Some Interesting Self-assembled nano-structures of Pb/Si(111)

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Outline

1. **QSE driven growth mode**

   a. multi-layered islands

   b. comparison between different interfaces

2. **Stability of QSE driven islands**
3. **Applicability of nucleation theory**
4. **Low coverage structures**

$$\sqrt{3}\times\sqrt{3}-\beta \qquad \sqrt{7}\times\sqrt{3} \qquad \sqrt{43}\times\sqrt{3}$$

$$\sqrt{3}\times\sqrt{3}-\mathrm{HIC} \qquad \sqrt{3}\times\sqrt{3}-\mathrm{SIC}$$

Anything else?

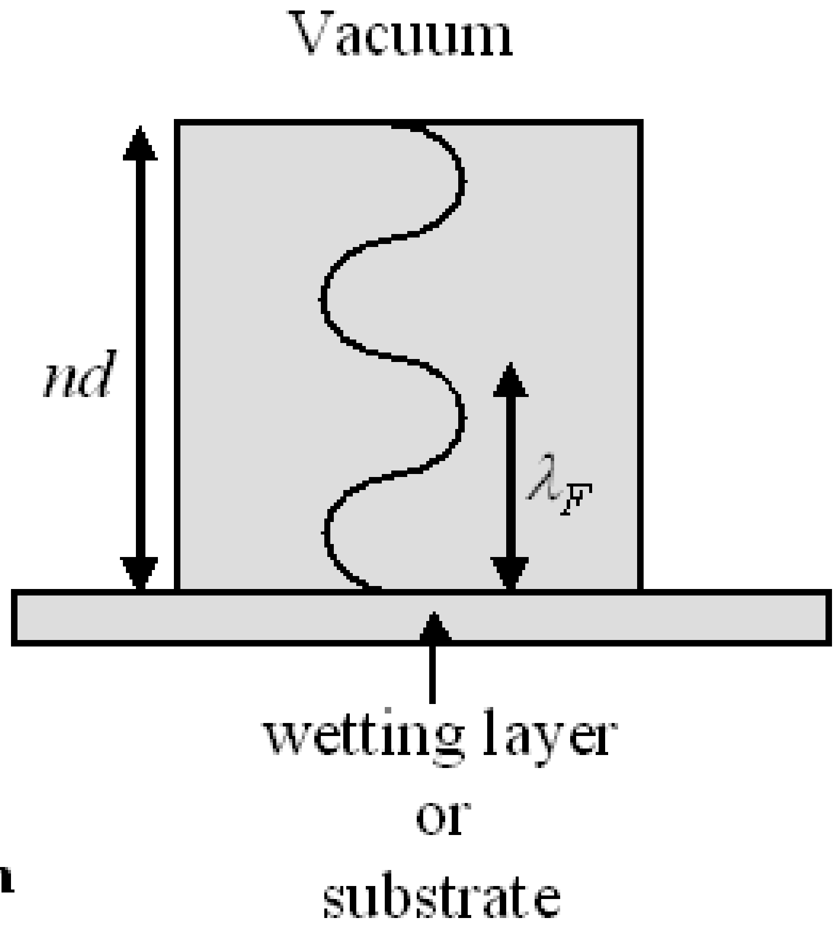
QSE

Driven

Growth Mode

Quantum size effects (QSE)

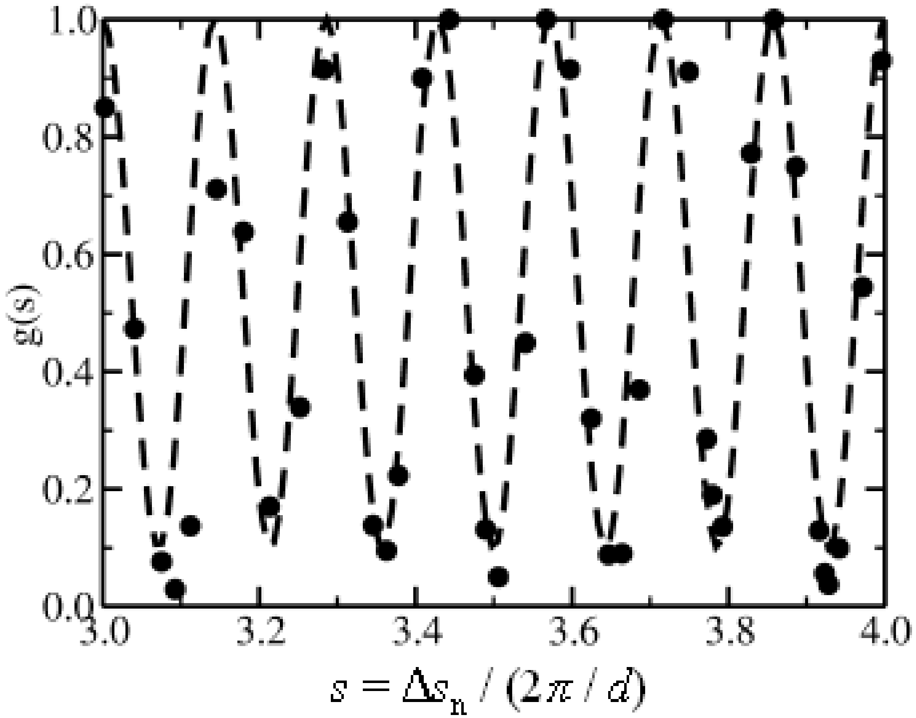
$$nd = s\lambda_F / 2 \quad (n, s \text{ integers})$$



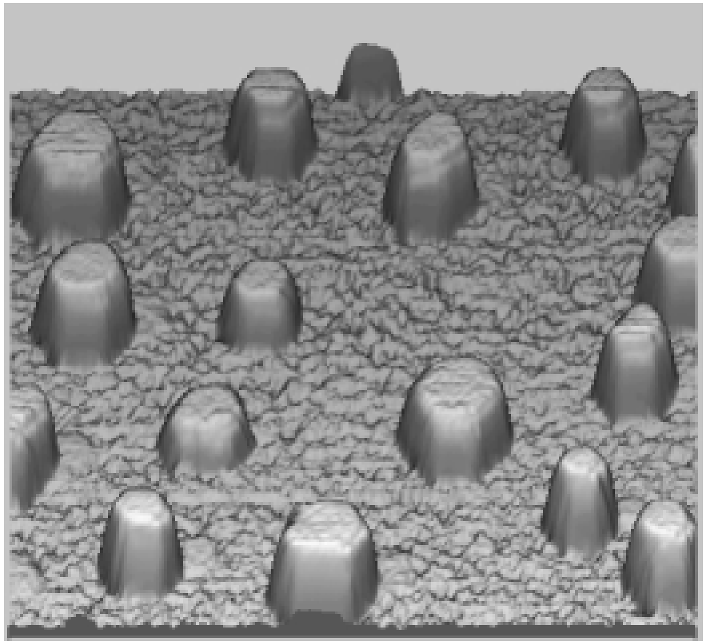
$d = 0.286$ nm

$\lambda_F = 0.395$ nm

$\theta = 4$ ML, $T = 192$ K



$\theta = 3.5$ ML, $T = 190$ K



Novel growth mode: 7-layer high Pb islands with steep edges and flat tops grown on Si(111)-7×7 between 180K and 240K.

K. Budde, E. Abram, V. Yeh, and M.C. Tringides, PRB 61, 10602 (2000)

Quantum well states of Pb islands

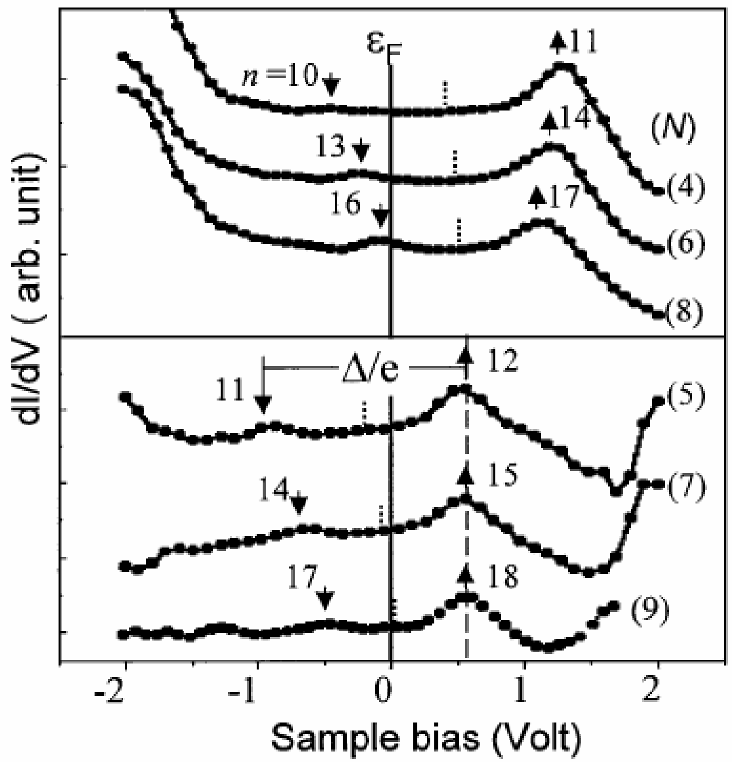


FIG. 2. A series dI/dV spectra taken on individual islands of varying thickness numbered in parentheses from $(N) = (4)$ to (9). The lower case n indicates the quantum number associated with the quantized state. The HOS are marked with point-down arrows and the LUS are marked with point-up arrows. The short-dotted lines mark the middle positions between the HOS and the LUS.

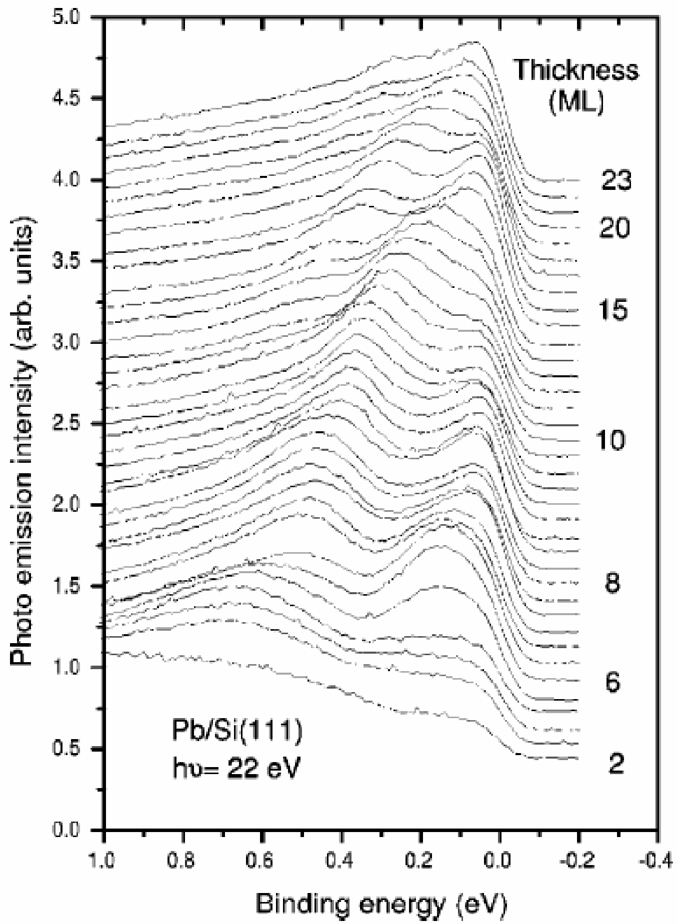
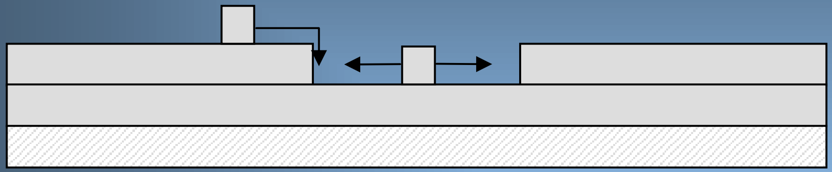


FIG. 1. Normal-emission photoemission spectra of Pb on Si(111)7×7. The coverage ranges from 2 to 23 ML.

W.B. Su, S.H. Chang, W.B. Jian, C.S. Chang, L.J. Chen, and T.T. Tsong, PRL 86, 5116 (2001)

A. Mans*, J.H. Dil, and A. R. H. F. Ettema, H. H. Weitering, PRB 66, 194410 (2002)

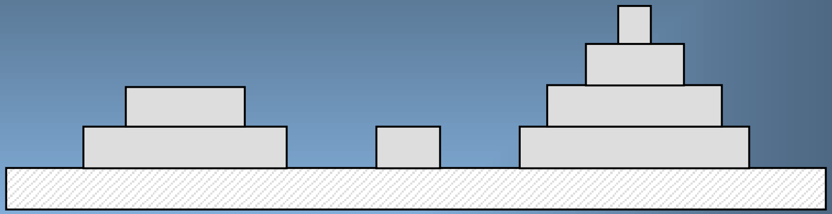
Different modes of thin film growth



Layer by layer

$\Delta\gamma <= 0$

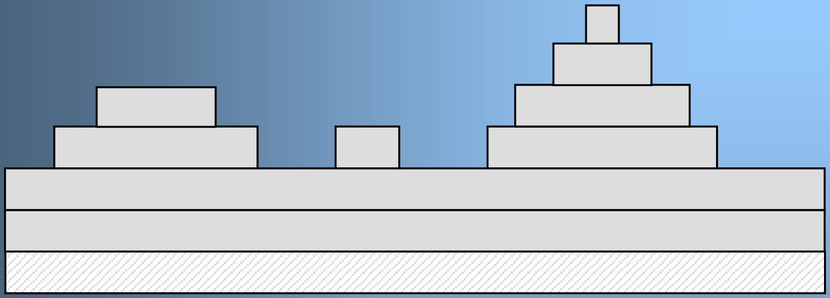
(Frank-van der Merwe)



Three dimensional

$\Delta\gamma > 0$

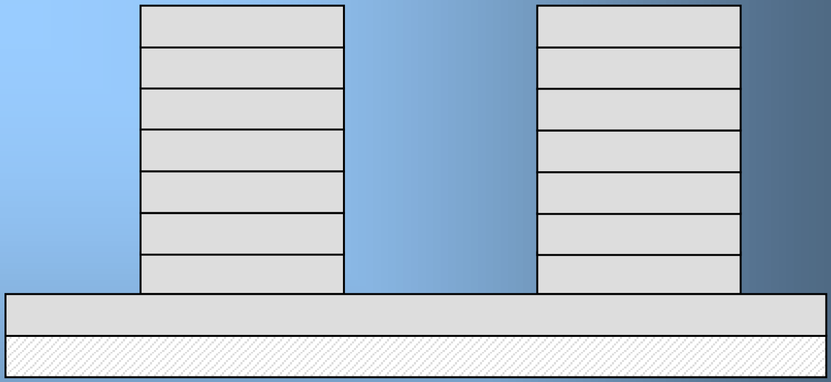
(Volmer-Weber)



Mixed

$\Delta\gamma < 0$, then change sign

(Stranski-Kratanov)

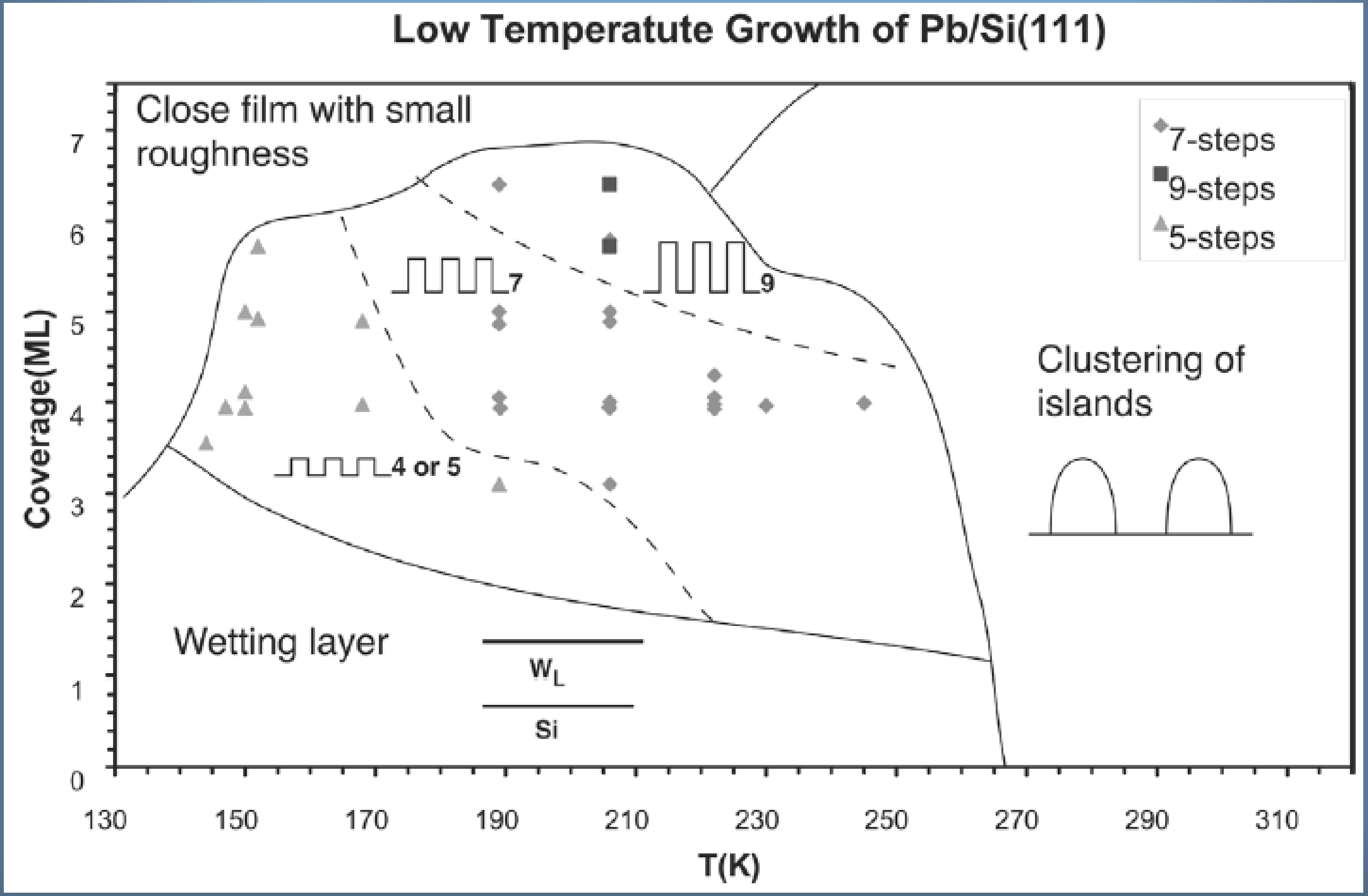


Newly discovered

7-layer high, flat top, steep edge

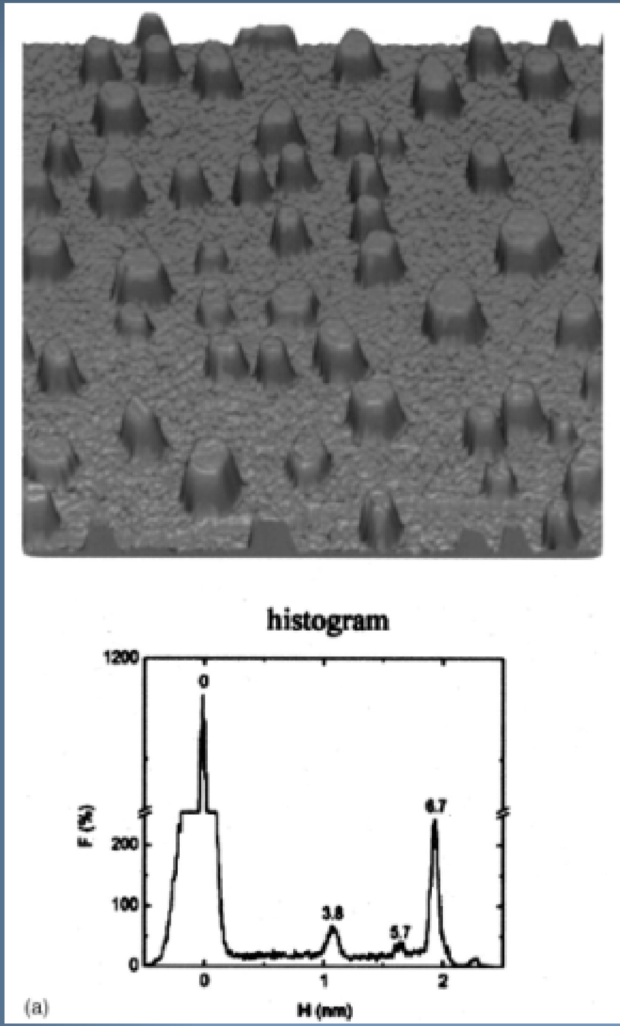
Quantum size effects (QSE)

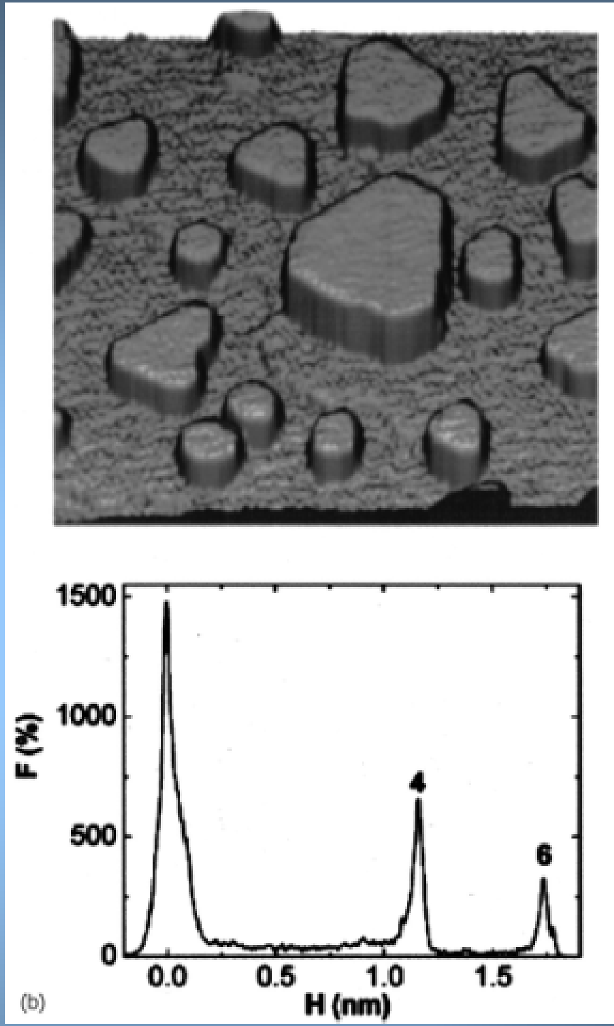
Different stable heights with bi-layer difference under different conditions



M. Hupalo, S. Kremmer, V. Yeh, L. Berbil-Bautista, E. Abram, M.C. Tringides, Surf. Sci. 493 (2001) 526

Comparison between interfaces.

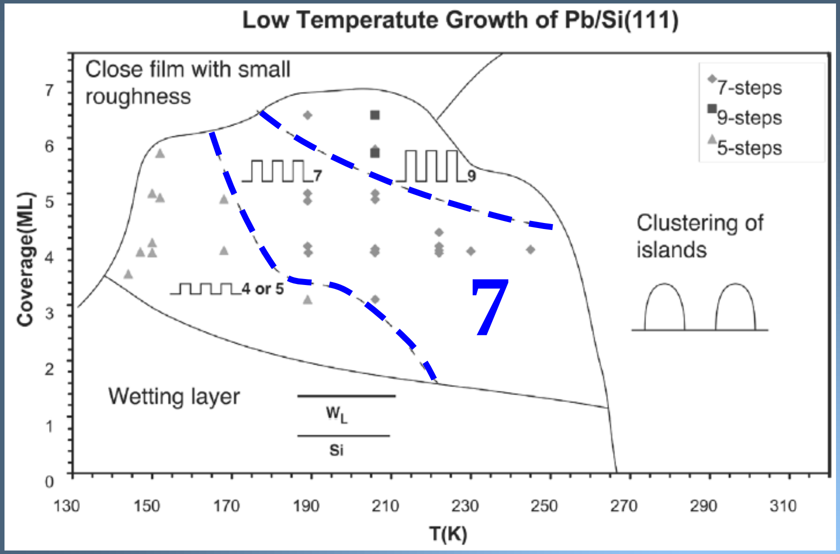




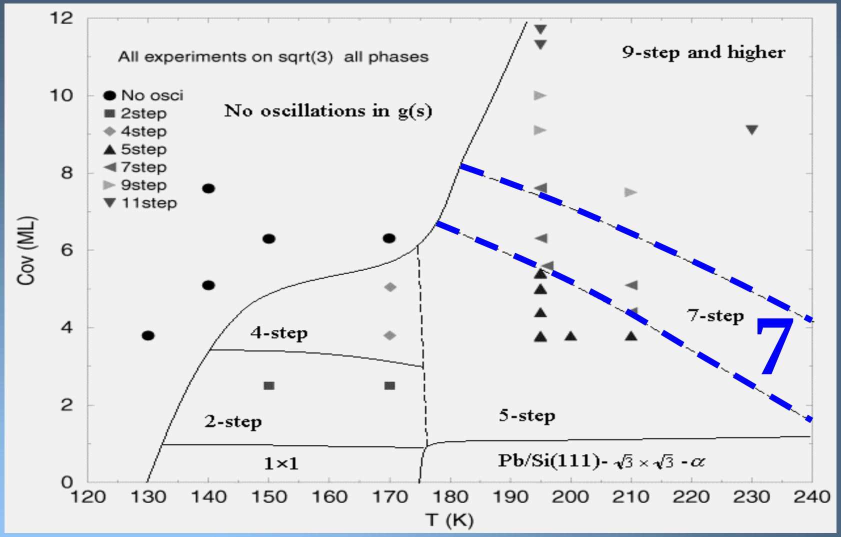
Si(111)-7 × 7 **200 *nm* × 200 *nm*** **Pb/Si(111)-$\sqrt{3}\times\sqrt{3}$ -β**

θ = 3.3 ML ***T* = 200K** **θ = 3 ML**

V. Yeh, L. Berbil-Bautista, C.Z. Wang, K.M. Ho, and M.C. Tringides, PRL 85, 5158 (2000)



Si(111)-7×7



Pb/Si(111)-$\sqrt{3}\times\sqrt{3}$

- **Wetting layer: from 3 ML at 130K to 1 ML at RT**
- **4- to 9-step islands observed**
- **For θ < 6 ML and T between 190K and 200K, 7-step islands with mean size 90Å**
- **At T = 195K, islands tend to stay at 7-step and cover the whole surface at θ ~ 8ML, then new islands grow on top**

- **First layer used to cover substrate to dense phase**
- **2-step to 11-step islands observed**
- **For θ < 6 ML and T between 190K and 200K, 5-step islands with mean size 180Å**
- **At T = 195K, islands tend to grow higher, in bi-layer increments, than grow larger for up to 20ML**

M. Hupalo, S. Kremmer, V. Yeh, L. Berbil-Bautista, E. Abram, M.C. Tringides, Surf. Sci. 493 (2001) 526

$$E = E_0 - E_C$$

E_0 : the energy of the Pb film (after subtracting the bulk energy)

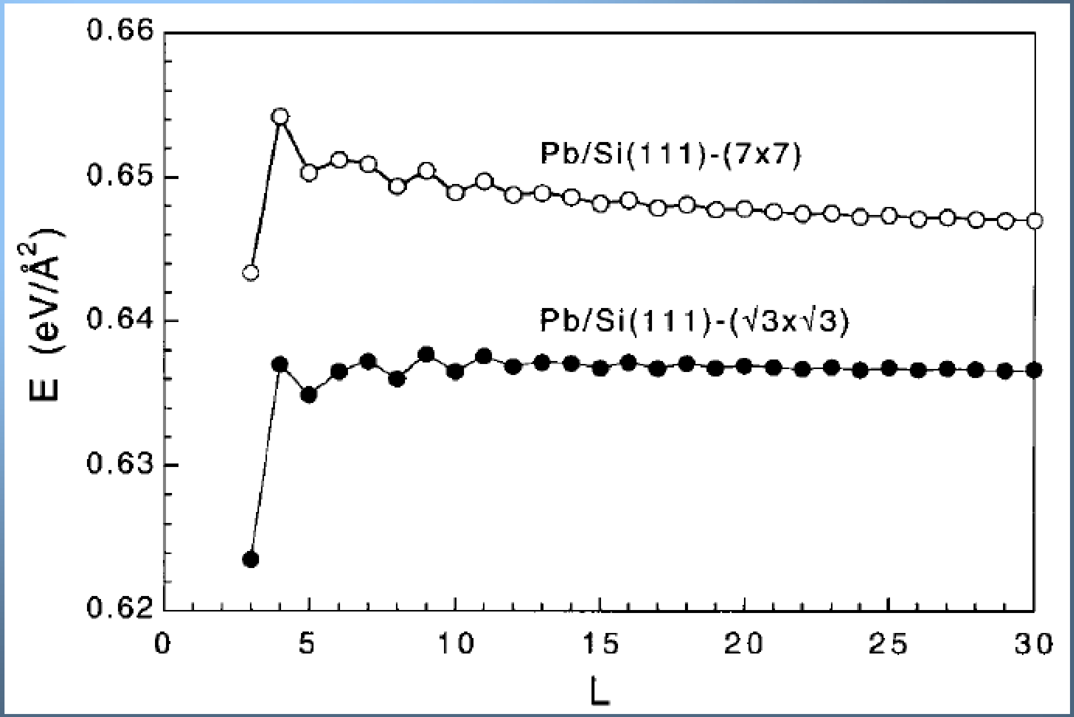
$E_C = C(\Delta E_f)^2$: energy gain due to the charge transfer

ΔE_f = 4.75 eV for 7×7 and 4.98 eV for $\sqrt{3} \times \sqrt{3}$

C = 0.033 eV/Å^2

(see Zenyu Zhang, et al, PRL 80, 5381
D.R. Heslina, et al, PRL 64, 1589 and H.H. Weitering, et al, PRB 45, 9126.)

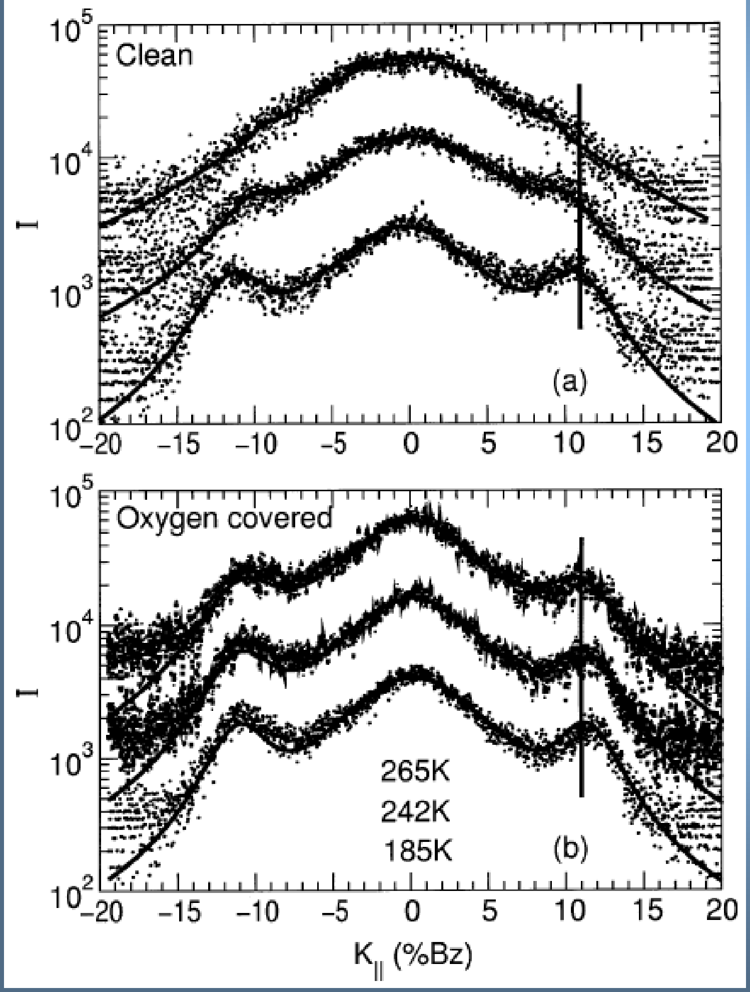
Calculated film energy vs film thickness *L* for the two interfaces showing the $\sqrt{3} \times \sqrt{3}$ phase has a lower curve and different minima due to larger charge transfer term.

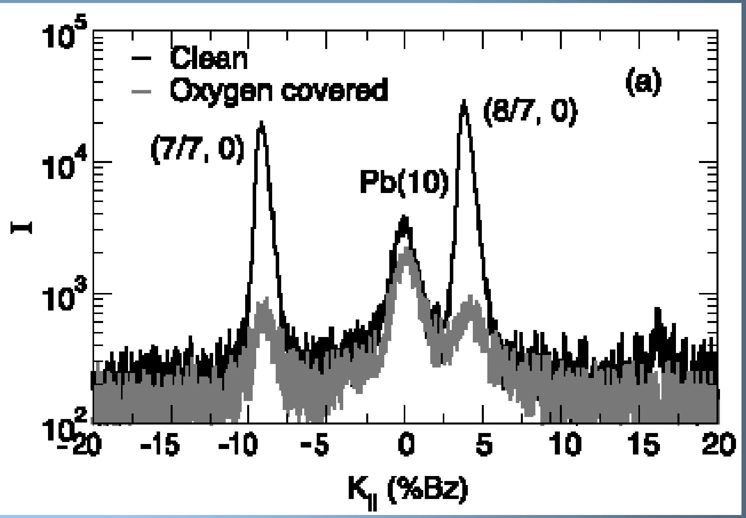


V. Yeh, L. Berbil-Bautista, C.Z. Wang, K.M. Ho, and M.C. Tringides, PRL 85, 5158 (2000)

Stability
of
QSE driven islands.

Oxygen covered Pb islands retain their height to a higher temperature





Oxygen suppressed atom diffusion to the islands

S. Stepanovskyy, V. Yeh, M. Hupalo, and M.C. Tringides, Surf. Sci. 515 (2002) 187

Controlling the Thermal Stability of Thin Films by Interfacial Engineering

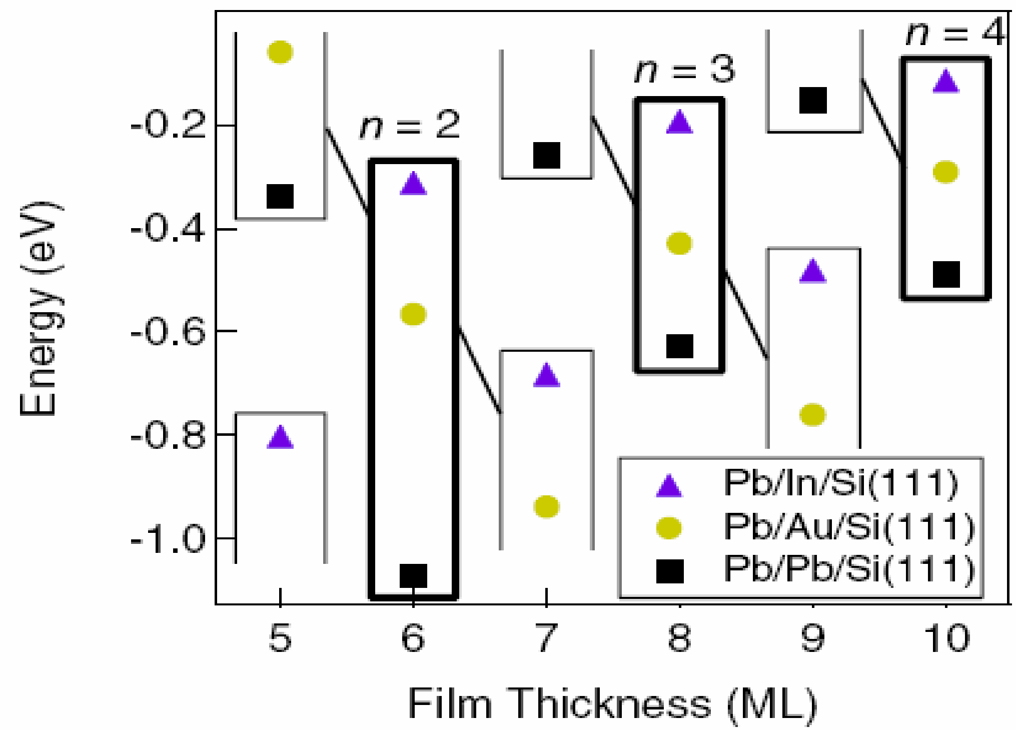


FIG. 1 (color online). Measured quantum well state energy levels for Pb films grown on the In-, Au-, and Pb-terminated Si(111) surfaces as a function of film thickness N. States are grouped by quantum number n using boxes for each thickness.

D.A. Ricci, T. Miller, and T.-C. Chiang, PRL 95, 266101 (2005)

Applicability of nucleation theory.

For 2D islands and complete saturation

$$n_x = \eta(\theta, i)\left(\frac{D}{F}\right)^{-\chi} \exp\left(\frac{E_i}{(i+2)kT}\right) \qquad \chi = \frac{i}{i+2}$$

$$\ln(n_x) = \ln\eta - \chi \ln\left(\frac{\nu_0}{4F}\right) + \frac{\chi}{kT}\left(E_m + \overline{E}_i\right)$$

$$\ln(n_x) = \chi \ln F + const2$$

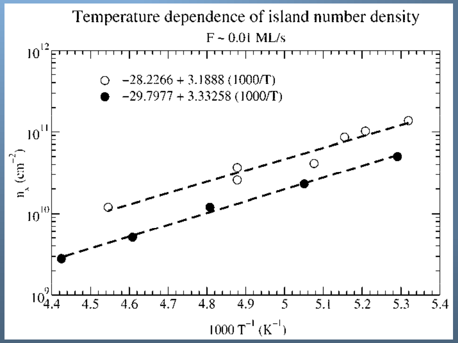
Rate dependence

$$\ln(n_x) = \frac{\chi}{kT}\left(E_m + \overline{E}_i\right) + const.$$

$$s = \frac{\chi}{k}\left(E_m + \overline{\overline{E}}_i\right)$$

$$\ln(n_x)\big|_{1/T=0} = \ln\eta - \chi \ln\left(\frac{\nu_0}{4F}\right)$$

Temperature dependence



STM measurements

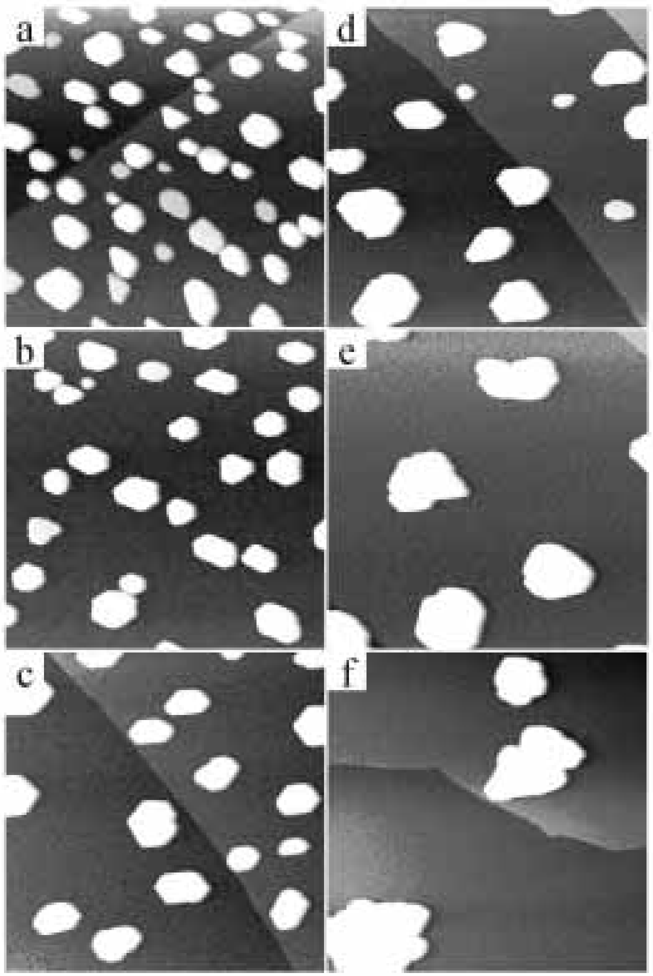


FIG. 5. The growth of Pb islands at (a) 189 K, (b) 198 K, (c) 208 K, (d) 217 K, (e) 226 K, and (f) 254 K at a coverage of 3.2 ML. The image size of (a)–(c) is 300× 300 nm², (d) and (e) is 500 ×500 nm², (f) is 1000×1000 nm².

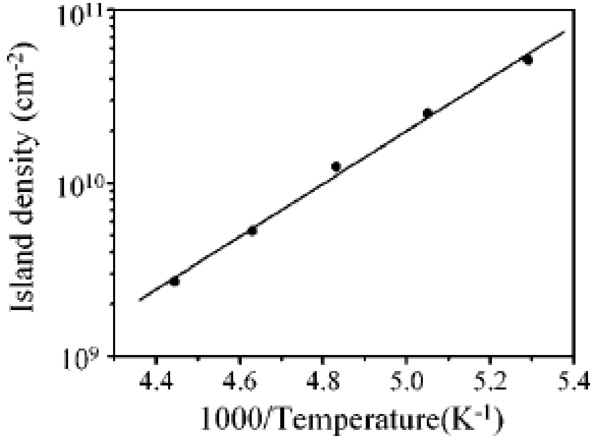


FIG. 6. The Arrhenius plot of the island density versus temperature, showing a linear relationship.

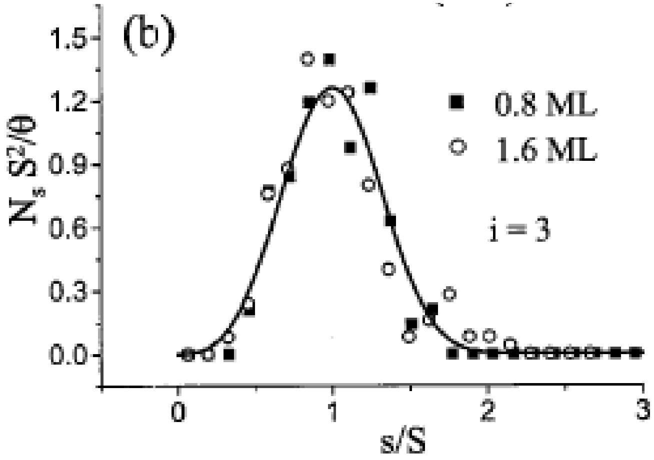


FIG. 4. (a) Pb island size distributions at the coverage of 0.8 and 1.6 ML above the wetting layer. The solid and dash-dotted lines are smooth fitting lines to the data points. (b) The two size distributions in (a) can be scaled into nearly identical distributions. The solid curve is the theoretical scaling function of three-atom critical size.

S.H. Chang, W.B. Su, W.B. Jian, C.S. Chang, L.J. Chen, and T.T. Tsong, PRB 65, 245401 (2002)

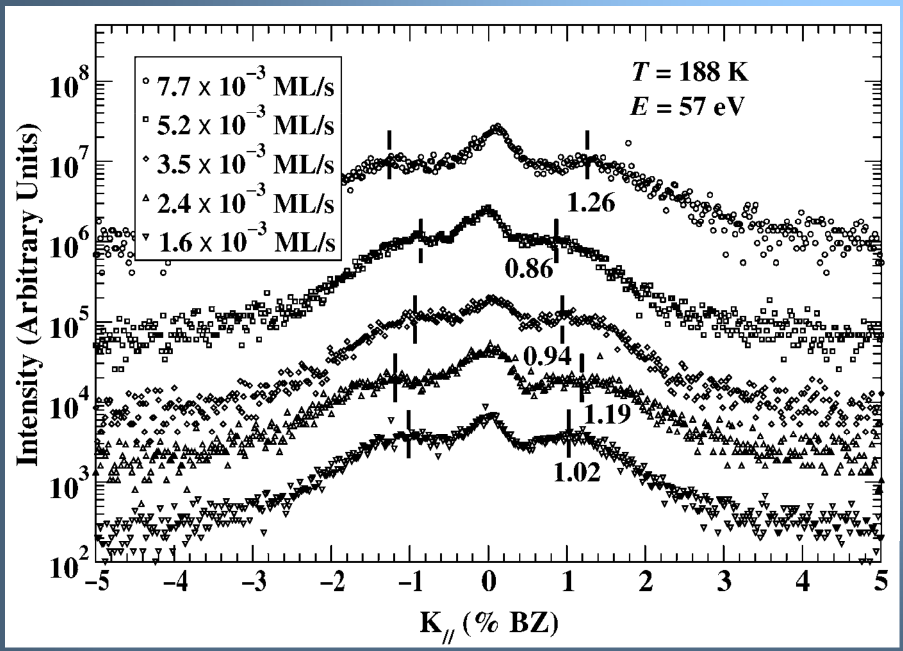
W.B. Su, S.H. Chang, H.Y. Lin, Y.P. Chiu, T.Y. Fu, C.S. Chang, and T.T. Tsong, PRB 68, 033405 (2003)

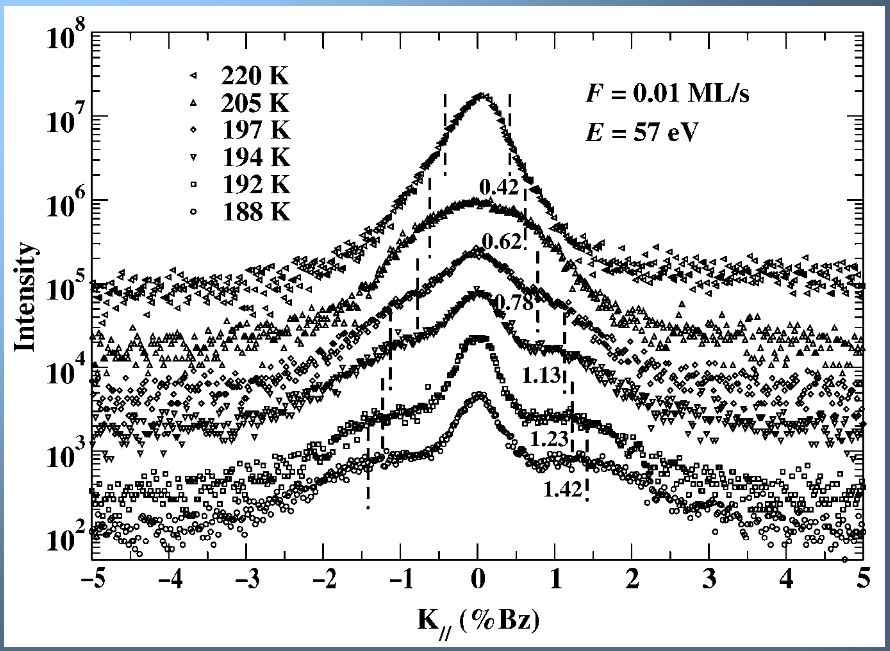
SPA-LEED measurements

$$n_x \approx \left(\frac{1}{S}\right)^2 = \left(\frac{K_1}{2\pi}\right)^2 = \left(\frac{K_1}{K_{10}}\right)^2 \left(\frac{1}{a_0}\right)^2$$

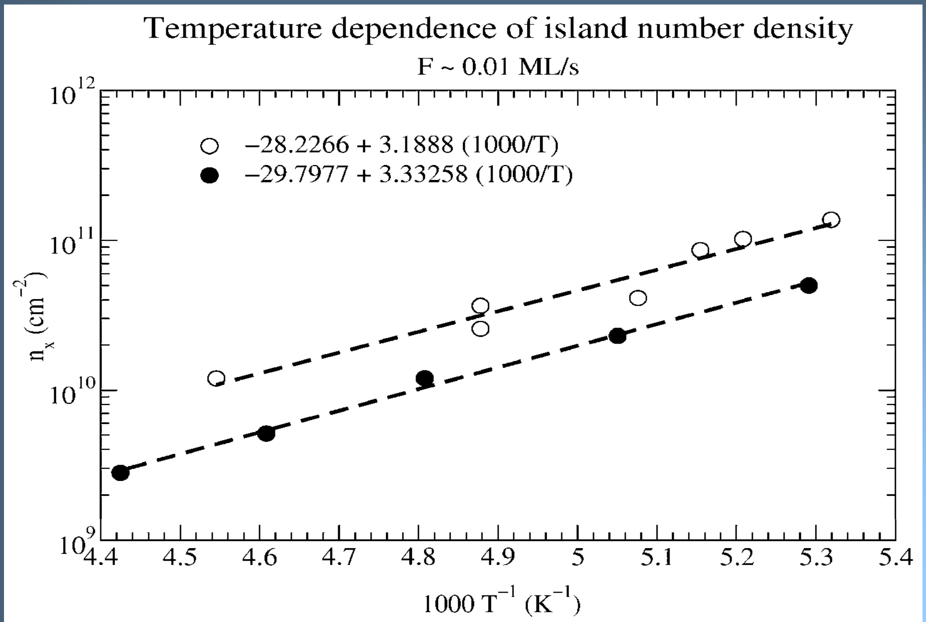
Assumption

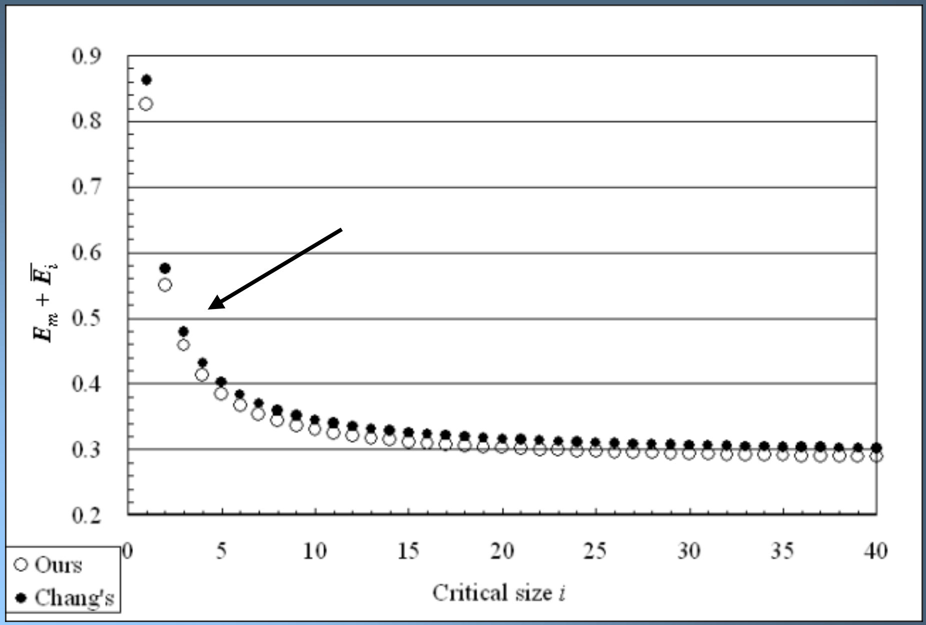
Measured island density = saturation density

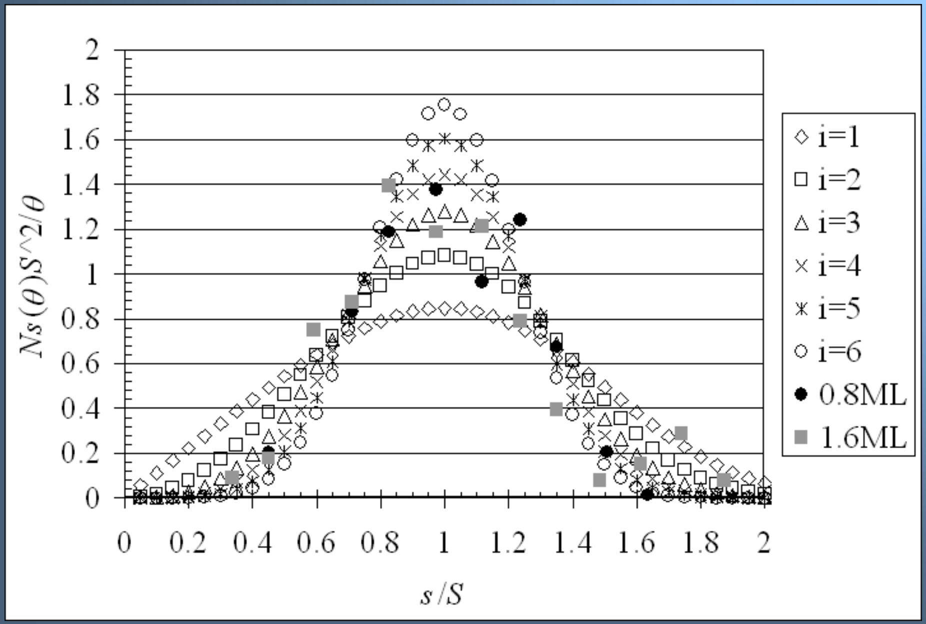


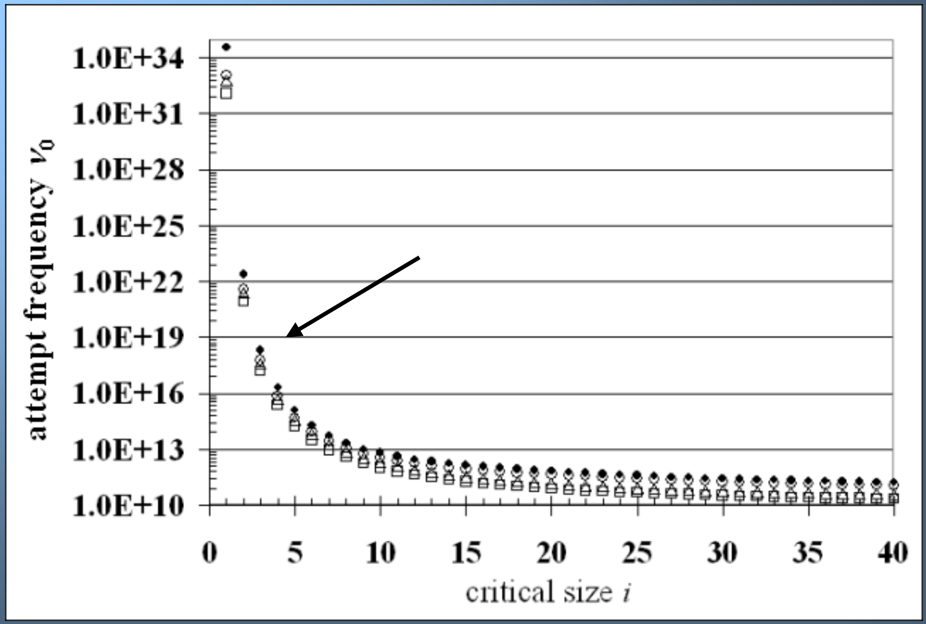


Rate dependence **Temperature dependence**


Temperature dependence of island number density
F ~ 0.01 ML/s
−28.2266 + 3.1888 (1000/T)
−29.7977 + 3.33258 (1000/T)
n_x (cm^-2)
1000 T^-1 (K^-1)


E_m + Ē_i
Critical size i
Ours
Chang's


Ns(θ)S^2/θ
s/S
i=1
i=2
i=3
i=4
i=5
i=6
0.8ML
1.6ML


attempt frequency ν0
critical size i

Low θ Structures

$\sqrt{3}\times\sqrt{3}-\beta$

$\sqrt{7}\times\sqrt{3}$

$\sqrt{43}\times\sqrt{3}$

$\sqrt{3}\times\sqrt{3}-\text{HIC}$

$\sqrt{3}\times\sqrt{3}-\text{SIC}$

Anything else?

Low coverage structures

Inconsistent results between different works

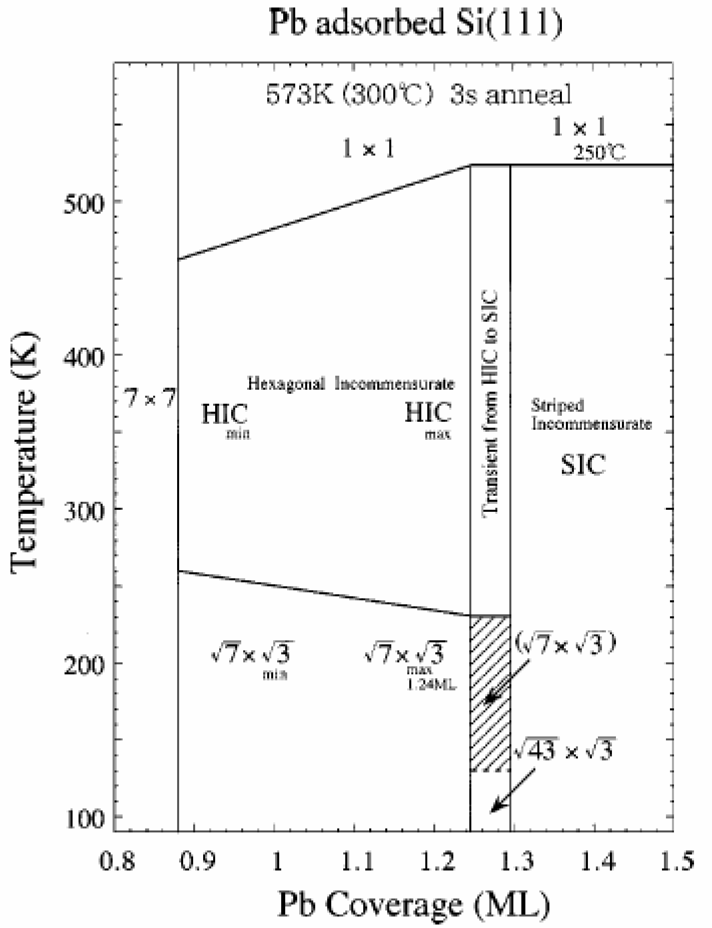


FIG. 1. A structure map, showing the surface phases that appear by Pb deposition.

Kotaro Horikoshi, Xiao Tong, Tadaaki Nagao, Shuiji Hasegawa, PRB 60, 13287 (1999)

STM study on HIC phase

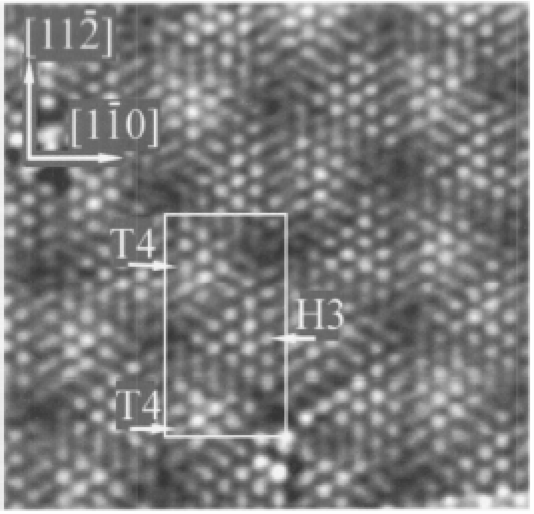


FIG. 1. 17.5 nm×15 nm STM image of the HIC phase showing the sixfold domain degeneracy, with alternating triangular domains occupying different binding sites H3 vs T4.

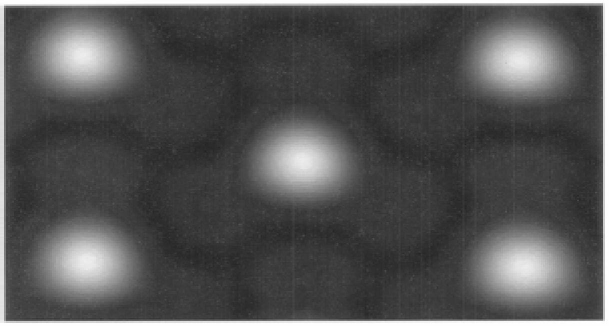


FIG. 3. First-principles calculation of the STM image of the α-$\sqrt{3}\times\sqrt{3}$ for H3 site occupation with $V=+1.5$ V.

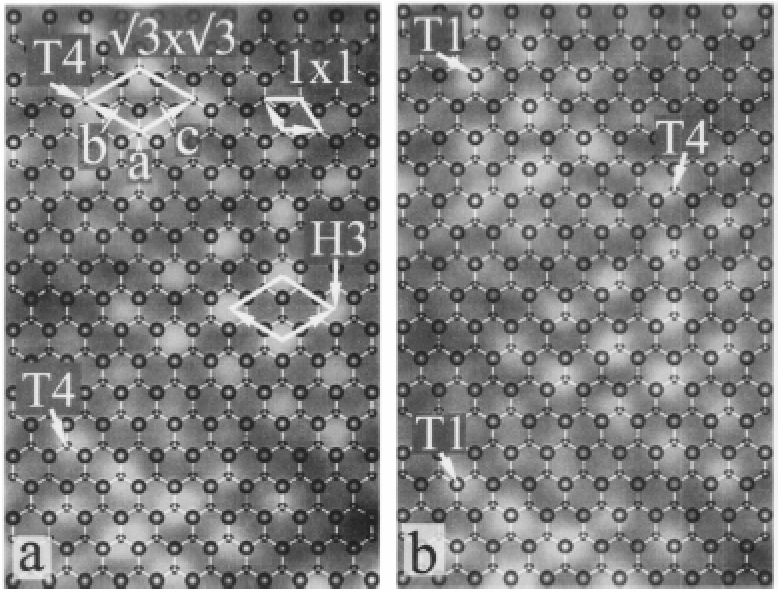


FIG. 2. A Si(1×1) lattice overlayed on top of the area in Fig. 1 enclosed by the white rectangle, can be used to deduce the type of site occupied in a given triangular domain, i.e., whether H3 or T4. Figure 2(a) is the correct assignment.

M. Hupalo, T.L. Chan, C.Z. Wang, K.M. Ho and M.C. Tringides, PRB 66 161410 (2002)

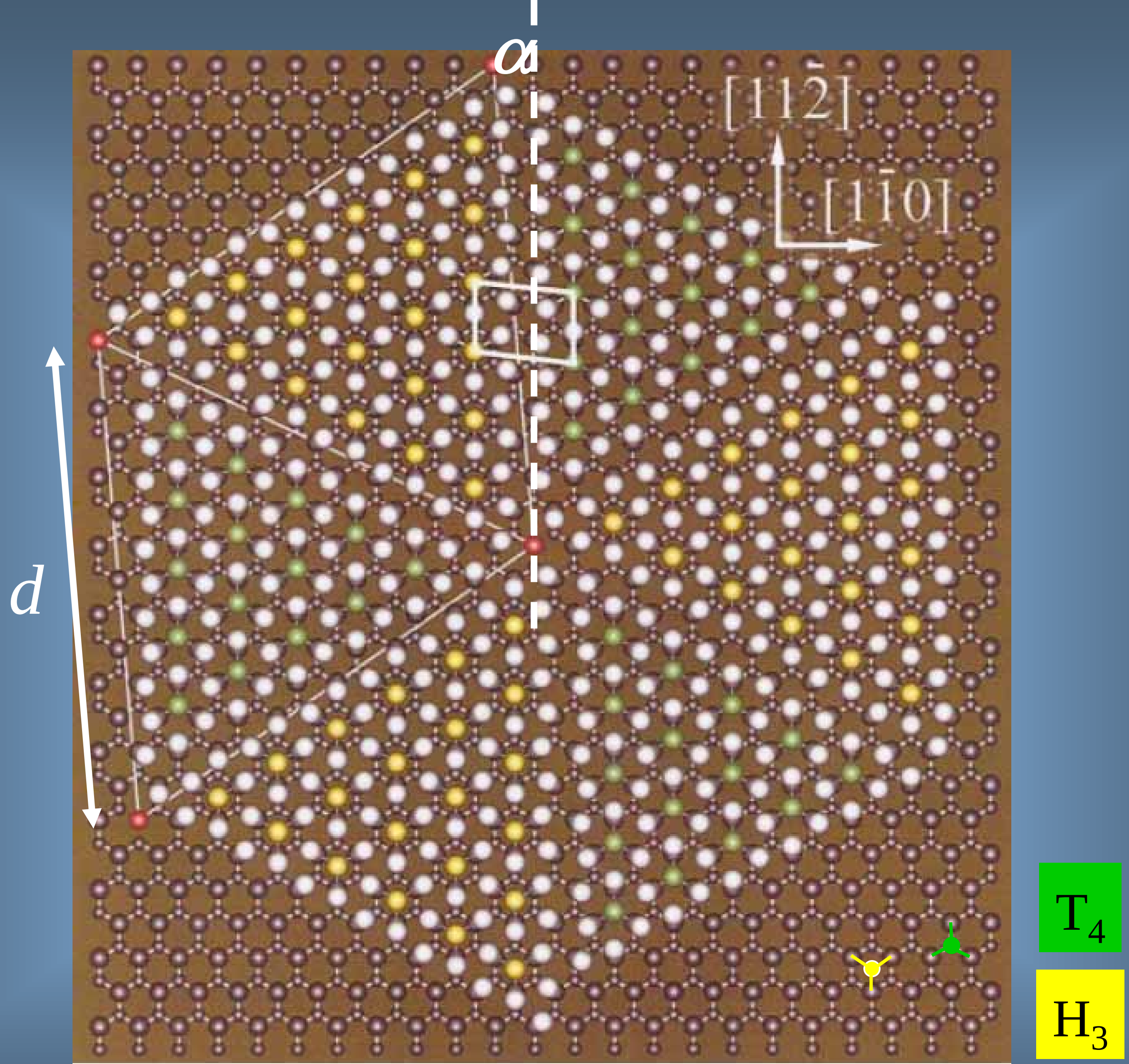
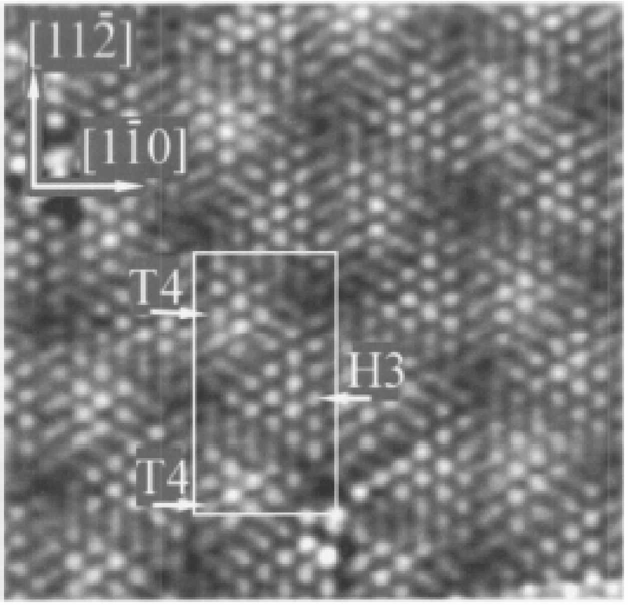


FIG. 4. (Color) Atomic model of the HIC phase shown in Fig. 1.

M. Hupalo, T.L. Chan, C.Z. Wang, K.M. Ho and M.C. Tringides, PRB 66 161410 (2002)

LEED study on HIC phase



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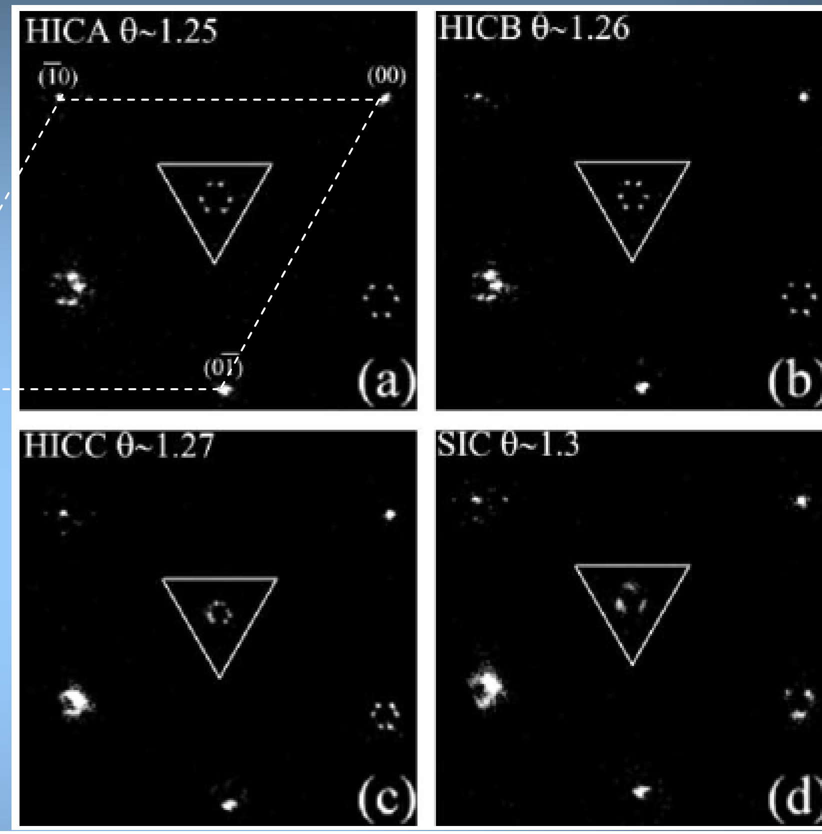
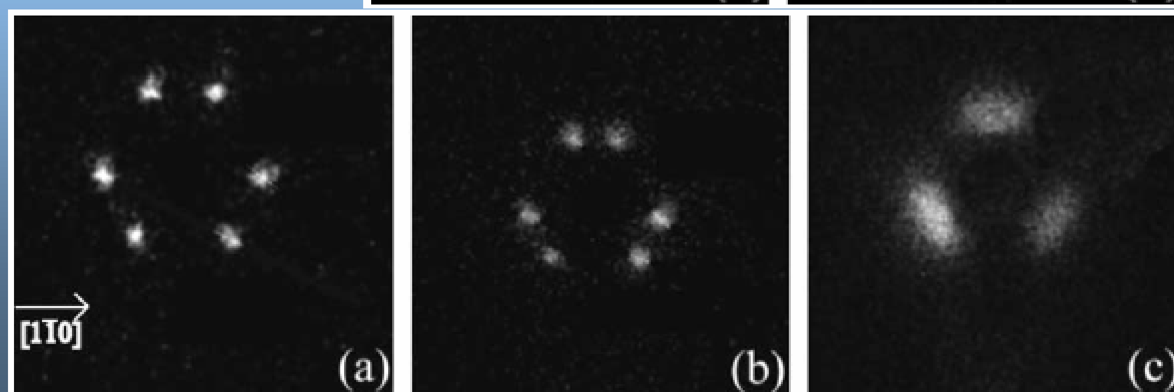
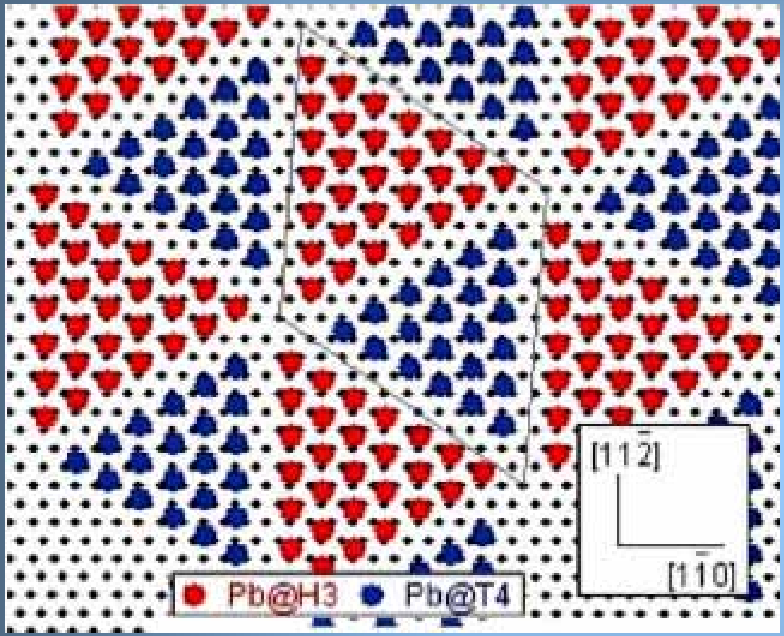
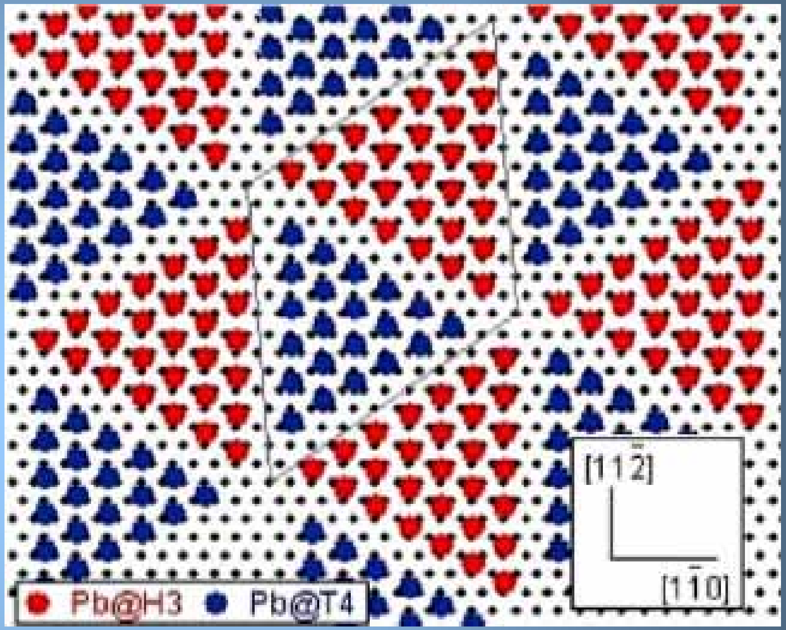


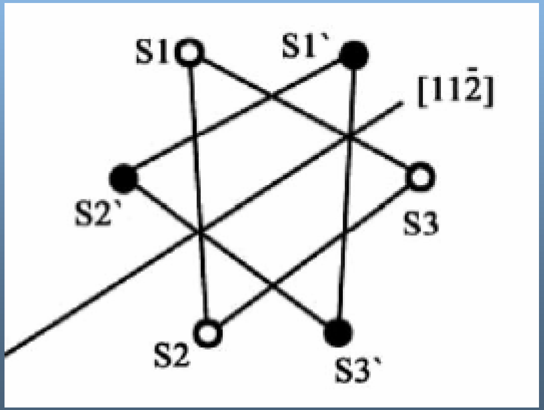


S. Stepanovskyy, M. Yakes, V. Yeh, M. Hupalo and M.C. Tringides, Surface Science 600, 1417 (2006)

Kinematical calculations of LEED pattern for HIC phase



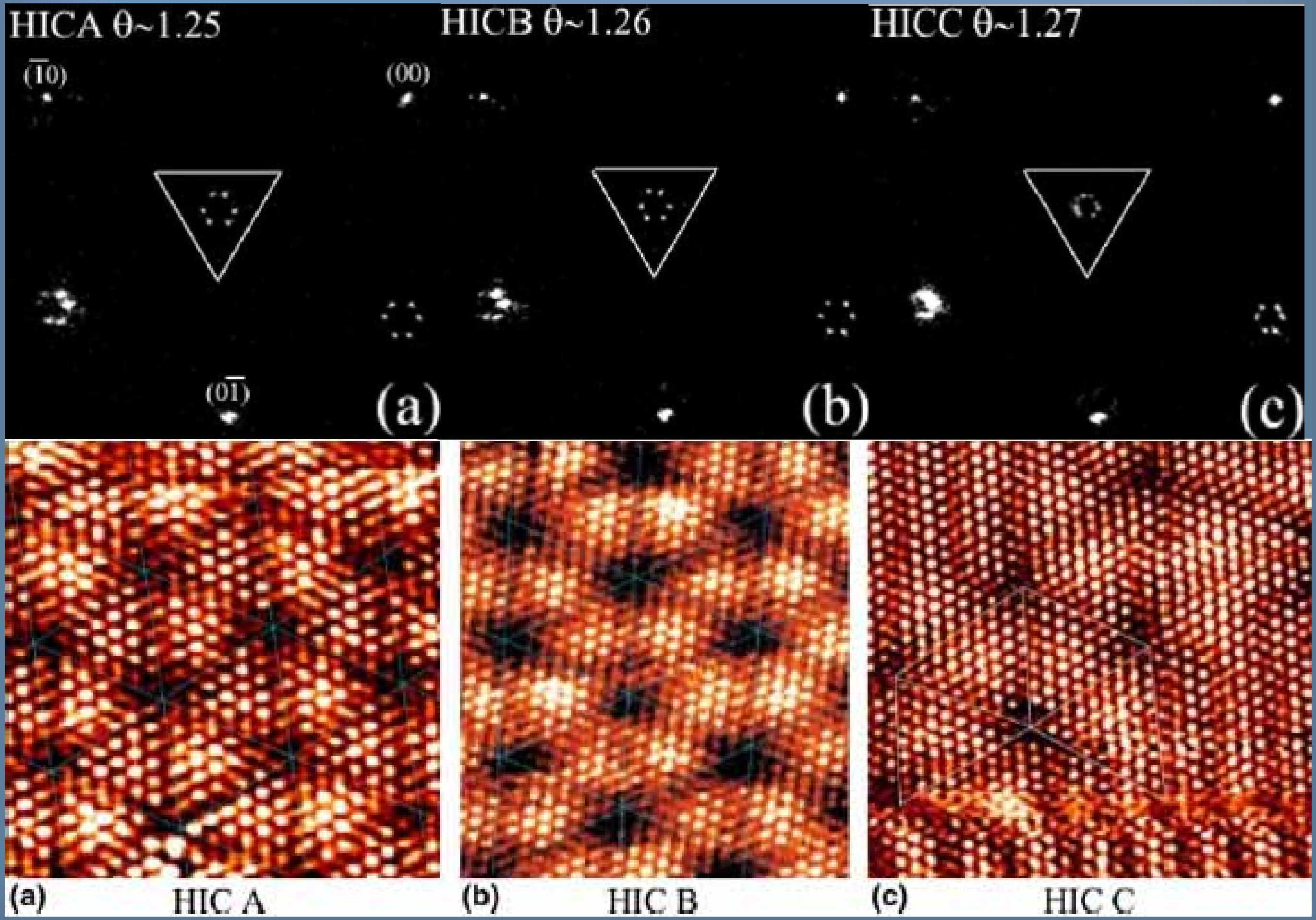




S. Stepanovskyy, M. Yakes, V. Yeh, M. Hupalo and M.C. Tringides, Surface Science 600, 1417 (2006)

Three HIC phases

$d \sim 48$Å $\alpha \sim 5.4^\circ$ | $d \sim 53$Å $\alpha \sim 4.3^\circ$ | $d \sim 61$Å $\alpha \sim 3.0^\circ$



$d \sim 47$Å $\alpha \sim 4.7^\circ$ | $d \sim 53$Å $\alpha \sim 4.1^\circ$ | $d \sim 59$Å $\alpha \sim 3.7^\circ$

$\sqrt{43} \times \sqrt{3}$?

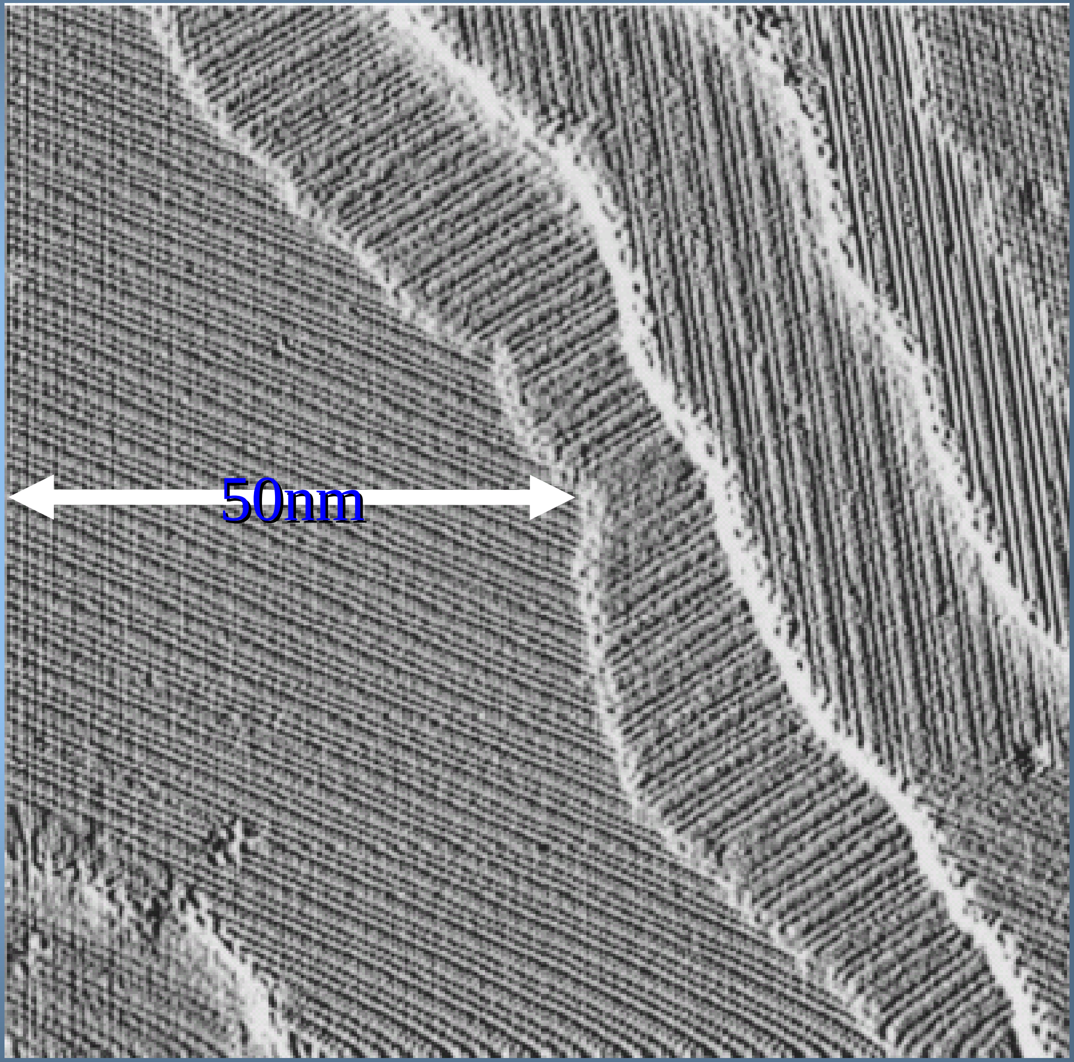
In Pb/Si(111)

$\Delta\theta = 0.006ML$

$T_{dep} = 120K$

Huge number of atoms move collectively

Two phases covers 80% of 0.5mm



1.5V, 100nm × 100nm

In a tiny change of θ

$\sqrt{7}\times\sqrt{3}$ **θ = 1.2 ML**

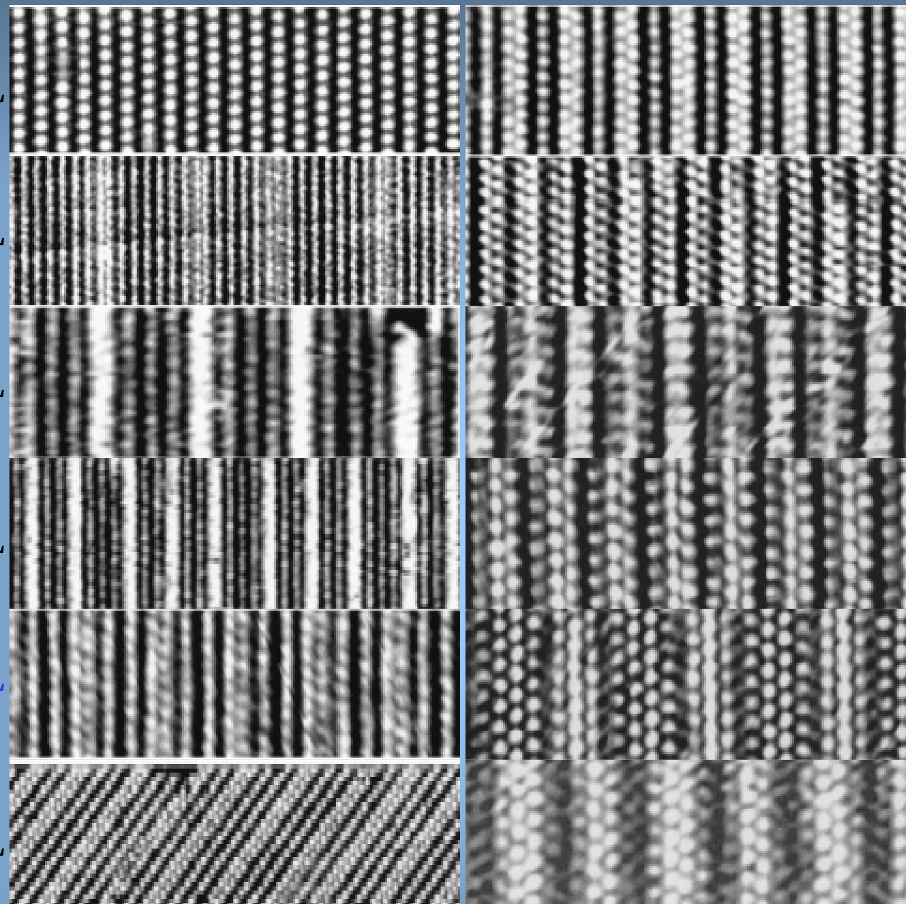
(7, 1) and (5, 1), θ = 1.21 ML

(4, 1), θ = 1.217 ML

(4, 1) and (3, 1), θ = 1.220 ML

(3, 1), θ = 1.222 ML

(3, 1) and (2, 1), θ = 1.223 ML



$\sqrt{43}\times\sqrt{3}$

(2, 1), θ = 1.231 ML

(1, 1), θ = 1.25 ML

(1, 1) and (1, 2), θ = 1.263 ML

(1, 2), θ = 1.27 ML

(1, 2) and (1, 3), θ = 1.28 ML

(1, 3), θ = 1.285 ML

M. Hupalo, J. Schmalian, M.C. Tringides, PRL 90, 216106-1 (2003)

STM images of twelve distinct, commensurate phases observed within a small coverage range 1.2 ML < θ < 1.3 ML.

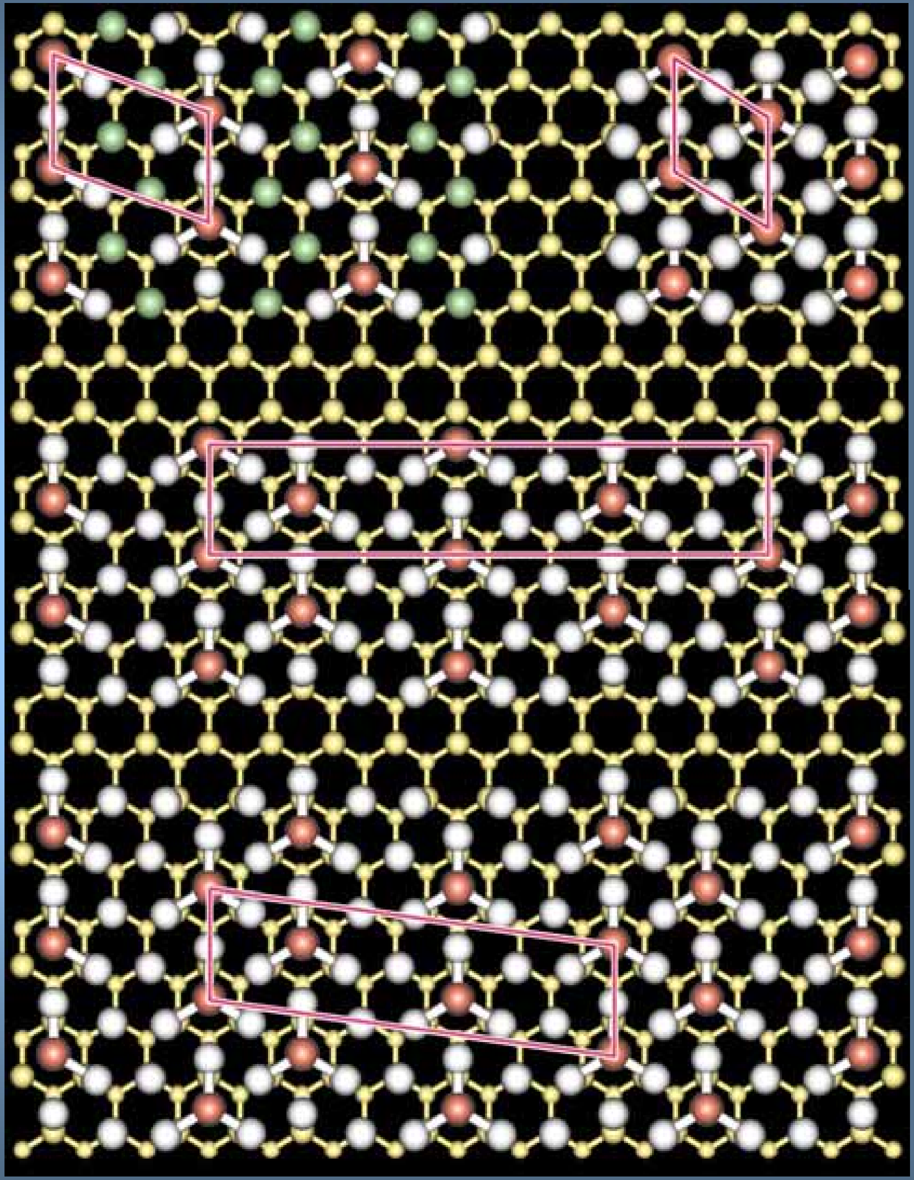
Detailed structures of

(3, 1)

and

(2, 1)

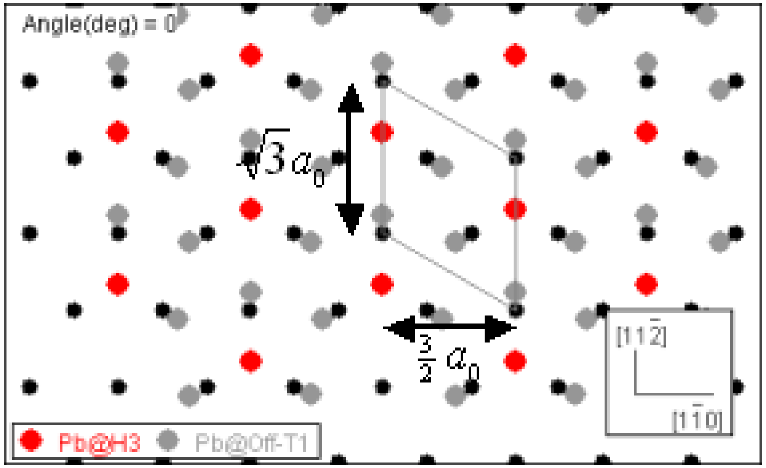
phases



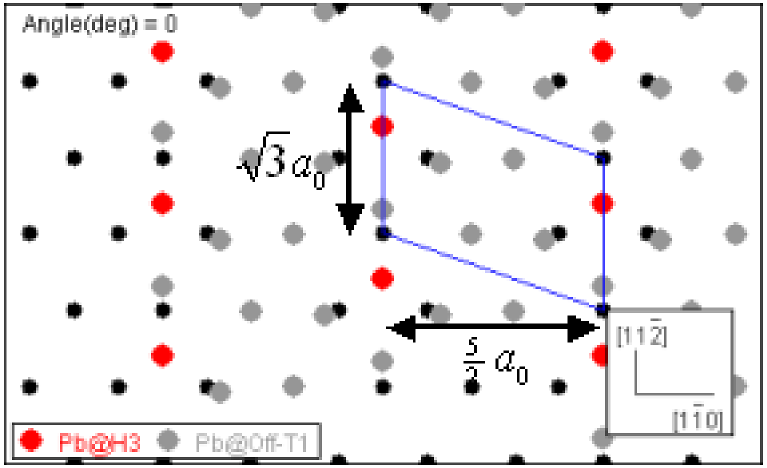
What are the detailed structures of those separations?

Smaller separations
Θ = 4/3 ML

Larger separations
Θ = 6/5 ML



$\sqrt{3} \times \sqrt{3}$



$\sqrt{7} \times \sqrt{3}$

C.Kumpf, etc., Scurf. Sci. **448**, L213 (2000)
T.L. Chan, etc., Phys. Rev. B **68**, 45410 (2003)

$a_0 = 3.84$Å

Periodicity and coverage of a (n, m) phase

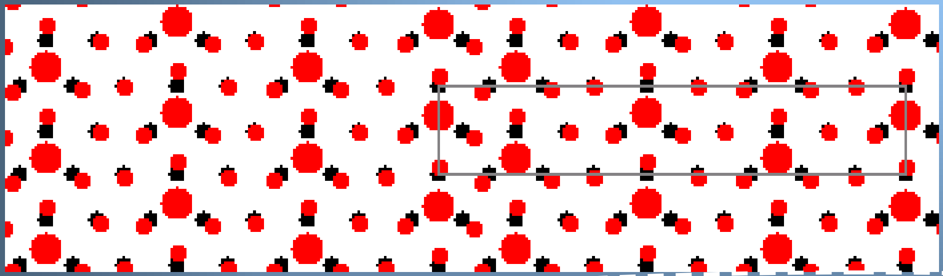
Periodicity

$$q_{(n,m)} = (5\times n + 3\times m)\frac{a_0}{2}$$

Coverage

$$\theta_{(n,m)} = \frac{6n+4m}{5n+3m} = 1 + \frac{n+m}{5n+3m} = 1 + \frac{p}{q}$$

For example, $(n, m) = (3, 1)$

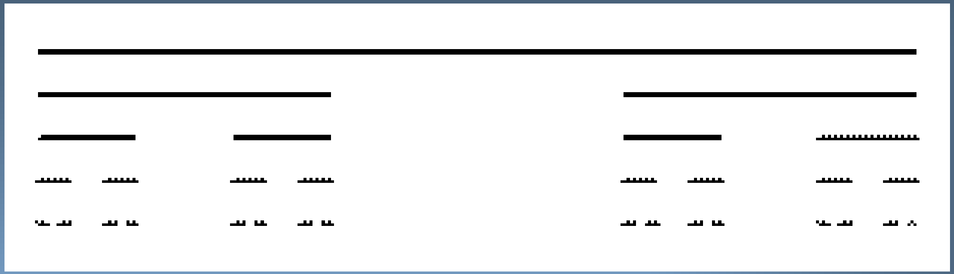


$$q_{(3,1)} = (5\times 3 + 3\times 1 = 18)\frac{a_0}{2}$$

$$\theta_{(3,1)} = 1 + \frac{(3+1)}{q} = 1 + \frac{4}{18}$$

$$\theta_{(3,1)}\left(=\frac{22}{18}\right) \oplus \theta_{(2,1)}\left(=\frac{16}{13}\right) = \theta_{(5,2)=(3,1)\oplus(2,1)}\left(=\frac{38}{31}\right)$$

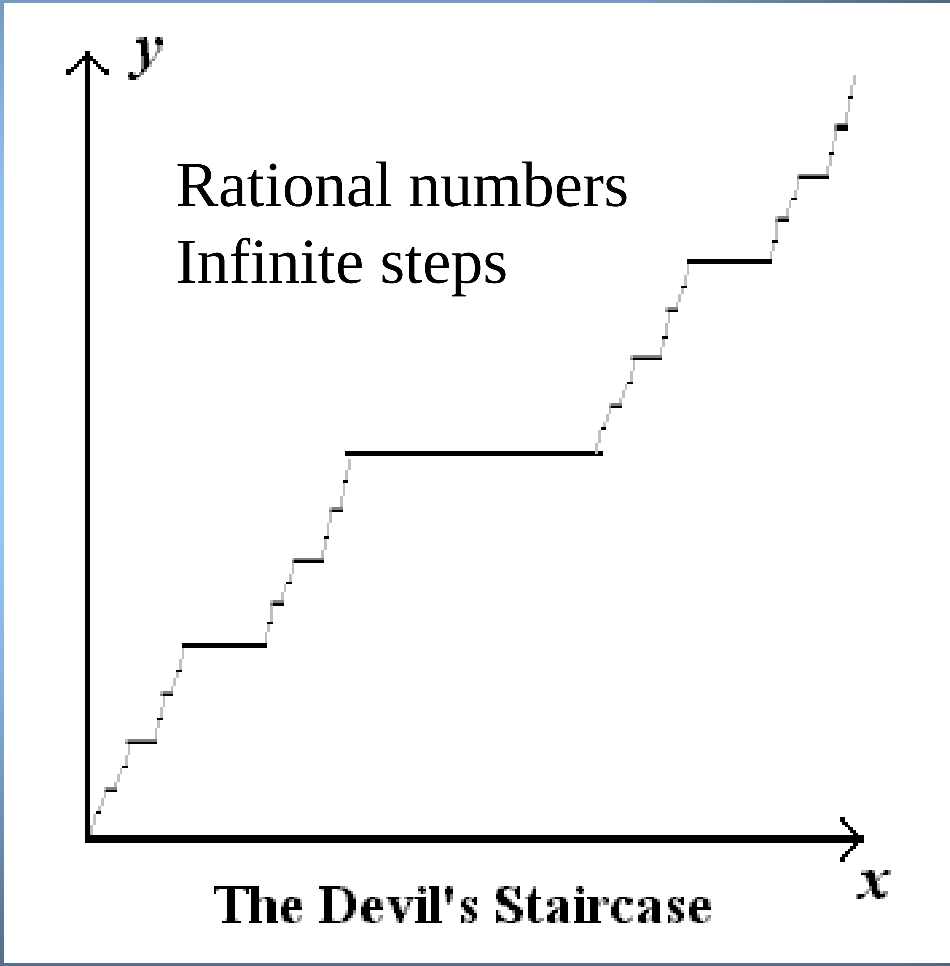
Cantor comb



Cantor function

Infinite steps in
a finite interval

All steps take
rational numbers



http://www.mathacademy.com/pr/prime/articles/cantset/

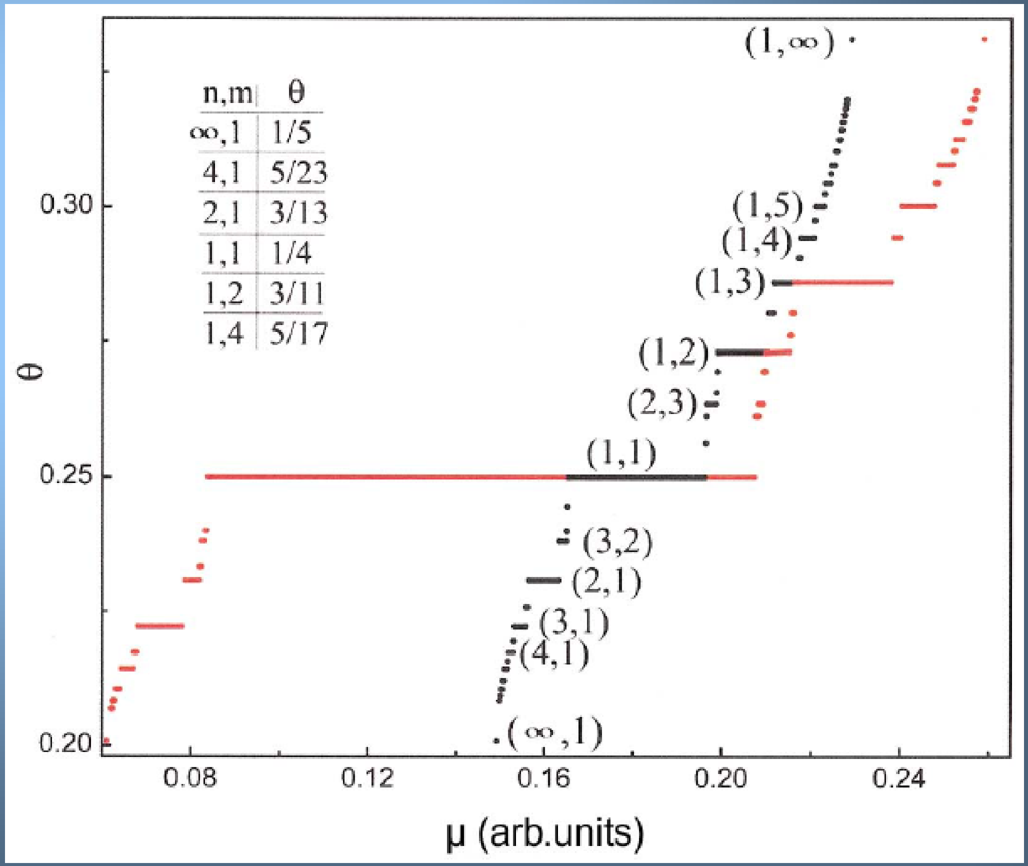
The devil's staircase in Pb/Si(111) reveals itself in θ vs μ stability curve

$$\Delta\mu(p/q) = \frac{1}{2}\sum_{l=1}^{\infty} lq\{J(lq+2) + J(lq-2) - 2J(lq)\}$$

Competing interactions
Chemical: immiscibility
Elastic: lattice mismatch

Steeper steps due to double separation

Additional 3-fold rotational symmetry gives rise to phases other than the DS phases at finite temperatures



M. Hupalo, J. Schmalian, M.C. Tringides, PRL 90, 216106-1 (2003)

Examples of physical systems that show a devil's staircase (I)

Competing interactions
Ferromagnetic for NN
Anti-ferromagnetic for NNN

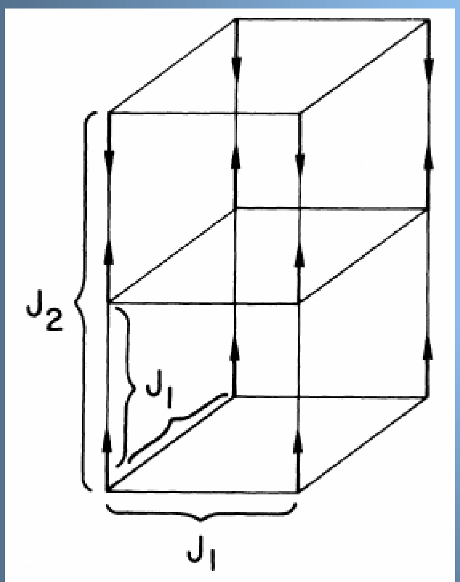
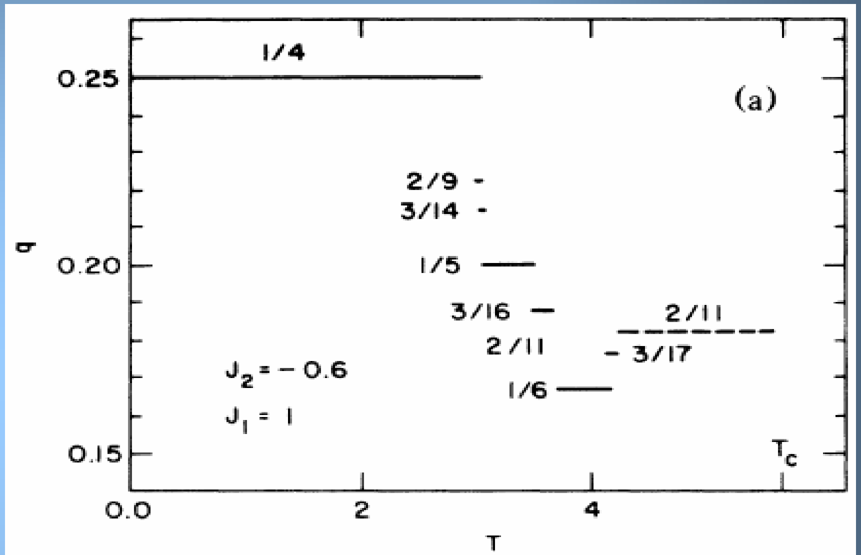
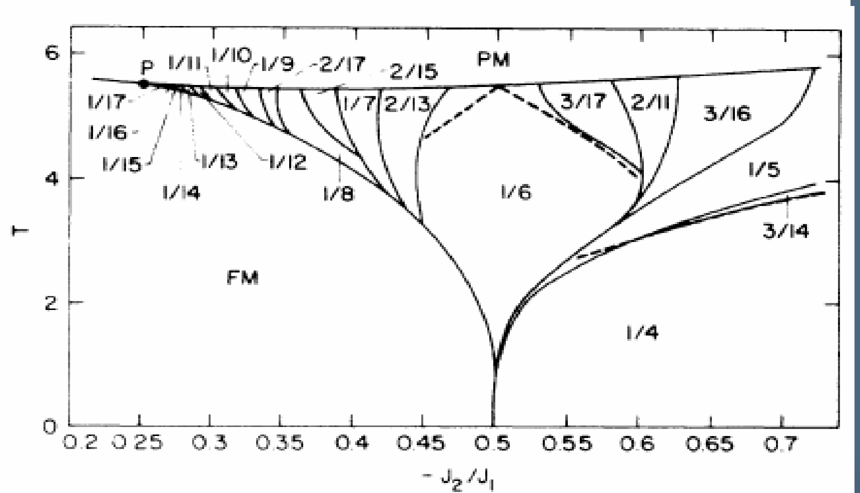


FIG. 1. Ising model with competing interactions.

Per Bak, J. von Boehm, PRB **21**, 5297 (1980)



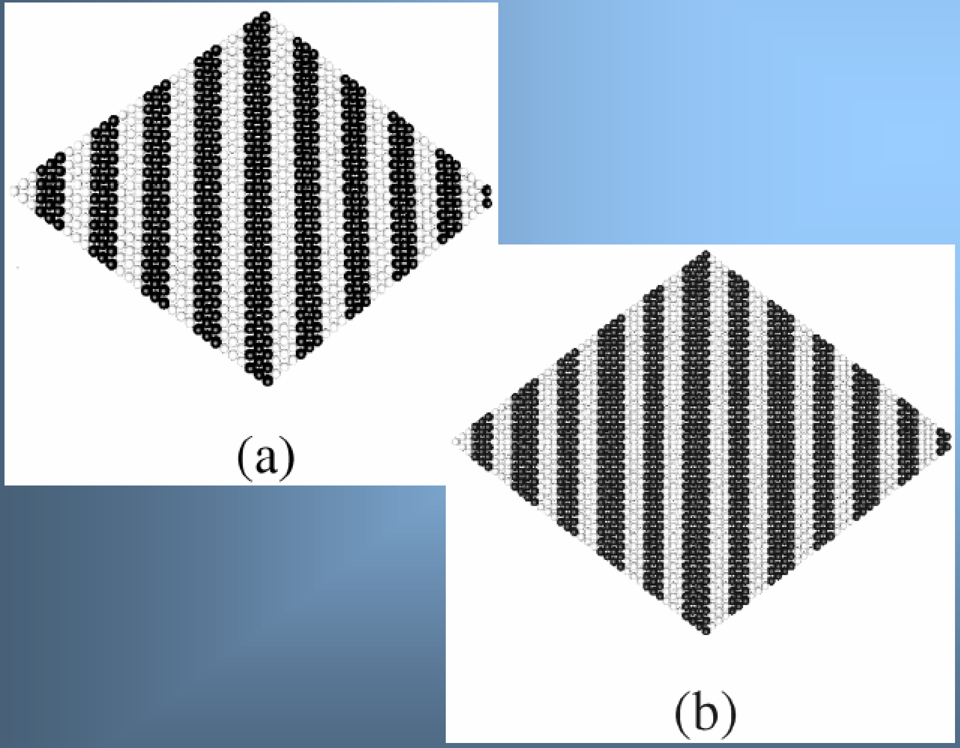


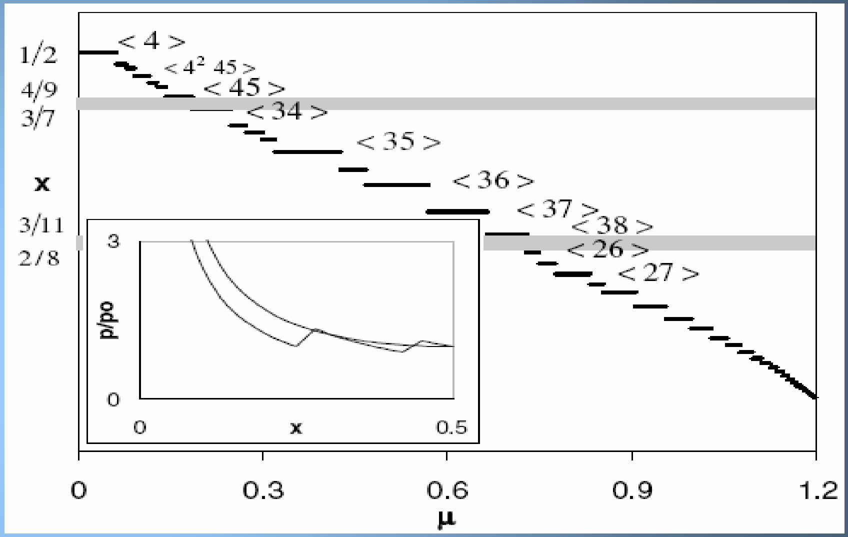
Examples of physical systems that show a devil's staircase (II)

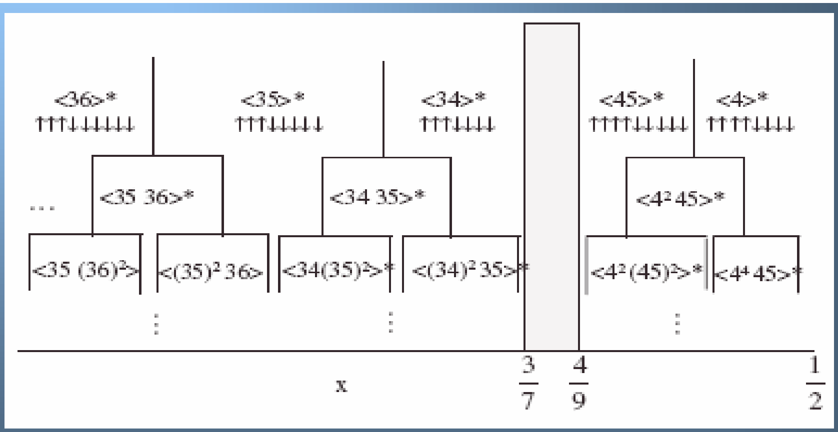
Competing interactions

Chemical: bulk immiscibility

Elastic: lattice mismatch







B.D. Krack, V. Ozolins, M. Asta, and I. Daruka, PRL **88**, 186101 (2002)

The T-θ phase diagram

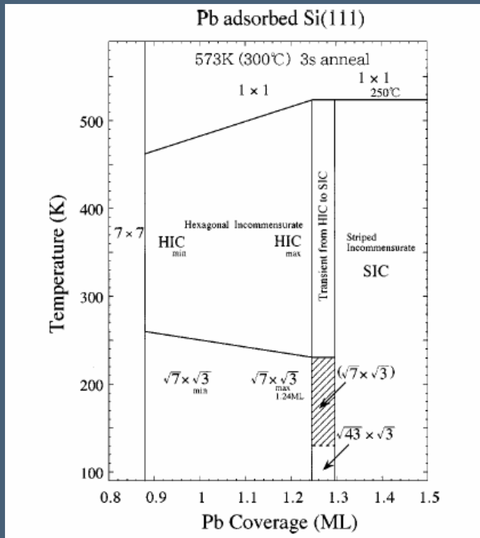
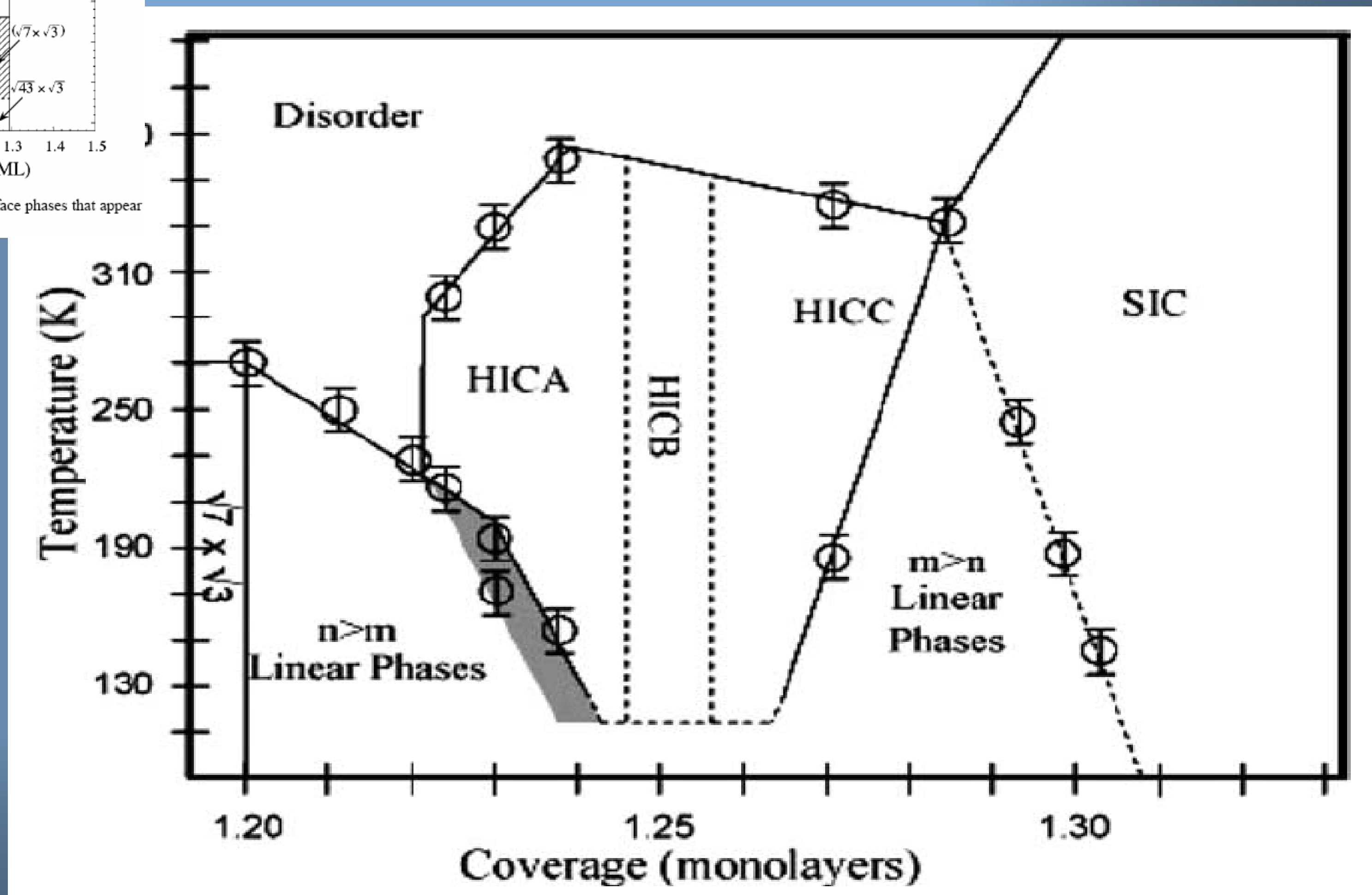


FIG. 1. A structure map, showing the surface phases that appear by Pb deposition.

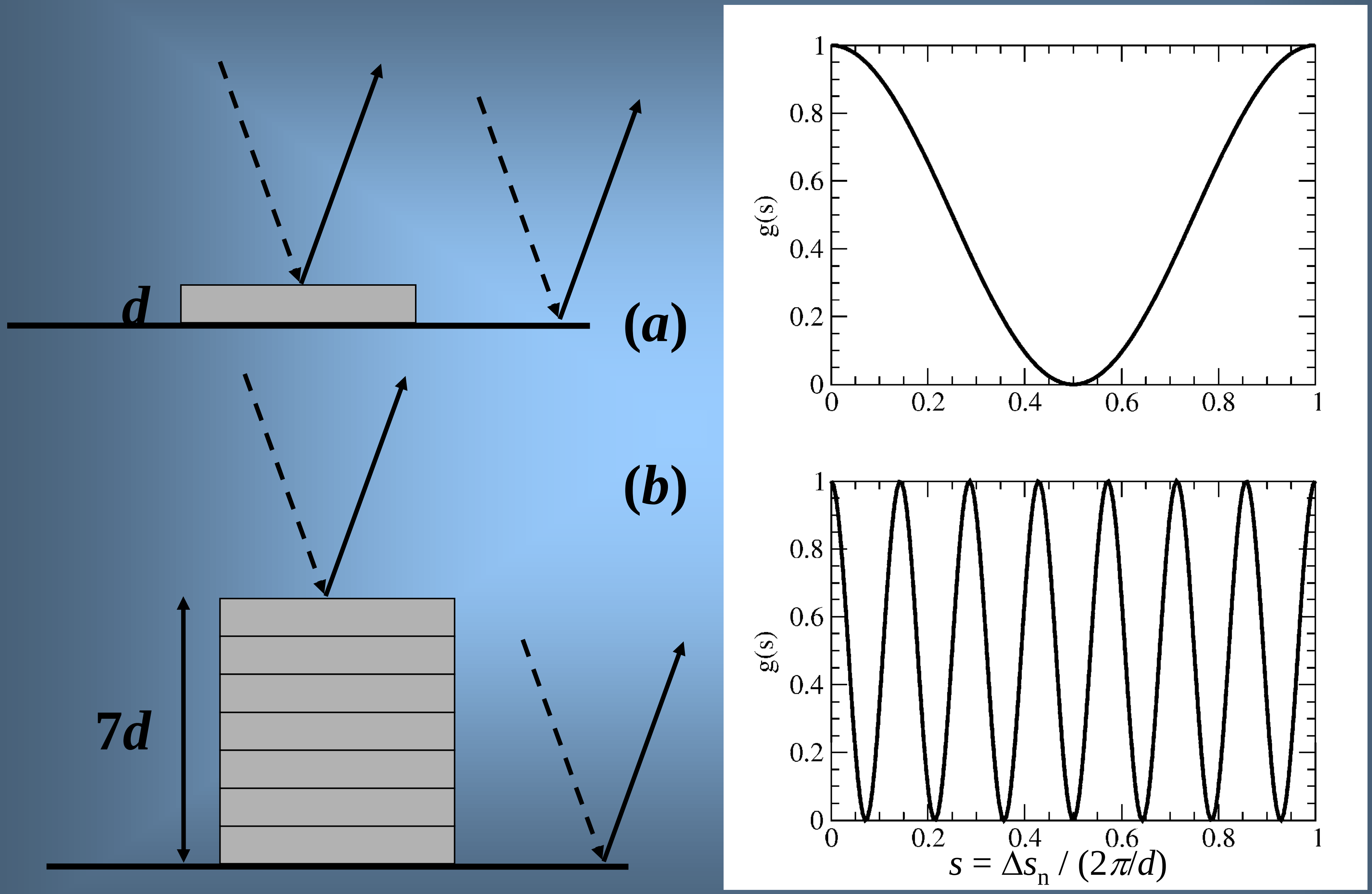


Thank you

for

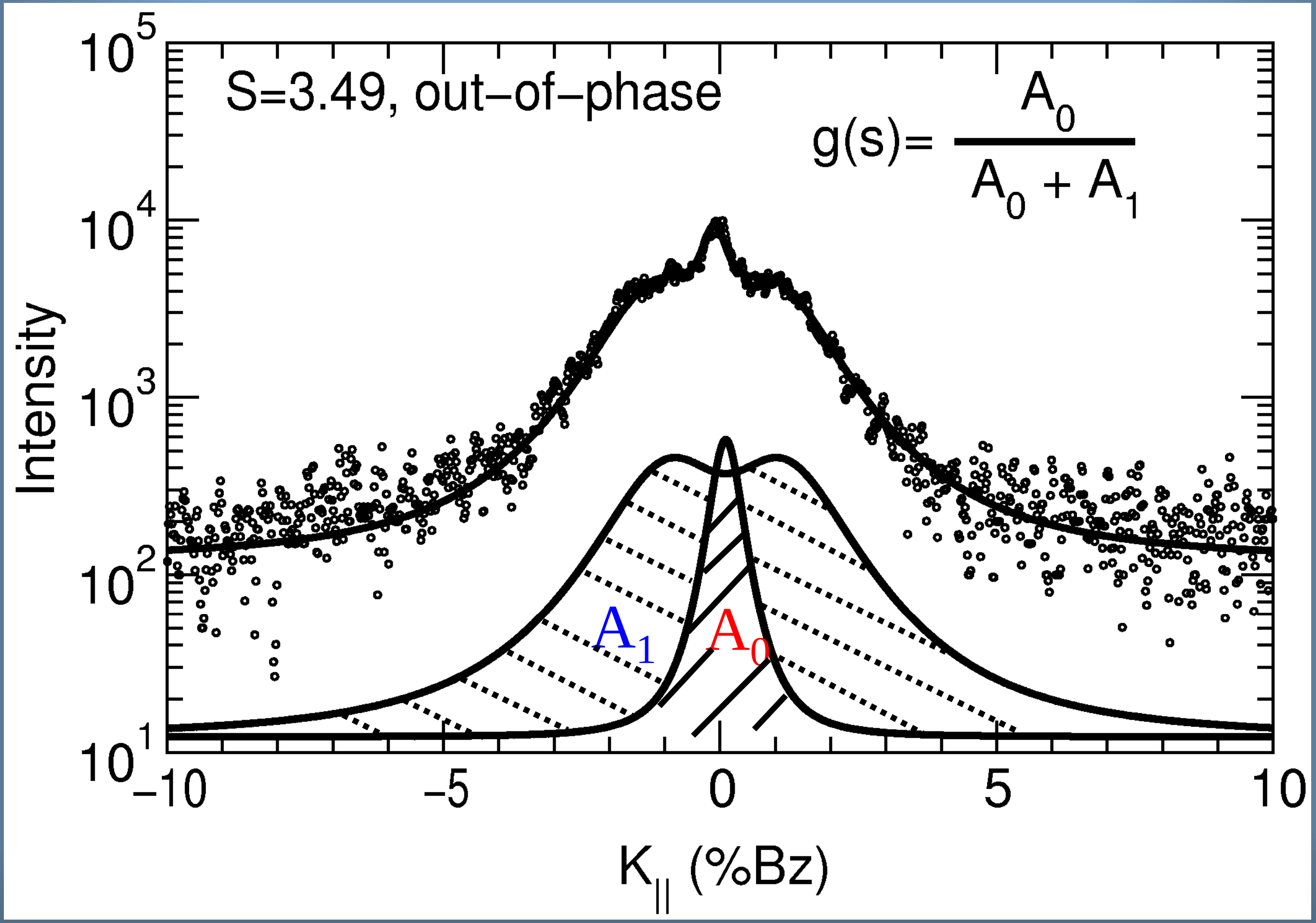
your attention.

g(s): Intensity of (00) spot vs electron beam energy



Δs_n: normal component of momentum of incident waves

g(s) definition



Binary phase diagram Pb vs Si