Influence of the buffer layers on growth and quality of graphene films grown by pulsed laser deposition

Pramod Kumar1,2, Indranil Lahiri2 and Anirban Mitra1

1 High Power Laser Lab, Department of Physics, Indian Institute of Technology Roorkee, Roorkee -247667, India
2 Nanomaterials and Applications Lab, Department of Metallurgical and Materials Engineering, Indian Institute of Technology Roorkee, Roorkee-247667, India

E-mail: mitrafph@gmail.com

Keywords: graphene, PLD, raman spectroscopy, SEM, XPS

Abstract

Graphene films were grown onto two different thickness levels of SiO2/Si substrates, with the help of Ni film from highly ordered pyrolytic graphite through pulsed laser deposition. In this work, the influence of thickness of buffer layer of SiO2 on quality, growth, and crystallinity of graphene films in terms of substrate temperatures has been studied. All graphene samples were characterized by using Raman spectroscopy, Scanning electron microscopy (SEM), and x-ray photoelectron spectroscopy (XPS). The micro-Raman spectroscopic results clearly reveal that each SiO2/Si substrate does not behave in same manner during PLD growth process. From the Raman spectra, it was noticed that the quality, growth and crystallinity of graphene films was rapidly improved on commercially available SiO2 (100 nm)/Si substrates, while gradually improved on thermally grown SiO2 (400 nm)/Si substrates upon increasing growth temperature from 700 °C to 800 °C. Moreover, it was also observed from the Raman spectra that the number of graphene layers was found to be quite similar on both SiO2 layer at higher temperature of 800 °C. The microstructure of the as-synthesized graphene films also confirmed that the quality of graphene layers continuously improved over commercially available SiO2 substrates with the increment in substrate temperature. XPS results obviously verify that crystallinity and carbon-carbon bond strength increased while defects reduced on SiO2 (100 nm)/Si substrates with the increase growth temperature from 700 °C to 800 °C. Thus, the presented study provides a facile, rapid, and effective technique to control the quality, growth, and crystallite size of graphene films onto different thickness SiO2 layer on Si.

1. Introduction

Graphene is two-dimensional (2D) structural material formed by single layer of \(sp^2\) bonded carbon atoms [1, 2], which has seen increasing research interest toward the synthesis in the past few years because of its outstanding thermodynamic stability [3], extremely high carrier mobility [4], low optical absorbance [5], and superior mechanical flexibility [6]. Therefore, graphene is expected to be a highly promising next generation electronic material in near future potential applications such as transparent electrodes [7], field effect transistor [8], solar cell [9], highly sensitive sensors [10], supercapacitors [11], Li-ion batteries [12] etc. Thus, it is essential to synthesize high-quality, large-area graphene for better performance in various devices.

In the last decade, many processing routes were developed to synthesize high quality monolayer graphene, since the isolation of single layer graphene from highly ordered pyrolytic graphite by micromechanical exfoliation in 2004 [13]. This process produces better quality graphene in limited size, which does not satisfy the requirement of large-scale electronic application. Another common method is epitaxial growth on silicon carbide, which requires ultrahigh vacuum conditions and high process temperature [14]. Moreover, this method is expensive due to high cost of SiC substrate. Additionally, it is not also easy to transfer the epitaxial
graphene from SiC substrate to any other desired substrates, due to high chemical stability of SiC [15]. Chemical vapor deposition (CVD) has been extensively investigated as an attractive approach in synthesis of graphene using Ni and Cu metal substrates [16, 17]. However, the traditional CVD growth of graphene usually involves high temperature decomposition of carbonaceous gas, which results in the growth of graphene with a high rate and makes the growth process less controllable. Furthermore, the growth of graphene from carbon source using molecular beam epitaxy (MBE) is a very promising approach to overcome some of the limitations of the CVD technique such as the probability to grow graphene directly over a broad range of substrate surfaces including semiconductors and insulators [18–20]. However, this technique also requires ultra-high vacuum (10⁻⁶–10⁻¹² torr) as well as high process temperature.

In order to address these issues, a facile, rapid, effective and controllable technique was developed to obtain graphene films through pulsed laser deposition (PLD). This technique is known to offer the energetic carbon species, up to a few hundred eV and control the thickness per pulse, up to 0.1 Å, by controlling of laser power [21, 22]. In comparison to Cu, Ni has been better successful in forming few layer graphene by PLD technique, due to the high carbon solubility in Ni and large lattice mismatch [23]. Apart from benefits of polycrystalline Ni film, there are some complication including presence of grain boundaries that limit the size of graphene flakes, leading to restriction on electrical properties of graphene. Furthermore, Ni grains can also lead to form clusters as well as exposing surface of the substrate at extreme substrate temperature which results in Ni film dewetting [24, 25]. To reduce the effect of dewetting, Ni films more than 400 nm thickness have been used for the growth of graphene layers [26]. Nevertheless, thicker Ni films increase more carbon species in the film solution, which leads to multilayer graphene formation.

As an alternate approach, a buffer layer of SiO₂ over Si can be used to prevent the diffusion of atoms inside the silicon wafer as well as to reduce the effect of dewetting and surface roughness which has been mostly used in chemical vapor deposition of graphene [27]. Furthermore, metal over an oxide buffer layer demonstrates much less mobility due to increment in adhesion between the film and the substrate [28]. Recently, graphene was grown on complex Ni-Cu substrate from highly ordered pyrolytic graphite using PLD technique at room temperature [29]. Only a limited number of studies were performed on PLD growth parameters that affect quality and growth of graphene films. One of those parameters is substrate temperature. Researchers have given contradictory outlooks about the effect of substrate temperature in PLD growth of graphene layers. Some scientists have reported that quality and growth of graphene film would improve with an increase in substrate temperature [30], while other study have demonstrated degradation in the quality of graphene with an increase in substrate temperature [31, 32]. Therefore, it is believed that substrate temperature has also crucial function in PLD growth of graphene.

In this study, to clarify this controversial issue, few layer graphene was grown on two different thickness levels of SiO₂ on Si substrates, such as commercially available 100 nm thick SiO₂ layer over Si and thermally grown 400 nm thick SiO₂ layer over Si. Graphene was grown with the help of 250 nm Ni film using highly ordered pyrolytic graphite target through pulsed laser deposition technique. The effect of buffer layers of SiO₂ thickness on the PLD growth, quality, thickness, and crystallite size of graphene film in terms of substrate temperatures was also studied. This growth process demonstrates a significant enhancement in the quality, growth, and crystallite size of resulting graphene film, while number of graphene layers were reduced over the SiO₂ (100 nm)/Si substrates by increasing the substrate temperature from 700 °C to 800 °C. Thus, the quality, growth, crystallite size as well as the number of graphene layers can be well controlled on SiO₂ (100 nm)/Si substrates by varying the substrate temperature from 700 °C to 800 °C.

2. Experimental details

Approximately 250 nm thick Ni film was sputtered onto two different types of SiO₂/Si substrates: commercially available SiO₂ (100 nm)/Si and thermally grown 400 nm thick SiO₂ layer over Si. Ni deposition was performed by dc sputtering (Excel Instruments, India) using 100 W Argon plasma at 6.9 mtorr pressure, and 30 sccm Ar flow rate. The purity of the Ni target material was 99.99%. Prior to loading the sample into the system, both SiO₂ wafers were successively immersed in acetone, isopropyl alcohol and distilled water for 5 min each to remove organic impurities from the upper surface. Then, the SiO₂/Si wafers were dried in flowing nitrogen after cleaning.

After Ni deposition, both SiO₂/Si wafers were taken out from the sputtering system and the samples were cut into 1 cm × 1 cm pieces. Then, the Ni coated SiO₂/Si substrates and the commercially available HOPG (SPI Supplies, USA, 446HP-AB, purity 99.999%) target were loaded into the PLD chamber (Excel Instruments, India). The PLD chamber was initially evacuated to a base pressure of 1 × 10⁻⁸ torr approximately using a turbo molecular pump backed up with a rotary vacuum pump. The distance from target to substrate was kept at 5 cm. The substrate temperature was raised from room temperature to 800 °C at the rate of 10 °C minute⁻¹. Prior to
deposition, each sample was heated up to higher temperature of 700 °C, 750 °C, and 800 °C under vacuum and the samples were thermally annealed at higher temperature for 10 min to improve the crystallinity of Ni film. For the graphene growth, the laser energy and the number of laser pulses were set to be 100 mJ/pulse and 1000, respectively. The graphene growth was carried out for 100 s using 532 nm wavelength of Nd:YAG laser with pulse width 8 ns and a pulse repetition rate of 10 Hz. Table 1 summarizes all the PLD processing parameters used in the present study. In this work, two series of the graphene samples were prepared under the same conditions by varying temperature from 700 °C to 800 °C in steps of 50 °C. First series was deposited on 250 nm thick Ni film over commercially available 100 nm thick SiO2 layer over Si, while second series was deposited on 250 nm thick Ni film over thermally grown 400 nm thick SiO2 layer over Si. After deposition, the PLD heater was turned off, and then the samples were naturally cooled down to room temperature under vacuum.

The growth, quality, number of graphene layers, defect density and crystallite size of graphene film were examined by micro-Raman spectroscopy (Reinshaw microscope). The micro-Raman spectroscopy studies were performed using an Ar-ion laser with an excitation wavelength of 514.5 nm. The laser power was kept at a very low value of 10% of its maximum power to save the samples from unwanted heating. All Raman spectra were collected at the magnification of 50X in the frequency range of 1000 to 3000 cm\(^{-1}\). The Raman spectra were analyzed using a peak fitting procedure to extract the peak positions, full width half maximum (FWHM), and integrated intensity. The microstructure of graphene samples was observed in field emission scanning electron microscope (FESEM, Carl, Zeiss). Graphene films were also characterized using x-ray photoelectron spectroscopy (XPS, PHI 5000 Versa Probe III). XPS studies were carried out using monochromatic Al K\(\alpha_1\) radiation in an ultra-high vacuum (UHV) conditions.

### 3. Results and discussion

Raman spectroscopy is well known technique to determine quality, growth, the number of graphene layers, and crystallite size of graphene films [28, 33, 34]. Therefore, the influence of buffer layers on growth, quality, and number of graphene layers, as well as on crystallinity of graphene films in terms of growth temperatures was first studied by micro-Raman spectroscopy after PLD process. Figures 1(a)–(b) shows the typical Raman spectra collected from the graphene samples as-synthesized on commercially available 100 nm thick SiO2 layer and thermally grown 400 nm thick SiO2 layer, at three different substrate temperatures of 700 °C, 750 °C, and 800 °C, respectively. Intensities ratios (I\(_D\)/I\(_G\) and I\(_{2D}\)/I\(_G\)) and crystallite size of both substrates as a function of growth temperature are given in table 2.

The Raman spectra of graphene samples grown on both Ni (≈250 nm)/SiO2(100 nm)/Si and Ni(≈250 nm)/SiO2(400 nm)/Si substrate consist of three major peaks approximately at 1368 cm\(^{-1}\), 1595 cm\(^{-1}\), and 2718 cm\(^{-1}\), corresponding to D, G, and 2D bands. First, the D-peak appeared at ~1368 cm\(^{-1}\), which signifies the existence of amount of structural defects or defect density in graphene film [35]. Second, the G-peak is raised at ~1595 cm\(^{-1}\) corresponds to in-plane sp\(^2\) carbon-atom vibrations [36], which increases almost linearly with the number of graphene layers increasing [17]. Finally, the presence of 2D-peak at ~2718 cm\(^{-1}\) is the signature of formation graphene layers, arising from double resonance process in K points in the first Brillouin zone [37]. At growth temperature of 700 °C, higher intensity D and broader G peaks were observed, whereas 2D peak was found to be hardly visible on both substrates, as shown in figures 1(a)–(b). In contrast, the

| Laser used | Nd:YAG (Neodymium: yttrium aluminium garnet) |
|------------|---------------------------------------------|
| Target used | HOPG (99.999% purity) |
| Laser Wavelength | 532 nm |
| Laser energy | 100 mJ |
| Pulse duration | 8 ns |
| Pulse Repetition Rate | 10 Hz |
| No. of shots | 1000 |
| Deposition time | 100 s |
| Substrate | 700, 750, and 800 °C |
| Substrate Temperature | Ni (≈250 nm)/SiO2(100 nm)/Si |
| Target to substrate distance | 50 mm |
The intensity of D-peak and full width half maximum (FWHM) of G-peak linearly decreases on Ni/SiO$_2$ (100 nm)/Si with the increment in substrate temperature from 700 °C to 800 °C as expected, while D-peak becomes less intensive and FWHM of G-peak does not reduce more on Ni/SiO$_2$ (400 nm)/Si with the increase substrate temperature. A comparison between all these Raman spectra clearly indicates that lower structural disorder and higher crystallinity are present in graphene films grown on Ni/SiO$_2$ (100 nm)/Si substrate at the higher substrate temperature of 800 °C, while higher defects and lower crystallinity were present in graphene sample grown on Ni/SiO$_2$ (400 nm)/Si substrate at 800 °C. The reason behind this phenomenon is that carbon atoms might be more disordered on SiO$_2$ (400 nm) layer as compared to SiO$_2$ (100 nm) layer due to different adsorption ability of carbon atoms on substrates. Another reason is that the surface of commercially available SiO$_2$ (100 nm) layer seems to be smooth and superior quality as compared to thermally grown of SiO$_2$ (400 nm) layer as evident from SEM images, which may produce more oxygen functional group over the Ni/SiO$_2$ (400 nm)/Si as compared to Ni/SiO$_2$ (100 nm) substrates. Furthermore, it is also clearly observed from the Raman spectra that 2D-peak continuously becomes higher intensive and narrower over Ni/SiO$_2$ (100 nm)/Si as compared to Ni/SiO$_2$ (400 nm)/Si substrate with the increment in substrate temperature, clearly indicating higher crystallinity in graphene film grown on Ni/SiO$_2$ (100 nm)/Si as compared to Ni/SiO$_2$ (400 nm)/Si substrate. Thus, it is obvious that buffer layers play significant role in constructing graphene films. At higher substrate temperature, ablated clusters of carbon atoms have sufficient energy to segregate in nickel, and then precipitate out on Ni surface to form graphene film during cooling process [38]. Therefore, it is clear sign from the Raman spectra that the quality, growth, and crystallinity of graphene film significantly improves with the increment in substrate temperature from 700 °C to 800 °C [30]. As a result, it clearly verifies from the Raman spectrum that substrate temperature also plays a major role to enhance the quality, growth, and crystallinity of graphene film by annealing structural defects with the increment in substrate temperature.

**Table 2.** Intensity ratio of D, G and 2D bands of graphene films deposited on Ni/SiO$_2$ (100 nm)/Si [1st] and Ni/SiO$_2$ (400 nm)/Si [2nd] substrates at three different growth temperatures.

| Growth temp. (°C) | Laser energy (mJ)/pulse | Wave length (nm) | No. of shots | I$_D$/I$_G$ | I$_{2D}$/I$_G$ | Crystallite size (L$_a$ (nm)) |
|------------------|------------------------|-----------------|--------------|-------------|---------------|-----------------------------|
|                  |                        |                 |              | 1st  | 2nd  | 1st  | 2nd  | 1st  | 2nd  | 1st  | 2nd  |
| 700              | 100                    | 532             | 1000         | 2.85 | 1.59 | 0.10 | 0.02 | 5.90 | 10.57 |
| 750              | 100                    | 532             | 1000         | 1.26 | 1.32 | 0.40 | 0.32 | 13.35 | 12.74 |
| 800              | 100                    | 532             | 1000         | 0.34 | 1.16 | 1.70 | 0.43 | 49.46 | 14.50 |

**Figure 1.** Raman spectra of graphene grown at three different substrate temperatures; (a) Ni ($\approx$250 nm)/SiO$_2$ (100 nm)/Si (b) Ni ($\approx$250 nm)/SiO$_2$ (400 nm)/Si.
The plot of crystallite size as a function of growth temperatures is shown. Substrate at higher temperature of 800°C shows a plot of ID/IG and I2D/IG as a function of growth temperature. It is well evident from the figure 2(a) and table 2 that ID/IG peak ratio decreases and I2D/IG peak ratio continuously increases on commercially available SiO2 (100 nm)/Si substrate with increasing growth temperature from 700 to 800 °C, which clearly indicates that the amount of structural defects and the number of graphene layers rapidly reduce over 100 nm thick SiO2 with the increase of growth temperature. While ID/IG decreases very slowly, I2D/IG peak ratio increases gradually on thermally grown SiO2 (400 nm)/Si substrates as shown in figure 2(b) and table 2, which obviously indicates that the amount of defects and the number of graphene layers gradually decrease on 400 nm thick SiO2 substrate with the increase of growth temperature. The intensity ratio of the 2D-peak and the G-peak (I2D/IG) is usually utilized to provide qualitative information about the number of graphene layers [37]. The peak intensity ratio (I2D/IG) of graphene samples grown on commercially available SiO2 (100 nm) substrate was found to be 0.10, 0.40, and 1.70 at three different substrate temperatures of 700 °C, 750 °C, and 800 °C respectively, which correspond to bi- or trilayer graphene [30]. On the other hand, the I2D/IG ratio of graphene samples grown on thermally grown SiO2 (400 nm) substrate was found to be 0.02, 0.32, and 0.43 at substrate temperature of 700 °C, 750 °C, and 800 °C respectively, which are corresponded to few-layer graphene [30]. In comparison, the number of graphene layers seems to be quite lower on SiO2 (100 nm) substrate as compared to SiO2 (400 nm) substrate at 800 °C. Also, the quality of graphene films rapidly improves on commercially available SiO2 (100 nm) substrates as compared to thermally grown SiO2 (400 nm) substrates.

Furthermore, the full width at half maximum (FWHM) of 2D-band increases with the number of graphene layers, which is also used to determine the number of graphene layer [39]. In the present study, the FWHM of 2D-peak for the graphene sample grown on commercially available SiO2 substrate and thermally grown SiO2 substrate at higher temperature of 800 °C was found to be 75 cm⁻¹ and 90 cm⁻¹ respectively, which are very close to 74 cm⁻¹, previously reported for 3–4 layer of graphene grown on SiC [40]. From the peak intensity ratio and FWHM, it is very much clear indication that film grown on Ni coated SiO2 substrate (commercially available and thermally grown) are bi- or tri-layer graphene. Therefore, it is well evident from the Raman spectra that the number of graphene layer is almost different on both SiO2 substrates at higher temperature of 800 °C while graphene layer seems to be almost similar on both SiO2 substrates at lower temperature of 750 °C. However, defect density of graphene film, as calculated from D-peak to G-peak intensity ratio (ID/IG), rapidly reduced on commercially SiO2 (100 nm) substrate as compared to thermally grown SiO2 (400 nm), as shown in figures 2(a)–(b) and table 2. The intensity ratio ID/IG is also widely used to measure the crystallite size. The crystallite size (Lc) of graphene can be obtained from empirical formula by Cancado’s formula [41]:

\[ (L_c \text{ nm}) = \left( 2.4 \times 10^{-10} \right) \text{X} \left( \frac{\text{ID}}{\text{IG}} \right)^{-1} \]

The plot of crystallite size as a function of growth temperatures is shown figures 3(a)–(b). It is obvious from table 2 and figures 3(a)–(b) that crystallite size increases rapidly on commercially available SiO2 substrate while gradually increases on thermally grown SiO2 substrates upon increasing substrate temperature from 700 °C to 800 °C, which is again well evident that crystallinity increases with the increment substrate temperature.

The surface morphology of graphene films grown on two different thicknesses of SiO2 substrates with the help of Ni film was investigated using field emission scanning electron microscope. Figures 4(a)–(c) shows SEM micrographs of graphene film grown on commercially available SiO2 (100 nm)/Si substrates at three different substrate temperature of 700 °C, 750 °C, and 800 °C, while figures 4(d)–(f) shows SEM micrographs of graphene...
film grown on thermally grown SiO₂ (400 nm)/Si substrates at growth temperature of 700 °C, 750 °C, and 800 °C. At the substrate temperature of 700 °C, from the SEM micrograph of figure 4(a), the graphene layers with too much wrinkles were seen on commercially available SiO₂ (100 nm) substrates. As growth temperature increases, the number of wrinkles is reduced in graphene films. At the higher substrate temperature of 800 °C, it was noticed from the SEM images that the graphene sheet seems to be transparent with wrinkle free over SiO₂ (100 nm)/Si substrate. Thus, it is clear from the SEM micrograph that quality of graphene films continuously...
improves on commercially available SiO$_2$ (100 nm)/Si substrates. On the other hand, it was observed from SEM micrographs that graphene layers seem to be transparent on the thermally grown SiO$_2$ (400 nm)/Si substrates at lower substrate temperature of 700 °C. At substrate temperature of 750 °C, graphene films seem to be more transparent, but graphene layers becomes foldable with more wrinkles at higher temperature of 800 °C that makes more defects in graphene film. Therefore, graphene films become more defective on thermally grown SiO$_2$ (400 nm) substrates at higher temperature of 800 °C.

X-ray photoelectron spectroscopy (XPS) technique was used to investigate the C–C $sp^2$ bonding, and crystallinity in graphene films. Figures 5(a)–(c) shows analyzed XPS spectra of graphene samples as synthesized on commercially available 100 nm thick SiO$_2$ layer covered Si with the help of Ni film at three different substrate temperatures of 700 °C, 750 °C, and 800 °C while figures 5(d)–(f) shows XPS spectrum of graphene films deposited on thermally grown 400 thick SiO$_2$ over Si at substrate temperature of 700 °C, 750 °C, and 800 °C. The XPS spectra show the C1s peak of graphene film as the main peak appeared around ~284.5 eV [42] and two other shoulder peaks appeared around 285.6 eV and 288.4 eV, corresponding to C–O and C = O respectively [43, 44]. The existence of higher intensive peak at binding energy of 284.5 eV probably indicates C–C $sp^2$ bonding in graphene film [42], while oxygen functional group appeared either from the structural disorder of graphene layers or environment conditions [43, 45]. From the XPS results as shown in figures 5(a)–(c), it was noticed that the C1s peak continuously becomes intensive strongly and its FWHM constantly decreases as well as the intensity originated from the oxygen functional group gradually disappeared and slightly shifted to lower binding energy with the increment in substrate temperature from 700 °C to 800 °C. This observation clearly indicates an improvement in crystallinity as well as appearance of stronger C–C $sp^2$ bonding of graphene films [19, 43]. On the other hand, it was observed from figures 5(d)–(f) that the C1s peak becomes slightly weaker intensive due to formation of oxygen functional group and its FWHM increases as well as the intensity emerged from the oxygen functional group becomes intensive with the increase substrate temperature from 700 to 800 °C. This clearly shows reduction in quality and crystallinity of graphene films. The reason behind this phenomenon is that the surface of commercially available of SiO$_2$ (100 nm) seems to be smooth and superior quality as compared to thermally grown of SiO$_2$ (400 nm), which produce intensive peak of oxygen functional group and weaker C1s peak. Therefore, the buffer layer of SiO$_2$ thickness along with substrate temperature also affects the quality, growth, and crystallinity of graphene films. Thus, these XPS results are very good in agreement with the micro-Raman spectroscopy and SEM results.

Figure 5. (a)–(c) XPS analysis of PLD-derived graphene samples on Ni (≈250 nm)/SiO$_2$(100 nm)/Si substrate and (d)–(f) a XPS spectrum of graphene samples grown on Ni (≈250 nm)/SiO$_2$(400 nm)/Si substrate, at three different substrate temperature.
4. Conclusion

In summary, few layer graphene were successfully obtained on two different levels of thickness of SiO$_2$ on SiO$_2$/Si substrates, with the help of Ni film and from highly ordered pyrolytic graphite using pulsed laser deposition. Raman spectroscopic results clearly show that the buffer layers of SiO$_2$ play a significant role in graphene growth. As a result, a preferentially high-quality graphene film was formed on commercially available 100 nm thick SiO$_2$ layer as compared to thermally grown 400 nm thick SiO$_2$ layer. Moreover, it was also pointed out from the Raman spectra that substrate temperature also affects the growth, quality, number of graphene layers, and crystallite size of graphene sheet. Furthermore, micro-Raman spectroscopy results reveal that quality and crystallite size rapidly improve on commercially available SiO$_2$ (100 nm)/Si substrate while crystallite size gradually increases on thermally grown SiO$_2$ (400 nm)/Si substrate with the increment in substrate temperature from 700 °C to 800 °C. The XPS results also verified that crystallinity of graphene film increases and C–C bond becomes stronger with the increase in substrate temperature from 700 to 800 °C. Thus, our experimental results showed that high growth temperature is required to deposit graphene films with smaller number of graphene layers and lower amount of defects.

Acknowledgments

Pramod Kumar would like to gratefully acknowledge Ministry of Human Resource Development (MHRD), Government of India, for providing the research fellowship. IL would like to acknowledge partial financial support by Science and Engineering Research Board (SERB), India through grant no. EMR/2016.001282. The support from Dr Sanjeev Manhas and Mr Narendra Kumar (Research Scholar), Microelectronics Lab, Department of Electronics and Communication Engineering, IIT Roorkee acknowledged for their help in sputtering system and Dr M K Maurya (Head of Department, Department of Chemistry), IIT Roorkee for performing Raman experiment.

ORCID iDs

Pramod Kumar @ https://orcid.org/0000-0001-7351-7033
Indranil Lahiri @ https://orcid.org/0000-0002-5301-0072
Anirban Mitra @ https://orcid.org/0000-0003-3678-177X

References

[1] Geim A K and Novoselov K S 2007 The rise of graphene Nat. Mater. 6 183–91
[2] Geim A K 2009 Graphene: Status and Prospects Science 324 1530–1534
[3] Balandin A A, Ghosh S, Bao W, Calizo I, Tezel P, et al 2008 Superior thermal conductivity of single-layer graphene Nano Lett. 8 902–7
[4] Bolotin K I, Sikes K J, Jiang Z, Klima M, Fudenberg G, Hone J, Kim P and Stormer H L 2008 Ultrahigh electron mobility in suspended graphene Solid State Commun. 146 351–5
[5] Nair R R, Blake P, Grigorenko A N, Novoselov K S, Booth T J, Stauber T, Peres N M R and Geim A K 2008 Fine structure constant defined via transparency of graphene Science 320 1308
[6] Lee C, Wei X, Kysar J W and Hone J 2008 Measurement of the elastic properties and intrinsic strength of monolayer graphene Science 321 385–8
[7] Kim K S, Zhao Y, Jang H, Lee S Y, Kim J M, Kim K S, Ahn J H, Kim P, Choi J Y and Hong B H 2009 Large-scale pattern growth of graphene films for stretchable transparent electrodes Nature 457 706–10
[8] Lin Y M, Dimitrakopoulos C, Jenkins K A, Farmer D B, Chiu H Y, Grill A and Avouris P 2010 100-GHz transistors from wafer-scale epitaxial graphene Science 327 662
[9] Miao X, Tongay S, Pettersson M K, Berke K, Rinzel G A, Appleton B R and Hebard A F 2012 High efficiency graphene solar cells by chemical doping Nano Lett. 12 2745–50
[10] Tian H, Shu Y, Wang X F, Mohammad M A, Bie Z, Xie Q Y, Li C, Mi W T, Yang Y and Ren T L 2015 A graphene-based resistive pressure sensor with record-high sensitivity in a wide pressure range Sci. Rep. 5 8603–8
[11] Liu C, Yu Z, Neff D, Zhan A A and Jang B Z 2010 Graphene-based supercapacitor with an ultrahigh energy density Nano Lett. 10 4863–8
[12] Wang G, Shen X, Yao J and Park J 2009 Graphene nanosheets for enhanced lithium storage in lithium ion batteries Carbon 47 2049–53
[13] Novoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V and Firsov A A 2004 Electric field effect in atomically thin carbon films Science 306 666–9
[14] Huang H, Chen W, Chen S, Thye A and Wee S 2008 Bottom-up growth of epitaxial graphene on 6H-SiC (0001) ACS Nano. 2 2513–8
[15] Winterlin J and Boquet M L 2009 Graphene on metal surfaces Surf. Sci. 603 1841–52
[16] Reina A, Jia X, Ho J, Neizich D, Son H, Bulovic V, Dresselhaus M S and Kong J 2009 Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition Nano Lett. 9 930–5
[17] Li X et al 2009 Large-area synthesis of high-quality and uniform graphene films on copper foils Science 324 1312–4
[18] Wang S, Santos L F D, Wurstbauer U, Wang L, Pfeiffer L N, Hone J, García J M and Pinczuk A 2014 Single- and bi-layer graphene grown on sapphire by molecular beam epitaxy Solid State Commun. 189 15–20
Mater. Res. Express 6 (2019) 125625

19. Oliveira M H, Schumann T, Gargallo-Caballero R, Fromm E, Seyller T, Ramsteiner M, Trampert A, Geelhaar L, Lopes J M J and Riechert H 2013 Mono- and few-layer nanocrystalline graphene grown on Al₂O₃(0 0 0 1) by molecular beam epitaxy Carbon 56 339–50

20. Moreau E, Godey S, Ferrer F J, Vignaud D, Wallart X, Avila J, Asensio M C, Bourtrel F and Gallet J J 2010 Graphene growth by molecular beam epitaxy on the carbon-face of SiC Appl. Phys. Lett. 97 241907–8

21. Pappas D L, Saenger K L, Bruley J, Krakow W, Cuomo J J, Gu T and Collins R W 1992 Pulsed laser deposition of diamond-like carbon films J. App. Phys. 71 5675–84

22. Akhsakhalyan A D, Bityurin Y A, Gaponov S V, Godkov A A and Luchin V I 1982 Processes occurring in an erosion plasma during laser vacuum deposition of films. I. Properties of a laser erosion plasma in the inertial-expansion stage Sov Phys. Tech. Phys. 27 969–973

23. Koh A T T, Foong Y M and Chua D H C 2012 Comparison of the mechanism of low defect few-layer graphene fabricated on different metals by pulsed laser deposition Diam. Relat. Mater. 25 98–102

24. Petersen J and Mayer S G 2008 Dewetting of Ni and NiAg solid thin films and formation of nanowires on ripple patterned substrates J. App. Phys. 103 023520–8

25. Luber E J, Olsen B C, Ophus C and Mitlin D 2010 Solid-state dewetting mechanisms of ultrathin Ni films revealed by combining in situ time resolved differential reflectometry monitoring and atomic force microscopy Phys. Rev. B 82 085407–16

26. Gong Y et al 2012 Layer-controlled and wafer-scale synthesis of uniform and high-quality graphene films on a polycrystalline nickel catalyst Adv. Funct. Mater. 22 3153–9

27. Delamaro A, Rabot C, Vallee C and Zenasni A 2014 Wafer scale catalytic growth of graphene on nickel by solid carbon source Carbon 66 48–56

28. Saiz E, Cannon R M and Tomson A P 2008 High-temperature wetting and the work of adhesion in metal/oxide systems Annu. Rev. Mater. Res. 38 197–226

29. Abd Elhamid A M, Aboufotouh A M, Hafez M A and Azzouz I M 2017 Room temperature graphene growth on complex metal matrix by PDL Diam. Relat. Mater. 80 162–7

30. Wang K, Tai G, Wong K H, Lau S P and Guo W 2011 Ni induced few-layer graphene growth at low temperature by pulsed laser deposition AIP Adv. 1 022141–9

31. Kumar P, Kanaujia P K, Vijaya Prakash G, Dewasi A, Lahiri I and Mitra A 2017 Growth of few- and multilayer graphene on different substrates using pulsed nanosecond Q-switched Nd:YAG laser J. Mater. Sci. 52 12295–306

32. Kumar I and Khare A 2016 Optical nonlinearity in nanostructured carbon thin films fabricated by pulsed laser deposition technique Thin Solid Films 611 56–61

33. Ferrari A C and Basko D M 2013 Raman spectroscopy as a versatile tool for studying the properties of graphene Nat. Nanotechnol. 8 235–46

34. Ferrari A C 2007 Raman spectroscopy of graphene and graphite: disorder, electron-phonon coupling, doping and nonadiabatic effects Solid State Commun. 143 47–57

35. Dresselhaus M S, Jorio A, Hofmann M, Dresselhaus G and Saito R 2010 Perspectives on carbon nanotubes and graphene Raman spectroscopy Nano Lett. 10 751–8

36. Malard L M, Pimenta M A, Dresselhaus G and Dresselhaus M S 2009 Raman spectroscopy in graphene Phys. Rep. 473 51–87

37. Ferrari A C et al 2006 Raman spectrum of graphene and graphene layers Phys. Rev. Lett. 97 187401–4

38. Koh A T T, Foong Y M and Chua D H C 2010 Cooling rate and energy dependence of pulsed laser fabricated graphene on nickel at reduced temperature Appl. Phys. Lett. 97 114102–4

39. Karamat S, Sonu A, Čelik Y, Uysalı, Özgönlü E and Oral A 2015 Synthesis of few layer single crystal graphene grains on platinum by chemical vapour deposition Prog. Nat. Sci. Mater. Int. 25 291–9

40. Lee D S, Biedl C, Krauss B, Von Klitzing K, Gunte U and Smet J H 2008 Raman spectra of epitaxial graphene on SiC. and of epitaxial graphene transferred to SiO₂ Nano Lett. 8 4320–5

41. Cançado L G, Takai K, Enoki T, Endo M, Kim Y A, Mizusaki H, Jorio A, Coelho L N, Magalhães-Paniago R and Pimenta M A 2006 General equation for the determination of the crystallite size Lₕ of nanographite by Raman spectroscopy App. phys. Lett. 88 163106–8

42. Kovács G J, Bertóti I and Radnóczi G 2008 X-ray photoelectron spectroscopic study of magnetron sputtered carbon–nickel composite films Thin Solid Films 516 7942–6

43. Kwak J H, Lee S S, Lee H J, Ainoop G, Lee H J, Kim W S, Ryu S W, Kim H S and Jo J Y 2016 Direct growth of nano-crystalline graphite films using pulsed laser deposition with in situ monitoring based on reflection high-energy electron diffraction technique App. Phys. Lett. 108 123107–11

44. Bhauumik A and Narayan J 2016 Wafer scale integration of reduced graphene oxide by novel laser processing at room temperature in air J. App. Phys. 120 105304–16

45. Mortazavi S Z, Parvin P and Reyhani A 2012 Fabrication of graphene based on Q-switched Nd:YAG laser ablation of graphite target in liquid nitrogen Laser Phys. Lett. 9 547–52