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Detector development for a high-flux neutron reflectometer

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Abstract Neutron reflectometry is a powerful tool for investigating the surface and interfacial structures of various materials on the nanometer to submicrometer spatial range. We have been developing a horizontal-type neutron reflectometer, named SOFIA, at BL16 in J-PARC/MLF for investigating such interfacial structures, including those of free liquid surfaces. Utilizing the high-flux neutron beam at J-PARC, we have, until now, efficiently performed reflectivity measurements with SOFIA. However, the maximum count rate of the two-dimensional (2-D) detector currently in use will be insufficient for the full-power operation of J-PARC (1 MW) in the near future. In order to increase the count rate, we developed a new detector comprising 21 one-dimensional (1-D) detectors. 1-D detectors were used because neutron counts for a two-dimensional (2-D) detector can be reduced by dividing it into an array of 1-D detectors. In this study, we evaluated the performance of the newly developed detector and successfully carried out neutron reflectivity measurements using it.

1. Introduction

Neutron reflectometry is widely used for investigating surface structures and buried soft material interfaces. Beam line 16 (BL16) at Materials and Life Science Facility (MLF) in Japan Proton Accelerator Research Complex (J-PARC) is dedicated to a horizontal-type neutron reflectometer in which two downward neutron beams (2.22° and 5.71°) are transported from a coupled hydrogen moderator to irradiate a free surface such as the air–water interface. In order to measure neutron reflectivity by using the high-flux neutron beam at J-PARC/MLF as soon as possible, we utilized an old reflectometer, named ARISA [1], which was relocated from KENS facility to MLF in 2008. However, the range of movement of the slit, sample, and detector stages was limited and only the 2.22° beam line at BL16 was available for neutron reflectivity measurements, because the distance between the two downward beams (2.22° and 5.71°) was too large for ARISA. This is a serious disadvantage that prevented the use of the 5.71° beam line for measuring neutron reflectivity at air–liquid interfaces measurement with a high incident angle. To overcome this disadvantage, the components of ARISA were replaced by a new reflectometer—SOFt-Interface Analyzer (SOFIA)—in 2011 through a joint collaboration between JST/ERATO and KEK [2-4]. The slit, sample, and detector stages of SOFIA allowed us to utilize both the 5.71° and 2.22° beam lines. This, in turn, enabled us to measure neutron reflectivity over a wide momentum-transfer region on free liquid surfaces.

We employed a two-dimensional (2-D) photomultiplier tube equipped with a ^{6}LiF/ZnS scintillator as the detector for SOFIA [5]. Using this detector, we were able to measure specular reflection, off-specular reflection, and background at a time. Neutron signals were recorded by a NeuNET module [6] and stored as event data with time stamps inserted by a GateNET module. This recording system enabled us to change time slicing bins for kinetics measurement even when the original measurement had been completed. In addition, SOFIA provided the double frame mode (12.5 Hz operation) that extended the wavelength band up to 1.76 nm; this is significantly higher than the maximum wavelength available in the single frame mode (0.88 nm at 25 Hz operation). This setup of SOFIA reflectometer enabled us to efficiently perform kinetics measurements, and thus, numerous kinetics studies were carried out using this setup [7, 8].

However, this detector started showing pulse pile-up at around 10,000 cps (counts per second) because of scintillator afterglow. This count rate would have been insufficient for the full-power operation of J-PARC (1 MW) in the near future. In order to increase the count rate, we developed a new photon detector consisting of an opto-semiconductor device, multi-pixel photon counter (MPPC). Figure 1 (a) schematically illustrates the new detector comprising 21 one-dimensional (1-D) detectors and a readout module. The sensitive area of each detector was 5 mm in width and 128 mm in length, and the total detector area, including the areas of all 21 1-D detectors, was 105 mm × 128 mm. Since the neutron beam used for reflectivity measurements is collimated along one dimension but is very wide along the other dimension, neutron counts for a detector can be reduced by dividing a 2-D detector into an array of 1-D detectors. Figure 1 (b) shows the side view of a single 1-D detector. The neutron position along the length direction was evaluated by a charge division method, and the spatial resolution was about 1 mm. Detailed information is provided in our previous report [9].

In this study, we evaluated the performance of the new detector and carried out neutron reflectivity measurements using this detector with the SOFIA reflectometer.

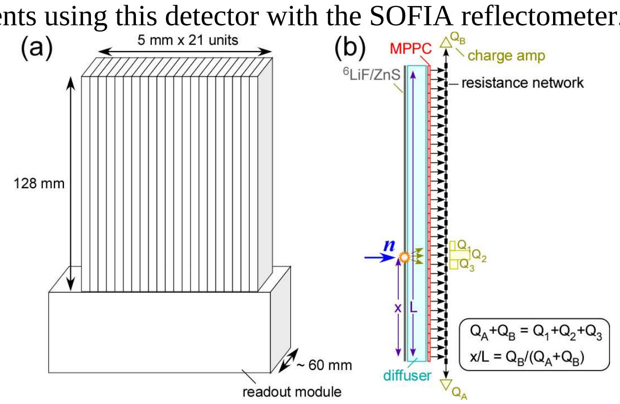

Figure 1. Schematic illustration of the new detector. (a) Dimensions of the 1-D detector array. (b) Side view of a single 1-D detector.

2. Performance and reflectivity measurements

To determine the position linearity and spatial resolution of the new detector, we performed a position scan with a beam width of 0.1 mm and a step of 4 mm. Figure 2 (a) shows the obtained beam profile for a single detector. The peak position and full width at half maximum (FWHM) were evaluated by multi-peak fitting performed using a Gaussian function. The position linearity translated very well, as shown in Figure 2 (b), and the slope was evaluated to be 0.183 mm/channel (total number of channels: 1024). Using this slope, we also evaluated the FWHM depending on the beam position, as shown in Figure 2 (c). The FWHM was about 1.4 and 2.0 mm at the center and edge of the detector, respectively; this resolution is sufficient for neutron reflectivity measurements.

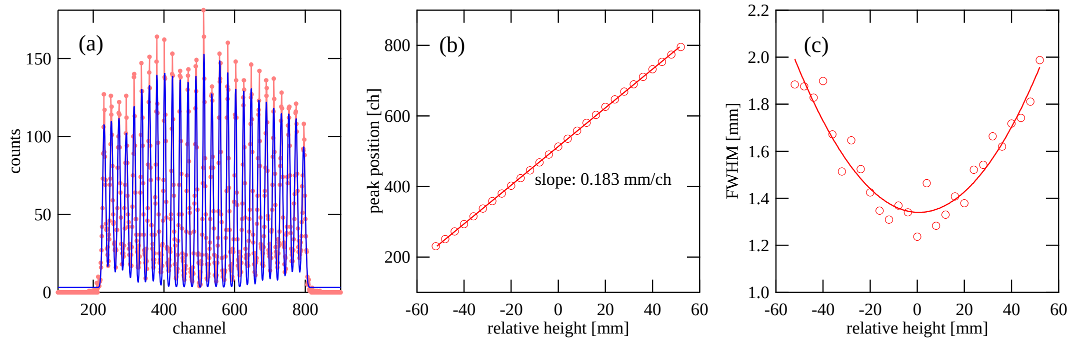

Figure 2. Results of a detection position scan for a single detector. (a) Beam profile and fitting

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results. (b) Peak position depending on the beam position. (c) FWHM depending on the beam position.

Next, we evaluated the homogeneity of the neutron detection sensitivity by directing a homogeneous beam on the detector. For this purpose, the neutron beam was first scattered by a poly(methyl methacrylate) plate and incoherent scattering from the plate was measured by the detector at a distance of 1.8 m from the plate. Figures 3 (a) and (b) show the obtained intensity map obtained for various detection positions and frequency distribution of the neutron count, respectively. The intensity map appears flat. Further, considering the standard deviation of the frequency distribution, it was concluded that about 10% of the average frequency was sufficient for use in reflectivity measurements.

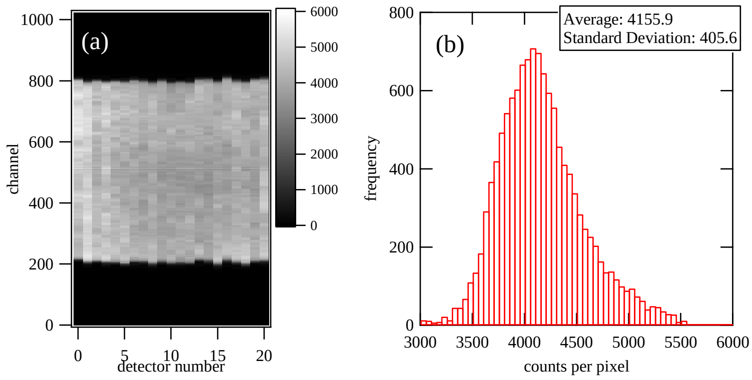

Figure 3. Results of homogeneity evaluation for neutron detection sensitivity. (a) Intensity map based on the detection position. (b) Frequency distribution of the neutron count.

Furthermore, we evaluated the count rate of the detector. A neutron beam was collimated by double slits and neutrons directly counted by the detector. The horizontal width of the slits was adjusted according to the beam width at a sample position of 40 mm (width at the detector was 55 mm). This means that in the case of the new detector, eleven 1-D detectors should work because the width of each 1-D detector was 5 mm. The vertical width of the downstream slit was fixed at 0.2 mm and that of the upstream slit was varied between 0.2 mm and 8.0 mm. Figures 4 (a) and (b) show the time-of-flight profiles for various beam collimations obtained using the new and old detector, respectively. All the profiles in Figure 4 (a) show a Maxwellian peak around 11 ms; however, the peak height was suppressed at 3.2 mm and completely disappeared at 6.4 mm in Figure 4 (b). This indicates that the maximum count rate of the new detector was considerably higher than that of the old one. Figure 4 (c) shows the count rate of the new detector at the Maxwellian peak as a function of the slit width. For low slit widths, the count rate is proportional to the slit width. With increasing slit width, however, the count rate lessens than that indicated by the fit line around 40,000 cps because of counting loss. Although the eleven detectors worked, this count rate was only four times that of the old detector. The region at the detector that exhibited high flux was 20 mm in width. This is the reason for the maximum count rate being limited to 40,000 cps, and the count rate in reflectivity measurements should be maintained at a value less than this.

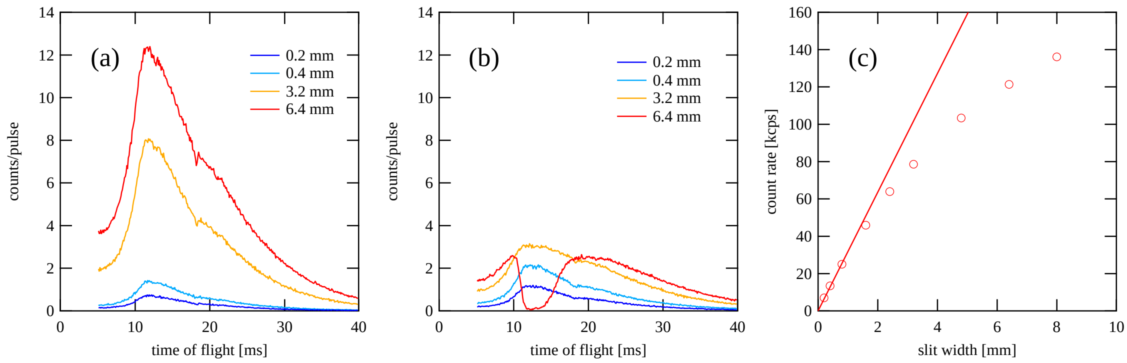

Figure 4. Results of count rate evaluation. (a) Time-of-flight (TOF) profile obtained using the new detector. (b) TOF profile obtained using the old detector. (c) Count rate of the new detector at the Maxwellian peak with varying slit width.

Finally, we performed reflectivity measurements using the new detector on a 65-nm-thick deuterated polystyrene thin film coated on a silicon wafer. Figure 5 (a) shows the TOF profiles for a direct beam and the reflection at an incident angle of 0.6°. The reflection intensity modulates for up to 30 ms owing to interference from the polystyrene film, and overlaps with that of the direct beam above 30 ms because of total reflection at the film surface. To compare the obtained reflectivity data with the measured data obtained using the old detector, the reflection intensity was normalized by the direct beam profile. Figure 5 (b) shows the reflectivity profile depending on the momentum transfer normal to the substrate, Q_z. The profile obtained using the new detector agreed well with that obtained using the old detector. This is direct evidence that the new detector is suitable for neutron reflectivity measurements.

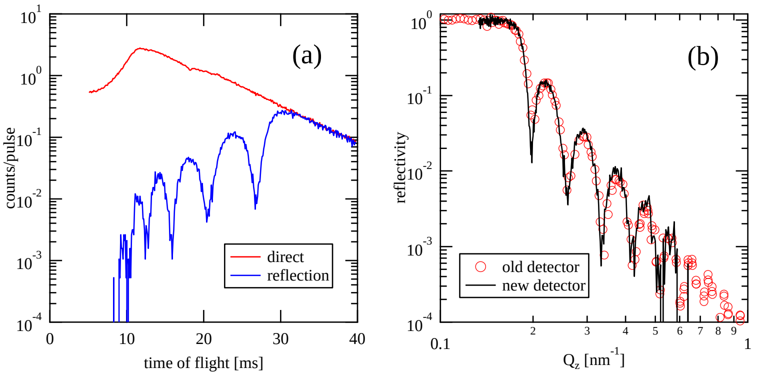

Figure 5. Reflectivity measurements for a deuterated polystyrene thin film performed using the new detector. (a) TOF profiles for a direct beam and reflection at an incident angle of 0.6°. (b) Reflectivity profile depending on momentum transfer normal to the substrate, Q_z.

3. Conclusion

We developed an array of one-dimensional, position-sensitive detectors for use with a neutron reflectometer, SOFIA, at J-PARC. The FWHM of the detector was about 1.4 mm at the center, and the homogeneity of neutron sensitivity was about 10%. These values are sufficient for using the detector for neutron reflectivity measurements. Remarkably, the maximum count rate of the new detector was four times that of the old detector used with SOFIA. This improved count rate is insufficient to accept a direct beam with a wide slit width, but it is enough for time-slicing reflectivity measurements with a short accumulation time performed using the high-flux neutron beam operating at 1 MW in J-PARC. Furthermore, we used the new detector for performing reflectivity measurements and confirmed that the detector is suitable for this purpose. Installation of the new detector in SOFIA has been planned in the near future.

The neutron scattering experiment was approved by the Neutron Scattering Program Advisory Committee of IMSS, KEK (Proposal No. 2009S08).

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