A High Resolution SNS Powder Diffractometer

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The HRPD proposed for the Rutherford SNS[1] is essentially Steichele's original design[2,3,4], as developed on a pulsed source by Windsor and Sinclair[5],but with the following features. For comparison with a conventional HRPD see also Fender[6] and Hewat[7].

Advantages over Steichele's Munich machine

i) Higher resolution, because with a 20 K moderator the pulse width is as low as 7λ μsec for $\lambda < 2.2$ Å, giving $\frac{\Delta t}{t} = 2.8 \times 10^{-4}$ at $\ell = 100$ metres. (Steichele has 100 μsec at 143 metres giving $\frac{\Delta t}{t} \sim \frac{3\times10^{-3}}{\lambda}$).

ii) Constant resolution for $\lambda < 2.2$ Å, since the neutron velocity is $\frac{4000}{\lambda}$ m.sec^{-1} (Steichele has $\frac{\Delta d}{d}$ increasing with $\frac{\sin\theta}{\lambda}$, whereas one would like it to decrease as on a conventional HRPD).

iii) A larger $\Delta\lambda$(0.8 Å) can be used without frame overlap, because of the shorter flight path. (Steichele must be limited to $\sim$0.5 Å). This means that sufficient data for most crystal structure problems could be collected with a single $\Delta\lambda$ slice.

iv) Of course, much higher intensity (a factor of $\sim 10^3$), eliminating the need for the very large samples ($\sim$100 cm^3) used at Munich, and making possible experiments on quite small (< 1 cm^3) hydrogenous samples of complex structure, dilute precipates in alloys etc.

Disadvantage

(i) The pulse shape from the SNS moderator has a rapid increase, corresponding to the time for thermalization, followed by an exponential decay as the moderator empties. This is less favourable than the clean triangular pulse produced by a chopper, since the exponential tails will add to form a background under closely spaced peaks. The effect can be minimized by optimum design of the moderator, and this is one of the main reasons for choosing a moderating temperature of 20 K. It does not seem feasible to use a pulse shaping chopper, since this would have to be within centimeters of the moderator if a wide wavelength band was to be shaped efficiently.

Design

a) Guide tube

i) We assume a guide of internal width d = 2.5 cm ; on a reactor there is an advantage in making it narrow, because of limitations on the pulse width chopper speed, but there is no problem with the slower wavelength and pulse suppression choppers, which are all that is needed on the SNS. The guide might be of square or even circular section, unless there is any chance of using a focussing guide.

ii) A radius of R = 35 km as at Munich gives a lower cut-off of $\lambda = 0.34$ Å. The critical cut-off angle ϕ_c is given by

$$\cos\phi_c = \frac{R-d}{R}$$

iii) The first 10 metres, containing the two choppers, and the final 40 metres, would be straight ; the central ℓ_c = 50 metres would be curved with a radius of 35 km to give a deflection δ from the straight through beam of 7.14 cm, since

$$\delta = \ell_c^2/R$$

This is sufficient to remove the γ-flash and epi-thermal neutrons.

iv) The need for expensive boron glass guides is not obvious ; the guide would of course be screened externally along its length.

b) Choppers

The basic pulse rate is 50 hertz, or 20 000 μsec between pulses. At a point ℓ from the moderator, the time delay between wavelengths separated by

$\Delta\lambda$ is

$$\Delta t = \frac{\ell \Delta\lambda}{4000}$$

since the neutron velocity is $\frac{4000}{\lambda}$ metre.sec^{-1}. The function of the first chopper is to pass a wavelength band $\Delta\lambda$ just so wide that there is no frame overlap at ℓ = 100 m. This gives a bandwidth $\Delta\lambda$ = 0.8 Å if Δt = 20 000 μsec (50 hertz). The bandwidth can be multiplied by a factor p if the fundamental pulse rate is divided by p, using a second chopper. Values of p = 1,2,3,4 are adequate, and for most purposes p = 1 (with the second chopper stopped) will be sufficient. It should be noted that the γ-flash will occur at times corresponding to measurements of λ = 0.8, 1.6, 2.4, 3.2 Å etc. A single one of these windows might be sufficient for most experiments, and then the counters might be "switched off" to avoid each γ-flash.

i) Wavelength Chopper

This chopper can conveniently be situated 8 m from the moderator, since then $\Delta\lambda$ = 10 Å for Δt = 20 000 μsec. The λ range of the instrument is then 0.34 to 10.34 Å. To produce from this a bandwidth of 0.8 Å, the wavelength chopper must open for Δt = 1600 μsec, and of course close for the remainder of the 20000 μsec.

The pulse shape for a simple chopper of window ω crossing the guide of diameter d is the truncated triangle shown in figure 2. For ~~neutrons of~~ chopper velocity v, the flat transmission time $(\omega-d)/v$ should be long compared to the rise or fall time d/v.

A cylindrical chopper of the type used by Steichele (diameter D = 68 cm) must turn at f = 25 hertz, since the beam may enter either window. Then $v = \pi D f$ and $\Delta t = 1600 \times 10^{-6} = \frac{\omega + d}{v} = \frac{\omega + d}{\pi D f}$ yields a window of 6 cm. The flat transmission region is then comparable with the rise or fall time, but could be improved by a larger chopper diameter D, or a smaller guide diameter d.

If a disk chopper were used instead, the frequency f could be doubled, since there need only be one window which could be twice as wide for the same diameter. However the absorption of such a relatively thin chopper must be sufficient to stop neutrons of $\lambda > 0.34$ Å. There is also the problem of stopping very slow neutrons ($\lambda + 10n$ Å) coming from the nth earlier pulse : these would be stopped by a cylindrical chopper, but not by a single disk chopper. A second phased disk, which could also serve as a pulse rate chopper, is an obvious solution.

ii) Pulse rate chopper

With cylindrical choppers cutting out very slow neutrons, the pulse rate chopper would not be required for many experiments ; the fundamental 50 hertz pulse rate yields a sufficiently large $\Delta\lambda = 0.8$ Å at 100 m without frame overlap. If $\Delta\lambda$ is multiplied by using a pulse divider, the γ-flash could not then be avoided, but with adequate shielding on a curved guide there should be no real problem. The pulse rate chopper should probably be identical to the wavelength chopper, so that even if one failed, the other could be used to trim the wavelength and the machine could continue to run normally at 50 hertz. This second chopper should immediately follow the first, with say 2 metres separation ; at 10 metres from the source the fundamental waveband $\Delta\lambda = 0.8$ Å occupies $\Delta t = 2000$ µsec, which is easily passed by a second identical chopper running at a fraction of the speed of the first (or stationary).

c) Counters

The net resolution of the instrument is the RMS sum of the temporal part $\frac{\Delta t}{t}$ and the angular part $\Delta\theta \cot\theta$, which should then be approximately equal

$$\frac{\Delta d}{d} = \left[\left(\frac{\Delta t}{t}\right)^2 + (\Delta\theta\cot\theta)^2\right]^{\frac{1}{2}}$$

Clearly, for small $\frac{\Delta t}{t}$ this near equality can only be achieved for large scattering angles θ, and then only if the resolution $\Delta\theta$ of the counter system varies inversely as $\cot\theta$. Table 1 shows how the spatial resolution Δy of a spherical counter array centred on a sample 1 m away should change with the distance y from the beam axis.

(i) High angle counters

According to table 1, 1 cm counters could be used to fill the area up to 10 cm from the beam axis ; however such an array would subtend an angle of only 0.25 % of 4Π , and as well, part of this area must be cut away for the incident beam. For many problems, even this small solid angle might be sufficient with such a high flux source ($\sim 10^{16}$ $n.cm^{-2}.sec^{-1}$). For example, it would be about 5 times the solid angle subtended by the 10 x 10' collimators on D1A, except that on a conventional guide tube diffractometer, a factor of at least 5 can be recovered by focusing the incident beam. Furthermore a multidetector on a conventional guide tube machine can accept at least 0.75 % of 4Π (D1B). The full potential of the TOF machine can then only be realized using a large multi-detector with radial resolution increasing towards the perimeter, as shown in fig. 3. Angular resolution would also be useful for the study of textures, which will not normally be isotropic. A flat detector , with an electronic time delay as a function of radius, seems more practical than a detector curved to give time focusing (e.g. as the counters are arranged on the Munich machine). As well, with a flat detector the sample position could be moved along the beam to permit an easy and rapid change of resolution.(Clearly the pulse time width cannot be changed, as on the Munich machine, to relax the resolution). In the standard high resolution geometry, a counter of diameter 1 m and 1 m from the sample will subtend a solid angle of 6.25 % of 4Π , almost an order of magnitude better than on a conventional diffractometer such as D1B.

ii) Low angle counters

The most important low angle counter will be the low efficiency monitor directly behind the sample. This counter is needed, not only to measure the incident spectrum, but more importantly the spectrum transmitted by the sample. This latter spectrum will contain a sharp Bragg cut-off for each reflected wavelength, so that the back of the sample does not see the same spectrum as the front. Unless corrected for , this gives a "secondary extinction" effect for the TOF machine which does not exist for the conventional diffractometer (e.g. Hewat 1975).

A second type of low angle counter is needed to collect low resolution data at small values of $\frac{\sin\theta}{\lambda}$, especially since primary extinction may become important for the very long wavelengths needed in the backscattering geometry. However, there are very few experiments for which both high resolution backscattered data and low resolution forward scattered data are needed. Even then, the data sets could be collected sequentially. Then the same multidetector could be used in the forward direction : detector and sample might be mounted on rails, and the counter simply lifted and turned, such as on D11 at the ILL. In fact such an arrangement would make an interesting small angle scattering option available on the diffractometer.

iii) 90° counters

The main interest of detectors at $2\theta=90°$ would be for avoiding scattering from the sample surroundings -air, cryostats, furnaces etc. The incident and diffracted beams, if orthogonal, may define a very small scattering volume. However, backscattering would still be superior in at least some situations ; for example, a pressure cell or cryostat might be made with only two small single crystal windows.

The chief difficulty of using 90° scattering is that the resolution $\Delta\theta \cos\theta$ is not good, and the density in θ of reflexions is a maximum (Hewat 1975) -a conventional diffractometer is therefore arranged to have maximum resolution for $2\theta \gtrsim 90°$. In fact, conventional söller collimator/ counters might be a good choice for those (few ?) experiments where 90° scattering might be needed. A multidetector ring would automatically throw away the main advantage of 90° scattering, since it would accept scattering from a large sample volume.

Comparison with the Proposed High Flux SNS Diffractometer

The only way that one can obtain higher flux from the machine described above is to increase either the incident or scattered solid angles : clearly the pulse width cannot be relaxed as on the Munich machine. With a multidetector, the scattered solid angle is already very large, but in principle the incident solid angle can be increased by greatly reducing the flight path length of 100 m. This is the basis of the proposal for the high intensity SNS powder diffractometer[1).

However, the closest approach to the moderator is limited by the shielding and the space required for choppers and counters. The current proposal[1) calls for a flight path of 12 m. Then the angle subtended at the sample by a moderator of 10 cm is 28.7'. Since the measured divergence of a nickel guide is $\alpha_v = 0.015\ \lambda/2\Pi$, a guide tube machine will give greater incident divergence for $\lambda > 3.59$ Å. It follows that apart from losses due to guide tube imperfections, the SNS high resolution machine should also have higher intensity for $\lambda > 3.59$ Å or $d = 1.8$ Å for backscattering. In fact $d \sim 1.5$ Å is just the region of most interest for chemical kinetics type experiments, where very high flux is most needed.

The HRPD has a fundamental advantage over the 12 metre machine for high intensity measurements. The latter machine has a bandwidth of 6.7 Å with no frame overlap. However, it is difficult to see when such a large bandwidth would really be needed : usually for chemical kinetics, a bandwidth smaller even than the 0.8 Å of the HRPD is adequate. The critical point is then the following : on the 12 metre machine, the neutrons in a bandwidth of 0.8 Å all arrive within 2,400 μsec, whereas on the 100 metre machine, these same neutrons are spread out over the entire cycle of 20,000 μsec. Then for the same useful neutron intensity, the counter on the 12 metre machine must be almost an order of magnitude faster to avoid dead time counting losses.

It should be remembered that dead time losses are already a problem for chemical kinetics experiments with large multidetectors on the ILL reactor. They originate from the electronic logic rather than from the multidetector itself, and the difficulties are proportional to the number of channels. On the SNS machines, these difficulties will be compounded because time channels are needed as well as space channels, and because all of the interesting neutrons arrive together on short flight path machines. Only on the 100 m HRPD will the useful neutron intensity be averaged out over time in a reasonable manner.

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References

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Table 1

y_{mm}	Δy_{mm}
50	22.4
100	11.2
200	5.6
500	2.2
1000	1.0

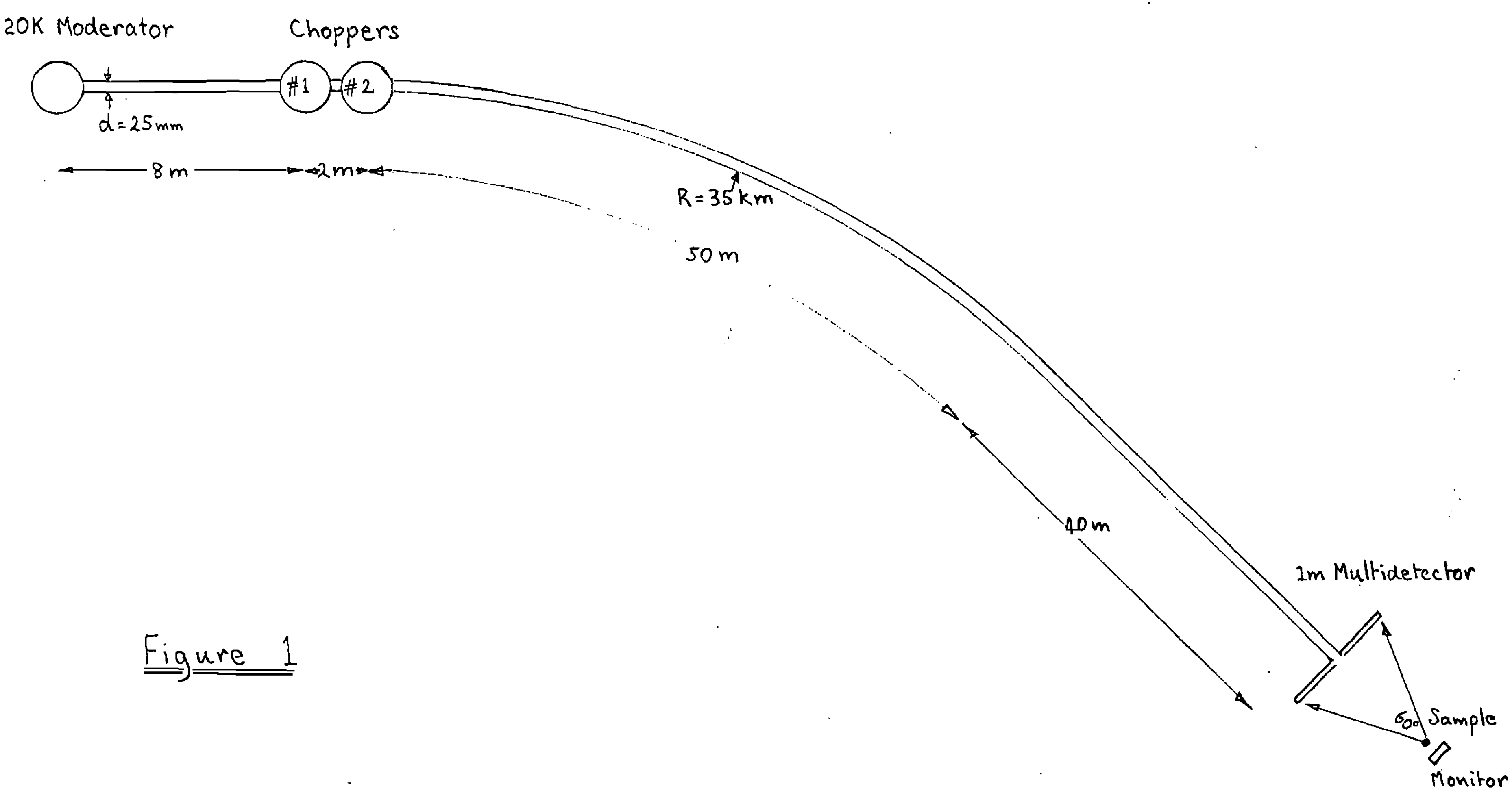
The radial resolution Δy required of a backscattering counter system as a function of the distance y from the beam axis. A multidetector of radius 0.5 metre and maximum resolution 2.2 mm seems practical ; this would subtend a solid angle of 6.25 % of 4Π at the sample 1 metre away i.e. an order of magnitude greater than a multidetector on a conventional powder diffractometer (D1B).

Figure Captions

Figure 1 Basic lay out of the proposed SNS high resolution powder diffractometer.

Figure 2 Pulse shape passed by a window ω chopping a beam of width d. This corresponds with the λ transmission of the 1st chopper, and should be as square as possible.

Figure 3 Schematic design for the 1 m diameter multidetector, showing the radial resolution increasing towards the circumference, with constant angular resolution.



20K Moderator
Choppers
#1
#2
d=25mm
8 m
2m
R=35 km
50 m
40 m
1m Multidetector
60°
Sample
Monitor

Figure 1

Figure 2

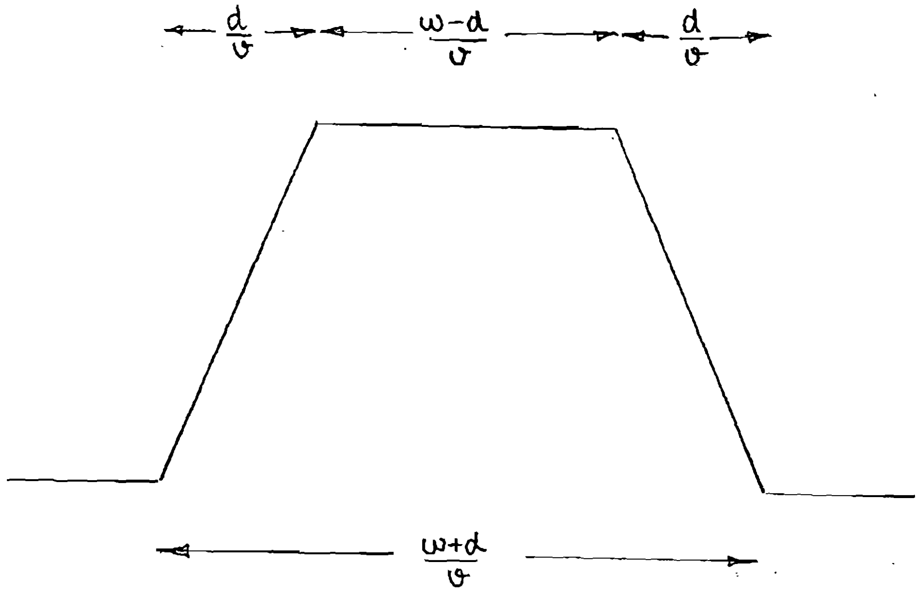


Figure 3