

Strained Superlattice photocathodes with CBE

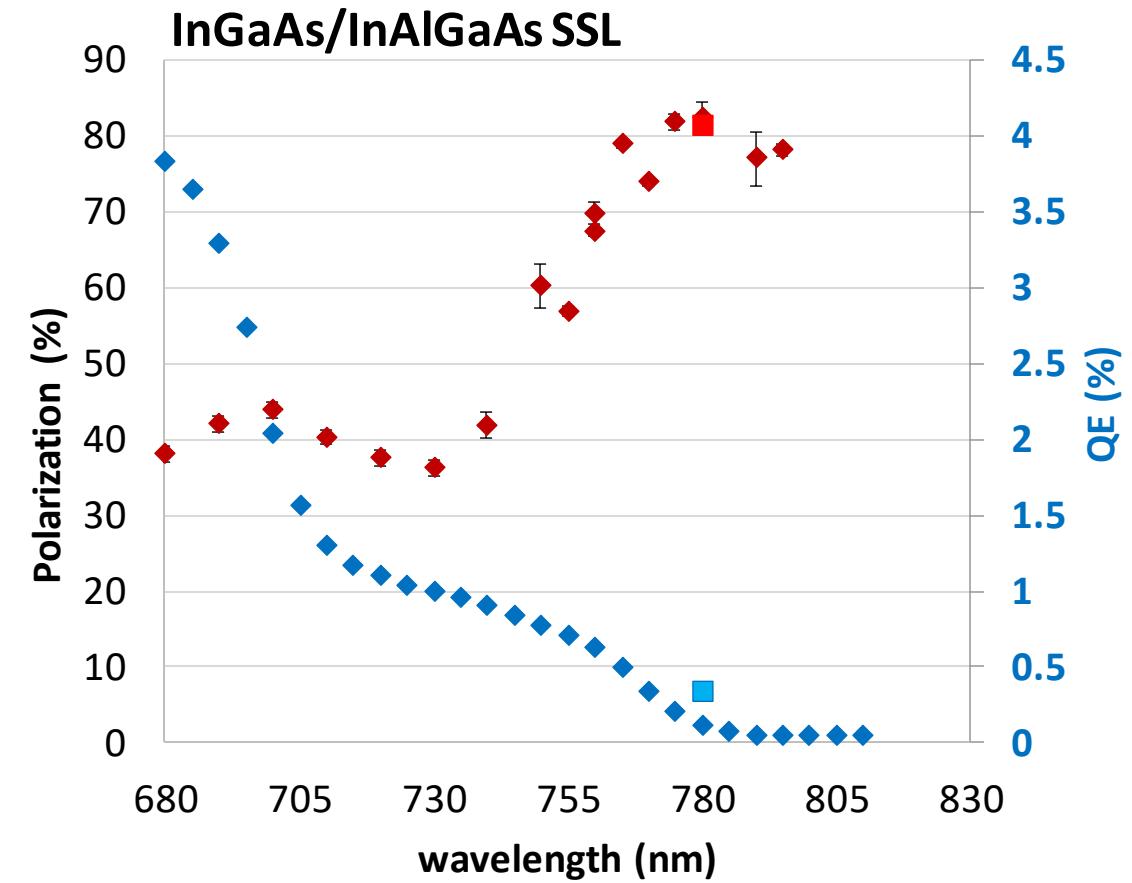
SPIN 2023

25th International Symposium on
Spin Physics

Marcy Stutzman, Jefferson Lab

Chris Palmstrøm and Aaron Engel, UCSB

Greg Blume, Old Dominion University



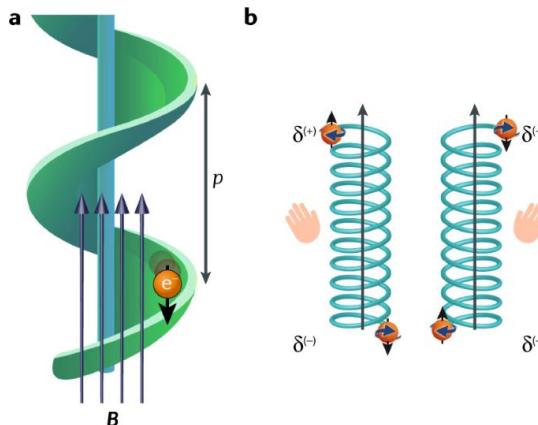
Why we want polarized electros



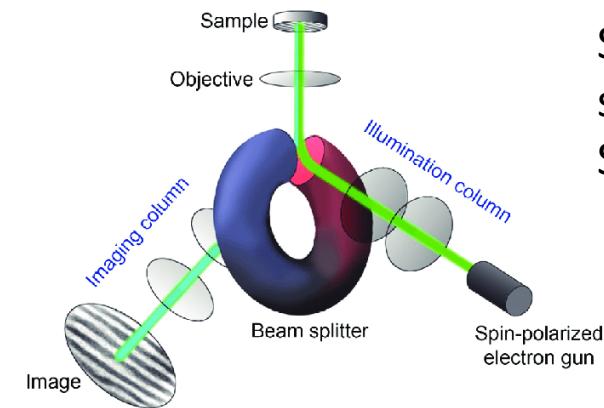
25TH INTERNATIONAL SPIN PHYSICS SYMPOSIUM

- Nuclear and High Energy physics experiments
- Atomic physics
- Surface analysis: Spin polarized LEEM, TEM, ...
- Spintronics

Chiral Molecules

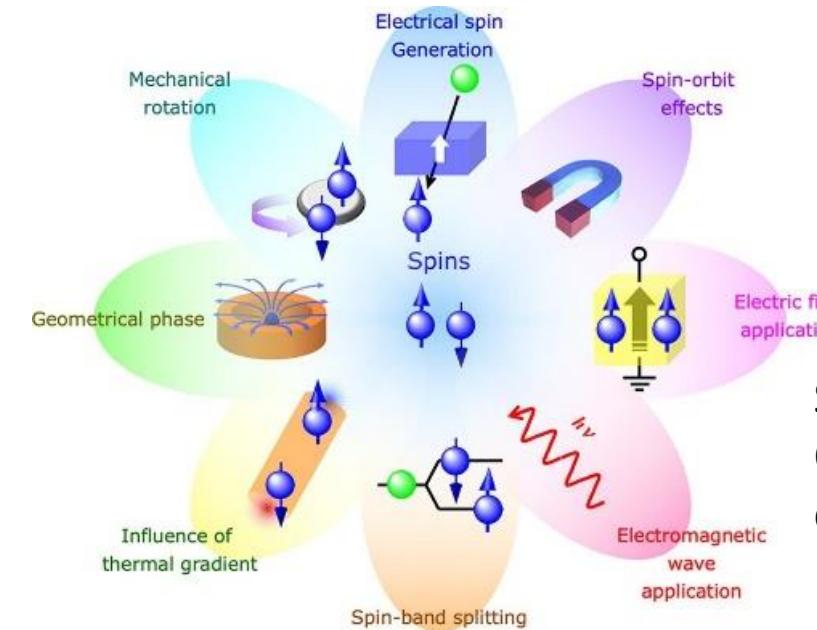


Naaman, R., Paltiel, Y. & Waldeck, D.H., *Nat Rev Chem* **3**, 250–260 (2019).
<https://doi.org/10.1038/s41570-019-0087-1>



Spin resolved surface analysis:
SPLEEM

Rougemaille, N. & Schmid, A., (2010)
The European Physical Journal Applied Physics. 50. 10.1051/epjap/2010048.



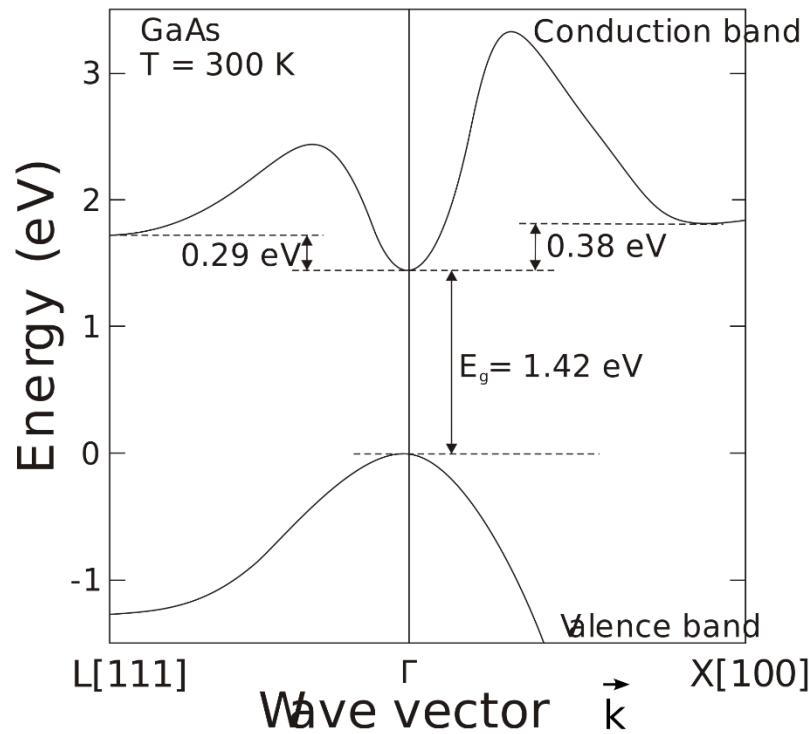
Spintronics,
Quantum
computing

A. Hirohata et al., *Journal of Magnetism and Magnetic Materials*, **509**, (2020), 166711.
<https://doi.org/10.1016/j.jmmm.2020.166711>.

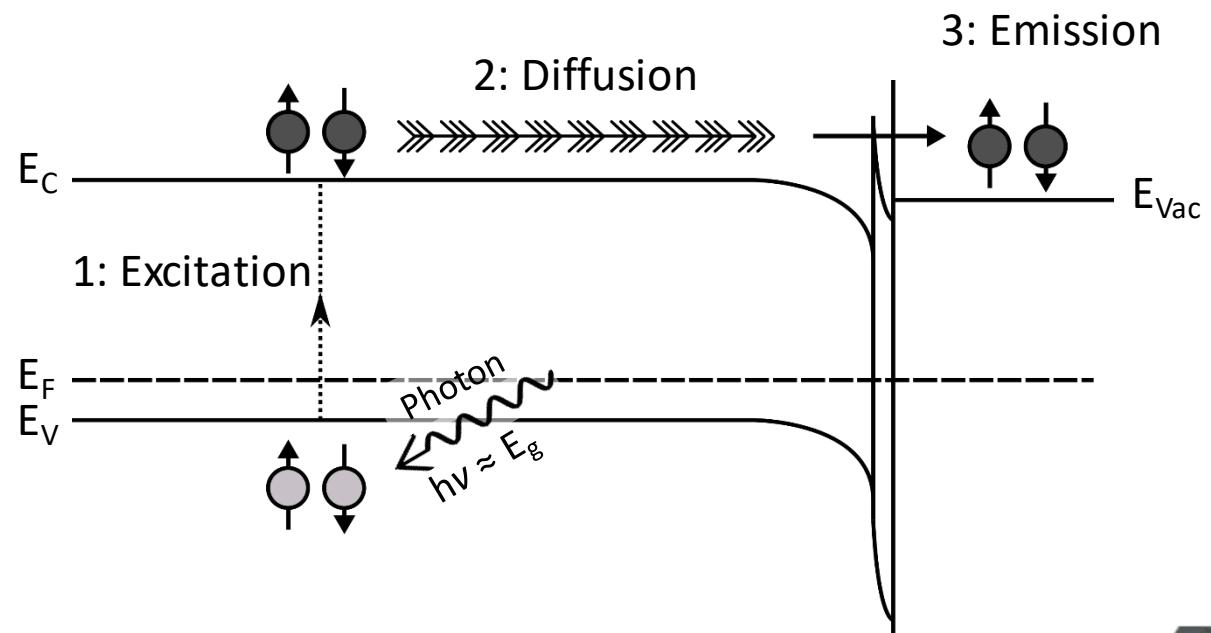
Photoemission from GaAs

Direct Band Gap

- Photoexcitation promotes carriers to conduction band



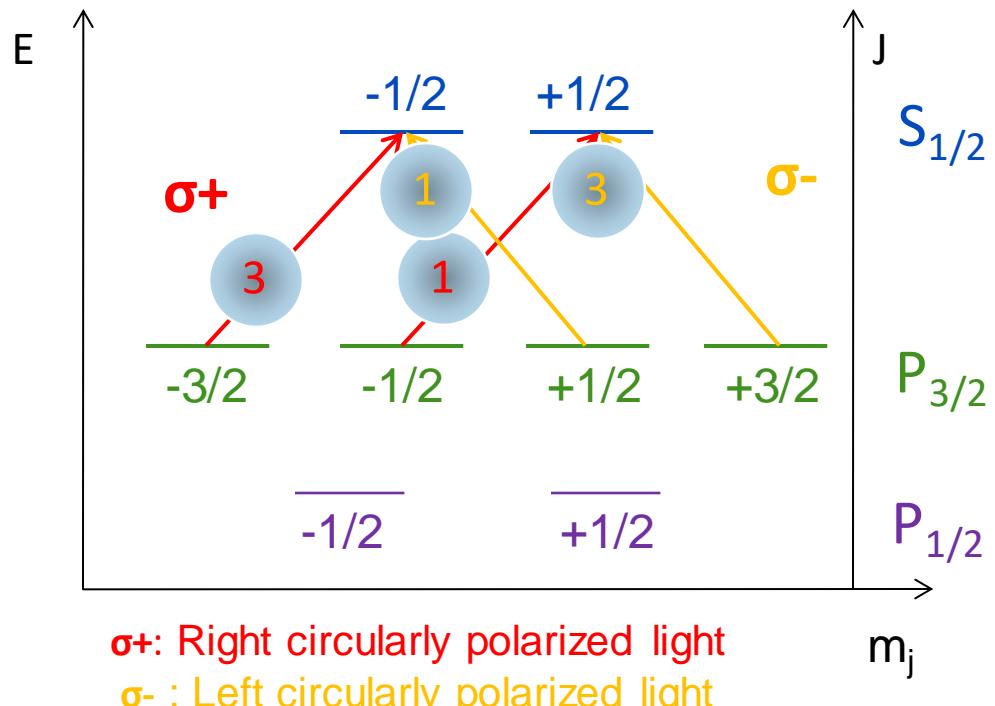
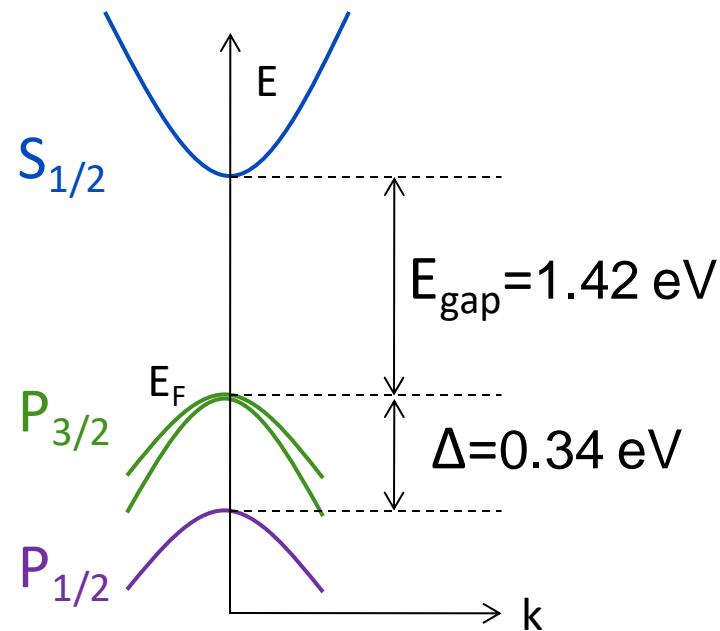
Three step photoemission from GaAs



W.E. Spicer, *Phys Rev* **112**, 114 (1958)
C.N. Berglund and W.E. Spicer, *Phys Rev* **4A**, A1030 (1964)



Spin Polarized Photoemission from Bulk GaAs

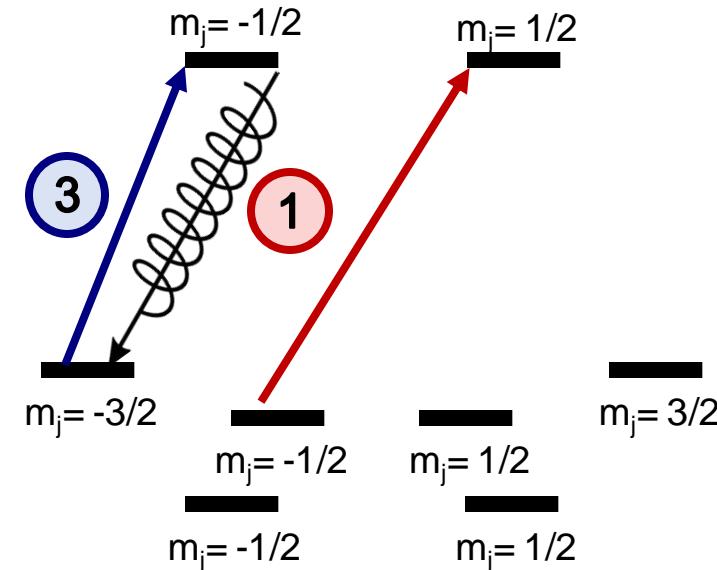
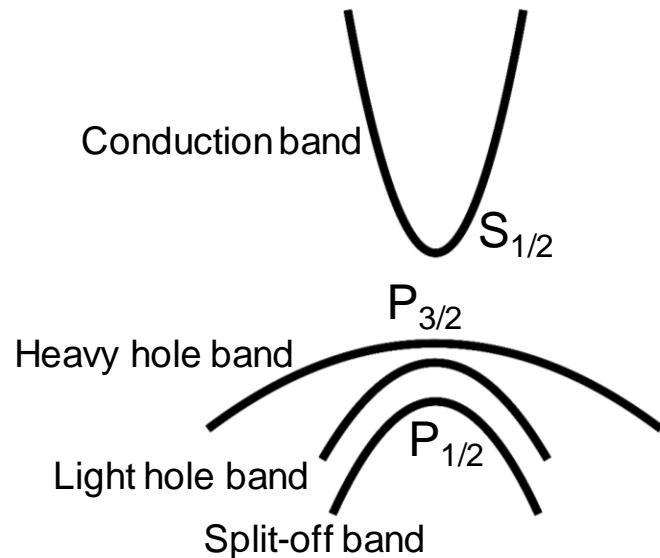


- Laser excitation from $P_{3/2}$ to $S_{1/2}$: $E_{\text{gap}} < E_{\gamma} < E_{\text{gap}} + \Delta$
- Electron Polarization: $P_e < \frac{3-1}{3+1} = 50\%$
- Reverse electron polarization by reversing light polarization



How does spin selectivity arise in III-Vs?

- Circularly polarized light couples to electron angular momentum
- Degeneracy limits the theoretical maximum spin polarization
- **Confinement and strain break heavy hole/light hole degeneracy**



$$\eta_{Electron\ Spin\ Polarization} = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}$$

$$\eta_{ESP} = \frac{3 - 0}{3 + 0} = 100\% ! \quad \eta_{ESP} =$$



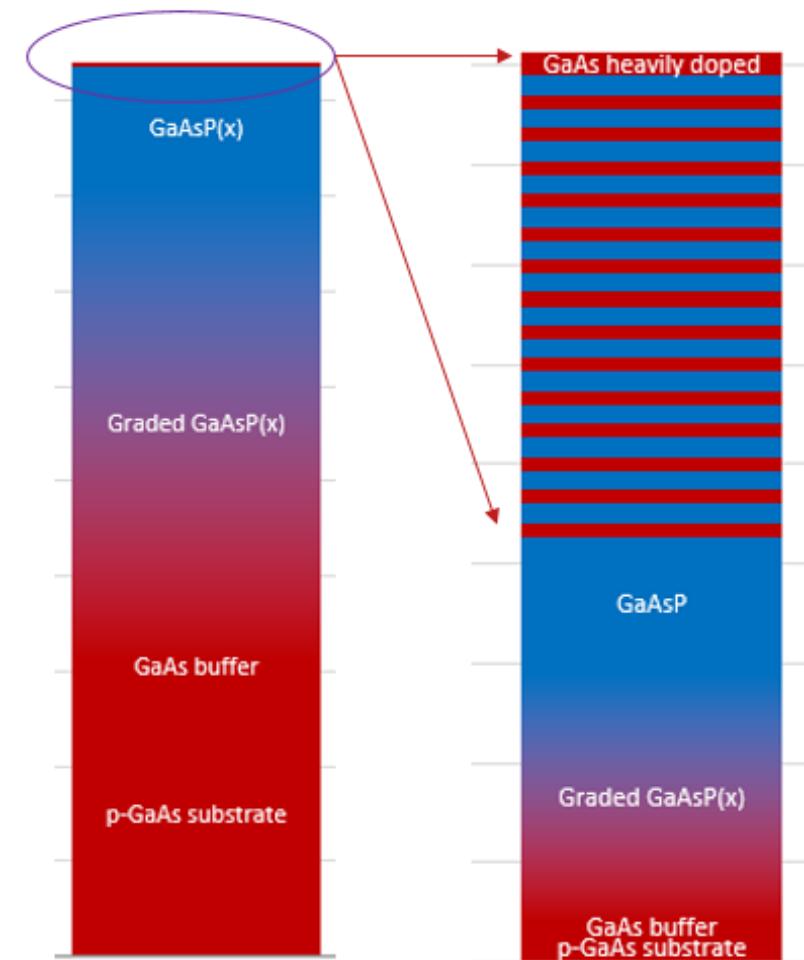
Strained Superlattice Development through SBIR program

- SVT SBIR Partnerships with SLAC or JLab for high polarization photocathodes:
 - Phase I: 2001, 2005, 2007, 2012, 2013
 - Phase II: 2002, 2008, 2013, 2014
- Compositions
 - **GaAs/GaAsP**
 - GaAsSb
 - AlGaAs/GaAs
 - *Distributed Bragg Reflector*
- Parameters
 - Quantum Well thickness
 - Barrier thickness
 - Dopant concentration
 - Number of periods

SVT no longer producing



FOA 20-2310: Initiative to restore high polarization photocathode supply



To scale

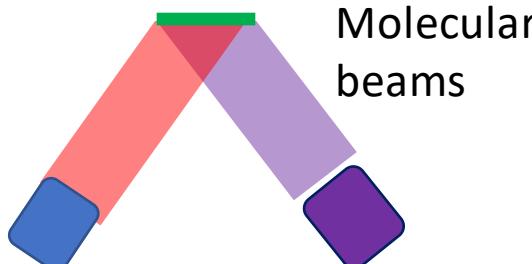
Superlattice
scaled up

MBE, GSMBE, CBE and MOCVD

MBE

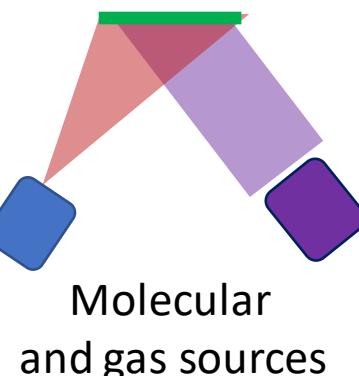
Molecular Beam Epitaxy
elemental As, P, Ga

- Pressure $\sim 10^{-8}$ mbar
- Growth rates $\sim 1 \mu\text{m}/\text{hr}$
- Very precise control



GSMBE

Gas Source Molecular Beam Epitaxy
 AsH_3 , PH_3 , elemental Gallium

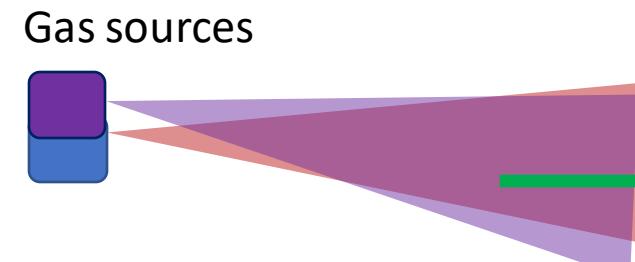


CBE

Chemical Beam Epitaxy

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

- Pressure $< 10^{-4}$ mbar
- Growth rates $0.5\text{-}1 \mu\text{m}/\text{hr}$



MOCVD

Metal organic chemical vapor deposition

AsH_3 , PH_3 , trimethylgallium (TMGa)

- Pressures > 100 mbar during growth
- Growth Rates $10 \mu\text{m}/\text{hr}$
- Traditionally difficult to get sharp interfaces

Photocathode Growth at UCSB

**U California
Santa Barbara
Semiconductor
Deposition System**

- CBE and MBE growth
- ARPES, XPS, STM, LEED, Auger analysis
- Half-metal Heusler Alloys – potential 100% photocathode
- Collaborators for growing GaAs/GaAsP SSL

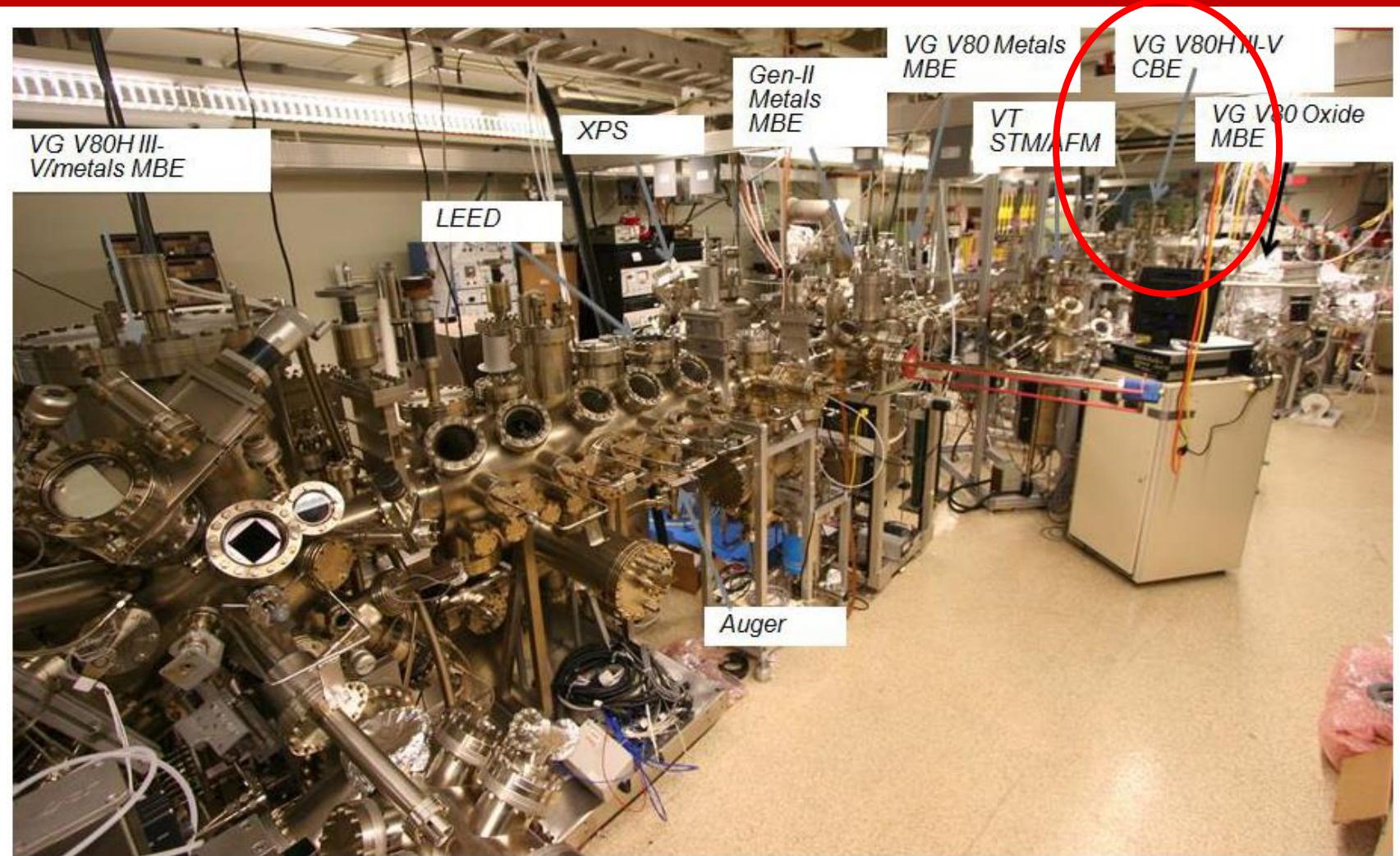
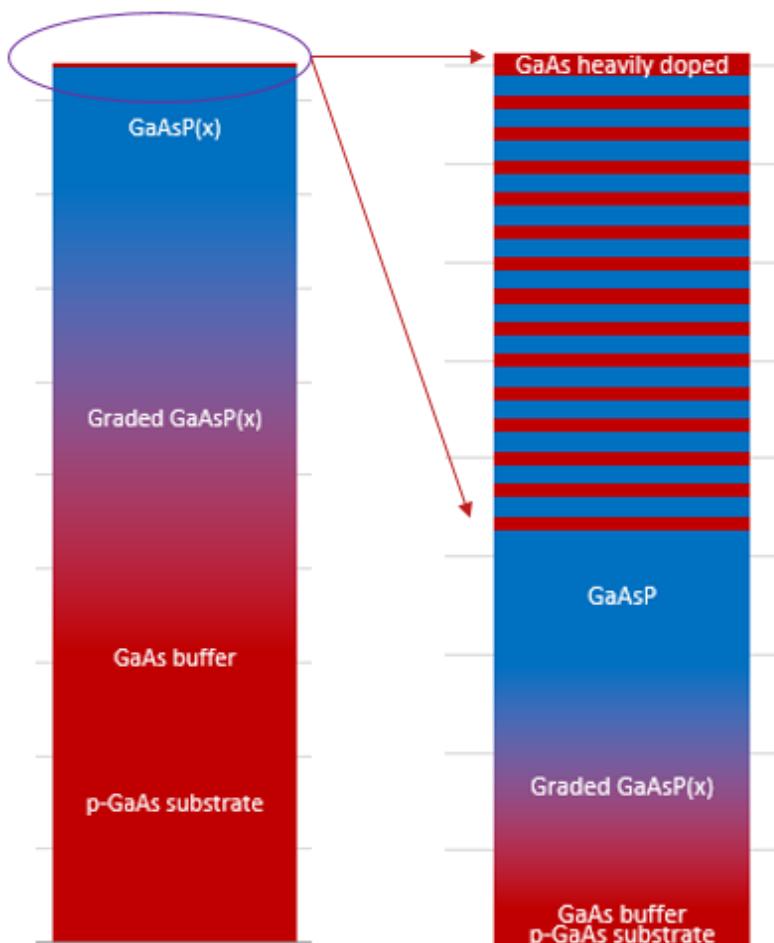


Figure 2 Semiconductor deposition system at Chris Palmstrom's lab at UCSB. The CBE system for the growth of this material is shown at the back and labelled "VG V80H III-V CBE".

Original Research Plan



To scale

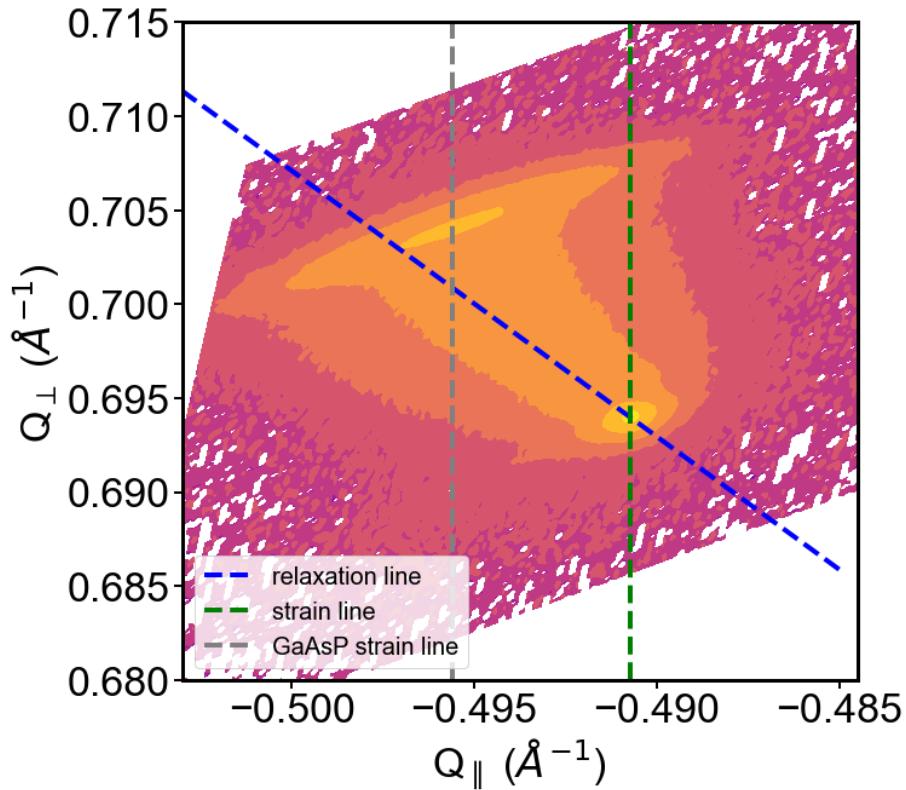
Superlattice
scaled up

1. Grow GaAs/GaAsP: UCSB CBE instead of MBE
2. Measure Polarization: JLab
3. Use Photocathodes!

Obstacles -> Innovation



UCSB Highlights: Graded layer GaAs to GaAsP



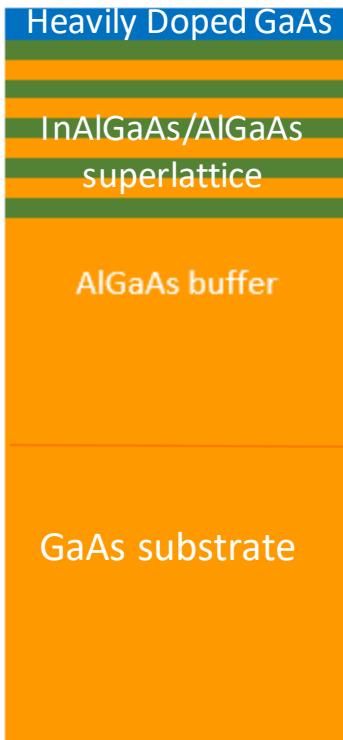
X-ray Reciprocal space mapping

- Plot of lattice distance during growth
- Graded Layer with minimal strain
- GaAs layer (5-10 nm) strained: lattice constant that of GaAsP

- Graded layer: slow, \$\$, defects
- Triethyl-gallium + P: high vapor pressure residue
 - Return to solid source Ga
 - CBE becomes MBE
- Rebuild system, recalibrate growth parameters with new heaters & sources
- Meanwhile Literature Review
 - Try InAlGaAs/AlGaAs
 - Photoemissive Layer
 - Non-emissive lattice mismatched layer

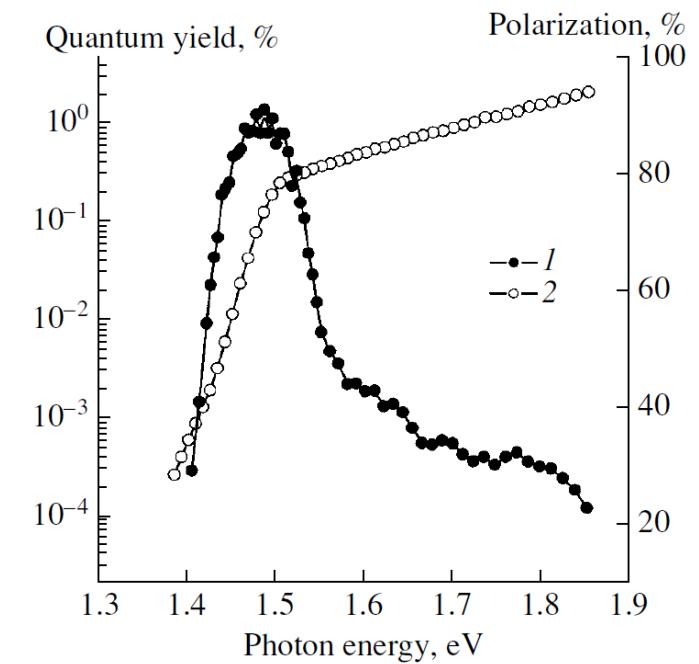


Benefits of InAlGaAs/AlGaAs



- No virtual substrate necessary
 - AlGaAs almost perfectly lattice matched to GaAs:
Grow directly on GaAs
 - No lateral undulations from virtual substrate
- Best cathodes comparable with best GaAs/GaAsP
 - Large sample-sample variations
 - Terminated with GaAs

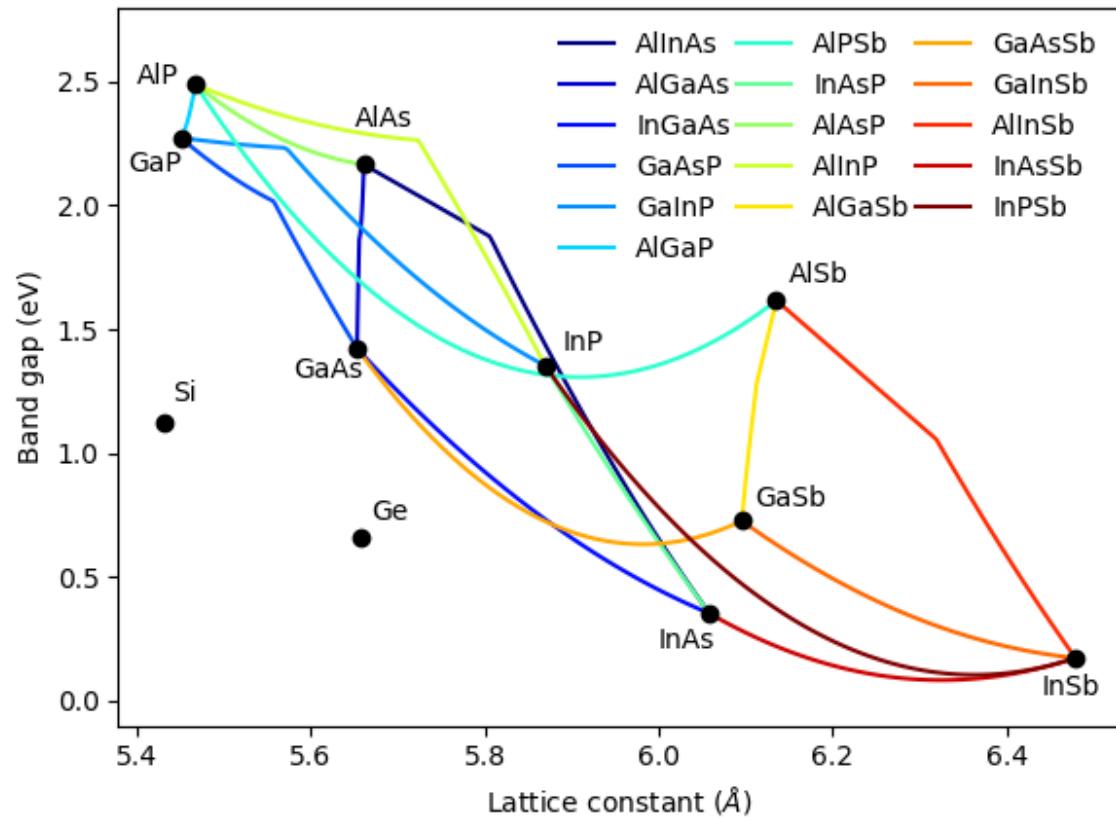
Easily tunable DBRs
AlAs/AlGaAs for DBR
well characterized optical constants
abrupt interfaces



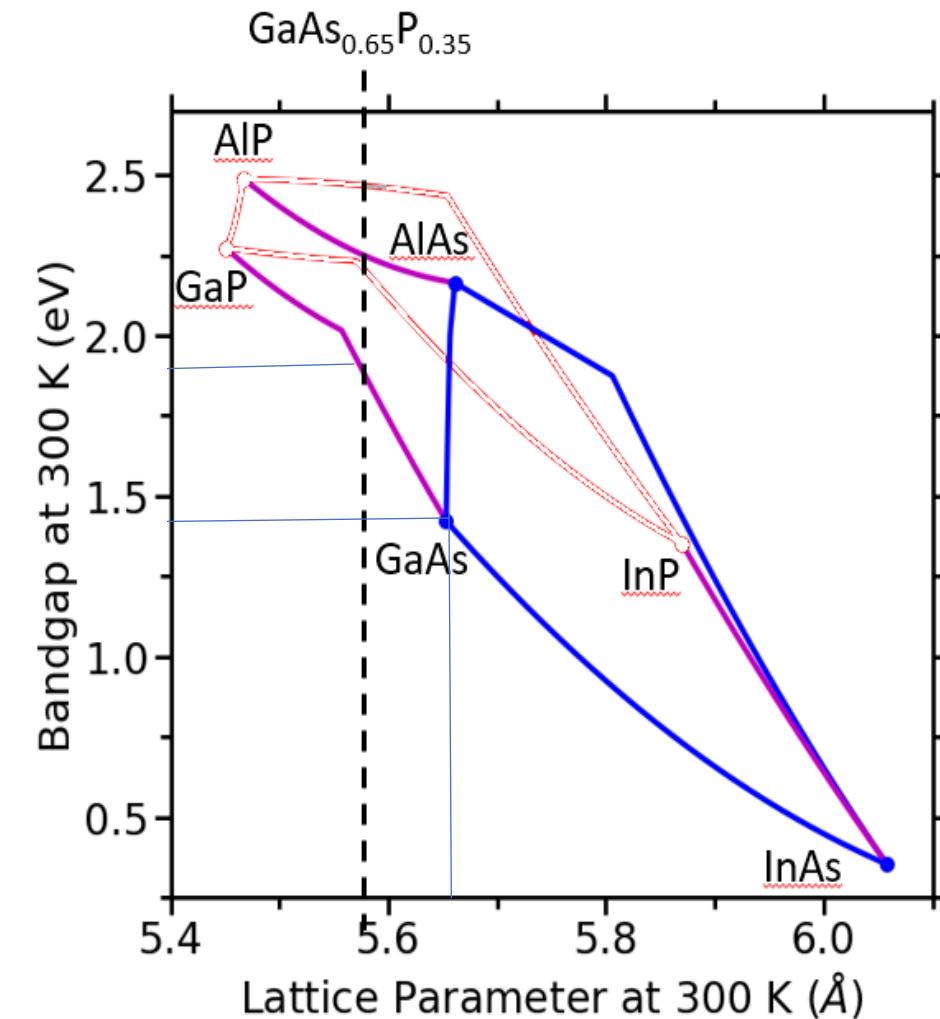
[1] L. G. Gerchikov, et al. *Semiconductors* **40**, 1326–1332 (2006)

Strained well: GaAs/GaAsP

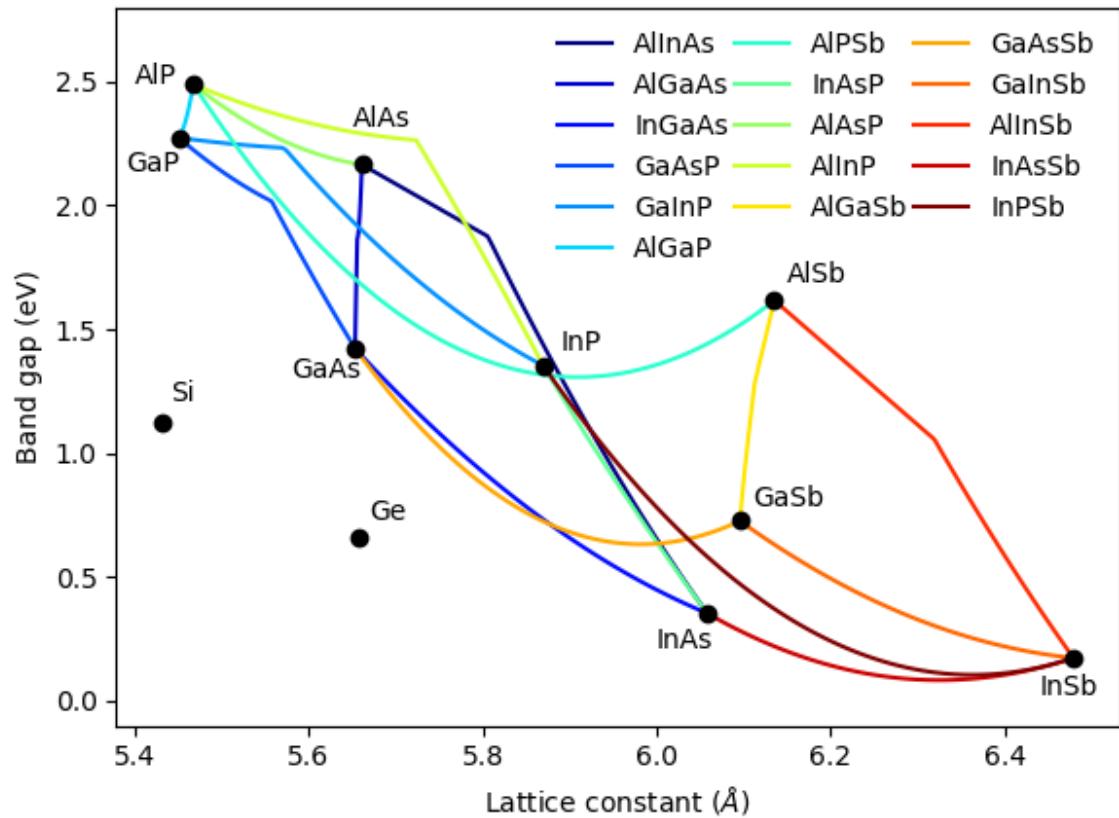
- Strain and valence band offset coupled: both fixed by virtual substrate



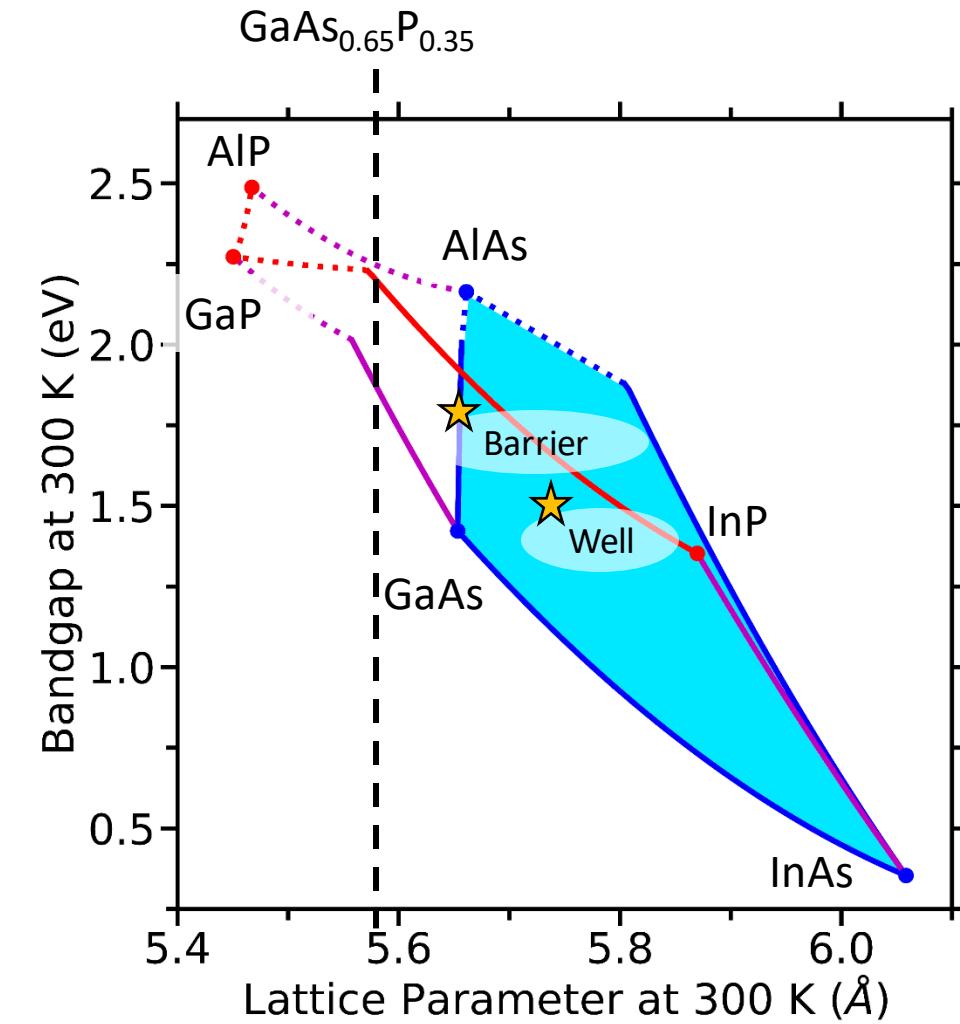
III-IV semiconductor alloys: Band gaps and lattice constants



Strained well: InAlGaAs/AlGaAs



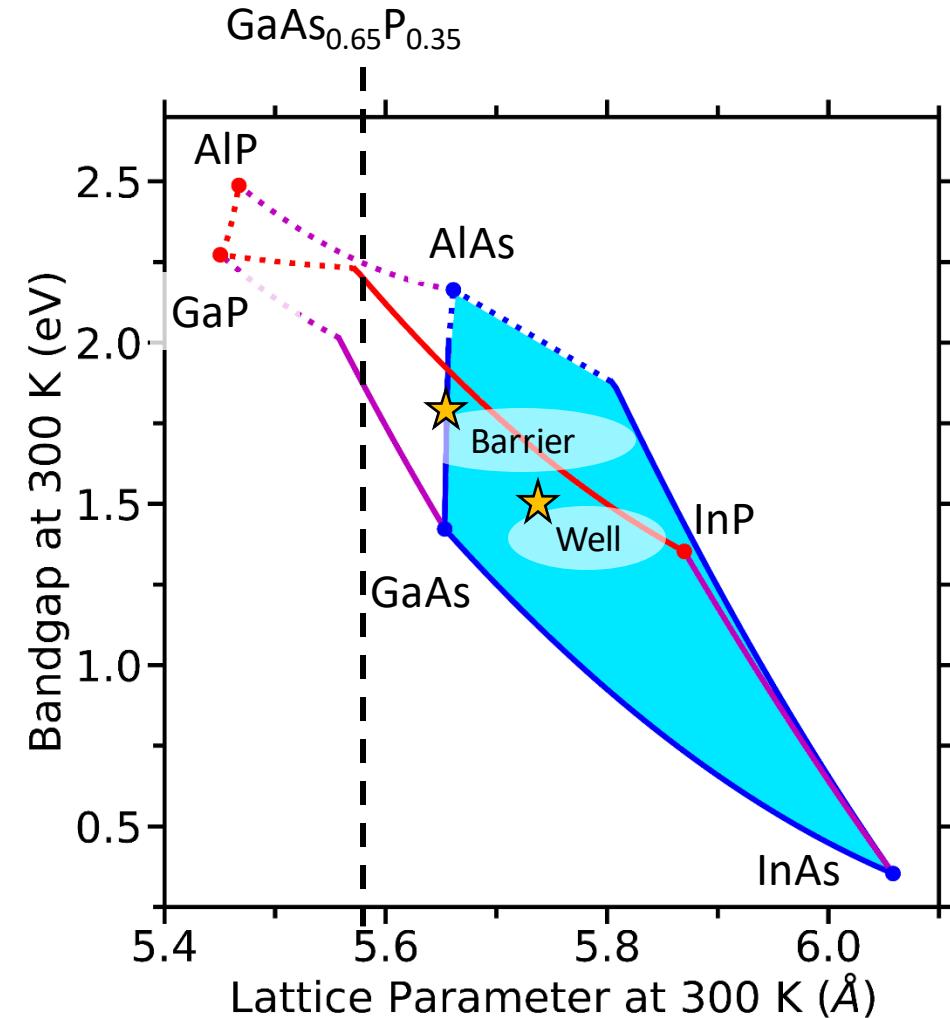
III-IV semiconductor alloys: Band gaps and lattice constants



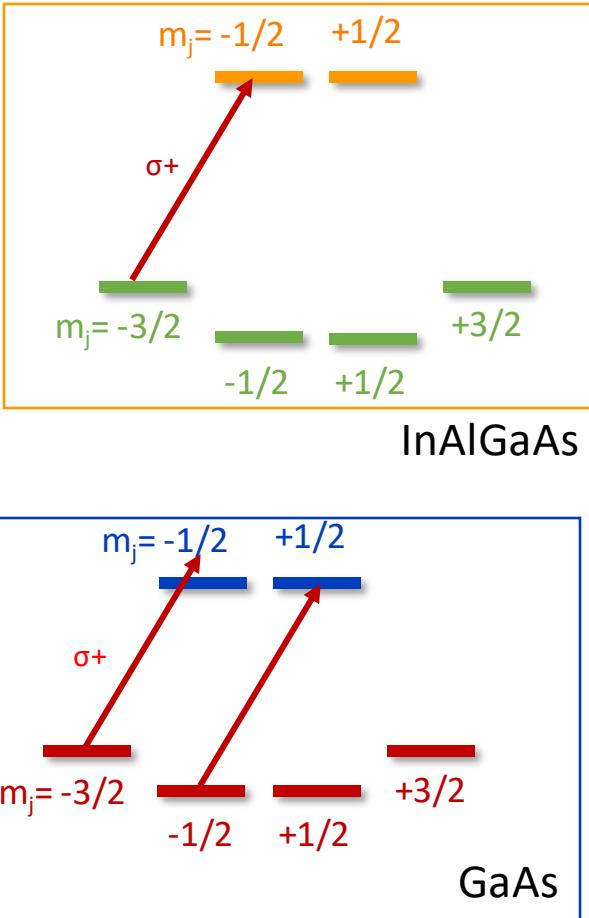
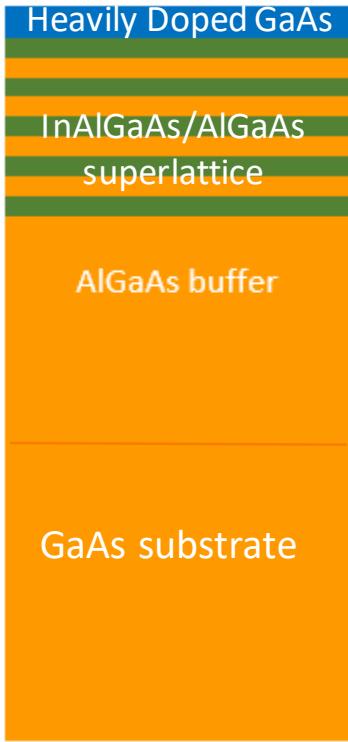
Strained well: InAlGaAs/AlGaAs

- No virtual substrate necessary!
- Much more band engineering possible
 - Zero conduction band offset [1]
 - Graded barrier heights
- Better growth temperature agreement
- Best reported InAlGaAs/AlGaAs photocathodes are comparable to GaAs/GaAsP
- Easily tunable DBRs in AlAs/AlGaAs system

[1] L. G. Gerchikov, et al. *Semiconductors* **40**, 1326–1332 (2006)

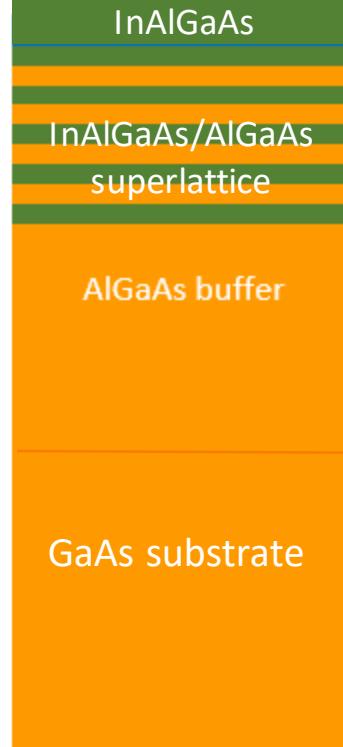


Polarized Emission from InAlGaAs/AlGaAs



- InAlGaAs has larger band gap than GaAs
- Additional photon energy will excite both GaAs bands InAlGaAs/GaAs superlattice

“Excites to opposite channel as superlattice”



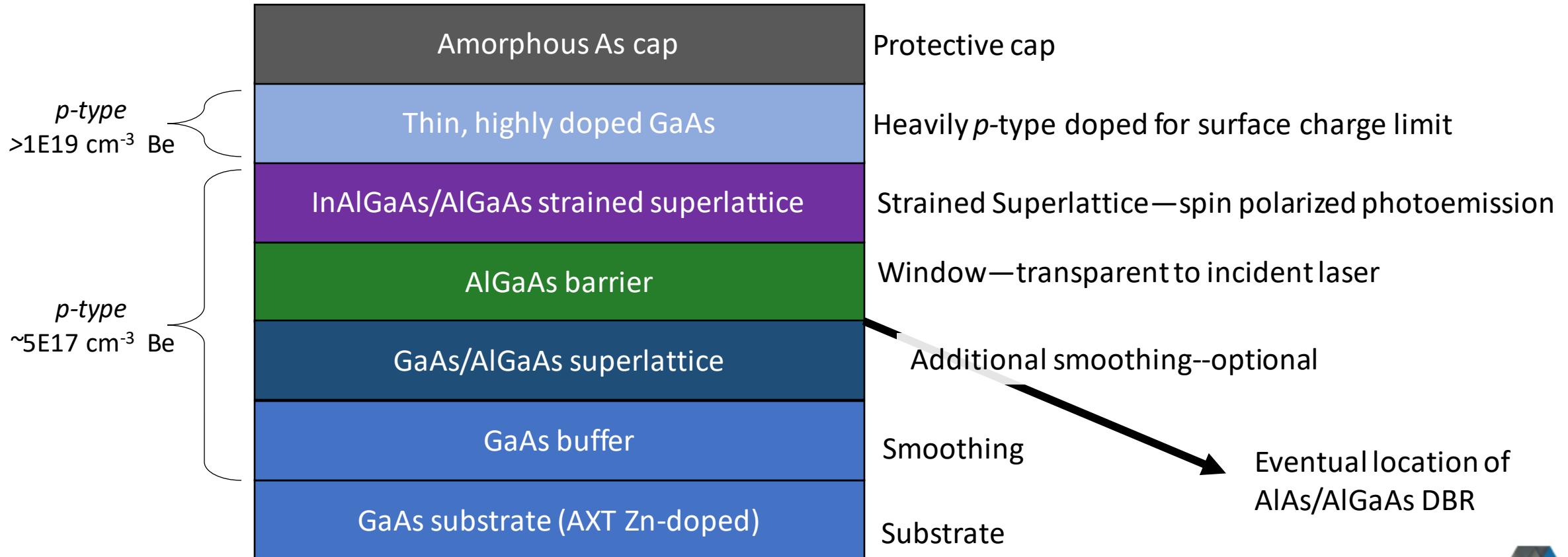
Excites to same channel as superlattice



Completely transparent to pump light



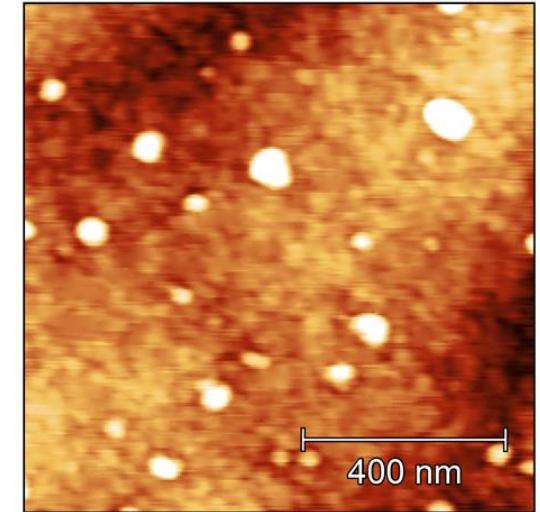
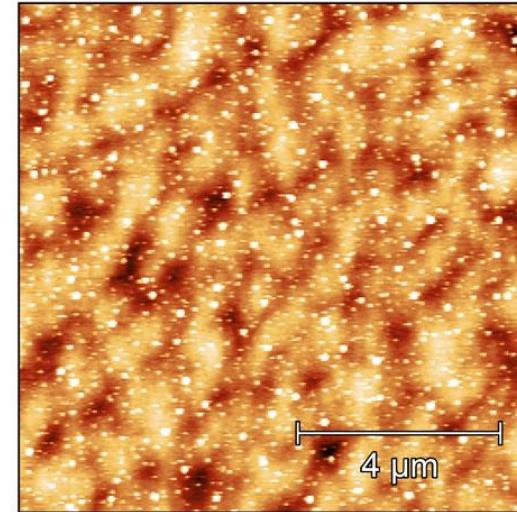
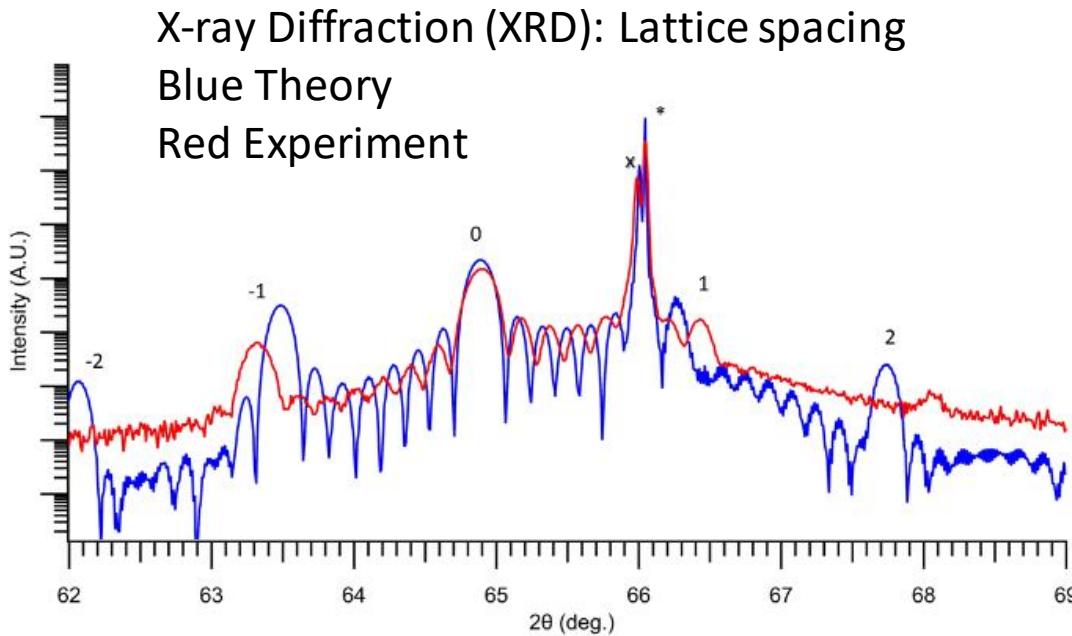
UCSB InAlGaAs/AlGaAs Structure



Based on Mamaev et al., Appl. Phys. Lett. 93, 081114 (2008) and
<https://www.slac.stanford.edu/pubs/slacpubs/11250/slac-pub-11403.pdf>



First photocathode



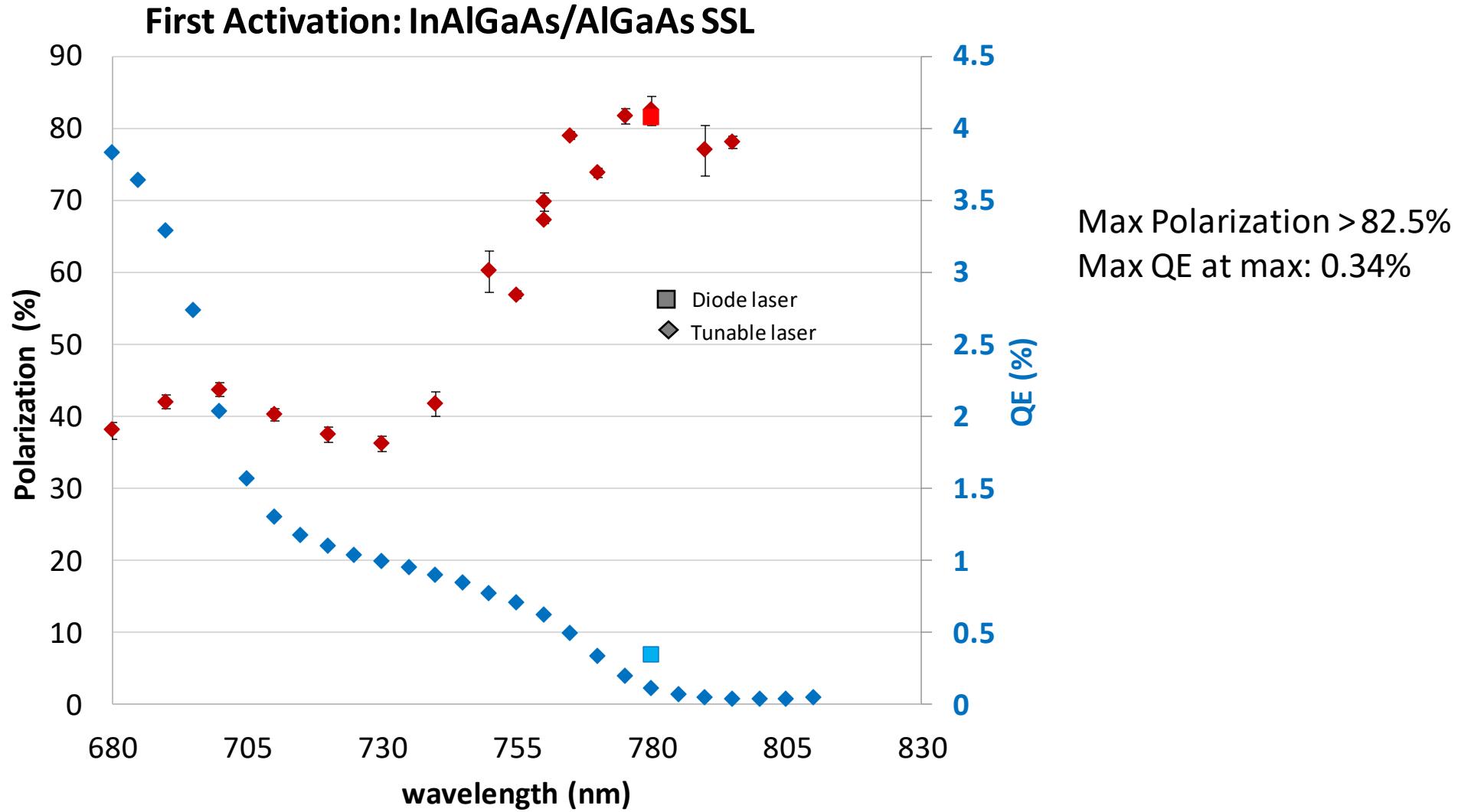
X-ray diffraction measurement of Superlattice

- Fully strained
- Superlattice period good - 8% less than goal

Atomic Force Microscope surface morphology

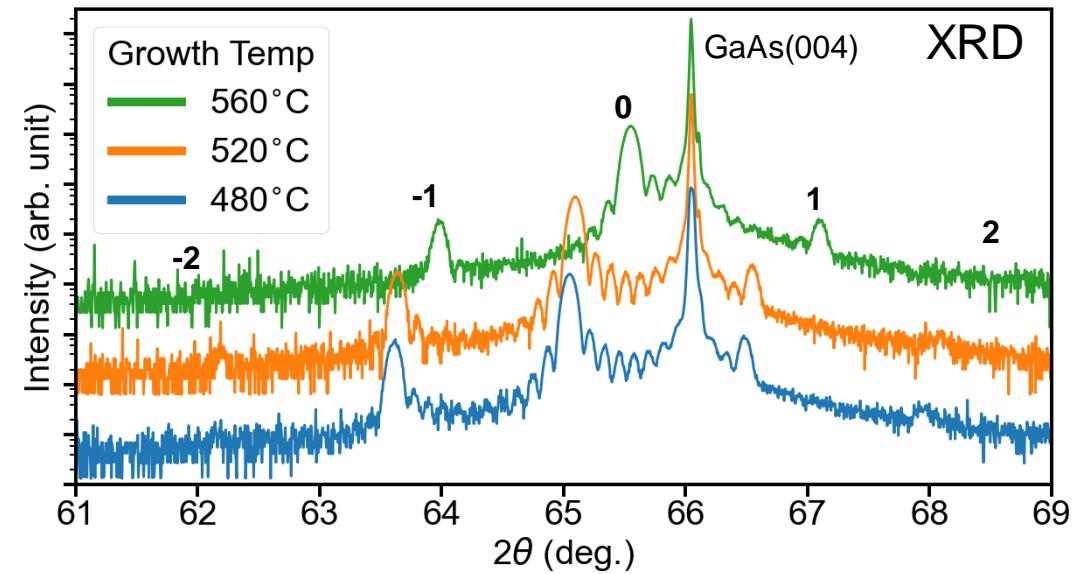
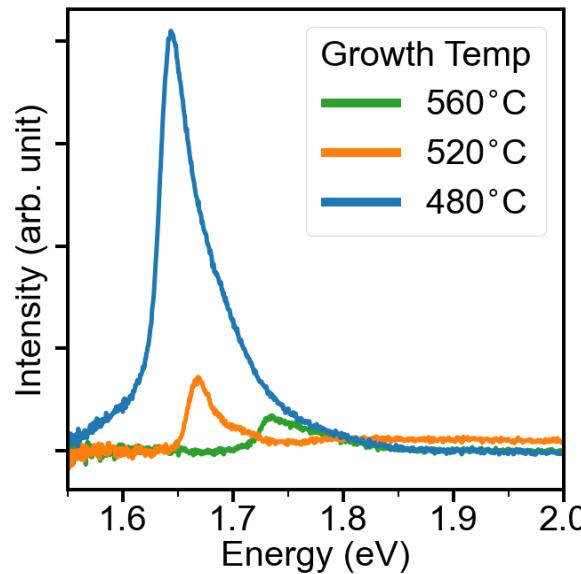
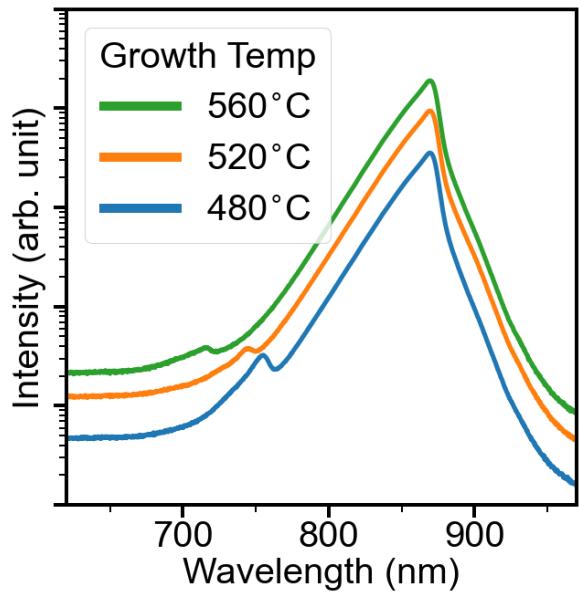
- Verification of arsenic cap coverage
- Some excess As – will desorb in first heat cycle

First Activation InAlGaAs/AlGaAs SSL



Vary Growth Temperature for InAlGaAs/AlGaAs

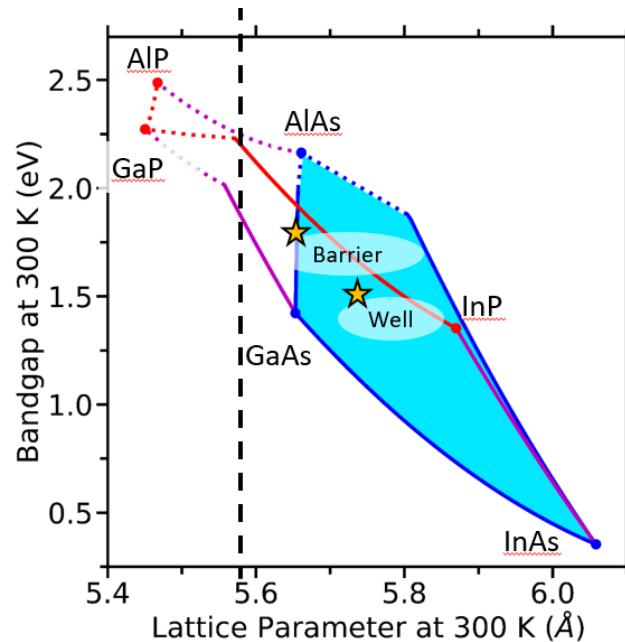
Photoluminescence



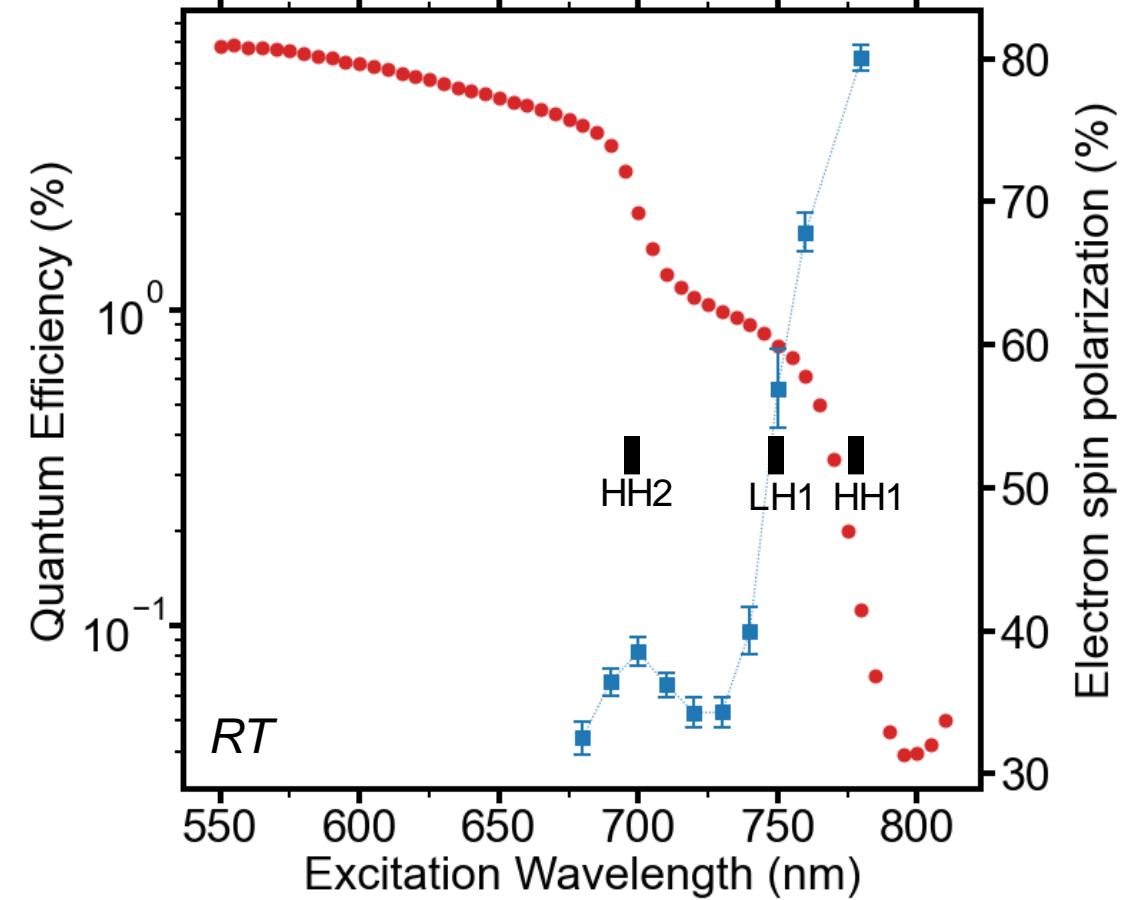
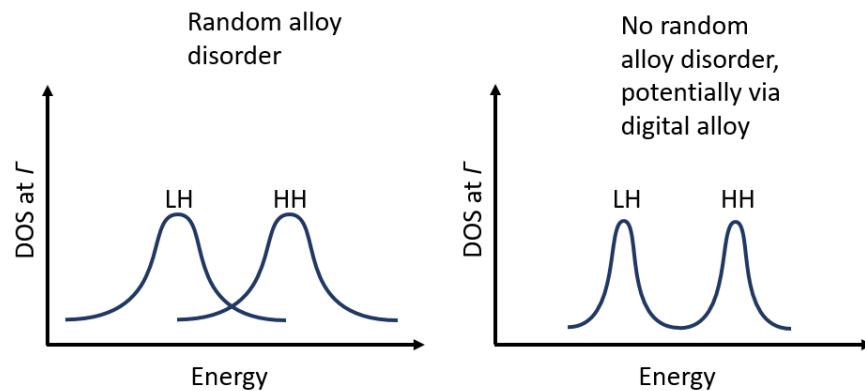
- Lower temperature growth produces more intense PL from superlattice
- XRD shows more visible fringes in films grown at lower temperature, indicating sharper interfaces

520°C sample tested, 480 and 560°C in the queue

Random Alloy Disorder



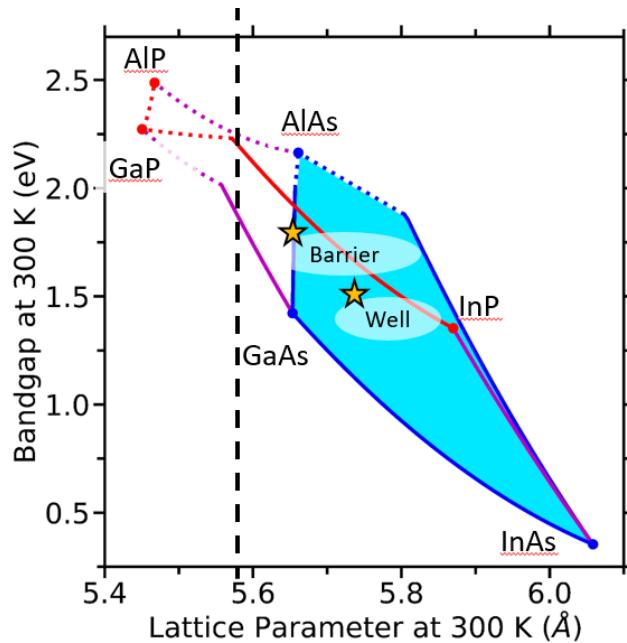
- Quaternary well (InAlGaAs) adds random alloy disorder
 - increased bandwidth
 - decreased spin polarization



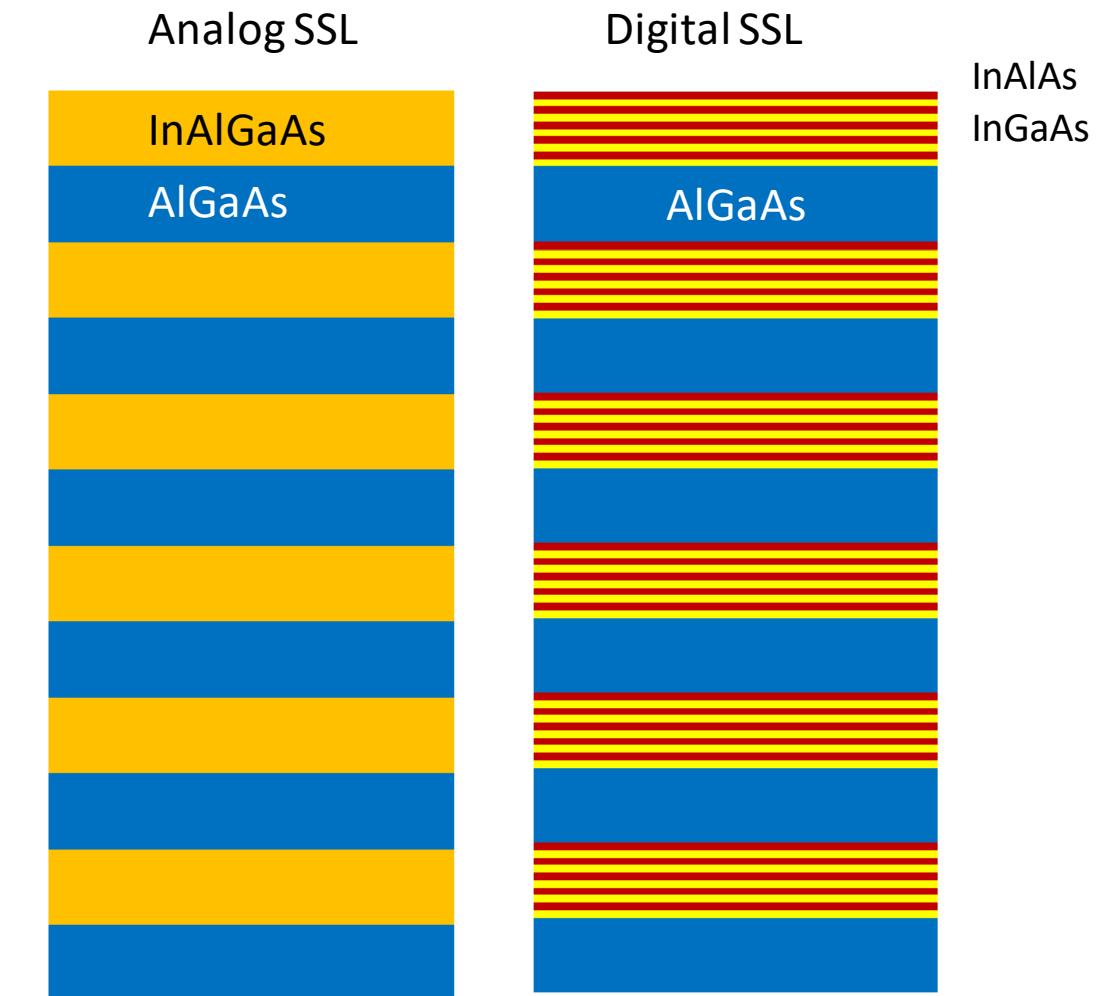
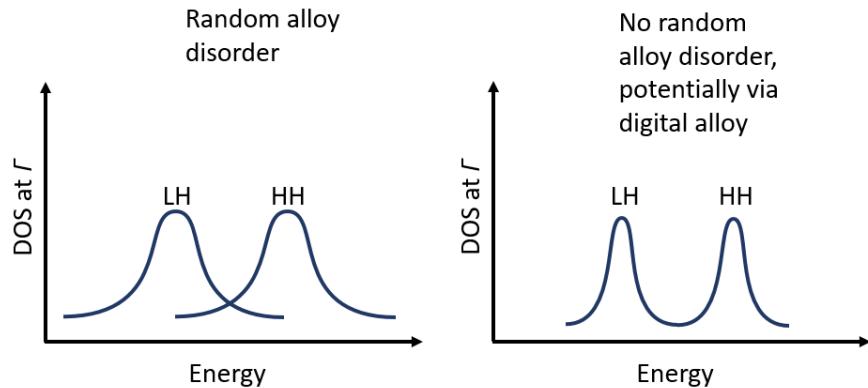
- ~50 meV hole splitting



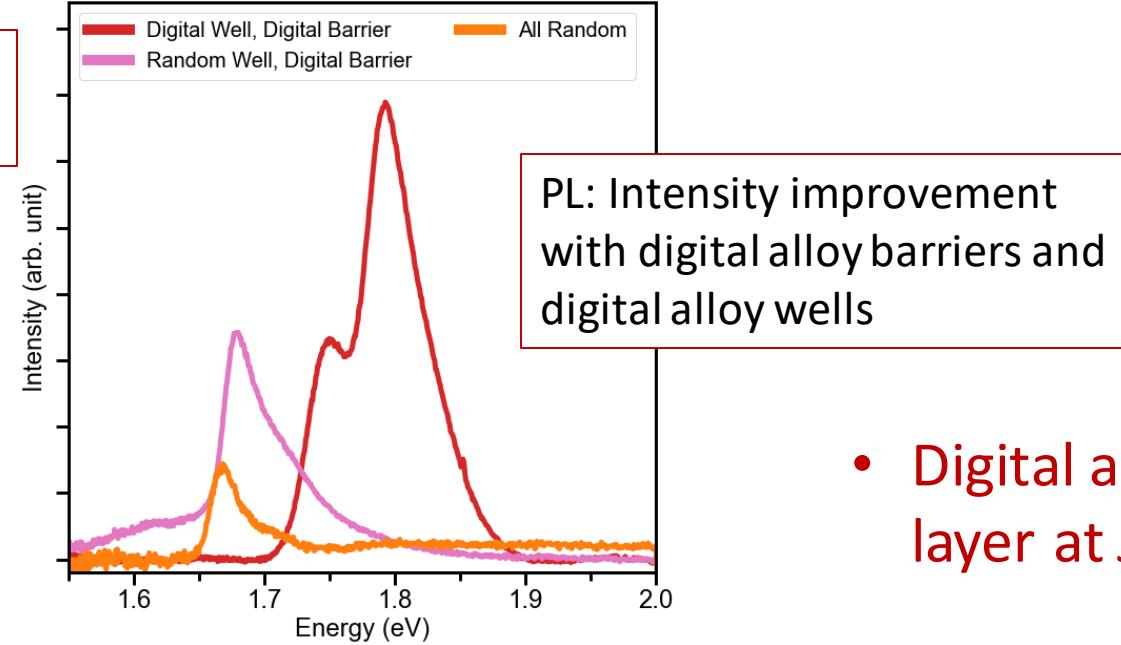
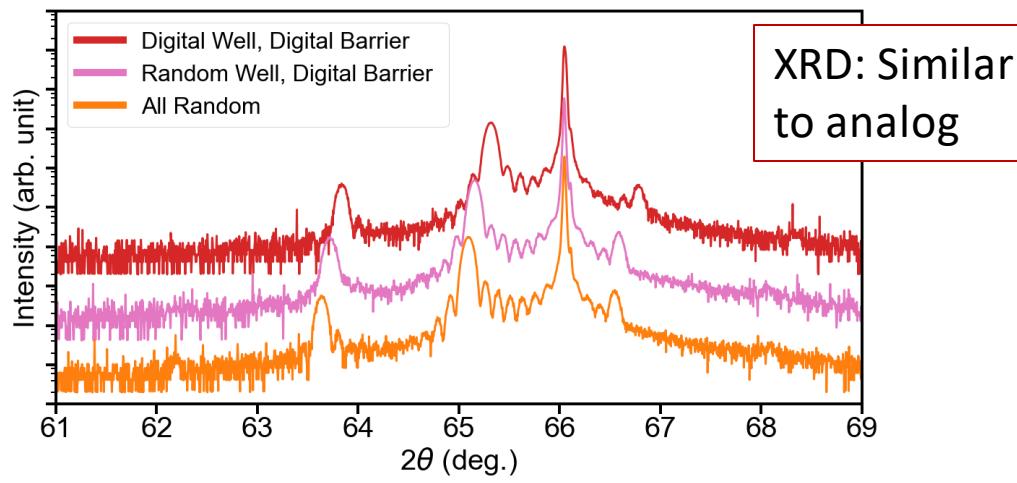
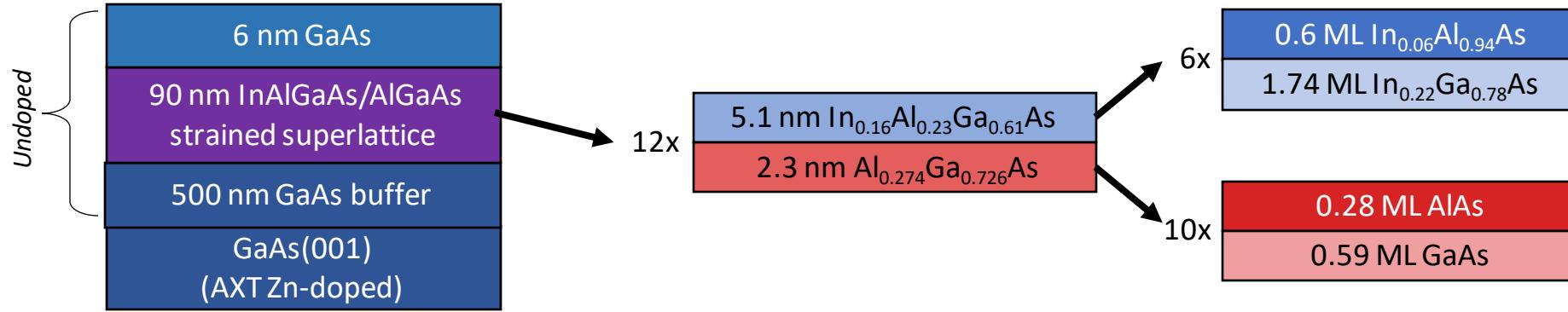
Random Alloy Disorder



- Quaternary well (InAlGaAs) adds random alloy disorder
 - increased bandwidth
 - decreased spin polarization



Digital Alloy Test



- Digital alloy barrier layer at JLab to test



Successful DBR Structures

Distributed Bragg Reflector

- Enhance QE by reflecting light for several passes through SSL
- Designed for peak reflectivity at 770 nm
- Analog and Digital AlAs/AlGaAs DBR structures designed and tested
- Digital Alloy: better uniformity across wafer

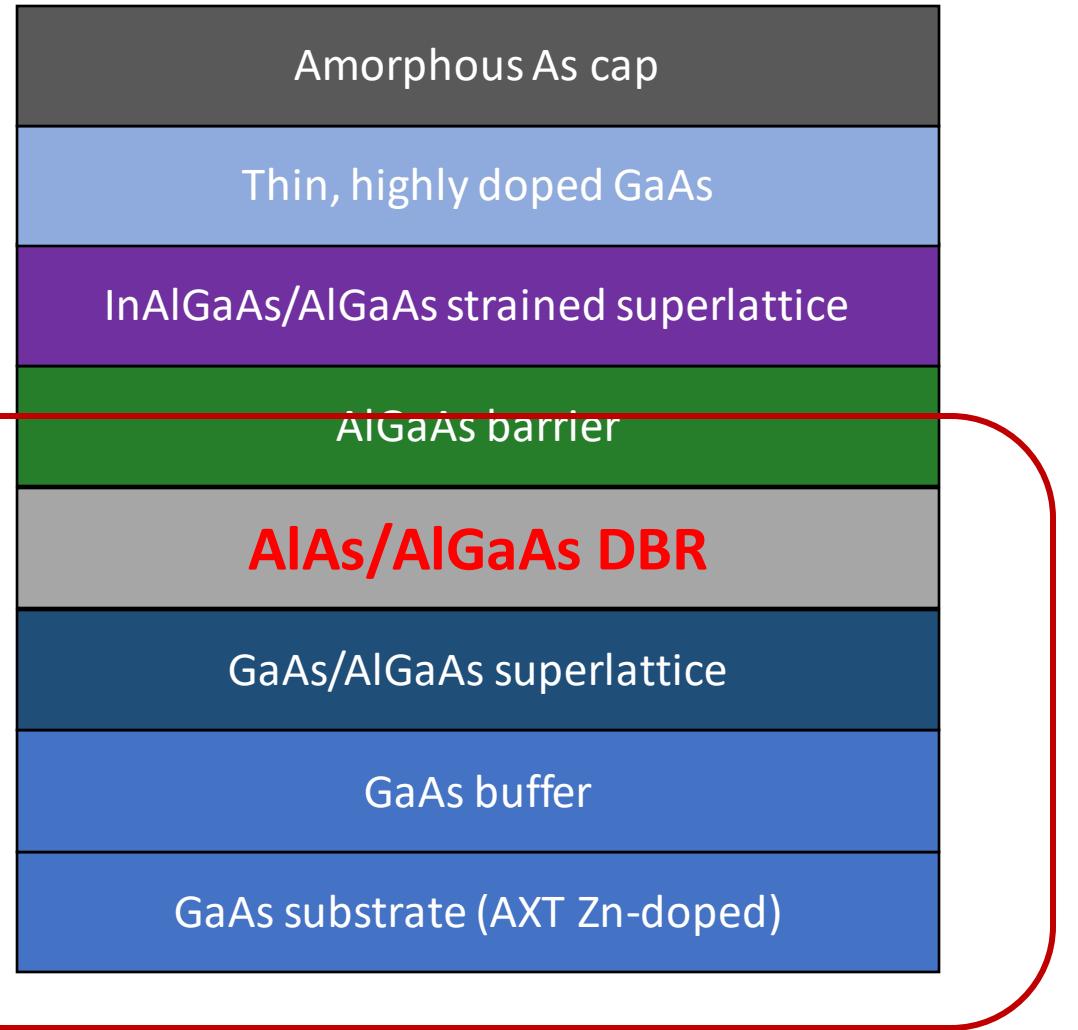
Analog DBR



Digital DBR



GaAs/AlAs superlattice



UCSB Photocathodes

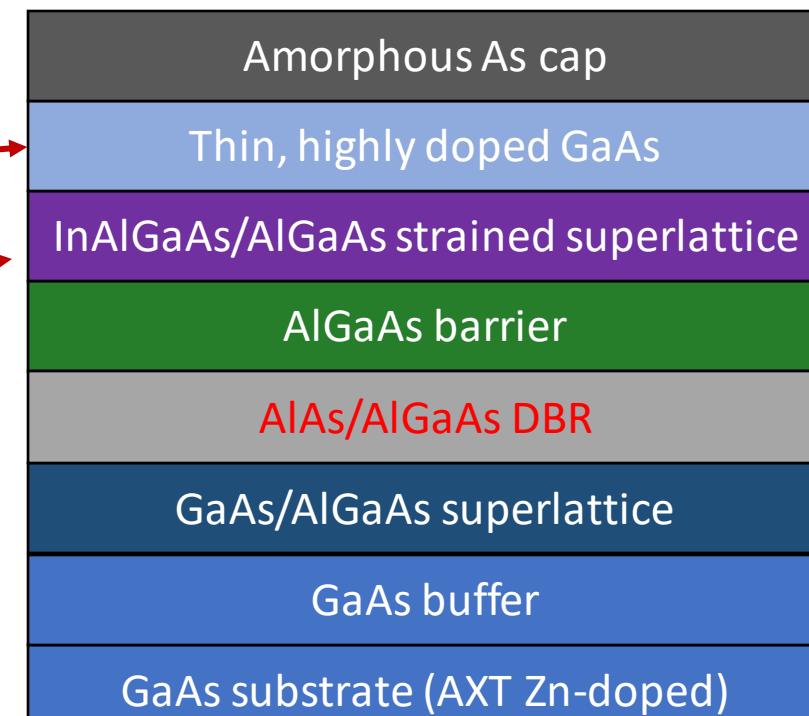
- Varied growth temp: Samples 198, 199
- Increase strain: Sample 144
- Higher dopant top & band gap shift: Sample 143
- Digital alloy barrier layer: Sample 202

At JLab awaiting testing

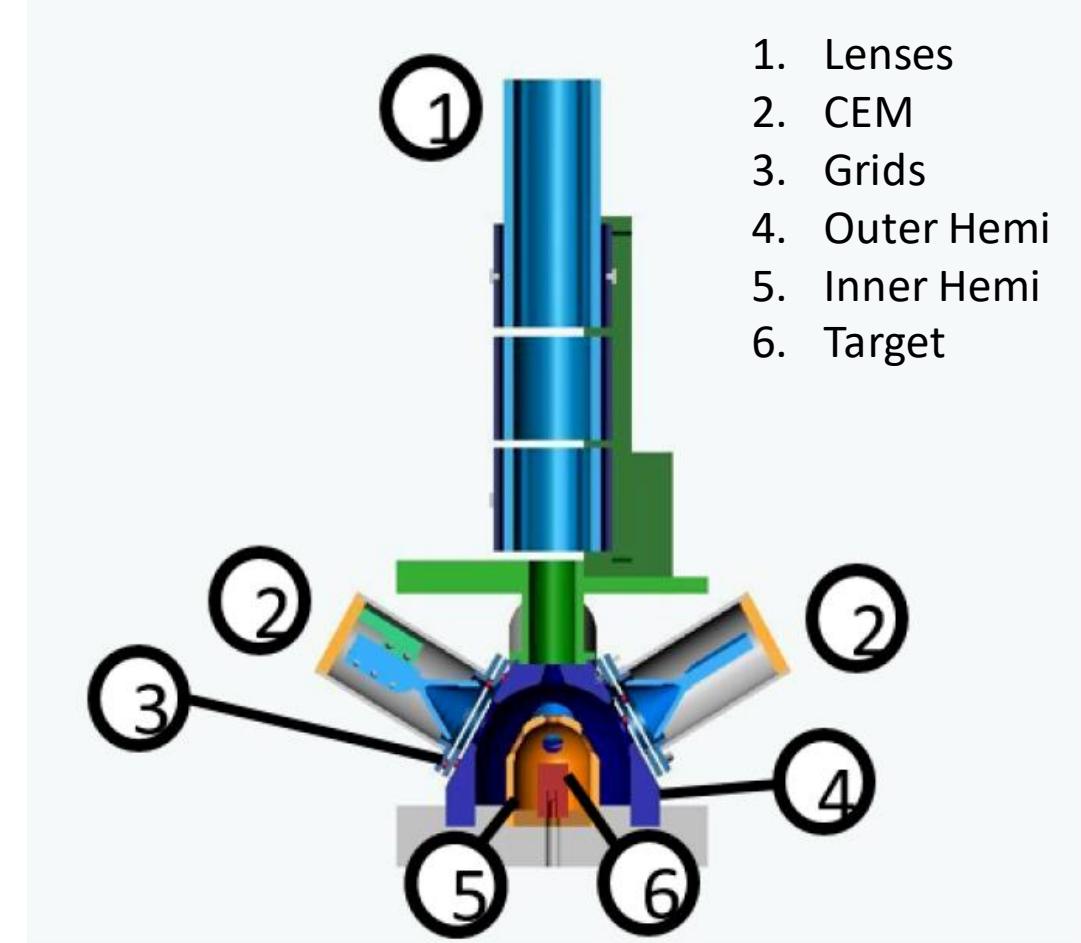
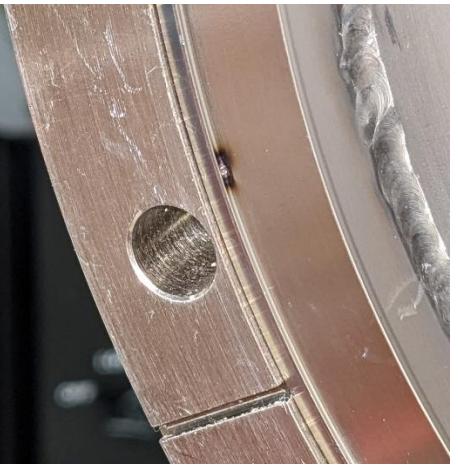
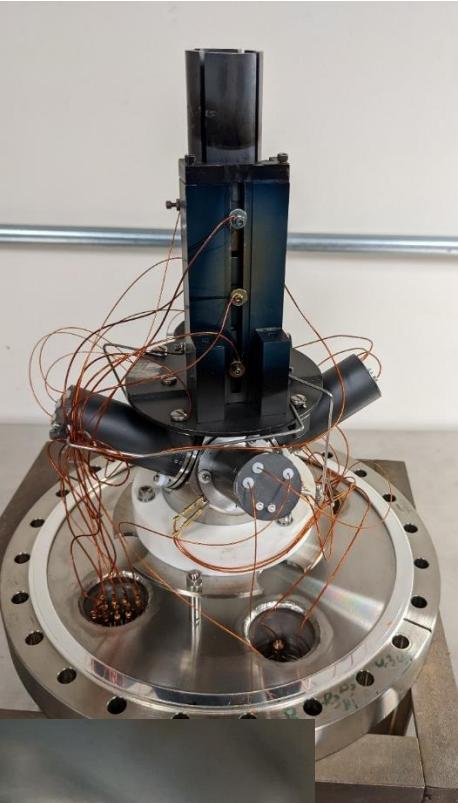
Next

- Different Top Layer
- Distributed Bragg Reflector Photocathode
- Lots of parameters to optimize

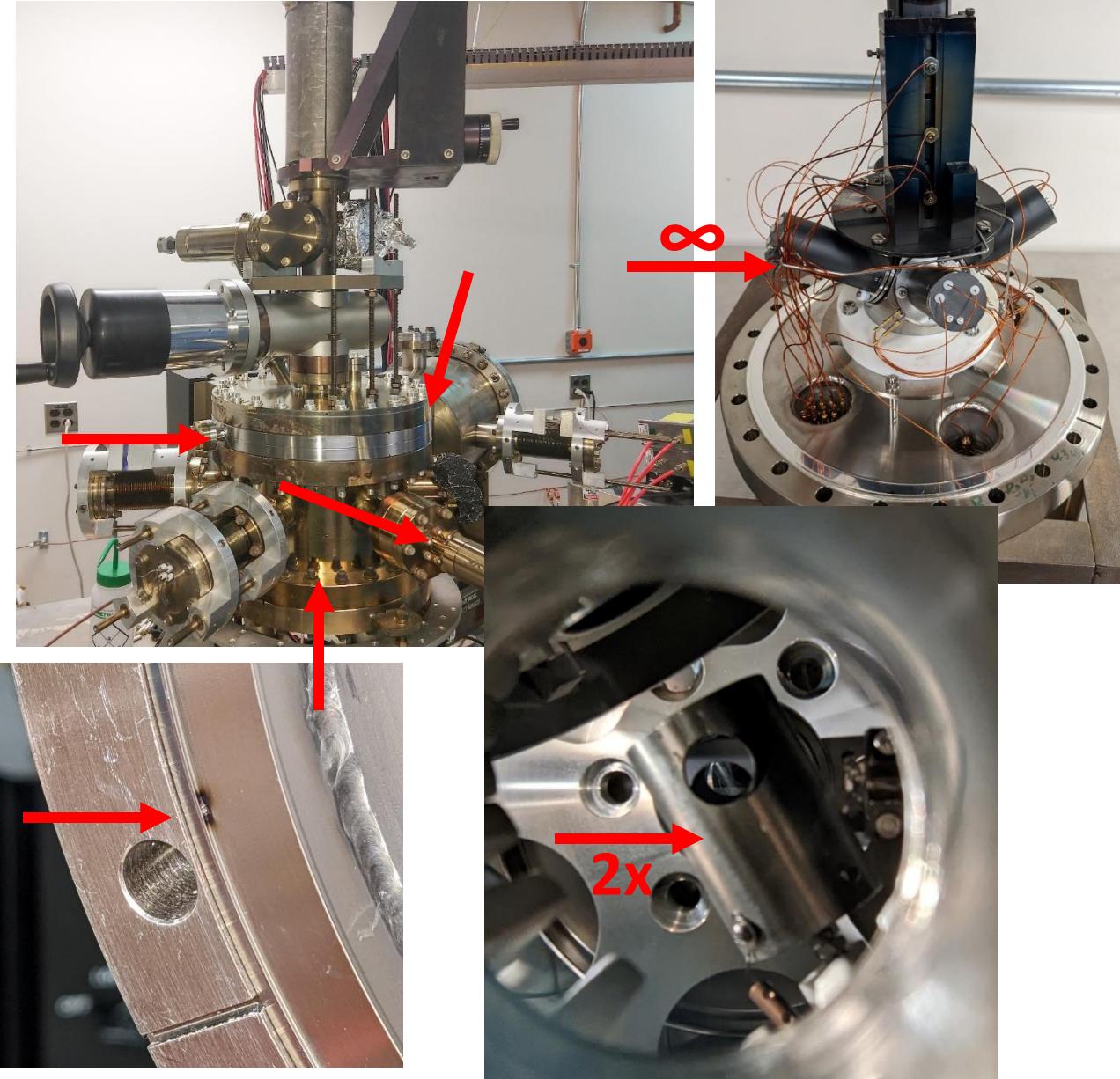
FOA timeline over but more samples coming
- awaiting JLab polarization measurements



JLab Tests: MicroMott



JLab Tests: MicroMott



MicroMott Polarimeter - A Series of Unfortunate Events

- Multiple issues during refurbishment
- Nearly there!
- Wish List: more robust polarimeter with JLab puck load-lock



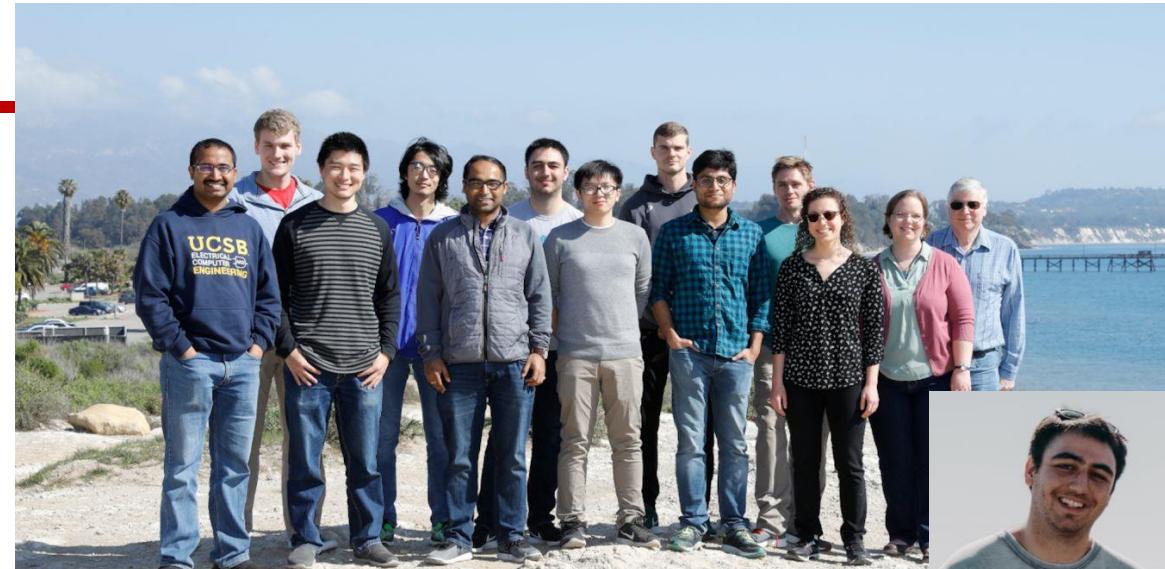
Project Summary

JLab: First UCSB sample tested
Polarimeter nearly ready to continue

UCSB

- Initial GaAs/GaAsP growth characterized
 - Extensive chamber maintenance to remove phosphorous compounds
- InAlGaAs/AlGaAs superior in many aspects
 - Temperature compatibility
 - Strain and band gap independent
 - Higher dopant potential
 - Digital structures for both SSL and DBR
- More to come

Many Thanks to Aaron Engle for photocathode growth,
characterization and slides, and Chris Palmstrøm for guidance



Questions?