結晶表面でのステップダイナミクス
Step dynamics on crystal surfaces

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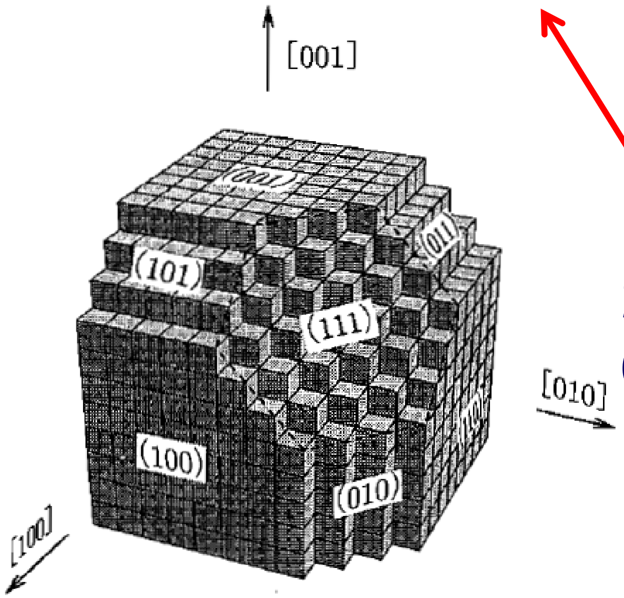
NTT Basic Research Laboratories

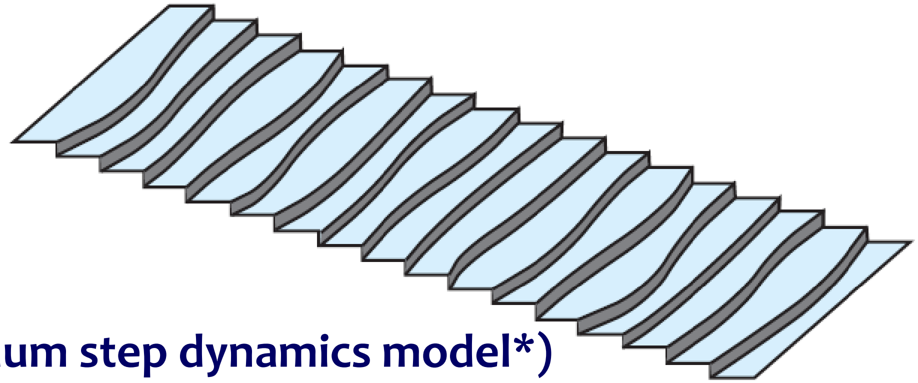
表面形状の階層的解釈

Atom (atomistic lattice-gas dynamics model*)

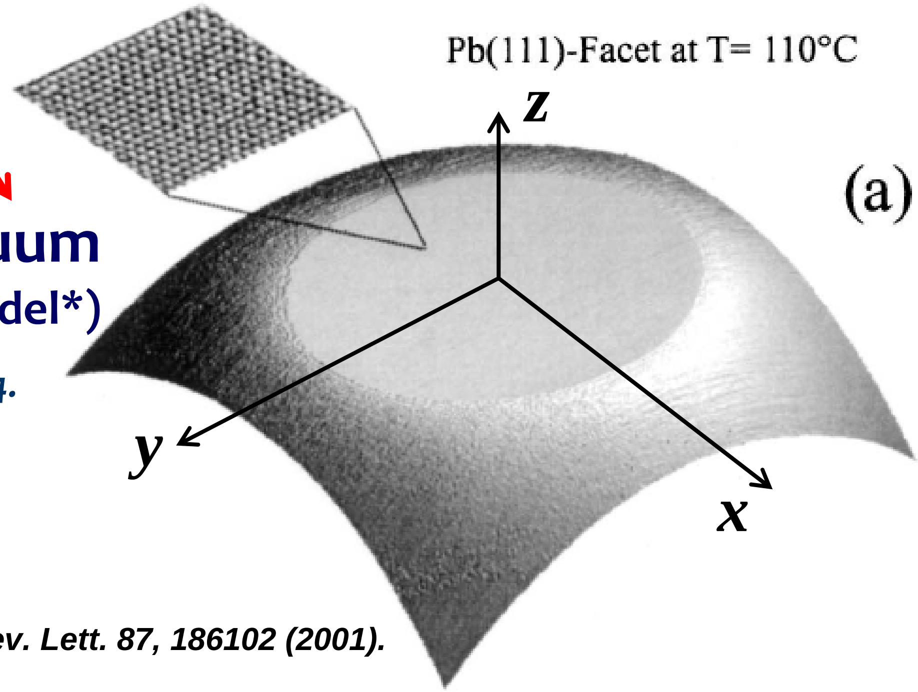


M. Uwaha: “Dynamics and pattern formation in crystal growth”

Step
(2D continuum step dynamics model*)



Continuum
(3D continuum partial differential equation model*)



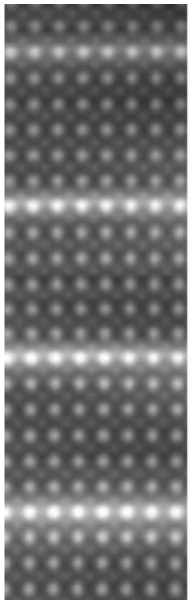
** J. W. Evans, ISSCG-14.*

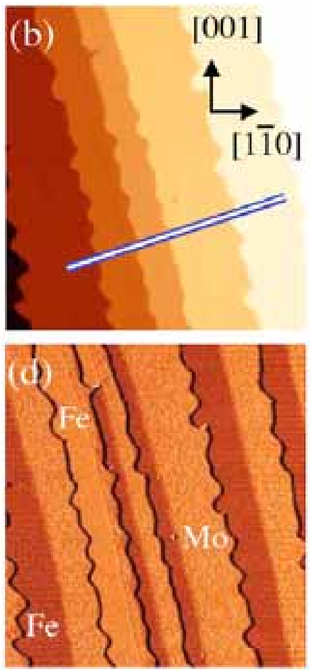
K. Thurmer et al., Phys. Rev. Lett. 87, 186102 (2001).

ステップダイナミクス

基礎科学　応用技術

✓新物質創製





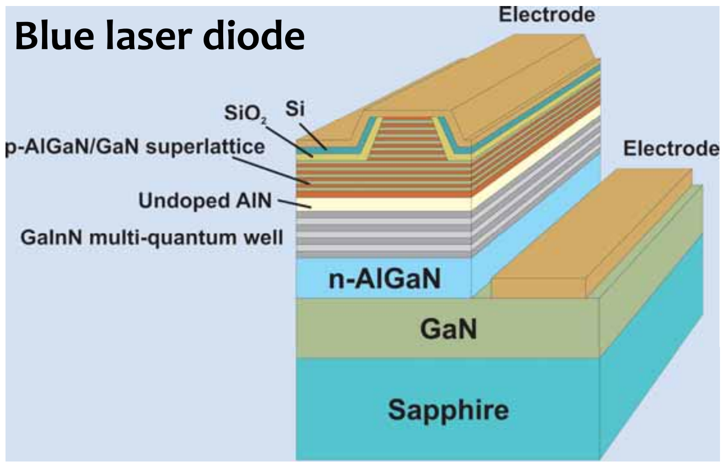
LaTiO3 layers (bright)
spaced by SrTiO3 layers

Template for magnetic nanowire

A. Ohtomo et al., Nature 419, 378 (2002).

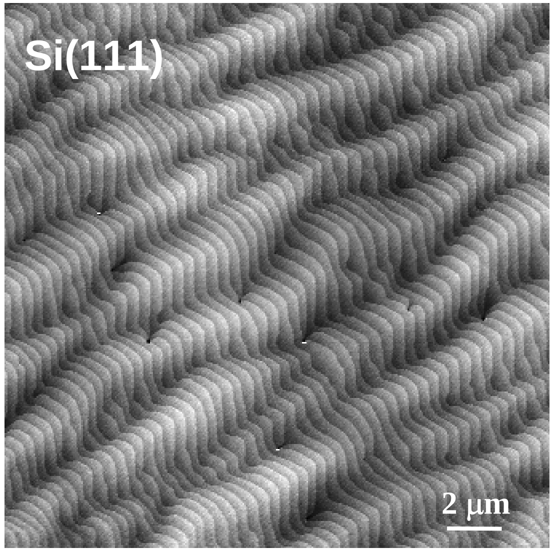
J. Prokop et al., PRB 73, 014428 (2002).

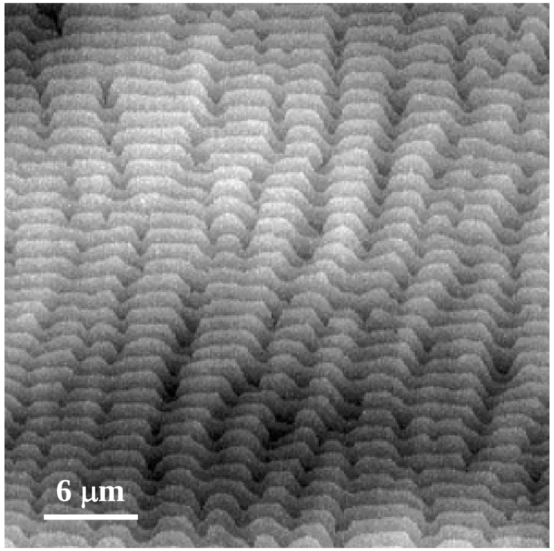
✓高機能デバイス

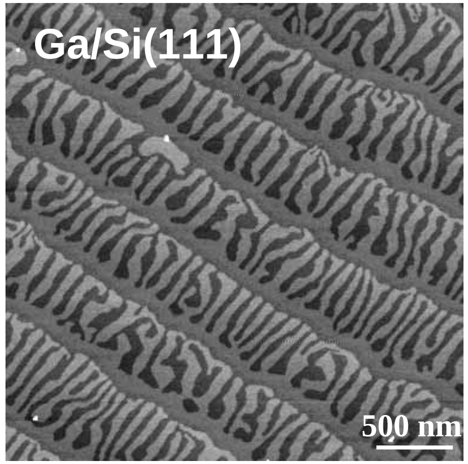


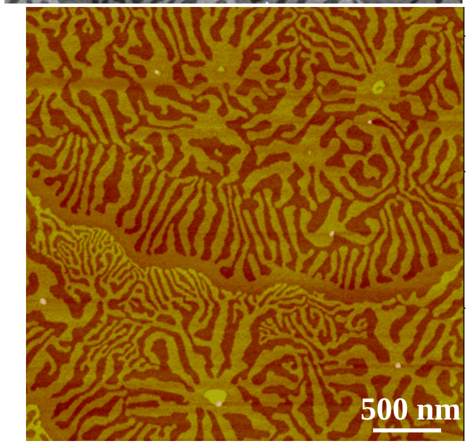
www.semiconductor-today.com/features/Semiconductor%20Today%20-%20The%20wide%20blue%20yonder.pdf

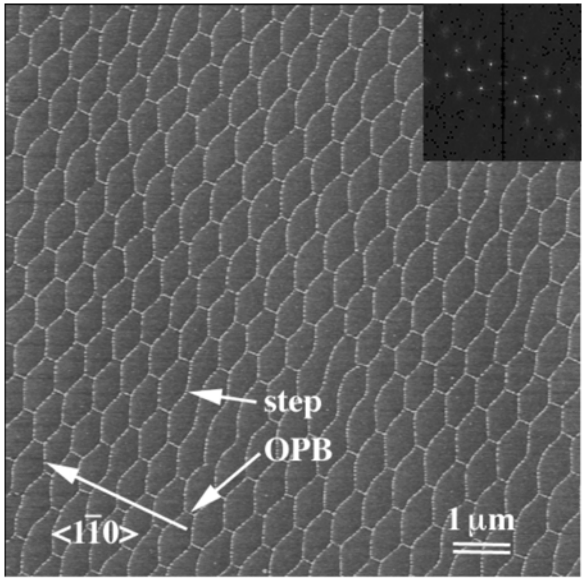
Step structure: my work











内容

(1) ステップの揺らぎ

(2) ステップの蛇行

(3) ステップバンチング

References

上羽牧夫：結晶成長のダイナミクスとパターン形成（培風館、2008）

上羽牧夫：結晶成長のしくみを探る（共立出版、2002）

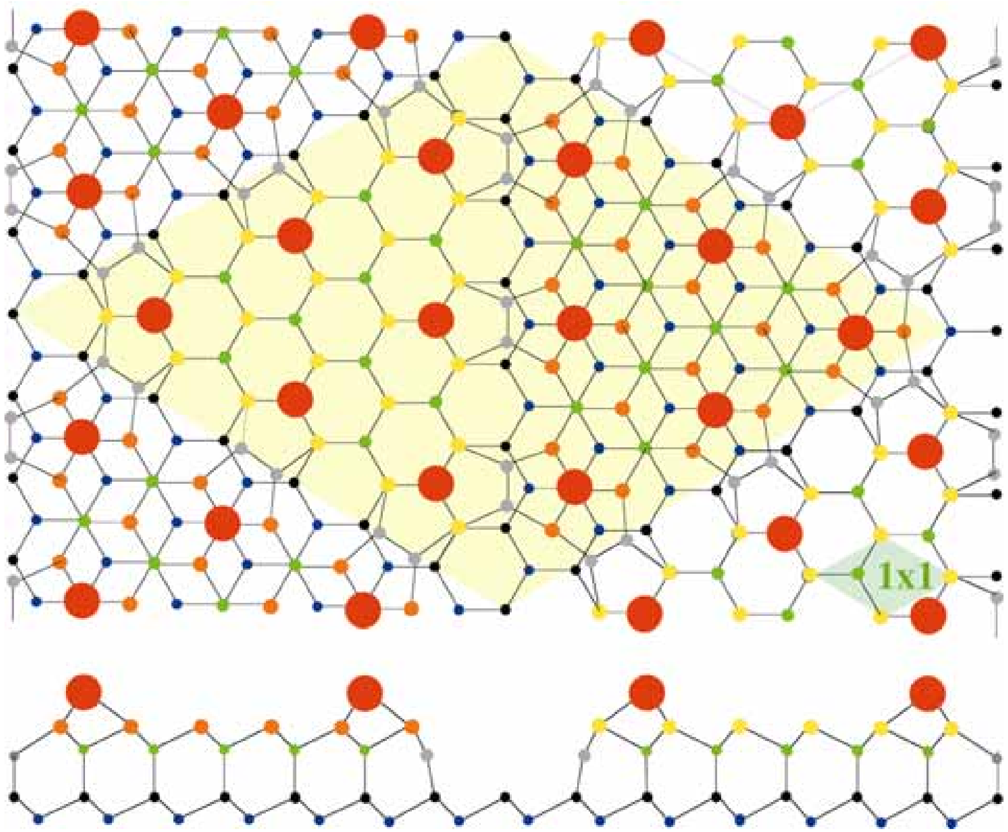
Yukio Saito, "Statistical Physics of Crystal Growth," (World Scientific, 1996)

C. Misbah, O. Pierre-Louis, and Y. Saito, "Crystal surfaces in and out of equilibrium: A modern view," Review of Modern Physics 82, 981 (2010).

表面構造

Surface reconstruction on Si(111)

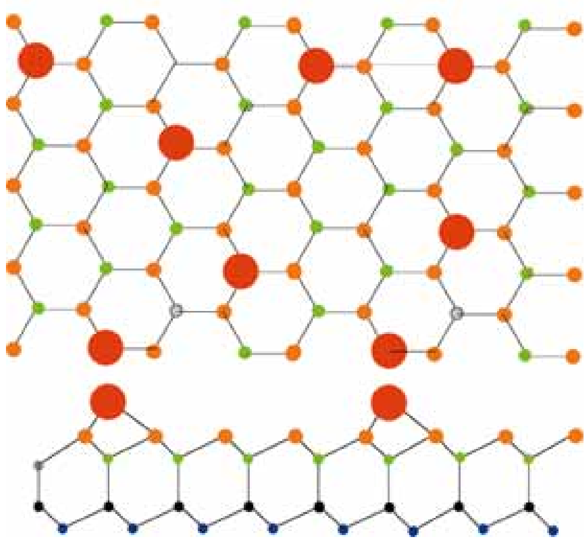
Si(111)-7×7

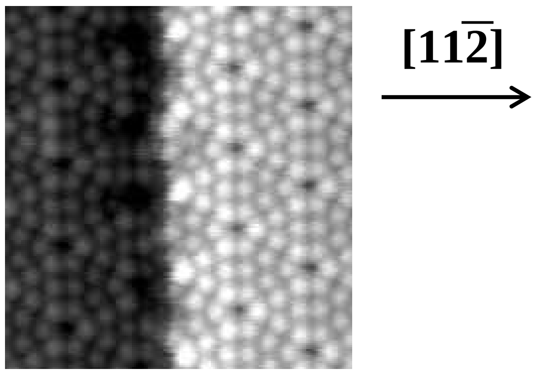


Dimer-Adatom-Stacking fault (DAS) model

K. Takayanagi et al., J. Vac. Sci. Technol. A 3, 1502 (1985).

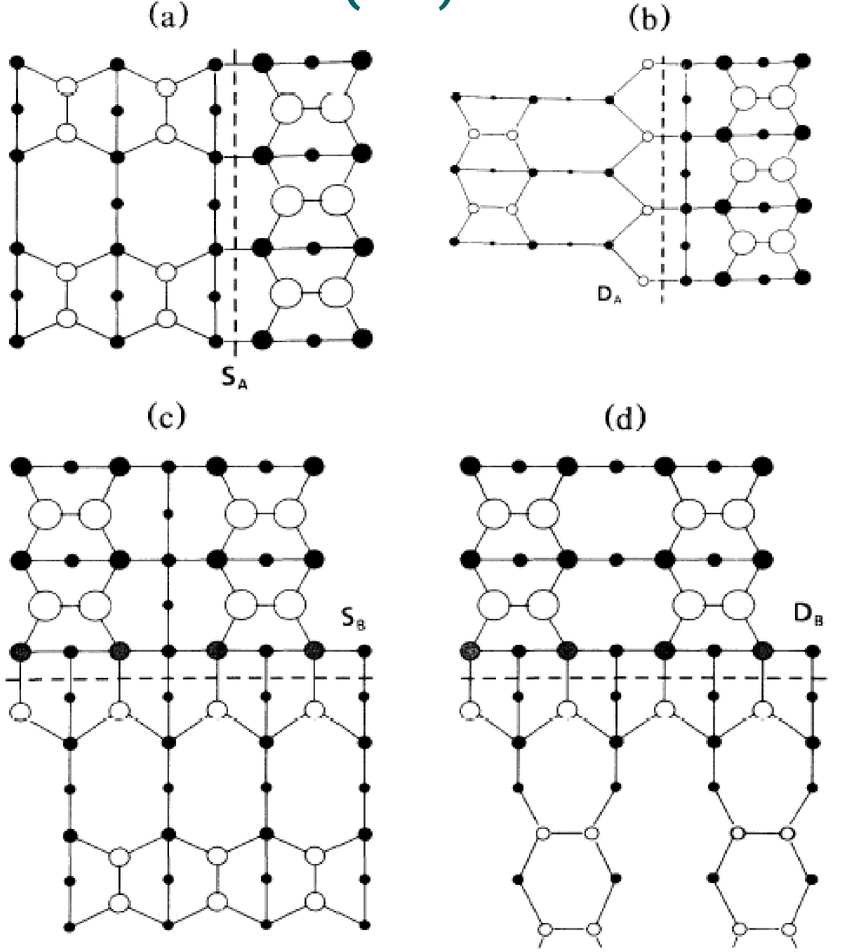
Si(111)-"1×1"





Surface reconstruction on Si(001)

Si(001)-2×1



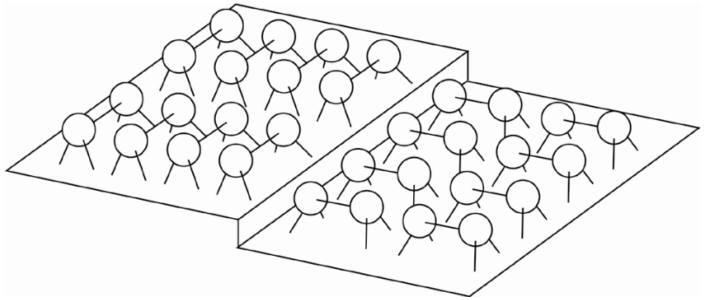
$\lambda(S_A) \simeq (0.01 \pm 0.01\ \mathrm{eV})/a,$

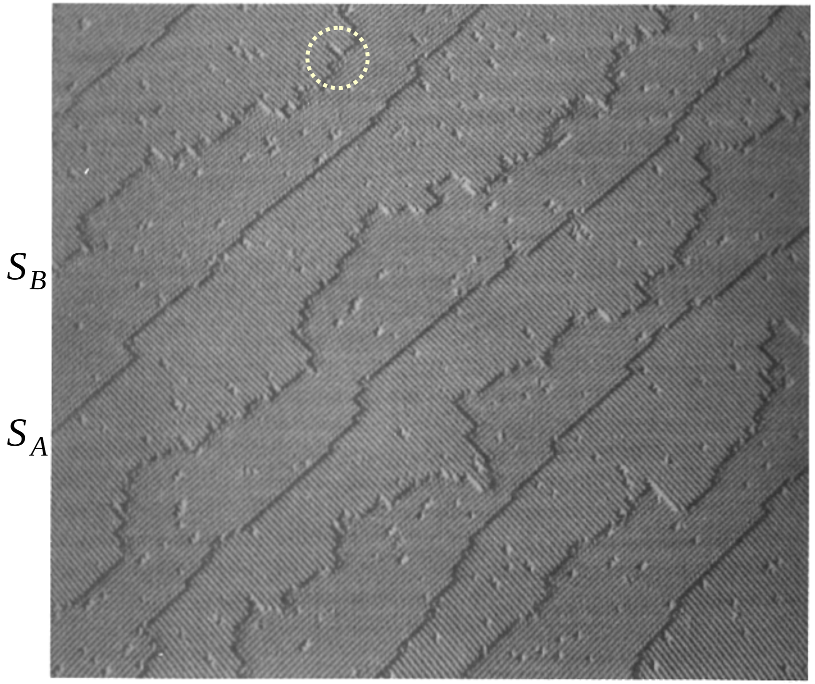
$\lambda(S_B) \simeq (0.15 \pm 0.03\ \mathrm{eV})/a,$

$\lambda(D_A) \simeq (0.54 \pm 0.10\ \mathrm{eV})/a,$

$\lambda(D_B) \simeq (0.05 \pm 0.02\ \mathrm{eV})/a.$

D. J. Chadi, Phys. Rev. Lett. 59, 1691 (1987).



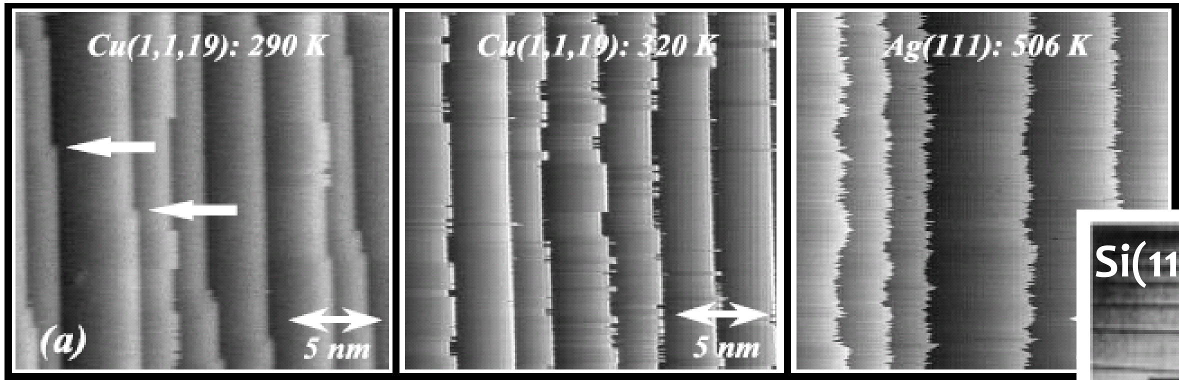


A. Pimpinelli and J. Villain, "Physics of Crystal Growth", p. 2, (Cambridge University Press, 1998)

観察装置

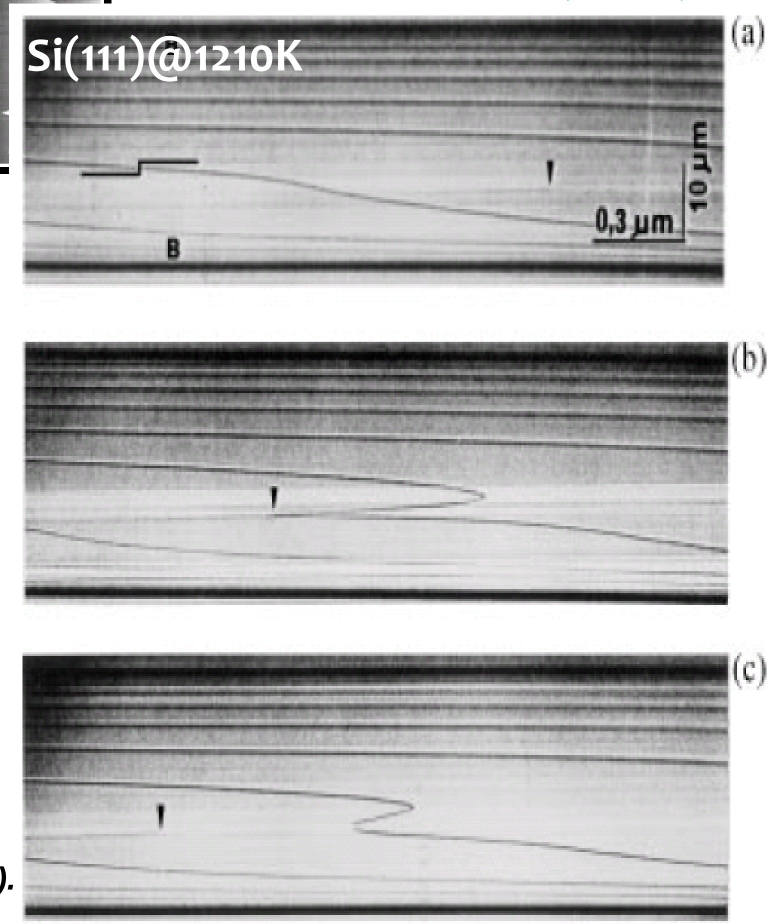
ステップのその場観察

走査トンネル顕微鏡(STM)

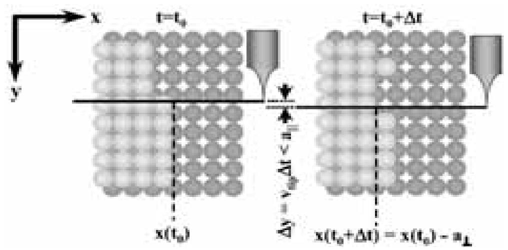


M. Giesen, Prog. Surf. Sci. 68, 1 (2001).

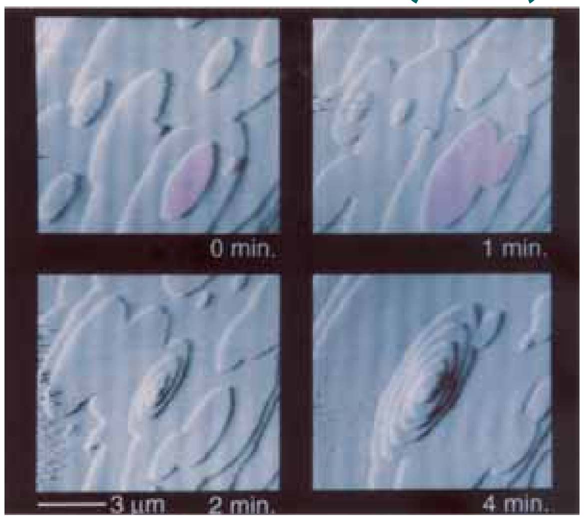
反射電子顕微鏡(REM)



A. V. Latyshev et al., PRL76, 94 (1996).

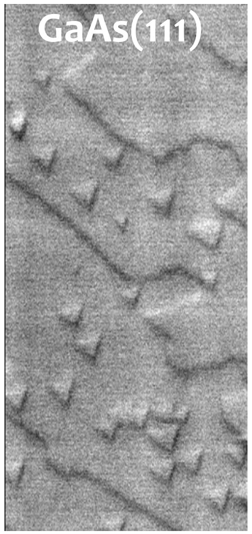


原子間力顕微鏡(AFM)



Lysozyme crystal

R. Lal et al., Am. J. Phios. 266, C1-& (1994).

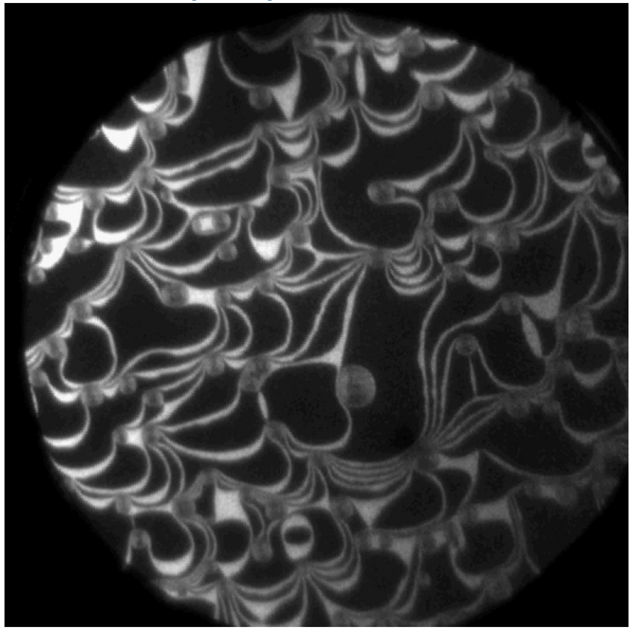


走査電子顕微鏡(SEM)

低エネルギー電子顕微鏡(LEEM)

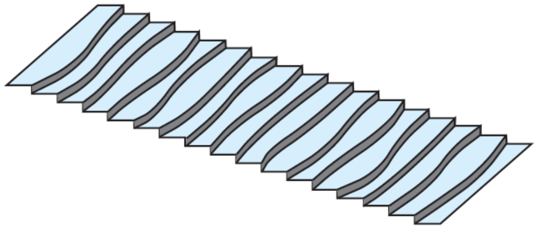
表面構造変化の動的観察例

Si(001)表面ステップ

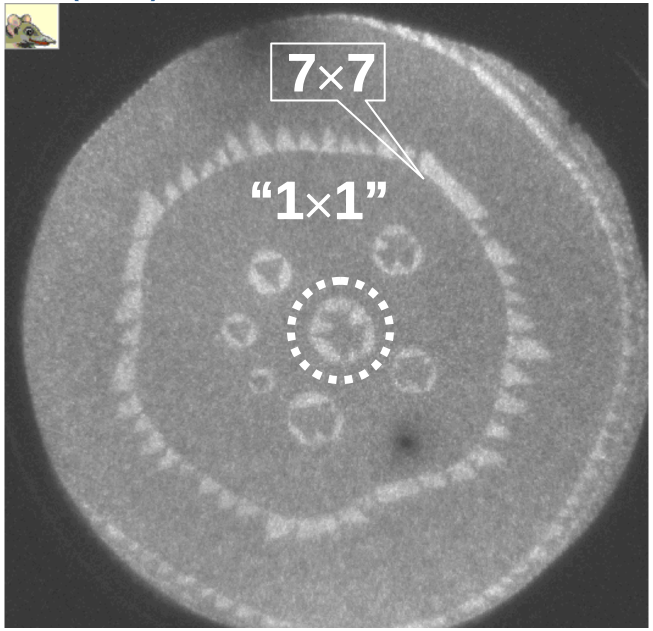


1 μm

20倍速

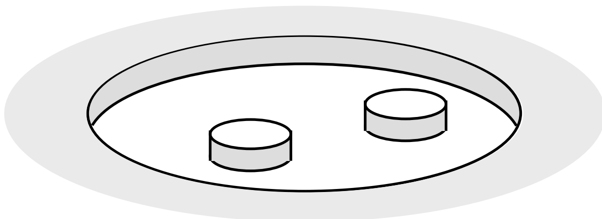


Si(111)表面上の二次元島の消失

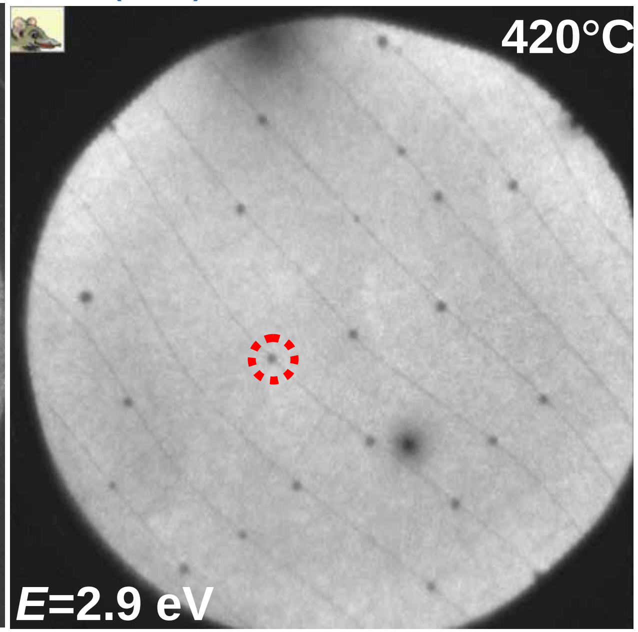


1 μm

5倍速

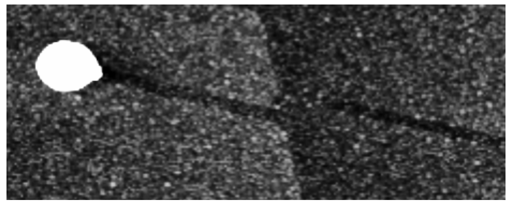


Si(111)表面上のAuナノ粒子

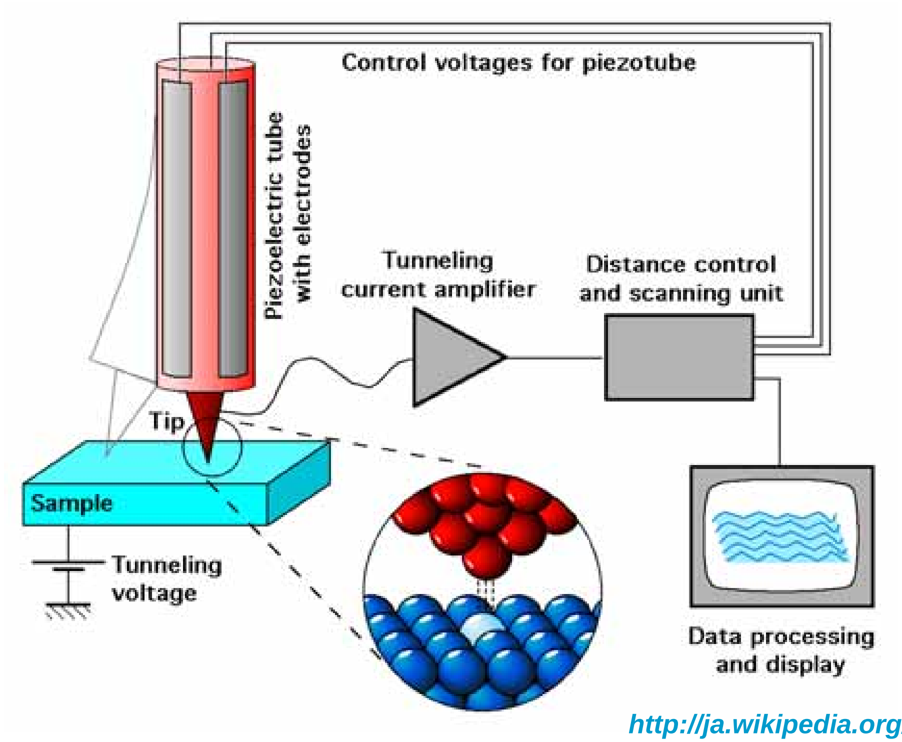


視野径4.4 μm

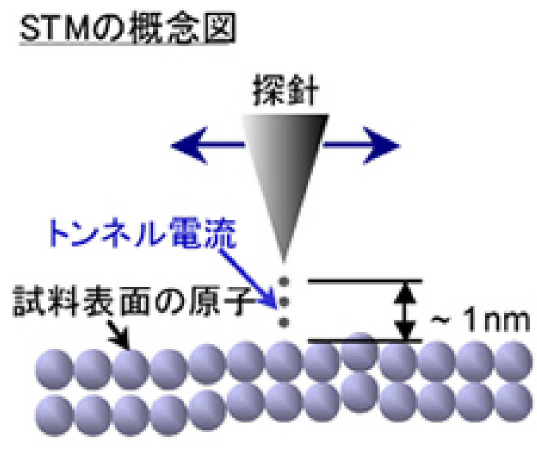
10倍速

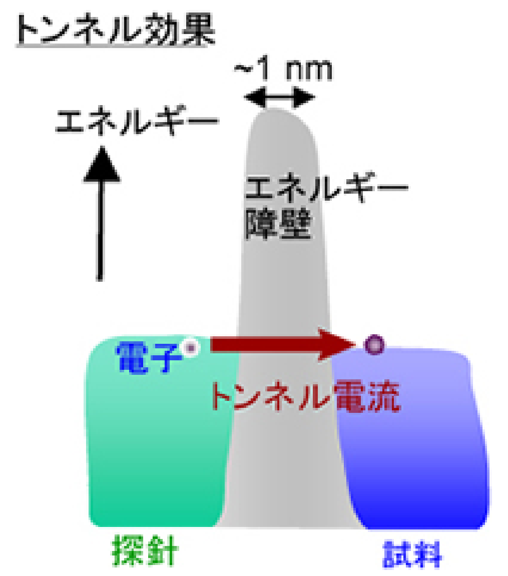


走査トンネル顕微鏡(STM)



http://ja.wikipedia.org/wiki/





$$I = \frac{4\pi e}{\hbar}|T|^2\int_{-\infty}^{\infty}\rho_t(E)\rho_s(E+eV)[f(E)-f(E+eV)]dE$$

$$|T|^2 \propto \exp\left(-2z\sqrt{2m\phi/\hbar^2}\right)$$

$$\frac{dI}{dV} \propto \rho_s(eV)$$

http://kelvin.phys.s.u-tokyo.ac.jp/fukuyama_lab/japanese/research/STM.html

原子間力顕微鏡(AFM)

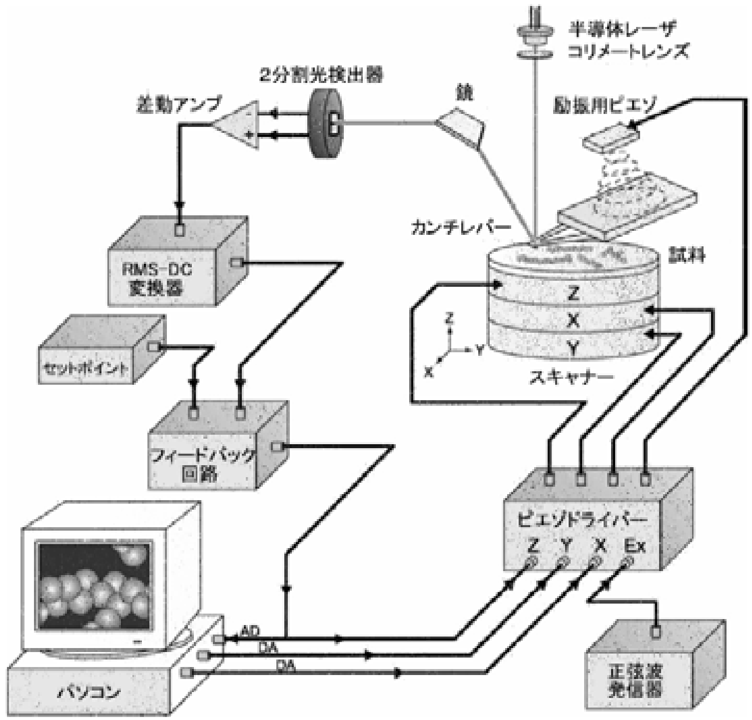
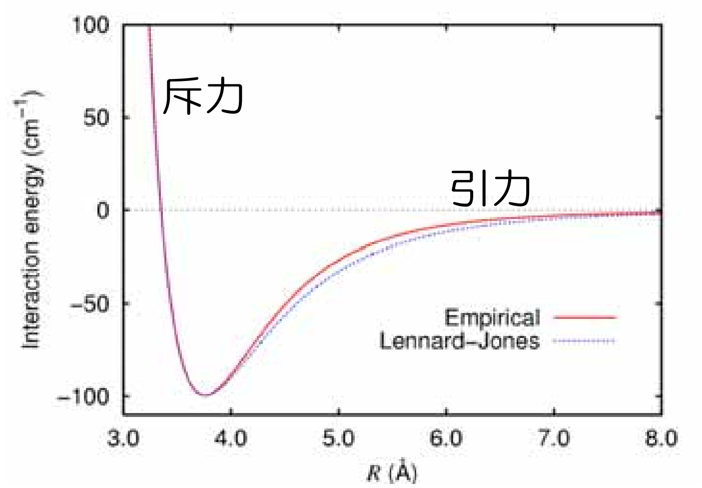


図 1　通常の AFM 装置の概略図

安藤敏夫、古寺哲幸、制御と計測 45(2), 1 (2005).



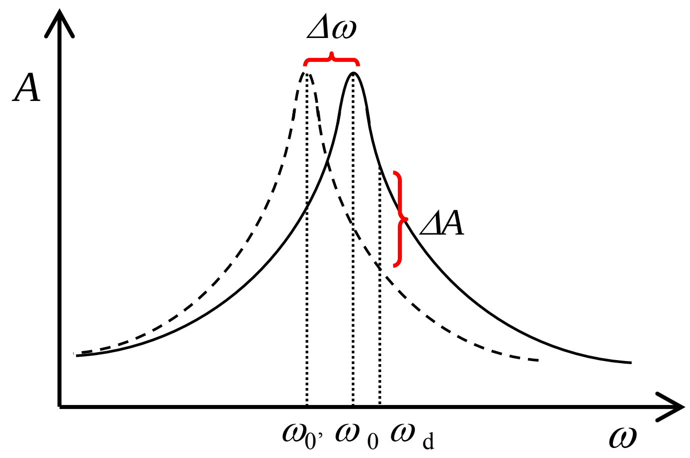
http://ja.wikipedia.org/wiki/

$$m\ddot{z} + (m\omega_0/Q)\,\dot{z} + m\omega_0^2 z = F_0 \cos(\omega_d t)$$

$$z(t) = A_0 \cos(\omega_d t + \vartheta_0)$$

$$A_0 = \frac{F_0/m}{\sqrt{(\omega_0^2 - \omega_d^2)^2 + (\omega_0\omega_d/Q)^2}}$$

$$\vartheta_0 = \tan^{-1}\left(\frac{\omega_0\omega_d}{Q(\omega_0^2 - \omega_d^2)}\right)$$

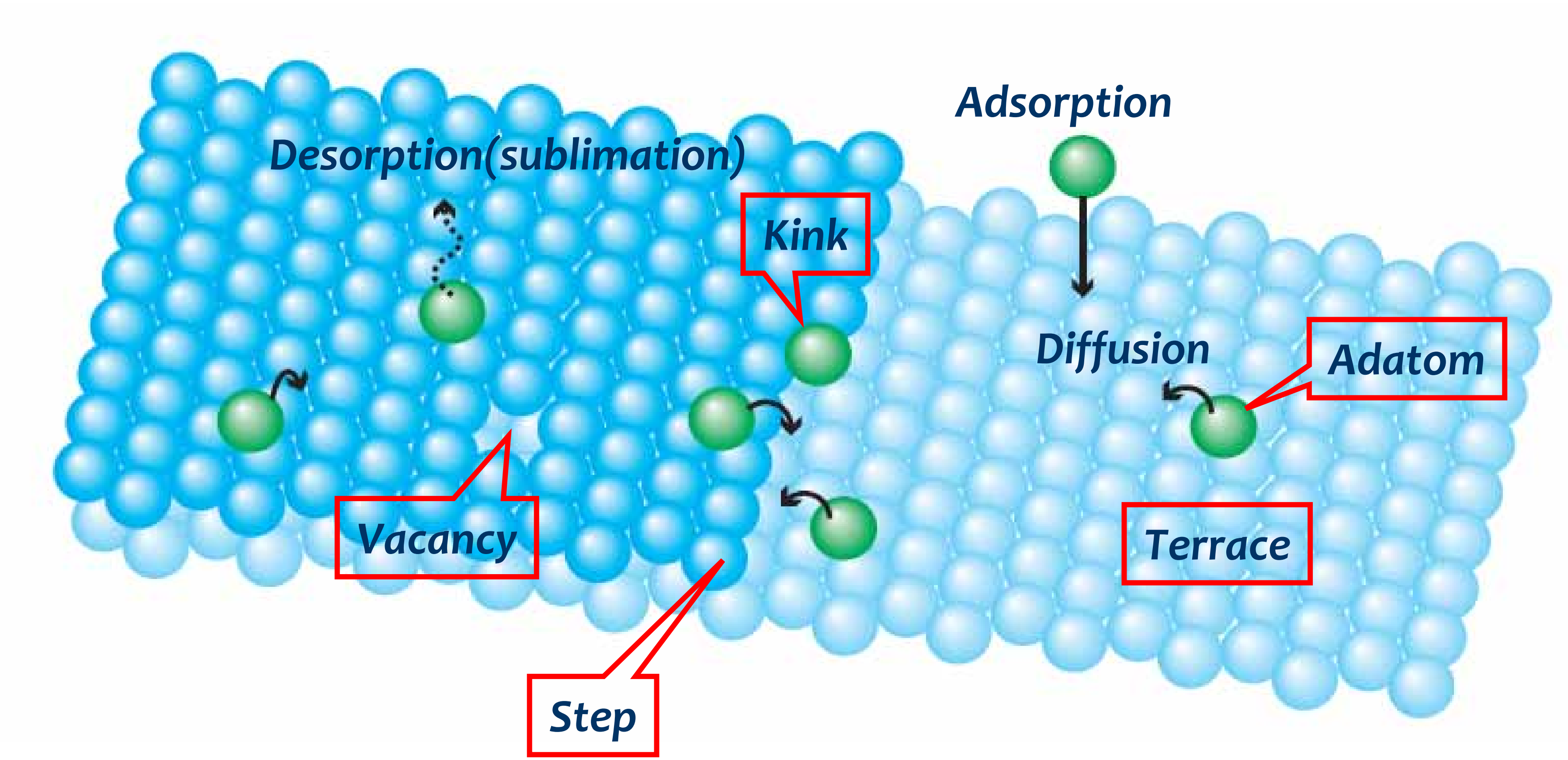


位相計測

端針・試料間相互作用力
端針・試料間の粘性抵抗係数

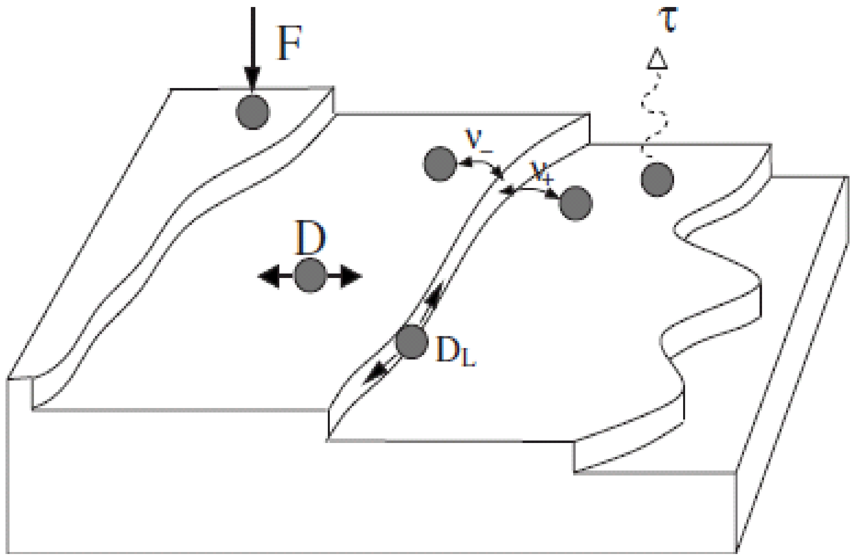
拡散方程式

結晶表面の模式図


Adsorption
Desorption(sublimation)
Kink
Diffusion
Adatom
Vacancy
Terrace
Step

拡散方程式

拡散方程式:
$$\frac{\partial c(x,y,t)}{\partial t} = \underbrace{D\nabla^2 c(x,y,t)}_{\text{拡散}} + \underbrace{F}_{\text{蒸着}} - \underbrace{\frac{1}{\tau}c(x,y,t)}_{\text{脱離}} - \underbrace{\vec{v}_{\text{drift}} \cdot \nabla c}_{\text{ドリフト}}$$



ドリフト流

外部電場中の帯電アドアトムのドリフト: $v_{\text{drift}} = \dfrac{DZ}{k_B T} E$

$$J_{\text{drift}} = v_{\text{drift}} c = \mu F c = -\mu c \frac{dU}{dx}, \quad J_{\text{diffusion}} = -D\frac{dc}{dx}$$

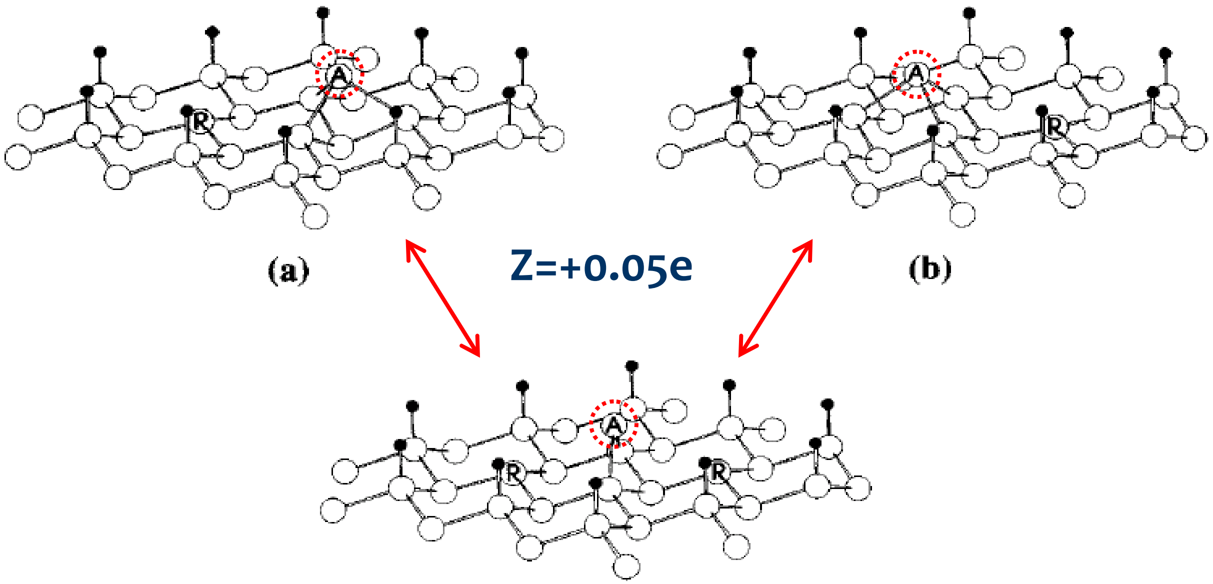
平衡分布

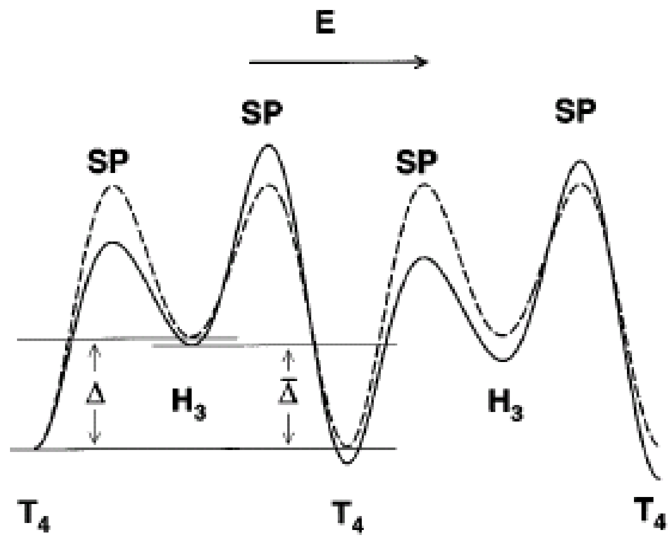
$$J_{\text{drift}} + J_{\text{diffusion}} = 0$$

$$c = Ae^{-U/kT} \longrightarrow \frac{dc}{dx} = -\frac{A}{kT}\frac{dU}{dx}e^{-U/kT} = -\frac{c}{kT}\frac{dU}{dx}$$

$$\longrightarrow \mu = \frac{D}{k_B T}$$

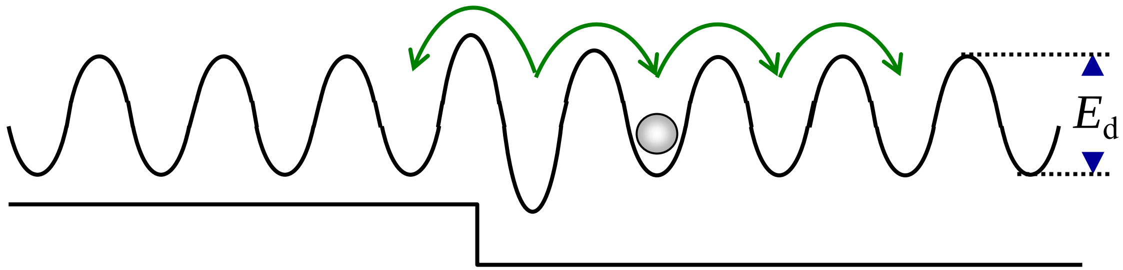
Diffusion of Si adatom





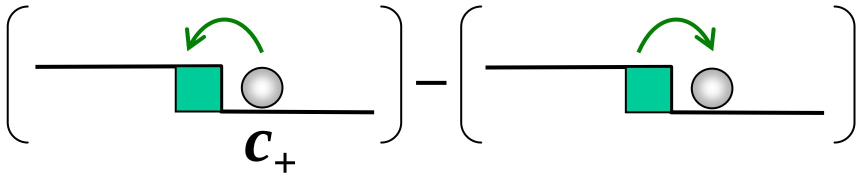
D. Kandel and E. Kaxiras, Phys. Rev. Lett. 76, 1114 (1996).

ステップでの境界条件



下段テラスからの拡散流によるステップ前進速度： $v_+ = \Omega D \left. \dfrac{\partial c(x,y,t)}{\partial x} \right|_+$

ステップで結晶化する速度：



両者がバランスされているときアドアトム濃度= c_{eq}^0

$$v_+ = \Omega K_+ \left(c_+ - c_{eq}^0 \right)$$

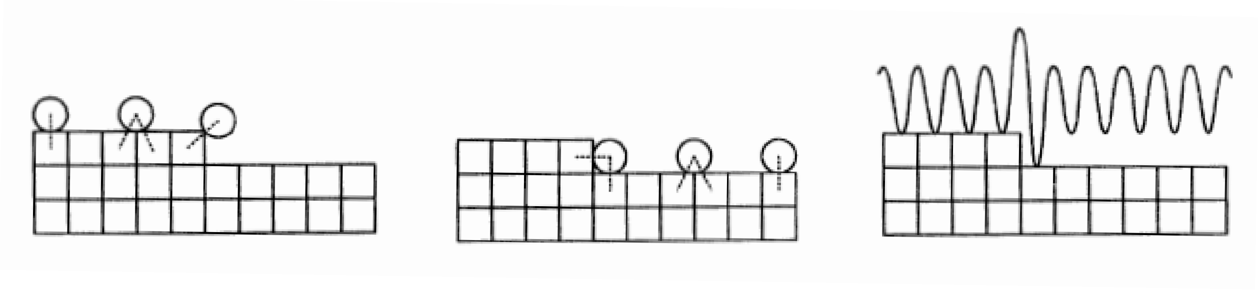
境界条件：$D \left. \dfrac{\partial c(x,y,t)}{\partial x} \right|_+ = K_+ \left(c_+ - c_{eq} \right)$

$$\pm D \vec{n} \cdot \nabla c \big|_\pm = K_\pm \left(c_\pm - c_{eq} \right)$$

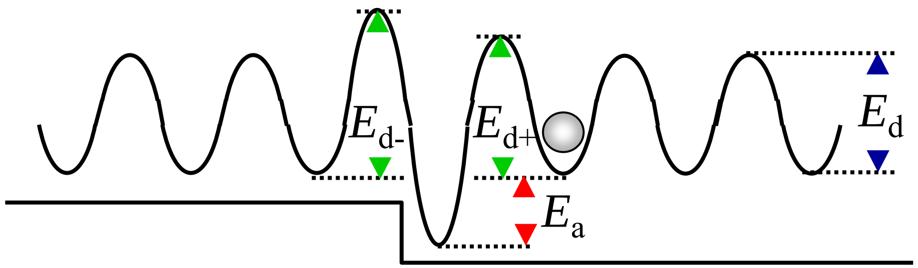
カイネティク係数

エーリッヒ-シュワーベル効果

ステップへの原子の取り込み速度に対する上下の非対称性



R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. 44 (1966) 3682.
G. Ehrlich and F. G. Hudda, J. Chem. Phys. 44 (1966) 1039.

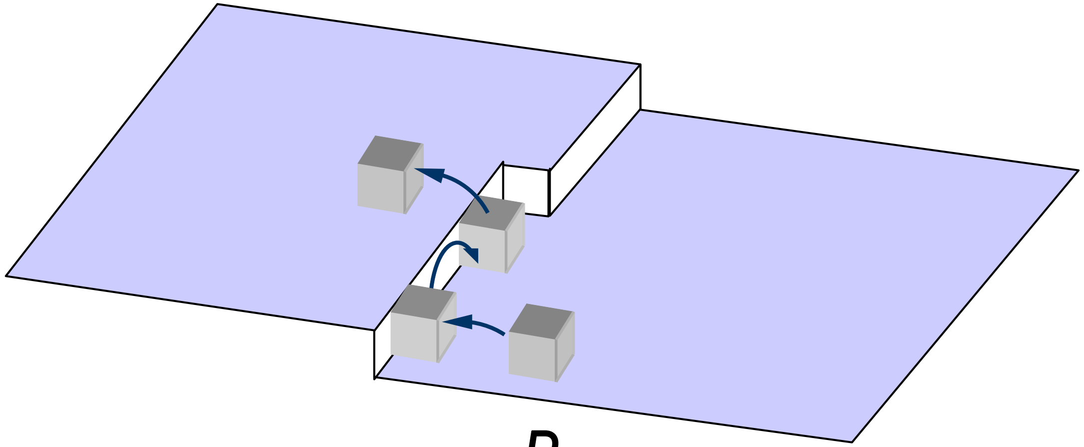


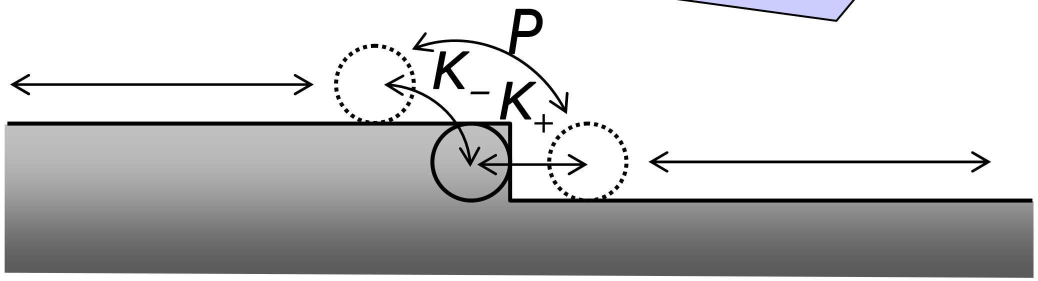
$$v_+ = c_+ \exp\left(-\frac{E_{d+}}{k_B T}\right) - \exp\left(-\frac{E_{d+}+E_a}{k_B T}\right) = K_+\left(c_+ - c_{eq}^0\right)$$

$$c_{eq}^0 = \exp\left(-\frac{E_a}{k_B T}\right) \qquad K_+ = \exp\left(-\frac{E_{d+}}{k_B T}\right)$$

ステップ透過率(permeability)

原子が結晶に取り込まれること無く通過





$$\begin{cases} j_- = j_{s-} + j_p \\ j_+ = -j_{s+} + j_p \end{cases} \qquad \begin{cases} j_{s\pm} = K_\pm\left(c(x_\pm) - c_{eq}\right) \\ j_p = P\left(c(x_-) - c(x_+)\right) \end{cases}$$

ステップ透過率

$$\vec{n}\cdot\left(\nabla c|_+ - \vec{v}_{drift} c_+\right) = K_+\left(c_+ - c_{eq}\right) + P\left(c_+ - c_-\right)$$

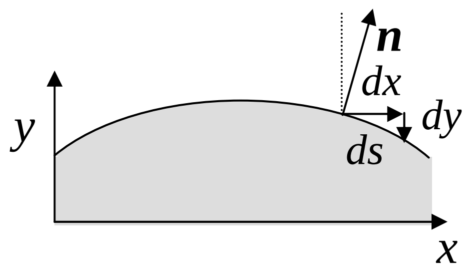
$$-\vec{n}\cdot\left(\nabla c|_- - \vec{v}_{drift} c_-\right) = K_-\left(c_- - c_{eq}\right) + P\left(c_- - c_+\right)$$

ギブス-トムソン効果

直線ステップの平衡アドアトム濃度 c_{eq}^0

湾曲したステップの平衡アドアトム濃度 c_{eq}

c_{eq} の c_{eq}^0 からの変化量は、原子１個が固化するときにどれだけ余分の仕事が必要になるかで決まる。仕事＝力$F\times\Omega$



$$c_{eq} = c_{eq}^0 \exp\left(-\frac{\Omega F}{k_B T}\right) \approx c_{eq}^0\left(1-\frac{\Omega F}{k_B T}\right)$$

$$F_{step} \approx \int\left(\beta(0)+\frac{1}{2}\tilde{\beta}(0)p^2\right)dx$$ 、ここで $p=\frac{\partial y}{\partial x}$

スティフネス

$$\delta F_{step} \approx \int\left(\tilde{\beta}(0)p\,\delta p\right)dx = \int\left(-\tilde{\beta}(0)\frac{\partial^2 y}{\partial x^2}\delta y\right)dx$$

$$\Rightarrow F = -\frac{d\left(\delta F_{step}\right)/dx}{\delta y} = \tilde{\beta}(0)\frac{\partial^2 y}{\partial x^2} \approx -\frac{\tilde{\beta}(0)}{R}$$

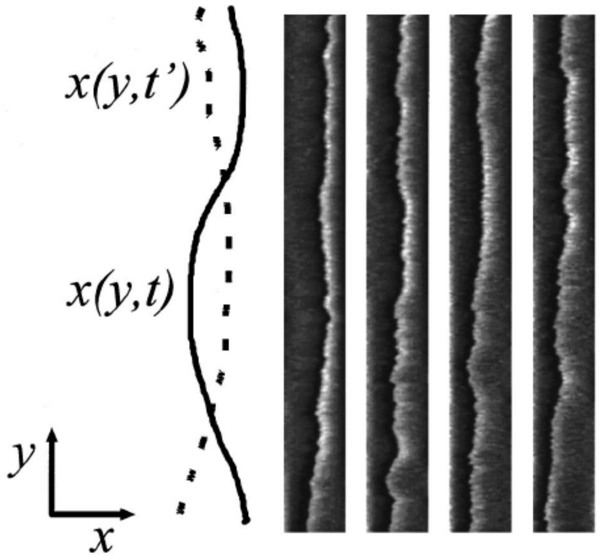
曲率半径

$$c_{eq} = c_{eq}^0 \exp\left(\frac{\Omega\tilde{\beta}}{k_B T R}\right) \approx c_{eq}^0\left(1+\frac{\Omega\tilde{\beta}}{k_B T R}\right)$$

ステップの熱平衡揺らぎ

ステップの揺らぎ(TSKモデル)

ステップの揺らぎの微視的に理解する。

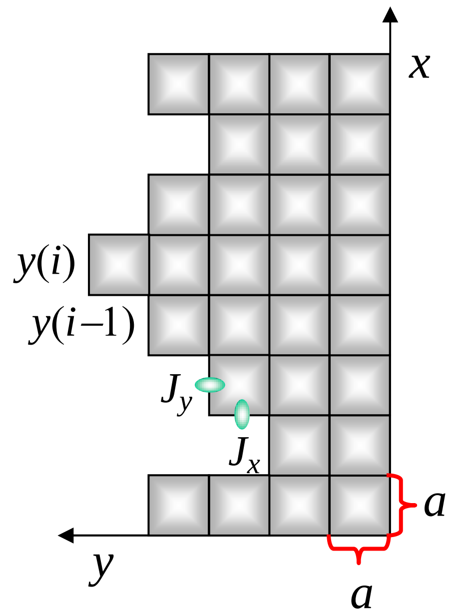


Four 60nm×3500nm STM images of Ag(110), each accumulated in 18 s of a fluctuating isolated step are shown at 30-min intervals.

W.W. Pai et al., Phys. Rev. B53, 15991 (1996).

テラス-ステップ-キンク(TSK)モデル

J_x ：キンクエネルギー



TSKモデルで、x=0からL=Naまで走るステップのエネルギーが

$$E = NJ_y + J_x \sum_{i=1}^{N} |n_i|$$

と書けるとき、xの異なる点でのキンクには相関が無いとして、ステップの自由エネルギー密度βを求める。

ステップの揺らぎ(TSKモデル)

分配関数 $$Z=\left[\sum_{n=-\infty}^{+\infty}\exp\left(-\frac{J_y+J_x|n|}{k_{\mathrm{B}}T}\right)\right]^N=\exp\left(-\frac{NJ_y}{k_{\mathrm{B}}T}\right)\left(\coth\frac{J_x}{2k_{\mathrm{B}}T}\right)^N$$

$$F=-k_{\mathrm{B}}T\ln Z=NJ_y-Nk_{\mathrm{B}}T\ln\left(\coth\frac{J_x}{2k_{\mathrm{B}}T}\right)$$

ステップ自由エネルギー密度 $$\beta=-\frac{F}{Na}=\frac{1}{a}\left(J_y-k_BT\ln\left(\coth\frac{J_x}{2k_BT}\right)\right)$$

$J_x=J_y=J$ のとき、$T_R=J/\left[k_B\ln\left(\sqrt{2}+1\right)\right]$ において、$\beta=0$ （表面ラフニング）

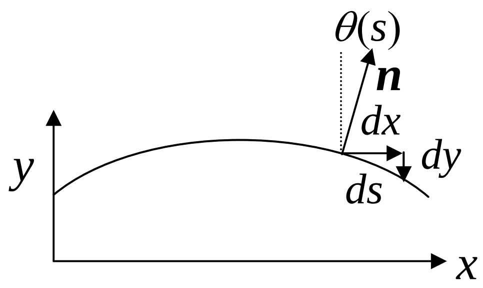
平均キンク数 $$\langle n^2\rangle=\frac{1}{2\sinh\dfrac{J_x}{2k_BT}}$$

ステップ位置の差の相関関数 $$G(x)\equiv\left\langle\left(y(x)-y(0)\right)^2\right\rangle=\left\langle\left(\sum_{j=1}^{N}an_j\right)^2\right\rangle$$

$$=a^2\left\langle\sum_{j=1}^{N}n_j^2\right\rangle=Na^2\langle n^2\rangle=ax\langle n^2\rangle$$

ステップの揺らぎ(連続体モデル)

揺らいでいるステップの全自由エネルギー



$$F_{step} = \int ds\beta(\theta(s)) = \int dx\beta(\theta(s))\sqrt{1+\left(\frac{dy}{dx}\right)^2}$$

$dy/dx = \tan\theta \approx \theta$ →

$$\approx \int\left(\beta(0)+\beta'(0)y'(x)+\frac{1}{2}\beta''(0)|y'(x)|^2\right)\left(1+\frac{1}{2}|y'(x)|^2\right)dx$$

$$\approx \beta(0)L+\frac{1}{2}(\beta(0)+\beta''(0))\int|y'(x)|^2dx = \beta(0)L+\frac{1}{2}\tilde{\beta}(0)\int|y'(x)|^2dx$$

スティフネス　$\tilde{\beta}(0)=\beta(0)+\beta''(0)$

ステップの揺らぎ(連続体モデル)

フーリエ分解

$$y(x) = \sum_k y_k e^{ikx}$$

周期境界条件: $y(x) = y(x+L)$

➡ $k = 2\pi m/L \quad \text{with} \quad m = \pm 1, \pm 2, \ldots., \pm \infty$

$$F = \frac{1}{2}\tilde{\beta}\int |y'(x)|^2 dx = \sum_k \frac{1}{2}\tilde{\beta}k^2 |y_k|^2$$

等分配則： $\left\langle \frac{1}{2}L\tilde{\beta}k^2 |y_k|^2 \right\rangle_{eq} = \sqrt{\frac{L\tilde{\beta}k^2}{2\pi k_B T}} \int \frac{1}{2}L\tilde{\beta}k^2 |y_k|^2 e^{-\frac{L\tilde{\beta}k^2}{2\pi k_B T}y_k^2} dy_k = \frac{1}{2}k_B T$

$$\left\langle |y_k|^2 \right\rangle_{eq} = \frac{k_B T}{L\tilde{\beta}k^2}$$

ステップの揺らぎ(連続体モデル)

相関関数

$$G(x) \equiv \left\langle \left(y(x) - y(0) \right)^2 \right\rangle$$

$$= 2\sum_k \left\langle |y_k|^2 \right\rangle_{eq} \left(1 - \cos kx\right) = 4\sum_k \left\langle |y_k|^2 \right\rangle \sin^2\left(\frac{kx}{2}\right)$$

$$= 4\frac{k_B T}{L\tilde{\beta}} \int_{-\infty}^{\infty} \frac{\sin^2(kx/2)}{k^2} \frac{L dk}{2\pi} = \frac{k_B T}{\tilde{\beta}} x$$

ステップの揺らぎの幅は？

Periodic step with a length *L*

$$w_{eq}^2 \equiv \frac{1}{L}\int_0^L \left\langle \left(y(x) \right)^2 \right\rangle_{eq} dx = \sum_k \left\langle |y_k|^2 \right\rangle = \sum_m \frac{k_B T}{L\tilde{\beta}} \left(\frac{L}{2\pi m} \right)^2 = \frac{k_B T}{12\tilde{\beta}} L$$

Step with the both ends fixed

$$w_{eq}^2 = \frac{k_B T}{6\tilde{\beta}} L$$

ステップの揺らぎ

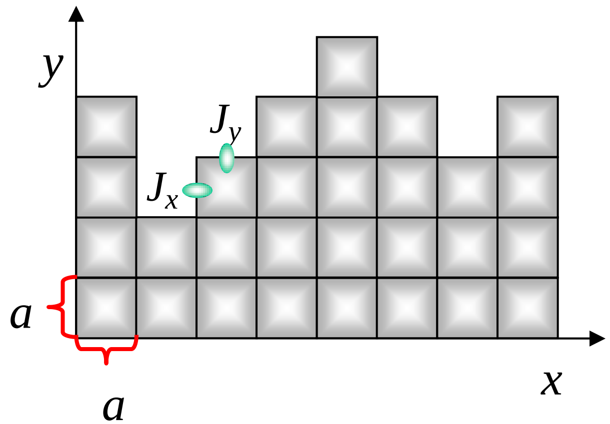
ステップスティフネスとキンクエネルギーの関係

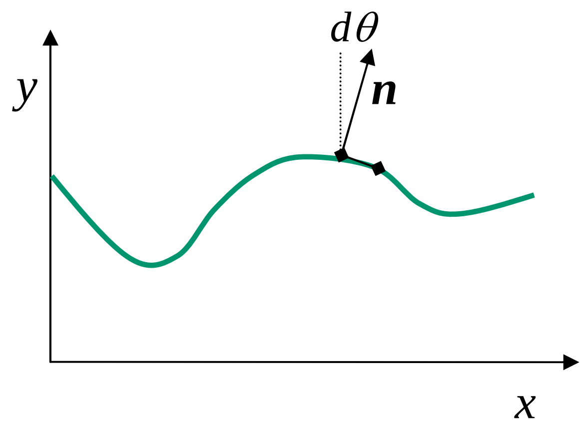
$$G(x) = ax\langle n^2 \rangle = \frac{k_B T}{\tilde{\beta}} x$$

$$\Rightarrow \tilde{\beta} = \frac{k_B T}{a\langle n^2 \rangle}$$

$$\langle n^2 \rangle = \frac{1}{2\sinh^2(J_x/2k_B T)}$$

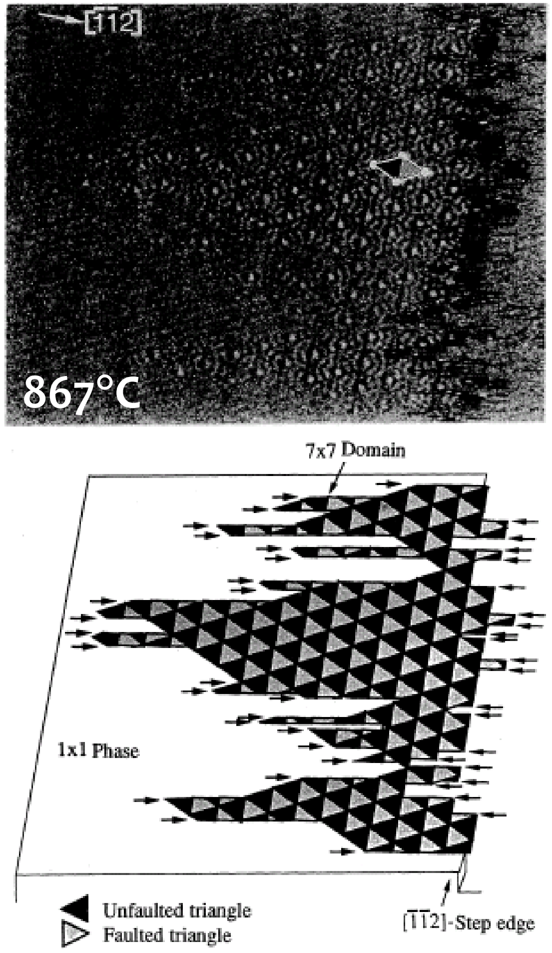
$$\tilde{\beta} = \frac{2k_B T}{a}\sinh^2(J_x/2k_B T)$$



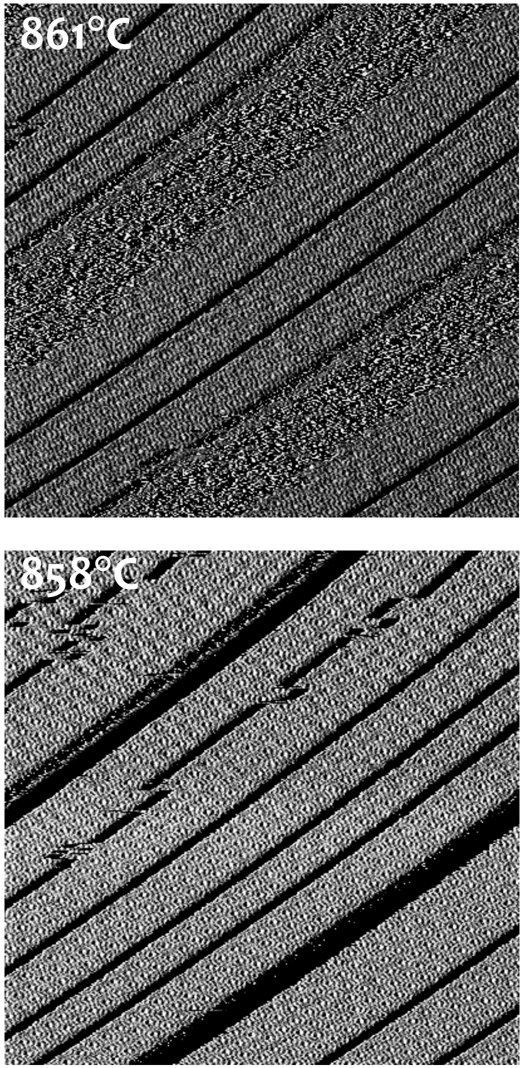


ステップの揺らぎ(実験)

Si(111)

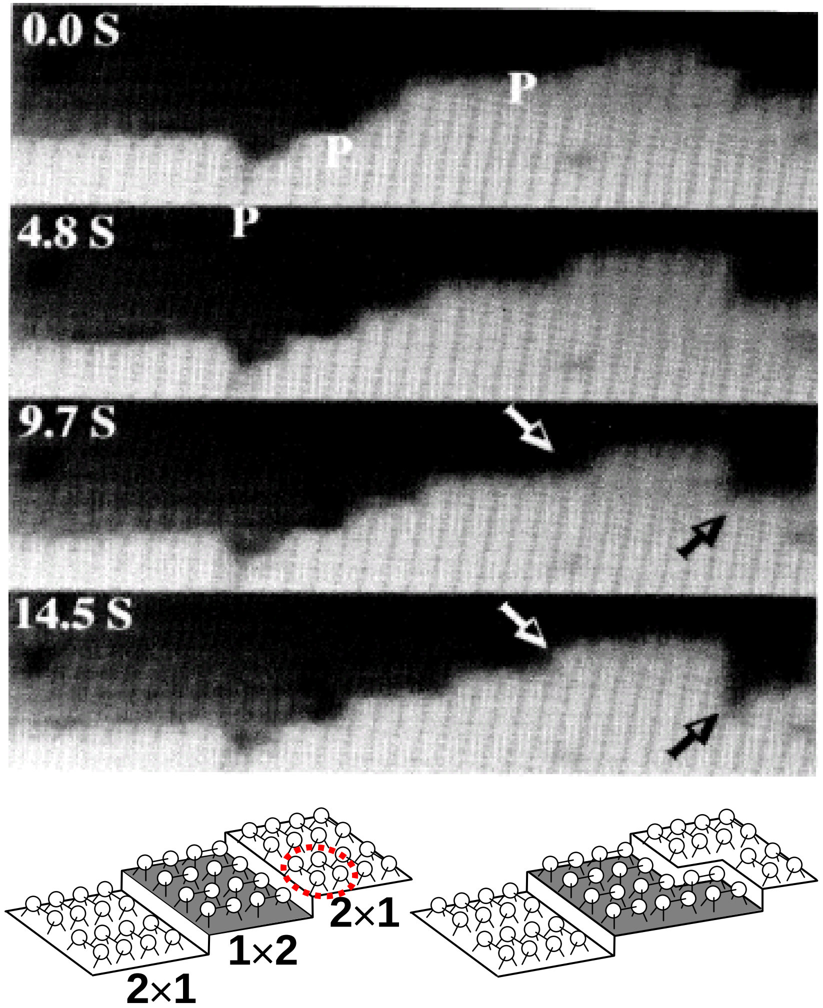


H. Tokumoto and H. Iwatsuki, Jpn. J. Appl. Phys. 32, 1368 (1993).



H. Hibino and T. Ogino, Phys. Rev. Lett. 72, 657 (1994).

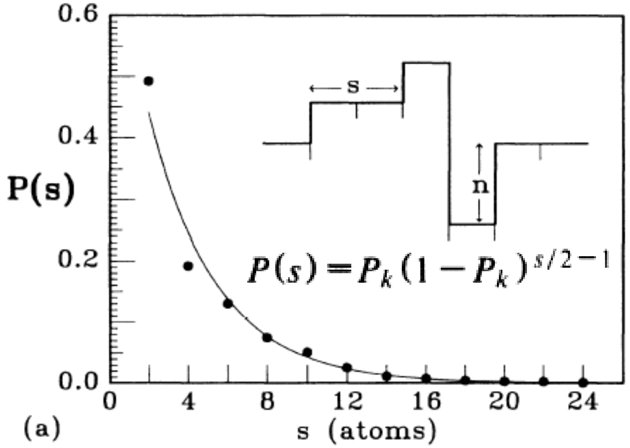
Si(001)

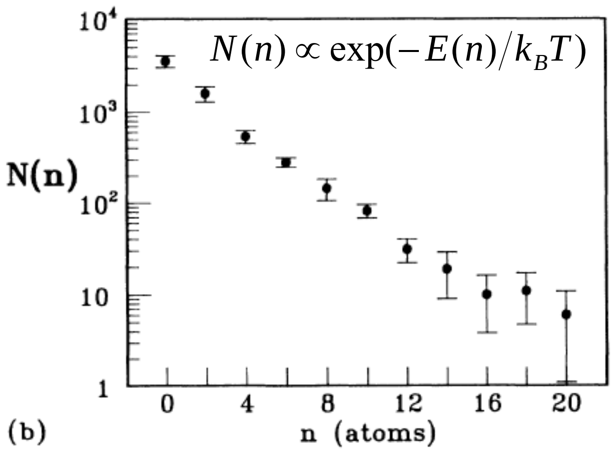


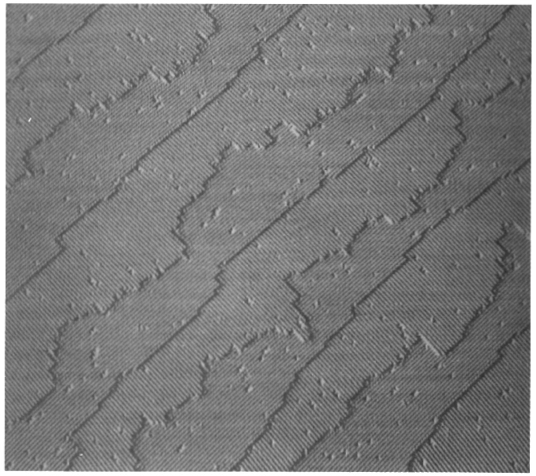
C. Pearson et al., Phys. Rev. Lett. 74, 2710 (1995)

キンクエネルギーの実測例

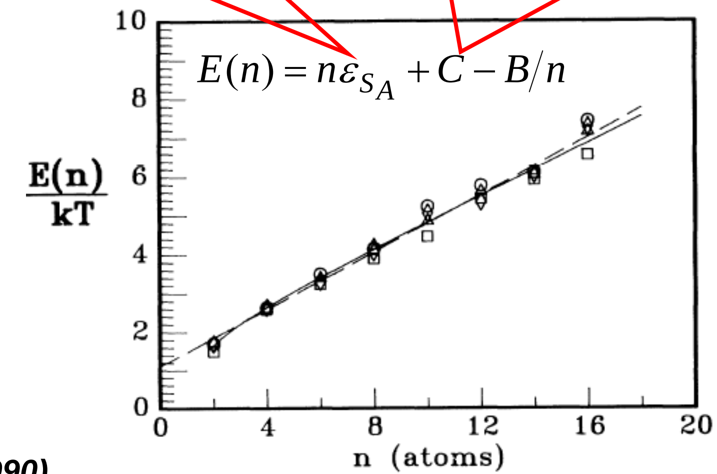
Si(001)-2×1表面







キンクエネルギー　キンクコーナーエネルギー



$\epsilon_{S_A} = 0.028 \pm 0.002$ eV/atom

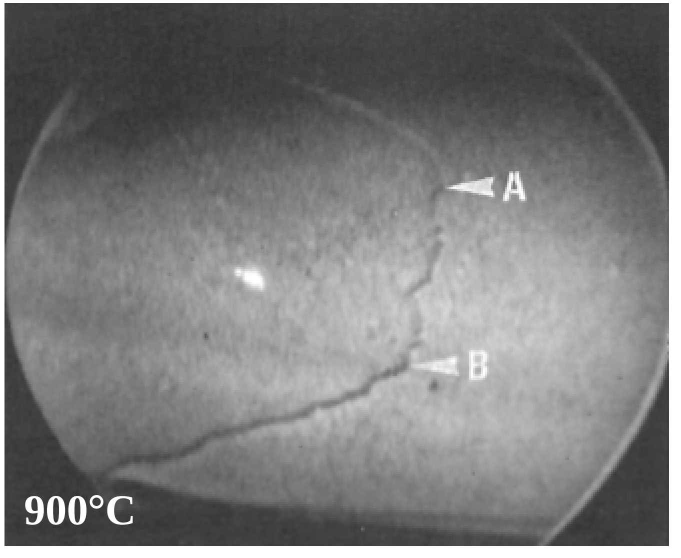
$\epsilon_{S_B} = 0.09 \pm 0.01$ eV/atom

B. S. Swartzentruber et al., Phys. Rev. Lett. 65, 1913 (1990).

スティフネスの実測例

Si(111)-”1×1”表面

反射電子顕微鏡(REM)像



C. Alfonso et al., Surf. Sci. 262, 371 (1992).

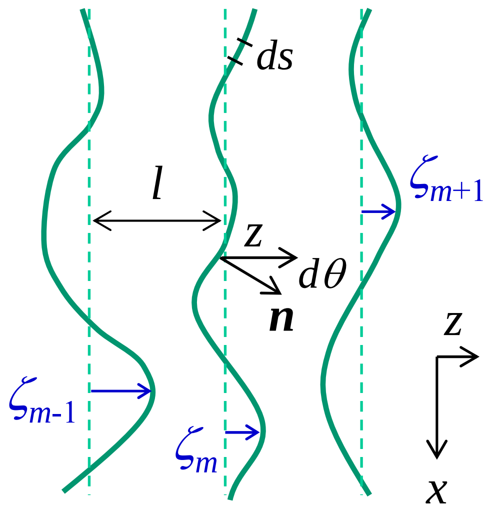
$$\left\langle (y(x))^2 \right\rangle = \frac{k_B T}{6\tilde{\beta}} L$$

$$\tilde{\beta} = 0.8 - 1.4\ \mathrm{eV/nm}$$

$$J_x \approx 0.25\ \mathrm{eV/bond}$$

P. Pimpinelli et al., Surf. Sci. 295, 143 (1993).

ステップ列のエネルギー

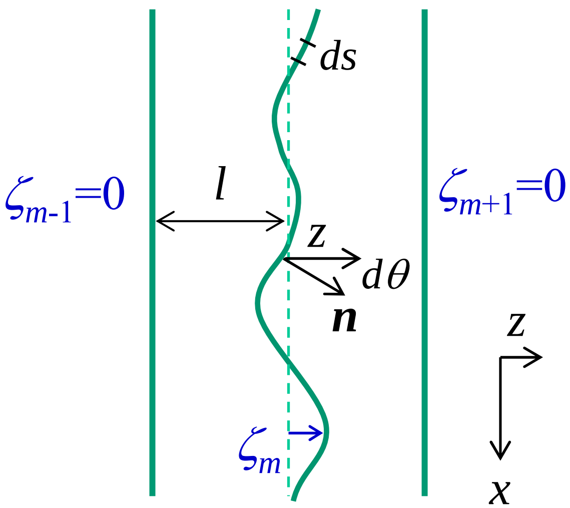


$$z_m(x) = ml + \zeta_m(x)$$

ステップ相互作用:

$$F = \sum_m \int dx \left[\frac{\tilde{\beta}}{2}\left(\frac{\partial z_m}{\partial x}\right)^2 + U\left[z_{m+1}(x) - z_m(x)\right] + U(l) \right]$$

$$\approx \sum_m \int dx \left[\frac{\tilde{\beta}}{2}\left(\frac{\partial z_m}{\partial x}\right)^2 + \frac{U''(l)}{2}\left[\zeta_{m+1}(x) - \zeta_m(x)\right]^2 \right]$$



Harmonic potential

$$w_h^2 = \frac{k_B T}{\sqrt{8U''(l)\tilde{\beta}}}$$

$$U(l) = A/l^2 \longrightarrow w_h^2 = \frac{k_B T}{\sqrt{48A\tilde{\beta}}} l^2$$

ステップ列のエネルギー

$$\zeta_m(x) = \int_0^{2\pi} \frac{d\phi}{2\pi} \int_{-\infty}^{\infty} \zeta_{k\phi} e^{i(kx+m\phi)} \frac{dk}{2\pi}$$

$$F = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \left[\frac{1}{2} \tilde{\beta} k^2 + U''(l)(1-\cos\phi) \right] \left| \zeta_{k\phi} \right|^2$$

ϕ=0 $$F = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \frac{1}{2} \tilde{\beta} k^2 \left| \zeta_{k\phi} \right|^2$$

⟶ **Identical with an isolated step**

ϕ=π/2 $$F = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \left[\frac{1}{2} \tilde{\beta} k^2 + U''(l) \right] \left| \zeta_{k\phi} \right|^2$$

⟶ **The effect from the neighboring steps disappears.**

ステップ列の揺らぎ

$$F = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \left[\frac{1}{2} \tilde{\beta} k^2 + U''(l)(1-\cos\phi) \right] \left| \zeta_{k\phi} \right|^2 \longrightarrow \left\langle \left| \zeta_{k\phi} \right|^2 \right\rangle_{eq} = \frac{k_B T}{\tilde{\beta} k^2 + 2U''(l)(1-\cos\phi)}$$

$$w_{eq}^2 \equiv \frac{1}{L} \int_0^L \frac{1}{N} \sum_m \left\langle [\zeta_m(x)]^2 \right\rangle_{eq} dx = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \left\langle \left| \zeta_{k\phi} \right|^2 \right\rangle_{eq}$$

@ϕ=π/2 $$w_{eq}^2 = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \frac{k_B T}{\tilde{\beta} k^2 + \frac{12A}{l^4}} = \frac{k_B T}{\sqrt{48 A \tilde{\beta}}} l^2$$

General case $$w_{eq}^2 = \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \frac{k_B T}{\tilde{\beta} k^2 + \frac{12A}{l^4}(1-\cos\phi)}$$

Cutoff ($\phi_0 = \frac{2\pi}{N}$ $k_0 = \frac{2\pi}{L}$ **)** $\tilde{\beta} k_0^2 > \frac{12A}{l^4} \phi_0^2 \xrightarrow[A \approx 10^{-30}\ \mathrm{Jm}]{\tilde{\beta} \approx 10^{-10}\ \mathrm{J/m}} \frac{Nl^2}{a} > L$

$$w_{eq}^2 \approx \int \frac{d\phi}{2\pi} \int \frac{dk}{2\pi} \frac{k_B T}{\tilde{\beta} k^2 + \frac{12A}{l^4} \phi^2} \approx \frac{k_B T l^2}{\sqrt{48 \tilde{\beta} A}} \int \frac{1}{k} \frac{dk}{2\pi} \approx \frac{k_B T l^2}{2\pi \sqrt{48 \tilde{\beta} A}} \ln\left(\frac{L}{L_e} \right)$$

where $L_e = \pi \sqrt{\tilde{\beta} A / l^4}$

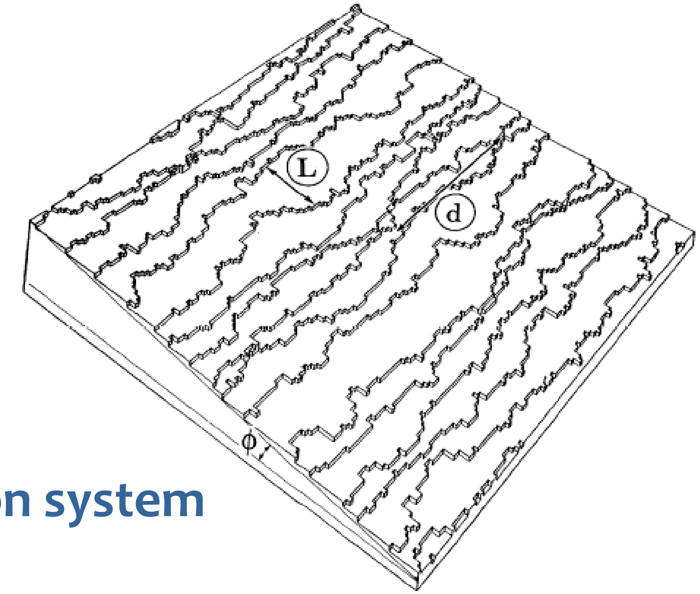
ステップ間斥力相互作用

エントロピー相互作用

B. Joos et al., Phys. Rev. B 43, 8153 (1991).

ステップが交差しない（揺らぎが制限される）
ことによるエネルギー上昇

$$0 \le z_1(x) < z_2(x) < \cdots < z_N(x) \le L_z$$



Noncrossing condition = one-dimensional fermion system

$$\frac{1}{2}\sum_{m=1}^{N}\int_0^L \tilde{\beta}\left(\frac{dz_m(x)}{dx}\right)^2 dx \iff H = \frac{k_BT}{2\tilde{\beta}}\sum k^2 \hat{a}_k^+ \hat{a}_k$$

$$E_1 = \frac{k_BT}{2\tilde{\beta}}\sum_{|k|\le k_F} k^2 \longrightarrow \frac{k_BT}{2\tilde{\beta}} L_z \frac{\pi^2}{3l^3}$$

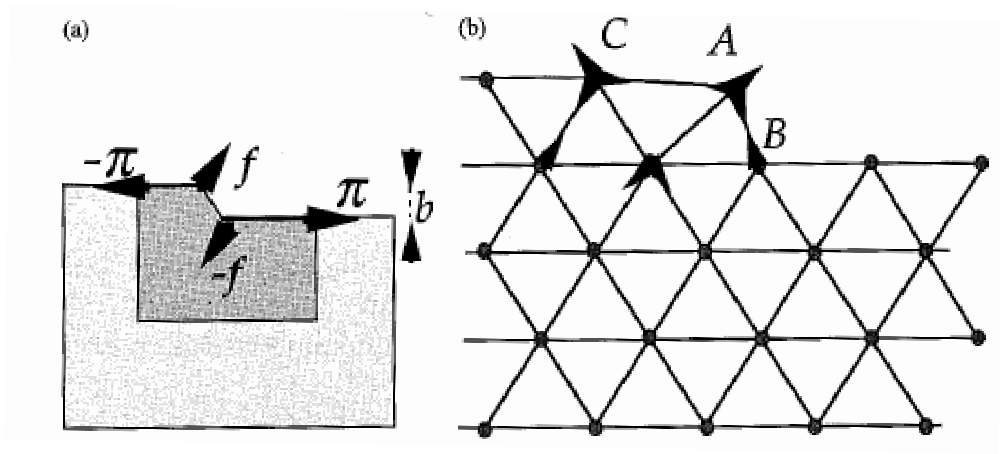
$$f_{\text{int}} = \frac{k_BT}{L_z}E_1 = \frac{\pi^2 (k_BT)^2}{6\tilde{\beta}(\theta) l^3} = \frac{\pi^2 (k_BT)^2}{6\tilde{\beta}(\theta)}\rho^3$$

$$\longrightarrow \quad f = f_0 + \beta(\theta)\rho + \frac{\pi^2 (k_BT)^2}{6\tilde{\beta}(\theta)}\rho^3$$

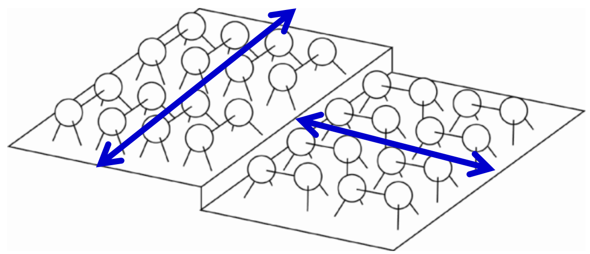
ステップ間斥力相互作用

弾性相互作用１　Force dipole

単位長さの相互作用エネルギー $U(l) = A/l^2 \propto \theta^2$



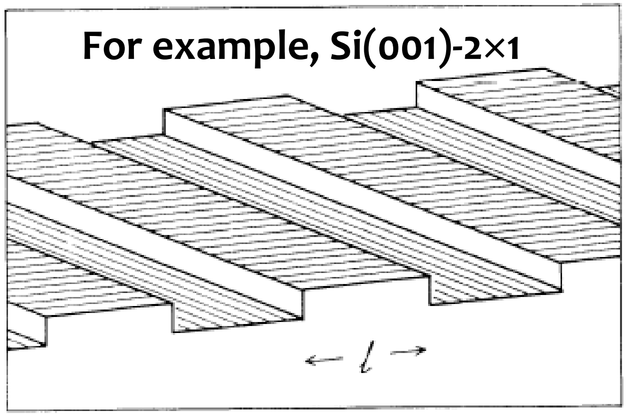
A. Pimpinelli and J. Villain, "Physics of Crystal Growth" (Cambridge University Press, 1998).



弾性相互作用２　Force monopole

$$U(l) = C_1 - C_2 \ln(l/\pi a)$$

O. L. Alerhand et al., Phys. Rev. Lett. 61, 1973 (1988).



ステップ間斥力相互作用

Fluctuation of steps interacting with an elastic interaction

⟶ **One-dimensional interacting fermion system**

$$H = -\sum_{m=1}^{N} \frac{k_B T}{2\tilde{\beta}} \frac{\partial^2}{\partial z_m^2} + \frac{A}{k_B T} \sum_{m<m'} \frac{1}{|z_m - z_{m'}|^2} = \frac{k_B T}{2\tilde{\beta}} \left[-\sum_{m=1}^{N} \frac{\partial^2}{\partial z_m^2} + g \sum_{m<m'} \frac{1}{|z_m - z_{m'}|^2} \right]$$

where $g = \dfrac{2\tilde{\beta}A}{(k_B T)^2}$

$$f_{\mathrm{int}}(g) = \frac{\pi^2 (k_B T)^2}{6\tilde{\beta}(\theta) l^3} \bar{\lambda}^2(g)\text{, where } \bar{\lambda}^2(g) = \frac{1}{2}\left(1 + \sqrt{1+2g}\right)$$

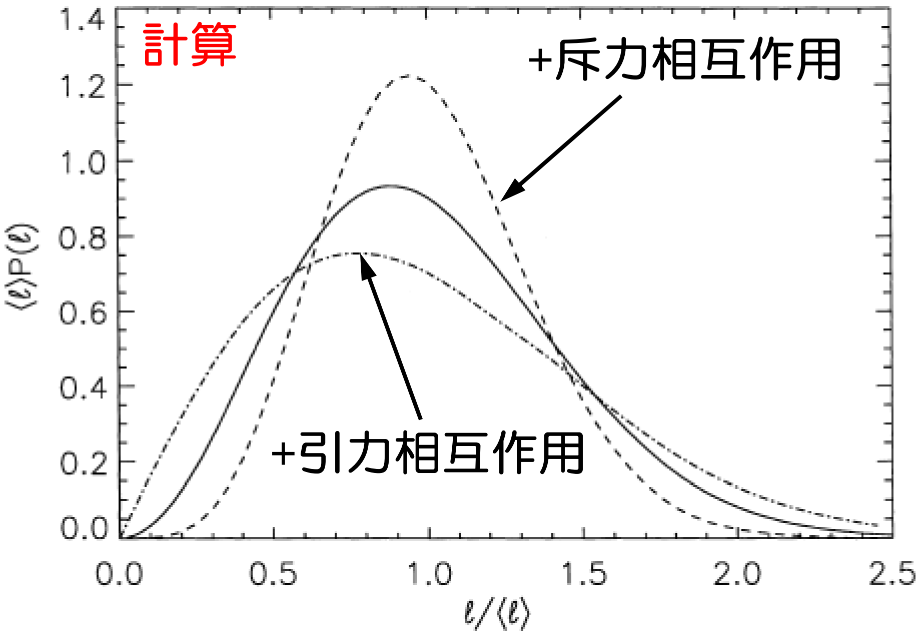
$$U''_{\mathit{eff}}(g) = \frac{\left(\pi k_B T \bar{\lambda}(g)\right)^2}{6\tilde{\beta}(\theta) l^2}$$

$$= \frac{(\pi k_B T)^2}{24\tilde{\beta}(\theta) l^2} \left[1 + \sqrt{1 + \frac{4\tilde{\beta}A}{(k_B T)^2}} \right]^2$$

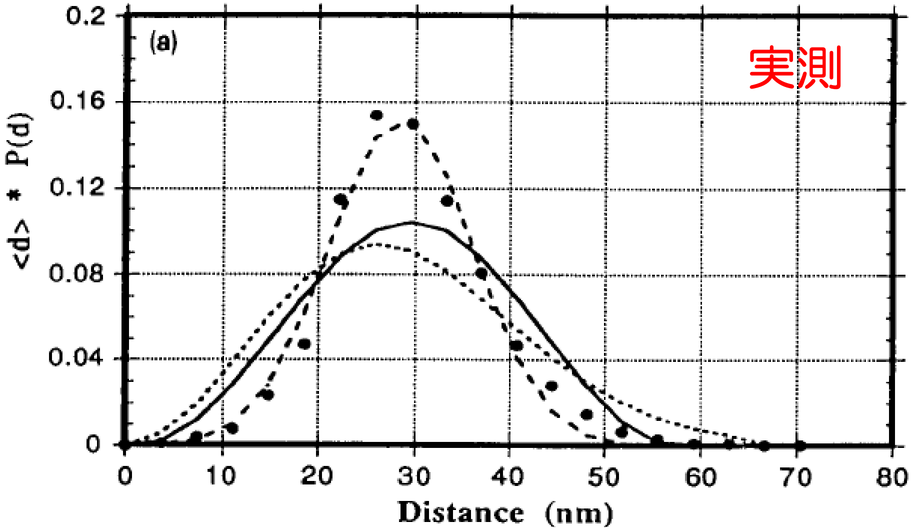
ステップ相互作用エネルギーの測定

$$U(l) = A/l^2 \Rightarrow \beta_2 = \frac{(\pi k_B T)^2}{24 l^2 \tilde{\beta}}\left[1 + \sqrt{1 + \frac{4A\tilde{\beta}}{(k_B T)^2}}\right]^2$$

テラス幅の分布

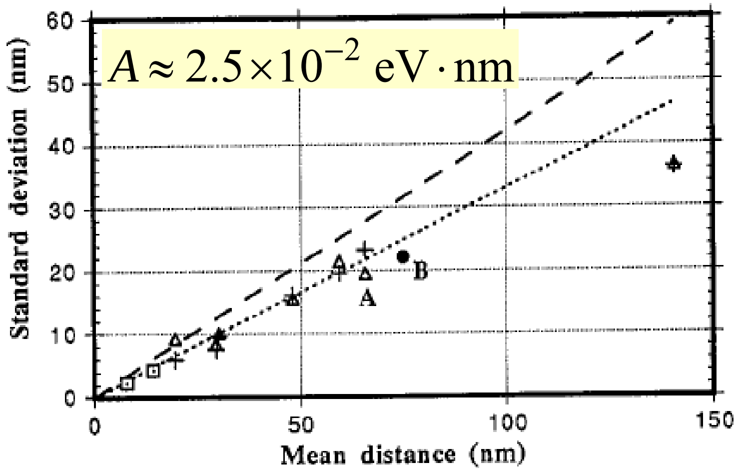


H.-C. Jeong, E.D. Williams / Surface Science Reports 34 (1999) 171–294



弾性的双極子相互作用が支配的

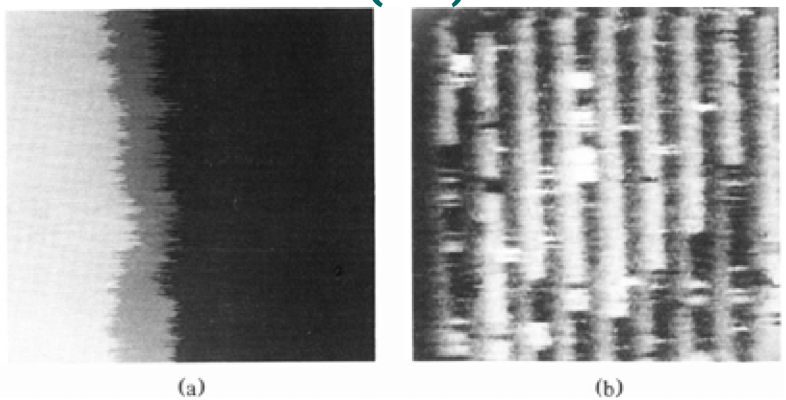
$$\Rightarrow \frac{\sigma}{<l>} = \left(\frac{(k_B T)^2}{48\tilde{\beta}A}\right)^{1/4}$$

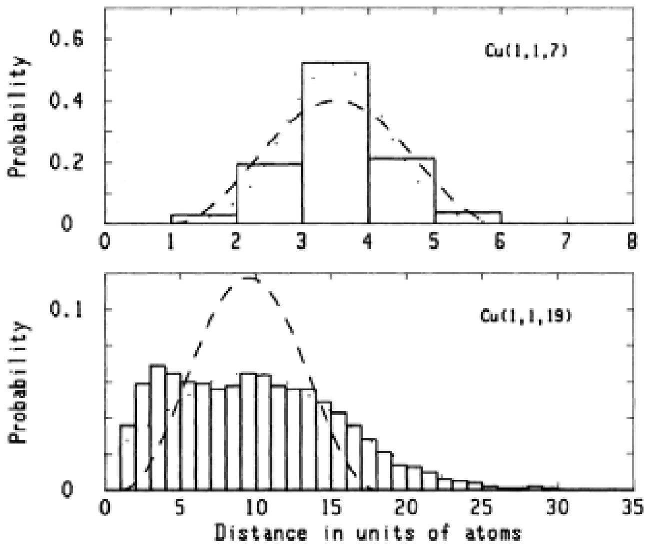


C. Alfonso, J.M. Bermond, J.C. Heyraud, J.J. Métois, Surf. Sci. 262 (1992) 371.

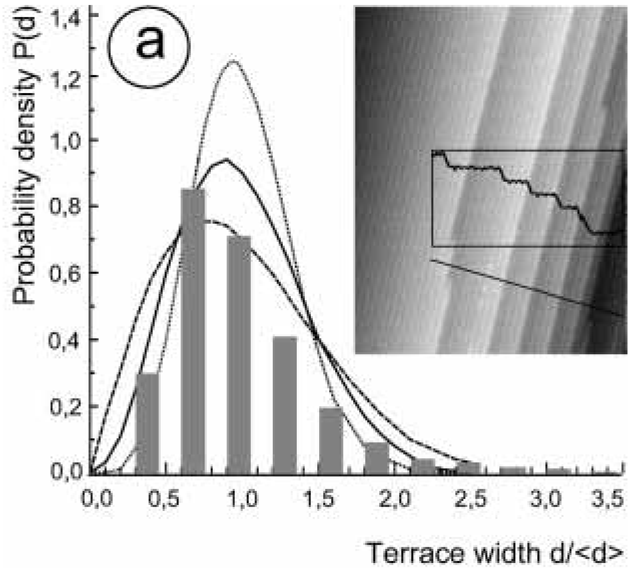
ステップ間引力相互作用

Cu(11n)



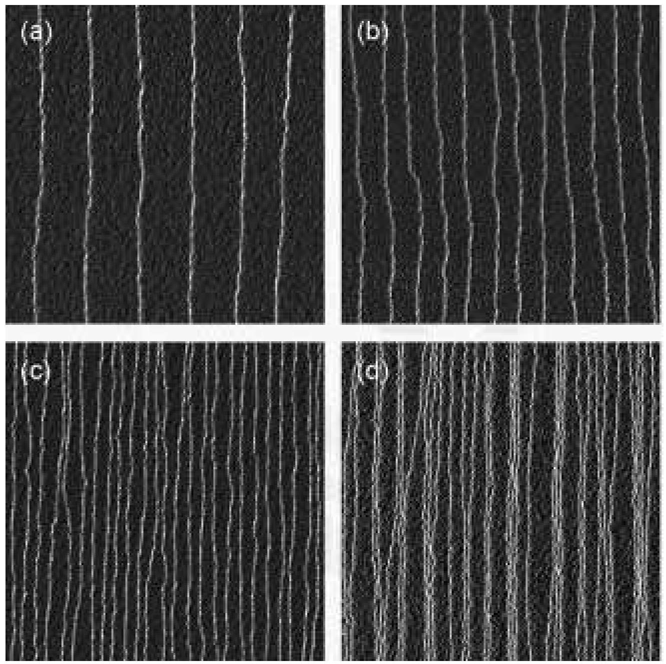


J. Frohn et al., Phys. Rev. Lett. 67, 3543 (1991).

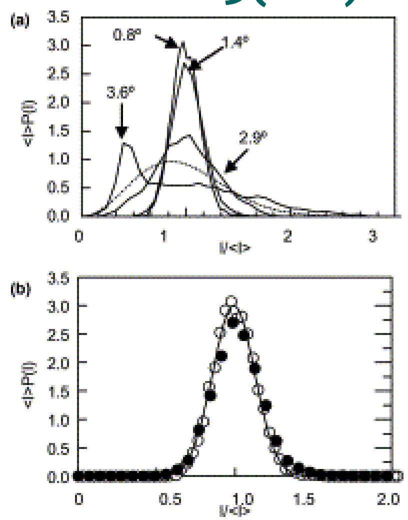


Pt(110)

K. Swamy et al., Surf. Sci. 425, L369 (1999).



SrTiO3(001)



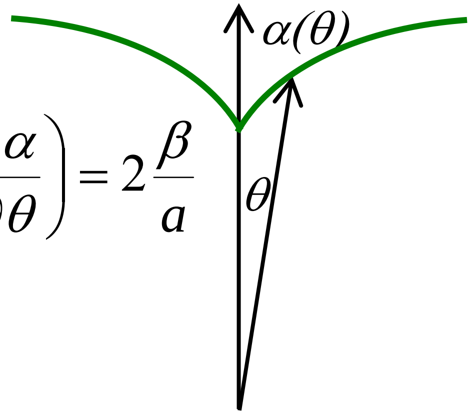
K. Sudoh et al., Surf. Sci. 557, L151 (2004).

ファセット近傍の結晶の形

射影自由エネルギー密度

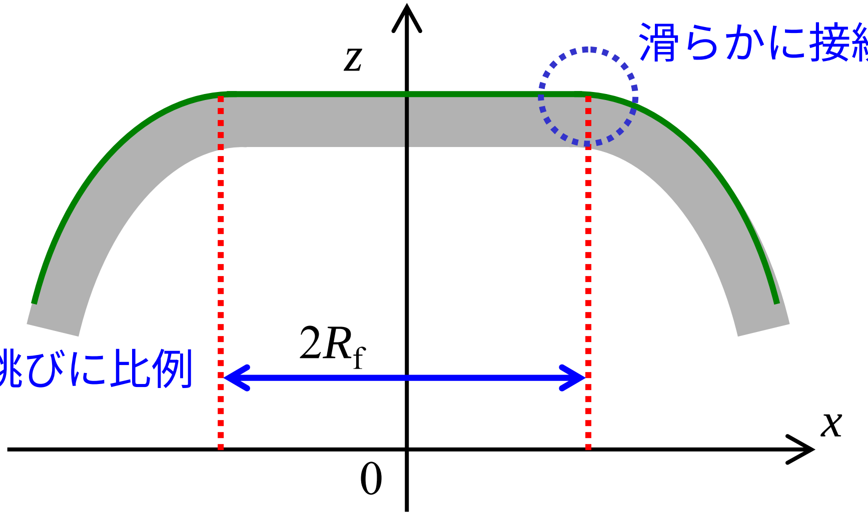
$$f\,(p_x,0)=\alpha(0,0)+\frac{\beta}{a}|p_x|+\frac{\phi}{a^3}|p_x|^3$$

$$\Delta\left(\frac{\partial\alpha}{\partial\theta}\right)=2\frac{\beta}{a}$$



平衡形

$$\lambda z(x)=\begin{cases}\alpha(0,0) & \left(0\le\lambda x\le\dfrac{\beta}{a}\right)\\ \alpha(0,0)-\left(\dfrac{4a^3}{27\phi}\right)^{1/2}\left(\lambda x-\dfrac{\beta}{a}\right)^{3/2} & \left(\dfrac{\beta}{a}<\lambda x\right)\end{cases}$$



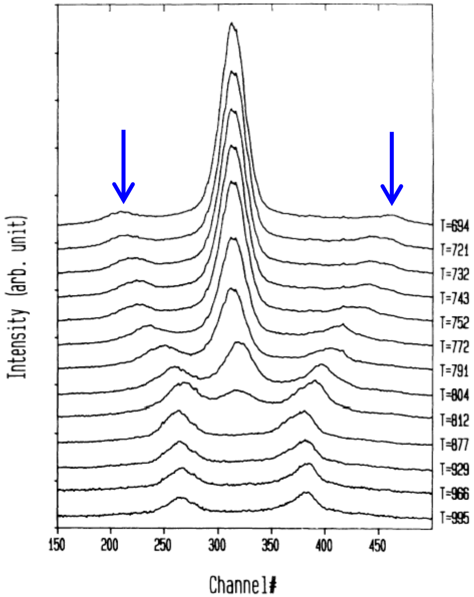
ファセットの幅は$\alpha(0)$の傾きの跳びに比例

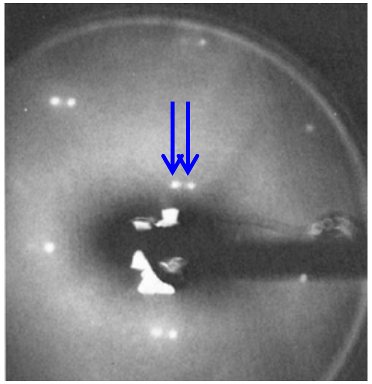
$$2\lambda R_f=2\frac{\beta}{a}=\Delta\left(\frac{\partial\alpha}{\partial\theta}\right)$$

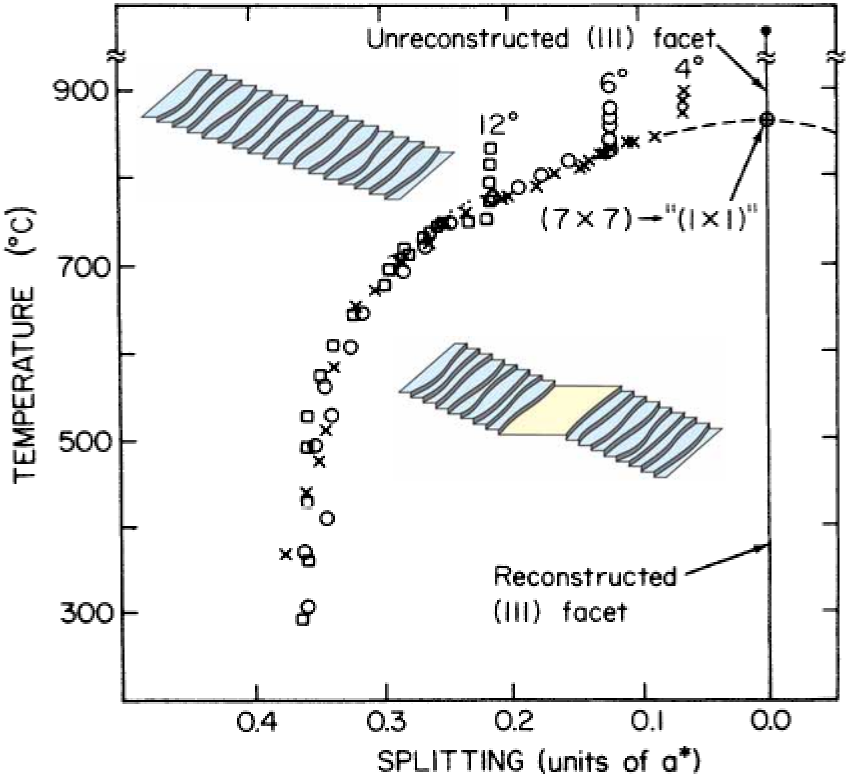
ステップ領域の傾斜角の温度依存性

$[1\bar{1}0]$方向に傾斜したSi(111)表面でのステップバンチング

$$\tan\theta = \left(\frac{a\Delta\gamma}{2\beta''_{1\times1}}\right)^{1/3} \propto \left(\frac{T_c - T}{T_c}\right)^{1/3}$$







R. J. Phaneuf and E. D. Williams, Phys. Rev. Lett. 58, 2563 (1987).   *R. J. Phaneuf at el., Phys. Rev. B 38, 1984 (1988)*

熱平衡揺らぎのダイナミクス

熱平衡でのステップ揺らぎダイナミクス

$$\zeta_m(x)=\int_0^{2\pi}\frac{d\phi}{2\pi}\int_{-\infty}^{\infty}\frac{dk}{2\pi}\zeta_{k\phi}e^{i(kx+m\phi)}$$

Linear Langevin equation

$$\frac{\partial\zeta_{k\phi}(t)}{\partial t}=i\omega_{k\phi}\zeta_{k\phi}(t)+\eta_{k\phi}(t)$$

Thermal noise

$$\left\langle\eta_{k\phi}(t)\eta_{k'\phi'}(t')\right\rangle=-8\pi^2\left\langle\left|\zeta_{k\phi}\right|^2\right\rangle_{eq}i\omega_{k\phi}\delta(k+k')\delta(\phi+\phi')\delta(t-t')$$

Step fluctuation width

$$w^2(t)=\left\langle\zeta_m(x,t)^2\right\rangle=\int_0^{2\pi}\frac{d\phi}{2\pi}\int_{-\infty}^{\infty}\frac{dk}{2\pi}\left\langle\left|\zeta_{k\phi}(t)\right|^2\right\rangle$$

Terrace width fluctuation

$$W^2(t)=\left\langle[\zeta_{m+1}(x,t)-\zeta_m(x,t)]^2\right\rangle=\int_0^{2\pi}\frac{d\phi}{2\pi}\int_{-\infty}^{\infty}\frac{dk}{2\pi}\left\langle\left|\zeta_{k\phi}(t)\right|^2\right\rangle(1-\cos\phi)$$

Time correlation

$$G(t)=\left\langle[\zeta_m(x,t)-\zeta_m(x,0)]^2\right\rangle_{eq}=2\int_0^{2\pi}\frac{d\phi}{2\pi}\int_{-\infty}^{\infty}\frac{dk}{2\pi}\left\langle\left|\zeta_{k\phi}\right|^2\right\rangle_{eq}\left(1-e^{i\omega_{k\phi}t}\right)=2w^2(t/2)$$

熱平衡でのステップ揺らぎダイナミクス

$$i\omega_{k\phi} = i\omega_k = -A_0|k|^n$$

$$w^2(t) = \frac{G(2t)}{2} = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk \left\langle \left|\zeta_{k\phi}\right|^2 \right\rangle_{eq} \left(1 - e^{-2A_0 k^n t}\right)$$

$$\downarrow \quad \left\langle \left|\zeta_{k\phi}\right|^2 \right\rangle_{eq} = \frac{k_B T}{\tilde{\beta} k^2}$$

$$= \frac{k_B T}{2\pi\tilde{\beta}} 2\int_0^{\infty} \frac{dk}{k^2}\left(1 - e^{-2A_0 k^n t}\right)$$

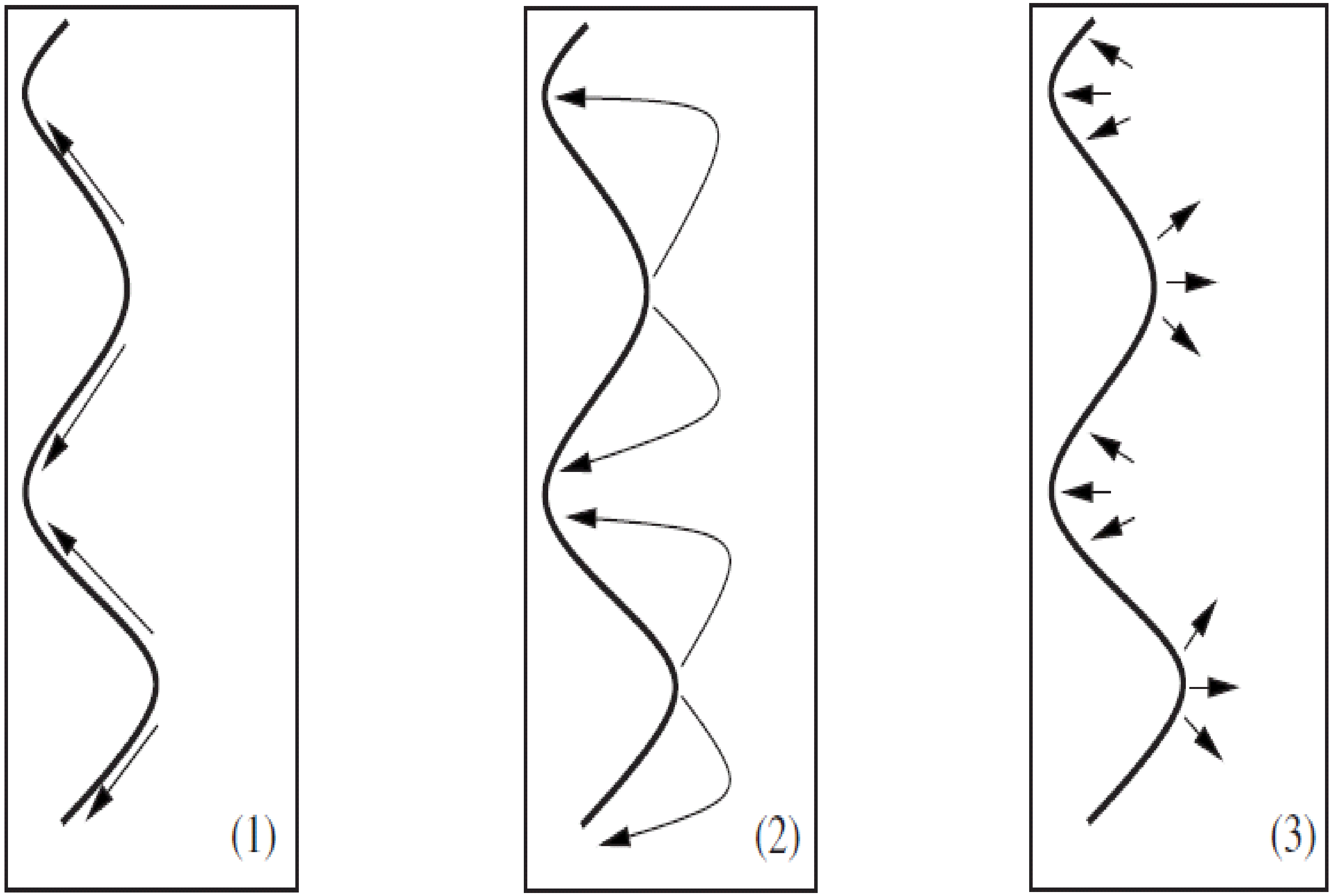
$$= \frac{k_B T}{\pi\tilde{\beta}} n(2A_0 t)\int_0^{\infty} dk k^{n-2} e^{-2A_0 k^n t}$$

$$= \frac{k_B T}{\pi\tilde{\beta}} \Gamma\left(1 - \frac{1}{n}\right)(2A_0 t)^{1/n}$$

, where $\Gamma(z) = \int_0^{\infty} x^{z-1} e^{-x} dx$

$$\Gamma\left(\frac{1}{2}\right) = \sqrt{\pi},\ \Gamma\left(\frac{2}{3}\right) = 1.35175...,\ \Gamma\left(\frac{3}{4}\right) = 1.22541...$$

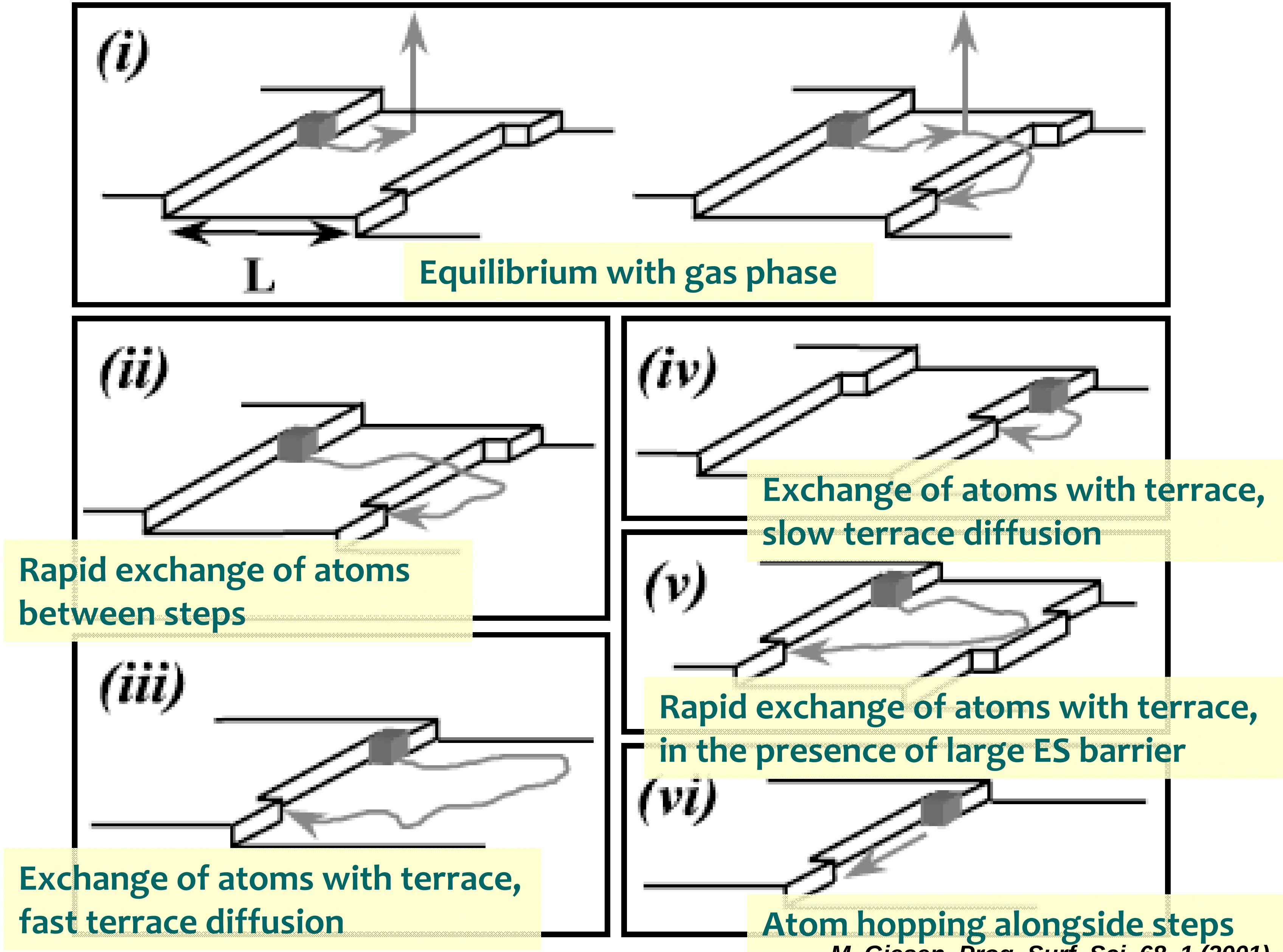
質量輸送機構



Edge diffusion　Terrace diffusion　Attachment/detachment

C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

質量輸送機構



M. Giesen, Prog. Surf. Sci. 68, 1 (2001).

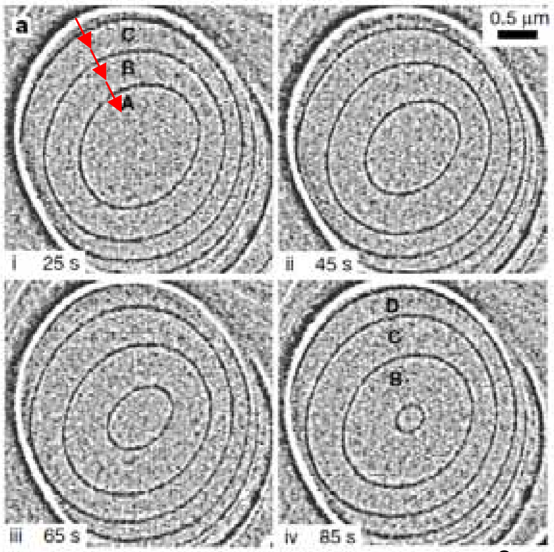
固体内の空孔拡散

Vacancies in solids and the stability of surface morphology

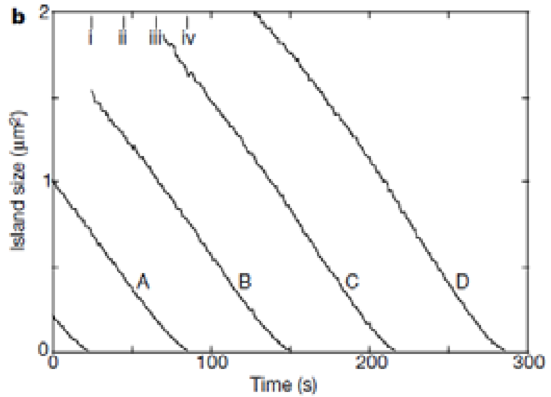
NiAl(110)

K. F. McCarty, J. A. Nobel & N. C. Bartelt NATURE | VOL 412 | 9 AUGUST 2001 **622**

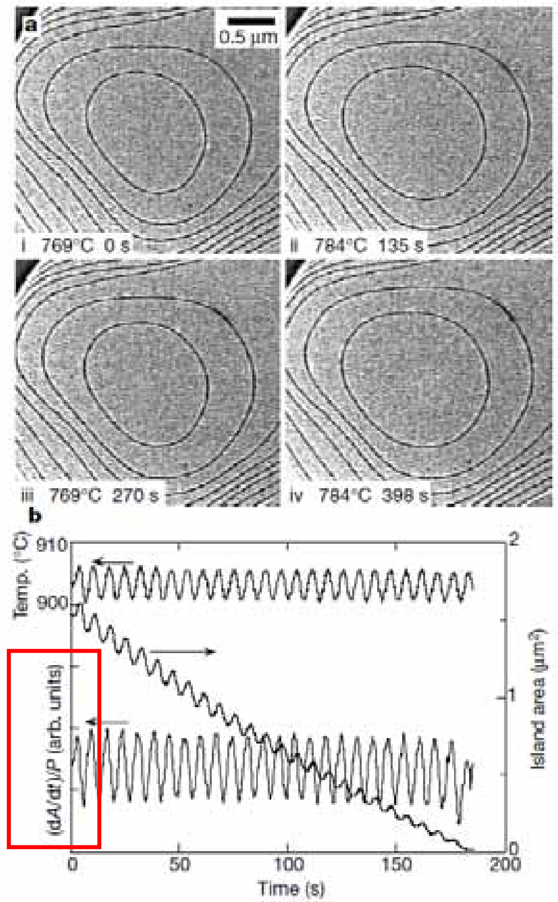
difficult. Here we show that vacancy generation (and annihilation) on the (110) surface of an ordered nickel–aluminium intermetallic alloy does not occur over the entire surface, but only near atomic step edges. This has been determined by



957°C



$$\frac{dA}{dt} = -\frac{2\pi a^4 h_{B-S} c_B \beta}{kT} = \text{constant}$$



表面でのアドアトム・空孔対形成

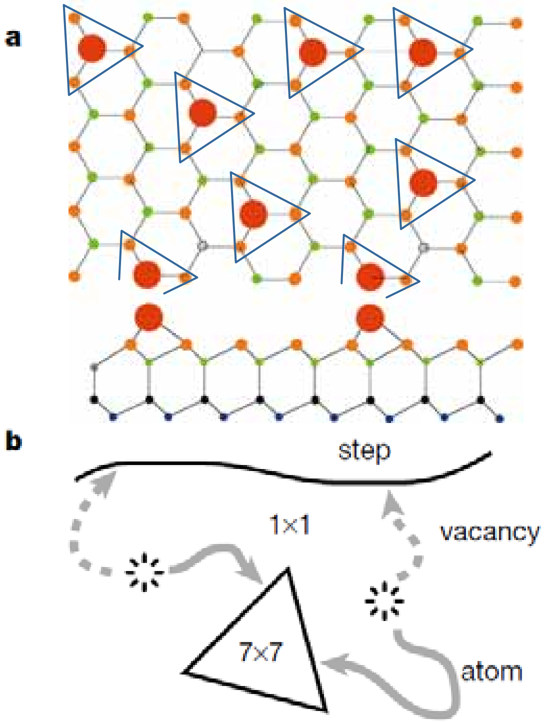
Dynamics of the silicon (111) surface phase transition

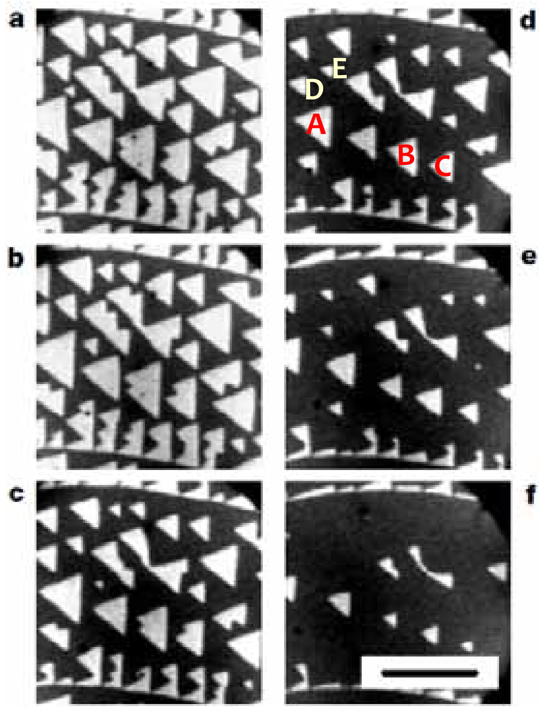
Si(111)

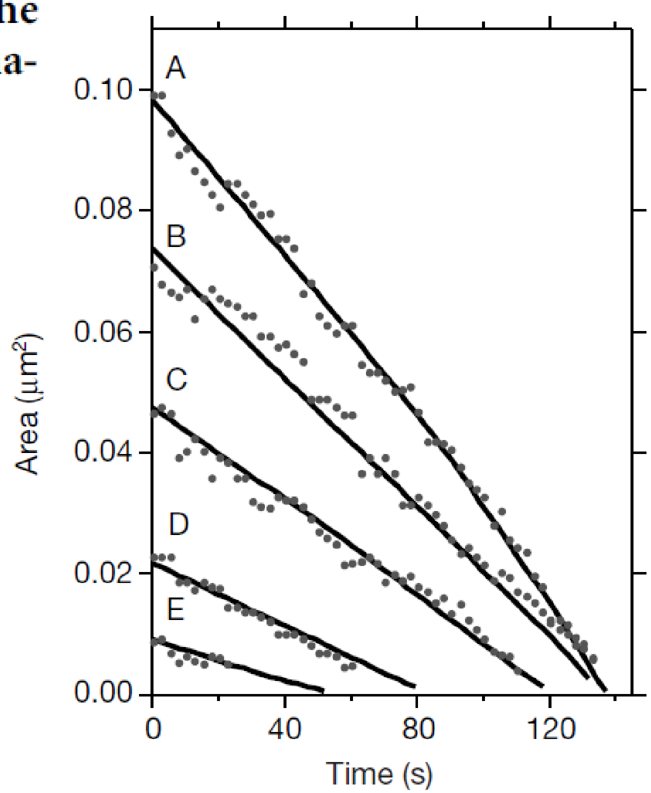
J. B. Hannon*, H. Hibino†, N. C. Bartelt‡, B. S. Swartzentruber§, T. Ogino† & G. L. Kellogg§

NATURE | VOL 405 | 1 JUNE 2000 | 552

to the silicon (111) surface. We show that the transformation is governed by the rate at which material is exchanged between the first layer of the crystal and the surface. In bulk phase transforma-







付着脱離律速

Step velocity

$$V_{\pm} = \Omega K_{\pm}\left(c_{\pm} - c_{eq}\right) + \eta_{\pm} \qquad V = V_{+} + V_{-}$$

Equilibrium concentration

$$c_{eq} = c_{eq}^{0} e^{\mu/k_B T} \approx c_{eq}^{0}\left[1 + \frac{\mu}{k_B T}\right]$$

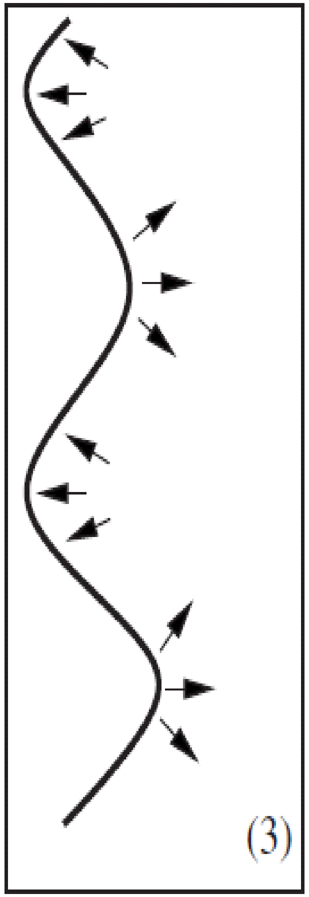
$$\mu = \frac{\delta F}{\delta N} = \frac{\delta F}{\delta \zeta}\frac{\delta \zeta}{\delta N}$$

$$F = \frac{1}{2}\tilde{\beta}\int\left(\frac{\partial \zeta}{\partial x}\right)^{2} dx$$

Curvature: $\kappa = -\dfrac{\partial^2 \zeta}{\partial x^2} \Big/ \left[1 + \left(\dfrac{\partial \zeta}{\partial x}\right)^{2}\right]^{3/2} \approx -\dfrac{\partial^2 \zeta}{\partial x^2}$

$$\mu = \Omega\tilde{\beta}\frac{\partial^2 \zeta}{\partial x^2} = \Omega\tilde{\beta}\kappa$$

$$c_{eq} \approx c_{eq}^{0}\left[1 + \Omega\frac{\tilde{\beta}\kappa}{k_B T}\right]$$



C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

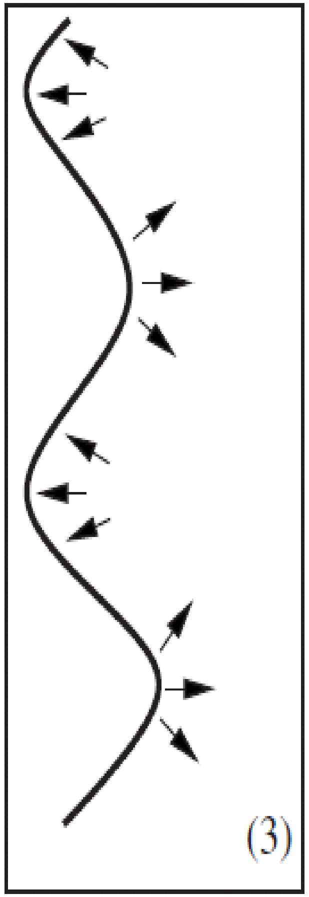
付着脱離律速

$$\frac{\partial \zeta}{\partial t} = V_+ + V_- = (K_+ + K_-)\Omega c_{eq}^0 \Gamma \frac{\partial^2 \zeta}{\partial x^2} + (\eta_+ + \eta_-)\Omega$$

$$\zeta = \sum_k \zeta_k e^{ikx + i\omega_k t}$$

$$i\omega_k = -(K_+ + K_-)\Omega c_{eq}^0 \Gamma k^2$$

$$w^2(t) = \frac{G(2t)}{2} = \frac{k_B T}{\pi \tilde{\beta}} \Gamma\left(\frac{1}{2}\right)\left[2(K_+ + K_-)\Omega c_{eq}^0 \Gamma t\right]^{1/2}$$



C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

エッジ拡散律速

Normal velocity

$$V_n = -\Omega \frac{\partial J_L}{\partial s}$$

$$J_L = -\left(\frac{M}{\Omega}\right)\frac{\partial \mu}{\partial s} \quad \textbf{with a mobility of } M = \frac{aD_L}{k_B T}$$

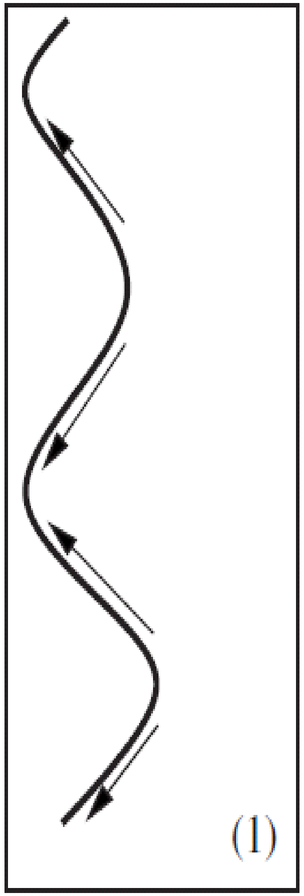
$$\mu = \Omega \tilde{\beta} \kappa$$

$$V_n = \frac{\partial}{\partial s}\left[aD_L \Gamma \frac{\partial \kappa}{\partial s}\right]$$

$$\frac{\partial \zeta}{\partial t} = -aD_L \Gamma \frac{\partial^4 \zeta}{\partial x^4} + \eta_e$$

$$\longrightarrow \quad i\omega_k = -aD_L \Gamma k^4$$

$$w^2(t) = \frac{G(2t)}{2} = \frac{k_B T}{\pi \tilde{\beta}} \Gamma\left(\frac{3}{4}\right)\left[2aD_L \Gamma t\right]^{1/4}$$



C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

表面拡散律速

Diffusion equation

$$\frac{\partial c}{\partial t} = D\nabla^2 c + F - \frac{c}{\tau} = 0$$

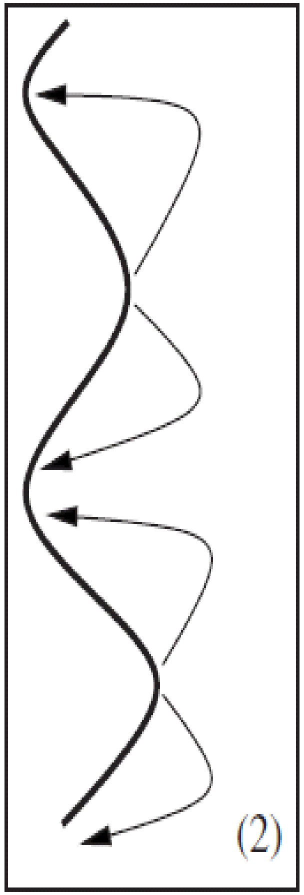
$$\downarrow F = \tau^{-1} = 0$$

$$D\nabla^2 c = 0$$

$$\left.\begin{aligned} \zeta(x,t) &= \zeta_k(t)e^{ikx} \\ c(x,z,t) &= c_k(z,t)e^{ikx} \end{aligned}\right\}$$ **Fluctuation with the same wavelength**

$$D\nabla^2 c = 0 \longrightarrow \frac{\partial^2 c}{\partial z^2} - k^2 c = 0$$

$$\rightarrow \begin{cases} c(x,z>0,t) = c_{eq}^0[1+\Gamma\kappa] = c_{eq}^0\left[1+\Gamma k^2\zeta_k(t)e^{ikx-|k|z}\right] \\ c(x,z<0,t) = c_{eq}^0\left[1+\Gamma k^2\zeta_k(t)e^{ikx+|k|z}\right] \end{cases}$$



C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

表面拡散律速

Step velocity

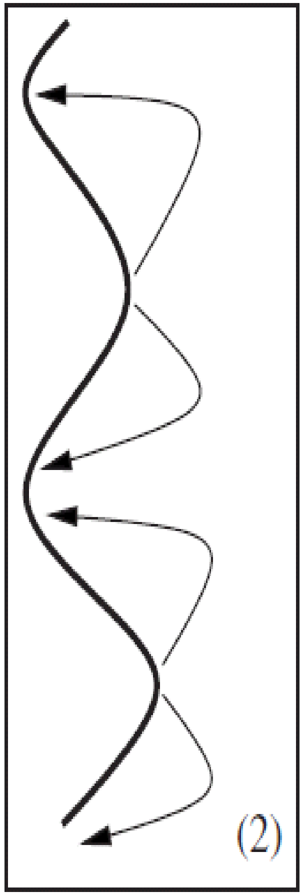
$$V_{\pm} = \pm \Omega D \vec{n} \cdot \nabla c_{\pm} \approx \pm \Omega D \frac{\partial c_{\pm}}{\partial z}$$

$$\vec{n} = \left(-\frac{\partial \zeta}{\partial x}, 1 \right) \Big/ \sqrt{1 + \left(\frac{\partial \zeta}{\partial x} \right)^2} \approx \left(-\frac{\partial \zeta}{\partial x}, 1 \right)$$

$$\zeta = \sum_k \zeta_k e^{ikx + i\omega_k t}$$

$$i\omega_k = -2\Omega D c_{eq}^0 \Gamma |k|^3 = -2 D_S \Gamma |k|^3$$

$$w^2(t) = \frac{G(2t)}{2} = \frac{k_B T}{\pi \tilde{\beta}} \Gamma\left(\frac{2}{3}\right) [4 D_s \Gamma t]^{1/3}$$



C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

質量輸送の律速過程の判別

Edge diffusion

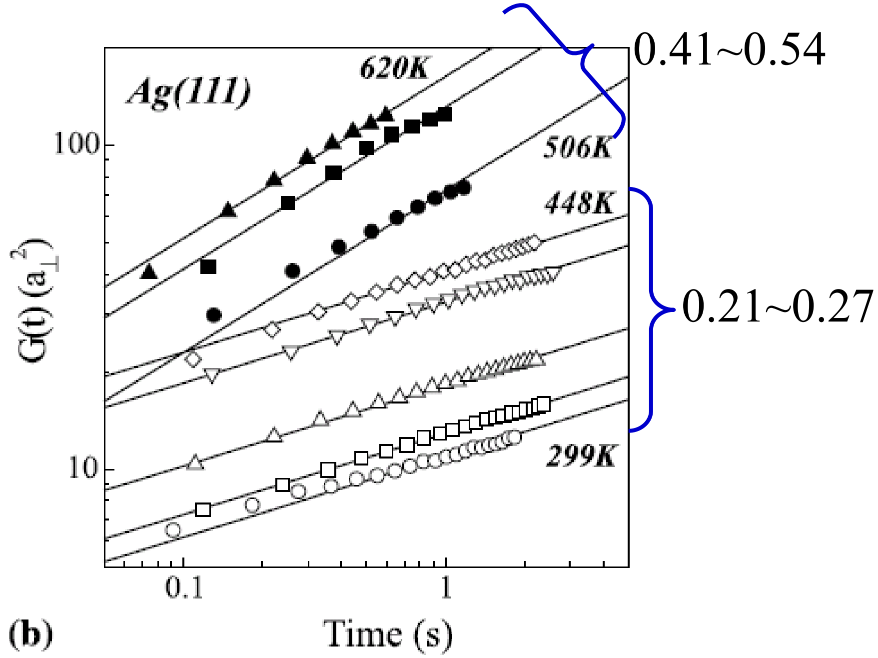
$$G(t) = 2\frac{k_B T}{\pi\tilde{\beta}}\Gamma\left(\frac{3}{4}\right)\left[aD_L\Gamma t\right]^{1/4}$$

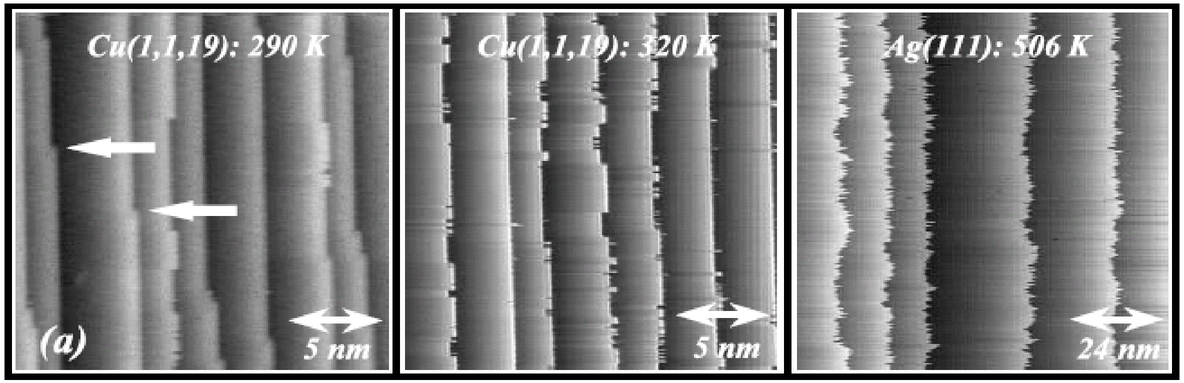
Terrace diffusion

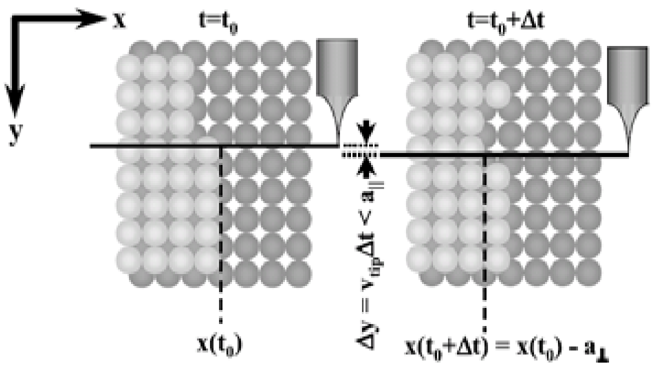
$$G(t) = 2\frac{k_B T}{\pi\tilde{\beta}}\Gamma\left(\frac{2}{3}\right)\left[2D_s\Gamma t\right]^{1/3}$$

Attachment/detachment

$$G(t) = 2\frac{k_B T}{\pi\tilde{\beta}}\Gamma\left(\frac{1}{2}\right)\left[(K_+ + K_-)\Omega c_{eq}^0\Gamma t\right]^{1/2}$$







M. Giesen / Progress in Surface Science 68 (2001) 1–153

質量輸送の律速過程の判別

$$w^2(t) = \frac{G(2t)}{2} = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk \left\langle \left|\zeta_{k\phi}\right|^2 \right\rangle_{eq} \left(1 - e^{-2A_0 k^{n} t}\right)$$

$$G_k(t) = \frac{k_B T}{L\tilde{\beta}k^2}\left(1 - e^{-A_0 k^{n} t}\right) = \frac{k_B T}{L\tilde{\beta}k^2}\left(1 - e^{-t/\tau(k)}\right)$$

$$\tau(k) = 1/A_0 k^n$$

Edge diffusion

$$\tau(k) = \frac{k_B T}{\Omega a D_L \tilde{\beta} k^4}$$

Terrace diffusion

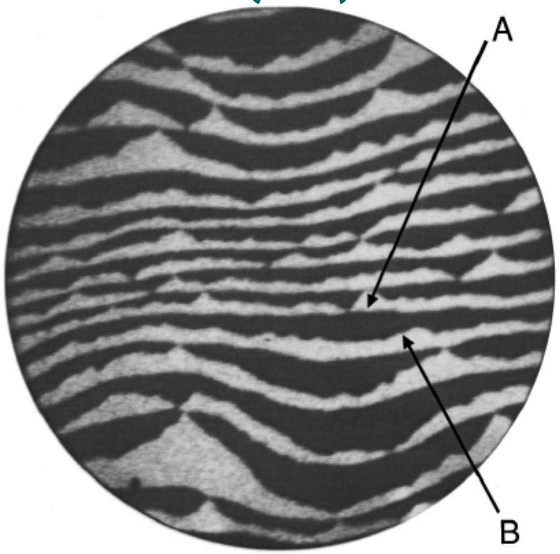
$$\tau(k) = \frac{k_B T}{2\Omega^2 D c_{eq}^0 \tilde{\beta} k^3}$$

Attachment/detachment

$$\tau(k) = \frac{k_B T}{\Omega^2 (K_+ + K_-) c_{eq}^0 \tilde{\beta} k^2}$$

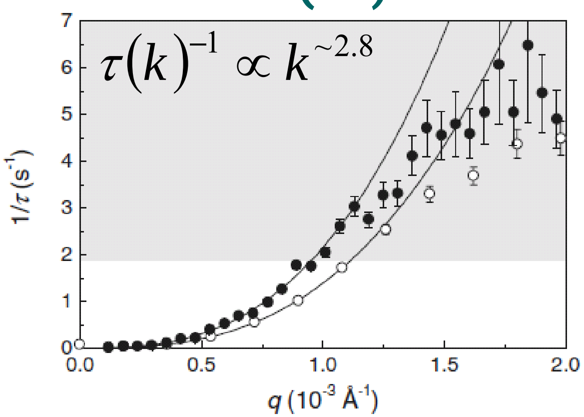
質量輸送の律速過程の判別(実験)

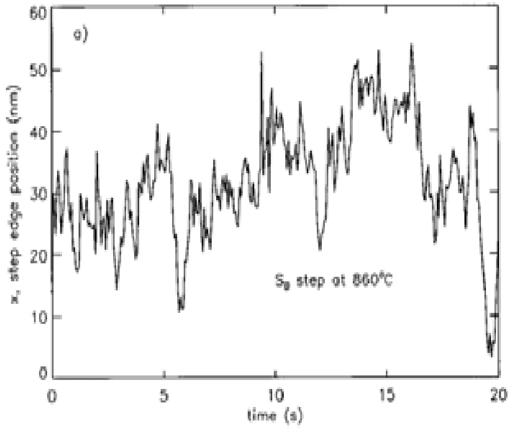
Si(001)

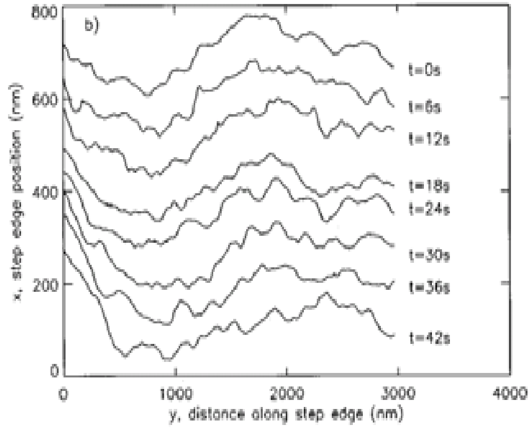


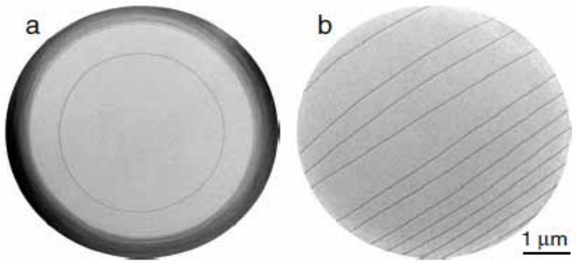
N. C. Bartelt and R. M. Tromp, Phys. Rev. B 54, 11731 (1996).

Si(111)



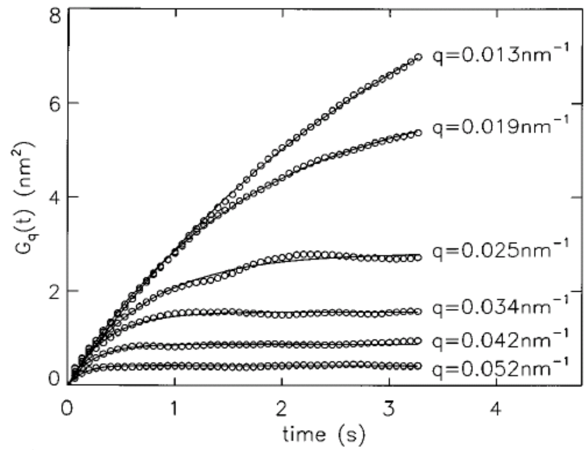




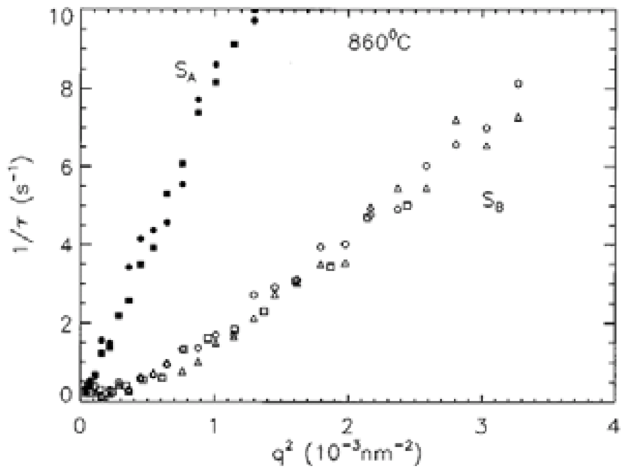


A. B. Pan et al., Phys. Rev. B 77, 115424 (2008).

$$G_k(t) = \frac{k_B T}{L\tilde{\beta}k^2}\left(1 - e^{-t/\tau(k)}\right)$$

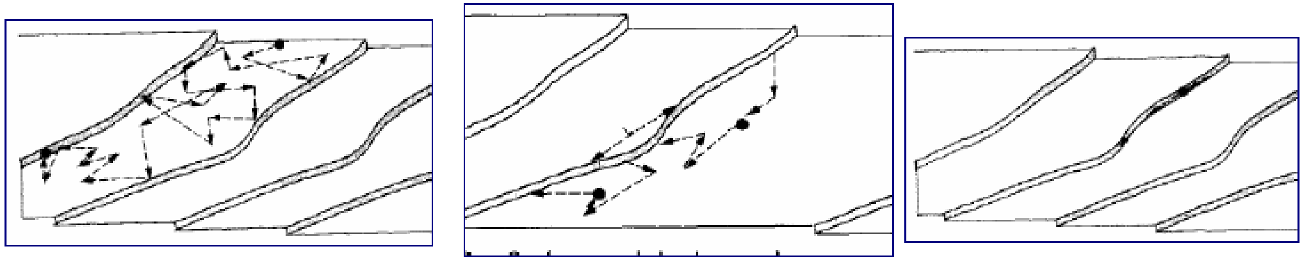


$$\tau(k)^{-1} \propto k^2$$



質量輸送機構

質量輸送の律速過程の判別法



	EC or AD (ADL)	TD (DL)	PD
Limited by	At/de/tach at step	Terrace diffu'n	**Step-edge diffu'n**
Fluctuation healing time--width y	y^2	y^3	y^4
Size dep. of island diffu'n, $R \propto \sqrt{\text{area}}$	R^{-1}	R^{-2}	R^{-3}
$w^2(t)$	$t^{1/2}$	$t^{1/3}$	$t^{1/4}$
Island area decay	t^1	$t^{2/3}$	N/A
Evolution of atom/ vacancy island	Shrink to round point *(Grayson's Thm)*		Wormlike, pinch-off
Height decay of cone ["facet"]	$t^{1/4}$	$t^{1/4}$	N/A
Height decay of paraboloid [rough]	$t^{1/3}$	$t^{2/5}$	N/A

www2.physics.umd.edu/~einstein/talks/UNHColloq.pdf

表面形状の緩和１

$$\frac{\partial c(x,y,t)}{\partial t} = D\nabla^2 c(x,y,t) + F - \frac{1}{\tau_d} c(x,y,t)$$

準静的近似　　　成長なし、脱離なし

$$\nabla^2 c = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c}{\partial r}\right) = 0$$

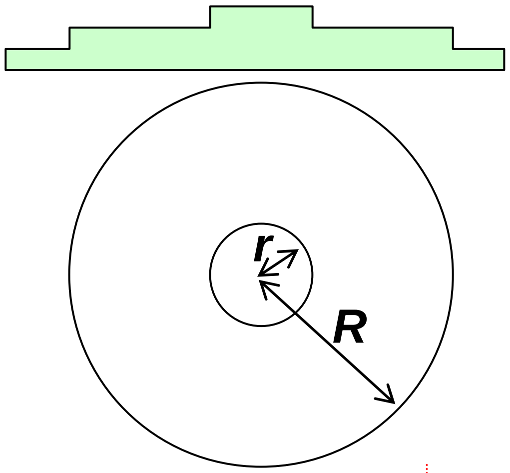
$$D\left.\frac{\partial c}{\partial r}\right|_r = K_+\left(c(r) - c_{eq}^0 - c_{eq}^0\frac{\tilde{\beta}\Omega}{k_B T r}\right)$$

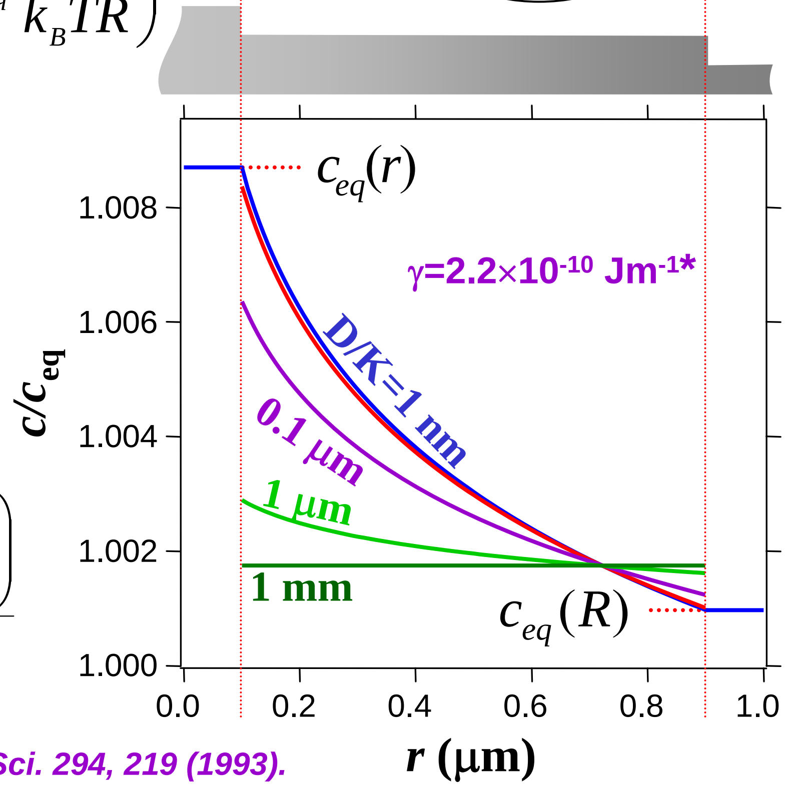
$$-D\left.\frac{\partial c}{\partial r}\right|_R = K_-\left(c(R) - c_{eq}^0 - c_{eq}^0\frac{\tilde{\beta}\Omega}{k_B T R}\right)$$

Solution: $c(r) = a \cdot \ln r + b$

$$a = \frac{-c_{eq}^0\left(1+\frac{\tilde{\beta}\Omega}{k_B T r}\right) + c_{eq}^0\left(1+\frac{\tilde{\beta}\Omega}{k_B T R}\right)}{\frac{D}{K_+ r} + \frac{D}{K\_R} + \ln\frac{R}{r}}$$

$$b = c_{eq}^0\frac{\left(1+\frac{\tilde{\beta}\Omega}{k_B T R}\right)\left(\frac{D}{K_+ r} - \ln r\right) + \left(1+\frac{\tilde{\beta}\Omega}{k_B T r}\right)\left(\frac{D}{K\_R} + \ln R\right)}{\frac{D}{K_+ r} + \frac{D}{K\_R} + \ln\frac{R}{r}}$$





*E. D. Williams et al., Surf. Sci. 294, 219 (1993).

表面形状の緩和２

Area change rate of 2D island

$$\frac{dA}{dt} = 2\pi r\Omega D\frac{dc}{dr} = 2\pi r\Omega D = 2\pi\Omega D\frac{-c_{eq}^{0}\left(1+\frac{\Omega\tilde{\beta}}{k_B Tr}\right)+c_{eq}^{0}\left(1+\frac{\Omega\tilde{\beta}}{k_B TR}\right)}{\frac{D}{K_+ r}+\frac{D}{K\_R}+\ln\frac{R}{r}}$$

Kinetic distance

Diffusion-limited: $r >> \frac{D}{K_+} = d_+ \quad r << R$

$$\Rightarrow \frac{dA}{dt} = -\frac{2\pi\Omega^2\tilde{\beta}c_{eq}^{0}D}{k_B T\ln R}\frac{1}{r} \Rightarrow A(t) \propto (t_0 - t)^{2/3}$$

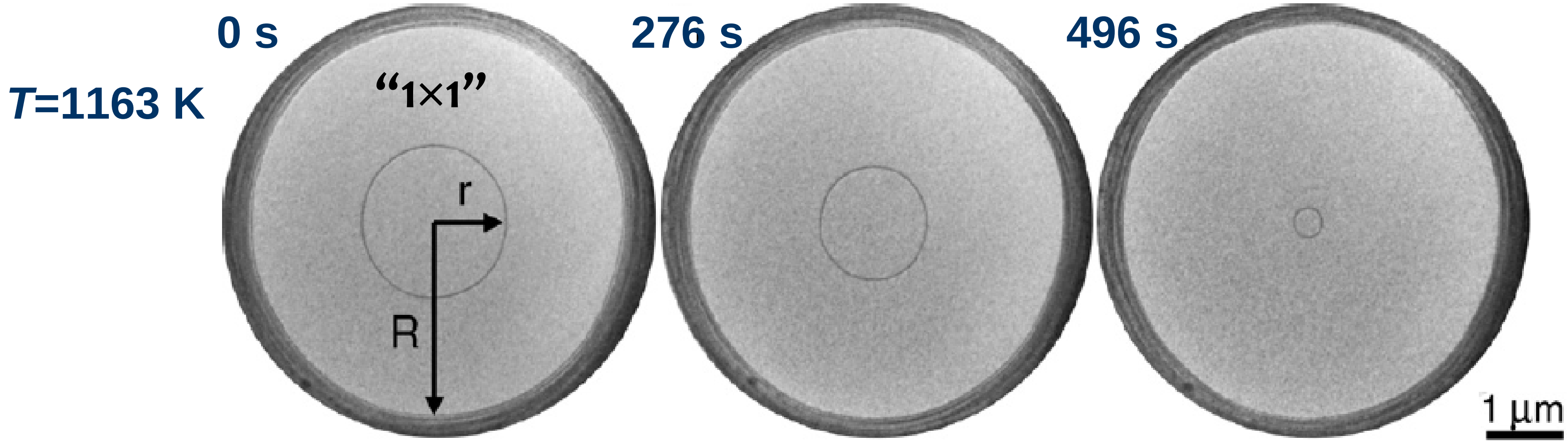
Concave function

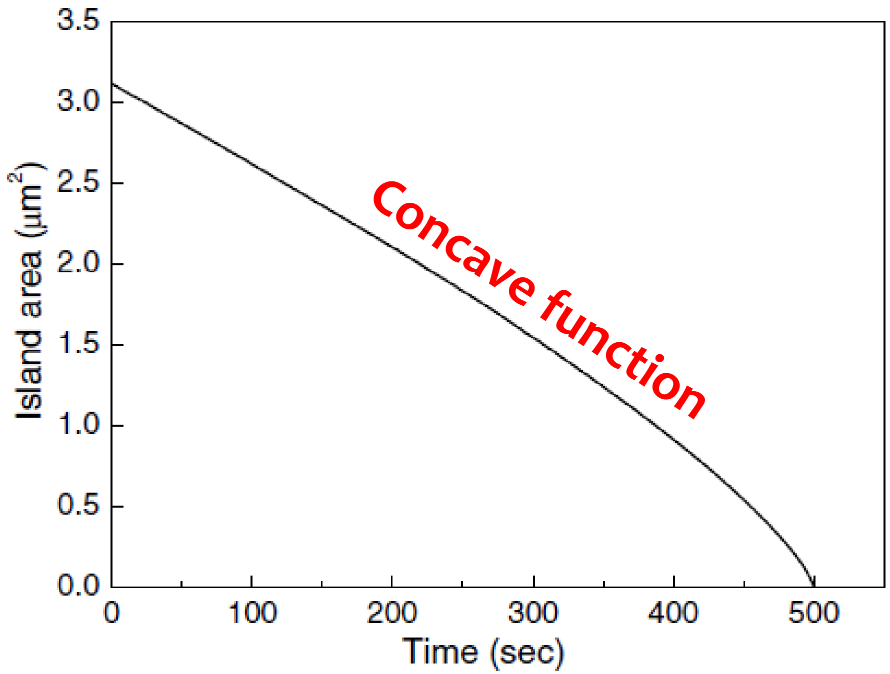
Attachment/detachment limited: $r << \frac{D}{K_+} = d_+ \quad r << R$

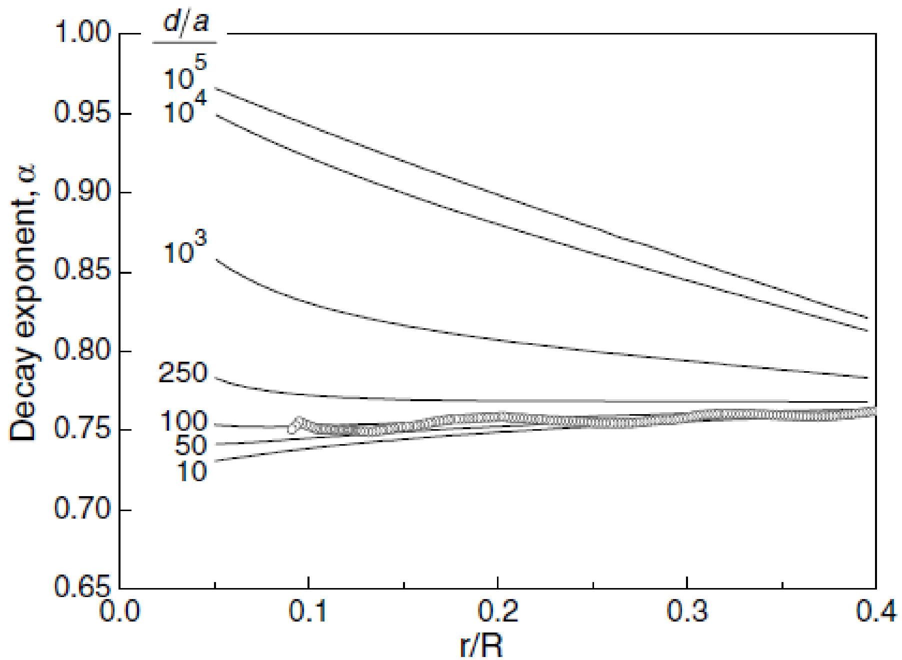
$$\Rightarrow \frac{dA}{dt} = -\frac{2\pi\Omega^2\tilde{\beta}c_{eq}^{0}D}{k_B Td_+} \Rightarrow A(t) \propto (t_0 - t)$$

Linear function

Decay of 2D islands on Si(111)



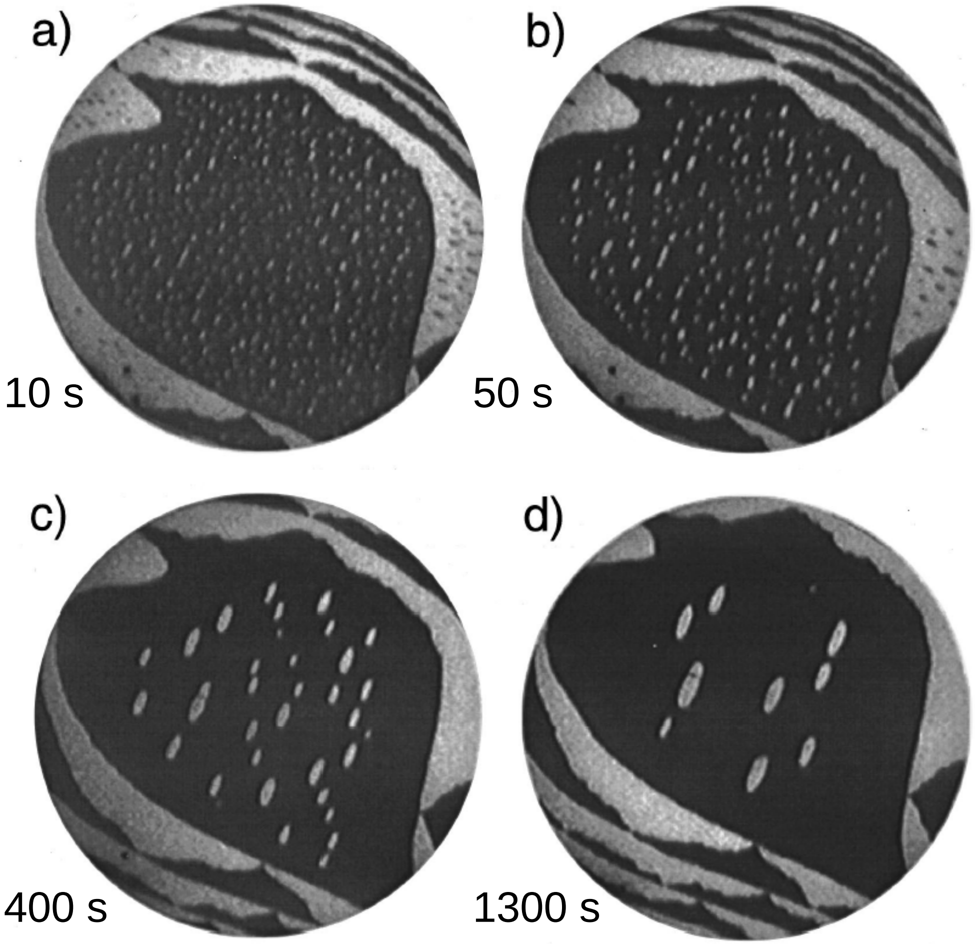




K. L. Man et al., Surf. Sci. 601, 4669 (2007).

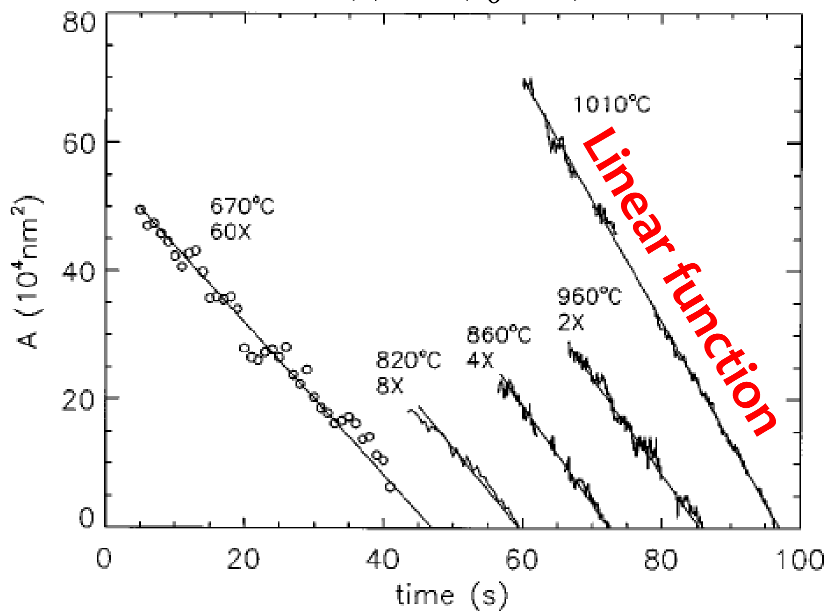
Decay of 2D islands on Si(001)

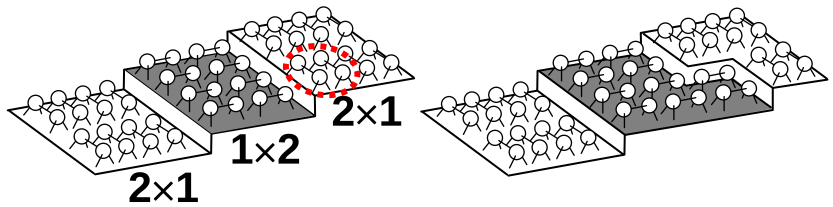
Si(001)-2×1 ➡ Attachment/detachment limited



T=670°C
FOV=5.5 μm

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

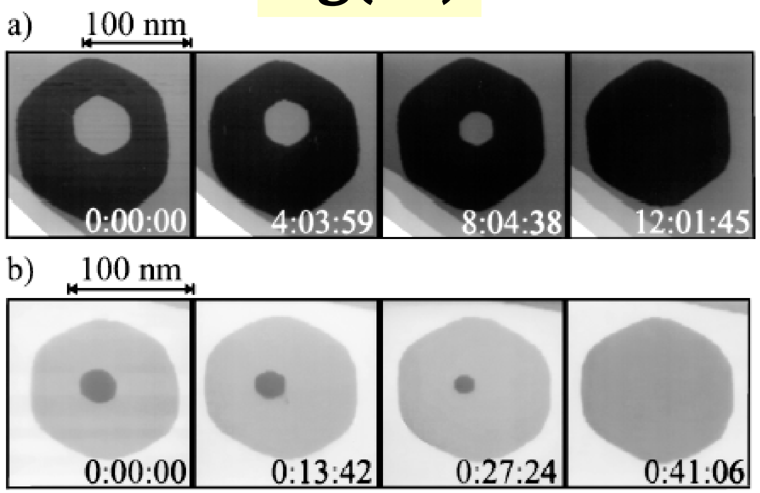


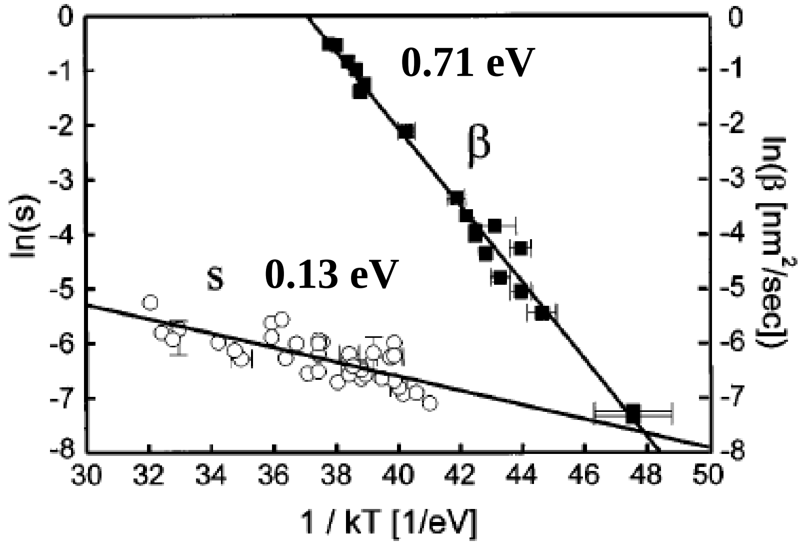


N. C. Bartelt et al., Phys. Rev. B 54, 11741 (1996).

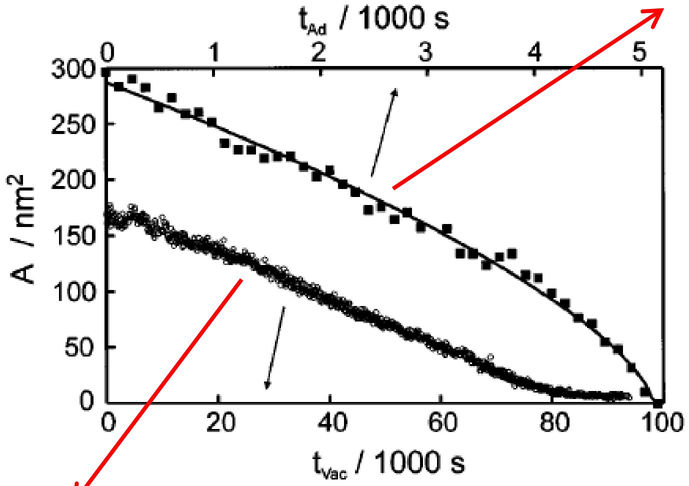
エーリッヒ-シュワーベル効果

Ag(111)





$$\frac{d(\pi r^2)}{dt} = -\beta\left[\frac{a}{r} + \ln(R/r) + \frac{a}{R}\right]^{-1} \times \left[\exp\left(\frac{\gamma}{kTnr}\right) - \exp\left(-\frac{\gamma}{kTnR}\right)\right]$$



$$\beta = 2\pi\Omega D c_{eq}^{0} / K$$

$$s = \nu_S / \nu_0 \exp(-E_S / k_B T)$$

$$\frac{d(\pi r^2)}{dt} = \beta\left[\frac{a}{sr} + \ln(R/r) + \frac{a}{R}\right]^{-1} \times \left[\exp\left(-\frac{\gamma}{kTnr}\right) - \exp\left(-\frac{\gamma}{kTnR}\right)\right]$$

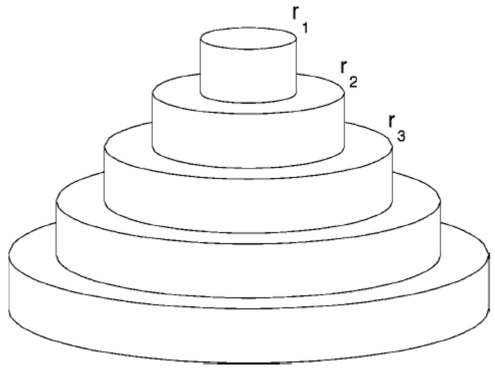
K. Morgenstern et al., Phys. Rev. Lett. 80, 556 (1998).

マウンドの形状変化（理論）

$$\dot{\rho}_i \equiv \frac{d\rho_i}{d\tau} = \frac{a_i - a_{i-1}}{\rho_i}, \qquad \text{with}$$

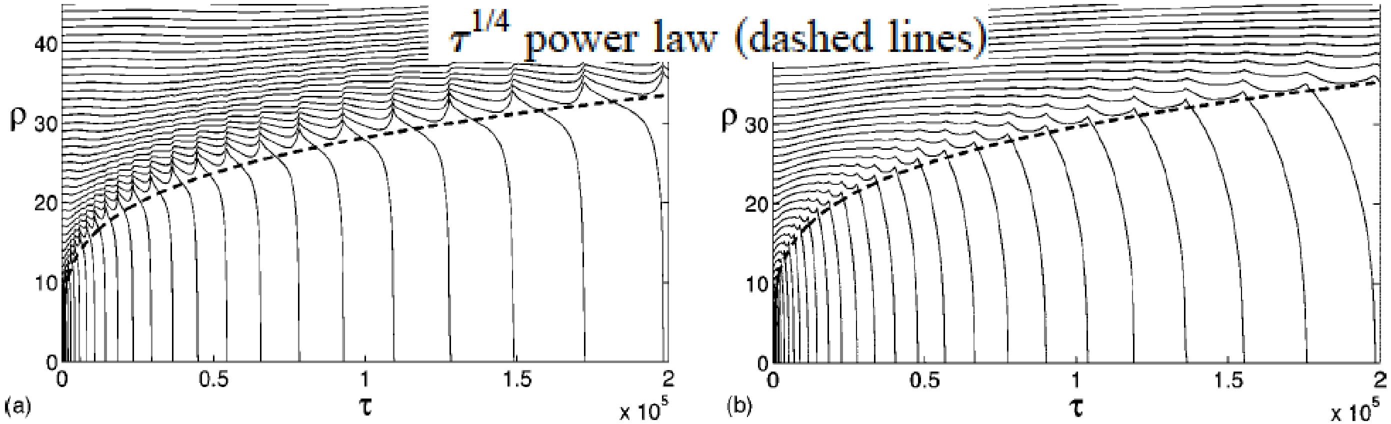
$$a_i = \frac{\frac{1}{\rho_i} - \frac{1}{\rho_{i+1}} + 2g\left(\xi_i - \frac{\rho_{i-1}}{\rho_i+\rho_{i-1}}\xi_{i-1} - \frac{\rho_{i+2}}{\rho_{i+2}+\rho_{i+1}}\xi_{i+1}\right)}{(1-q)\ln\frac{\rho_i}{\rho_{i+1}} - q\left(\frac{1}{\rho_i} + \frac{1}{\rho_{i+1}}\right)},$$

$$\xi_i = (\rho_{i+1} - \rho_i)^{-3},$$



付着・脱離律速　　拡散律速

Cone-shaped mound

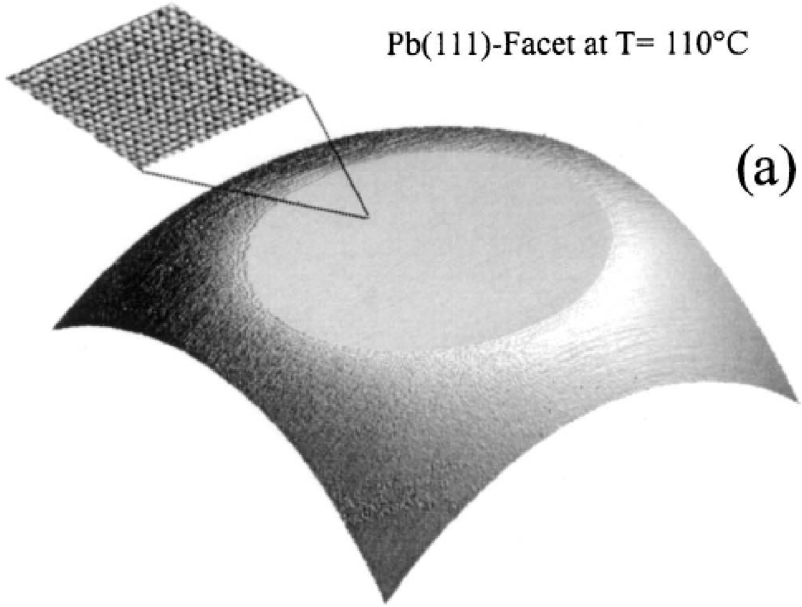


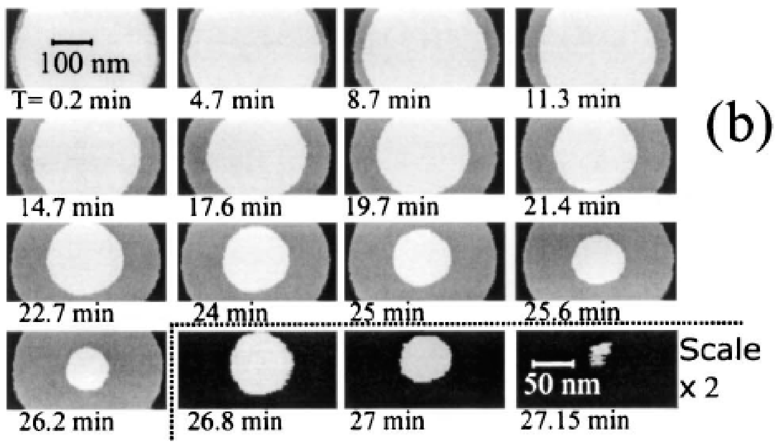
N. Israeli and D. Kandel, Phys. Rev. Lett. 80, 3300 (1998); Phys. Rev. B 60, 5946 (1999).

	Initial shape	Kinetics limited	Diffusion limited
$[R(h) = A_1(h_0 - h)]$	Cone	1/4	1/4
$[R(h) = A_2(h_0 - h)^{1/2}]$	Paraboloid	1/6	1/5
	Single layer	1/2	1/3

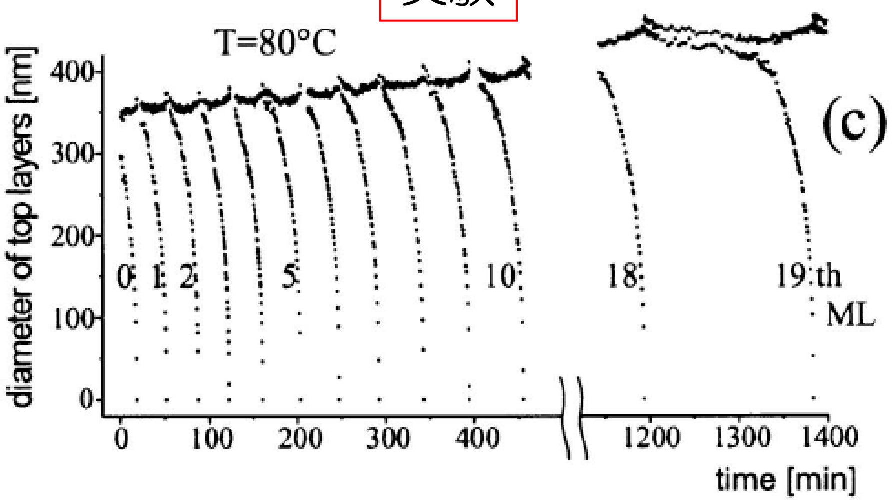
A. Ichimiya et al., Phys. Rev. Lett. 84, 3662 (2000).

マウンドの形状変化（実験）

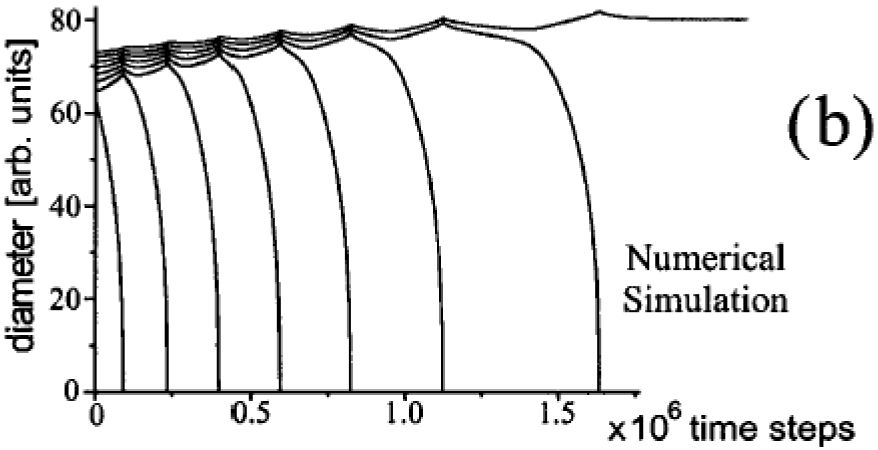




実験



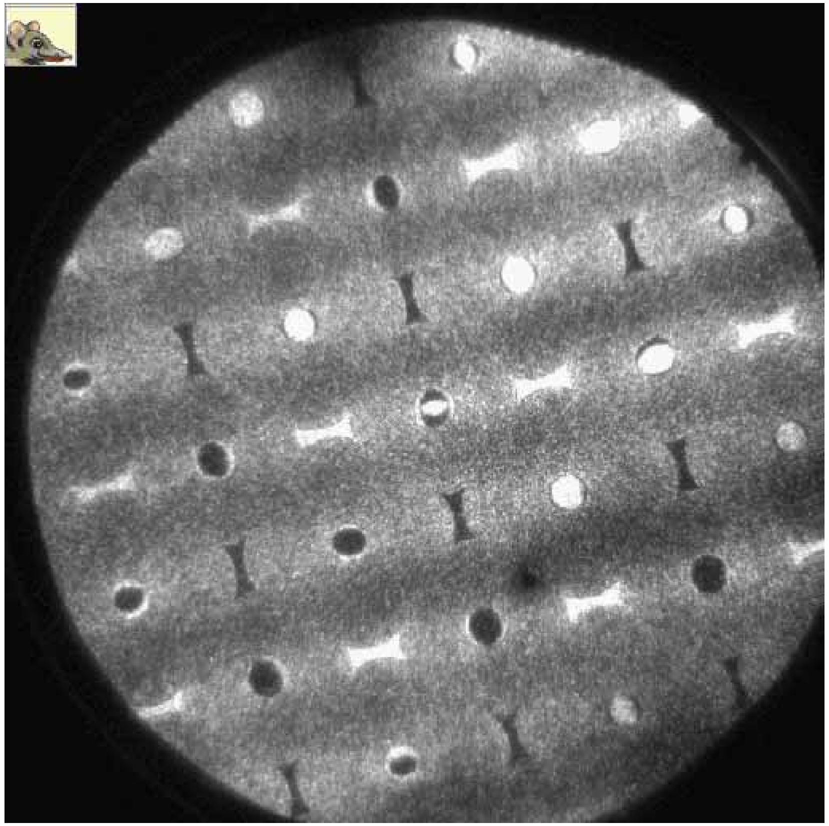
シミュレーション



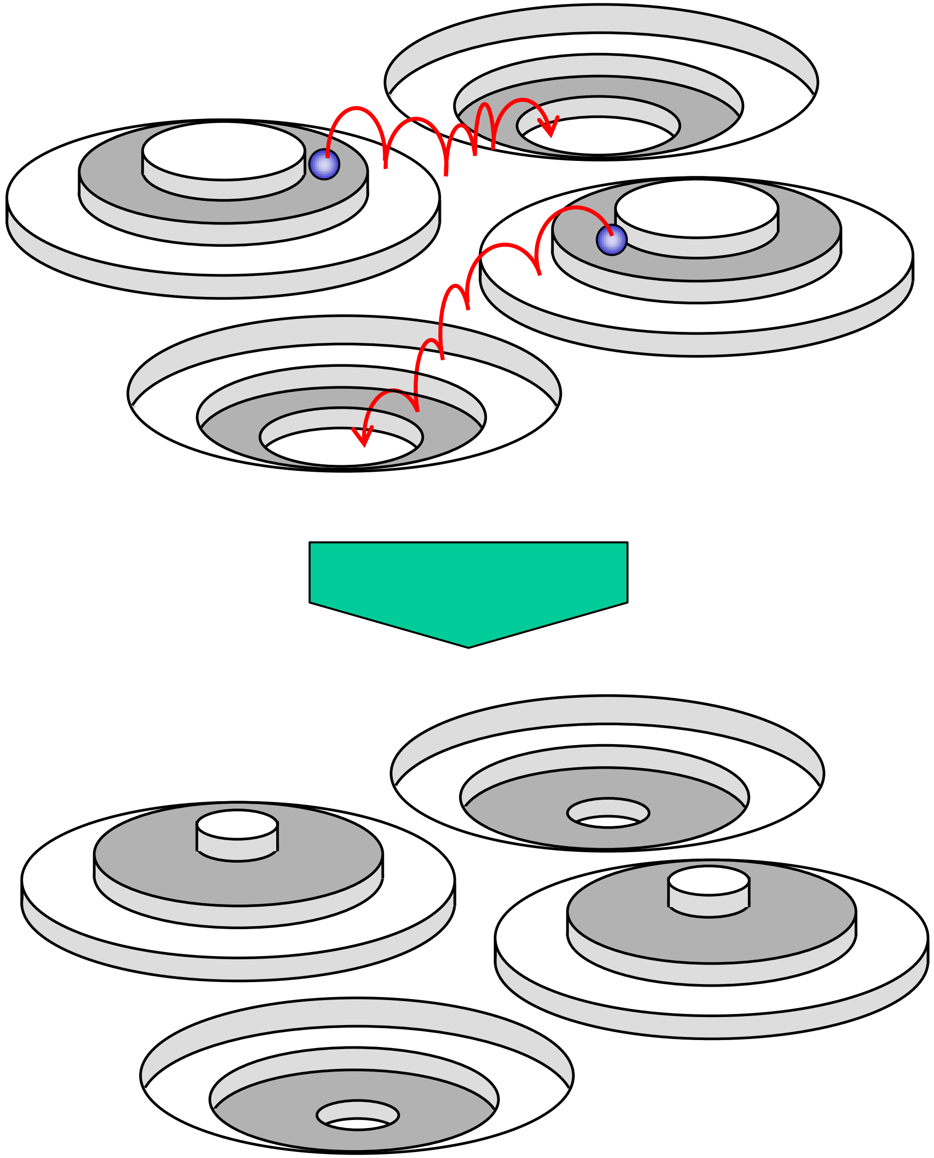
K. Thurmer et al., Phys. Rev. Lett. 87, 186102 (2001).

マウンドの形状変化 on Si(001)

LEEM movie



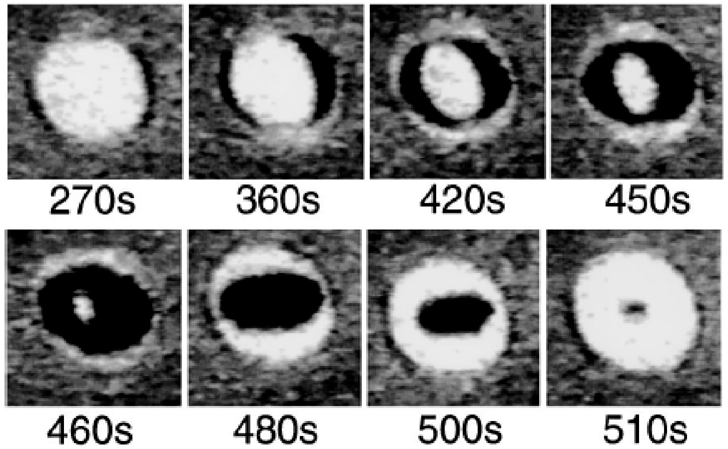
5.7倍速
視野径~8μm



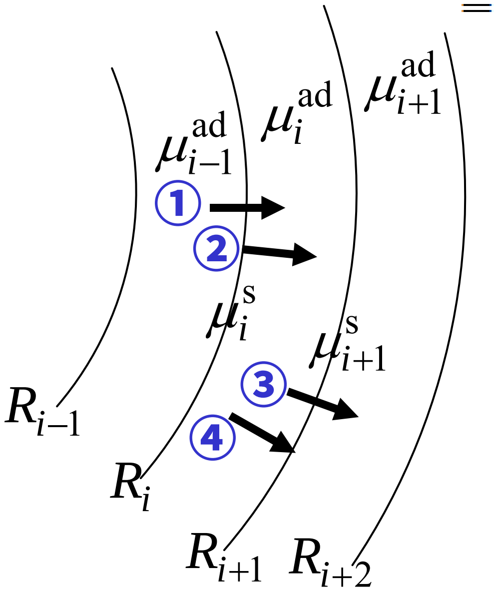
ステップ透過率(permeability)

吸着子がステップを透過する過程

Si(001)

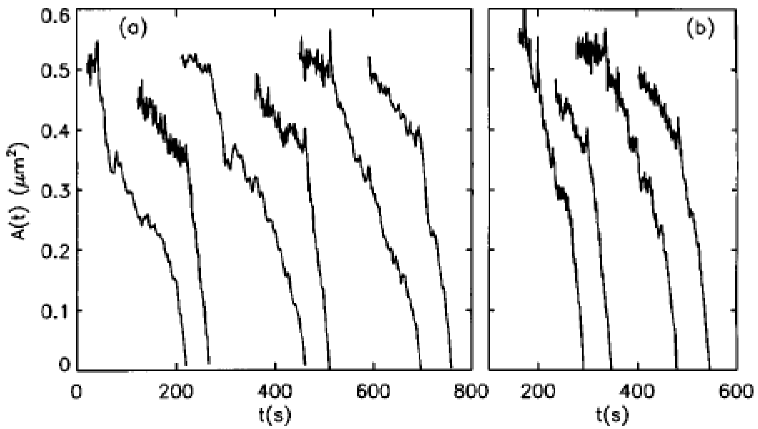


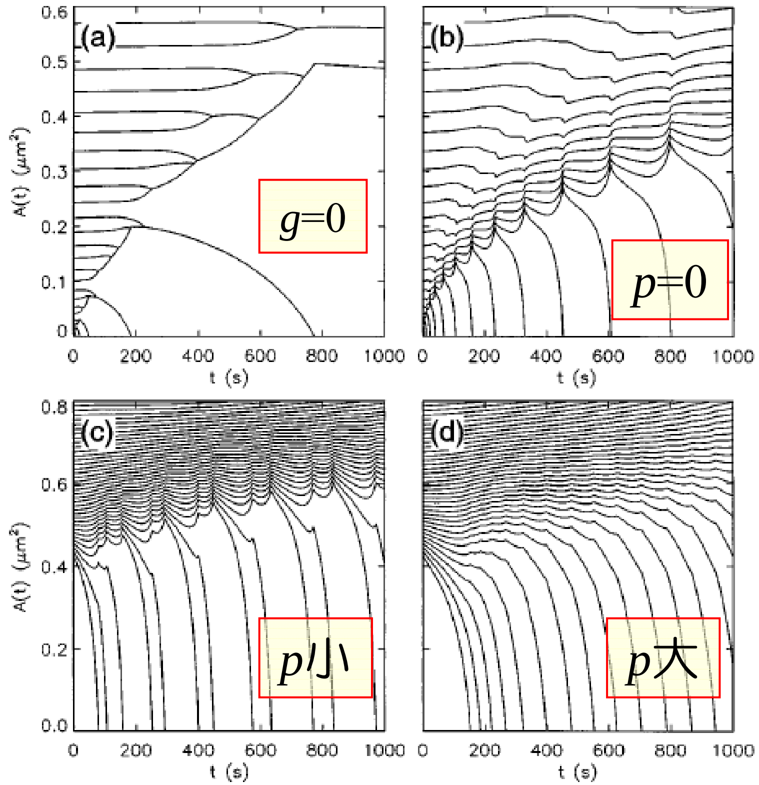
$$① \; 2\pi R_i[p(\mu_{i-1}^{\rm ad} - \mu_i^{\rm ad}) \; + \; \Gamma_-(\mu_i^{\rm s} - \mu_i^{\rm ad})] \; ②$$
$$= 2\pi R_{i+1}[p(\mu_i^{\rm ad} - \mu_{i+1}^{\rm ad}) \; ③$$
$$+ \; \Gamma_+(\mu_i^{\rm ad} - \mu_{i+1}^{\rm s})]. \; ④$$



p：ステップ透過率

S. Tanaka et al., Phys. Rev. Lett. 78, 3342 (1997).





二次元島の拡散

$$g(\theta, t) = [\tilde{r}(\theta, t) - R]/R$$

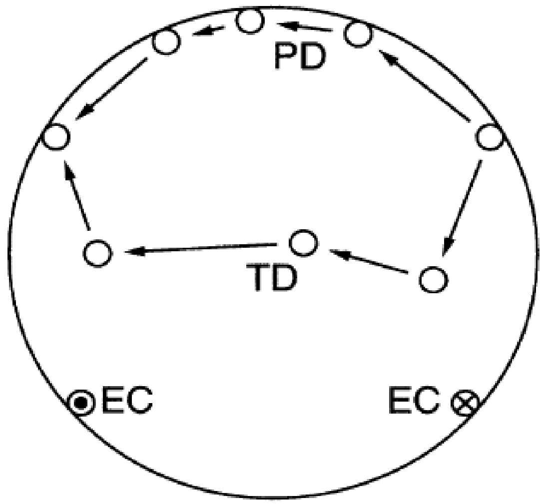
$$D_c \equiv \frac{\langle \vec{\mathbf{r}}^2_{\mathrm{CM}}(t) \rangle}{4t}$$

$$g(\theta, t) = \sum_n g_n(t) \exp(in\theta)$$

$$\frac{\partial g_n(t)}{\partial t} = -\tau_n^{-1} g_n(t) + \zeta_n(t)$$

$$\langle |g_n(t)|^2 \rangle = k_B T / 2\pi \tilde{\beta} R n^2$$

$$D_c = k_B T R / \pi \tilde{\beta} \tau_1$$



Edge diffusion

$$\tau_n^{-1} = D_{\mathrm{st}} c_{\mathrm{st}} \Omega^2 \tilde{\beta} n^4 / k_B T R^4$$

Terrace diffusion

$$\tau_n^{-1} = 2 D_{\mathrm{su}} c_{\mathrm{su}} \Omega^2 \tilde{\beta} |n|^3 / k_B T R^3$$

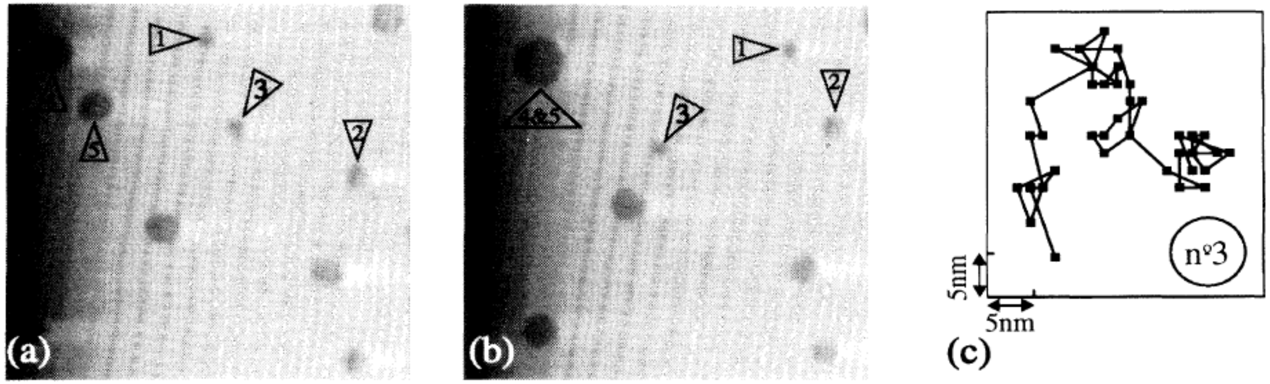
Attachment/detachment

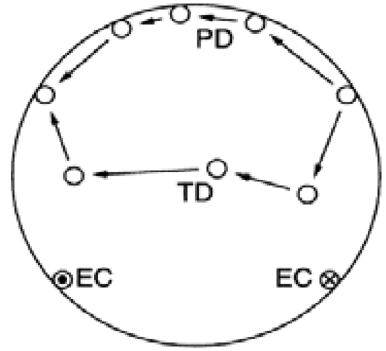
$$\tau_n^{-1} = \Gamma \tilde{\beta} n^2 / k_B T R^2$$

S. V. Khare et al., PRL 75, 2148 (1995).

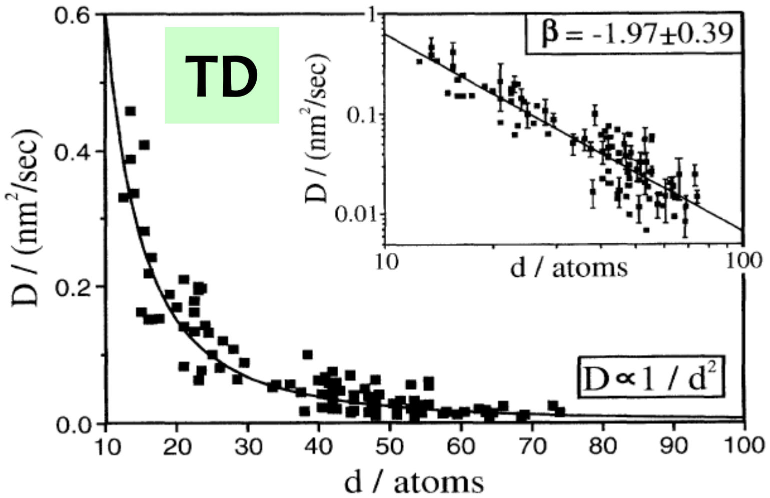
二次元島の拡散の例

Vacancy islands on Ag(111)



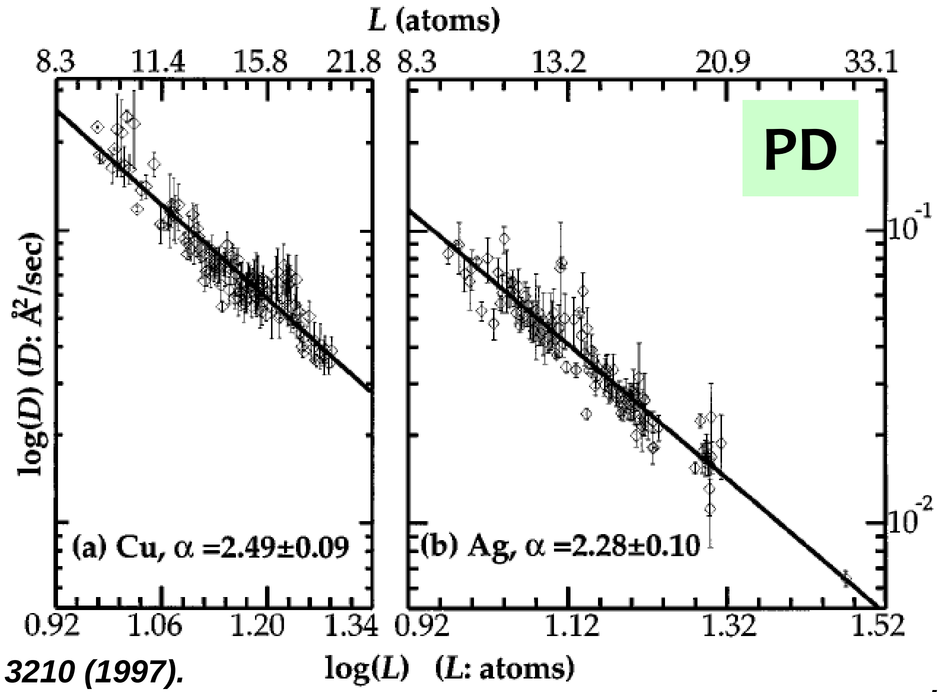


S. V. Khare et al., PRL 75, 2148 (1995).



K. Morgenstern et al., Phys. Rev. Lett. 74, 2058 (1995).

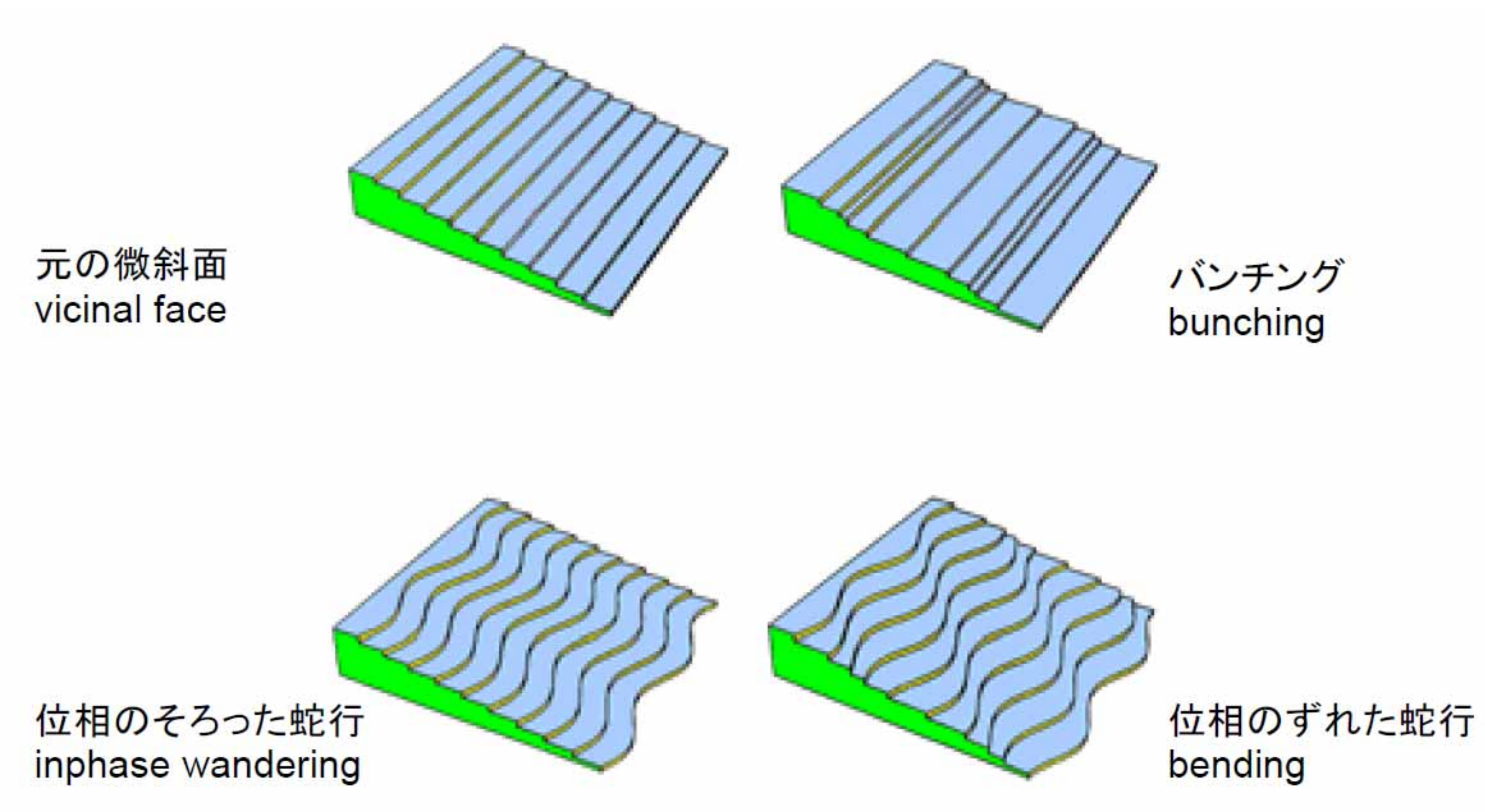
Islands on Cu(100) and Ag(100)



W. W. Pai et al., Phys. Rev. Lett. 79, 3210 (1997).

結晶成長中のステップの不安定化

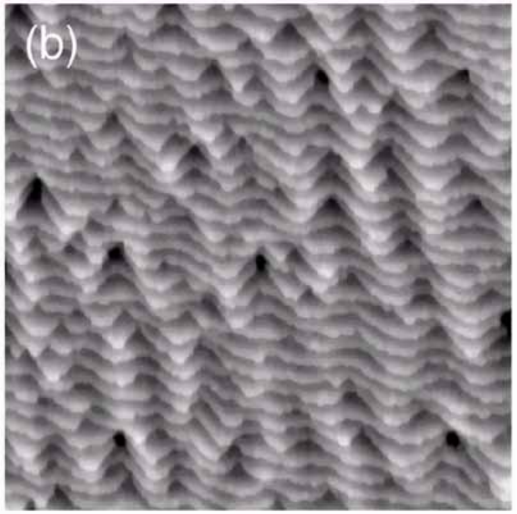
ステップの不安定化



http://www.slab.phys.nagoya-u.ac.jp/uwaha/gairon08.pdf

ステップの蛇行パターン

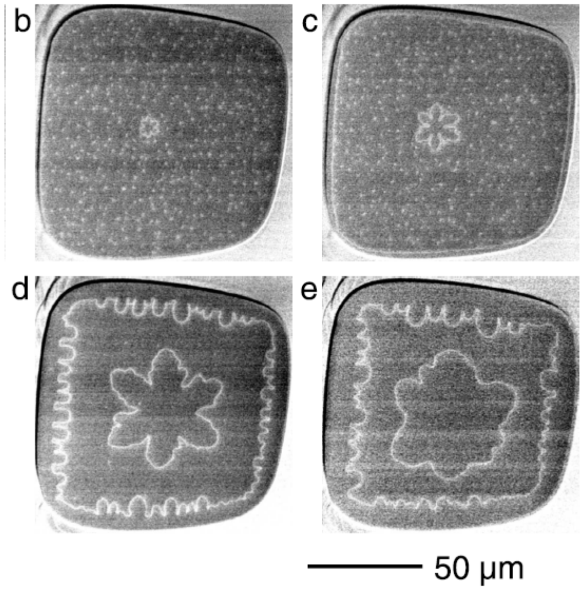
Cu(1,1,17)



Asymmetry in the kinetic coefficient
(Ehrlich-Schwoebel effect)

T. Maroutian L. Douillard, and H.-J. Ernst, Phys. Rev. Lett. 83, 4353 (1999).

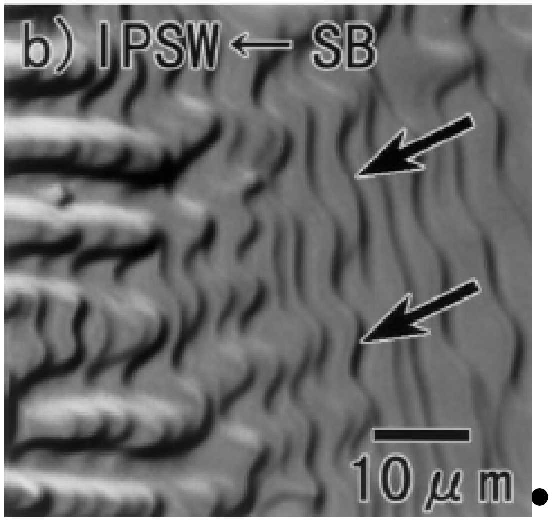
超平坦Si(111)



Asymmetry in the terrace size

Y. Homma, P. Finnie, and M. Uwaha, Surf. Sci. 492, 125 (2001).

Si(111)-"1×1"

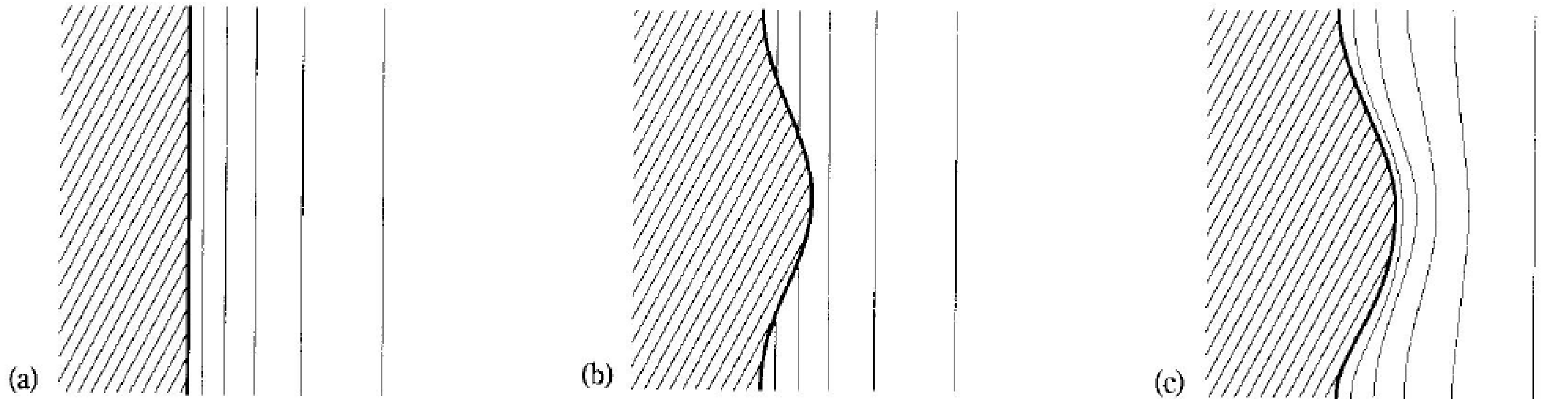


Step-down current at 950°C

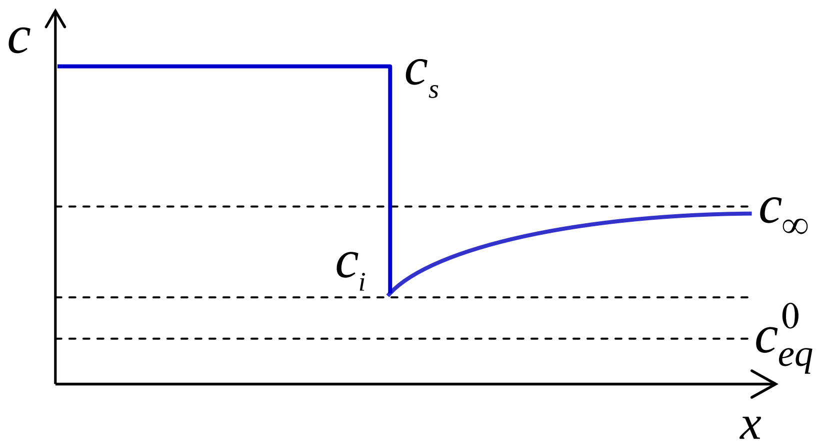
K. Yagi et al., Surf. Sci. Rep. 43, 45 (2001).

Mullins-Sekerka不安定性

拡散場中での成長界面の不安定化

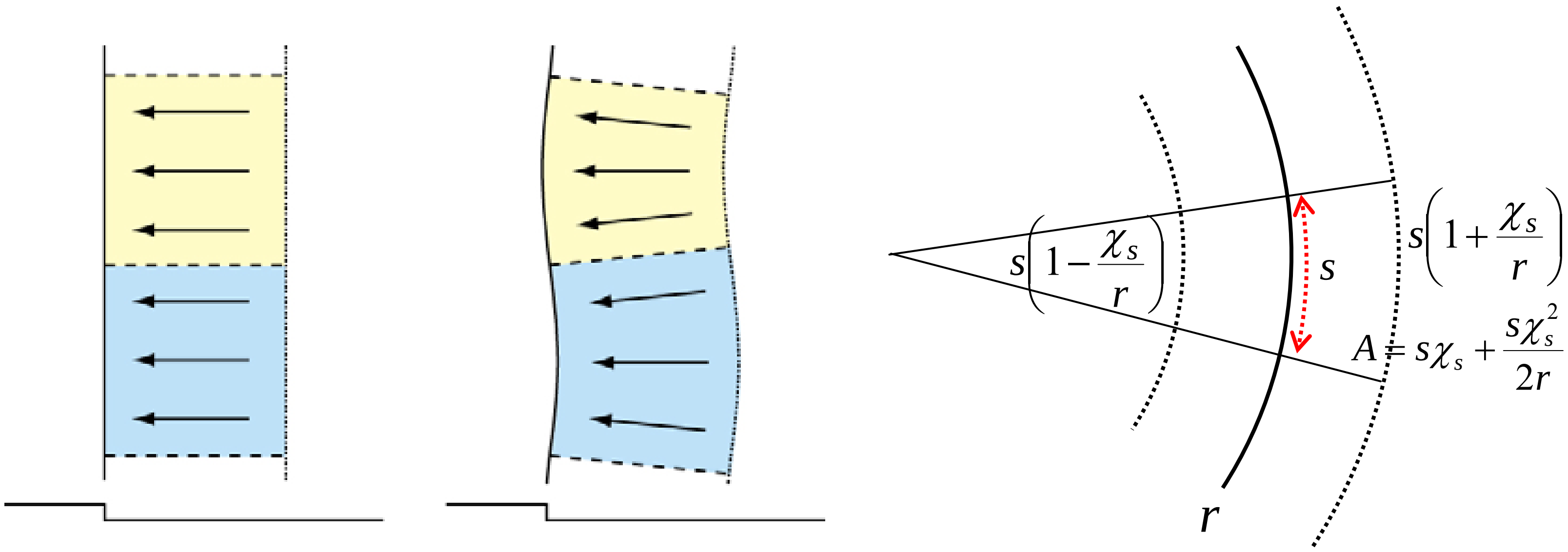


上羽牧夫, シリーズ「結晶成長のダイナミクス」第2 巻「結晶成長のしくみを探る—その物理的基礎」(共立出版, 東京, 2002).



成長中の直線ステップの不安定化

孤立ステップ＋片側モデル



$$v_0 = (F - F_{eq})\Omega\chi_s$$

$$\delta v_d = (F - F_{eq})\Omega \cdot \frac{\chi_s^2}{2r}$$

$$\delta v_s = -F_{eq}\frac{\Omega\tilde{\beta}}{k_B T r}\Omega\chi_s$$

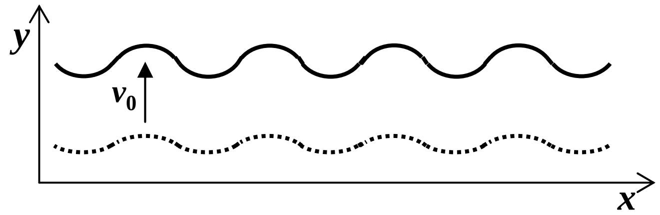
$$\delta v_d + \delta v_s = (F_c - F_{eq})\Omega \cdot \frac{\chi_s^2}{2r} - F_{eq}\frac{\Omega\tilde{\beta}}{k_B T r}\Omega\chi_s = 0$$

臨界フラックス：$F_c = F_{eq}\left(1 + \frac{2\Omega\tilde{\beta}}{\chi_s k_B T}\right)$

Yukio Saito, "Statistical Physics of Crystal Growth", (World Scientific, 1996).

直線ステップの不安定化(線形安定性)

線形安定性理論（片側モデル）



ステップ位置： $\zeta(x,t) = v_0 t + \delta\zeta_q e^{iqx+\omega_q t}$

吸着子濃度： $c(x,y;t) = c_\infty + (c_{eq}^0 - c_\infty) e^{-(y-v_0 t)/x_s} + \delta c_q e^{iqx - \Lambda_q (y - v_0 t) + \omega_q t}$

$$v_0 = \Omega \frac{D}{x_s}\left(c_\infty - c_{eq}^0\right)$$

拡散方程式 $\left(\nabla^2 - \frac{1}{x_s^2}\right)(c - c_\infty) = 0$ を満足するためには：

$$\nabla^2 c = \frac{\left(c_{eq}^0 - c_\infty\right)}{x_s^2} e^{-(y-v_0 t)/x_s} + \delta c_q \left(-q^2 + \Lambda_q^2\right) e^{iqx - \Lambda_q (y - v_0 t) + \omega_q t}$$

$$-)\quad \frac{(c - c_\infty)}{x_s^2} = \frac{\left(c_{eq}^0 - c_\infty\right)}{x_s^2} e^{-(y-v_0 t)/x_s} + \frac{\delta c_q}{x_s^2} e^{iqx - \Lambda_q (y - v_0 t) + \omega_q t}$$

$$\Lambda_q = \sqrt{q^2 + x_s^{-2}}$$

Yukio Saito, "Statistical Physics of Crystal Growth", (World Scientific, 1996).

直線ステップの不安定化(線形安定性)

線形安定性理論

ギブス-トムソン効果： $c_{step} = c_{eq}^0\left(1+\frac{\Omega\tilde{\beta}}{k_BTR}\right) = c_{eq}^0\left(1-\frac{\Omega\tilde{\beta}}{k_BT}\frac{\partial^2 y}{\partial x^2}\right)$

$$c\left(v_0 t + \delta\zeta_q e^{iqx+\omega_q t}\right) = c(\zeta_0 + \delta\zeta) \approx c_0(\zeta_0) + \left.\frac{\partial c_0}{\partial y}\right|_{\zeta_0}\delta\zeta + \delta c(\zeta_0)$$

$$\delta c(x,y;t) = \delta c_q e^{iqx-\Lambda_q(y-v_0 t)+\omega_q t}$$

$$\delta\zeta(x,t) = \delta\zeta_q e^{iqx+\omega_q t}$$

$$= c_{eq}^0 - \frac{c_{eq}^0 - c_\infty}{x_s}\delta\zeta_q e^{iqx+\omega_q t} + \delta c_q e^{iqx+\omega_q t}$$

$$c_{step} = c_{eq}^0\left(1-\frac{\Omega\tilde{\beta}}{k_BT}\frac{\partial^2 y}{\partial x^2}\right) = c_{eq}^0 + \Gamma q^2\delta\zeta_q e^{iqx+\omega_q t}, \text{ where } \Gamma = \frac{c_{eq}^0\Omega\tilde{\beta}}{k_BT}$$

$$\delta c_q = -\delta\zeta_q\left(\frac{c_\infty - c_{eq}}{x_s} - \Gamma q^2\right)$$

Yukio Saito, “Statistical Physics of Crystal Growth”, (World Scientific, 1996).

直線ステップの不安定化(線形安定性)

$$v_n = \frac{\partial\zeta/\partial t}{\sqrt{1+(\partial\zeta/\partial x)^2}} \approx \frac{\partial\zeta}{\partial t} = v_0 + \omega_q \delta\zeta_q e^{iqx+\omega_q t}$$

$$v_n = D\Omega(\hat{n}\cdot\nabla c)_+ \approx D\Omega \left.\frac{\partial c}{\partial y}\right|_+ = D\Omega\left[\frac{c_\infty - c_{eq}}{x_s} e^{-(\zeta - v_0 t)/x_s} - \Lambda_q \delta c_q e^{iqx-(\zeta-v_0 t)/x_s + \omega_q t}\right]$$

$\zeta - v_0 t = \delta\zeta_q e^{iqx+\omega_q t}$ を代入して展開

$$\approx \frac{D\Omega}{x_s}(c_\infty - c_{eq}^0)\left(1 - \frac{\delta\zeta_q}{x_s} e^{iqx+\omega_q t}\right) + D\Omega\Lambda_q\delta\zeta_q\left(\frac{c_\infty - c_{eq}^0}{x_s} - \Gamma q^2\right) e^{iqx+\omega_q t}$$

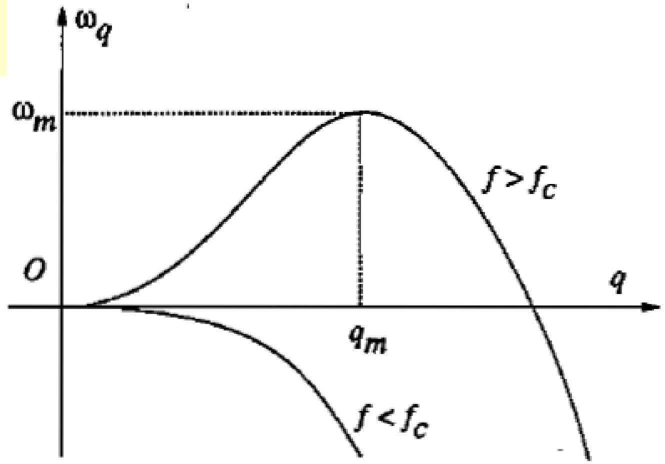
$$\omega_q = -D\Omega\left[\frac{c_\infty - c_{eq}}{x_s^2} - \Lambda_q\left(\frac{c_\infty - c_{eq}}{x_s} - \Gamma q^2\right)\right] = v_0\left(\Lambda_q - x_s^{-1}\right) - D\Omega\Gamma\Lambda_q q^2$$

$$\Lambda_q = \sqrt{q^2 + x_s^{-2}}$$

Bales-Zangwill instability

$$\omega_q \approx \left(\frac{1}{2}v_0 - \frac{D\Omega\Gamma}{x_s}\right)q^2 - \left(\frac{1}{8}v_0 x_s^3 + \frac{1}{2}D\Omega\Gamma x_s\right)q^4$$

$$f_c = f_{eq}\left(1 + \frac{2\Omega\tilde{\beta}}{x_s k_B T}\right) \qquad \left(\Gamma = \frac{c_{eq}^0 \Omega\tilde{\beta}}{k_B T}\right)$$



Yukio Saito, "Statistical Physics of Crystal Growth", (World Scientific, 1996).

直線ステップの不安定化(線形安定性)

$$\omega_q = -D\Omega\left[\frac{c_\infty - c_{eq}}{x_s^2} - \Lambda_q\left(\frac{c_\infty - c_{eq}}{x_s} - \Gamma q^2\right)\right] = v_0\left(\Lambda_q - x_s^{-1}\right) - D\Omega\Gamma\Lambda_q q^2$$

$$\left(\Lambda_q = \sqrt{q^2 + x_s^{-2}}\right)$$

蒸発が無視できると、$x_s \to \infty, \Lambda_q \to q$ のため、

$$\omega_q = v_0 q - D\Omega\Gamma q^3$$

$$\Rightarrow \quad q_{\max} = \sqrt{\frac{v_0}{3D\Omega\Gamma}} = \sqrt{\frac{k_B T v_0}{3D\Omega^2 c_{eq}^0 \tilde{\beta}}}$$

最も不安定なモードの波長は、入射強度あるいは
前進速度の1/2乗に反比例する。

直線ステップの不安定化(非線形効果)

摂動展開による非線形効果の解析:

$$\varepsilon = \frac{f - f_c}{f_c - f_{eq}} = \frac{v_{st}^0}{v_{stc}} - 1$$

$$q_{\max} \sim \varepsilon^{1/2}$$

$$\omega(q_{\max}) \sim \varepsilon q^2 \sim \varepsilon^2$$

逓減摂動法:

$$X = \sqrt{\varepsilon}\frac{x}{x_s}, \quad Z = \frac{\zeta}{x_s}, \quad T = \varepsilon^2 \frac{t}{\tau}$$

$$\frac{\zeta}{x_s} = \varepsilon H = \varepsilon H_0 + \varepsilon^2 H_1 + \cdots$$

$$u = \Omega(c - c_\infty) = u_0 + \varepsilon u_1 + \varepsilon^2 u_2 + \cdots$$

直線ステップの不安定化(非線形効果)

拡散方程式

$$\left(\nabla^2 - \frac{1}{x_s^2}\right)(c - c_\infty) = 0 \longrightarrow \varepsilon u_{XX} + u_{ZZ} - u = 0$$

質量保存

$$v_{st} = \Omega j_{s+} = -\Omega D\vec{n} \cdot \nabla c(x_{st+}) \longrightarrow V_c(1+\varepsilon) + \varepsilon^3 H_T = u_Z - \varepsilon^2 u_X H_X$$

$$v_{st} = \frac{\partial\zeta / \partial t}{\sqrt{1 + (\partial\zeta / \partial x)^2}} \qquad \vec{n} = \frac{(-\partial\zeta / \partial x, 1)}{\sqrt{1 + (\partial\zeta / \partial x)^2}}$$

平衡濃度

$$c_{st} = c_{eq}^0\left(1 + \frac{\tilde{\beta}\Omega}{k_B T}\kappa\right) \longrightarrow u = -V_c(1+\varepsilon) - \frac{V_c}{2}\frac{\varepsilon^2 H_{XX}}{\left[1 + \varepsilon^3 (H_X)^2\right]^{3/2}}$$

$$\kappa = -\frac{\partial^2\zeta / \partial x^2}{\left[1 + (\partial\zeta / \partial x)^2\right]^{3/2}}$$

where $V_c = \frac{v_{stc}\tau}{x_s} = \frac{2\Omega^2 f_{eq}\tilde{\beta}}{k_B T}\frac{\tau}{x_s}$

直線ステップの不安定化(非線形効果)

Order ε^0

$$\varepsilon u_{XX} + u_{ZZ} - u = 0 \longrightarrow u_{0ZZ} - u_0 = 0 \longrightarrow u_0 = A_0 e^{-Z}$$

$$V_c(1+\varepsilon) + \varepsilon^3 H_T = u_Y - \varepsilon^2 u_X H_X \longrightarrow u_{0Z} = V_c$$

$$u = -V_c(1+\varepsilon) - \frac{V_c}{2}\frac{\varepsilon^2 H_{XX}}{\left[1+\varepsilon^3 (H_X)^2\right]^{3/2}} \longrightarrow u_0 = -V_c$$

$$\Rightarrow A_0 = -V_c$$

Order ε^1

$$\varepsilon u_{XX} + u_{ZZ} - u = 0 \longrightarrow u_{1ZZ} - u_1 = 0 \longrightarrow u_1 = A_1 e^{-Z}$$

$$V_c(1+\varepsilon) + \varepsilon^3 H_T = u_Z - \varepsilon^2 u_X H_X$$

$$\longrightarrow V_c + \varepsilon V_c = u_{0Z} + \varepsilon u_{1Z} = u_{0Z} + \varepsilon H_0 u_{0ZZ} + \varepsilon u_{1Z}$$

$$\longrightarrow V_c = H_0 u_{0ZZ} + u_{1Z}$$

$$u = -V_c(1+\varepsilon) - \frac{V_c}{2}\frac{\varepsilon^2 H_{XX}}{\left[1+\varepsilon^3 (H_X)^2\right]^{3/2}} \longrightarrow u_1 + H_0 u_{0Z} = -V_c$$

$$\Rightarrow \frac{A_1}{A_0} = 1 + H_0(X,T)$$

直線ステップの不安定化(非線形効果)

Order ε^2

$$\varepsilon u_{XX} + u_{ZZ} - u = 0 \longrightarrow u_{2ZZ} - u_2 = -A_{1XX} e^{-Z}$$

$$\longrightarrow u_2 = A_2 e^{-Z} + \frac{ZA_{1XX}}{2} e^{-Z}$$

$$V_c(1+\varepsilon) + \varepsilon^3 H_T = u_Z - \varepsilon^2 u_X H_X$$

$$\longrightarrow H_1 u_{0ZZ} + \frac{1}{2} H_0^2 u_{0ZZZ} + H_0 u_{1ZZ} + u_{2Z} = 0$$

$$u = -V_c(1+\varepsilon) - \frac{V_c}{2} \frac{\varepsilon^2 H_{XX}}{\left[1 + \varepsilon^3 (H_X)^2\right]^{3/2}}$$

$$\longrightarrow H_1 u_{0Z} + \frac{1}{2} H_0^2 u_{0ZZ} + H_0 u_{1Z} + u_2 + \frac{V_c}{2} H_{0XX} = 0$$

$$\Longrightarrow \frac{A_2}{A_0} = H_0 + \frac{1}{2} H_0^2 + \frac{1}{2} H_{0XX} + H_1$$

直線ステップの不安定化(非線形効果)

Order ε^3

$$\varepsilon u_{XX} + u_{ZZ} - u = 0 \longrightarrow u_{3ZZ} - u_3 = -\left[A_{2XX} + \frac{ZA_{1XXXX}}{2}\right]e^{-Z}$$

$$\longrightarrow u_3 = A_3 e^{-Z} + \frac{ZA_{2XX}}{2}e^{-Z} + \frac{A_{1XXXX}}{8}\left(Z + Z^2\right)e^{-Z}$$

$$V_c(1+\varepsilon) + \varepsilon^3 H_T = u_Z - \varepsilon^2 u_X H_X$$

$$u = -V_c(1+\varepsilon) - \frac{V_c}{2}\frac{\varepsilon^2 H_{XX}}{\left[1+\varepsilon^3 (H_X)^2\right]^{3/2}}$$

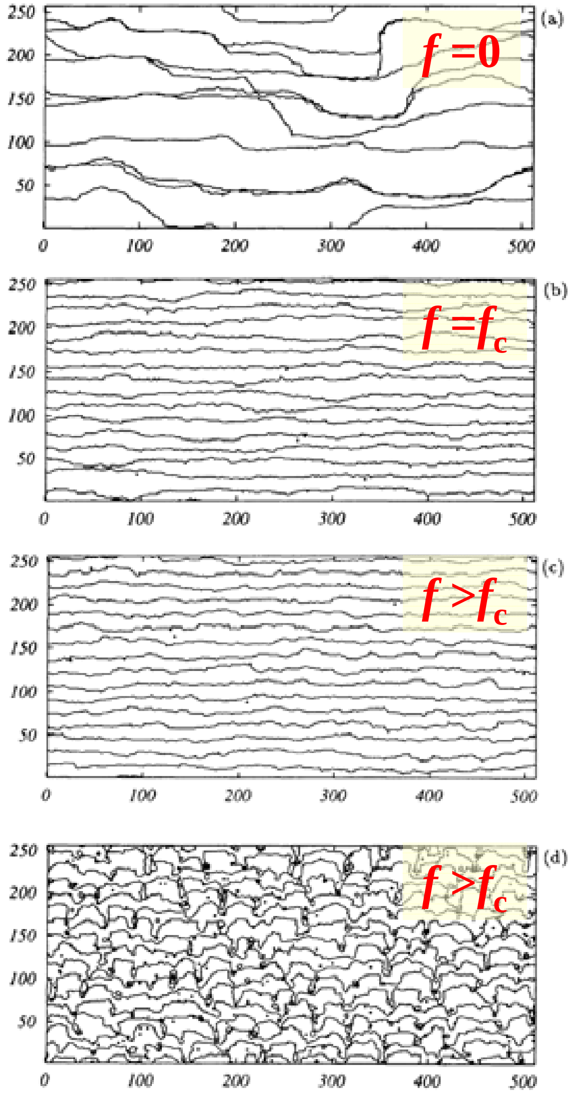
$$\longrightarrow \frac{A_3}{A_0} = \frac{1}{2}H_0^2 + \frac{1}{6}H_0^3 + H_1 + H_0 H_1 + \frac{1}{2}H_{1XX} + H_2$$

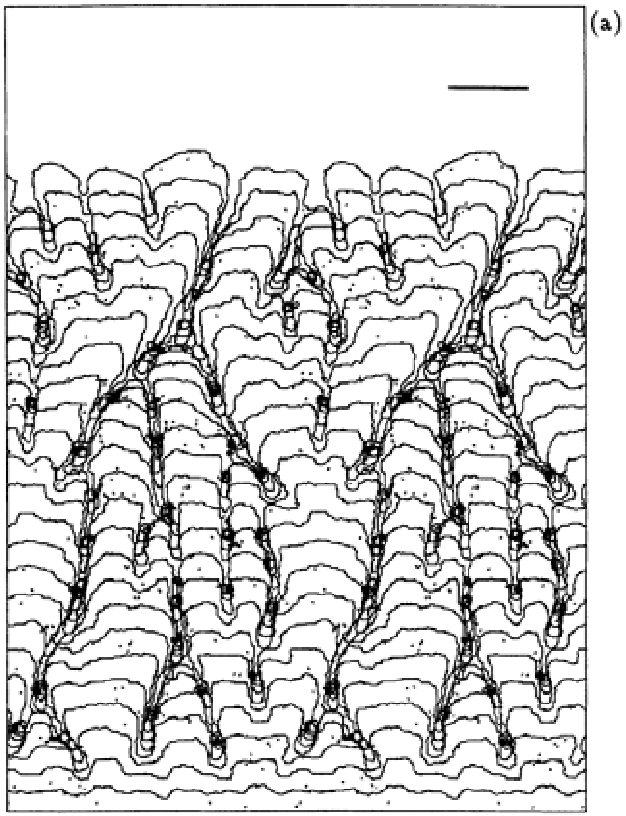
$$\longrightarrow \frac{1}{V_c}\frac{\partial H_0}{\partial T} = -\frac{1}{2}\frac{\partial^2 H_0}{\partial X^2} - \frac{3}{8}\frac{\partial^4 H_0}{\partial X^4} + \frac{1}{2}\left(\frac{\partial H_0}{\partial X}\right)^2$$

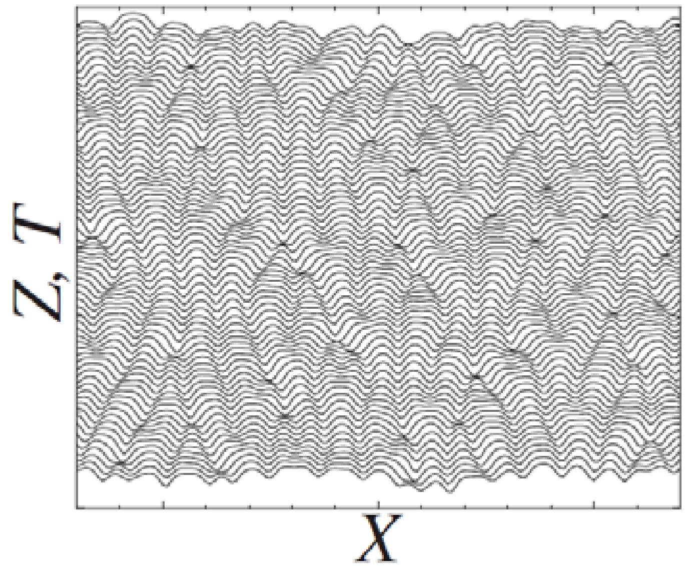
$$\longrightarrow \frac{\partial y}{\partial t} = -\frac{\partial^2 y}{\partial x^2} - \frac{\partial^4 y}{\partial x^4} - \frac{1}{2}\left(\frac{\partial y}{\partial x}\right)^2$$

Kuramoto-Sivashinsky equation

直線ステップの不安定化(非線形効果)







モンテカルロシミュレーション

Y. Saito and Y. Uwaha, PRB 49, 10677 (1992).

KS方程式の数値解

C. Misbah et al., Rev. Mod. Phys. 82 981 (2010).

成長中(保存系)のステップ列の不安定化

ステップ列、位相のそろった蛇行、保存系

Linear stability analysis

$$\omega_k = \frac{\varepsilon D}{2} k^2 - D\Gamma l k^4 \equiv \alpha k^2 - \beta k^4 \quad \Rightarrow \quad k_{\max} = \sqrt{\alpha/\beta}$$

$$\varepsilon = \frac{\Omega F l^2}{D} << 1 \qquad \Gamma = \frac{\tilde{\beta}\Omega^2 c_{eq}^0}{k_B T}$$

$$X = \sqrt{\varepsilon} x, \quad \zeta = H/\sqrt{\varepsilon}, \quad T = \varepsilon^2 t$$

Nonlinear equation

$$\partial_t \zeta = -\partial_x \left[\frac{\alpha \partial_x \zeta}{1 + (\partial_x \zeta)^2} + \frac{\beta}{1 + (\partial_x \zeta)^2} \times \partial_x \left\{ \frac{\partial_{xx} \zeta}{\left(1 + (\partial_x \zeta)^2\right)^{3/2}} \right\} \right]$$

Reduction of mobility due to
the crowding of the deformed steps

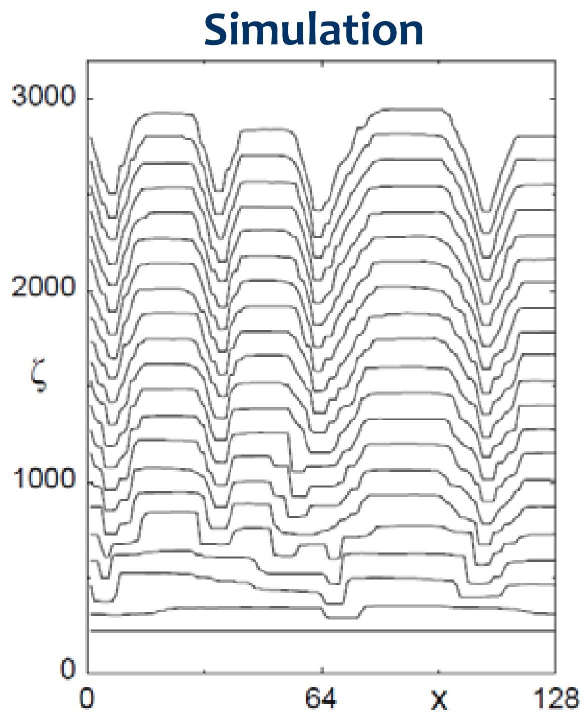
O. Pierre-Louis et al., PRL 80, 4221 (1998).

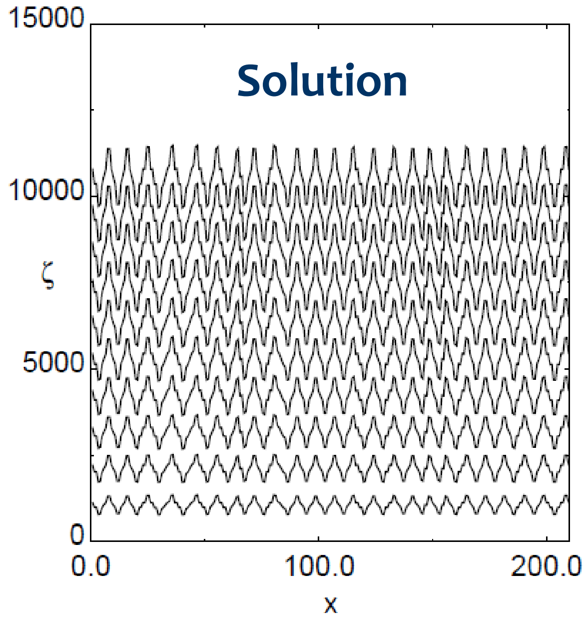
成長中のステップ列の不安定化

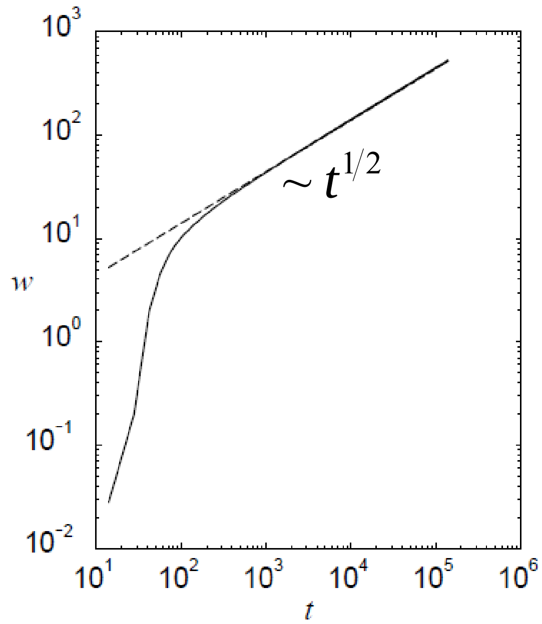
$$\partial_t \zeta = -\partial_x \left[\frac{\alpha \partial_x \zeta}{1+(\partial_x \zeta)^2} + \frac{\beta}{1+(\partial_x \zeta)^2} \times \partial_x \left\{ \frac{\partial_{xx} \zeta}{\left(1+(\partial_x \zeta)^2\right)^{3/2}} \right\} \right]$$

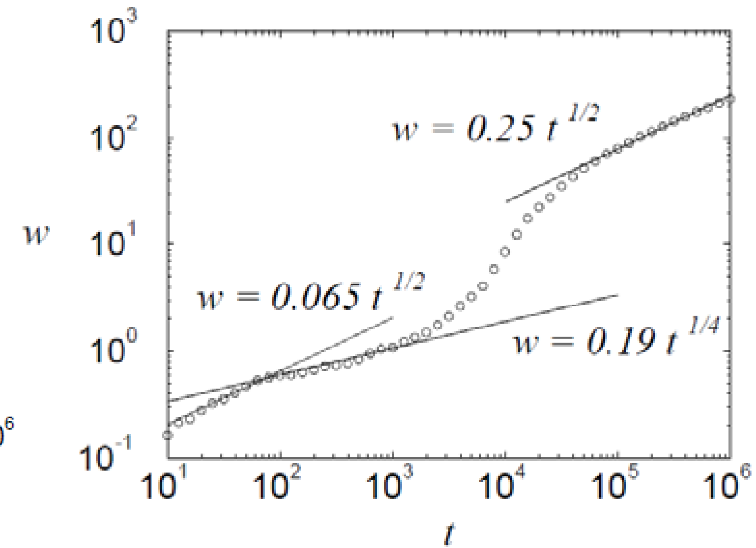
The main reason for this "singular scaling" of ζ with respect to ϵ is that departure from equilibrium coincides with the occurrence of instability. This appeared above in the fact that F scaled as ϵ. This strongly contrasts with the case where a finite critical flux exists.

$$\varepsilon = \frac{f - f_c}{f_c - f_{eq}},$$









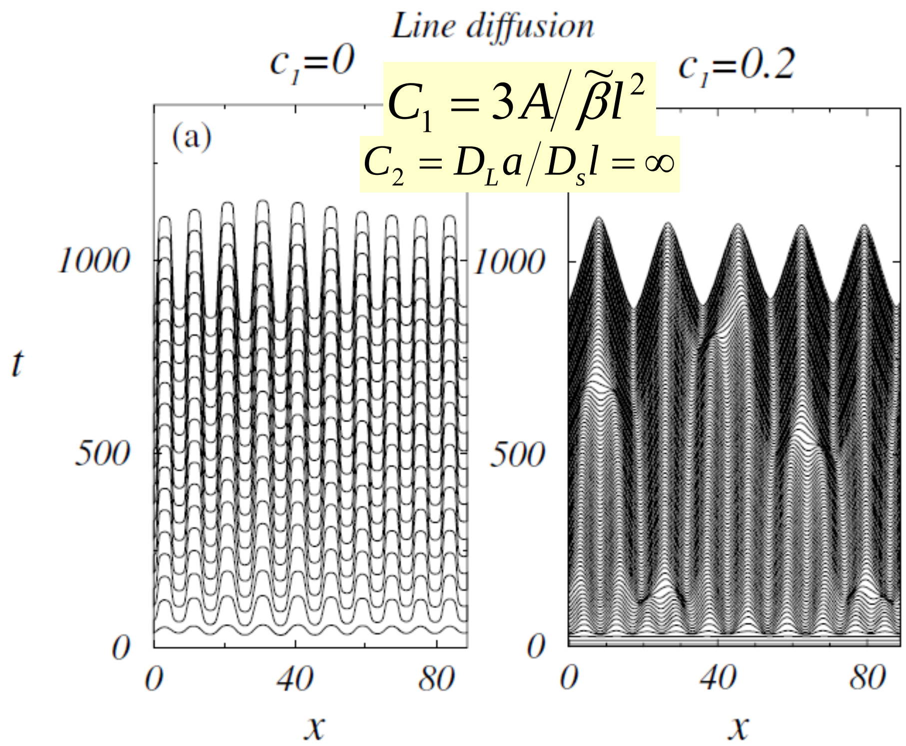
O. Pierre-Louis et al., PRL 80, 4221 (1998).

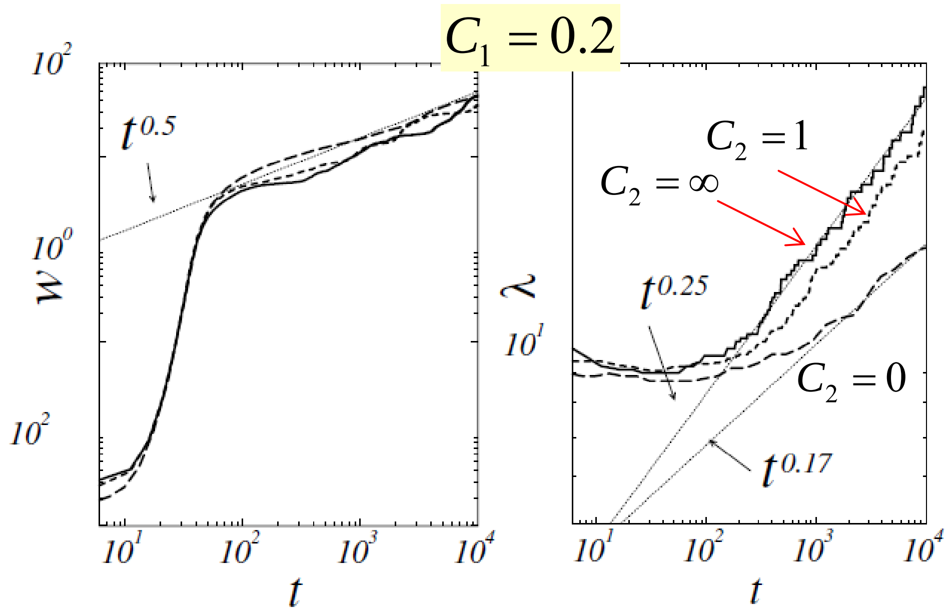
蛇行へのステップ相互作用の影響

Nonlinear equation

$$\partial_t \zeta = -\partial_x \left\{ \frac{\partial_x \zeta}{1+(\partial_x \zeta)^2} + \frac{1}{1+C_2} \left(\frac{1}{1+(\partial_x \zeta)^2} + \frac{C_2}{\left[1+(\partial_x \zeta)^2\right]^{1/2}} \right) \right.$$

$$\left. \times \partial_x \left[\left(\frac{1+C_1\left[1+(\partial_x \zeta)^2\right]\left[1+2(\partial_x \zeta)^2\right]}{1+C_1} \right) \frac{\partial_{xx} \zeta}{\left[1+(\partial_x \zeta)^2\right]^{3/2}} \right] \right\}$$





S. Paulin et al., PRL 86, 5538 (2001).

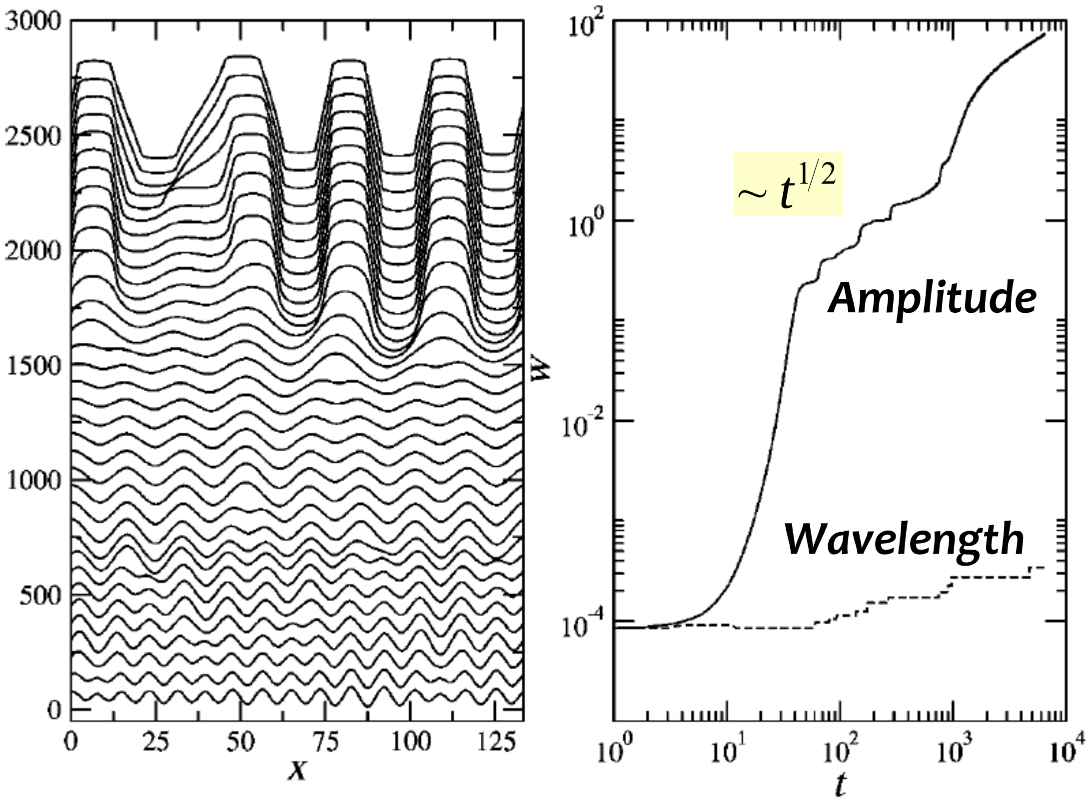
蛇行への異方性の影響

Linear stability analysis

Most unstable mode (in phase) $\lambda_m = 4\pi\left\{\Gamma(\bar{\theta})\left[D_S l + D_L(\bar{\theta})a\right]/\Omega F l^2\right\}^{1/2}$

Nonlinear equation (slope $m = \partial_x \zeta$)

$$\partial_t m = -\partial_{xx}\left\{\frac{\sigma_0 m}{1+m^2} - \widetilde{M}_0(\theta)\partial_x\left[A_\Gamma(\theta)\kappa\right]\right\}$$



Anisotropic line diffusion

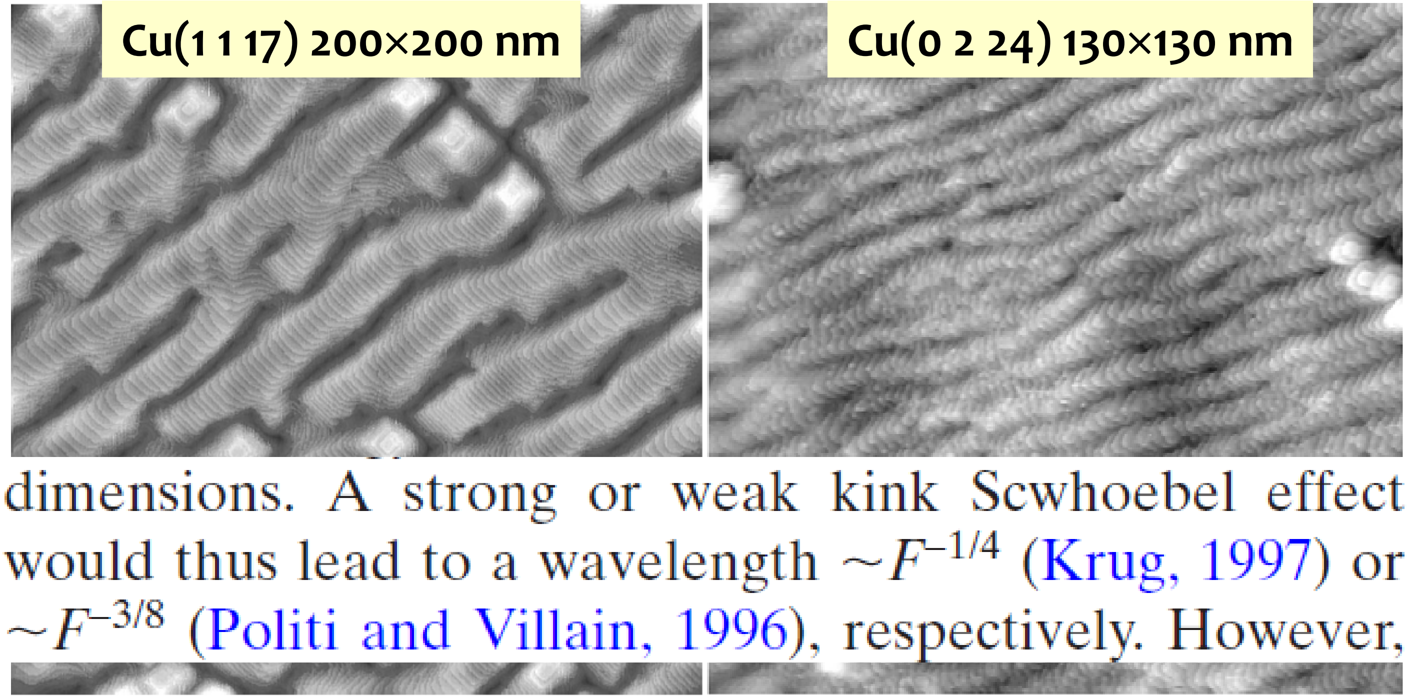
$$D_L(\theta) = D_{L0}\left[1+\varepsilon_L \cos\left[4(\theta-\theta_L)\right]\right]$$

$$\theta_L = \pi/4$$

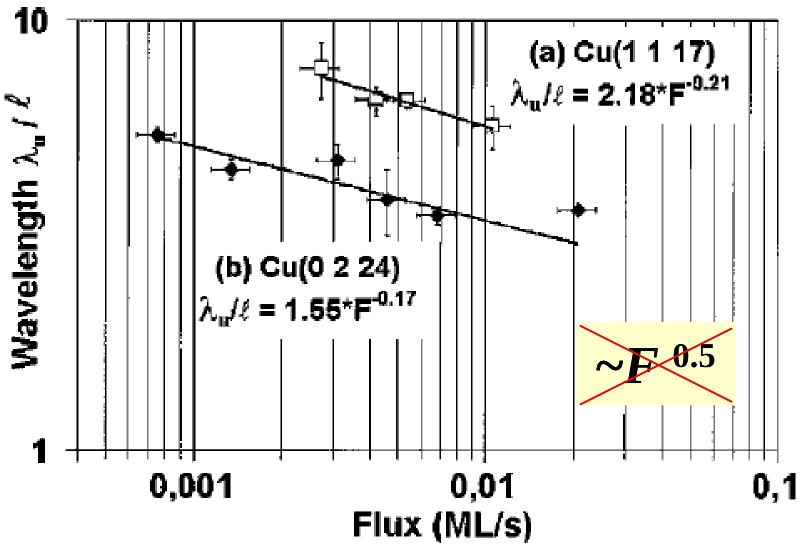
$$\varepsilon_L = 0.92$$

G. Danker et al., PRE 68, 020601 (2003).

成長中のステップ列の不安定化(実験)



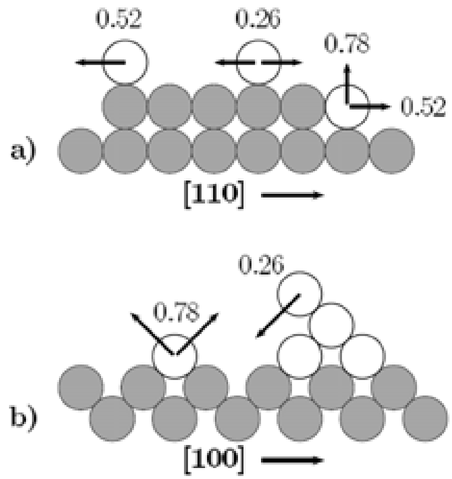
dimensions. A strong or weak kink Scwhoebel effect would thus lead to a wavelength $\sim F^{-1/4}$ (Krug, 1997) or $\sim F^{-3/8}$ (Politi and Villain, 1996), respectively. However,

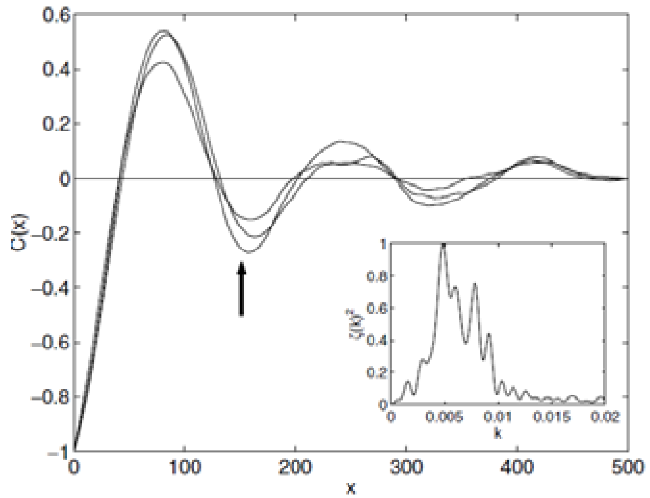


T. Maroutian et al.,
Phys. Rev. B 64, 165401 (2001).

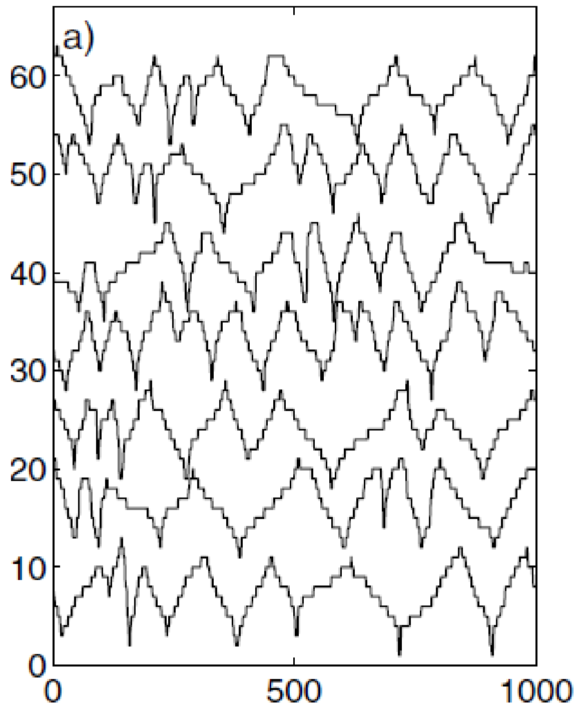
Kink Ehrlich-Schwoebel effect

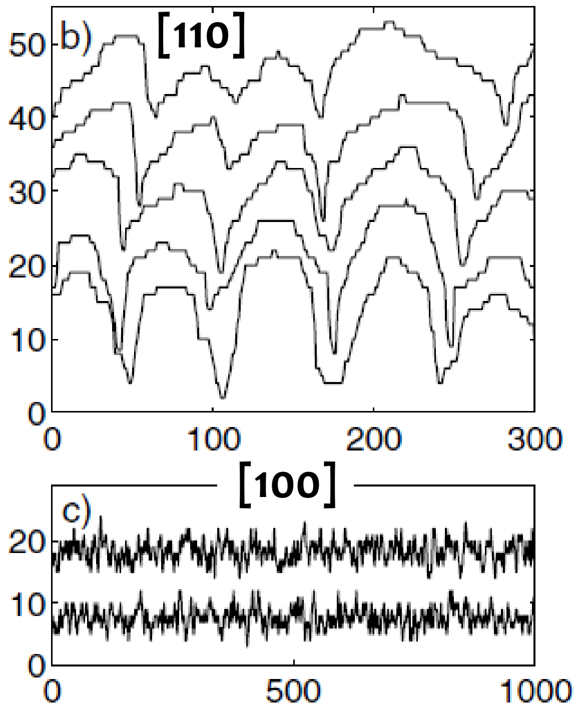
Monte Carlo simulation on metal fcc(001) surface

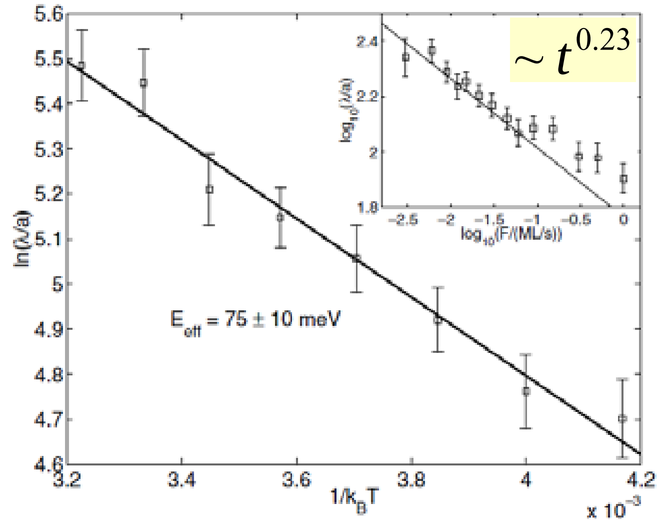




[110]





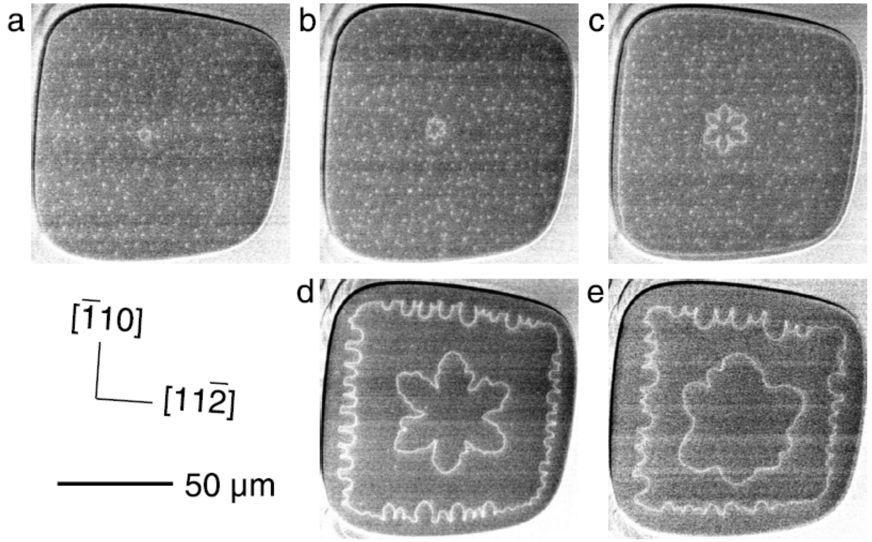


M. Rusanen et al., PRL 86, 5317 (2001).

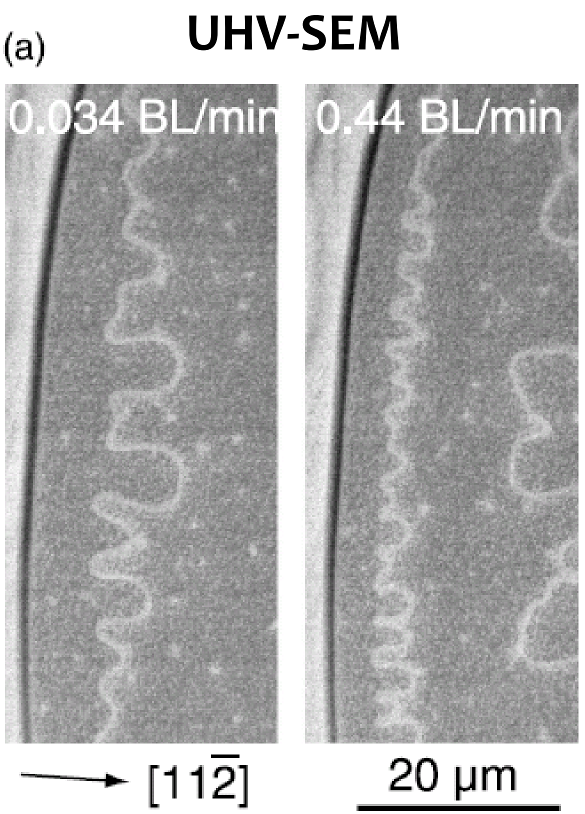
成長中のステップ列の不安定化(実験)

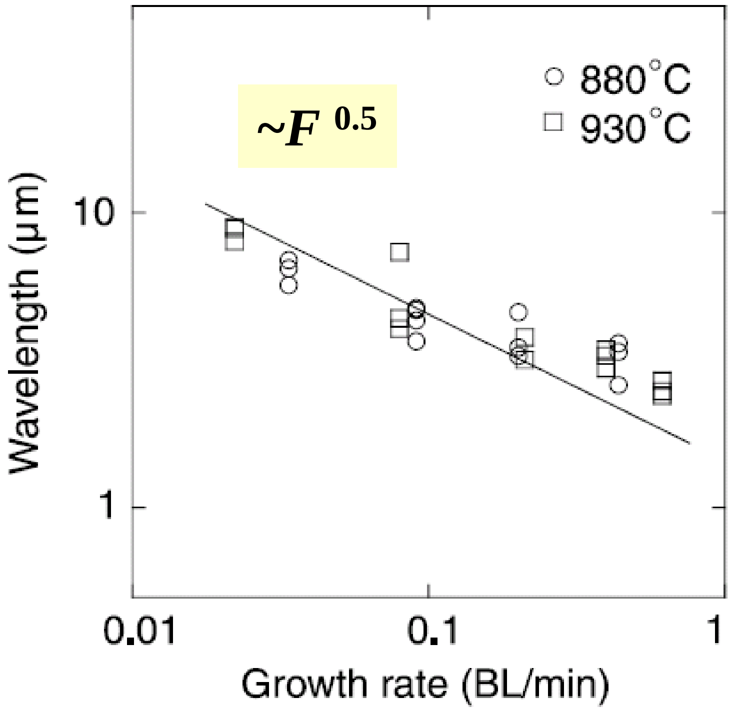
超平坦Si(111)面上でのSi昇華

Y. Homma et al., Surf. Sci. 492, 125 (2001).



UHV-SEM

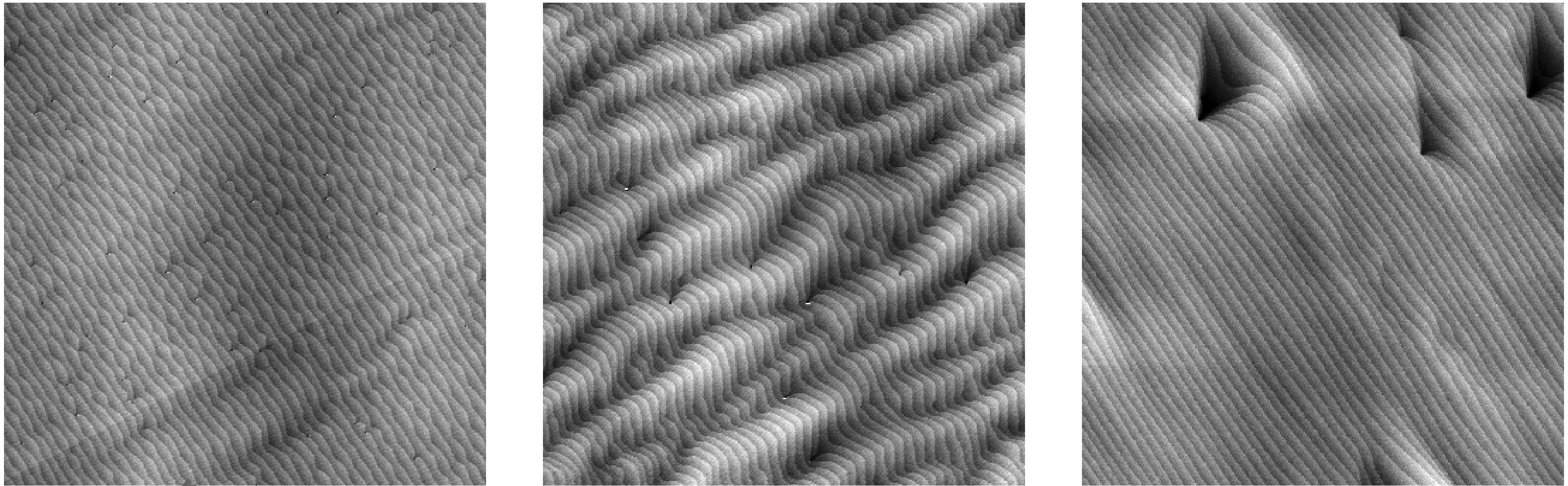




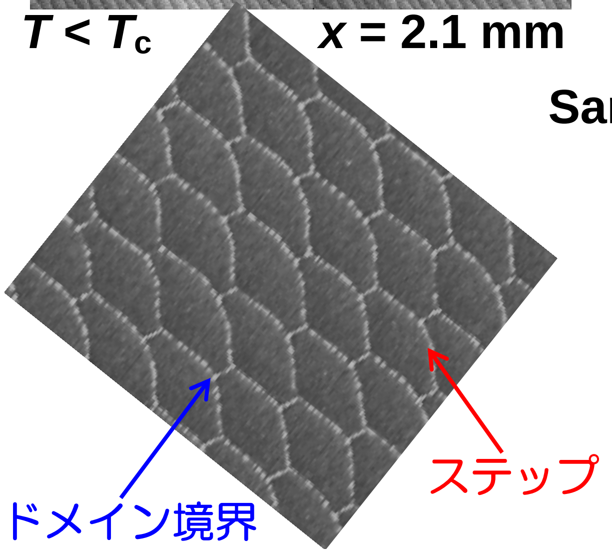
二相共存表面での成長中のステップの蛇行

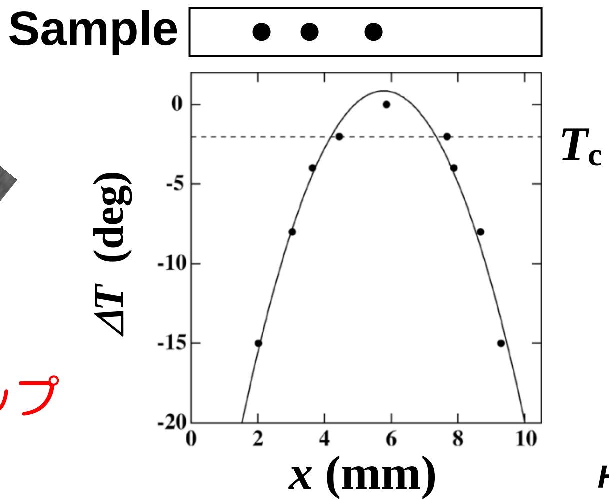
Si(111)表面でのSi成長

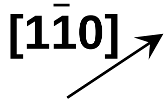
成長後の表面の大気中AFM images



$T < T_c$ x = 2.1 mm $T \leq T_c$ x = 3.9 mm $T > T_c$ x = 5.4 mm





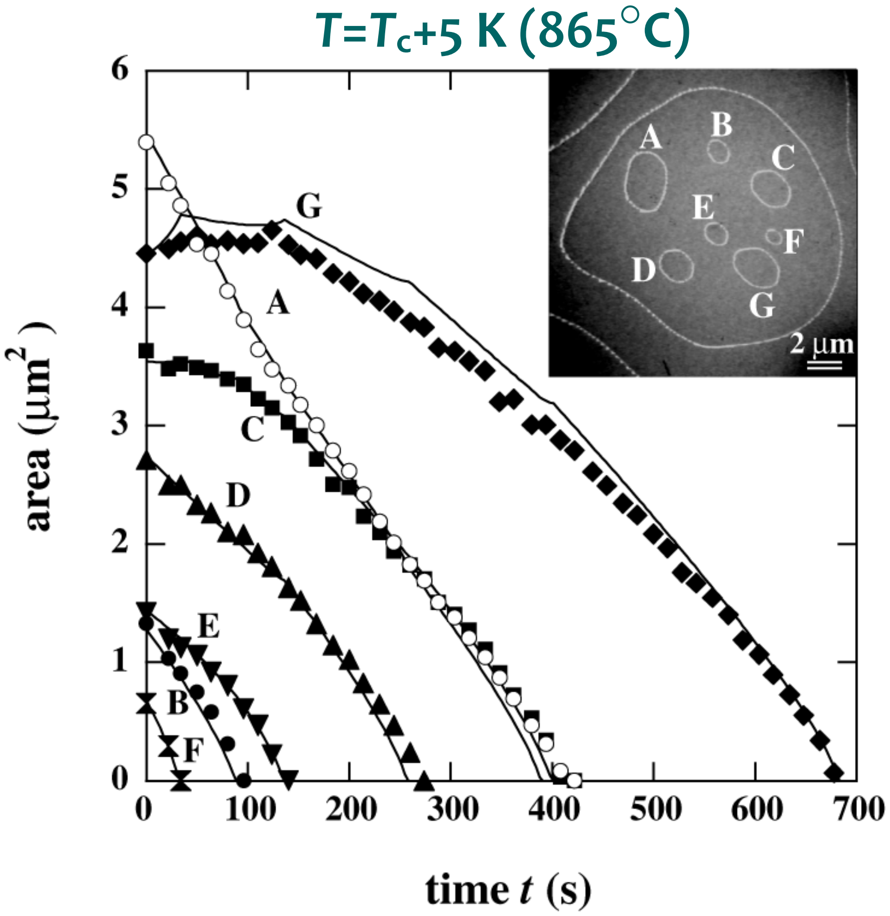


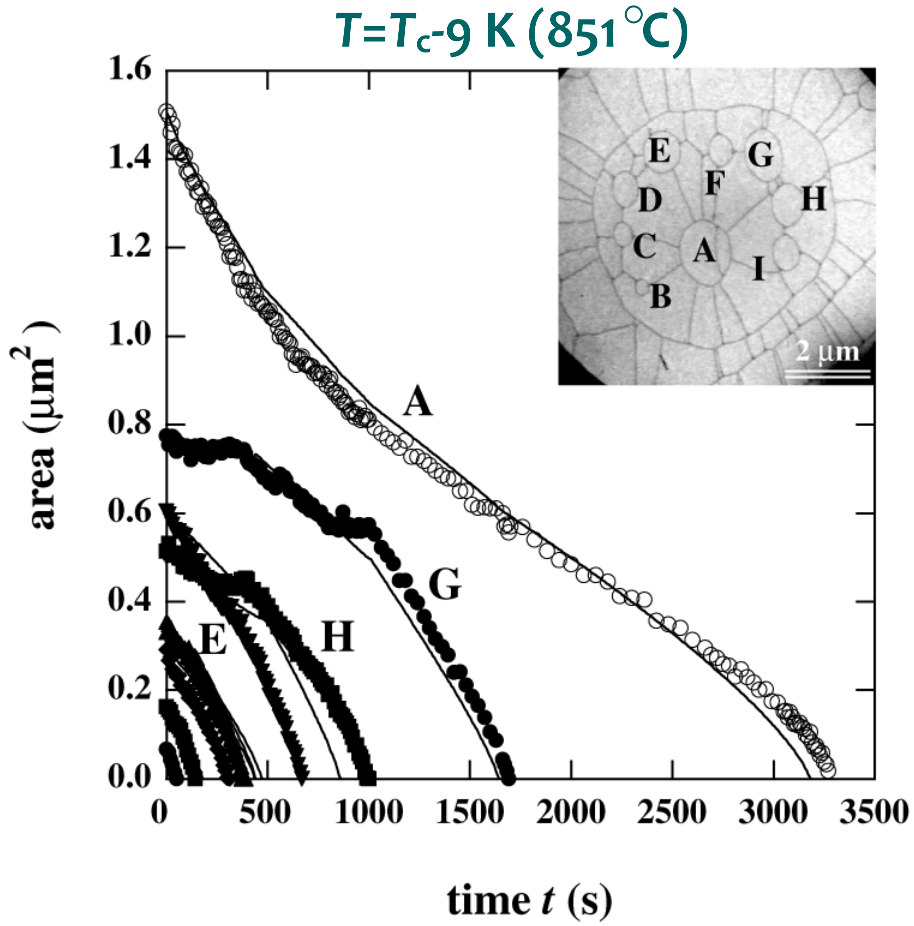
Thickness = 20 nm

Scan area = 20×20 μm

H. Hibino et al. , Surf. Sci. 527, L222 (2003).

Si(111)表面での質量輸送

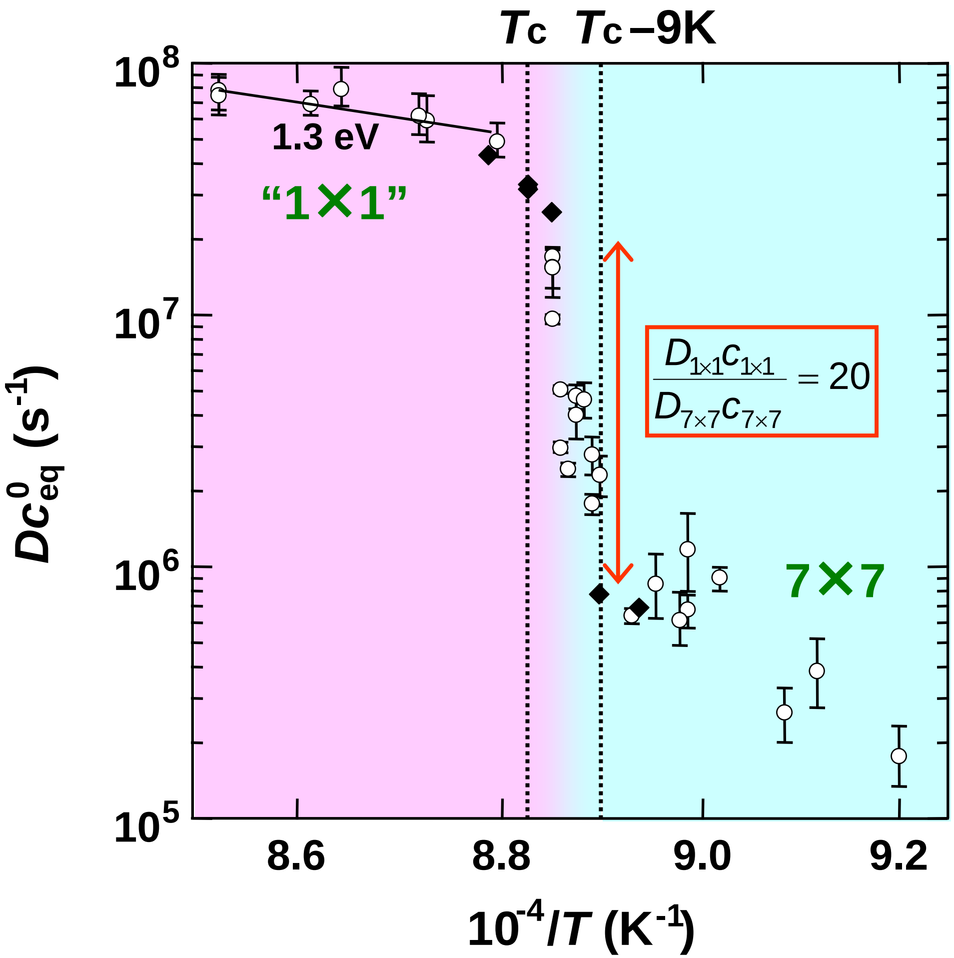




(1) 面積変化はサイズが小さいほど速い。
(2) 島と穴は、ほぼ同様に変化する。
(3) “1×1”表面での面積変化は、7×7表面での面積変化より速い。

H. Hibino, C.-W. Hu, T. Ogino, and I. S. T. Tsong, Phys. Rev. B 63, 245402 (2001).

表面質量輸送係数の温度依存性



表面質量輸送係数は、相転移で階段状に変化し、7×7表面において、"1×1"表面に比べ小さい。

H. Hibino et al., Phys. Rev. B 63, 245402 (2001).

二相共存表面での拡散方程式

境界条件（拡散律速）：

◆ ステップ

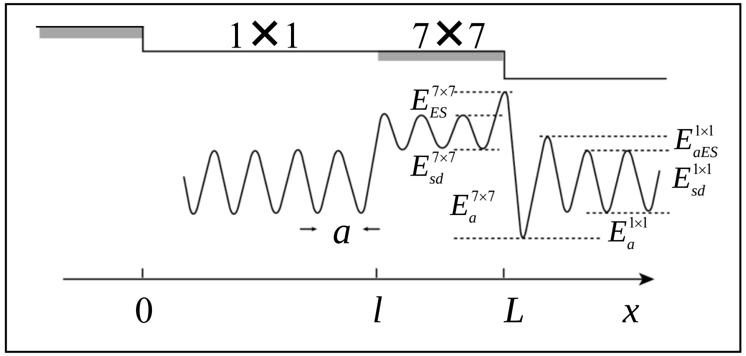
◆ $c(0) = c_{1\times1}$

◆ $c(L) = c_{7\times7}$

◆ 相境界

◆ $D_{1\times1} \left.\frac{dc}{dx}\right|_{l_-} = D_{7\times7} \left.\frac{dc}{dx}\right|_{l_+}$

◆ $\frac{c|_{l_-}}{c_{1\times1}} = \frac{c|_{l_+}}{c_{7\times7}}$

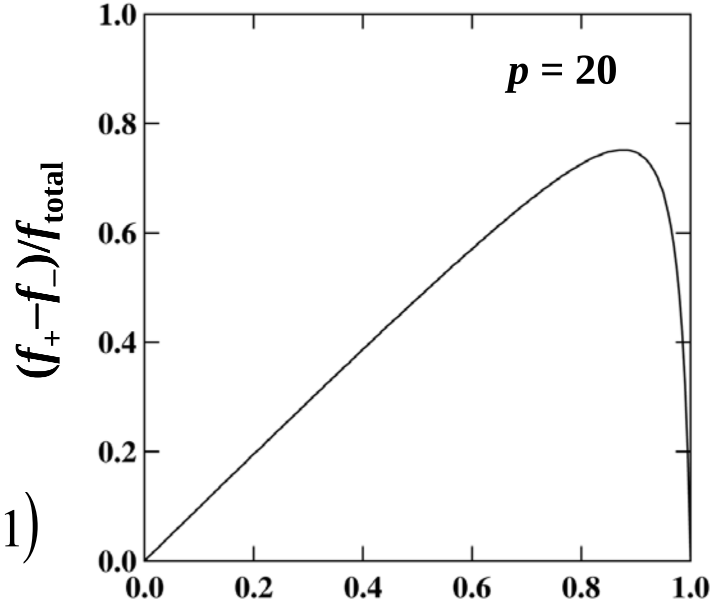


下側テラスからの原子流：

$$f_+ = D_{1\times1} \left.\frac{\partial c}{\partial x}\right|_{0+} = \frac{FL}{2} \cdot \frac{pa^2 - a^2 - p}{pa - a - p}$$

$$\text{, where } a = \frac{l}{L},\quad p = \frac{D_{1\times1} c_{1\times1}}{D_{7\times7} c_{7\times7}}.$$

f_+ / f_{total} は、$a = \sqrt{p} / \left(\sqrt{p} + 1\right)$ で最大値 $\sqrt{p} / \left(\sqrt{p} + 1\right)$

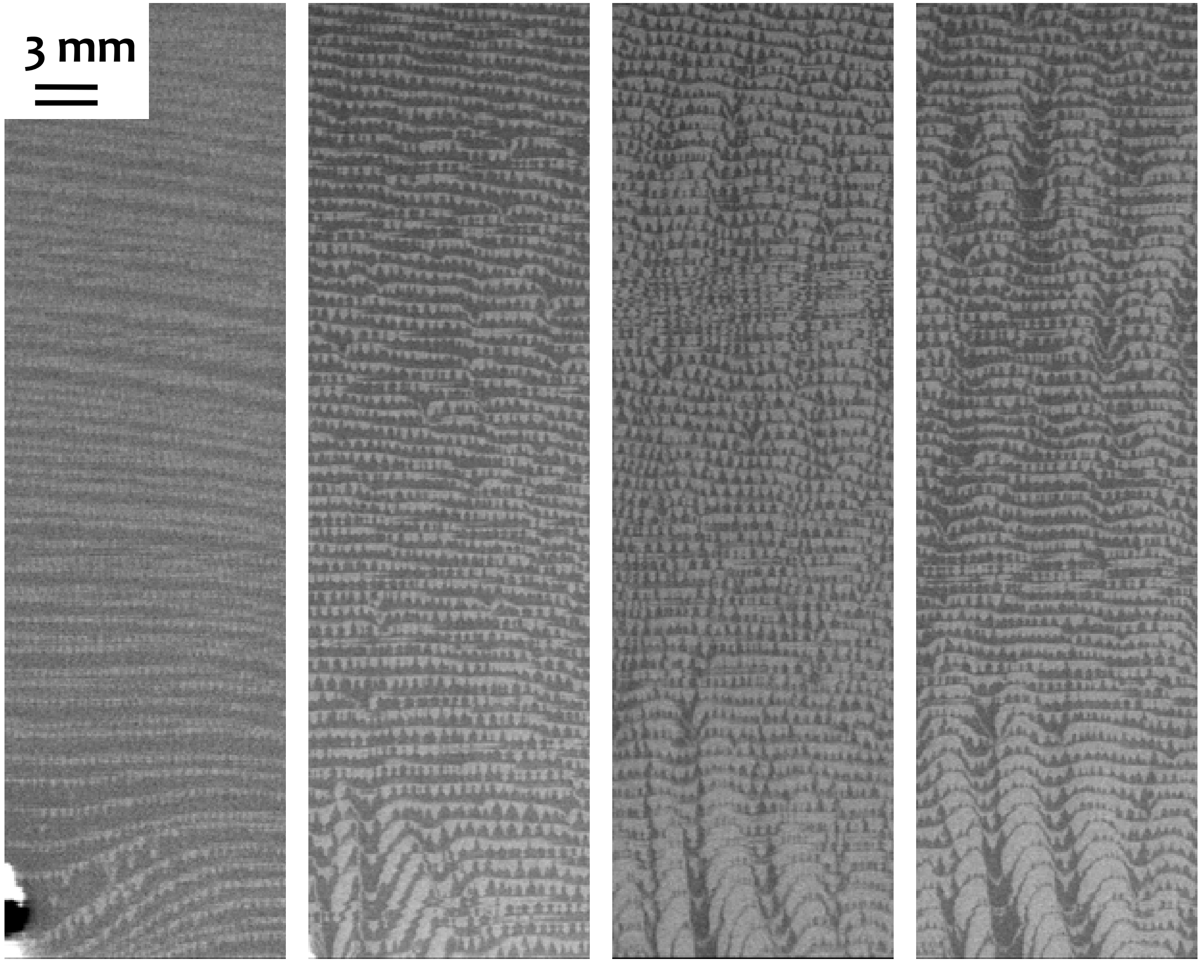


H. Hibino et al. , Surf. Sci. 527, L222 (2003).

成長中のステップの蛇行(ステップ方位の影響)

Si(111)表面でのSi成長

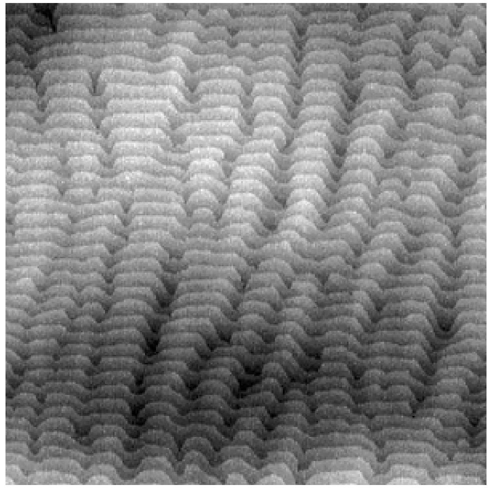
SEMその場観察



before growth　6 nm　10 nm　15 nm

$[11\bar{2}]$

20 nm成長後の表面
(大気中 AFM 像)



6 μm

H. Hibino et al., Surf. Sci. 527, L222 (2003).

成長中のステップの蛇行(その場観察)

Si(111)表面でのSi成長

成長中のLEEM観察

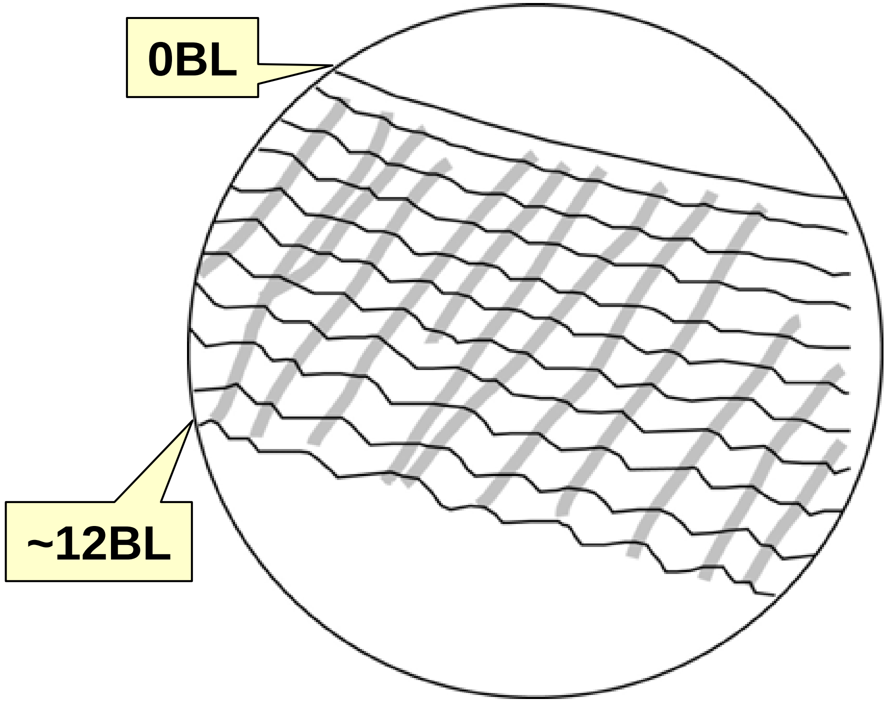
Substrate preparation

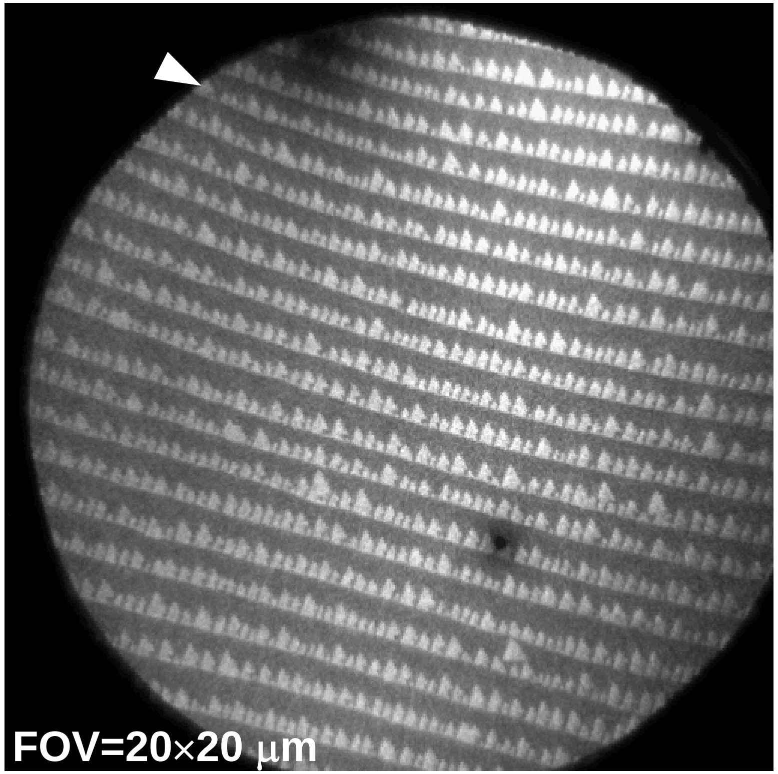
♦**After Co deposition at 780°C, annealed at 800°C.**

Growth conditions

♦**Substrate temperature = 780°C**

♦**Growth rate = ~0.5 BL/min**





100 times faster than real

成長中のステップの蛇行(加熱電流の影響)

H. Hibino et al., Surf. Sci. 527, L222 (2003).

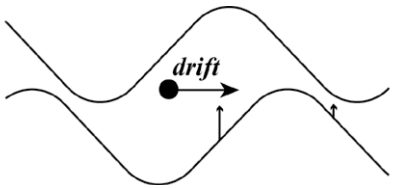
Si(111)表面でのSi成長

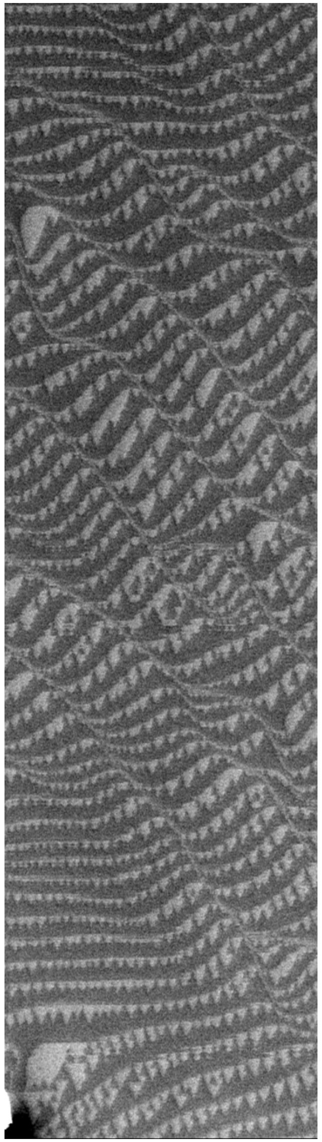
加熱電流の影響

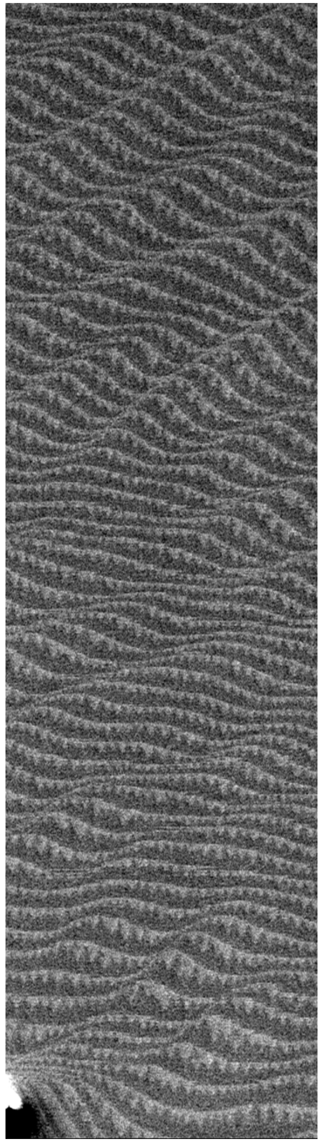
SEMその場観察

i →

***t* = 3.7 nm**

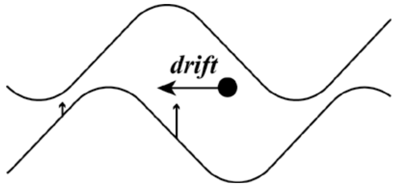






i ←

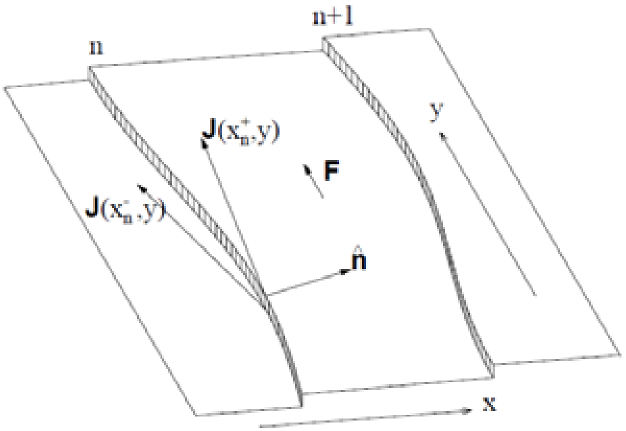
***t* = 3.1 nm**



3 μm

Bending instability?

D.-D. Liu et al., PRL 81, 2743 (1998).



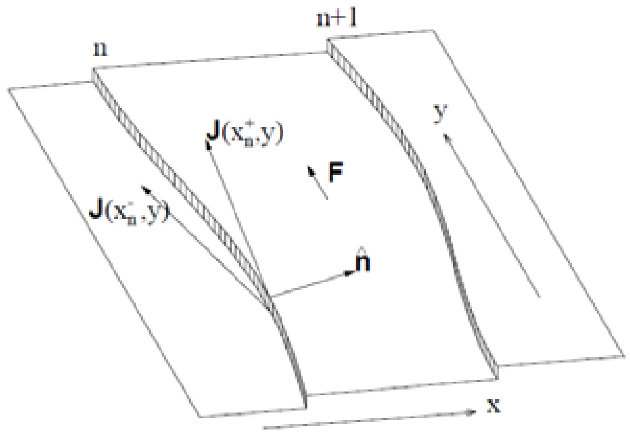
$$D_s\left(\frac{\partial^2 c}{\partial x^2}+\frac{\partial^2 c}{\partial y^2}\right)-\frac{D_s F}{kT}\frac{\partial c}{\partial y}=0$$

ベンディング

拡散方程式

$$D_s\left(\frac{\partial^2 c}{\partial x^2}+\frac{\partial^2 c}{\partial y^2}\right)-\frac{D_s F}{kT}\frac{\partial c}{\partial y}=0. \qquad (2)$$

$$\delta x_n(y,t)=\sum_{q,\phi}\delta x(q,\phi)e^{iqy+in\phi+\omega(q,\phi)t}+\text{c.c.}$$



境界条件

$$\pm\kappa[c_n^{\mathrm{eq}}(y)-c(x_n^{\pm}(y),y)]=\hat{\mathbf{n}}\cdot\mathbf{J}(x_n^{\pm}(y),y), \qquad (3)$$

$$\mathbf{J}(x,y)=-D_s(-\partial/\partial x,-\partial/\partial y+f)c(x,y), \qquad (4)$$

質量保存

$$\begin{aligned}v_n&=a^2\hat{\mathbf{n}}\cdot[\mathbf{J}(x_n^+,y)-\mathbf{J}(x_n^-,y)]\\&=D_s a^2[2c_n^{\mathrm{eq}}(y)-c(x_n^+,y)-c(x_n^-,y)], \qquad (5)\end{aligned}$$

平衡濃度

$$c_n^{\mathrm{eq}}(y)=c_0^{\mathrm{eq}}\exp\left(\frac{\mu_n(y)-\mu_C}{kT}\right), \quad \mu_n=a^2[V'(w_n)-V'(w_{n-1})+\tilde{\beta}\partial^2 x_n/\partial y^2]+\mu_C. \qquad (7)$$

D.-D. Liu et al., PRL 81, 2743 (1998).

ベンディング

$$\omega(q,\phi) = \frac{-2D_s c_{eq}^0 a^2 \Lambda_q [f q d \sin\phi + g(q,\phi)]}{2\Lambda_q d \cosh(\Lambda_q w_0) + (\Lambda_q^2 d^2 + 1)\sinh(\Lambda_q w_0)}, \quad (8)$$

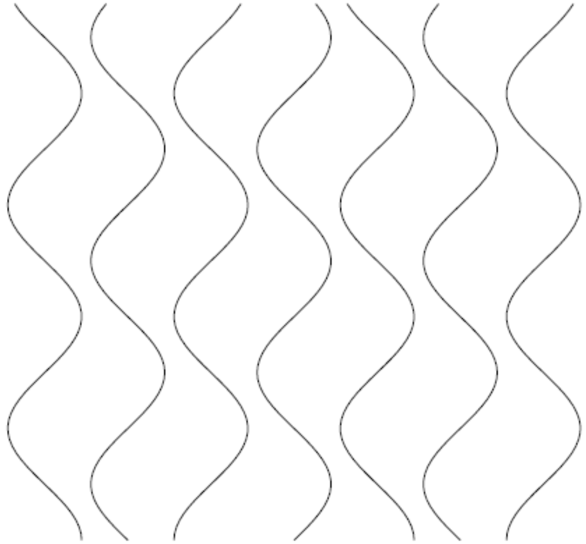
where

$$g(q,\phi) = g_x(1 - 2\cos\phi + \cos 2\phi) - g_y q^2 \cos\phi + [g_y q^2 + 2g_x(1-\cos\phi)] \times [\cosh(\Lambda_q w_0) + \Lambda_q d \sinh(\Lambda_q w_0)], \quad (9)$$

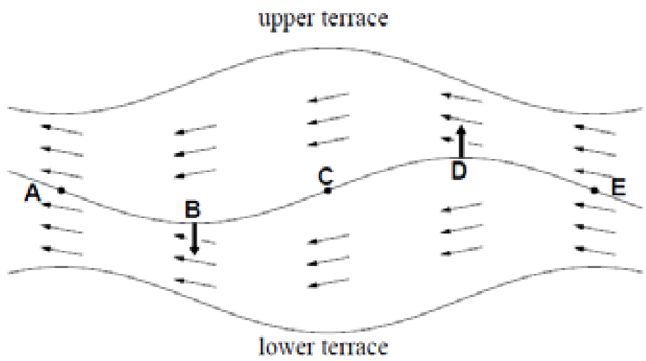
$$\Lambda_q = \sqrt{q^2 + ifq}, \quad (10)$$

and

$$g_x = \frac{a^2}{kT}\frac{d^2V}{dw^2}\bigg|_{w=w_0}, \qquad g_y = \frac{\tilde{\beta}a^2}{kT}. \quad (11)$$



$-\phi = \pi/2$



$f > 0$ (+ y direction)

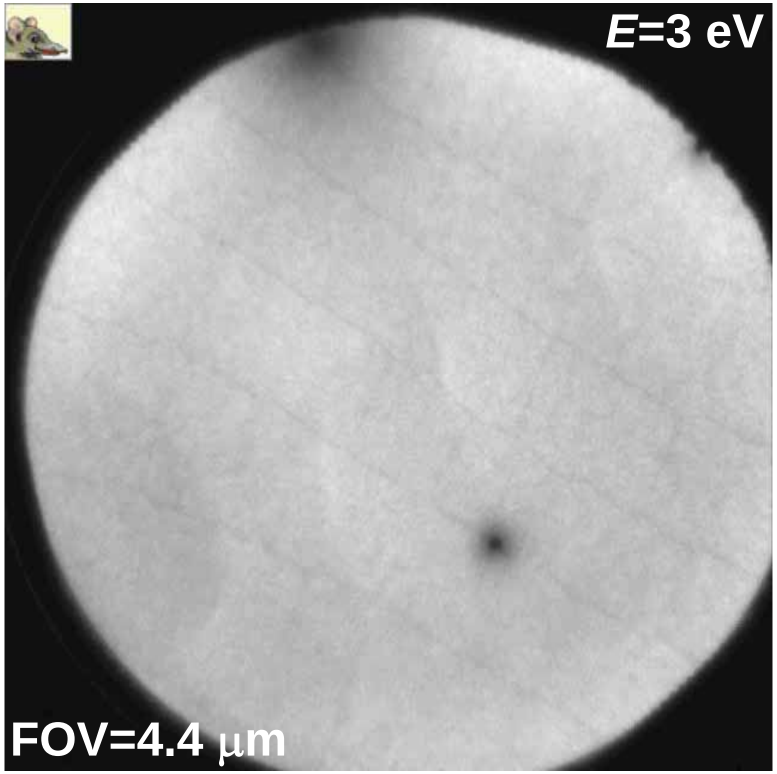
$\longrightarrow$ $\mathrm{Re}\,\omega > 0$ for $-\pi < \phi < 0$

Maximally unstable when $\phi = -\pi/2$

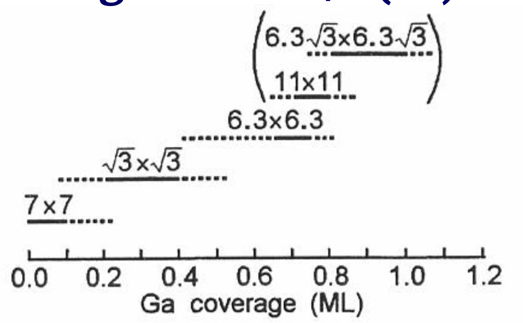
D.-D. Liu et al., PRL 81, 2743 (1998).

Ga蒸着中のSi(111)ステップ不安定化

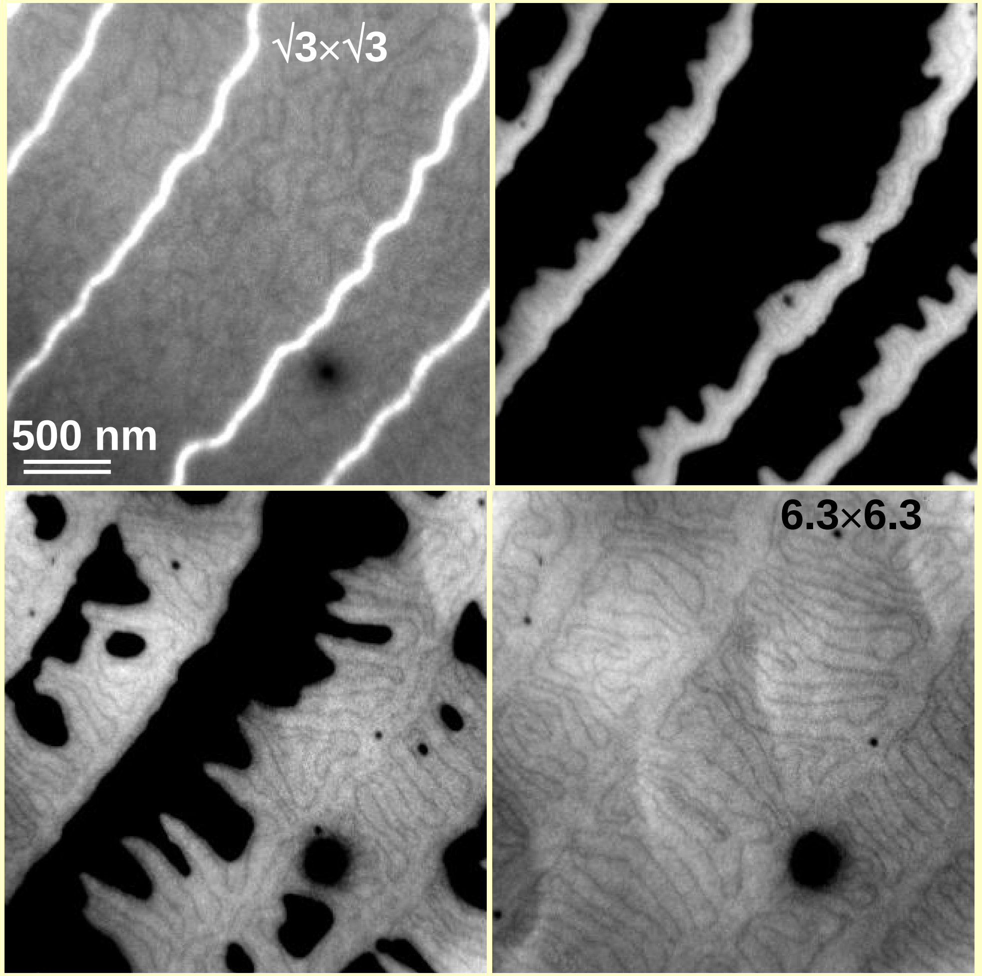
Ga deposition at 580°C



Phase diagram of Ga/Si(111)

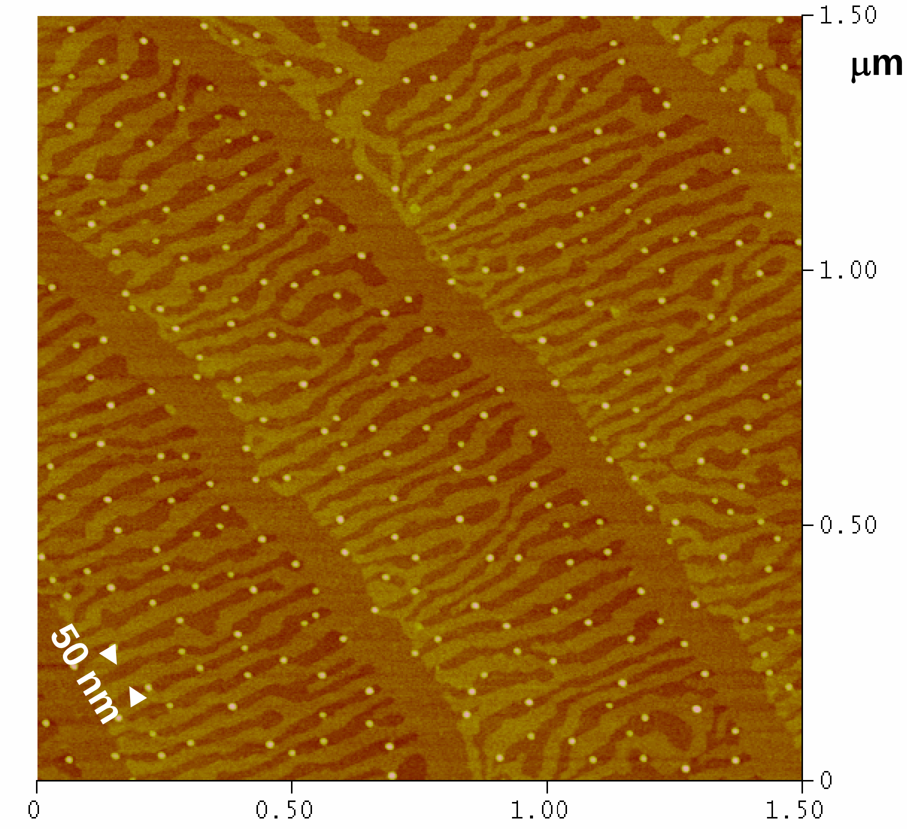


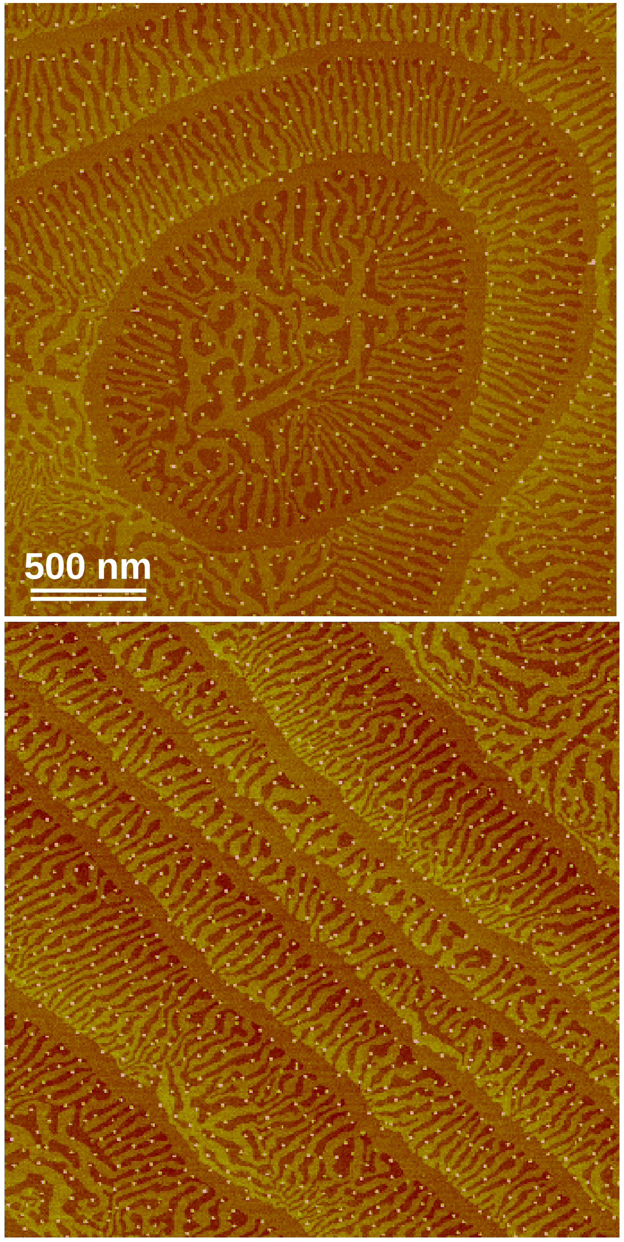
Ga deposition at 623°C



Ga蒸着後の櫛型ステップ

***Ex-situ* AFM images**
after Ga deposition at 581°C





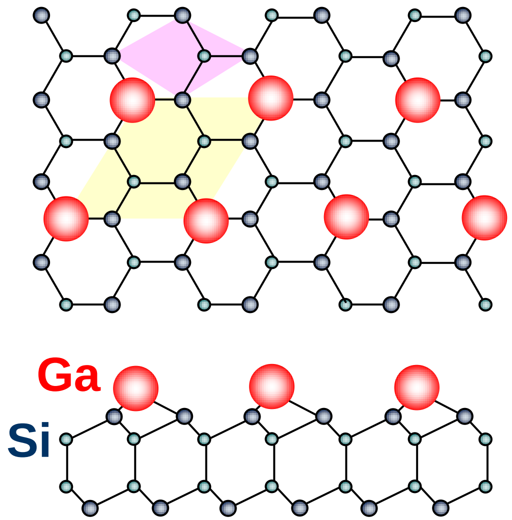
H. Hibino et al., Surf. Sci. 602, 2421 (2008).

Ga蒸着によるステップ形状不安定化

◆**Si(111)√3×√3-Ga**

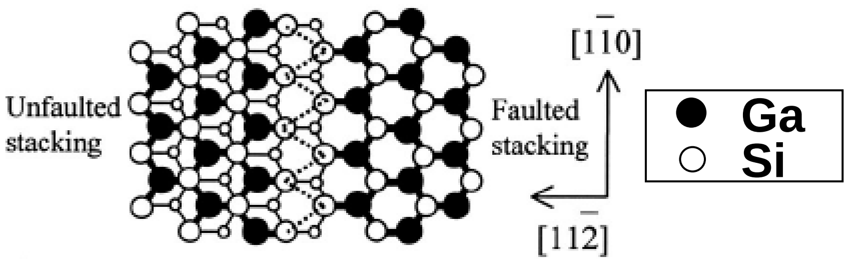
Ga T4 adatom model

➡ **Ga coverage: 0.33 ML**
Si atom density: 2.0 atoms/(1×1) unit



◆**Si(111)6.3×6.3-Ga**

Ga-Si double layer structure model

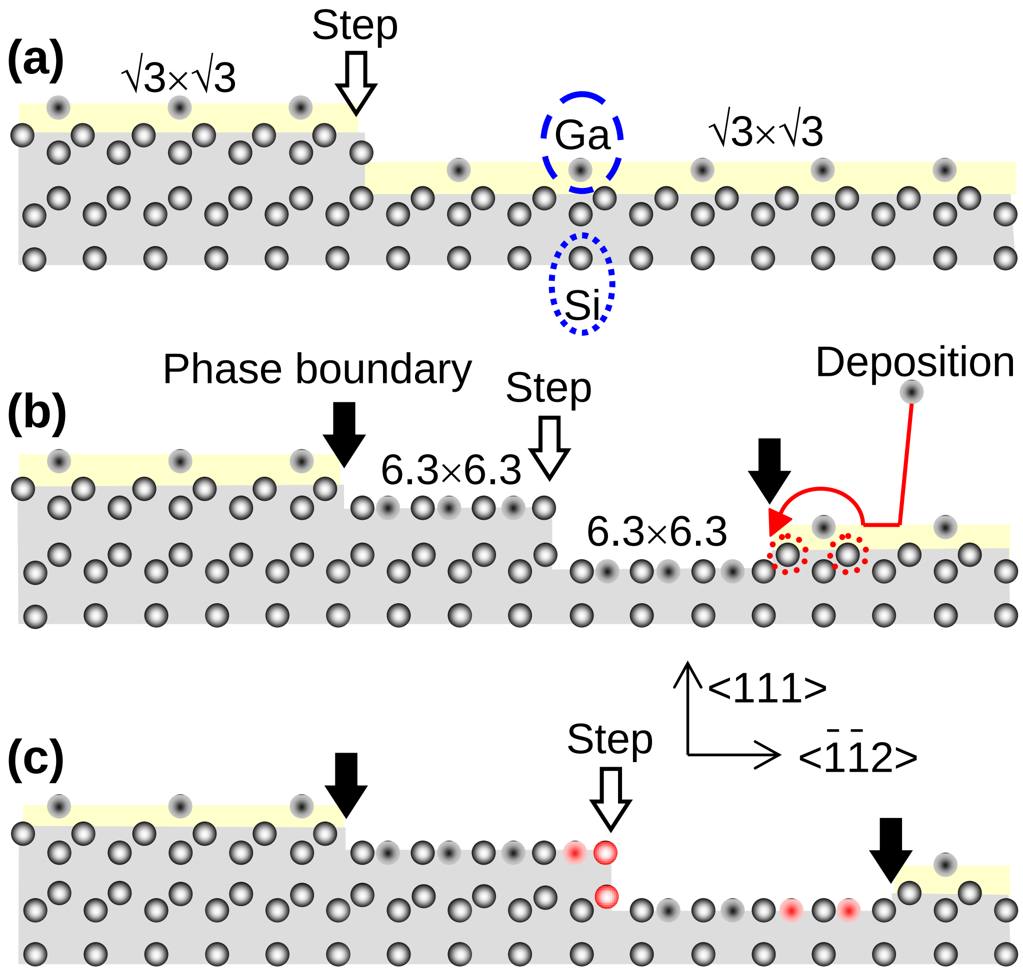


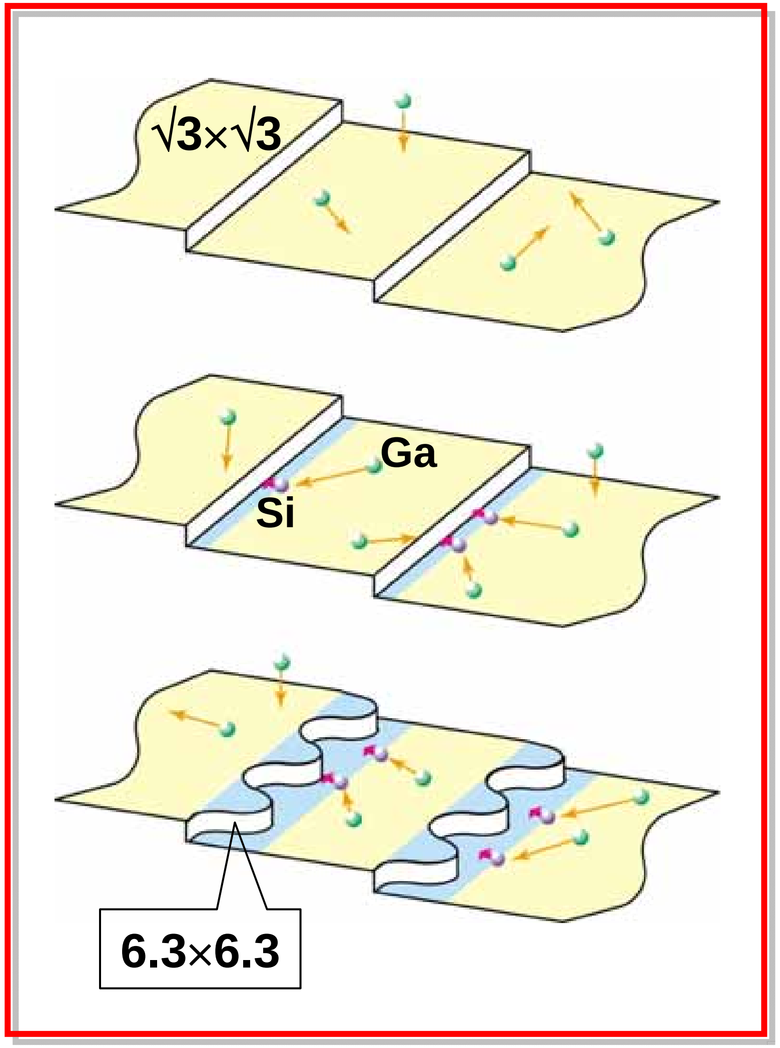
➡ **Ga coverage: ~0.7 ML**
Si atom density: ~1.3 atoms/(1×1) unit

LEEM image

➡ **Si atom density:**
~0.94 atoms/(1×1) unit

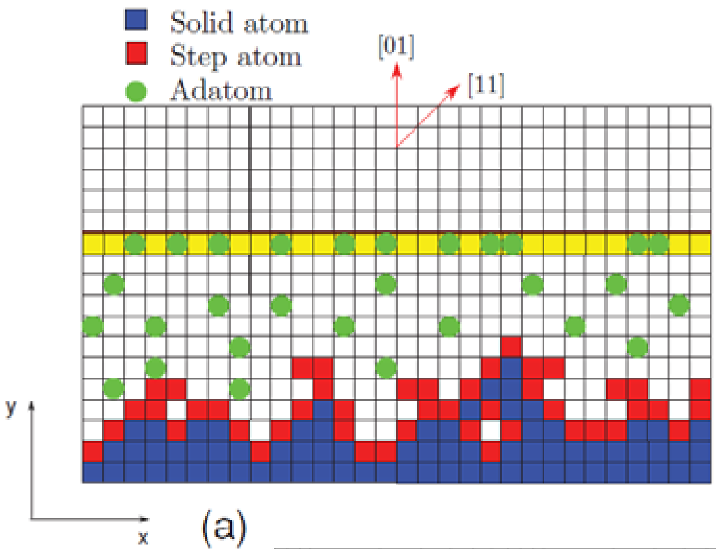
Ga蒸着による構造変化モデル

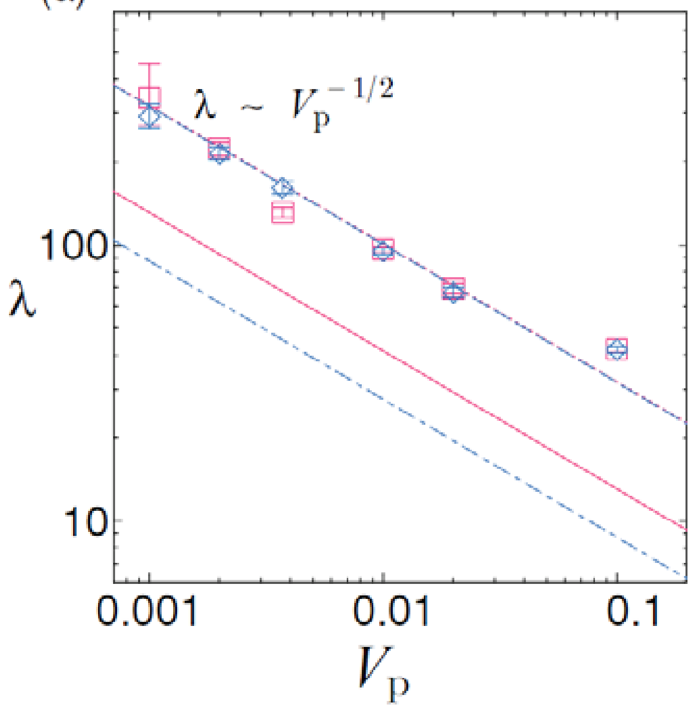


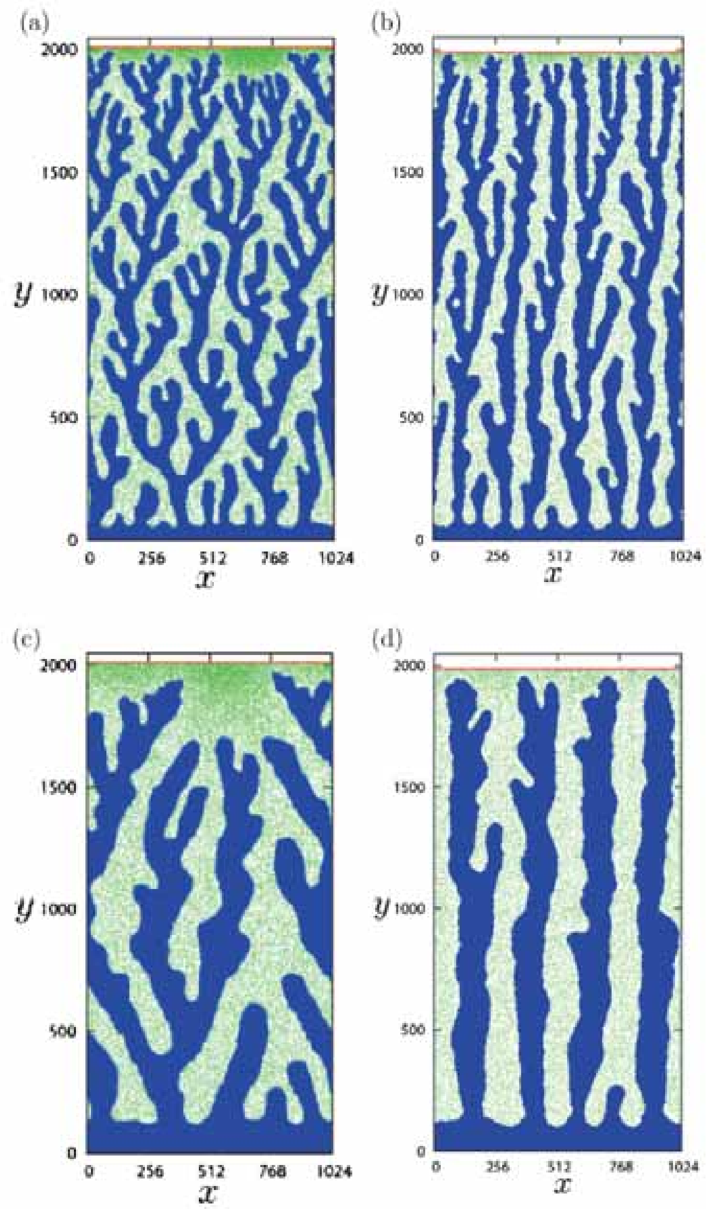


H. Hibino et al., Surf. Sci. 602, 2421 (2008).

Pattern formation induced by moving linear source

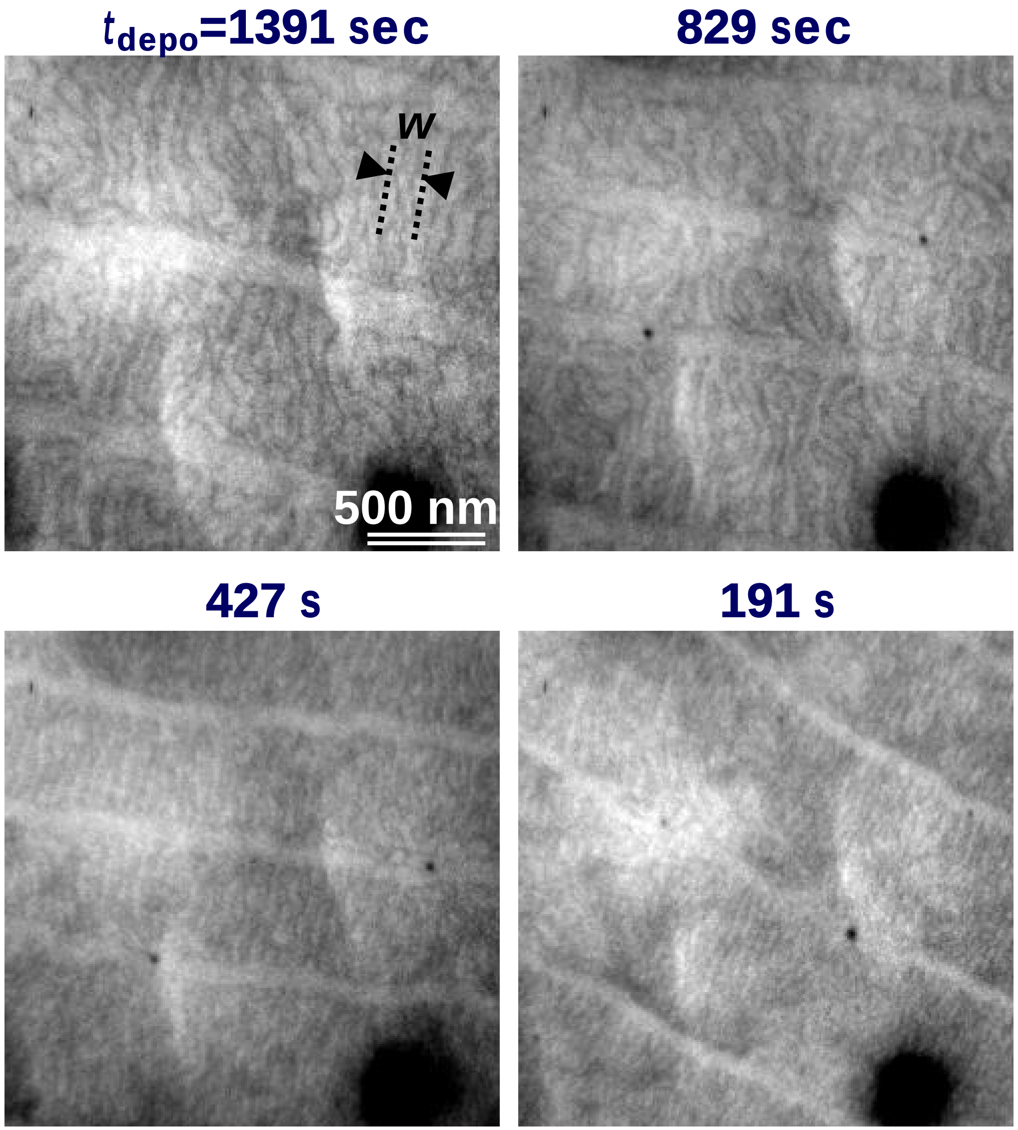






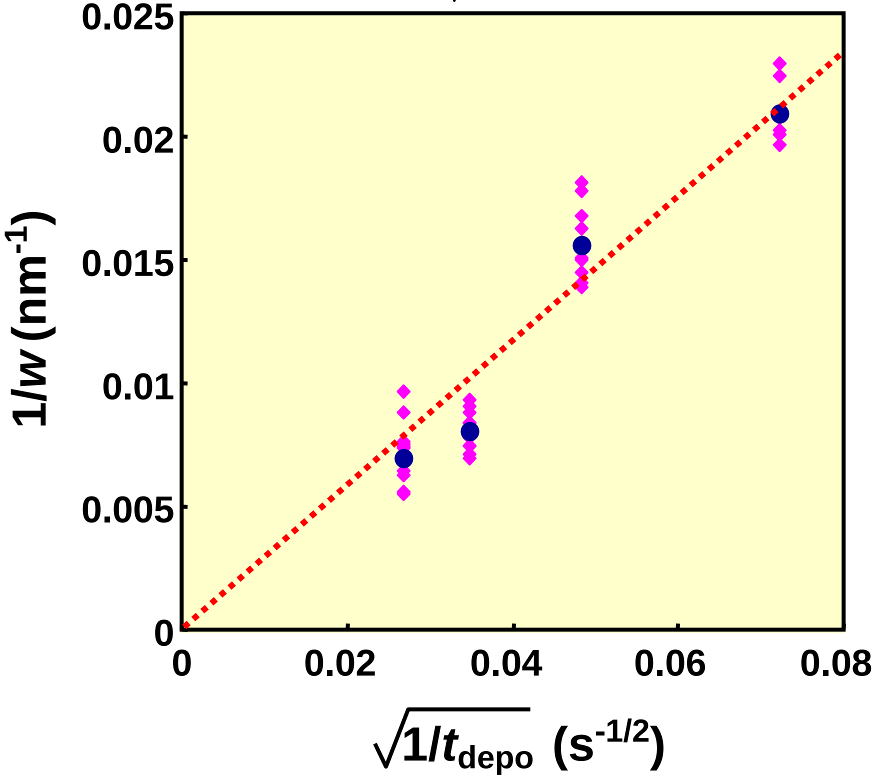
S. Kondo et al., PRB 84, 045420 (2011).

Ga蒸着によるステップ形状不安定化



T_{sub}=581°C　　E=2-2.7 eV

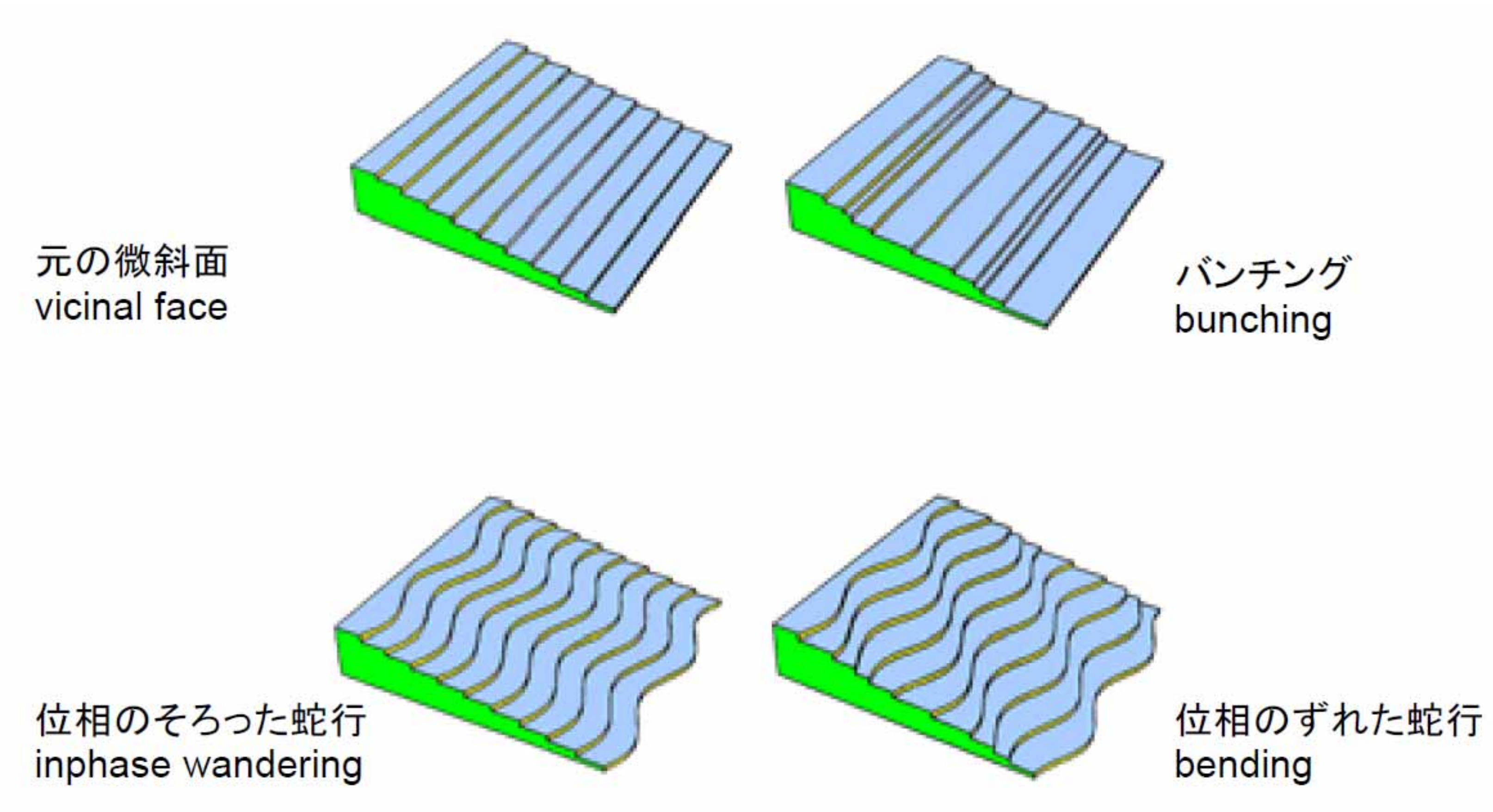
$$k_{max} = \sqrt{\frac{k_B T \nu_0}{3\Omega^2 D c_{eq} \tilde{\beta}}}$$



H. Hibino et al., Surf. Sci. 602, 2421 (2008).

ステップバンチング

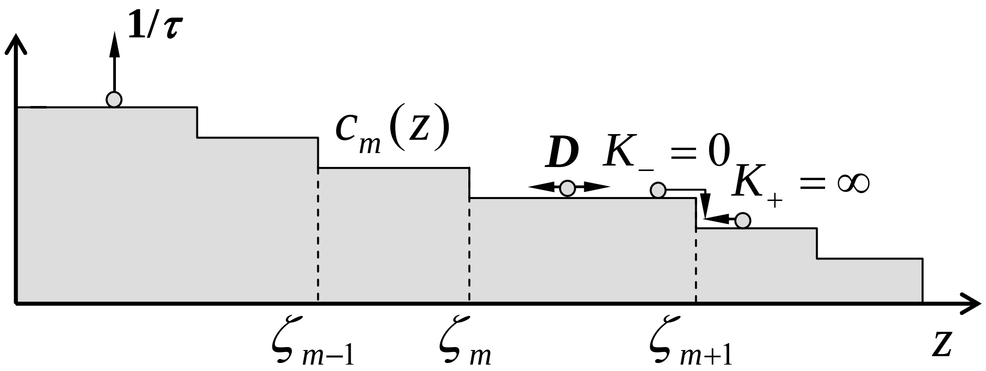
ステップの不安定化



http://www.slab.phys.nagoya-u.ac.jp/uwaha/gairon08.pdf

ステップ前進速度(片側モデル)

One-sided model



$$D\partial_{zz}c - c/\tau = 0$$

$$\partial_z c_- = 0 \quad (K_- = 0)$$

$$c_+ = c_{eq}^m \quad (K_+ = \infty)$$

where $c_{eq}^m = c_{eq}^0\left[1 + \frac{\Omega A}{k_B T}\left(\frac{1}{l_m^3} - \frac{1}{l_{m-1}^3}\right)\right]$

$$c(z) = C_1 e^{z/x_s} + C_2 e^{-z/x_s}$$

$$\begin{pmatrix} e^{l_m/x_s} & -e^{-l_m/x_s} \\ 1 & 1 \end{pmatrix}\begin{pmatrix} C_1 \\ C_2 \end{pmatrix} = \begin{pmatrix} 0 \\ c_{eq}^m \end{pmatrix} \longrightarrow \begin{pmatrix} C_1 \\ C_2 \end{pmatrix} = \frac{2c_{eq}^m}{\cosh(l_m/x_s)}\begin{pmatrix} e^{-l_m/x_s} \\ e^{l_m/x_s} \end{pmatrix}$$

$$V_m = \Omega D \partial_z c_+\big|_m = (\Omega D/x_s)(C_1 - C_2) = -\left(\Omega c_{eq}^m x_s/\tau\right)\tanh(l_m/x_s)$$

V はl_m の単調増加関数=ステップ幅の揺らぎを増幅(バンチング)

等間隔ステップの安定性(片側モデル)

Linear stability analysis

$$z_m = ml + \bar{V}t + \zeta_m$$

0th order in ζ

$$\bar{V} = -\left(\Omega c_{eq}^0 x_s / \tau\right)\tanh\left(l_m / x_s\right)$$

1st order in ζ

$$\dot{\zeta}_m = \underbrace{a_{ne}\left(\zeta_m - \zeta_{m+1}\right)}_{\text{diffusion}} - \underbrace{a_{eq}\left(2\zeta_m - \zeta_{m+1} - \zeta_{m-1}\right)}_{\text{elastic interaction}}$$

diffusion — stabilizing

elastic interaction — destabilizing

where $a_{ne} = -\partial\bar{V}/\partial l = \left(\Omega c_{eq}^0 / \tau\right)\cosh^{-2}\left(l/x_s\right) > 0$

$$a_{eq} = 3\left(\Omega c_{eq}^0 / \tau\right)\tanh\left(l/x_s\right)Ax_s / l^4 > 0$$

等間隔ステップの安定性(片側モデル)

Linear stability analysis

$$\zeta_{\omega\phi} = \int dt \sum_m e^{-i\omega t - im\phi} \zeta_m(t)$$

$$\zeta_m(t) = \int \frac{d\omega}{2\pi} \int \frac{d\phi}{2\pi} e^{i\omega t + im\phi} \zeta_{\omega\phi}$$

$$\dot{\zeta}_m = a_{ne}(\zeta_m - \zeta_{m+1}) - a_{eq}(2\zeta_m - \zeta_{m+1} - \zeta_{m-1})$$

$$\longrightarrow \quad i\omega = [a_{ne} - 2a_{eq}](1 - \cos\phi) - i a_{ne} \sin\phi$$

When $\underbrace{a_{ne} > 2a_{eq}}$ **,** $\mathrm{Re}[i\omega] > 0$ $\longrightarrow$ **Instability**

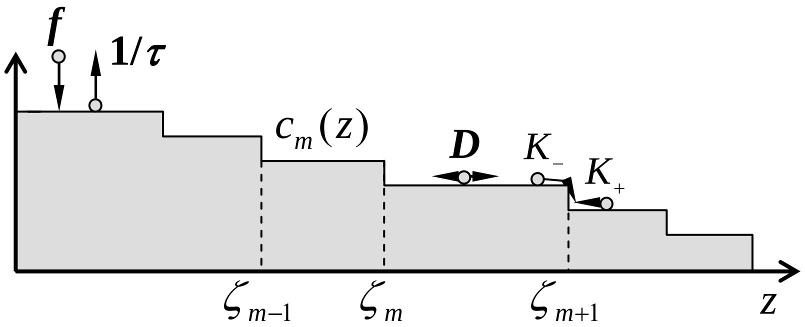
$$r \equiv l^3 / 3A(x_s / l) \sinh(2l / x_s) > 1$$

The most unstable mode $\phi = \pi$

等間隔ステップの安定性(両側モデル)

$$v_{n\pm} = \Omega \frac{D}{x_s} \frac{\left(\cosh\left(\frac{l_{n\pm}}{x_s}\right) + \frac{d_\mp}{x_s}\sinh\left(\frac{l_{n\pm}}{x_s}\right)\right)(c_\infty - c_n) - (c_\infty - c_{n\pm1})}{\left(\frac{d_+}{x_s} + \frac{d_-}{x_s}\right)\cosh\left(\frac{l_{n\pm}}{x_s}\right) + \left(1 + \frac{d_- d_+}{x_s^2}\right)\sinh\left(\frac{l_{n\pm}}{x_s}\right)}$$

where $x_s = \sqrt{D\tau}, \quad d_\pm = D/K_\pm$



Linear stability analysis $\left(y_m(t) = v_0 t + ml + \delta y_k e^{w_k t + ikml}\right)$

$$\omega_k = -\left(\frac{\pi^2}{2l} + \frac{1}{2}\frac{d_- - d_+}{d_+ + d_-} v^0 l\right)k^2 - \frac{\pi^2 x_s}{2(d_+ + d_-)}k^4 + iv^0\left[1 - \frac{1}{6}(kl)^2\right]k$$

ステップの疎密波の伝播 $v^0 - \mathrm{Im}(\omega_k)/k \approx \frac{1}{6}(kl)^2 v^0$

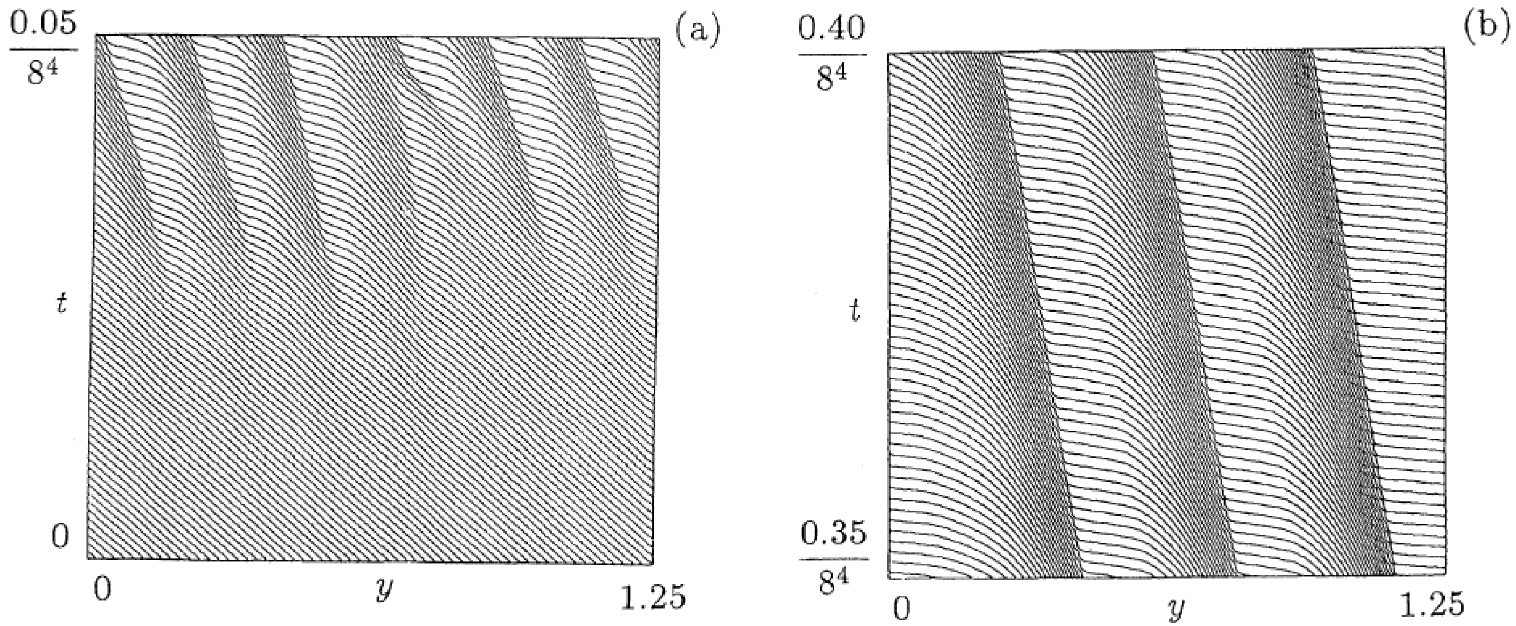
$$d_- < d_+, v^0 > v_c \longrightarrow -\left(\frac{\pi^2}{2l} + \frac{1}{2}\frac{d_- - d_+}{d_+ + d_-} v^0 l\right) > 0 \longrightarrow \textbf{Instability}$$

$k_{\max}$ はES効果（$K_+/K_- = d_-/d_+$）が大きくなると短波長にシフト

上羽牧夫：結晶成長のダイナミクスとパターン形成（培風館、2008）

等間隔ステップの安定性(両側モデル)

$$v_{n\pm} = \Omega \frac{D}{x_s} \frac{\left(\cosh\left(\frac{l_{n\pm}}{x_s}\right) + \frac{d_{\mp}}{x_s} \sinh\left(\frac{l_{n\pm}}{x_s}\right) \right)(c_\infty - c_n) - (c_\infty - c_{n\pm 1})}{\left(\frac{d_+}{x_s} + \frac{d_-}{x_s}\right)\cosh\left(\frac{l_{n\pm}}{x_s}\right) + \left(1 + \frac{d_- d_+}{x_s^2}\right)\sinh\left(\frac{l_{n\pm}}{x_s}\right)}$$ の数値解



M. Sato and M. Uwaha, Phys. Rev. B 51, 11172 (1995).

Benny方程式

$$v_{n\pm} = \frac{\left(\cosh(l_{n\pm}) + \lambda_{\mp}\sinh(l_{n\pm})\right) f_n - f_{n\pm 1}}{(\lambda_+ + \lambda_-)\cosh(l_{n\pm}) + (1 + \lambda_+\lambda_-)\sinh(l_{n\pm})}$$

Linearized equation for the step density variation

$$\frac{\partial \tilde{\rho}}{\partial t} = -\left(\frac{d\xi}{d\rho_0} + \frac{1}{2}\frac{\lambda_- - \lambda_+}{\lambda_+ + \lambda_-}\frac{f_0}{\rho_0^2}\right)\frac{\partial^2 \tilde{\rho}}{\partial z^2} + \frac{1}{6}\frac{f_0}{\rho_0^3}\frac{\partial^3 \tilde{\rho}}{\partial z^3} - \left(\frac{1}{\lambda_+ + \lambda_-}\frac{1}{\rho_0}\frac{d\xi}{d\rho_0} + \frac{1}{24}\frac{\lambda_- - \lambda_+}{\lambda_+ + \lambda_-}\frac{f_0}{\rho_0^4}\right)\frac{\partial^4 \tilde{\rho}}{\partial z^4}$$

Dispersion eq. $\omega_k = -\left(\frac{d\xi}{d\rho_0} + \frac{1}{2}\frac{\lambda_- - \lambda_+}{\lambda_+ + \lambda_-}\frac{f_0}{\rho_0^2}\right)k^2 - \frac{1}{\lambda_+ + \lambda_-}\frac{1}{\rho_0}\frac{d\xi}{d\rho_0}k^4 - i\frac{1}{6}\frac{f_0}{\rho_0^3}k^3$

$$f_0 \le f_c = -2\frac{\lambda_+ + \lambda_-}{\lambda_- - \lambda_+}\rho_0^2\frac{d\xi}{d\rho_0} \longrightarrow$$ **Instability**

$$\varepsilon \equiv \left|1 - \frac{f_c}{f_0}\right| \xrightarrow[k_{\max} \sim \sqrt{\varepsilon},\quad \mathrm{Re}\,\omega_k \sim \varepsilon^2]{} Z \equiv \sqrt{\varepsilon} z, \quad T \equiv \varepsilon^2 t$$

$$\xrightarrow{\tilde{\rho} \equiv \varepsilon^{3/2} N}$$

$$\frac{\partial N}{\partial \tilde{T}} + \frac{1}{3\sqrt{\varepsilon}}\frac{\lambda_- + \lambda_+}{\lambda_+ - \lambda_-}\frac{1}{\rho_0}\frac{\partial^3 N}{\partial \tilde{Z}^3} + \left(\frac{1}{\lambda_+ + \lambda_-}\frac{1}{\rho_0}\frac{\partial^4 N}{\partial \tilde{Z}^4} + \frac{\partial^2 N}{\partial \tilde{Z}^2} + \frac{4\lambda_-\lambda_+}{\lambda_- - \lambda_+}\frac{1}{\rho_0} N \frac{\partial N}{\partial \tilde{Z}}\right) = 0$$

M. Sato et al., Europhys. Lett. 32, 639 (1995).

Benny方程式

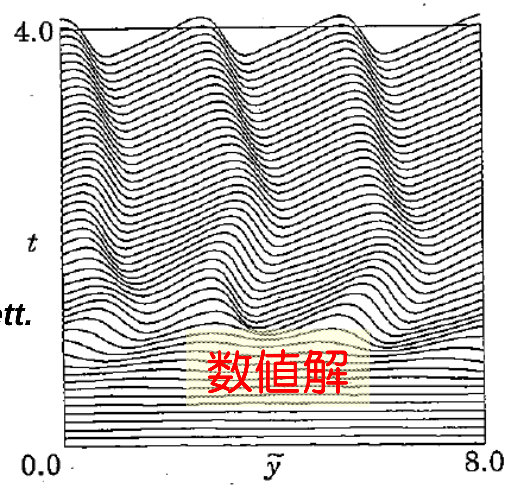
Benny equation

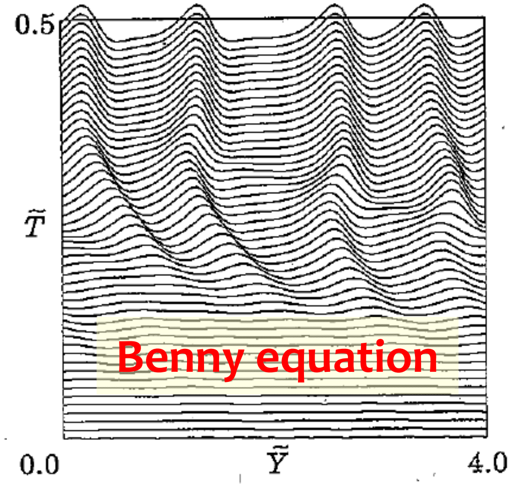
$$\frac{\partial N}{\partial T}+\frac{\partial^4 N}{\partial Z^4}+\delta\frac{\partial^3 N}{\partial Z^3}+\frac{\partial^2 N}{\partial Z^2}+N\frac{\partial N}{\partial Z}=0$$

where $\delta \equiv \frac{1}{3}\frac{d_- + d_+}{d_- - d_+}\sqrt{\frac{d_- + d_+}{n^0 x_s^2}}\frac{1}{\sqrt{\varepsilon}}$

上羽牧夫：結晶成長のダイナミクスとパターン形成（培風館、2008）

M. Sato et al., Europhys. Lett. 32, 639 (1995).





KS equation

$$\frac{\partial y}{\partial t}=-\frac{\partial^2 y}{\partial x^2}-\frac{\partial^4 y}{\partial x^4}-\frac{1}{2}\left(\frac{\partial y}{\partial x}\right)^2 \longrightarrow \frac{\partial y'}{\partial t}=-\frac{\partial^2 y'}{\partial x^2}-\frac{\partial^4 y'}{\partial x^4}-y'\frac{\partial y'}{\partial x}$$

KS equation=Benny equation with δ=0

When δ is large,

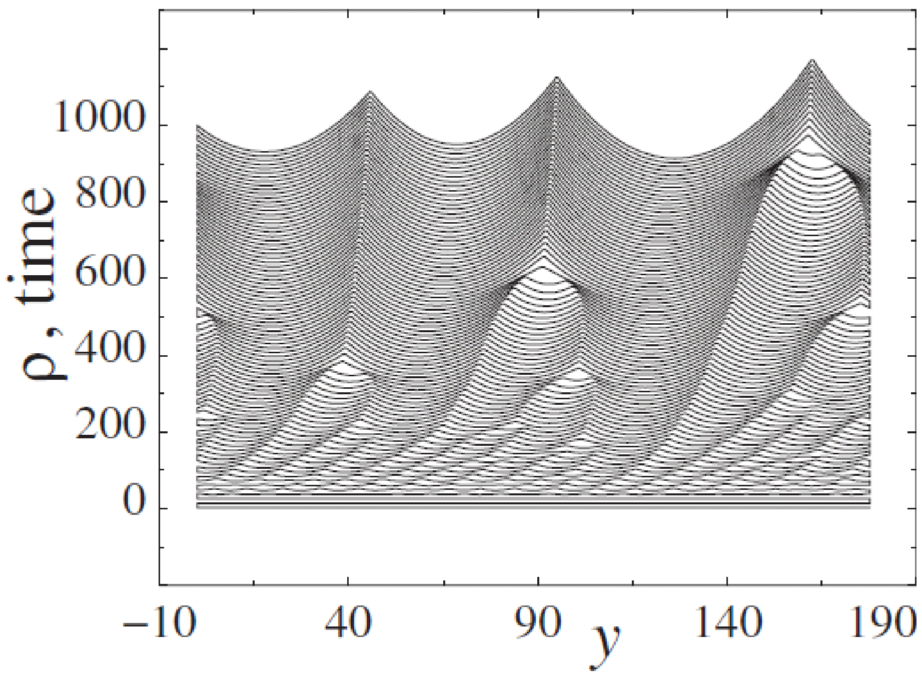
$$\frac{\partial N}{\partial T}+\frac{\partial^4 N}{\partial Z^4}+\delta\frac{\partial^3 N}{\partial Z^3}+\frac{\partial^2 N}{\partial Z^2}+N\frac{\partial N}{\partial Z}=0 \longrightarrow \frac{\partial N}{\partial T}+\delta\frac{\partial^3 N}{\partial Z^3}+N\frac{\partial N}{\partial Z}=0$$

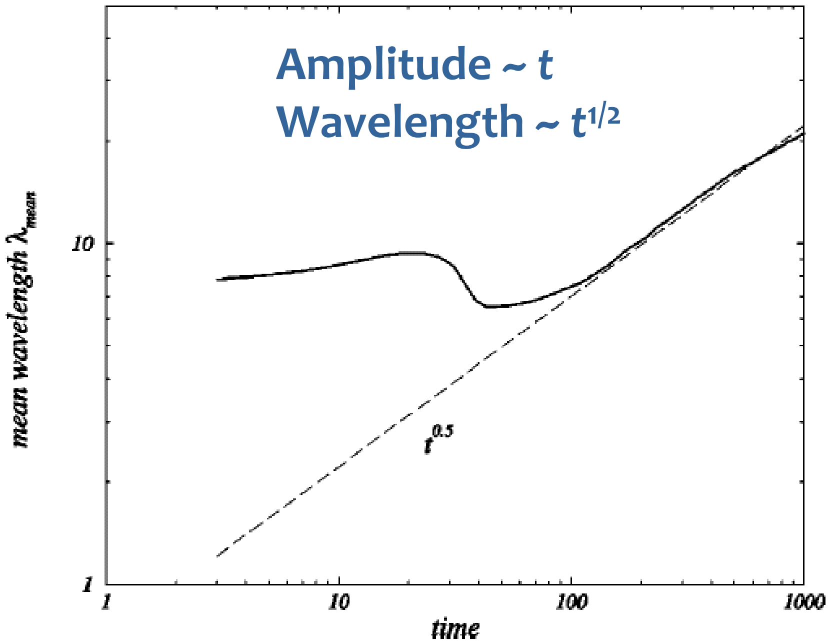
Korteweg-de Vries (KDV) equation

保存系のBenny方程式

In the limit of large desorption length and weak ES effect,

$$\frac{\partial \zeta}{\partial t} = -\frac{\partial^2}{\partial z^2}\left[\zeta + b\frac{\partial \zeta}{\partial z} + \frac{\partial^2 \zeta}{\partial z^2} - \left(\frac{\partial \zeta}{\partial z}\right)^2\right]$$

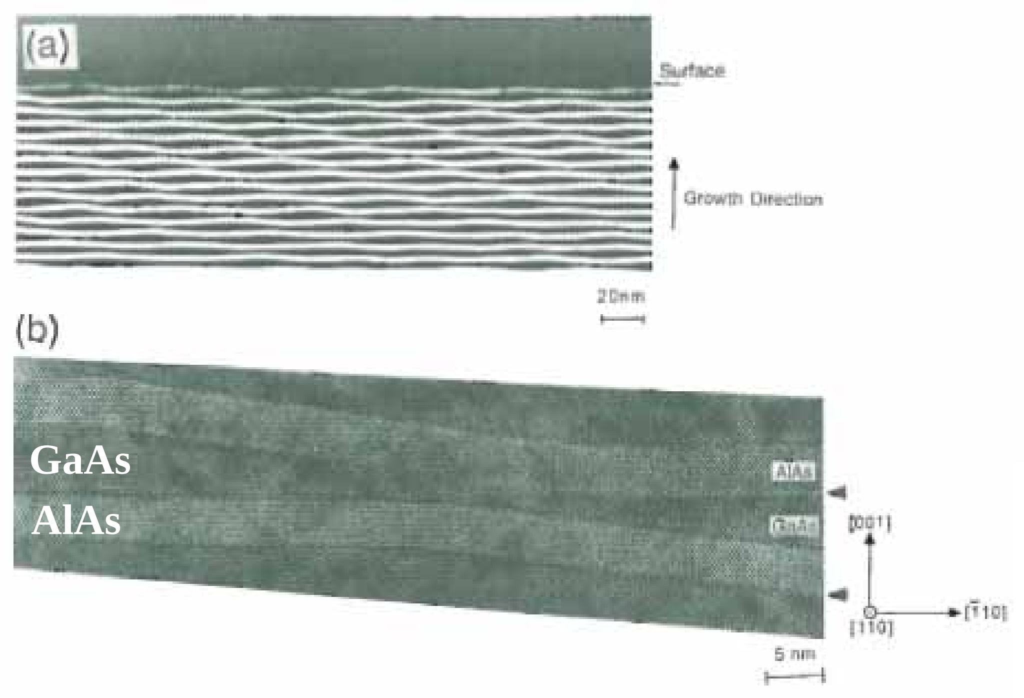




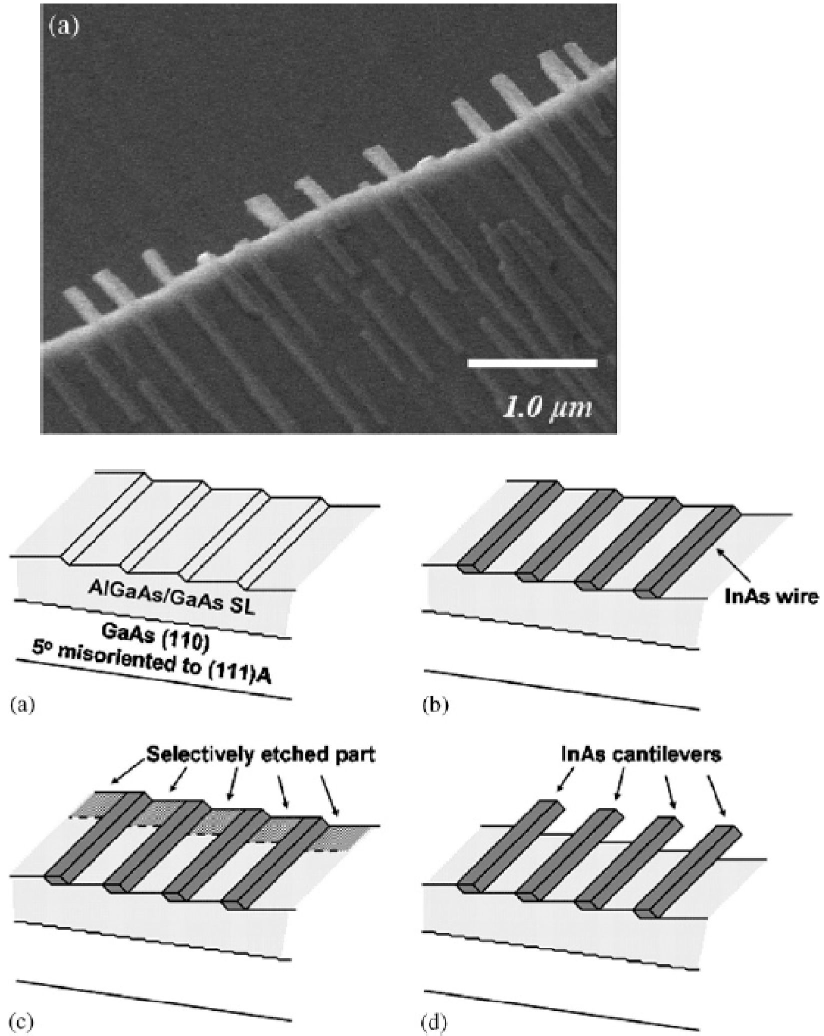
F. Gillet et al., PRB 63, 241401(R) (2001).

成長中のステップバンチング(実験)

Anti-ES effect?



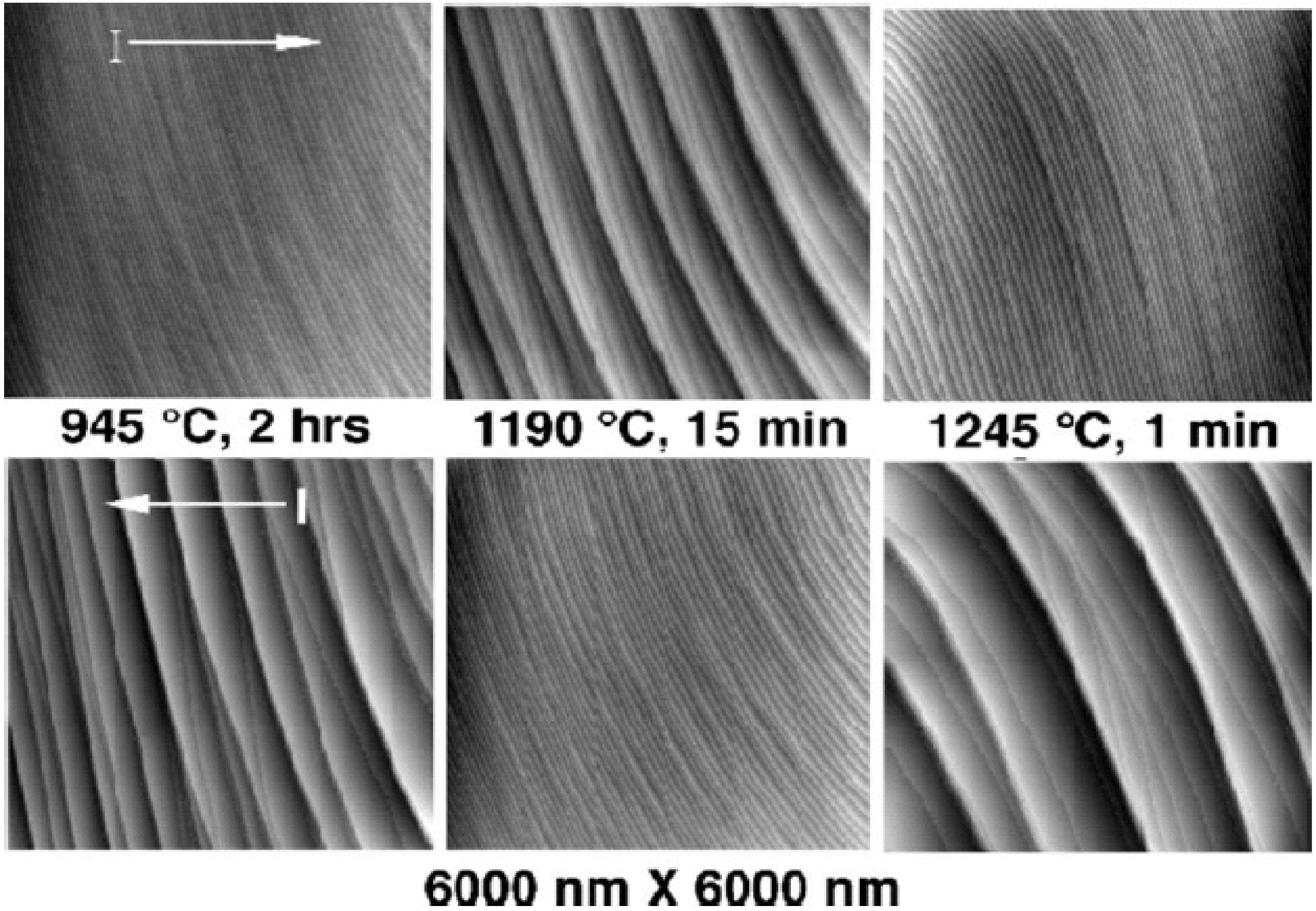
M. Kasu and N. Kobayashi, Appl. Phys. Lett. 62, 1262 (1993).



H. Yamaguchi and Y. Hirayama, J. Crystal Growth 251, 281 (2003).

通電加熱によるステップバンチング

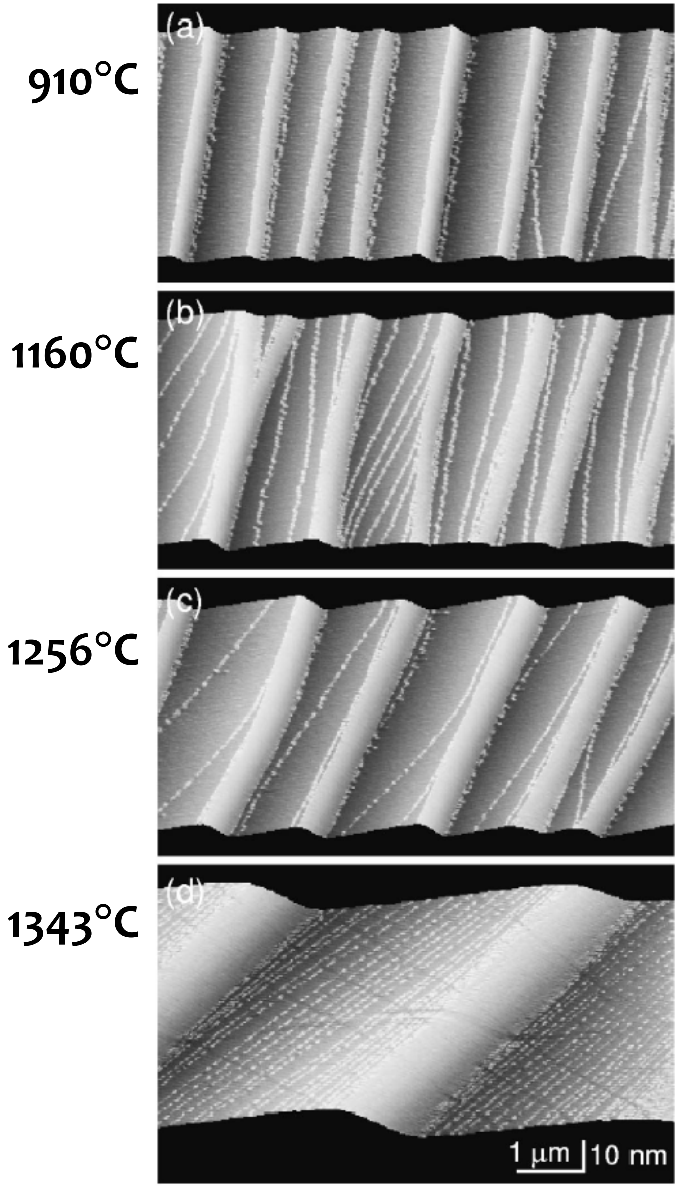
STM images of Si(111)

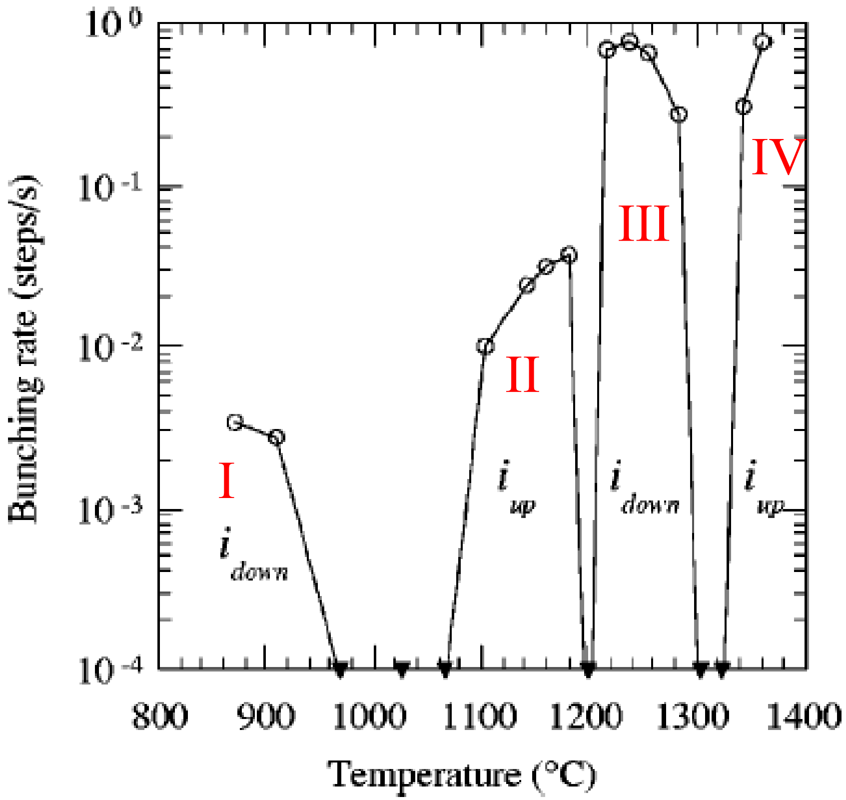


Y. N. Yang et al., Surf. Sci. 356, 101 (1996).

通電加熱によるステップバンチング

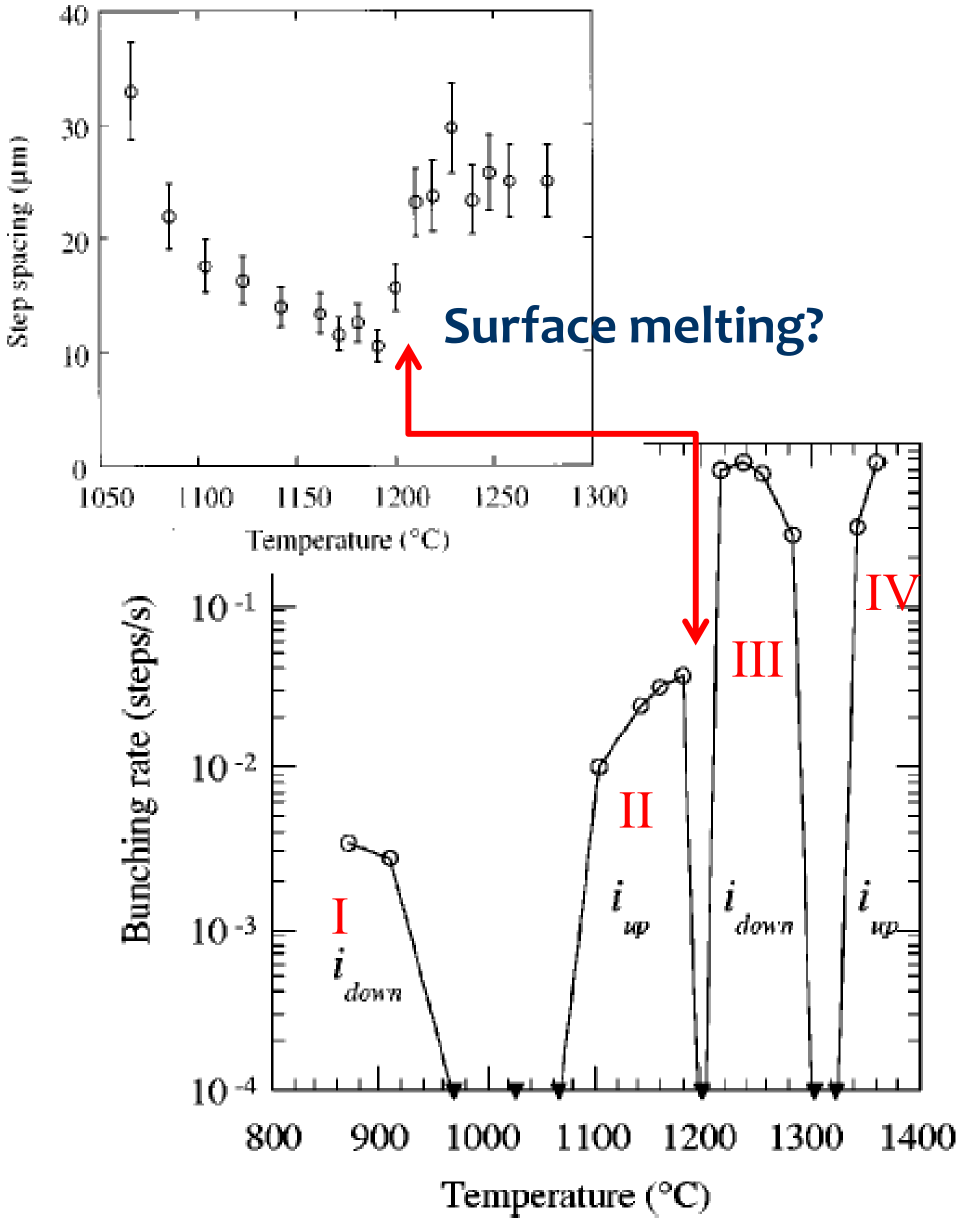
AFM images of Si(111)



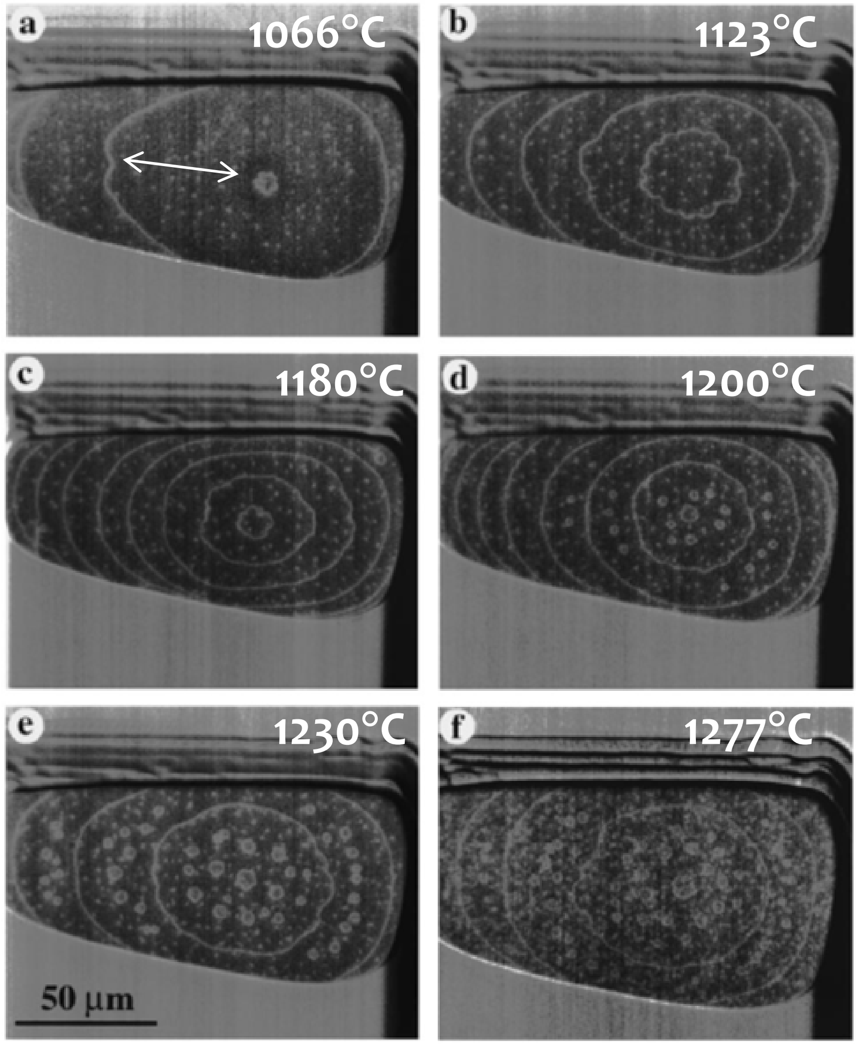


Y. Homma and N. Aizawa, Phys. Rev. B 62, 8323 (2000).

通電加熱によるステップバンチング



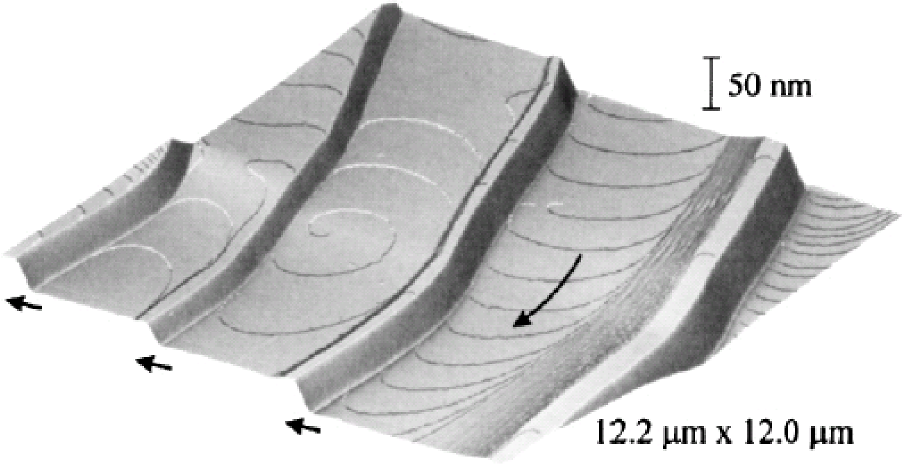
Y. Homma and N. Aizawa, Phys. Rev. B 62, 8323 (2000).

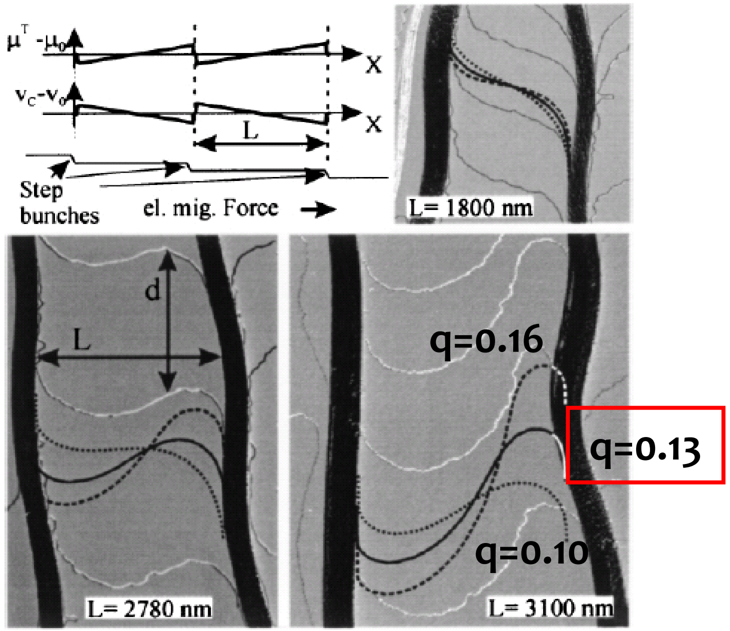


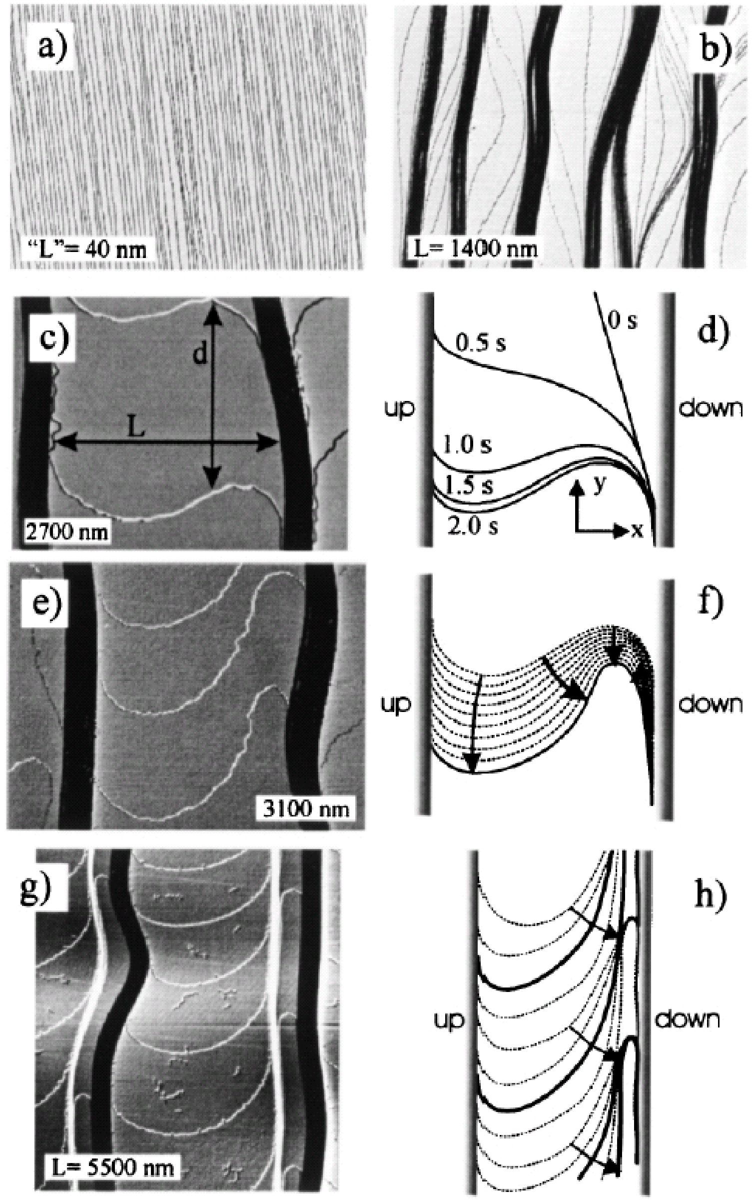
Y. Homma, H. Hibino et al., Phys. Rev. B 55, R10237 (1997).

通電加熱によるステップアンチバンディング

Step Antibanding Instability







K. Thurmer et al., PRL 83, 5531 (1999).

ドリフトによるステップバンチング

境界条件

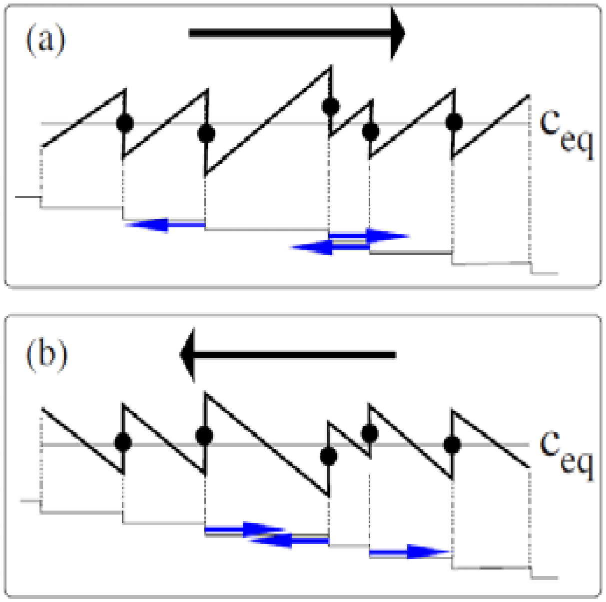
$$\vec{J}_{+}\cdot\vec{n} = -D\partial_z c_{+} + Dc_{+}/\xi = -\overline{K}\left(c_{+} - c_{eq}\right)$$

where $\xi = k_B T/qeE$

$$\vec{J}_{-}\cdot\vec{n} = -D\partial_z c_{-} + Dc_{-}/\xi = \overline{K}\left(c_{-} - c_{eq}\right)$$

$$V = -\Omega\left(\vec{J}_{+} - \vec{J}_{-}\right)\cdot\vec{n} = \Omega\overline{K}\left[\left(c_{+} - c_{eq}\right) + \left(c_{-} - c_{eq}\right)\right] = 2\Omega\overline{K}\left(\bar{c} - c_{eq}\right)$$

Bunching



Debunching

拡散方程式

$$D\partial_{zz}c - \left(D/\xi\right)\partial_z c = 0$$

$$\vec{J}_{+} \approx 0 \longrightarrow -\partial_z c + c/\xi = 0$$

$$\xrightarrow[c \approx c_{eq}]{} \partial_z c = c_{eq}^{0}/\xi \longrightarrow c = c_{eq}^{0}\left(1 + z/\xi\right)$$

$$\bar{c} = c_{eq}^{0}\left(1 + \left(l_{-} - l_{+}\right)/4\xi\right)$$

$$V_m \approx \left(\Omega c_{eq}^{0}\overline{K}/2\xi\right)\left(l_{-} - l_{+}\right) = \left(\Omega c_{eq}^{0}\overline{K}/2\xi\right)\left(\zeta_{m+1} + \zeta_{m-1} - 2\zeta_{m}\right)$$

C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

ドリフト流によるステップバンチング

拡散方程式 $$D\frac{\partial^2 c}{\partial z^2} - v_d \frac{\partial c}{\partial z} = 0$$

Impermeable step → No ES effect

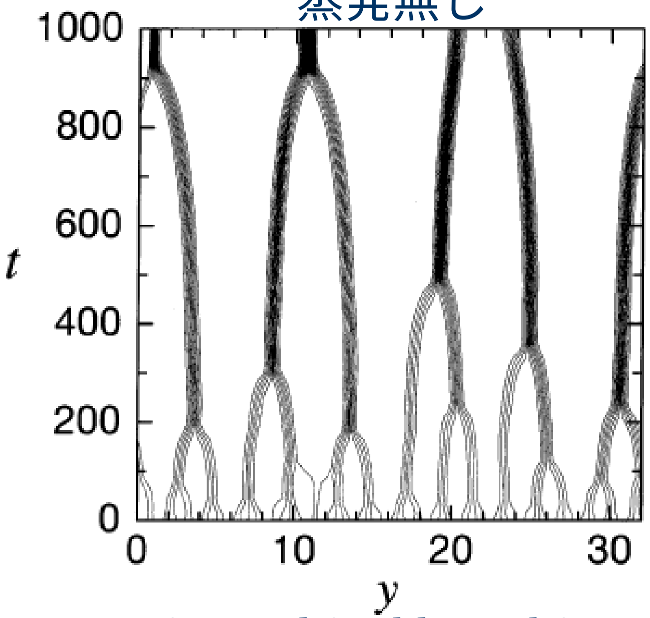
$$j_n = K\frac{\left(c_n e^{v_d l_n/D} - c_{n+1}\right)v_d}{\left(e^{v_d l_n/D} - 1\right)K + \left(e^{v_d l_n/D} + 1\right)v_d}$$

$$v_n = \Omega\left(j_{n-1} - j_n\right)$$

下段向きの流れ $v_d > 0$ では、$l_{n+1} > l_n$ のとき $j_{n+1} > j_n$ ⟶ ステップバンチング

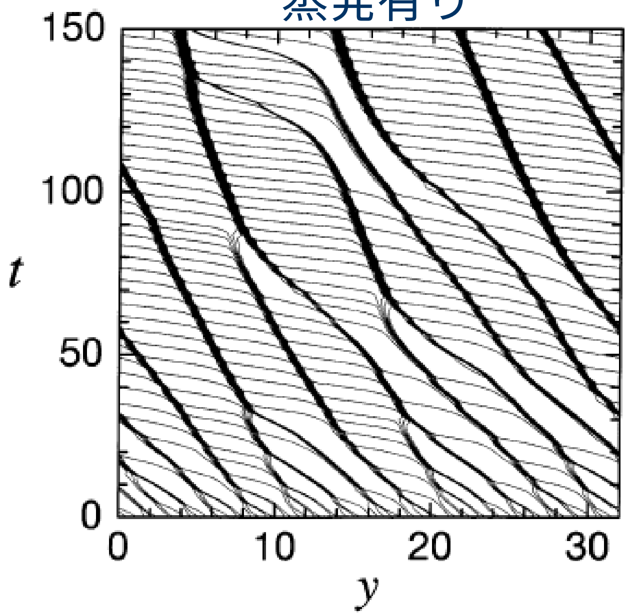
上羽牧夫：結晶成長のダイナミクスとパターン形成（培風館、2008）

蒸発無し



Hierarchical bunching

蒸発有り



M. Sato, M. Uwaha, Surface Science 442 (1999) 318–328.

ドリフト流によるステップバンチング

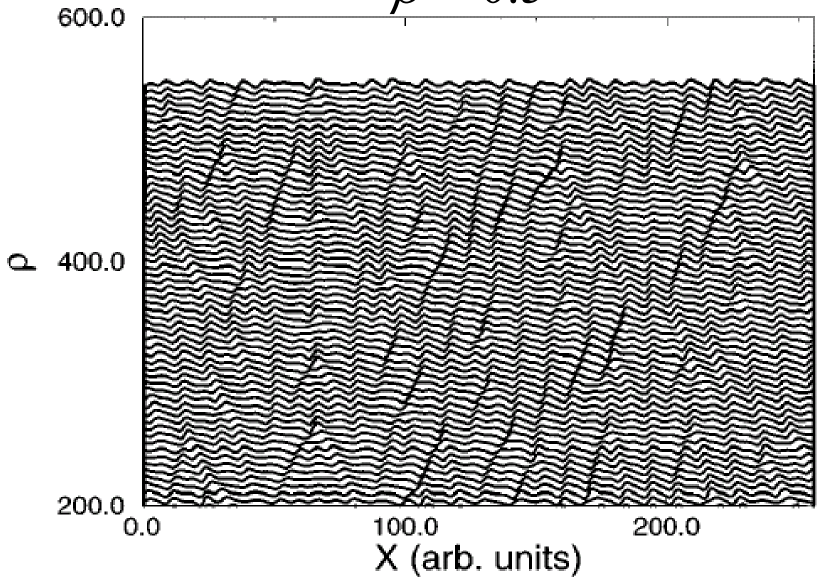
非線形方程式

非保存系 ——→ Benny方程式

$$\frac{\partial \rho}{\partial T} = -\frac{\partial^4 \rho}{\partial Z^4} - \beta \frac{\partial^3 \rho}{\partial Z^3} - \frac{\partial^2 \rho}{\partial Z^2} - \rho \frac{\partial \rho}{\partial Z}$$

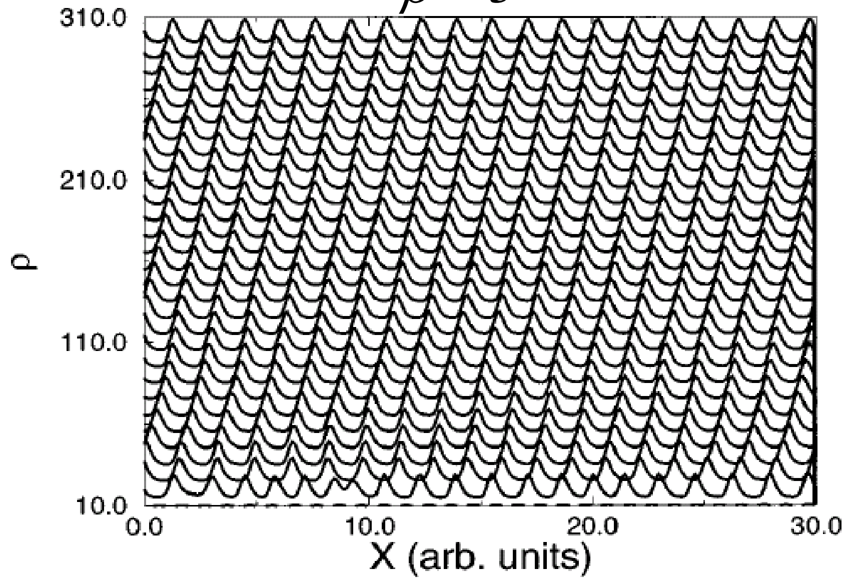
$$\begin{aligned}(1/\Omega D c_{eq})[W_t - (\Omega c_{eq}/\tau) W_x] &= -\beta_2 W_{xx} - \left(\frac{l_0^2 \beta_2}{12} + \frac{A}{2 l_0^4 d \Omega c_{ceq}}\right) W_{xxxx} \\ &\quad - \left(\frac{1}{6 x_s^2} - \frac{A}{2 \Omega c_{eq} \xi d l_0^3}\right) W_{xxx}\end{aligned}$$

$\beta = 0.5$



Spatiotemporal chaos

$\beta = 3$



"Soliton"

C. Misbah and O. Pierre-Louis, PRE 53, R4318 (1996).

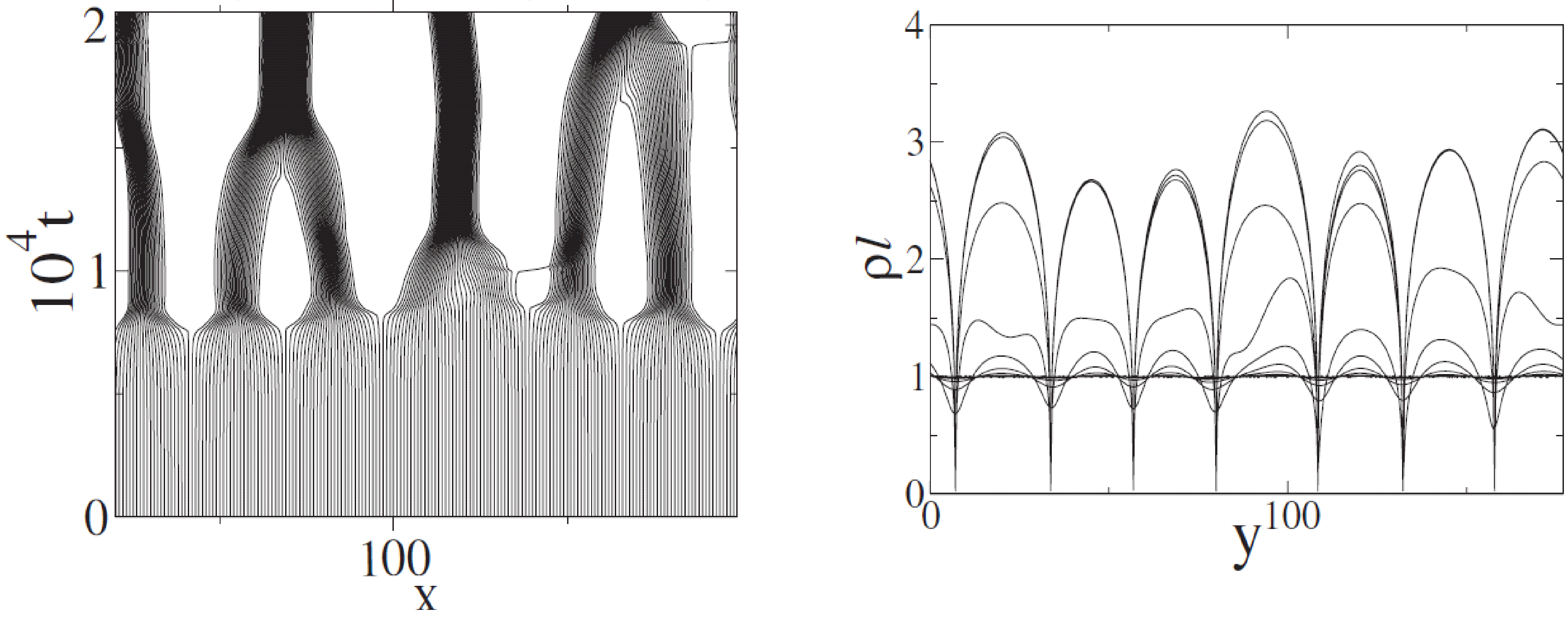
ドリフト流によるステップバンチング

非線形方程式

保存系 ⟶ $$\partial_t \zeta = -a\partial_z \left[\frac{\Omega D c_{eq}^0}{1+d\rho} \left(\frac{1}{\xi} - A\rho a^2 \partial_{zz} \rho^3 \right) \right]$$

where $\rho = 1/(l + a\partial_z \zeta)$

No tangential matching (faceting)

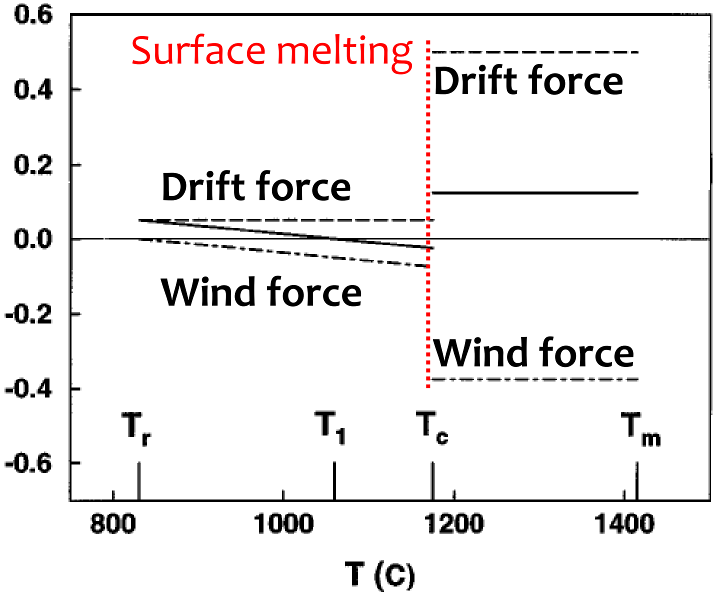


J. Chang et al., PRL 96, 195901 (2006).

直接通電加熱中のステップバンチングモデル

Balance between drift and wind forces

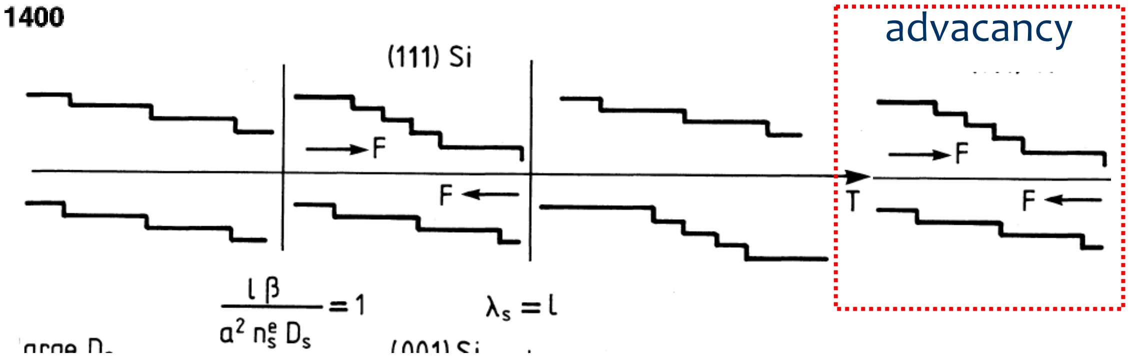
D. Kandel and E. Kaxiras, Phys. Rev. Lett. 76, 1114 (1996).



$$D_s \frac{d^2 n_s}{dx^2} - \frac{D_s F}{kT} \frac{dn_s}{dx} - \frac{n_s}{\tau_s} = 0,$$

Incorporation of advacancy

C. Misbah et al., Phys. Rev. B 51, 17283 (1995).



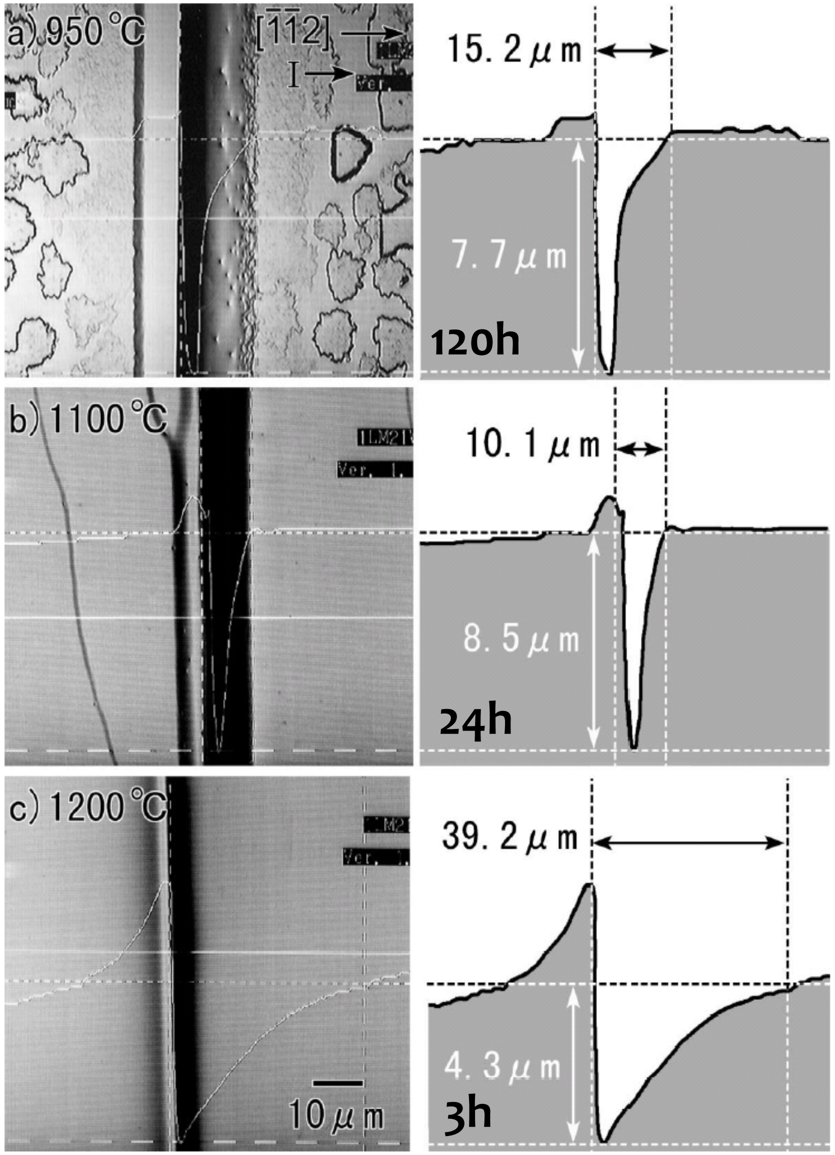
Changes in permeability

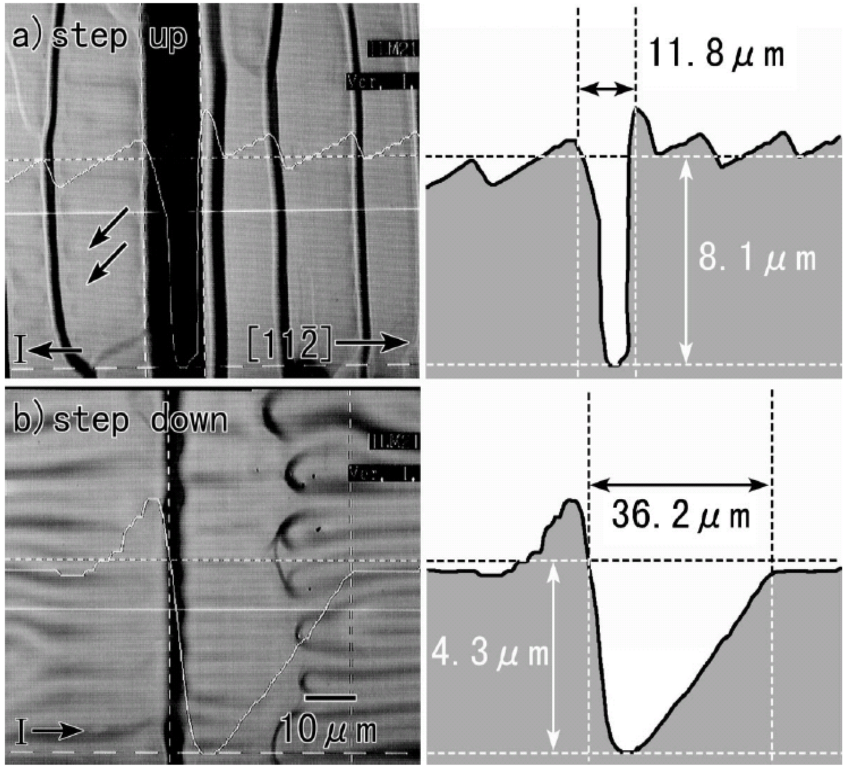
An isolated bunch of permeable steps is stable with the step-up drift.
Impermeable in ranges I and III
Permeable steps in range II

S. Stoyanov, Surf. Sci. 416, 200 (1998); M. Sato et al., Phys. Rev. B 62, 8452 (2000).

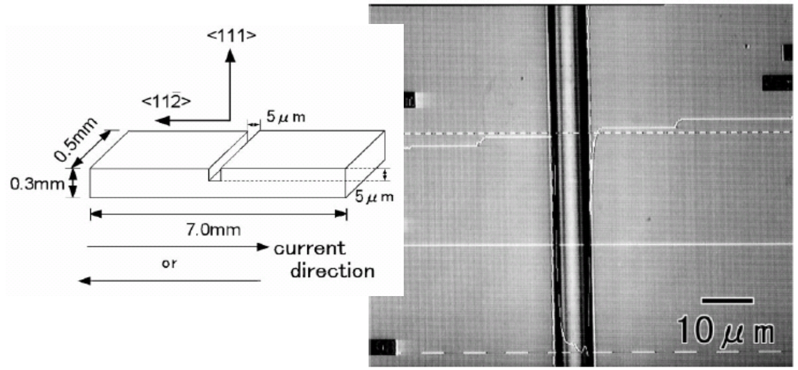
吸着子の帯電状態

Si(111) optical microscopy images





1160°C, 24h

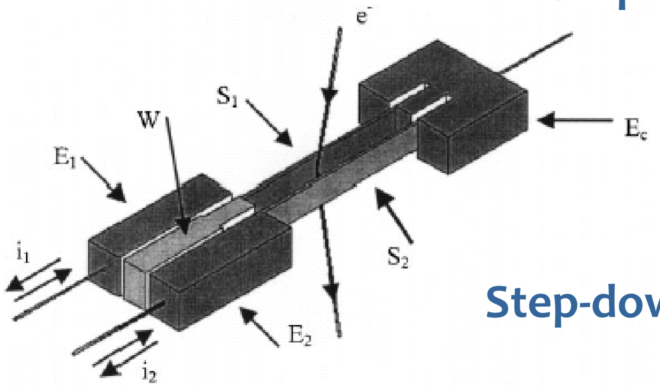


M. Deagawa et al., Surf. Sci. 461, L528 (2000).

電流方向と昇華/成長とバンチングの関係

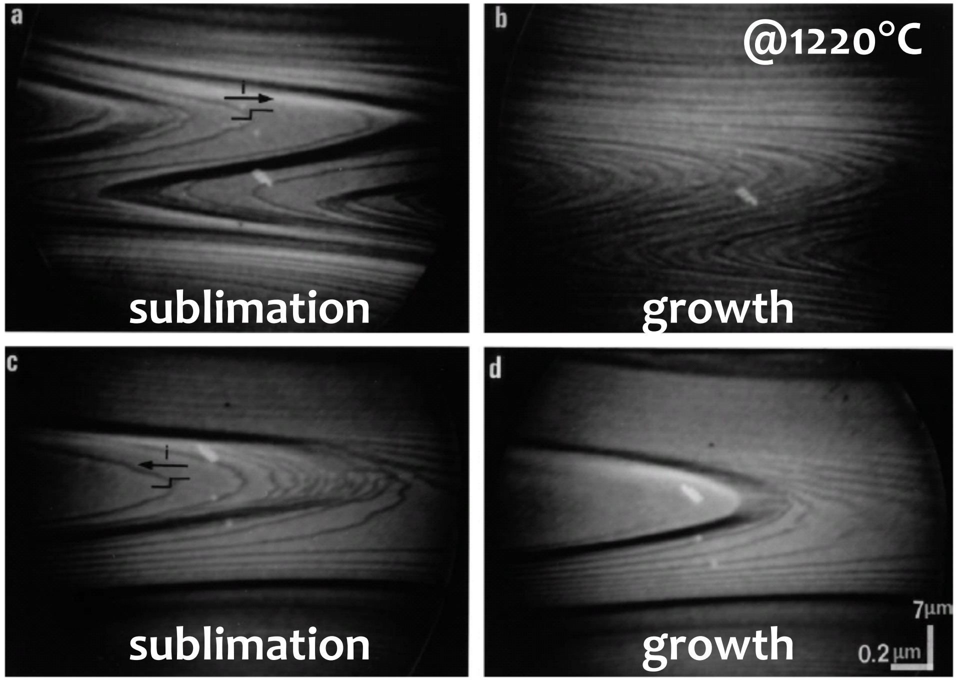
Si(111) REM images

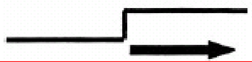
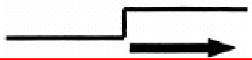
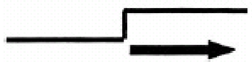
Step-up current



Step-down current

J. J. Metois et al., Surf. Sci. 440, 407 (1999).

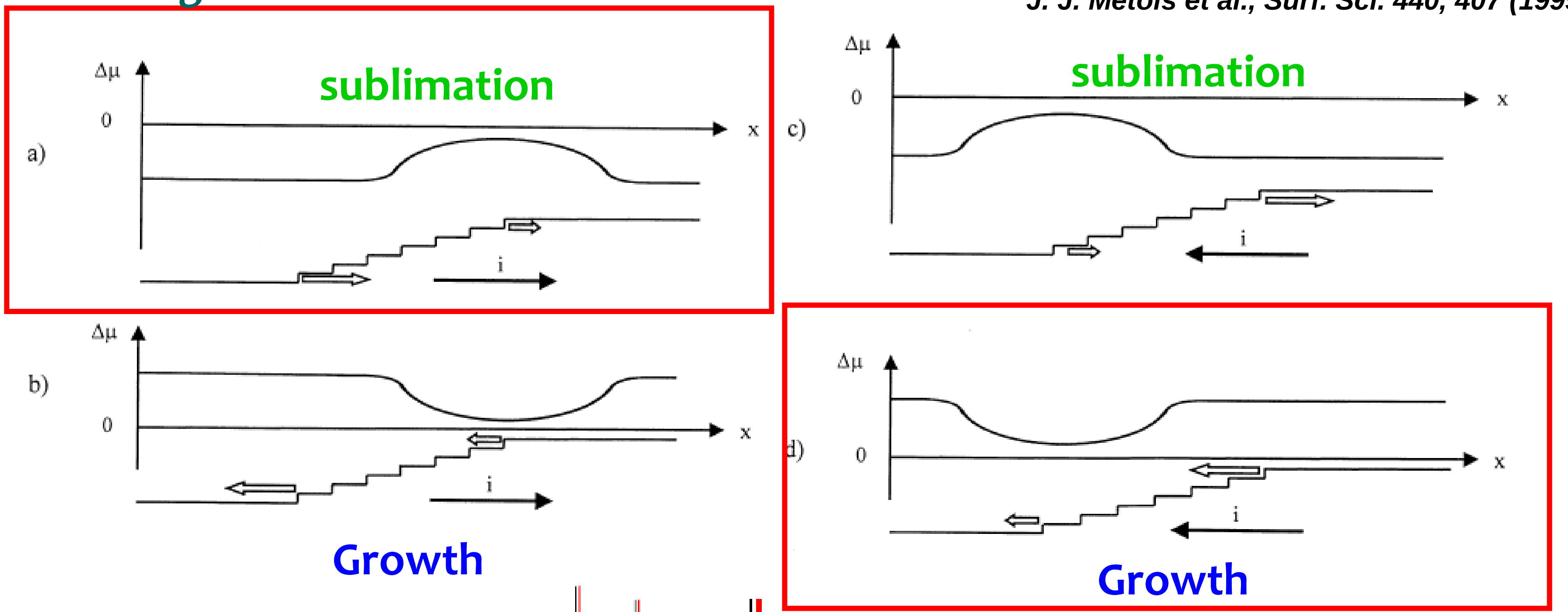


Temperature domains	Current direction[a]	Deposition	Sublimation
Low Temperature (data from Ref. [2])		Bunching	Bunching
		No Bunching	No Bunching
Middle Temperature		Bunching	No Bunching
		No Bunching	Bunching
High Temperature		Bunching	Bunching
		No Bunching	No Bunching

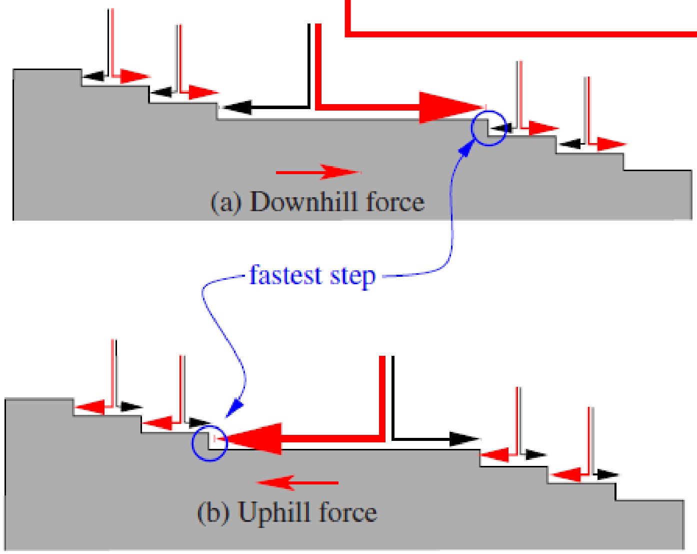
透過ステップの運動

Bunching

J. J. Metois et al., Surf. Sci. 440, 407 (1999).



Bunching



C. Misbah et al., Rev. Mod. Phys. 82, 981 (2010).

ドリフトによるステップ不安定化

TABLE I. Conditions to induce instabilities.

Growth condition	Permeability	Terrace	Kinetics	Drift	Instability
sublimation	permeable	isolated	fast/slow	down	wandering
		$l/x_s \ll 1$	fast/slow	down	wandering
			not too slow ($\lambda \ll x_s/l$)	up	bunching
	impermeable	isolated	fast/slow	down	wandering
		$\alpha l/x_s \gg 1$	fast/slow	up	bunching
				down	wandering
		$\alpha l/x_s \ll 1$	fast/slow	down	wandering/bunching
growth	permeable	isolated	fast/slow	up	wandering
		$l/x_{ms} \ll 1$	fast/slow	up	wandering
			not too slow ($\lambda \ll x_s/l$)	down	bunching
	impermeable	isolated	fast ($\lambda \ll 1$)	up	wandering
			slow ($\lambda \gg 1$)	down	wandering
		$\alpha l/x_s \gg 1$	fast ($\lambda \ll l/x_s$)	down	bunching
			slow ($\lambda \gg l/x_s$)	down	wandering /bunching
		$\alpha l/x_s \ll 1$	fast ($\lambda \ll l/x_s$)	up	wandering
				down	bunching
			slow ($\lambda \gg l/x_s$)	down	wandering/bunching

$$x_s = \sqrt{D\tau}, \quad d_\pm = D/K_\pm, \quad \lambda_\pm = d_\pm/x_s$$

M. Sato et al., PRB 62, 8452 (2000).

透過ステップの対形成

拡散方程式

$$D\partial_{zz}c - \frac{D}{\xi}\partial_z c + F - \frac{1}{\tau} = 0$$

境界条件

$$D\partial_z c_{\pm} - \frac{D}{\xi}c_{\pm} = \pm\nu_{\pm}\left(c_{\pm} - c_{eq}\right) + \nu_0\left(c_{+} - c_{-}\right)$$

平衡濃度

$$c_{eq}^{n} = c_{eq}^{0}\left[1 + \frac{A}{3}\left(\frac{1}{l_n^3} - \frac{1}{l_{n+1}^3}\right)\right]$$

質量保存

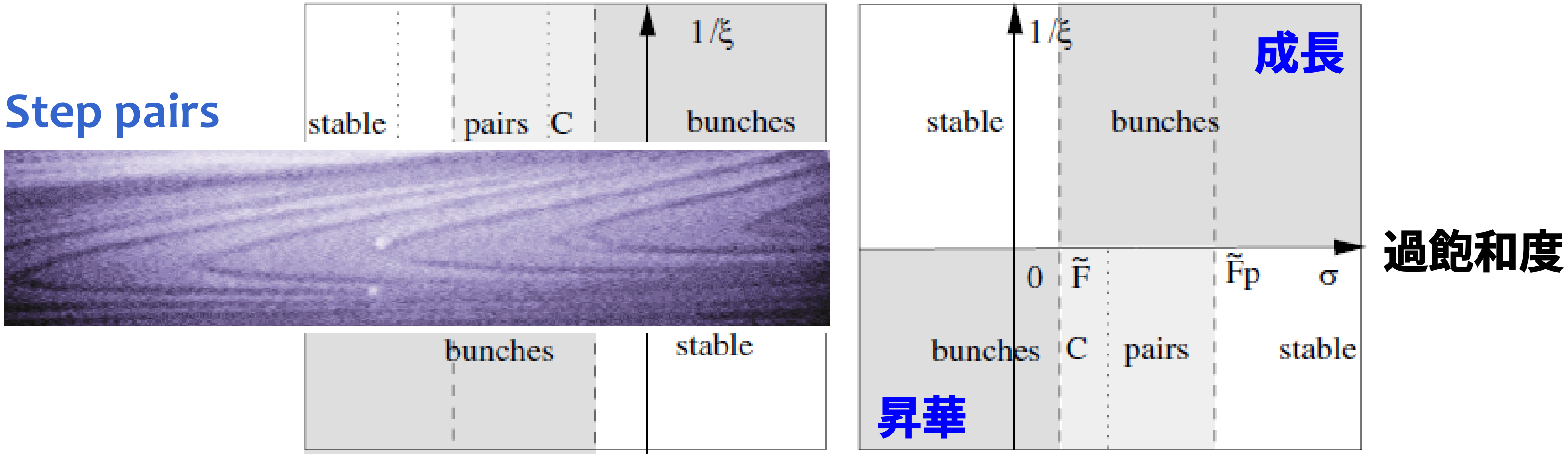
$$\frac{V}{\Omega} = \left[D\partial_z c - \frac{D}{\xi}\right]_{+} - \left[D\partial_z c - \frac{D}{\xi}\right]_{-}$$

→ **分散関係** $\mathrm{Re}[i\omega] = 2[1 - \cos\phi]B_2 - 4[1 - \cos\phi]^2 B_4$

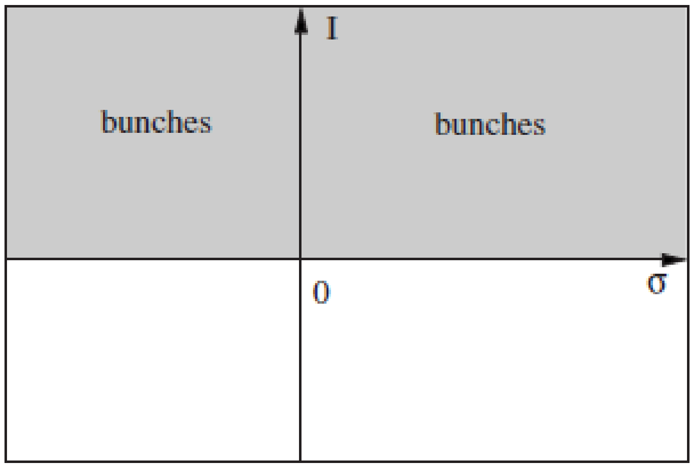
O. Pierre-Louis et al., PRL 93, 165901 (2004).

透過ステップの対形成

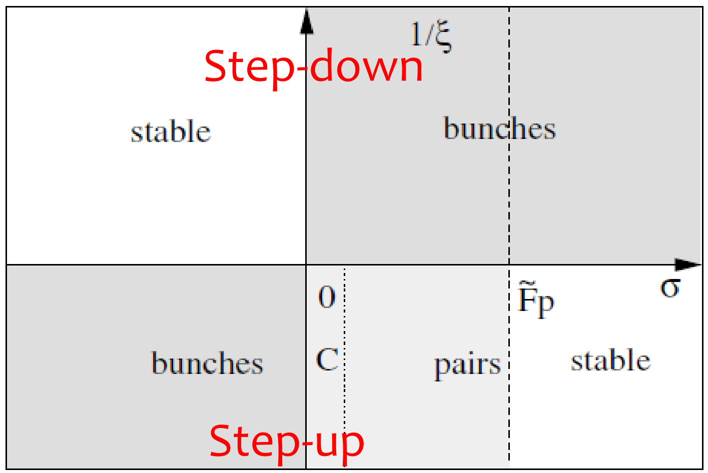
$\xi^{-1} = F/kT$



$d_0 > 0$ $d_0 = D/\nu_0$ $d_0 < 0$



Regime I



Regime II

O. Pierre-Louis et al., PRL 93, 165901 (2004).

Si(001)表面でのステップ不安定化(一次元)

拡散方程式

$$D_{a(b)}\partial_{xx}c^n_{a(b)}(x,t) = -F$$

境界条件

$$c^n_a\left(x^{n-1}_b\right) = c^n_{eq,b} \quad D_a\frac{\partial c^n_a}{\partial x}\left(x^n_a\right) = -\nu\left[c^n_a\left(x^n_a\right) - c^n_{eq,a}\right]$$

$$c^n_b\left(x^n_b\right) = c^{n+1}_{eq,b} \quad D_b\frac{\partial c^n_b}{\partial x}\left(x^n_a\right) = \nu\left[c^n_b\left(x^n_a\right) - c^n_{eq,a}\right]$$

質量保存

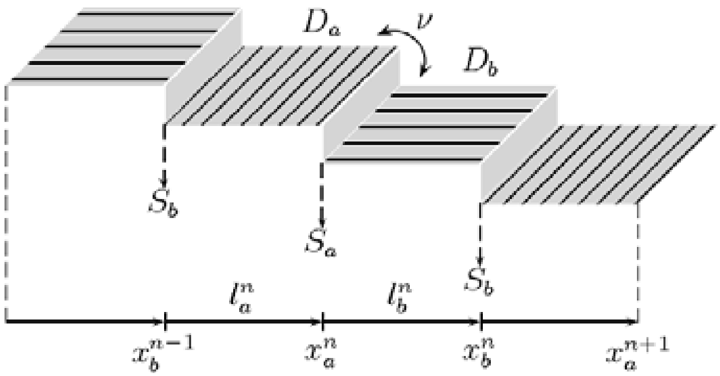
$$v^n_a = \Omega D_b\frac{\partial c^n_b}{\partial x}\left(x^n_a\right) - \Omega D_a\frac{\partial c^n_a}{\partial x}\left(x^n_a\right)$$

$$v^n_b = \Omega D_a\frac{\partial c^{n+1}_a}{\partial x}\left(x^n_b\right) - \Omega D_b\frac{\partial c^n_b}{\partial x}\left(x^n_b\right)$$

平衡濃度

$$c^n_{eq,b} = C_0 + \frac{\Omega C_0 A}{k_B T}\left(\frac{1}{l^n_a} - \frac{1}{l^{n-1}_b}\right)$$

$$c^n_{eq,a} = C_0 + \frac{\Omega C_0 A}{k_B T}\left(\frac{1}{l^n_b} - \frac{1}{l^n_a}\right)$$



T. Frisch and A. Verga, PRL 94, 226102 (2005).

Si(001)表面でのステップ不安定化(一次元)

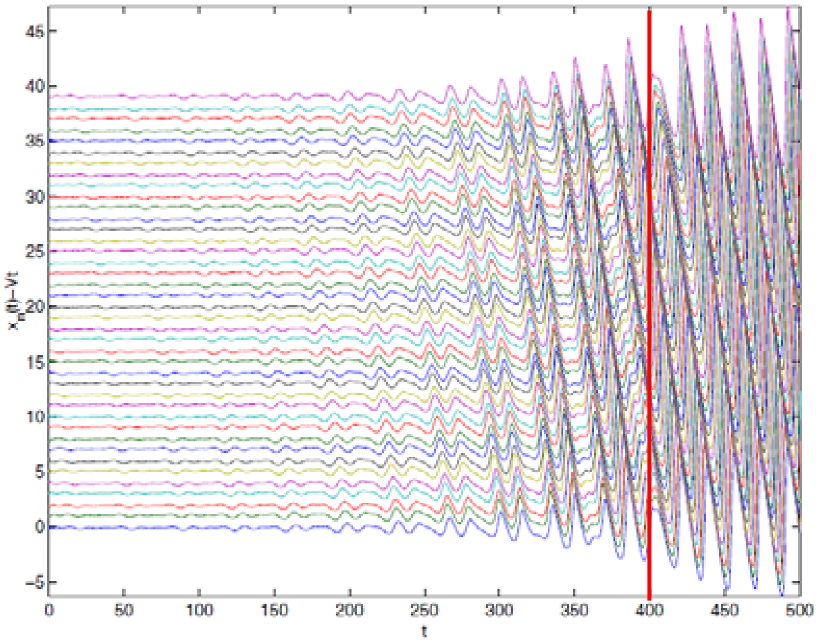
$$\sigma = Af^2k^2 - Bk^4, \qquad \omega = \frac{1}{2}fk - Cfk^3, \qquad (12)$$

where $f = Fl_a^2/\nu E$, $\alpha_a = \nu l_a/D_a$, and $\alpha_b = \nu l_a/D_b$.

$$A = \frac{1}{16}\frac{(\alpha_a + \alpha_b + 3)}{(\alpha_a + \alpha_b + 2)^2}$$

$$B = \frac{1}{1536(\alpha_a + \alpha_b + 2)^3} \times [768(1 + \alpha_a + \alpha_b) + 192(\alpha_a + \alpha_b)^2 - 24(8 + 6\alpha_a + 14\alpha_b + \alpha_a^2 + 6\alpha_a\alpha_b + 5\alpha_b^2)f - (72 + 86(\alpha_a + \alpha_b) + 19\alpha_a^2 + 80\alpha_a\alpha_b - 11\alpha_b^2)f^2]$$

$$C = \frac{1}{384}\frac{8\alpha_a + 8\alpha_b + 16 - 9(\alpha_a - \alpha_b)f}{2 + \alpha_a + \alpha_b}.$$



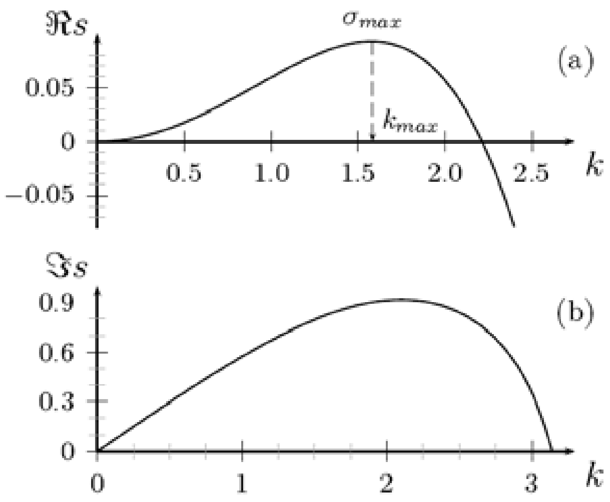
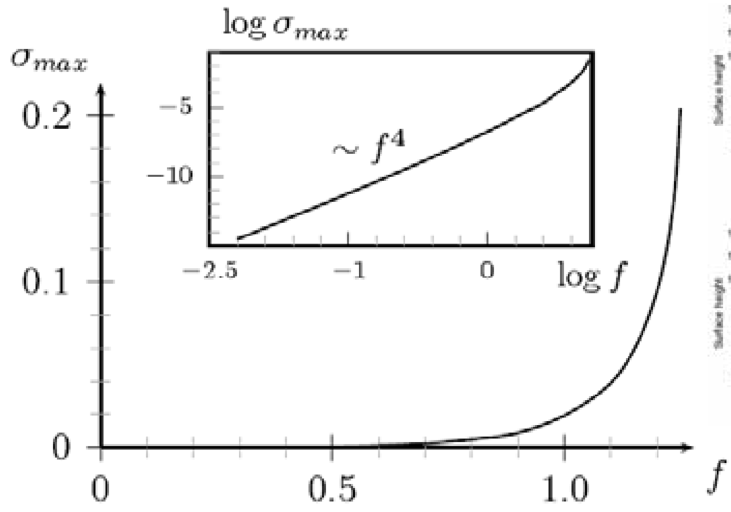
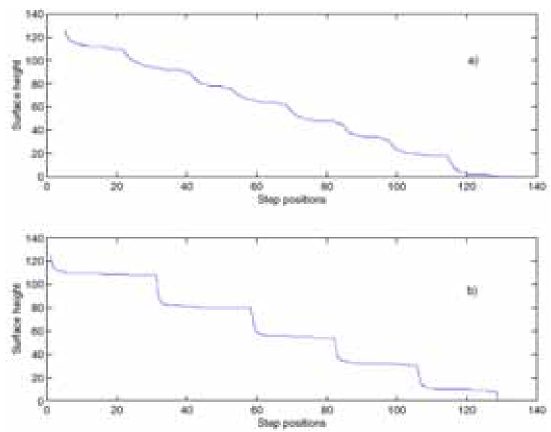


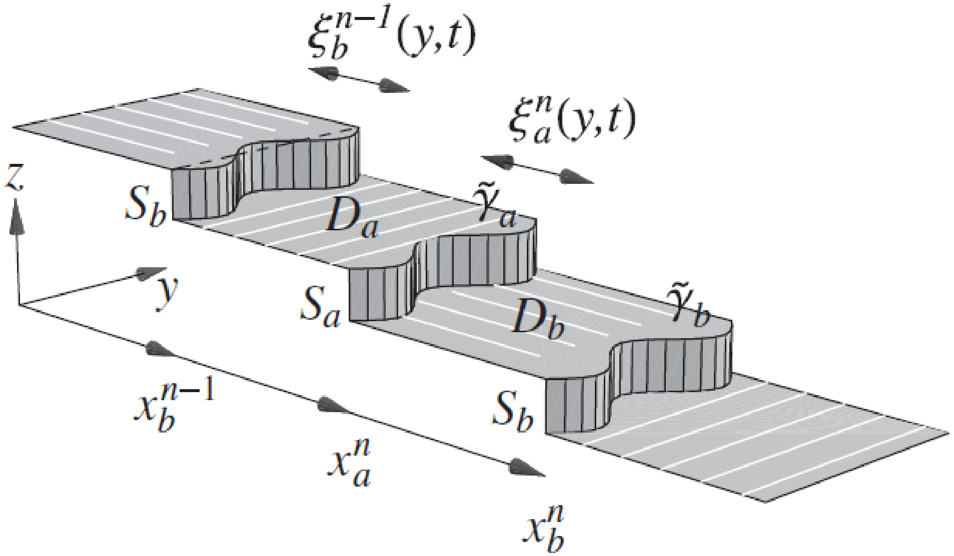
FIG. 3. The growth rate s versus k, (a) real and (b) imaginary parts, for the l_b^- branch with $\alpha_a = 1$, $\alpha_b = 0.1$, and $f = 1.2 < f_c$.





T. Frisch and A. Verga, PRL 94, 226102 (2005).

Si(001)表面でのステップ不安定化(二次元)



拡散方程式

$$D_a \partial_x^2 C_a^n + D_b \partial_y^2 C_a^n = -F,$$

$$D_b \partial_x^2 C_b^n + D_a \partial_y^2 C_b^n = -F,$$

境界条件

$$C_a^n(x_b^{n-1}) = C_{\mathrm{eq},b}^{n-1}, \qquad C_a^n(x_a^n) = C_{\mathrm{eq},a}^n,$$

$$C_b^n(x_a^n) = C_{\mathrm{eq},a}^n, \qquad C_b^n(x_b^n) = C_{\mathrm{eq},b}^n,$$

平衡濃度 $C_{\mathrm{eq},a}^n = C_0 \Gamma_a \kappa_a^n, \qquad C_{\mathrm{eq},b}^n = C_0 \Gamma_b \kappa_b^n,$

質量保存

$$\dot{x}_a^n = \Omega\{[D_b \partial_x C_b^n - D_a(\partial_y x_a^n)\partial_y C_b^n] - [D_a \partial_x C_a^n - D_b(\partial_y x_a^n)\partial_y C_a^n]\}_{x=x_a^n},$$

$$\dot{x}_b^n = \Omega\{[D_a \partial_x C_a^{n+1} - D_b(\partial_y x_b^n)\partial_y C_a^{n+1}] - [D_b \partial_x C_b^n - D_a(\partial_y x_b^n)\partial_y C_b^n]\}_{x=x_b^n}.$$

T. Frisch and A. Verga, PRL 96, 166104 (2006).

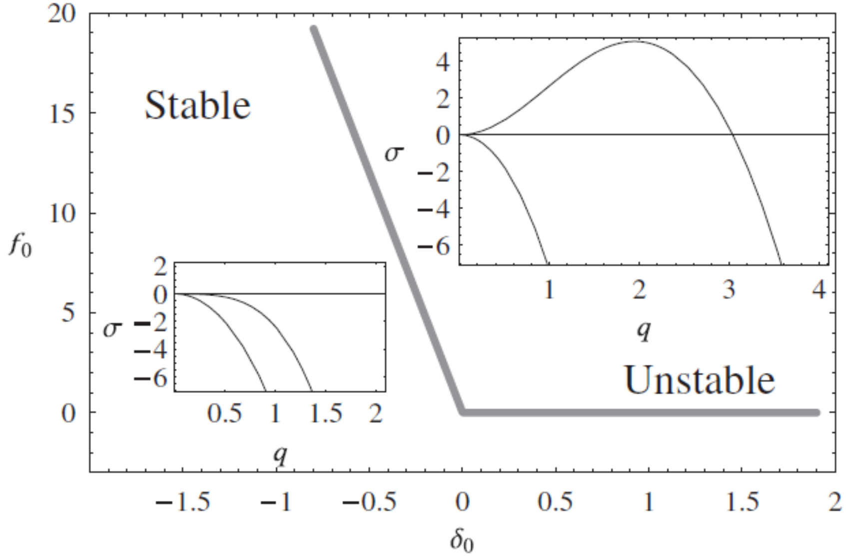
Si(001)表面でのステップ不安定化(二次元)

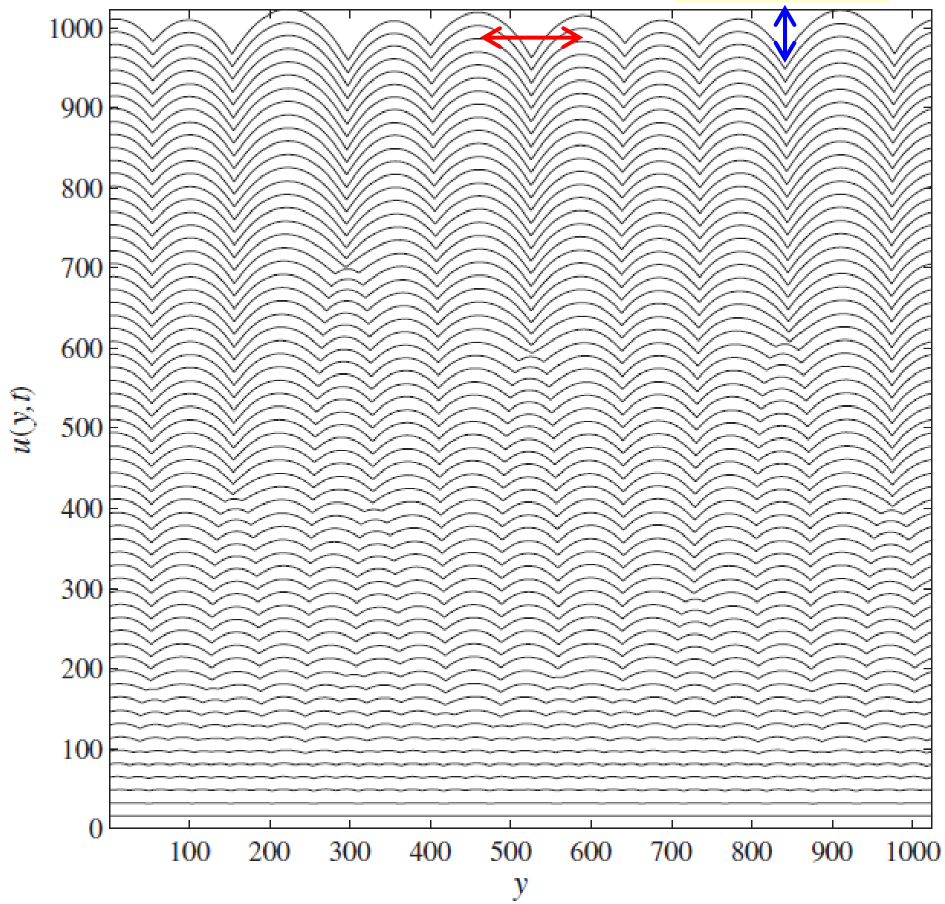
不安定化 $f_0 > f_{0c} = -12\alpha_0\delta_0/(\alpha_0 - 1),\ f_0 = \frac{Fl_0^3}{C_0\Gamma_a D_a},\quad \alpha_0 = \frac{D_b}{D_a},\quad \delta_0 = \frac{\Gamma_a - \Gamma_b}{\Gamma_a}$

Conserved version of the KS equation

$$\partial_t u = -\partial_y^2\left[\frac{f\alpha}{48}(f\alpha + 12\delta)u + \partial_y^2 u + \frac{f}{12}(\partial_y u)^2\right],$$

$w \sim t^{1/2}$ $\quad \left\langle u^2 \right\rangle^{1/2} \sim t$





T. Frisch and A. Verga, PRL 96, 166104 (2006).

応力下のステップバンチング

Step-Bunching Instability of Vicinal Surfaces under Stress
(transcription of Asaro-Tiller-Grinfeld instability)

Force per unit length on the *m*th step

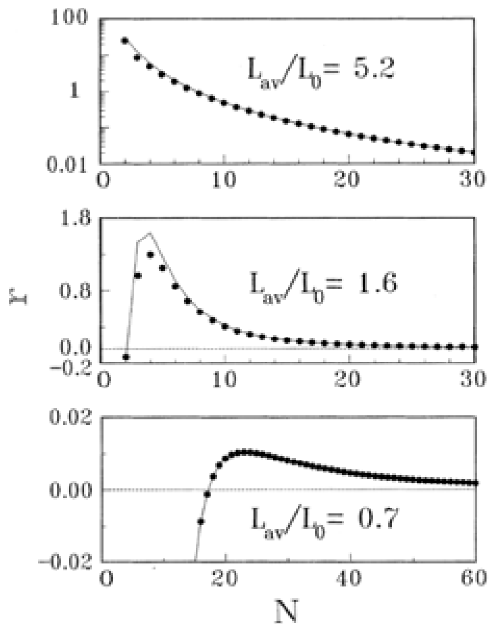
$$f_m = \sum_{n \neq m} \left[\frac{\alpha_1}{(z_n - z_m)} - \frac{\alpha_2}{(z_n - z_m)^3} \right]$$

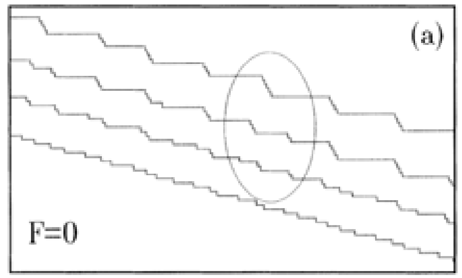
引力

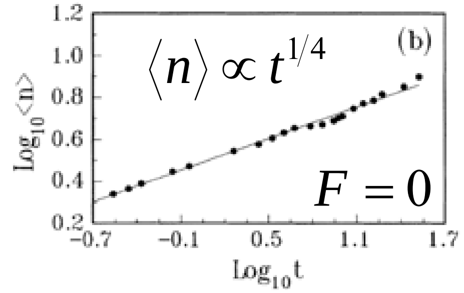
$$u_m(t) \approx e^{rt} \Delta \cos\left(\frac{2\pi}{N} m + \frac{2\pi}{N} F t \right),$$

where

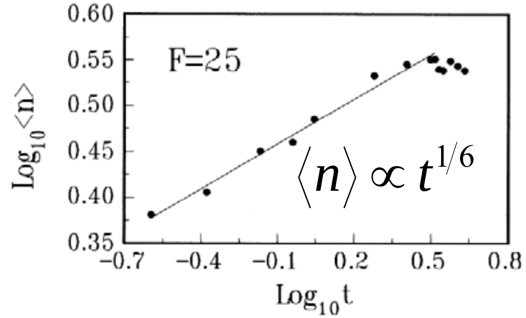
$$r = \left(\frac{B\alpha_1^2}{\alpha_2} \right) \frac{L_0^2}{L_{\mathrm{av}}^3} \frac{8\pi^4}{N^3} \left\{ 1 - \frac{1}{N} + \frac{L_0^2 \pi^2}{L_{\mathrm{av}}^2 N} \left(-1 + \frac{2}{N} - \frac{1}{N^2} \right) \right\}, \quad (8)$$







Minimum energy separation for an isolated pair $L_0 = \sqrt{(\alpha_2/\alpha_2)}$



r is sensitive on L_o/L_{av}.

For large L_{av}, *r* >o for all N.

J. Tersoff et al., PRL 75, 2730 (1995).

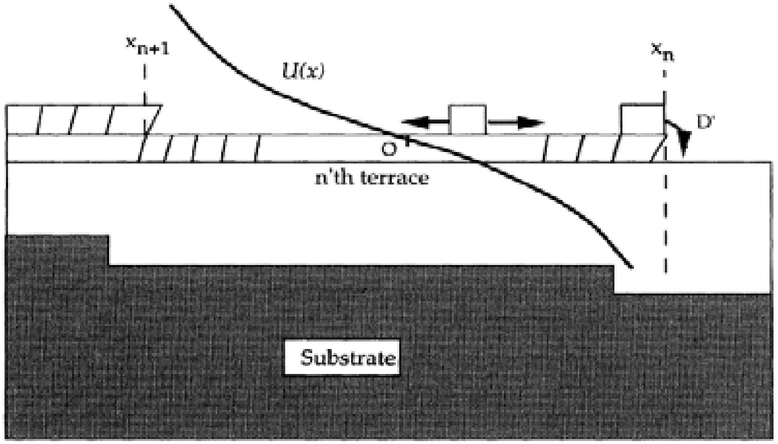
弾性的アドアトム-ステップ相互作用

Drift due to elastic interaction (similar to an effective ES effect)

$$D\partial_z\left[\partial_z c + \frac{c}{kT}\partial_z U\right] + F = 0$$

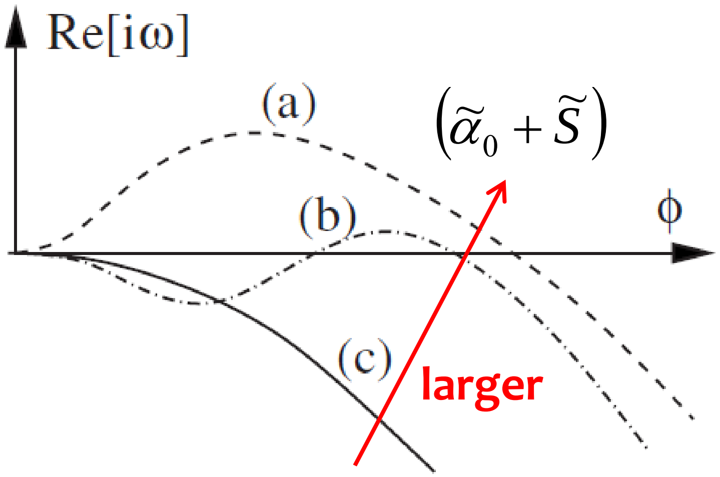
Elastic interaction between adatom and step

$$U = -\sum_{m=-\infty}^{\infty}\frac{\alpha_0}{z - z_m}$$



Adatom is attracted to the upper or lower terrace because of elasticity.

$$\mathrm{Re}[i\omega] \sim F\left(\tilde{\alpha}_0 + \tilde{S}\right)\phi^2 + \tilde{\alpha}_1\varepsilon|\phi|^3 - \tilde{\alpha}_2\phi^4$$



C. Duport et al., PRL 74, 134 (1995).

まとめ

　結晶表面では、成長や昇華の際中に、ステップの運動に伴い、様々な不安定化現象が現れる。

　ステップの蛇行とバンチングを中心に、不安定化現象のメカニズムの解明に向けた実験的・理論的研究を概説した。