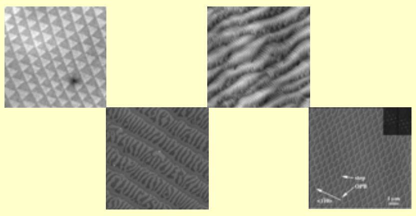

Dynamics of Si surface morphology

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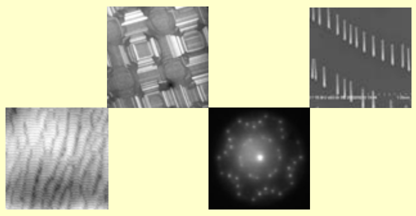
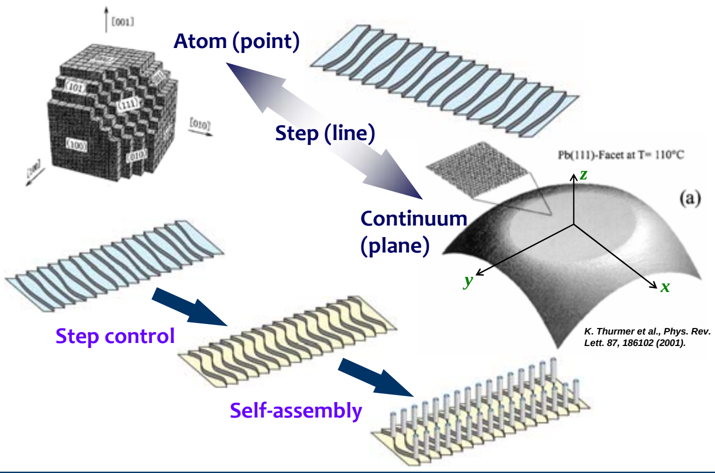


Motivation

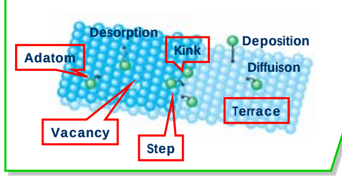
Control of nanostructure self-assembly

- Understanding step motion during annealing/growth/sublimation
- Position control of nanostructures using steps as templates



Approach

Many complicated processes involving a vast number of atoms



A few parameters in diffusion equation

Diffusion equation: $\frac{\partial c}{\partial t} = D\nabla^2 c - \frac{c}{\tau} + F$

Boundary condition: $\mp D\vec{n}\cdot\nabla c|_{\pm} = K_{\pm}(c - c_{eq})$

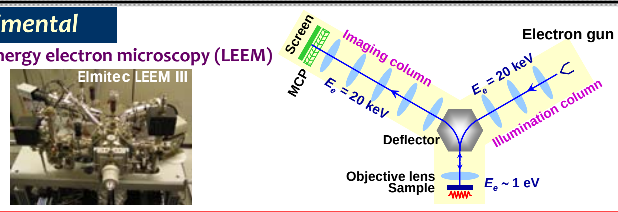
Gibbs-Thomson effect: $c_{eq} = c_{eq}^0 \exp\left(\frac{\gamma\Omega}{k_B T}\kappa\right)$

Kinetic distance: $d_k = D/K$

Surface mass diffusion constant: Dc_{eq}^0

Experimental

Low-energy electron microscopy (LEEM)

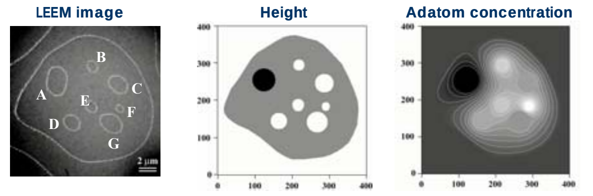


LEEM can routinely image surface structures in a spatial resolution of ~20 nm at a temporal resolution of ~0.1 s.

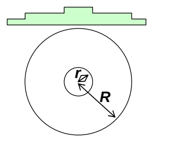
LEEM is suitable for studying surface dynamical processes.

Diffusion equation

1. Solved in the actual geometries using the finite element method.



2. Solved analytically in simple geometries.



$$\frac{dA}{dt} = 2\pi r D\frac{dc}{dr} = 2\pi \Omega D = 2\pi \Omega D \frac{-c_{eq}^0\left(1+\frac{a^2\tilde{\beta}}{kTr}\right)+c_{eq}^0\left(1+\frac{a^2\tilde{\beta}}{kTR}\right)}{\frac{D}{K_r r}+\frac{D}{K_R R}+\ln\frac{R}{r}}$$

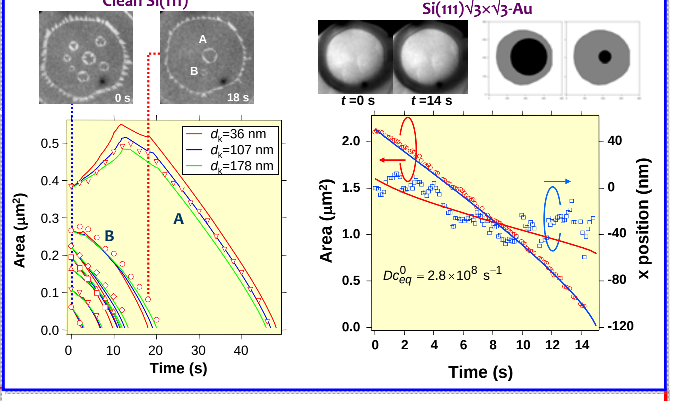
Diffusion limited: $r >> \frac{D}{K_+} = d_+$

$A(t) \propto (t_0 - t)^{2/3}$

Attachment/detachment limited: $r << \frac{D}{K_+} = d_+$

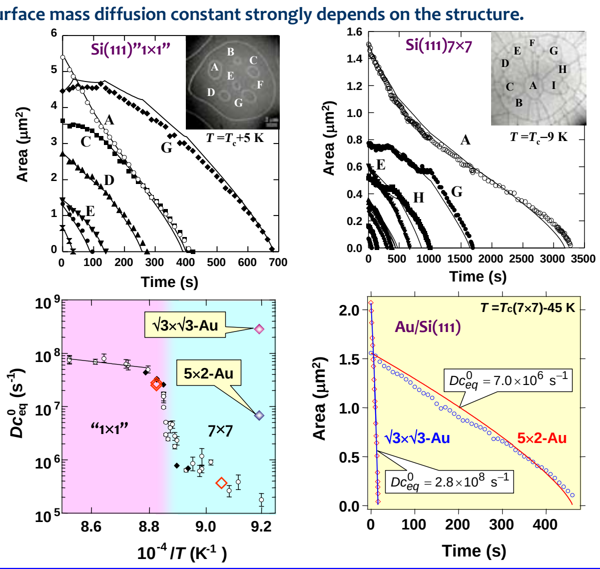
$A(t) \propto (t_0 - t)$

➤The diffusion equation analyses well reproduce the experiments.

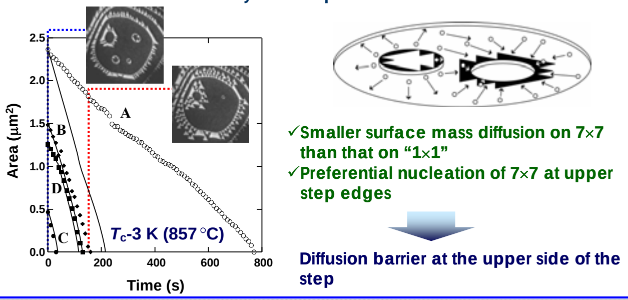


Surface mass transport

➤Surface mass diffusion constant strongly depends on the structure.



➤Diffusion barrier caused by the two phase coexistence

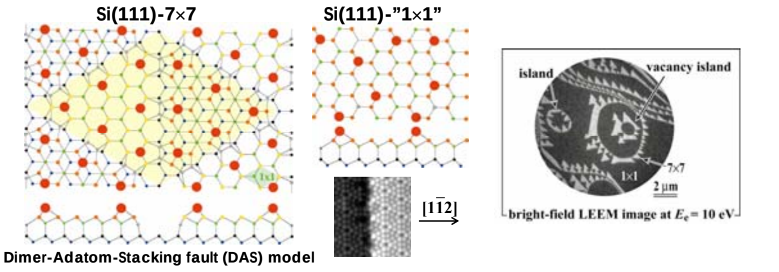


✓Smaller surface mass diffusion on 7×7 than that on "1×1"

✓Preferential nucleation of 7×7 at upper step edges

Diffusion barrier at the upper side of the step

Si(111)

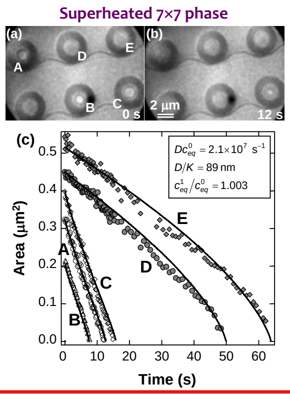


Dimer-Adatom-Stacking fault (DAS) model

➤Faster decay of islands with metastable structures

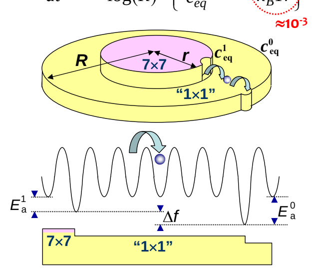
⇒ Difference in the surface energy

Superheated 7×7 phase



$$\frac{dA}{dt} \cong -\frac{2\pi\Omega Dc_{eq}^0}{\log(R)}\left\{\frac{c_{eq}^1}{c_{eq}^0} - 1\right) + \frac{\gamma\Omega}{k_B Tr}\right\}$$

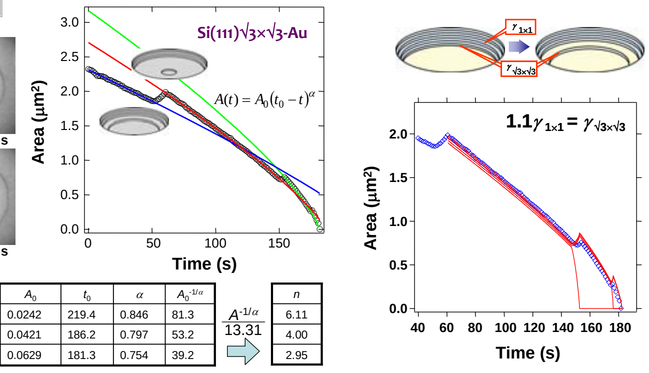
≈10^{-3}



$$c_{eq}^1/c_{eq}^0 = \exp\left(\frac{\Delta f \Delta T\Omega}{k_B T}\right) = 1.003$$

$f_{7\times7}^0 - f_{1\times1}^0 = 170$ meV/(1×1) cell

➤Decay of multi-step crater



Time (s)	A_0	t_0	α	$A_0^{-1/\alpha}$
0-51	0.0242	219.4	0.846	81.3
61-152	0.0421	186.2	0.797	53.2
154-175	0.0629	181.3	0.754	39.2

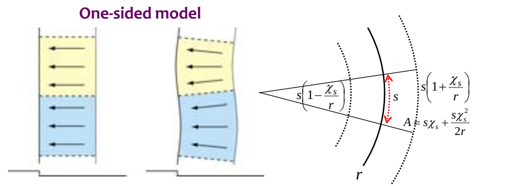
$A^{-1/\alpha}$ 13.31 ⇒

n
6.11
4.00
2.95

Step wandering

➤Step wandering induced by homoepitaxy on Si(111) during "1×1"-7×7 phase transition.

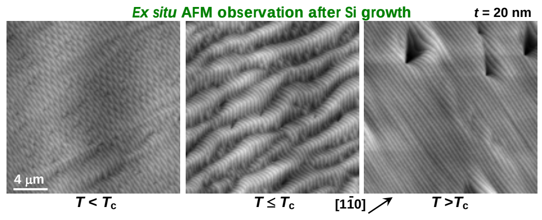
One-sided model



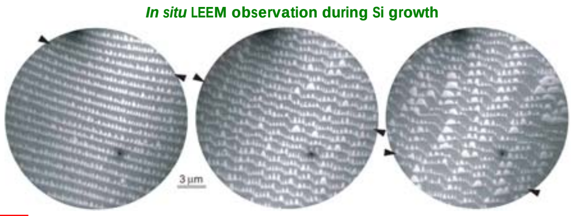
$$\delta_d + \delta_s = (F_c - F_{eq})\Omega \cdot \frac{\chi_s^2}{2r} - F_{eq}\frac{\Omega\tilde{\beta}}{k_B Tr}\Omega\chi_s = 0$$

$$F_c = F_{eq}\left(1 + \frac{2\Omega\tilde{\beta}}{\chi_s k_B T}\right)$$

Ex situ AFM observation after Si growth



In situ LEEM observation during Si growth



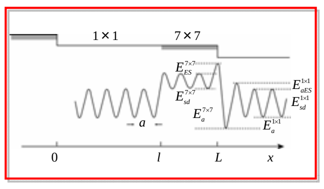
Boundary conditions (diffusion-limited case + continuum model)

◇step

- $c(0) = c_{1\times1}$
- $c(L) = c_{7\times7}$

◇phase boundary

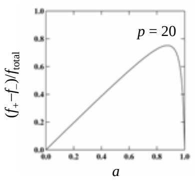
- $D_{1\times1}\frac{dc}{dx}\Big|_{l-} = D_{7\times7}\frac{dc}{dx}\Big|_{l+}$
- $\frac{c|_{l-}}{c_{1\times1}} = \frac{c|_{l+}}{c_{7\times7}}$



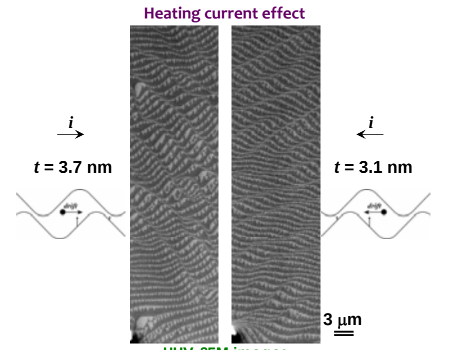
Flux from lower-side terrace

$$f_+ = D_{1\times1}\frac{\partial c}{\partial x}\Big|_{0+} = \frac{FL}{2}\cdot\frac{pa^2 - a^2 - p}{pa - a - p} \quad \left(a = \frac{l}{L},\ p = \frac{D_{1\times1}c_{1\times1}}{D_{7\times7}c_{7\times7}}\right)$$

f_+ has a maximum value $\sqrt{p}/(\sqrt{p}+1)$ at $a = \sqrt{p}/(\sqrt{p}+1)$.



Heating current effect

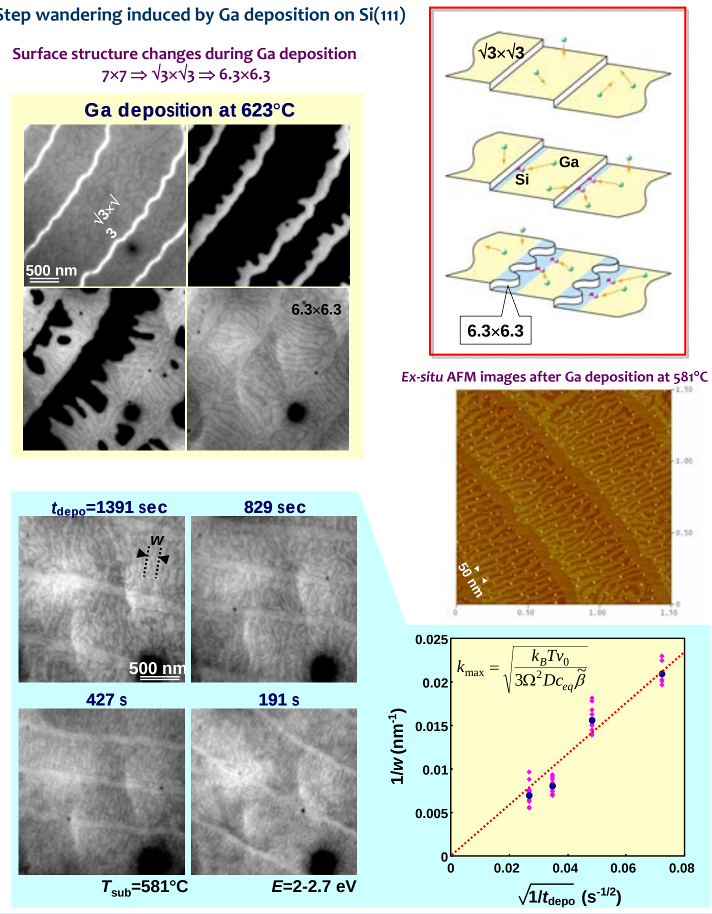


UHV-SEM images

➤Step wandering induced by Ga deposition on Si(111)

Surface structure changes during Ga deposition

7×7 ⇒ √3×√3 ⇒ 6.3×6.3



Domain coarsening

➤Von Neumann's law well explains the coarsening of 7×7 domains.

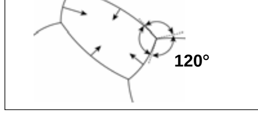
Curvature rule $S = K\sigma\mu$

- K: curvature
- σ: free energy per unit length
- μ: mobility
- S: speed

The enclosed area A decreases at a constant rate.

$\frac{dA}{dt} = -2\pi\sigma\mu$

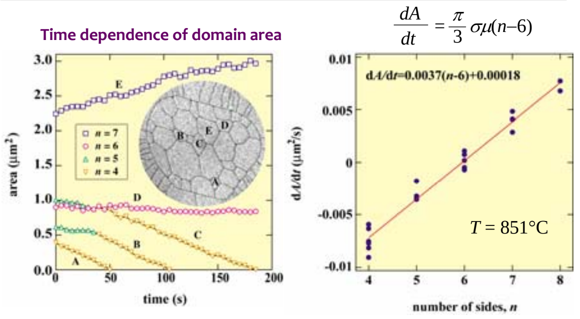
Von Neumann's law



The rate of change of area of the cell with n sides

$$\frac{dA}{dt} = \frac{\pi}{3}\sigma\mu(n-6)$$

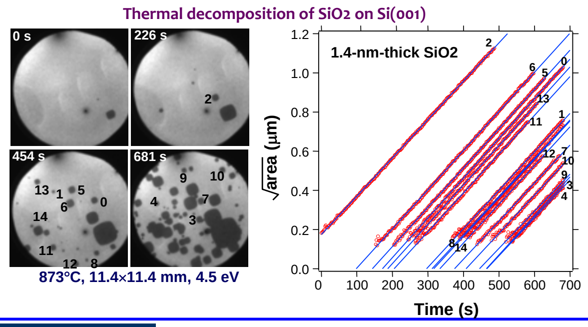
Time dependence of domain area



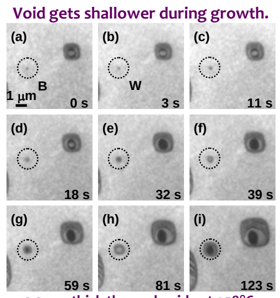
Void growth

➤dA/dt∝√A: The periphery reaction of the Si monomer with the oxide controls the void growth.

Thermal decomposition of SiO2 on Si(001)



Void gets shallower during growth.



2.2-nm-thick thermal oxide at 958°C

Si(001); attachment/detachment limited case

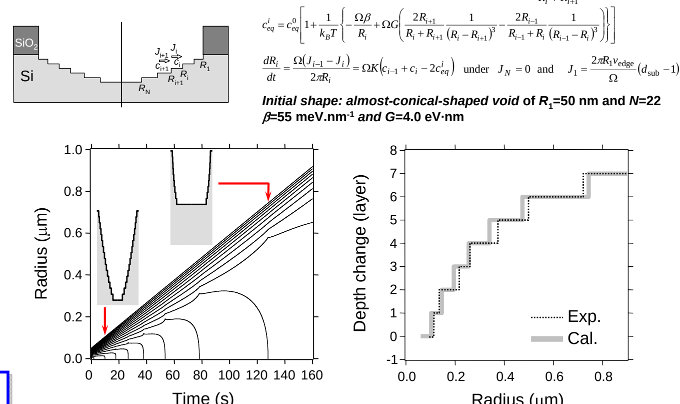
$$J_i = 2\pi R_{i+1}K(c_i - c_{eq}^{i+1}) = 2\pi R_i K(c_{eq}^i - c_i) \longrightarrow c_i = \frac{R_i c_{eq}^i + R_{i+1}c_{eq}^{i+1}}{R_i + R_{i+1}}$$

$$c_{eq}^i = c_{eq}^0\left[1 + \frac{1}{k_B T}\left(-\frac{\Omega\beta}{R_i} + \Omega G\left(\frac{2R_{i-1}}{R_i + R_{i-1}}\frac{1}{(R_i - R_{i-1})^2} - \frac{2R_{i+1}}{R_{i+1} + R_i}\frac{1}{(R_{i+1} - R_i)^2}\right)\right)\right]$$

$$\frac{dR_i}{dt} = \frac{\Omega(J_{i-1} - J_i)}{2\pi R_i} = \Omega K(c_{i-1} + c_i - 2c_{eq}^i) \quad \text{under} \quad J_N = 0 \ \text{and} \ J_1 = \frac{2\pi R_1 v_{edge}}{\Omega}(d_{sub} - 1)$$

Initial shape: almost-conical-shaped void of R_1=50 nm and N=22

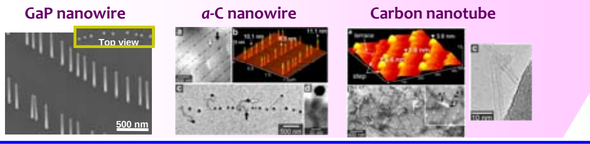
β=55 meV.nm^{-1} and G=4.0 eV·nm



Summary

➤ Understanding the surface mass transport

➤ Control of the step morphology



Control of nanostructure self-assembly

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