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Study on the graphene transfer process from graphitized SiC substrates

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1. Introduction

Graphene has attracted a great deal of attention due to excellent electrical properties and unique physical characteristics. However, the physical and chemical exfoliation of graphite can produce only a tiny piece of graphene [1]. Thus, the processes to fabricate a large area, high quality, single layer graphene have been studied intensively. One of early approaches was a thermal graphitization of 4H-SiC substrate at a very high temperature range (1400°C - 1600°C)[2-4]. However, the control of layer by layer thickness was difficult in this process due to the high process temperature. Also, the transfer of a graphene layer from the graphitized SiC substrate was not as easy as the process using metal catalysts substrate such as Ni and Cu [5-6]. Recently, Unarunotal et al showed that e-beam evaporated Au/polyimide film can be used to peel off a graphene from the graphitized 6H-SiC [7]. In this work, Ni and Au adhesion layers are compared to understand the process of graphene transfer from a graphitized SiC and potential damage to the graphene is examined.

2. Experiments

In this work, Si face 4H SiC substrates were used as a template for a graphene fabrication. After an initial clean using 100:1HF, the surface of SiC substrate was graphitized using a high power KrF laser in a vacuum ambient (<5E-7Torr). Same graphitization processes were used for all samples to compare the differences due to the metal adhesion layer. Then, 100nm of Ni, Au films are deposited using an e–beam evaporation process.

Fig.1 shows a schematic flow of the graphene transfer process used in this work. After the metal deposition, polyimide layer was applied as a supporting material for metal thin films deposited on the graphitized SiC. The sidewall of metal layer was protected from the polyimide to allow the metal wet-etch. The, the combined stack of polyimide/metal/graphene was detached from the SiC surface and bonded to SiO_2 substrate. The bonding was performed at sub-atmospheric pressure to minimize the air gap between the graphene and SiO_2. Finally, the metal/polyimide stack was etched off in TFA and $FeCl_3$ solution leaving the graphene on SiO_2. Typical process time was 5 min for 100nm thick metal layers. SEM photograph of the transferred graphene is shown in Fig.2. Then, the quality of transferred graphene layers were analyzed using SEM, AFM and Raman Spectroscopy (λ=514nm).

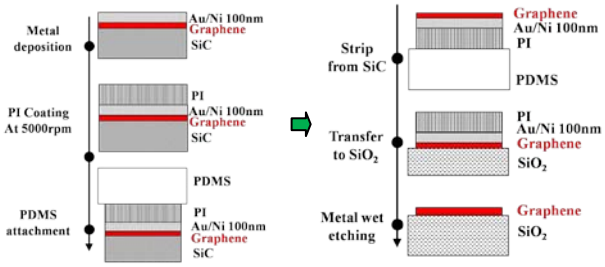


Fig.1 Schematic flow and picture of samples showing the process of graphene transfer from the SiC substrate

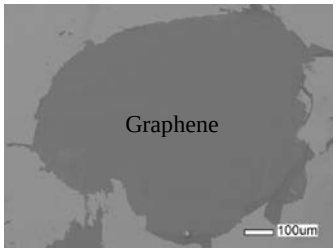


Fig.2 SEM photograph of transferred graphene on SiO_2

3. Physical analysis of graphene transferred on SiO_2

After the graphitization, a few nm of multi layer graphene seems to be formed on the SiC surface as indicated by the strong 2D peak of Raman spectrum shown in Fig.3 (a) [8]. Raman spectrum of SiC surface still shows the signature of multi layer graphene even after the graphene transfer, but the height of 2D peak is slightly lowered as the thickness of graphitized layer is reduced. AFM indicates that the thickness of transferred graphene is more uniform with Au adhesion layer than at Ni adhesion layer (Fig.4).

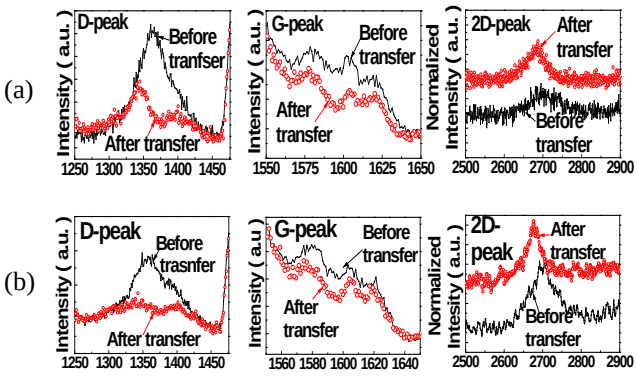
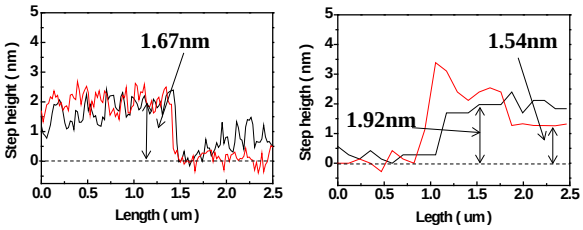


Fig.3 Raman Spectrum of graphitized SiC ; (a) Au (before / after) (b)Ni(before/after) the graphene transfer



(a) (b)

Fig.4 Step heights of graphene transferred on SiO_2, measured by Atomic Force Microscope(AFM); (a) Au, (b) Ni

In case of Au, the step heights of transferred graphene were around 1-1.5nm, close to the thickness of monolayer graphene on SiO_2 while those of Ni sample varied in wide range.

The thickness of transferred graphene is further investigated with multi-point Raman spectrum. The Raman spectrum of graphene transferred using Au adhesion layer is uniform and shows lower G/2D peak ratio indicating the signature of monolayer (Fig.5 (a))[9]. On the other hand, the graphene transferred with a Ni adhesion layer shows the scattered G/2D peak ratio indicating a wide thickness distribution.

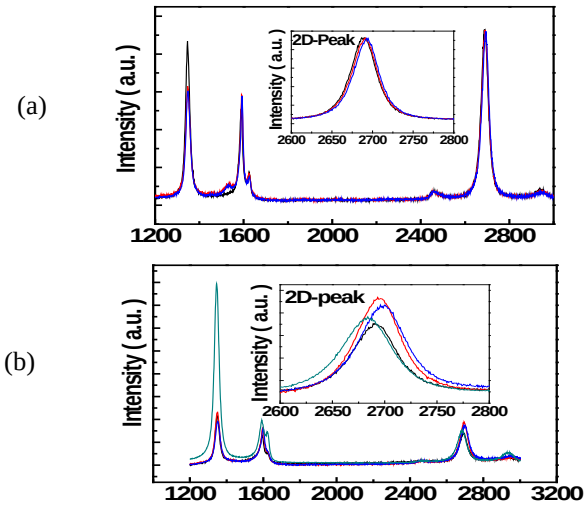


Fig.5 Raman Spectrum of graphene films transferred to SiO_2 300nm/Si substrate; (a) Au, (b) Ni

Based on the above observation, the graphene transfer process using a metal adhesion layer appears to be related to the local adhesion between metal and graphene as shown in the Fig.6. Formation of graphene on SiC is typically initiated at the surface of facets and the graphene islands on neighboring facets are gradually connected after successive anneals to form a wide area graphene. Then, further anneals increase the thickness of graphitized layer. When the metal films deposited on the graphitized surface of SiC is detached from the surface, a mono layer graphene or a few layers of graphene are detached from the surface. Au adhesion layer appears to take only a mono layer graphene while Ni adhesion layer seems to hold multi layer graphene at this detaching step as shown above with AFM and Raman analysis.

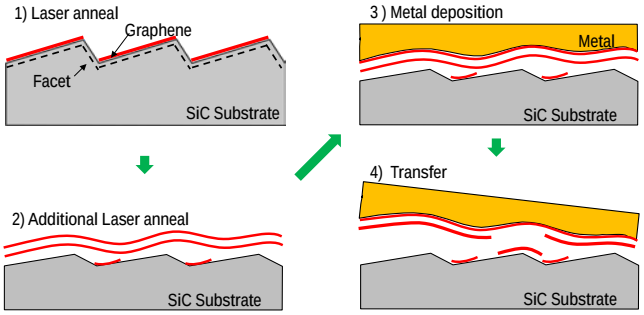


Fig.6 Schematic diagram of graphene transfer from a grahitized SiC surface using a metal thin film adhesion layer

What makes the difference between Au and Ni is not clearly understood. Experimentally, Ni adhesion seems to be stronger than Au because more force should be applied to detach the Ni layer. Theoretical estimation of the van der Waals forces between metal and graphene is 16.7×10^{-20}J for Au and 10.6×10^{-20}J for Ni, while that for graphene to graphene is 7×10^{-20}J. Thus, the adhesion forces between Au/graphene and Ni /graphene allows the detachment of thin graphene layer from the graphitized surface, but the difference in van der Waals force is not enough to explain the thickness variations and material dependence. Metal deposition process, pressure applied to polyimide and other process related parameters should be more carefully studied to develop a stable graphene transfer process.

3. Conclusions

A process to transfer a monolayer graphene to SiO_2 substrate from a graphitized SiC surface is successfully developed. The differences between Au and Ni adhesion layer are examined using SEM, AFM, and Raman spectroscopy. The graphene transferred using the Au film appears be a mono layer while those transferred using Ni shows wide range of thickness from mono layer to multi layer graphene.

Acknowledgements

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