Inelastic neutron scattering

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Outline

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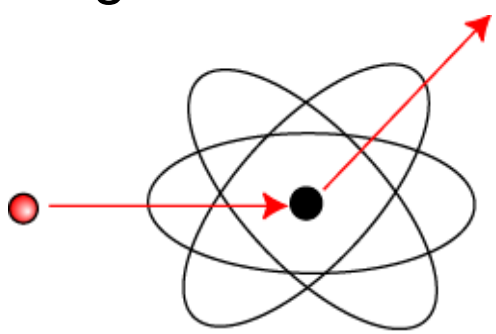
I. **Interaction of the neutron**
   A. Nuclear
   B. Magnetic
II. **General inelastic scattering**

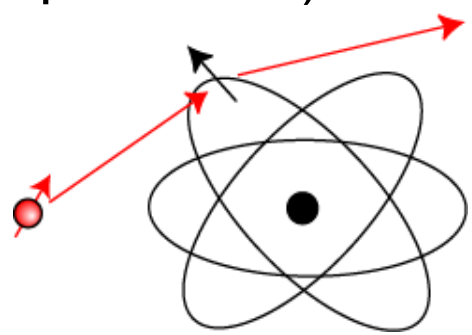
III. **Nuclear inelastic scattering**
   A. Correlation functions
   B. Examples
IV. **Magnetic inelastic scattering**
   A. Correlation functions
   B. Examples

Neutron interaction with matter

- **Properties of the neutron**
  - Mass $m_n = 1.675 \times 10^{-27}$ kg
  - Charge 0
  - Spin-1/2, magnetic moment $\mu_n = -1.913\ \mu_N$
- **Neutrons interact with…**
  - Nucleus
    - Crystal structure/excitations (eg. phonons)
  - Unpaired e^- via dipole scattering
    - Magnetic structure/excitations (eg. spin waves)



Nuclear scattering



Magnetic dipole scattering

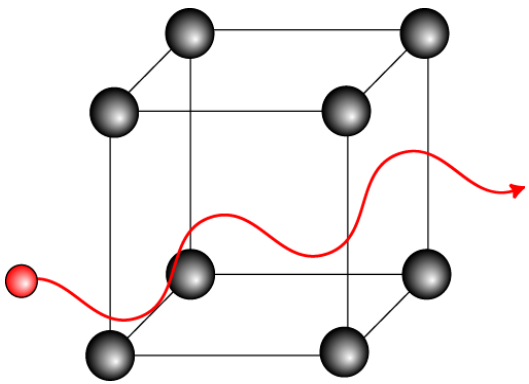
Wavelength-energy relations

- **Neutron as a wave …**
  - Energy (E), velocity (v), wavenumber (k), wavelength (λ)

$$k = \frac{m_n \mathrm{v}}{\hbar} = \frac{2\pi}{\lambda}$$

$$E = \frac{\hbar^2 k^2}{2m_n} = \frac{\hbar^2}{2m_n}\left(\frac{2\pi}{\lambda}\right)^2 = \frac{81.81\,\mathrm{meV}\cdot\mathrm{Å}^2}{\lambda^2}$$

$$E = k_B T = \left(0.08617\,\mathrm{meV}\cdot\mathrm{K}^{-1}\right)T$$



λ ~ interatomic spacing → E ~ excitations in condensed matter

	Energy (meV)	Temperature (K)	Wavelength (Å)
Cold	0.1 – 10	1 – 120	4 – 30
Thermal	5 – 100	60 – 1000	1 – 4
Hot	100 – 500	1000 – 6000	0.4 – 1

Dynamical (time) scales

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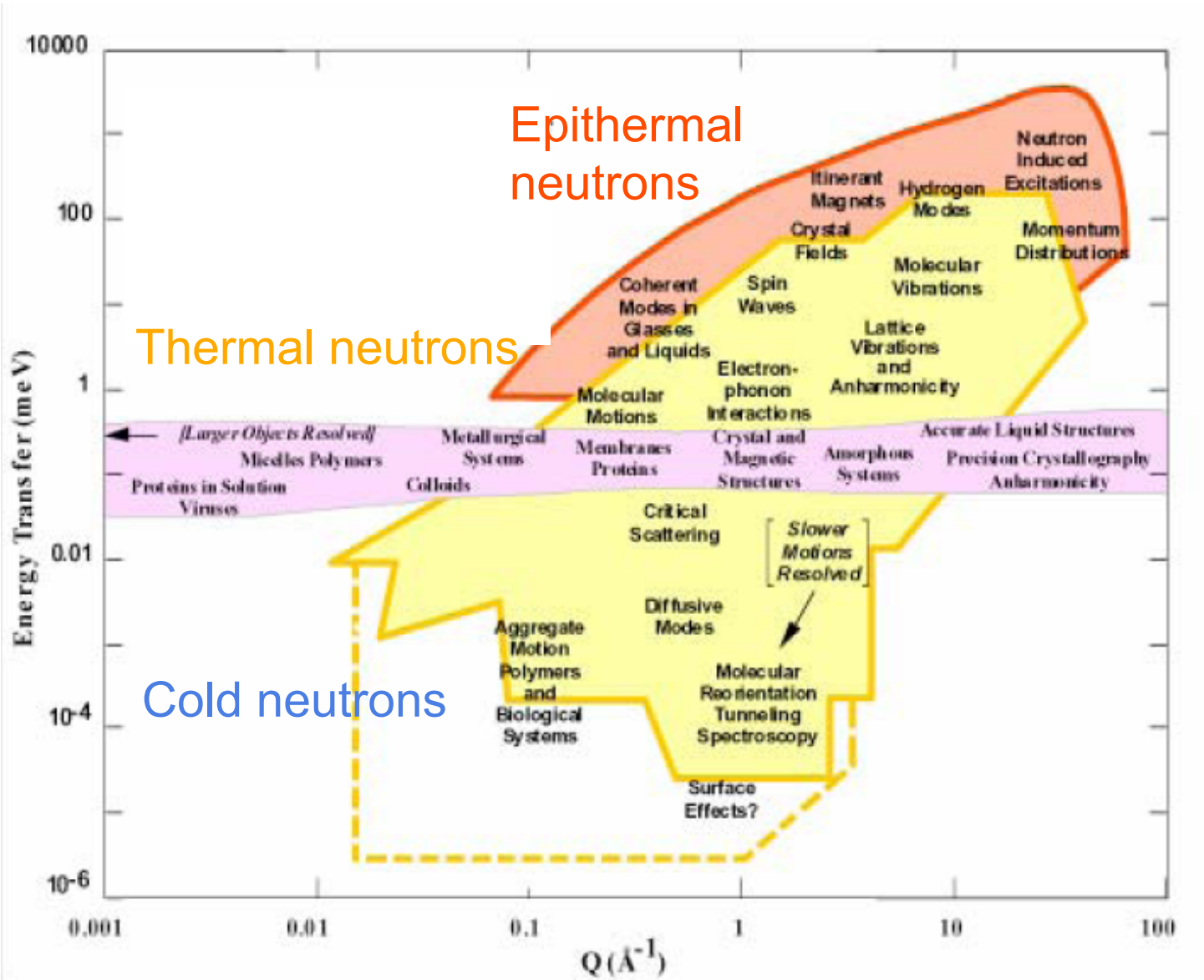
Faster processes

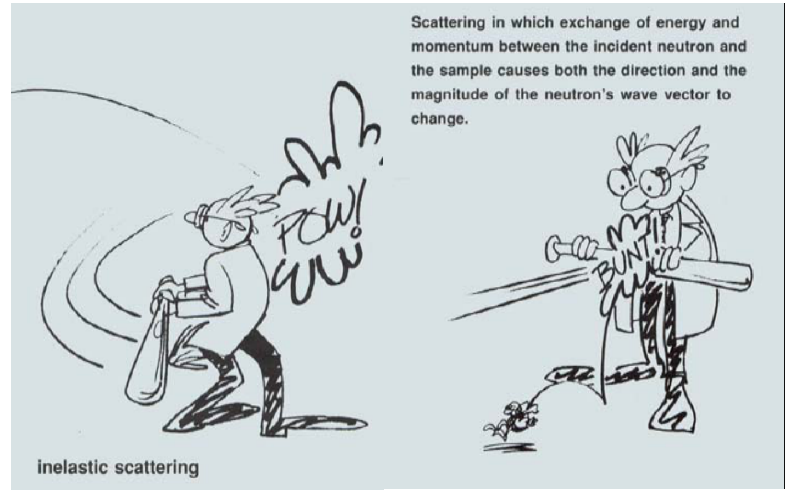


Elastic scattering

Shorter length scales

Inelastic scattering

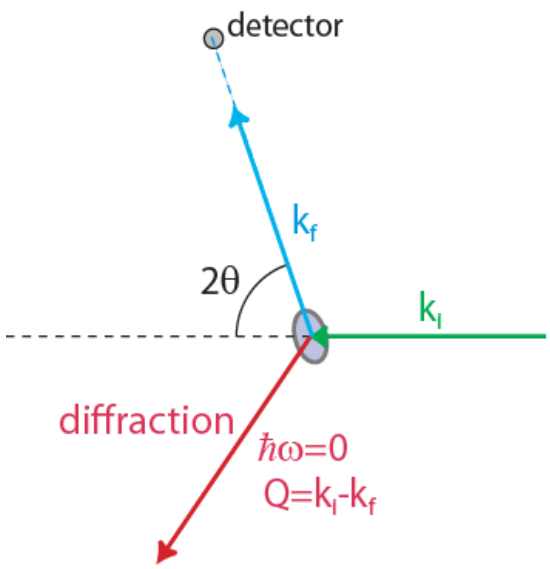
- Scattering process that changes the energy of the neutron



- Conservation of energy and momentum

$$\hbar\omega = E_i - E_f \qquad \mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$

- Scattering triangle



Elastic scattering

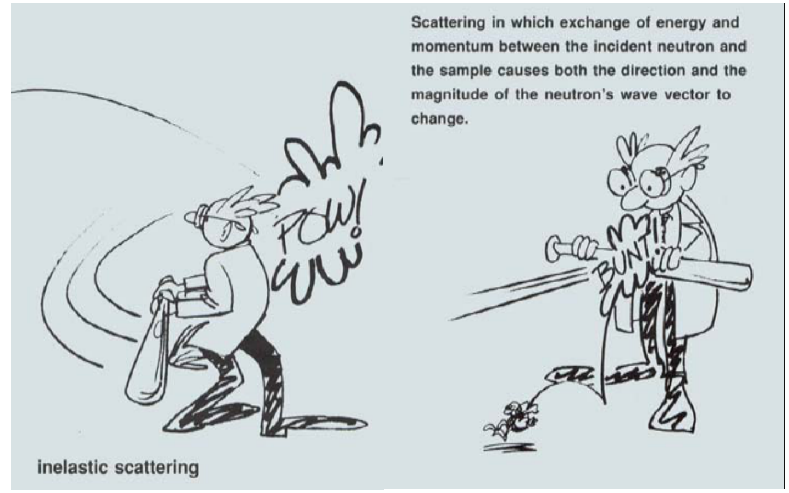
$$\hbar\omega = 0$$

$$|\mathbf{k}_i| = |\mathbf{k}_f|$$

$$Q = 2k_i \sin\theta$$

Inelastic scattering

- Scattering process that changes the energy of the neutron



- Conservation of energy and momentum

$$\hbar\omega = E_i - E_f \qquad \mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$

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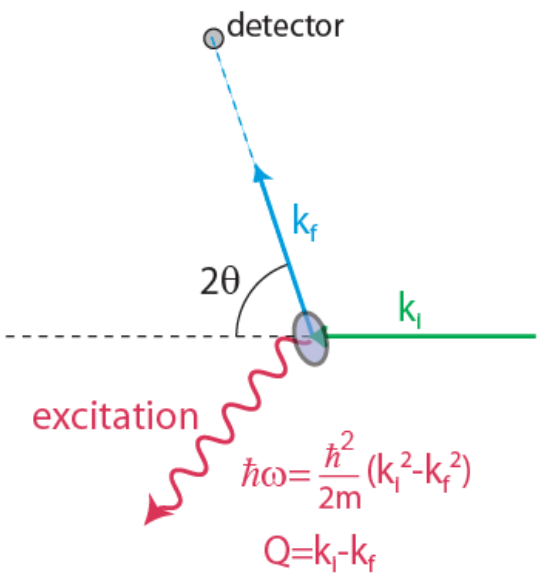

<u>Inelastic scattering</u>

$$\hbar\omega > 0$$

$$|\mathbf{k}_i| \neq |\mathbf{k}_f|$$

$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$

Nuclear correlation functions

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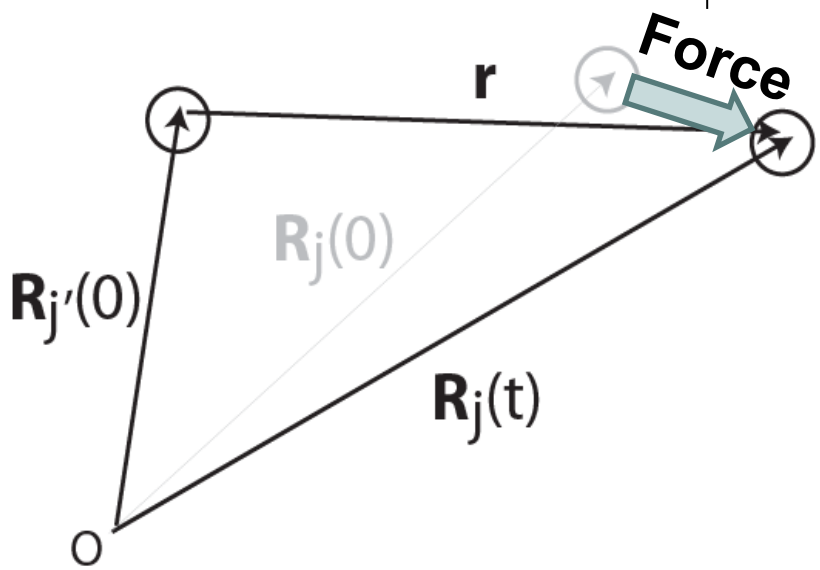

Pair correlation function

$$G(\mathbf{r},t) = \frac{1}{N}\int \sum_{jj'} \delta\left(\mathbf{r}' - \mathbf{R}_{j'}(0)\right)\delta\left(\mathbf{r}' + \mathbf{r} - \mathbf{R}_j(t)\right) d\mathbf{r}'$$

Intermediate function

$$I(\mathbf{Q},t) = \int G(\mathbf{r},t) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} = \frac{1}{N}\sum_{jj'} \exp\left(-i\mathbf{Q}\cdot\mathbf{R}_{j'}(0)\right)\exp\left(i\mathbf{Q}\cdot\mathbf{R}_j(t)\right)$$

Scattering function

$$S(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar}\int I(\mathbf{Q},t) e^{-i\omega t} dt$$

Differential scattering cross-section

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{\sigma_{scat}}{4\pi}\frac{k_f}{k_i} NS(\mathbf{Q},\omega)$$

Nuclear (lattice) excitations

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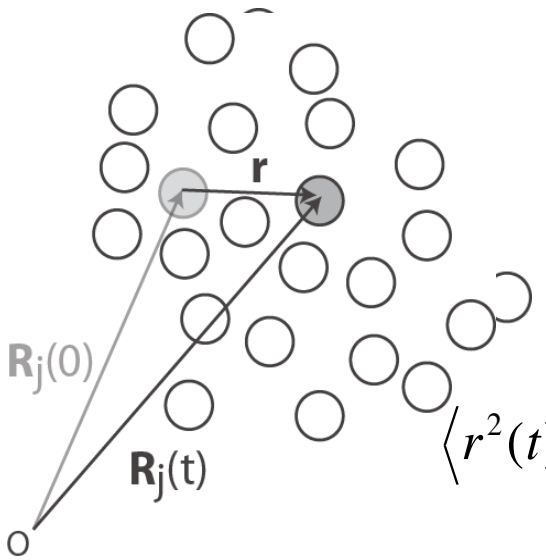
Neutron scattering measures simultaneously the wavevector and energy of **collective excitations** → dispersion relation, $\omega(\mathbf{q})$
In addition, **local excitations** can of course be observed

- **Commonly studied excitations**
  - Phonons
  - Librations and vibrations in molecules
  - Diffusion
  - Collective modes in glasses and liquids
- **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)

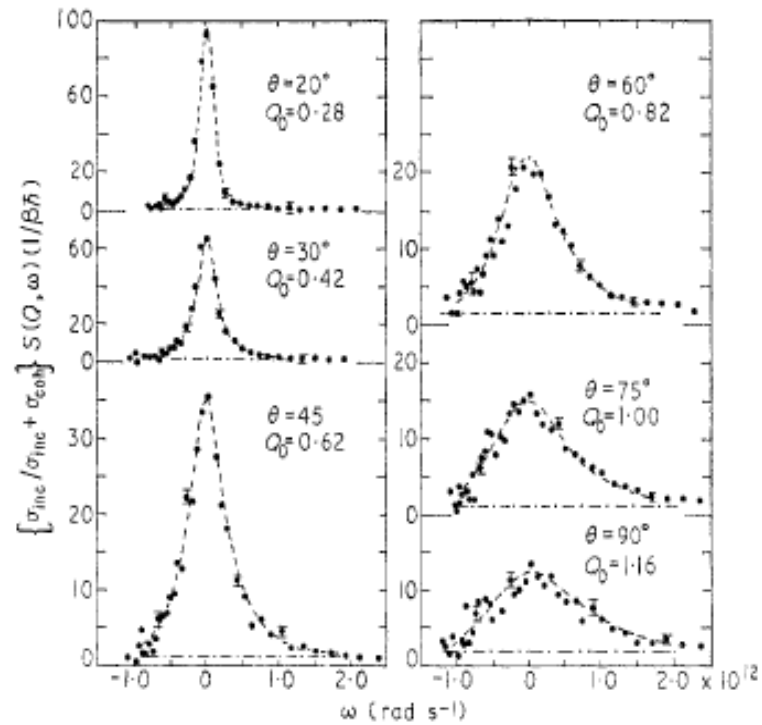
Atomic diffusion

For long times compared to the collision time, atom diffuses



$$\left\langle r^2(t)\right\rangle \approx 6Dt$$

Liquid Na



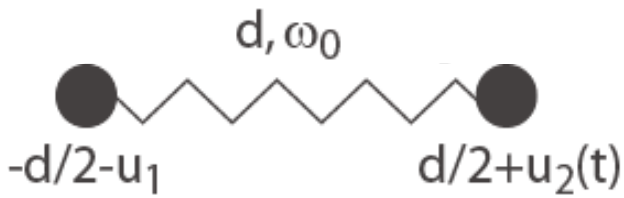
Cocking, *J. Phys. C* **2**, 2047 (1969)..

Auto-correlation function

$$G_s(r,t) = \left\{6\pi\left\langle r^2(t)\right\rangle\right\}^{-3/2} \exp\left(-\frac{r^2}{6\left\langle r^2(t)\right\rangle}\right)$$

$$S(Q,\omega) = \frac{1}{\pi\hbar}\exp\left(\frac{\hbar\omega}{2k_BT}\right)\frac{DQ^2}{\omega^2 + (DQ^2)^2}$$

Diatomic molecule



$$R_1(0) = -\frac{d}{2} - u(0)$$

$$R_2(t) = \frac{d}{2} + u(t)$$

$$u(t) = \sqrt{\frac{\hbar}{2M\omega_0}}\left[\hat{a}e^{-i\omega_0 t} + \hat{a}^{+}e^{i\omega_0 t}\right]$$

$$S(Q,\omega) = \frac{1}{2\pi\hbar N}\int_{-\infty}^{\infty} dt e^{-i\omega t}\sum_{jj'}\left\langle \exp\left(-iQR_{j'}(0)\right)\exp\left(iQR_j(t)\right)\right\rangle$$

$$S(Q,\omega) = \frac{1}{2\pi\hbar}e^{-Q^2\langle u^2\rangle}\int_{-\infty}^{\infty} dt e^{-i\omega t}\left[e^{Q^2\langle u(0)u(t)\rangle} + \cos(Qd)e^{-Q^2\langle u(0)u(t)\rangle}\right]$$

SHO correlation functions

$$\langle u^2\rangle = \frac{\hbar}{2M\omega_0}\coth(\hbar\omega_0\beta)$$

$$\langle u(0)u(t)\rangle = \frac{\hbar}{2M\omega_0}\left[\left(1+\frac{1}{e^{\hbar\omega_0\beta}-1}\right)e^{i\omega_0 t} + \frac{1}{e^{\hbar\omega_0\beta}-1}e^{-i\omega_0 t}\right] = \frac{\hbar}{2M\omega_0}\frac{\cosh\omega_0(it+\hbar\beta/2)}{\sinh(\hbar\omega_0\beta/2)}$$

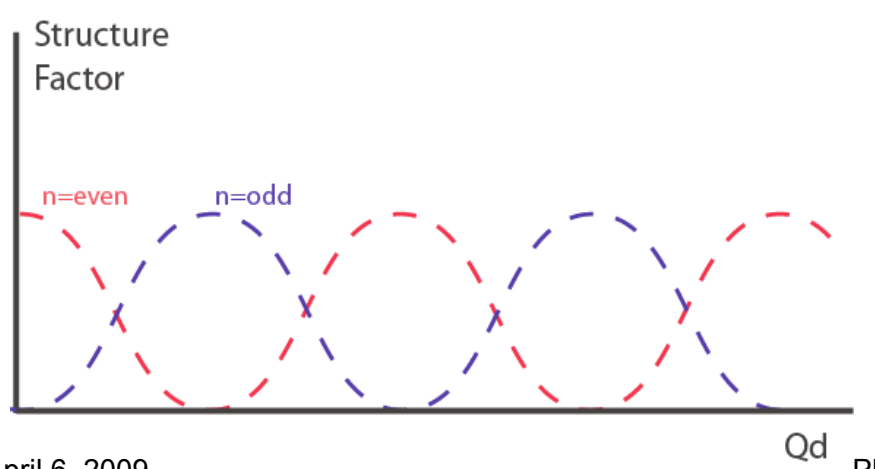
$$e^{y\cosh x} = \sum_{n=-\infty}^{\infty} I_n(y)e^{nx} \qquad y = \frac{\hbar Q^2}{2M\omega_0}\operatorname{csch}\left(\frac{1}{2}\hbar\omega_0\beta\right)$$

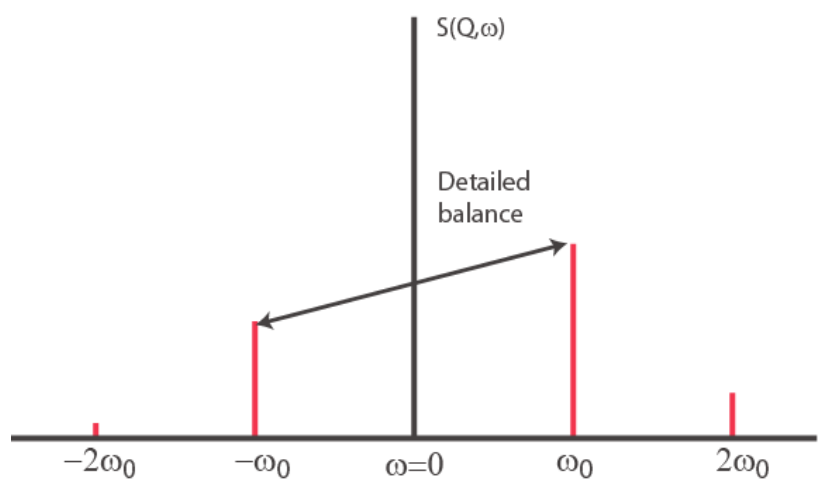
Diatomic molecule

$$S(Q,\omega) = \underbrace{\frac{1}{\hbar} e^{-Q^2\langle u^2\rangle}}_{\text{Debye-Waller}} \underbrace{e^{\hbar\omega\beta/2}}_{\text{Detailed balance}} \underbrace{\sum_{n=-\infty}^{\infty}\left[1+(-1)^n\cos(Qd)\right]}_{\text{Structure factor}} \underbrace{I_n(y)}_{\text{Form factor}} \underbrace{\delta(\omega - n\omega_0)}_{\text{Excitation energy}}$$

Small-amplitude approximation, y small

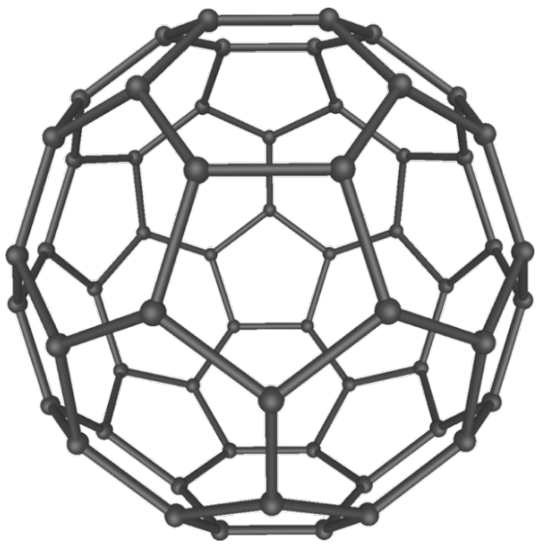
$$S(Q,\omega) \approx \frac{1}{\hbar} e^{-Q^2\langle u^2\rangle} \left\{ \underbrace{\left[1+\cos(Qd)\right]\delta(\omega)}_{\text{Elastic scattering}} + \underbrace{\frac{Q^2}{2M\omega_0}\left[1-\cos(Qd)\right]\left[\left(\frac{1}{2} \pm \frac{1}{2} + n(\omega_0)\right)\delta(\omega \mp \omega_0)\right]}_{\text{One quantum}} + \underbrace{\ldots}_{\text{Multi-quanta}} \right\}$$



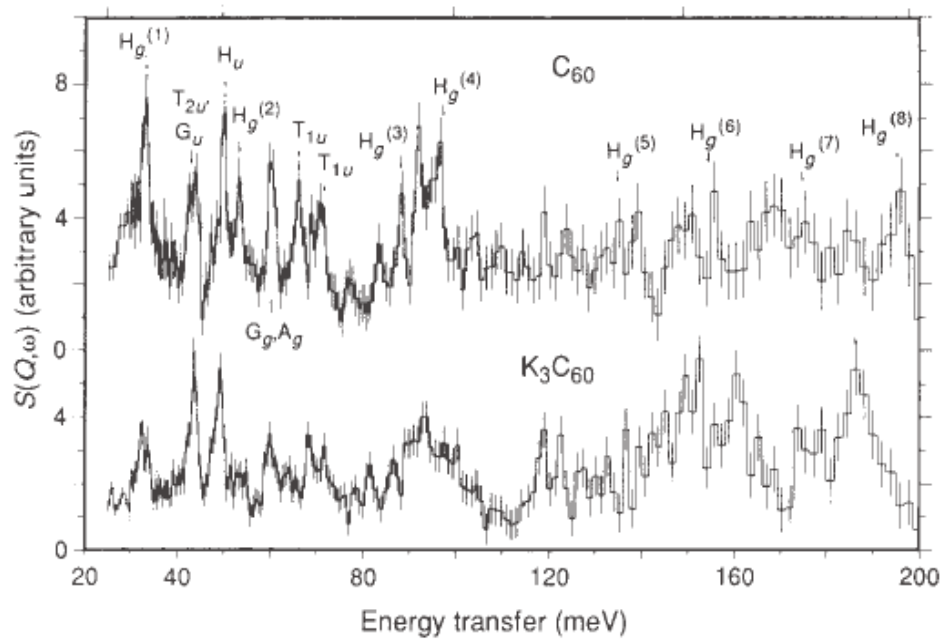


Molecular vibrations

- Large molecule, many normal modes
- Harmonic vibrations can determine interatomic potentials



C60 molecule



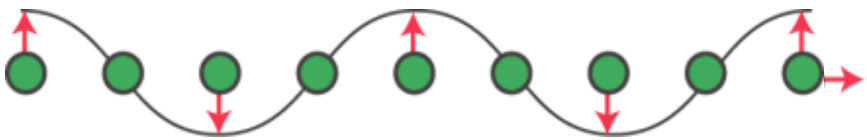
Prassides *et al., Nature* **354**, 462 (1991).

Phonons

- Normal modes in periodic crystal → wavevector

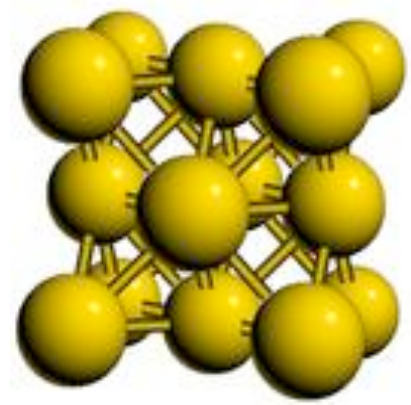
$$\mathbf{u}(l,t) = \frac{1}{\sqrt{NM}} \sum_{j\mathbf{q}} \boldsymbol{\varepsilon}_j(\mathbf{q}) \exp(i\mathbf{q}\cdot\mathbf{l}) \hat{B}(\mathbf{q}j,t)$$

- Energy of phonon depends on **q** and polarization

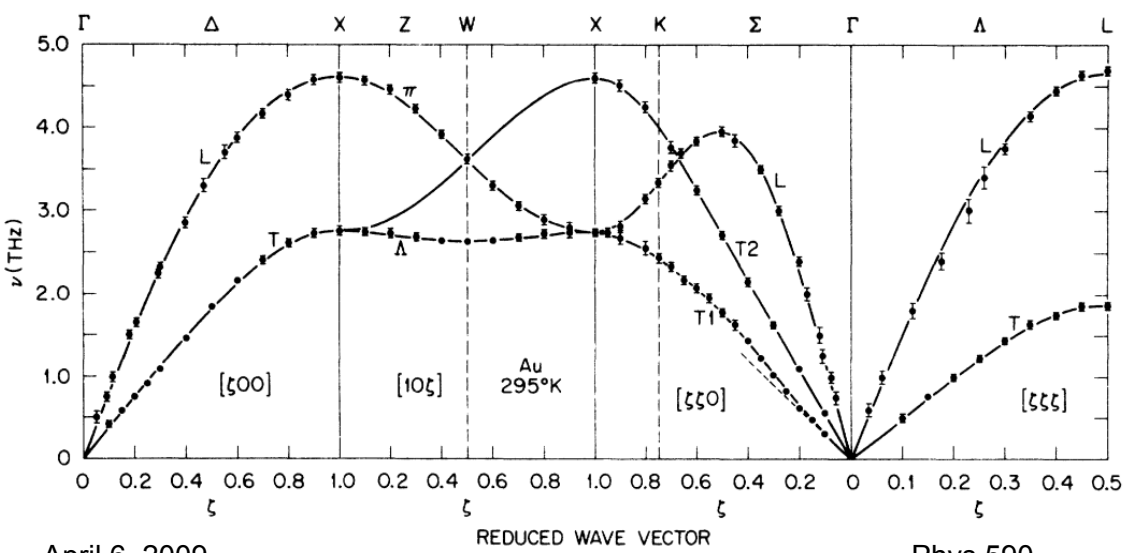


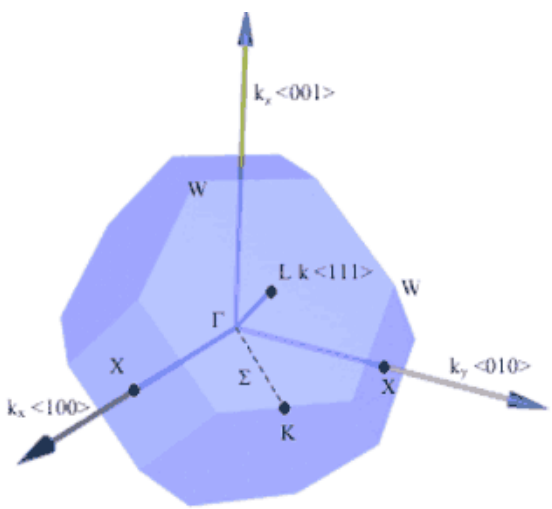
Longitudinal mode

Transverse mode



FCC structure





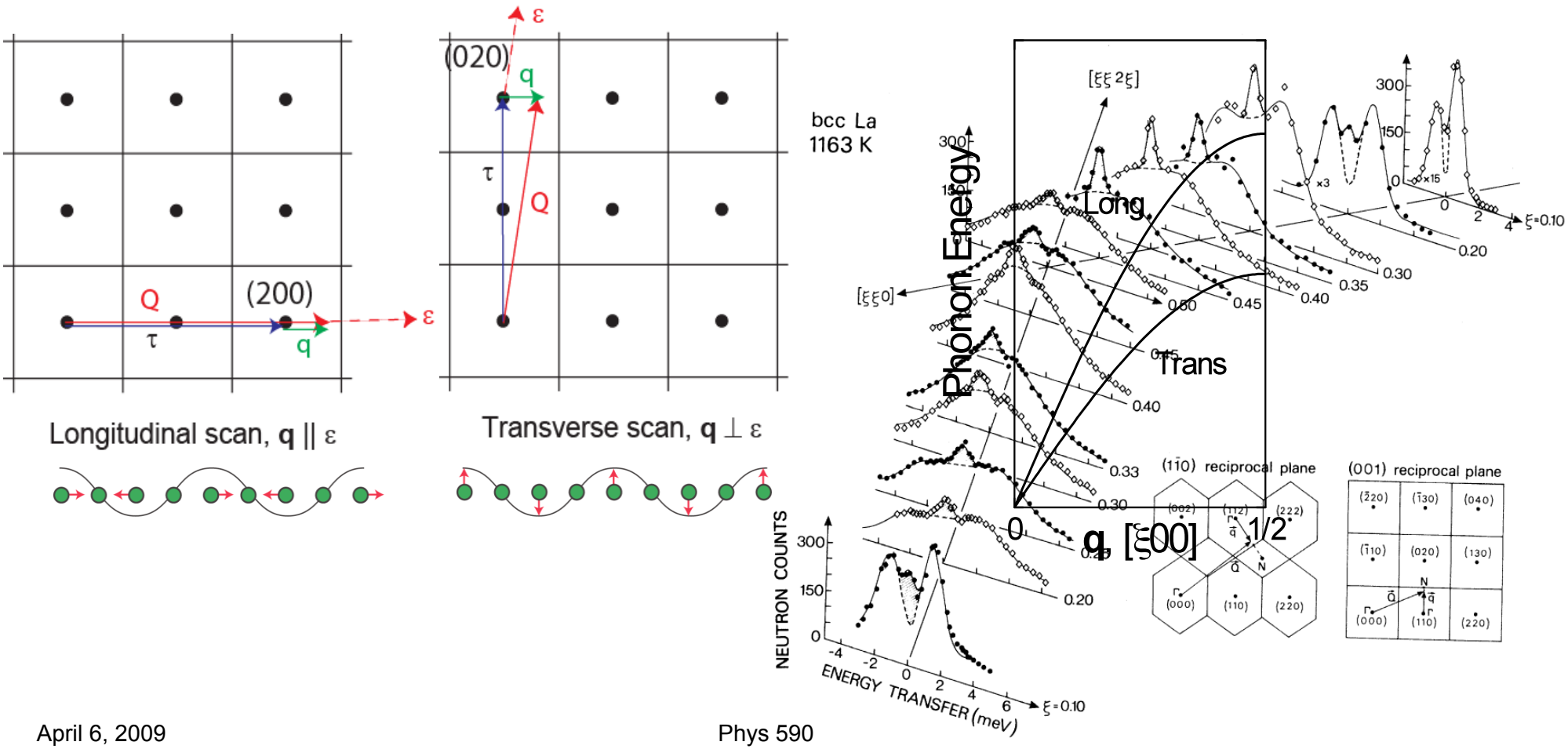
FCC Brillouin zone

Lynn, *et al.*, *Phys. Rev. B* **8**, 3493 (1973).

Phonon intensities

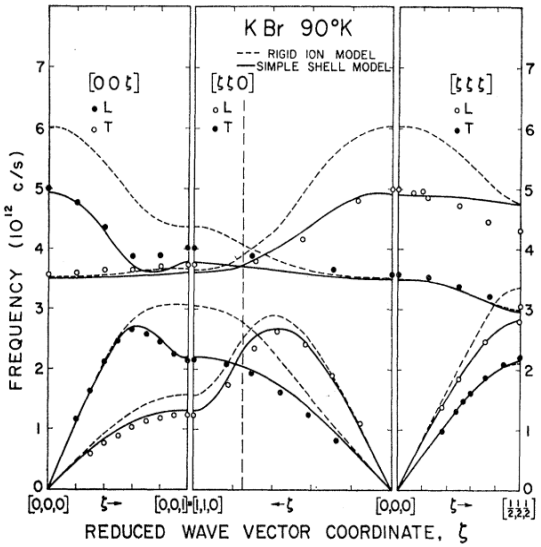
Structure (polarization) factor

$$S_{1+}(\mathbf{Q},\omega) = \frac{1}{2NM} e^{-Q^2\langle u^2\rangle} \sum_{j\mathbf{q}} \frac{\left|\mathbf{Q}\cdot\boldsymbol{\varepsilon}_j(\mathbf{q})\right|^2}{\omega_j(\mathbf{q})} \left(1+n(\omega)\right)\delta(\mathbf{Q}-\mathbf{q}-\boldsymbol{\tau})\delta\left(\omega-\omega_j(\mathbf{q})\right)$$

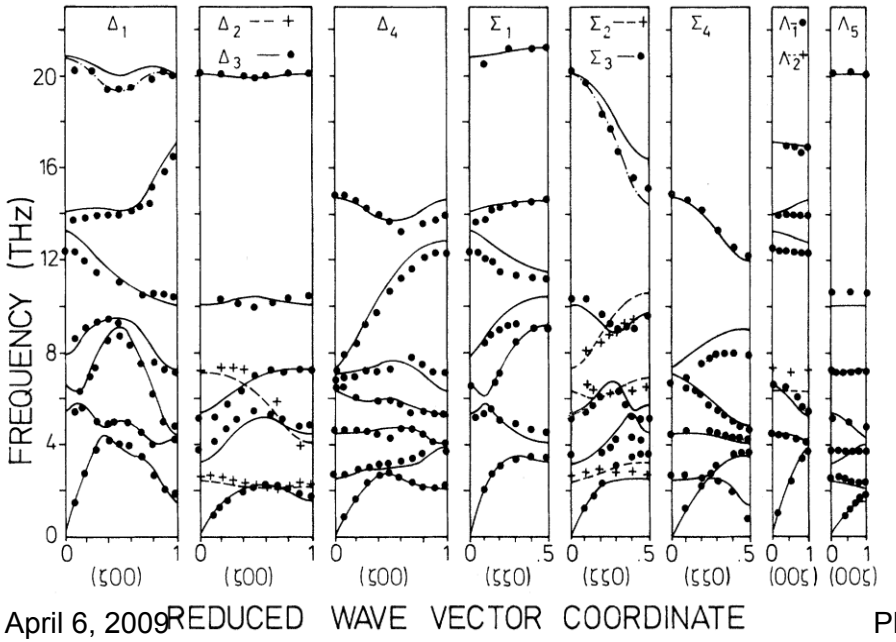


Guthoff *et al., Phys. Rev. B* **47**, 2563 (1993).

More complicated structures

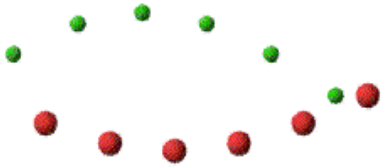


Woods, *et al., Phys. Rev.* **131**, 1025 (1963).

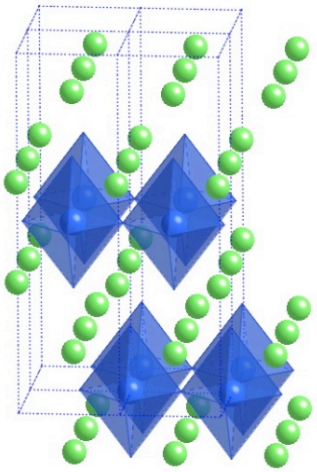


Chaplot, *et al., Phys. Rev.* B **52**, 7230(1995).

Acoustic phonon



Optical phonon



La_2CuO_4

Spin correlation functions

$$\frac{d^2\sigma}{d\Omega dE_f} = \underbrace{\frac{k_f}{k_i}\left[\frac{1}{2}\gamma r_0 g F(Q)\right]^2}_{\text{Scattering cross-section}} \underbrace{\sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right)}_{\text{Dipole interaction}} \sum_{jj'} e^{i\mathbf{Q}\cdot(\mathbf{R}_{j'} - \mathbf{R}_j)} \frac{1}{2\pi\hbar}\int dt e^{-i\omega t} \underbrace{\left\langle S_j^\alpha(0) S_{j'}^\beta(t)\right\rangle}_{\text{Spin-spin correlation function}}$$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right|_{inel} = \frac{k_f}{k_i}\left[\frac{1}{2}\gamma r_0 g F(Q)\right]^2 \sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right)\left(1 - e^{\hbar\omega/kT}\right)^{-1} \frac{1}{\pi(g\mu_B)^2}\mathrm{Im}\left\{\chi^{\alpha\beta}(\mathbf{Q},\omega)\right\}$$

The cross-section is proportional to the magnetic susceptibility,
i.e. it is the response of the system to spatially & time varying magnetic field

Spin excitations

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

- **Magnetic inelastic scattering can tell us about**
  - Exchange interactions
  - Single-ion and exchange anisotropy (determine Hamiltonian)
  - Phase transitions & critical phenomena
  - Quantum critical scaling of magnetic fluctuations
  - Other electronic energy scales (eg. CF & SO)
  - Interactions (eg. spin-phonon coupling)

Paramagnetic scattering

$$\left\langle S_j^\alpha S_{j'}^\beta \right\rangle = 0 \; \left(j \neq j' \right)$$

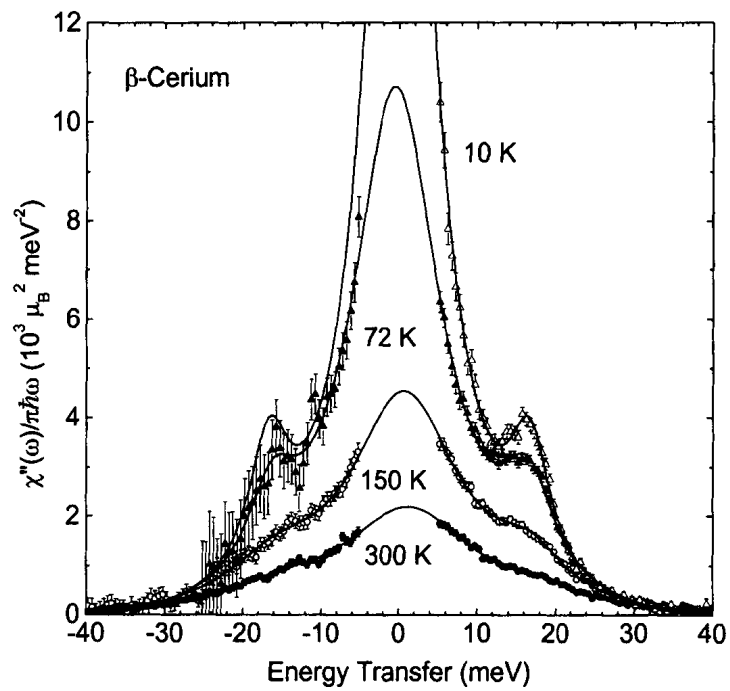
Single ion scattering

$$\left\langle S_j^z(0) S_j^z(t) \right\rangle = \left\langle \left(S_j^z \right)^2 \right\rangle e^{-\Gamma t} = \frac{1}{3} \left\langle \left(\mathbf{S}_j \right)^2 \right\rangle e^{-\Gamma t} = \frac{1}{3} S(S+1) e^{-\Gamma t}$$

$$\frac{\mathrm{Im}\left\{ \chi^{zz}(0,\omega) \right\}}{\pi \hbar \omega} = \frac{g^2 S(S+1) \mu_B^2}{3 k_B T} \frac{1}{\pi} \frac{\Gamma}{\Gamma^2 + (\hbar\omega)^2}$$

- Inverse width, $1/\Gamma$, gives relaxation time
- Note crystal field excitation

$$\chi_0 = \int_{-\infty}^{\infty} \frac{\mathrm{Im}\left\{ \chi^{zz}(0,\omega) \right\}}{\pi \hbar \omega} d\omega = \frac{g^2 S(S+1) \mu_B^2}{3 k_B T}$$



McQueeney *et al., Phil. Mag*. B **81**, 675 (2001).

Spin waves

$$H = -\sum_{<i,j>} J_{ij} \mathbf{S}_{\mathrm{i}} \cdot \mathbf{S}_{\mathrm{j}}$$

Heisenberg Hamiltonian

Linear spin wave theory

$$S^z \approx S$$

$$S^{\pm} = S^x \pm iS^y$$

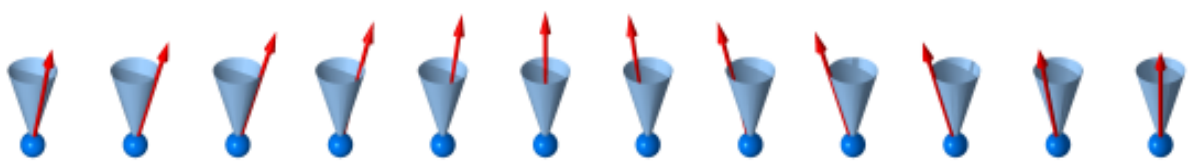
$$S_j^+(t) = \frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{Q}\cdot\mathbf{R}_j} S_{\mathbf{q}}^+(t)$$

$$\left\langle S_j^{\alpha}(0) S_{j'}^{\beta}(t) \right\rangle = \left\langle S_j^{-}(0) S_{j'}^{+}(t) \right\rangle$$

For a simple ferromagnet

$$\left\langle S_{\mathbf{q}}^{-}(0) S_{\mathbf{q}}^{+}(t) \right\rangle = \frac{S}{2N}\left(1 - e^{-\hbar\omega/kT}\right)^{-1} e^{i\omega(\mathbf{q})t}$$

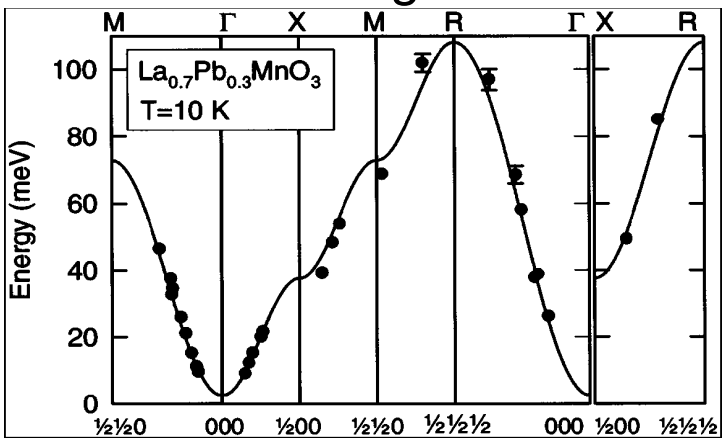
$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{1}{2}\left(1 + \hat{Q}_z^2\right)\frac{k_f}{k_i}\left[\frac{1}{2} g r_0 F(Q)\right]^2 \frac{S}{1 - e^{-\hbar\omega/kT}} \sum_{\mathbf{q}} \delta(\mathbf{Q} - \mathbf{q} - \boldsymbol{\tau})\delta(\omega - \omega(\mathbf{q}))$$



Linear spin waves

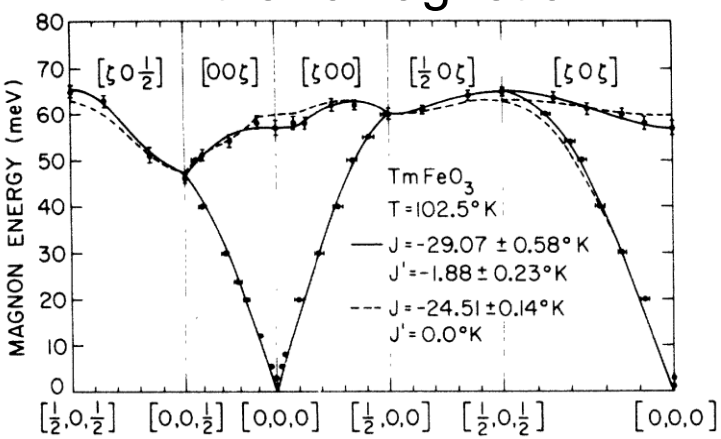
Spin waves

Ferromagnetic



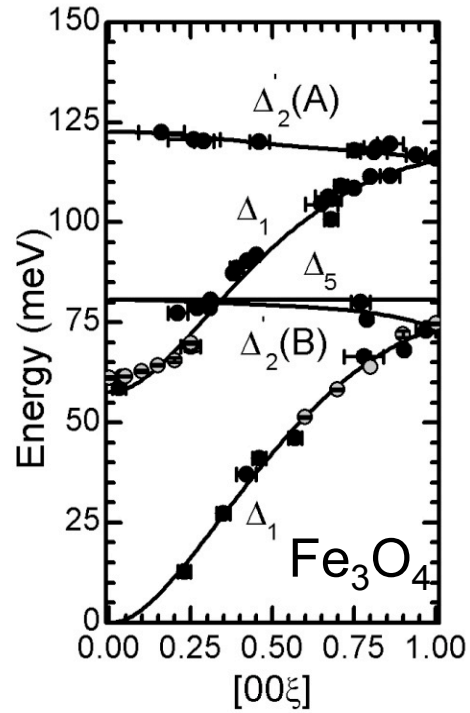
Perring *et al., Phys. Rev*. Lett. **77**, 711 (1996).

Antiferromagnetic



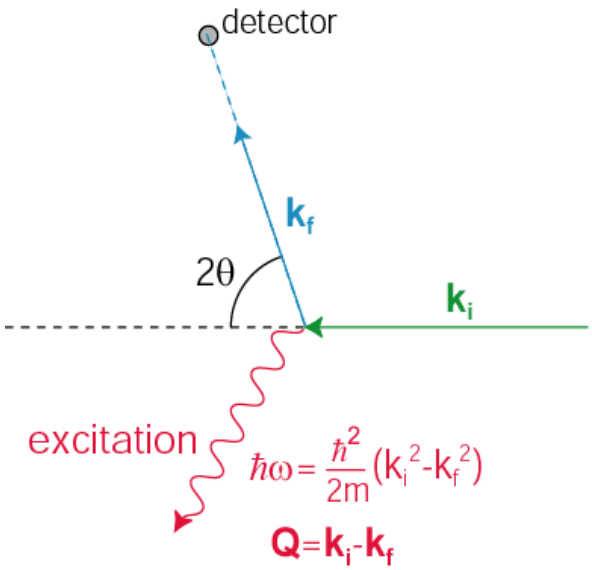
Shapiro *et al., Phys. Rev*. B **10**, 2014 (1974).

Ferrimagnetic

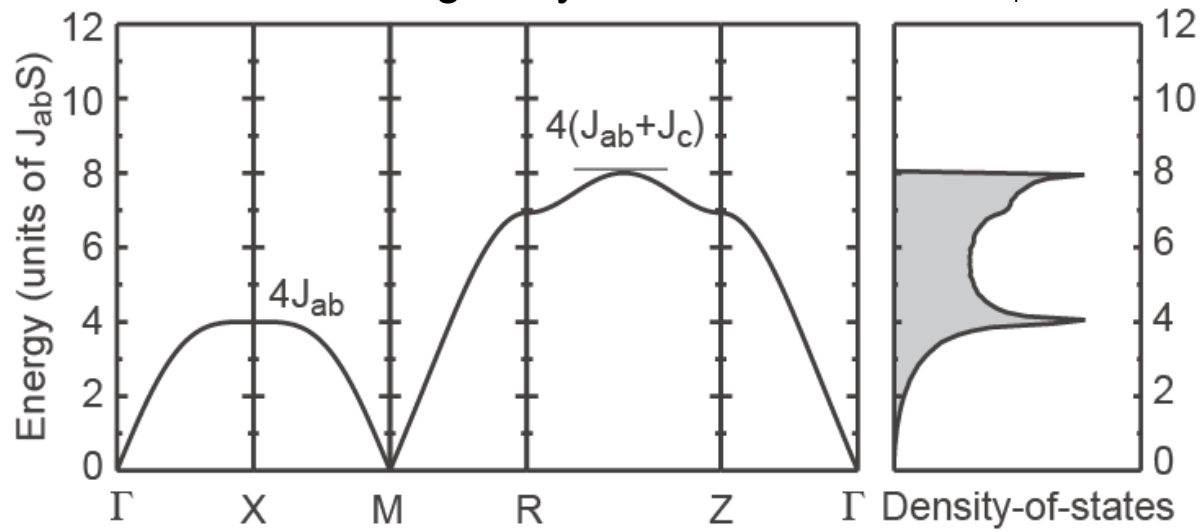


McQueeney *et al., Phys. Rev*. Lett. **99**, 246401 (2007).

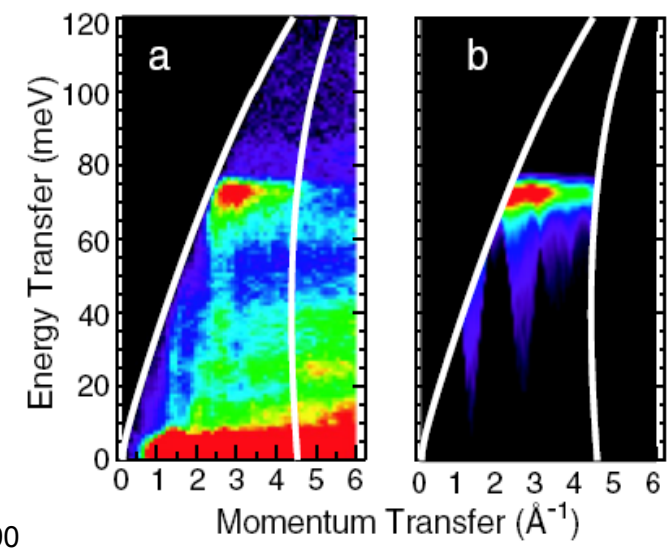
Scattering experiments



Single-crystal



Powder S(|**Q**|,ω)

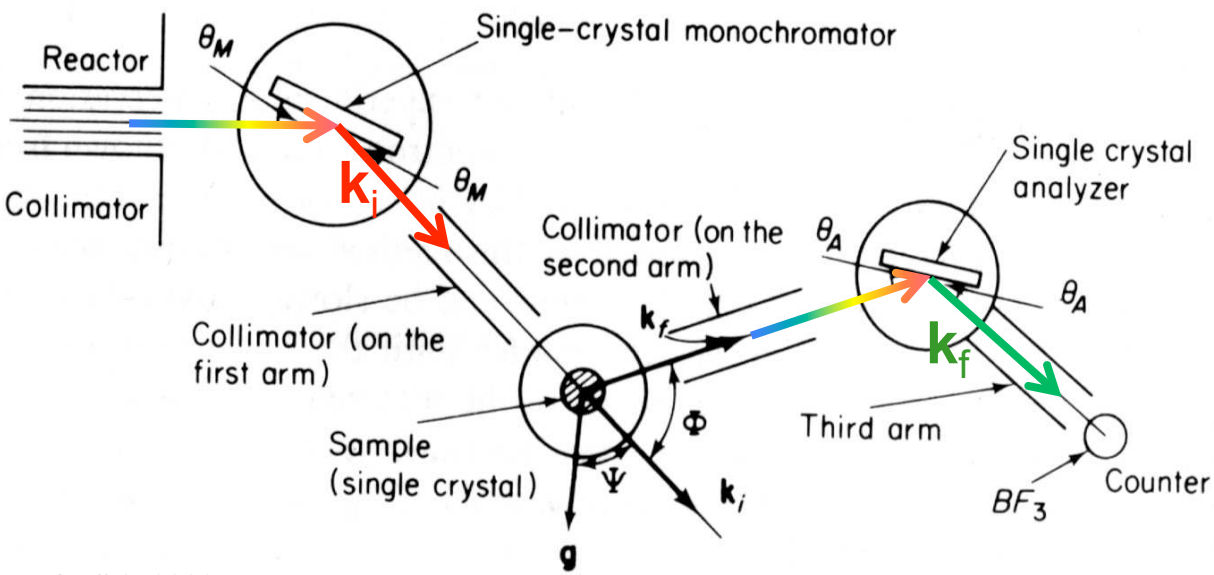


Instrument and sample (powder or single-crystal) determine how (**Q**,ω) space is sampled

Triple-axis instruments

High flux isotope reactor - ORNL

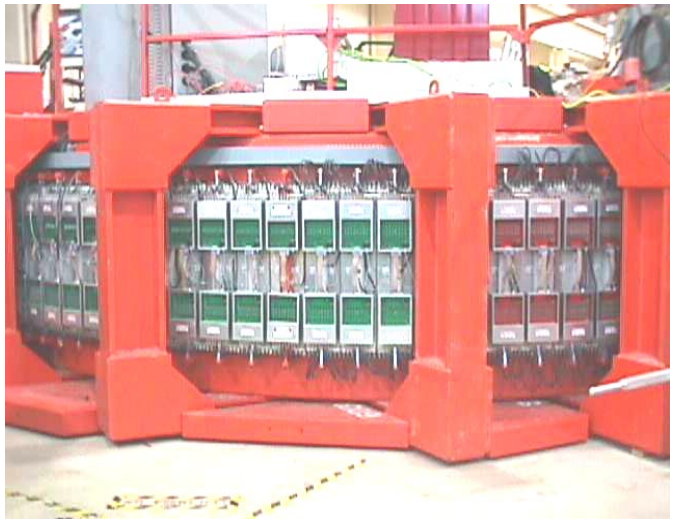
HB-1A 3-axis spectrometer



- Hardware flexibility
- Constant-**Q** (or E) scans
- Ideally suited for single-xtals

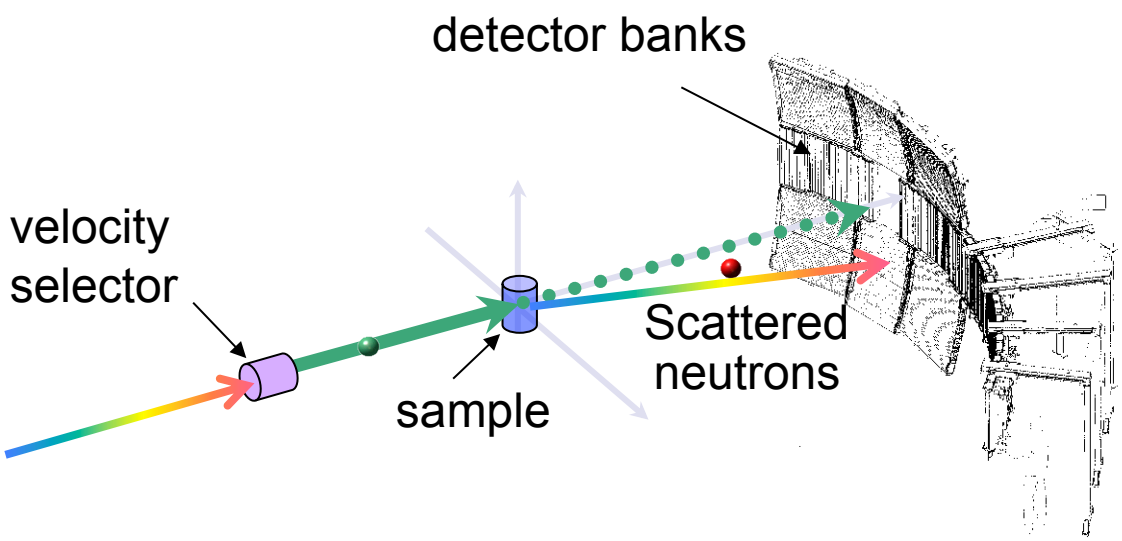
Time-of-flight methods

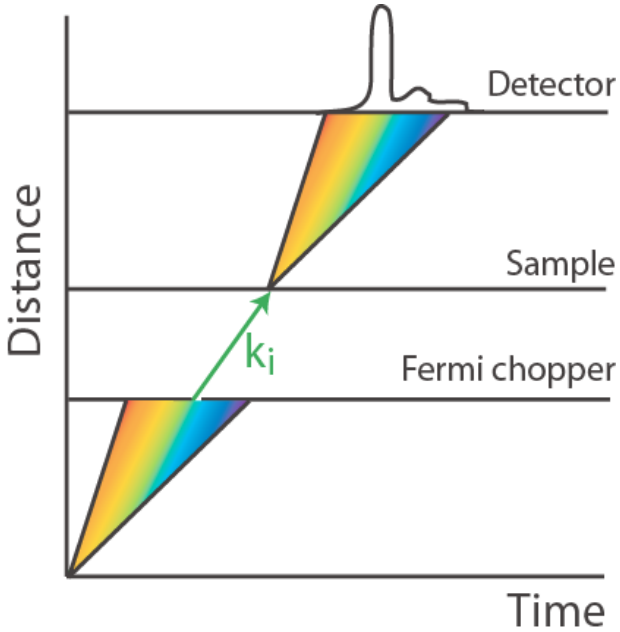
Spallation neutron source



Pharos – Lujan Center

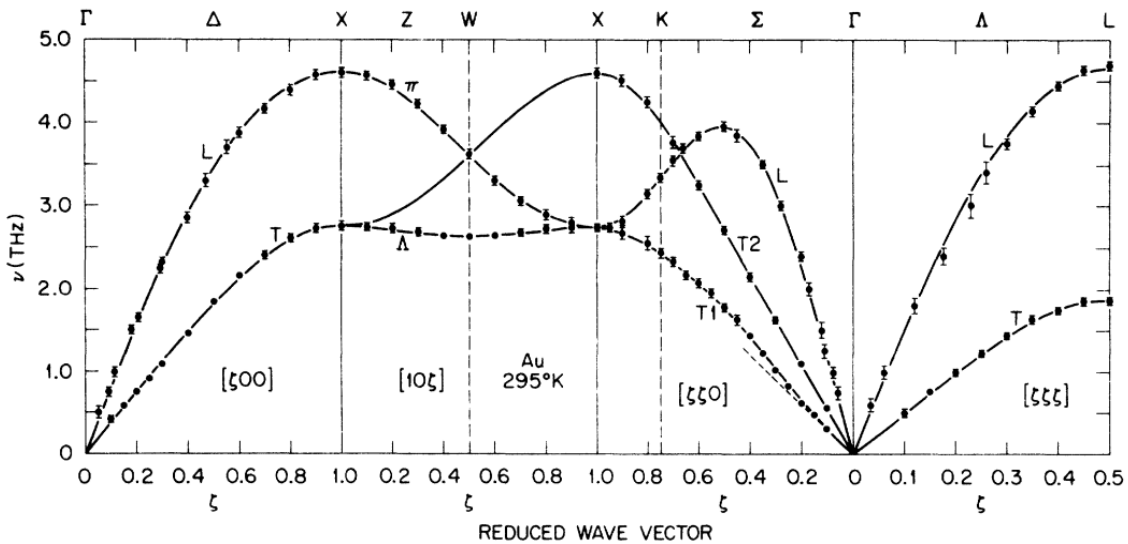
- Hardware inflexible
- Effective for powders
- Complicated **Q**,E-scans a challenge for single-xtals



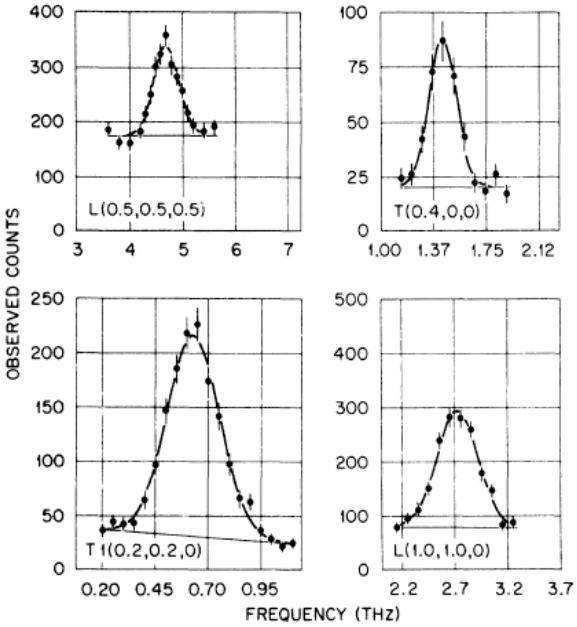


INS data

- Intensities as a function of **Q** and ω



Lynn, *et al.*, *Phys. Rev. B* **8**, 3493 (1973).



Reciprocal space

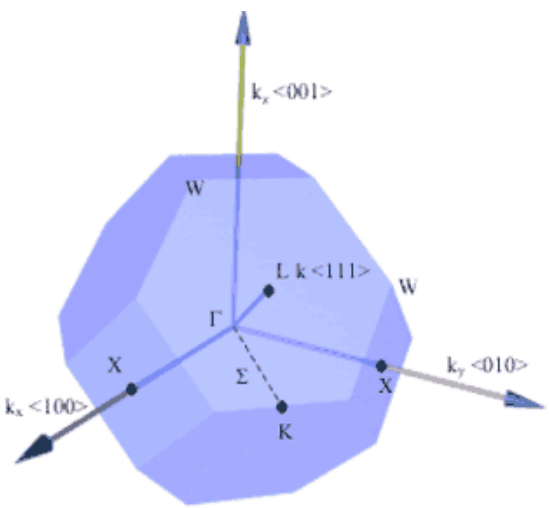
$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$

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

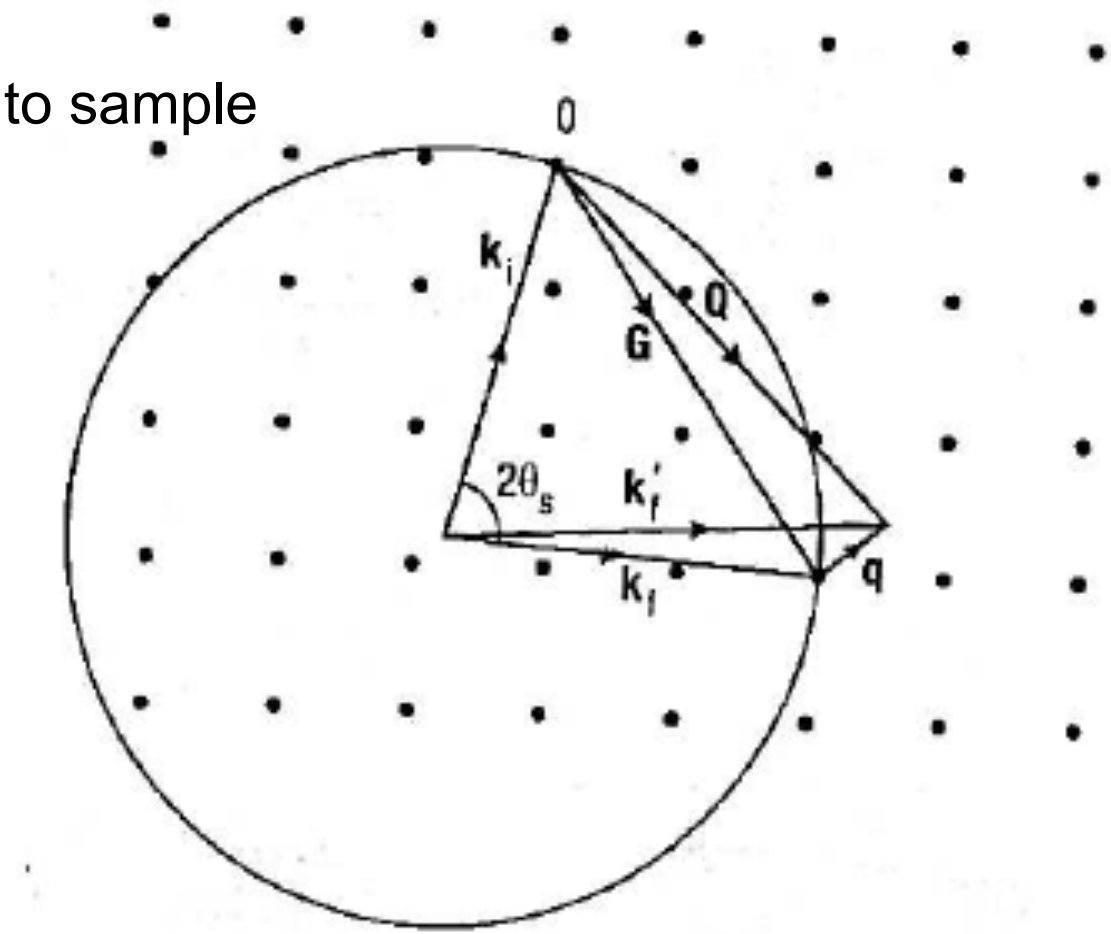
Momentum/energy transferred to sample

$$\mathbf{q} = \mathbf{Q} - \tau$$

Wavevector in 1st Brillouin zone



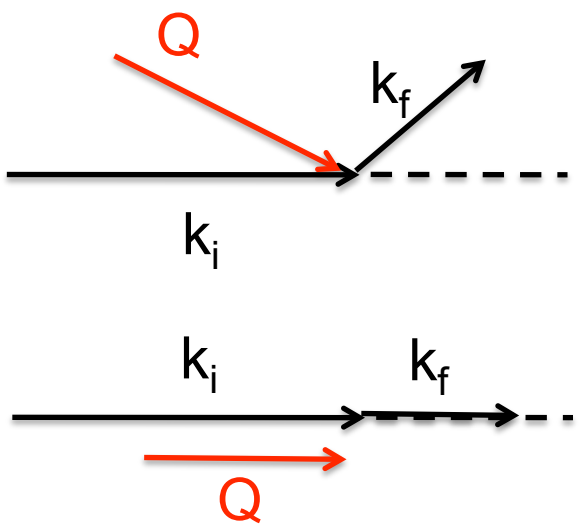
FCC Brillouin zone



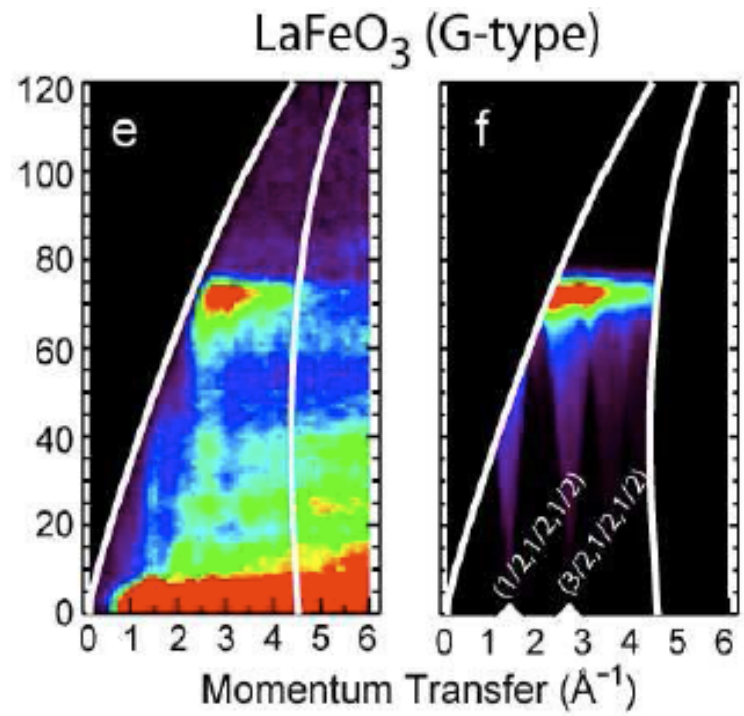
Scattering triangle

Kinematic limitations

- **Many combinations of k_i,k_f for same Q,ω**
  - Only certain configurations are used (eg. E_f-fixed)
- **Cannot "close triangle" for certain Q,ω due to kinematics**



Minimum accessible Q

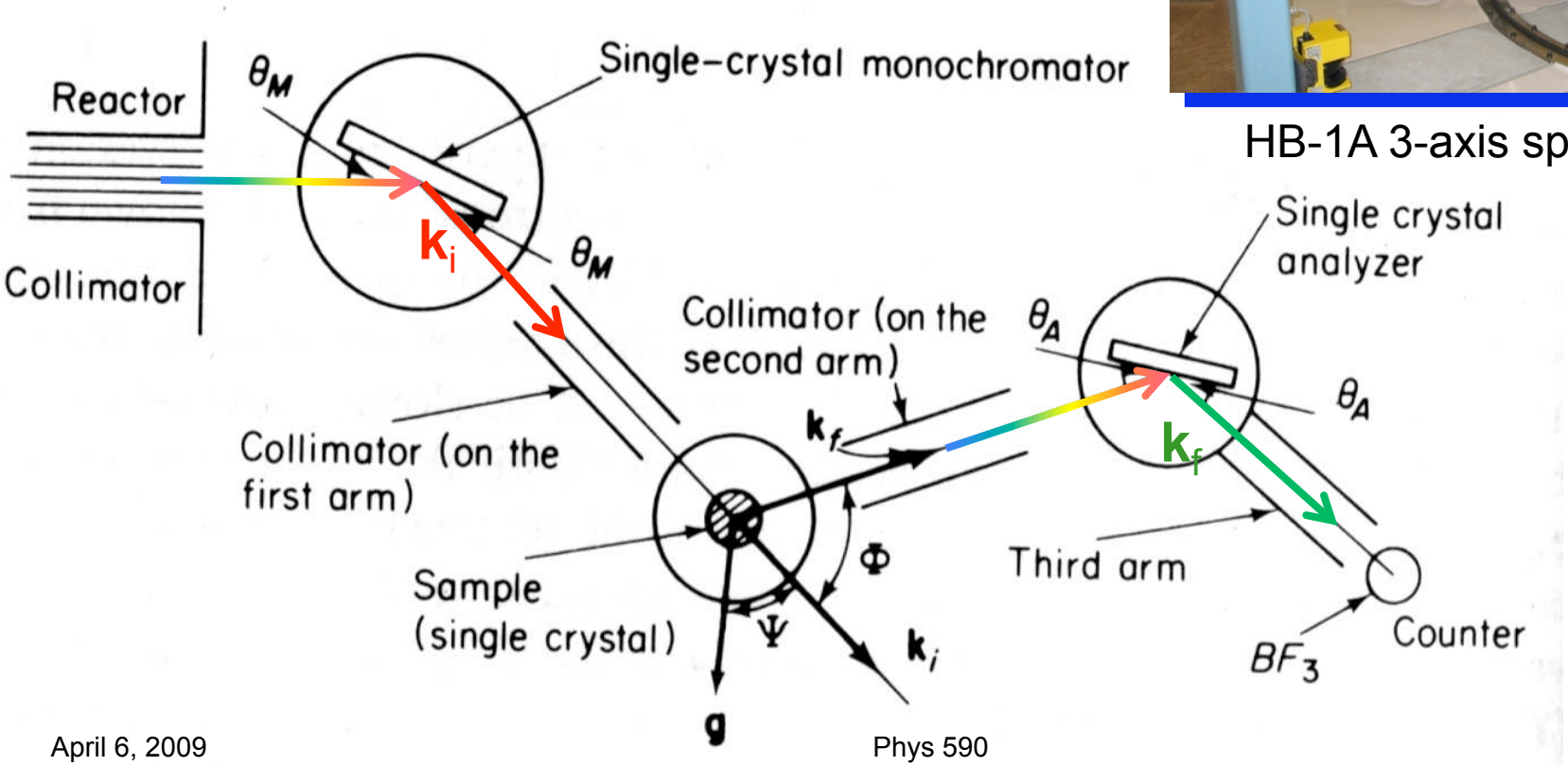


Kinematic limits, E_i=160 meV

Triple-axis instruments

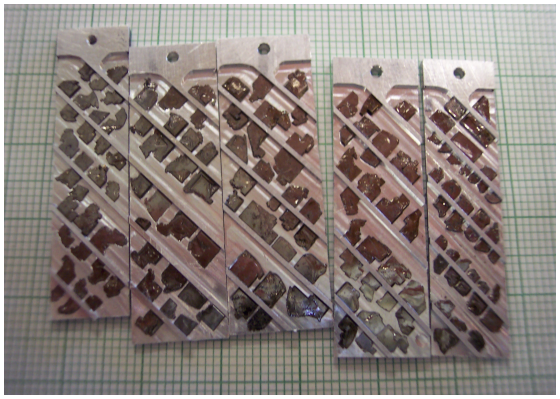
- **Workhorse INS instrument**
  - $\mathbf{k}_i$,$\mathbf{k}_f$ defined by Bragg scattering
  - Sample goniometer
  - Detector
  - Resolution/collimators

HB-1A 3-axis spectrometer



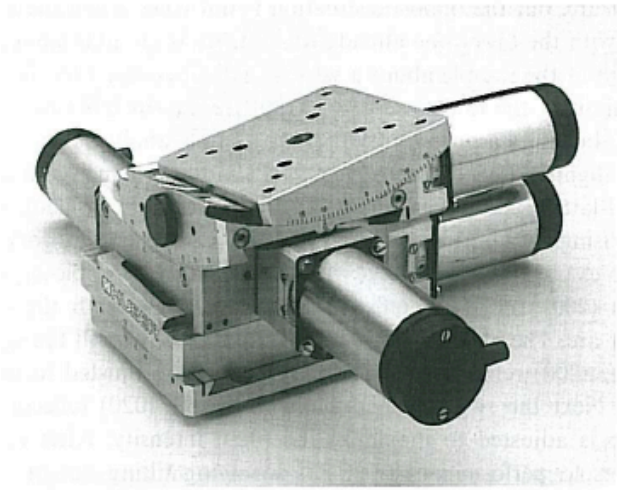
Samples

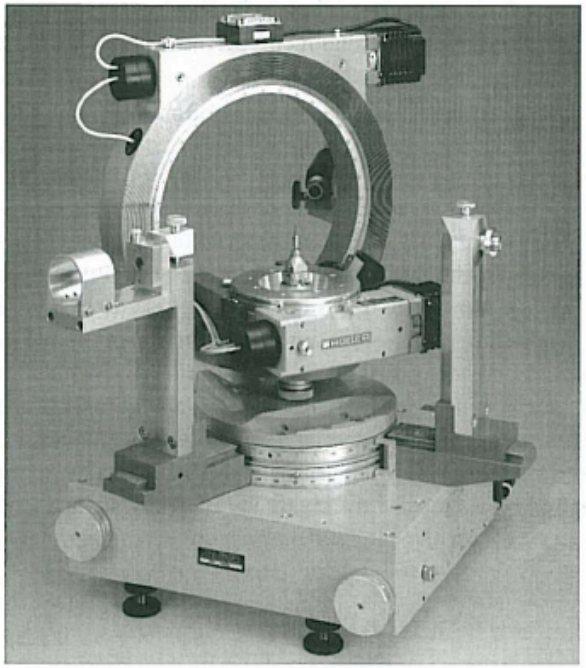
- **Samples need to be BIG**
  - ~ gram or cc
  - Counting times are long (mins/pt)



Co-aligned $CaFe_2As_2$ crystals

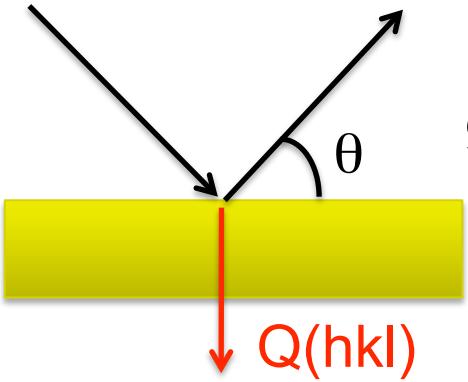
- **Sample rotation**
- **Sample tilt**



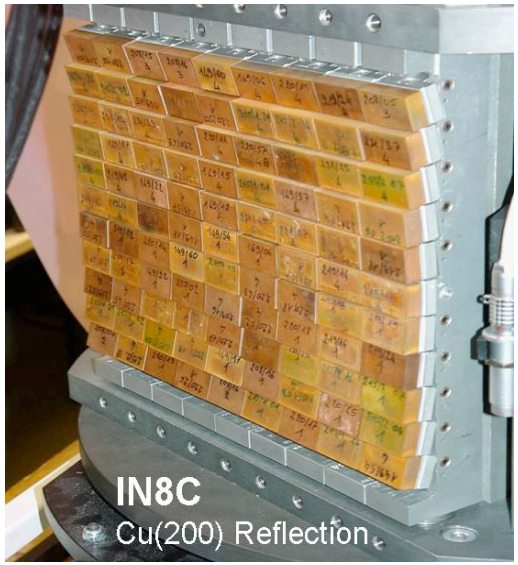


Monochromators

- **Selects the incident wavevector**



$$Q(hkl) = \frac{2\pi}{d(hkl)} = 2k_i \sin\theta$$

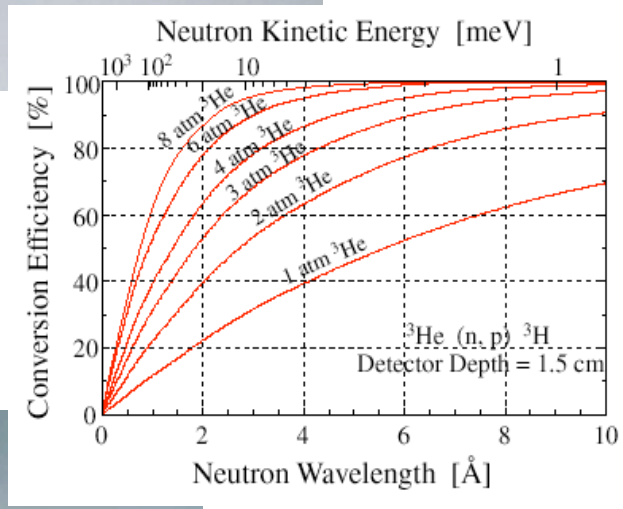


- Reflectivity
- focusing
- high-order contamination eg. λ/2 PG(004)

Mono	d(hkl)	uses
PG(002)	3.353	General
Be(002)	1.790	High k_i
Si(111)	3.135	No λ/2

Detectors

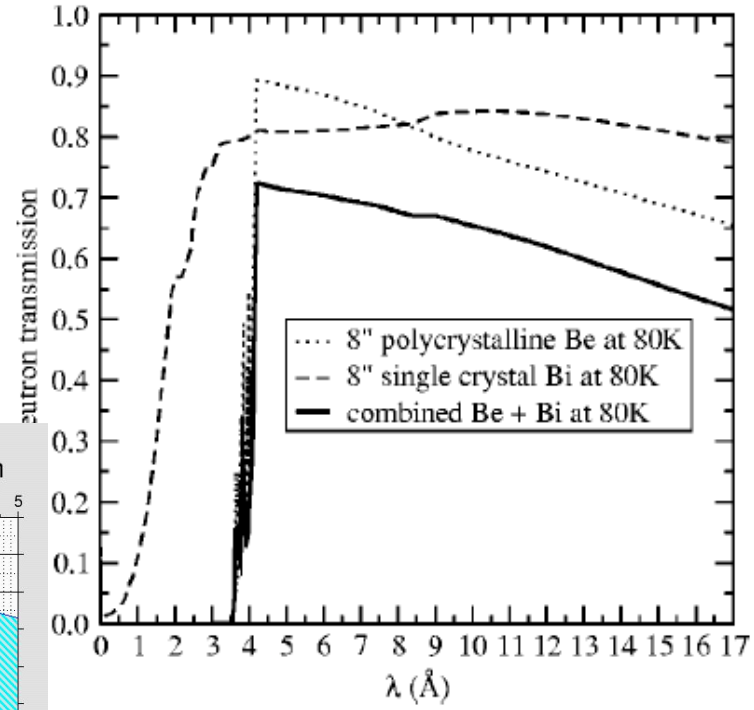
- **Gas Detectors**
- n + ^{3}He → ^{3}H + p + 0.764 MeV
- Ionization of gas
- e^- drift to high voltage anode
- High efficiency

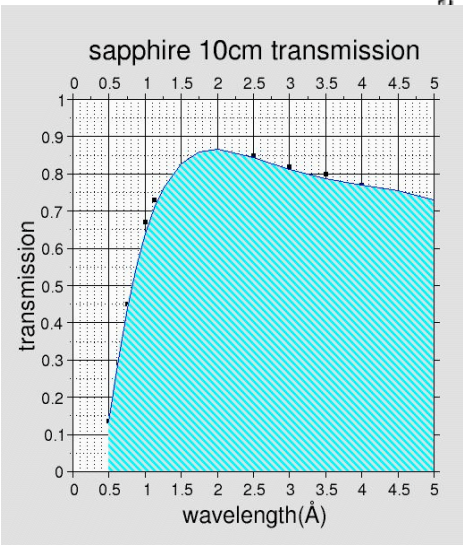


- **Beam monitors**
- Low efficiency detectors for measuring beam flux

Other triple-axis stuff

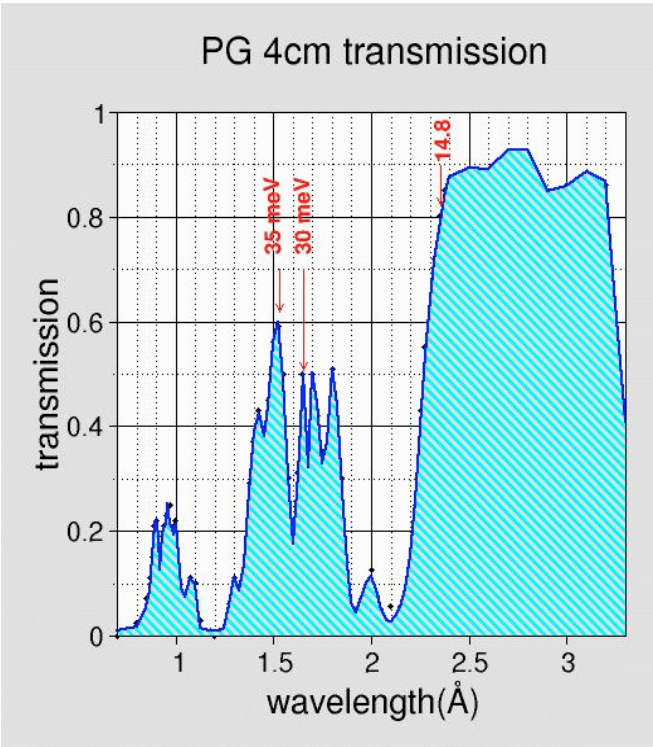
- **Soller Collimators**
  - Define beam divergence
  - Q,ω resolution function
- **Filters**
  - Xtal Sapphire: fast neutron background
  - Poly Be: low-energy (5 meV) band pass
  - PG: higher order contamination
- **Masks**
  - Beam definition
  - Background reduction

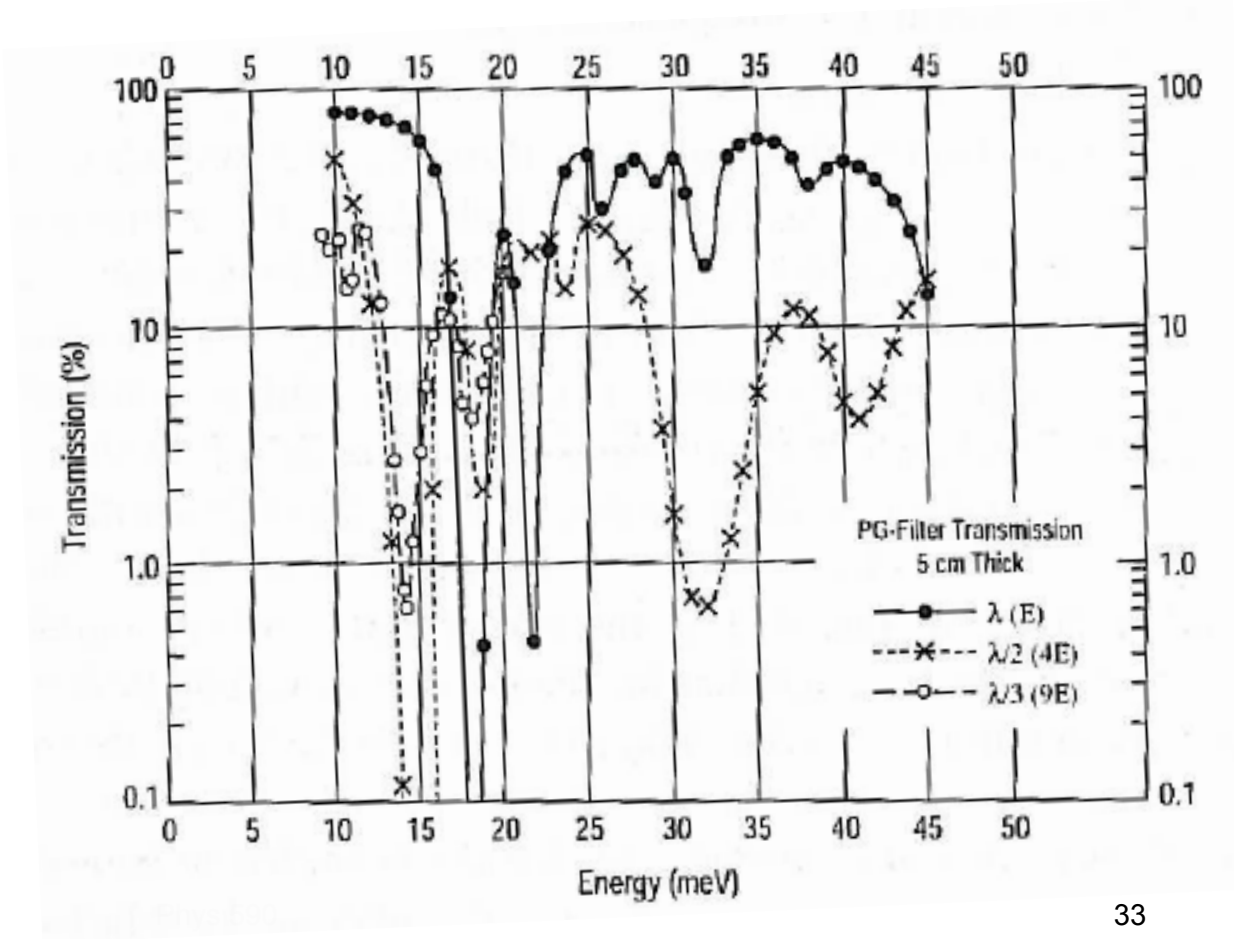




PG filter

- **Magic numbers**
  - Best filter for rejection of $\lambda/2$ contamination
  - E_f = 13.7, 14.7, 30.5, 41 meV



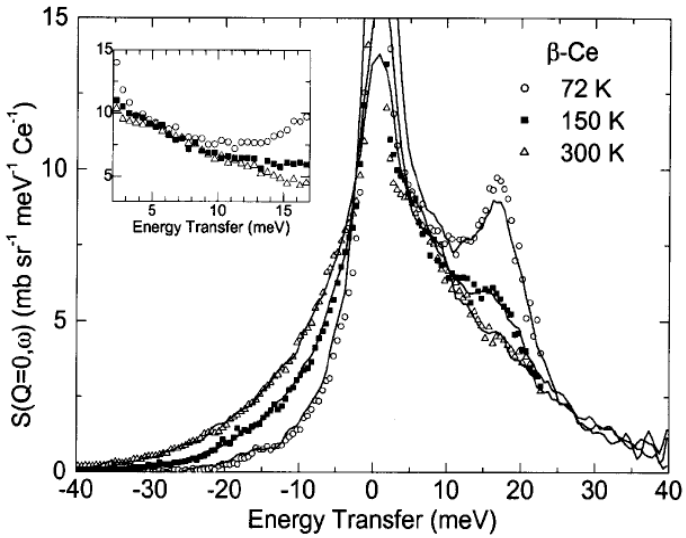


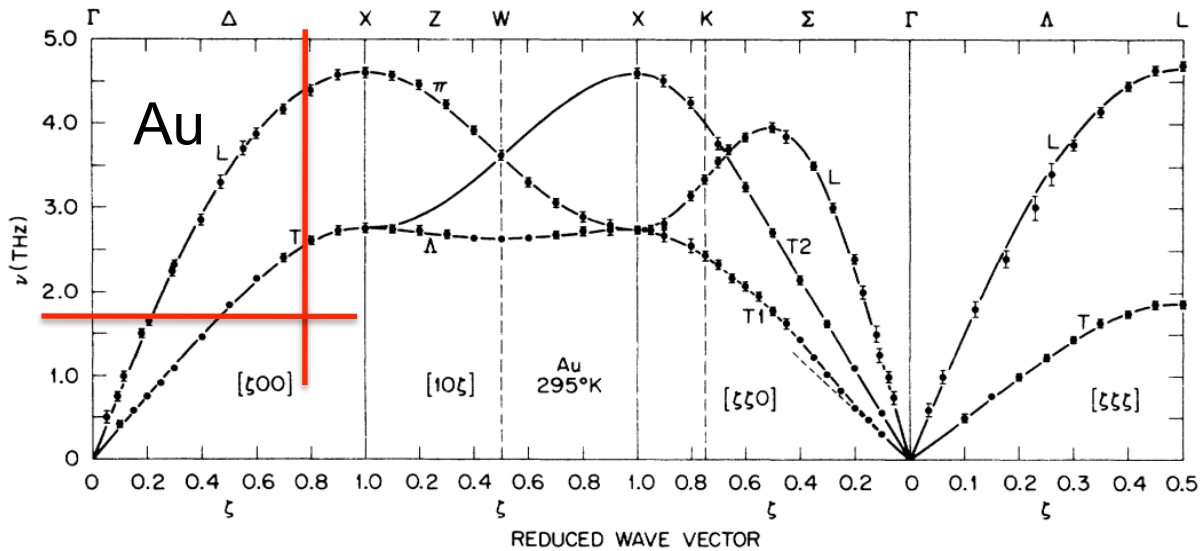
Sample environment

- **Temperature, field, pressure**
- **Heavy duty for large sample environment**
  - CCR
  - He cryostats
  - SC magnets
  - …

Acquiring data

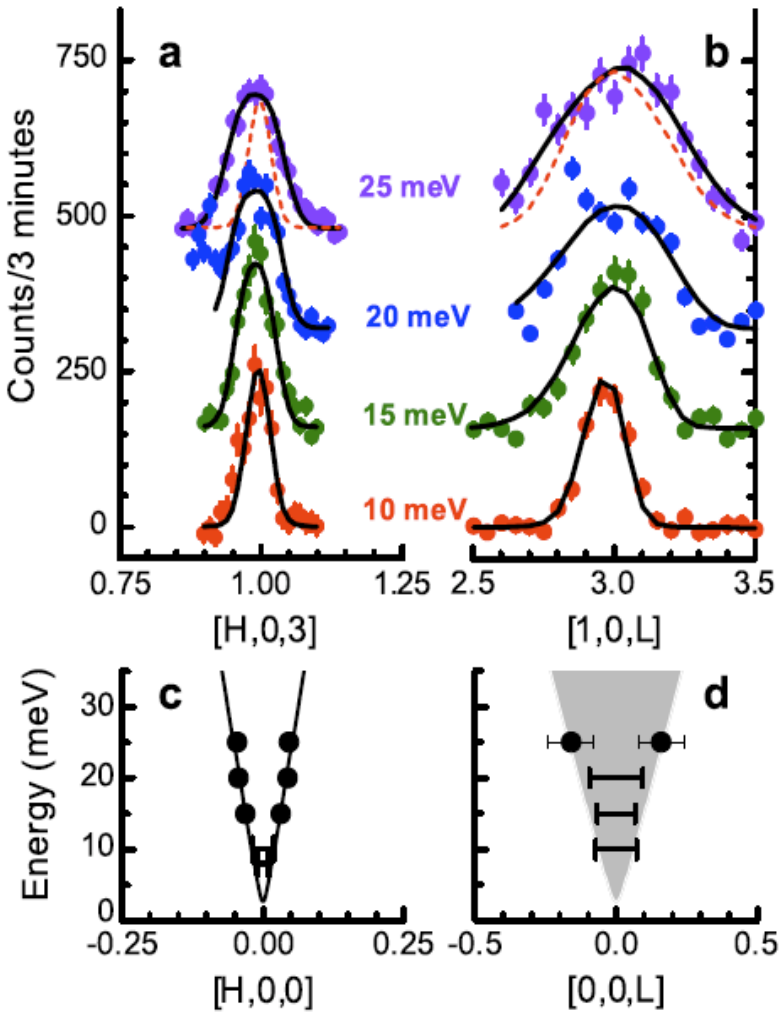
- **Energy gain, energy loss**
  - Detailed balance
- **Constant-Q scans**
  - Most common
- **Constant-ω scans**
  - Used for steep dispersions

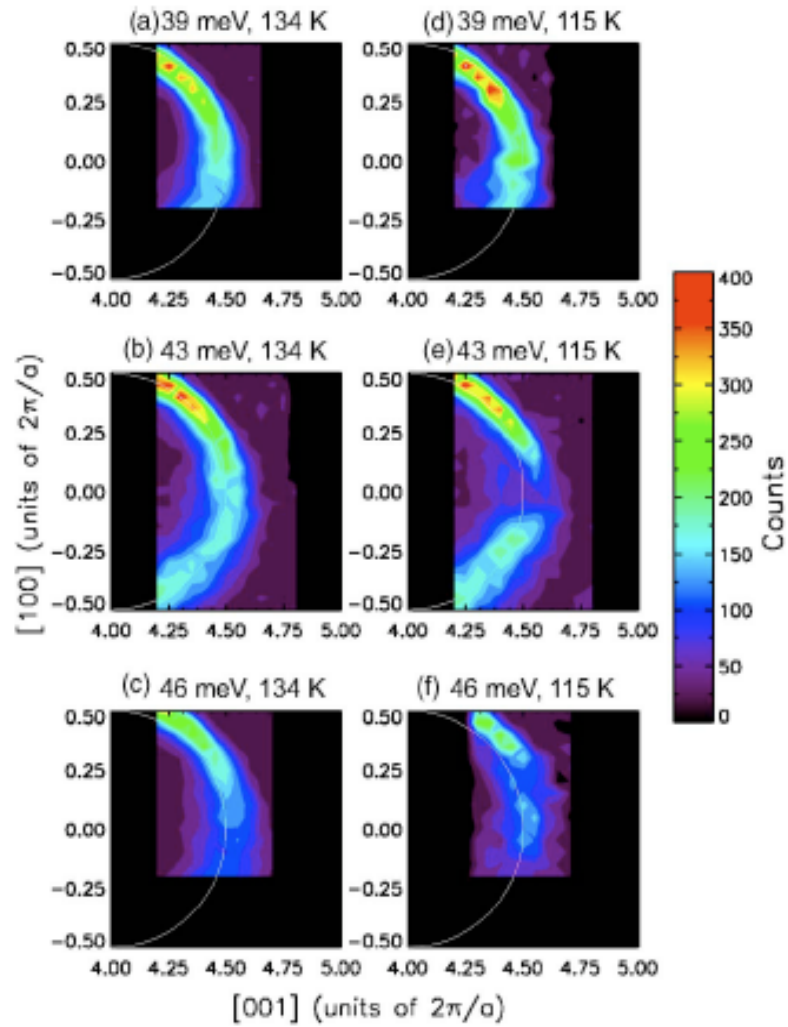




Lynn, *et al.*, *Phys. Rev. B* **8**, 3493 (1973).

Constant-ω scans

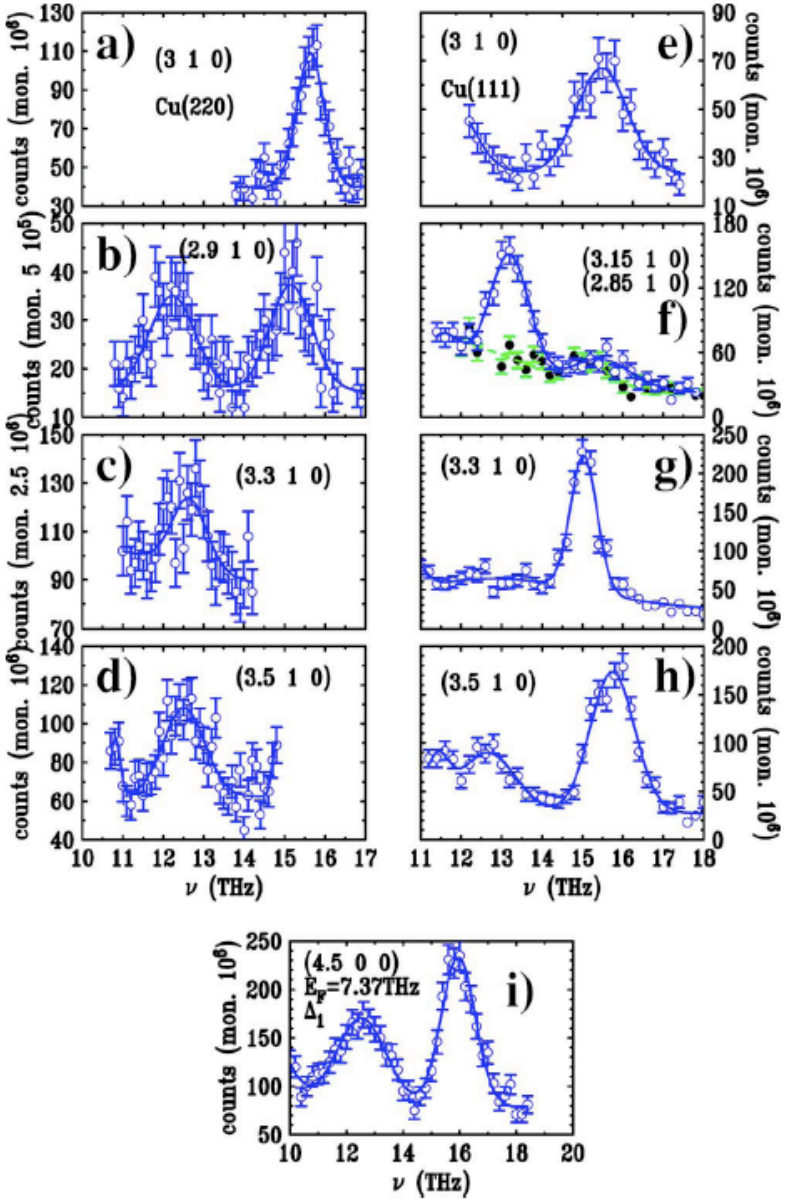


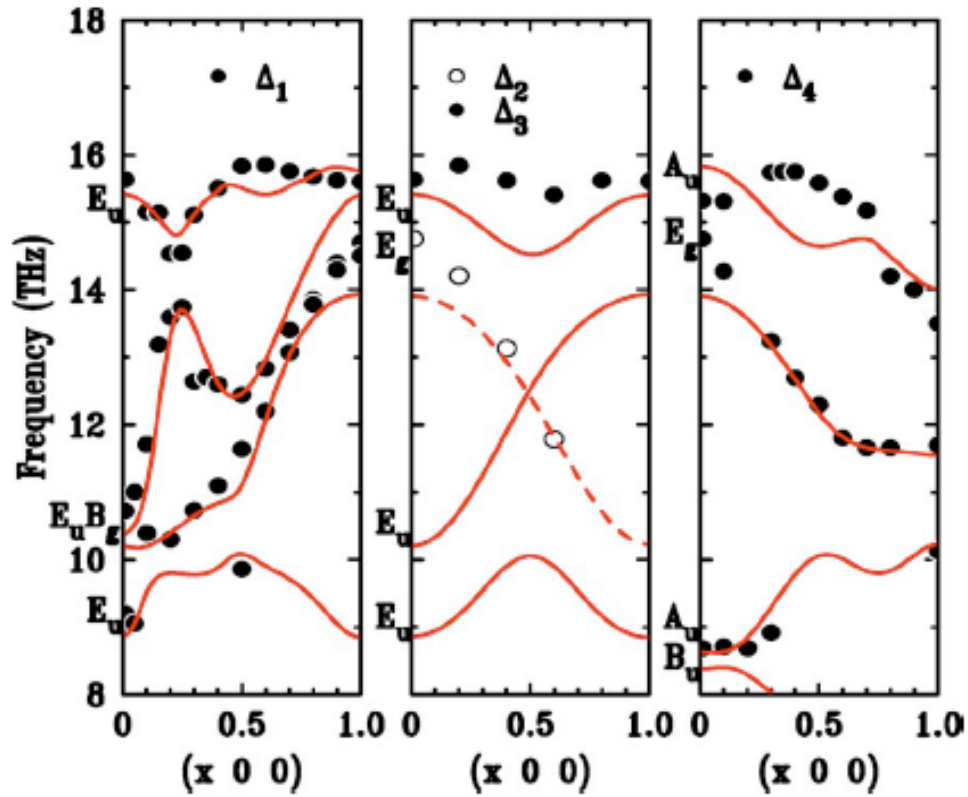


Steep AF spin waves in $CaFe_2As_2$
McQueeney et al, PRL 101, 227205 (2008)

Slice of spin wave cone in Fe_3O_4
McQueeney et al, PRB 73, 174409 (2006)

Constant-Q scans

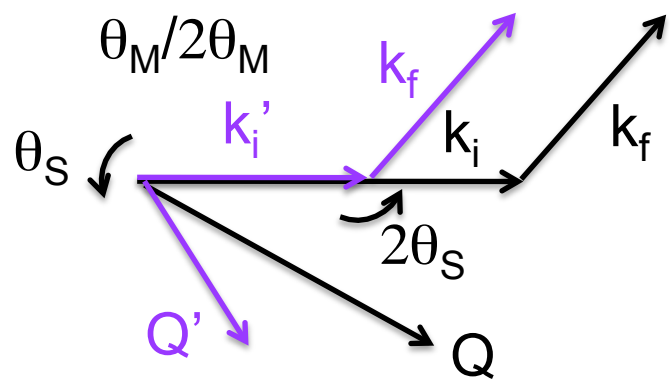




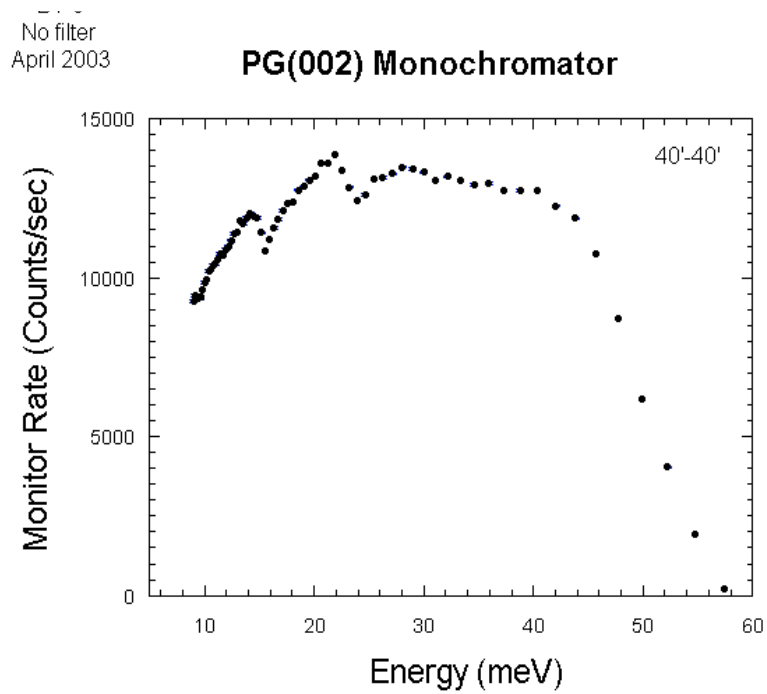
Optical phonons in $Nd_{0.85}Ce_{0.15}CuO_4$
Braden, et al, PRB, 72 184517 (2005).

Configurations

- **E_f-fixed mode**
  - Mono moves during ω-scan
  - Beam monitor accounts for variations in incident flux
- **E_i-fixed mode**
  - Analyzer moves during ω-scan
  - Useful for expts requiring low background
  - Analysis more complicated
- **Magic numbers**
  - λ/2 contamination
  - E_i or E_f = 13.7, 14.7, 30.5, 41 meV

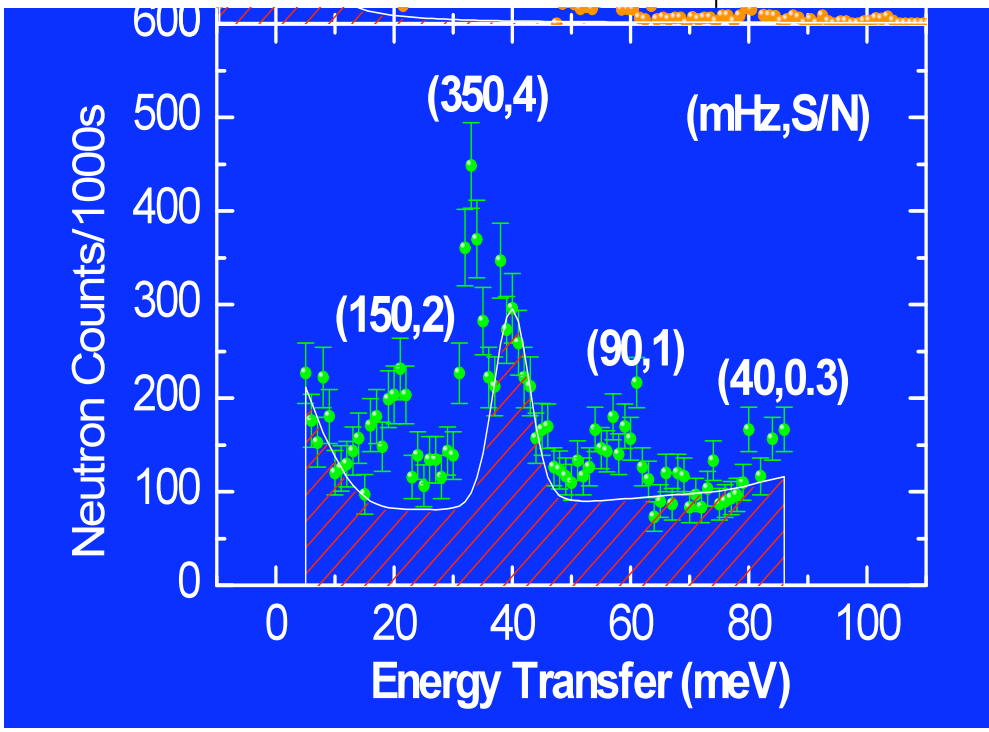


Fixed-E_f mode



Spurions

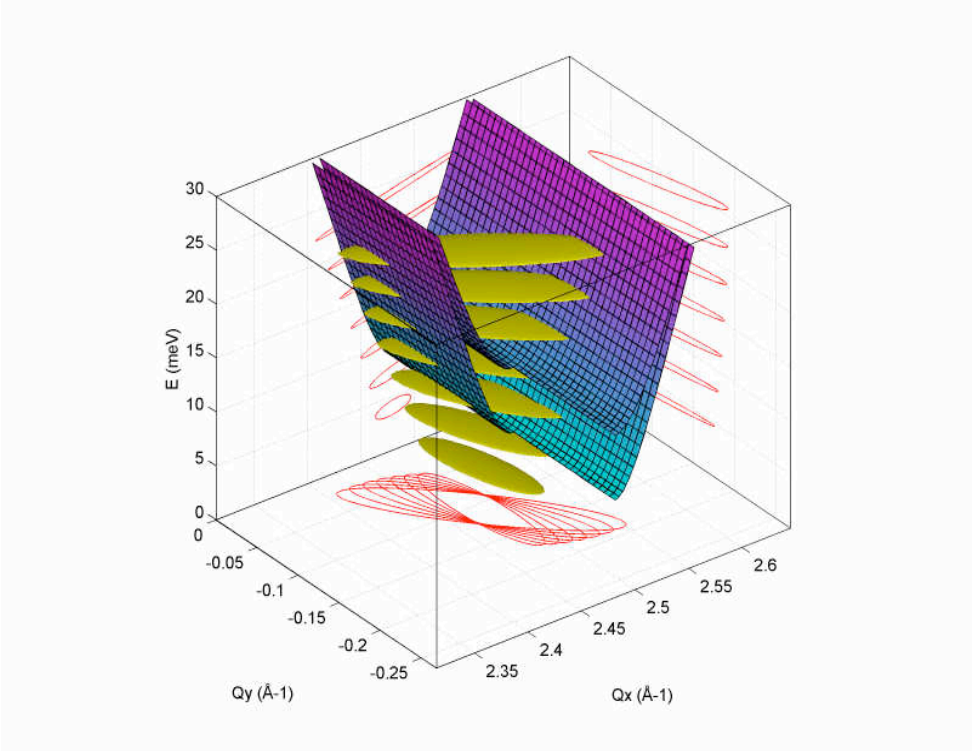
- **Bragg – incoherent – Bragg**
  - Eg. $k_i - 2k_f$
    - $\hbar\omega$ = 41.1 meV
    - E_f = 13.7 meV
      E_i = 54.8 meV
      $4E_f$ = 54.8 meV
    - Incoherent elastic scattering visible from analyzer $\lambda/2$



- **incoherent – Bragg – Bragg**
  - Sample 2θ in Bragg condition for k_f-k_f
  - Even for inelastic config, weak incoherent from mono

Resolution

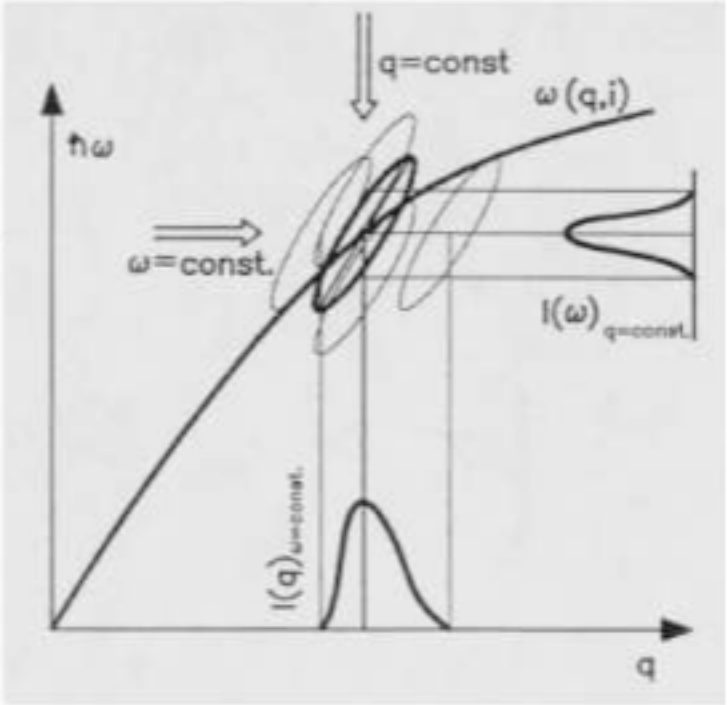
- **Resolution ellipsoid**
  - Beam divergences
  - Collimations/distances
  - Crystal mosaics/sizes/angles

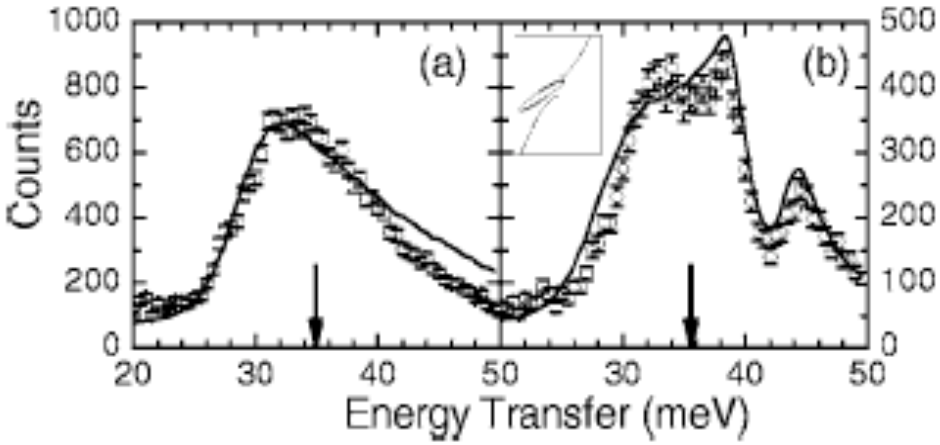


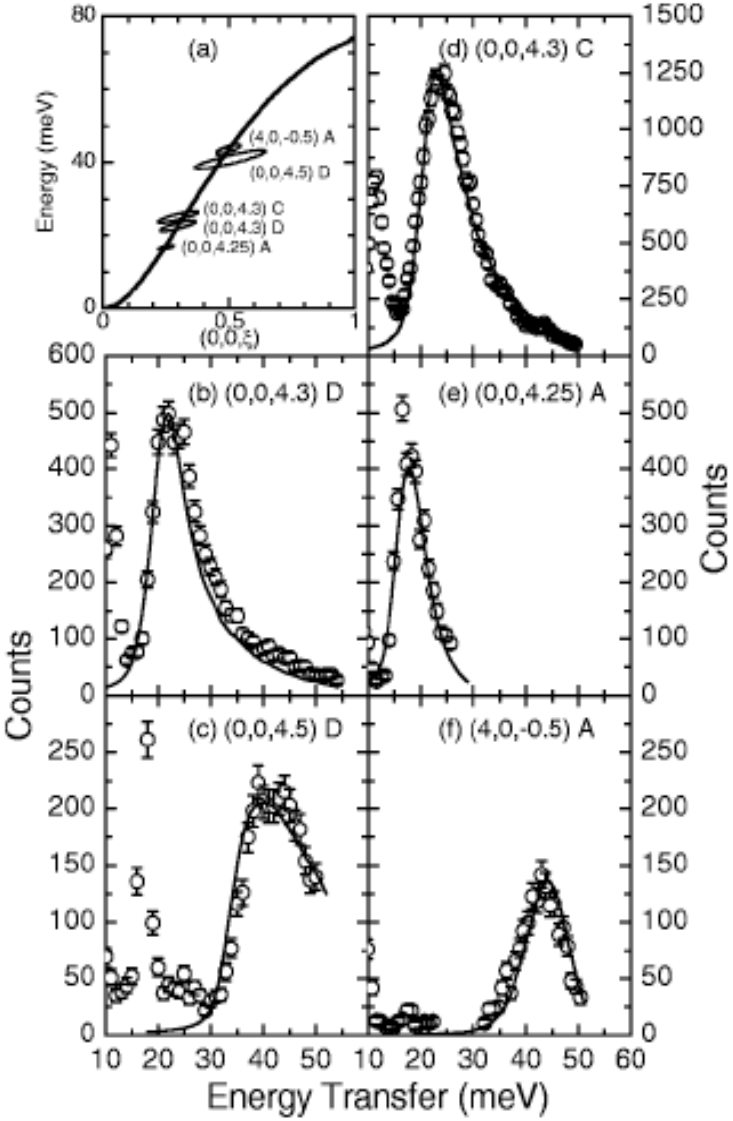
- **Resolution convolutions**

$$\mathrm{I}(\mathbf{Q}_0,\omega_0) = \int \mathrm{S}(\mathbf{Q}_0,\omega_0) R(\mathbf{Q}-\mathbf{Q}_0,\omega-\omega_0)\, d\mathbf{Q}\, d\omega$$

Resolution effects

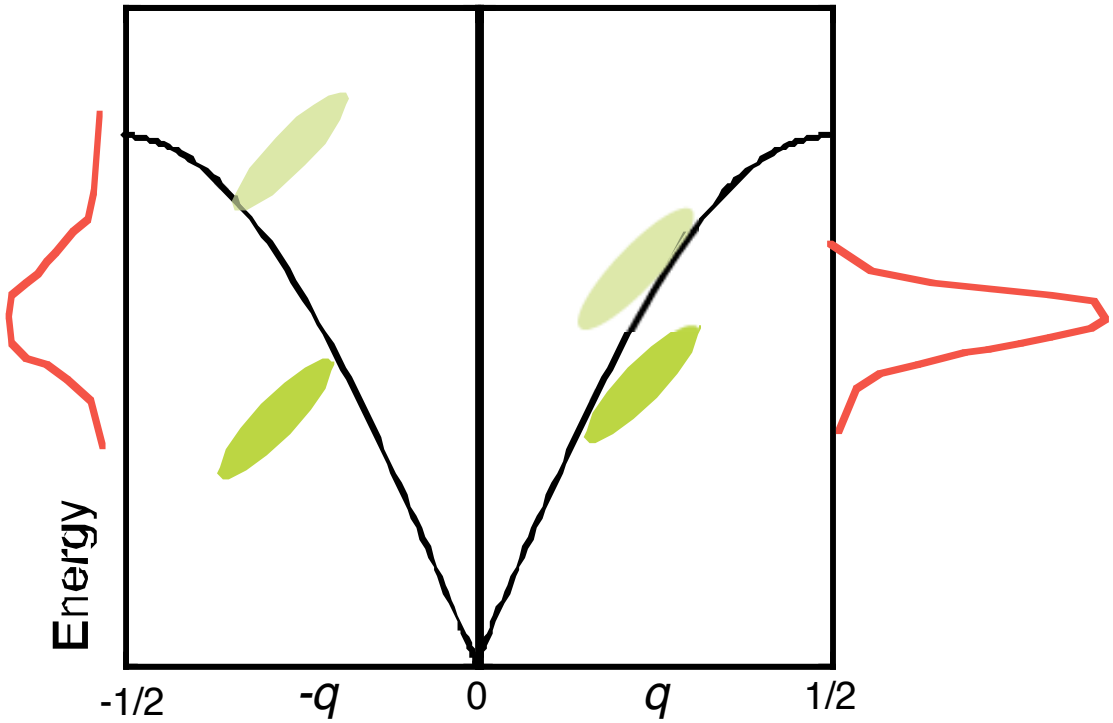






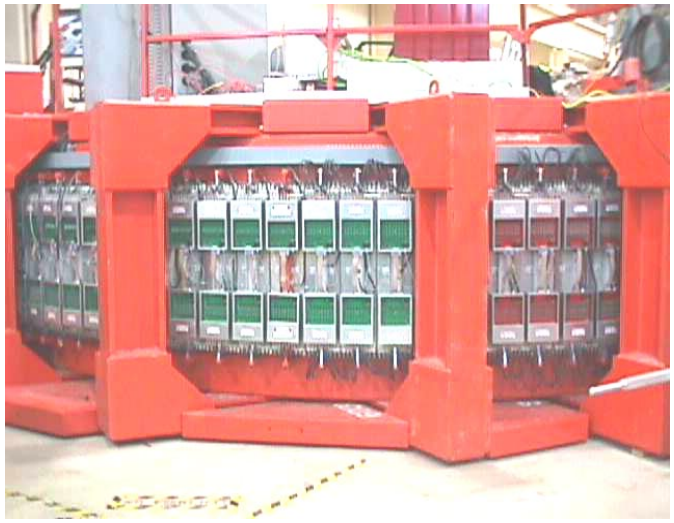
Resolution focusing

- Optimizing peak intensity
- Match slope of resolution to dispersion



Time-of-flight methods

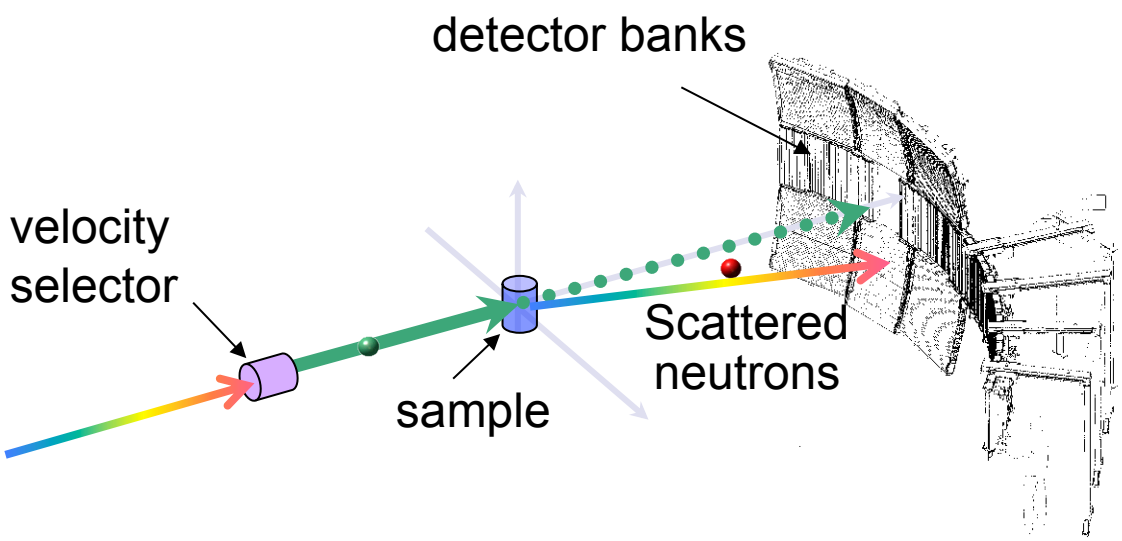
Spallation neutron source

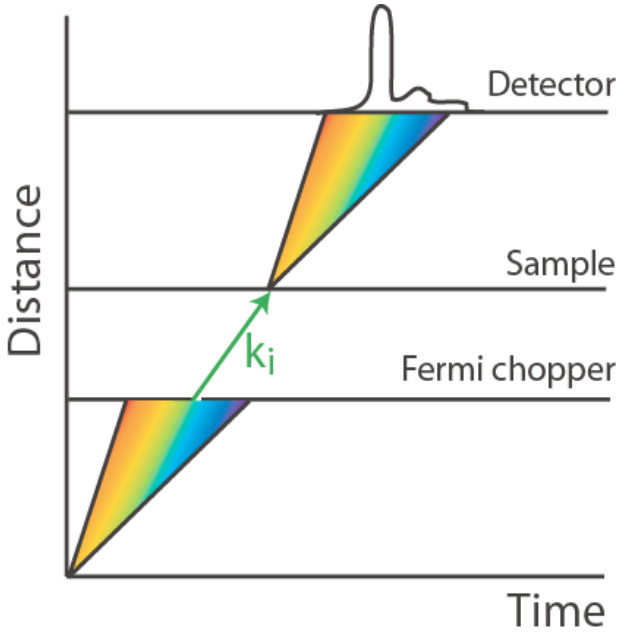


Pharos – Lujan Center

- Effectively utilizes time structure of pulsed neutron groups

$$t = \frac{d}{v} = \left(\frac{m}{h} d\right)\lambda$$

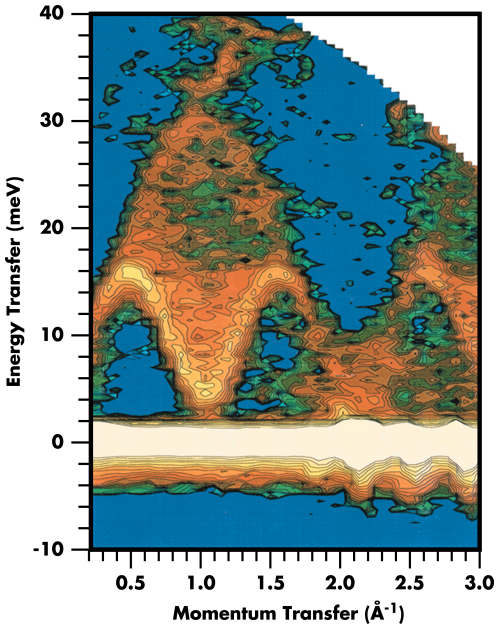


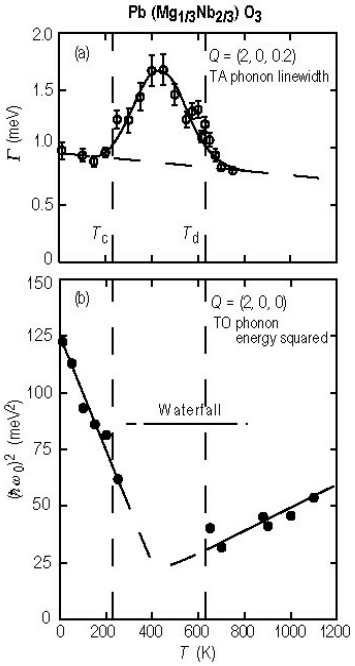


TOF vs. 3-axis

- epithermal (up to 2 eV)
- Total spectra (esp. powder samples)
- Absolute normalization
- Low-dimensional systems
- Hardware inflexible
- Software intensive

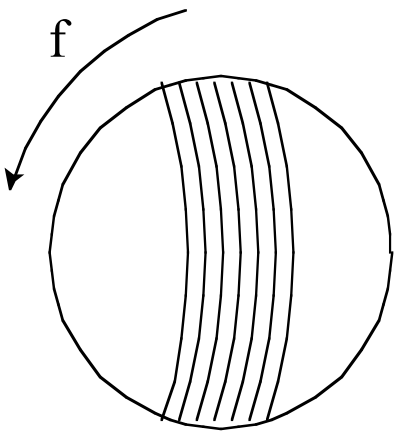
- High flux of thermal neutrons
- Focused studies in Q,ω (soft modes, gaps, etc.)
- Three-dimensional systems
- Hardware intensive
- Software inflexible





Fermi Choppers

- Body radius ~ 5 cm
- Curved absorbing slats
  - B or Gd coated
  - ~mm slit size
- f = 600 Hz (max)
- Acts like shutter, $\Delta t \sim \mu s$



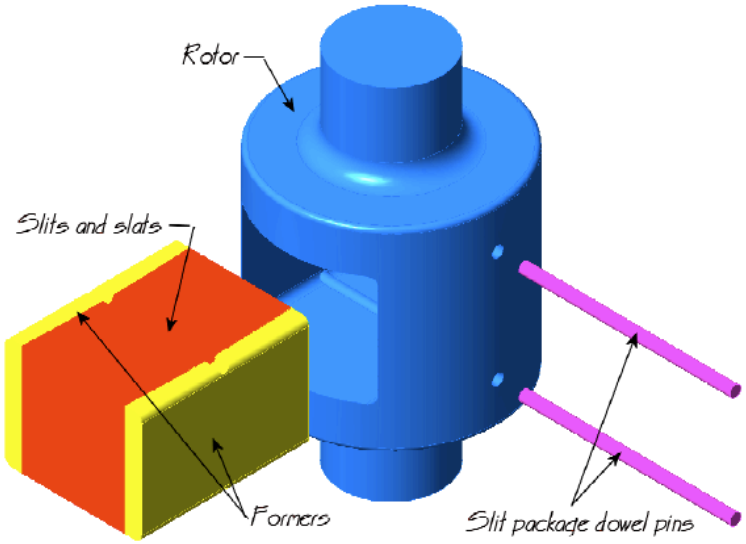
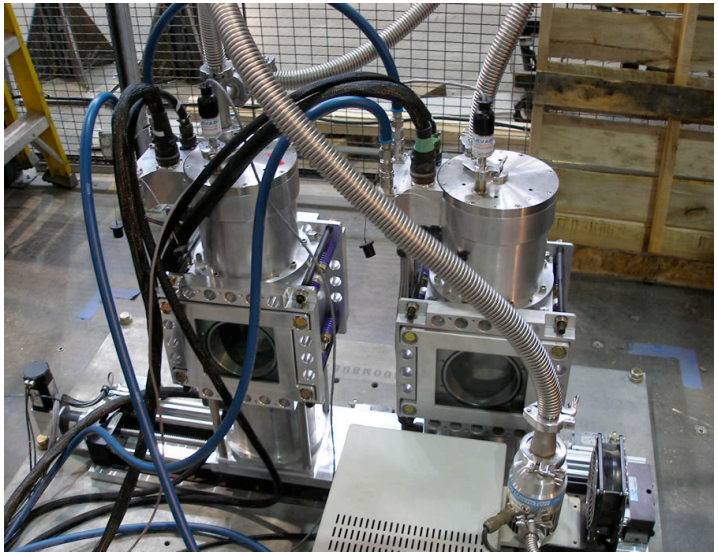


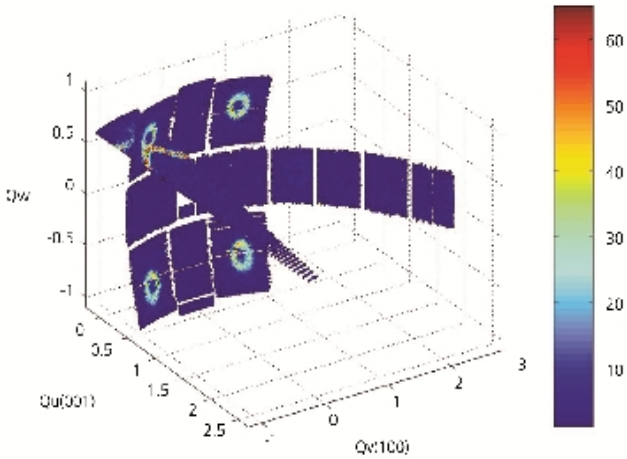
Figure 1. ISIS MAPS chopper and slit package assembly – exploded view



Position sensitive detectors

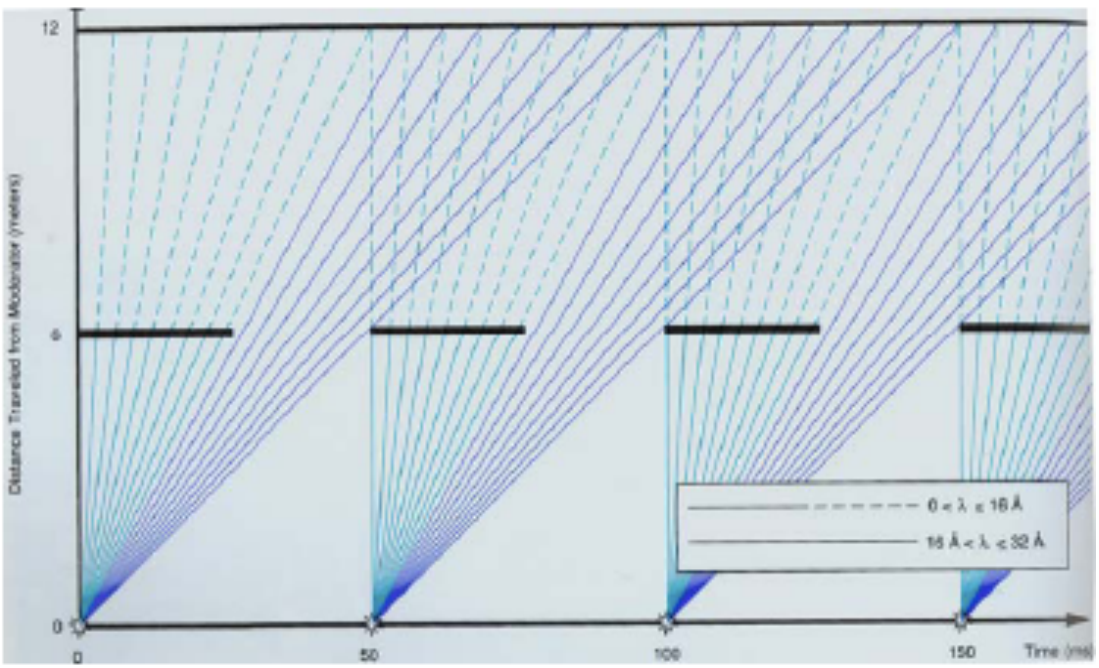
- ^{3}He tubes (usu. 1 meter)
- Charge division
- Position resolution ~ cm
- Time resolution ~ 10 ns

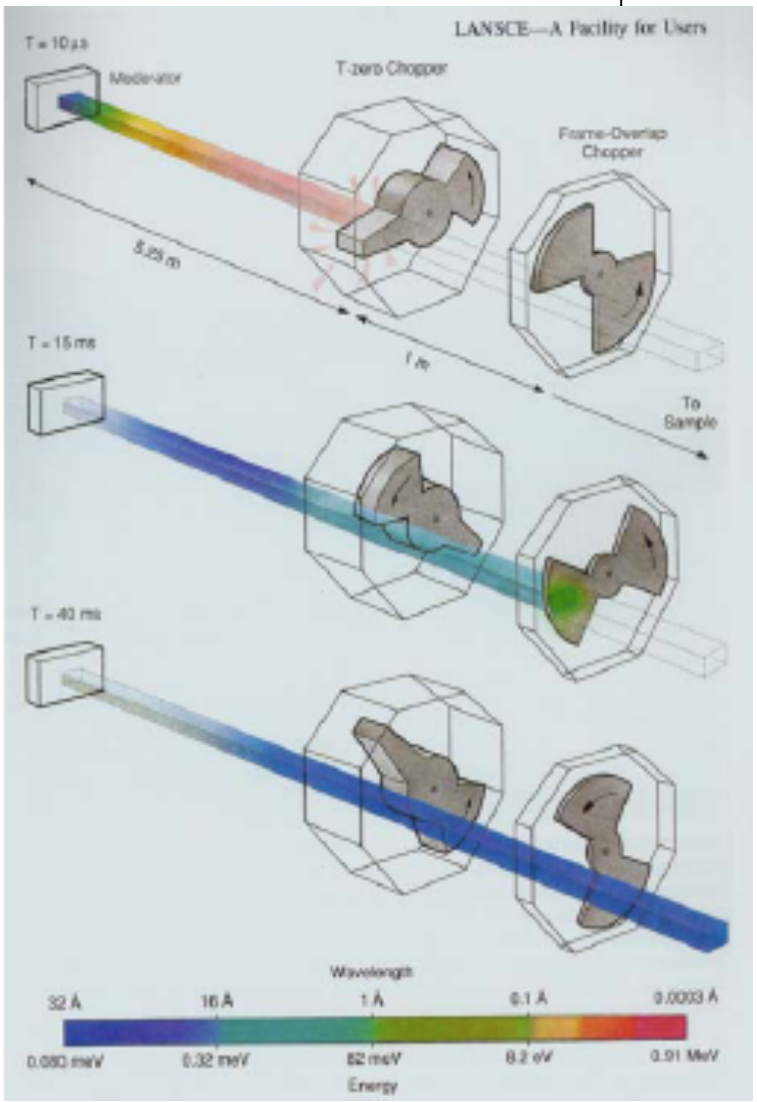
MAPS detector bank



T-zero chopper

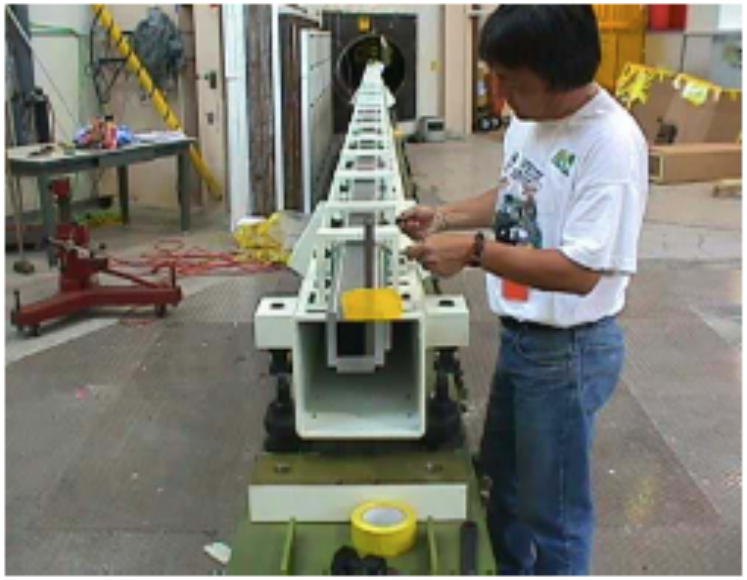
- **Background suppression**
- **Blocks fast neutron flash**

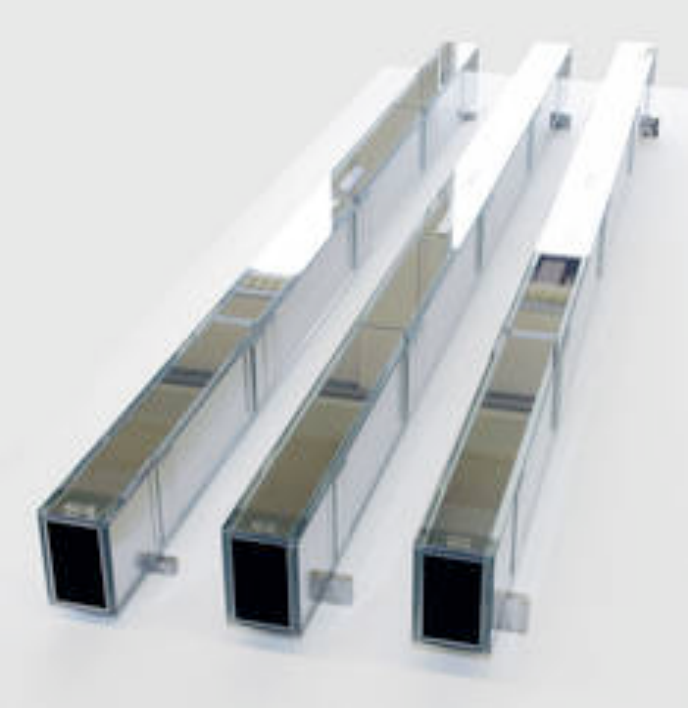




Guides

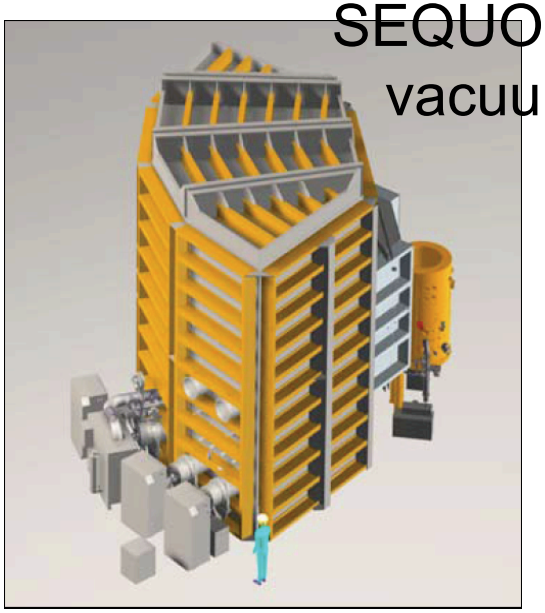
- **Transport beam over long distances**
- **Background reduction**
- **Total external reflection**
  - Ni coated glass
  - Ni/Ti multilayers (supermirror)





Size matters

- **Length = resolution**
  - Instruments ~ 20 – 40 m long
  - E-resolution ~ 2-4% E_i
- **More detectors**
  - SEQUOIA – 1600 tubes, 144000 pixels
  - Solid angle coverage 1.6 steradians
- **Huge data sets**
- 0.1 – 1 GB

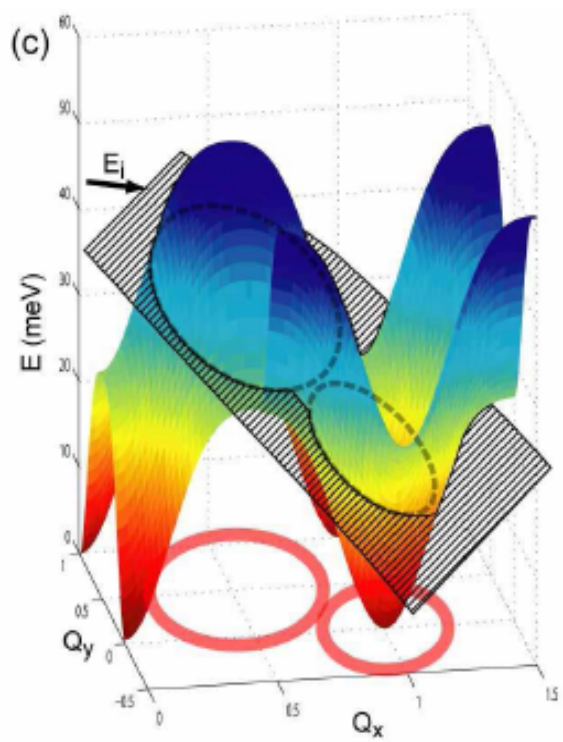


SEQUOIA detector vacuum vessel

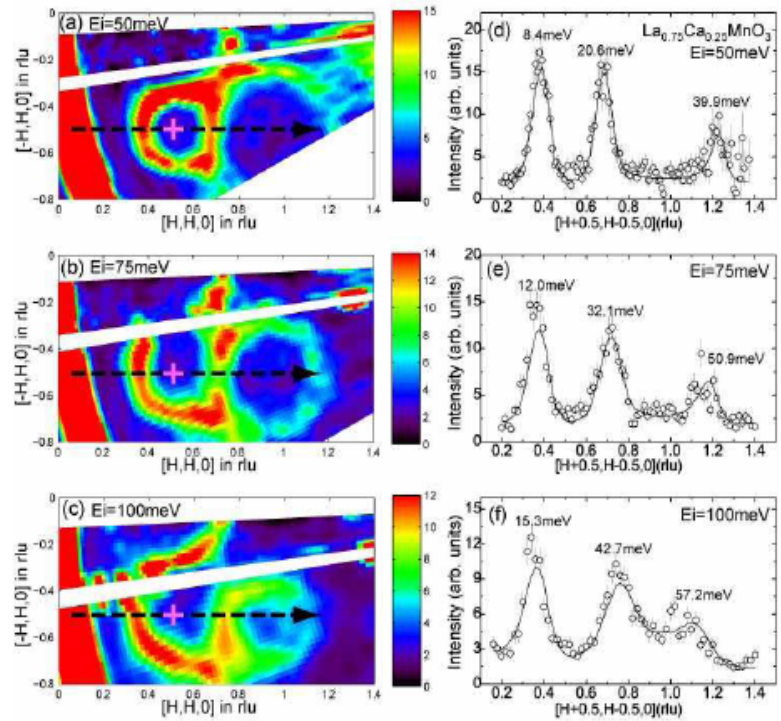
Data visualization

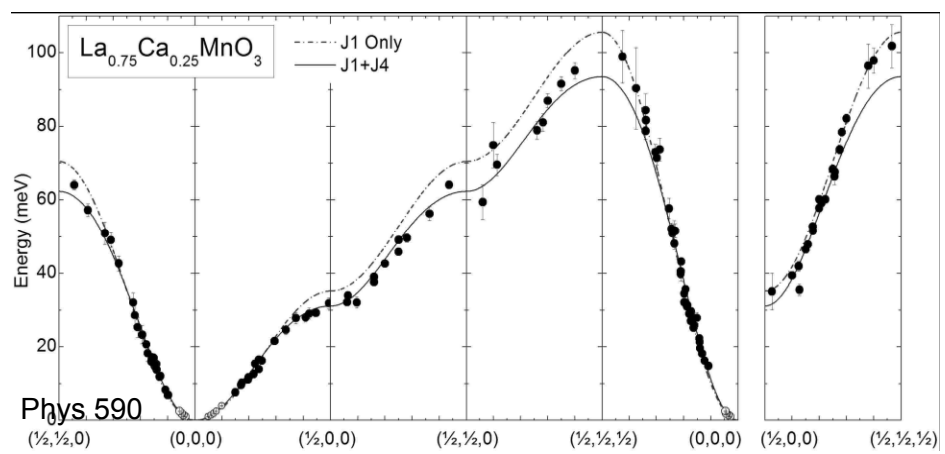
- Large, complex data from spallation sources
- Measure S(**Q**,ω) – 4D function

$La_{1-x}Ca_xMnO_3$

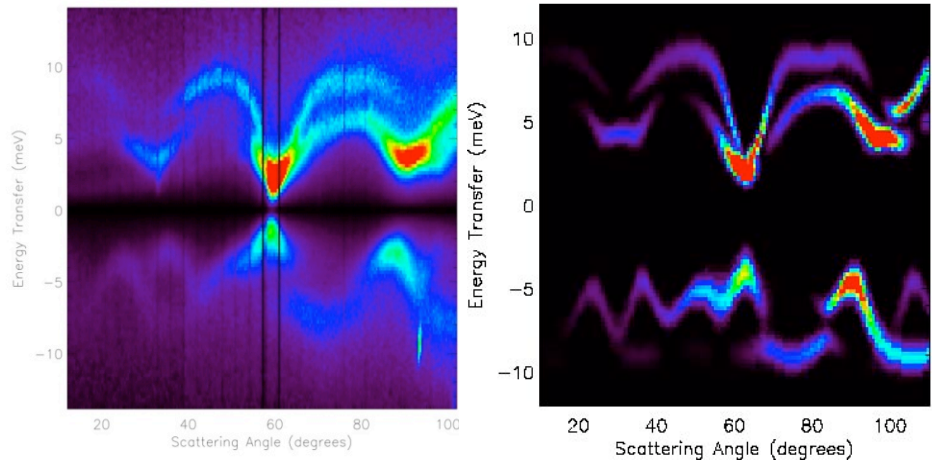


Ye *et al., Phys. Rev. B*, **75** 144408 (2007).

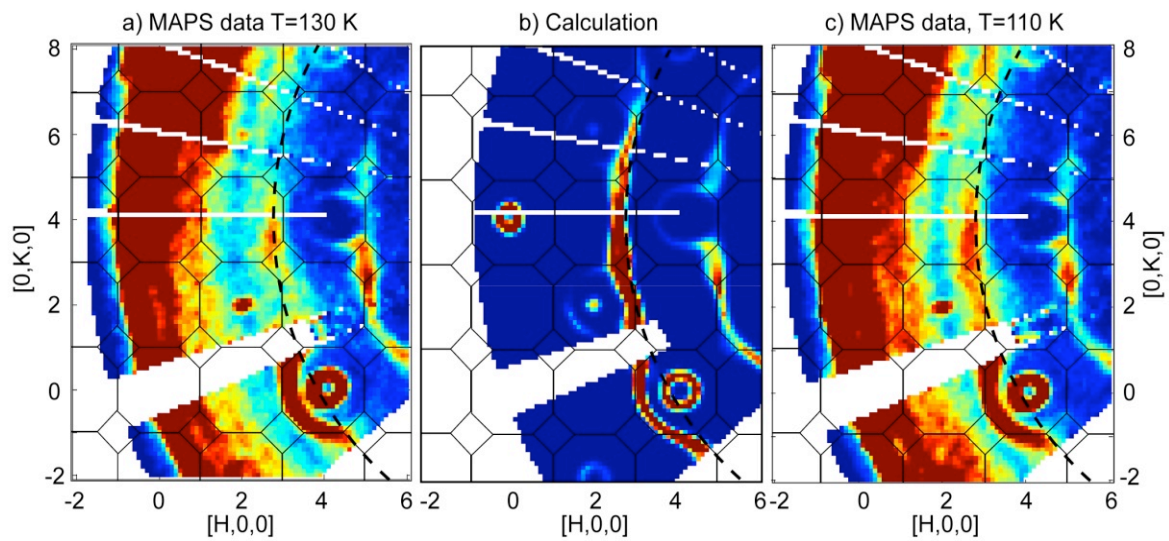




Computation



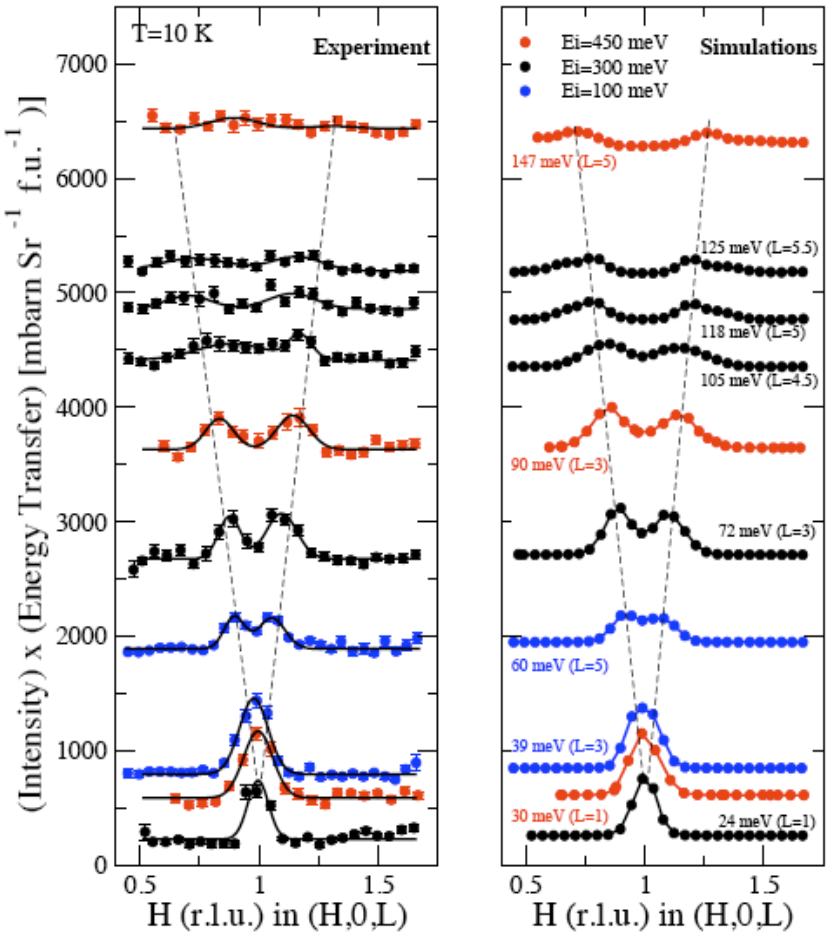
Pb phonons



Fe_3O_4 spin waves

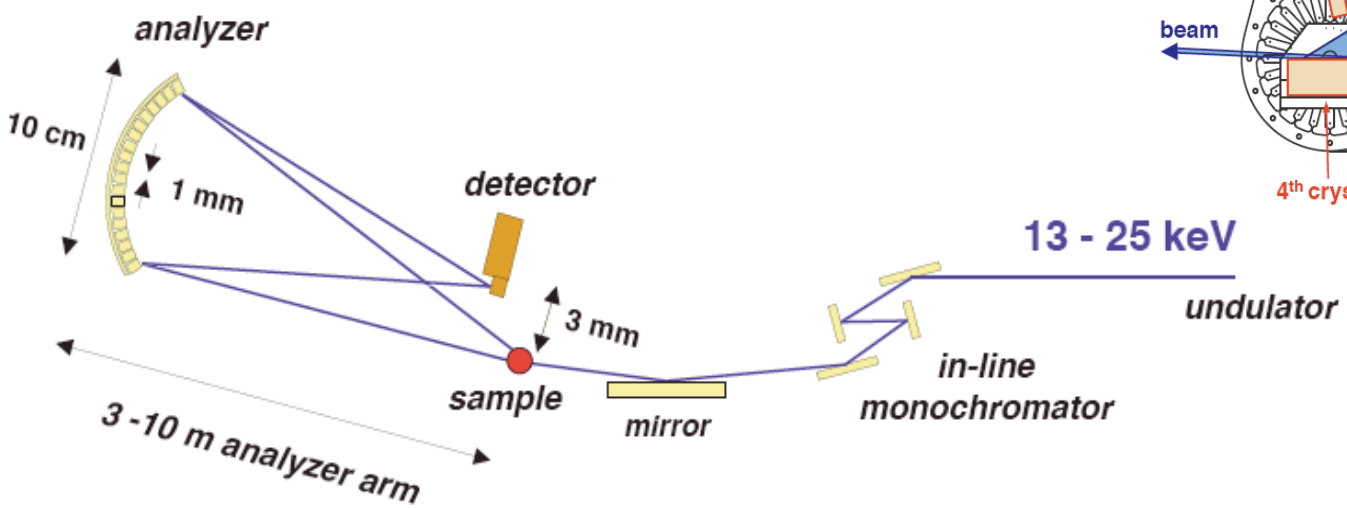
Absolute normalization

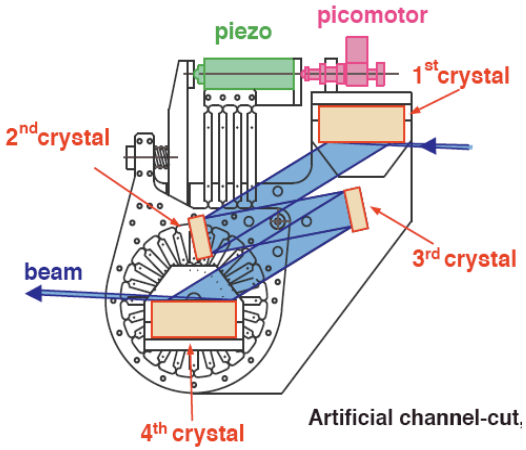
- **Absolute normalization**
  - Using incoherent scattering from vanadium
  - $\sigma/4\pi$ = 404 mbarns/Sr



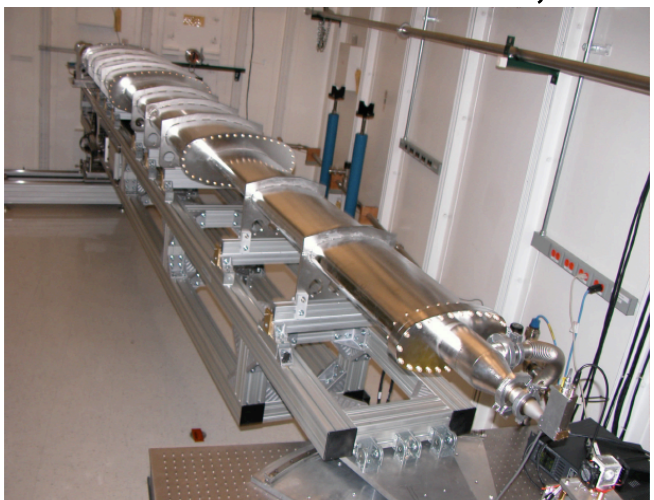
Inelastic x-ray scattering

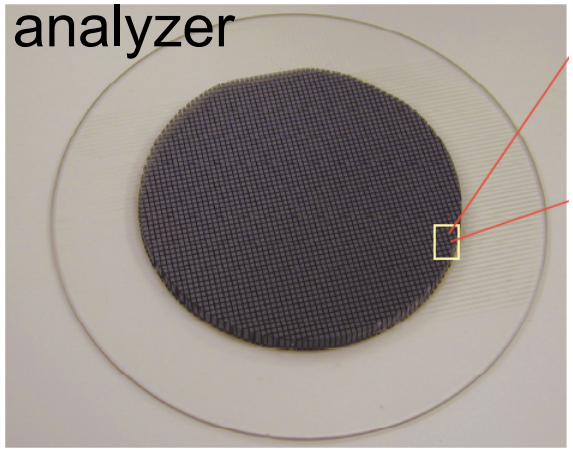
- Ei=20 keV, need 1 meV resolution
- $\Delta E/E_i \sim 10^{-7}$!!!





Sector-3, APS





ϕ-scan of monochromator
1 meV $\Rightarrow$ μrad

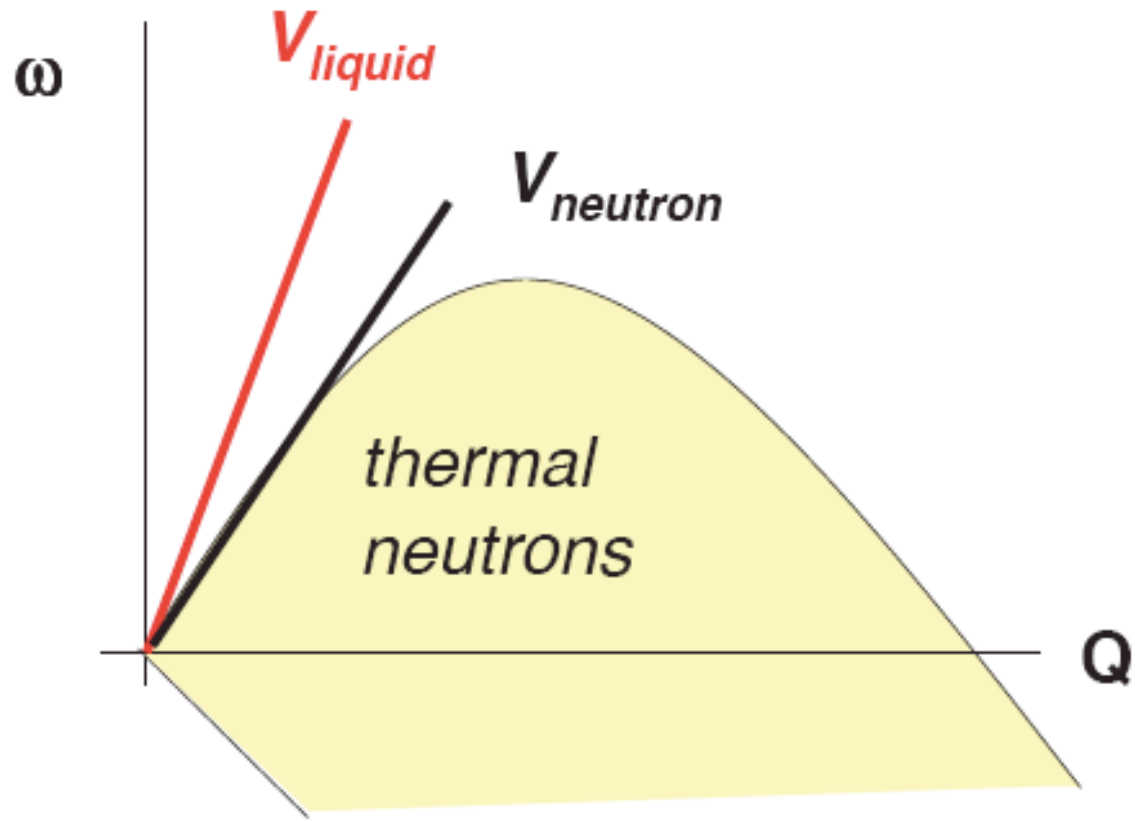
T-scan of monochromator
1 meV $\Rightarrow$ 0.02 K

Kinematics

- Essentially elastic scattering
- No kinematic limits

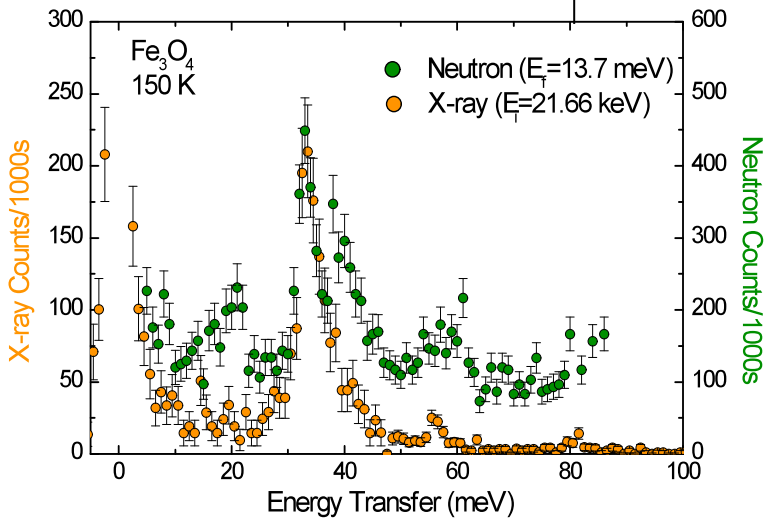
$$Q \approx 2k_i \sin\theta$$

$$\hbar\omega = \hbar c(k_i - k_f)$$

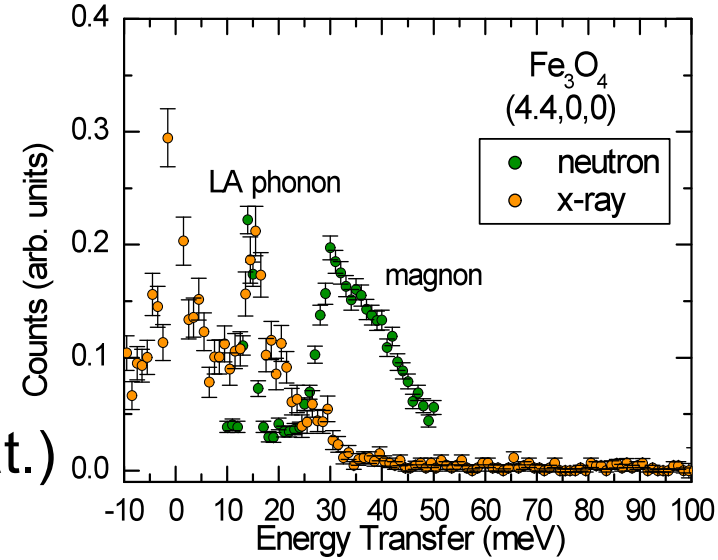


IXS vs. INS

- SAMPLE SIZE



- **IXS**
  - Simple scattering geometry ($k_i \approx k_f$)
  - Resolution function simpler
    (most angles fixed, E-scans only)
  - No spurions
    (high-order refs. keV, no incoherent scat.)
  - *Can only do lattice excitations*



References

AMES LABORATORY

General neutron scattering

G. Squires, “Intro to theory of thermal neutron scattering”, Dover, 1978.
S. Lovesey, “Theory of neutron scattering from condensed matter”, Oxford, 1984.
R. Pynn, http://www.mrl.ucsb.edu/~pynn/.

Polarized neutron scattering

Moon, Koehler, Riste, Phys. Rev **181**, 920 (1969).

Triple-axis techniques

Shirane, Shapiro, Tranquada, “Neutron scattering with a triple-axis spectrometer”, Cambridge, 2002.

Time-of-flight techniques

B. Fultz, http://www.cacr.caltech.edu/projects/danse/ARCS_Book_16x.pdf