X-ray Powder Diffraction Camera for High-Field Experiments

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Abstract. We have designed a high-field X-ray diffraction (HF-XRD) camera which will be inserted into an experimental room temperature bore (100 mm) of a conventional solenoid-type cryocooled superconducting magnet (10T-CSM). Using the prototype camera that is same size of the HF-XRD camera, a XRD pattern of Si is taken at room temperature in a zero magnetic field. From the obtained results, the expected ability of the designed HF-XRD camera is presented.

1. Introduction

Recently, high performance materials controlled by magnetic fields such as ferromagnetic shape memory alloys [1,2], giant magnetostrictive compounds [3], magnetic refrigerants [4-7] and so on have been intensively studied all over the world. In these compounds, a magnetic field plays an important role for application-oriented research. Many compounds of the above system exhibit a structural transformation or a large lattice distortion, accompanied by the magnetic phase transition at the working temperature. Therefore, it is very important for developing these materials to clarify structural properties microscopically in magnetic fields. Especially, it requires high magnetic fields to investigate the potential of these materials at the early stage of the development.

So far, neutron and synchrotron X-ray diffraction experiments have been performed in high magnetic fields over 15 T using split-pair type superconducting magnets [8] and 25 T using pulsed magnets [9, 10], respectively. In addition, a 30 T hybrid magnet has been also planned for neutron experiments at the Hahn-Meitner Institute, Germany, collaborating with the National High Magnetic Field Laboratory, USA [11]. However, it is difficult to investigate many materials in detail using such big facilities, because of limited machine time and experimental cost.

An X-ray powder diffraction (XRD) experiment in house is a very general and useful method to investigate the structural properties of materials. Some researchers have independently reported compact X-ray diffraction systems using split-pair type superconducting magnets [12-14]. In the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University (HFLSM), the high-field X-ray powder diffraction (5T-HF-XRD) apparatus has been developed [13]. This apparatus consists of a cryocooled split-pair superconducting magnet and \( \theta -2\theta \) scan-type X-ray diffractometer. The apparatus has provided unique experiments in magnetic fields \( B \) up to 5 T and 8 – 320 K temperature \( T \) range. By using this system, several important results have been reported up to now [15-17]. However, it is difficult to upgrade an X-ray diffractometer combined with high field magnets over 10 T. Development of a split-pair superconducting magnet, which can generate more
than 10 T with a horizontal room temperature gap for X-ray beam path, has a lot of problems like a mechanical strength issue to be solved and needs a huge cost to construct.

In this work, we have started to develop an X-ray diffraction apparatus in high field measurements. A diffractometer is to be inserted into an experimental room temperature bore (100 mm) of a magnet. In this case, we abandoned the conventional diffractometer method but adopted the Debye-Scherrer camera, which is simple and not influenced by the magnetic field. However, it is necessary to examine whether the experimental conditions are realistic in high magnetic fields, before developing the camera system. Furthermore, it should be carefully examined that the camera system with a power supply, an X-ray source, the controlling device, and safety devices will work in large leakage fields (~0.1 T) of the high field magnet. This paper describes a basic design of a HF-XRD camera system and some results obtained using its prototype camera in a zero magnetic field. The obtained results suggest that the XRD pattern is obtained in high magnetic fields.

2. Design of a high-field X-ray powder diffraction camera system

Figure 1 shows the schematic illustration of the high-field X-ray powder diffraction (HF-XRD) camera system at present, which is utilized for a 10T-100 mm bore cryocooled superconducting magnet (10T-CSM). The apparatus mainly consists of a Debye-Scherrer camera (TRY SE Co. Ltd), an X-ray source, a power supply, a chiller and 10T-CSM.

An X-ray source is set outside from the magnet end, which is 472 mm apart from the center (sample position) of the magnet. We adopt a vacuum-tube type X-ray source. Therefore, we can easily change the wavelength by selecting a target among Cu, W, Fe, Co and Mo. The maximum output power of an X-ray source is 2 kW. A collimator size is ø 1.0 or 0.5 mm. The X-ray pass from the source to the collimator is evacuated by a vacuum pump.

A diffraction pattern is collected on an image plate (IP) detector in the Debye-Scherrer camera. The pixel size of the IP leader will be 0.025 mm × 0.025 mm. The diameter of the camera is 80.2 mm. A measurement range in 2θ is from -140˚ to 140˚. A powder sample is set in a capillary (ø 0.5 mm) on the center axis of the camera. In our system, the IP can be changed with the sample fixed in high magnetic fields.

![Figure 1](image.png)

**Figure 1.** Schematic illustration of the high-field X-ray powder diffraction (HF-XRD) camera system which is combined with a 10T-100 mm bore cryocooled superconducting magnet (10T-CSM).
3. Estimation of the performance of HF-XRD using a prototype camera

Before installing the HF-XRD camera in 10T-CSM, we estimated the performance of the camera using a prototype one without the magnet. The diameter (80.2 mm) of the prototype camera is the same size of the HF-XRD camera. Figure 2 shows the XRD pattern of Si powders at room temperature in a zero magnetic field, which is collected by the IP detector (Fujifilm BAS III) using the prototype camera. The obtained pattern can be indexed, as shown in this figure. This measurement is performed in air using Cu Kα radiation (1.2 kW), a collimator (ø 1.0), a Ni filter and IP. The X-ray beam path from the source to sample is 150 mm, and the exposure time is 15 minutes.

Figure 2. X-ray powder diffraction pattern of Si at room temperature in a zero magnetic field. The pattern is collected by an image plate detector using a prototype camera that is same scale of the HF-XRD camera.

Figure 3 shows the 400 and 331 diffraction patterns, which is a close-up of the pattern in Fig. 2. We confirmed that the pixel size is 0.1 mm × 0.1 mm, indicating that the 2θ–position is determined by the resolution of 0.13°. As we know, the pixel size is limited by an IP-reader. In this measurement, we utilized an IP-reader (TRY-XIA). However, we will use another high resolution IP-reader with the pixel size of 0.025 mm × 0.025 mm in our HF-XRD camera system. Therefore, we will determine the diffraction position with 0.033° on 2θ unit, indicating that lattice parameters can be determined with high significant figures.

Figure 3. 400 and 331 diffraction patterns, which are close-up of the pattern in Fig. 2. A pixel size is 0.1 mm × 0.1 mm.

The beam pass of the HF-XRD camera is about three times larger than that of the prototype one, indicating that the intensity of the X-ray beam decreases to about 10%. From this result, we roughly estimated that the exposure time is required about 60 minutes for one picture when the output power of the X-ray source is 2 kW and the X-ray pass is evacuated in the HF-XRD camera. This experimental condition is realistic, and the systematic measurement is sufficiently possible in high magnetic fields.
As seen in Fig. 3, the line width of the 331 diffraction is 0.9 mm, which corresponds to 1.2° on 2θ unit. This is two times larger than that obtained by our 5T-HF-XRD. However, it is expected to observe the structural transformation, because the XRD patterns are drastically changed. If we utilize the collimator of ø 0.5 mm, the line width decreases further. From these results, we expect that field-induced structural transformation can be observed using the HF-XRD camera.

We should consider influence of leakage fields from the magnet on the HF-XRD camera. We measured the leakage field distribution of 10T-CSM. The leakage field at 472 mm away from the magnet center is less than 0.07 T for the center field of 10 T. In the case of our 5T-HF-XRD apparatus, an X-ray tube is in the leakage field of 0.08-0.02 T and works well without a magnetic shield in such a condition. Therefore, it is expected that no trouble will be caused on X-ray source by the leakage field in this design, but we also consider the preparation of the magnetic shield for the X-ray source of HF-XRD camera.

From these examinations using the prototype camera, the HF-XRD camera was produced. Figure 4 shows a photo of the HF-XRD camera system that is combined with the 10T-CSM. The HF-XRD camera system examination in high magnetic fields is now in progress. The results will be published in elsewhere in the near future.

**Figure 4.** Overall view of the high-field X-ray powder diffraction (HF-XRD) camera system that is combined with a 10T-100 mm bore cryocooled superconducting magnet (10T-CSM).

### 4. Summary
A High-field X-ray powder diffraction (HF-XRD) camera was designed in order to carry out HF-XRD experiments in 10 T generated by a cryocooled superconducting magnet (10T-CSM) at a laboratory site. Using the prototype camera that is the same scale of the HF-XRD camera, we measured a XRD pattern of Si at room temperature in a zero magnetic field. From the results, we expected that one picture of a XRD pattern will be taken about 60 minutes using the HF-XRD camera in 10 T. In addition, obtained results indicate that the high resolution HF-XRD pattern is obtained and the structural transformation can be observed.

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