Structure and interface of graphene films grown on SiC using propane-hydrogen-argon CVD

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Graphitization of annealed SiC, first studied in the mid 70's, has been widely explored last years as a way of producing graphene films on semi-insulating substrates for both fundamental studies and graphene-based applications. In this method, graphene grows from carbon atoms of the SiC substrate. More recently, graphene growth from external sources of carbon has been demonstrated under ultra-high vacuum conditions [1,2] or argon atmosphere CVD conditions [3,4]. For all these methods, graphene/SiC interface and graphene structural properties depend mainly on the SiC surface orientation: on the Si-face, graphene/SiC interface is $(6\sqrt{3}\times6\sqrt{3})$-R30° reconstructed and graphene layers are bernally stacked, while on the C-face, no graphene/SiC interface reconstruction is observed and graphene layers are disoriented with respect to others (rotational disorder).

In previous works, we have shown that propane-hydrogen CVD (i.e. using propane as the carbon precursor and hydrogen as the carrier gas) is an exception, as this method allows growing graphene films presenting either Bernal or disoriented stacking on the Si-face [5,6]. In this contribution, this original behavior is studied through samples grown using different hydrogen/argon mixtures for the carrier gas. We also discuss and compare our results with what can be obtained using SiC annealing to underline differences between propane-hydrogen-argon CVD and annealing methods.

Samples are grown on 6H-SiC(0001) at 1450°C and 800 mbar with a propane flow of 5 sccm for 5 minutes. We use for the carrier gas different argon/hydrogen mixture with hydrogen ratio R_{H2} ($R_{H2} = F_{H2} / (F_{H2} + F_{Ar})$) ranging from 38% to 100% (7 samples). Figure 1 presents low energy electron diffraction (LEED) patterns and atomic force microscopy (AFM) images of samples grown with R_{H2} = 38%, 53%, and 100%. For R_{H2} = 38%, AFM image shows SiC atomic steps (0.75 nm) or sometime, SiC ± graphene atomic steps (0.75 ± 0.35 nm), while LEED pattern presents graphene and $(6\sqrt{3}\times6\sqrt{3})$-R30° interface reconstruction spots. When increasing the hydrogen ratio (R_{H2} = 53%), $6\sqrt{3}$ spots become more shadow, while new graphene orientations (0°, 30 ± 2°) and a ring appear. The appearance of rotational disorder on the LEED pattern is correlated to the appearance of few isolated wrinkles on the AFM view. Finally, for R_{H2} = 100% (pure hydrogen), the $(6\sqrt{3}\times6\sqrt{3})$-R30° interface vanishes totally and graphitic ring appears clearly on the LEED pattern. On the AFM view, wrinkles join to form a network on the surface.

In our contribution, we will analyze this set of samples using X-ray photoemission spectroscopy (XPS), LEED, AFM, and Raman spectroscopy. LEED and AFM study will allow to discuss on the links between graphene/SiC interface and wrinkle formation, and Raman spectroscopy will be used to study the influence on graphene/SiC interface on the strain. Finally, we will compare our results to what can be observed using the annealing method on the Si or C-face in order to evidence the numerous differences between the samples grown using both techniques.

References

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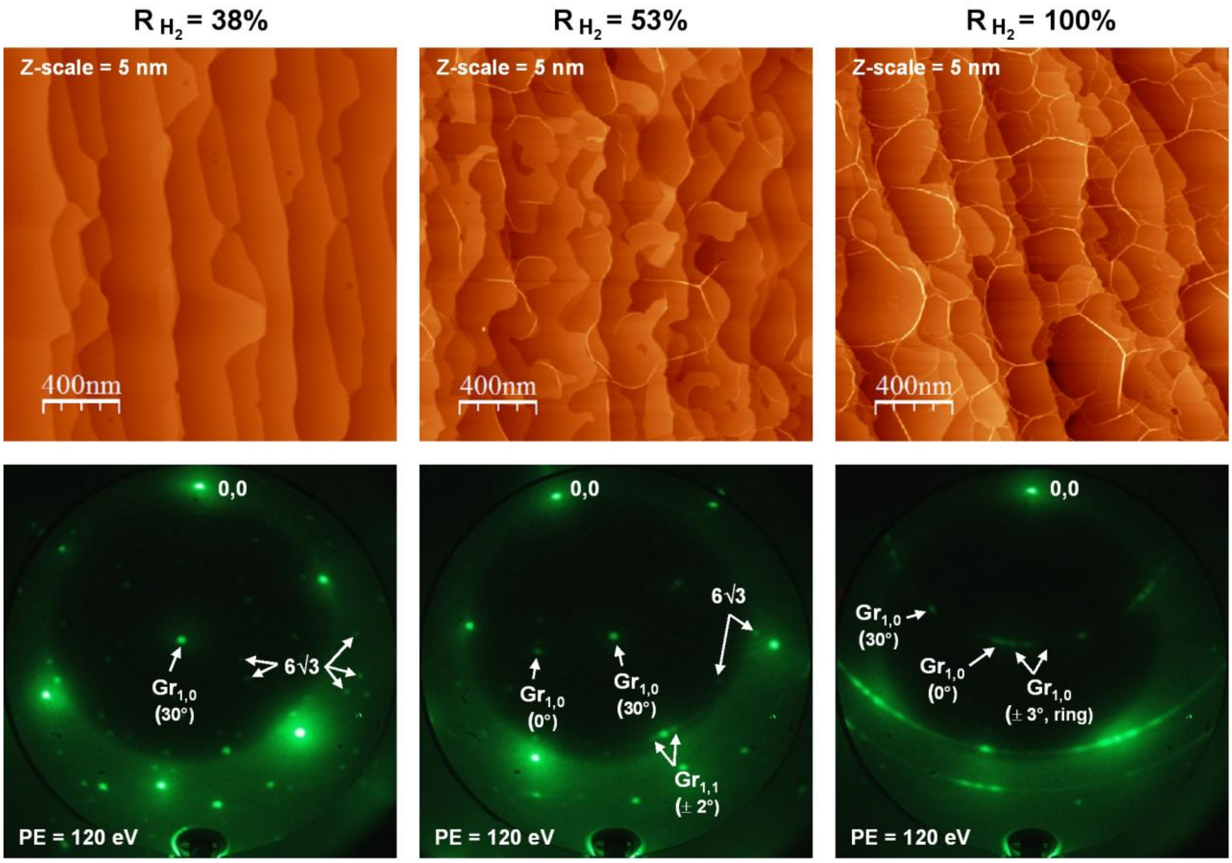
Figure 1



AFM images and LEED patterns from samples grown by propane-hydrogen-argon CVD on 6H-SiC(0001) with different hydrogen/argon mixtures used as the carrier gas.