# Magnetic Excitations II

Andrew Wildes
Institut Laue-Langevin

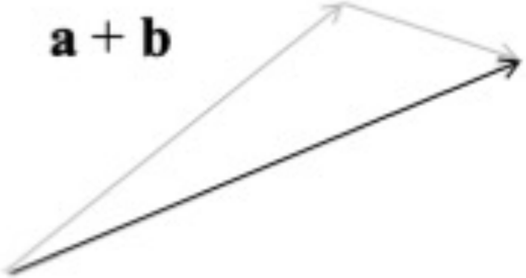
# Plan:

- Reminders
- Measurements of powders
- Measurements of single crystals

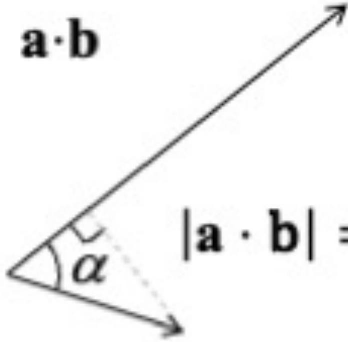
# Learn to work with vectors

**a**

**a + b**



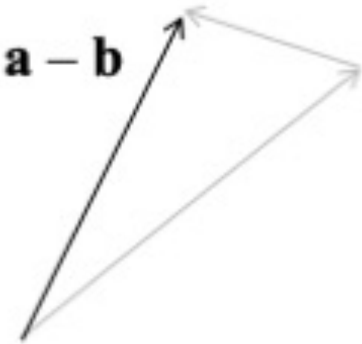
**a·b**



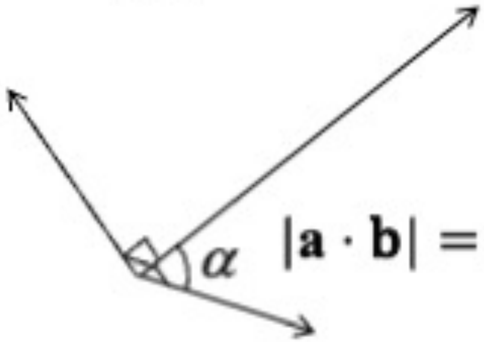

$$|\mathbf{a} \cdot \mathbf{b}| = |\mathbf{a}||\mathbf{b}| \cos \alpha$$

**b**

**a – b**



**a×b**



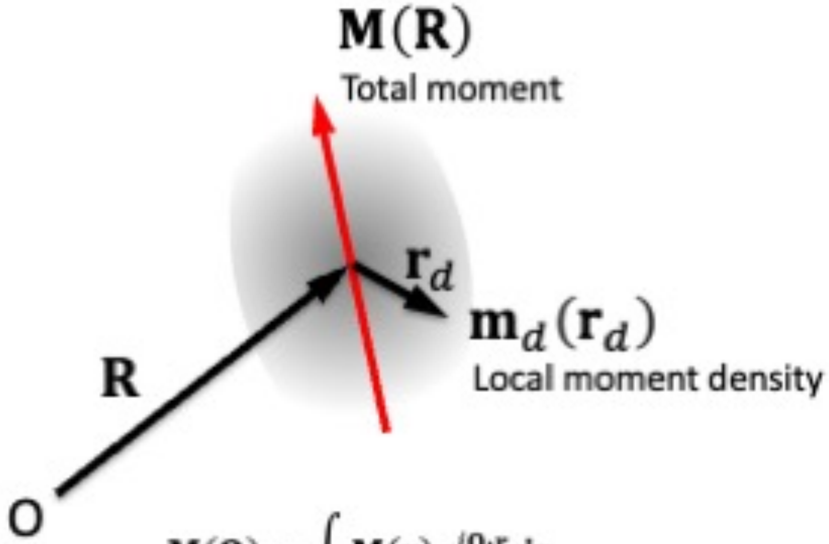

$$|\mathbf{a} \cdot \mathbf{b}| = |\mathbf{a}||\mathbf{b}| \sin \alpha$$

Reminder 1

## The fundamental rule of neutron magnetic scattering

Neutrons *only ever* see the components of the magnetization that are *perpendicular* to the scattering vector!

# Magnetic scattering has *a form factor* $f(\mathbf{Q})$

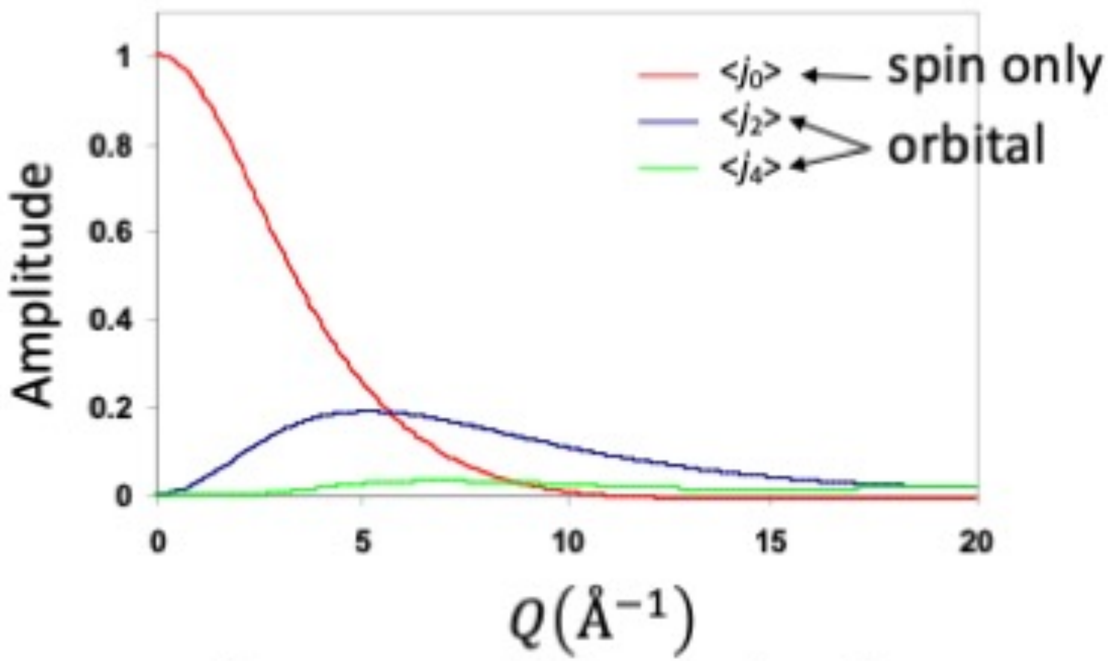



$$\mathbf{M}(\mathbf{Q}) = \int \mathbf{M}(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r}$$

$$= \int \mathbf{m}_d e^{i\mathbf{Q}\cdot\mathbf{r}_d} d\mathbf{r}_d \int \mathbf{M}(\mathbf{R}) e^{i\mathbf{Q}\cdot\mathbf{R}} d\mathbf{R}$$

$$= \underline{f(Q)} \int \mathbf{M}(\mathbf{R}) e^{i\mathbf{Q}\cdot\mathbf{R}} d\mathbf{R}$$

Form factors for iron



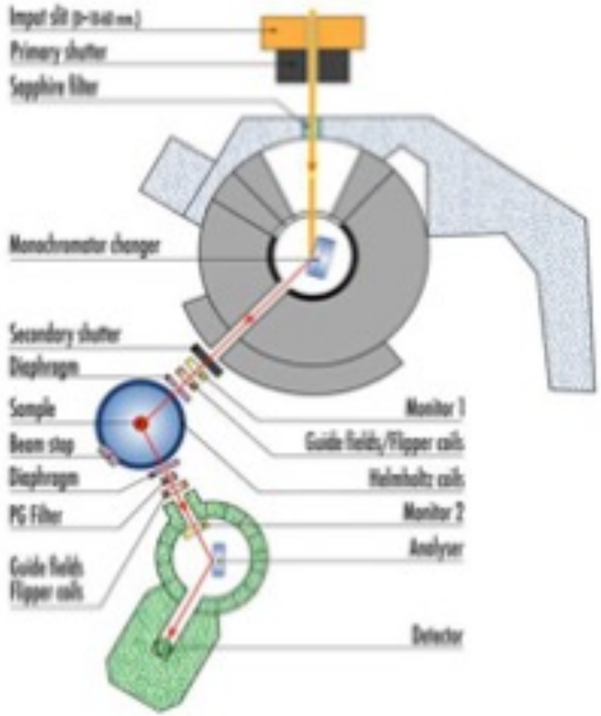

*Deceases* with *increasing Q*

A measurement of $\frac{d^2\sigma}{d\Omega dE_f}$ requires a knowledge of $\mathbf{k}_i$ and $\mathbf{k}_f$

Conventional instrumentation

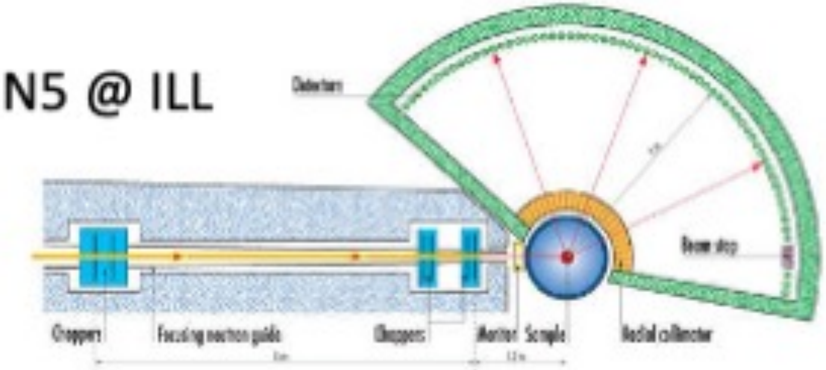
Three-axis spectrometry

IN20 @ ILL

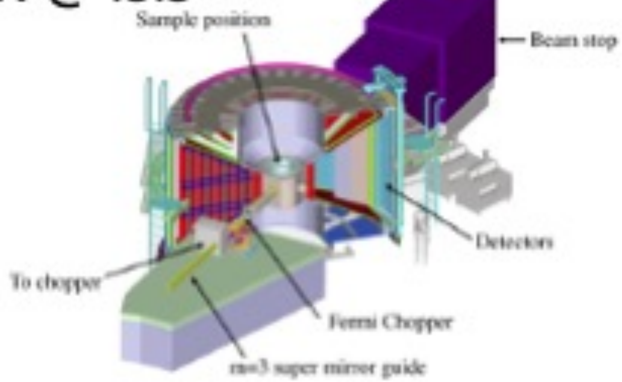



Time-of-flight spectrometry

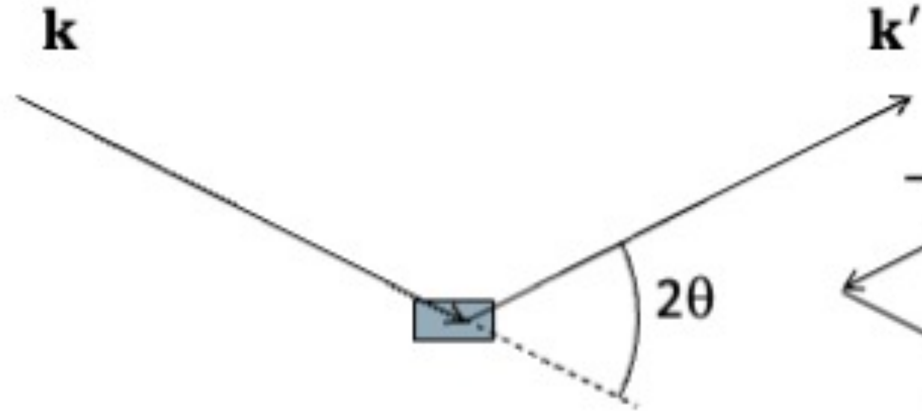
IN5 @ ILL



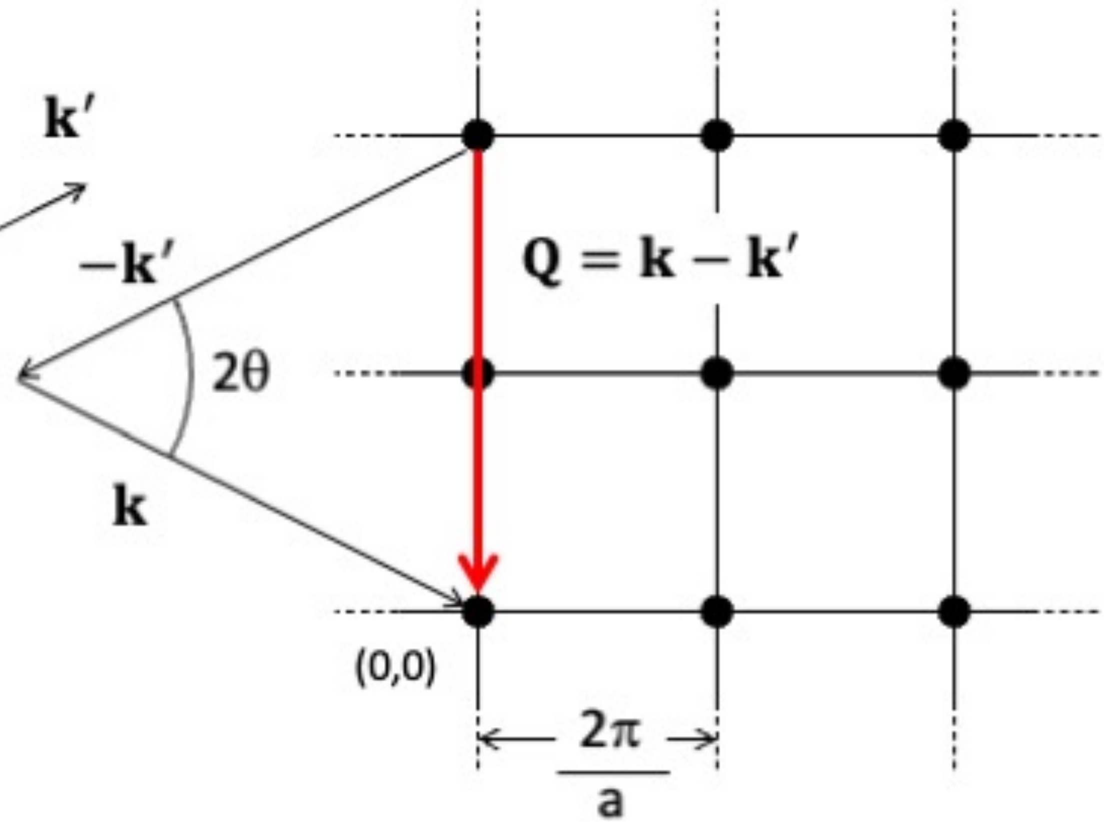

MERLIN @ ISIS



# Inelastic scattering




Fourier transformed structure




Momentum transfer:

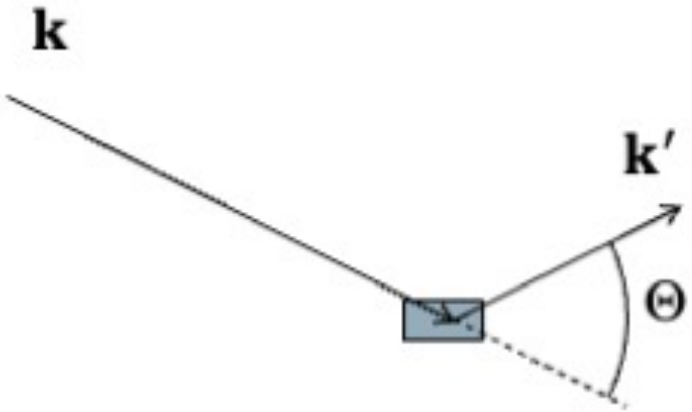
$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

Energy transfer:

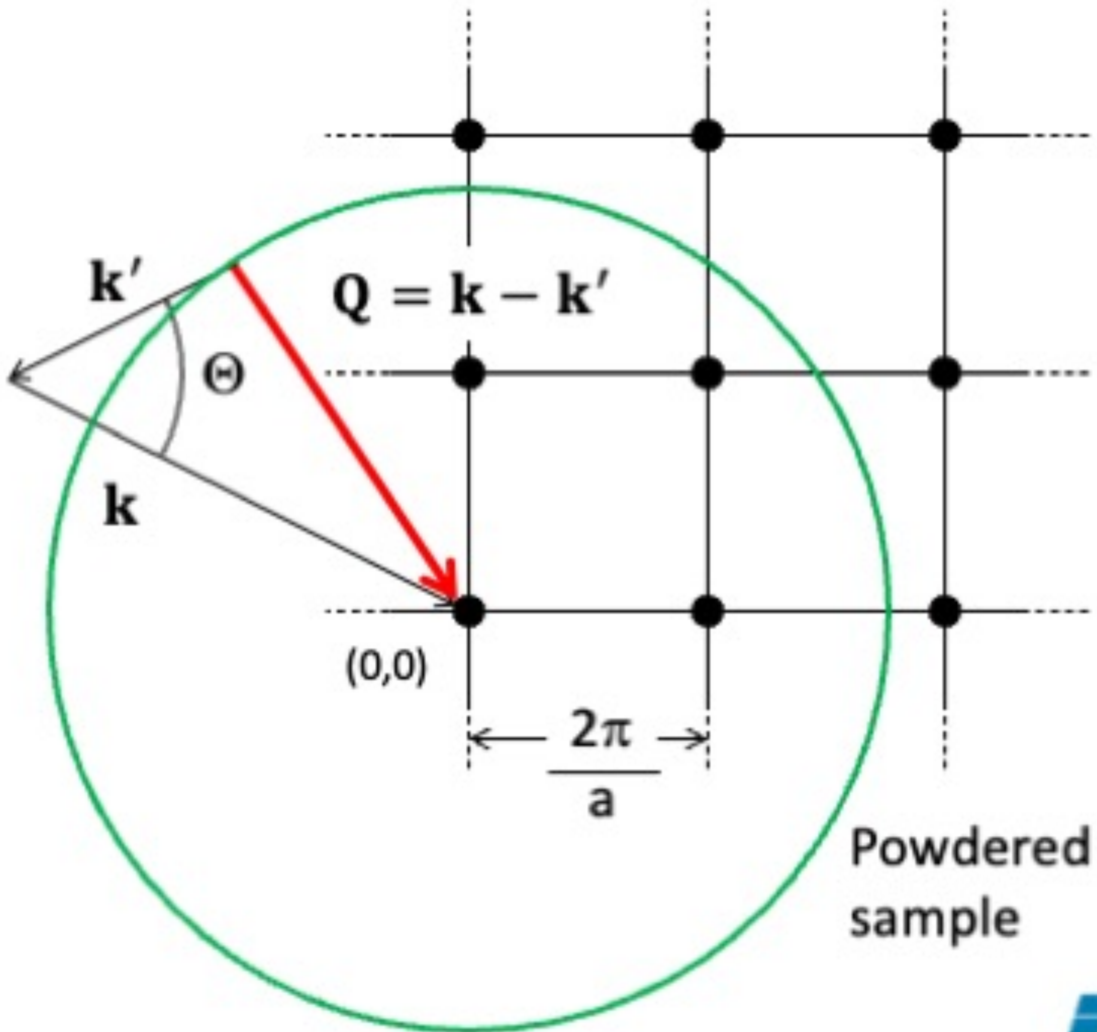
$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

Bragg's Law: $2d\sin\theta = \lambda$

# Inelastic scattering




Fourier transformed structure




Momentum transfer:

$$Q^2 = k^2 + k'^2 - 2kk' \cos\Theta$$

Energy transfer:

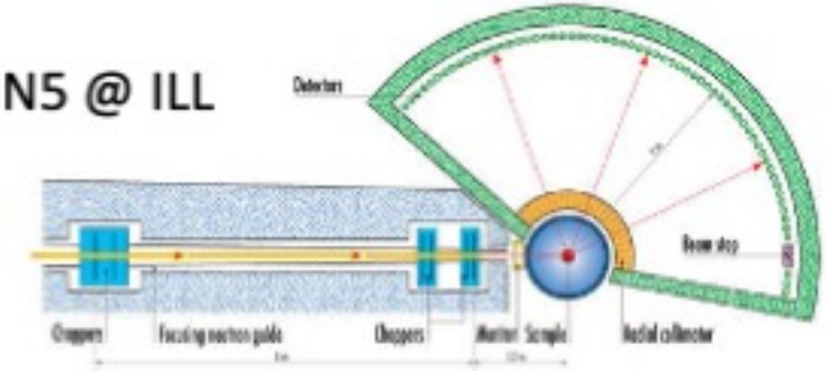
$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

# Measuring powder samples

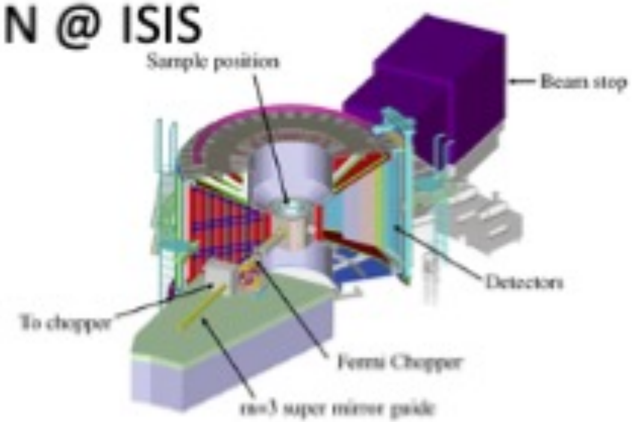
Usually best done on time-of-flight instrument

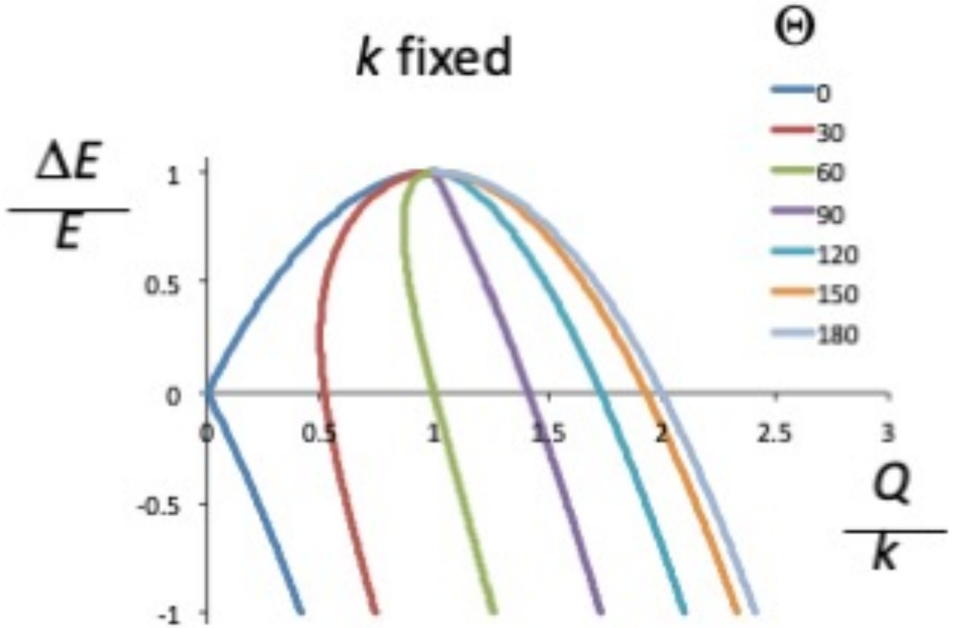
Time-of-flight spectrometry

IN5 @ ILL




MERLIN @ ISIS



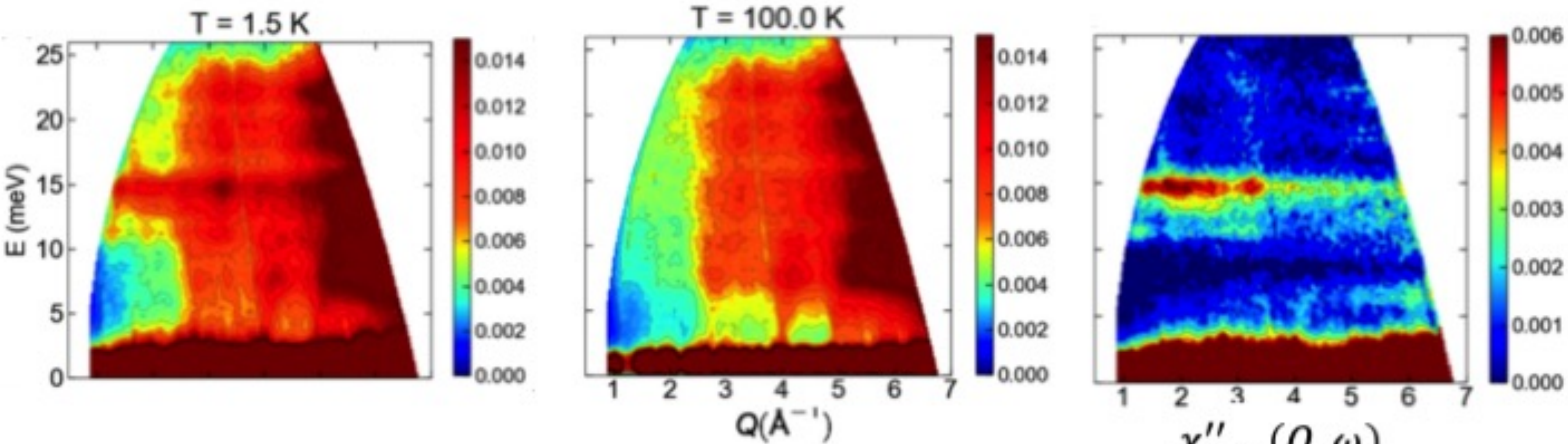



# Temperature subtractions

Triplet excitations in $Li_2Cu_2O(SO_4)_2$

O. Vacccarelli *et al.*, PRB **99** (2019) 064416

$$S(Q,\omega) = \frac{1+n(\omega)}{\pi}\chi''(Q,\omega)$$

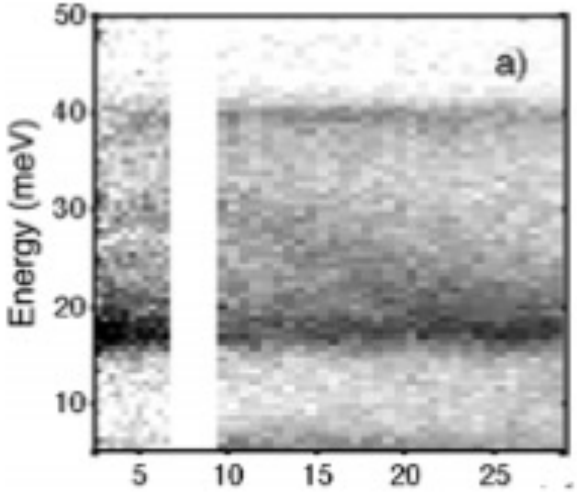


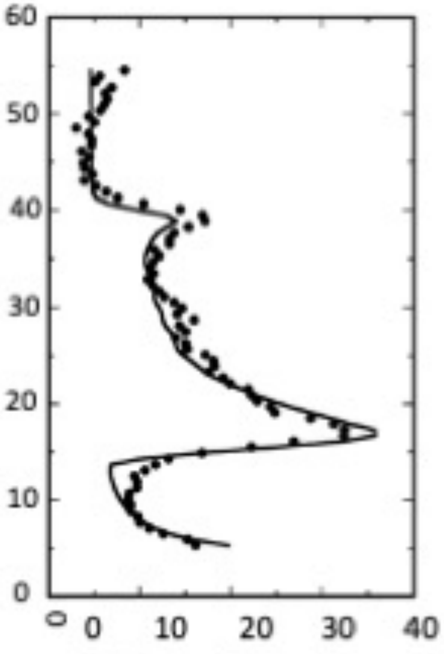

$$\chi''_{\text{mag}}(Q,\omega) = \chi''_{1.5\text{K}}(Q,\omega) - \chi''_{100\text{K}}(Q,\omega)$$

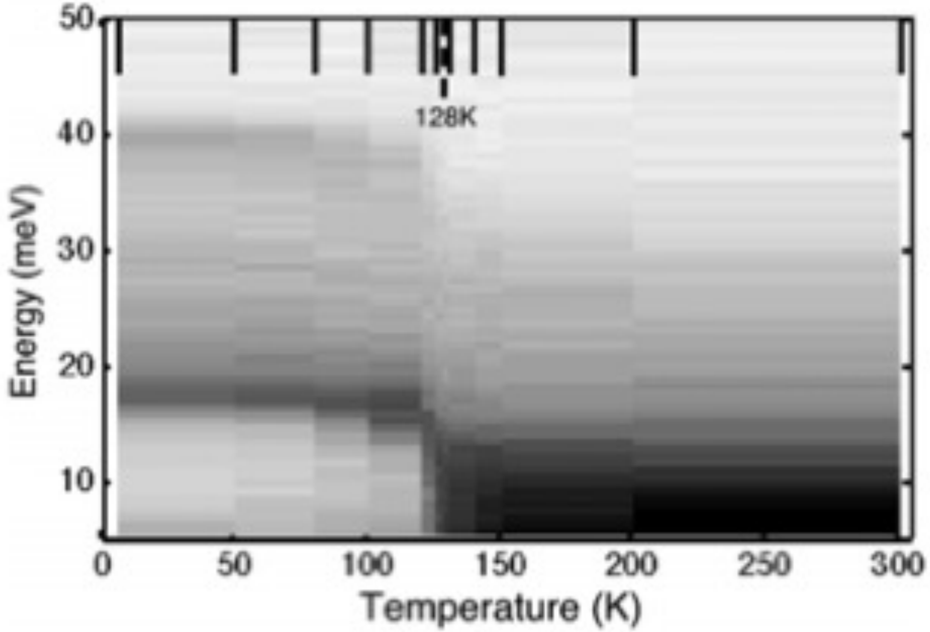
# Temperature subtractions

## Magnons in $FePS_3$

A. R. Wildes *et al.*, JPCM **24** (2012) 416004



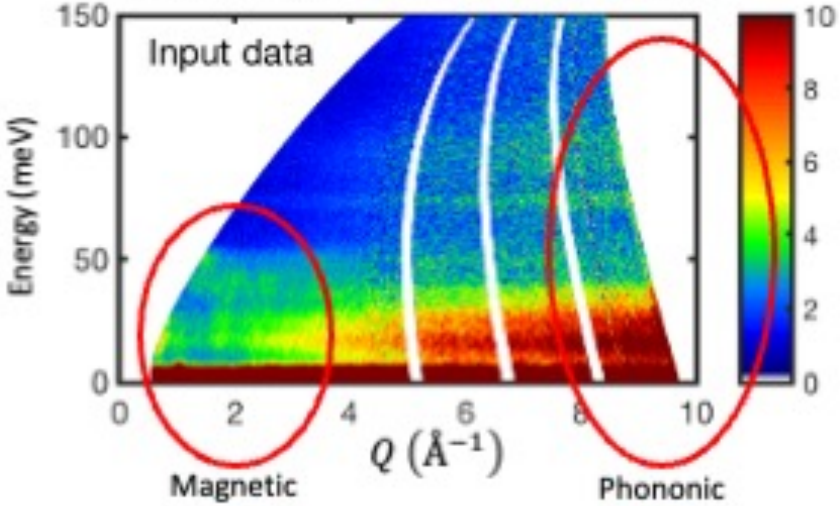
# Q-dependence

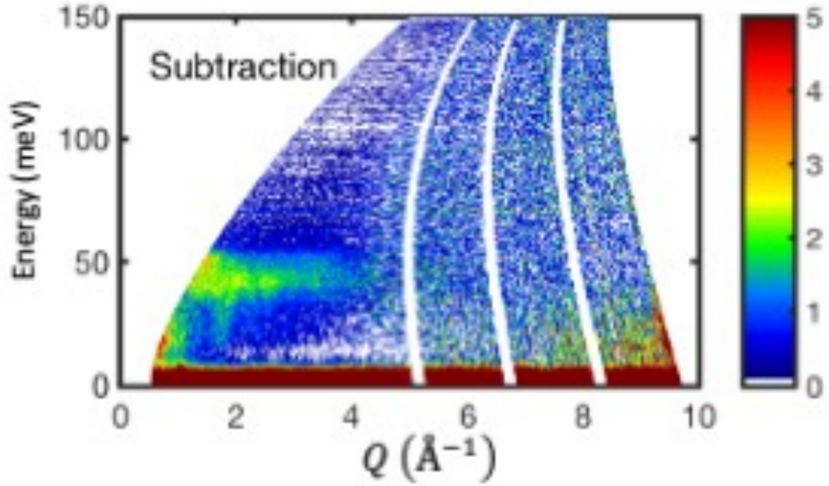
Magnetic scattering $\propto f^2(Q)$

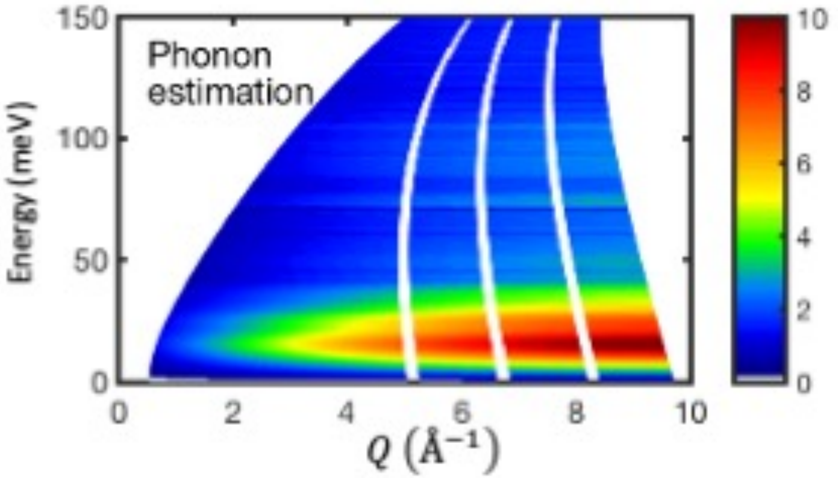
Phonon scattering $\propto Q^2 e^{-DQ^2}$

## Spin waves in $NiPS_3$

D. Lançon *et al.*, PRB **98** (2018) 134414




## Powder phonons in $LaFe_4Sb_{12}$

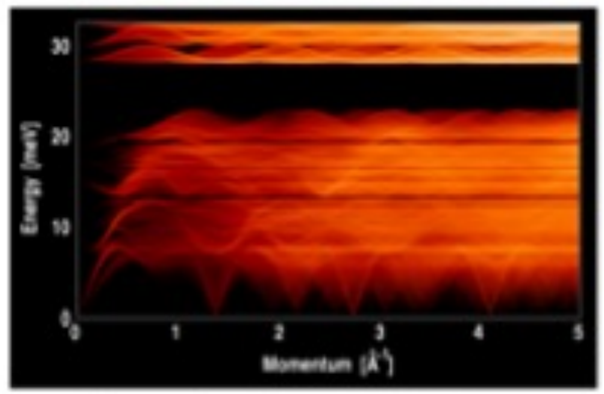


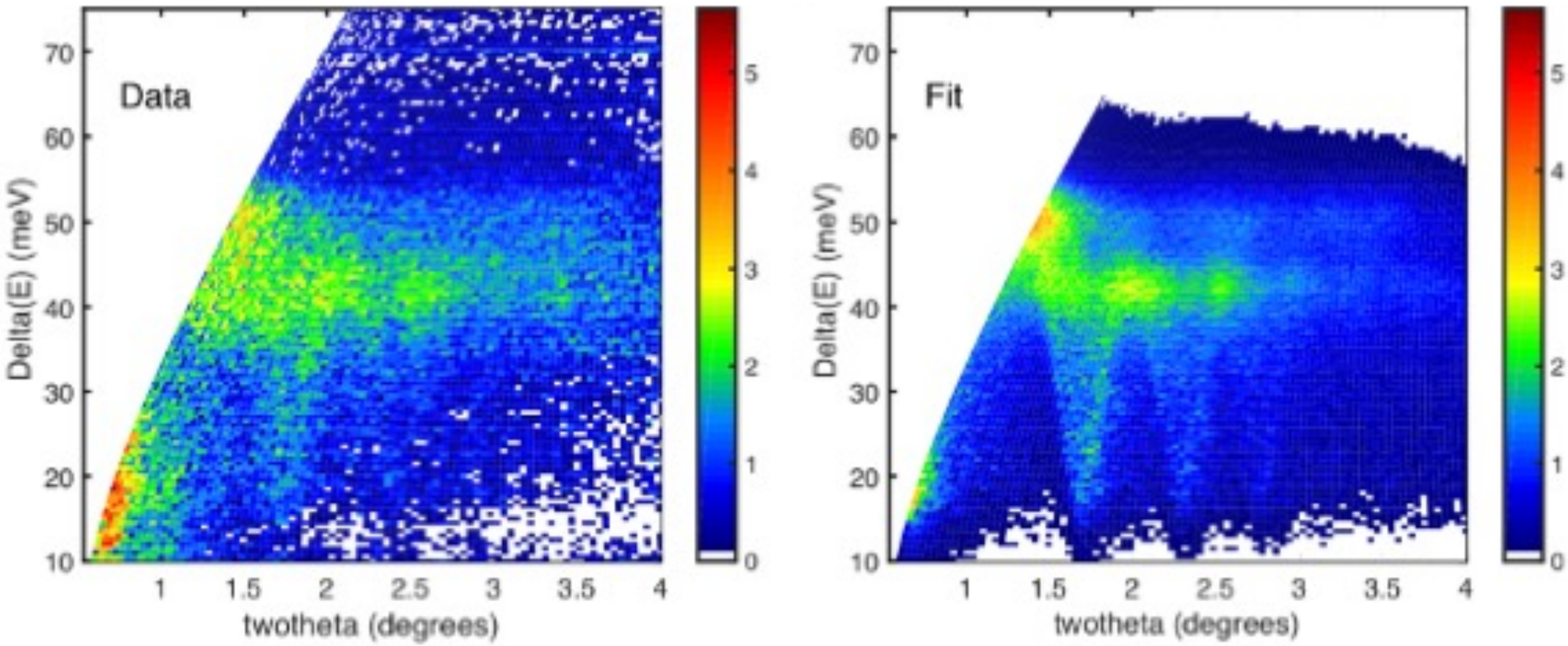

Figure thanks to M. M. Koza

# Q-dependence

## Spin waves in $NiPS_3$

D. Lançon *et al.*, PRB **98** (2018) 134414

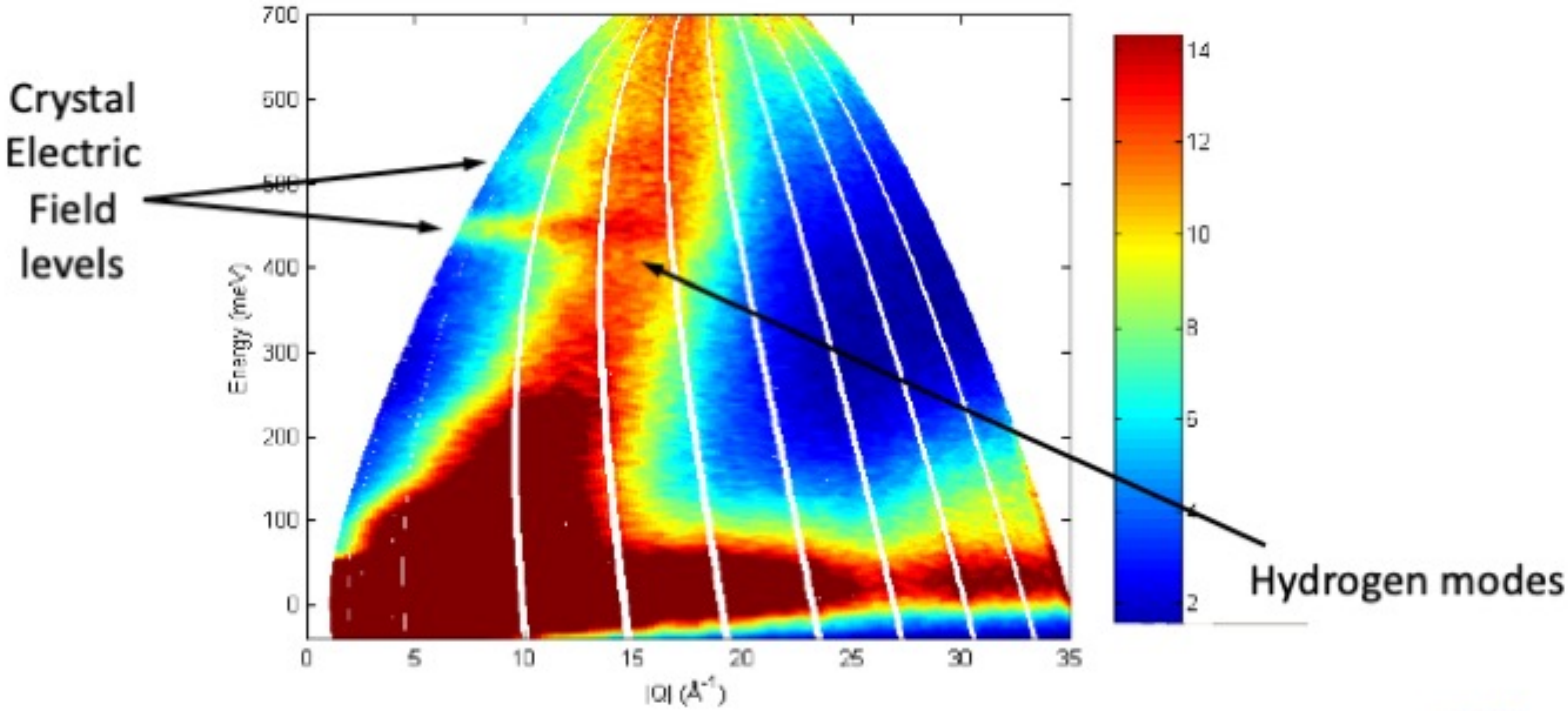
$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\left(\frac{\gamma r_0}{2\mu_B}\right)^2 \sum_{\alpha,\beta}(1 - \hat{Q}_\alpha \hat{Q}_\beta) S_{\alpha\beta}(\mathbf{Q}, \omega)$$



# Q-dependence

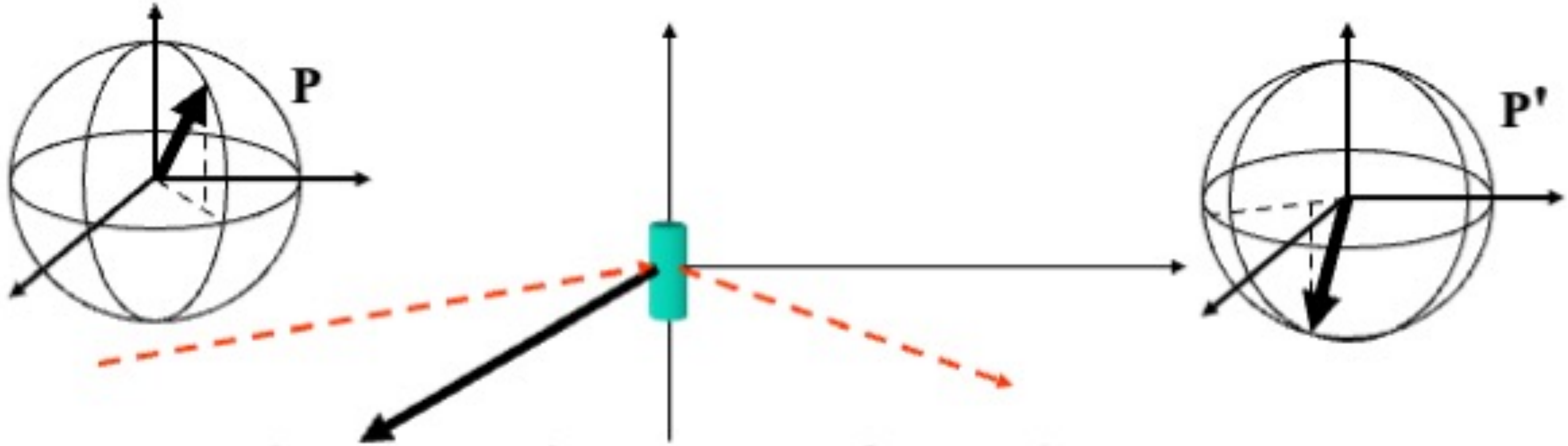
Powdered $Sr_2PO_4$

J. Taylor and D. McK. Paul



R. M. Moon, T. Riste and W. C. Koehler, Phys. Rev. **181** (1969) 920
J. R. Stewart *et al.*, J. Appl. Cryst. **42** (2009) 69

Polarization is the ensemble average of all the neutrons in the beam




There are three sets of coordinates:

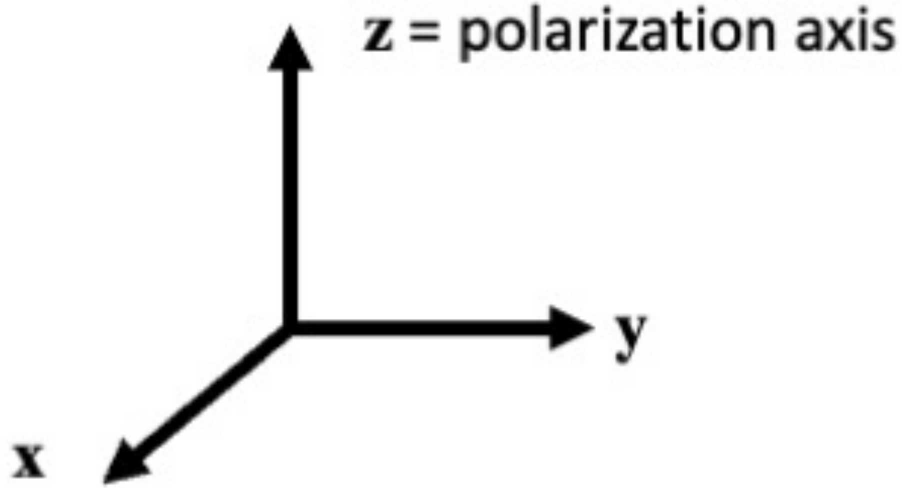
i) Coordinates for the instrument

ii) Coordinates for the magnetism
(Needed to define $\mathbf{M}_\perp$)

iii) Coordinates for the polarization

# Polarized neutrons

R. M. Moon, T. Riste and W. C. Koehler, Phys. Rev. **181** (1969) 920
J. R. Stewart *et al.*, J. Appl. Cryst. **42** (2009) 69




Potential $V \rightarrow U^{\alpha\beta}$

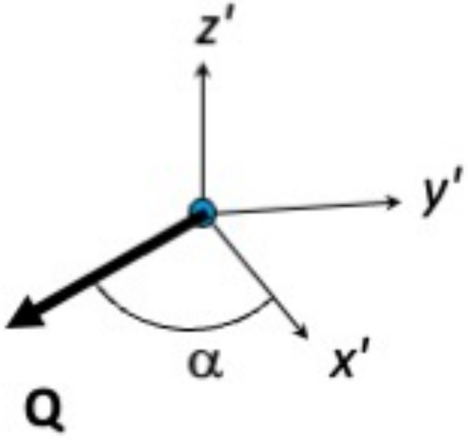
$$U^{++} = b - M_{\perp z} + BI_z$$

$$U^{--} = b + M_{\perp z} - BI_z$$

$$U^{+-} = -\left(M_{\perp x} + iM_{\perp y}\right) + B\left(I_x + iI_y\right)$$

$$U^{-+} = -\left(M_{\perp x} - iM_{\perp y}\right) + B\left(I_x - iI_y\right)$$

# 'XYZ' Polarization Analysis




$$\left(\frac{\mathrm{d}\sigma^{\mathrm{NSF}}}{\mathrm{d}\Omega}\right)_{x'} = \frac{\mathrm{d}\sigma_{\mathrm{Nuc}}}{\mathrm{d}\Omega} + \frac{1}{3}\frac{\mathrm{d}\sigma_{\mathrm{NSI}}}{\mathrm{d}\Omega} + \frac{1}{2}\frac{\mathrm{d}\sigma_{\mathrm{PM}}}{\mathrm{d}\Omega}\sin^2\alpha$$

$$\left(\frac{\mathrm{d}\sigma^{\mathrm{SF}}}{\mathrm{d}\Omega}\right)_{x'} = \frac{2}{3}\frac{\mathrm{d}\sigma_{\mathrm{NSI}}}{\mathrm{d}\Omega} + \frac{1}{2}\frac{\mathrm{d}\sigma_{\mathrm{PM}}}{\mathrm{d}\Omega}(\cos^2\alpha + 1)$$

$$\left(\frac{\mathrm{d}\sigma^{\mathrm{NSF}}}{\mathrm{d}\Omega}\right)_{y'} = \frac{\mathrm{d}\sigma_{\mathrm{Nuc}}}{\mathrm{d}\Omega} + \frac{1}{3}\frac{\mathrm{d}\sigma_{\mathrm{NSI}}}{\mathrm{d}\Omega} + \frac{1}{2}\frac{\mathrm{d}\sigma_{\mathrm{PM}}}{\mathrm{d}\Omega}\cos^2\alpha$$

$$\left(\frac{\mathrm{d}\sigma^{\mathrm{SF}}}{\mathrm{d}\Omega}\right)_{y'} = \frac{2}{3}\frac{\mathrm{d}\sigma_{\mathrm{NSI}}}{\mathrm{d}\Omega} + \frac{1}{2}\frac{\mathrm{d}\sigma_{\mathrm{PM}}}{\mathrm{d}\Omega}(\sin^2\alpha + 1)$$

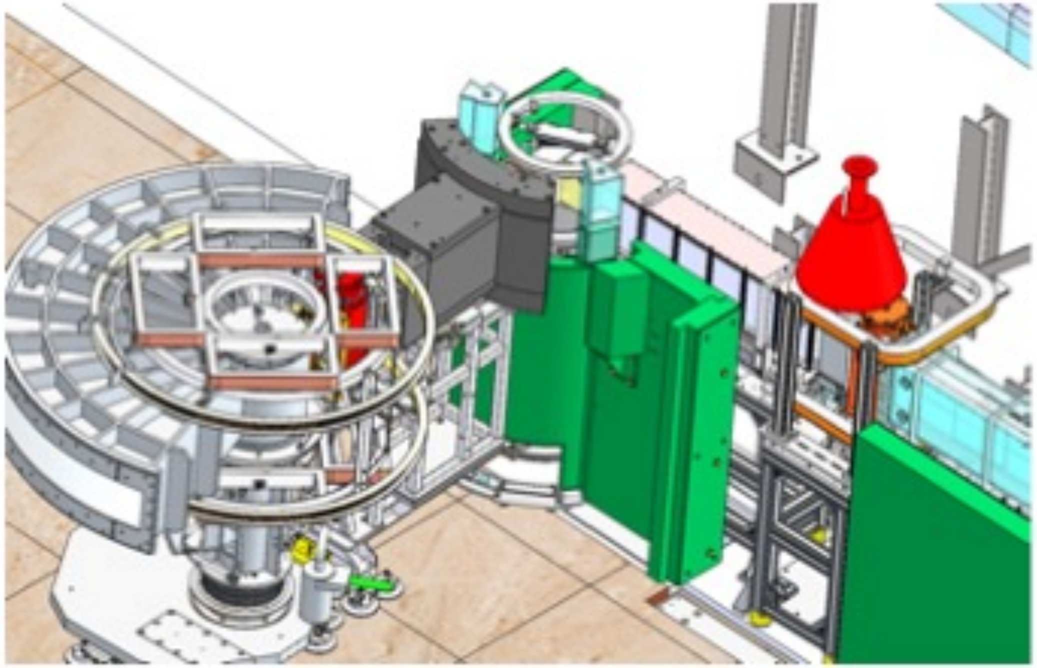
$$\left(\frac{\mathrm{d}\sigma^{\mathrm{NSF}}}{\mathrm{d}\Omega}\right)_{z'} = \frac{\mathrm{d}\sigma_{\mathrm{Nuc}}}{\mathrm{d}\Omega} + \frac{1}{3}\frac{\mathrm{d}\sigma_{\mathrm{NSI}}}{\mathrm{d}\Omega} + \frac{1}{2}\frac{\mathrm{d}\sigma_{\mathrm{PM}}}{\mathrm{d}\Omega}$$

$$\left(\frac{\mathrm{d}\sigma^{\mathrm{SF}}}{\mathrm{d}\Omega}\right)_{z'} = \frac{2}{3}\frac{\mathrm{d}\sigma_{\mathrm{NSI}}}{\mathrm{d}\Omega} + \frac{1}{2}\frac{\mathrm{d}\sigma_{\mathrm{PM}}}{\mathrm{d}\Omega}$$

O. Schärpf and H. Capellmann, Phys. Stat. Sol a **135** (1993) 359
J. R. Stewart *et al.*, J. Appl. Cryst. **42** (2009) 69

# D007

G. J. Nilsen *et al.*, NIMA **951** (2020) 162990

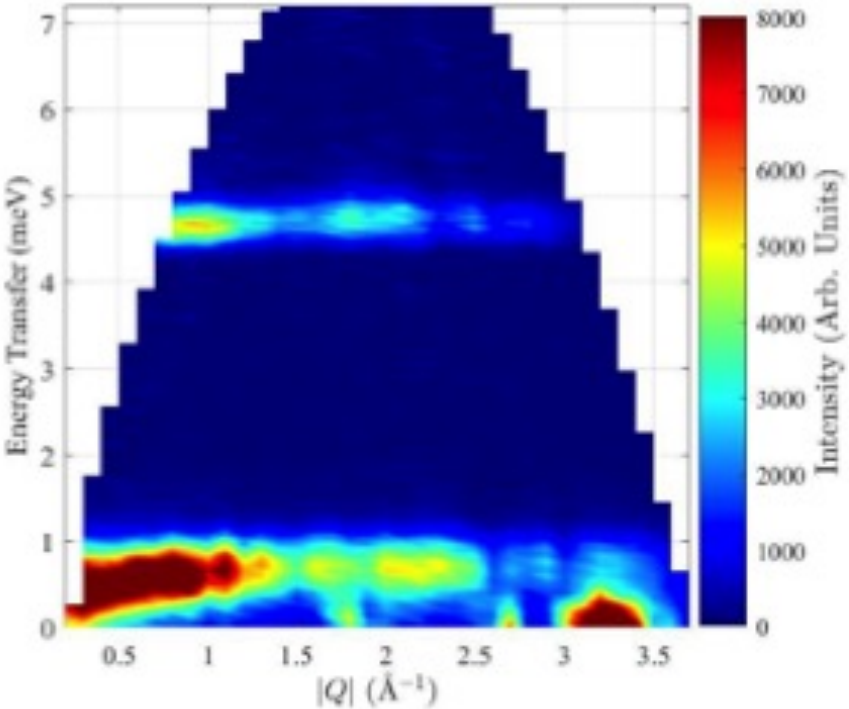


# 'XYZ' Polarization Analysis

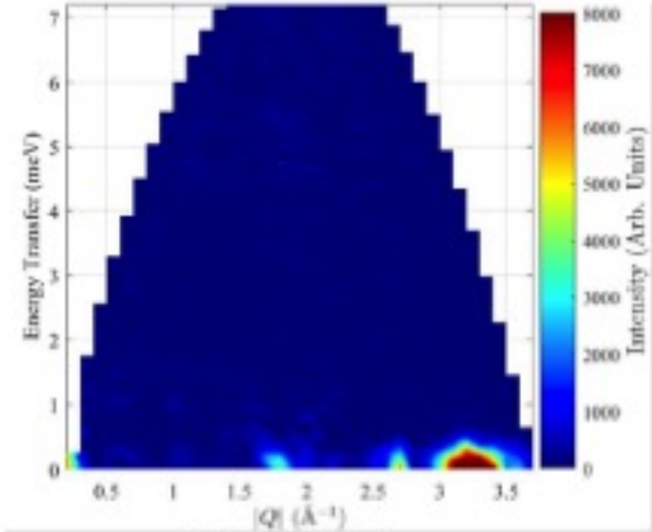
Inelastic scattering from powdered $HoF_3$

R. Dixey *et al.*, APL Mater **11** (2023) 041126

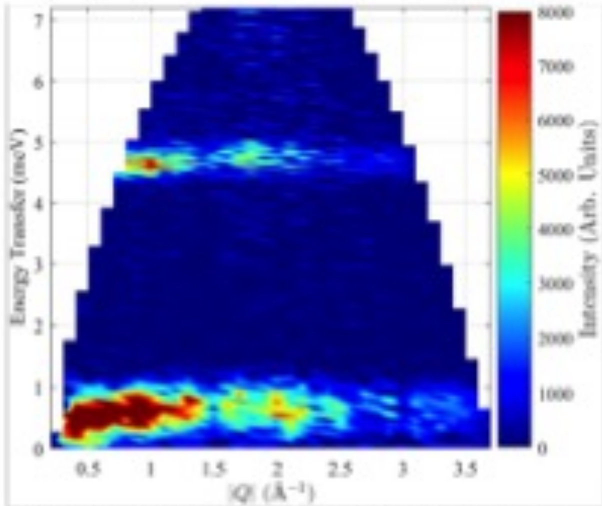
Total (unpolarized) scattering



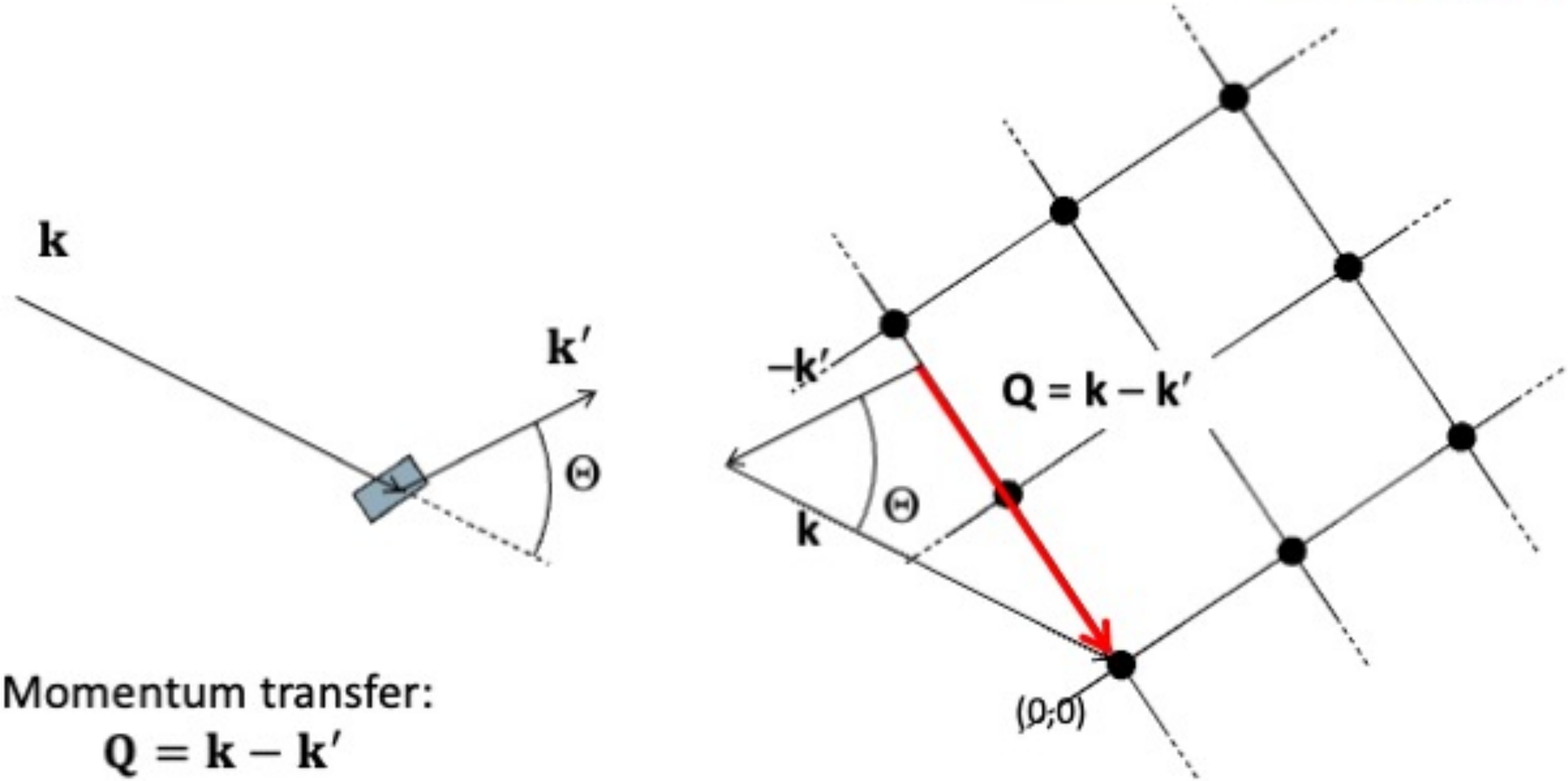

Nuclear coherent




Magnetic



# Inelastic scattering on single crystals



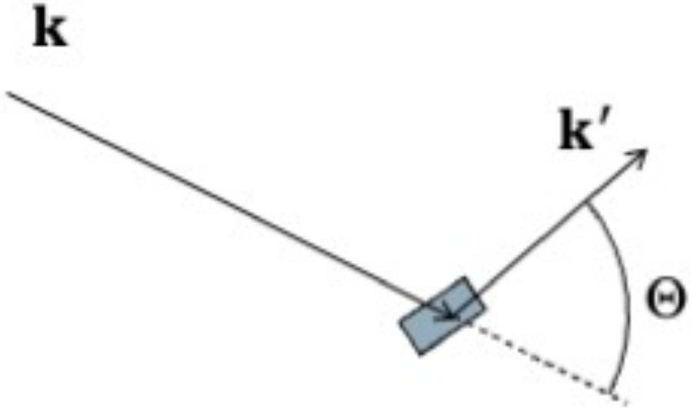

Momentum transfer:

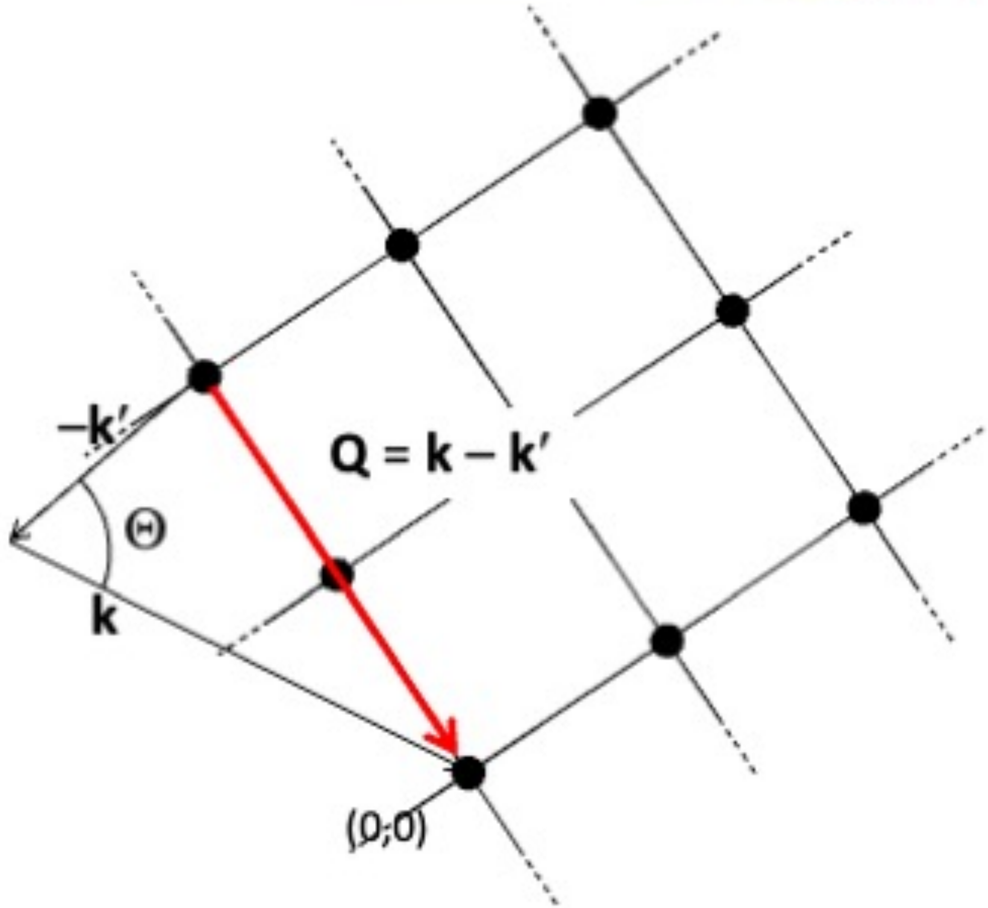
$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

Energy transfer:

$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

# Inelastic scattering on single crystals




Momentum transfer:

$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

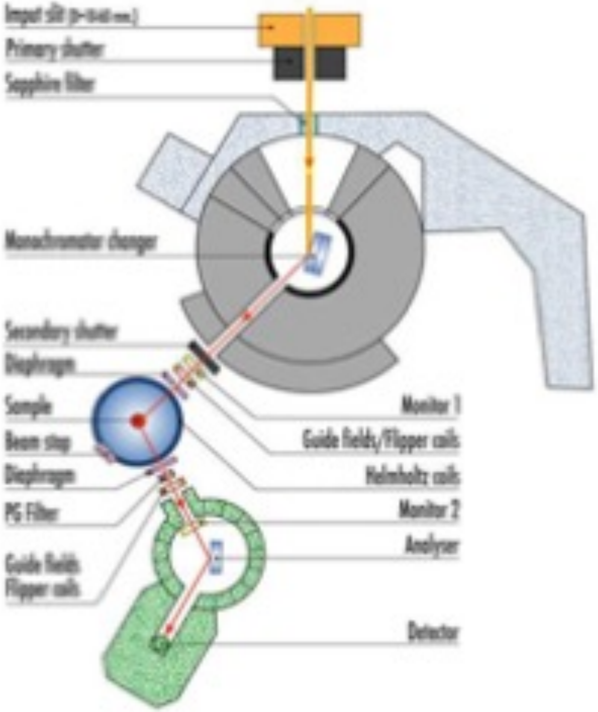
Energy transfer:

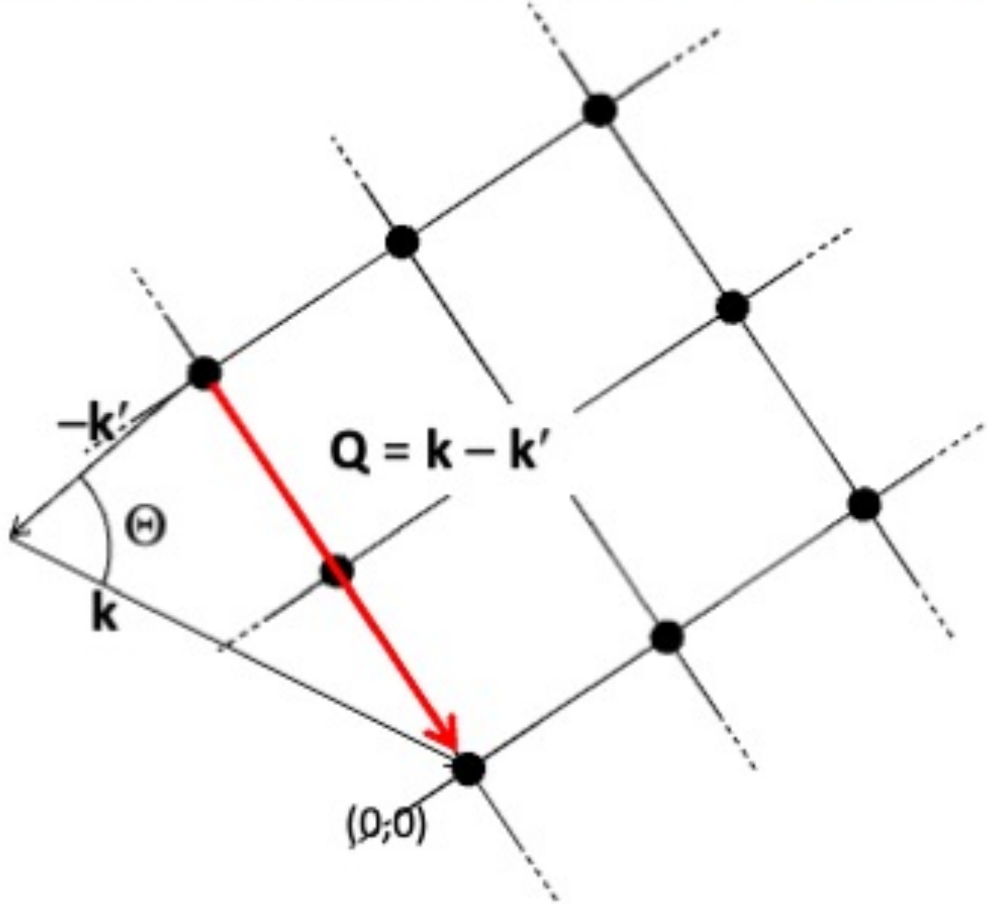
$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

# Inelastic scattering on single crystals

Straight-forward to visualize on a three-axis

IN20 @ ILL

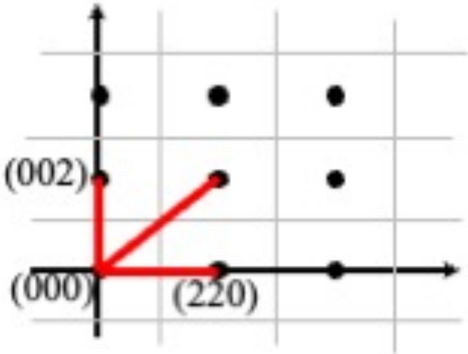


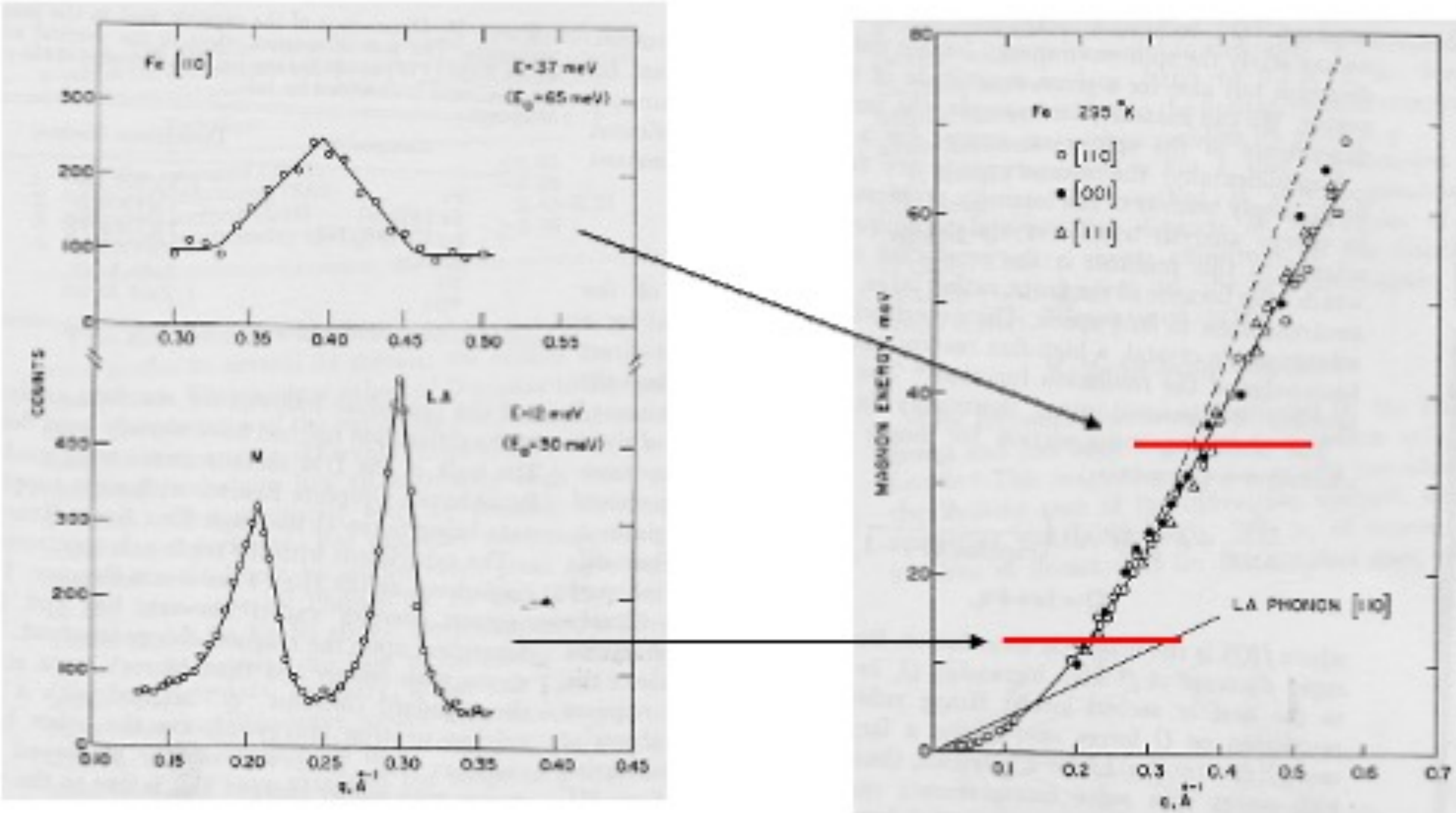



# Magnons in crystalline iron

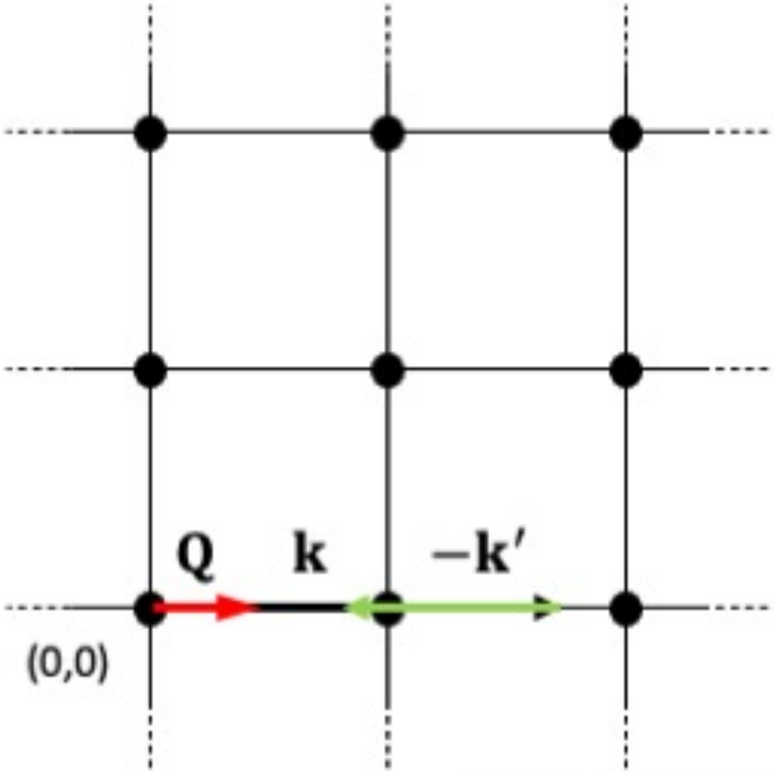
G. Shirane *et al.*, J. Appl. Phys. **39** (1968) 383

## How to discriminate against other contributions?



# Verifying magnetic signals




Magnetic intensity $\propto f^2(Q)$
Phonon intensity $\propto Q^2$

Momentum transfer:

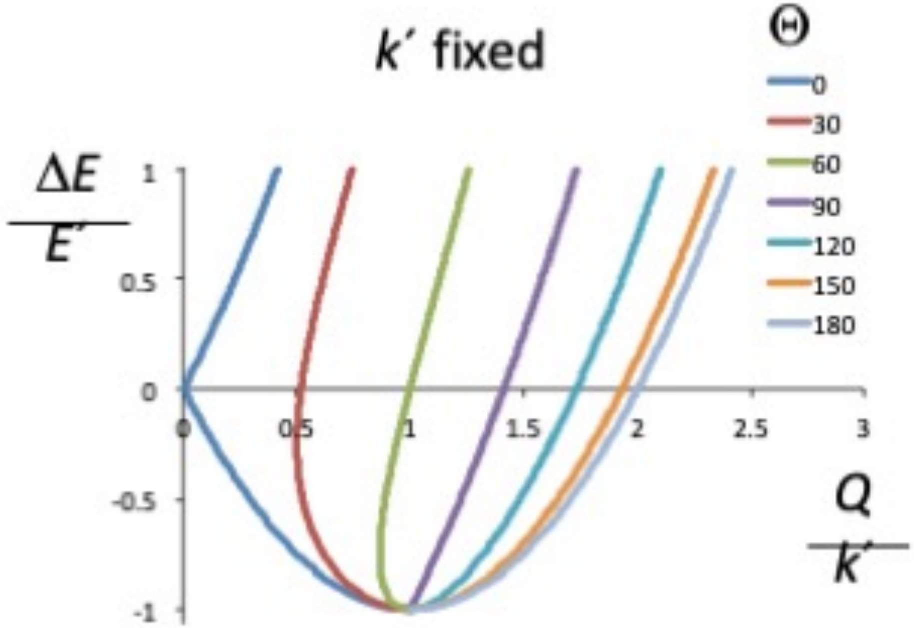
$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

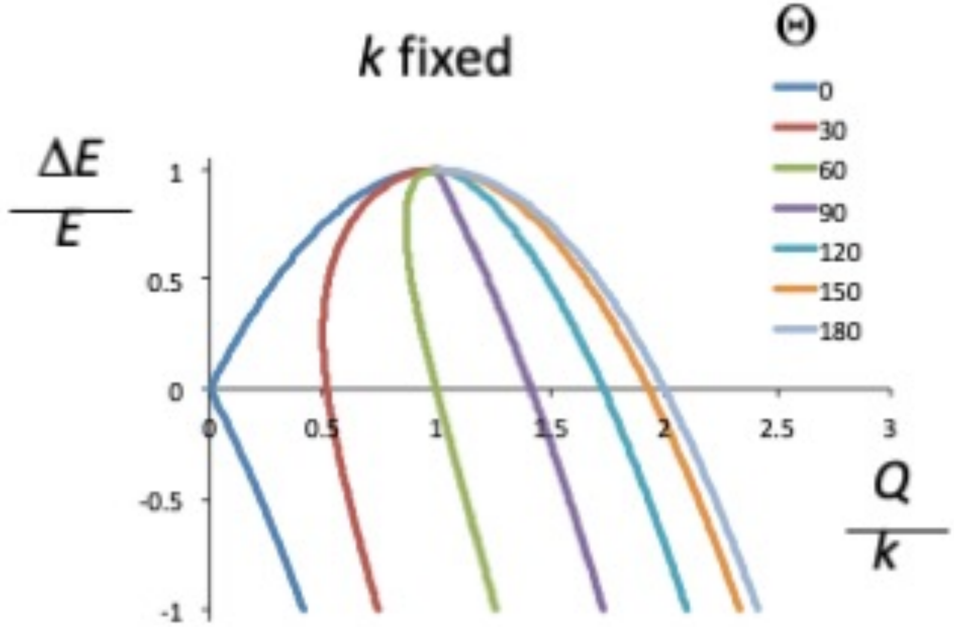
$$Q^2 = k^2 + k'^2 - 2kk' \cos\Theta$$

Energy transfer:

$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

# Kinematic constraints




Momentum transfer:

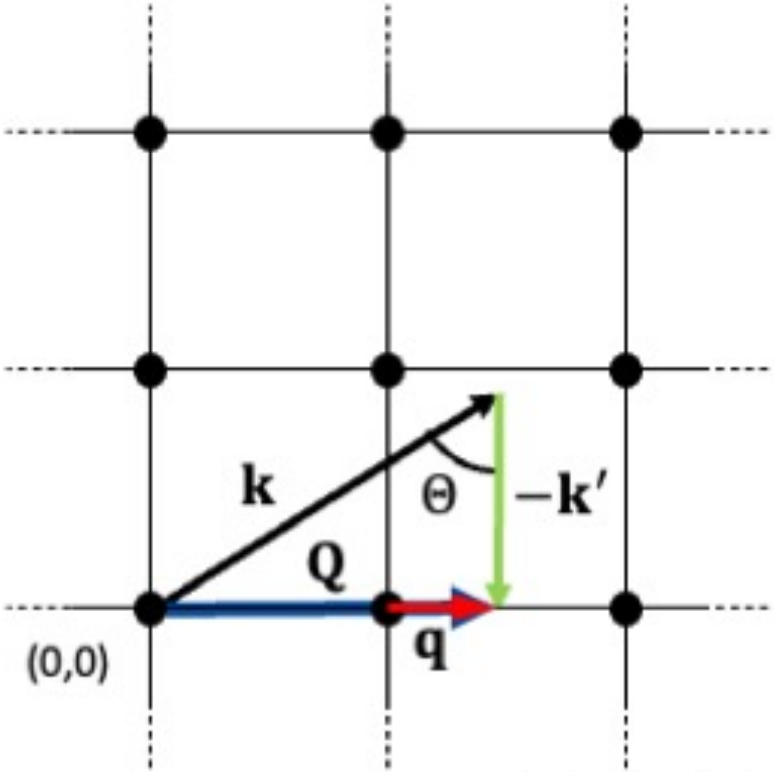
$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

$$Q^2 = k^2 + k'^2 - 2kk'\cos\Theta$$

Energy transfer:

$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

## Verifying magnetic signals




Magnetic intensity $\propto f^2(Q)$
Phonon intensity $\propto Q^2$

Momentum transfer:

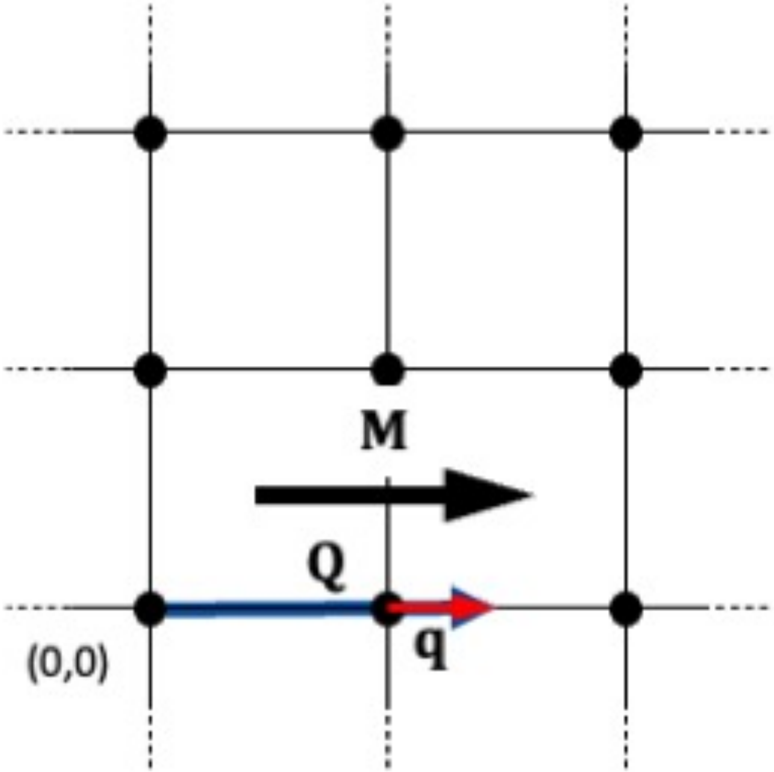
$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

$$Q^2 = k^2 + k'^2 - 2kk' \cos\Theta$$

Energy transfer:

$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

# Verifying magnetic signals




Magnetic intensity $\propto f^2(Q)$
Phonon intensity $\propto Q^2$

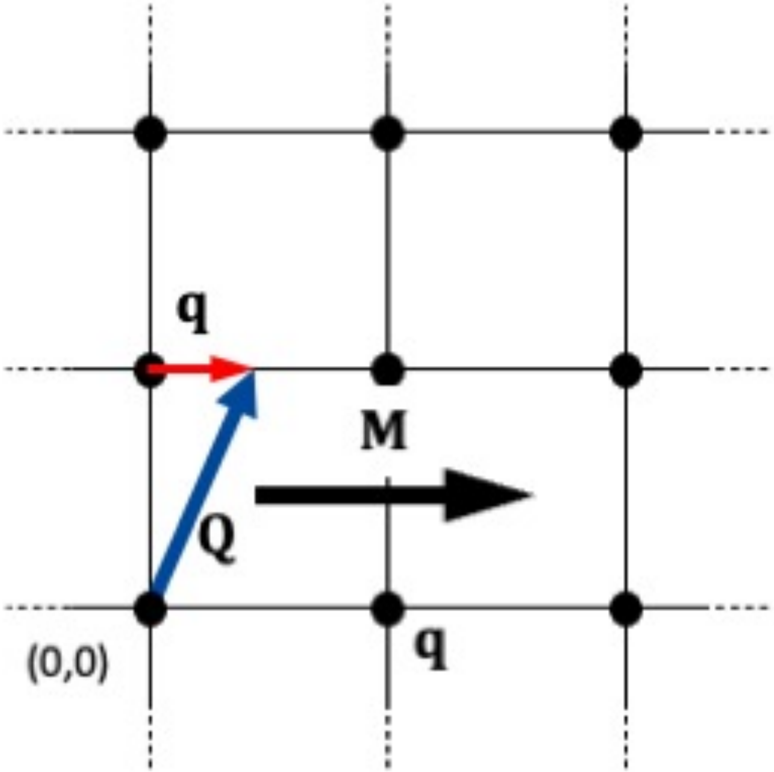
Magnons have $\mathbf{M}_\perp$

Intensity from *both* spin wave components

Phonons have $\mathbf{Q} \cdot \mathbf{e}$

Longitudinal mode

# Verifying magnetic signals

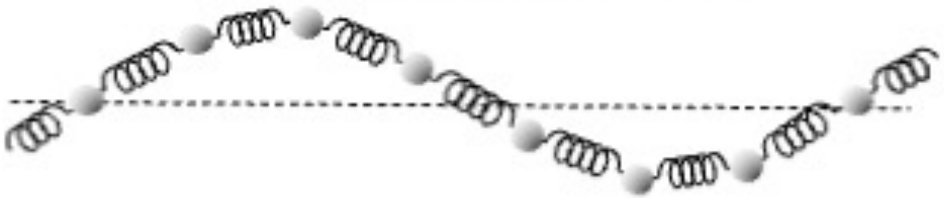



Magnetic intensity $\propto f^2(Q)$
Phonon intensity $\propto Q^2$

Magnons have $\mathbf{M}_{\perp}$

Intensity from *one* spin wave component

Phonons have $\mathbf{Q} \cdot \mathbf{e}$

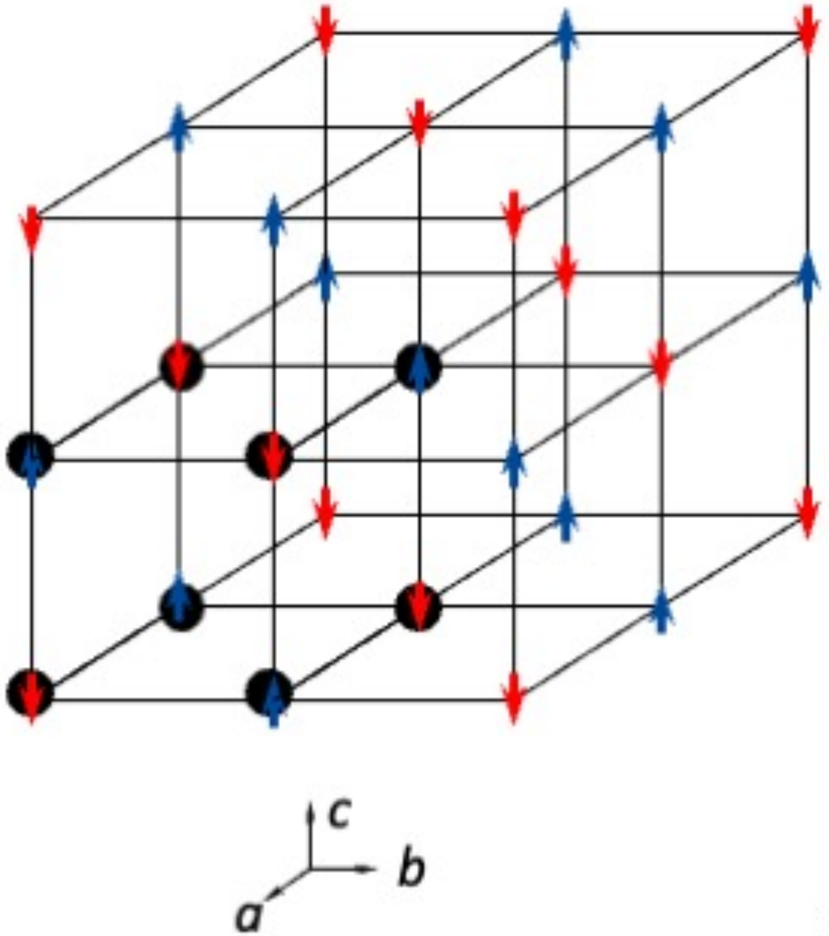


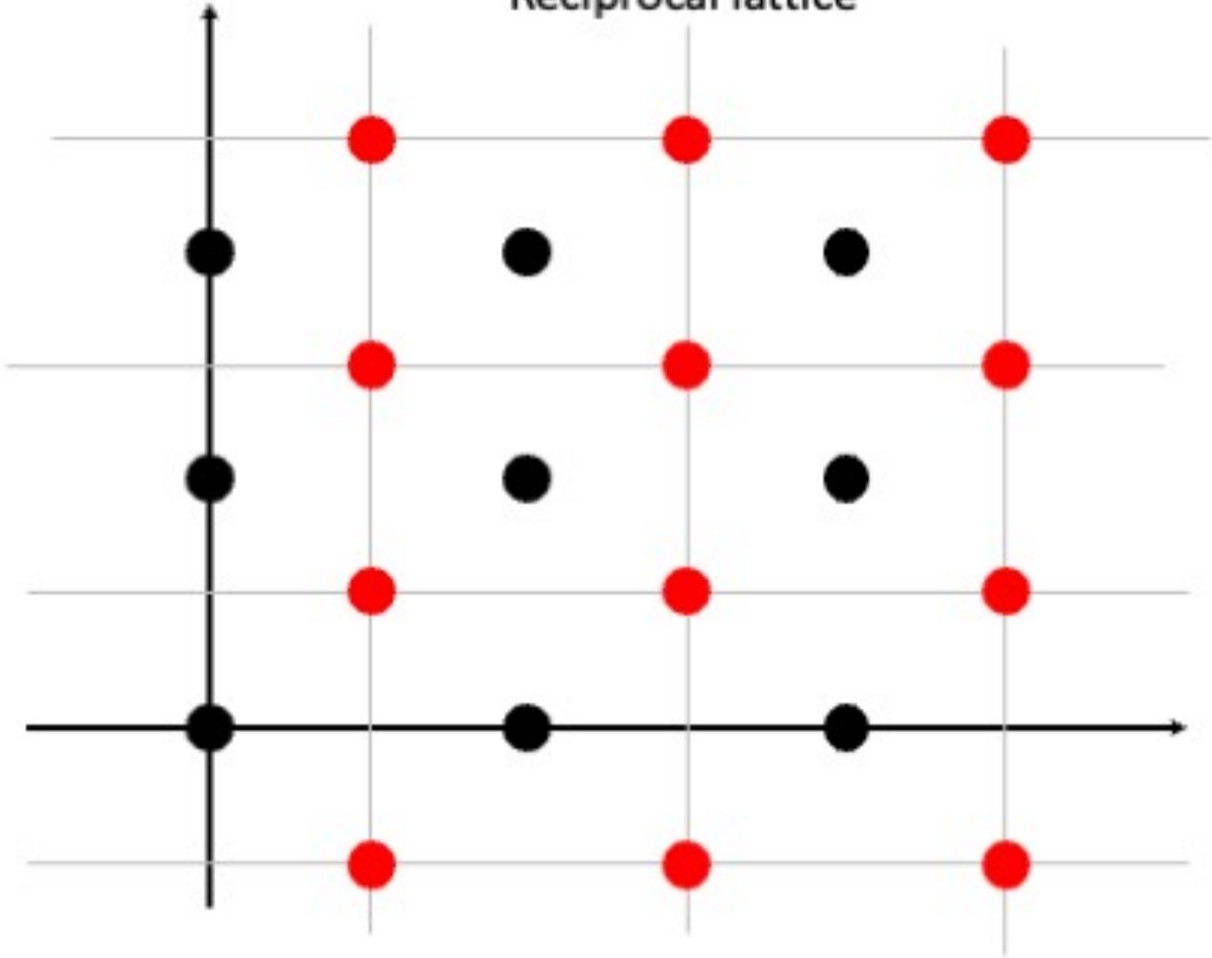
Transverse mode

**What about antiferromagnets?**

Antiferromagnetic magnon energies $\propto q$ at small $q$

Acoustic phonon energies $\propto q$ at small $q$



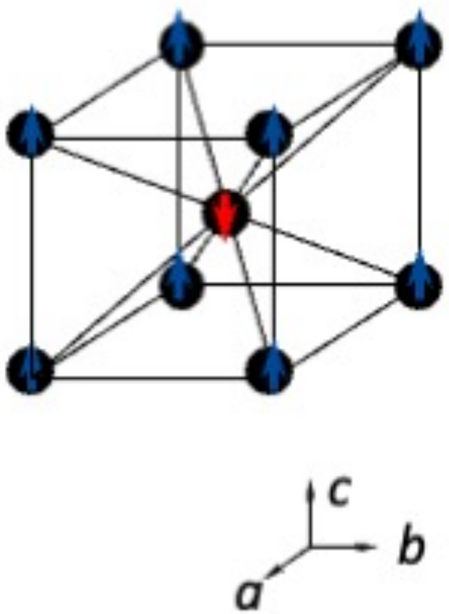

Periodicity is different

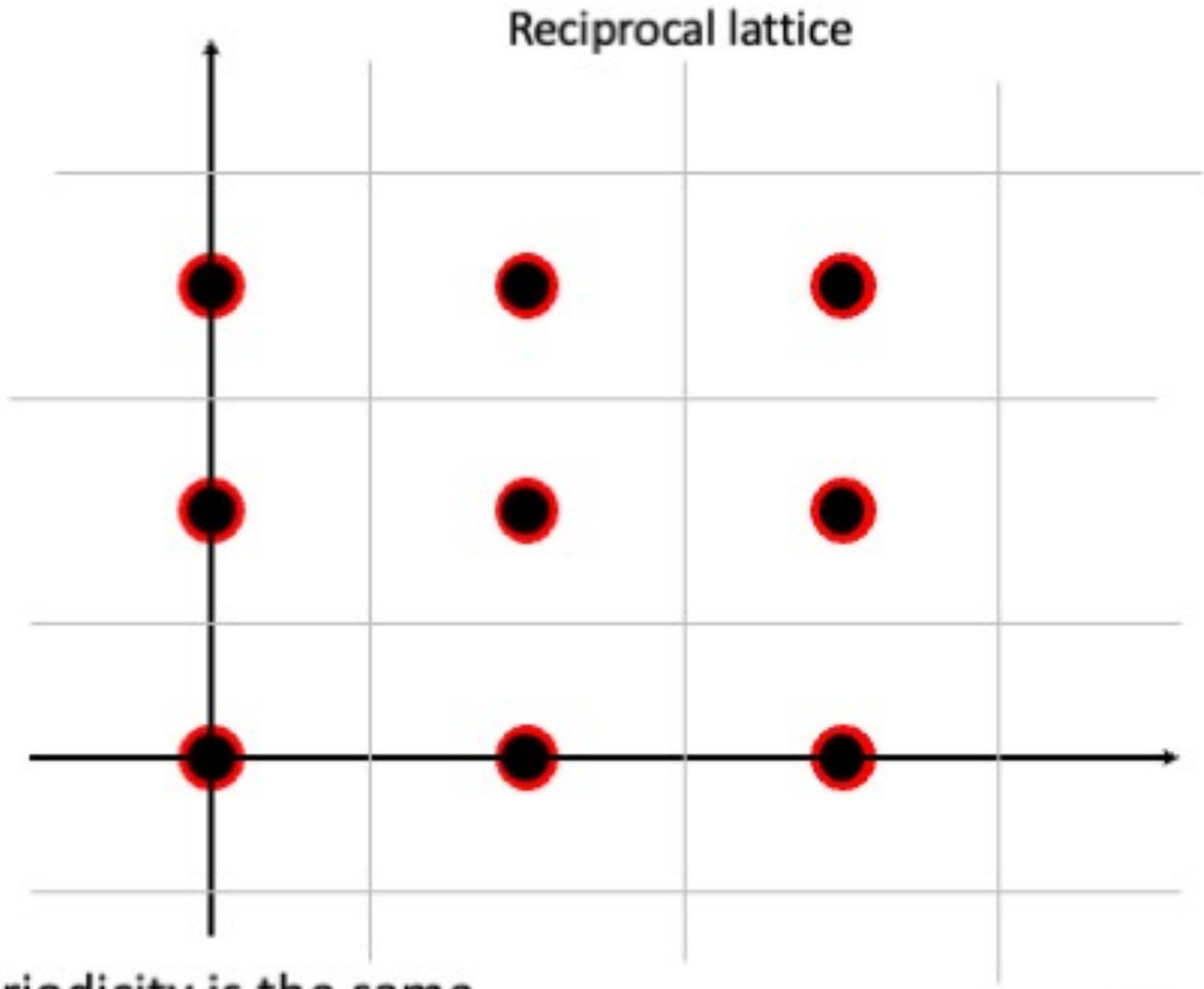
## What about antiferromagnets?

Antiferromagnetic magnon energies ∝ $q$ at small $q$

Acoustic phonon energies ∝ $q$ at small $q$

Propagation vector = 0



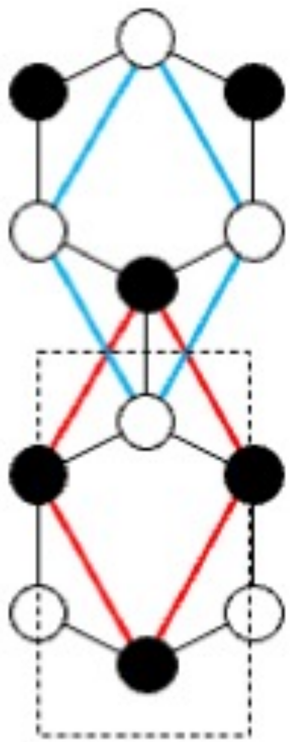



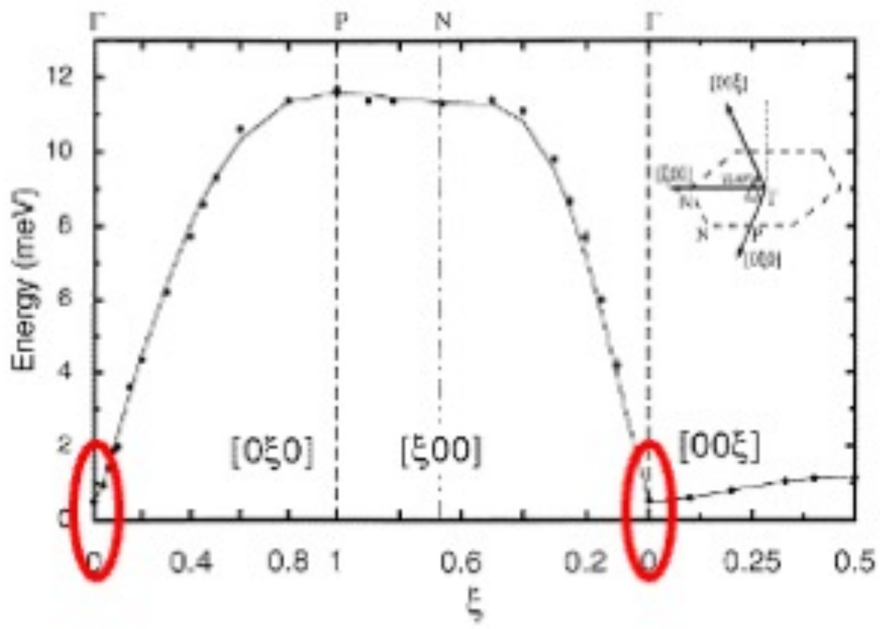

Periodicity is the same

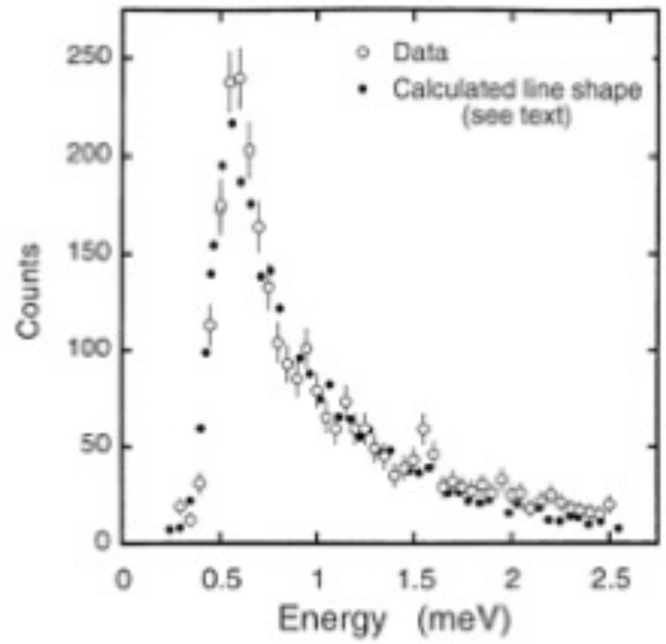
# The presence of gaps

## Spin waves in $MnPS_3$

A. R. Wildes *et al.*, JPCM **10** (1998) 6417



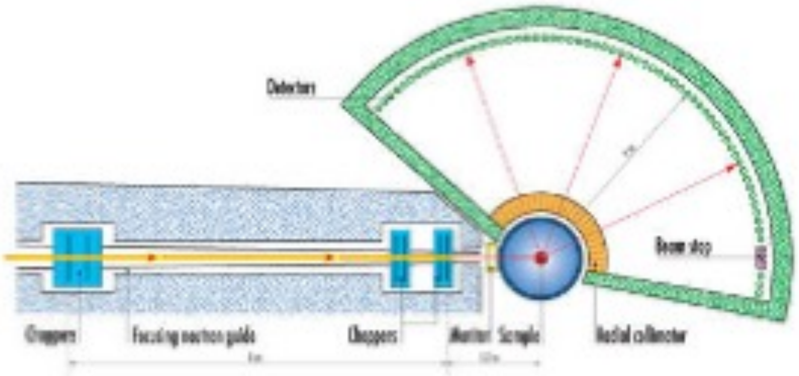


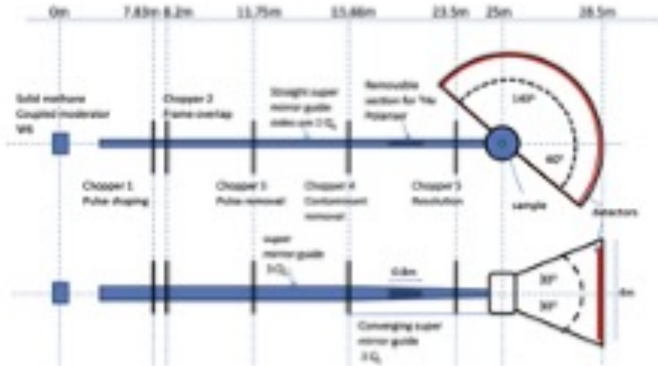

~260 s/pt

# Measuring single crystals: time-of-flight

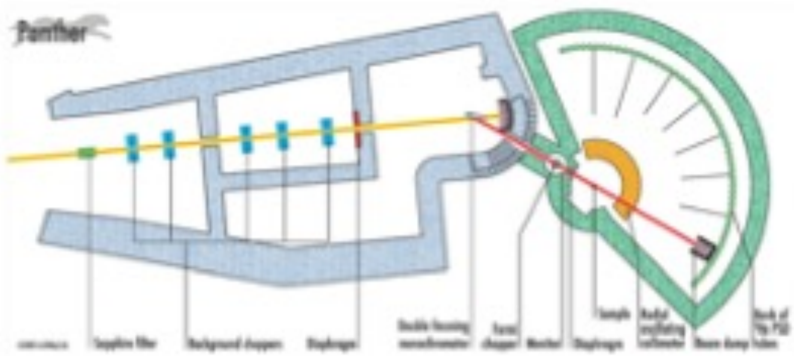
## IN5 @ ILL



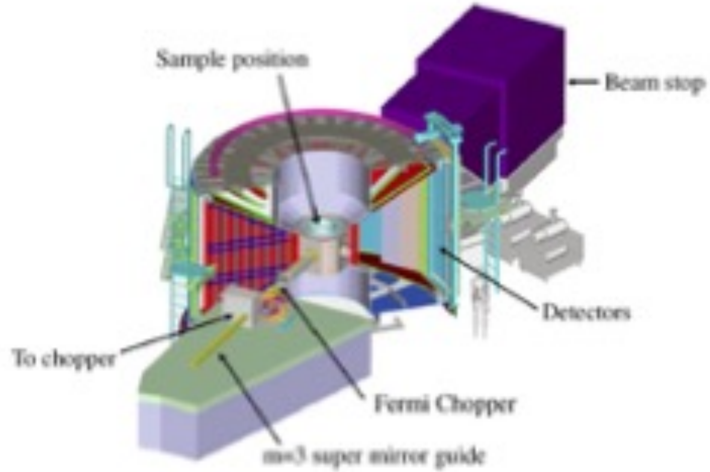

## LET @ ISIS



## PANTHER @ ILL

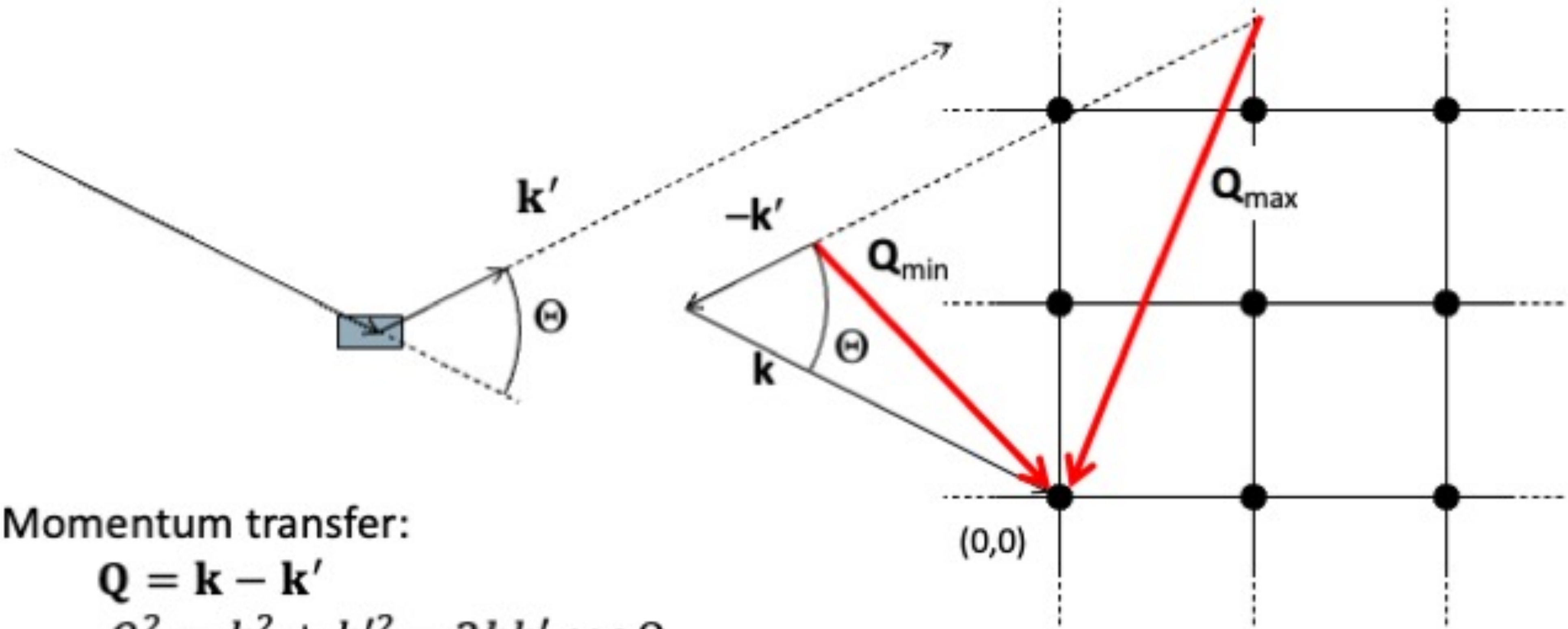


## MERLIN @ ISIS



## Inelastic scattering on single crystals

Doing it on a TOF instrument




Momentum transfer:

$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

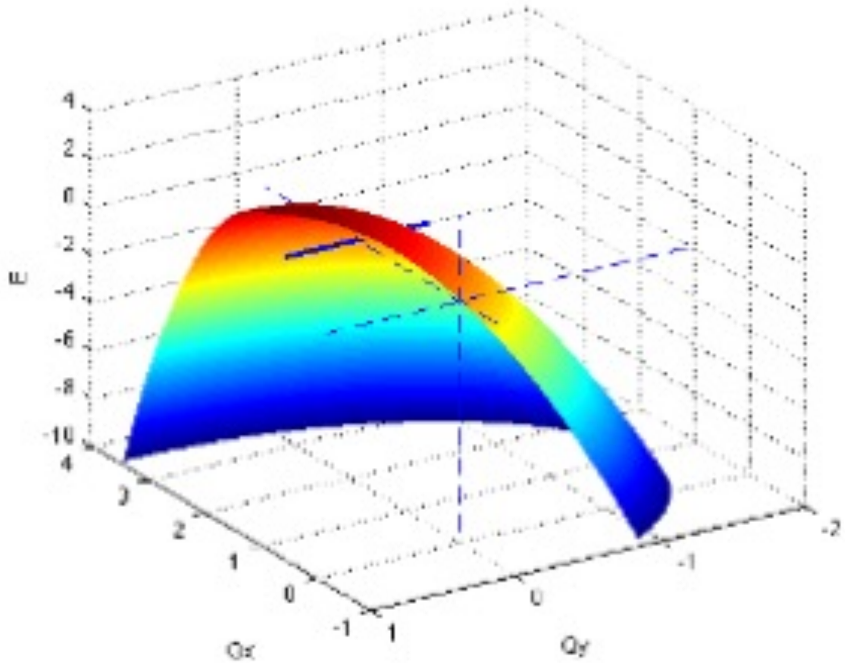
$$Q^2 = k^2 + k'^2 - 2kk' \cos\Theta$$

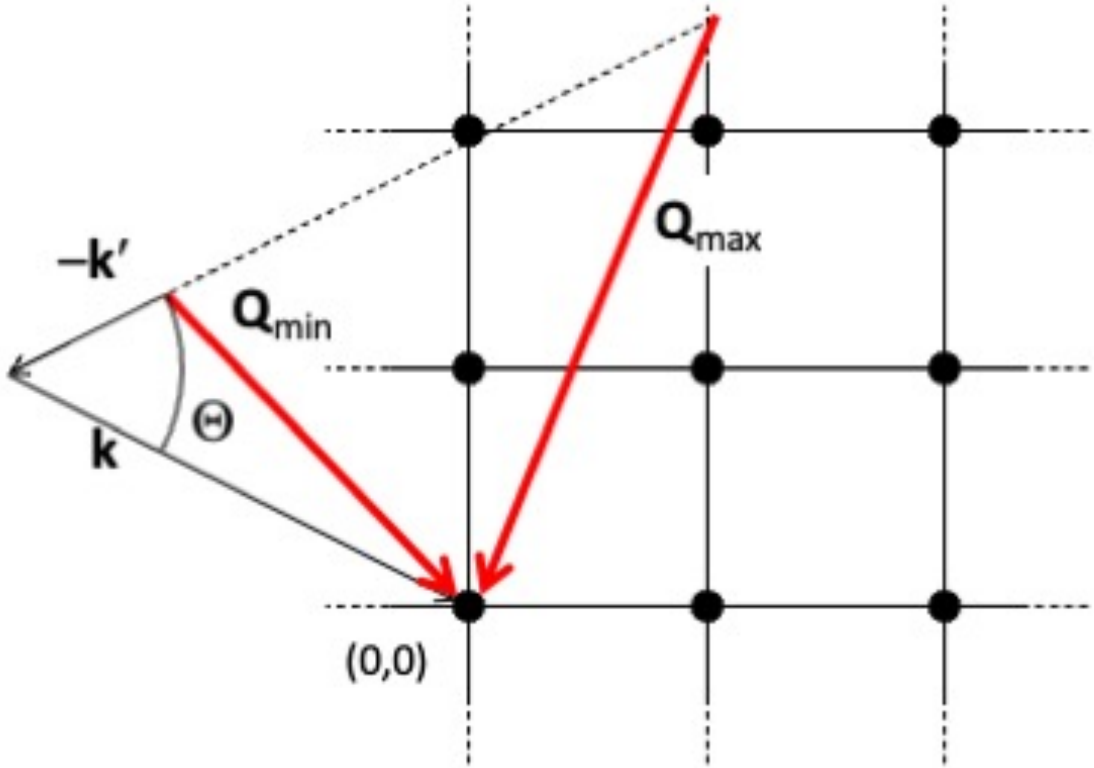
Energy transfer:

$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

## Inelastic scattering on single crystals

Doing it on a TOF instrument




Momentum transfer:

$$\mathbf{Q} = \mathbf{k} - \mathbf{k}'$$

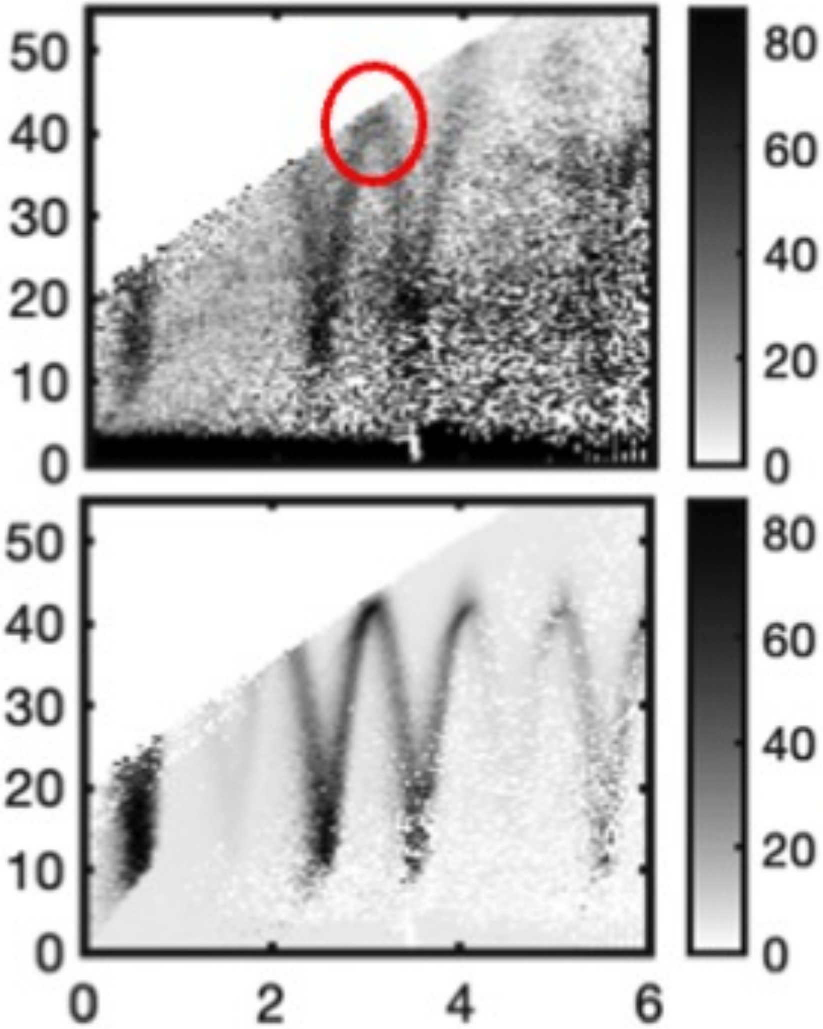
$$Q^2 = k^2 + k'^2 - 2kk' \cos\Theta$$

Energy transfer:

$$\Delta E = \hbar\omega = \frac{\hbar}{2m_n}(k^2 - k'^2)$$

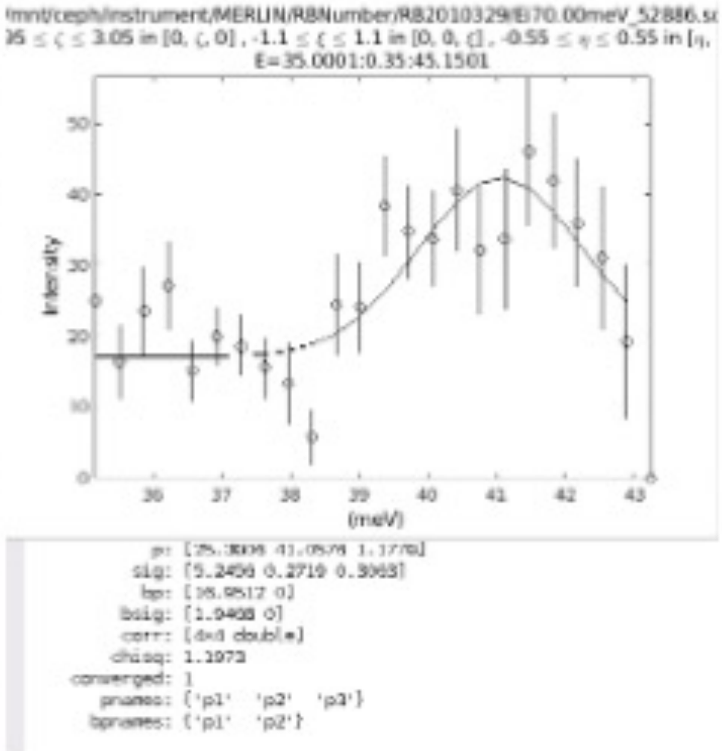
# HORACE scans

https://pace-neutrons.github.io/Horace/v4.0.0/


$$\sum_{h=\pm\frac{1}{2}} h,k,\ \sum_{l=0,\pm 1} l$$

3 – 4 days



# Polarised neutrons on single crystals

Spin-dependent potentials

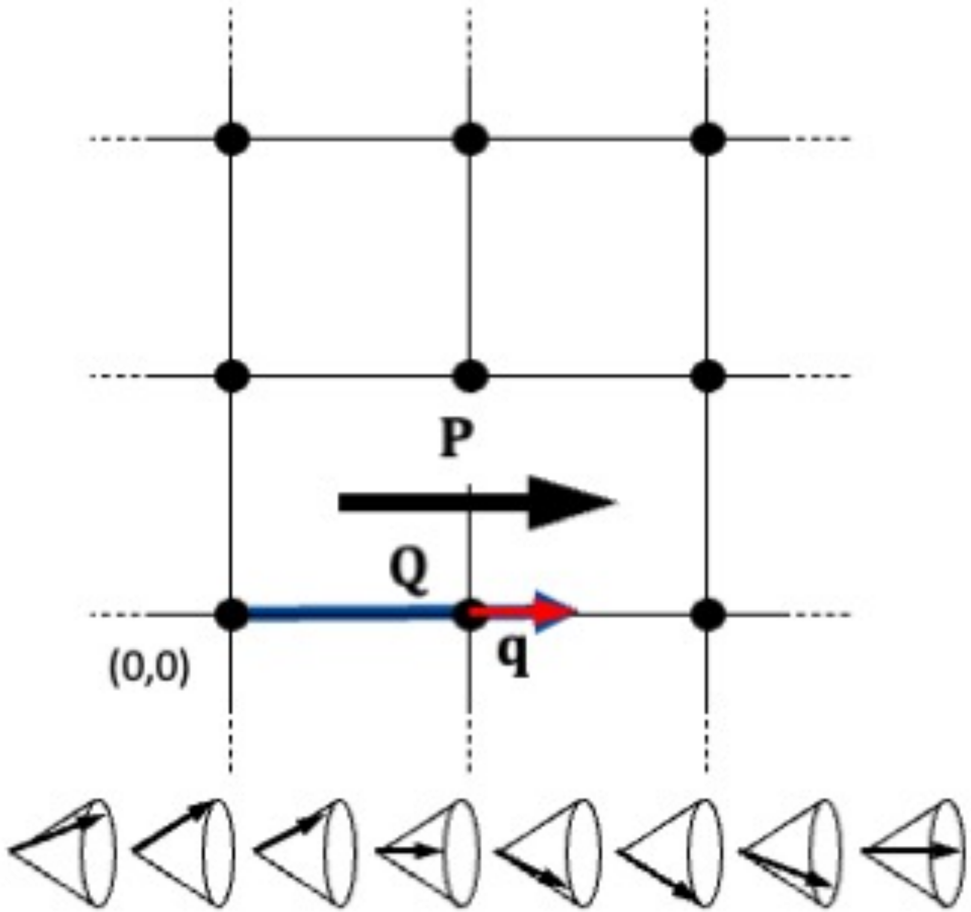
$$U^{++} = b - M_{\perp z} + BI_z$$

$$U^{--} = b + M_{\perp z} - BI_z$$

$$U^{+-} = -\left(M_{\perp x} + iM_{\perp y}\right) + B\left(I_x + iI_y\right)$$

$$U^{+-} = -\left(M_{\perp x} - iM_{\perp y}\right) + B\left(I_x - iI_y\right)$$

$z$ is the polarization axis, $\mathbf{P}$




If $\mathbf{P}||\mathbf{Q}$

$$M_{\perp z} = 0$$

Nuclear coherent scattering is only NSF ($\pm\,\pm$)

Magnetic scattering is only SF ($\pm\,\mp$)

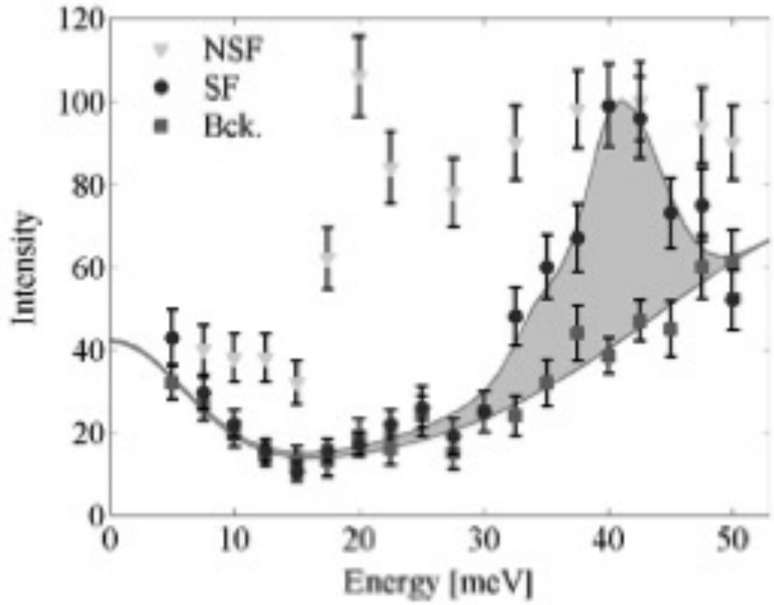
# Magnetism: **P** ||**Q**

Inelastic scattering

Separation of magnetic and nuclear contributions

$$U^{++} = b - M_{\perp z} + BI_z$$
$$U^{--} = b + M_{\perp z} - BI_z$$
$$U^{+-} = -\left(M_{\perp x} + iM_{\perp y}\right) + B\left(I_x + iI_y\right)$$
$$U^{-+} = -\left(M_{\perp x} - iM_{\perp y}\right) + B\left(I_x - iI_y\right)$$

$YBa_2Cu_3O_{6.85}$
**Q** = (1.5, 0.5, 1.7)




Regnault *et al.*, Physica B **335** (2003) 19

# Magnetism: P ||Q

Spin-dependent potentials

$$U^{++} = b - M_{\perp z} + BI_z$$

$$U^{--} = b + M_{\perp z} - BI_z$$

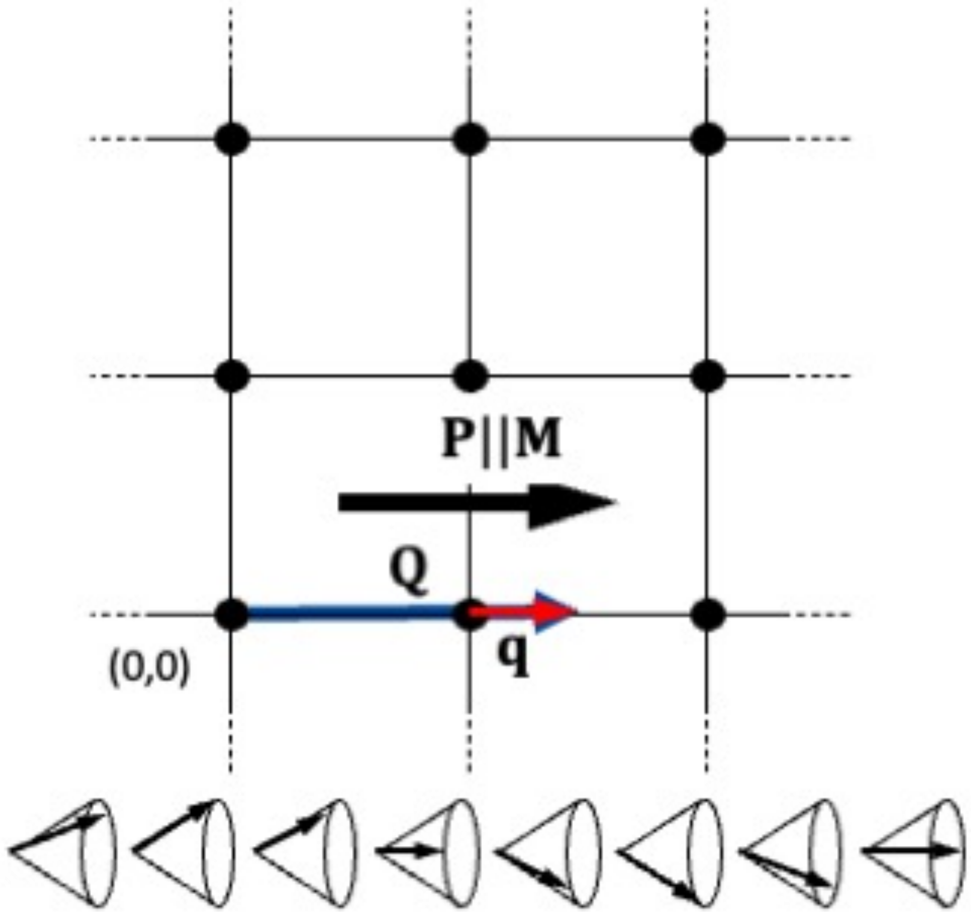
$$U^{+-} = -\left(M_{\perp x} + iM_{\perp y}\right) + B\left(I_x + iI_y\right)$$

$$U^{+-} = -\left(M_{\perp x} - iM_{\perp y}\right) + B\left(I_x - iI_y\right)$$

$z$ is the polarization axis, $\mathbf{P}$

If $\mathbf{P}||\mathbf{Q}$

$$M_{\perp z} = 0$$

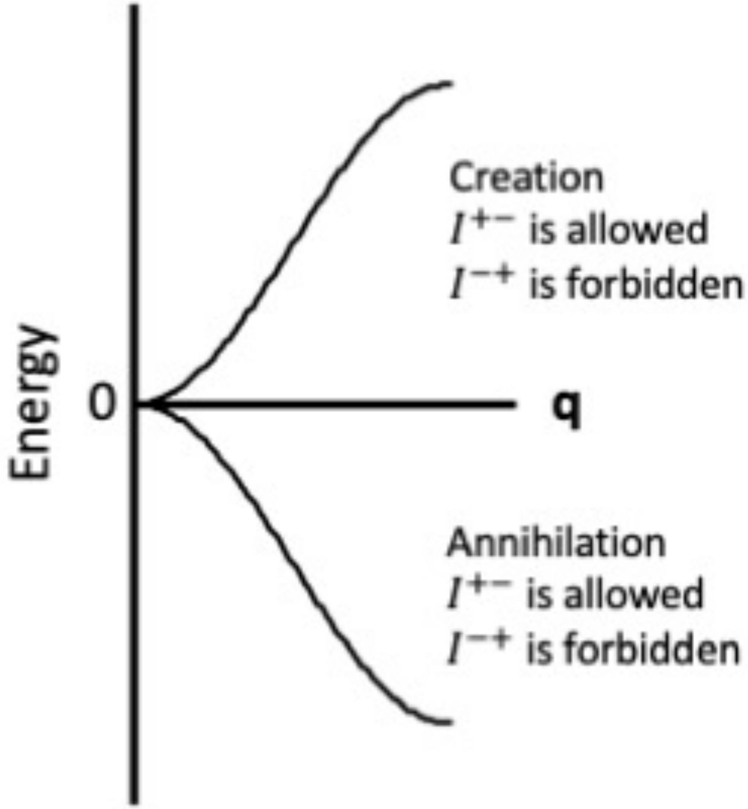



Nuclear coherent scattering is only NSF (± ±)
Magnetic scattering is only SF (± ∓)

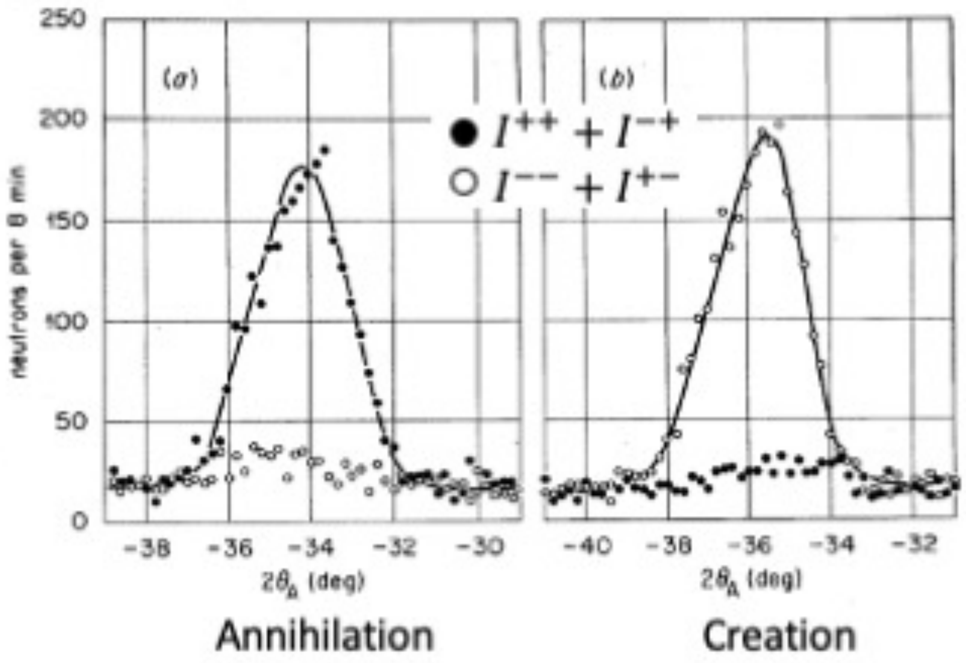
# Magnetism: **P** ||**Q**

Inelastic scattering

Creating or annihilating a ferromagnetic magnon requires a transfer of angular momentum.
The neutron spin must flip.




Spin wave scattering from $Fe_{2.5}Li_{0.5}O_4$



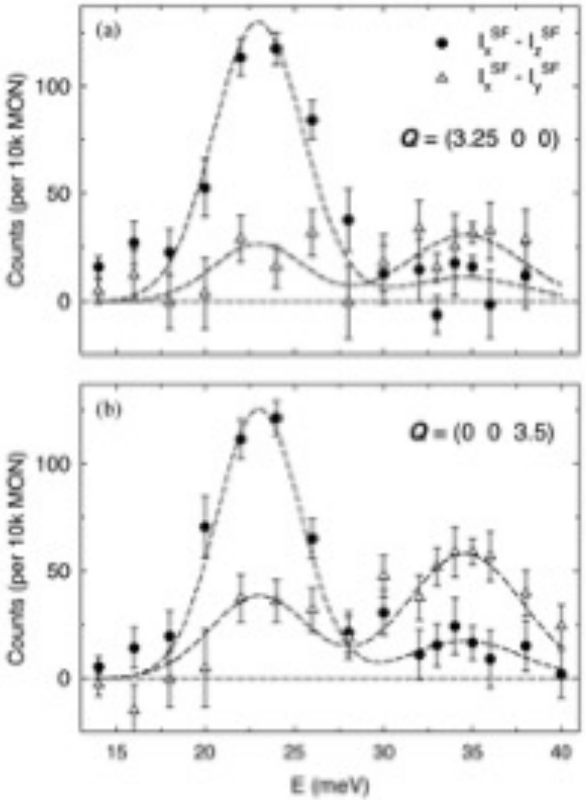

T. Riste *et al.*, PRL **20** (1968) 997

# Magnetism: Component separation

## CEF in CePtSn

B. Janoušová *et al.*, Physica B **335** (2003) 26

$$U^{++} = b - M_{\perp z} + BI_z$$
$$U^{--} = b + M_{\perp z} - BI_z$$
$$U^{+-} = -\left(M_{\perp x} + iM_{\perp y}\right) + B\left(I_x + iI_y\right)$$
$$U^{-+} = -\left(M_{\perp x} - iM_{\perp y}\right) + B\left(I_x - iI_y\right)$$



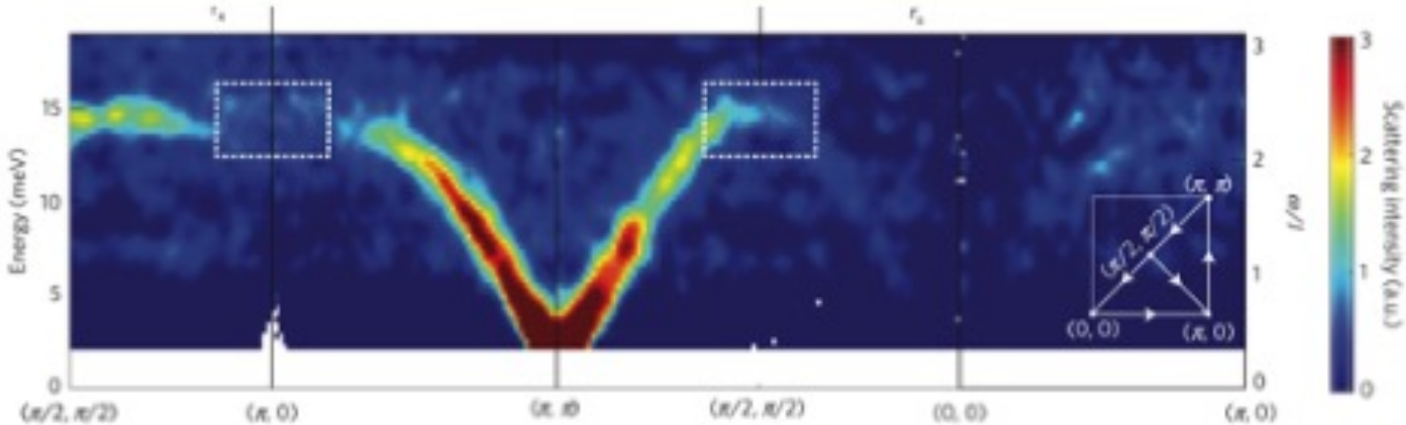

Neutron intensities (arbitrary scale)

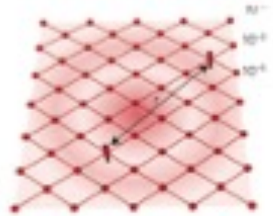
| | | |
|---|---|---|
| $E_{12} = 23.0\,\mathrm{meV}$ | $\lvert\langle 1\lvert J_a\rvert 2\rangle\rvert^2$ | 262(71) |
| | $\lvert\langle 1\lvert J_b\rvert 2\rangle\rvert^2$ | 777(54) |
| | $\lvert\langle 1\lvert J_c\rvert 2\rangle\rvert^2$ | 166(40) |
| $E_{13} = 34.6\,\mathrm{meV}$ | $\lvert\langle 1\lvert J_a\rvert 3\rangle\rvert^2$ | 474(69) |
| | $\lvert\langle 1\lvert J_b\rvert 3\rangle\rvert^2$ | 113(56) |
| | $\lvert\langle 1\lvert J_c\rvert 3\rangle\rvert^2$ | 245(32) |

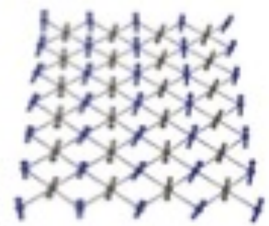
# Magnetism: Component separation

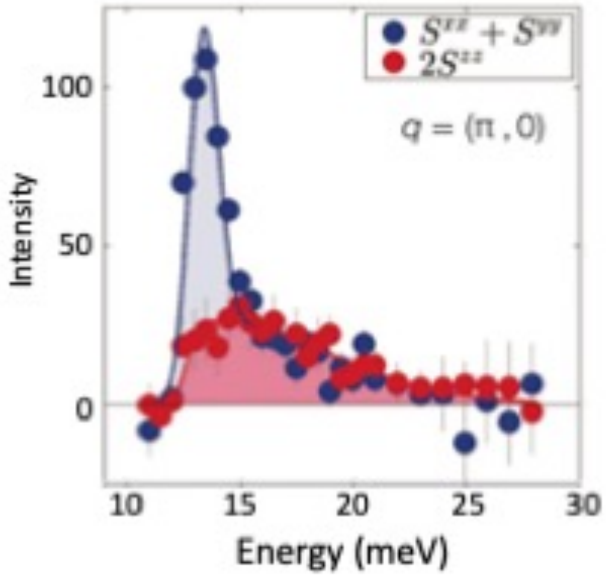
## Magnetic fluctuations in $Cu(DCOO)_2.4D_2O$

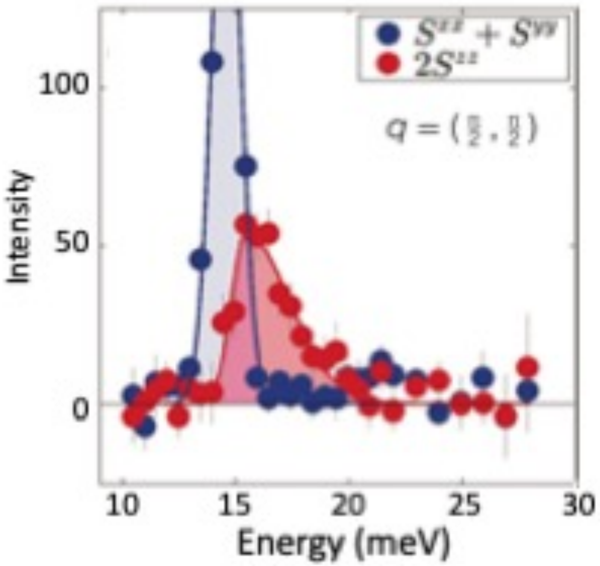
B. Dalla Piazza *et al.*, Nature Physics **11** (2015) 62







## Take home messages

- Work in reciprocal space!
- Neutrons see $\mathbf{M}_\perp$
- Neutrons have a form factor, $f(Q)$
- Polarized neutrons are good for magnetism