Current Status of a New Polarized Neutron Reflectometer at the Intense Pulsed Neutron Source of the Materials and Life Science Experimental Facility (MLF) of J-PARC

Takeda Masayasu,1,2,3,* Yamazaki Dai,1,2 Soyama Kazuhiko,1,2,3 Maruyama Ryuji,2 Hayashida Hiroshi,2 Asaoka Hidehito,1,2,4 Yamazaki Tatsuya,1 Kubota Masato,1,2 Aizawa Kazuya,2 Arai Masatoshi,2 Inamura Yasuhiro,2 Itoh Takayoshi,5 Kaneko Koji,1,2,3 Nakamura Tatsuya,2 Nakatani Takeshi,2 Oikawa Kenichi,2 Ohhara Takashi,1 Sakaguchi Yoshifumi,1 Sakasai Kaoru,2 Shinohara Takena,2 Suzuki Junichi,1 Suzuki Kentaro,2 Tamura Itaru,2,3 Toh Kentaro,2 Yamagishi Hideshi,2 Yoshida Noboru,5 and Hirano Tatsumi6

1 Quantum Beam Science Directorate, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
2 J-PARC Center, Tokai, Ibaraki 319-1195, Japan
3 Department of Research Reactor and Tandem Accelerator, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
4 Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
5 Research Center for Neutron Science and Technology, Comprehensive Research Organization for Science and Society, Tokai, Ibaraki 319-1106, Japan
6 Hitachi Research Laboratory, Hitachi Ltd., Hitachi, Ibaraki 319-1292, Japan

(Received December 6, 2011)

The construction of a new polarized neutron reflectometer is now in progress at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). MLF has the world’s brightest pulsed neutron and muon sources (JSNS and MUSE). The user program of MLF has been already started in 2008, and now nine neutron and two muon spectrometers are in operation. Installation of the new reflectometer was expected to be completed in March 2011. However, the construction was interrupted by the massive earthquake hitting northeast Japan, including Tokai-mura where J-PARC is located. We expect to restart the user program of the new polarized neutron reflectometer at the beginning of next year (2012).

PACS numbers: 61.05.fj, 07.60.Hv, 68.65.Ac, 68.35.Ct

I. INTRODUCTION

Polarized neutron reflectometry is a very powerful and non-destructive tool for the structural investigation of magnetic multilayers, which are important in high-density magnetic recording devices and spintronics [1]. A neutron reflectometer is a type of spectrometer which measures the depth profile of the refractive indexes of layered materials, using neu-

*Electronic address: takeda.masayasu@jaea.go.jp

tron optical effects. A neutron behaves like visible light when it impinges on a surface with a shallow angle, and this phenomenon is called neutron optics. In the framework of neutron optics, a neutron is regarded as a plane wave, and it reflects and refracts at the interface, or the boundary between two different media. The reflected waves from every interface are superimposed outside the sample and interfere with each other according to the phase differences. The phase of each reflected plane wave is governed by the refractive index and the thickness of the layer. The refractive index is uniquely determined by the density, nuclear scattering length, and magnetic scattering length of each layer. The important point is that the refractive index is dependent on the neutron spin-state, which is parallel or antiparallel to the sample magnetization. The advantage of using polarized neutrons is that the sensitivity of the magnetization can be enhanced. In addition, the depth profile of the in-plane components of the magnetization vector can be decisively obtained using polarized neutron reflectometry.

II. NEW NEUTRON REFLECTOMETER AT JAPAN PROTON ACCELERATOR RESEARCH COMPLEX (J-PARC)

J-PARC is an acronym for Japan Proton Accelerator Research Complex; it is located at the Tokai campus of the Japan Atomic Energy Agency (JAEA), 130 km northeast of Tokyo [2]. J-PARC consists of four facilities: the Accelerators, the Hadron Experimental Facility, the Neutrino Experimental Facility, and the Materials and Life Science Experimental Facility (MLF). MLF has an intense pulsed neutron source, which is placed in a 50-GeV main ring (MR) in J-PARC [3]. The first neutron extraction was successfully performed in 2007, and the user program started in 2008, using several neutron spectrometers. The MLF building is 120 m long, 70 m wide, and 32 m in height. There is an intense and efficient pulsed neutron source inside the MLF building. The neutron target is circulating mercury, where proton beams of up to 1 MW are injected. There are 23 beam holes, or beam lines, and most of them are already occupied by spectrometers. Nine of them are in operation, three are at the commissioning stage, five are now under construction, and the remaining six are undecided.

At the end of 2009, the Japanese government decided to construct a second neutron reflectometer at MLF by allocating a supplementary budget in the 2009 stimulus package. We already have our first neutron reflectometer, SOPHIA (formerly ARISA-II), at beam line 16 (BL16) in the second experimental hall of MLF [4]. BL16 is a special beam line which is optimized to extract the neutron beam downwards, and SOPHIA is capable of measuring the reflectivities from a free surface and a free interface with a specially designed beam line. We have therefore decided to focus on solid-state physics, particularly on magnetism, in designing a new reflectometer as the second neutron reflectometer at MLF.
III. BASIC DESIGN OF THE NEW REFLECTOMETER

The new reflectometer is installed at BL17 in the second experimental hall of MLF. Next to this beam line, there is SOPHIA, at BL16, which is the first reflectometer at MLF. The key parameters of the new reflectometer are listed in Table 1.

### TABLE I: Specifications of the new polarized neutron reflectometer installed at BL17 of MLF

<table>
<thead>
<tr>
<th>1. Beam line</th>
<th>BL17</th>
</tr>
</thead>
<tbody>
<tr>
<td>2. Total flight-path</td>
<td>18 m (moderator–sample 15.5 m, sample–detector 2.5 m)</td>
</tr>
<tr>
<td>3. Moderator</td>
<td>coupled hydrogen moderator</td>
</tr>
<tr>
<td>4. Wavelength band in the first time-of-flight frame</td>
<td>(\lambda_g) 0.2–0.84 nm (polarized mode), 0.1–0.88 nm (unpolarized mode)</td>
</tr>
<tr>
<td>5. (q)-range</td>
<td>(q = 0.1–12\ \text{nm}^{-1}) (polarized mode), 0.05–25 nm(^{-1}) (unpolarized mode)</td>
</tr>
<tr>
<td>6. Scattering-angle range</td>
<td>(2\theta = -5^\circ) to (20^\circ)</td>
</tr>
</tbody>
</table>
| 7. Detectors        | zero-dimensional \(^3\)He gas detector  
                        two-dimensional multi-wire proportional counter  
                        (sensitive area \(128 \times 128\ \text{mm}^2\))  
                        two-dimensional scintillator detectors  
                        (sensitive area \(256 \times 256\ \text{mm}^2\)) |

This reflectometer sees a coupled moderator, and mainly uses cold neutrons. The distance between the surface of the coupled moderator and the sample position is 15.5 m, and the length of the \(2\theta\) arm is 2.5 m. The total length of the flight path of the neutrons is 18 m. The repetition rate of the 3-GeV synchrotron is 25 Hz. The combination of the total flight-path and the repetition rate enables us to use neutrons in the wavelength band between 0.1 and 0.88 nm in the first time-of-flight (TOF) frame. The maximum wavelength can be extended to 2.2 nm using the second TOF frame. The wavelength band narrows when polarized neutrons are used because of the limited effective bandwidth of a polarizing supermirror. The scattering angle can go up to \(20^\circ\). Consequently, the covered \(q\)-range is between 0.05 and 25 nm\(^{-1}\) in the case of specular reflectivity measurements using the first TOF frame. Here, \(q\) is the magnitude of the scattering vector in the scattering plane.

A T0 fast chopper eliminates burst neutrons, and three disk choppers are also installed on the beam line to avoid frame overlapping effects up to the ninth TOF frame, as shown in Fig. 1. Between the two disk choppers, there is a changer which can easily switch the neutron polarizer for reflectivity measurements using polarized neutrons to a simple collimator for reflectivity measurements using unpolarized neutrons, and vice versa. There are two spin-flippers: one is for incoming neutrons to the sample, and is set before the sample, and the other is for the reflected neutrons after the sample. The reflectometer has a sample table, several beam slits, and detectors at the very end of the reflectometer. As described later, it has four different kinds of neutron-detectors.
IV. DEVICES FOR POLARIZED NEUTRONS

The polarizing devices and components are very important for a polarized neutron reflectometer. The key components are a polarizer, an analyzer, and a spin-flipper. There are several issues in choosing the method for polarizing and analyzing neutron spin in a broad wavelength band. The most feasible ways of polarizing and analyzing the spins of pulsed neutrons are using a polarizing supermirror or using a $^3$He spin filter. Each method has both advantages and disadvantages. We decided to use a polarizing Fe/Si ($m = 4$) supermirror as the polarizer and analyzer because of their ease of operation and because they can be purchased commercially. We use the supermirror in a double-transmission geometry to avoid changing the beam line between measurements using polarized neutrons and those using unpolarized neutrons.

The analyzer is composed of stacking mirrors in the single-reflection geometry. In the case of the analyzer, the available space is restricted, so we had to make it as compact as possible. We estimated the figures of merit in two cases, i.e., reflection geometry and transmission geometry, and finally found that the reflection geometry is preferable. However, it is clear that we need a metastable exchange optical pumping (MEOP) $^3$He spin filter system for analysis of the reflected neutrons.

Fig. 2 is a schematic illustration of the polarizer and the analyzer. The total length of the polarizer is 1500 mm, and the maximum beam size is 60 mm (height) by 10 mm (width). The available polarized neutron wavelength band is from 0.21 to 0.71 nm. The size of the analyzer is compact in comparison with that of the polarizer. This combination of polarizer and analyzer enables us to measure the polarized neutron reflectivity with full polarization analysis (the measurement of four kinds of reflectivity, i.e., $R_{++}$, $R_{+-}$, $R_{-+}$, and $R_{--}$) in the wavelength band between 0.21 and 0.64 nm. Here, $R$ is the polarized
neutron reflectivity, and the first sign in the suffix indicates the spin-state of the incident neutrons, and the second one the spin-state of the reflected neutrons.

FIG. 2: Schematic representations of (a) neutron polarizer and (b) analyzer. Approximate scales are indicated.

We also spent time discussing the best spin-flipper for incident and reflected neutrons in different kinds of reflectivity measurements, namely specular reflectivity measurements and off-specular reflectivity measurements. It was important to take the significant influence of a large leakage of magnetic flux from the superconducting magnet on the flipper into account. Finally, we decided on the current design of the polarizing and analyzing system; this is shown in Fig. 1. A non-adiabatic two-coil spin-flipper is used for flipping the spin of incident neutrons. We use two kinds of spin-flippers for the reflected neutrons. Another non-adiabatic two-coil spin-flipper is used in the specular reflectivity measurements, and the so-called Mezei spin-flipper is used for off-specular reflectivity measurements. The Mezei flipper makes use of the Larmor precession to flip the neutron spins. The flipper has two coils: a precession coil, and a compensation coil. The condition of 180°-rotation of spin (spin-flip) using the Larmor precession differs for neutrons with different wavelengths. The current in the precession coil is periodically changed so that the magnetic field is inversely proportional to the wavelength of the pulsed neutrons.
V. DETECTORS

The supplementary budget asked for this neutron reflectometer to be available for a variety of surface science applications. We therefore planned to equip the new reflectometer with the capability of performing grazing incidence small-angle neutron scattering (GISANS) and the grazing incidence diffraction (GID) measurements. Consequently, we decided to install four different kinds of detectors to cover a wide \( q \)-range with an appropriate \( q \)-resolution. The detectors are listed in Table 2. One is an orthodox \(^3\)He gas tube detector without special resolution, and the other three are two-dimensional position-sensitive-detectors (2D-PSDs). Here, we focus on the 2D-PSDs.

<table>
<thead>
<tr>
<th>Detector Type</th>
<th>Effective area</th>
<th>Spatial resolution</th>
<th>Measurement type</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>MWPC</td>
<td>2D 128 \times 128 \text{mm}^2</td>
<td>2.0 \times 2.0 \text{mm}^2</td>
<td>specular and off-specular individual read-out reflectivities</td>
<td></td>
</tr>
<tr>
<td>WLSF</td>
<td>2D 256 \times 256 \text{mm}^2</td>
<td>4.0 \times 4.0 \text{mm}^2</td>
<td>off-specular reflectivity (^{10})B(_2)O(_3) + ZnS and GISANS</td>
<td></td>
</tr>
<tr>
<td>PS-PMT</td>
<td>2D \phi 100 \text{mm}</td>
<td>0.5 \times 0.5 \text{mm}^2</td>
<td>GID (^{10})B(_2)O(_3) + ZnS</td>
<td></td>
</tr>
<tr>
<td>Gas tube</td>
<td>0D 150 \times 25.4 \text{mm}^2</td>
<td>NA</td>
<td>specular reflectivity (^3)He gas (7 atm)</td>
<td></td>
</tr>
</tbody>
</table>

The first is a multiwire proportional neutron counter (MWPC). This detector has better spatial resolution than other detectors have, a high counting-rate, and a high efficiency. However, it is difficult to make a large detector of this type. We therefore use two other types of 2D-PSD. One is a 2D scintillator detector using a wavelength-shifting-fiber (WLSF) readout. Its spatial resolution is worse than that of the MWPC but it is easy to make such a detector with a large effective-area. In our case, the effective area is 256 mm \( \times \) 256 mm, with a 4 mm \( \times \) 4 mm spatial resolution. The other is a scintillator detector based on a position-sensitive photomultiplier. The effective area is 100 mm\( \phi \), with a 0.5 mm \( \times \) 0.5 mm spatial resolution.

We combined these three detectors to cover a wide \( q \)-region, as shown in Fig. 3. In this paper, the three components of the \( q \)-vector are defined as

\[
\begin{align*}
q_x &= \frac{2\pi}{\lambda} (\sin \theta_f + \sin \theta_i) \\
q_y &= \frac{2\pi}{\lambda} \sin \theta_f \cos \theta_s \\
q_z &= \frac{2\pi}{\lambda} (\cos \theta_f \cos \theta_s - \cos \theta_i)
\end{align*}
\] (1)

Here, \( q_x \) is the specular component, \( q_y \) is the GISANS component, \( q_z \) is the off-specular component of the scattering vector, \( \theta_i \) is the glancing angle, and the relations among \( \theta_i \), \( \theta_s \),...
FIG. 3: Illustration of setup for reflectivity measurements using three two-dimensional position sensitive detectors: one multi-wire proportional counter (MWPC), and two wavelength-shifting-fiber scintillation detectors (WLSF).

and $\theta_f$ are illustrated in Fig. 4(a). An MWPC with a high spatial-resolution of 2 mm is located at the center of a detector bank. The $q$-range shown in Fig. 4(b) can be measured in the off-specular reflectivity measurements using the MWPC only. Two large-area WLSF scintillation detectors with a medium spatial resolution of 4 mm are mounted above and on the left side of the MWPC from the viewpoint of the neutron target. The condition of the specular reflection is satisfied along the line $q_y = q_z = 0$. This combination of detectors can cover a wide $q$-region in GISANS measurements, as shown in Fig. 4(c). This area is dependent on the glancing angle. The glancing angle used in calculating the area in Fig. 4(c) is 0.4°.

VI. DATA ACQUISITION SYSTEM, DATA STORAGE, AND SOFTWARE

MLF has its own standard data acquisition (DAQ) system [3, 5], and this new reflectometer uses this DAQ system. The data storage format used at MLF is unique. Signals are recorded event-by-event using a clock; this enables us to know the history of the neutrons from production to detection. We call this an event data format. In this format, each neutron is given its own ID, and every neutron has its own event data. The event data contain the time of neutron production at the target, the time detected by the detector, the detector ID which detects the neutrons, the pixel position on the detector, and so on. Information on the sample environment such as sample temperature and external magnetic
field is also contained in the event-recording format. In the DAQ system, there are data acquisition servers, data storage servers, and data analysis servers.

We have developed a software group which controls the hardware, manages a variety of measurements, monitors the real-time data during measurements, visualizes the reflectivity data in reciprocal space, and analyzes the data. Most of the programs run on Linux (Scientific Linux), and a few run on Windows XP because several of the hardware drivers only work on the Windows platform.

VII. SAMPLE ENVIRONMENT EQUIPMENT

An external magnetic field is indispensable to reflectivity measurements of magnetic multilayers. A conventional electromagnet is used for the experiments. This magnet is water cooled and produces magnetic fields of up to 10 kOe. The direction of the field, horizontal or vertical, can be changed by rotating the magnet through 90°. The polarity of the magnetic field can be continuously switched using a bipolar power supply. We have a cryocooler which can control the sample temperature between 4 K and the ambient temperature. An additional superconducting magnet is also available. Reflectivity measurements are performed down to 50 mK under magnetic fields up to 70 kOe in combination with a $^3$He variable temperature insert (VTI). The superconducting magnet was specially designed to minimize leakage flux and to avoid depolarization of the polarized neutrons along the beam path inside the magnet (an asymmetric mode).
VIII. EXPECTED THROUGHPUT

Fig. 5 shows the expected reflectivity curves, with error bars, and measuring times for unpolarized neutron reflectivity measurements of Ni thin films of thickness 300 nm on Si substrates, estimated using a neutron-ray-trace simulation package McStas [6, 7], when the proton beam power is assumed to be 1 MW. The reflecting areas of the samples are (a) $30 \times 30 \text{ mm}^2$, and (b) $10 \times 10 \text{ mm}^2$, respectively. These graphs suggest that a typical unpolarized neutron reflectivity measurement for a sample will be completed within a few minutes, assuming typical sample size at BL17 (Fig. 5(a)), or several hours for a tiny sample (Fig. 5(b)). In the case of polarized neutron reflectivity measurements without polarization analysis, the duration time is four times longer than those indicated in Fig. 5 because more than half of the incident neutrons are filtered out by the supermirror polarizer, and two kinds of reflectivity, according to the spin-flipper states (active and inactive), are measured. Furthermore, the time is doubled (a total of more than eight times longer than the times indicated in Fig. 5) in the case of polarized neutron reflectivity measurements with analysis of the spins of the reflected neutrons using the additional spin-flipper and the analyzer. Four kinds of reflectivity ($R_{++}$, $R_{+-}$, $R_{-+}$, and $R_{--}$) are measured using polarized neutrons.

IX. CONCLUSION

We have contributed extensively to the design and construction of the new polarized neutron reflectometer at the MLF at J-PARC. At the beginning of March 2011, the installation was expected to be completed at the end of March as scheduled. However, construction was interrupted by the massive earthquake on March 11, 2011. None of the devices and beam line components was seriously damaged. However, most of biological shielding blocks around the neutron target vessel were unexpectedly moved. We had to remove all the
components which had already been installed along the beam line, and reinstall them after fixing and repairing the bulk shielding blocks. We expect to recover the neutron beams with proton powers of 200 kW and to start the user program on this polarized neutron reflectometer at the beginning of next year (2012). J-PARC, including MLF, is the international experimental facility which is open to both domestic and international researchers. We are very much looking forward to performing neutron reflectivity measurements with both domestic and international users at BL17.

Acknowledgements

We would like to express our heartfelt thanks to all the people who gave us kind messages and supported Japan in our difficult days after the earthquake.

References