Peer-reviewed version available at Materials 2018, 11, 1120; doi:10.3390/ma11071120

Article

A Novel Method of Synthesizing Graphene for Electronic Device Applications

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Abstract: This article reports a novel and efficient method to synthesize graphene by thermal decomposition process. In this method, silicon carbide (SiC) thin films grown on Si(100) wafers with an AlN buffer layer were used as substrates. A CO_2 laser beam heating without vacuum or controlled atmosphere was applied for SiC thermal decomposition. The physical, chemical, morphological, and electrical properties of the laser-produced graphene were investigated for different laser energy densities. The results demonstrate that graphene was produced in form of small islands with quality, density and properties depending on the applied laser energy density. Furthermore, the produced graphene exhibits a sheet resistance characteristic similar to graphene grown on mono-crystalline SiC wafer, which indicates its potential for electronic device applications.

Keywords: graphene synthesis; silicon carbide; thin film; high-power impulse magnetron sputtering; thermal decomposition, electronic devices.

1. Introduction

Nowadays, the synthesis of high-quality graphene has been the focus of several researches due to great potential applications of this material, as for example in electronic devices, sensors and flexible displays [1]. Among the graphene synthesis methods, the thermal decomposition processes have been successful used to grow graphene layers on silicon carbide (SiC) [2-5]. In general, these studies show the use of SiC wafers as substrates to be decomposed by heating using an induction furnace at vacuum or at atmospheric pressure with an inert gas flow [6-10]. The kinetics of graphene formation and properties, such as structure and morphology, show to be dependent on the reactor pressure, type of gas atmosphere, orientation and face termination of the SiC wafer [10-12].

Most recently, the use of a laser beam as heating source for graphene formation from SiC has been reported [13-15]. The focus of these studies was to investigate the growth of graphene on Si- and/or C-face of SiC wafers termination. Perrone et al. used a near infrared Nd:YVO_4 (1064 nm) laser to promote the heating of SiC surface [13]. They reported a possible presence of graphene when process was performed using an argon flow or vacuum at a pressure of 10^{-5} Torr. Using ultra-high vacuum (UHV), only a disordered graphite phase was observed [13]. Lee et al. using an UV laser (248 nm) noticed not only that it is possible to grow epitaxial graphene (EG) from Si-terminated SiC (0001),

as pointed out that EG has a structure comparable to thermally grown graphene in UHV using the same substrate [14]. Unlike the previous two works, Yannopoulos et al. obtained graphene on SiC without the use of vacuum environment or pre-treatment of SiC substrate [15]. In their work, a carbon dioxide (CO_2) laser beam was used as the heating source and the argon gas flow at atmospheric pressure was applied to form few layers of EG. An advantage of the use of CO_2 laser is the cooling effect during pulse and the possibility of writing graphene patterns on SiC, which eliminates the lithographic step [15]. Nevertheless, the aforementioned studies and processes have a serious drawback that is the use of high-cost SiC wafers as substrate [16, 17].

In other recent study, Galvão et al. reported on the growth of graphene layers on a low cost polycrystalline SiC substrate obtained from powder metallurgy using a CO_2 laser beam [18]. Although the graphene obtained may be applied in several areas, the SiC substrate is not ideal for microelectronic applications. In order to improve the quality of the graphene samples without significant increase in the production costs, we explore the use of SiC thin films as substrates in combination with the CO_2 laser beam heating technique for EG growth. Herein, SiC thin films were grown by high-power impulse magnetron sputtering (HiPIMS) on silicon substrates covered with an aluminum nitride (AlN) buffer layer. To the best of our knowledge, the formation of graphene from SiC thin films grown on AlN/Si substrates using CO_2 laser beam has not yet been reported in literature [19]. In this work, different levels of laser energy density were applied during SiC thermal decomposition and the chemical properties and quality of the graphene were evaluated using Raman spectroscopy. Moreover, to further understand the material characteristics, morphological and electrical properties of the samples were investigated using AFM and four points probe method, respectively.

2. Materials and Methods

2.1. SiC thin film growth

SiC thin films were deposited by HiPIMS technique on pieces of polished p-type Si(100) wafer covered with AlN buffer in a high-vacuum chamber with a background pressure of 6×10^{-6} Torr. More details of the magnetron sputtering system can be found elsewhere [19]. The working pressure of the argon gas (99.999%) was maintained at 3×10^{-3} Torr for a corresponding flow rate of 20 sccm. A high-purity SiC (99.5%, Kurt J. Lesker) target with a 4-inch diameter was used. The sputtering reactor was powered by a HiPIMS power supply (Solvix HIP^3 5kW) with an applied power of 200 W and duty cycle of 5%. The target-to-substrate distance was approximately 65 mm and the deposition was performed during 10 min. The substrate holder was in a floating potential. Before starting the SiC deposition, a pre-sputtering period (10 min, at 200 W) was performed to remove the contamination from the target surface. The obtained SiC film on AlN/Si substrate had an average thickness around 240 nm.

The AlN buffer layer was grown on the Si(100) substrates using the HiPIMS technique at "Institut des Matériaux Jean Rouxel in Nantes University". More details can be found elsewhere [20, 21]. The AlN film on Si substrate had an average thickness around 1300 nm and the main crystallographic orientation was (002).

2.2. SiC sublimation by CO_2 laser heating

The heating of the SiC thin film was carried out by using a CO_2 laser (Synrad Evolution – 125) with a beam diameter of 200 μm, which emits infrared laser radiation at a wavelength band of 10.6 μm. The samples were positioned at 5 mm below the laser focal point (see schematic diagram of the laser irradiation process in [18]). For all the processes, a beam overlap of 50% was used. Three different laser scanning velocity rates were used: 2300, 2500 and 2600 mm s^{-1}. For all conditions, the laser power applied was 50 W, which corresponded to 40% of the overall power. The entire process occurred under ambient atmosphere and pressure conditions. The energy densities of each scanning velocity were calculated and are shown in Table 1. One can note that in this investigation the energy densities were between 127-145 J cm^{-2}, whereas in the previous works the values were between 132-200 J cm^{-2} [18]. The choice of a lower energy density range is due to the properties of the synthesized

graphene at higher laser energy density did not present significant difference in comparison with samples produced with lower energy density [18].

Table 1. Scanning velocities and energy densities applied for each condition.

	Condition 1 (C1)	Condition 2 (C2)	Condition 3 (C3)
Scanning velocity (mm s^{-1})	2300	2500	2600
Energy density (J cm^{-2})	145.25	136.95	127.69

2.3. Material characterization

The structure of the as-deposited SiC films was investigated using Grazing Incidence XRD with an incidence angle (ω) of 0.3° operated with a PANalytical X'pert Pro x-ray diffractometer with CuKα radiation.

Raman analyses were performed using a Horiba Raman microprobe system equipped with an argon ion laser (514.5 nm). Raman spectra of the SiC and graphene samples were obtained at room temperature in the range of 200 to 1100 cm^{-1} and 1200 to 2900 cm^{-1}, respectively. The Raman spectral imaging or Raman mapping were obtained focusing on the main characteristic bands (or peaks) of graphene. The type of the defects presented in the graphene film was inferred according to Eckmann et al. [22], which reported on a relation between the intensity (height) of the ID and ID' peaks with the type of defect present in the graphene sample. The maximum ID/ID' ratio (~13) would correspond to sp^3-defects, (~7) for vacancy-like defects and, (~3.5) for boundaries defects [22]. In this work, the D' peak is merged with the G peak. To determine the intensity of D' peak, a Lorentzian double peaks function fitting was applied to each Raman spectrum.

The surface morphology was investigated by atomic force microscopy (AFM Veeco Multimode with Nanoscope V control station). The tapping mode was used for all samples except for sample C2, where contact mode was applied because it provided images with better resolutions.

3. Results and Discussion

3.1. SiC thin film structure

In the literature, there is only one report about the SiC growth on Si substrate using the HiPIMS technique [23]. However, due to the large mismatch between SiC and Si (~ 20%), the grown film exhibited an amorphous characteristic and a high residual stress [23, 24]. To reduce these effects, several studies have demonstrated that using a sacrificial layer (buffer layer) on Si substrate before the deposition of SiC is an effective alternative. In this work, we chose to use the AlN buffer, which presents a mismatching in the lattice constant of less than 1% comparing to SiC. Figure 1 shows the GIXRD spectrum of the SiC thin film grown on AlN/Si(100) substrate. As expected, the identified peaks reveal the polycrystalline nature of the SiC.

In addition, the orientations of SiC indicated in GIXRD spectrum is in agreement with some studies in the literature [25-27]. We found that the peaks of the SiC matched well with those of α-SiC (6H-SiC), however, it can be not excluded that the existence of β-SiC (3C-SiC).

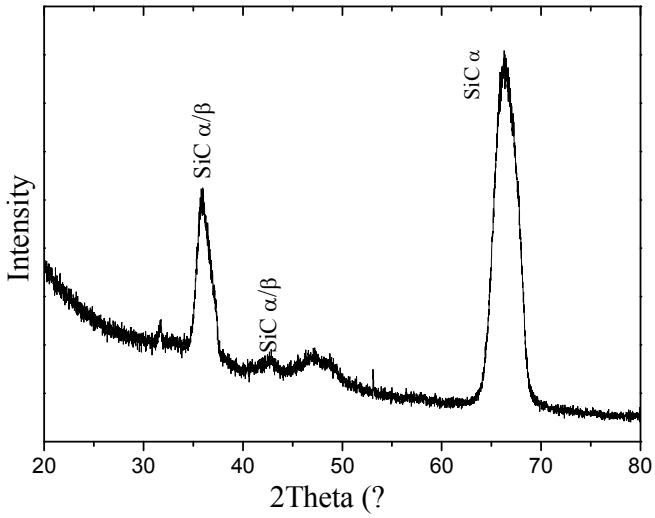

Figure 1. Grazing incidence XRD spectrum of SiC film grown on AlN/Si(100) substrate.

3.2. Graphene characterization

Raman spectroscopy is a very useful technique to obtain important information about inherent features of carbon materials, such as graphene [28-31]. The Raman spectra of the graphene contain three main in-plane vibrational bands: (i) G-band (~1584 cm^{-1}), corresponding to the doubly degenerate E_{2g} phonon mode at the Brillouin zone center; (ii) D-band (1200-1400 cm^{-1}) that arises from TO phonons around the K point and requires a defect for its activation; and (ii) 2D-band (2400-2800 cm^{-1}), that is the second order of the D-band and has been widely used to evaluate the number of layers and structural quality of graphene [28, 32].

Figure 2 shows the Raman spectra of each SiC sample that was laser treated. In addition, the Raman spectra of the unexposed SiC area are also presented. The presence of graphene G and 2D-bands and a significant amount of defects (D-band) is observed (Figure 2b). Conversely, we notice the absence of peaks related to the Si-C band (Figure 2a). The absence of the Si-C bands can be an indication that, at this point, all the SiC was decomposed to form graphene. To verify the growth behavior and distribution of graphene in the treated sample, a Raman mapping was performed (Figures 3 and 4).

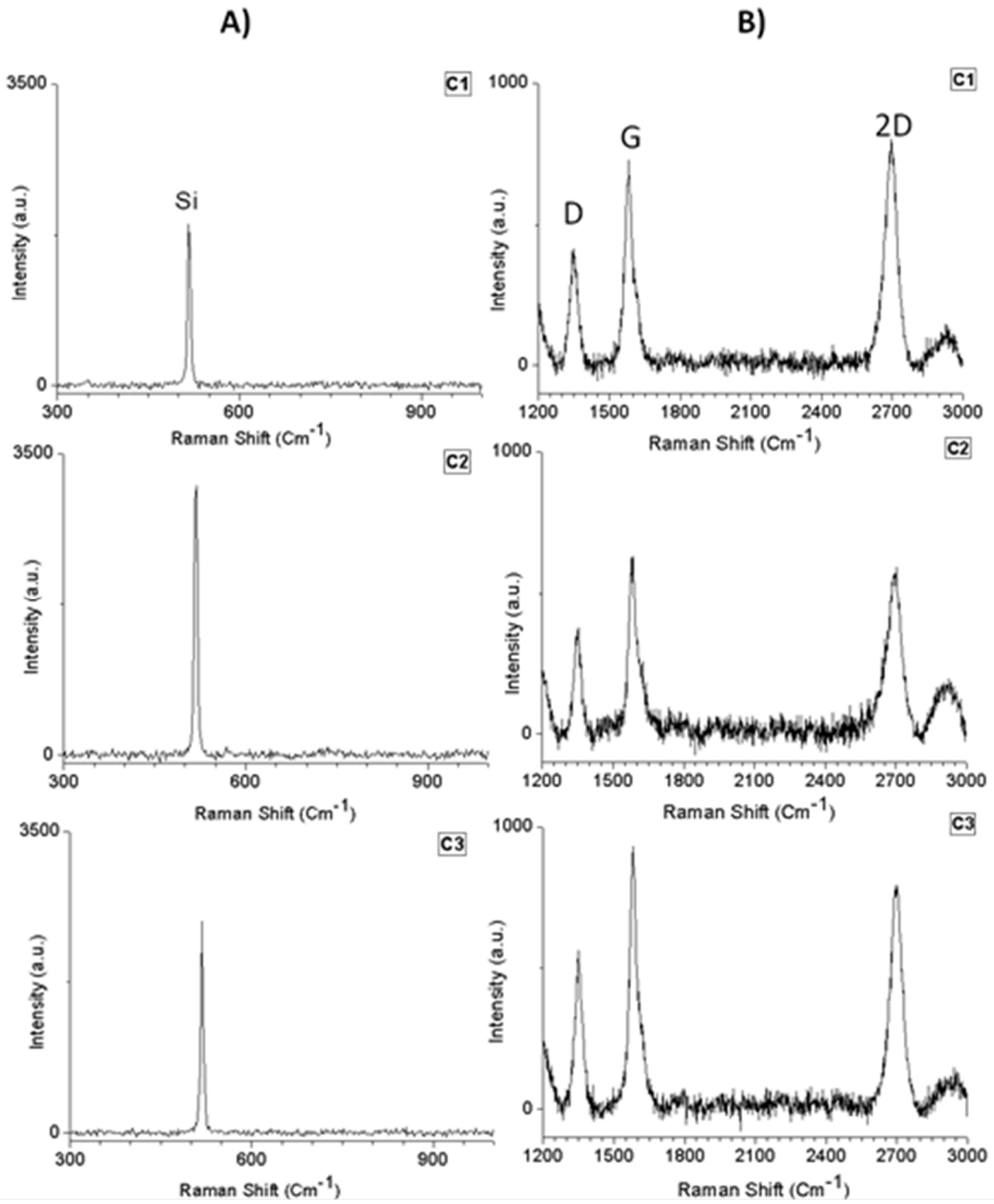

Figure 2. Raman spectra of the samples C1, C2 and C3: (a) scanning in the SiC area (column A); (b) scanning in the graphene area (column B).

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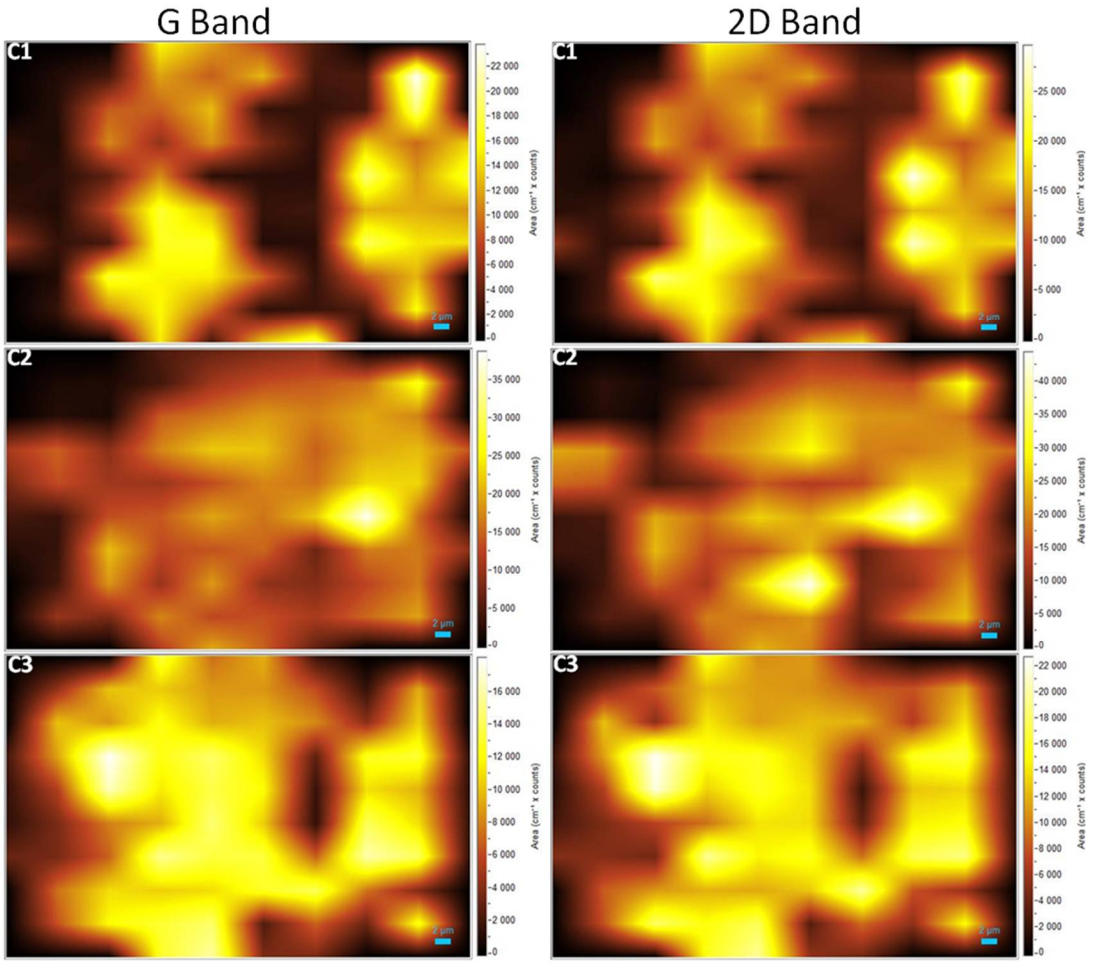

Figure 3. Raman mapping of the samples corresponding to G and 2D bands.

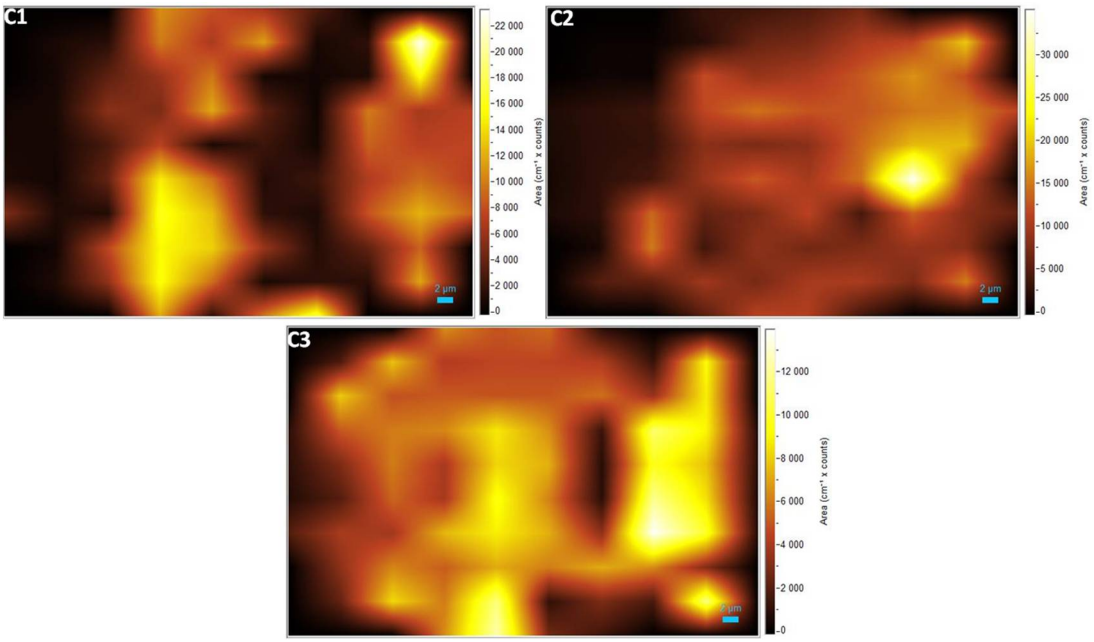

Figure 4. Raman mapping of the samples corresponding to D band.

The bright spots observed in the Raman maps (Figures 3 and 4), which are due to the presence of bands related to graphene, are in agreement with the Raman spectra shown in Figure 2. The dark

areas correspond to the absence of bands related to graphene. The mapping images also reveal that graphene did not expand enough to cover the entire region of analysis, forming some "graphene islands". Furthermore, it is possible to observe a non-uniform distribution of higher intensity regions. This variation is probably related to the quality of graphene. Several studies reported that the kinetics of graphene growth on Si- or C-face of SiC is distinct [3, 32, 33, 34]. When EG is grown on the Si-face in the UHV environment, the rate of sublimation is reduced and therefore it is possible to control the growth of graphene layers. This leads to a large and homogeneous monolayer [36, 37].

When the C-face is considered, the growth rate is higher, and its control is more difficult, which usually results in an inhomogeneous graphene [3, 33, 34]. However, Hass et al. reported that graphene grown on C-face of the SiC substrate, in a RF furnace, can present an exceptional quality [37]. This indicates that depending on the technique it is possible to obtain good quality graphene on both faces. In our studies, graphene was grown on SiC thin film with undefined face-termination (Si–C face). Currently, there is a clear lack of studies reporting on the kinetics of graphene growth on SiC thin films using thermal decomposition by CO_2 laser heating. Thus, according to the behavior observed in Raman mapping and considering that the SiC film is polycrystalline and contains some amorphous areas, it is possible to presume that graphene was grown from both Si–C face terminations. These two possible faces growth can result in graphene regions with high defect concentration and different layers. In addition, the laser heating may also have influenced the growth and quality of graphene. As obtained by Galvão et al. [18], the limited growth of graphene and non-dissociation of SiC in some regions of the material may have been influenced by the heat transfer along and across the heterogeneous surface. Inhomogenities on the surface cause a non-uniform temperature distribution. A detailed Raman analysis performed on sample C2 revealed that the dark areas on the maps are composed by SiC films that did not get enough energy to dissociate and form graphene; however, crystallization started to take part instead, which can be verified by the well-defined SiC peak showed in Figure 5.

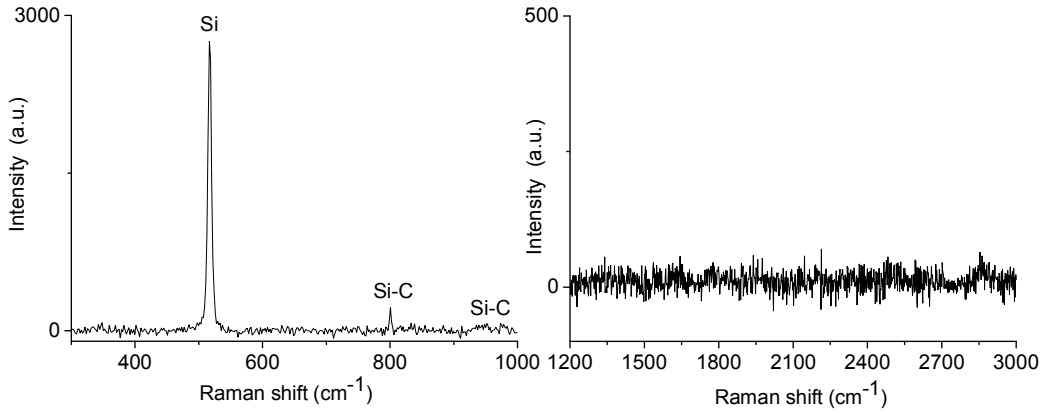

Figure 5. Raman spectra of the dark areas for C2 sample: (a) scanning in the SiC range (column A); (b) scanning in the Graphene range (column B).

3.3. Sheet resistance and morphology of SiC thin film and graphene layers

As graphene is a material of great interest for electronic device applications, we have also analyzed the sheet resistance for each sample (Table 2) produced by the developed method. In all conditions, the sheet resistance is lower than the sheet resistance of SiC, but only C2 sample presents properties consistent with graphene. The high sheet resistance presented by C1 and C3 may have occurred because the "islands" of graphene produced are small and not interconnected. Both Raman maps and AFM surface morphology images can endorse this fact.

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Table 2. Sheet resistance of SiC thin film and graphene inferred by the four points probe method.

	C1	C2	C3	SiC[1]
Sheet resistance (Ω/□)	30900	26	29320	60000

[1] Reference measurement made on SiC film substrate.

Before analyzing the surface morphology of the treated samples, it is important to verify the morphology of the SiC surface. Figure 6 shows the AFM images of the surface of the SiC films and the AlN buffer layer before laser treatment. As can be noticed, the SiC film follows the morphology of the AlN layer with grains than 100 nm. These results indicate high quality SiC and AlN thin films.

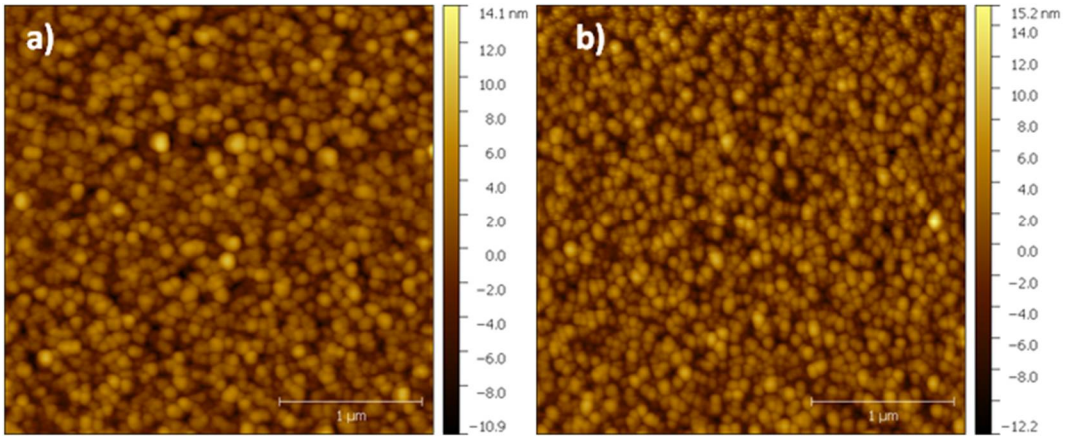

Figure 6. Atomic force microscopy images of SiC (a) and AlN (b) films.

Figure 7 shows the surface morphology of the samples C1, C2 and C3. For samples C1 and C3 (Figures 7a and 7b), it is only possible to observe small isolated graphene islands. On the other hand, for the sample C2 (Figures 7c and 7d), the graphene layers are larger and well distributed on surface area in comparison with C1 and C3. The growth of the "islands" can be visualized in the profile of Figure 7d (sample C2), where stacking of the multilayers can be easier perceived. This result indicates that even a small difference of the energy density of the CO2 laser has a strong influence on the quality of graphene grown on SiC film, which allows for the control of up to 3 orders of magnitude of material resistivity.

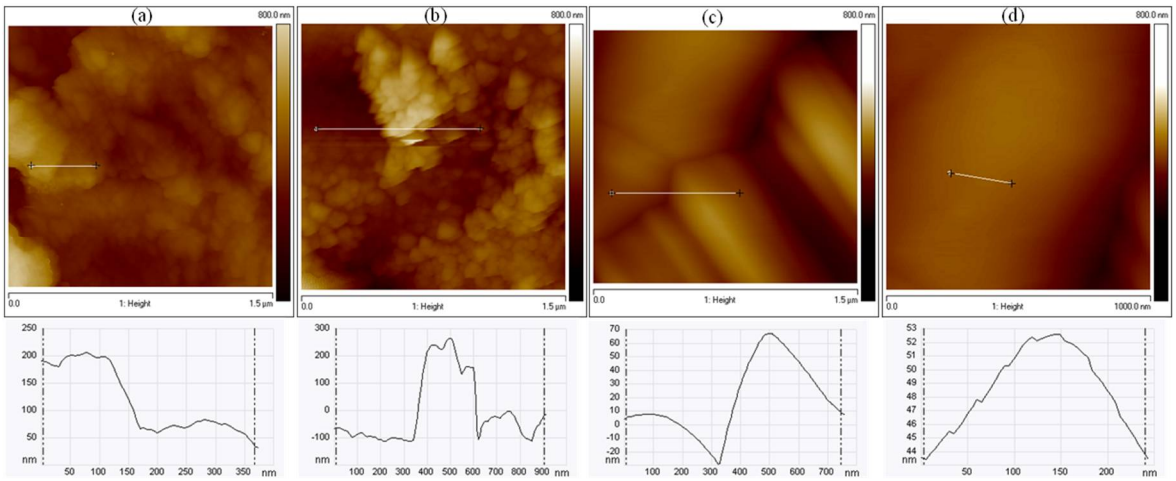

Figure 7. Atomic force microscopy images of samples C1 (a); C3 (b); and C2 (c, d). White line = profile traced to check the stacking of islands whose result is shown in the chart below each image.

5. Conclusions

Herein, for the first time, we approached a feasible route for graphene growing based on the thermal decomposition of polycrystalline SiC thin films deposited by HiPIMS technique on AlN/Si substrates. For this purpose, we used a CO_2 laser beam heating without vacuum or controlled atmosphere. Raman mapping along with AFM measurements revealed the formation of islands of graphene on the SiC surfaces. The quality and density of graphene islands showed to be strongly dependent on the energy density of laser process. It was observed that an energy density of the order of 137 J cm^{-2} allowed for the obtaining of graphene layers interconnected but with some defects along the surface, resulting in a sheet resistance characteristic of graphene grown on pure crystal SiC wafer. Conversely, when the energy density was increased to 145 J cm^{-2}, the density of defects was considerably reduced. Finally, the results of this work demonstrate the feasibility of using the laser beam technique as a heating source for graphene formation from SiC thin films, as well as give rise to new possibilities to explore the development of graphene layers on different substrates using the same methodology proposed here.

Author Contributions: N. G., G. V., R. P., J. M., M. G. and J. C. provide the investigation and methodology, N. G., R. P., M. F., B. R., M. D. and H. M. white the original draft.

Funding: This work was supported by the Brazilian funding agencies: Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES), grants No. 23038.005802/2014-98 and No. 88881.064970/2014-01, Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), grants No 473043/2012 and 301982/2015-5, and Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP), grants No. 2011/07468-2, 2018/01265-1 and (FAPESP/CNPq/PRONEX) 2011/50773-0.

Acknowledgments: We gratefully acknowledge the Associate Laboratory of Sensors and Materials, Instituto Nacional de Pesquisas Espaciais (INPE), for Raman measurements.

Conflicts of Interest: The authors declare no conflict of interest.

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