Principles of X-ray and Neutron Scattering

Lecture 8: Inelastic Neutron Scattering to Investigate Dynamics

14. 02. '24

Lectures by: Prof. Philip Willmott, Prof. Johan Chang and **Dr. Artur Glavic**

Course Outline

Monday	Tuesday	Wednesday	Thursday	Friday
Lecture 1 10-10h45 Philip	**Lecture 4** 10-10h45 Philip	**Lecture 7** 10-10h45 Artur	**Lecture 10** 10-10h45 Artur	**Lecture 13** 10-10h45 Johan
Lecture 2 11-11h45 Philip	**Lecture 5** 11-11h45 Philip	**Lecture 8** 11-11h45 Artur	**Lecture 11** 11-11h45 Artur	**Lecture 14** 11-11h45 Johan
Lunch - Mensa	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**	**Lunch - Mensa**
Lecture 3 13h00-13h45 Philip	**Lecture 6** 13h00-13h45 Philip	**Lecture 9** 13h00-13h45 Artur	**Lecture 12** 13h00-13h45 Artur	**Lecture 15** 13h00-13h45 Johan
		Exercise Class 14h30-16		Exercise Class 14h30-16

X-ray scattering

Neutron Scattering

Resonant x-ray scattering

Neutron Lectures:

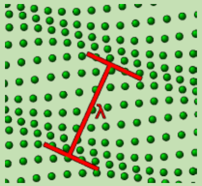
7: Neutrons & Scattering to Determine Structure
8: Inelastic Neutron Scattering to Investigate Dynamics
9: Magnetic Scattering
10: Neutron Polarization Analysis
11: Studying quantum matter for nanoscale applications
12: Neutron Instrument Development

Lecture 9: Inelastic Neutron Scattering to Investigate Dynamics

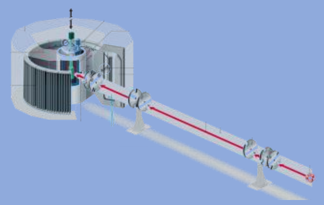
Theoretical Background

- Scattering from time dependent structures
- The correlation function and quasi particle excitations

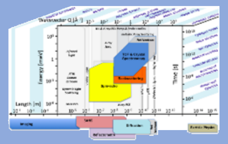
$S(\vec{Q},\omega)$



Practical Implementation

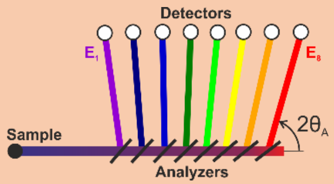


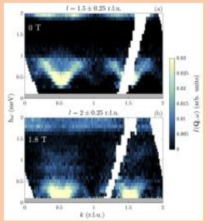
- Neutron time of flight technique and pulsed sources
- Inelastic neutron scattering techniques and range of application



Example Application

- SDW in frustrated magnet Cs_2CoBr_4





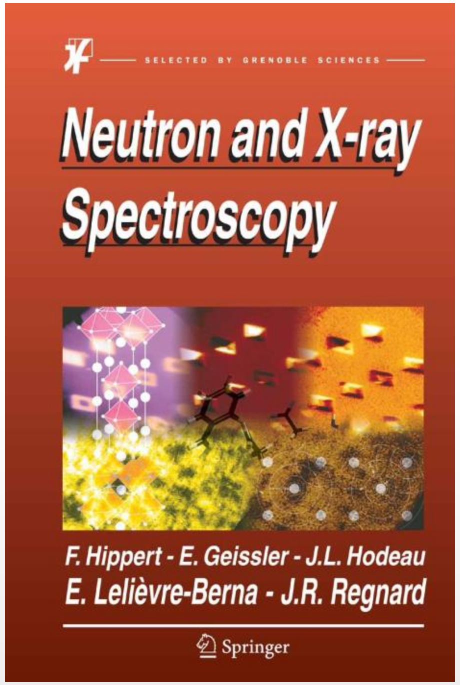
Further Reading

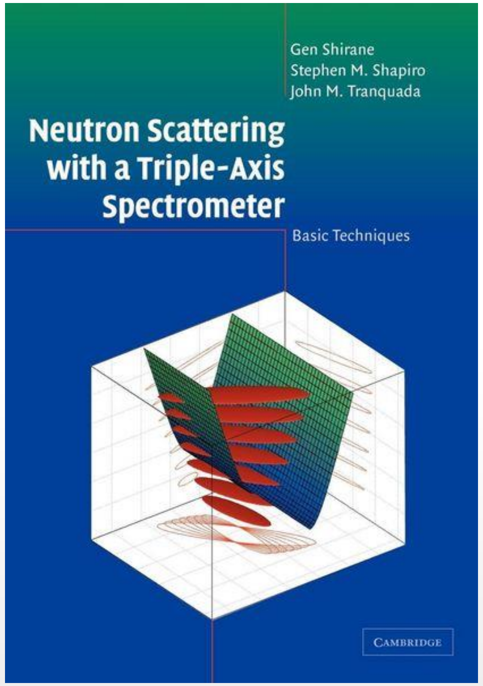
- "Introduction to the Theory of Thermal Neutron Scattering"
  G. L. Squires
  Dover Publication (1978)
- "Theory of Neutron Scattering from Condensed Matter" Vol.I/II.
  S. W. Lovesey
  Oxford Science Publications (1984).

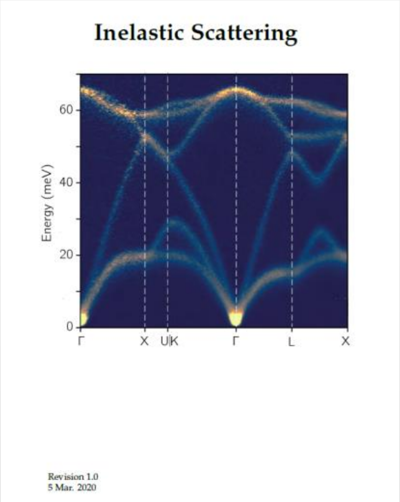
- "Neutron Scattering with a Triple-Axis Spectrometer: Basic Techniques"
  G. Shirane, S. M. Shapiro, J. M. Tranquada
  Cambridge University Press

- "Inelastic Scattering" (more TOF-centric)
  B. Fulz *et al.*
  https://www.its.caltech.edu/~matsci/btfgrp/Inelastic_Neutron_Book.pdf

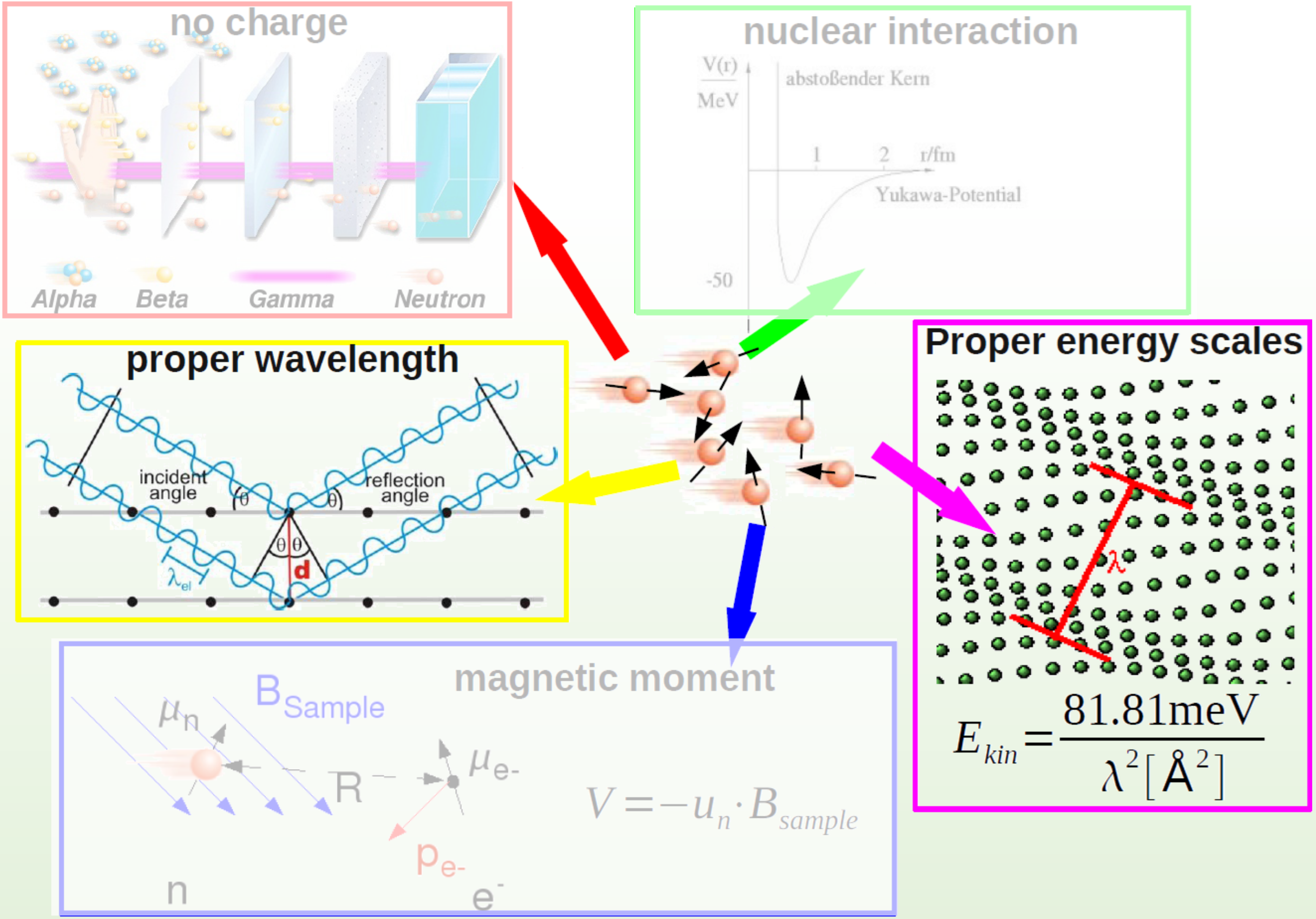
- "Neutron & X-ray Spectroscopy"
  F. Hippert, E. Geissler, J. L. Hodeau, E. Lelievre-Berna, J. R. Regnard
  Grenoble Sciences, Springer







Reminder: Why Neutrons?



Double Differential Cross-Section

Fermi's Golden Rule (elastic $(k = k')$):

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar}\right)^2 \Big|\underbrace{\langle\vec{k}'|V|\vec{k}\rangle}_{\int e^{i(\vec{k}-\vec{k}')\vec{r}'} V(\vec{r'})\ d^3\vec{r}'}\Big|^2 \delta\left(E_{k'} - E_k\right)$$

Change in sample state *I* to *F* (inelastic) :

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\left(\frac{m_n}{2\pi\hbar}\right)^2 \left|\langle\vec{k}', F|V|\vec{k}, I\rangle\right|^2 \delta\left(E_{k'} - E_k + \hbar\omega_{I\to F}\right)$$

Using the Fermi pseudo-potential one can derive (see e.g. Squires):

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\sum_{j,j'} b_j b_j' \sum_{k,k'} P(I) \left|\langle F|e^{i\vec{Q}\vec{r}_j}|I\rangle\right|^2 \delta\left(E_{k'} - E_k + \hbar\omega_{I\to F}\right)$$

With the probability for the system to be thermally excited to the initial state E_I:

$$P(I) = \frac{e^{-\frac{E_I}{k_B T}}}{\sum_n e^{-\frac{E_n}{k_B T}}}$$

Double Differential Cross-Section

Fermi's Golden Rule (elastic $(k = k')$):

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar}\right)^2 \left|\underbrace{\langle\vec{k}'|V|\vec{k}\rangle}_{\int e^{i(\vec{k}-\vec{k}')\vec{r}'} V(\vec{r}')\ d^3\vec{r}'}\right|^2 \delta\left(E_{k'} - E_k\right)$$

Change in sample state *I* to *F* (inelastic) :

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\left(\frac{m_n}{2\pi\hbar}\right)^2 \left|\langle\vec{k}', F|V|\vec{k}, I\rangle\right|^2 \delta\left(E_{k'} - E_k + \hbar\omega_{I\to F}\right)$$

Using the Fermi pseudo-potential and a few standard tricks one can derive (see e.g. Squires):

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\frac{1}{2\pi\hbar}\sum_{j,j'} b_j b'_j \int \left\langle e^{i\vec{Q}(\vec{r}_{j'}(t)-\vec{r}_j(0))}\right\rangle e^{-i\omega t} dt$$

The measured double differential scattering cross-section is a Fourier transform in space and time!

It measures "where atoms are and how they move".

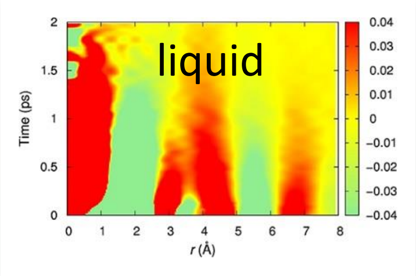
Correlation Functions

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\frac{\sigma_{coh}}{4\pi}\sum_{j,j'}\int\left\langle e^{i\vec{Q}(\vec{r}_{j'}(t)-\vec{r}_j(0)}\right\rangle e^{-i\omega t}dt = \frac{k'}{k}\frac{\sigma_{coh}}{8\pi^2\hbar}NS(\vec{Q},\omega)$$

scattering potential

$$V(\vec{r},t) \xrightarrow[\text{correlation}]{\text{time and space}} G(\vec{r},t) = \frac{1}{(2\pi)^3}\int I(\vec{Q},t)e^{-i\vec{Q}\vec{r}}d^3\vec{Q}$$

pair correlation function

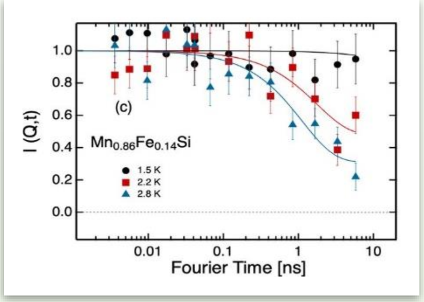


$\mathscr{F}(\vec{r})$

$\mathscr{F}(\vec{r},t)$

$$I(\vec{Q},t) = \frac{1}{N}\sum_{j,j'}\left\langle e^{i\vec{Q}(\vec{r}_{j'}(t)-\vec{r}_j(0)}\right\rangle e^{-i\omega t}$$

Intermediate scattering function

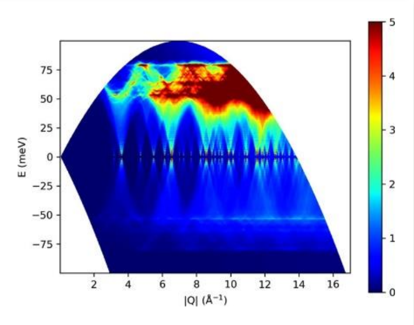


$\mathscr{F}(t)$

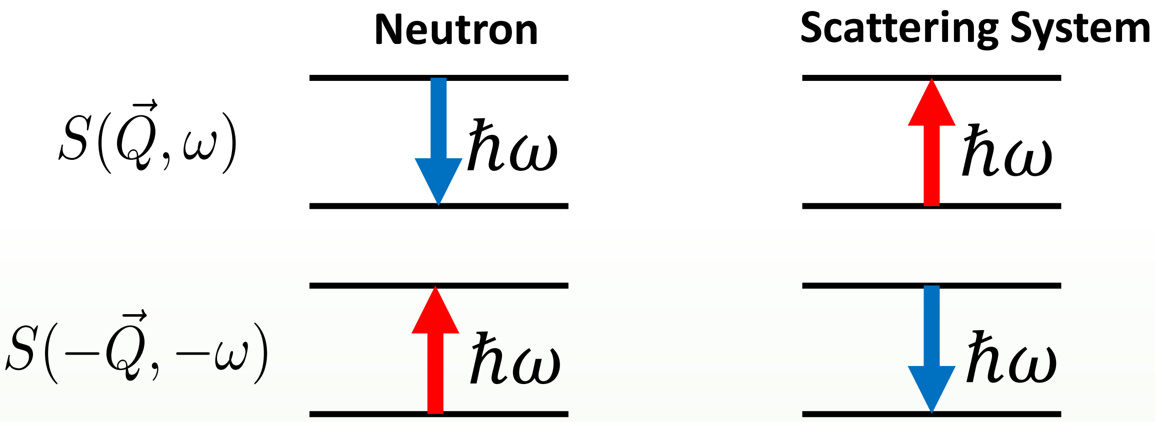
$$\Phi(\vec{Q},\omega) \xrightarrow[\text{squared}]{\text{absolute}} S(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\int I(\vec{Q},t)e^{-i\omega t}dt$$

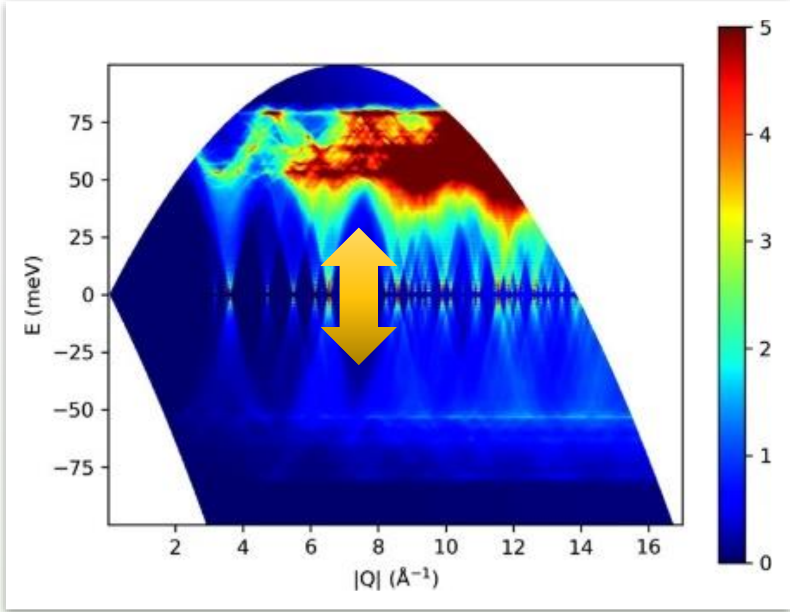
scattered wave function

scattering function



Detailed Balance





$S\left(\vec{Q},\omega\right)$ has an important property known as ***detailed balance***

$$S\left(-\vec{Q},-\omega\right) = e^{-\frac{\hbar\omega}{k_B T}} S\left(\vec{Q},\omega\right)$$

Meaning:

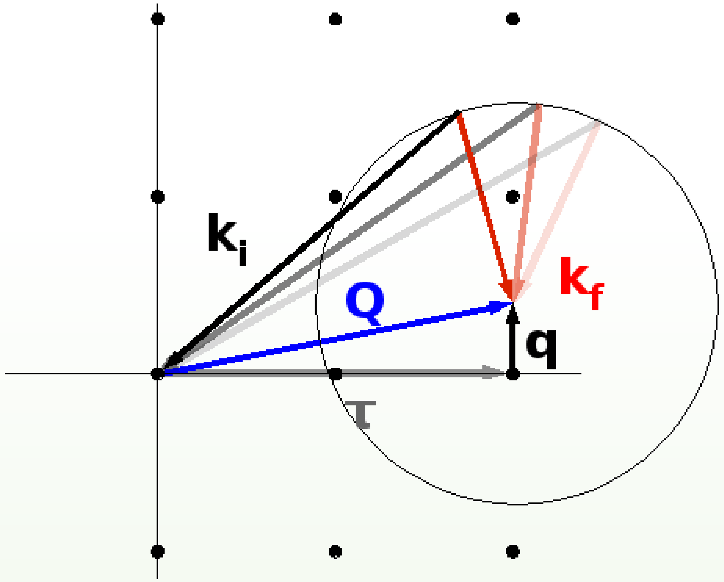
- ω > 0: Creation of excitation in scattering system (down scattering of neutron)
- ω < 0: Annihilation of excitation (up scattering of neutron)
- Weight for both cases depends on temperature!
- At T = 0 K no modes can be annihilated, because there are none!

Can be derived directly from $\frac{d^2\sigma}{d\Omega dE'}$

and thermal state population:

$$P(I) = \frac{e^{-\frac{E_I}{k_B T}}}{\sum_n e^{-\frac{E_n}{k_B T}}}$$

Quasi-particle Excitations



$$S(\vec{Q},\omega) = \frac{\chi''(\vec{Q},\omega)}{1 - e^{-\hbar\omega/k_B T}}$$

where χ'' is imaginary part of dynamic susceptibility (see fluctuation-dissipation theorem)

Creation Annihilation Phonon Mode *s*

$$\chi''(\mathbf{Q},\omega) = \frac{1}{2}\frac{(2\pi)^3}{v_0}\sum_{\tau,q}\delta^3(\mathbf{Q}-q-\tau)\sum_s \frac{1}{\omega_{qs}}|F(\mathbf{Q})|^2\left[\delta(\omega-\omega_{qs})-\delta(\omega+\omega_{qs})\right]$$

With the dynamic structure factor:

$$F(\mathbf{Q}) = \sum_j \frac{b_j}{\sqrt{m_j}}(\mathbf{Q}\cdot\xi_{js})e^{i\mathbf{Q}\cdot\mathbf{d}_j}e^{-W_j}$$

Debye-Waller factor of atom *j*

Scattering length of atom *j*

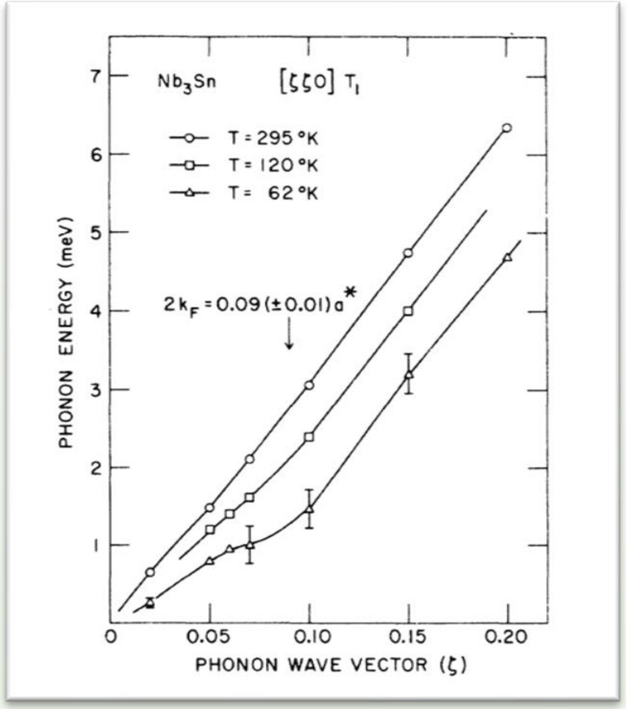
Mass of atom *j*

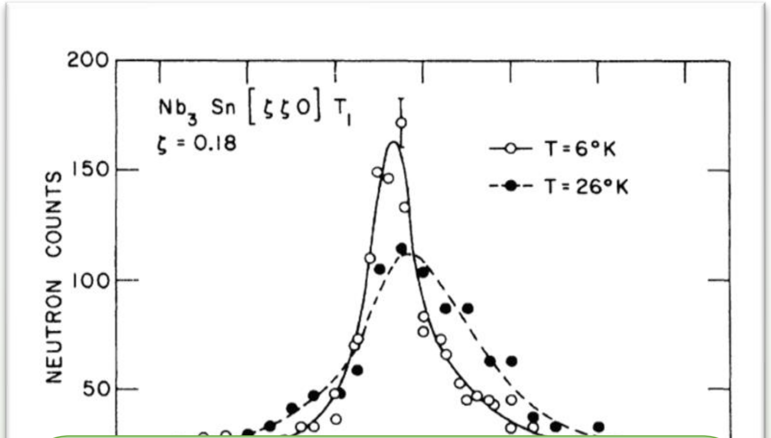
Polarization of Mode *s*

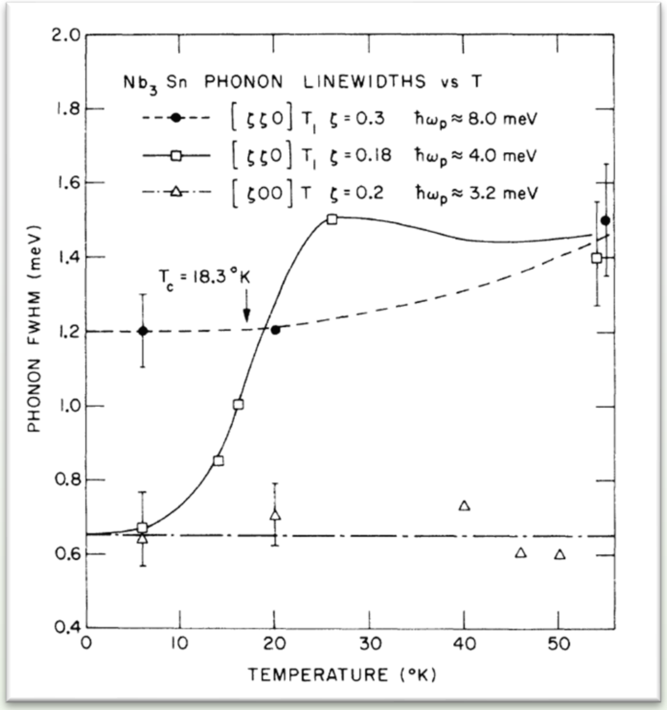
Pos. of atom *j* in unit cell

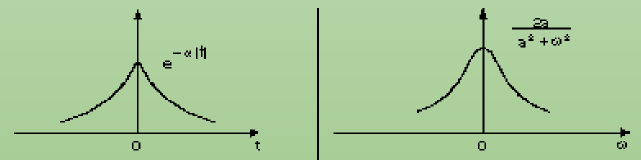
Phonon Lifetime

Phonons in conventional SC Nb_3Sn: J. D. Axe and G. Shirane, Phys. Rev. Lett. **30**, 214 *(1973);* Phys. Rev. B **8,** 1965 (1973).







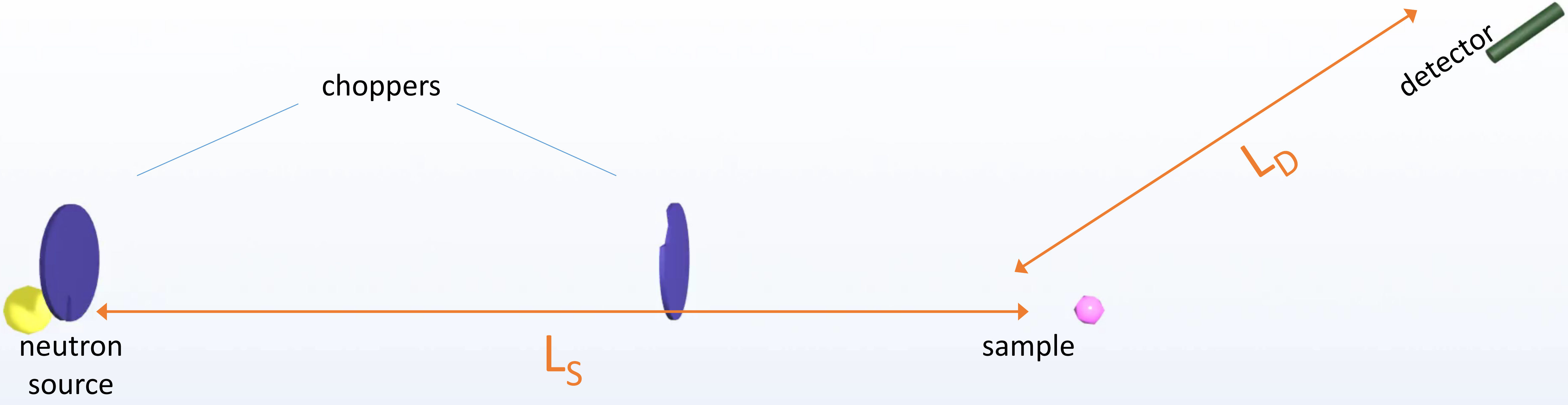


remember Frouier transform

$$\frac{1}{\omega_{qs}}\delta\left(\omega\pm\omega_{qs}\right)\rightarrow\frac{1}{\pi\omega'_{qs}}\cdots\frac{\cdots}{\left[\omega\pm\omega'_{qs}\right]^2+\Gamma^2_{qs}}\quad \text{with } \omega'^2_{qs}=\omega^2_{qs}-\Gamma^2_{qs}$$

Γ_{qs} is the half-width at half-maximum (HWHM) and is related to the phonon lifetime via $\tau=\frac{\hbar}{2\Gamma_{qs}}$

Neutron time of flight technique



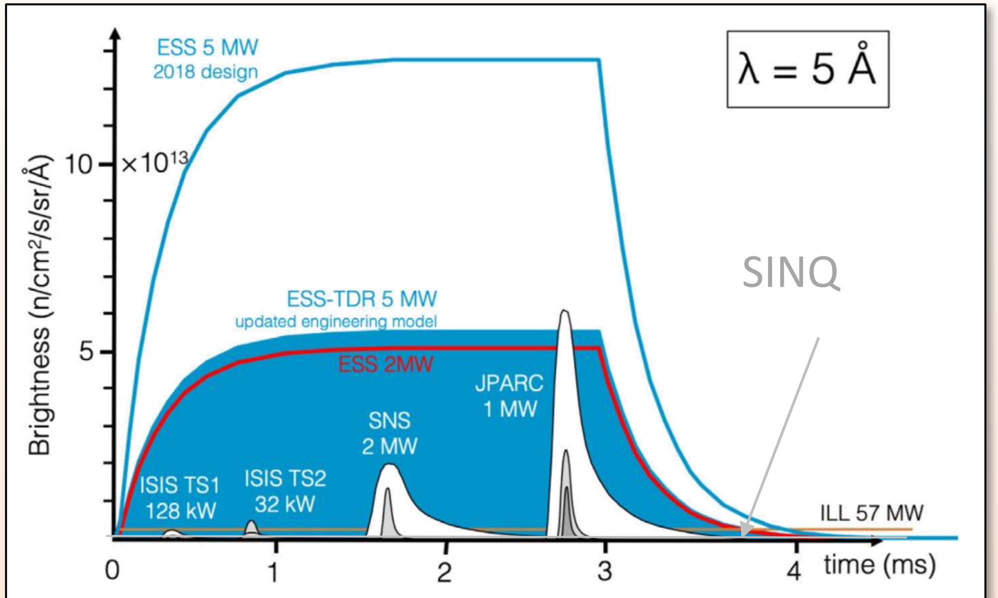
$$\lambda_n = \frac{h}{v_n m_n} = \frac{t_{ToF}}{L_S + L_D} \cdot 3956 \left[\frac{\text{Å}}{\text{m/s}}\right]$$

Pulsed neutron sources

Higher efficiency for ToF based instruments (usable / produced neutrons)

Example: European Spallation Source (ESS)

- Beam power up to 5 MW (5x SINQ)
- Usable cold neutrons 100x SINQ



K.H. Andersen, et al., **Nuc. Inst. Meth. A.**, 957 (2020), 163402

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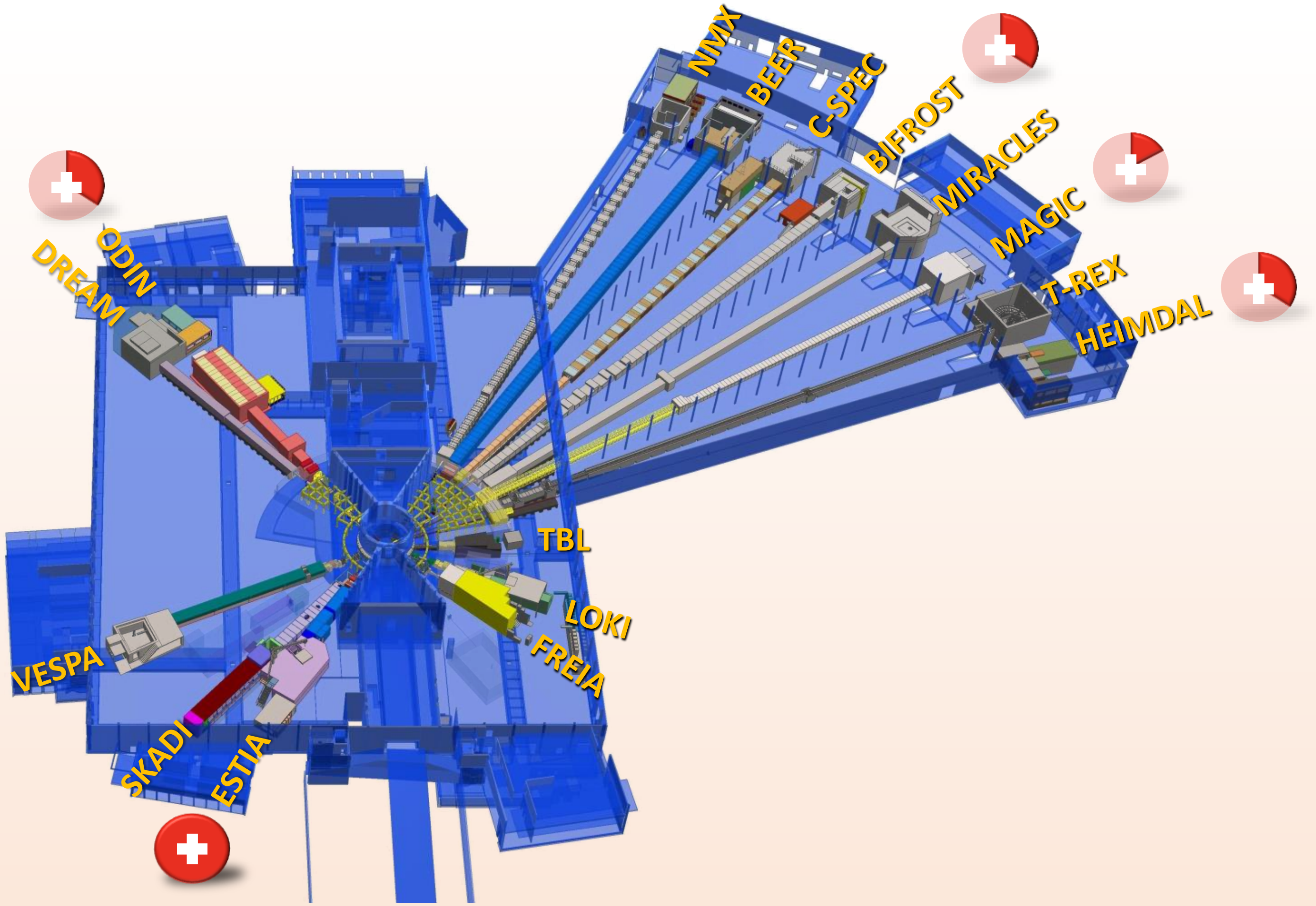

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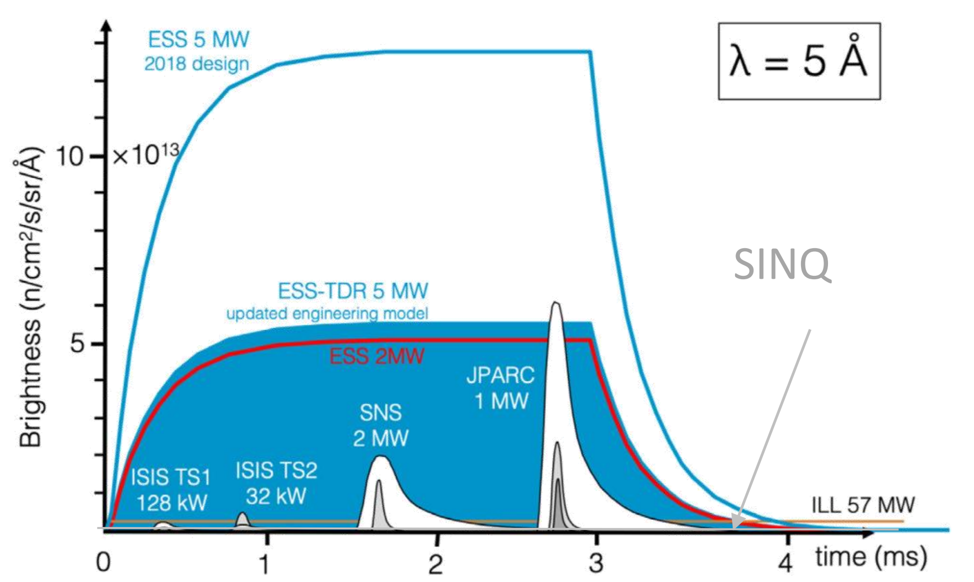
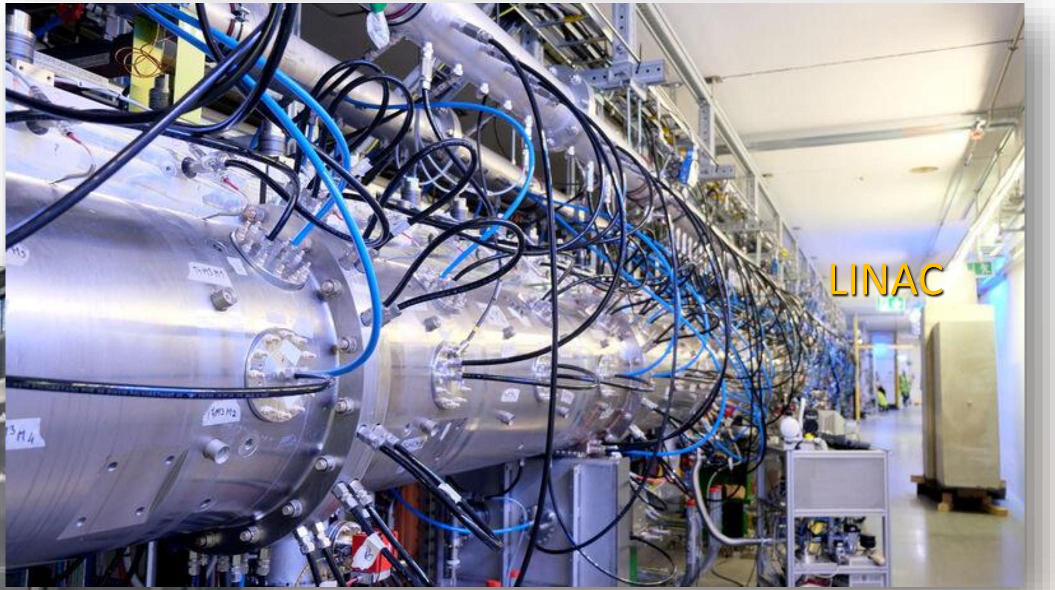
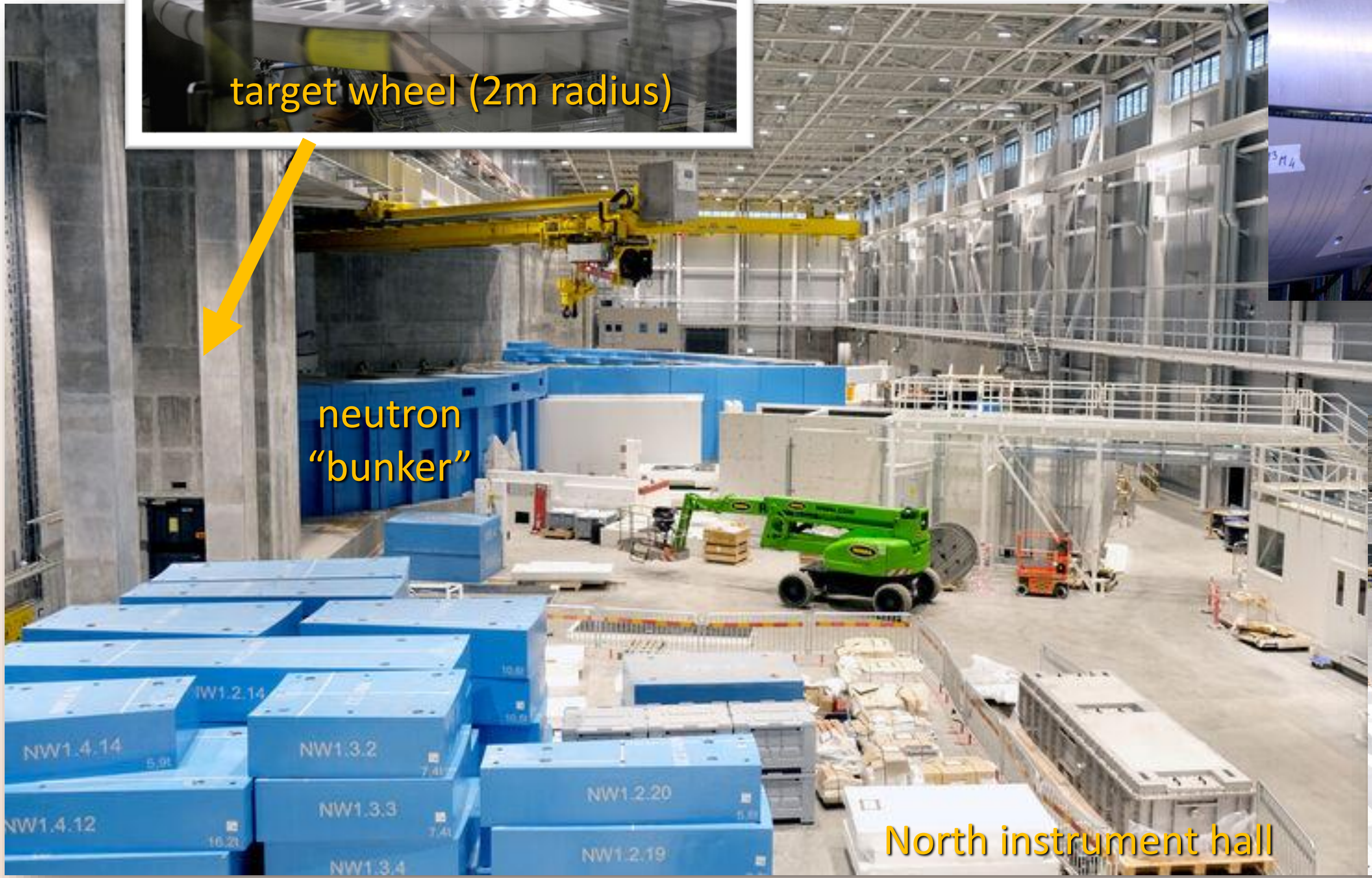


K.H. Andersen, et al., **Nuc. Inst. Meth. A.**, 957 (2020), 163402

ESS is huge


target wheel (2m radius)


LINAC

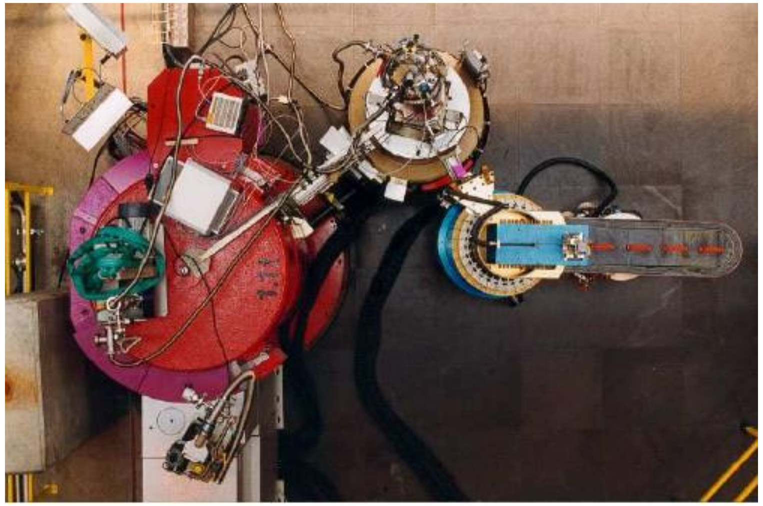

neutron
“bunker”
NW1.4.14
NW1.3.2
NW1.2.20
NW1.4.12
NW1.3.3
NW1.2.19
NW1.3.4
North instrument hall


long neutron guide hall (~150m)

Neutron spectrometers

Time-of-Flight Spectrometer

Triple-Axis Spectrometer



Spin-Echo Spectrometer

Backscattering Spectrometer

Neutron spectrometers (inelastic)

Triple Axis Spectroscopy

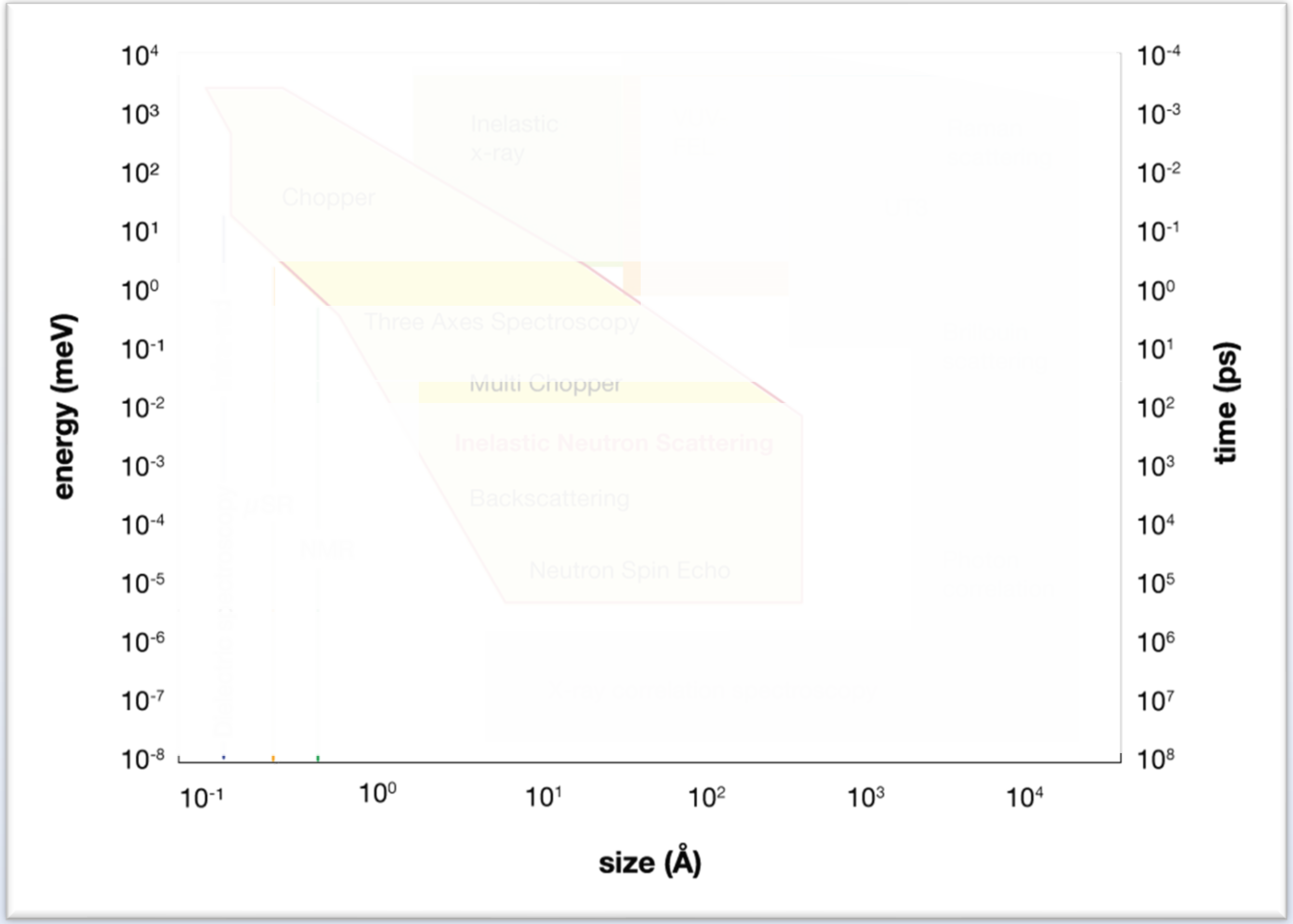
- High resolution
- Low background
- Simpler analysis
- Single point measurement

Time of Flight Spectroscopy

- Large energy- and q-range
- Fast measurement
- Profit from modern, pulsed sources
- Flexible binning
- Complex data reduction
- Possible spurious signals (spurion)

Specialized Techniques

- Backscattering spectroscopy
    - High absolute energy resolution
- Spin-echo
    - High energy transfer resolution



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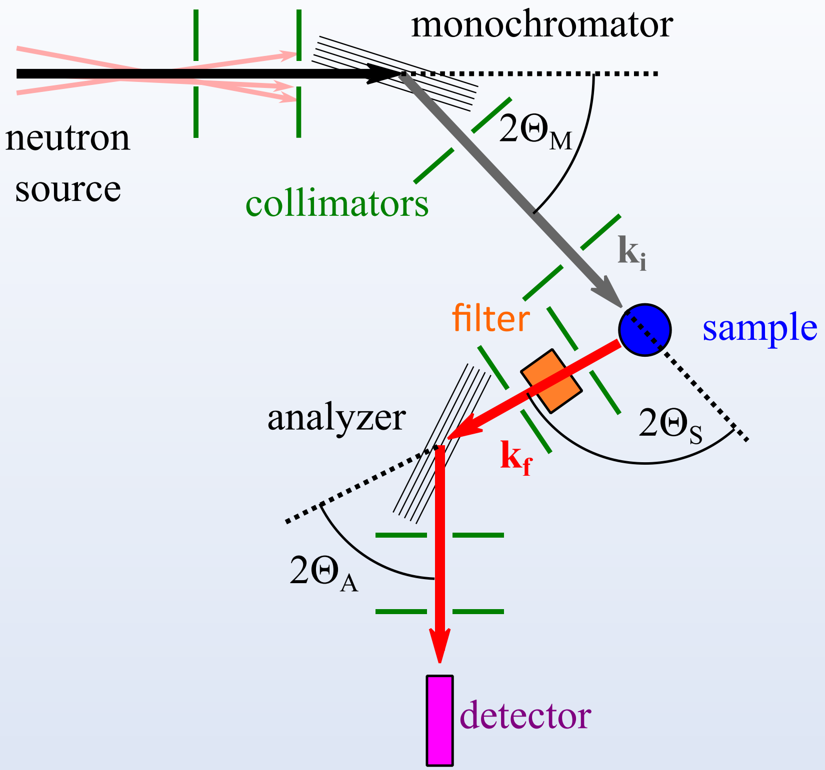

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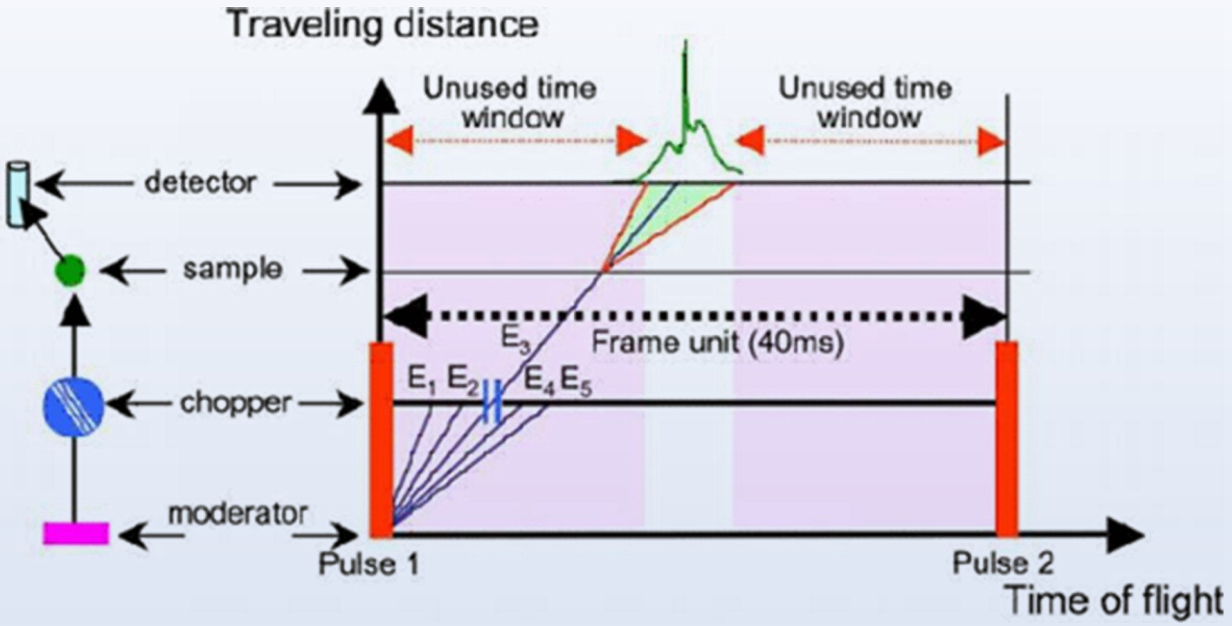
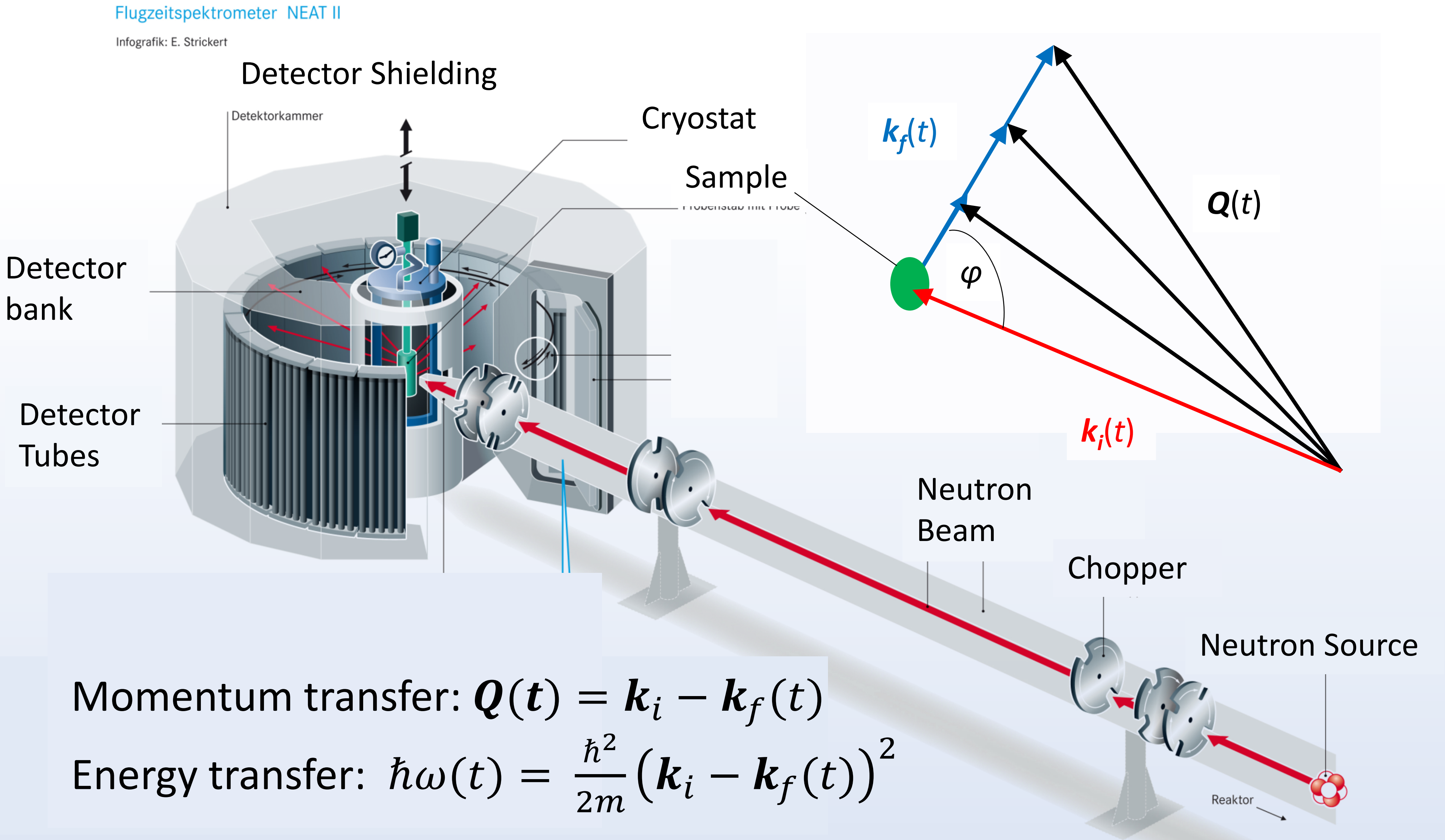


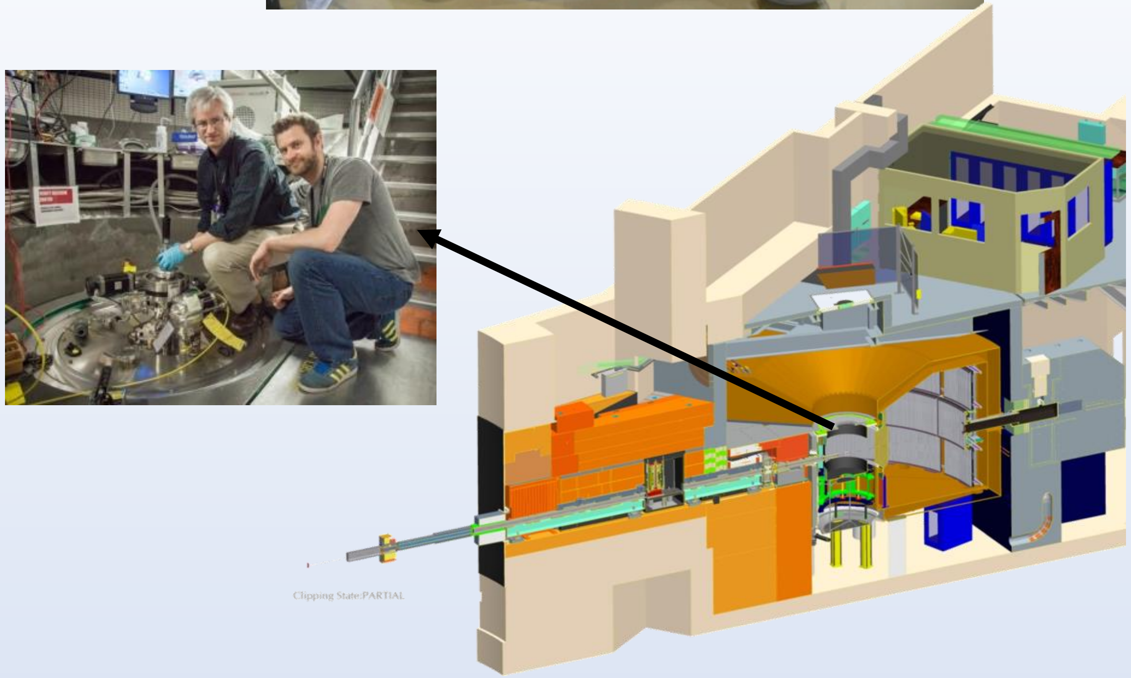
Time of Flight spectroscopy



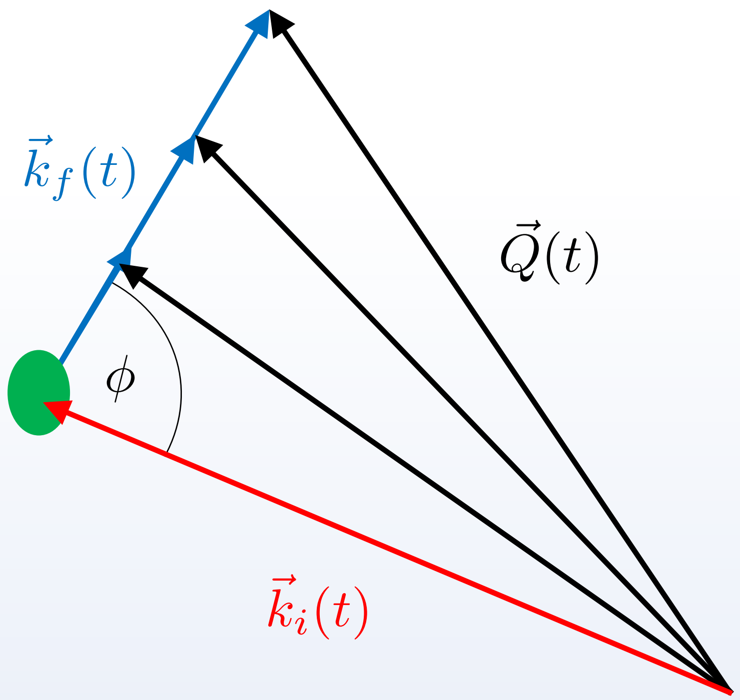
Momentum transfer: $\boldsymbol{Q}(\boldsymbol{t}) = \boldsymbol{k}_i - \boldsymbol{k}_f(t)$

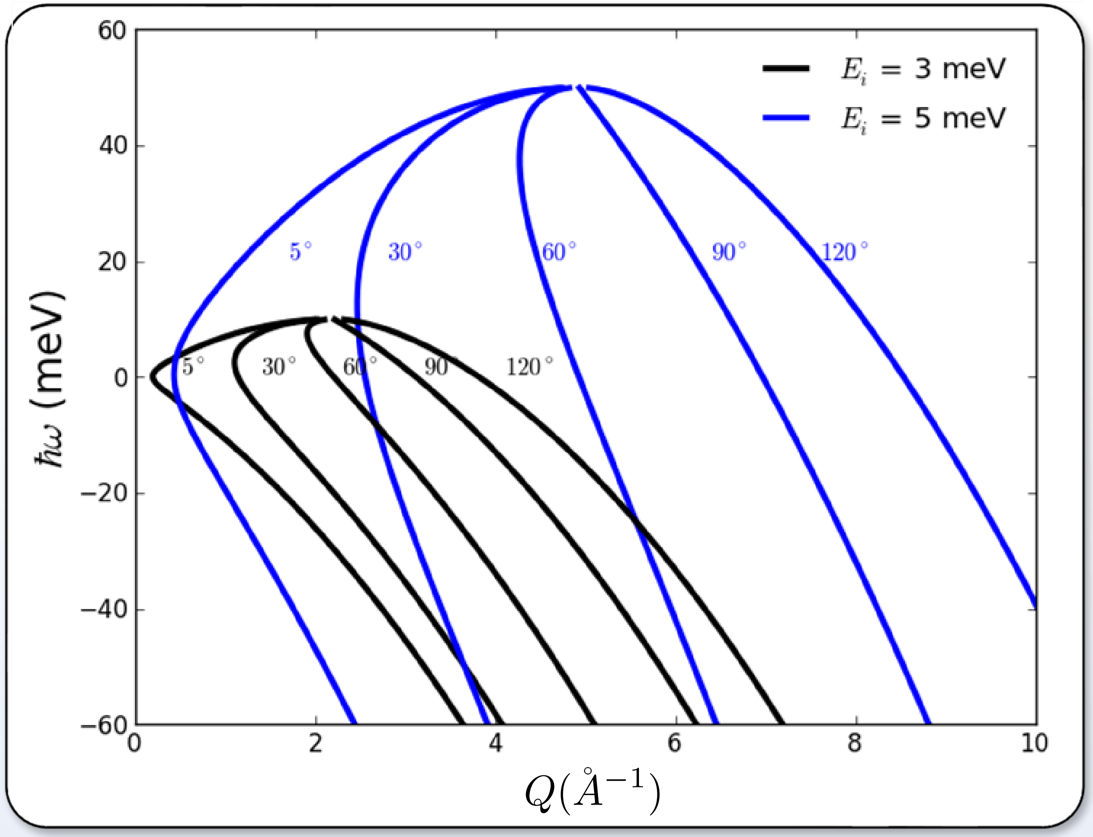
Energy transfer: $\hbar\omega(t) = \frac{\hbar^2}{2m}\left(\boldsymbol{k}_i - \boldsymbol{k}_f(t)\right)^2$

ToF spectrometers at different facilities



Time-Of-Flight Spectroscopy: Kinematic Conditions



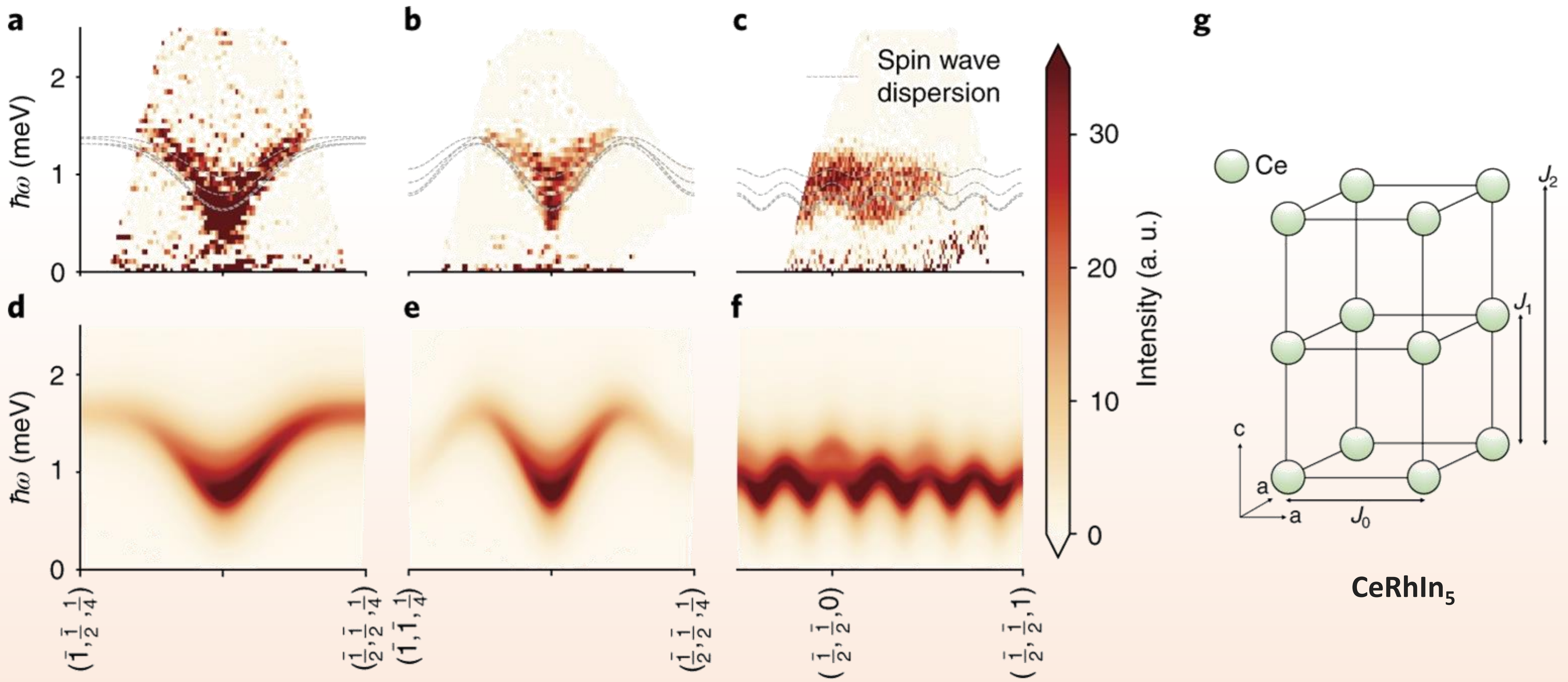


Cosine rule provides measurement range:

$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos\phi \qquad \hbar\omega = E_i - E_f$$

$$\frac{\hbar^2 Q^2}{2m_n} = E_i + E_f - 2\sqrt{E_i E_f}\cos\phi = 2E_i - \hbar\omega - 2\cos\phi\sqrt{E_i(E_i - \hbar\omega)}$$

Time-Of-Flight Spectroscopy: Quick Data Collection



CeRhIn$_5$

P. Das *et al.*, Phys. Rev. Lett. 113, 246403 (2014); D. M. Fobes *et al.*, Nature Physics 14, 456–460 (2018)

Spurious Scattering : Sample Environment in TOF

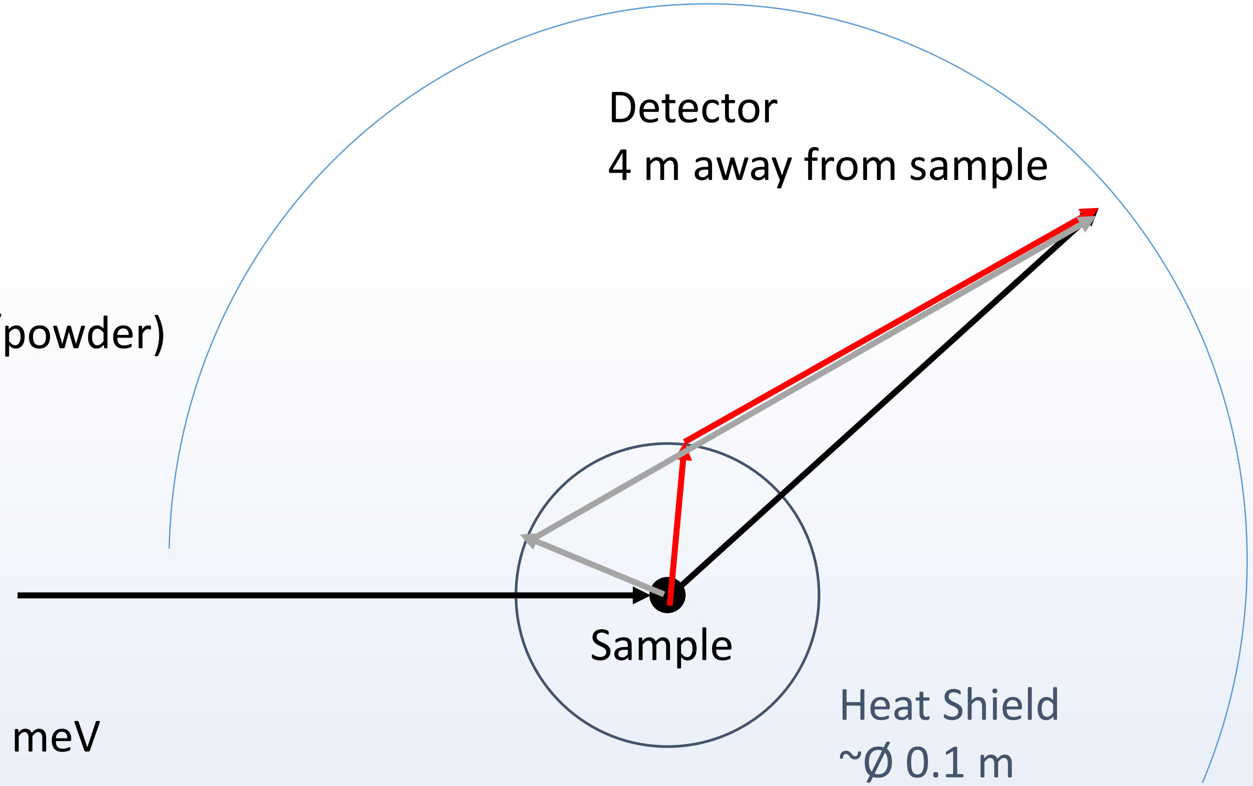
E_i = 500 meV ↔ v_i = 9.781 m/s

Typical TOF for E_i=E_f: t = 4 m / 9.781 m/s = 409 µs

Double scattering at container (elastic, incoherent/powder)
→additional flight path ~ 10 mm

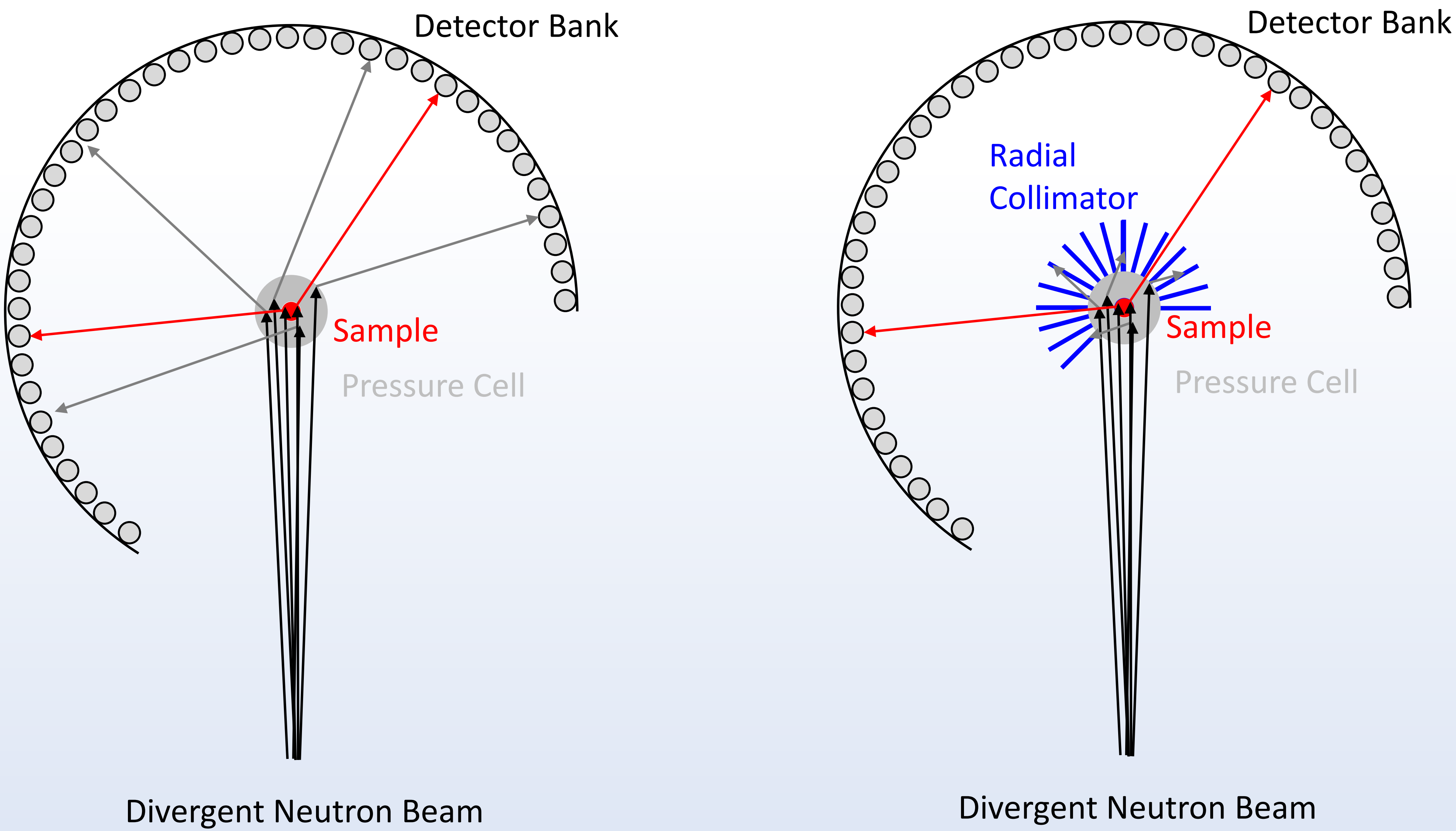
Additional flight time:
Δt = 0.1 m / 9.781 m/s = 10.2 µsec

Corresponds to virtually slow neutron with speed
v_f = 4 m / (t + Δt) = 9.542 m/s ↔ E_f= 476 meV

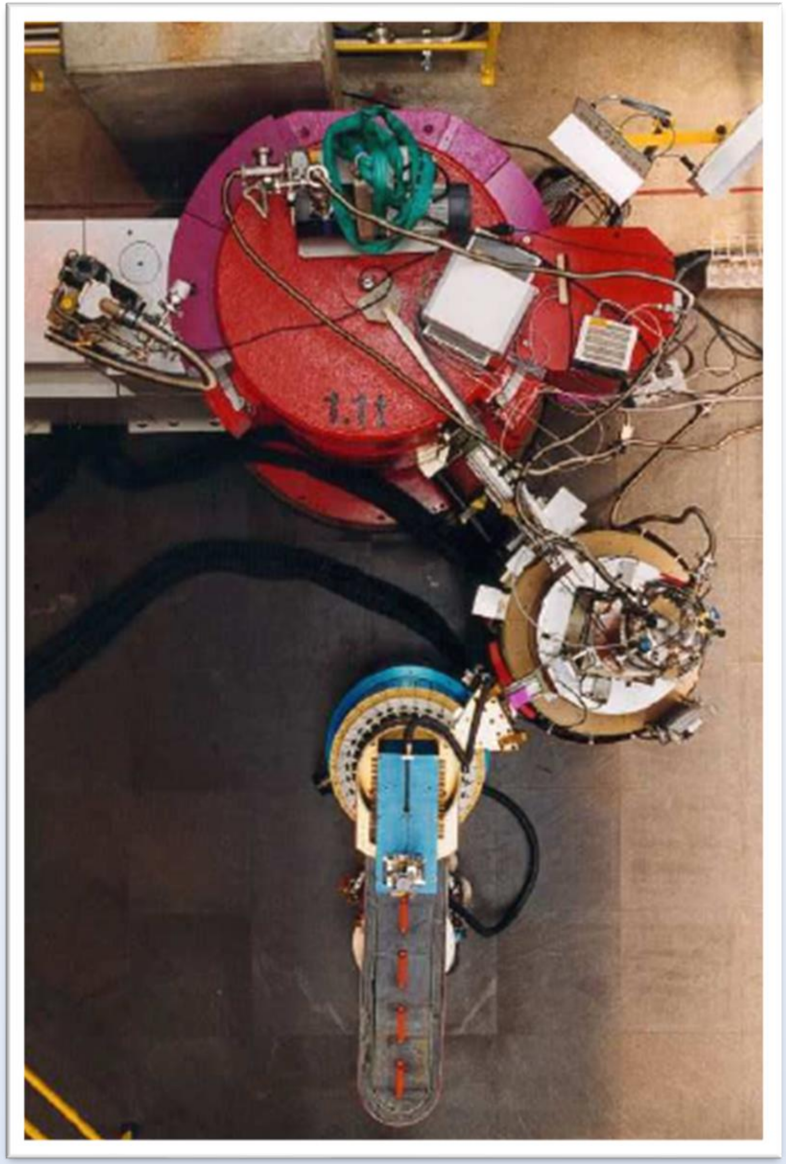


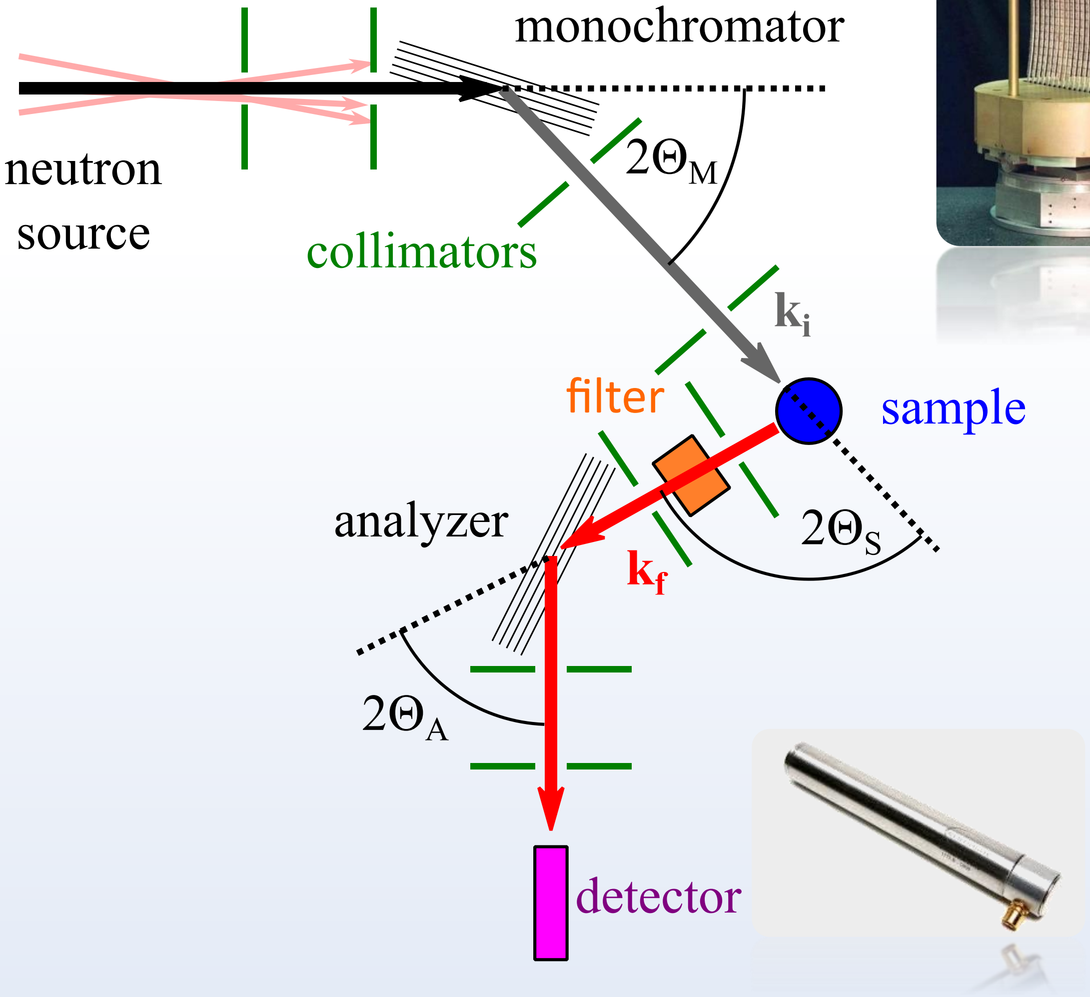
Easy to produce "virtual" energy transfers between 0-50 meV even for elastic scattering!!!!

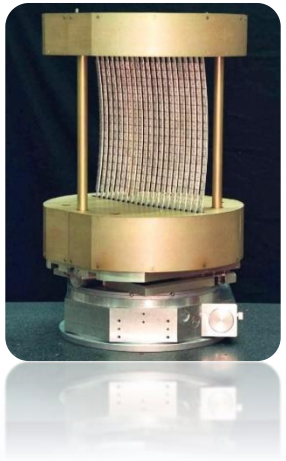
Improve background with Radial Collimator



Triple Axis Spectroscopy



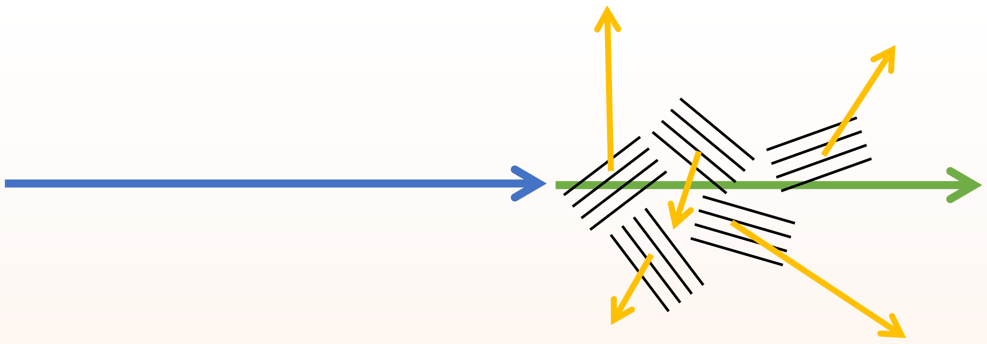




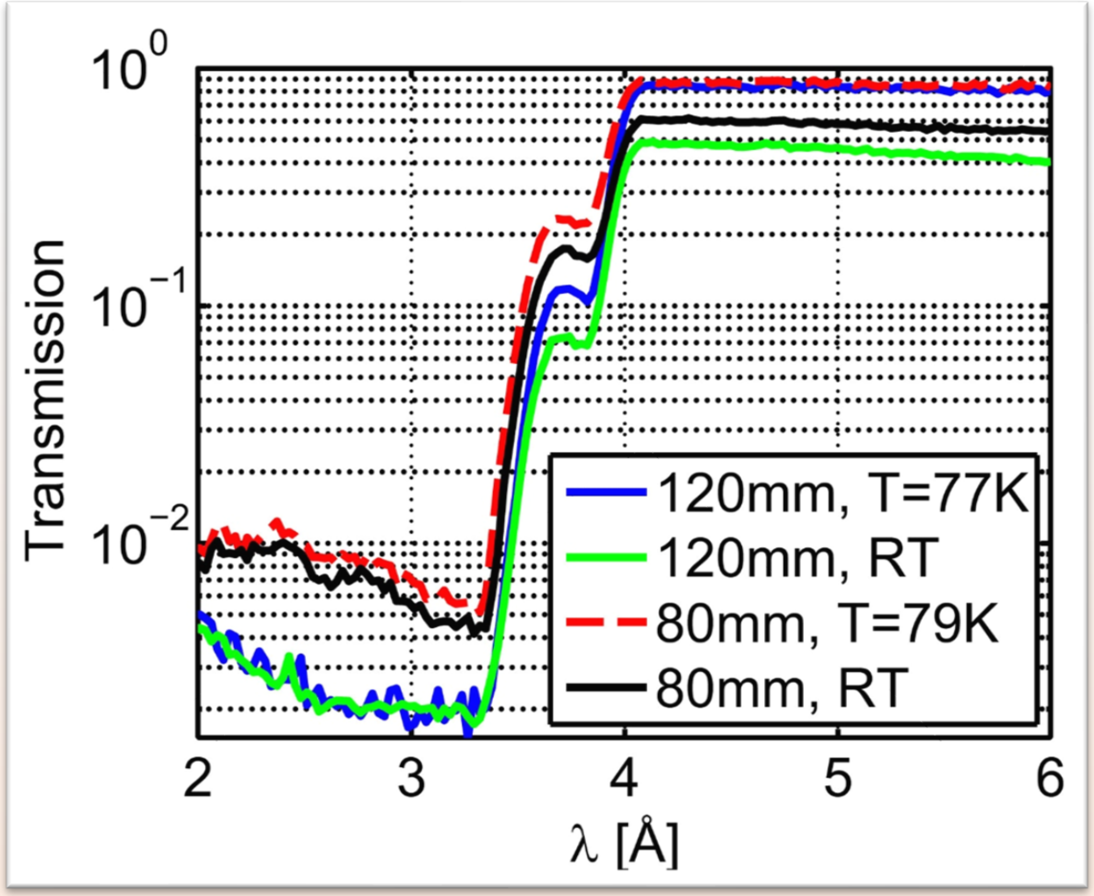
Note:
Clifford Shull & Bertram Brockhouse received
1994 Nobel Prize in Physics for invention of this technique!

Intermezzo – Bragg-edge imaging and filters

Bragg: $n\lambda = 2d\sin\theta$

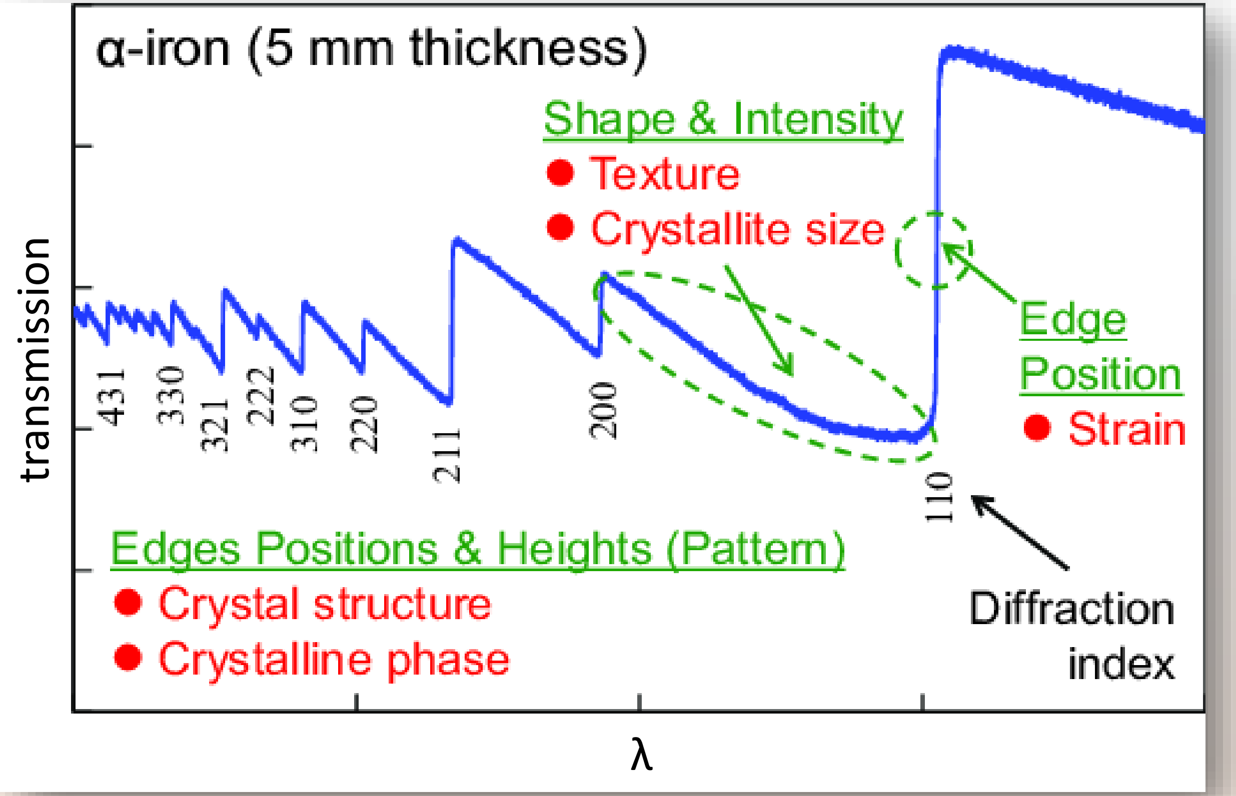


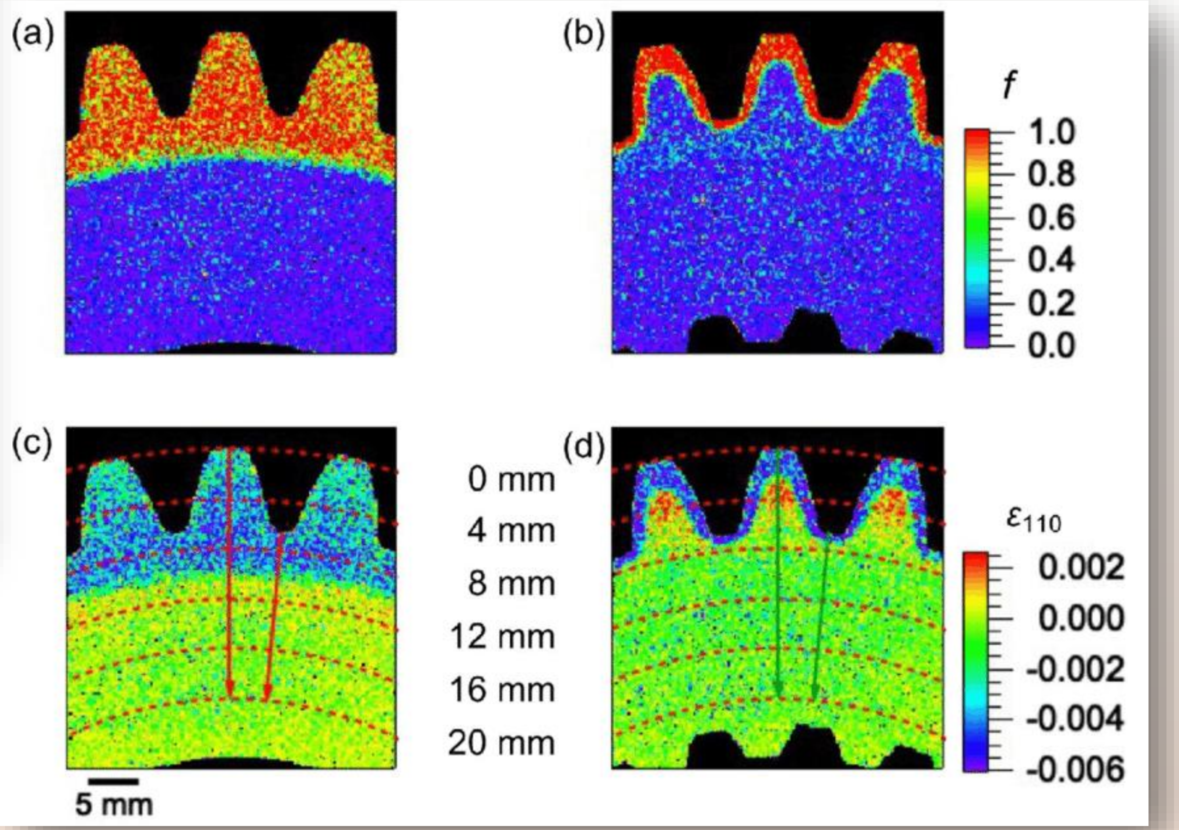
There is a cut-off for wavelengths larger than 2d!



If we use a crystal, why is filter needed?

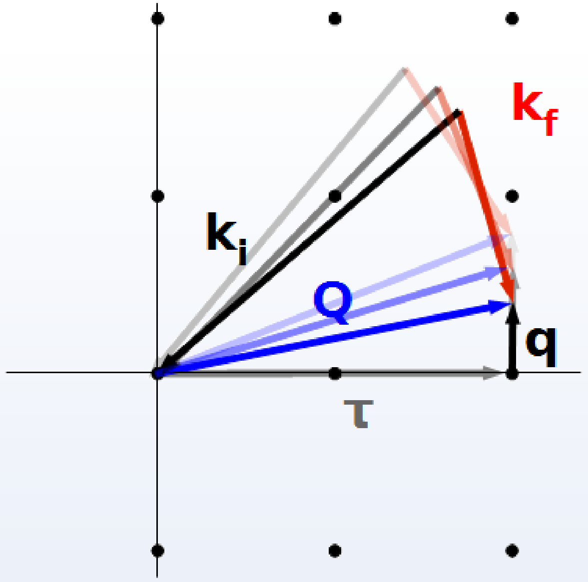
Intermezzo – Bragg-edge imaging and filters

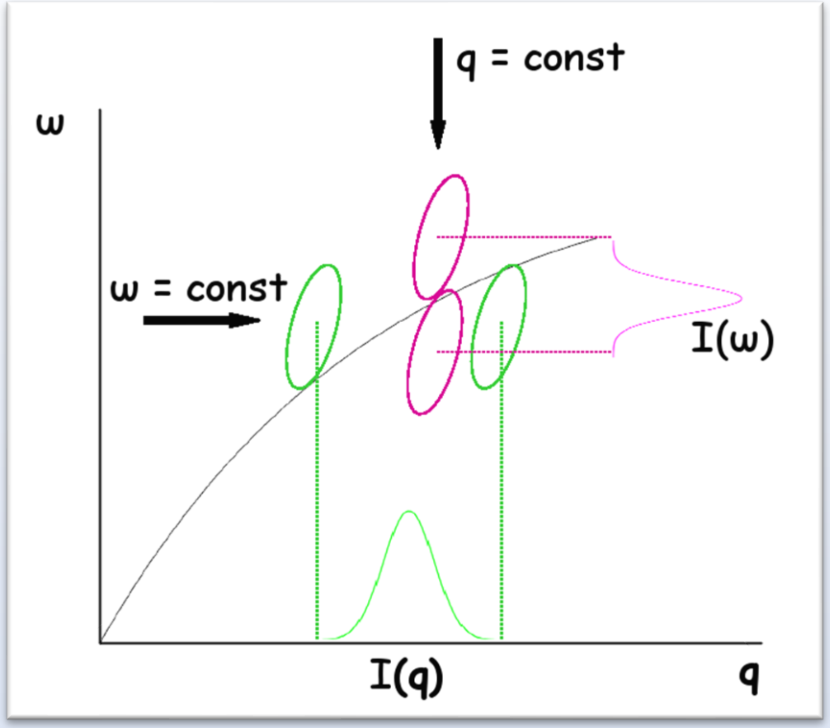




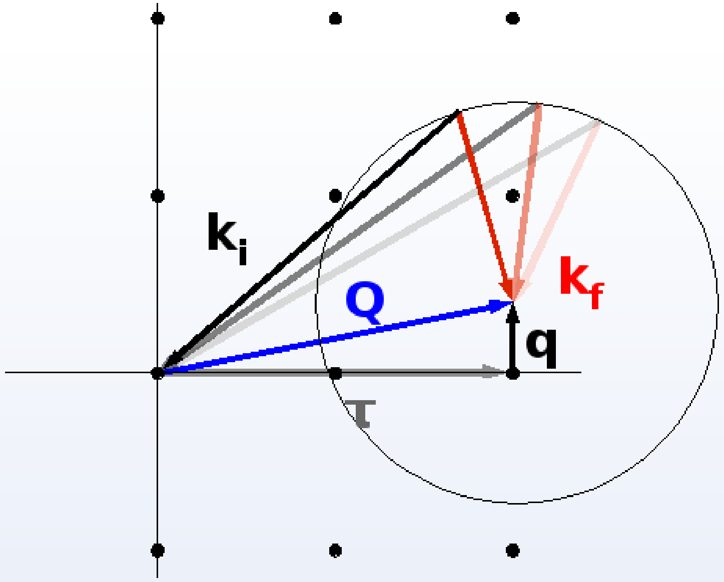
Triple Axis Spectroscopy - Modes of Operation

Constant-Momentum Scan
(q = constant)

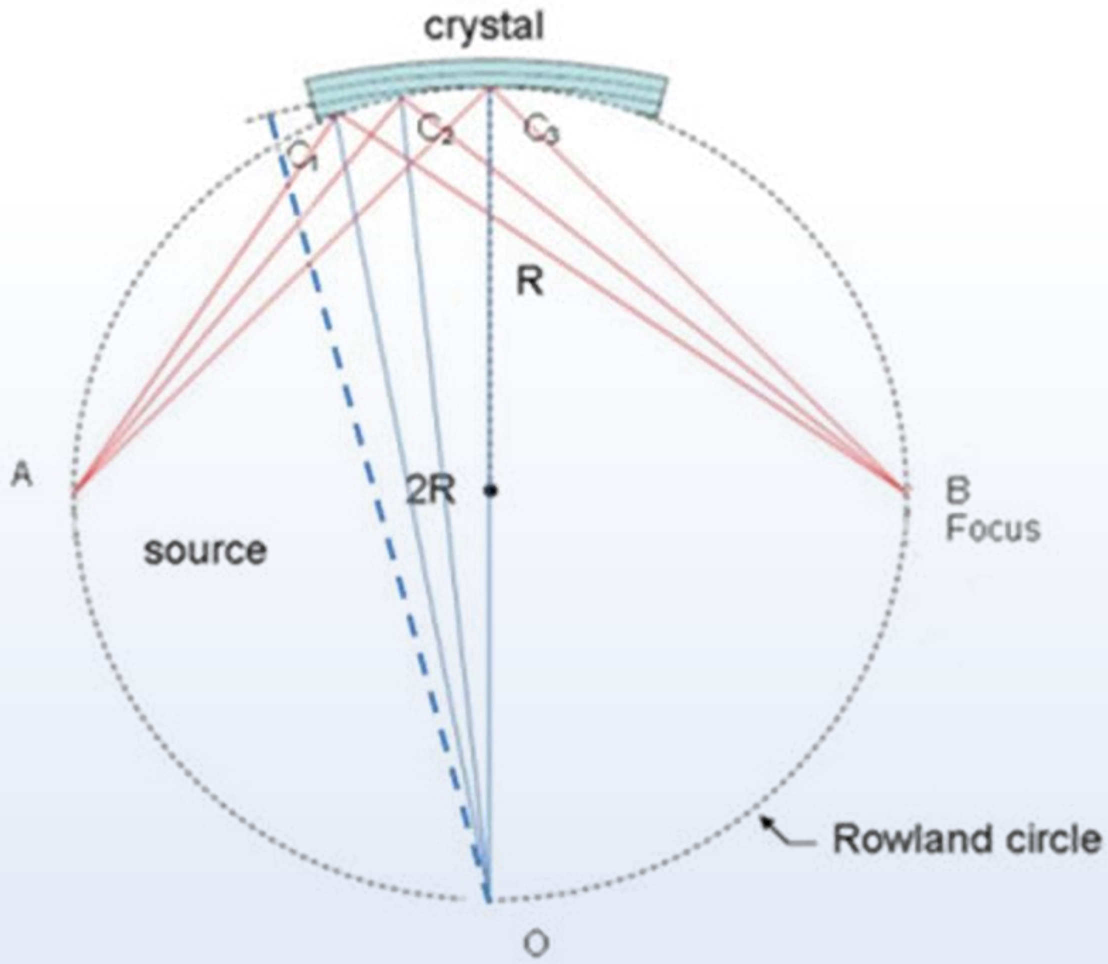


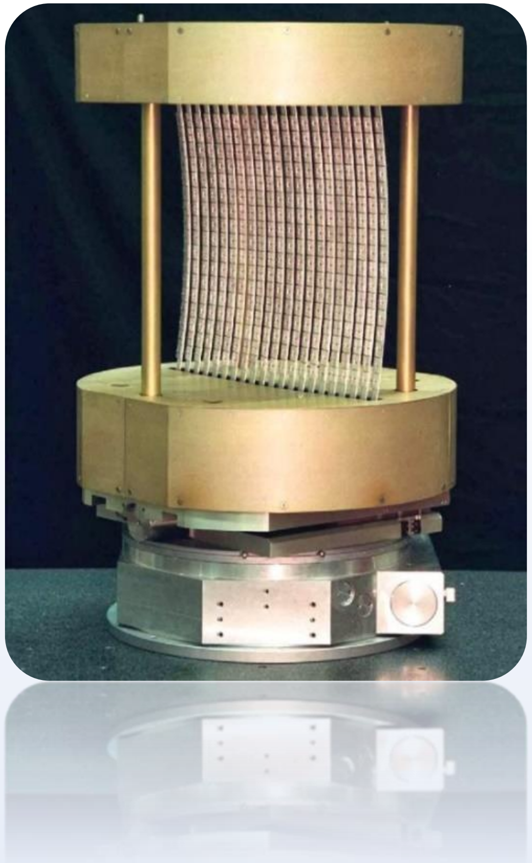


Constant-Energy Scan
(ω = constant)



Triple Axis Spectroscopy – Rowland Focusing





➔ Focusing increases the flux on the sample.

➔ What happens to the resolution?

Triple Axis Spectroscopy – Rowland Focusing

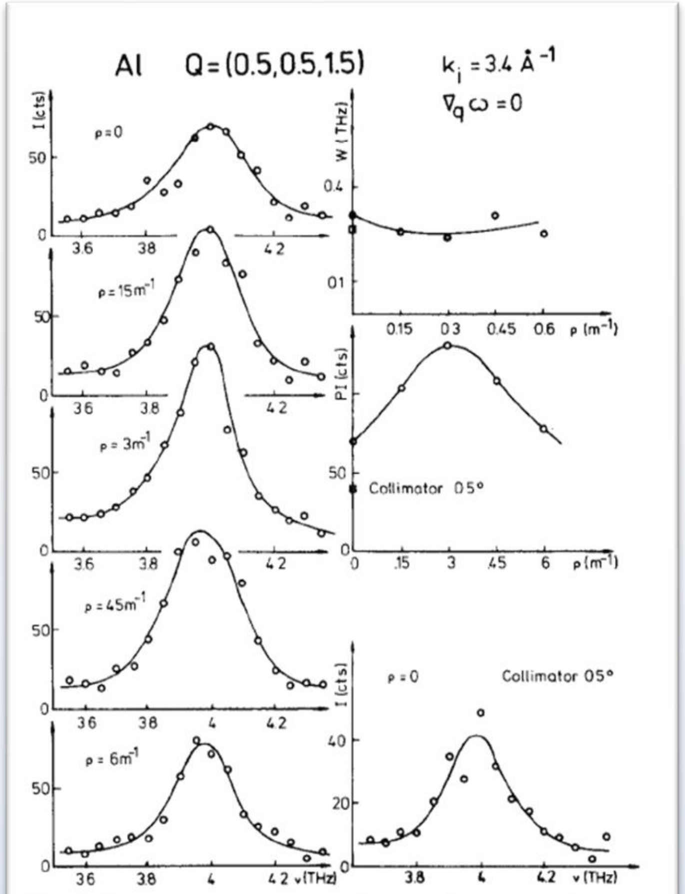


Fig. 8. Neutron groups corresponding to coherent one-phonon scattering involving the transverse mode at the L point in Al, for several values of the curvature ρ. Also shown are peak intensity PI, peak width W and a comparison scan with flat analyser and collimator (note different scale).

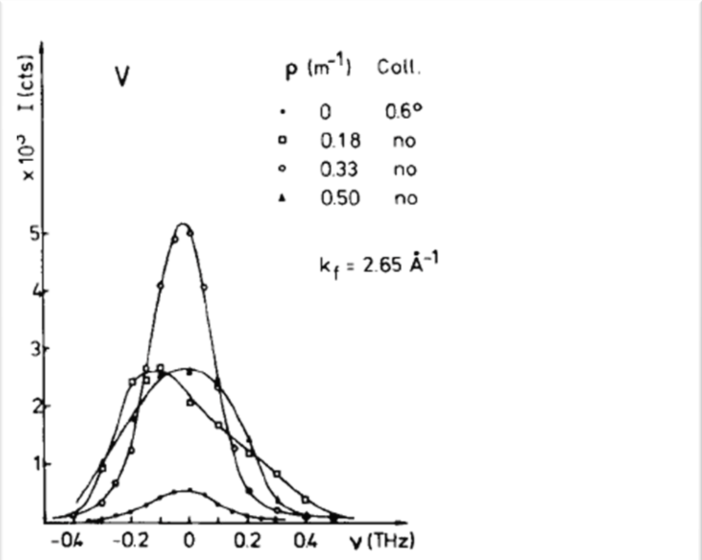
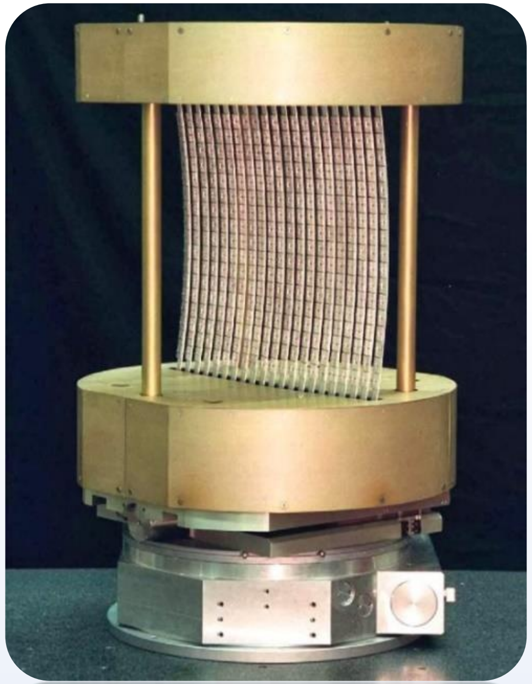


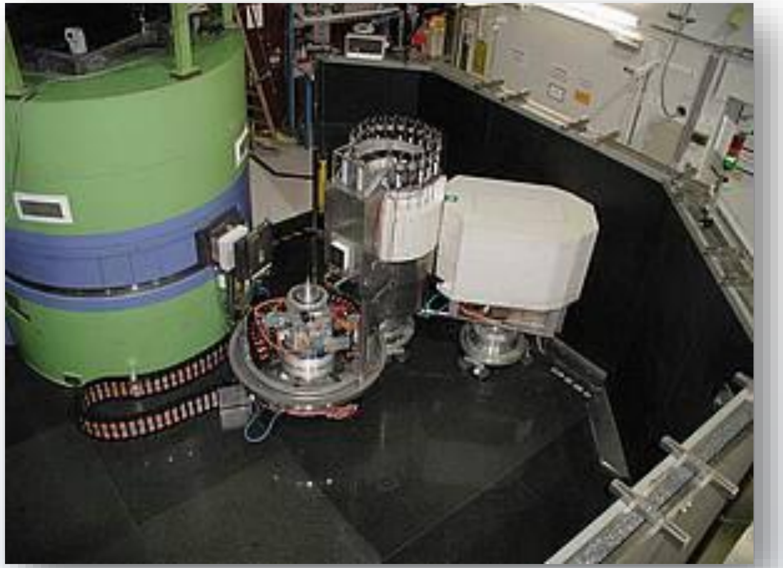
Fig. 7. Energy distributions of neutrons scattered elastically from a vanadium sample. Circles correspond to measurements with the curved analyser set at different curvatures and no collimator. A comparison scan with flat analyser and a 0.6° Soller collimator is shown with full dots.



➔ Momentum resolution decreases (high flux).

➔ However, energy resolution increases!!!

Recap: Advantages/Disadvantages ToF/3A



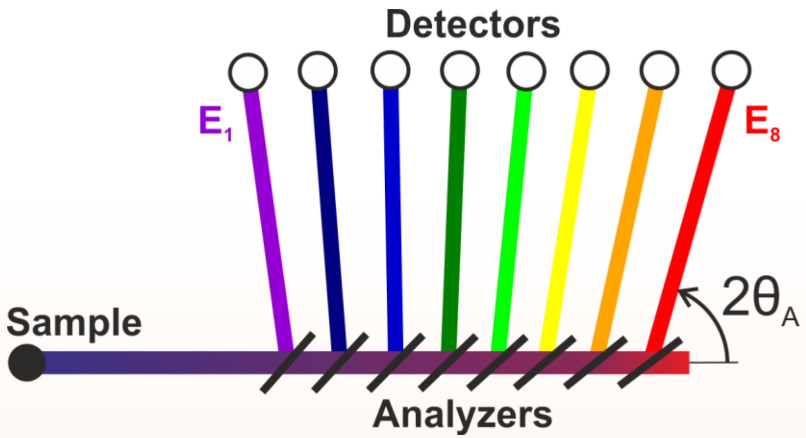
Time-of-Flight Spectrometer

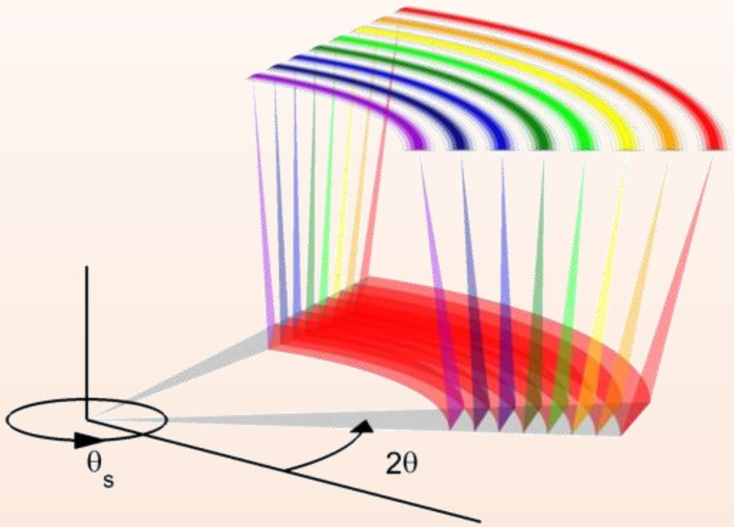
- Rapid data collection.
- Energy resolution mostly controlled via choice of incident energy (simple)
- Energy resolution mostly controlled via incident energy (limited options).
- Spurious Scattering is harder eliminate (multiple scattering,…).
- Sample environment such as magnets or pressure cells are difficult to use (background).

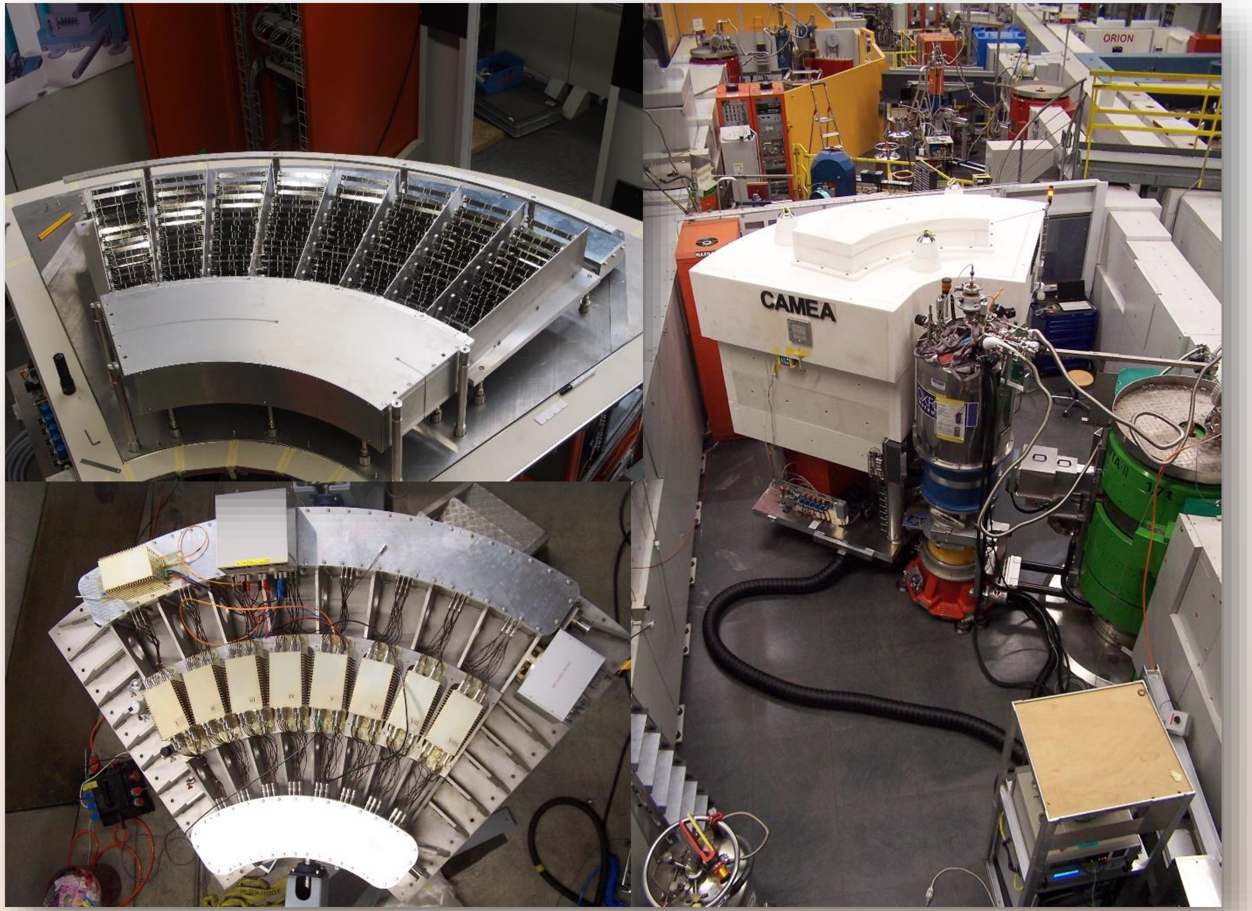
Triple-Axis Spectrometer

- Very versatile
- Clever choice of configurations can increase resolution while maintaining intensity.
- Background is more controlled.
- Can be used with large variety of sample environment.
- Slow data collection (single spot in Q-ω).
- Resolution function is complex (but can be calculated)

Extending TAS – CAMEA@SINQ



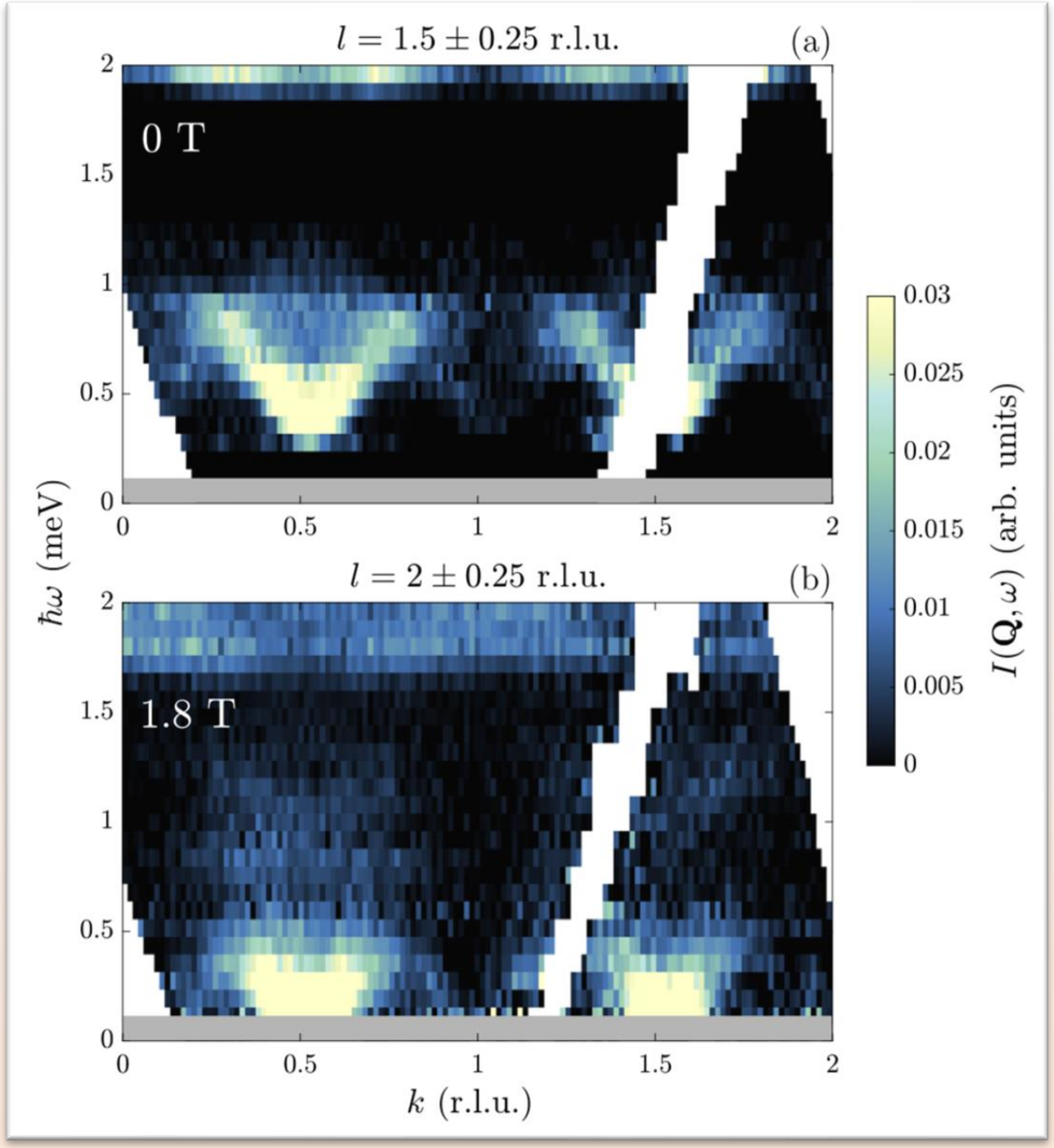


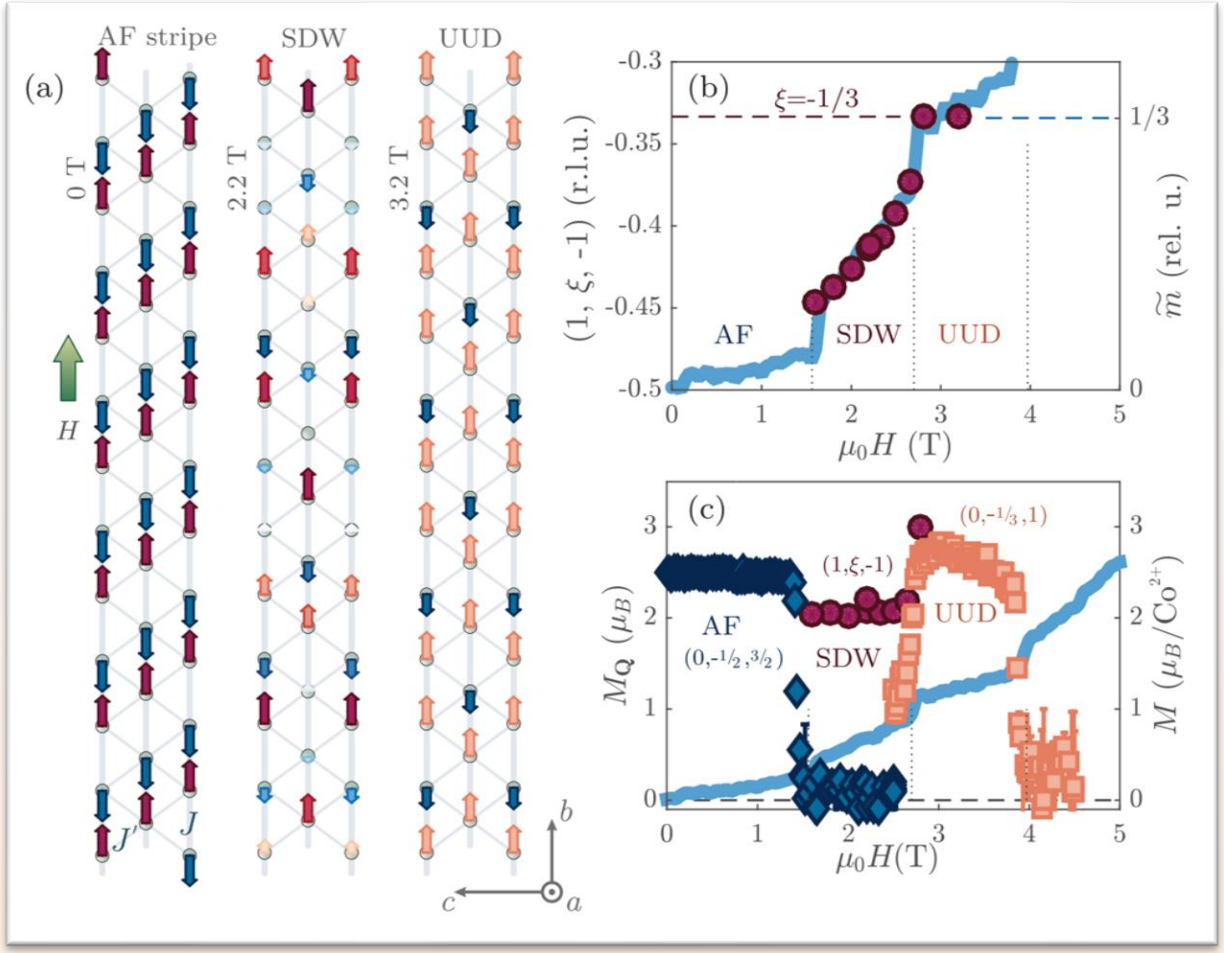


C. Niedermayer

P. Keller

SDW in frustrated magnet Cs2CoBr4





System: Cs_2CoBr_4
L. Facheris, et al., *Phys. Rev. Lett.* **129**, 087201 (2022)