Modern cathode materials for LCLS-II HE

And other applications

John Smedley

Thanks to J. Lewellen, T. Xu, M. Gaowei, J. Maxson, R. Tromp, T. Vecchione, the MSU gun team and the cathode teams at ASU, BNL, Cornell, HZDR, LANL and SLAC

Outline

LCLS-II and LCLS-II-HE

The SLAC/FRIB/HZDR/Argonne SRF gun development program

Cathodes – the first frontier of beam quality

- Three Step Model
- Cathode R&D – towards single crystal, low MTE cathodes
  - Cesium Antimonide Family
  - Cesium Teluride
- Epitaxy!

Other Cathode applications

- Ultrafast Electron Diffraction
- Optical Near Field Electron Microscopy

Injector

Cryoplant

SC Linac

NC Linac

Spreader

Bypass

Beam Transport

Undulators &
X-Ray Transport

X-Ray Experimental Area
(NEH and FEH)

LCLS-II

BERKELEY LAB

Fermilab

Argonne
NATIONAL LABORATORY

Jefferson Lab

CORNELL UNIVERSITY
FOUNDED A.D. 1865

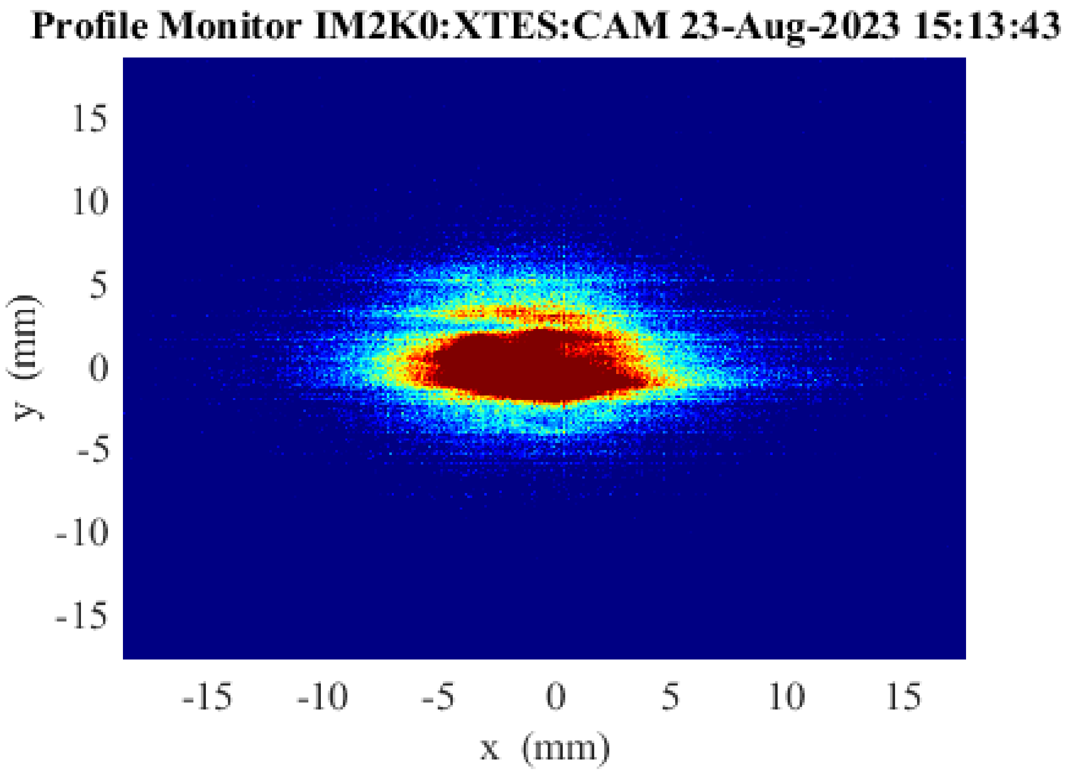
SLAC NATIONAL ACCELERATOR LABORATORY

CD-0: 2010
TTO: 2023
→ 13 years

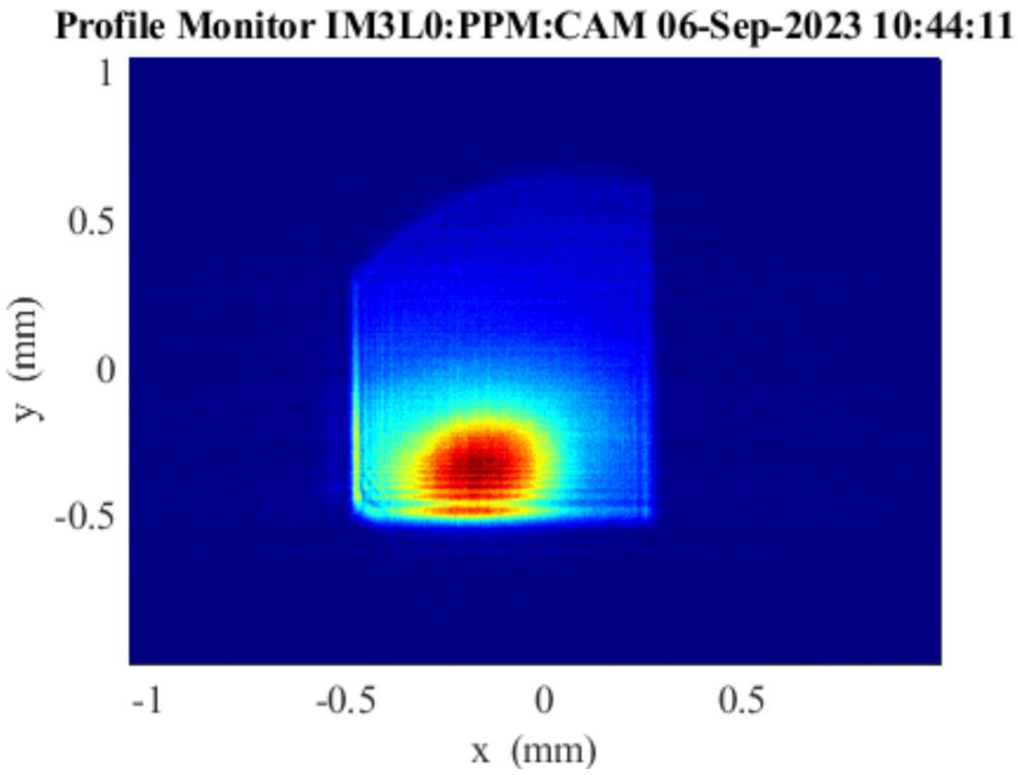
~ 5M hours
~ $1B

LCLS-II First Light

8/23/23
Soft X-ray Undulator, 450 eV

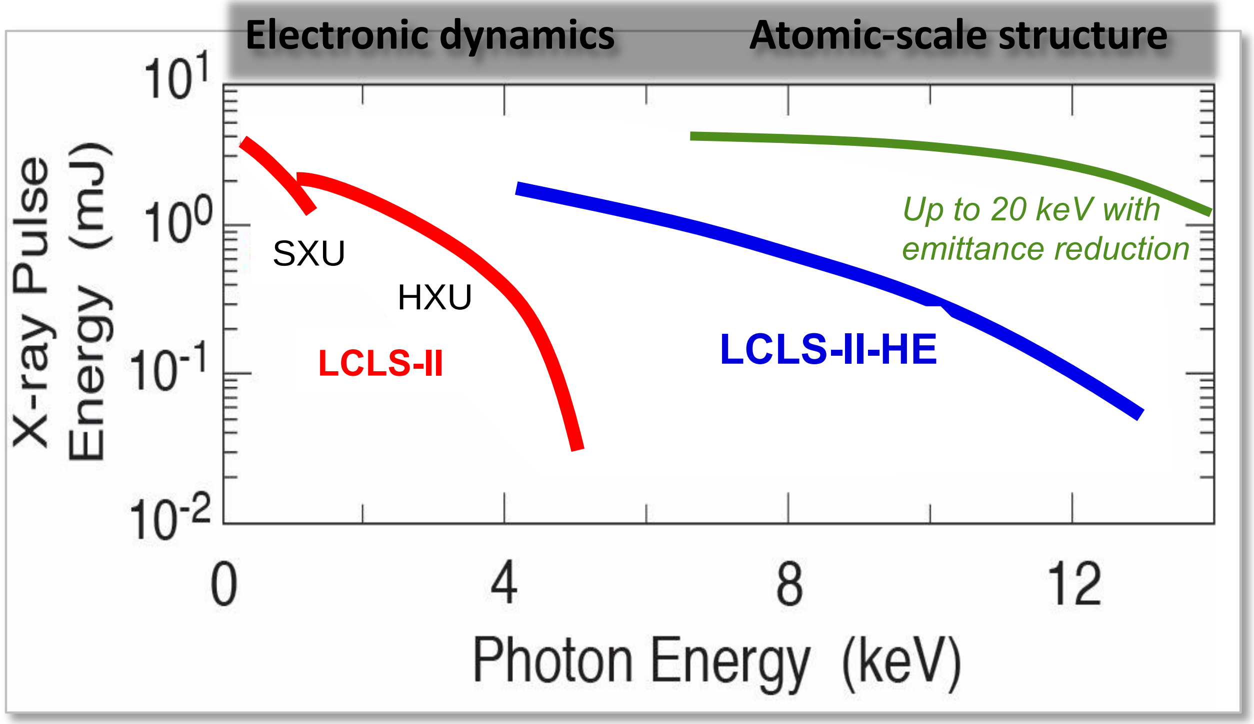


9/6/23
Hard X-ray Undulator, 1050 eV



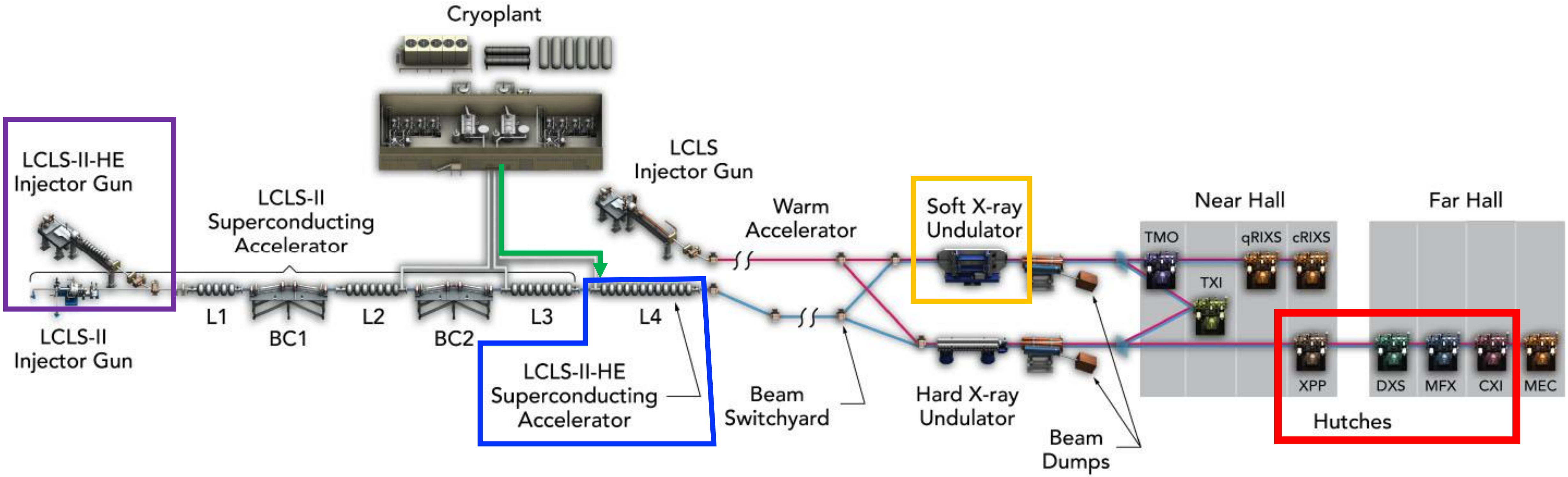
LCLS-II-HE Mission

Deliver the ability to observe and understand the structural dynamics of complex matter at the atomic scale with hard x-rays, at ultrafast timescales, and in operational environments.



LCLS-II-HE provides high-rate, FEL radiation at Ångstrom wavelengths

LCLS-II-HE Overview



1. Add 23 additional cryomodules (L4 linac) to double the LCLS-II accelerator energy: 4 GeV to 8 GeV
2. Install new cryogenic distribution box and transfer line between the cryoplant and the new L4 linac
3. New long period soft X-ray undulator
4. Upgrade the LCLS hard X-ray instruments for MHz beam and data rates
5. Design low-emittance injector and SRF gun for extended hard X-ray performance

Preliminary Key Performance Parameters

Performance Measure	Threshold	Objective
Superconducting linac electron energy	7 GeV	8 GeV
Electron bunch repetition rate in linac	93 kHz	929 kHz
Charge per bunch in SC- linac	0.02 nC	0.1 nC
Photon energy range	250 – 8,000 eV	250 to ≥ 20,000 eV
High rep-rate-capable HXR instruments	≥ 1	≥ 4
FEL photon quantity per bunch (10^{-3} BW)	5×10^{8} (50× spont. @ 8 keV)	> 10^{11} @ 8 keV (200 μJ) AND > 5×10^{9} @ 20.0 keV (20 μJ)

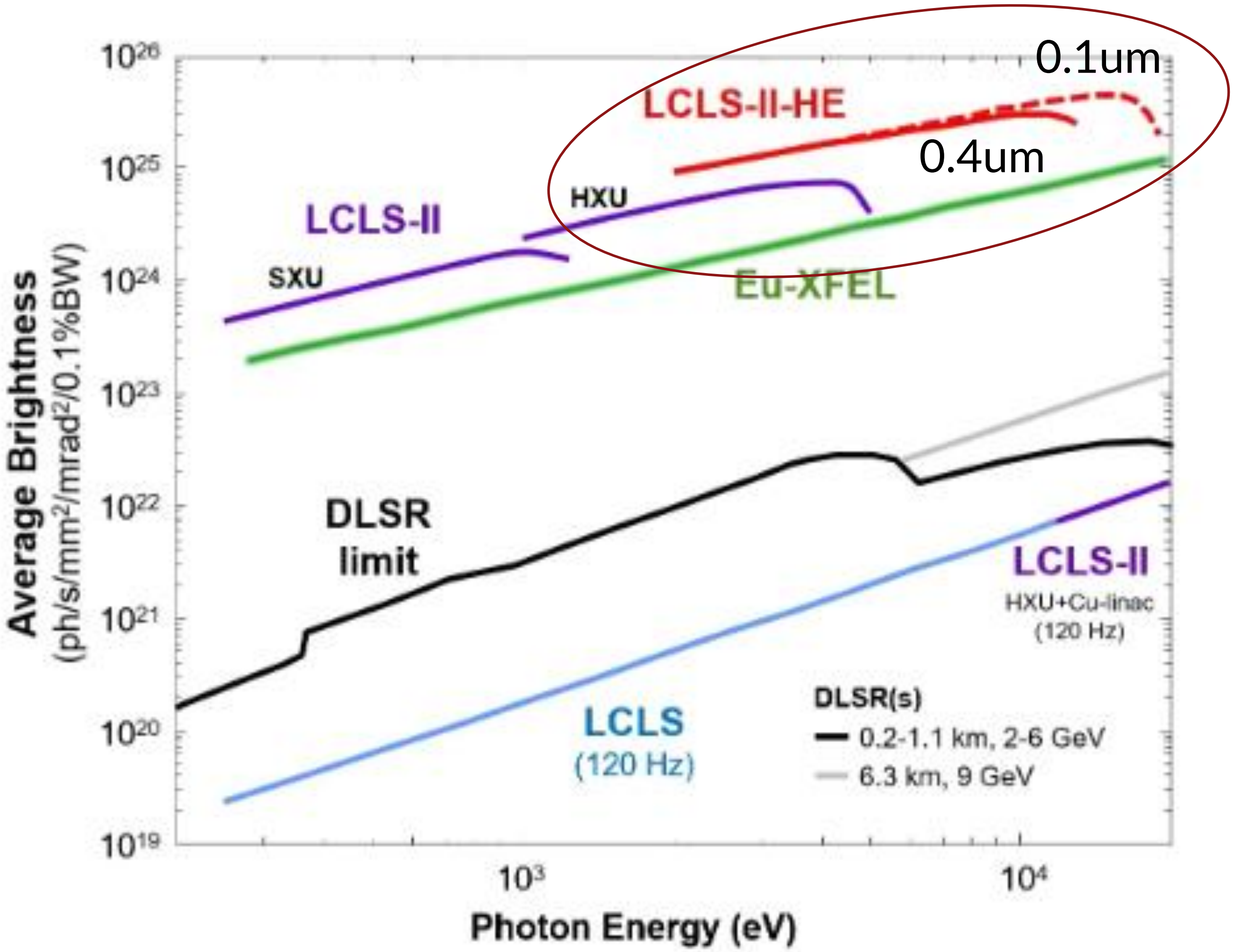
Motivation for the Low Emittance Injector (LEI)

Provide lower emittance beams to extend photon energy reach, enabling a broader photon physics program

- Less costly than increasing beam energy, which is currently limited by space constraints

Provide redundant electron source

- Qualify second injector while current one operates
- Once online, could quickly revert to original injector if problems occur
- In the interim, a spare LCLS-II gun with modest improvements is being built



Electric field on cathode 30 MV/m is required to achieve 0.1 um emittance.

Parallel effort to develop an SC gun and injector

Step 1: Develop low emittance, high-gradient SC gun

- Design, build and test a ~ 186 MHz quarter-wave SRF gun over 3 years
- Deliverable is to demonstrate 30 MV/m gradient performance
- Development cost: $10M

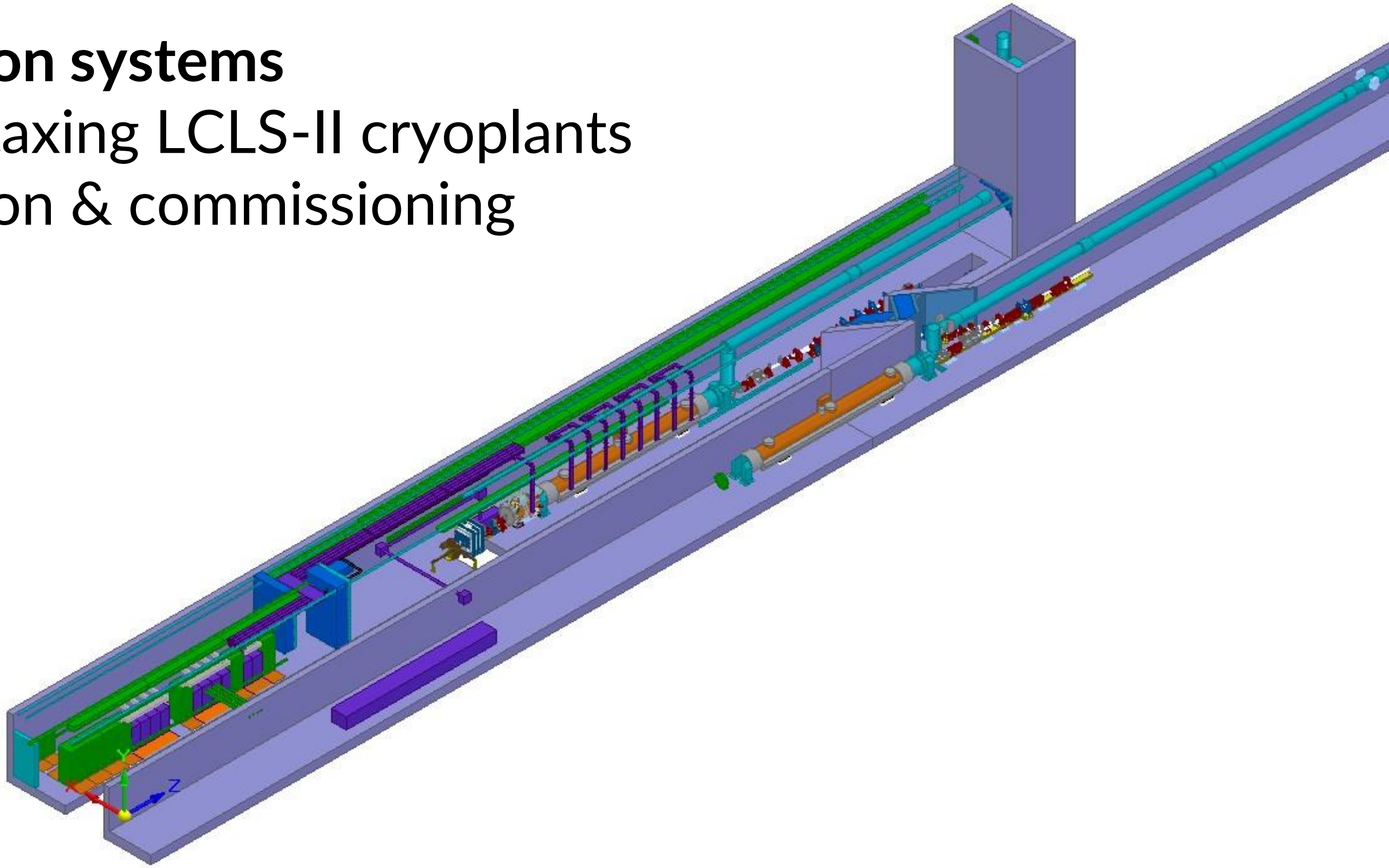
Step 2: New injector and tunnel for LCLS-II-HE

- Expand project scope to include SC gun, buncher cavities, beamline, and new injector tunnel.
  - Provides infrastructure to commission a new gun without interruption to science operations
  - Provides spare gun capability
  - Enhances science program with pump-probe operation capability
- Build SRF gun based on lessons from Step 1

A 2-step process is needed to demonstrate the gun technology before inclusion into the project scope.

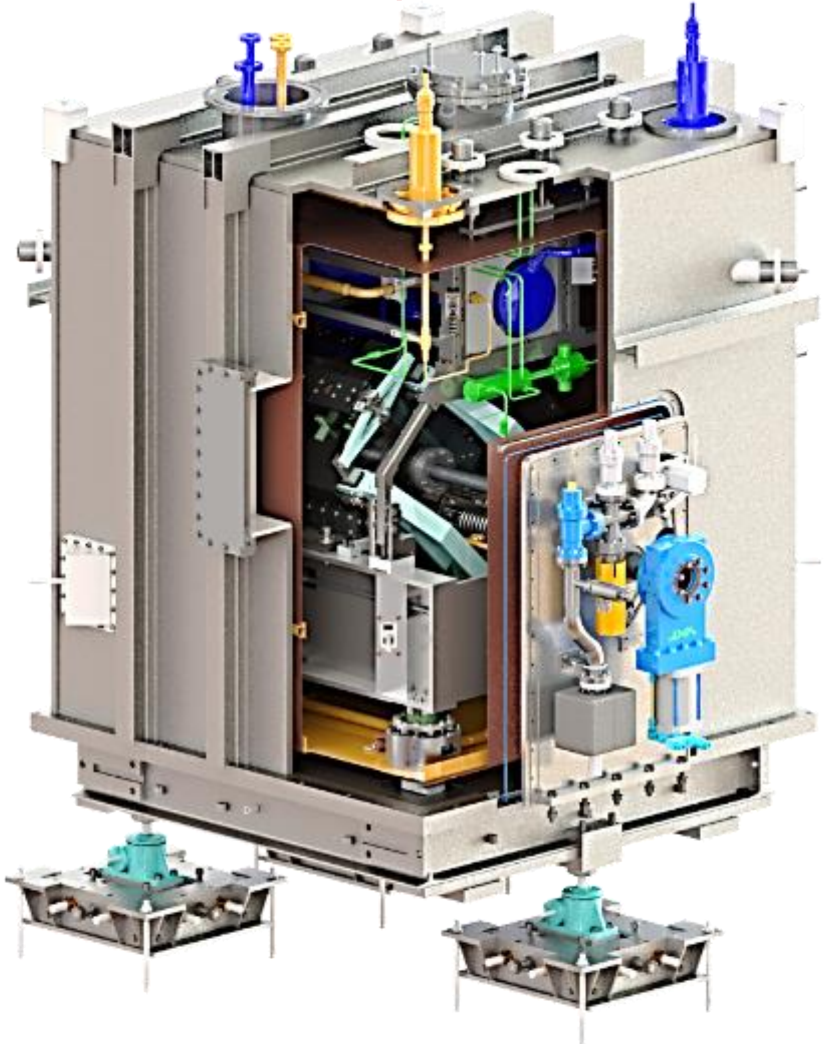
Injector Scope – currently contingency scope

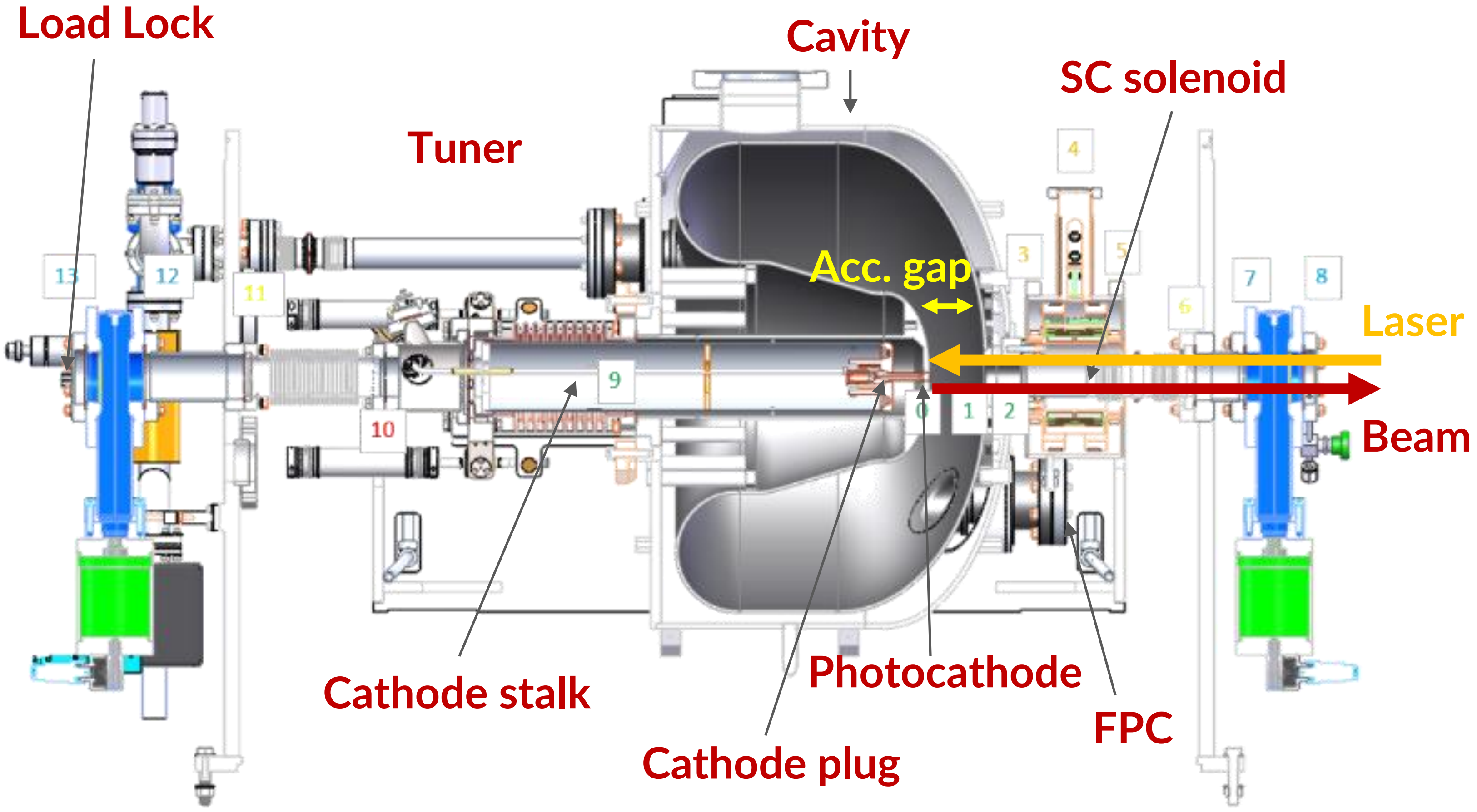
- **New injector tunnel adjacent to the existing accelerator housing**
  - New tunnel at grade-level
  - Enables independent and parallel operation
- **Separate cryoplant and distribution systems**
  - Increases performance with taxing LCLS-II cryoplants
  - Enables independent operation & commissioning
- **High brightness injector**
  - SRF gun
  - NC and SRF buncher cavities
  - L0’ cryomodule
  - Diagnostics beamline
  - Local e^- dump



SRF Gun Cryomodule

Cryomodule

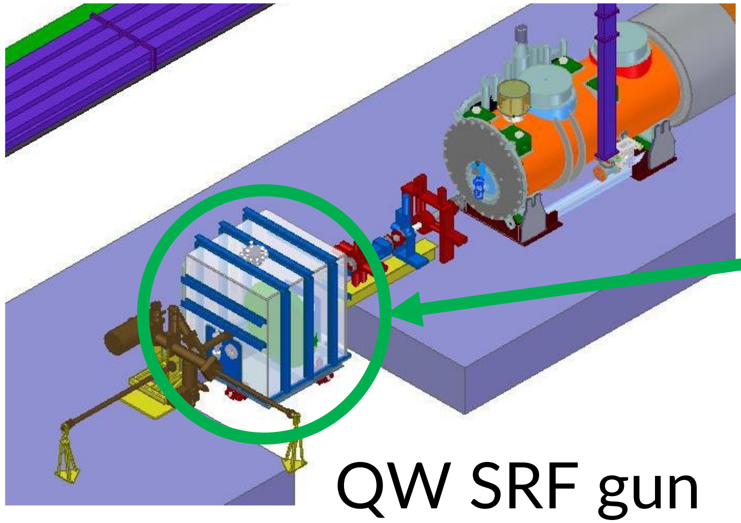




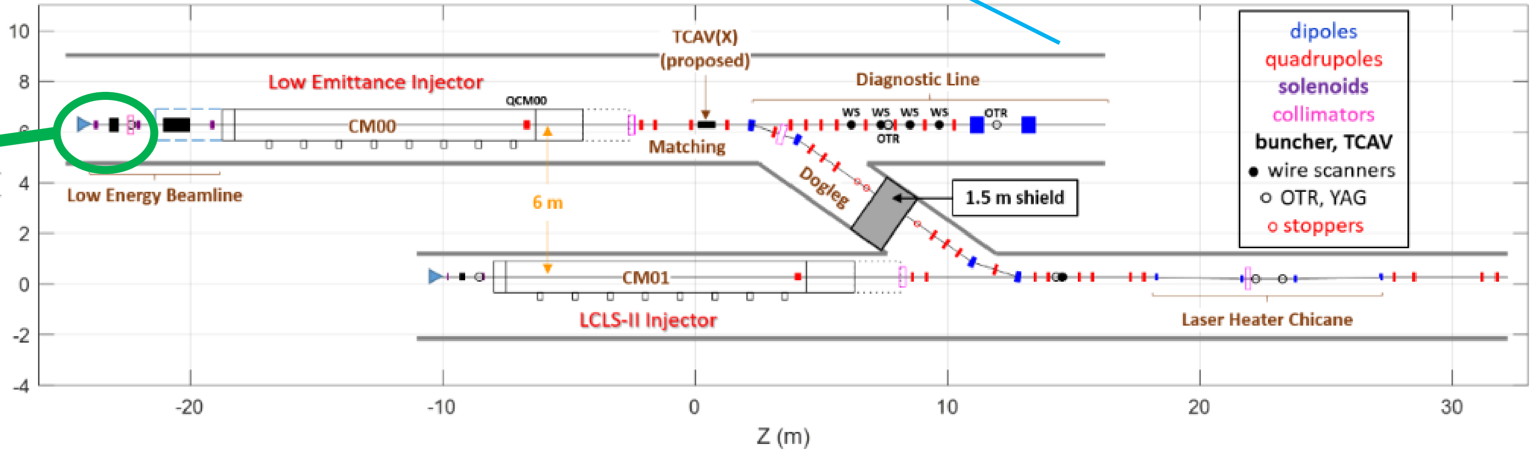
What do we need from the cathode?

LCLS-II-HE *may* install a new, low-emittance injector in a new tunnel at the west end of the SLAC linac. Goal: $\varepsilon_{n,rms,95\%} < 0.1\ \mu m$ @ 100 pC

- 30 MV/m on cathode
- MTE < 184 meV
- Lifetime > 1 week
- “green” illumination



QW SRF gun



Semiconductor Photocathodes

The primary path to high average current in photoinjectors

The good points:

- QE can be >10%
- Many use visible light
- Polarized cathodes possible

However:

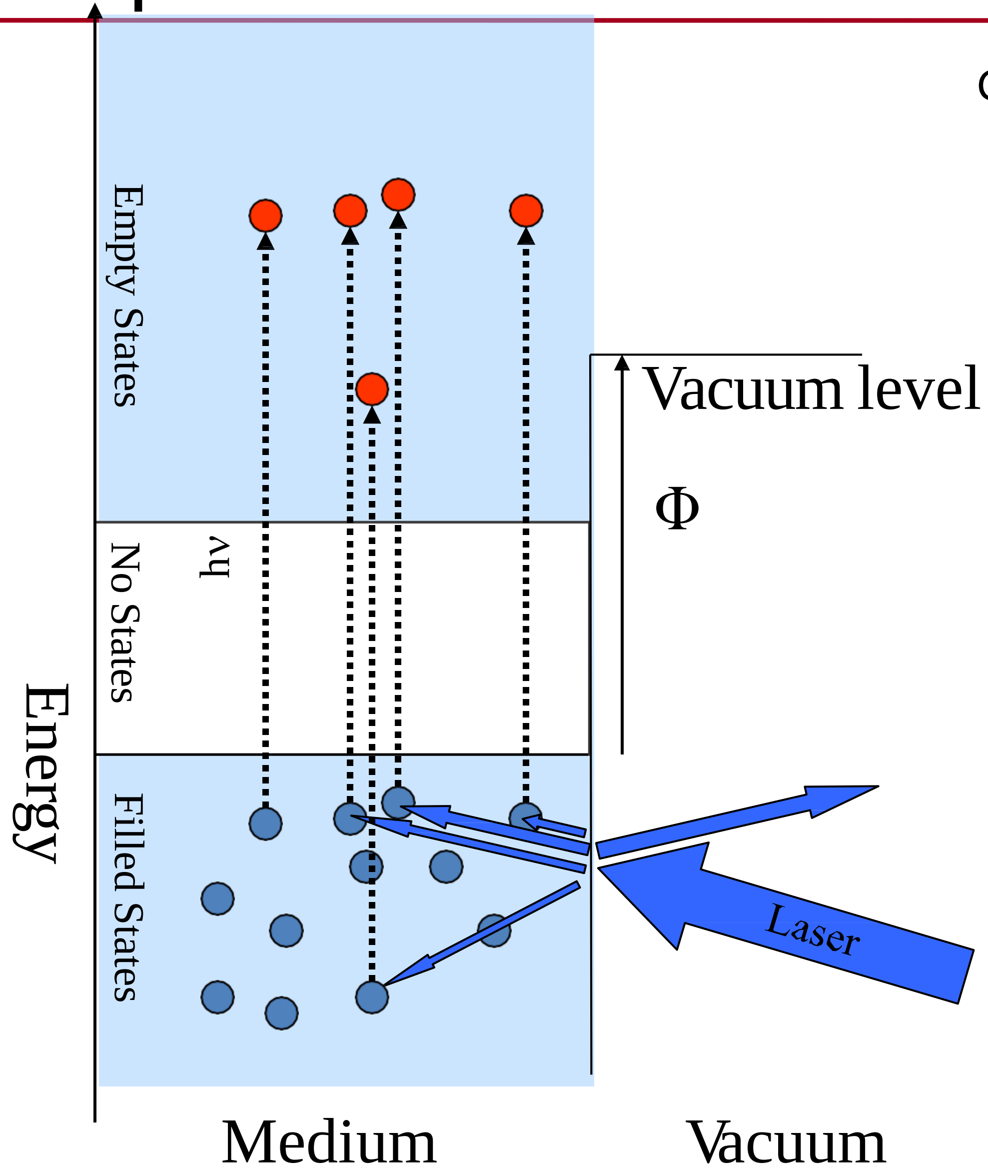
- Require UHV (<0.1 nTorr)
- Limited Lifetime
- Response time
- Complicated

Common types:

- Cs_2Te – QE ~7% @ 262 nm, Lifetime ~1yr
- K_2CsSb – QE >4% @ 532 nm, Lifetime ~weeks
- Cs:GaAs – QE ~0.5% @ 800 nm (polarized), >6% @ 527 nm

W.E. Spicer & A. Herrera-Gomez, Modern Theory and Applications of Photocathodes, SLAC-PUB-6306 (1993)

Three Step Model of Photoemission - Semiconductors



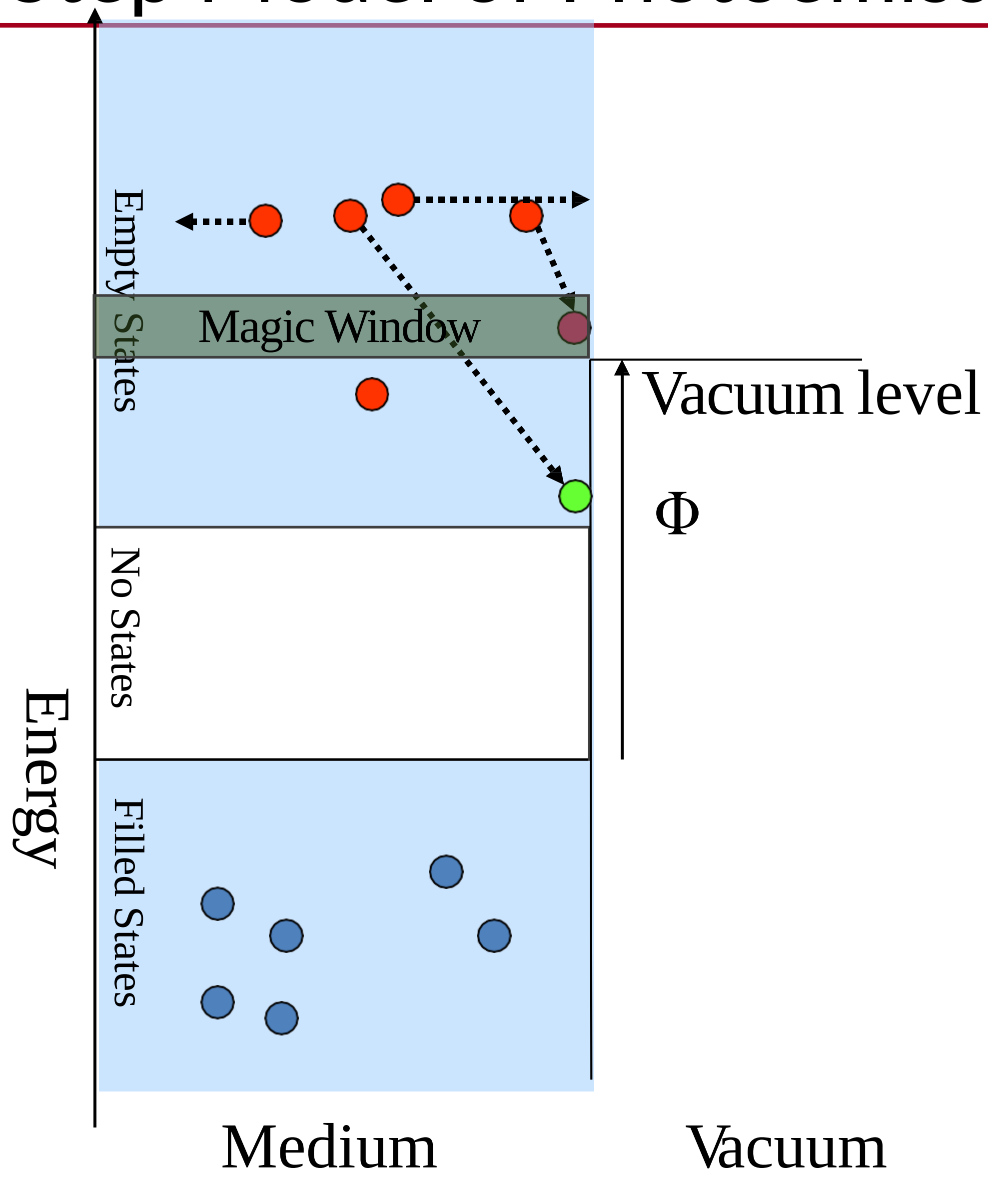
Goal: understand the physics of
electron excitation and emission

1) Excitation of e$^-$

Reflection, Transmission

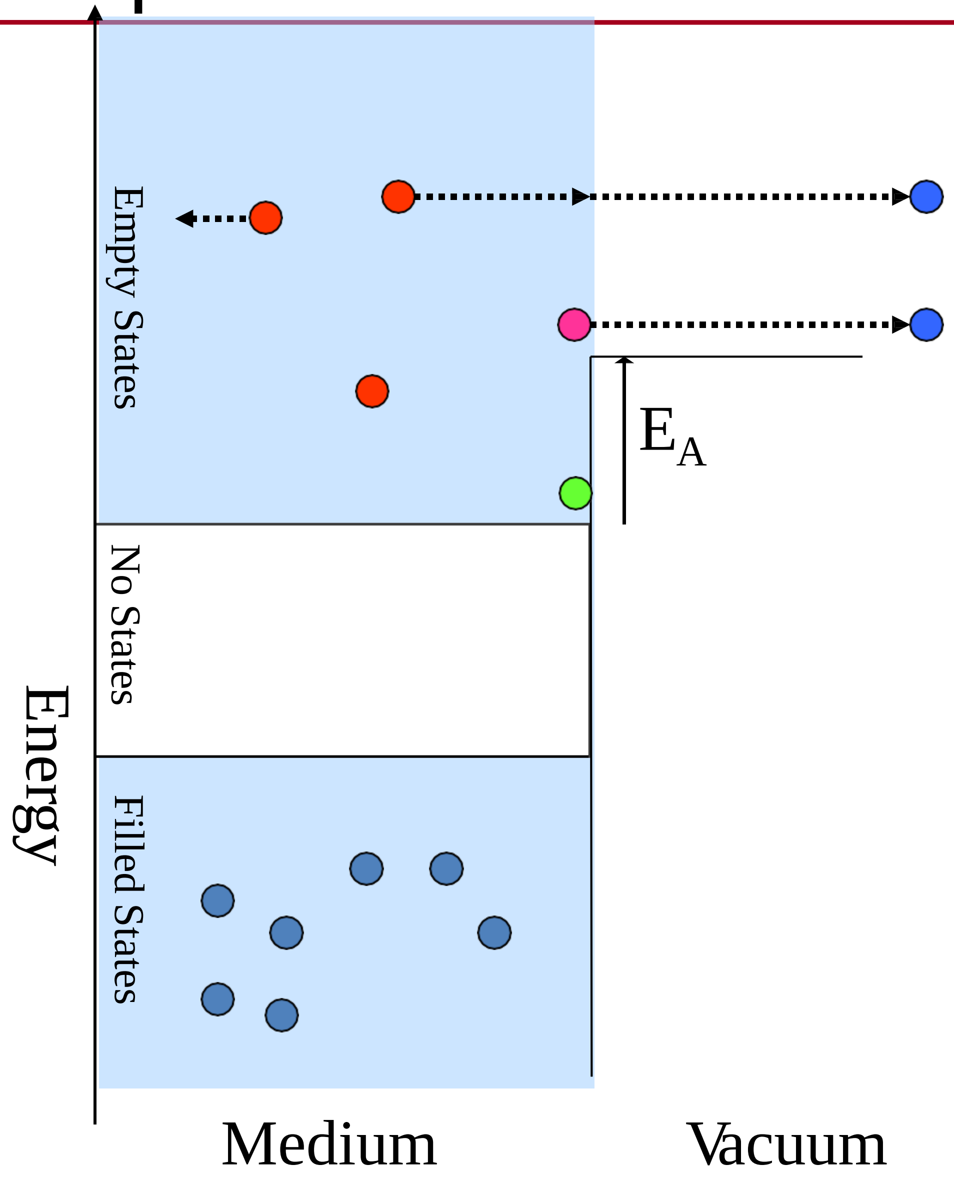
Energy distribution of excited e$^-$

Three Step Model of Photoemission - Semiconductors



1) Excitation of e^-
   - Reflection, Transmission
   - Energy distribution of excited e^-
2) Transit to the Surface
   - e^--phonon scattering
   - e^--defect scattering
   - e^--e^- scattering
     - Random Walk

Three Step Model of Photoemission - Semiconductors

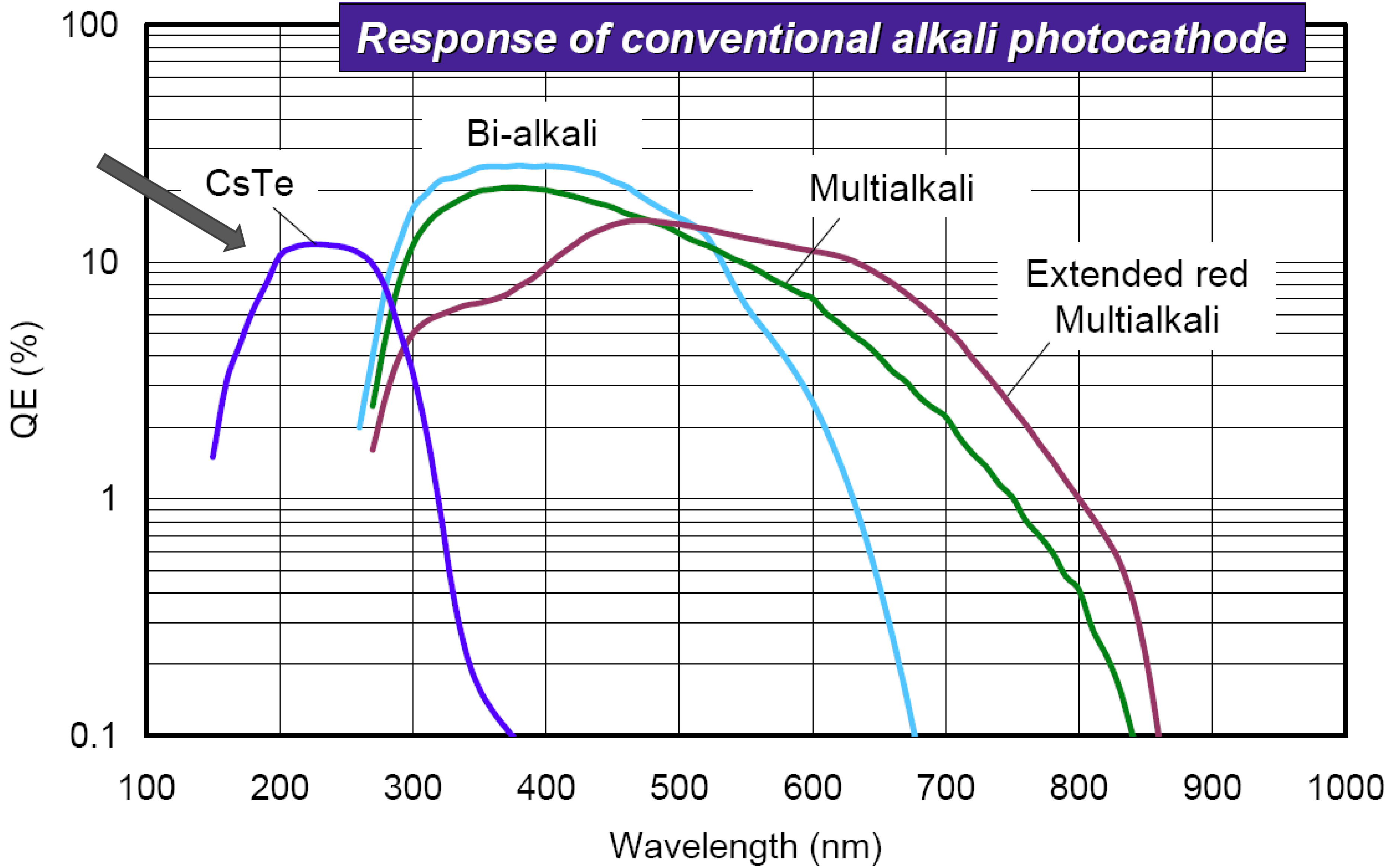


1) Excitation of e$^-$
   - Reflection, Transmission
   - Energy distribution of excited e$^-$
2) Transit to the Surface
   - e$^-$-phonon scattering
   - e$^-$-defect scattering
   - e$^-$-e$^-$ scattering
   - Random Walk
3) Escape surface
   - Overcome Electron affinity
   - Surface termination impacts
   - Multiple tries

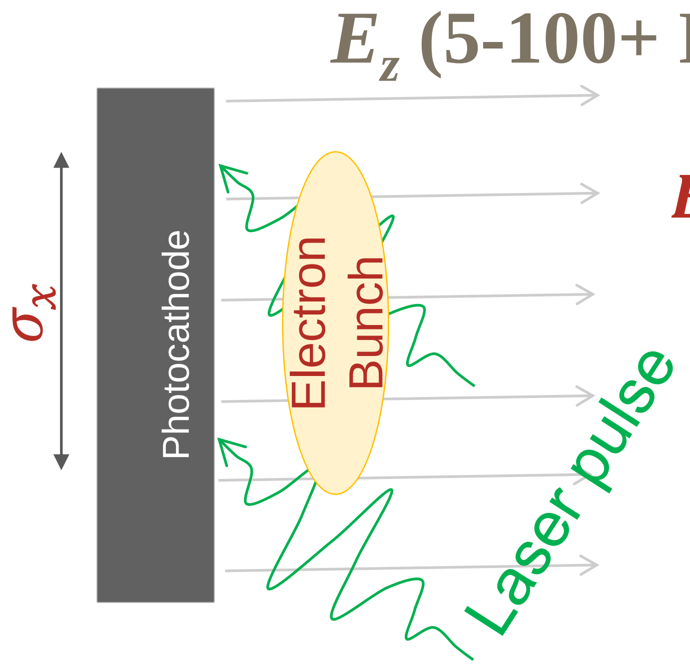
Need to account for Random Walk in cathode suggests Monte Carlo modeling

Integrating probabilities at each step provides framework to predict QE

Response of conventional alkali photocathode



Maximum Achievable Brightness



I. Bazarov *et al.*, Phys. Rev. Lett. **102**, 104801 (2009)

Child-Langmuir law

$$B_{4D} \propto \frac{N_{bunch}}{\varepsilon_n^2} \quad \text{where } N_{bunch} \propto \sigma_x^2 E_z \quad \propto \frac{E_z}{\mathrm{MTE}}$$

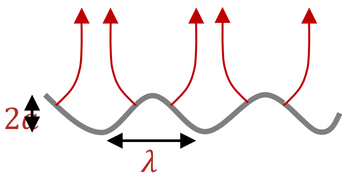
$$\varepsilon_n = \sigma_x \sqrt{\frac{\mathrm{MTE}}{m_e c^2}}$$

$$\mathrm{MTE} = \frac{1}{2} m v_\perp^2$$

Brightness limited by photocathode MTE

MTE limiting factors -Surface non-uniformities

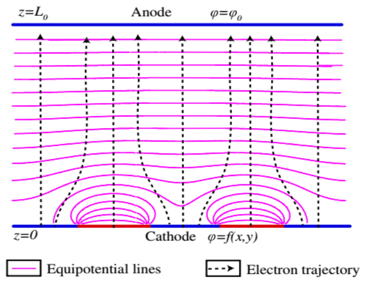
Physical Roughness



$$\mathrm{MTE}_{field} = \frac{\pi^2 a^2 E_0 e}{2\lambda}$$

J. Feng, S. Karkare *et al*, J. Appl. Phys. 121, 044904 (2017)

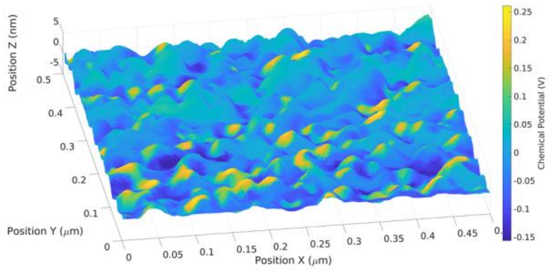
Chemical Roughness



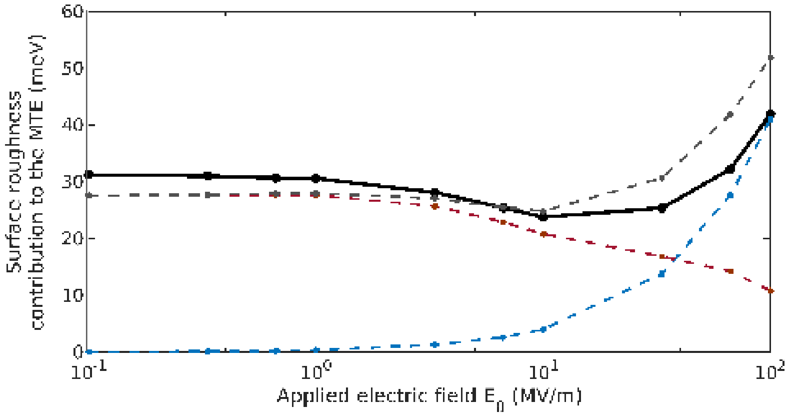
$$\mathrm{MTE}_{wf} = \frac{\pi^2 h^2 e}{4\sqrt{2} a E_0}$$

S. Karkare and I. Bazarov, Phys. Rev Applied, 4, 024015 (2015)

Combined Physical and Chemical Roughness



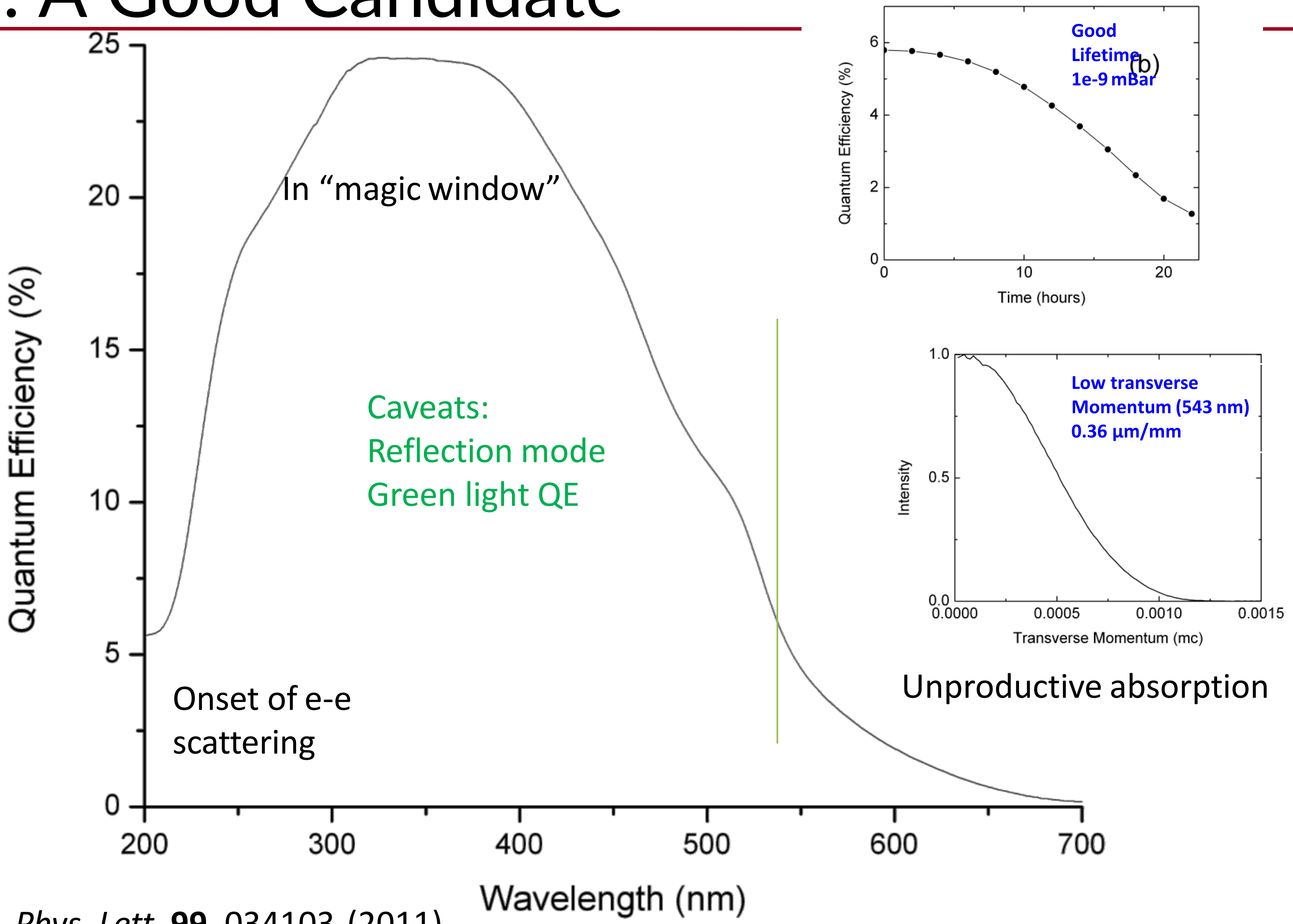
(a) AFM with KPFM surface potential



G. Gevorkyan et al. Phys. Rev. ST-Accel and Beams, 21, 093401(2018)

Need single crystal ordered surfaces to get sub-10 meV scale MTE

K_2CsSb: A Good Candidate



In "magic window"

Caveats:
Reflection mode
Green light QE

Onset of e-e scattering

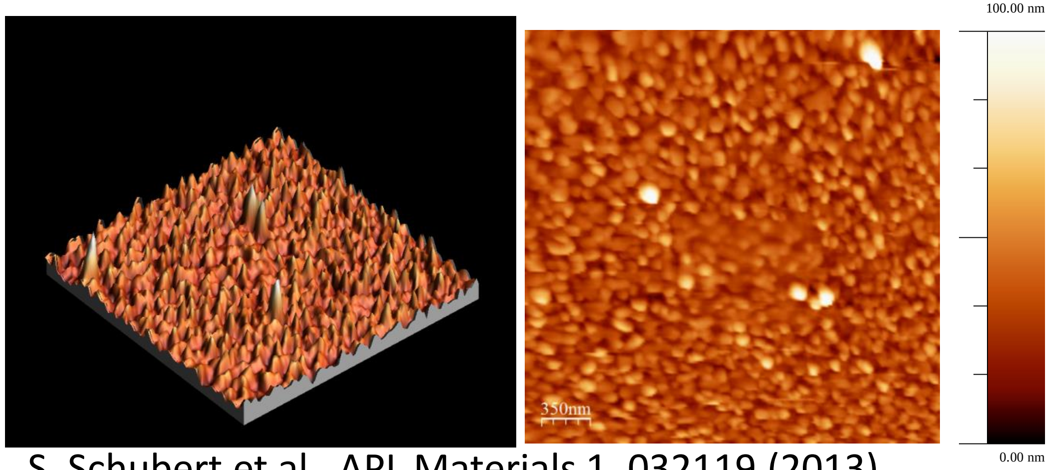
Good Lifetime 1e-9 mBar

Low transverse Momentum (543 nm) 0.36 µm/mm

Unproductive absorption

T. Vecchione et al., *Appl. Phys. Lett.* **99**, 034103 (2011)

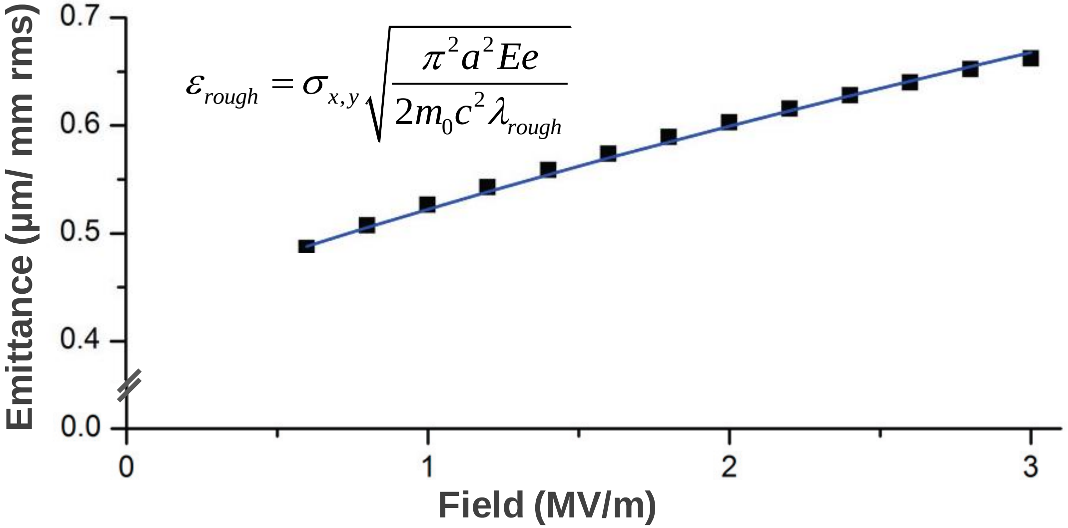
Traditional Sequential Deposition of K_2CsSb High QE and Rough Surface



S. Schubert et al., APL Materials 1, 032119 (2013)

**25 nm roughness,
100 nm spatial period**

**Emittance vs field
measured with
Momentatron, 532 nm light**



$$\varepsilon_{rough} = \sigma_{x,y}\sqrt{\frac{\pi^2 a^2 Ee}{2m_0c^2\lambda_{rough}}}$$

T. Vecchione, et al, Proc. of IPAC12, 655 (2012)

In operando analysis during growth

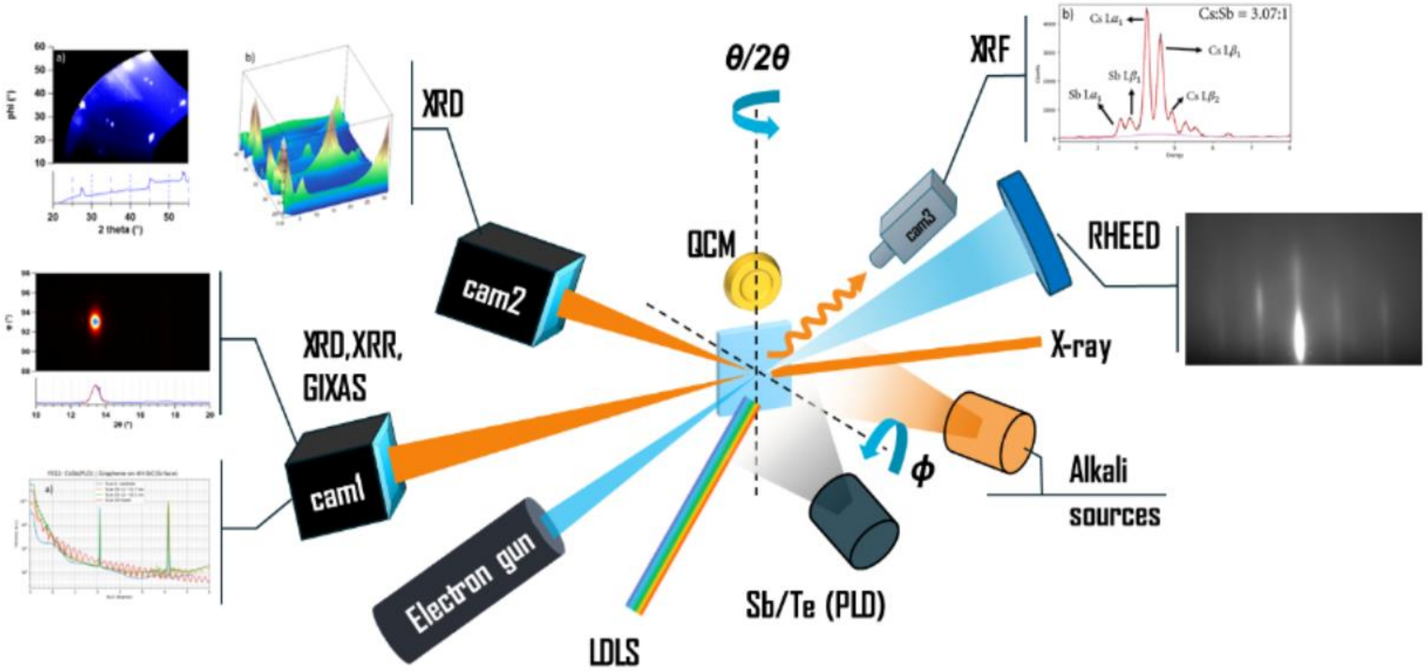
Our growth method:

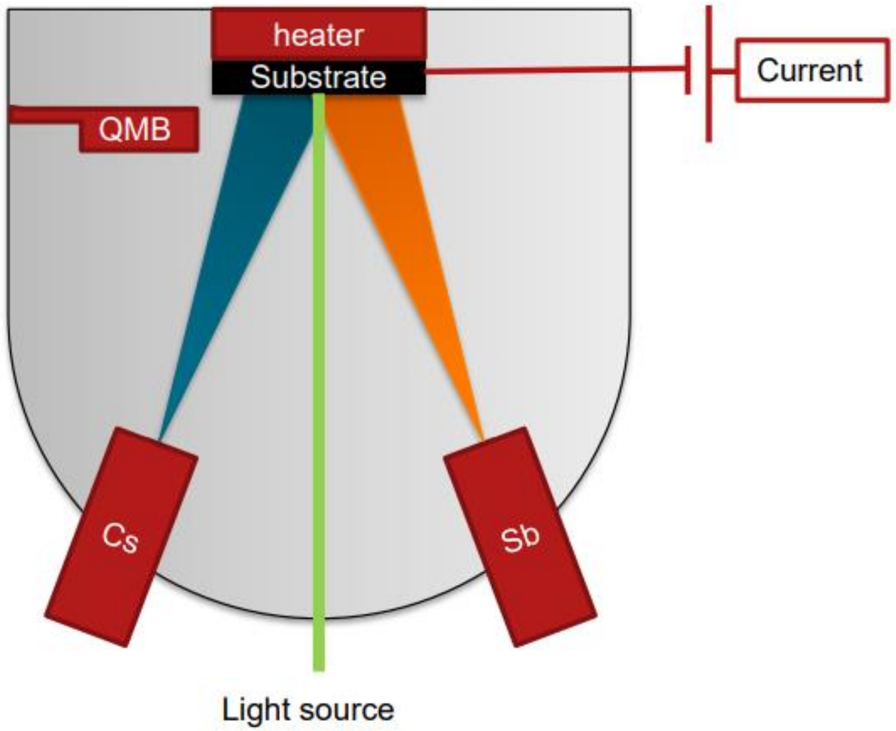
Real-time thickness, stoichiometry, XRD measurements

Conventional photocathode growth:

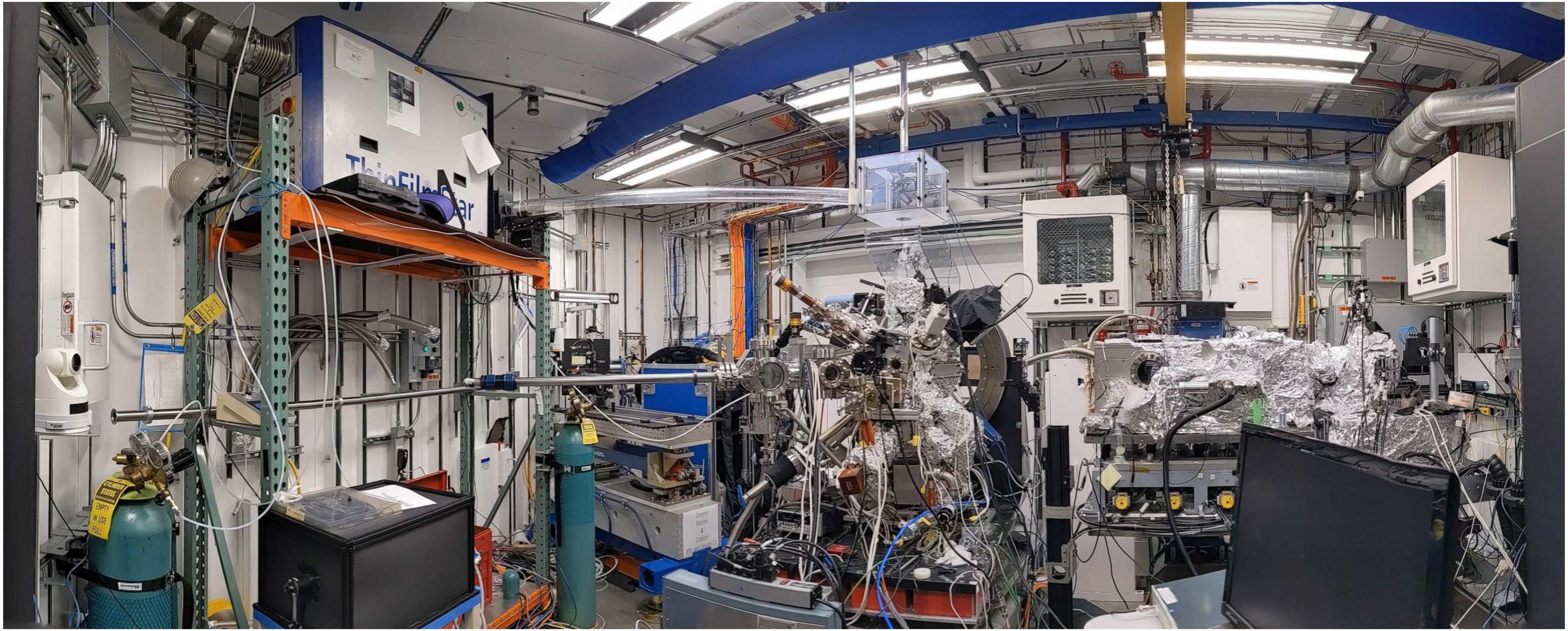
Photocurrent oriented.

Maximize quantum efficiency.



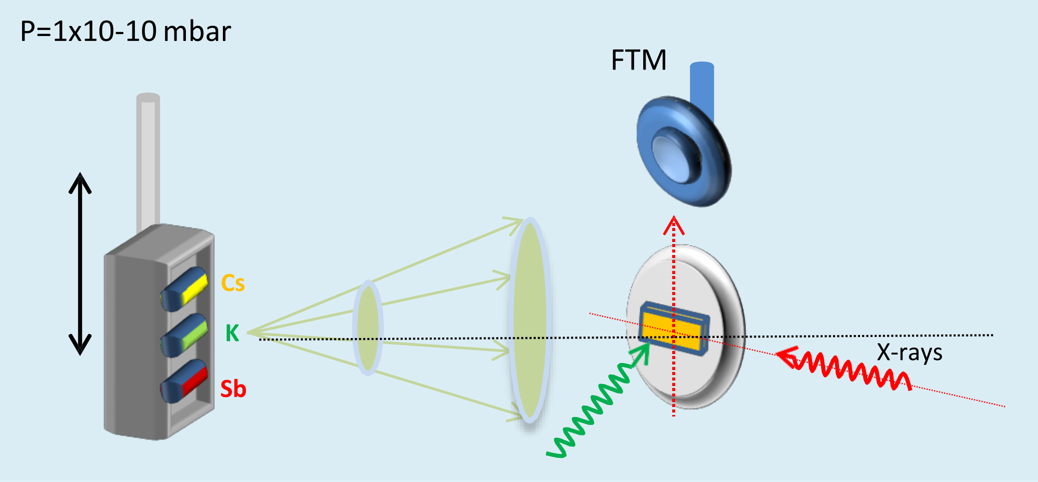


Experimental setup NSLS-II/ISR

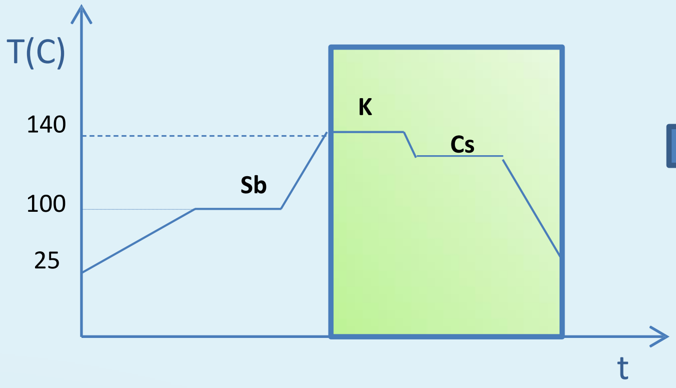


Experimental set up: K_2CsSb cathode growth

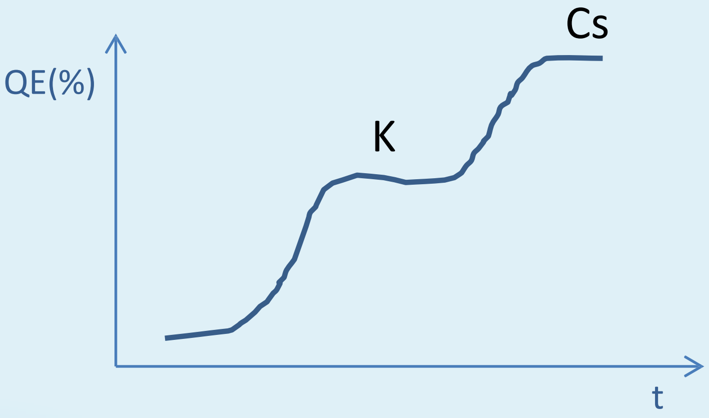
Horizontal evaporation of three sources:



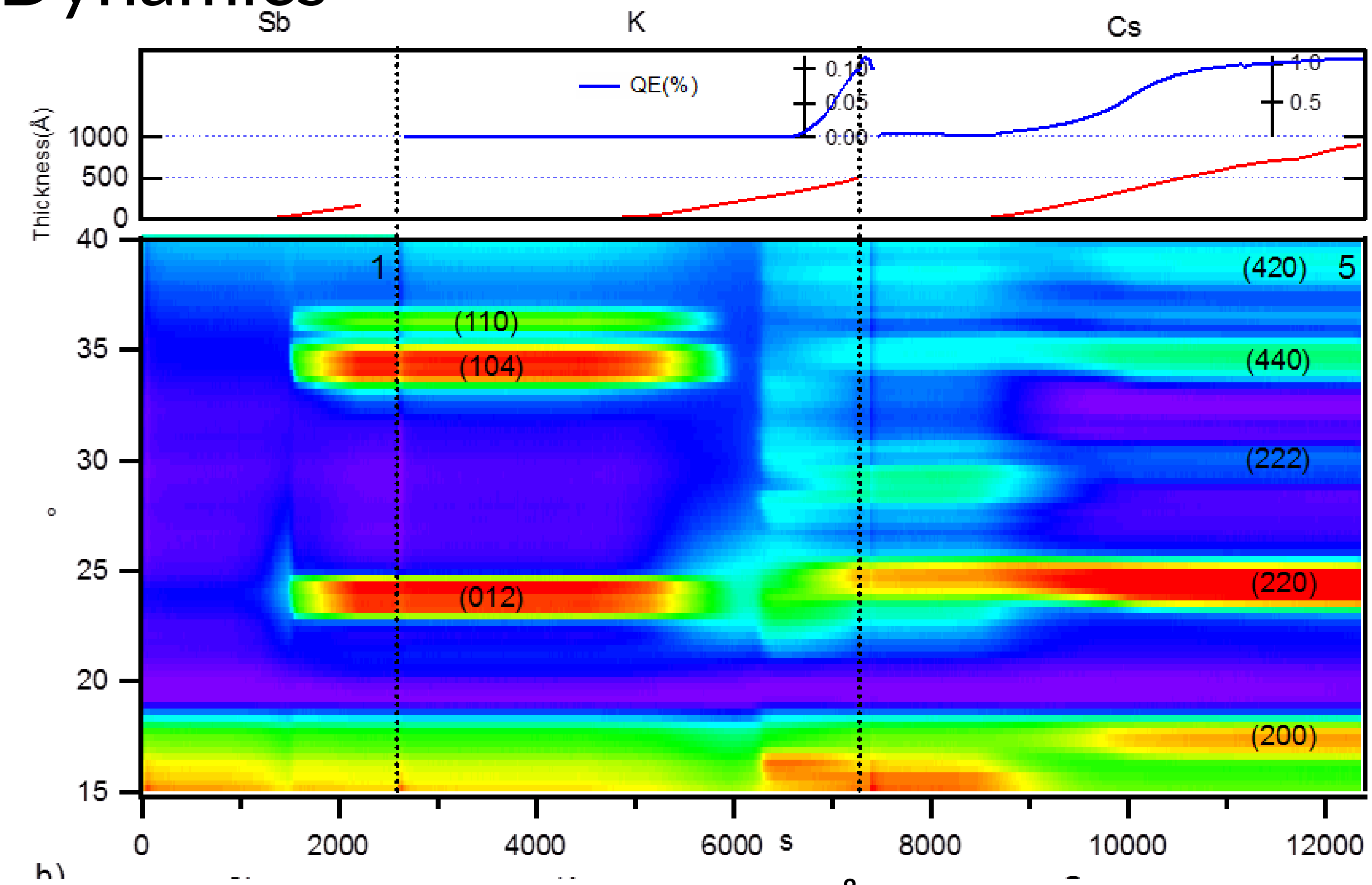
Recipe:



QE during growth (532 nm laser)



Reaction Dynamics



Antimony evaporated on Si, 0.2 Å/s; crystallize at 4nm

K deposition dissolves Sb layer - This is where roughening occurs!

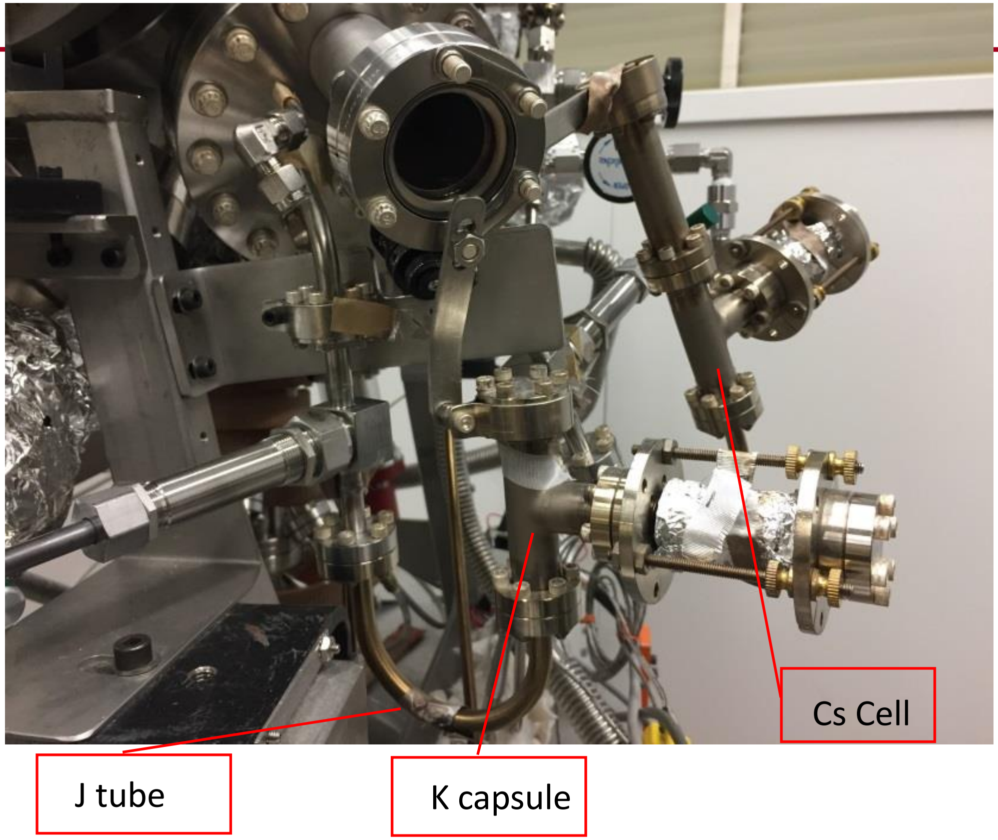
QE increase corresponds with K_xSb crystallization

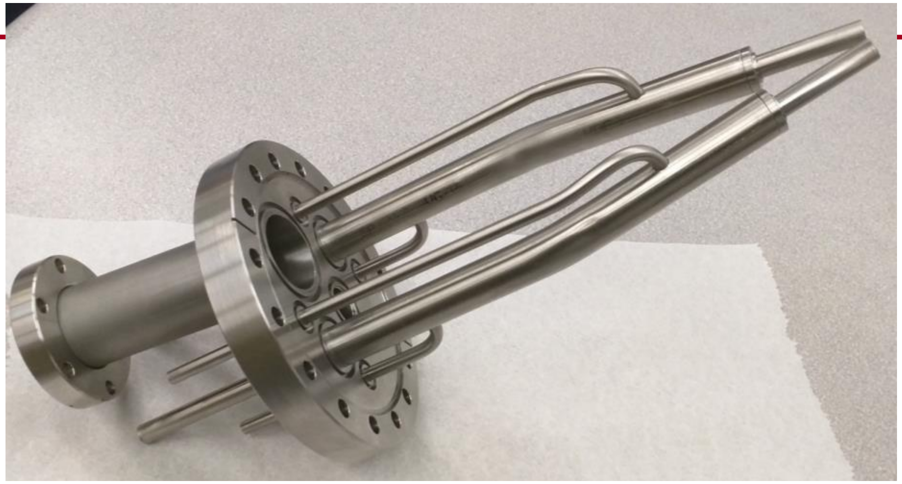
Cs increases lattice constant and reduces defects

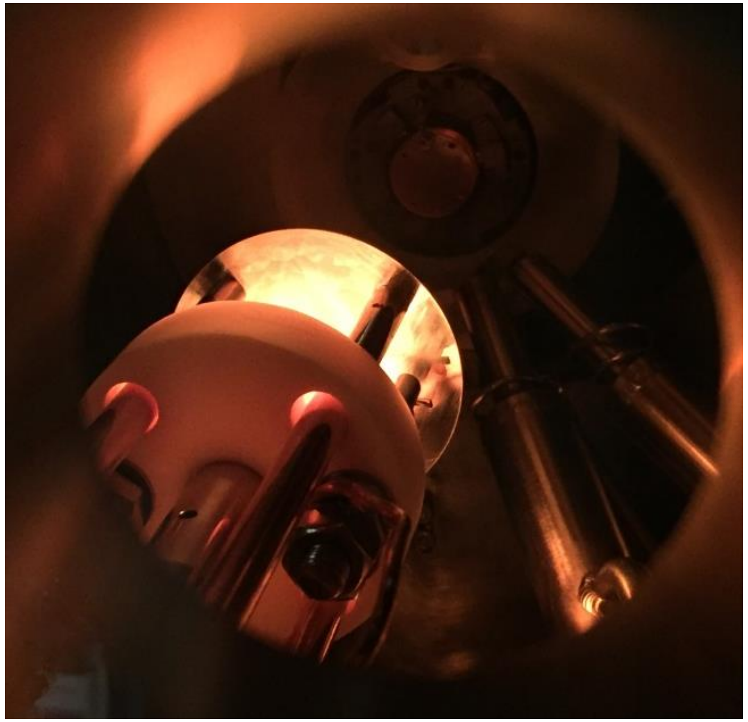
M. Ruiz-Osés et al., APL Mat. 2, 121101 (2014)

Ternary Co-evaporation

Simultaneously evaporate from Sb evaporator and K,Cs effusion cells







Growth rate are controlled by J tube temperature, valve and shutter

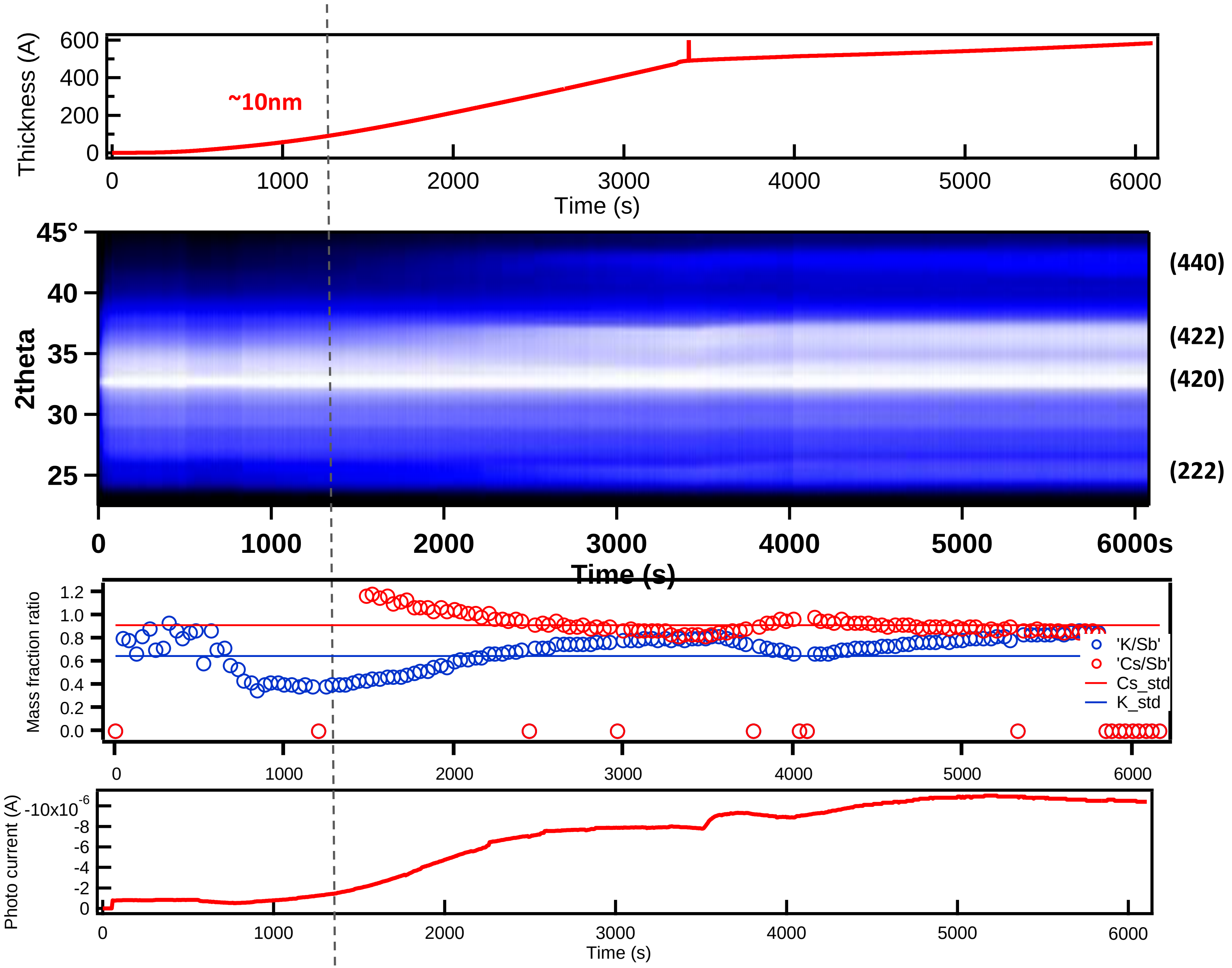
Stoichiometry controlled by real time XRF

Thickness

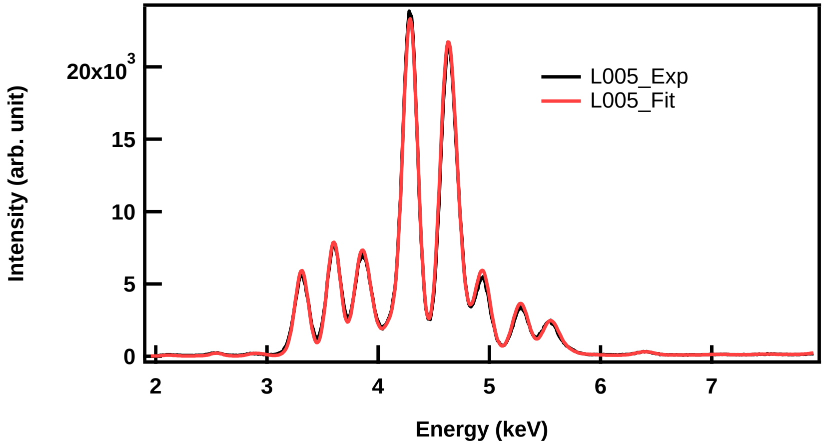
Real time XRD

Real time Fluorescence

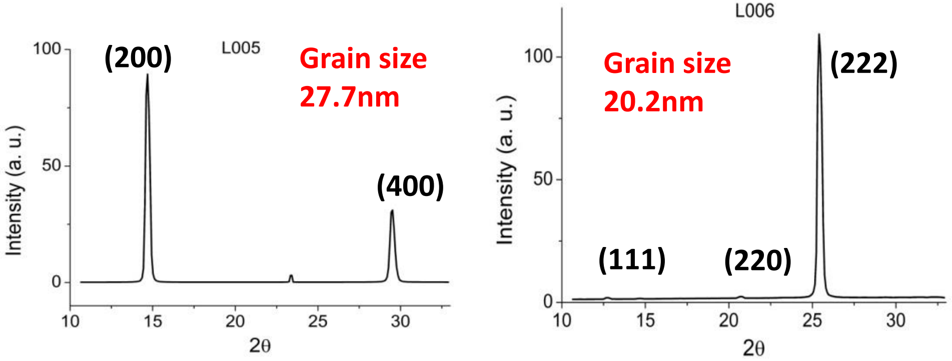
QE



Stoichiometry & Structural Analysis



Camera 1



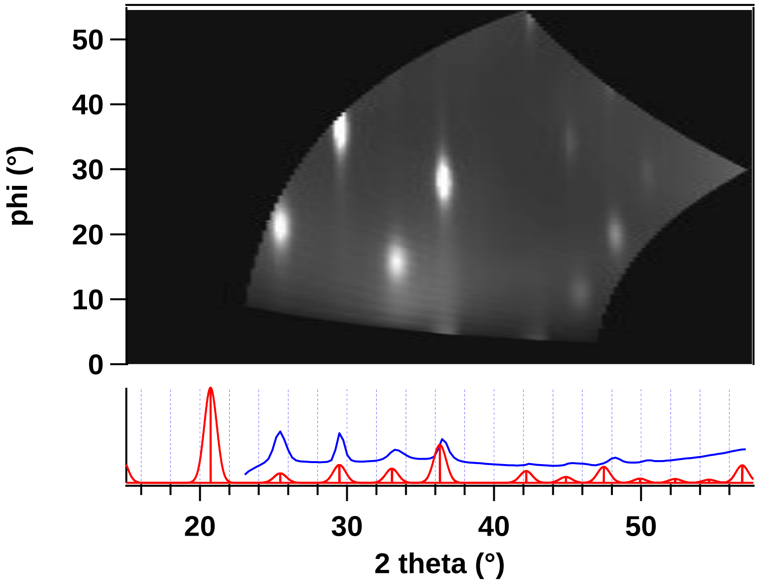
Grain size 27.7nm

Grain size 20.2nm

	K	Sb	Cs
L004 Si	2.50	1.00	1.16
L005 Si	2.37	1.00	0.91
L006 Si	2.21	1.00	0.95
L011 Si	2.07	1.00	0.94
L012 MgO	1.98	1.00	0.88

Good K/Cs/Sb ratio!

Camera 2

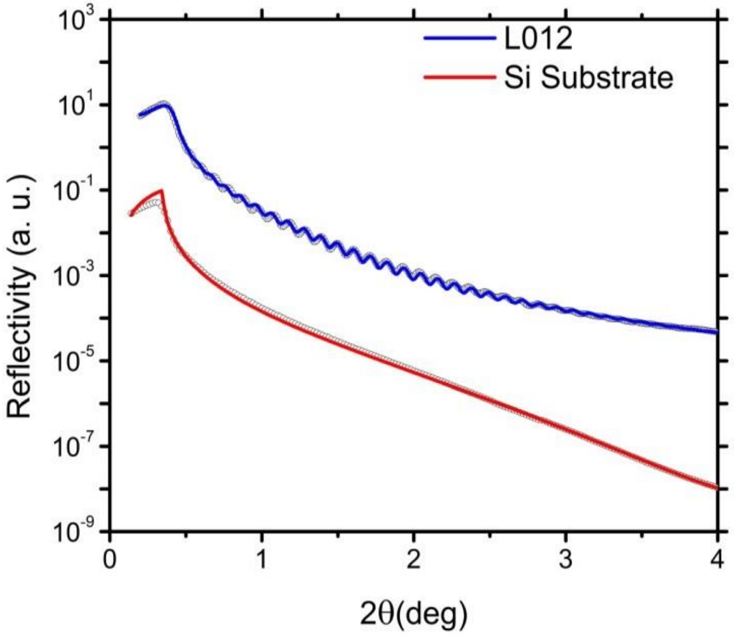


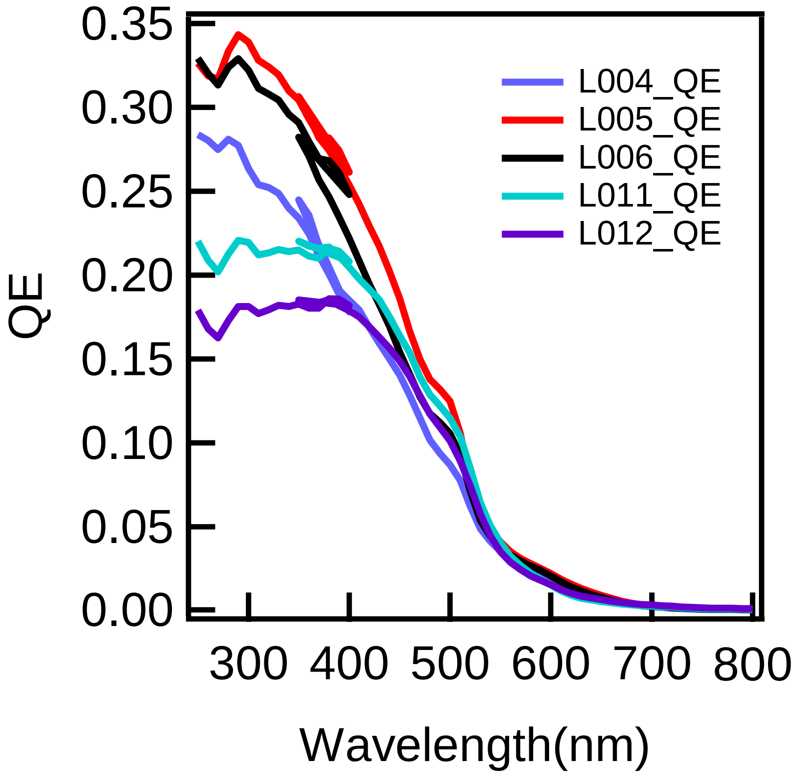
Highly textured K_2CsSb phase!

mm size lateral grains

This works for the entire Alkali antimonide family – we've created pseudo single crystals with a wide range of stoichiometries

Surface Roughness & QE





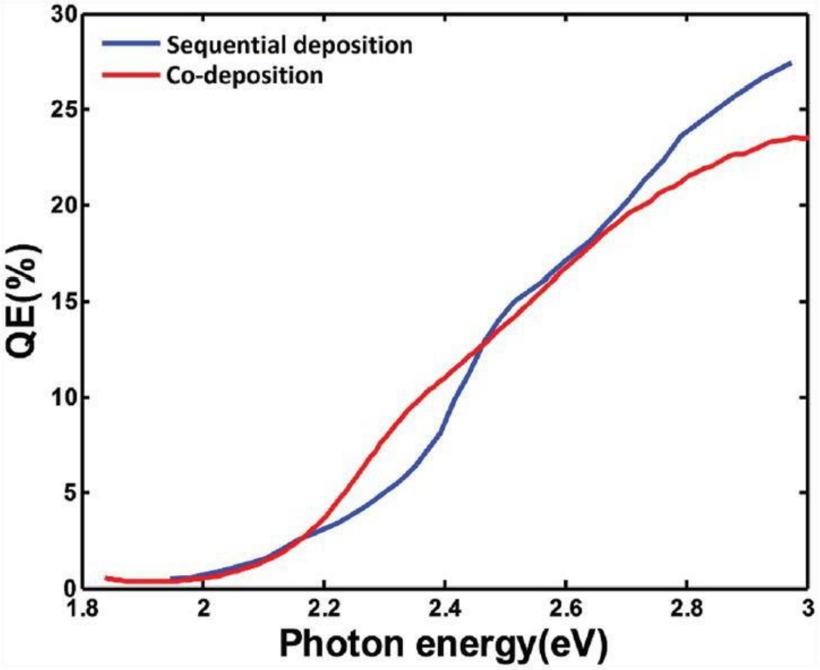
	QE@532nm(%)	Roughness(A)	Thickness (A)	Grain size (A)
L004 Si	4.9	3.5	234	155
L005 Si	5.8	11.5	815.3	277
L006 Si	5.4	13.8	757.5	202

Simultaneous evaporation of all constituents results in no crystal phase transformation

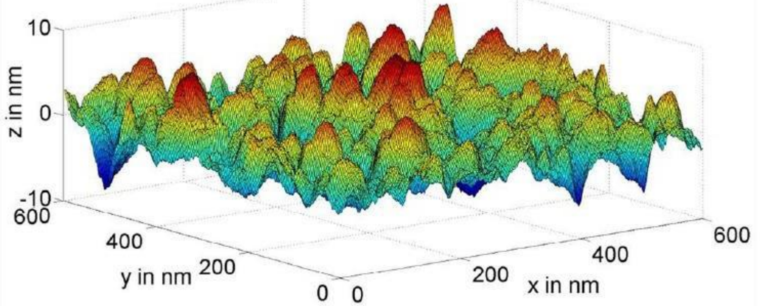
Smooth, reproducible and ultra-high QE. Highly Crystalline!

Co-deposition leads to efficient and ultra-smooth cathodes

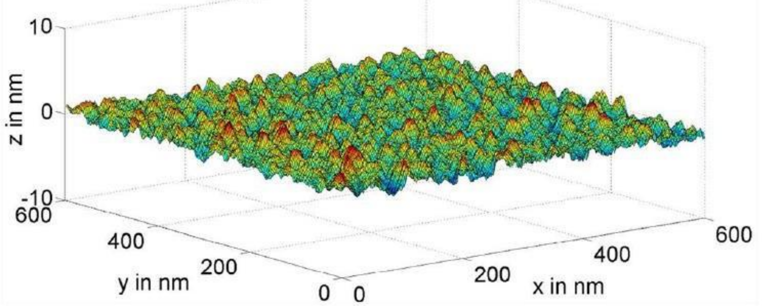
Co-deposited K2CsSb yield spectrum

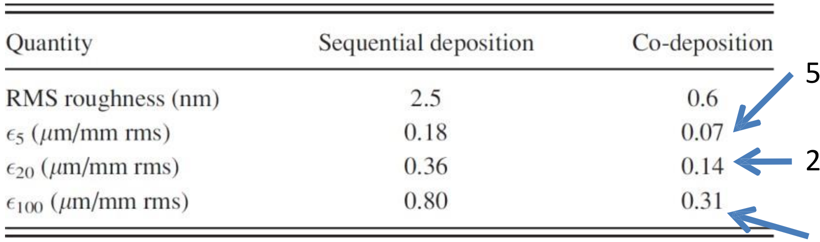


(a) Sequential deposition



(b) Co-deposition





Quantity	Sequential deposition	Co-deposition
RMS roughness (nm)	2.5	0.6
ϵ_5 (μm/mm rms)	0.18	0.07
ϵ_{20} (μm/mm rms)	0.36	0.14
ϵ_{100} (μm/mm rms)	0.80	0.31

5 MV/m

20 MV/m

100 MV/m

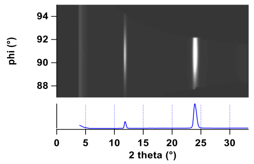
J. Feng et al., J. Appl. Phys., 121(4) 044904, 2017

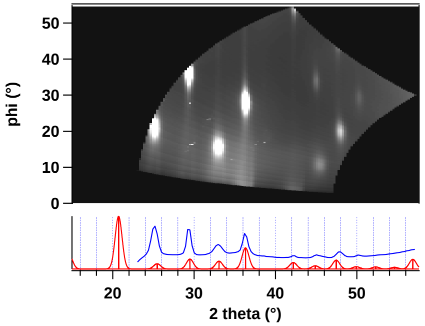
20 MV/m

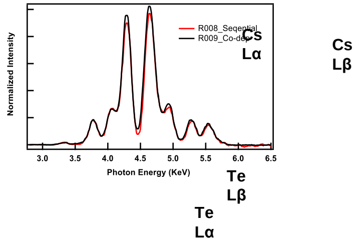
- significant emittance degradation for sequential method
    - no practical degradation with co-deposition

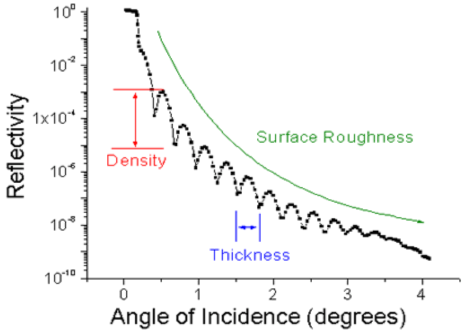
- LN2 threshold emission
    - significant degradation, even with co-deposition
    - we need films smoother than 0.6 nm!!

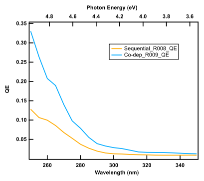
Let’s apply this to Cs-Te cathode growth

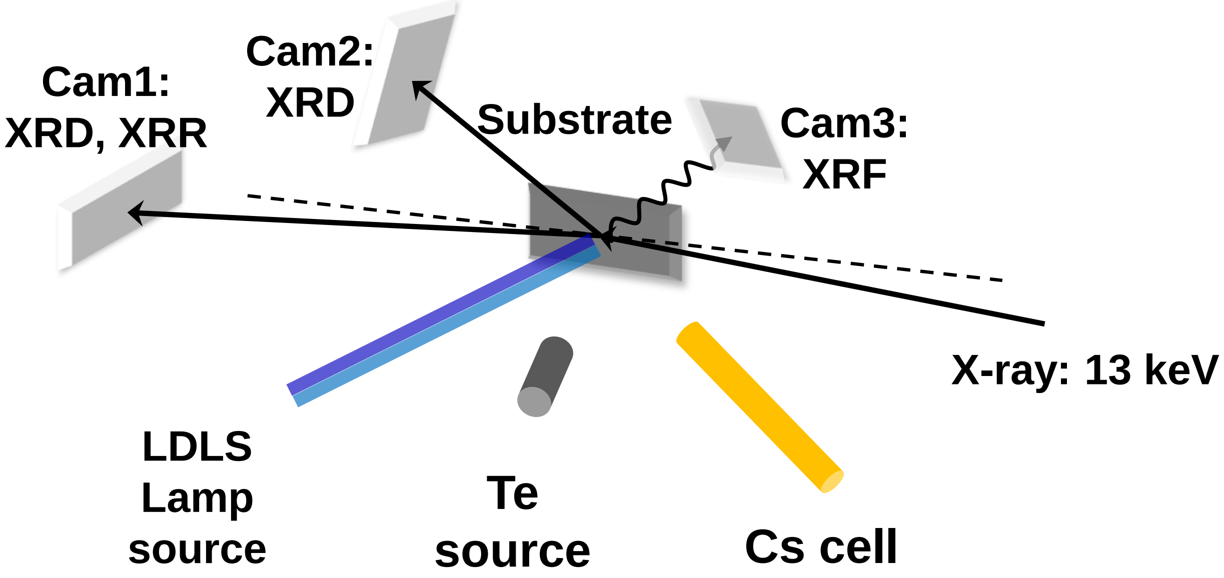






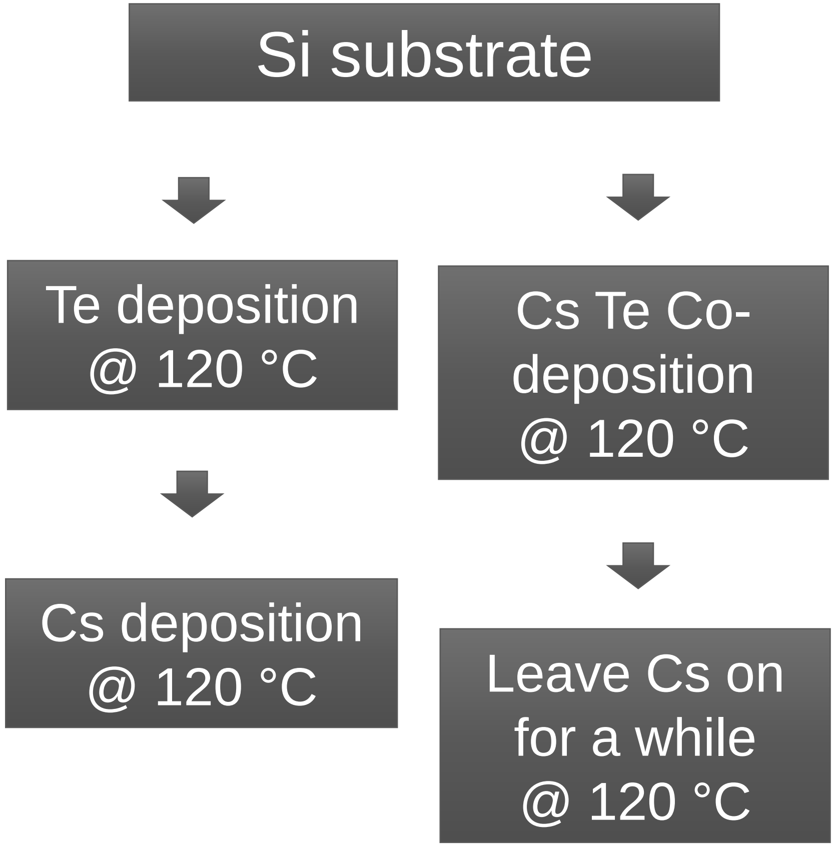






M. Gaowei, J. Sinsheimer, D. Strom, J. Xie, J. Cen, J. Walsh, E. Muller, and J. Smedley
Phys. Rev. Accel. Beams 22, 073401 – Published 23 July 2019

Sequential vs Co-dep CsTe -- XRF



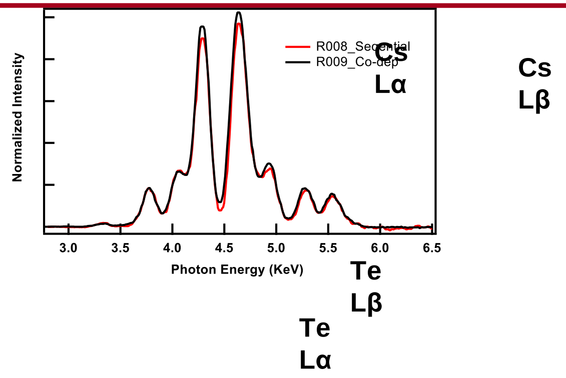
Si substrate

Te deposition @ 120 °C

Cs deposition @ 120 °C

Cs Te Co-deposition @ 120 °C

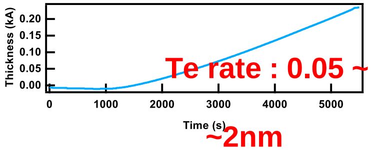
Leave Cs on for a while @ 120 °C



Sample	Te	Cs (+/- 0.05)
R008 Sequential	1.00	1.75
R009 Co-dep	1.00	1.98

At same substrate temperature; co-dep method incorporates more Cs.

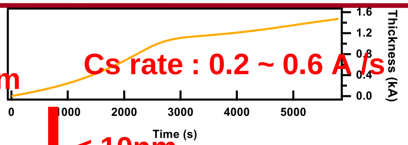
Realtime analysis : Sequential growth



Te rate : 0.05 ~ 0.07 A/s

~2nm

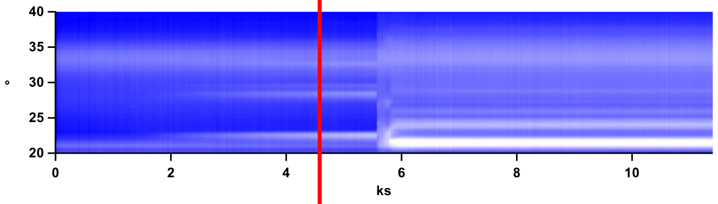
23.5nm

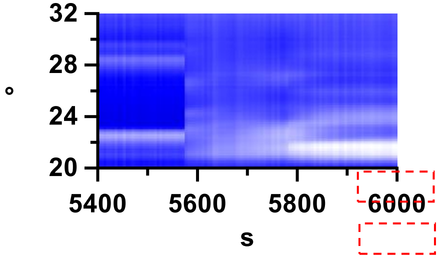


Cs rate : 0.2 ~ 0.6 A /s

< 10nm

~150 nm

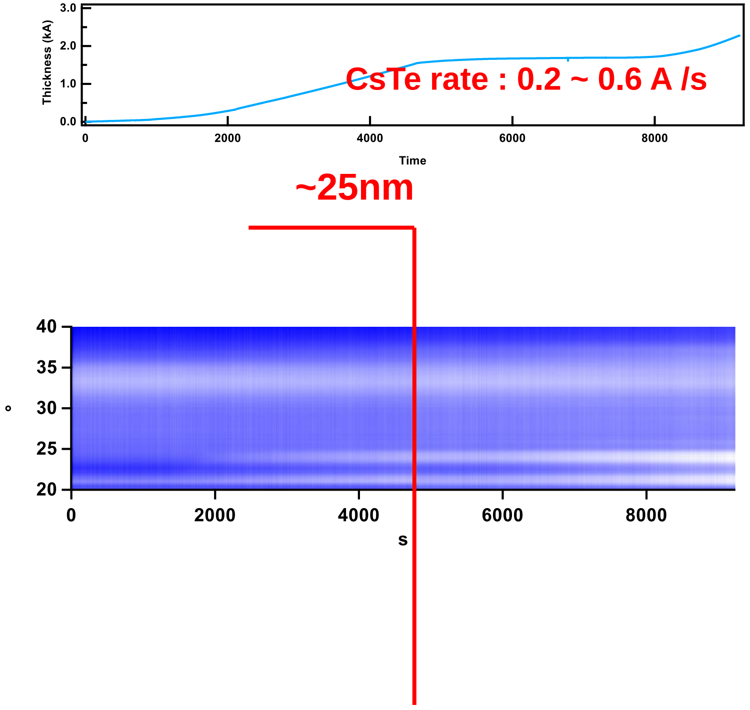




In less than 10 nm of Cs deposition, at least 2 intermediate phases appeared and disappeared, then a stable product of CsTe forms.

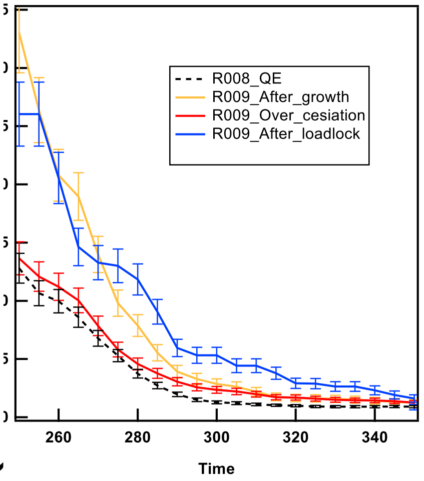
Realtime analysis : Co-deposition

~230nm



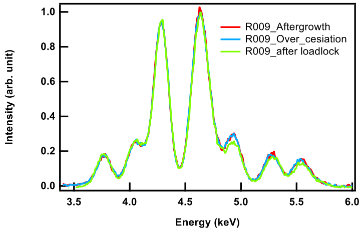
- **Starts to crystalize ~ 25 nm of total thickness.**
- **Same phase throughout the growth.**

CsTe cathode Quantum efficiency



QE

Wavelength (nm)



- **Co-dep can result in much higher QE than sequential**
- **over cesiation might lead to Cs build-up on the cathode surface and lower the QE**
- **QE can be recovered by removing the access Cs**

M. Gaowei, J. Sinsheimer, D. Strom, J. Xie, J. Cen, J. Walsh, E. Muller, and J. Smedley
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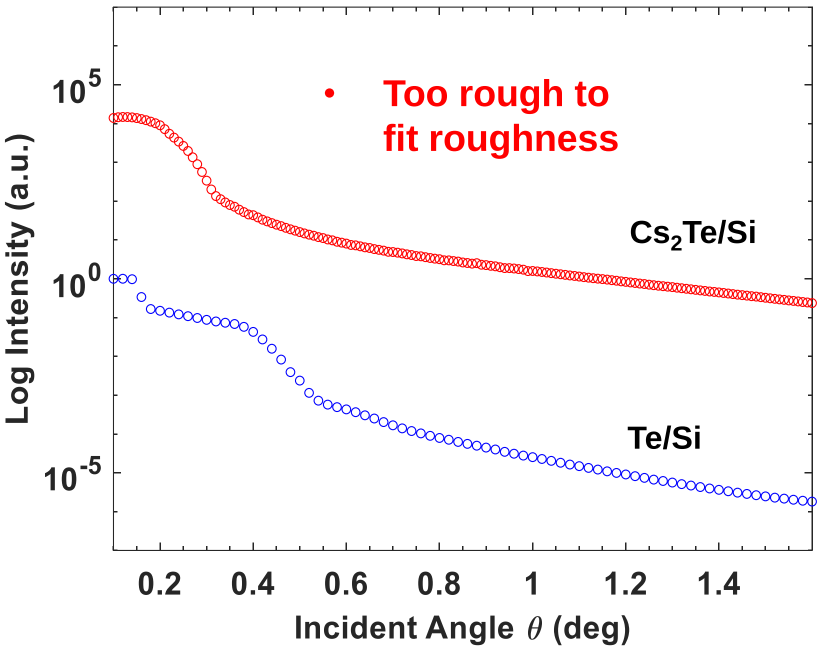
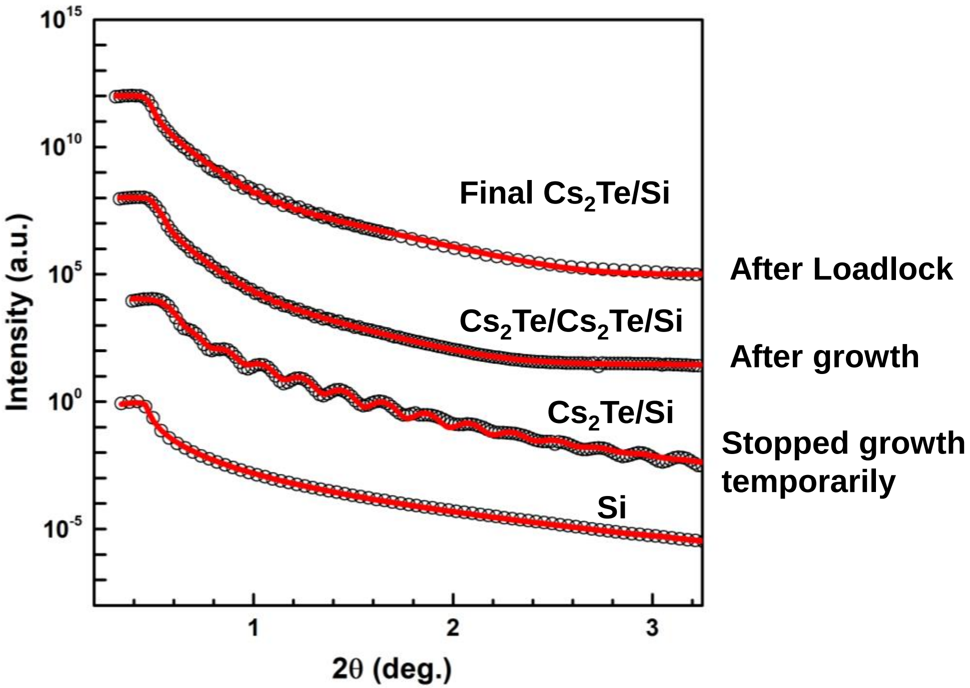
CsTe cathode surface roughness: XRR analysis

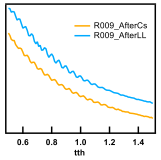


R008 Sequential - CsTe on Si



R009 Co-dep - CsTe on Si

	Thickness (Å)	Roughness (Å)
FINAL Cs2Te/Cs2Te	968.3 ± 2.9 (total Cs_2Te)	19.1 ± 0.2
Cs2Te/Cs2Te	1026.1 ± 1.6 (total Cs_2Te)	19.10 ± 0.07
Cs2Te	245.5 ± 1.7	9.55 ± 0.14
Si Substrate	-	3.75 ± 0.02

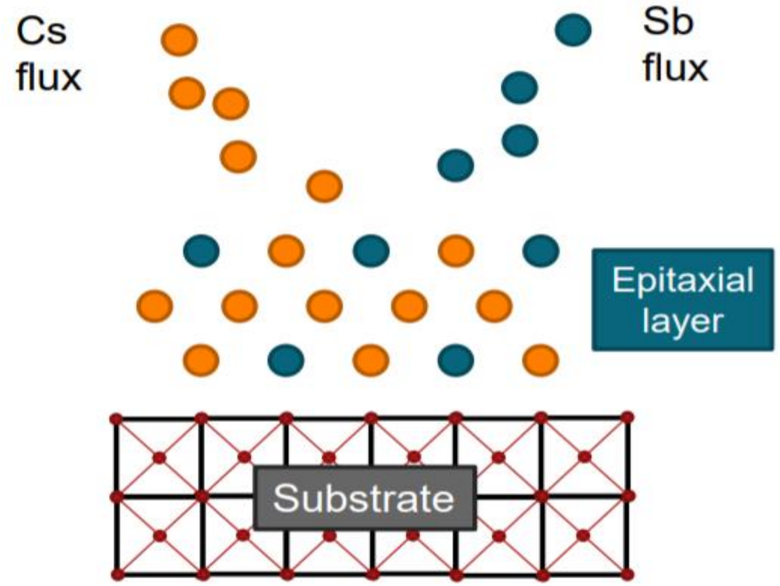


Epitaxial Growth

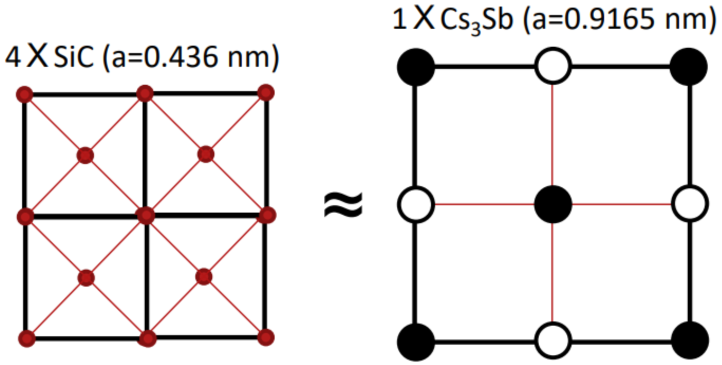
Cs_3Sb photocathodes are conventionally grown polycrystalline with disordered surfaces.

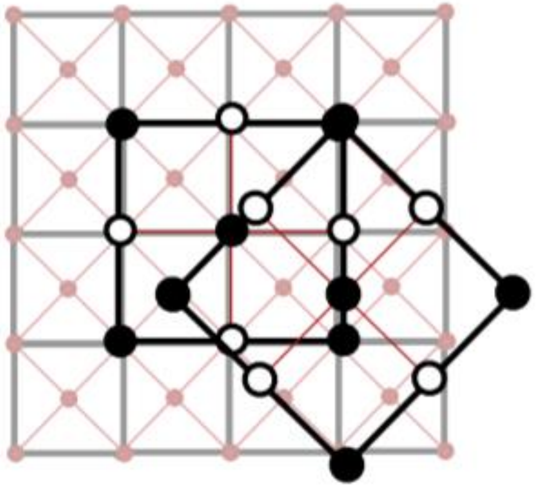
Reducing the intrinsic emittance may be limited by the surface disorder in the form of defects, grain boundaries, and roughness.

Epitaxy is the alignment of crystal layers with respect to an underlying crystal seed layer.



Cs_3Sb is grown on lattice matched, 3C-SiC substrates.





Thanks to Chad Pennington

Why epitaxy?

- Towards higher brightness: a figure for the quality of the electron beam.

$$B_n = \frac{2m_e c^2 I}{\sigma_x^2 MTE}$$

Beam current: quantum efficiency, laser fluence

Mean transverse energy: Intrinsic momentum spread + roughness + laser heating + …

  - Material ordering eliminates defects (roughness, grain boundaries) that contribute to electron momentum spread (MTE).

  - Epitaxy opens the door to band structure and/or QE engineering – similar to work on single crystal GaAs and GaN.

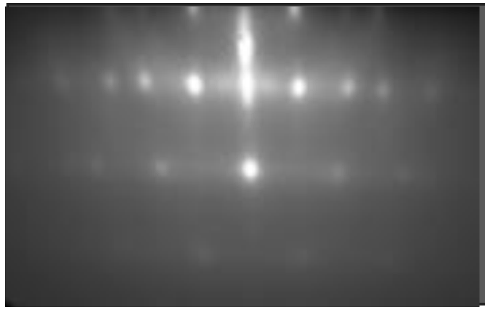
-. W. Liu, *et.* , Appl. Phys. Lett. 109, 252104 (2016).
- J. Marini,, Polarization engineered N-polar Cs-free GaN photocathodes, J. Appl. Phys. 124, 113101 (2018).

Thanks to Chad Pennington

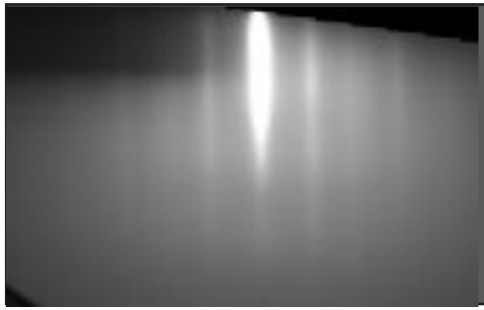
Flat, ordered Cs_3Sb on 3C-SiC(001)

- RHEED patterns indicate flat and fiber-textured films.

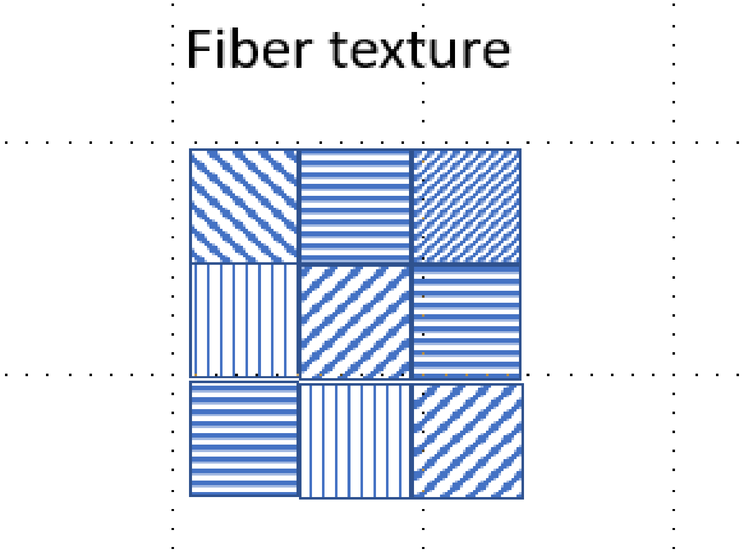
Deposition temperature: 90 C.
Grown using PLD.
Fiber textured film.
Thickness ~8 nm.



Deposition temperature: 80 C.
Grown using PLD.
Ordered film with 3D islands.
Thickness = 5.6 nm.



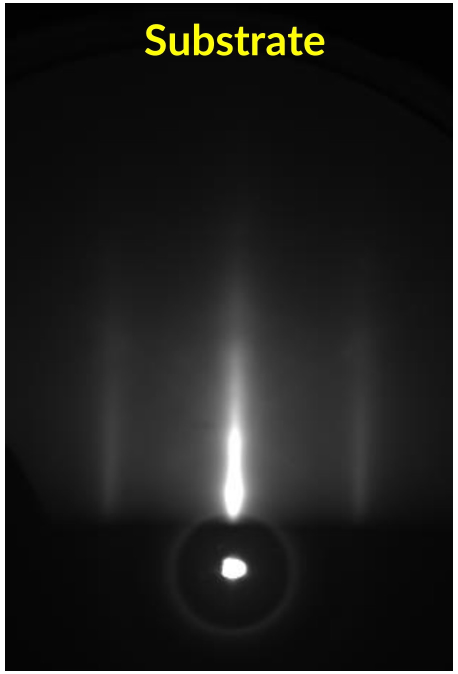
Deposition temperature: 85 C.
Grown using thermal evaporation.
Fiber textured film.
Thickness = 8.3 nm.

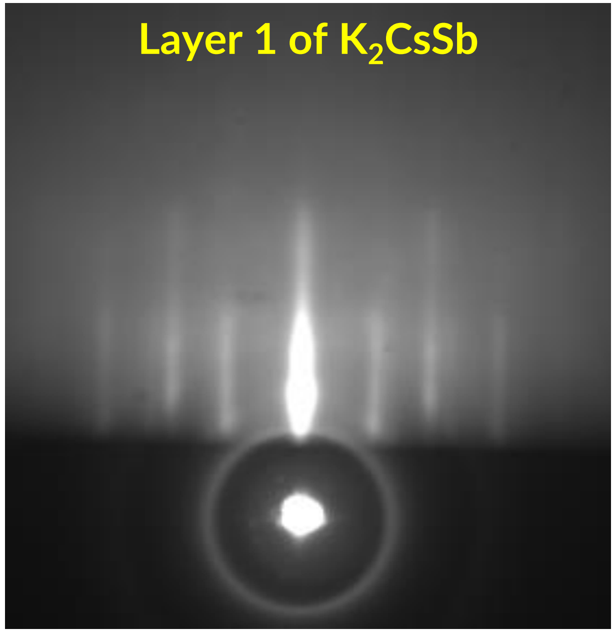


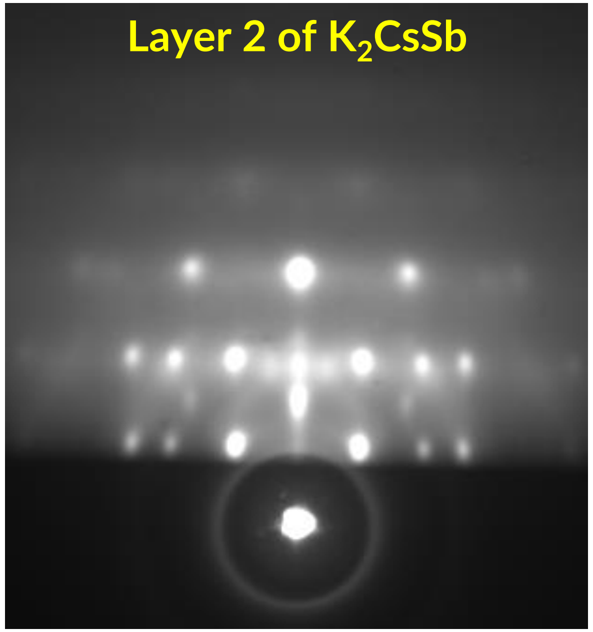
- Flat, thin, and ordered films with near percent level QE at 530 nm are easily grown with PLD and x-ray/RHEED diagnostics.
- A Cs:Sb stoichiometry ratio of 3:1 is derived from XRF for ordered Cs_3Sb films.

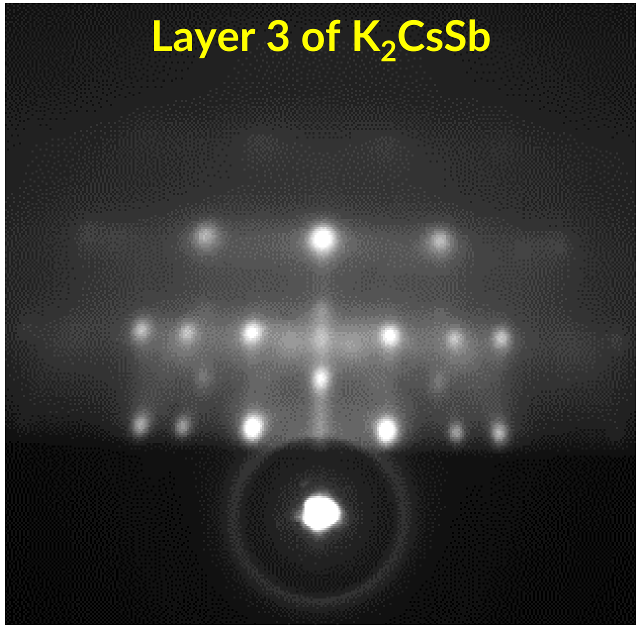
Thanks to Chad Pennington

RHEED of K_2CsSb/New Substrate (Sample-2)


Substrate


Layer 1 of K_2CsSb


Layer 2 of K_2CsSb


Layer 3 of K_2CsSb

RHEED of K_2CsSb

- ☐ RHEED image shows epitaxial growth.
- ☐ Streaks represent smooth surfaces with small domains.
- ☐ Dots represents 3D islands.

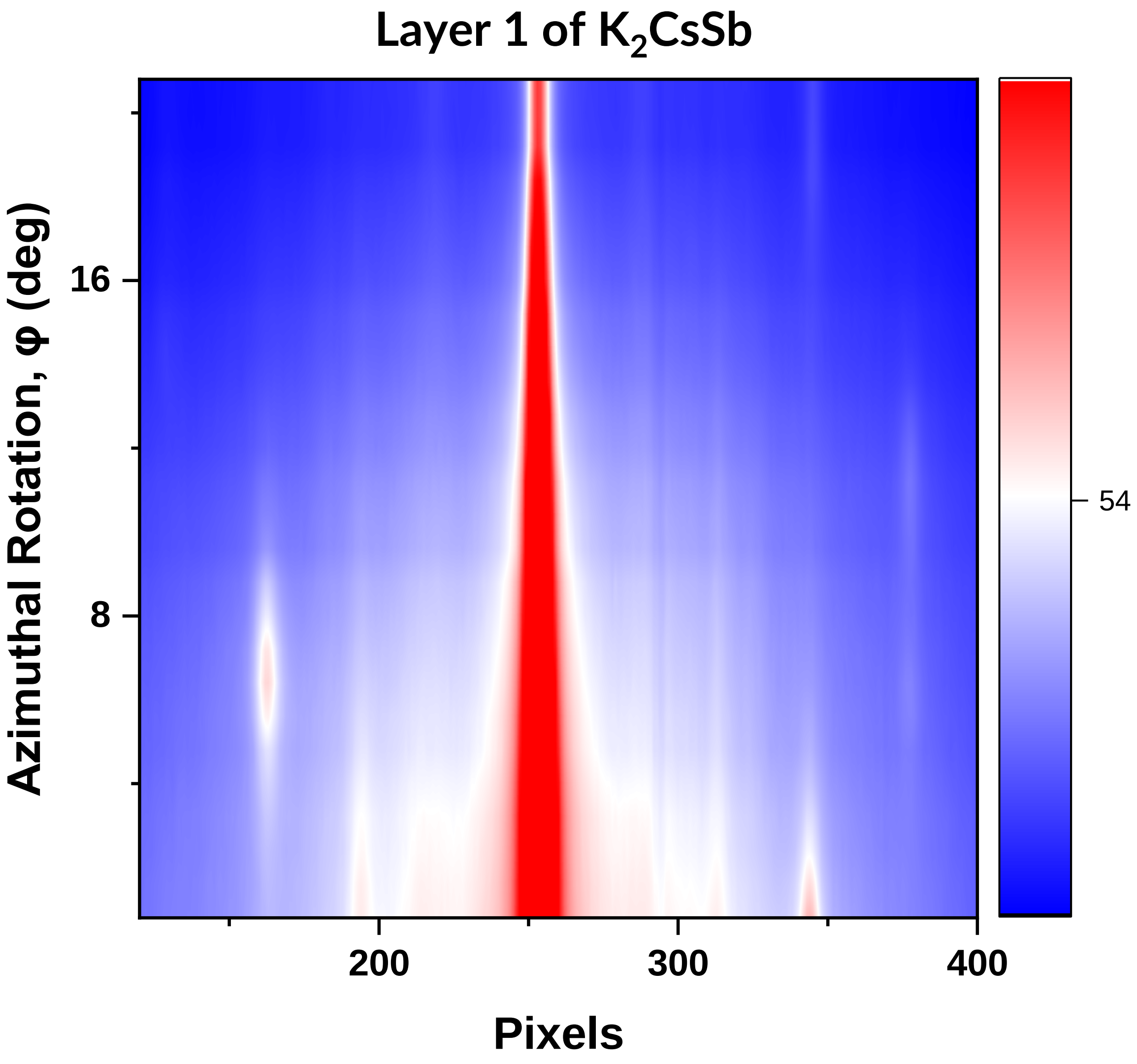
K_2CsSb on Substrate

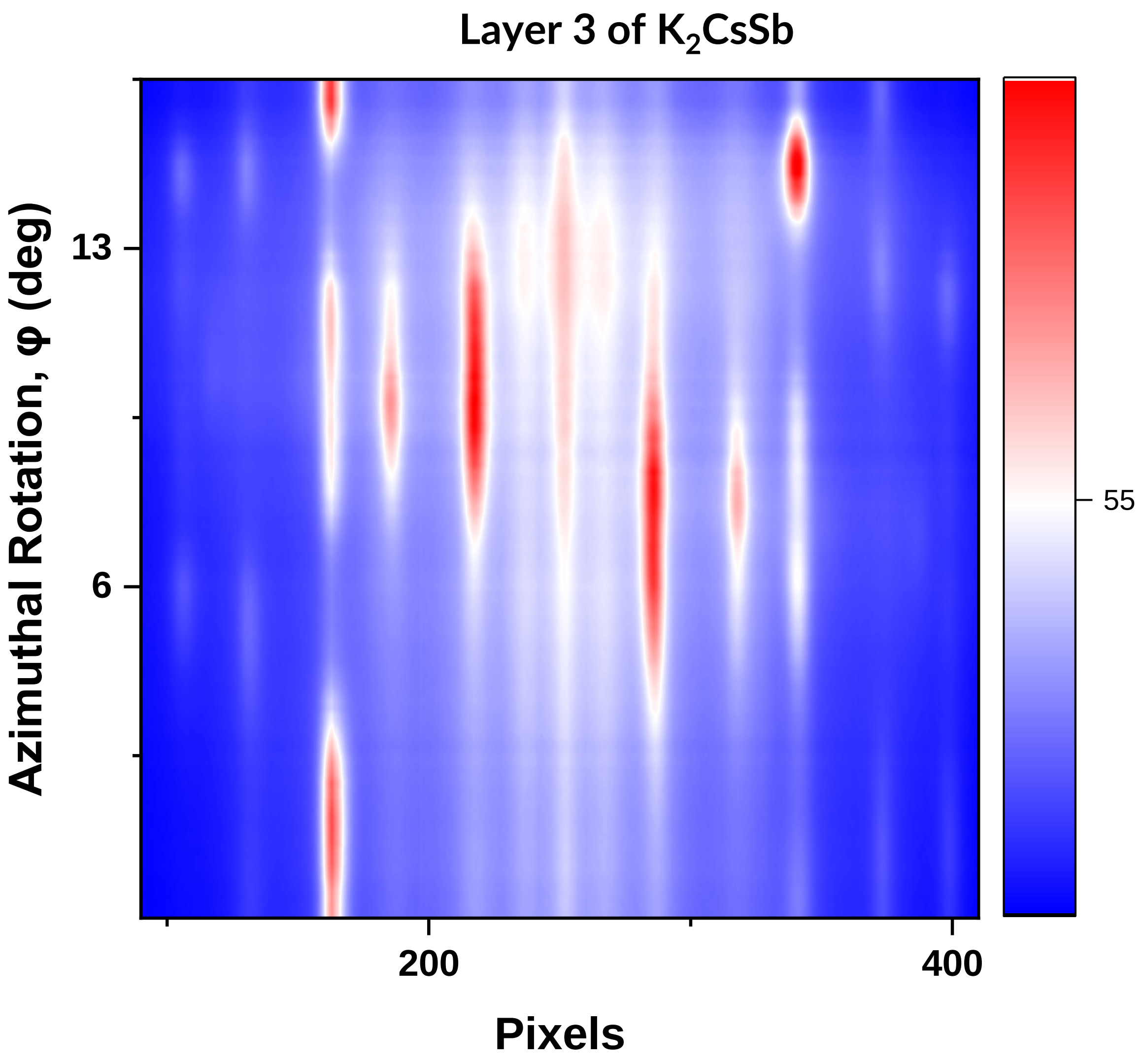
Peak # from center	D(pixel)	d (Å)	Close to Planes
1st	34.55	4.96	111
2nd	59	2.91	122
3rd	69.25	2.48	222
4th	90.4	1.90	133

Thanks to Kali Prasanna Mondal

Brookhaven National Laboratory

Azimuthal angular dependence of RHEED from Sample-2

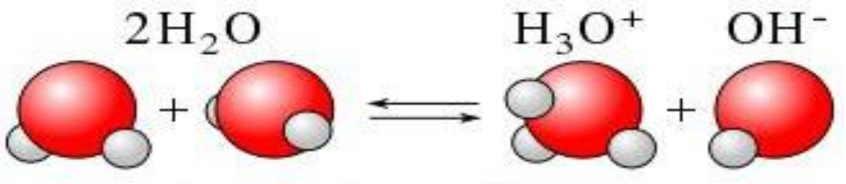




Azimuthal angular dependence observed

Thanks to Kali Prasanna Mondal

MHz MeV-UED to Enable Grand Challenge Science



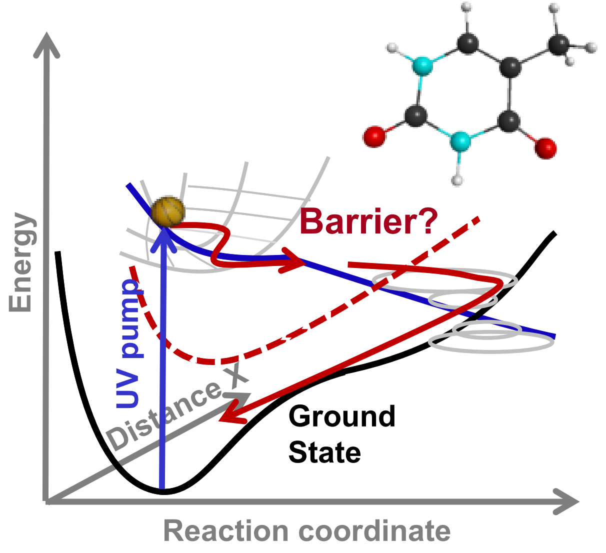
water molecule dissociation

The transfer of a proton or a hydrogen atom from one group (OH, -NH,) to another (C=O, -N=) has been referred to as 'the most general and important reaction in chemistry'

Proton-transfer reaction dynamics

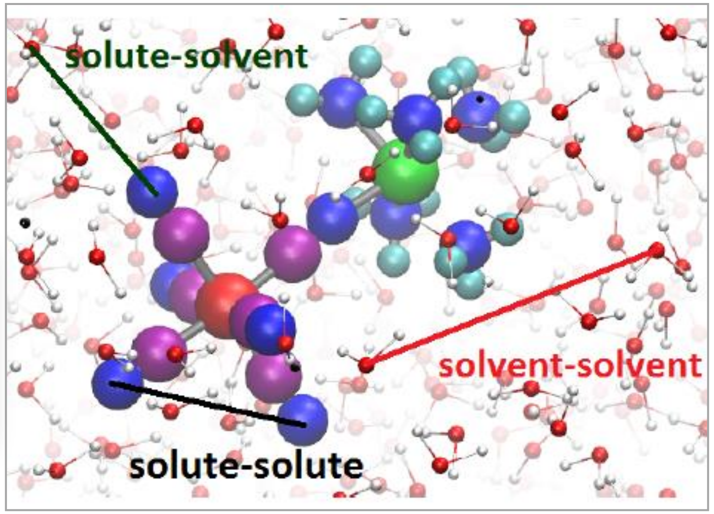
Abderrazzak Douhal [a], Françoise Lahmani [b], Ahmed H. Zewail [c,*]

Chemical Physics 207 (1996) 477–498



- **Real-space observation of proton dynamics requires both proton sensitivity and high time resolution (< ~50 fs).**
- **Neutron scattering lacks time resolution, X-ray diffraction lacks proton sensitivity. Electron diffraction is the only viable path.**

MHz MeV-UED based on the SRF gun will possess exquisite sensitivity and atomic spatial temporal resolutions to address one of the grand challenge sciences:
Probing proton-transfer of chemical and biological process in its native environments.



Thanks to XJ Wang

Optical Near Field Electron Microcopy

Optical microscopy with significantly **sub wavelength** resolution

Eventually fast enought to observe biological dynamics

Key inovation:

Convert scattered photons into electrons in the optical near-field

Resolution is limited by the sample to electon emitting surface distance -> **need ultra-thin photocathodes!**

Good QE in the visible → $\mathrm{Cs_3Sb}$ or $\mathrm{K_2CsSb}$ candidates

>1% @ 530 nm for good signal

Ultra-smooth ultra-thin layer

<10 nm film; <1 nm roughness to not degrade resolution

Suitable transparent substrate for ONEM back-illumination

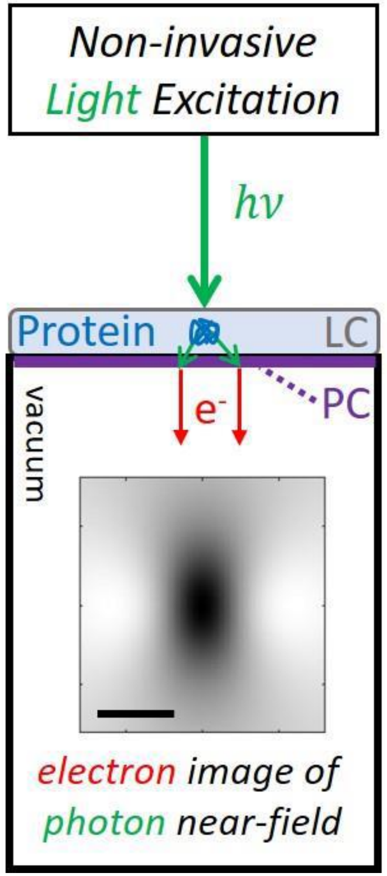
SiN, SiC, graphene

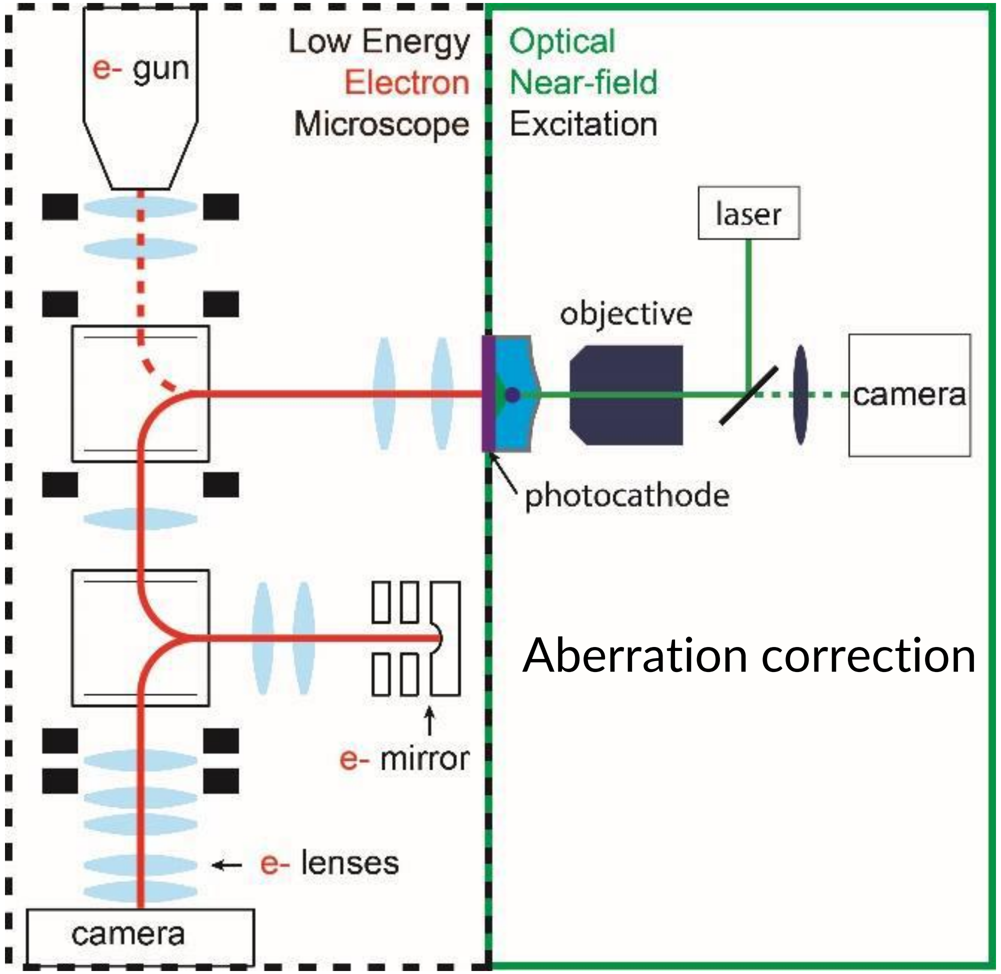
Universiteit
Leiden

Thanks to Guido Stam

Phys. Rev. Applied 16, 014008 (2021)

Positioning ONEM in LEEM

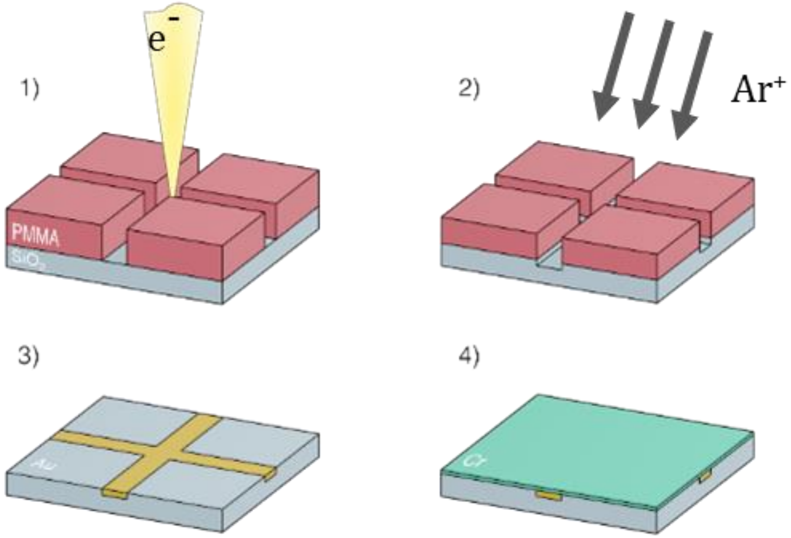




Aberration correction

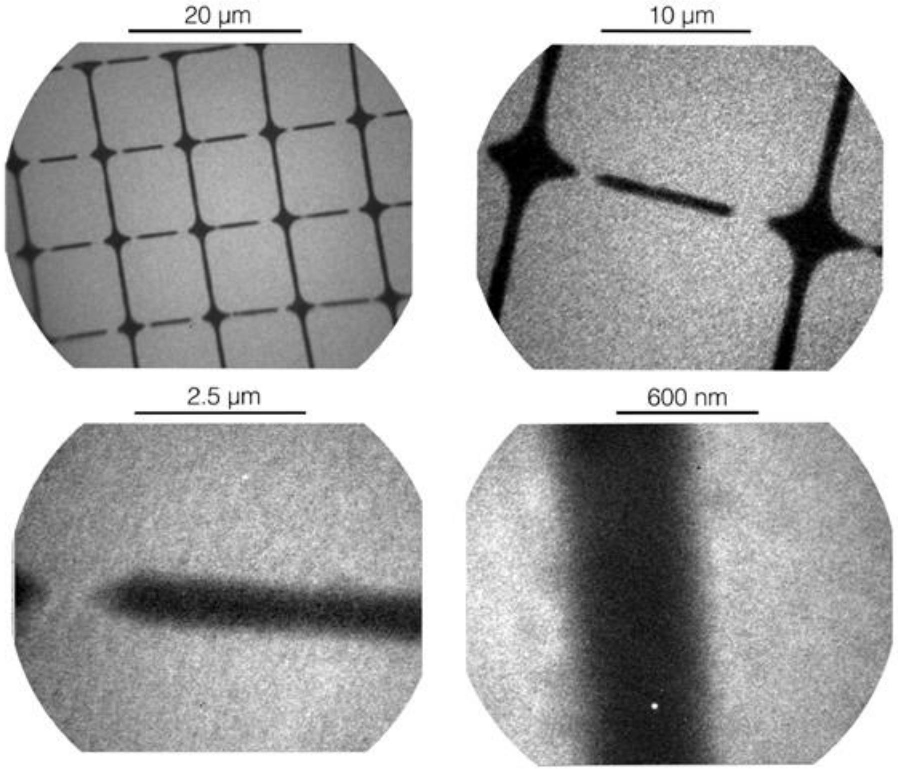
UV-ONEM using LED (275 nm)

1) Create embedded Au nanostructures



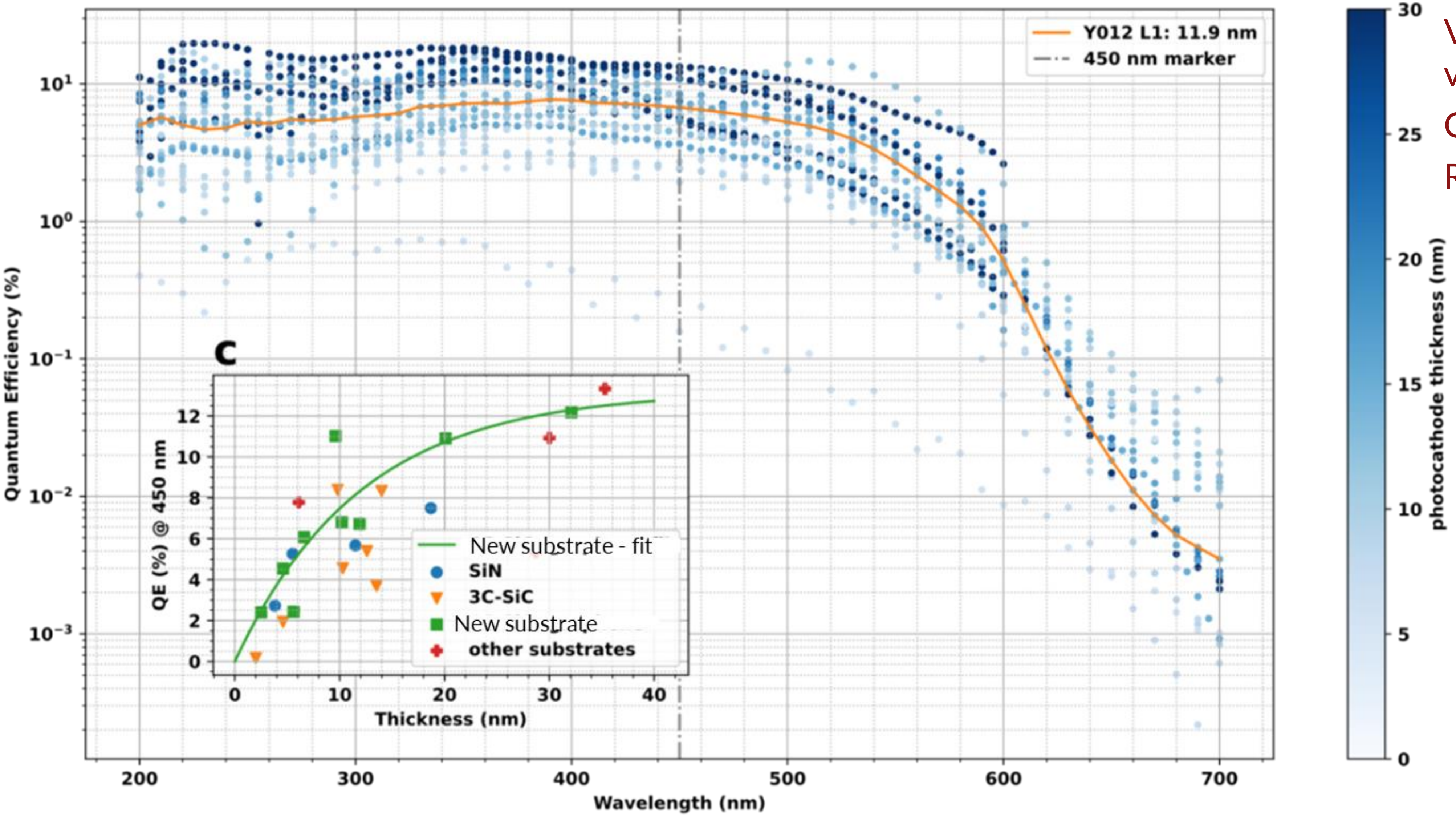
2) Cover with photocathode layer:
Chromium

ONEM **Proof of Principle**!



Illumination time: **10-35 min.**
Estimated resolution: 37 nm = λ/7

Quantum Efficiency



Very good already at very low thicknesses

Cutoff above 600 nm

Roughness < 1 nm

Conclusions and Thanks

LCLS-II HE will bring world class hard x-ray performance to LCLS-II

A new gun and injector are an option to further increase this reach

- Gun development and production underway
- Hopefully tunnel for LEI will be complete in early 2027

Semiconductor cathodes are the path to low emittance and high QE

- Ultrasmooth cathodes now available
- Epitaxy has been achieved on ordered substrates

Applications at the cutting edge

- UED/UEM
- ONEM

MSU has the potential to bring cathode science and superconducting guns together

Thank you

BERKELEY LAB
Fermilab
FACILITY FOR RARE ISOTOPE BEAMS
Jefferson Lab
SLAC NATIONAL ACCELERATOR LABORATORY
Stanford University
U.S. DEPARTMENT OF ENERGY

Trends and observations

Modern photocathode development is focused in two areas:

- High average current (e^- cooling, CW FEL – FLASH, EuXFEL, LCLS-II)
- Ultra high brightness (UED/UEM)

These are driving development of semiconductor cathodes

- **Spatial uniformity (better than 0.6nm RMS), Single crystal**

  A. Galdi et al., Appl. Phys. Lett. 118, 244101 (2021)
  M. Gaowei, et al., Phys. Rev. Accel. Beams 22, 073401 (2019)

- **Ultra thin/optical etalon**

  A. Alexander, et al., AIP Advances 11, 065325 (2021)

- **Sealed photocathode systems for commercial delivery**

  J. Smedley, et al., Alkali Antimonide Photocathodes for Everyone, PAC 2013, 1178

- **Graphene or HBN covered cathodes**

  J. Biswas, et al., APL Materials 10, 111115 (2022)
  H. Yamaguchi, et al., npj 2D Materials and Applications, 12, (2017)