## Correlation between morphology and transport properties of quasi-free-standing monolayer graphene

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Quasi-free-standing monolayer graphene (QFMLG), obtained by intercalating hydrogen at the interface of buffer layer and SiC(0001), is efficiently decoupled from the substrate and a promising material for wafer-scale graphene-based nanoelectronics [1-3]. However, the mobility of QFMLG is limited to a value lower than epitaxial monolayer graphene (~3000 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>), and the carrier scattering has not been fully understood. Recently it has been reported that the mobility of QFMLG depends on the substrate temperature during the hydrogen intercalation process, and the highest mobility is obtained at 700-800°C [4]. These measurements suggested that the carrier scattering is mainly caused by charged impurities at 600 and 800°C, and by defects at 950°C [4]. We have used scanning tunneling microscopy (STM) to study the surface structure of QFMLG formed at several hydrogen intercalation temperatures, and investigated the relationship with transport measurements.

Our STM observations reveal that the QFMLG formed at  $600^{\circ}$ C and  $800^{\circ}$ C shows defects (dark spots) with a diameter of 1.5 nm, depth of 20-80 pm, and density of  $1\times10^{13}$  cm<sup>-2</sup>, while samples formed at  $1000^{\circ}$ C show dark spots with diameter 4-10 nm, depth 250 pm, and density  $6\times10^{10}$  cm<sup>-2</sup>. The dark spots at  $600^{\circ}$ C and  $800^{\circ}$ C partially align with a periodicity of 1.8 nm, corresponding to the quasi-(6x6) reconstruction of the buffer layer.

This implies that hydrogen intercalation in our samples is not complete at 600°C and 800°C, and the remaining patches of buffer layer are observed as dark spots. Since the depth of the dark spot defects at 1000°C corresponds to the height of a SiC(0001) bilayer, they are identified as holes in the SiC substrate, probably due to etching by hydrogen or redistribution of surface atoms at high temperature. This is consistent with transport measurements and suggests that the incomplete hydrogen intercalation at 600 and 800°C scatters carriers as charged impurities, while the holes in the SiC substrate at 1000°C act as defects.

A higher mobility of QFMLG could therefore be obtained by optimizing the hydrogen intercalation temperature which should be as high as possible to intercalate more hydrogen, but lower than the temperature at which holes appear.

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