Polytype-Stabilized Solution Growth of 3C-SiC

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1. Introduction

Silicon carbide (SiC), which have wide band gaps, high thermal conductivities, and high electrical break-down field strengths, can be utilized for power devices. SiC exists in a large number of polytypes due to the different stacking sequences of Si-C double atomic planes. 3C-SiC, the target of the present study, is a promising polytype for MOSFETs with a higher channel mobility since the electron trapping by "near-interface-traps" in the $SiO_2/3C$ -SiC system can be much suppressed [1, 2]. Nevertheless, few device studies on 3C-SiC have been reported because no high-quality substrates exist. In our previous study [3], we grew a high quality needle-like 3C-SiC crystal by using a low-temperature solution growth technique. Then, to obtain a larger crystal, the top-seeded solution growth was performed on a (111) surface of the high quality needle-like 3C-SiC. However, the polytype of the grown crystal changed to 6H-SiC. The polytype transformation from 3C-SiC to 6H-SiC is considered to occur easily on the (111) surface due to the stacking errors since the (111) 3C-SiC is equivalent to the (0001) 6H-SiC. In this study, we grew stably large-scale 3C-SiC crystals by the following two approaches: (1) One is the homogeneous growth of 3C-SiC on (001) 3C-SiC thick epilayer. The concept of this approach is similar to the polytype stabilization in the step-controlled epitaxy on CVD method by utilizing the step-flow growth on off-oriented SiC substrates. (2) The other is a different approach that we intentionally induce the stacking error just at the surface of the 6H-SiC seed crystal and grow the crystal under the growth condition that 3C-SiC was thermodynamically stable. Consequently, the growth of a large-scale 3C-SiC crystal on the 6H-SiC substrate was achieved.

2. Experimental

A top-seeded dipping method was performed in a carbon crucible. For homogeneous growth, a seed crystal was a thick free-standing (001) 3C-SiC epilayer grown on an undulated Si substrate [4]. For heterogeneous growth, a seed was a (0001) 6H-SiC substrate. The seed crystal was mounted on the top of graphite rod and dipped to a Si solvent or Si-Sc solvent. The growth temperature was ranged from 1300 to 1700 °C.

3. Results and Discussion

Homogeneous growth of 3C-SiC on (001) 3C-SiC

Figure 1 shows the cross-sectional optical-microscope image of the SiC crystal grown on the free-standing (001)

3C-SiC epilayer at 1700 °C for 4 h. The surface of the grown layer partially consists of $\{111\}$ facet planes. The thickness of the grown layer ranged from 80 to 180 μ m corresponding to the growth rate of 22-49 μ m/h. The polytypes of both the seed crystal and the grown crystal were identified to be 3C-SiC from a transmission electron diffraction patterns. Thus, the use of (001) 3C-SiC seed made it possible to grow 3C-SiC crystals stably avoiding the polytype transformation.

However, some problems are still present. The stripes parallel to {111} planes are clearly observed in the optical microscopic image. The typical micro-Raman spectra obtained from the dark and light stripe regions are shown in Fig. 1 (b). In the micro-Raman spectrum at the dark region, the transverse optical (TO) mode of the 3C-SiC appears at 796 cm⁻¹. On the other hand, in the spectrum at the light region, the measured Raman spectrum is similar to the spectrum of 6H-SiC although the electron diffraction pattern indicates 3C-SiC. This discrepancy means that the high-density stacking faults with the density of $\sim 10^6$ cm⁻¹ exist in the crystal [5]. We also observed the high-density stacking faults with TEM. The (001) 3C-SiC epilayers used as seeds slightly include stacking faults of the density of 5×10^3 cm⁻¹ due to the lattice mismatch between SiC and Si [6]. These stacking faults drastically increased and extended to the grown crystals during the high temperature growth process.

Heterogeneous growth of 3C-SiC on (0001) 6H-SiC

For the "heterogeneous" growth, the following three points were considered: (1) A 4H- or 6H-SiC on-axis



Fig. 1 (a) Cross-sectional optical microscopic image near the interface between the grown and seed crystal of SiC, (b) typical micro-Raman spectra obtained from the light region and the dark region. 3C-SiC grew on the seed crystal homogeneously. However, high-density stacking faults existed in the light region.

(a) Cross-sectional image of 3C-SiC on 6H-SiC



Fig. 2. (a) Cross-sectional optical-microscope image of the crystal grown on the 6H-SiC (000-1) C-face for 50 hours at 1300 °C. The grown crystal on the 6H-SiC was identified as 3C-SiC. The green 6H-SiC crystal was grown from the edge of the seed. (b) Optical-microscope images of $18 \times 18 \text{ mm}^2$ 3C-SiC on 6H-SiC seed crystals. In the center black area, the carbon rods mounting seed crystals are seen through the transparent wafers.

(0001) substrate was used as a seed to avoid the continuity of the polytype of the seed crystal; (2) low-temperature growth was performed by using a Si-Sc solvent of the eutectic composition; (3) growth was carried out on the (000-1) C-face. Figure 2 shows the optical-microscope image of 3C-SiC with the area of 18×18 mm² grown on 6H-SiC (000-1) C-face for 50 hours at 1300 °C. The grown crystal on the 6H-SiC was transparent yellow, and this crystal was identified as 3C-SiC with Raman spectrum measurement. The green 6H-SiC crystal grew from the edge of the seed. This indicates the use of (000-1) was important to induce intentionally the stacking error. Under the various growth conditions without the successful example of Fig. 2, 3C-SiC did not stably grow or the other polytype grew. In other words, it is possible to grow the 3C-SiC and the 6H-SiC selectively on 6H-SiC substrate by optimizing the growth condition. This developed technique can grow large-area 3C-SiC crystals using commercial 2 or 3 inch 6H-SiC substrates made by the sublimation method. Moreover, the crystal quality is expected to be better since the stacking fault density in 6H-SiC substrate is very low.

4. Conclusions

We grew the large-area 3C-SiC crystals by the two

approaches. In the case of growth on (001) 3C-SiC epilayer, the 3C-SiC could grow stably. However, the high density stacking faults were induced in the grown crystal at 1700 °C by the extension of the stacking faults originally included in the seed crystal. In the growth on (000-1) 6H-SiC substrate, the $18 \times 18 \text{ mm}^2$ 3C-SiC could grow heterogeneously under the specific growth condition. We consider that the former technique is better since it is easy to grow large-area 3C-SiC crystals using commercial 2 or 3 inch 6H-SiC substrates without stacking fault. The surface morphology of the crystal on (000-1) 6H-SiC is smoother than that on (001) 3C-SiC. In addition, it is possible to grow the various polytype selectively by optimizing the growth condition.

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