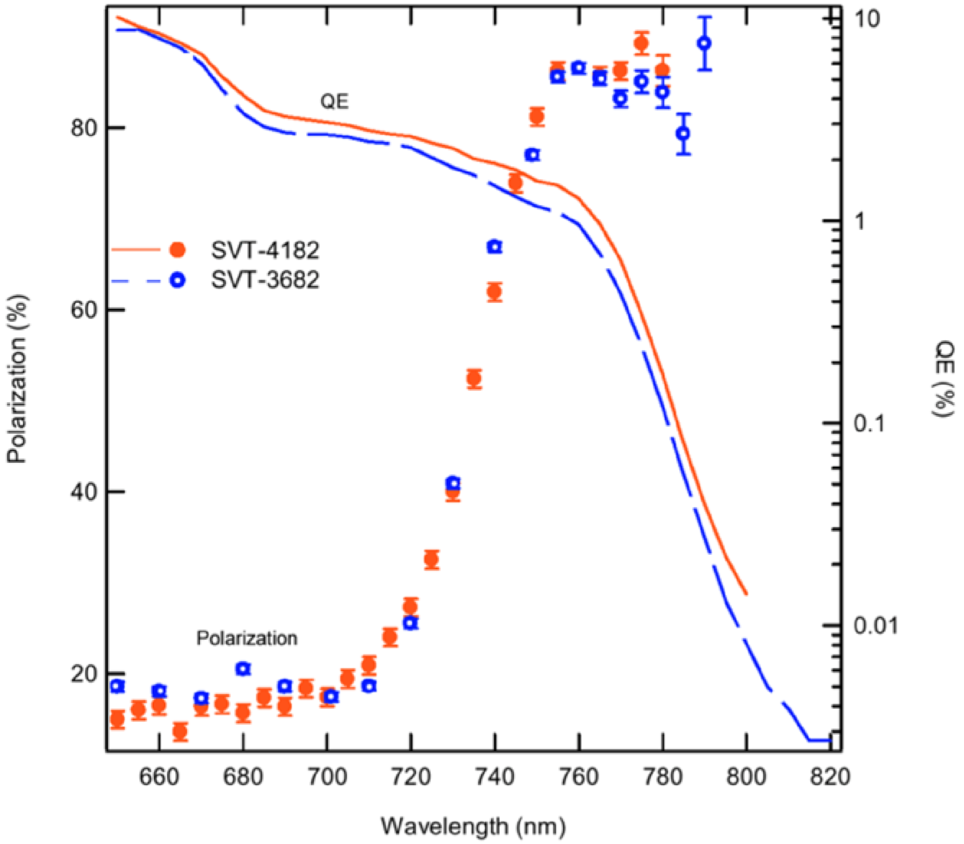
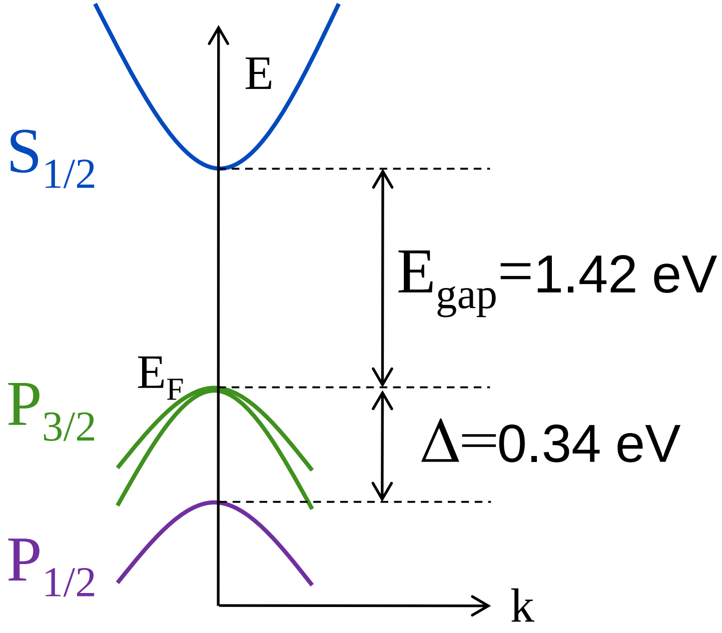
Strained Superlattice GaAs/GaAsP

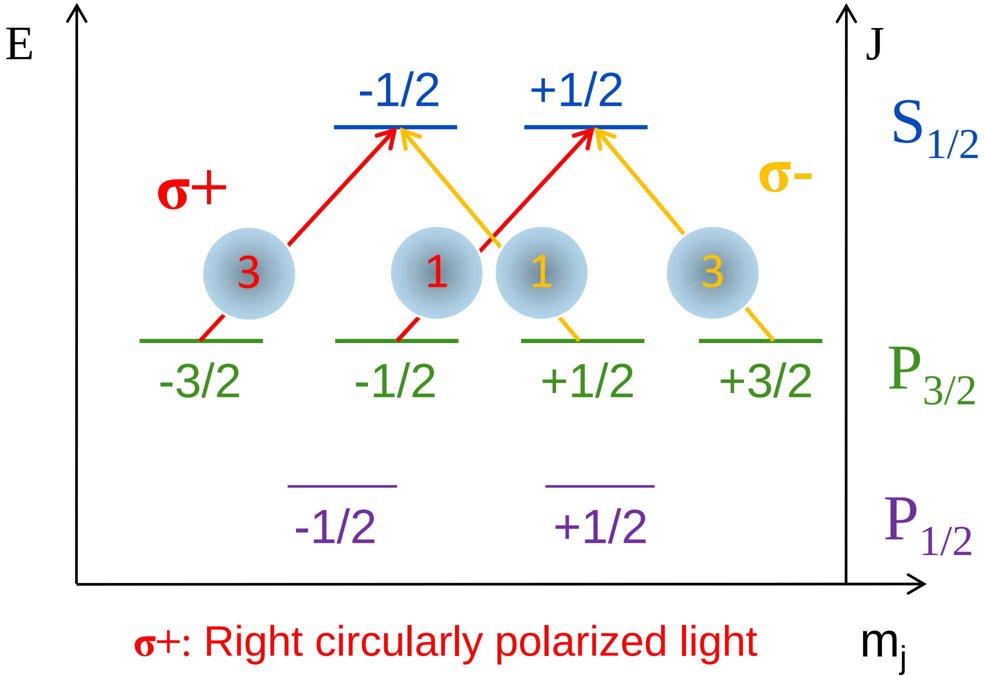
DOE funded efforts to restore High Polarization Photocathode production: Goals and Updates



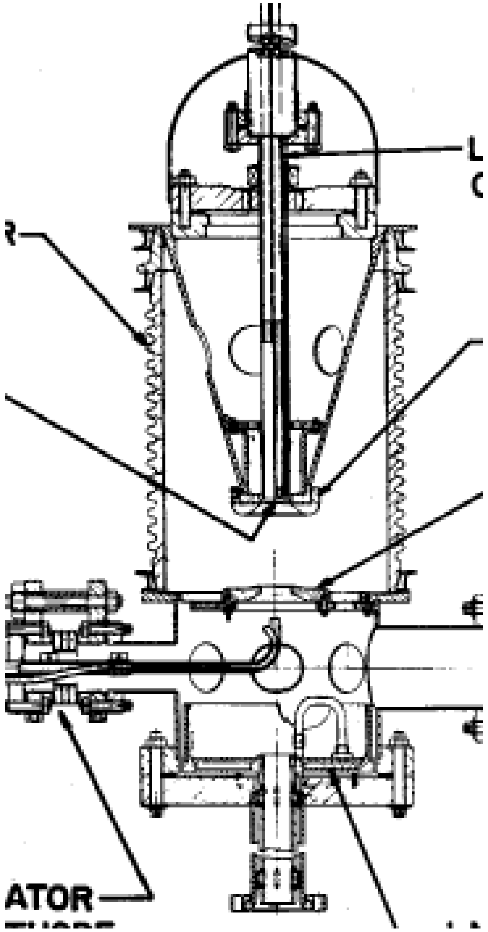
Marcy Stutzman, Jefferson Lab

Spin Polarized Photoemission from Bulk GaAs





σ+: Right circularly polarized light
σ- : Left circularly polarized light

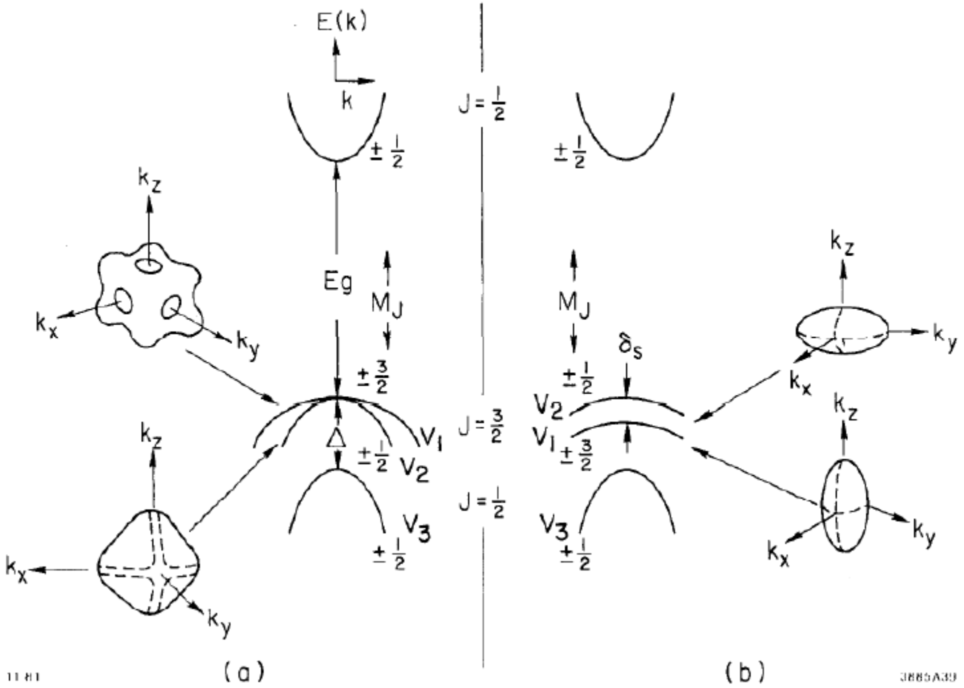


SLAC 1977

Strained GaAs

Breaking the 50% barrier

PhD thesis, Paul Zorabedian, SLAC Report 248, 1982



Application of a uniaxial strain removes the degeneracy of the $P_{3/2}$ state

Eliminate degeneracy of $P_{3/2}$ state via "Interface Stress Method"

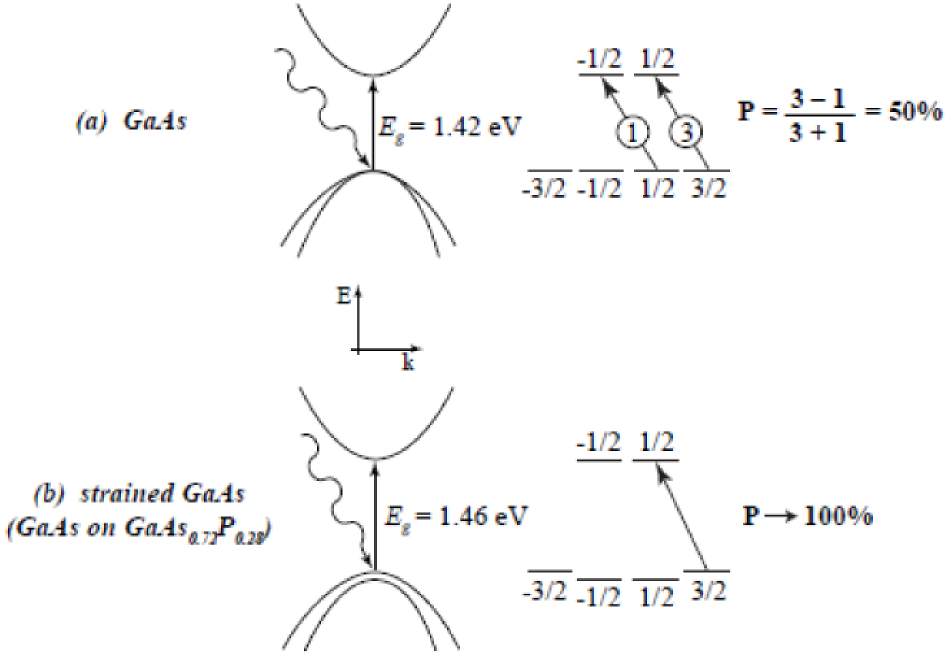
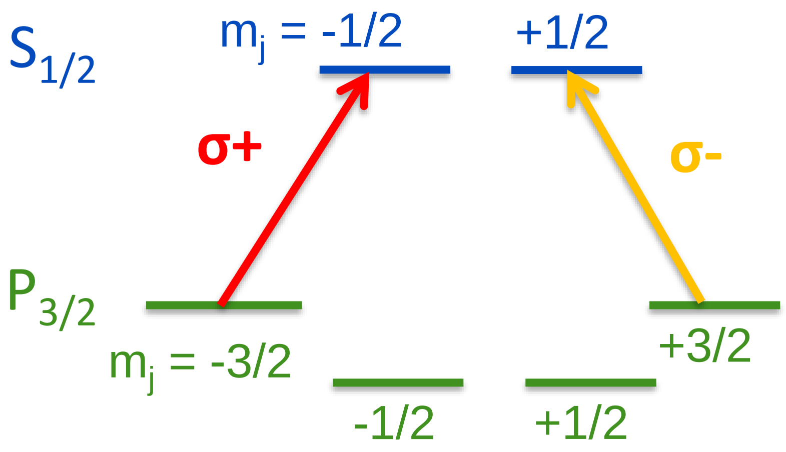
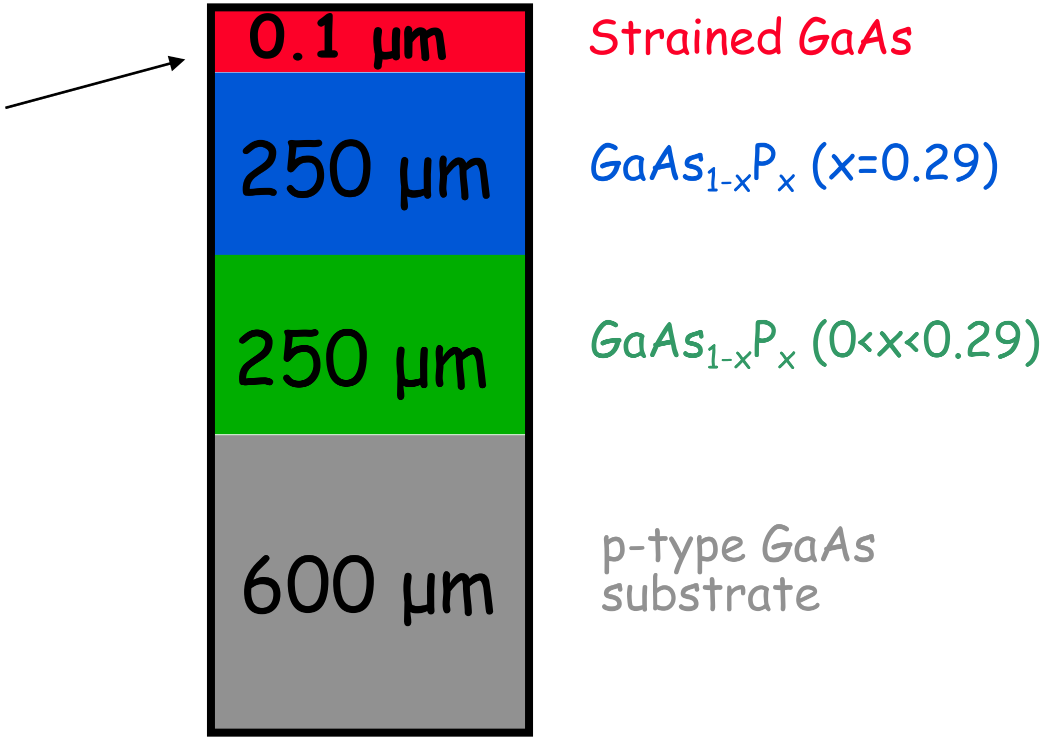


Image from Pablo Saez, PhD Thesis, Stanford University, SLAC Report 501, 1997

Strained layer GaAs





0.1 µm — Strained GaAs

250 µm — $GaAs_{1-x}P_x$ (x=0.29)

250 µm — $GaAs_{1-x}P_x$ (0<x<0.29)

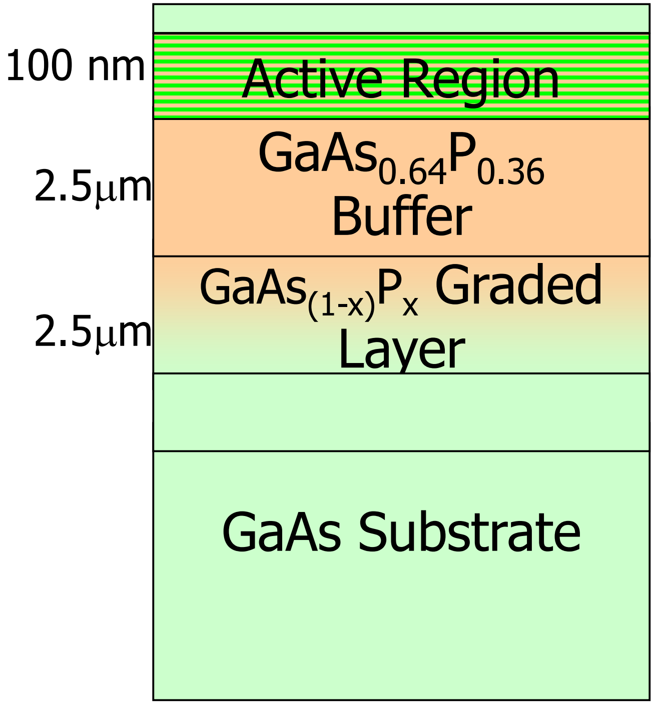
600 µm — p-type GaAs substrate

- ✓ Polarization 75% >> 50% ☺
- ✓ Strain relaxes in 100 nm layer
- ✓ QE 0.1%

MOCVD-grown epitaxial spin-polarizer wafer

Maruyama et al., Phys. Rev. B, **46** 4261 (1991)

Strained layer superlattice GaAs/GaAsP

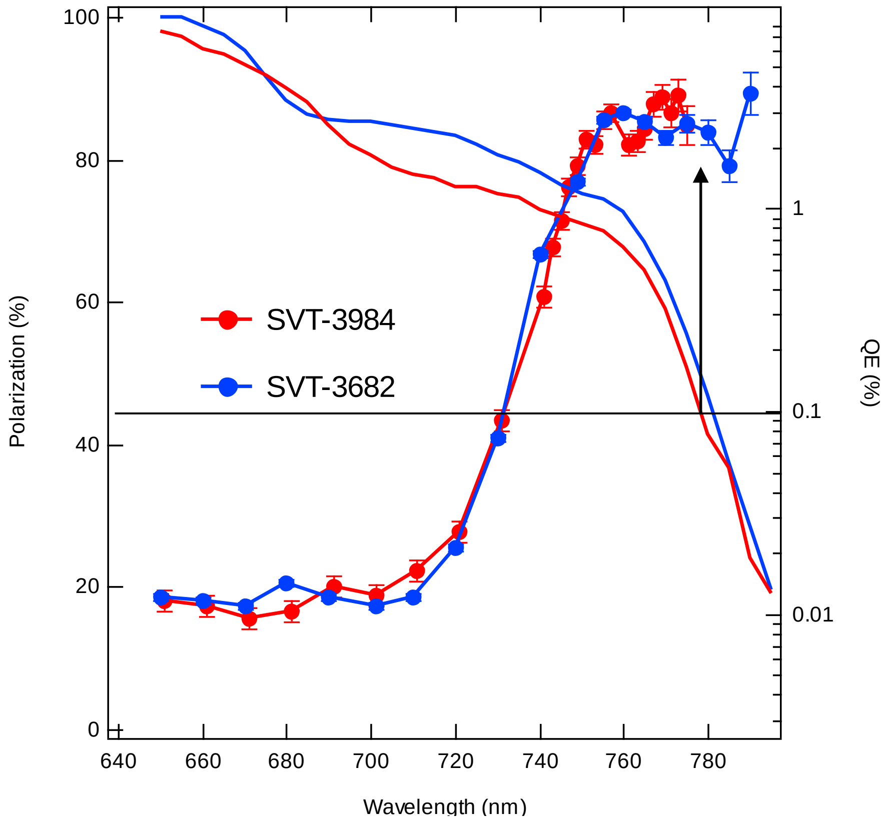


100 nm Active Region

2.5μm $GaAs_{0.64}P_{0.36}$ Buffer

2.5μm $GaAs_{(1-x)}P_x$ Graded Layer

GaAs Substrate

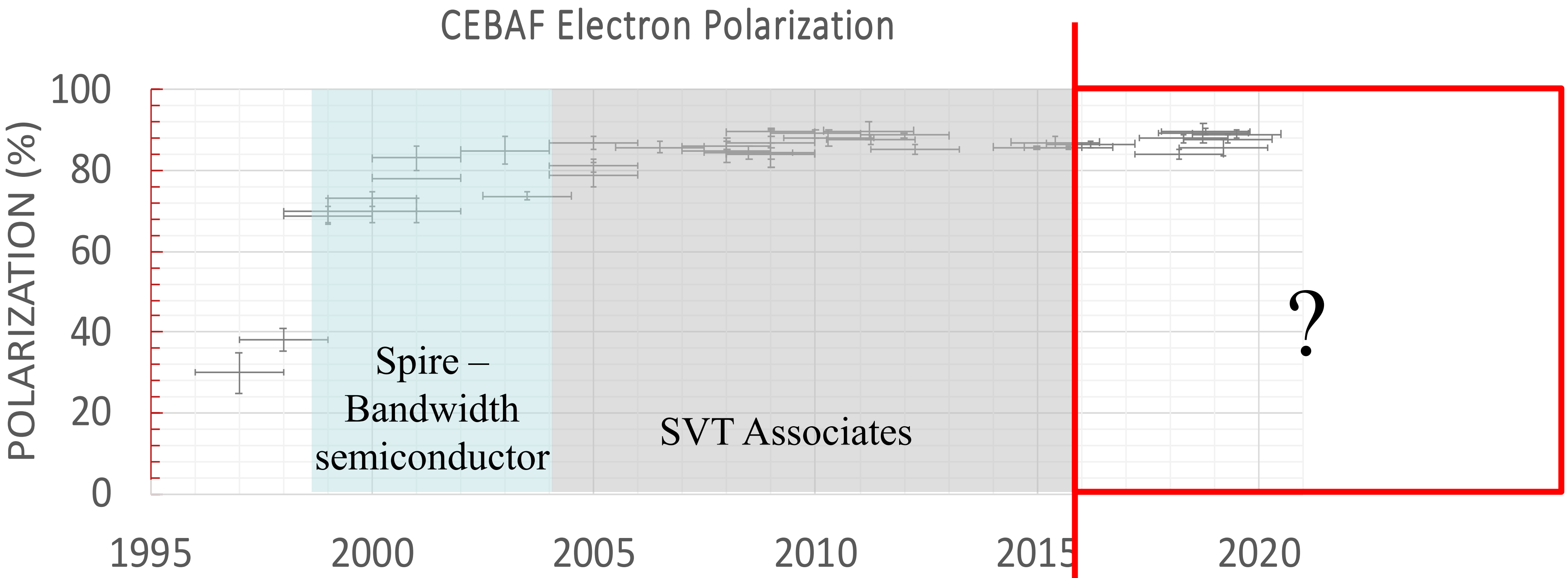


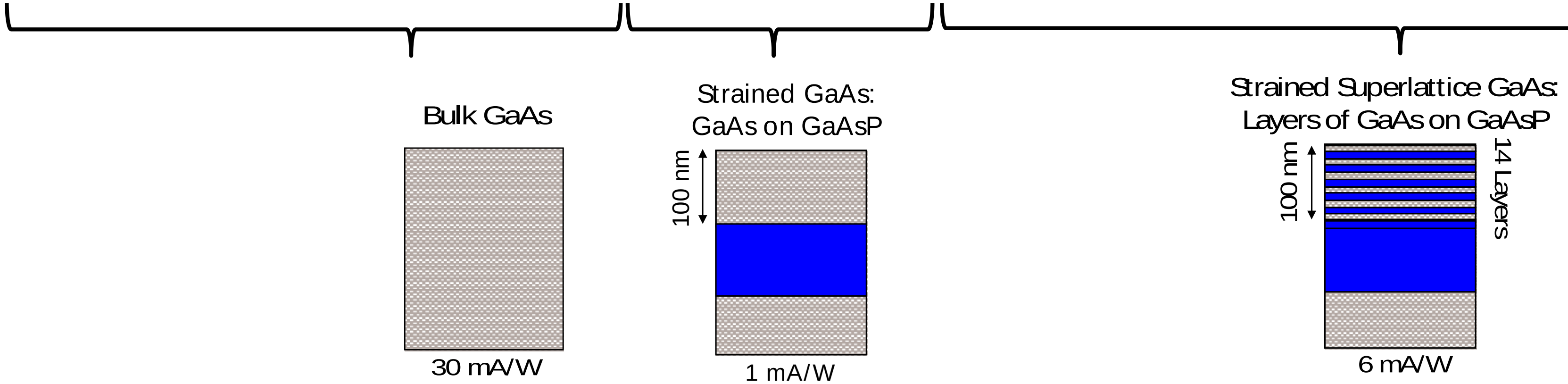
D. Luh et al, SLAC, PESP2002

QE 1% and Polarization 85%

From Aaron Moy, SVT Assoc and SLAC, PESP2002

JLab polarization : since 1990’s





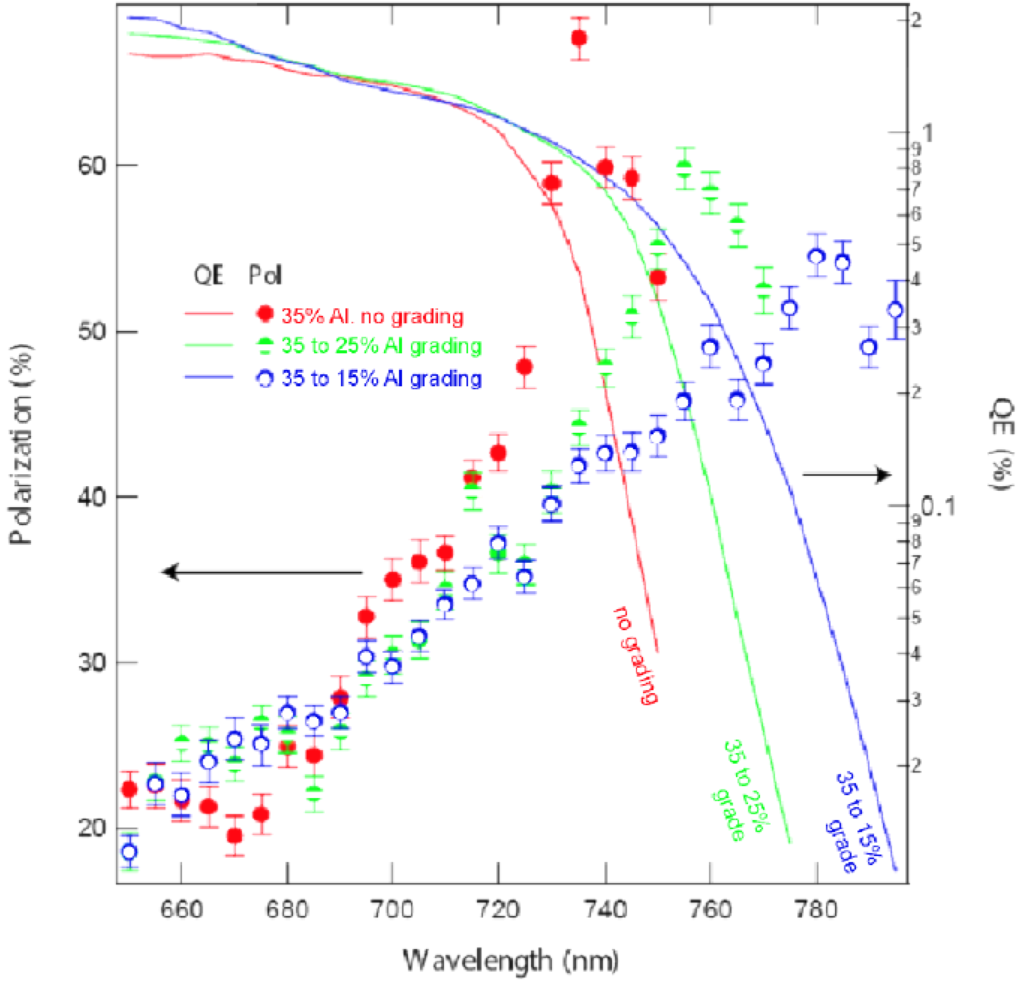
2016: Last SSL Photocathode received at JLab from SVT

Innovation through SBIR program

- SVT SBIR Partnerships with SLAC or JLab for high polarization photocathodes:
  - Phase 1: 2001, 2005, 2007, 2012, 2013
  - Phase II: 2002, 2008, 2013, 2014
- Various Superlattice Structures
  - **GaAs/GaAsP**
  - GaAsSb
  - AlGaAs/GaAs
  - *Distributed Bragg Reflector*

Variations

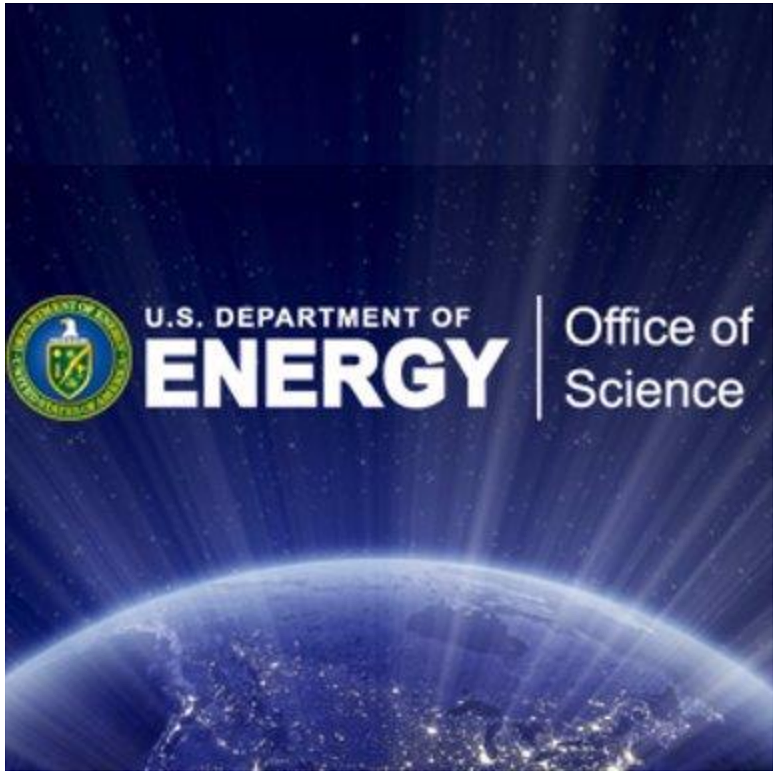
- Quantum Well thickness
- Barrier thickness
- Strain layer concentration
- Number of periods



AlGaAs/GaAs, A. Moy 2009

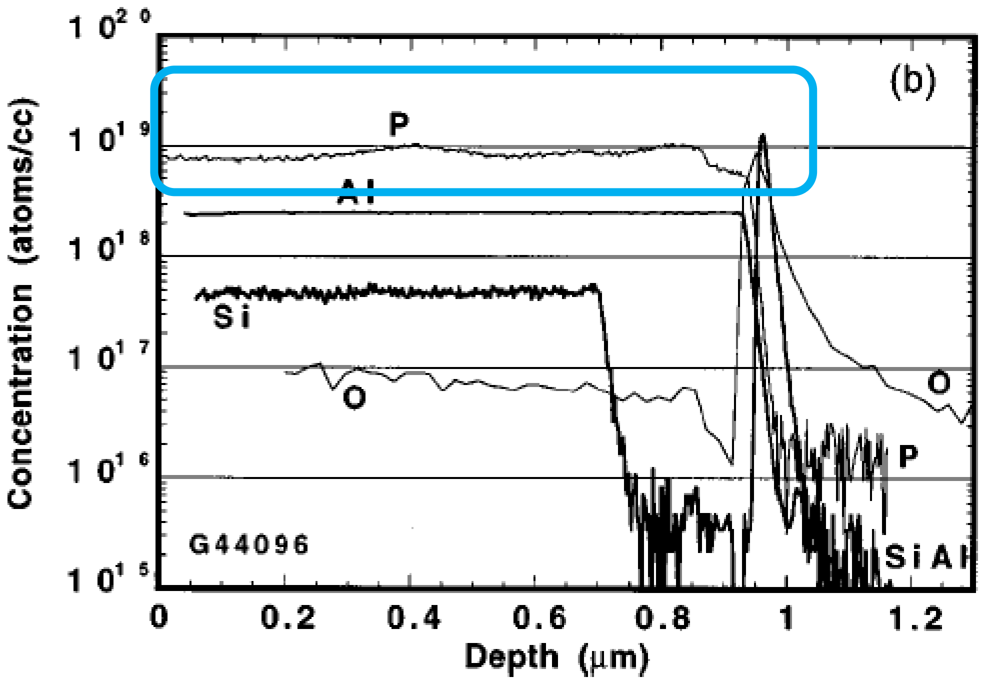
SBIR research program lifetime

- SBIR Program Goals include
  - Stimulate technological innovation
  - **Use small business to meet Federal R/R&D needs**
- SBIR Phases
  - Phase I explores the feasibility of innovative concepts with awards up to $250,000 and 12 months.
  - Phase II is the principal R&D effort, with awards up to $1,600,000 and 2 years.
  - *Phase III: pursue commercial applications of their R&D with non-SBIR/STTR funding.*
    - Market for high polarization photocathode material is small
    - Commercialization not financially viable



Technical Challenges of Growing GaAs/GaAsP using GSMBE

- GSMBE (Gas source MBE) uses crackers for AsH_3 (arsine) and PH_3 (phosphine)
  - Both gasses Toxic, Flammable
  - Phosphorus grows on MBE walls
    - Generates phosphine gas & phosphoric acid when venting
    - Absorbs water and has high water vapor pressure when pumped back down
    - Residue cannot be scraped off - ignites
      - Careful degassing can solve this
  - Phosphine residue can cause high background in subsequent samples



SIMS of AlGaAs grown after Phosphorus contamination

W.E.Hoke and P.J. Lemonias JVSTB **17** 1999, p. 2009.

Efforts to restore supply

- DOE Funding Opportunity 20-2310
  - MOCVD (*metal organic chemical vapor deposition*)
    - JLab: M. Poelker and M. Stutzman
    - BNL: E. Wang
    - ODU: S. Marsillac, B. Belfore
  - CBE (Chemical Beam Epitaxy)
    - JLab: M. Stutzman
    - UCSB: C. Palmstrøm, A. Engel
- MBE SSL GaAs/GaAsP Distributed Bragg Reflector
  - Sandia National Lab: Center for Integrated Nanotechnology
    - BNL: L. Cultrera
- Acken Optoelectronics Ltd., Suzhou China
  - Yiqiao Chen, formerly of SVT Associates
  - SSL GaAs/GaAsP photocathodes on order for evaluation

MBE, GSMBE, CBE and MOCVD

MBE

Gas Source Molecular Beam Epitaxy

elemental As, P, Ga

- Pressure ~10^{-8} mbar
- Growth rates ~ 1 µm/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

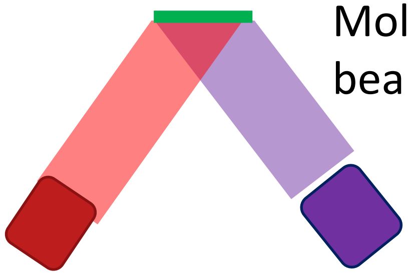
- Pressure <10^{-4} mbar
- Growth rates 0.5-1 µm/hr

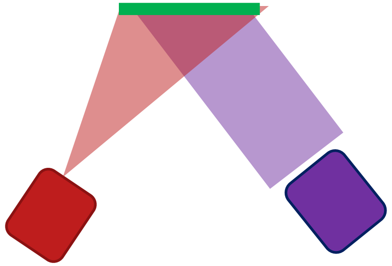
MOCVD

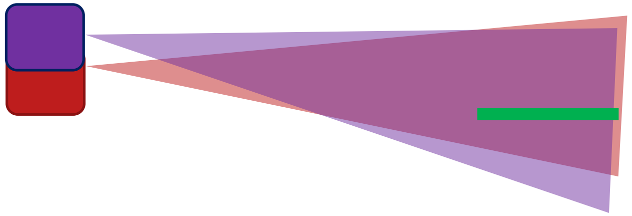
Metal organic chemical vapor deposition

AsH_3, PH_3, trimethylgallium (TMGa)

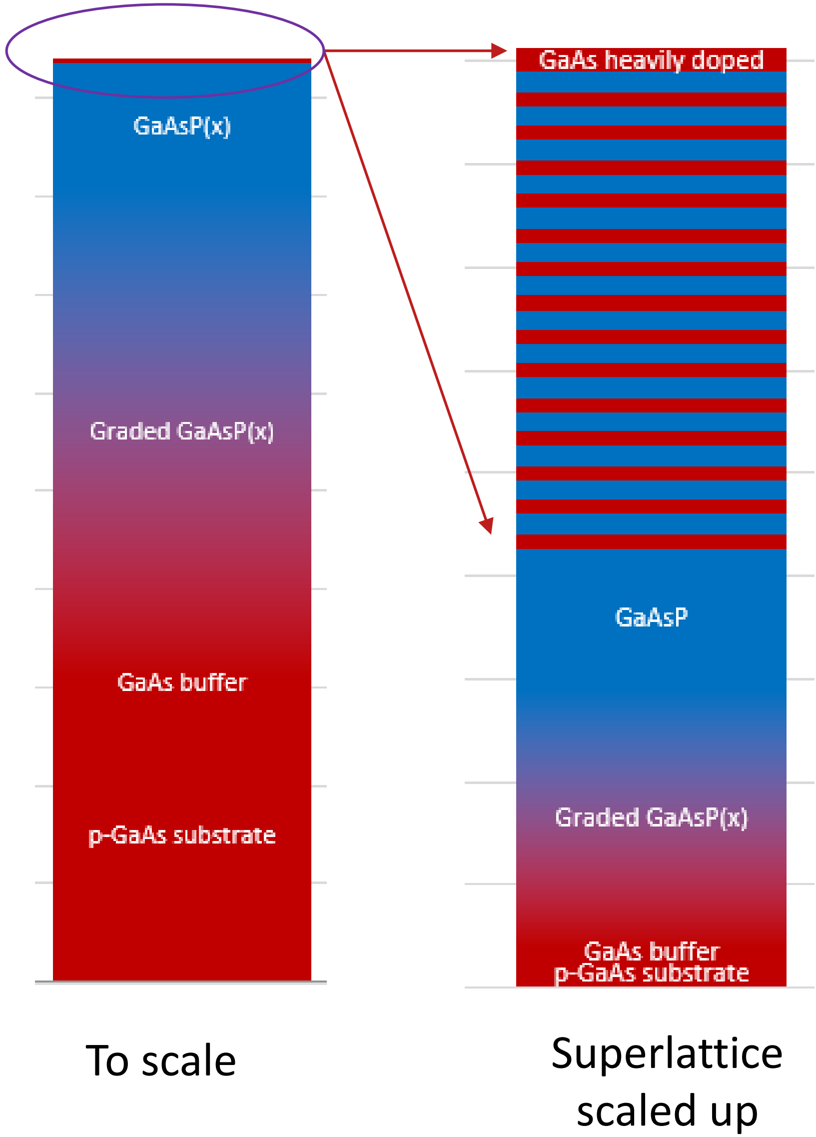
- Pressures >100 mbar during growth
- Growth Rates 10 µm/hr
- Traditionally difficult to get sharp interfaces







Wafer growth steps



- Epitaxial Buffer Layer grown on GaAs
- Graded GaAs to $GaAs_{(1-x)}P_x$
- $GaAs_{(1-x)}P_x$ layer
- Superlattice
- Heavily doped top layer

Parameters to vary

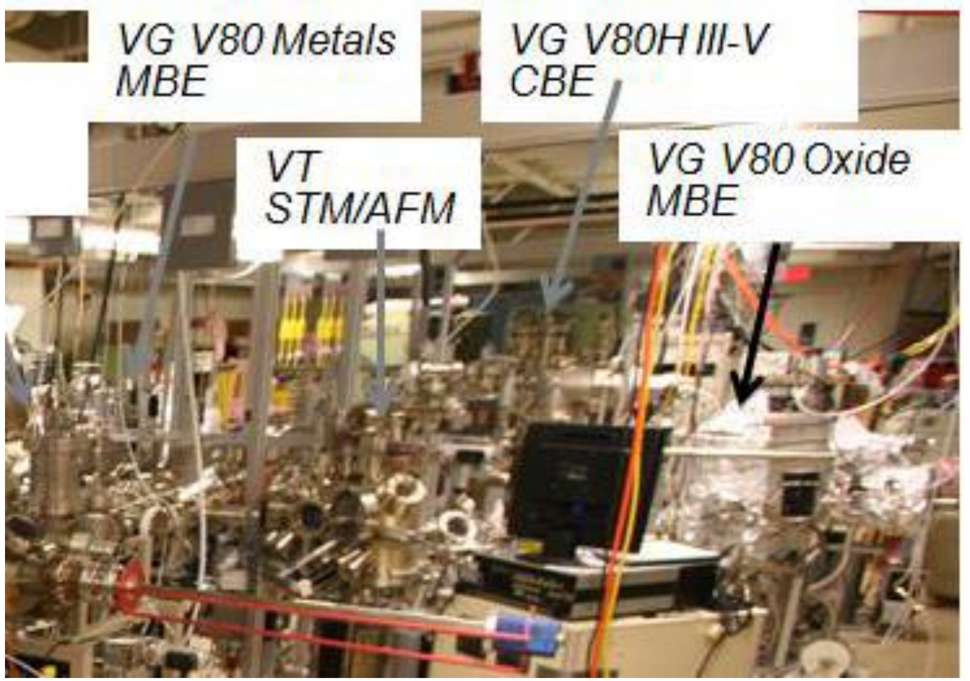
- Substrate Temperature
- Source Temperature/Pressures
- Time
- Grading profile
- Underlying crystal orientation

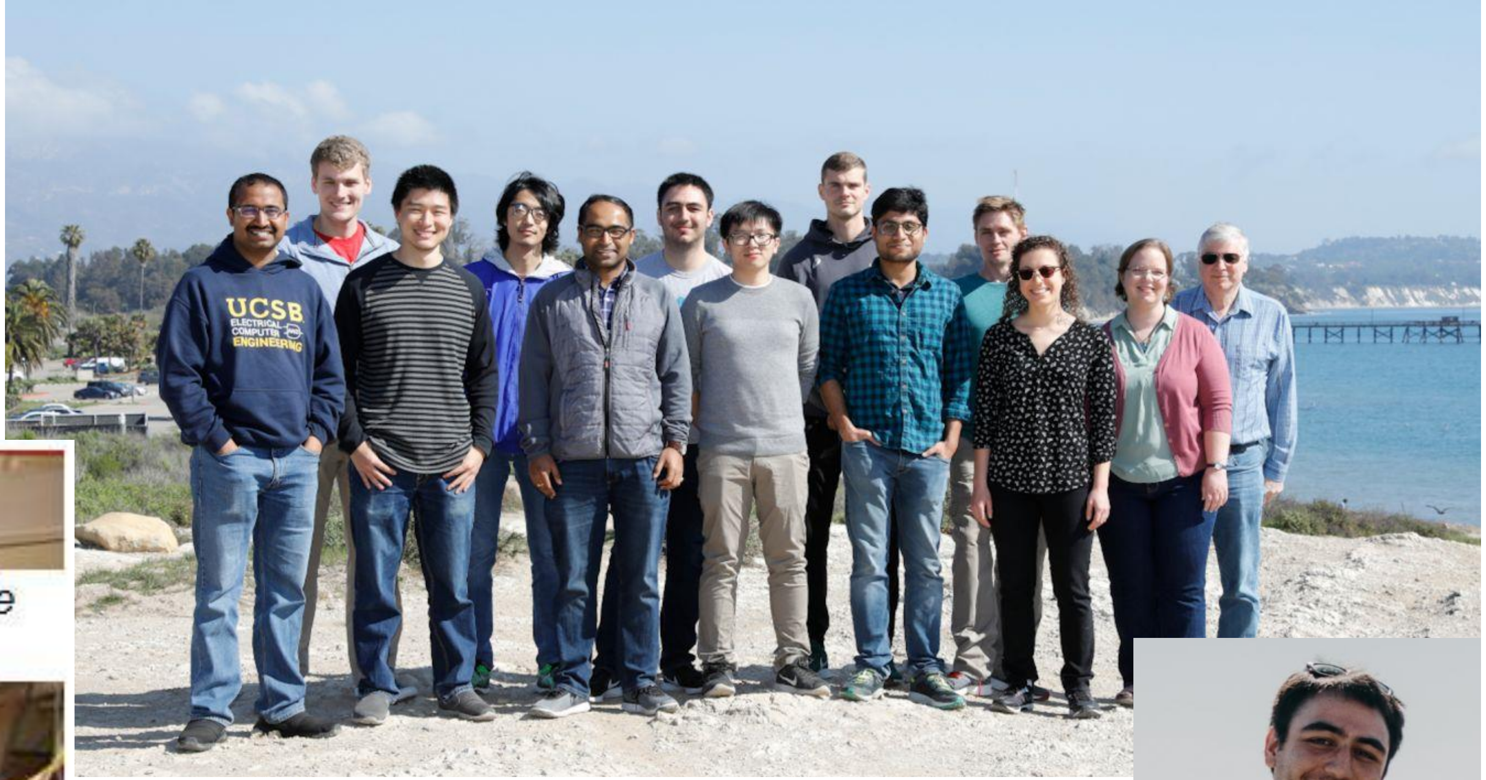
CBE: Photocathode progress

Chris Palmstrøm Group, UCSB

- Aaron Engel, graduate student

- Chemical Beam Epitaxy System

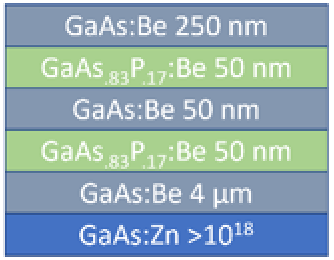


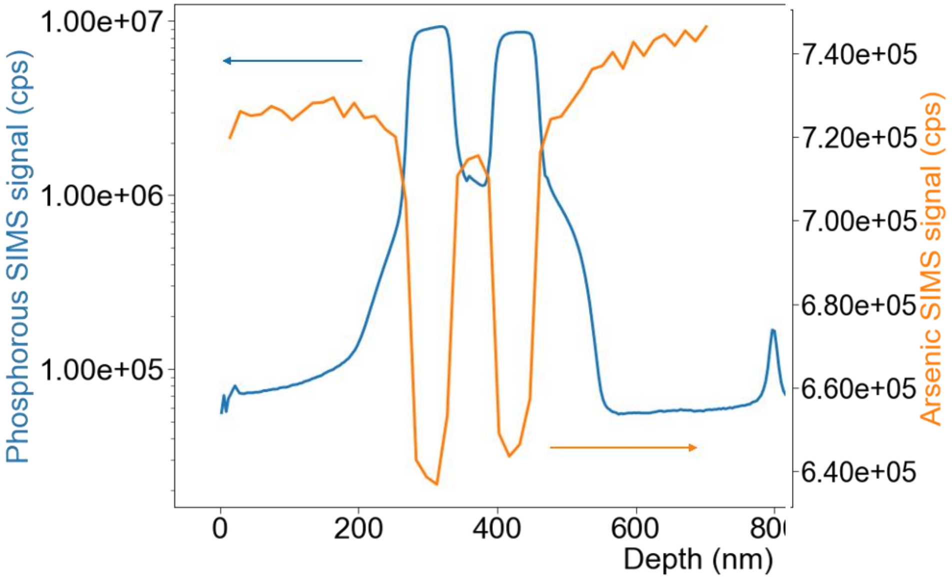


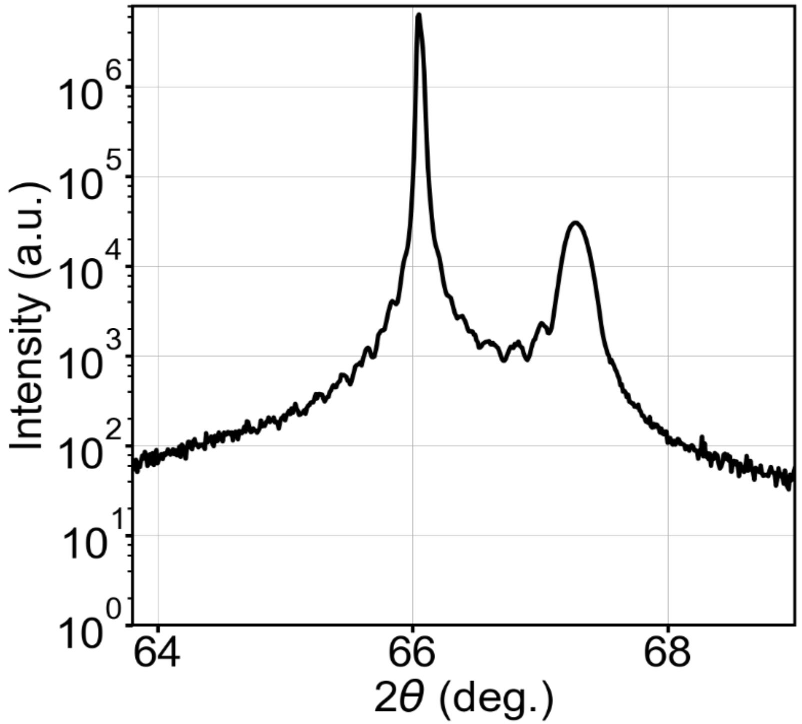
CBE: Photocathode progress

- Computerized control developed for GaAs->GaAsP graded layer
- Interface quality between GaAs and GaAsP measured
    - SIMS analysis, x-ray diffraction
- Sample temperature, gas flux optimized for proper stoichiometry
- Testing strained GaAsP on GaAs initially

SIMS:
Composition
vs. Depth


GaAs:Be 250 nm
$GaAs_{.83}P_{.17}$:Be 50 nm
GaAs:Be 50 nm
$GaAs_{.83}P_{.17}$:Be 50 nm
GaAs:Be 4 µm
GaAs:Zn >10^{18}

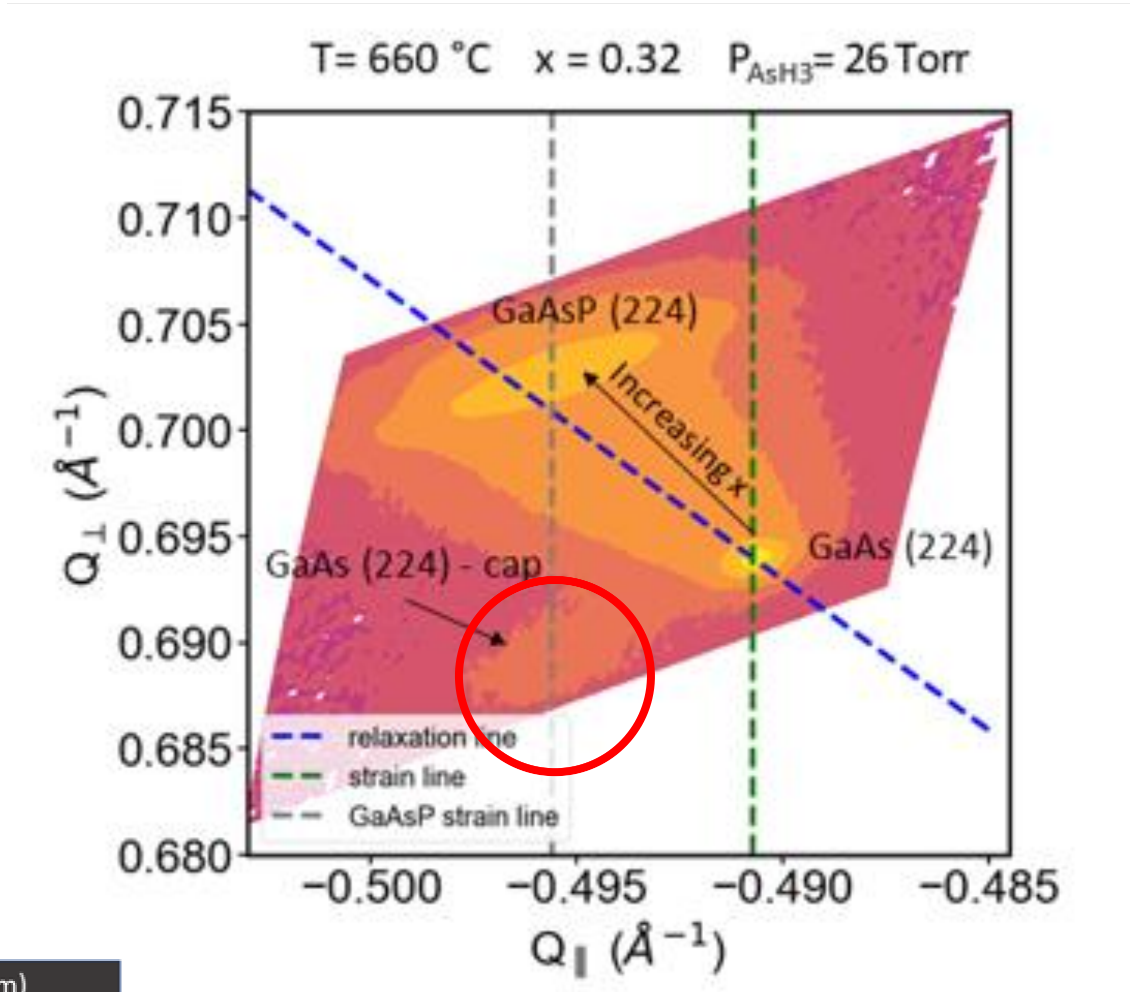




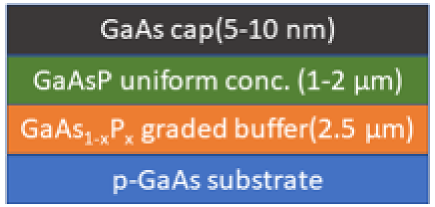
x-ray diffraction:
1 micron $GaAs_{0.75}P_{0.25}$ on GaAs
Testing analysis tools for interfaces

CBE: Photocathode progress

- Computerized control developed for GaAs->GaAsP graded layer
- Interface quality between GaAs and GaAsP measured
  - SIMS analysis,
- Sample temperature, gas flux optimized for proper stoichiometry
- 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



X-ray reciprocal space map for single 5-10 nm GaAs layer on GaAsPx



GaAs cap(5-10 nm)

GaAsP uniform conc. (1-2 μm)

$GaAs_{1-x}P_x$ graded buffer(2.5 μm)

p-GaAs substrate

CBE: Photocathode progress

Next Steps

- Triethylgallium and phosphine create high vapor pressure background
  - Move to elemental Ga source?
  - Upgrade sample bonding from indium to gallium
- Grow photocathode material to test & test at JLab

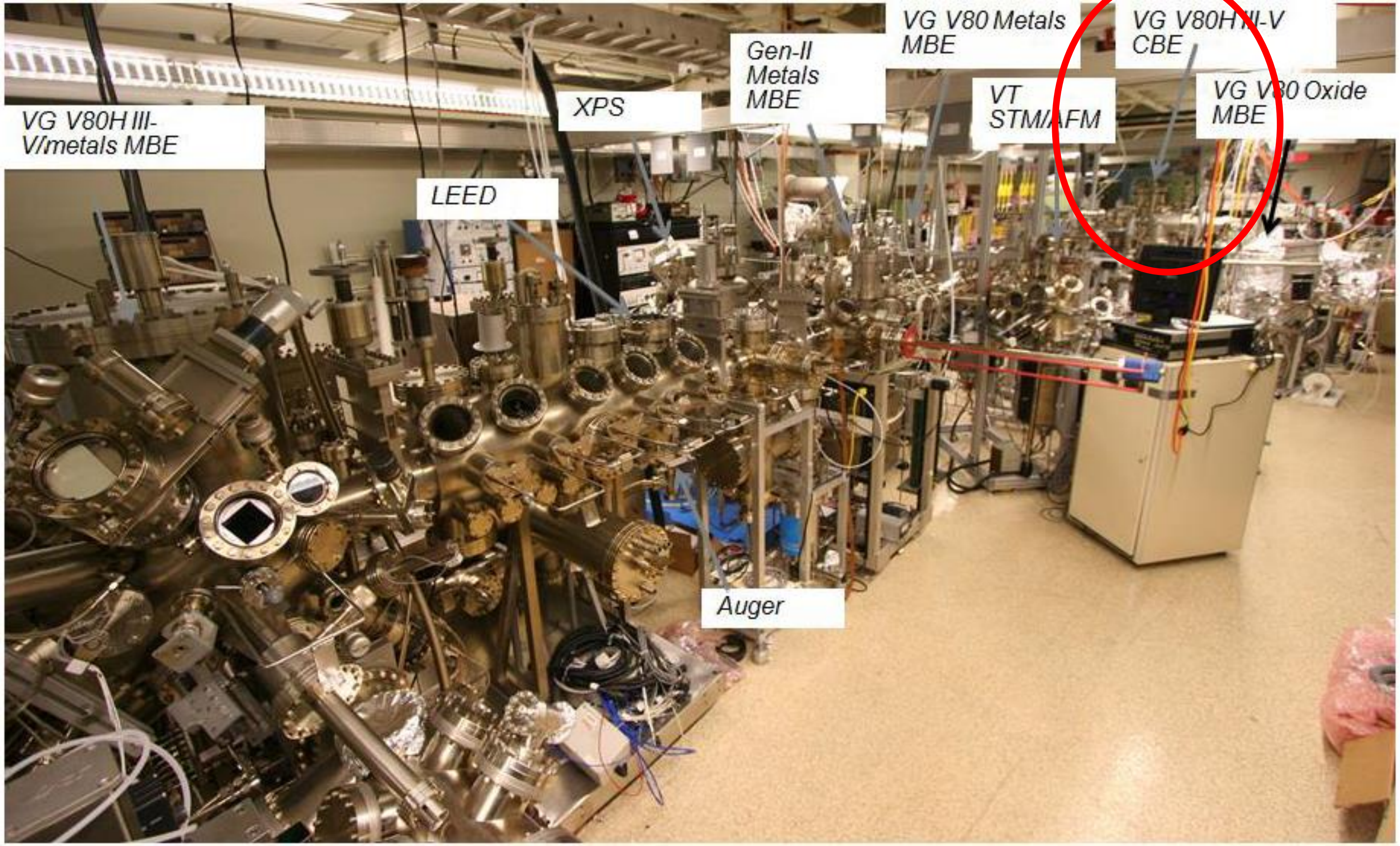


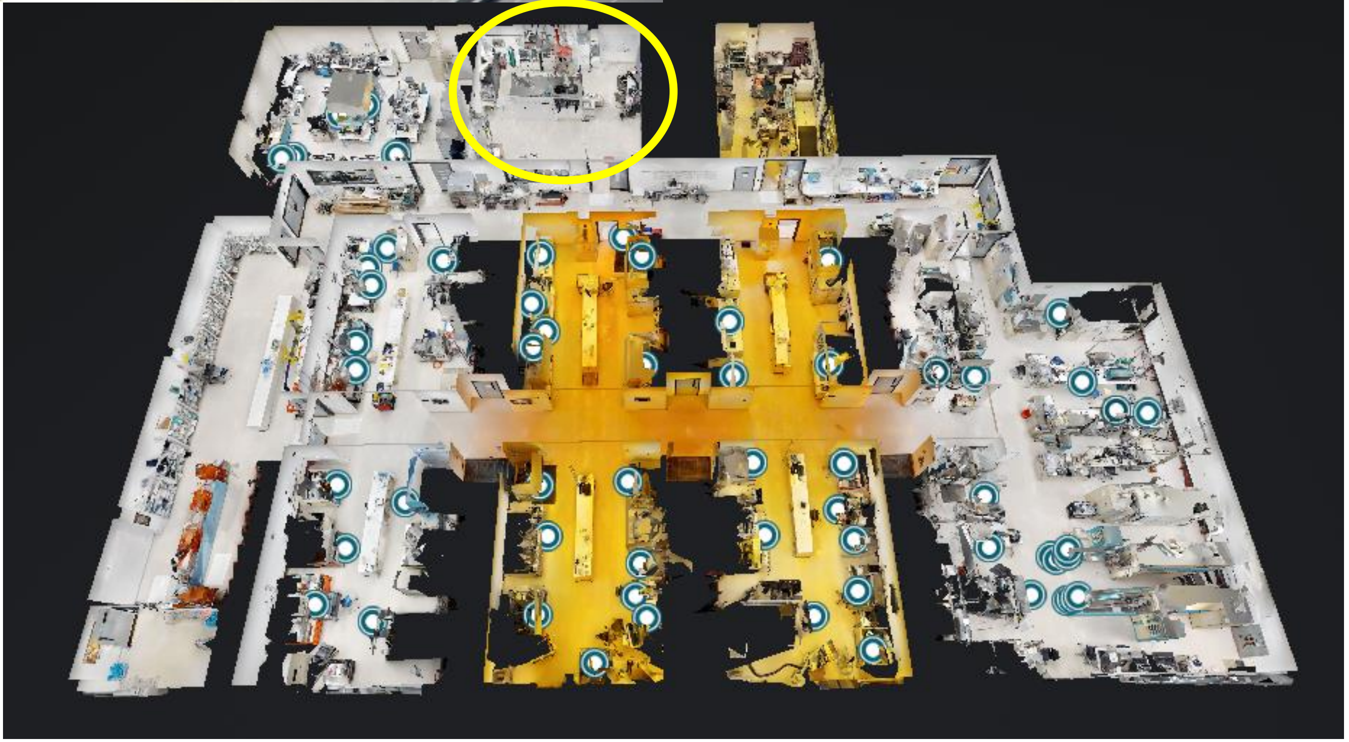
Figure 2 Semiconductor deposition system at Chris Palmstrom's lab at UCSB. The CBE system for the growth of this material is shown at the back and labelled "VG V80H III-V CBE".

MOCVD: Photocathode progress

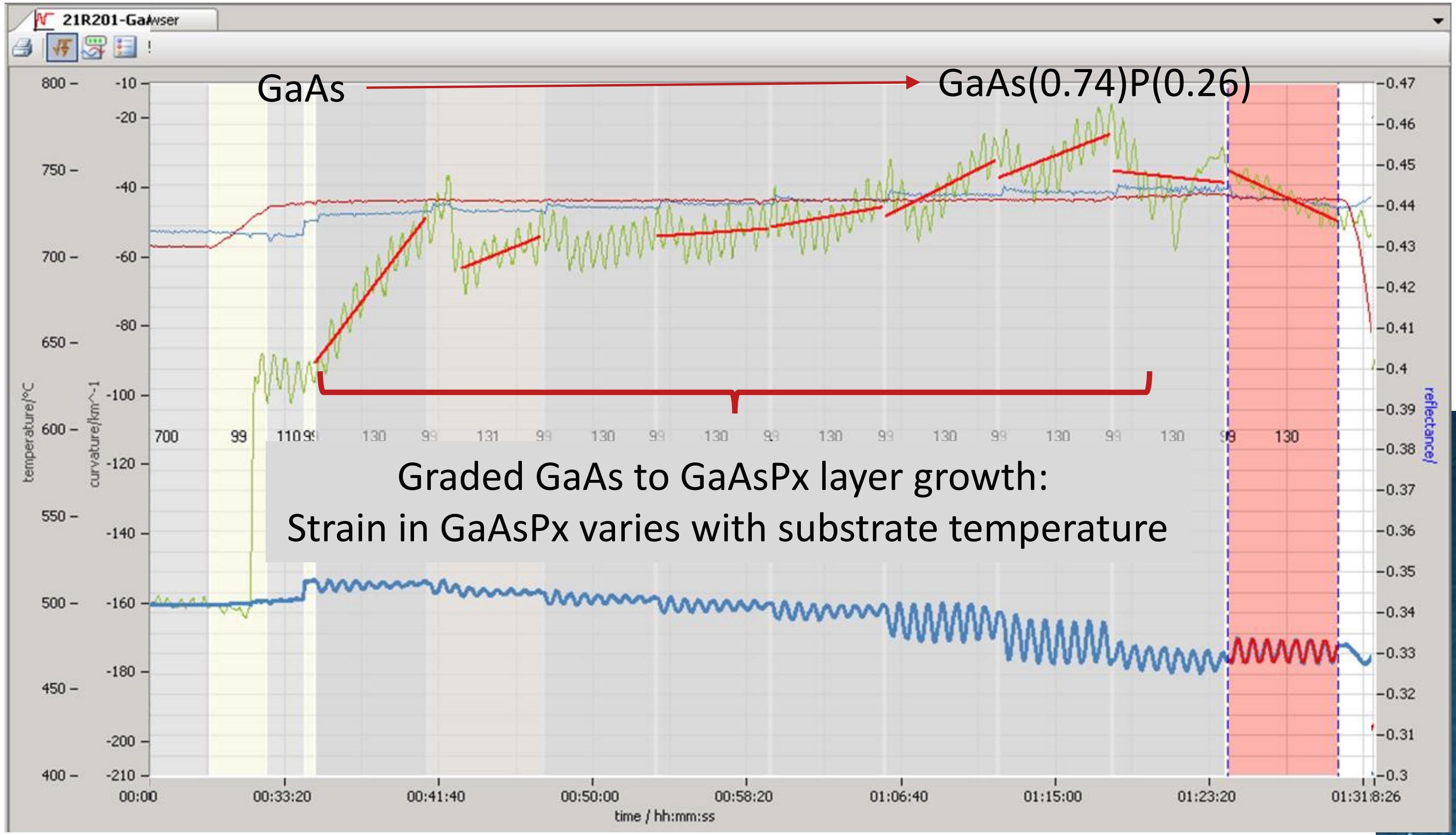
The Rochester Institute
of Technology
III-V EPICenter

Dr. Sylvain Marsillac,
Old Dominion University

Ben Belfore
ODU Graduate
Student

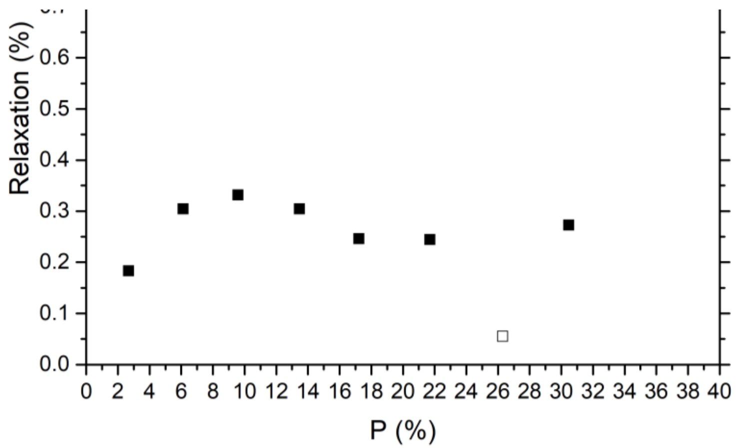


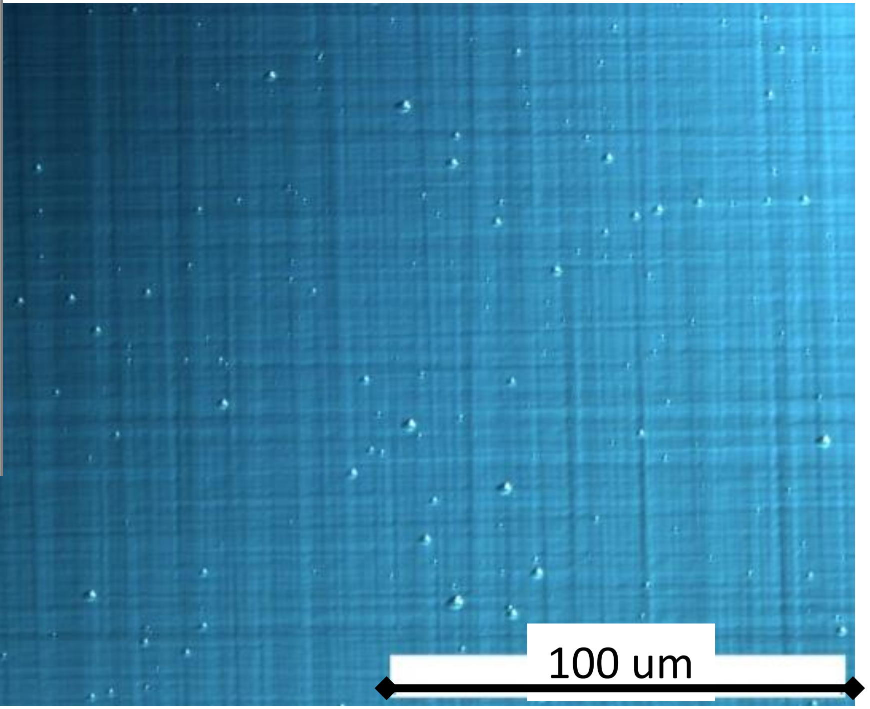
Results: MOCVD



GaAs → GaAs(0.74)P(0.26)

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





Higher temperatures yield improved surface with moderate relaxation throughout

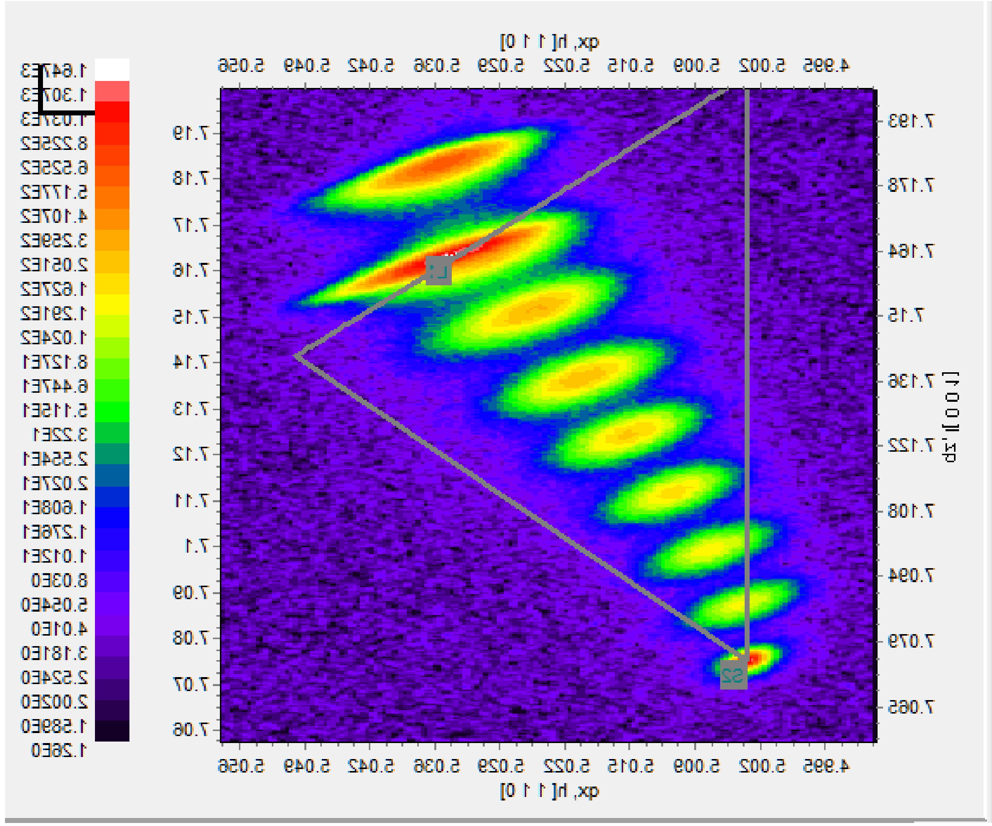
730°C growth temperature

Optimizing temperatures, graded layer profile

Results: MOCVD photocathode progress

- Graded layer “metamorphic” test runs
  - —Optimizing parameters for highest relaxation
  - —Hall effect measurements for dopant characterization
- Superlattice runs
  - —Growing superlattice on each metamorphic run
  - —Optimizing parameters with zinc dopant
- Characterization
  - —Surface analysis (SIMS, TEM) planned
  - —**Ready for first polarization measurements**

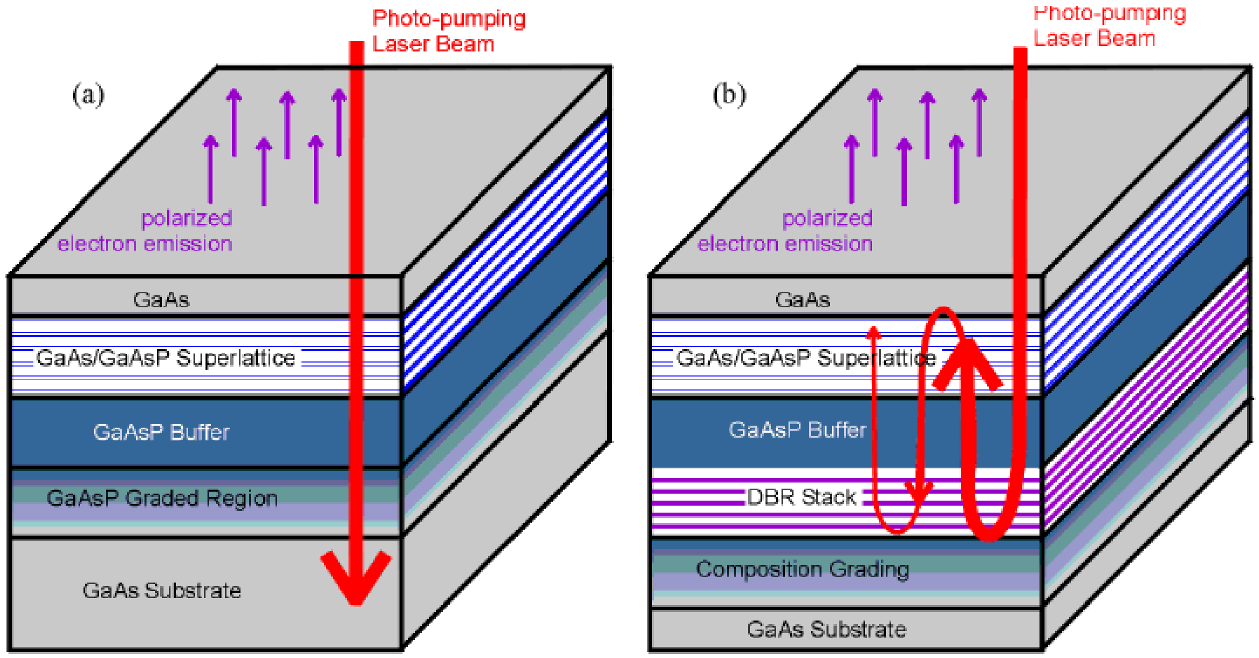
  - —JLab: MicroMott Polarimeter
  - —BNL: Specs Mott Polarimeter
    - Operational, testing various samples



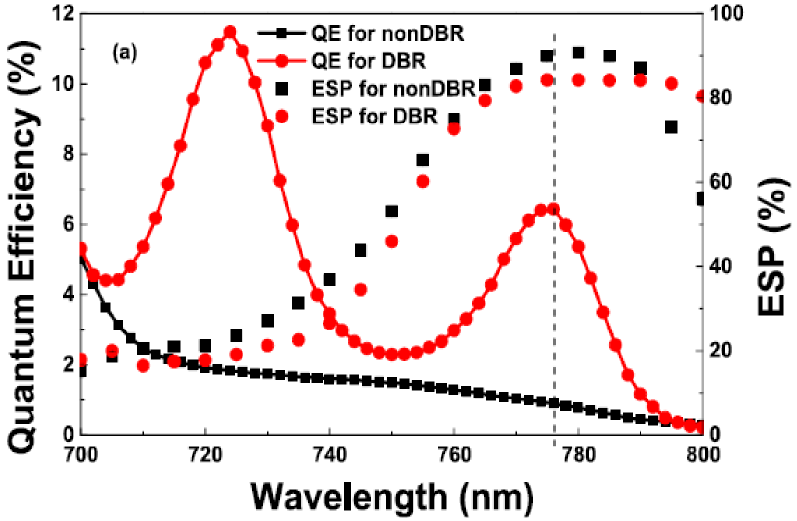
X-ray reciprocal space mapping

Crystal growth by Ben Belfore under the supervision of Sylvian Marsillac, Old Dominion University
MOCVD system at Rochester Institute of Technology

Distributed Bragg Reflector



SVT DBR:
Polarization >80%
QE 6% (6x typical)



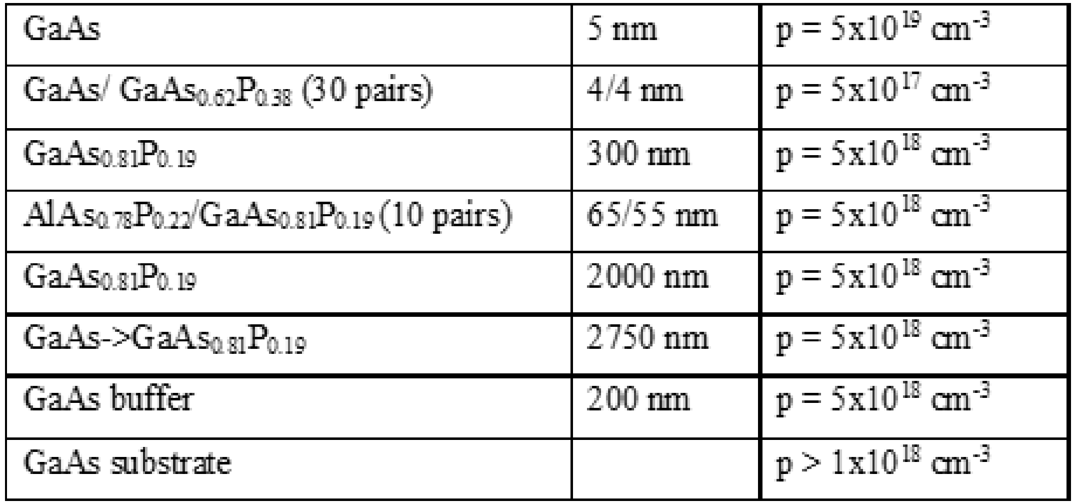
DBR structure has Fabry Perot structure

- Thickness tuned to desired wavelength
- Multiple passes -> more excited photons
- Polarization of light preserved

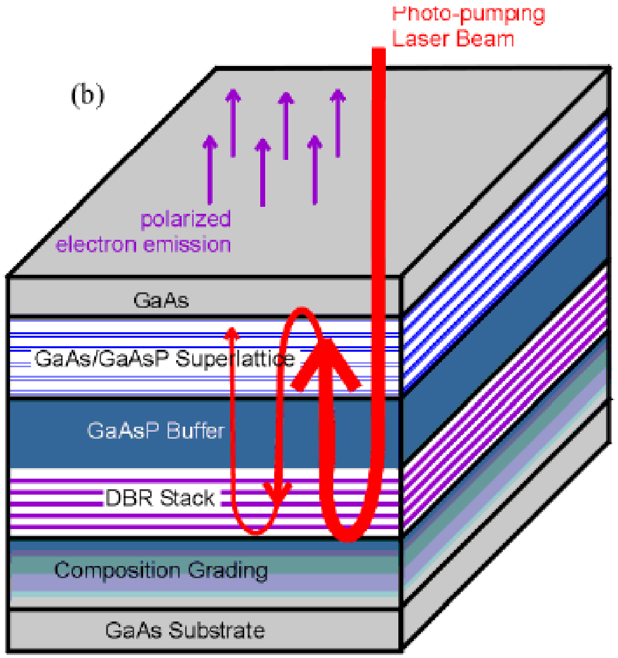
Wei Liu, Yiqiao Chen, Wentao Lu, Aaron Moy, Matt Poelker, Marcy Stutzman, and Shukui Zhang, "*Record-level quantum efficiency from a high polarization strained GaAs/GaAsP superlattice photocathode with distributed Bragg reflector*", Appl. Phys. Lett. **109**, 252104 (2016).

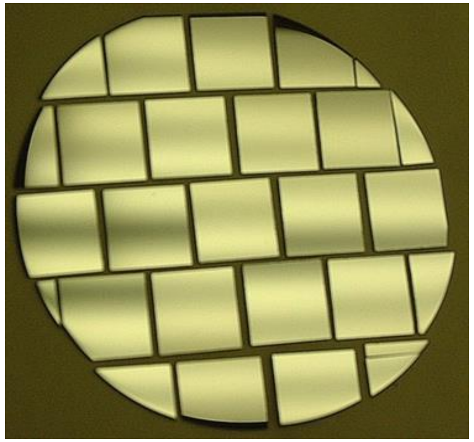
Results: MBE DBR photocathode progress

- BNL: Luca Cultrera
    - Funding: Center for Integrated Nanotechnology
- Sandia National Lab
    - Several MBE growth systems
    - 3” wafers
- 240 nm Superlattice (vs. ~100 nm)
- First & second samples complete



GaAs	5 nm	p = 5×10^{19} cm^{-3}
GaAs/ GaAs$_{0.62}$P$_{0.38}$ (30 pairs)	4/4 nm	p = 5×10^{17} cm^{-3}
GaAs$_{0.81}$P$_{0.19}$	300 nm	p = 5×10^{18} cm^{-3}
AlAs$_{0.78}$P$_{0.22}$/GaAs$_{0.81}$P$_{0.19}$ (10 pairs)	65/55 nm	p = 5×10^{18} cm^{-3}
GaAs$_{0.81}$P$_{0.19}$	2000 nm	p = 5×10^{18} cm^{-3}
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}





Representative diced GaAs wafer

BNL Mott polarimeter measurements

- Good QE (>1%)
- Polarization ~80%

Issues: non-uniformity across photocathode

Future: Optimize and test

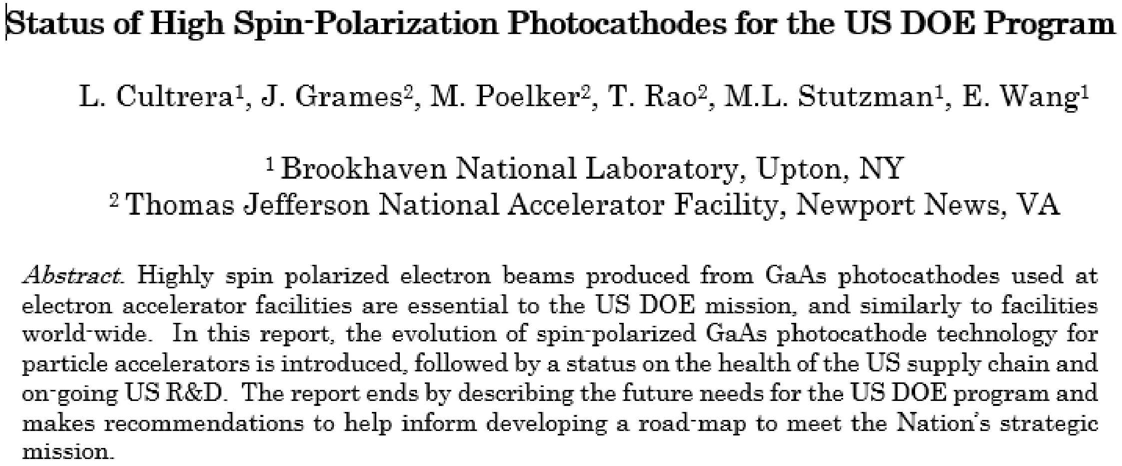
Where do we go from here?

Recent Whitepaper sent to David Asner, BNL

Recommendations

- Short Term
  - We don’t have a domestic supplier
  - Both CEBAF at JLab and EIC will need high polarization photocathodes
  - Partnership with commercial vendors (such as SVT) or DOE labs (Sandia) could restore supply

    ** Acken Optoelectronics Ltd., Suzhou China*
- Longer Term
  - Useful to explore different technologies like MOCVD or CBE: university partnerships
  - Continue research: Novel materials, structures, activations
  - Future machines may have different demands



Status of High Spin-Polarization Photocathodes for the US DOE Program

L. Cultrera[1], J. Grames[2], M. Poelker[2], T. Rao[2], M.L. Stutzman[1], E. Wang[1]

[1] Brookhaven National Laboratory, Upton, NY
[2] Thomas Jefferson National Accelerator Facility, Newport News, VA

Abstract. Highly spin polarized electron beams produced from GaAs photocathodes used at electron accelerator facilities are essential to the US DOE mission, and similarly to facilities world-wide. In this report, the evolution of spin-polarized GaAs photocathode technology for particle accelerators is introduced, followed by a status on the health of the US supply chain and on-going US R&D. The report ends by describing the future needs for the US DOE program and makes recommendations to help inform developing a road-map to meet the Nation’s strategic mission.

Conclusions

- Two university research partnerships in progress: MOCVD and CBE
- Sandia Laboratory MBE growth in progress: first samples tested
- Polarimeters and test beamlines available at JLab and BNL
  - JLab: Acken SSL on order
  - BNL tests: Sandia SSL DBR, Nagoya SSL, SVT SSL samples

Jefferson Lab
Center for
Injectors and
Sources

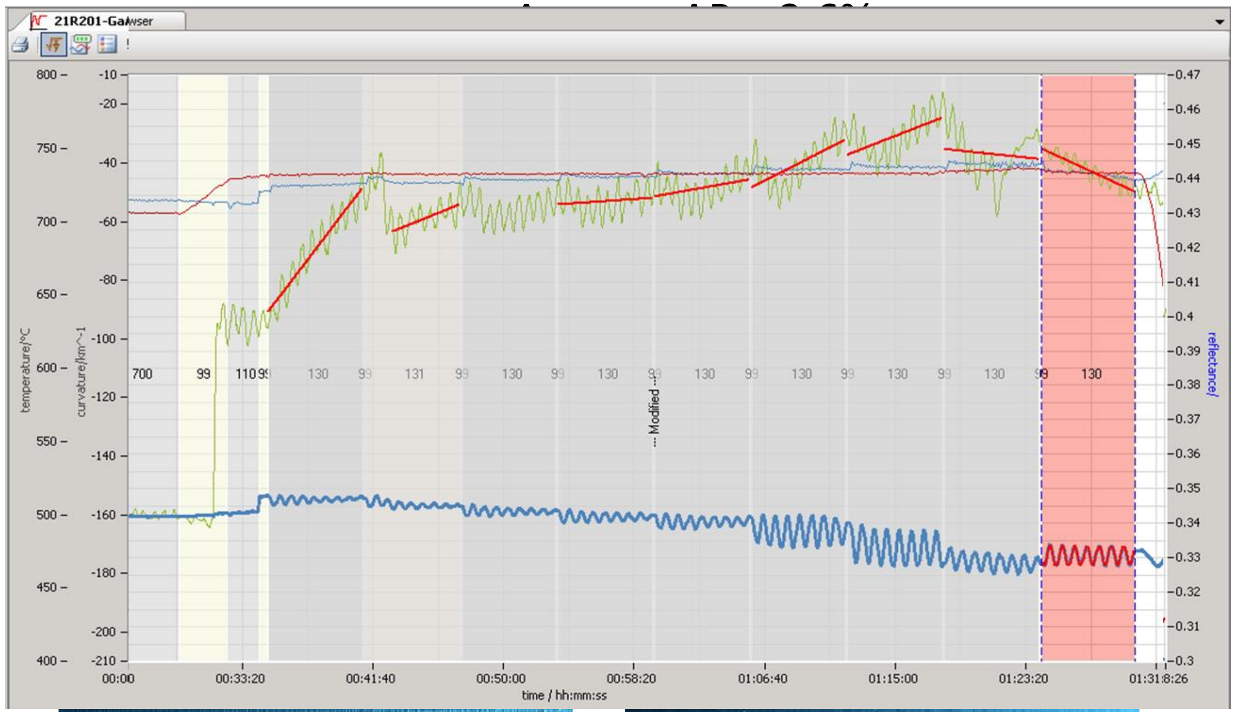
Questions?

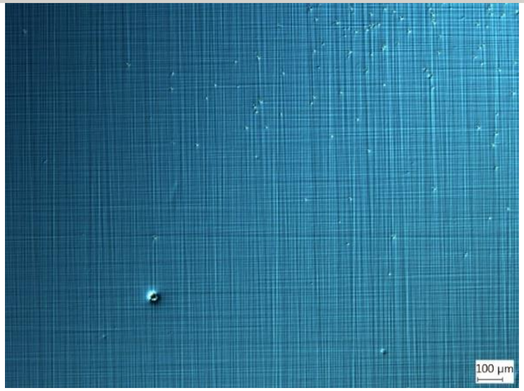
email: marcy@jlab.org

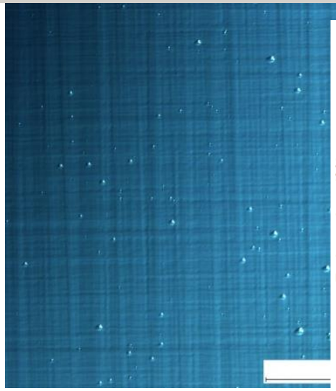
backup

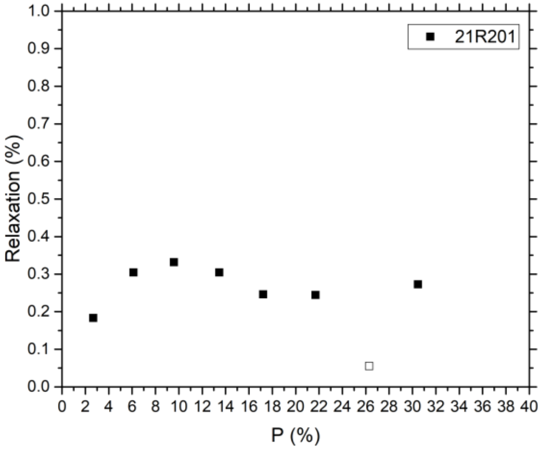
- ***Recommendations***
- A reliable source of highly spin-polarized photocathodes is essential to the success of the DOE Nuclear Physics programs at the CEBAF and EIC. We recommend a strategy with both Short Term and Long Term components to develop a reliable and healthy US supply chain.
-
- ***Highest Recommendation – Short Term (immediate)***
- Our highest recommendation is to immediately support a competent and capable industrial partner (like SVT Associates) and/or a suitable DOE facility (like Sandia National Laboratories) to restore the reliable production of well-known superlattice GaAs/GaAsP photocathodes. This recommendation is meant to mitigate the risk to CEBAF operations or the timely development of the EIC at Brookhaven National Laboratory. Photocathodes may be stockpiled for future operation of CEBAF and EIC, however we urge a strategy which avoids future circumstances as exists today, with no US supply chain option. Relying on a single vendor/source is not ideal. If anything were to happen preventing the sole source of GaAs/GaAsP photocathodes to sustain production the chain of supply might break again.
-
- ***Long Term (sustained)***
- Multiple industries, national labs and academia exploring alternative fabrication methods should be supported to explore versatile growth techniques. Growth techniques which are less expensive than MBE to maintain (like MOCVD, CBE), or those which may yield improved performance over existing superlattice photocathodes should be encouraged and funded. This approach may attract more diverse industrial and university partners, resulting in a potential greater applications. An ideal consequence of a more diverse infrastructure is a healthier technology industry utilizing spin polarized photocathodes. Our proposed strategy is not meant to not constrain activities only to GaAs technology. Rather we strongly support R&D for alternative polarized electron materials or structures which may outperform and supersede GaAs technology. Coupled with the progress in laser and SRF technology, this long term development will be critical to meet requirements of future science programs which could have requirements far more demanding than imagined today.
-

MOCVD: Increase growth temperature







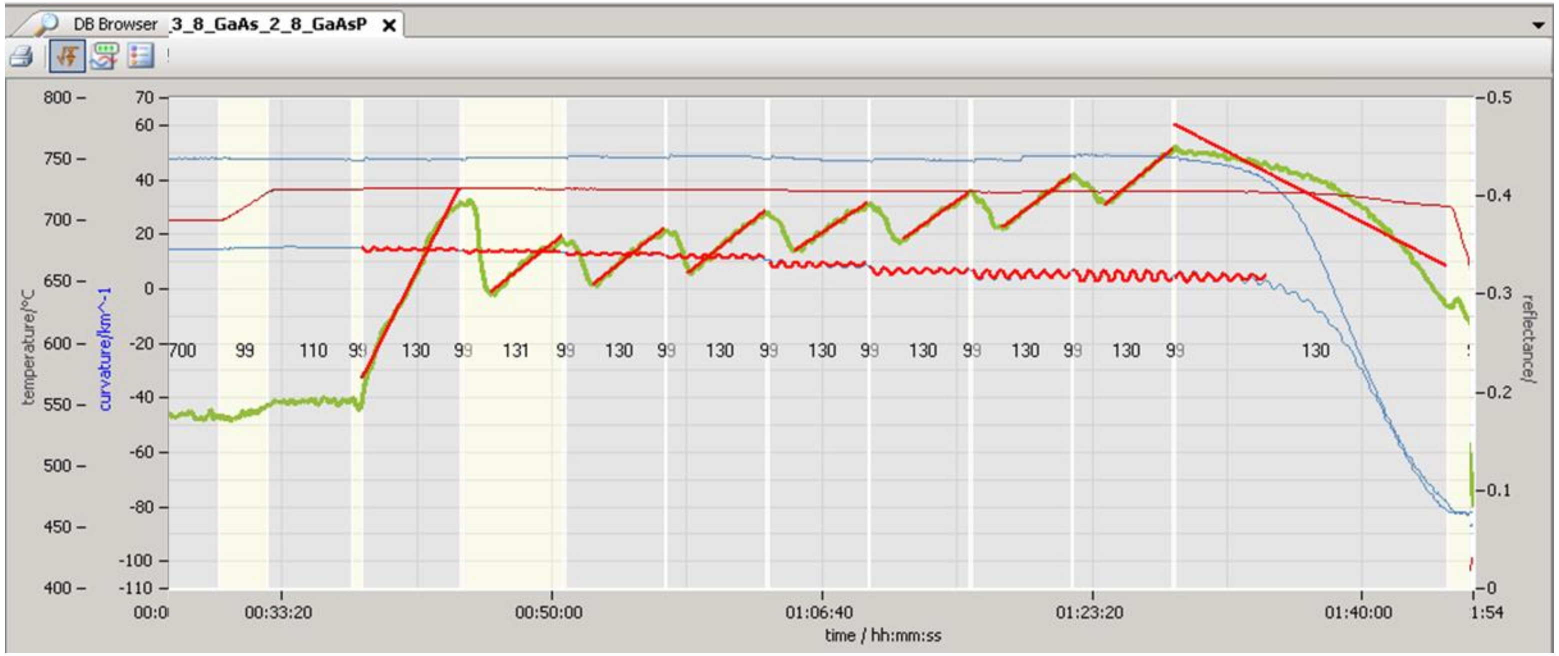


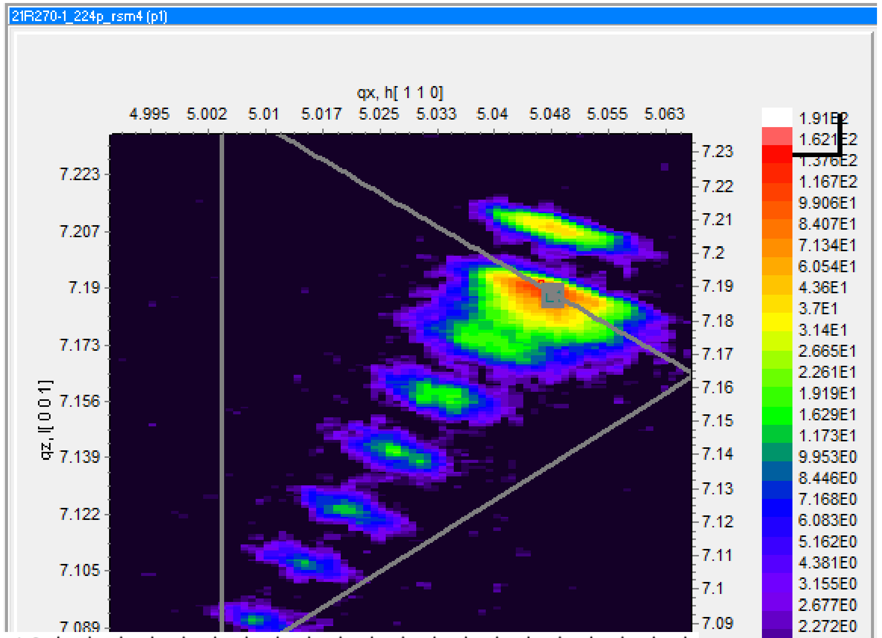
Ends at ~26 % P

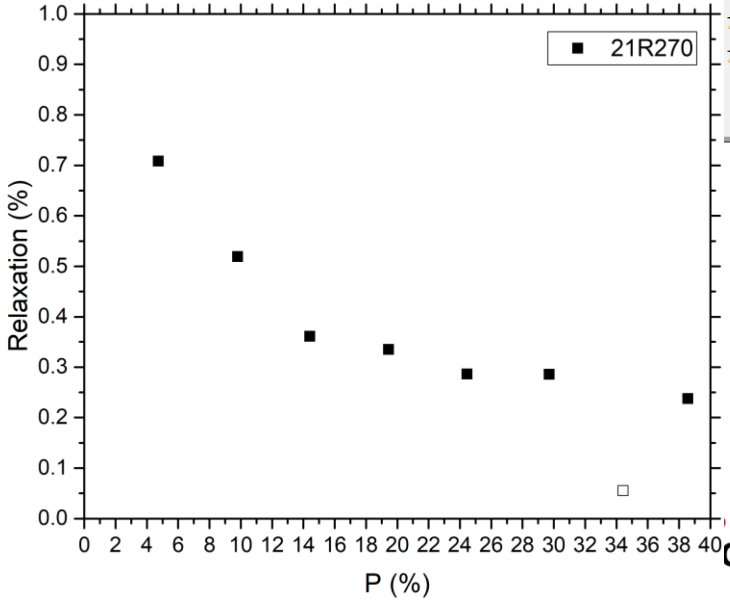
~10 µm/hr graded layer growth rate
V/III: 20-25
730°C growth temperature

Higher temperature has considerably improved surface, moderate relaxation throughout.

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