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## 主 論 文 の 要 旨

論文題目 Growth of Epitaxial Graphene on 6H-SiC and Its Characterization by Surface Analysis Methods ( 6H-SiC 上のエピタキシャルグラフェンの成長と表面分析法によるその評価)

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## 論 文 内 容 の 要 旨

Graphene is an allotrope of carbon which is arranged into a honeycomb lattice with one carbon atom thick. Graphene was first produced in 2004 by Andre Geim, Konstantin Novoselov and their collaborators from the University of Manchester, UK and the Institute for Microelectronics Technology in Chernogolovka, Russia [1]. In 2010, The Nobel Prize in physics was awarded to Andre Geim and Konstantin Novoselov. Graphene has many exotic properties such as its high strength and flexibility. Namely, graphene is about 100 times stronger than steel and it can be stretch by 20 percent. And yet, graphene is flexible like rubber [2]. In addition, graphene also has many exceptional electronic properties such as high mobility of carriers and linear dispersion (Dirac cone) at the K-point in the Brillouin zone where the valence and conduction bands touch with each other. However, graphene electronic properties vary with the graphene thickness. For example, the monolayer graphene has no band gap but the bilayer graphene has the band gap with parabolic band dispersion due to the interaction of adjacent layers.

Graphene can be formed by many ways such as mechanical exfoliation of graphite, chemical vapor deposition (CVD) of carbon-bearing gases on the surface of copper films [3, 4], and cutting open nanotubes [5]. The growth of graphene by annealing silicon carbide (SiC) substrate is also one of the efficient approaches for a large scale production of graphene. Graphene on SiC can be applied for many applications such as field-effect transistors (FETs) [6], optical antennas [7] and so on.

Silicon carbide (SiC) is a semiconductor with attractive properties such as wide band gap, high saturated electron mobility, high break-down electric field, good chemical and thermal stability and so on [8]. Since SiC is a polar crystal, SiC has 2 types of surface i.e. Si-terminated and C-terminated faces. SiC substrate that ends up with the Si layer is called Si-face SiC. Similarly, SiC substrate that ends up with the C layer is called C-face SiC. Both faces form graphene, but graphene grown on the two polar surfaces is different in

quality and thickness.

In this thesis, we study on the growth of epitaxial graphene on Si-face and C-face 6H-SiC (0001). Thickness (or layer number) of the epitaxial graphene film on SiC is estimated by X-ray diffraction technique. In addition, we also study the growth of embedded and protrusive striped graphene and ring-like shaped graphene on 6H-SiC. The followings show outline of the study in this thesis.

In chapter 2, the basic knowledge of SiC and graphene structures, analytical instruments and X-ray diffraction is written by using simple words to understand the instrument and principle clearly.

In chapter 3, the experimental procedure is thoroughly written including synthesis and characterization of graphene.

It is important to know the layer number of graphene film since the electronic properties of graphene depend on graphene film thickness. In chapter 4, the efficient method to determine graphene film thickness on 6H-SiC by using X-ray diffraction is demonstrated. We offer a simple and effective master equation to estimate graphene film thickness on 6H-SiC. The accuracy of the results is confirmed by ultra-high vacuum scanning electron microscope (UHV-SEM) and angle-resolved photoemission spectroscopy (ARPES). The contents in this chapter were published briefly in *Applied Surface Science*, Vol. 282, pp. 297-301 (2013).

In chapter 5, we anneal C-face 6H-SiC (000-1) substrates under argon gas to produce epitaxial graphene films with average thickness of 3 monolayers on the substrate. We also characterize the surface structure and morphology by reflection high-energy electron diffraction (RHEED) and atomic force microscopy (AFM). Moreover, Raman mapping measurement reveals that the graphene layer has high uniformity in doping concentration and strains. After graphitization we found that the step height of SiC surface is lower than 9 nm and graphene grows continuously across these steps to form large domains. AFM phase images indicate that the SiC surface is completely covered by graphene. The statement in the chapter was published in *Surface and Interface Analysis*, Vol. 44, pp. 793-796 (2012).

In chapter 6, we find the growth of protrusive striped graphene on Si-face 6H-SiC (0001) by annealing the SiC substrate under Ar gas. This result shows a strong contrast against the epitaxial graphene on SiC substrate grown in UHV; the epitaxial graphene is embedded in SiC surface with bonds between graphene edge and Si or C atoms of SiC substrate. For the graphene grown in Ar, the graphene edge structures approach free standing graphene because there is no bonding between graphene edge and SiC substrate. The context was briefly published in *International Journal of Engineering and Innovative Technology* Vol. 3, Issue 7, pp. 34-38 (2014).

In chapter 7, we display the growth of graphene rings on Si-face 6H-SiC (0001) since graphene rings have many attractive properties such as high electromagnetic confinement. This property implies that graphene rings are suitable for applying to optical antennas. Moreover, this growth method can preserve the structure of ring edges intact unlike electron-beam lithographic methods. The contents in this chapter were published in *Applied Surface*

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