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An Elastic Effect in Crystal Growth: Change of Growth Modes with Dislocations in Heteroepitaxy

—in an elastic lattice model—

H. Katsuno and M. Uwaha*

(work done in collaboration with Y. Saito**and H. Uemura*)

Department of Physical Sciences, Ritsumeikan University
* Department of Physics, Nagoya University
** Department of Physics, Keio University

Background picture by H. Hibino

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Introduction: Epitaxial growth modes and elastic interaction

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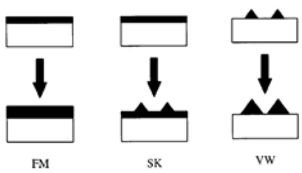
Growth modes in heteroepitaxy

Frank-van der Merwe (FM) mode: layer growth

Stranski-Krastanov (SK) mode: island growth after layer growth

Volmer-Weber (VW) mode: island growth

Lattice mismatch small ↔ large



Surface energy low ↔ high

Strain energy high ↔ low

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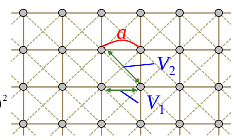
Two-dimensional elastic lattice model

- Interaction potential between the atoms

$$V_1(r) = -J_1 + V_1'(a)(r-a) + \frac{1}{2}K_1(r-a)^2$$

$$V_2(r) = -J_2 + V_2'(\sqrt{2}a)(r-\sqrt{2}a) + \frac{1}{2}K_2(r-\sqrt{2}a)^2$$



Atoms are connected by two kinds of springs.

- Stability condition for the square lattice

$$\sigma_0 = -V_1'(a) = \sqrt{2}V_2'(\sqrt{2}a)$$

Spring V_1 is compressed, Spring V_2 is stretched.

- Condition for isotropic elastic body

$$K_1 = 2K_2$$

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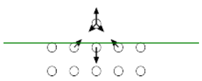
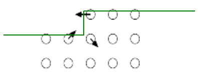
Elastic interaction between "defects"



an adatom

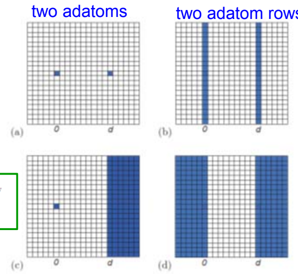
force dipole
elastic dipole

$$m_{\alpha\gamma} \equiv \sum_{\boldsymbol{r}} r_\alpha f_{\boldsymbol{r}}^\gamma$$



a step

$U \sim 1/d^3$ $U \sim 1/d^2$



an adatom and a terrace

$U \sim 1/d.$

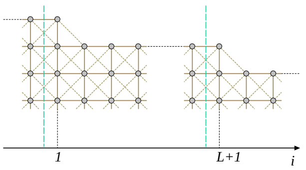
two terraces
two steps of opposite sign

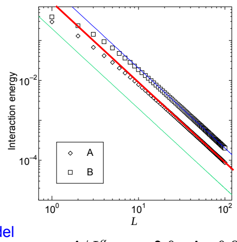
$U \sim \ln d.$

$$U_{\text{step-step}} \sim \frac{1}{d^2}$$

Step interaction in the two-dimensional elastic lattice model

model A





lattice model A/L^{α}, $\alpha = 2.0$, $A = 0.88$

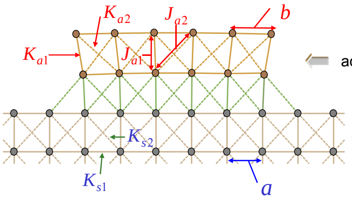
Effective force dipoles are several times stronger than expected.

continuum elasticity (force dipole on flat terrace) A/L^{α}, $\alpha = 2$, $A = 0.1963$

H. Uemura, Y. Saito, M. Uwaha, J. Phys. Soc. Jpn. **70** (2001) 743

Two-dimensional elastic lattice model and heteroepitaxial growth modes

Epitaxial island in the two-dimensional elastic lattice model



- natural bond energy of adsorbate

$J_{a1}(b) = 2J_{a2}(b) = 0.1$

- spring constants

$K_{a1} = K_{s1} = 2.0,\ K_{a2} = K_{s2} = 1.0$

- misfit parameter

$-0.2 \le f = \dfrac{b-a}{a} \le 0.2$

- strength of spontaneous stress

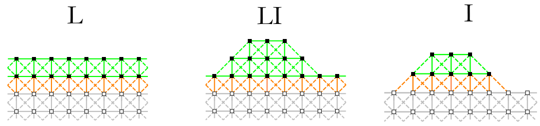
$\sigma_0 \sim 0.1$

H. Uemura, Y. Saito, M. Uwaha, J. Phys. Soc. Jpn. **72** (2003) 2856

Possible equilibrium shapes

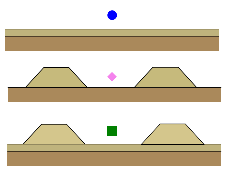
The shape of the island is the Wulff shape.

- Without dislocations



layer	Layer + island	island
FM mode	SK mode	VW mode

Growth modes for the square lattice model

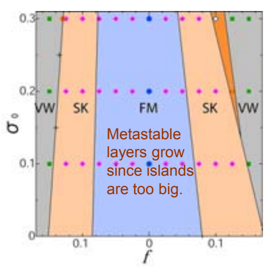


spontaneous stress σ_0

lattice misfit parameter

$f = \dfrac{b-a}{a}$

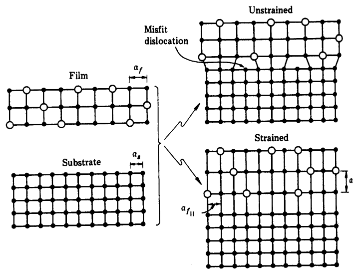
growth mode diagram in spontaneous stress-misfit plane



Katsuno, Uemura, Uwaha, Saito, J. Cryst. Growth **275** (2005) e283

Misfit dislocation in the two-dimensional elastic lattice model

Relaxation of the stress by introduction of misfit dislocations



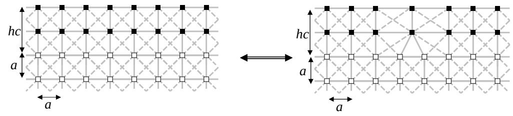
introduction of dislocations

accumulation of stress

Theoretical estimation of the elastic energy and the critical thickness for the introduction of a dislocation

coherent adlayer

adlayer with a dislocation



change of the elastic energy → estimation of the critical thickness

$$E_{\mathrm{el}} = \frac{E\,|\boldsymbol{b}|^2}{8\pi\left(1-\zeta^2\right)} \ln\left(\alpha\frac{h}{|\boldsymbol{b}|}\right) - \frac{E}{1-\zeta} f\,|\boldsymbol{b}|\,h$$

misfit parameter $f = \frac{b-a}{a}$

Young coefficient	E
Poisson ratio	ζ
Burgers vector	$\lvert\mathbf{b}\rvert$

b natural lattice constant of the adsorbate
a natural lattice constant of the substrate

J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth, **27** (1974) 118

Two-dimensional lattice model of misfit dislocations

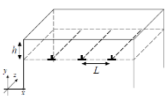
Interatomic potentials

V_1	NN bond energy	K_1	NN spring const.	σ_x σ_y	NN force
V_2	NNN bond energy	K_2	NNN spring const.	σ_2	NNN force

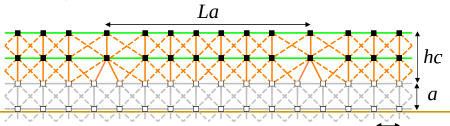
$f = \frac{b-a}{a}$ misfit

isotropic solid

$$K_1 = 2K_2$$



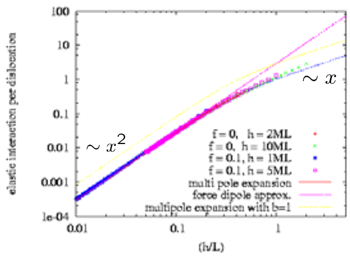
The interaction at the interface is the same as the adsorbate.



Elastic interaction energy between dislocations

$$E^{\mathrm{con}}_{\mathrm{int}}(h,L) = \tilde{E}^{\mathrm{con}}_{\mathrm{int}}\left(x=\frac{h}{L}\right) = \frac{{d^{(1)}_{xx}}^2}{8\pi\mu}\left[2\pi x \coth 2\pi x - 1 - \frac{\lambda}{\lambda+\mu}\ln\frac{\sinh 2\pi x}{2\pi x}\right] \quad \left(x<\frac{1}{2}\right)$$

$d^{(1)}_{xx}$: force dipole moment for $h=1$ $\quad x=\frac{h}{L}$



Period dependence

small x $\rightarrow \tilde{E}^{\mathrm{con}}_{\mathrm{int}}(x) \sim x^2 \sim \frac{1}{L^2}$

large x $\rightarrow \tilde{E}^{\mathrm{con}}_{\mathrm{int}}(x) \sim x \sim \frac{1}{L}$

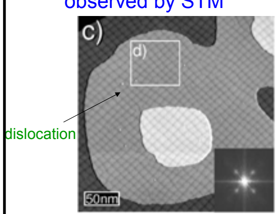
The effect of misfit is very small.

Katsuno, Uwaha, Saito, J. Phys. Soc. Jpn. **76** (2007) 044605

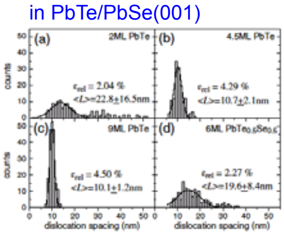
Application (1): Web of misfit dislocations

Network of misfit dislocations

PbTe/PbSe(001) surface observed by STM



distance between dislocations in PbTe/PbSe(001)

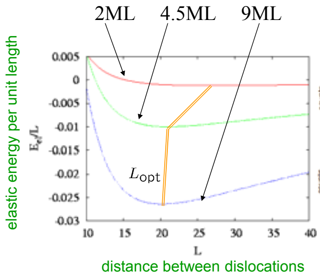


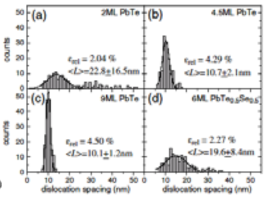
•Periodic network of dislocations is formed.

•thicker adsorbate: closer dislocations narrower distribution

K. Wiesauer and G. Springholz, Phys. Rev. Lett., **88** (2002) 015507

Equilibrium distance between dislocations





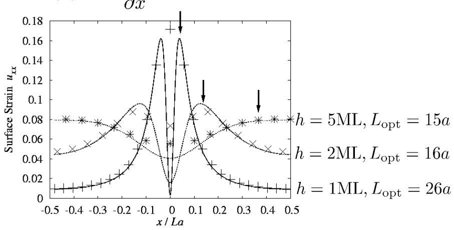
K. Wiesauer and G. Springholz, Phys. Rev. Lett. **88** 015507 (2002)

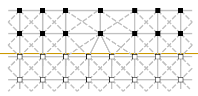
Katsuno, Uwaha, Saito, J. Phys. Soc. Jpn. **76** (2007) 044605

Application (2): Prediction of nucleation site

Surface strain

strain $u_{xx}(x) = \dfrac{\partial u_x(x)}{\partial x}$ misfit : $f = 0.075$





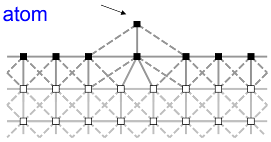
Elastic energy of an adatom

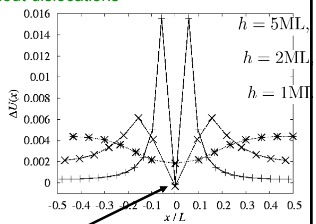
$$\Delta U(x) = U(x) - E_{\mathrm{d}} - E_{\mathrm{a}} + Ef^2hL/2$$

E_{d} : dislocation energy without adatoms

E_{a} : adatom energy without dislocations

adsorbate atom





location of the atom

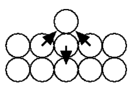
Nucleation will occur on top of the dislocation.

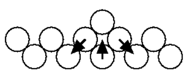
Interaction between an adatom and a misfit dislocation

elastic interaction energy $\sim -u_{xx}d_{xx}$

surface strain by a dislocation u_{xx} → the sign depends on ***f***

force dipole of an adsorbate atom d_{xx} → the sign depends on the face (or site)





(01) face : $d_{xx} < 0$
protuberant site

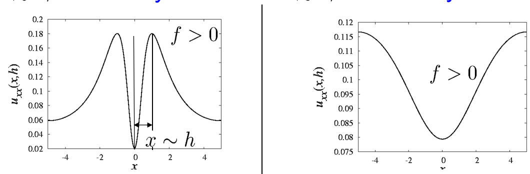
(11) face : $d_{xx} > 0$
hollow site

Position of the nucleation site of the new layer

elastic interaction: $-d_{xx}u_{xx}(h, L)$

$h \lesssim L/2$: thin layer

$h \gtrsim L/2$: thick layer



nucleation site:

$d_{xx}f < 0 \Rightarrow$ on top or midpoint

$d_{xx}f > 0 \Rightarrow x \sim h$ near the dislocation

nucleation site:

$d_{xx}f < 0 \Rightarrow$ on top

$d_{xx}f > 0 \Rightarrow$ midpoint

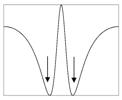
f : misfit parameter

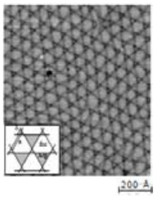
Growth of Ag on 2ML Ag/Pt(111)

Pt fcc (111) face hollow site $\rightarrow d_{xx} > 0,$

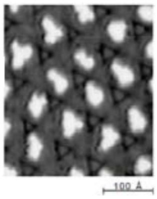
$a_{\mathrm{Ag}} > a_{\mathrm{Pt}} \rightarrow f > 0$

$\Rightarrow d_{xx} f > 0$ Nucleation near but not on top of the dislocations is predicted.





coverage 0.1ML →



H. Brune, M. Giovannini, K. Bromann and K. Kern, Nature **394** 451 (1998)

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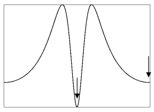
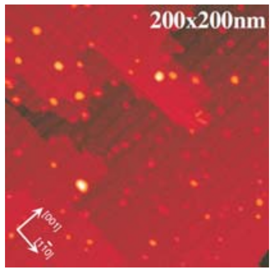
Growth of Ga on 5ML InAs/GaAs(110)

$\begin{pmatrix} d_{xx} < 0 \\ f > 0 \end{pmatrix} \Rightarrow d_{xx} f < 0$

$\Rightarrow$ on top and between the dislocations

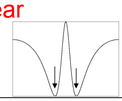
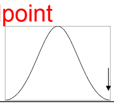
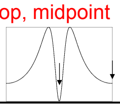
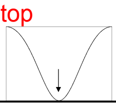




N. Oyama, A. Ohtake, S. Tsukamoto, N. Koguchi, and T. Ohno, Jpn. J. Appl. Phys. **43** L1422 (2004)

Prediction of the nucleation site

Location of the nucleation site depends on the force dipole *dxx* and the misfit parameter *f*.

	$h \lesssim L/2$ thin	$h \gtrsim L/2$ thick
$d_{xx} f > 0$	near 	midpoint 
$d_{xx} f < 0$	on top, midpoint 	on top 

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Application (3):
Prediction of growth modes

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Parameters

binding energies: $J_1 = 2J_2 = 0.1$

elastic constants: $K_1 = 2K_2 = 2$

adsorption strength: $\frac{J_{1,2}^{\mathrm{interface}}}{J_{1,2}^{\mathrm{Adsorbate}}} = 1.1$

misfit parameter: $f = \frac{b-a}{a} (> 0)$

Modification of parameters due to misfit

$$J_{1x} = J_1 - \sigma_0 f a - \frac{1}{2} K_1 (fa)^2$$

$$J_{1y} = J_1 + \frac{K_2}{K_1 + K_2} \sigma_0 f a - \frac{K_1 K_2^2}{2(K_1 + K_2)^2} (fa)^2$$

$$J_2' = J_2 + \frac{K_1}{2(K_1 + K_2)} \sigma_0 f a - \frac{K_1^2 K_2}{4(K_1 + K_2)^2} (fa)^2$$

$$\sigma_{1x} = \sigma_0 + K_1 f a$$

$$\sigma_{1y} = -2\sigma_2 = \sigma_0 - \frac{K_1 K_2}{K_1 + K_2} f a$$

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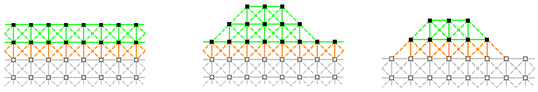
Possible equilibrium shapes

The shape of the island is the Wulf shape.

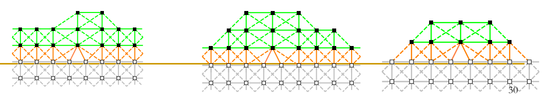
- Without dislocations

L LI I



- With dislocations

L+D LI+D I+D



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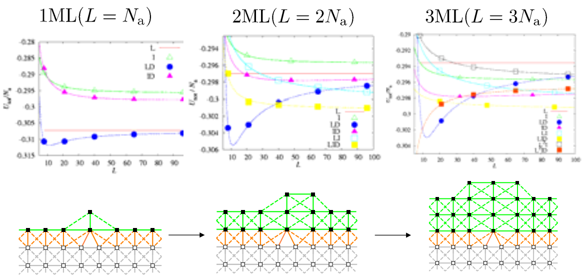
Search for minimum energy shape in increasing adsorbate *f* =0.1



layer growth with dislocations at the interface

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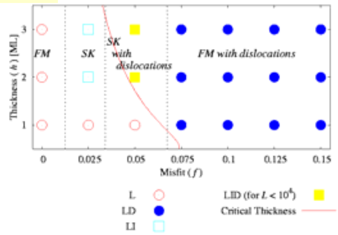
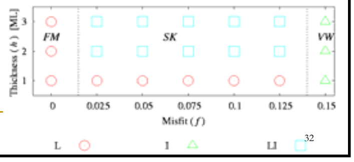
Possible growth modes

- For large misfit, growth modes with dislocations may appear.
- Islands and dislocations can coexist.
- VW does not appear. ($f < 0.2$)



If dislocations are not allowed →



Katsuno, Uwaha, Saito, Surf. Sci. **602** (2008) 3461

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Summary: Dislocations in epitaxial growth

- The period of the web of dislocations and its fluctuation is theoretically understood.
- On the web of dislocations
  Location of the nuclei of a new layer is determined by the sign of *f* and the sign of the force dipole d_{xx} of an adatom.
  $d_{xx} f < 0 \Rightarrow$ nucleation above dislocations
  $d_{xx} f > 0 \Rightarrow$ nucleation between dislocations
- As the misfit f increases, epitaxial growth mode changes:
  FM→ SK→ SK with Dislocations→ FM with Dislocations

  cf: Without dislocations FM→ SK→ VW

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