Influence of the initial stepped SiC surface on epitaxial graphene formation

M.H. Oliveira Jr., T. Schumann, R. Hey, J.M.J. Lopes and H. Riechert

Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117 Berlin, Germany
oliveira@pdi-berlin.de

Due to its singular properties [1,2], graphene is currently considered to be one of the most promising materials for future electronic applications. Regarding its synthesis, several methods have been employed to prepare graphene layers, from which we can mention mechanical and chemical exfoliation, CVD on catalytic metal surfaces, and epitaxial growth on SiC by surface graphitization. This last approach offers particular potential since it can be realized on wafer size scale [3], without the need of further transferring the graphene layer to another insulating substrate (due to the insulating/semi-insulating nature of the SiC). However, in order to achieve an optimized growth of uniform and high-quality epitaxial graphene on SiC substrates, it is fundamental to understand the phenomena behind the graphene formation and its dependence on the surface step structure of the initial SiC surface. Several details of the nucleation and growth process, including step bunching effects resulting in large and homogeneous graphene terraces, are still under debate. In this work, we will present a study of the formation of epitaxial graphene on stepped SiC surfaces having different surface morphologies obtained as a function of the temperature during H-etching.

The samples investigated in this work were prepared using 6H-SiC(0001) substrates. H-etching and graphene growth were both performed in a cold wall reactor equipped with an induction heating system. The H-etching was carried out in a forming gas (95 % Ar and 5 % H_2) atmosphere at 900 mbar for 15 min. Different temperatures ranging from 1350 ºC to 1550 ºC led to SiC surfaces with different step heights and terrace widths. The step heights observed on the substrate before graphene preparation ranged from about 0.3 nm (non-etched sample) up to about 16 nm for the sample etched at 1550 ºC, while the terrace widths varied from 160 nm up to 5 μm, respectively. After the etching, high-quality epitaxial graphene layers were grown at 1600 ºC in a 900 mbar Ar atmosphere, as proposed by Emtsev *et al.* [4].

Figures 1 shows AFM profiles of a non-etched SiC surface before [1(a)] and after [1(b)] graphene formation, as well as an image of the graphene surface obtained by AFM [1(c)]. It is observed that due to step bunching, even a SiC surface with narrow terraces (~160 nm wide) and very small step heights (~0.3 nm) gives rise to a graphene layer showing macro-terraces (up to 4 μm wide) and large steps (up to 8 nm). The same effect [see figures 1(d),(e), and (f)] is observed for graphene prepared on SiC surfaces offering larger terraces (~0.5 μm wide) and step heights around 0.75 nm (i.e. half of $c_{6H\text{-}SiC}$). However, it is interesting to observe that for a stepped SiC surface with very wide terraces (~5 μm) and even higher steps (16 nm – more than 10 times $c_{6H\text{-}SiC}$), the subsequent graphene growth does not modify the initial surface morphology. As shown in Figs. 2 (a) and (b), there is no significant change on the terrace dimensions before and after graphene growth. Consequently, the profiles in Figs. 2 (c) and (d) reveal that the step heights remain almost unaltered as well. Both findings suggest that the step bunching process is suppressed in this case. Raman spectroscopy analysis reveal that, even though the graphene surface morphology seems to be much more regular for the sample initially etched at 1550 ºC (the case where no additional step bunching is observed during graphene formation), it presents much broader G (29 cm^{-1}) and 2D (61 cm^{-1}) peaks and a 2D peak shift of 42 cm^{-1} compared to the other samples [see Fig 2(e)]. One possible explanation for this is that, due to the absence of step bunching, the multilayer graphene formation at step edges becomes more pronounced. However, this hypothesis

is currently under investigation. Finally, the effects of a graphene growth on SiC with or without step bunching will be also discussed taking into account the resulting transport properties of the layers.

References

[1] K.S. Novoselov et al. Nature, **438** (2005) 197.
[2] Y. Zhang et al. Nature, **438** (2005) 201.
[3] Y.M. Lin et al. Science, **327** (2010) 662.
[4] K.V. Emtsev et al. Nature Materials 8, **3** (2009) 203.

Figures

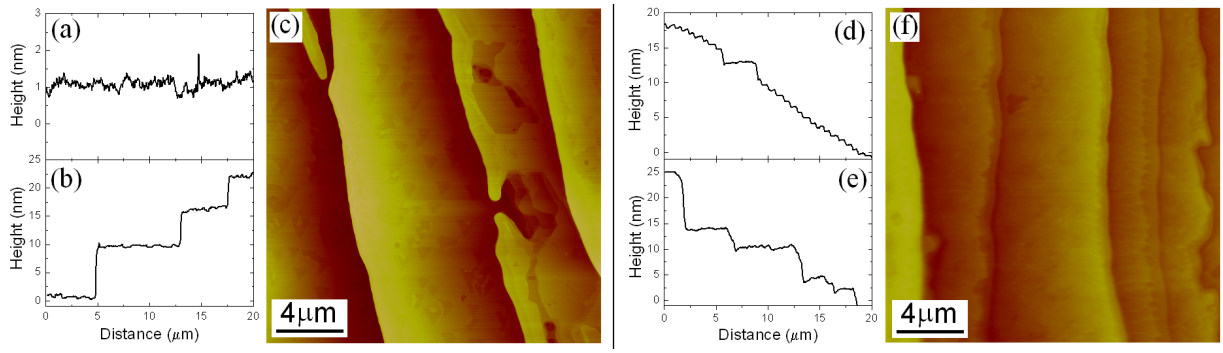

Figure 1

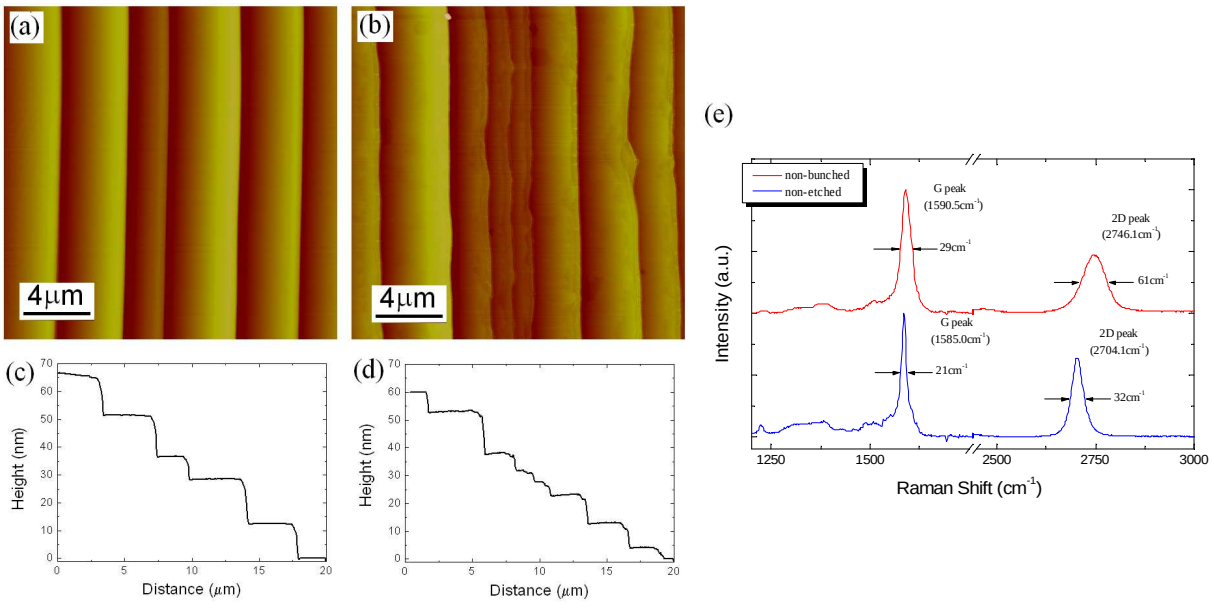

Figure 2

Figure caption

*Figure 1 – Comparison between the profiles for a non-etched SiC sample before **(a)** and after **(b)** the graphene growth, as determined by AFM. AFM image of the surface after graphene formation **(c)**. The same comparison is presented [**(d)** and **(e)**] for a SiC sample etched at 1500 ºC. The respective AFM image of the surface after graphene formation is shown in **(f)**.*

*Figure 2 – Surface image obtained by AFM for the sample etched at 1550 ºC before **(a)** and after **(b)** graphene preparation, and their respective profiles **(c)** and **(d)**. Even though the graphene surface morphology in this case seems to be more regular as compared to the other samples [see Figs. 1 (c) and (f)], its Raman spectrum **(e)** shows broader G and 2D peaks, what may be related to the existence of multilayer graphene on the step edges due to the absence of a step-bunching process.*