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Collimation and background reduction design for the time of flight neutron reflectometer SPEAR at *LANSCE*

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ABSTRACT

The effectiveness of neutron reflection measurements as a probe of surface and interface characteristics depends on the dynamic range of wave vector transfers over which the reflectivity can be measured, and upon the resolution possible within that range. We discuss some aspects of the design and our experience of the performance of the Surface ProfilE Analysis Reflectometer SPEAR at *LANSCE* with respect to collimation, wave vector transfer range and resolution and suppression of background.

I. INTRODUCTION

The reflection coefficient for neutrons (illustrated schematically in figure 1) is related to the profile of the neutron scattering length density, β, normal to the reflecting surface. For low reflectivities the relation reduces to the relatively simple form [1,2]:

$$R \approx \frac{[4\pi]^2}{Q_R^4} \left| \int_{-\infty}^{\infty} \frac{d\beta}{dz} \exp[iQ_R z]\, dz \right|^2 \qquad (1)$$

In which the reflectivity is proportional to the modulus squared of the Fourier transform of the gradient of the β profile and falls off as the fourth power of the reflection wave vector transfer perpendicular to the reflecting surface or interface. $Q_R = 4\pi\sin\theta/\lambda$, where θ is the angle of incidence and λ is the

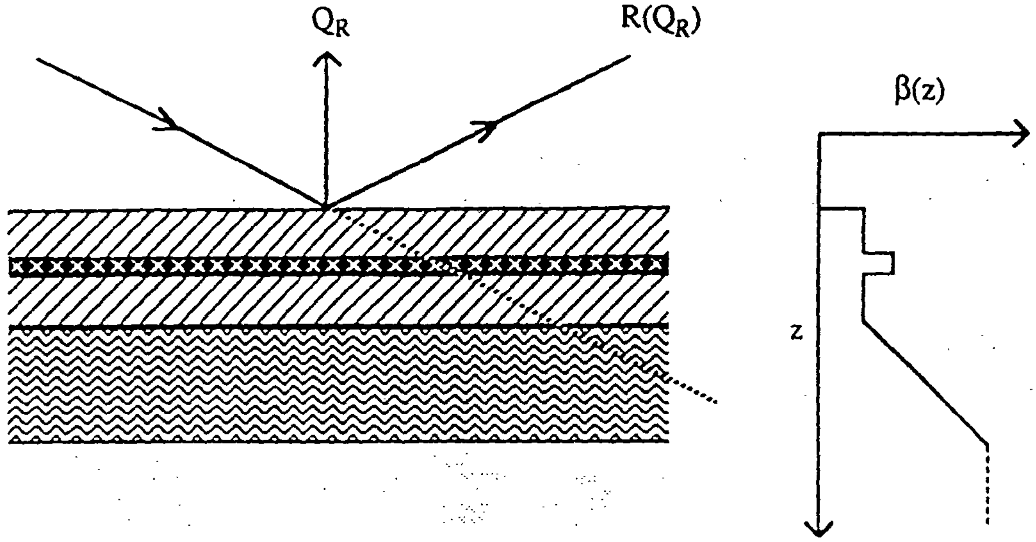

Figure 1. A schematic illustration of a reflectivity measurement. We wish to infer the scattering length density profile $\beta(z)$ from measurements of the reflectivity R as a function of the scattering vector Q_R.

neutron wavelength. Neutron reflectivity experiments measure the specular reflection as a function of Q_R, yielding information about the composition and density gradients at surfaces and interfaces.

As equation (1) makes clear, the information available from a neutron reflection experiment depends largely the dynamic range in Q_R over which the reflectivity can be measured. The maximum value of Q_R essentially determines the depth resolution of the technique. Firstly, this will be limited by available neutron wavelengths and scattering angles. Secondly, since reflectivity falls so rapidly with increasing scattering vector, instrumental backgrounds form various sources can combine to limit the minimum reflectivity, and hence maximum Q_R. Finally, the ultimate limits to the useful Q_R range are determined by sample dependent effects, such as incoherent scattering from the bulk [3].

II. SPEAR at *LANSCE*

On November 2-3, 1987, a workshop was held at the Los Alamos National Laboratory to discuuss the design criteria for a time of flight reflectometer based on the spallation neutron source at the Manuel Lujan, Jr Neutron Scattering Center (*LANSCE*) [4]. A feature that was overwhelmingly supported at the meeting was a vertical scattering plane, which allows reflection measurements from unconstrained liquid surfaces. Given the design restrictions associated with necessarily horizontal samples a number of measures were suggested to maximize the accessible range of Q_R. It was decided that the instrument should view a cold moderator to make available a large a range of neutron wavelengths as possible. Also, since backgrounds associated with spallation neutron production are intense, tight collimation of the neutron beam within the bulk biological shield was advised. Since the level of shielding required necessarily fixes the incident angle upon a liquid surface, it was further decided that , the instrument should provide two independently operable beams at different angles to the horizontal. Thus allowing a choice of high and low angles of incidence and a consequent flexibility in Q_R ranges depending on the reflecting strength of samples.

The configuration of SPEAR as built on flight path 9 at *LANSCE* [5] is shown in Figure 2. The instrument views the liquid hydrogen moderator. Maintained at 20K, produces observable neutron flux to about 50 Å, well beyond the range necessary for reflection experiments. The moderated neutrons are collimated into two beams within the LANSCE target's bulk biological shield. These beams are inclined downward at angles of 1.5° and 1.0° to the horizontal and converge at a common sample position 8.73 m downstream from the moderator. A specially designed mercury shutter allows the beams to be operated either independently or simultaneously. As defined by the in-shield collimation the vertical angular full width of neutron illumination at the sample position of the beams is 0.15° and 0.1°, 10% of the nominal angles of incidence on a horizontal sample, with a horizontal divergence of 0.25°. The standard detector position is a further 3.65 m downstream from the sample. The moderator to detector distance of the instrument is therefore 12.38 m. At the *LANSCE* source's 20 Hz repetition rate this leads to a first frame overlap wavelength of 16.0 Å.

III.T_0 FLASH AND FRAME OVERLAP BACKGROUND SUPPRESSION

Equipment in the beam handling area between the bulk shield and the sample position is used to further tailor the beams' wavelength composition and horizontal collimation, and to suppress background contributions.

The first component encountered in this region is a T_0 chopper, located in a heavily shielded cave just outside shutter exit, about 4.5 m from the moderator. This device rotates to bring a 300 mm Nickel alloy (Inconel) slug into the beam paths during the initial flash of high energy neutrons and gamma rays from the spallation target, blocking them from the instrument cave proper. Altering the rotation direction and phasing of the chopper with respect to the neutron pulse allows the chopper to be opened as soon as desired after the flash for either beam. Typically we set the chopper to open at about 3 ms, admitting neutrons with wavelengths greater than 1 Å.

The fast neutrons eliminated by this chopper are a particularly significant background at short times, since they can be moderated in the instrument cave shielding and scatter into the detector arriving at the same time as neutrons reflected from the sample, overwhelming a weak reflection signal from short wavelength neutrons at high wave vector transfers. Our measurements of cave backgrounds show a background reduction by about three orders of magnitude when this chopper is in operation.

A further advantage of this chopper is that is enables direct measurement of the incident illumination upon the sample at short wavelengths using the same detector as we use for reflection measurements. Without the chopper it proved impossible to shield a normal ^{3}He tube detector sufficiently to prevent saturation due to the flash, from which the detector took 5 to 10 ms to recover, without blocking the moderated neutron signal altogether. This made a direct incident beam spectrum

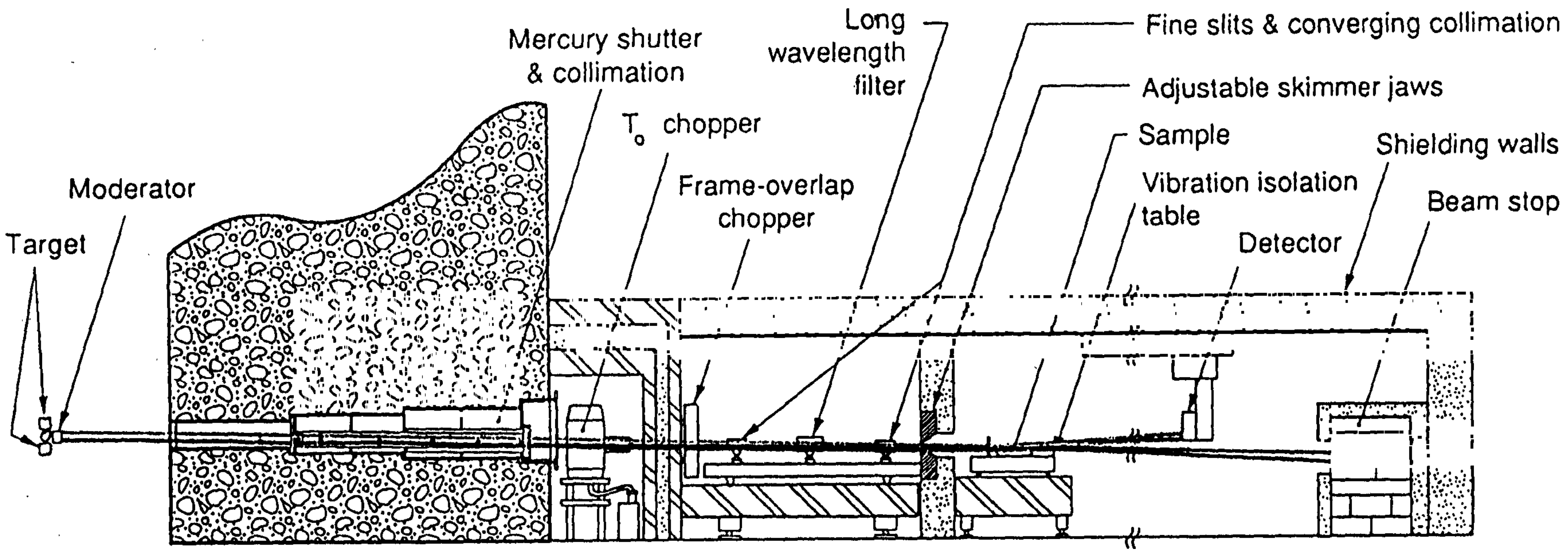

Figure 2 Physical layout of the Surface ProfilE Analysis Reflectometer SPEAR at *LANSCE*. The operation of the moderator, bulk shield collimation system, the T_0 chopper, the frame overlap chopper and mirrors, and detector are discussed in the text.

impossible for wavelengths below about 3 Å, and this region had to be measured with a less sensitive monitor detector. This introduced errors due to efficiency corrections between detectors into our determination of the reflection coefficient in this region. With the frame overlap chopper even our rather more fragile linear position sensitive detector can be used in the direct beam path with only cadmium slits in the beam handling area and at the sample position to reduce the neutron signal to suitable rates. Measurement of the incident illumination over the range 1 to 3 Å, increases our the reliably normalizable dynamic range range in Q_R at a given angle of incidence by a factor of three.

The neutron beams leave the enter the instrument cave proper from the T_0 chopper cave passing through narrow channels in the heavy shielding wall, and encounter a frame chopper mounted on the far side of that wall at the midpoint of the beam line (6.19 m). This chopper consist of a lightweight neutron absorbing disc (Boral backed by Cadmium) with two opposing 90° segments removed. Rotating at one half of the source repetition rate this chopper admits neutrons from either 0-16 Å or 16-32 Å depending on its direction of rotation. This has two effects. Primarily, this increases by a factor of two the range of Q_R accessible in measurements at a single angle, and allows us to measure reflectivities from a horizontal sample down to wave vector transfers of 0.007 $Å^{-1}$ using the 1.0° beam. It is also reduces frame overlap contamination of the reflected beam signal. Frame overlap contamination is a critical problem for time of flight reflection measurements since the falloff in intensity at longer wavelengths is offset to a large extent by the rise in reflectivity (at least until total reflection is reached at the critical value of Q_R). While this has a favorable consequence in that it evens out count rates and hence statistics across Q_R range of a reflection measurement, it also allows relatively few slow neutrons from a previous pulse reflected since they reflect very strongly from the sample to result in a signal comparable to that from the weak reflection of many more faster neutrons from the current pulse, when they arrive at the detector at the same time (1/v cross section contributions to detector efficiencies also work to exacerbate this problem). The frame overlap chopper acts as our first line of defence against this effect, blocking cross talk between the reflection signals the neutrons from two time frames.

As it happens, the hydrogen moderator is efficient enough that the 1-16 Å reflection signal can also be affected by frame overlap contamination from at least the beginnings of the 32-48 Å wavelength band admitted at the same time by the chopper. The simplest way to eliminate this final long wavelength contamination is to take advantage of their high reflecivity to filtering them out of the beam. We use a set of Nickel film on Silicon mirrors set at a relatively high angle (~3°) such that neutrons beyond 32 Å are totally reflected out of the beam onto the sample.

IV. BULK SHIELD COLLIMATION AND INSTRUMENT RESOLUTION

The choice of a vertical scattering plane and the use of neutrons of wavelengths of tens of Å has consequences not usually encountered in conventional neutron scattering. Our collimation, to a first approximation a geometric optics system of the simplest kind, has had to be designed to take into account gravitational "aberration" effects which become significant in our 16-32 Å frame.

Since we deal with such small angles of incidence ($\theta < 2^\circ$) the acceptance of most samples is small, of order a millimeter or so. Effectively acting as a narrow slit, this strongly defines the collimation of the neutrons In order to reduce background in the instrument we decided to do as much initial collimation as possible within the bulk shield. In the absence of gravitational affects neutrons the small acceptance of the sample would (with slight corrections for non-zero sample acceptances) allow us to define the angular spread ($\Delta\theta$) of the neutron beam upon the sample with a system of collimating slits converging to the sample position. A schematic of the actual situation is shown in Figure 3. Slow neutrons will actually fall appreciably away from a line of sight path in the time it takes them to travel from the moderator to the sample - a 32 Å neutron travels at 124 ms^{-1} and would fall 24 mm below the sample. To reach the sample slower neutrons need to start out at an angle above the direct path and from lower down on the moderator and these paths are blocked by line of sight collimation. This will cause us to lose illumination at longer wavelengths and will also cause the collimation angle to vary drastically with angle becoming tighter as a greater fraction of the paths are blocked eventually cutting off altogether. (This collimation variation is the main reason that this "gravitational filtering" of long wavelength neutrons was rejected as a solution to the 32-48 Å frame overlap contamination, in favour of the Nickel on Silicon reflection filters mentioned in the previous section.) To allow such paths the

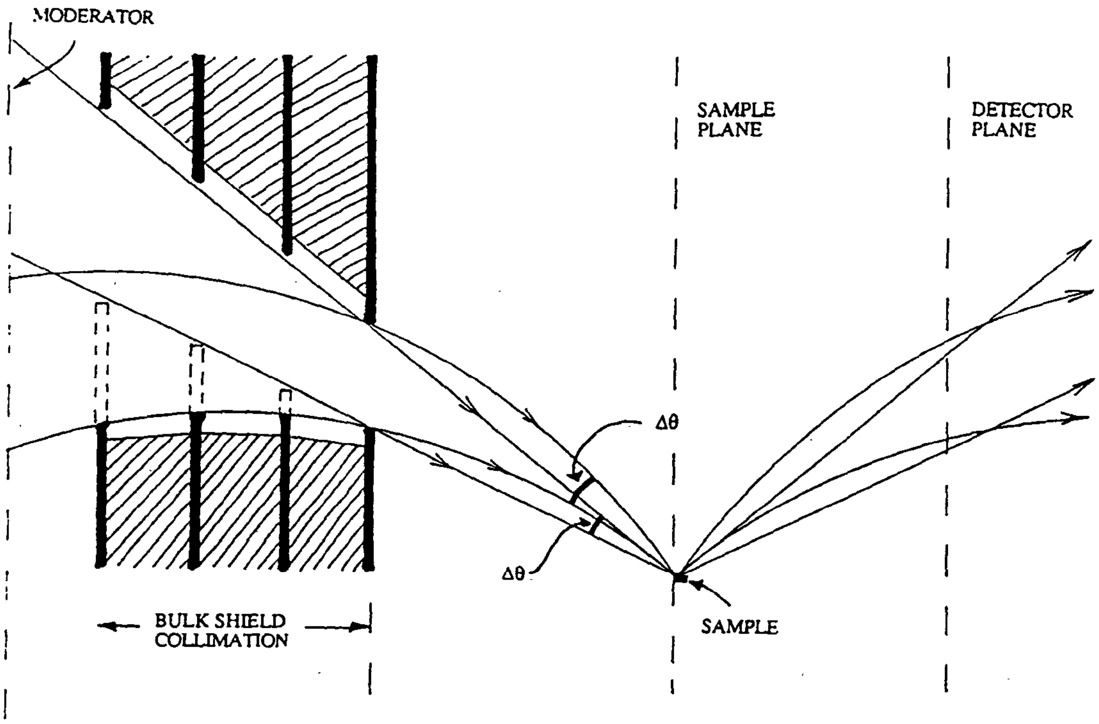

Figure 3. Gravitational effects on slow neutron trajectories (the vertical scale is exaggerated), showing the necessary breaking away of the bulk shield collimation elements to allow longer wavelength neutron to reach the sample position.

lower half of the collimation system has to break away from the line of sight path as it nears the moderator. At the entrance to the our collimation about 1 m from the moderator the slit edges of the collimation system have to be 7 mm below the fast neutron line of sight paths to allow a full angular spread, $\Delta\theta$, of 32 Å neutrons to reach the sample unimpeded.

One obvious consequence of the noticeably parabolic trajectories followed by longer wavelength neutrons is that they are incident upon the the sample at a higher angle of incidence than the line of sight collimation. At the 4.47 m separation between the exit slit of our collimation system and our sample position the center of the incident angle distribution is higher than the nominal line of sight value up by 0.08°, representing a +5% shift in Q_R for the 1.5° beam incident upon a horizontal sample. Another consequence of this higher angle of incidence is that when the slower reflected neutrons leave the sample they are initially aimed higher on the detector plane than the nominal straight line reflection trajectory. However, en route to the detector they will fall back towards and eventually cross its level as far from the sample as the sample is from the collimator exit (independent, to first order, of the angle of incidence upon the sample). Thus, if the collimation exit to sample distance were the same as the sample to detector distance, the reflected beam position on the detector plane would be essentially independent of wavelength. On SPEAR this was not quite realized, the distances are 4.47 m and 3.65 m respectively, resulting in a slight shift upward over the nominal postion on the detector of 1 mm for the reflection of 32 Å neutrons. Both these effects are easily dealt with in software when the reflection signal is normalized to the incident spectrum (as are related effects: a slight amount of gravitational focussing due the increase in angle, and an effective increase in the sample's acceptance).

SPEAR's current detector is an Ordela 1202N linear position sensitive detector with a resolution of 2 mm FWHM and an active length of 200 mm [6]. We acquired this detector mainly to enable the measurement of diffuse scattering from rough surface and interface samples and to allow us to measure and correct for instrument backgrounds by measuring in the off specular region simultaneously with our specular reflection measurement. For many good quality (flat) samples the detector has provided us with an unexpected bonus, allowing relatively high resolution measurements to be made without further collimating the incident beam and suffering a consequent loss of incident intensity.

Since our reflection plane is vertical this is the usual orientation of the detector allowing us to determine angles from the sample position 3.65 m away with a resoltion of ~0.03° FWHM. Data collected from by this detector is usually displayed by our analysis software as a two dimensional colour image. Figure 4 shows a typical data set, in this case reflection showing interference fringes from a thin film of Nickel on a Sapphire substrate. With angle on vertical scale and time of flight horizontally, the figure clearly shows that we can resolve features in wave vector transfer across the width of the beam, with the interference minima clearly crossing at an oblique angle. In fact, for many samples, such as this one, which show very little diffuse scattering and no significant broadening of the beam due to ripples on the sample surface, we can assume to a good approximation that all the scattering we see is specular, relative to the mean plane of the sample. If this is so the angular structure of the beam is undistorted by reflection from the sample and angular relations across the beam relate directly to relations of angles of incidence. In which case reflection features are aligned in directions of constant

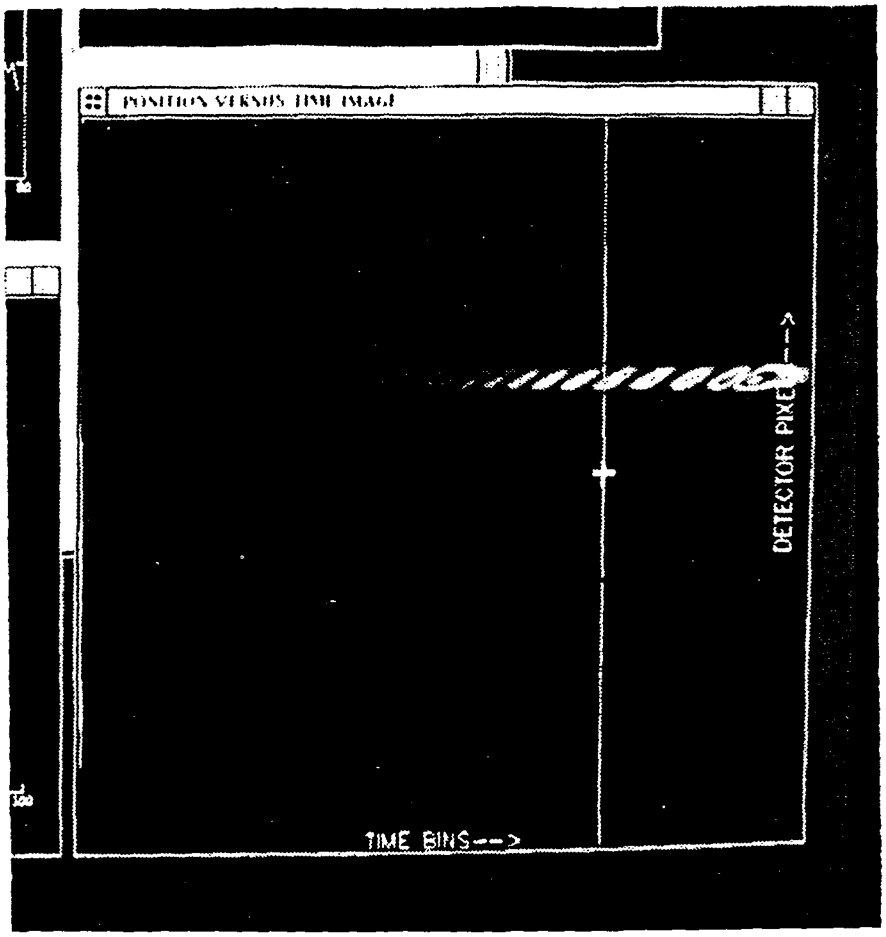

Figure 4. Reflected beam signal display from the SPEAR data acquisition system. The 0.15° collimated incident beam is clearly modulated by finer constant Q_R structure upon reflection, representing higher resolution reflectivity information.

$Q_R = 4\pi\sin\theta/\lambda$. Since $\sin\theta$ is proportional to height on the detector and λ is prortional to the time of flight these are lines of constant height/T, which is what we observe to be the case. (The gravitational shifts mentioned in the previous section distort this relation slightly).

This observation has meant that in general we have needed to do very little fine collimation beyond the bulk shield for good quality samples. Using the postion sensitive detector and the constant Q_R transform outlined above, we are able to determine the reflectivity to a resolution, typically $\Delta Q_R/Q_R$ ~1-2%, limiteded mainly by the angular resolution at the detector (determined by the detector resolution, the sample acceptance and the sample detector separation) and the wavelength resolution (determined by the neutron pulse width and the time of flight binning), and largely independent of our incident beam collimation.

V. CONCLUSION

We have presented some of the design features of the neutron reflectometer SPEAR. Using a T_0 and frame overlap choppers we have shown that background can be significantly reduced allowing measurements of much lower reflectivities. We have found that we can make routine use of neutrons up to 32 Å on pulsed source instrument by proper design of a collimation system which allows for gravitational effects on slow neutron trajectories. Finally, for many samples the use of a position sensitive detector enables us to make reflectivity measurements at good resolution and high incident intensity using a relatively coarsely collimated beam.

References

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Q(M.Furusaka): What is the type of detector you use?

A(R.Pynn): An ORDELA linear PSD with a nominal positional resolution of about 1.5mm.

Q(W.G.Williams): At what wavelength does the gravity correction become significant? Our calculations at RAL suggested gravity effects could be ignored at $\lambda<10$Å.

A(R.Pynn): I would agree. We can just see the correction towards the end of the first frame, around the 14Å mark.