CO1-1 Current Status of Versatile Compact Neutron Diffractometer (VCND) on the B–3 Beam Port of KUR, 2021

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INTRODUCTION: Neutron diffraction is a powerful tool to precisely determine the positions of light elements (H, Li, etc.) in solids. This is the main reason why neutron powder diffractometers are critical for structural investigations of energy storage materials such as rechargeable lithium-ion batteries and hydrogen-absorbing materials. The B–3 beam port of Kyoto University Research Reactor (KUR) had long been used as a four-circle single-crystal neutron diffractometer (4CND). For the last decade, however, the 4CND was so old that its research activity on neutron science was quite low. Nowadays, the versatile compact neutron diffractometer (VCND) has been installed instead of the 4CND, as shown in Fig. 1 [1]. The neutron wavelength, λ, which is monochromatized by the (220) plane of a Cu single crystal (i.e., Cu monochromator), is 1.0 Å. To cover the detector area of 6 ° ≤ 2θ ≤ 130 °, twenty-five ^{3}He tube detectors (1/2 inch in diameter) are used, where 2θ is the scattering angle. A detector bank including twenty-five ^{3}He tube detectors is placed on the arm of the HUBER-440 goniometer. The distance from the Cu monochromator to the sample is approximately 2 m, and the distance from the sample to the detector is 1.2 m.

CRYSTAL STRUCTURE ANALYSIS: Aragonite is one of the crystal forms of calcium carbonate ($CaCO_3$). For biominerals such as shells, eggshells, pearls, and corals, aragonite plays an important role in their mechanical properties [2]. The crystal structure of the synthetic aragonite can be described by an orthorhombic system (space group *Pnma*). In Fig. 2, an excellent fit was obtained between the observed and calculated intensities (where R_{wp} = 14.992% and S = 3.4488). It is worth noting that the background intensities were omitted from the ND data. The values of a, b, and c were estimated to be 5.739(3), 4.962(2), and 7.968(4) Å, respectively. The atomic positions (x, y, z) and isotropic atomic displacements (B_{iso}) for each element were precisely determined as follows: Ca (0.236(3), 1/4, 0.417(2)) and B_{iso}(Ca) = 0.67(38) Å^2 in the 4c site; C (0.086(3), 1/4, 0.7619(14)) and B_{iso}(C) = 0.21(29) Å^2 in the 4c site; O1 (0.096(3), 1/4, 0.9247(18)) and B_{iso}(O1) = 0.48(27) Å^2 in the 4c site; and O2 (0.087(3), 0.477(2), 0.6821(12)) and B_{iso}(O2) = 0.77(22) Å^2 in the 8d site.

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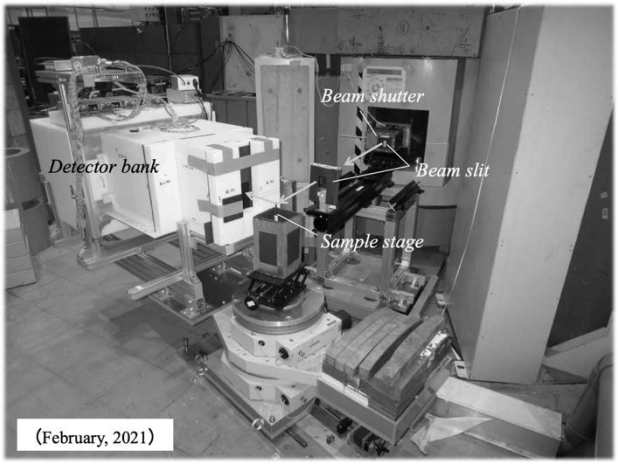

Fig. 1. Versatile compact neutron diffractometer (VCND) installed at the B-3 beam port of Kyoto University Research Reactor (KUR) [1].

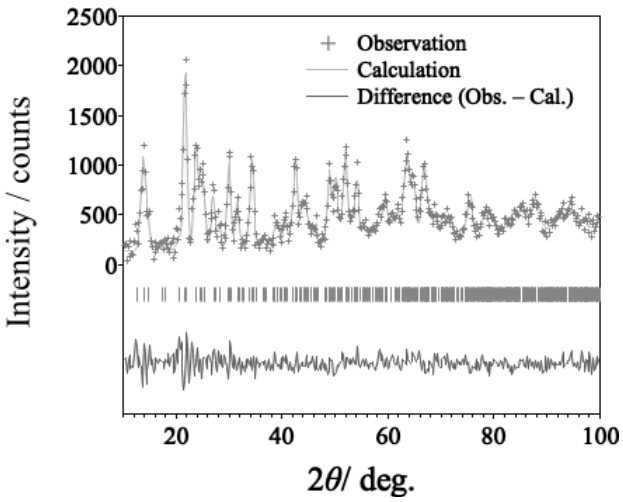

Fig. 2. Rietveld refinement using ND data for synthetic aragonite ($CaCO_3$) at room temperature.

CO1-2 Towards to mass production of high-*m* neutron focusing supermirrors

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INTRODUCTION: Progress of neutron optical devices is very significant. We have established fabrication method for aspherical focusing supermirror with metal substrate [1-3]. The metallic substrate is robust and ductile, to which able to fabricate steeply curved surface with high form accuracy. It is also applicable to use under high radiation irradiation and high-temperature filed, even at a place close to the neutron target and moderator. Furthermore, it is possible to fabricate a large focusing mirror by combining multiple segmented mirrors with mechanical fastening entailing the usage of screw holes and fixture tabs. We have solved the problem of required surface roughness for neutron mirror. The roughness should be smaller than 0.3 nm for high-*m* supermirror coating. Here *m* is the maximum critical angle of the mirror in units of critical angle of natural nickel. By using electroless nickel-phosphorus (Ni-P) plating, we overcame the problem and are establishing fabrication process for aspherical focusing supermirror. There is still a problem of peeling off for high-*m* supermirror coating on metal substrate with steep curvature. It is also important to improve reflectivity of the supermirror. In this study, we report a status of mass production for high-*m* neutron focusing supermirrors.

EXPERIMENTS: We fabricated ellipsoidal metallic substrates with the Ni-P plating, based on the technology using ultrahigh precision cutting with correction processing, followed by mechanical precision polishing. The first precise manufacturing was conducted at a CNC machine for development of neutron optical devices at workshop of the KURNS. The ultra-precise manufacturing, polishing and cleaning of the metallic substrate were conducted at RIKEN. The supermirror coating was conducted with ion beam sputtering machine at the KURNS (KUR-IBS) [4]. Figure 1 shows the photograph of ellipsoidal supermirror deposited on the latest lot (LOT-23) and silicon substrates. The semi-major and semi-minor axes of the ellipsoidal supermirror were 1250 mm and 65.4 mm, respectively. The acceptable angle of the minor axis arc of the ellipsoidal supermirror is 20 degree. In this study, we had to reconfirm the importance of the cleaning process and storage period because many failures were made in depositing on a substrate with a long storage period. Eventually, we have fabricated *m*=6 NiC/Ti(C) supermirrors in which effective number of layers was 9750, where the half of the layers were very thin carbon interlayers. The thickness of interlayer is constant and approximately a few sub-nanometers. In the point of view of the reflectivity performance, the effective number of periodic layers is 4875. The neutron experiments were conducted at the BL06 (VIN ROSE) beam port at J-PARC MLF.

RESULTS: Figure 2 shows reflectivity profiles of neutrons by the *m*=6 NiC/Ti (C) supermirror deposited on the monitor mirror (silicon substrate). The measurement was conducted by four incident angles, 0.8, 1.6, 3.2 and 4.8 degrees. By adding very thin carbon interlayer between NiC and Ti layers, we have realized *m*=6 supermirror with high reflectivity. Here the number of effective layers, 4875, is not so many enough for *m*=6 supermirror, and there is still room for improvement of reflectivity of the supermirror. Noting that it is not reflectivity of ellipsoidal supermirror. The evaluation of ellipsoidal supermirror is now analyzing.

Fig. 1. The photograph of LOT-23 ellipsoidal supermirrors with two monitor mirrors.

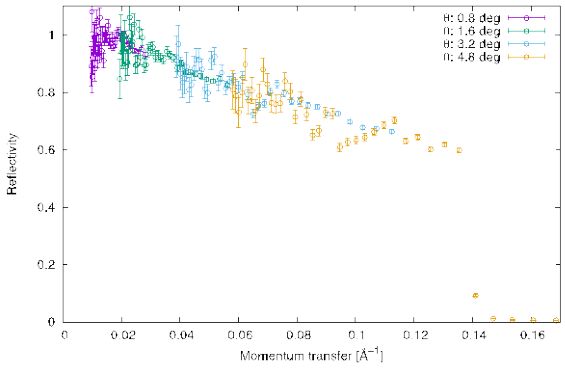

Fig. 2. The measured reflectivity of *m*=6 NiC/Ti (C) supermirror deposited on a monitor mirror (silicon substrate).

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CO1-3 Development of multilayer mirrors for neutron interferometer

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INTRODUCTION: Neutron interferometry is a powerful technique for studying fundamental physics. Numerous interesting experiments [1] have been performed since the first successful test of a single-crystal neutron interferometer [2]. However, the single-crystal interferometer is inherently not able to deal with a neutron that has a wavelength longer than twice its lattice constant. In order to investigate problems of fundamental physics, including tests of quantum measurement theories and searches for non-Newtonian effects of gravitation, the interferometry of cold neutrons is extremely important, since the sensitivity of interferometer for small interaction increases with the neutron wavelength. A large scale of interferometer also has the advantage to increase the sensitivity to small interactions.
One of the solutions is an interferometer using neutron multilayer mirrors [3]. We succeeded in developing a multilayer interferometer for cold neutrons in which two paths are completely separated for the first time using wide-gap etalons [4]. We can easily control parameters such as Bragg angle, reflectivity, and Bragg peak width by selecting the deposited material and tuning the bilayer thickness and the number of layers.
We have started the development of multilayer interferometer at the beamline 05 NOP in MLF. From 2019, we are continuing the experiments with etalons with monochromatic mirrors in order to demonstrate the performance of the interferometer. Figure 1 shows the interference fringes with etalons according to time-of-flight. The phase of interferogram depends on the wavelength of neutrons. We are testing the practical application of the interferometer. Neutron coherence scattering length of the material can be measured by inserting the sample into a path of the interferometer. The results of the trial measurements were consistent with the literature values.
Because the mirrors have narrow bandwidth of the neutron reflectivity, the number of neutrons contributing to the interference is limited. When the neutron supermirrors whose lattice constants vary gradually are utilized in the interferometer, the effective range of neutron wavelength can be broadened to be applicable to a pulsed source. In addition, the wavelength dependence of the interactions can be measured simultaneously by using pulsed neutrons.

EXPERIMENTS AND RESULTS: We are trying to fabricate the neutron mirrors with wide band for the interferometer by using Ion Beam Sputtering facility in KURNS. The mirrors should have the wide and smooth top of the reflectivity with the range of momentum transfer from 0.4 nm^{-1} to 1.0 nm^{-1}. Especially, half mirrors with the wide range of neutron wavelength are needed for the interferometer. We tried to make the half mirrors and to measure the reflectivity at MINE2 in JRR3. Figure 2 shows the reflectivity of the half mirrors on the fused silica substrates. Neutron wavelength was 0.88 nm and the bandwidth of the beam was 2.7% of the wavelength. Mirrors with more uniform reflectivity were created. We are continuing the development of the mirrors for the neutron interferometer.

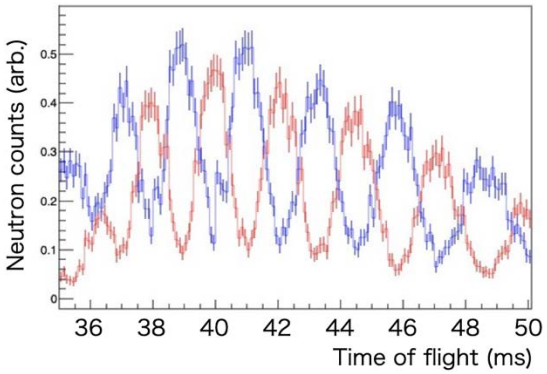

Fig. 1. Interference fringes with multiplayer mirrors for pulsed neutrons. The contrast was improved.

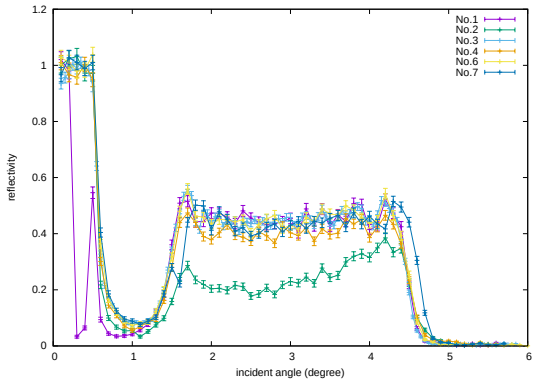

Fig. 2. Reflectivity of the half mirror with wide band of neutron wavelength. Colors represent the sample ID.

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CO1-4 Development of high-resolution cold/ultracold neutron detectors using nuclear emulsion

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INTRODUCTION: Nuclear emulsion is a 3D-tracking detector for charged particles with a submicron resolution. It consists of silver halide crystals with diameter of several ten – several hundred nm. It works as a slow neutron detector by combining it with nuclides which absorb neutrons and emit charged particles. A cold/ultracold neutron detector with spatial resolution less than 100 nm has been developed by combining nuclear emulsion with a thin converter layer including ^{10}B[1]. An experiment (Experiment 1) was conducted at PF2, Institut Laue-Langevin (ILL) in order to obtain a spatial distribution of UCNs in the Earth's gravitational field using a detector fabricated by coating a nuclear emulsion on a converter layer sputtered by an ion beam sputtering system (KUR-IBS) in KURRI[2]. Nuclear emulsion can also be used for fundamental studies of radiology such as that of proton boron capture therapy (PBCT)[3]. A related experiment (Experiment 2) was conducted at CN-3 beam line. Studies for applications of emulsion detectors to neutron imaging are going on[4,5]. Also applications of fluorescent nuclear track detectors (FNTD) [9] to neutron imaging has been studied in parallel. An experiment with the latter detector (Experiment 3) was conducted at CN-3.

Experiment 1: Layers of $^{10}B_4C$(200 nm)-NiC-C were sputtered on double-side-polished Si plates with thickness of 500 μm with KUR-IBS and were coated with 10 μm-thick fine-grained nuclear emulsion[6] at Nagoya University. They were exposed to UCNs of 9.5 m/s at the downstream of a mirror-absorber system of the qBOUNCE experiment[7] which is in a similar configuration as described in the Ref. [8] with keeping the gap between the mirror and the absorber 24 μm. The number of arrived UCNs at the fiducial area (6.5 cm × 24 μm) of one of the detector was 1.1×10^4. The detector was developed and tracks of α-particles and ^{7}Li from neutron absorptions that are in the angle acceptance of $\theta < 0.9$ rad were read out by an automatic track readout system at Nagoya University. The acceptance guarantees the spatial resolution of the absorption points less than 100 nm.
Results: The number of the tracks read out from the fiducial area was 1674, which was reasonable considering the detection efficiency[1] and the fraction of tracks to be found in the angle acceptance. Analyses will be continued and a final spatial distribution will be obtained.

Experiment 2: The sample consists of fine-grained emulsion and a 16 μm thick polyvinyl alcohol sheet immersed in sodium tetrachloride solution. The sheet contains natural boron of 31 μg/cm^2. The sample was exposed to 20 meV neutrons at the CN3 beamline. The beam density was 5×10^8 cm^{-2}. The exposed film was developed and observed under an epi-illumination optical microscope.
Results: The track density of α and ^{7}Li produced from ^{10}B(n, α)^{7}Li reaction was expected to be 1×10^6 cm^{-2} from the amount of sodium tetrachloride and the reaction cross section. The number of observed tracks was $(1.3 \pm 0.1) \times 10^6$ cm^{-2}, which is approximately consistent with the expectation. Furthermore, the number of silver grains along the observed tracks was counted manually. It showed a bimodal distribution reflecting the difference in energy deposition. We can extract α tracks produced in the proton boron capture reactions by counting the silver grains in a track.

Experiment 3: A sample of a neutron imaging device that combines an FNTD and a $^{10}B_4C$ (200 nm)-NiC-C film was created to study the fundamental characteristics of the detector. The FNTD is a single crystal of aluminum oxide doped with carbon and magnesium. It records tracks of ions, and the tracks are observed as fluorescence features against the background under a confocal laser microscope with a resolution of sub-micrometers. This detector can be reused repeatedly because UV laser irradiation can erase the recorded tracks. The sample was irradiated with the neutron beam for 10^4 sec under one megawatt thermal power of the reactor and recorded the tracks of neutron capture events from the $^{10}B_4C$-converter and background γ-ray. The authors are tuning the UV laser conditions for the track erasure.

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CO1-5 Development of a Spin Analyzer for Ultra-Cold Neutron

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INTRODUCTION: Existence of non-zero permanent electric dipole moments (EDM) of the fundamental particles violates time reversal symmetry. Under CPT conservation, T violation implies CP violation. Thus, a precise measurement of an EDM may reveal the origin of matter dominant universe. The TUCAN (TRIUMF Ultra-Cold Advanced Neutron source) collaboration aims to measure a neutron EDM with a sensitivity of 10^{-27} ecm, which is more than one order better sensitivity than the current best measurement.

The neutron EDM is measured by the precise measurement of spin precession frequency of neutrons. Ultra-Cold Neutrons, whose kinetic energy is less than a few 100 neV, is used for the measurement. One of the key components of the measurement is a spin analyzer of the UCN. Since the kinetic energy of an UCN is so low that magnetic potential can is used as a spin filter. When iron, which has a large saturation magnetization of 2.2 T, is used for the spin filter, the effective potential V_{eff} is

$$V_{eff} = V_{Fe} \mp |\mu| \cdot |B| = 90 \text{ neV, or } 330 \text{ neV}$$

Where $V_{Fe} = 210$ neV is the Ferimi potential of the iron, $\mu = 60$ neV/T is the magnetic moment of the neutron, and $B = 2.2$ T. Only one spin state of UCNs with kinetic energies between 90 neV to 330 neV can transmit the iron magnetic potential. Therefore, magnetized iron functions as an UCN spin filter. In order to reduce UCN absorption, the iron should be as thin as an order of 100 nm.

EXPERIMENTS: The thin iron films are prepared by ion beam sputtering facility at the Institute for Integrated Radiation and Nuclear Science, Kyoto University (KURNS). We produced thin iron films with thicknesses of 30, 50, and 90 nm on Al foils and Si substrates.

Magnetic properties of the samples were measured by a vibrating sample magnetometer at KURNS. Samples on the Si substrates were also characterized by the cold neutron reflectometry. The measurements were carried out at the J-PARC/BL05. Polarized cold neutron beam was incident on the sample and the reflectivity was measured. A spin-flipper was used to change the spin state of the incident neutrons.

RESULTS: Figure 1 shows the magnetization curves of the samples. The iron films are fully saturated by the magnetic field of 12 kA/m for the samples on the Al foils and 4 kA/m for those on the Si substrates.

Figure 2 shows the reflectivity of the iron film with 90 nm thickness on the Si substrate. The reflectivity depends on the momentum transfer of the reflected neutrons. The polarization of the incident neutron beam was evaluated by another experiment. From the fitting, the critical momentum transfer was extracted. This is translated to the effective potential of the samples 328 neV for the one spin state and 65 neV for the other spin state. This means the iron film has ability to select spin state for the UCNs in an energy range of 65 – 328 neV. This result is very promising for the development of the spin analyzer for UCN. We plan to evaluate the samples using UCNs in the spring 2022.

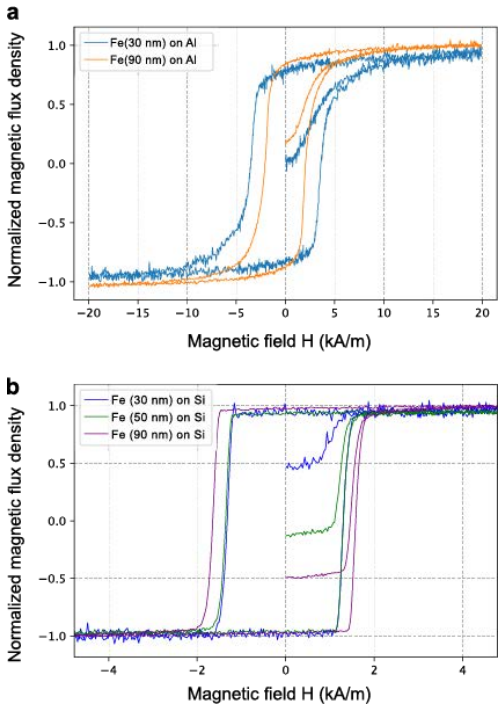

Fig. 1. Magnetization curves of iron films on Al foils (a) and on Si substrates (b). Figure from [1]

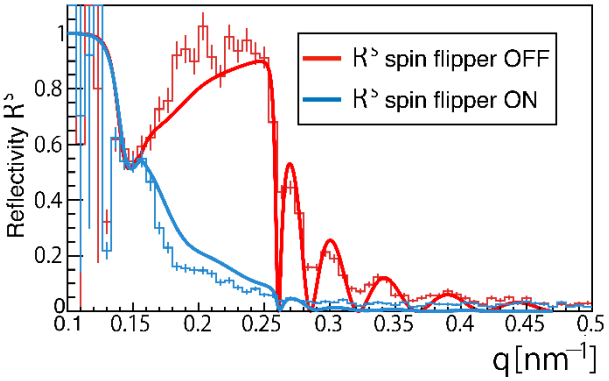

Fig. 2. Neutron reflectivity Red shows the data with spin flipper OFF and Blue shows the data with spin flipper OFF. Solid lines are indicates fit result. Figure from [1]

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