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Design Concepts and Advanced Research Application of the J-PARC Neutron Reflectometer with Horizontal Sample Geometry

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A high-performance and unique neutron reflectometer with horizontal sample geometry was proposed for a high-intensity pulsed neutron source of the Japan Proton Accelerator Research Complex (J-PARC). The design of downward beam lines is optimized quantitatively by using the Monte Carlo ray-tracing simulation code, McStas, so as to measure specular reflectivity easily and effectively up to 0.5 $Å^{-1}$ in the magnitude of neutron momentum transfer along the depth direction for the samples with free interfaces. The high-intensity pulsed neutron source makes it possible to detect weak signals from low-contrast sample and of off-specular scattering with high precision during a short period of time in minutes. The reflectometer is equipped with a few unique options: polarized neutron option, grazing incidence diffraction (GID) and neutron spin-echo options. The polarized neutron option is used to detect the spatial distribution of weak magnetic moment, and also distinguish incoherent scattering originating from the samples. The GID option utilizes evanescent wave propagating near surface, and is favorable to observe the detailed surface structure with the scale of nm. The spin-echo option is effective for the studies on dynamics of interface and surface in neV order, and is expected to develop new science using reflectivity measurement.

Key words: a neutron reflectometer, horizontal sample geometry, J-PARC, a high-intensity pulsed neutron source, unique optional functions

1. INTRODUCTION

It has been widely recognized that reflectivity measurement using x-ray and neutron is powerful and essential technique for the studies on surface and interface of materials due to its high spatial resolution, especially along the sample depth direction. [1,2]

The neutron is a complementary probe to x-ray for studying material structures, since the neutron interacts with nucleus (nuclear scattering) while x-ray does with electron. The degree of interaction of neutron with nucleus, which is expressed by the sign and magnitude of scattering length, *b*, changes irregularly with atomic number, and is also different between isotopes. There is a relatively large difference in *b* between light hydrogen (H, -0.374×10^{-12} cm) and deuterium (D, 0.667×10^{-12} cm) atoms for neutron, although they are not distinguishable by x-ray. Thus, the deuterium-labeling method is utilized for the researches on the materials containing many hydrogen atoms, such as polymers, surfactants, biomaterials, etc. Also, the neutron has spin and interacts with unpaired electrons in magnetic atoms (magnetic scattering), so that the neutron can detect the spatial distribution of magnetic moment for magnetic thin film or artificial lattice. Another feature is that the neutron exhibits high permeability to materials. The interfaces buried inside materials, e.g., the solid/liquid or liquid/liquid interfaces, can be investigated by using the neutron. Moreover, the neutron is non-destructive probe because of carrying no charge.

There exist a few neutron reflectometers in Japan: ARISA [3] and PORE [4] at the KENS pulsed neutron source, and MINE [5] and LTAS [6] at the JRR-3M research reactor of Japan Atomic Energy Research Institute (JAERI). Among them, only the ARISA reflectometer adopts horizontal sample geometry, so that it is applicable for the samples with free interface. Though various material interfaces have been studied on these domestic neutron reflectometers so far, their application is limited to the measurement of specular reflectivity of 10^{-6} at most by the flux of the existing neutron sources.

The joint project of High Energy Accelerator Research Organization (KEK) and JAERI, named Japan Proton Accelerator Research Complex (J-PARC), has been started to construct the experimental facilities based on a series of high-intensity proton accelerators at the Tokai campus of JAERI, Ibaraki. [7] One of the major facilities, the Material and Life Science Experimental

Facility, has a 1MW pulsed neutron source, which exceeds the present world's brightest pulsed neutron source of the ISIS Facility, Rutherford Appleton Laboratory, UK with the accelerator power of 800 MeV × 200 μA. [8] A neutron reflectometer with horizontal sample geometry was proposed as one of indispensable instruments for the J-PARC high-intensity pulsed neutron source. Here, the conceptual design and current status of beam-line design are reported on the proposed J-PARC neutron reflectometer.

2. THE MERIT OF A PULSED-NEUTRON SOURCE

At the pulsed neutron source, white neutrons with a wide wavelength band are used for measurement, and the Time-of-Flight (TOF) method, in which the neutron wavelength is determined by measuring the time when it takes that the neutrons emerging from the moderator reach a detector, is applied to the data analysis. The merit of the reflectometer at the pulsed-neutron source is that it can cover the wide space in neutron momentum transfer, Q, at one time, without the scanning measurement, such as the so-called θ-2θ scan for specular reflectivity, performed at the reactor source using the monochromatic neutron. This implies that the pulsed-neutron reflectometer adopting horizontal sample geometry is favorable for the studies on free interfaces such as liquid surface and liquid/liquid interface, since the frequency to move the sample itself or the relevant goniometers, of which movement could induce vibration on the sample, is much less compared with the reflectometer using the monochromatic beam. The above merit of pulsed-neutron reflectometer, also, leads to the great advantage to make the time-dependent measurement with a short period of time in minutes or less, though it requires extremely high neutron flux. Further, the use of a one-dimensional position-sensitive detector on the pulsed-neutron reflectometer makes it possible to observe specular and off-specular reflectivity simultaneously in wide Q-space, though the off-specular reflection is measured by combining different types of scans such as the rocking scan, the detector scan and the off-set scan, or choosing the suitable scan from them according to the purpose on the monochromatic-neutron reflectometer.

3. THE CURRENT DESIGN OF THE J-PARC REFLECTOMETER

The proposed reflectometer is installed at one of the cold neutron beam lines viewing the J-PARC coupled liquid hydrogen moderator. The neutron flux, predicted by the Monte Carlo calculation, at the moderator surface is shown as a function of wave number, k ($=2\pi/\lambda$) where λ is the neutron wavelength, in Fig.1. It is found that the coupled liquid-H_2 moderator exhibits peak flux at k around 2 $Å^{-1}$, which is corresponding to about 3 Å in λ.

The use of neutrons with a wider λ band is advantageous for reflectivity measurement, and the neutron flux at the lower limit of the λ band is required to be as high as possible for measuring low reflectivity at higher neutron momentum transfer, Q_z ($=4\pi\sin\theta/\lambda$), along the direction normal to the sample surface. Eventually, the lower λ limit was chosen to be 2.5 Å, which gives a neutron flux slightly lower than the peak at λ of 3 Å, but satisfies well the requirements mentioned above. On the other hand, the upper limit of the λ band was determined by considering the frame-overlapping, in which the slow neutrons are passed by the prompt ones emitted in the next pulse. The critical λ value for the frame-overlapping was evaluated to be 9.0 Å from the operational frequency, 25 Hz, of the J-PARC accelerator and the total length, 17.5 m, from the moderator to a detector, for this reflectometer. Hereafter, the neutron λ band was supposed to be ranging 2.5 - 9.0 Å for the beam-line design.

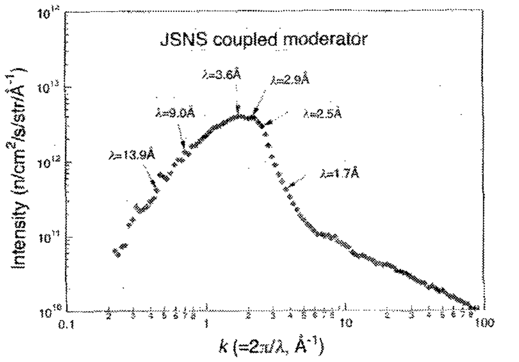

Fig.1 The neutron flux at moderator surface for the J-PARC coupled liquid H_2 moderator as a function of k.

The J-PARC reflectometer adopts horizontal sample geometry for free interfaces, so that special downward neutron beams are required to make the grazing-incident beam onto the sample surface keeping the sample horizontal, since the samples with free interfaces can't be tilted at all. The beam-line design is optimized for satisfying the requirement that specular reflection can be observed easily and effectively for free interfaces at least up to 0.5 $Å^{-1}$ in Q_z. It was found that the four angles, i.e., 5.71, 2.22, 0.862 and 0.335 deg., are needed for the downward beams by considering the overlap in Q_z coverage for each angle of the neutrons with λ ranging from 2.5 to 9.0 Å. Fig.2 demonstrates the Q_z-coverage for each angle of the neutrons. The beam lines with the two higher angles, i.e., 5.71 and 2.22 deg., are guided viewing the moderator surface directly without bending by optical devices such as a supermirror bender, since the use of optical devices leads to the loss of neutron intensity to no small extent. The other two with the lower angles, i.e., 0.862 and 0.335 deg., are produced from the 2.22deg beam by using the supermirror bender.

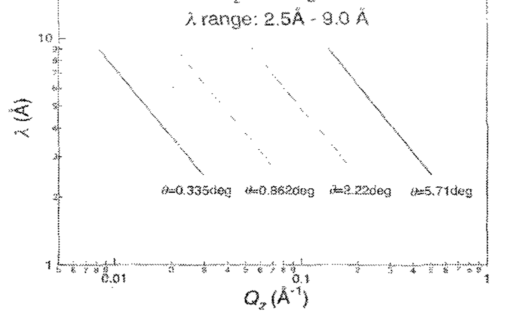

Fig.2 The Q_z coverage for each angle of the neutrons with λ ranging from 2.5 to 9.0 Å.

Fig.3 shows a schematic drawing of the two downward beam lines viewing the moderator surface directly, and the arrangement of basic instrumental components. The two beam-holes for the 5.71- and 2.22-deg. downward beams are prepared inside a biological shield extending about 7 m from the center, and either of them is selected by adjusting the vertical position of a beam shutter. A T0-chopper is installed just outside of the biological shield for reducing the background attributed to the high-energy neutrons coming along the beam line. The final selection of the beam angle from 5.71, 2.22, 0.862 and 0.335 deg. is made by adjusting the vertical position of or rotating the second shutter, located between the T0-chopper and the first slit, with four different optical devices corresponding to each beam, i.e., two supermirror guide sections for the higher-angle beams and two bender ones for the lower angles. At the sample position, the two downward beams with higher angles are vertically separated by about 90cm, and the 5.71deg beam runs below the floor level. The pit with 1.5m depth is prepared along the beam line, and all the instrumental components, i.e., the first slit, a sample stage, a detector tower, etc. on the same optical bench are softly and stably moved up and down according to the selected beam angle.

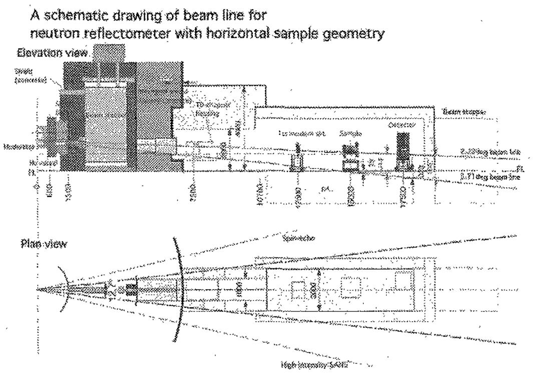

Fig.3 A schematic drawing of the two downward beam lines viewing moderator surface directly, and the arrangement of basic instrumental components.

Table I. The specifications of the J-PARC reflectometer

Moderator type	Coupled type, liquid H_2
Operational frequency	25 Hz
Moderator-sample	15 m
Sample-detector	2.5 m
Optical device	Supermirror bender
Occupied area	-6.35 deg < θ < 6.35 deg
Wavelength range	2.5 Å < λ < 9.0 Å
Downward beam angle	5.71, 2.22, 0.862, 0.335 deg
Q_z range	$Q_z < 0.5$ Å^{-1}
Q_z resolution	$\Delta Q_z/Q_z < 10$ %
Moderator viewed area	5 cm (V) × 10 cm (H)
Sample size	Max. 5 cm × 5 cm
Detector	2D-PSD (1mm×1mm pixel)
Chopper	T0-chopper
Optional measurement	Off-specular reflection Grazing incident diffraction Polarized neutron option Neutron spin-echo option

The beam-line design is optimized quantitatively by using the Monte Carlo ray-tracing simulation code, McStas (version 1.5) [9], to obtain the higher neutron flux at the sample position. Fig.4 shows one example of the simulation results. The figure exhibits neutron flux at the sample position for the downward beam line with 5.71 degrees as a function of λ. In the calculation, the long $3Q_c$-supermirror guide was installed downstream from the beam shutter, just before the first incident slit. The angular resolution, $\Delta\theta/\theta$, determined by a pair of the incident slits was supposed to be 5 %. It is found from this figure that the peak flux of 2.3×10^8 n/sec/Å is obtained at λ of 2.8 Å by the supposed beam extraction.

The present instrumental parameters of the proposed reflectometer are summarized in Table I.

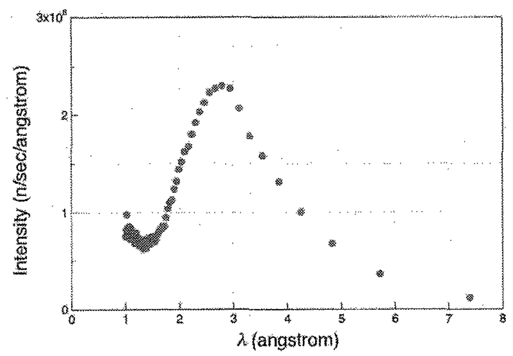

Fig.4 The expected neutron flux at the sample position for the 5.71deg downward beam line with $\Delta\theta/\theta$ of 5 % as a function of λ.

4. THE SCIENTIFIC FEASIBILITY OF THE J-PARC REFLECTOMETER

The reflectometer proposed here adopts horizontal sample geometry, so that it can be applied for any kinds of material interface including free interfaces, such as liquid surface or liquid/liquid interface, which are not measured by using the reflectometer with vertical sample geometry. By installing the reflectometer at the high-intensity pulsed-neutron source, the weak intensity from low-contrast sample or off-specular scattering can be precisely measured with high resolution during a short period of time in minutes or less. This makes it possible to observe the structural response of material interfaces to the change of the various outer fields such as temperature, surface pressure, magnetic field, light, etc. using neutron reflectometer. The reflectometer is equipped with a polarized neutron option, i.e., a polarizer, an analyzer and a spin-flipper, so that the spatial distribution of weak magnetic moment can be detected for magnetic thin film or artificial lattice. Also, the polarized-neutron is used to distinguish incoherent scattering. This is very effective especially for the samples containing many hydrogen atoms, such as polymer or bio-membrane, because it reduces the background intensity from incoherent scattering so that the structural information is obtained more precisely and reliably. Further, another two unique and challenging options, i.e., grazing-incidence diffraction (GID) and spin-echo options, are introduced. So far, there has been no precedent for installing these techniques on the pulsed-neutron reflectometer. The GID option utilizes evanescent wave propagating near surface, and is favorable to observe the detailed surface structure with the scale of nm. A two-dimensional position-sensitive detector with high spatial resolution needs to be scanned in the horizontal plane for the GID measurement. The spin-echo option is effective for the studies on the dynamics of surface and interface in neV order, and is expected to develop new science using reflectivity measurement. Here, a neutron resonance spin-echo method using radio-frequency flippers is adopted, since large precession coils are unnecessary and the respective components can be made compact. The R&D work for realizing the spin-echo option on the reflectometer is now in progress.

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