Hot pressing of Ge crystals toward a reflection-plane-selective neutron monochromator

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Abstract. We determined a hot-pressing condition of temperature and pressure to fabricate a Ge-crystal monochromator. After hot pressing under the optimal condition, a reflected beam by the Ge crystals shows an appropriate peak width (~0.3° in FWHM) and an enhanced peak reflectivity (~40%). The feasibility of reflection-plane-selective monochromator, which consists of Ge crystals, is confirmed by magnetic powder neutron diffraction in the high-Q survey mode as well as in the fine Q-resolution mode.

1. Introduction

Tohoku University has sufficiently contributed to a high growth of neutron science in Japan since 1970’. At present, Institute for Materials Research (IMR) of Tohoku University owns two neutron-scattering instruments, AKANE and HERMES, at thermal neutron beam ports of a research reactor of JRR-3 in Tokai, Japan. AKANE is a triple-axis spectrometer [1] and HERMES is a powder diffractometer with multi detectors [2]. There are two missions for IMR neutron group. One is the scientific and material research under a user program in corporation with Universities in Japan. The other is to develop new experimental techniques for neutron scattering.

Here, we present recent activities for neutron monochromator renovation using Ge crystals. One characteristic feature of Ge is that, in principle, no second-harmonic waves contaminate in the reflected beam from a monochromator Ge (hhl) owing to the forbidden reflection (2h 2h 2l) of diamond structure, where h and l are odd integers. Hence, any incident energy is available without filters. Another feature of Ge should be a high quality of single crystals. Owing to nearly perfect crystals, a well collimated neutron beam is extracted from the crystal monochromator. On the other hand, due to extinction effects in such the high-quality crystal, a neutron reflectivity is severely suppressed. To overcome this disadvantage in Ge, we introduce mosaic crystals by pressing at high temperatures below its melting point (938°C). In this manuscript, we first report an optimal hot-pressing condition for neutron monochromator with respect to temperature (T) and pressure (P). Second, we introduce a reflection-plane-selective (RPS) Ge element, by which (hhl) and (h'hl') Bragg reflections are easily switchable with a small crystal-angle rotation. In preliminary measurements of magnetic neutron diffraction, we confirmed a good performance of the RPS monochromator.
2. Experimental methods

2.1. Procedure of hot pressing

High-quality Ge single crystals with (hhl) plane were purchased from a private company in a disk form (φ100 mm × t6 mm). The cutting faces were selected to (113) plane and (331) plane for AKANE and HERMES, respectively. To survey an optimal hot-pressing condition, the crystals were cut into nearly square pieces (20×18×6 mm³) along [1, 1, 0] and [1, 1, −(2h/l)] axes, as shown in Fig. 1(a). A vertical furnace, which consists of hinged two semicircular parts with a maximum power of ~1.2 kW, was prepared for present hot pressing [Fig. 1(b)]. A flat temperature distribution is attainable locally; for example, 800±1°C in area of 20×20 mm² at the furnace center. During hot pressing, the Ge crystals were placed inside the furnace. One Ge piece was set between alumina plates and put further between stainless-steel blocks [Fig. 1(b)]. The procedure of hot-pressing is as follows. We raise the temperature to a target value in 2.5 hours, wait 20 minutes for thermal equilibrium, and press to a target pressure in 10 seconds. After keeping the hot-pressing state in 30 seconds, the furnace heater is turned off first. When the temperature decreases to 650°C, the pressure is released in a second and then the Ge crystal is naturally cool down in the furnace. The effect of hot pressing on neutron Bragg reflections was characterized on AKANE under a double-axis mode by using a well-collimated (guide-15'-S-30') and small-sized (2.5×3 mm²) beam.

![Figure 1](image)

Figure 1. (a) Orientation of Ge crystals used for hot pressing. (b) Inside the hot-pressing furnace, showing a sandwich structure about Ge with alumina plates and SUS blocks. (c) Principle of the RPS monochromator, using (331)-cutting-face crystals. The offset angle between (331) and (551) scattering vectors is about 5°.

2.2. Principle of RPS monochromator

By using a currently running Ge monochromator at HERMES, we tested how far the RPS function works for neutron scattering measurements. The (331)-reflection monochromator was set up with [1, 1, 0] axis aligned vertically, so that (331) and (551) reflections were switchable through a crystal-angle rotation by ΔθM ≈ 5° (ΔθM/θM ~ 10%) [Fig. 1(c)]. Owing to the small ΔθM, an effect of asymmetric reflection of (551) using (331)-face-cutting crystals may be weak. Since the scattering angle is fixed to 2θM = 90° at HERMES, the wave length of reflected beam corresponds to λ = 1.82 and 1.11 Å for (331) and (551) reflections, respectively. Note that no filter was inserted in the beam path.

Neutron powder diffraction was measured on HERMES with such a RPS monochromator using a rare-earth metal compound ErB₂C₂, in which an isotope ¹¹B is enriched. The compound shows antiferromagnetic transitions below T_N = 15.9 K and T_I = 13.0 K with a large magnetic moment of 8.3μ_B/Er [3]. Single crystals of ErB₂C₂ were grown by the Czochralski method with a tri-arc furnace, and then we powdered the crystals under He gas with a mortar. The powder sample of 8 g in weight was packed into a vanadium cell of φ9 mm, and set in a refrigerator.
Figure 2. (a) (113)-reflection profiles before and after hot pressing at \( P = 36 \) MPa and \( T = 750 \)°C. A transmission data after hot pressing is also displayed. (b,c) Hot-pressing effects on the FWHM and the peak intensity in \((P,T)\) parameter phase. The contour lines are drawn as a guide, based on 10 data as shown by circles. The closed circles represent the condition for a sample in (a). (d) The peak intensity (open circle) and the angle-integrated intensity (closed circle) as a function of the FWHM. The data points shown by a triangle originate from Ge crystals with a different shape \((12 \times 80 \times 16 \text{ mm}^3)\). The reflected-beam feature in the hatch is suitable for our neutron monochromator.

3. Results of hot pressing

Hot-pressing conditions for Ge (113)-face crystals were surveyed in several \((P,T)\) states, where \( P \) and \( T \) correspond to the target pressure and temperature, respectively. A typical rocking curve from a hot-pressed sample is shown in Fig. 2(a), together with a result from a non-pressed sample. A substantial change in both the peak width and intensity manifests an introduction of mosaic crystals through the hot pressing. Figures 2(b) and 2(c) show the FWHM and the peak intensity of (113) Bragg peak, respectively, in a limited region of the \((P,T)\) phase. A drastic increase (decrease) in the FWHM (the peak intensity) occurs above 750 °C (below 650 °C). Concerning the FWHM, no significant difference was observed between \([1,1,0]\) and \([1,1,−2/3]\) directions, unlike a previous report [4]. The isotropic mosaic in present case could be due to the homogeneous temperature distribution. Figure 2(d) displays a re-plot of Figs. 2(b) and 2(c), showing the peak intensity and the angle-integrated intensity as a function of the FWHM. The peak intensity rapidly increases when the FWHM becomes wider than the angle resolution \((\sim 0.08^\circ)\), and takes a maximum at around \( 0.2^\circ \); this is consistent with a beam divergence of guided neutrons \((\sim 0.2^\circ)\). On the other hand, the angle-integrated intensity increases linearly with the FWHM and becomes ten-times as large as before hot pressing. The flat behavior seen at beyond \( 0.3^\circ \) suggests a saturation in effective volume of the monochromator element; namely, a limit of mosaic-crystal introduction in this method. On account of a medium \( Q \) resolution of HERMES and inelastic scattering experiments at AKANE, we regard \( \sim 0.3^\circ \) as an acceptable FWHM for the IMR neutron instruments. Therefore, a thermal treatment under \((P,T) \sim (35−40 \text{ MPa, 650−750 K})\) is found to be suitable for us in producing Ge monochromators. In this case, the peak reflectivity of Ge (113) reflection attains to \( \sim 40 \% \) [Fig. 2(a)], which corresponds to more than 80% of theoretical peak reflectivity [5].

4. Performance of the RPS monochromator

As a preliminary step, magnetic neutron powder diffraction of antiferromagnetic \( \text{ErB}_2\text{C}_2 \) was measured by using the RPS monochromator. Figures 3(a) and 3(b) show magnetic cross sections in cases of \( \lambda = 1.11 \text{ Å} \) and 1.82 Å, respectively. First, the agreement between the two profiles
at the low-Q region guarantees validity of the RPS function. Second, as shown in a magnified plot [Fig. 3(c)], magnetic cross sections are likely observable up to $\sim 8$ Å$^{-1}$, or (630) and (631) magnetic reflections when the short wave-length beam is used. This maximum $Q$ is quite large for unpolarized neutron diffraction if we consider a squared magnetic form factor $f_m^2$, which falls off substantially at high $Q$.

At present, however, the much longer observation time is inconvenient for measurements when the short wave-length neutrons are used [Fig. 3(a)]. According to a simple estimation of reflected beam intensity based on $\lambda^3$ and a beam flux in the supermirror-guided neutrons, there is room to shorten the observation time into a half in case of Ge (551) reflected beam. In a new RPS monochromator of Ge (hhl), which we are now fabricating, a vertical focussing function is reinforced and a dimension of monochromator becomes vertically twice as large as that of current running one. On the other hand, the fine $Q$ resolution in the long wave-length case [Fig. 3(d)] is proper for detailed structural analysis. Therefore, along each experimental purpose we can choose a suitable measurement mode if the RPS monochromator is available.

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