Compositional Mapping of Semiconductor Quantum Dots by X-ray Microscopy

Stefan Heun

Laboratorio TASC INFM-CNR

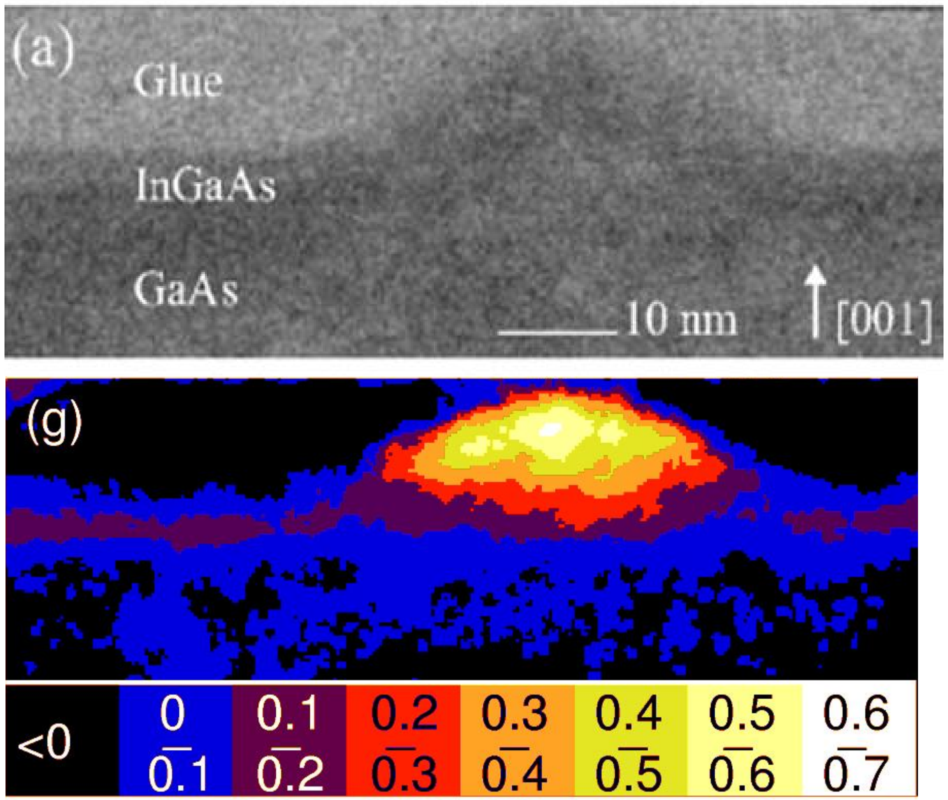
34012 Trieste, Italy

Motivation

- Quantum Dot Applications based on their particular electronic properties (confinement)
- Strain-driven self-assembly (SK-growth)
- Model systems: InAs/GaAs, Ge/Si
- Intermixing and alloying allow for partial strain relaxation
- Control of composition of individual QD, which determines their physical properties

Concentration Profiles

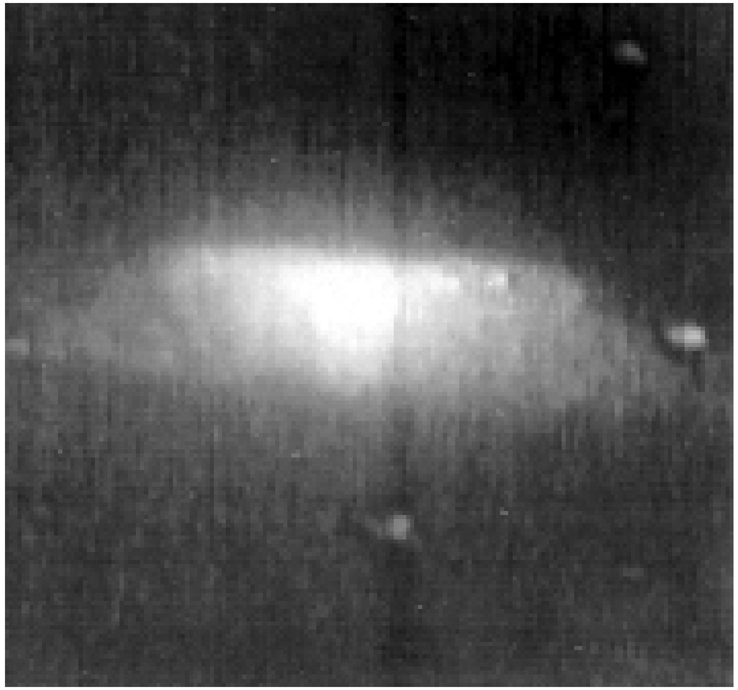
- Concentration maps
  - in cross-section: TEM



Walther et al.,
PRL **86** (2001) 2381

Concentration Profiles

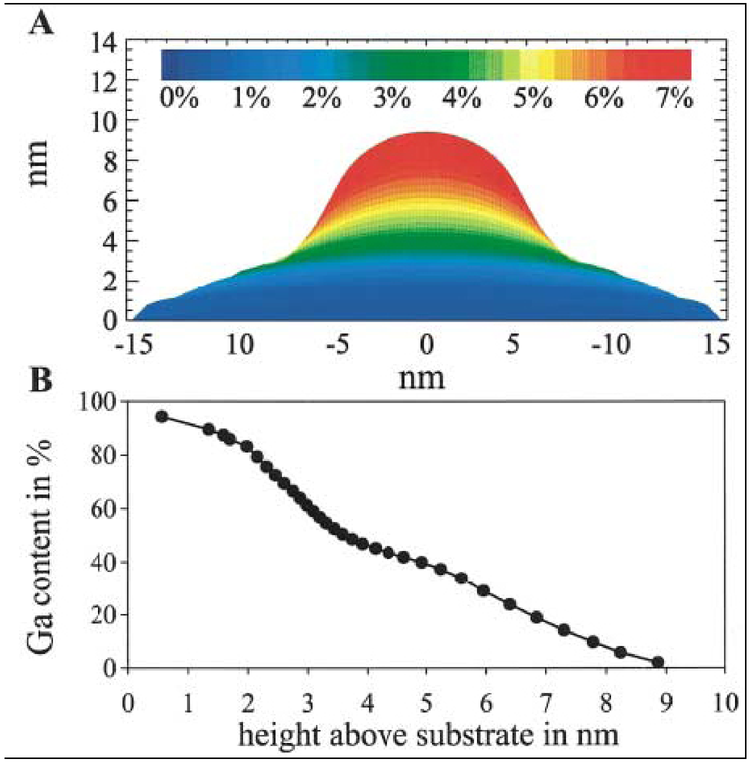
- Concentration maps
  - in cross-section: TEM, STM



Liu et al., PRL **84** (2000) 334

Concentration Profiles

- Concentration maps
  - in cross-section: TEM, STM, XRD

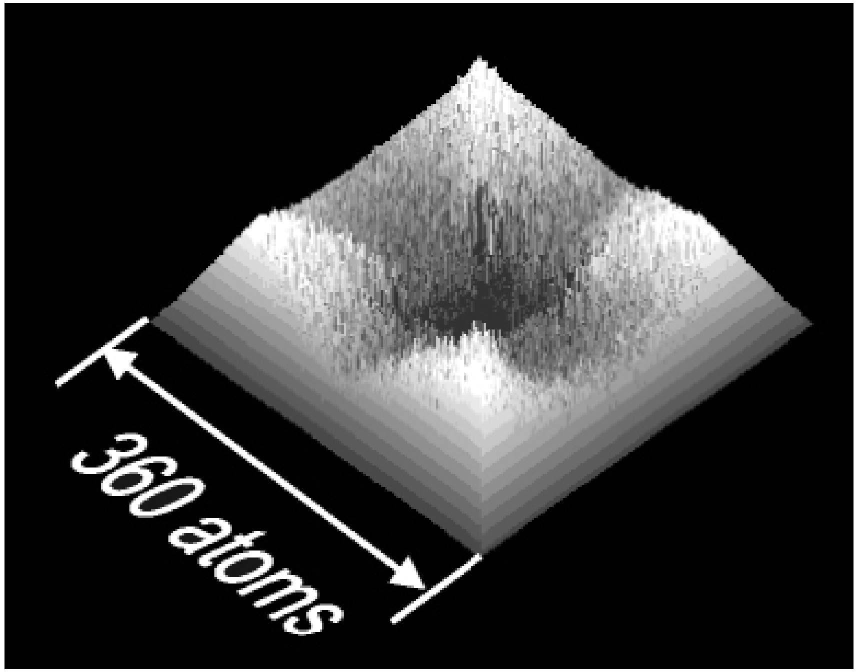


Kegel et al., PRL **85** (2000) 1694

Concentration Profiles

- Concentration maps
  - in cross-section: TEM, STM, XRD
  - in top-view: etching (Ge > 65%)

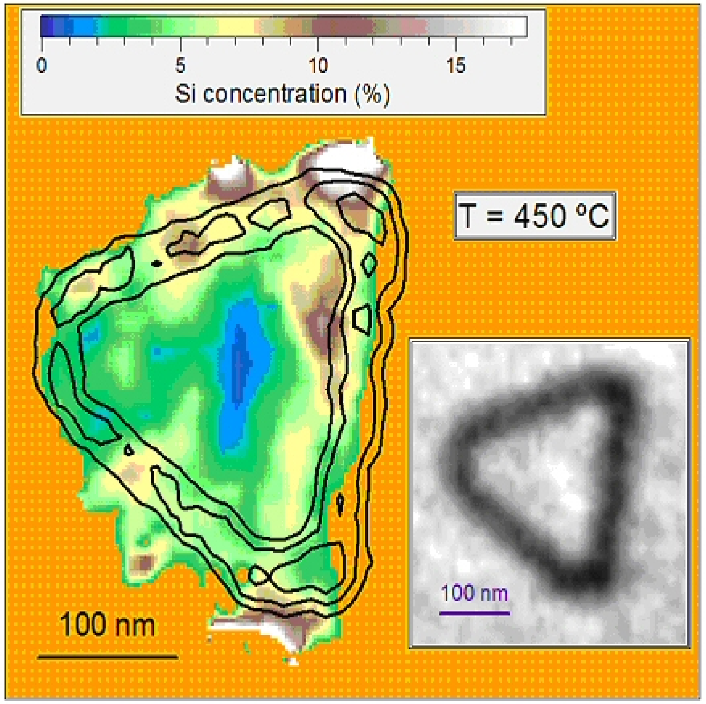
Complementary views
Full 3D mapping



Denker et al., PRL **90** (2003) 196102

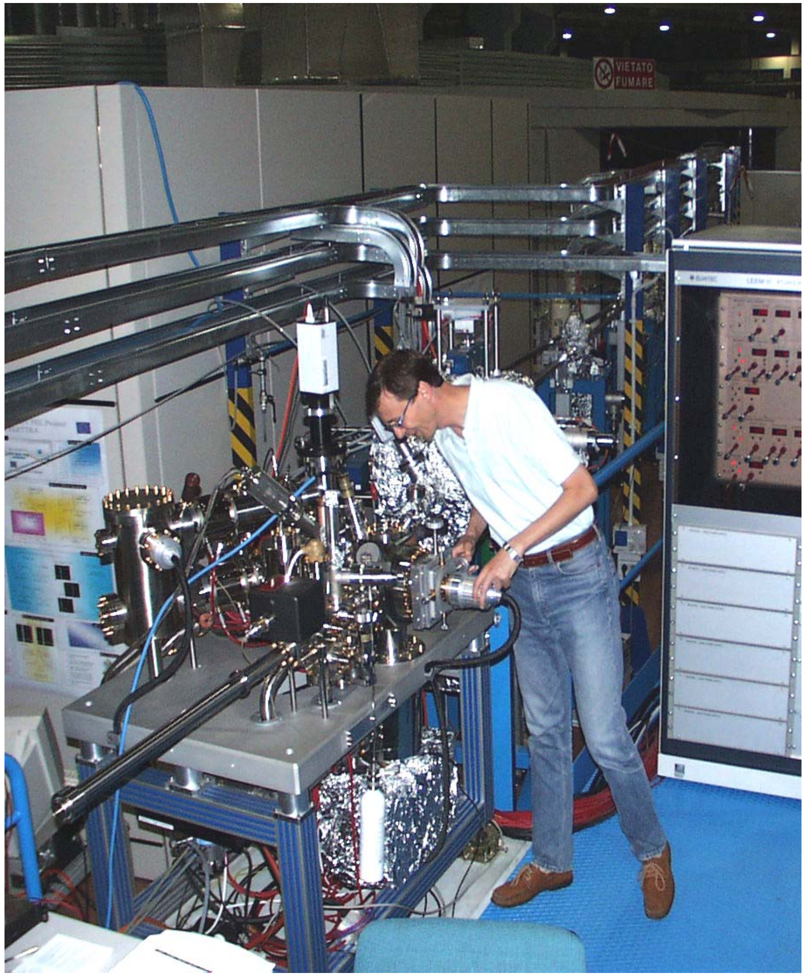
Concentration Profiles

- Concentration maps
  - in cross-section: TEM, STM, XRD
  - in top-view: etching, XRM



F. Ratto, S. Heun et al.: Small **2** (2006) 401.

The SPELEEM at ELETTRA

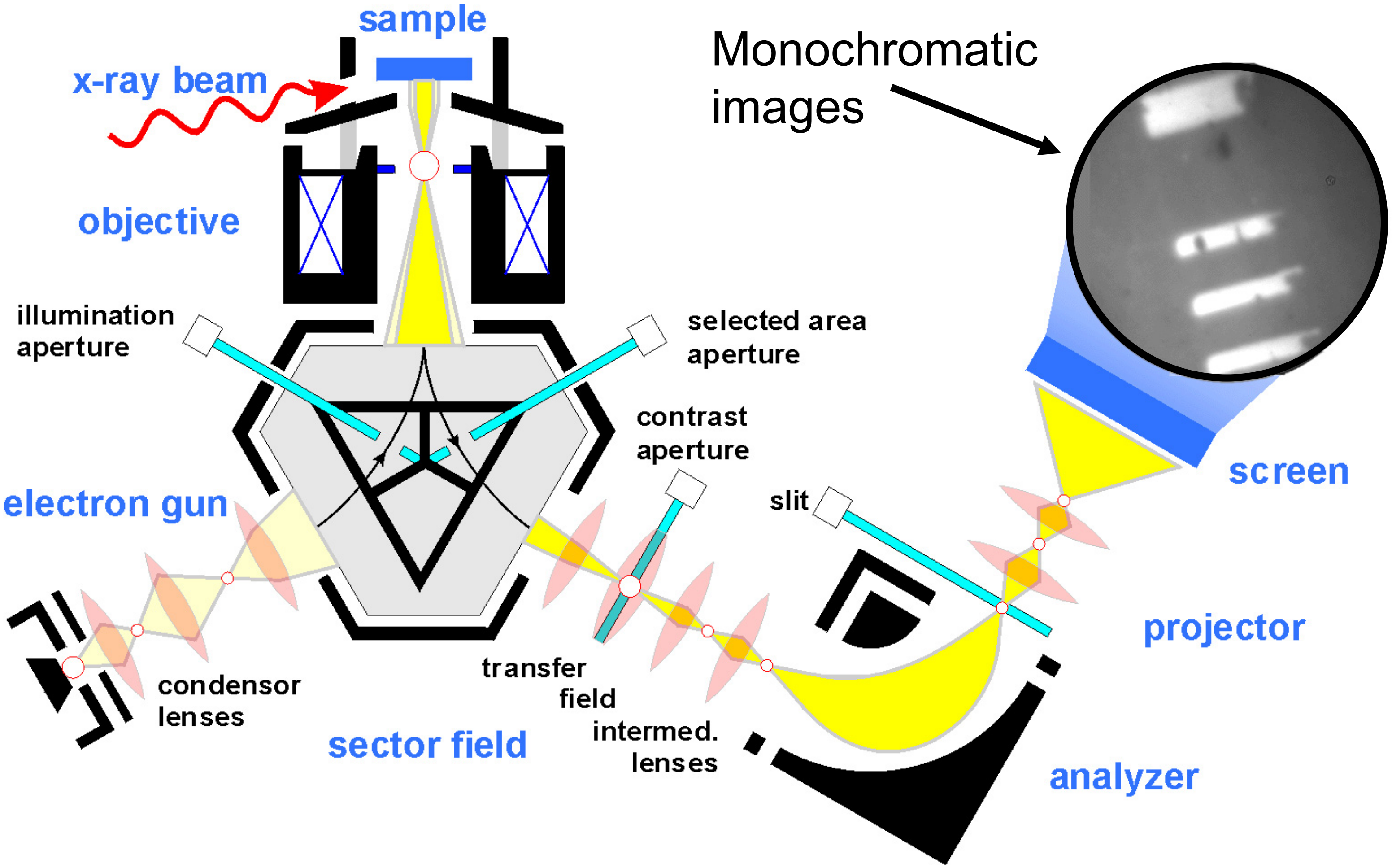


- Best energy resolution: 250 meV
- Best lateral resolution: 25 nm

- Variable polarization
- 20 - 1000 eV
- Photon flux 10^{13} ph/s
- Small spot (2µm x 25µm)

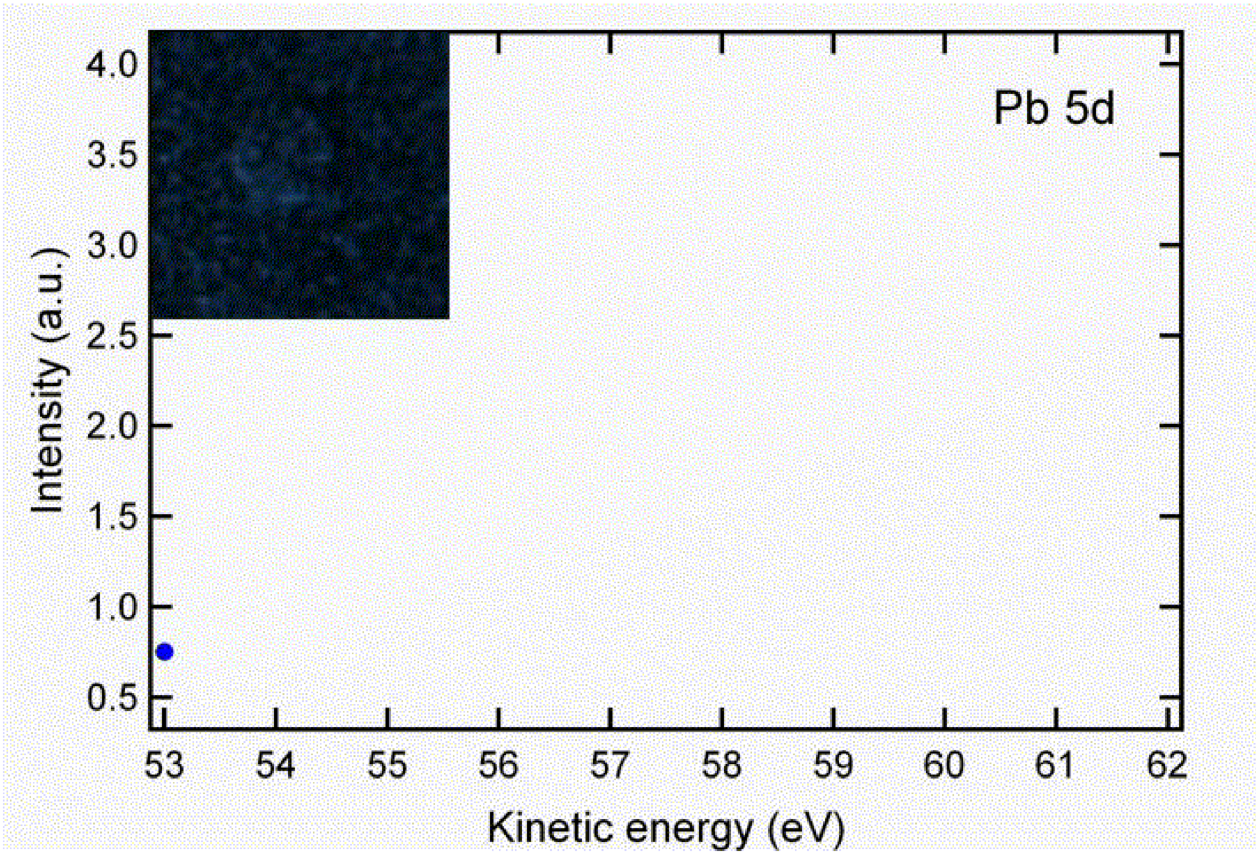
The SPELEEM instrument

Spectroscopic Photo-Emission and Low Energy Electron Microscope

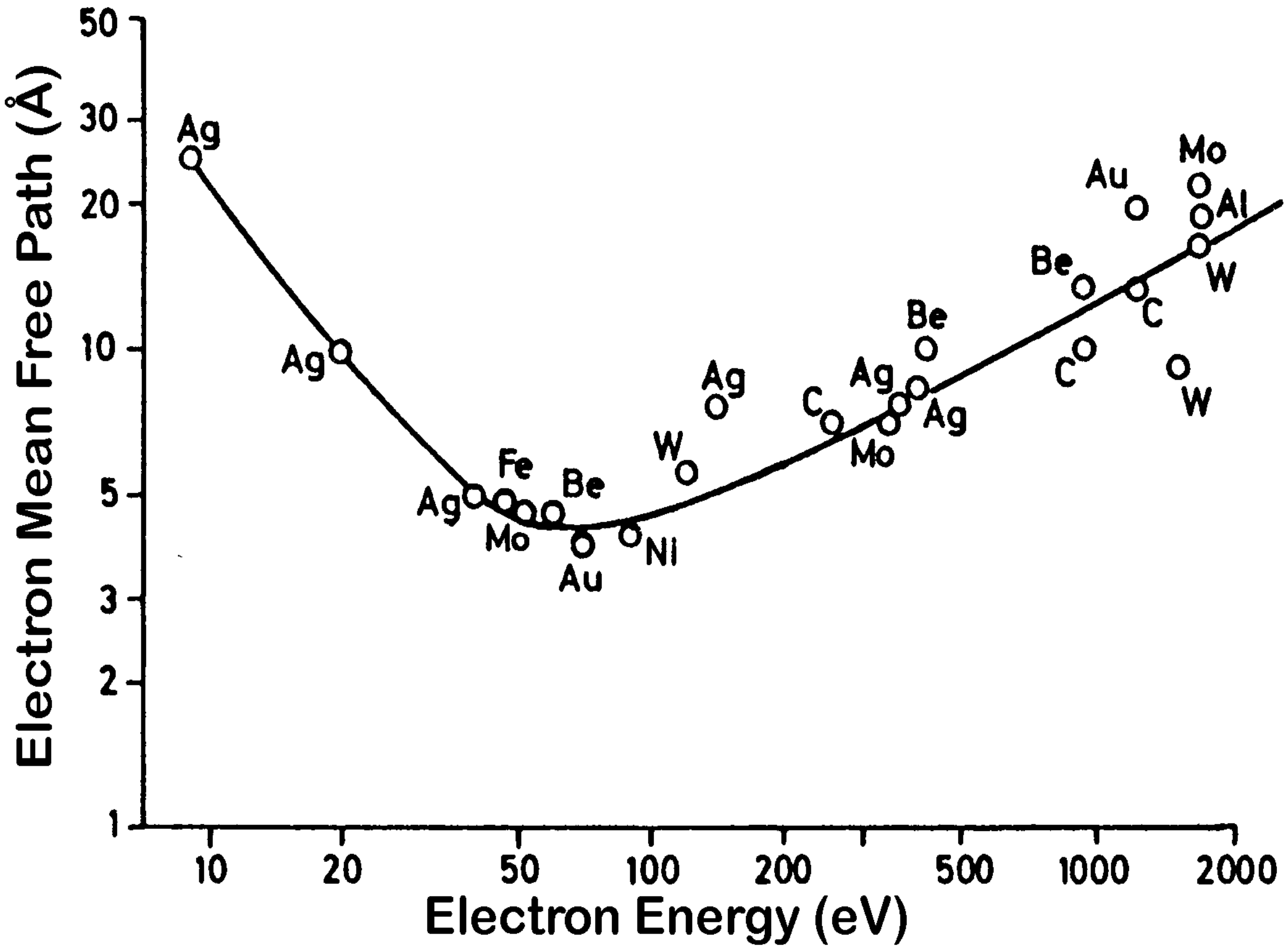


XPEEM: Core Level Spectroscopy

- Pb/W(110), Pb 5d core level, hν = 80 eV
- Best energy resolution: 250 meV

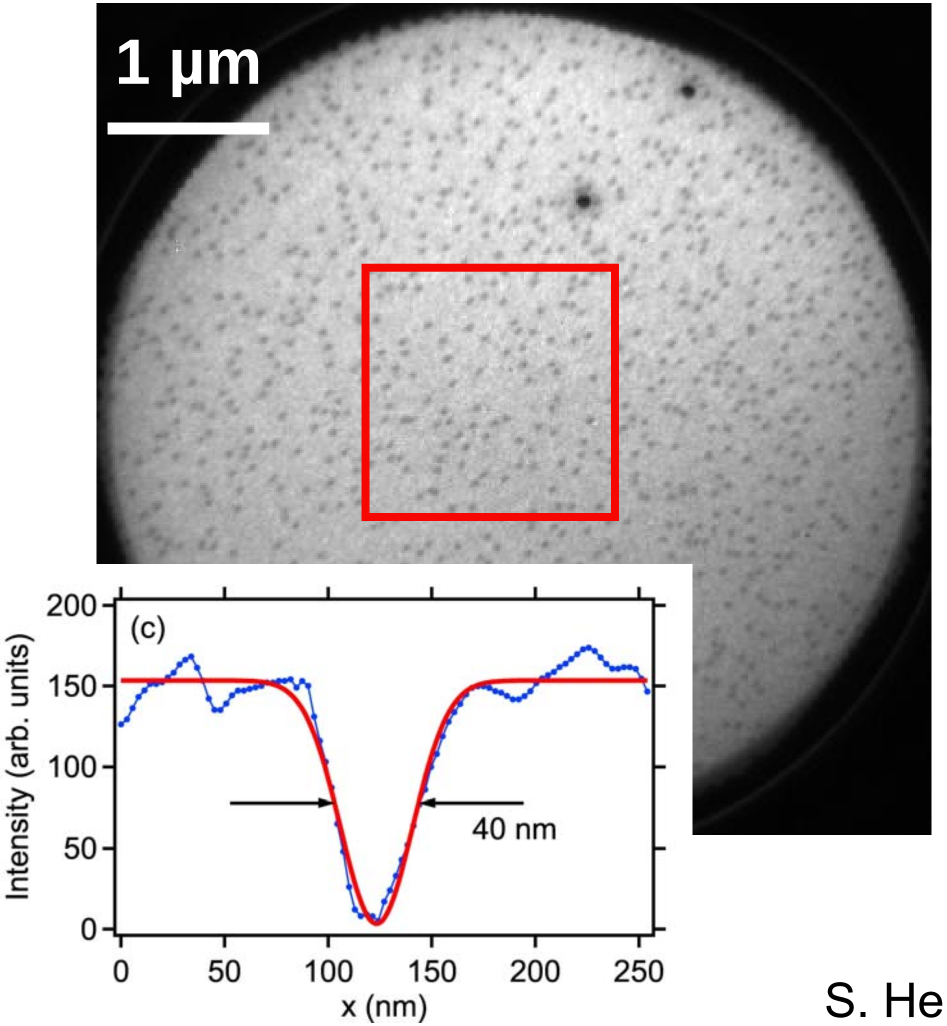


Photoelectron Mean Free Path

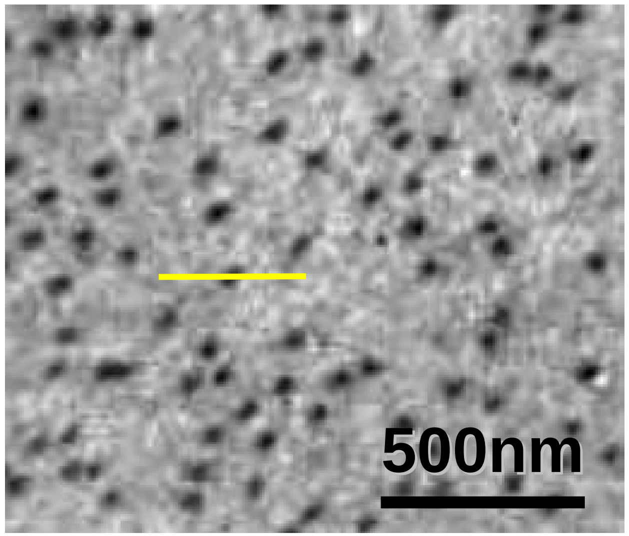


M. P. Seah and W. A. Dench: Surf. Interface Anal. **1** (1979) 2.

InAs/GaAs Islands (LEEM)

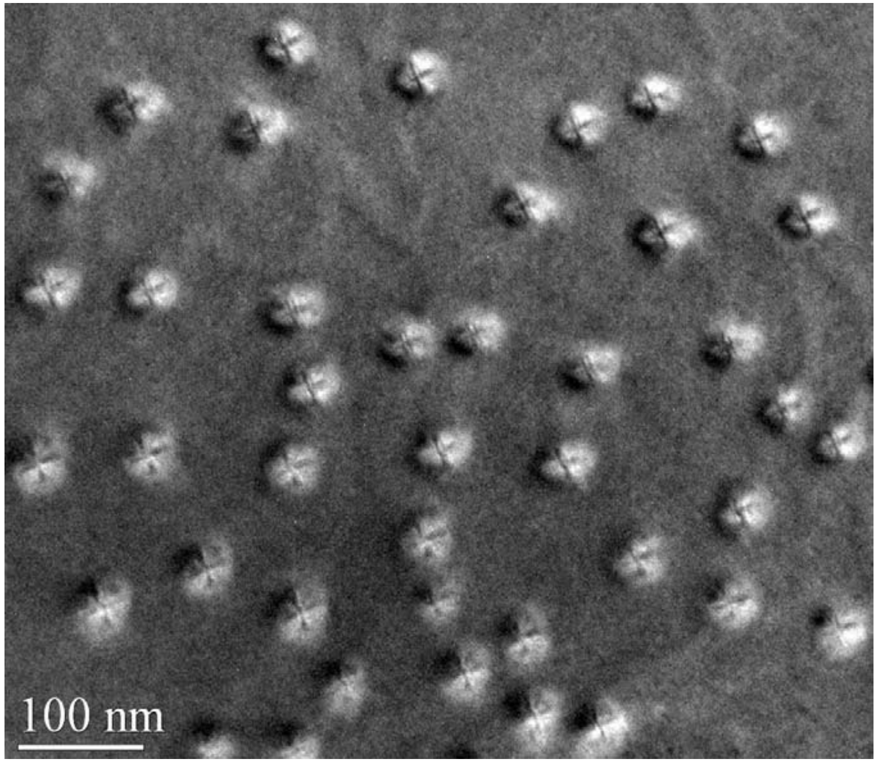


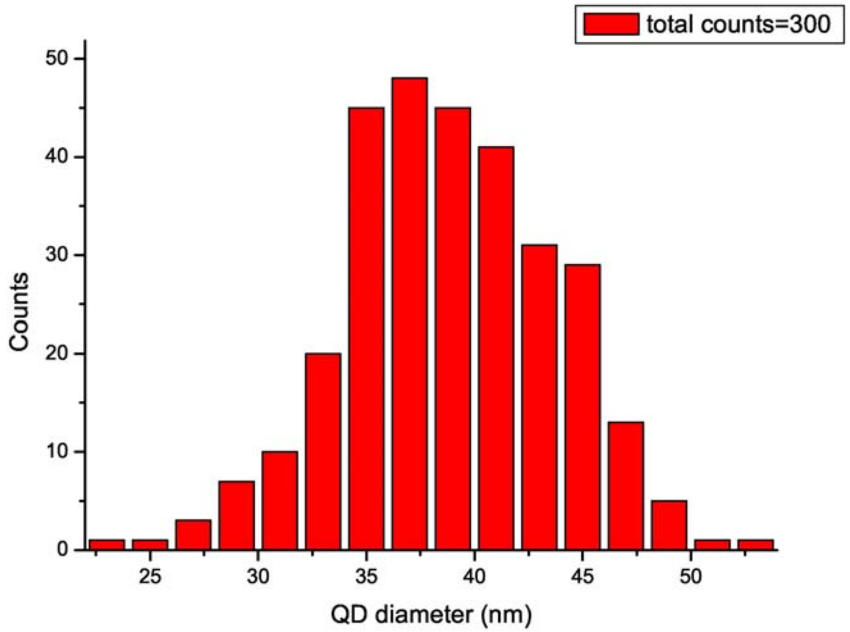
- Electron Microscopy (LEEM)
- 5 µm FOV
- E_{kin} = 7.6 eV



S. Heun et al.: J. Nanosci. Nanotech., in press.

TEM investigation

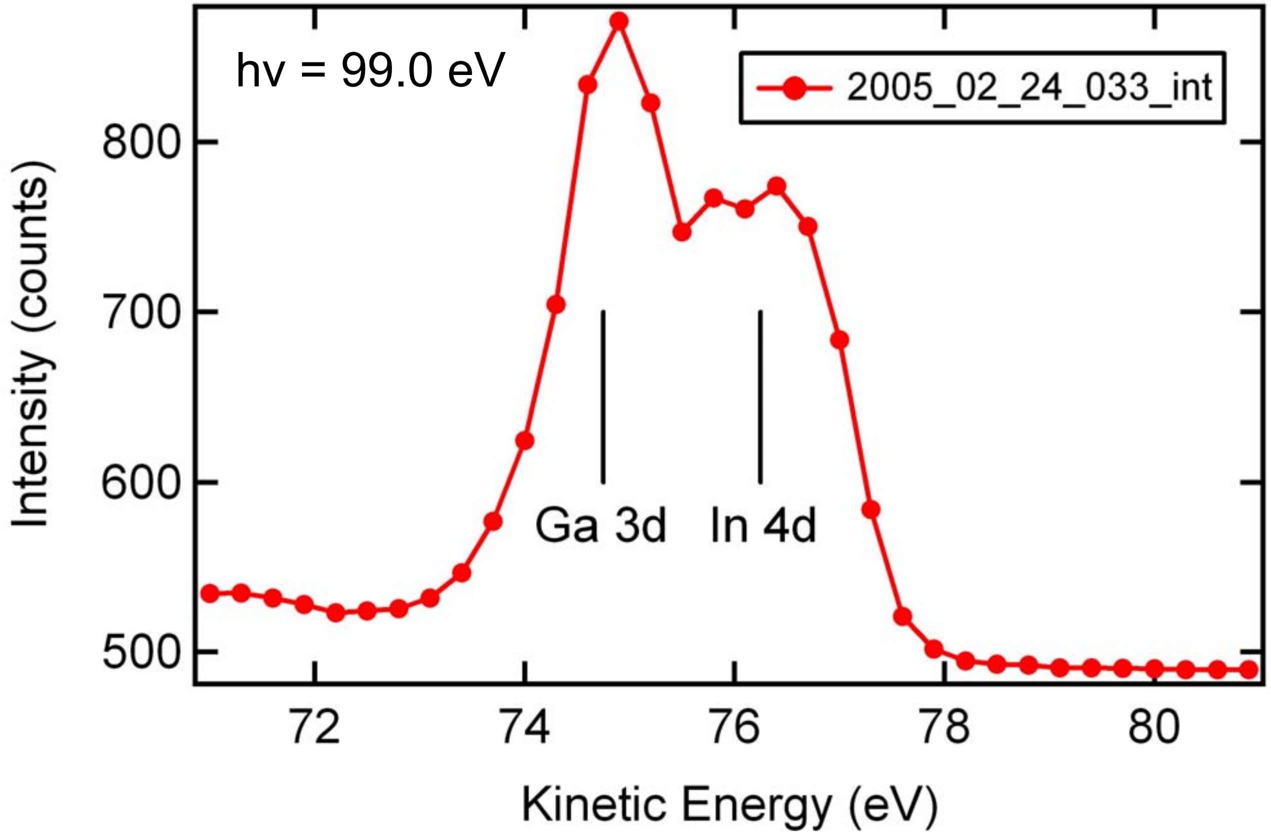




- Sample imaged in [001] plan view geometry
- Islands are coherent, i.e. no dislocations

S. Heun et al.: J. Nanosci. Nanotech., in press.

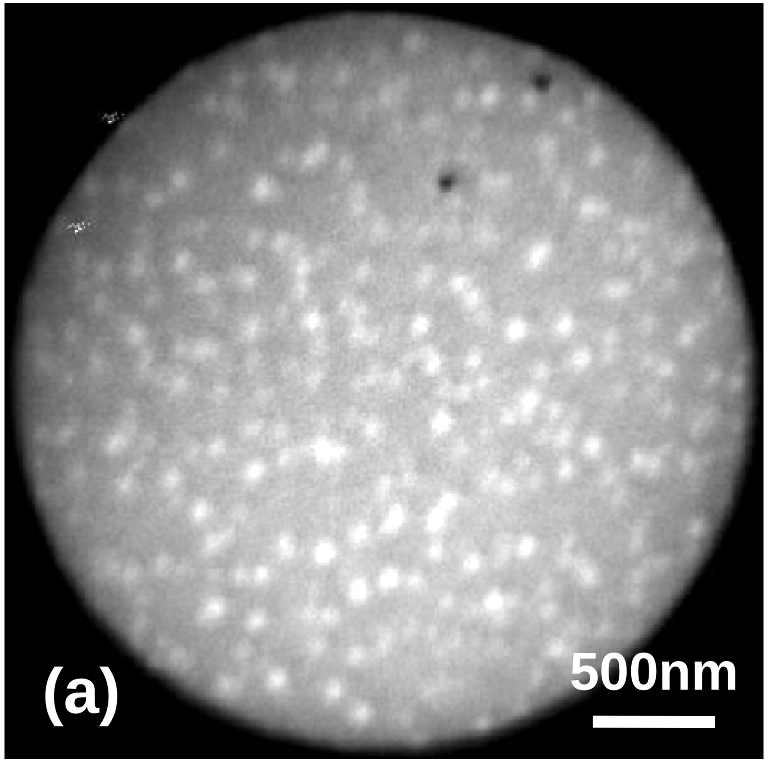
“Integral” Core Level Spectra

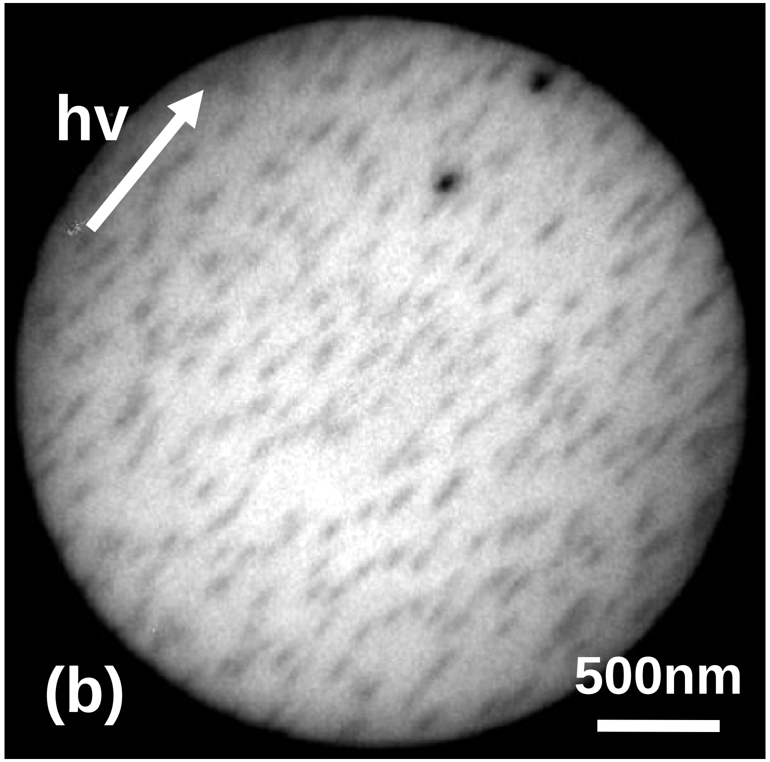


- Spectra taken from a 1 µm x 1 µm sample area.

G. Biasiol, S. Heun et al.: Appl. Phys. Lett. **87** (2005) 223106.

XPEEM Core Level Imaging



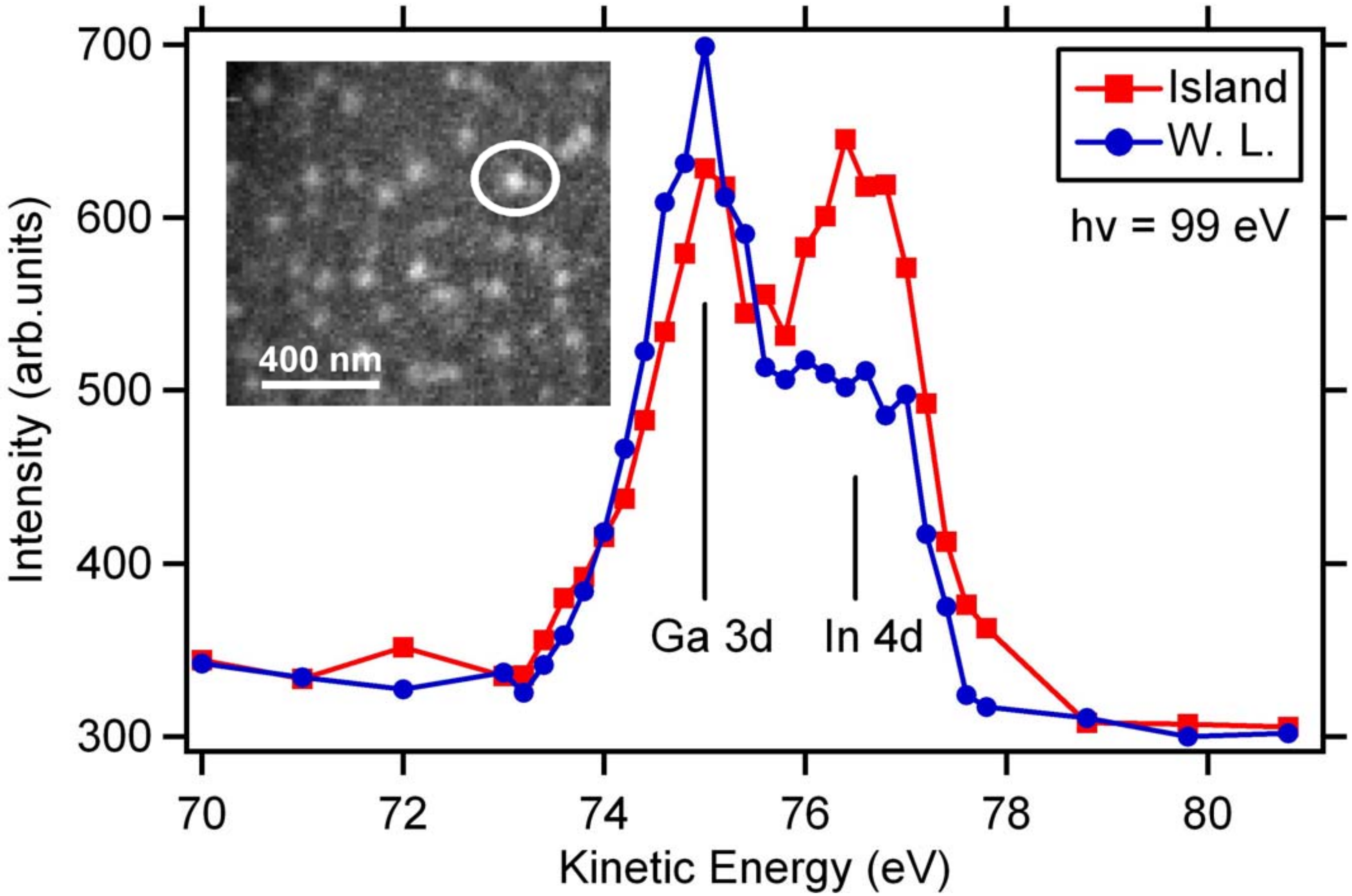


In 4d XPEEM image
hv = 99.0 eV, E_{kin} = 76.25 eV

Ga 3d XPEEM image
hv = 99.0 eV, E_{kin} = 74.75 eV

G. Biasiol, S. Heun et al.: Appl. Phys. Lett. **87** (2005) 223106.

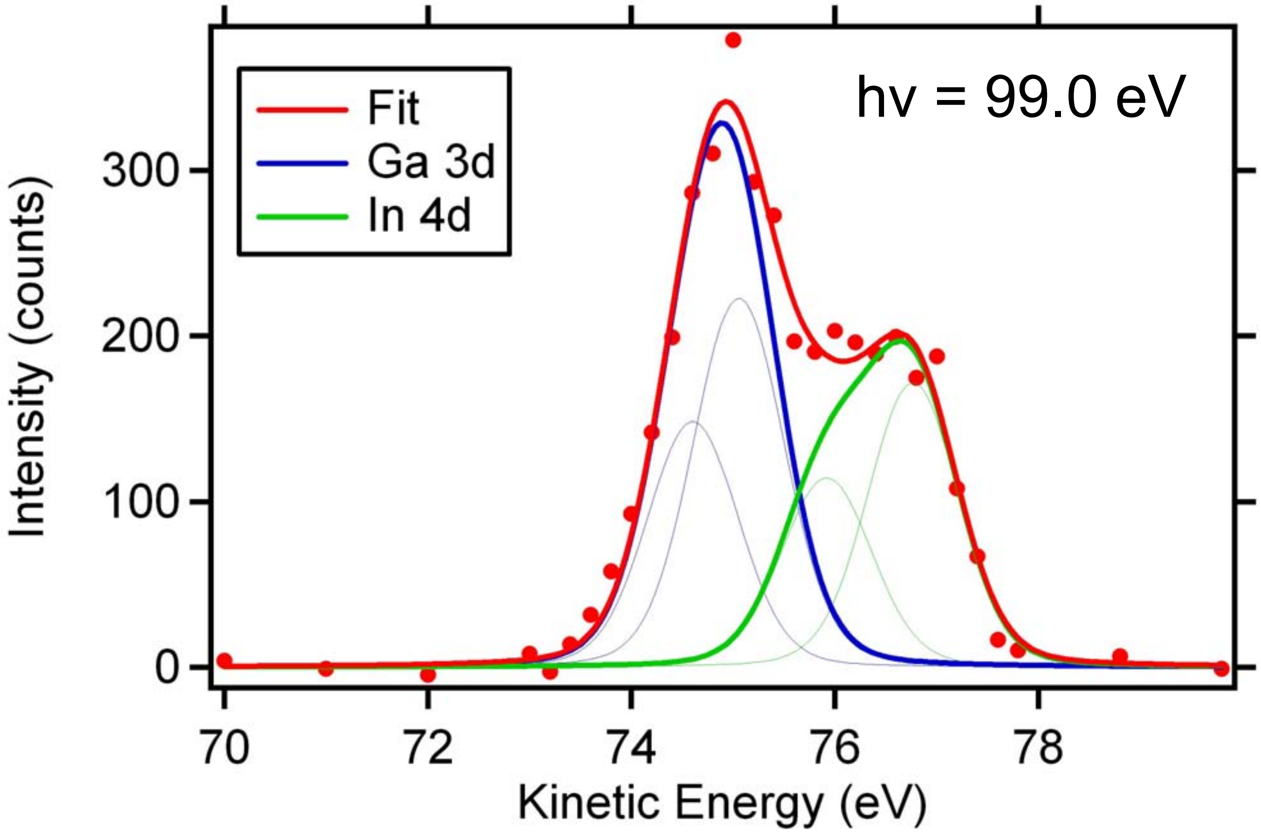
XPEEM Local Spectra



Integration area 25 nm x 25 nm, energy resolution ≈ 1 eV

G. Biasiol, S. Heun et al.: Appl. Phys. Lett. **87** (2005) 223106.

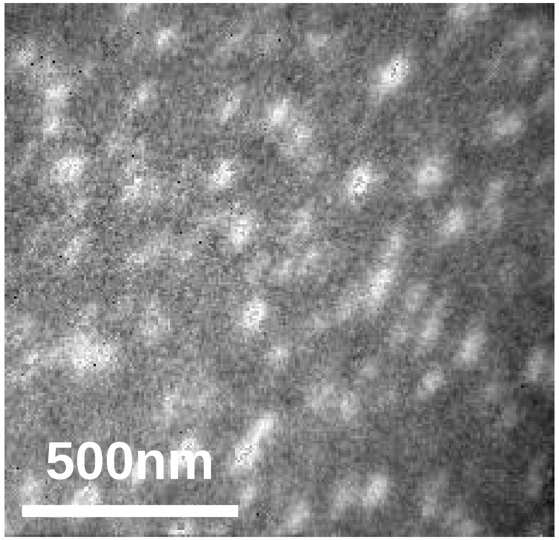
Core Level Line Profile Analysis



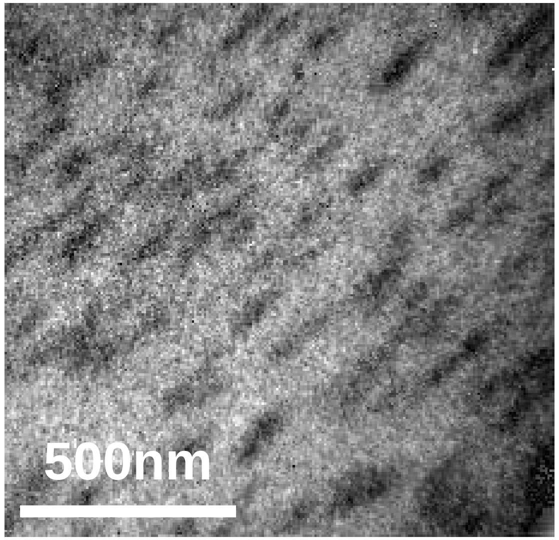
Spectrum from Wetting Layer, Shirley Background subtracted
Gauss 1 eV, Lor 0.16 eV, BR 1.5, SO: Ga 3d 0.45 eV, In 4d 0.85 eV

G. Biasiol, S. Heun et al.: Appl. Phys. Lett. **87** (2005) 223106.

2D Fit of XPEEM Data



In 4d peak area
Min: 220, Max: 520



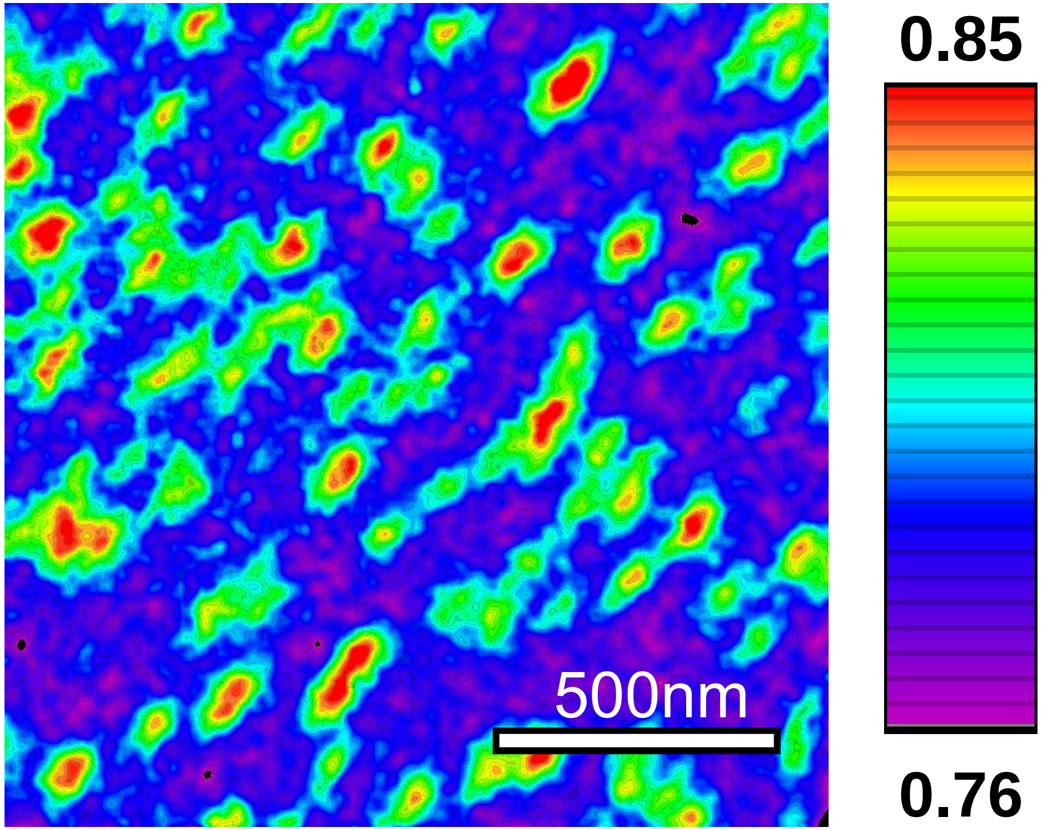
Ga 3d peak area
Min: 270, Max: 470

Ratio of Number of Atoms: $$\frac{n_{In}}{n_{Ga}} = \frac{I_{In}}{I_{Ga}} \cdot \frac{\sigma_{Ga}}{\sigma_{In}}$$

G. Biasiol, S. Heun et al.: Appl. Phys. Lett. **87** (2005) 223106.

Indium Surface Concentration Map

$$\frac{n_{In}}{n_{tot}} = \frac{I_{In}\sigma_{Ga}}{I_{In}\sigma_{Ga} + I_{Ga}\sigma_{In}}$$



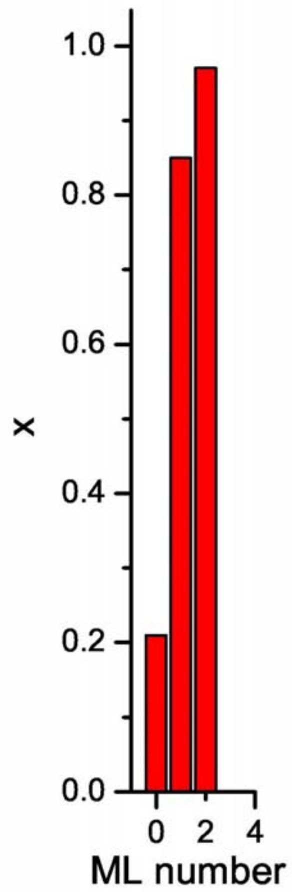
G. Biasiol, S. Heun et al.: Appl. Phys. Lett. **87** (2005) 223106.

Wetting Layer Composition

- Segregation models predict the following In concentration profile:
- Measured composition is average across topmost layers:

$$< x > = \sum x_i e^{-\frac{d_i}{\lambda}} \Big/ \sum e^{-\frac{d_i}{\lambda}}$$

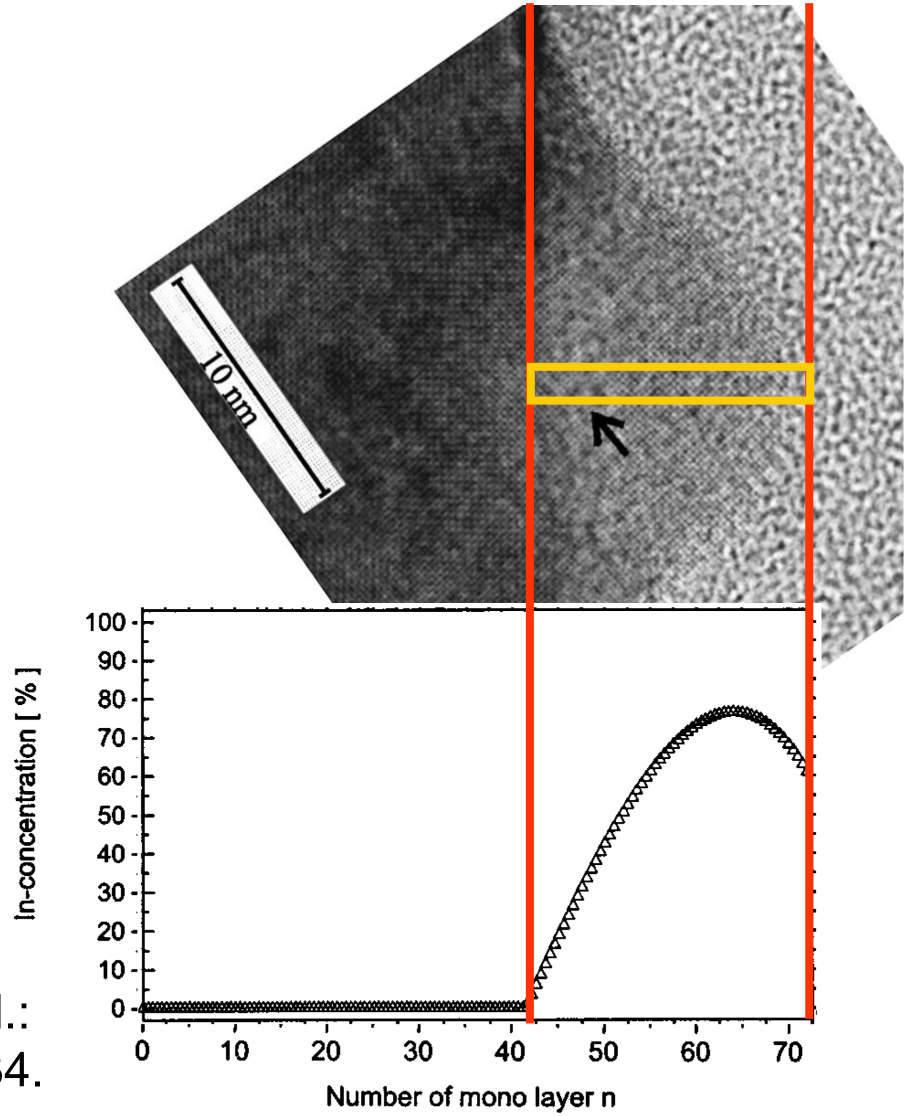
- Shown profile would be measured as x ~ 0.75, in agreement with our data.



O. Dehaese et al.:
Appl. Phys. Lett. **66** (1995) 52.

Dot Composition from TEM

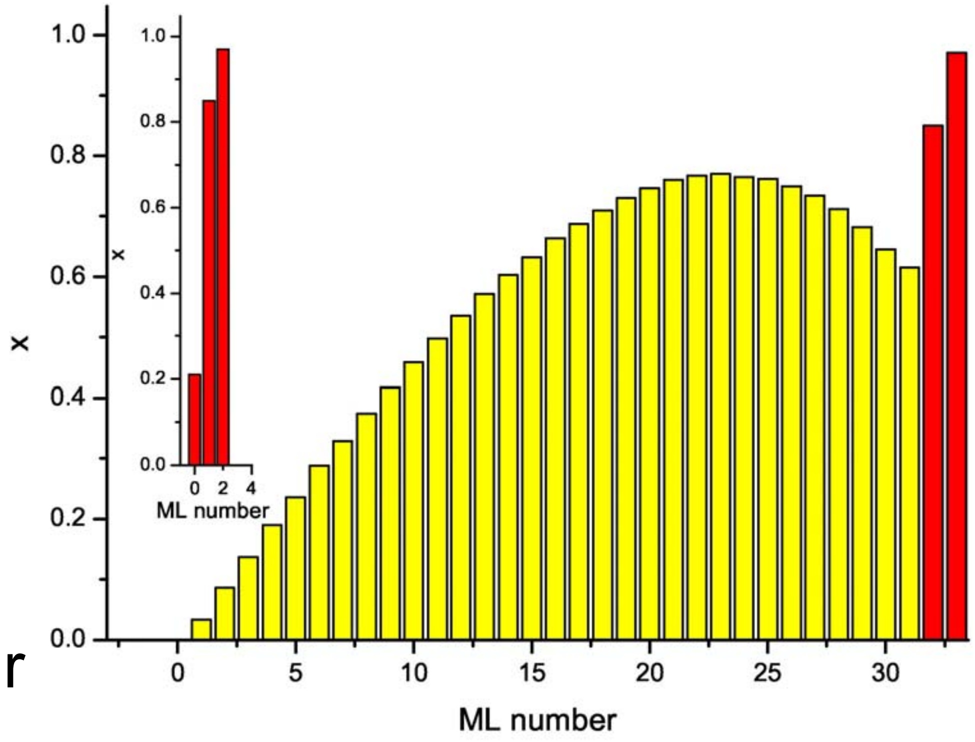
- At surface: x ~ 0.6
- Max of x ~ 0.8 at 10 ML below the surface
- We would measure this profile as x ~ 0.65
- Our data: x ~ 0.85



A. Rosenauer et al.:
Phys. Rev. B **64** (2001) 245334.

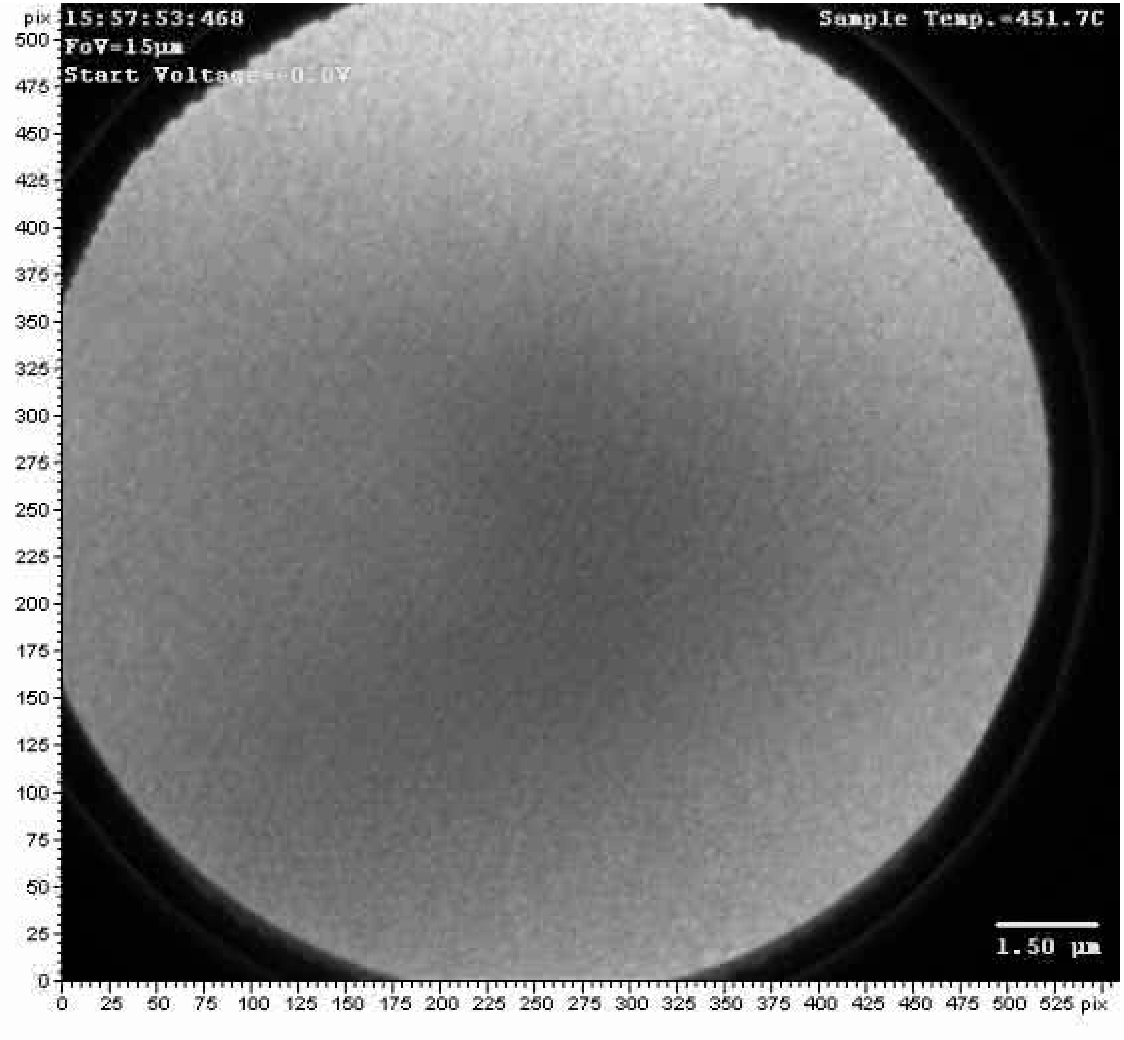
Indium depth concentration profiles

- Strong In segregation also on surface of dots.
- Add double layer with x ~ 0.85 - 0.97 (like WL) to surface.
- We would measure this profile as x ~ 0.85, in good agreement with our data.



S. Heun et al.: J. Nanosci. Nanotech., in press.

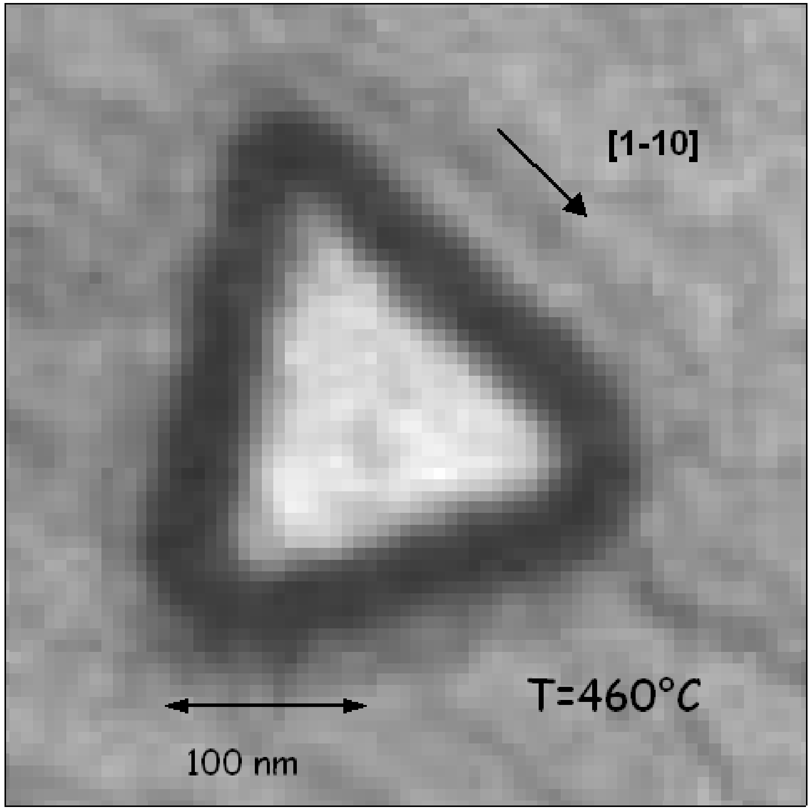
Ge/Si(111) growth by LEEM



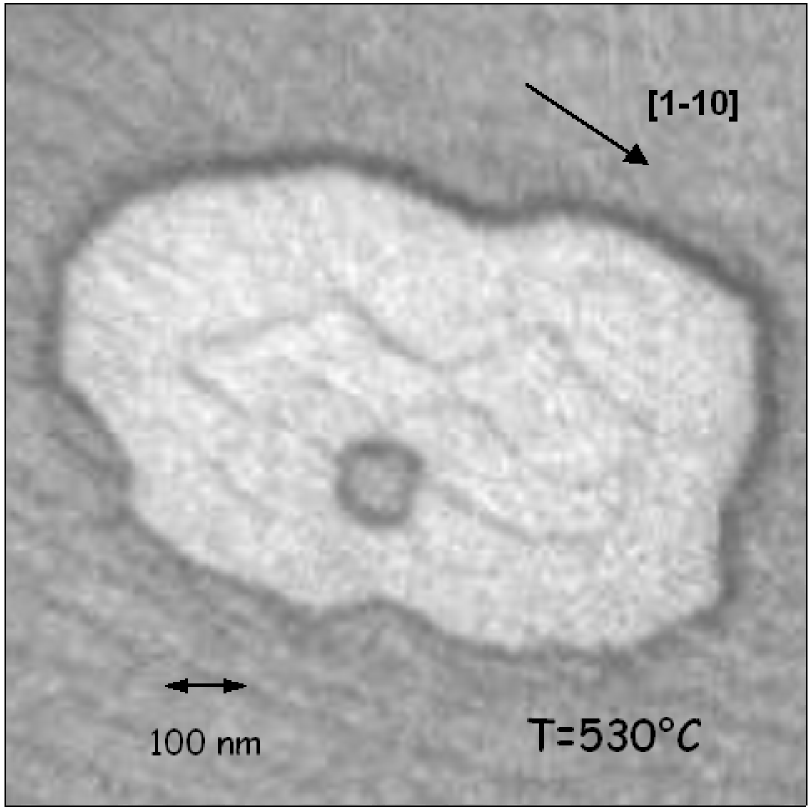
- LEEM Movie
  - FOV 15 µm
- MBE growth
  - T = 550ºC
  - 3 to 8 ML Ge on Si(111)
- Has been used to study diffusion dynamics during the nucleation and growth of Ge/Si nanostructures on Si(111)

F. Ratto, S. Heun et al.: Phys. Rev. Lett. **96** (2006) 096103.

Island shape evolution



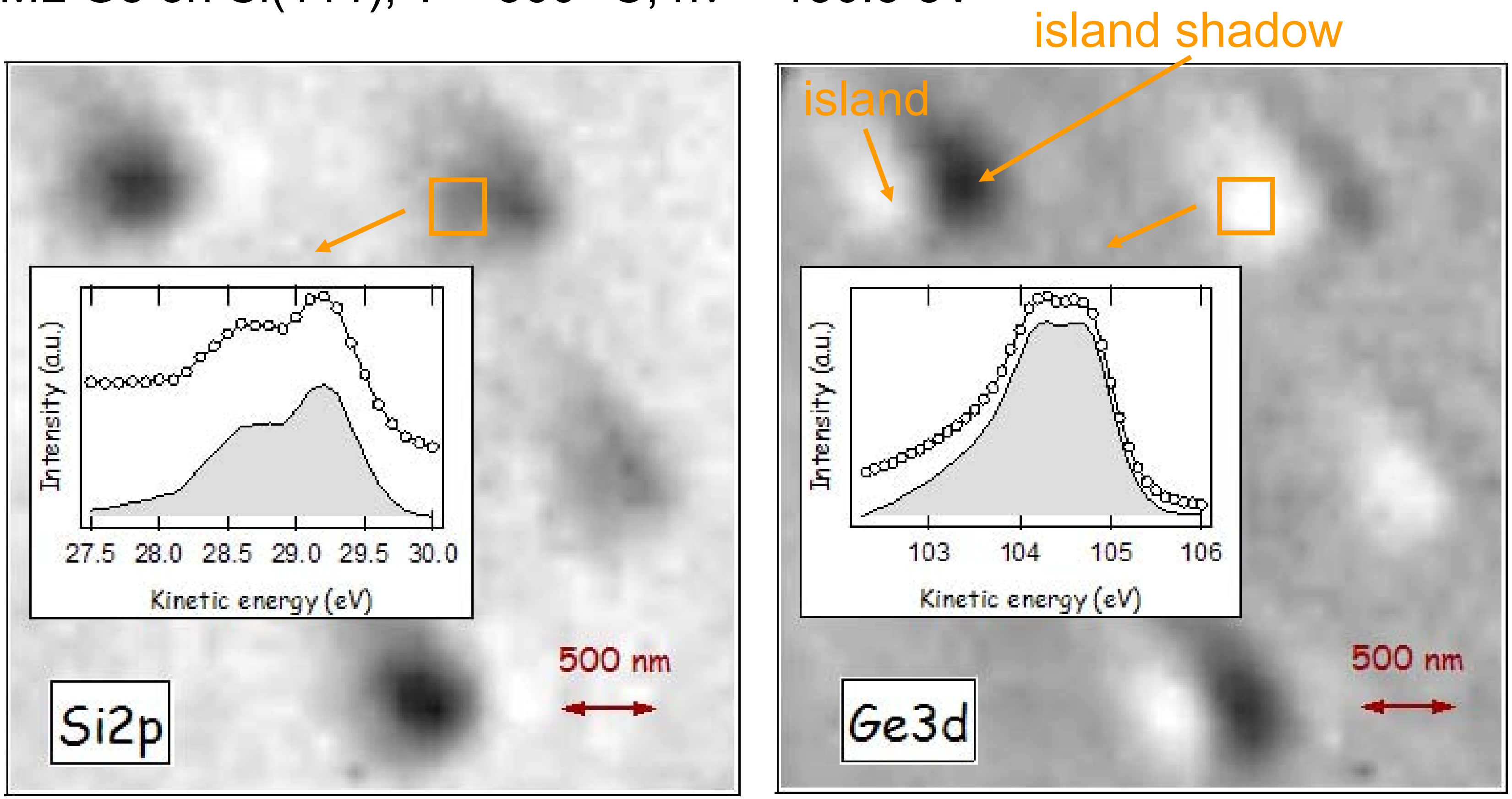
Truncated Pyramid:
strained, coherent



Atoll-like Morphology

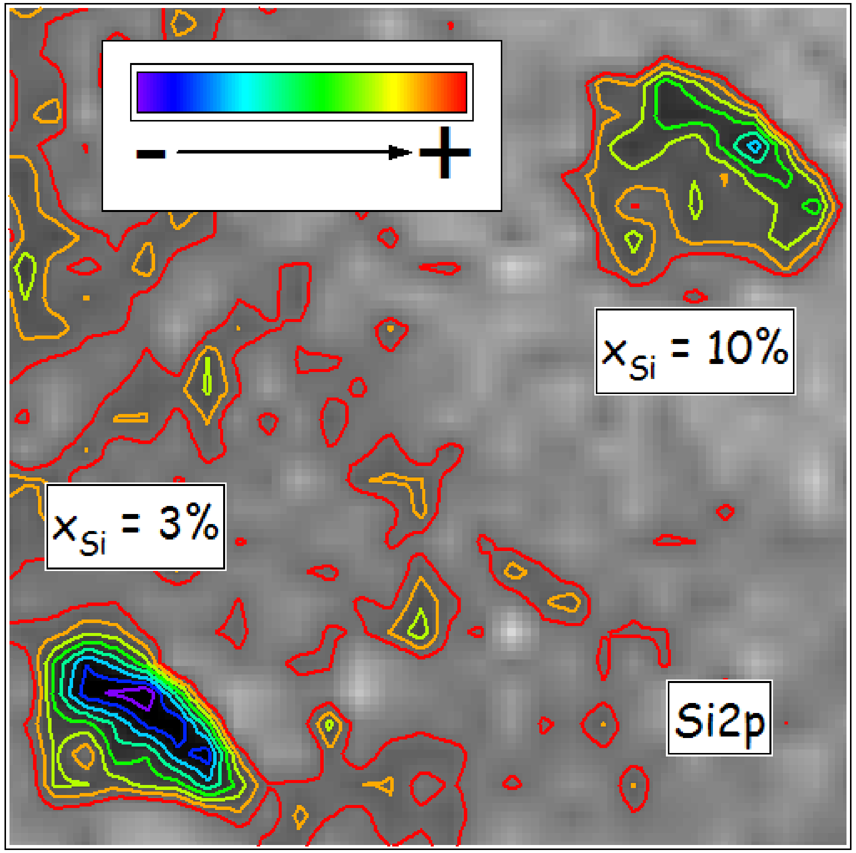
Chemical contrast by XPEEM

10 ML Ge on Si(111), T = 560 °C, hv = 130.5 eV



F. Ratto, S. Heun et al.: Appl. Phys. Lett. **84** (2004) 4526.

Intensity contour maps of islands

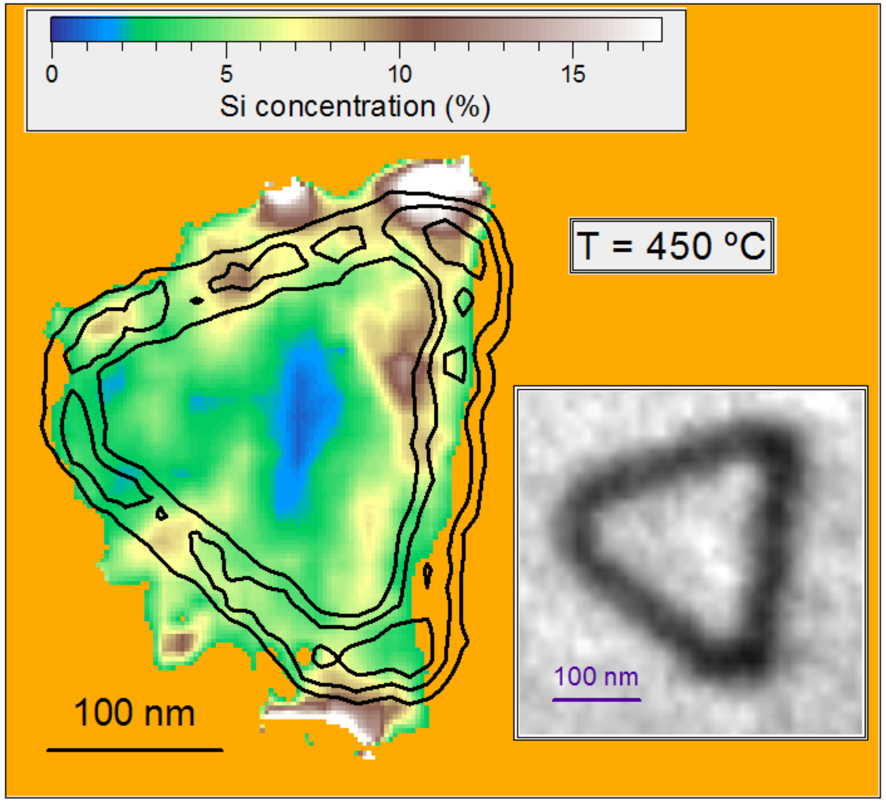


- Intensity contour maps of a more (top) and a less (bottom) ripened island.
- Photoelectron yields are increasing from blue (lowest) to red (highest).
- Darkest regions: shadows of the 3 D islands, due to the 16° X–Ray incidence angle.
- The WL is highly inhomogeneous.

2×2 μm^2 Si2p core level integrated XPEEM image
Growth at T = 530 °C

F. Ratto, S. Heun et al.: Appl. Phys. Lett. **84** (2004) 4526.

Comp. mapping of Ge/Si islands

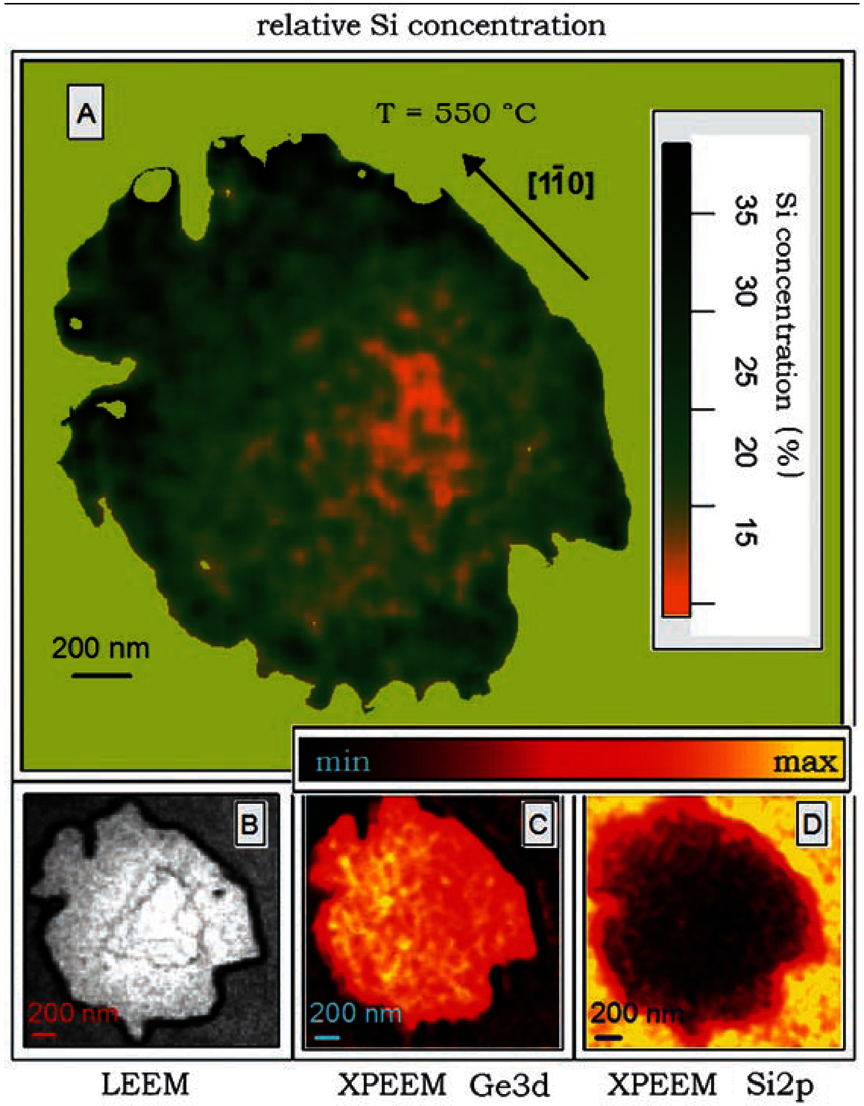


Island height: about 25 nm

- Relative Si surface concentration in a Ge(Si) island on Si(111).
- The composition mapping is obtained by combining sequences of Si2p and Ge3d XPEEM micrographs with a lateral resolution of ~30 nm.
- Inset: LEEM image of the same 3D structure (~10 nm lateral resolution).
- 10 MLs Ge
- Rate: 0.2 ML/s
- T = 450 °C

F. Ratto, S. Heun et al.: Small **2** (2006) 401.

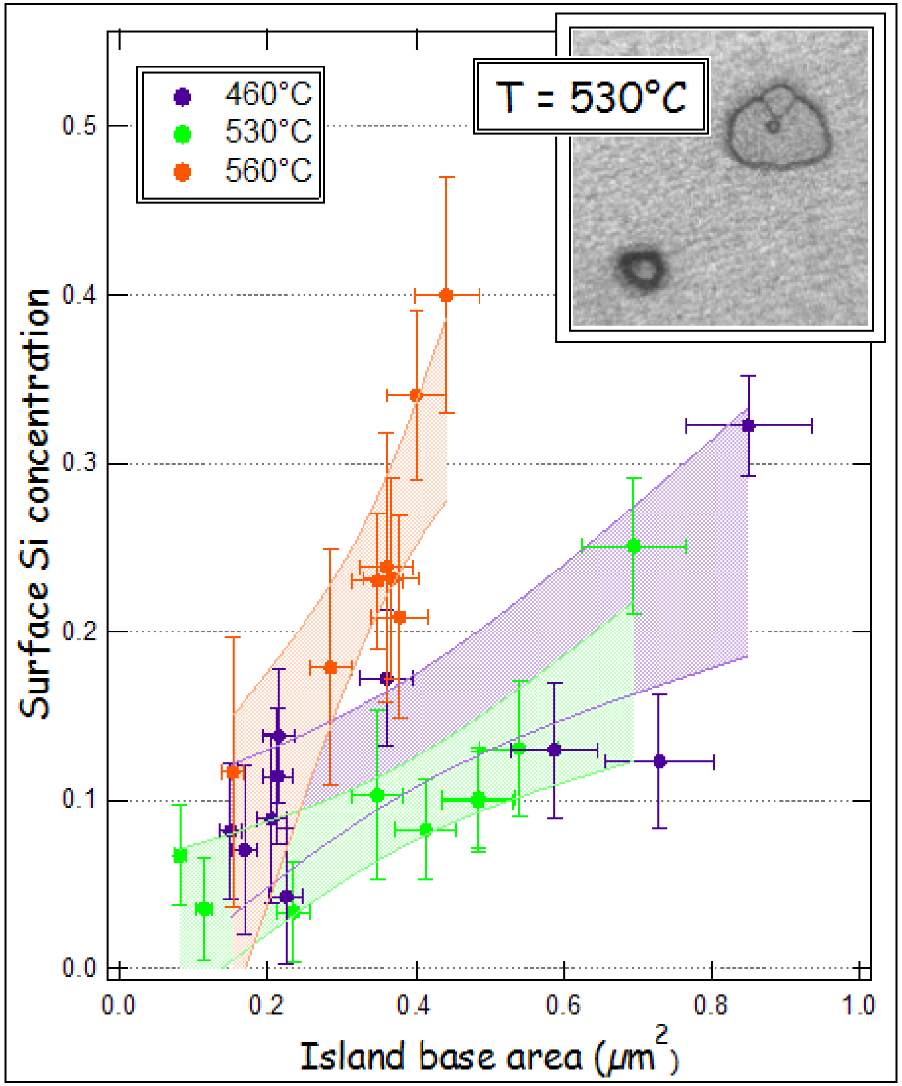
Top view concentration mapping



- B: LEEM image of a Ge/Si 3D island (T = 550 °C).
- C: Normalised XPEEM Ge3d intensity.
- D: Normalised XPEEM Si2p intensity. The chemical contrast in the image shows that the WL is richer in Si than the islands.
- A: Estimated surface Si concentration map (combining the information from C and D).
- The concentrations are an average over 0.5÷2 nm from the surface (absolute error: ~2%).
- The Si content increases with island lateral dimensions and deposition temperature.

F. Ratto, S. Heun et al.: Small **2** (2006) 401.

Si conc. vs. island morphology



- Si surface concentration as a function of island base area.
- At each deposition temperature, the stoichiometry is uniquely determined by the island's lateral dimensions.

F. Ratto, S. Heun et al.:
J. Appl. Phys. **97** (2005) 043516.

Conclusions

- Surface concentration maps of InAs/GaAs and Ge/Si quantum dots by SPELEEM.
- Dot composition neither pure InAs (Ge) nor homogeneous $In_xGa_{1-x}As$ (SiGe).
- In (Ge) concentration decreases from center (high) to borders (low) of dots.
- In segregation (x ~ 0.9) on surface of InAs dots and WL.
- The Si content in Ge/Si dots increases with island lateral dimensions and deposition temperature.
- X-ray microscopy powerful tool for surface chemical mapping of individual nanostructures

Ackonwledgements

- InAs/GaAs:
  - G. Biasiol, G. B. Golinelli, V. Grillo, E. Carlino, L. Sorba, *Laboratorio TASC INFM-CNR, Trieste, Italy*
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  - A. Locatelli, T. O. Mentes, *Sincrotrone Trieste, Italy*
- Ge/Si:
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  - S. Kharrazi, S. Ashtaputre, S. K. Kulkarni, *University of Pune, India*
  - S. Fontana, A. Locatelli, *Sincrotrone Trieste, Italy*