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Current status of a TOF-Laue single crystal neutron diffractometer SENJU

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Abstract. A TOF-Laue single crystal neutron diffractometer at the BL18 of MLF/J-PARC, SENJU, was designed for precise crystal and magnetic structure analyses under extreme environments such as low-temperature, high-pressure and high-magnetic field. On-beam test measurements of 4K cryostat, high-pressure cell and vertical-field superconducting magnet were carried out to evaluate the background from each device. All devices worked as planned and single crystal neutron diffraction measurements under extreme conditions on SENJU were successfully achieved.

1. Introduction

SENJU is a time-of-flight (TOF) Laue single crystal neutron diffractometer which was launched in March 2012 at the BL18 of MLF/J-PARC. SENJU was designed for crystal and magnetic structure analyses of inorganic/organic crystalline materials with small single crystal less than 1.0 mm^3 in volume [1,2]. To achieve the purpose, SENJU has 37 scintillation area detectors [3] to measure many Bragg reflections simultaneously as shown in figure 1 and use relatively short wavelength incident neutron (λ > 0.4 Å) to cover wide range of reciprocal space. SENJU was also designed for measurement under extreme conditions such as low-temperature, high-pressure and high-magnetic field [1,2]. Single crystal neutron diffraction measurement under extreme condition is an important analytical technique in the field of materials science because this technique is one of the best way to obtain the precise crystal and magnetic structures of many materials which shows interesting properties such as magnetism, superconductivity or ferroelectricity under various extreme conditions. Many single crystal diffractometers which have wide area detectors, for example, SXD [4] at ISIS, VIVALDI [5] and CYCLOPS [6] at ILL, KOALA [7] at ANSTO have a flange at the top of the sample chamber to accept various types of sample environment devices. SENJU also has a vacuum sample chamber with the MLF standard size flange to accept various types of sample environment devices which are developed and used in MLF to achieve various types of extreme conditions.

Some test measurements in the commissioning phase showed that 0.4~4.4 Å wavelength incident neutrons are available as the 1st frame and 4.6~8.8 Å as the 2nd frame, estimated neutron intensity at the sample position with 1MW accelerator power is 0.6 x 10^6 $n \cdot s^{-1} \cdot mm^{-2}$ in the standard mode and 1.3 x 10^6 $n \cdot s^{-1} \cdot mm^{-2}$ in the focused mode (with a insertion focusing guide). In addition, the measurements with empty vacuum sample chamber showed that the background is 1/1000 lower than the neutron scattering from a ϕ5 mm V-Ni null alloy [2]. Figure 2 shows a typical diffraction image at SENJU, a TOF Laue neutron diffraction image of a ϕ2 mm size ruby single crystal.

After the initial commissioning of SENJU, commissioning and test measurements of three sample environment devices, 4K cryostat with 2-axes goniometer, piston cylinder type high-pressure cell and vertical-field superconducting magnet were conducted to check the operation and to evaluate the background from the devices and S/N ratio of diffraction spots when a sample crystal was mounted on the devices. This paper describes the results of commissioning and on-beam test measurements of those sample devices.

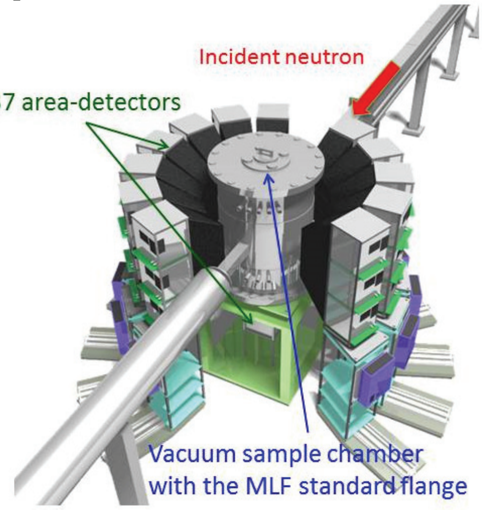

Figure 1. Schematic view of SENJU.

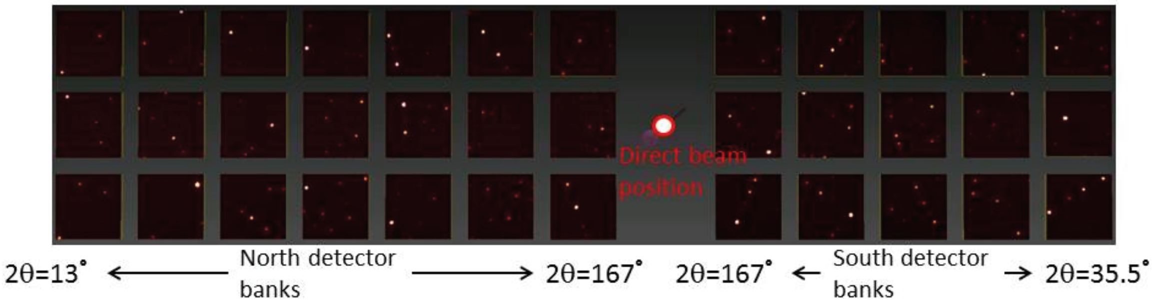

Figure 2. A typical diffraction image at SENJU, a TOF Laue neutron diffraction image of a ϕ2 mm size ruby single crystal.

2. Commissioning and on-beam test measurements of sample devices

2.1. 4K cryostat with 2-axes goniometer

4K cryostat is one of the most important sample environment devices for diffraction measurements of materials science. In usual low temperature experiment with 4-circle single crystal neutron diffractometer, a cryostat is mounted on a large goniometer equipped with ω, χ and ϕ axes to rotate the sample crystal to access to arbitrary points in the reciprocal space. However, because SENJU has no space to set a large goniometer at the sample position, we developed a new piezo-based low temperature 2-axes fixed-χ goniometer system [8]. Figure 3 shows the photograph of the goniometer at the cold-head of a closed-cycle cryostat. This goniometer has two piezo rotating devices (ω, ϕ) and a fixed-χ arm to change the crystal orientation under vacuum and low temperature condition. Because of the poor thermal conductivity of the piezo rotators, additional copper mesh wires were attached between the cold-head and the χ arm, and the χ arm and the sample holder (XY-stage) as heat paths.

The result of a cooling test showed that the lowest temperature at the sample position was 3.8 K and the cooling time from room temperature to the lowest was 4.5 hours as shown in figure 4. Two piezo rotators, attocube systems AG ANR240/RES for the ω axis and ANR220v/RES for the ϕ axis, stably work even in the lowest temperature. An on-beam test measurement of this cryostat showed that there was no additional background from the cryostat except weak powder diffractions from two cylindrical radiation shields in the vacuum chamber in which four 5 μm thickness Al foils are on the direct beam path.

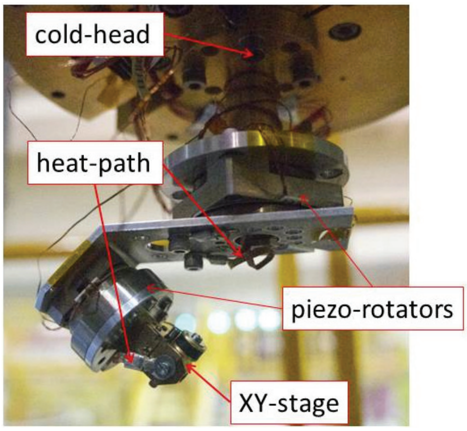

Figure 3. Photograph of the 2-axes goniometer at the cold-head of the cryostat.

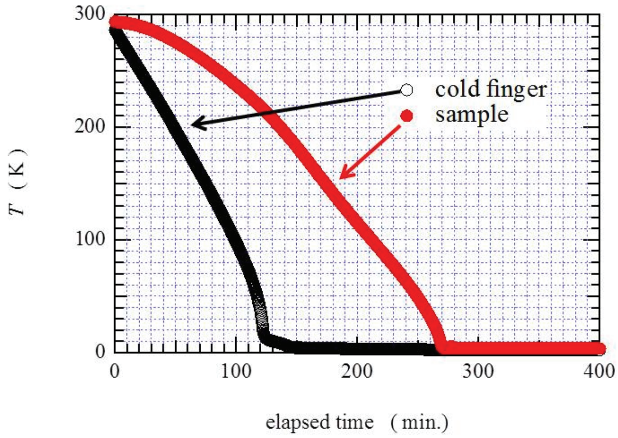

Figure 4. Cooling curves of the 4 K cryostat of SENJU. Red dots show the temperature change of the sample position.

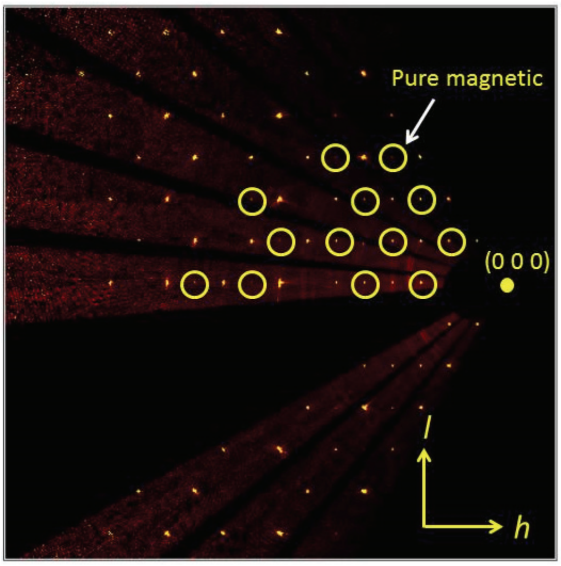

Figure 5. *(h0l)* plane of the diffraction from MnF_2 at 4.3 K. Yellow circles indicate pure magnetic reflections.

To confirm the performance of the cryostat, we carried out a structure analysis of the low-temperature phase of a MnF_2 single crystal, which shows paramagnetic – antiferromagnetic phase transition at 75 K [9]. The reported space group is $P4_2/mmm$ and cell parameters are a = b = 4.87 Å and c = 3.30 Å. 2 x 2 x 2 mm MnF_2 single crystal was mounted on the XY-stage at the top of the goniometer and temperature of the cold-head was set to 4.0 K. Finally, the temperature gauge at the XY-stage in the figure 3 indicated 4.3 K. In the diffraction measurement, neutron exposure time was 10 hours. Figure 5 shows an observed diffraction image of the reciprocal space, (h0l) plane. Many pure magnetic reflections of the antiferromagnetic phase (white circles) were observed. Powder pattern from the cryostat is very weak comparing to the Bragg peaks from the sample crystal. In this measurement, 2048 reflections including 51 pure magnetic reflections (I > 3σ(I)) were observed. After the refinement of the 14 structure parameters including magnetic structure parameters, final R value was 5.28% (I > 3σ(I)) and obtained magnetic moment of Mn was μ(Mn) = 4.7(1) μ_B. Those were acceptable values. This result indicates that the cryostat has enough cooling capacity and the neutron scattering from the cryostat rarely disturbs the structure analysis.

2.2. High-pressure piston-cylinder cell

Varying pressure is sometimes very important to explore new properties of a functional material. A compact piston-cylinder type high-pressure cell, which has an ideal geometry because the apparatus has a wide opening accessible direction for incoming and scattered neutrons, made from copper-beryllium alloy (< 2 GPa) was tested on SENJU. A taurine single crystal with 1.5 x 1.5 x 1.5 mm size was enclosed in the pressure cell together with deuterated glycerol and pressurized up to 1 GPa. Accelerator power was 300 kW and the exposure time was 6 hours. Even through the intensity of neutrons scattered from the pressure cell was relatively high, many distinct Bragg reflections from the taurine crystal were observed as shown in figure 6. The result shows that this piston-cylinder cell can be used as one of the high-pressure environments of SENJU.

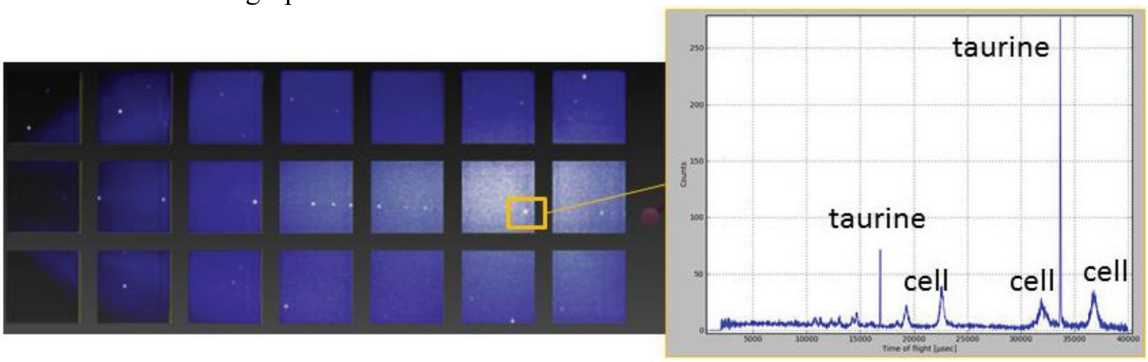

Figure 6. A neutron Laue image and a TOF profile of the taurine crystal in the piston-cylinder type high-pressure cell measured at SENJU.

2.3. Vertical-field superconducting magnet

Magnetic field is an important sample environment in researches of magnetism. To realise a neutron scattering experiment in magnetic field, a vertical-field magnet was developed as a MLF standard sample environment device. The first test of the magnet showed that the maximum magnetic field was 6.85 T and minimum temperature was 50 mK respectively. Then, the magnet was set in the vacuum chamber of SENJU without sample and exposed to neutron beam at room temperature to check the background from the magnet. Figure 7(a) shows a diffraction image of all detectors. Whereas the upper and the lower detectors were hidden by the magnet as expected, all the equatorial plane detectors and the bottom detector are available for diffraction measurement with the magnet. Figure 7(b) shows the 1st frame (0.4~4.4 Å) time-of-flight neutron spectrum of one of the highest angle detectors from the magnet. As expected, many Bragg peaks from the magnet were observed. On the other hand, as shown in figure 7(c), there is no Bragg peak from the magnet in the spectrum of the 2nd frame (4.4~8.8 Å), important wavelength region for measurements of magnetic reflections in the field of materials science.

A test measurement of the magnet with a sample crystal was also carried out. A single crystal of $CeCoGe_3$ [10] with 3.0 x 1.5 x 1.5 mm size was set in the magnet. Measurement temperature was 1.5 K, magnetic field was 0.5 T, accelerator power was 300 kW and neutron exposure time was 2.5 hours. The diffraction image and a TOF profile in figure 8 shows that Bragg peaks of the sample crystal were much higher than the peaks of Al from the magnet even in the 1st frame.

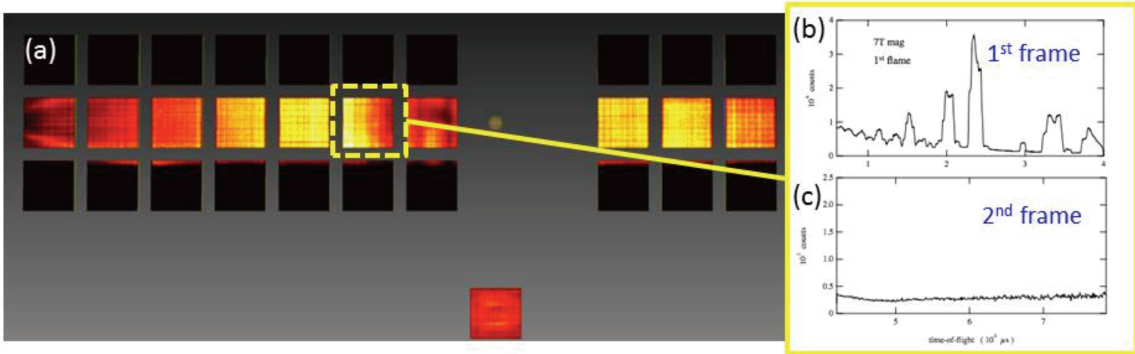

Figure 7. Neutron scattering from the magnet measured at SENJU. (a) Scattering image of the area detectors. (b) The 1st frame (0.4 – 4.4 Å) TOF profile of the detector enclosed by the dashed line. (c) The 2nd frame (4.6 – 8.8 Å) TOF profile of the detector enclosed by the dashed line.

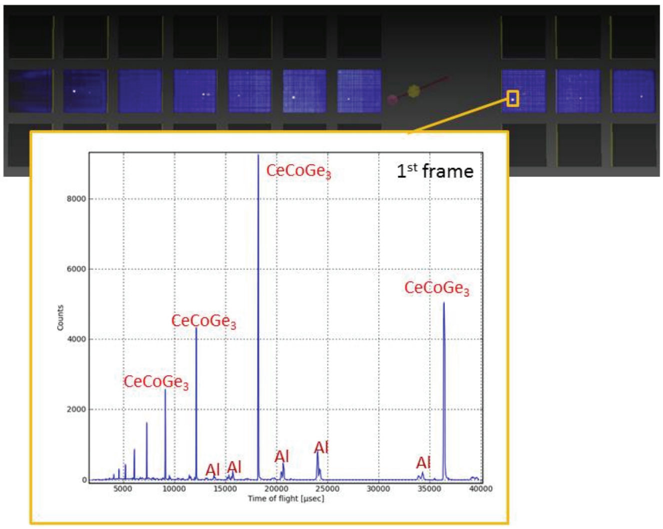

Figure 8. A neutron Laue image and a TOF profile of the $CeCoGe_3$ crystal in the magnet measured at SENJU.

3. Summary

Commissioning and on-beam test measurements of the devices at SENJU showed that the all three devices worked as planned on SENJU. The 4 K cryostat has enough cooling capacity and the powder pattern from the cryostat rarely disturbed the structure analysis. In cases of the magnet and high-pressure cell, powder patterns from the devices were observed but Bragg peaks from the samples were clearly observed. Those results indicate that the main purpose of SENJU, crystal and magnetic structure analyses of inorganic/organic crystalline materials under extreme conditions, was successfully achieved. The devices described in this paper are available for the general user program at SENJU. In near future, we will develop new collimators for the magnet and the high-pressure cell to reduce the powder patterns and improve the quality of diffraction data.

References

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