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Epitaxial Silicon Carbide Films Grown by New Method of Replacement of Atoms on the Surface of High-resistivity (111) Oriented Silicon

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Abstract. The nanolayers of single crystal SiC were grown on the surface of a high-resistance n-type silicon substrates by replacement of the atoms in the crystal lattice of silicon on the carbon atoms at the temperatures of 1250, 1330 °C and CO gas pressures 264, 395 Pa, respectively. The formation of crystalline β-SiC phase in films by electron diffraction and Raman spectroscopy techniques was shown. The SiC films are epitaxial and do not contain twins on the surface. By Atomic Force Microscopy is shown that two set of SiC films have pyramidal and step-like structure of the surface with clear-cut fragmentation of grains with sizes between 100 and 200 nm, and this is due to the composition of carbon and silicon atoms in the layer. Two set of SiC films have a granular surface structure with indistinct grain fragmentation. The influence of synthesis condition on the microstructure of film surface is discussed.

1 Introduction

In a series of papers, generalized in review [1], it has been developed theoretically and experimentally implemented a new method of growth of high quality thin SiC films on Si. The method consists of replacing part of the atoms of the matrix of silicon on the carbon atoms to form molecules of silicon carbide. The process of replacement occurs gradually without destroying the crystal structure of silicon. Moreover, the "old" original crystalline structure of the silicon matrix defines the orientation of the film, while using conventional techniques only the surface of the silicon substrate defines the film orientation. According [1] the process of silicon atoms replacement on the carbon atoms can be realized by using such chemical reaction as $2Si + CO = SiC + SiO$.

It is known [2] that SiC has more than 170 different polytypes. The most common forms are cubic (3C-SiC), as well as hexagonal 4H, 6H (α-SiC) [2-5]. Raman spectroscopy is an efficient and non-destructive technique which may be used to identify the structure of the SiC polytype. In the case of 3C-SiC typically two strong Raman peaks are observed: at 972 cm^{-1}, corresponding to the peak of longitudinal optical (LO) phonons; at 796 cm^{-1}, corresponding to the peak of transverse optical (TO) phonons [5]. Hexagonal modes for 6H-SiC are indicated by splitting the TO mode into three peaks: TO_2 at 767 cm^{-1} and 789 cm^{-1}, TO_1 at 797 cm^{-1}, LO_1 at 965 cm^{-1} [5].

In this work, using Raman spectroscopy, electron microscopy and Atomic Force Microscopy the nanolayers of single crystal SiC grown by this method, have been investigated.

2 Experimental

The nanolayers of single crystal SiC were grown on the surface of a high-resistance n-type silicon substrates. The thickness of the original (111) oriented silicon substrates was about 1300 microns. Before synthesis, the surface of silicon wafers were ground, polished and specially treated to obtain a smooth mirror surface. SiC films were synthesized in a special installation, developed by the authors [6]. The synthesis was carried out under the following conditions. Two series of samples, marked № 1 and № 2, were grown at the temperature of 1250 °C and at pressure of 264 Pa in the atmosphere of CO. The growth time of these samples was 15 min. Two other series of films, marked № 3 and № 4, were synthesized for 7 minutes at the temperature of 1330 °C and at pressure of 395 Pa in the atmosphere of CO.

The composition of the films was investigated by Raman spectroscopy using a confocal Raman microscope Alpha 300R (WITec, Germany). The microstructure of the surface of the films was studied using the atomic force microscope JSM 5200 (Jeol, Japan) by semi-contact (AFM AC) method. Resolution of the microscope on a plane is 0.14 nm and vertical resolution - 0.01 nm.

3 Results

Figure 1 presents the Raman spectra of the crystalline silicon carbide films on silicon substrates (samples № 1 (a), № 2 (b), № 3 (c) and № 4 (d)), synthesized by replacement of the atoms in the crystal lattice of silicon.

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The spectra show the presence of first and second order transverse acoustical phonon (2TA) peaks from the crystalline Si substrate [7] at 519.7 and 303.3 cm^{-1}, respectively. A clear peak due to the Si second order features [7] appearing at 971.4 cm^{-1} is also presented.

Figure 1a - d shows that in the Raman spectra of the SiC films № 1 and № 2, synthesized at a temperature of 1250 °C and a pressure of 264 Pa, and the SiC films № 3 и № 4, synthesized at a temperature of 1330 °C and a pressure of 395 Pa, there are broad peaks at 730 - 860 cm^{-1} centered in the TO mode of β-SiC at ~ 796 cm^{-1} [8] consisting of two components with maxima at 792.6 ± 0.3 cm^{-1} and 822.7 ± 1.1 cm^{-1} (Table 1). It is evident that the greatest value of the amplitude of the peaks characteristic of the films of SiC № 2 and № 3.

The LO mode of β-SiC at 969 cm^{-1} is impossible to identify due to the superimposed Si-second-order Raman background.

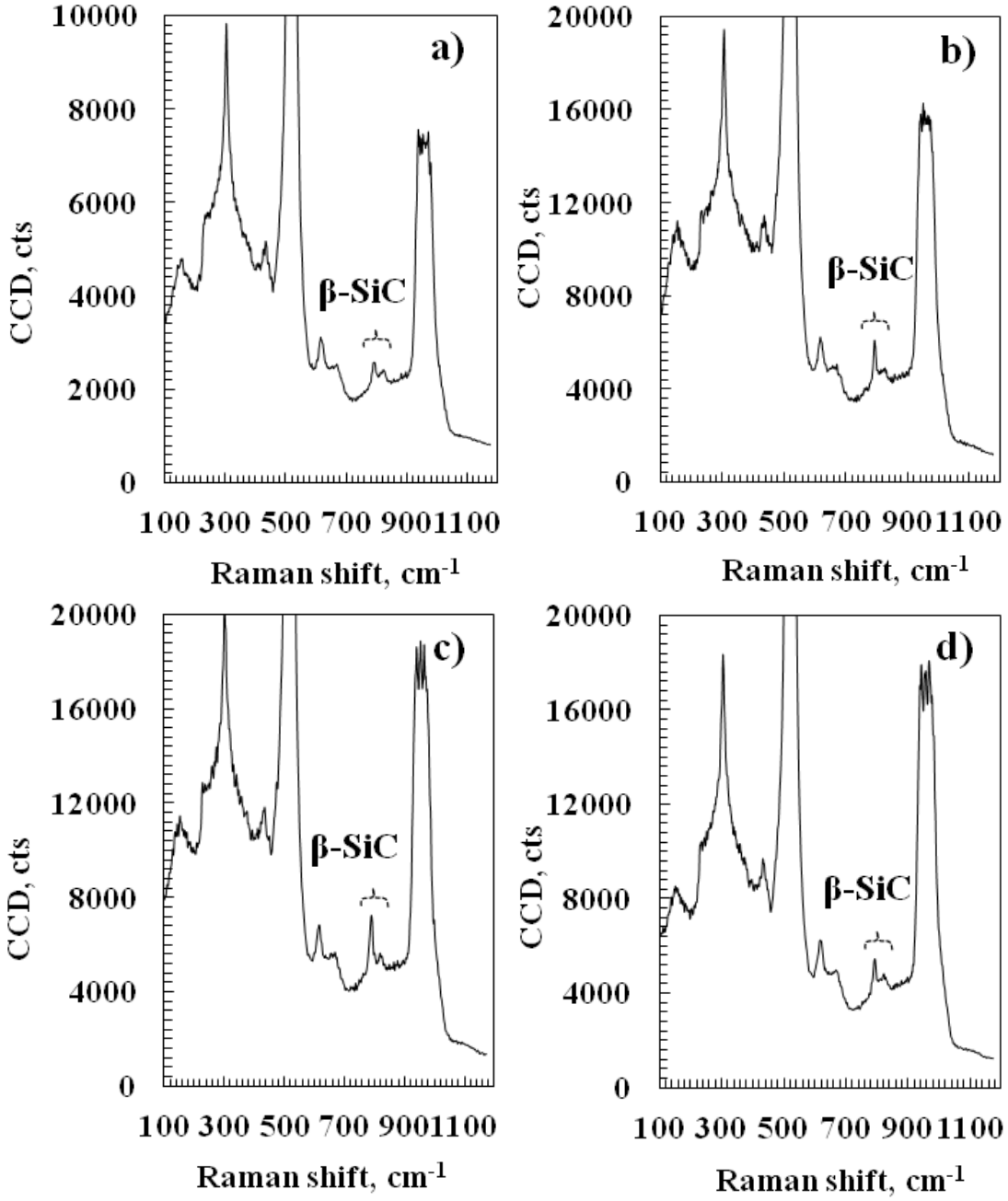

Figure 1. Raman spectra of crystalline SiC films on silicon substrates (samples № 1 (a), № 2 (b), № 3 (c) and № 4 (d)), synthesized by replacement of atoms in the crystal lattice of silicon

Table 1. The position of the maxima of two components of β-SiC TO mode in the Raman spectrum

Option	SiC film № 1		SiC film № 2		SiC film № 3		SiC film № 4	
Position of maxima, cm^{-1}	791.8	822.7	791.8	826.1	792.4	821.6	792.9	822.7

Studies by Atomic Force Microscopy of the surface microstructure of the film № 1 in areas with dimensions of 500 × 500 nm (Figure 2a) and 200 × 200 nm (Figure 2b) show that the film surface after synthesis has a pyramidal structure and is composed of large crystals with sizes ~ 100 × 100 nm, has clear-cut fragmentation of grains and height variations within 19 nm. Protruding portions of the surface have bright color, low surface areas, from which height countdown begins, have a dark color.

The surface of the film № 2 (Figure 2c, d) at similar areas has a step-like structure, variation of height up to 38 nm and more clear-cut fragmentation of large crystals with sizes of about 200 × 150 nm. SiC films № 3 (Figure 2e, f) and № 4 (Figure 2g, h) have a granular surface structure with variations of height up to 46 nm and 19 nm, respectively, with indistinct grain fragmentation.

In general, it is seen that the films № 1 and № 2 have a similar structure with a distinct fragmentation of the grains on the surface and different from films № 3 and № 4 having insufficient grain fragmentation. This may be due to excessive content of carbon atoms [9, 10] in the films № 3 and 4 due to the higher synthesis temperature and a high pressure of CO gas.

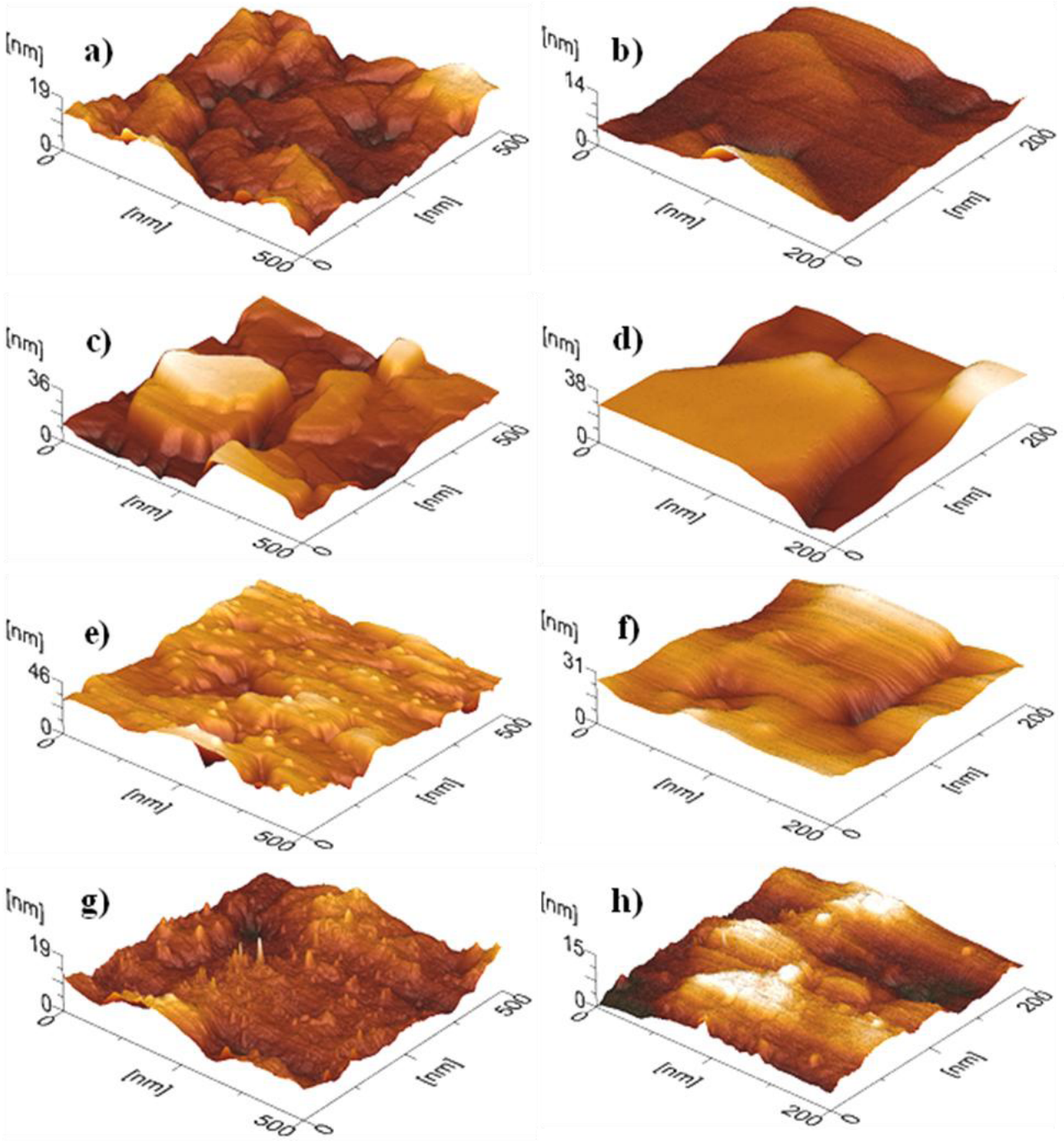

Figure 2. Atomic Force Microscopy of the surface microstructure of the SiC films № 1(a,b), № 2(c,d), № 3(e,f), № 4(g,h), in areas with dimensions of 500 × 500 nm (a,c,e,g) and 200 × 200 nm (b,d,f,h)

Figure 3 shows the electron diffraction patterns of the SiC films of samples № 1, № 2, № 3 and № 4, measured in the <110> zone axis projection.

Point reflexes of electron diffraction patterns clearly indicate that the (111) oriented 3C-SiC layer lies on the surfaces of Si samples № 1, № 2, № 3 and № 4, and the direction [111] of this layer perpendicular to the substrate surface. It is also evident from electron diffraction patterns that the SiC films are epitaxial and do not contain twins on the surface.

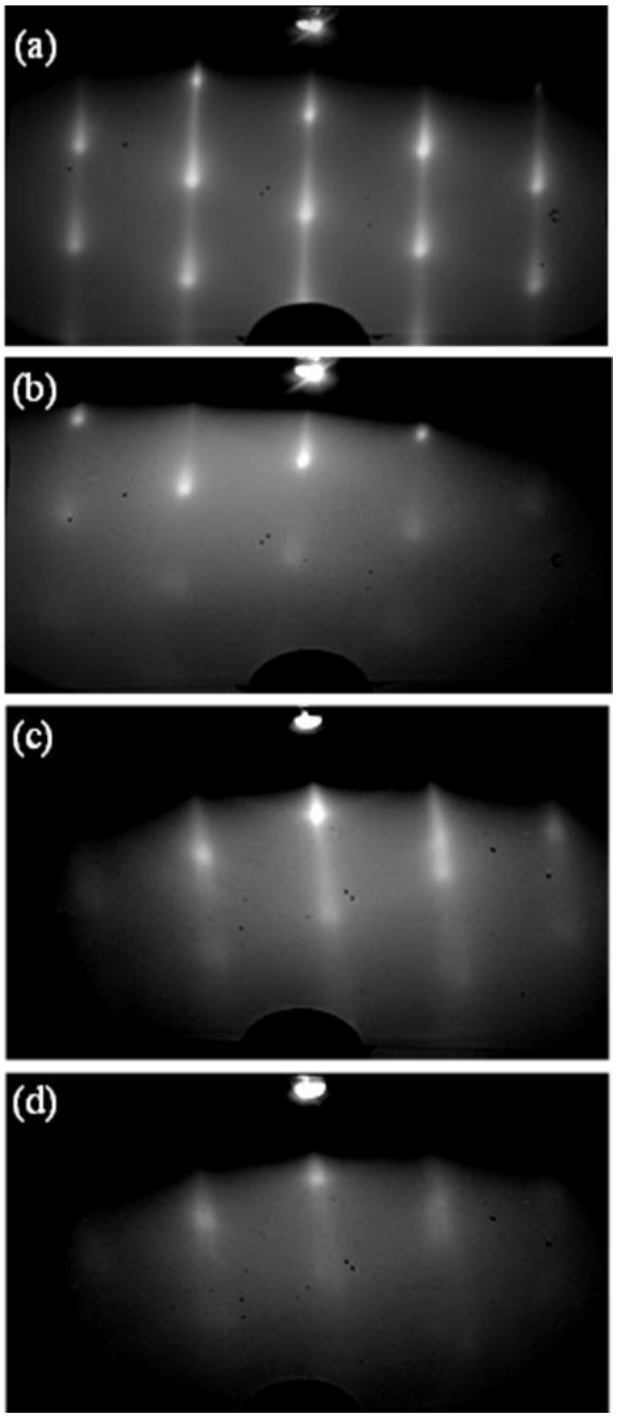

Figure 3. Reflection electron diffraction patterns of the SiC films recorded in the <110> zone axis projection for samples № 1 (a), № 2 (b), № 3 (c), № 4 (d)

4 Conclusion

The nanolayers of single crystal SiC were grown on the surface of a high-resistance n-type silicon substrates by replacement of the atoms in the crystal lattice of silicon on the carbon atoms.

The formation of crystalline β-SiC phase in films synthesized at the temperature of 1250 °C and the pressure of CO gas 264 Pa or 1330 °C, 395 Pa, were shown. The (111) oriented 3C-SiC films on the (111) oriented silicon substrates are epitaxial and do not contain twins on the surface.

It is shown that the films № 1 and № 2 have a similar pyramidal or step-like structure of the surface with variations of height up to 19 and 38 nm, respectively, and with clear-cut fragmentation of grains with sizes between 100 and 200 nm, and this is due to the composition of carbon and silicon atoms in the layer. SiC films № 3 and № 4 have a granular surface structure with indistinct grain fragmentation and this is due to high content of carbon atoms in the films due to the higher synthesis temperature and a high pressure of CO gas.

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