Multi- Incident Beam Compact Time-Of-Flight SANS Instrument with 1 Millimeter Diameter beams

M Furusaka¹, S Takeda¹, A Homma¹, F Fujita¹, T Miyata¹, Y Kiyanagi¹ and M Ohnuma²

¹Faculty of Engineering, Hokkaido University, Kita-13 Nishi-8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan
²Quantum Beam Unit, National Institute for Materials Science, 1-2-1 Sengen, 305-Tsukuba 0047, Japan

E-mail: furusaka@eng.hokudai.ac.jp

Abstract. Although scanning small-angle x-ray scattering (SAXS) instruments are available, almost no attempt has been made to realize scanning small-angle neutron scattering (SANS) instruments because of rather low flux available in the case of neutron scattering. We propose the use of a multi-pinhole collimator–pinhole combination to reduce the beam size to about 1–2-mm diameter immediately in front of a sample. With such small pinholes, we can produce a rather short instrument, on the order of 1–2 m, to have access to conventional minimum $Q$ of about 0.03 nm$^{-1}$. Because multiple beams hit the same detector at different parts of the detector, the obtained SANS patterns mutually overlap. However, using a wavelength-dependent SANS pattern, we were able to resolve the overlap in theory. We conducted a proof-of-principle type of experiment at the Hokkaido University electron linear accelerator based pulsed cold neutron source (HUNS) facility. We made several multi-pinhole plates with thin beam holes arranged in hexagonal patterns. By changing the moderator and detector pixel size and the instrument overall size, we can optimize the pinhole size and their layout. We conducted experiments using welded steel samples and seek the difference in SANS pattern in the welded and non-welded parts, as well as the heat-affected zone between them. We demonstrated the principle of resolving overlapped SANS patterns from the adjacent beams using wavelength-dependent scattering.

1. Introduction

Scanning X-ray SAS [1–5] is a standard method used for analyzing position-dependent nanostructures in objects that have hierarchical structures such as bone, but people usually avoid performing scanning “NEUTRON” SAS because of the low flux available to neutron instruments in general. In fact, SAS measurements necessitate tight angular collimation of about a few millirads, resulting in low flux at the sample position. The only available means of overcoming this is to enlarge the sample size, i.e. just opposite to scanning concept.

We are proposing the use of multiple pinholes of approximately 1 mm diameter for collimation. Because of the small pinhole size, the use of rather short path lengths of about 1–2 m can provide the necessary collimation. Minimum $Q$ that are typically available using conventional SANS instruments, such as several times 0.01 nm$^{-1}$ to 0.1 nm$^{-1}$, can be realized by the configuration. The intensity drops rapidly and concomitantly with the (pinhole diameter)$^{-2}$. To compensate for this, we propose the use of
many small pinholes for the entrance of the collimator section to gain intensity and use a single pinhole immediately in front of the sample position, and a detector at the right end. The possibility also exists of using a multi-pinhole aperture plate immediately in front of the sample.

We then confront overlapping small-angle scattering from different pinholes. Separating them is expected to be difficult in the case of a reactor instrument, but in the case of a time-of-flight instrument, it is possible to resolve the overlap because we can obtain multiple $I(q)$ information for the same $q$ from different wavelengths and position regions.

We therefore made a proof-of-principle mockup instrument to study whether such a concept actually works at the Hokkaido University electron linear accelerator based pulsed cold neutron source facility (HUNS).

2. Principle of multi-incident beam instrument

The $Q$ resolution $\Delta Q$ of a pinhole-geometry SANS instrument can be expressed as

$$\Delta Q = \frac{4\pi}{\lambda} \cdot \Delta \theta$$

$$= \frac{2\pi}{\lambda} \sqrt{\frac{2\ln 2}{3} \left( \frac{A_0^2 + A_5^2}{L_2^2} + \frac{A_5^2 + A_0^2}{L_1^2} \right)} \cdot \frac{\Delta Q}{Q} \cdot$$

where $\lambda$ is the wavelength, $\Delta \theta$ the angular resolution, $A_0$ is the first aperture, $A_5$ is the aperture immediately before the sample, $A_0$ is the detector position resolution, $L_1$ the length between $A_0$ and $A_5$, and the $L_2$ sample to detector distance. In the case of time of flight (TOF) instrument, $\Delta \lambda$ is negligible in general.

Typically, $A_5$ is chosen as nearly 10 mm or slightly less in diameter. When $L_2 \approx 5$ m and $\lambda \approx 0.6$ nm is assumed, $\Delta Q$ becomes about 0.01–0.02 nm$^{-1}$ and $Q_{\text{min}}$ of about 0.03–0.06 nm$^{-1}$ can be measured. If we were able to reduce $A_5$ to about 1 mm, then we would be able to obtain the same $Q$-range using only $L_2 = 0.5$ m, with the sacrifice of intensity to 1/100.

To regain the intensity, we can use a multi-pinhole aperture for the first or second aperture, as presented in figures 1 and 2. For the first case (intensity gain type, (type-1)), all neutrons passed through the pinholes converge onto the single pinhole immediately before the sample position that is
placed exactly halfway between the incident pinholes and the detector position at the end. Every single beam generates SANS and on the detector, all the SANS patterns from every beam overlap. A method of unwrapping the degenerated SANS patterns will be presented later in this paper. We are using the TOF method. Consequently, we can make use of wavelength dependence to unwrap the pattern. For example, with short wavelength neutrons, the SANS patterns are expected to be concentrated near each direct beam and are expected to accommodate the unwrapping fairly easily.

In the second case (scanning SANS or imaging type, (type-2)), the beam goes through a single pinhole at the entrance of the collimator section. Because the multiple pinholes are immediately in front of the sample, neutrons hit different positions of the sample. It is like scanning SANS experiment, meaning that we observe different patterns from each beam. Again the SANS patterns overlap, but we can unwrap them as described later.

Both types of configuration can be mixed. Some examples are related to the first aperture: we can use a seven-pinhole plate and put three pinholes immediately before the sample to produce 21 different SANS patterns from three different positions on the sample.

Because all SANS become the same, it is far easier to resolve the SANS scattering if we use type-1 geometry. The time required to scan is the same for both type-1 and type-2 measurements.

3. Instrument design principle
One example of multi-pinhole patterns is presented in figure 3.

![Figure 3. Example of multi-pinhole patterns.](image)

The number of pinholes $n_{\text{pinhole}}$ is calculated as

$$n_{\text{pinhole}} = 1 + \sum_{i=1}^{m-1} 6i,$$  \hspace{1cm} (2)

where $m$ is an integer parameter related to the approximate distance between the center to the position of pinholes specifying the number of layers, starting from 1. For $m=1$, there is only one pinhole at the center. For $m=2$, six pinholes are added, making the total number 7. The total numbers of pinholes therefore become 1, 7, 19, 37, 61, ... as parameter $m$ increases.

When designing the pinhole pattern, a constraint arises from the detector pixel size $d_{\text{pix}}$, pixel number $n_{\text{pix}}$, and the moderator size $d_{\text{mod}}$. We first assumed the direct beam size $d_{\text{dir}}$ as $r_1$ times larger than $d_{\text{pix}}$. Here, $r_1$ should be a number greater than 2.

$$d_{\text{dir}} = r_1 d_{\text{pix}}$$  \hspace{1cm} (3)
The distance between the direct beam images on the detector can be expressed as

\[ d_{\text{patt}} = r_2 \, d_{\text{dir}}. \]  

(4)

When the detector accommodates \( m \) layers of the pinhole pattern, the overall detector size \( d_{\text{det}} \) can be expressed as

\[ d_{\text{det}} = (2 \, m - 1) \, d_{\text{patt}}. \]  

(5)

allowing a \( 1/2 \) \( d_{\text{patt}} \) margin outside of the pinholes.

Then, the sample to detector distance \( L_{\text{sd}} \) and the collimator length \( L_{\text{cs}} \) can be related to the moderator to sample distance \( L_{\text{ms}} \) and the moderator size \( d_{\text{mod}} \), using \( d_{\text{det}} \) as

\[ d_{\text{mod}} \approx \frac{L_{\text{ms}}}{L_{\text{sd}}} \, d_{\text{det}}, \quad L_{\text{sd}} \approx \frac{d_{\text{det}}}{d_{\text{mod}}} \, L_{\text{ms}} \]  

(type 1 case),

\[ d_{\text{mod}} \approx \frac{L_{\text{mc}}}{L_{\text{cs}} + L_{\text{sd}}} \, d_{\text{det}}, \quad L_{\text{cs}} + L_{\text{sd}} \approx \frac{d_{\text{det}}}{d_{\text{mod}}} \, L_{\text{mc}} \]  

(type-2 case),

respectively corresponding to type-1 and type-2 cases. \( L_{\text{ms}} \), which is not a free parameter, is usually of the order of 5–10 m. When the pinhole size at the sample position \( d_{\text{patt}} \) is fixed, \( L_{\text{cs}} \) or \( L_{\text{sd}} \) is determined according to the requirement from the \( Q \) resolution \( \Delta Q \).

\( \Delta Q \) is a function of the pinhole size and \( L_{\text{sd}} \). When we choose collimator length \( L_{\text{cs}} \) as equal to \( L_{\text{sd}} \), the optimum ratios of the pinholes become

\[ A_0 : A_s : A_d = 2 : 1 : 4. \]  

(8)

Actually, \( \Delta Q \) of about 0.01 nm\(^{-1}\) can be attained using long-wave neutrons of 0.3–1.2 nm if we choose \( A_s = 2 \) mm and \( L_{\text{mc}} = L_{\text{sd}} = 1.5 \) m.

We can use 7–19 or more pinholes for the incident aperture that helps in recovering the intensity. Of course we face overlapping of small-angle scattering from different pinholes. Separating them is expected to be difficult in the case of a reactor instrument, but in case of time-of-flight instrument, it will be possible because we can get multiple \( I(q) \) information for the same \( q \) from different wavelengths and position regions. The possibility also exists of using a multi-pinhole aperture plate immediately in front of the sample.

4. Experiment

We tested the principle of the instrument type-2 at HUNS. It has an extremely efficient solid methane cold moderator, but the time-integrated neutron flux at sample position is rather low: a few times \( 10^3 \) n/cm\(^2\)/s.

![Figure 4](image)

**Figure 4.** Instrument layout of the test experiment.
immediately before the sample was 2 mm. The sample was a welded steel (SS400) sample as shown in figure 6, and the central part of the multi-pinhole pattern is placed at the welded position.

For the detector, we used a Li-6 loaded ZnS scintillator coupled with a position-sensitive photomultiplier tube (R3292; Hamamatsu Photonics K.K.)[6]. The detector resolution is less than 1 mm and the effective area is about 100 mm diameter. The whole system together with a data acquisition electronics based on VME modules and control software (LabView; National Instruments Corp.) was obtained from Japan Neutron Optics Inc.

Figure 7 shows measured direct beam patterns and SANS patterns for the welded steel sample. Enlarged images of the central parts of the images on the left columns are also shown in the right columns. The detector image is oriented as though one is looking from the back of the detector: it has 256 × 256 position channels with 0.45 mm/channel. Unfortunately, some image distortion is apparent towards the periphery of the detector image. Data are integrated over wavelengths of 0.35–0.62 nm.

Despite the lack of a vacuum chamber and despite the exposure to atmosphere in between the first pinhole to the detector, the direct beam profile shows little indication of diffuse tailing down to about 3.5 orders from the peak. Because of the pinhole camera geometry, the intensity distribution on the moderator surface directly reflects the intensity distribution of the pinhole images, but it is upside down and flipped left to right on a vertical axis. Although there were 19 pinholes, the top and bottom images at the central column are missing because the beams corresponding to the pinholes are at the outside edges of the moderator.

5. Position-dependent SANS using 1-mm-diameter pinholes

We also conducted a 1-mm-diameter pinhole experiment. The sample was also welded steel, but this time there was a heat-affected zone (HAZ) of about 1 mm width between the welded and non-welded parts. We put a multi-pinhole plate upstream that had two sets of nine pinholes and two pinholes immediately before the sample was 2 mm. The sample was a welded steel (SS400) sample as shown in figure 6, and the central part of the multi-pinhole pattern is placed at the welded position.

For the detector, we used a Li-6 loaded ZnS scintillator coupled with a position-sensitive photomultiplier tube (R3292; Hamamatsu Photonics K.K.)[6]. The detector resolution is less than 1 mm and the effective area is about 100 mm diameter. The whole system together with a data acquisition electronics based on VME modules and control software (LabView; National Instruments Corp.) was obtained from Japan Neutron Optics Inc.

Figure 7 shows measured direct beam patterns and SANS patterns for the welded steel sample. Enlarged images of the central parts of the images on the left columns are also shown in the right columns. The detector image is oriented as though one is looking from the back of the detector: it has 256 × 256 position channels with 0.45 mm/channel. Unfortunately, some image distortion is apparent towards the periphery of the detector image. Data are integrated over wavelengths of 0.35–0.62 nm.

Despite the lack of a vacuum chamber and despite the exposure to atmosphere in between the first pinhole to the detector, the direct beam profile shows little indication of diffuse tailing down to about 3.5 orders from the peak. Because of the pinhole camera geometry, the intensity distribution on the moderator surface directly reflects the intensity distribution of the pinhole images, but it is upside down and flipped left to right on a vertical axis. Although there were 19 pinholes, the top and bottom images at the central column are missing because the beams corresponding to the pinholes are at the outside edges of the moderator.

5. Position-dependent SANS using 1-mm-diameter pinholes

We also conducted a 1-mm-diameter pinhole experiment. The sample was also welded steel, but this time there was a heat-affected zone (HAZ) of about 1 mm width between the welded and non-welded parts. We put a multi-pinhole plate upstream that had two sets of nine pinholes and two pinholes immediately before the sample was 2 mm. The sample was a welded steel (SS400) sample as shown in figure 6, and the central part of the multi-pinhole pattern is placed at the welded position.

For the detector, we used a Li-6 loaded ZnS scintillator coupled with a position-sensitive photomultiplier tube (R3292; Hamamatsu Photonics K.K.)[6]. The detector resolution is less than 1 mm and the effective area is about 100 mm diameter. The whole system together with a data acquisition electronics based on VME modules and control software (LabView; National Instruments Corp.) was obtained from Japan Neutron Optics Inc.

Figure 7 shows measured direct beam patterns and SANS patterns for the welded steel sample. Enlarged images of the central parts of the images on the left columns are also shown in the right columns. The detector image is oriented as though one is looking from the back of the detector: it has 256 × 256 position channels with 0.45 mm/channel. Unfortunately, some image distortion is apparent towards the periphery of the detector image. Data are integrated over wavelengths of 0.35–0.62 nm.

Despite the lack of a vacuum chamber and despite the exposure to atmosphere in between the first pinhole to the detector, the direct beam profile shows little indication of diffuse tailing down to about 3.5 orders from the peak. Because of the pinhole camera geometry, the intensity distribution on the moderator surface directly reflects the intensity distribution of the pinhole images, but it is upside down and flipped left to right on a vertical axis. Although there were 19 pinholes, the top and bottom images at the central column are missing because the beams corresponding to the pinholes are at the outside edges of the moderator.

5. Position-dependent SANS using 1-mm-diameter pinholes

We also conducted a 1-mm-diameter pinhole experiment. The sample was also welded steel, but this time there was a heat-affected zone (HAZ) of about 1 mm width between the welded and non-welded parts. We put a multi-pinhole plate upstream that had two sets of nine pinholes and two pinholes immediately before the sample was 2 mm. The sample was a welded steel (SS400) sample as shown in figure 6, and the central part of the multi-pinhole pattern is placed at the welded position.

For the detector, we used a Li-6 loaded ZnS scintillator coupled with a position-sensitive photomultiplier tube (R3292; Hamamatsu Photonics K.K.)[6]. The detector resolution is less than 1 mm and the effective area is about 100 mm diameter. The whole system together with a data acquisition electronics based on VME modules and control software (LabView; National Instruments Corp.) was obtained from Japan Neutron Optics Inc.

Figure 7 shows measured direct beam patterns and SANS patterns for the welded steel sample. Enlarged images of the central parts of the images on the left columns are also shown in the right columns. The detector image is oriented as though one is looking from the back of the detector: it has 256 × 256 position channels with 0.45 mm/channel. Unfortunately, some image distortion is apparent towards the periphery of the detector image. Data are integrated over wavelengths of 0.35–0.62 nm.

Despite the lack of a vacuum chamber and despite the exposure to atmosphere in between the first pinhole to the detector, the direct beam profile shows little indication of diffuse tailing down to about 3.5 orders from the peak. Because of the pinhole camera geometry, the intensity distribution on the moderator surface directly reflects the intensity distribution of the pinhole images, but it is upside down and flipped left to right on a vertical axis. Although there were 19 pinholes, the top and bottom images at the central column are missing because the beams corresponding to the pinholes are at the outside edges of the moderator.
immediately before the sample. The direct-beam patterns are shown in figure 8. In figure 9, the pattern with the sample is shown. A strong SANS contribution is apparent. We are conducting analyses now.

6. Resolving overlapped scattering

We are now developing a method of resolving overlapping SANS from multiple pinholes, although it is in a preliminary stage. Figure 10 shows wavelength-dependent circular-averaged SANS patterns from a pinhole with wavelength regions of 0.28–0.35 nm, 0.31–0.39 nm, and 0.35–0.44 nm. In each figure, thin upper curves show SANS with the sample and lower ones without the sample. The red thick curves the background subtracted SANS patterns. Because of scattering from adjacent pinholes, upturn behavior is shown towards the higher-$Q$ region from about 0.025 nm$^{-1}$. With shorter wavelength regions, we assume that the data are reliable for higher-$Q$ regions. With longer wavelength regions, lower-$Q$ regions would be reliable. We combined data from good regions and obtained the SANS curve shown in figure 11. The upper three curves show scattering from welded parts and the lower three show that from non-welded parts. Although the analysis remains in a preliminary stage, it is apparent that welded parts produce higher scattering intensities than non-welded parts do.

Figure 8. Direct beam pattern from a couple of nine pinholes upstream and two 1-mm-diameter pinholes immediately in front of the sample.

Figure 9. SANS pattern from a couple of the nine pinholes upstream and two 1-mm-diameter pinholes immediately before the sample.

Figure 10. Wavelength-dependent SANS patterns from a pinhole with the wavelength regions of 0.28–0.35 nm, 0.31–0.39 nm, and 0.35–0.42 nm from left to right. The upper curves show that of the scattering with sample and the lower ones without the sample. The red curves show scattering corrected for the background. The thick green parts on the red curves show $Q$-ranges where the data are reliable with the selected wavelength ranges.
Acknowledgments
This work was partially supported by the Quantum Beam Technology Program of JST and (JSPS) KAKENHI (23226018).

References
[1] Bunk O, Bech M, Jensen TH, Feidenhans'l R, Binderup T, Menzel A, and Pfeiffer F 2009 New Journal of Physics 11 123016
[2] Fratzl P, Weinkamer R, 2007 Nature's hierarchical materials, Progress in Materials Science, 52 1263-1334
[3] Cedola A, Mastrogiacomo M, Lagomarsino S, Cancedda R, Giannini C, Guagliardi A, Ladisa M, Burghammer M, Rustichelli F, and Komlev V 2007 Spectrochimica Acta Part B: Atomic Spectroscopy, 62 642-647
[4] Dinapoli R, Bergamaschi A, Henrich B, Horisberger R, Johnson I, Mozzanica A, Schmid E, Schmitt B, Schreiber A, and Shi X 2010, Nucl. Instrum. and Meth. A, doi:10.1016/j.nima.2010.12.005 in press.
[5] Cancedda R, Cedola A, Giuliani A, Komlev V, Lagomarsino S, Mastrogiacomo M, and Peyrin F, Rustichelli F 2007 Biomaterials 28 2505-2524
[6] Hirota K, Shinohara T, Ikeda K, Mishima K, Adachi T, Morishima T, Satoh S, Oku T, Yamada S, Sasao H, Suzuki J-i, and Shimizu HM 2005 Phys. Chem. Chem. Phys. 7 1836-1838

Figure 11. Reconstructed SANS patterns in welded parts (upper three curves) and in non-welded parts (lower six curves).