Direct growth of graphene on sputter-deposited Al_2O_3 thin layer on SiO_2/Si substrate by thermal chemical vapor deposition method

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Introduction

Graphene is a single atomic layer of carbon atoms that form a honeycomb structure, and thus, it is the ultimate thin layer. Thanks to its high carrier mobility, graphene is one of the materials with most promise to open a new era of electrical and optical integrated circuits. A method needs to be developed to grow graphene directly on sub-nm-order flat insulating layers in order to fabricate electrical and optical devices that utilize graphene. In addition, using a Si wafer as a substrate is strongly desirable. The current big Si-electronics industry has accumulated enormous amounts of knowledge and know-how concerning Si. Thus, starting graphene-electronics on a Si substrate seems practical.

This paper discusses the development of a new method for growing graphene directly on an insulating layer fabricated on a SiO_2/Si substrate without any metal catalysts. We recently reported on a growth method for graphene on a sapphire substrate [1, 2]. This time we applied this growth method to grow graphene on an Al_2O_3 layer sputter-deposited on a Si substrate, and then we found that a sputtered Al_2O_3 layer has the catalytic ability to grow graphene just like sapphire substrates.

Experimental

An Al_2O_3 thin layer was sputter-deposited on a SiO_2 (200-nm thick) /Si (525-µm thick) substrate by using magnetron sputtering equipment (E-400S-TY, ANELVA). An aluminum oxide target was sputtered using 400-W power and was deposited on a SiO_2/Si substrate that was facing the target at a tilted angle of 30˚. The pressure during sputtering was maintained at 2×10^{-1} Pa using a mixed flow of argon and oxygen. The flow ratio of argon to oxygen was 95/5.

The surface roughness of the sputter-deposited Al_2O_3 layer was measured by using an atomic force microscope (AFM, Nano-R, Pacific Nanotechnology). The patterned Al_2O_3 layer was fabricated by using a lift-off method with two-layer photo-resists to measure its thickness by using an AFM.

Graphene was grown directly on a sputtered Al_2O_3/SiO_2/Si substrate by using thermal chemical deposition without any metal catalysts. The substrate was treated at 800˚C for 30 min with a flow of dried air to burn off organic contaminations on the surface of the substrate before the growth. The growth temperature was 800˚C using propylene flows (10 ml/min) as the source gas and argon (800 ml/min) as the carrier gas. The temperature was kept at 800˚C for 30 min with an argon flow after the growth to prevent additional deposition of organic compounds at lower temperatures, and then the temperature decreased naturally.

The morphology of the grown graphene was observed by using a scanning electron microscope (SEM, S-4800, Hitachi) to evaluate its domain size. The number of graphene layers grown on a sputtered Al_2O_3/SiO_2/Si substrate was determined by measuring the optical transmittance of the graphene grown on a sputtered Al_2O_3/sapphire substrate under the same growth conditions. It is assumed that the growth rate of graphene on the two substrates is the same. The number of layers is calculated based on the fact that the optical absorbance of a single graphene is 2.3%. The details are described by Okai et al. [2]. The electrical conductance of the grown graphene was measured by using the four-probe method and the electrical conductivity was calculated using the measured averaged number of graphene layers and by assuming that the thickness of one layer was 0.34 nm thick.

Results and Discussion

A nm-order flat substrate surface is essentially required for growing graphene to be used in electrical devices, because surface fluctuation affects the carrier mobility and generates electrical localized levels in grown graphene. The surface roughness for a 1-µm-square area of a sputtered Al_2O_3 thin layer at a thickness of 34 nm on a SiO_2/Si substrate was measured by using an AFM. The root-mean-square roughness is 0.15 nm, which is as good as that for a SiO_2/Si substrate. The roughness increases along with the increase in the thickness of the Al_2O_3 layer as expected, whereas it is notable that the roughness is unchanged for thicknesses up to 150 nm. We used an approximately 30-nm thick Al_2O_3 layer sputtered on a SiO_2/Si substrate for the graphene growth for this study.

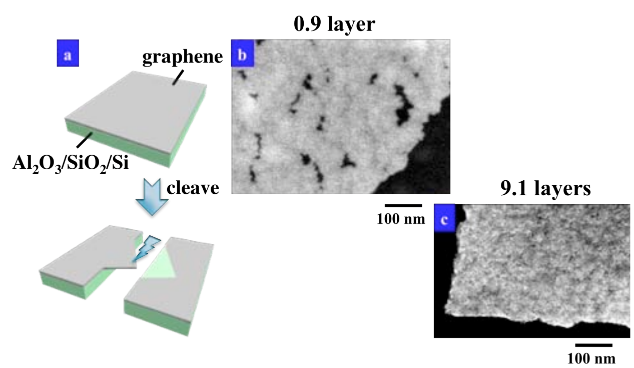

Fig. 1 (a) Our method to observe the grown graphene on sputtered Al_2O_3 layer on SiO_2/Si substrate by a scanning electron microscope (SEM). SEM views of the grown graphene with 0.9 layer (b) and with 9.1 layers (c).

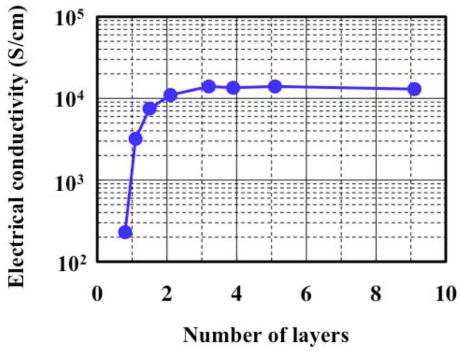

Fig. 2 Electrical conductivity dependence on the number of layers of graphene

We observed the graphene growth on an $Al_2O_3/SiO_2/Si$ substrate by using SEM to clarify its growing process. Figure 1(a) shows our method to observe the grown graphene by using SEM. It is difficult to observe the grown graphene on SiO_2/Si substrate due to the charge up effect caused by the insulating SiO_2 layer. We then cleaved the graphene-grown substrate and observed suspended graphene layers near the cleaved facets of the substrate. Figure 1(b) shows a SEM view of the grown graphene with 0.9 layer measured by using an optical transmission method. The grown graphene is not a continuous film and there are some empty spaces between the graphene platelets. The size of the graphene platelets, which is the domain size, is more than 30 nm. Figure 1(c) shows a SEM view of the suspended grown graphene with 9.1 layers. It is a continuous film consisting of graphene platelets.

The electrical conductivity dependence on the number of layers is shown in Fig. 2. The electrical conductivity increases with an increase in the number of layers and it saturates to 1.4×10^4 S/cm for a grown graphene of more than three layers. We confirmed by looking at the SEM that a grown graphene with 2.l layers has small empty regions in it, whereas one with 3.1 layers is a continuous film. These results show that the growth process of graphene is as follows: Graphene platelets start to grow at reactive points on the surface of a sputtered Al_2O_3 layer. We suppose that the hydroxyl group on the surface of a sputtered Al_2O_3 layer is the reactive point. Graphene platelets grow to a domain size of 30 nm. The determining factor for the domain size seems to be the distance between reactive points. Neighboring platelets grow and collide against each other to stop the growth. Before the whole surface of an Al_2O_3 layer is covered with graphene platelets, the second layer of growth starts on the first layer consisting of graphene platelets. We guess that the reactive points for the second layer growth are the carbon radicals on the edges of graphene platelets in the first layer. The third and succeeding layers grow in the same way. The grown graphene layer becomes a continuous film when the number of layers exceeds three.

We succeed in growing graphene selectively on a line-patterned Al_2O_3 film fabricated on a SiO_2/Si substrate without growing it on SiO_2 surface. Figure 3(a) shows a schematic of selective-area growth of graphene on a patterned Al_2O_3 film. SEM images of a 4-µm-wide line-patterned Al_2O_3 film before and after graphene growth are shown in Fig. 3(b) and (c), respectively. The surface of line-patterned Al_2O_3 film (center) and SiO_2 film (both sides) looks very flat before the growth. On the other hand, after the growth, the surface of a line-patterned Al_2O_3 film has a morphology shown in Fig. 3(d) while the SiO_2 surface remains looking flat. We also confirmed that line-patterned Al_2O_3 film has electrical conductivity and SiO_2 film still insulates after selective-area growth of graphene.

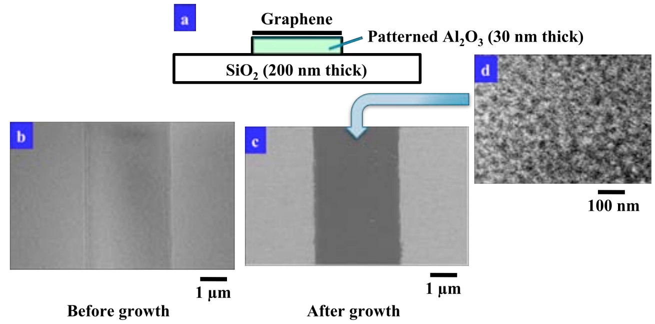

Fig. 3 (a) Schematic of selective-area growth of graphene on patterned Al_2O_3 film. SEM images of a 4-µm-wide line-patterned Al_2O_3 film before (b) and after (c) graphene growth. (d) A high-resolution SEM image of the grown graphene on the line-patterned Al_2O_3 film.

Conclusions

We have successfully grown graphene on the sputter-deposited Al_2O_3 thin layer on a SiO_2/Si substrate by using thermal chemical deposition at 800˚C with propylene used as the source gas. The surface of the sputtered Al_2O_3 layer is as flat as that of a SiO_2/Si substrate at a thickness of less than 150 nm. The grown graphene has a piled structure of graphene platelets with a domain size of more than 30 nm. The graphene film is continuous when there are three or more layers and its electrical conductivity is as high as 1.4×10^4 S/cm.

We succeed in growing graphene selectively on a line-patterned Al_2O_3 film fabricated on a SiO_2/Si substrate without growing it on SiO_2 surface.

References

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