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Optical and electrical properties of graphene layers directly-grown by Alcohol-CVD

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1. Introduction

Graphene has been attracted interest due to its high transparency and conductivity. It is a promising material for transparent electrodes and solar cells. Chemical vapor deposition (CVD) from gaseous hydrocarbon[1] and lately alcohol[2] sources has shown promises for large-scale graphene growth. Recently, a direct growth of graphene using propane[3] and alcohol[4] has attracted due to an advantage of passing through a transfer process. In this study, we report on an optical transmittance and a sheet resistance of directly-grown graphene sheet on sapphire substrate. SiO_2/Si and *n*-6H-SiC substrates were also examined for graphene direct growth.

2. Experimental

Graphene layers were grown on sapphire substrate consisting of various orientations such as *c*-plane (0001), *r*-plane (01-12) and *a*-plane (11-20) substrate. The sapphire substrates were annealed at 1000 ^{o}C for 3 hour to obtain a step morphology. SiO_2 (95nm)/*p*-Si and both of silicon and carbon face of *n*-6H-SiC (0001) substrate were used as references. 1-10 sccm of ethanol was used as a source with Ar and H_2 mixture gas ambient at 10 Torr, the temperature range of 900 to 1000 ^{o}C for 1-20 min. The thickness was determined by atomic force microscope (AFM). The optical characterizations have been done by optical microscope, Raman spectroscopy (532nm), and transmittance by UV-visible spectrometer. The sheet resistance was measured by a four-point probe method and Hall effect measurement performed in a van der Pauw geometry. The samples on SiO_2/Si was prepared Hall bar configuration with back gate electrode.

3. Result and discussions

Fig.1 shows (a) the typical AFM image of annealed *a*-sapphire substrate, (b) optical microscope image of graphene layer on *a*-plane sapphire substrate, (c) the AFM image of the thickness and (d) scan profile, showing that the direct-growth of graphene had been realized. Fig.2 shows the typical Raman spectra of graphene layers. The graphene layers on *a*-sapphire substrate showed G-band (~1598cm^{-1}), D-band (~1347cm^{-1}), and small 2D-band (~2700cm^{-1}) peaks, which are originated to graphite mode. The layers had a large number of edge defects and disorder as evidenced by the weakly defined presence of D' band at 1620cm^{-1}. The characteristics of highly disordered sp^3 hybridization of carbon lattice was not observed in graphene layer synthesized.

Fig.3 (a) shows the relationship between the thickness of graphene layers and the optical transmittance. The line shows a calculation of graphite film. The transmittance on each sample showed a dispersed tendency and weakly accorded to the theoretical value. It suggests that the directly grown layers have non-uniform thickness. Fig.3 (b) shows the relationship between the thickness and the sheet resistance. The line shows a calculated value of graphite. The observed sheet resistance decreased with the thickness, and was higher as 1-2 orders than the calculation. However, the sheet resistance had improved after annealing of sapphire substrate compared with the previous results which *a*-plane sapphire substrates were used as as-received condition.

Table I summarized the typical Raman peaks and Hall effect measurement with different substrate. The conductivity type showed *p*-type conductivity on insulating substrate. The sheet resistance showed similar value of ~4 kΩ/sq in both of sapphire substrate and SiO_2/Si substrate. The Hall mobility showed ranging of 55.3 to 99.3 cm^2/Vs, which are smaller than that value of Ref.9 in spite of similar value of carrier density as ~10^{13} cm^{-2}. The low Hall mobility may originate to the nano-domain (~10nm) of the direct grown graphene layers. The modulation of the drain current by the gate electric field was observed in change of 5% with varied gate voltages from -50 V to +50 V. The typical field effect mobility was calculated as 1.7 cm^{2}V^{-1}s^{-1}.

4. Conclusions

For 1.5-4.3 nm thick graphene layers on sapphire substrates, the sheet resistance varies from 7 to 2 kΩ/sq with an optical transparency from 89.6 % to 80.5 % at the wavelength of 550nm. The direct graphene layers have sensitive characteristics in the surface conditions of substrate and could be performed high conductivity by optimizing the direct growth conditions including a substrate pretreatment.

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References

[1] K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J-H. Ahn, P. Kim, J-Y. Choi, and B. H. Hong, Nature, **457**, 706 (2009). [2] Y. Miyasaka, A. Matsuyama, T. Nogi, A. Nakamura, and J. Temmyo, Phys. Status Solidi c **8**, 577 (2011). [3] A. Michon, S. Vezian, A. Ouerghi, M. Zielinski, T. Chassagne, and M. Portail, Appl. Phys. Lett., **97**, 171909 (2010). [4] Y. Miyasaka, A. Nakamura, and J. Temmyo, Jpn. J. Appl. Phys. **50**, 04DH12 (2011). [5] Y. Wang, X. Chen, Y. Zhong, F. Zhu, and P. Loh, Appl. Phys. Lett., **95**, 063302 (2009). [6] A. Reina, X. Jia, J. Ho, D. Nezich, H. Son, V. Bulovic, M. S. Dresselhaus and J. Kong, Nano Lett. **9**, 30 (2009). [7] T. Sun, Z. L. Wang, Z. J. Shi, G. Z. Ran, W. J. Xu, Z. Y.

Wang, Y. Z. Li, L. Dai, and G. G. Qin, Appl. Phys. Lett., **96**, 133301 (2010). [8] J. Kim, M. Ishihara, Y. Koga, K. Tsugawa, M. Hasegawa, and S. Iijima, Appl. Phys. Lett., 98, 091502 (2011). [9] T. Fujii, M. Sato, Y. Yonezawa, and Y. Ichikawa, The 71st Fall Meeting, The Japan Society of Applied Physics, 16a-ZQ-8, Nagasaki Univ., Sept. 2010.

Table I. Typical Raman peaks and Hall measurements with different substrates

substrate	orientation	G (cm^{-1})	D (cm^{-1})	2D (cm^{-1})	I_D/I_G	I_G/I_{2D}	Trans mittance (%)	Thickness (nm)	Sheet R (Ω/sq)	Conductivity (S/cm)	Carrier density (cm^{-2})	Mobility μ_H (cm^2/Vs)	type	comments and reference
sapphire	*c*-(0001)	1599	1351	2698	1.7	2.7	89	3	4719	757	2.4×10^{13}	55.3	*p*	sub. annealed ~100nm step, Tg=1000°C, $F_{ethanol}$=10sccm, 10min
	a-(11-20)	1598	1347	2700	1.4	3	93	2	4146	1148	2.1×10^{13}	69.5	*p*	sub. annealed ~20nm step, Tg=900°C, $F_{ethanol}$=10sccm, 10min
(previous)	*a*-(11-20)	1598	1351	2706	2.1	2.2	85	5	9790					sub. as-received surface[4], Tg=900°C, $F_{ethanol}$=50sccm, 15min
6H-SiC	(0001)	1602	1353	2696	1.4	7.2		16 (260μm)	(12)	(3.2)	(1.1×10^{16})	(45.8)	(*n*)	*n*-type substrate
SiO_2/Si	-	1600	1345	2686	1.8	4		3	4467	799	1.4×10^{13}	99.3	*p*	Hall-bar configulation
Ni film, quartz							76.3		~280					CH_4[1]
Ni film, glass							85	6-7	310		6.1×10^{13}	330	*p*	CH_4 for electrode of OLED[7]
Cu foil, acryl							~80		3000					CH_4, plasma CVD[8]
Cu foil 25μm, PMMA								~1	520	18324	4.1×10^{13}	434	*p*	CH_4[9]

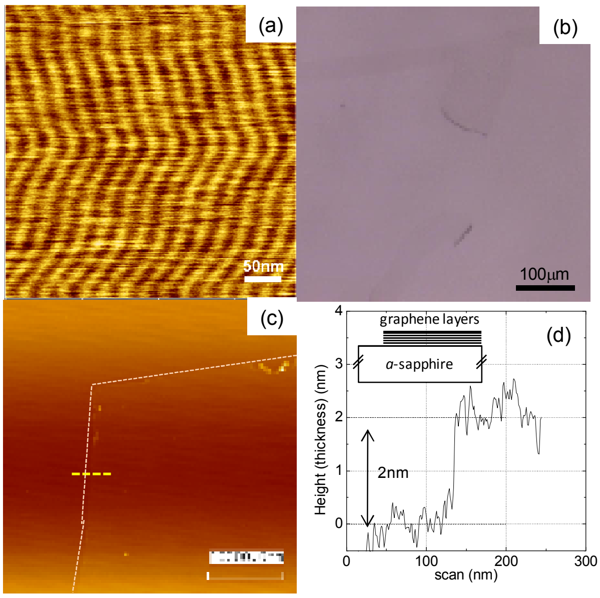

Fig.1. (a) AFM image of *a*-plane sapphire substrate annealed at 1000°C for 3h. (b) optical microscope image, (c) AFM image for thickness measurement, (d) the scan profiles.

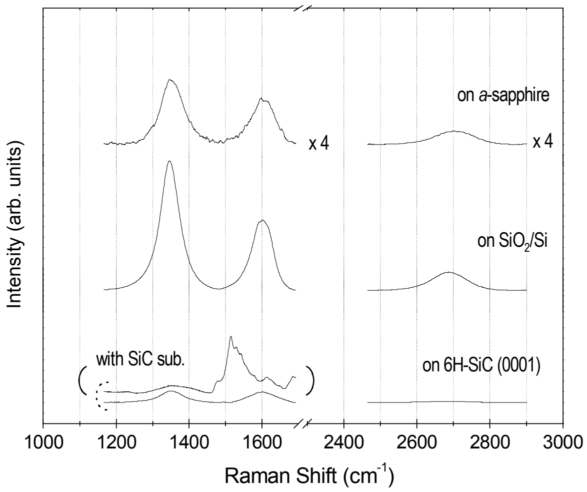

Fig.2. Typical Raman spectra of graphene layers directly grown at 1000°C for 10 min.

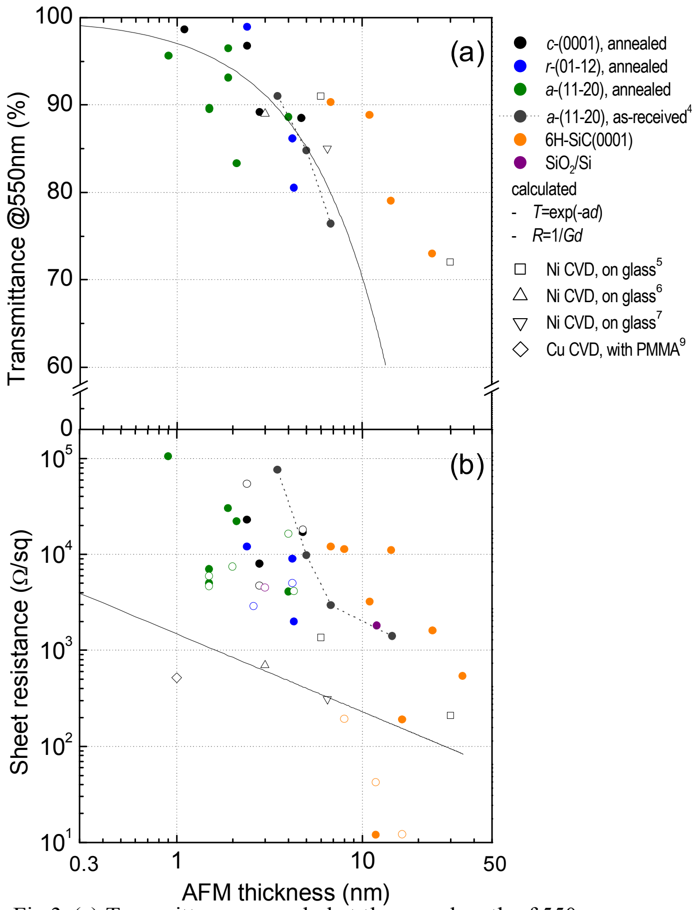

Fig.3. (a) Transmittance recorded at the wavelength of 550nm with thickness, (b) Sheet resistance with different thickness. The solid circles were taken by four probe method, and the open circles were taken by van der Pauw method of Hall measurement.