JP0150542

ICANS-XV
15th Meeting of the International Collaboration on Advanced Neutron Sources
November 6-9, 2000
Tsukuba, Japan

11.1
Real Time Neutron Reflectometry Using Neutron Optical Imaging

Gregory S. Smith* and Jaroslaw Majewski

Manuel Lujan Jr. Neutron Scattering Center, MS H805, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
*Email: gsmith@lanl.gov

Abstract

We will describe recent improvements to the SPEAR reflectometer at the Manuel Lujan Jr. Neutron Scattering Center at Los Alamos. One of the changes consists of wider convergent, incident-beam, collimation to take advantage of optical imaging for specular scattering. In addition, the instrument now views a partially coupled liquid hydrogen moderator as opposed to the decoupled moderator that was previous in-place. While the wavelength distribution is poorer, it matches the time (wavelength) resolution of the reflectometer more closely with the angular resolution. Since the integrated intensity of the partially coupled moderator is higher than the decoupled moderator, we show a similar gain in incident beam flux on the sample without loss of the ability to separate fringes. The increases in intensity from the moderator gain and the improved collimation combine to allow us to measure reflectivities with good statistics down to 10^{-4} in a matter of minutes and reflectivities of 10^{-6} in an hour. Examples of measurements showing the gain in data accumulation rates are presented.

1. Introduction
Within the last decade, X-ray and Neutron reflectometry have emerged as primary non-destructive tools for investigating the fine-scale architecture of interfaces, mainly because they provide excellent resolution in the most relevant range of length scales (10-1000Å). Fully exploiting these methods in soft-matter research is a prerequisite to realizing "engineering" organic interfaces for a broad range of applications, including high performance synthetic coating, "smart" synthetic surfaces, and a variety of biomedical applications.

Neutrons offer two distinct advantages over X-rays in soft matter research: (1) they generally have much higher transmission through condensed organic media, which permits the study of buried interfaces, and (2) contrast can be varied by deuterium labeling without large chemical perturbations. The primary disadvantage of neutrons is the relatively low intensity of typical neutron sources. The neutron flux at the sample is generally several orders of magnitude lower than the photon flux at a synchrotron X-ray source. This precludes real time studies of many systems and limits the range of length scales which may be examined. Therefore,

gaining neutron intensity while maintaining the instrumental resolution has the potential for opening new areas of science for neutron reflectometry.

Figure 1 shows the geometry for the specular reflection process. The scattering vector only has a component normal to the surface, Q_z where

$$Q_z = 4\pi \sin(\theta)/\lambda \qquad (1)$$

and the Q_z resolution, σ_{Qz}, is given by:

$$(\frac{\sigma_{Q_z}}{Q_z})^2 = (\frac{\sigma_\theta}{\theta})^2 + (\frac{\sigma_\lambda}{\lambda})^2 . \qquad (2)$$

The process of specular scattering results in an angular delta function response. It has been shown that the angular portion of the Q_z resolution of a reflectometer is independent of the divergence of the incident beam [1]. By measuring only the angle of reflection, since the angle of incidence is equal to the angle of reflection, one may determine the value of the perpendicular scattering vector, Q_z (Fig 1). This technique is referred to as neutron optical imaging. Using a position sensitive detector, one can determine the angle to within the resolution of the detector. In this case, the detector's resolution is the only contribution to σ_θ in Eqn. 2. Therefore, by opening the incoming divergence of the beam we can gain intensity without the loss of resolution.
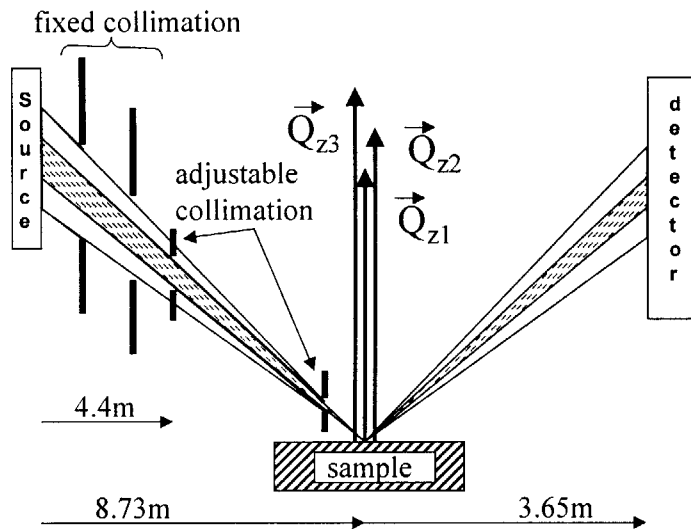

Figure 1 A schematic representation of specular scattering on SPEAR. The different values of the scattering vector are for a constant wavelength where Q_{z1}, Q_{z2}, and Q_{z3} are associated with the lower, middle and upper rays, respectively.

Given that the angular resolution is determined by the detector resolution and that the contributions to the resolution add in quadrature, ideally one would prefer to match the wavelength resolution with the angular resolution to optimize the overall performance of the instrument. This is accomplished by matching the first term in equation 2 equal to the second term. For a spallation neutron source, the wavelength is measured using time-of-flight techniques. In that case,

$$(\frac{\sigma_\lambda}{\lambda})^2 \approx (\frac{\sigma_{t1}}{t})^2 + (\frac{\sigma_{t2}}{t})^2 , \qquad (3)$$

where σ_{t1} is the uncertainty in the time-of-flight, t due to the electronic measuring errors and σ_{t2} is the uncertainty in the time of flight due to the finite pulse width of the neutron source. The largest contribution to σ_λ for the SPEAR reflectometer at the Manuel Lujan Jr. Neutron Scattering Center (the Lujan Center) is the pulse width of the neutron source, σ_{t2}. Therefore, if one closely matches the resolution contribution due to pulse width with the angular resolution, the intensity can be maximized without degrading the Q_z resolution.

The Lujan Center produces neutrons through the spallation process by injecting 800MeV protons into a tungsten target. The resulting high-energy neutrons have such short wavelengths that they are not useful for condensed matter research. The neutrons are moderated to lower energies though interactions with hydrogenous material (the SPEAR moderator is liquid hydrogen). This produces a thermal neutron energy spectrum. For neutrons in a given wavelength range there is a spread in emission time from the moderator. By changing the geometry and the materials surrounding the target and moderators, both the pulse width and the spectral intensity may be varied. For instance, if a heavy material (reflector) surrounds the moderator/target area, neutrons can be scattered back into the moderator. This increases the intensity of the neutrons at a spectrometer viewing that moderator. At the same time, the delay in neutron arrival at the moderator increases the pulse width. The moderator may be decoupled from the reflector if a thermal neutron absorbing material surrounds the moderator on the sides not facing the spectrometer. Therefore, by changing the geometry of the target/moderator/reflector system (TMRS), one may match the temporal width of the neutron pulse to the desired resolution for the instrument.

2. Experimental Details
Instrument resolution
As noted above, the geometrical resolution will be determined by the detector pixel resolution. The SPEAR detector is an Ordela model 1202N linear position sensitive detector. For this detector, $\sigma_{pixel} \sim 0.75mm$, and the sample to detector distance is nominally 3.65 m. This yields $\sigma_\theta = .012°$ and $\sigma_\theta/\theta = 0.024$ for $\theta = 0.5°$.

The Lujan Center pulsed source repetition rate is 20 Hz. This defines the data collection time frame to be 50 ms. The minimum time for the data collection in a frame is limited to ~3ms due to the opening time of a T_0 chopper which blocks the initial burst of high energy neutrons. Since 3ms<t<50ms (1Å< λ<16Å), the time (wavelength) resolution will vary from $0.06<\sigma_t/t< 0.003$ compared to the 1997 value of $.028< \sigma_t/t<.0017$. This means that the resolution will be matched to the geometrical resolution for times (wavelengths) greater than 7ms (2.3Å). The resolution will still be dominated by the geometrical resolution for most of the wavelength range so that we have gained intensity without a detrimental effect on the resolution.

Calculated intensity gain
SPEAR views a liquid hydrogen neutron moderator. In 1997, this moderator was decoupled from neutrons reflecting back from the surrounding materials. This was accomplished by covering all but the
front face of the liquid hydrogen vacuum vessel with a neutron adsorbing material. The target/moderator/reflector system was upgraded in 1998 where the new design for this moderator included removing the decoupler and improving the coupling between the moderator and the reflector. The calculated increase in intensity from the 1996 coupled to the 1998 decoupled moderator was 2.5 times, and the calculated rms pulse width increased from 85 to 170 μsec [2]. The geometry of the 1997 decoupled moderator was not the same as the calculated comparison. The gain from 1997 to 1998 is expected to be higher than 2.5 times with a similar change in the pulse width[3]. At the same time the proton current on target was increased from 70 μamps to 100 μamps. This yields a minimum expected gain in intensity from the source, $G_{source} = 3.6$.

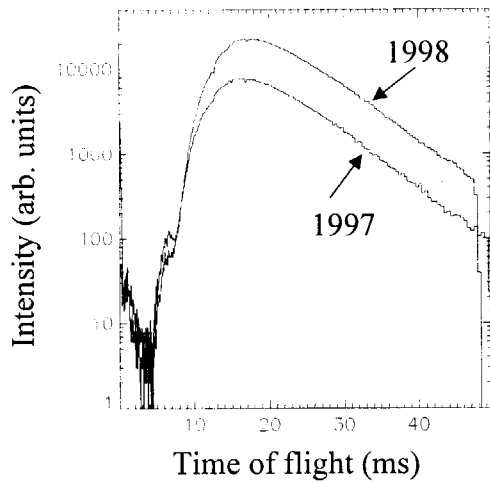Since the detector resolution determines the geometrical resolution, we can take advantage of the techniques of optical imaging [1] to further increase the flux at the sample. To do this, we changed the amount of the moderator viewed by the sample by changing the fixed collimation inside the bulk biological shielding. Since SPEAR has a vertical scattering plane, the collimation is designed to converge vertically at the sample position (8.73m from the moderator) and at the detector horizontally (12.38m from the moderator). In 1997 the view of the moderator was an area 4.5cm wide and 3.1cm high defining a 0.21°horizontal and a 0.20°vertical divergence. This was increased to 9.1cm wide and 7.9 cm high yielding a 0.42°horizontal and 0.52° vertical beam divergence. The height of the sample slit was the same for 1997 and 1998, but the width of the slit

Figure 2 The raw data from two similar samples measured with the slits set as in 1997 and for a full view of the moderator available in 1998.

increased from 1.3cm to 2.3cm. The intensity can be calculated from the solid angle viewed by the sample. This is simply given by:

$$I \approx \frac{A_1 \times A_2}{s^2} \tag{4}$$

where A_1 is the area of the first slit, A_2 is the area of the second slit and s is the distance between slits. Here we assume that the moderator area is the first slit although this view is actually determined by a set of slits in the bulk shield. The ratio of the calculated intensities for 1997 to 1998 gives an expected gain from increasing the moderator view, $G_{geom}=9.1$. Combined with the gain from the moderator we expect an overall gain in flux at the sample $G= G_{geom} \times G_{source}= 33$.

Measured intensity gains

To determine the actual gains on SPEAR two sets of measurements were made. One to assess the source intensity gain and another to assess the intensity gain from a larger view of the moderator. In each case, the reflectivity from a sample consisting of a film of a polymer on a copper coated silicon substrate was measured. A similar sample was run in 1997, so this provides a basis for comparison of the intensities. The adjustable slits at 4.4m from the moderator and at the sample position were used to define the view of the moderator. First to measure the affect of the moderator gain, a 1998 measurement was made of the reflectivity using the same slit settings as in 1997. The results of the reflectivity measured in 1998 and for a similar sample with the same slits settings made in 1997 are shown in Figure 2. The data was collected in 1998 to obtain similar statistics to the 1997 run. The calculated Q_z resolution is nearly the same in each case as well. By summing the raw data in Figure 2 over similar time ranges and normalizing to the same number of proton pulses on target, we get a ratio of the intensities (normalized to the same amount of time) of 5.9. If we take out the fact that the average current in 1998 was 100 µamps versus 70 µamps, the gain factor for the moderator coupling is 4.1.

To assess the geometry gain, we performed two runs on the polymer sample in 1998. One with the adjustable slits set at the 1997 values and one with the slits open to view the largest area on the moderator. The adjustable slits are located at the sample position 8.73m from the moderator and at 4.4m from the moderator (Figure 1). The slit settings are shown in Table I. Again, the ratio of the number of counts in the raw data for each set was used to indicate the intensity gain. This ratio of the 1998 intensity to the 1997 intensity normalized to the number of proton pulses was 5.7. The predicted gain from the slit settings was shown in Table I as 9.1.

Table I.

	SPEAR 1997	SPEAR 1998
Dimensions of viewed moderator	4.5cm wide, 3.1cm high	9.1cm wide, 7.9cm high
Slit1 settings (4.4m from moderator)	15mm high, 30mm wide	40mm high, 60mm wide
Slit 2 settings (sample position at 8.73m from moderator)	1.134mm high, 15mm wide	1.134mm high, 30mm wide
Calculated intensity gain from moderator view	1	9.1
Calculated intensity gain from brightness gain and current increase	1	3.6
Measured intensity gain from moderator view	1	5.9
Measured intensity gain from brightness gain and current increase	1	5.7

3. Results and Discussion

Table I lists some of the instrument parameters and the calculated and measured gains. We see that the geometrical gain is lower than predicted. This may be due to some uncertainties in the slit sizes for the small slits or uncertainties in scaling the beam current from one year to the next. In addition, the calculation doesn't take into account the convolution of the reflectivity with the incident beam spectrum. If the slits are opened symmetrically then some intensity for a given value of Q_z is gained at shorter wavelengths and some at longer wavelengths. Since there are more neutrons at shorter wavelengths, this can bias the prediction. On the other hand, the gain in intensity of the moderator and current are higher than predicted. This may be due to the same effects discussed for the geometrical gain. In addition, the calculations for the partially coupled moderator [2] were for a different geometry that existed in 1997. The 1997 moderator was a temporary decoupled moderator, which was expected to have a lower intensity than the calculated value. The gain factor is expected to be larger than the 2.5 times [3]. Therefore, it is fortuitous that

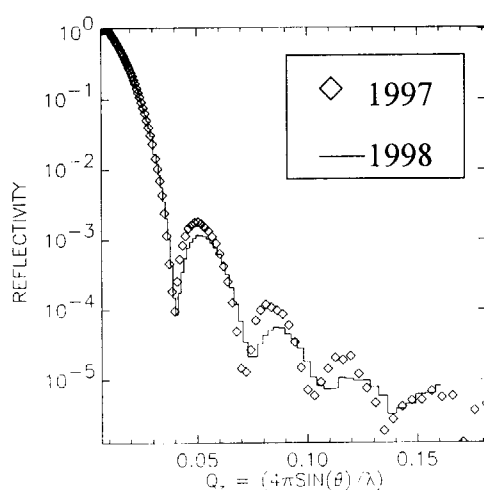**Figure 3 Comparison of the reduced data from two similar samples. The data show that the resolution is similar in the two cases.**

the measured total gain is equal to the prediction. To obtain more accurate predictions, one must perform detailed Monte Carlo calculations that include the spectral and geometrical values for the instrument. The uncertainties in the instrumental settings and beam current measurements must also be controlled more precisely.

4. Conclusions

Overall, the preliminary data presented here show that one does gain intensity by making the changes described above. The reflectivity data in Figure 2 can be reduced, and the 1998 data (on a similar sample) is comparable to the data obtained by using a decoupled moderator and tighter incoming beam divergences (Figure 3). This supports the idea that runs that took hours in the past will now take minutes to obtain the same statistical accuracy. Conversely, the increase in intensity should allow one to measure lower values of the reflectivity for experimental runs lasting several hours. This opens new possibilities for non-equilibrium systems and pushes the measured reflectivity to lower values and smaller length scales.

References

[1] W. A. Hamilton, J.B. Hayter, and G.S. Smith, J. Neutron Res., 2 (1), 1(1994).

[2] P. D. Ferguson, G. J. Russell, and E.J. Pitcher, in Proceedings of the International Workshop on Cold Moderators for Pulsed Neutron Sources, J.M. Carpenter and E. B. Iverson eds., Argonne National Laboratory, 67(1997).

[3] Private communications with P. D. Ferguson at Los Alamos National Laboratory.

Acknowledgements