Characterization of Synthetic IIa Diamonds for Third and Fourth Generation X-ray Sources

Diamond has been expected as a good candidate for optical elements in synchrotron radiation science because of their superior thermal and optical properties, i.e., a low linear thermal expansion coefficient, a high thermal conductivity, and a low X-ray absorption. However, the use of diamonds is limited than has been expected. Most high-heat-load monochromators are still using silicon crystals rather than diamonds, because the available quality and size of diamonds are believed to be insufficient.

The needs for perfect diamonds may be more serious for forthcoming self-amplified spontaneous emission free-electron-laser (SASE FEL) sources, such as SCSS at SPring-8 [1], which will deliver FEL pulses with a huge peak power of several gigawatts. Although the average power is easily manageable, the peak energy dose to the optical elements will be close to the melt limit for most materials, e.g., the melt limit of silicon is estimated to be 0.4 eV/atom [2] and the peak energy dose is calculated to be 0.1 eV/atom for 6 keV FEL beams at SCSS. Only low-Z materials, such as Li, Be, B, and C, can be used due to their smaller photoionization cross sections. Considering the preservation of the high spatial coherence of the FEL beams, perfect diamond crystals should be a unique solution.

We characterized (111) synthetic IIa diamonds at the 1-km beamline BL29XU [3]. These crystals were synthesized by Sumitomo Electric Industries Ltd. under high pressure and high temperature conditions [4]. The as-cleaved surface had a sufficiently large area (more than $7 \times 4 \text{ mm}^2$) for the monochromators.

Figure 1 shows the rocking curves of the 111 reflection in the reflection geometry of two samples, i.e., samples 1 and 2. Both the rocking curves measured on the entire surface showed narrow widths comparable to the theory, indicating a high crystalline perfection. The rocking curve of sample 2 almost agreed with the theoretical curve for a smaller beam size ($0.5 \times 0.5 \text{ mm}^2$). However, the width of the rocking curve depended slightly on the type of sample. When we compared quasi-plane wave topographs, sample 1 was found to include more defects, especially stacking faults, than sample 2 (Fig. 2). The width of the rocking curve seems to be related to the defect density.

To confirm such a relationship, we performed reciprocal space mapping. The 111 reciprocal space point of sample 1 did not spread uniformly, but was characterized by two strong streaks (Fig. 3(a)), which broadened the rocking curve. From a geometrical consideration, we found that the streak denoted as S1 is directed towards the projection of [1-11] and S2 is the projection of [11-1]. The findings that S1 was not observed and S2 showed an interference effect for sample 2 (Fig. 3(b)) indicate that the $<111>$ streaks originated from (111) stacking faults and not from the rough surface formed by cleavage.

We then investigated the crystal of high quality, sample 2, using the quasi-plane wave topograph (Fig. 2(b)). Two or three sets of stacking faults, many outcrops of dislocations (black spots), several surface scratches (black lines), and several dark streaks were observed. Such contrast was the origin of the broadening of the rocking curve. The dark streaks that extended from the lower part near the seed crystal were not isolated defects, such as dislocations, but were considered as strain bands. We detected small amounts of Fe and Co by scanning fluorescent X-ray analysis. These impurities included in the early stage of crystal growth were considered as the origin of the dark streaks.
In conclusion, synthetic type IIa diamonds of sufficient quality and dimensions for the high-heat-load monochromators at the third generation facilities are now available, when one selects crystals with a lower stacking fault density and uses a nearly defect-free area. Further reduction in defect density is desired for the future use of diamonds under fully spatial coherent radiation from SASE FEL.

Fig. 2. Reflection topographs with the 111 reflection of samples 1 (a) and 2 (b) measured at the lower tail of the rocking curve.

Fig. 3. Reciprocal space maps of samples 1 (a) and 2 (b) within the [111]-[01-1] plane around the 111 reciprocal space point.

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References