# Influence of Annealing Atmosphere on the Epitaxial Graphene Growth on 3C-SiC (111)/Si (111)

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### 1. Introduction

Due to its extremely high electron mobility and field generated band gap, graphene based electronics have potential in high speed and low power device applications [1]. However, its viable application on electronic devices depends on a large scale availability of good quality homogenous graphene. Graphene synthesis by preferential silicon (Si) atom sublimation from silicon carbide (SiC) is one of the popular techniques to get epitaxial graphene. Hibino et al. [2] had reported few-layer graphene by ultra high vacuum (UHV) annealing of SiC suggesting the possibility of single crystal growth. And on the other hand, Emtsev et al. [3] had reported improved quality of epitaxial graphene on SiC by atmospheric pressure graphitization than that of UHV treatment. But, a large scale structural quality is limited due to the morphological changes of the surface in the course of high temperature annealing. This method has also its own problems of cost- effectiveness and size limitation of the SiC wafers. In order to overcome these problems, growth of graphene on Si wafer has been initiated by Suemitsu et al. [4]. The process consists of formation of thin 3C-SiC film on Si substrate and sublimation of Si atom from the film to get graphene on it. The quality of graphene, however, still remains a big challenge. Full understanding on the change of surface morphology due to annealing is necessary to optimize the growth conditions so as to get uniform and homogenous graphene. Presently, our efforts concern on the formation of high quality wafer scale graphene on 3C-SiC (111)/Si (111) substrate [5]. In this paper, we would discuss about the surface states and present structural characterization of atmospheric pressure (ATM) and UHV grown epitaxial graphene on 3C-SiC/Si substrate.

## 2. Experimental

Epitaxial graphene were grown on 3C-SiC (111)/SiC (111) substrate. The growth of 3C-SiC (111) epilayers' thin film (600nm) on Si (111) follows the carbonization of Si substrate which results the formation of thin (< 50 nm) crystalline 3C-SiC that serve as a template for 3C-SiC growth. Details of 3C-SiC growth is reported elsewhere [6]. Substrates were cleaned by acetone and propanol, dried by nitrogen blow and loaded to the annealing chamber directly without any pre-treatment. Atmospheric pressure inside the annealing chamber was maintained by flowing argon gas (250 sccm) for

atmospheric pressure annealing, and UHV annealing was performed at about  $10^{-7}$  Pa. Annealing were performed during 5 minutes for all successive experiments. XPS measurements were carried out by utilizing Alka monochromatic source of 1486.6 eV at normal incidence. Raman spectra were taken by green laser (533nm) excitations at room temperature.

### 3. Results and Discussions



Fig. 1 XPS C 1s core level spectra of epitaxial graphene grown on 3C-SiC (111)/ Si (111) substrate by ATM (1300  $^{\circ}$ C) and UHV (1000  $^{\circ}$ C) annealing. The spectra were taken by utilizing Alka monochromatic source of 1486.6 eV at normal incidence.

In order to track the chemical environment of the as grown graphene, XPS C1s core level spectra were probed. Fig. 1 compares the XPS spectra of graphene grown by atmospheric pressure graphitization at 1300 <sup>o</sup>C and ultra high vacuum graphitization at 1000 <sup>o</sup>C. In the case of ATM graphitization, the C 1s spectrum shows a prominent shoulder peak centered at 284.5 eV binding energy. This peak is associated with graphene signals [3]. We noticed graphene signal above 1250 <sup>o</sup>C annealing temperature in the case of ATM graphitization. While

annealing at UHV condition, graphene related signal appears from 950  $^{\rm O}$ C. Graphene related signal of UHV annealed (1000  $^{\rm O}$ C) sample is weak while comparing with ATM annealed one. And shows a prominent peak, around 285.8 eV binding energy which corresponds to the signals from interface layer.



Fig. 2 Raman spectra of 3C-SiC (111)/Si (111) before and after graphitization. Graphitizations were performed at 1000 and 1300  $^{\circ}$ C under UHV and ATM condition respectively.

Raman spectra of graphene formed by UHV and ATM annealing are presented in Fig. 2 along with the spectrum of the sample without annealing. For ATM grown samples, G and 2D peaks are observed at 1584 and 2692 cm<sup>-1</sup>respectively. The UHV grown sample shows a broader 2D peak comparing to ATM and is centered at 2704 cm<sup>-1</sup>. This phenomenon shows that the ATM grown graphene experiences less compressive strain than that of UHV samples. Strong D peak signal of the UHV annealed sample shows many defects and domain boundaries comparing to that of ATM sample. Significant improvement of graphene growth was observed in the case of ATM growth. In the presence of atmospheric pressure by argon atmosphere no sublimation of Si is observed below 1250 °C, whereas Si sublimation started at 950 °C in UHV. The significant high growth temperature results in an enhancement of surface diffusion such that reconstructing of the surface is completed before graphene is formed. This fact is supported by XPS C 1s spectra presented in Fig. 1, where intensity peak related to interface layer is more distinct in UHV than in ATM graphitization. This result is in consistent with other reports for the graphitization of 6H-SiC [3].

#### 4. Conclusion

We have grown graphene on 3C-SiC (111)/Si (111) substrate by UHV and ATM annealing. Graphene signal detected at 1000  $^{\rm O}$ C in UHV and 1300  $^{\rm O}$ C in ATM

annealing were discussed. Enhancement of surface diffusion at ATM annealing results the improvement of graphene quality while comparing with UHV annealing. The improved quality of graphene would lead towards the formation of high quality wafer scale graphene for its viable device applications.

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