SAND2022-8670C

GaAs(Sb) nanostructures formed by arsenic-induced in-situ etching of III-Sb surfaces

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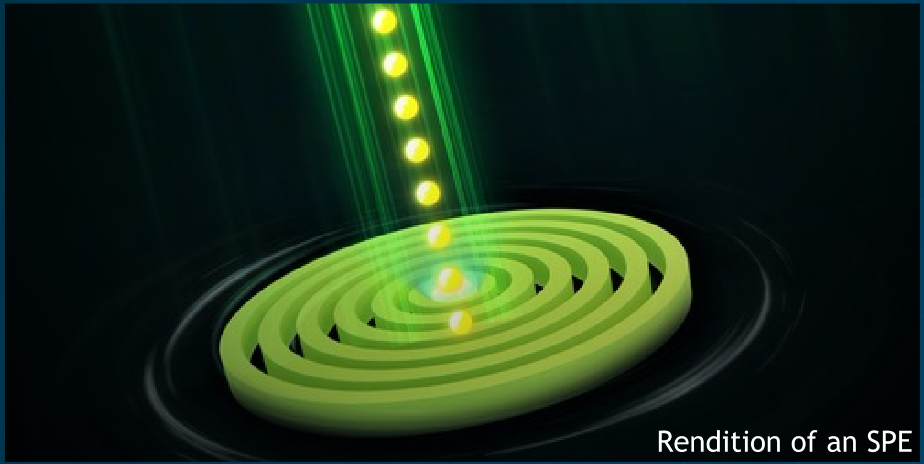
Electronic Materials Conference - The Ohio State University, Columbus, Ohio

June 30th 2022

This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. The views expressed in the article do not necessarily represent the views of the U.S. DOE or the United States Government. SJA acknowledges funding from Sandia's LDRD program.

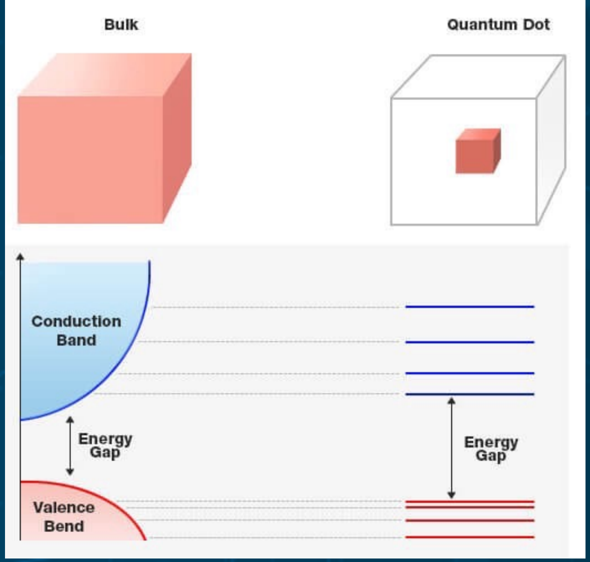
Motivation

Big picture: realizing efficient quantum emitters of single photons


Rendition of an SPE

Key ingredient for quantum information technologies

- Solid-state SPEs – combine optical properties of atoms with scalability
  - Semiconductor QDs, fluorescent atomic defects, 2D materials, carbon nanotubes
- Semiconductor QDs:
  - Quantum confinement in all spatial directions - discrete energy states
  - Explored to improve optoelectronic devices - suitable for integration



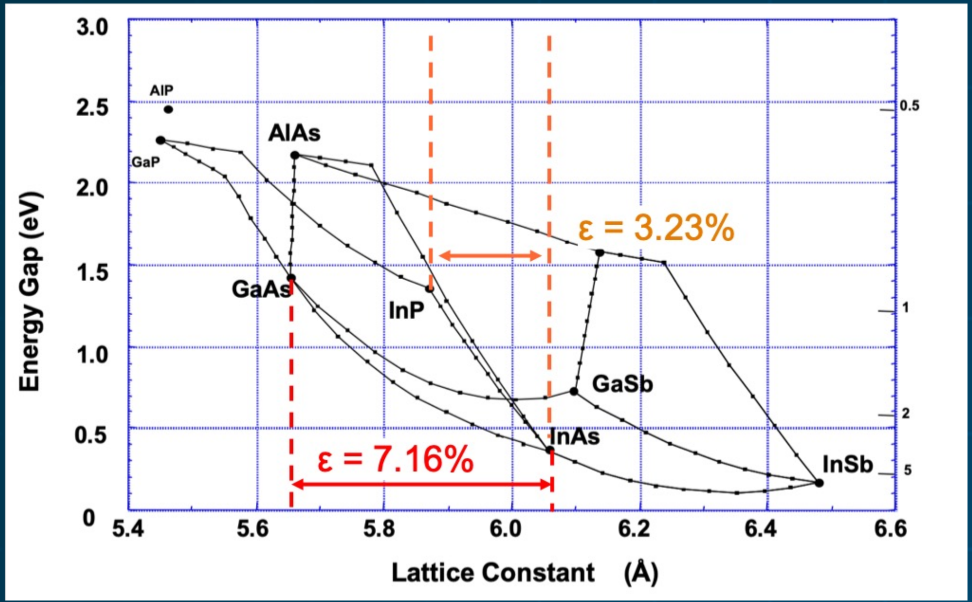
Energy levels: bulk vs QD

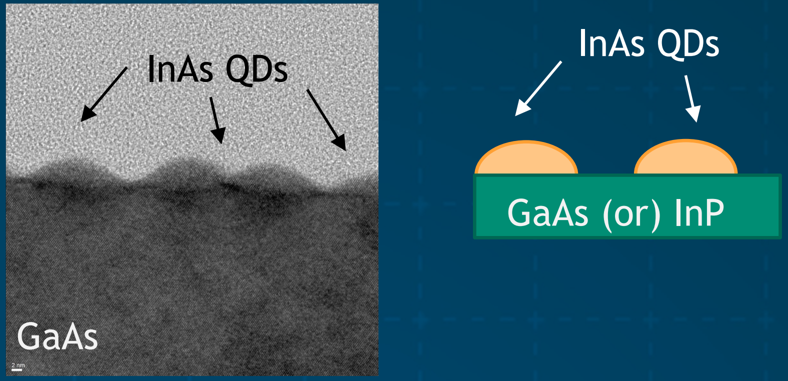
APS physics (2020); Medium (2019)

State-of-the-art

III-V QDs exhibit some of the highest all-around SPE performance*

- Stranski-Krastanov QDs (InAs, InGaAs)

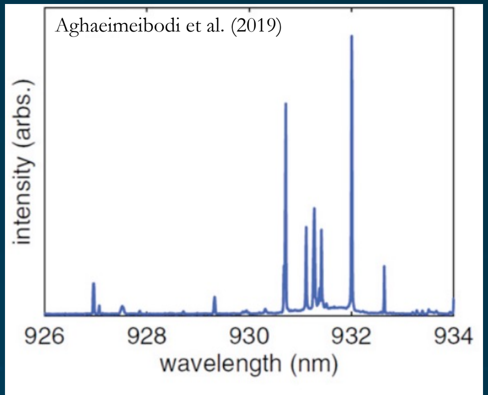


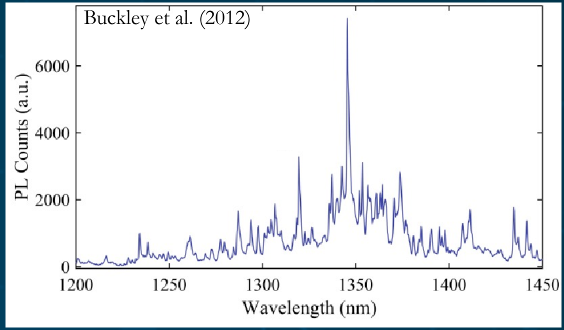


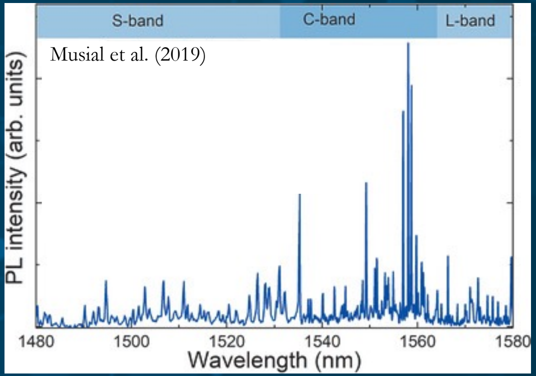
X-sectional TEM & schematic showing InAs/GaAs QDs

- Strain-driven formation mechanism
- Spans telecom λ range

- *Drawbacks:
  - Presence of a 2D layer interconnecting QDs
  - Self-assembly → limited range and control over shape, size and density
  - Strain-driven → limited combinations → limited λ







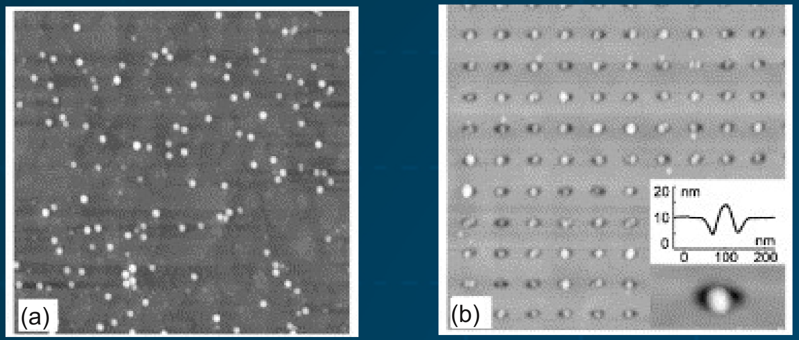
PL results from various works

State-of-the-art

S-K QDs : Efficient SPEs and widely used, but mechanism limits control/tunability

Alternative : Instead of self-assembly, QDs could be grown in pre-defined patterns → Reduces randomness; No material-choice issue

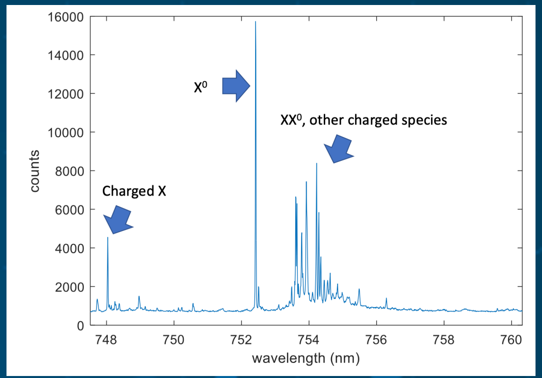
- **Lithographic patterning:**



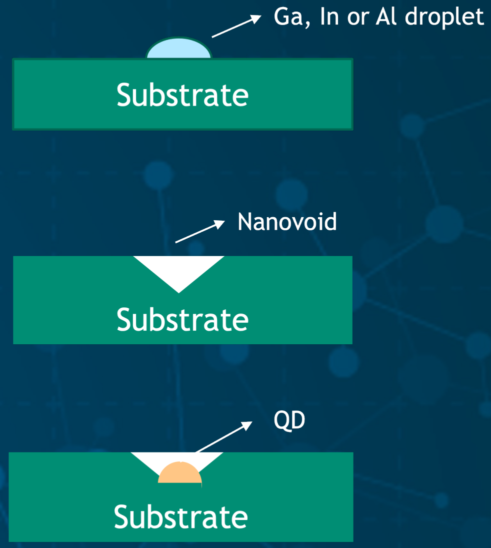
InAs/GaAs QDs grown on (a) unpatterned and (b) ex-situ patterned substrate

  - Pattern controls site, shape, size of the QD
  - Wavelength could be tuned by varying QD material
  - **Drawback**: ex-situ patterning introduces contamination and interface issues

- **In-situ etching:**
  - Group III-droplet assisted etching
  - Defect and impurity-free nanopatterning
  - High symmetry QDs – ideal for SPEs
  - Even lattice-matched QDs grown



Typical PL spectrum for GaAs/AlGaAs LDE QDs

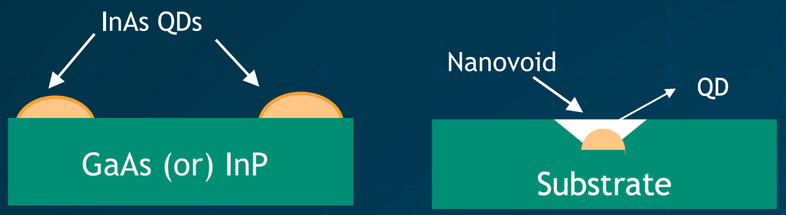


Schematic showing LDE growth process

  - **Drawback**: Complicated/sensitive growth process

Kiravittaya et al. (2004)

Approach

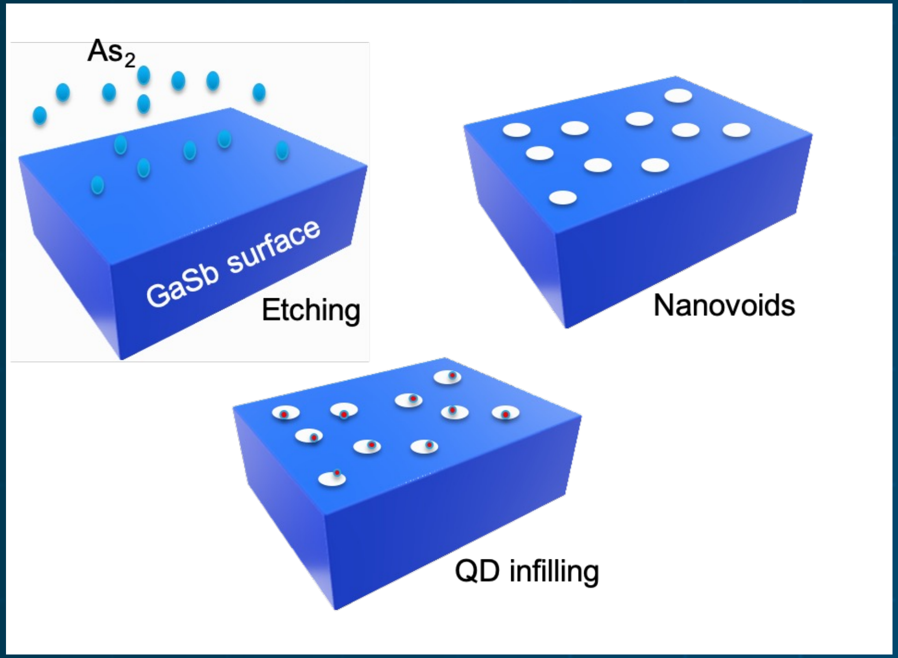


Schematic showing S-K and LDE QD growth

- S-K QDs work well → limited by formation mechanism
- QD growth in pre-defined patterns solves S-K issues
- In-situ patterning is better, but, LDE is a complex growth process

- **Best way forward:**
  - Use in-situ patterning – control over size & shape of QDs
    – NO material choice constraints
  - Alternative patterning/ QD growth process:

Arsenic-induced displacement etching of antimonide surfaces + infilling



Possible mechanism for As_2-induced etching + QD formation

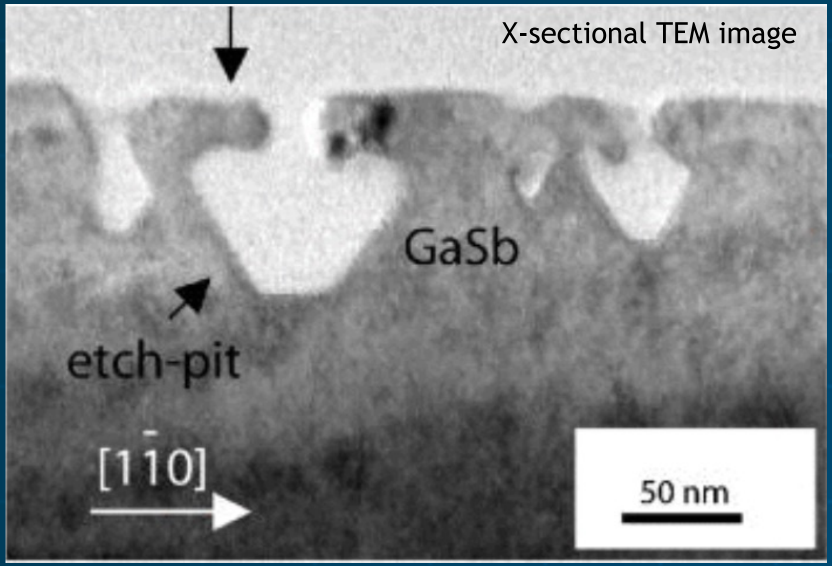
Background

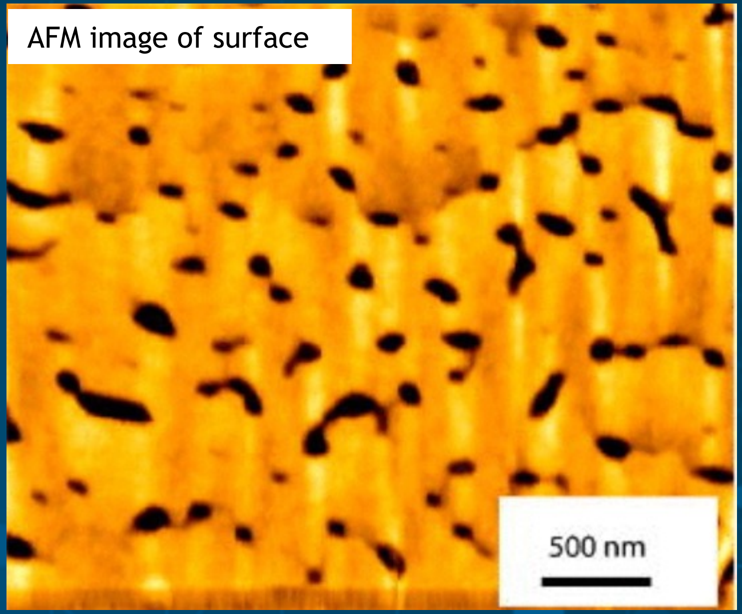
- As_2 reacts aggressively with GaSb surfaces through two reactions:

$$2GaSb + As_2 \rightarrow 2GaAs + Sb_2 \qquad \Delta H^0 = -47.6kJ/mol$$ (Anion exchange Surface AFM

$$GaSb + As_2 \rightarrow GaAs + AsSb \qquad \Delta H^0 = -33.9kJ/mol$$ (Isoelectronic AsSb formation)

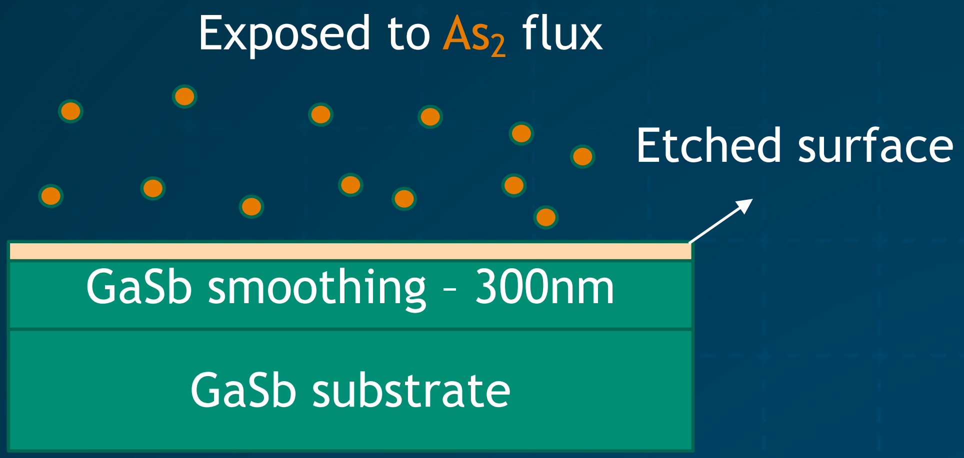
- Nanovoid formation observed previously on GaSb surfaces exposed to As_2 flux – not used for infilling

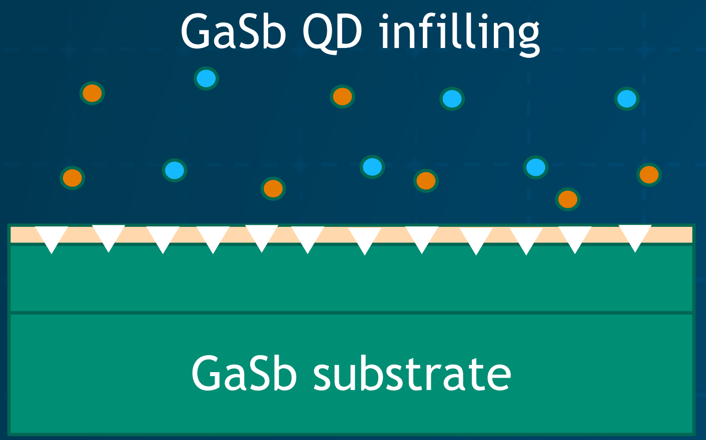




Losurdo et al. (2006) ; Huang et al. (2007)

Experiment: structural





- **Procedure:**
  - GaSb native oxide desorption: 540℃ for 30min
  - 300nm GaSb smoothing layer grown – Sb:Ga = 3 at 505℃
  - Excess Sb desorbed from surface
  - Surface exposed to As_2 – varying flux, times & tempature

Nanovoid formation

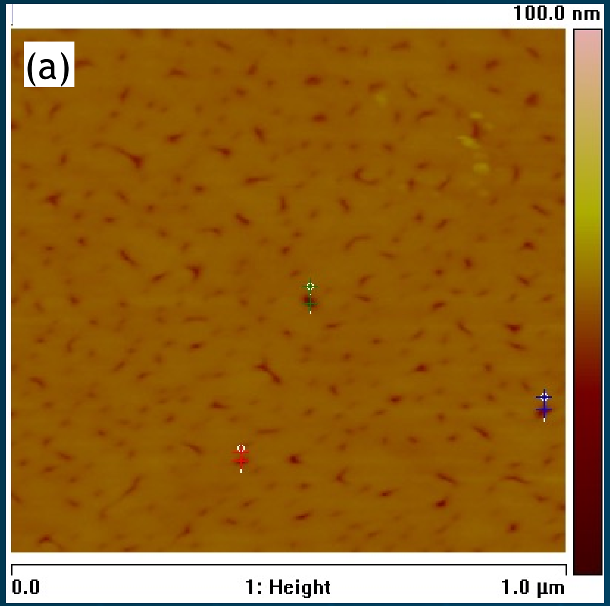
- Sample cooled down under Sb flux

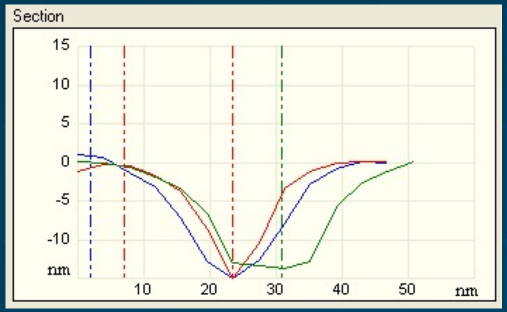
Infilling with QDs

- Sb soak – 5min
- Migration-enhanced GaSb QD growth
- Sample cooled under Sb flux

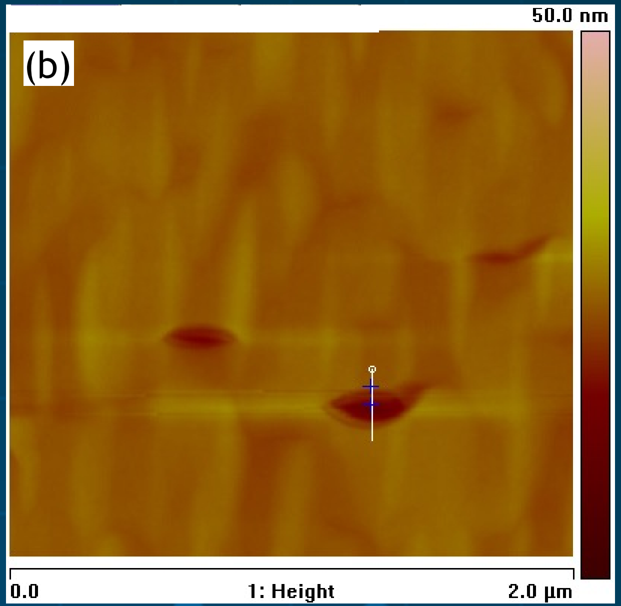
Initial findings

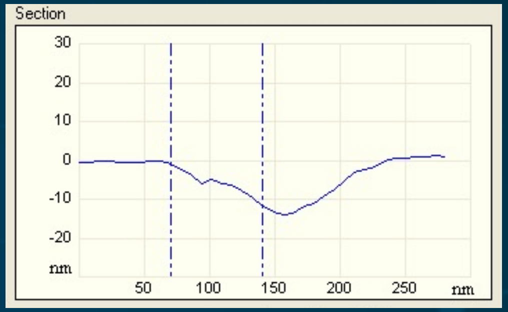
- Prolonged exposure (> 2 min) at high As_2 fluxes (> 1e-6 Torr) → nanovoid formation
- Infilling observed in a relatively low number of nanovoids





Vertical distances (nm):
- 16.58
- 15.21
- 13.72





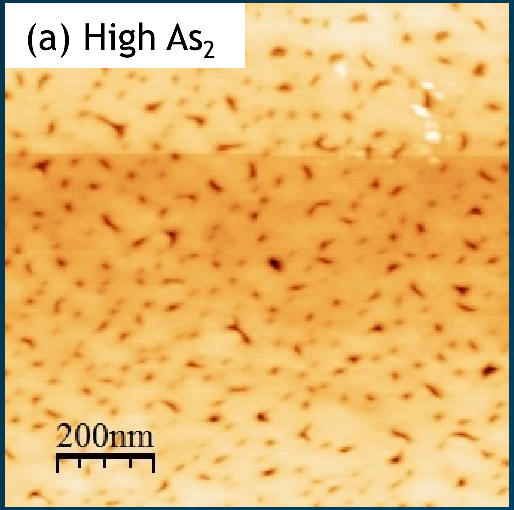
- Depth: ~15nm
- QD height: 7-8nm

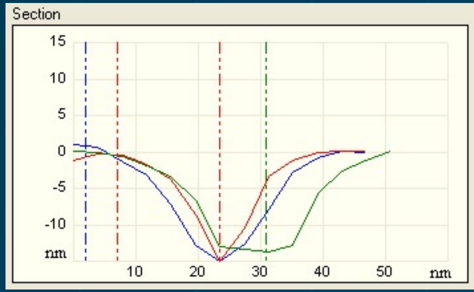
AFM scans of (a) etched nanovoid surface and (b) infilled nanovoid with sectional analysis

- Nanovoids show LOW uniformity in size & shape
- An extended study is carried out to determine if growth conditions influence:
  - Nanovoid uniformity, size and shape
  - Infilling of voids

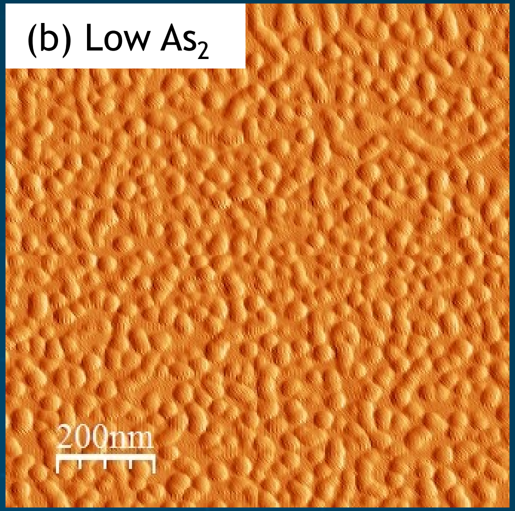
Structural characteristics

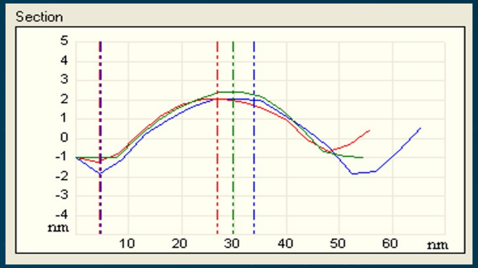
- Nanovoid etch mechanism highly dependent on As_2 exposure – both flux and time





Nanovoid dimensions:
- Width : ~40nm
- Height : ~15nm





"QD" dimensions:
- Width : ~25nm
- Height : ~3nm

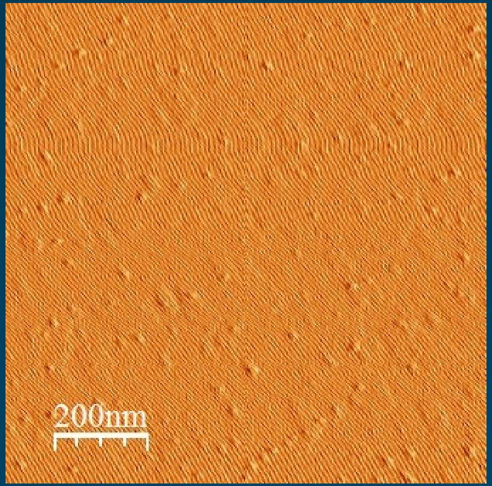
1X1 µm AFM image of (a) high and (b) low As_2 (flux and time) etched GaSb surface

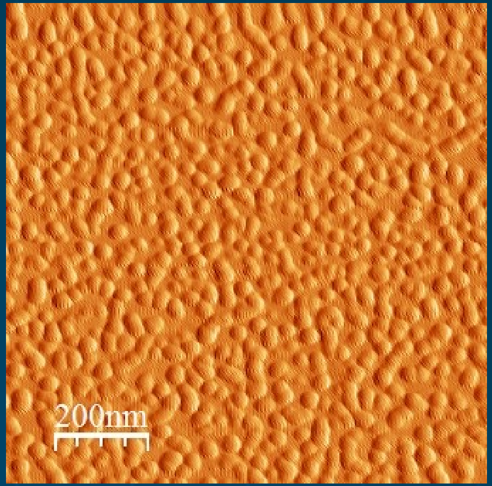
- **High As_2 → Nanovoids**
  - High density
  - Non-uniform void sizes and profiles
- **Low As_2 → "QDs"**
  - High density
  - Profiles show similar sizes

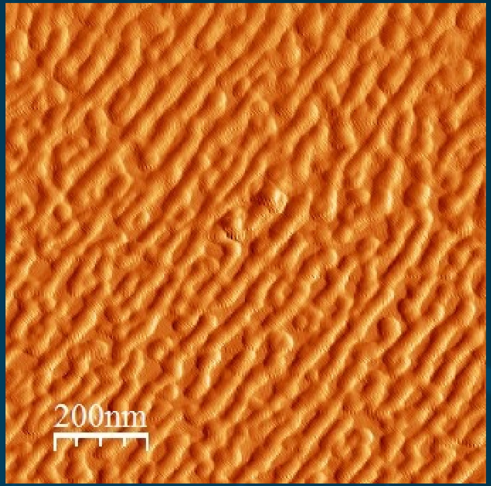
Structural characteristics

- As_2 exposure (controlled by flux, time or growth temperature) determines etch mechanism

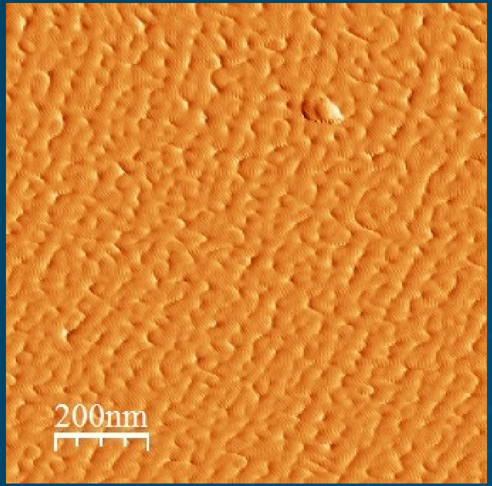
Increasing As_2 exposure

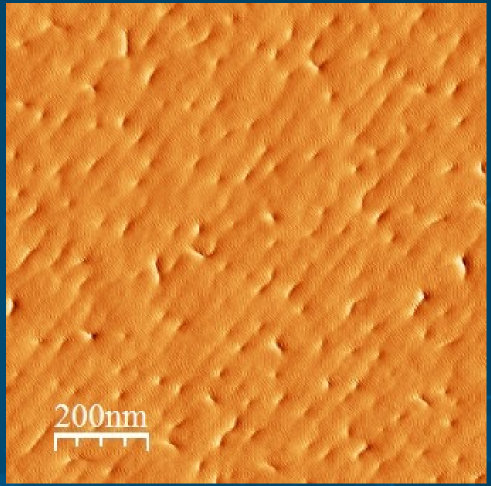


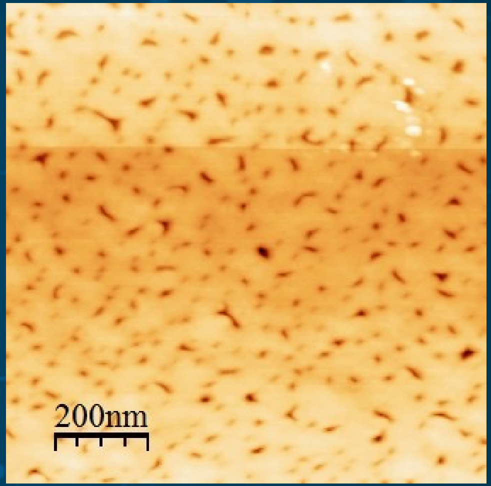




1X1 μm AFM image of etched GaSb surface with varying As_2 exposure



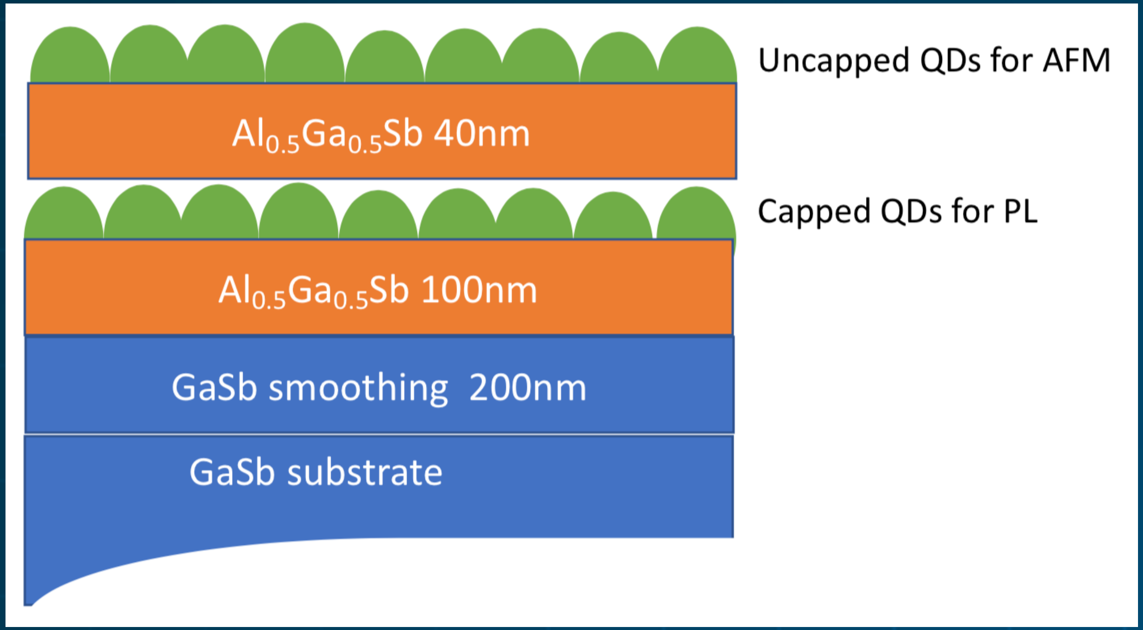




- In “QD” regime, density can be controlled
- With increasing As_2, nanostructures coalesce before void formation
- For further studies, high-density “QD” conditions used

Experiment: optical

- QDs embedded in higher bandgap material ($Al_{0.5}Ga_{0.5}Sb$) for analyzing optical signature
  - Also for x-sectional and composition analysis using TEM & SIMS

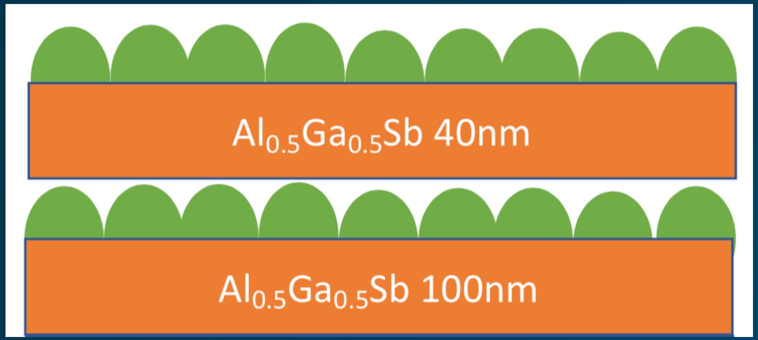


Schematic showing structure used for optical studies

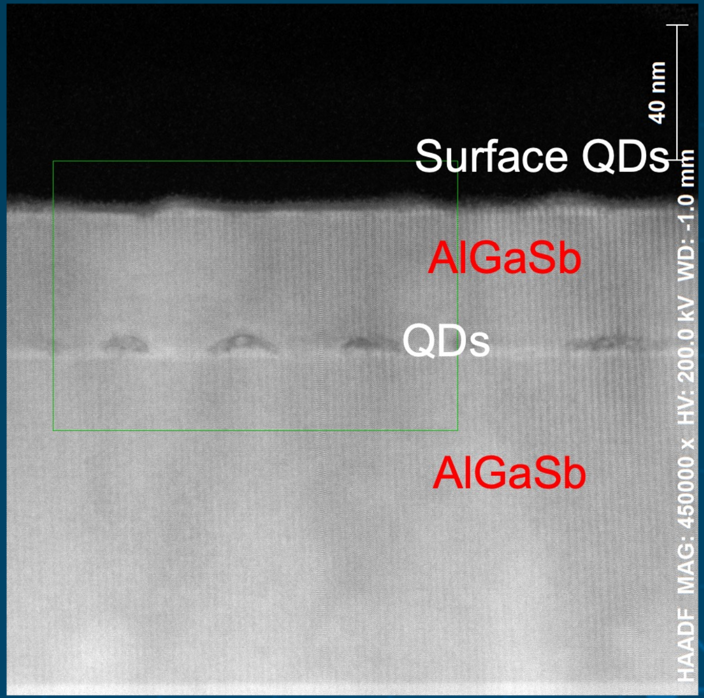
- **Procedure:**
  - GaSb native oxide desorption: 540°C for 30min
  - 300nm GaSb smoothing layer grown – Sb:Ga = 3 at 505°C
  - 100nm $Al_{0.5}Ga_{0.5}Sb$ barrier layer grown
  - Excess Sb desorbed from surface
  - Surface exposed to As_2 - flux, time and temperature adjusted for QD growth
  - 5 min Sb soak
  - QDs buried in a 40nm $Al_{0.5}Ga_{0.5}Sb$ layer
  - QD growth process repeated on the top surface for AFM measurements.

X-sectional analysis using TEM

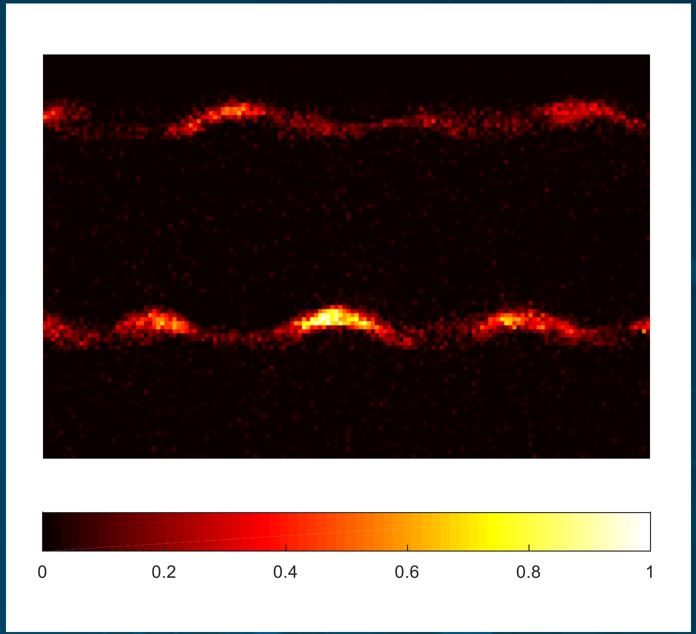
- TEM analysis shows spatially separated 3-dimensional nanostructures with a mostly GaAs(Sb) composition



Schematic showing QDs & surrounding material



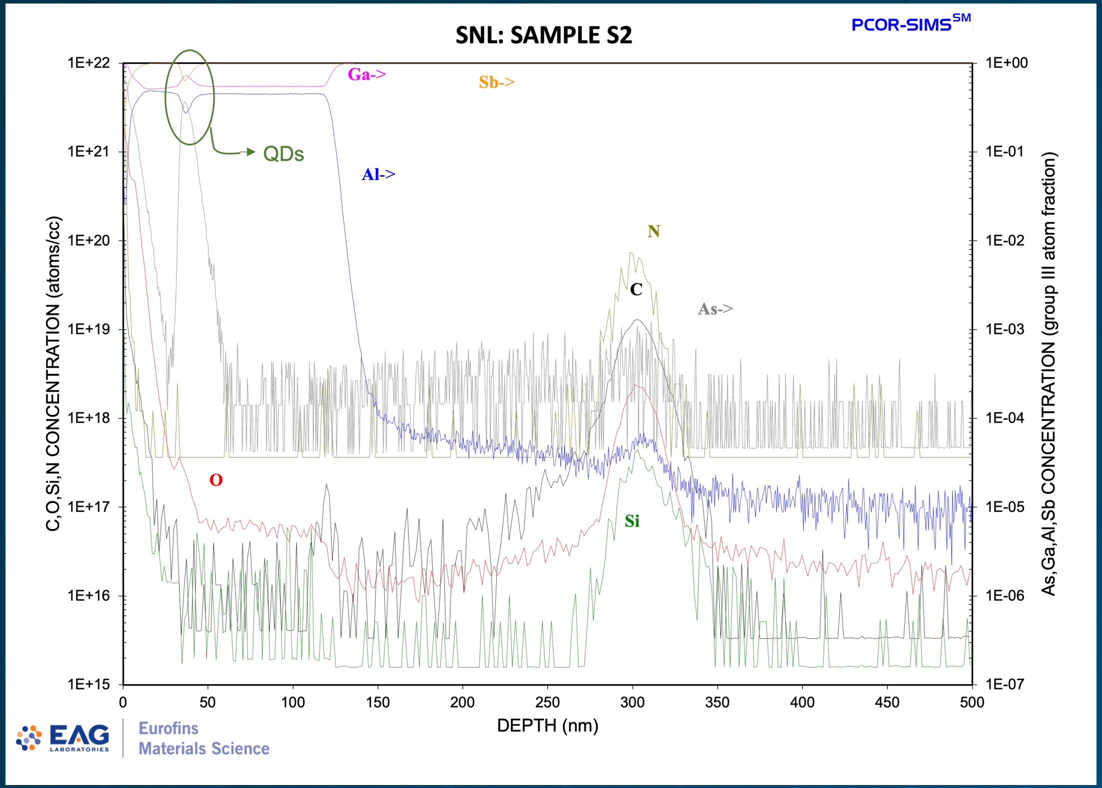
HAADF STEM image of QDs and surrounding material



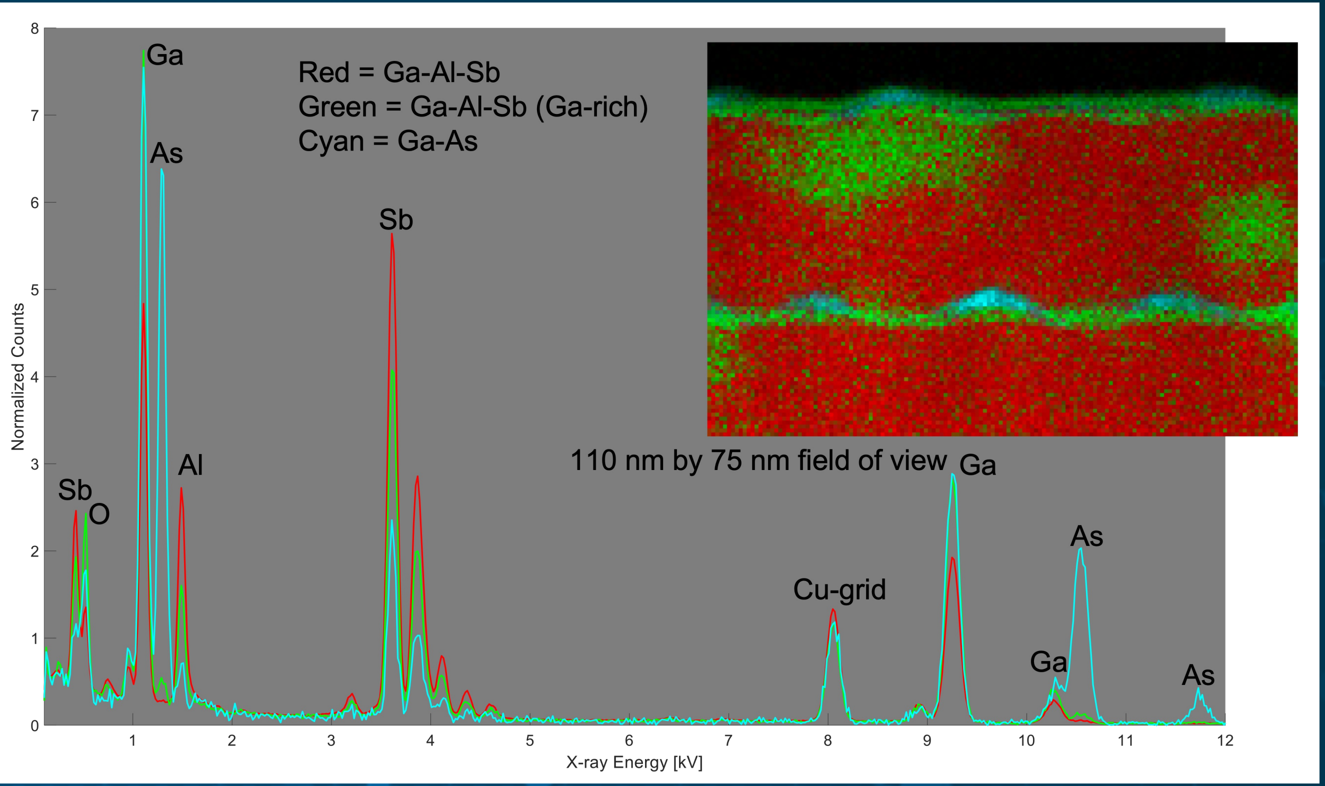
Ga-As component image

Material composition (SIMS & TEM)

- Both SIMS and EDS analysis suggest a QD composition of GaAs with low % of Sb



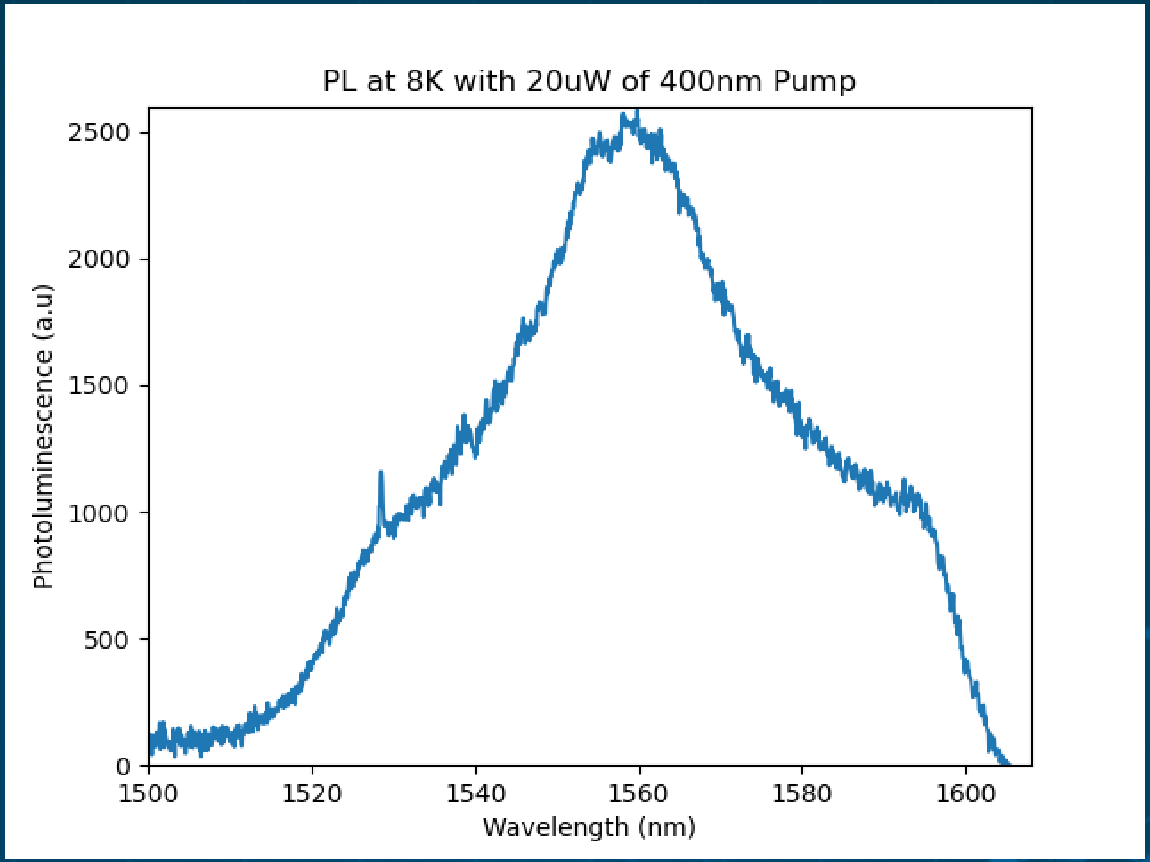
SIMS profile from GaAs(Sb) QD PL sample



EDS analysis of GaAs(Sb) QDs and surrounding material

Optical characteristics

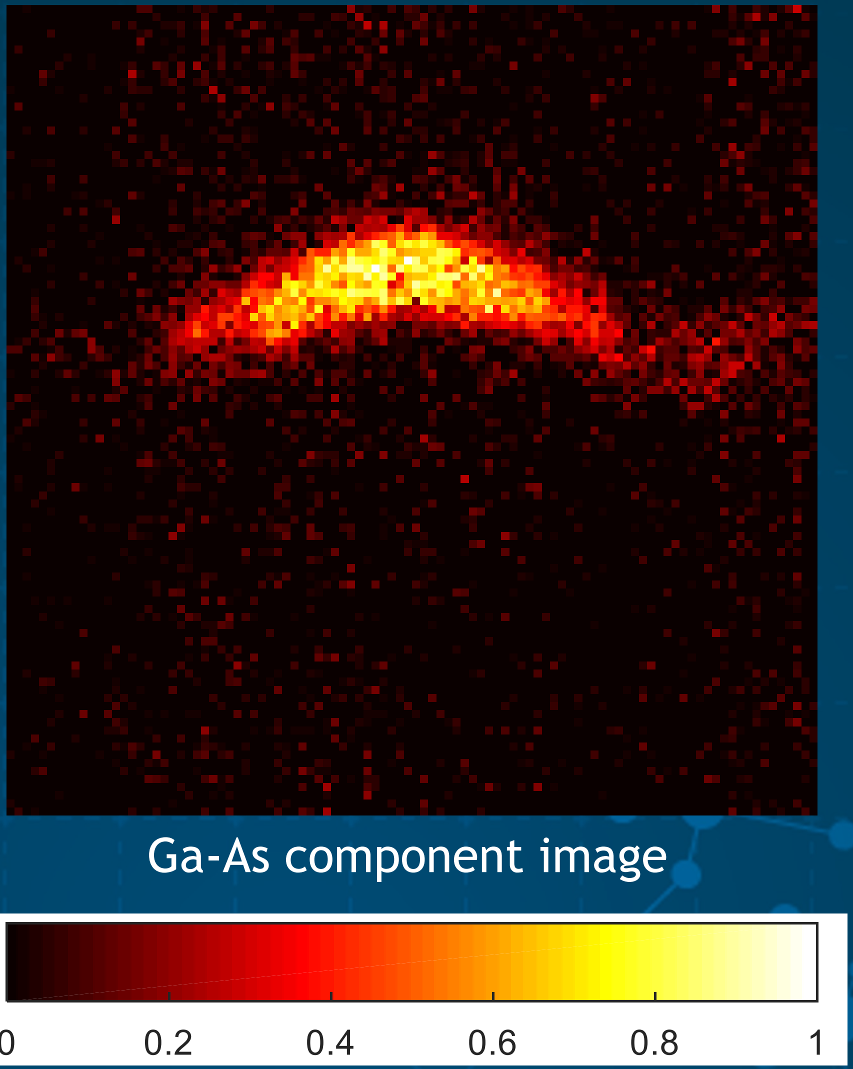
- Preliminary optical studies carried out
- Extended analysis ongoing including PL comparison between etched and non-etched GaAs(Sb) samples



Conclusions

- Arsenic-induced in-situ etching of III-Sb surfaces explored as a QD formation mechanism
- Growth conditions for nanovoid formation + QD infilling determined
  - High As_2 exposure (controlled by flux, time and temperature) leads to nanovoid formation
  - Nanovoids (both before and after infilling) show high nonuniformity in size & shape
- Low As_2 exposure results in 3-dimensional nanostructure (QD) formation with As_2 flux controlling QD density and size with high uniformity.
- Cross-sectional TEM analysis shows spatially separated QDs and coupled with SIMS reveals the composition to be GaAs(Sb).
- Preliminary optical analysis shows a distinct optical signature ~1.55μm (8K measurement)

Questions?



Ga-As component image