

Supplementary Information

Epitaxial Graphene morphological imperfections: from a hindrance to the generation of new photo-responses in visible domain

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Here, we report part of our photoresistance measurement, currently under study, of un-oxidised 4H-SiC epitaxial graphene grown on face terminated carbon. We present the XPS measurements to confirm the located layer numbers and the photoresistance measurements.

I- XPS measurements

We performed XPS measurements for graphene grown on 4H-SiC (000 $\bar{1}$). The major graphene component is assigned to the C_{1s} core level. The C_{1s} spectrum was fitted with three components: the first peak is assigned to the sp² hybridised C atoms in graphene positioned at 284.32 eV,¹⁴ the second peak is related to the sp³ hybridised C atoms appearing at 285.1 eV and the third small peak located at 286.6 eV is associated with the C-O bonds (see, Fig. 1S).¹⁴ The C_{1s} illustrates the graphene layers across the sample surface. The intensity ratio of sp³- and sp²- hybridised carbon components gives a better determination of the graphene layer numbers **n**, especially, for high layer numbers. We found an intensity ratio of sp³- and sp²-hybridised carbon components of about 0.26 (**n**>5). This is also confirmed by the SiC component behaviour usually located at lower binding energy $\approx$ 282.6 eV. As the graphene intensity reading increases (i.e. the number of graphene layers increases) the intensity of the SiC decreases. No buffer layer was observed here. On the other hand, we notice the presence of a small oxygen component at 286.6 eV. It should be noted that the presence of oxygen can be accounted for by the contamination of the graphene layer. In fact, epitaxial growth of graphene on face terminated carbon is associated with unintentional oxidation of the C-terminated face, as reported in previous studies.¹⁶ However, the presence of oxygen is very small for it to induce an oxidation. In fact, oxidation occurs for a well-defined percent of oxygen in the system that varies between 5-8%.

For epitaxial graphene with face terminated carbon, the films oxidation is determined through a large change of the Si_{2p} peak alongside where a SiO₂ peak must appear, as reported in previous studies.²⁰ This was also observed for the 6H-SiC substrate in the main paper. We have also investigated the Si_{2p} components here. The Si_{2p} peak shows exclusively a single chemical environment of Si. In fact, we located a unique Si_{2p} which is consistent with the SiC at $\sim$ 100.0 eV binding energy. No extra Si products (such as SiO₂), besides SiC, exist. This shows the graphene is not oxidised and there is only weak contamination.

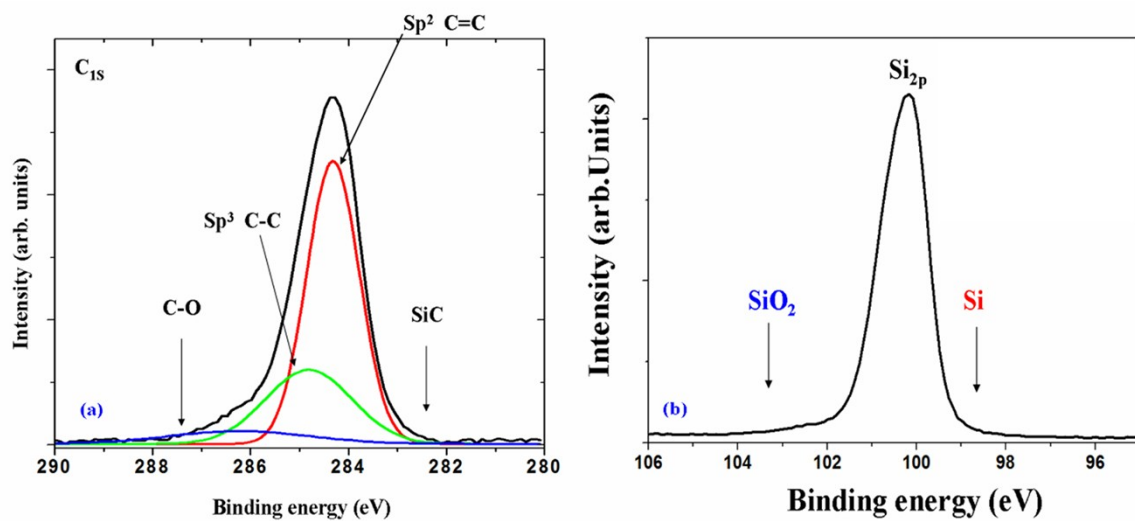


Figure 1S: XPS components of epitaxial graphene layers grown on 4H-SiC face terminated carbon 4H-SiC (000-1) respectively: **a-** C_{1s} core level and **b-** Si_{2p} .

II- Photoresistance measurements

We measured the photoresistance of the n graphene layers grown on 4H-SiC face terminated carbon (4H-SiC (000-1)). We proceed similarly to the photoresistance measurements detailed in the main paper. We studied the photoresistance effect using violet wavelength. Nevertheless, we did not locate any photoresponse (see, Figure 2S). Here, the epitaxial graphene response is similarly to a semiconductor. As reported in the main paper, the photoresistance response is mainly obtained due to the high presence of the impurities (oxidation). Here, the present oxygen is very weak that can be neglected. This does not oxidise the layers and induces a slight increase of the local defects. Small amount of defects remain unable to change the state of the system. In fact, electrons still have a long scattering time. Contrary to the case of oxidation, a new state of the epitaxial graphene system is defined and the scattering time of electrons increases. This results in the enhancement of the resistance as we discussed in the main paper. This study illustrates how important the effects of the oxidation on the photo response are.

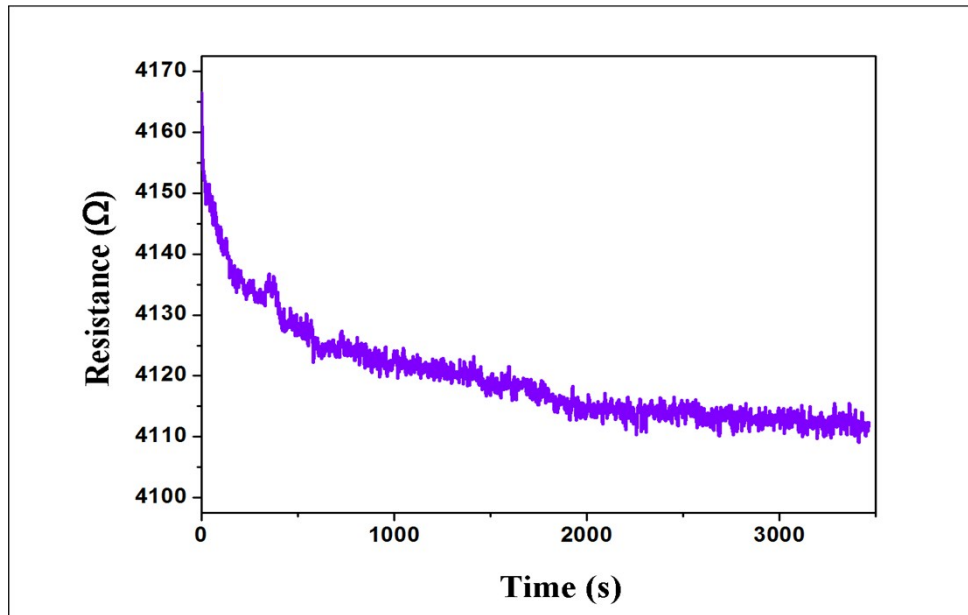


Figure 2S: Photoresistance of five layers of epitaxial graphene grown on face terminated carbon - 4H-SiC (000-1) - under violet laser excitation.