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# Confirmation of the growth mechanism of the buffer layer in epitaxial graphene on SiC.

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Epitaxial graphene on SiC or so-called epigraphene has emerged as a highly promising candidate for nanoelectronic applications [1], [2]. Integration of graphene and SiC allows for synergistic benefits by harnessing the unique properties of both materials. SiC is a wide band-gap semiconductor with excellent thermal and chemical stability, making it suitable for high-power and high-temperature applications [3]. On the other hand, graphene is a two-dimensional material with remarkable electrical, mechanical, and optical properties [4–6]. This integration enables the development of novel electronic devices that can take advantage of the wide band-gap characteristics of SiC and the unique properties of graphene. For instance, one application is the development of graphene-based field-effect transistors (GFETs) on SiC substrates [7]. The SiC provides a robust and thermally stable platform, while the graphene channel offers high carrier mobility and compatibility with CMOS (complementary metal-oxide-semiconductor) processing techniques. This combination allows for the realization of high-performance and low-power electronic devices.

The successful application of graphene on SiC relies on a comprehensive understanding of the growth process, which significantly influences the quality, structure, and properties of the resulting graphene layers, ultimately affecting their suitability for various applications [8].

In current study, we conducted an experiment confirming the hypothesis regarding the epigraphene growth process on Si-face of 4H-SiC. Epigraphene growth on SiC occurs through thermal decomposition, also known as thermal decomposition epitaxy (TDE), where the SiC substrate acts as a carbon source. In the TDE process, high temperature causes Si atoms to vaporize from the SiC surface, whereas carbon partial vapor pressure is negligible. The remaining carbon atoms rearrange themselves into a graphene lattice structure, forming the first graphene "floating" at specific distance from the SiC surface. Due to this distance, following Si sublimation is possible and it results in the creation of a next graphene layer underneath the existing one. Thus, TDE process leads to the formation of a multilayer graphene stack, where every subsequent graphene layer originates from the SiC substrate pushing up previous graphene layers and keep "floating" on top of SiC surface. The process window of this TDE is determined by the temperature and partial vapor pressure of Si ( $P_{Si}$ ) in the proximity of SiC surface. Particularly for the Si-face, the  $P_{Si}$  is also related to the total number of grown graphene layers, providing a well-known self-limited epigraphene growth on SiC Si-face [8].

We assumed that as the SiC is cooled down to room temperature, the bottommost graphene layer of the stack approaches the SiC surface, transitioning into what is commonly known as the "buffer" layer: graphene layer partially covalently bonded to the underlying Si-face of SiC substrate. We show that throughout subsequent temperature rise, the grown stack of layers maintains its integrity with SiC substrate up to certain temperature threshold restricting silicon sublimation and thus further graphene growth. Figure 1 presents AFM scans taken at the exact same position on the sample after graphene growth and after subsequent annealing, providing evidence that there is no discernible change in the topography.

In order to confirm that the cool down step is responsible for this shift of the growth process window we conducted a control experiment, where temperature was raised directly without a cool down to the room temperature. We observed the growth of graphene along with a change in the SiC surface morphology (Fig 1c), confirming the continued sublimation of silicon with increasing temperature. Figure 2 presents Raman spectra of graphene with consecutive and direct annealing after the initial growth. The pronounced increase in G-band intensity indicates a corresponding rise in the number of graphene layers in case of direct annealing process.

Additionally, this experiment provides evidence that the bare SiC surface can be annealed (up to 1700°C) without significantly degrading its roughness value, even in the absence of an external deposited carbon capping (initial RMS value = 2.42 nm vs 1700°C RMS value = 2.80 nm).

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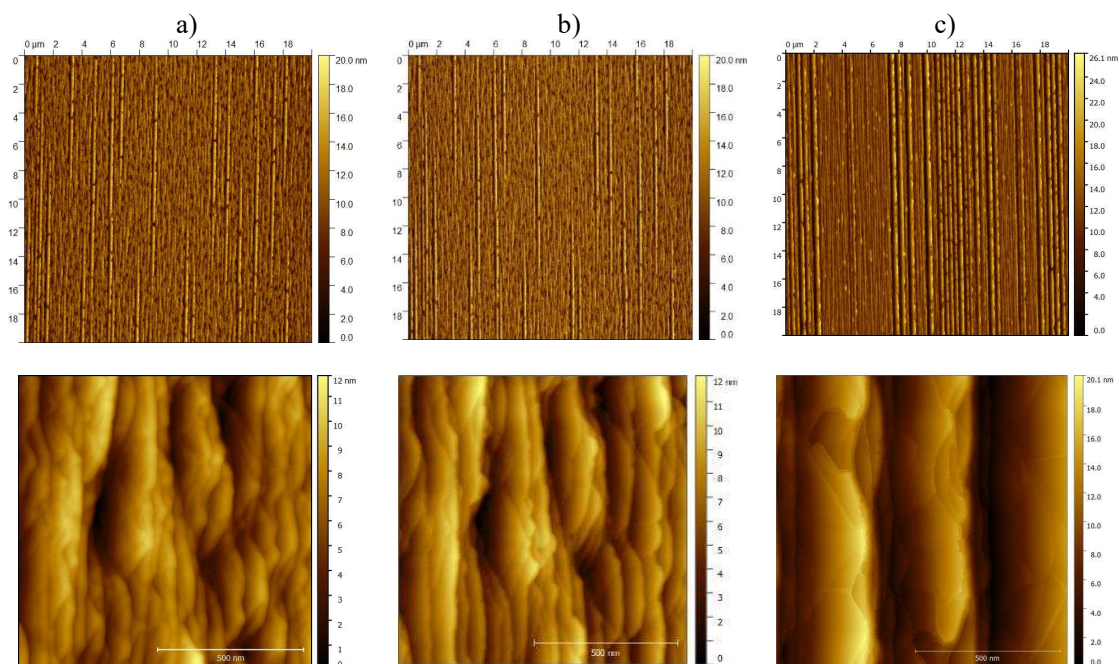


Fig. 1. AFM scans of 20x20  $\mu\text{m}$  (top panels) and zoomed 1x1  $\mu\text{m}$  (bottom panels) of 4H-SiC Si-face a) after initial graphene growth by sublimation at 1300°C; b) after subsequent annealing at 1700°C scanned at the same place as in (a); c) after consecutive growth at 1300°C and annealing at 1700°C without cooling down to the room temperature in between. Colorbar corresponds to the full z-scale of each image. RMS roughness values for top panels: a) 2.42 nm b) 2.80 nm c) 3.80 nm.

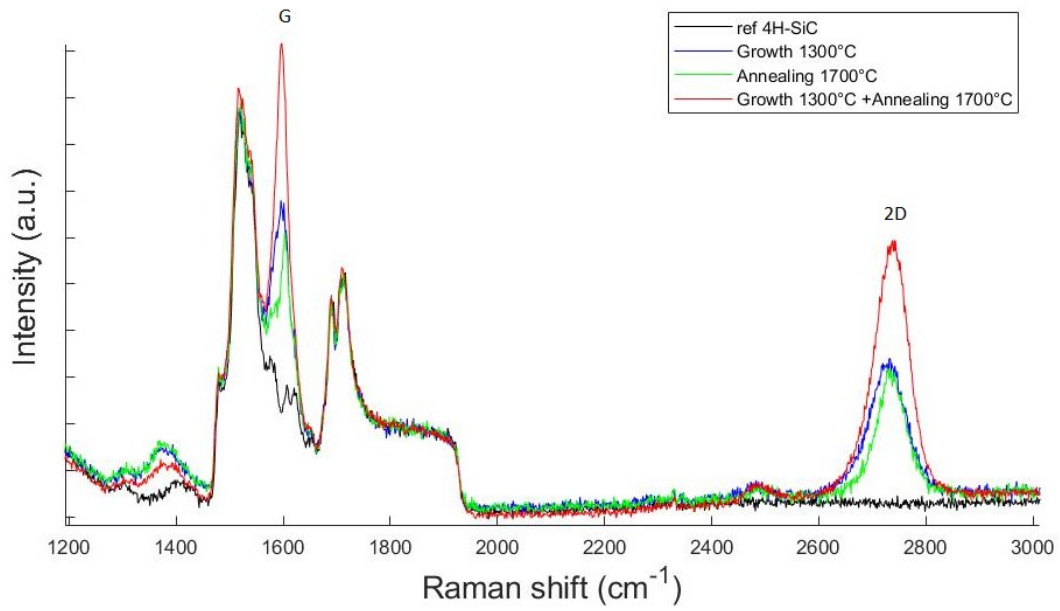


Fig. 2. Raman spectra of grown graphene (blue), graphene after cool down and annealing (green), graphene after consecutive growth and annealing without cool down step (red) and spectra of bare 4H-SiC (black). Laser excitation wavelength 532 nm.