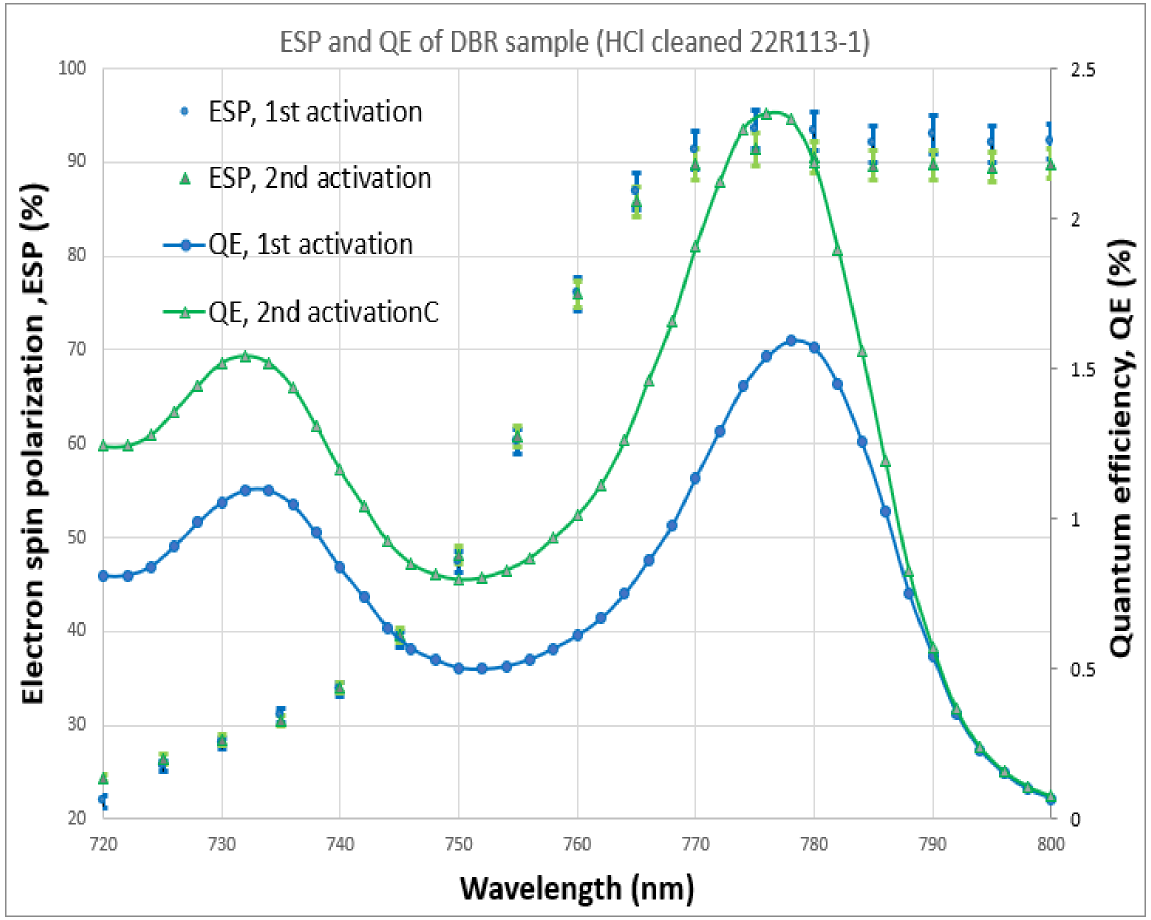
Photocathodes with 90% polarization and QE greater than 1% for DOE NP

Annual NP Accelerator R&D virtual PI Exchange meeting - November 29, 2022



Matt Poelker and M. Stutzman, Jefferson Lab
Sylvain Marsillac, ODU
Erdong Wang, BNL

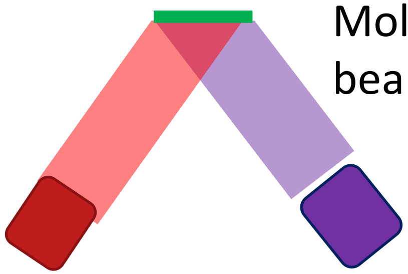
MBE, GSMBE, CBE and MOCVD (aka MOVPE)

MBE

Gas Source
Molecular Beam
Epitaxy

elemental As, P, Ga

- Pressure ~10^{-8} mbar
- Growth rates ~ 1 µm/hr
- Very precise control

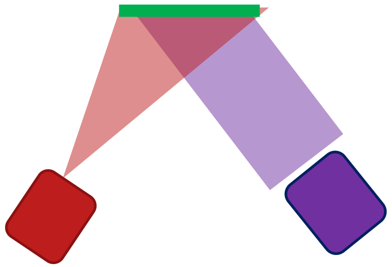


Molecular beams

GSMBE

Gas Source
Molecular Beam
Epitaxy

AsH_3, PH_3, elemental Gallium



Molecular and gas sources

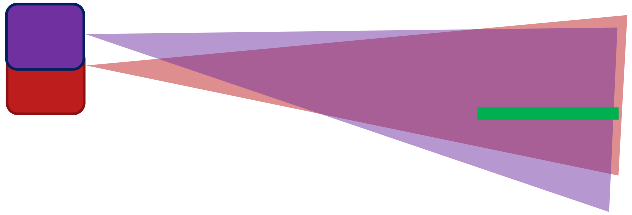
CBE

Chemical Beam Epitaxy

AsH_3, PH_3, triethyl gallium (TEGa) or elemental Gallium

- Pressure <10^{-4} mbar
- Growth rates 0.5-1 µm/hr

Gas sources



MOCVD

Metal organic chemical vapor deposition (metal organic vapor phase epitaxy)

AsH_3, PH_3, trimethylgallium (TMGa)

- Pressures >100 mbar during growth
- Growth Rates 10 µm/hr
- Some claim difficult to get sharp interfaces

Background

- SPIRE/Bandwidth Semiconductor used MOCVD to grow single-strained-layer GaAs/GaAsP photocathodes (good)
- SVT used MBE to grow strained-superlattice GaAs/GaAsP photocathodes (better)
- In this work - JLab, ODU and BNL - focus on MOCVD
  - Strained Superlattice photocathodes
  - Strained Superlattice photocathodes with DBR
- We were granted 3 months no-cost extension, project officially ends 12/31/2022

Project Goals

	Tasks Year 1	**Q1**	**Q2**	**Q3**	**Q4**
✓	Calibration of p-$GaAs_{0.65}P_{0.35}$ (ODU)	🟦			
✓	Calibration of metamorphic grade from GaAs to $GaAs_{0.65}P_{0.35}$ (ODU)	🟦			
✓	Calibration of $GaAs/GaAs_{0.65}P_{0.35}$ strained superlattice (ODU)		🟦		
✓	**Mott system: assemble and pump down** (BNL)	🟦	🟦	🟦	
✓	**System commissioning and calibration** (BNL & J✗b)	🟦	🟦	🟦	
✓	Fabrication runs strained-superlattice (ODU & JLab & BNL)			🟦	
✓	Strained-superlattice Photocathodes **Evaluation (JLab)**				🟦

	Tasks Year 2	**Q1**	**Q2**	**Q3**	**Q4**
✓	Calibration of p-$AlAs_{0.6}P_{0.4}$ (ODU)	🟩			
✓	Calibration of $GaAs_{0.65}P_{0.35}$ /$AlAs_{0.6}P_{0.4}$ DBR (ODU)		🟩		
✓	Strained-superlattice Photocathodes Evaluation (BNL)	🟩	🟩	🟩	
✓	Fabrication runs strained superlattice with DBR (ODU & JLab & BNL)			🟩	
✗	**Measure CsTe/CsI coated SL-GaAs** (BNL)		🟩	🟩	🟩
✓	Strained superlattice/DBR Photocathodes Evaluation (J✗b & BNL)				🟩

Project Conclusion

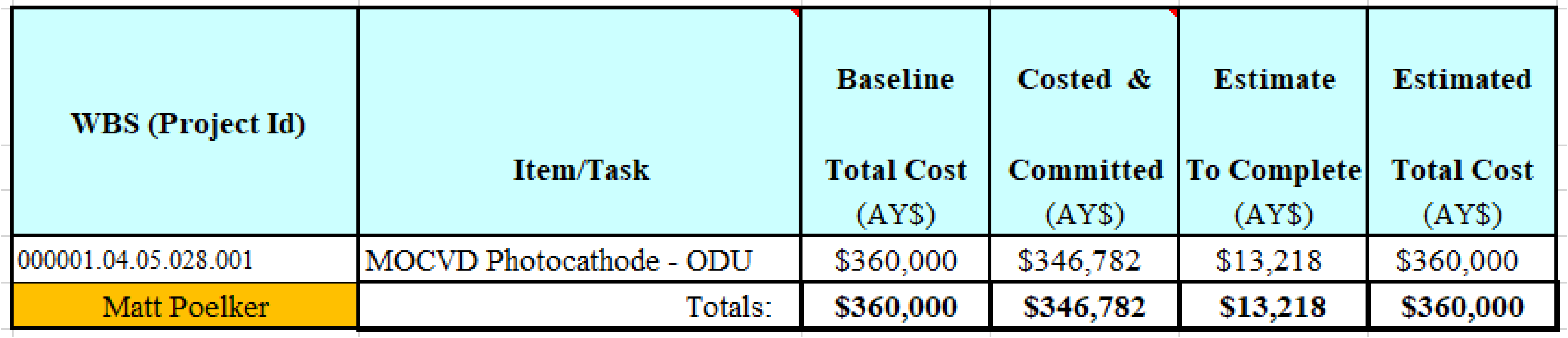
Stated clearly:

- We successfully fabricated both kinds of photocathodes, SSL and SSL with DBR, via MOCVD. Exceptional work by ODU
- We achieved our central goal: Polarization > 90% and QE > 1%
- Between JLab and BNL, we have nearly 200k$ worth of photocathode material, our photocathode stores now replenished! Enough material for polarimeter cross checks, to support CEBAF and EIC physics programs, and to support polarized source R&D
- BNL has a fully functional microMott polarimeter, and this device was essential for project success
- JLab’s microMott polarimeter is very close to being operational again
- The cross calibration of the two microMott polarimeters will happen
- Having both source groups – JLab and BNL – participating in this project has been wonderful, a big selling point (IMHO)

Budget

JLab FY20 + FY21 loaded budget = 360k$

- Support for ODU
- Procurements: sample evaluation, Mott apparatus upkeep
- Some labor for Matt and Marcy

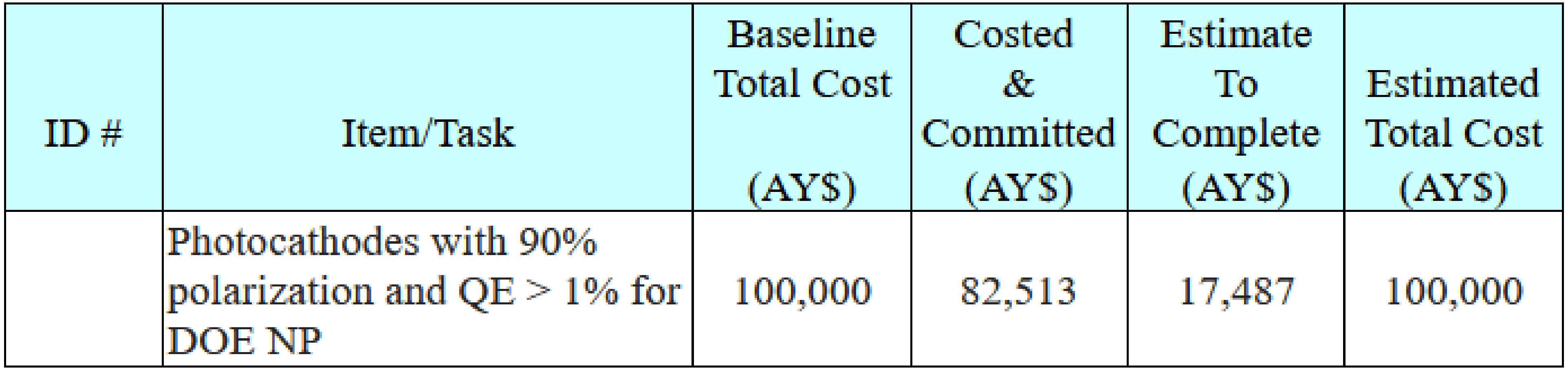


WBS (Project Id)	Item/Task	Baseline Total Cost (AY$)	Costed & Committed (AY$)	Estimate To Complete (AY$)	Estimated Total Cost (AY$)
000001.04.05.028.001	MOCVD Photocathode - ODU	$360,000	$346,782	$13,218	$360,000
Matt Poelker	Totals:	**$360,000**	**$346,782**	**$13,218**	**$360,000**

Budget

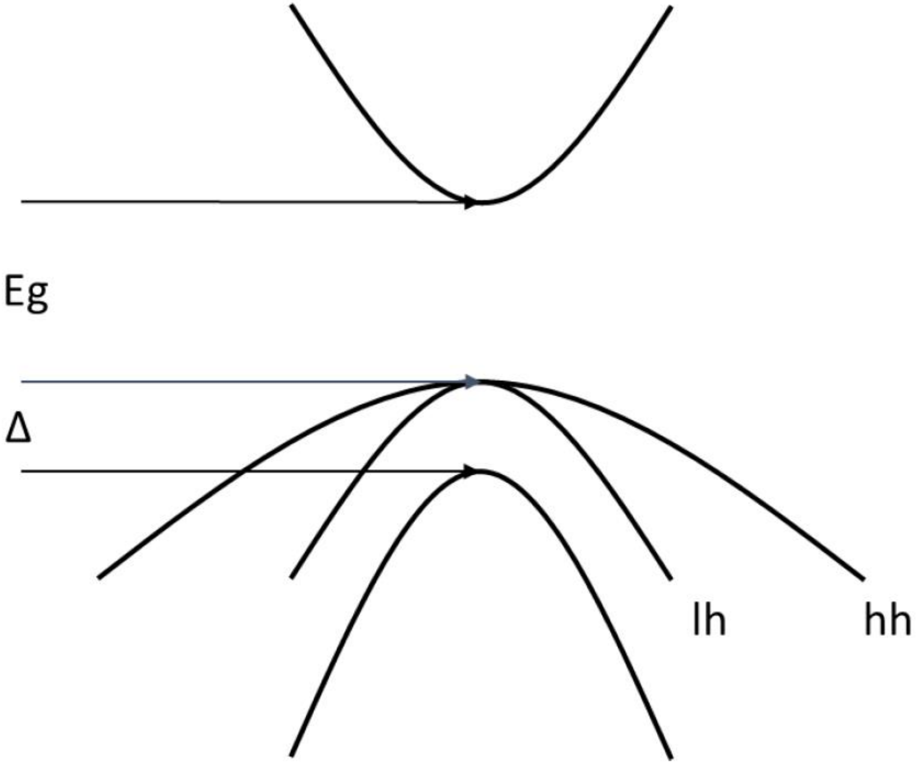
BNL FY20 + FY21 loaded budget = 100k$

- Sample evaluation
- Mott apparatus upkeep

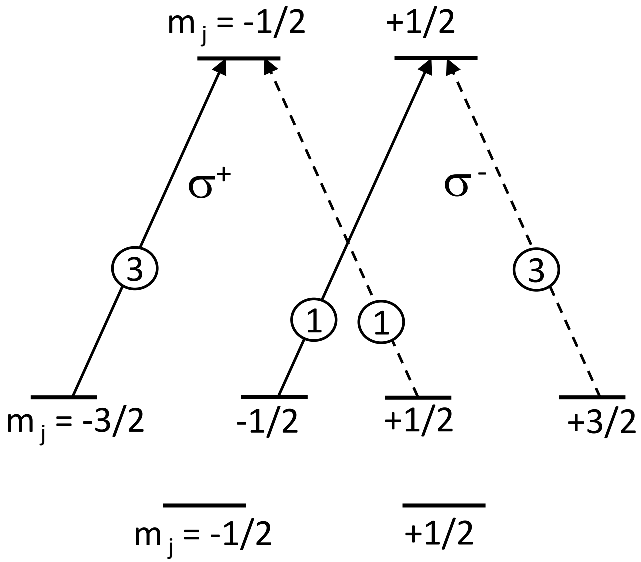


ID #	Item/Task	Baseline Total Cost (AY$)	Costed & Committed (AY$)	Estimate To Complete (AY$)	Estimated Total Cost (AY$)
	Photocathodes with 90% polarization and QE > 1% for DOE NP	100,000	82,513	17,487	100,000

Making polarized electron beams with GaAs

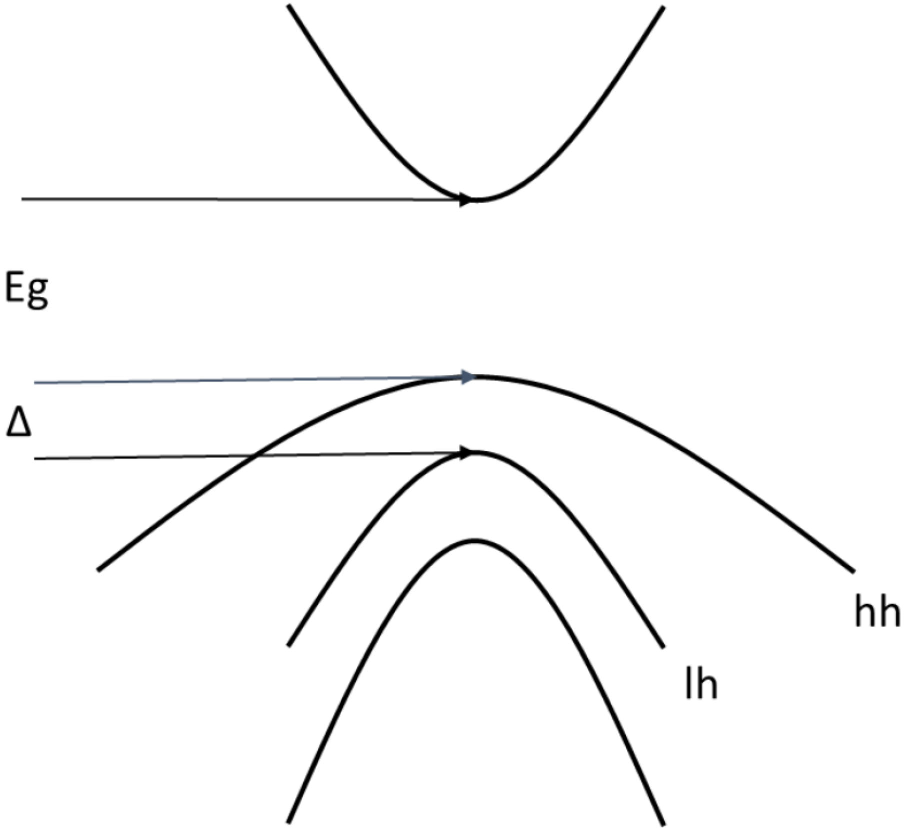


Bulk GaAs – no strain

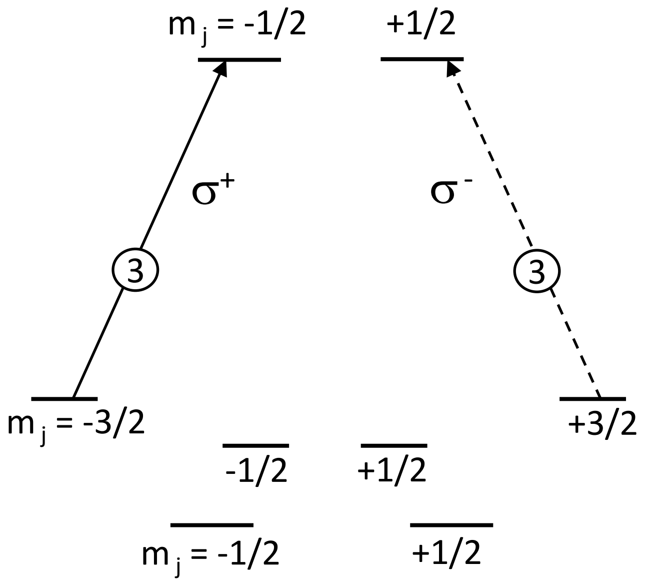


Maximum Polarization 50%

Making polarized electron beams with GaAs



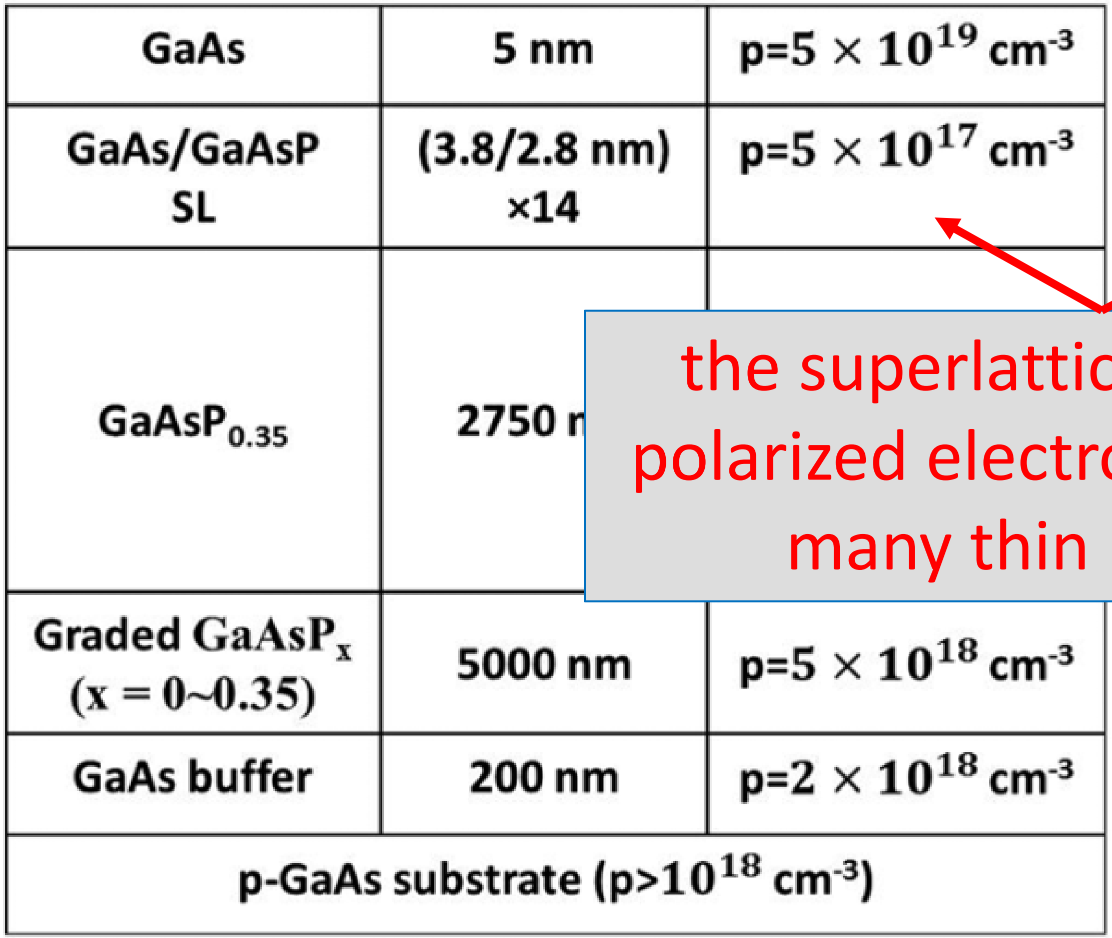
Strained GaAs



Maximum Polarization 100%

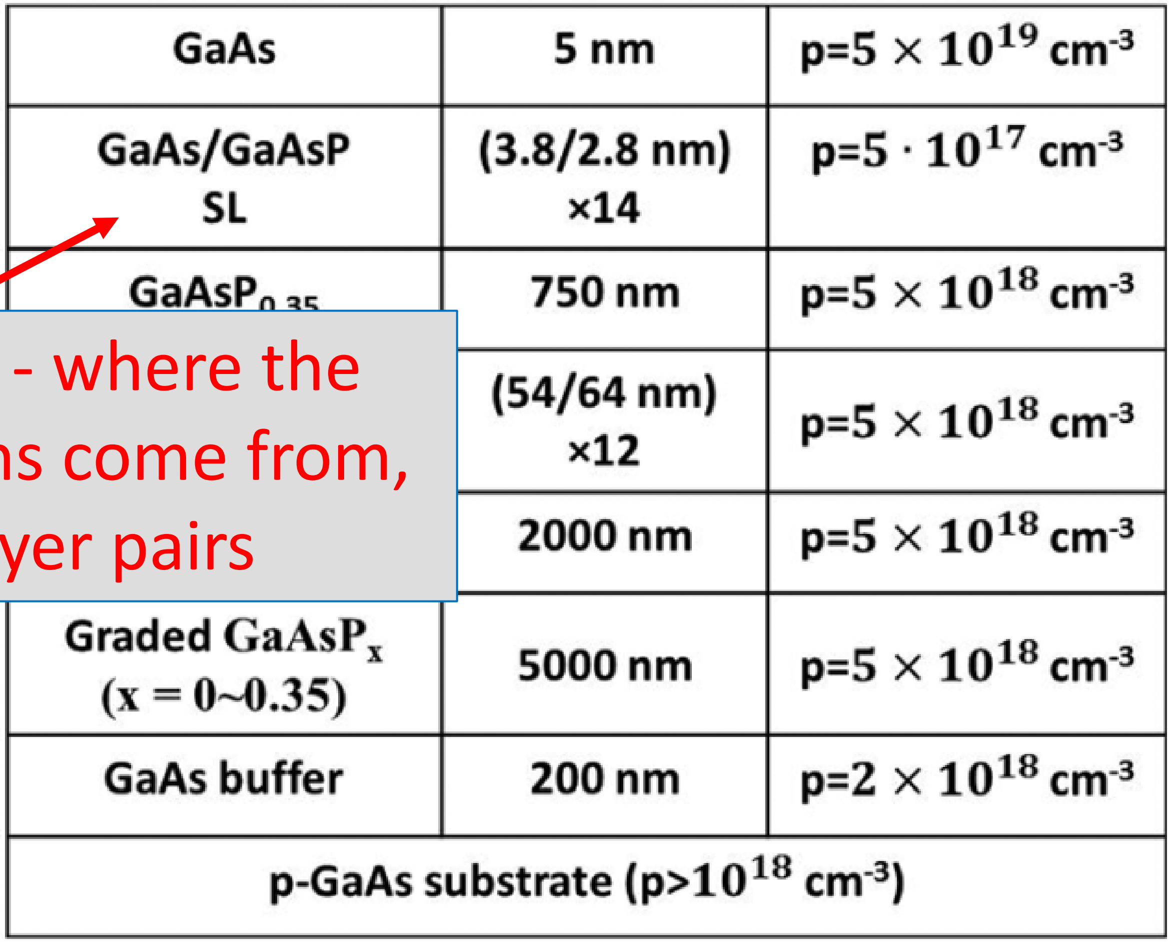
Device Structure

Strained Superlattice



GaAs	5 nm	p=5 $\times 10^{19}$ cm^{-3}
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	p=5 $\times 10^{17}$ cm^{-3}
GaAsP$_{0.35}$	2750 n	
Graded GaAsP$_x$ $(x = 0\sim0.35)$	5000 nm	p=5 $\times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	p=2 $\times 10^{18}$ cm^{-3}
p-GaAs substrate (p>10^{18} cm^{-3})		

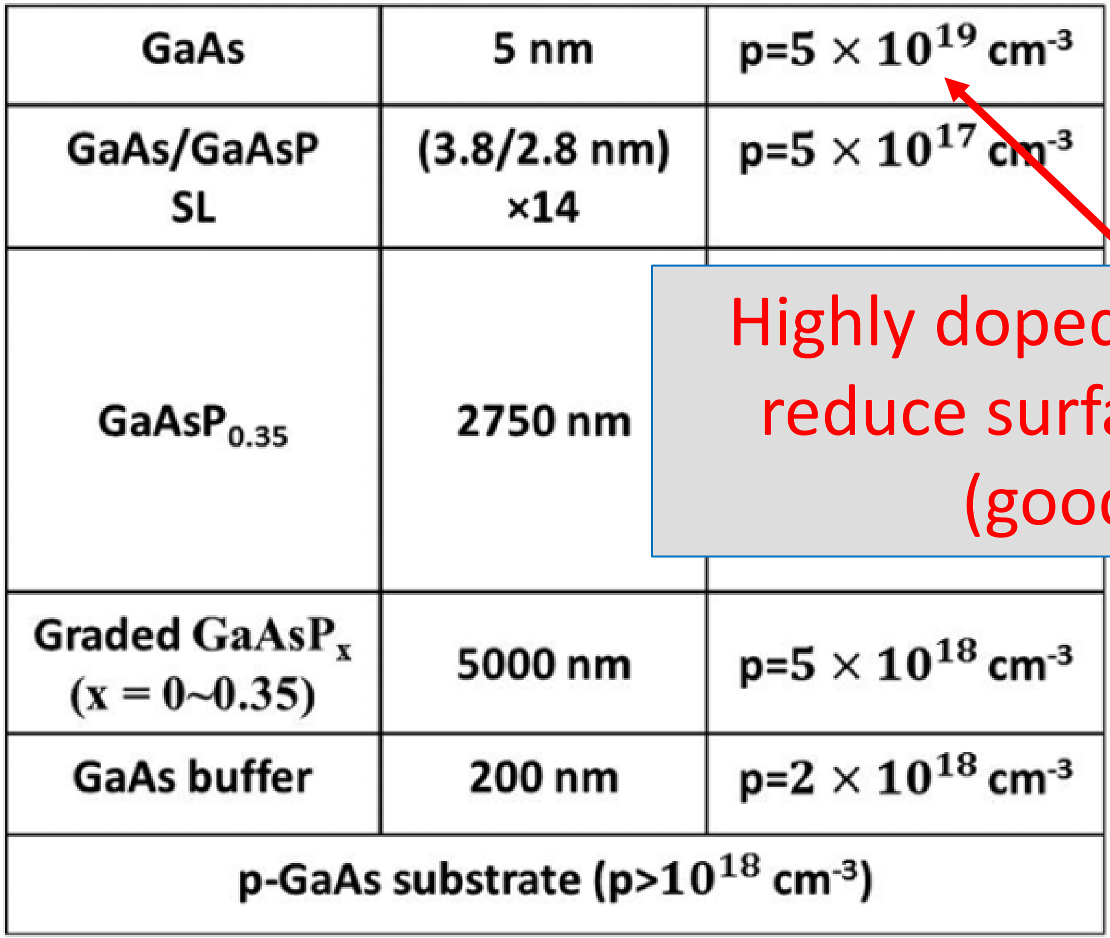
Strained Superlattice with Distributed Bragg Reflector



GaAs	5 nm	p=5 $\times 10^{19}$ cm^{-3}
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	p=5 $\cdot 10^{17}$ cm^{-3}
GaAsP	750 nm	p=5 $\times 10^{18}$ cm^{-3}
	(54/64 nm) ×12	p=5 $\times 10^{18}$ cm^{-3}
	2000 nm	p=5 $\times 10^{18}$ cm^{-3}
Graded GaAsP$_x$ $(x = 0\sim0.35)$	5000 nm	p=5 $\times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	p=2 $\times 10^{18}$ cm^{-3}
p-GaAs substrate (p>10^{18} cm^{-3})		

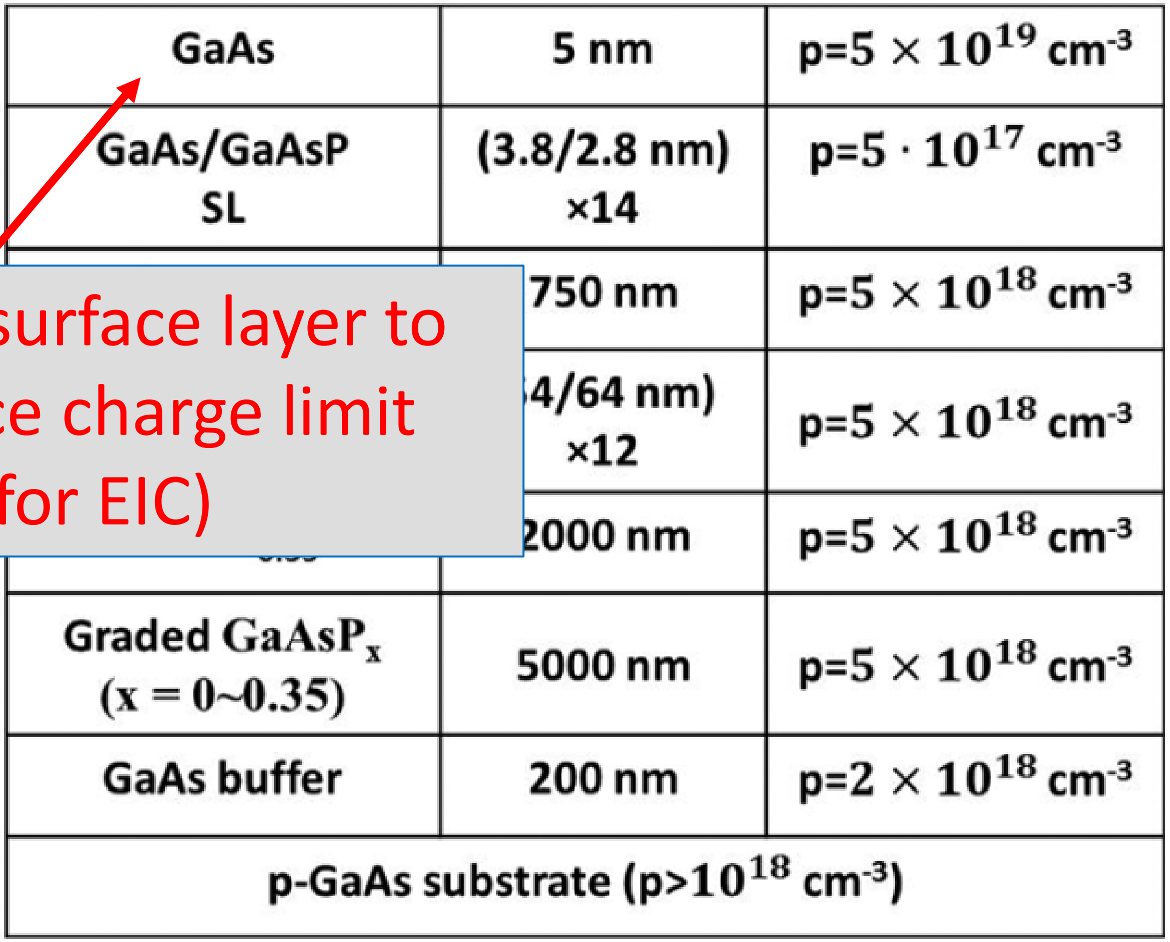
the superlattice - where the polarized electrons come from, many thin layer pairs

Device Structure



Strained Superlattice

GaAs	5 nm	$p=5 \times 10^{19}\ \mathrm{cm^{-3}}$
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	$p=5 \times 10^{17}\ \mathrm{cm^{-3}}$
$GaAsP_{0.35}$	2750 nm	
Graded $GaAsP_x$ ($x = 0\sim0.35$)	5000 nm	$p=5 \times 10^{18}\ \mathrm{cm^{-3}}$
GaAs buffer	200 nm	$p=2 \times 10^{18}\ \mathrm{cm^{-3}}$
p-GaAs substrate ($p>10^{18}\ \mathrm{cm^{-3}}$)		



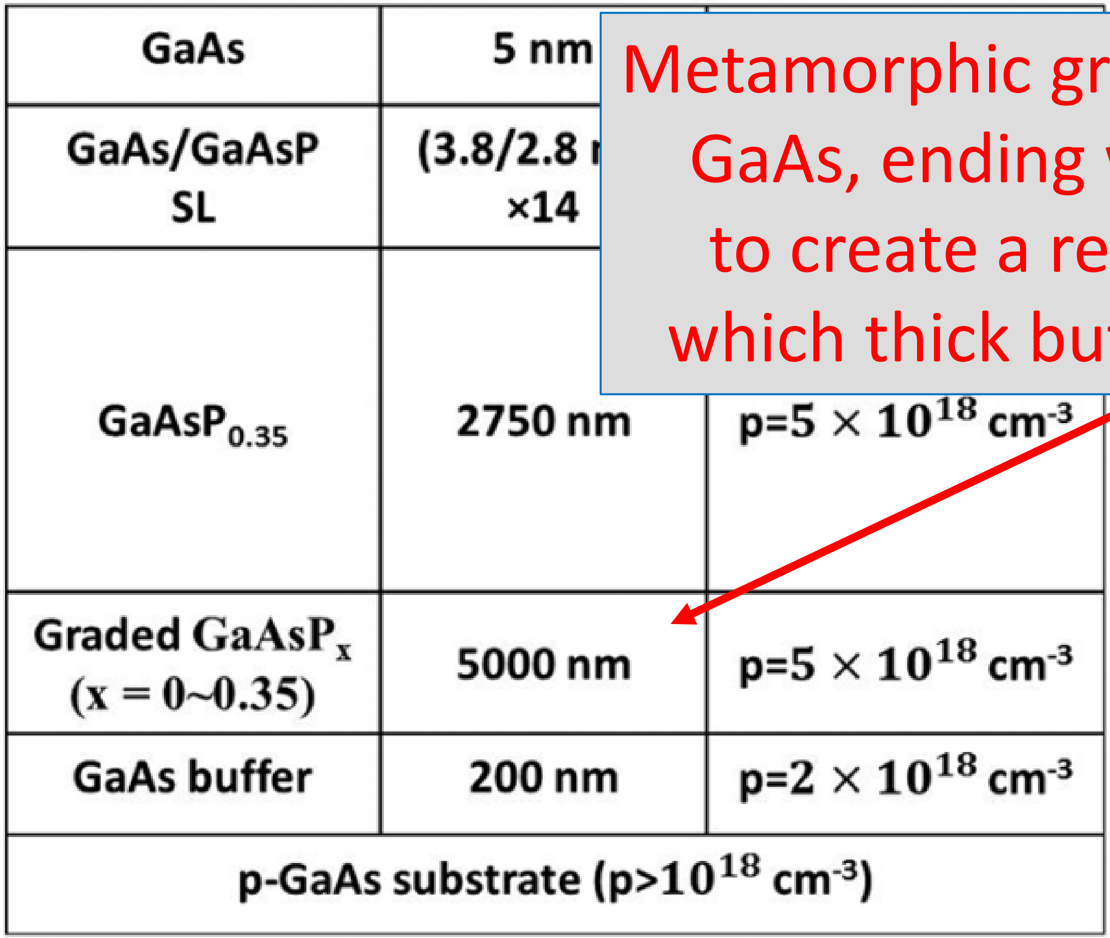
Strained Superlattice with Distributed Bragg Reflector

GaAs	5 nm	$p=5 \times 10^{19}\ \mathrm{cm^{-3}}$
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	$p=5 \cdot 10^{17}\ \mathrm{cm^{-3}}$
	750 nm	$p=5 \times 10^{18}\ \mathrm{cm^{-3}}$
	4/64 nm) ×12	$p=5 \times 10^{18}\ \mathrm{cm^{-3}}$
0.35	2000 nm	$p=5 \times 10^{18}\ \mathrm{cm^{-3}}$
Graded $GaAsP_x$ ($x = 0\sim0.35$)	5000 nm	$p=5 \times 10^{18}\ \mathrm{cm^{-3}}$
GaAs buffer	200 nm	$p=2 \times 10^{18}\ \mathrm{cm^{-3}}$
p-GaAs substrate ($p>10^{18}\ \mathrm{cm^{-3}}$)		

Highly doped surface layer to reduce surface charge limit (good for EIC)

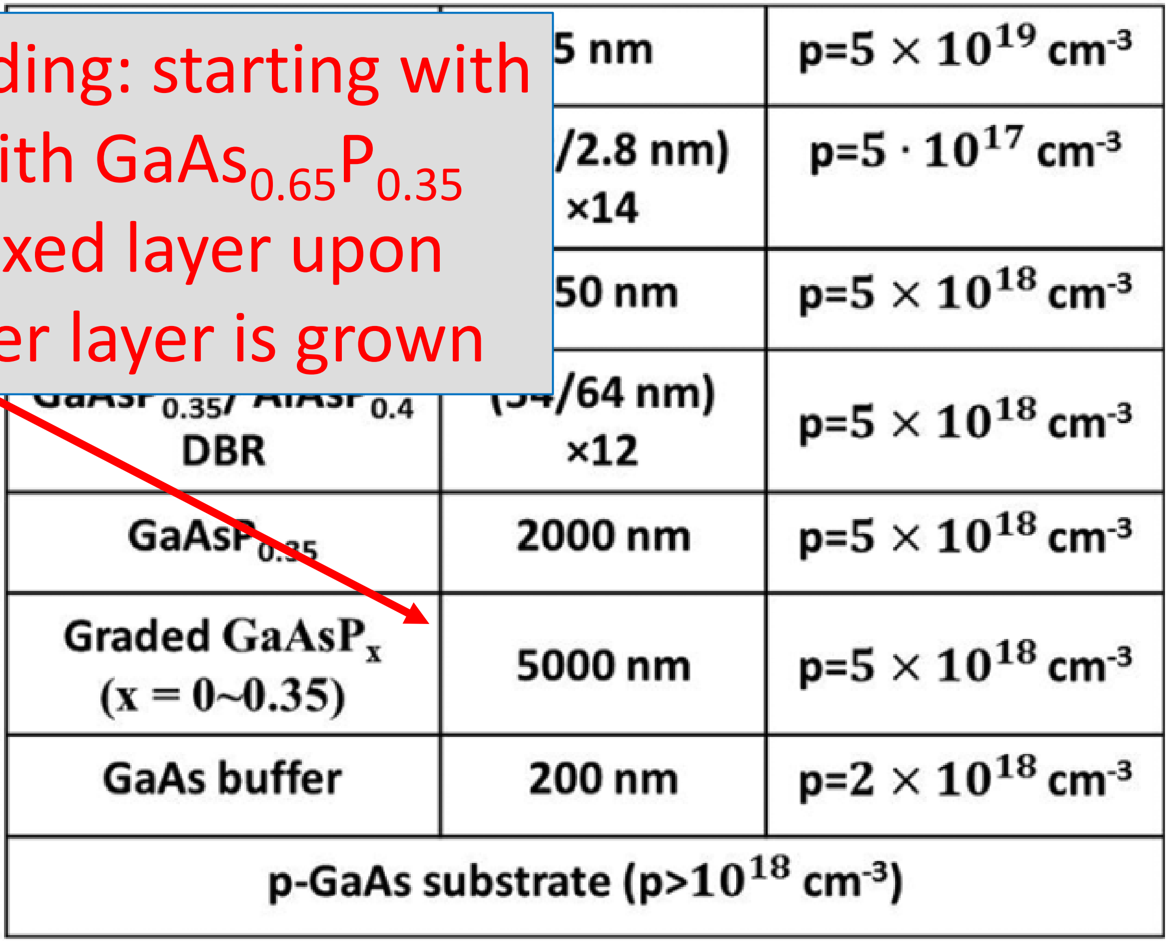
Device Structure

Strained Superlattice



GaAs	5 nm	
GaAs/GaAsP SL	(3.8/2.8 ×14	
$GaAsP_{0.35}$	2750 nm	$p=5 \times 10^{18}$ cm^{-3}
Graded $GaAsP_x$ $(x = 0 \sim 0.35)$	5000 nm	$p=5 \times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	$p=2 \times 10^{18}$ cm^{-3}
p-GaAs substrate ($p>10^{18}$ cm^{-3})		

Strained Superlattice with Distributed Bragg Reflector



	5 nm	$p=5 \times 10^{19}$ cm^{-3}
	/2.8 nm) ×14	$p=5 \cdot 10^{17}$ cm^{-3}
	50 nm	$p=5 \times 10^{18}$ cm^{-3}
$GaAsP_{0.35}/AlAsP_{0.4}$ DBR	(54/64 nm) ×12	$p=5 \times 10^{18}$ cm^{-3}
$GaAsP_{0.35}$	2000 nm	$p=5 \times 10^{18}$ cm^{-3}
Graded $GaAsP_x$ $(x = 0 \sim 0.35)$	5000 nm	$p=5 \times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	$p=2 \times 10^{18}$ cm^{-3}
p-GaAs substrate ($p>10^{18}$ cm^{-3})		

Metamorphic grading: starting with GaAs, ending with $GaAs_{0.65}P_{0.35}$ to create a relaxed layer upon which thick buffer layer is grown

Device Structure

Strained Superlattice

GaAs	5 nm	p=5 $\times 10^{19}$ cm^{-3}
GaAs/GaAsP	(3.8/2.8 nm)	p=5 $\times 10^{17}$ cm^{-3}
		cm^{-3}
Graded GaAsP$_x$ ($x = 0{\sim}0.35$)	5000 nm	p=5 $\times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	p=2 $\times 10^{18}$ cm^{-3}
p-GaAs substrate (p>10^{18} cm^{-3})		

Another complicated superlattice which forms the distributed Bragg reflector (DBR)

Strained Superlattice with Distributed Bragg Reflector

GaAs	5 nm	p=5 $\times 10^{19}$ cm^{-3}
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	p=5 $\cdot 10^{17}$ cm^{-3}
GaAsP$_{0.35}$	750 nm	p=5 $\times 10^{18}$ cm^{-3}
GaAsP$_{0.35}$/ AlAsP$_{0.4}$ DBR	(54/64 nm) ×12	p=5 $\times 10^{18}$ cm^{-3}
GaAsP$_{0.35}$	2000 nm	p=5 $\times 10^{18}$ cm^{-3}
Gr		
p-GaAs substrate (p>10^{18} cm^{-3})		

Etalon formed between DBR and top surface: stores light, more light absorbed, higher QE

MOCVD growth parameters

- Key Precursors
  - Trimethyl Gallium ($Ga(CH_3)_3$)
  - Arsine (AsH_3) and Phosphine (PH_3)
  - Diethyl Zinc ($Zn(CH_3CH_3)_2$)
  - Carbon Tetrachloride (CCl_4)
  - Lower diffusivity of carbon in GaAsP should improve lifetime of device surface
- Substrate: 2” GaAs wafers with either 0 or 2^o offcut in the 110 direction
- Growth rate range: 3-8 μm/hr
- Temperature: 650-750^{o}C

MOCVD system at Rochester Institute of Technology

Issues that needed to be resolved

- Sample temperature and growth rate for best metamorphic grading
- Phosphorus concentration for optimum strain and polarization
- Defect mitigation
- Cleave plane orientation, offcut or no offcut

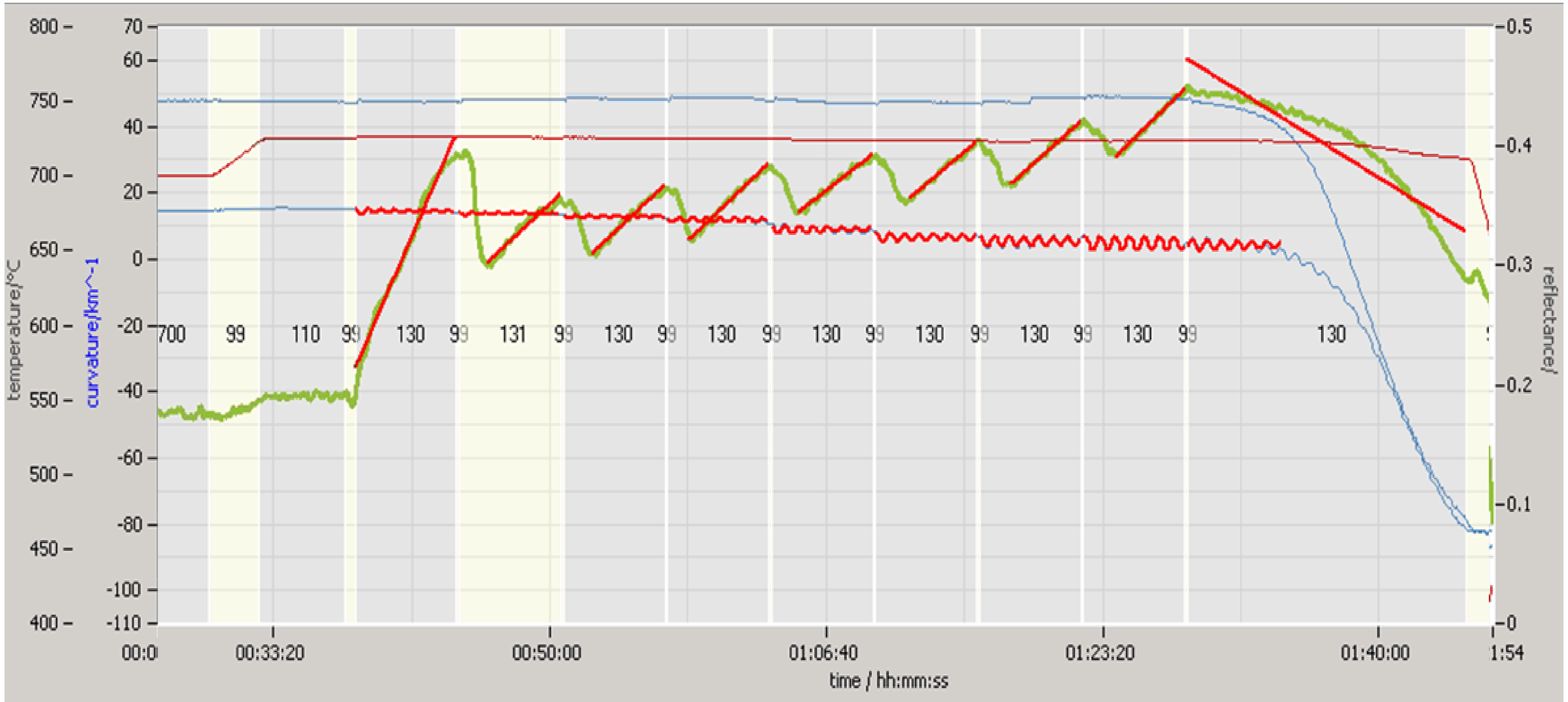
Tools Used

- Telemetry: in-situ measurement
  - Pyrometry
  - Curvature
  - Reflectivity

🟥 Temperature

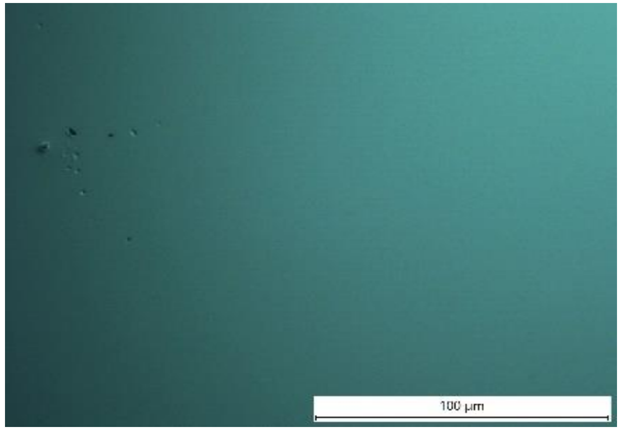
🟦 Reflectivity

🟩 Curvature

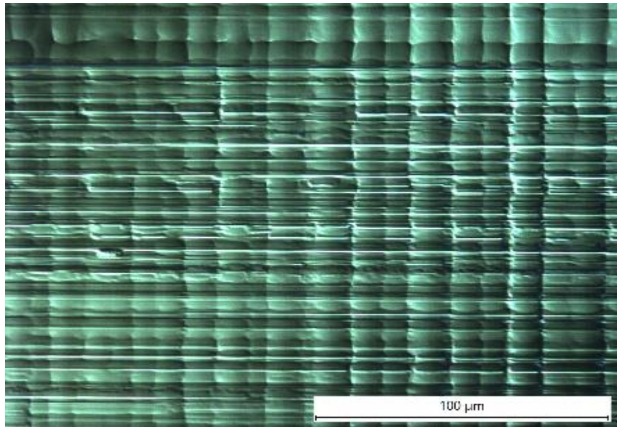


Tools Used

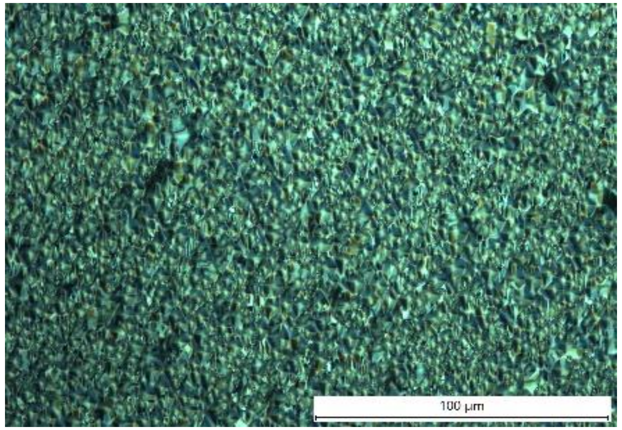
- Nomarski Microscopy : ex-situ measurement
  - Differential interference contrast (DIC) microscopy
  - Surface topography



$GaAs_{0.95}P_{0.05}$
minimal strain so minimal surface features



Textured photocathode, good, there's uniform strain

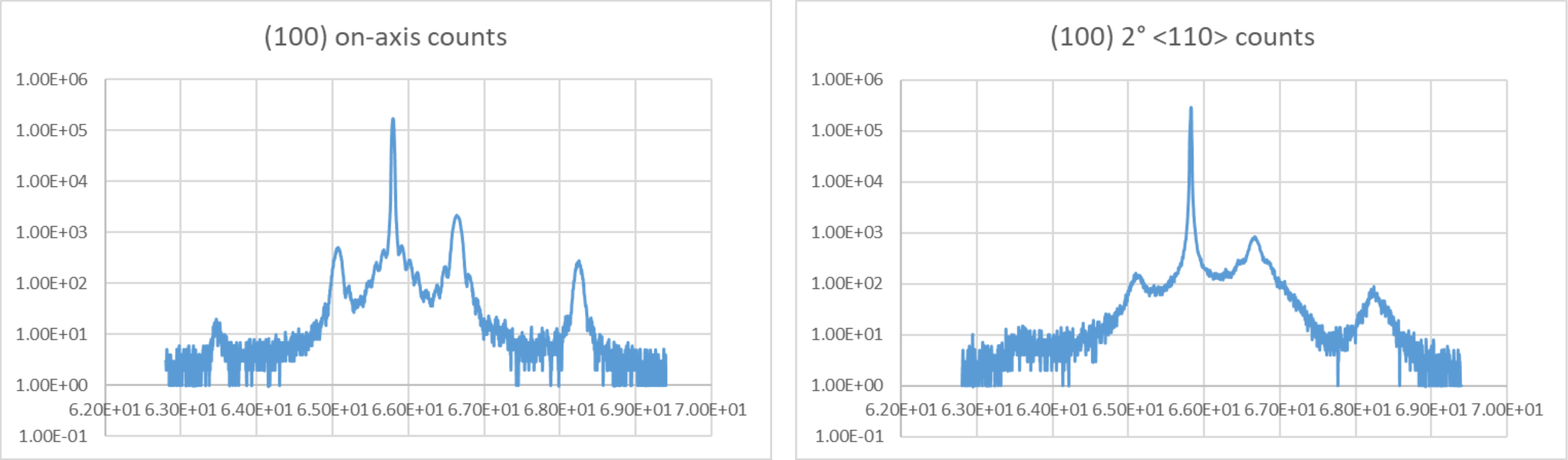


poorly relaxed growth

Tools Used

- X-ray Diffraction : ex-situ measurement
  - Used to measure material strain and composition
  - destructive and constructive interference that occurs when X-rays impinge on sample

GaAs/GaAs$_{0.65}$P$_{0.35}$ superlattice samples grown on different substrates:

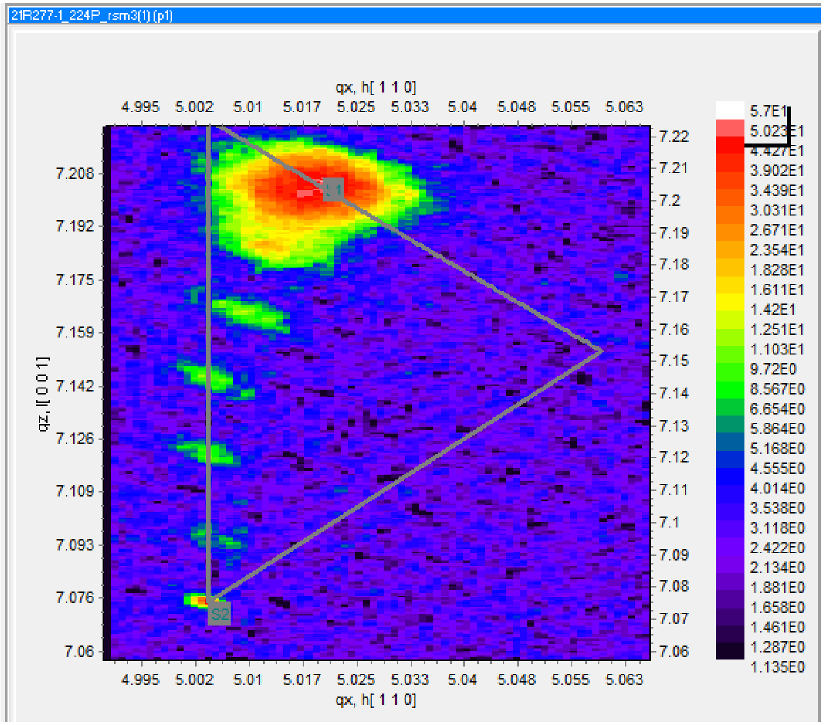


(100) substrate, on-axis

(100) substrate, 2 degree offcut in the (110) direction

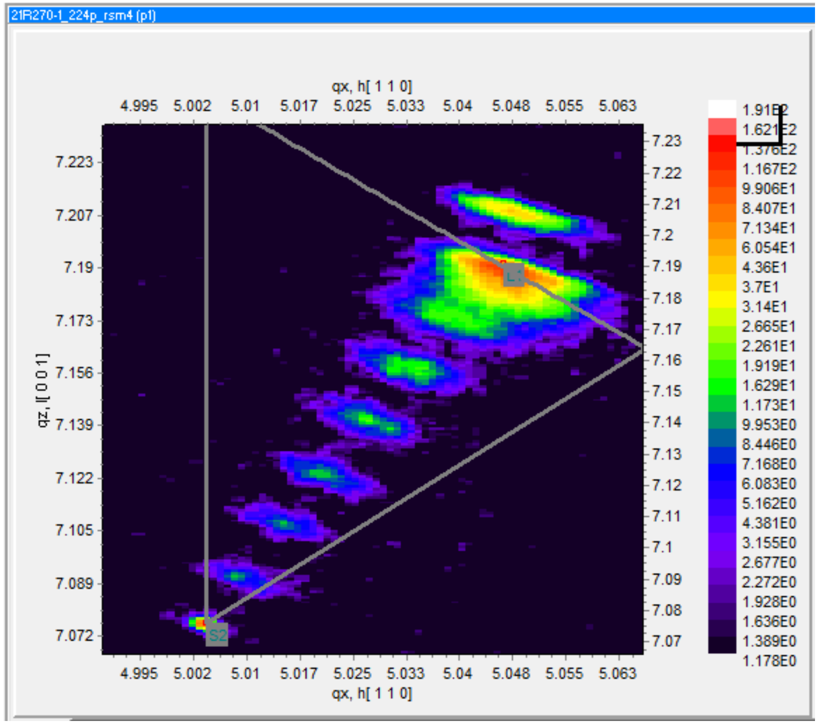
Tools Used

- X-ray Diffraction : ex-situ measurement
  - Reciprocal phase space mapping
  - Shows relaxation of deposited films, key to determining quality of the deposited metamorphic grading



3µm/hr 730°C Growth Temp

Close to vertical line: more strain

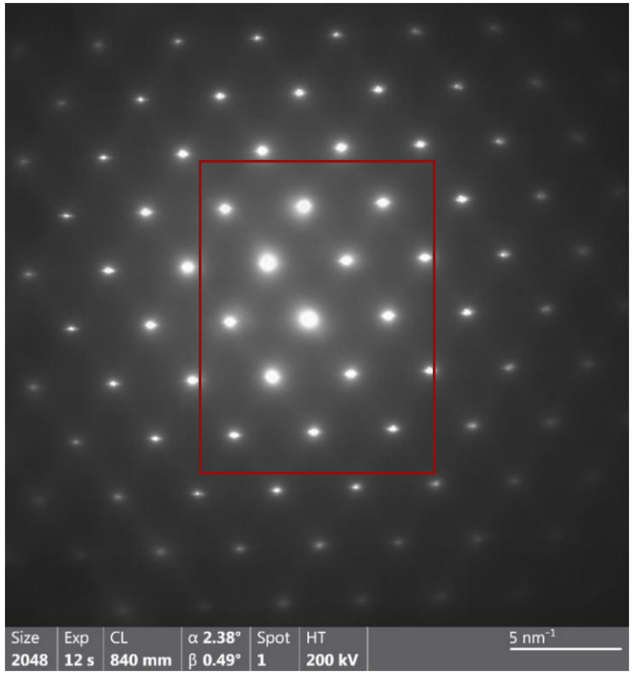


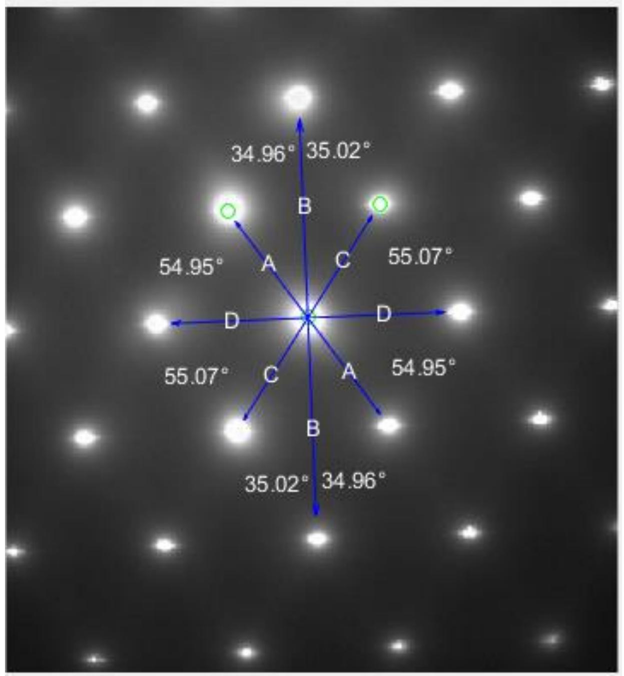
10µm/hr 730°C Growth Temp

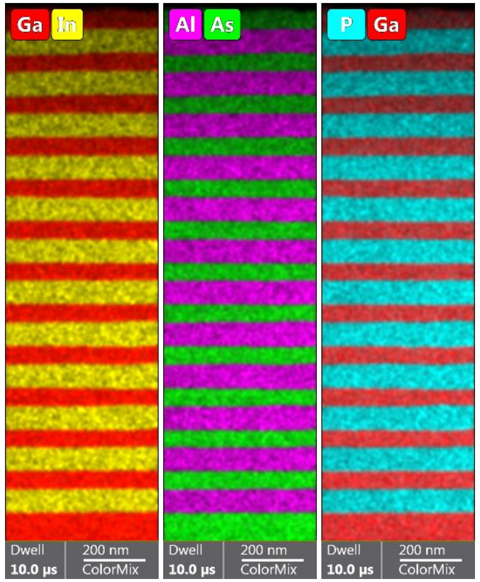
Close to diagonal line: less strain

Tools Used

- Transmission Electron Microscopy : ex-situ measurement
  - TEM micrographs (Images)
  - Selected Area Electron Diffraction (SAED)
  - Energy Dispersive X-Ray Spectroscopy (EDS)

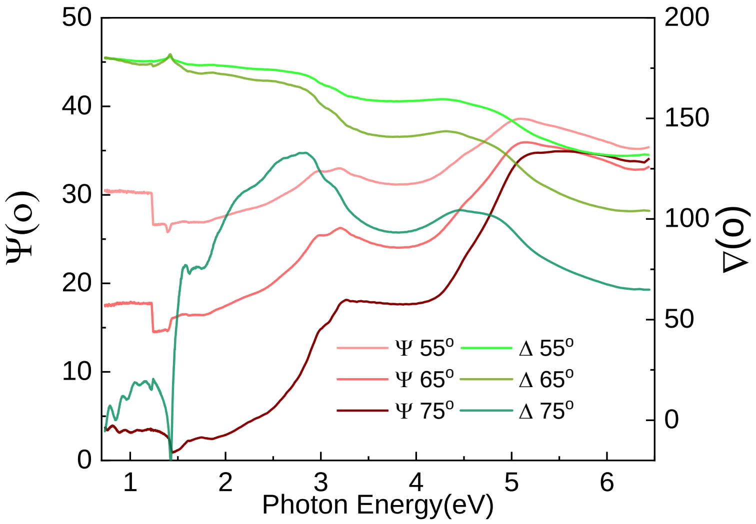


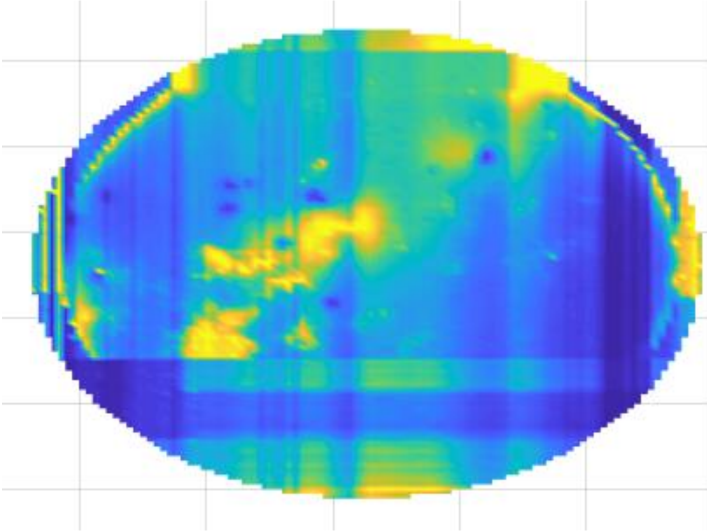


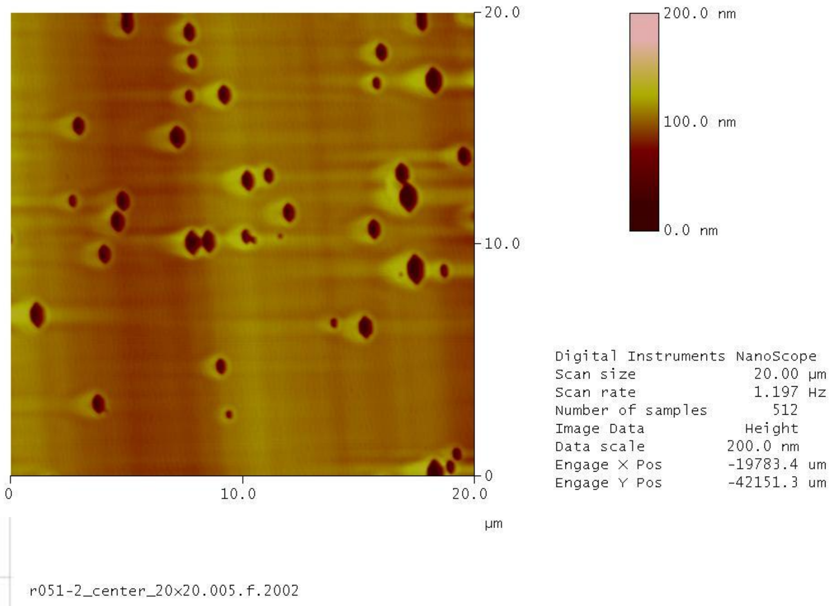


Tools Used

- Ellipsometry (optical method, layer thickness)
- Photoluminescence (PL) mapping (uniformity)
- Hall Effect (measure dopant concentration)
- Atomic Force Microscopy (surface defects)







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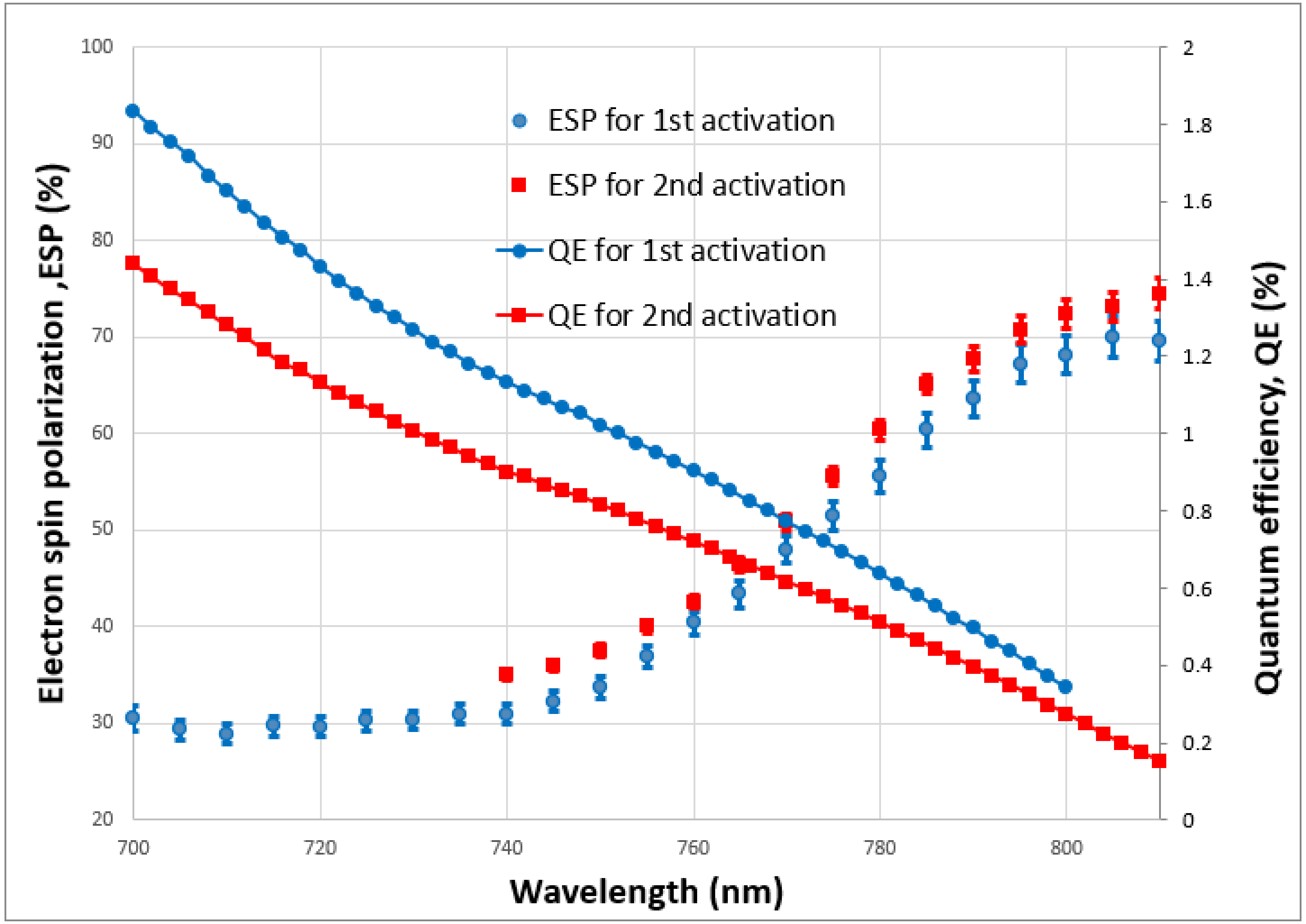
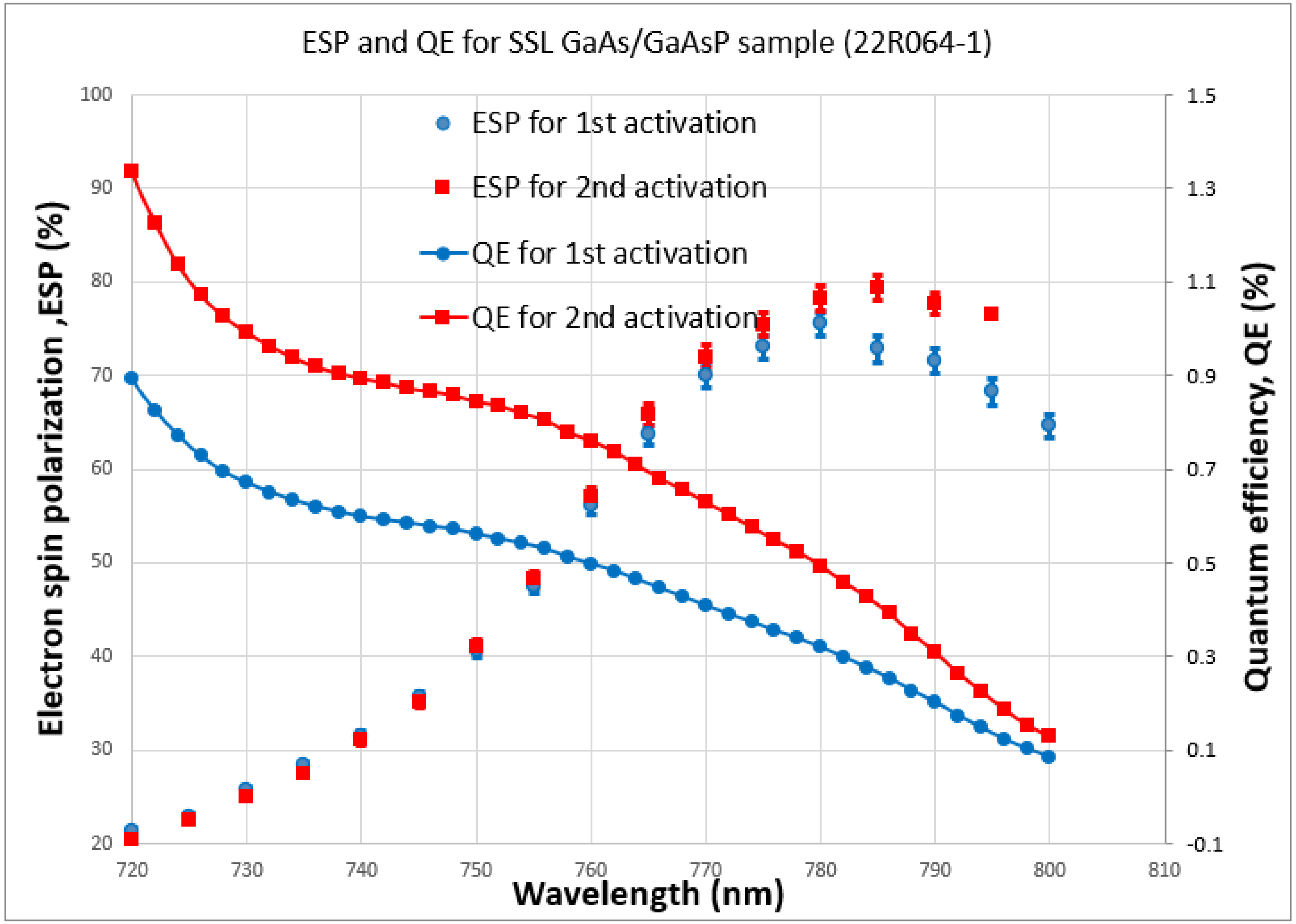
Old Dominion University, Ben Belfore's first set of photocathodes, SSL



Pol ~ 70%
QE ~ 0.3%
λ_{peak} ~ 820 nm

BNL microMott,
measurements by Wei Liu

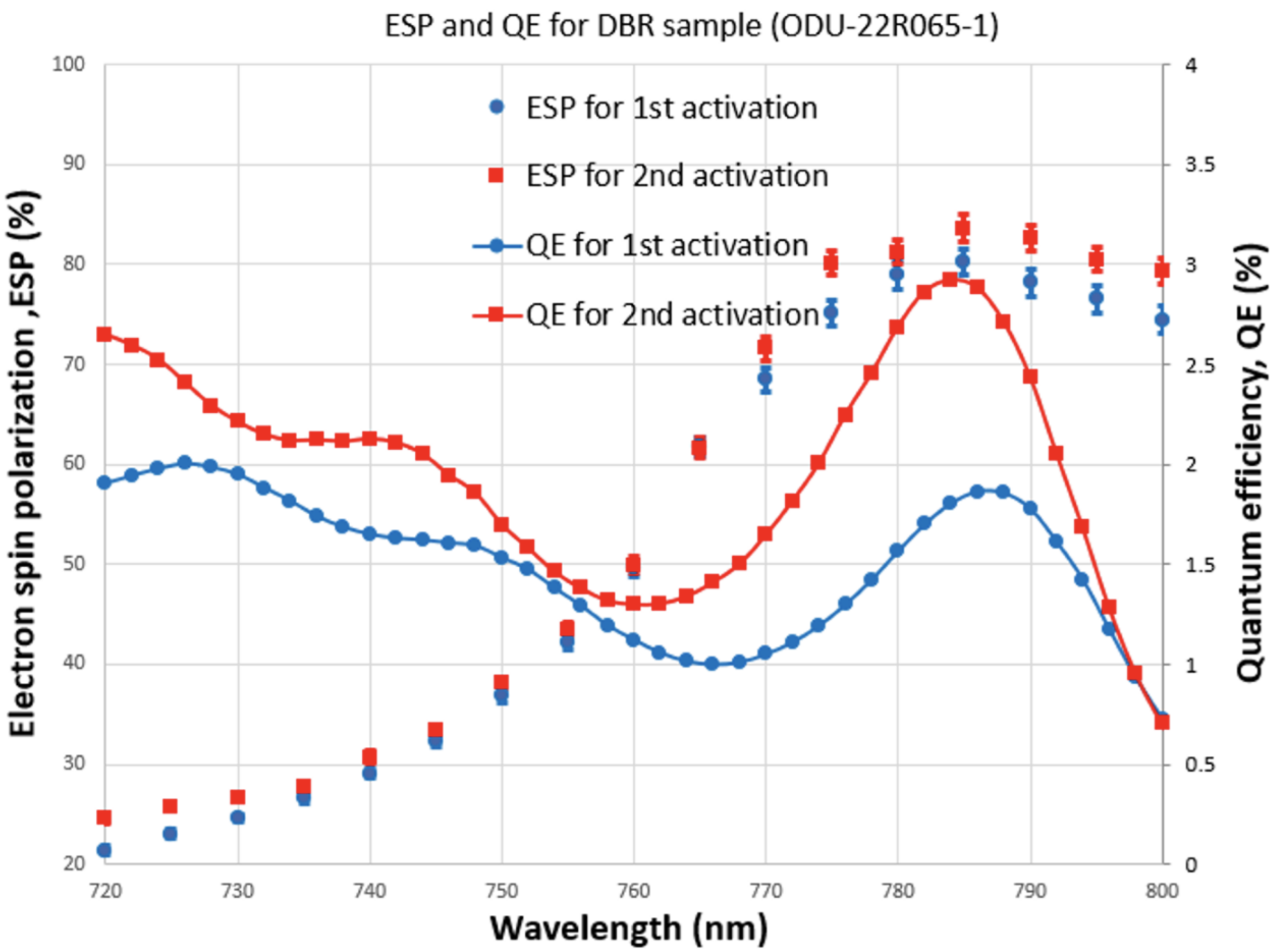
ODU Ben’s second set of photocathodes, SSL



Pol ~ 80%
QE ~ 0.4%
λ_{peak} ~ 785 nm

BNL microMott,
measurements by Wei Liu

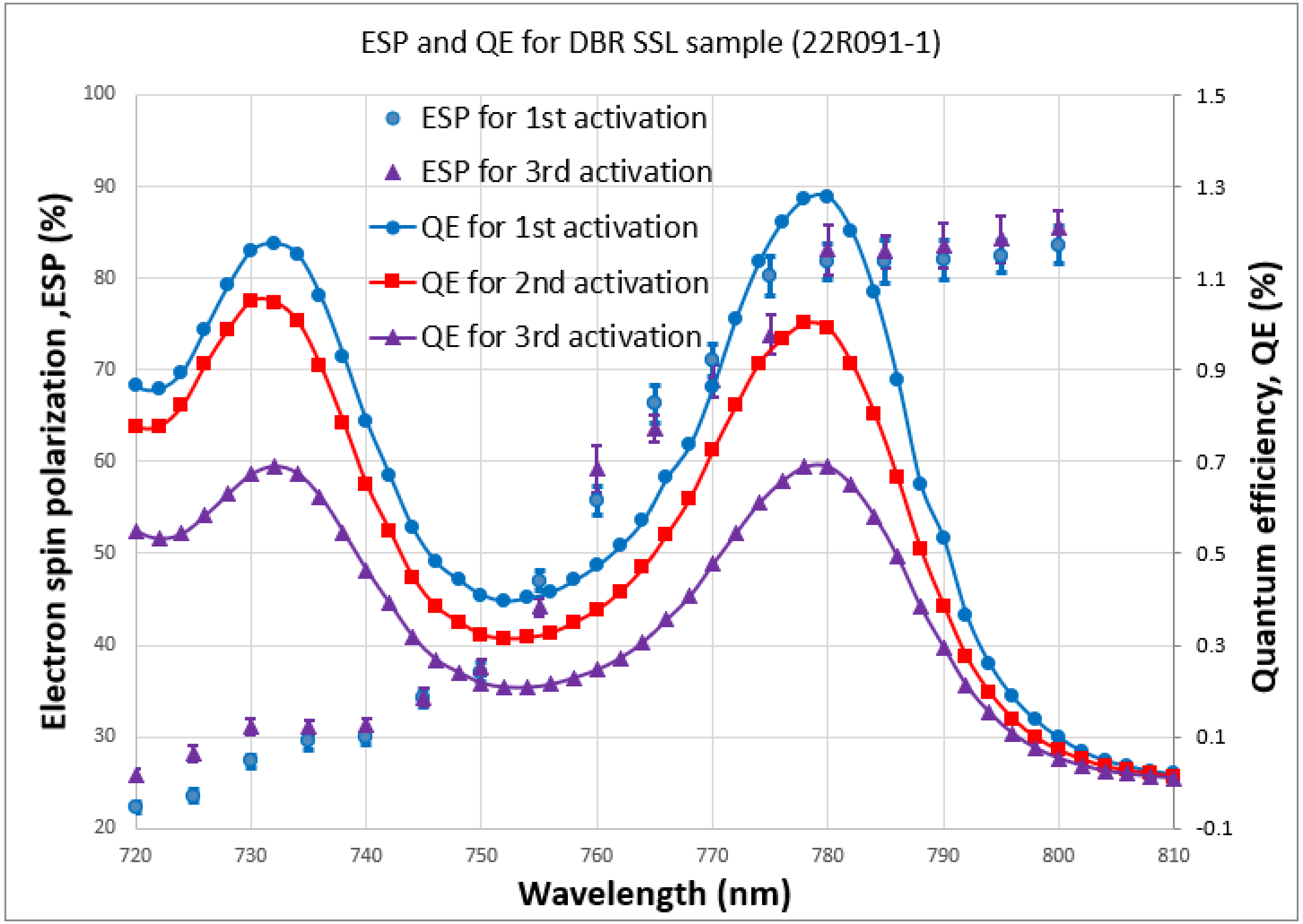
ODU Ben’s third set of photocathodes, SSL with DBR



Pol ~ 83%
QE ~ 3%
λ_{peak} ~ 785 nm

BNL microMott, measurements by Wei Liu

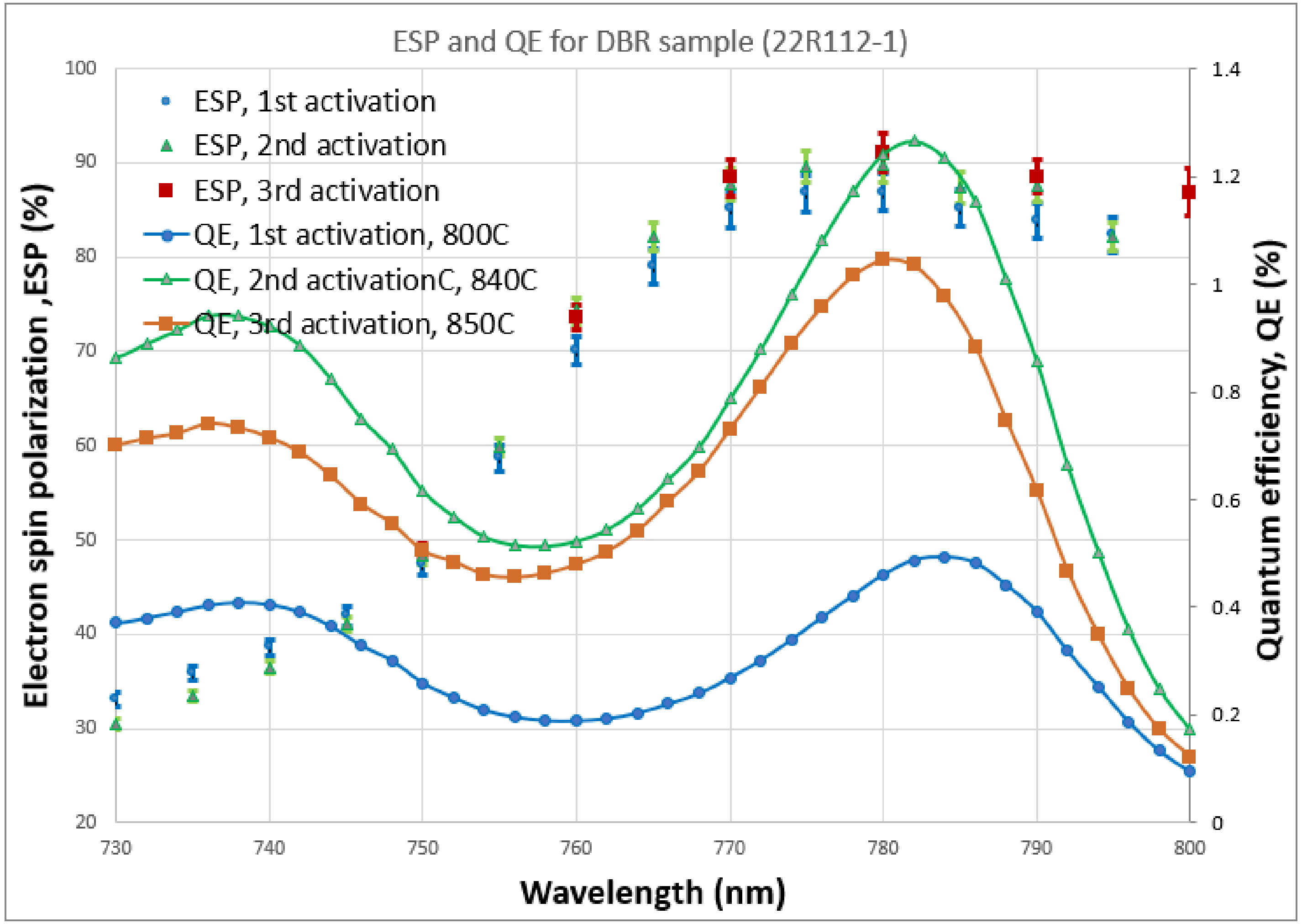
ODU Ben’s fourth set of photocathodes, SSL with DBR



Pol ~ 83%
QE ~ 1.3%
λ_{peak} ~ 775 nm

BNL microMott,
measurements by Wei Liu

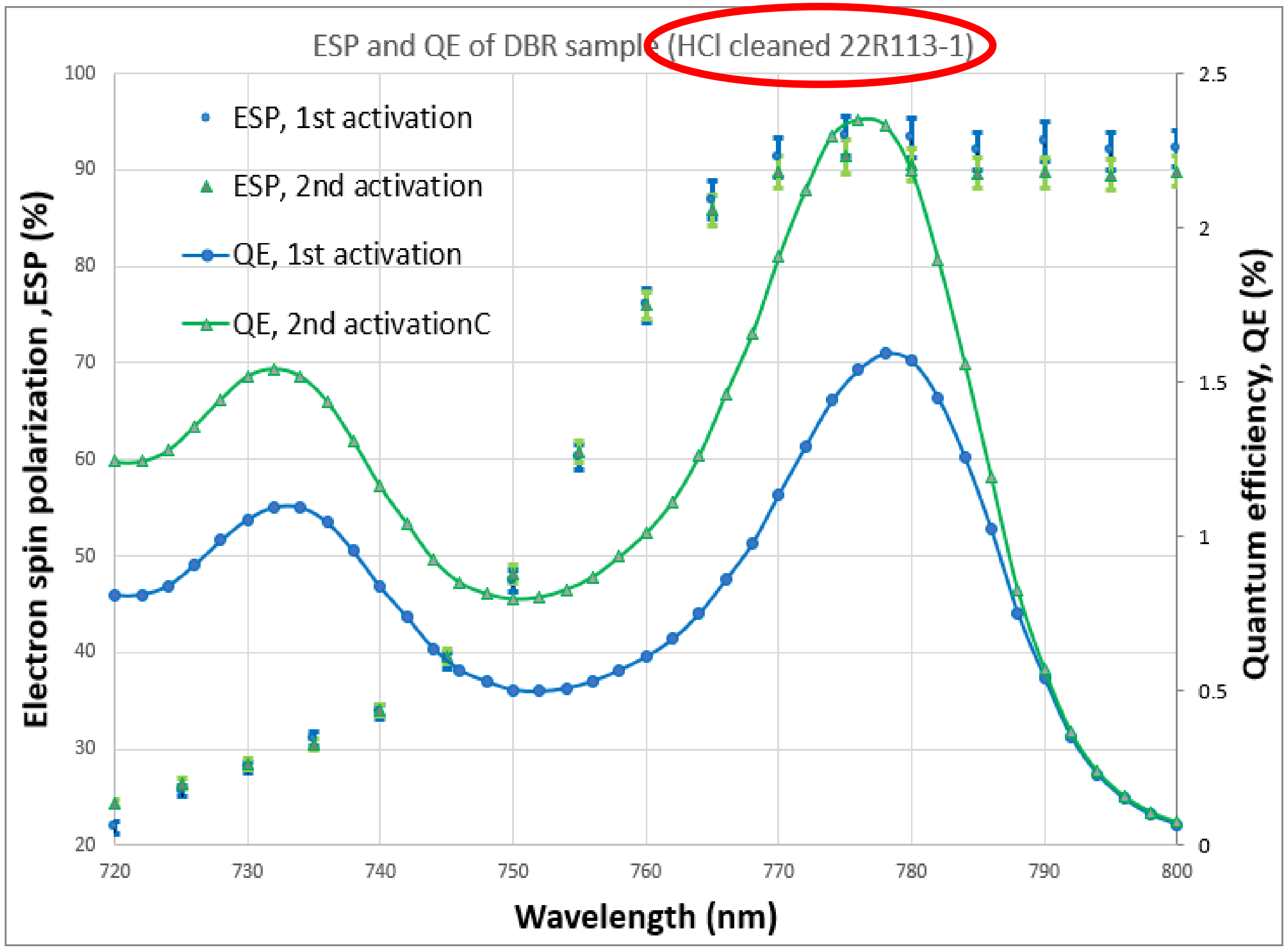
Old Dominion University, Adam Masters' first set of photocathodes, SSL with DBR



Pol ~ 90%
QE ~ 1%
λ_{peak} ~ 785 nm

BNL microMott, measurements by Wei Liu

ODU Adam’s first set of photocathodes, SSL with DBR



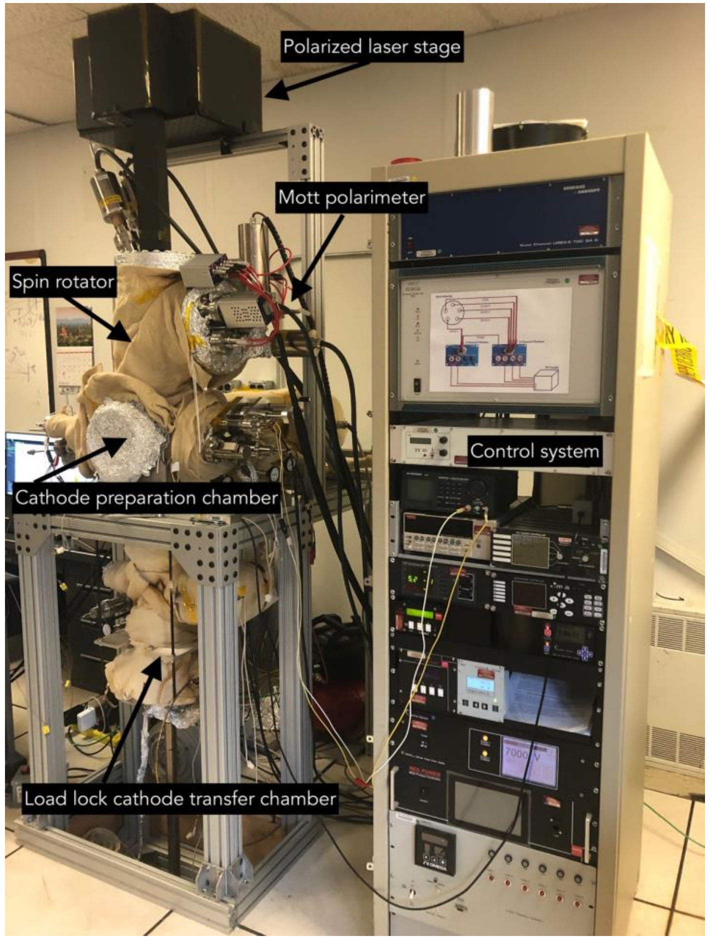
Higher QE following HCl rinse hints at surface contamination

Pol ~ 92%
QE ~ 2.3%
λ_{peak} ~ 785 nm

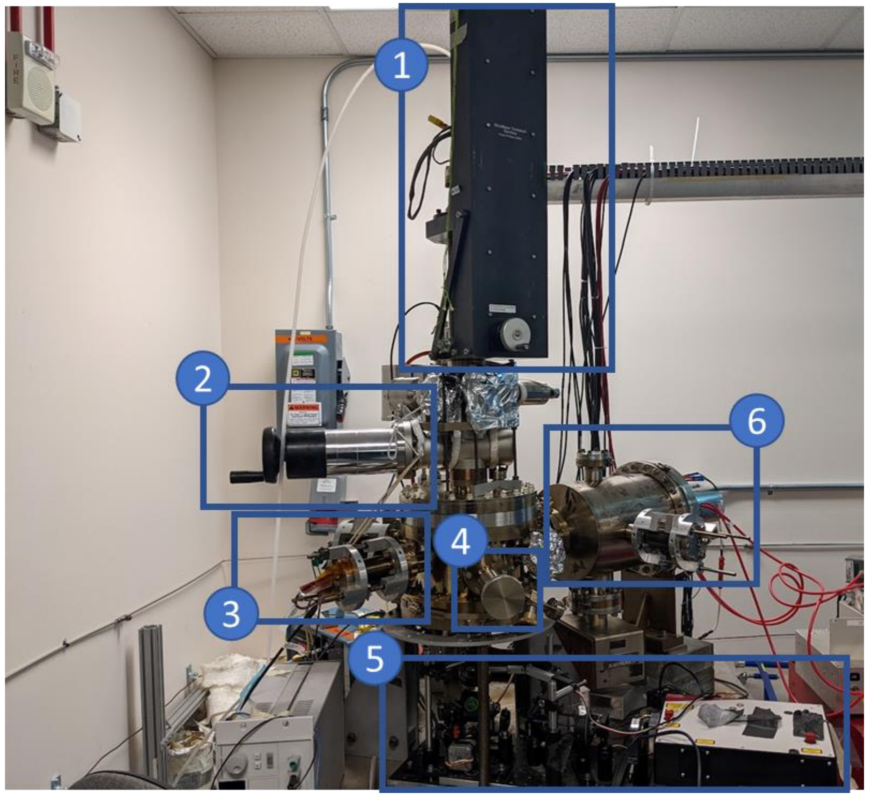
BNL microMott,
measurements by Wei Liu

‘microMott’, ‘retarding field’, low voltage polarimeters

BNL



JLab



Conclusion

- Multiple devices grown - superlattice and superlattice samples with distributed Bragg reflector
- Successful interaction with “service providers”: North Carolina State University and Rochester Institute of Technology
- Problems solved in timely manner: successful doping and compositional calibration of all layers, viable and reliable growth recipes identified
- MOCVD is definitively a reliable method for fabricating high polarization photocathodes with good QE
- Future Work: arsenic cap layer, hydrogen cleaning, higher QE (higher polarization too, why not?)

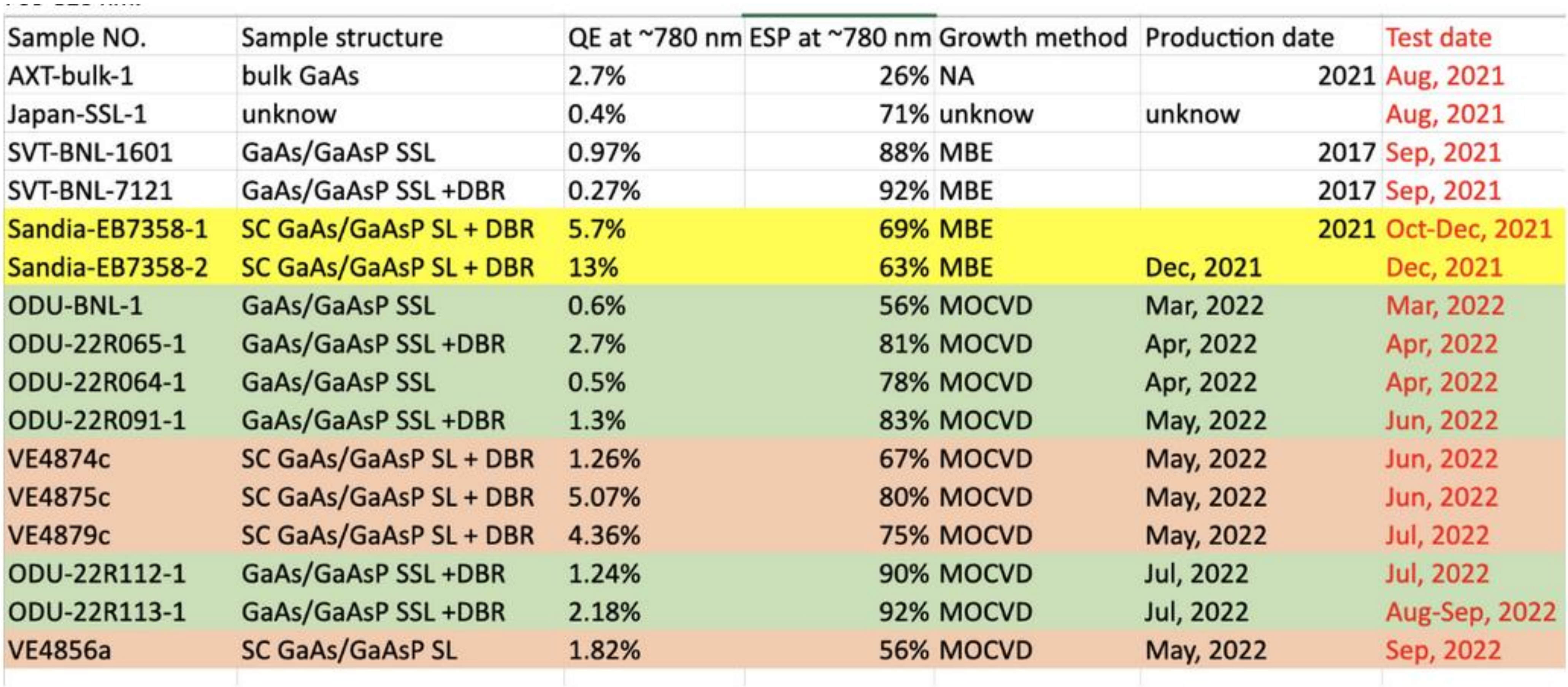
Project Conclusion

Stated clearly:

- We successfully fabricated both kinds of photocathodes, SSL and SSL with DBR, via MOCVD. Exceptional work by ODU
- We achieved our central goal: Polarization > 90% and QE > 1%
- Between JLab and BNL, we have nearly 200k$ worth of photocathode material, our photocathode stores now replenished! Enough material for polarimeter cross checks, to support CEBAF and EIC physics programs, and to support polarized source R&D
- BNL has a fully functional microMott polarimeter, and this device was essential for project success
- JLab's microMott polarimeter is very close to being operational again
- The cross calibration of the two microMott polarimeters will happen
- Having both source groups – JLab and BNL – participating in this project has been wonderful, a big selling point (IMHO)

Back up slides

Measurements made at BNL



Sample NO.	Sample structure	QE at ~780 nm	ESP at ~780 nm	Growth method	Production date	Test date
AXT-bulk-1	bulk GaAs	2.7%	26%	NA	2021	Aug, 2021
Japan-SSL-1	unknow	0.4%	71%	unknow	unknow	Aug, 2021
SVT-BNL-1601	GaAs/GaAsP SSL	0.97%	88%	MBE	2017	Sep, 2021
SVT-BNL-7121	GaAs/GaAsP SSL +DBR	0.27%	92%	MBE	2017	Sep, 2021
Sandia-EB7358-1	SC GaAs/GaAsP SL + DBR	5.7%	69%	MBE	2021	Oct-Dec, 2021
Sandia-EB7358-2	SC GaAs/GaAsP SL + DBR	13%	63%	MBE	Dec, 2021	Dec, 2021
ODU-BNL-1	GaAs/GaAsP SSL	0.6%	56%	MOCVD	Mar, 2022	Mar, 2022
ODU-22R065-1	GaAs/GaAsP SSL +DBR	2.7%	81%	MOCVD	Apr, 2022	Apr, 2022
ODU-22R064-1	GaAs/GaAsP SSL	0.5%	78%	MOCVD	Apr, 2022	Apr, 2022
ODU-22R091-1	GaAs/GaAsP SSL +DBR	1.3%	83%	MOCVD	May, 2022	Jun, 2022
VE4874c	SC GaAs/GaAsP SL + DBR	1.26%	67%	MOCVD	May, 2022	Jun, 2022
VE4875c	SC GaAs/GaAsP SL + DBR	5.07%	80%	MOCVD	May, 2022	Jun, 2022
VE4879c	SC GaAs/GaAsP SL + DBR	4.36%	75%	MOCVD	May, 2022	Jul, 2022
ODU-22R112-1	GaAs/GaAsP SSL +DBR	1.24%	90%	MOCVD	Jul, 2022	Jul, 2022
ODU-22R113-1	GaAs/GaAsP SSL +DBR	2.18%	92%	MOCVD	Jul, 2022	Aug-Sep, 2022
VE4856a	SC GaAs/GaAsP SL	1.82%	56%	MOCVD	May, 2022	Sep, 2022

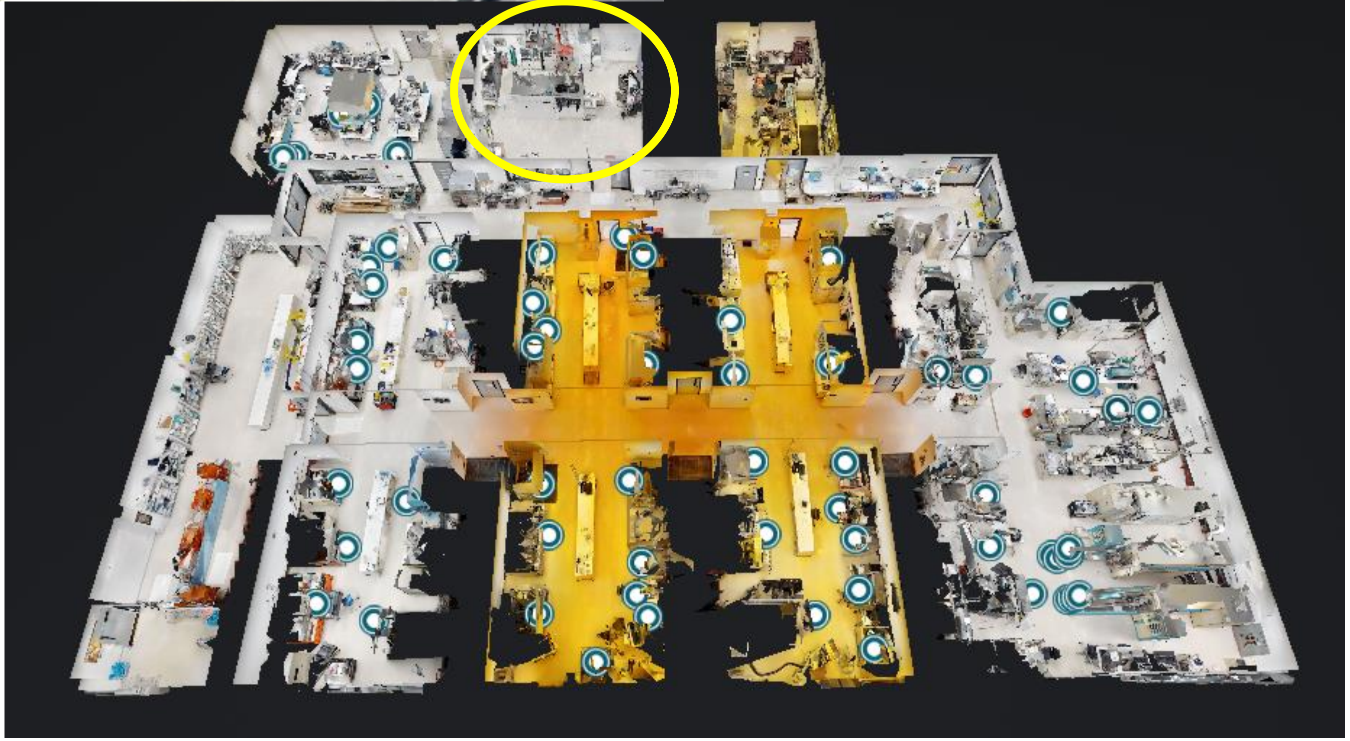
MOCVD: Photocathode progress

Virginia Center for Photovoltaics

The Rochester Institute
of Technology
III-V EPICenter

Dr. Sylvain Marsillac,
Old Dominion University

Ben Belfore
ODU Graduate
Student

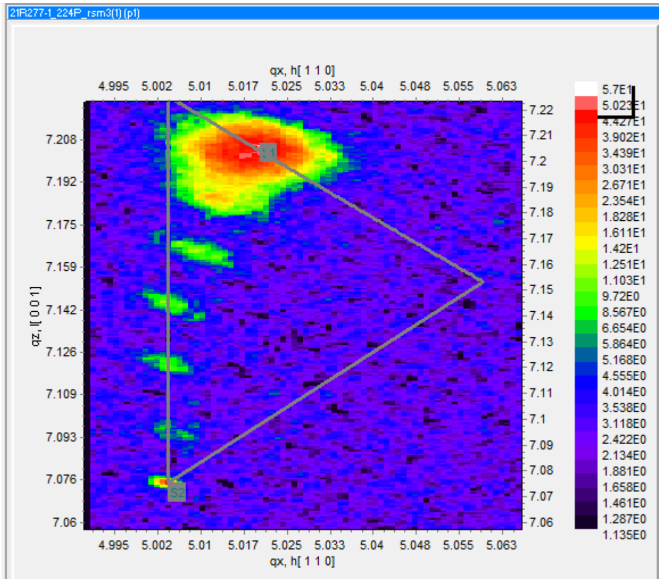


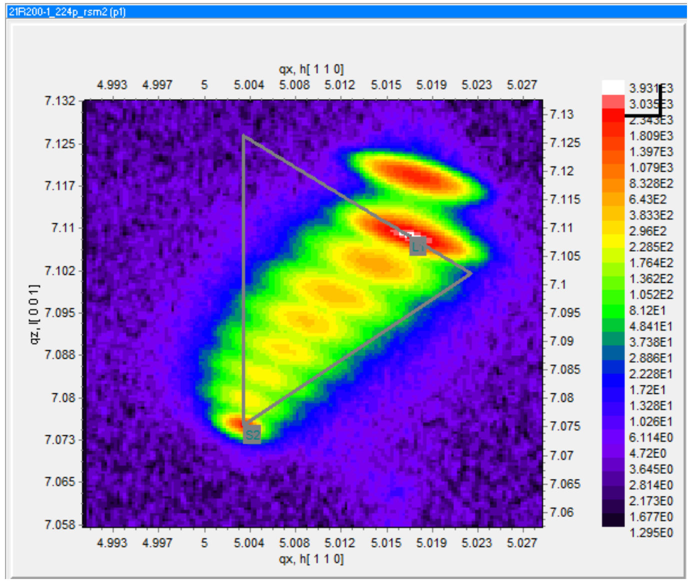
Results: Metamorphic Grading

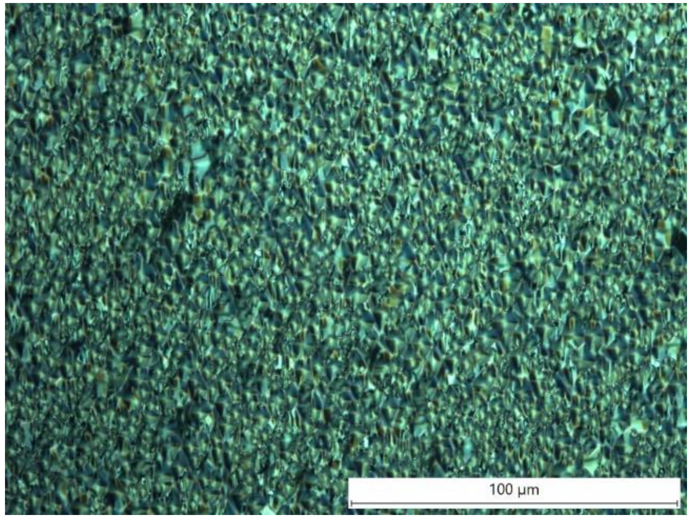
- High-quality strain relaxation of underlying layers is key to getting intended GaAs strain in the emitting region
- Necessary because of growing on lattice mismatched substrate
- RSM used to characterize extent of relaxation in metamorphic layers
- Key parameters changed:
  - Growth Rate
  - Growth Temperature
  - Arsine/Phosphine Ratio

Results: Metamorphic Grading

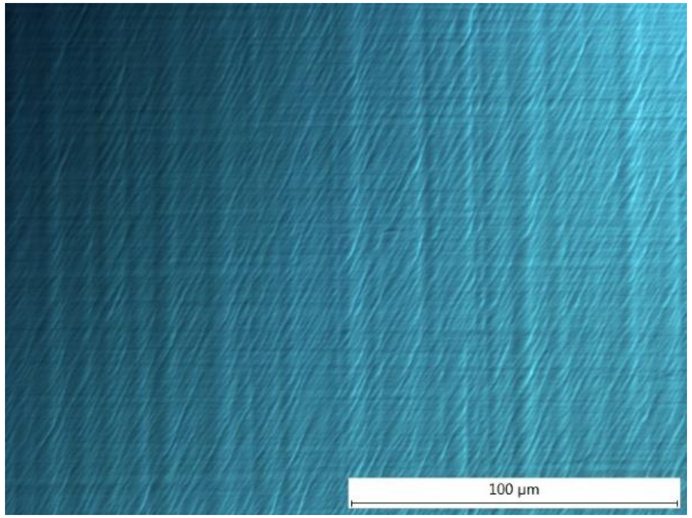
- Slower Growth Rate resulted in highest strained films (left)
- High Temperature with smaller composition change between steps is more promising (right)





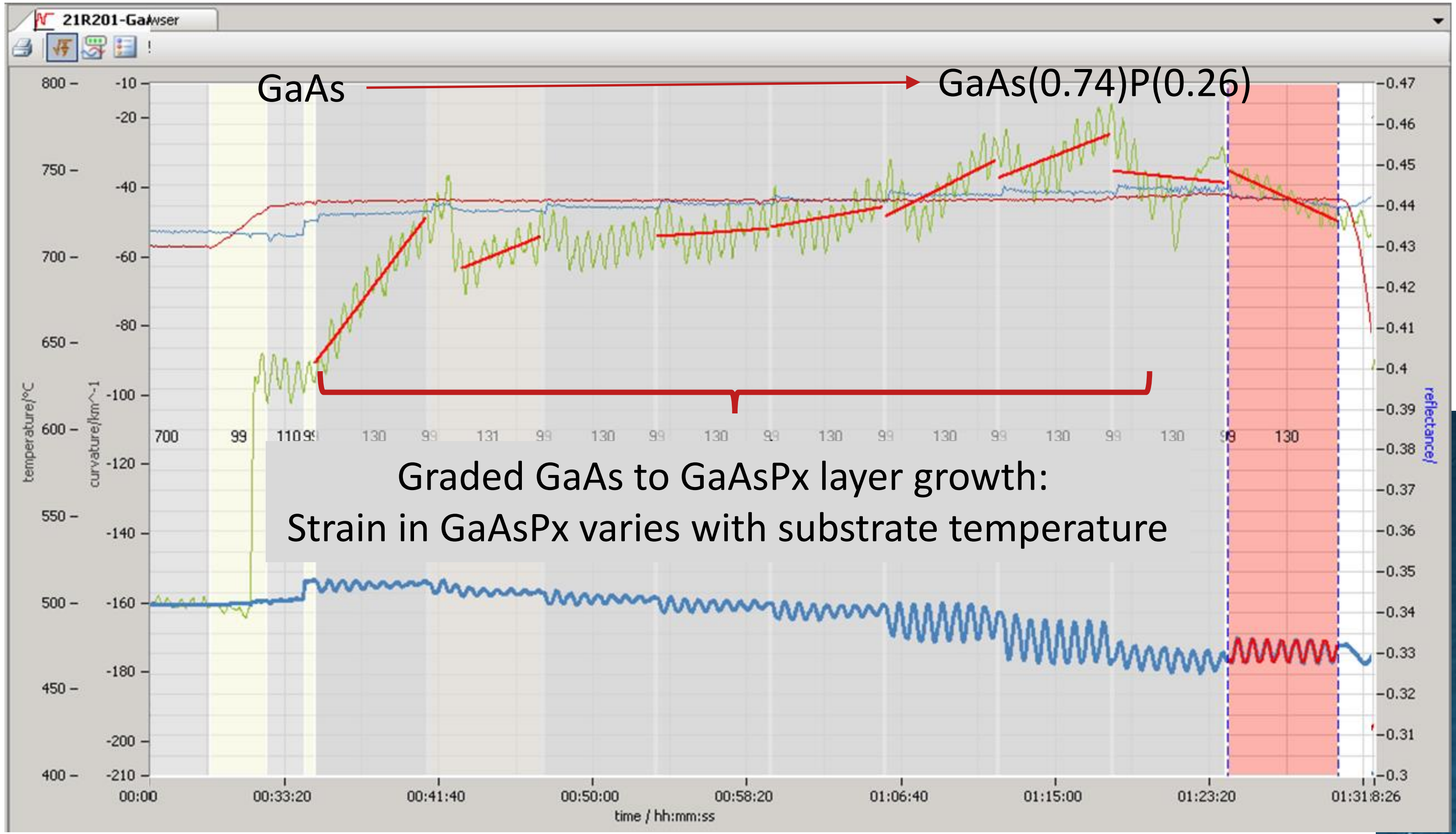


3µm/hr 730°C Growth Temp



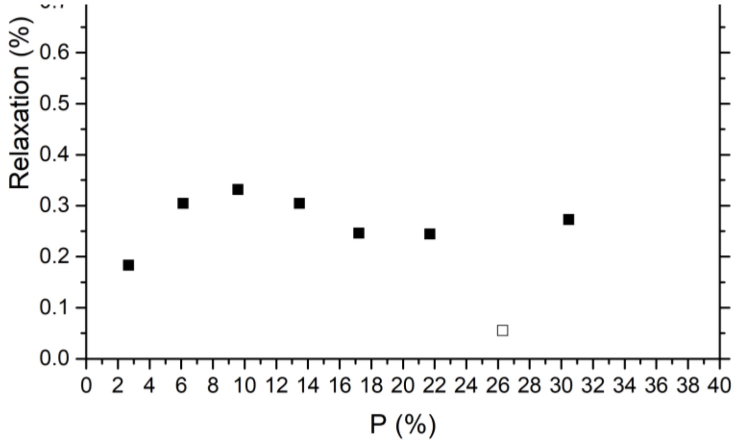
10µm/hr 650°C lower phosphorous grade

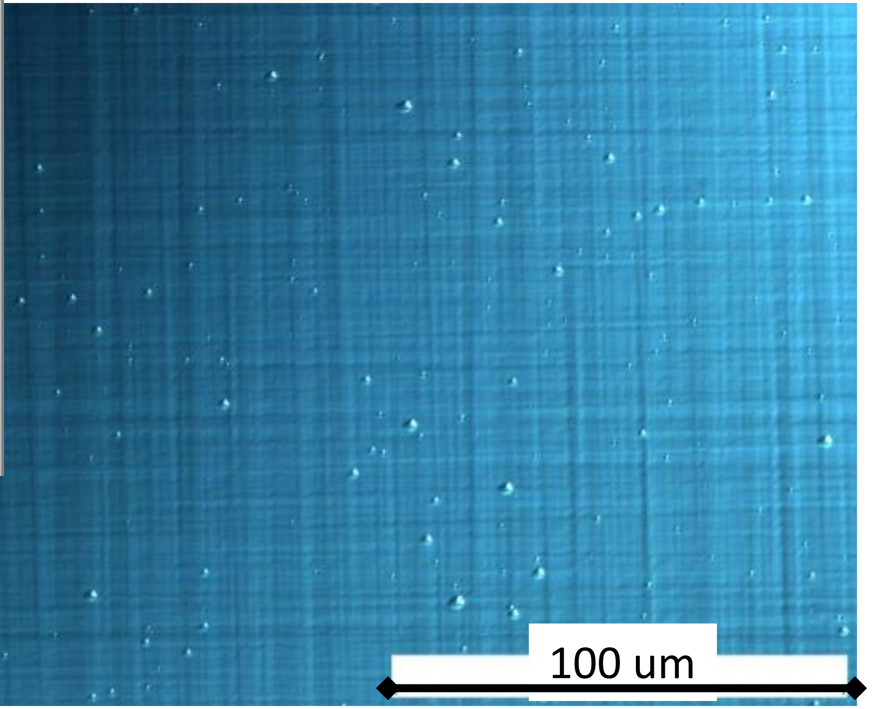
Results: MOCVD



GaAs → GaAs(0.74)P(0.26)

Graded GaAs to GaAsPx layer growth:
Strain in GaAsPx varies with substrate temperature





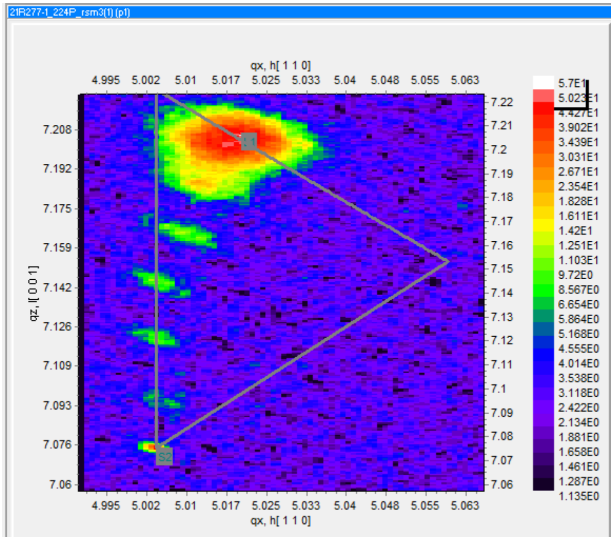
Higher temperatures yielded improved surface with moderate relaxation throughout

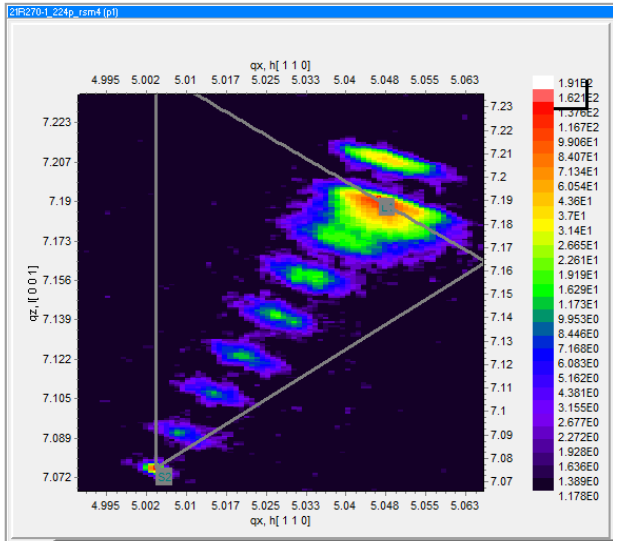
730°C growth temperature

Optimizing temperatures, graded layer profile

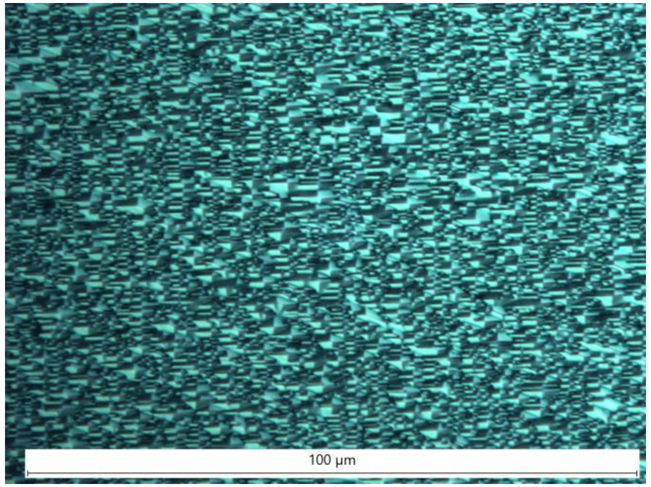
Results: Device Continued

- Surface quality initially poor but subsequent runs improved surface quality
- Refining runs to improve relaxation

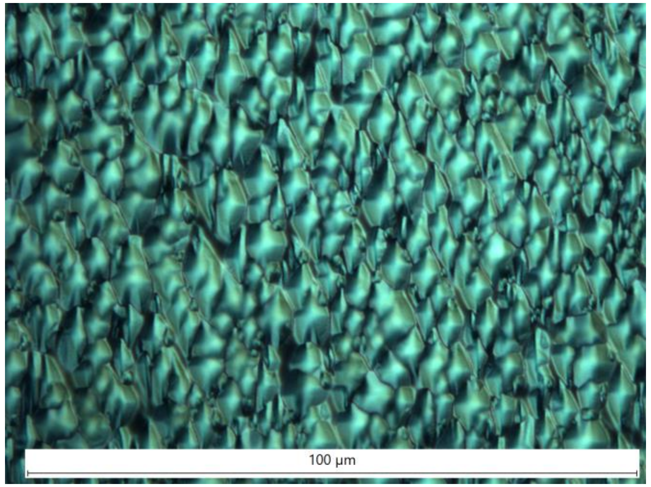




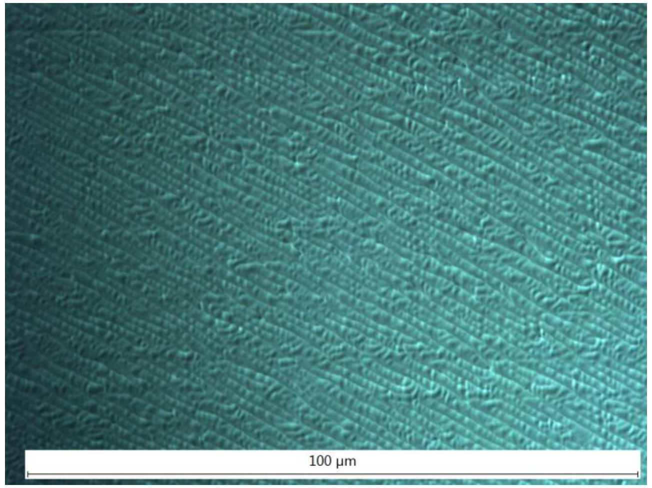
Surface completely matted,
RSM not performed



10µm/hr 650°C Growth Temp

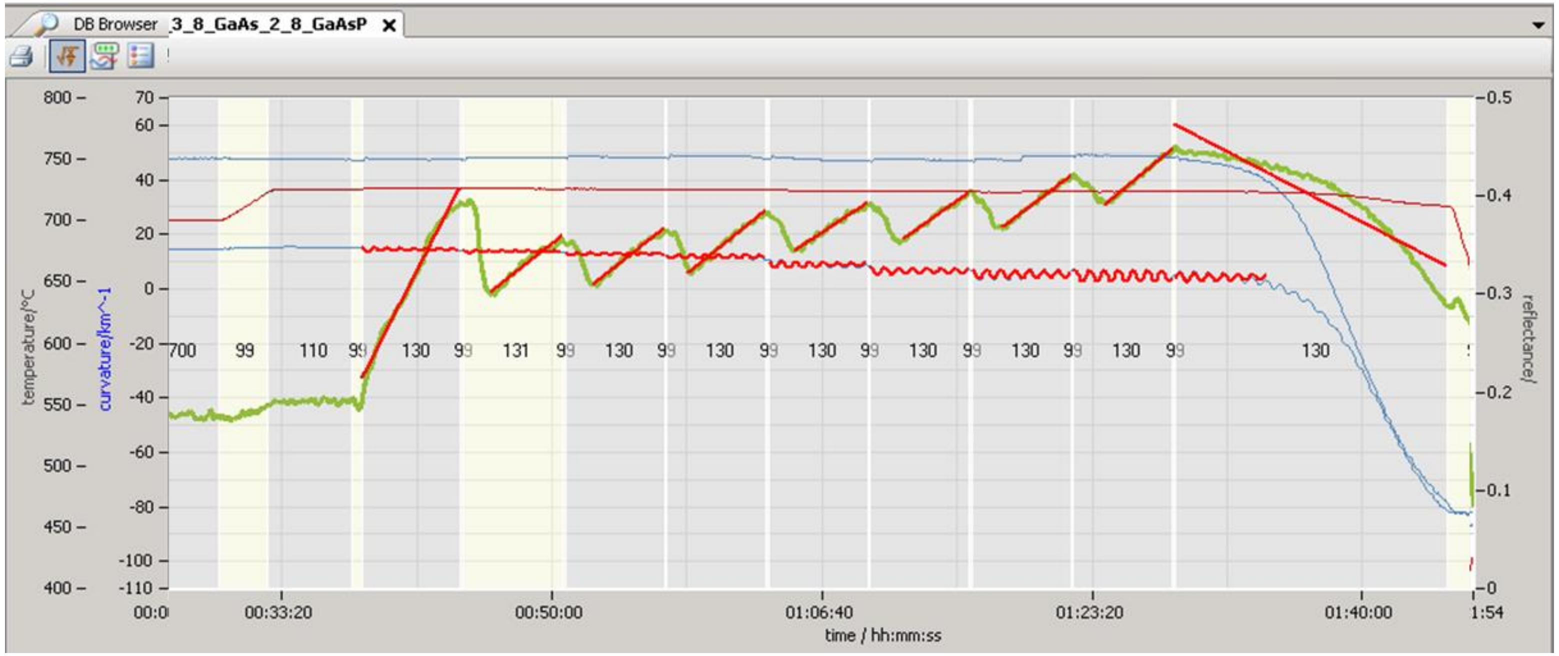


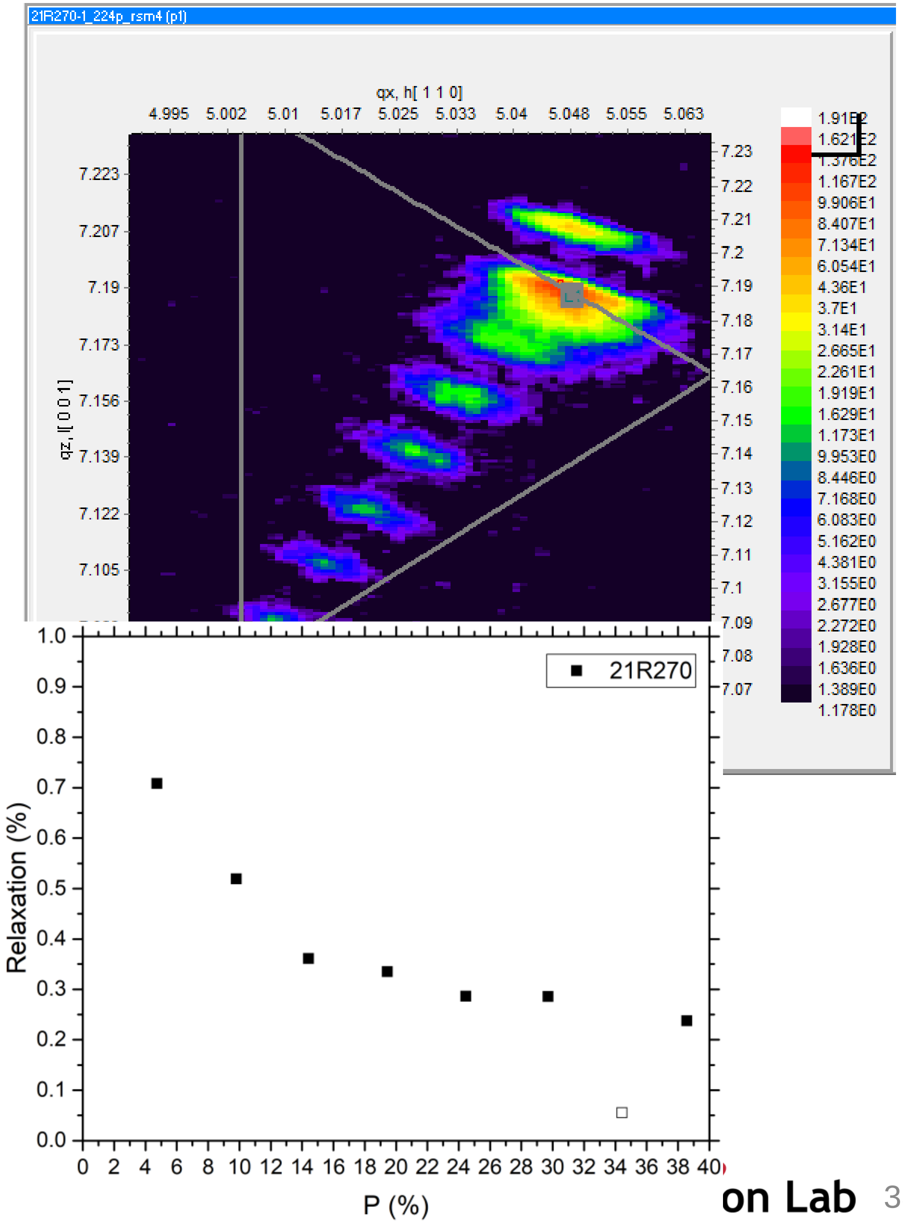
3µm/hr 730°C Growth Temp



10µm/hr 730°C Growth Temp
PL Map taken on this sample

MOCVD monitoring: graded layer optimization





(100) 2° <110>

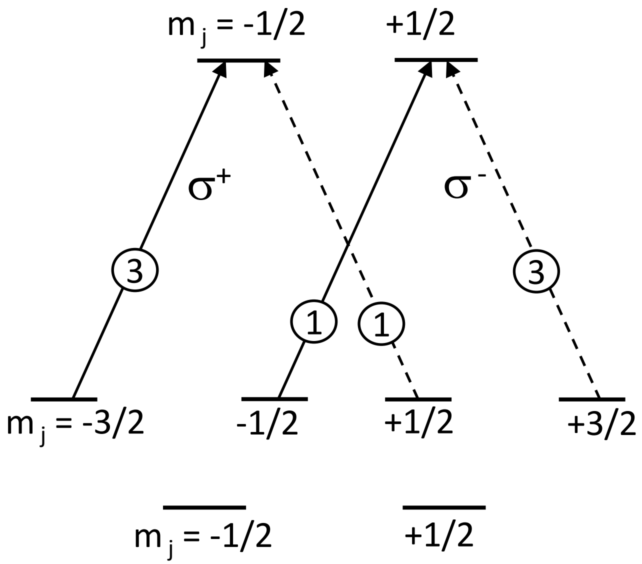
Average ΔP = 4.95%

Higher temperature and higher ΔP steps begins well, levels out to 30% relaxation, and surface degrades during thick 35% buffer

~10 µm/hr graded layer growth rate
V/III: 20-25
730°C growth temperature

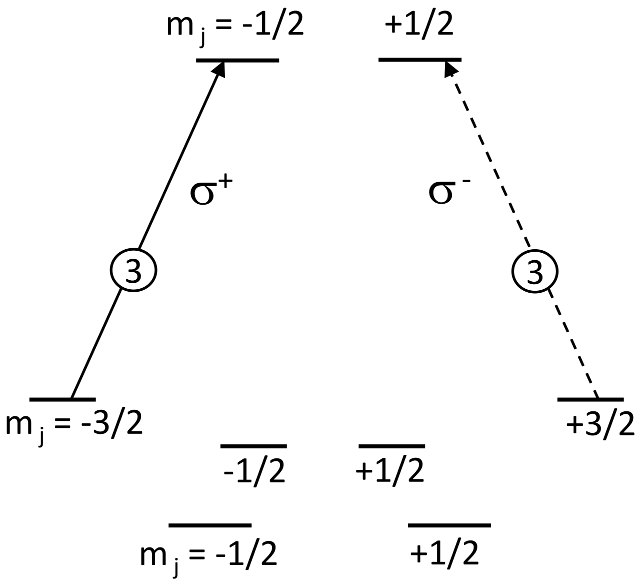
Making polarized electron beams with GaAs

Bulk GaAs – no strain



Maximum Polarization 50%

Strained GaAs



Maximum Polarization 100%