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To cite this article: A V Avramchuk *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **475** 012036

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Local laser annealing of 3C-SiC film deposited on the silicon substrate by CVD

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Abstract. In this work, we try to suite an approach, which concerns of epitaxial graphene growth by laser irradiation of 3C-SiC (111) film deposited on silicon substrate (111) by chemical vapor deposition method. Laser treatment was performed by pulsed 1064 nm laser with 20 Hz repetition rate and 15 ns pulse duration, the fluency was varied 0-1.5 J/cm². Raman spectroscopy studies show that for fluence above 0.8 J/cm² 2D band is noticeable revealing formation of high crystallographic quality graphitic (graphene) film.

1. Introduction

Despite over decade history of graphene intensive study the problem of its integration in electronics remains open. One of the issues which limit the applicability of graphene in modern electronics is the lack of the reliable technology to produce cheap and wafer- scale graphene which electronic properties allow to replace the existing on the market materials. Among the variety of the methods for graphene production CVD and epitaxial growth of graphene on SiC surface are the methods where the most progress has been achieved. Indeed, single-crystals of graphene grown by CVD method possess high charge carrier mobility (up to 16000 cm²B⁻¹c⁻¹) [1, 2, 4], compatible with mechanically exfoliated graphene. Moreover, up to cm scale of graphene single-crystal can be grown by CVD [3-4]. However, the need for transfer graphene on target substrate from catalyst surface and problems accompanying this process (wrinkles, charge transfer) [5-6] seriously affect the device performance. On another hand, epitaxial growth of graphene on silicon carbide through thermal sublimation Si from SiC provides direct graphene technology on the isolated substrate. Furthermore, epitaxial graphene grown on different polytypes of SiC substrate is commercially available. It is worth mentioning that except structural quality, the SiC substrates are expensive and size-limited. 3C-SiC film deposited on silicon can act as an alternative to the use of expensive single-crystal substrate of silicon carbide of 4H and 6H polytypes for epitaxial synthesis of graphene. However, because of relatively low melting temperature of silicon (~1400 °C) conventional annealing techniques may not provide optimal conditions for high quality graphene growth [7-8]. The laser annealing could be an alternative technique for ensuring a high local temperature, which could be sufficient to transform silicon carbide into graphene. The possibility of epitaxial growth of graphene by laser annealing on single crystal SiC surface already were reported. S Lee et al grew epitaxial graphene on SiC (0001) using UV laser (KrF



248 nm radiation, $\lambda=248$ nm, pulse length 25 ns) in HV conditions $\sim 10^{-6}$ Torr [9]. M. Hattori et al reported results of patterning of SiC (0001) with 248 nm excimer KrF-laser, pulse duration of 55 ns and repetition rate 100Hz [10]. Recently N K A M Galvão et al grew epitaxial graphene by 10.6 μm CO-laser annealing of polycrystalline SiC with 125 W power of laser radiation in atmospheric pressure. They showed that graphene growth as mosaic structure on grains of SiC [11].

In this work we report our structural investigation of 1064 nm pulsed YAG: Nd + laser annealed 3C-SiC film grown by on Si (111) substrate.

2. Sample fabrication

SiC film with ~ 300 nm thickness was grown in cold wall type CVD reactor. Before the growth process, the Si wafer was chemically treated, then annealed at 1100°C in H_2 in CVD chamber. Prior the deposition of silicon carbide, the substrate was pre-carbidized in a 5 cm^3/min C_2H_4 gas for 2 minutes. The silicon carbide film was grown at $T=1300$ °C using SiH_4 and C_2H_4 precursors for 2 minutes. The flow rate of each gas was 5 cm^3/min .

The local laser heating of silicon carbide films was carried out in a vacuum chamber at a pressure of 10^{-3} Pa in an Ar gas using 1064 nm YAG: Nd + laser. All experiments were carried out with pulse repetition frequency of 20 Hz, pulse duration of 15 ns. The fluence was varied from 0 to 1.5 J/cm^2 . The laser was normally focused on the surface of the film. Finally, 10000 shots were used for each experiment.

The crystalline structure of the silicon carbide film was studied by Ultima IV X-ray diffractometer ($\text{CuK}\alpha 1$ $\lambda = 0.154056$ nm, tube voltage 40 kV, current 40 mA). A parallel primary beam was applied to reduce the effect of surface roughness on the measurement results. Graphite monochromator was used on diffracted radiation to improve the signal-to-noise ratio. We investigate the structure of the films irradiated with different fluence using Raman spectroscopy (Nanofinder HE (LOTIS TII) spectrometer). The measurements were conducted in the backscattering geometry (spectral resolution of 3 cm^{-1}) with wavelength 473 nm and normal environmental conditions. The surface morphology of the grown films was evaluated by SEM (TESCAN LYRA3 SEM-FIB) and atomic force microscopy (NT-MDT Solver Open).

3. Results and discussion

The diffraction spectra, ω -2 θ scan, of 3C-SiC film deposited on Si substrate is shown in figure 1. Three characteristic peaks of 3C-SiC (111), (220) and (222) are visible. The ratio of reflections corresponding to 3C-SiC (111) and 3C-SiC (220) crystals indicates the growth of the (111) orientation of the predominantly single-crystal 3C-SiC film.

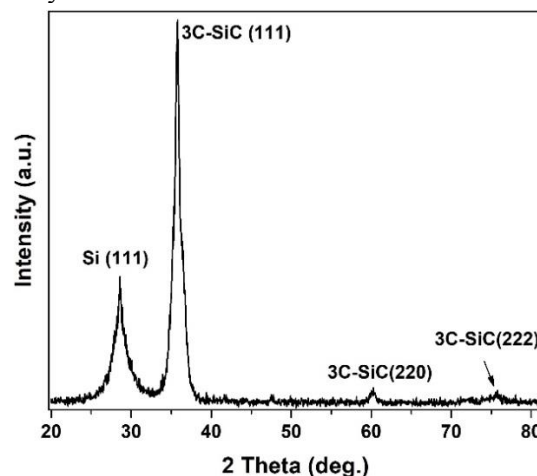


Figure 1. XRD spectra of 3C-SiC film on Si.

In figure 2 we collect the images of as deposited and annealed with 1 J/cm^2 fluence surfaces of 3C-SiC film, a and b respectively.

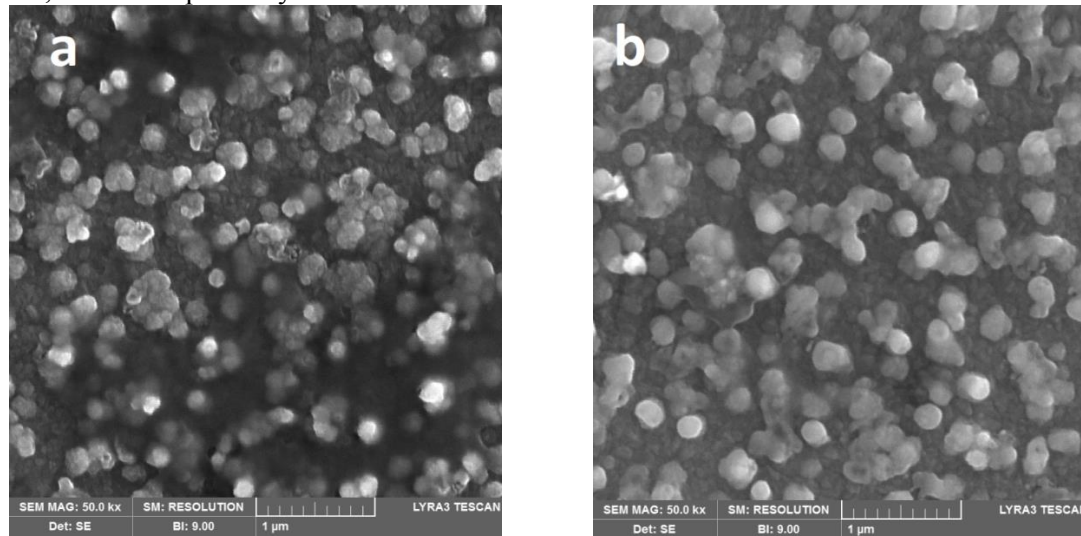


Figure 2. The SEM images of as deposited (a) and annealed with 1 J/cm^2 fluence (b) surfaces of 3C-SiC film.

No significant change in the surface morphology can be noticeable. However, our AFM studies reveal that RMS roughness is decreased from 40 nm for as deposited film to 30 nm for annealed film. This fact already demonstrates significant effect of laser annealing at used conditions on 3C-SiC film.

Our Raman studies (not present here) for the fluence in the $0 - 0.8 \text{ J/cm}^2$ range demonstrate only the changes in $1200 - 1700 \text{ cm}^{-1}$ domain, which may be associated with formation of disordered graphitic like structure. But for the fluences above 0.8 J/cm^2 an additional peak in $2700 - 2750 \text{ cm}^{-1}$ range associated with 2D band starts to be noticeable. We collect Raman spectra for 3C-SiC surface annealed with laser fluence above 0.85 J/cm^2 in figure 3. First, one may notice significant increasing of the intensity of the peak 1450 cm^{-1} . Because of the lack of additional structural information, we may only assume the relation of this peak to SiC phase. We believe that intensity increasing of this peak is caused by temperature induced structural changes in SiC. What would be rather in agreement with our assumption for high surface temperature induced by laser annealing. Another significant feature observable in the spectra is the change of the 2D band intensity. Moreover, for the highest fluence we used in our work, FWHM 2D is 31 cm^{-1} . This fact can be explained by formation of much more structurally ordered graphitic like structure on the top of disordered. Unfortunately, strong overlap of G bands ($\sim 1603 \text{ cm}^{-1}$) associated with disordered graphite structure and G band from ordered one does not allow to draw any solid conclusion based on G/2D ratio. However, symmetrical Lorentz shape of 2D line together with its position 2731 cm^{-1} support the fact that ordered part of the graphitic layer is fewlayered graphene with turbostratic structure [12]. Nevertheless, the conclusion we made from Raman investigation requires crosschecking by other structural techniques. We put our discussion here only as possible scenario.

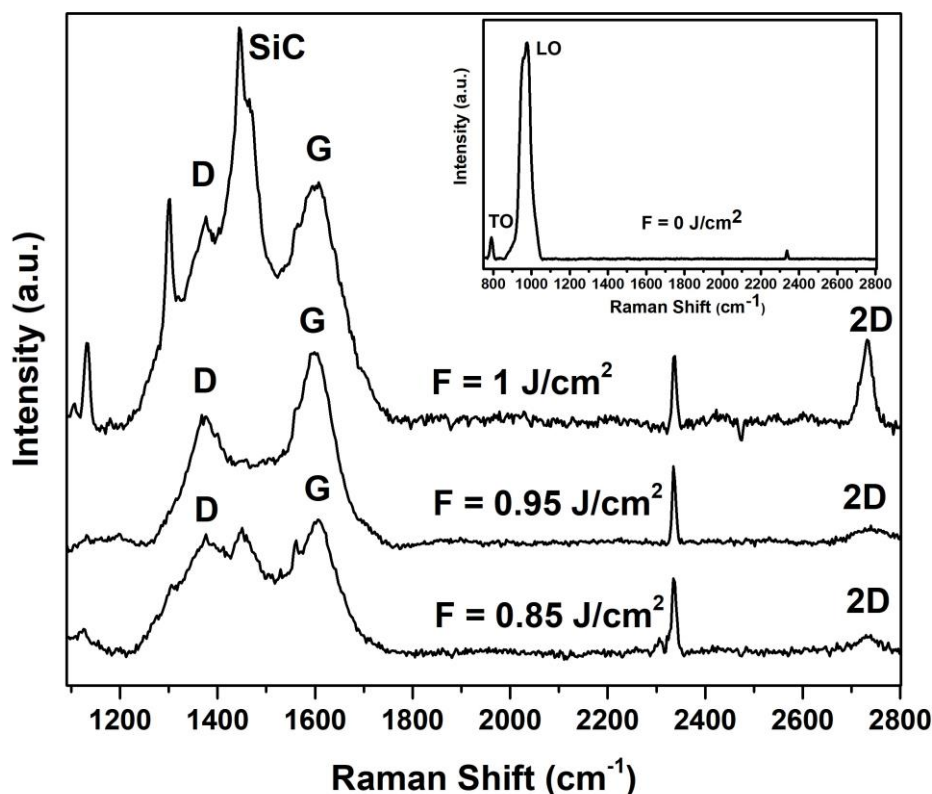


Figure. 3 Raman spectra of irradiated areas of 3C-SiC film with different fluence. The insert shows Raman spectra of as grown 3C-SiC film. LO and TO mark modes for SiC.

4. Conclusion

We demonstrate that local annealing of 3C-SiC film deposited on Si performed by 15 ns pulsed laser with 1064 nm with 20 Hz repetition rate with fluence above 0.8 J/cm^2 leads to formation in Raman spectra of 2D band with FWHM $\sim 31 \text{ cm}^{-1}$. This fact is explained by formation of few layered graphene film with turbostratic structure. The given experimental results show the perspective of the laser annealing for graphene epitaxial growth on surface of 3C-SiC/Si heterostructure.

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