CONF-9510212--10

LA-UR- 95-3905

Title: THE PERFORMANCE OF REFLECTOMETERS AT CONTINUOUS WAVE AND PULSED-NEUTRON SOURCES

Author(s): M. R. Fitzsimmons, R. Pynn

Submitted to: Proceedings of ICANS XIII, Villigen, Switzerland, 11-14 October 1995

Los Alamos
NATIONAL LABORATORY

ICANS-XIII
13th Meeting of the International Collaboration on
Advanced Neutron Sources
October 11-14, 1995
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

THE PERFORMANCE OF REFLECTOMETERS AT CONTINUOUS WAVE AND PULSED-NEUTRON SOURCES

M. R. Fitzsimmons and Roger Pynn
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

ABSTRACT

To quantify gains from time-of-flight (TOF) methods, identical reflectometers viewing a continuous wave (CW) neutron source and a variety of pulsed-neutron sources were simulated using a Monte Carlo technique. Reflectivity profiles obtained for a simple thin-film, reflecting sample were nearly identical in all simulations, and models fitted to the simulated data yielded parameters (film thickness, surface roughness, and scattering length density) that were equally accurate and precise in all cases. The simulations confirm the power of the TOF method and demonstrate that the performance of pulsed sources for reflectometry does not scale simply as the inverse duty factor of the source. In the case of long-pulse sources, the simulations suggest that pulse tails have little effect on results obtained from specular reflectometry and that maximum brightness of the neutron source should be the primary design criterion.

1. The Reflectometers Simulated

This paper presents the results of Monte Carlo simulations of a traditional reflectometer at different neutron sources — CW sources, short pulse spallation sources (SPSS) and long pulse spallation sources (LPSS). The same reflectometer was used in all simulations. Only the characteristics of the neutron source, and the technique used to measure neutron wavelength were changed. In the case of the CW simulation, a monochromating crystal was used to select a nearly monochromatic beam (MB) from the neutron spectrum. In the simulations using the pulsed-sources, time of flight (TOF) was used to determine neutron wavelength.

A "reflectometer of traditional geometry" (cf Figure 1) is an instrument which uses a pair of slits of height h and width w separated by a distance L_S to define an incident beam that is highly collimated in the reflection plane [1]. Since studies of liquid surfaces are not to be excluded, the reflection plane is taken to lie in the vertical plane, so h is small and w is large.

In the simulations, each reflectivity profile was accumulated by following the same procedure that one would use in an actual experiment. The sample was lowered as the angle of incidence of neutrons on the sample, α_i, was increased so the footprint of the beam remained constant and centered on the sample. The widths of the slits defining the incident beam were equal (w = 40mm) and were not changed during the simulation. The vertical dimensions of the slits were also equal, but were adjusted to be proportional to the magnitude of the scattering vector, $Q=4\pi\sin(\alpha_i)/\lambda$, and equaled 1mm when $Q=0.1Å^{-1}$. By changing the vertical opening of the slits in this manner, the geometrical contribution, $\delta\alpha/\alpha_i$, to the resolution, $\delta Q/Q$ was kept constant as the wave-vector transfer perpendicular to the reflecting sample, Q, was varied.

Keywords: Neutron Sources, Reflectometry, Performance, MCLIB

Since the acceptance of the neutron beam by the slits varies as $\phi=(hw)^2/L_S^2$, the intensity of the neutron beam reaching the sample increases with Q^2. This increase partly negates the loss of signal owing to the decrease of the sample reflectivity which varies as approximately Q^{-4}.

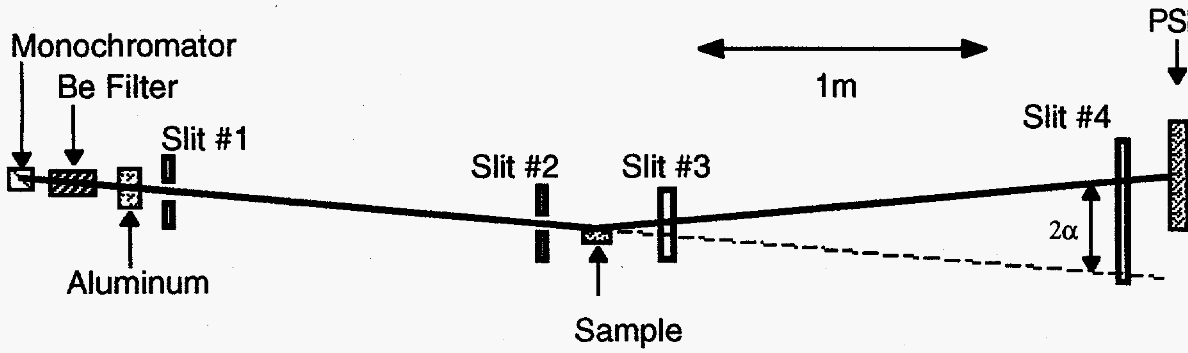

Figure 1: A reflectometer of traditional geometry is depicted. The reflectometer is configured with a monochromator and Be filter, which together produce a monochromatic neutron beam that is used in the MB simulation. The monochromator and Be filter are removed for the TOF simulations.

In the simulations of the MB technique, a graphite crystal was used to select a nearly monochromatic beam with a mean wavelength of 4 Å from a neutron spectrum that is typical of that produced by liquid H_2 moderators. The angular divergence of the neutron beam emanating from the moderator was chosen to be the same as that at the end of a Ni-coated neutron guide for λ_0=4.1Å neutrons, or 0.8° (FWHM). The mosaic spread of the monochromator in the diffraction plane of the crystal was 1.4° (FWHM). The crystal mosaic, the divergence of neutrons from the moderator, and the acceptance of the horizontal slits (in the diffraction plane of the monochromator) fixed the wavelength spread of the incident beam to be $\delta\lambda/\lambda_0$=2% (FWHM). The reflectivity of the monochromator was 80%.

The TOF studies involved simulations of the performance of the reflectometer at ten spallation sources with different proton pulse lengths and different target/moderator configurations. The neutron pulse shapes and spectra have been calculated by Pitcher et al [2] for various target/moderator configurations. The neutron spectra are tabulated and the shapes are fitted to simple functional forms for input into the present simulation of reflectometer performance [3]. The most significant operating parameters of each source are the pulse repetition frequency, the proton pulse width, δt, and the thermal time constant, τ, of the moderator [4]. These parameters are given in Table 1 for the different pulsed sources. In all cases, the proton power incident on the spallation target was held constant at 1 MW. The time-averaged brightness of the sources with τ = 370ms (PSS-1, -3, -5, -6, and -7) are the same as that of the CW source. Sources with longer time constants have larger time-averaged brightness. These are given in the Table.

In the TOF simulations, the length of the incident beam path was chosen so that FWHM wavelength resolution at the minimum wavelength used matched that used in the MB simulations. The operating parameters of the instruments investigated in the simulations are also given in Table 1.

In order to achieve good resolution in the TOF simulations either the time of flight must be large, or the neutron pulse duration must be small. The former can be accomplished by using very slow neutrons that travel a short distance (PSS-1), or by using shorter wavelength neutrons that travel a long distance (PSS-2 through -6). Alternatively, good temporal resolution can be achieved by using a combination of short proton pulses and moderator time constants (PSS-8 through -10), or by using a chopper (PSS-7). The chopper pulse width, δt_c, can be made as small or as large as needed for the measurement.

The TOF instruments vary in length from 16 to 60 m. Only the last 4 m of each instrument is the same for all simulations.

Table 1: Operating Parameters of the Instruments and Sources

Source	f [Hz]	δt [ms]	τ [us]	δt_n [ms]	$B \cdot 10^{16}$ n/st/m^2/s	L [m]	λ_{min} [Å]	$\Delta\lambda$ [Å]	F	t_{min} [ms]	M	Technique
CW	steady	-	-	-	5.5	4	4.1	$=\delta\lambda$	-	n/a	100	monochromator
PSS-1	60	1	375	1	5.5	18	7.85	3.25	3	35	8	short flight path, long λ, (cold neutrons)
PSS-2	60	1	230	1	3.9	60	3.38	0.94	4	51	13	long flight path, moderate λ, small τ
PSS-3	60	1	375	1	5.5	60	3.38	0.94	4	51	13	long flight path, moderate λ, moderate τ
PSS-4	60	1	490	1	6.7	60	3.38	0.94	4	51	13	long flight path, moderate λ, large τ
PSS-5	120	0.5	370	0.56	5.5	30	3.41	0.95	3	26	13	twice the freq. and half the width of PSS-3
PSS-6	120	0.735	370	0.76	3.9	36	3.76	0.62	4	34	21	somewhat longer pulse than PSS-5
PSS-7	60	1	370	0.33	5.5	23	2.86	2.76	2	16.7	4	short flight path, pulse-imaging chopper
PSS-8	10	0.3	230	0.33	3.9	30	1.96	11.3	1	15	1	moderate flight path, PSS-2 at 10Hz and 0.3ms pulse
PSS-9	10	0.1	230	0.20	3.9	16	1.64	23.2	1	6.6	1	SNQ facility
PSS-10	10	0.001	230	0.19	3.9	16	1.64	23.1	1	6.6	1	1MW low frequency target

f	source frequency = $1/\Delta T$
δt	proton pulse width
τ	composite moderator time constant
δt_n	neutron pulse width
B	source brightness
L	flight path length
λ_{min}	minimum neutron wavelength available
$\Delta\lambda$	wavelength bandwidth available
F	end of detection frame in units of 1/f (detection frame is 1/f long)
t_{min}	time-of-flight of a neutron with wavelength λ_{min} traveling a distance L
M	number of macro-measurements needed to measure the reflectivity profile from Q_c to 0.106Å^{-1}

The TOF instruments also use up to three choppers to condition the neutron beam. The first is called the t_0 chopper and is placed at 5 m from the moderator in our simulations. The purpose of the t_0 chopper is to stop most high energy neutrons and γ-radiation from reaching the experimental area. The second chopper is the frame overlap chopper and intercepts most of the very slow neutrons generated during earlier pulses [5]. The frame overlap chopper was located 6 m from the moderator. The third chopper is called the frame definition chopper and, as its name implies, serves to define the frame, F, in which neutrons are detected relative to their creation at t_0. The position of the frame definition chopper (between the frame overlap chopper and the reflectometer) was chosen to select neutrons with wavelengths between λ_{min} and λ_{max}. The frame definition chopper was not needed for the simulations PSS-8, -9, and -10, since these instruments use neutrons produced in the first frame.

2. The Reflecting Sample

The reflecting sample used in the simulations was a 600Å thick film of d-polystyrene (d-PS) on a Si substrate. The roughnesses of the air-PS and PS-Si interfaces were 5Å (FWHM). The selection of such a relatively simple sample tends to favor the simulation of the MB technique over the TOF technique, since information over a very broad region of Q is not needed to fit a simple model. By favoring the MB technique, the intensity gains realized when employing the TOF technique over the MB technique are believed to be conservative estimates.

3. The Simulations

The reflectivity profile for the MB technique was simulated from Q=0.009 to 0.106Å^{-1} by taking equal steps in α_i of 0.02°. For Q>0.02Å^{-1}, the counting time was increased with Q^2. This increase combined with the increase of the acceptance of the slits yielded an exposure that varied as Q^4 and largely negated the Q^{-4} decay of the reflectivity profile. The result was a reflectivity curve whose fringe maxima had equal statistical precision.

The Monte Carlo simulations of the TOF instruments proceeded in the same manner as the MB simulation, except that neutrons traveled along a neutron guide and through choppers before entering the reflectometer. One important difference between the simulations involves the manner in which the data were simulated for different ranges of Q. In the MB simulation, λ_0 was fixed, so Q was sampled by changing α_i. For a TOF instrument, which utilizes a relatively small bandwidth, such as those for the simulations of an LPSS, the simulation can be conducted in one of two ways. The first, like the MB simulation, involves changing Q by changing α_i a few times. The second fixes α_i and increments the mean wavelength used. The best choice is not immediately clear, since the MB method always utilizes the most intense part of the spectrum at the expense of slit and/or sample acceptance.

The ability to select either constant-angle or constant-wavelength methods is an added degree of flexibility that is not found for reflectometers at CW or SPSS (e.g. PSS-8, -9 and -10). This extra degree of freedom is an advantage of the narrower bandwidth. For example, the ability to measure the reflectivity profile by scanning wavelength permits studies of samples in fixed geometry and is a virtue of the TOF technique. On the other hand, the ability to examine small portions of reciprocal space, e.g. a Bragg reflection from a multilayer sample, without suffering a loss of performance, is an advantage of the reflectometers at the longer pulsed-sources and CW sources.

Whereas the MB simulation required 100 separate measurements to sample the Q-range from 0.009 to 0.106Å^{-1}, a dozen or fewer separate measurements (called macro-measurements) were needed for the TOF simulations (within one macro-measurement are many more micro-measurements made as a function of neutron wavelength). For the SPSSs, a single macro-measurement sufficed for each of our simulations, whereas more such measurements are required to accumulate the full range of required data at an LPSS.

4. The Stopping Criterion

There are several criteria that one can imagine using to terminate each simulation. For example, one might assert that the total number of neutrons used in each simulation should be the same or one might demand equal statistical precision of the reflectivity profile at the largest value of Q interrogated. However, since experimenters are usually interested in fitting reflectivity data to a model, we have chosen to terminate each simulation when models fitted to the simulated data give equal statistical precision in the fitted parameters. These parameters were chosen to be the thickness, scattering length density and interfacial roughness of a film on a substrate whose scattering power was set equal to the known value for silicon. We found that, with this criterion, each simulation gave results of similar accuracy (i.e., there were no obvious systematic errors).

The measurement times deduced from the simulations depend, of course, on the stopping criteria used. However, the only really significant (i.e. > 10%) effects we have observed in this regard occur when the second stopping criterion (equal statistical precision at high Q) is used. In this case, the SPSS instruments need 4 to 6 times longer to collect data than they do when the third criterion (equal precision of fitted parameters) is used. In order not to introduce this sort of bias, we have chosen to present data only for the third criterion.

5. Gains Obtained at Pulsed Sources

In Table 2, we give the measurement times deduced from each simulation. The inverse of these numbers, normalized to the data collection time at the CW source, gives the overall gain, G_{Π}, for the pulsed sources. While the major part of this gain derives from the use of the TOF method, there are other factors that also need to be considered. All of these additional factors — monochromator reflectivity, transmission through slits of a given area, guide and filter transmissions, intensity variations across the neutron spectrum, and source brightness — can be quantified with a high degree of confidence. Noting that the total gain is the product of all gain factors, the TOF gain can be deduced by dividing the total calculated gain by the product of the additional gain factors noted above. The TOF gain calculated in this way is also given in Table 2, where it is compared with the naive values that would be deduced either from the inverse of the source duty cycle or a comparison of useful wavelength bandwidth and wavelength resolution at the minimum wavelength used.

Table 2: Collection Times, T, Performance, G_{Π}, and TOF Gain, G_{TOF} Factors

Source	T [s]	G_{Π}	G_{TOF}	$\frac{\Delta T}{\delta t_n}$	$\frac{\Delta\lambda}{\delta\lambda}$
CW	994	1	-	1	1
PSS-1	106	9.4	8	17	21
PSS-2	115	8.6	14	17	14
PSS-3	90	11	13	17	14
PSS-4	63	15.8	15	17	14
PSS-5	95	10.5	11	15	14
PSS-6	126	7.9	7	11	8
PSS-7	66	15	21	51	48
PSS-8	17	58	176	303	288
PSS-9	17	58	225	500	707
PSS-10	17	58	225	526	707

Several results are clear from Table 2:

- All of the pulsed sources give shorter collection times than the CW source, even though all but one of the pulsed sources (PSS-4) have equal or lower average source brightness than the CW source.

- The performance of pulsed source reflectometers does not scale simply as the inverse source duty cycle, because not all information obtained during a single TOF measurement is equally useful. For the simulations reported here, a logarithmic dependence of performance on inverse duty cycle appears to describe the results, but further simulations will be needed to establish whether this behavior is in any way universal.

- Comparing PSS-4 and PSS-10 shows that a reflectometer at a 1 MW, 10 Hz SPSS is likely to outperform that at a 1 MW, 60 Hz LPSS by about a factor of 4.

- Comparing PSS-8 and PSS-10 shows that there is no performance improvement obtained by shortening the proton pulse at an SPSS to a value below 300 μsec. Much of the effort and technical complexity that such sources invest in accumulator rings or rapid cycling synchrotrons is thus wasted for reflectometry.

- For reflectometers at an LPSS, the best performance is obtained by using the largest integrated intensity, even though this also implies that the neutron pulses have long tails.

- Comparing PSS-3 and PSS-5 shows that similar performance is obtained at 120 Hz and 60 Hz LPSS with equal duty cycles.

- Simulations PSS-3 and PSS-7 show that a pulse shortening chopper is advantageous at an LPSS, at least in the case considered. The added performance and flexibility provided by this chopper makes it attractive.

6. Final Remarks

The results presented here apply strictly to the performance that would be obtained for reflection experiments with a simple, single-layer sample and an experimental protocol that measures reflectivity out to Q = 0.1 Å^{-1}. One might argue that this is not a typical case. For example, studies of a Bragg reflection from a multilayer would normally not require such a large Q range, thus the performance of the SPSS compared to the LPSS would be more greatly

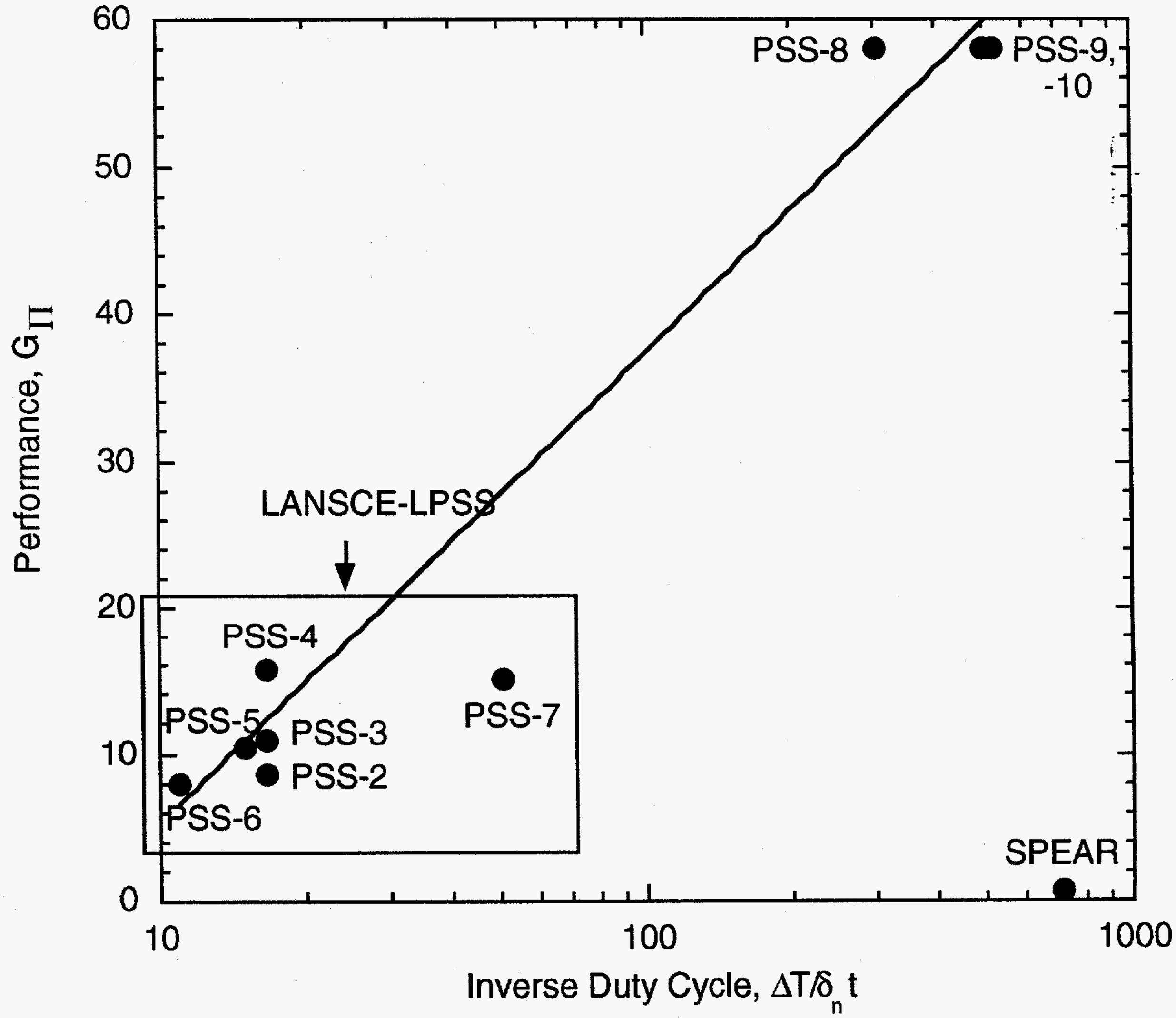

Figure 2: The performance of reflectometers at various spallation sources is plotted versus inverse duty cycle. For comparison purposes the performance of the reflectometer, SPEAR, at the existing LANSCE source (0.06MW) is also shown. The performance of a reflectometer at the ILL on this scale is taken as unity.

reduced than our results would indicate, since the CW and LPSS measurements would be quicker, ie. fewer macro-measurements would be required. On the other hand, there are many experiments that require an even wider range of Q than that studied here as well as high statistical accuracy of the data at large Q. The advantage of using a shorter pulse in this case is that most of the reflectivity profile, albeit the easiest and least time-consuming part of the profile to measure is measured during the time spent measuring the low reflectivity region.

7. Acknowledgments

We wish to thank P.A. Seeger for providing and maintaining the Monte Carlo simulation code with which this study was possible. We also acknowledge valuable discussions with L. Daemon, R. P. Hjelm, C. F. Majkrzak, E. Pitcher, S. Satija and G. S. Smith. This work was supported by the U.S. Department of Energy, BES-DMS, under Contract No. W-7405-Eng-36.

8. References

[1] For measurements of specular reflectivity, the use of a collimated beam is highly non-optimal. In fact, for specular scattering, the optimal condition for obtaining data is to use a highly divergent beam that converges at the sample position, and to measure the angle of incidence from the scattered radiation with a position sensitive detector. This procedure is employed with the reflectometer at the Los Alamos Neutron Scattering Center called SPEAR and decreases collection times dramatically (a factor of 2 is conservative).

[2] E.J. Pitcher, G.J. Russell, P.A. Seeger and P.D. Ferguson, "Performance of Long-Pulse Source Reference Target-Moderator-Reflector Configurations," these proceedings.

[3] P.A. Seeger, "The MCLIB Library: Monte Carlo Simulation of Neutron Scattering Instruments," these proceedings.

[4] The additional parameters used for the pulse shape were the same for all cases. The epithermal time constant (proportional to wavelength) was 8μs/Å and the switch function between the epithermal and thermal was exp[-(0.9Å/λ)2.5]. Both terms were convoluted with Gaussians proportional to wavelength. The rms width for the Gaussian representing the epithermal component was 1.9μs/Å and for thermal was 2.9μs/Å. These spectra were earlier versions of these tabulated in ref. [2], with composite Be/Ni reflectors.

[5] Alternatively, the frame overlap chopper can be replaced by a mirror whose angle of incidence relative to the neutron beam is set to reflect very long wavelength neutrons out of the beam. This approach has the advantage of removing one of the choppers; thus, reducing the cost of the instrument.