THEORY OF INTERMIXING IN Ge/Si(100) QUANTUM DOTS

P.C. KELIRES

PHYSICS DEPARTMENT, UNIVERSITY OF CRETE

&

FOUNDATION FOR RESEARCH AND TECHNOLOGY - HELLAS (FORTH)

IN COLLABORATION WITH: Ph. SONNET

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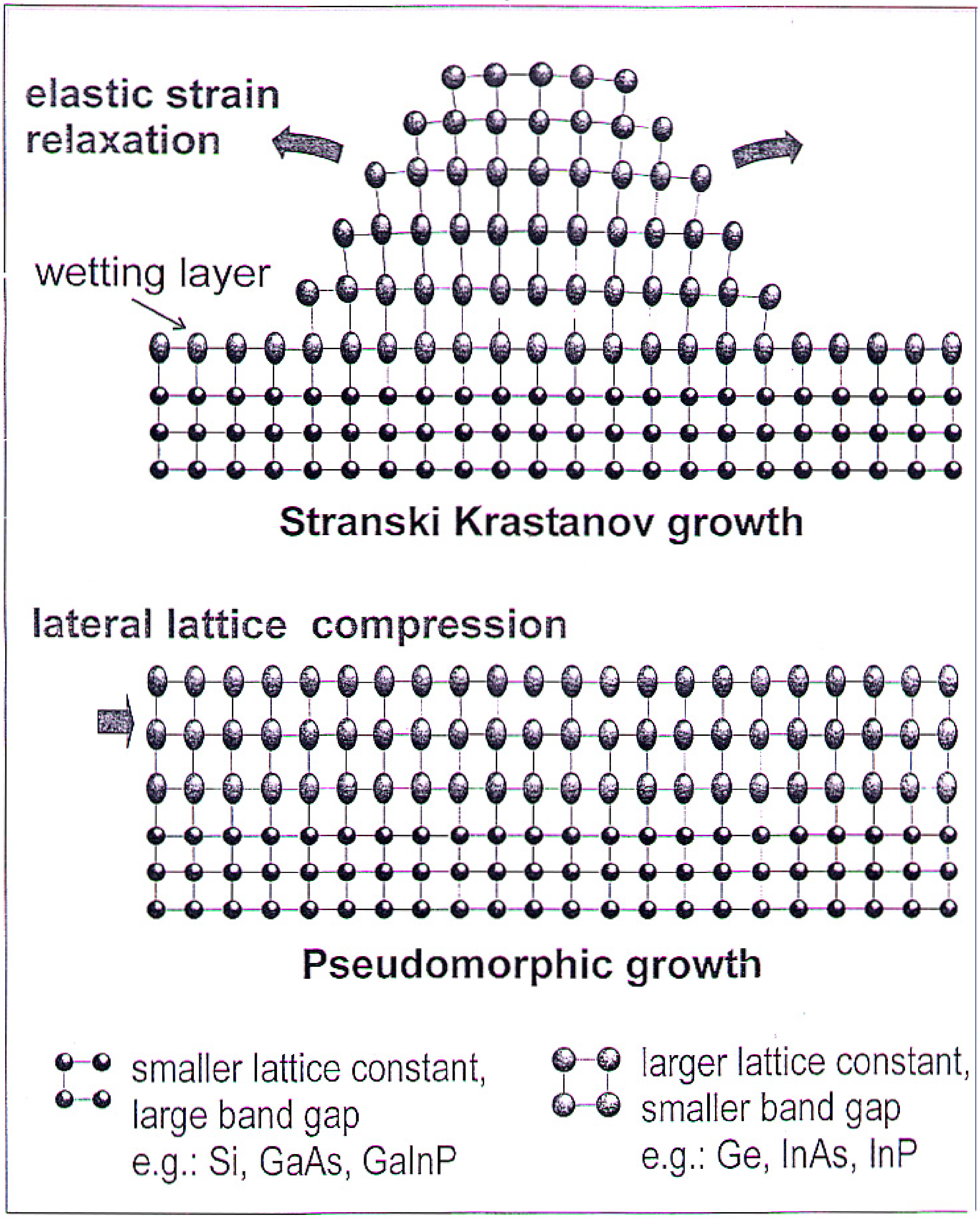
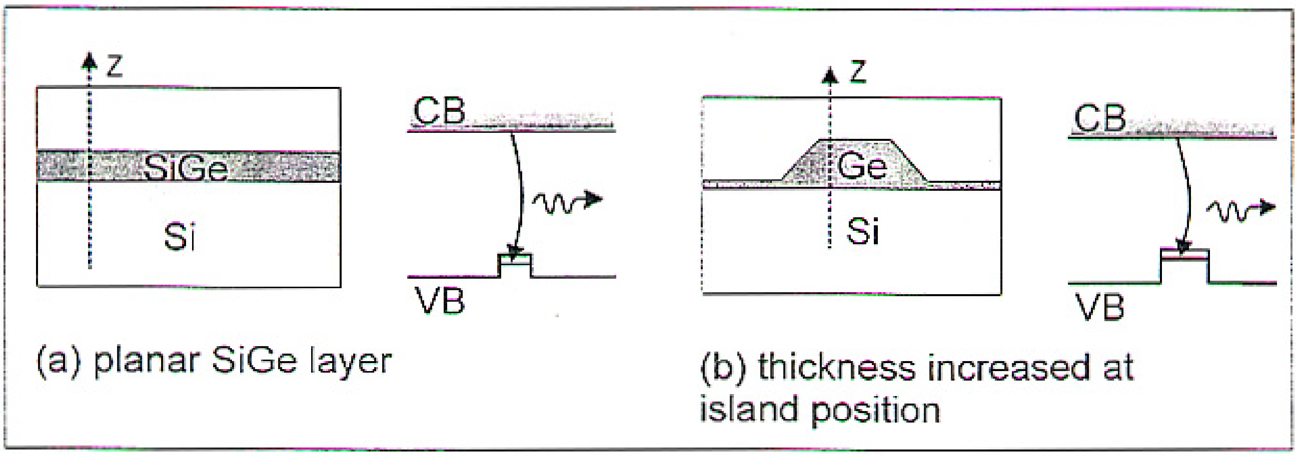

elastic strain relaxation
wetting layer
Stranski Krastanov growth
lateral lattice compression
Pseudomorphic growth
smaller lattice constant, large band gap e.g.: Si, GaAs, GaInP
larger lattice constant, smaller band gap e.g.: Ge, InAs, InP


z
SiGe
Si
CB
VB
(a) planar SiGe layer
z
Ge
Si
CB
VB
(b) thickness increased at island position

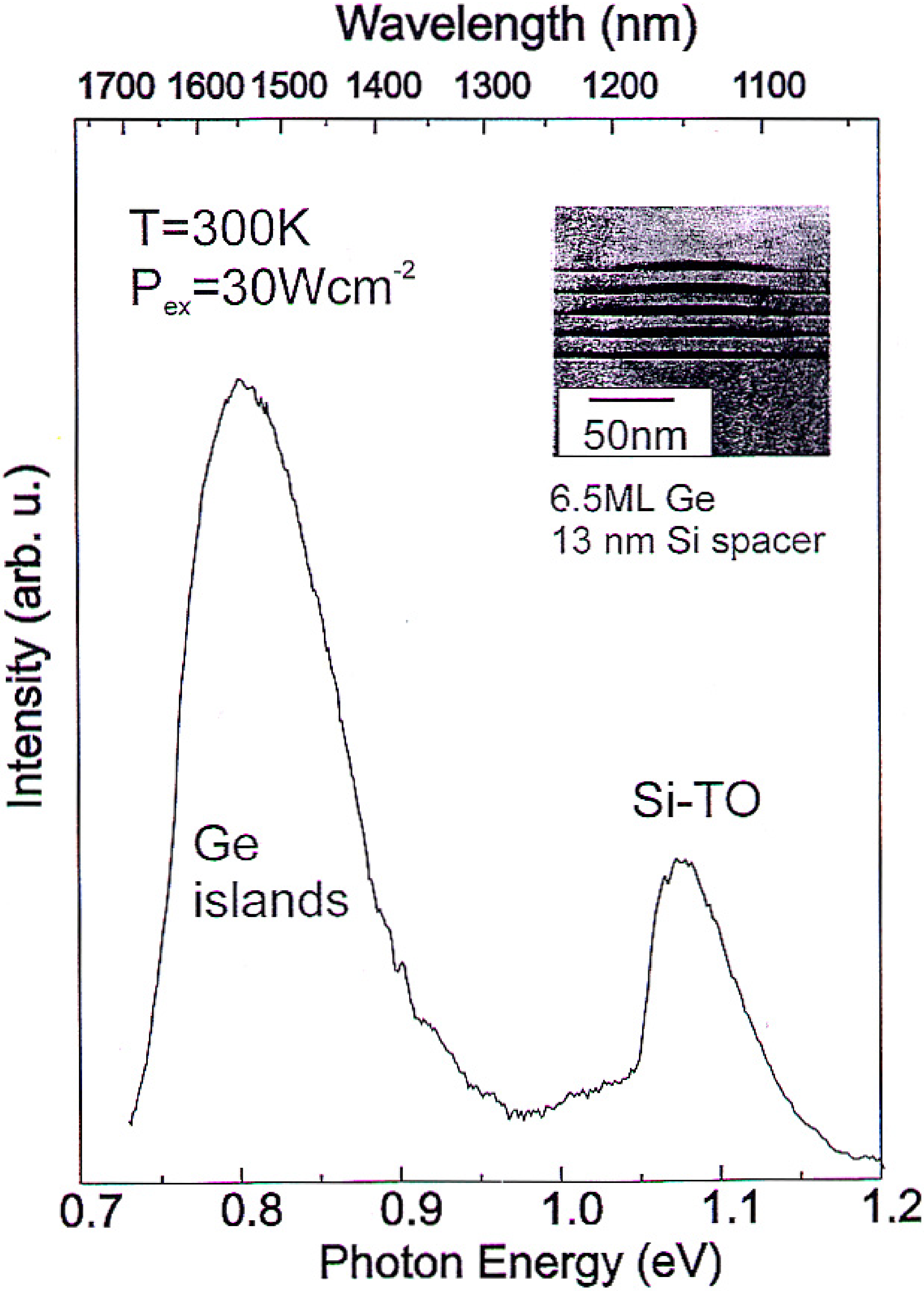


Fig. 4 K. Eberl et al.

Crucial parameters: size of QDs, alignment, strain, composition.

Well studied: island nucleation processes, strain-driven mechanisms within MTE (Tersoff)

Significant gap: interdiffusion, compositions, stress fields.

→ shows the difficulty in theory + exp. to equilibr inhomogeneous environments in semiconductors.

Here, for the first time: study intermixing, extract compositions at typical growth T's and correlate to stresses.

Ingredients:

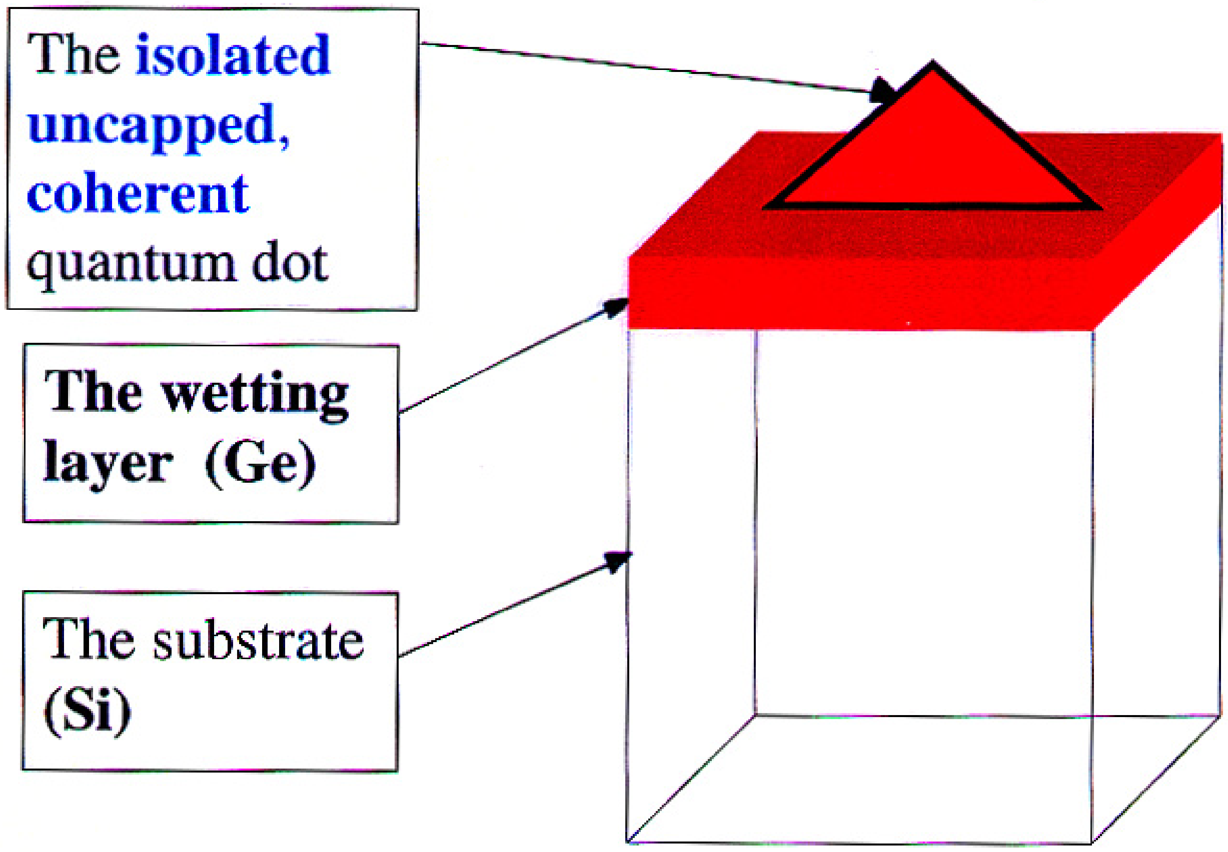
→ Novel Monte Carlo simulations

→ Empirical potential approach (Tersoff's)

→ Minimization of Gibb's FE at finite T's.

MODEL

=> **Supercell** consisting of 10 layers of Si (substrate), 3 layers of Ge (the wetting layer) and the Ge quantum dot



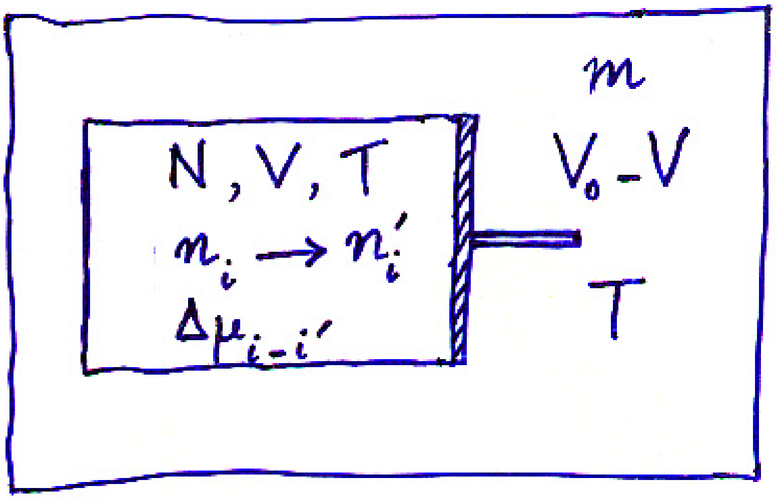
→ The two last layers are fixed

=> Total number in the cell: **8000-15000 atoms**

Semi-grand canonical ensemble

(ἡμι-μεγαλοκανονική συλλογή)

(Foiles, 1985; metals — Kelires & Tersoff, 1989; semiconductors)



$(\Delta\mu, N, P, T)$

Exchanges of particles within the system & exchanges of volume with heat bath.

- SGC ≡ GC + (N, P, T)

- Besides usual random atomic + volume moves,

we have Ising-type identity flips:

$$Si \leftrightarrow Ge \leftrightarrow C \leftrightarrow Si$$

∴ model "diffusion"; inaccessible by MD.

Metropolis scheme :

Traditional moves

$U \longrightarrow U'$: atom moves $(\vec{r}^N \longrightarrow \vec{r}'^N)$

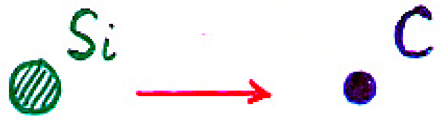
$V \longrightarrow V'$: volume moves

Acceptance probability :

$$P_{acc} = \text{Min}\left[1, \exp(-\beta \Delta W)\right] \sim e^{-\Delta W / k_B T}$$

$$\Delta W = (U' - U) + P(V' - V) - N k_B T \ln\left(\frac{V'}{V}\right)$$

Identity flips

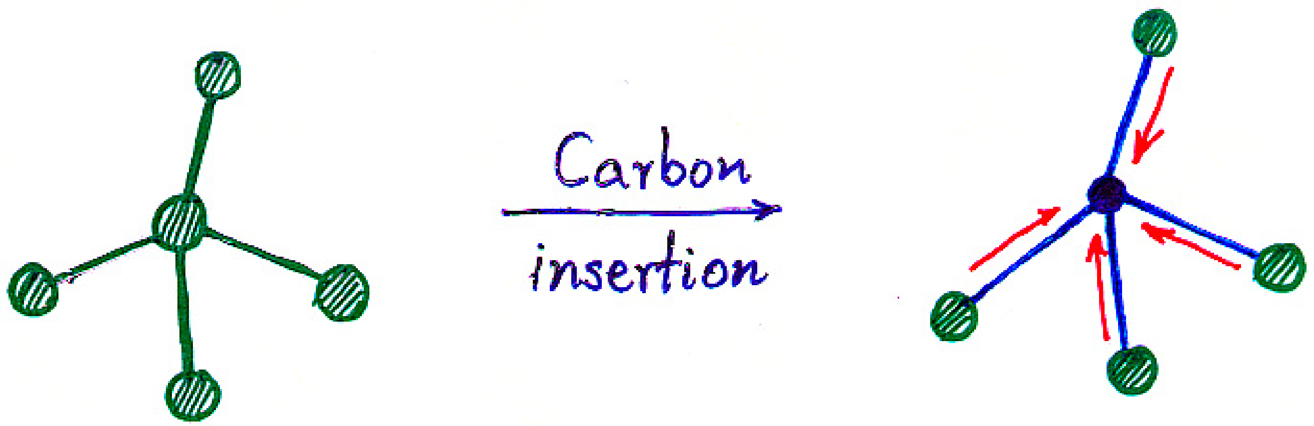


$$P_{acc}(i \longrightarrow i') = \text{Min}\left[1, \frac{e^{\beta \mu_i'}}{e^{\beta \mu_i}} \exp(-\beta \Delta U)\right]$$

$$\sim e^{\beta \Delta \mu} \; e^{-\beta \Delta U(\vec{r}^N)}$$

$$\Delta \mu = \mu_{Si} \Delta n_{Si} + \mu_{Ge} \Delta n_{Ge} + \mu_C \Delta n_C$$

For systems with large atomic size mismatch



$$\Delta U(\vec{r}^N) = E_{cluster}\left\{ i \to i', \sum_{k}^{nn} \Delta\vec{r}_k(\vec{r}_{0k}) \right\} - E^0_{cluster}$$

relaxations of nn

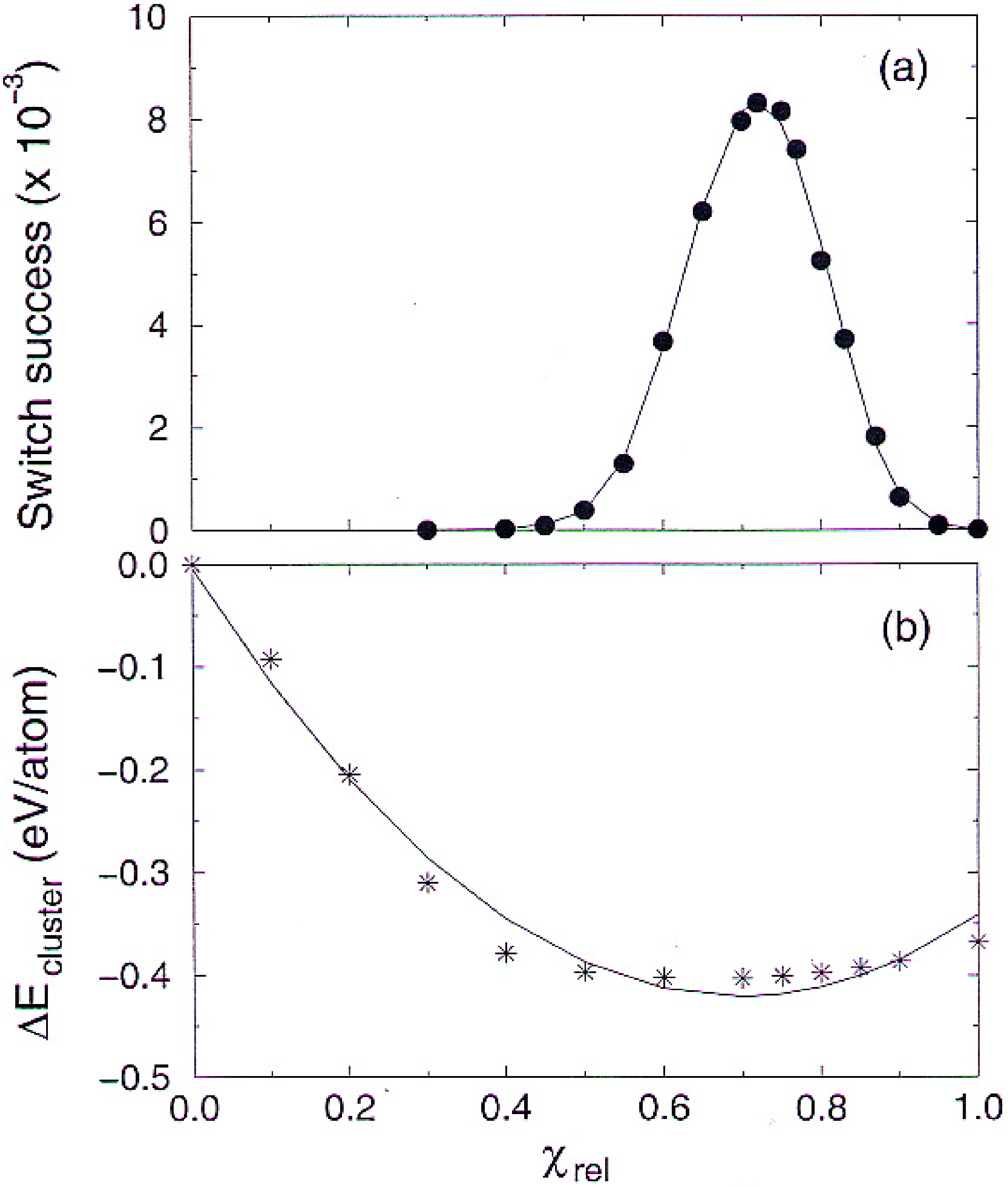
$$\Delta\vec{r}_k(\vec{r}_{0k}) = A_{bond}\,\vec{r}_{0k}$$

$$A_{bond} = \left\{ b_{0k}[i'(0), i(k)] - |\vec{r}_{0k}| \right\} x_{rel} / |\vec{r}_{0k}|$$

$x_{rel} = 0.0, \ldots\ldots, 1.0$: relaxation parameter

[P.C. Kelires, PRL 75, 1995] "Biased" Monte Carlo

Effect of relaxations on acceptance rate



Atomic Level Stresses (Vitek & Egami, 1987)

- Under no external forces, due to local incompatibilities – in networks with non-equivalent atoms and in any disordered structure
- By analogy to the macroscopic pressure, define atomic stress as a local compression:

$$\sigma_i = -\frac{dE_i}{d\ln V} \sim p_i \Omega_i$$

E_i : atomic energy (decomposition of total energy)

V : system volume

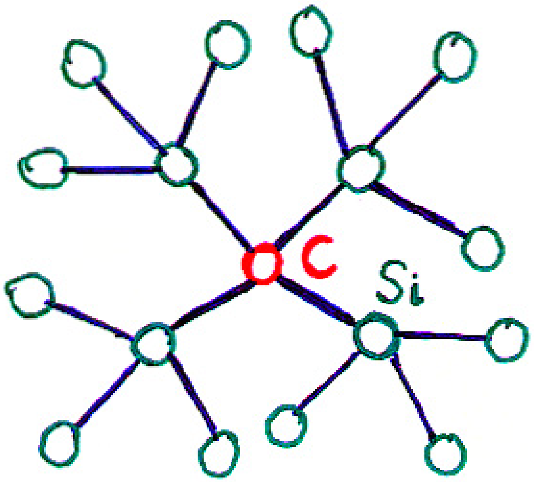
p_i : local hydrostatic pressure (trace of the stress tensor)

Ω_i : appropriate atomic volume

(Kelires & Tersoff, PRL 63, 1989)

Atomic level stresses

Vitek & Egami (1987): under no external forces, due to local incompatibilities



$c\text{-}Si_{1-x}C_x$ alloy

networks with non equivalent atoms and in any disordered structure

Formally, in the general quantum-mechanical case

$$\mathcal{H} = \sum_n \frac{p_n^2}{2m_n} + V_{int} \quad \text{(exclude external influences)}$$

Total stress:

$$\sigma^{\alpha\beta} = \sum_n \left(\Psi \middle| r_n^\beta \frac{dV_{int}}{dr_n^\alpha} - \frac{p_n^\alpha p_n^\beta}{m_n} \middle| \Psi \right)$$

α, β: Cartesian coordinates

Kelires & Tersoff (1989): a simplified way - for isotropic systems deal with the trace of the stress tensor, which defines a local compression (or hydrostatic pressure).

So, define atomic stress

$$\sigma_i = -\frac{dE_i}{d\ln V} \sim p_i\,\Omega_i$$

E_i : atomic energy (decomposition of total energy)

V : system volume

p_i : local hydrostatic pressure - describes local density fluctuations

Total stress : $\sigma = \sum_i \sigma_i$

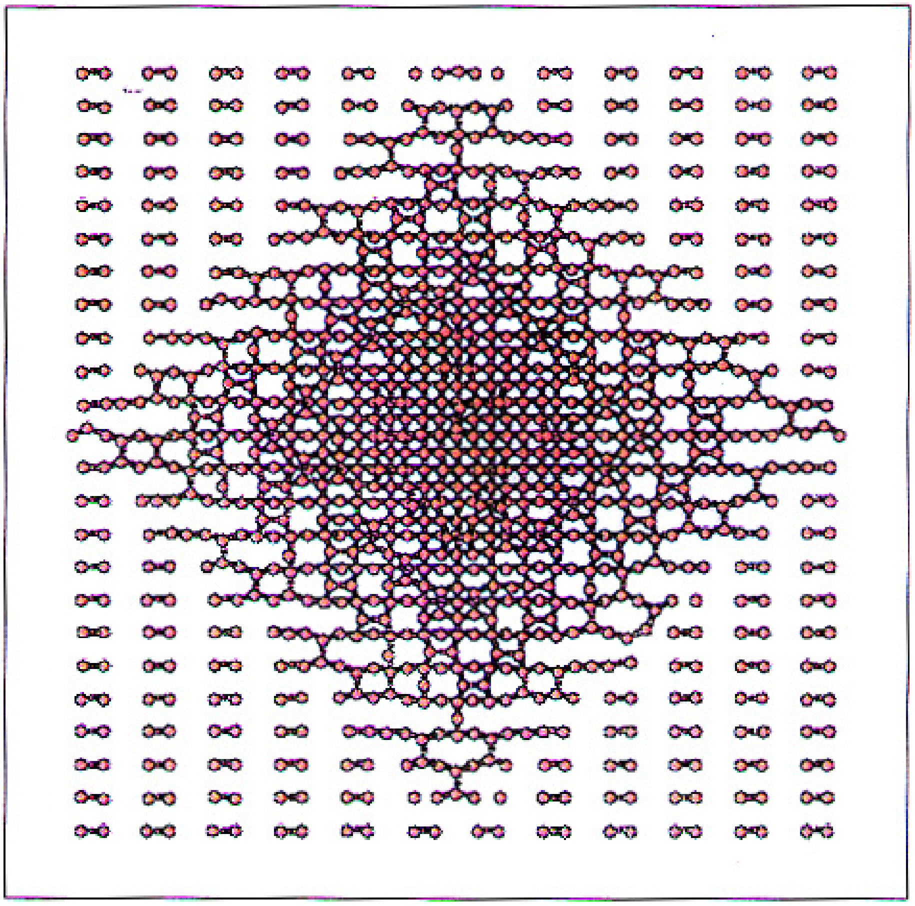
Could be zero if σ_i compensate each other

Does not mean that σ_i are diminished.

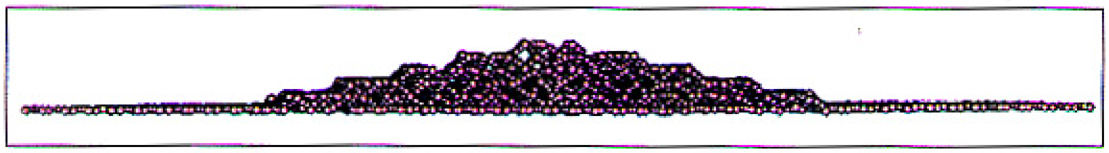
GEOMETRIC PROPERTIES

pyramidal shape – square base

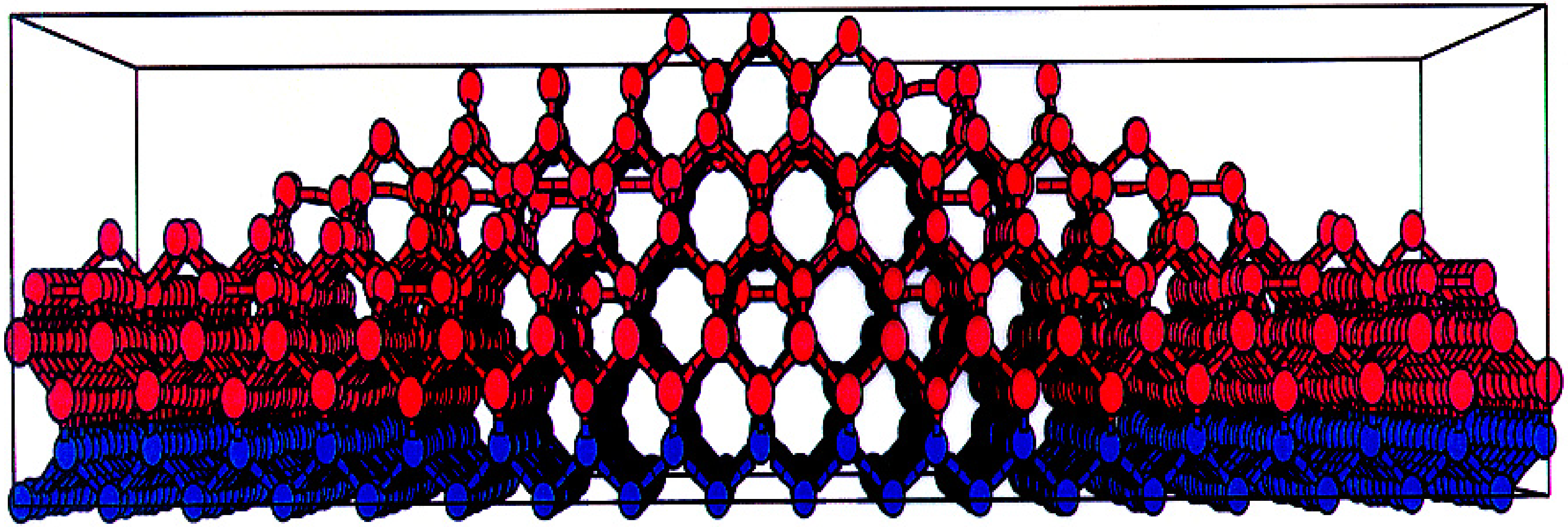
Top View



Side View

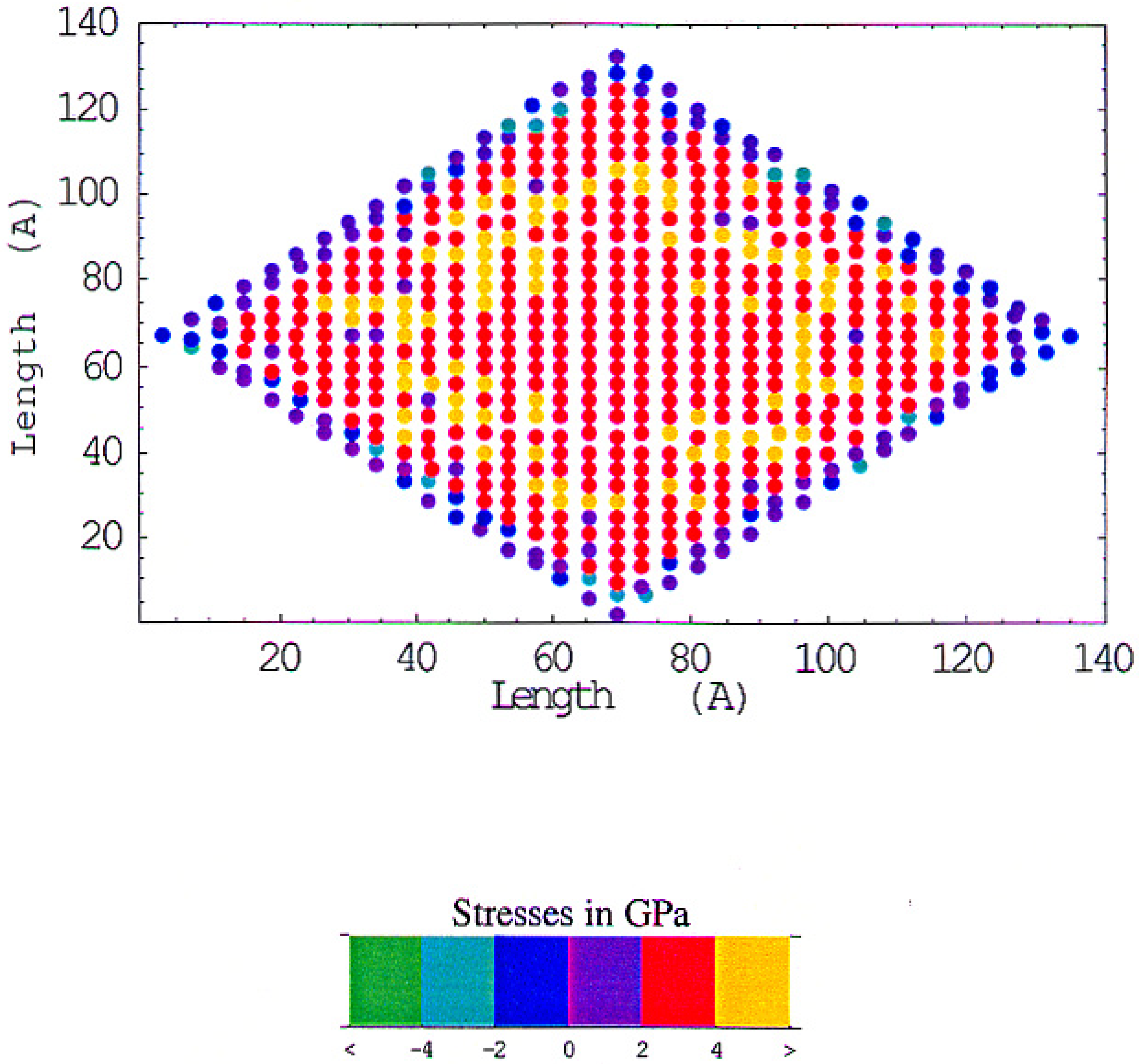


Ideal Ge QD on Si (100)



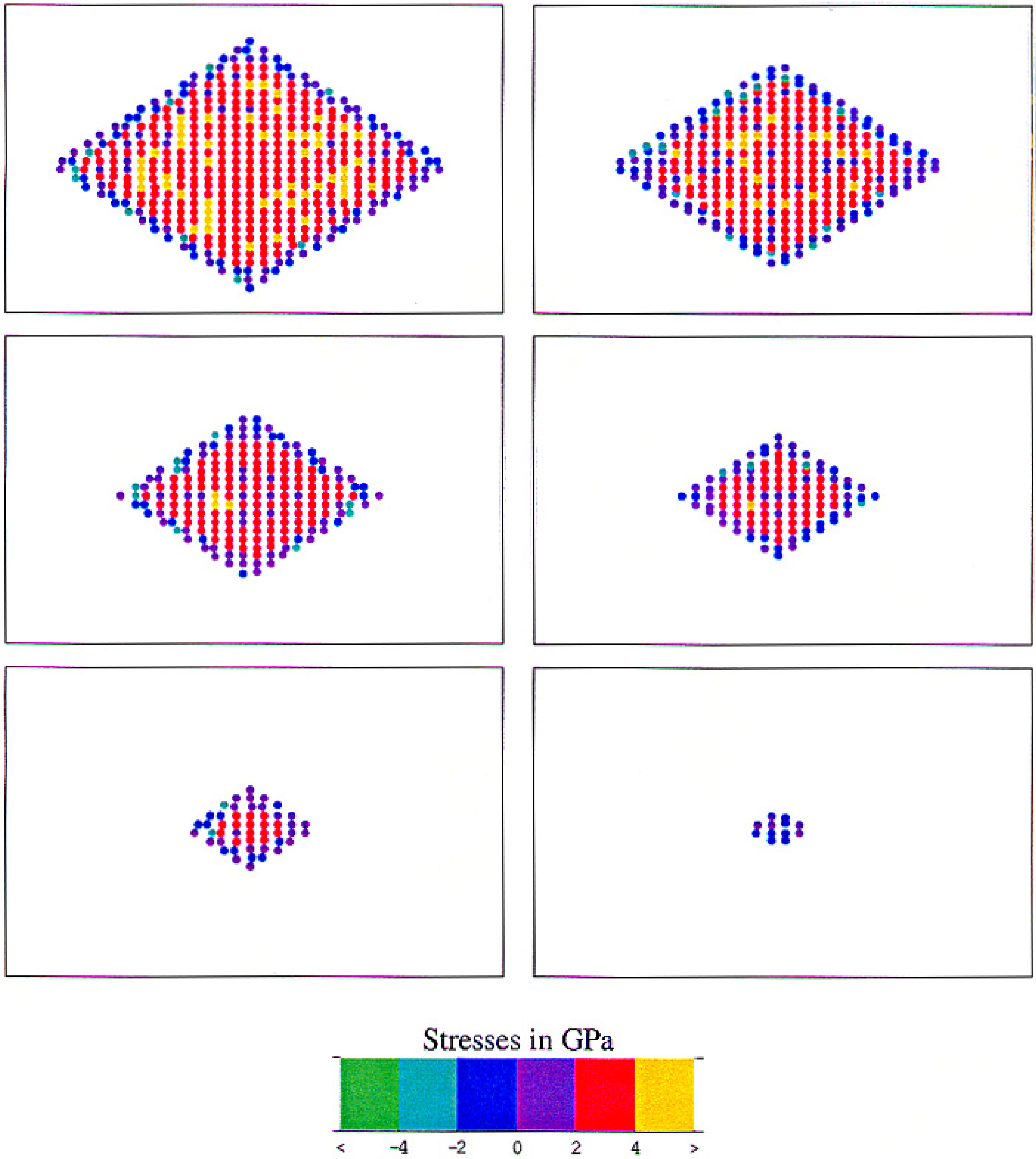
Atomic Stresses

1st QD Layer

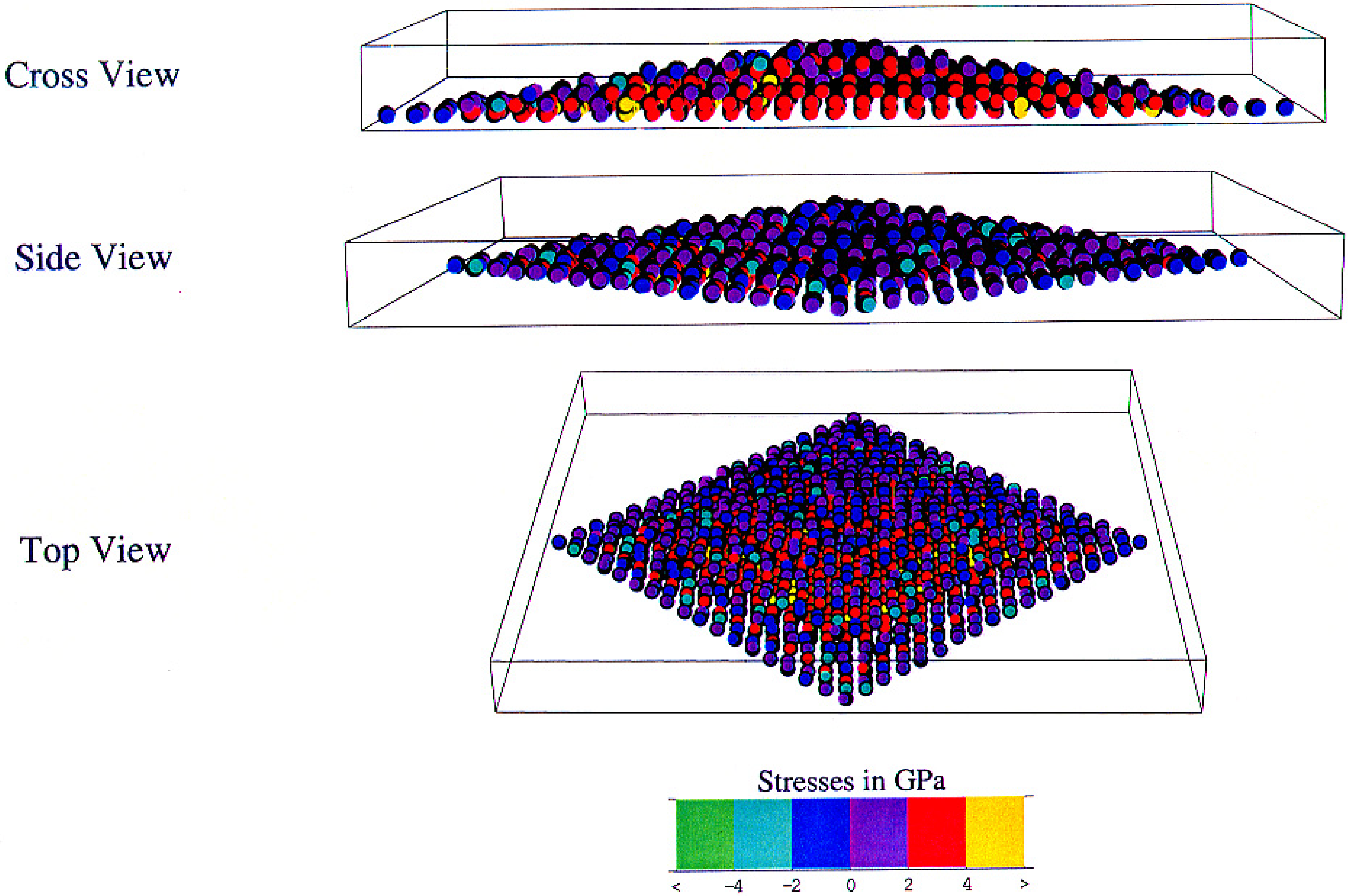


Atomic Stresses Layer by Layer

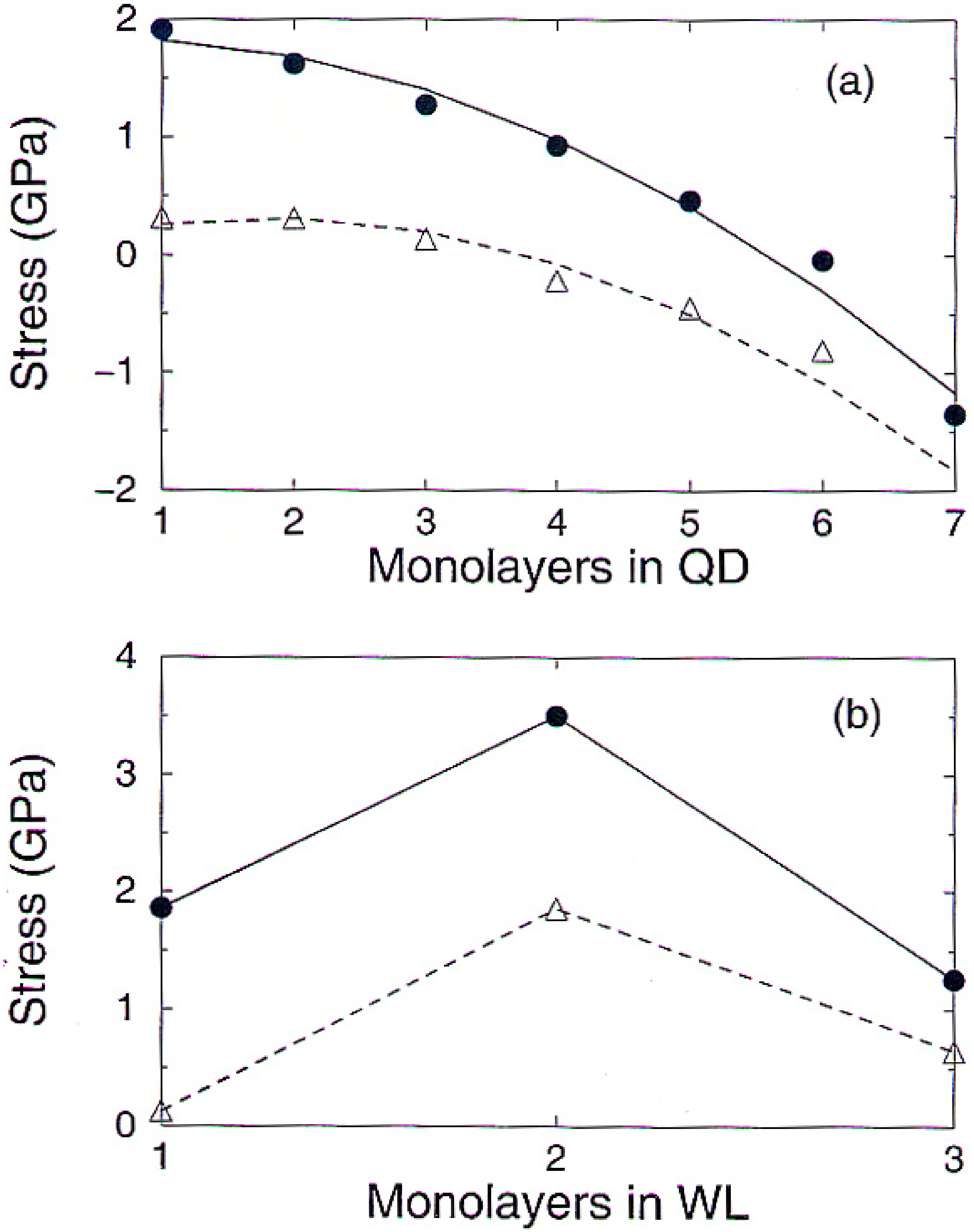
2nd – 7th QD Layer



Atomic Stresses in QD

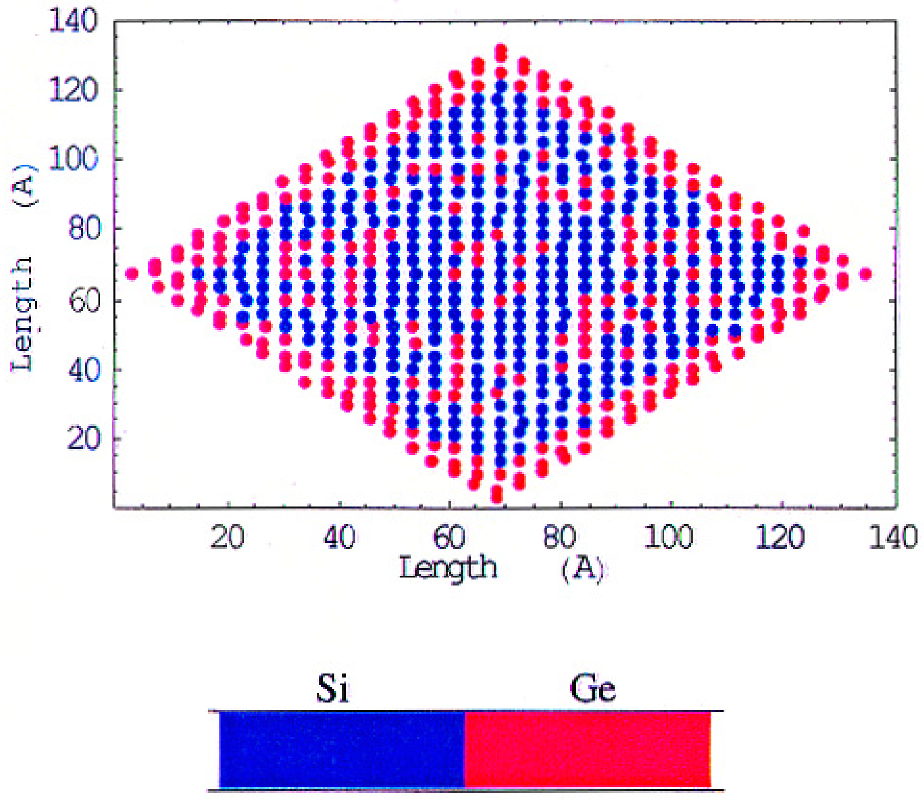


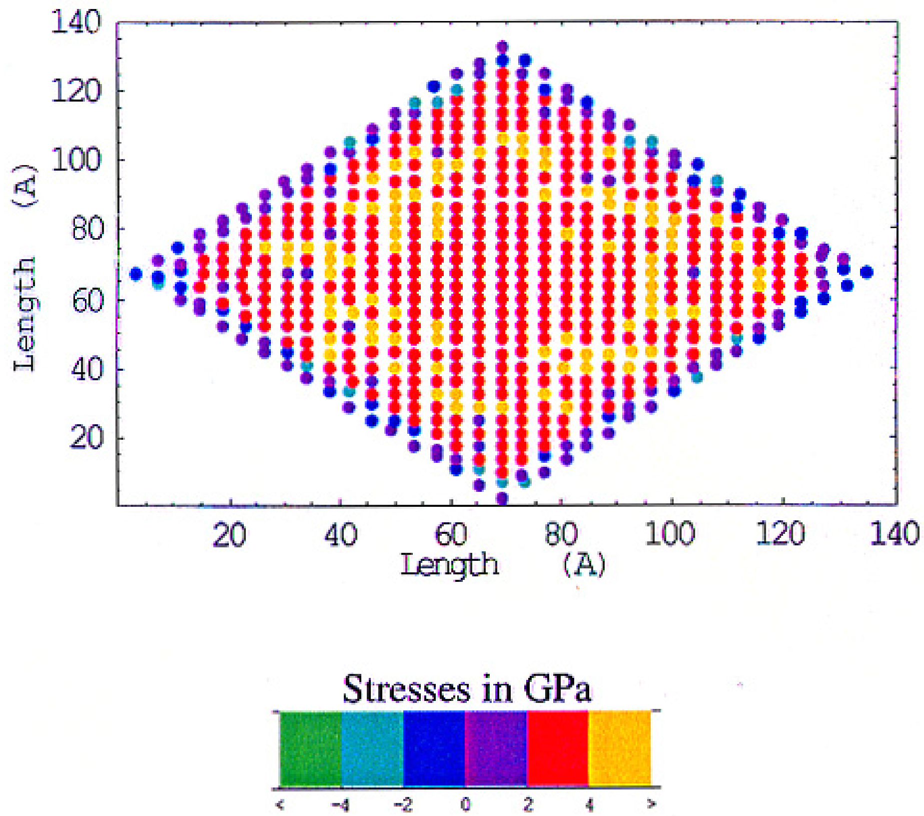
Stress per layer in QD and WL



Equilibrium Compositions vs Atomic Stresses in QD

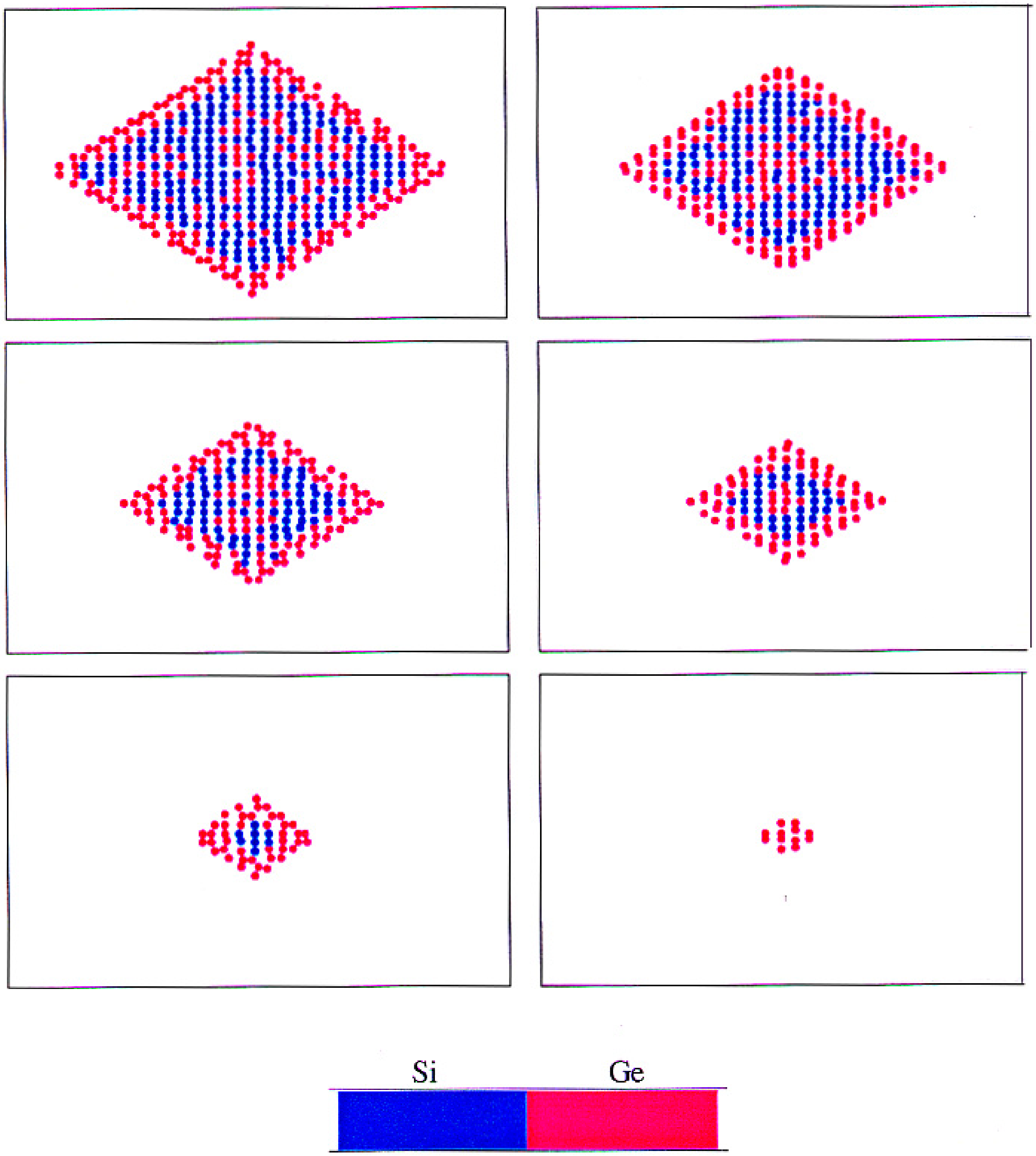
1st QD Layer





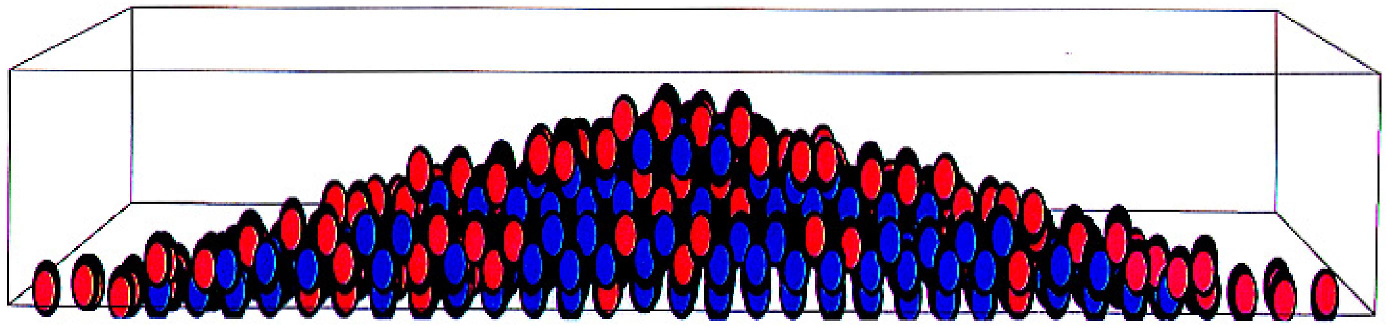
Equilibrium Compositions Layer by Layer

2nd – 7th QD Layer

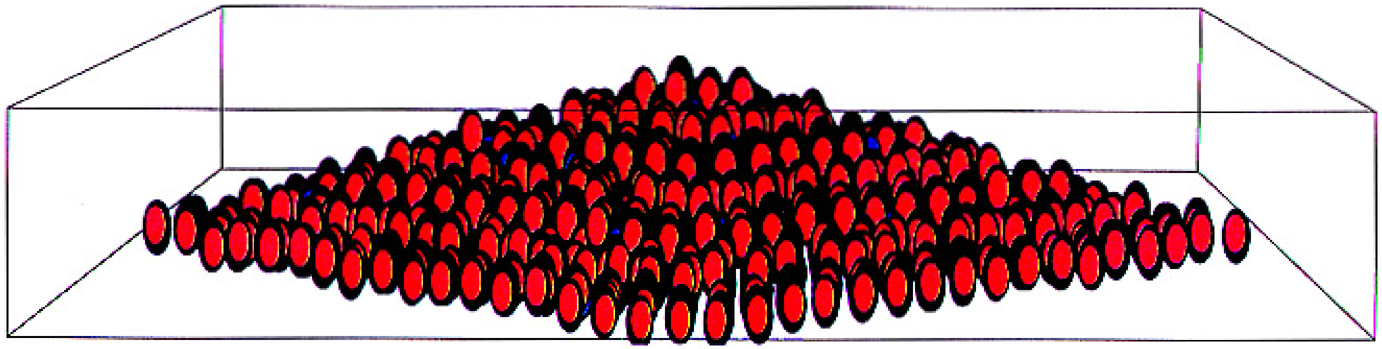


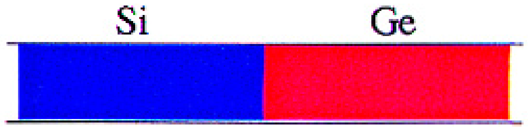
Equilibrium Compositions in QD

Cross View



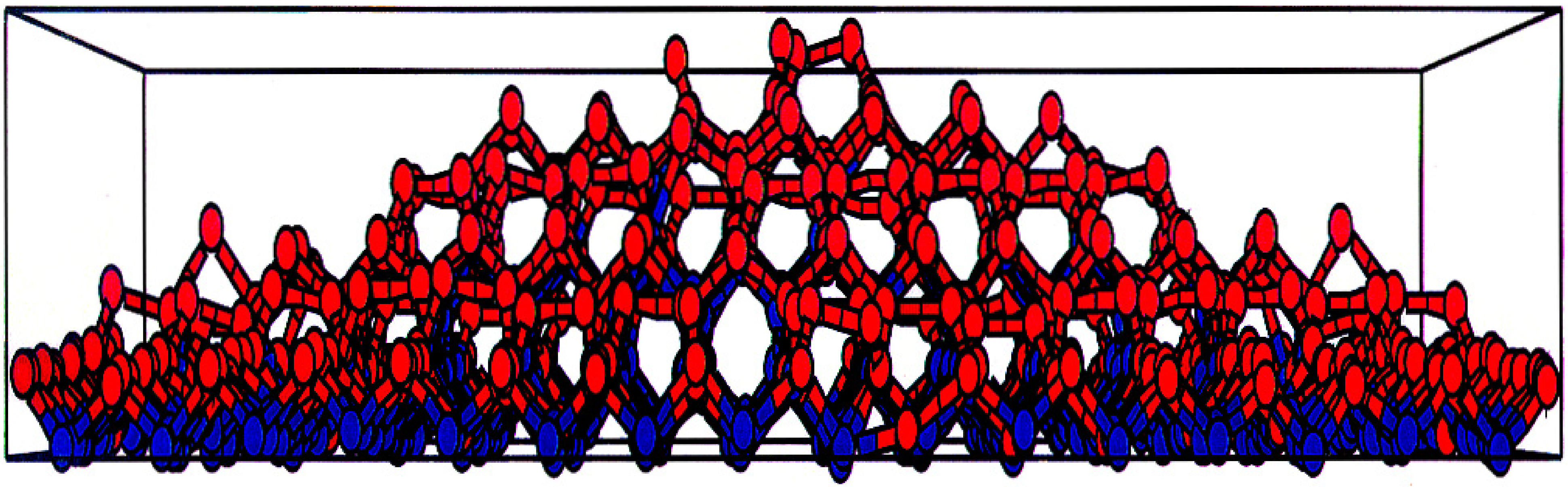
Side View





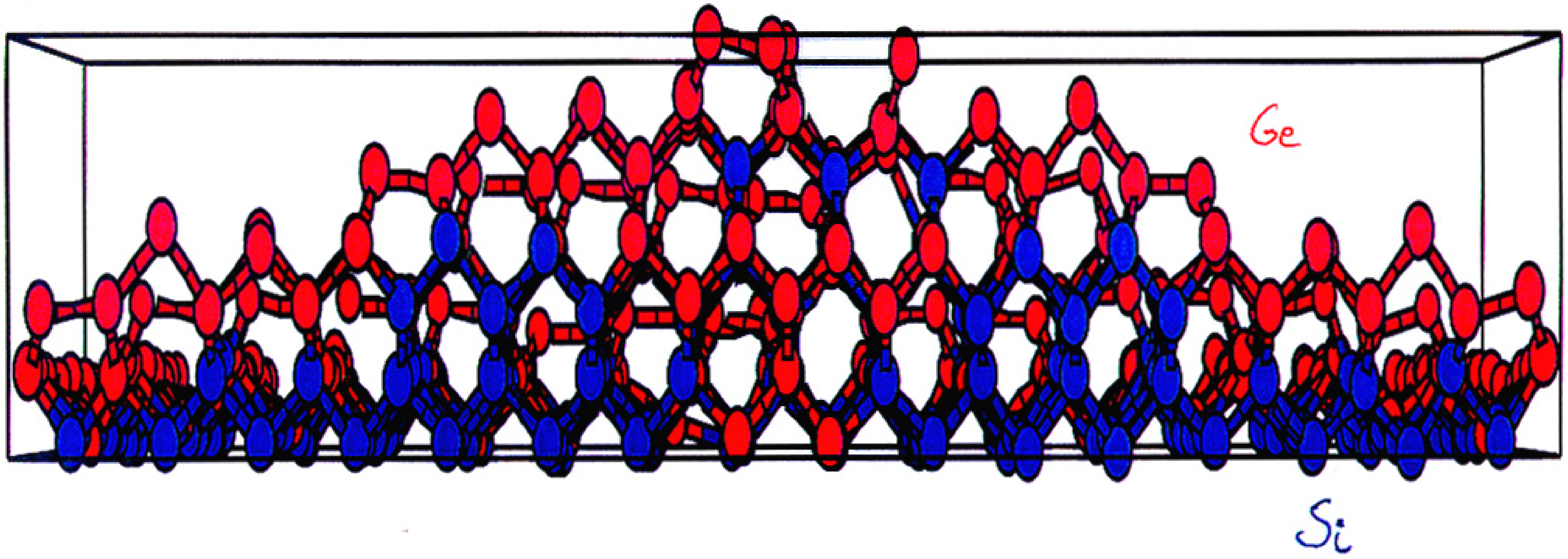
Intermixing in QD and WL at 800 K

(Side view)

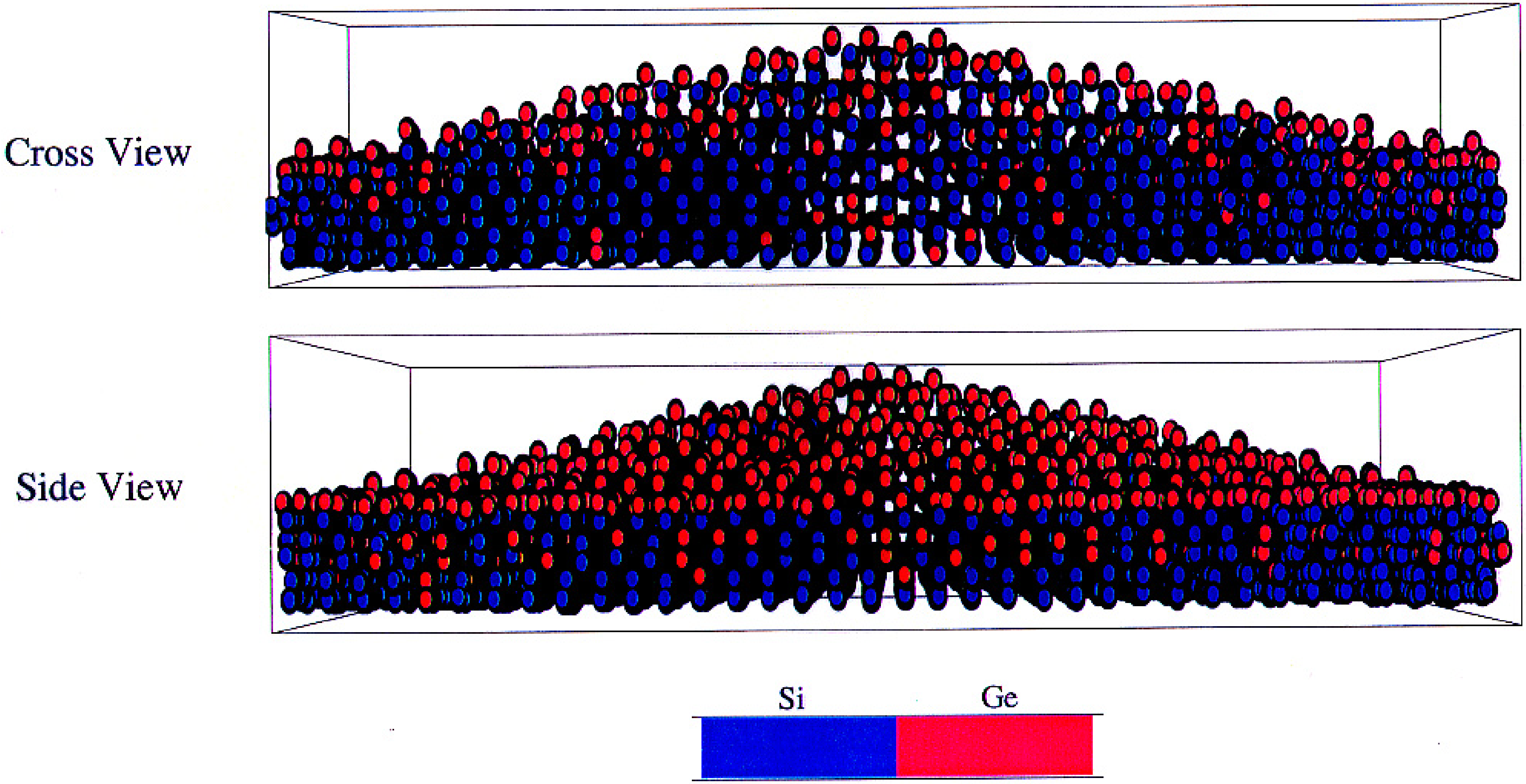


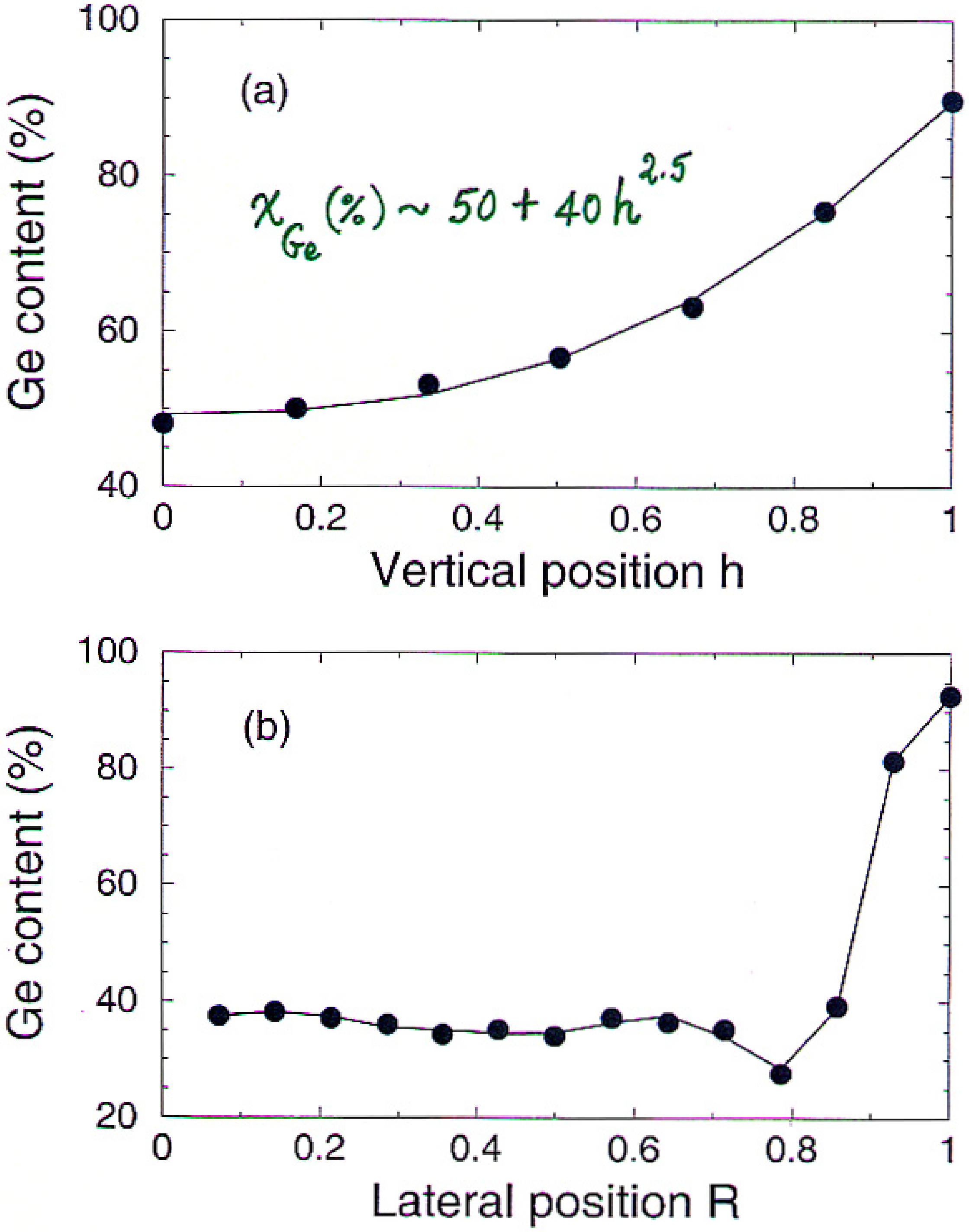
Intermixing in QD and WL at 800 K

(Cross view)



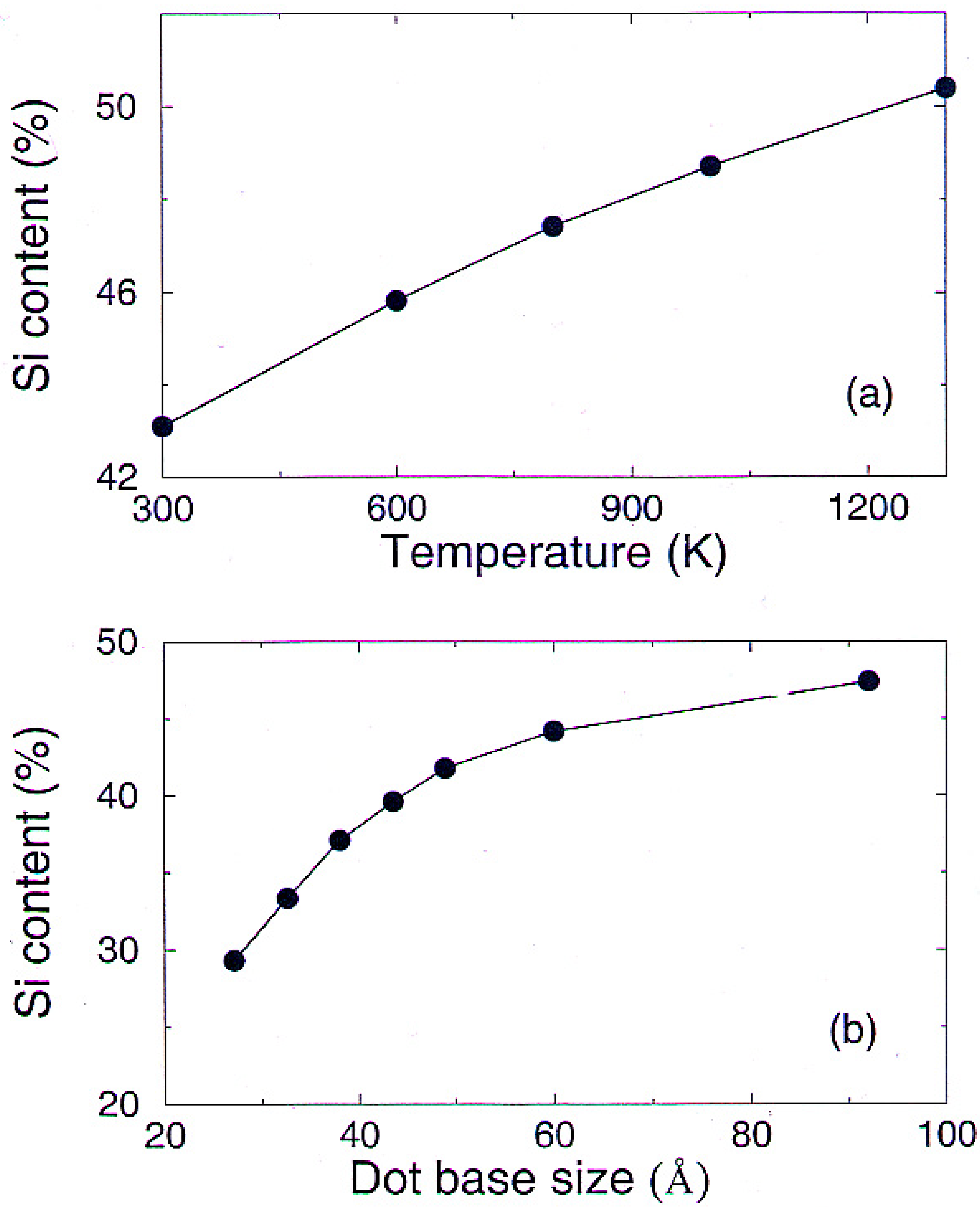
Equilibrium Compositions in QD / WL / Substrate System





Vertical and lateral (1st layer) variation of Ge content in QD

Temperature and size variation of composition



Appl. Phys. Lett., Vol. 78, No. 3, 15 January 2001

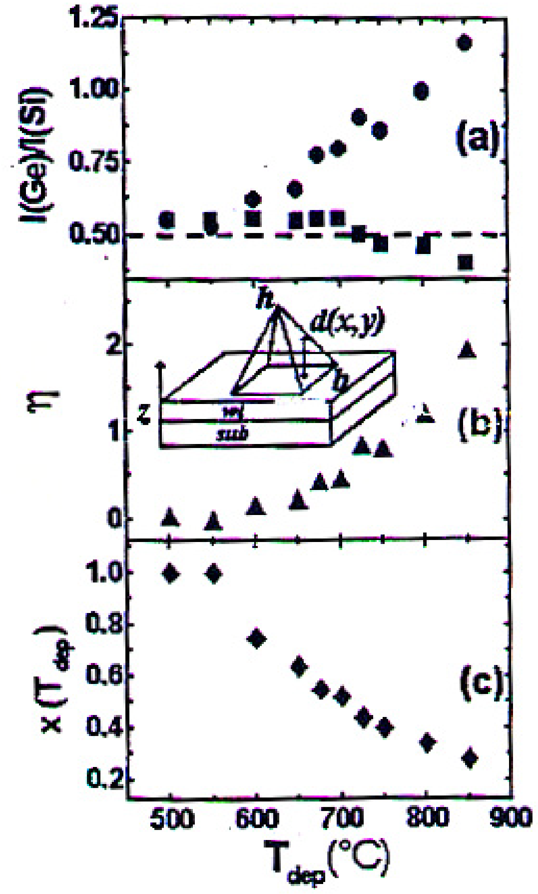


FIG. 2. (a) XPS intensity ratios of the Ge3*d* and Si 2*p* core levels as a function of T_{dep}: (■), experimental ratio R_{expt} calculated ratio R_{calc} with the reported model in the assumption of no intermixing. (●) (b) Normalized difference $\eta=(R_{expt}-R_{calc})/R_{expt}$ as a function of T_{dep} in the $x=1$ limit. (c) Ge average content x in the epilayer as a function of T_{dep}.

Capellini, De Seta, Evangelisti (APL 78, 2001)

Boscherini et al., 2000 (EXAFS)

Conclusions

- Compositional equilibration in Ge islands on Si(10
made possible by Monte Carlo simulations.
- Atomic stresses probe the stress field locally, site by site.
- Interdiffusion driven by stress relaxation both in the QD and the wetting layer.
- Stresses strongly coupled with average site occupancies
- Inhomogeneous compositions both vertically and laterally.
- Rather weak temperature dependence of compositions interplay between surface energies, stress fields, and chemical randomness.
- Limiting Si content as size of QD increases.
- Future studies: Carbon-induced Ge dots; strain coupled with strong chemical effects.