Fabrication of X-ray absorption grating using an ultracentrifuge machine

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We propose a novel approach to fabricate hard-X-ray optics using an ultracentrifuge machine. As a demonstration, we fabricated an X-ray absorption grating made of gold with a pitch of 30 μm and an effective depth of 44 μm in 67-μm-depth grooves (a filling rate of 65 %, as same as the filling rate of random packing for equal-sized spheres in a jammed state (64 %)). Our approach is cost-efficient and has a high production-rate and material-selectivity, and will be widely used for fabricating optics not only for X-rays but also neutrons in the near future.

Hard-X-ray optics requires a sufficient thickness in the X-ray propagation direction because of weak interaction of hard X-rays with matters, while, fine patterning is often desired in the plane perpendicular to the X-ray propagation direction. For example, absorption transmission gratings used in X-ray grating interferometry,[1–23)] which is a phase-contrast imaging technique,[24–26)] requires thicknesses more than a few tens of μm even if heavy elements such as Au, Pt, and Pb are used, while, pitches of a few or a few tens of μm are required for effectively using spatial coherence of X-rays. Other examples are hard X-ray lenses,[27–36)] which are desired to have well-controlled fine structures with a precision less than 10 nm in the plane perpendicular to the X-ray propagation direction, while at least a few μm thickness is necessary even in the case of phase-type hard X-ray optics. Thus, hard-X-ray optics often takes a high-aspect-ratio shape, which is generally not easily fabricated.

Although it is difficult to fabricate high-aspect-ratio hard-X-ray optics, several approaches have so far been reported for X-ray absorption grating.[37–56)] Electroplating techniques combined with X-ray lithography[38–41,43,45,50,54] and deep etching[37,42,57] have so far been widely used. They have provided high-quality and large-area X-ray absorption gratings, and the technique combined with deep dry etching[42,57] should be promising in terms of cost-effectiveness, but electroplating normally requires more than a few hours for a few tens of μm thickness. Recently, high-throughput techniques such as microcasting[46,47,51,55,56] and metallic glass imprinting[48,52,53] were proposed. Although they are low-cost and the imprinting technique can also be used even for neutron absorption grating,[58,59] specific materials, in which atomic density of heavy atoms are not necessarily high, are required.

In this paper, we propose a novel approach to fabricate hard-X-ray optics using an ultracentrifuge machine. Our approach has not only a high throughput and high production-rate but also a high material-seclectivity. As a demonstration, we fabricated an X-ray absorption grating with a pitch of 30 μm by filling 67 μm-depth grooves with gold particles using the ultracentrifuge machine.

The fabrication process for the X-ray absorption grating is very simple, consisting of only two steps, mold fabrication and centrifugal filling. First, a 8 mm $\times$ 8 mm-size 30-μm-pitch Si grating (mold) whose grooves have a width of 20 μm and a depth of 67 μm was prepared on a 525-μm-thick Si wafer by inductively coupled plasma (ICP) etching (Fig. 1 (a)). Next, an ultracentrifugal filling of Au particles was carried out by using an ultracentrifuge machine (CS120FNX, himac Koki Holdings Co., Ltd.) with a swinging bucket rotor (S50ST, himac Koki Holdings Co., Ltd.). Au particles (diameter of 1.5 μm, TAU-150, Tokuriki Honten Co., Ltd.) were used with no further modification. The Au particles were dispersed in water (concentration of the particles was 1.65 wt%) and sonicated for 1 hour to be monodispersed by using high-power ultrasonic generator (UT-605HS, Sharp Corporation). 7 mL of Au particles suspension was poured into centrifugal tubes, in which the Si grating mold was placed on a holder that was custom-made for this purpose (Fig. 1(b)). The ultracentrifugal filling was made at a rotation speed of 40,000 rpm (an acceleration of 162,000 G) for 1 hour at 4°C. After Au filling, the transparent supernatant water was collected gently from the tubes with pipettes to avoid the particles from moving out of the grating and the tubes were put into an oven heated at 70°C for completely drying the Si mold filled with Au particles.

Figure 2 (a) shows a photograph of the fabricated grating, and Figs. 2 (b) and (c) are cross-sectional images obtained by scanning electron microscopy (SEM). It can be seen that the grooves of the mold were uniformly filled with the Au particles.

The fabricated X-ray absorption grating was evaluated by using a one-dimensional X-ray Bragg magnifier.[60–68] The experiment was performed at BL-14C, Photon Factory, KEK, Japan, where an X-ray beam from a vertical wiggler monochromatized by a Si (2 2 0) double-crystal monochromator is available. We used a 6 mm (horizontal) × 70 mm (vertical) X-ray beam (photon density: 10^{8-9} photons/mm^2/sec) with an energy of 33.3 keV, which was determined from the K-edge of iodine. Figure 3 (a) shows the experimental setup of the Bragg magnifier, consisting of a sample, a highly asymmetric Si crystal, and an X-ray image detector. The grating was used as the sample, which was located in front of an asymmetrically cut Si(2 2 0) crystal with a miscut angle of 5.285°. The projection image of the sample was magnified 38.7 times in the horizontal direction by the crystal to be captured by a charge-coupled device (CCD)-based X-ray image detector with an effective pixel size of 9 μm (Hamamatsu Photonics C9300-124A), where the CCD (4000 × 2672 pixels) was connected to a 15-μm-thick P43 (Gd_2O_2S:Tb) screen with a 1:1 fiber coupling. Figure 3 (b) is the transmittance image of the sample obtained from the magnified projection image, and Fig. 3 (c) is the line profile along the dotted line in Fig. 3 (b). From the transmittance image, it turned out that effective thickness of Au in the grooves was 44 μm. Since the depth of the groove was 67 μm, the filling ratio of Au was 65 %, which is as same as the random packing ratio for equal-sized spheres in a jammed state (64 %).[69] The standard deviation of the effective thickness was estimated to be 1.5 μm, which allows the grating to be used in X-ray grating interferometry.

The advantage of our approach over the previously reported casting and imprinting methods is material-selectivity. In the above demonstration, we used Au monodispersive particles with a diameter of 1.5 μm, but other particles should also be available: there should be many variations of materials for the suspension. We should be able to use smaller particles with diameters up to 100 nm, which enables to fabricate a finer pitch grating, but it is expected that Brownian motion of particles after stoping centrifugal rotation limit the smallest size of the particles available. Wettability of the surface of mold to dispersion liquid should another factor that affects filling rate. Note that we should be able to fabricate a neutron absorption grating with an unachieved effective thickness by using, for example, particles of gadolinium oxide.

The only problem for X-ray absorption grating used in X-ray grating interferometry is that we can use a mold with a size less than 10 mm × 10 mm in the case of a normal centrifuge machine, which is sufficient for synchrotron X-ray imaging but insufficient for medical diagnostics using a laboratory X-ray source. Tiling[50,54] is a solution of this problem. Because, in the case of the centrifugal machine, we used, 4 or 8 samples can be simultaniously rotated, we can fabricate a larger grating by tiling small pieces of gratings fabricated in a centrifuge process. Thus, our approach has a large potential for fabricating low-cost and unprecedent effective thickness gratings with a high-throughput in the near future.

In summary, we proposed an approach to fabricate hard-X-ray optics using an ultracentrifuge machine. As a demonstration, we successfully fabricated an X-ray absorption grating made of gold particles with a filling of 65 %, which is as same as the random packing ratio for equal-sized spheres in a jammed state (64 %). Our approach is cost-efficient and has a high production-rate and material-seclectivity, and will be widely used for fabricating optics not only for X-rays but also neutrons in the near future.

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Fig. 1. Fabrication process of X-ray absorption grating with centrifugal machine. (a) Fabrication of Si mold by using inductively coupled plasma (ICP) etching. (b) High speed rotation of a centrifugal tube, in which Au particle suspension and a Si mold placed on a custom-made holder for high-speed rotation and ultracentrifugal filling.

Fig. 2. (a) Photograph of fabricated grating. (b) Cross-sectional image obtained by scanning electron microscopy (SEM) ((c) magnified image of rectangular area in (b)).

Fig. 3. (a) Top view of experimental setup of one-dimensional Bragg magnifier at BL-14C, Photon Factory, Japan. (b) X-ray transmittance image of fabricated grating magnified in horizontal direction (X-ray energy: 33.3 keV, gray scale: 0-1). Scale bar shows size in horizontal direction before magnified. (c) Line profile along dotted line in Fig. 3 (b). Horizontal axis represents distance before magnified.

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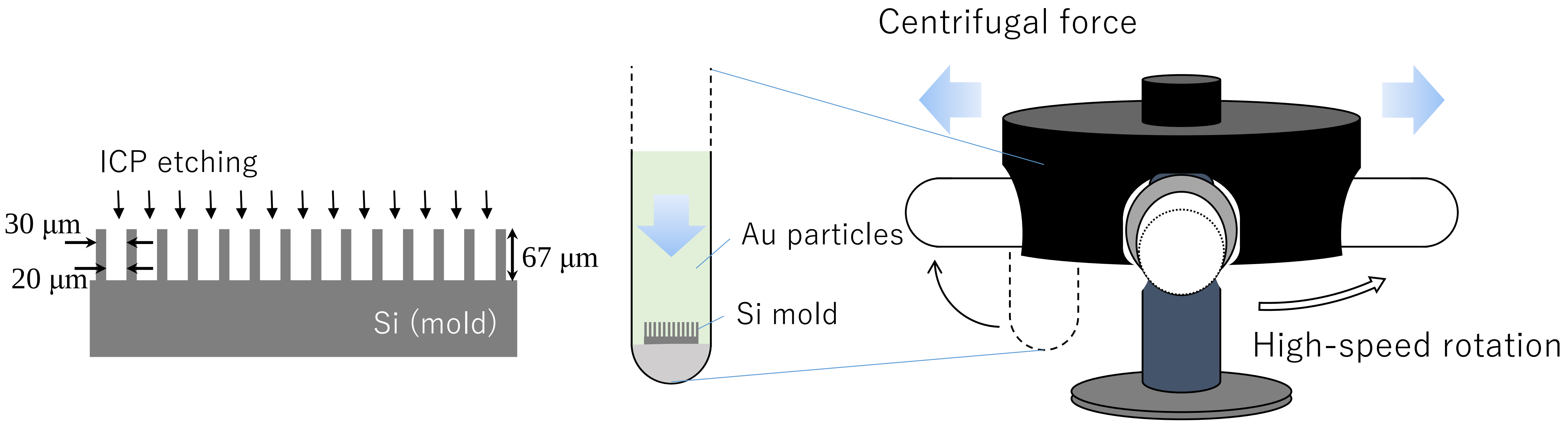
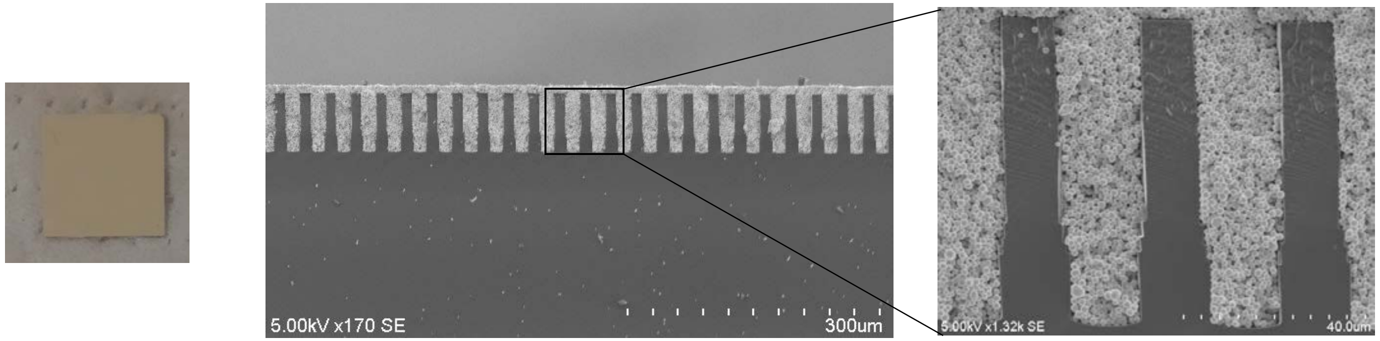


(a)

(b)

Fig. 1 Hojo et al.



(a) (b) (c)

Fig. 2 Hojo et al.

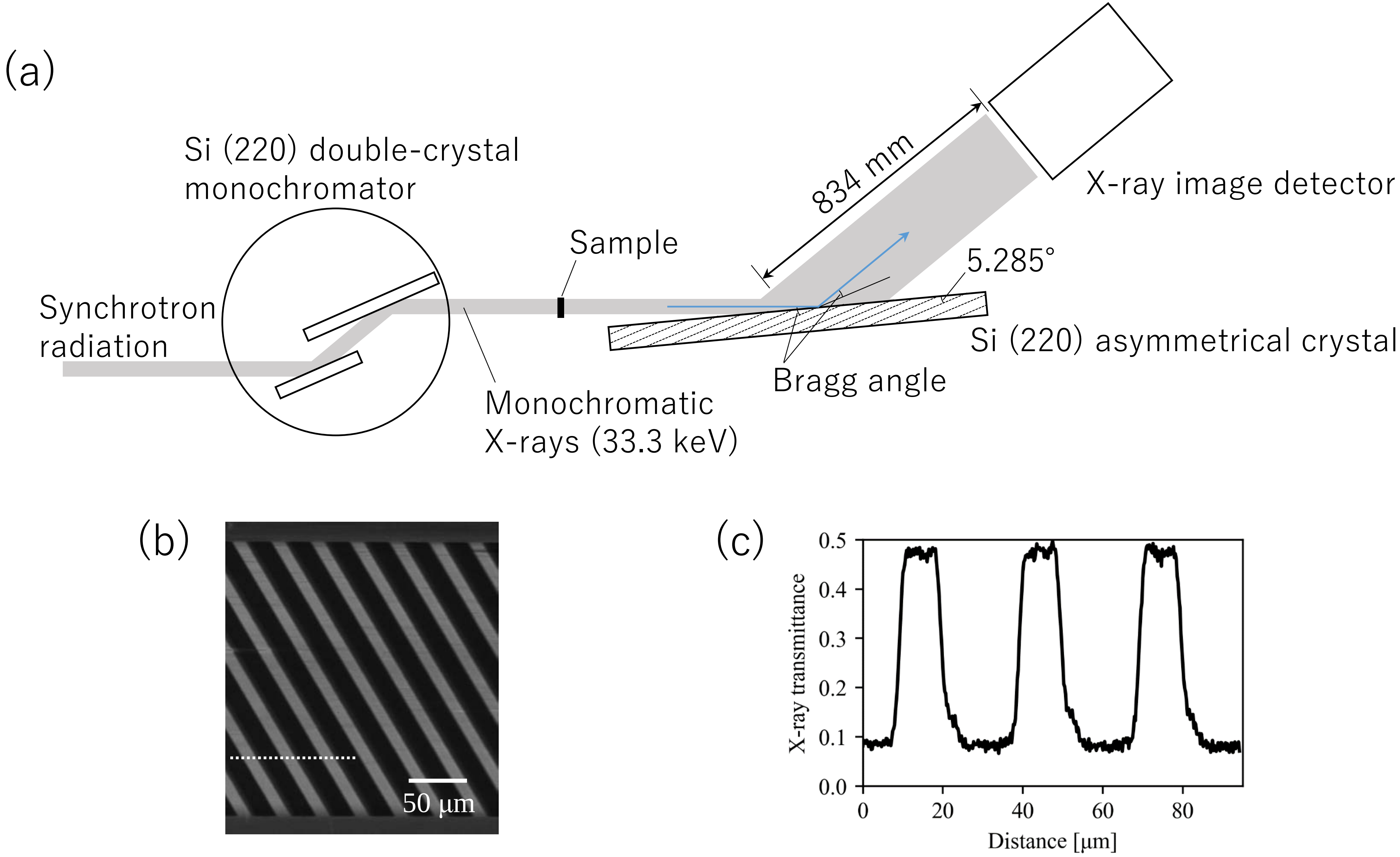


Fig. 3 Hojo et al.