Photoelectron spectroscopy from individual heteroepitaxial nanocrystals on GaAs(001)

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Motivation

Nanocrystals formed spontaneously during strained-layer epitaxy
• Quasi zero-dimensional nature (quantum dots)
• Semiconductor lasers and memory applications
• No lithography: cost-effective fabrication of devices

Problems:
• Size fluctuations: need for nano-scale spectroscopy
• Segregation and interdiffusion observed for Ge/Si and for InAs/GaAs

Purpose of this work:
• Determination of the elemental composition of the nanocrystals
• Photoelectron spectroscopy with high lateral resolution
• Electronic structure of a single InAs nanocrystal

Sample Preparation

Nanocrystal fabrication process (in Tsukuba, Japan):

- Substrate: n-type GaAs(001), growth of GaAs buffer layer
- Se-treatment (forming a 2-3 ML-thick film of Ga$_2$Se$_3$ on top of bulk GaAs)
- Deposition of nominally 2 ML InAs by MBE
- As capping to protect samples during transfer in air

Characterization with SPELEEM at beamline 6.2LL at Elettra
InAs / Se / GaAs after capping and decapping.

Typical island size: 50 nm.
Typical island height: 20 nm.
Typical island density: 20 µm⁻²

Field of view: 2µm x 2µm.
InAs / Se / GaAs surface reconstruction after growth: 2x1

Reference: GaAs(100)-(2x4) surface

2x1 reconstruction of InAs on Se/GaAs after capping / decapping

Reproducibility of surface stoichiometry

Before As capping

After As decapping in UHV

- Virtually identical spectra before and after decapping
- No oxidized components
- Intensity ratio Ga 3d / In 4d not changed

The SPELEEM at ELETTRA

Spectroscopic photoemission and low energy electron microscope

Lateral Resolution of XPEEM

Evaluated spatial resolution in XPEEM mode: 22nm

Energy Resolution of XPEEM

Energy resolution better than 0.5 eV

Pb/W(110)

XPEEM

nanocrystals have square base oriented along $<110>$

Island Size Distribution

Sampled area: 20 μm²
Nanocrystal density: 25 μm⁻²
Average diameter: 53 nm

Core Level Spectra

Integral photoemission spectra from a 3 µm² area, obtained by integration of a series of SPELEEM images.

Good agreement with literature values
↑ samples not changed by capping / decapping

\( h\nu = 52 \text{ eV}, \text{ FoV} = 2\mu\text{m} \)

nanocrystal height from their shadow length: 22 nm

Laterally resolved core level spectroscopy

Integral spectrum

Laterally resolved spectra

(a) Ga 3d / In 4d
hv = 52 eV

(b) Ga 3d / In 4d
hv = 52 eV

Indium on substrate ↑ SK growth mode
Gallium on nanocrystals

Sample topography vs. photoelectron yield

Sample topography vs. photoelectron yield

For emission along the sample normal:
photoelectron intensity ratio from inclined to flat unit areas of the sample:

$$\frac{I_{fn}}{I_s} = \frac{\sin(\beta_f + \alpha)}{\sin \alpha}$$

In transmission:

$$\frac{I_{bn}}{I_s} = \frac{\sin(\beta_b - \alpha)}{\sin \alpha} e^{-\ell/\lambda}$$

with \(\lambda\): x-ray attenuation length

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Laterally resolved core level spectroscopy

Integral spectrum

Laterally resolved spectra

Intensity correction factor for these nanocrystals: 1.24
↑ Ratio nanocrystals / substrate = 1.30 for In 4d, 0.92 for Ga 3d

Selenium on the nanocrystals

Valence band and work function data:
Further evidence for the presence of Selenium at the surface of the nanocrystals

$hv = 121 \, \text{eV}, \, E_b = 54.7 \, \text{eV}, \, \text{FoV} = 2\mu\text{m}$

Se 3d core level spectra

- Ga$_2$Se$_3$: zincblende structure with ordered Ga vacancies.
- A: Se with no Ga vacancy as nearest neighbor
- B: Se with one Ga vacancy as nearest neighbor
- B species closer to the surface than A species
- clean Se/GaAs: A / B = 1.51

Here: intensity ratio A / B = 2.26
↑ less B species after deposition of InAs
↑ some material moved from surface to nanocrystals

From LEEM: volume of nanocrystals: $6 \times 10^5$ nm$^3$ per µm$^2$
2 ML InAs correspond to $6 \times 10^5$ nm$^3$ per µm$^2$
SK growth mode
↑ nanocrystal volume is greater than expected
↑ additional material from another source (Ga$_2$Se$_3$)

Discussion

- Reaction between InAs and Ga$_2$Se$_3$
- No GaAs reaction (growth at 200°C)

- Formation of a quaternary unlikely (As and Se from different chemical groups)
- Alloying on cation sublattice (strain minimization)
- Phase separation on anion sublattice (like in InAs$_x$Sb$_{1-x}$)

- No bulk inclusions of Ga$_2$Se$_3$ in the InAs nanocrystals (10% lattice mismatch)

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Summary

• During heteroepitaxy, the InAs reacts with the Ga$_2$Se$_3$.

• Phase separation on anion sublattice, alloying on cation sublattice.

• A wetting layer of In$_x$Ga$_{1-x}$As is formed covered by (In$_y$Ga$_{1-y}$)$_2$Se$_3$.

• (In$_y$Ga$_{1-y}$)$_2$Se$_3$ covered nanocrystals are formed on this surface.