# Photocathodes for Polarized Electron Beams at Jefferson Lab

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**December 3, 2021**

Thomas Jefferson National Accelerator Facility is managed by
Jefferson Science Associates, LLC, for the U.S. Department of Energy's Office of Science

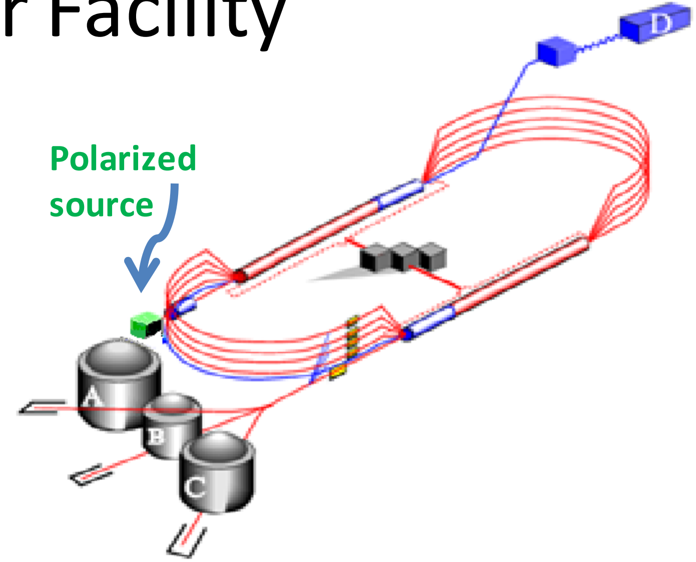
Jefferson Lab

# Outline

- Jefferson Lab, EIC and polarized electrons
- Photoemission from GaAs
- High polarization photocathodes
  - Structure
  - Supply chain issues
  - Initiatives for new growth methods
- Photocathode damage mechanisms
  - Ion acceleration
  - Vacuum improvements
  - Damage evaluation
- Novel ideas for photocathodes
- Conclusions

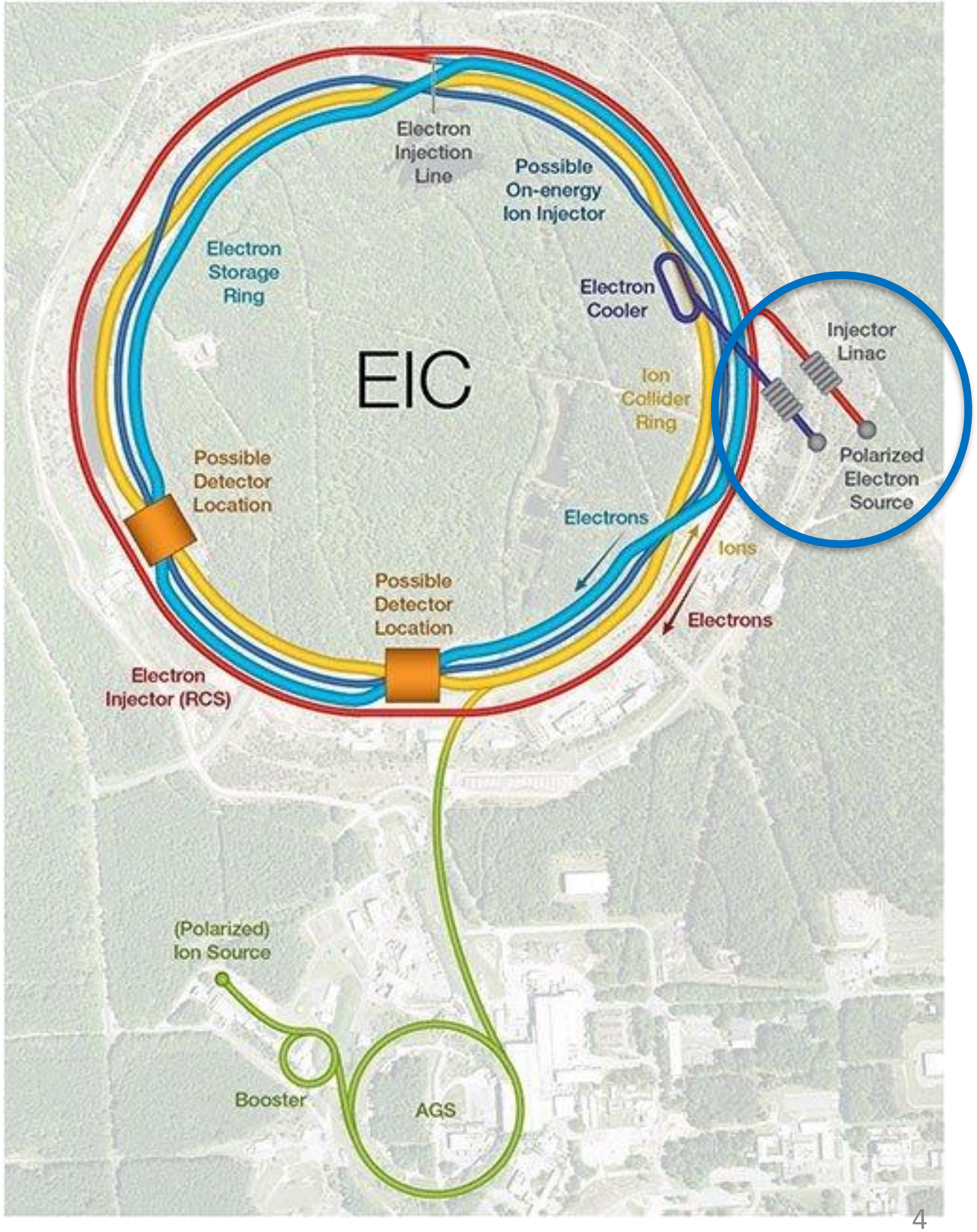
# CEBAF at Thomas Jefferson National Accelerator Facility

- 12 GeV electron accelerator for Nuclear Physics (DOE)
- Mission: To understand the transition from hadronic (protons and neutrons) to partonic (quarks and gluons) physics.
  - Electron beam, with polarization nearly 90% and current to 200 μA
  - Investigate spin dependent nuclear processes
  - ~1 mile racetrack design accelerator
  - 4 experimental halls




# Polarized electrons for EIC

- Electron Ion Collider
  - Next major DOE NP project
  - Location: Brookhaven Natl. Lab
  - BNL RHIC tunnel: 2.4 mile circumference
  - Add polarized electron beam
    - New capability for BNL
- Physics motivation
  - *Strong nuclear force*
  - *Proton spin origin*
  - *Proton mass origin*



# Why do we need polarized electrons?

- Nuclear physics electron scattering
  - Parity violation: differences in scattering rate between electron and nuclear spin states
    - QWeak
    - HAPPEX, PREX, CREX
    - MOLLER
  - Proton spin structures
    - Nature Physics **17**, p.736 (2021)
  - Nucleon electromagnetic structure
    - Nature **566**, p.354 (2019)

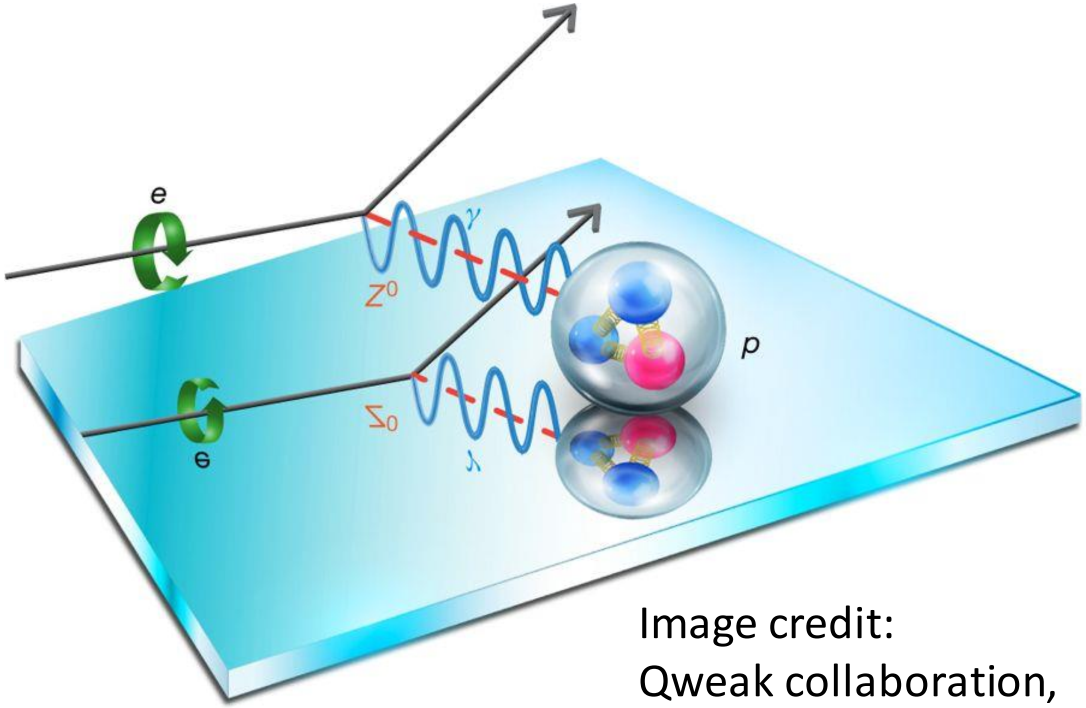
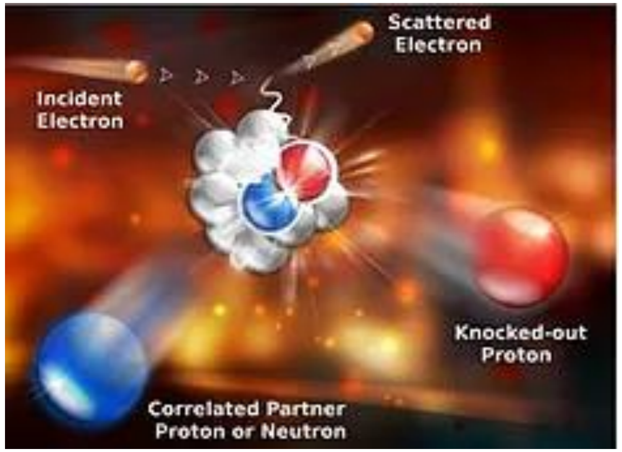
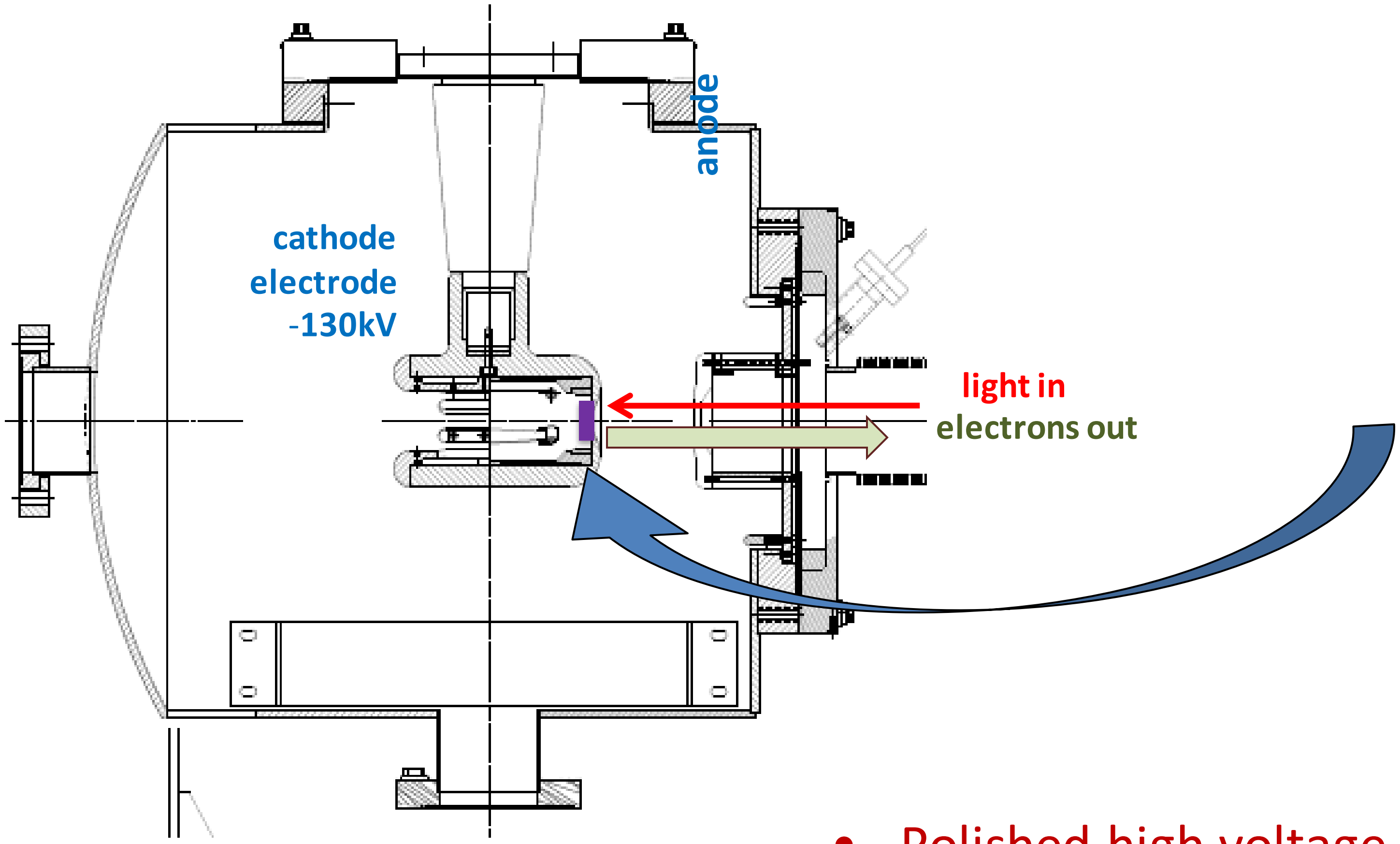
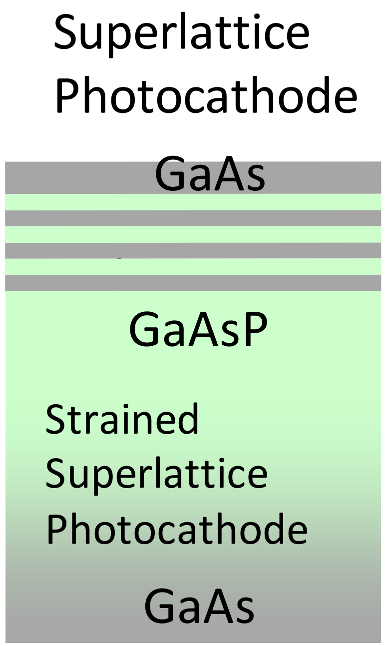



Image credit:
Qweak collaboration,
*Nature* **557**, p207 (2018).



# DC Photoemission Source

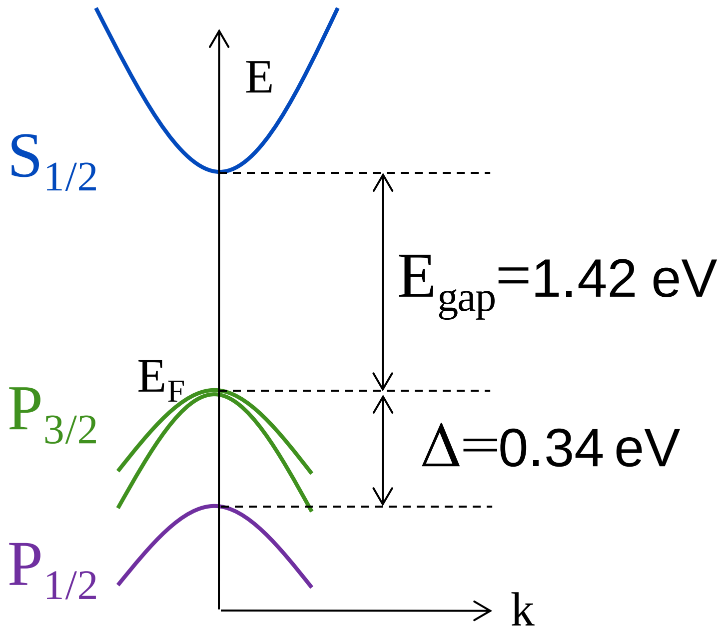


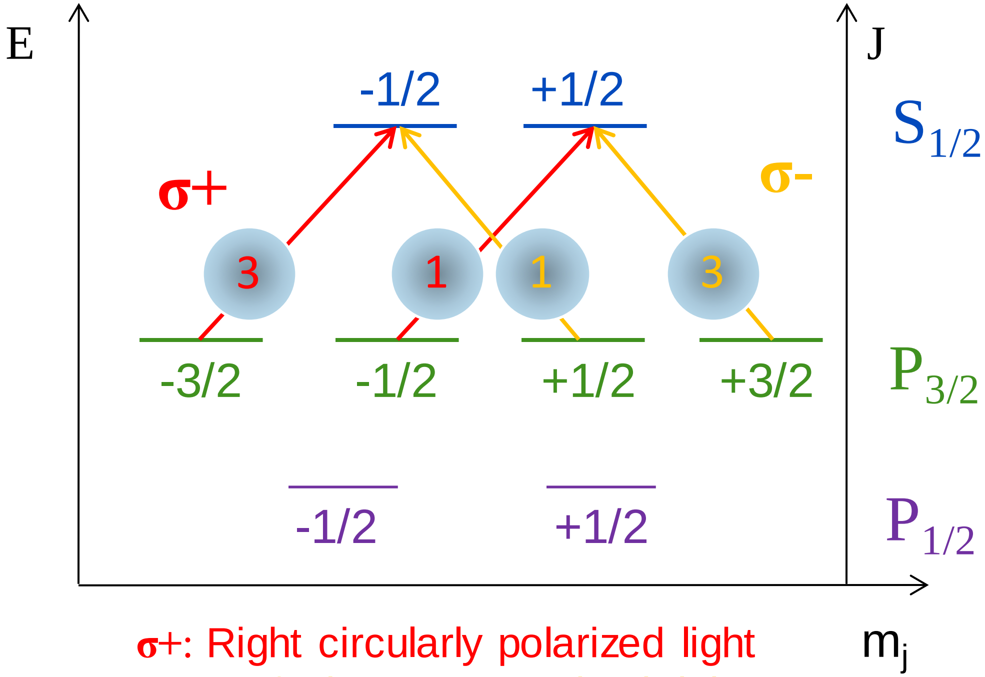





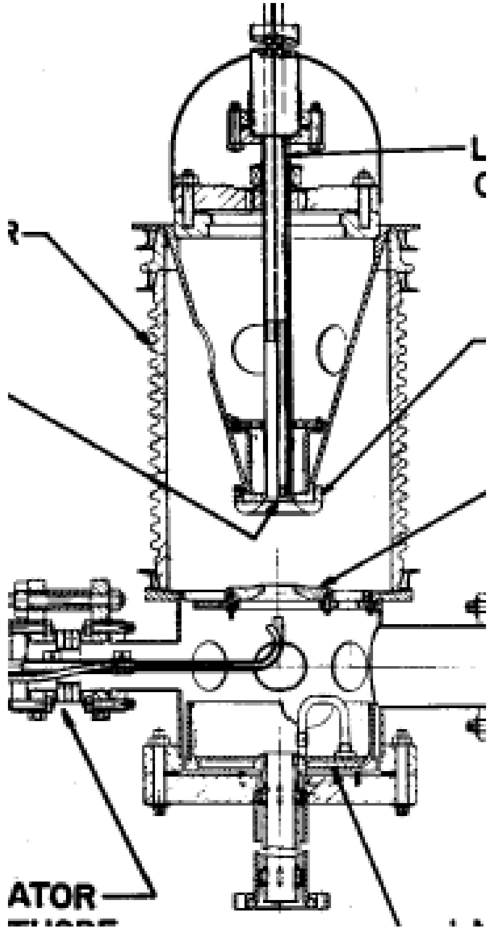
- Polished high voltage electrodes
- Strained superlattice GaAs/GaAsP photocathode
- Surface preparation: Cs, oxygen or fluorine
- Base pressure approaching XHV ≡ $P < 1x10^{-10}$ Pa

# Spin Polarized Photoemission from Bulk GaAs




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




SLAC 1977

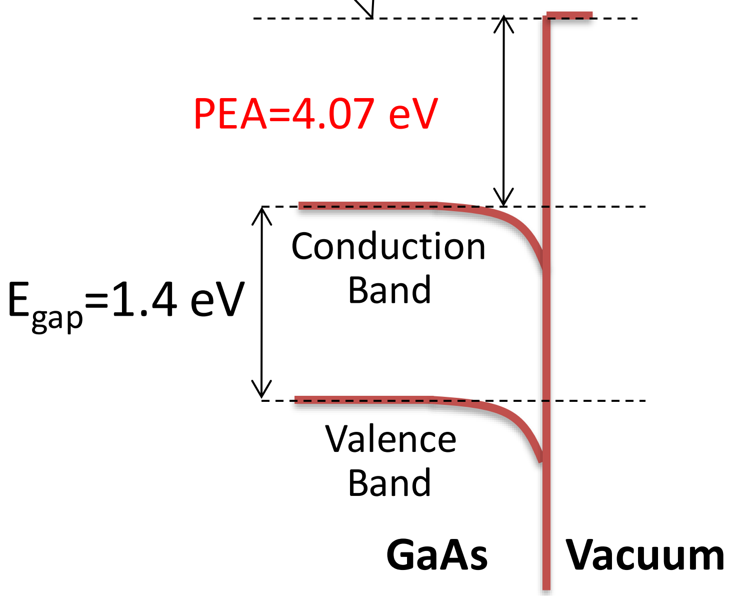
Note: Flip electron spin using laser polarization. Rate 30-1000 Hz

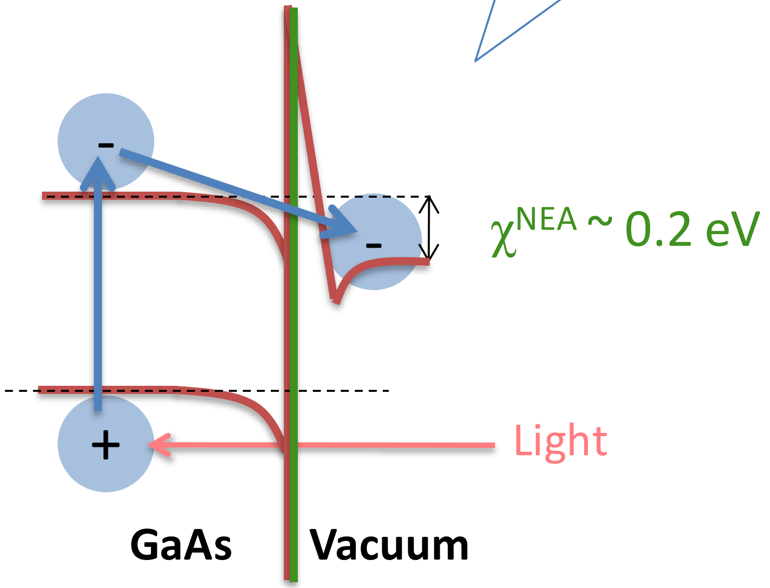
# Photoemission from GaAs

**Bare GaAs surface**
- Large Work Function
- Positive Electron Affinity (PEA)

NEA Surface Activation

**Dipole layers of Cs + $NF_3$ or Oxygen**
- Reduces Work Function
- Negative Electron Affinity (NEA)



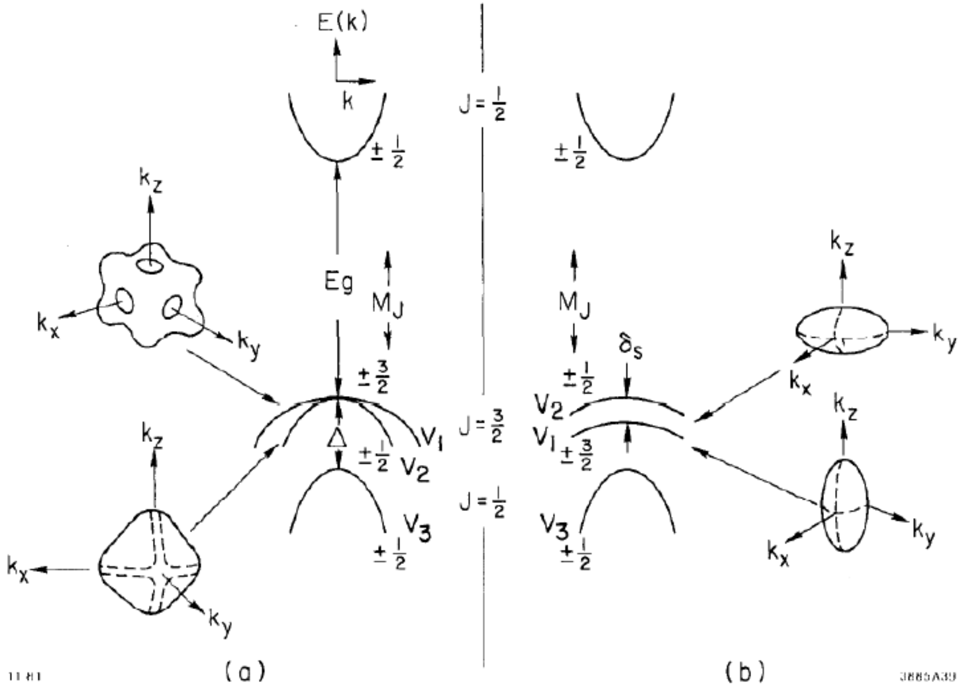




**GaAs Spin Polarized Electron Source, D.T. Pierce, R.J. Celotta, G.-C. Wang, W.N. Unertl, A. Galejs, C.E. Kuyatt, and S.R. Mielczarek, Review of Scientific Instruments 51(4), 478 (1980).**

# 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"

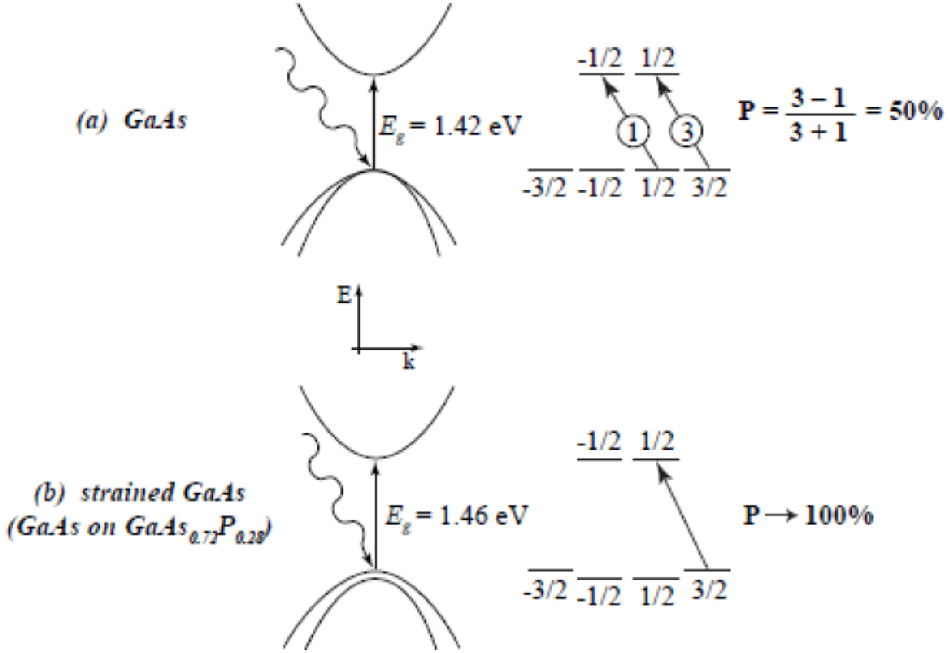
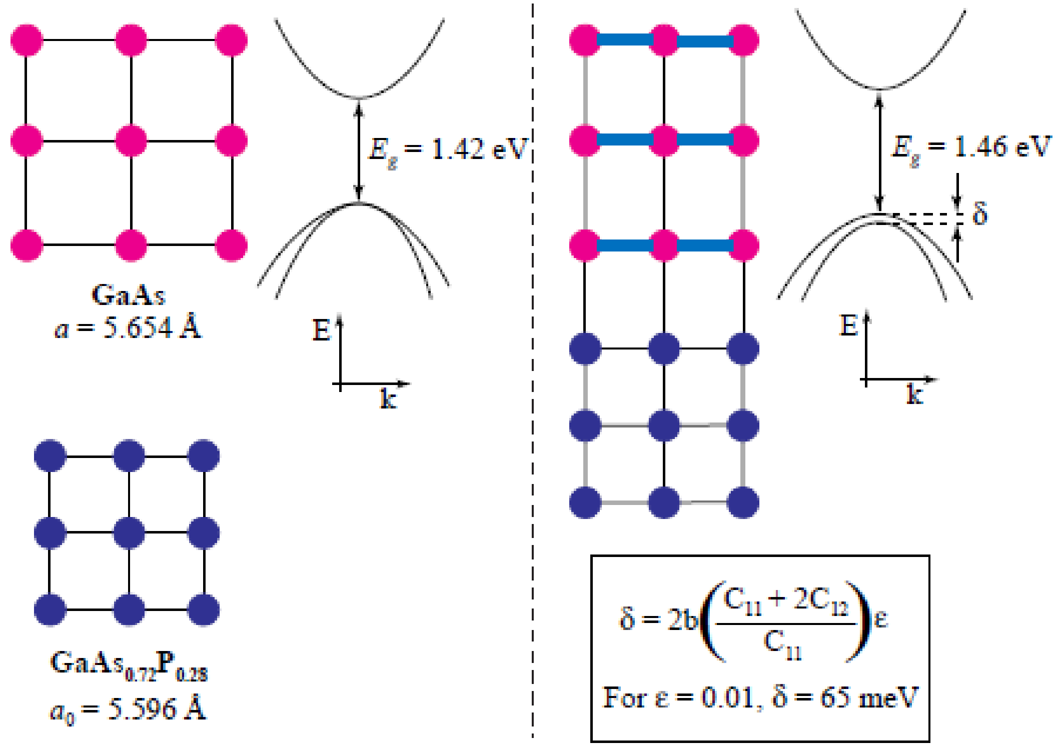



Image from Pablo Saez, PhD Thesis, Stanford University, SLAC Report 501, 1997

# Lattice mismatch provides stress

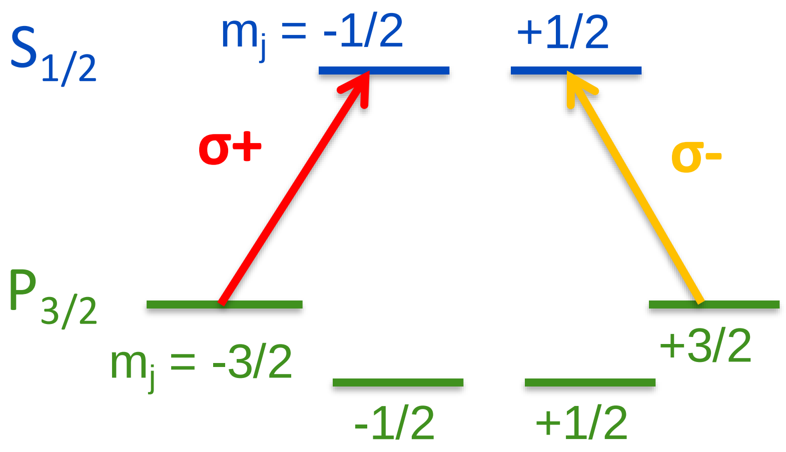


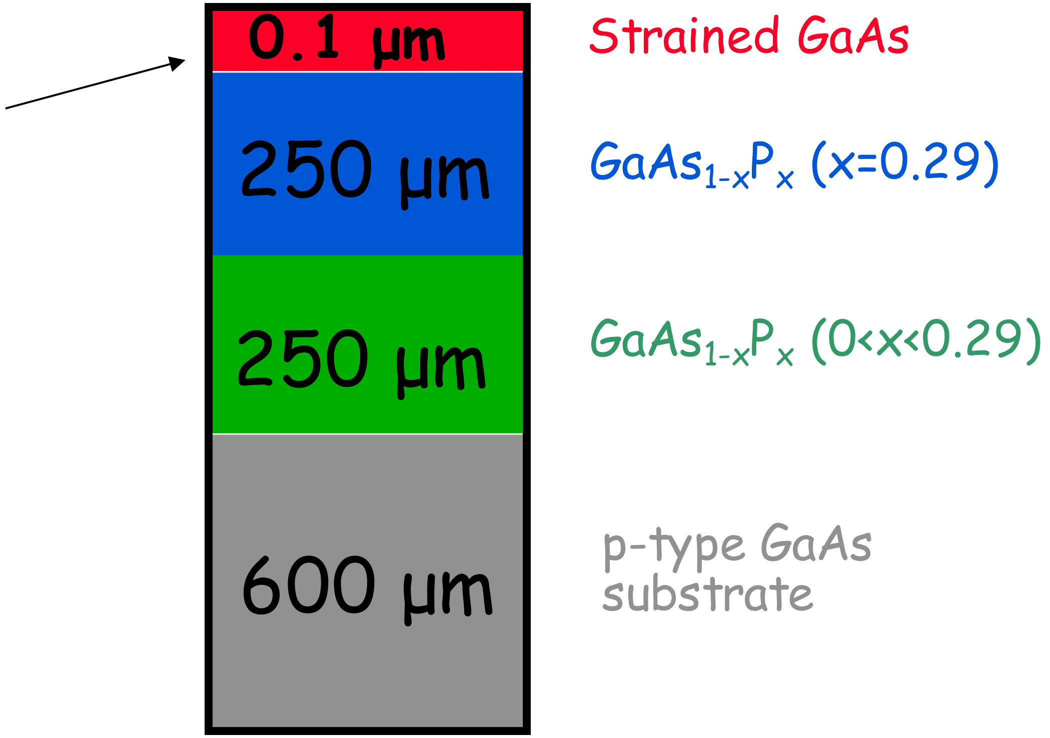

Pablo Saez, PhD Thesis, SLAC Report 501, 1997

- The band gap of the substrate layer must be larger than surface layer
- Lattice constants must differ enough to introduce suitable strain
- Adjust lattice constant of substrate by varying concentration of third element

1% lattice mismatch provides equivalent force as hydraulic press!

# Strained layer GaAs



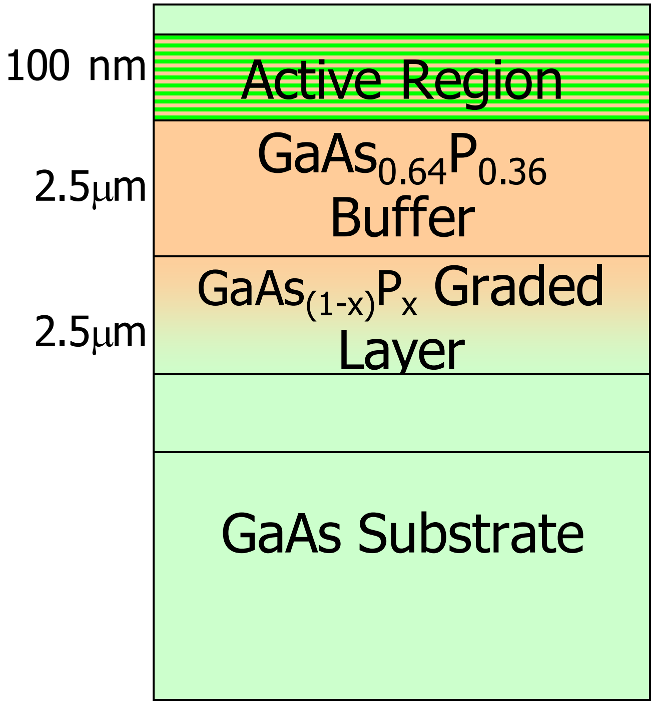



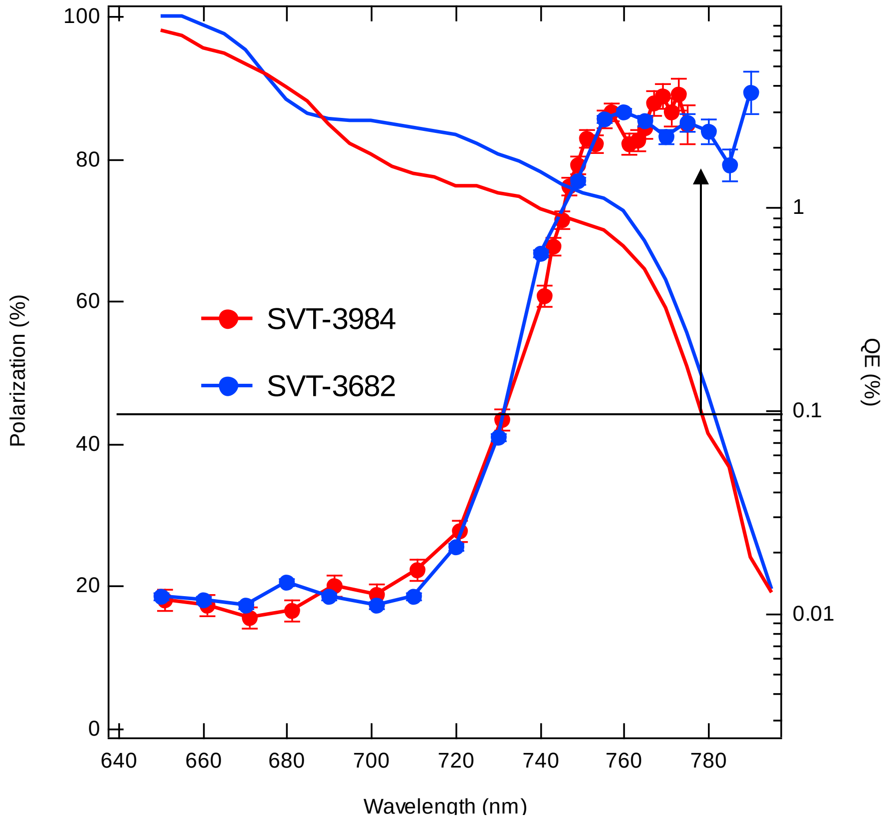

- ✓ Polarization 75% >> 50% ☺
- ✓ Strain relaxes in 100 nm layer
- ✓ QE 0.1%

MOCVD-grown epitaxial spin-polarizer wafer

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

# Strained layer superlattice GaAs/GaAsP




D. Luh et al, SLAC, PESP2002

QE 1% and Polarization 85%

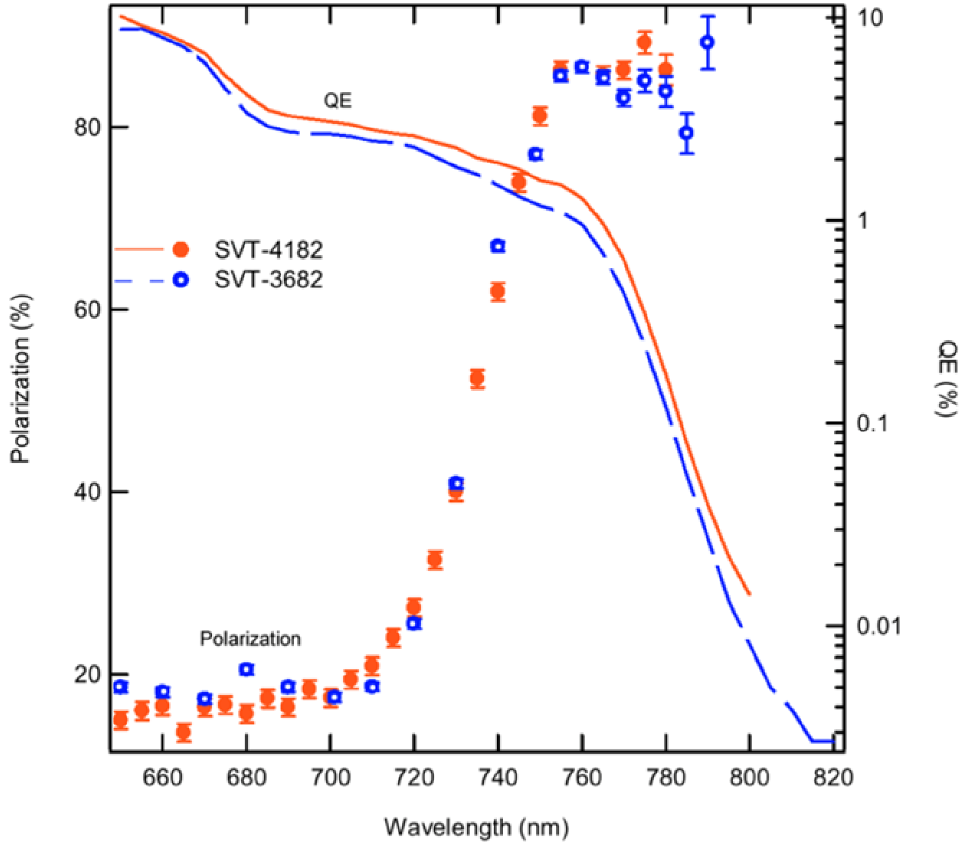
From Aaron Moy, SVT Assoc and SLAC, PESP2002

# Innovation 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
- Various Superlattice Structures
  - **GaAs/GaAsP**
  - GaAsSb
  - AlGaAs/GaAs
  - Distributed Bragg Reflector for high QE

Variations

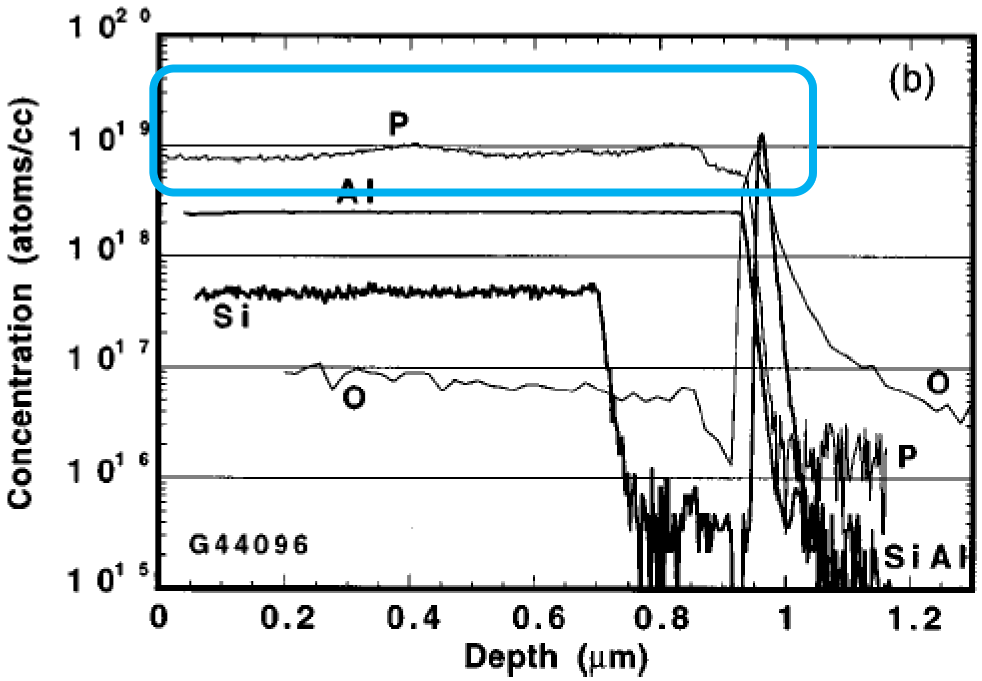
- Quantum Well thickness
- Barrier thickness
- Strain layer concentration
- Number of periods




AlGaAs/GaAs, A. Moy 2009

# 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

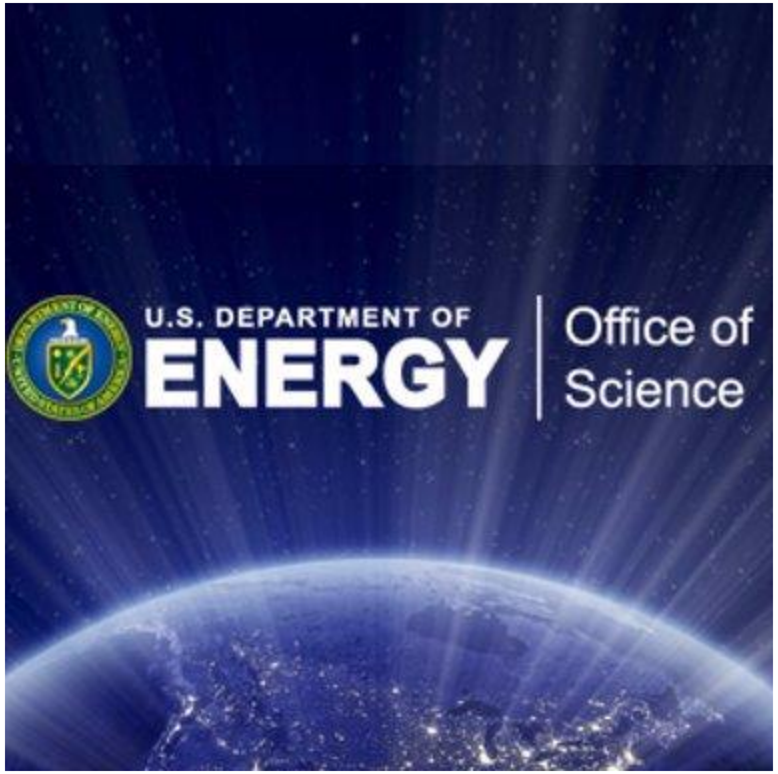



*SIMS of AlGaAs grown after Phosphorus contamination*

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

# 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 (yet?) financially viable



# 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

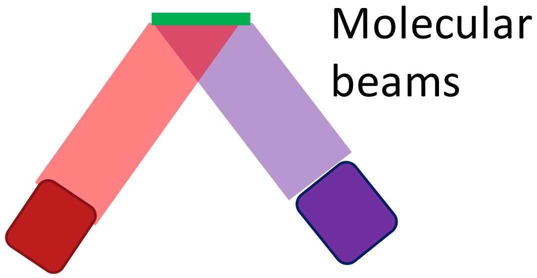
# MBE, GSMBE, CBE and MOCVD

## MBE

Gas Source Molecular Beam Epitaxy

elemental As, P, Ga

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

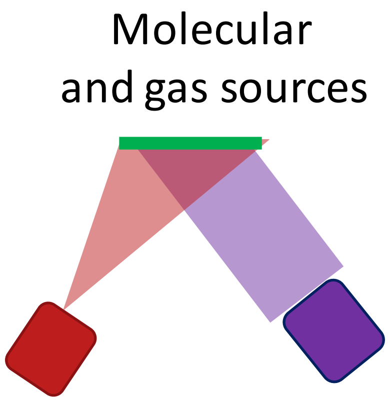



## GSMBE

Gas Source Molecular Beam Epitaxy

$AsH_3$, $PH_3$, elemental Gallium

**Used at SVT**

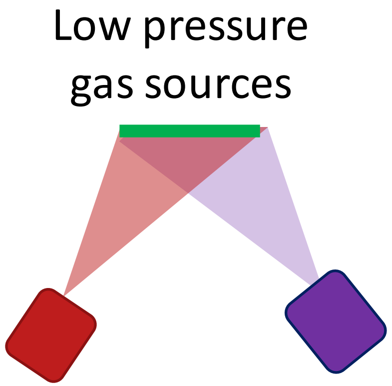



## CBE

Chemical Beam Epitaxy

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

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



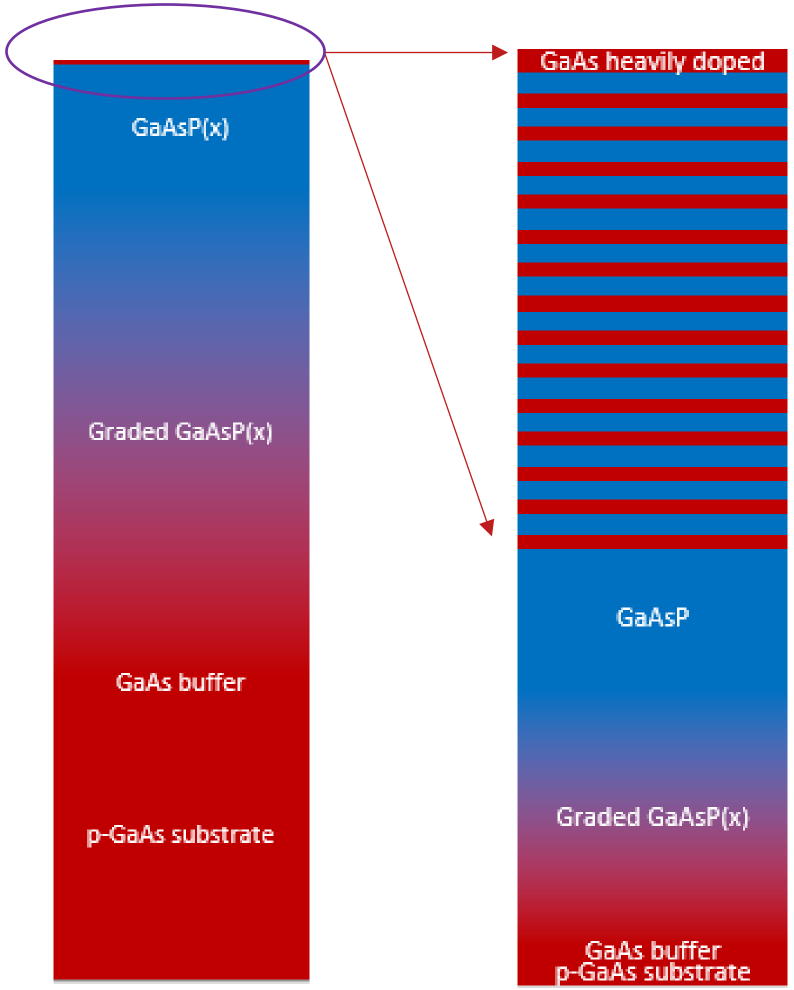

## MOCVD

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




To scale

Superlattice scaled up

- 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

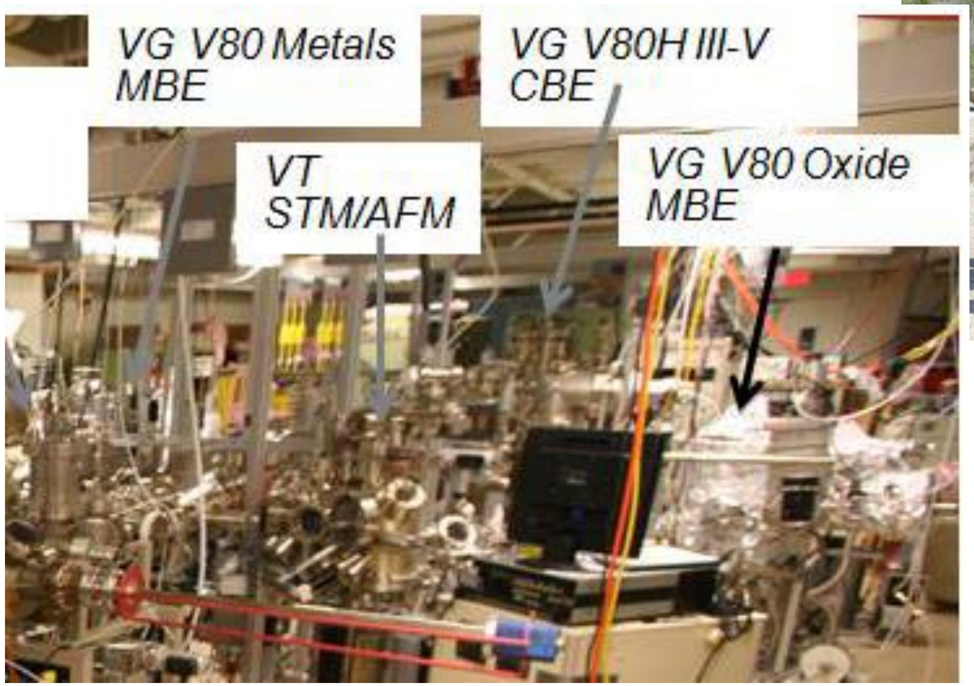
- Substrate Temperature
- Source Temperature/Pressures
- Time
- Grading profile
- Underlying crystal orientation
- Superlattice layer thickness

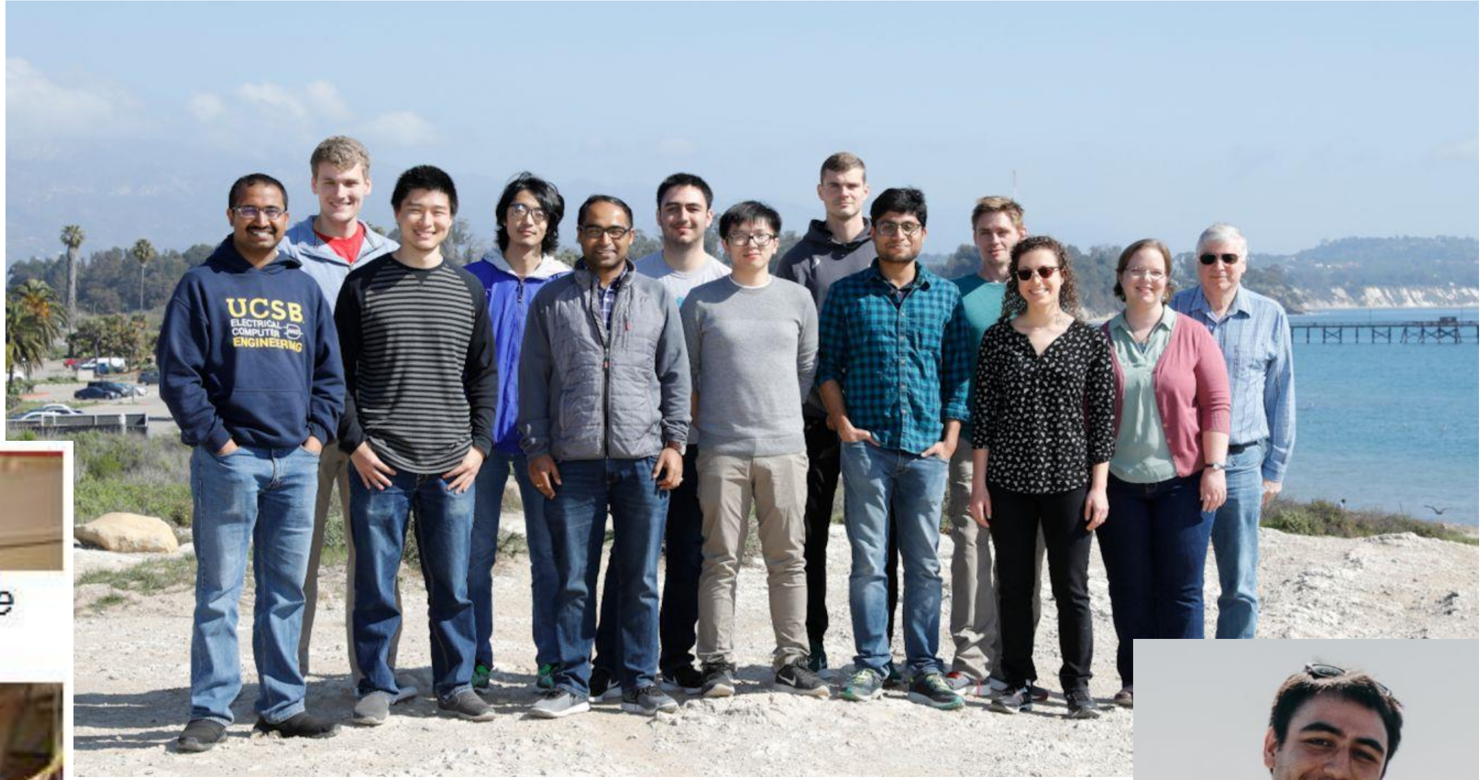
# CBE: Photocathode progress

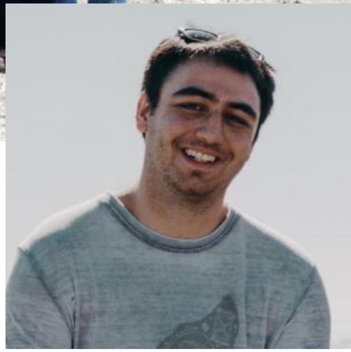
Chris Palmstrøm Group, UCSB

- Aaron Engel, graduate student

- Chemical Beam Epitaxy System



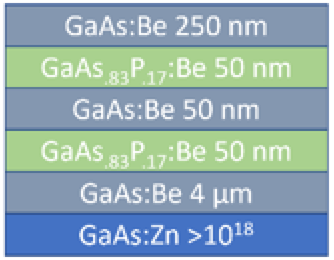



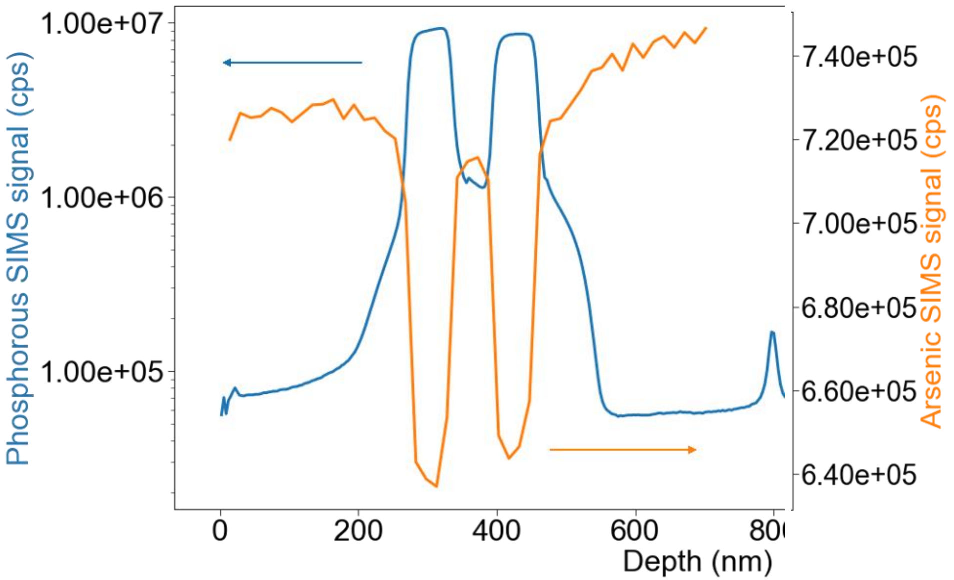



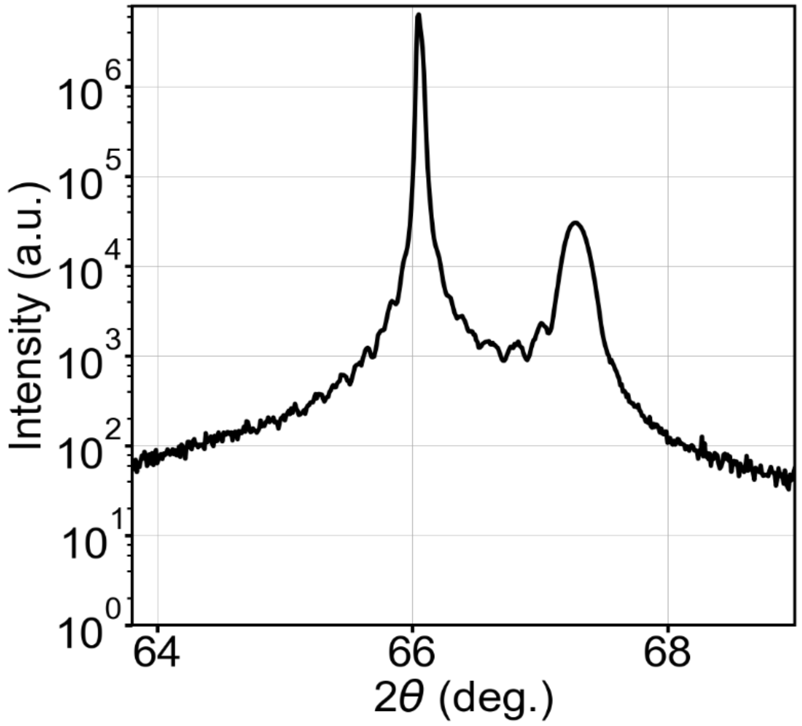
# CBE: Photocathode progress

- 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



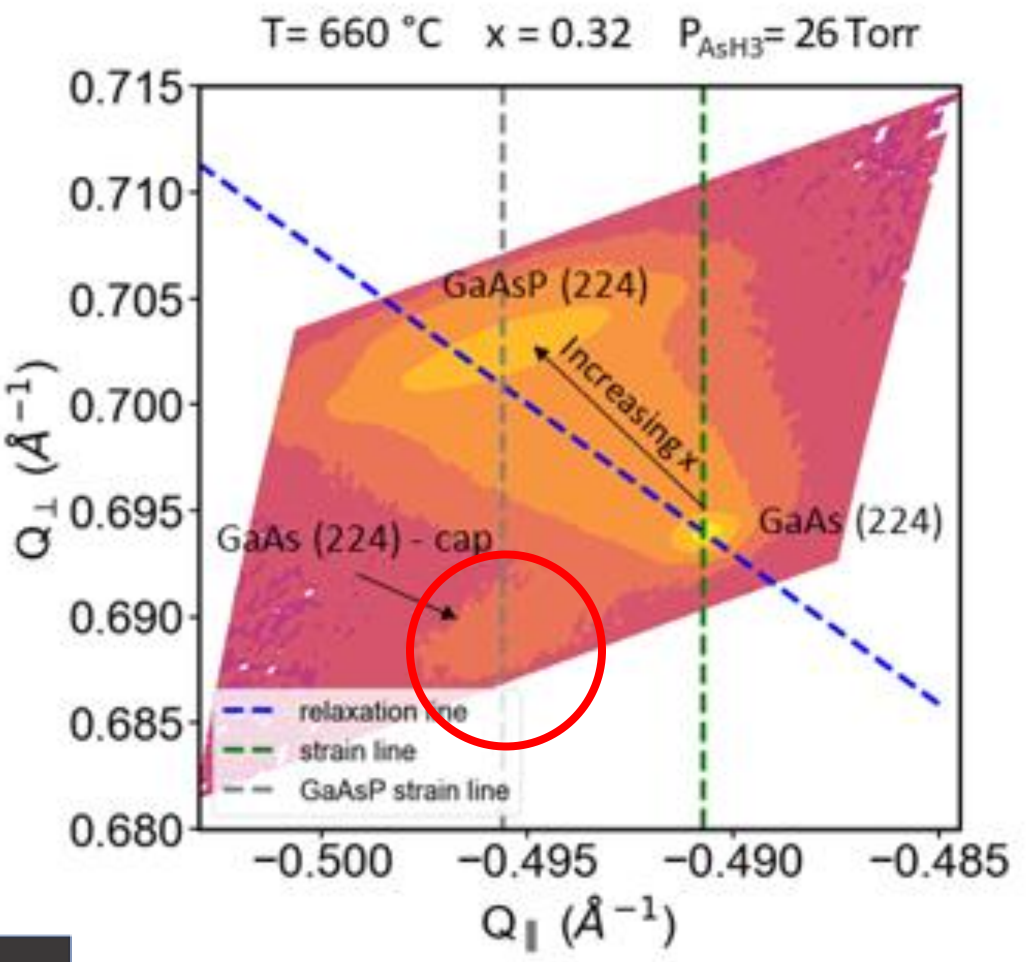



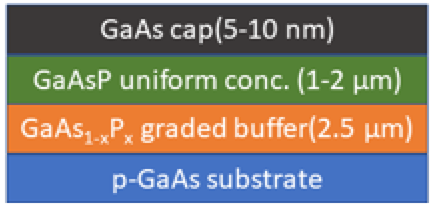




x-ray diffraction:
1 micron $GaAs_{0.75}P_{0.25}$ on GaAs
Testing analysis tools for interfaces

# 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

# 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

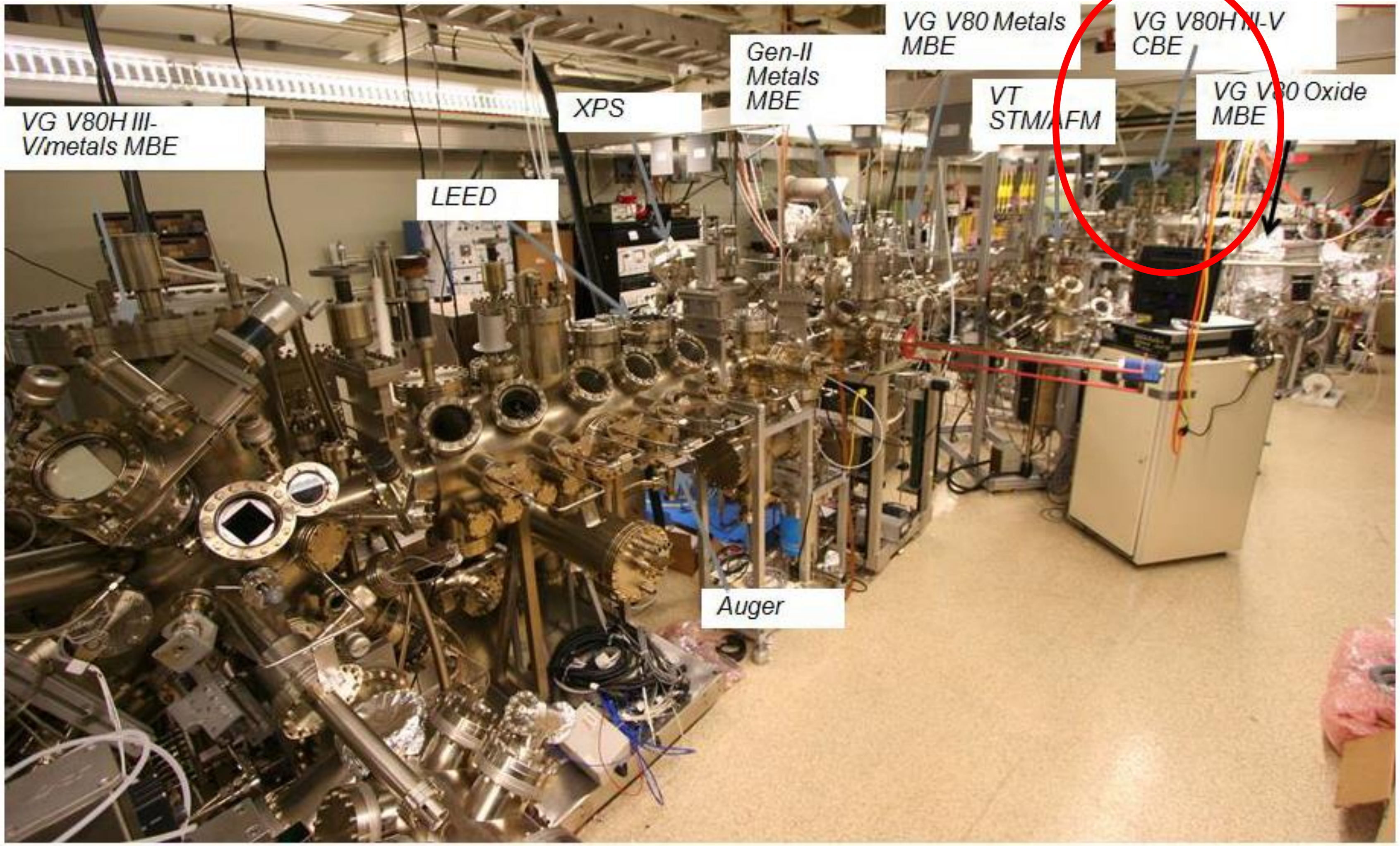


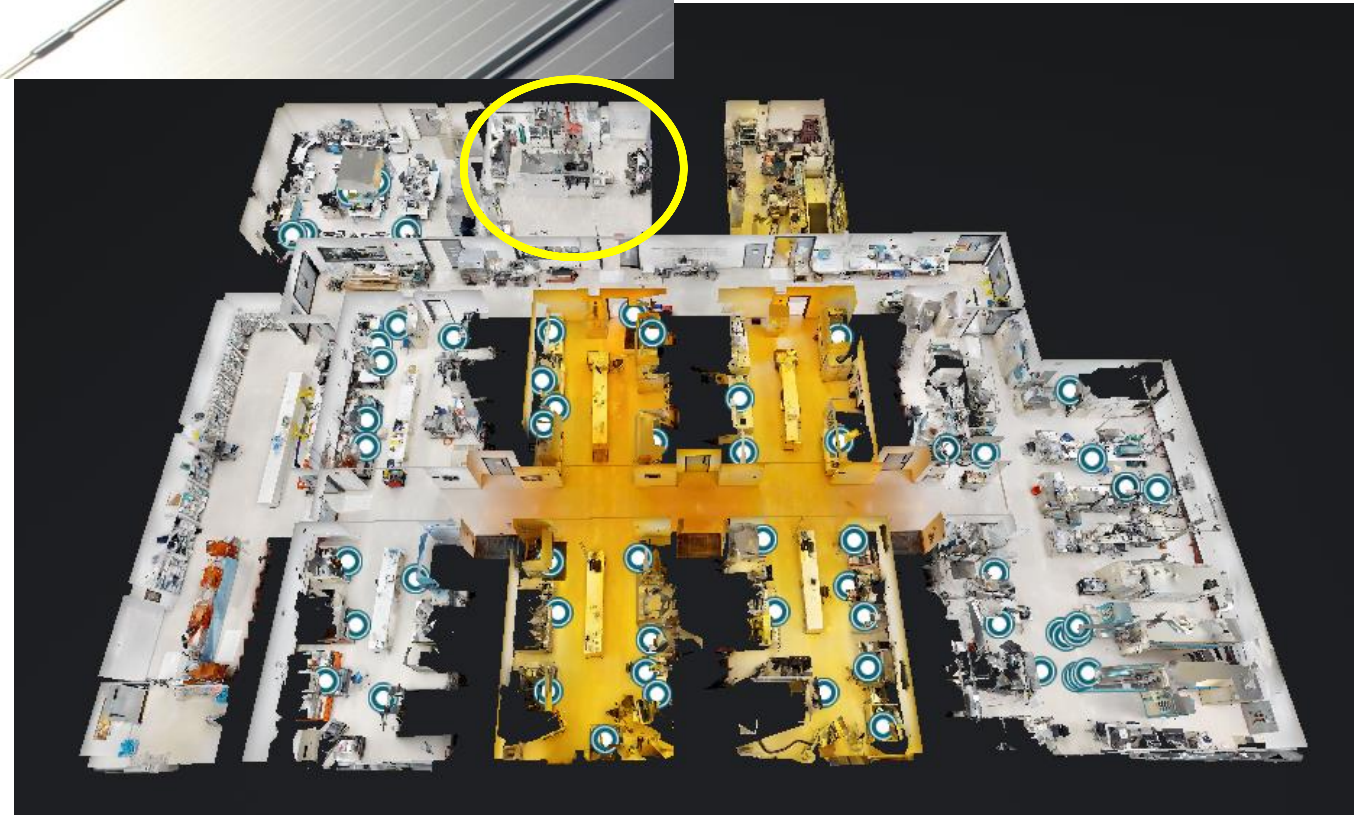

*Figure 2 Semiconductor deposition system at Chris Palmstrom's lab at UCSB. The CBE system for the growth of this material is shown at the back and labelled "VG V80H III-V CBE".*

# MOCVD: Photocathode progress

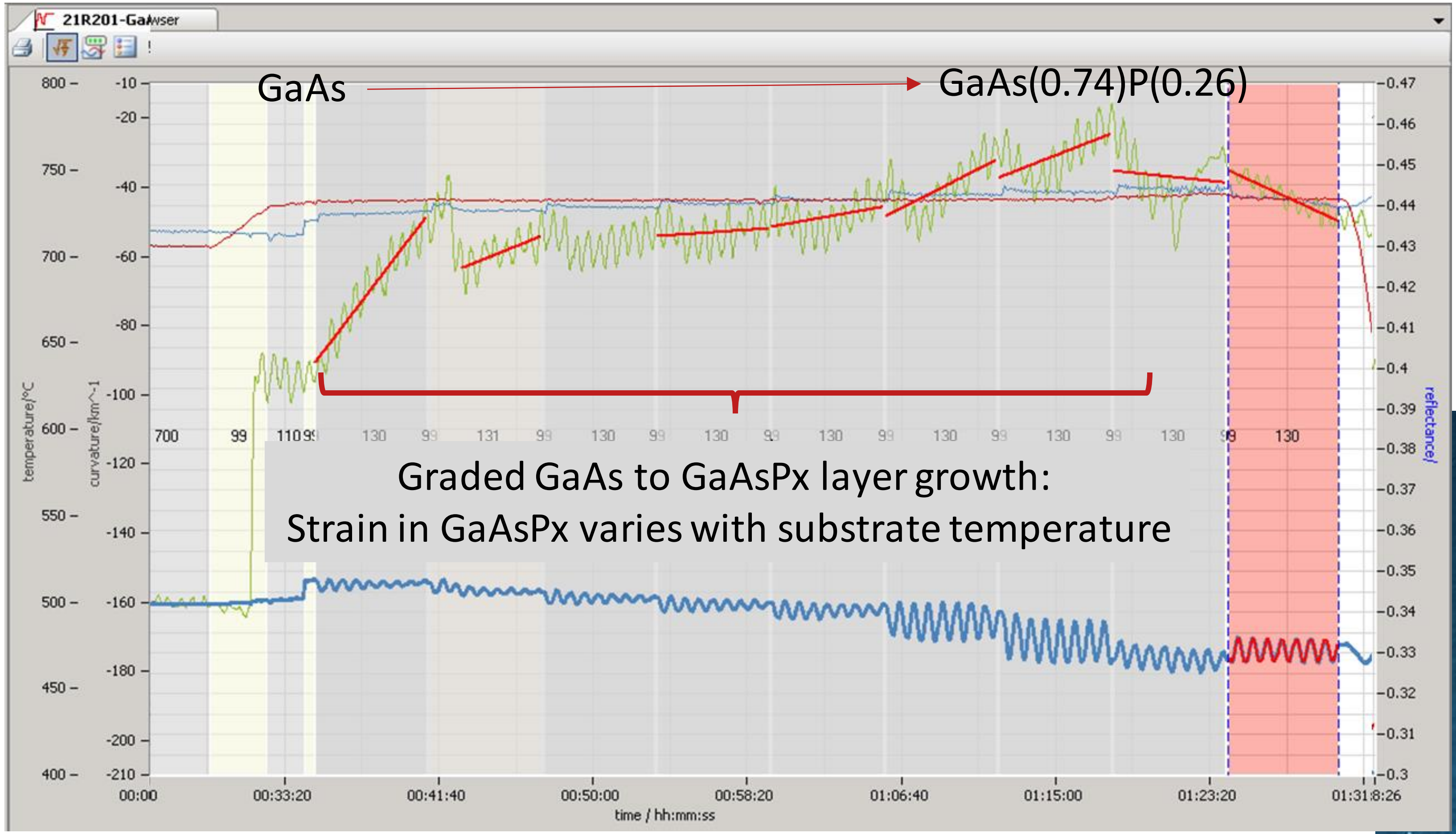

The Rochester Institute of Technology
III-V EPICenter

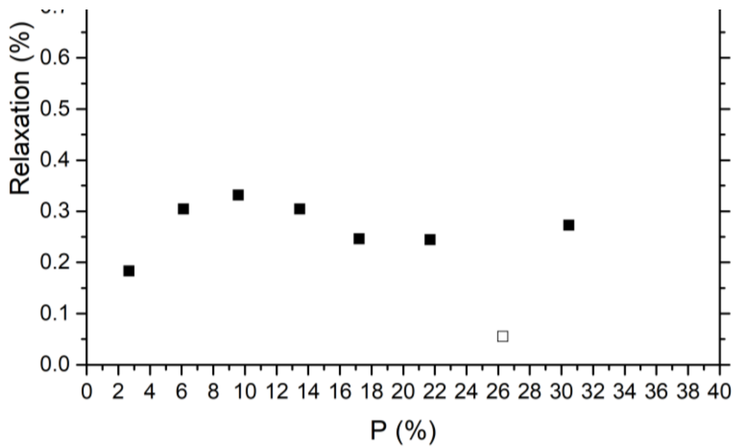
Dr. Sylvain Marsillac,
Old Dominion University

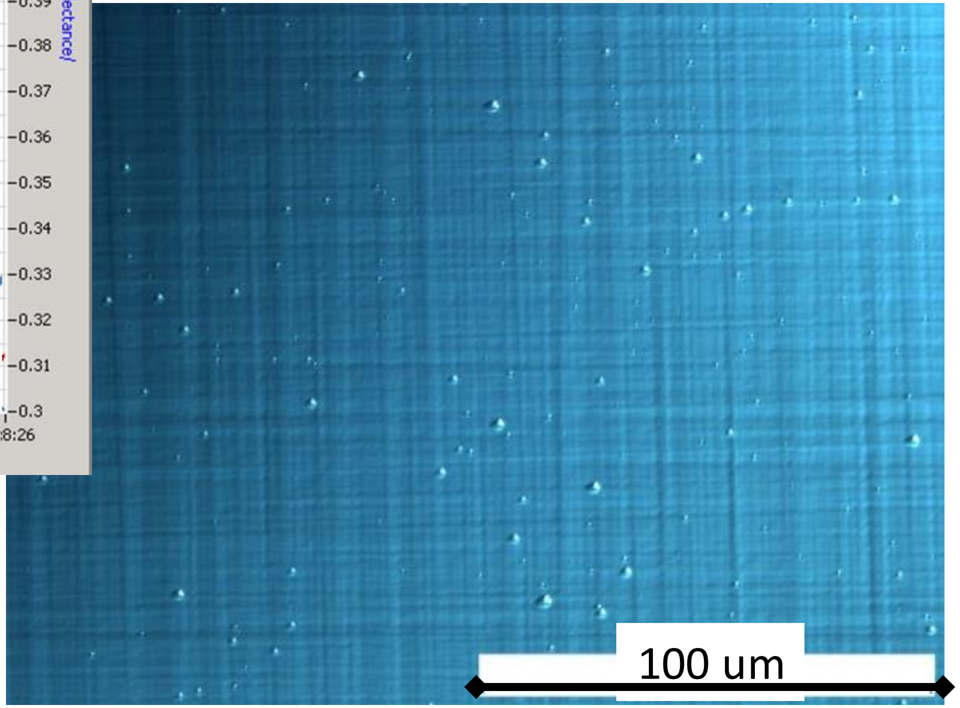
Ben Belfore
ODU Graduate Student



# Results: MOCVD



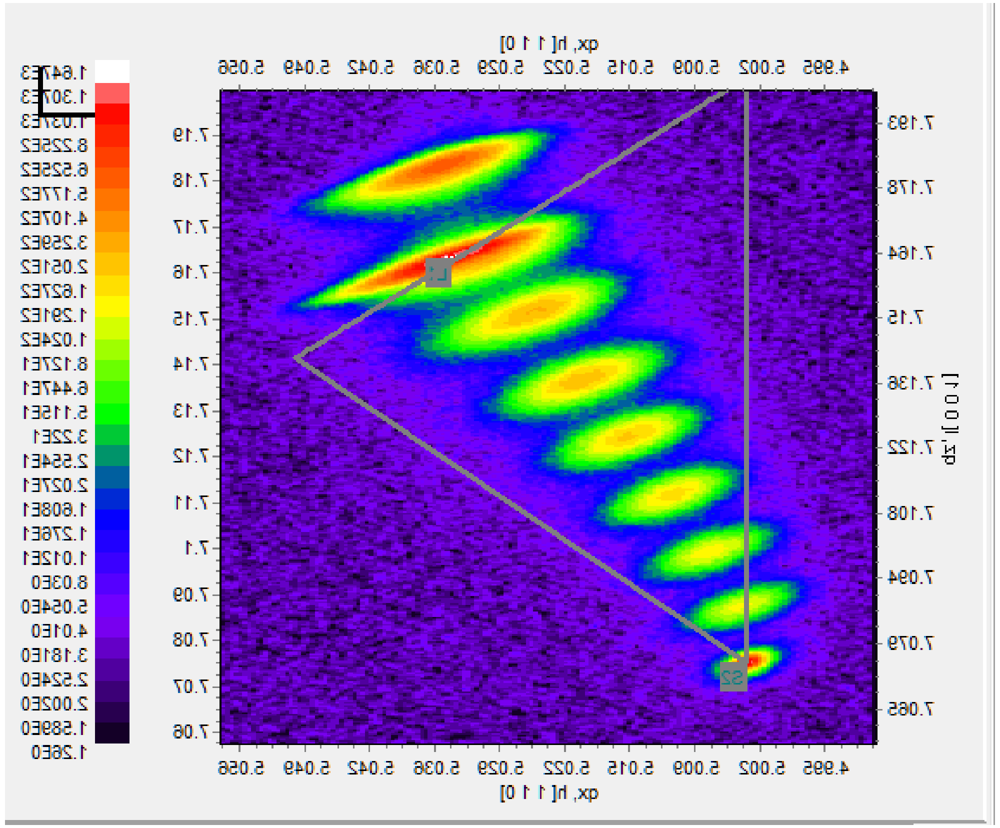

Higher temperatures yield improved surface with moderate relaxation throughout

730°C growth temperature

Optimizing temperatures, graded layer profile

# Results: MOCVD photocathode progress

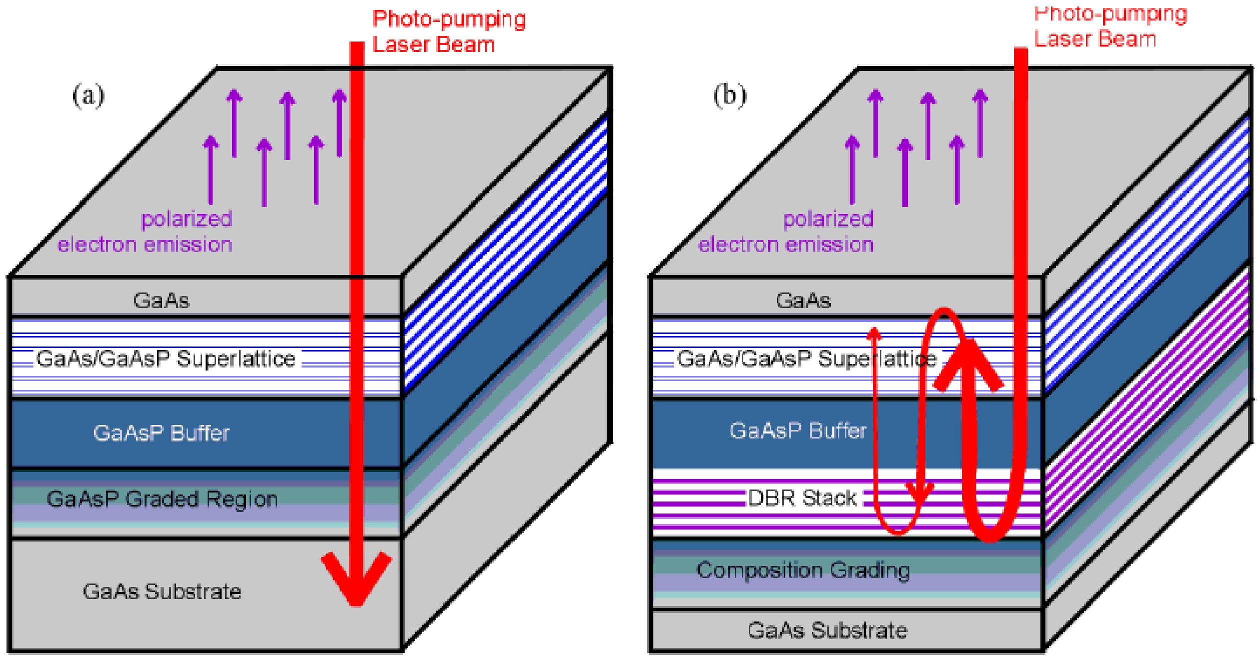
- Graded layer "metamorphic" test runs
  - Optimizing parameters for highest relaxation
  - Hall effect measurements for dopant characterization
- Superlattice runs
  - Growing superlattice on each metamorphic run
  - Optimizing parameters with zinc dopant
- Characterization
  - Surface analysis (SIMS, 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

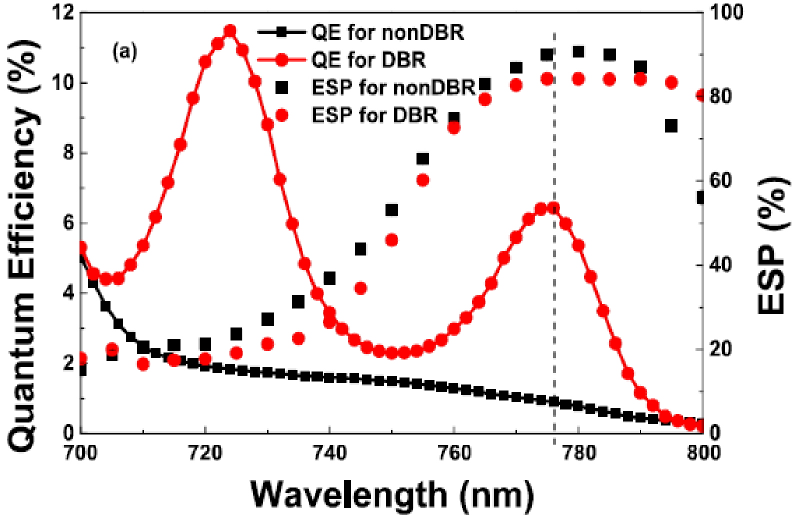
# Distributed Bragg Reflector




SVT DBR:

Polarization >80%

QE 6% (6x typical)



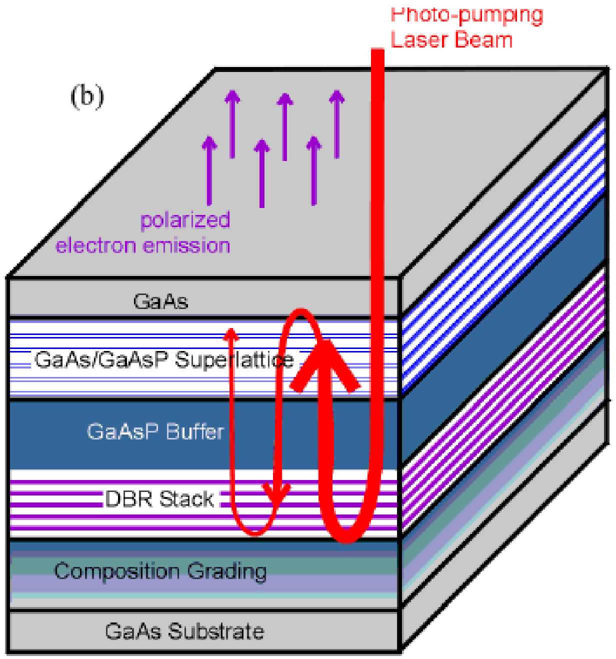

DBR structure has Fabry Perot structure

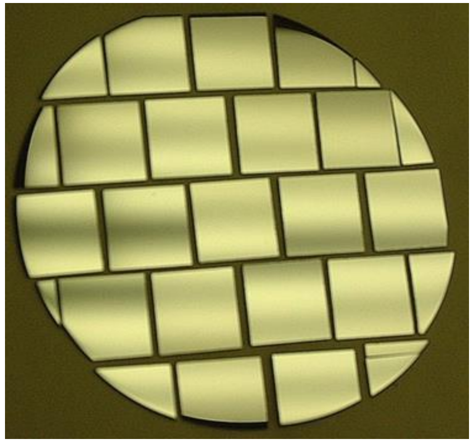
- Thickness tuned to desired wavelength
- Multiple passes -> more excited photons
- Polarization of light preserved

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



Representative diced GaAs wafer

| | | |
|---|---|---|
| GaAs | 5 nm | $p = 5x10^{19}$ $cm^{-3}$ |
| GaAs/ $GaAs_{0.62}P_{0.38}$ (30 pairs) | 4/4 nm | $p = 5x10^{17}$ $cm^{-3}$ |
| $GaAs_{0.81}P_{0.19}$ | 300 nm | $p = 5x10^{18}$ $cm^{-3}$ |
| $AlAs_{0.78}P_{0.22}$/$GaAs_{0.81}P_{0.19}$ (10 pairs) | 65/55 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 | | $p > 1x10^{18}$ $cm^{-3}$ |

BNL Mott polarimeter measurements

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

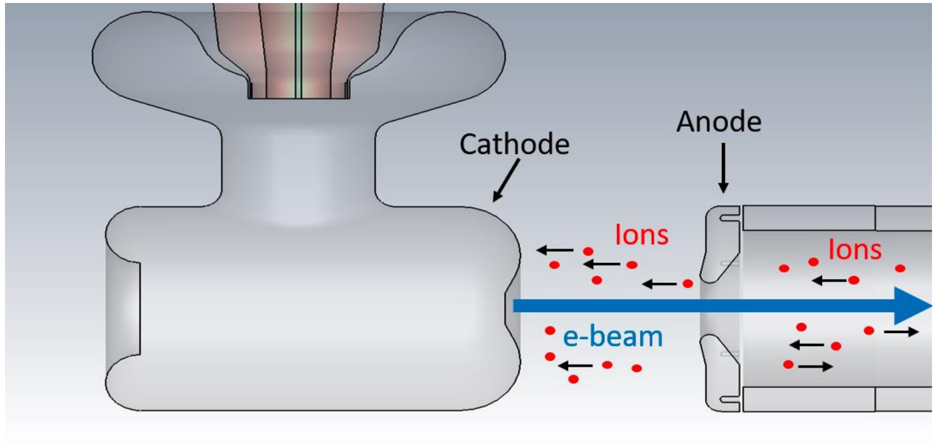
Issues: non-uniformity across photocathode

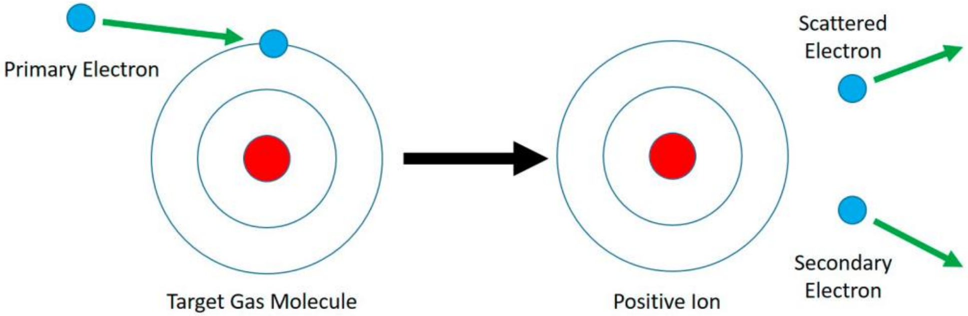
Future: Optimize and test

# Outline

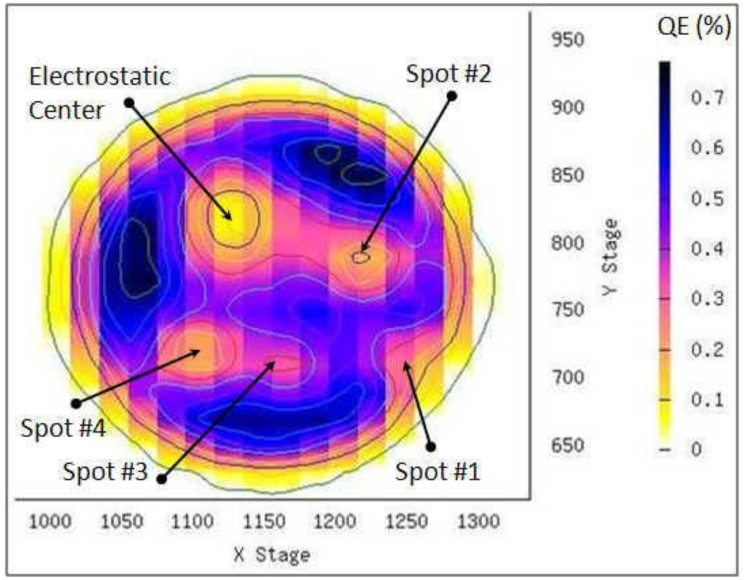
- Jefferson Lab, EIC and polarized electrons
- Photoemission from GaAs
- High polarization photocathodes
  - Structure
  - Supply chain issues
  - Initiatives for new growth methods
- **Photocathode damage mechanisms**
  - Ion acceleration
  - Vacuum improvements
  - Damage evaluation
- Novel ideas for photocathodes
- Conclusions

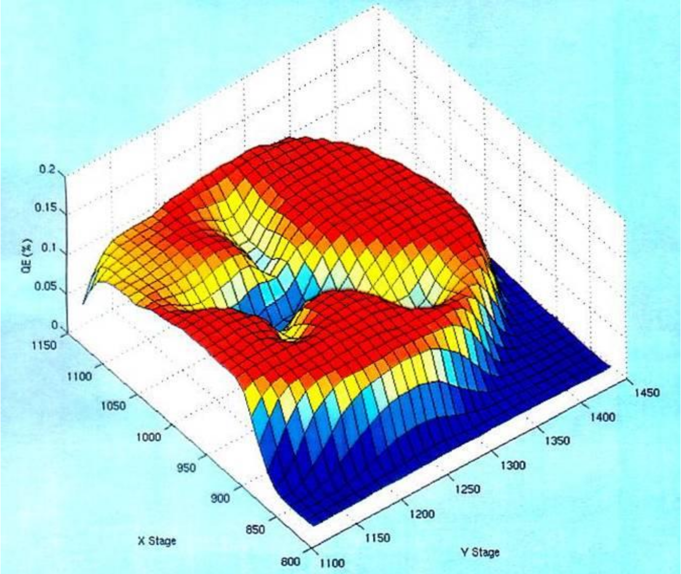
# Photocathode Lifetime limitations: Ion back-bombardment




Ionized residual gas limits lifetime
1. Implantation/surface chemistry
2. Liberates gas: x-ray or secondary e-



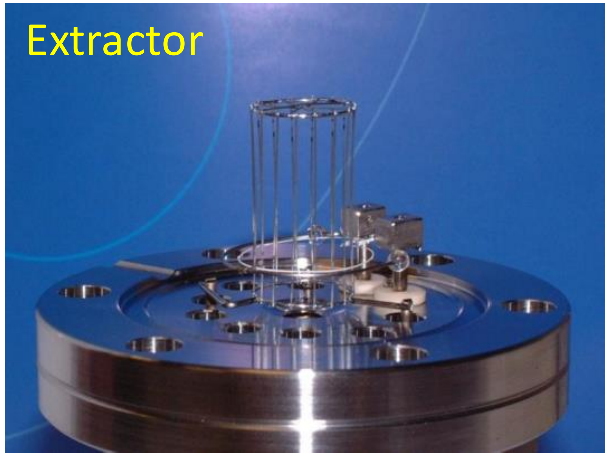



# Photocathode Lifetime improvements: Vacuum system

Installed pumping

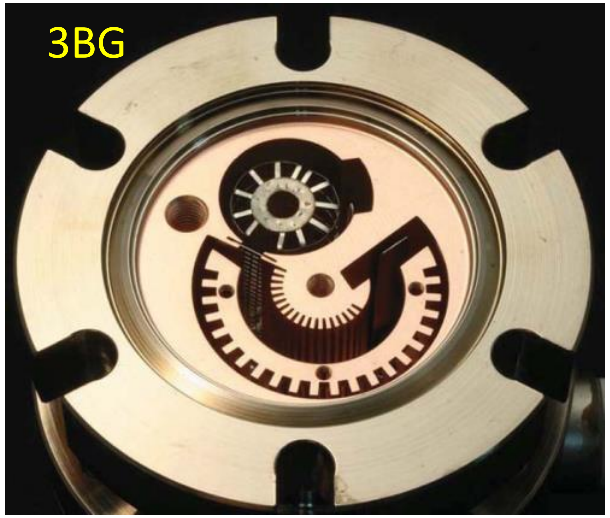
- SAES WP1250 modules x 10 (5600 L/s total)
- Gamma SEM/XHV ion pump (45 L/s)

~ 25 micron TiZrV coating

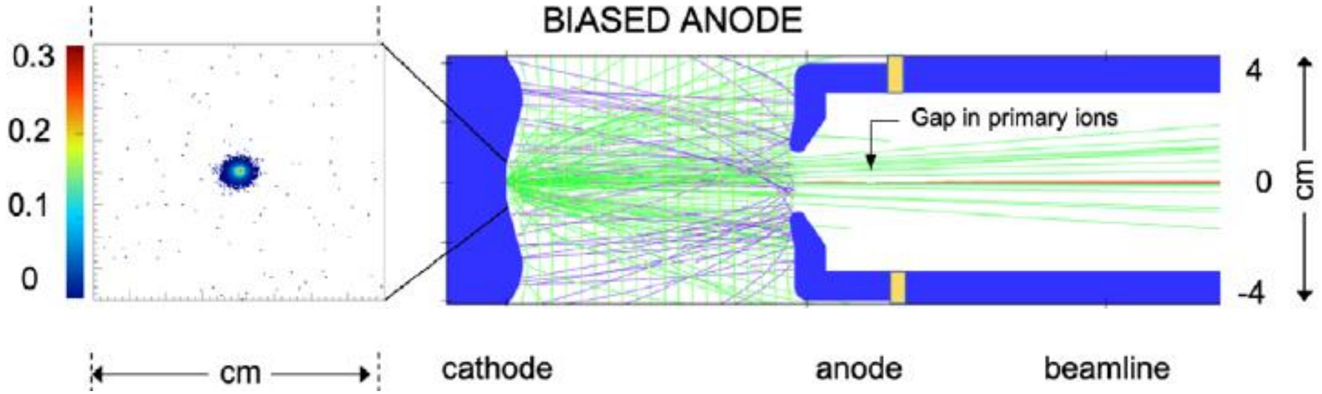




Measured pressure ~$1.3 \times 10^{-10}$ Pa
(extractor gauge, x-ray limit measured and subtracted)
Charge Lifetime: **~200 Coulombs**

Extractor gauge
x-ray limit reduced through geometry
Watanabe 3BG (Bent Belt Beam) gauge
230° deflector BeCu housing
JVSTA **28**, 486 (2010)

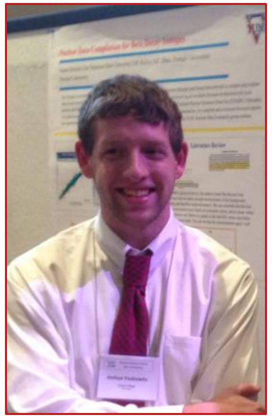


# Biased Anode for Lifetime Enhancement

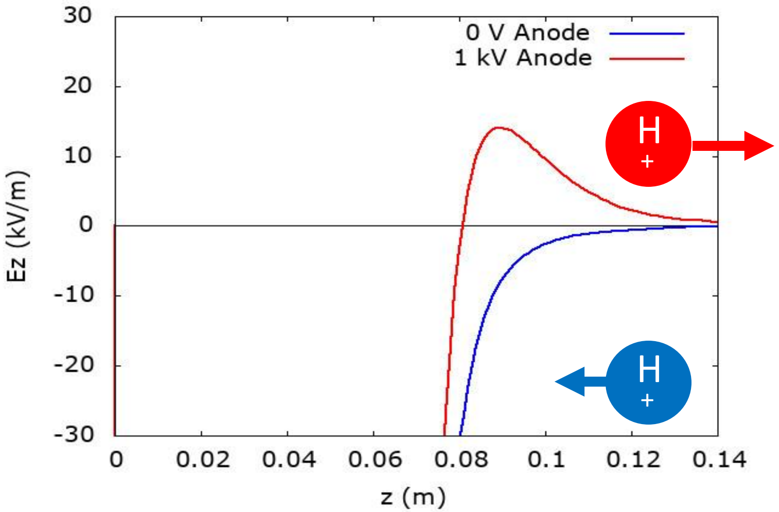



Simulations by C. Valerio in C. Hernandez-Garcia et al., Phys. Rev. Accel. Beams **22**, 113401 (2019)

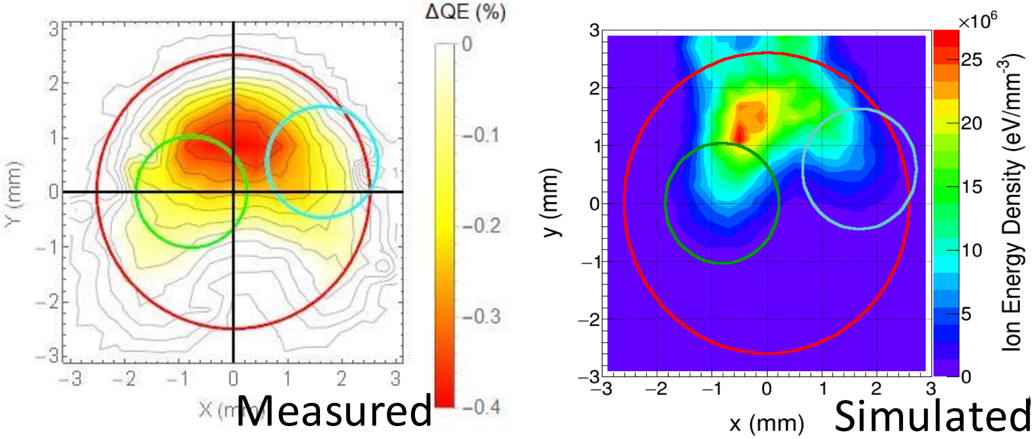
- Lifetime limitation: Ionized gas from beamline incident on photocathode
- JoshYoskowitz, Ph.D. research Anode bias in CEBAF gun
  - Unbiased Lifetime < 200 Coulombs
  - Biased Lifetime ~300 Coulombs
- Injector beamline rebuild 2021
  - Lifetime consistently over 250 C during 4 hall beam operation



Josh Yoskowitz
PhD thesis work

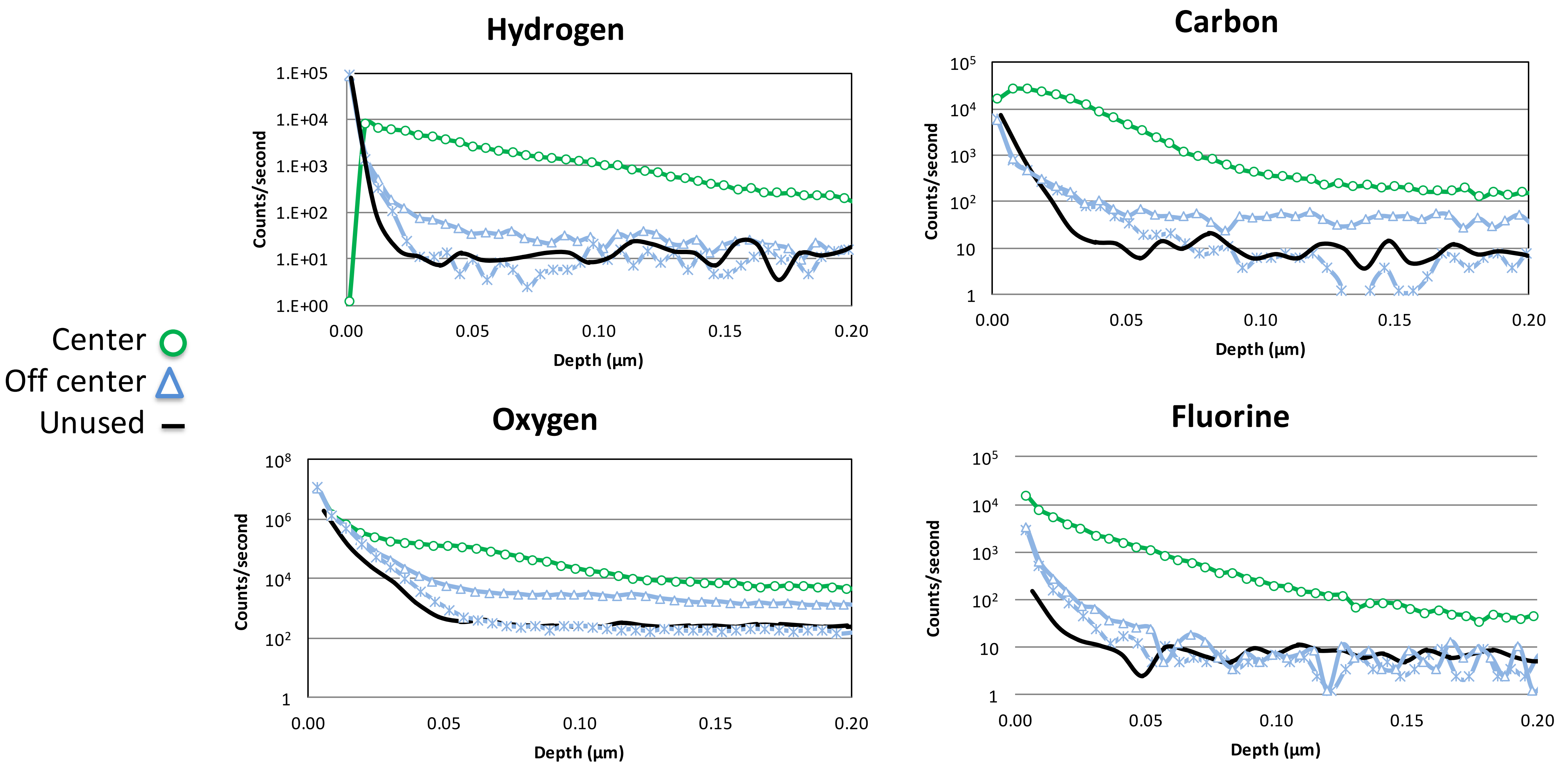



cathode damage



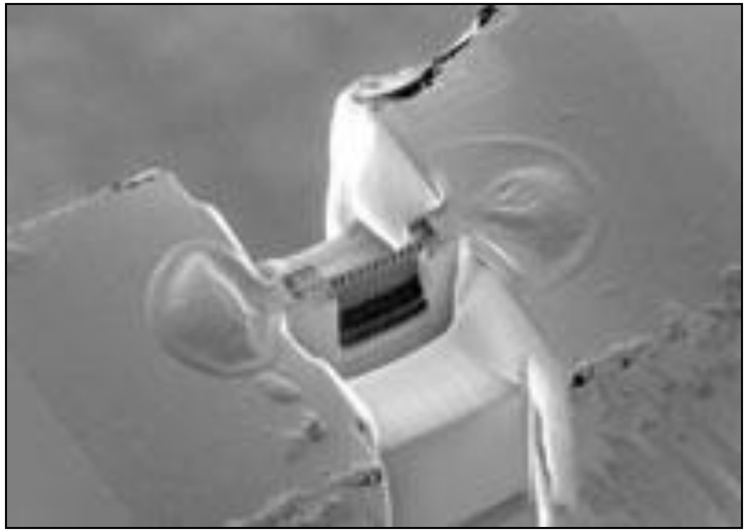

Measured Simulated

# Residual gas implantation: SIMS analysis

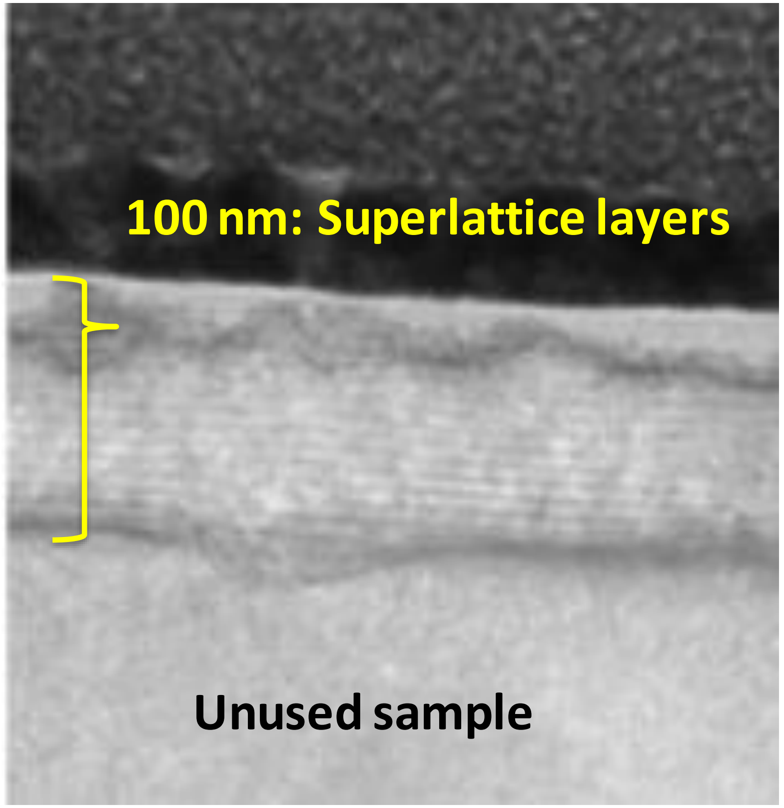


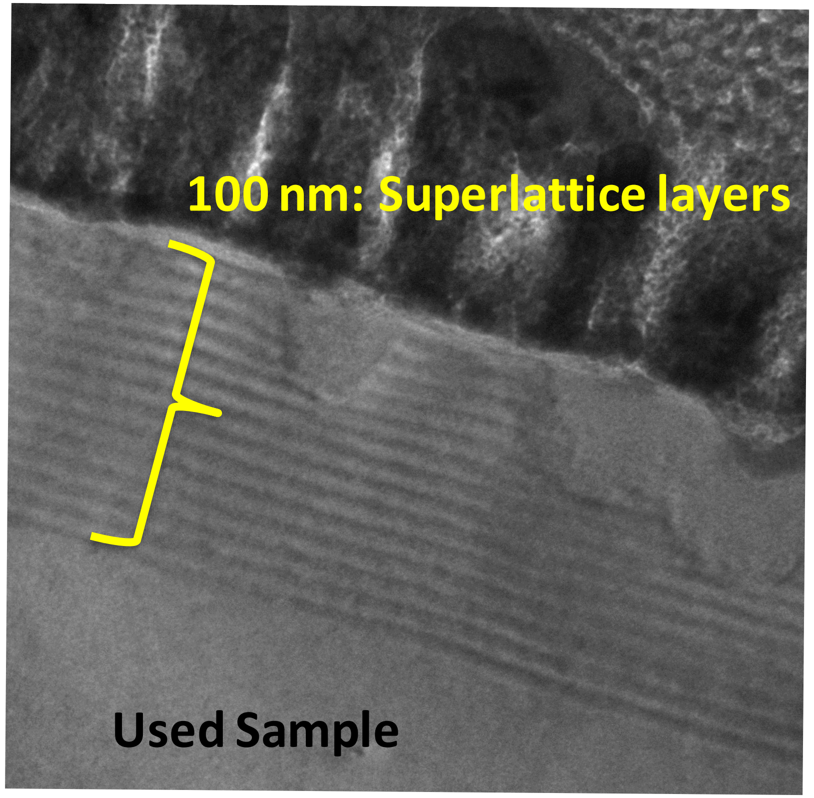

V. Shutthanandan, …, M.L. Stutzman, et al., Surface science analysis of GaAs photocathodes following sustained electron beam delivery, PRSTAB **15** 063501 (2012)

# Liftout FIB / TEM analysis



F.A.Steve et al., Surf. Interface Anal. **31**, 345, (2001)




- Focused Ion Beam used to mill cross sectional depth profile for TEM
- Au/Pd evaporated, Pt sputtered to protect surface (misalignment = shadows)
- Uniform layers, no darkening

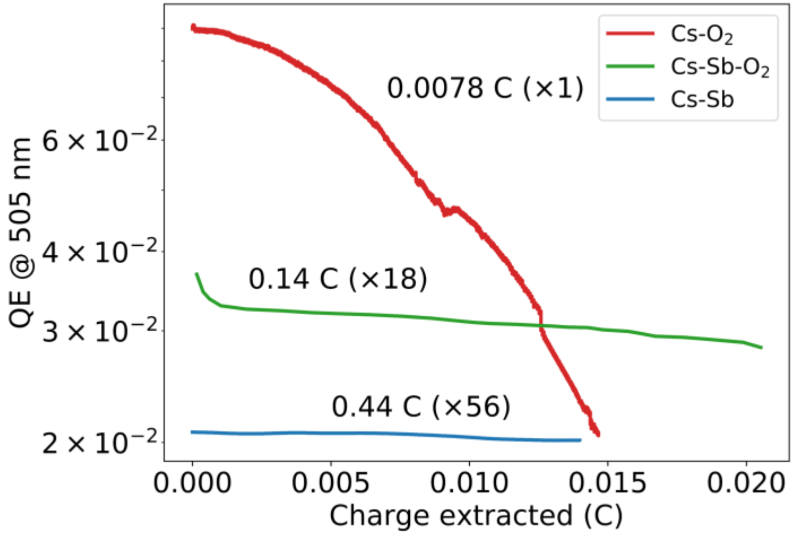
Obvious damage

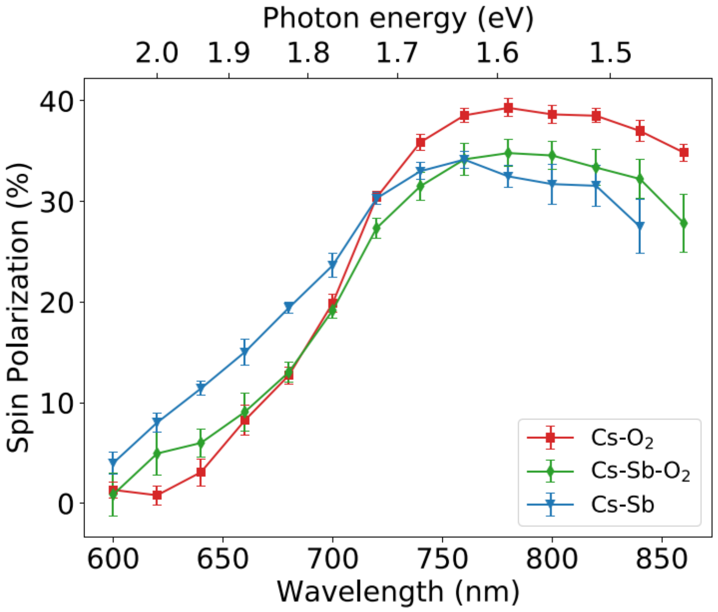
- Why non-uniform?
- Heat cycle vs. implantation role?

Phys. Rev. ST Accel. Beams 15, 063501 (2012)

# Robust Activation for Lifetime Enhancement: Sb-Cs-O J.K. Bae, Cornell

- Typical Photocathode activation: Cs & O
  - Sensitive to vacuum conditions
- Antimony activations goals
  - Reduce vacuum sensitivity
  - Increase lifetime
  - Preserve QE and polarization
- Cs, Sb and O




Cornell measurements

- Low voltage test stand
  - Vacuum not optimal
- Strained Superlattice and Bulk GaAs tested

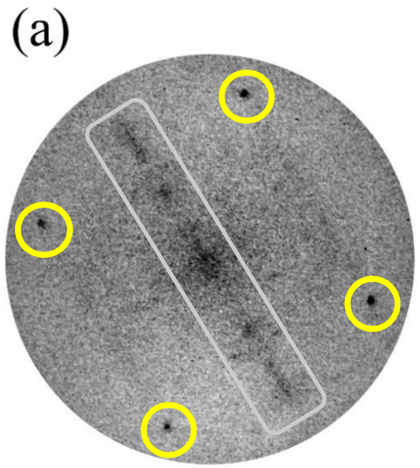
JLab collaboration

- High Voltage UITF tests
  - Polarization
  - Lifetime
  - Extreme High Vacuum
  - 200 keV Mott Polarimeter

**Jai Kwan Bae, Cornell**

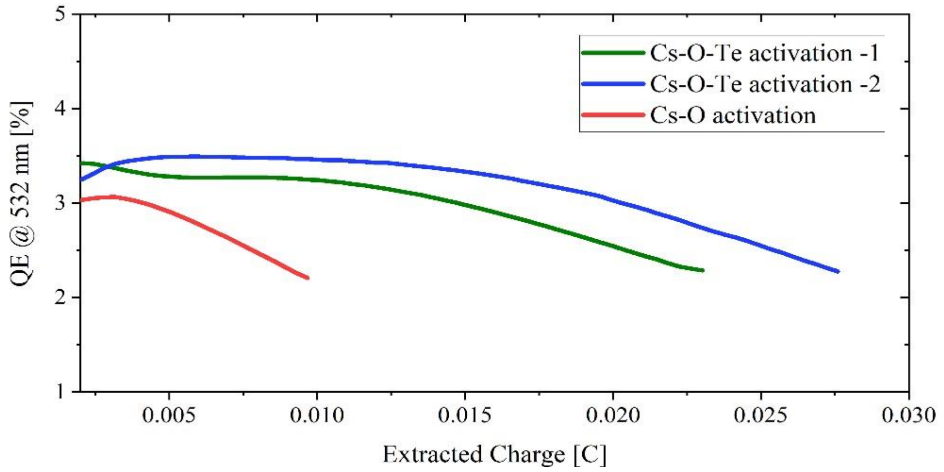
Bae *et al, J. Appl. Phys* **127**, 124901 (2020)

# Robust Activation for Lifetime Enhancement: $Cs_2Te$, J. Biswas, BNL

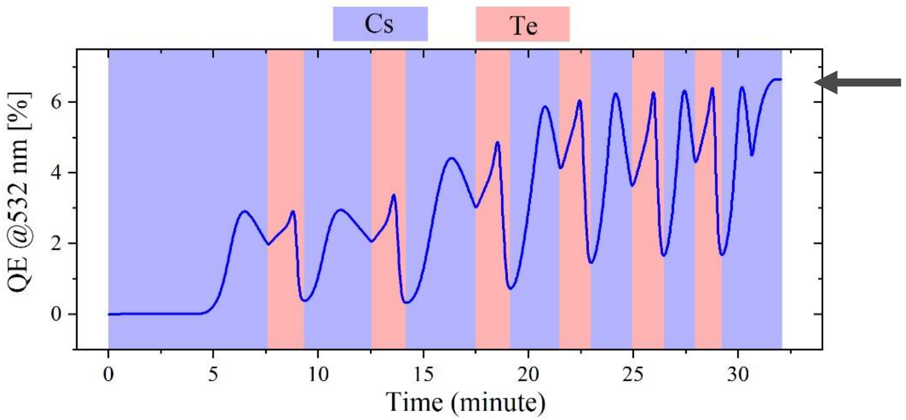



(1x1), & defused (4x6) reconstruction

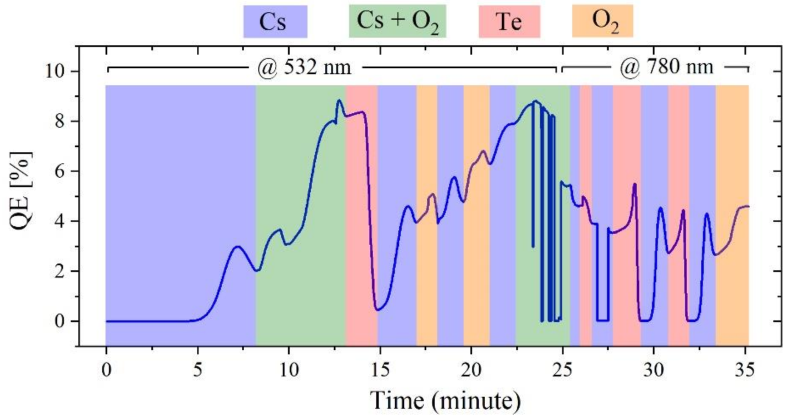
Oxide desorption – LEED




Longer charge lifetime in a test chamber as compared to Cs-O/GaAs




Cs-Te activation on GaAs




Cs-O-Te activation on GaAs

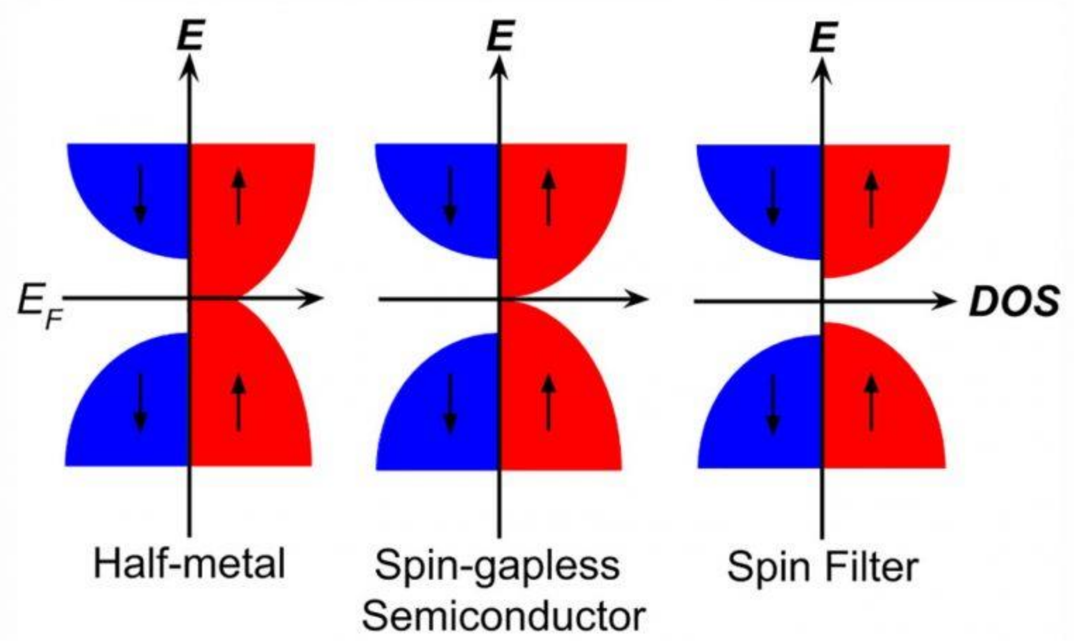
Biswas et al. AIP Advances 11, 025321 (2021); https://doi.org/10.1063/5.0026839

# Novel Polarized Photocathodes

Spintronic materials

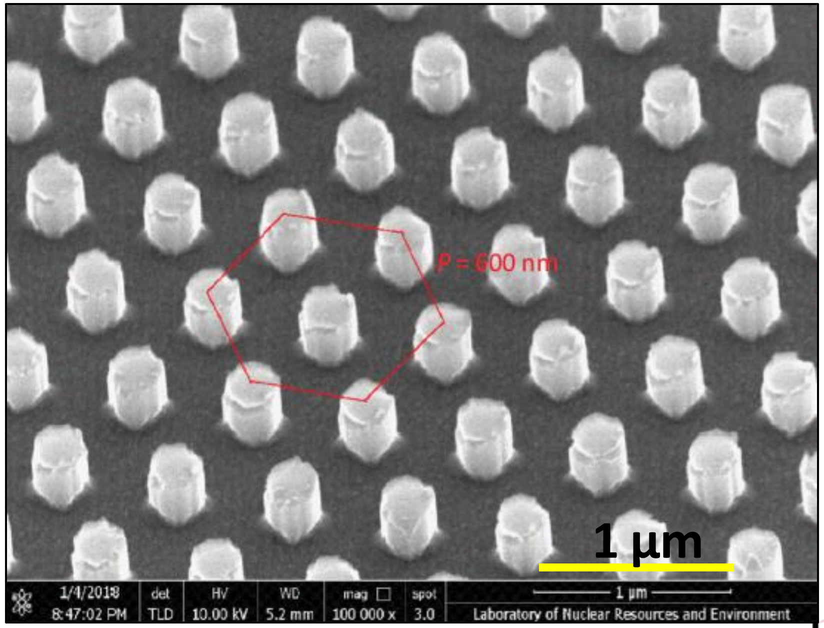
- Half metals
- Heusler alloy/ half-Heusler

- Surface plasmon polaritons to enhance emission
  Z.Li et al., Scientific Reports 7(1):44335 (2017).
- Spin filters: Euclid Techlabs SBIR projects




Nanopillar arrays: Goal: Enhanced QE

- X. Peng et al., Opt. Express **28**(2), 860-874 (2020)
- X. Peng et al., Phys. Rev. Applied **12**, 064002 (2019)



# Conclusions

- High polarization SSL photocathodes for nuclear physics accelerators
  - Supplier issues, new growth method initiatives, looking for industry or DOE partners
- Superlattice lifetime limitations
  - Vacuum improvements, biased anode, robust activations
- Novel photocathode materials
  - Spin filters, half-metals, nano-pillar arrays

Jefferson Lab
**C**enter for
**I**njectors and
**S**ources

Questions?

email: marcy@jlab.org