

UNIVERSITÉ
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STEM Cathodoluminescence of Individual GaN/AlN Quantum Disks within a single Nanowire

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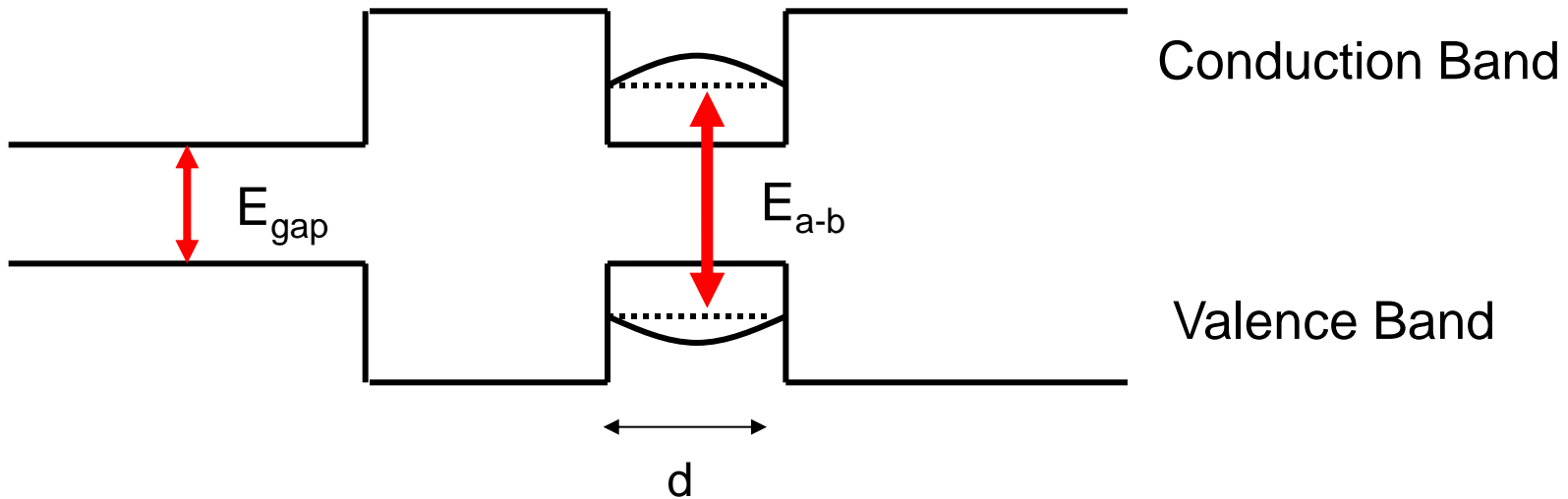
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Introduction

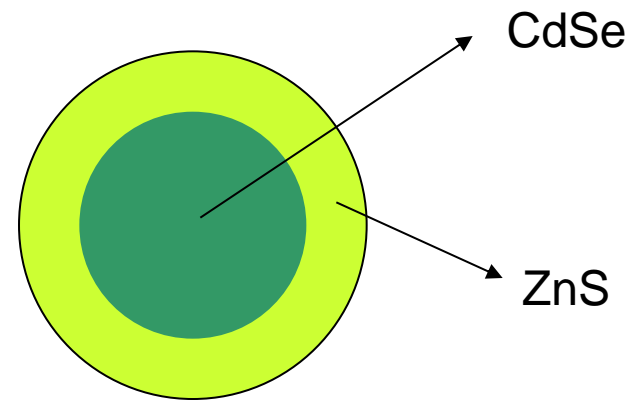
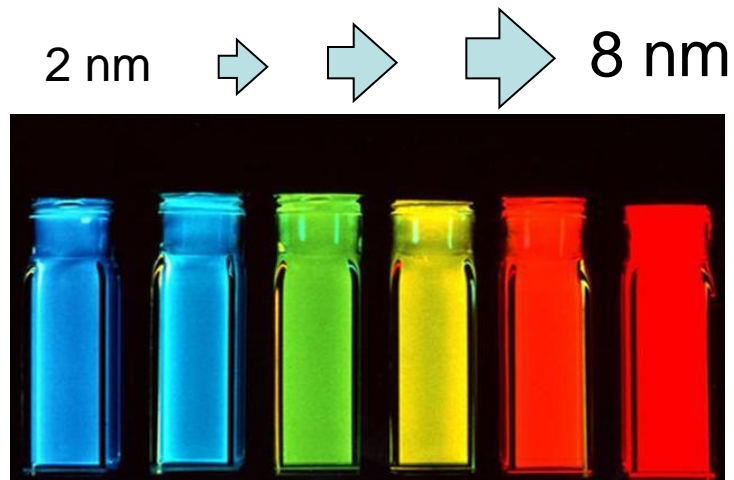
Quantum Confinement



Correlation of quantum object dimension and its transition energy.

Introduction

CdSe nanoparticles

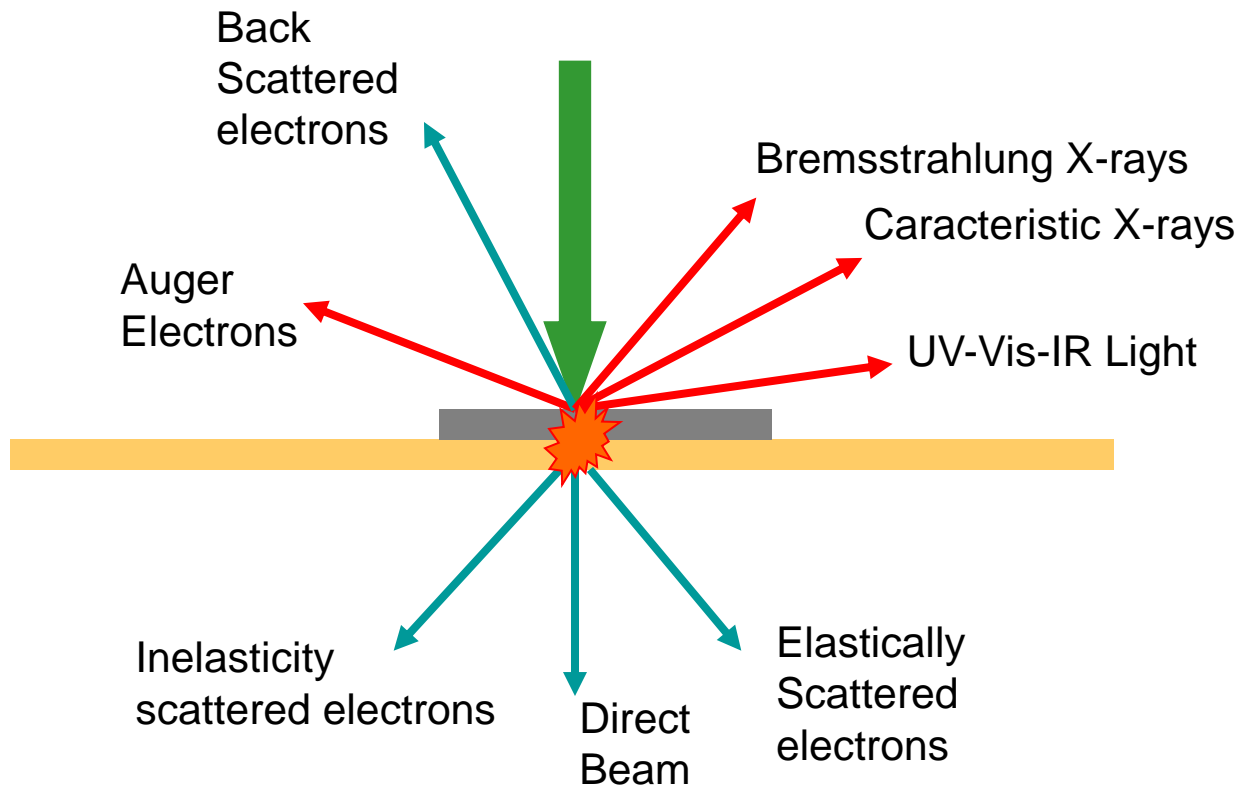


M. G. Bawendi

J. Phys. Chem. B **1997**, *101*, 9463–9475

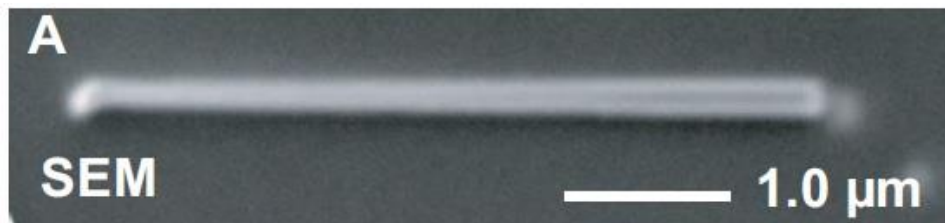
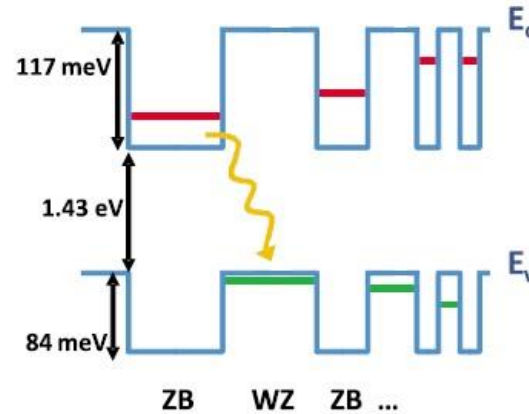
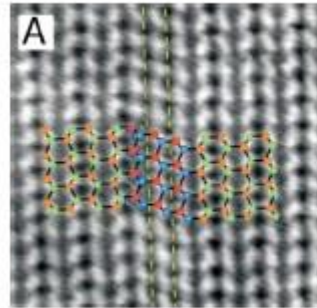
Cathodoluminescence

Electron – Matter interactions



Cathodoluminescence

zinc-blende/wurtzite
GaAs nanowire
heterostructures

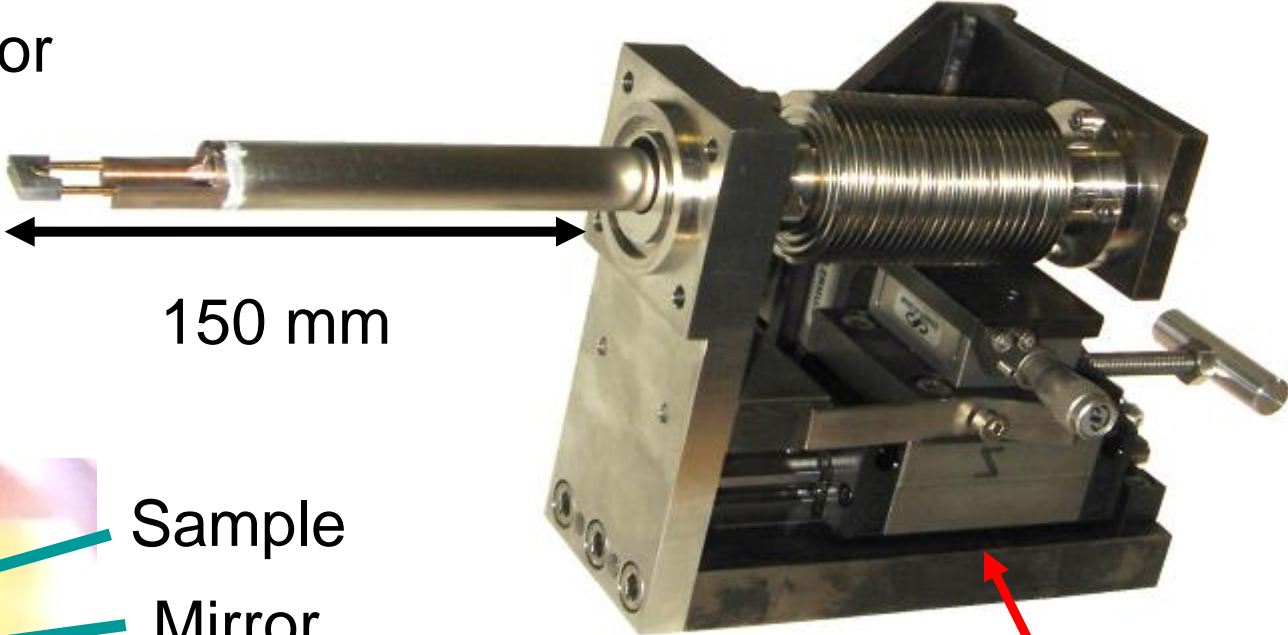


D. Spirkoska et al., PHYSICAL REVIEW B **80**, 245325 2009

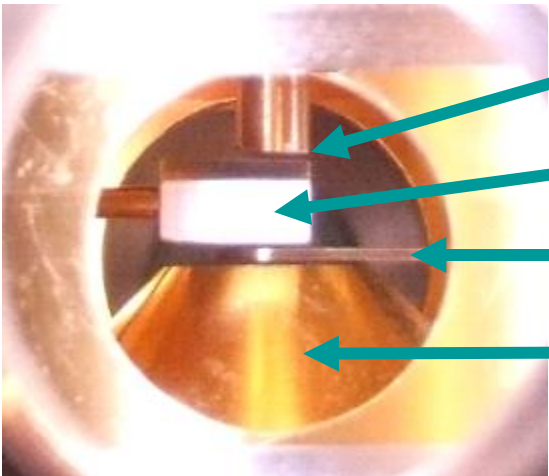
Cathodoluminescence System

Positioning the mirror is crucial to its efficiency.

Parabolic Mirror



150 mm



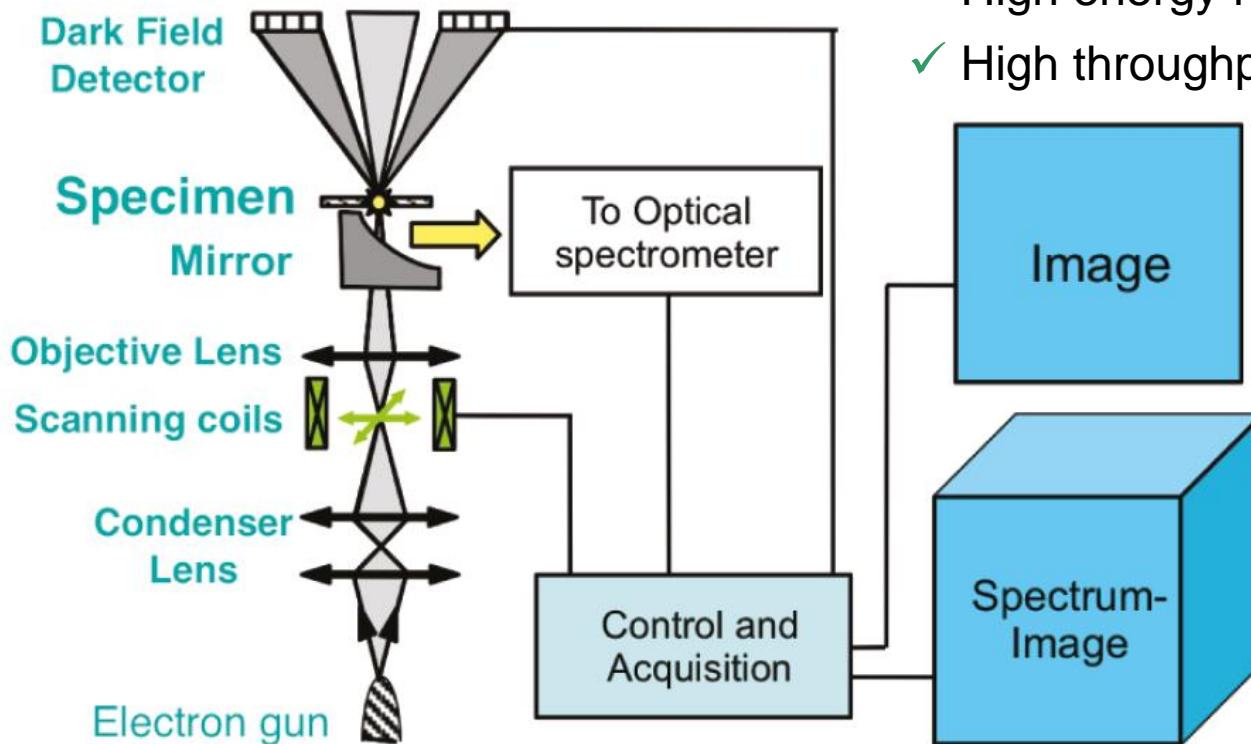
Sample
Mirror
Aperture
Objective Lens

Micrometric screws for 3D Position adjustment

Cathodoluminescence System

An Optimized CL detector:

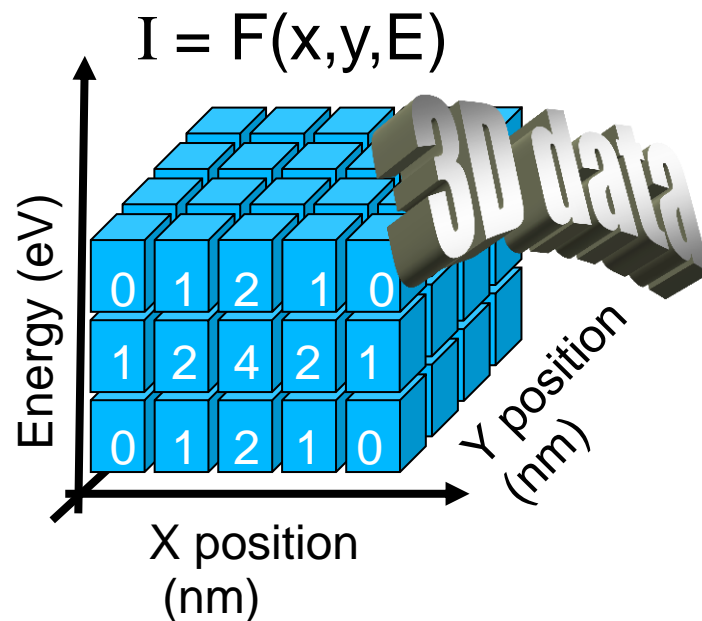
- ✓ High collecting angle (35% of 4π sr).
- ✓ High energy resolution (~ 5 meV).
- ✓ High throughput.



Cathodoluminescence System

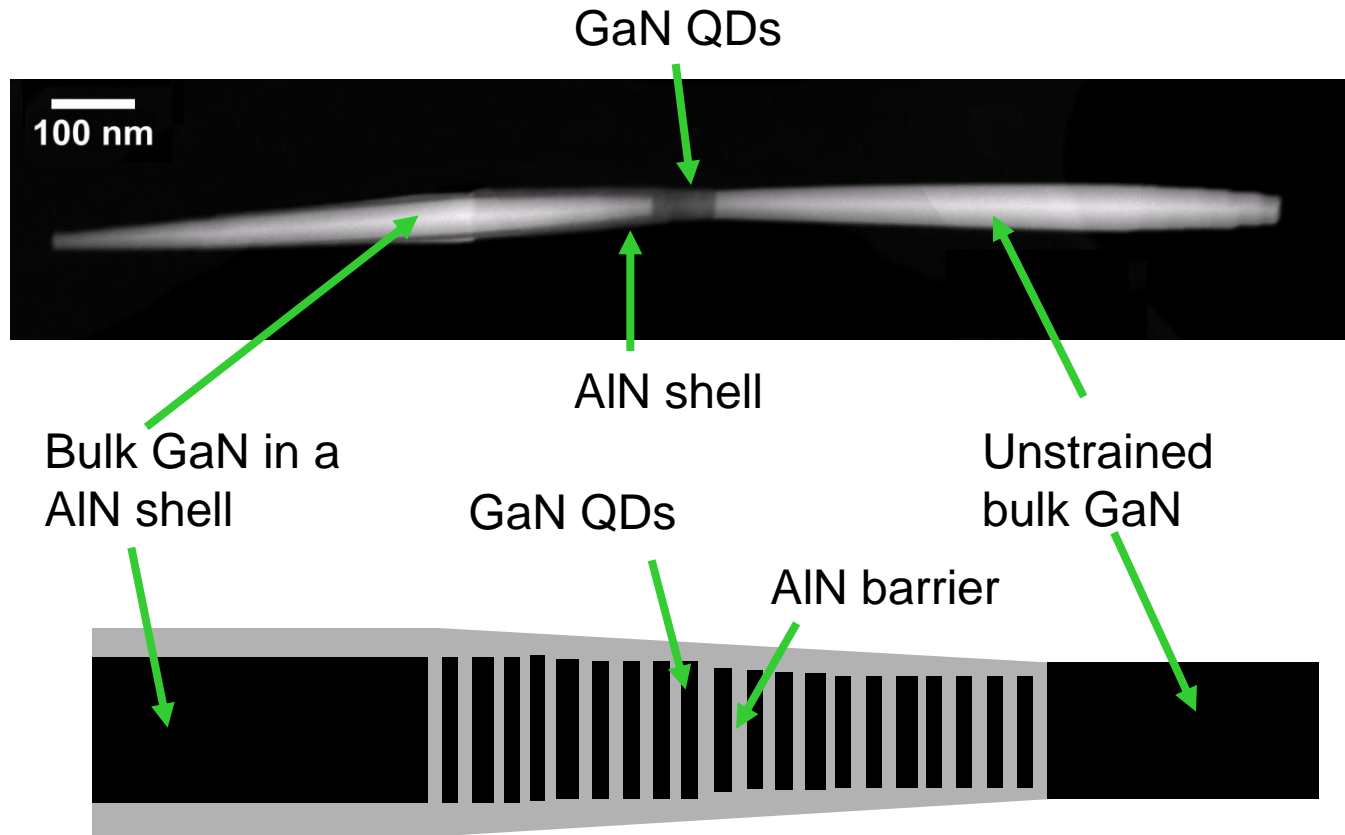
Microscopic Probe – STEM:

- **STEM HB 501 by VG operating at 60kV**
- Spatial resolution of <1 nm.
- Probe current of about ~200pA.
- Liquid Nitrogen Sample stage (150K).
- 3D Spectrum-Image acquisition.

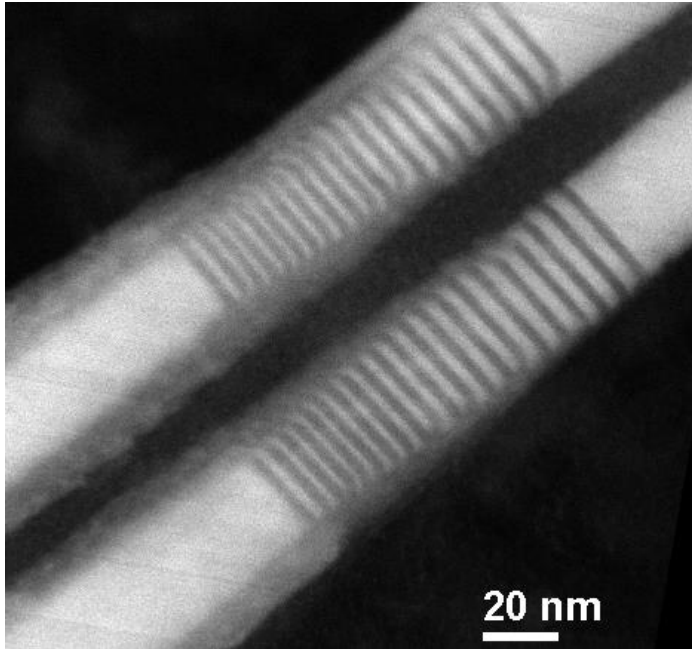


Sample: GaN quantum discs

GaN nanowires with GaN quantum discs with AlN barrier and shell.



Sample: GaN quantum discs



Catalyst-free growth on Si (111)
(RF-PA-MBE).

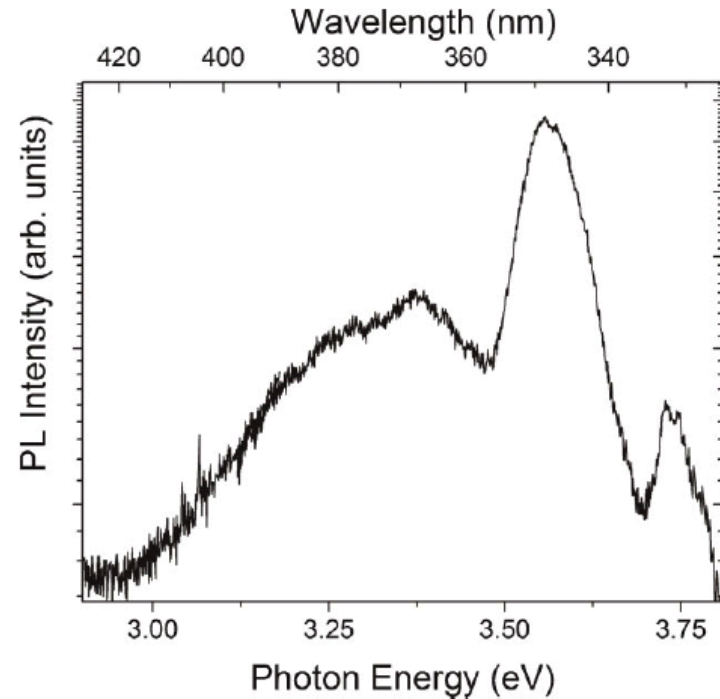
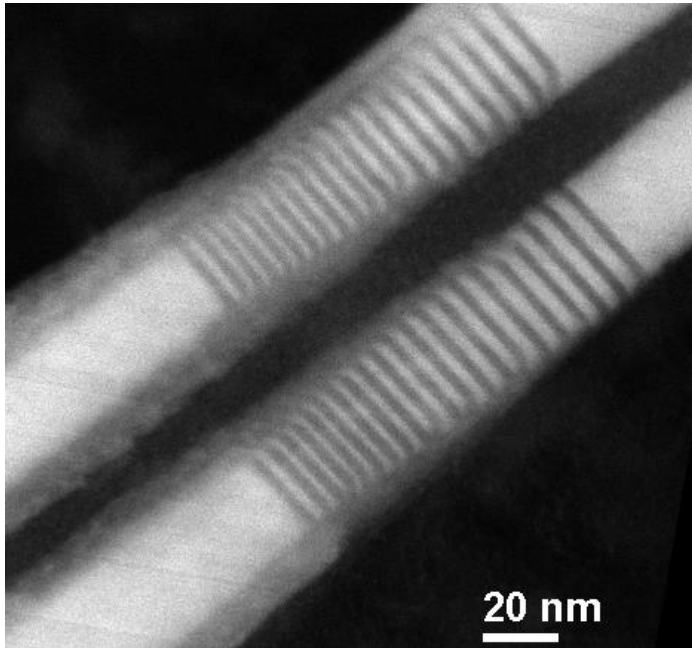
GaN nanowires with GaN
quantum discs with AlN barrier
and shell.

Nanowire length = $\sim 2.1 \mu\text{m}$
Nanowire diam. = 20 to 50 nm

Number of QD's = 20.

Disc Thickness: $\sim 3.5 \text{ nm}$
Barrier Thickness: $\sim 4.1 \text{ nm}$

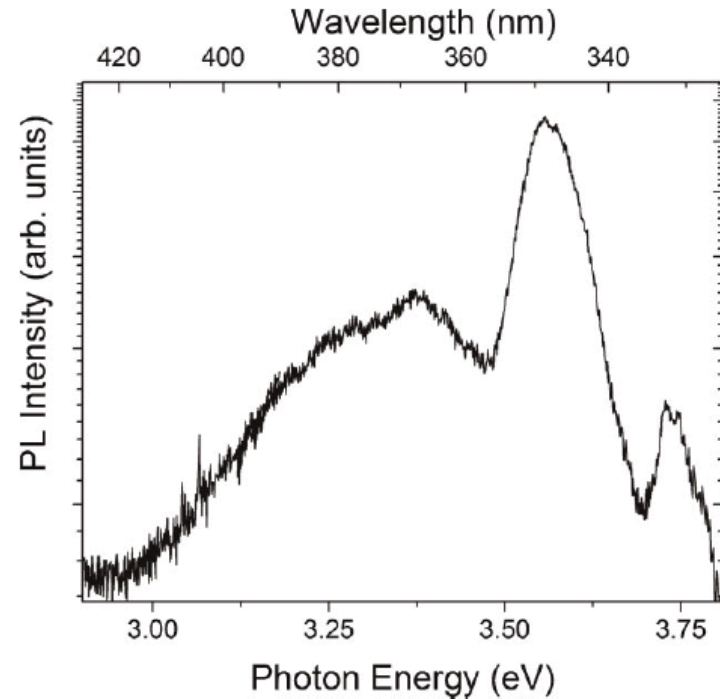
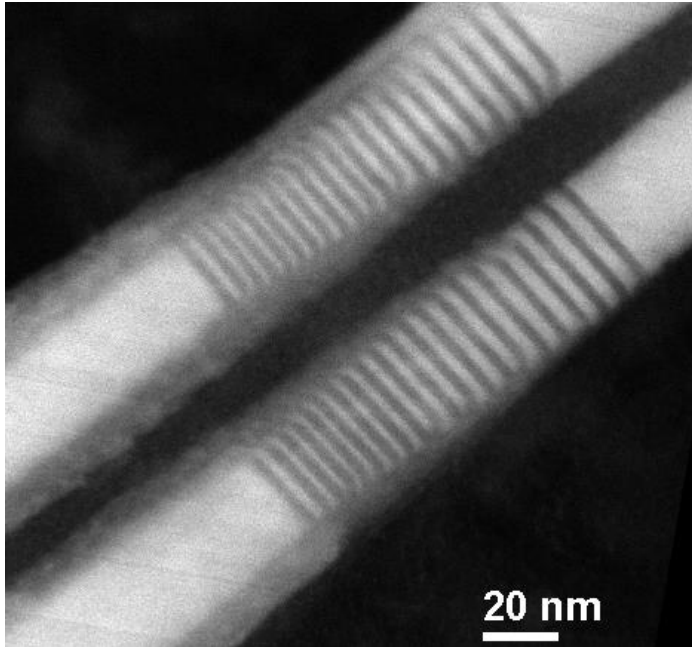
Sample: GaN quantum discs



Emissions below and above the GaN Band gap

L. Rigutti et al. Nano Lett. 2010, 10, 2939–2943

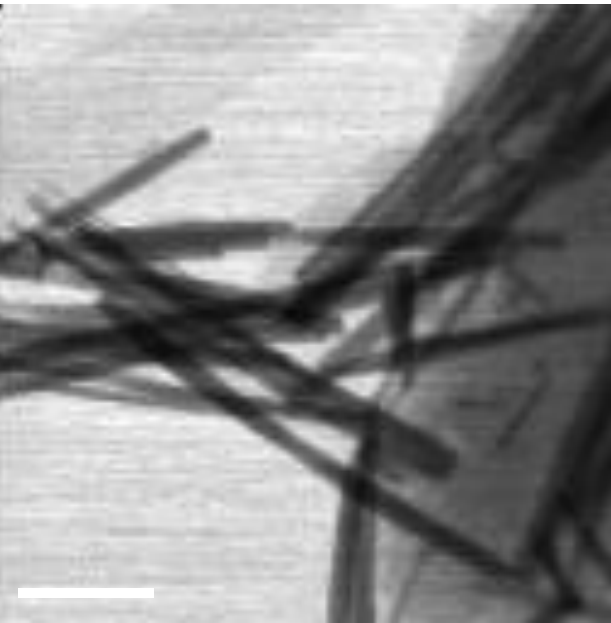
Sample: GaN quantum discs



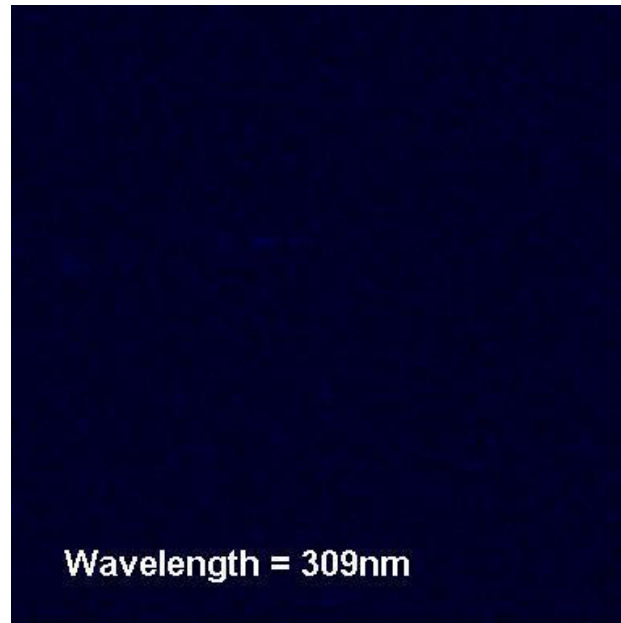
Where do they come from?

L. Rigutti et al. Nano Lett. 2010, 10, 2939–2943

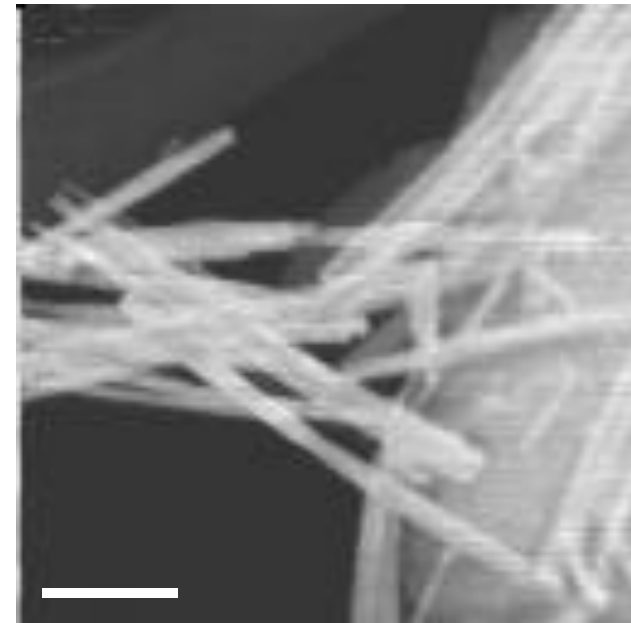
GaN Quantum discs



Bright field image

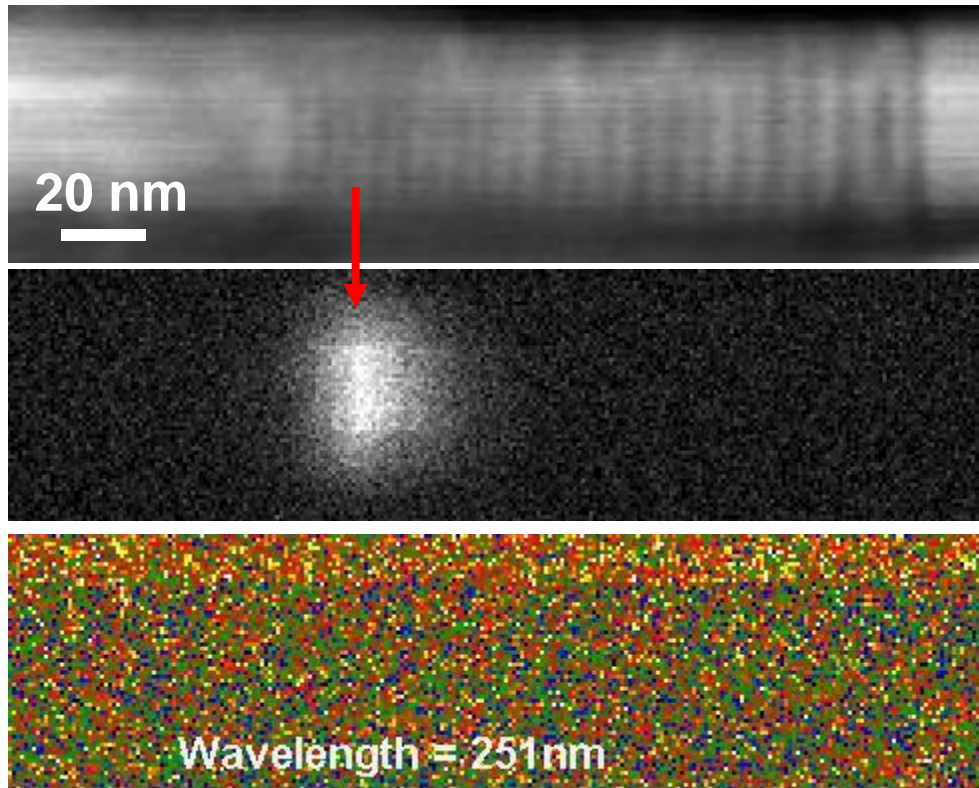


CL Spectrum Image



Dark field image

GaN quantum discs



HADF acquired simultaneously with CL spectrum-image

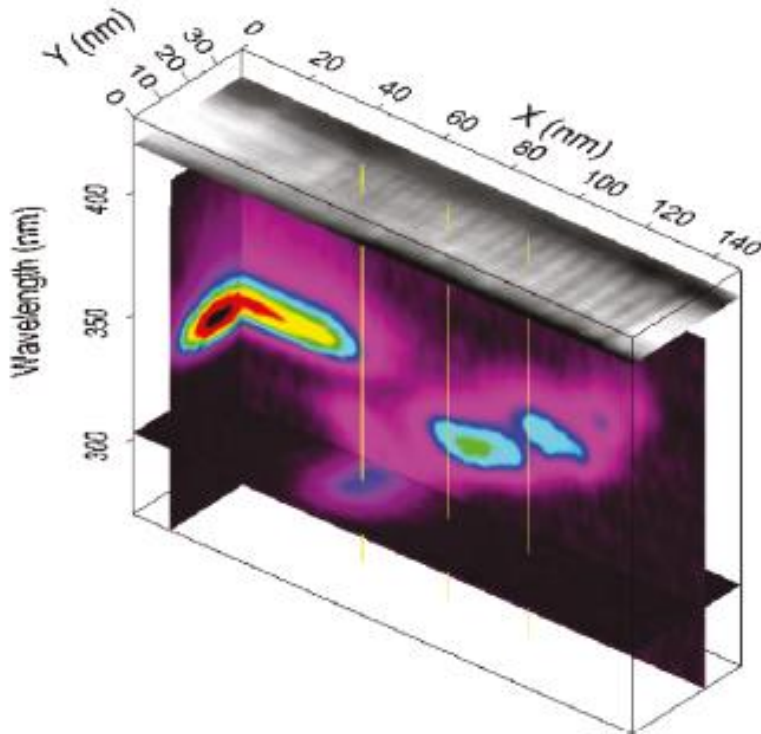
Monochromatic image extracted from the Datacube at 300 nm.

Datacube from 271 to 390 nm.

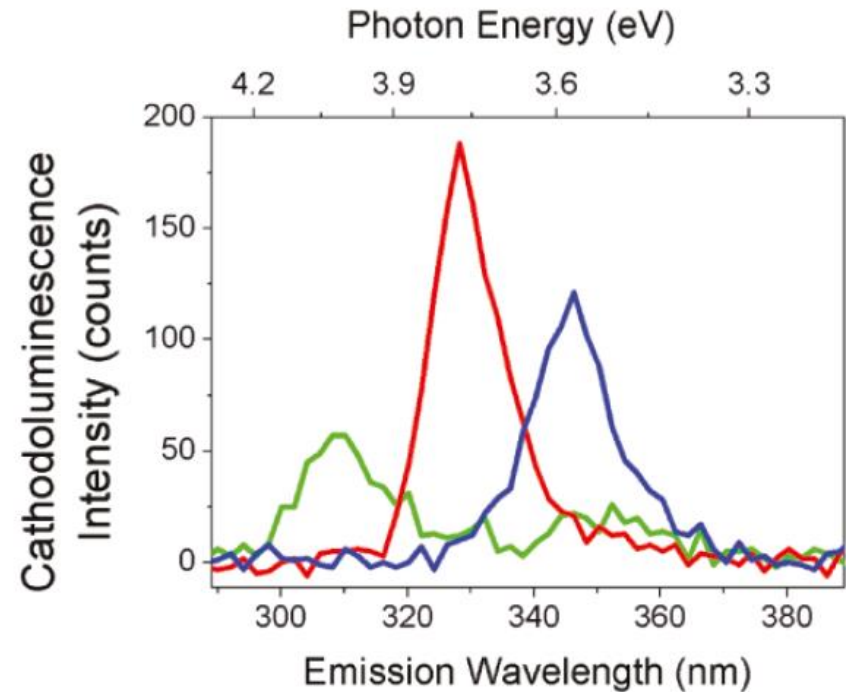
Spatial sampling: 0.6 nm per pixel
Spectral sampling: 2 nm per pixel
Dwell time: **20 ms per pixel**

Spectrum image size: 256 x 64 x 256 pixels

GaN quantum discs

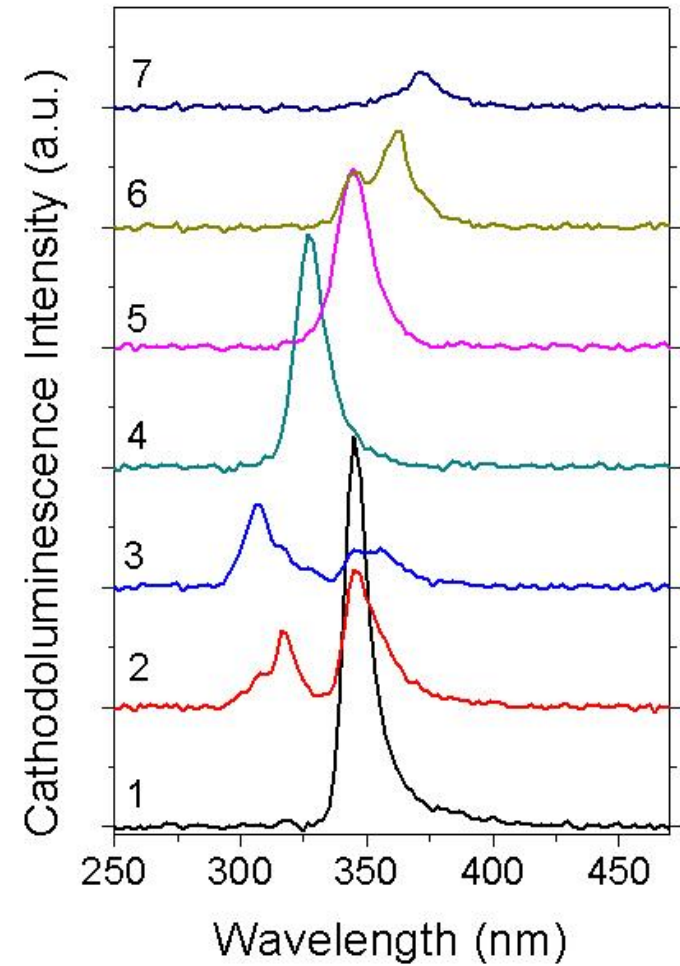
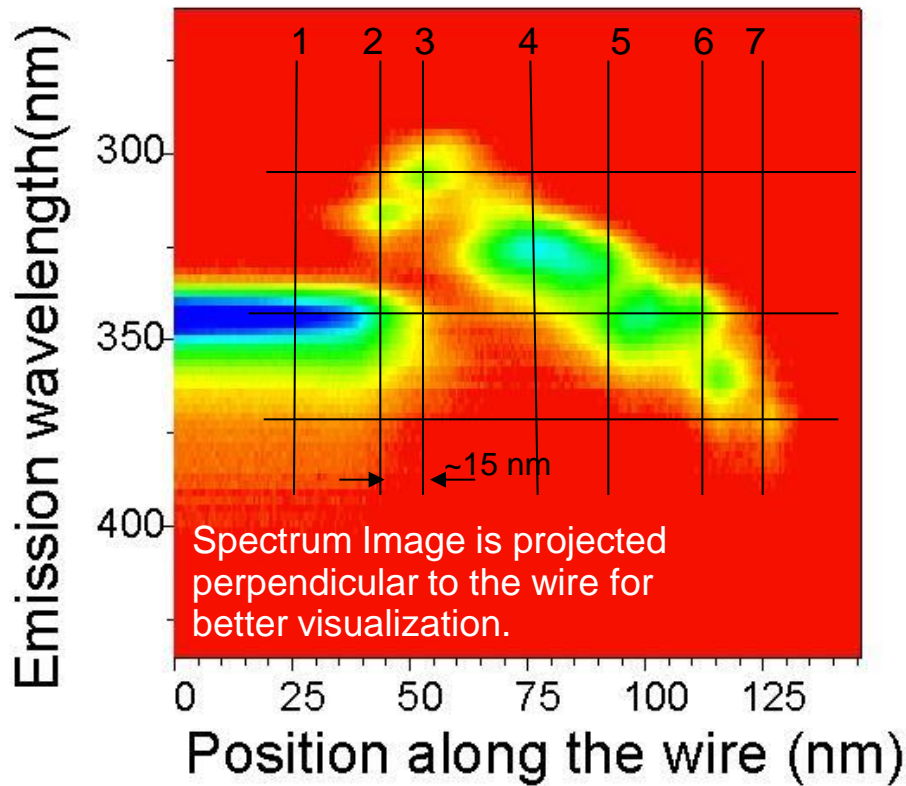
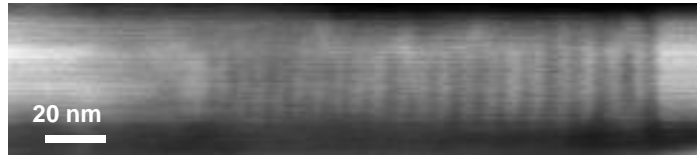


HAADF Image and the cathodoluminescence datacube.

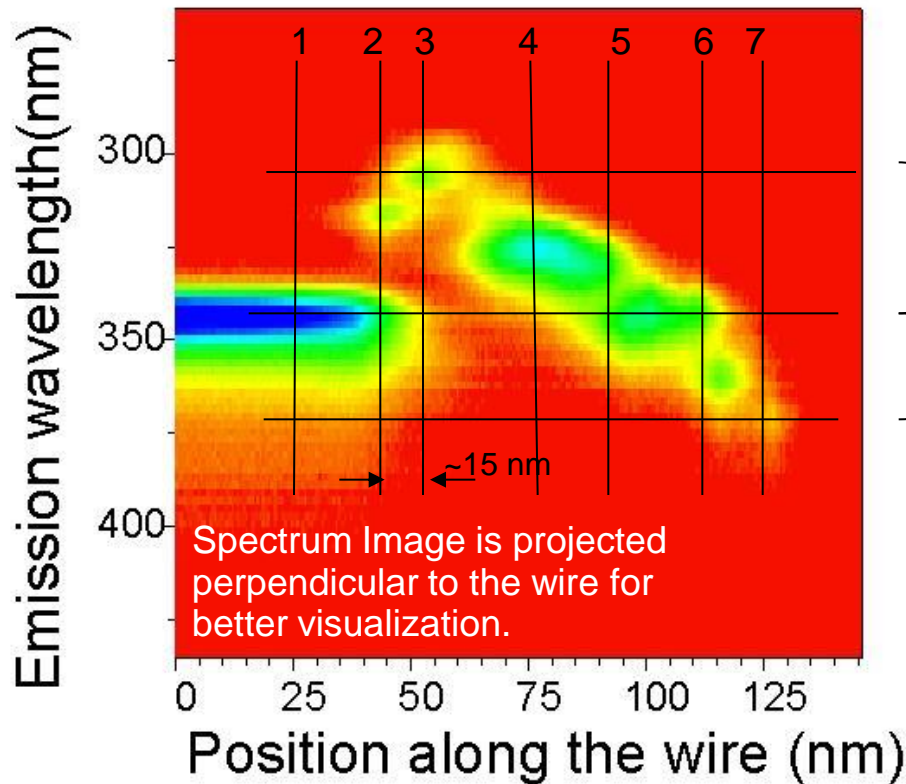
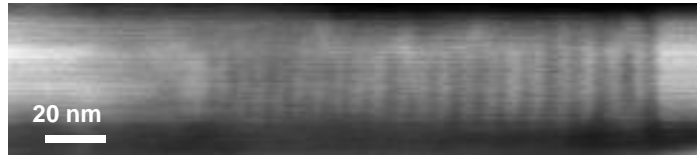


Single pixel spectra.

GaN quantum discs



GaN quantum discs



Only the QDs and the bulk GaN are emitting.

Energy higher than Gap Band Gap:
Dominated by Quantum Confinement

Energy lower than Gap Band Gap:
Dominated by Stark Effect

Nanowire Morphology

Quantum confinement effect is more important in smaller discs.

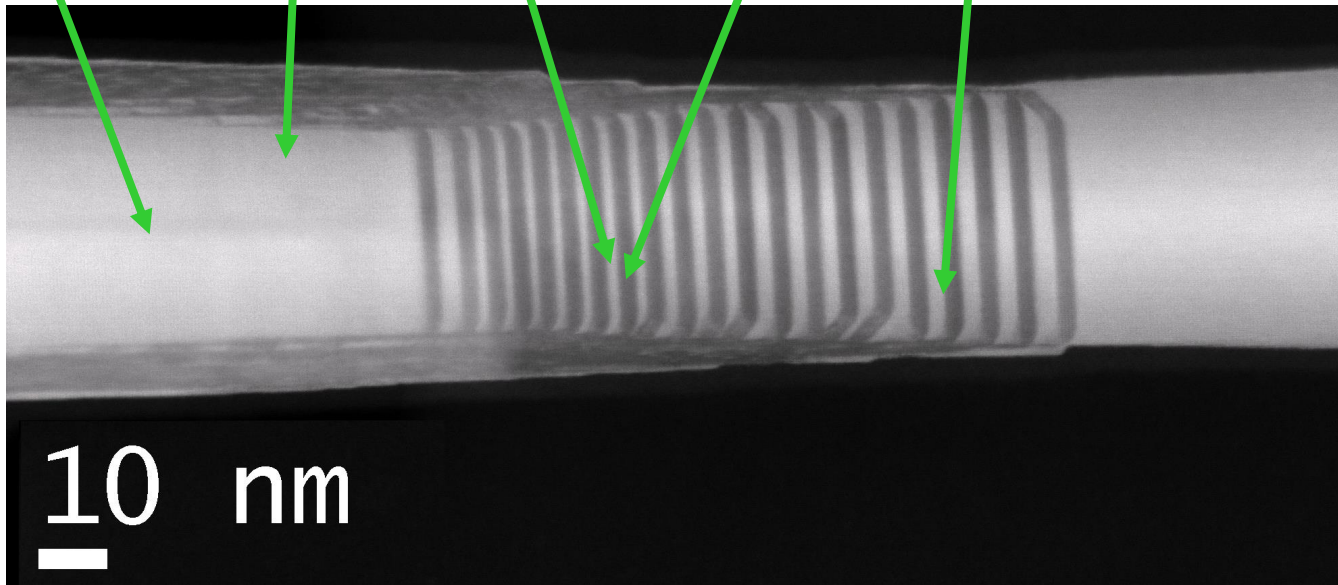
Bulk GaN in a
AlN shell

AlN shell

GaN QDs

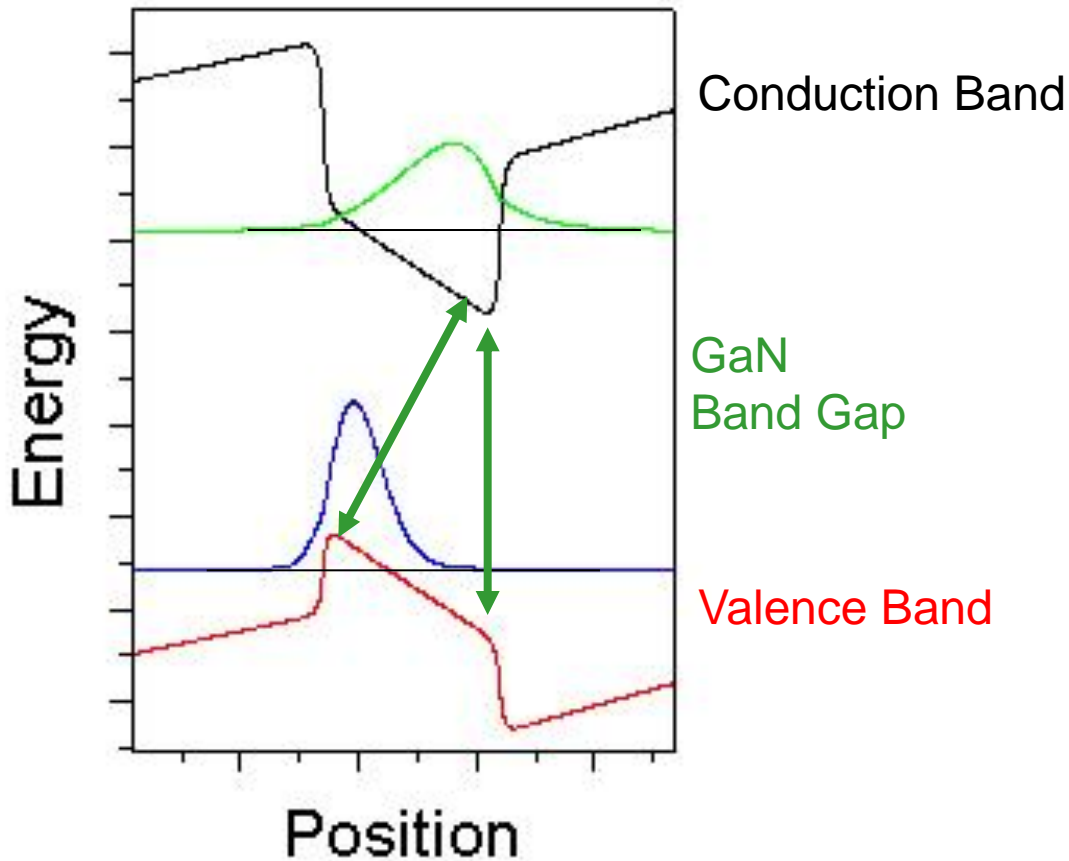
AlN Barrier

Unstrained
bulk GaN



Disc Diameter:
Increases from
1 nm (4 ML)
to 3.4 nm (13 ML)

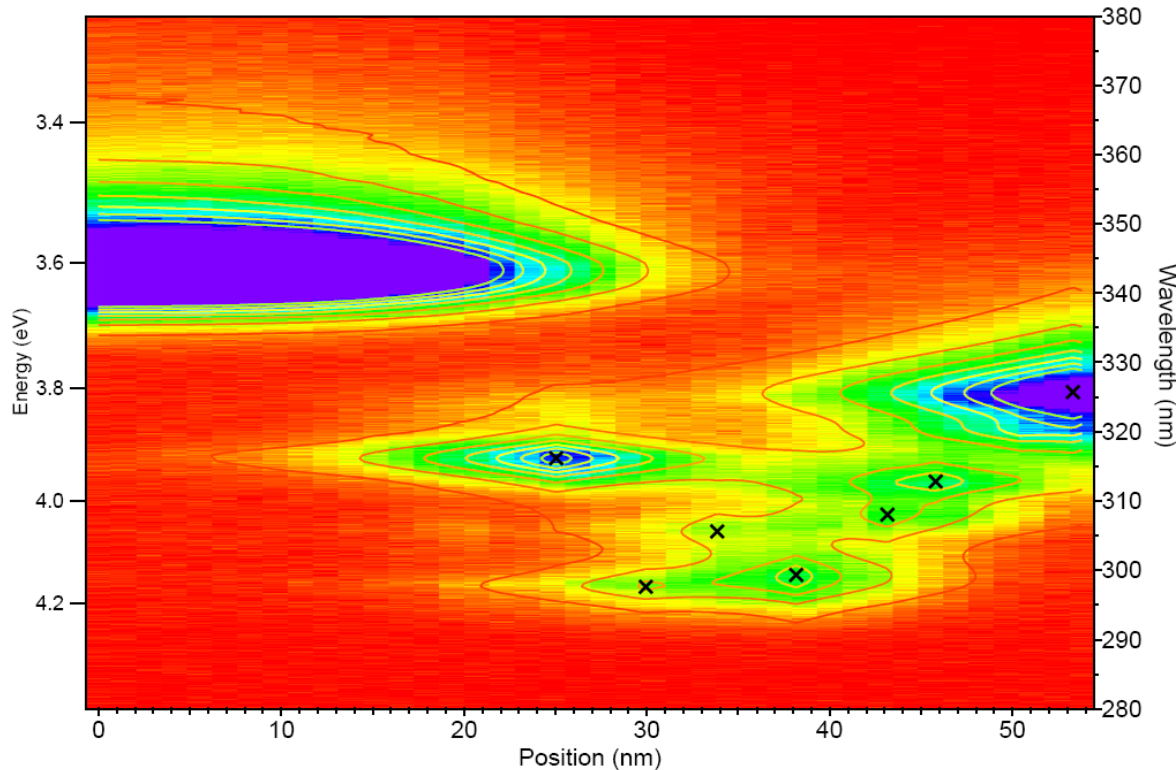
Quantum confinement Stark Effect



Gap GaN = 3.47 eV
Gap AlN = 6.20 eV

The band bending due to internal electric field is the origin of the observed QCSE and therefore the redshift emission for large QDs.

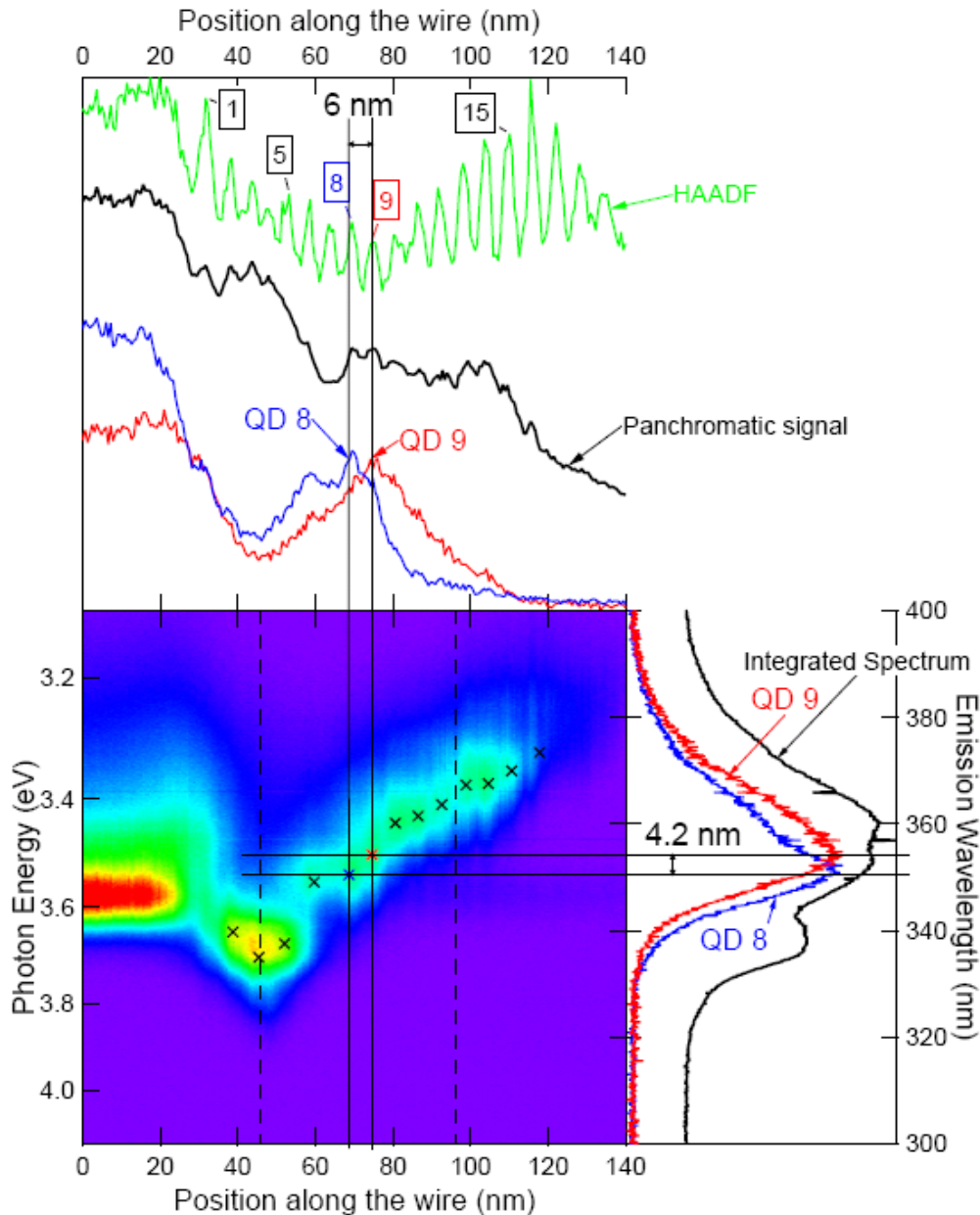
Identifying single QD emissions



The partial overlapping of all QD emission can be distinguished by fitting.

**L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573**

Identifying single QD emissions

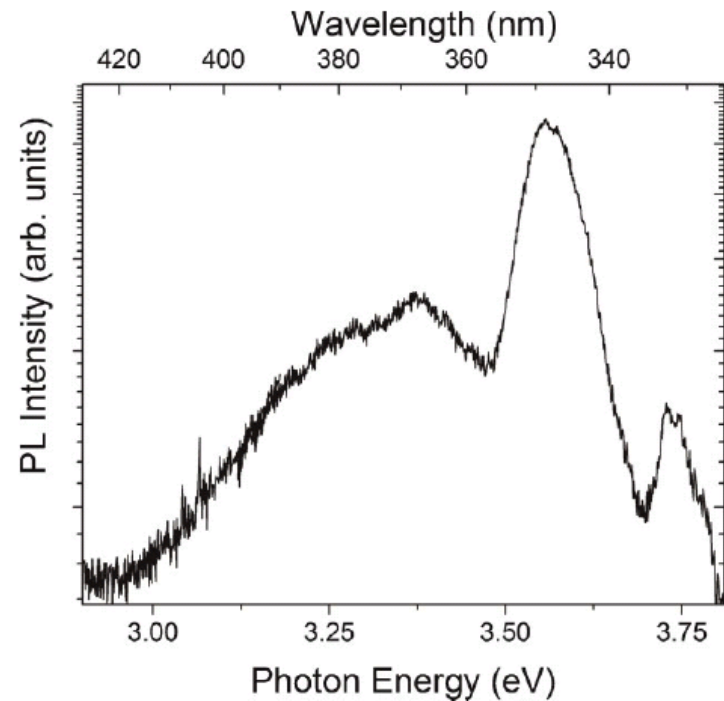
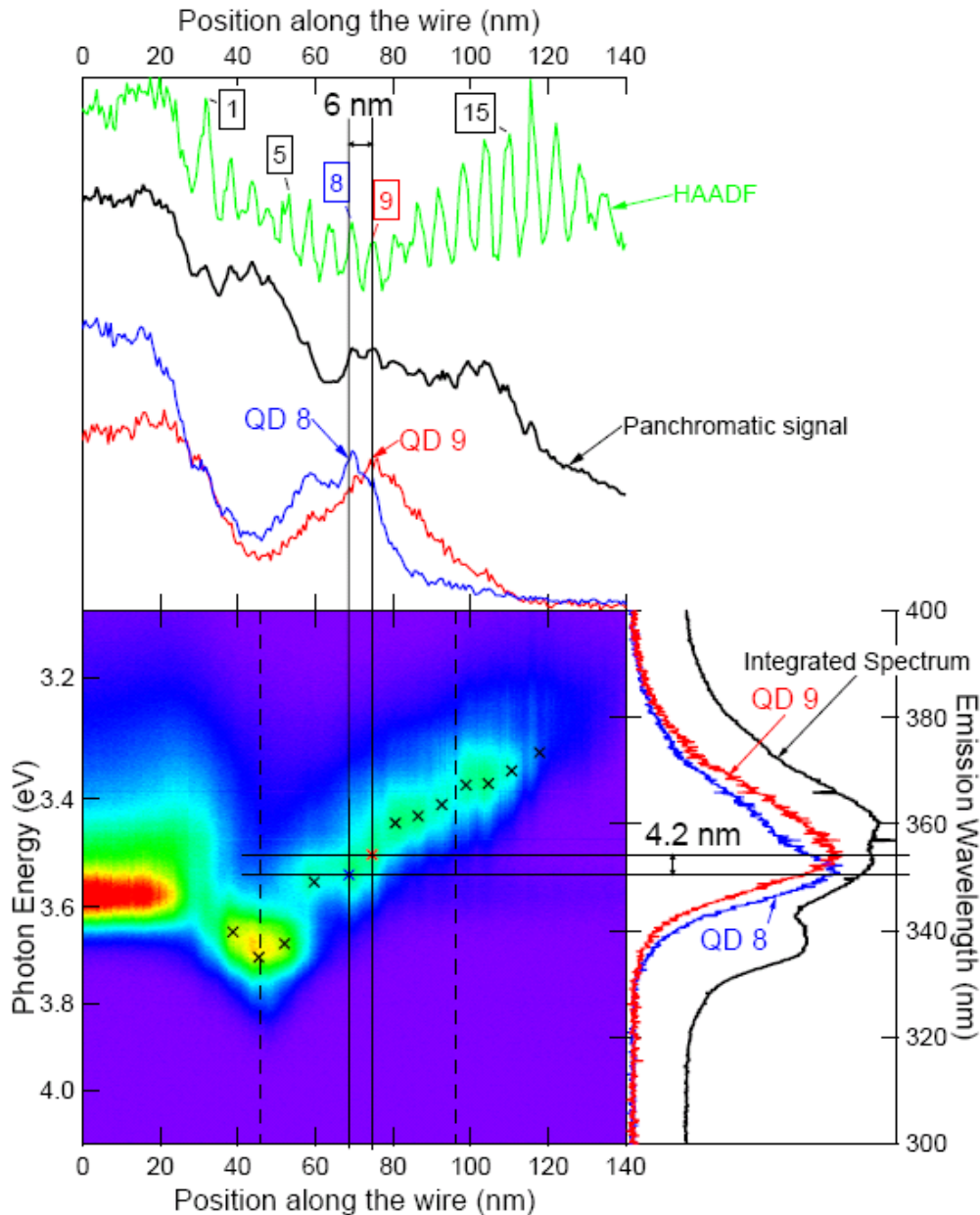


Each individual QD appears as a peak in the combined spatial-spectral plot.

High spatial and spectral resolutions and sampling are needed to find and distinguish each of these peaks.

L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573

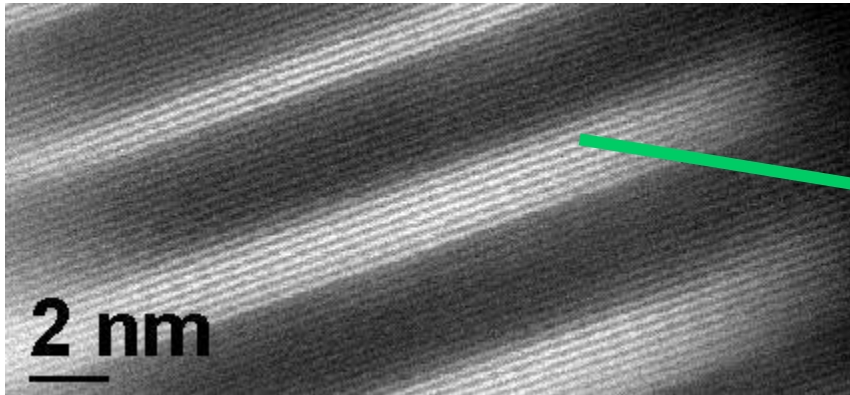
Identifying single QD emissions



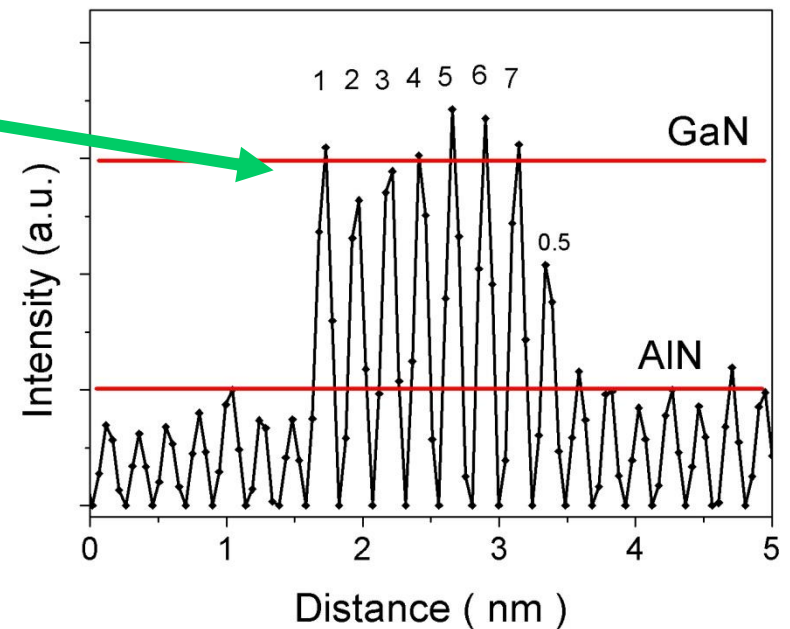
Previous micro-PL data compare well with spatially integrated signal on CL.

**L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573**

Measurement of QD thickness

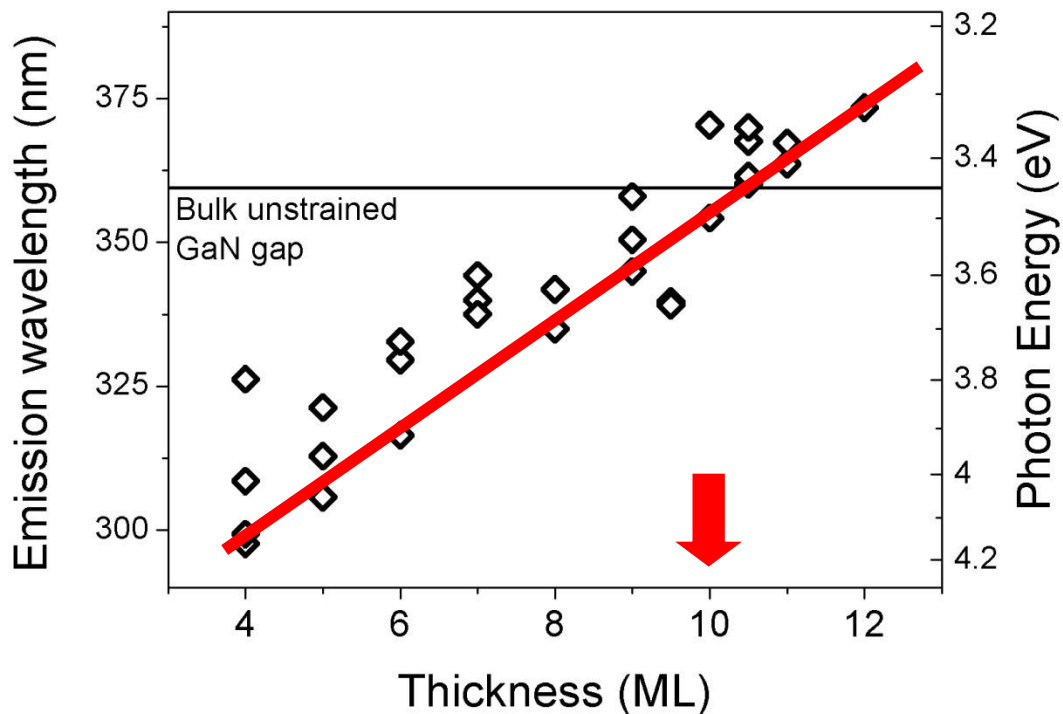


HAADF HR-STEM images were acquired to determine the thickness of each QD.



L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573

Relation QD thickness vs. emission energy

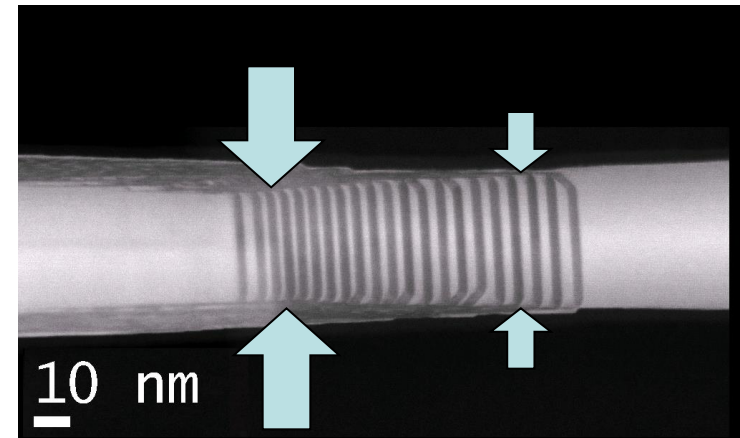
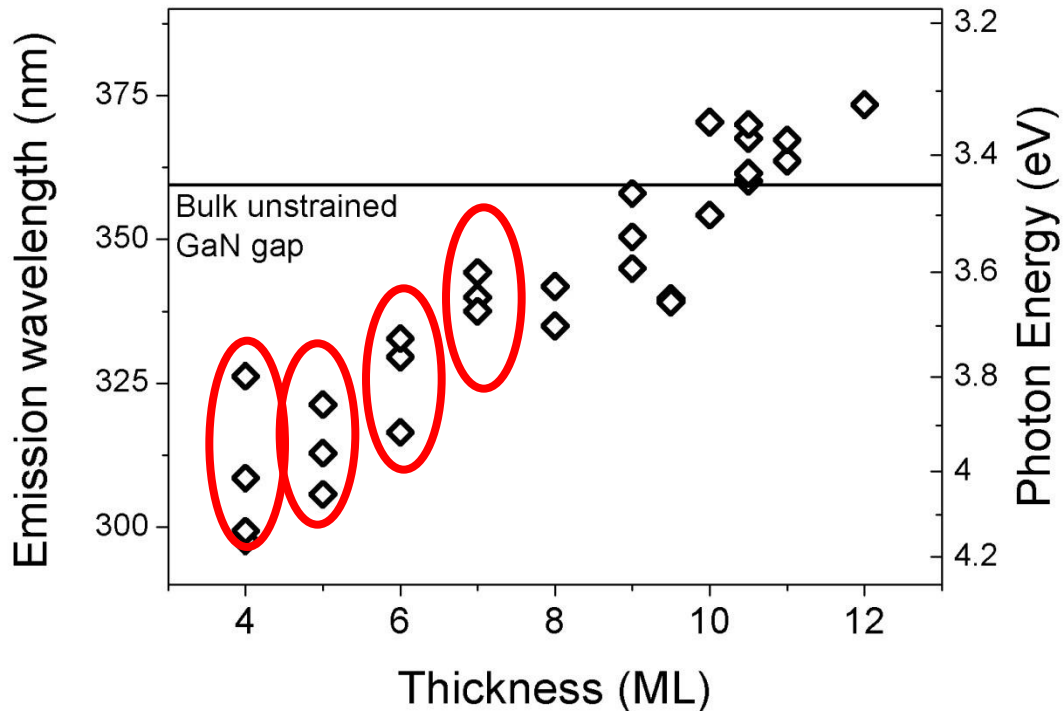


Each data point represents a single QD of known size and emission energy!

The Quantum Confinement is clearly evidenced by the relation of QD thickness and emission energy.

L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573

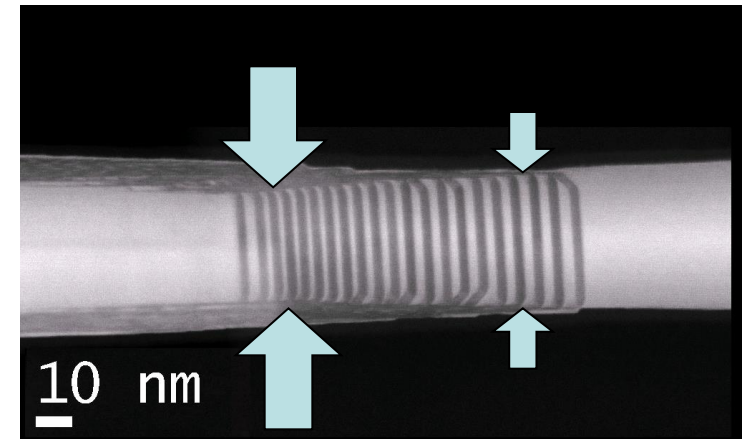
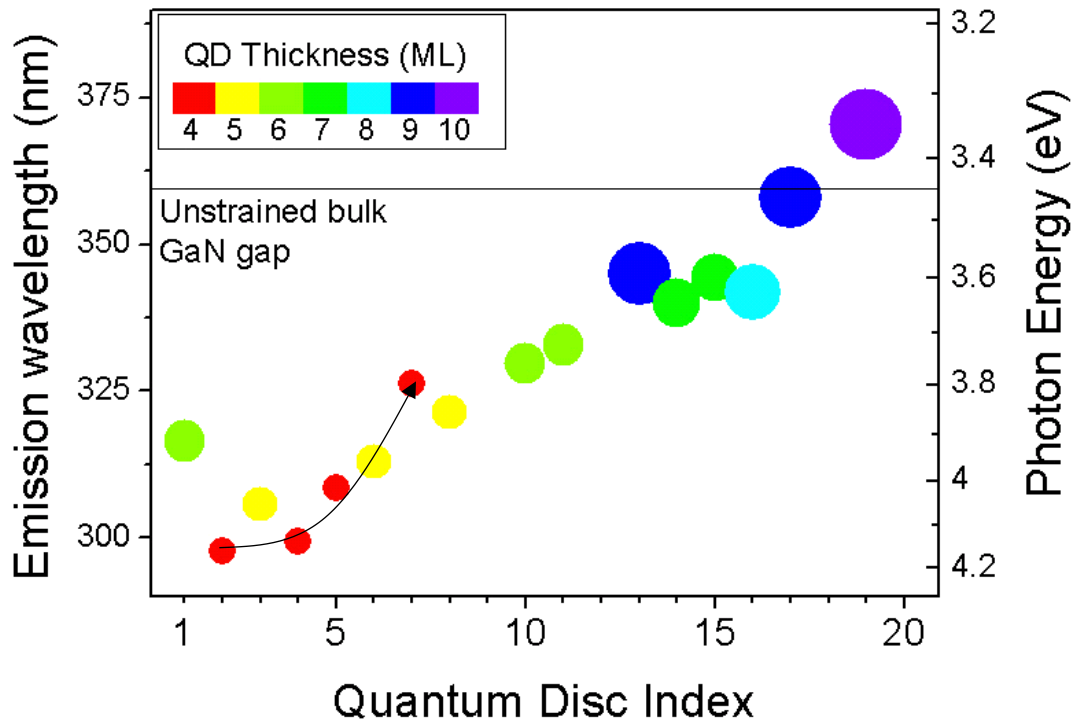
Relation QD thickness vs. emission energy



The dispersion in the curve is possibly caused by strain.

L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573

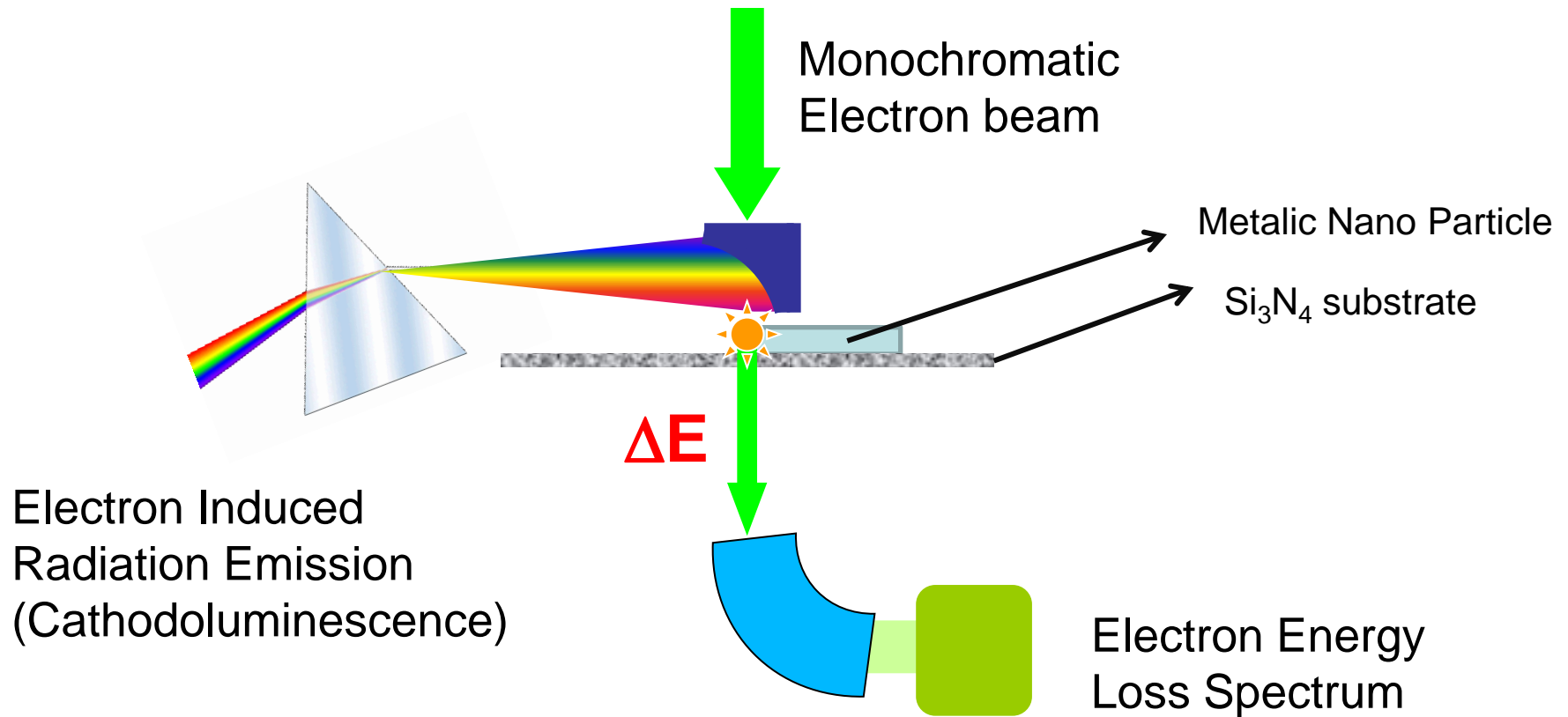
Effect of the AlN Shell



At equal thickness, QD's emission red-shift from the begin to the end of the QD stack.

L. F. Zagonel et al.,
Nano Lett. 2011, 11, 568–573

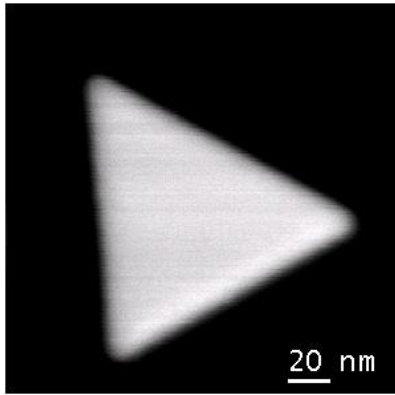
Emission and absorption with electrons



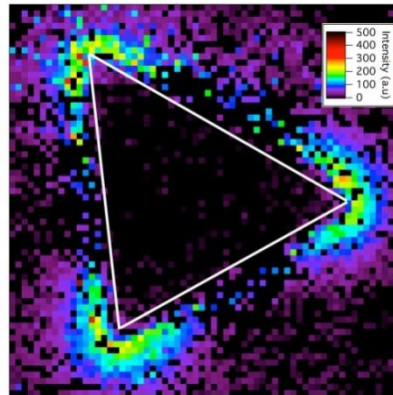
Emission by CL

Absorption by EELS

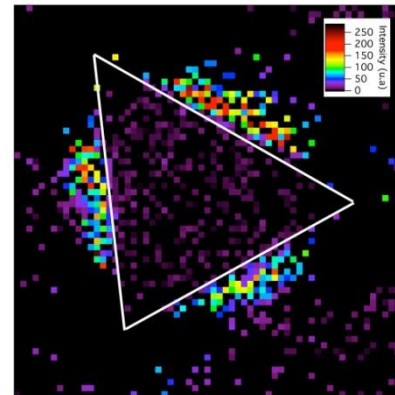
Plasmon modes on Au Triangles



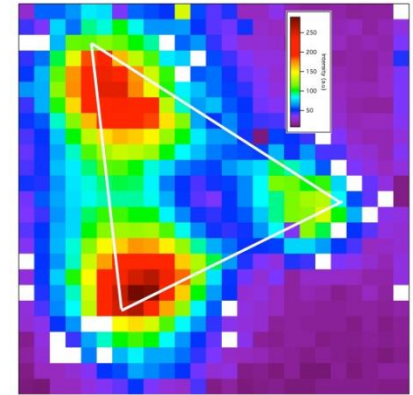
STEM VG
DF Image



Absorption (EELS)
mode at ~2.4 eV



Absorption (EELS)
mode at ~2.7 eV



Emission (Cathodo)
mode at ~2.3 eV

Collaboration with



Luis M. Liz-Marzán
J. B. Rodriguez
Vigo University
Spain

Summary

Quantum confinement and Stark effect is clearly evidenced in individual GaN quantum discs.

High localization of CL signal is shown.

Emission and absorption on the same metallic nanoparticle has been performed.

Analysis, simulations and new experiments are in progress.

Thank you for your attention!

Merci de votre attention!

Acknowledgements

Financial Support

