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U.S. DEPARTMENT OF ENERGY

Superlattice structures with Distributed Bragg Reflector for intense spin polarized electron beams

Luca Cultrera

PI Meeting, Nov 17 2025

Outline

- III-V photocathode for high QE and ESP
  - Lifetime, QE, and ESP tradeoffs
- Design of the SL-DBR
- Experimental Results on QE and ESP
- Future Developments
- Conclusions

Objectives and Budget

Milestone 1: *Demonstrate SL-DBR GaAsP/GaAs photocathode capable to achieve QE>10% and ESP >80% at ~780 nm.*

Milestone 2: *Demonstrate SL-DBR GaAsP/InGaAs photocathode lattice matched on GaAs capable to achieve QE>10% and ESP > 90% at ~800 nm.*

Milestone 2: *Demonstrate SL-DBR GaAsP/GaAsSb photocathode lattice matched on GaAs capable to achieve QE>10% and ESP > 90% at ~1030 nm.*

WBS or ID #	Item/Activity	Baseline Total Cost (FY24)	Baseline Total Cost (FY25)	Costed & Committed (AY$)	Estimate To Complete (AY$)	Estimated Total Cost (AY$)
1	Photocathode design	$50,000.00	$50,000.00	$80,000.00	$20,000.00	$100,000.00
2	Photocathode growth	$50,000.00	$150,000.00	$50,000.00	$150,000.00	$200,000.00
3	Photocathode characterization	$27,000.00	$173,000.00	$122,212.82	$77,787.18	$200,000.00
Totals:		$127,000.00	$373,000.00	$252,212.82	$247,787.18	$500,000.00

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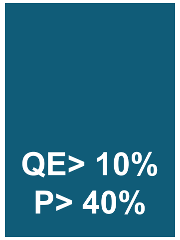
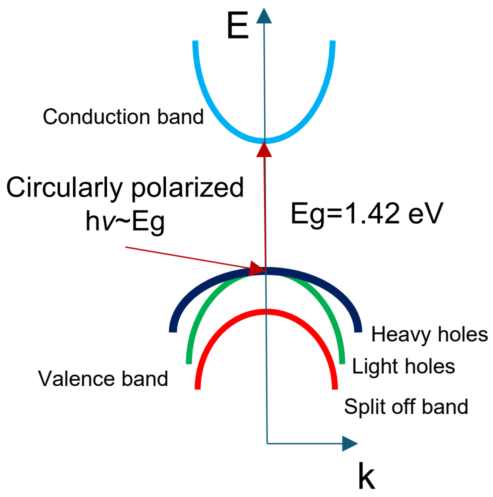
GaAs based photocathodes

Bulk GaAs
(no layer - 1 material)

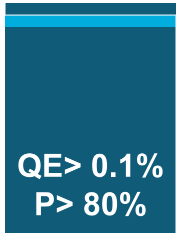


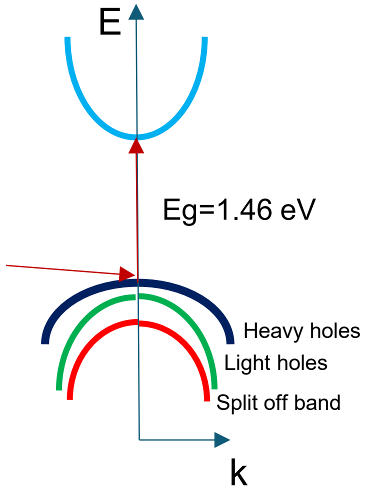


Polarization limited by energy degeneracy

D.T. Pierce *et al.*, *Phys. Lett.* **51A**, 465 (1975)

Strained GaAs
(2 layer – 2 materials)

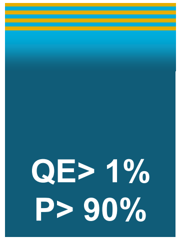


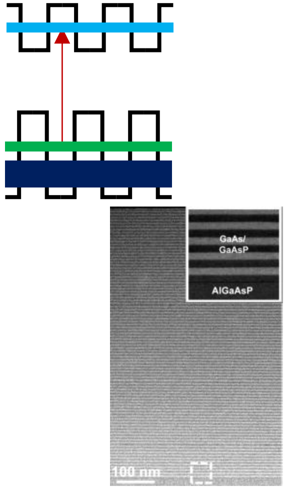


QE and P limited by optical absorption and crystal defects

T. Maruyama *et al.*, *Phys. Rev. B*, ***46*** , 4261 (1992)

III-V GaAs SuperLattices
(~30 layers – 3 materials)

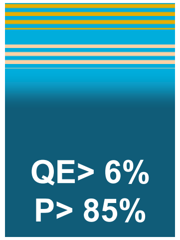


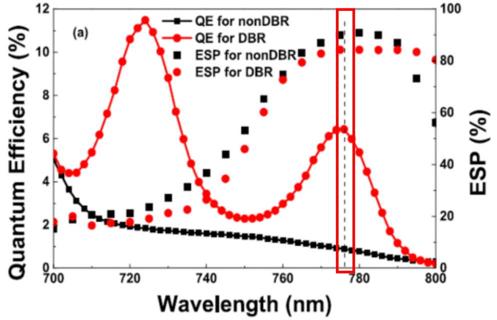


QE limited by optical absorption and crystal defects

X. Jin *et al.*,*Appl. Phys. Lett.* **105**, 203509 (2014)

III-V GaAs SL with DBR
(~60 layers – 5 materials)





Inspirational work

W. Liu *et al., Appl. Phys. Lett.* **109**, 252104 (2016)

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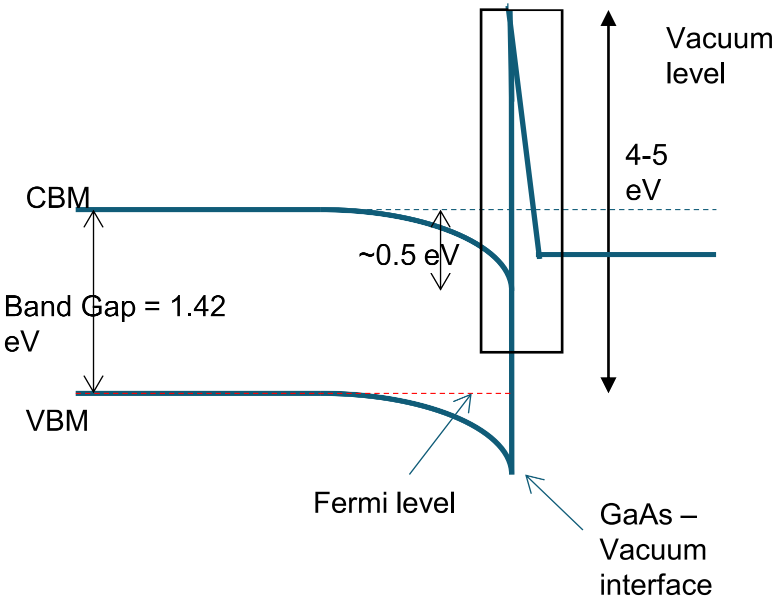
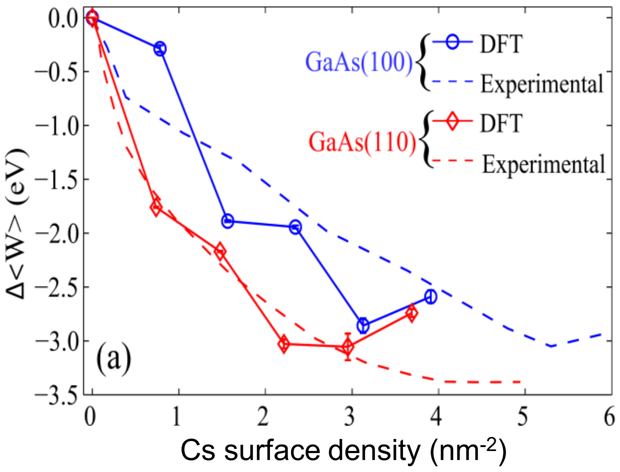
Extracting electrons requires Negative Electron Affinity conditions

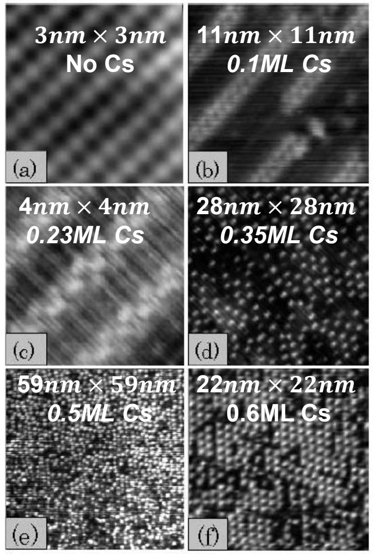
Negative Electron Affinity

All GaAs based photocathodes *operated at photon energies near the band gap must be activated to* ***Negative Electron Affinity (NEA)*** *condition using Cs and O (or other methods) to form a strong surface dipole lowering the electron affinity below the conduction band minimum*



A.J. Van Bommel and J.E. Crombeen, *Surf. Sci.*, **57**, 109 (1976)





S. Karkare *et al.*, *Phys. Rev. B* **91**, 035408 (2015)

Less than 1 monolayer to achieve NEA

Extremely vacuum sensitive => vacuum better than 10^{-11} Torr are required

Vacuum requirements have so far limited operation in HV DC gun

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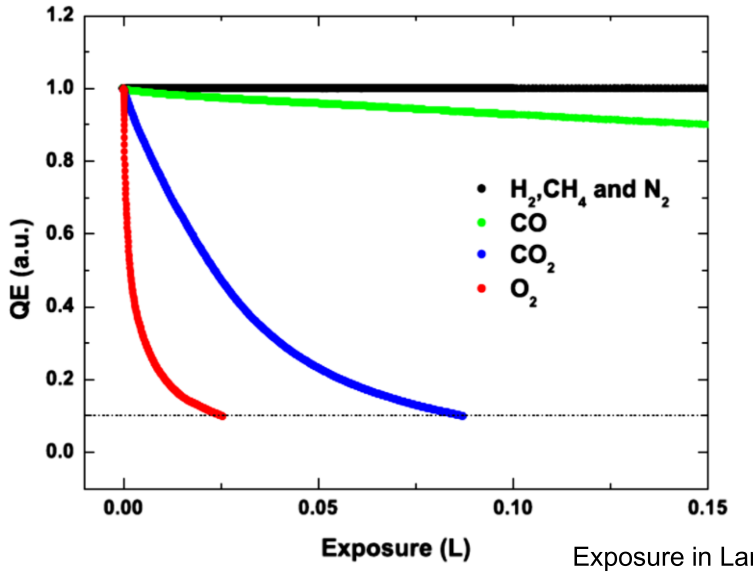
GaAs QE degradation mechanisms

- Photocathode QE degradation can be induced by the following mechanisms:

Chemical poisoning

Oxygen is a killer

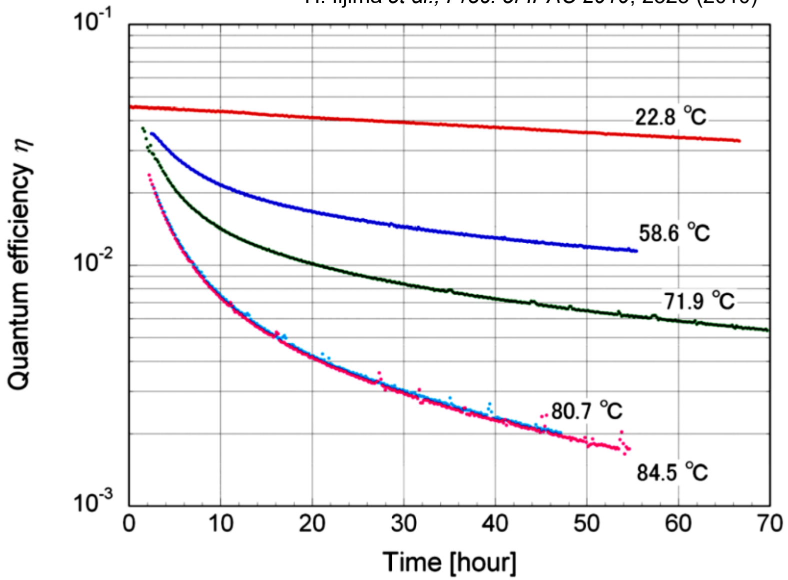


Exposure in Langmuir
1L = 10^{-6} Torr*s

N. Chanlek *et al., J. Phys. D: Appl. Phys.* **47**, 055110 (2014)

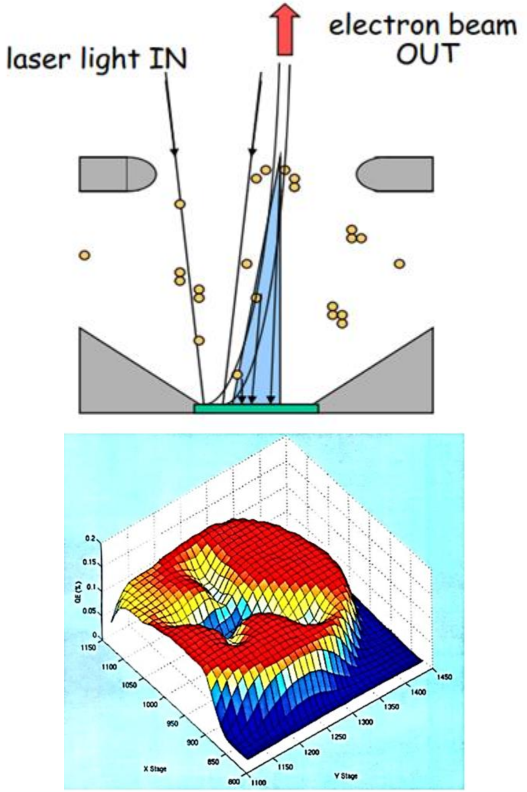
Alkali desorption

H. Iijima *et al., Proc. of IPAC 2010*, 2323 (2010)



Relevant for high average power or tightly focused lasers beams

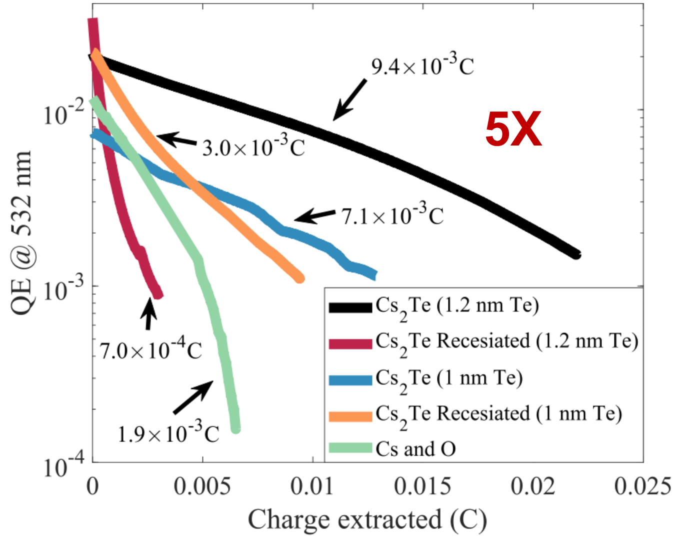
Ion back bombardment



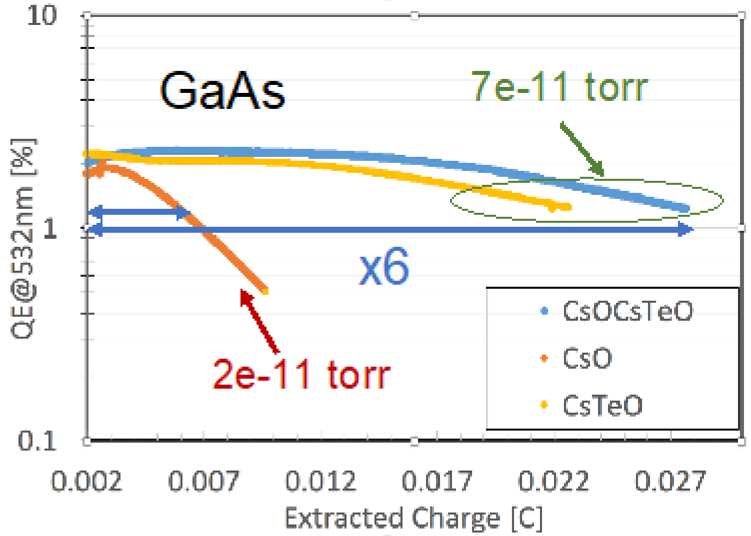
J Grames *et al., AIP Conf. Proc.* **980**, 110 (2008)

Robust activation layer yield longer lifetimes

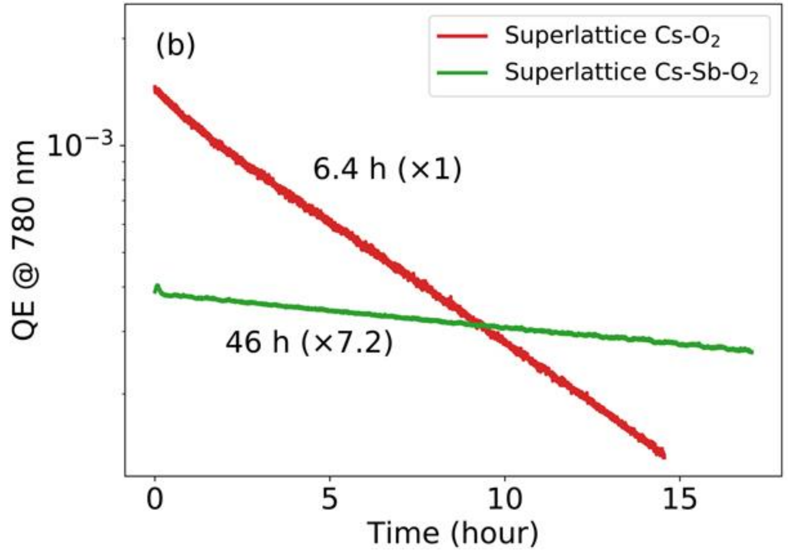
J. Bae et al., Appl. Phys. Lett. **112** (2018) 154101

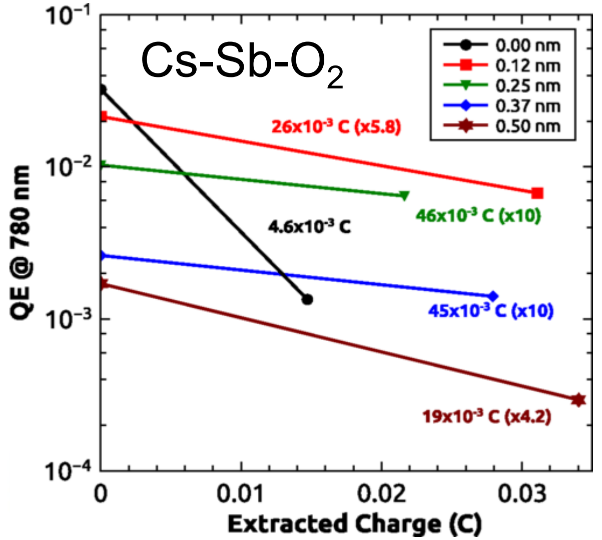


O. Rahman et al., IPAC 2019, TUPTS102

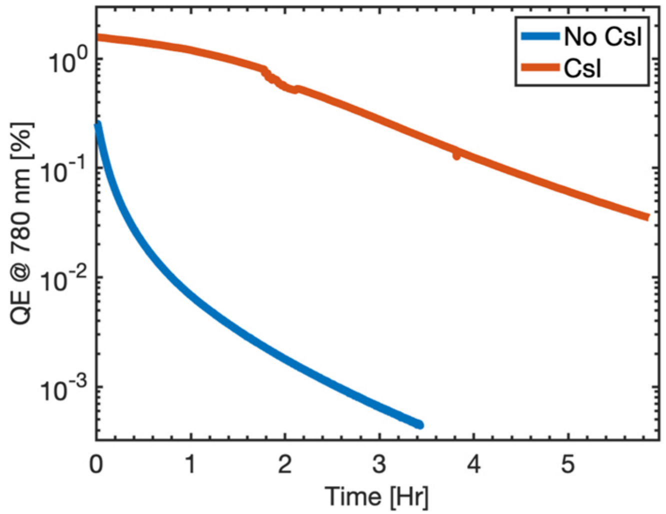


J Bae et al., J. Appl. Phys. **127**, 124901 (2020)

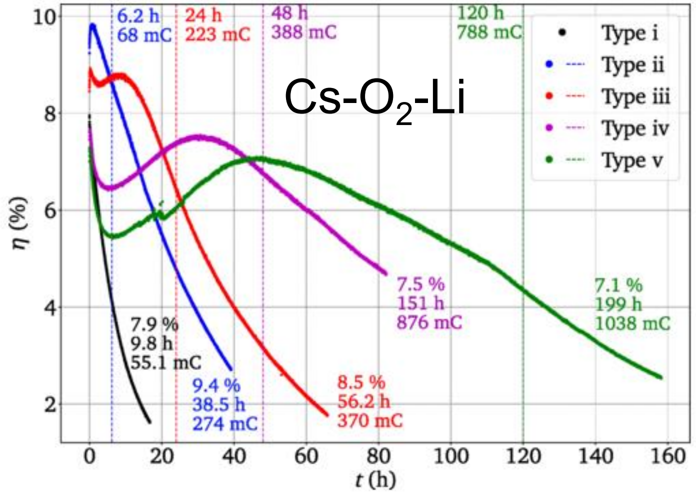




L. Cultrera et al., Phys. Rev. Accel. Beams **23**, 023401 (2020)

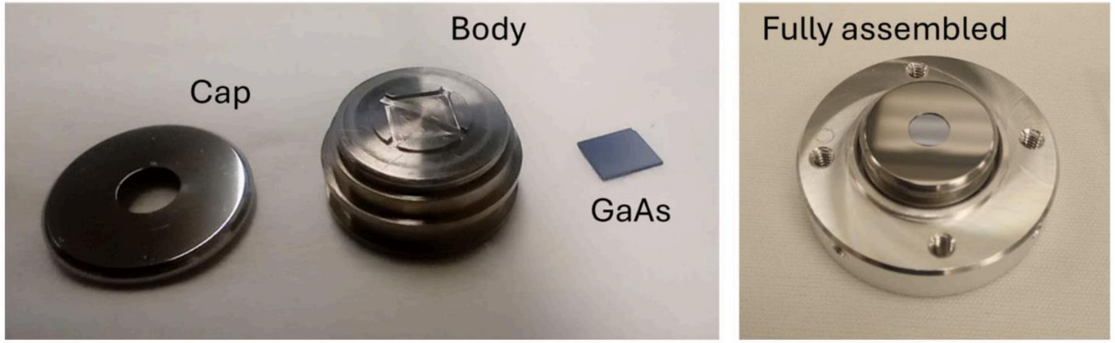


S.J. Levenson et al., J. Appl. Phys. **137**, 224901 (2025)



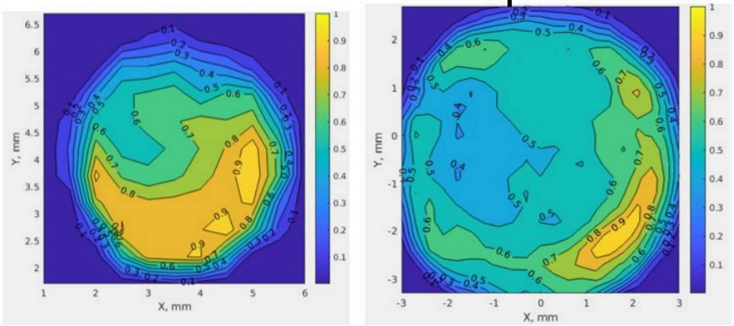
M. Herbert et al., Phys. Rev. Accel. Beams **28**, 013401 (2025)

CsO-Te-CsO activated photocathode in SRF gun

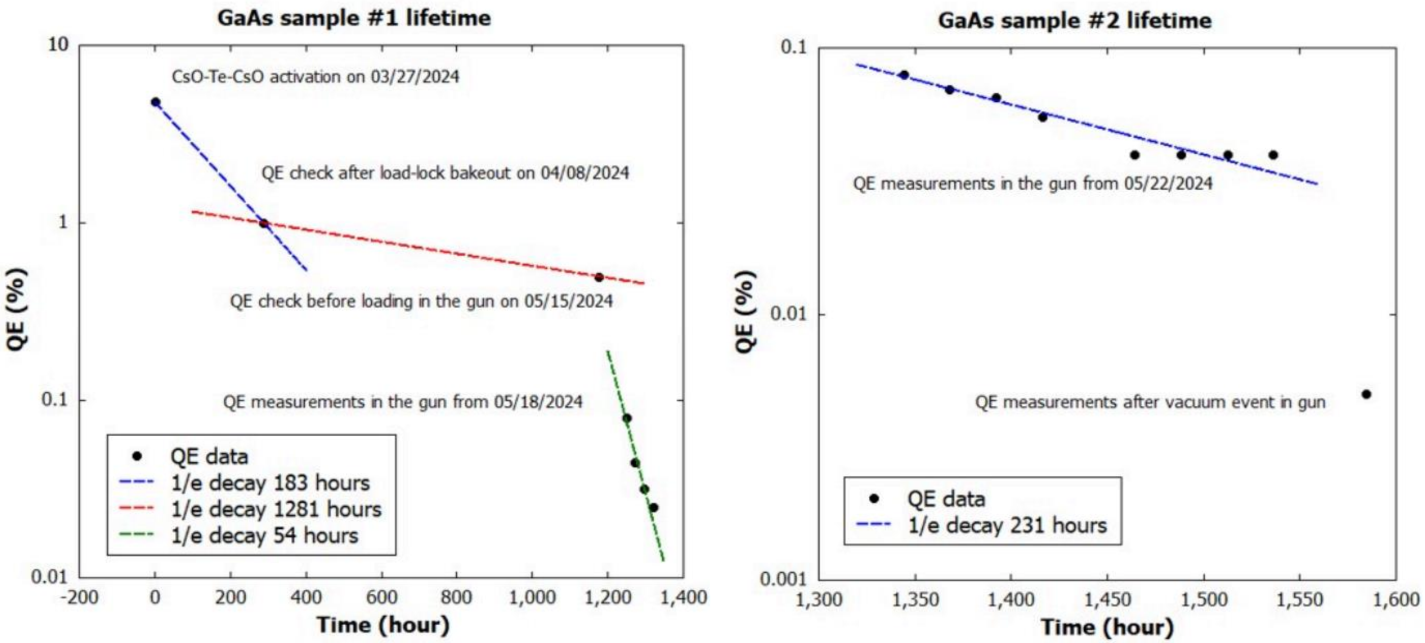


(a) (b)

Relative QE maps


(a) (b)

- Cathode holder specially designed for SRF 112 MHz electron gun;
- Cathodes were transferred using a vacuum suitcase from production system to RHIC tunnel;
- Operation was only performed at 532 nm;
- **Cathodes survived transfers and were operated for hundreds of hours;**
- Max E_{field}=12 MV/m
- Max Charge/bunch=1.2 nC



(a) (b)

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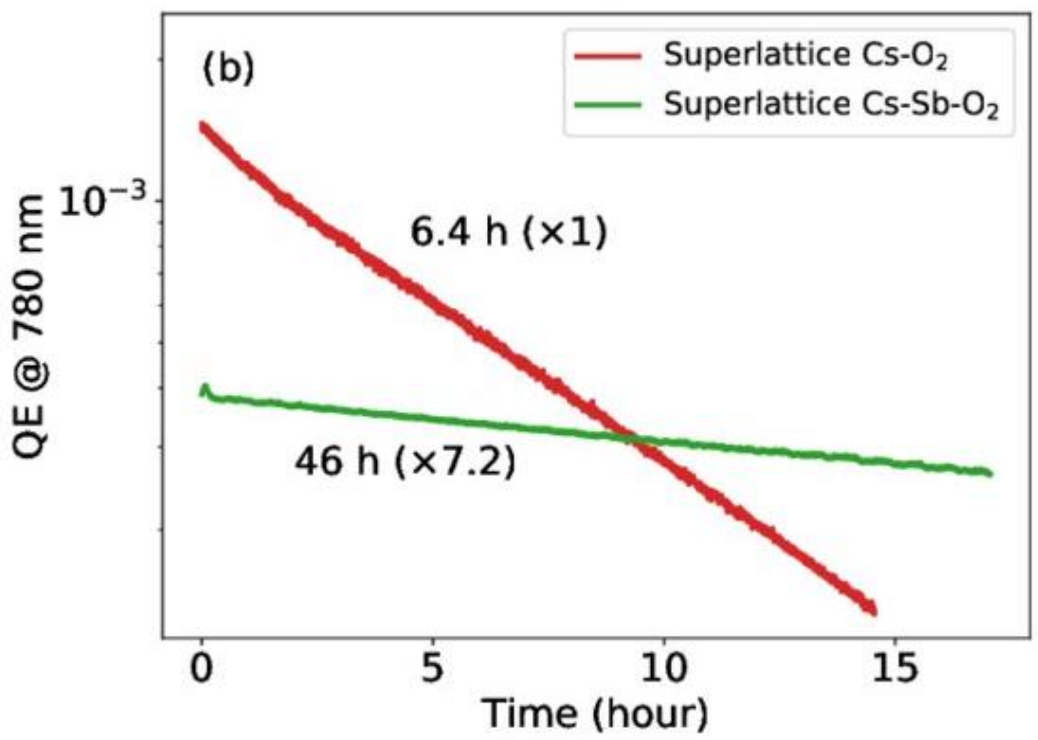
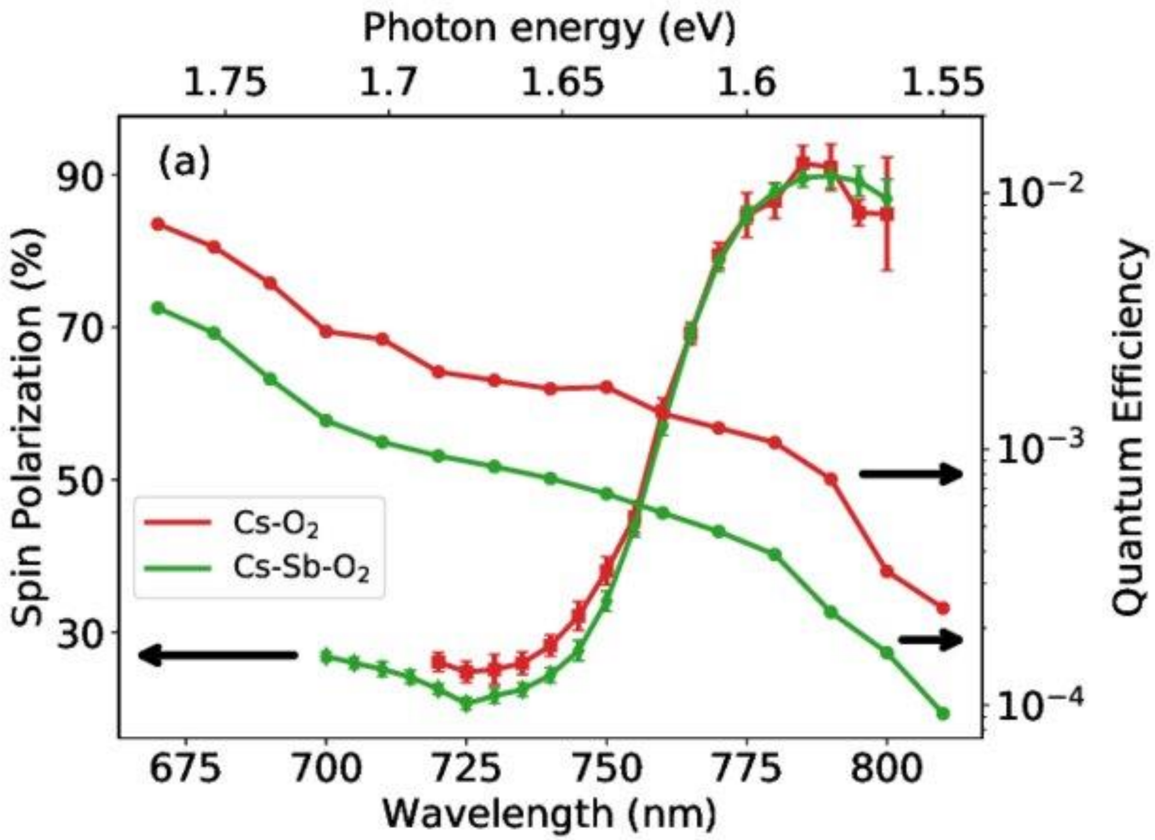
V. Litvinenko et al., "Towards a New Class of Polarized Electron Sources: An RF Gun with GaAs Photocathode", Nature Physics, accepted

QE and ESP tradeoffs

J Bae et al., *J. Appl. Phys.* **127**, 124901 (2020)





The use of robust activating layers allows to increase lifetime at expenses of QE

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Increasing QE with strain compensated SL-DBR

GaAs	5 nm	p = 5×10^{19} cm^{-3}		EMISSION LAYER
$GaAs_{0.62}P_{0.38}$	4 nm	p = 5×10^{17} cm^{-3}	30 pairs	STRAIN COMPENSATED SL
GaAs	4 nm	p = 5×10^{17} cm^{-3}		
$GaAs_{0.81}P_{0.19}$	300 nm	p = 5×10^{18} cm^{-3}		FABRY-PEROT SPACING LAYER
$AlAs_{0.78}P_{0.22}$	65 nm	p = 5×10^{18} cm^{-3}	10 pairs	DISTRIBUTED BRAGG REFLECTOR
$GaAs_{0.81}P_{0.19}$	55 nm	p = 5×10^{18} cm^{-3}		
$GaAs_{0.81}P_{0.19}$	2000 nm	p = 5×10^{18} cm^{-3}		PSEUDO-SUBSTRATE
GaAs->$GaAs_{0.81}P_{0.19}$	2750 nm	p = 5×10^{18} cm^{-3}		GRADED COMPOSITION LAYER
GaAs buffer	200 nm	p = 5×10^{18} cm^{-3}		BUFFER LAYER
GaAs substrate		p > 1×10^{18} cm^{-3}		SUBSTRATE

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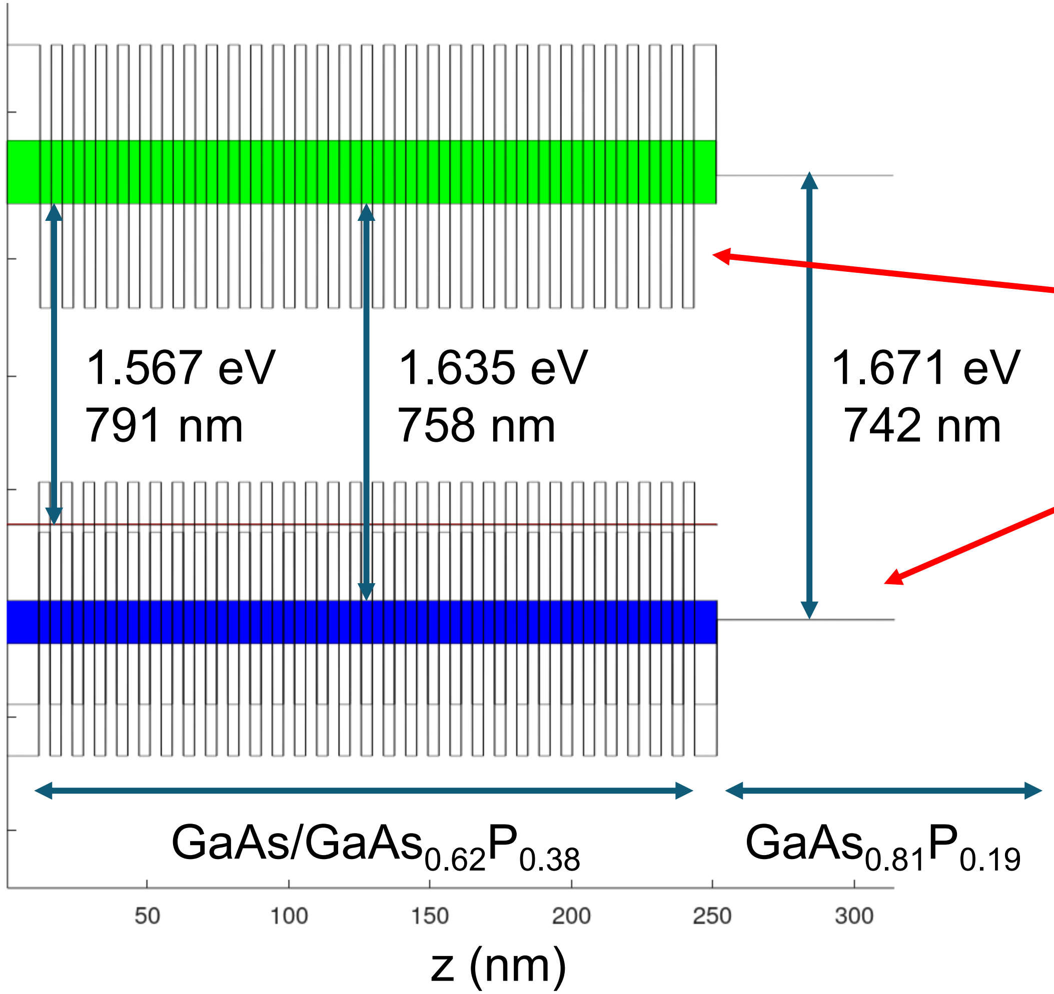
84 layers!! With sub-nm resolution

Miniband calculation



In plan strain estimated at +/- 0.7%
HH-LH separation is estimated at 66 meV

GaAs	5 nm	p = 5×10^{19} cm^{-3}	
$GaAs_{0.62}P_{0.38}$	4 nm	p = 5×10^{17} cm^{-3}	30 pairs
GaAs	4 nm	p = 5×10^{17} cm^{-3}	
$GaAs_{0.81}P_{0.19}$	300 nm	p = 5×10^{18} cm^{-3}	
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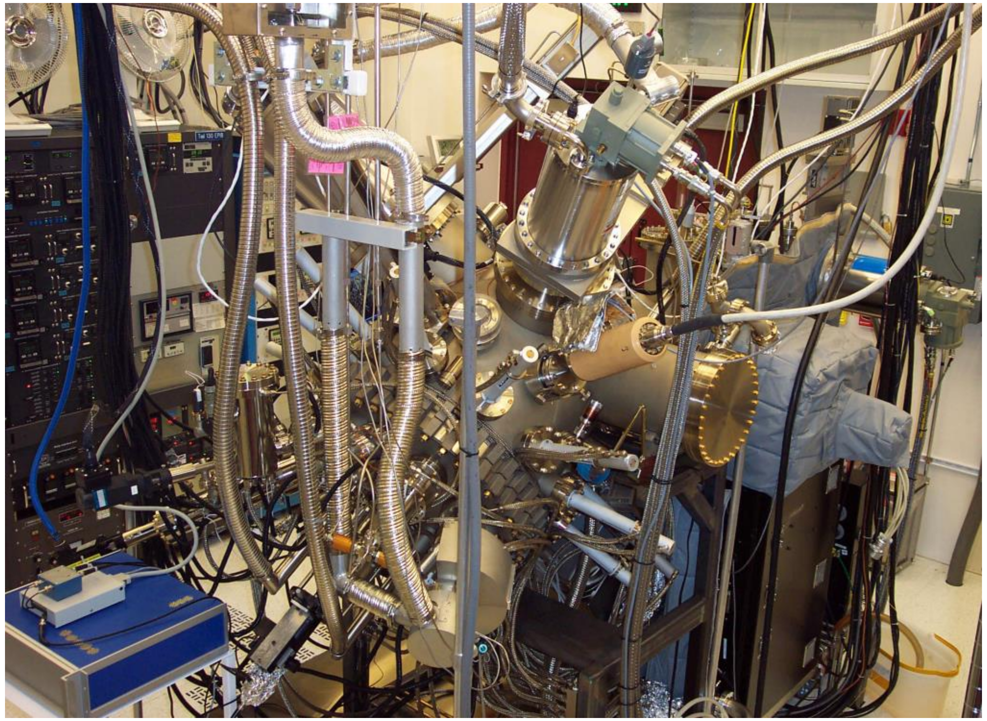
GaAs->$GaAs_{0.81}P_{0.19}$	2750 nm	p = 5×10^{18} cm^{-3}	
GaAs buffer	200 nm	p = 5×10^{18} cm^{-3}	
GaAs substrate		p > 1×10^{18} cm^{-3}	

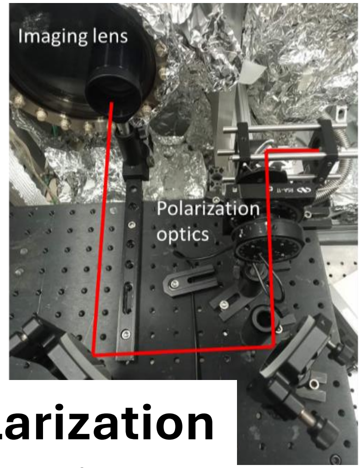
Molecular Beam Epitaxy

Sandia National Laboratories has state of the art MBE infrastructure, and the expertise required to perform the growth of III-V layered structures

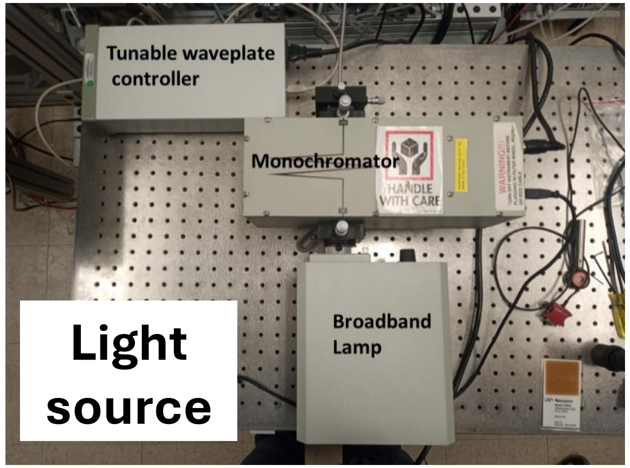


- Highly stable flux control:
    - As is supplied from high purity solid sources
    - P is supplied by phosphine thermal crackers
- Dual-zone substrate heater provides better temperature uniformity over the 3” wafer
- P-Type doping is achieved with carbon obtained using a CBr_4 gas injector
- Wafers were capped with arsenic to protect the surface from oxidation.

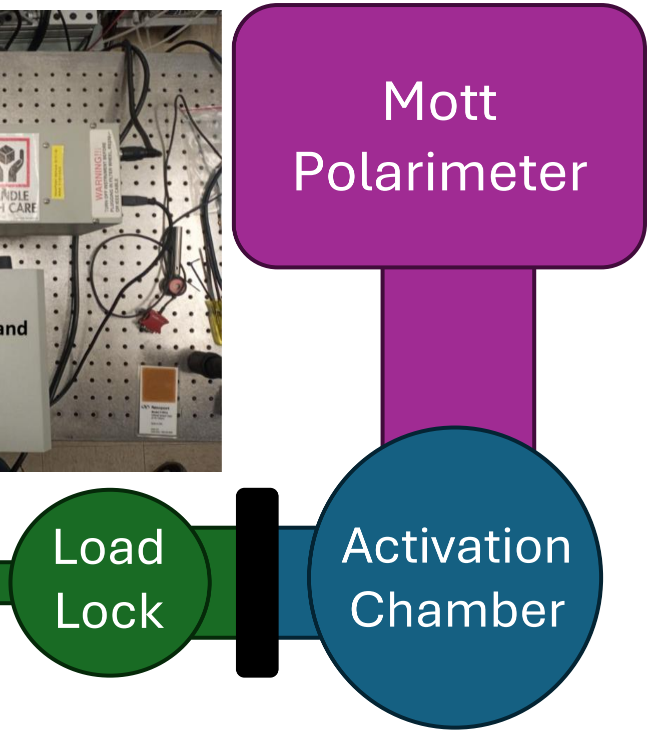
Experimental setup at BNL

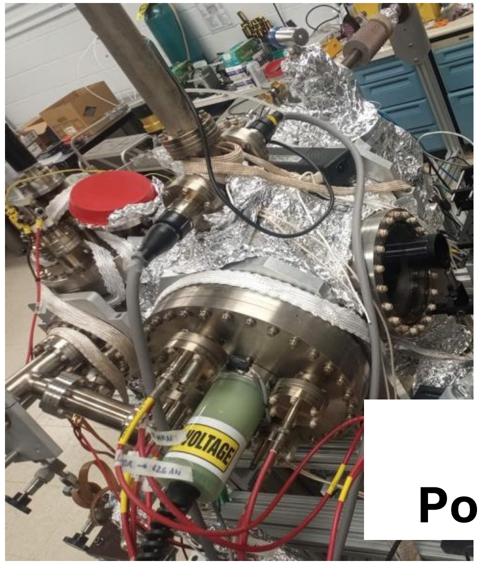


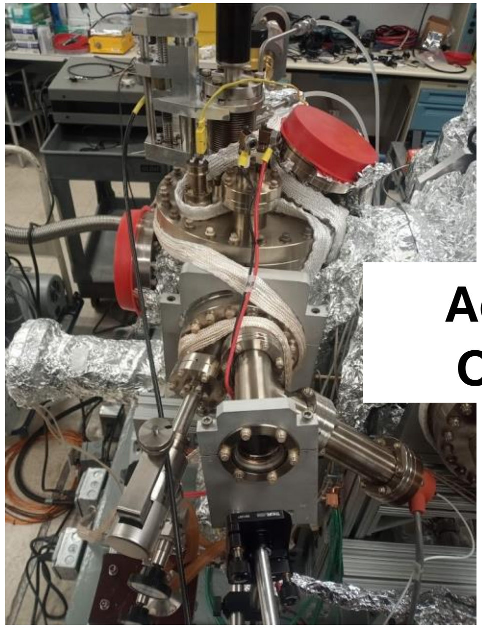
Polarization optics



Light source



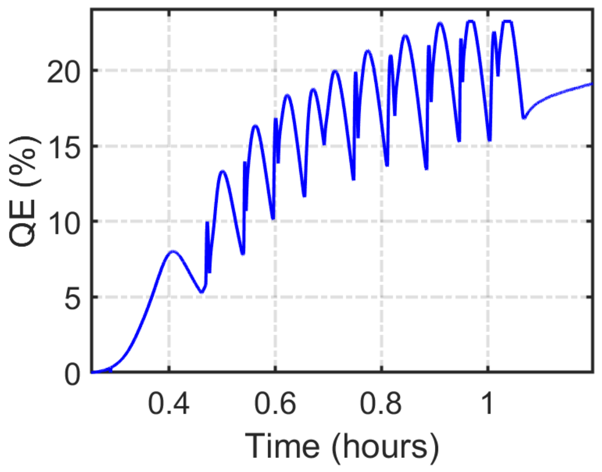

Mott Polarimeter


Activation Chamber


Load lock

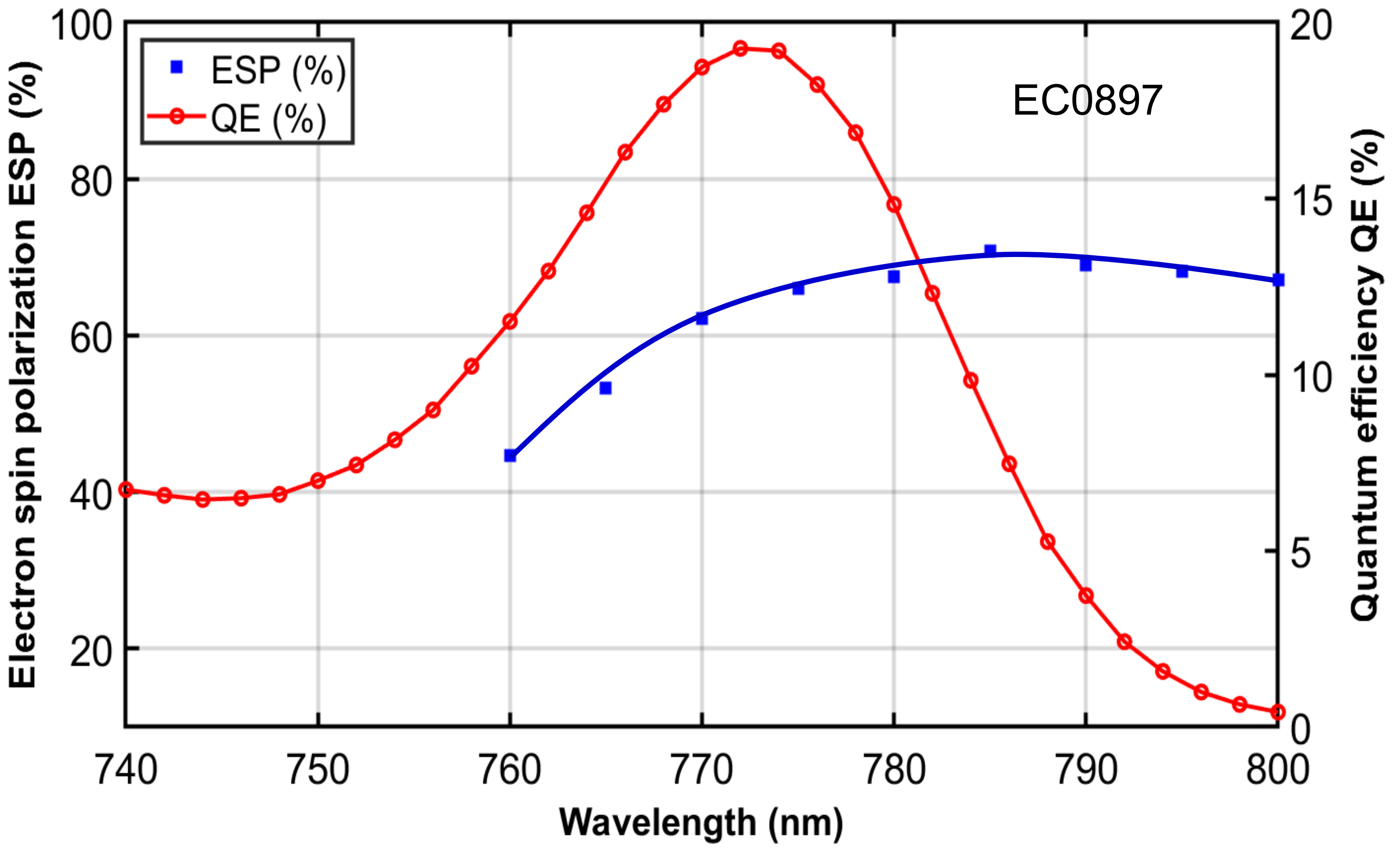
- Base pressure: **low 10^{-11} Torr**
- Spectral range: **400 - 1200 nm**
- Degree of circular polarization: **98-99 %**
- Retarding field Mott polarimeter:
  - Target: **Au**
  - Bias: **25 kV**

QE and ESP – initial design



- Photon vavelength – **532 nm**
- Alternate exposure to Cs and O_2 until peak QE saturates
- Maximum QE at the end of activation ~ 20%

- In-plane strain ~0.7%
- HH-LH estimated at about 70 meV



ESP = ~70%, QE = 15% at 780 nm

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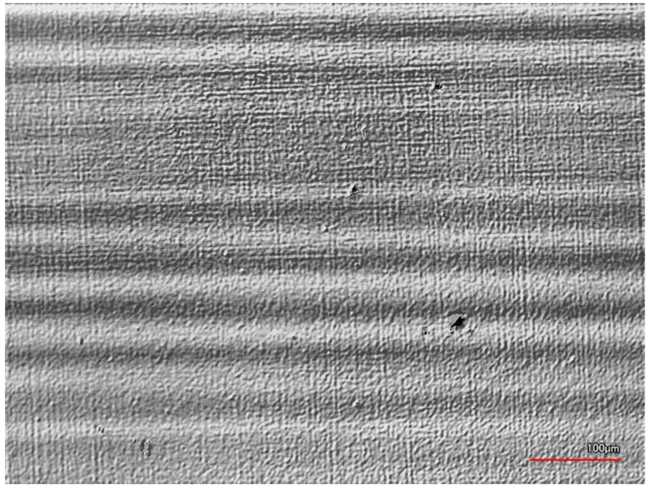
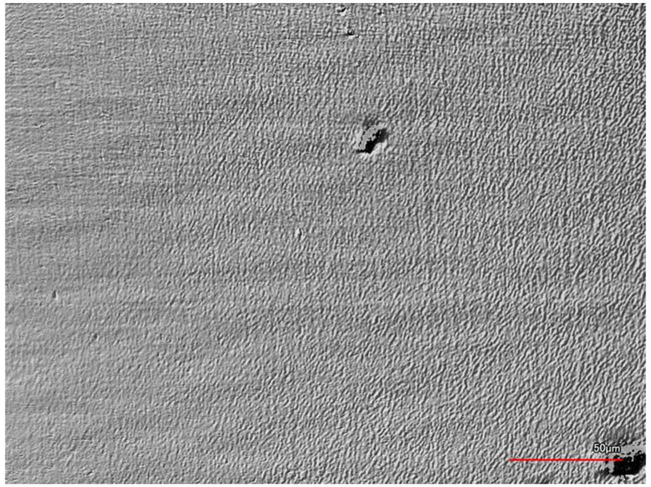
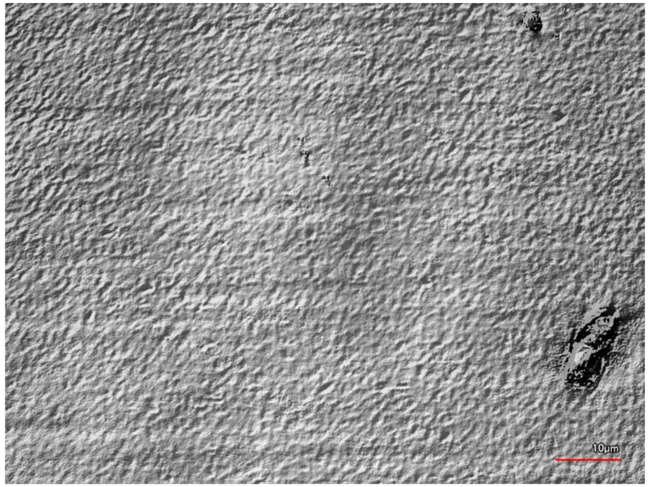
ECO-897 Surface Morphology



20X



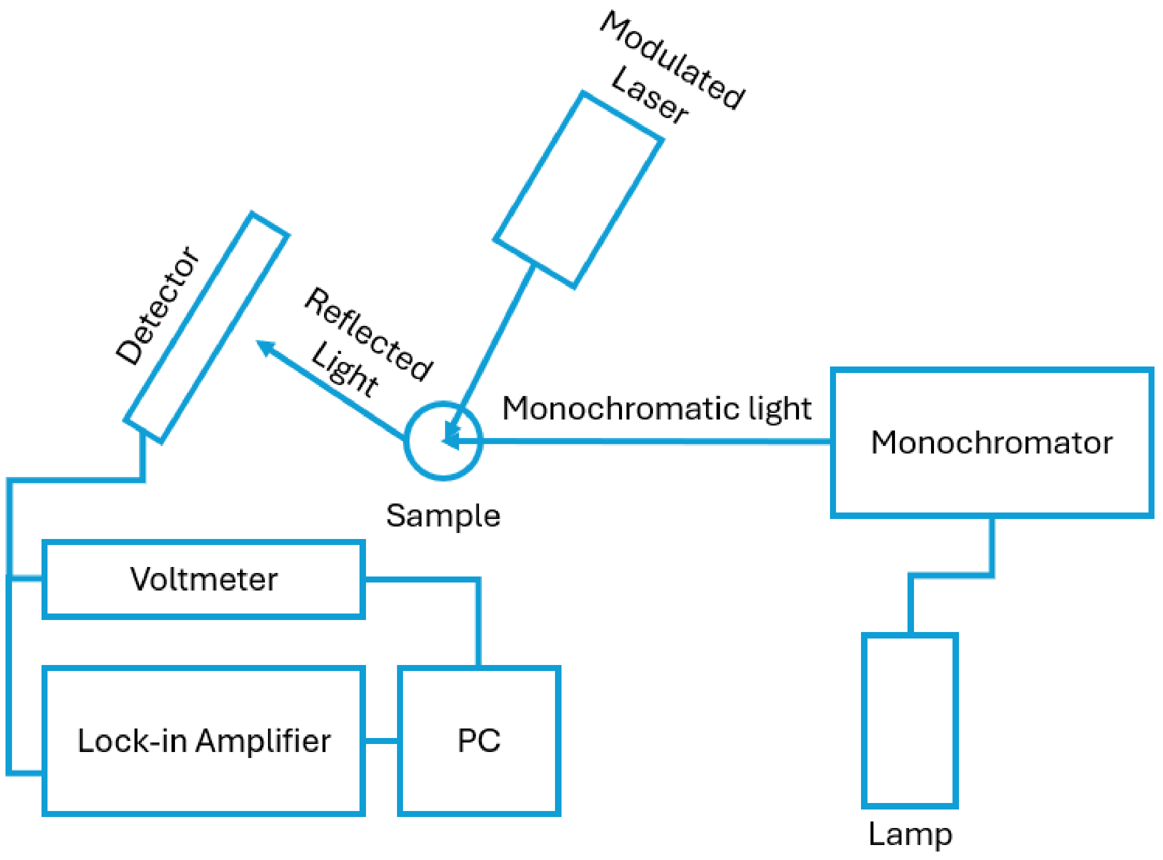
50X



150X

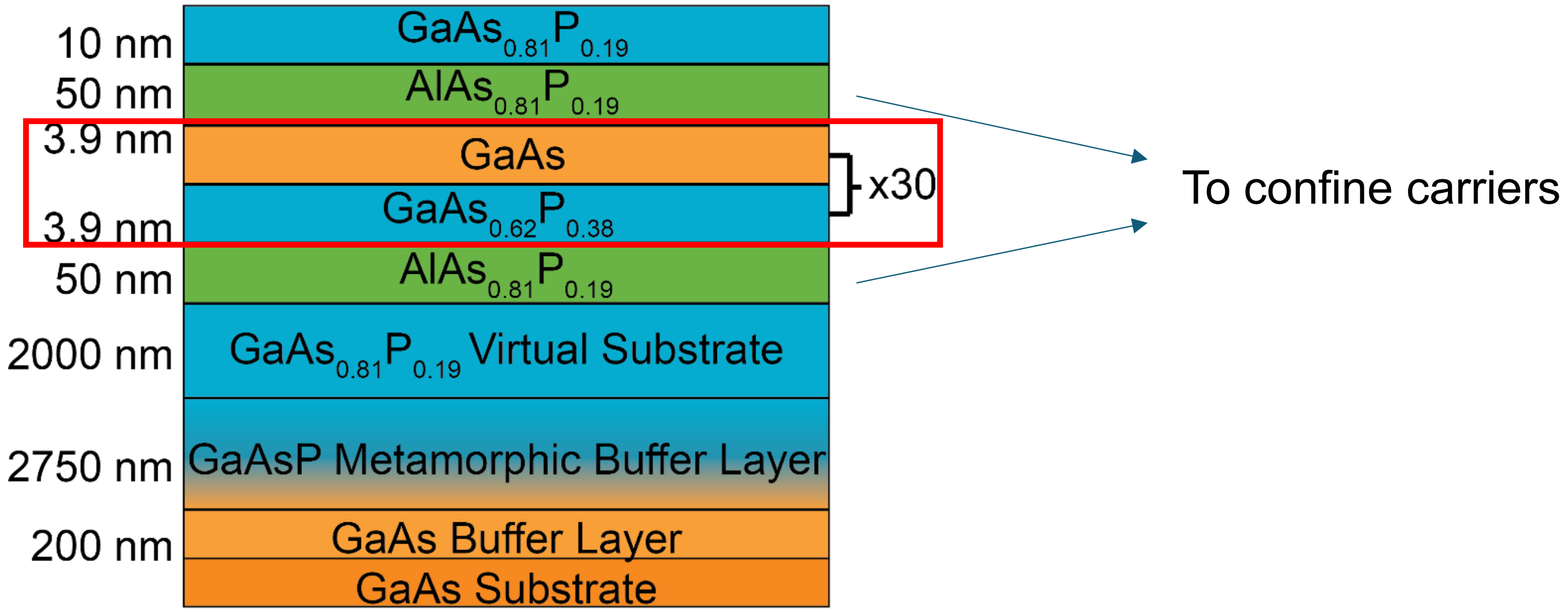
Photoreflectance

- Consists of a probe (white) light and a pump (green laser)
- Pump beam modulates the electric field in the sample
- Measure the change in reflectivity of the sample and normalize by dividing out the reflectance



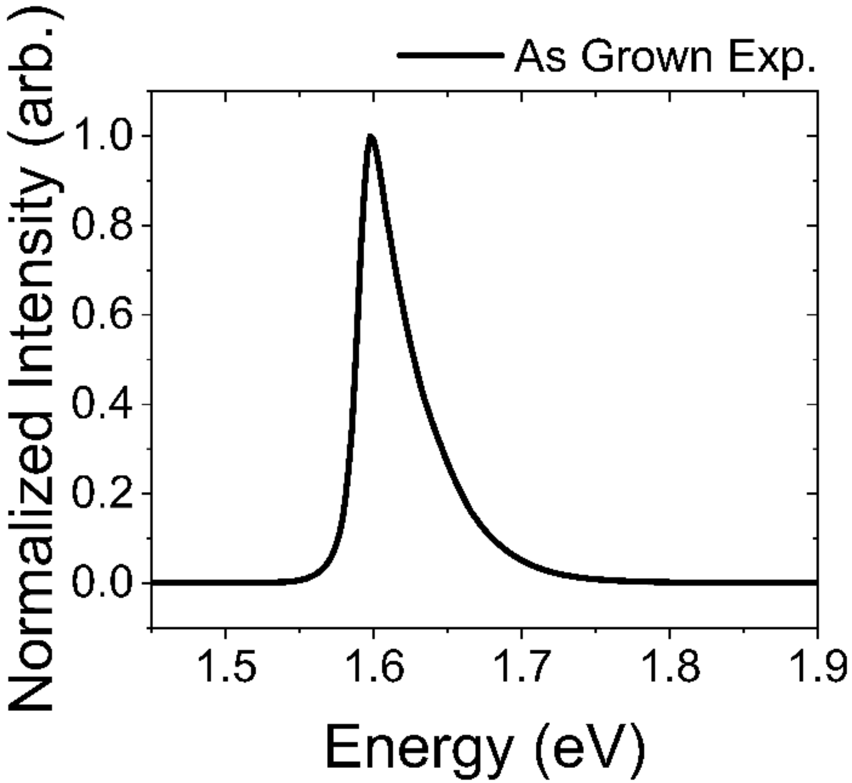
Structure

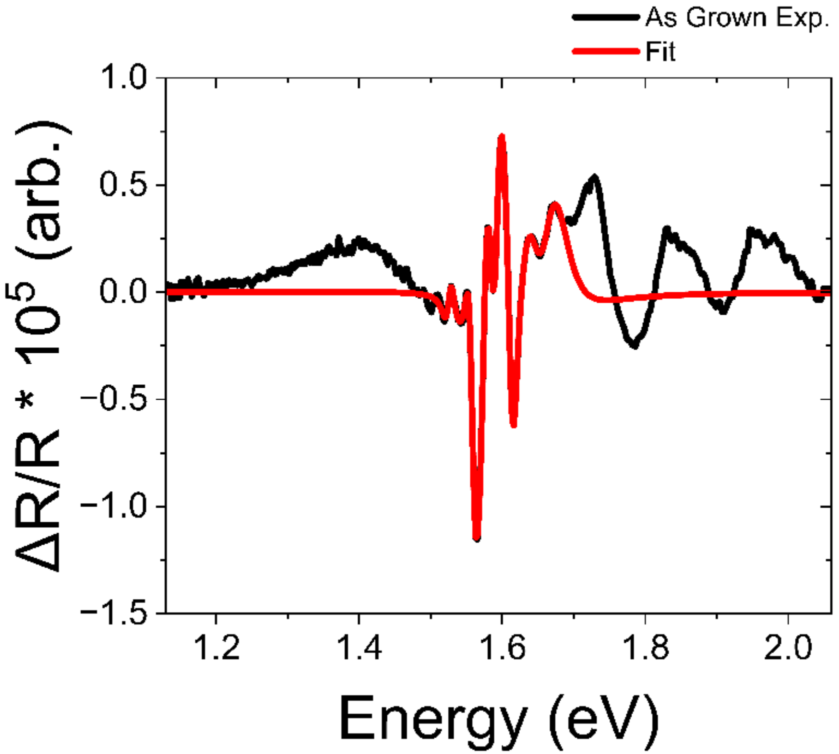
Nominally the same structure from the record QE photocathode, without the DBR



Courtesy of A. Muhowski, M. McDonough

As-Grown Sample





Courtesy of A. Muhowski, M. McDonough

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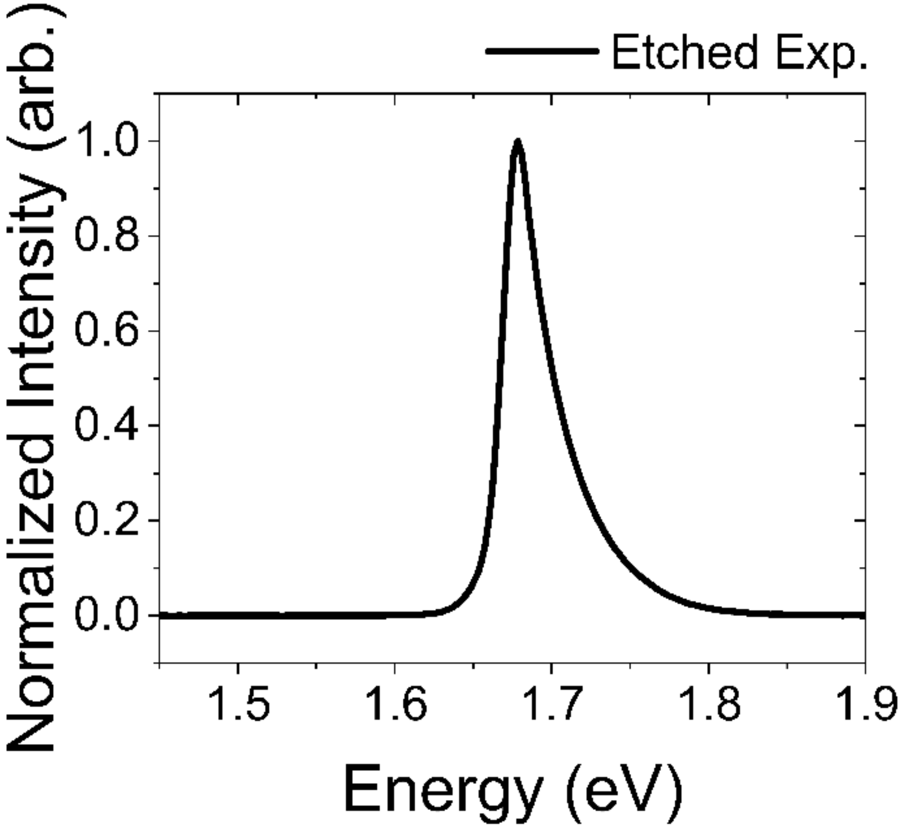
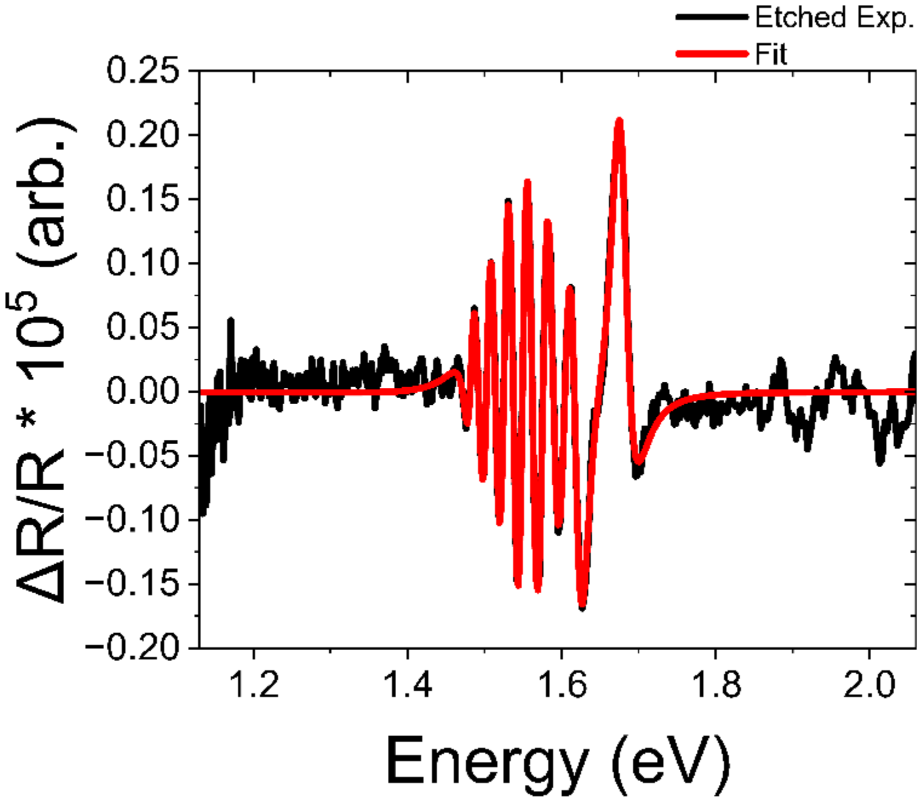
Etched Sample

Sample was etched down about 300 nm into the virtual substrate to remove the superlattice structure





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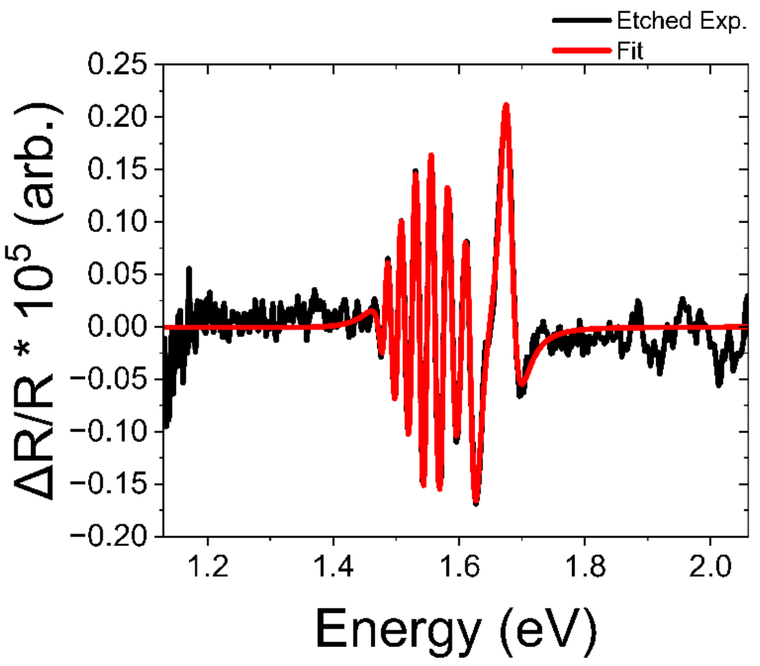
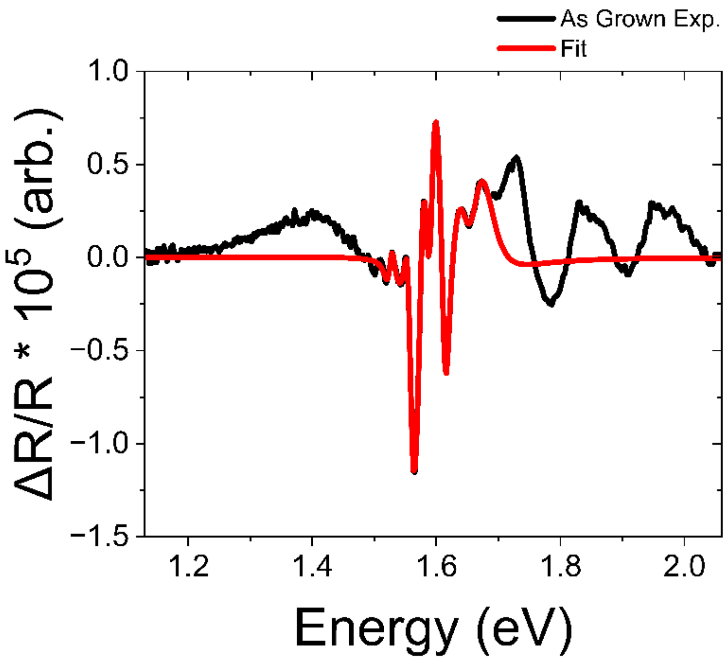
Courtesy of A. Muhowski, M. McDonough

Comparison of Fitting Results



E_{CP} (eV)	Identity
1.483	Metamorphic Buffer Layer
1.503	Metamorphic Buffer Layer
1.524	Metamorphic Buffer Layer
1.547	Metamorphic Buffer Layer
1.564	Metamorphic Buffer Layer
1.582	Metamorphic Buffer Layer
1.616	Metamorphic Buffer Layer
1.679	E_g of Virtual Substrate



E_{CP} (eV)	Identity
1.527	Metamorphic Buffer Layer
1.557	Metamorphic Buffer Layer
1.579	CB/HH
1.614	Metamorphic Buffer Layer
1.636	CB/LH
1.662	E_g of Virtual Substrate

HH/LH Splitting of 57 meV

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Courtesy of A. Muhowski, M. McDonough

Modification to the FP spacing layer

GaAs	5 nm	p = 5x10^{19} cm^{-3}
$GaAs_{0.62}P_{0.38}$	4 nm	p = 5x10^{17} cm^{-3}
GaAs	4 nm	p = 5x10^{17} cm^{-3}
$GaAs_{0.81}P_{0.19}$	300 nm	p = 5x10^{18} cm^{-3}
$AlAs_{0.78}P_{0.22}$	65 nm	p = 5x10^{18} cm^{-3}
$GaAs_{0.81}P_{0.19}$	55 nm	p = 5x10^{18} cm^{-3}
$GaAs_{0.81}P_{0.19}$	2000 nm	p = 5x10^{18} cm^{-3}
GaAs->$GaAs_{0.81}P_{0.19}$	2750 nm	p = 5x10^{18} cm^{-3}
GaAs buffer	200 nm	p = 5x10^{18} cm^{-3}
GaAs substrate		p > 1x10^{18} cm^{-3}

Changed the composition of the Fabry Perot spacing layer

GaAs	5 nm	p = 5x10^{19} cm^{-3}
$GaAs_{0.62}P_{0.38}$	4 nm	p = 5x10^{17} cm^{-3}
GaAs	4 nm	p = 5x10^{17} cm^{-3}
$Al_{0.15}Ga_{0.85}As_{0.81}P_{0.19}$	300 nm	p = 5x10^{18} cm^{-3}
$AlAs_{0.78}P_{0.22}$	65 nm	p = 5x10^{18} cm^{-3}
$GaAs_{0.81}P_{0.19}$	55 nm	p = 5x10^{18} cm^{-3}
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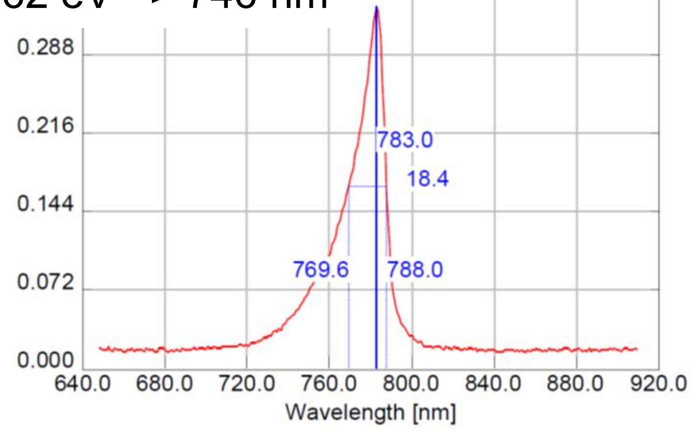
GaAs->$GaAs_{0.81}P_{0.19}$	2750 nm	p = 5x10^{18} cm^{-3}
GaAs buffer	200 nm	p = 5x10^{18} cm^{-3}
GaAs substrate		p > 1x10^{18} cm^{-3}

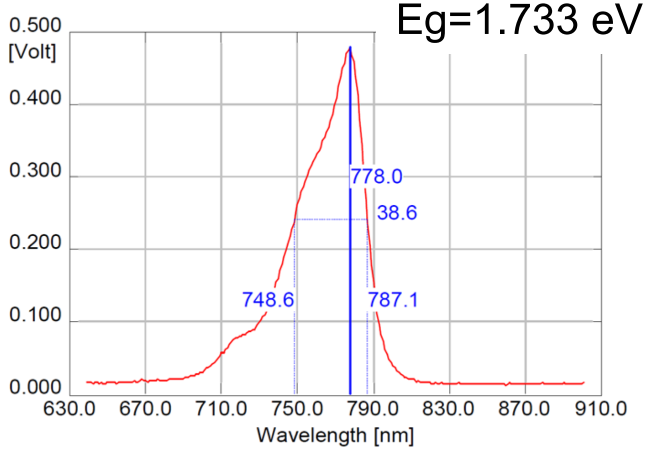
The increased band gap increases the energy required for electrons excitation in this layer

Eg=1.662 eV => 746 nm

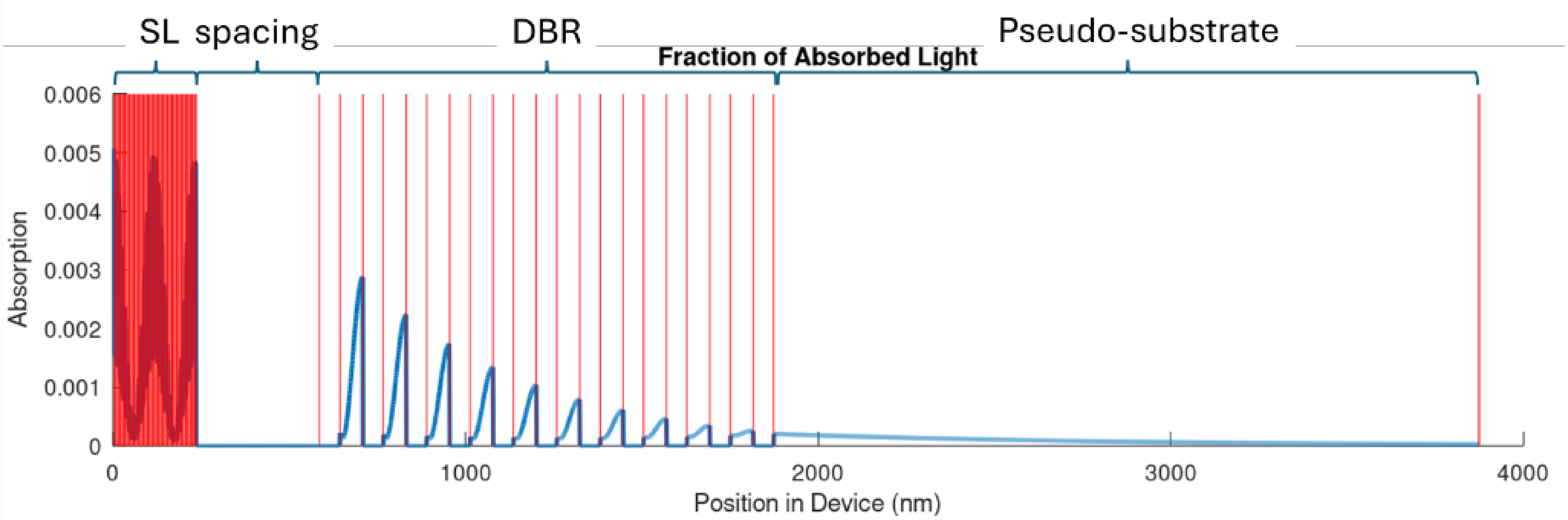


Photoluminescence measurements shows expected longer tail at shorter wavelengths

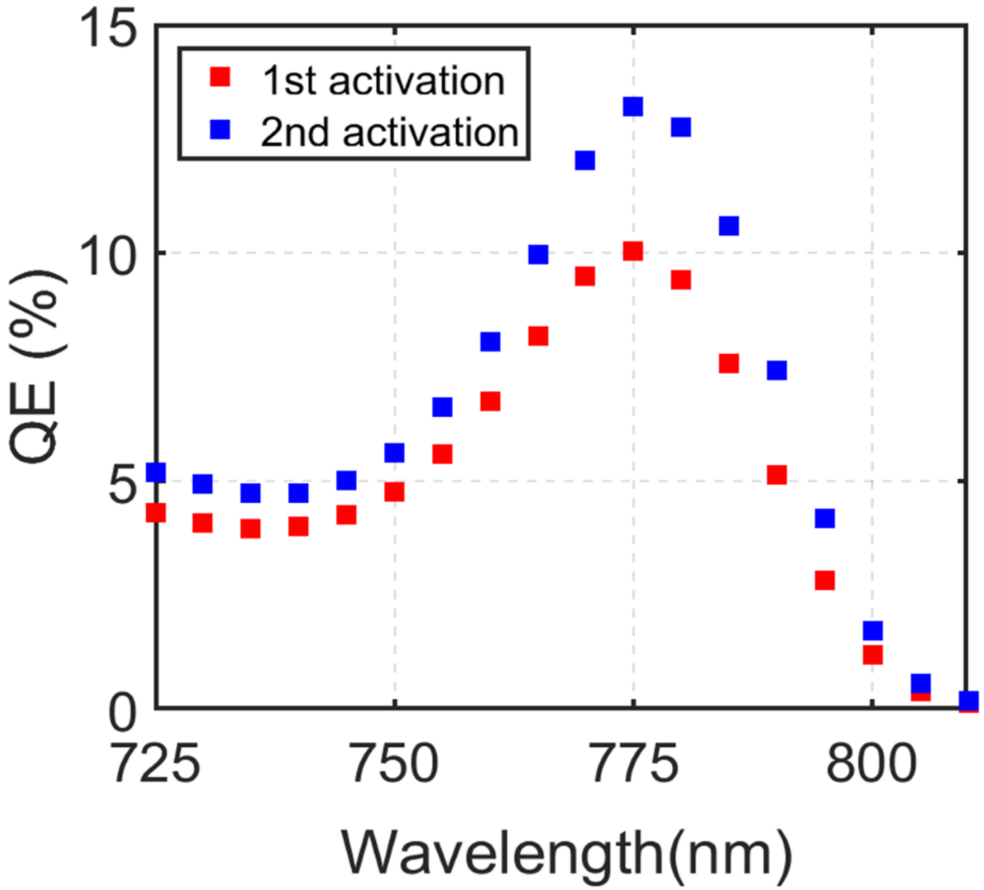
Eg=1.733 eV => 715 nm

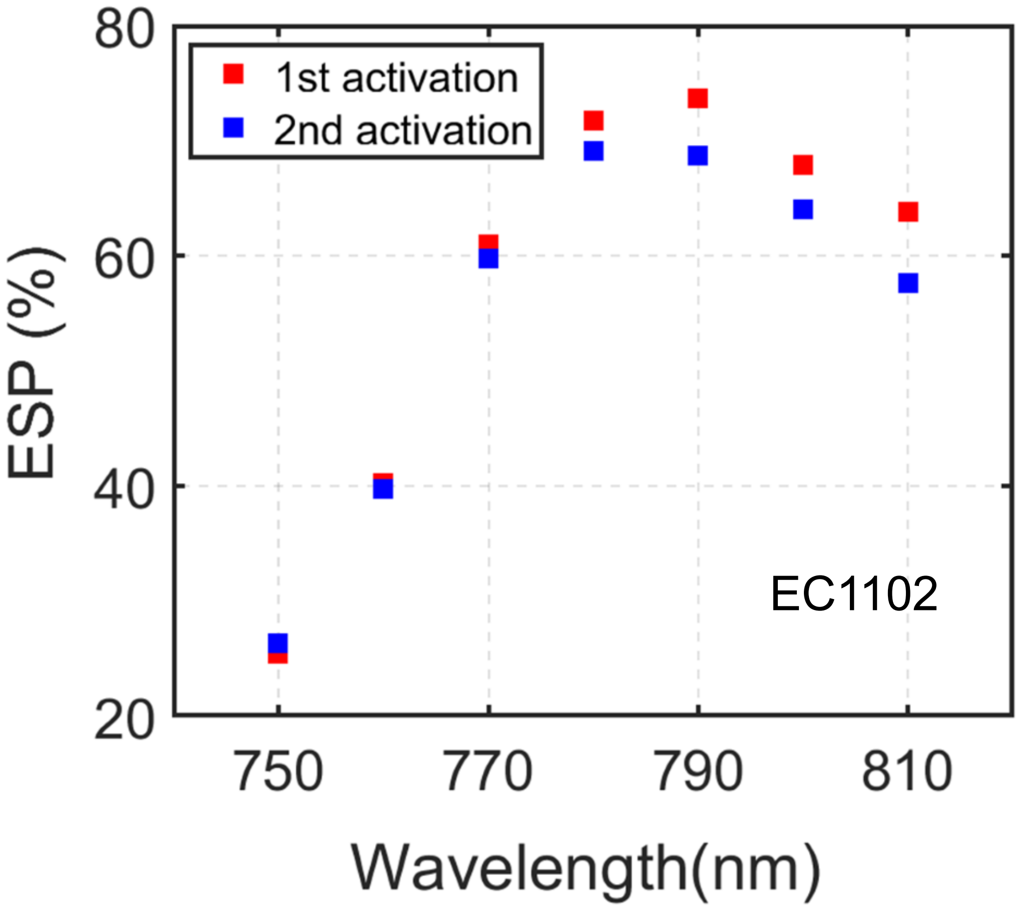


Optical absorption within the structure



QE and ESP from 2nd iteration





We still observe very high QEs but not the expected improvement in the ESP

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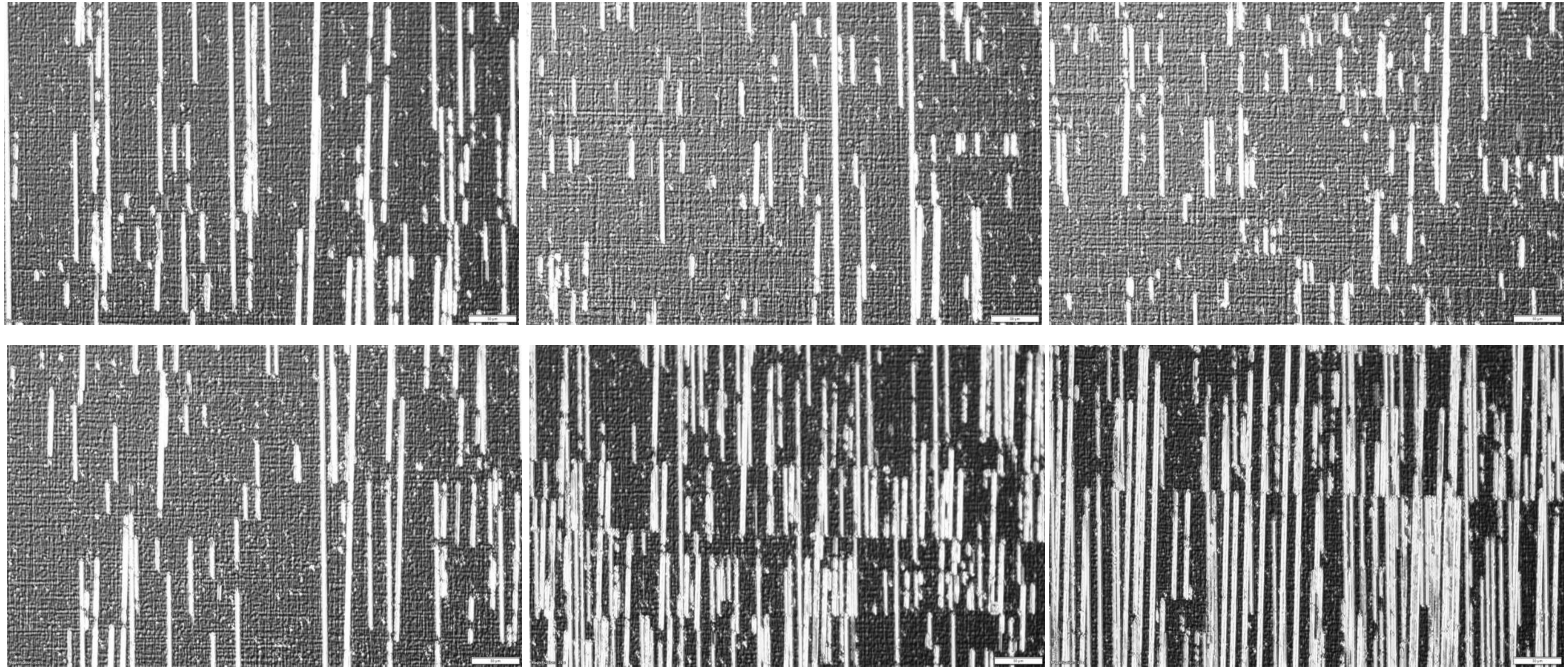
Morphology of the samples

High density of slip-planes possibly from thread dislocations
Originated on the underlying DBR layers?
TBC from Transmission Electron Microscopy measurement



Known challenge when growing samples on pseudo-substrates like the $GaAs_{0.81}P_{0.19}$

Courtesy of A. Muhowski

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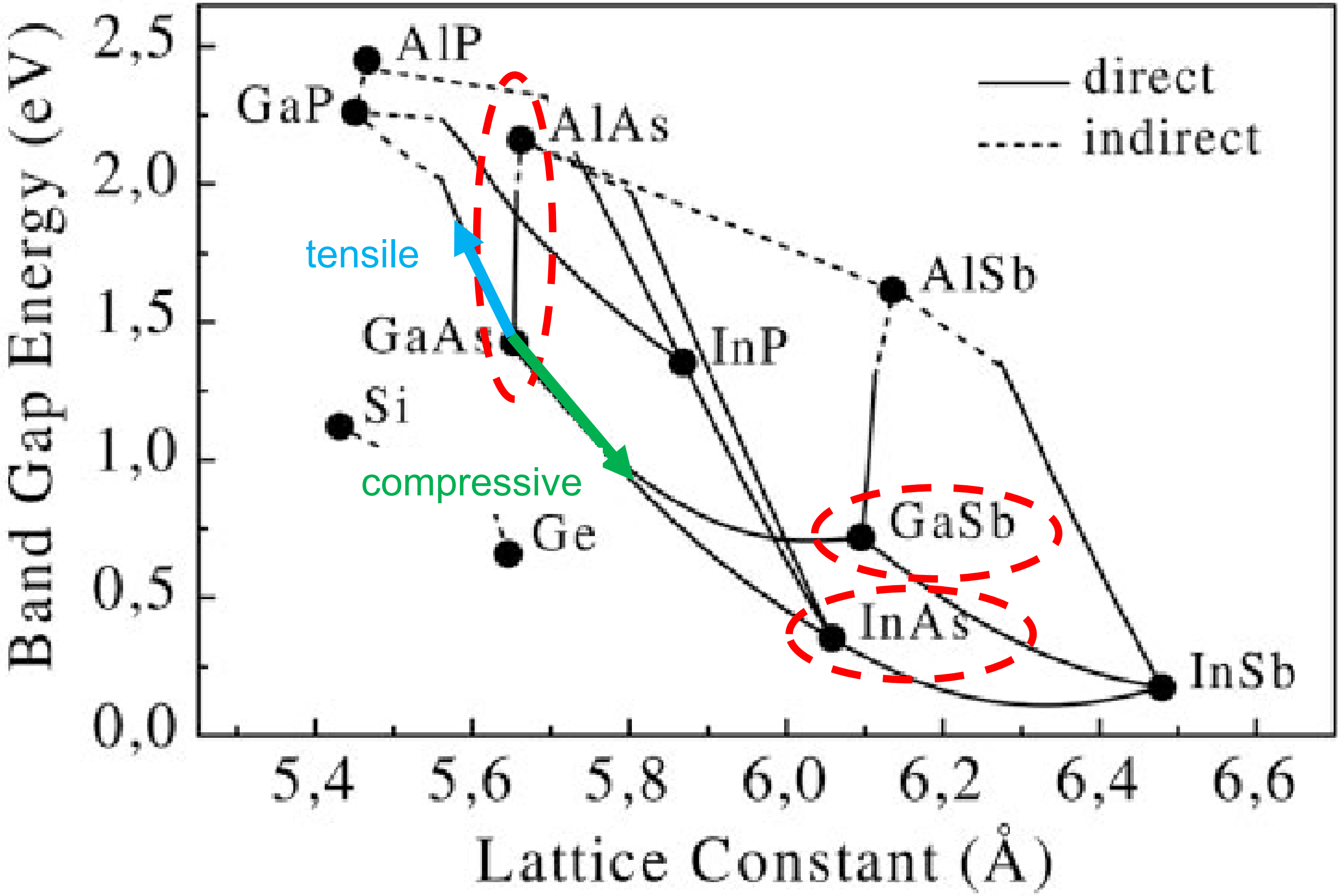
Can we use GaAs pristine surface to grow upon it?



Grow directly on GaAs

- No need to grow the pseudo-substrate
- Less material to be grown
- Higher substrate quality

DBR made out with GaAs/AlAs

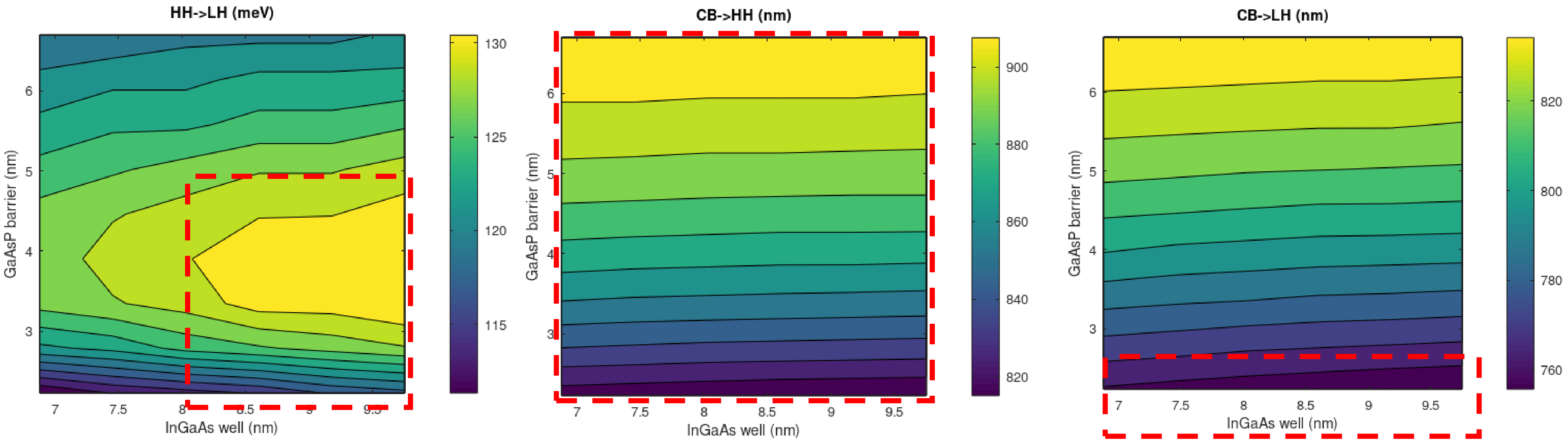
- Binary compounds

Two option for the SL layers

- GaAsP/InGaAs
- GaAsP/GaAsSb

SL based on $GaAs_{0.62}P_{0.38}/In_{0.20}Ga_{0.80}As$

Parametric studies varying the width of barrier and wells in strain compensated SL



HH-LH separation is large
=> high ESP

(about 2x w.r.t. our SL-DBR
which has ~70 meV)

CB-HH => Absorption edge

Larger than 800 nm is good as it
ensure large QE @800nm

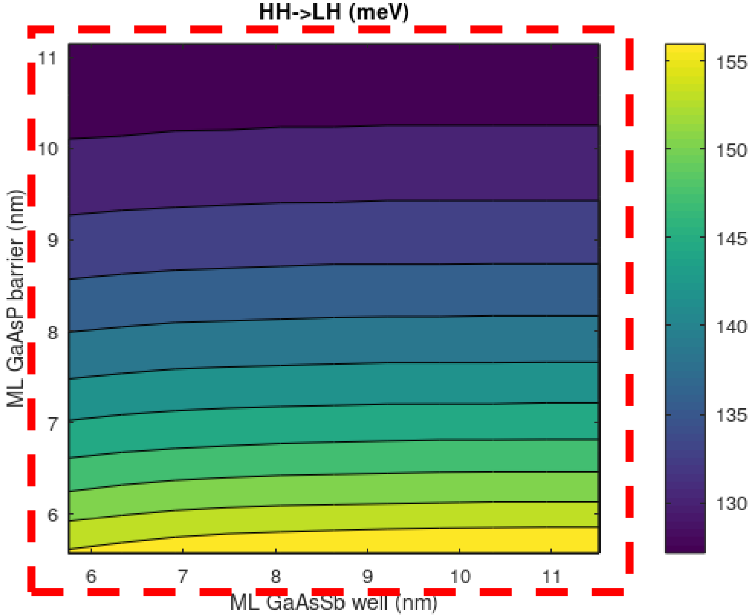
CB-LH => onset of ESP
depolarization

We wanted to have this to be
shorter than 800 nm

SL based on $GaAs_{0.60}P_{0.40}/GaAs_{0.75}Sb_{0.25}$

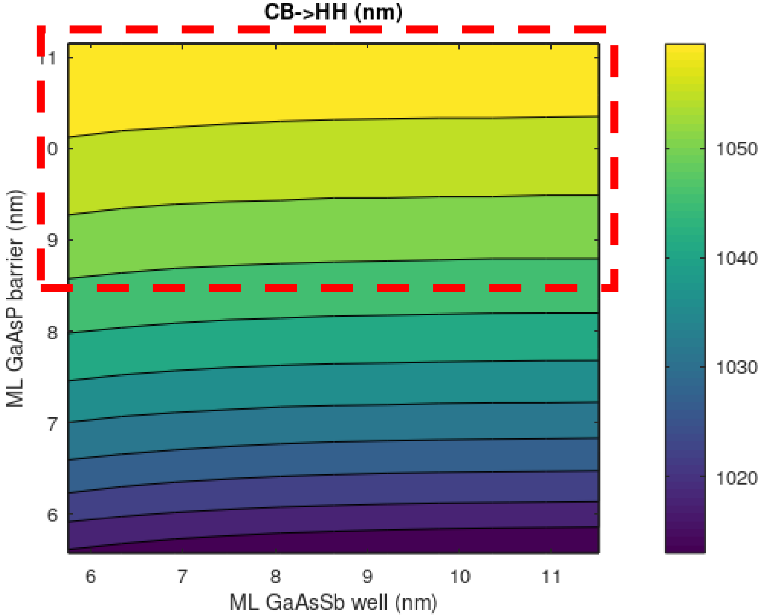
Parametric studies varying the width of barriers and wells in strain compensated SL

Designed to be conveniently operated with fiber lasers at 1030nm



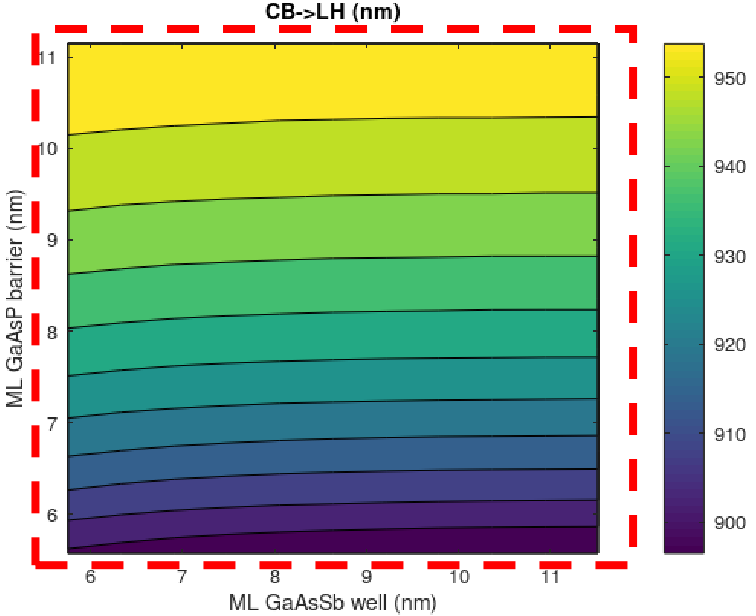
HH-LH (>130 meV)
⇒ high ESP

-2x the currently used SL-



SL band edge is larger than 1030 nm

Ensure large optical absorption for Yb fiber laser operation



Wavelength for the onset of ESP depolarization is shifted towards wavelengths much shorter than 1030 nm

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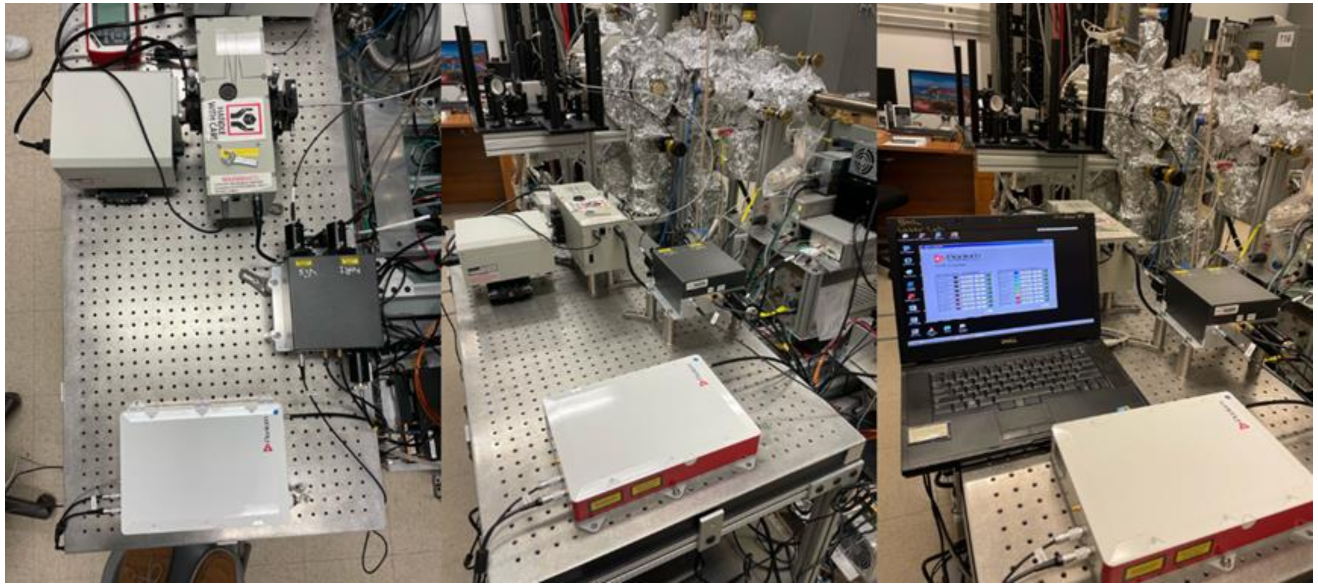
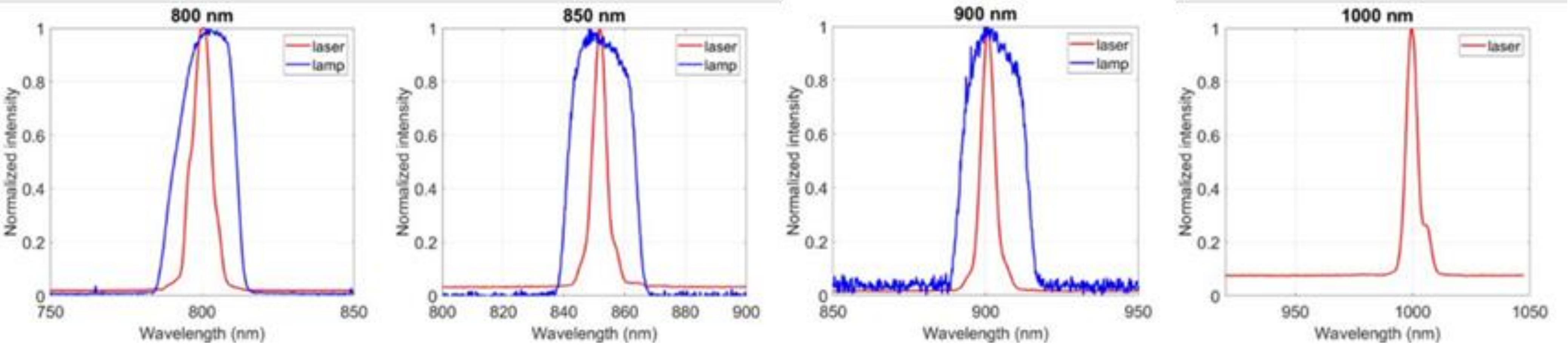
Optical system upgrade



- The intensity vs. bandwidth of our lamp system is not optimal.
- The tungsten lamp does not provide enough photons at wavelength larger than 900 nm.
- We installed and re-commissioned a supercontinuum laser which provides more photons and a narrower bandwidth for next measurement



Conclusions

- Several robust activating layers yielded longer dark lifetimes
- QE and ESP tradeoffs call for higher QEs to increase operational lifetime
- Achieved record high QE from a SL DBR with high ESP
- Lattice matching on GaAs can simplify growth
  - GaAsP/InGaAs operated at ~800 nm
  - GaAsP/GaAsSb operated at ~1030 nm

Collaboration	Wavelength (nm)	QE (%)	ESP (%)	FoM ($QE*ESP^2$)
Nagoya/KEK/LBNL	780	1.6	92	1.35
SVT/Jlab	775	6.4	84	4.51
BNL/SNL rev.1	775	18.8	66	8.18
BNL/SNL rev.2	780	9	72	4.66

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Thank you!

- **BNL team:** P. Saha, M. Bouchickha, T. Tsang, J. Walsh, R. Begay
- **SNL team:** A. Muhowski, S. Hawkins, V. Patel, M. McDonough

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