Design of neutron reflectometer in the angle-resolved dispersive mode for compact accelerator based neutron sources

Y. Yamashita, S. Tasaki*, Y. Abe, K. Hironaka

Development of Nuclear Engineering, Kyoto University, Japan

Abstract

Compact accelerator-driven neutron sources take advantage of their readiness of operation and high adaptability to various experiments, although their neutron flux is low. We are now working on installing a reflectometer to Kyoto University Accelerator based Neutron Source (KUANS). Neutron reflectometer (NR) is one of the most simple but powerful spectrometer, in which neutron reflectivity from the surface of a sample is measured and analyzed to give so-called ‘depth profile’ of the sample, which represents the distribution of materials along the surface normal direction. Usually, reflectometer requires narrow well-collimated beam. For the present reflectometer, however, incident neutron beam is narrowed at the sample position but not so much collimated to allow variety of incident angle to the sample simultaneously assuming the specular reflection at the sample surface. This multi-incident angle measurement helps to compensate weak intensity of KUANS. The motivation of this study is development of the NR with the readiness of operation for a preliminary experiment. In this study, the design of the NR is presented and the reflectivity is estimated via Monte-Carlo simulation. It was found that measurable lowest reflectivity is down to 10⁻².

Keywords: reflectometer; depth profile; wave-vector transfer.

1. Introduction

Neutron reflectometer (NR) is one of the most simple but powerful spectrometer, in which neutron reflectivity from the surface of a flat sample is measured and analyzed to give so-called ‘depth profile’ of

* Corresponding author. Tel.: +81-75-383-3912; fax: +81-75-383-3912.
E-mail address: tasaki@nucleng.kyoto-u.ac.jp.
the sample, which represents the distribution of materials along the surface normal direction. In this method, the structure of thin films and multilayer film is inspected non-destructively. The technique accesses relatively small values of the wave-vector transfer $Q (= k_f - k_i$, equals to $(4\pi\sin\theta)/\lambda$ in the specular condition), corresponding to spatial length scales down to the nanometer range [1]. Neutron reflectivity is analyzed for each $Q$ that is determined by $\lambda$ and $\theta$, where $\theta$ and $\lambda$ are incident glancing angle and wavelength of neutrons, respectively. $\lambda$ is determined by Time-Of-Flight (TOF) $t$ and $\theta$ is obtained from neutron detecting position. It is necessary that the resolution of $\theta$ and $t$ is reduced in order to obtain reflectivity of high precision.

We are now working on installing a NR to Kyoto University Accelerator based Neutron Source (KUANS). Neutron intensity of KUANS is 3300 cps/cm² at the position 130 cm away from surface of the moderator for average proton current of 31 $\mu$A, pulse width of 100 $\mu$s and pulse repetition rate of 100 Hz. In such compact accelerator based neutron source of low intensity and limit of short flight-path (~ 300 cm at KUANS), NR normally requires a narrow well-collimated beam neutron beam, is not suitable because of its low neutron flux. In the preset NR, however, the neutron beam incident to the sample has relatively large divergence to allow variety of incident angle simultaneously. This multi-angle incident measurement helps to compensate weak intensity of KUANS. We adopted the RPMT system composed of ZnS scintillator, PSPMT (position sensitive PMT, Hamamatsu Photonics Co., the spatial resolution is 0.2325 mm [2]) and the electronics as a detector. RPMT system allows us to get the detected position (corresponding to the reflection angle $\theta$) and the time of flight of a neutron $t$ can be measured at the same time. Analyzing reflected neutron intensity $I(\theta, t)$, the reflectivity of the sample is obtained. The advantage of this NR is that measuring time can be reduced to use simultaneously the neutrons with different $\theta$ and $t$.

In this paper, the reflectivity measured with this NR is evaluated using Monte-Carlo simulation and the resolution of $Q$ and the accuracy of the reflectivity is evaluated.

2. Neutron reflectometer

In the developed NR, incident neutron beam is not so much collimated as shown in Fig. 1 in order to compensate low neutron flux. A neutron beam line (cross section of 10 cm wide × 10 cm high) transports the neutrons from the polyethylene moderator to the instrument. Neutrons emitted from the moderator at leftmost of Fig.1 are collimated with Slit1 placed at 1 m away from surface of moderator and irradiate the sample. Slit1 as well as Slit2 in front of the detector, is composed of 1mm Cadmium and 5 mm B₄C rubber. The dimension of Slit1 is 1 mm (width) × 60 mm in order to limit the neutron beam to the sample. Neutrons reflected at the sample are separated from incident beam and detected by the detector placed at 1 m away from the sample. Assuming that only specular reflection occurs at the sample surface, the reflection and incident angle $\theta$ is equal. Therefore using the RPMT system, we obtain the reflection angle $\theta$ from the detected position and the neutron wavelength $\lambda$ from the time of flight $t$, and hence momentum transfer $Q$ of each neutron. The spatial resolution of the RPMT system is about 0.5 mm estimated experimentally[3]. $\theta$ is limited from 5 to 10 mrad by Slit2 in front of the detector. The time resolution $\Delta t$ of this NR is determined by the channel width $\Delta t_{ch}$ of time analyzer (1 $\mu$s) and the proton pulse width $\Delta t_p$ which is changeable from 20 to 120 $\mu$s. Neutron reflectivity is obtained by the division of the reflected beam spectrum by the direct beam spectrum and the multiplication of the normalization factor obtained by the ratio of either neutron monitor counts or the reflectivity for neutrons with long wavelength. Fitting the reflectivity calculated from the model potential to the measured reflectivity, the depth profile of the sample can be obtained.
3. Simulation of neutron reflectometer

3.1. Simulation conditions

Performance evaluation of this NR is carried out using Monte Carlo code McStas [4]. Overview of set-up is shown in Fig. 1 and the simulation conditions are shown in Table 1. Neutron spectrum was assumed to be Maxwellian distribution with the temperature of 350 K. Fast neutrons were not considered in this simulation because the detector efficiency is quite low for the fast neutron region. 21 wavelength monitor components arranged at 0.5 mm intervals are employed as detectors. $\Delta Q$ resolution is represented as

$$ (\Delta Q/Q)^2 = (\Delta t/t)^2 + (\Delta \theta/\theta)^2 $$

where $t$, $\Delta t$, $\theta$ and $\Delta \theta$ are the TOF, the TOF resolution, the reflection angle and the reflection angle resolution, respectively. $\Delta t$ is affected by the TOF channel width $\Delta t_{ch}$ and the neutron pulse width $\Delta t_n$ which is represented as a neutron pulse function convoluted the proton pulse width to Ikeda-Carpenter function. And $\Delta \theta$ is the angle resolution due to the spacial resolution of the RPMT $\theta_D$ and the sample size $\theta_s$. In the present simulation, $\Delta \theta_D/\theta$ and $\Delta \theta_s/\theta$ is 3~10% and 6%. $\Delta t$ was set to be 80 $\mu$s which is a typical proton width for KUANS. Measuring time is assumed to be 3 hours to bet sufficient statistics.

In the simulation, we adopt as a sample 3Q-supermirror, whose the critical angle of total reflection is 3 times as large as that of Ni-mirror, in order to estimate the measured reflectivity in a wide range of $Q$. The reflectivity is represented as the following equation:

$$ R = \begin{cases} R_0 \{1 - \tanh[(Q - mQ_c)/W]\} \{1 - \alpha(Q - Q_c)\} & Q \leq Q_c \\ 0.5R_0 \{1 - \tanh[(Q - mQ_c)/W]\} \{1 - \alpha(Q - Q_c)\} & Q > Q_c \end{cases} $$

where $R_o=0.99$, $m=3$, $W=0.003$ Å$^{-1}$, $\alpha=6.1$ Å and $Q_c$ is the critical wave-vector transfer for Ni 0.021 Å$^{-1}$. The value of $\alpha$ and $W$ give the gradient of reflectivity between $Q_c$ and $mQ_c$, and around $mQ_c$, respectively. Lowering of the reflectivity starts at $Q>Q_c$ and it decreases rapidly at $Q=mQ_c$. When $Q=mQ_c$, $\lambda=\lambda_m$ and critical wavelength for the supermirror $\lambda_m$ is given by

$$ \lambda_m = \lambda_c/m = 4\pi sin\theta/mQ_c $$

where $\lambda_c$ is the critical wavelength for Ni.
### Table 1. Calculation conditions.

| Description                                      | Value                  |
|--------------------------------------------------|------------------------|
| Neutron intensity on front of slit1              | 3300 cps/cm²           |
| Temperature of Maxwellian distribution           | 350 K                  |
| History                                          | $10^{10}$              |
| Beam line cross section                          | $10.0 \times 10.0$ cm  |
| Slit1 width and height                           | 1.0 mm width $\times$ 60.0 mm height |
| Slit2 width and height                           | 10.0 mm width $\times$ 60.0 mm height |
| Flight path from the moderator to the detector   | 230.0 cm               |
| Neutron detector                                 | Wavelength monitor component |
| Detectors size                                   | 0.50 mm width $\times$ 60.0 mm height |
| Number of detectors                              | 21                     |
| Sample                                           | 3Q-Supermirror         |
| Sample size                                      | 6.0 cm square          |
| Angle of inclination from the vertical plane of the sample | 5.0 mrad               |
| Reflection angle                                 | $\theta = 5.0$~$15.0$ mrad |
| Resolution of reflection angle due to the spatial resolution of the detector | $\Delta \theta_s = 0.50$ mrad |
| Resolution of reflection angle due to the sample size | $\Delta \theta_r = 6.0\%$ |
| Resolution of TOF channel width                  | $\Delta t_{ch} = 80$ $\mu$s |
| Detection efficiency of neutrons                 | 100%                   |

#### 3.1. Results and discussions

Spectrum of the incident beam (solid line) to the supermirror and the reflected beam to the detectors (dotted lines) corresponding to the measurement for 3 hours are shown in Fig. 2. The intensity of the incident neutrons to the sample is about 1800 cps and this spectrum follows the Maxwellian distribution. The total intensity of the reflected neutrons to the detectors is 12.4 cps and this reflected spectrum also follows the Maxwellian distribution when $\lambda > \lambda_m$. This reflected spectrum shows clearly the critical wavelength $\lambda_m$ varies as $\theta$ changes. The statistical error of the reflected neutron of each reflection angle is less than 5% in the vicinity of $\lambda_m$.

The reflectivity is obtained the ratio the reflected intensity of the incident intensity multiplied by the normalizing factor and represented as

$$R(\lambda, \theta) = \left[\frac{I_{ref}(\lambda, \theta)}{I_{inc}(\lambda, \theta)}\right] \times \left[\frac{I_{inc}(\theta)}{I_{ref}(\theta)}\right]$$  \hspace{1cm} (4)

where $I_{inc}$ is the intensity of incident beam, $I_{ref}$ the intensity of reflected beam. The normalization factor, which the reflectivity for neutrons with long wavelength is unity, is calculated by integrating neutron intensity of the incident and reflected beam in $\lambda > \lambda_c$. And $\hat{I}(\theta)$ is given as

$$\hat{I}(\theta) = \int_{\lambda_c}^{\infty} I(\lambda, \theta) d\lambda$$  \hspace{1cm} (5)
The reflectivity $R(Q)$ as a function of $Q$ in linear and log scale are shown in upper and lower figure in Fig.3. $I_{ref}(\theta, \lambda)$ having over 25 counts for 3 hours is used in order to obtain $R(Q)$. The reflectivity could be measured down to $10^{-2}$ with this NR. The counting error of the reflectivity is about $5\sim20\%$ in $10^{-2}$ range.

$\Delta Q/Q$ for minimum and maximum reflection angle at $\lambda_m$ are shown in Table2 and $\Delta t/t$ and $\Delta \theta/\theta$ are the same degree. $\Delta Q/Q$ differs significantly depending on the angle of reflection. It is also shown in Fig. 3 that $\Delta Q/Q$, which is represented by the horizontal bars of the several experimental points, at a low-angle is much larger than those at a high-angle. The length of the bar is equal to $\Delta Q$ and makes the precision of the reflectivity worse. The change of the simulated reflectivity is moderated comparing to that of the model reflectivity because of smoothing effect of large $\Delta Q/Q$.

Fig. 2. The number of incident neutrons and reflected neutrons with each reflection angle.

Fig. 3. Measured reflectivity of 3Q-Supermirror in linear and log scale.
The reflectivity curve of the supermirror is obtained from the least-square fitting to Eq. (2) with the values $\alpha_f$, $W_f$ and $m_f$ as fit parameters. The fit parameters and the model value $\alpha_0$, $W_0$ and $m_0$ are presented in Table 3. $\alpha_f$ and $W_f$ are well reproduced within the error of less than 5%. There is however discrepancy of about 50% between model and reproduced values because it is easily affected by $\Delta Q$. $\sqrt{W^2 - (\Delta Q/2)^2}$ is very close to $W_0$, provided that and $Q = 0.007 \, \text{Å}^{-1}$. If we remove the blurring effect due to $\Delta Q$, $W$ is reproduced perfectly.

The number of the reflected neutrons is larger at a low-angle than at a high-angle (see Fig. 2). Therefore at a low-angle, the spatial resolution can be small without increasing the measurement time despite the decreased number of detected neutrons. It is desirable that the spatial resolution is changed according to the reflection angle so that $\Delta \theta/\theta$ is constant. And more accurately measurement of the low reflectivity at a constant angle resolution is expected.

Table 3. The fit parameters and the model values.

| $\alpha$ [Å] | $W$ [Å⁻¹] | $m$ [-] | $\alpha_0$ [Å] | $W_0$ [Å⁻¹] | $m_0$ [-] |
|-------------|--------|------|-------------|--------|-----|
| 5.8±0.4     | 0.0046±0.0001 | 2.98±0.01 | 6.1        | 0.003  | 3.0 |

4. Conclusion

In this work we have designed the neutron reflectometer for KUANS. The concept of this NR is to make TOF measurement for various incident angles simultaneously. In order to realize such measurement, incident neutron beam is collimated 1.0×60 mm and the position sensitive detector is employed. It was found that the reflectivity of 3Q-Supermirror can be measured down to order of 10⁻² with the present NR. It is possible to get proper reflectivity down to 10⁻² under suitable consideration of the instrumental resolution.

References

[1] M. P. Seah, S. J. Spencer, F. Bensebaa et al., Surf. Interface Anal. 36 (2004) 1269.
[2] S. Satoh and S. Muto, Nucl. Instrum. Methods A (2013).
[3] Yoshinaga, private communication.
[4] McStas project website at (http://www.mcstas.org).
[5] K. Skold and D. L. Price, Methods of Experimental Physics, Vol. 23 Neutron Scattering, Part A, Academic Press, New York (1986).
[6] S. Ikeda and J. M. Carpenter, Nucl. Instrum. Methods A 239 (1985) 536.
[7] O. Schaerpf, Physica B 156-157 (1989) 631.
[8] O. Schaerpf, Physica B 156-157 (1989) 639.
[9] O. Schaerpf and N. Stuesser, Nucl. Instrum. Methods A 284 (1989) 208.