Practical Experiment

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Presentation Outline:

From ideas.....

- ❑ **write a good proposal**
- ❑ **make a successful experiment**
- ❑ **analyse your data**

.....to papers (or thesis)

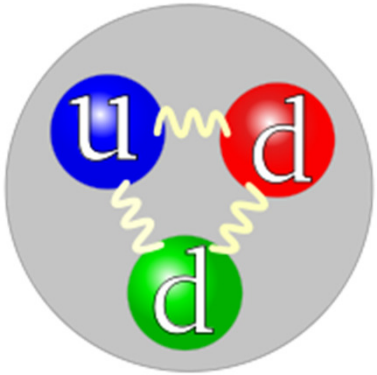
Step 1: Idea

You (or your supervisor) have an idea

Step 2: Proposal

You need neutrons (or you believe you do)

You have to write a proposal:



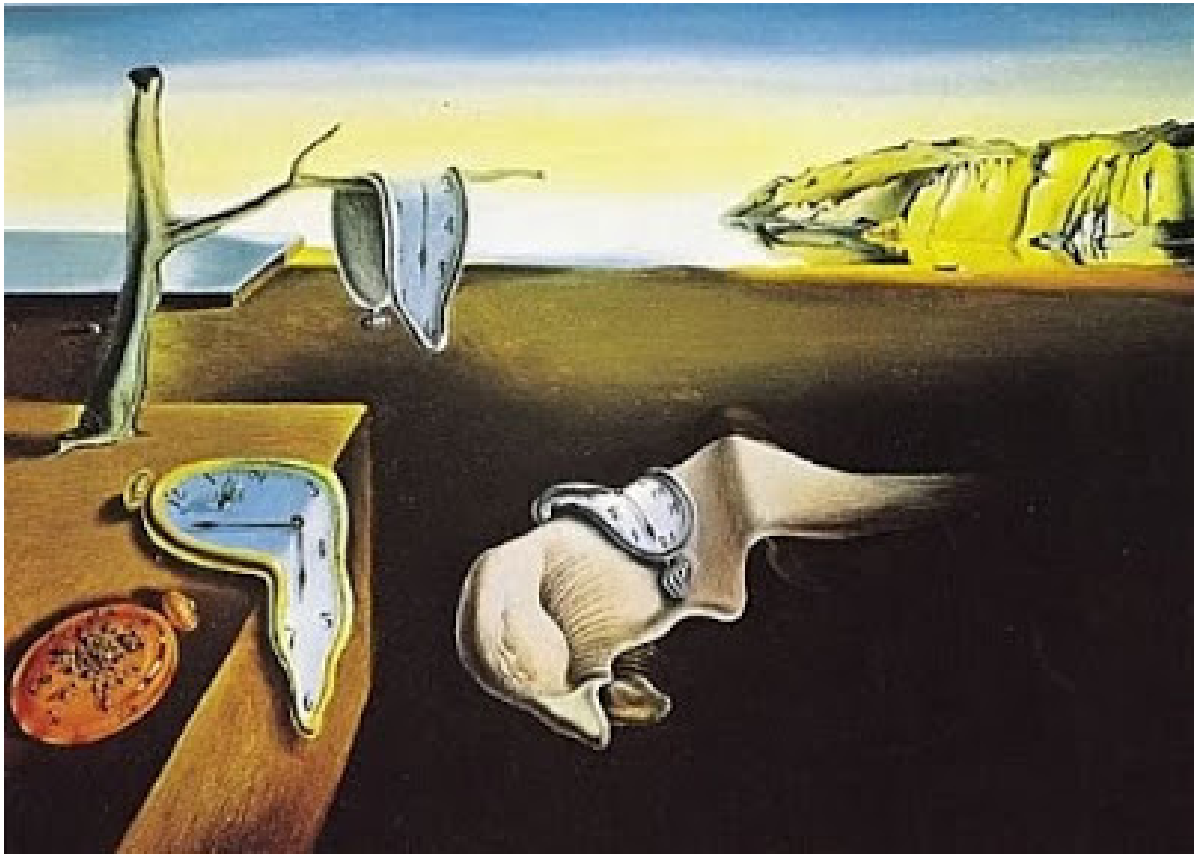
- ❑ **Characterize your sample**
- ❑ **Read instrument website**
- ❑ **talk with instrument scientist**
- ❑ **Make clear it is part of a PhD's thesis**
- ❑ **Etc….**

Step 3: Proposals

Your proposal is accepted!!!!

Step 4: Experiment

No matter how long you wait, too soon it is time for the experiment …..



Step 5: sample preparation

Get your sample ready:

- **For standard samples, it means make sure you have right amount, that they are oriented if single crystal, that appropriate sample holder are available, that they can be kept at right temperature humidity etc….**

- **For cultural heritage it is another story….**
  you need an incredible amount of paperwork and time!

Step 6: get the objects

What you need to do in order to be able to bring your precious artefacts from museums to facilities:

- ❑ **Convince a curator that you absolutely need an object from his museum (before proposal possibly!!!!!)**
- ❑ **Then you and the curator have to convince the museum director that your experiment is worth the trouble of moving an object from his place. It is not easy because in many cases the director is not interested in what you do**
- ❑ **If successful, go to next page, otherwise start from scratch!**

Step 7: insure the objects

- **Find an insurance that doesn't cost the earth for the transport and the period of time the object will be away (it can be long due to activation). Be careful, people can be afraid of activation so explain very clearly that the object are absolutely safe when non active**

- **If successful, go to next page, otherwise start from step 6!**

Step 8: move the objects

Arrange for the transport of the artefacts:

- ❑ **Find a company that meets all the requirements of the museum (first choice for curator, last for us, too expensive)**
- ❑ **The curator brings the objects with her/him. Much better, since most likely the curator needs to be there anyway "to supervise" you when you handle the objects, or do it, if they are very strict.**
- ❑ **Bring the object with you. The curator trusts you and doesn't come**
- ❑ **If successful, go to next page, otherwise start from scratch!**

More on paperwork...

Paperwork needed to transport the objects:

- ❑ **Authorization from Ministry of Cultural Heritage (can takes weeks)**
- ❑ **Letter stating that the objects are transported for <u>scientific purposes only</u>. It is useful to prevent you from going in jail for illegal trafficking of cultural heritage**
- ❑ **Now you are ready for your experiment**

Step 9: optimal samples

Calculating and optimizing the size and shape of your sample is important in neutron scattering

- ❑ **Need to know roughly how long you need to count for**
- ❑ **Need to minimise multiple scattering**
- ❑ **Need to minimise self attenuation**

In order to calculate and correct for these, the sample should ideally be a regular shape. Archaeological sample are never regular shapes…….

Scattering unit

Most commonly this is the sample formula
e.g. sample of polythene (monomer is ethylene)

	barns			
	σ_{coh}	σ_{inc}	σ_{abs}*	M_f
C	5.550	0.001	0.3326	12.011
H	1.7568	80.26	0.0035	1.0079
C_2H_4	18.127	321.04	0.6792	28.054

Cross sections found in tables, e.g. V F Sears, Neutron News 3 1992
Can also choose atoms, unit cells, etc. Must be self consistent
*** Remember that σabs is wavelength dependent**

Scattering unit

Number of scattering units in the sample (mass, m) is given by

$$N_s = \frac{m}{M_f} N_A$$

where M_f is the mass of the formula unit and N_A=6.022 x 10^{23} mol^{-1} is Avogadro's Number. Similarly the number density is given by

$$n_s = \frac{\rho}{M_f} N_A$$

where ρ is the density of the sample.

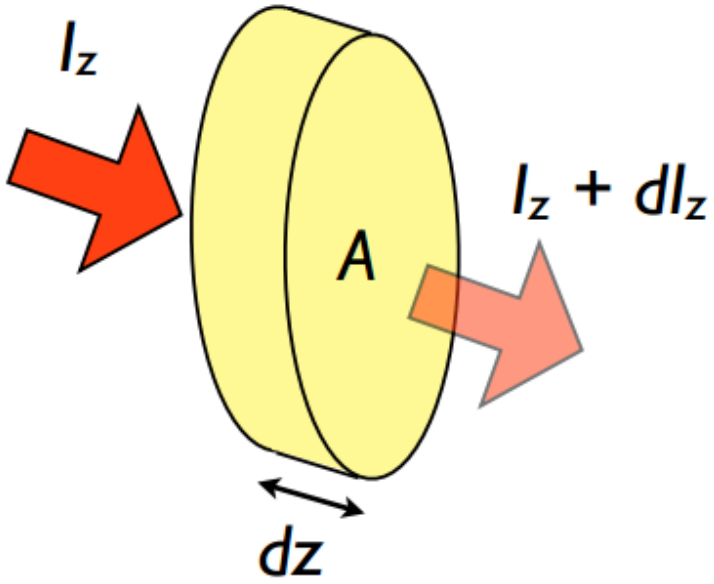
If we express N_A in units of x 10^{24} mol^{-1} then we get the number density in units of x 10^{24} cm^{-3} ≡ $(barn.cm)^{-1}$ which is useful later on. i.e.

$$n_s = \frac{\rho}{1.661 M_f}$$ (with ρ in g cm^{-3} and M_f in g)

e.g. Polythene: ρ = 0.93 g cm^{-3}, M_f = 28.054 g

so: n_s = 0.02 $(barn.cm)^{-1}$ ≡ 2 x 10^{22} cm^{-3}

Transmission



Assume thin slab of material with molecules having an absorption cross-section, σ

n_s x A x dz molecules in the slab

so total absorbing area is $\sigma n_s A dz$

and fraction of neutrons absorbed is the total absorbing area divided by the total area, therefore

$$\frac{dI_z}{I_z} = -\sigma n_s dz$$

Integrating both sides: $\ln(I_z) = -\sigma n_s z + C$

So for thick slab of thickness *t*, we have $I_z = I_0$ at $z = 0$ and $I_z = I_1$ at $z = t$, and

$$\ln(I_1) - \ln(I_0) = (-\sigma n_s t + C) - (-\sigma n_s 0 + C) = -n_s \sigma t$$

Finally, exponentiating both sides, we find the expression for the neutron transmission

$$T = \frac{I_1}{I_0} = \exp(-n_s \sigma t)$$

Beer's Law

Optimal size

Experience shows that a good neutron sample is one which scatters around 10% of the incoming beam. Much less than this, the count-rate is too small. Much more, *self-attenuation* and especially *multiple scattering* become problematic

To calculate the thickness of a 10% scattering sample we write

$$T = \frac{I_1}{I_0} = \exp(-n_s \sigma t) = 0.9 \qquad \text{90 \% un-scattered}$$

$$\Rightarrow n_s \sigma t = 0.1$$

In this case the cross-section we need is the total scattering cross-section, σ_T

e.g. Polythene: σ_T = 339.167 barns, n_s = 0.02 (barn.cm)$^{-1}$

so: $t_{10\%}$ = 0.1 / (339.167 x 0.02)

= 0.015 cm

So only around 0.1 mm thickness of polythene scatters 10% of the beam
This indicates the strength of the scattering of neutrons from hydrogen

Strong absorbers

There are a few "boagie" materials which prove challenging for neutron experiments due to their massive absorption cross-sections

element	σ_{abs}
Gd	49700 b
B	767 b
Cd	2520 b
Li	70 b
Ir	425 b
Dy	994 b
Sm	5922 b
^{3}He	5333 b

The numbers here are given for "thermal neutrons" (λ = 1.8 Å)

In general at low energies, the absorption cross-section is linear with wavelength

$$\sigma_{\mathrm{abs}}(\lambda) = \frac{\lambda \sigma_{\mathrm{abs}}^{\lambda=1.8}}{1.8}$$

Sometimes the high absorption can be avoided by choice of isotope - e.g. ^{160}Gd and ^{11}B have low absorption

Many of these elements are used in neutron detectors

Strong absorbers

Big absorbers are very useful in neutron shielding design

e.g. How much Cd is needed to absorb 99.9% of thermal neutrons?

$n_s = 0.046$ (barn.cm)$^{-1}$, $\sigma_{abs} = 2520$ b

$t = -\ln(T) / n_s\sigma_{abs} = -\ln(0.001) / 115.9 = 0.6$ mm

For absorbing samples the 10% rule is generally not used

e.g Mn metal: $\sigma_T = 2.15$ b, $n_s = 0.08$ (barn.cm)$^{-1}$, $\sigma_{abs} = 13.3$ b

so: $t_{10\%} = 6$ mm

Suppose we're doing an experiment on IN6 with a wavelength of 5.1 Å

$\sigma_{abs}(5.1Å) = 5.1 * 13.3 / 1.8 = 37.7$ barns

so the fraction of neutrons absorbed in a 10% scattering sample is

$1 - \exp(-n_s\sigma t) = 1 - \exp(-1.8) = 0.84$

i.e. 84% of the incoming **& scattered** neutrons are absorbed

So in this case we need a compromise solution

Strong absorbers

Assuming uniform sample shape (e.g. slab for SANS or cylinder for diffraction) then we can make the rough approximation that the neutrons all traverse a similar path through the sample

Then we can write the fraction of neutrons scattered as

$$\begin{aligned} \Sigma &= T\left(1-\exp[-n_s\sigma_T t]\right) \\ &= \exp[-n_s\sigma_{\rm abs} t]-\exp[-n_s(\sigma_{\rm abs}+\sigma_T)t] \end{aligned}$$

To maximise this, we differentiate and set to zero to find

$$\frac{d\Sigma}{dt} = n_s(\sigma_{\rm abs}+\sigma_T)\exp[-n_s(\sigma_{\rm abs}+\sigma_T)t]-n_s\sigma_{\rm abs}\exp[-n_s\sigma_{\rm abs}t]=0$$

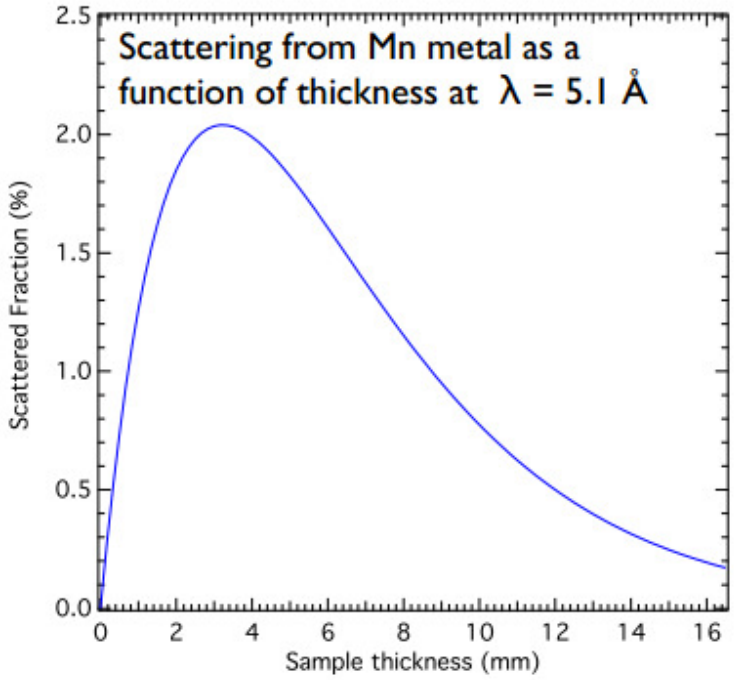
$$\Rightarrow \quad \exp[-n_s\sigma_T t]=\frac{\sigma_{\rm abs}}{\sigma_{\rm abs}+\sigma_T}$$

$$\Rightarrow \quad t=\frac{\ln(\sigma_{\rm abs}+\sigma_T)-\ln(\sigma_{\rm abs})}{n_s\sigma_T}$$

Strong absorbers

So taking the previous example of Mn metal at 5.1 Å, the optimum thickness is

$$t = \frac{\ln(37.7 + 2.15) - \ln(37.7)}{0.08 * 2.15} \simeq 3.2 \text{ mm}$$



At this thickness the fraction of scattered neutrons is around 2%

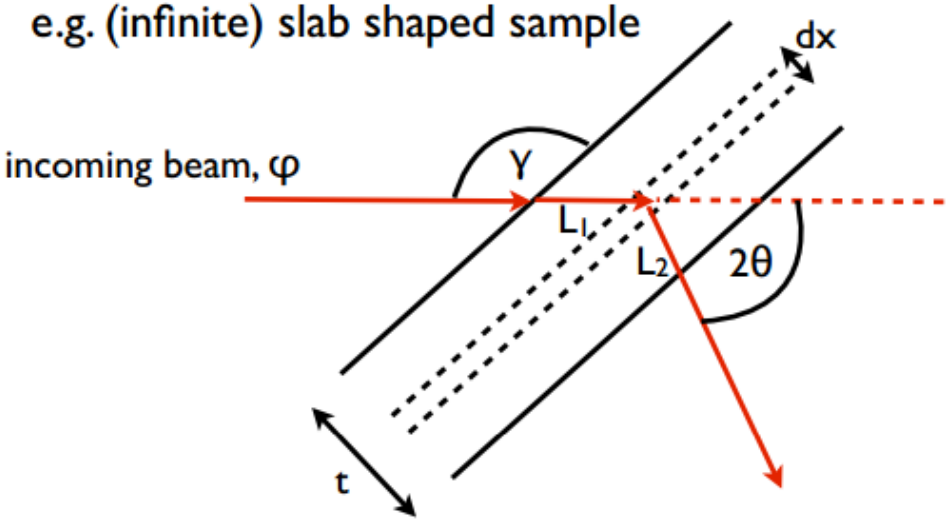
This occurs at a sample transmission of 38 % - which is roughly 1/e

More difficult to optimise sample size for broad wavelength bands - choose lowest useful wavelength in band and optimise there

Transmission - Slab

In order to calculate the attenuation of neutrons through a sample *as a function of scattering angle* we need to know the shape of the sample, and average over all possible paths through the sample.

e.g. (infinite) slab shaped sample



Neutron scattered from layer thickness dx at depth x

$$L_1 = \frac{x}{\sin\gamma}$$

$$L_2 = \frac{t-x}{\sin(\gamma - 2\theta)}$$

Incoming flux φ attenuated along L_1

$$\phi = \phi_0 \exp(-N\sigma L_1)$$
$$= \phi_0 \exp(-\frac{N\sigma x}{\sin\gamma})$$

The outgoing beam is attenuated along L_2

$$T_2 = \exp\left(-N\sigma \frac{t-x}{\sin(\gamma - 2\theta)}\right)$$

(assuming diffraction condition $k_i = k_f$)

Transmission - Slab

Outgoing beam proportional to cross-section into solid angle $\Delta\Omega$, flux, n_s, thickness of slab Δt and is attenuated along L_2

$$\Delta N = \phi n_s \Delta t \left(\frac{d\sigma}{d\Omega}\right) T_2 \Delta\Omega$$
$$= \phi_0 n_s \exp\left(-\frac{n_s\sigma x}{\sin\gamma}\right) \frac{dx}{\sin\gamma} \left(\frac{d\sigma}{d\Omega}\right) \exp\left(-\frac{n_s\sigma(t-x)}{\sin(\gamma-2\theta)}\right) \Delta\Omega$$

So total neutron scattered is found by integrating above wrt. x between x=0 and x=t

$$N = \phi_0 n_s \left(\frac{d\sigma}{d\Omega}\right) \Delta\Omega \frac{1}{\sin\gamma n_s\sigma(\mathrm{cosec}\gamma - \mathrm{cosec}(\gamma-2\theta))} \left\{\exp\left(-\frac{n_s\sigma t}{\sin(\gamma-2\theta)}\right) - \exp\left(-\frac{n_s\sigma t}{\sin\gamma}\right)\right\}$$

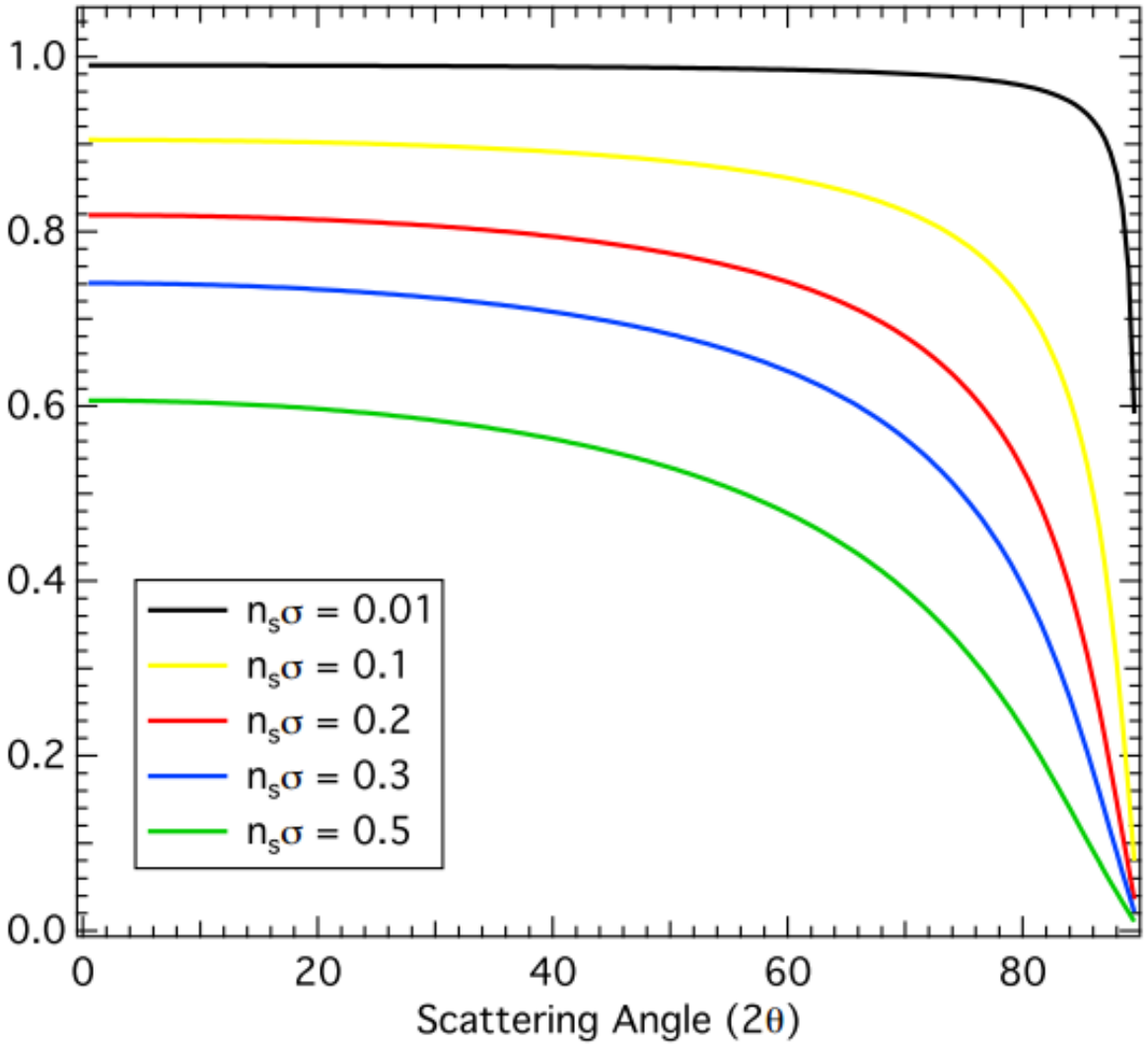
In order to get the attenuation factor we need to know the number of counts in the limit of zero absorption, N_0 and divide

Finally we get

$$T = \frac{1}{n_s\sigma t(\mathrm{cosec}\gamma - \mathrm{cosec}(\gamma-2\theta))} \left\{\exp\left(-\frac{n_s\sigma t}{\sin(\gamma-2\theta)}\right) - \exp\left(-\frac{n_s\sigma t}{\sin\gamma}\right)\right\}$$

This is one of the very few solvable cases for transmission as a function of 2θ

Transmission - Slab



Transmission plotted for slab normal to incident beam (e.g. SANS)

Thickness of sample = 1 cm

Slab geometries for samples are also commonly used in Neutron spin-echo experiments

Beyond 90° - in reflection geometry - another (similar) expression exists

Transmission - Cylinder

With the advent of large detector arrays which commonly surround the sample (in the equatorial plane) cylindrical sample geometries have become more common. Sometimes an annular cylinder is used to fill beam, but minimise absorption

In these cases there is no analytic expression for the transmission as a function of angle and the attenuation must be calculated numerically (i.e. lookup tables or Monte-Carlo algorithms are commonly used

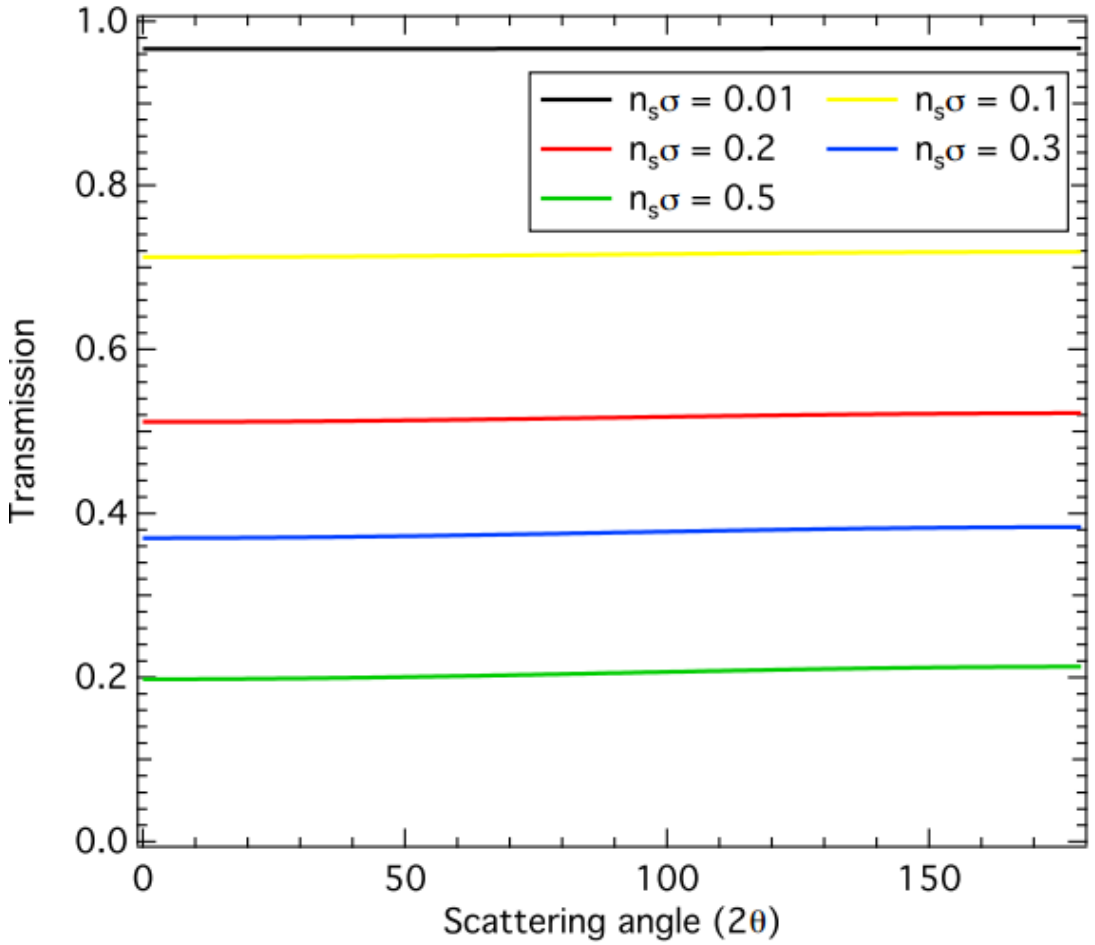
In the case of a solid cylinder or sphere with $n_s\sigma \leq 1$ a good approximation (better than 0.5% accuracy) is given by the expression

$$T = \exp\left\{-(a_1 + b_1 \sin^2\theta) n_s\sigma R - (a_2 + b_2 \sin^2\theta)(n_s\sigma R)^2\right\}$$

where R is the radius of the cylinder/sphere, and the coefficients, a_1, b_1, a_2 and b_2 have the values

	a_1	b_1	a_2	b_2
cylinder	1.7133	-0.0368	0.0927	-0.0375
sphere	1.5108	-0.0315	-0.0951	-0.2898

Transmission - Cylinder



Much flatter transmission as a function of angle - so friendly for wide-angle instruments

Generally transmission is assumed to be constant as a function of angle in these cases

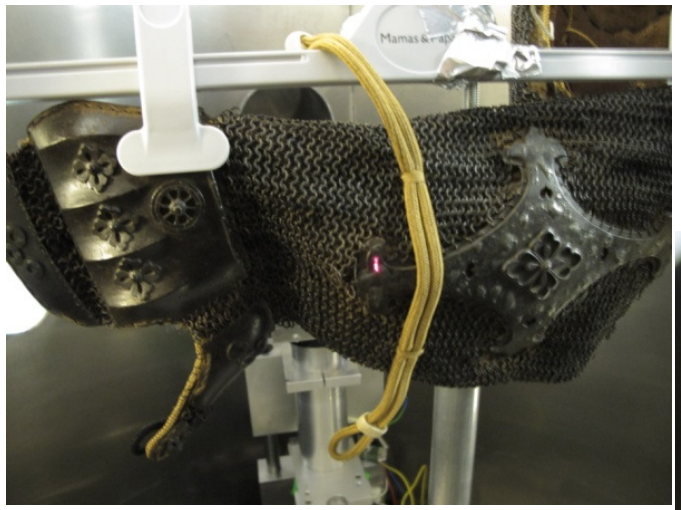
Sample holder

Choice of material for sample container / holder is crucial - and depends strongly on type of neutron experiment. Should be made as thin as possible to reduce mass in beam (typically 0.1 mm - 1 mm thickness)

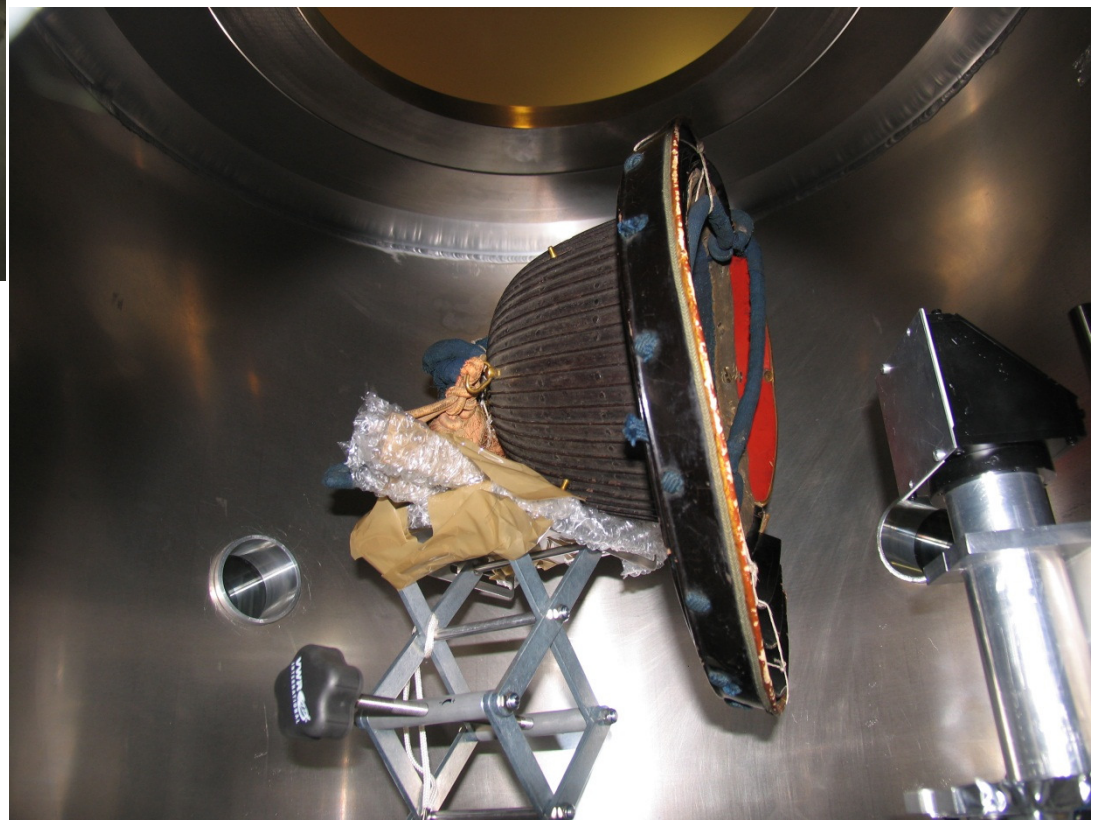
	Material	Comment
Powder diff.	vanadium	no Bragg peaks, large σ_{inc} some absorption
Inelastic scattering	Al / Cu	low-ish scattering cross-sections, low incoherent scattering (Cu for low T)
Polarized neutrons	Al / Cu	
Liquids / glasses	vanadium / TiZr	no Bragg peaks TiZr is a "null-matrix"
SANS	Quartz	very low small angle scattering
Single crystals	Small pin (Al)	sample holds own shape

Many thanks to Ross Stewart (ISIS)

Sample holder for archaeometry



Ideally, you put your samples in V pocket… Not always possible



Be creative!

Sample alignment

It is very important that the sample are aligned properly
To make sure that:

- **You are measuring what you want**

- **The distance between moderator and detector is correct**
  (to convert from ToF to d or wavelenght or Energy…)

  L=L1+L2, with L1=distance moderator – sample, and L2=distance sample – detectors

ToF equation:

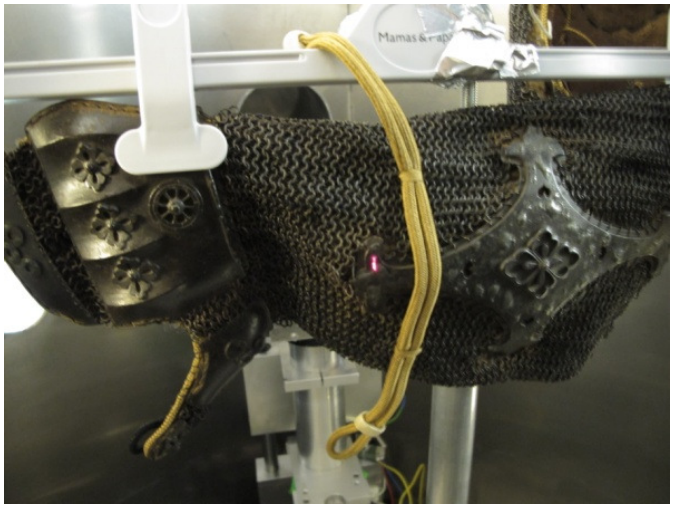
$$d(\text{Å}) = \frac{h}{2m_n}\frac{t(\mu s)}{L(m)\sin\Theta} = \frac{1}{505.56}\frac{t(\mu s)}{L(m)\sin\Theta}$$

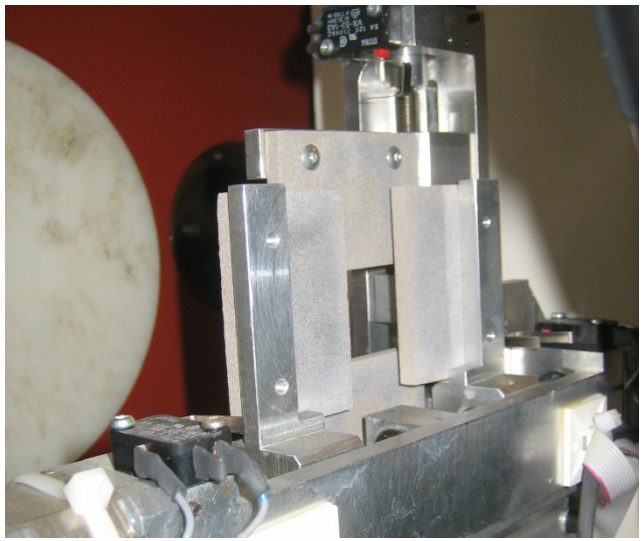
Time of flight

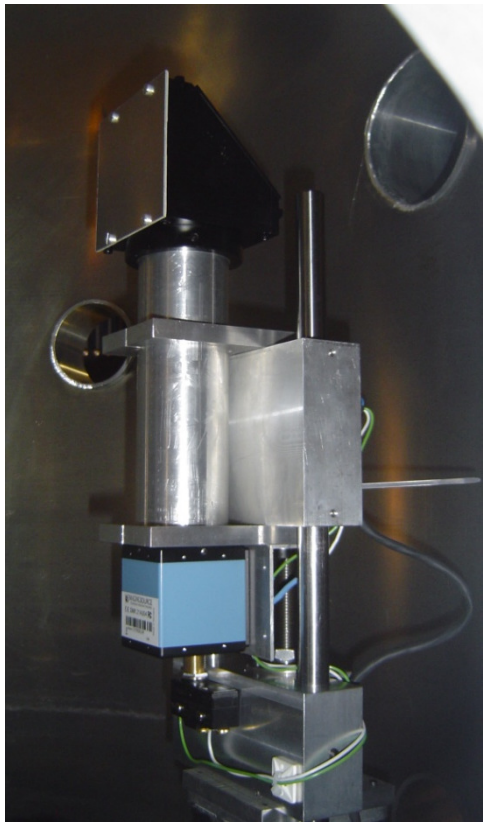
Distance

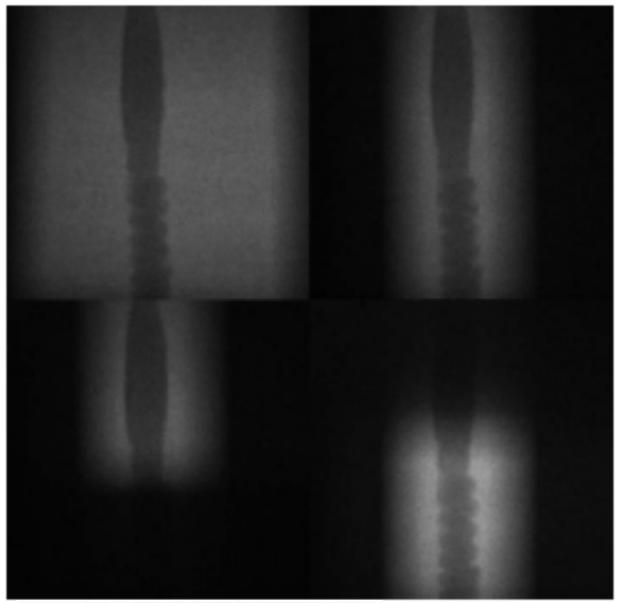
Scattering angle

Sample alignment









Data collection

- Hint: look at your data during experiment
- Better too long than too short, but be reasonable
- Do not be afraid to change your plan
- Try to get final results on less samples
- You can always measure more samples later, but, especially for CH, you cannot always have back the same samples

Counting time

The error bars in the measurements are given by Poisson Statistics $\delta(N) = \sqrt{N}$

and therefore the fractional error is $\frac{\delta(N)}{N} = \frac{1}{\sqrt{N}}$

so in order to improve the fractional error by a factor of x, we need to count a factor of x^2 more neutrons

Counting time

e.g. After 20 minutes we've counted 1000 in a detector. What's the fractional error bar?

$$\frac{\delta(N)}{N} = \frac{1}{\sqrt{1000}} = 0.032$$

How long do we need to count to get the error bar below 1%?

$$\frac{1}{\sqrt{N_{1\%}}} = 0.01$$

$$\Rightarrow N_{1\%} = 10000$$

the count rate is 50 counts per minute, so we need to count for 200 minutes.
NB In order to improve the error bars from around 3% to 1% we need to count 10x longer !

Data normalization

The total number of neutrons counted during a measurement depend on uninteresting factors, such as:

- **the counting time**
- **the solid angle coverage of the detectors**
- **the efficiency of the detectors**

Data normalization

To correct for counting time we can either:

- **Divide the measured counts by the counts measured in an incident beam montior**
- **Divide the measured counts by the counting time**

It's generally better to use a monitor since the flux may not be constant over time (e.g. ISIS). It is even more important if you instrument is downstream another (e.g. INES)

Data normalization

Monitor counters are generally low efficiency detectors made from:

- **Low pressure 3He**
- **^{235}U foils (fission chambers)**
- **Low density scintillator materials**

Data normalization: step 1

Divide your diffraction spectra by the incident monitor spectrum:

$$S_{norm}=S/S_{mon}$$

S_{norm}=Spectra normalized by monitor
S=Spectra
S_{mon}=Monitor Spectrum

Data normalization

- Vanadium has very high incoherent cross section compared with its negligible coherent cross section, therefore it scatters neutron all over the place homogeneously
- This feature is very useful since it allow us to correct for detectors efficiency and solid angle
- Vanadium measurement corrects also for the incident flux profile

Data normalization: step 2

Divide your diffraction spectra normalized by the incident monitor spectrum, by the Vanadium spectra normalized by monitor:

$$S_{norm2}=S_{norm}/V_{norm}$$

S_{norm2}=Spectra normalized by monitor and Vanadium
S_{norm}=Spectra normalized by monitor only
V_{norm}=Vandium spectra normalized by monitor

Background subtraction

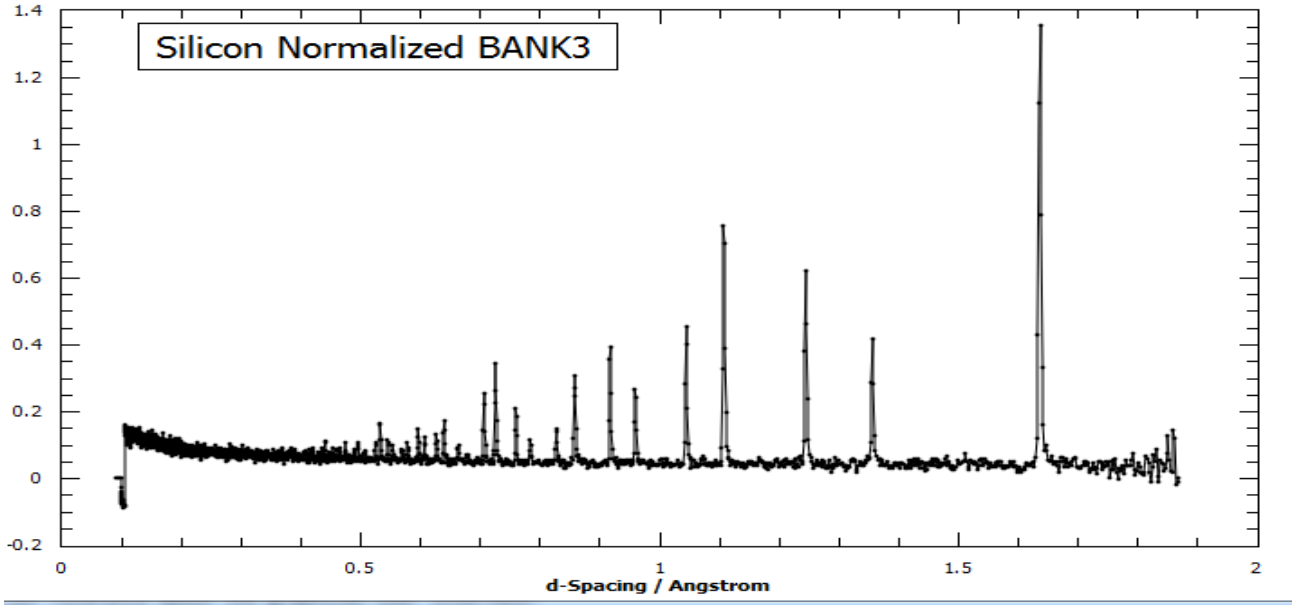
- In order to take into account the sample holder, one should measure an empty sample holder and subtract it from the data

- It is necessary to make a run with the instrument empty, and subtract it from the Vanadium

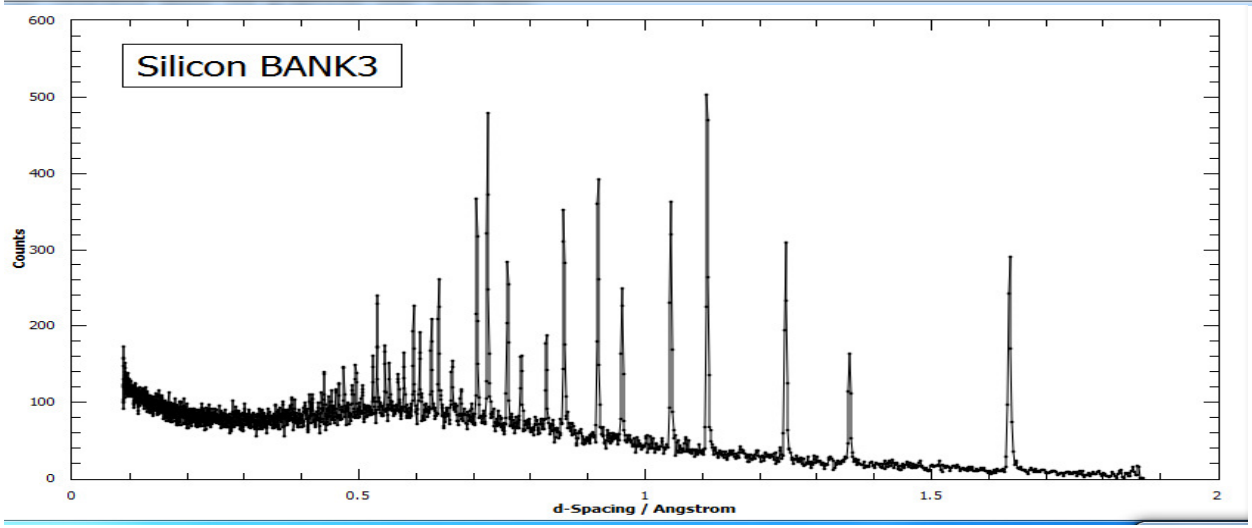
$$S_{norm3} = (S_{norm} - Sample\_holder) / (V_{norm} - Background)$$

S_{norm3}=data normalized, S_{norm}=data normalized by monitor
V_{norm}=V normalized by monitor

Comparison (Silicon)



Normalized



Not Normalized

Data analysis

- **Data analysis is the process of obtaining meaningful information from our measurements**

- **For diffraction experiment, this usually means refining your data (Rietveld method)**

Good refinement hints

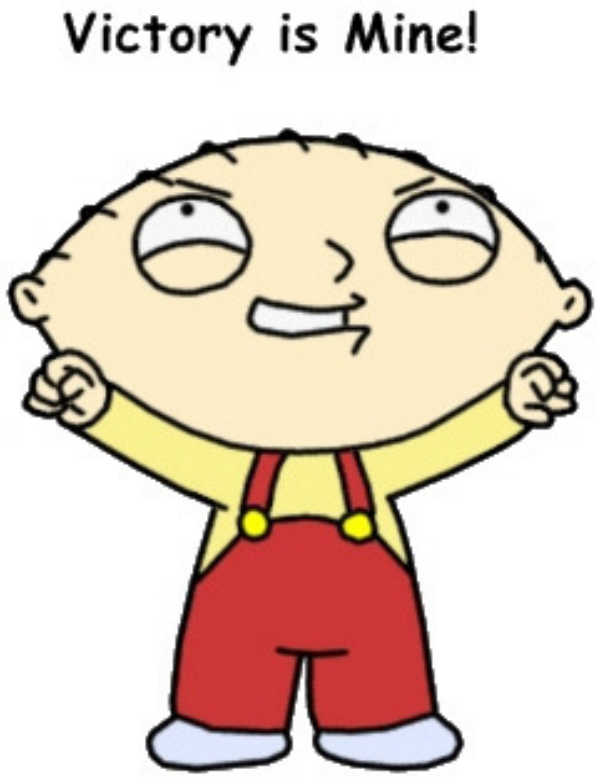
- First get a good experiment/spectrum
- Do not refine too many parameters
- Never stop at the first result
- Look carefully and constantly to the visual fit/plot and residuals during refinement process (no “blind” refinement)
- Zoom in the plot and look at the residuals. Try to understand what is causing a bad fit

End of the process

You have finished!!!

Write a paper, your thesis, be happy

Let's start again……



Thank you for your attention!