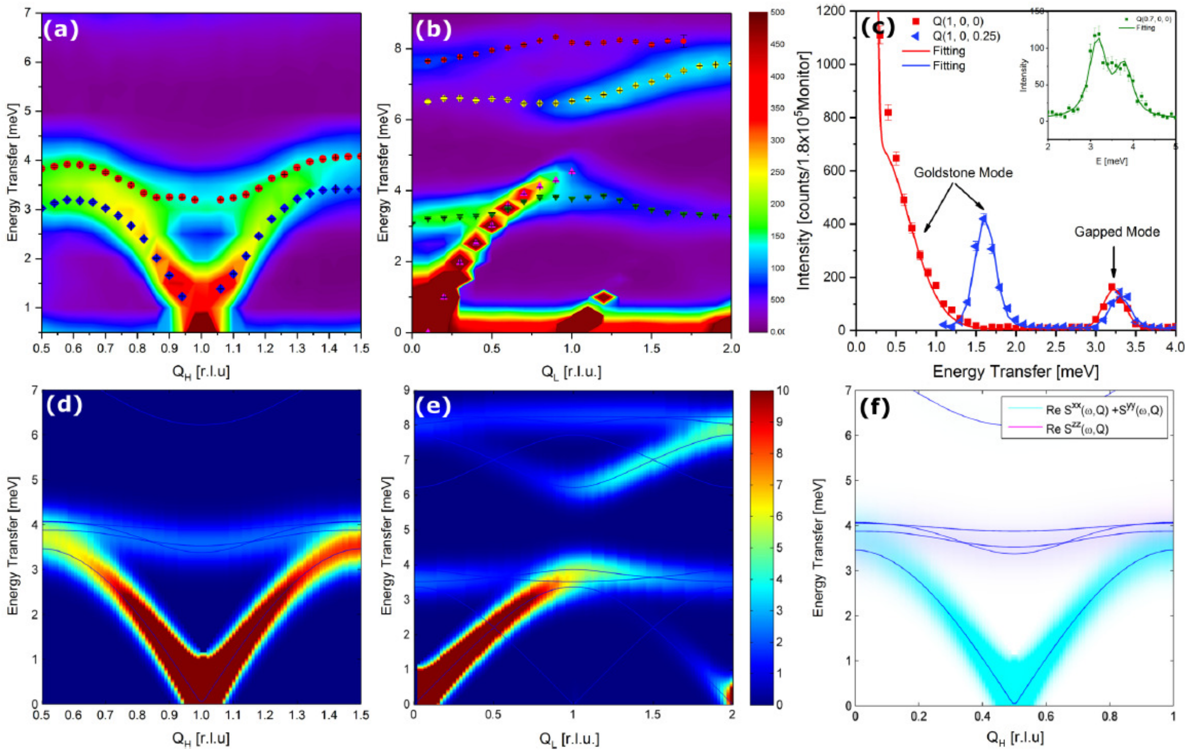
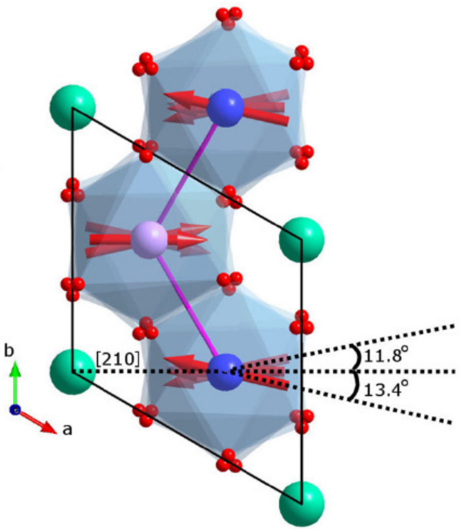
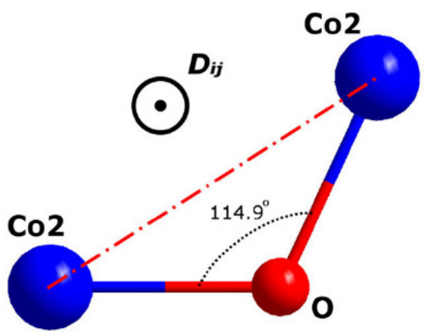
Co1 = 2.32 μB
Tilt = 1.3°

Co1 = 2.52 μB
Tilt = 25.2°









$J_{FM1} = -0.70$ meV, $J_{FM2} = -0.15$ meV,
$J_{FMp} = -0.16$ meV, $J_{AFMb} = 0.42$ meV,
$J_{AFMbp} = 0.52$ meV, $D_1 = D_2 = 1.8$meV

Deng… Rule… et al., PHYSICAL REVIEW B **97**, 085154 (2018)

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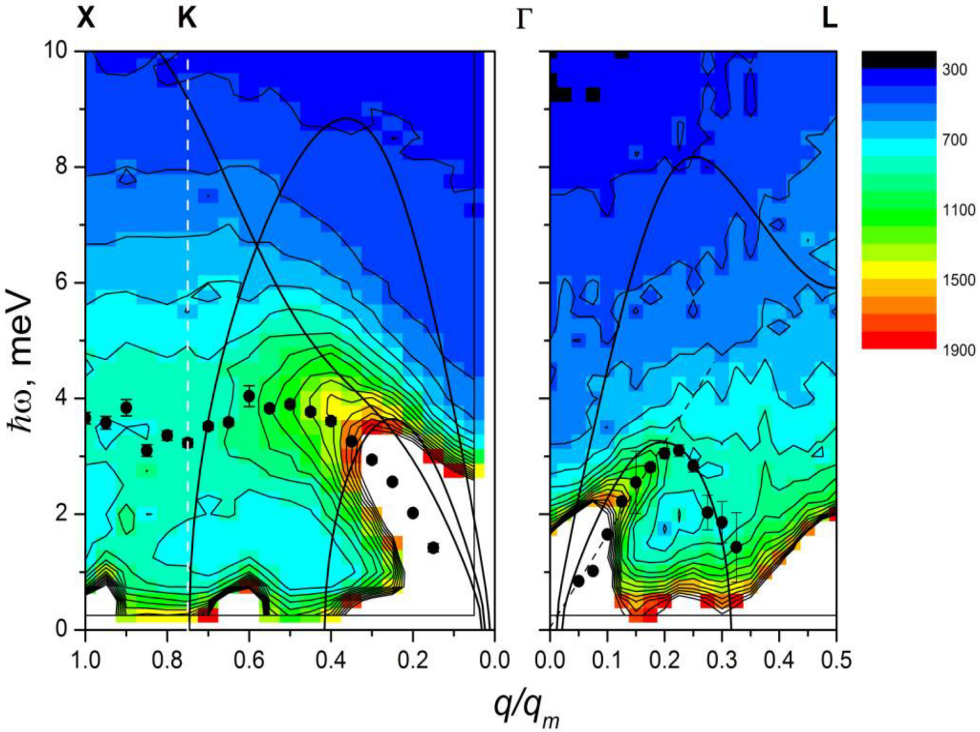
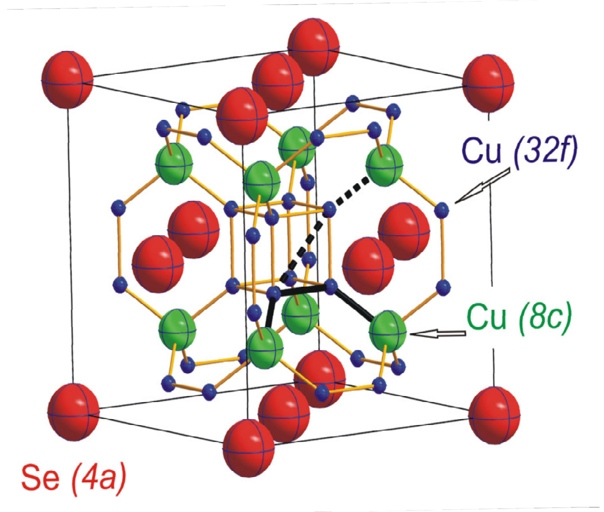
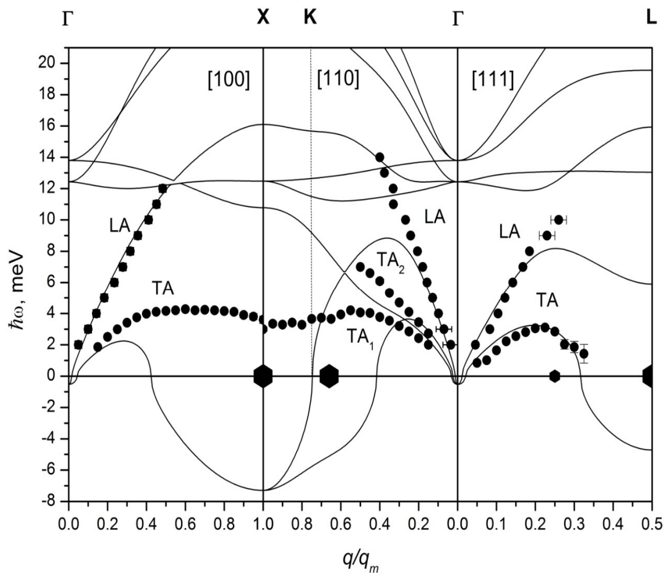
Superionic Conductor $Cu_{1.8}Se$







$Cu_{2-\delta}Se$ - mixed i+e conductor

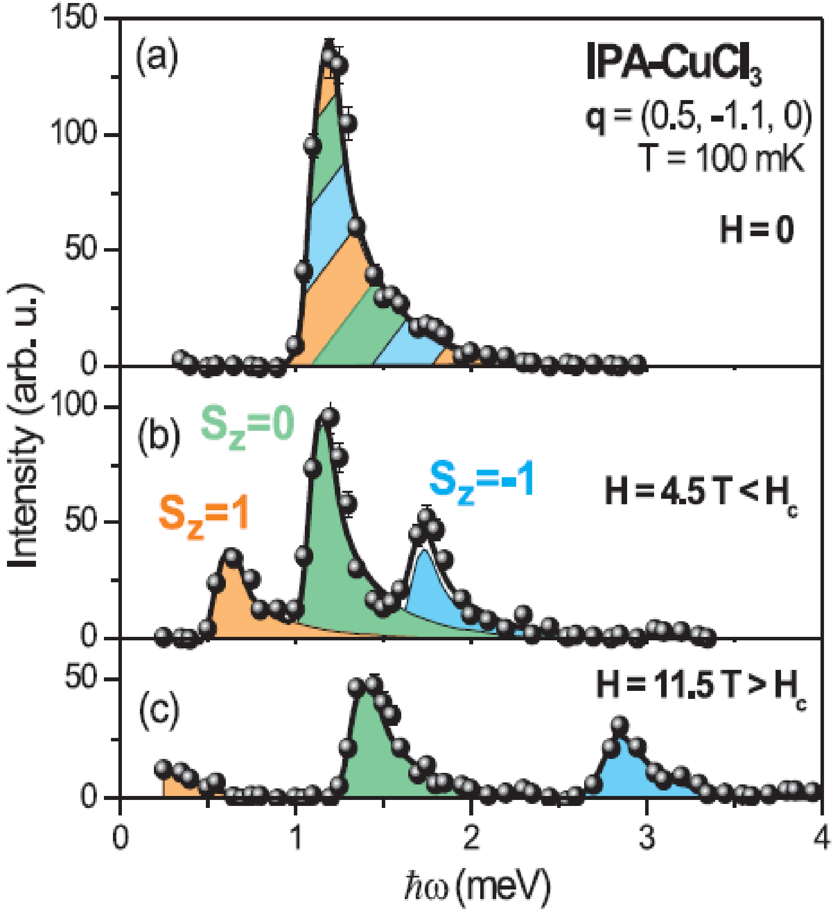
T_S= 414 K for Cu_2Se

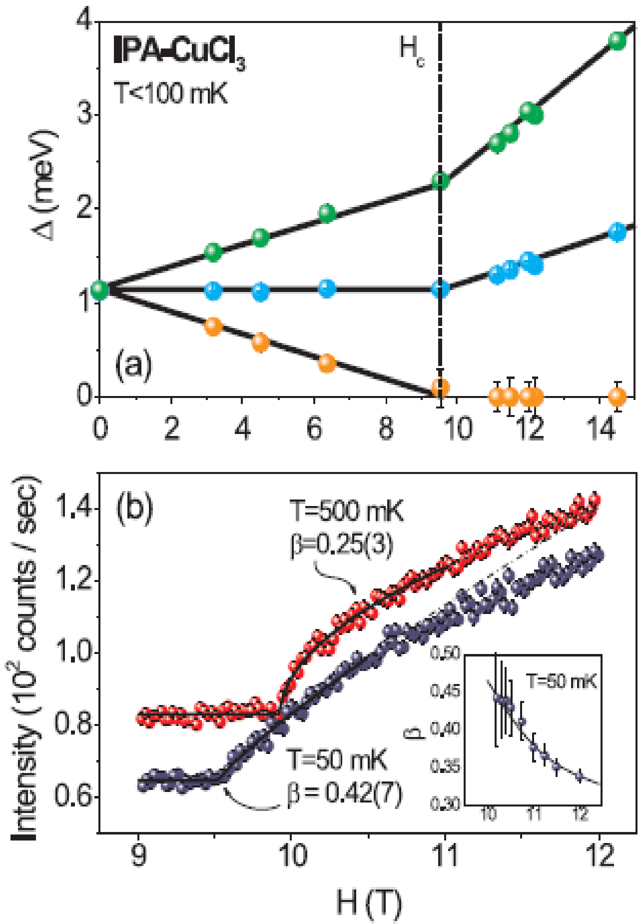
σ_i= 2.4 $\Omega^{-1}cm^{-1}$ @ 670K

High-T phase stable @ RT in the range δ = 0.15 – 0.25

Danilkin et al., J. Phys. Conf. Series **340** 012003 (2012)

Bose Einstein Condensate of Magnons

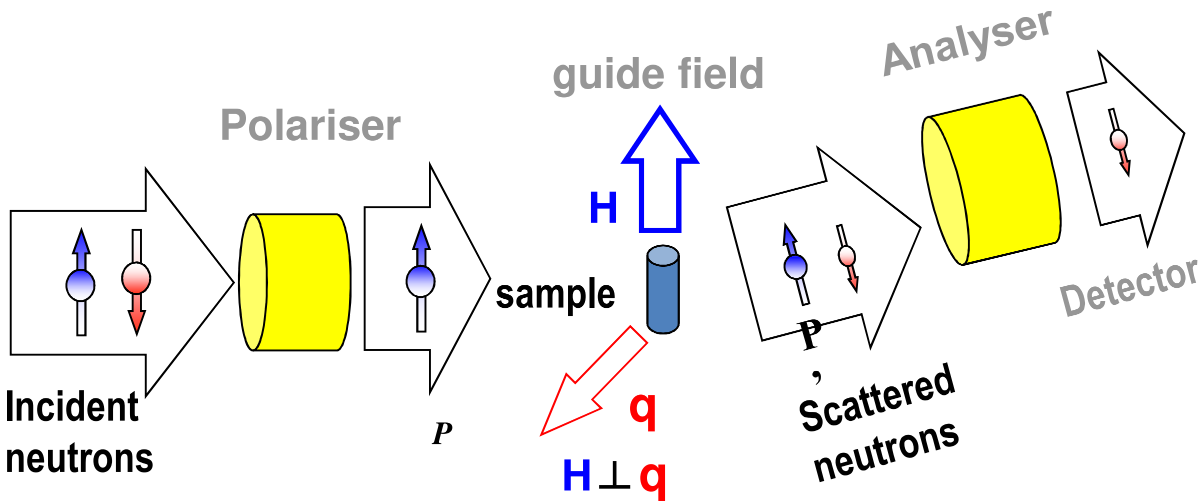


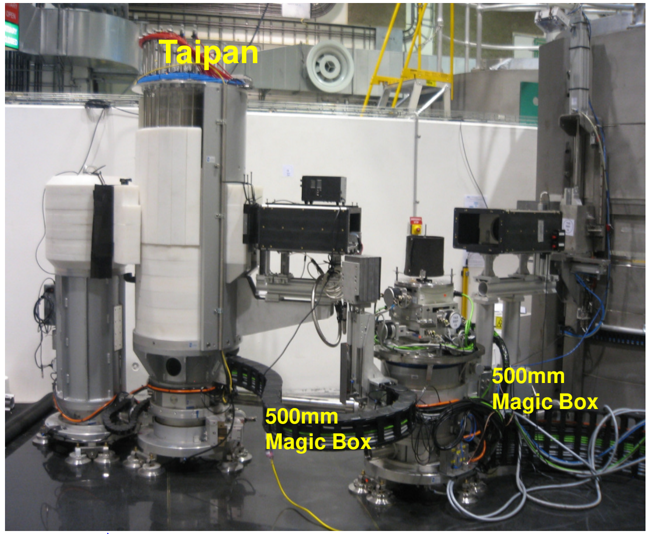


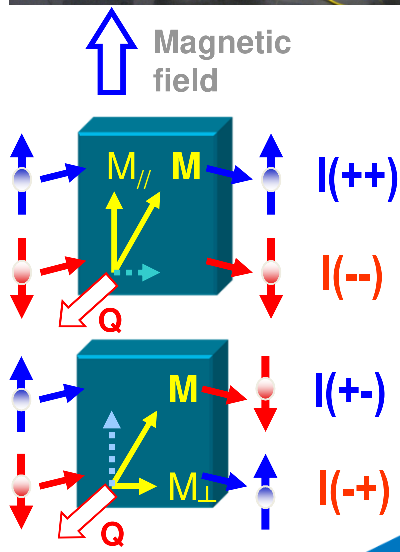
Field dependence of inelastic scattering in the quasi-1D antiferromagnet (AF) IPA-$CuCl_3$ measured at the 1D AF zone center q_1=(1.5,0,0).

Garlea et al., PRL **98** 167202 (2007)

Polarised Neutron Scattering – The basic principle







Nuclear scattering (coherent or isotope-incoherent): $P' = P$

Non-spin-flip scattering

Both magnetic and nuclear scattering: I(++) – I(--) ∝ $M_{//}$ (q)

Spin-flip scattering

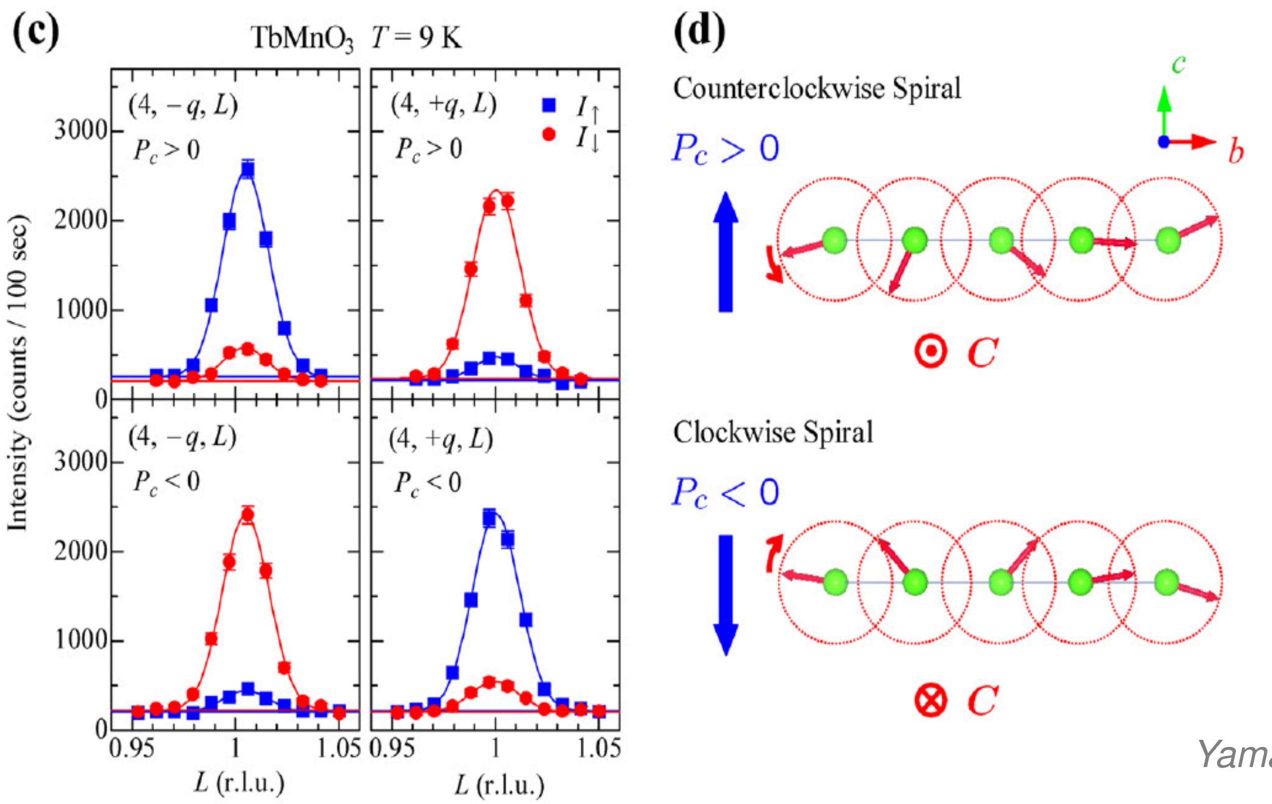
Contains only magnetic scattering: I(+-), I(-+) ∝ |M⊥(q)|.

Special case I(+-)=I(-+), Spiral magnetic structure I(+-)≠I(-+)

Spin-incoherent scattering: $P' = -\frac{1}{3} P$

Suppress 2/3 of the background from Hydrogen

Magnetic ordering in $TbMnO_3$



control
spins
with
applied voltages

Yamasaki et al., PRL 98 (2007) 147204.

Essential: Neutron Diffraction
with polarization analysis was used to determine the
helicity of the magnetic spiral

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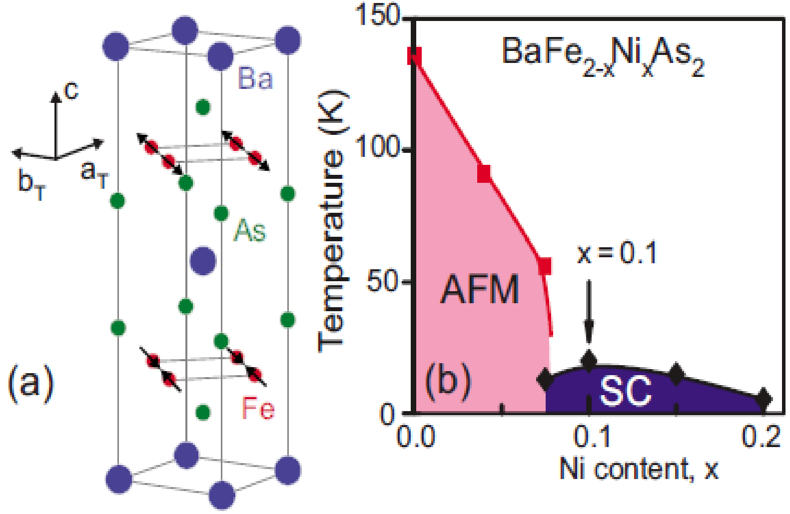
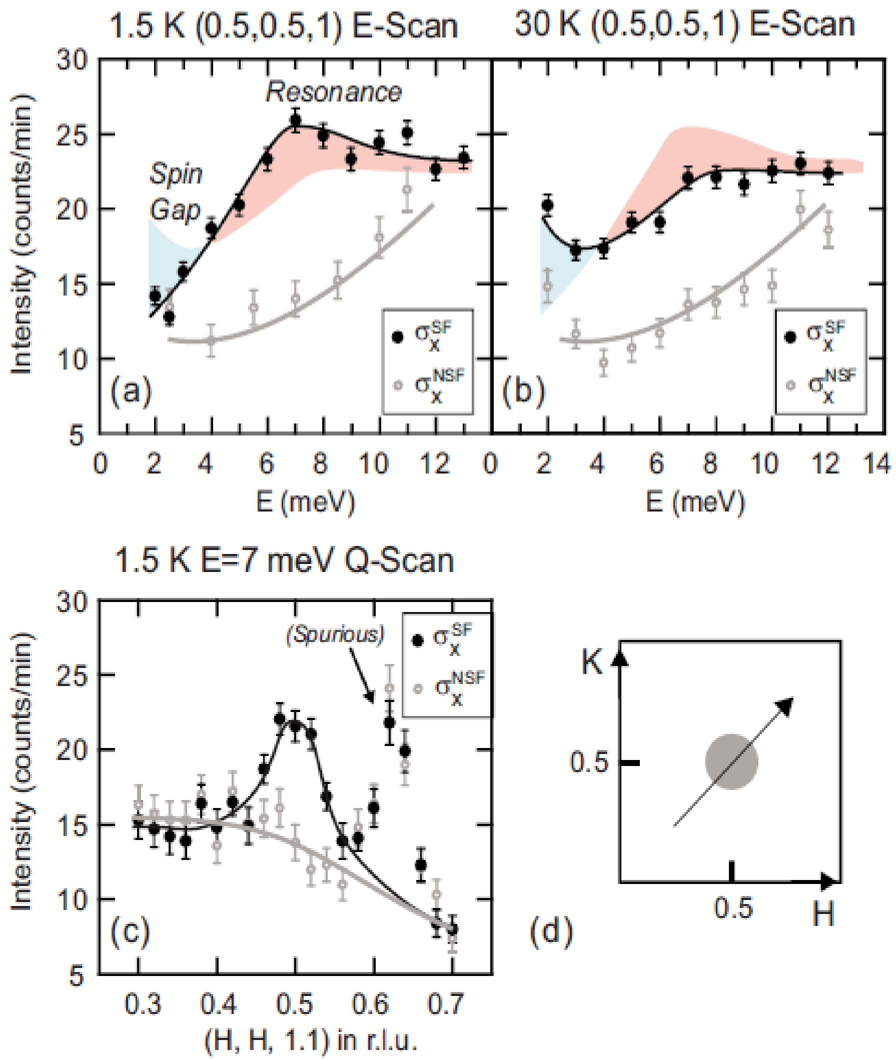
Polarised Inelastic Neutrons





Polarized inelastic neutron scattering was used to show that the neutron spin resonance below *Tc* in superconducting $BaFe_{1.9}Ni_{0.1}As_2$ (*Tc* = 20 K) is purely magnetic in origin.

Lipscombe et al., PRB **82** 064515 (2010)

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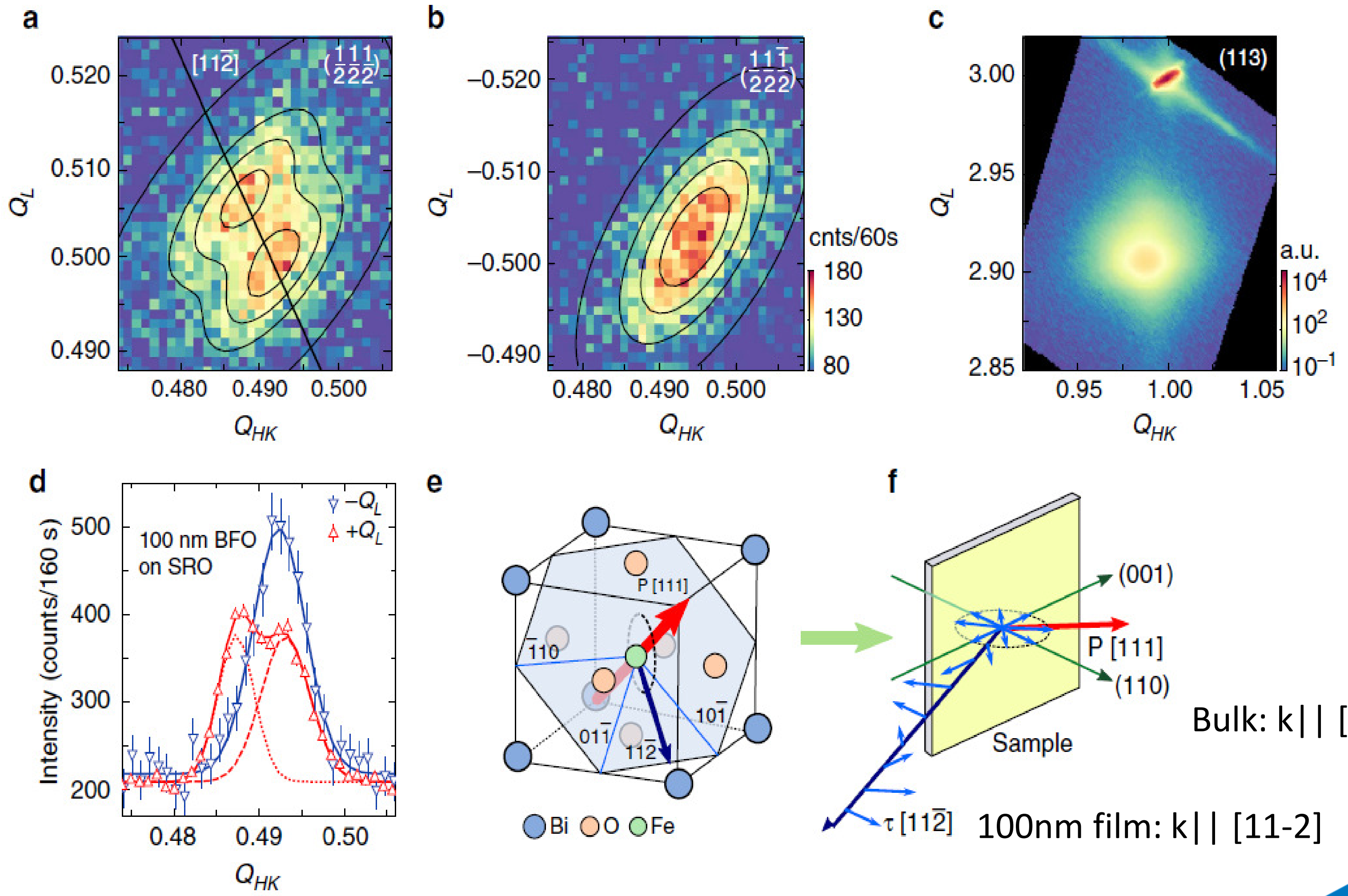
Spin cycloid in a 100nm $BiFeO_3$ film



Bulk: k|| [110]

100nm film: k|| [11-2]

NATURE COMMUNICATIONS | 7:12664 | DOI: 10.1038/ncomms12664

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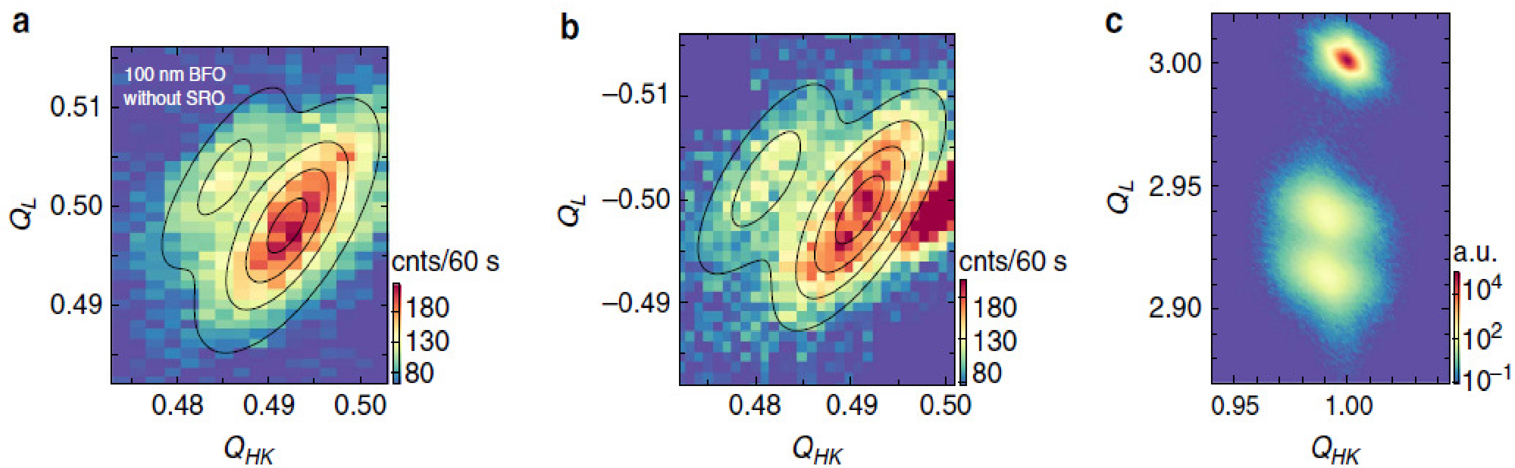

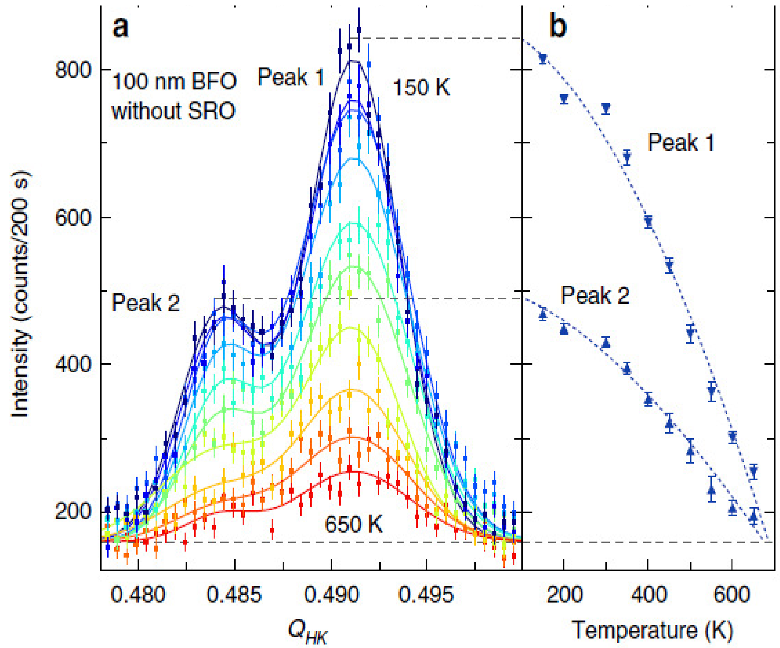

<u>ICM magnetic satellite peaks</u>

Cycloid propagation length = 64.7 nm, which is consistent with the bulk value.

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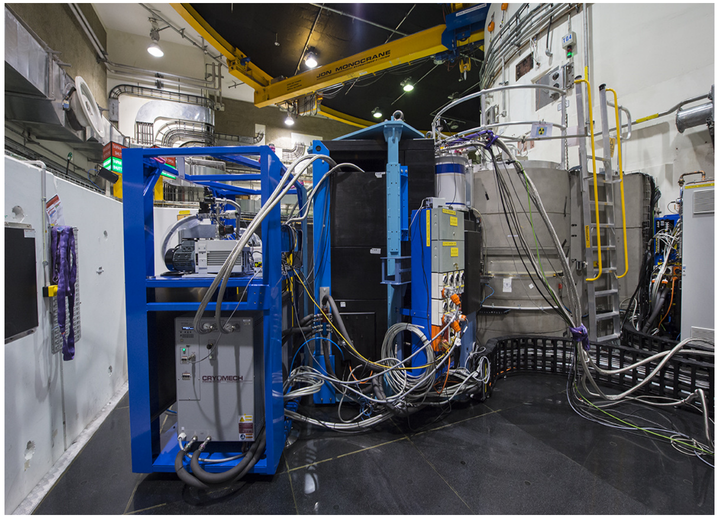
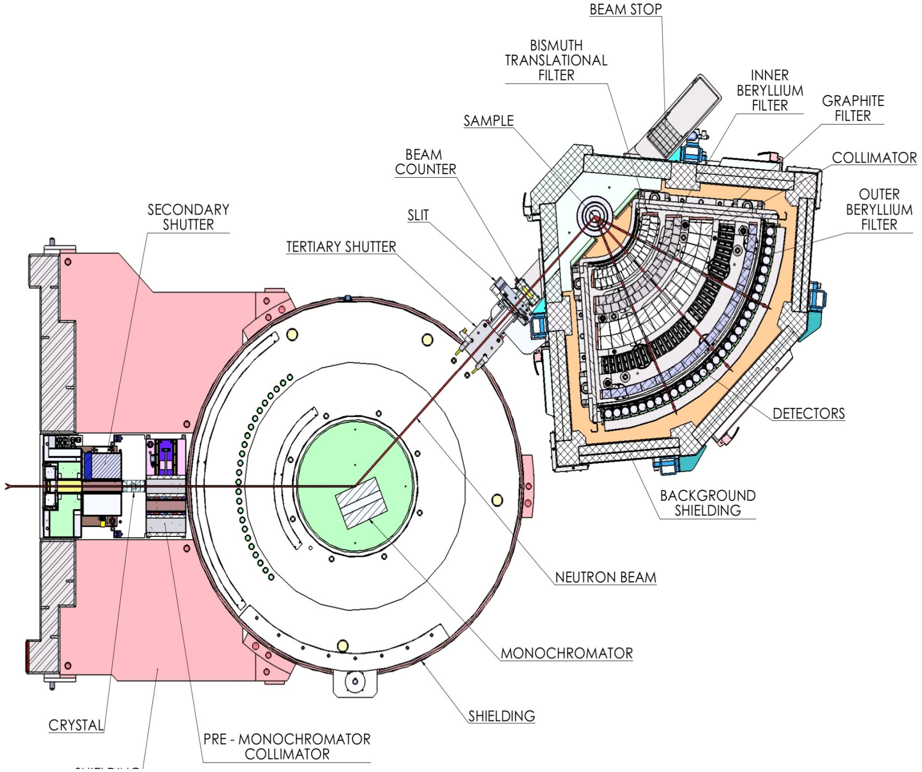
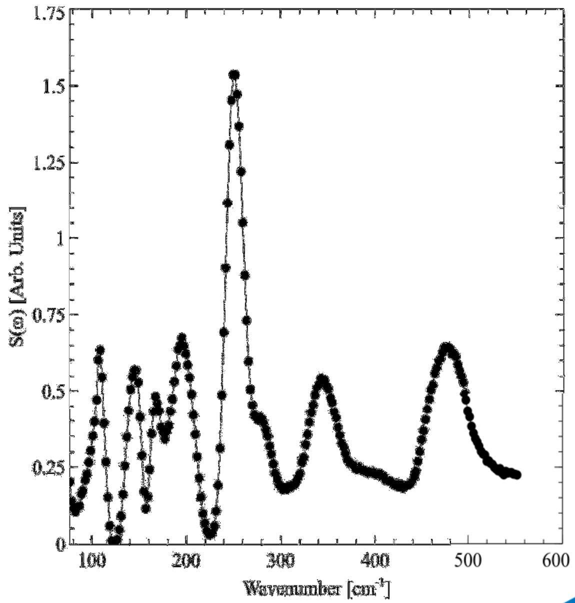
NATURE COMMUNICATIONS | 7:12664 | DOI: 10.1038/ncomms12664

TAIPAN – Be-filter



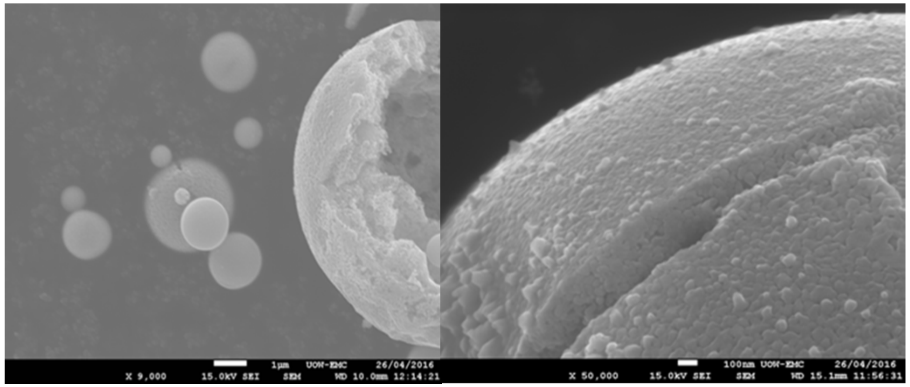
- Lattice and molecular excitations in complex materials in the form of phonon density of states
- Molecular vibrations as "fingerprints" of surroundings
  - Future energy storage (hydrides)
  - Oil and chemical industry catalysts (zeolites)
  - Nano-crystalline materials for industry
  - Coal studies
  - Nuclear Fuels





Ag doped nanoparticles of $LaMnO_3$

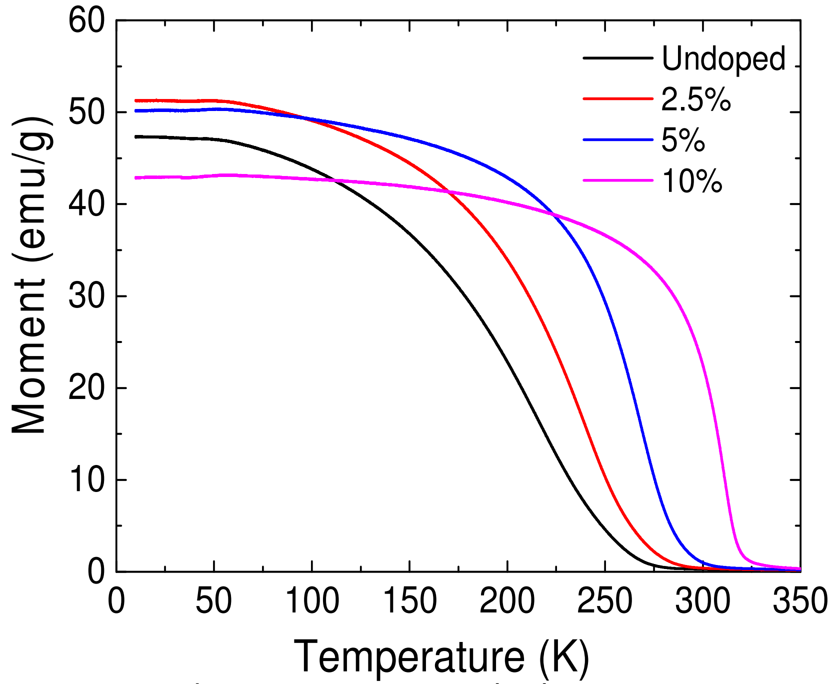
TAIPAN & PELCIAN



Hollow sphere aggregates made via the spray pyrolysis method – diameters ~ 70nm

Desired Ag Concentration	EDS Result
2.5%	2.65%
5%	4.78%
10%	10.5%

- What role does the Ag play in: Magnetism, stoichiometry, Jahn-Teller distortions and phonon propagation?
- How does this compare to the bulk?



Ag doping gave an O-rich phase

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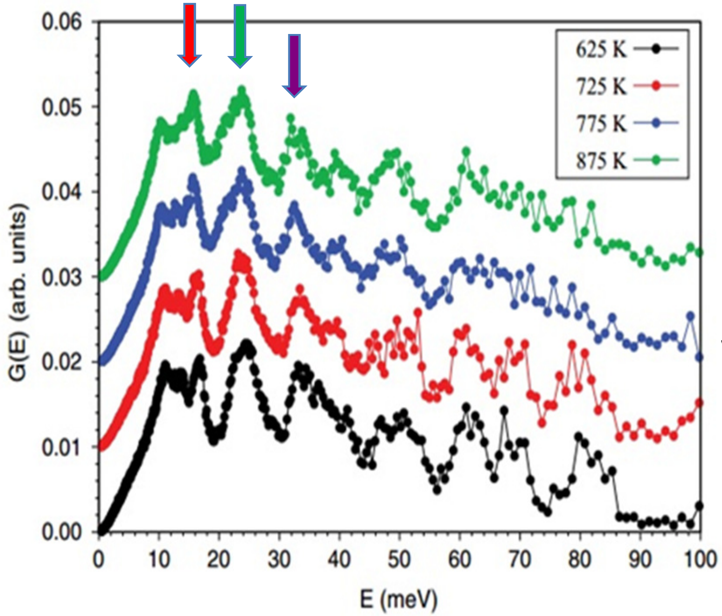
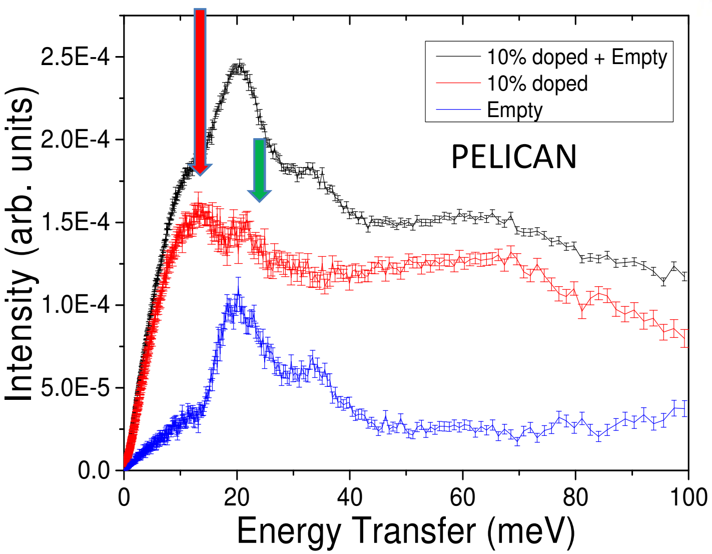
Dynamic properties – $LaMnO_3$

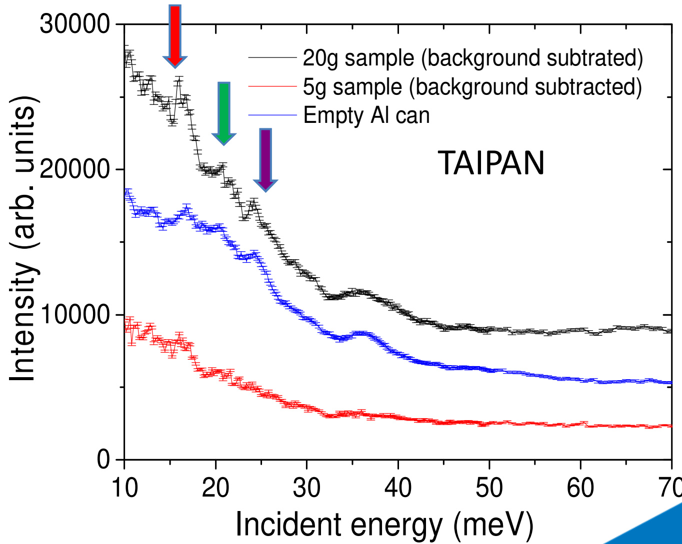
TAIPAN & PELCIAN



- ToF neutron energy loss data
- Bulk, stoichiometric $LaMnO_3$
- ~750K = Jahn-Teller transition

Wdowik et al., PRB **86**, 174305 (2012)





10% Ag sample
300K

Westlake, Rule, et al., In preparation

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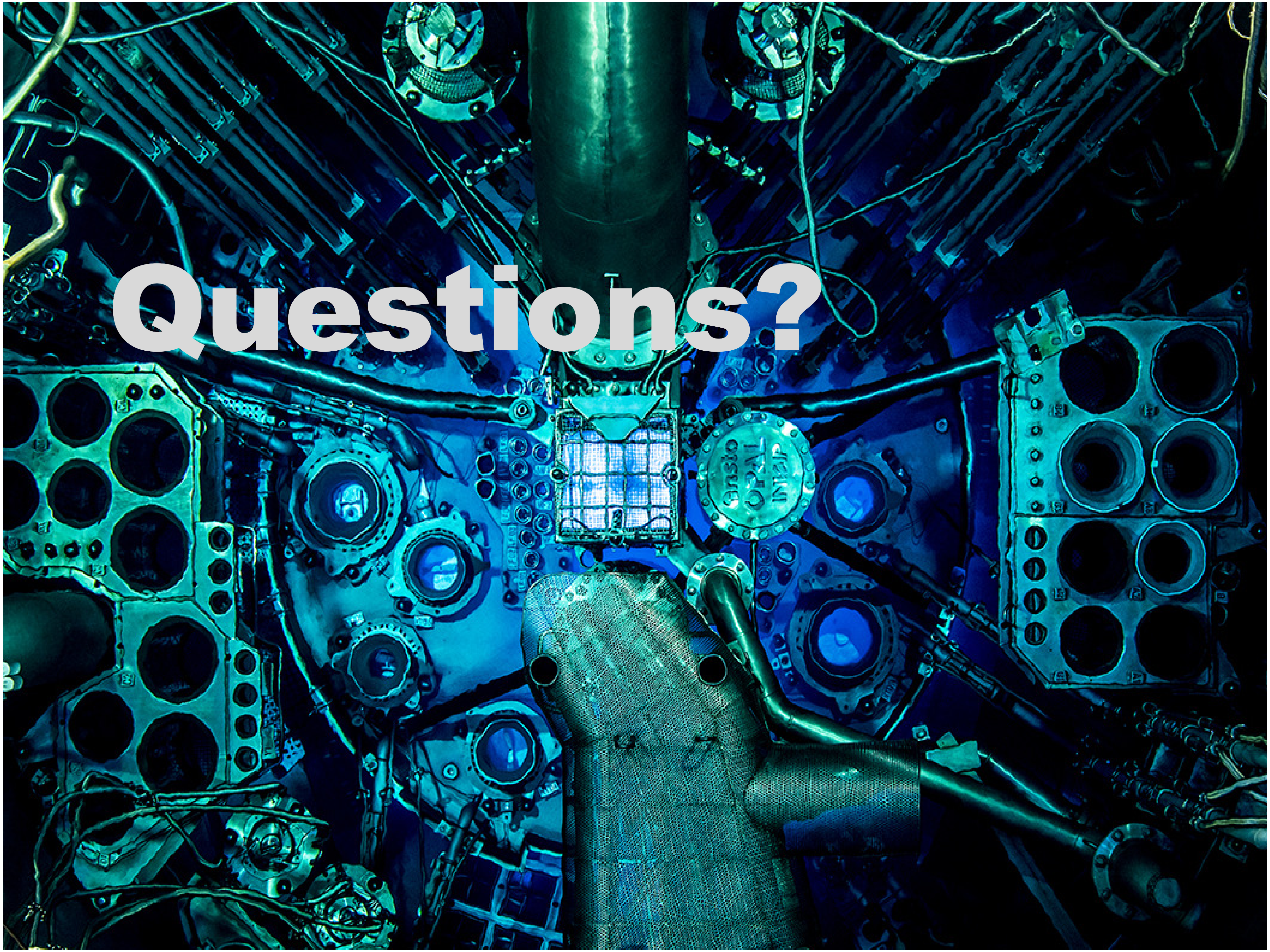
Other Applications…

- Vibrational and rotational excitations in disordered solids.
- Magnetic and vibrational excitations in artificial structures
- Multiferroics
- High Tc Superconductors
- One and two dimensional magnets.
- Magnets with frustrated interactions.
- Magnets with characteristic energy scales below 10 K.
- Spin interactions in diluted magnetic systems.
- Spin glasses.
- Spin correlations in heavy fermion metals.
- Spin correlations in Kondo insulators.
- …


Questions?

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