Abstract

The molecular beam epitaxy process can produce single crystal and smooth surface at atomic level as well as synthesizing the desired material by supplying the multiple materials on substrates. This paper deals with an application of the hetero-epitaxial process of silicon carbide (SiC) on silicon (Si) substrate, and aims to make clear the attainable surface roughness and its properties. It was found that the steep pits were formed during the carbonization process before the epitaxy and that they strongly affected the final roughness. The attainable roughness was 0.4nm rms. Finally, the applicability to toroidal mirror optics was discussed.

Keywords: Silicon carbide, Mirror surface, Deposition process

1 INTRODUCTION

Silicon carbide (SiC) has good mechanical properties such as thermal and cryogenic stability and is expected to be applied as the structural material for optics in severe environment [1]. However, the production of SiC and its surface finish are difficult because of the high melting point and hardness.

Chemical vapor deposition (CVD) process has been well used to produce SiC layers on the sintered SiC substrate. By polishing SiC layer, the surface became smooth up to 0.41nm rms [2]. The recent grinding process can finish the surface up to 0.266nm rms [3]. As to the hard X-rays optics, it has been found that not only the roughness with nm wavelength but also the waviness with several mm wavelength affects the focusing properties [4]. As a result, the smoothing process is required that can attenuate the profile error over wide spatial frequency range.

The processing properties are also affected by the material's structure, that is, polycrystalline, single crystal or amorphous. Theoretically, single crystal material is considered as the best among them if the anisotropy does not disturb the functionality. The small size SiC wafers are already commercially available. In order to produce the large size wafers, the SiC/Si hetero-structure is often used because large size single crystal silicon is easily obtained for the substrate. The low pressure CVD method (LPCVD) has been established for four-inch size wafers [5].

Epitaxy is one of the crystal growth processes and can be used as the smoothing process at the atomic level. For example, the surface roughness of 0.2nm Ra was obtained in the silicon homo-epitaxy [6, 7]. In the hetero-epitaxy process, the difference between the lattice constant of grown material and that of the substrate often causes strain and there appear islands of single crystal area [8]. In addition, there appear the so-called "anti-phase boundaries" between them, where the lattice phase differs as shown in Figure 1. As a result, it is difficult to obtain perfect surface with single crystal and consistent structure. However, the crystal orientation of the islands is consistent due to the principle of epitaxy, and the theoretical height of the step at a boundary is only 0.066nm. Thus, this process can be applied to the surface finish of optical elements.

In this paper, the surface finish properties of epitaxial growth are investigated, and the attainable roughness is to be made clear. Then, its applicability to X-ray mirror element is discussed.

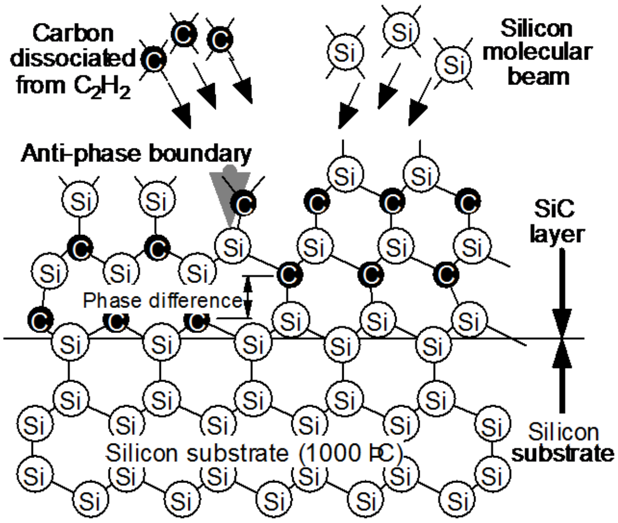


Figure 1: Hetero-epitaxial growth model of SiC on Si substrate and anti-phase boundary.

2 EXPERIMENTAL SETUP AND PROCEDURE

Figure 2 shows the apparatus for experiments. A four-inch silicon wafer is held upside down in a vacuum chamber and heated from its backside. The molecular beams of source materials are supplied from the bottom in the chamber, where the silicon is supplied by melting the silicon ingot using an electron beam evaporator while the carbon is supplied by dissociating the acetylene gas. The crystal structure of the substrate can be examined by the reflection high-energy electron diffraction (RHEED) patterns. The main specifications are also shown in tabular form.

Figure 3 shows a typical process procedure. The process is divided into two stages, namely, the carbonization and the epitaxial growth process. In the carbonization stage, only the acetylene gas is emitted into the chamber. The acetylene molecules approached to the substrate are dissociated by the heat radiated from the wafer, and the carbon molecular beams reach to the substrate. This process is necessary for inhibiting the migration and dissociation of the silicon atoms from the surface. After the carbonization, the silicon molecular beams are supplied to synthesize SiC layer together with the acetylene gas.

Table 1 shows the experimental conditions. The original substrate was (111) silicon wafer of 450μm thickness with the surface roughness of 0.3nm rms. The partial pressure of the acetylene gas during the carbonization was set at 10^{-4} or 10^{-2}Pa, by adjusting both the flow rate of acetylene gas and that of liquid nitrogen fed into the shroud (see Figure 2). The heating and cooling rates before and after the epitaxial growth are important in avoiding the distortion of the substrate and set between 2-10˚C/min empirically. The growth thickness of SiC was 50nm in all experiments.

3 EVALUATION OF THE RESULTS

3.1 Crystal structure

Figure 4 shows the results of the RHEED observations. From these observations, both the regularity of crystal structure and its lattice constant of several atomic layers thickness over 100μmx2cm area can be examined. The pattern becomes spotty in case the structure has regularity as single crystal. The spacing of the streaks, the vertical stripes, is inversely proportional to the lattice constant.

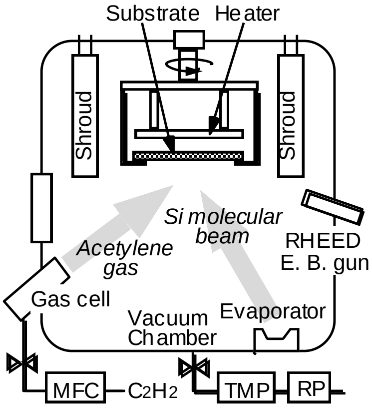
Figure 4(a) shows the patterns of the original silicon substrate. The substrate has regular crystal structure. Figure 4(b) shows the pattern after the carbonization at 10^{-2}Pa. The spots are not clear, thus the regularity of crystal structure deteriorated. The spacing of the streaks becomes wider than that of the original substrate, which means the lattice constant has become smaller. The ratio of the spacings is about 1.2 as shown in the Figure 4, which coincides with the theoretical ratio 1.245.

Figure 4(c) shows the result after the epitaxial growth. The pattern has become spotty and the spacing of the streak pattern becomes clear so that it is concluded that the regularity of crystal structure was improved and the single crystal silicon carbide layer has been produced.

3.2 Carbonization process

Figure 5 shows the top views of the carbonized surface observed by an atomic force microscope (AFM), and compares the effect of the acetylene pressure in case of low and high during the carbonization. The examined area was 30μm by 30μm, and a portion is shown in the figures. The deep and steep pits appeared regularly. In case the acetylene supplying pressure is higher, the density of the pits becomes lower and the size becomes smaller.

The shape of the pit is triangular and though the size differs, its orientation is almost same. Considering these regularities, the finished surface is considered as one of the textured surfaces [9]. Such surface characteristic may affect the optical properties such as reflectivity.



Specifications

Pressure	$>10^{-9}$ Pa
Substrate size rotation temp.	 < ϕ100mm < 13rpm < 1200 ˚C
Evaporator voltage current	 -4 – -10kV < 500mA
C_2H_2 supply rate	< 50sccm (cm³/min)
Trap of contaminant	Liquid N_2 shroud

Figure 2: Apparatus for SiC epitaxy.

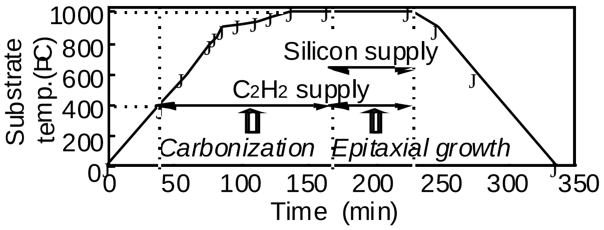
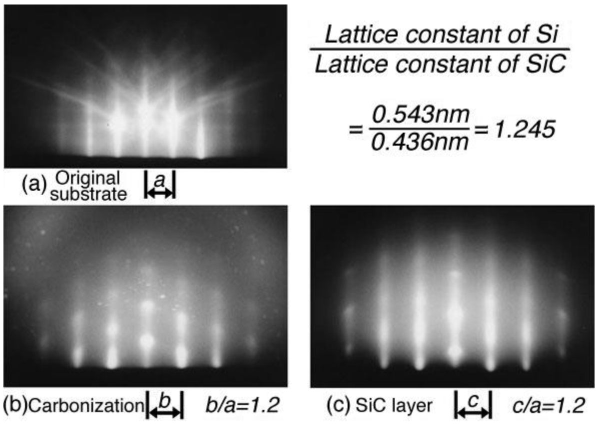


Figure 3: Typical process procedure.

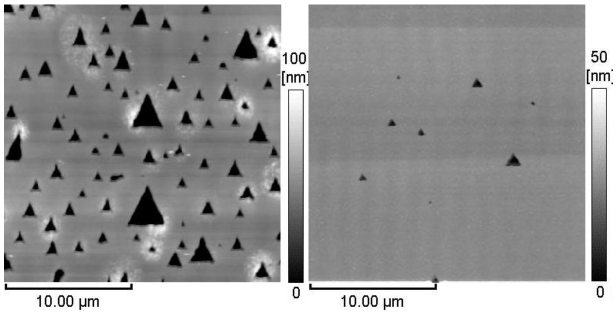
Substrate	Single crystal silicon (111), 100mm diameter	
Carboni-Zation	Temp. at carbonization starts	400˚C
	Partial pressure of C_2H_2	10^{-4}, 10^{-2}Pa
Epitaxial growth process	Substrate temp.	1000˚C
	Nominal supply rate of Si	0.02nm/s
	Partial pressure of C_2H_2	10^{-3}Pa
	Growth time	60min

Table 1: Experimental conditions.



$$\frac{\text{Lattice constant of Si}}{\text{Lattice constant of SiC}} = \frac{0.543nm}{0.436nm} = 1.245$$

Figure 4: RHEED observations.



(a) C_2H_2:10^{-4}Pa(19.1nm rms) (b) C_2H_2:10^{-2}Pa (5.1nm rms)

Figure 5: AFM observation of carbonized surface.

The crystal facet of the inner surface of the pit is identified by its orientation as {111} crystal plane. This is reasonable because the surface energy of this facet is the minimum among the others. From the gray-scale in the figure, it can be seen that the pits become deeper in case the acetylene pressure is low (Figure 5(a)). The maximum depth of the pits is about 100nm, which is much deeper than the expected carbonized layer thickness of 1nm. The reason will be discussed later.

3.3 Epitaxial growth process

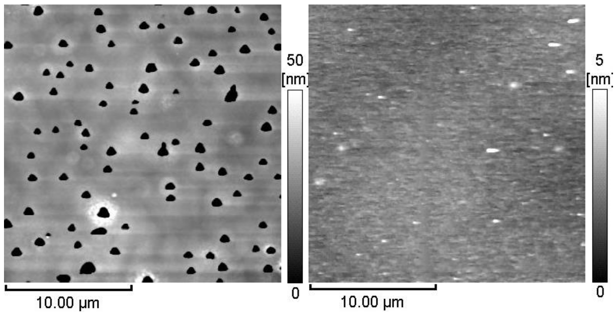
Figure 6 shows the top views of the epitaxially grown SiC surface observed by the AFM, and compares the effect of the partial pressure in case of low and high during the carbonization. In case the acetylene gas pressure is high and the density of the pits is low, the surface became smooth, 1.1nm rms, as shown in Figure 6(b). Whereas, in case the acetylene pressure is low, the surface did not become smooth due to the existence of the pits as shown in Figure 6(a). The pits look smaller and more rounded than those on the carbonized surface shown in Figure 5(a). From these results, it is found that the prevention of the pits formation is essential in obtaining smooth surfaces.

Figure 7 shows both the top view of the wide area observation and the cross sectional profile. The sample was the same as in Figure 6(a) but the examined area was 100μm by 100μm and only a portion of the area is shown. Larger pits than those of Figure 6(a) can be seen and their edges are higher than the height of the other area like a protrusion. The cross sectional profile measured by the AFM shows it clearly. The depth of the pits reaches 300nm, whereas the depth after the carbonization was about 100nm by another measurement. The protrusion at the pit edge was about 60nm, whereas it was about 30nm after the carbonization. Thus, the surface became rougher along with the process, and the pits formed in the carbonization process did not disappear but grew. It was estimated that the silicon atoms migrate from the bulk to the surface through the pits, and react with the carbon atoms to produce SiC molecules if the carbon supply is insufficient.

Figure 8 shows the cross sectional profile of the smoothest surface observed by the AFM. Surface roughness is calculated at 0.4nm rms along 10μm length. The short wavelength components can be seen in the profile. Local and microscopic crystal growth might have caused this fluctuation, though some of them have come from the noise in the measurement. This smooth surface is considered to be applicable to mirror optics.

3.4 Surface Finishing Properties

Figure 9 shows the relation between the pit size and the frequency that were analyzed by a general-purpose image-processing program. The upper graph shows the result of carbonization stage analyzed from the top view of Figure 5, and the lower graph shows that of the epitaxy stage analyzed from top view of Figure 6. The sampled area was 30 by 30 μm. The vertical axis denotes the number of the pits in specific size ranges. In case the acetylene pressure is low, the number of smaller pits increase. However, the larger pits become dominant after the epitaxy process. On the other hand, if the pressure is high, the whole number of the pits decreases, while small pits are observed on the epitaxially grown surface. It was found that regardless of the pit size, the dominant pits were left on the surface.



(a) Epi. after 10^{-4}Pa carbo. (28nm rms) (b) Epi. after 10^{-2}Pa carbo. (1.1nm rms)

Figure 6: AFM observation of SiC grown surface.

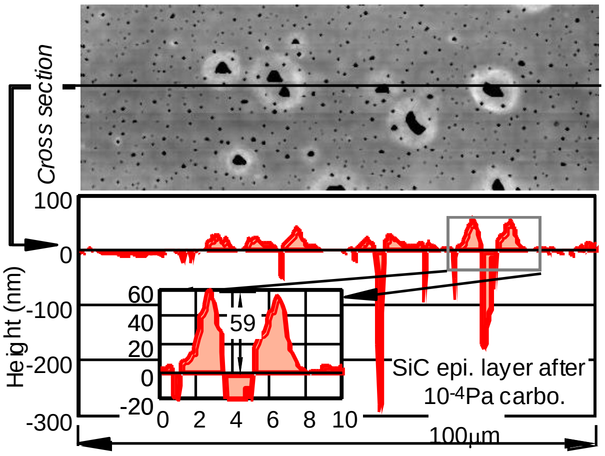


Figure 7: Cross section of the pits on SiC surface.

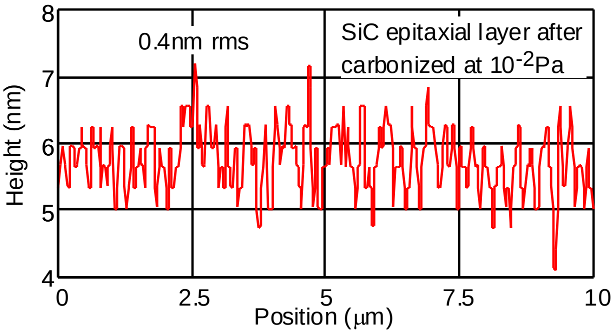


Figure 8: Cross section of the smooth SiC surface.

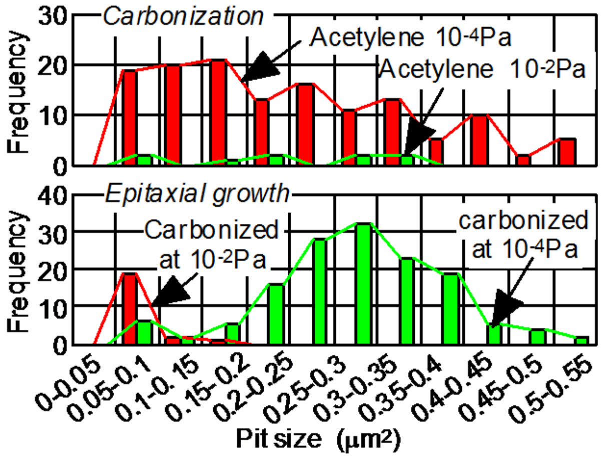


Figure 9: Relation between pit size and frequency.

Figure 10 shows the results of the Fourier analysis of the cross sectional profile obtained by the AFM observations. The horizontal axis denotes the spatial frequency, the inverse of the wavelength. The vertical axis denotes the power spectral density. The magnitude has no quantitative meaning but represents the relative amplitude at specific frequency.

The results show that in case the acetylene pressure during the carbonization is high, the epitaxial process smoothes the surface by an order of magnitude over almost all frequency ranges. On the other hand, if the acetylene pressure is low, the surface does not become smooth but become rough. In both cases, the epitaxially grown surfaces have a low frequency component, in other words, long wavelength component. This means that the geometrical accuracy with several tens μm wavelength cannot be improved. The epitaxial process can be applied to the surface smoothing process only in short wavelength.

4 APPLICABILITY TO OPTICAL ELEMENTS

Epitaxially grown surface is considered to be applicable for finishing plane mirrors because the specific crystal plane appears autonomously, that is, the flat and smooth surface at atomic level can be obtained.

However, it is difficult to finish the three-dimensional profile by the epitaxy because this process is much affected by the crystal orientation of the substrate [7]. Figure 11 shows a typical design of toroidal mirror for X-ray optics. From the specifications, the Miller indices at the edges of the mirror surface can be calculated. In this case, the crystal plane is almost same as the original substrate (111), because the geometrical deviation from the plane is small. Thus it is considered the epitaxial process can smooth the roughness with short wavelength all over the surface area. However, it is necessary to form the aspheric profile using other processes in advance. The machinability of the silicon is superior to that of SiC, so this combined process is considered as feasible. Experimental verification has not been completed. This is one of the future subjects.

5 CONCLUSIONS

The hetero-epitaxial growth of silicon carbide on silicon substrate was introduced for the use of surface smoothing process, and its properties were examined. The results are summarized as follows:

1. The pits that were formed during the carbonization process govern the final roughness.
2. Sufficient carbon supply during the carbonization process is crucial in suppressing the formation of pits.
3. The best result of the surface roughness was 0.4nm rms and considered as applicable to mirror optics, though the profile error with long wavelength is not necessarily improved.

6 ACKNOWLEDGEMENT

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7 REFERENCES

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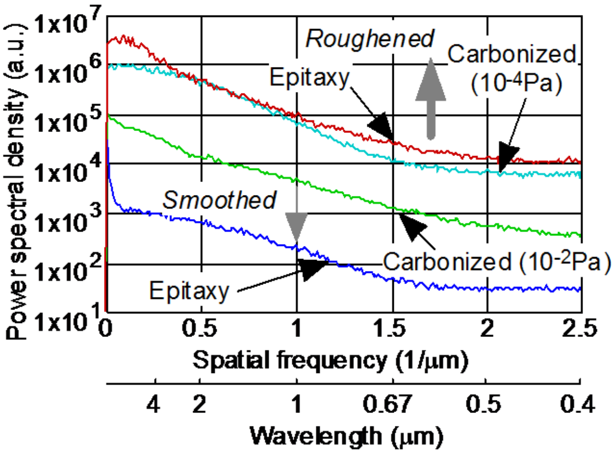

Figure 10: Power spectrum of the cross sectional profile.

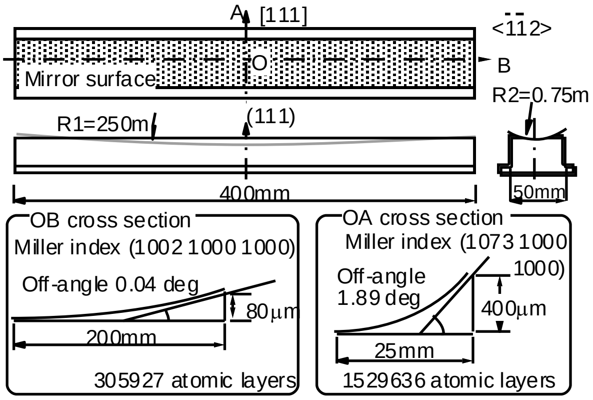

Figure 11: Typical specifications of a toroidal mirror.

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