sp^2 Materials

(graphene, silicene, phosphorene, germanene, dichalcogenide,…..2D layered Materials)

1D Nano-Ribbons and 2D Sheet Silicene: Structural and Electronic Properties

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Aim of the talk:

- Brief introduction on Dirac Fermions system (graphene, silicene)

- Display what the theory predicts on Silicene system

- Show the Structural and Electronic properties of:

- Self-organized (1D), (2D grating) and (3D) Multilayer Silicene/Ag(110)
- 2D Silicene sheet on Ag(111) and multilayer silicene o Ag(111)

by Angle-resolved Photoemission spectroscopy (ARPES), which is the principal technique to study Silicene
and by: STM, LEED and AUGER techniques

-O_2 on Silicene/Ag(110) and Silicene/Ag(111) (studied by STM and core level specroscopy)

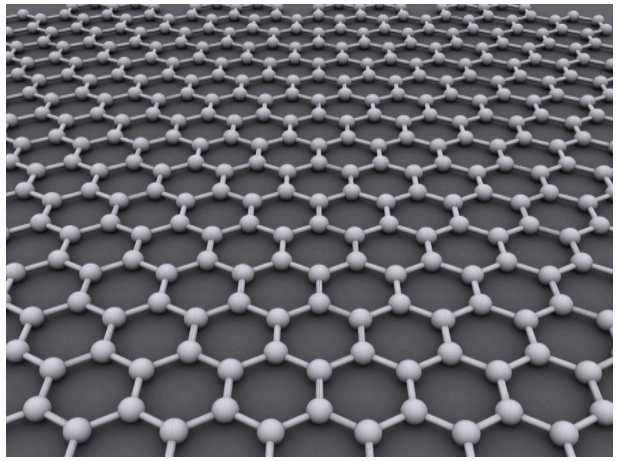
- XRD, EDXR and Raman spectroscopy on multilayer silicene grown on Ag(111)

- Summary and Perspectives

- Brief introduction on Dirac Fermions system(graphene/silicene (and in general 2D Materials)

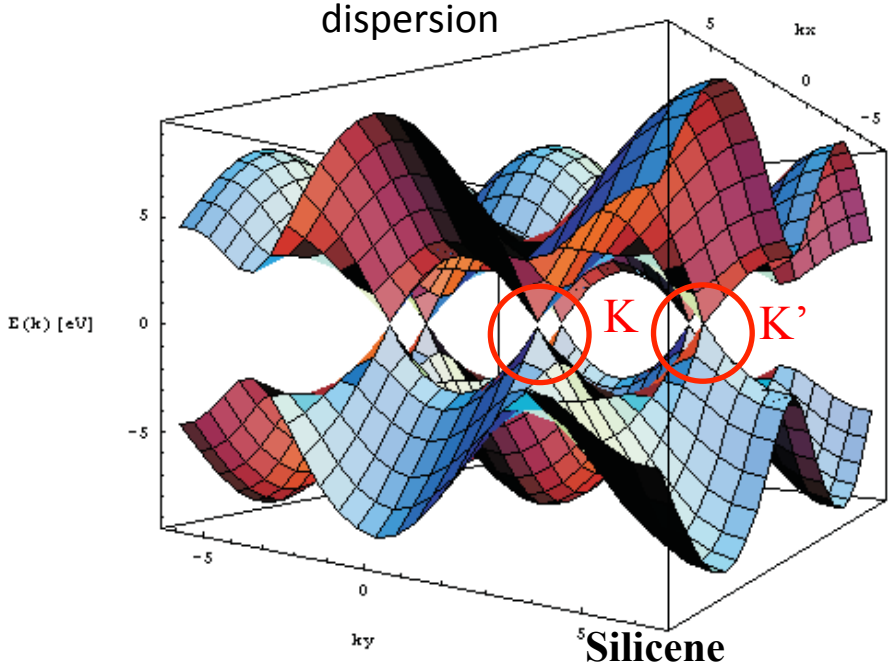
Silicene, is a hexagonally Si sheet in which sp^2 hybridized electrons (σ electrons) form a honeycomb structure and the remaining π electrons follow the massless particles Dirac equation.

Honeycomb lattice-sp^2



Silicene

3D E(k) dispersion



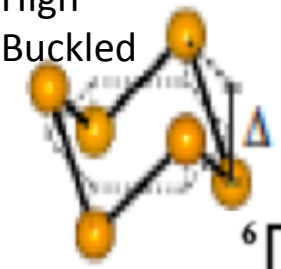
Silicene

From an electronic point of view, **Silicene** might be equivalent to graphene with the advantage that it is probably more easily interfaced with existing electronic devices and technologies.

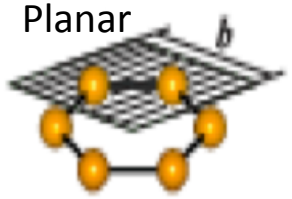
BUT, the sp^2 bonded Si is much less common than for C, because the natural Si has thetrahedral structure, where the Si atoms are sp^3 bonded. We have to synthesize Si in honeycomb structure!

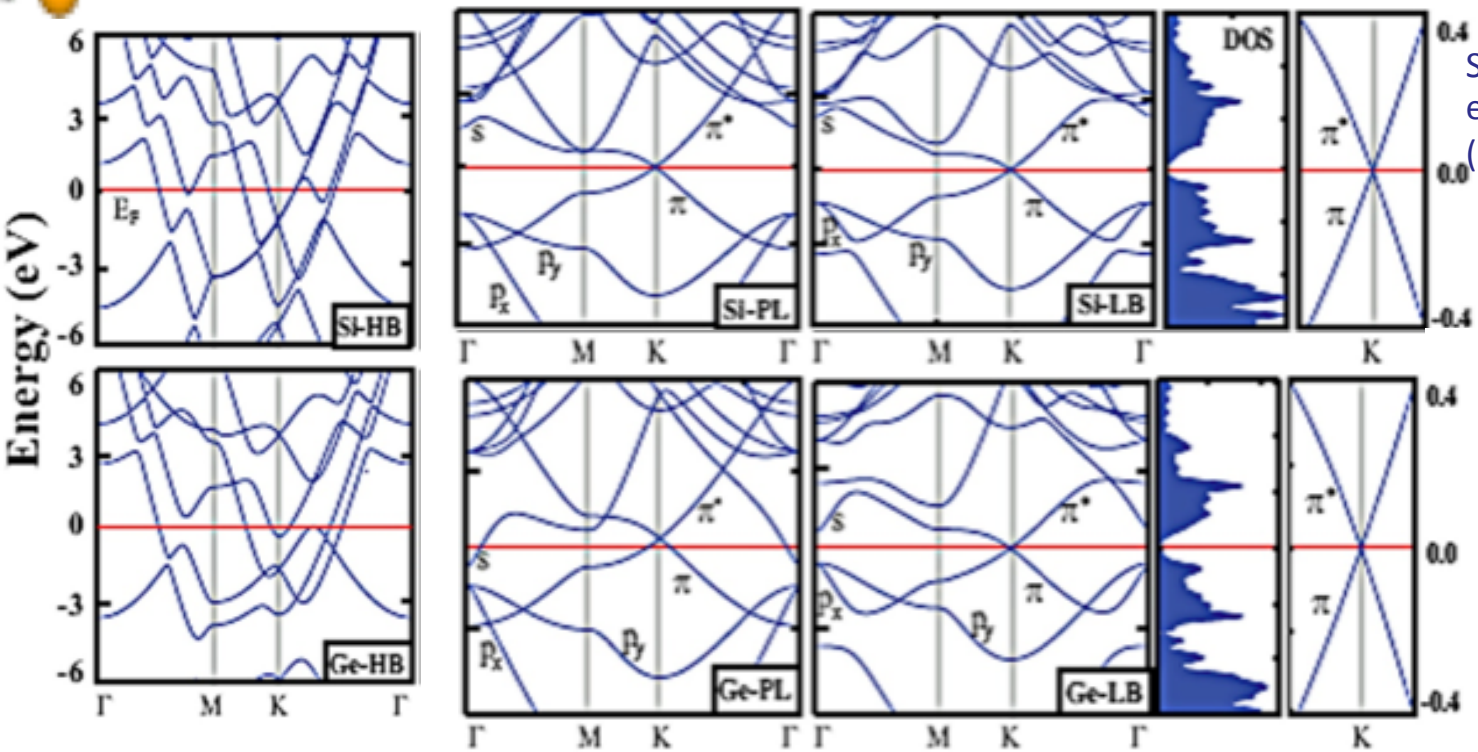
Theoretical prediction of Silicene

High Buckled



Planar





S. Cahangirov et al. PRL. 102 (2009) 236804

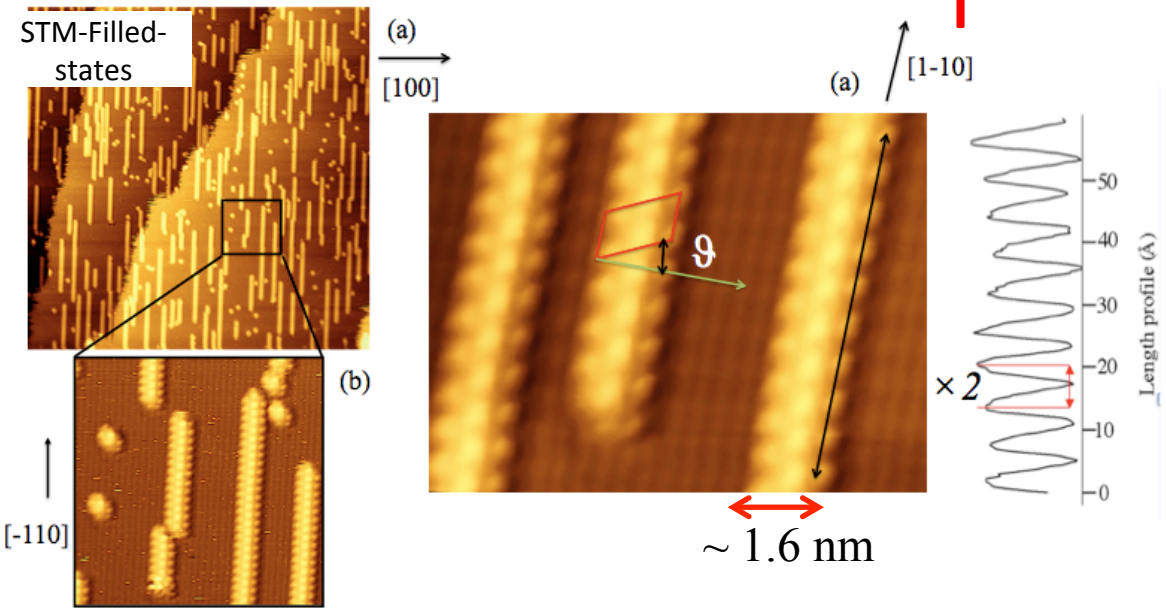
Energy band structures of **silicene** and of **germanene** calculated for low-buckled (LB) and planar (PL) geometries.
In the LB case the density of states is also presented.
The crossing of **the π and π* bands at the K points of the BZ** is amplified to show they **are linear near the crossing point.**

Silicene, has an electronic structure very similar to the one of graphene, making them possibly equivalent.
N.B.: The term « silicene » was first introduced by Gian G. Guzmán-Verri and L. C. Lew Yan Voon, PRB 76 (2007) 075131

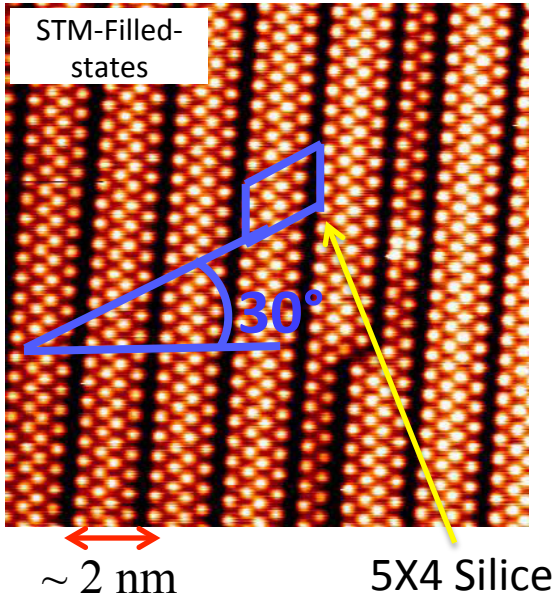
Self-organized Silicene Nanoribbons on Ag(110)

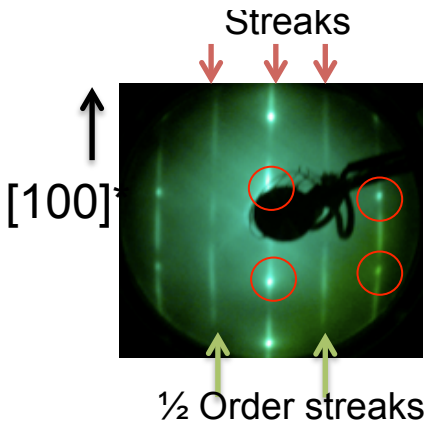
1D → 2D grating

RT

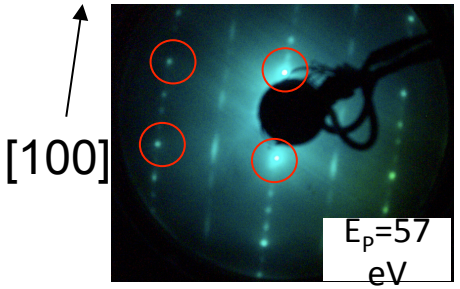


470 K





LEED 5x2/5x4

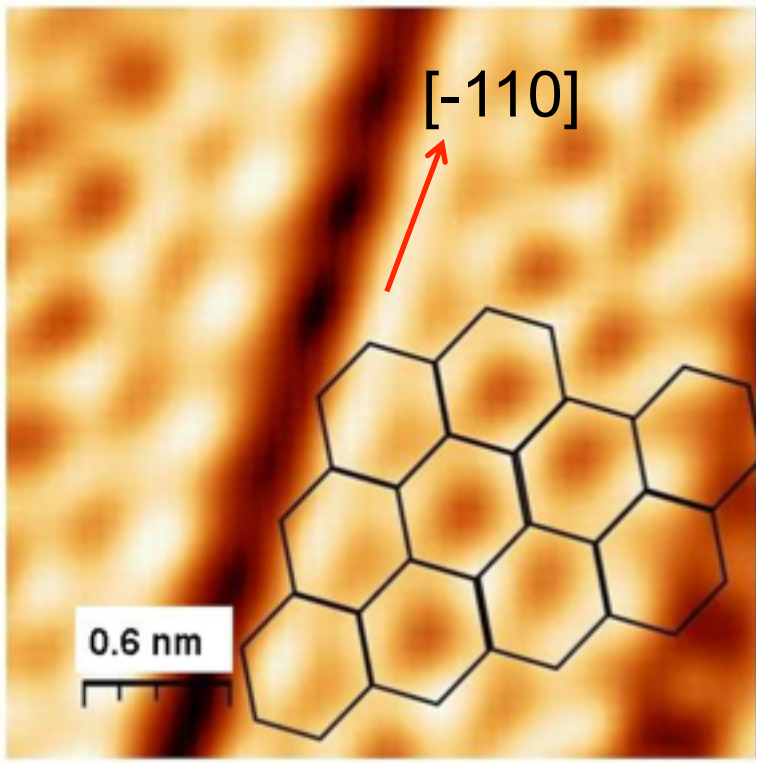


Léandri et al., Surf. Sci. 2005, **574**, L9
De Padova et al. Nano Lett. 2008, **8**, 271
De Padova et al. J. Phys.: Cond. Mat. 2012, **24**, 223001

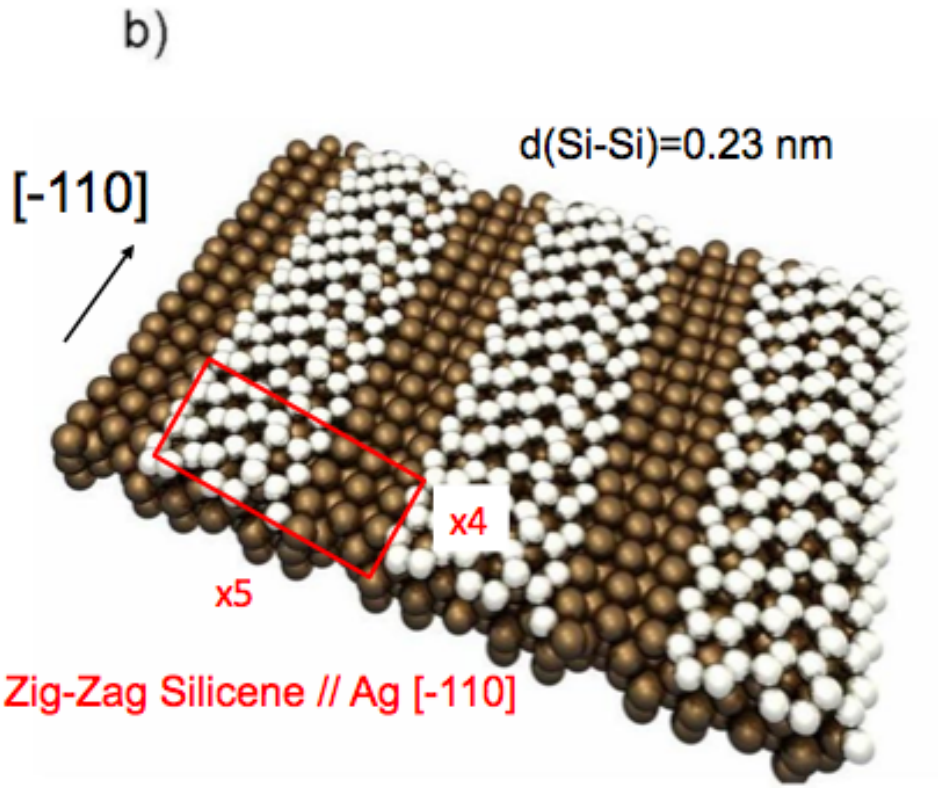
H. Sahaf et al., Appl. Phys. Lett., 2007, **90**, 263110
F. Ronci et al., Physica Status Solidi C 2010, **7**, 2716

Atomic structure of Self-organized 2D Silicene grating NRs on Ag(110)

Filled-states STM



MODEL

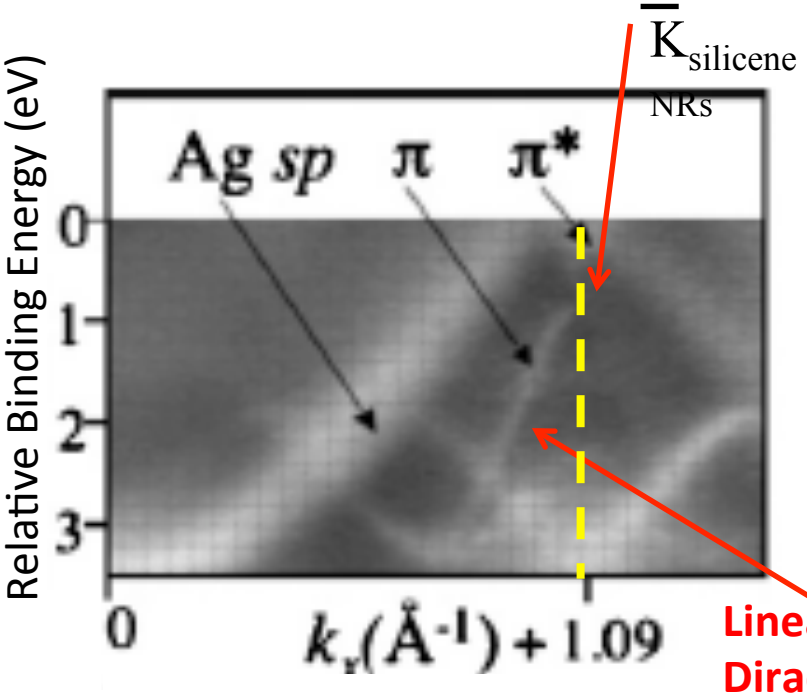


The honeycomb structure of the nanoribbons reveals that they are stripes of graphene-like silicon sheets, i. e., silicene nanoribbons

B. Aufray, A. Kara, S. Vizzini, H. Oughaddou,C. Léandri, B. Ealet and G. Le Lay, Appl. Phys. Lett. **96**, 183102 (2010); Kara et al. J- Supercond. Novel Mag. 22, 259 (2009); A. Kara, S. Vizzini, C. Leandri, B. Ealet, H. Oughaddou, B.Aufray, and G. Le Lay, J. Phys.-Cond. Mat. **22** (2010) 045004

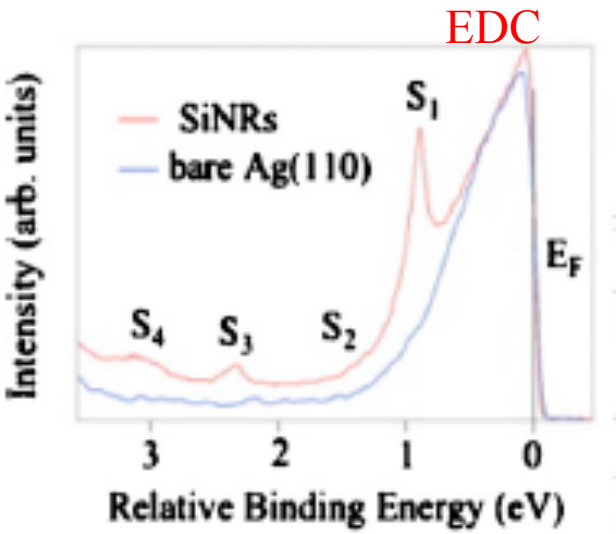
} DFT calculations

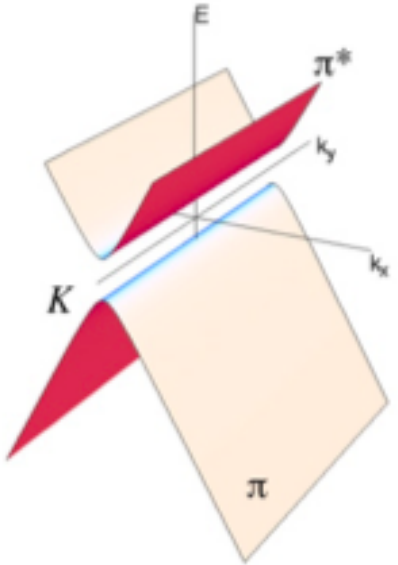
Dirac cone of Silicene NRs



Linear dispersion: Dirac fermions

$\bar{\Gamma} \rightarrow \bar{K}_{silicene\ NRs}$





Fermi velocity ≈1 10^6 m/s

(graphene: ~1 10^6 m/s)

GAP≈0.5 eV

De Padova et al., APL 96 (2010) 261905

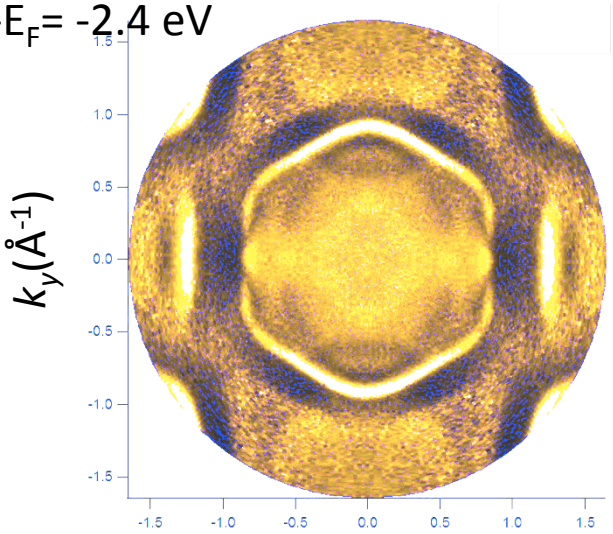
Energy Contour Section

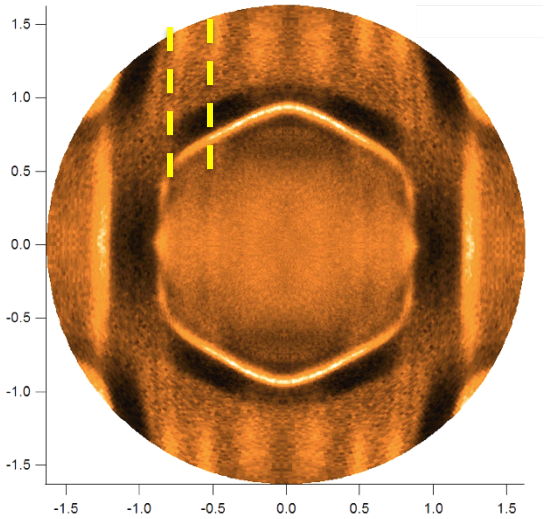
bare Ag(110)

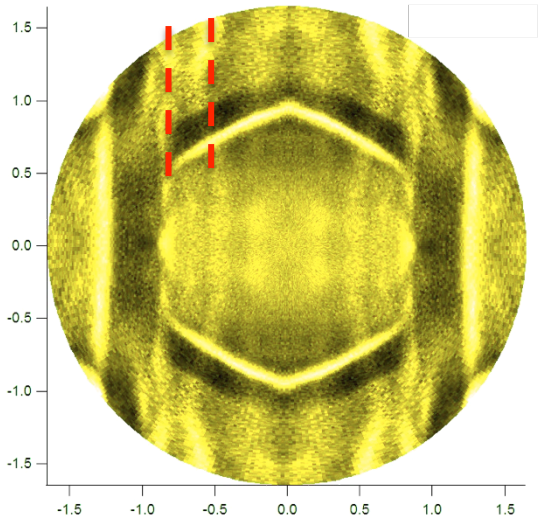
1D SiNRs

2D Grating SiNRs

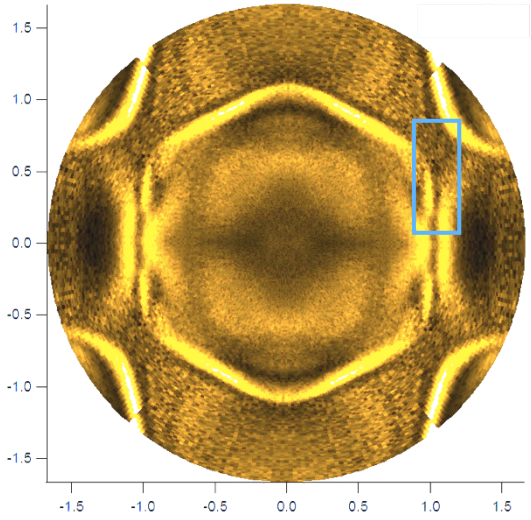
$E-E_F$= -2.4 eV

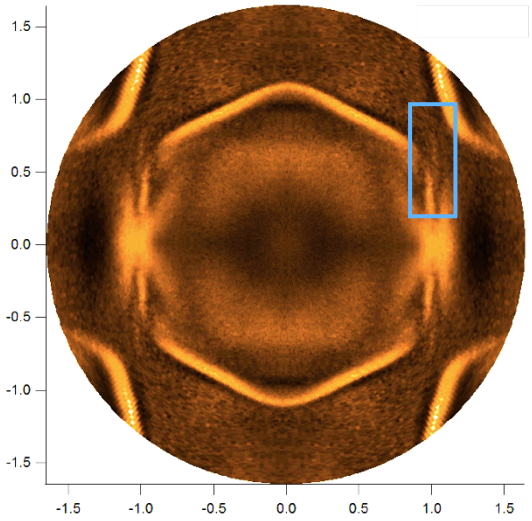


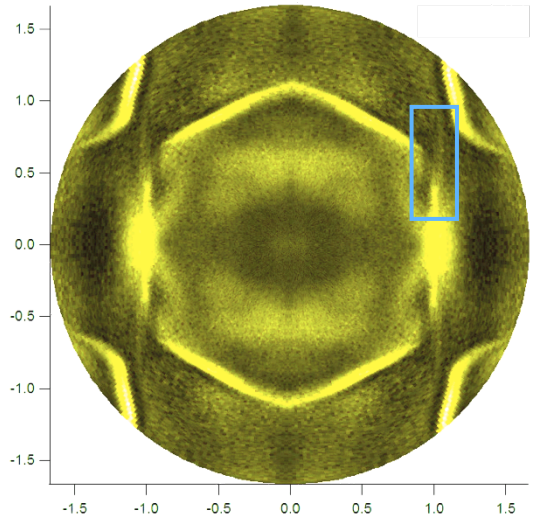




$E-E_F$= -0.9 eV

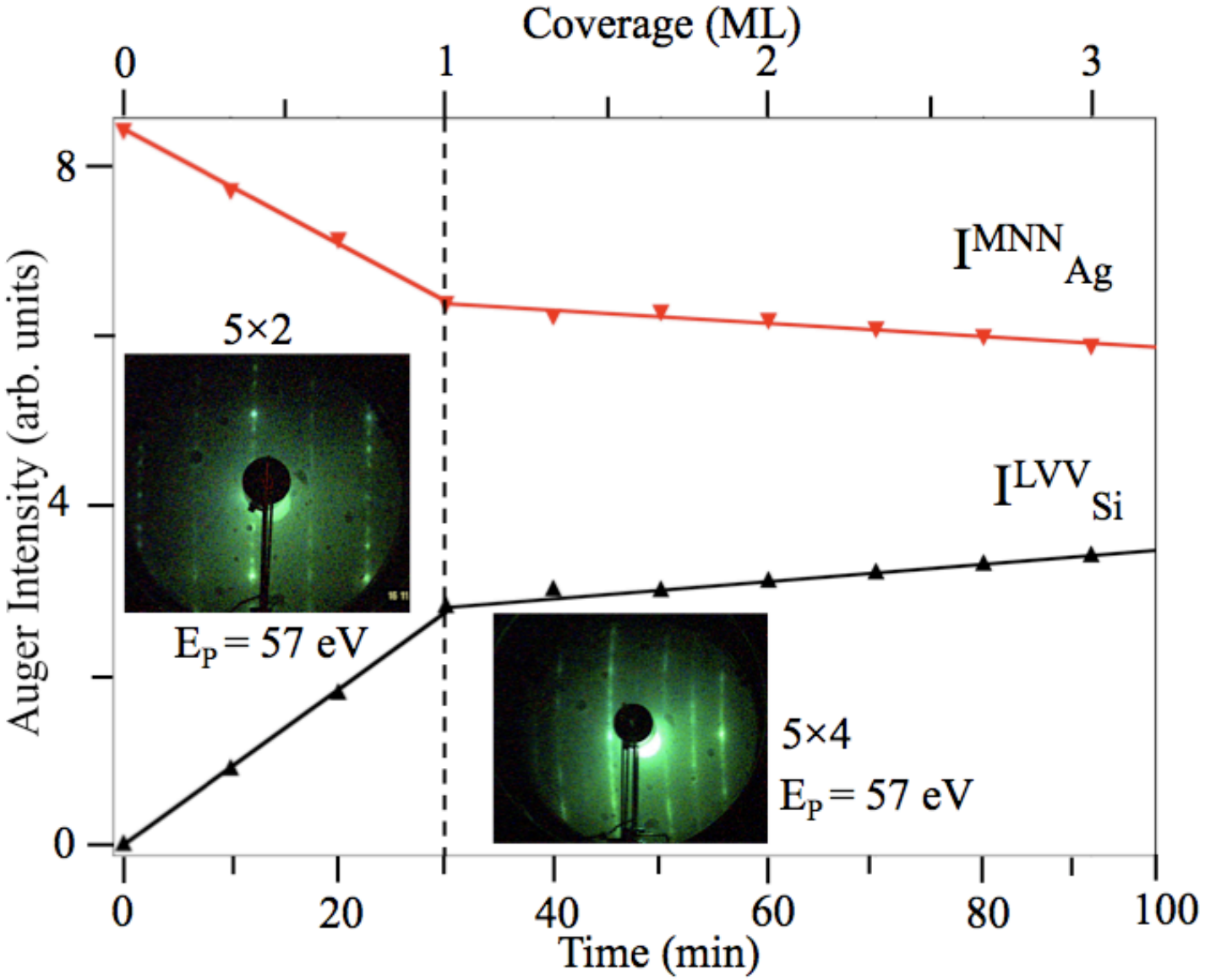






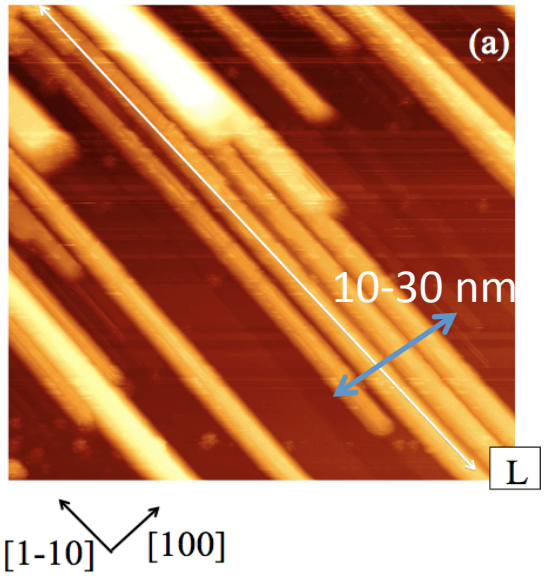
hu = 78 eV

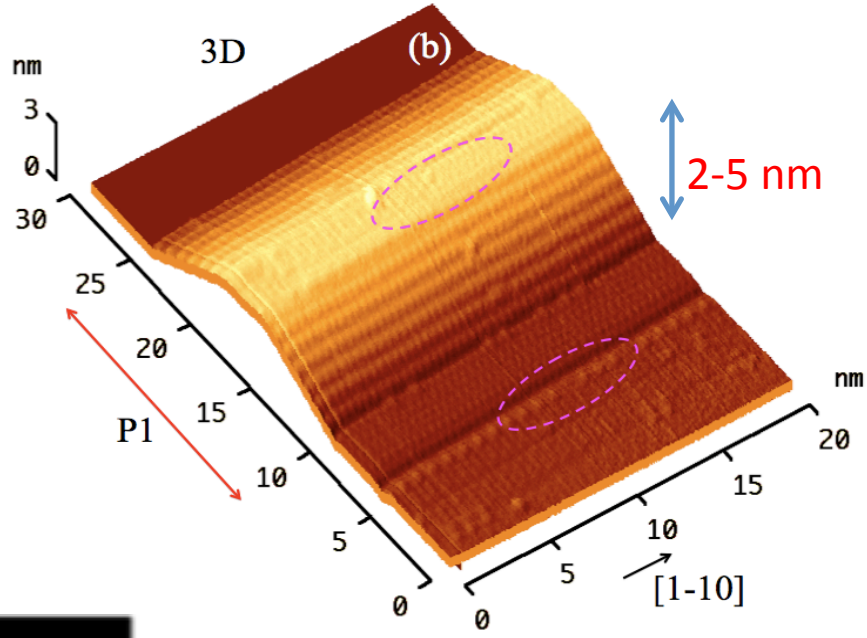
Self-organized 2D Silicene NRs on Ag(110): 2D→3D

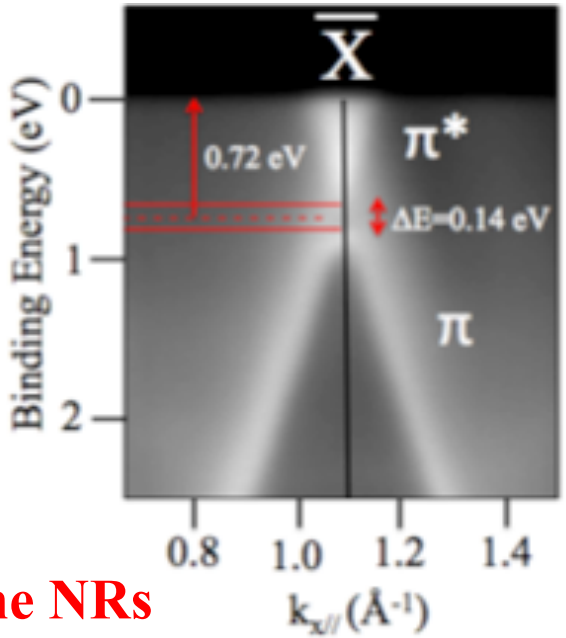


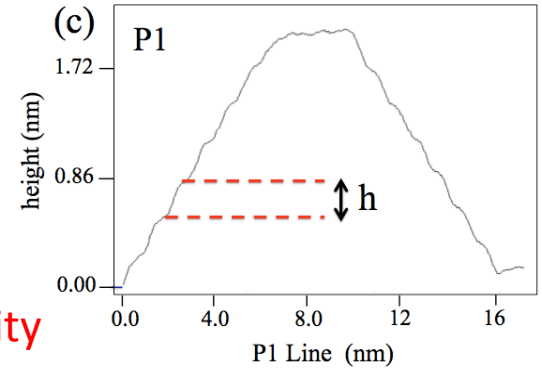
De Padova et al., Nano Letters 2012, **12**, 5500

STM Self-organized 3D Silicene NRs on Ag(110)









Fermi Velocity
≈ 1×10^{6} ms^{-1}
GAP=0.14 eV

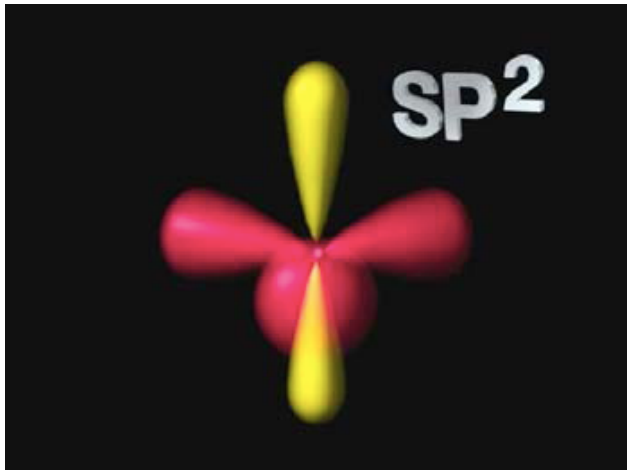
h ≈ 0.29 nm

Γ→K silicene NRs

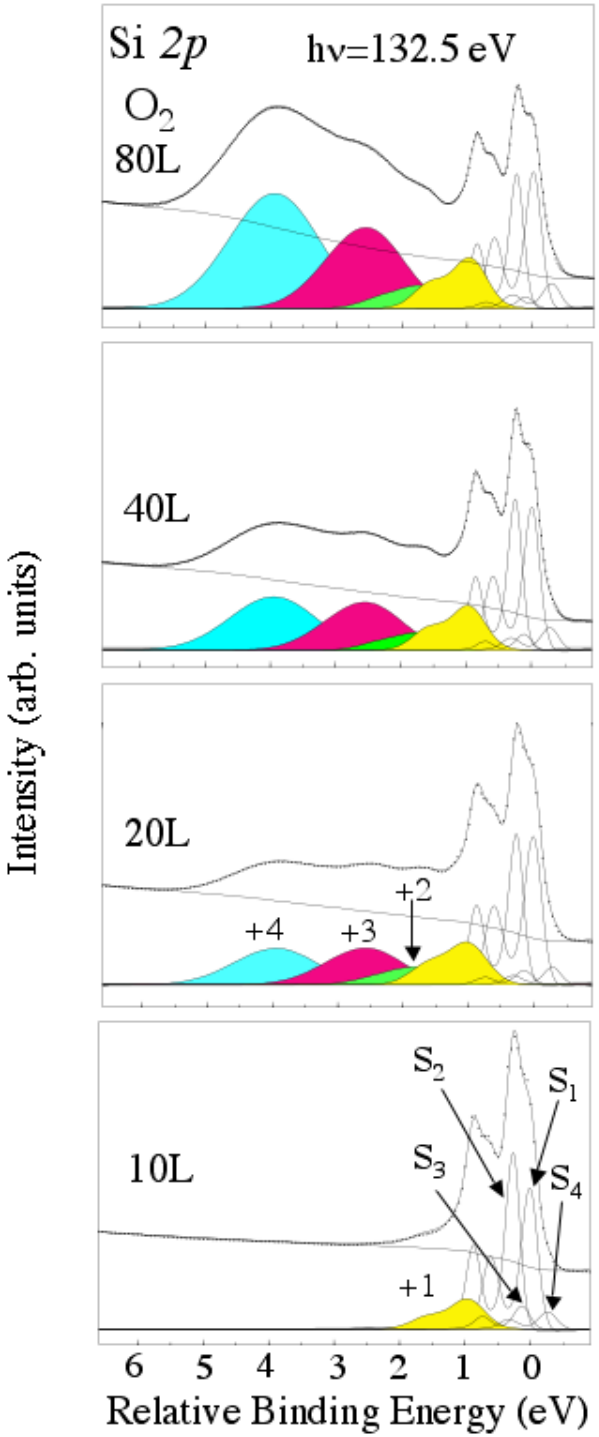
hυ=126 eV

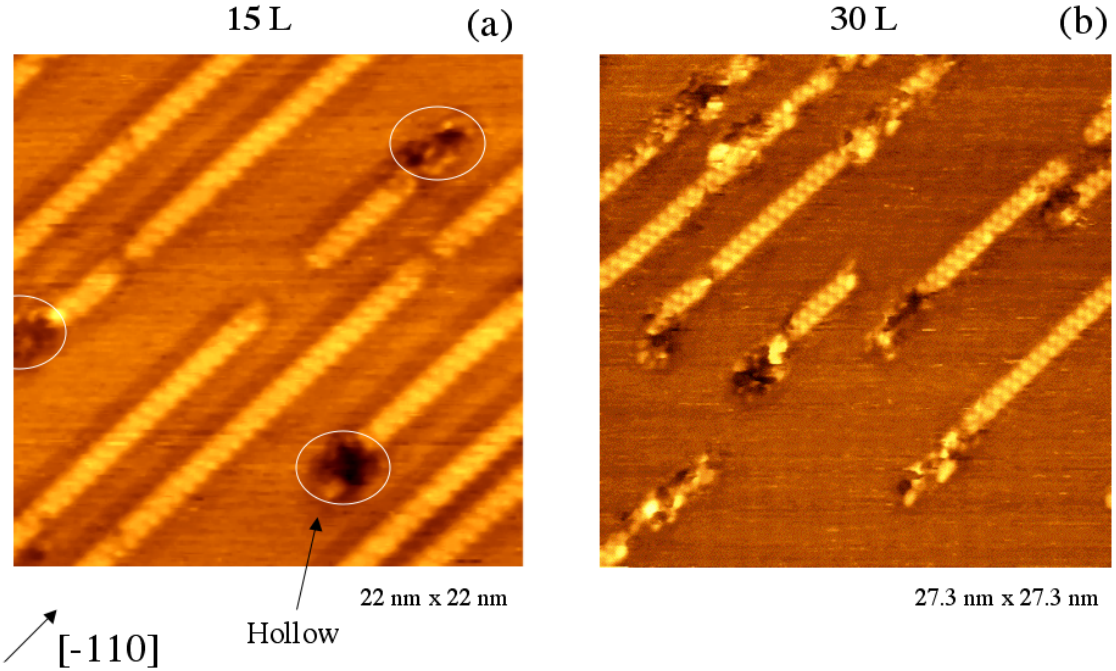
De Padova et al., Nano Letters 2012, **12**, 5500

Chemistry: strong resistance toward oxidation



O_2 on SiNRs/Ag(110) at RT





Filled-state STM images (V=-1.8 V; I=1.2 nA)

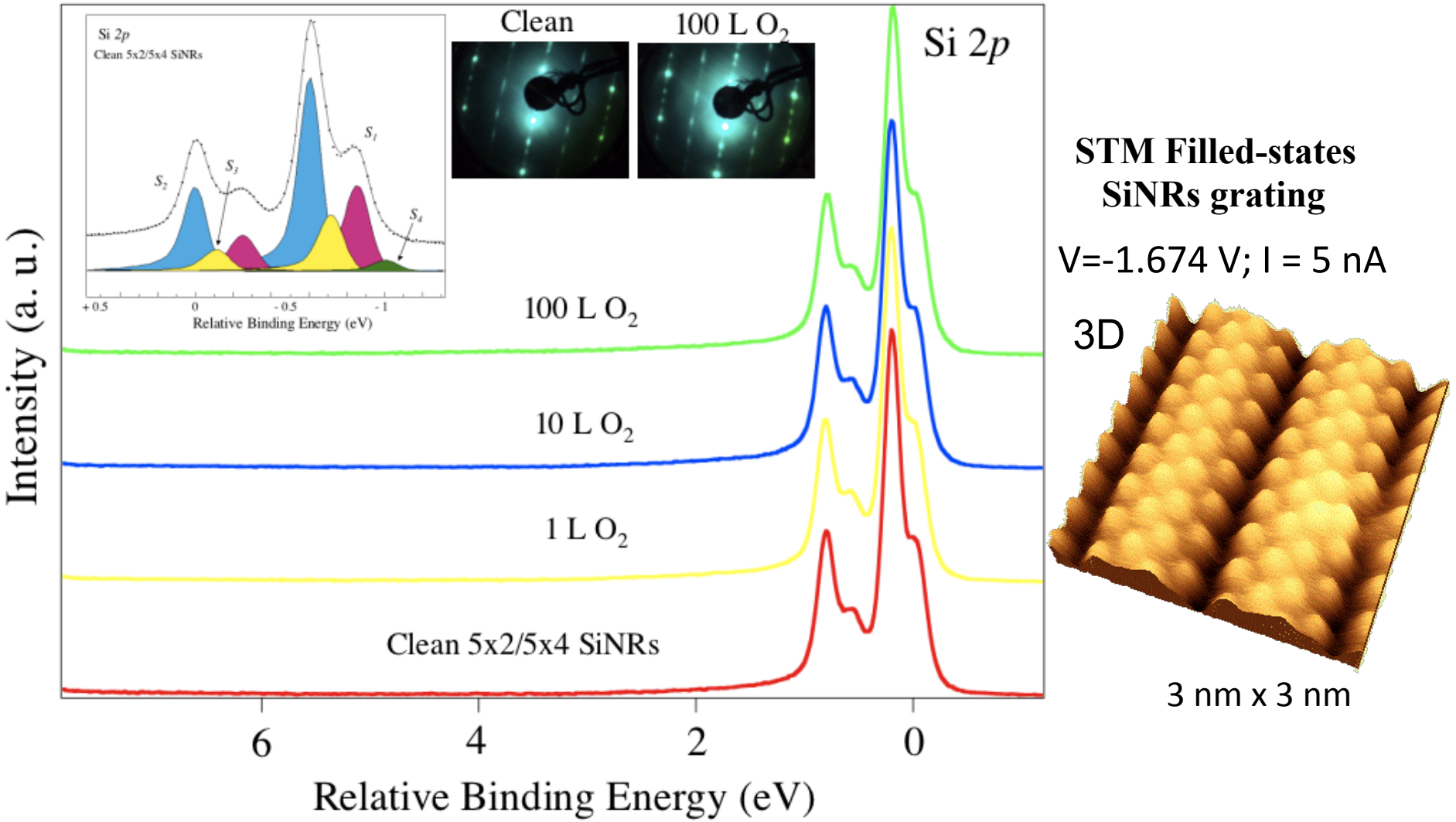
Photoemission Measurements

S^0+

- Si^{4+} → + 3.85 eV
- Si^{3+} → + 2.42 eV
- Si^{2+} → + 1. 71 eV
- Si^{1+} → + 0.95 eV

P. De Padova et al. Nano Lett. 8 (2008) 2299

Strong resistance of silicene nanoribbons towards oxidation



STM Filled-states SiNRs grating

V=-1.674 V; I = 5 nA

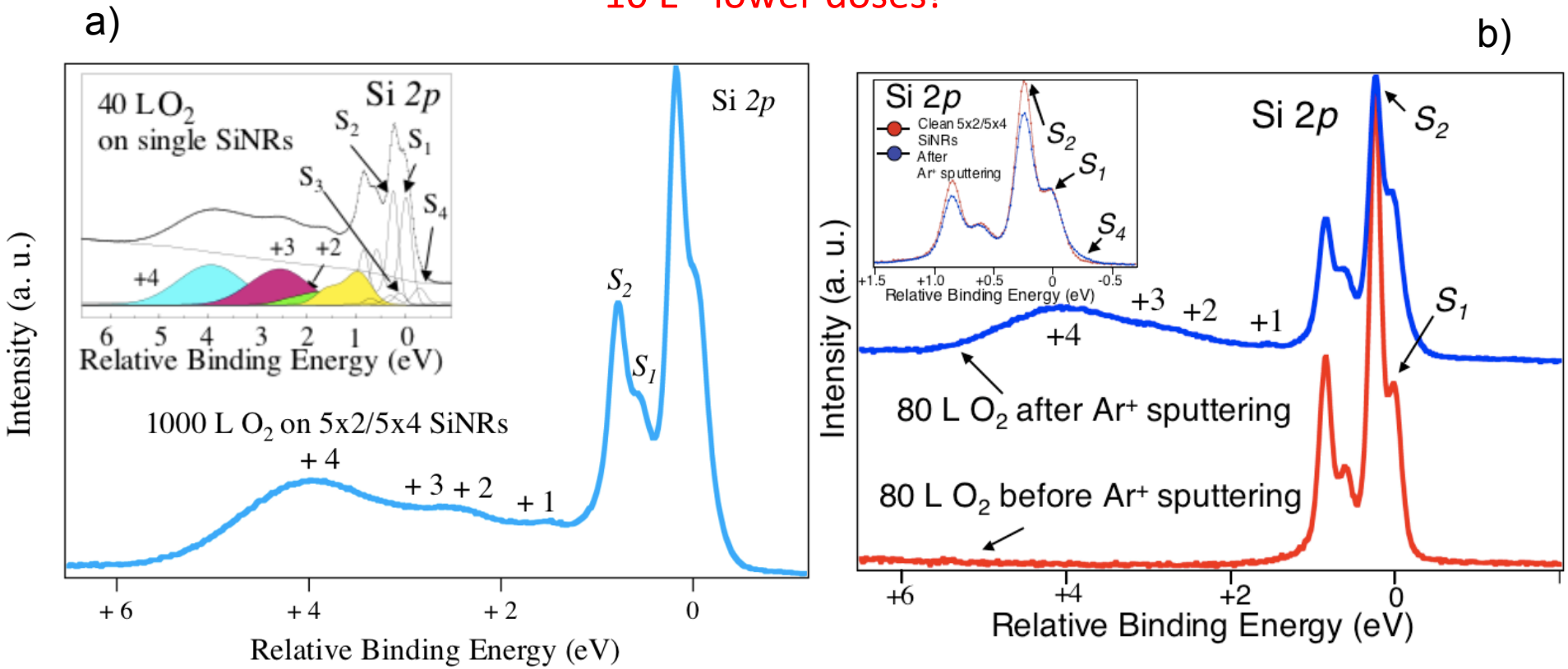
3D

3 nm x 3 nm

Normal emission Si *2p* core-levels from a dense array of silicene NRs on Ag(110) with 5x2/5x4 superstructure and after 1, 10 100 L of oxygen exposures. The inset is the Si *2p* convolution on clean 5x2/5x4 SiNRs. The LEED patterns (E_p= 57 eV) were collected on clean 5x2/5x4 SiNRs and after the oxygen exposition of 100 L.

P. De Padova, C. Quaresima , B. Olivieri, P. Perfetti and G. Le Lay; J. Phys. D: Appl. Phys. 44 (2011) 312001
Fast Track Communication

On Si(100) or Si(111)
oxidation starts at
~10 E^{-4} lower doses!



a) Si *2p* core level of the dense 5x2/5x4 grating after 1000 L of oxygen exposure. On the Si 2*p* spectrum the S_1, S_2 peaks of the virgin part and the oxidation states (+1 to +4) are indicated. Inset: convoluted Si *2p* core level of isolated SiNRs after 40 L of oxygen exposure. The colored components (+1 to +4) related to the different oxidation states (+1 to +4) grow at the expense of the four initial components (S_1, S_2, S_3, S_4) of the clean SiNWs. b) Si 2*p* core level spectrum of the silicene grating with 80 *L* of oxygen exposure, before and after Ar^+ sputtering. Inset: Si 2*p* spectra before (clean spectrum) and after Ar^+ sputtering .

The robustness of the SiNRs toward oxidation compares favourably to the inertness of the SiNTs. It is a strong indication of sp2-like bonding!

2D Si growth on Ag(111)

Ag(110)

unit cell

→

Ag(111)

unit cell

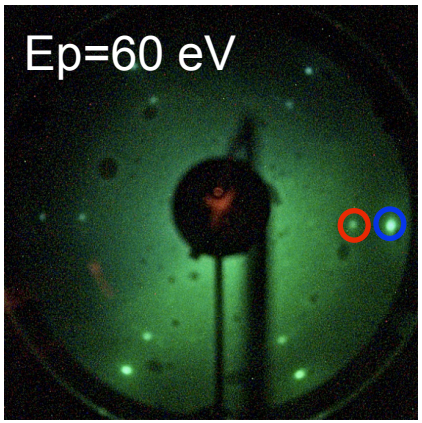
→

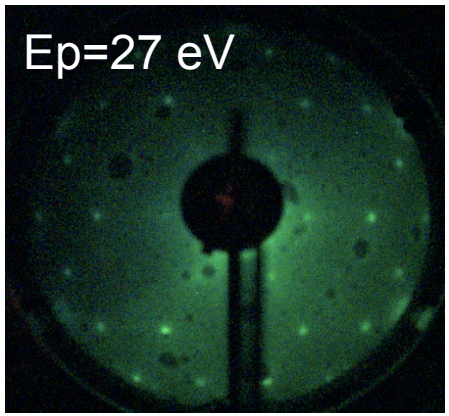
2D Silicene

unit cell

a_{Ag} = 2.89 Å

a_{Si} = 2.23 Å





Ag (1×1) spots

Si 3/4 spots

Si/Ag(111)

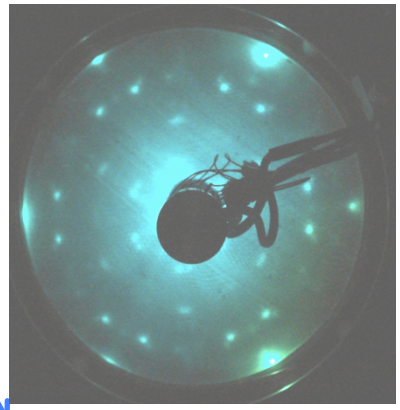
(depending on T and thickness)

Pure $(4\times4)_{Ag}$

$(3\times3)_{Silicene}$

T ≈ 200 °C

LEED[(2√3 x 2√3)R30°



E_P=42 eV

Pure (2√3×2√3)R30°

T ≈ 300 - 400°C

[Si diffusion on Ag(111)]

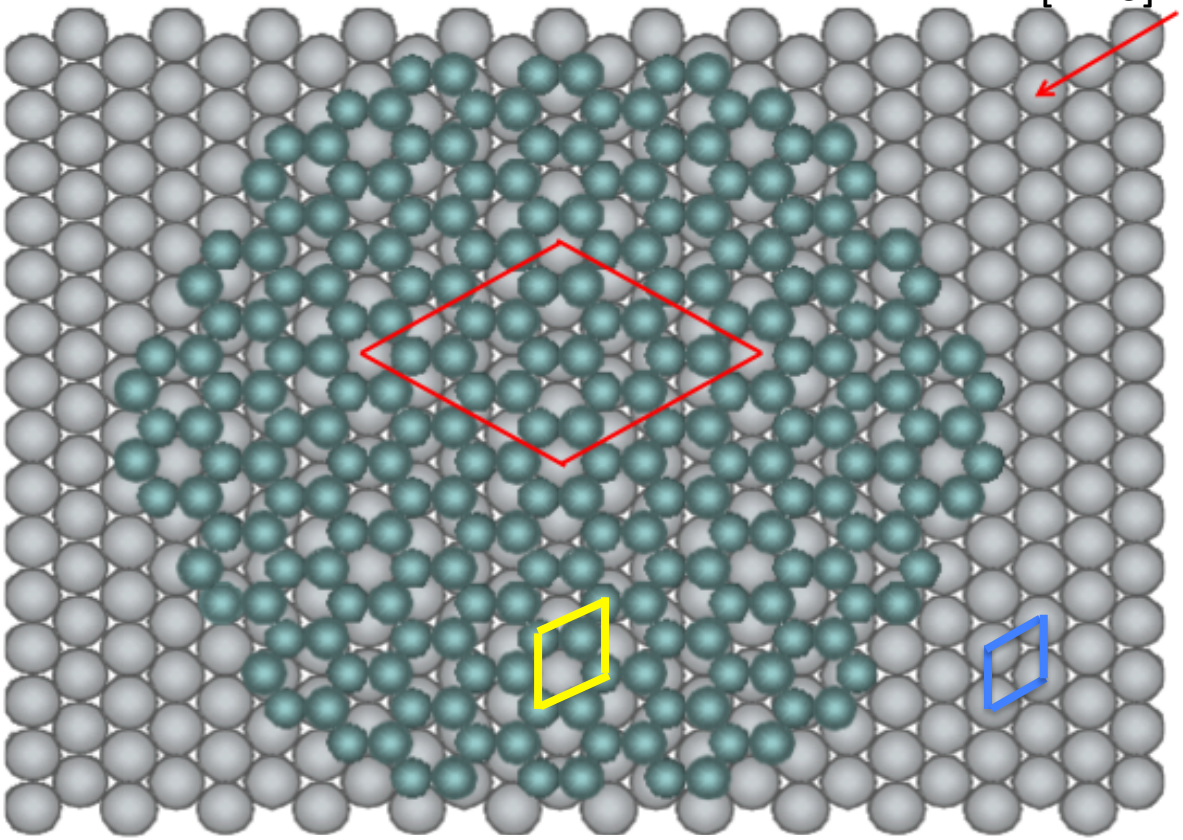
P. Vogt *et al.* PRL, 2012, 101, 155501

Z. L. Liu et al. APL materials 2014, 2, 092513

MODEL of 2D honeycomb Si on Ag(111)

Si-Si=2.23 Å

[-110]



Zig-Zag Silicene // Ag [-110]

Unit cells

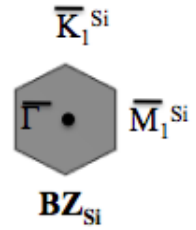
$(1\times1)_{Ag}$

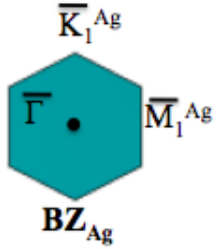
$(1\times1)_{Silicene}$

$(4\times4)^{(respect\ to\ Ag\ 1\times1)}_{Silicene}$

≡

$(3\times3)^{(respect\ to\ Silicene\ 1\times1)}_{Silicene}$





$$\overline{\Gamma K_1}^{Si} = 3/4\,\overline{\Gamma K_1}^{Ag}$$

STM of 2D Si layer and DFT calculations

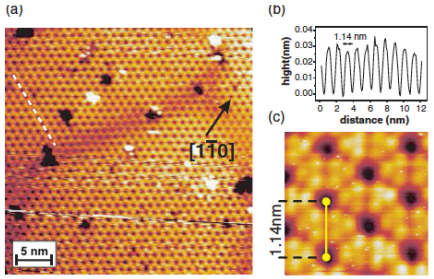
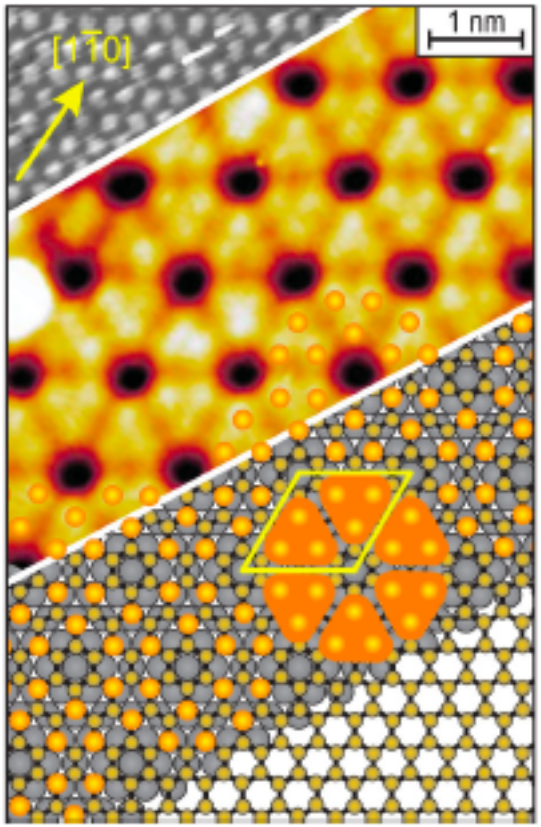


FIG. 2 (color). (a) Filled-states STM image of the 2D Si layer on Ag(111)-(1 × 1) ($U_{bias} = -1.3$ V, $I = 0.35$ nA). Clearly visible is the honeycomblike structure. (b) Line profile along the dashed white line indicated in (a). The dark centers in the STM micrograph are separated by 1.14 nm, corresponding to 4 times the Ag(111) lattice constant, in agreement with the (4 × 4) symmetry. (c) High-resolution STM topograph (3 × 3 nm, $U_{bias} = -1.3$ V, $I = 0.35$ nA) of the Si adlayer.

STM-Ag

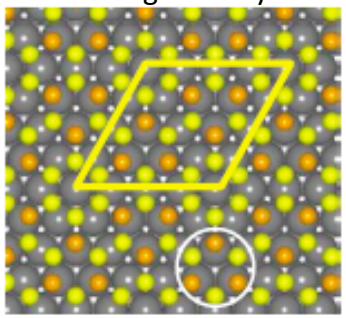


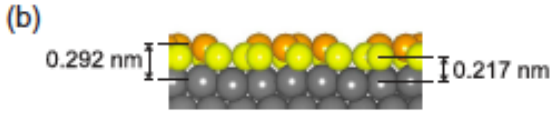
STM-(3×3) Si

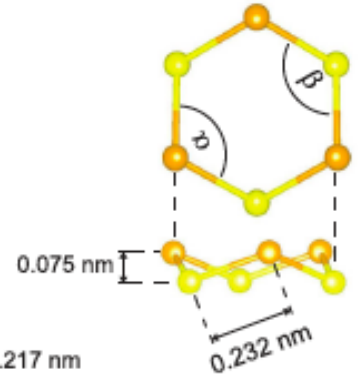
MODEL

DFT Calculations

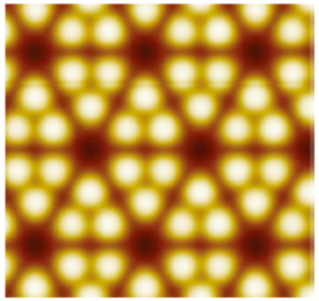
Top and side-view
Fully relaxed (3×3) Si
atomic geometry



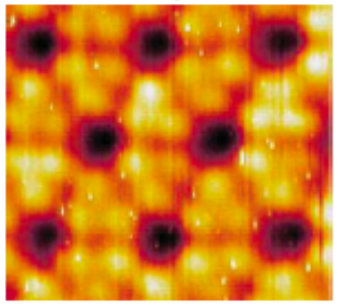




simulated image



measured image



The six top Si atoms of the (3×3) unit cell, bond angles of α= 110 °, which is very close to an angle of 109.5 for an ideally sp3-hybridized Si atom. From the remaining 12 bottom Si atoms, six are purely sp2 hybridized (bond angle β of 120° and six have bond angles of between 112° and 118, indicating a sp3=sp2 hybridization.

P. Vogt, et al. PRL, **2012**, 108, 155501

Beyond single silicene layer: the √3×√3 multilayer silicene on Ag(111)

Theory

Standalone bilayer silicene

R. Hoffmann, PCC., **2013**, 52, 93

M. Ezawa, JPSJ., **2012**, 81, 1047131

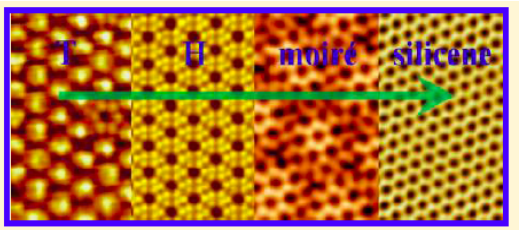
Multilayer silicene

K. Takeda C. and Shiraishi, PRB **1994**, 50, 14916

Kamal *et al.*, JPCM., **2013**, 25, 085508-1

S. Cahangirov PRB 2014, 90, 035448

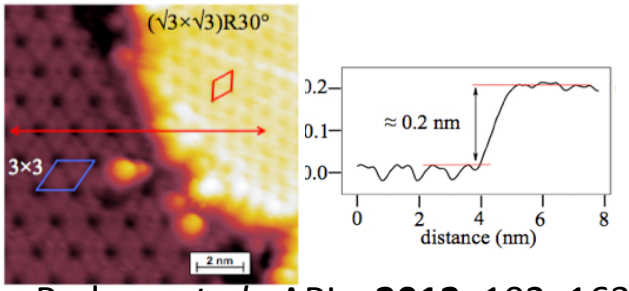
√3×√3-1×1 Ag(111) silicene



L. Chen *et al.*, PRL., **2012**, 102, 056804-1;

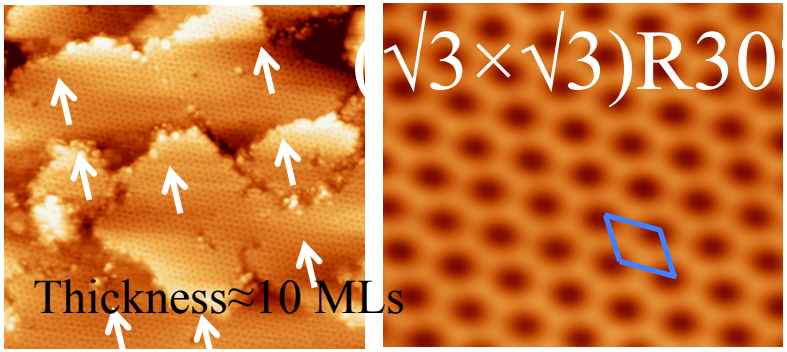
B. Feng *et al.*, Nano Lett. **2012**, 11, 3507.

V=-1.1 V; I=0.33 nA



P. De Padova *et al.*, APL., **2013**, 102, 163106;

P. De Padova *et al.*, JPCM., **2013**, 25, 382202



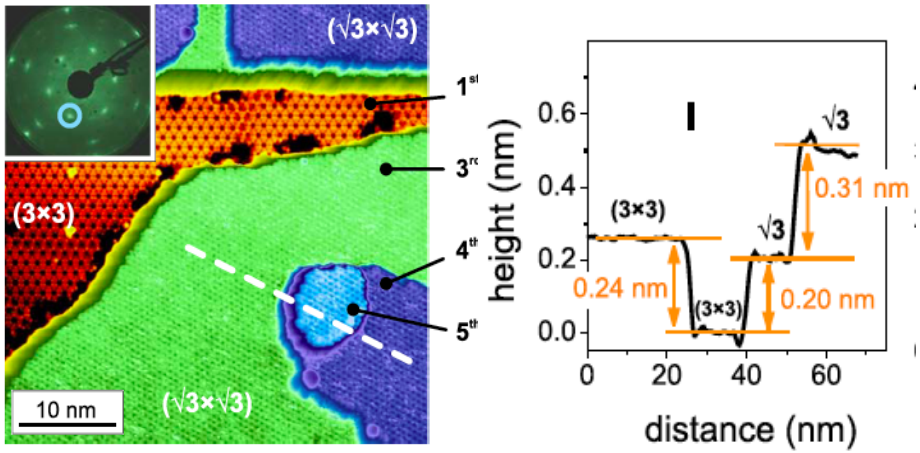
40 nm×40 nm V=-0.1 V; I=0.16 nA 5 nm×5 nm

P. De Padova *et al.*, 2D Materials, 1 (2014) 021003;

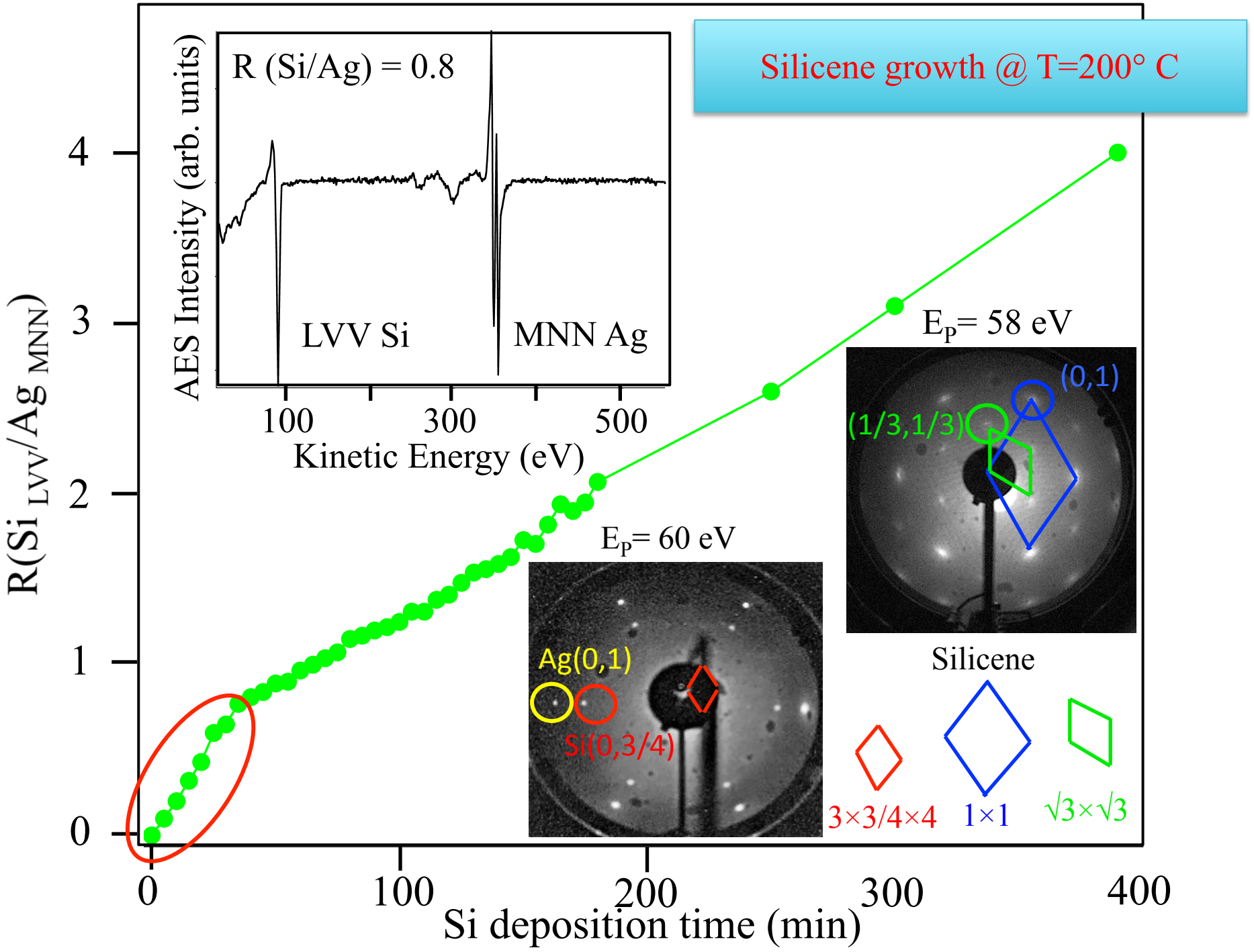
E. Salomon *et al.*, JPCM., **2014**, 7, 185003.

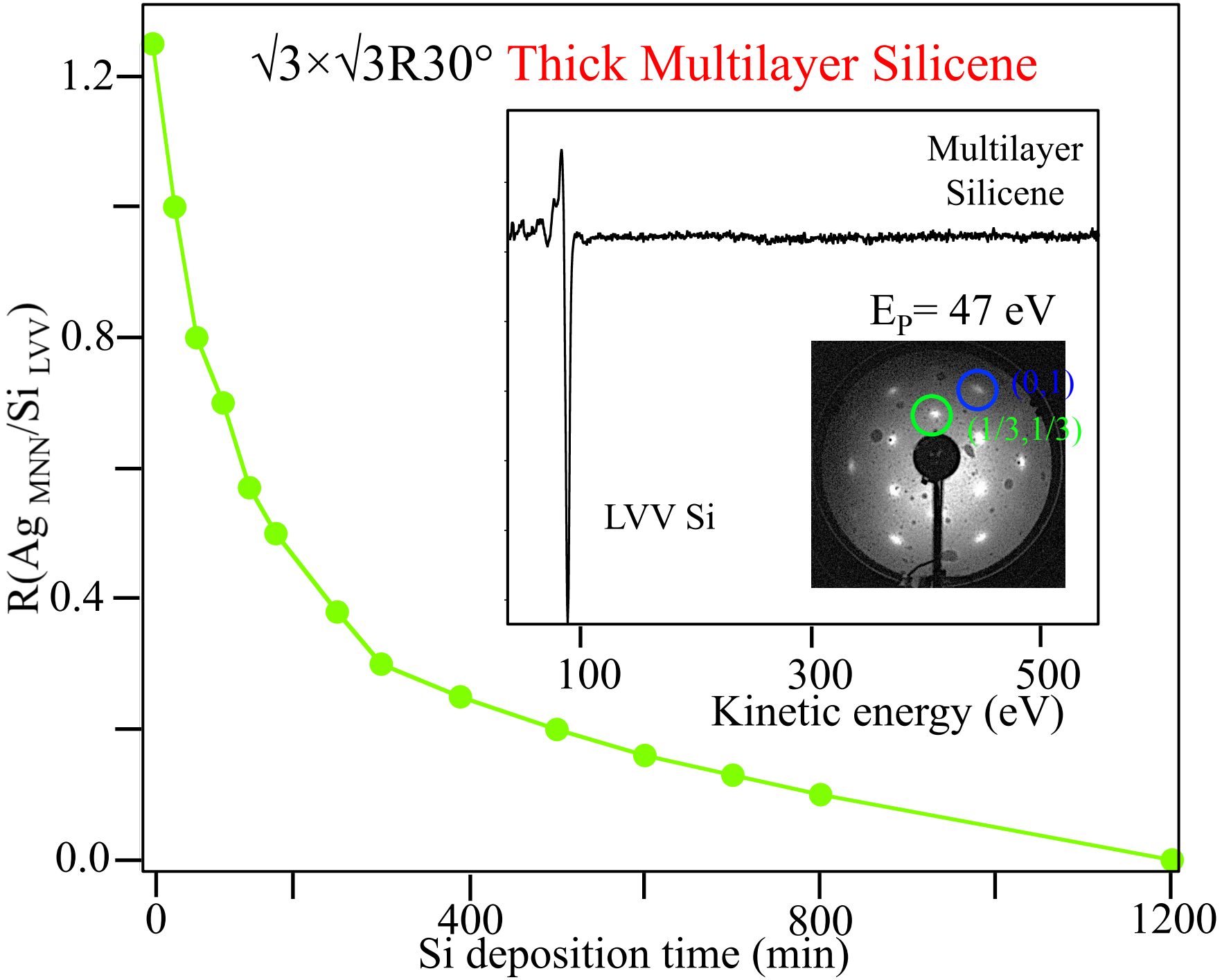
√3×√3 multilayer silicene on Ag(111)

V=-1.1 V; I=0.54 nA

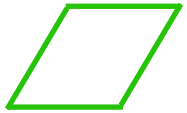


P. Vogt *et al.*, APL., **2014**, 104, 021602.


Silicene growth @ T=200° C
R (Si/Ag) = 0.8
AES Intensity (arb. units)
LVV Si
MNN Ag
100
300
500
Kinetic Energy (eV)
R(Si LVV/Ag MNN)
0
1
2
3
4
0
100
200
300
400
Si deposition time (min)
E_P= 58 eV
(0,1)
(1/3,1/3)
E_P= 60 eV
Ag(0,1)
Si(0,3/4)
Silicene
3×3/4×4
1×1
√3×√3


√3×√3R30° Thick Multilayer Silicene
1.2
0.8
0.4
0.0
R(Ag MNN/Si LVV)
0
400
800
1200
Si deposition time (min)
Multilayer
Silicene
E_P= 47 eV
(0,1)
(1/3,1/3)
LVV Si
100
300
500
Kinetic energy (eV)

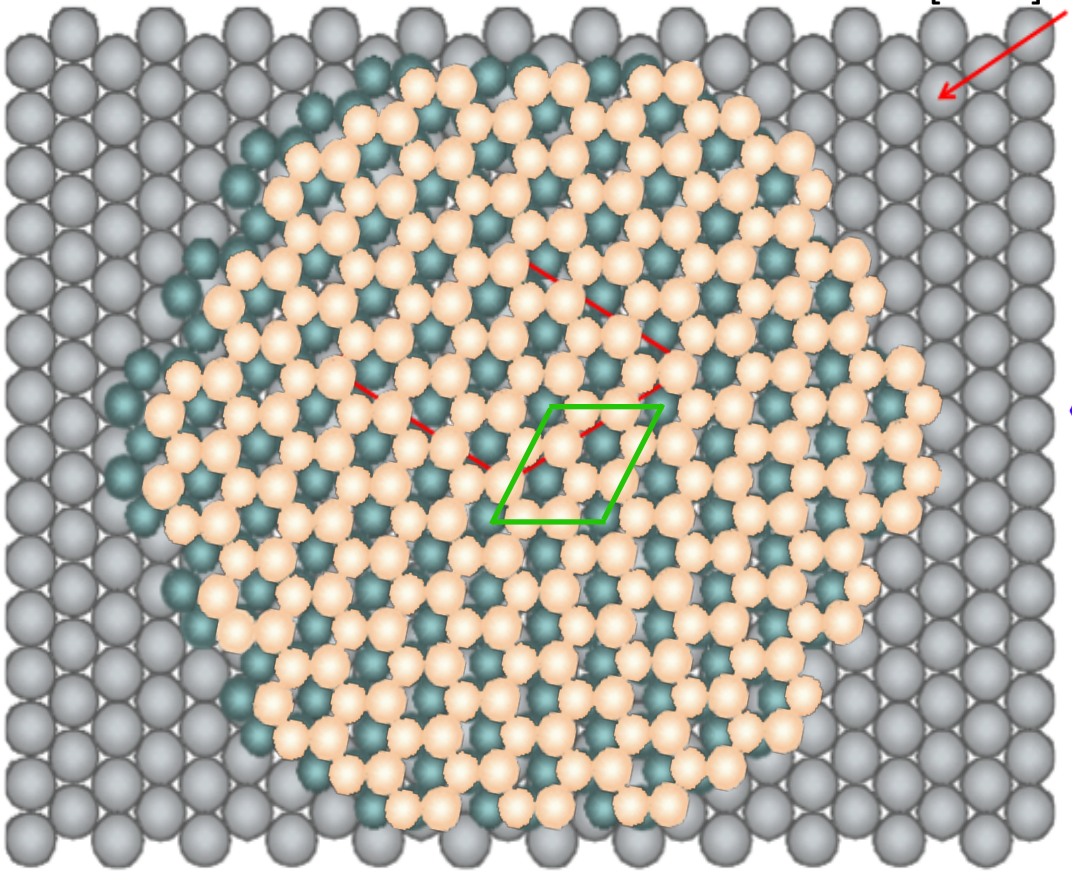
MODEL: 2nd Si layer----on top of first Si 3×3 Layer



$(\sqrt{3}\times\sqrt{3})_{\text{Silicene}}$ (respect to Silicene 1×1)

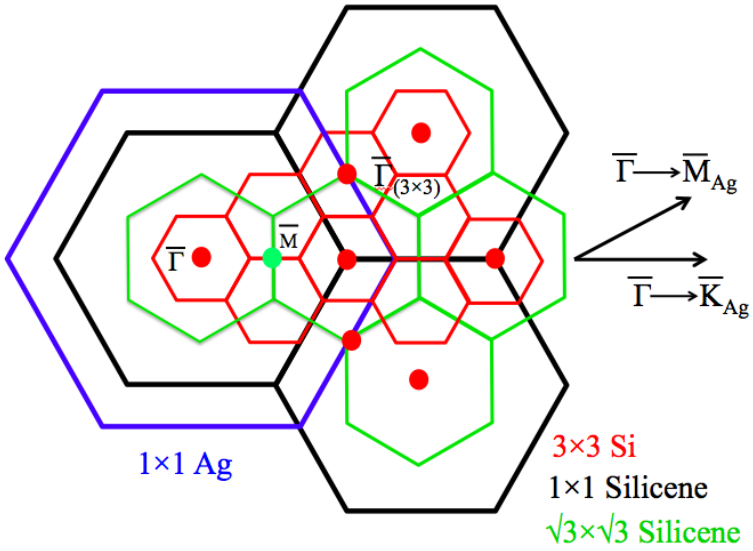
Si-Si=2.23 Å

[-110]

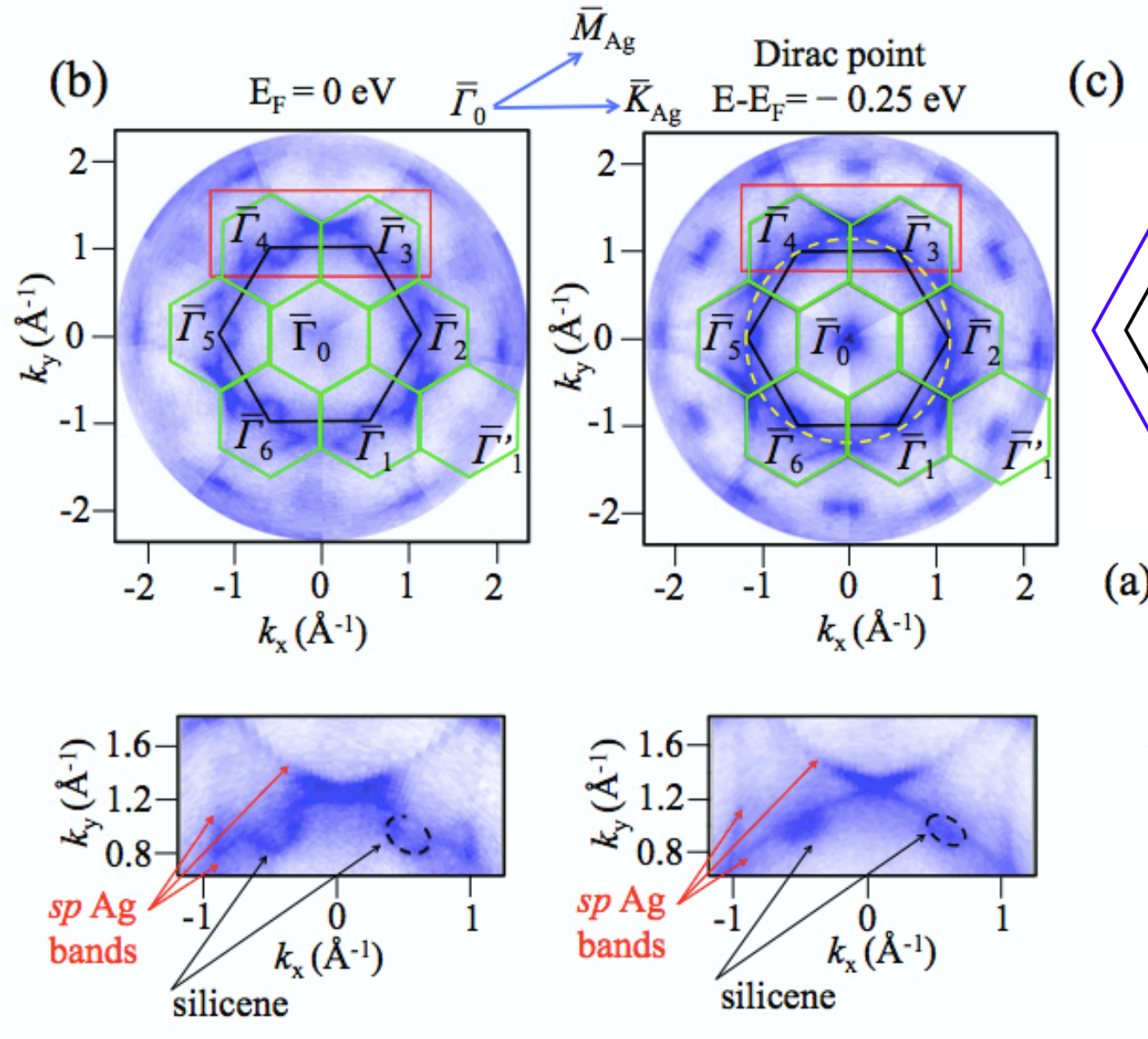


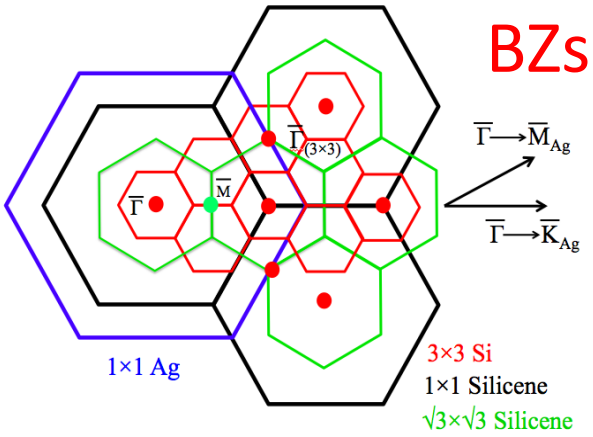
Zig-Zag Silicene // Ag [-110]

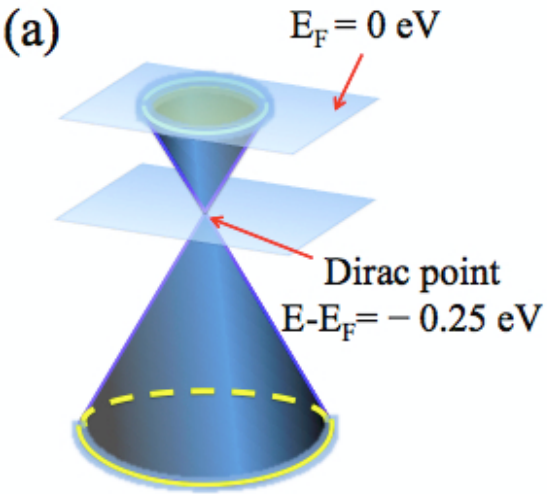
BZs



Energy Contour Section on √3×√3 Silicene/Ag(111) (1.5 MLs)

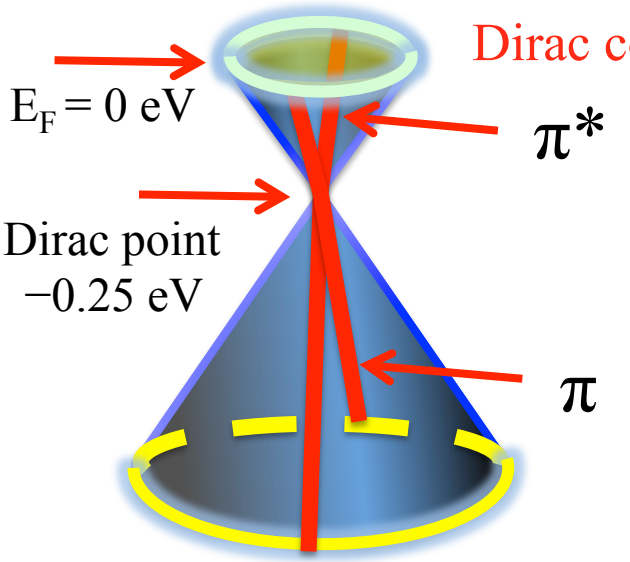


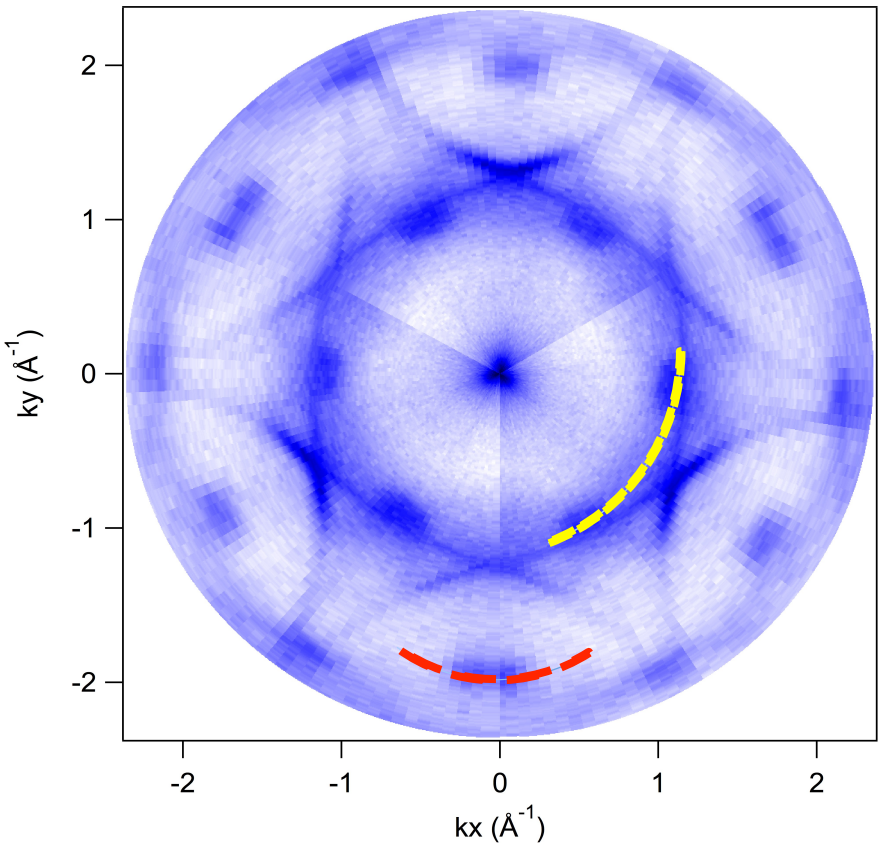




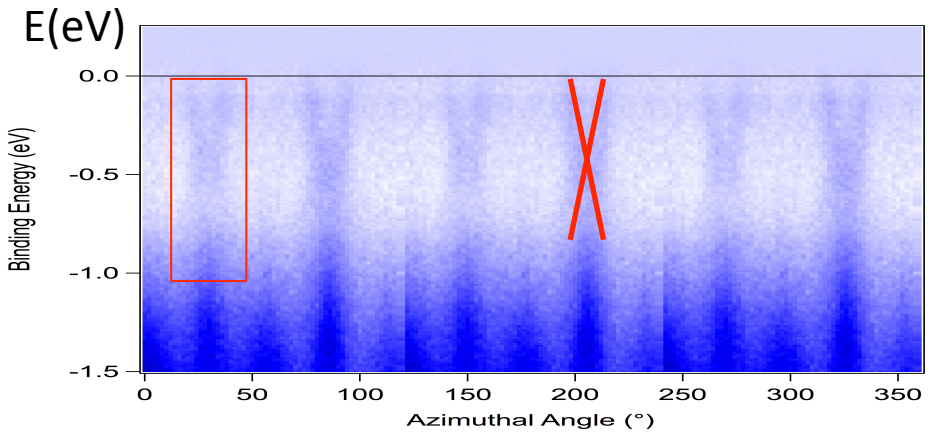
P. De Padova *et al.*, JPCM., **2013**, 25, 382202.

Dirac cone Vertical Section on √3×√3 Silicene/Ag(111) (1.5 MLs)

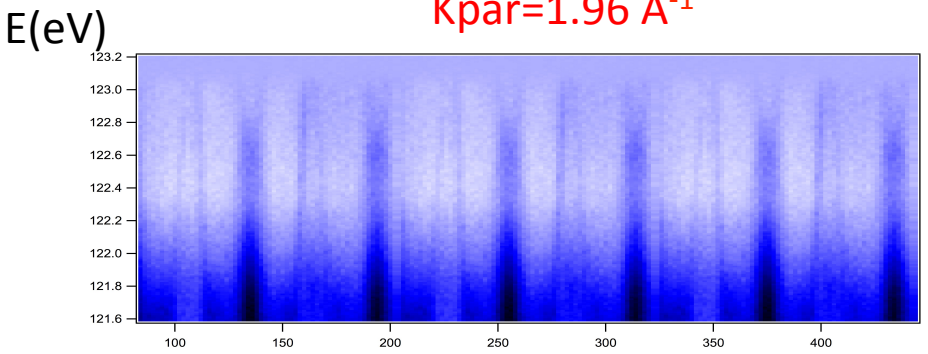




Kpar=1.09Å^{-1}



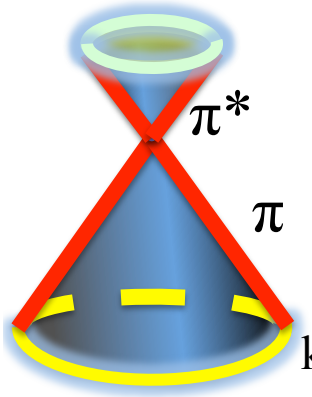
Kpar=1.96 Å^{-1}

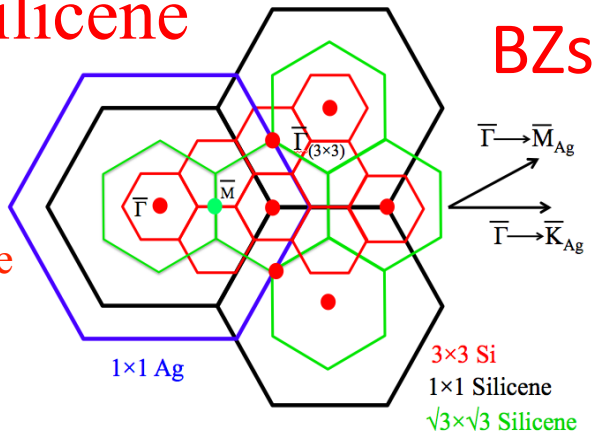


P. De Padova *et al.*, JPCM., **2013**, 25, 382202.

ANTARES-SOLEIL

Dirac cone of 2D multilayer Silicene (4.5 MLs)

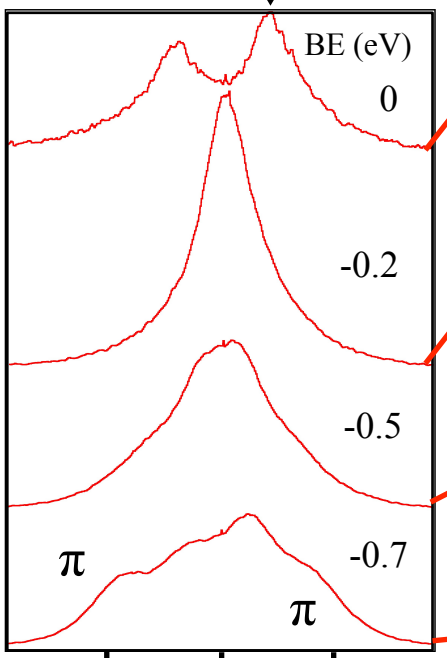


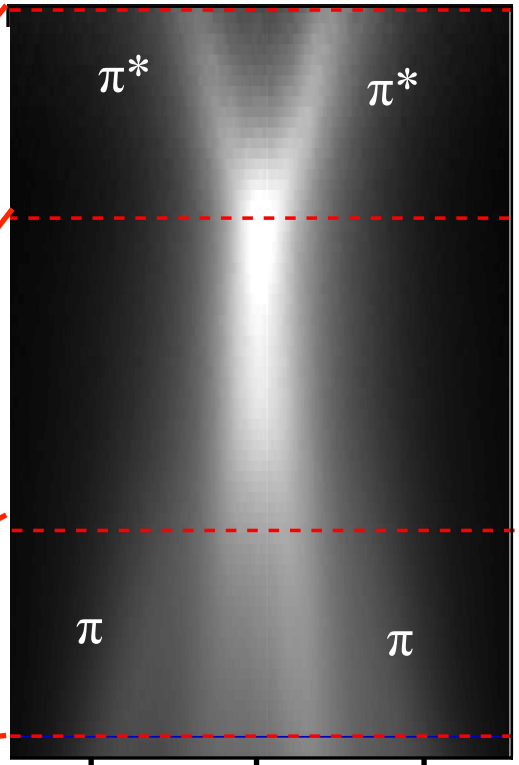


E_F= 0 eV

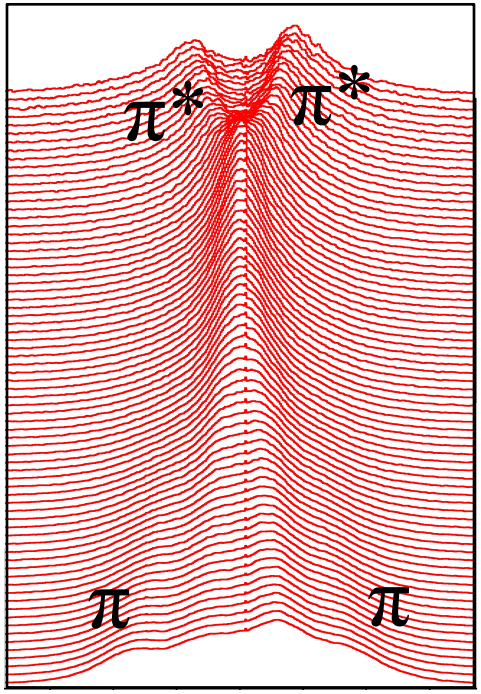
k_F=0.2 Å^{-1}

$\overline{\Gamma}$ √3×√3 Silicene





0.5



-0.4 0.0 0.4

$k_{//}$ (Å^{-1})

-0.4 0.0 0.4

$k_{//}$ (Å^{-1})

$\overline{\Gamma}\rightarrow\overline{K}_{silicene}$

$V_F \approx 0.3\ 10^{6}$ m sec^{-1}

Dirac point ≈ - 0.25 eV

No GAP!

P. De Padova *et al.*, APL., **2013**, 102, 163106;

Multilayer silicene or Ordinary Si(111) √3×√3-Ag reconstruction?

Silicon Growth at the Two-Dimensional Limit on Ag(111)

Andrew J. Mannix,[†,‡,∥] Brian Kiraly,[†,‡,∥] Brandon L. Fisher,[†] Mark C. Hersam,[‡,§,*] and Nathan P. Guisinger[†,*]

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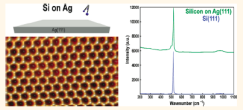
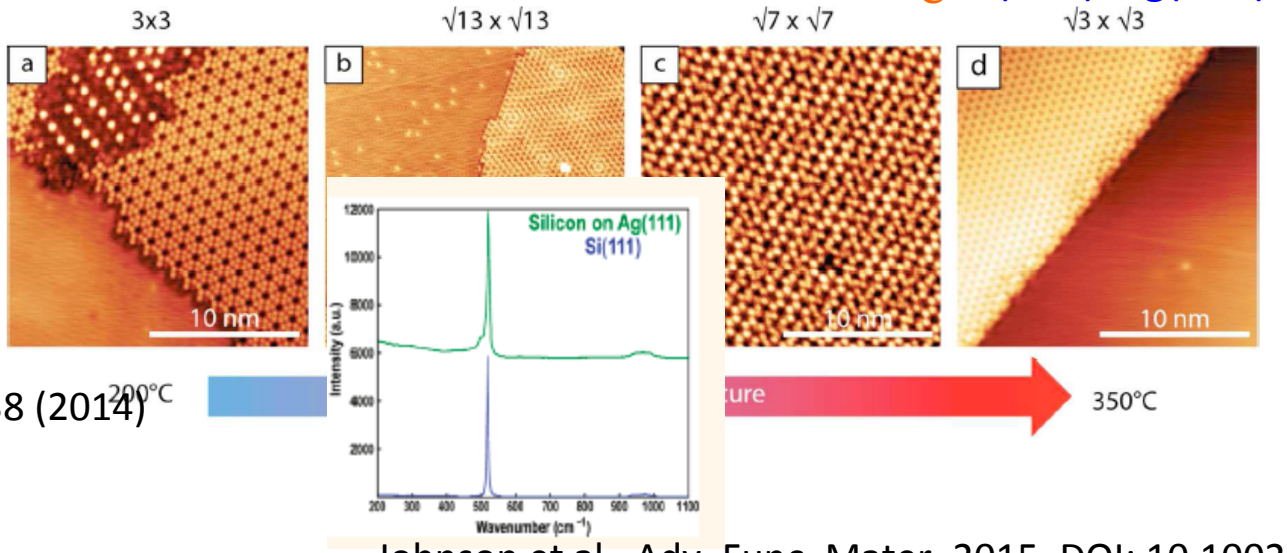
ABSTRACT Having fueled the microelectronics industry for over 50 years, silicon is arguably the most studied and influential semiconductor. With the recent emergence of two-dimensional (2D) materials (*e.g.*, graphene, MoS_2, phosphorene, *etc.*), it is natural to contemplate the behavior of Si in the 2D limit. Guided by atomic-scale studies utilizing ultrahigh vacuum (UHV), scanning tunneling microscopy (STM), and spectroscopy (STS), we have investigated the 2D limits of Si growth on Ag(111). In contrast to previous reports of a distinct sp^2-bonded silicene allotrope, we observe the evolution of apparent surface alloys (ordered 2D silicon−Ag surface phases), which culminate in the precipitation of crystalline, sp^3-bonded Si(111) nanosheets. These nanosheets are capped with a $\sqrt{3}$ honeycomb phase that is isostructural to a $\sqrt{3}$ honeycomb-chained-trimer (HCT) reconstruction of Ag on Si(111). Further investigations reveal evidence for silicon intermixing with the Ag(111) substrate followed by surface precipitation of crystalline, sp^3-bonded silicon nanosheets. These conclusions are corroborated by *ex situ* atomic force microscopy (AFM), transmission electron microscopy (TEM), Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS). Even at the 2D limit, scanning tunneling spectroscopy shows that the sp^2-bonded silicon nanosheets exhibit semiconducting electronic properties.



Munnix et al. , ACS NANO 8, 7538 (2014)

√3×√3-Ag-Si(111)/Ag(111)



Johnson et al., Adv. Func. Mater. 2015, DOI: 10.1002/ADFM.201501029

PHYSICAL REVIEW B **89**, 241403(R) (2014)

Structure determination of multilayer silicene grown on Ag(111) films by electron diffraction: Evidence for Ag segregation at the surface

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[1]*Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan*

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(Received 2 May 2014; revised manuscript received 28 May 2014; published 10 June 2014)

The structure of multilayer silicene formed on Ag(111) films was studied by low-energy electron diffraction. It turned out that the experimental data cannot be explained by proposed models that only consider buckling of silicon atoms. We have rather found that multilayer silicene on Ag(111) is actually a thin film of bulklike silicon terminated with the Si(111)$\sqrt{3} \times \sqrt{3}$-Ag surface. The results are compared to previous works and clearly show the importance to properly understand the structure of the system when discussing its electronic properties.

J. Chen Sci Rep. DOI:101038/Srep/13590

Stability and Electronic Characteristics of Epitaxial Silicene Multilayers on Ag(111)

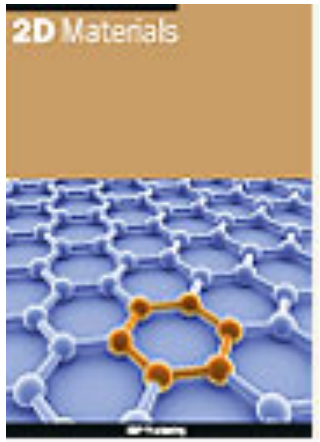
Neil W. Johnson, David Muir, Ernst Z. Kurmaev, and Alexander Moewes*

In this study, the stability and electronic characteristics of epitaxial silicene bilayers and multilayers on the Ag(111) surface are investigated through synchrotron-based soft X-ray emission and absorption spectroscopy and first-principles, full-potential density functional theory simulations. The calculations predict a novel tristable AA-stacked bilayer structure that can explain the (√3 × √3)R30° honeycomb topography commonly observed through scanning tunneling microscopy and noncontact atomic force microscopy. It is reported that the electronic structure of this epitaxial bilayer is similar to those of epitaxial monolayers on Ag(111), namely, metallic and showing significant interaction with the underlying substrate. However, the soft X-ray spectroscopy experiments suggest that during multilayer growth a majority of the epitaxial silicon reverts to a bulk-like state, a result that has significant implications toward the existence of large-area epitaxial silicene multilayers.

2D Materials
1 (2014) 021003

Letter

24 h stability of thick multilayer silicene in air



Paola De Padova[1], Carlo Ottaviani[1], Claudio Quaresima[1], Bruno Olivieri[2], Patrizia Imperatori[3], Eric Salomon[4], Thierry Angot[4], Lucia Quagliano[5,6], Claudia Romano[6], Alessandro Vona[6], Maurizio Muniz-Miranda[7], Amanda Generosi[1], Barbara Paci[1] and Guy Le Lay[1,4]

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doi:10.1088/2053-1583/0/0/000000

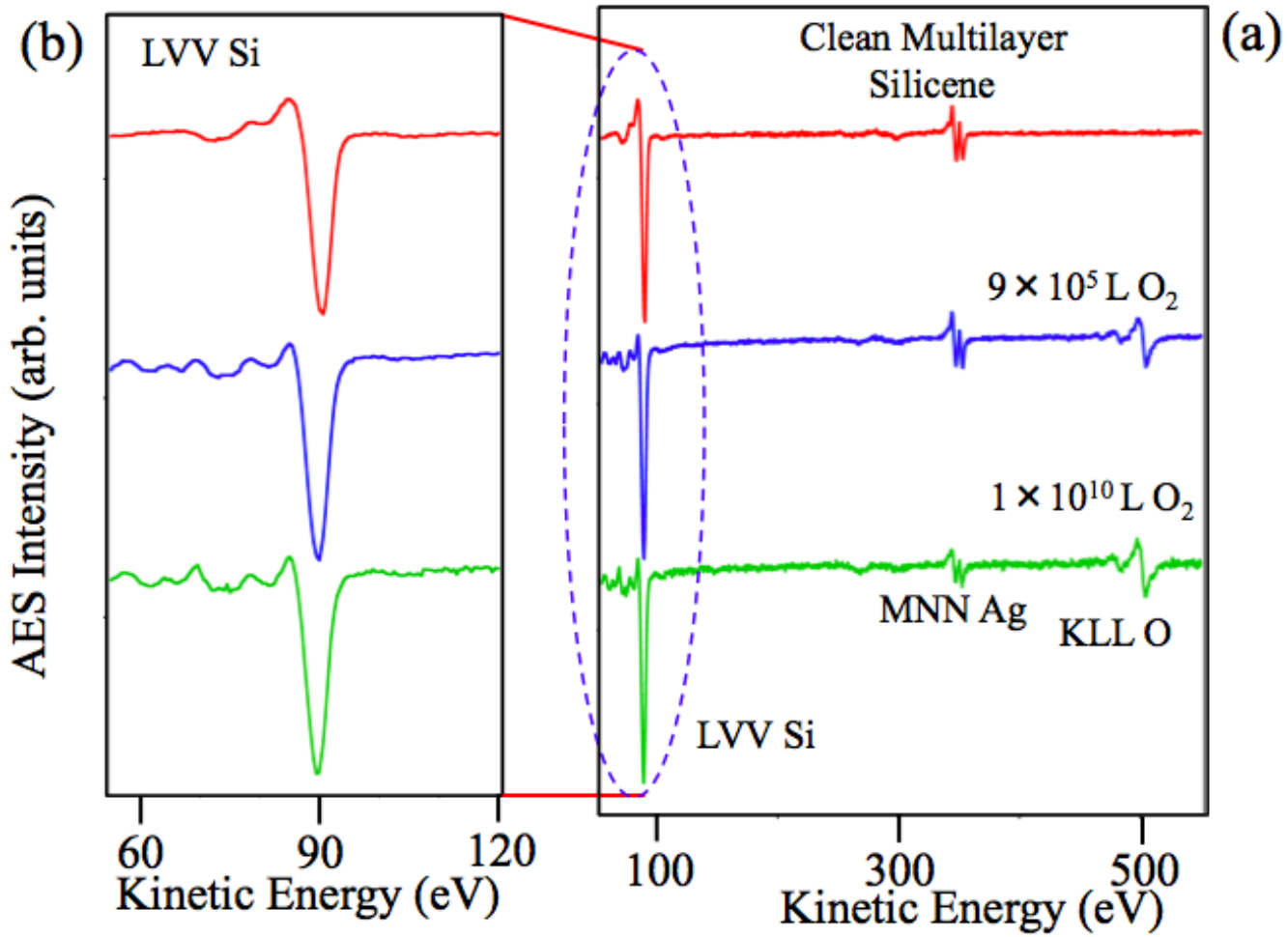
IOP | 2D Materials

Abstract

Thick epitaxial multilayer silicene films with a $\sqrt{3} \times \sqrt{3}R(30°)$ surface structure show only mild surface oxidation after 24 h in air, as measured by Auger electron spectroscopy. X-ray diffraction and Raman spectroscopy measurements performed in air *without any protective capping*, as well as, for comparison, with a thin Al_2O_3 cap, showed the (002) reflection and the G, D and 2D Raman structures, which are unique fingerprints of thick multilayer silicene.

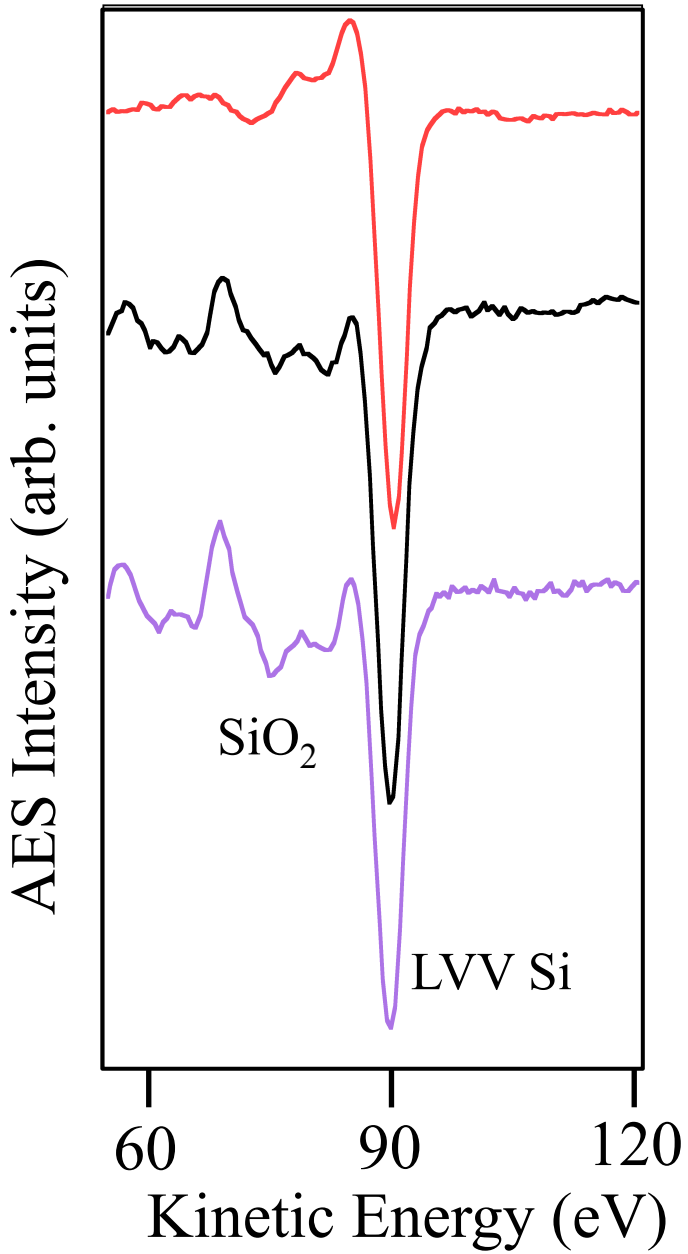
$\sqrt{3}\times\sqrt{3}R30°$ silicene

Pure O_2 exposure in UHV: No Oxidation

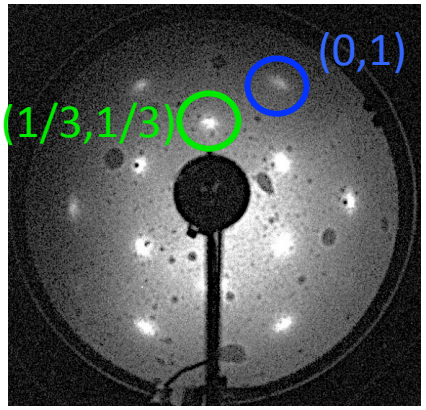


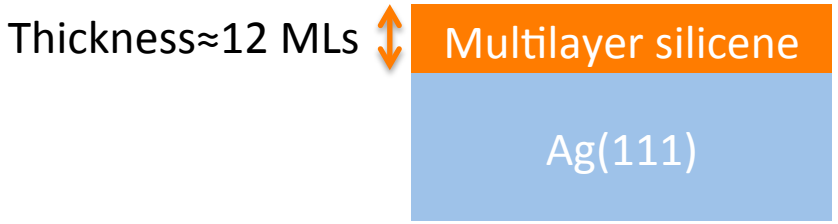
P. De Padova *et al.*, 2D Materials, 1 (2014) 021003

Strong resistance towards oxidation of $\sqrt{3}\times\sqrt{3}R30°$ multilayer silicene



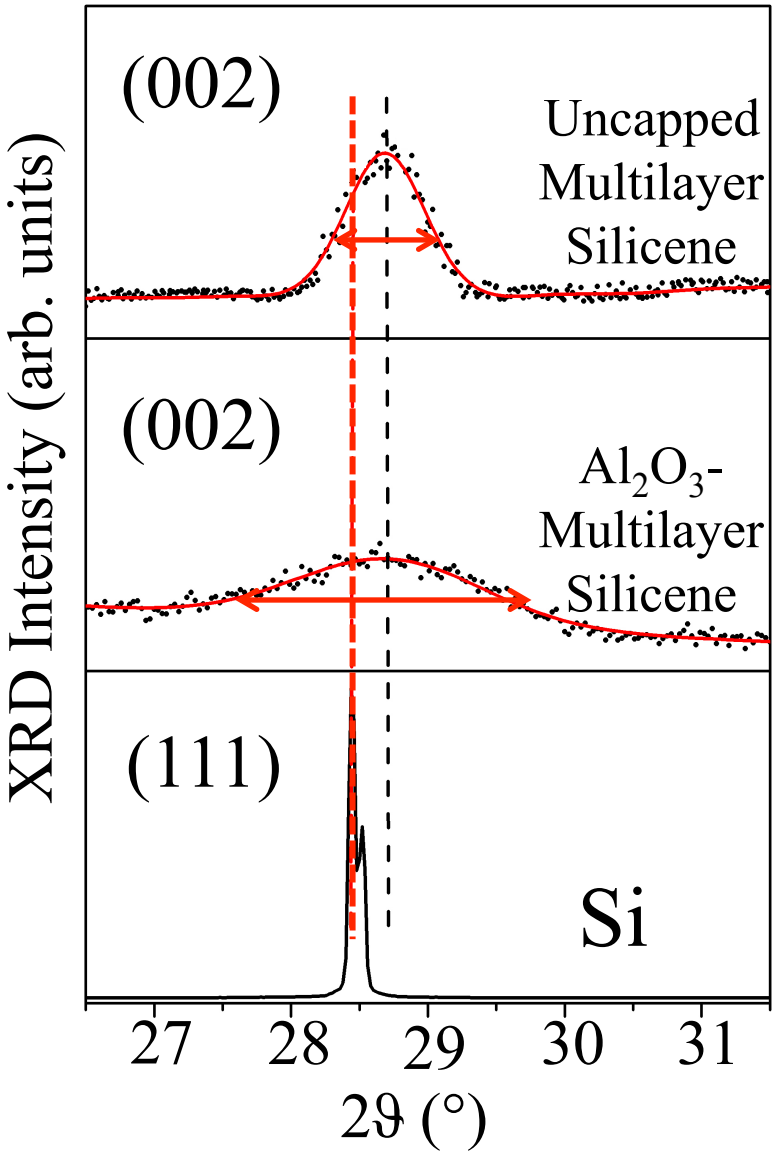
E_P= 47 eV

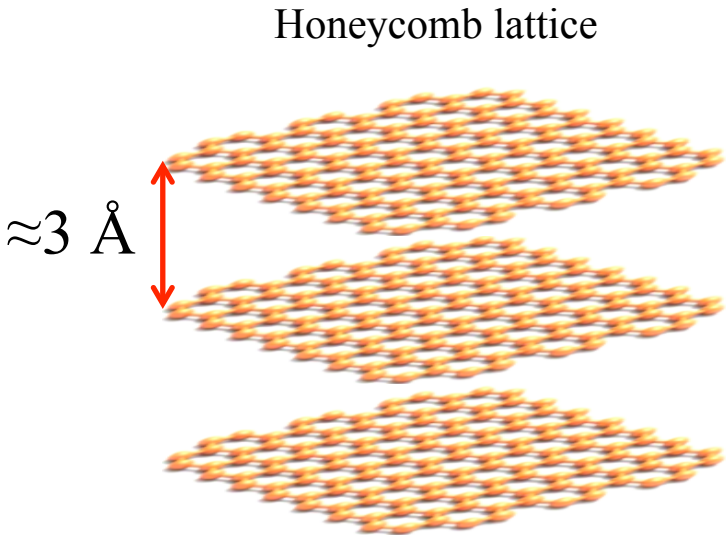




P. De Padova *et al.*, 2D Materials, 1 (2014) 021003

XRD on √3×√3R30° thick multilayer silicene





Scherrer's equation
$(0.9 \bullet \lambda / FWHM_{rad} \bullet \cos\vartheta)$

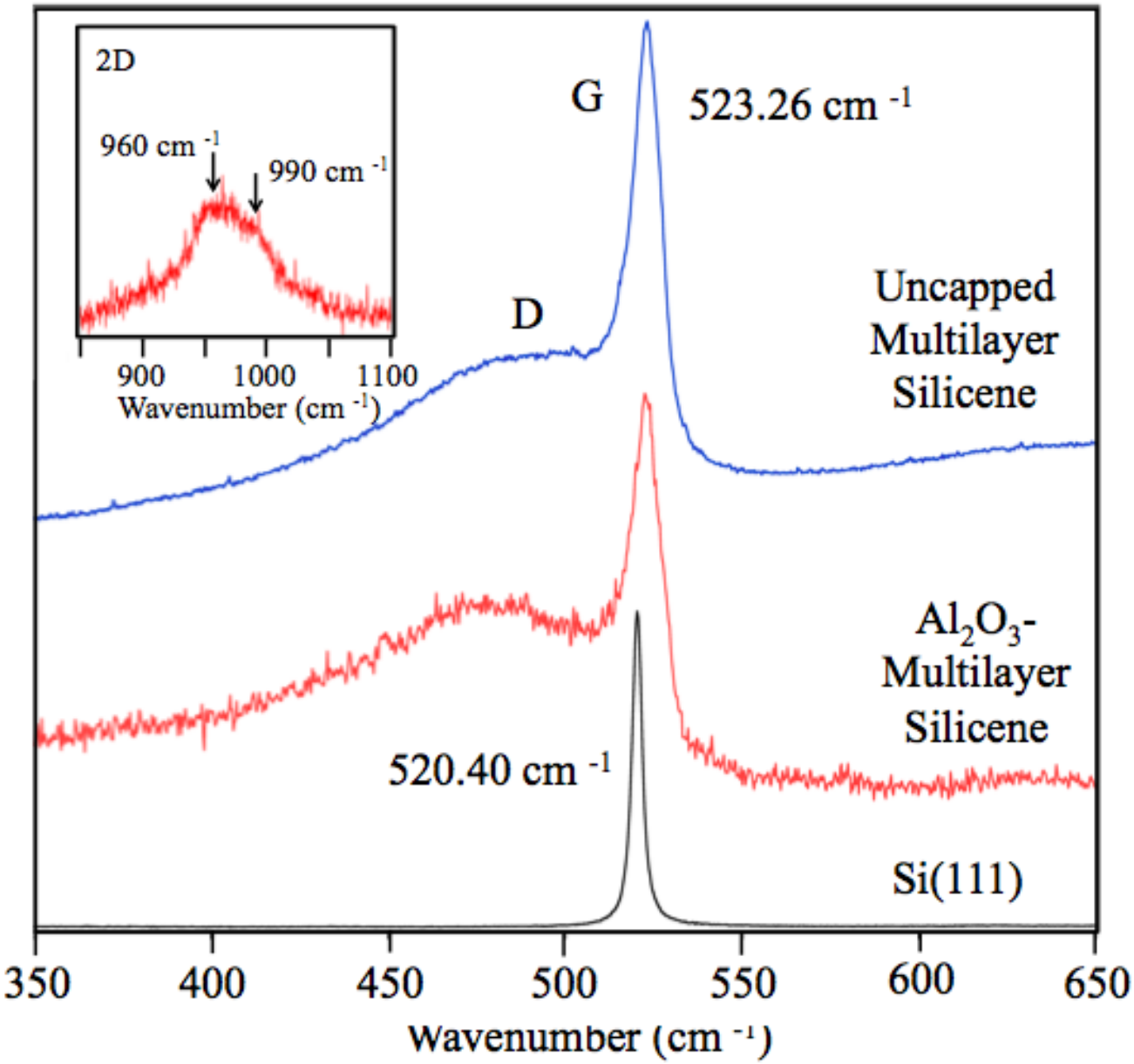
FWHM=1.63°
0.63°
2ϑ=28.71°
λ=1.5418 Å

$t^{\text{Multilayer silicene}} \approx$ 5MLs;
12 MLs

law $(2d \bullet \sin\vartheta = n\lambda)$ → Stacking between silicene layers ≈3 Å

P. De Padova *et al.*, 2D Materials, 1 (2014) 021003

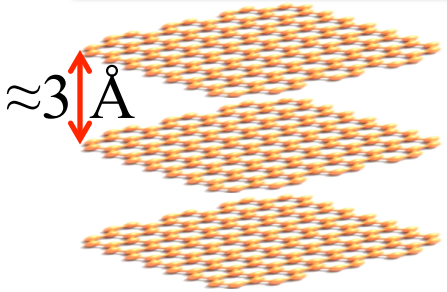
Raman spectroscopy on $\sqrt{3}\times\sqrt{3}R30°$ thick multilayer silicene



G—E_{2g} mode
Si-Si bond Stretghing
atom pair
honeycomb-like structure

D—Ph
Defects

2D—second order Ranan
Si(111) @≈900 cm^{-1}



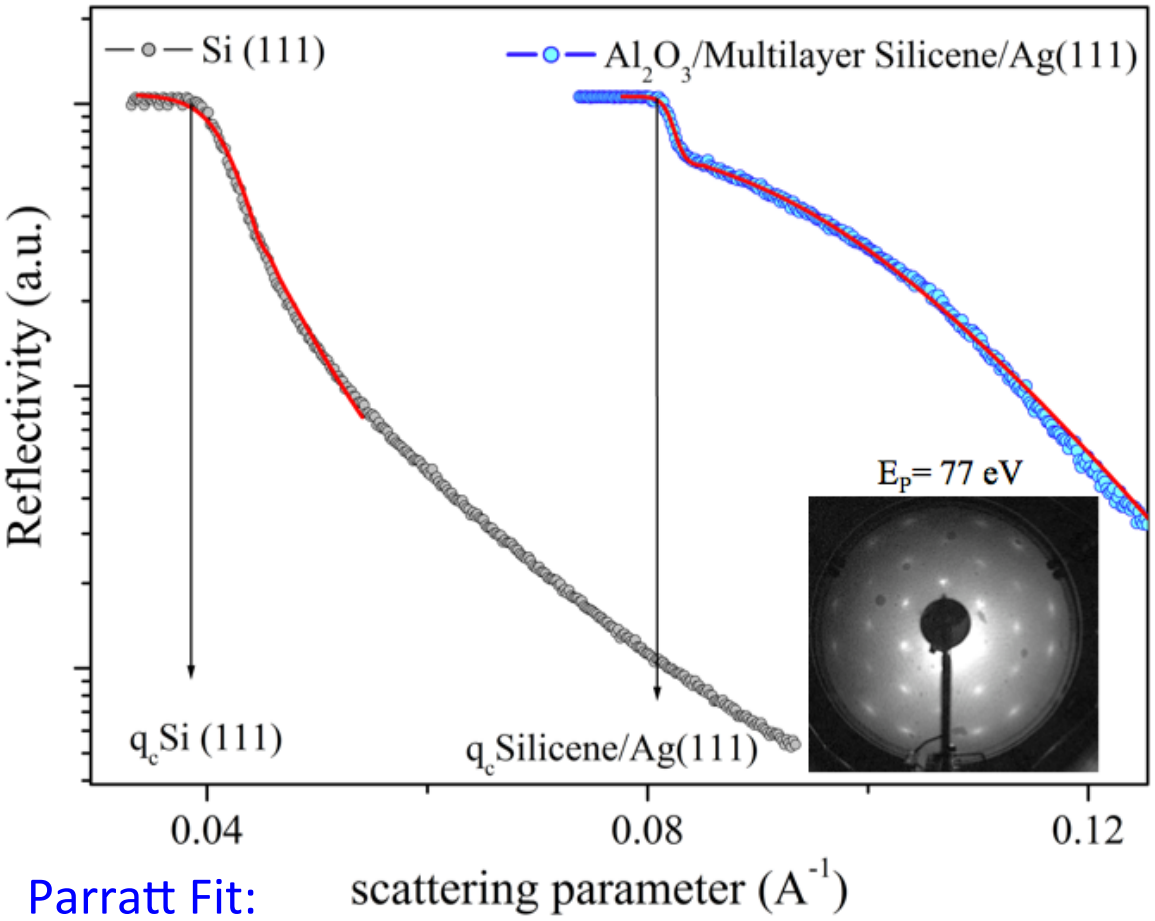
t Multilayer silicene
e≈ 5 (12) MLs

Red Laser
@633 nm

P. De Padova *et al.*, 2D Materials, 1 (2014) 021003

E. Cinquanta et *al.*,J. Phys. Chem C 117, 16719 2013.

$\sqrt{3}\times\sqrt{3}R30°$ silicene (EDXR)



Parratt Fit:

q_c Si(111) = 0.037 Å^{-1} and q_c Silicene/Ag(111) = 0.080 Å^{-1};

SLD (ρ):

ρ Si(111) = 2.10E-5 Å^{-2}; ρ Ag(111) = 7.70E-5 Å^{-2};

ρ silicene = 1.75E-5 Å^{-2}:

d(silicene) = (60 ± 10) Å

P. De Padova *et al.*, 2D Materials, 1 (2014) 021003

EDXR apparatus, CNR-ISM, Rome, Italy

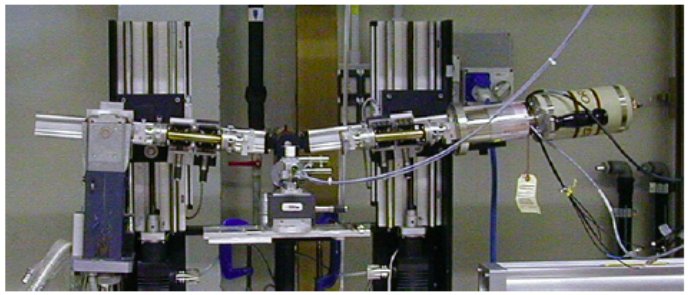


Figure 5. Sketch of the energy dispersive x-ray reflectometer. The fundamental components are as follows: (1) x-ray source (W anode tube), (2) collimation slits, (3) sample holder and (4) Ge single crystal energy-sensitive detector.

Multilayer Silicene thickness≈4.5 MLs

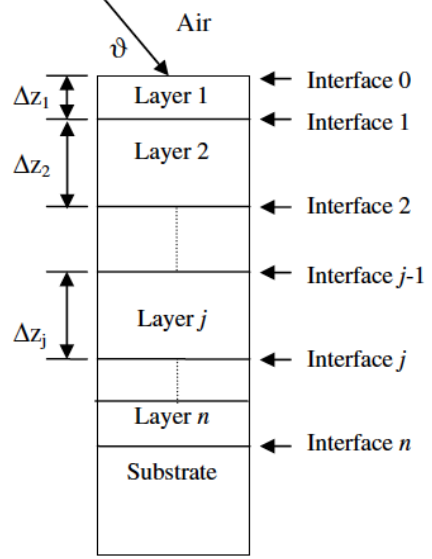


Figure 2. A schematic diagram showing the definitions used in the Parratt formalism for a multilayered film.

Rossi et al.

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J. Phys. D: Appl. Phys. **39** (2006) R461–R486

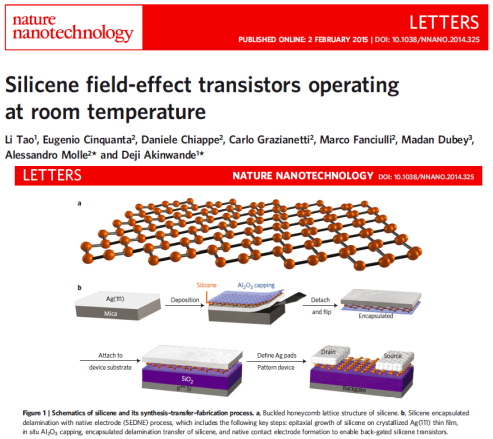
Summary

- We studied the structural and electronic properties of:

- Self-organized (1D), (2D grating) and (3D) Multilayer Silicene/Ag(110), and
- 2D Silicene sheet on Ag(111) and multilayer silicene on Ag(111)

by: STM/LEED/AES/XRD/EDXR/RAMAN/ARPES/core level spectroscopy

Showing that we are in presence of interesting 2D layered material and
For multilayer silicene we exclude the presence of Si(111).



- WORK IN PROGRESS:
  - Device multilayer silicene-based (in collaboration with CNR-IMM);
  - XRD/GISAXS on multilayer silicene grown on Ag(111) and Ag/Si(111);
- STM/STS and in situ Raman spectroscopy (in collaboration with Wollongong University

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