

報告番号	甲 第 12319 号
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主 論 文 の 要 旨

論文題目 Growth of epitaxial graphene on SiC (0001) by thermal decomposition in argon and its etching in oxygen atmosphere
(アルゴン雰囲気中 SiC(0001)熱分解法によるエピタキシャルグラフェン成長とその酸素エッチングに関する研究)

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論 文 内 容 の 要 旨

Graphene is a one-atom-thick sheet of carbon atoms arranged in a honeycomb lattice. The unique electric properties make graphene the most ideal two-dimensional material for next generation electronics. Thermal decomposition of silicon carbide (SiC) has been frequently used for fabrication of epitaxial graphene directly on insulating substrates. High quality epitaxially grown graphene and graphene nanoribbons can be formed on SiC (0001) surfaces, which gives a significant potential for electric devices.

Nevertheless, there are still many issues to be solved when manufacturing graphene/SiC devices. Due to complex parameters on the graphene growth on SiC (0001), such as surface morphology, atmosphere and annealing temperature, the growth mechanism is not well understood, so that it is necessary to study graphene growth control technology. In addition, during the graphene formation on SiC (0001), a monolayer graphene layer always grows with a connection to surface buffer layer, which will influence the electron states at the graphene edges and limits the application of graphene nanoribbons on SiC. To improve the graphene growth method of thermal decomposition of SiC (0001) and solve these issues, in this research, graphene formation in argon atmosphere on SiC (0001) and its etching in oxygen were investigated by *in situ* ultra-high vacuum scanning electron microscopy and scanning tunneling microscopy (UHV-SEM/STM) system. Followings show the outline of the studies in this research.

In chapter 1, the basic of graphene and its fabrication methods are introduced. Especially, a current situation and remained issues of graphene growth on SiC substrates are described in detail.

In chapter 2, principles and experimental methods of samples' preparations and characterizations are given.

In chapter 3, different stages of graphene formation on SiC (0001) was investigated by UHV-SEM/STM system. Step-free SiC substrates (terrace width: $>100\ \mu\text{m}$) and vicinal SiC substrates (terrace width: $\sim 100\ \text{nm}$) were annealed at various condition. Graphene growth was firstly carried out on the step-free SiC substrate, that graphene with domain size of about $10\ \mu\text{m}$ was formed in the pit. Due to the uncontrollability of pit formation, the existence of step edges on SiC surfaces is indispensable to grow graphene uniformly. On the vicinal surface, we found the largest increasing rate of terrace widths took place at about 1400°C of annealing temperature. Thus, as a pretreatment for graphene growth, annealing at 1400°C for 3 hours was firstly carried out to enlarge the terrace widths. Subsequently, by heating 1550°C , monolayer graphene grown only from the step edges on wide terrace without pit formation. By such method step-free and pit-free single crystalline monolayer graphene has been fabricated with the size of $4\times 100\ \mu\text{m}$. We also observed graphene growth processes at the same position by *in situ* SEM. We found that the monolayer graphene firstly grew uniformly from step edges with a width of $800\ \text{nm}$, and the buffer layer with a striped morphology began to appear by the subsequent graphene growth. The formation of striped buffer layer is considered to play a role as the pass ways during the diffusion of Si atoms between a monolayer graphene and a newly formed buffer layer.

In chapter 4, we proposed a selective oxygen etching method to remove surface buffer layers without oxygen intercalation below graphene layers by using an extreme low partial pressure oxygen diluted by argon atmosphere. This result must be significant for the application of graphene to electric/spintronic devices. We also found that an etching rate of surface buffer layers depends on a distance from a step edge, which suggests an existence of crystallinity distribution of surface buffer layer on a terrace.

In chapter 5, the growth processes of graphene nanoribbon on SiC (0001) were investigated and we found that nanoribbons at step edges only grow after the surface buffer layer covered on the whole terraces. Free edge monolayer graphene nanoribbons with the width of $200\ \text{nm}$ were also formed by the selective oxygen etching.

Finally, in chapter 6, this research is summarized, and perspectives about graphene and carbon nanomaterials are explained. The new findings obtained in this research may greatly contribute to the development of graphene-based device fabrication technology.