Practical Neutron Experiments

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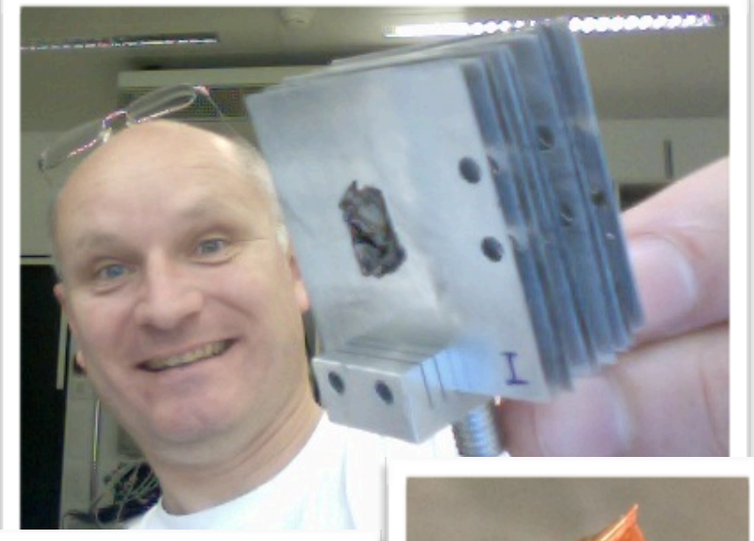
Outline

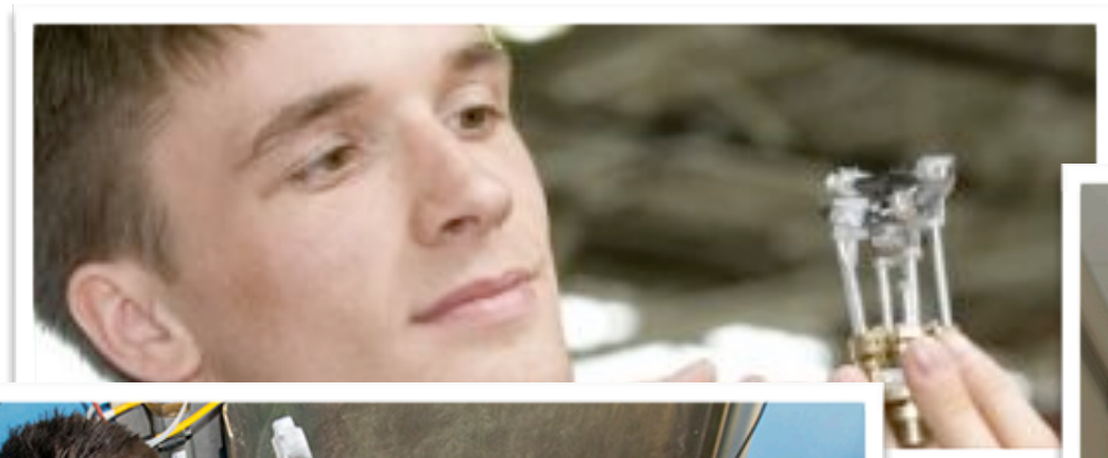
- Optimising sample size
- Sample shape - transmission of neutrons
- Normalisation of data
- Background Subtraction
- Counting time

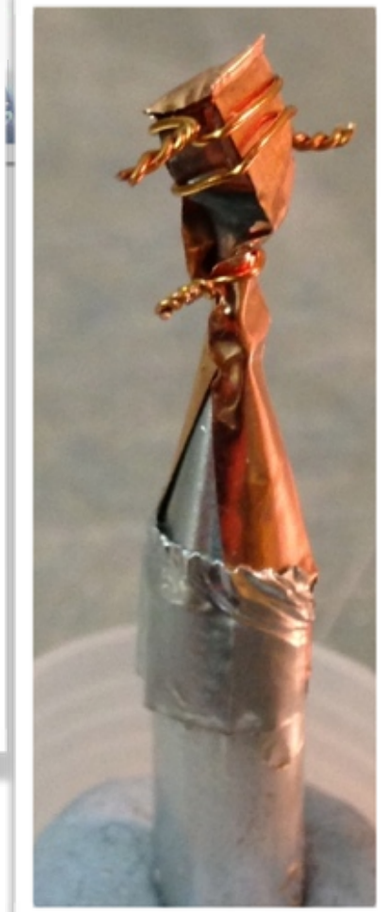
Sample preparation

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

Scattering unit

Most commonly this is the sample formula.

e.g. sample of polythene (monomer is ethylene)

C_2H_4

	barns			
	σ_{coh}	σ_{inc}	σ_{abs}*	M_f
C	5.55	0.001	0.3326	12.011
H	1.7568	80.26	0.0035	1.0079
C_2H_4	18.1272	321.042	0.6792	28.0536

per f.u.

Can also choose atoms, unit cells, etc. But must be *self consistent*

Cross sections found in tables, e.g. V F Sears, Neutron News **3** 1992

* Remember that σ_{abs} is wavelength dependent

Number Density of Sample

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}

Scattering Length Density

Commonly used in SANS and reflectometry, defined as the total scattering length in a given volume (e.g. choose volume of one f.u.)

$$\mathrm{SLD} = \frac{\sum_{i=1}^{N} b_i}{V_m}$$

where the subscript i runs over the number of atoms in the volume of interest, V_m . Since this volume is arbitrary, it makes sense to recast it in terms of the mass of a formula unit, Mf of the material divided by the density,

$$V_m = \frac{M_f}{\rho N_a}$$

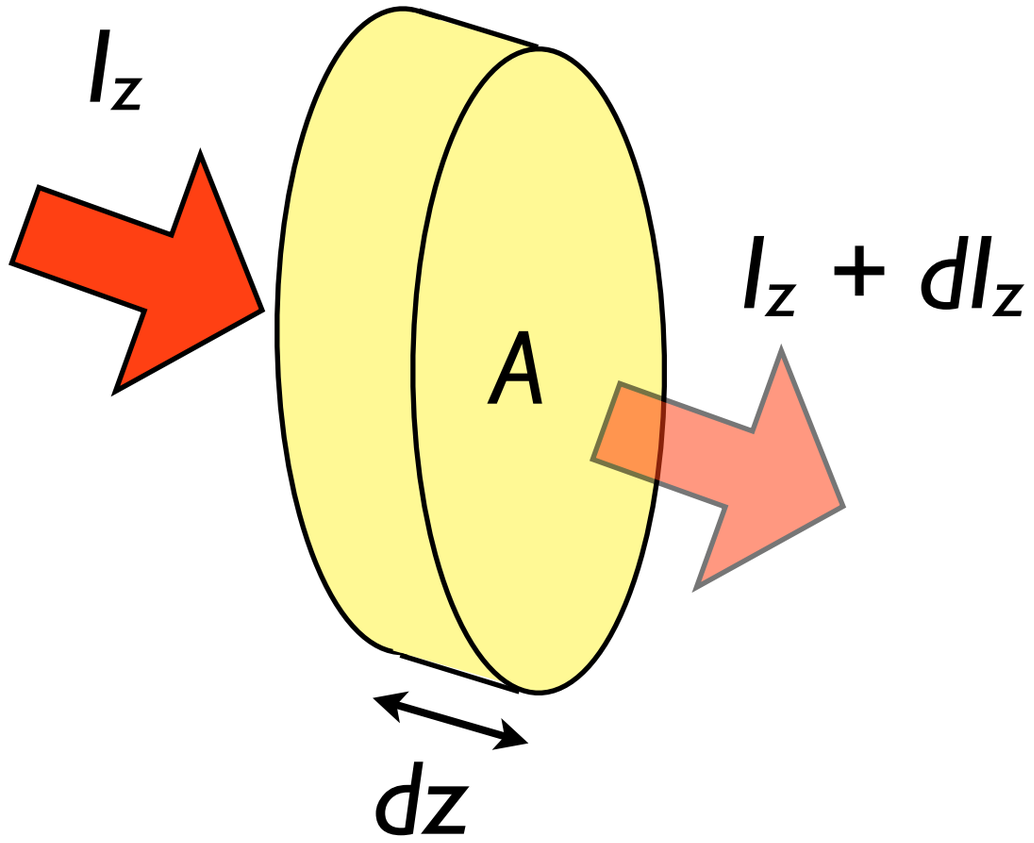
where ρ is the density of the sample.

We can now write the SLD as,

$$\mathrm{SLD} = \frac{\rho N_a \sum_{i=1}^{N} b_i}{M_f} = n \sum_{i=1}^{N} b_i$$

e.g. Polythene: SLD = 2×10^{22} x {2 x 6.65×10^{-13} + 4 x -3.73×10^{-13}}
= -3.24×10^{-9} cm^{-2}

Transmission of neutrons



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-Lambert 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)$^{-1}$, σ_{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: σ_T = 2.15 b, n_s = 0.08 (barn.cm)$^{-1}$, σ_{abs} = 13.3 b

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

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

σ_{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

Optimised sample size

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

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

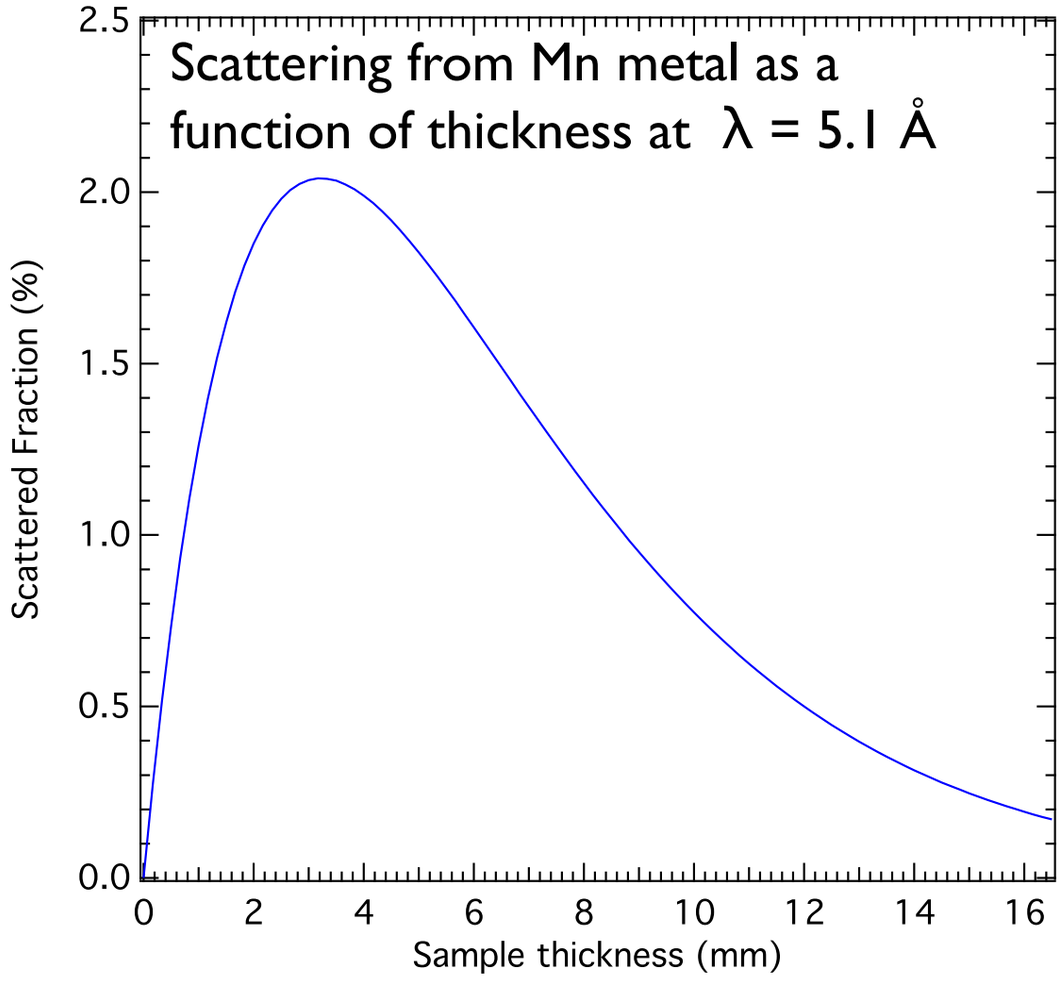
$$\Rightarrow \quad \exp[-n_s\sigma_T t]=\frac{\sigma_{\rm abs}}{\sigma_{\rm abs}+\sigma_T}$$

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

Optimised sample size

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%

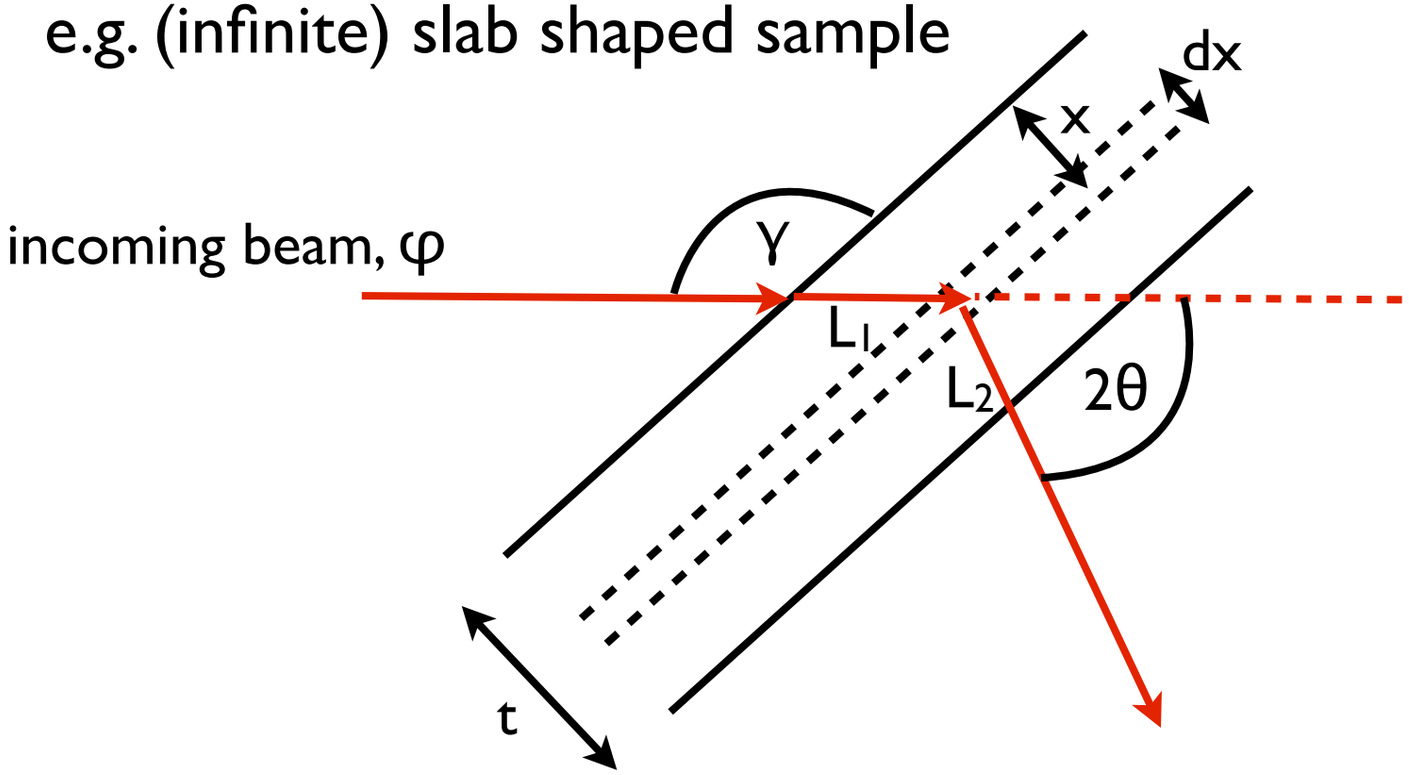
This occurs at a sample transmission of 38 %

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

Sample shape - 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

$$\phi = \phi_0 \exp(-n_s \sigma t)$$

$$= \phi_0 \exp\left(\frac{-n_s \sigma x}{\sin\gamma}\right)$$

The outgoing beam is attenuated along L_2

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

(assuming diffraction condition $k_i = k_f$)

Sample shape - slab

Outgoing number of neutrons is proportional to cross-section into solid angle ΔΩ, 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 neutrons scattered is found by integrating above wrt. x between x=0 and x=t

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

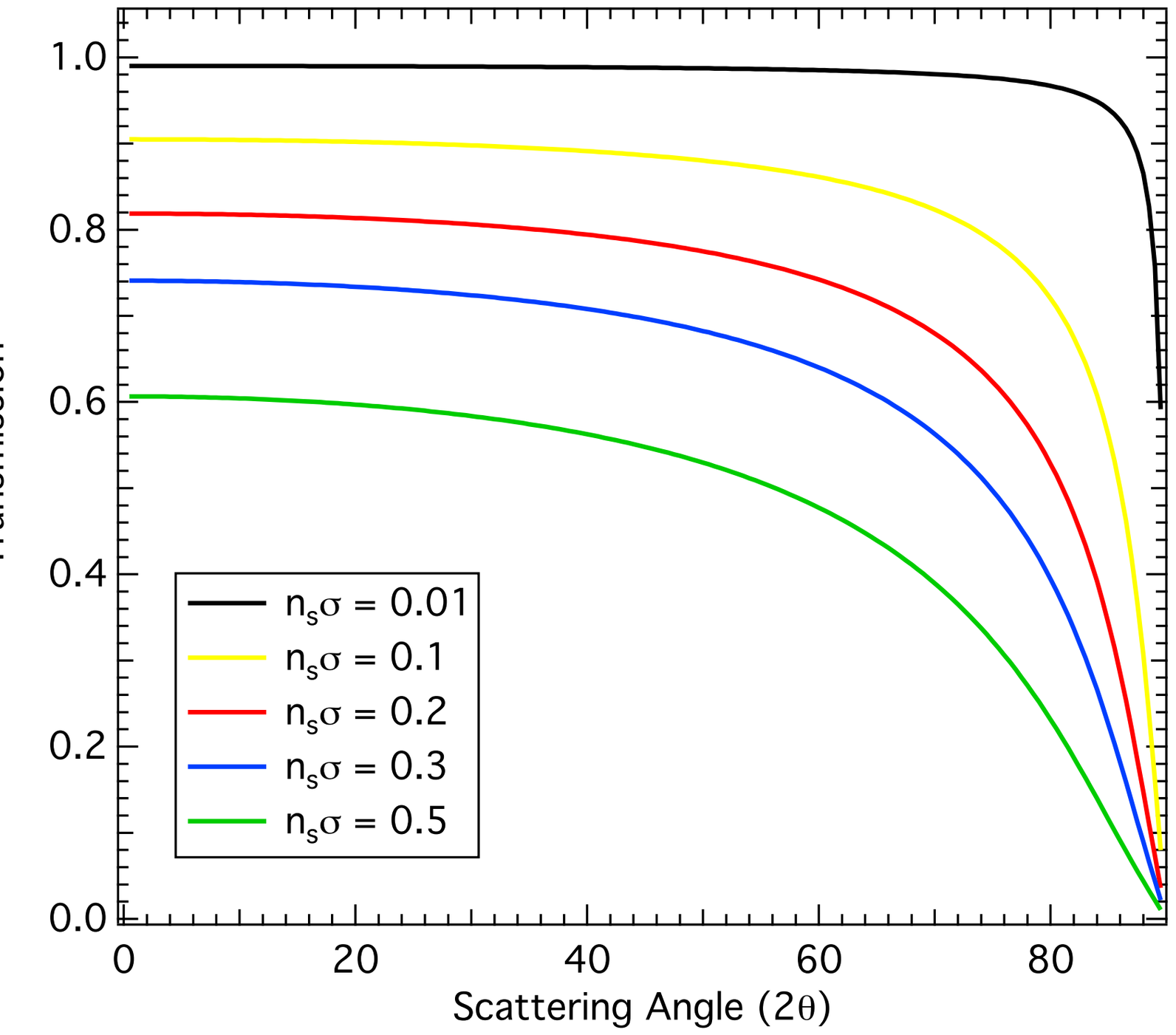
In order to get the attenuation factor we need to know the number of counts in the limit of zero absorption, N_0 and divide

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

Sample shape - 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

C G Windsor, Pulsed Neutron Scattering, Taylor and Francis (1981)

Sample shape - 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 analytical expression for the transmission as a function of angle - it 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

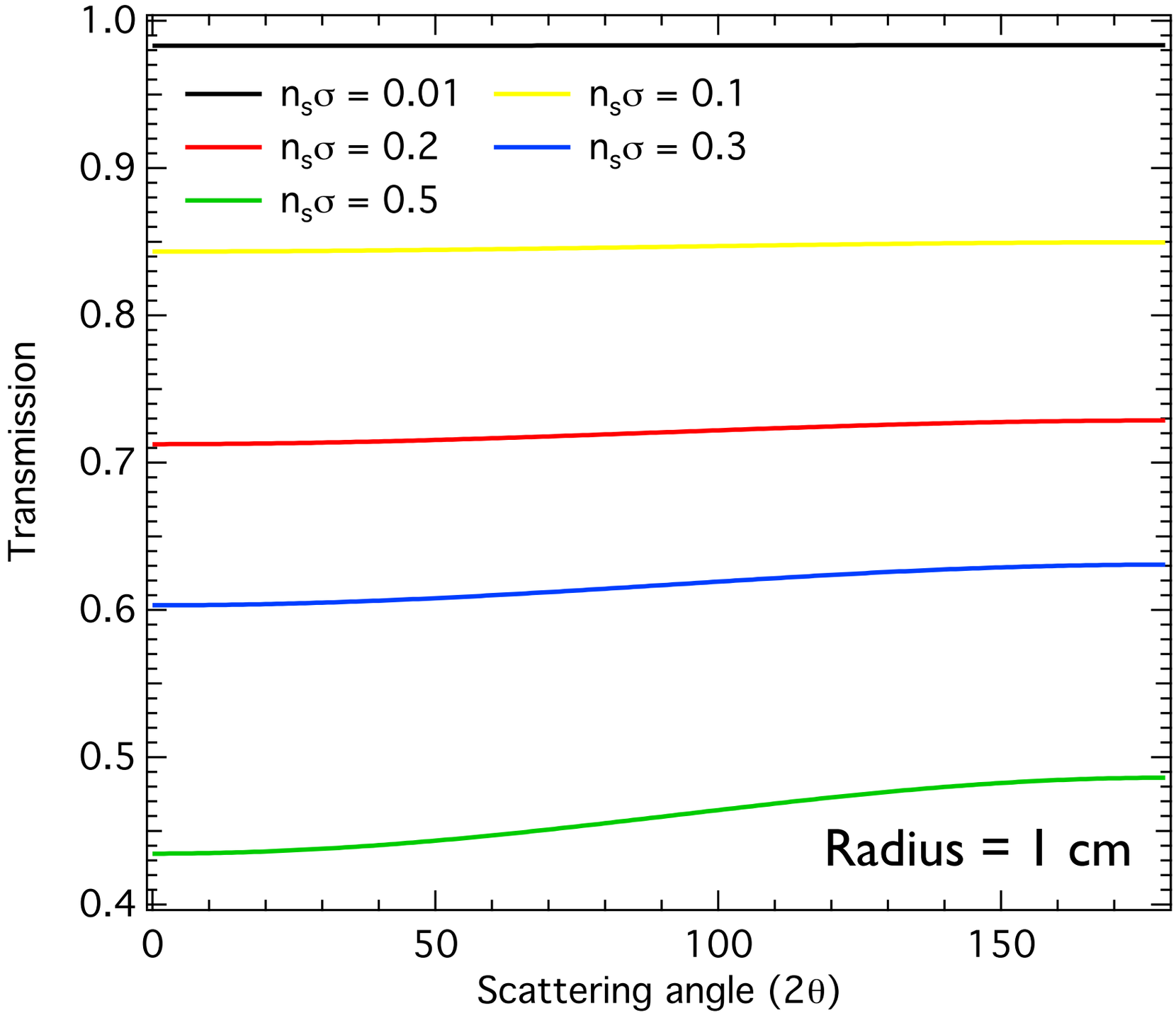
$$T = \exp\left\{-(a_1 + b_1 \sin^2\theta) n_s\sigma R - (a_2 + b_2 \sin^2\theta)(n_s\sigma R)^2\right\}$$

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	-0.3750
sphere	1.5108	-0.0315	-0.0951	-0.2898

A W Hewat, Acta. Cryst. A **35** (1975) 248

Sample shape - 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 Holders

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

Data Normalisation

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

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

To correct for counting time we can either:

1) Divide the measured counts by the counts measured in an *incident beam monitor*

2) 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)

Monitor counters are generally low efficiency (1 in 10^4) detectors made from:

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

Data Normalisation

The solid angle and detector efficiency can be corrected by dividing the measured counts by a measurement of vanadium

	abund. %	σ_{coh}	σ_{inc}	σ_{abs}
^{50}V	0.25	7.3	0.5	60
^{51}V	99.75	0.0203	5.07	4.9

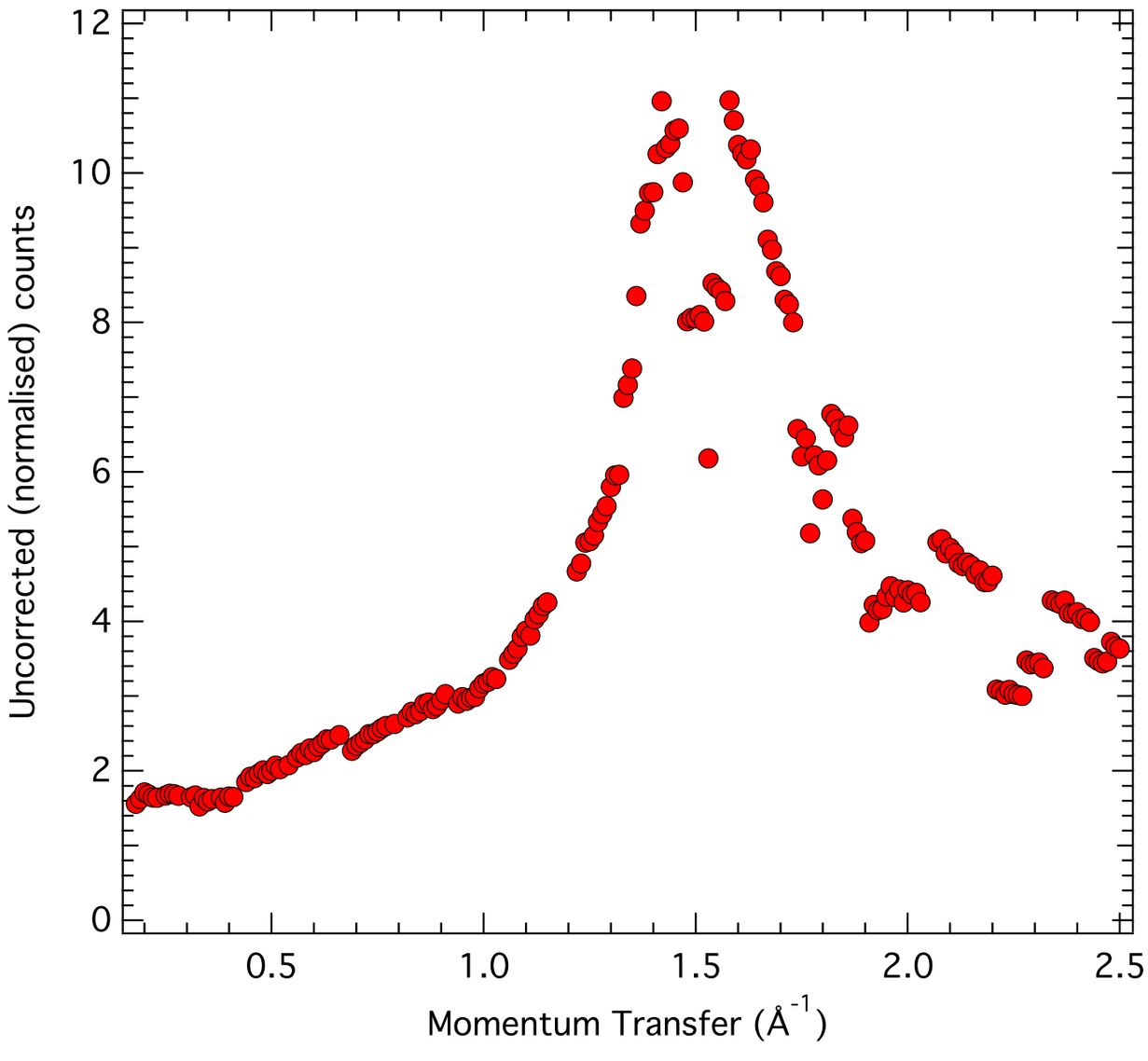
Due to the tiny coherent cross-section, and the abundance of the ^{51}V isotope V is approximated to be a purely incoherent scatterer

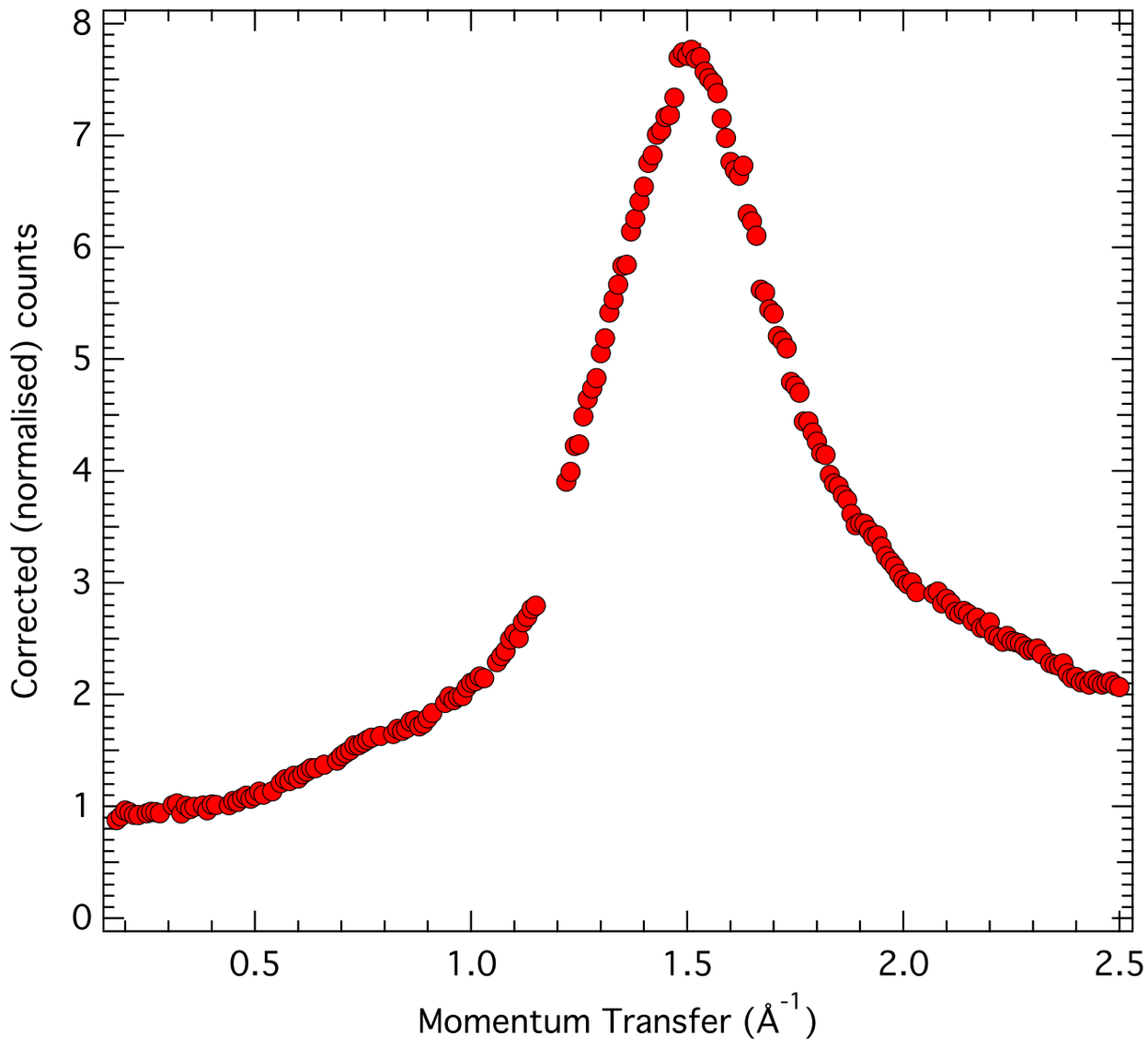
Therefore a vanadium count in a particular detector is proportional to the efficiency and solid angle coverage of that detector

Data Normalisation

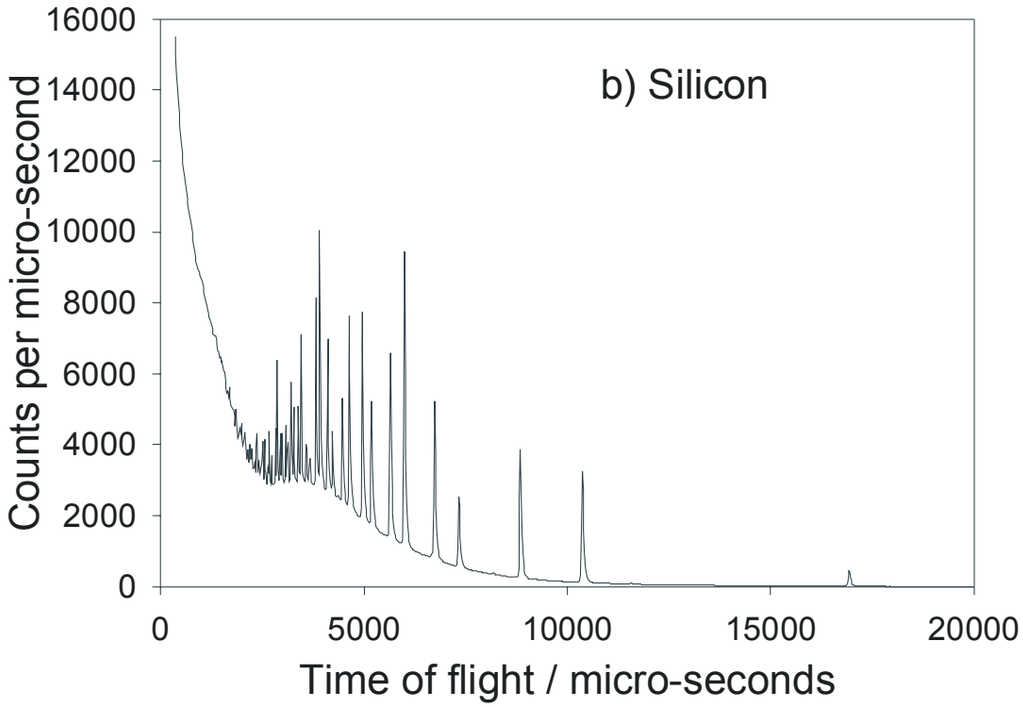
The solid angle and detector efficiency can be corrected by dividing the measured counts by a measurement of vanadium

Measurement of silica glass





Data Normalisation



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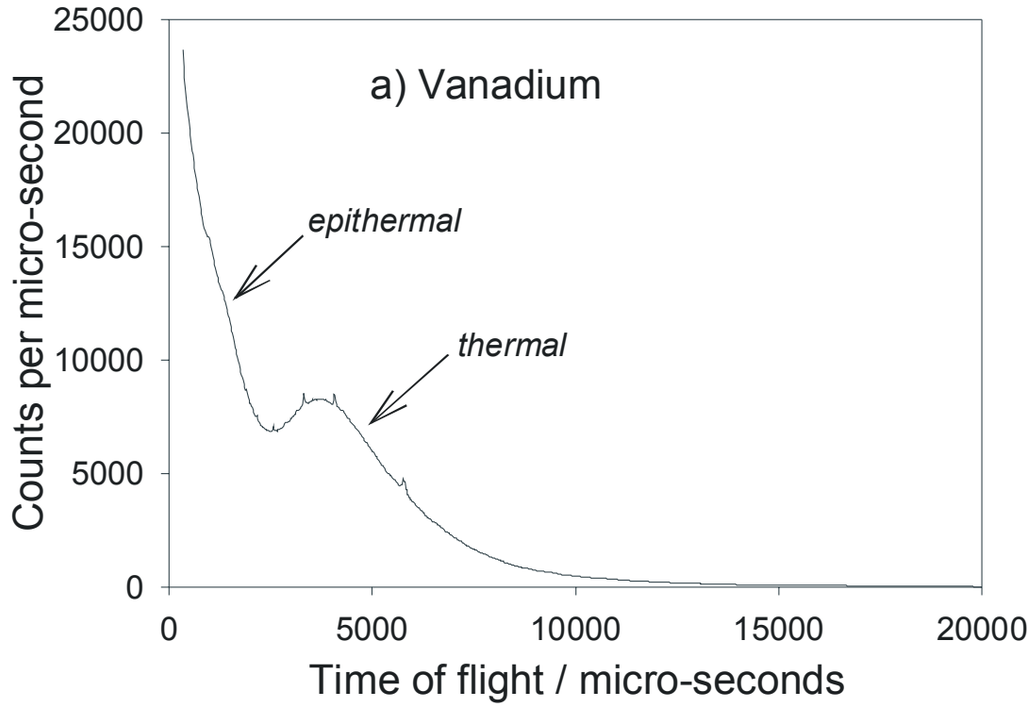

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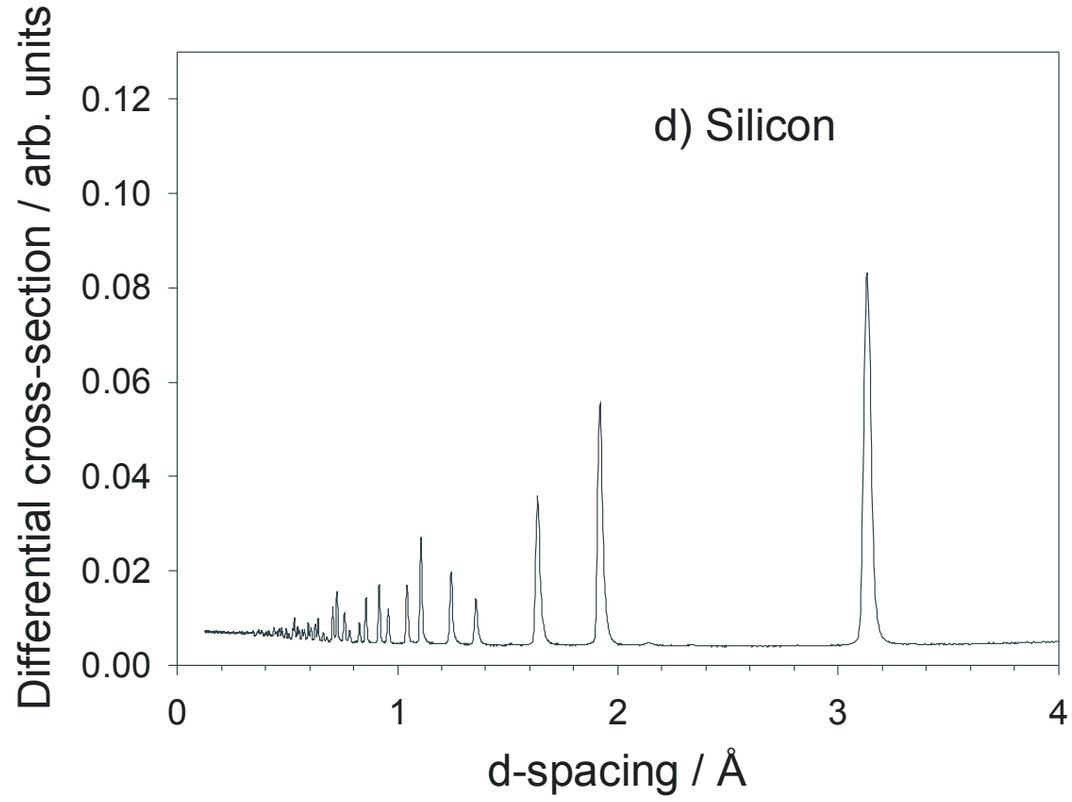
On a neutron time of flight diffractometer, the vanadium measurement additionally gives the flux profile as a function of wavelength. So division of the data by a vanadium measurement corrects also for the incident flux profile.



Absolute Normalisation

Since the spin incoherent cross-section of vanadium is well known, division of the data by vanadium can also be used to set the neutron data on an **absolute scale** of barns / ster. / scattering unit

For vanadium, $\sigma_{inc} = 5.07$
Therefore,

$$\left(\frac{d\sigma}{d\Omega}\right)_V = \frac{5.08}{4\pi}$$

The number of counts from the vanadium per second per solid angle is then (after transmission correction)

$$N_v = \phi n_{s(V)} t_V \frac{5.08}{4\pi} \Delta\Omega$$

So dividing the sample counts by the vanadium counts we get

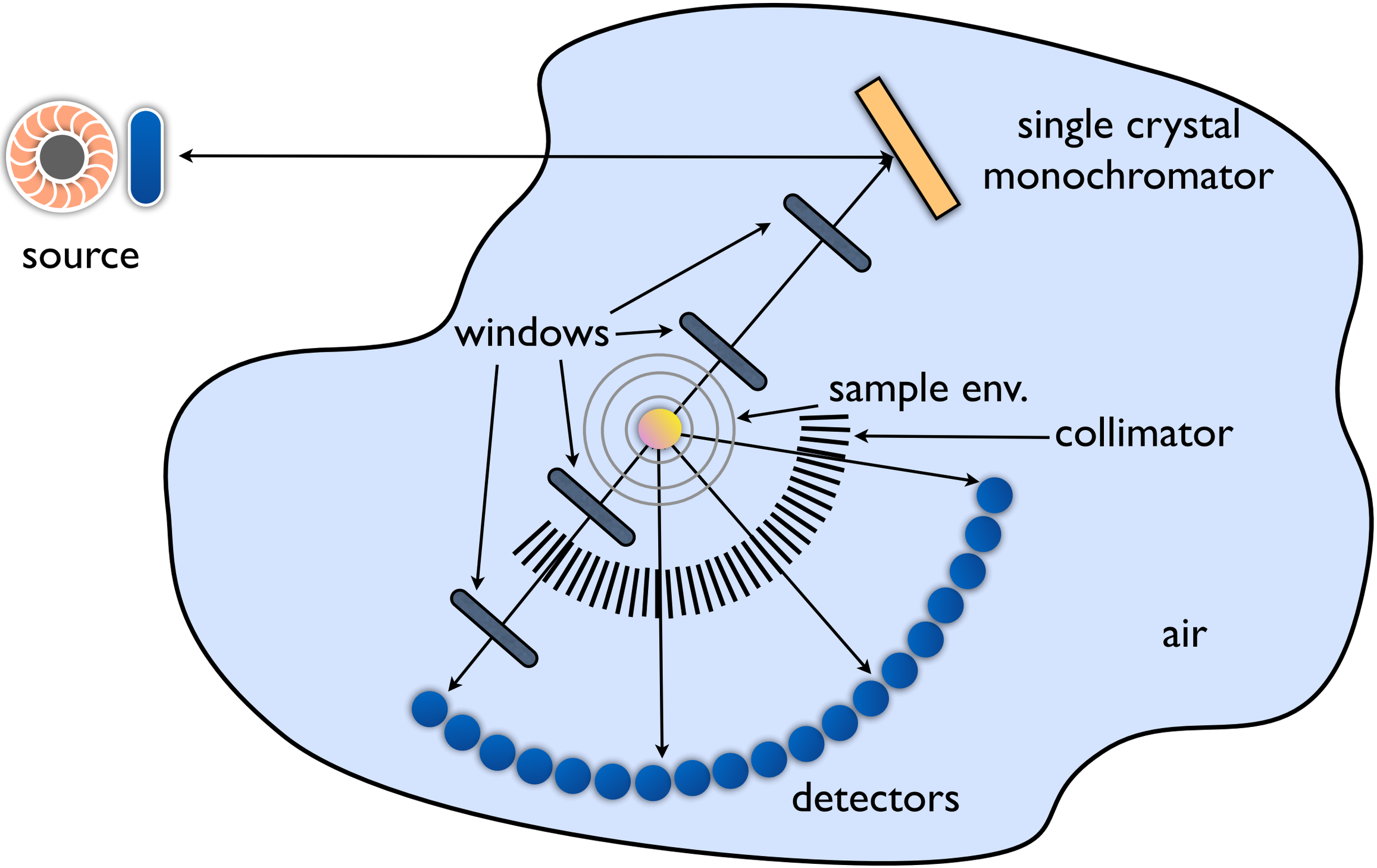
$$\frac{N_S}{N_V} = \frac{n_{s(S)} t_S \left(\frac{d\sigma}{d\Omega}\right)_S}{n_{s(V)} t_V \frac{5.08}{4\pi}}$$

and therefore

$$\left(\frac{d\sigma}{d\Omega}\right)_S = \frac{N_S}{N_V} \frac{n_{s(V)}}{n_{s(S)}} \frac{t_V}{t_S} \frac{5.08}{4\pi}$$

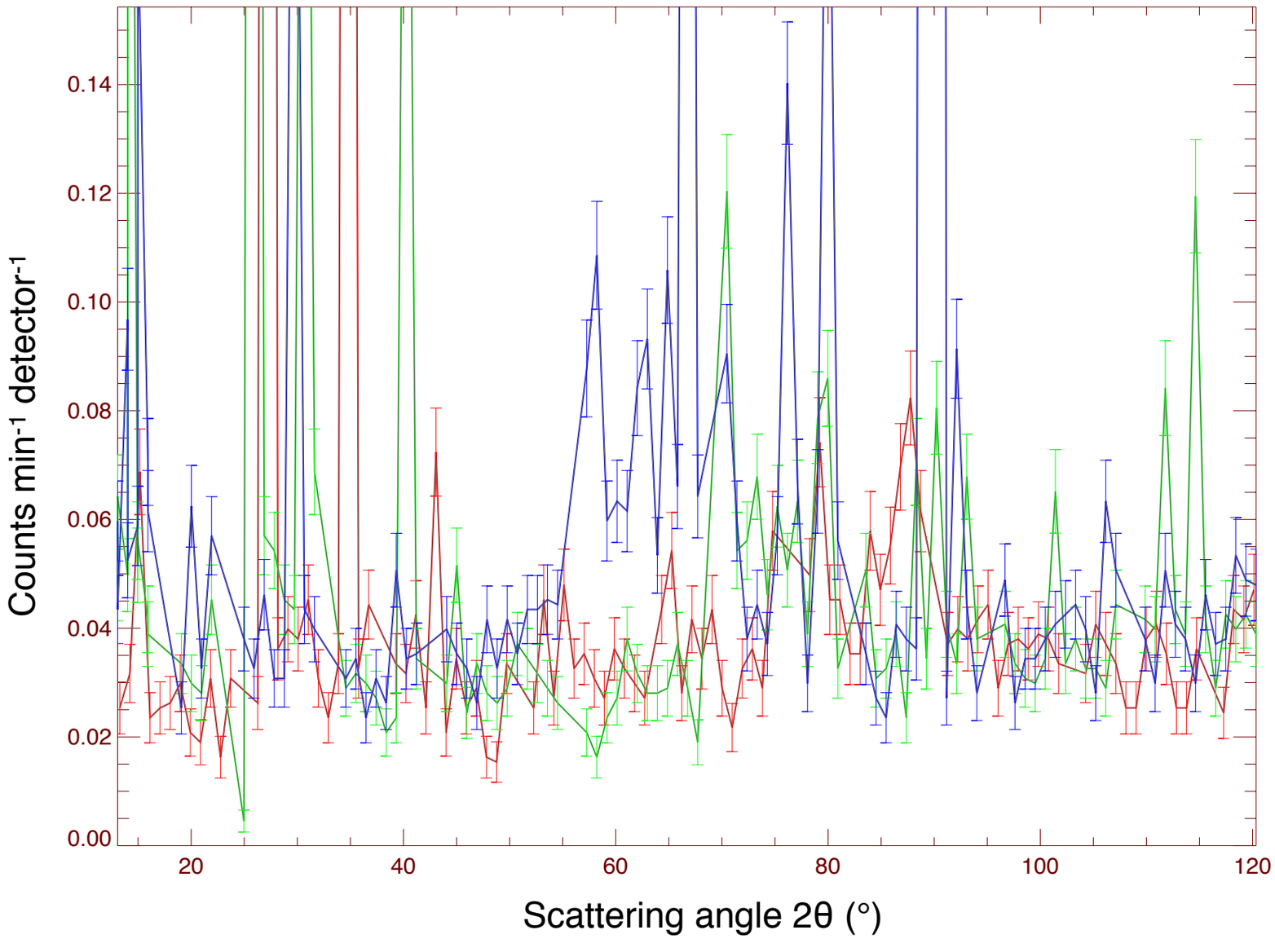
can substitute ratio of volumes here for non-slabs

Background subtraction



Case Study: IN4 (ILL)

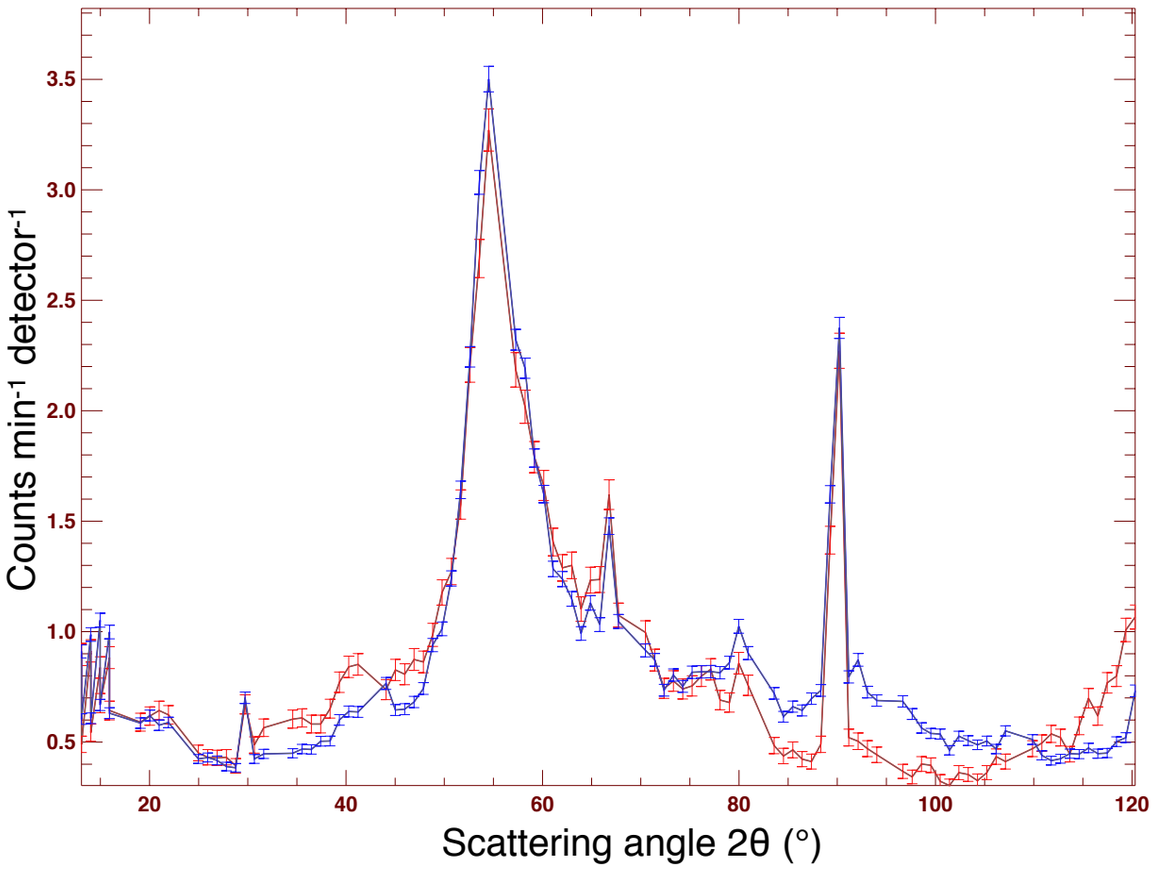
1) Reactor off



Background from electronic noise in detectors, cosmic rays or perhaps natural radioactivity

Case Study: IN4 (ILL)

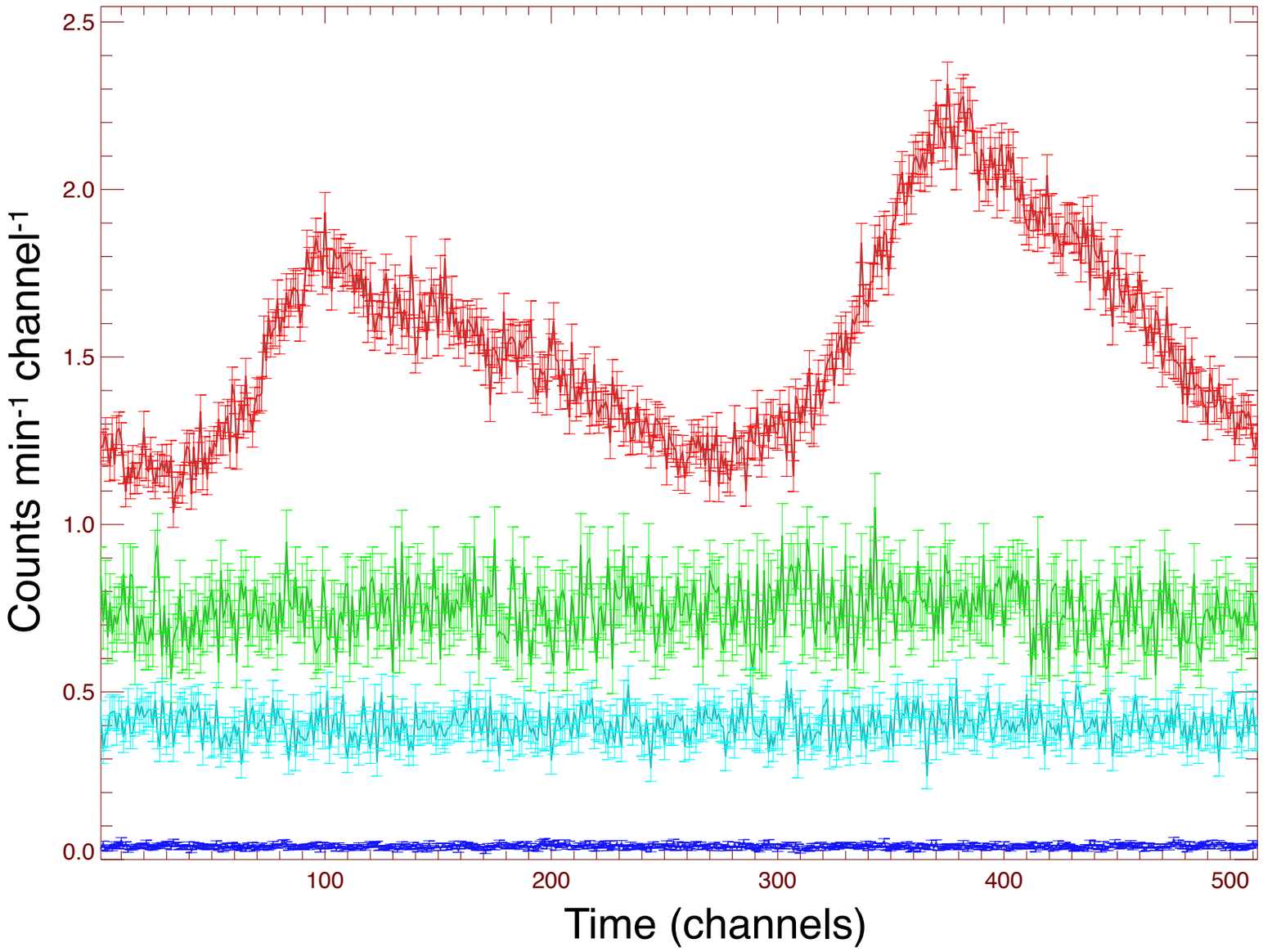
2) Reactor on: beam shutter closed



Background from "Neutron gas" in experimental hall

Case Study: IN4 (ILL)

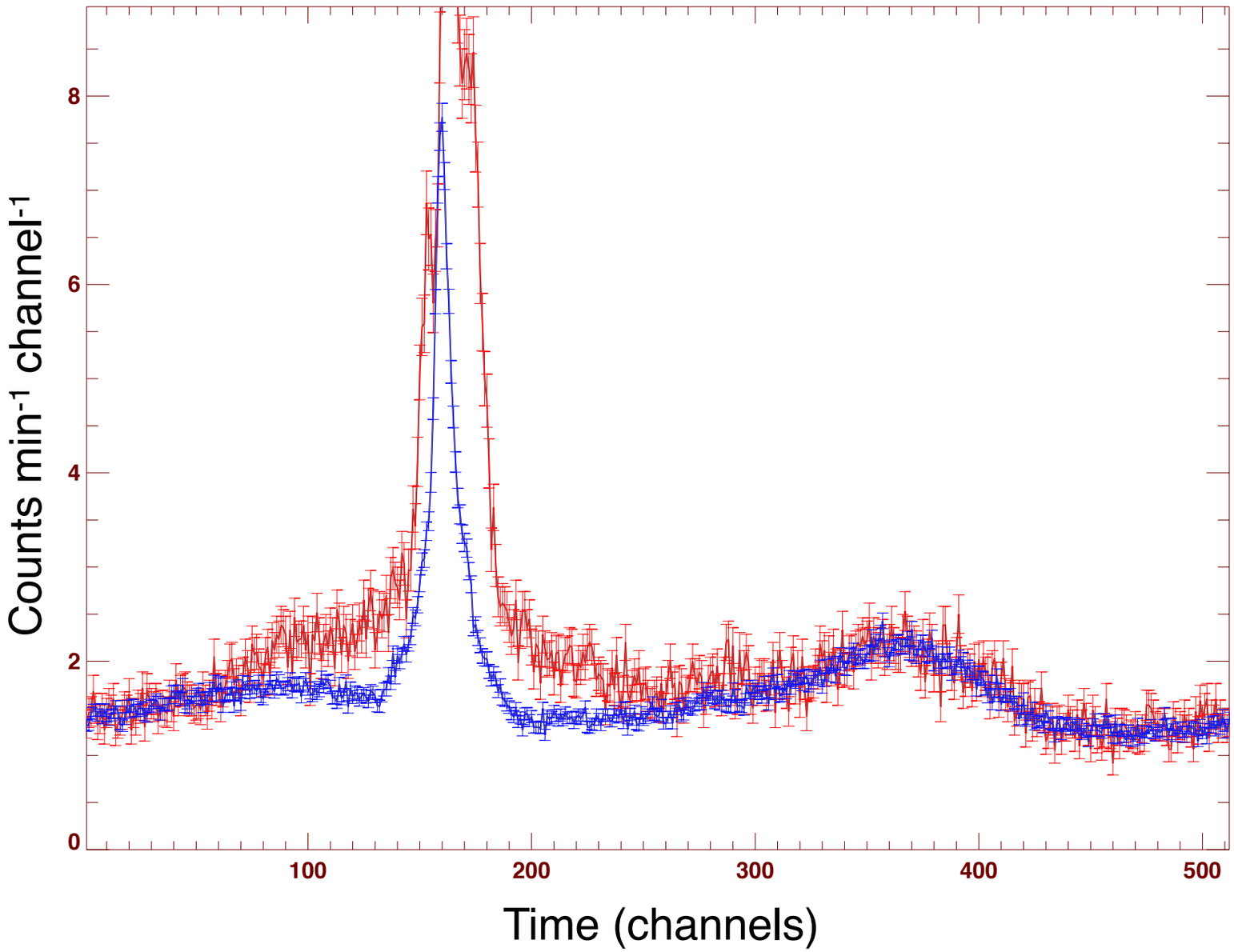
3) Reactor on: shutter open, nothing in beam



Scattering from windows, beam dumps and fast neutrons (thermalised in shielding). Can be time dependent on tof instruments

Case Study: IN4 (ILL)

4) Reactor on: shutter open, cryostat in beam

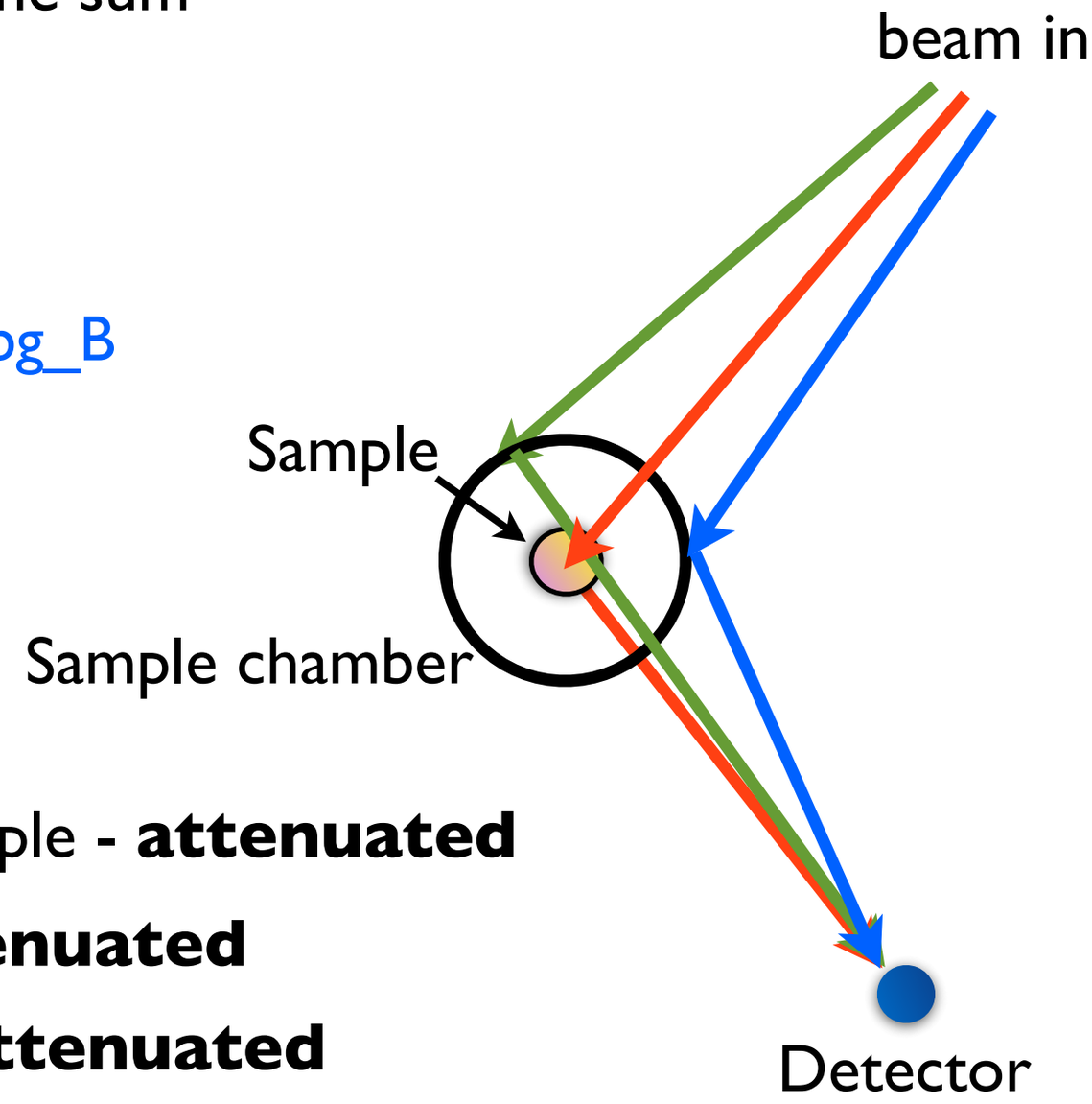


Scattering from cryostat tails, sample can (red). Will be time dependent on tof instruments

Background subtraction

Measured counts from the sample is the sum of three terms

$$N_{meas} = N_{sam} + N_{bg\_A} + N_{bg\_B}$$



N_{sam} = neutrons scattered from sample - **attenuated**

N_{bg_A} = background neutrons - **attenuated**

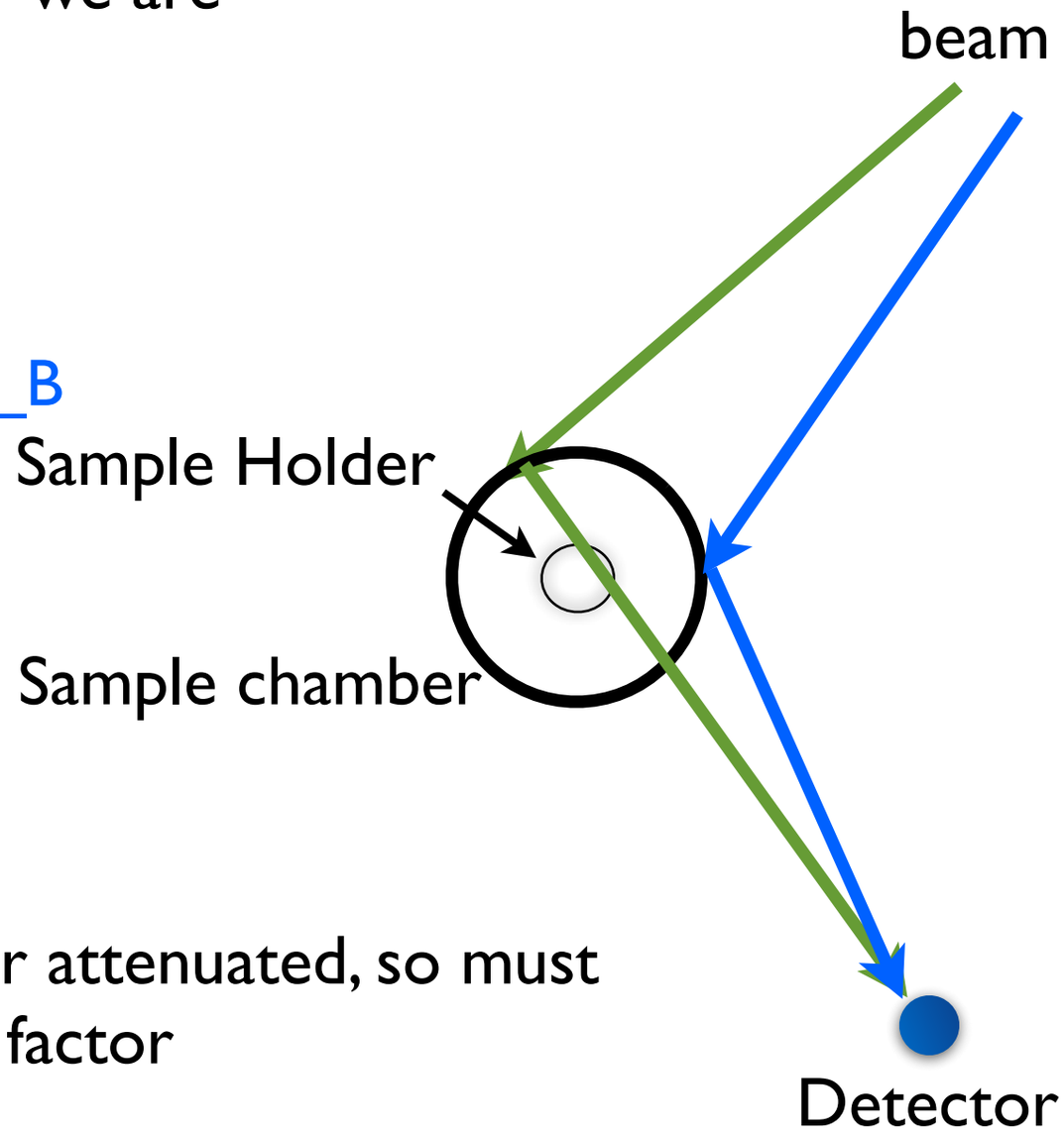
N_{bg_B} = background neutrons - **unattenuated**

Need to be able to separate the attenuated and unattenuated backgrounds for an accurate background subtraction

Background subtraction

If we measure an empty sample holder we are measuring

$$N_{empty} = (N_{bg\_A} / T) + N_{bg\_B}$$

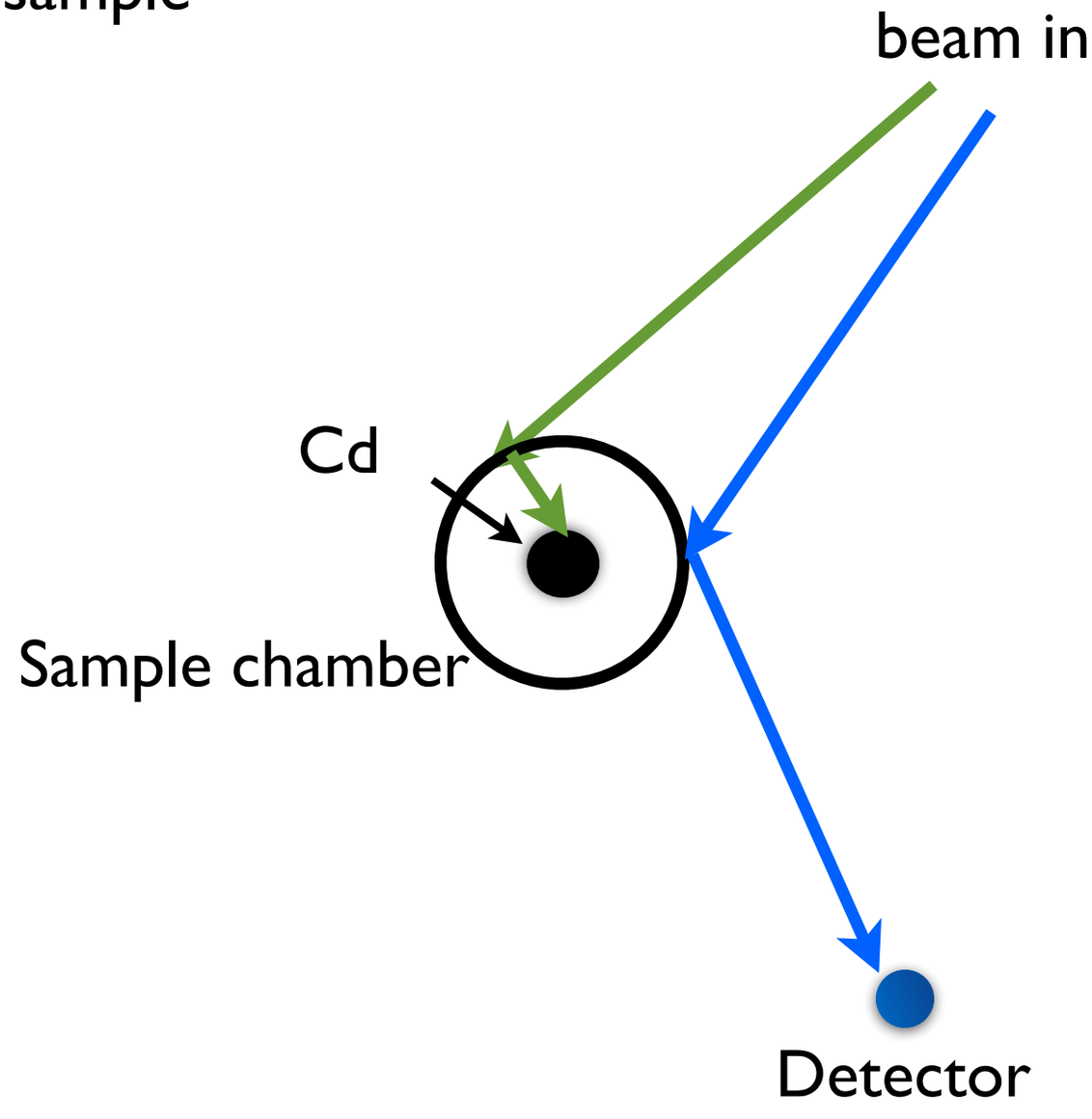


where the bg_A term is now no longer attenuated, so must be divided by the sample transmission factor

Background subtraction

If we measure a completely absorbing sample (such as cadmium) we are measuring

$$N_{Cd} = N_{bg\_B}$$



Background subtraction

Taking these together we have

$$N_{meas} = N_{sam} + N_{bg\_A} + N_{bg\_B}$$

$$N_{empty} = (N_{bg\_A} / T) + N_{bg\_B}$$

$$N_{Cd} = N_{bg\_B}$$

Combining these we get for the counts from the sample

$$N_{sam} = (N_{meas} - N_{Cd}) - T(N_{empty} - N_{Cd})$$

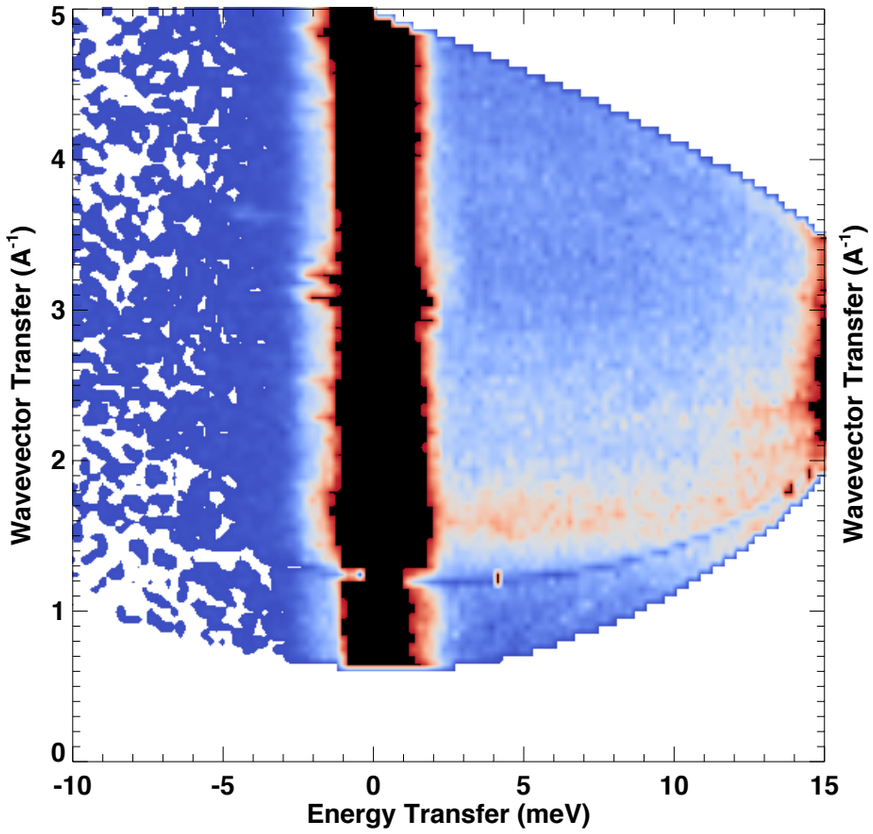
But to correct for sample attenutation, we need to divide by the transmission factor, therefore

$$N_{sam\_corr} = (N_{meas} - N_{Cd})/T - (N_{empty} - N_{Cd})$$

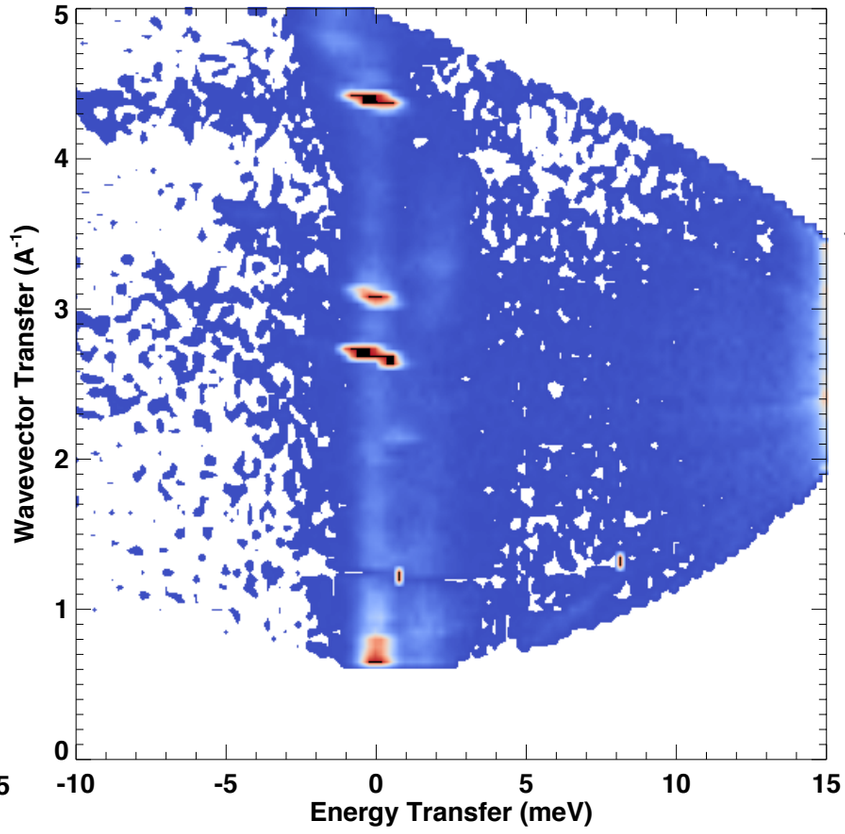
Things to note

- If the sample transmission is close to 1, then cadmium measurement not required
- N_{Cd} is often close to zero (e.g. SANS) -but still checked

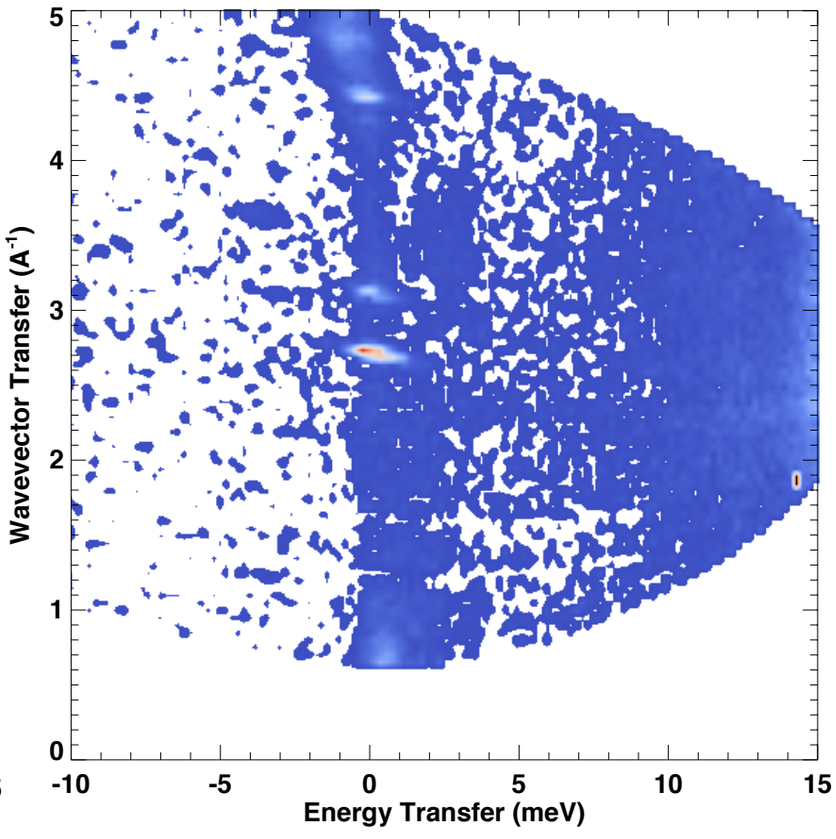
Sample (measured)



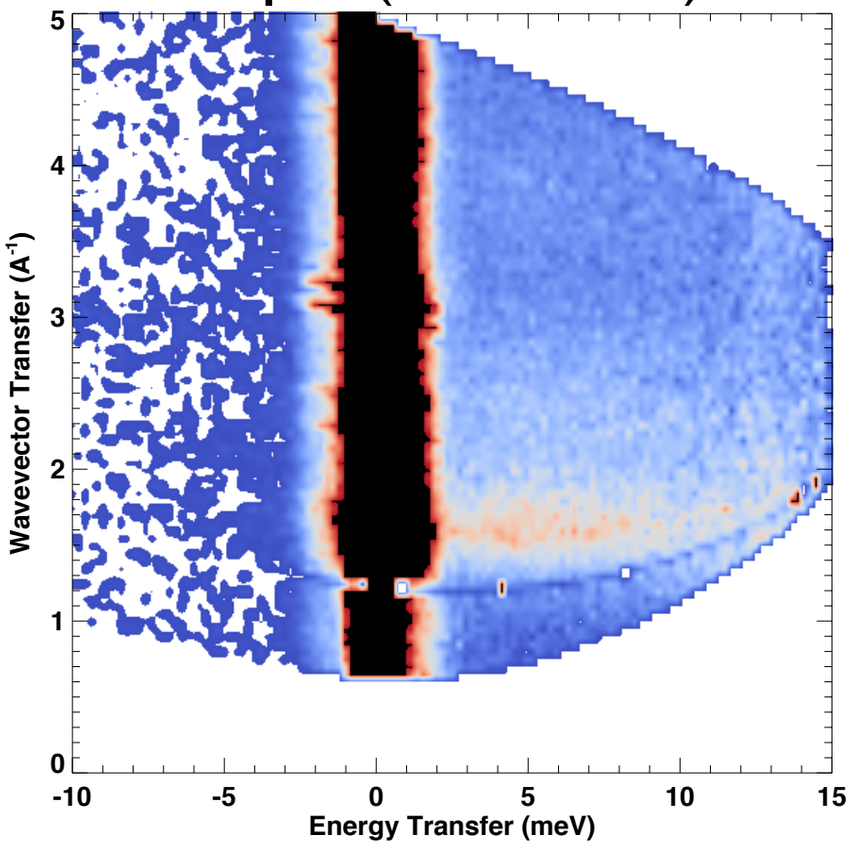
Empty



Cadmium



Sample (corrected)



Mn metal measured on IN4

Counting time

We need then to do 4 measurements to produce a corrected data set. (not counting experimental verification of the sample transmission)

- sample
- vanadium
- empty cell
- cadmium (or equivalent)

So the question now is how do we divide our time between these measurements?

This is an eternal question.... and opinion is divided.

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 !

Neutron fluxes are low, so it's important not to over-count our measurements. However - if we decide we need a certain level of error bar, we have to be prepared to "sit it out"

Counting time

The last question is - how much time do we have to "waste" measuring N_{empty}, N_{Cd} and N_v ?

Often N_v is already done by the instrument scientist (assuming sample size and shape doesn't alter very much)

If the backgrounds are close to zero - we don't need to count them at all
If they are large - they should be counted carefully.

The general "rule of thumb" is that the times spent on measuring sample and background runs should be in proportion to their count rates

$$\frac{t_{\mathrm{meas}}}{t_{\mathrm{empty}}} \simeq \frac{N_{\mathrm{meas}}}{N_{\mathrm{empty}}}$$

for the Cadmium run, the count time is modified by the transmission

$$\frac{t_{\mathrm{Cd}}}{t_{\mathrm{meas}}} \simeq \left(\frac{1-T}{T}\right) \frac{N_{\mathrm{Cd}}}{N_{\mathrm{meas}}}$$