

Short Term Mobility 2014 Report:

STM and Optical Investigation on silicene monolayer and multilayer on Ag(111) and Si(111)

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The work has been addressed on the measurements of the structural, electronic and optical properties of single and multilayer silicene synthesized on Ag(111) and on Si(111).

We realized *in situ* ($\sqrt{3}\times\sqrt{3}$)R30° multilayer silicene on Ag(111) after the first $3\times 3/4\times 4$ silicene layer, and on 1 ML of silver, with ($\sqrt{3}\times\sqrt{3}$)R30° reconstruction, grown on Si(111)7×7. We used scanning tunnelling microscopy, scanning tunnelling spectroscopy (STM/STS) and Raman optical measurements, as a function of temperature from RT down to 4 K, to investigate the epitaxial growth and the optical/electronic properties of multilayer 2D silicene on metallic Ag(111) and semiconducting Si(111) substrate.

Afterward the synthesis of single layer silicene on different metallic substrates ¹⁻³, multilayer silicene, the silicon correspondent of multilayer graphene ⁴, was synthesized *in situ* under ultra high vacuum (UHV) on silver (111) surfaces ⁵⁻⁷. These films grow in successive flat terraces and can cover the entire surface, past the initial formation of the dominant 3×3 reconstructed first silicene monolayer, which is in a 4×4 coincidence super cell with respect to the silver (111) unit cell ⁵⁻⁷. All terraces show a honeycomb $\sqrt{3}\times\sqrt{3}$ R(30°) surface structure, with respect to 1×1 silicene, as observed in Scanning

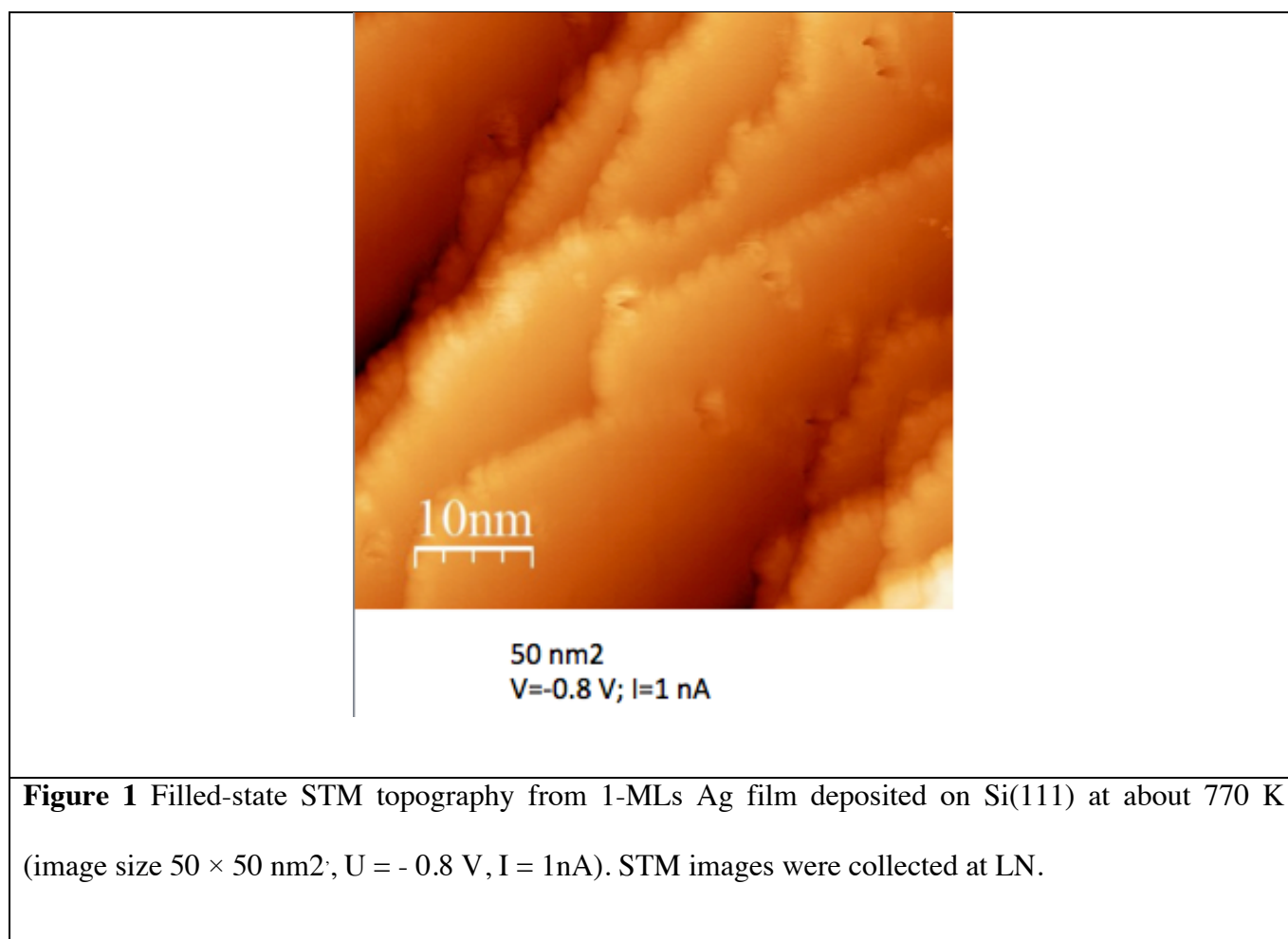
Tunnelling Microscopy⁵⁻⁸ and as also reflected in Low Energy Electron Diffraction patterns (LEED)⁵⁻⁸. Thick epitaxial multilayer silicene films with $\sqrt{3}\times\sqrt{3}R(30^\circ)$ surface structure grown on Ag(111) showed the G, D and 2D Raman structures, which are unique fingerprints of thick multilayer silicene⁹.

Interestingly, the paper was focused on the synthesis and study of multilayer silicene obtained on silicon supports (by solid phase deposition) after obtaining the interface Ag-Si(111) 7×7 with $\sqrt{3}\times\sqrt{3}R(30^\circ)$ symmetry beyond the actual state-of-the-art consisting in Si self-assembling on massive single crystal Ag(111) used as a template. This will open the way to realize a prototype of multilayer silicene directly on Si substrate, envisaging the possibility to use silicene-based devices in nano-technological and/or industrial applications.

Here, we report the preliminary results on the realization and study of 2D multilayers of silicene sheets first grown on Ag(111) and subsequently on Si(111), *via* a 1ML $\sqrt{3}\times\sqrt{3}R(30^\circ)$ silver used as a template, investigated by STM/STS and Raman spectroscopy, obtained during the short term mobility stage of last November 2014 at Institute for superconducting and Electronic Materials (ISEM) Australian Institute for Innovative Materials (AIIM) Innovation Campus University of Wollongong (Australia). The multilayer silicene sheets were grown on Ag(111), and on Ag/Si(111) in the UHV chamber (base pressure: 7.5×10^{-11} mbar) at about $T \sim 470$ K by Si source. Ag solid evaporation source was used to produce the $\sqrt{3}\sqrt{3}R(30^\circ)$ Ag/Si(111) 7×7 at ~ 770 K after repeatedly flashes and annealing of the Si(111) substrate at about 1350 K, while keeping the pressure below 2×10^{-10} mbar during heating. An infrared pyrometer was used to measure the sample temperature. Silicon (7×7) and Ag (1×1) structures were observed by STM before the Si evaporation at rate of ~ 0.03 ML/min from a Si source, up to few MLs, while the Ag(111) or/and Ag/Si(111) interface was kept at $T \sim 470$ K to produce the 2D multilayers silicene sheets. The STM images were recorded at liquid Nitrogen (LN) in constant-current mode at a bias voltage from ± 0.1 to ± 3 V and a tunnelling current from 0.5 to 10 nA.

We report here, the preliminarily results showing from 1ML $\sqrt{3}\times\sqrt{3}R(30^\circ)$ Ag/Si(111) 7×7 interface.

Figure 1 (a) is a typical STM topography image for 1-ML Ag film deposited at 770 K, where the silicon terraces having a size comprise between 30-100 nm are completely covered by silver atoms. This is the template for growing the multilayer silicene, as in the case of multilayer silicene on Ag(111)⁵⁻⁹, by showing the $\sqrt{3}\times\sqrt{3}R(30^\circ)$ silicene. Figure 1(b) displays the high resolution STM image from $\sqrt{3}\times\sqrt{3}R(30^\circ)$ Ag/Si(111).



8 nm², V=0.5, I=1nA

From Aizawa et al., Surf. Sci.429,509 (1999)

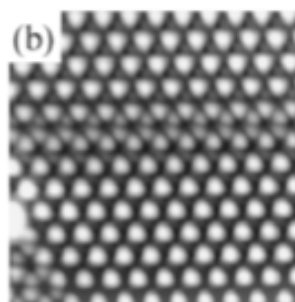
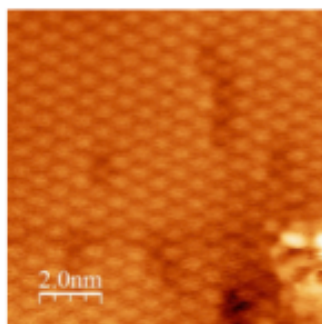
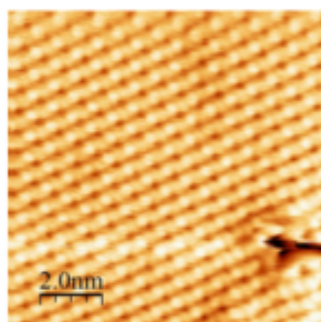


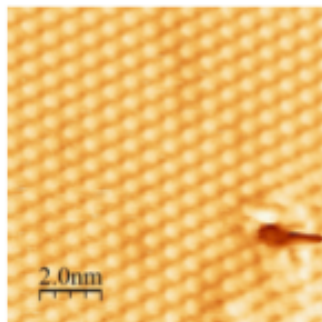
Fig. 4. Empty-state STM images measured at 62 K for two different regions of the Si(111)- $\sqrt{3}\times\sqrt{3}$ -Ag surface. The displayed areas, tunneling currents, and sample bias voltages were (a) $0.125 \times 0.125 \text{ Å}^2$, 1.3 nA, and 1.5 V, and (b) $80 \times 80 \text{ Å}^2$, 1.0 nA, and 0.5 V.



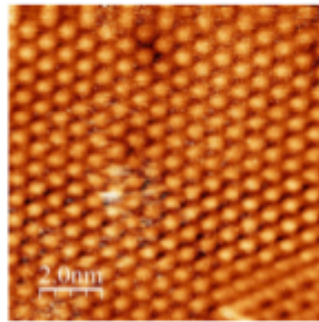
10 nm²
V=-0.8 V



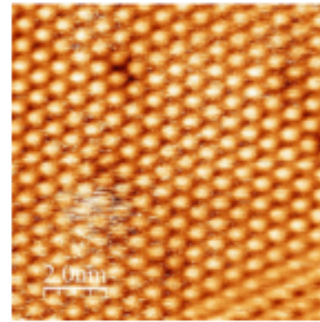
10 nm²
V=0.4 V



10 nm²
V=0.8 V



10 nm²
V=1.2 V



10 nm²
V=1.2 V

Figure 2 Empty (filled)-state high-resolution STM topographies for 1-MLs $\text{Ag}(\sqrt{3}\times\sqrt{3})\text{R}30^\circ$ deposited on Si(111) at about 770 K (image size $10 \times 10 \text{ nm}^2$, $U = 0.4\text{-}1.2 \text{ V}$, $I = 0.5\text{-}1 \text{ nA}$; the STM from Aizawa (Surf. Sci. 1999) was reported for comparison. STM images were collected at LN.

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