TECHNISCHE UNIVERSITÄT
CHEMNITZ

Epitaxial silicene: Silicon in the 2D world

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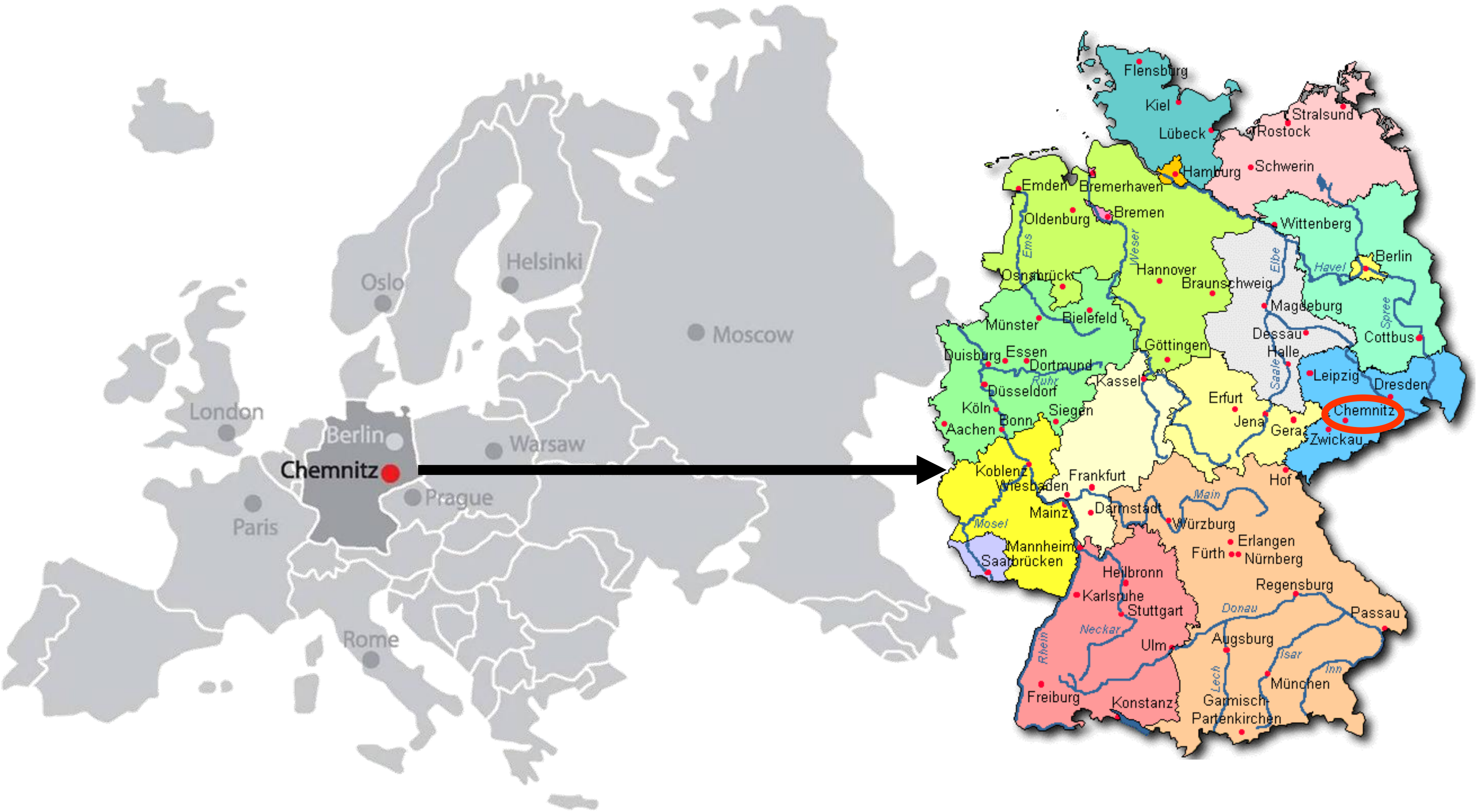
Winter school: New Frontiers in 2D Materials
Villard-de-Lans

January 15 - 20, 2017

Location | Saxony – City of Chemnitz



Impressions | City of Chemnitz

A new research centre in Chemnitz

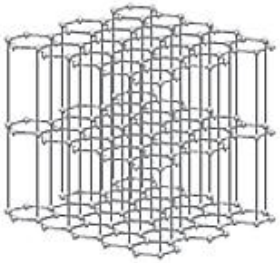
Center for **M**aterials, **A**rchitecture and **I**ntegration oif **N**ano-Membranes (**MAIN**)

- 43 million € funding
- 4000 m^2 lab and office space
- for ~100 scientists
- starting at the end of 2017

- nano-membranes
- 2D materials
- 2D organic layers
- hetero-structures

▶ For technological applications and flexible electronics

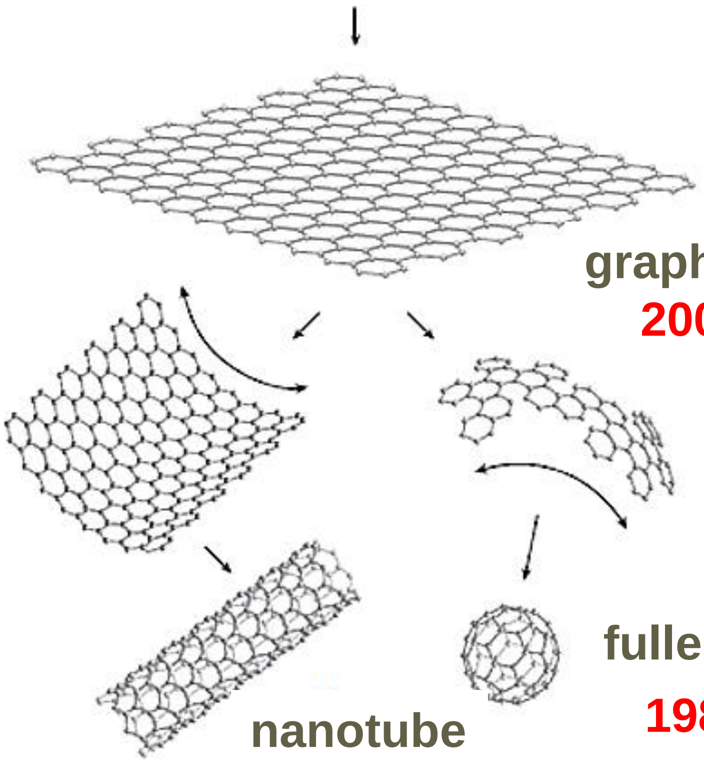
Motivation: carbon-structures



graphite **1564**

γράφειν = to write

Novoselov, Geim
Nobel Prize in Physics **2010**

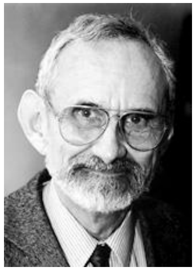


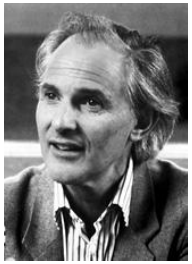
graphene
2004

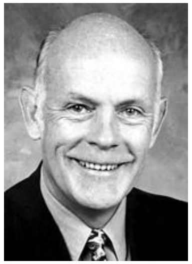
fullerene
1985

nanotube
Iijima
1991-1993

Curl, Kroto, Smalley
Nobel Prize in Chemistry **1996**

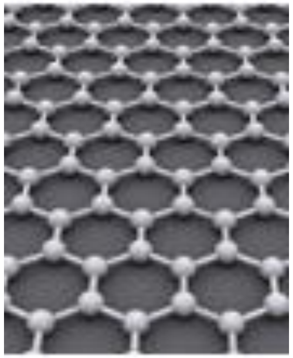




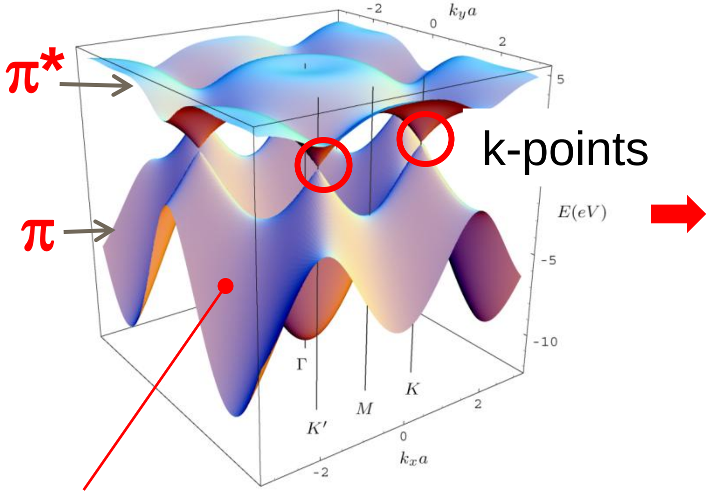


Motivation: graphene

Honeycomb structure

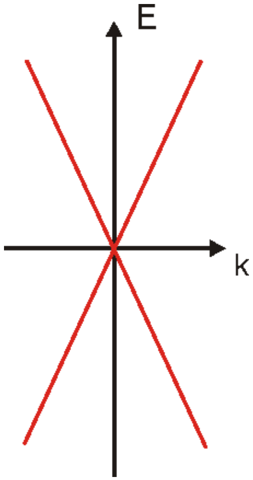


Energy dispersion relation E(k)



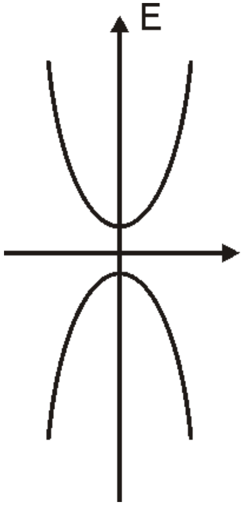
3D energy surface in k space.

“Dirac-cone”



$E_k = \nu_F k$

semiconductor



$E_k = h^2k^2/2m^*$

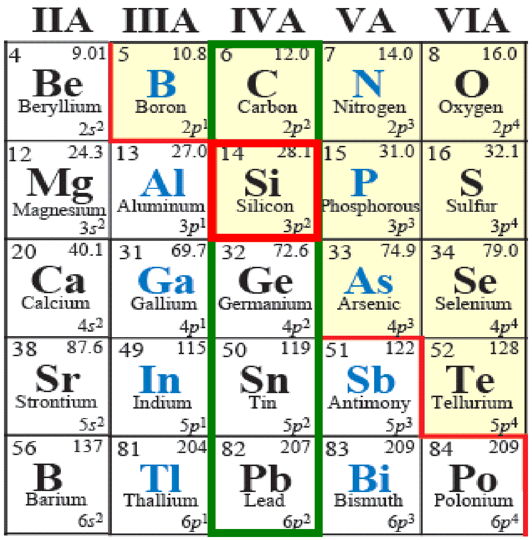
- linear dispersion of the π and π* bands close to the K points
- resembling properties of relativistic Dirac fermions
- Fermi velocity $\nu_F = 1.0 \times 10^6\ \mathrm{ms}^{-1}$

But:
- Graphene is semi-metallic and has no bandgap!
- Graphene is not (easily) applicable in logic and photonic devices.

Are there other similar elemental 2D materials?

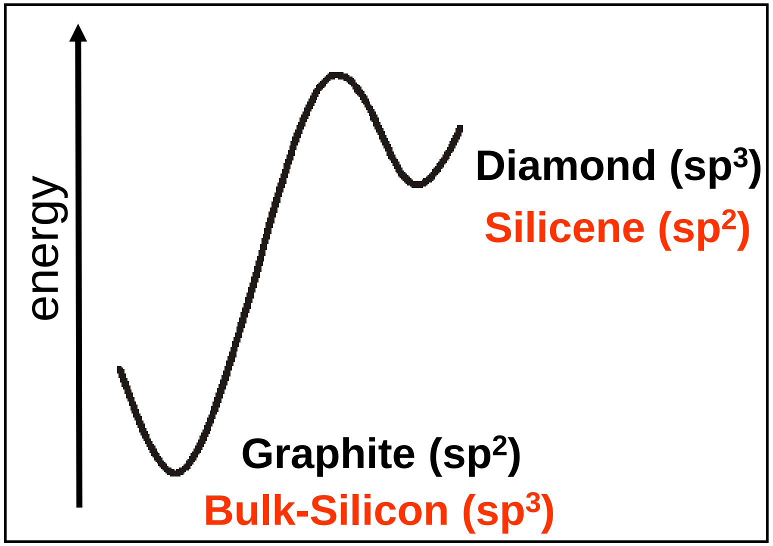
Motivation: 2D materials beyond graphene?

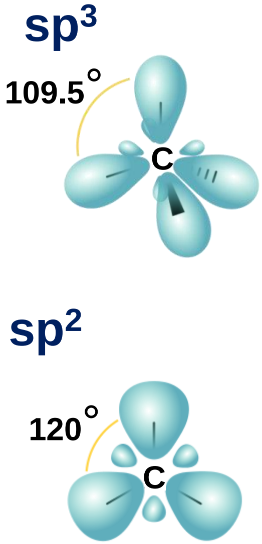
Other elemental 2D materials?



IIA	IIIA	IVA	VA	VIA
4 9.01 **Be** Beryllium $2s^2$	5 10.8 **B** Boron $2p^1$	6 12.0 **C** Carbon $2p^2$	7 14.0 **N** Nitrogen $2p^3$	8 16.0 **O** Oxygen $2p^4$
12 24.3 **Mg** Magnesium $3s^2$	13 27.0 **Al** Aluminum $3p^1$	14 28.1 **Si** Silicon $3p^2$	15 31.0 **P** Phosphorous $3p^3$	16 32.1 **S** Sulfur $3p^4$
20 40.1 **Ca** Calcium $4s^2$	31 69.7 **Ga** Gallium $4p^1$	32 72.6 **Ge** Germanium $4p^2$	33 74.9 **As** Arsenic $4p^3$	34 79.0 **Se** Selenium $4p^4$
38 87.6 **Sr** Strontium $5s^2$	49 115 **In** Indium $5p^1$	50 119 **Sn** Tin $5p^2$	51 122 **Sb** Antimony $5p^3$	52 128 **Te** Tellurium $5p^4$
56 137 **B** Barium $6s^2$	81 204 **Tl** Thallium $6p^1$	82 207 **Pb** Lead $6p^2$	83 209 **Bi** Bismuth $6p^3$	84 209 **Po** Polonium $6p^4$

Schematic energy diagram





Graphite (Graphene) is the most stable form of carbon

But:
sp^2 hybridized silicene, germanene,...
are less stable than the sp^3 hybridized bulk forms

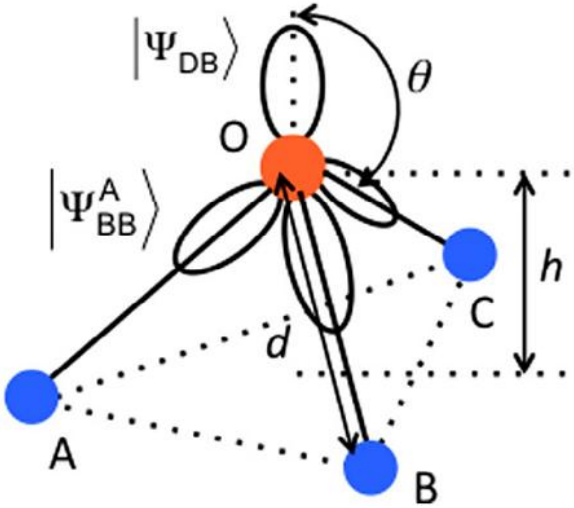
▼

Solution: a mixed sp^2/sp^3 hybridization state

Silicene: sp^2/sp^3 hybridization

N. Takagi, *et al.*, Prog. Surf. Sci. **90**, 1 (2015).

Analogy to the dangling bond state on Si (111)



$$E_{DB} = \frac{2q^2E_s + (1-3q^2)E_p}{1-q^2}$$

$$E_{BB} = \frac{(1-3q^2)E_s + 2E_p}{1-3q^2}$$

where ***q* = *h*/*d*** – “buckling ratio”

(buckling distance/Si-Si bond length)

Varying the buckling ratio ***q***:

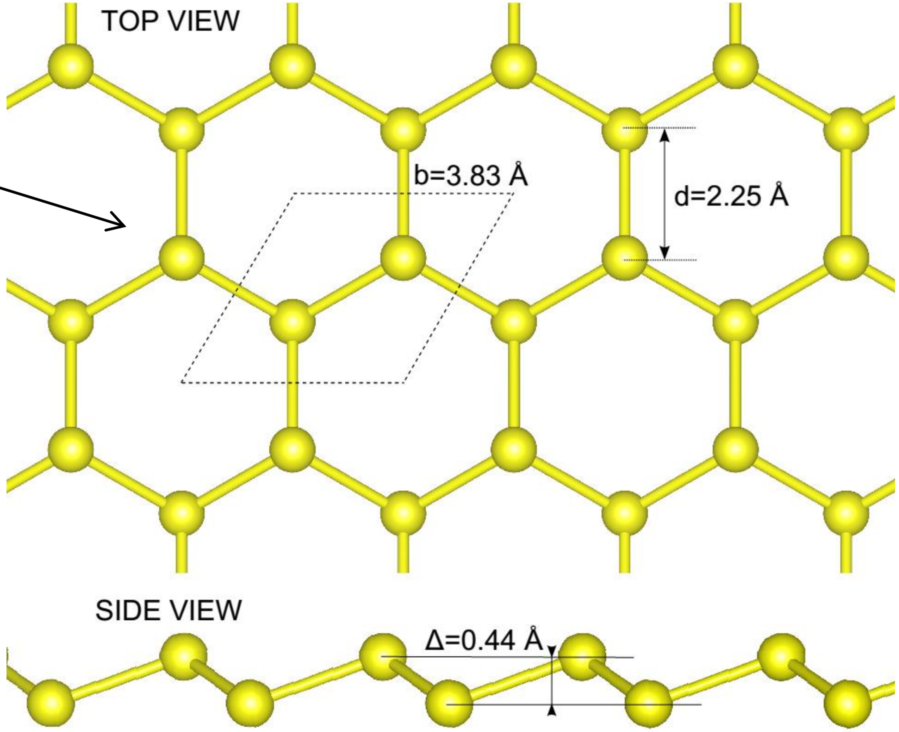
$q = 0$ ➔ DB=Ep; BB=Es+2Ep ➔ sp^2 hybridization

$q \sim 1/\sqrt{3}$ ➔ sp^2/sp^3 hybridization

$q = 1/3$ ➔ DB=BB=Es+3Ep ➔ sp^3 hybridization

Silicene: geometric structure

K. Takeda, K. Shiraishi, PRB **50,** 14916 (1994)
G. G. Guzmán-Verri, et al., PRB **76**, 075131 (2007)
S. Cahangirov et al.,PRL **102**, 236804 (2009)



Si atoms

Si-Si bond
diamond Si: 2.35 Å

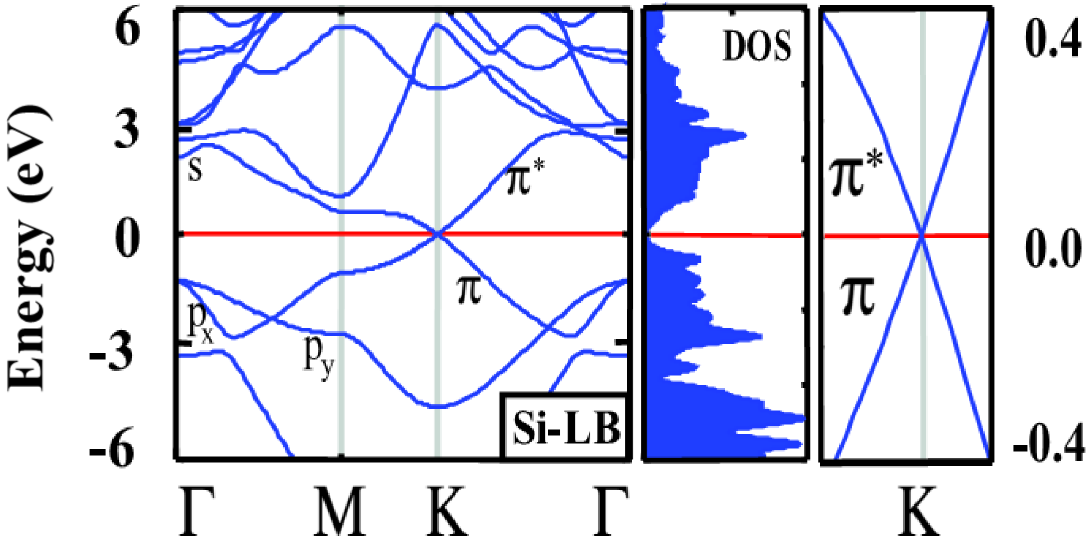
Buckling (Δ)
diamond Si (111)
plane: 0.29 Å

mixed sp^2/sp^3
hybridization

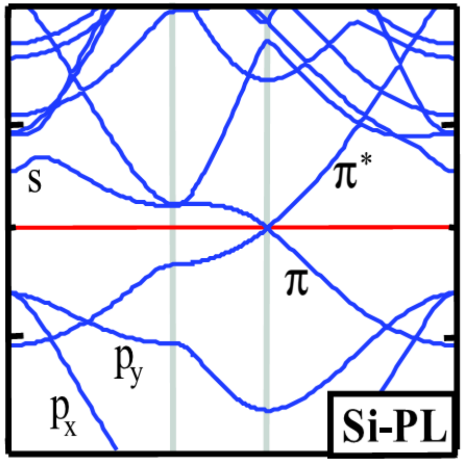
Silicene: electronic structure

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

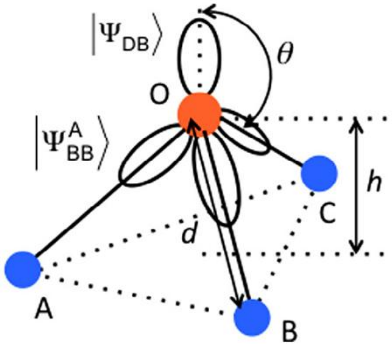
Low-buckled (LB)



Planar (PL)



N. Takagi, *et al.*, Prog. Surf. Sci. **90**, 1 (2015).



$q = h/d$:

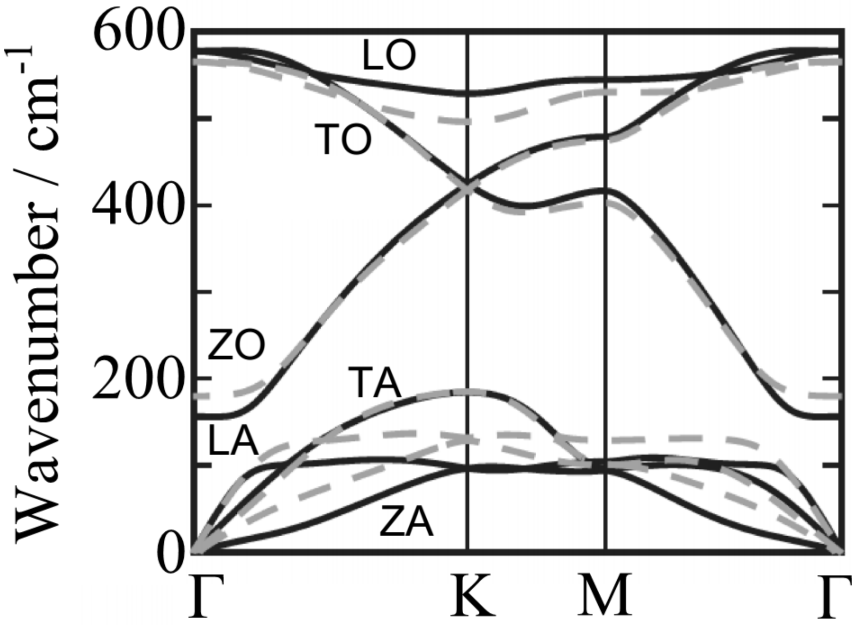
$q = 0$ ($\theta = 90°$) ➔ sp^2 hybr.

$q \sim 1/\sqrt{3}$ ➔ sp^2/sp^3 hybr. ➔

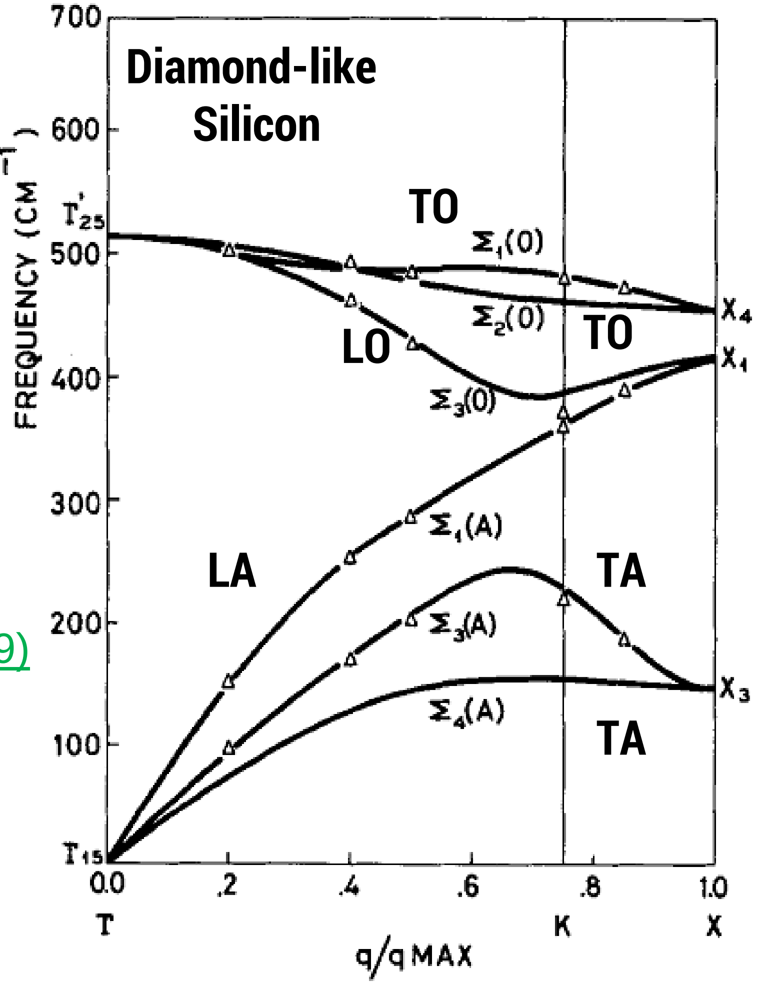
$q = 1/3$ ($\theta = 109.5°$) ➔ sp^3 hybr.

The bonds have a mixed sp^2 and sp^3 character

Silicene: vibrational structure



<u>S. Cahangirov, *et al.*, Phys. Rev. Lett. **102**, 236804 (2009)</u>
P. Gori, *et al.* Energy Proc. **45**, 512 (2014).
X. Li, *et al.* Phys. Rev. B **87**, 115418 (2013).

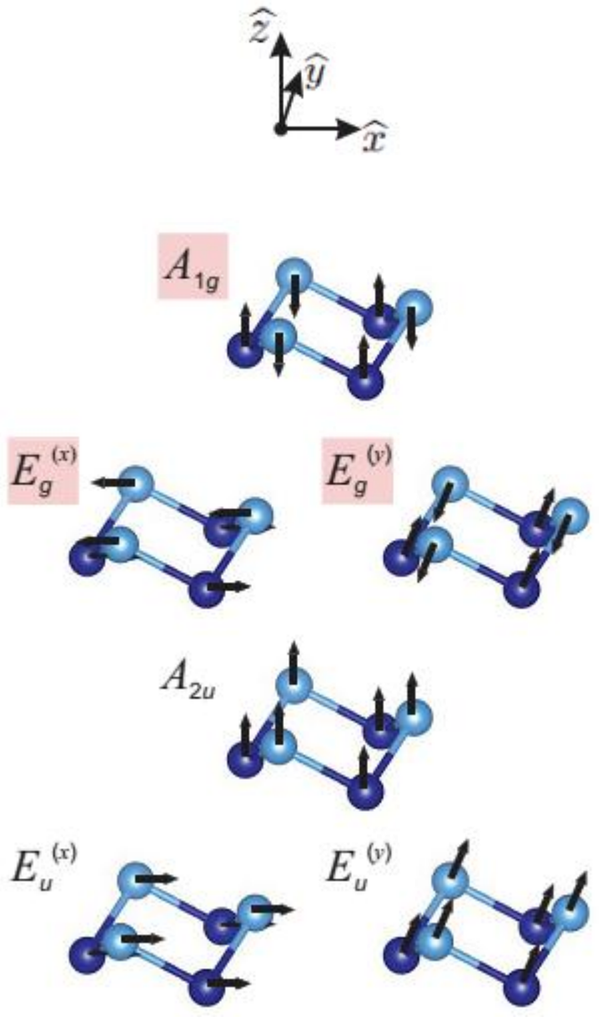


R. Tubino, *et al.*,
J. Chem. Phys. **56**, 1022 (1972).

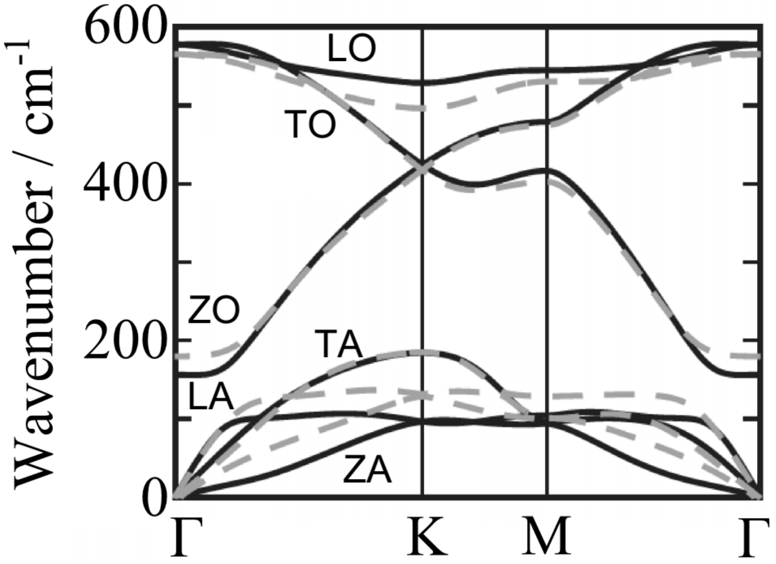
<u>Characteristics:</u>

- Higher energy of the L(T)O phonons
- Out-off-plane ZO phonon

Raman mode symmetries



J. Ribeiro-Soares, *et al.*, Phys. Rev. B **91**, 205421 (2015).



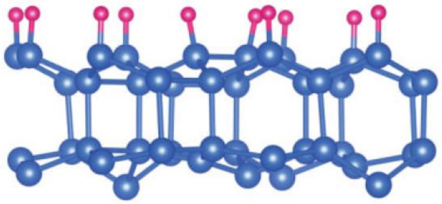
S. Cahangirov, *et al.*, Phys. Rev. Lett. 102, 236804 (2009)

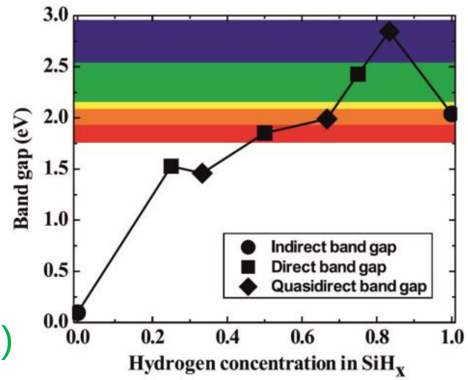
- A-symmetry phonons: out-of-plane motion
- E-symmetry phonons: in-plane motion

Expected properties of silicene

- Silicene could be superconducting - possibility of high Tc by B doping
- Because of the large spin-orbit coupling in Si compared to C, it is predicted to exhibit a quantum spin Hall-effect in an accessible temperature regime (10-20K)
- Silicene could be integrated easily in silicon-based nanotechnology (maybe)
- Predicted high carrier mobilities (~$2.6 \cdot 10^{5}$ $cm^{2}V^{-1}s^{-1}$)
- Band gap opening by an electric field or functionalization
- Novel silicene-based devices:

Hydrogenated bilayer-silicene for optical applications





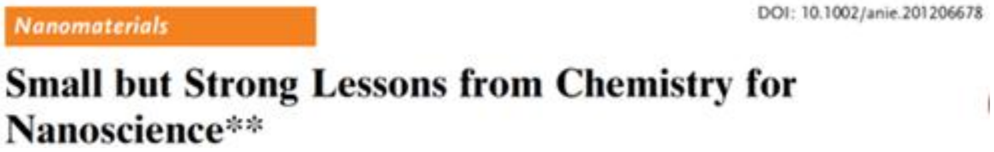
Hydrogenation of bilayer silicene opens a direct tunable bandgap between 1.5 – 2.9 eV

B. Huang et al, Phys. Rev. X. **4**, 021029 (2014)

How to synthesize silicene.....

Contrary to graphene, silicene does not exist in nature!

↓



Nanomaterials

DOI: 10.1002/anie.201206678

Small but Strong Lessons from Chemistry for Nanoscience**

Roald Hoffmann

1981

“Silicene exists and will be made only on a support of some sort, metal or semiconductor”

Angew. Chem. Int. Ed. **52**, 93 (2013)

↓

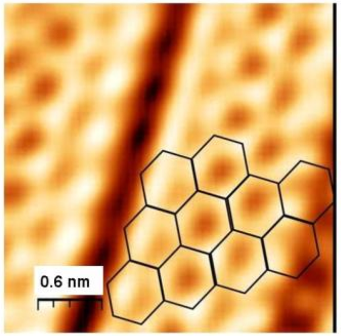
Synthesis on a nonreactive substrate preventing 3D growth

- Synthesis of 2D silicene on a templates
- Preparation under vacuum conditions (UHV)

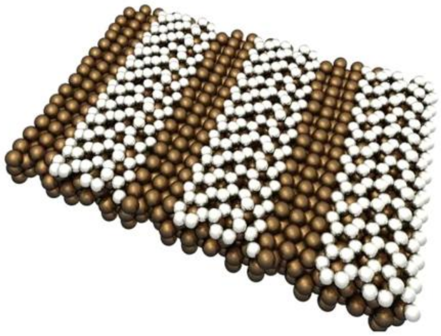
Synthesis of silicene on Ag surfaces

1D Si nano-ribbons on Ag(110)

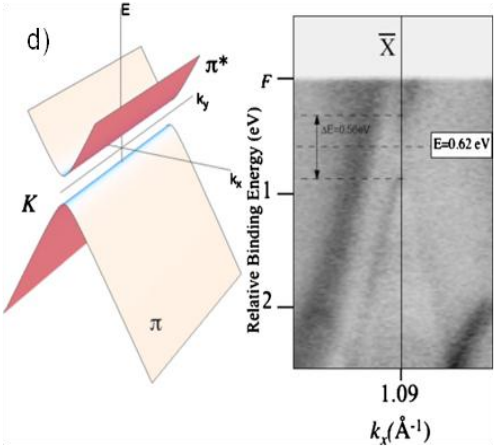
STM



structure model



cones at the Dirac points



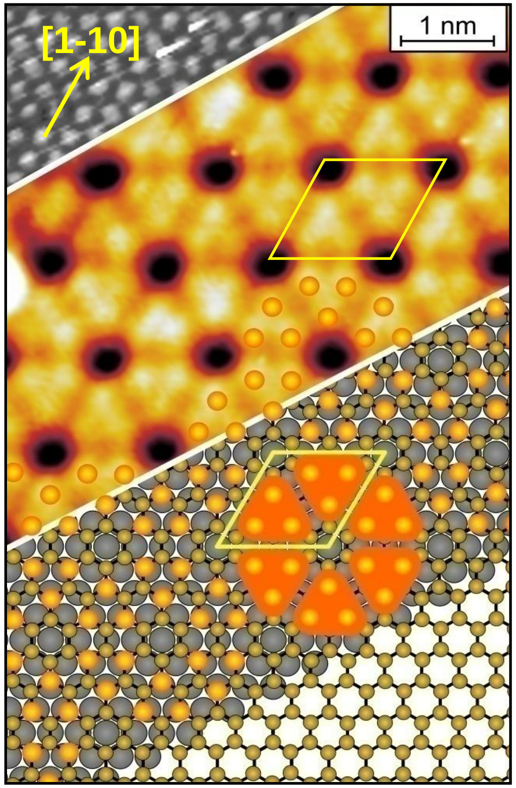
Ronci et al., pss c **7,** 2716 (2010)
Sahaf et al., APL **90,** 263110 (2007)
M.E. Davila et al., Nanotechnology **23,** 385703 (2012)
Le Lay et al., Appl. Surf. Sci. **256**, 524 (2009)
De Padova et al., APL **96**, 261905 (2010)

➡ No silicide formation!

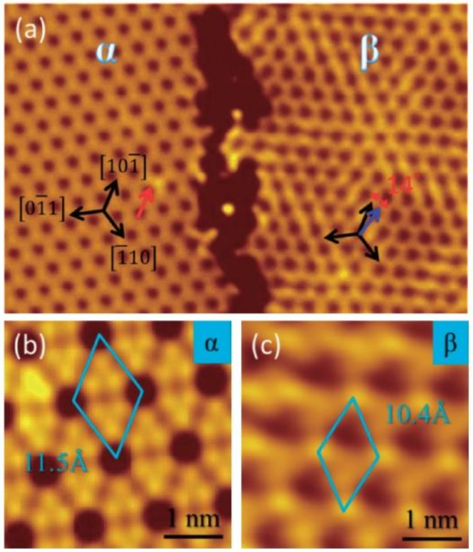
Ag(111) templates for the synthesis of 2D silicene

Synthesis of silicene on Ag(111) in 2012

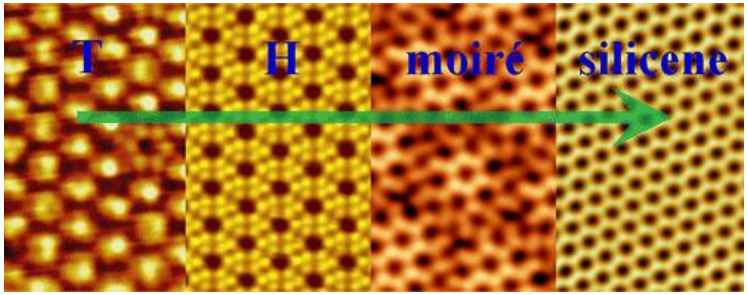
Phys. Rev. Lett. **108**, 155501 (2012)



C.-L. Lin et al.,
Appl. Phys. Express, **5** (2012) 045802



A. B. Feng et al., Nano Letters **12**, 3507 (2012)



Outline

- **part 1:** **- epitaxial growth of silicene on Ag(111)**

- **part 2:** **- Vibrational properties of epitaxial silicene / Ag(111)**

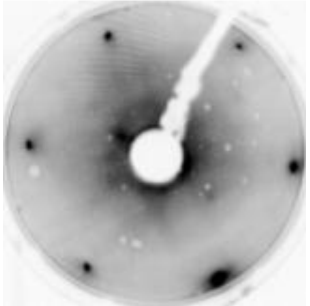
  - Other elemental 2D materials

Synthesis of 2D silicene on Ag(111)

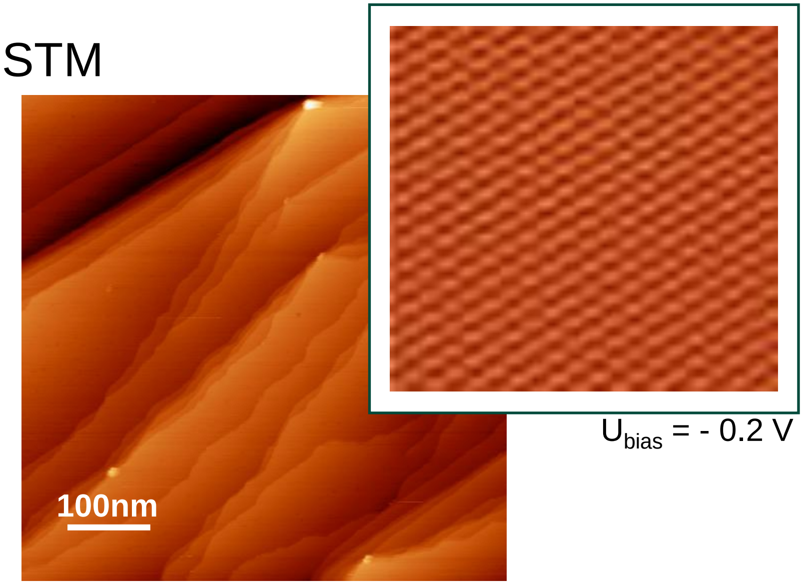
Preparation of Ag(111)

- sputtering for 1h (1.5kV, 5×10^{-5} mbar Ar^+)
- annealing at 560°C for 30 minutes

LEED: (1x1)



STM



U_{bias} = - 0.2 V

600nm x 600nm, U_{bias} = - 0.18 V

- flat surface with big terraces

Si deposition:

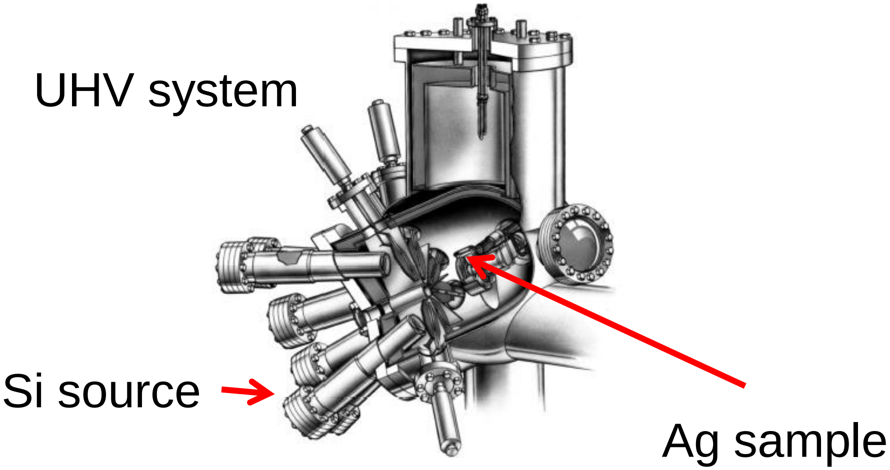
Si source:

- directly heated Si-wafer piece

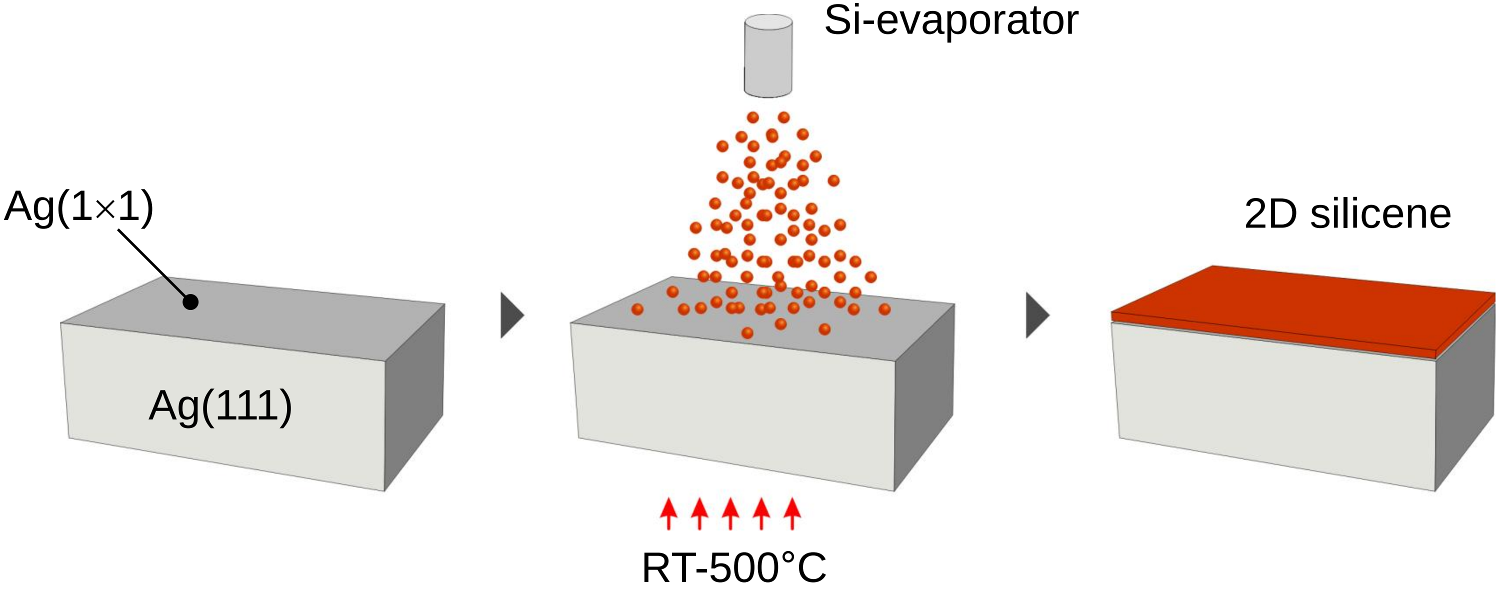
Growth parameters:

- sample temperature: RT – 450°C
- deposition rate and coverage

UHV system



Growth of 2D Si-layers on Ag(111)



Preparation of a clean
Ag(111)(1×1) surface

Si deposition at a certain
sample temperature

Formation of a
2D silicene layer

Growth Conditions: UHV

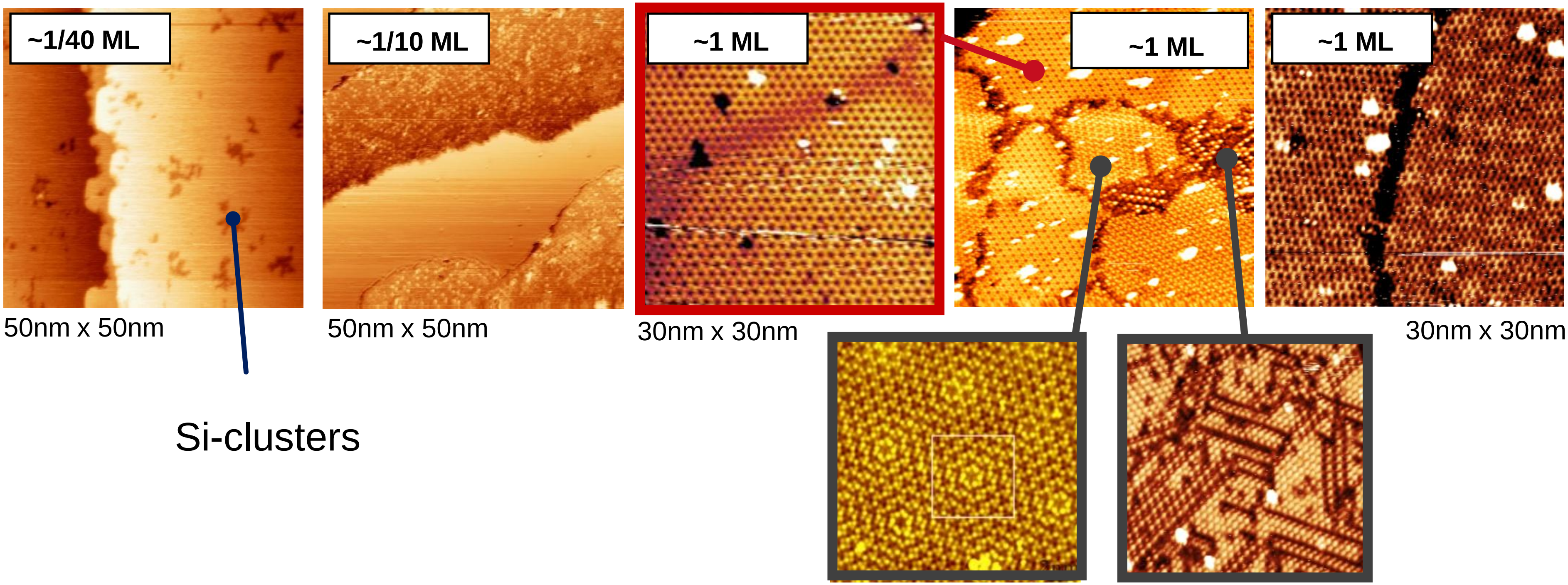
Growth parameters:

- sample temperature: RT – 500°C
- deposition rate and coverage

The sample temperature is crucial for the formation of Si-phases on Ag(111)

Si deposition on Ag(111) at different temperatures

RT	<180°C	~220°C	>220°C	>250°C
Si-clusters	**“disordered ”**	**(3×3)/(4×4)**	**multi-phase**	**“(2√3×2√3)”**



50nm x 50nm

50nm x 50nm

30nm x 30nm

30nm x 30nm

Si-clusters

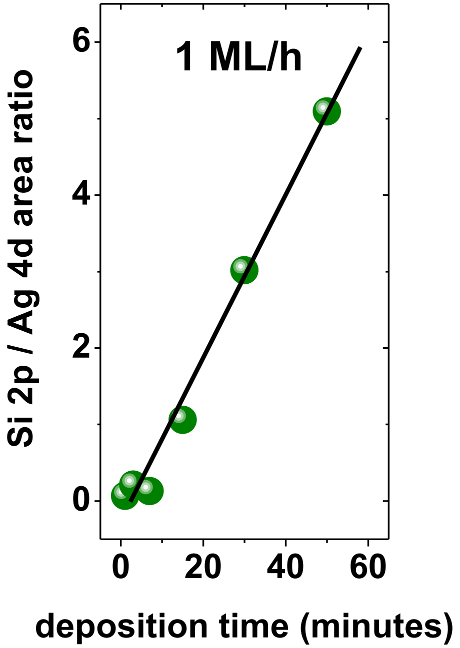
(√13×√13)-α

(√13×√13)-β

The formation 2D Si-layers / Ag(111) depends crucially on the temperature

Si deposition on Ag(111) at 220°C

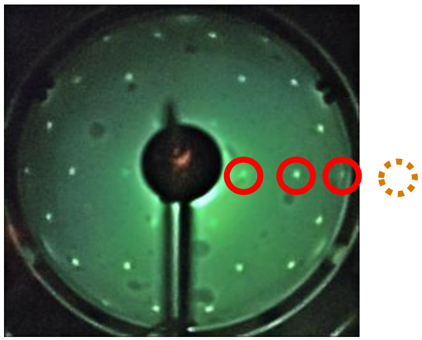
Growth mode



↓

linear growth mode

LEED



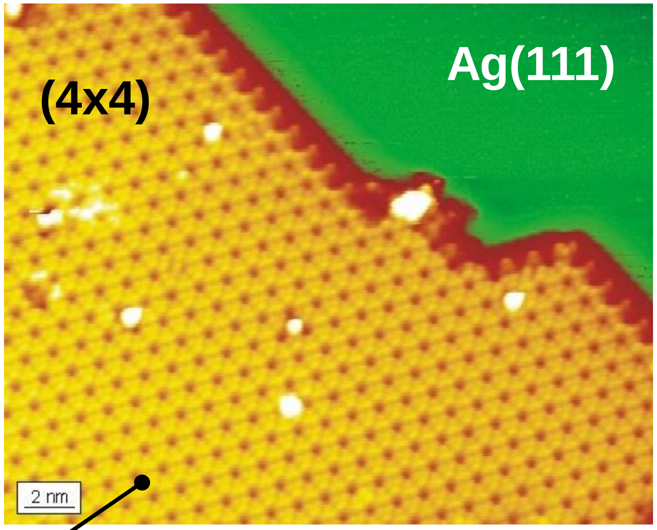
○ ¼ spots

◌ integer spots

↓

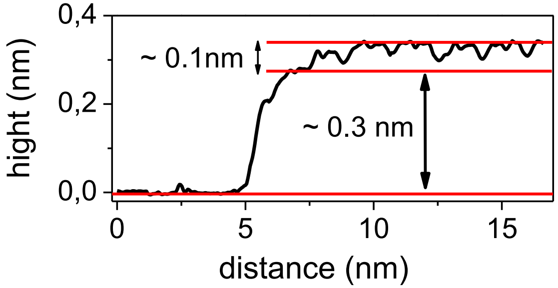
clear coincidence
(4x4) symmetry

STM



U = - 1.4 V

honeycomb-like
structure



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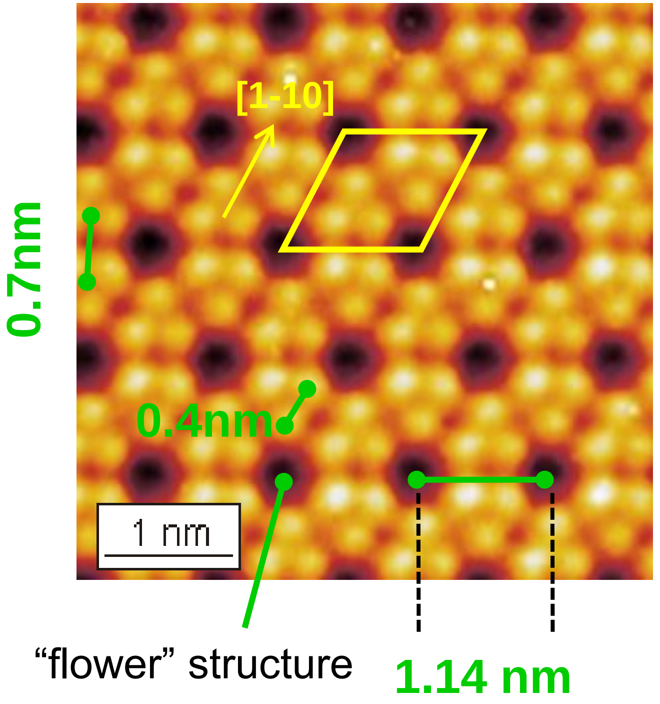
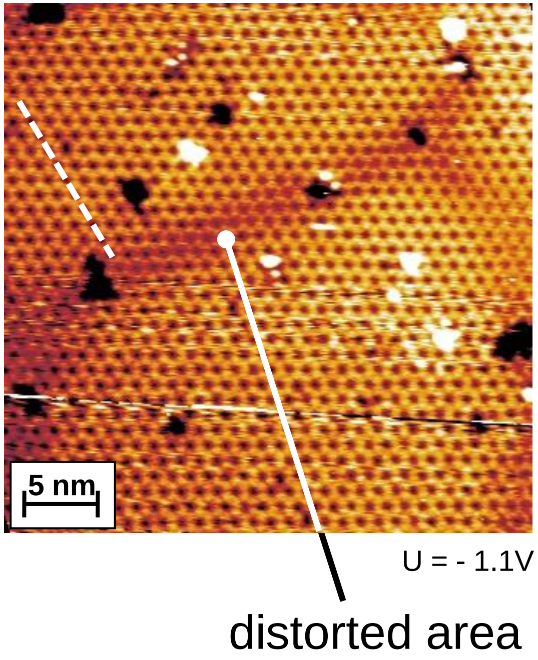
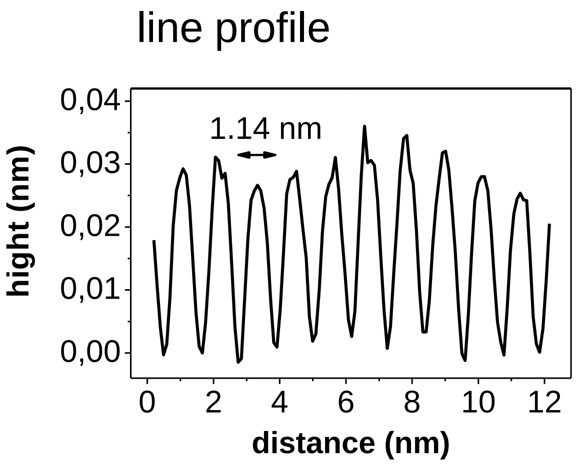
2D layer

2D growth of a 2D Si layer with a (4x4) periodicity (with respect to Ag 1x1)

STM of the Si “(4×4)” structure







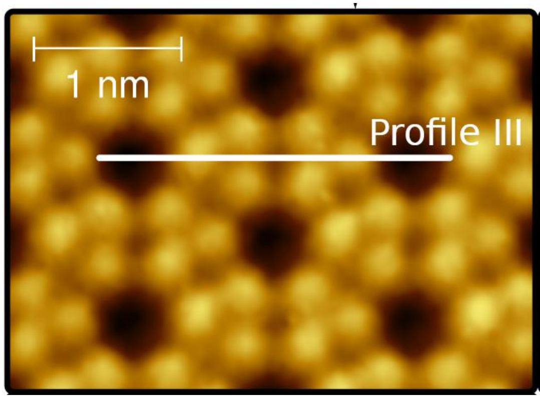
1.14nm ~ 4x Ag-Ag distance

The structure has a (4x4) periodicity with respect to Ag(111)
In agreement with LEED

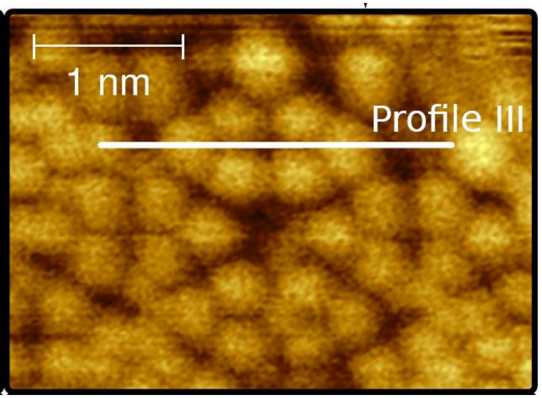
Phys. Rev. Lett. **108**, 155501 (2012)

Comparison of STM and nc-AFM

STM

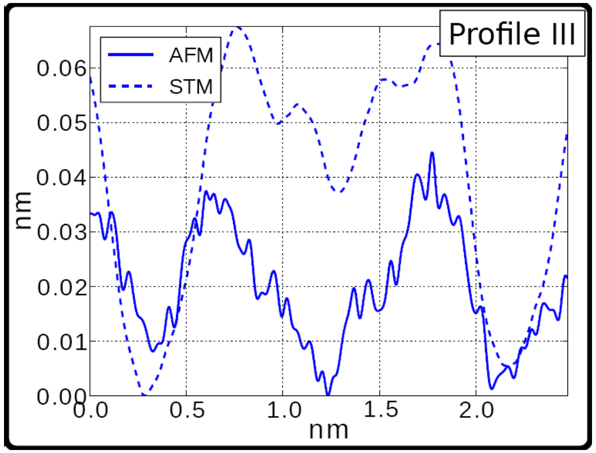


nc-AFM



Atomically resolved AFM show the same “flower”-like structure for the (3x3) silicene

line profile



Both images are clearly dominated by geometric factors

A. Resta et al., Sci. Rep. **3**, 2399 (2013)

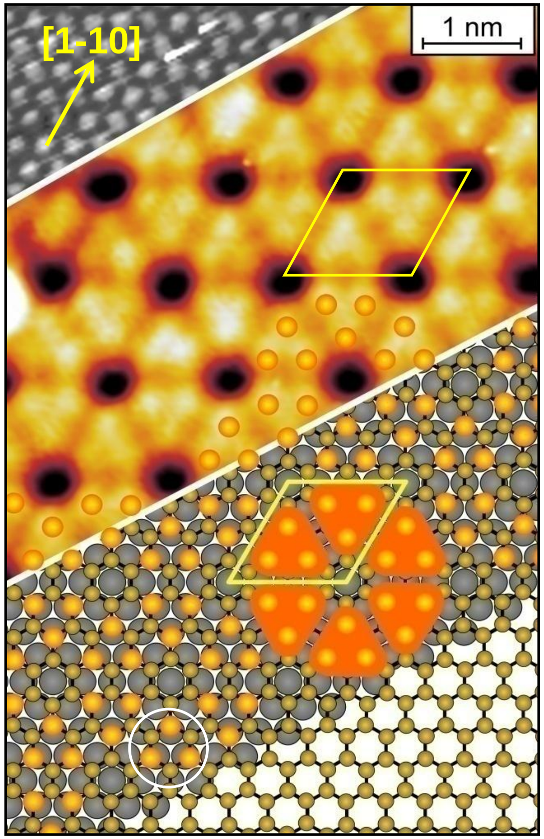
(3×3)/(4×4) silicene on Ag(111): structure model

Structure model

STM:
Ag(111)

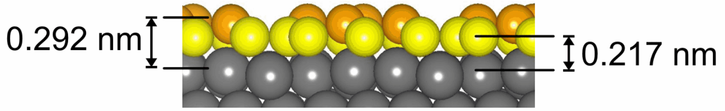
STM:
Si layer

Model

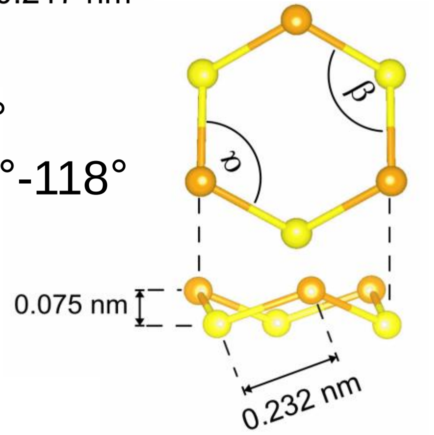


DFT results

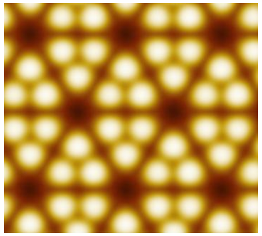
- TE calculation: structure is energetically stable



- sp^2/sp^3-character $\left\{ \begin{array}{l} \alpha \sim 110^\circ \\ \beta \sim 112^\circ\text{-}118^\circ \end{array} \right.$



- Si-Si distance: 0.23nm
- simulated STM image agrees well with the experimental ones



▶ DFT results support the model

The (3×3)/(4×4) structure indicates a significant Ag-silicene interaction!

Phys. Rev. Lett. **108**, 155501 (2012)

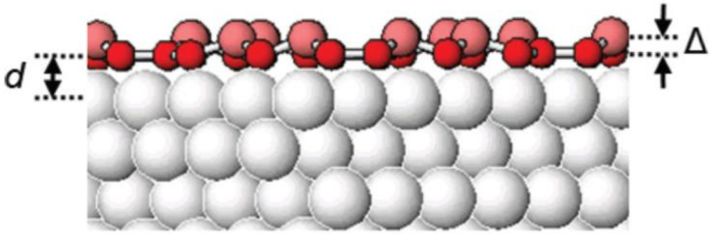
Confirmation of the model by diffraction methods

Positron diffraction (RHEPD)

Y. Fukuya et al.,Phys. Rev. B **88**, 205413 (2013)

Structure parameters

	Δ (Å)	d (Å)	α (°)	β (°)
This study	0.83	2.14	112	119
Our values	0.78	2.17	110	118



Dynamical LEED analysis

K. Kawahara et al., Surf. Sci. **623** ,25 (2014)

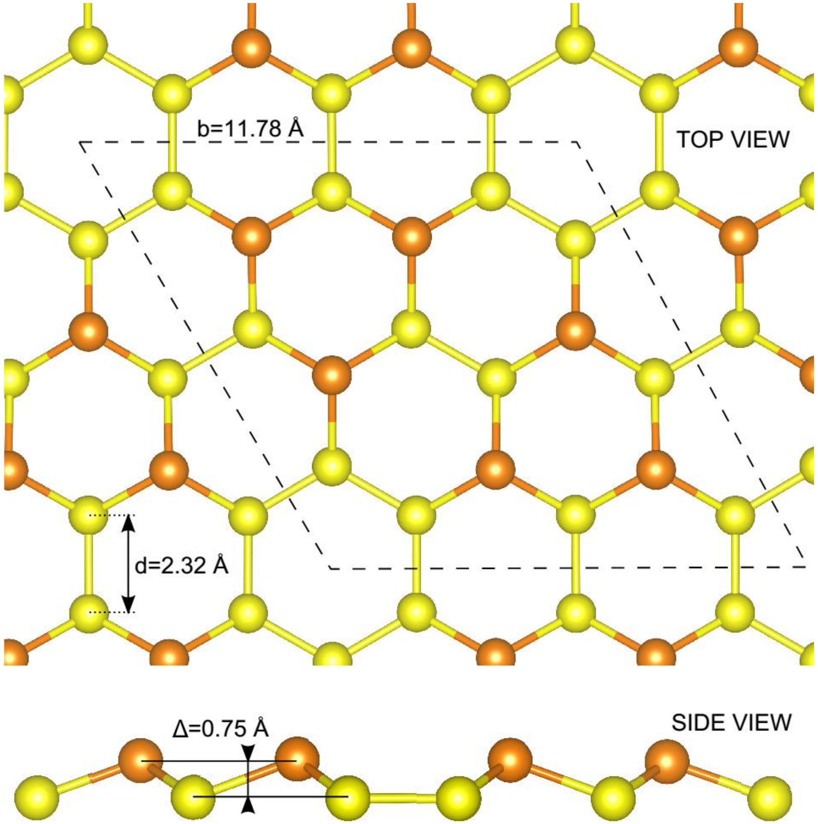
Structure parameters

	Δ (Å)	d (Å)	Si-Si (nm)
This study	0.74 - 0.77	2.18	0.23
Our values	0.78	2.17	0.23

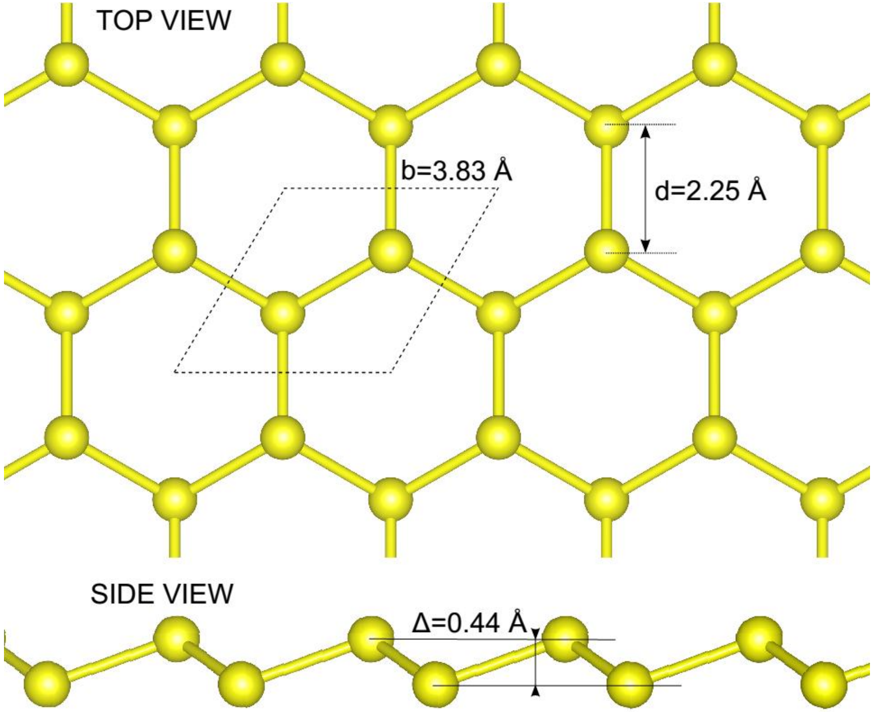
Diffraction methods confirm the(3×3) silicene model

Epitaxial silicene vs. free-standing silicene

“Epitaxial (3×3)/(4×4) silicene”



Free-standing silicene

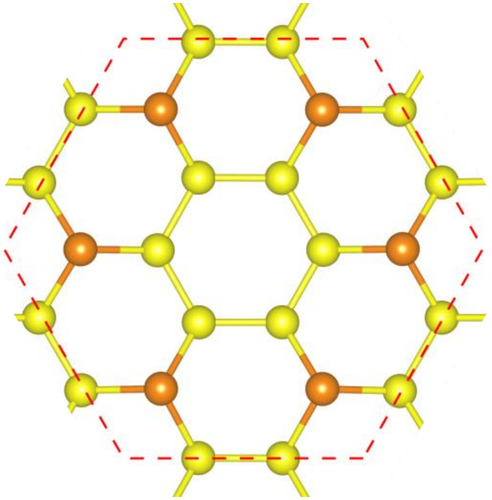


Interaction with a substrate ➤

- geometric structure: ≈
- electronic structure: **?**
- vibrational structure: **?**

Epitaxial silicene: C_{6v} Symmetry

Wigner-Seitz unit cell



Epitaxial (3x3)/(4x4) silicene:

- symmetry breaking by reconstructing

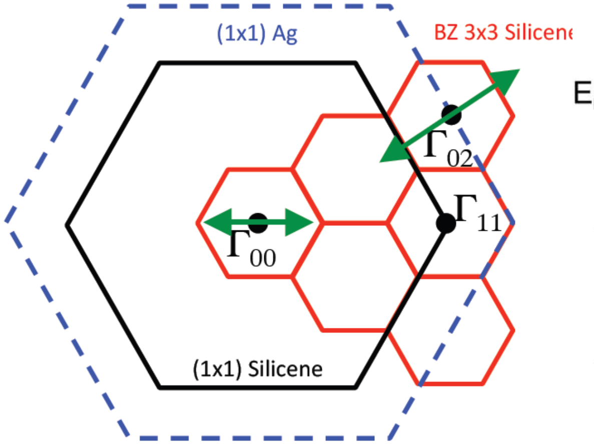
2 sub-lattices A and B

A: 12 Si atoms
B: 6 Si atoms

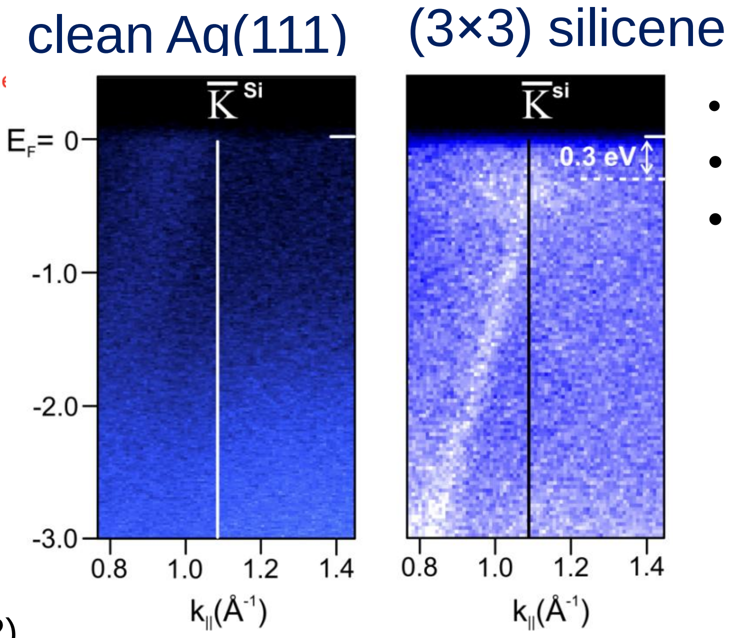
But: it has a highly symmetric C_{6v} symmetry

Influence on the electronic properties?

ARPES measurements on the (3×3) silicene



Phys. Rev Lett. **108**, 155501 (2012)



- No $k_\perp$ dependence
- $v_F = 1.3 \times 10^6\ ms^{-1}$
- Small band gap (≥ 0.3 eV)?

Dirac-cones for (3×3) silicene?

But: DFT calculations for the (3x3) silicene do not reproduce the Dirac cones!

C.-L. Lin et al., PRL 110, 076801 (2013), S. Huang et al. APL 102, 133106 (2013)
Z.-X. Guo et al., *J. Phys. Soc. Japan* **82**, 063714 **(2013),**
Y.-P. Wang, H.-P: Cheng, Phys. Rev. B **88**, 125428 (2013), P. Gori et al., *JAP* **114**,113710(2013)

But: No $k_\perp$ dependence: states are localization at the surface!

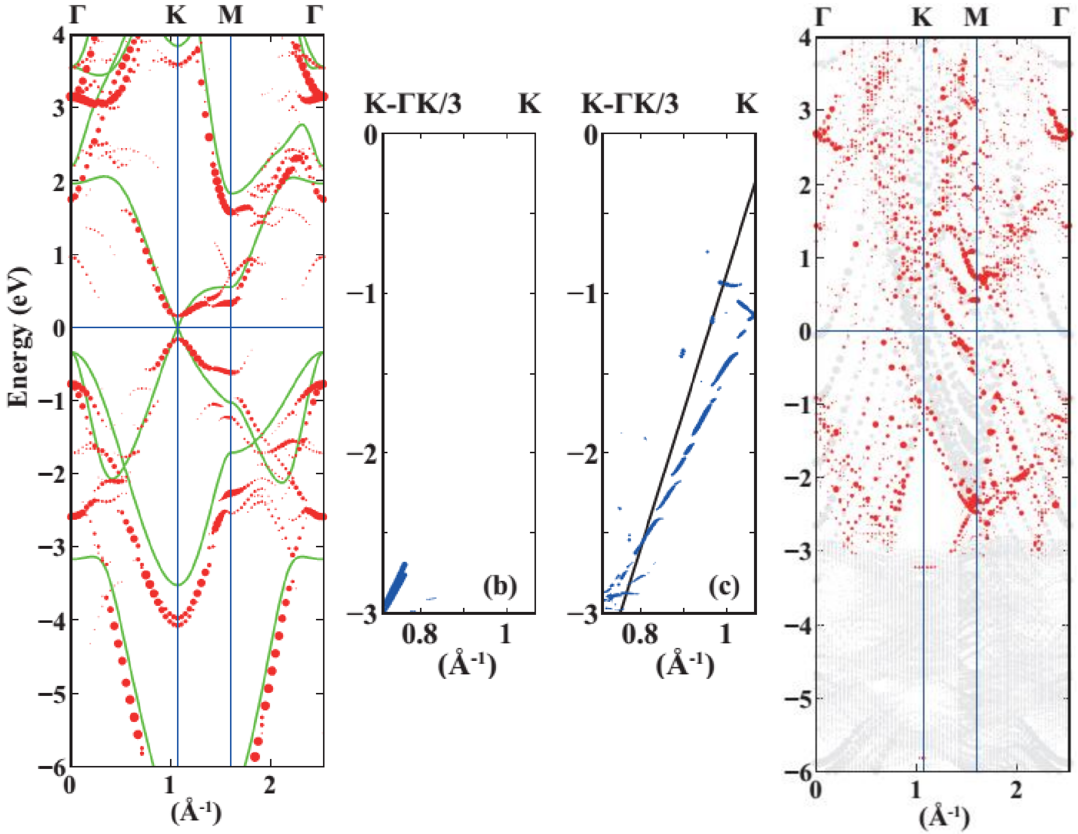
➡ *sp bands of bulk Ag cannot explain the ARPES results*

Epitaxial silicene: electronic structure

S. Cahangirov Phys, Rev. B **88, 035432 (2013):**

- Explanation of the experimental dispersion by: **hybridised states**

Reconstructed silicene (without Ag substrate)



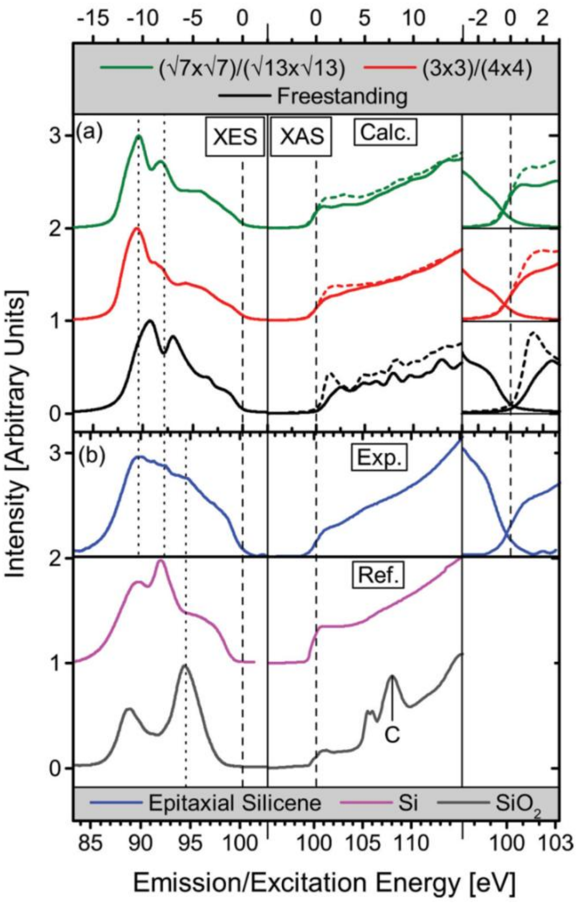
Ag substrate

- hybridized states are localized at the interface and give rise to a linear band
- in the absence of silicene these states disappear

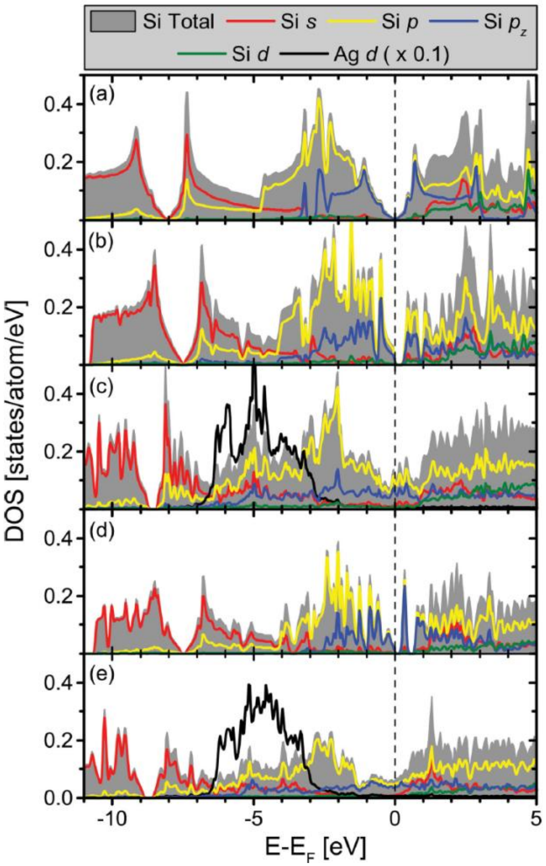
➡ *hybridized sates are in agreement with the experimental observations*

Soft X-ray absorption and emission spectroscopy

XAS and XES



Calculated DOS



(3×3)/(4×4)
without Ag(111)

(√13×√13)
without Ag(111)

(3×3)/(4×4)
with Ag(111)

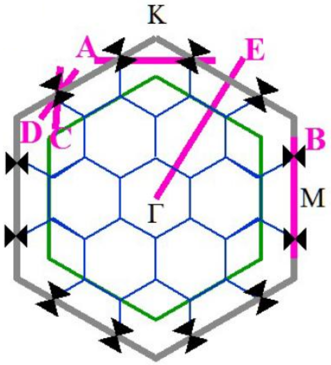
(√13×√13)
with Ag(111)

Confirmation of the metallic character of (4x4) silicene/Ag(111)

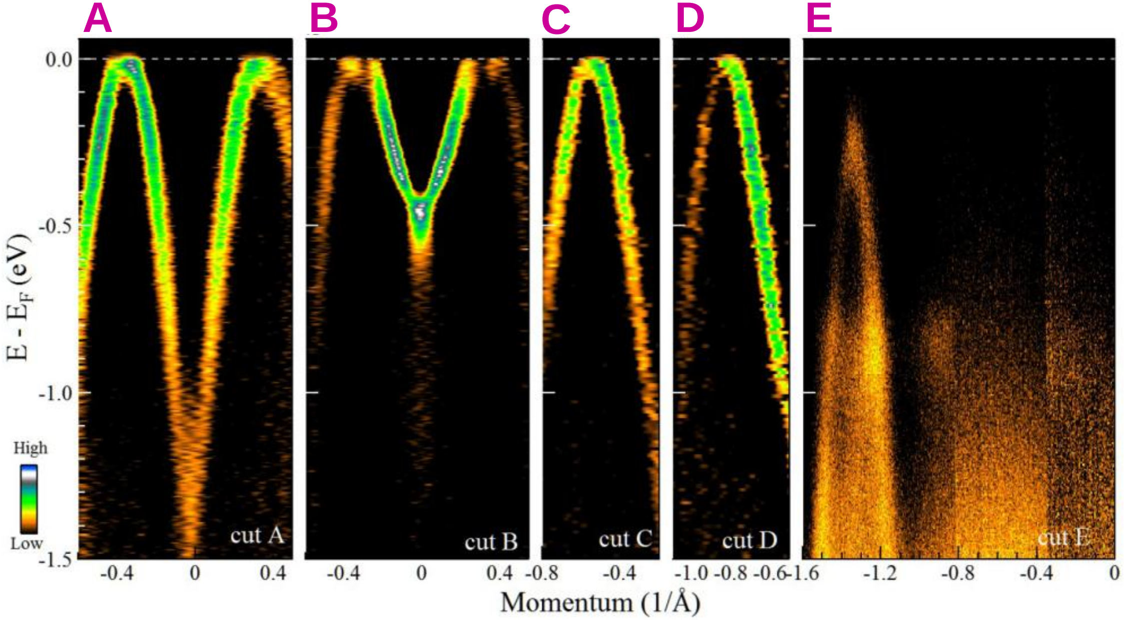
N.W. Johnson et al., Adv. Funct. Mater. **24**, 5253 (2014)

Recent ARPES results for (3×3) silicene

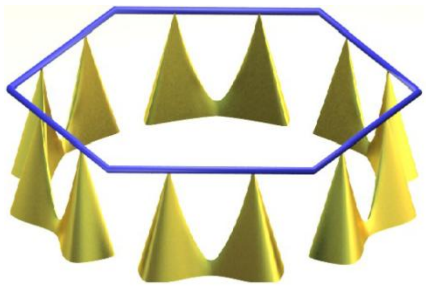
Feng et al., PNAS (2016), doi pnas.1613434114



Band structure along different cuts



Schematic 3D plot



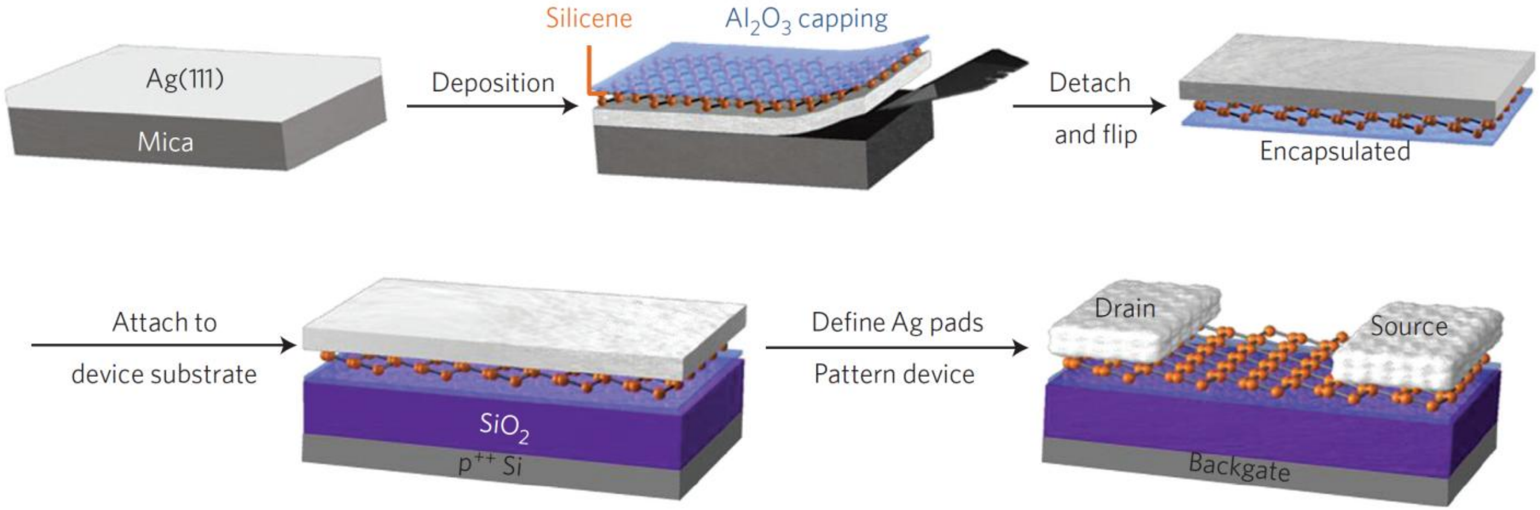
- Six paired Dirac cones at the edge of the Ag(111) BZ
- Does not exist for Ag(111) or free-standing silicene
- This dispersion is generated by the Ag-silicene interaction
- The band structure is not explaind by existing DFT calculations

The band structure for silicene / Ag(111) is still not fully understood

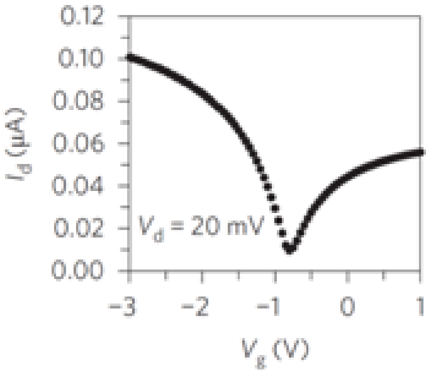
A FET based on silicene/Ag(111)

Li Tao et al., Nature Nanotechnol. **10**, 27 (2015)

“Silicene encapsulated delamination with native electrodes”



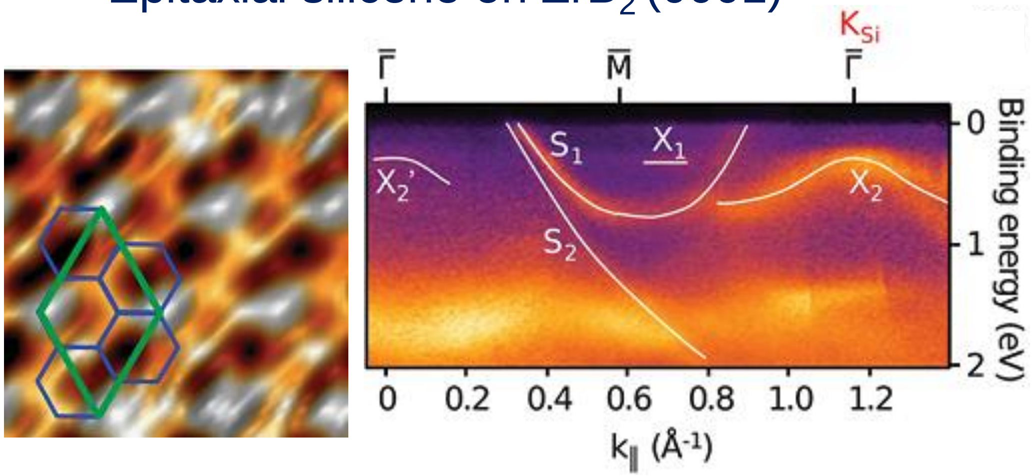
RT electrical characteristics



- ambipolar behavior, on/off ratio ~10
- mobility ~ $100 cm^2 V^{-1} s^{-1}$

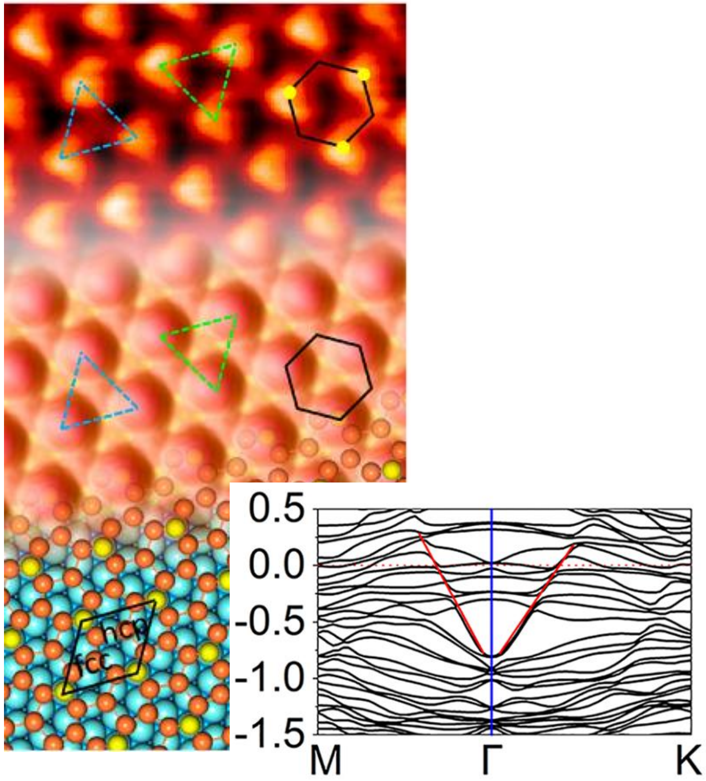
Epitaxial 2D Si layers (other substrates)

Epitaxial silicene on ZrB_2 (0001)



A. Fleurence, *et al.*, Phys. Rev. Lett. **108**, (2012).

Epitaxial silicene on Ir(111)



L. Meng, *et al.*, Nano Lett. **13**, 685 (2013)

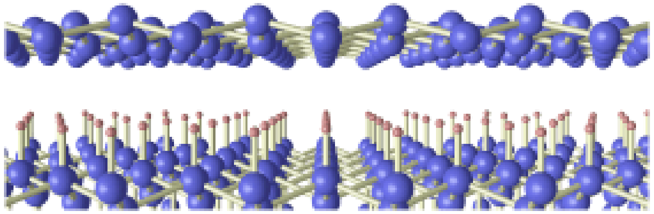
W. Wei, *et al.*,
J. of Phys. Chem. Lett. **6**, 1065 (2015)

Outlook: Alternative substrates for 2D materials

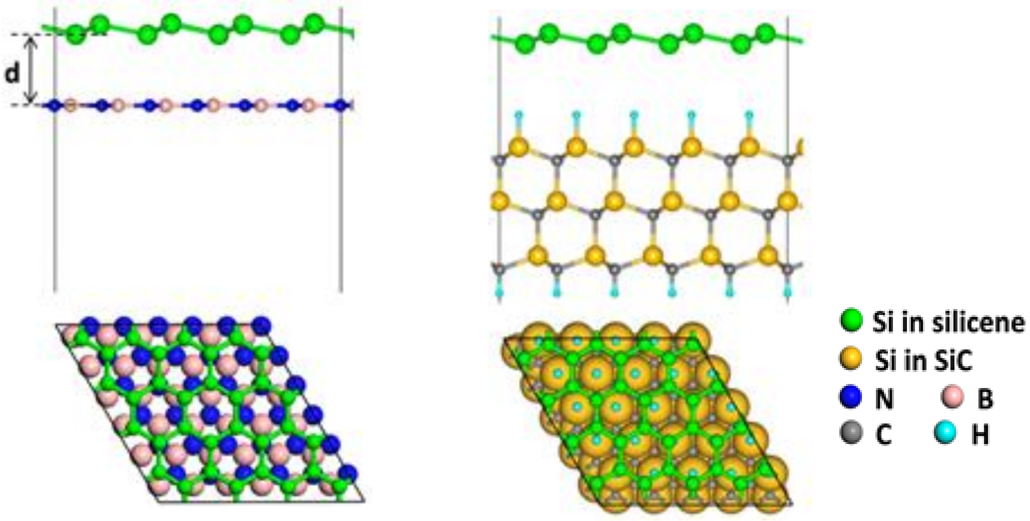
- A possible route for quasi free-standing elemental 2D Materials

Silicene on H- or Cl-terminated Si(111) or Ge(111)

S. Kokott et al., pss RRL (2013)
Guo et al., Phys. Rev. B **87**, 235435 (2013)



- weak interaction
- Dirac properties are preserved



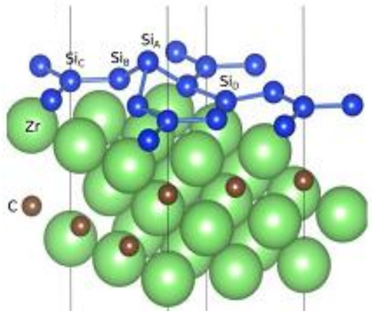
silicene/BN

silicene/H-Si−SiC

- Dirac properties are preserved

H. Liu et al., J. Phys. Chem. C **117**, 10353 (2013)

Epitaxial silicene on ZrC (111)



T. Aizawa, *et al.*,
J. of Phys. Chem. C **118**, 23049 (2014)

Conclusion: 2D epitaxial silicene on Ag(111)

▶ Single-layer 2D epitaxial silicene can be synthesized epitaxially on Ag(111)

- Symmetry and unit cell are modified with respect to ideal free-standing silicene as a result of the substrate interaction

- The atomic model is supported by experimental results and ab initio theory

- Hybridized states form at the silicene / Ag(111) interface
  → linear dispersion

- Dirac cones at the K-points are not preserved

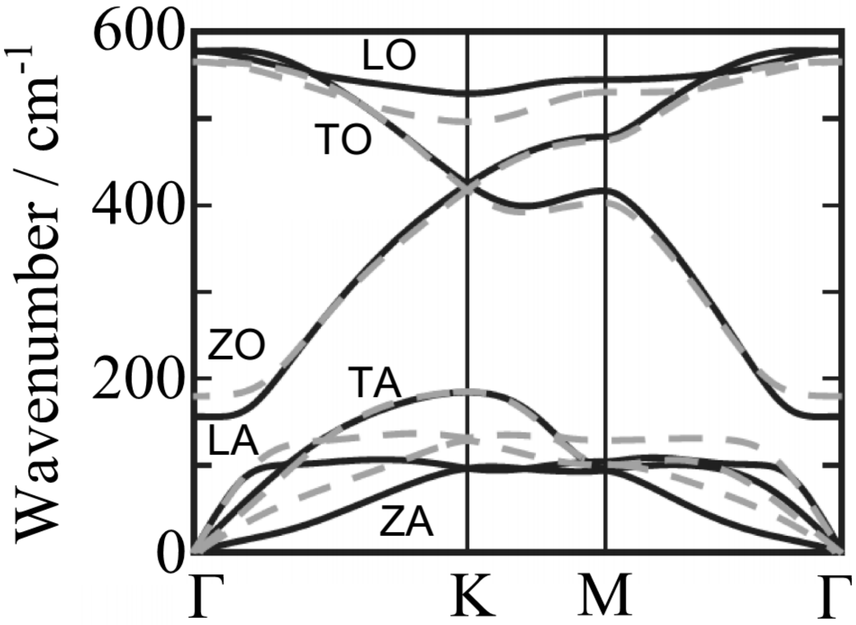
- But: the electronic structure is not fully understood yet!

Outline

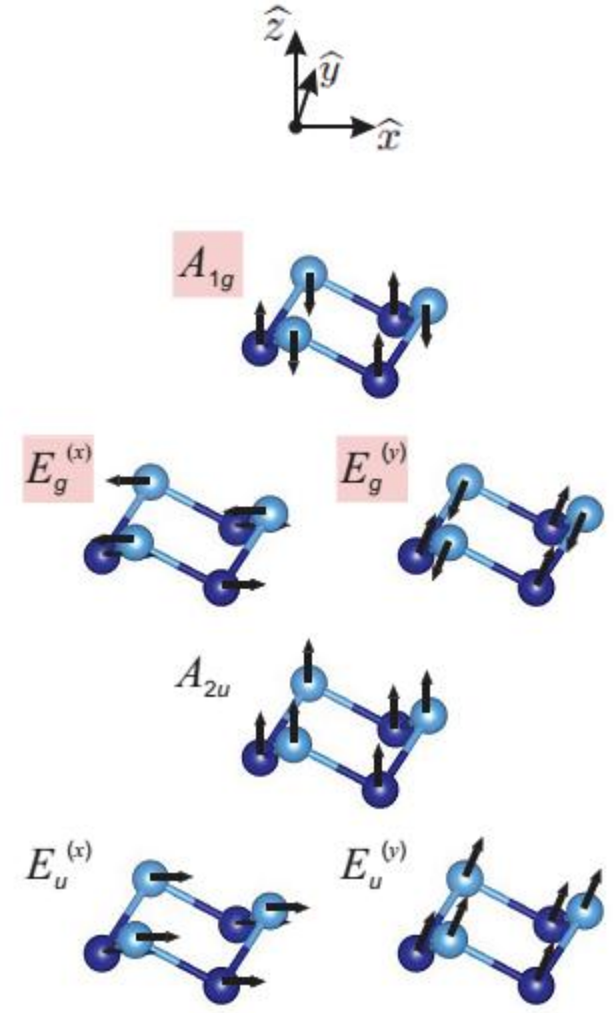
- **part 1:** **- epitaxial growth of silicene on Ag(111)**
- **part 2:** **- Vibrational properties of epitaxial silicene / Ag(111)**

- Other elemental 2D materials

Silicene: vibrational structure



S. Cahangirov, *et al.*, Phys. Rev. Lett. **102**, 236804 (2009)



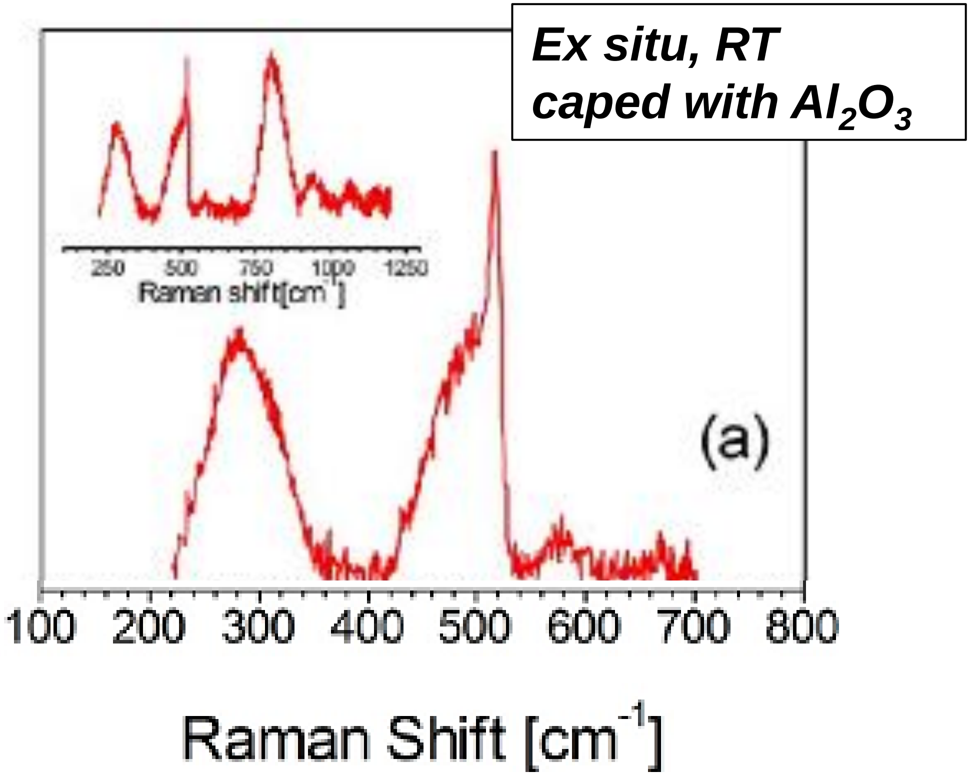
- A-symmetry phonons: out-of-plane motion
- E-symmetry phonons: in-plane motion

J. Ribeiro-Soares, *et al.*, Phys. Rev. B **91**, 205421 (2015)

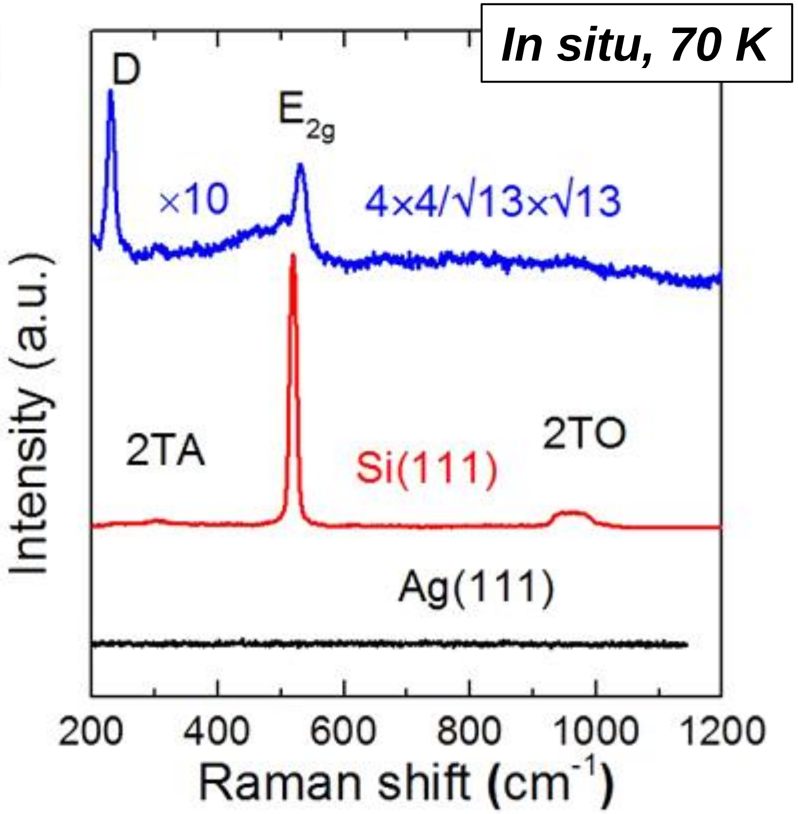
Raman spectra of epitaxial silicene

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

J. Zhuang, *et al.*,
Phys. Rev. B **91**, 161409(R) (2015)



Ex situ, RT
caped with Al_2O_3

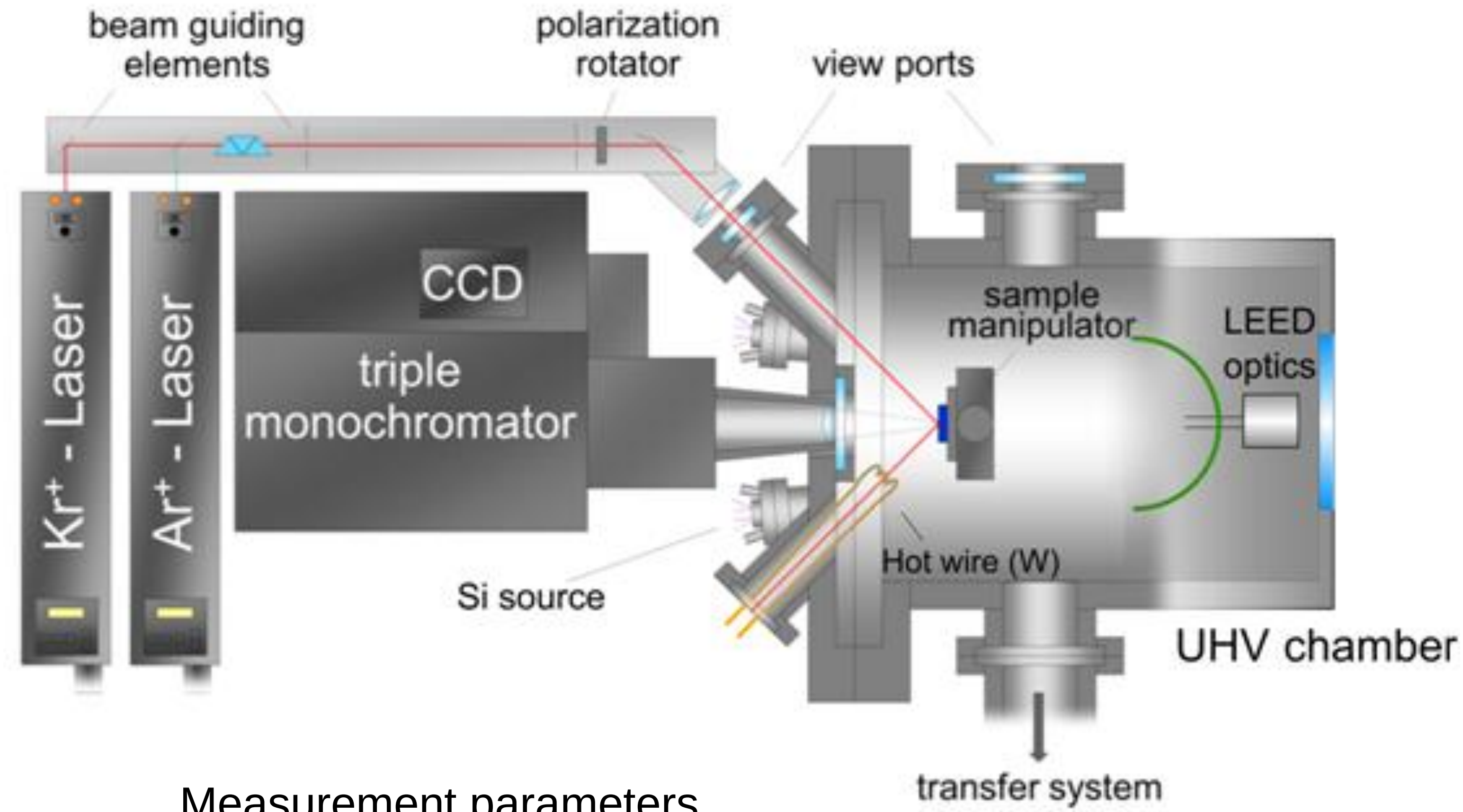


In situ, 70 K

<u>Ex situ, RT</u>
496 cm^{-1} – phonon confinement
516 cm^{-1} – E_{2g}
950 cm^{-1} – “2D”

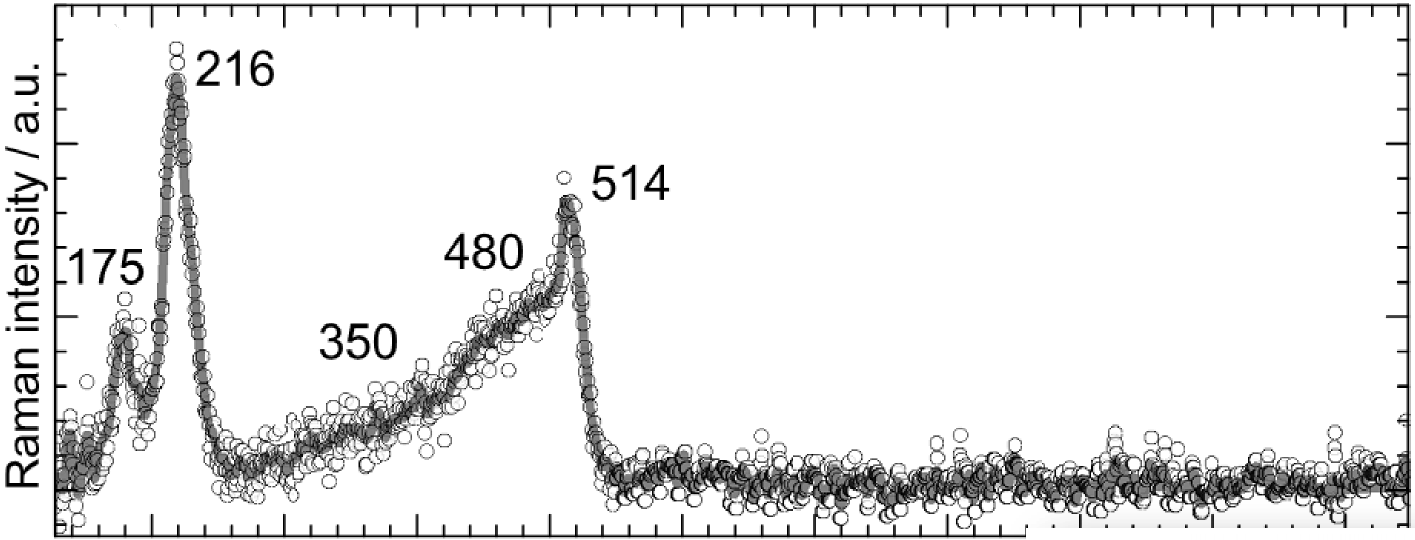
<u>In situ, T = 70 K</u>
230 cm-1 – “D band”
495-508 cm^{-1} – phonon confinement
530 cm^{-1} – E_{2g}

In situ Raman Spectroscopy

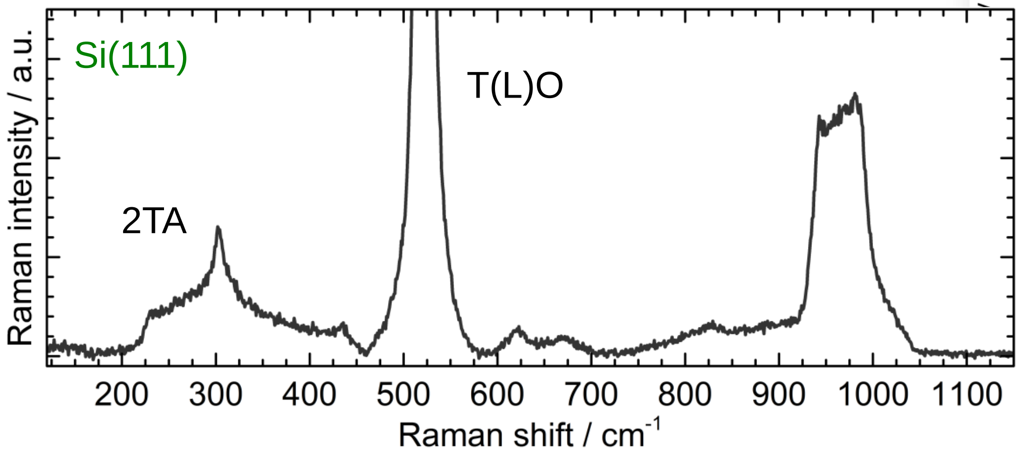


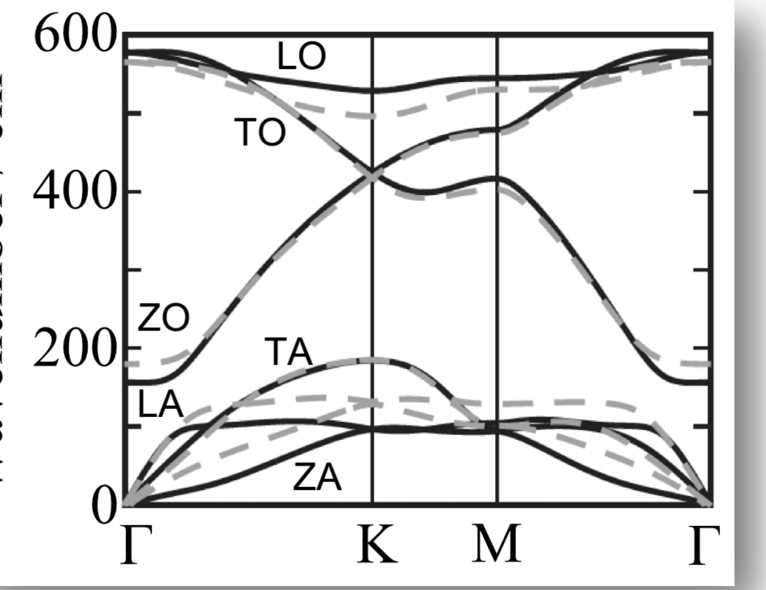
Measurement parameters
- Macro configuration
- Excitation energy: 2.41 eV (514.5 nm Ar laser)
- Laser power density: 10^3 W·cm^{-2}
- Exposure time: 144 s · 25 cycles (1 hour)
- Spectral resolution: ~ 3.3 cm^{-1}

Raman spectrum of epitaxial (3×3)/(4×4) silicene



Parameters
λ_{exc} = 514.5 nm
t_{exp} = 144 s · 25 acc
PD = $5 \cdot 10^2$ W/cm^2

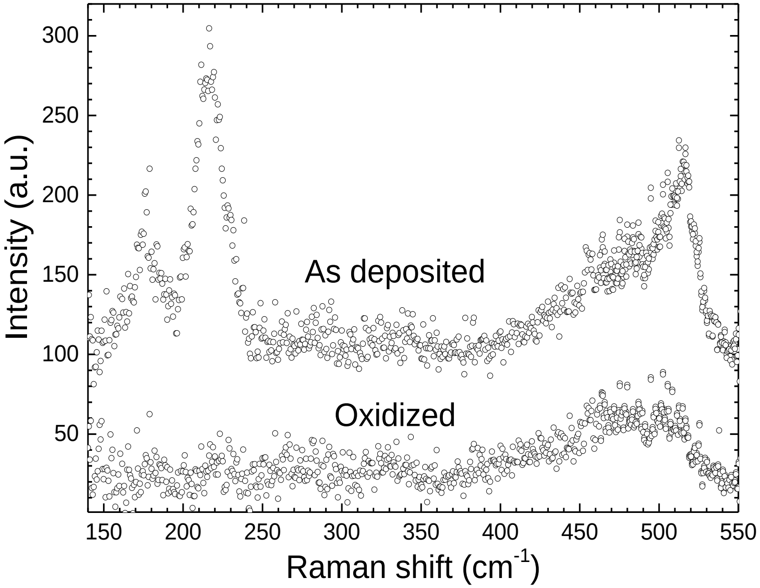




S. Cahangirov, *et al.*, Phys. Rev. Lett. 102, 236804 (2009)

Co-formation of A-Si and (3×3) silicene on Ag(111)

Oxidation

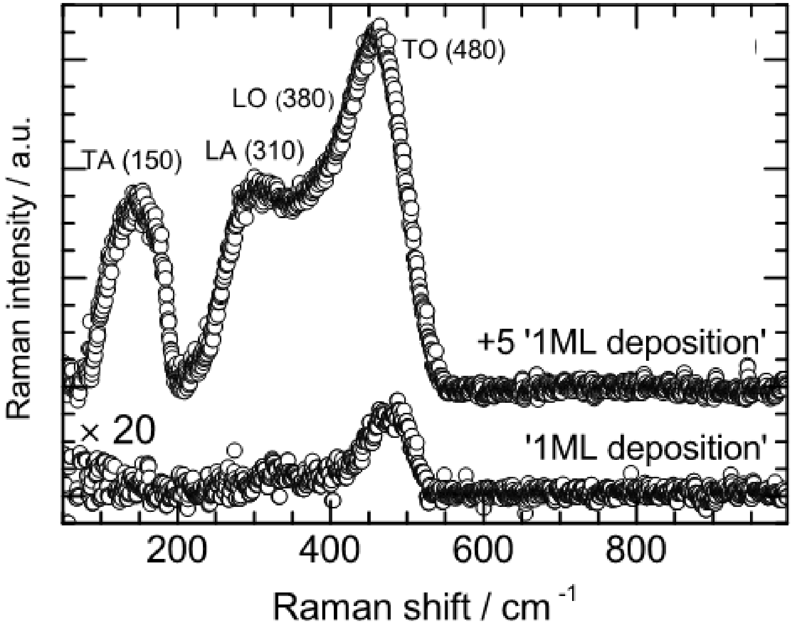


- The silicene related modes vanish upon oxidation

➡ Formation of Si_2O_3

- The signature of a-Si remains

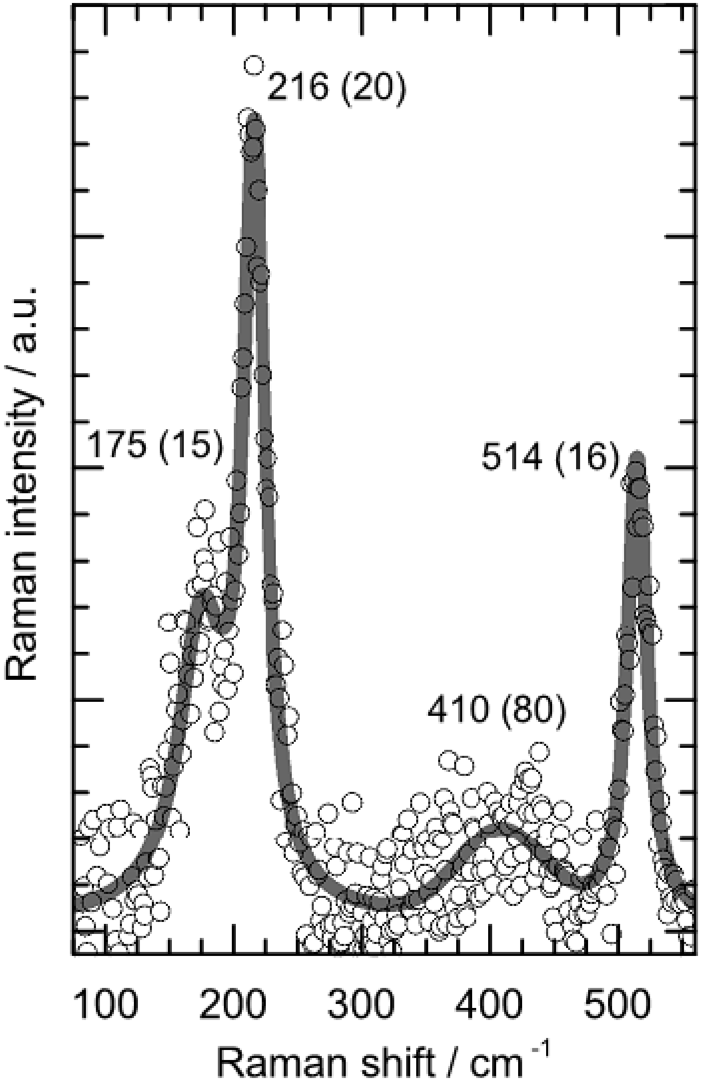
Si deposition at room temperature



- Formation of a-Si at RT

signature of a-Si can be substracted!

The vibrational signature of epitaxial silicene



- 3 Raman modes
- Weak broad mode at ~410 could be a 2nd-order band

D. Solonenko et al., 2D Mater. **4**, 15008 (2017)

Raman spectroscopy

Scattering efficiency:

$$\theta = N_i(\omega_s/c)^4 V L \mid \mathbf{e}_i \cdot \left(\frac{\partial \kappa}{\partial \mathbf{Q}}_0 \mathbf{Q}(\omega_0)\right) \cdot \mathbf{e}_s \mid^2$$

N_i - the number of scatterers;
ω_s - the frequency of the scattered light;
$\mathbf{e}_i$, $\mathbf{e}_s$ – polarization vectors of incident and scattered light
V - the scattering volume
L - the scattering length
$\mathbf{Q}$ - the displacement vector
κ - the electric susceptibility

Raman tensor:

$$\Re = \frac{\partial \kappa}{\partial \mathbf{Q}}_0 \mathbf{Q}(\omega_0)$$

→

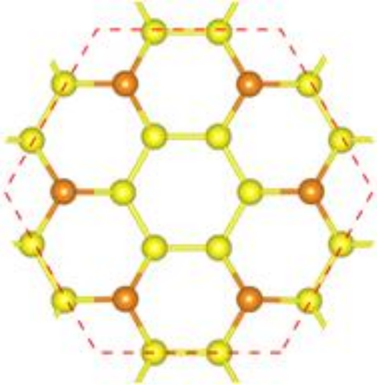
$$I \sim \mid \mathbf{e}_i \cdot \Re \cdot \mathbf{e}_s \mid^2$$

$$I_{Raman}(ij) \sim \sum_s |\alpha_{ij}|^2$$

α_{ij} are the matrix elements of the Raman tensor

Raman election rules for C_{6v}

Symmetry group: C_{6v}



Raman tensors:

www.cryst.ehu.es

$$A(z)=\begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix};\quad E_1=\begin{pmatrix} 0 & 0 & c \\ 0 & 0 & c \\ c & c & 0 \end{pmatrix};\quad E_2=\begin{pmatrix} d & -d & 0 \\ -d & -d & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Selection rules:

A symmetry

Parallel scattering configuration

$$\left|\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \times \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix} \times \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}\right|^2 = a^2$$

Crossed scattering configuration

$$\left|\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \times \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix} \times \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}\right|^2 = 0$$

E symmetry

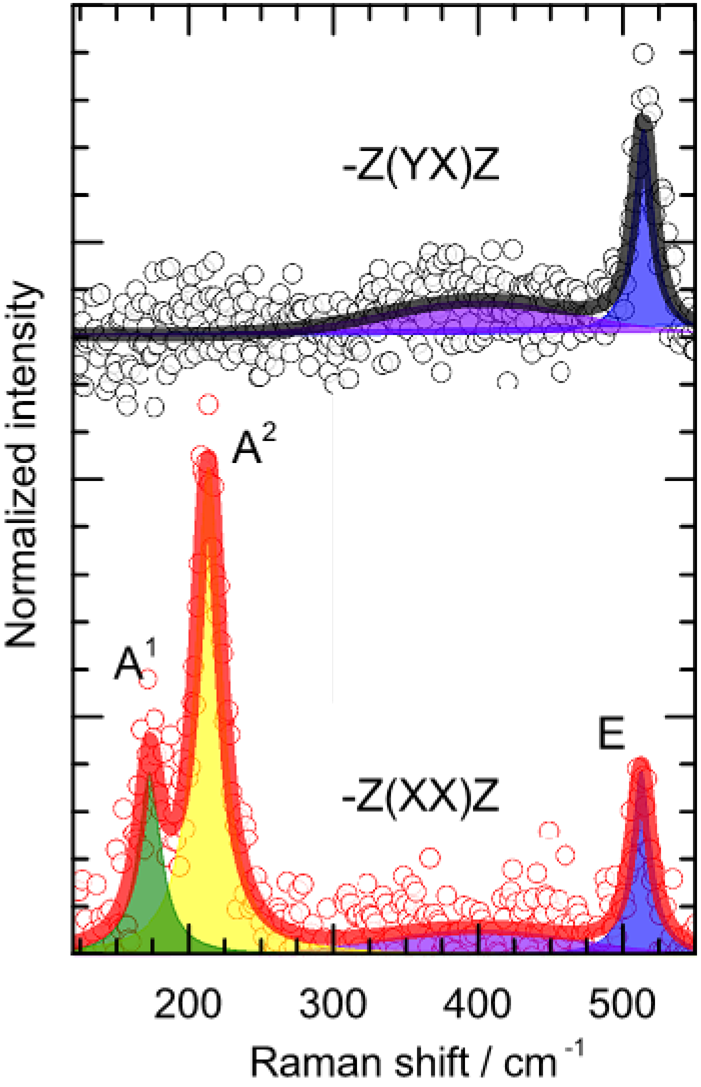
Parallel scattering configuration

$$\left|\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \times \begin{pmatrix} d & -d & 0 \\ -d & -d & 0 \\ 0 & 0 & b \end{pmatrix} \times \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}\right|^2 = d^2$$

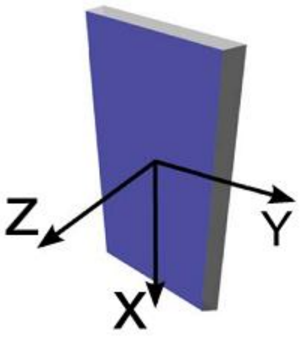
Crossed scattering configuration

$$\left|\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \times \begin{pmatrix} d & -d & 0 \\ -d & -d & 0 \\ 0 & 0 & b \end{pmatrix} \times \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}\right|^2 = (-d)^2$$

Polarization-dependent Raman measurements



Geometry

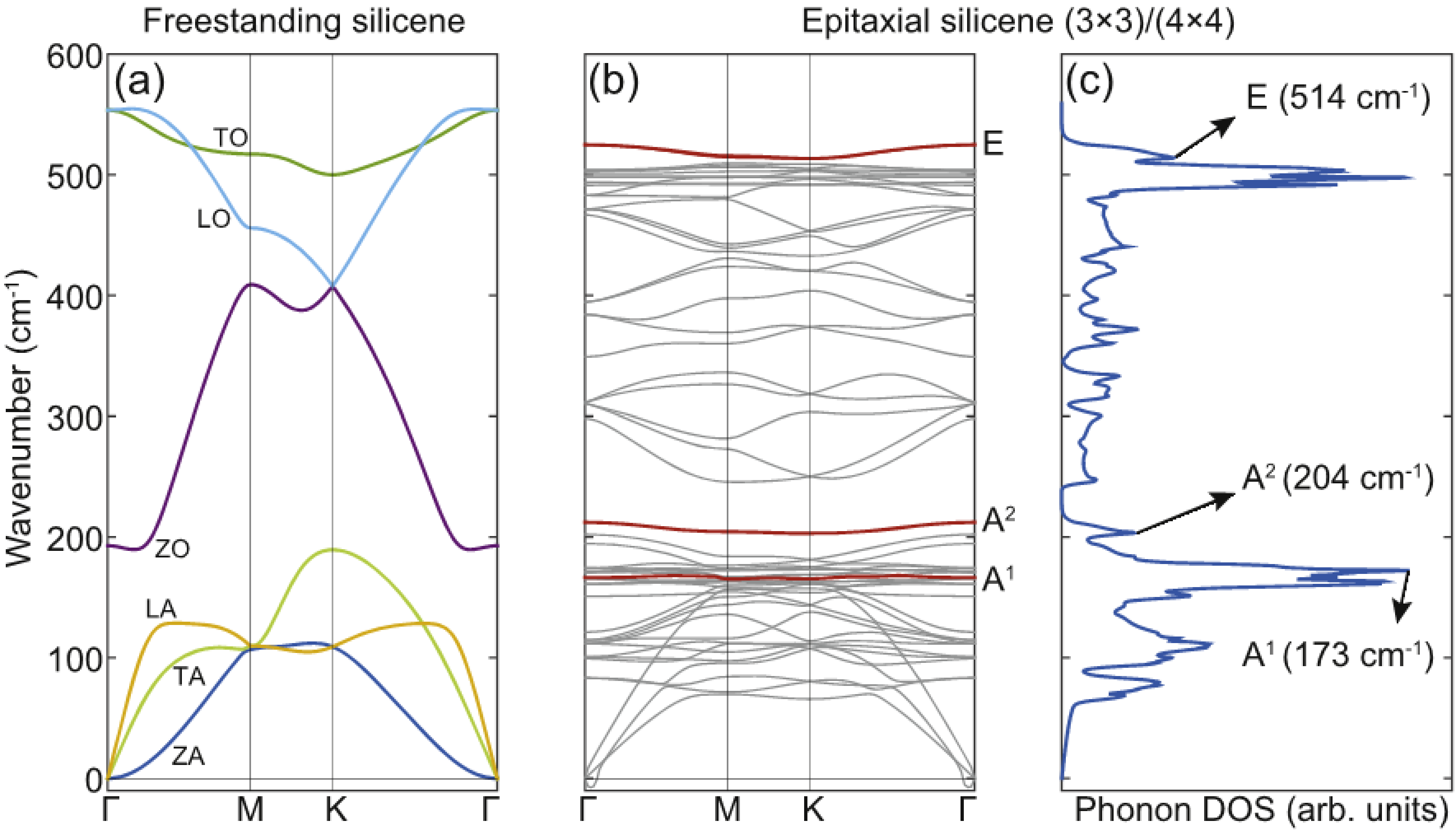


The observed Raman modes follow very clearly the selection rules

Epitaxial silicen / Ag(111) has:

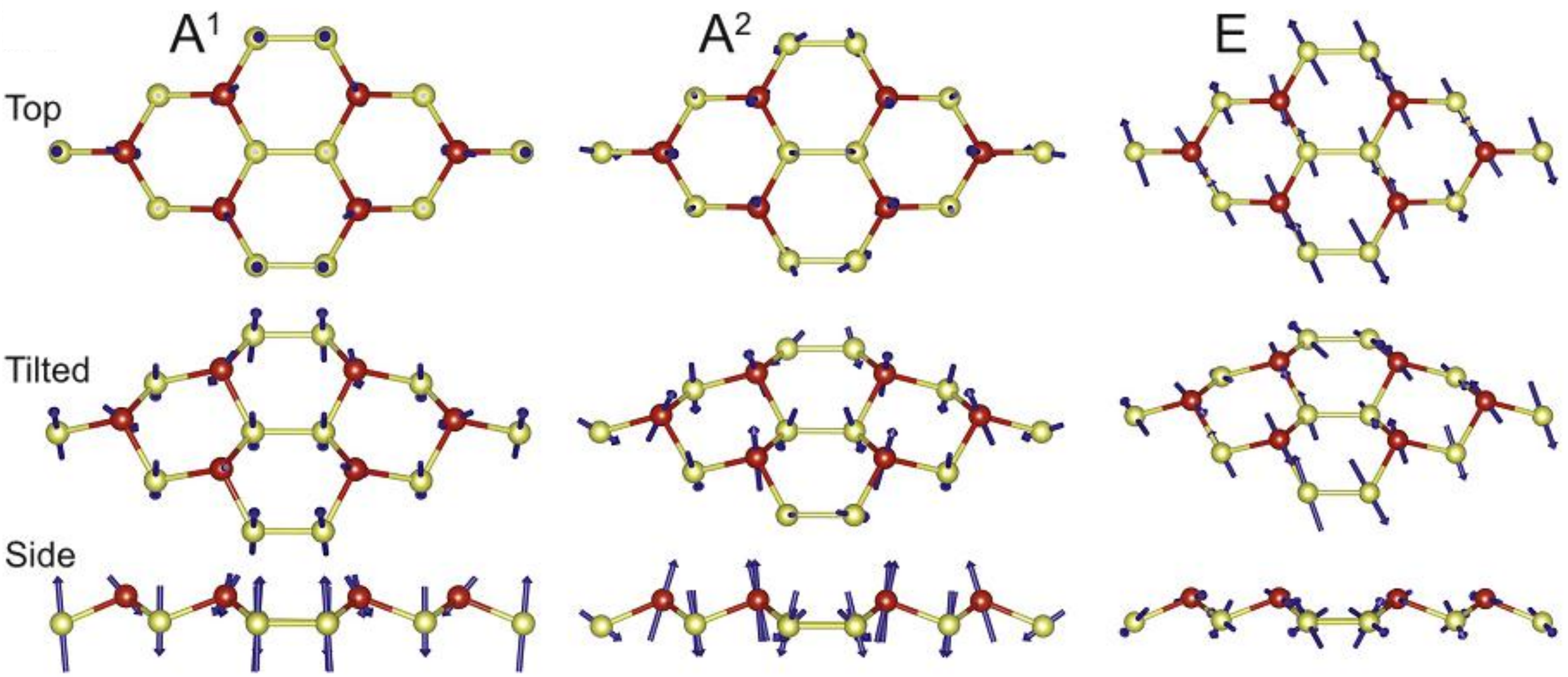
- 2 A-modes: 176 cm^{-1} and 216 cm^{-1}
- 1 E_{2g}-mode at 514 cm^{-1}

Vibrational properties of epitaxial silicene: DFT



2D Mater. **4**, 15008 (2017)

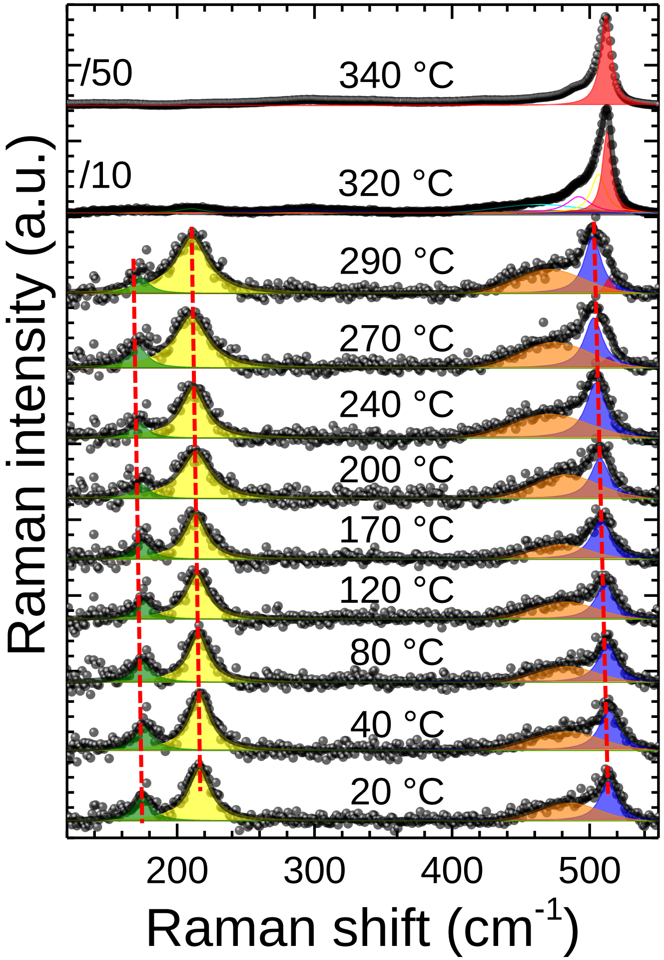
A and E modes: motion patterns



- A-symmetry phonons: out-of-plane motion
- E-symmetry phonons: in-plane motion

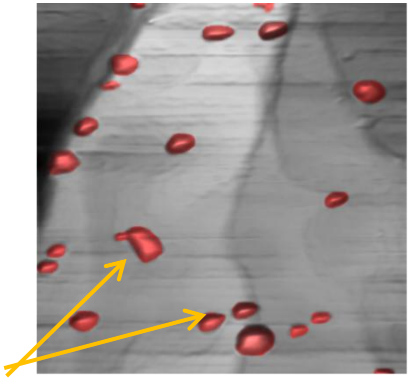
2D Mater. **4**, 15008 (2017)

Structural phase transition of (3×3) silicene



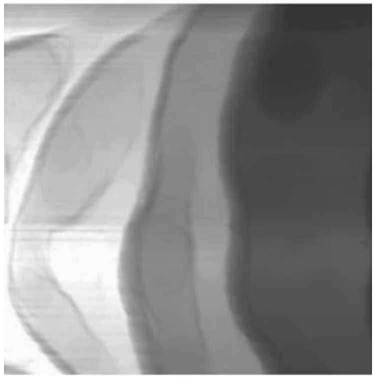
phase transition

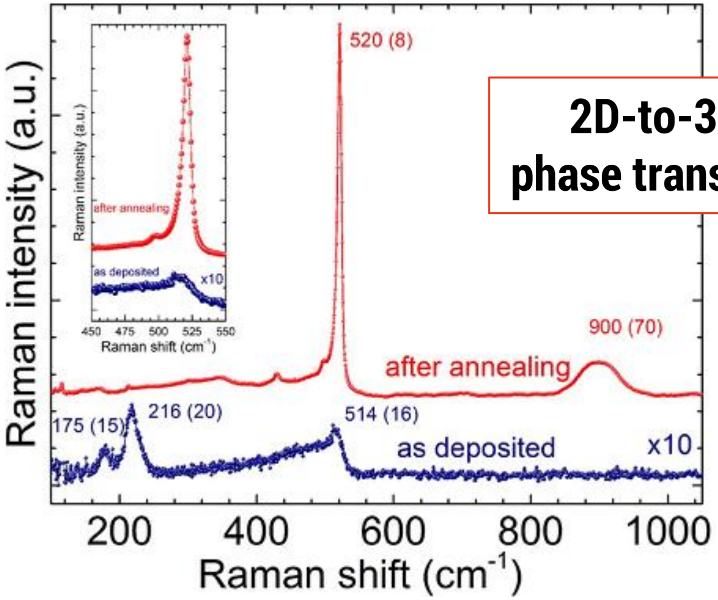
AFM after annealing



Si NPs

Initial Ag(111) surface



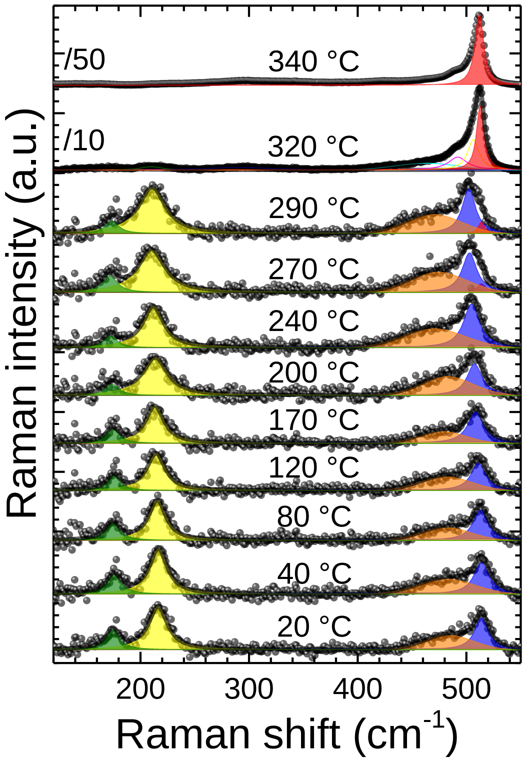


2D-to-3D phase transition

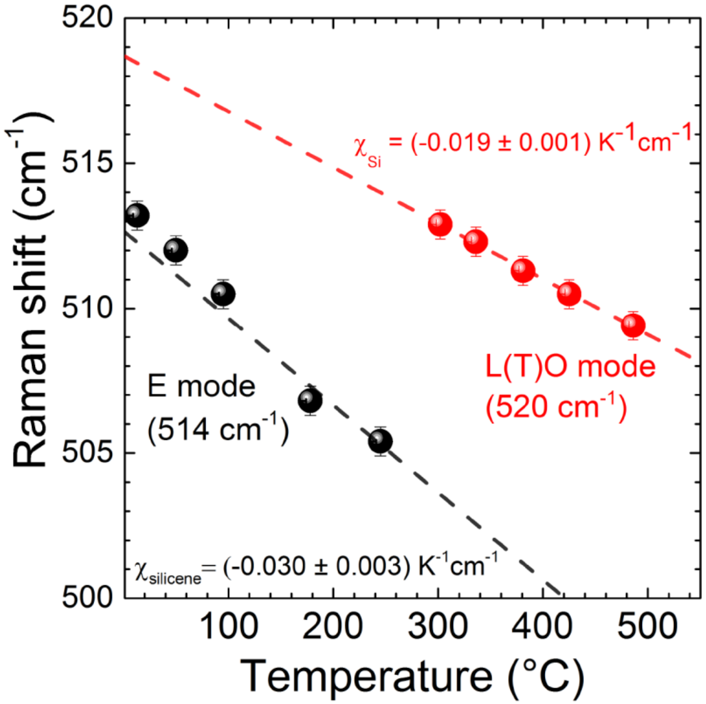
At ~300°C silicene transforms into Si nano-particles

Temp. dependence of the vibrational properties

In situ Raman



Thermal coefficient



- The thermal coefficient of the E-mode is clearly larger than the L(T)O
- Similar to: graphene : 0.016 $K^{-1}cm^{-1}$ ➡ diamond: 0.0078 $K^{-1}cm^{-1}$

Conclusion: Raman on epitaxial silicene

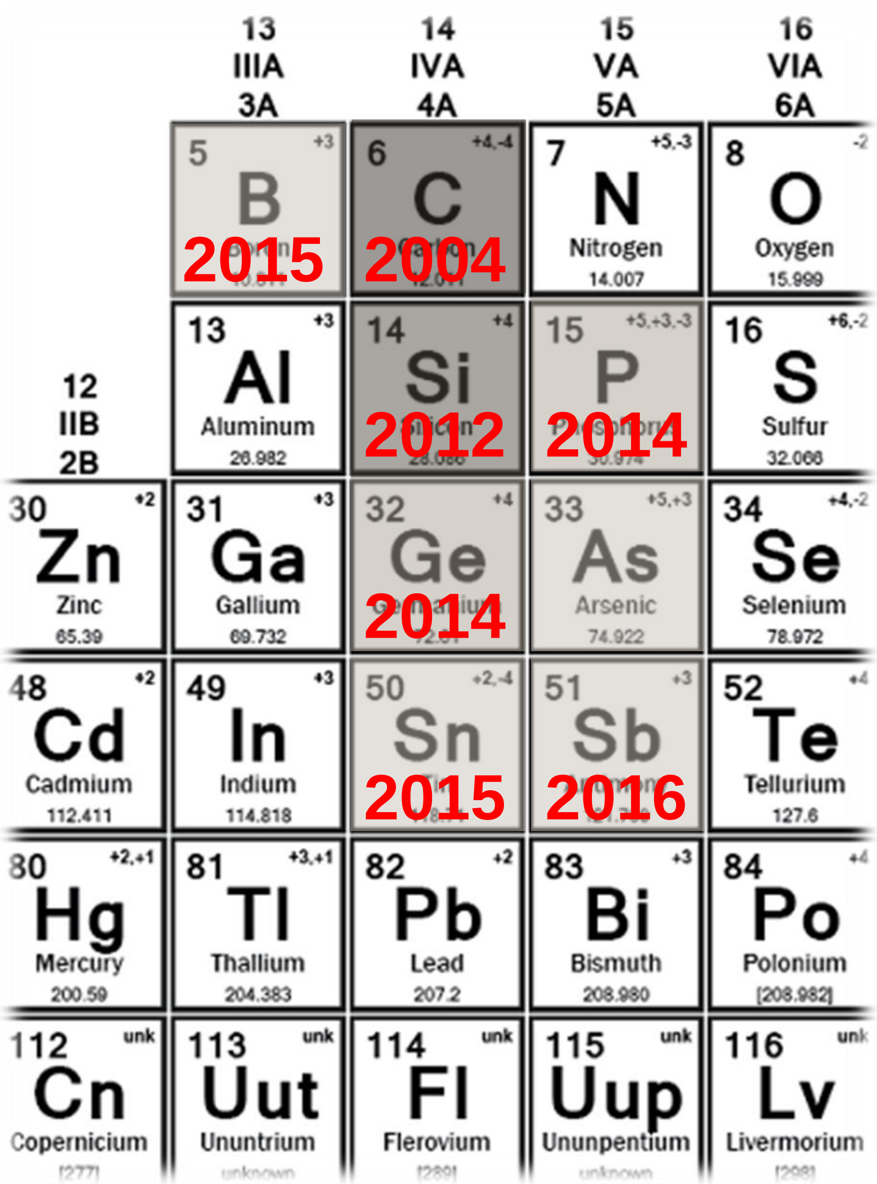
- The *in situ* Raman results confirm the 2D character of epitaxial (3×3) silicene/Ag(111)
  - out-of-plane A-modes
  - comparison to theory results
  - Temperature dependence
  - 2D/3D phase transition at ~ 300°C

Outline

- **part 1:** **- epitaxial growth of silicene on Ag(111)**
- **part 2:** **- Vibrational properties of epitaxial silicene / Ag(111)**

- Other elemental 2D materials

Elemental 2D Materials



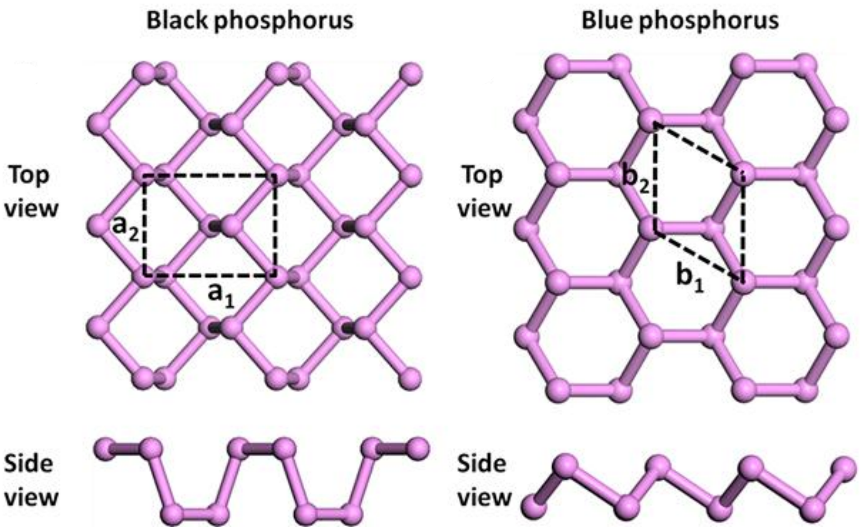
12 IIB 2B	13 IIIA 3A	14 IVA 4A	15 VA 5A	16 VIA 6A
	5 +3 B 2015	6 +4,-4 C 2004	7 +5,-3 N Nitrogen 14.007	8 -2 O Oxygen 15.999
	13 +3 Al Aluminum 26.982	14 +4 Si 2012	15 +5,+3,-3 P 2014	16 +6,-2 S Sulfur 32.066
30 +2 Zn Zinc 65.39	31 +3 Ga Gallium 69.732	32 +4 Ge 2014	33 +5,+3 As Arsenic 74.922	34 +4,-2 Se Selenium 78.972
48 +2 Cd Cadmium 112.411	49 +3 In Indium 114.818	50 +2,-4 Sn 2015	51 +3 Sb 2016	52 +4 Te Tellurium 127.6
80 +2,+1 Hg Mercury 200.59	81 +3,+1 Tl Thallium 204.383	82 +2 Pb Lead 207.2	83 +3 Bi Bismuth 208.980	84 +4 Po Polonium [208.982]
112 unk Cn Copernicium [277]	113 unk Uut Ununtrium unknown	114 unk Fl Flerovium [289]	115 unk Uup Ununpentium unknown	116 unk Lv Livermorium [298]

Phosphorene

Free-standing 2D black phosphorus

Band gap: 1.45 eV

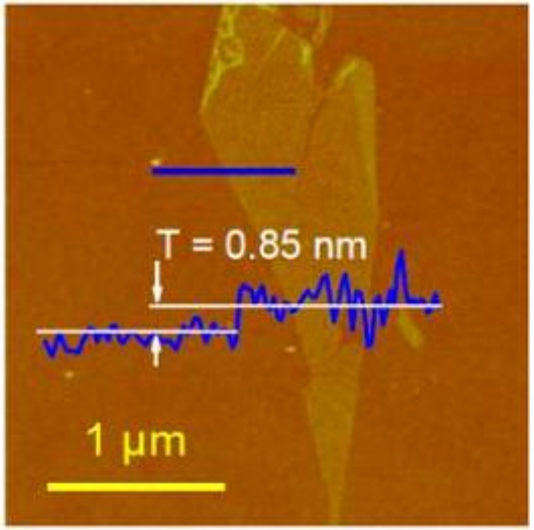
H. Liu, *et al.*,
ACS Nano **8**,
4033 (2014)

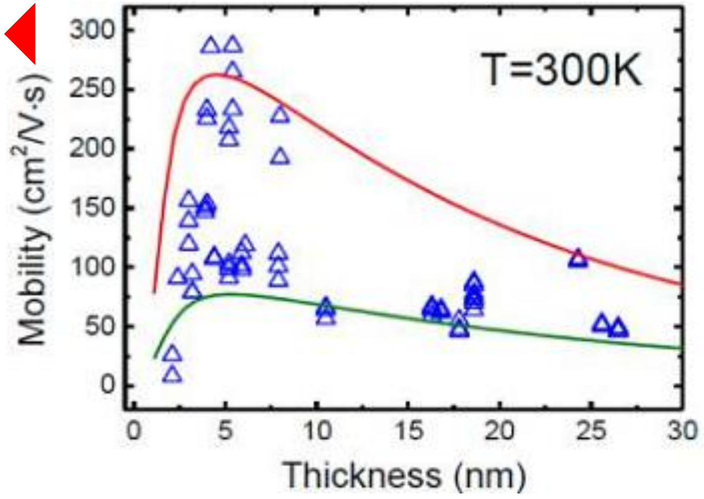


Blue phosphorus on Au(111)

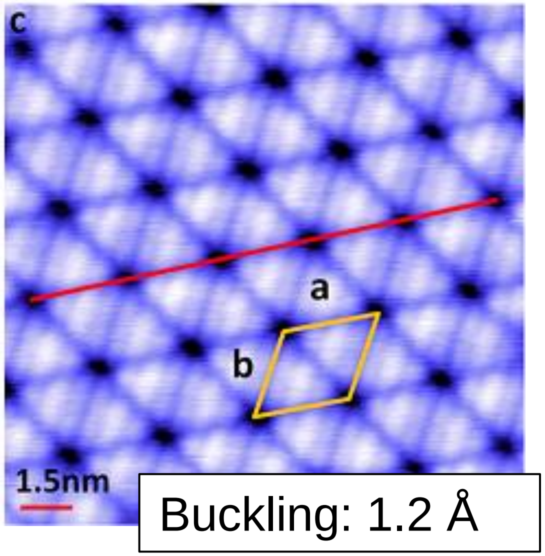
Band gap: 1.1 eV

J.L. Zhang, *et al.*,
Nano Letters **16**,
4903 (2016)





Its mobility is 4x higher than MoS_2

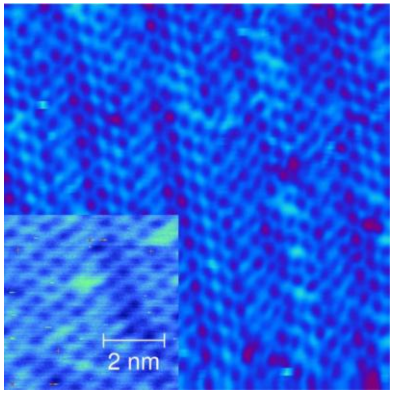


Buckling: 1.2 Å

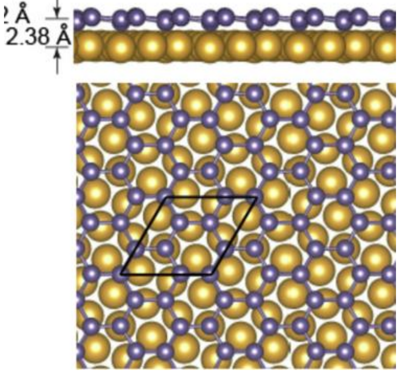
Outlook: Synthesis of germanene

Germanene/Au(111) Dávila et al. New J. Phys. **16**, 095002 (2014)

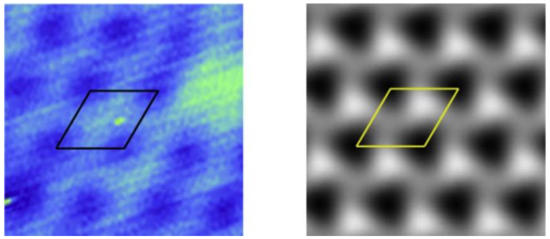
STM



√7×√7 germanene

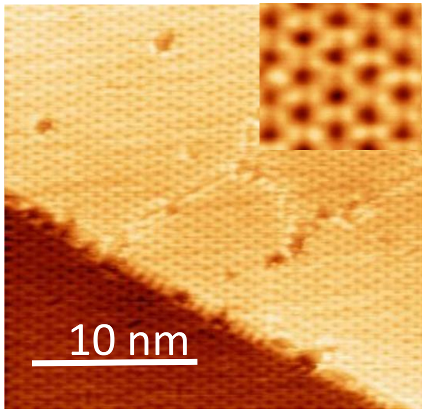


comparison with theory



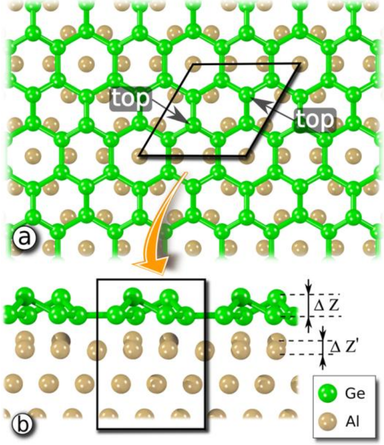
Germanene/Al(111) Derivaz et al., Nano Lett. **15**, 2510 (2015)

STM

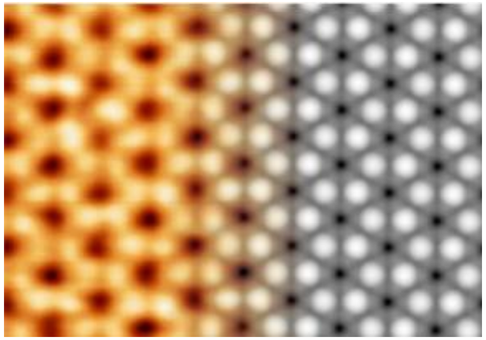


V = 1.3V

3×3 germanene



comparison with theory



Outlook: Stanene

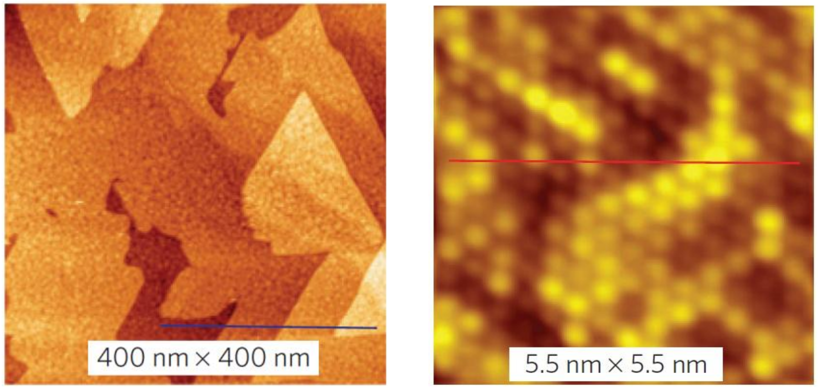
Expected properties of stanene

- Very strong effective spin-orbit coupling → topologically non trivial with a Quantum Spin Hall Effect markedly above room temperature
  Y. Xu et al., PRL **111**, 136804 (2013)
- High Tc superconductivity?

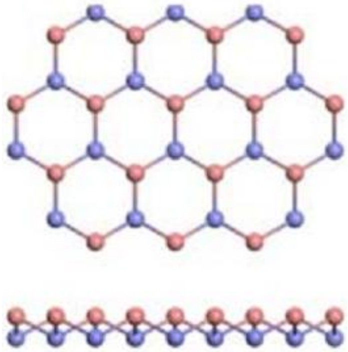
Stanene / $Bi_2Te_3(111)$

Zhu et al. Nature Mat.**14** 1020 (2015)

STM



Model



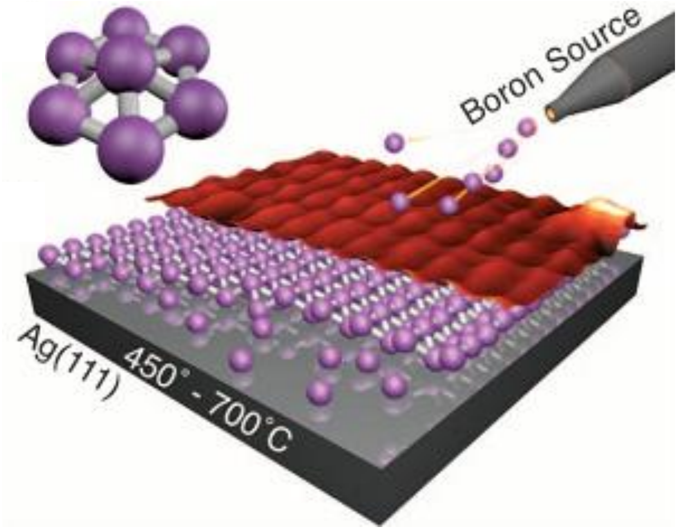
And: 3D topological states were observed for α-Sn(001) / InSb

Barfuss et al. PRL.**111**, 157205 (2015)   Ohtsubo et al. PRL.**111**, 216401 (2015)

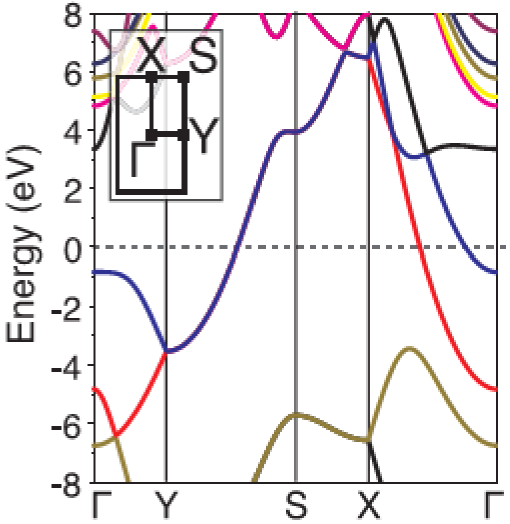
➡ Stanene could provide topological insulating states

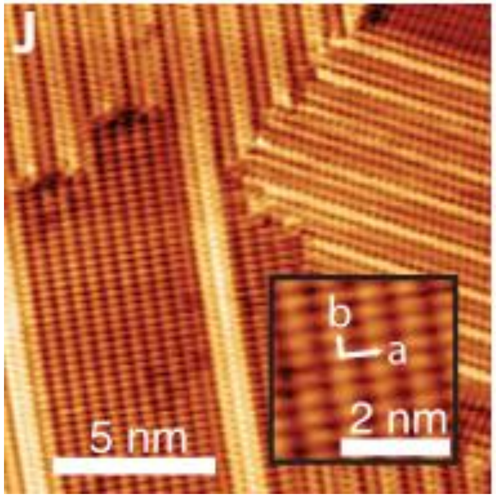
Borophene

A.J. Mannix, *et al.*, Science **350**, 1513 (2015).

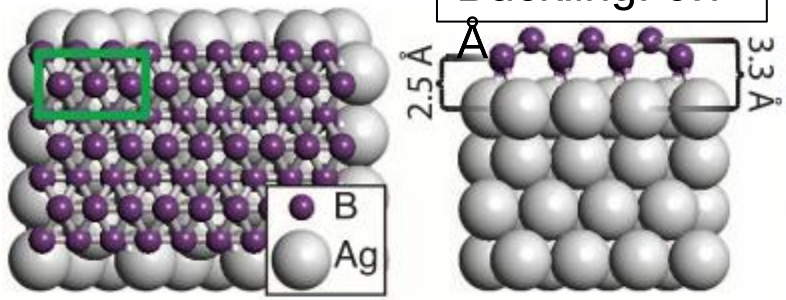


2D boron layers are metallic!



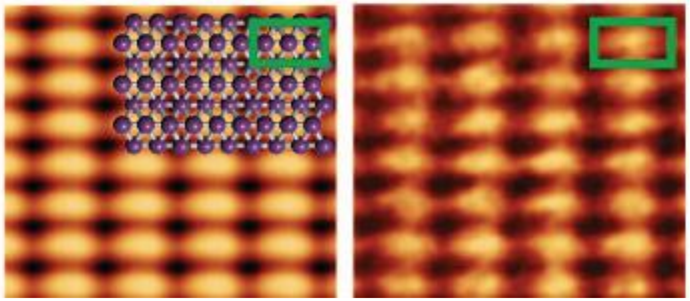


Buckling: 0.7 Å



Hexagonal lattice (no honeycombs)

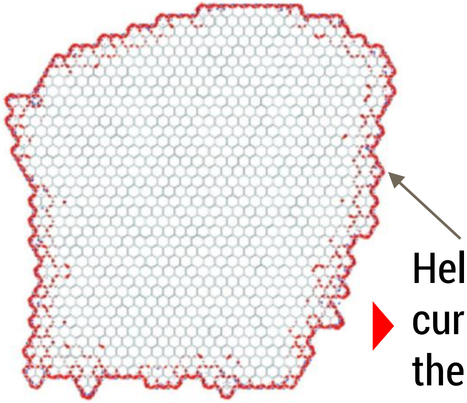
STM images of epitaxial borophene



Simulated Experimental

Topological properties of 2D materials

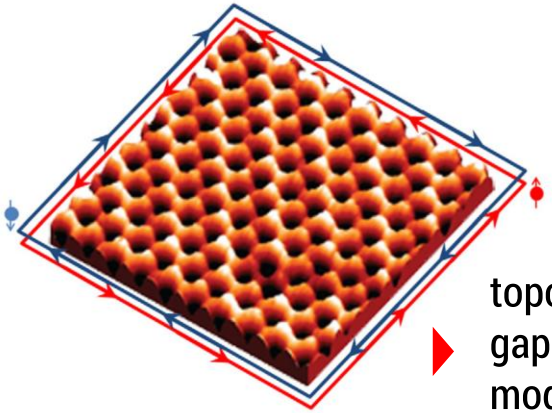
Edge states in silicene nanodisks



► Helical edge currents flow around the edge of a nanodisk

Kikutake et al., PRB **88**, 115432 (2013)

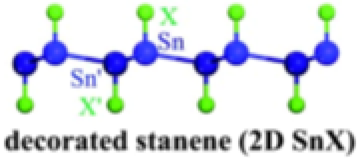
Quantum Spin Hall Effect in germanene

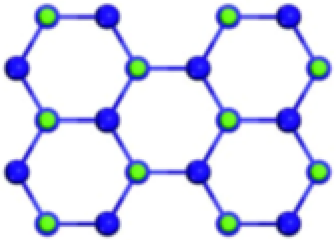


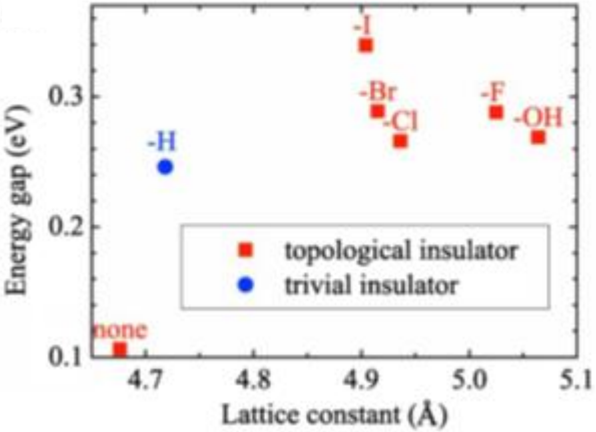
► topological protected gapless helical edge modes

A. Acun et al., J. Phys.: Condens. Mat. **27** (2015) 443002

Topological properties of decorated stanene







Xu et al., Phys. Rev. Lett 111, 136804 (2013)

Thank you for your attention!