tof-SANS Q resolution 25th March 2021

with some extra notes 12th April 2021

Some background, and thought provoking examples regarding Q resolution in time-of-flight SANS.

Other speakers today have considered, or will do so, aspects of Q resolution in much better detail than I have had time & opportunity to do!

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tof SANS data reduction in one slide!

$$Counts(R,\lambda) = I_0(\lambda)\frac{\partial\Sigma(Q)}{\partial\omega}\Omega(R)t.T(\lambda)\eta(\lambda)$$

Incident flux: $$I_0(\lambda) = \frac{M(\lambda)}{\eta_M(\lambda)}$$

Wavelength λ is proportional to arrival time at detector.
Need ratio of main detector efficiency compared to monitor.
e.g. Remove beam stop and put a small hole A_H at the sample to record:

$$D(\lambda) = \frac{C_H(\lambda)}{M_H(\lambda)} = \frac{\eta(\lambda)}{\eta_M(\lambda)}\frac{A_H}{A_S}$$

Rearranging:

$$I(Q) = \frac{\partial\Sigma(Q)}{\partial\Omega} = \frac{A_H}{A_S t}\frac{\sum_{R,\lambda\subset Q} C(R,\lambda)}{\sum_{R,\lambda\subset Q} M(\lambda)T(\lambda)D(\lambda)\Omega(R)}$$

R – radius on detector
t – sample thickness
T – transmission
η – detector efficiency

M – incident beam monitor
C – neutron counts
Ω – solid angle
A – beam area
$V_{sam} = A_s t$ – sample volume

D(λ) "direct beam" allows us to cross-normalise the incident spectrum to that empty beam seen on the main detector.

Numerator sums counts in a time and space "Q bin". There are some subtleties hidden here, including that we are combining data with varying Q resolution. Transmission T(λ) should include the SANS signal.

Proper statistics are *not* obtained by "averaging the reduced data from a series of wavelengths" .
P.A.Seeger & R.P.Hjelm J.Appl.Cryst. 24(1991)467-478

Q resolution

D.F.R.Mildner & J.M.Carpenter, J.Appl.Cryst. 17(1984)249-256.

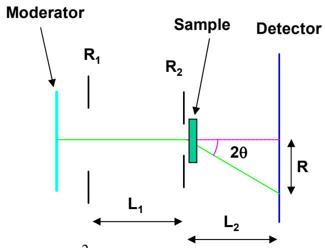
At a given Q, the radial σ_Q varies inversely with λ due to geometry,

e.g. for circular pinholes, isotropic scattering and assuming small angle approximations

With detector resolution of width ΔR at radius R

$$(\sigma_Q)^2 = \frac{1}{12}\left(\frac{2\pi}{\lambda}\right)^2\left[3\frac{R_1^2}{L_1^2} + 3\frac{R_2^2}{L'^2} + \frac{(\Delta R)^2}{L_2^2} + \frac{R^2}{L_2^2}\left(\frac{\Delta\lambda}{\lambda}\right)^2\right]$$

$$\frac{1}{L'} = \frac{1}{L_1} + \frac{1}{L_2} \qquad Q \approx \frac{2\pi\theta}{\lambda} \approx 2\pi\frac{R}{\lambda L_2}$$



$$\left(\frac{\sigma_Q}{Q}\right)^2 = \left(\frac{R_1 L_2}{2RL_1}\right)^2 + \left(\frac{R_2(L_1+L_2)}{2RL_1}\right)^2 + \frac{1}{12}\left(\frac{\Delta R}{R}\right)^2 + \frac{1}{12}\left(\frac{\Delta\lambda}{\lambda}\right)^2$$

"Optimal" SANS - best flux for best Q resolution: $L_1 = L_2$, $R_1 = 2R_2$

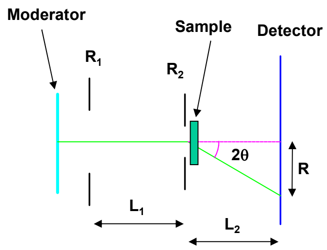
BUT this is a weak constraint, can get *almost* same resolution and count rate by adjusting aperture sizes if say $L_1 < L_2$, which increases band width in tof.

Notes

$$\frac{1}{L'} = \frac{1}{L_1} + \frac{1}{L_2} \qquad Q \approx \frac{2\pi\theta}{\lambda} \approx 2\pi\frac{R}{\lambda L_2}$$

$$(\sigma_Q)^2 = \frac{1}{12}\left(\frac{2\pi}{\lambda}\right)^2\left[3\frac{R_1^2}{L_1^2} + 3\frac{R_2^2}{L'^2} + \frac{(\Delta R)^2}{L_2^2} + \frac{R^2}{L_2^2}\left(\frac{\Delta\lambda}{\lambda}\right)^2\right]$$

$$\left(\frac{\sigma_Q}{Q}\right)^2 = \left(\frac{R_1 L_2}{2RL_1}\right)^2 + \left(\frac{R_2(L_1+L_2)}{2RL_1}\right)^2 + \frac{1}{12}\left(\frac{\Delta R}{R}\right)^2 + \frac{1}{12}\left(\frac{\Delta\lambda}{\lambda}\right)^2$$



This assumes that each distribution can be replaced by a Gaussian of the same standard deviation σ so that the convolution theorem can be used. $\sigma^2 = \sigma_a{}^2 + \sigma_b{}^2 + \sigma_c{}^2 + \sigma_d{}^2$

ΔR and $\Delta\lambda$ here are the width of a *rectangular* distribution, for which standard deviation σ is $\Delta/\sqrt{12} = \Delta/3.464$, hence the factors of 12 in the equation for σ^2, for other distribution shapes simply replace the σ^2,

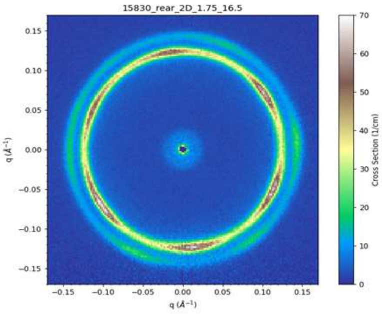
The FWHM, full width half maximum, of a Gaussian $f(x) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{x-\mu}{\sigma}\right)^2}$ is $(8\log_e(2))^{1/2}\sigma = 2.35482\sigma$

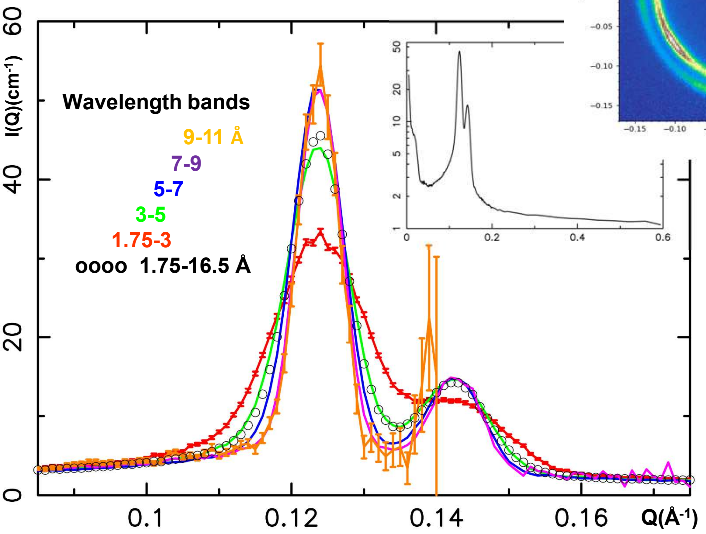
We need be clear whether we are talking about σ, FWHM or Δ.
(Personally I avoid using δx or ∂x or dx as they are ambiguous.)

e.g. Δx/x = 10% has σ = 2.9% with an equivalent Gaussian of fwhm = 6.8%

Tof SANS combines data from different radii and wavelengths, which may have a range of Q resolutions. Most often this matters little, here is one case where differences are apparent.

[Inverted hexagonal DOPE lipid (?) with silica particles on SANS2d, in 2012 W.Briscoe (Bristol), compare Bulpett et.al. Soft Matter 11(2015)8789-8800]





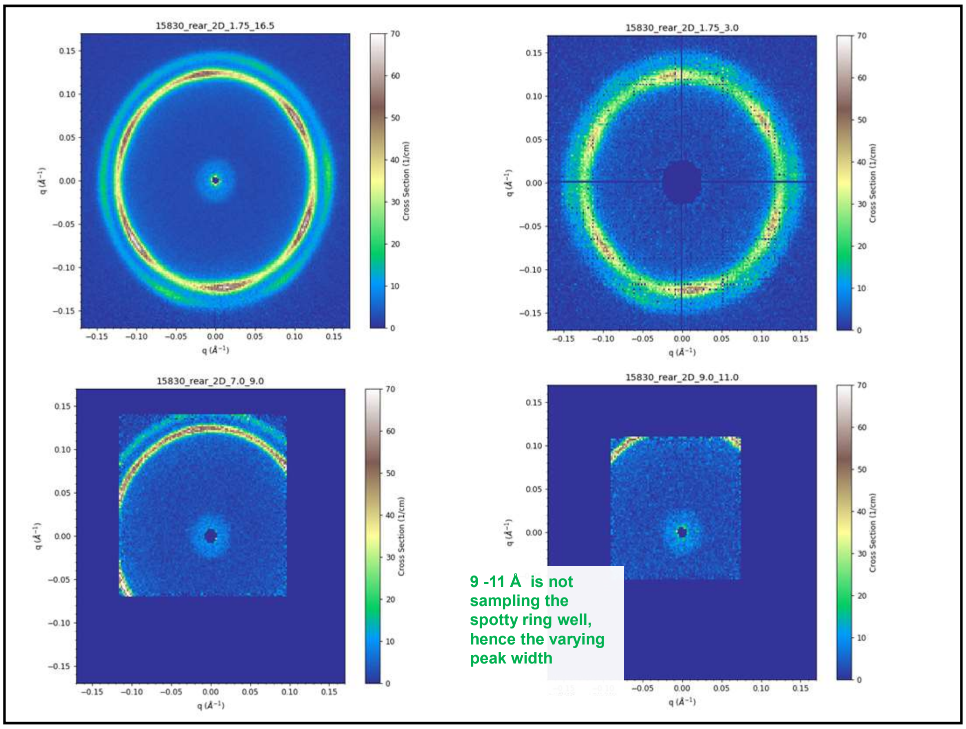
Fit the 2 peaks to Gaussians, on linear background

				FWHM % Resolution Mantid estimate		Fit smeared = fwhm(Bragg) ? Should be constant for each peak		Fit unsmeared	
Wav1	Wav2	Q1	Q2	M1	M2	1S	2S	1U	2U
1.75	3	0.1238	0.1440	8.6	7.6	10.3	5.8	13.4	9.6
3	5	0.1234	0.1424	6.2	5.6	6.7	5.4	9.1	7.8
9	11	0.1236		4.0		3.4		5.2	
1.75	16.5	0.1235	0.1423	5.8	5.6	6.1	5.5	8.6	8.1
Repeat reduction with 1% bins – see later									
9	11	0.1236		2.4		3.3		3.9	

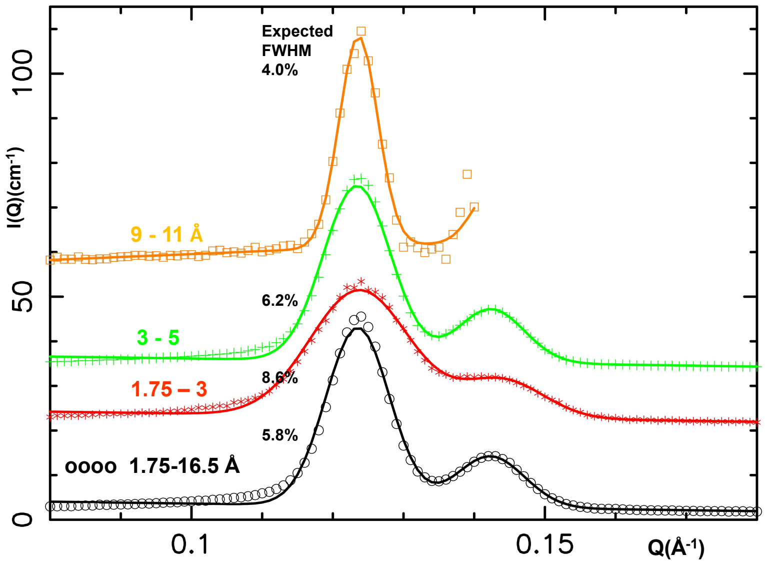
Mantid is averaging Mildner & Carpenter σ, weighted by number of neutrons at each wavelength.

Fit unsmeared minus smeared sqrt(U^2 - S^2) 1	2	from unsmeared fwhm(Bragg) sqrt(U^2 - M^2) 1	2
8.6	7.7	10.3	5.8
6.2	5.6	6.7	5.4
4.0		3.4	
6.1	5.9	6.4	5.8
2.1		3.1	

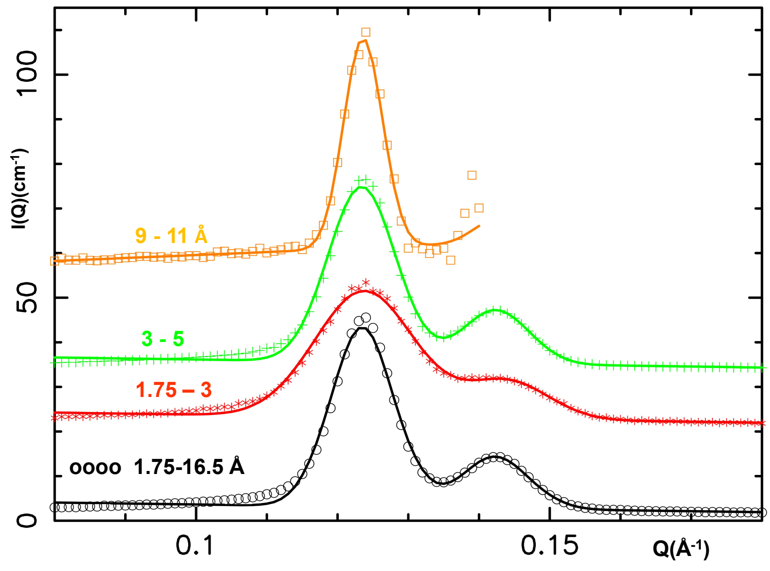
Checks here show sasview smearing is good!



Fit the 2 peaks to Gaussians, on linear background, I(Q) not smeared



Fit the 2 peaks to Gaussians, on linear background, I(Q) smeared by estimated "weighted mean" Q resolution



Why does σ_Q vary so much with wavelength?

$$\frac{1}{L'} = \frac{1}{L_1} + \frac{1}{L_2}$$

$$Q \approx \frac{2\pi\theta}{\lambda} \approx 2\pi\frac{R}{\lambda L_2}$$

$$(\sigma_Q)^2 = \frac{1}{12}\left(\frac{2\pi}{\lambda}\right)^2\left[3\frac{R_1^2}{L_1^2}+3\frac{R_2^2}{L'^2}+\frac{(\Delta R)^2}{L_2^2}+\frac{R^2}{L_2^2}\left(\frac{\Delta\lambda}{\lambda}\right)^2\right]$$

$$\left(\frac{\sigma_Q}{Q}\right)^2 = \left(\frac{R_1L_2}{2RL_1}\right)^2+\left(\frac{R_2(L_1+L_2)}{2RL_1}\right)^2+\frac{1}{12}\left(\frac{\Delta R}{R}\right)^2+\frac{1}{12}\left(\frac{\Delta\lambda}{\lambda}\right)^2$$

(a) Geometry, Q varies faster across the detector at short wavelengths

(b) which is another way of saying that the R_1/R, R_2/R and $\Delta R/R$ terms are larger at smaller radius

[and don't forget to allow for gravity at longer L_1 & L_2]

(c) The $\Delta\lambda/\lambda$ term may be larger at short wavelength, depending on the nature of the pulsed neutron source, and how long the beam line is. It may be very large for a long pulse source (i.e. ESS)

$$\left(\frac{\sigma_Q}{Q}\right)^2 = \left(\frac{R_1}{2R}\right)^2+\left(\frac{R_2}{R}\right)^2+\frac{1}{12}\left(\frac{\Delta R}{R}\right)^2+\frac{1}{12}\left(\frac{\Delta\lambda}{\lambda}\right)^2, \qquad \text{if } L_1 = L_2$$

$$\left(\frac{\sigma_Q}{Q}\right)^2 = 2\left(\frac{R_2}{R}\right)^2+\frac{1}{12}\left(\frac{\Delta R}{R}\right)^2+\frac{1}{12}\left(\frac{\Delta\lambda}{\lambda}\right)^2, \qquad \text{if } R_1 = 2R_2$$

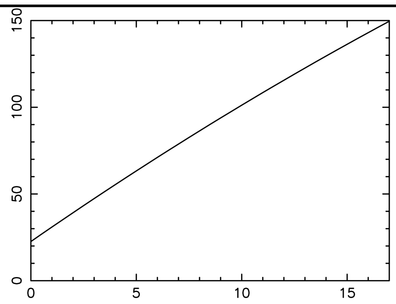
The $\Delta\lambda/\lambda$ term for pulsed sources

$\sigma_{moderator}$ [microsec] ISIS TS-2 coupled H_2

as measured on LET by R.Bewley

Values are extrapolated above 10 Å
– maybe they should level off ???

But ISIS TS-2 cold moderator is not quite fully moderated ?

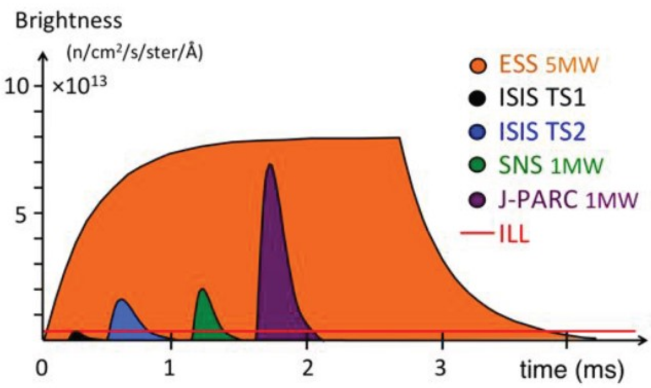


(i) For a short pulse accelerator based source, like ISIS, at any given wavelength there is a steep rise followed by an exponential tail in time due to moderation in the cold source. Thus what we see in tof at some *notional wavelength includes the tails of the distribution from slightly shorter wavelengths.* (In the extreme this may cause the resolution curve to broaden asymmetrically to higher Q.)
Note the standard deviation for exp{ - t/τ) is τ

(ii) For a chopped reactor source the wavelength distribution is usually rectangular.

(iii) In either case there may be minor effects due to the time it takes a relatively low speed chopper opening to cross the width of the beam.

(iv) The further we are from the source then Δt/t for a given λ reduces, so Δλ/λ reduces since λ [Å] ~ 3.956 time [msec] / L_{total} [m].
[Though resolution improves, the overall wavelength range and hence Q range decreases, so we compromise on the total length.]



(v) ESS is a "long pulse source", proton pulses are 2.86 msec

So $\sigma_{moderator}$ ~ Δ/ sqrt(12) ~ 3000 microsec/ 3.464 ~ 900 microsec

The Δλ/λ term for pulsed sources - more

(vi) ALSO may need to convolute one or more of:

Δλ due to data collection histogram time bin width (negligible in event mode) ,

Δλ due to rebinning to histogram in the data reduction.

$$(\Delta\lambda)^2 = (\Delta_{moderator})^2 + (\Delta_{collection})^2 + (\Delta_{rebin})^2$$

Aside: In the hexagonal phase example, it turned out that the data was collected in histogram mode with 0.75 msec bins (across the 100msec frame), and then the default was to rebin to Δλ/λ = 5% within Mantid. Repeat reduction with 1% bins, improved resolution.

Obviously the final Q bin size is also important, in the example this was 0.001 $Å^{-1}$ across the peaks, much smaller than "usual" for this part of Q range.

A further convolution for the final Q bin size ought to be included to allow for cases where the bin size dominates. Ideally the bin size should be say < σ/2 to give 4 or 5 bins across the FWHM, in which case this would have little effect.

Making the bin or rebin sizes too small has other consequences due to empty bins and "over-binning", strictly the resolution should be tracked separately from the bin step size.

Thus care is needed!

Postscript (added later)

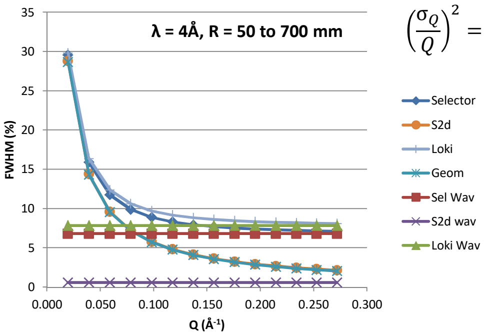
The simple average of σ_Q in the Mantid SANS reduction is currently weighted by the number of neutrons from each wavelength bin in that Q bin. However, as we saw in the example, a few long wavelength neutrons give a noticeably sharper peak (as the shorter wavelengths there are more smeared out). The current weighting gives only a tiny decrease in σ at the peak position, could try weighting as I(Q) instead???

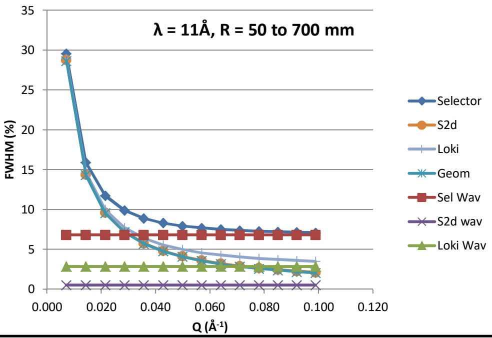
Note that the reduction software has no idea that the data "has a peak" !

The case of a "dip", such as in the form factor for monodisperse particles, is interesting since in the dip there are less neutrons and lower I(Q), so there seems no simple way to get a more representative mean for σ ? So will have to revert to throwing out the shorter wavelengths to improve resolution.

Though for convenience here resolution is described as FHWM(Q)/Q in %, Bragg peaks tend to be much the same width regardless of Q, so comparing FQHM(Q) or σ(Q) is actually better, else resolution at higher Q sounds better than it really is (especially for fixed wavelength, velocity selector, SANS).

Relative importance of the different terms





$$\left(\frac{\sigma_Q}{Q}\right)^2 = 2\left(\frac{R_2}{R}\right)^2 + \frac{1}{12}\left(\frac{\Delta R}{R}\right)^2 + \frac{1}{12}\left(\frac{\Delta\lambda}{\lambda}\right)^2, \quad \text{if } R_1 = 2R_2$$

Q resolution as FWHM %

Simple beam line
$L_1 = L_2$ = 4m
A_1 = 16mm, A_2 = 8mm
SANS2d at L_{total} =23m,
LoKI at 27.5m
ΔR = 8mm

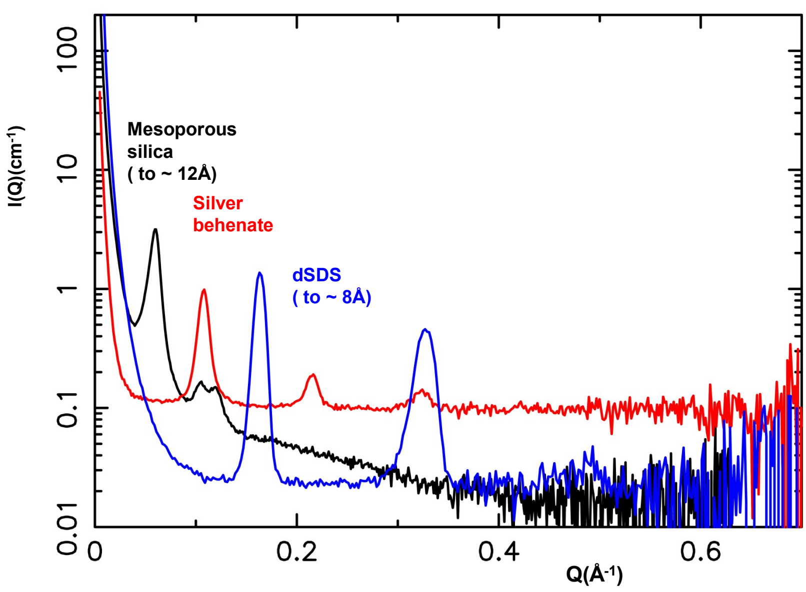
Δλ(selector) = 10%
SANS2d has σ_{mod} plus 0.5% rebin
Loki has Δt = 3.2msec

NOTE this *assumes* (a) we know scattering geometry to a few mm, else resolution may worsen when we combine wavelengths.

Especially so for higher angle banks out of the straight through beam.

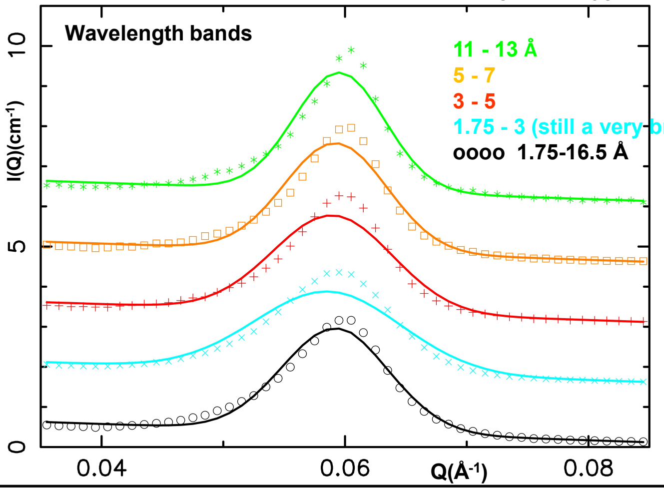
(b) we use the best value of λ for a given time of flight (not so obvious at a long pulse source)

Rotating powder samples L_2 = 4m on SANS2d rear detector, 1.75 – 16.5 Å, Oct 2020 (Najet Mahmoudi & SANS team)



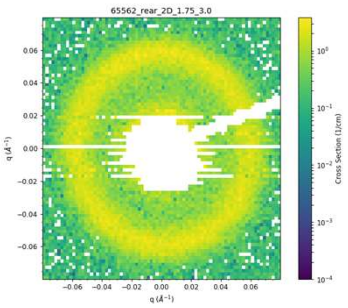
Better (?) example – mesoporous silica, peak at smaller Q, suits L_2 = 4m, but is broad compared to instrument resolution.
Perhaps not a Gaussian?
More detail needed near beam stop?

			resolution FWHM %	
			Mantid	Fit smeared = sigma(Bragg)
Wav1	Wav2	Qpeak	M1	?
1.75	3	0.0586	16.5	17.5
3	5	0.0590	7.4	16.3
5	7	0.0593	5.6	14.2
11	13	0.0595	3.8	15.5
1.75	16.5	0.0592	8.0	15.5

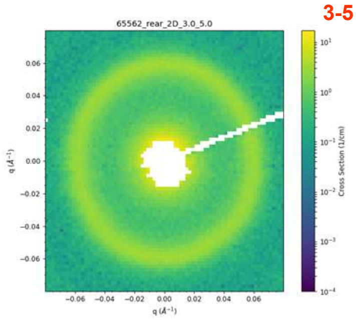


mesoporous silica (colour scale varies) – restricted Qmax

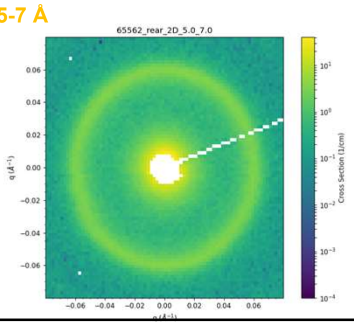
1.75-3 Å - VERY close to beam stop



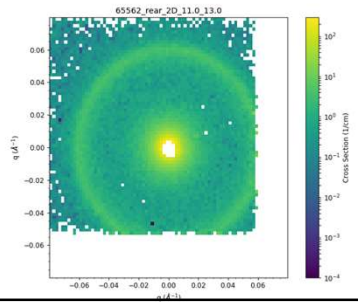
3-5 Å



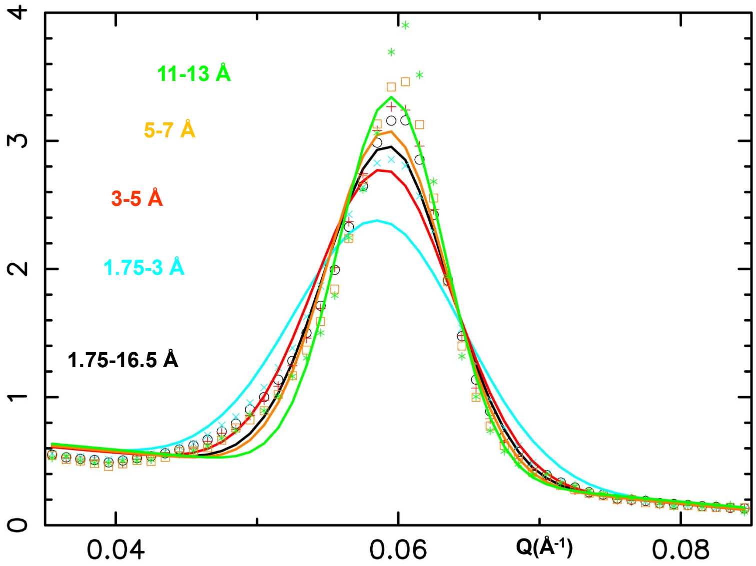
5-7 Å



11-13 Å



mesoporous silica



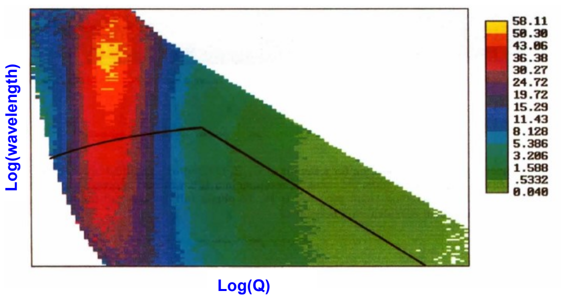
J. Appl. Cryst. (1991). **24**, 467–478

Remove short λ near beam stop - set criteria on resolution

Small-Angle Neutron Scattering at Pulsed Spallation Sources

By P. A. Seeger and R. P. Hjelm Jr

Manuel Lujan Jr Neutron Scattering Center, Los Alamos National Laboratory, Los Alamos, NM 87545, *USA*



0.6 x 0.6 m detector at 4.3m, λ ~ ?? to 15Å

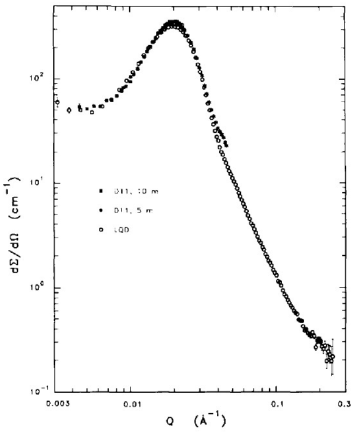
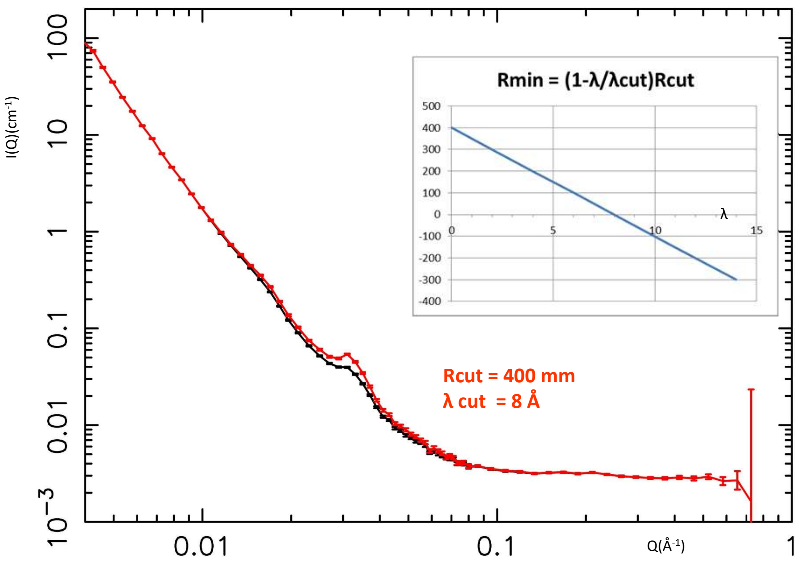


Fig. 7. Macroscopic differential cross section per unit volume for Vycor sample PT-5, as measured on LQD (○). Also shown are data from the same sample taken at the 10 m (■) and 5 m (*) detector positions on the D11 instrument at the Institut Laue–Langevin. From the quality of the data we judge the resolution to be equivalent to the 5 m position at D11. Pulsed-source instruments are advantageous when a large dynamic range of Q is required, since the entire range is collected with a single instrument setting.

Remove short λ near beam stop (SANS2d, SDS foam, 1.75 – 16.5 Å)

Could set criteria on resolution or use a more generic method as here:



tof Q resolution

- Combining data over a broad wavelength range gives a resolution curve with sharp peak but broad tails. Ideally need resolution curves stored with data. Use theory and/or simulation for the same set up *and* reduction scheme.
- In many cases a single parameter Gaussian seems not too bad, at least for a short pulse source.
- The code to average σ in Mantid could average estimated resolution curves, then output the whole curve or optionally say 3 or 4 "fit" parameters, starting with σ.
- In critical cases, take wavelength slices and fit them separately, or put in a resolution threshold in the reduction, at very least throw out short wavelength data near the beam stop.
- Take care that binning in the data collection and reduction (particularly geometry) do not degrade Q resolution, but bins must not be too small.
- Not considered 2d data, Q_x Q_y, resolution, suspect simulations are the way to go, again need an agreed method to store the information!
- Sharper Bragg peaks at lower Q would be good!