In-Situ Observation of Surface Kinetics During MBE Growth Using Synchrotron X-ray Diffraction

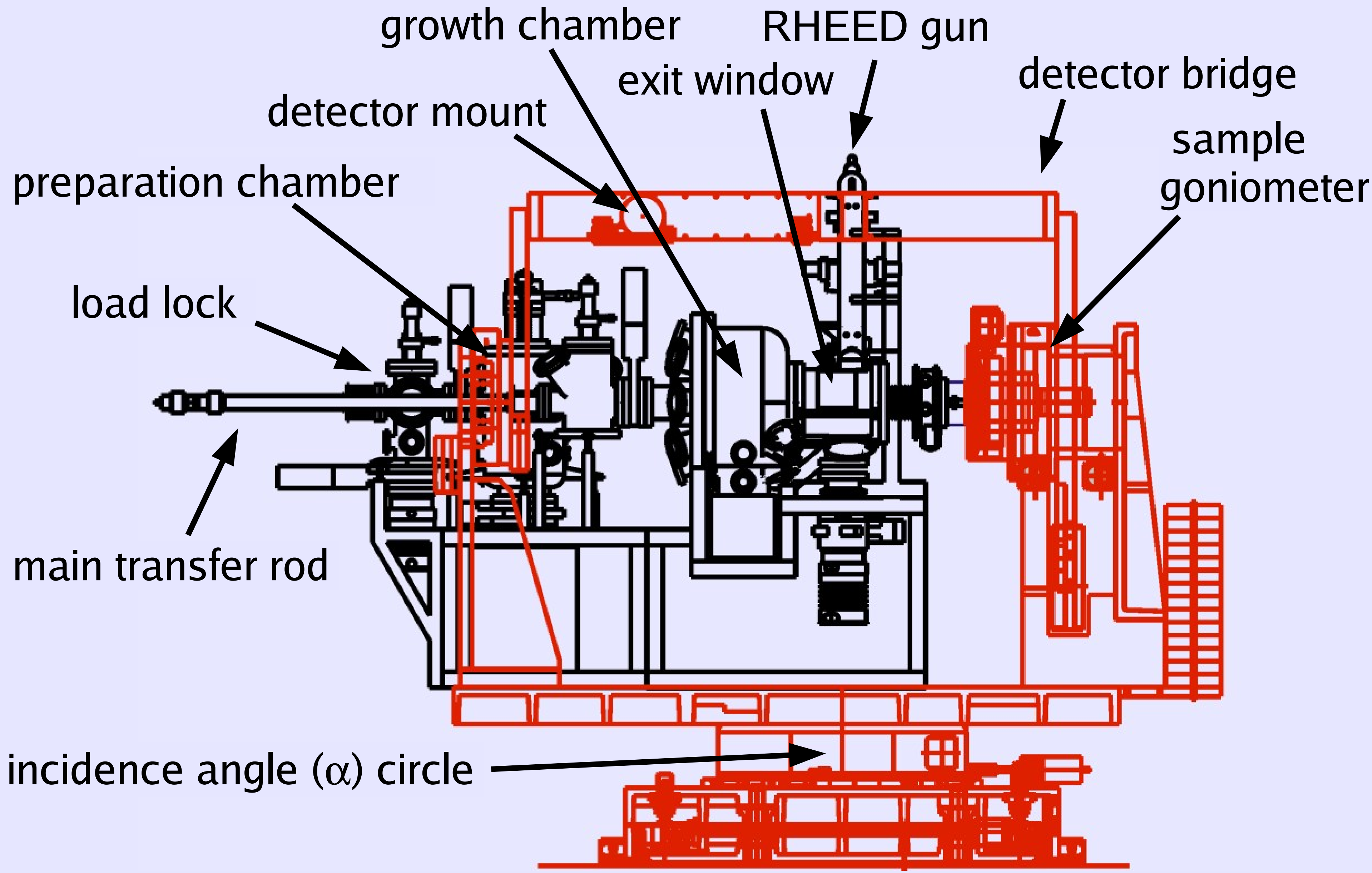
Wolfgang Braun

Paul-Drude Institute for Solid State Electronics, Berlin

Motivation

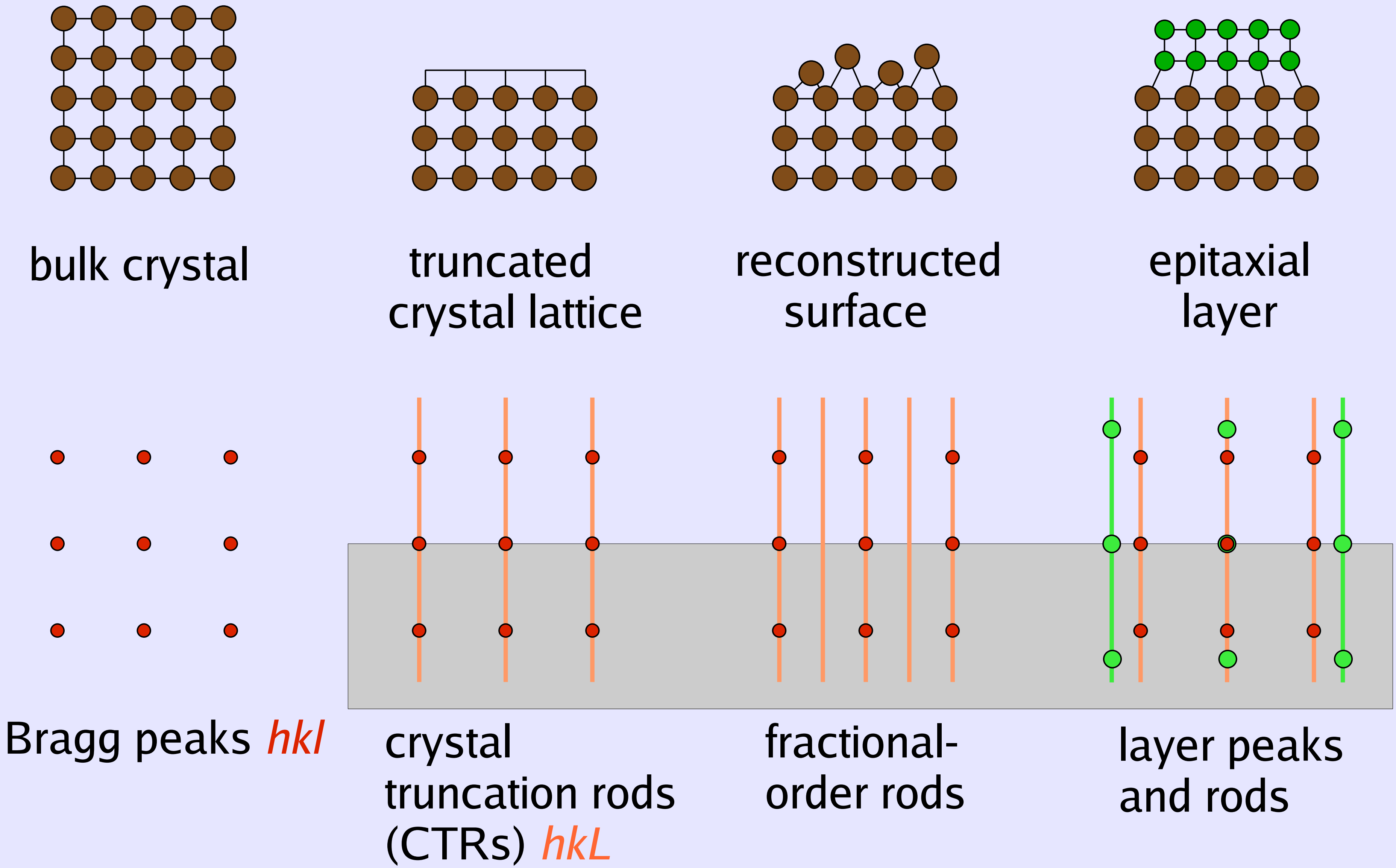
- Study MBE crystal growth in situ under typical vacuum conditions
- Quantitative analysis by kinematical theory
- Subsurface penetration: interface structures
- Large ensembles: reliable statistics
- Reciprocal space analysis: spatial filtering

Experiment

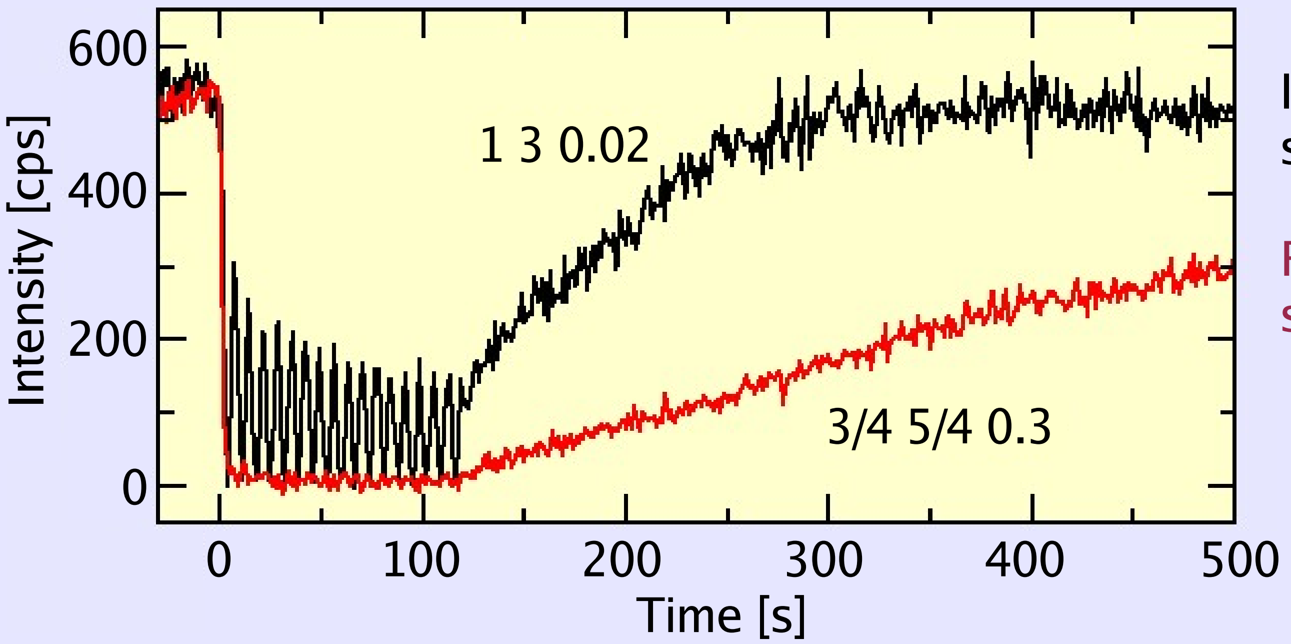


growth chamber
RHEED gun
exit window
detector bridge
detector mount
sample goniometer
preparation chamber
load lock
main transfer rod
incidence angle (α) circle

Bulk diffraction vs. surface diffraction



GaAs: deposition and recovery



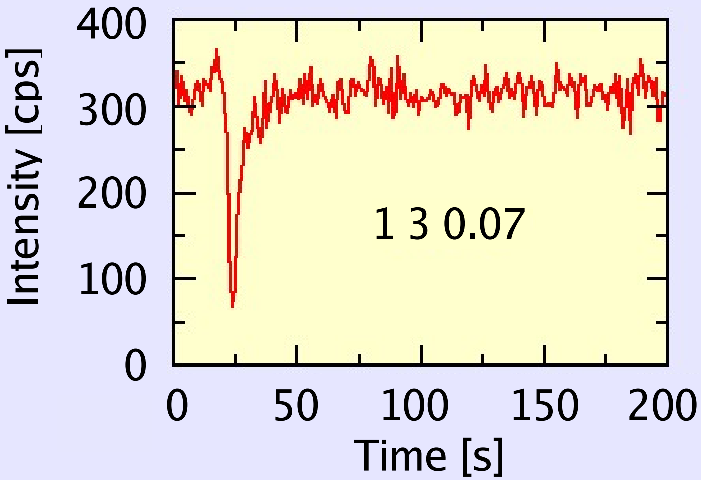
Integer-order reflection: surface morphology

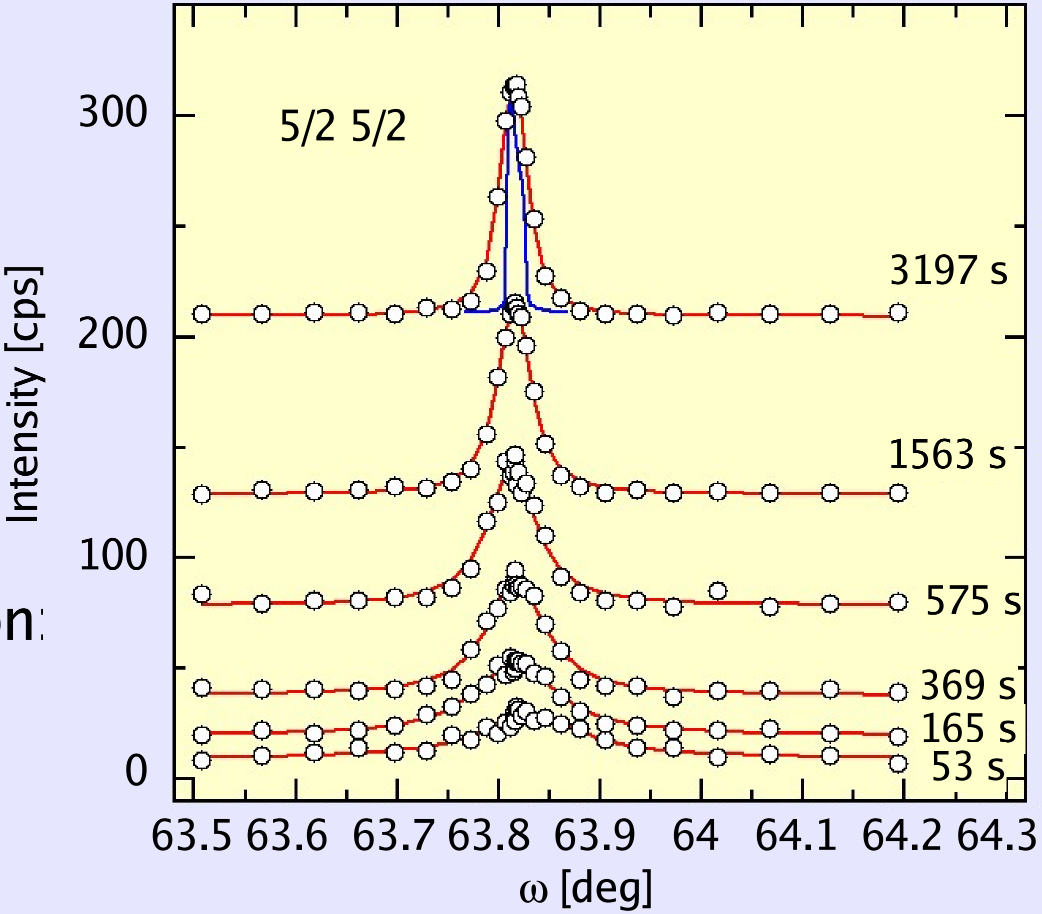
Fractional-order reflection: surface reconstruction

- deposition:
  - kinetics of layer-by-layer growth

- recovery:
  - kinetics of surface morphology
  - kinetics of reconstruction domains

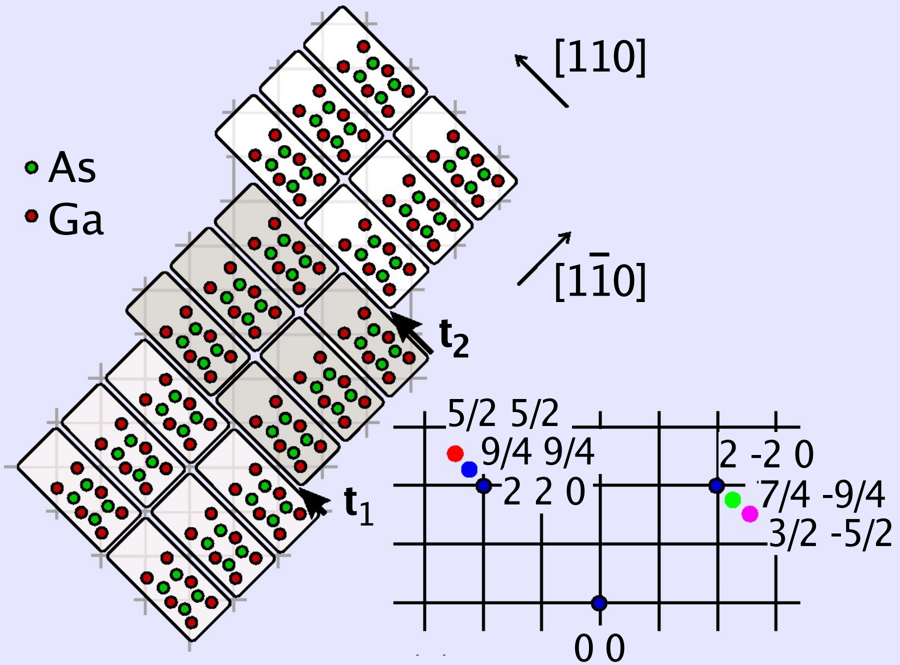
Reconstruction domain coarsening

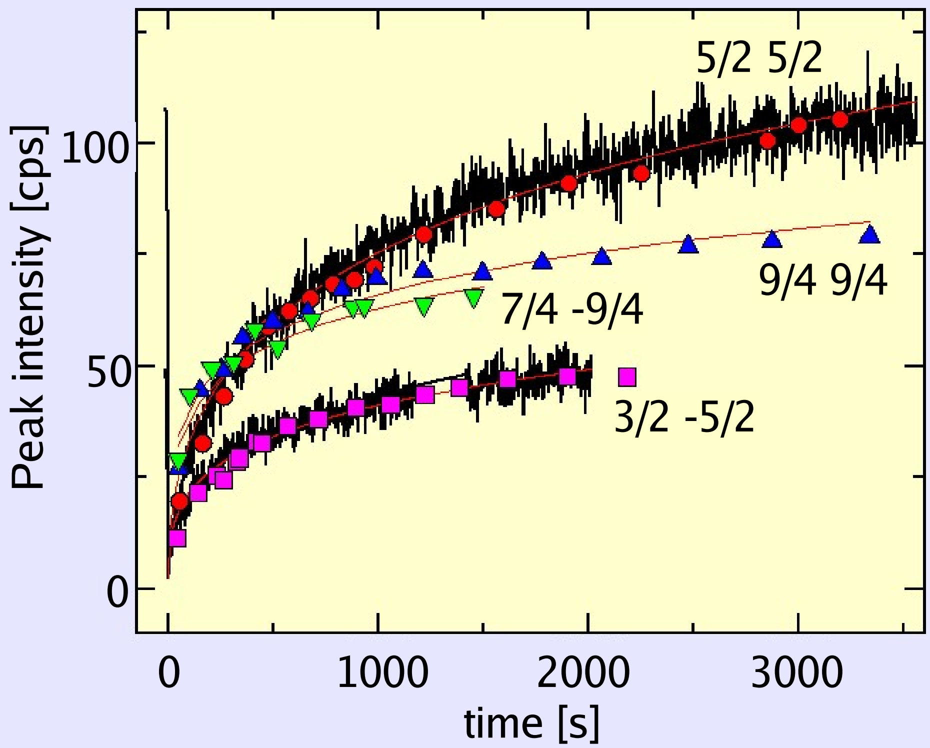




- Single monolayer deposition: unchanged morphology
- Independent nucleation of domains on 2D islands
- Monitor fractional-order profiles during recovery

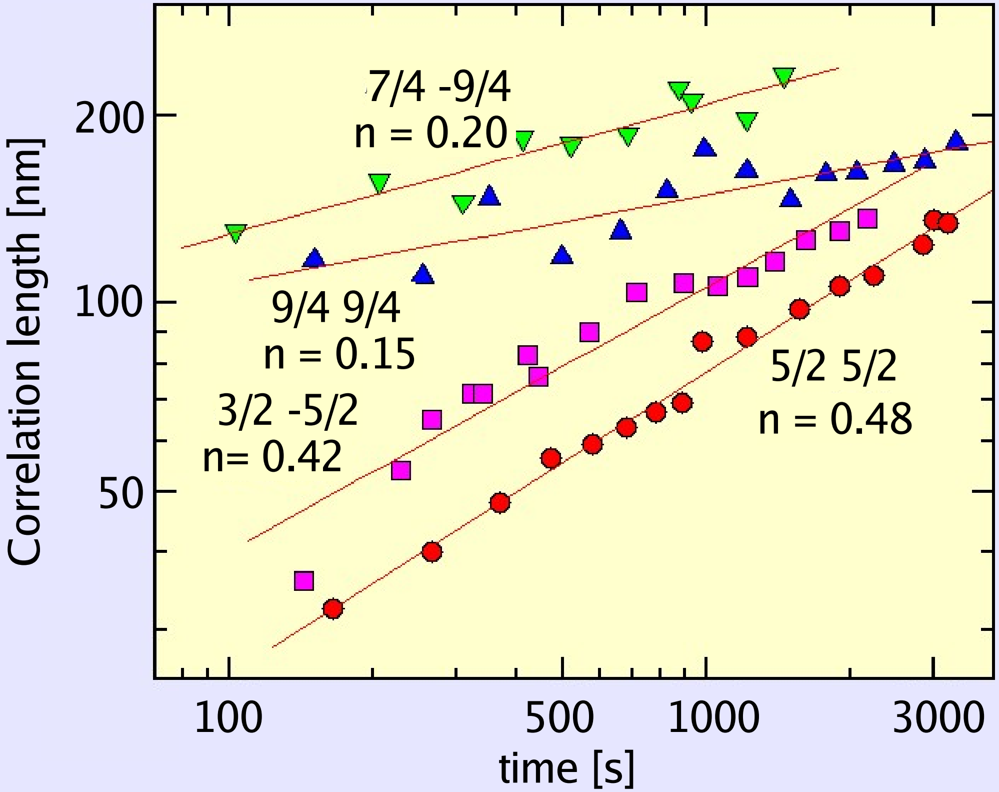
Reconstruction domain coarsening





- Compare two different azimuths
- Compare evolution of half-order and quarter-order reflections

GaAs: Nonequivalent Domain Boundaries



Correlation length increases as $l \sim t^{n}$

half-order reflections: $n = 0.42 \pm 0.05$

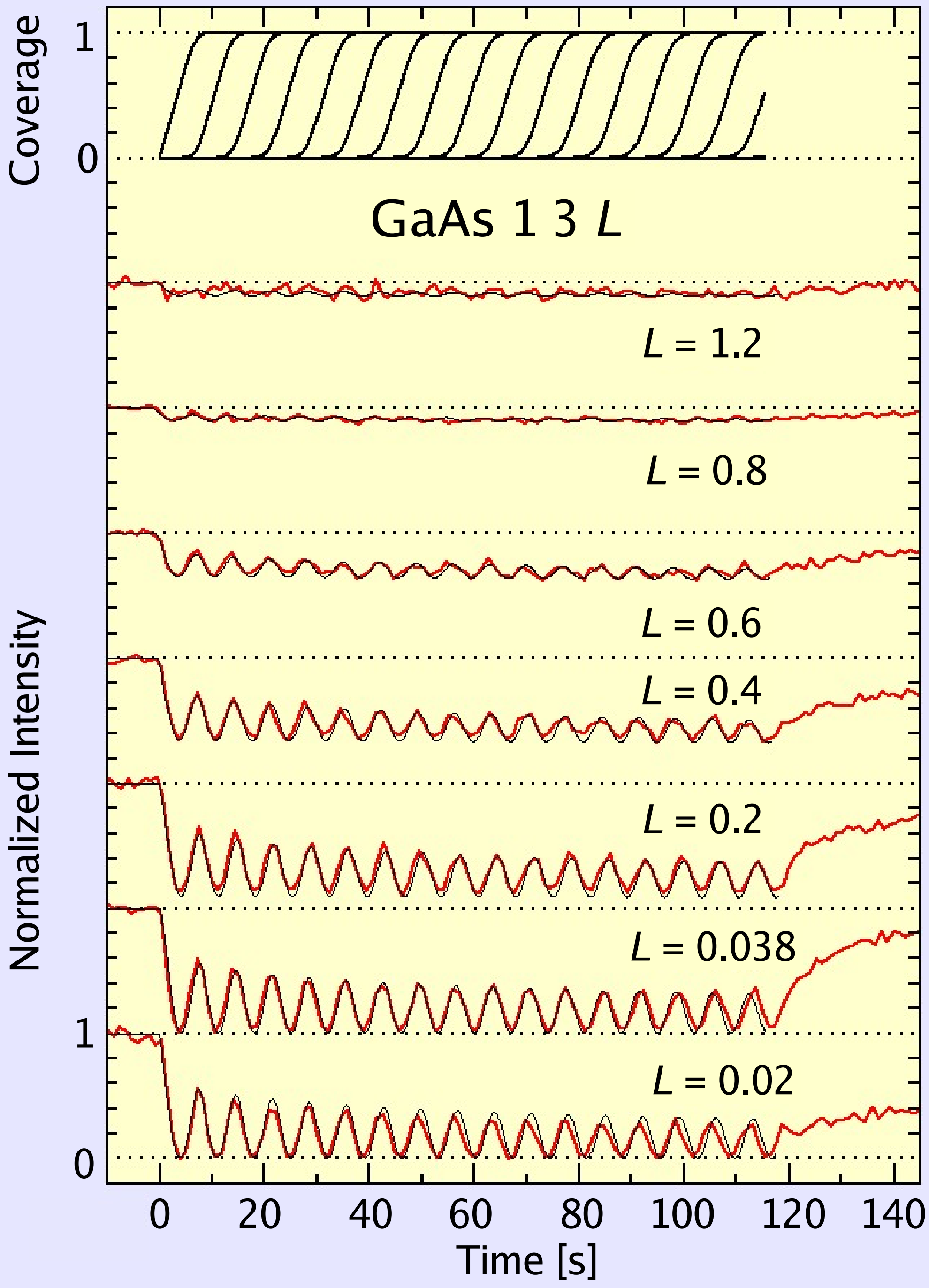
quarter-order reflections: $n = 0.22 \pm 0.05$

anisotropic reconstruction domains, aspect ratio ~1.5, elongated along [110]

Phys. Rev. Lett. **90** (2003) 016101

- single-kink and double-kink domain boundaries are energetically inequivalent
- The fraction of the higher energy double-kink domain boundaries increases logarithmically in time

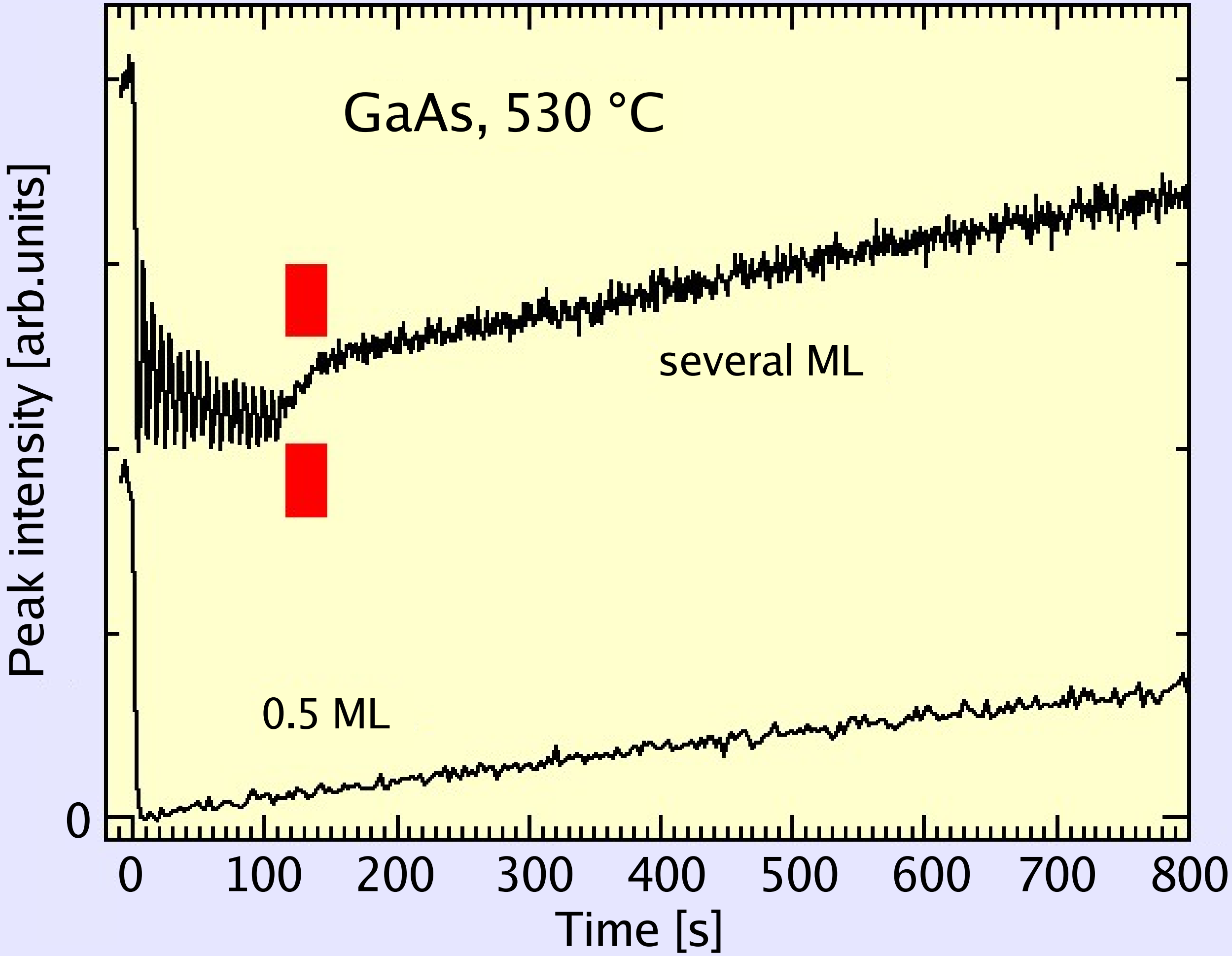
Layer-by-layer growth



- Simultaneous fit with same coverages for all *L*
- Intensity along crystal truncation rod does not change, therefore surface reconstruction does not change

Surf. Sci. **525** (2003) 126

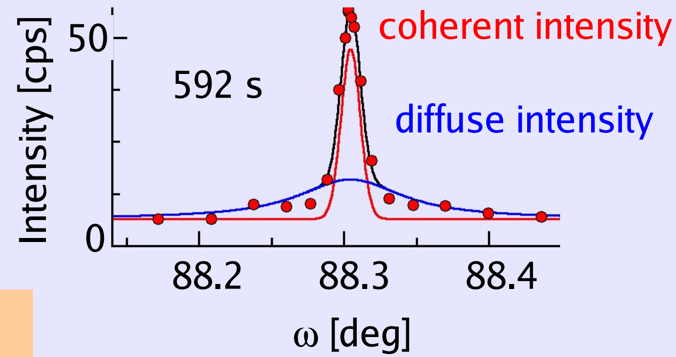
GaAs: two stages of recovery



- Multi-layer deposition: initial fast recovery, followed by slow recovery
- Below 1 ML: no fast recovery

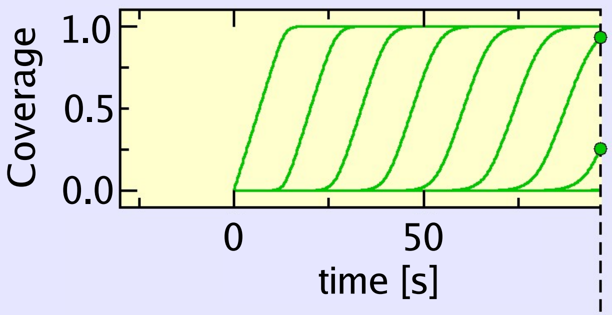
Analysis of a single growth run

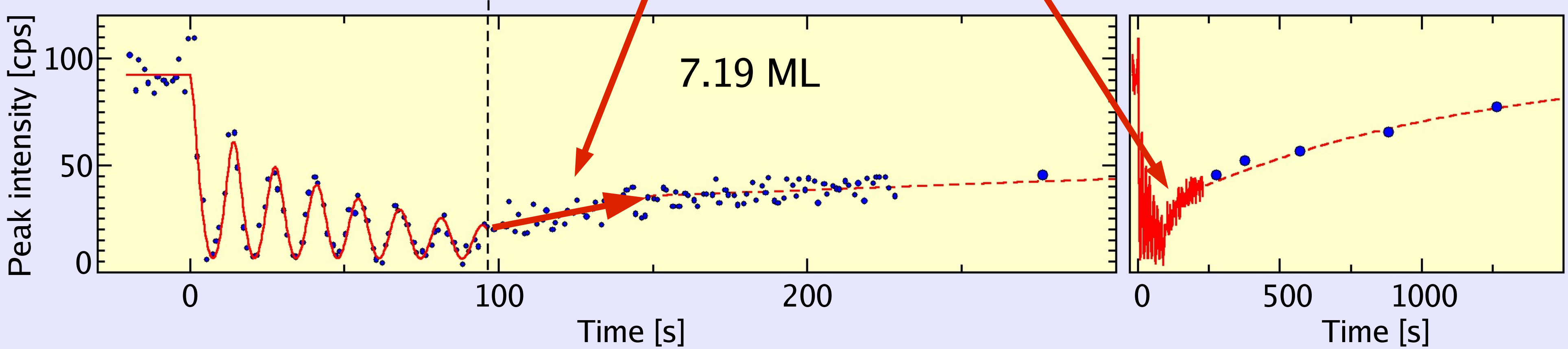
Deposition ends
at coverages:
7^{th} layer – 0.93 ML
8^{th} layer – 0.26 ML
total coverage: 7.19 ML



fast recovery is
the transformation
of a multi-level
surface to a
two-level surface

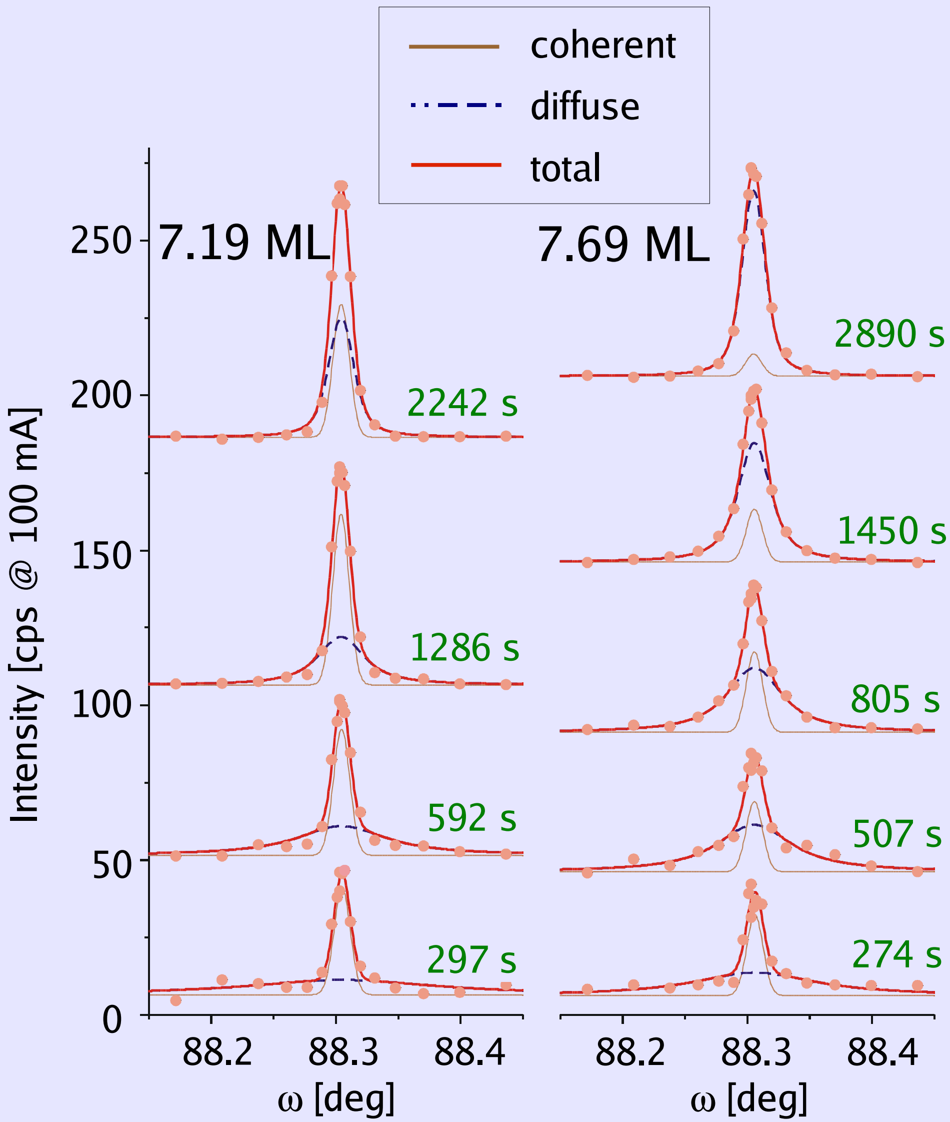
slow recovery starts
at coverage 0.22 ML

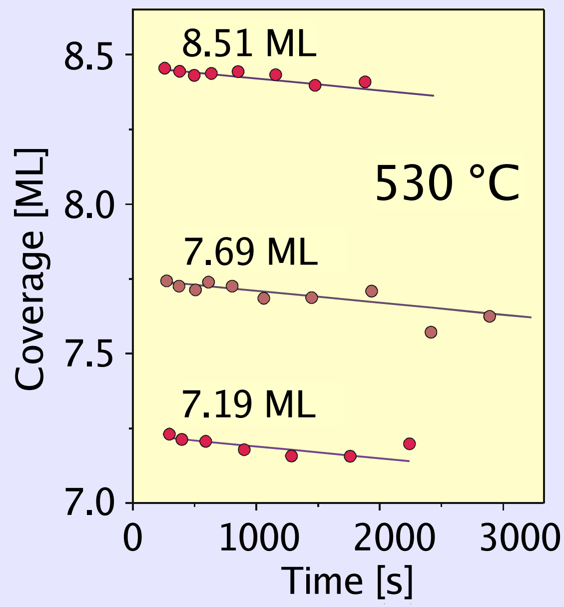




Surf. Sci. **560** (2004) 88

Coverages during recovery

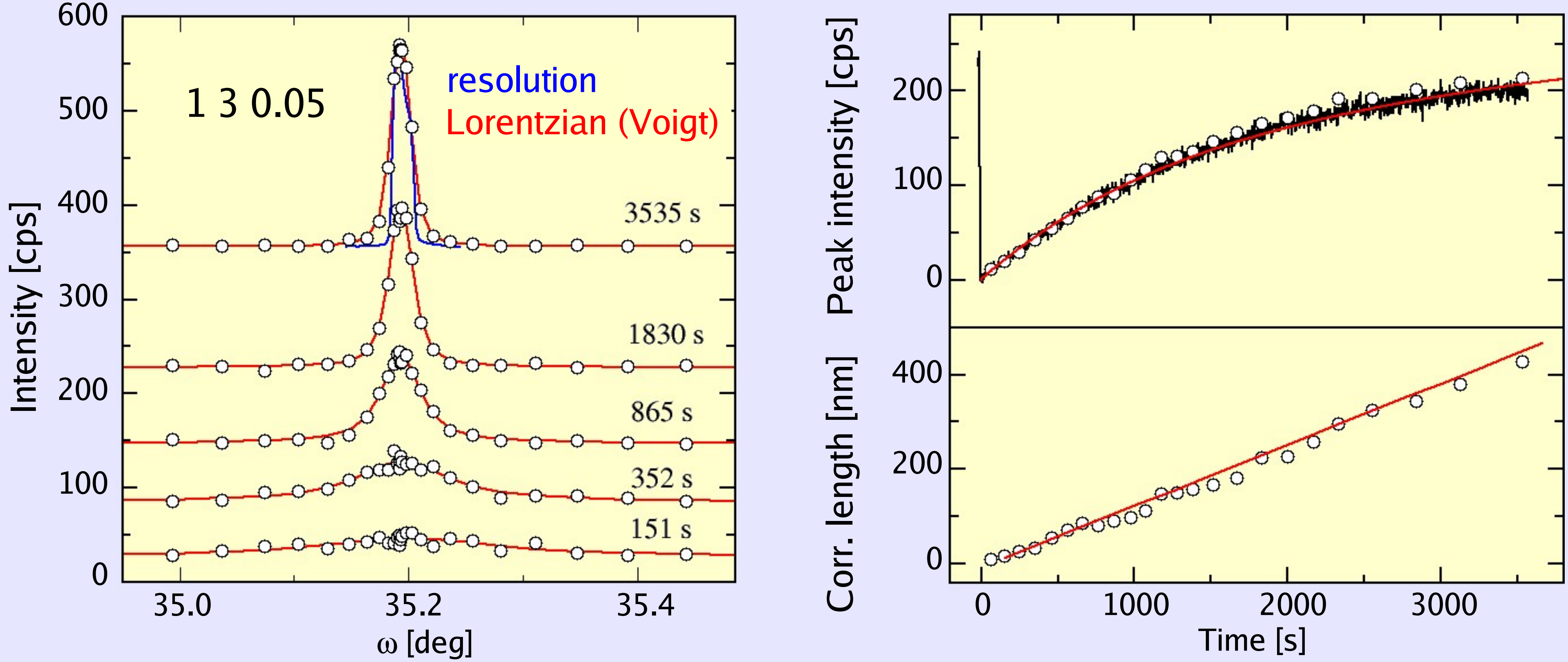




- analysis of coverage by separating coherent and diffuse intensities
- desorption as low as **1 ML in 7 hours** can be detected

GaAs: coarsening of 2D islands

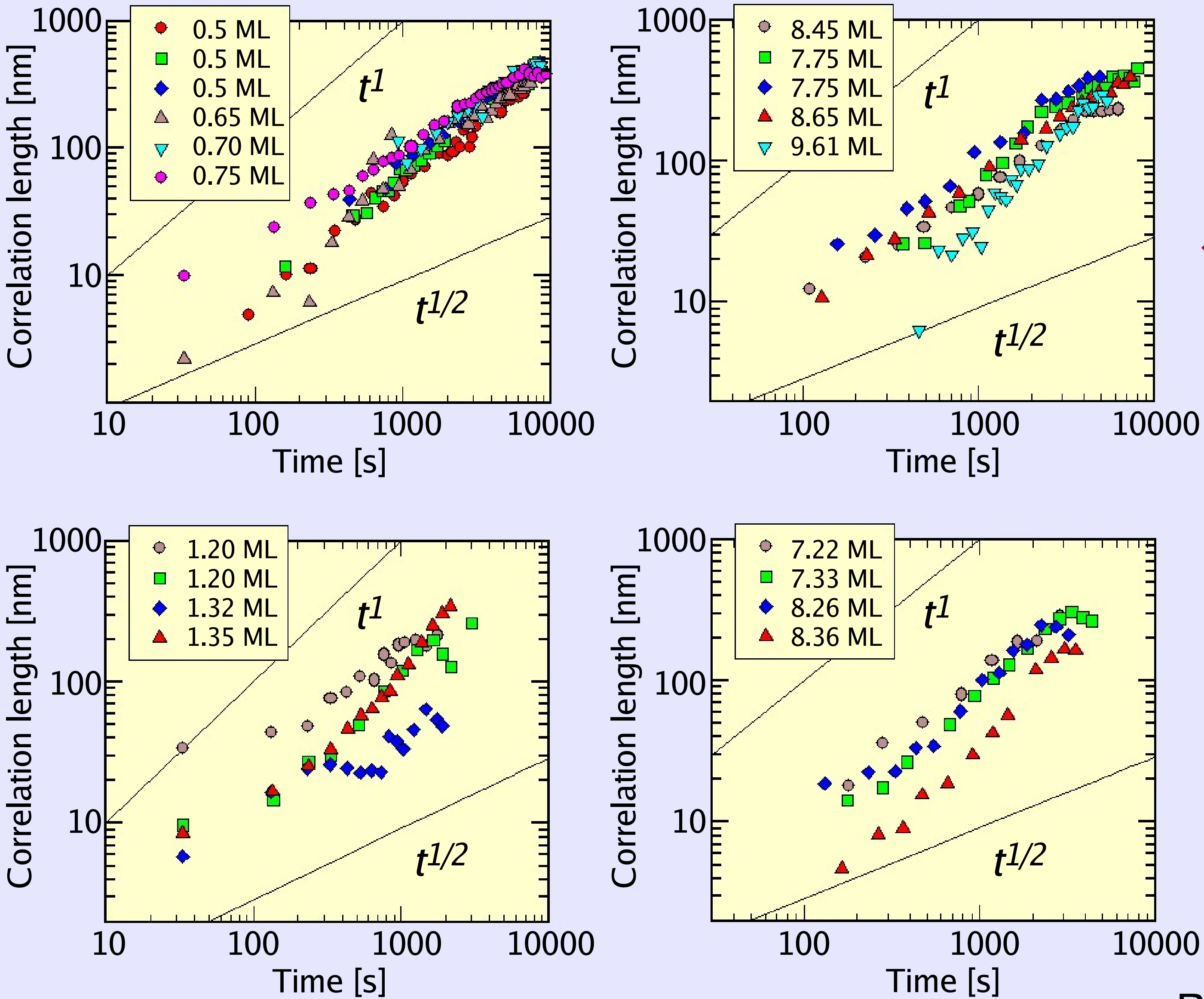
Coverage: 0.5 ML



- we expect: $l(t) \propto t^{1/3}$ (diffusion-limited, Lifshitz & Slezov, 1961)
  $l(t) \propto t^{1/2}$ (attachment-limited, Wagner, 1961)
- **we observe: $l(t) \propto t$**

Phys. Rev. B **69** (2004) 165405

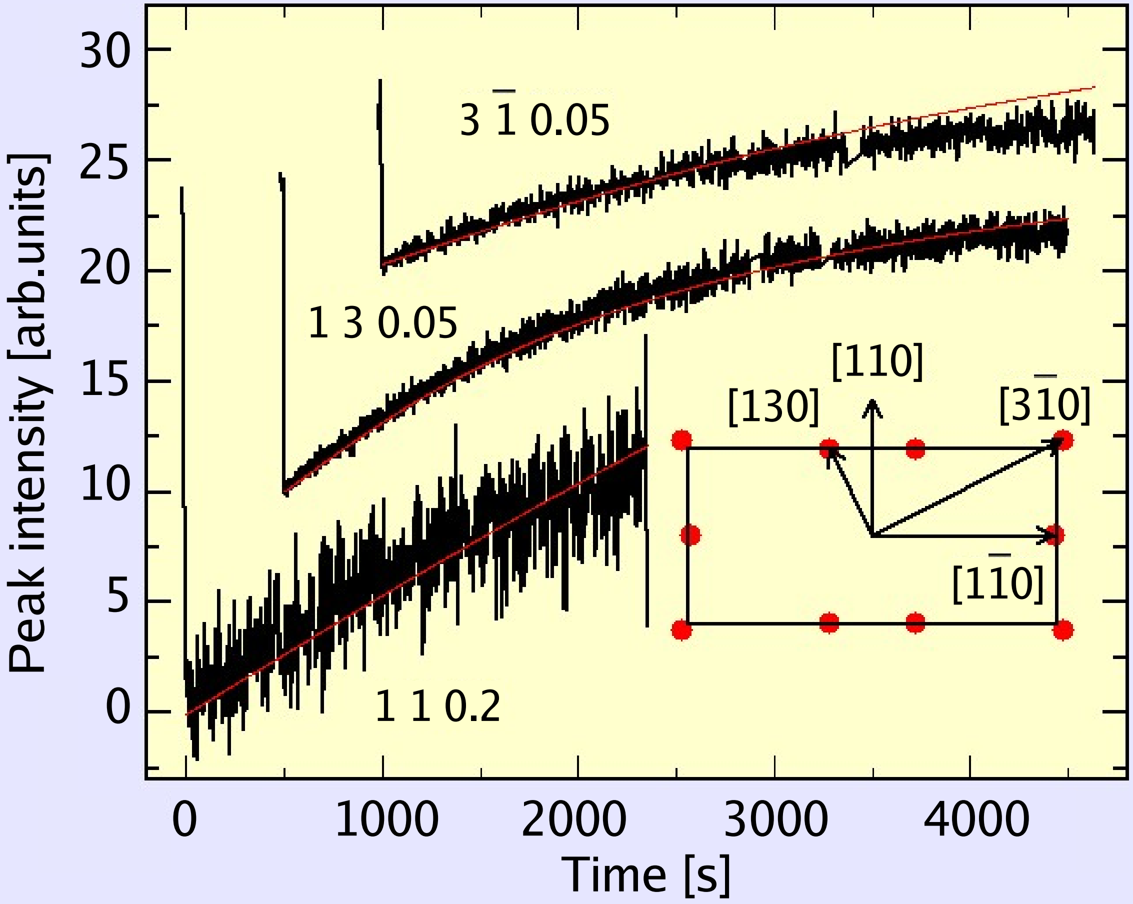
Kinetics of coarsening



- $l(t) \propto t^{n}$
  $n = 0.93 \pm 0.18$
- coarsening exponents do not depend on coverage

Physica B **357** (2005) 165

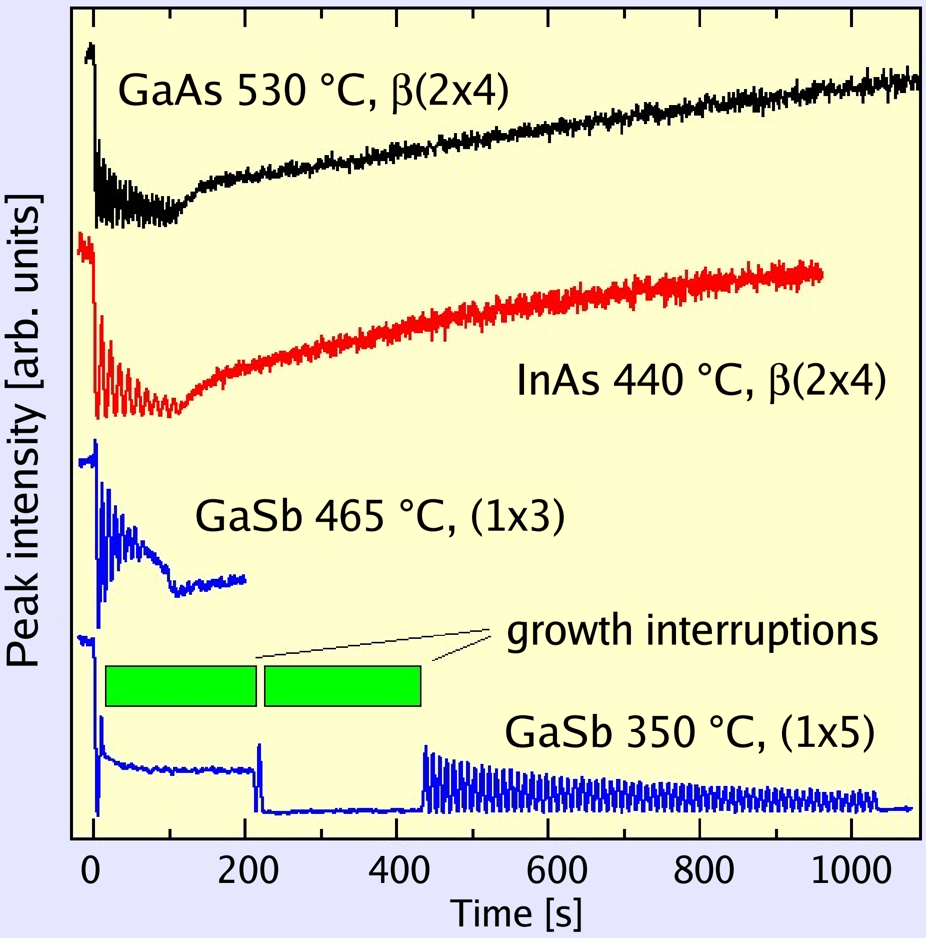
GaAs: self-similar coarsening



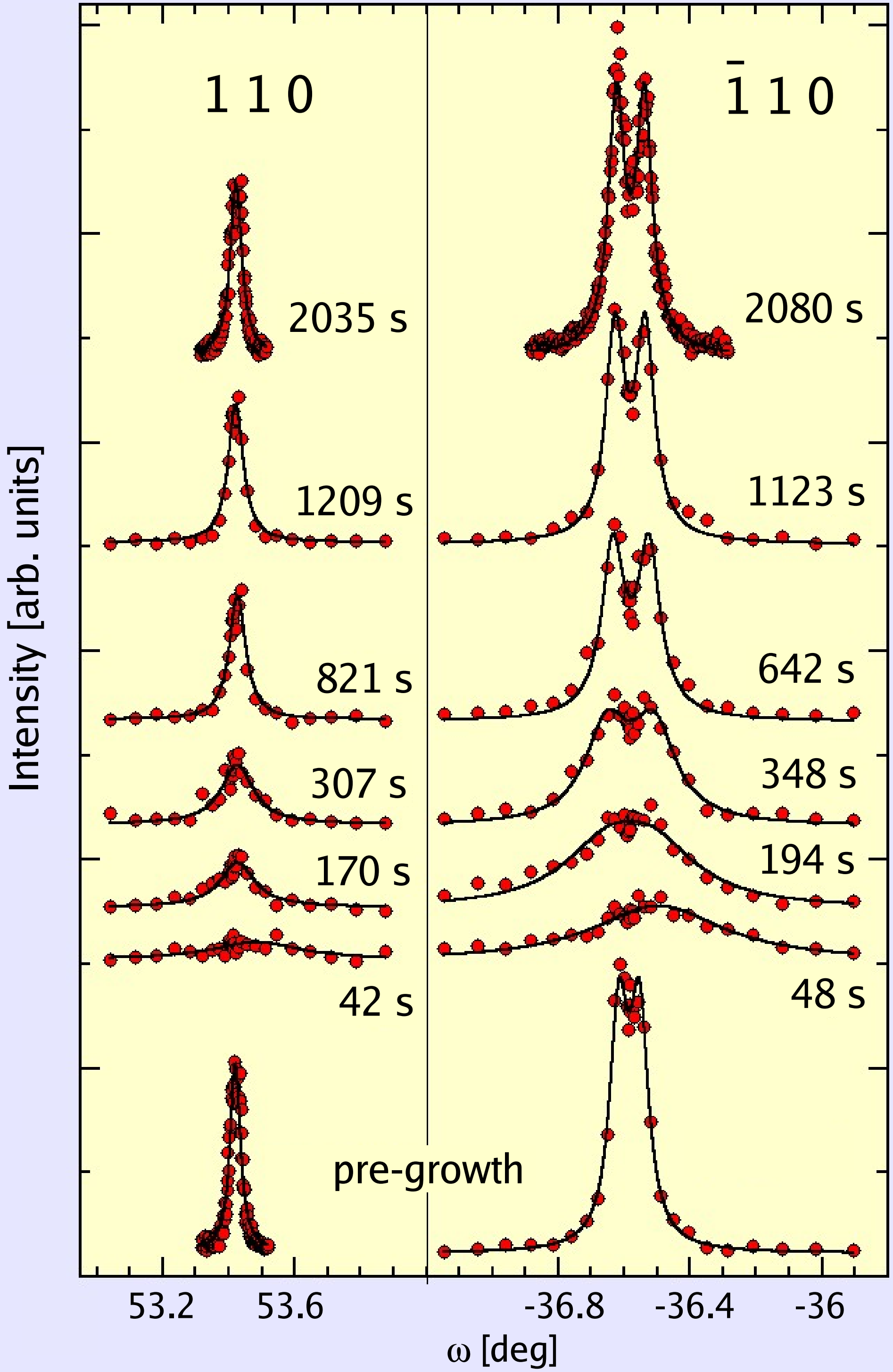
- identical time exponents and peak shapes in different azimuths along the surface: kinetic scaling
- different growth velocities
- Anisotropic surface correlations grow with self-similar shape

Phys. Rev. B **69** (2004) 165405

InAs and GaSb



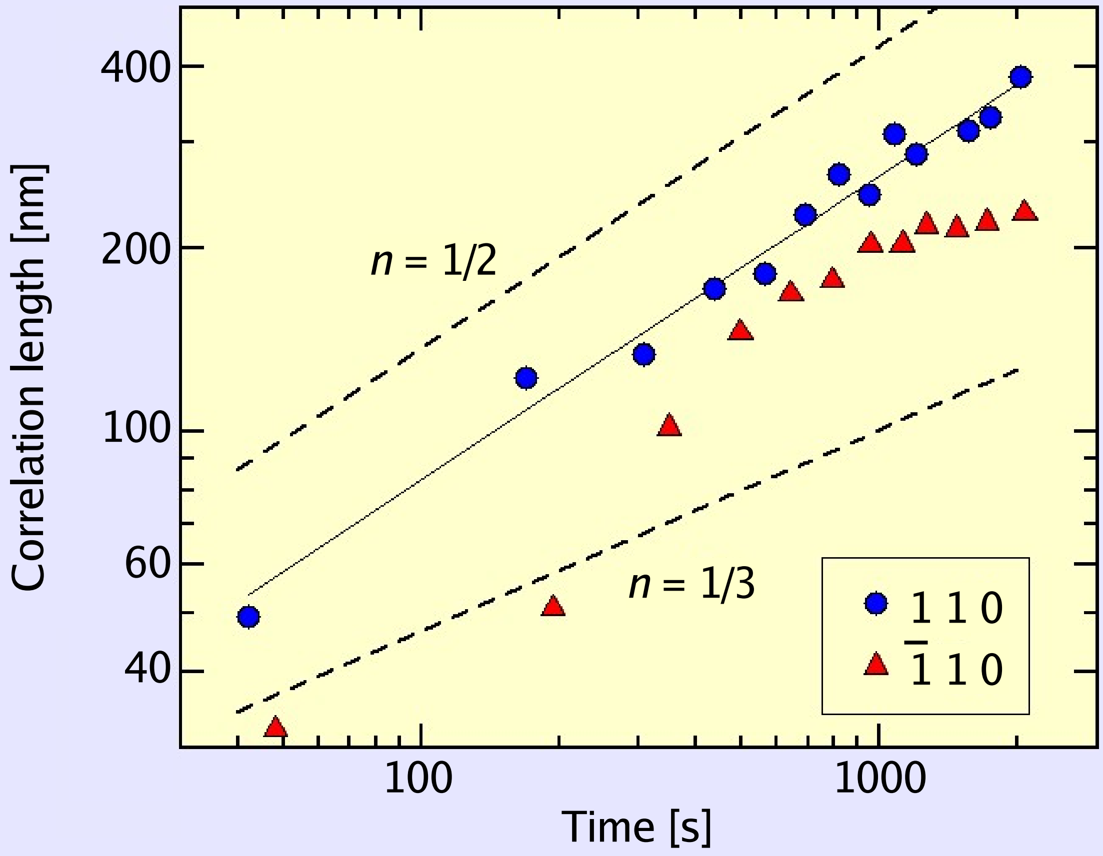
- similar deposition kinetics for GaAs and InAs
- GaSb strongly different
- low damping of growth oscillations: high surface mobility of adatoms
- GaSb growth unstable at high temperature
- recovery frozen for GaSb at low temperature



InAs (001)

- 0.5 ML coverage
- 1 1 0: narrow Lorentzian diffuse profile
- $\bar{1}$ 1 0: broader profile, split peak with constant separation: miscut producing 220 nm wide terraces

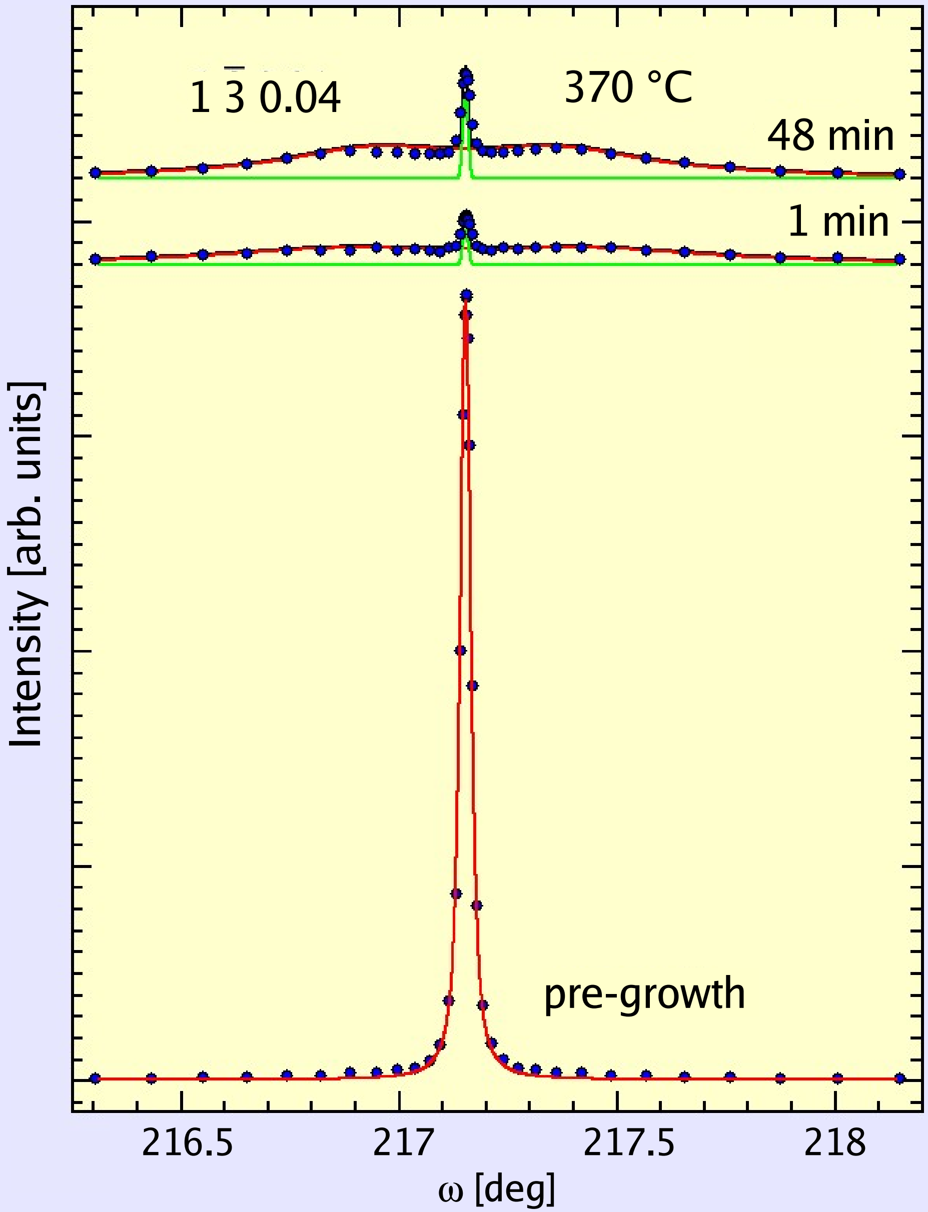
InAs (001): coarsening exponents



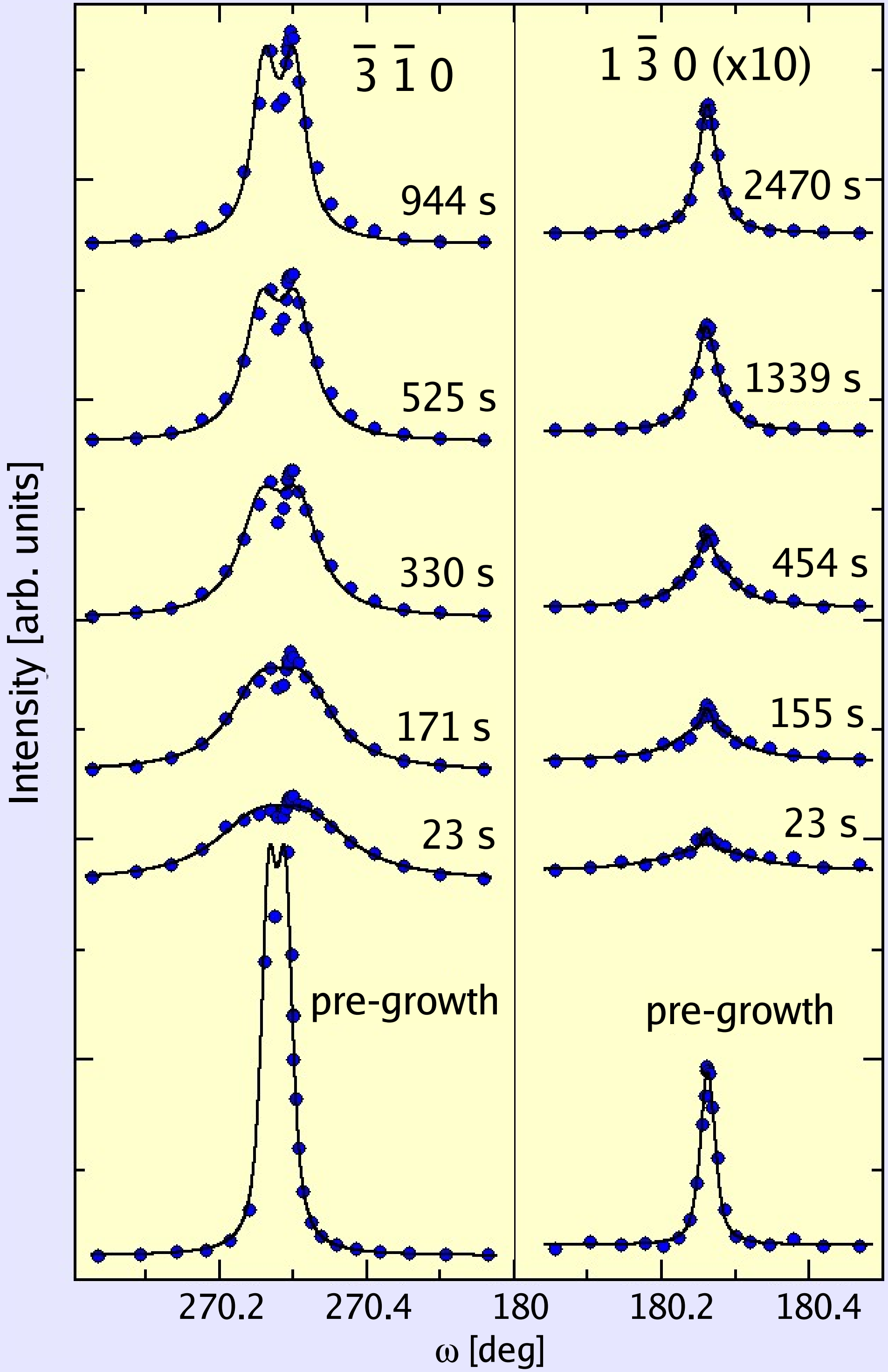
- parallel to steps: exponent $n = 0.5$
- fit with zero initial correlation length
- perpendicular to steps: saturation at 220 nm corres-ponding to miscut
- 1 1 0 and $\bar{1}$ 1 0 are independent

surface features elongated along [$\bar{1}$10] (perpendicular to diffraction vector)

GaSb (001): low temperature



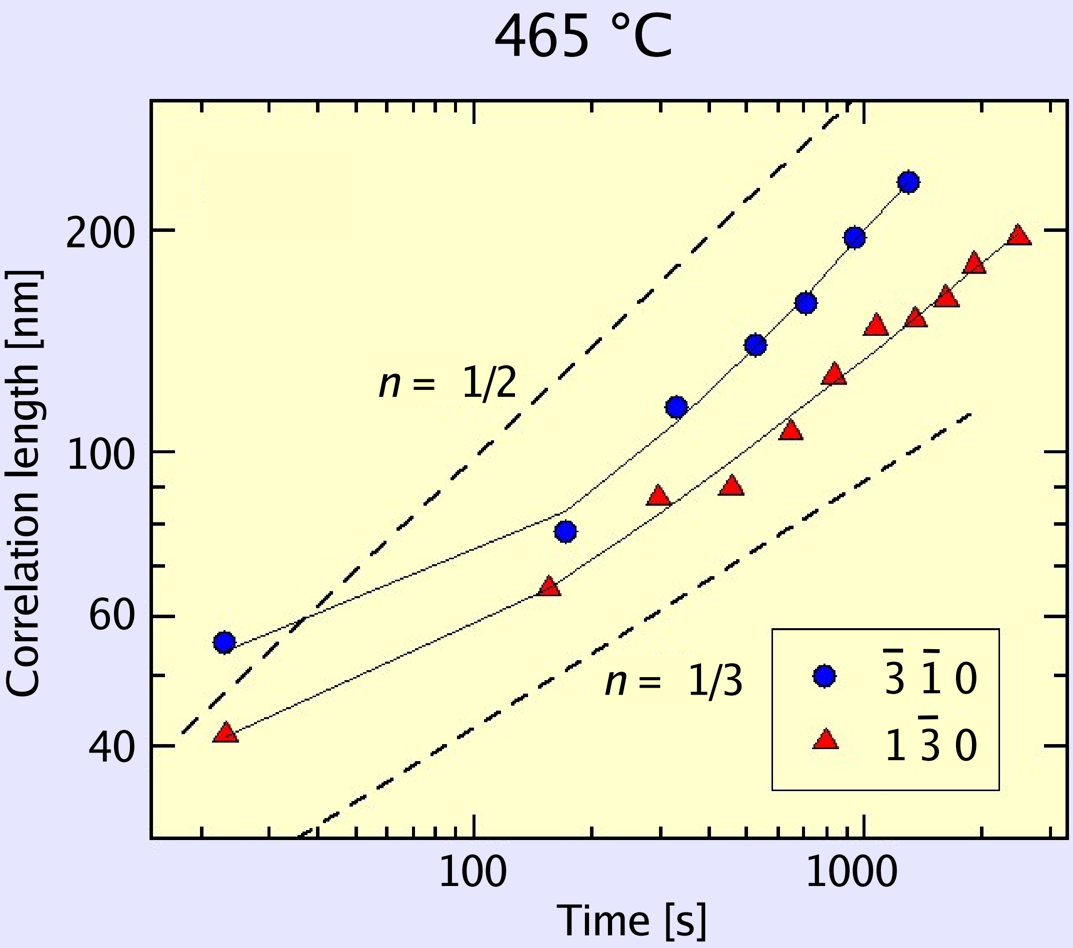
- 370 °C growth temperature
- Coverage 0.38 ML, non-vanishing coherent peak
- split diffuse component, presumably from nucleation
- extremely slow recovery
- BUT: low damping of growth oscillations: high surface mobility of ad-atoms



GaSb (001): 465 °C

- 0.5 ML coverage
- GaSb {1 1 0} reflections too weak, using {1 3 0} instead
- $1\,\bar{3}\,0$ unsplit, Lorentzian peak profiles: scaling
- different from low-temperature behavior
- $\bar{3}\,\bar{1}\,0$ split, peak separation *not* constant

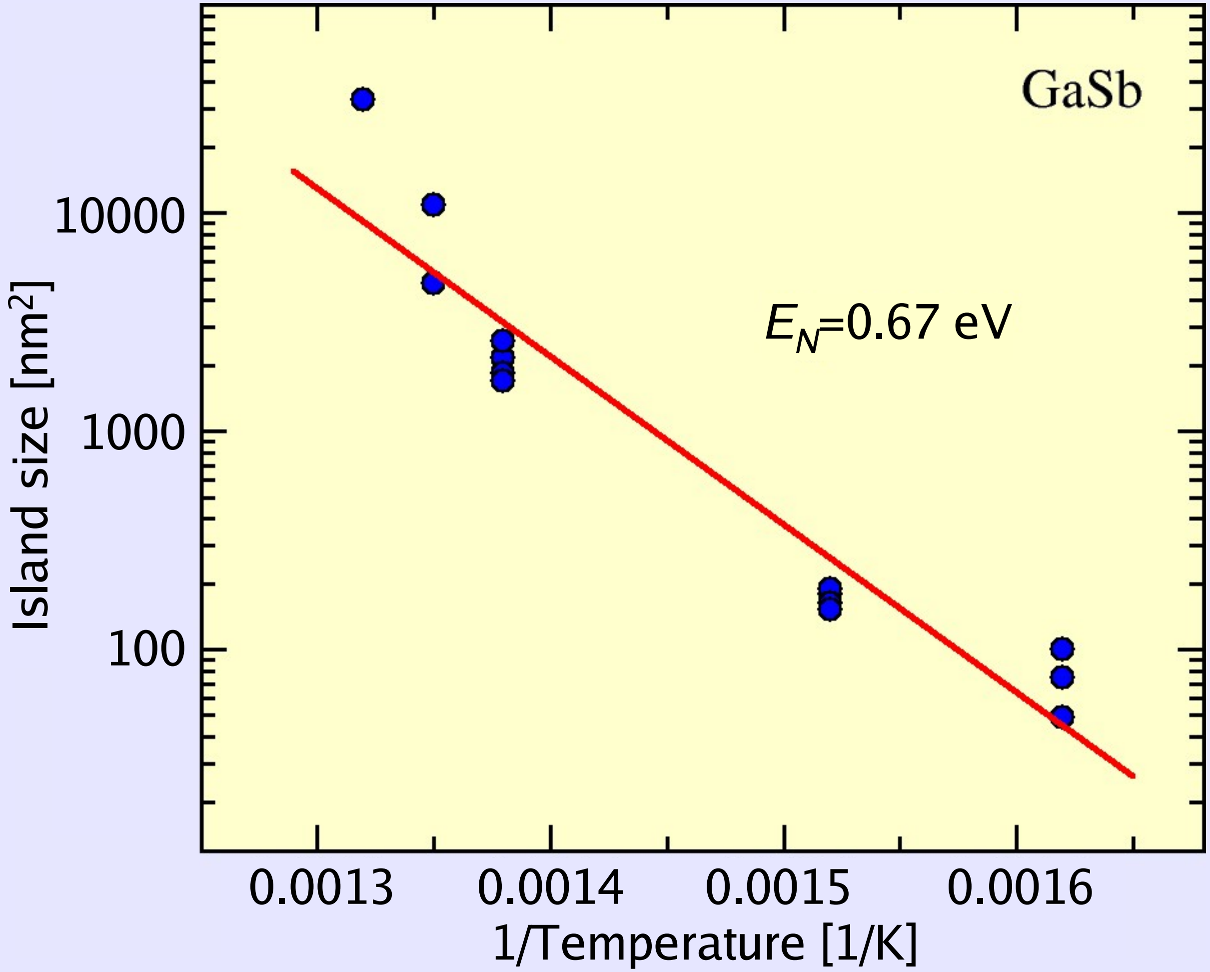
GaSb (001): coarsening exponents



- $\bar{3}\,\bar{1}\,0$ (close to 1 1 0): time exponent $n = 0.68$ with 50 nm initial size
- $1\,\bar{3}\,0$ (close to $1\,\bar{1}\,0$): time exponent $n = 0.44$ with 35 nm initial size
- split peak not saturat- ing: growth-induced (miscut 430 nm)
- Initially different island size distribution from nucleation

Surface features elongated along $[1\bar{1}0]$, anisotropy increasing in time

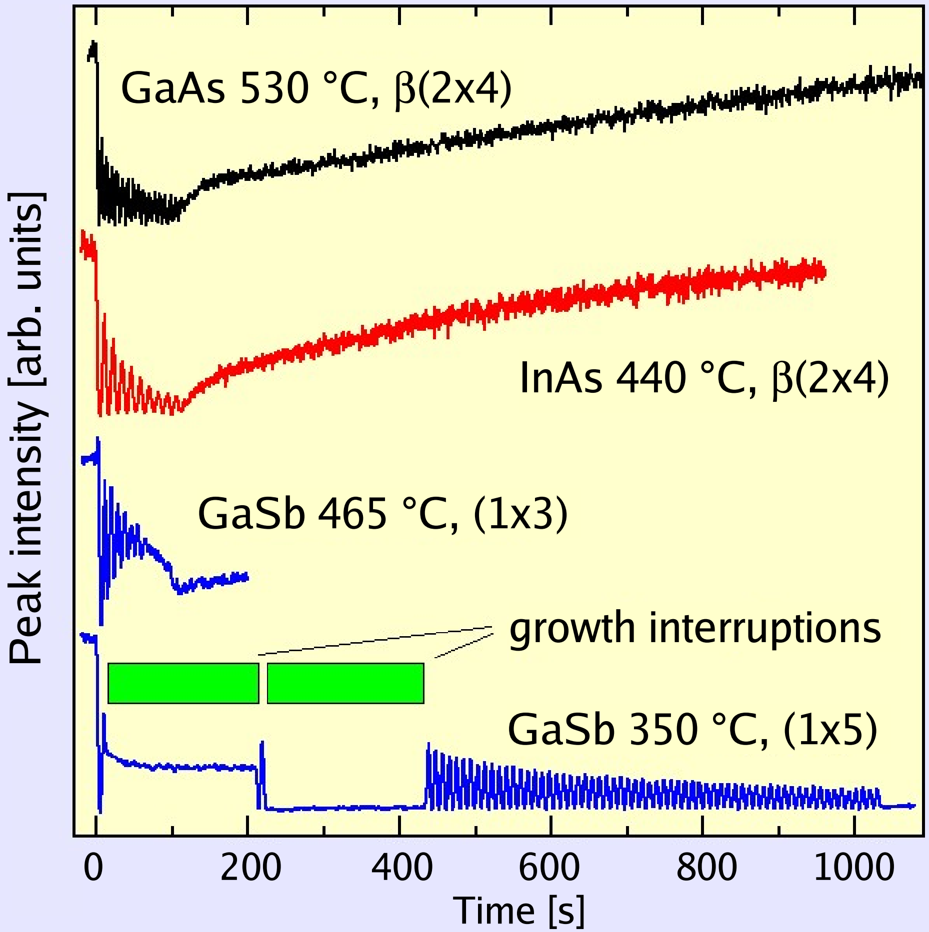
GaSb (001): nucleation

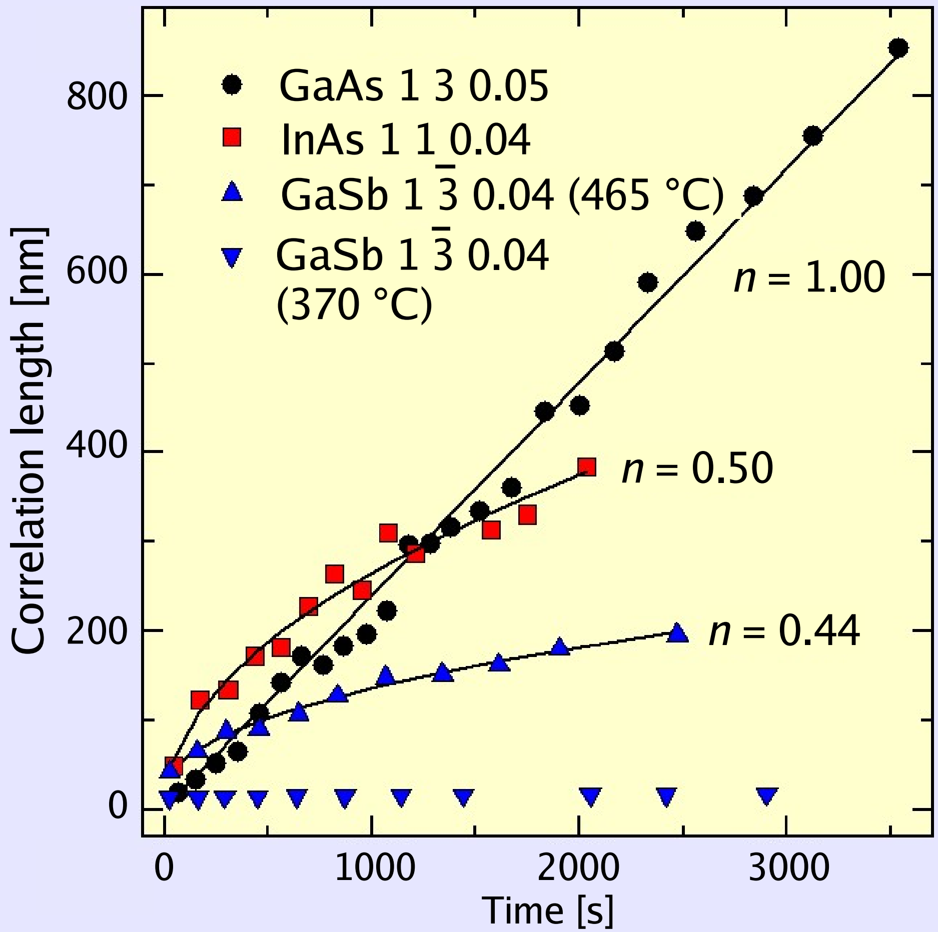


- Island sizes at the end of deposition
- Again shows high mobility of adatoms during deposition
- Wide temperature range, independent of surface reconstruction
- $E_D=3E_N=2.0\pm0.2$ eV*

*Phys. Rev. Lett. **69** (1992) 985

InAs and GaSb

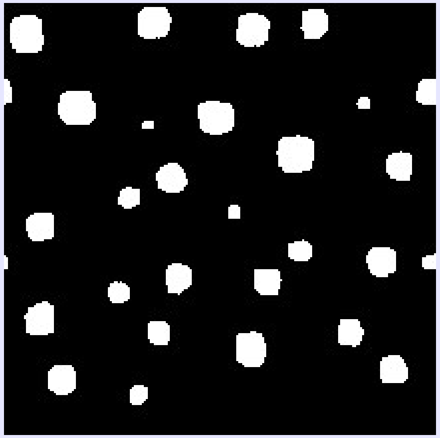


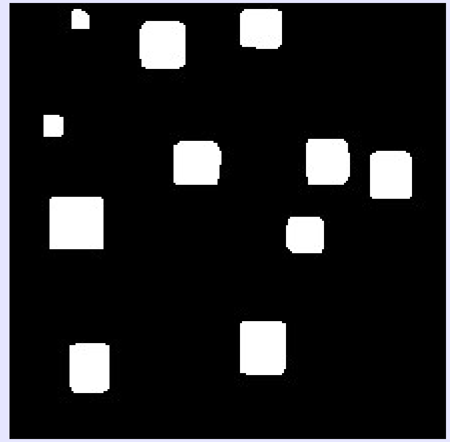


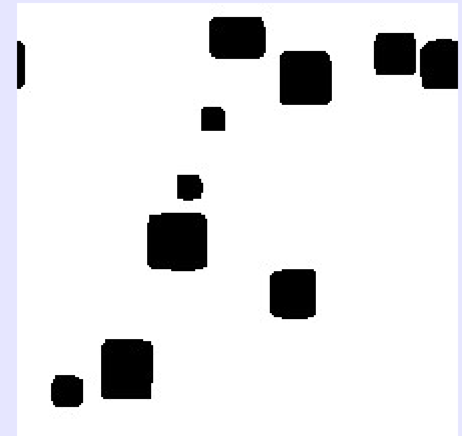
- *similar* deposition kinetics for GaAs and InAs, GaSb special features
- strongly *different* recovery kinetics

Z. Krist. **220** (2005) 225;
J. Cryst. Growth **278** (2005) 449

Generic Monte Carlo simulations



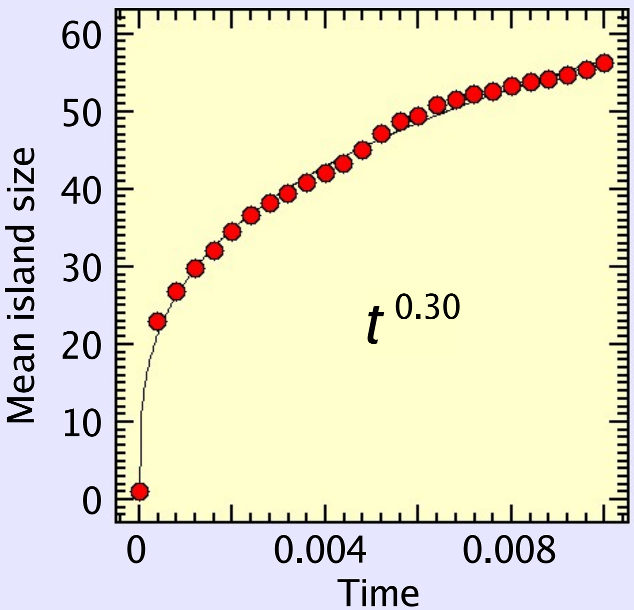


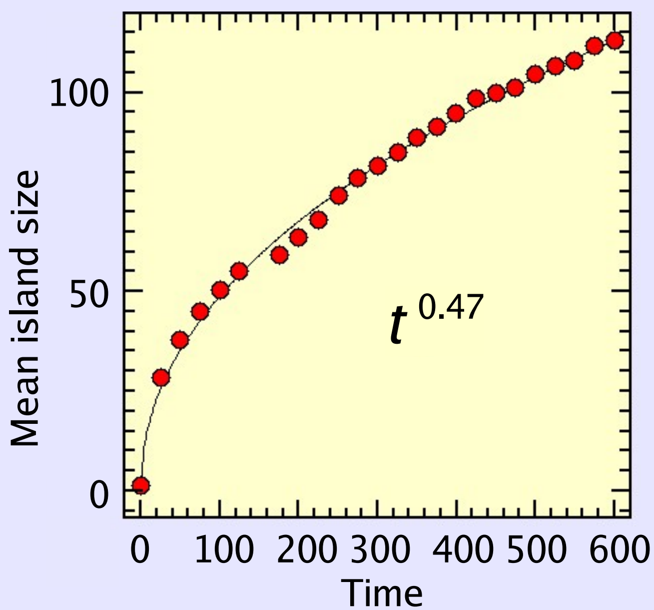


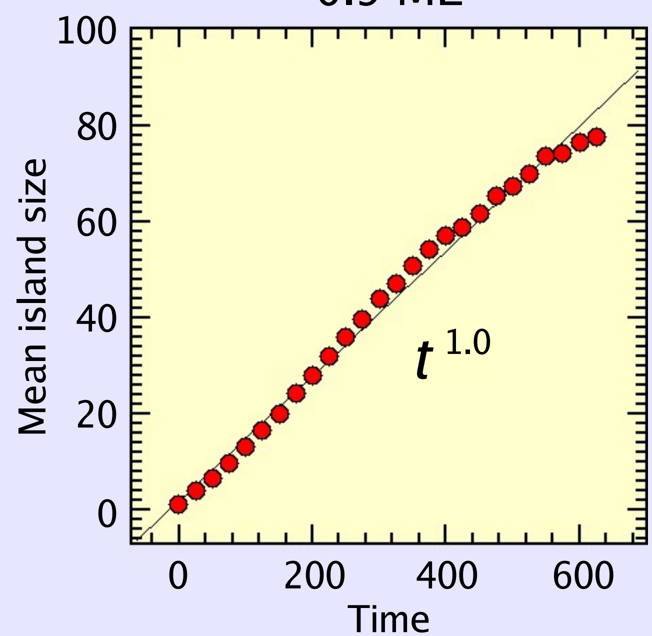
bond energy: 0.4 eV — 0.8 eV — 0.8 eV

T = 800 K — islands — pits

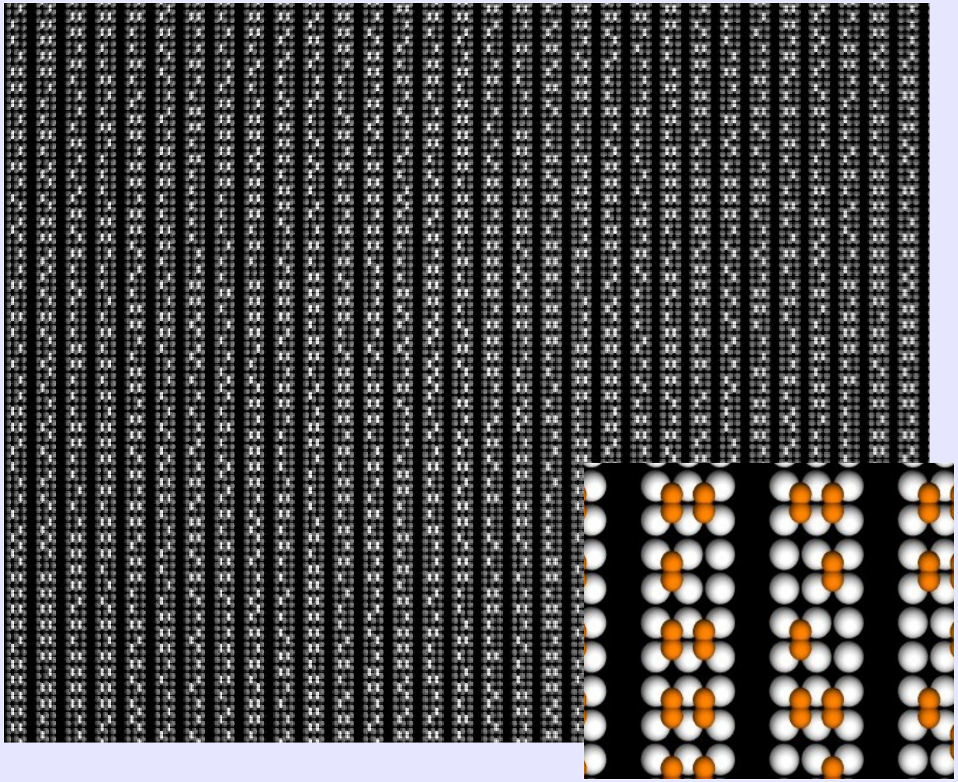
coverage: 0.1 ML — 0.1 ML — 0.9 ML

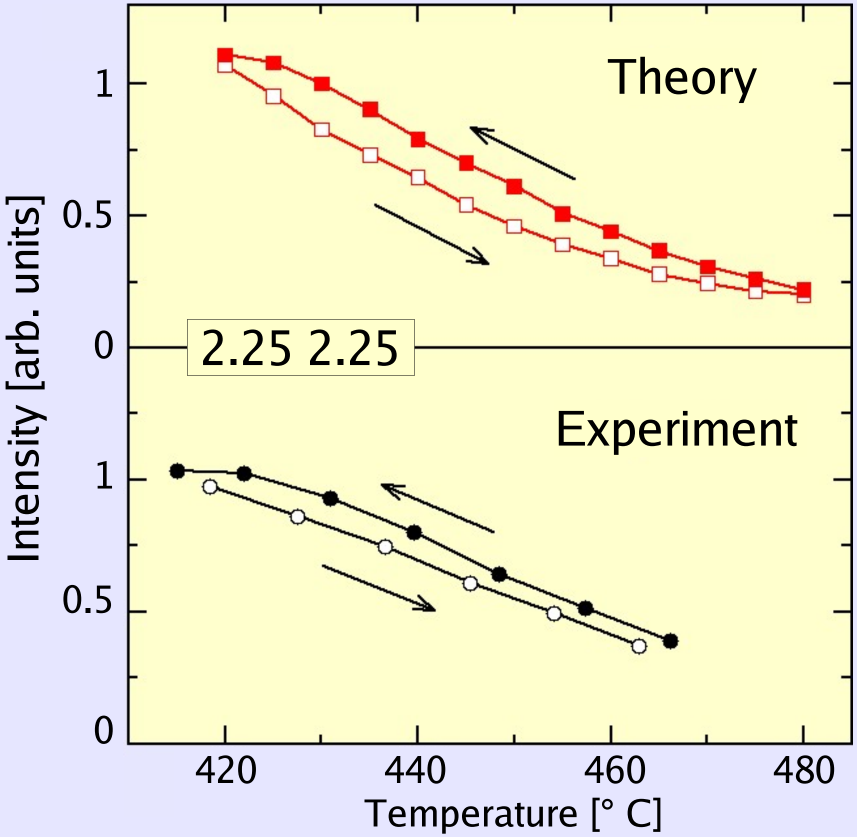






InAs: simulation and experiment





- InAs(001) α 2(2x4)–β 2(2x4) surface reconstruction transition
- comparison of 2.25 2.25 reflection calculated by first principle DFT-Monte Carlo simulations with experimental intensities

Collaboration with Frank Große, Humboldt University Berlin

Conclusions

- *Quantitative* analysis of growth kinetics on III-V semi-conductor surfaces
- Sufficient intensity to study kinetics of 2D systems with time resolution down to 1 s
- Two-stage coarsening: reduction to one incomplete level, lateral coarsening of 2D islands
- *Linear* coarsening for GaAs contradicts Ostwald ripen-ing mechanism
- Strongly different coarsening behavior for GaAs, InAs and GaSb, inhibited coarsening on GaSb

Acknowledgments

Vladimir M. Kaganer

Bernd Jenichen

Dillip K. Satapathy

Brad P. Tinkham

Xiangxin Guo

Masamitu Takahasi

Steffen Behnke

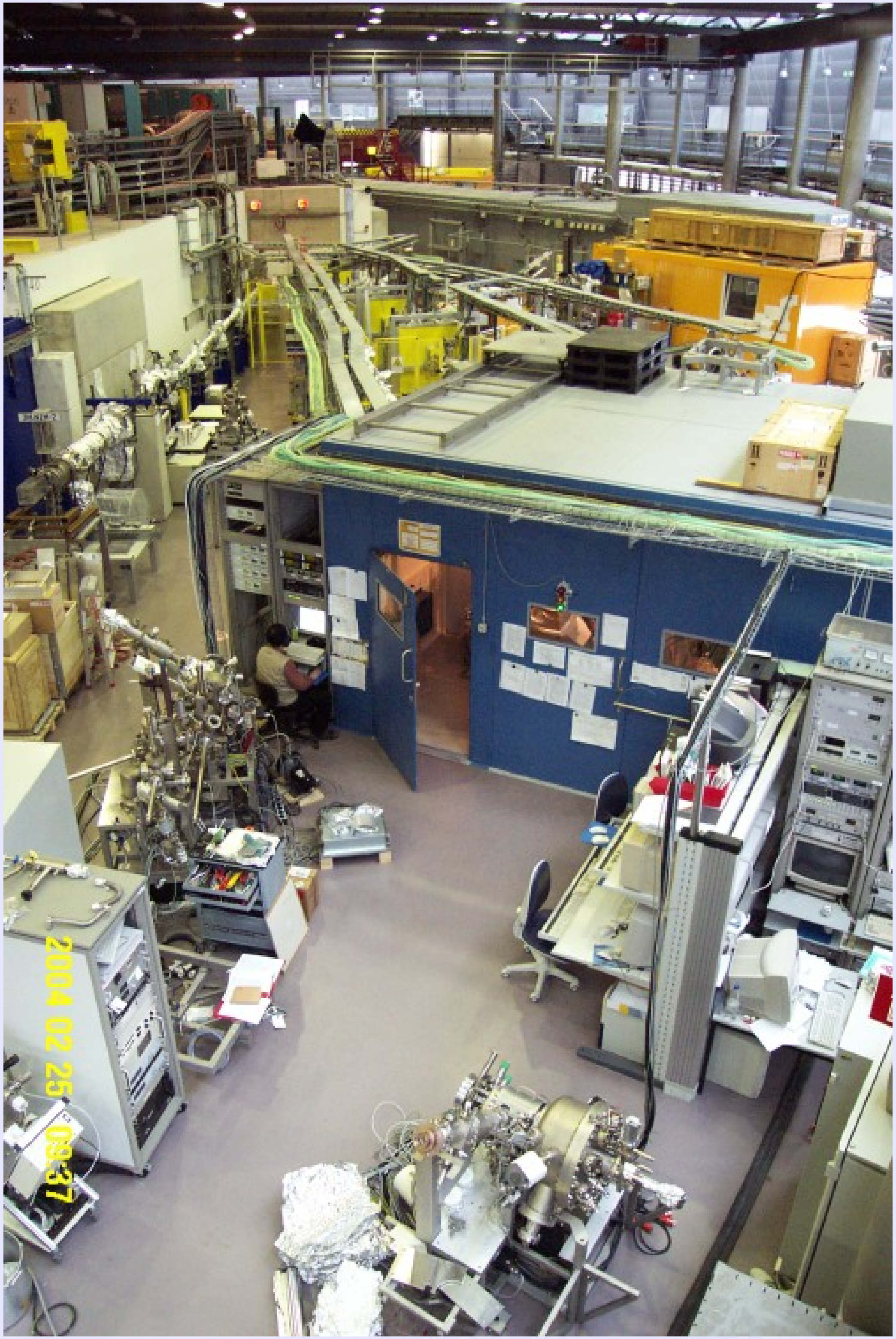
Klaus H. Ploog

Frank Große,
Humboldt University

BESSY, Berlin

The PHARAO beamline

- undulator beamline at BESSY, radiation safety hutch for experiment
- energy range 6-12 keV
- six-circle diffractometer for grazing incidence diffraction experiments
- three fully featured MBE systems for the growth of III-V compounds, metals and oxides
- liquid nitrogen cooling, up to 7 effusion cells per growth chamber



Diffractometer / MBE