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Formation of graphene/SiC/AlN multilayers synthesized by pulsed laser deposition on Si(110) substrates

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ABSTRACT

We have grown aluminum nitride (AlN) films on Si(110) substrates by pulsed laser deposition (PLD), and investigated the effects of laser power on the crystallinity and surface morphology of the AlN films. First, we epitaxially grew a fairly flat, high-quality AlN film, which contained no rotation domains, onto the Si(110) substrate in a well-lattice-matched relationship. Secondly, we formed a SiC interfacial buffer layer on the AlN film to grow a high-quality 3C-SiC film on the SiC buffer layer by PLD, which gave rise to a 3C-SiC(111)3×3 surface. The root-mean-square-roughness value of the SiC film was smaller than the previously reported values of SiC/AlN multilayers on Si(100) and Si(111) substrates. Thirdly, we grew graphene by annealing the SiC film at a high temperature in an ultra-high vacuum. It was demonstrated that the qualified graphene layer without rotation domains was grown on the SiC film. The formation of voids and the outdiffusion of Al and N atoms from the AlN film were successfully suppressed during the high-temperature annealing.

1. Introduction

Epitaxial growth of graphene on silicon carbide (SiC) is promising for fabricating a wafer-scale, high-quality graphene. To produce a graphene on a SiC substrate surface, the SiC substrate is annealed in an ultrahigh vacuum at temperatures of 1200–1600 °C to evaporate Si atoms from the surface and encourage remaining C atoms to form $C{=}C$ bonds. The growth of graphene on SiC has the advantages that it is composed of conventional semiconductor processes such as film deposition and thermal treatment and it gives an atomically flat graphene without wrinkles. This is because the growth of graphene on SiC does not involve the transfer processes used in chemical vapor deposition and mechanical exfoliation of graphite. The growth of graphene on SiC under high-temperature (~1600 °C) and high-pressure conditions (~1 atm) has been also carried out [1,2]. Emtsev et al. have reported that high-quality graphene films with domain sizes lager than 50 μm are successfully formed on Si-face SiC substrates by annealing at 1650 °C under an Ar gas atmosphere of ~1 atm [2]. Rana et al. have recently reported that graphene growth on SiC by Si selective etching using tetrafluorosilane (SiF_4) is introduced, where SiF_4 in Ar ambient selectively etches Si from the SiC surface at temperatures above 1400 °C, leaving the C as graphene [3].

The epitaxial graphene growth using thermal treatment of SiC substrates is highly compatible with conventional semiconductor processes. However, present SiC wafers are expensive and small in diameter, which would impede the industrialization of the epitaxial graphene. Formation of graphene by annealing cubic SiC on Si substrates (3C-SiC/Si) at high temperatures is one of the possible solutions to overcome the problem of the limitation in the size and cost-effectiveness of present SiC wafers [4,5]. This technique is known as graphene on silicon. However, a problem with the SiC epitaxial growth on Si and the graphene growth on SiC/Si is that Si outdiffusion from a Si substrate often occurs at high temperatures, leading to the formation of hollow voids at the SiC/Si interface [6–8]. In addition, it suffers the formation of dislocations and stacking defects in the SiC film because of the large mismatches in the lattice constant (~20%) and thermal expansion coefficient (~8%) between 3C-SiC and Si. To achieve qualified SiC/Si heteroepitaxy, insertion of an appropriate intermediate layer is strongly required, which bridges the lattice mismatch at the interface and impedes the Si outdiffusion from the substrate. It has been proposed that SiC/AlN/Si multilayered structures are formed to prevent the formation of hollow voids at high temperatures during SiC or graphene growth [9–11]. The relatively close lattice constant and thermal expansion coefficient of AlN with those of SiC make the material quite promising as an intermediate layer for high-quality SiC growth on Si. The primary role of the AlN layer on Si is on forming a diffusion barrier against Si atoms. The AlN layer on Si also functions as an insulating layer on which SiC and graphene devices can be fabricated. A field effect transistor (FET) can be fabricated on graphene/SiC/AlN/Si. The graphene FET consists of source, gate,

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and drain electrodes and a gate insulating layer formed on the graphene layer. The SiC and AlN layers play a role in insulating the FET from the Si substrate. A problem with the FET fabricated on graphene/SiC/Si is that leak current flows from the graphene to the Si substrate through the SiC layer owing to the presence of defects in the SiC layer [12]. As AlN is an excellent insulating material, insertion of the AlN layer between the SiC layer and the Si substrate would suppress the leak current.

Recently, we have conducted the heteroepitaxial growth of 3C-SiC(111) on a 2H-AlN(0001) intermediate layer on Si(100), and showed that the AlN layer prevents the formation of hollow voids during the SiC growth [13–15]. Additionally, we have found that the formation of a SiC interfacial buffer layer on AlN/Si at 650 °C by ultralow-pressure chemical vapor deposition (ULPCVD) using monomethylsilane (CH_3SiH_3, MMS) suppresses the outdiffusion of Al and N atoms from the AlN layer during the SiC growth and further improves the crystallinity and roughness of the SiC film [16]. A MMS molecule already contains a Si–C bond in itself and has the same stoichiometry as of SiC crystal. Previous studies of low pressure chemical vapor deposition and gas-source molecular beam epitaxy using MMS reported successful growth of 3C-SiC films on Si(100) substrates at low temperatures [17–23].

It has been reported that 3C-SiC(111) films are grown epitaxially on Si(110) substrates in a well-lattice-matched relationship [4,24]. Similarly, there is a well-lattice-matched relationship between AlN(0001) and Si(110) lattices. Fig. 1 shows the topmost atomic arrangements of (a) Si(110), (b) 2H-AlN(0001), and (c) 3C-SiC(111) surfaces, where the circles represent the elements: (a) Si atoms, (b) Al-N pairs, and (c) Si-C pairs. The Si atom spacing along the Si[001] axis with the length of $a_{\mathrm{Si}}[001]=0.543$ nm is close to the Al-N pair spacing along the AlN[01$\bar{1}$0] axis with the length of ($\sqrt{3}a_{\mathrm{AlN}}$) $[01\bar{1}0]=0.539$ nm. The Si atom spacing along the Si[$\bar{1}$10] axis with the length of ($a_{\mathrm{Si}}/2$) $[\bar{1}10]\times4=1.536$ nm nearly coincides with the Al-N pair spacing along the AlN[2$\bar{1}\bar{1}$0] axis with the length of $a_{\mathrm{AlN}}[01\bar{1}0]\times5=1.555$ nm. Thus, the lattice of the 2H-AlN(0001) surface almost conforms to that of the Si(110) surface, as indicated by the rectangles. Concurrently, the lattice of the 3C-SiC(111) surface almost conforms to that of the AlN(0001) surface, as indicated by the hexagons.

The heteroepitaxy of AlN on SiC substrates has been intensively carried out to fabricate AlN light-emitting diodes with the shortest wavelength and to form gate insulators for SiC field effect transistors as well as AlN buffer layers for gallium nitride optical devices [25–29]. However, few studies have clarified the crystallinity and surface morphology of SiC films grown on AlN [30–33]. The studies on epitaxial graphene on AlN/Si are especially limited to very few reports [9,10,14]. Although there are some studies of epitaxial growth of graphene on SiC/AlN/Si(111) and SiC/AlN/Si(100), the SiC films grown on the AlN intermediate layers show rugged surface with the root-mean-square roughness (RMS) of above 10 nm [9,10] The surface roughness of the SiC films is very crucial because the surface degradation causes the degradation of the crystalline quality of the graphene layers [9,10]. To the best of our knowledge, there is no study on the SiC growth on AlN/Si(110) and the growth of epitaxial graphene on the SiC/AlN/Si(110) multilayer structure.

In this study, in order to further improve the crystallinity and roughness of the SiC films on Si and epitaxial graphene layers thereon, we have grown AlN layers on Si(110) substrates by pulsed laser deposition (PLD), and investigated the effects of laser power on the crystallinity and surface morphology of the AlN layers. We then formed a SiC interfacial buffer layer at a low temperature on the highest-quality AlN layer obtained in this study by ultralow-pressure chemical vapor deposition (ULPCVD) using monomethylsilane (MMS, CH_3SiH_3) to grow a 3C-SiC film on the SiC buffer layer by PLD. Finally, we grew graphene by annealing the SiC film at a high temperature in an ultra-high vacuum.

2. Experimental methods

AlN and SiC films were grown in a high vacuum chamber, whose base pressure was 3.0×10^{-5} Pa. The substrate used was a boron-doped Si(110) (ρ=8–12 Ω cm) wafer, which was cut to 5×35 mm^2. Prior to introduction into the chamber, the substrate was chemically cleaned by the RCA method. Both the short side ends of the substrate were sandwiched between two tantalum plate electrodes of a sample holder. By causing a DC current to flow the substrate using a power supply (INSTEK PSW30-36), the substrate was resistively heated in a vacuum, and was degassed at 300 °C for 60 min. For the AlN growth, an AlN target (> 99.9%, 25 mm in diameter) was rotated at 7 rpm. A KrF* excimer laser with a wavelength of 248 nm was used for ablation of the AlN target. The pulse duration of the laser was 30 ns. The laser was focused onto the target in the vacuum chamber by means of lens with an incident angle of 45°. The laser repetition rate was set at 20 Hz. The area of the laser spot onto the target surface was 0.0123 cm^2. The laser power was varied from 50 to 250 mJ/pulse (4.07–20.4 J/cm^2). The substrate was resistively heated during the deposition process. The substrate temperature was kept at 750 °C. N_2 gas (99.9999%) pressure was fixed at 1 Pa. An SiC buffer layer was then formed onto the AlN layer grown at 200 mJ/pulse by ULPCVD using MMS (99.999%) at a

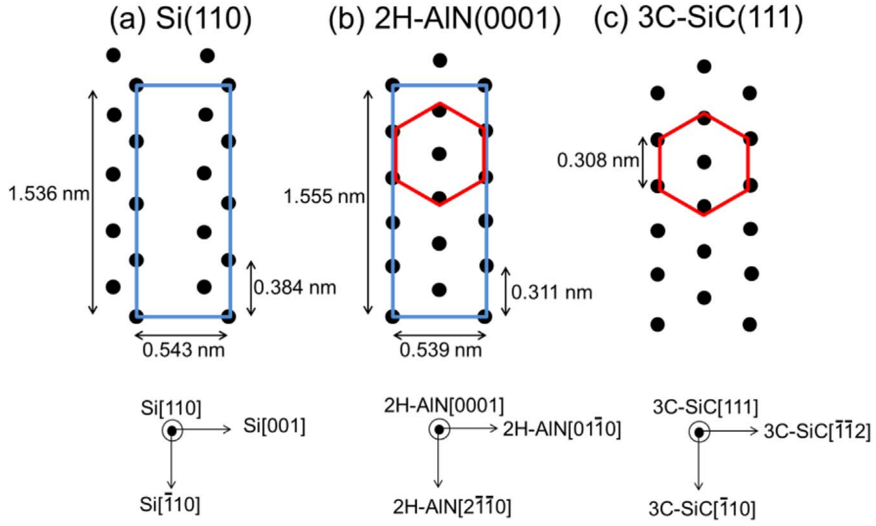

Fig. 1. The topmost atomic arrangements of Si(110), 2H-AlN(0001), and 3C-SiC(111) lattices, where the circles represent the elements: (a) Si atoms, (b) Al-N pairs, and (c) Si-C pairs.