Quantum Dots: formation

ECE 598XL

Fall 2009

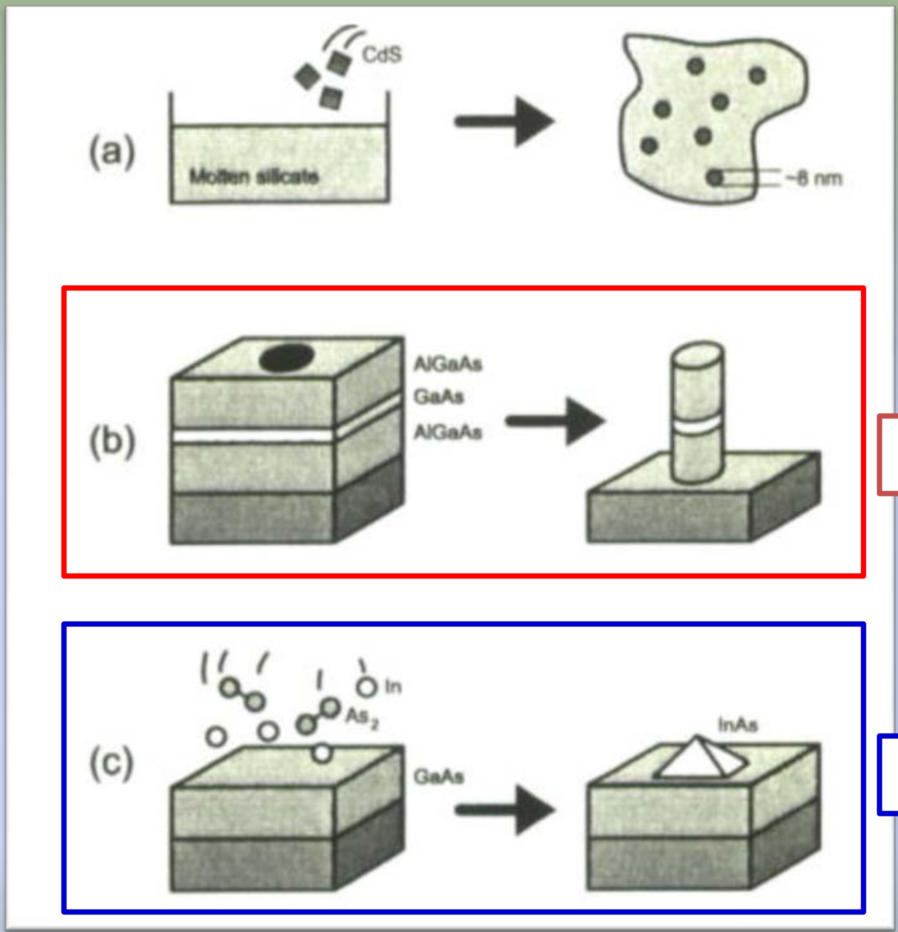
Outline

- Introduction
- Self-organized QDs
  - Critical thickness
  - S-K growth mode
- Patterned QDs
  - Lateral positioning
  - Multistack ordering
- QDs in QWs
  - Case study: InGaN QDs in InGaN QW
- Anti dots

QD Based Device Applications

- QD infrared photodetectors (interband and intraband optical transitions)
- QD lasers and LEDs
  - Edge emitters
  - VCSELs
  - Photonic crystals
- QD solar cells
- Single QD as building blocks for
  - Quantum information processing
  - single electron devices
- Dislocation filter for metamorphic growth

Methods of Nanostructure Formation



Top Down

Bottom Up

D. Bimberg, M. Grundmann, N. Ledenstov, "*Quantum Dot Heterostructures*"

Spontaneously Formed Nanostructures

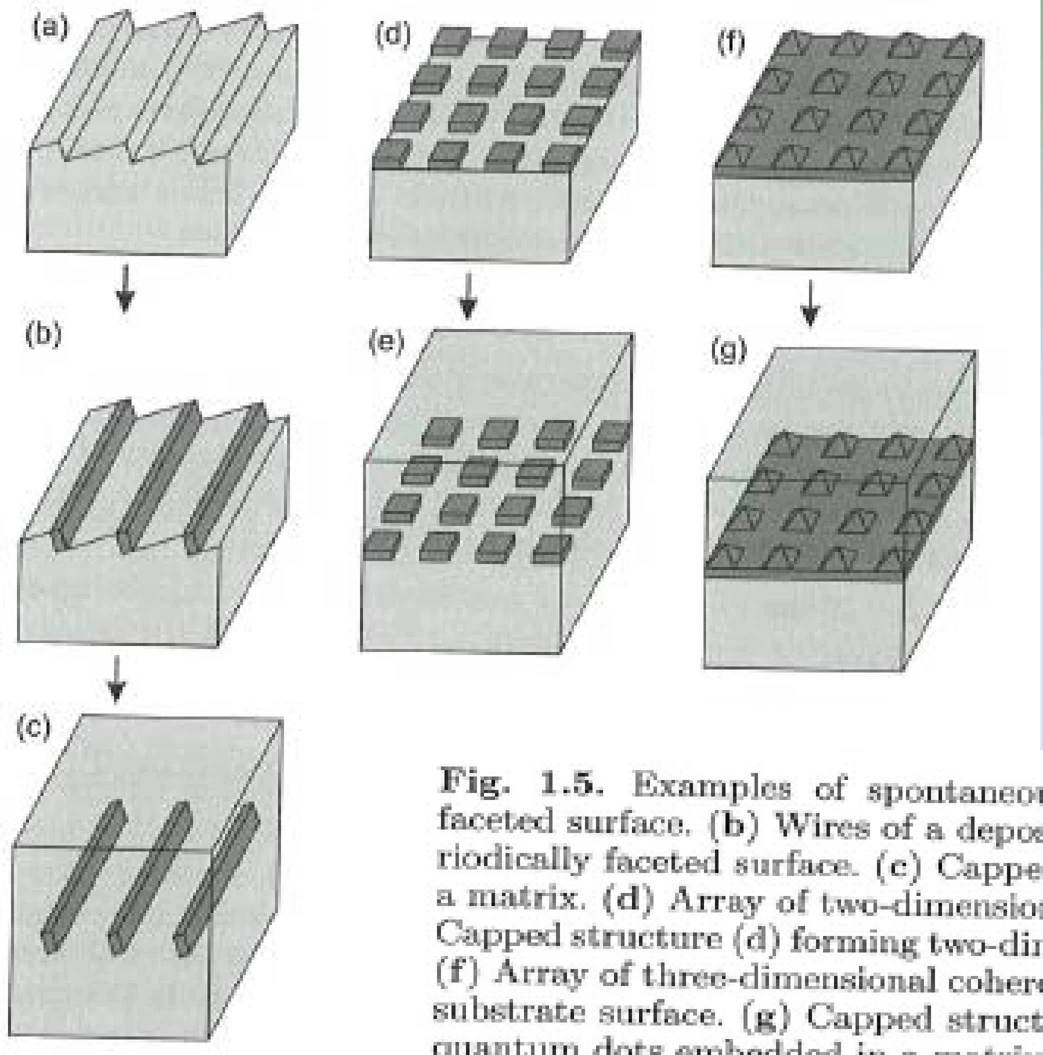


Fig. 1.5. Examples of spontaneously formed nanostructures. **(a)** Periodically faceted surface. **(b)** Wires of a deposited material formed in the grooves of the periodically faceted surface. **(c)** Capped structure (b) forming quantum wires within a matrix. **(d)** Array of two-dimensional islands in submonolayer heteroepitaxy. **(e)** Capped structure (d) forming two-dimensional quantum dots embedded in a matrix. **(f)** Array of three-dimensional coherently strained islands over a wetting layer on a substrate surface. **(g)** Capped structure (f) forming an array of three-dimensional quantum dots embedded in a matrix

QD Formation Methods

- **Self-organized QDs**
  - Stranski-krastanov growth:
    - Lattice mismatch
    - Strain
  - Growth methods:
    - MOCVD
    - MBE
- **Patterned QDs**
  - Lithography
  - Etching
  - Growth
- **Colloidal QDs**
  - Reaction T, t
  - Core-shell
  - Biological tag

Critical Layer Thickness

- **Pseudomorphic**: for thin layer where the overlayer is coherently strained (overlay adopt lattice constant of the substrate).
- **Dislocation formation**: for thick layers where the overlayer is relaxed (adopt its original lattice constant) through dislocation formation.
- **Critical layer thickness**: transition between the above two regions
  - Function of mismatch strain, etc.
  - Matthews-Blakeslee model (1970s)
  - Ge on Si (4%): 3- 4 ML
  - InAs on GaAs (7.2%): 1.7 ML
  - InAs on porous GaAs: 4.2 ML

$$E_{strain} = \lambda \left(\frac{\Delta a}{a} \right)^2 At$$

λ = elastic modulus

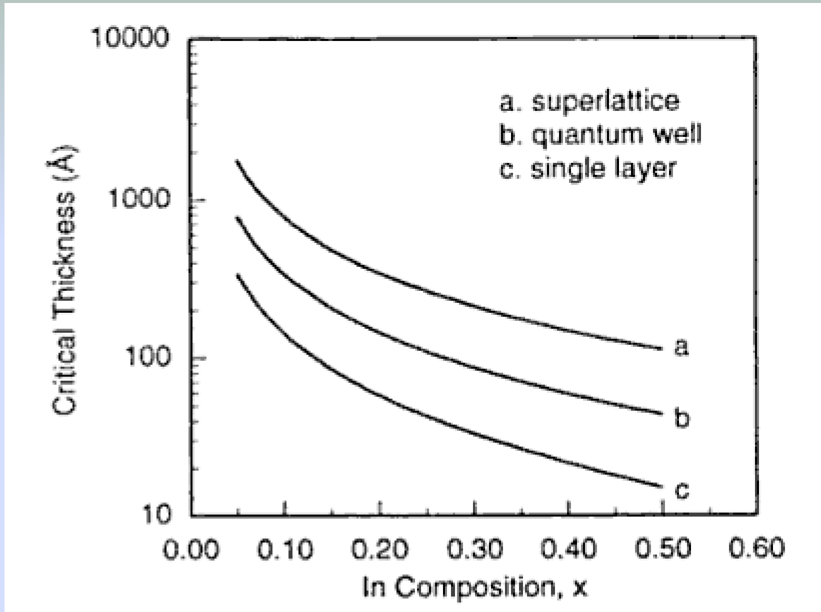
Δa = forced change in lattice constant

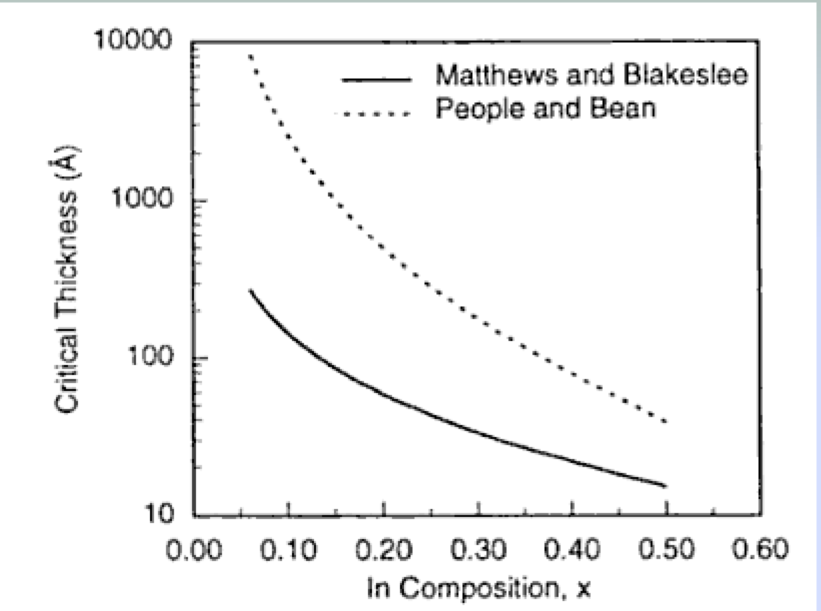
a = "free" lattice constant

A = area

t = thickness

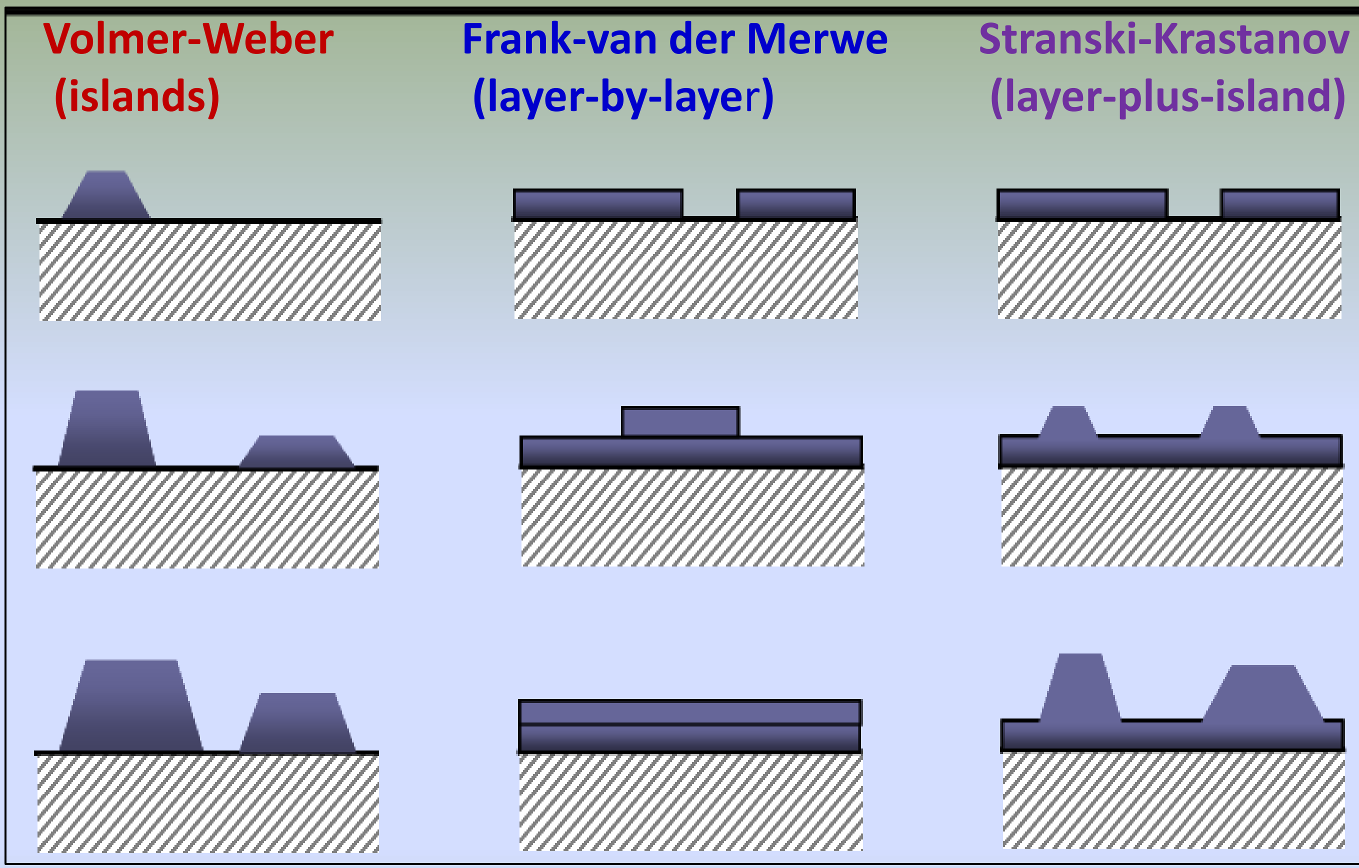
Critical Layer Thickness



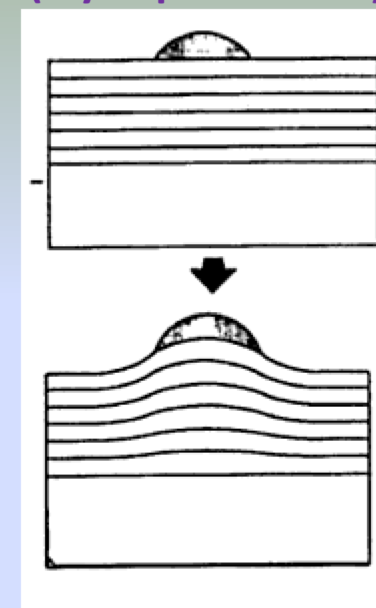


JJ Coleman, Strained Layer QW lasers

Three Modes of Epitaxial Growth



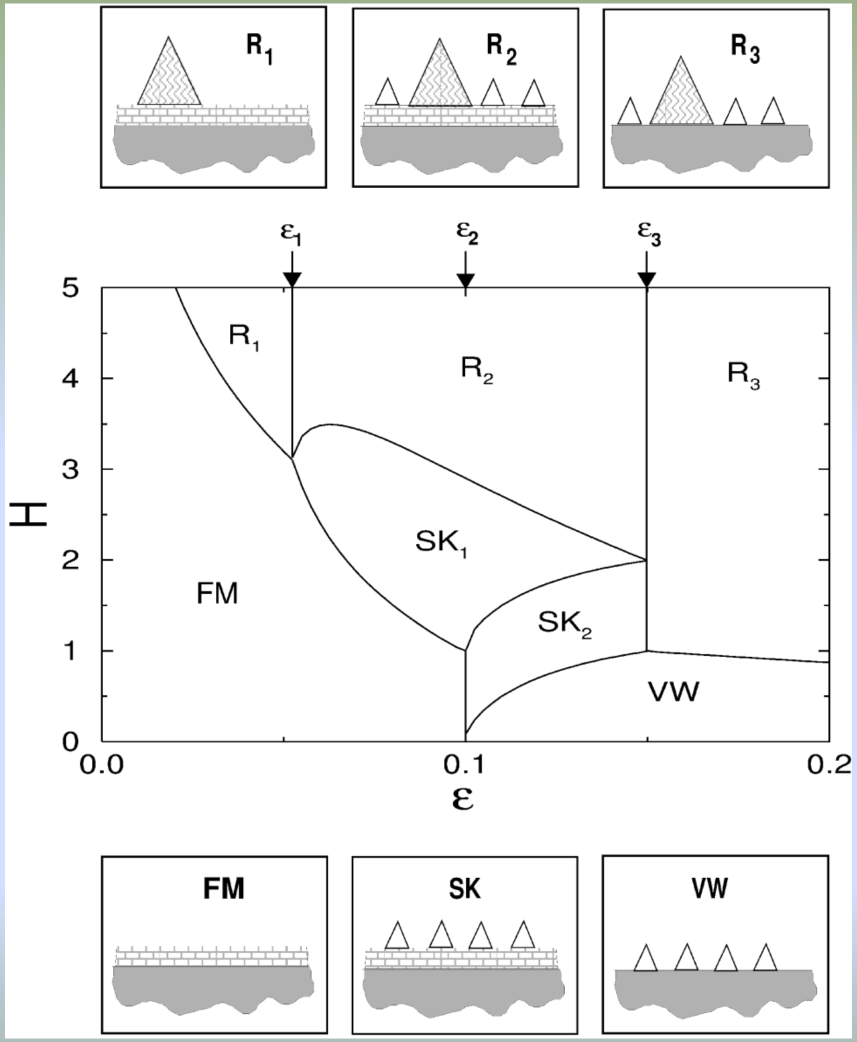
Stranski-Krastanov (layer-plus-island)



strain sharing

wikipedia

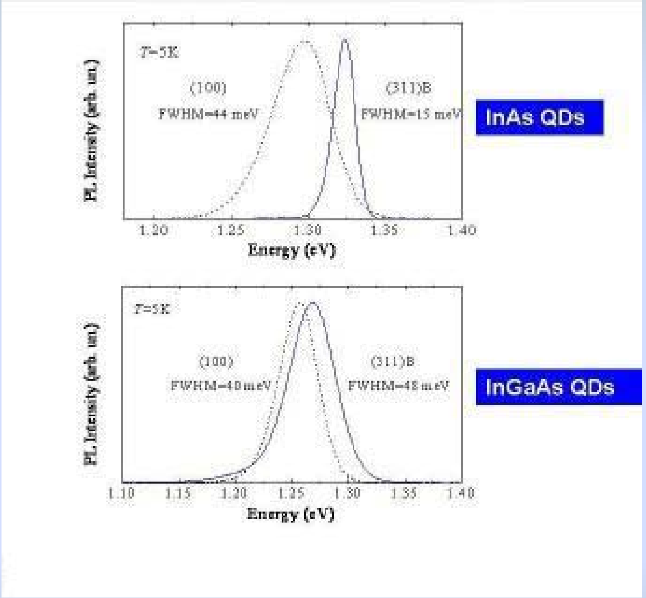
Phase Diagram
coverage H (ML) vs misfit strain ε



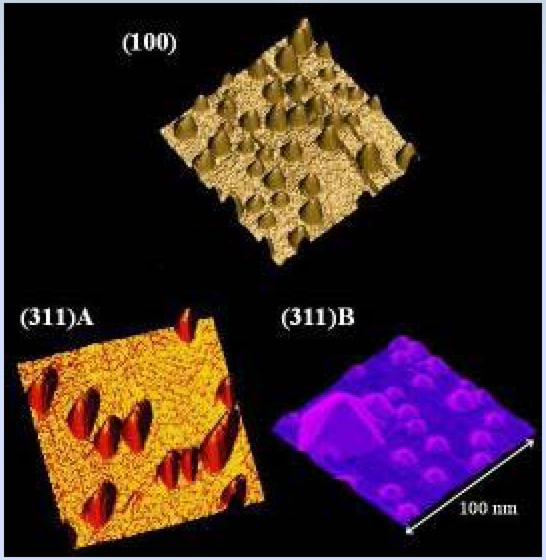
PRL, 79, 3708 (1997)

QDs – shape, size, size distribution

- Substrate orientation dependence



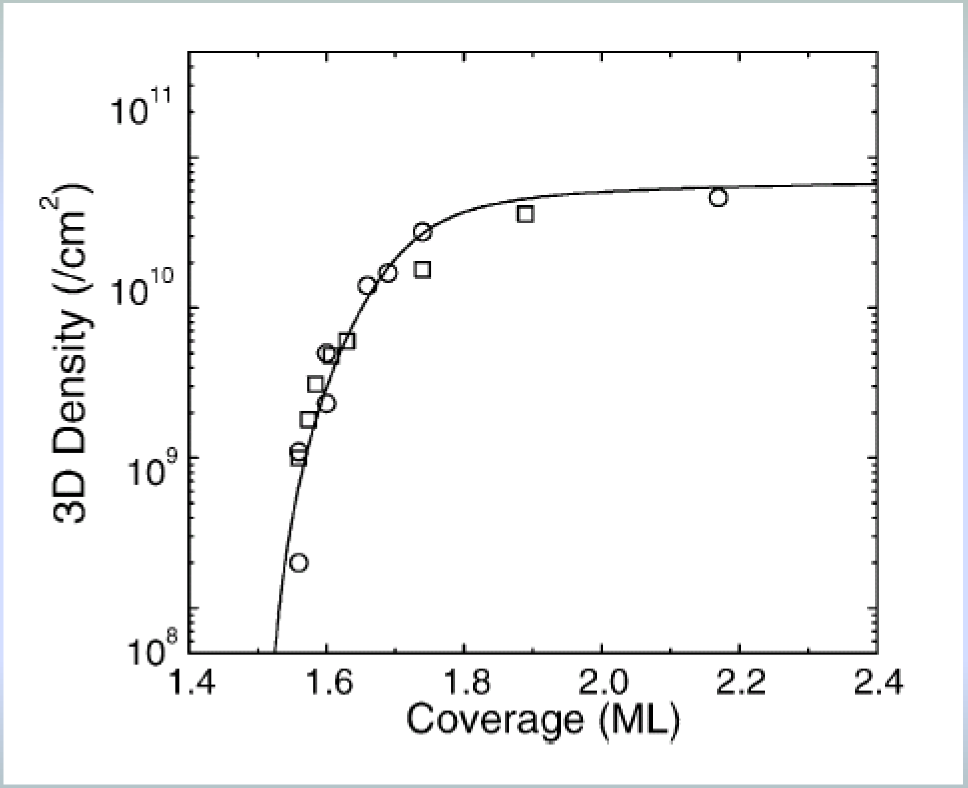
STM of InAs/GaAs QDs grown by MBE on (100), (311)A and (311)B GaAs substrates.



Nottingham

Coverage vs QD Density

- Density of 3D islands vs InAs coverage



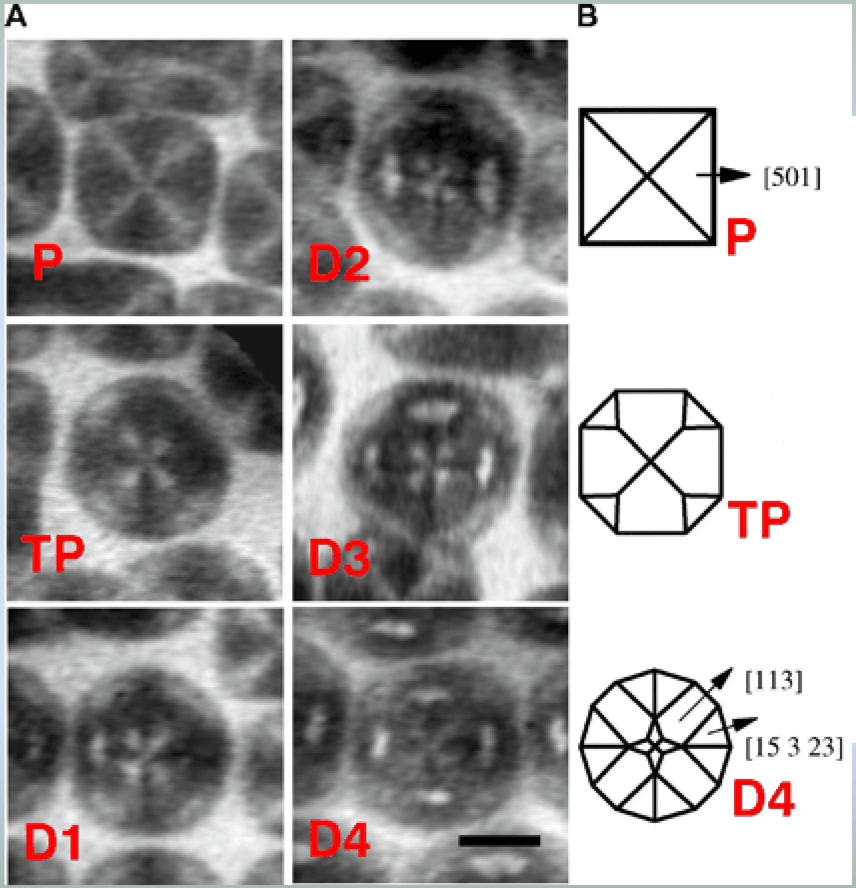
B.A. Joyce, D.D. Vvedensky / Materials Science and Engineering R 46 (2004) 127–176

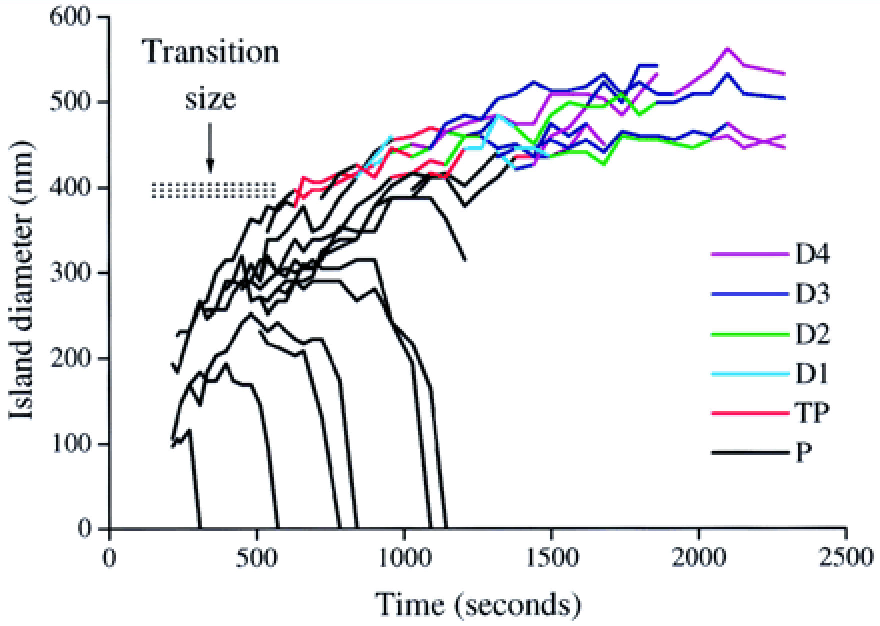
QDs – shape, size, size distribution

- Ge QDs/Si(100):
  - Competition between surface energy and strain relaxation in the island bulk
    - Pyramids (high aspect ratio, square base with {105} facets): more surface energy but relaxes more strain
    - Domes (low aspect ratio, multi-faceted):
    - Change from pyramids to domes w size
- InAs/GaAs
  - Pyramid: bounded by four steep {137} facets

QDs – shape, size, size distribution

- Growth conditions, size and composition dependence





Ross et al. Science, 286, 1931 (1999)

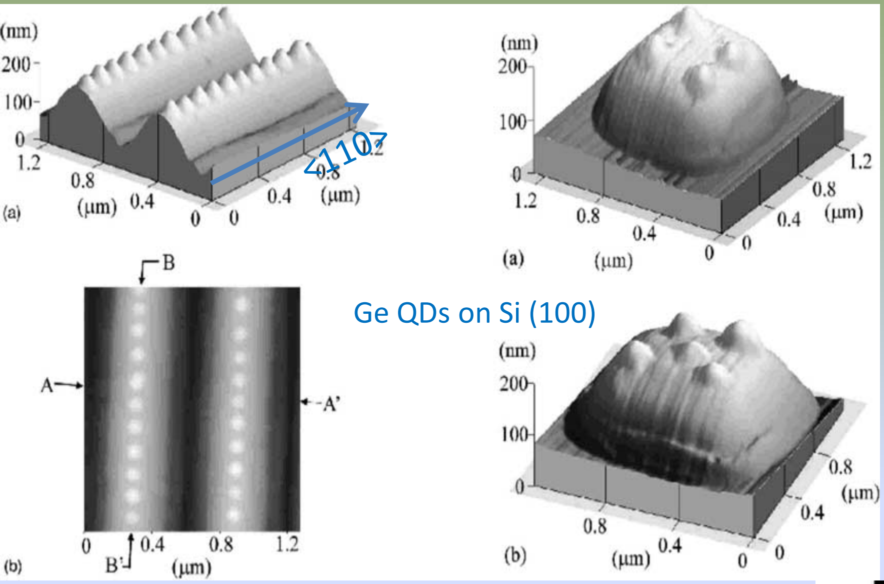
Criteria for QD Growth

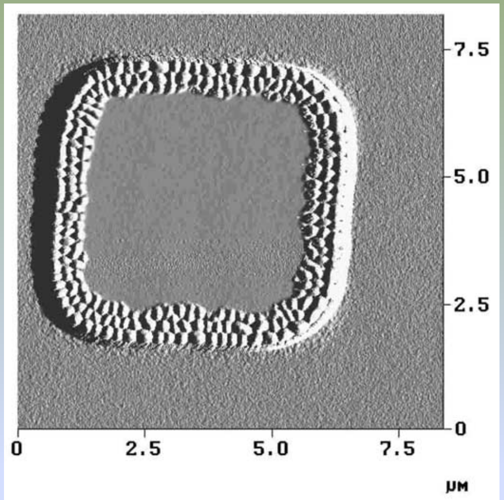
- Size controllability: size and uniformity
- Site controllability: positioning and density
- Vertical stacking and ordering

Lateral Positioning: patterned QDs

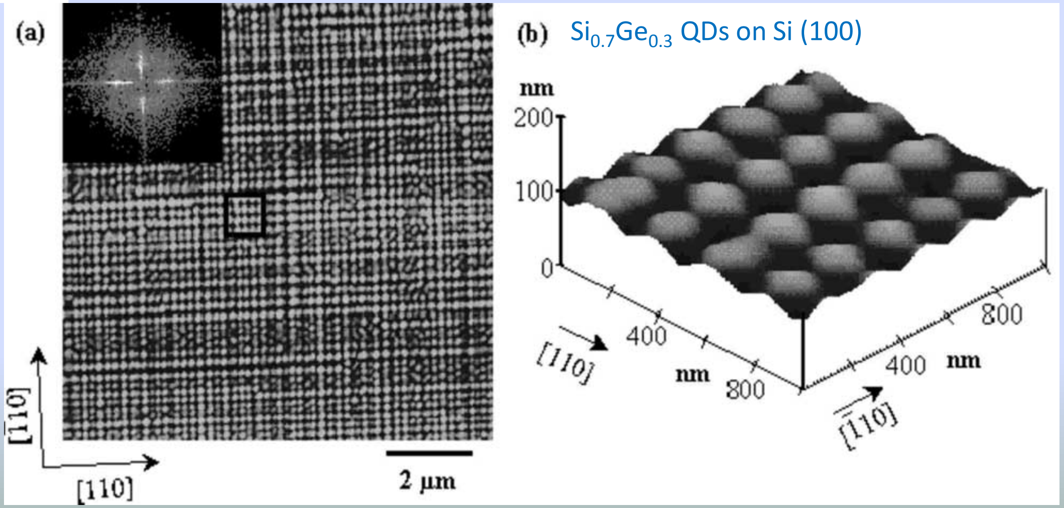
- Site control using preferential nucleation sites on a substrate
  - ex situ Lithography defined surface morphology
    - Maskless – self strain management
      - Raised stripes and squares
      - Pits/holes (ebeam, nanoporous Al_2O_3 template, diblock copolymer nanopatterning, etc.)
    - Masked (growth inhibition) selective area growth
      - Oxide
      - metal
  - Surface steps and bunches
  - Intentional formation of dislocations

Lateral Positioning: patterned QDs



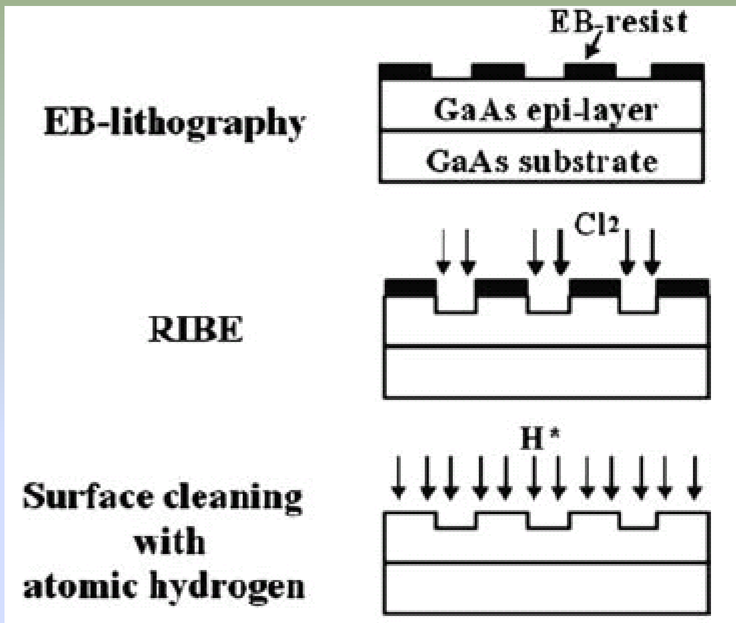


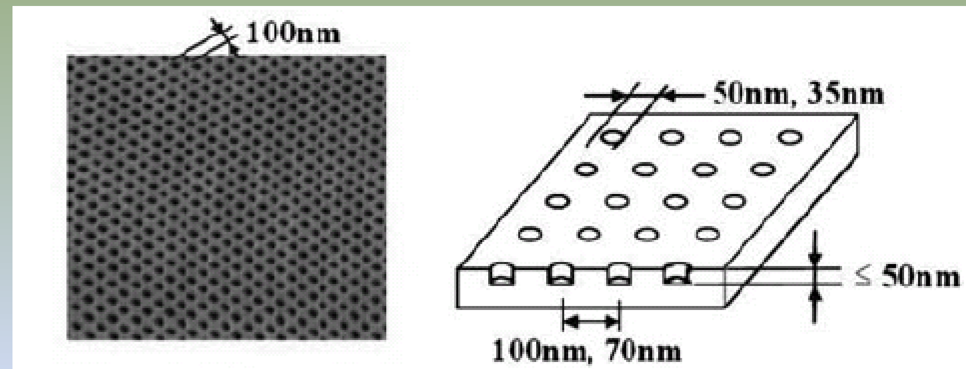
Strain Management

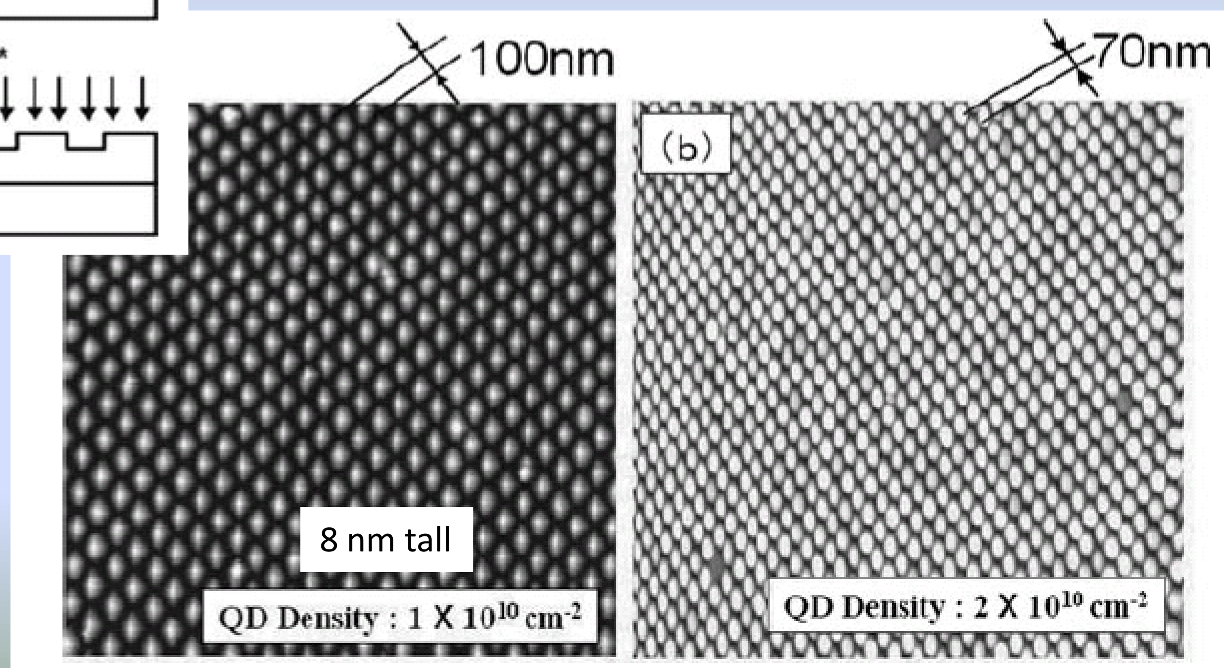


Woll, Rugheimer, Lagally, 2002

Patterned substrate with nano hole arrays







Localized modification of surface strain after in-filling

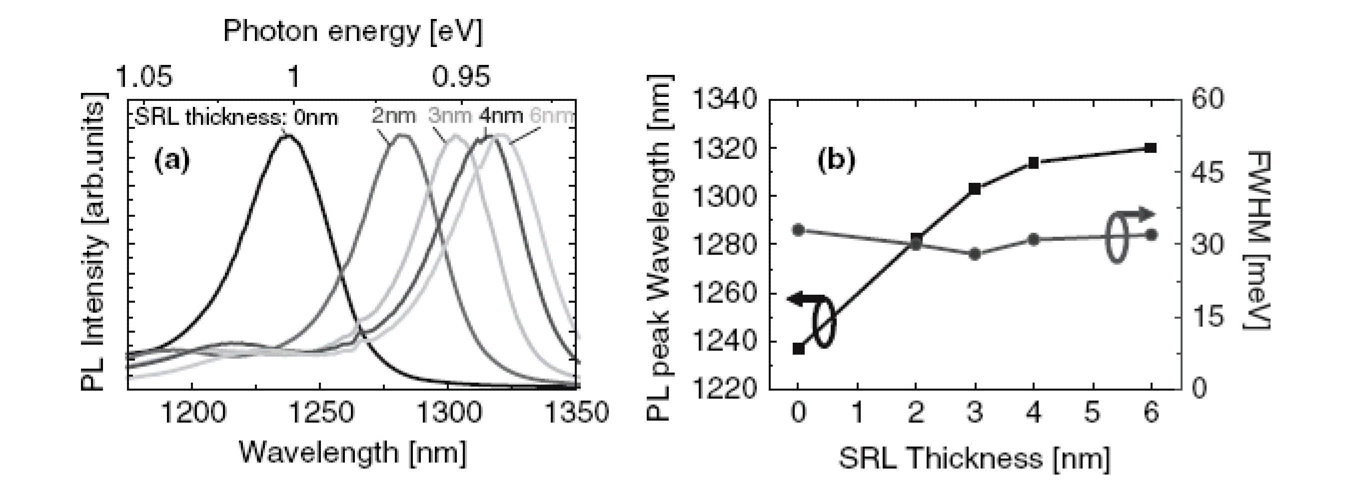
InGaAs QD nucleation is achieved by reducing T from 400 to 300 °C

100% occupancy!

Nobuhiko Ozaki, Shunsuke Ohkouchi, Yoshimasa Sugimoto, Naoki Ikeda, and Kiyoshi Asakawa

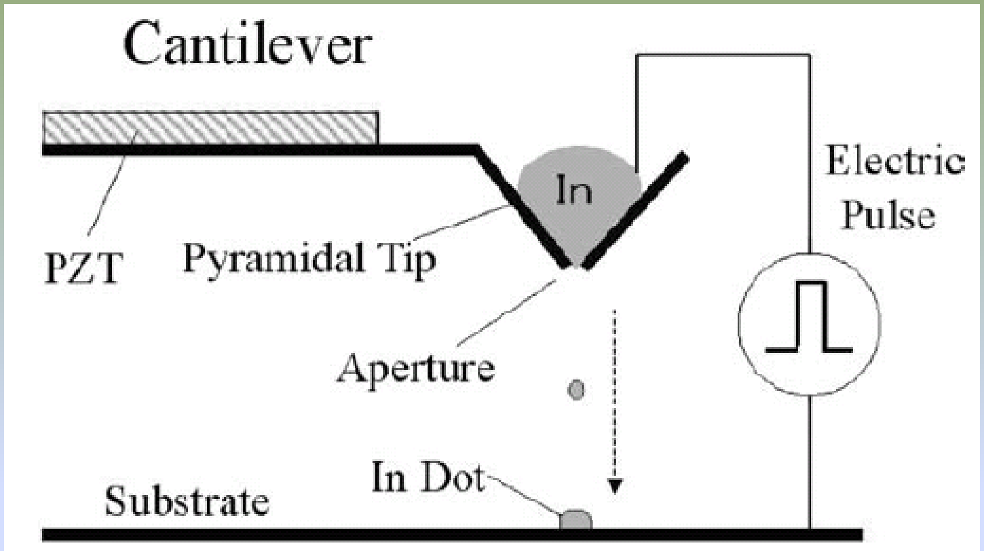
Site-controlled InAs QDs

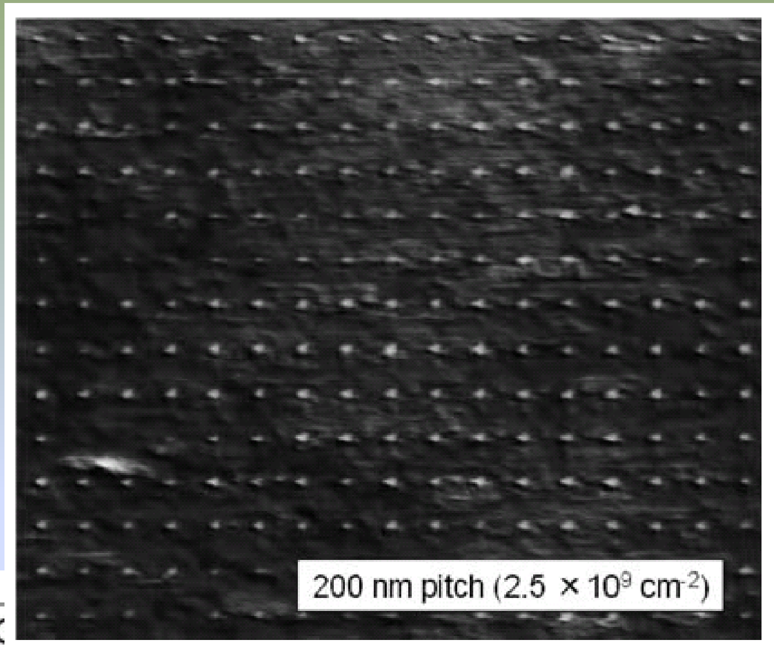
- **PL wavelength tuning using strain reducing layer $In_{0.2}Ga_{0.8}As$ (0 – 6 nm) on the InAs QDs**

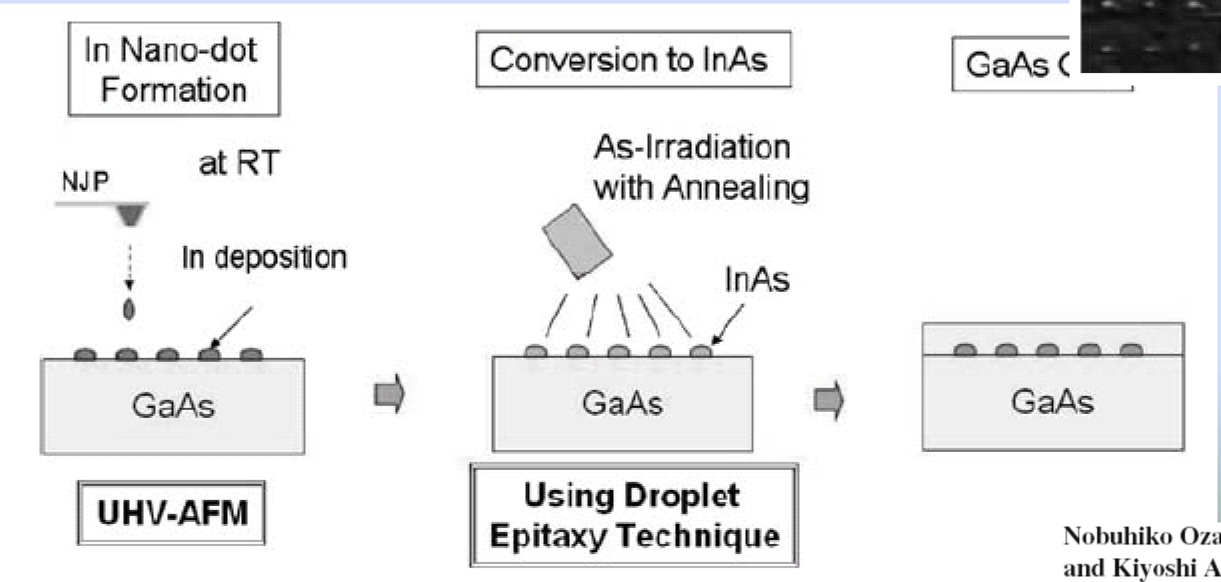


Nobuhiko Ozaki, Shunsuke Ohkouchi, Yoshimasa Sugimoto, Naoki Ikeda, and Kiyoshi Asakawa

InAs QDs by Nano Jet Probe Patterning







Nobuhiko Ozaki, Shunsuke Ohkouchi, Yoshimasa Sugimoto, Naoki Ikeda, and Kiyoshi Asakawa

Linear alignment of GaN QDs on vicinal AlN/SiC substrate by controlled nucleation on steps

- Vicinal SiC substrate preparation
- Stepped AlN layer
  - Height and width of AlN step – growth condition dependent

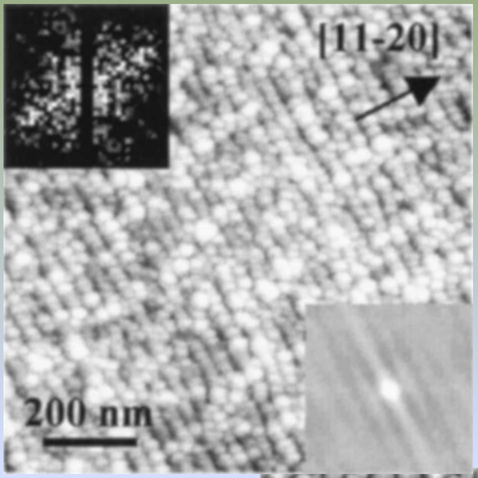


FIG. 4. AFM image of GaN quantum dots grown on a vicinal AlN~0001! surface. The upper inset gives the details of the 2D-FFT spectrum and the lower inset gives details of the center of the autocorrelation picture.

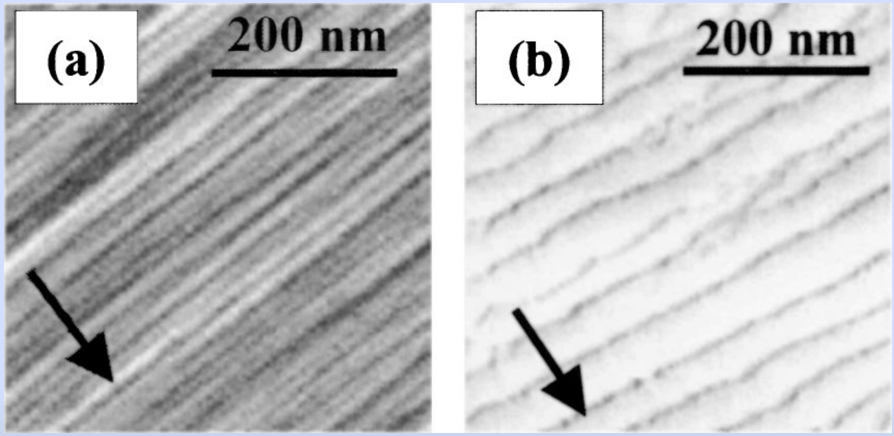


FIG. 1. AFM images of ~a! a 4H–SiC~0001! vicinal surface after HCl/H2 etching and ~b! a ;50 nm AlN layer grown on a 4H–SiC~0001! vicinal surface. The arrow on the images indicates the [11-20] direction.

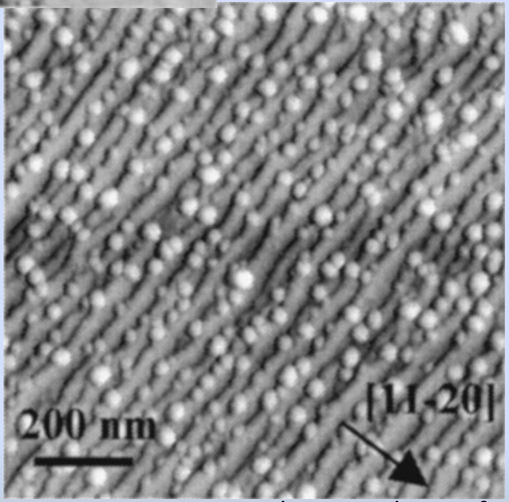
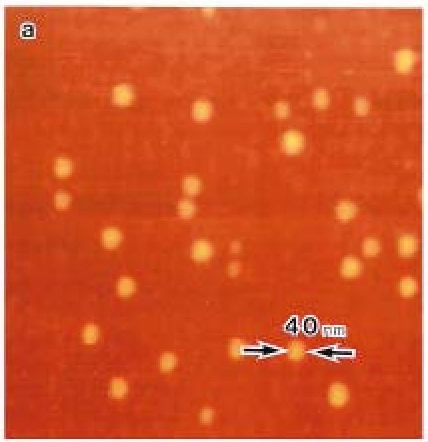


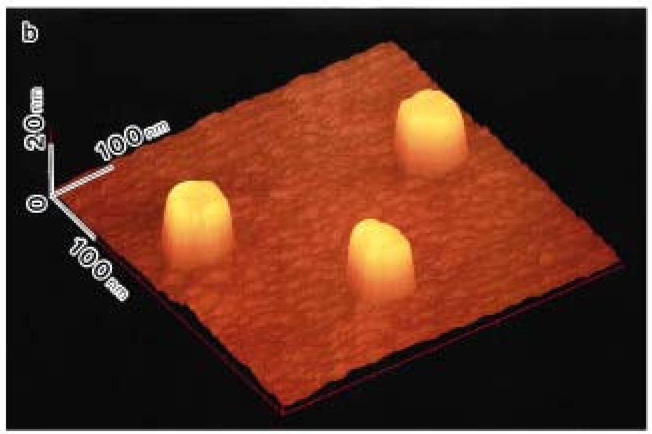
FIG. 2. AFM image showing the preferential alignment of GaN QDs along the AlN bunched steps.

JAP 93, 3108, 2003

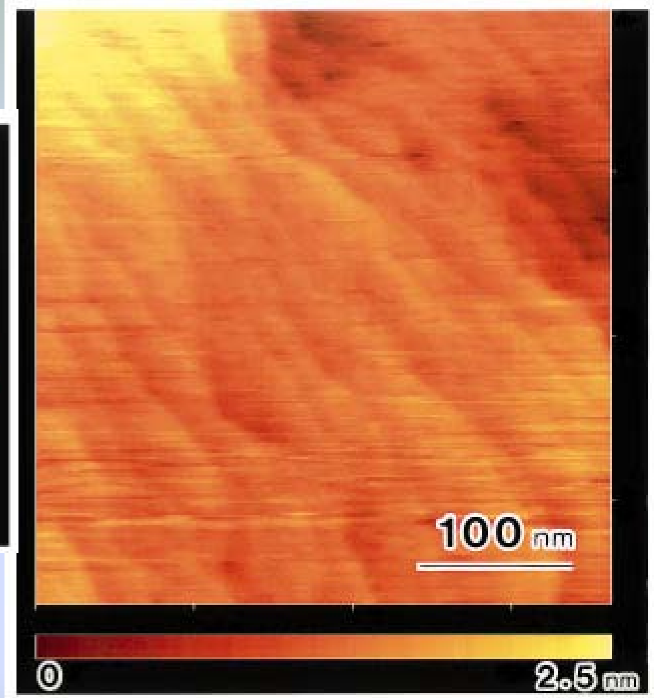
QDs growth using anti-surfactant

- Anti-surfactant: to promote 3D growth
  - Tetraethylsilane Si(C2H5)4 (TESi)
  - Modifies surface energy





w TESi, QDs 3e9 density

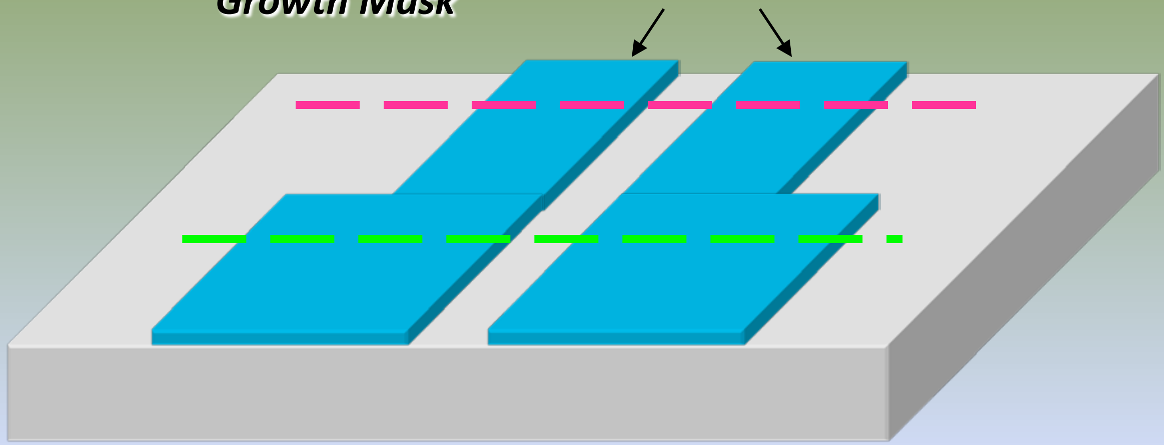


w/o TESi. No QDs

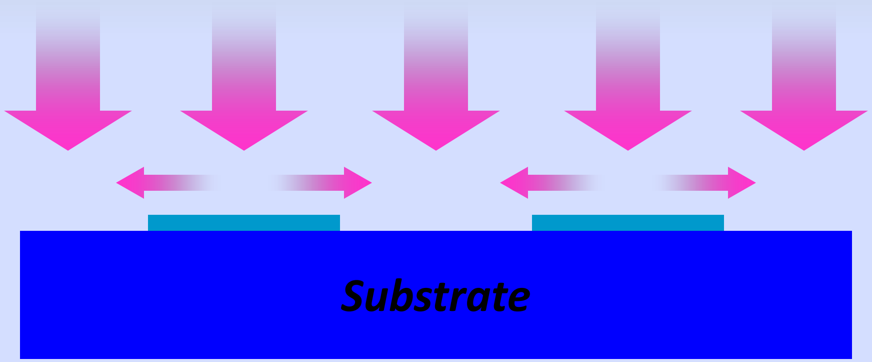
Appl. Phys. Lett. **69 (26), 23 December 1996**

Selective Area Epitaxy Process Overview

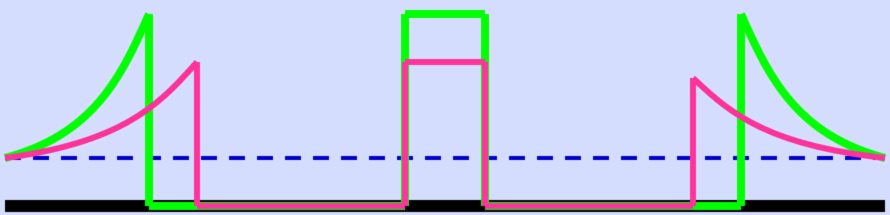
Dual Stripe Pattern
Growth Mask



Source Material

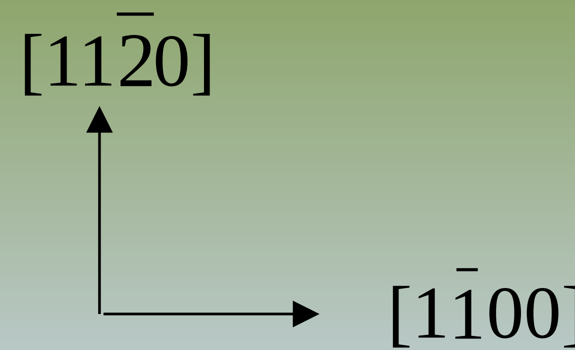


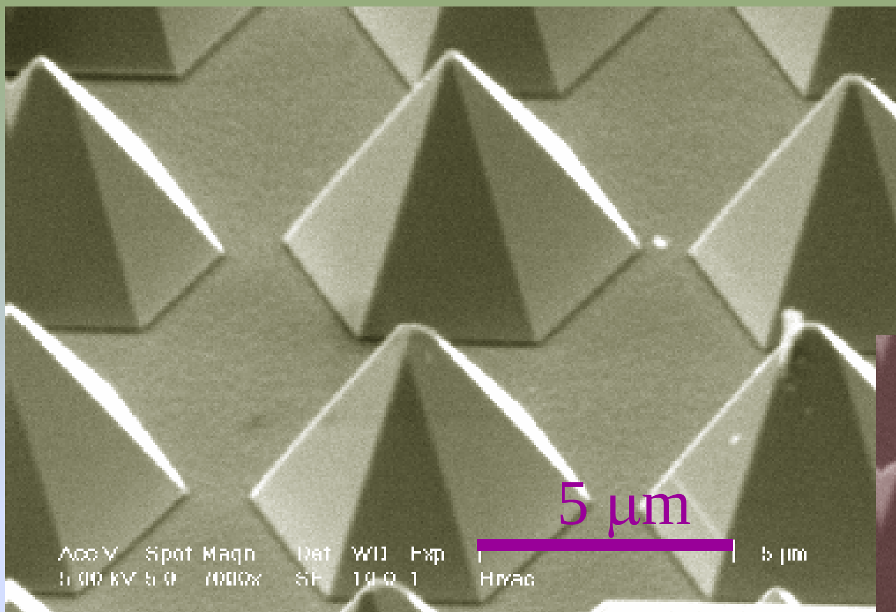
Growth occurs at exposed crystal surface only



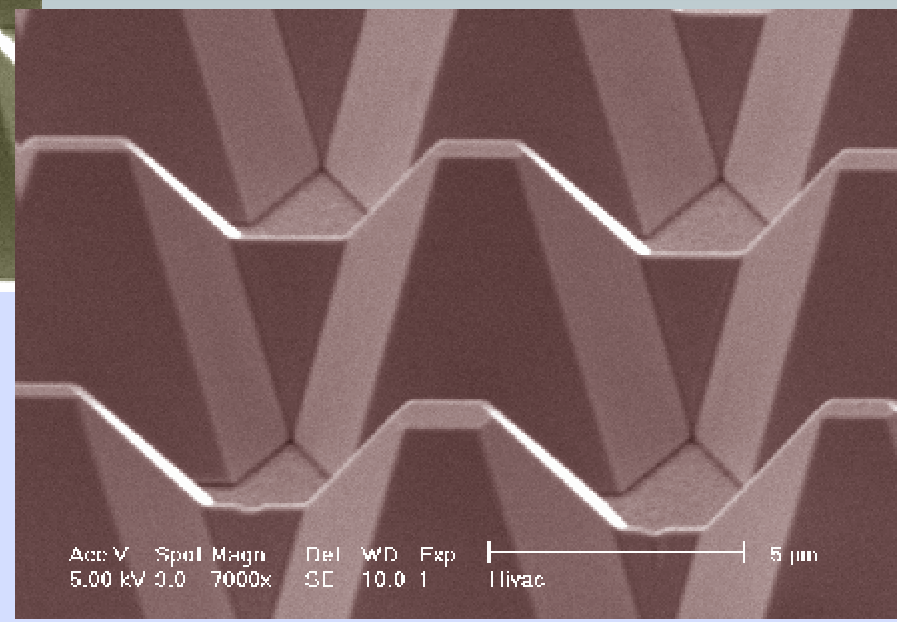
Epitaxial Growth Rate Enhancement

Selective area growth and lateral overgrowth





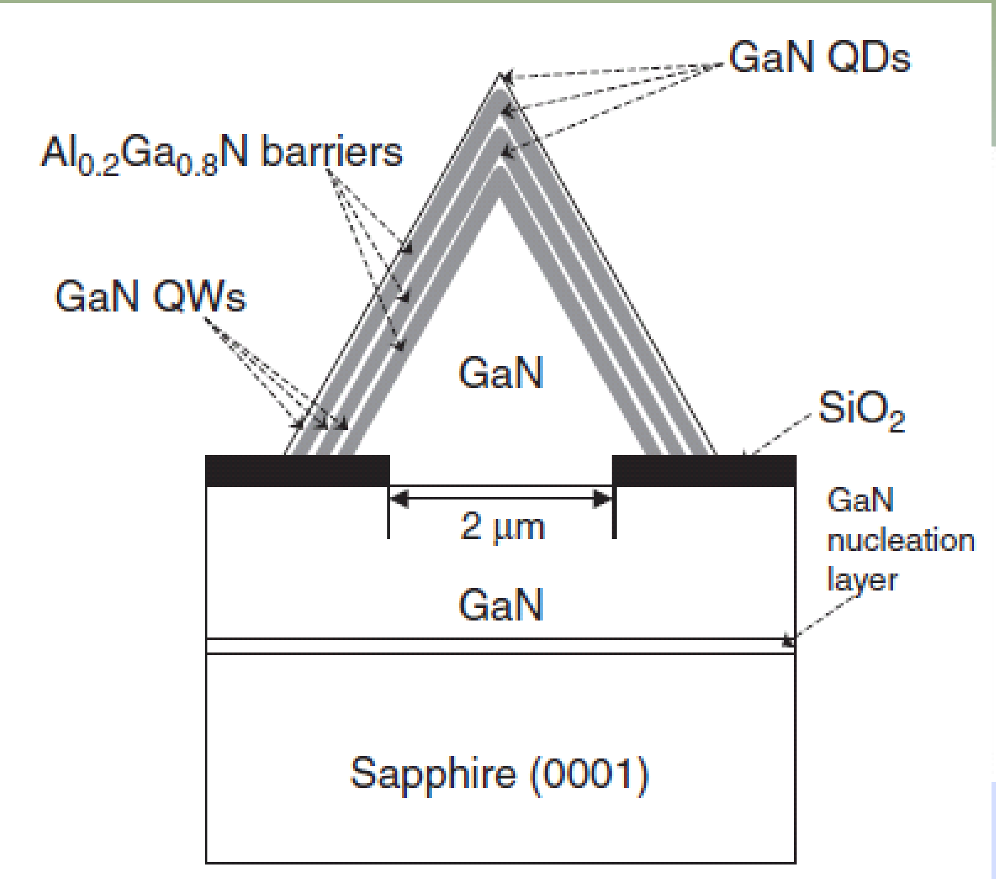
Isolated GaN pyramids

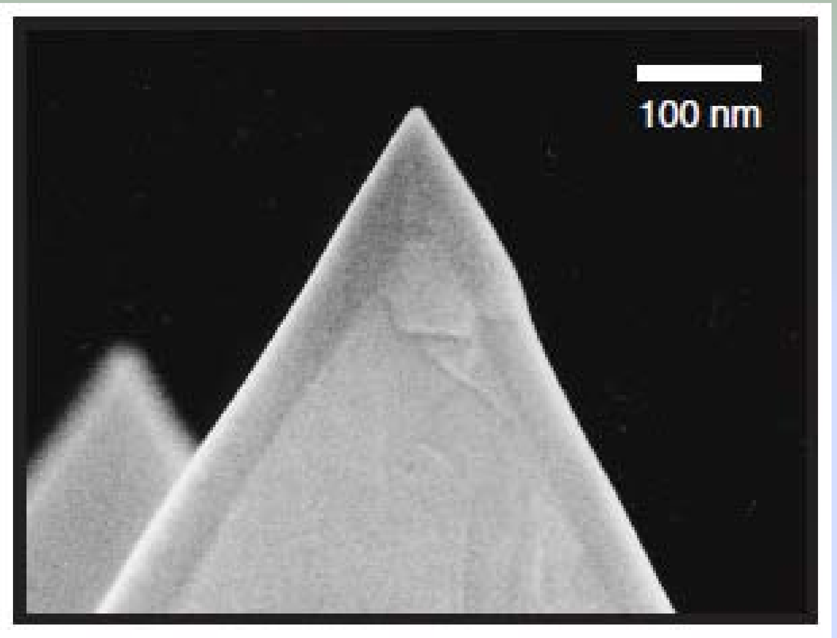


Partially coalesced

Li et al. APL, 73, 1179 (1998).

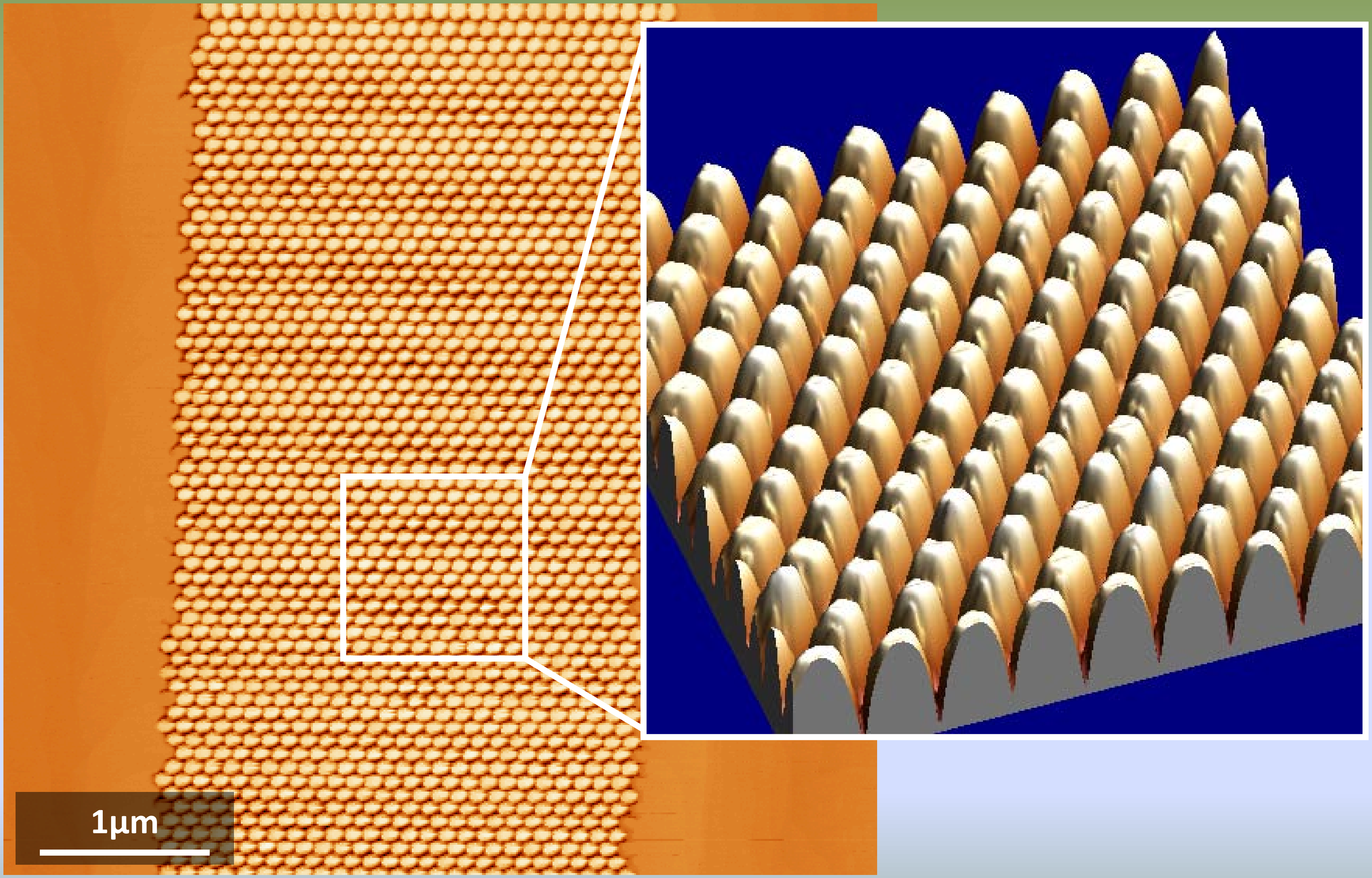
QDs at the Apex



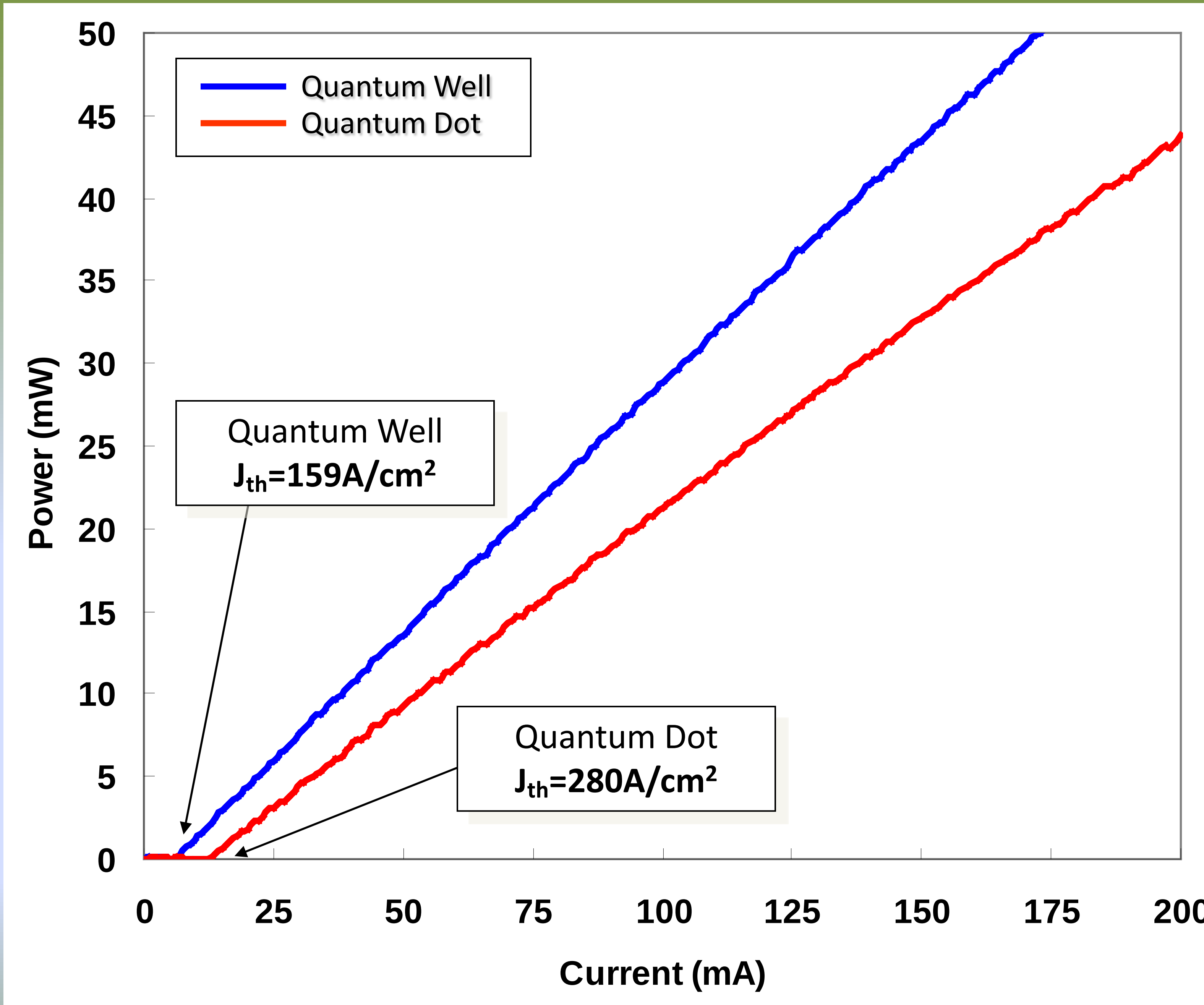


K. Tachibana et al. / Journal of Crystal Growth 237–239 (2002) 1312–1315

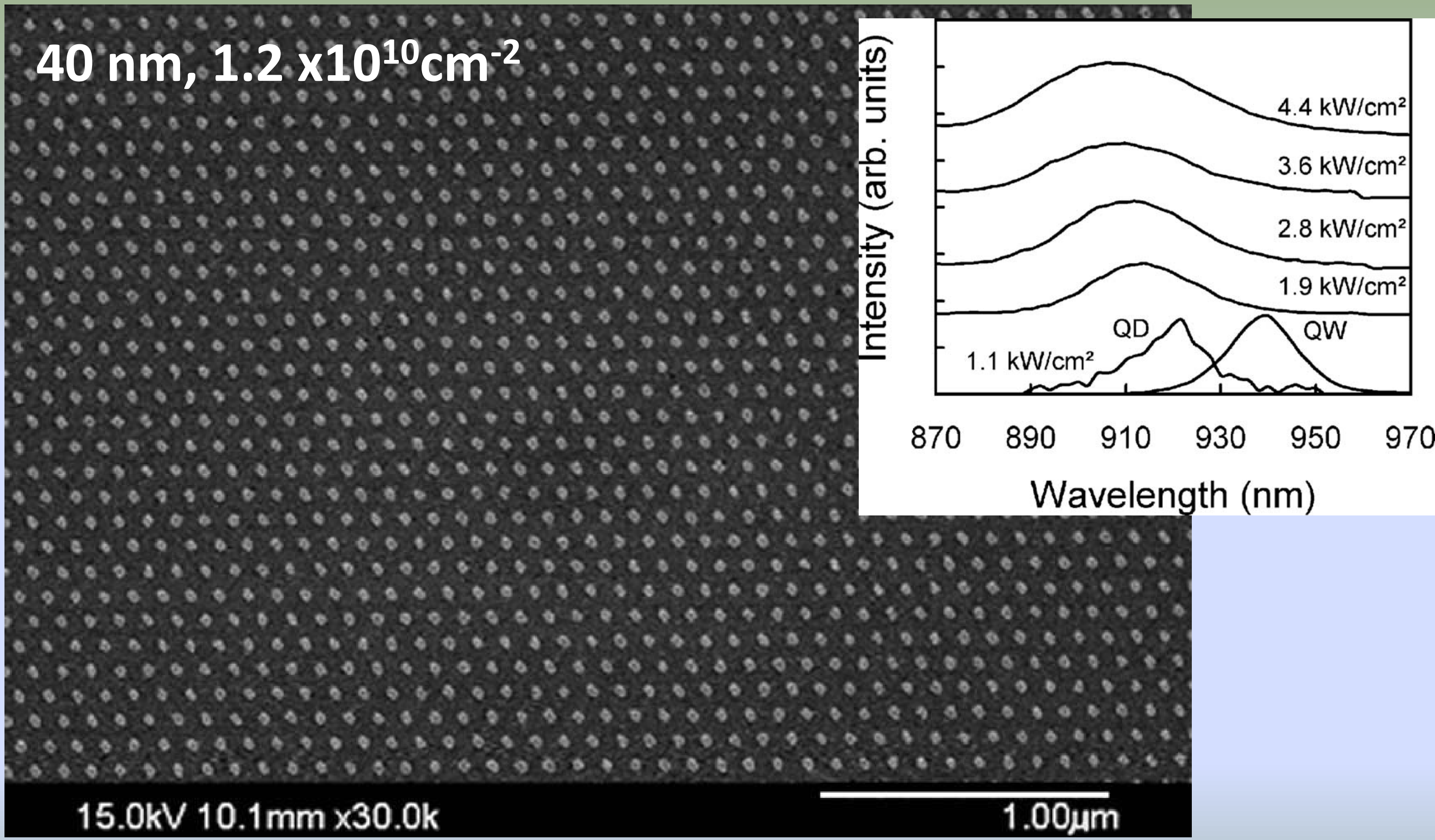
Oxide Patterned InGaAs QDs



Elarde thesis (Coleman)


Quantum Well
Quantum Dot
Quantum Well
J_{th}=159A/cm²
Quantum Dot
J_{th}=280A/cm²
Power (mW)
Current (mA)
0
5
10
15
20
25
30
35
40
45
50
0
25
50
75
100
125
150
175
200

InGaAs QDs by top-down e-beam patterning and wet etching using HSQ as mask

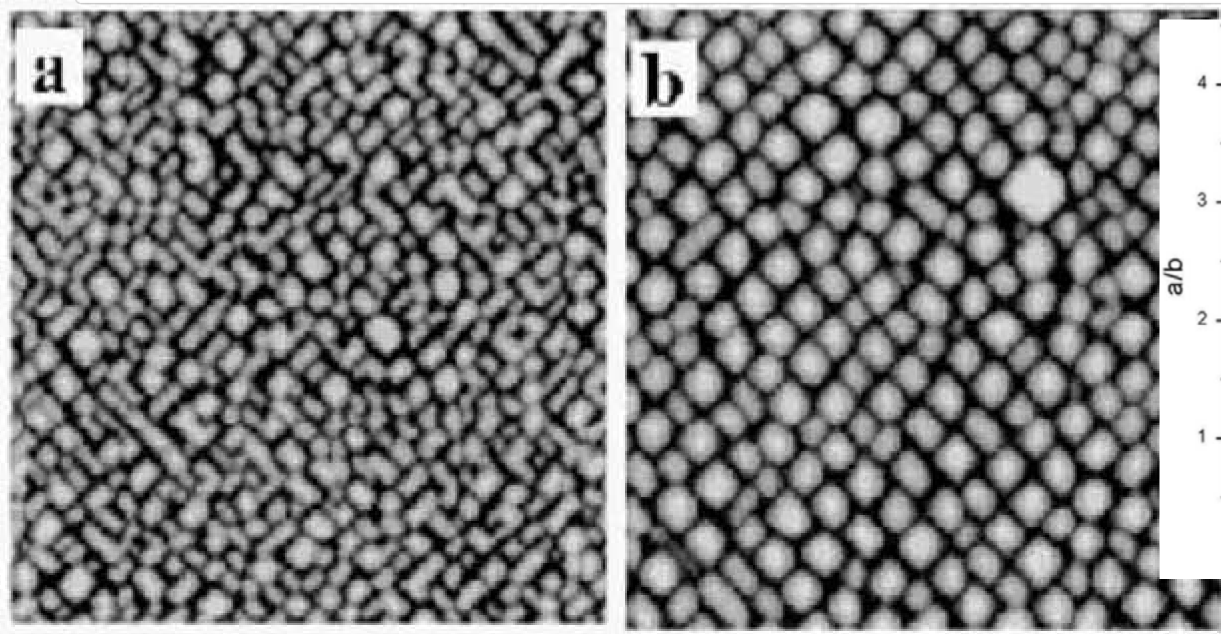


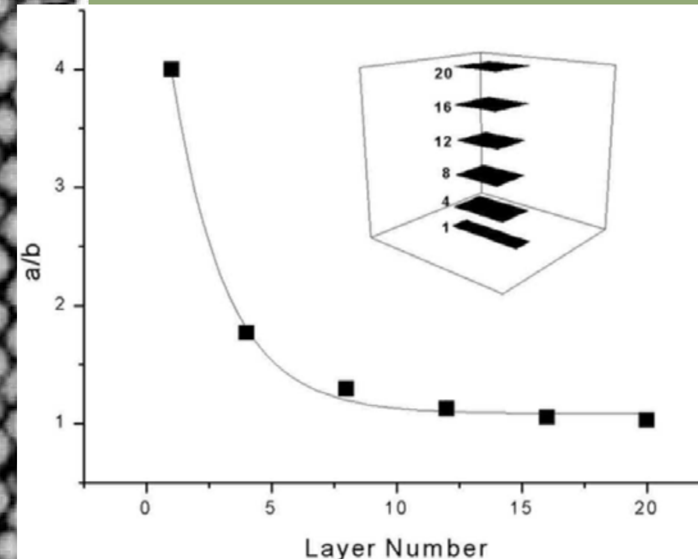
Verma, Coleman, APL, 93, 111117 (2008).

Multilayer Ordering: vertical positioning

- 3D arrays of QDs
- Self-assembly through strain management
- Alternating: GaAs/QD/GaAs/QD/GaAs
- Spacer layer
- Lateral strain inhomogeneity due to buried islands is transmitted to the surface through strain, so that nucleation of 3D islands in the 2^{nd} and subsequent layers is no longer spatially random.

Vertical Correlation: SiGe QDs on Si





- Spacer layer thickness dependence
- Islands become larger w layer #
- Islands become more regular in shape
- Increase in size $\propto$ 1/density
- Wetting layer thickness decrease w layer #
- Solution: reduce Ge dose w layer #

InGaAs QDs on GaAs: 50 periods

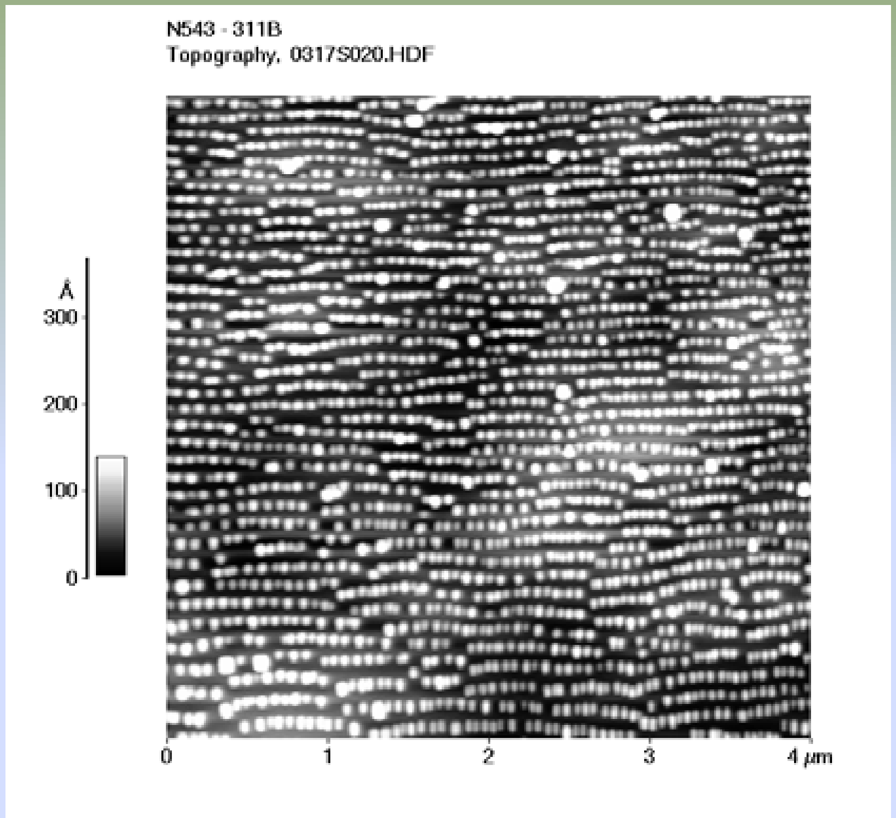


Fig. 4. AFM image of a 50-period undoped InGaAs (6.1 ML)/GaAs (20 nm) QD SL grown on {113}B GaAs substrate.

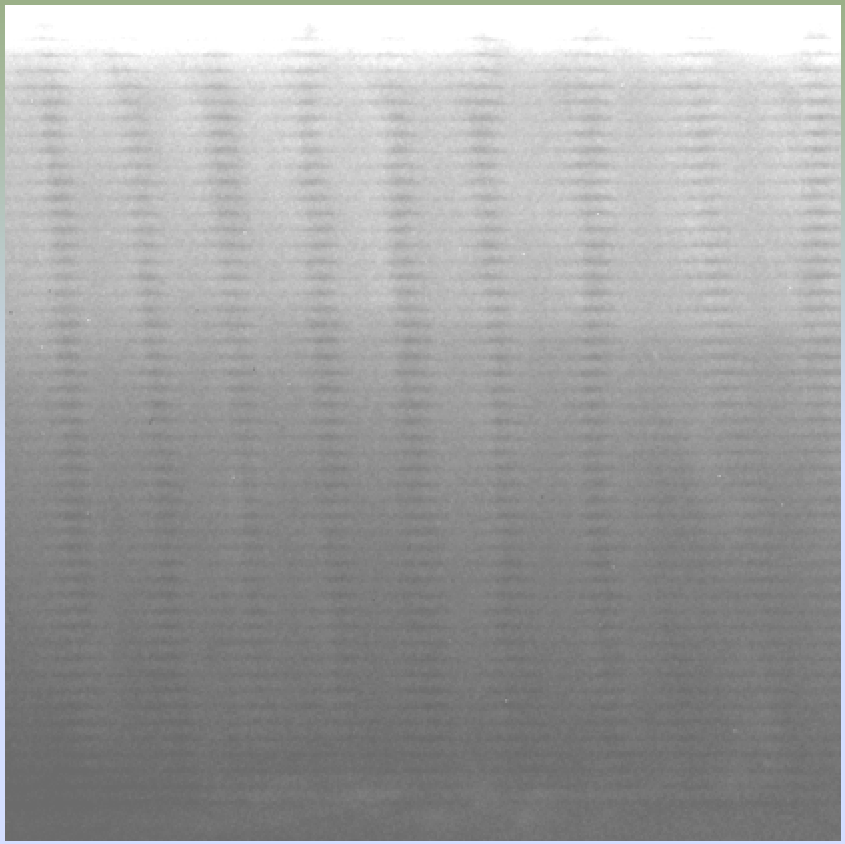
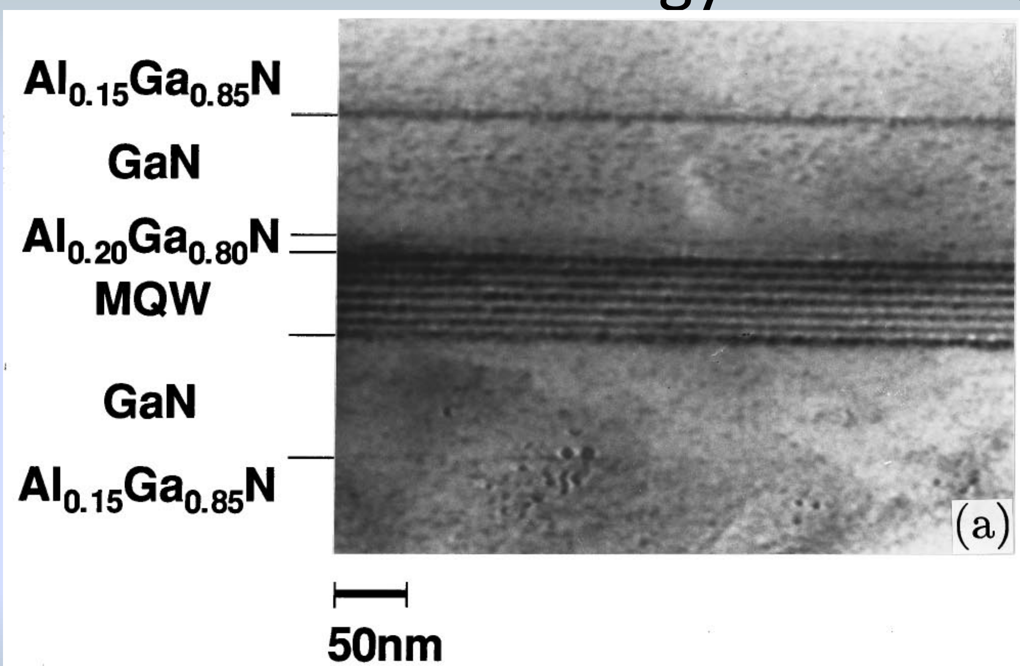
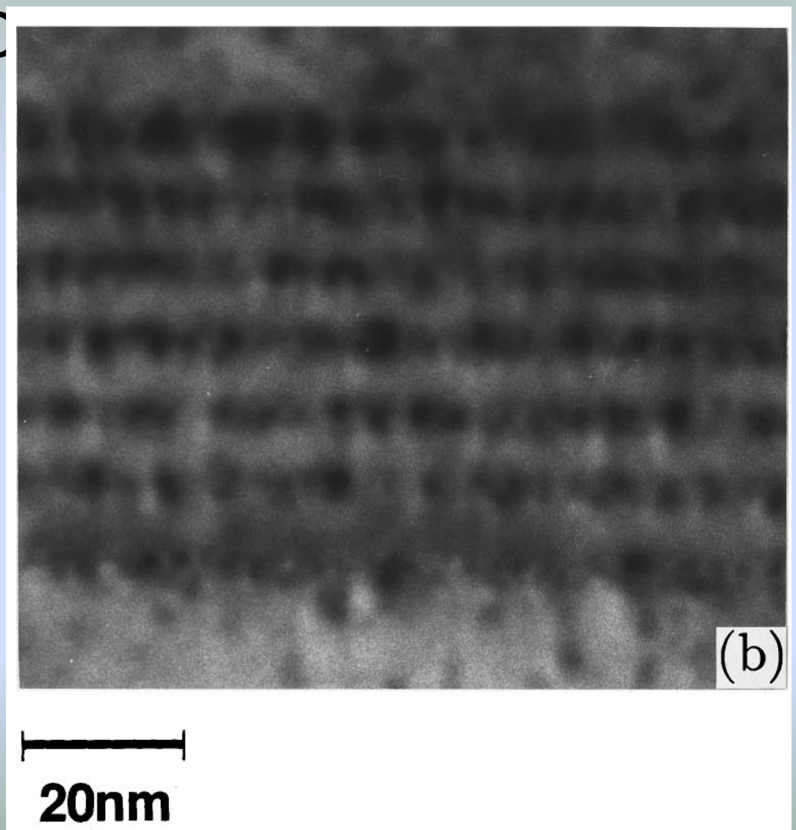


Fig. 5. <-110> pole bright-field TEM cross-section image of a 50-period undoped InGaAs (6.1 ML)/GaAs (20 nm) QD SL grown on {113}B GaAs substrate.

Composition Fluctuation in InGaN QW Blue LEDs

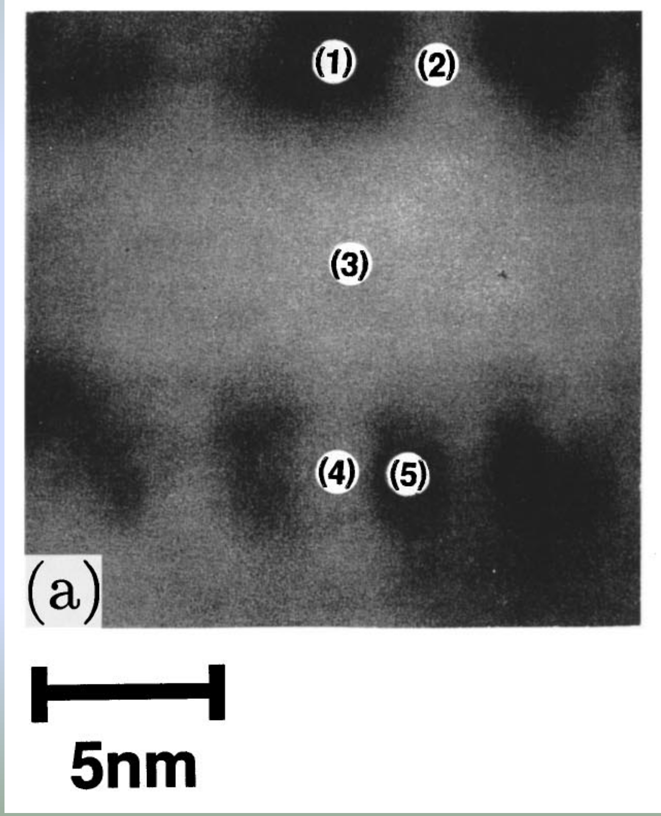
- Nominal quantum well structure:
  - $In_{0.2}Ga_{0.8}N$ 3nm well /$In_{0.05}Ga_{0.95}N$ 6 nm barrier
  - Localized energy states - QD

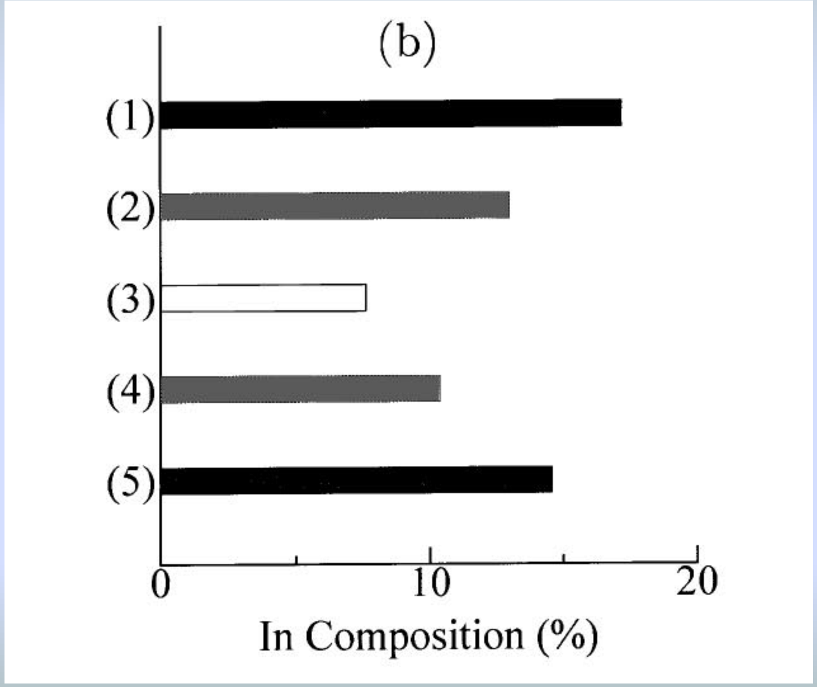




APL, 70, 981 (1997).

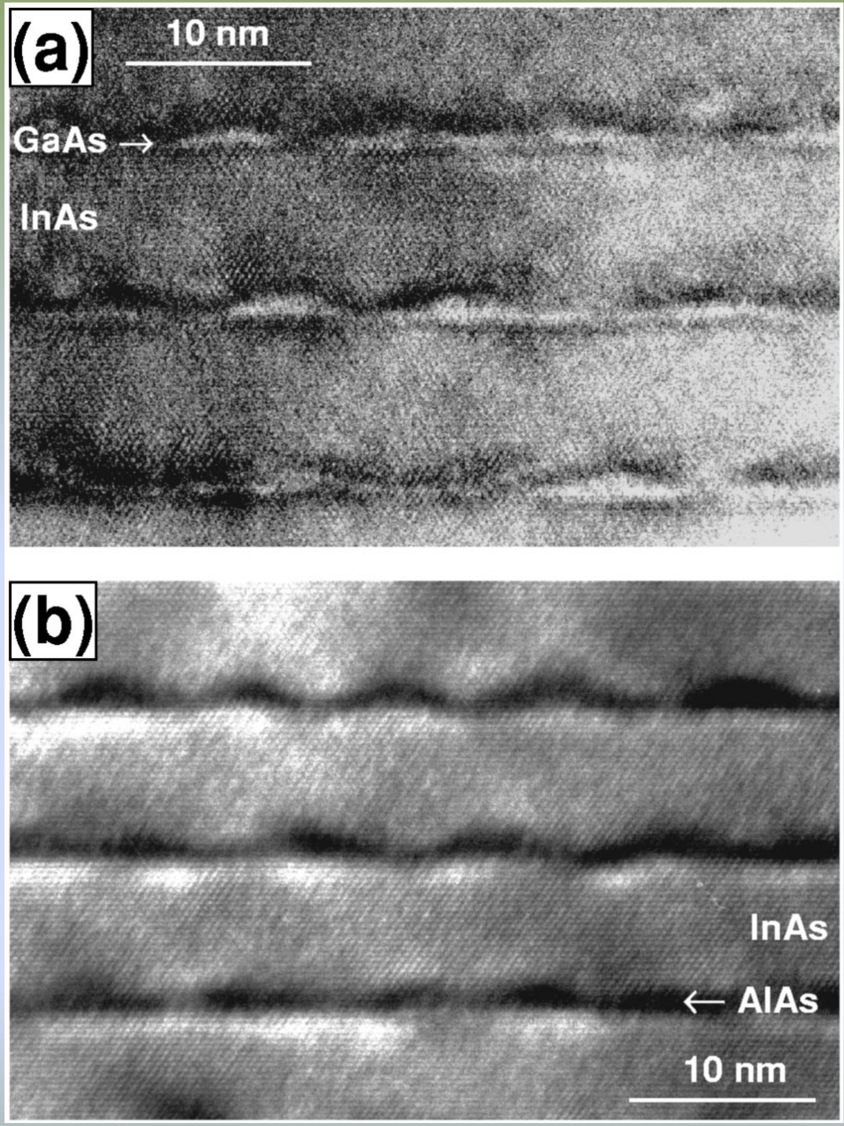
- Compositional modulation due to phase separation to produce In-rich QDs
- High quantum efficiency of the system despite > 1e8 cm-3 dislocation density is attributed to deep localization of excitons in QDs





Antidots: tensile strained QDs

- Only compressively strained materials readily form self-organized QDs
- Tensile strained QDs can only be formed under extreme non-equilibrium conditions
  - 0.06 (GaAs), 0.04 (AlAs) and 0.25 (InAs) ML/s
- Applications
  - Not fully confined e-/h$^+$ states
  - Yes confined optical phonon (phonon dots)



D. A. Tenne et al, "Raman study of self-assembled GaAs and AlAs islands embedded in InAs," *Physical Review B,* 61, 13785 (2000).

Summary

- Bottom-up self-organized QD formation
  - Critical thickness
  - S-K growth mode and coherently strained QDs
  - Energy minimization: size, shape, distribution
- Top-down patterned QD formation
  - Lithography defined growth template
    - Non-planar structure
    - Selective Area Epitaxy (SAE) or SAG
- Multi-stack QDs
- Composition modulation
- Anti-dots or nanopores