Coarsening of Pb Islands and Dynamics of Pb Wetting Layer on Si(111)

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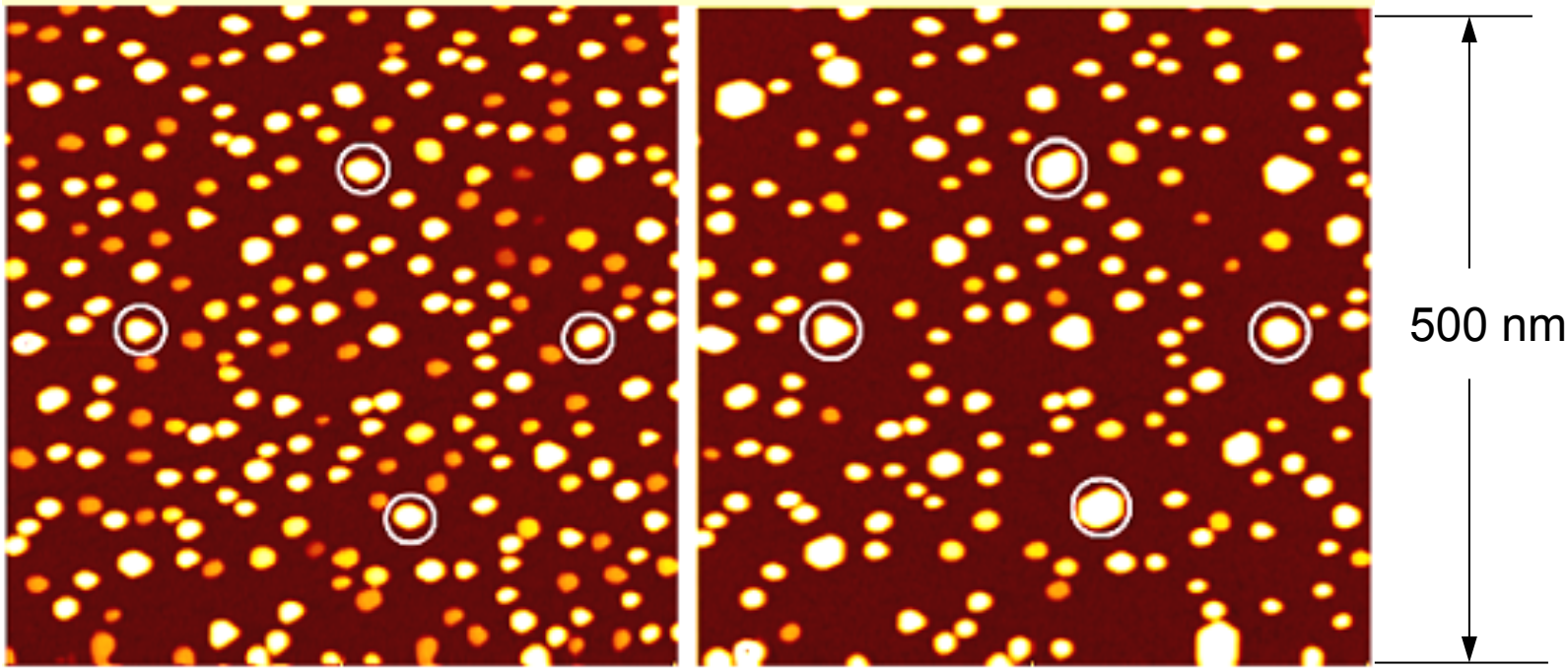
Coarsening of Pb Islands on Si(111)-7x7 Surface

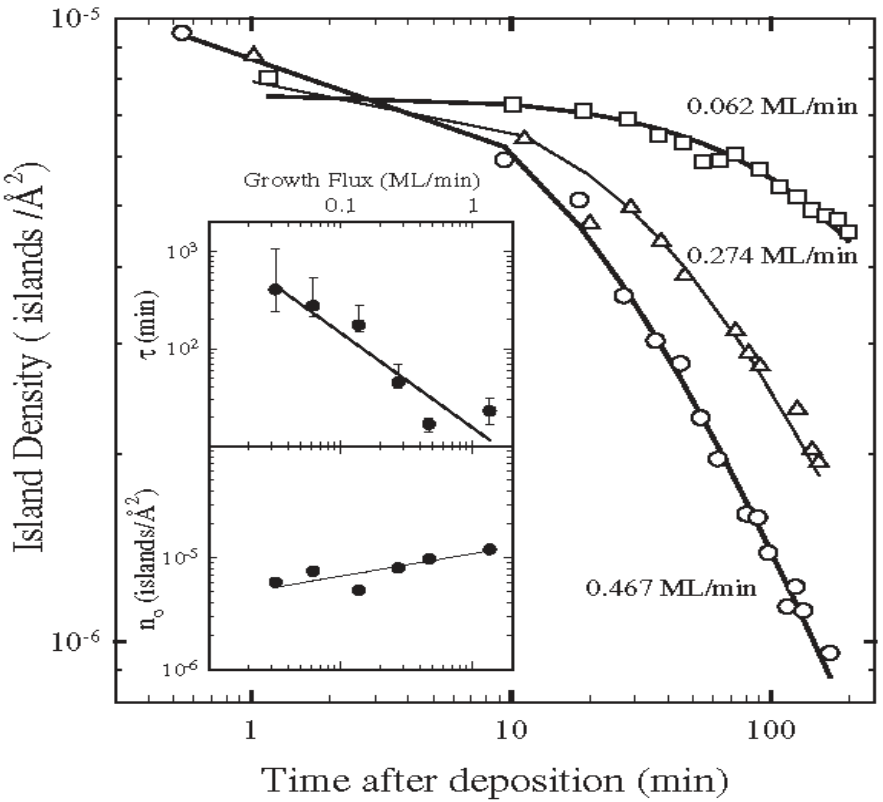
Hupalo and Tringides et al. 2006

System size: 500 x 500 nm^2 Initial number of islands: 330
Total coverage ~ 1.4 ML T=205 K



7-layer islands (circled) grow at the expense of lower height islands. After 2 hours, all 3-layer islands disappear.

Coarsening of Pb Islands on Si(111)-7x7 Surface



Contradictions to the classical theory:

* **Island density measured at different flux rates do not approach each other at long time.**
* **Island decay times τ are orders of magnitude faster than expected from the classical analysis.**

C. A. Jeffrey,E. H. Conrad, R. Feng P, M. Hupalo C. Kim, P. J. Ryan, P. F. Miceli, and M. C. Tringides, PRL **96**, 106105 (2006)

Classical Theory of Islands Coarsening

$$\mu(R) \approx \frac{\omega\gamma}{R}$$ **Gibbs-Thomson**

Evolution of Island Density $n(t) = n_0 \left(1 + t/\tau\right)^{-\beta}$

Time constant: $\tau \propto (n_0)^{-1/\beta}$ $\beta = 2/(m+2)$ m = 0,1,2

For $t >> \tau$ $n(t) \propto t^{-\beta}$ **Island density is Independent of n_0**

Classical Island *vs* Quantum Island: Quantum Size Effects

Classical Island:

- Chemical potential of the island is only dependent on the curvature of the island.
- Islands are surrounded by and communicate through a dilute lattice gas of adatoms.

Quantum Island:

- Chemical potential of the island is dependent on the height of the island due to QSE, as well as curvature.
- Islands are surrounded by and communicate through a dense and dynamic wetting layer.

Outline

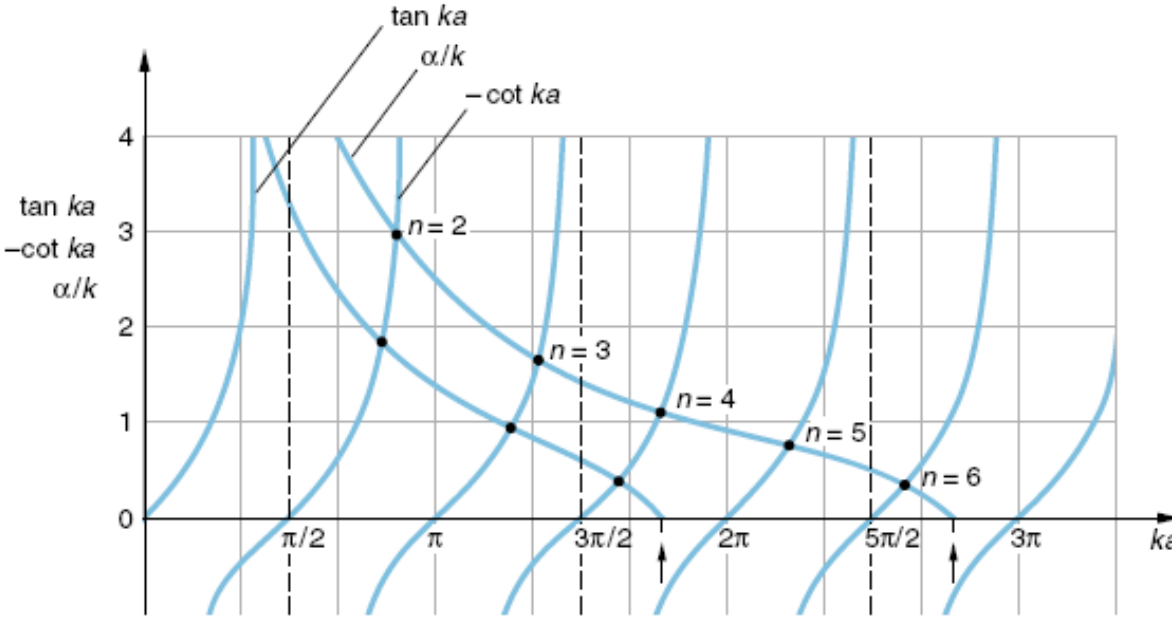
- Quantum size effects (QSE) in the stability and properties of Pb/Si(111)
- Theory of QSE in coarsening of Pb/Si(111)
- Collective dynamics of Pb wetting layer
- Summary

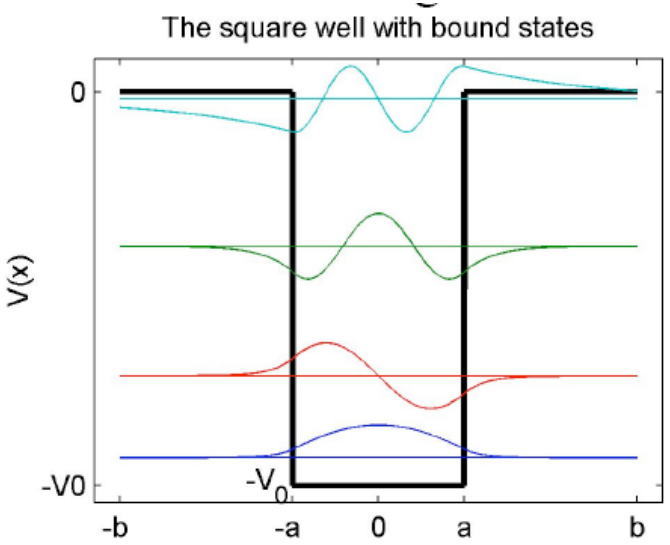
Quantum Size Effect (QSE)

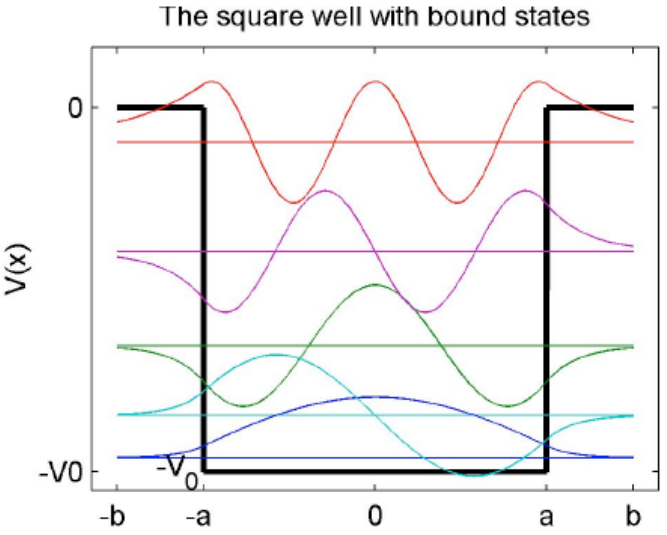
Square well potential

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V(x)\right]\psi(x) = E\psi(x)$$

$$V_0 \to \infty \qquad E_n = \frac{n^2\pi^2\hbar^2}{8ma^2}$$







Energy levels are discrete and dependent on the width of the well

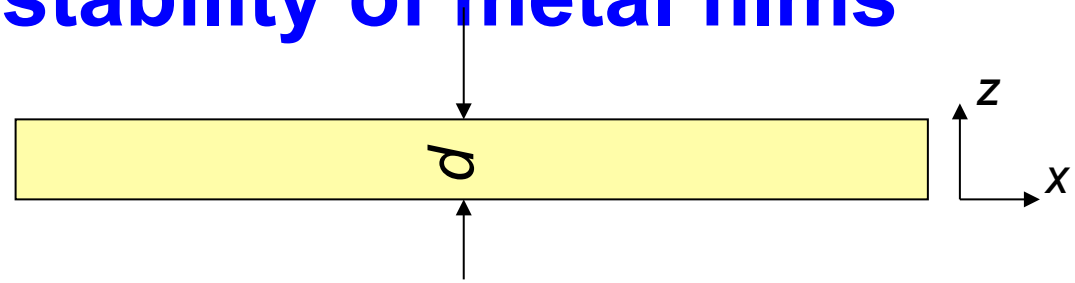
QSE on the stability of metal films

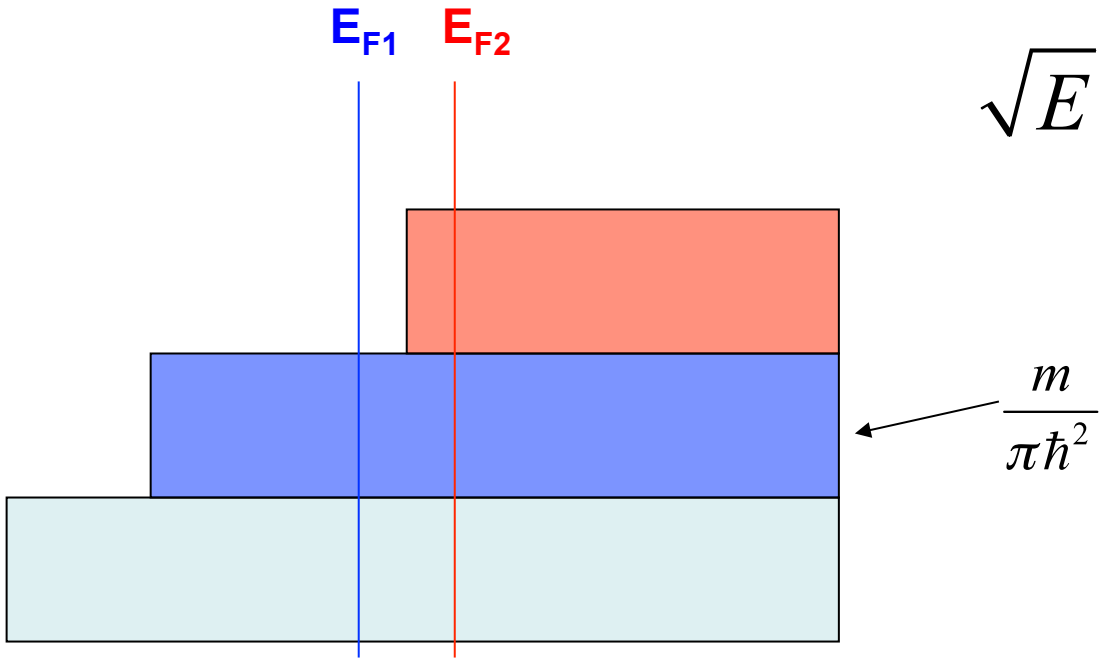
Schulte, *Surf. Sci.* **55**, 427 (1976);

Feibelman, *PRB* **27**, 1991 (1983); Feibelman and Hamann, *PRB* **29**, 6463 (1984);

Zhang, Niu, Shih, *PRL* **80**, 5381 (1998);

Yeh, Berbil-Bautista, Wang, Ho, and Tringides, *PRL* **85**, 5158 (2000).

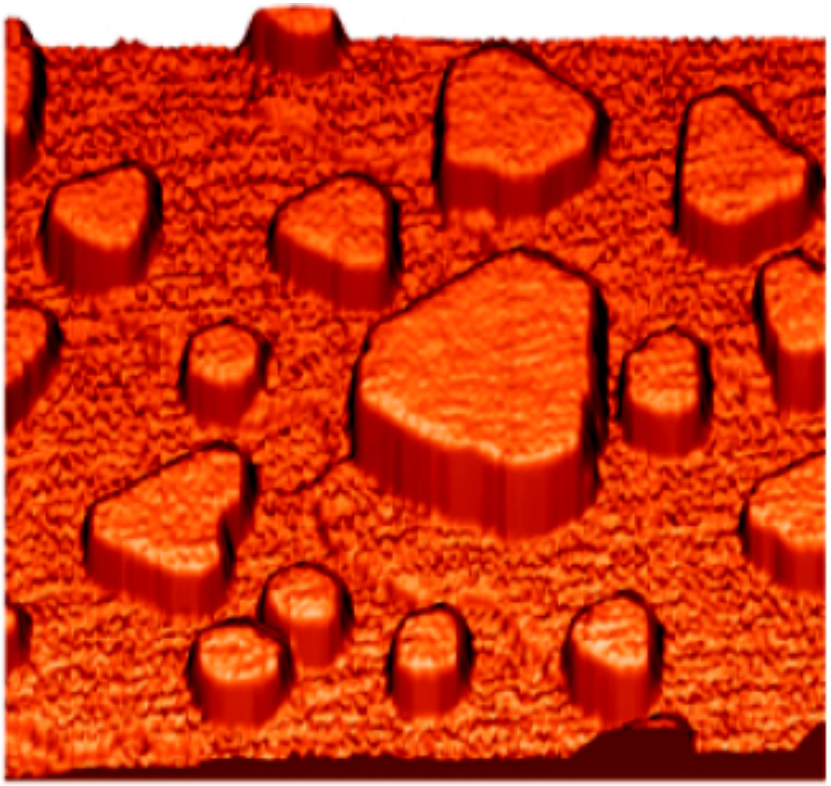




Condition for energy minima

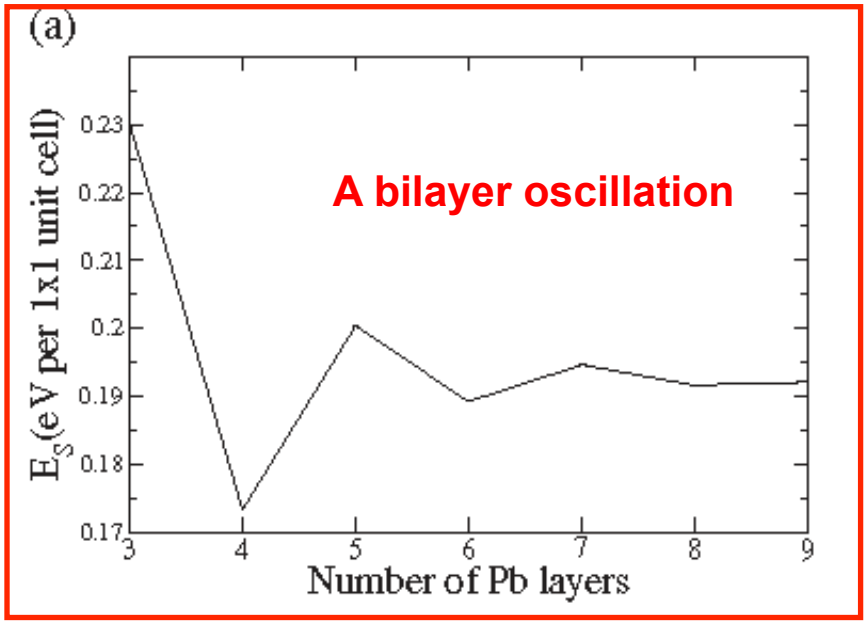
$$md = n\frac{\lambda_F}{2}$$

Formation and stability of Pb islands on Si(111) surface due to QSE



(Hupalo et al. PRB 64 155307(2001))

Quantum Size Effects in Pb/Si(111)



Condition for energy minima $md = n\frac{\lambda_F}{2}$

For Pb: $d = 2.86A, \lambda_F = 3.93A$

Therefore $2d \approx 3\frac{\lambda_F}{2}$

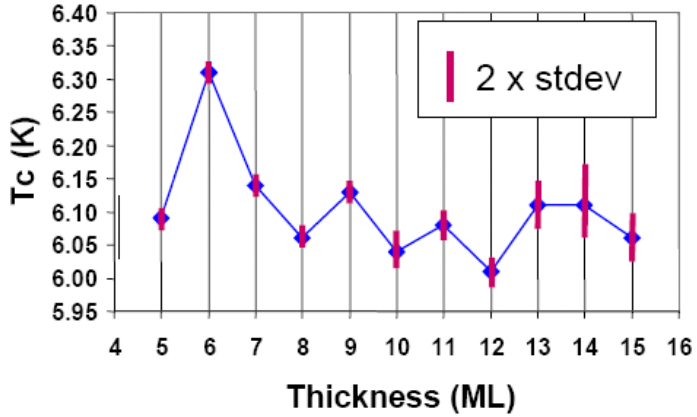
Z. Zhang, Q. Niu, and C.-K. Shih, Phys. Rev. Lett. 80, 5381 (1998).

Yeh, Berbil-Bautista, Wang, Ho, and Tringides, *PRL* 85, 5158 (2000).

C. M. Wei and M. Y. Chou, Phys. Rev. B 66, 233408 (2002).

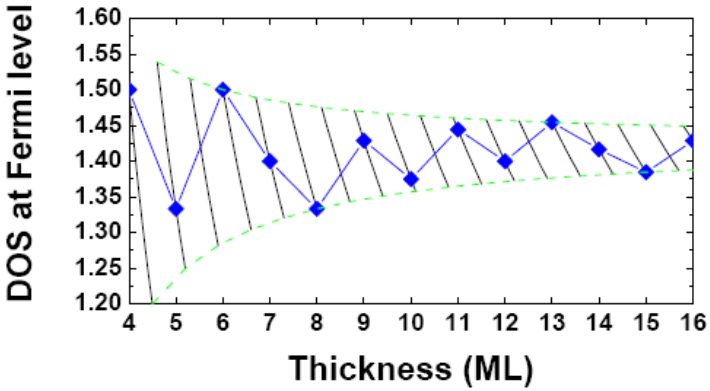
QSE in superconductivity: Pb/Si(111)

(a)



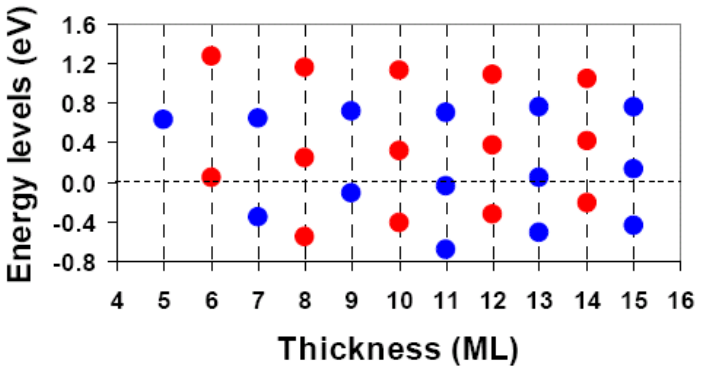
(a) A variation of critical temperatures, Tc determined from temperature-dependent gap measurements for 5-15 ML.

(b)



(b) The calculated density of states near a Fermi level, DOS (EF) as a function of the layer thickness.

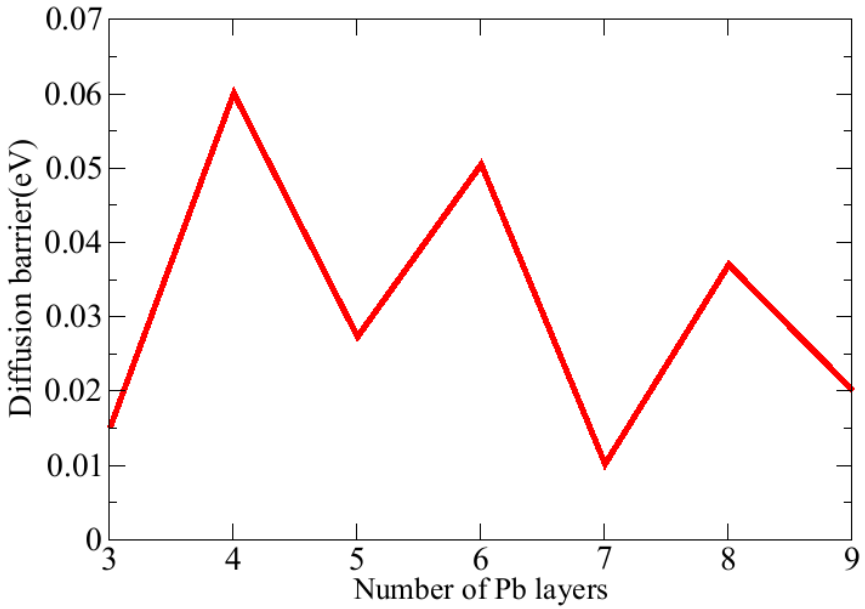
(c)



(c) The positions of QWS determined experimentally.

D. Eom, S. Qin, M. Y. Chou, and C.K. Shih, Phys. Rev. Lett. **96,** 027005/1-4 (2006).

QSE: Pb Adatom Diffusion on Pb(111) film

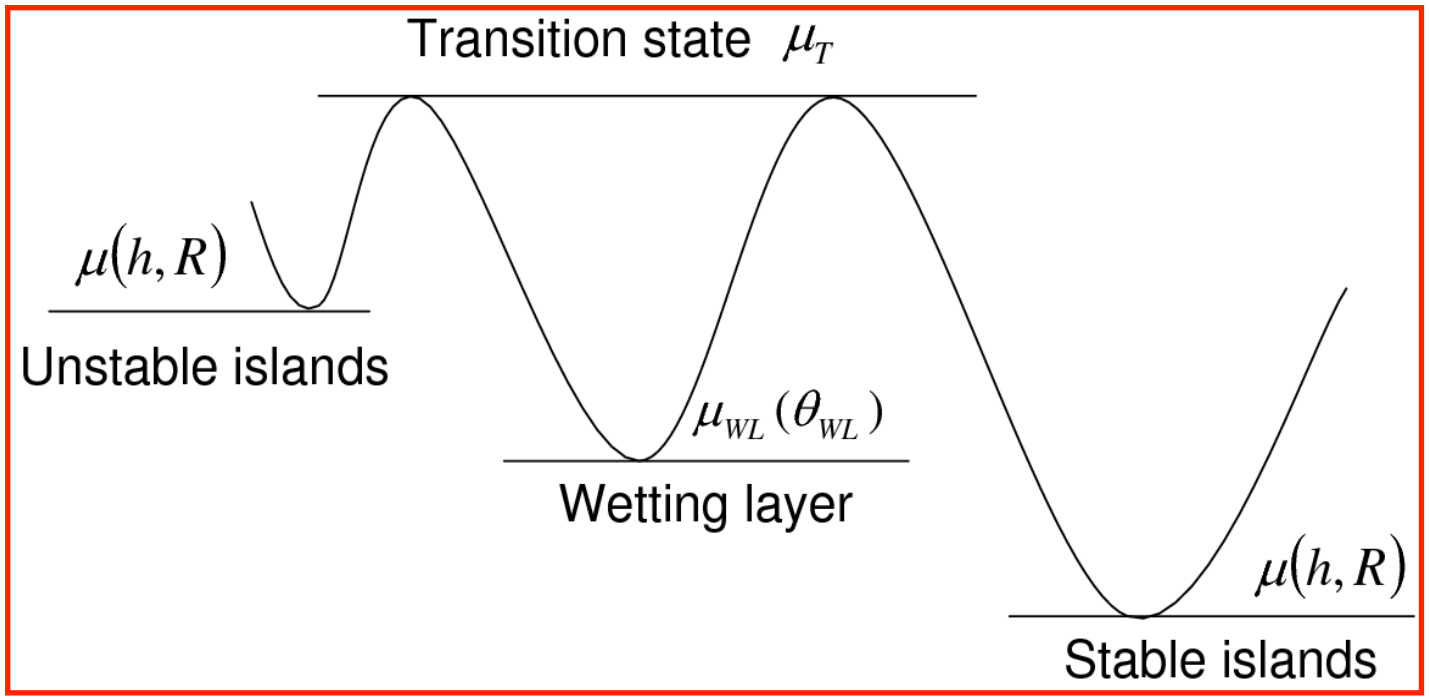


- **Small diffusion barriers: 0.01 – 0.06 eV**
- **Higher diffusion barrier for stable thickness**
- **Lower diffusion barrier for unstable thickness**

T. L. Chan, C. Z. Wang, M. Hupalo, M. C. Tringides, and K. M. Ho, Phys. Rev. Lett. **96**, 226102 (2006).

Experiment: Li et al., PRB 74, 075410 (2006)

Model for Coarsening of Quantum Islands



h: height of island

R: radius of island

1. Different levels of chemical potentials for islands and wetting layer.

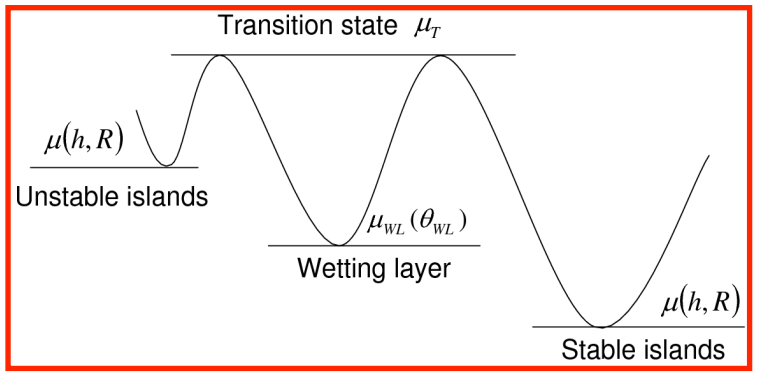
2. The transition state is the same for all islands, so the attachment barrier is the same, but detachment barrier is different for islands.

3. The wetting layer is assumed to be spatially uniform but varies with time.

Rate Equations for Coarsening of Quantum Islands

Size variation of island i with time:

$$\frac{dN_i}{dt} = 2\pi R_i \left(\sigma_i^+ - \sigma_i^-\right)$$

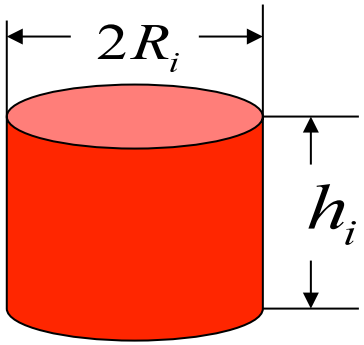


Attachment rate: $\sigma^+ = f_0 \exp\left[-\left(\mu_T - \mu_{WL}\right)/k_B T\right]$

Detachment rate: $\sigma^- = f_0 \exp\left[-\left(\mu_T - \mu_i\right)/k_B T\right]$

Islands are approximated to be mesas, so

$$N_i \approx \pi R_i^2 h_i$$



N_i is the size of island i

Attempt frequency $f_0 = 10^{13}/s$

Chemical Potential of Pb Islands

Free energy of island i

$$E_i = \pi R_i^2 h_i \varepsilon_b + \pi R_i^2 \gamma_i + 2\pi R_i h_i \gamma_s$$

γ_i and γ_s are surface energies of a mesa top and mesa sidewall, respectively.

ε_b is the bulk energy per atom.

During coarsening of Pb islands on Si(111), island height is assumed not change, so a partial chemical potential for radius growth is defined

$$\mu_i(R_i) = \frac{dE_i}{dN_i}\Big|_{h_i=const} = \Omega\left[\mu(h_i) + \frac{\gamma_s}{R_i}\right]$$

$\Omega = 1$ is the atomic volume and $\mu(h_i) = \varepsilon_b + \gamma_i / h_i$ $\gamma_s = 0.33$ eV

Define: $\mu_i = \gamma_i / h_i$ Then:

$\mu_T - \varepsilon_b = 0.66$ eV

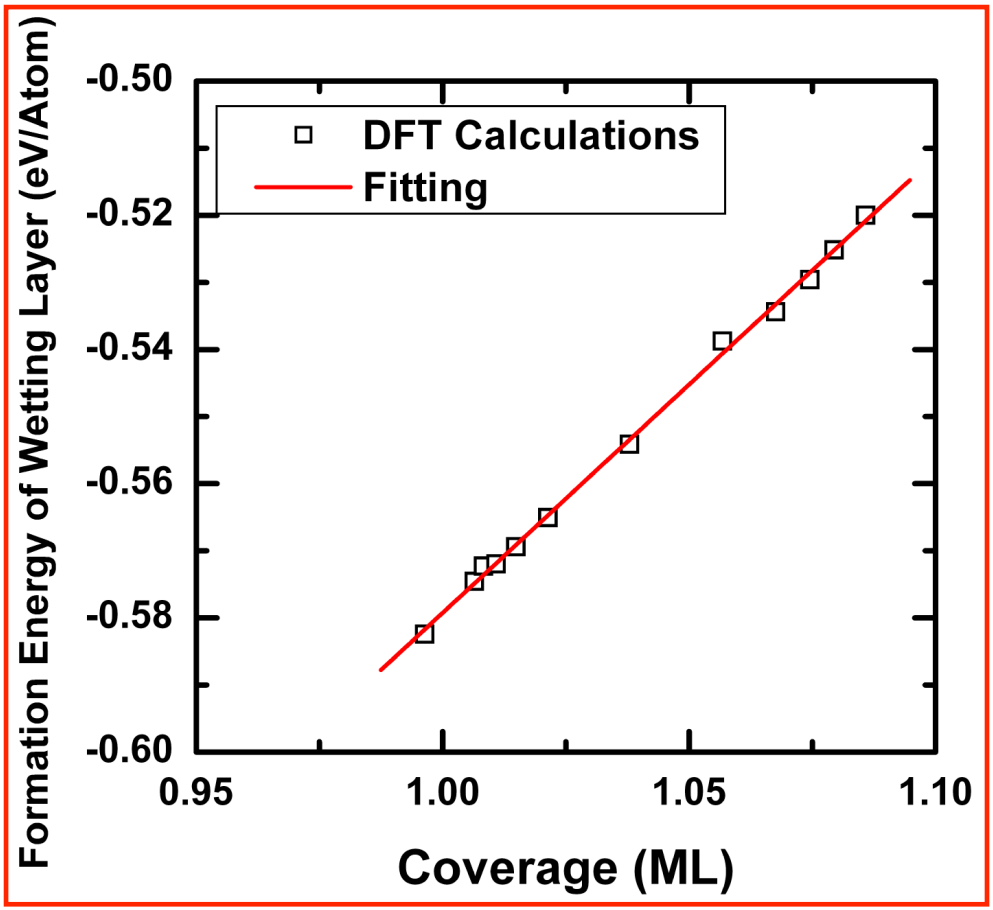
$\mu_3 = 0.0608$ eV $\mu_4 = 0.0491$ eV $\mu_5 = 0.0381$ eV

$\mu_6 = 0.0335$ eV $\mu_7 = 0.0274$ eV

Based on first-principles DFT calculations

Chemical Potential of the Wetting Layer

We model the dense wetting layer using ordered layers of Pb on Si(111) with various coverage. Chemical potential of wetting layer is determined based on the formation energy obtained from first-principles DFT calculations.



Energy of wetting layer:

$$E_{WL} = N_{WL}\varepsilon_b + N_{WL}\varepsilon$$

ε : Formation energy of wetting layer (per atom)

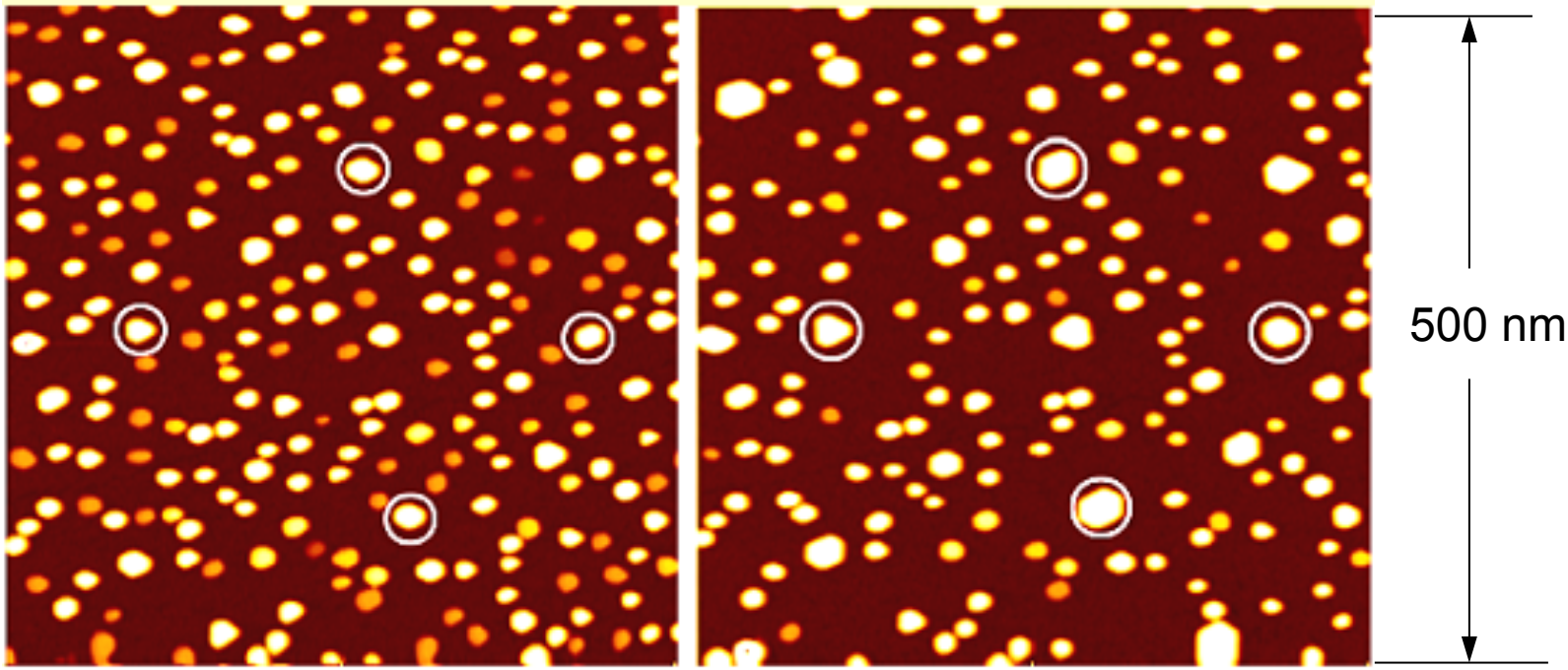
N_{WL}: number of atoms in wetting layer

$$\mu_{WL} = \varepsilon_b - 1.26 + 1.36\theta$$

Novel Coarsening of Pb Islands on Si(111)-7x7 Surface

Simulation is performed using the same condition as in the STM experiment

System size: 500 x 500 nm^2	Initial number of islands: 330
Total coverage ~ 1.4 ML	T=205 K



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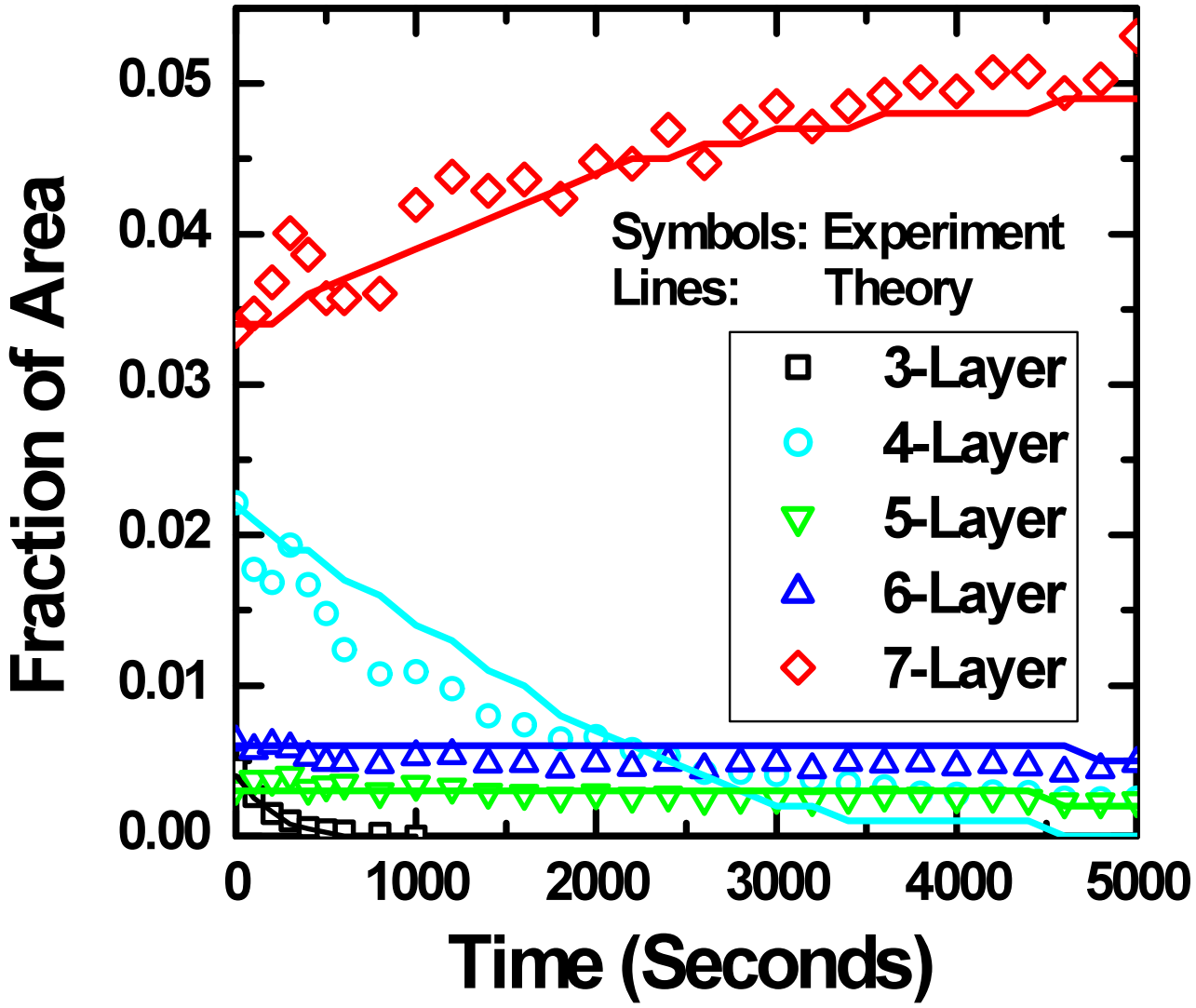
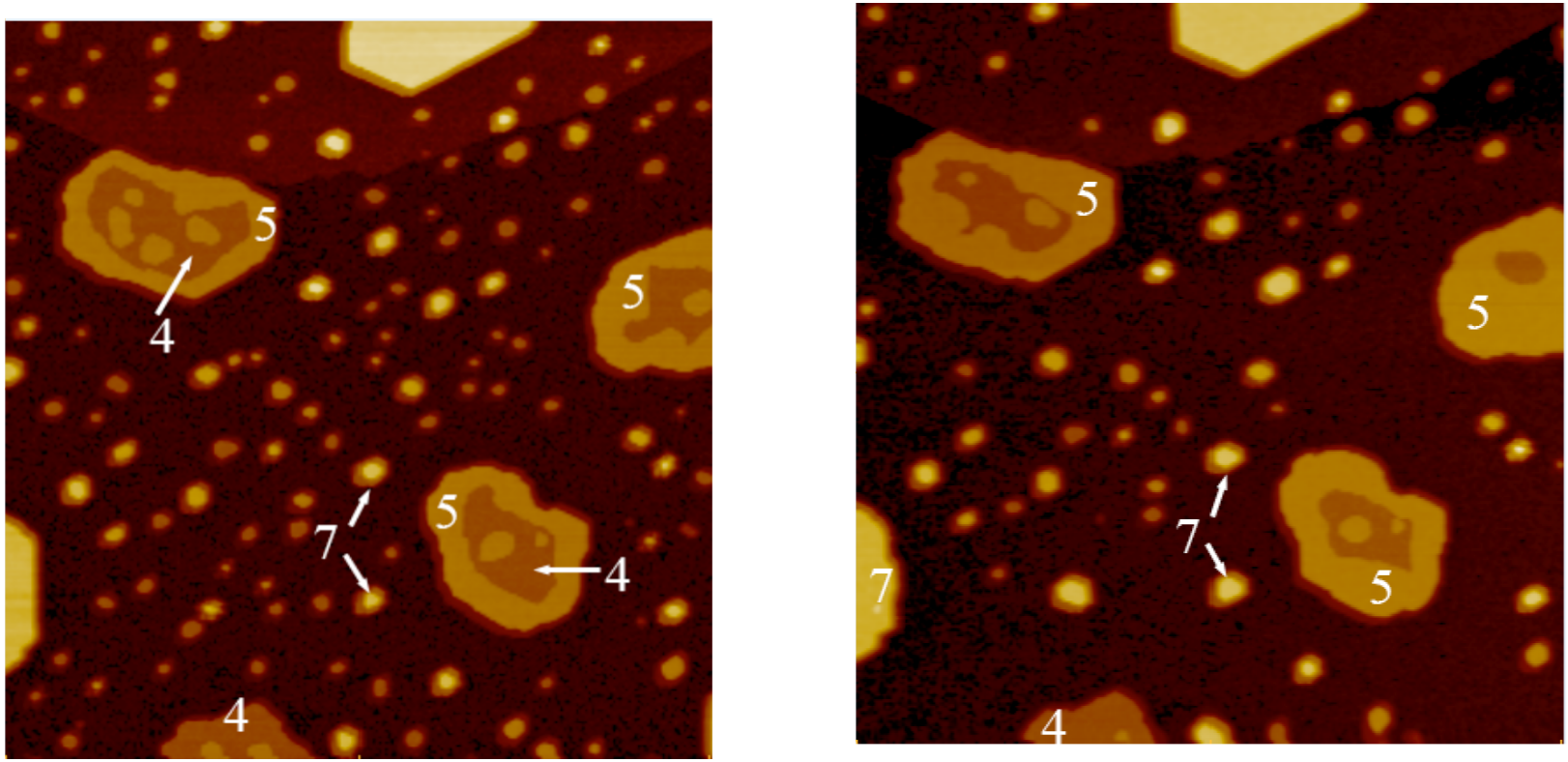


Li, Evans, Wang, Hupalo, Tringides, Chan, and Ho, *Surf. Sci. Lett*, **601**, L140 (2007)

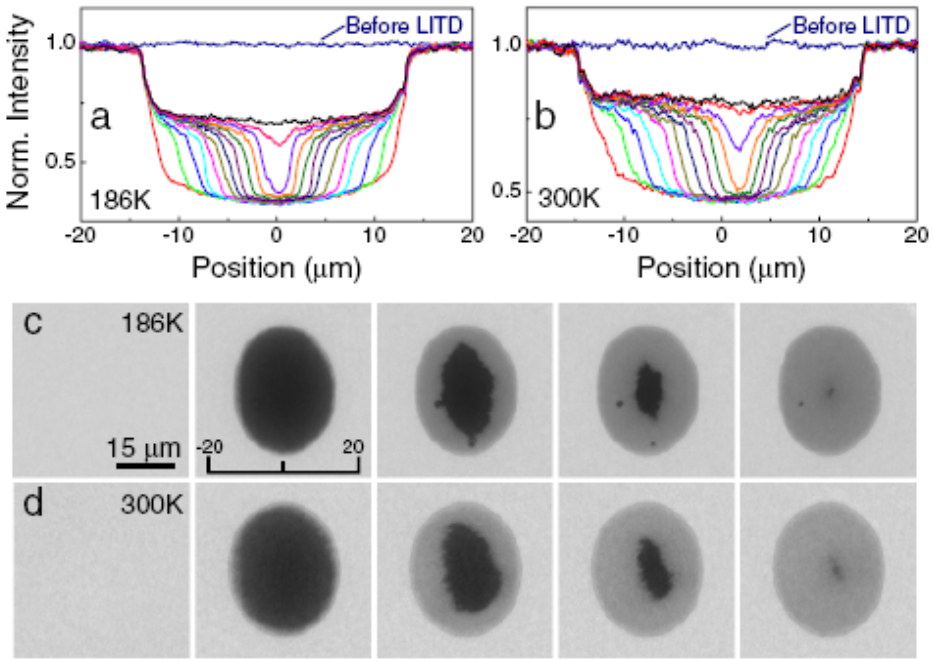
The same model can also describe the growth of large Pb islands on Si(111) surface including growth in height and ring expansion morphology

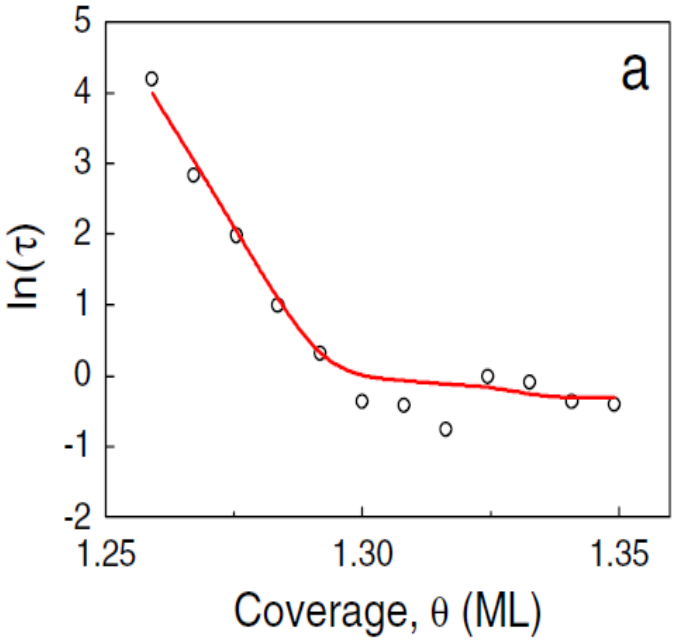


370 x 400 nm^2 T=190 K 92 min later

Li, Wang, Evans, Hupalo, Tringides, and Ho, *Phys. Rev. B (2009)*
Zhang, Li, Wang, Evans, Hupalo, Tringides, and Ho, *Phys. Rev. B (2011)*

Fast dynamics of Pb wetting layer on SI(111) observed by LEEM



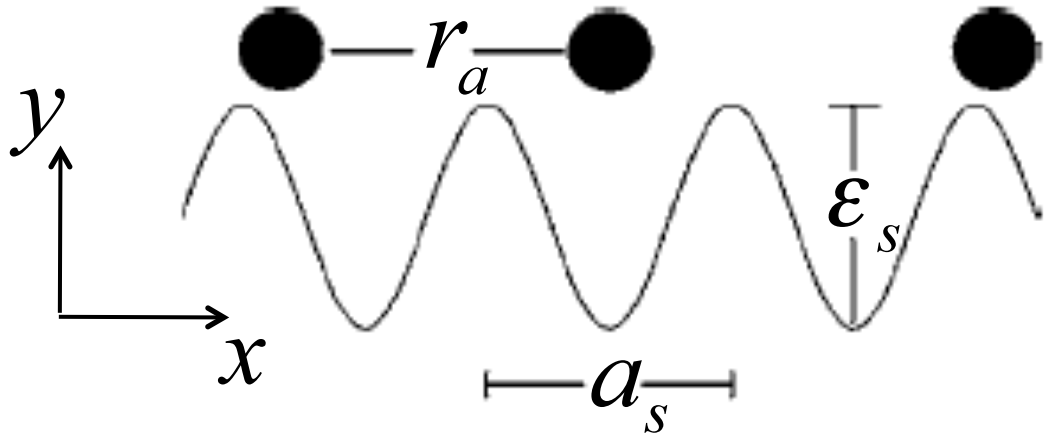


Profile evolution in the Pb α-phase on Si(111) following LITD for $\theta > \theta_c$ at (a), (c) T =186 K and (b), (d) T =300 K. The time intervals between LEEM intensity line profiles in (a) are Δt =4.1 sec and in (b) are Δt =0.2 sec. LEEM images are shown (l. to r.) in (c) at times t =0, 0.8, 15.7, 30.5, 45.4 sec, and in (d) at t =0, 0.01, 0.8, 1.4, 2.0 sec.

The dependence of the equilibration time τ, for LITD hole radius of 17 μm upon the initial wetting layer coverage θ, at 341 K

K. L. Man, M. C. Tringides, M. M. T. Loy, and M. S. Altman, PRL **101,** 226102 (2008)

MD simulation based on generalized Frenkel-Kontorova (FK) model

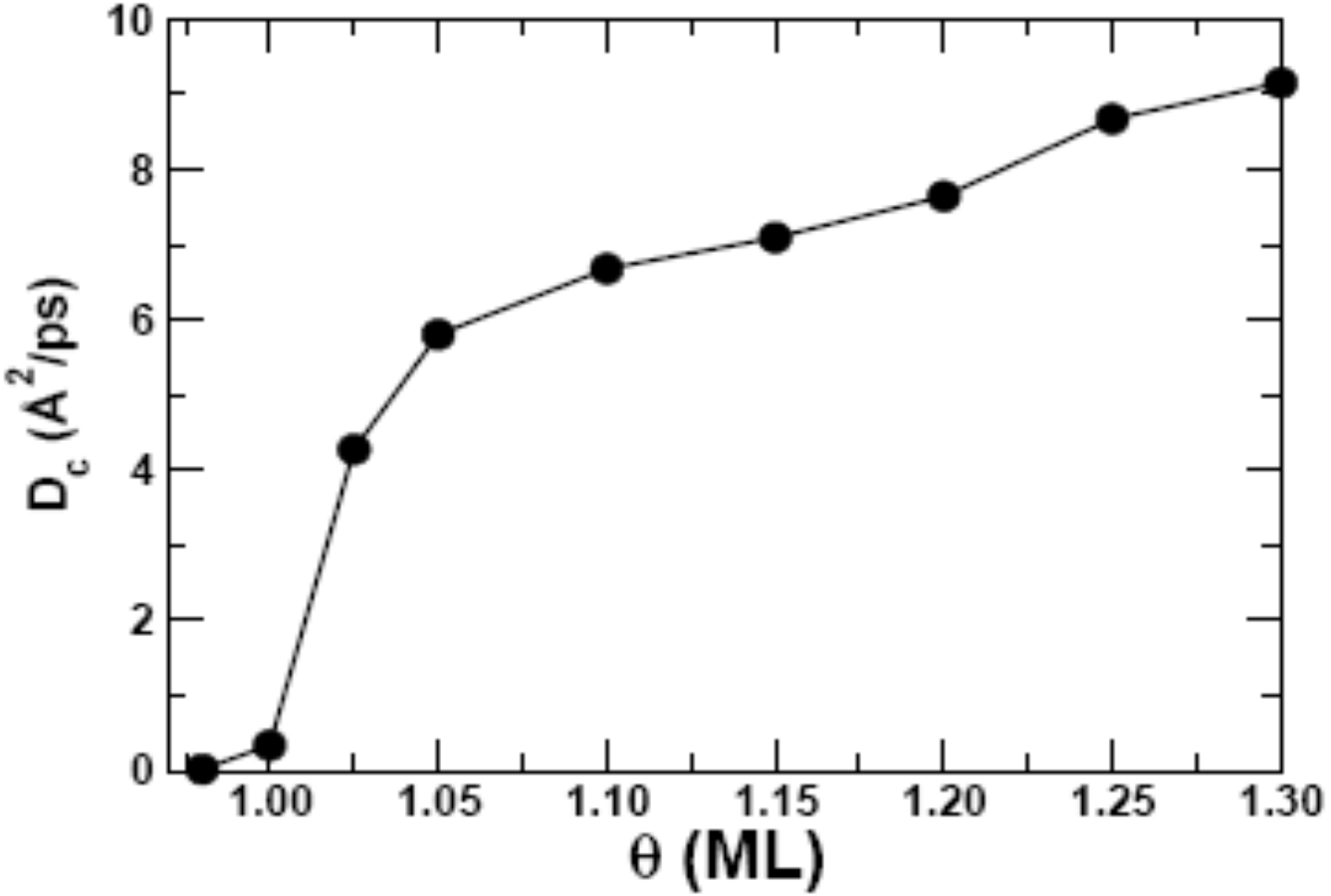


Calculate the Intermediate scattering function:

$$F(q,t) = \frac{1}{N}\left\langle \left(\sum_{j=1}^{N} e^{-i\vec{q}\cdot R_j(t+t_0)} \right)\left(\sum_{l=1}^{N} e^{i\vec{q}\cdot R_l(t_0)} \right) \right\rangle$$

Then determine the coverage-dependent collective diffusion constant $D_c(\theta)$

Coverage dependence of diffusion constant: Pb wetting layer on Si(111)



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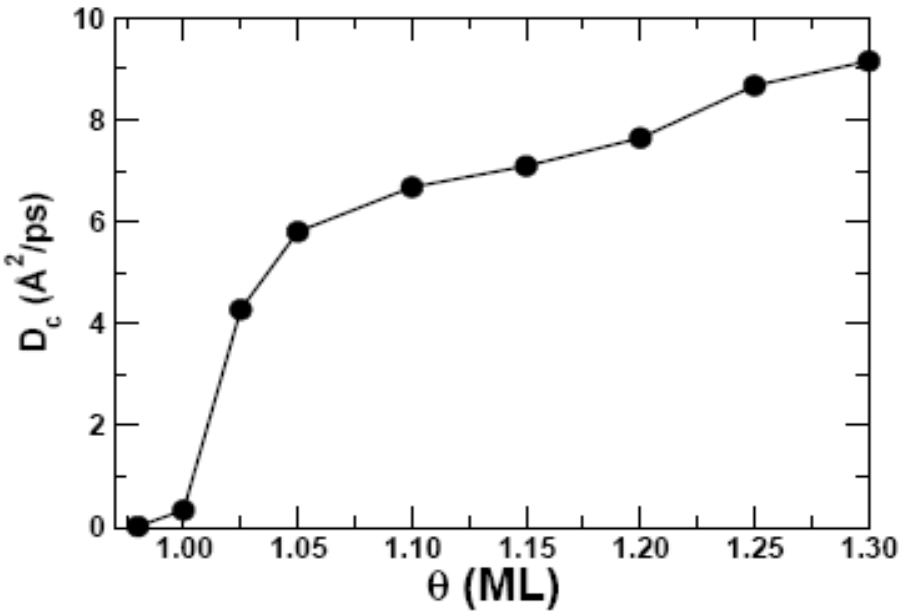

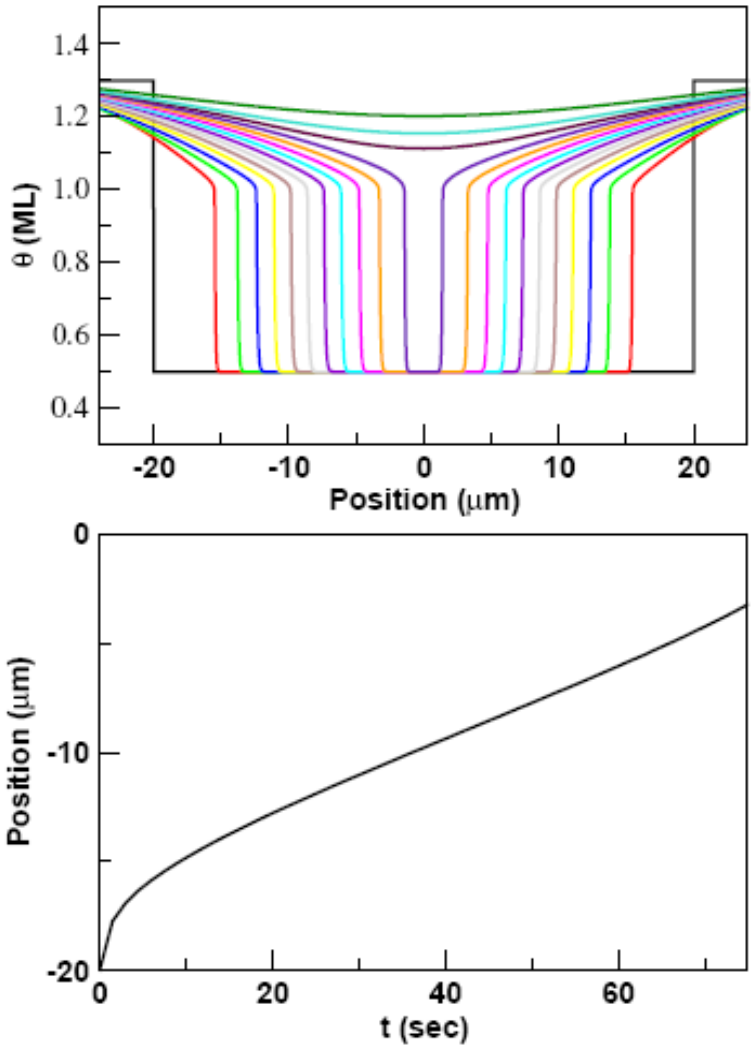

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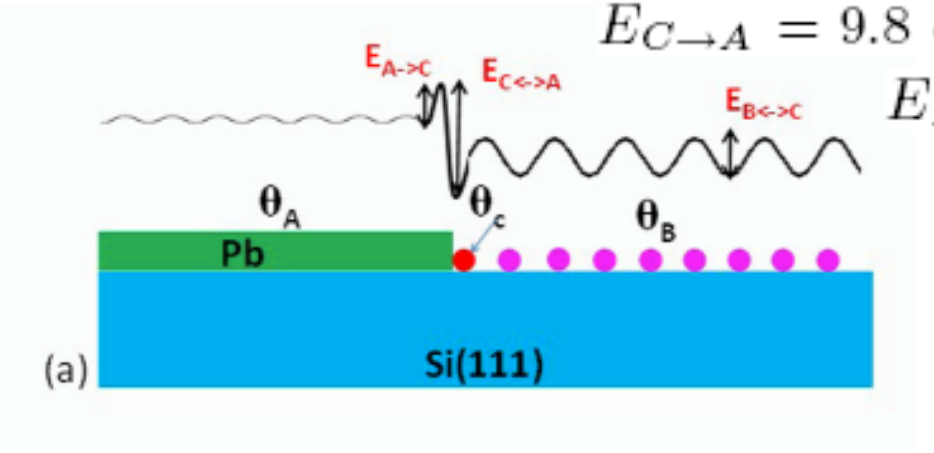
$$\frac{\partial \theta}{\partial t} = D_c(\theta)\left(\frac{1}{r}\frac{\partial \theta}{\partial r} + \frac{\partial^2 \theta}{\partial r^2}\right) + \frac{\partial D_c(\theta)}{\partial \theta}\left(\frac{\partial \theta}{\partial r}\right)^2$$

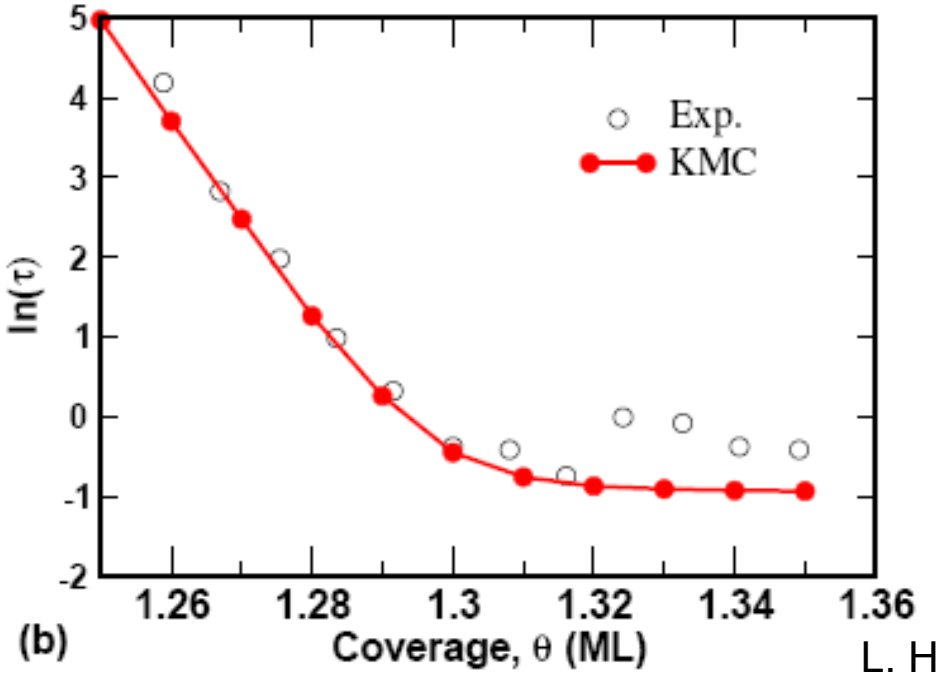
KMC Simulation

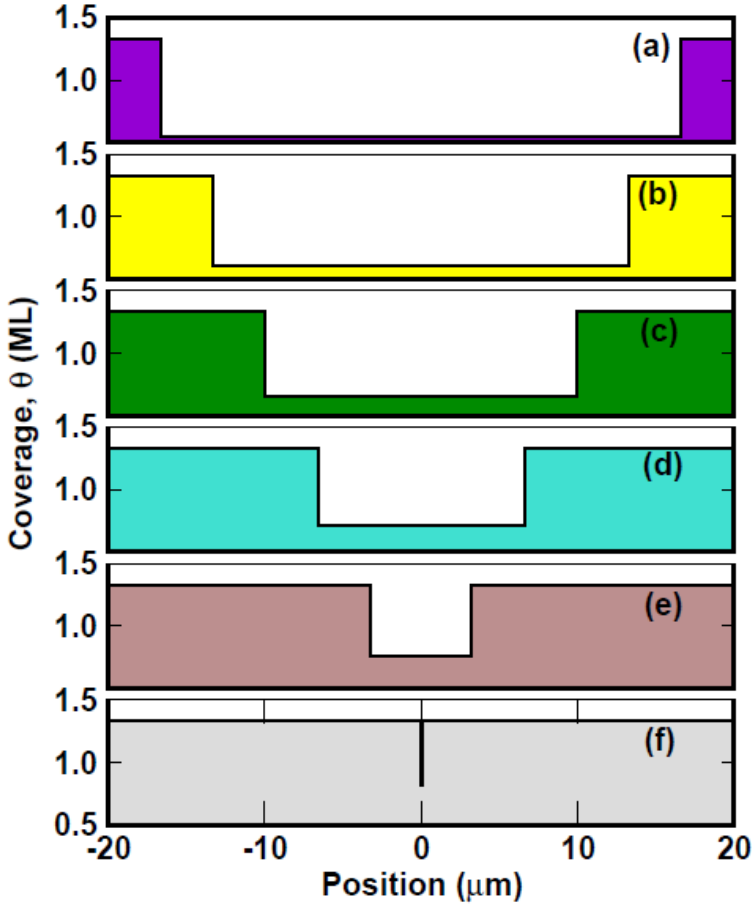
$$E_{A\to C} = -3.31\theta_A + 4.453 \text{ eV}$$

$$E_{C\to A} = 9.8 \text{ eV}$$

$$E_{B\leftrightarrow C, B\leftrightarrow B} = 0.18 \text{ eV}$$







Profile evolution with time

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Summary

♦Quantum size effects can be used to control the growth and properties of metal films on semiconductor surfaces.

♦Quantum size effects induce a strong dependence of chemical potential on island height, which affects the coarsening behavior.

♦The dense wetting layer in Pb/Si(111) acts as a fast mass transport medium.

Thank You