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Aspects of the Neutron 4-circle Diffractometer FONDER and the Bent Si Monochromator

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A neutron 4-circle diffractometer (FONDER) was constructed at the beam-port T22 of JRR-3M at JAERI. With a cryostat, we will be able to take diffraction intensities for structure analysis down to 8 K. We have also constructed and tested Si-553 and Si-422 double-bent monochromators. Sufficient beam-focus was achieved. For an oscillation photograph, neutron Image-plate is attachable in front of the detector. Several standard materials were used to test the performance of the system. Aspects of the diffractometer, the double-bent monochromator and the IP-camera are described.

KEYWORDS: neutron 4-circle diffractometer, instrumentation, bent Si monochromator

§1. Introduction

A four-cycle diffractometer is commonly used for structural studies such as structure analysis, measurements of intensity distribution in a reciprocal space etc., especially in the X-ray field. Recently, we constructed a neutron four-circle diffractometer at the T22 beam port of the guide hall of JRR-3M in Japan Atomic Energy Research Institute. In this paper, we will describe the current status, aspects and prospects of this diffractometer as well as a bent Si monochromator. The instrumentation was designed by looking the plan of replacing the guide-mirror to super-mirror. Then, the current stage is still underdevelopment. Very recently, the part of the guide mirror was replaced to a super-mirror. We tested the beam character for few days, and in this paper the preliminary result is given.

§2. Instrumentation

2.1 4-circle diffractometer

A four-circle diffractometer is composed of two Huber-440 for ω- and 2θ- axes, Huber-480 for χ-axis and Huber-420 for ϕ-axis. Essentially the system is the ordinary Four-circle Off-center-type Neutron Diffractometer(FONDER). The load limit of the diffractometer on the ϕ axis is about 360 kg, but the actual load limit is constrained by that of the goniometer-head(about 10kg) to align the x-, y- and z- position of a sample. Program *mxc* of MAC Science Co. is installed for data accumulation. Scans in a reciprocal space is performed by a home made program coded by Dr. Nakao of KEK. The main detector is ^{3}He counter horizontally aligned, and also a ^{3}He monitor counter for incident neutron is settled in front of shutter and absorber devices. Shutter is made of BN plate and absorber is consisted with two different thickness plastic plates. Between sample and counter, left- and top- side half slits are there for using of the peak centering procedures. Double narrower-slits made of BN plates are installed in front of the main detector and the acceptance angle is controlled by a computer. The large χ cradle blocks the incident beam above ω=30°. Above this region, we use the so-called ω-fix mode. The angle ω is fixed at 28°, and χ and ϕ angles are adjusted to get a target reciprocal position. We have three beam-holes from the monochromator position; $2\theta_M$=42.6°, 90° and 113.2°. Mechanically maximum angle of 2θ of detector is 90° for $2\theta_M$=42.6°, and 155° for $2\theta_M$=90°. Upon this angle, the detector arm hits the neutron guide tube. The 42.6° beam-hole is used for E_i=13.7meV(2.435 Å) neutrons with PG002 monochromator.

We tested the all system by using PG002 monochromator at the beginning when the guide-mirror is normal one. We prepared standard samples such as $SrTiO_3$, d-KDP, YAG, NaCl etc. The intensity is reasonably strong, but the wavelength (2.435 Å) is considerably long for the usual structure study. We introduced PG-filter to reduce the higher energy contamination. Without the filter, the ratio of $\lambda/2$ to λ is 5.2 %, while it turns out to 0.01 % with a filter. A cryostat was settled on the four-circle diffractometer, and the lowest temperature we obtained was 7 K. The feature of a cryostat on the goniometer is shown in Fig.1.

2.2 Performance of the diffractometer

As the test sample, we performed the experiments of structural phase transition of MeHPLN ($C_{14}H_{10}O_2$), which has many hydrogen atoms in a molecule. The crystal size is 0.98x1.21x5.15 mm^3. The space group is

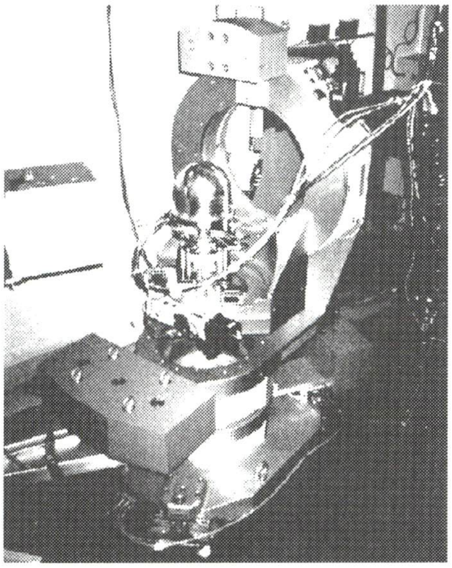

Fig. 1. The overview of the 4-circle diffractometer with a cryostat.

C2/c, and the obtained lattice parameters at room temperature are a=12.189(2), b=11.149(2), c=7.303(1) Å, α=89.99(2), β=95.11(1),γ=90.00(1)°. Peak search, peak refinement and determination of the UB-matrix are automatically performed as usually performed in a laboratory X-ray system. The accuracy of the lattice parameters obtained seems to be satisfactory. At room temperature, we measured Bragg intensities up to $2\theta_{max}$=89°, and 216-points were observed for 36 hours. The data F>2σ(F), 163-points, are used for structure analysis. Among them, the independent reflections are 73. We performed ψ-scan of 002 reflection, and we found that the intensity changes 50 % between maximum and minimum ones. This effect is coming from absorption and extinction effects. After extinction correction, absorption correction etc., the structure analysis was successfully performed. Since the number of the fitting parameters is close to the obtained data point, we combined the structure data taken by the X-ray diffraction. As the result, we could obtain R(F)=2.7 %. At 7 K, the low temperature phase was examined, in which 4-times modulated phase along the b-axis appeared. The intensity of the superlattice reflection $2\frac{1}{2}2$ is 10cps while the strongest Bragg intensity is 400cps. We have measured 575-point for 5 days, and 187 points were used for structure analysis. By combining X-ray data, we determined the position and population of hydrogen atoms. As shown here, neutron experiments give a clear information of lighter atoms such like hydrogen and oxygen atoms.

As a test material to observe magnetic intensity, we measured the antiferromagnetic phase transition of YMn_2O_5[1] at low temperature. It was revealed that the magnetic peak appears at an incommensurate **q**-vector ($\frac{1}{2}$+δ,0,$\frac{1}{4}$ + α), and the position changes and locks at several points associated with the successive phase transitions complicated way. These behaviors correspond to the anomalies of the magnetic susceptibility, dielectric susceptibility, and appearance of spontaneous polarization. This experiment clearly shows the capability of the four-circle diffractometer.

2.3 Si bent monochromator

We are developing Si bent monochromator to use shorter wavelength. First, we constructed 9-Si 553 plates on the goniometer head to get 1.18 Å neutrons. However, the focusing mechanism was so poor that we could not get satisfactory vertical focusing. As an improved one, we developed new focusing mechanism so that each stepping motor rotates all of plates. Since the guide tube is not still super-mirror, we constructed Si422 monochromator to get 1.5 Å neutrons. Figure 2 shows the rocking curve of 422 reflection of nine Si plates after the plates are bent horizontally up to 70 % of broken-point. The assembling and tuning of the monochromator was performed at TAS2(T24). After fine tuning, peak positions and peak intensities are almost the same for all of plates.

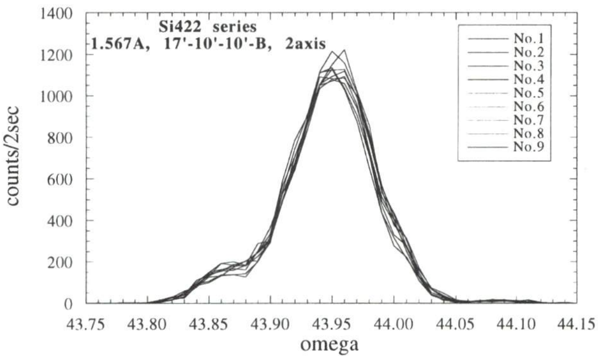

Fig. 2. Rocking curve of 422 reflection of 9-plates.

We have tested this monochromator quickly before the construction of replacing mirrors to super-mirror. The $2\theta_M$ angle is 90 ° and the wavelength is 1.557 Å. Vertical and horizontal focussing was beautifully performed, and the beam size became to 16x25mm^2 at about the receiving slit position.

Figure 3 shows the effect of vertical and horizontal bent of the monochromator. The left-hand side photographs show the beam intensity at 660mm and 1970mm positions from the monochromator. The right panels show the intensity profile as a section of the beam. The intensity distribution at 660 mm shows the structure coming from each plate since this position is not the focus point. In contrast, at about the focusing point, 1970mm, entire beam coming from 9-plates converging. In Fig.4, the beam size is shown as a function of the distance from the monochromator. As shown in the figure, the beam is doubly focused at about the sample position.

Because of the current neutron beam character in the guide-tube, the beam intensity of λ=1.557 Å with Si422 is about 20 times stronger than the case of Si553 (λ=1.18 Å), monitored by a standard sample. Si422 gives the $\lambda/2$ contamination. The observed $\lambda/2$ contamination is 0.13 %, slightly worse than that of PG002 monochromator with filter, but it seems practically satisfactory.

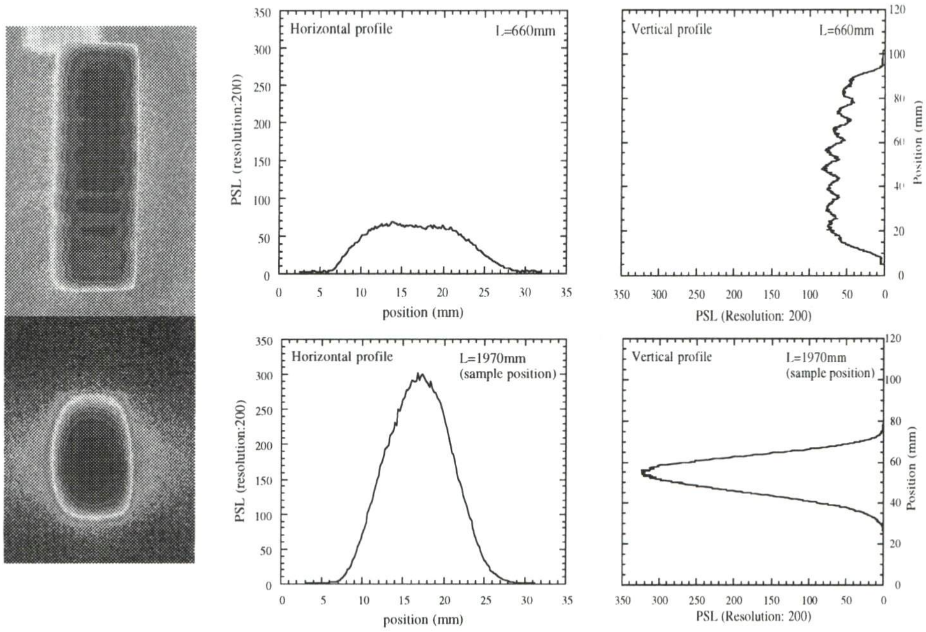

Fig. 3. Focusing effect of the bent monochromator.

2.4 *IP-camera*

As the demonstration of the short wavelength, we have taken an oscillation photograph of d-H2SQ by using a film made of a neutron Image-Plate[2] (BAS-ND2025 of Fuji Film Co.). Camera radius is 100mm, oscillation angle is 30 ° and exposure time is 2.5 hours. Beautiful picture was obtained with a good signal-to-noise ratio. The apparatus is still for test so that we can not discuss the data quantitatively. We plan to construct the dedicate camera-cassette to discuss more quantitatively.

2.5 *Effect of super-mirror*

We have tested the beam character for few days after a part of the beam guide was replaced to super-mirror. The monochromator is Si422 (λ=1.557 Å) and double focused. The beam profile is almost the same with the previous one. The intensity monitored by a standard sample shows a remarkable increasing, about three times stronger. On the other hand, the $\lambda/2$ contamination (λ=0.778 Å) also increases, about 10 times stronger, resulting in the ratio of the $\lambda/2$ contamination becomes 0.5 %. We can expect more gain after the entire beam guide is replaced to super-mirror, but the $\lambda/2$ contamination will also increase. In such a case, we will change the monochromator to Si553 (λ=1.18 Å).

§3. Summary

We have developed and constructed a new four-circle diffractometer(FONDER) and double-bent Si monochromator at T22 beam port of JRR-3M of JAERI. The performance of the diffractometer was evaluated and seems satisfactory. The test of the structure analysis was performed with λ=2.43 Å at the moment. Since the shorter neutron will be available soon, the number of diffraction observable will increase more than ten times. On that stage, this diffractometer becomes practically available for structure analysis. Even at the present stage, this four-circle diffractometer is also useful for the study of magnetic reflection, diffuse scattering etc. An oscillation photograph is also possible with a neutron Image-plate. This function will be a great help to search unknown new reflections, which will appear associated with a phase transition.

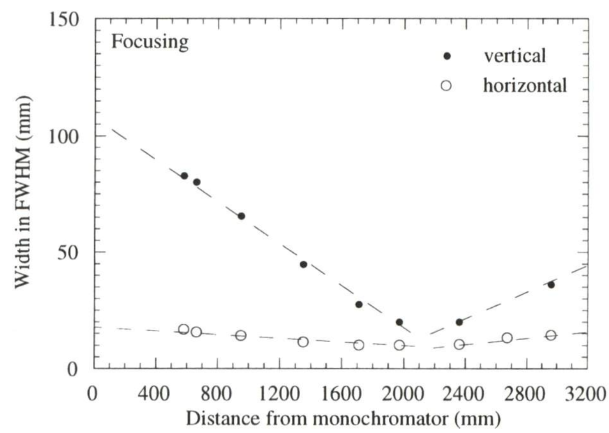

Fig. 4. Beam size as a function of the distance.

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