Neutron Spectroscopy (a.k.a. Inelastic Neutron Scattering)

ORNL Neutron School 2026

Plan: Lecture 1 Formalism and theory (60 mins)
What is $S(\vec{Q},\omega)$ and why do we care?

Lecture 2 Experiments and spectrometers (45 mins)
How neutron spectrometers measure $S(\vec{Q},\omega)$?

Warning: This is a conceptual overview and a first version

Lecturer: Martin Mourigal, Georgia Tech

- BSc. Physics and Materials, France
- M.Sc. Condensed Matter Theory, EPFL (Switzerland)
- Ph.D. Condensed Matter Experiment, EPFL
  - All my time at ILL, France (TAS group)
- Postdoc Quantum Matter, Johns Hopkins
  - Majority of experiments at ORNL (ToF)
- Faculty at GT since 2015

Spend time at a national lab if you can! (DOE SCGSR)

1.1 Structure versus dynamics

- Diffraction $\langle \hat{O} \rangle$

  static order

  Example: expectation value of operators in thermal equilibrium $\langle \vec{\hat{r}}_i \rangle$ or $\langle \vec{\hat{S}}_i \rangle$

$\Rightarrow$ key outcome of experiment is a set of order parameters and instantaneous spatial correlations

- Spectroscopy $\hat{O}(t) = e^{i\hat{\mathcal{H}}t/\hbar} \hat{O}(0) e^{-i\hat{\mathcal{H}}t/\hbar}$ $\quad \omega \neq 0$

  Hamiltonian $\hat{\mathcal{H}}$ generates the time evolution

  dynamical evolution

  Example: overlap or correlation between value of operators at different times $\langle \hat{r}_i^{\alpha}(0)\, \hat{r}_i^{\alpha}(t) \rangle$ or space-times $\langle \hat{S}_i^{\alpha}(0)\, \hat{S}_j^{\beta}(t) \rangle$

$\Rightarrow$ key outcome of experiment is a set of dynamical correlation functions

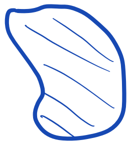
$\Rightarrow$ Both methods generalize to understanding two point correlation functions such as $\langle \hat{A}(0)\, \hat{B}(t) \rangle_{Q,T}$

operator/observables of interest

average over quantum and thermal ensembles

1.2 From the Hamiltonian to correlations and back

- Separation of energy scales in condensed matter



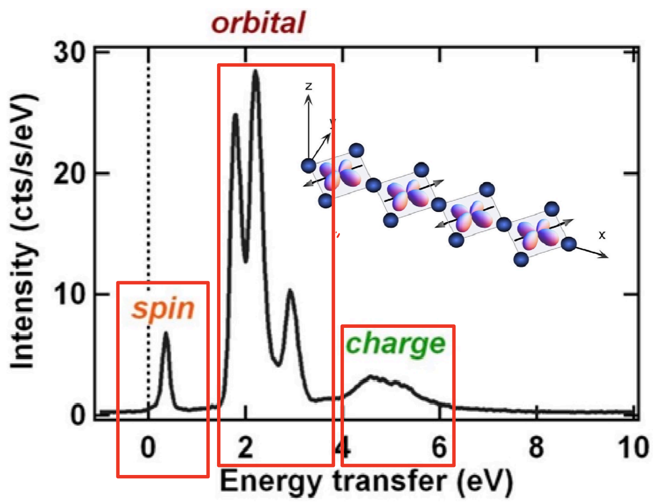
$$\hat{\mathcal{H}}_{mat} \simeq \hat{\mathcal{H}}_{orbital} + \hat{\mathcal{H}}_{charge} \overset{\text{Hubbard}}{\longleftrightarrow} + \hat{\mathcal{H}}_{spins} \overset{\text{magnetoelastic}}{\longleftrightarrow} + \hat{\mathcal{H}}_{lattice} + \dots$$

material

This is an idealization! Can recover some coupling!

Example: RIXS on 1D magnetic material Sr_2CuO_3

Nature 485, 82-85 (2012)



$\Rightarrow$ Each scale has its own isolated dynamics parametrized by an effective model

$$\hat{\mathcal{H}}_{orbital}(\{B_k^q\})\ ;\ \hat{\mathcal{H}}_{spins}(\{J_{ij}\})\ ;\ \hat{\mathcal{H}}_{lattice}(\{K_{ij}\})$$

crystal field parameters — exchange constants — Force constants

• So what's the job of a theorist?

$$\hat{\mathcal{H}}(\{p_i\}) \xrightarrow{\text{DIRECT PROBLEM}} \{|n\rangle, \mathcal{E}_n, i\Gamma_n\} \xrightarrow{\text{DIRECT PROBLEM}} \langle \hat{A}(0)\,\hat{B}(t)\rangle \xrightarrow{\text{DIRECT PROBLEM}} S^{AB}(\vec{Q},\omega)$$

wavefunction spectrum coherence

dynamical structure factor

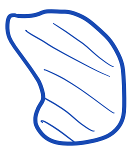
• So what's the job of a neutron spectroscopist?

$$\hat{\mathcal{H}}(\{p_i\}) \xleftarrow{\text{INVERSE PROBLEM}} \tilde{S}^{AB}(\vec{Q},\omega)$$

extract faithfull models (parameters + uncertainty)

perform accurate measurment

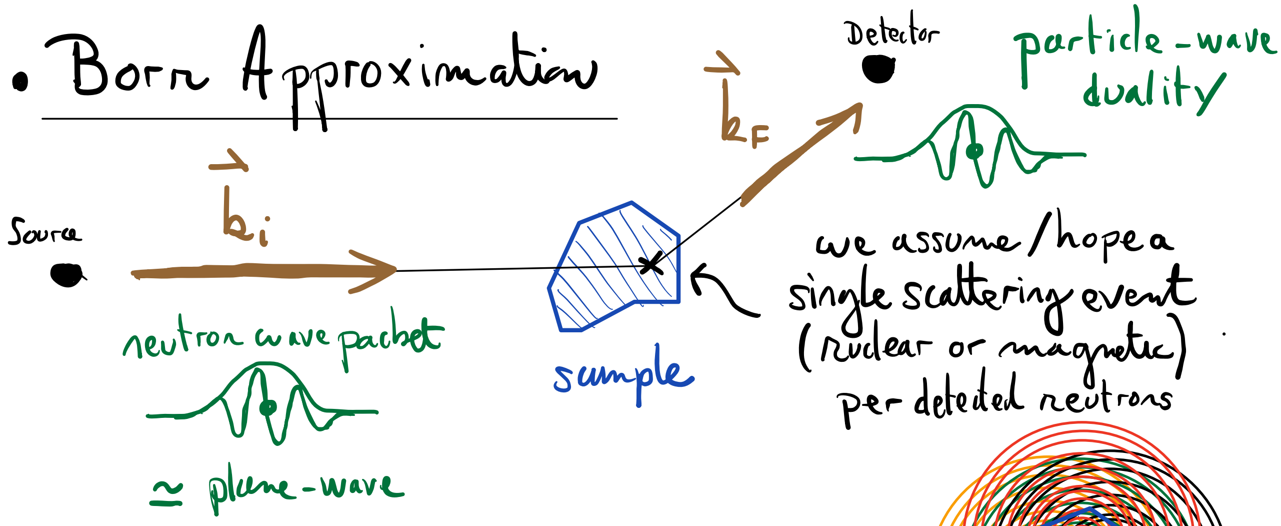
• Why? Make predictions from models!

 $+\ \hat{\mathcal{H}}(\{p_i\}) \Rightarrow$ Function

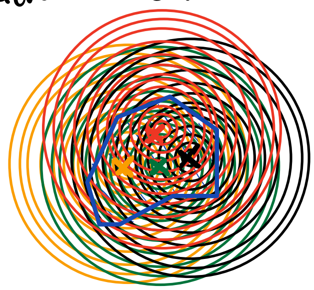
material model

1.3 Inelastic Scattering in a nutshell

- **Born Approximation**



it is an interference phenomenon



⚠ one neutron can experience <u>multiple scattering</u> events in sequence what complicates the analysis dramatically.

- **Neutron properties are ideal for condensed matter**

$$\left\{\begin{array}{l} \vec{p} = \hbar\vec{k} \\[2ex] E = \dfrac{\hbar^2 |\vec{k}|^2}{2m_n} \end{array}\right.$$

$$E[\text{meV}] = 2.072\, k^2 [\text{Å}^{-2}] = \frac{81.81}{\lambda^2} [\text{Å}^2]$$

$$5\ \text{meV} \rightarrow 1.55\ \text{Å}^{-1} \rightarrow 4\ \text{Å}$$

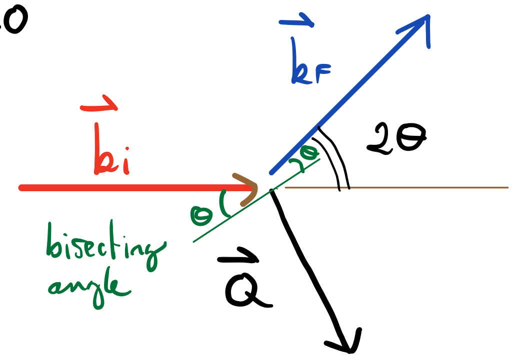
typical energy of CMP — match — typical lengthscale of CMP

$$1\ \text{meV} = 11.6\ \text{K} = 8\ \text{cm}^{-1} \qquad \hbar = 0.67\ \text{meV.ps}$$

if resolution $\Delta E = 1\ \text{meV}$ $\quad \tau_{res} = \hbar/\Delta E \approx 0.67\ \text{ps}$, slower dynamics is limited by resolution

• Scattering Triangle

$\hbar\omega = 0$



momentum transfer (to sample)

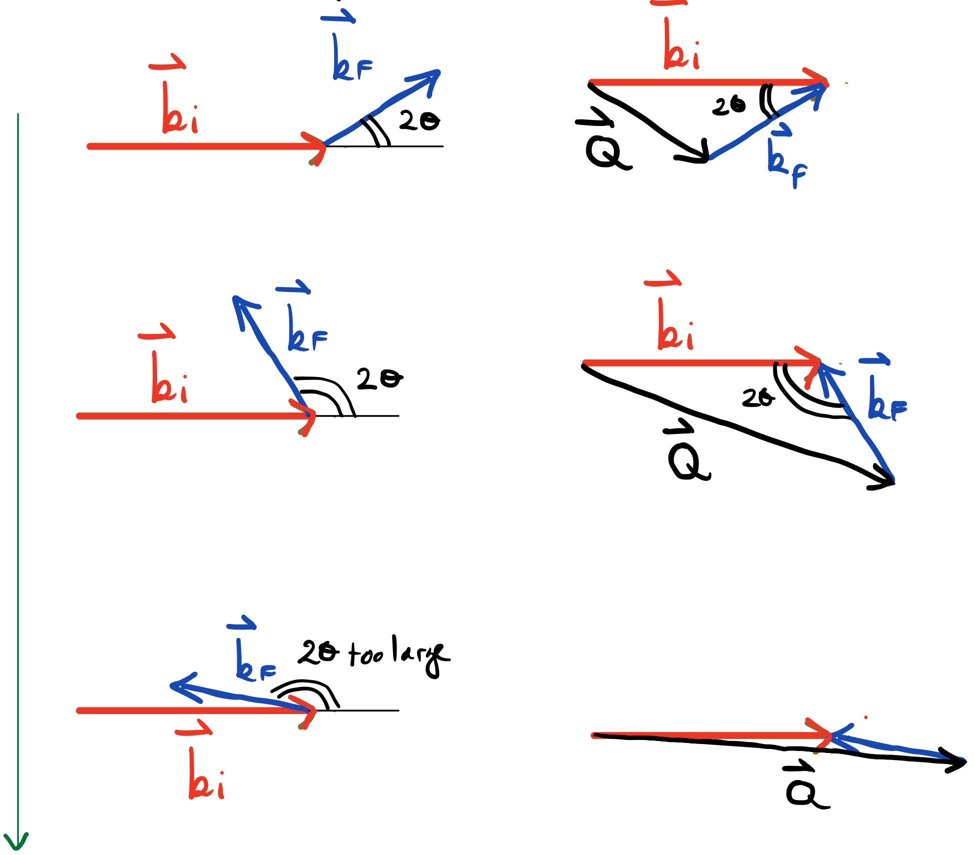
$$\vec{Q} = \vec{k}_i - \vec{k}_F$$

$$|\vec{Q}| = k_i^2 + k_F^2 - 2k_ik_F\cos 2\theta$$

energy transfer (to sample)

$$\hbar\omega = E_i - E_F = \frac{\hbar^2}{2m_n}\left(k_i^2 - k_F^2\right)$$

$\hbar\omega > 0$ ($k_F < k_i$)



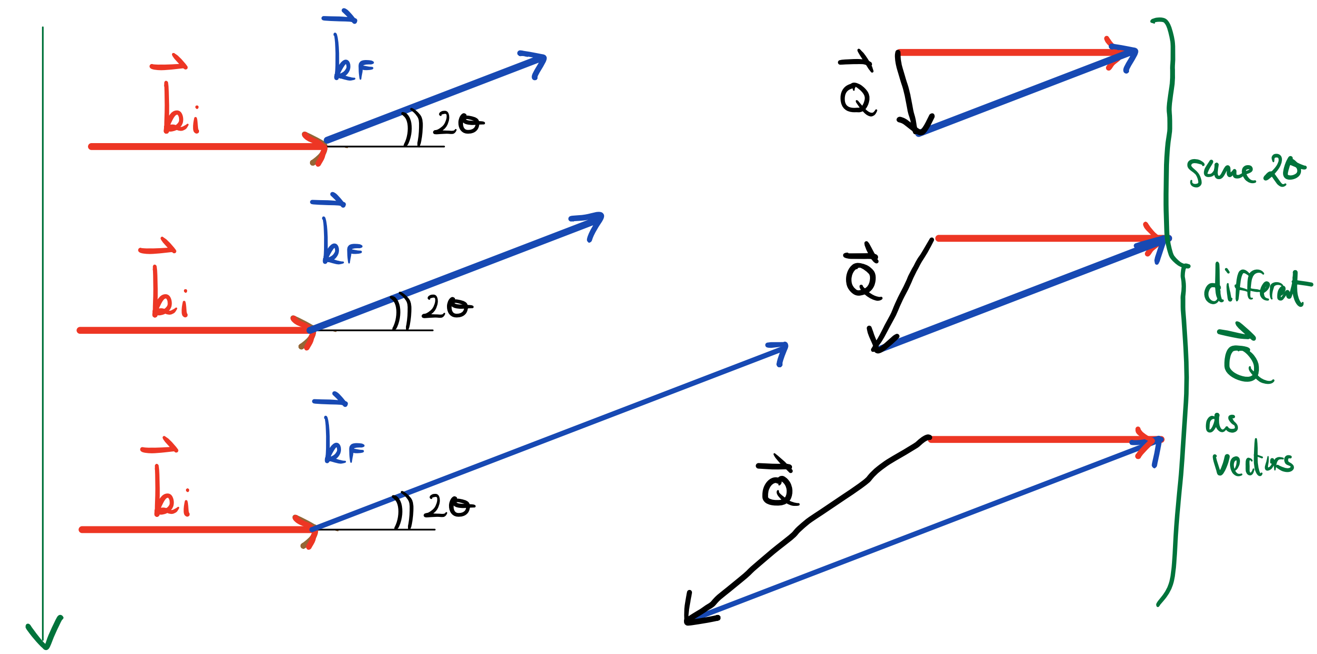
same $\hbar\omega$

different Q!

there is a maximum $|Q|$ one can reach!

This example: changing 2θ but keeping $\hbar\omega$ fixed

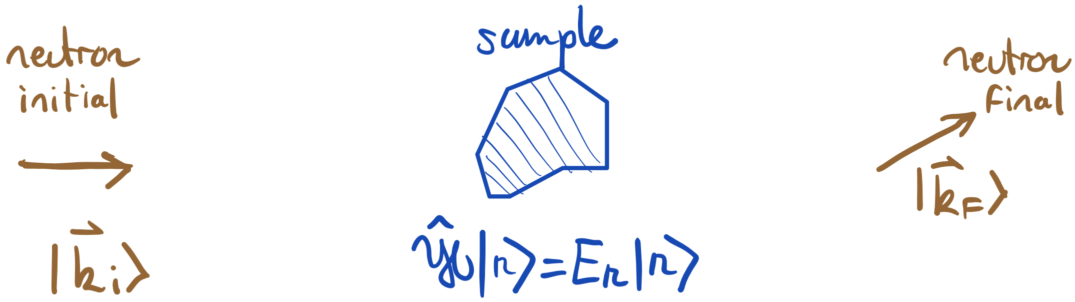
$\hbar\omega < 0 \quad (k_F > k_i)$



This example: keeping 2θ but changing $\hbar\omega$ through k_F

1.4 Quantum Treatment of Scattering

- set-up



Before

$$|i\rangle = |\vec{k}_i\rangle \otimes |n\rangle$$

After

$$|f\rangle = |\vec{k}_f\rangle \otimes |m\rangle$$

- transition matrix element: what does the neutron couple to?

Interaction between neutron and sample is essentially local:

$$\hat{V}_N(\vec{r}) \propto \sum_j b_j \, \delta(\vec{r} - \hat{\vec{R}}_j)$$

atomic positions

So we get:

$$\langle \vec{k}_f | V_N(\vec{r}) | \vec{k}_i \rangle \propto \int \sum_j b_j e^{-i\vec{k}_f\cdot\vec{r}} \delta(\vec{r}-\vec{R}_j)\, e^{+i\vec{k}_i\cdot\vec{r}}\, d^3\vec{r}$$

in position representation

$$\propto \sum_j b_j e^{i\underbrace{(\vec{k}_i-\vec{k}_F)}\cdot\vec{R}_j}$$

it's $\vec{Q}$!

plane waves select $\vec{Q}$

it's a spatial Fourier transform!

$$\propto \sum_j b_j e^{i\vec{Q}\cdot\vec{R}_j} \equiv \hat{A}_{\vec{Q}}$$

and in the end for the full system:

$$\langle i | V(\vec{r}) | f \rangle = \langle n | \otimes \langle k_i | V(\vec{r}) | k_F \rangle \otimes | m \rangle = \langle n | \hat{A}_{\vec{Q}} | m \rangle$$

$$\langle n | \hat{A}_{\vec{Q}} = \sum_j b_j e^{i\vec{Q}\cdot\vec{R}_j} | m \rangle$$

A measurement at $\vec{Q}$ probes the $\vec{Q}$-Fourier component of a sample operator.

- Fermi's Golden Rule: which transitions can occur?

$$W_{|i\rangle \to |f\rangle} = \frac{2\pi}{\hbar} \underbrace{|\langle f|\hat{V}|i\rangle|}_{\text{we just calculated this!}}{}^2 \delta(E_f^{tot} - E_i^{tot})$$

$$\left.\begin{aligned} E_f^{tot} &= E_m + E_f \\ E_i^{tot} &= E_n + E_i \end{aligned}\right\} E_f^{tot} - E_i^{tot} = -\underbrace{(E_i - E_f)}_{\uparrow \text{ it's } \hbar\omega \equiv E!} + (E_m - E_n)$$

so we get $\delta(E - [E_m - E_n])$ ↑ selects energy differences

and in the end:

$$W_{i \to f} = \frac{2\pi}{\hbar} |\langle m|\hat{A}_Q|n\rangle|^2 \delta(E - [E_m - E_n])$$

- Lehmann Representation and Finite Temperature

Fermi's Golden rule is about the transition between two states $|n\rangle$ and $|m\rangle$. But at finite temperature we need to consider a thermal ensemble where:

$$P_n = e^{-\beta E_n}/Z \quad \text{where } Z = \sum_n e^{-\beta E_n}$$

↖ $1/k_B T$

↖ probability for the system to be in $|n\rangle$

Furthermore we don't filter according to final state $|m\rangle$ so we need to sum over all of them: $\sum_m$

The tool to deal with this is the dynamical structure factor (DSF), here in Lehmann rep:

$$\boxed{\begin{gathered} S(\vec{Q},E) \equiv \frac{1}{Z} \sum_{n,m} e^{-\beta E_n} \left| \langle m | \hat{A}_Q | n \rangle \right|^2 \delta\left(E - [E_m - E_n]\right) \\ \text{contains all the neutron visible transitions at } (\vec{Q},E) \end{gathered}}$$

In general the neutron scattering cross-section takes the form $\dfrac{d^2\sigma}{d\Omega\, dE_F} \propto \dfrac{k_F}{k_i} S(\vec{Q},E)$

up to details of the interaction process.

1.5 What does the DSF encode

- What determines the visibility of an excitation?

(1) it exists! $E = E_m - E_n$

(2) matrix element allows it $\langle m | \hat{A}_{\vec{Q}} | n \rangle \neq 0$

(3) the initial state is populated! $p_n \neq 0$

(4) the spectrometer can reach $(\vec{Q}, E)$!

neutrons see it!

- Modes and continua at $T \to 0$ ($p_n = \delta_{n,0}$)

$$S(\vec{Q},E) = \sum_m |\langle m | \hat{A}_Q | 0 \rangle|^2 \delta(E - [E_m - E_0])$$

ground state

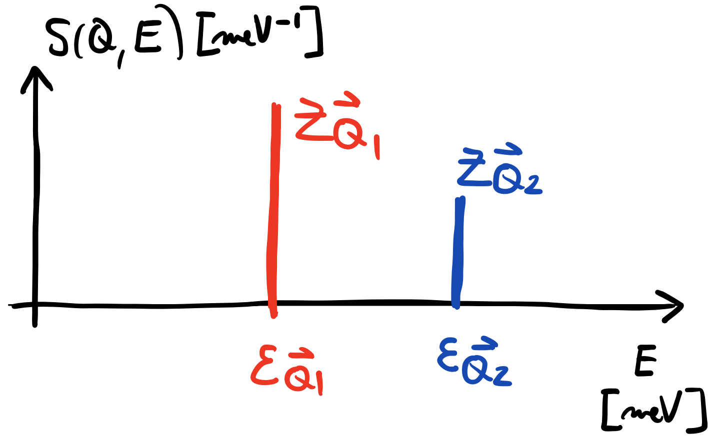
Coherent Mode $\hat{A}_{\vec{Q}} |0\rangle = \sqrt{Z_{\vec{Q}}}\, |\vec{Q}\rangle$

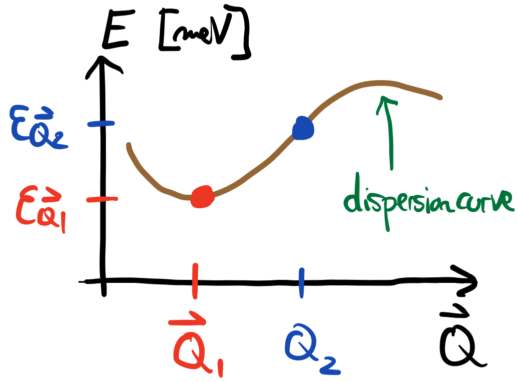
1-excitation with wavevector $\vec{Q}$ (phonon, magnon, ...)

then $S_{\text{mode}}(\vec{Q},E) = Z_{\vec{Q}}\, \delta(E - \varepsilon_{\vec{Q}})$

mode spectral weight

mode dispersion





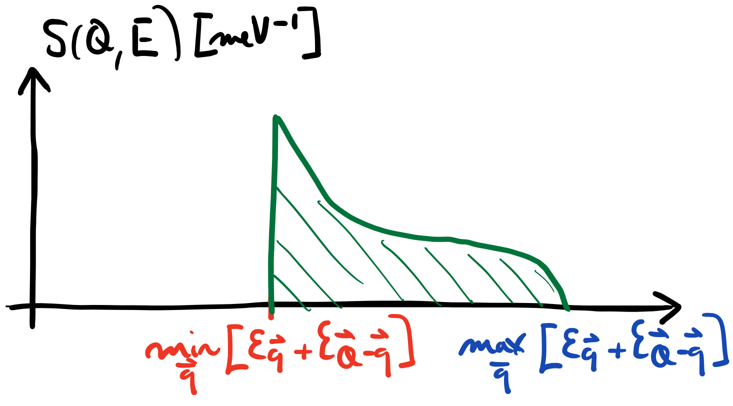
Excitation Continuum

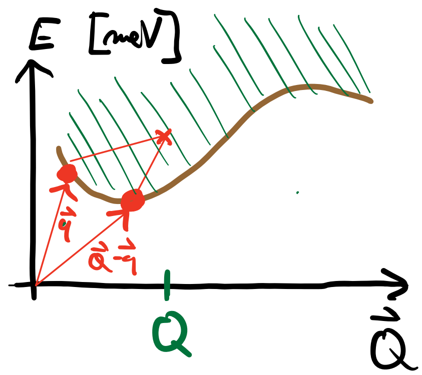
Multiple $|m\rangle$ states possible

then $$S_{cont}(\vec{Q},E) = \sum_m W_m(\vec{Q})\,\delta(E - [E_m - E_0])$$

For instance $E = \varepsilon_{\vec{q}} + \varepsilon_{\vec{Q}-\vec{q}}$

internal degree of freedom linked to all possible m's





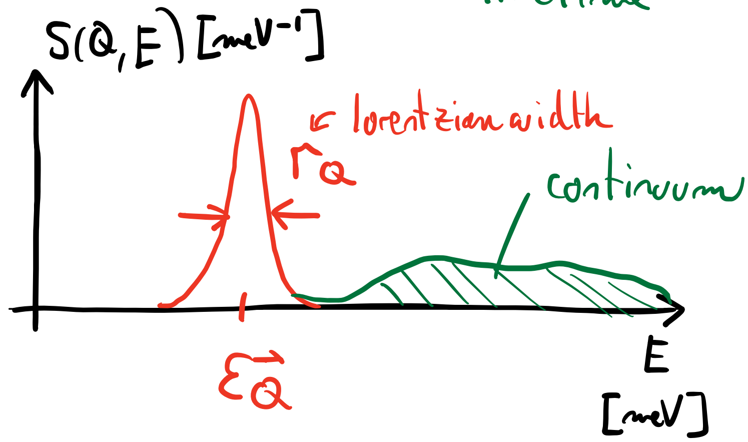
Many body system

$$S(\vec{Q},E) = Z_{\vec{Q}} \frac{1}{\pi} \frac{\Gamma_{\vec{Q}}}{(E-\varepsilon_{\vec{Q}})^2 + \Gamma_{\vec{Q}}^2} + S_{cont}(\vec{Q},E)$$

quasiparticle residue

quasiparticle lifetime

sideband



■ 1.6 Link to correlation Functions

- Back to space-time

We need to undo the $\vec{Q}$ and $\hbar\omega$ selections that we performed earlier

We use mathematical results:

(a) $$\delta(x) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt\, e^{-ixt/\hbar}$$

(b) $$\sum_m |m\rangle\langle m| = \hat{\mathbb{1}}$$

identity operator

Then $$S(Q,E) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt\, e^{-iEt/\hbar}\left\langle \hat{A}_{-\vec{Q}}(0)\,\hat{A}_{Q}(t)\right\rangle_T$$

(c) $$\hat{A}_{\vec{Q}} = \sum_j e^{i\vec{Q}\cdot\vec{R}_j}\,\hat{A}_j$$

then $$\left\langle \hat{A}_{-\vec{Q}}(0)\,\hat{A}_{Q}(t)\right\rangle = \sum_{ij} e^{i\vec{Q}(\vec{R}_j-\vec{R}_i)}\left\langle \hat{A}_i(0)\hat{A}_j(t)\right\rangle$$

For a system with translation symmetry

$$\vec{r} = \vec{R}_j - \vec{R}_i$$

$$S(\vec{Q},E) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt\, e^{-iEt/\hbar} \int e^{i\vec{Q}\cdot\vec{r}}\, d^3\vec{r}\, \underbrace{\langle \hat{A}(0,0)\,\hat{A}(\vec{r},t)\rangle}_{G(\vec{r},t)}$$

$\vec{r} \leftrightarrow \vec{Q}$ $\quad t \leftrightarrow E/\hbar$

so the DSF is the space-time Fourier Transform of two-point correlation functions

- Detailed balance

$$S(-\vec{Q},-E) = e^{-\beta E}\, S(\vec{Q},E)$$

goes to zero for $E<0$ when $T \to 0$

- Zero-energy spectral density

$$G(\vec{Q},t) = \underbrace{\underset{\text{Fluctuations}}{G(\vec{Q},t)}}_{\text{dynamics} \to 0 \text{ for } t\to+\infty} + \underbrace{G(\vec{Q},t\to+\infty)}_{\text{long-time asymptotic behavior}}$$

$$S(\vec{Q}, E=0) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt \; G(\vec{Q}, t)$$

$$S(\vec{Q}, E=0) = S(\vec{Q}, E=0) + \underbrace{G_\infty(\vec{Q})\,\delta(E)}_{\text{elastic scattering}}$$

- instantaneous correlations

$$\int_{-\infty}^{+\infty} S(\vec{Q}, E)\,dE = \frac{1}{2\pi\hbar}\int e^{i\vec{Q}\cdot\vec{r}} d^3\vec{r} \int_{-\infty}^{+\infty} dt \underbrace{\int_{-\infty}^{+\infty} dE\, e}_{\delta(t)} \; G(\vec{r}, t)$$

$$S(\vec{Q}) = \int e^{i\vec{Q}\vec{r}} d^3\vec{r} \;\; \vec{G}(\vec{r}, t=0)$$

$S(\vec{Q})$ = Instantaneous or Equal-time Structure Factor

"Snap shots"

- Generalization to different operators

$$S(\vec{Q}, \omega) \to S^{AB}(Q, \omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt\, e^{-iEt/\hbar} \langle \hat{A}_{-Q}(0)\; \hat{B}_{+Q}(t) \rangle$$

In neutron scattering:

$\hat{A}_Q = \hat{\rho}_{\vec{Q}}$ density (${}^4He$, liquids, etc)

$\hat{A}_Q = \hat{U}_{\vec{Q}}$ lattice displacement (crystals)

$\hat{A}_{\vec{Q}} = F_{\vec{Q}}\, \hat{M}^{\alpha}_{\perp Q}$ magnetic scattering (magnets)

For other experimental techniques:

$\hat{A}_{\vec{Q}} = \vec{J}_{\vec{Q}}$ current density (transport)

$\hat{A}_{\vec{Q}} = \hat{T}^{\alpha\beta}_{\vec{Q}}$ multipolar operator (RIXS)

Connection to experiments involve pre factors, for magnetism:

$$\frac{d^2\sigma}{d\Omega dE_F} = \frac{k_F}{k_i}\left(\frac{\gamma r_0}{2}\right)^2 \underbrace{|g\, F(\vec{Q})|^2}_{\text{magnetic ion}} \underbrace{P_{\alpha\beta}(\vec{Q})}_{\text{geometry}} \underbrace{S^{\alpha\beta}(\vec{Q},E)}_{\text{correlations } S^\alpha S^\beta}$$

■ 2.1 From materials dynamics to detector counts

• Recall the direct problem:

some properties of the material (shape, mosaic)

$$\underbrace{\hat{\mathcal{H}}_{\text{material}}(\{p_i\}) \to S(\vec{Q},E) \to \frac{d^2\sigma}{d\Omega dE_F}}_{\text{independent of the instrument}} \longrightarrow R \otimes \frac{d^2\sigma}{d\Omega dE_F} \to \underbrace{\text{Counts in detector vs time}}_{\text{"intensity"}}$$

properties of instrument

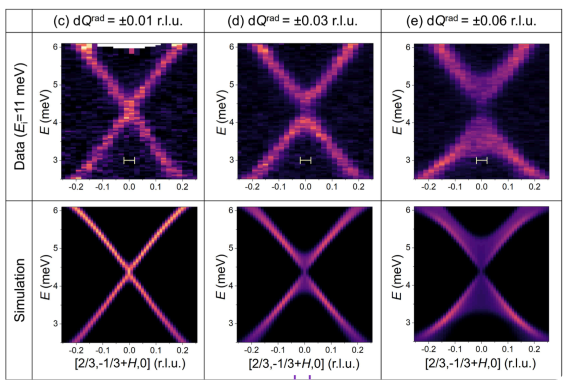
• We can represent this problem as:

$$\underbrace{\tilde{I}(\vec{Q}_0,E_0)}_{\substack{\text{reduced}\\ \text{scattering intensity}\\ \text{assigned to }(\vec{Q}_0,E_0)}} = \overbrace{A}^{\text{flux, sample, etc}} \iiiint d^3\vec{Q}'\,dE'\,\underbrace{R(\vec{Q}'-\vec{Q}_0,E'-E_0)}_{\substack{\text{resolution function}\\ \text{reflecting finite acceptance}\\ \text{in }\vec{Q}\text{ and }E\text{ around}\\ \text{nominal measurement point}}} \frac{d^2\sigma}{d\Omega dE_F} + \underbrace{B(\vec{Q}_0,E_0)}_{\substack{\text{instrument and}\\ \text{sample background}\\ \text{plus other unknowns}\\ \text{"spurions"}}}$$

Example: topological band crossing

gap or no gap?

Do et al. PRB 2022



■ 2.2 General Constraints on Neutron Spectrometers

- Recall the scattering triangle.

Our goal is to measure $\tilde{I}(\vec{Q}_0,\vec{E}_0)$ over a sufficiently broad range of $\vec{Q}_0$ and E_0, and with sufficient resolution

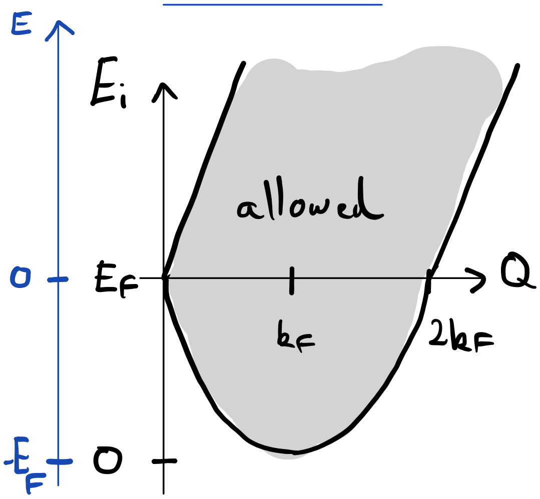
these two constraints strongly compete

...... to solve our scientific problem!

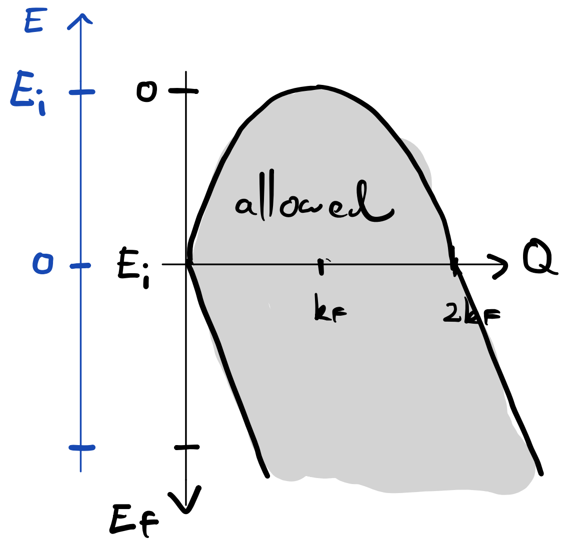
Fundamental constraint is: $|k_i - k_F| \leqslant Q \leqslant k_i + k_F$

You can imagine two simple choices to design and optimize a spectrometer, even before details come into play

Fixed k_F (or E_F)



Fixed k_i (or E_i)



- Flavors of Spectrometers

"INDIRECT-LIKE"

triple-axis : know k_i and k_F → fix k_i or k_F

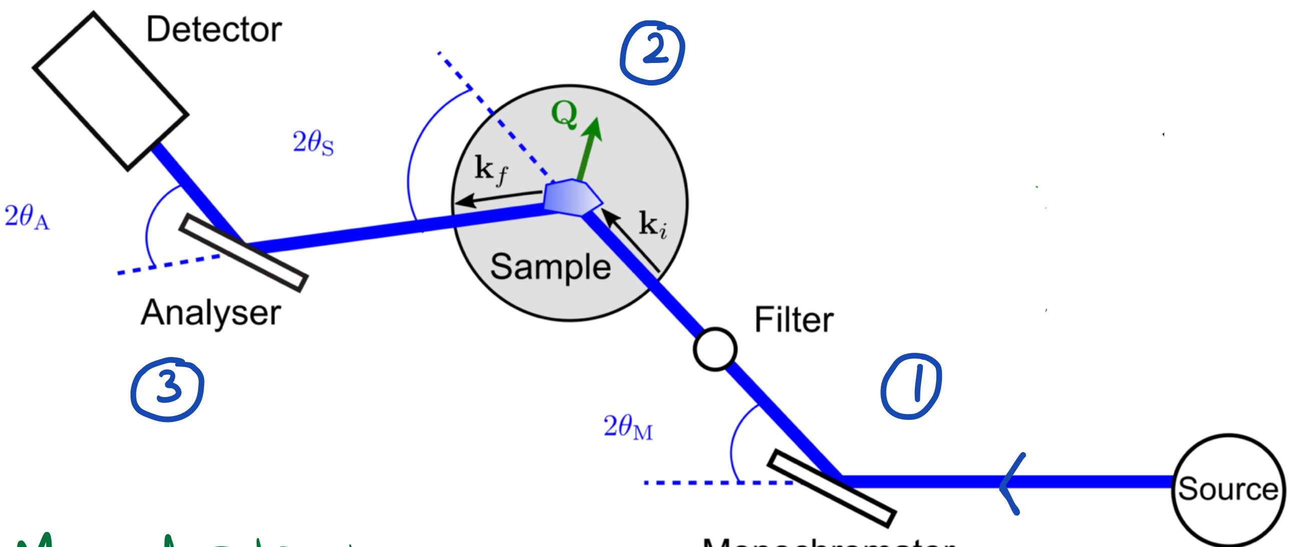
time-of-flight: know k_i or k_F → infer k_F / k_i from t.o.f

DIRECT INDIRECT

2.3 The triple-axis spectrometer

- Idea: Use Bragg reflections to select $\vec{k}_i$ and $\vec{k}_F$
  Use scattering angle 2θ to select $(\vec{Q}_0, E_0)$
  Usually operate with fixed $\vec{k}_F$ ← practical and historical reasons

- Schematic:



Mourigal, PhD Thesis, 2011

Bragg's Law from crystals is all you need:

Monochromator $2d_M \sin\theta_M = \lambda_i \rightarrow k_i = \frac{2\pi}{\lambda_i}$

Sample Scattering angle $2\theta_S \rightarrow$ angle between $\vec{k}_i$ and $\vec{k}_F$

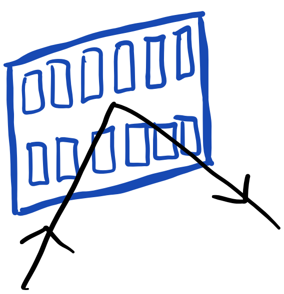
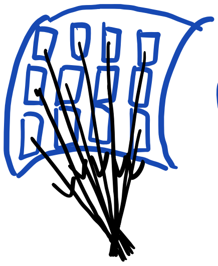
Analyzer $2d_A \sin\theta_A = \lambda_F \rightarrow k_F = \frac{2\pi}{\lambda_F}$

$$\text{TAS}: \overbrace{(\theta_M, 2\theta_S, \theta_A) + \text{sample orientation}}^{\text{events}} \Rightarrow (\vec{Q}_0, E_0)$$

few

[Canadian Invention
Chalk River, Ontario]

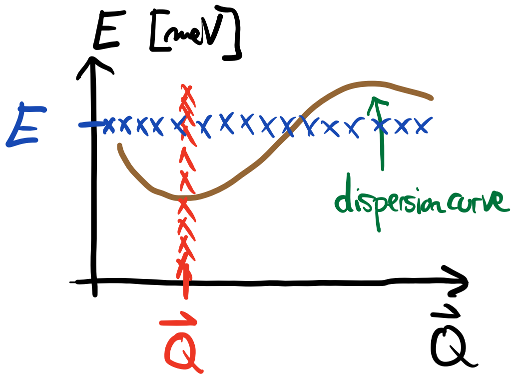
- **Monochromator / Analyzer materials**

Flat   Focusing

Graphite(002)
Silicon
Heusler

trade-off between reflectivity, wavelength etc

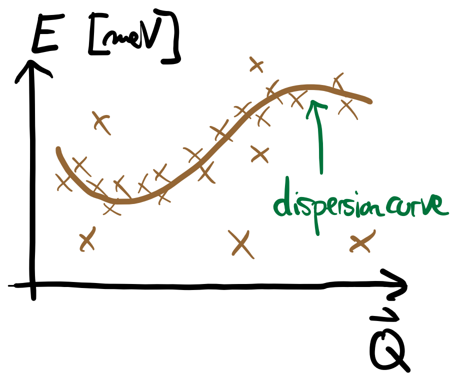
• Operations of a TAS spectrometer



constant-E cut

constant-$\vec{Q}$ cut

$$\underbrace{\begin{matrix}[\theta_M, 2\theta_s, \theta_m]_{i=1} \\ \vdots \\ [\theta_M, 2\theta_s, \theta_m]_{i=N}\end{matrix}}_{\text{coordinated}}$$



Bayesian search in $(\vec{Q}, E)$-space

• Filters

A major issue with crystal monochromators is <u>higher-order contamination</u>

Bragg's Law: $2d\sin\theta_M = n\lambda$ so at the same monochromator angle, the crystal may reflect

$\lambda, \lambda/2, \lambda/3, \ldots$ so $k, 2k, 3k, \ldots$

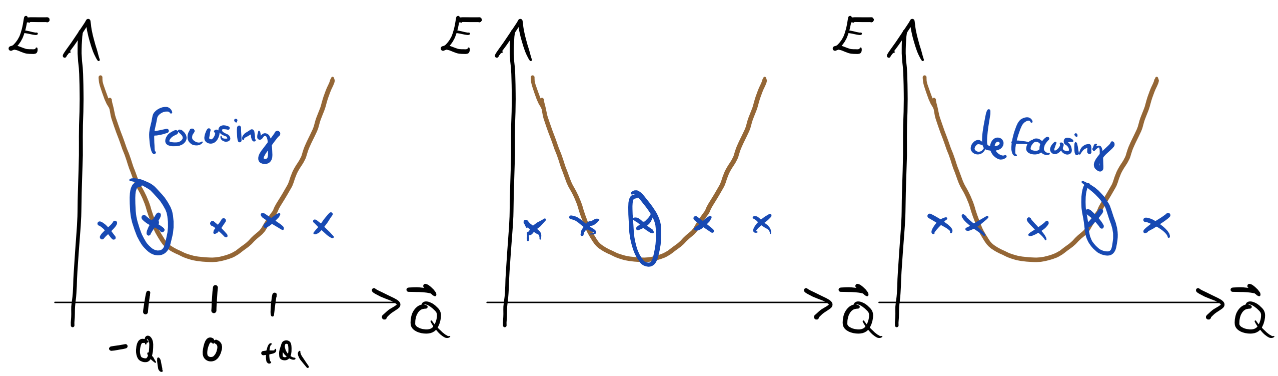
Filters such as PG or Cool Be or BeO work by transmitting k_i but not $2k_i$ or as a pass band filter

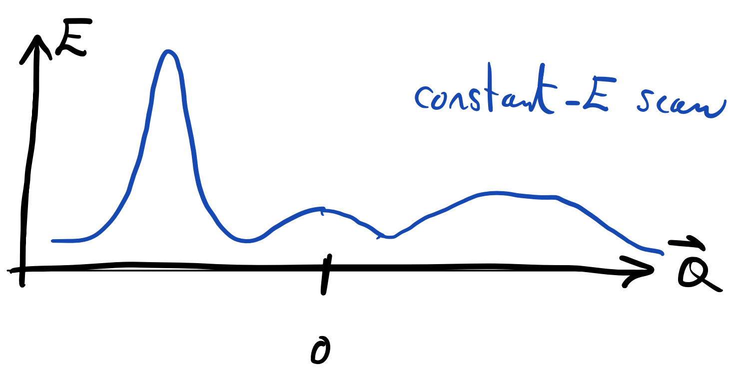
- **Resolution Function**

$$\tilde{I}(\vec{Q}_0, E_0) = A \iiiint d^3Q\, dE\, \underbrace{R(\vec{Q}-\vec{Q}_0, E-E_0)}\, \frac{d^2\sigma}{d\Omega\, dE_F} + B$$

Finite acceptance from beam divergence, mosaic and curvature of monochromator, sample size and mosaic, etc. etc.

Main feature is that $\Delta\vec{Q}$ and ΔE are correlated or in other words $\vec{R} \neq R_{\vec{Q}} \otimes R_E$.



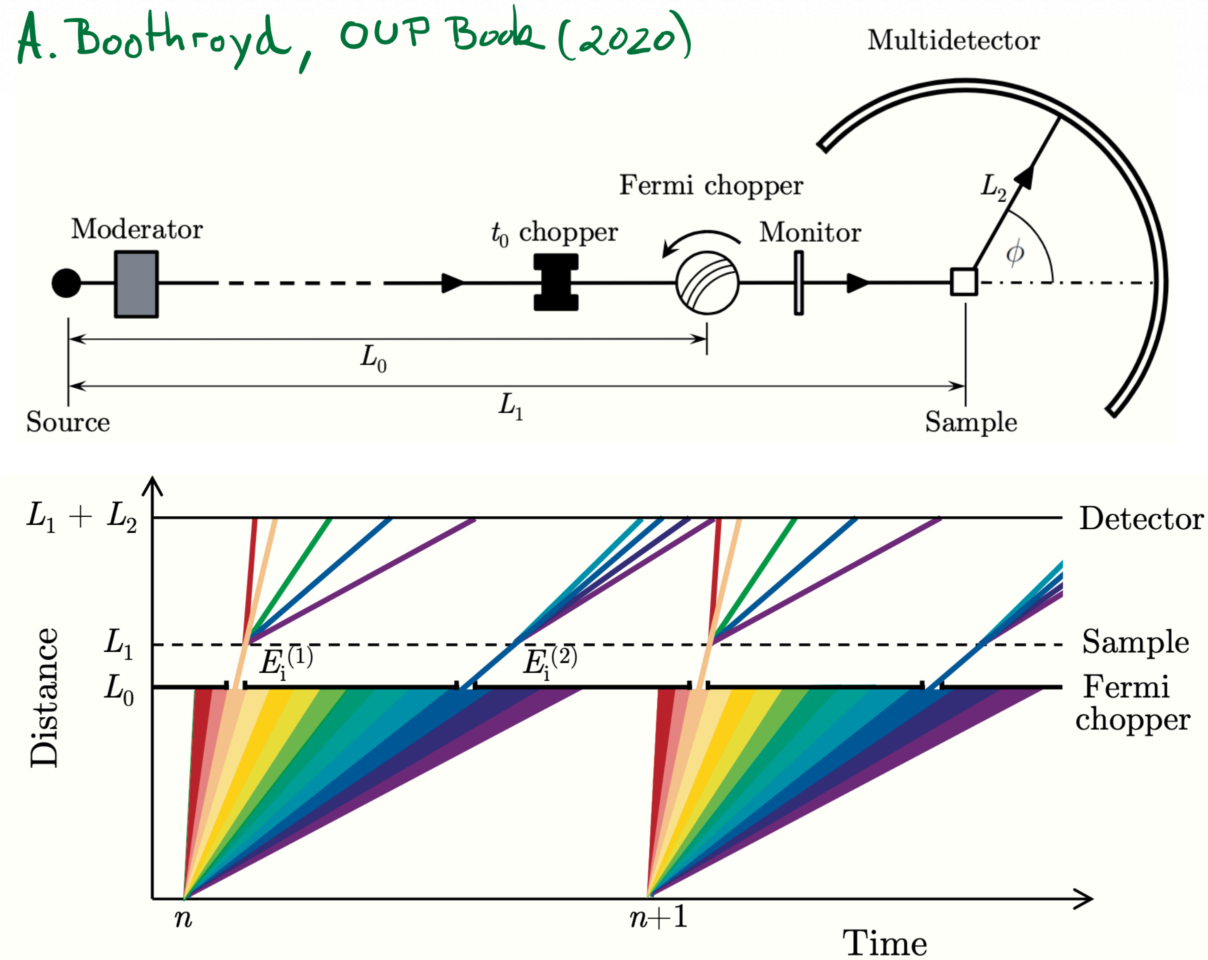


2.4 The direct-geometry time-of-flight spectrometer

- **Idea**: Use choppers to select k_i then time-of-flight and detector position to determine $\vec{k}_F$

- **Schematic**:

A. Boothroyd, OUP Book (2020)



Choppers: prepare E_i ($\vec{k}_i$) by filtering out

Flight time: determines E_F ($\vec{k}_F$)

many arrival times $\Rightarrow$ many E_F

Multiplexed detector: many detector pixels $\Rightarrow$ many $\vec{k}_F$ directions

$$t_{det} = t_i + t_F = \frac{L_1}{v_i} + \frac{L_2}{v_F} \quad \text{so } v_F = \frac{L_2}{t_{det} - L_1/v_i}$$

$$\text{DGS } \overbrace{\left(E_i,\ t_{det},\ \text{detector position}\right) + \text{sample orientation}}^{\text{events}} \Rightarrow \left(\vec{Q}_0,\ E_0\right)$$

many!

[??? Invention]

- ## Choppers and Resolution

In practice, many choppers used to control background, resolution, assign neutrons to the correct source pulse, etc

2.5 Conclusion: Solving the Inverse Problem

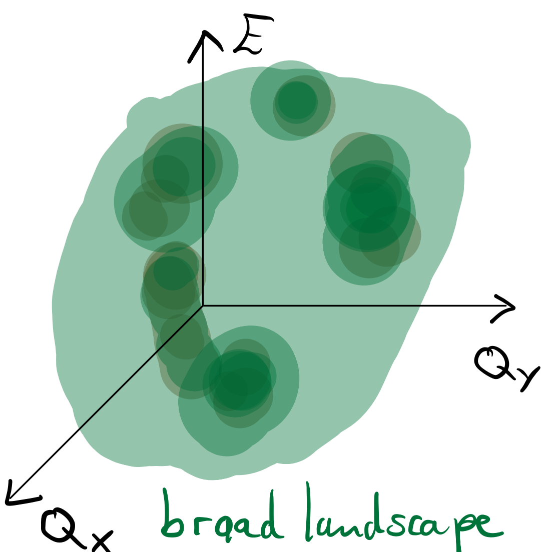
DIRECT PROBLEM

$$\hat{\mathcal{H}}(\{p_i\}) \to S^{AB}(\vec{Q},E) \to \frac{d^2\sigma}{d\Omega\, dE_F} \to \tilde{I}(\vec{Q},E) \to \text{Detector Counts}$$

INVERSE PROBLEM

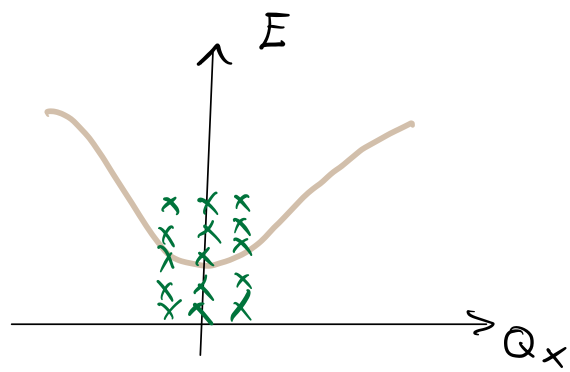
Goal: determine the most faithful $\hat{\mathcal{H}}(\{p_i\})$ and its uncertainty given background, resolution, kinematics, etc.

TOF: survey



broad landscape of $S^{AB}(\vec{Q},E)$

TAS: focus



Focus study of $(\vec{Q},E)$ points near signal of interest

Neutron Spectroscopy turns detector counts into physical models