Cathode Characterization and Fabrication

Applying the tools of modern material science to cathode design

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Snowmass2021 Electron Source Workshop
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Thanks to Mengjia Gaowei and Alice Galdi (and many others)

Talk Outline

- **Introduction – what makes a good photocathode?**
  - Focus on Alkali Antimonides and Tellurides
- **Deposition methods**
- **Characterization**
- **Path forward**

Photocathode needs in accelerator applications

Electron beam required for e-cooling

- High average current (> 100 mA)
- High bunch charge (1nC)
- Long lifetime (> 1 week)
- Reproducible

FEL sources

Moderate currents

Emittance improvement

(ideally 0.1 µm/mm)

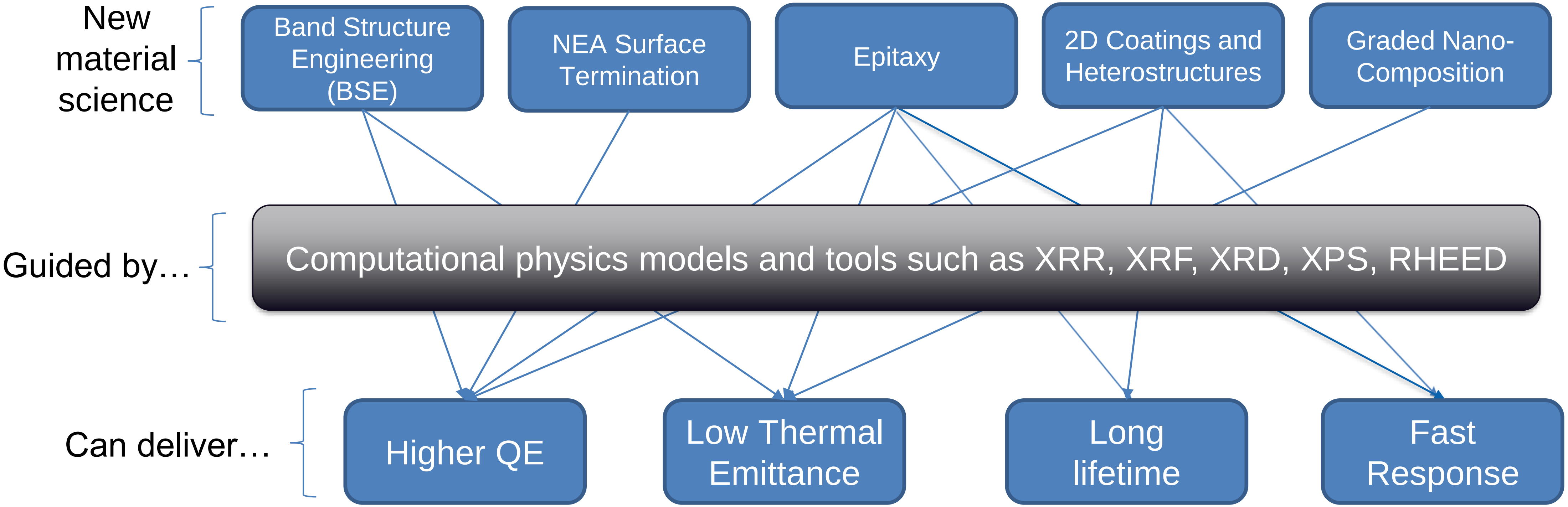
Ultrafast Electron Diffraction/Microscopy

High brightness

Very low current

Short pulse duration

New fabrication techniques are developed and applied to photocathodes motivated by theory and guided by real-time *in situ* x-ray analysis



New material science

- Band Structure Engineering (BSE)
- NEA Surface Termination
- Epitaxy
- 2D Coatings and Heterostructures
- Graded Nano-Composition

Guided by…

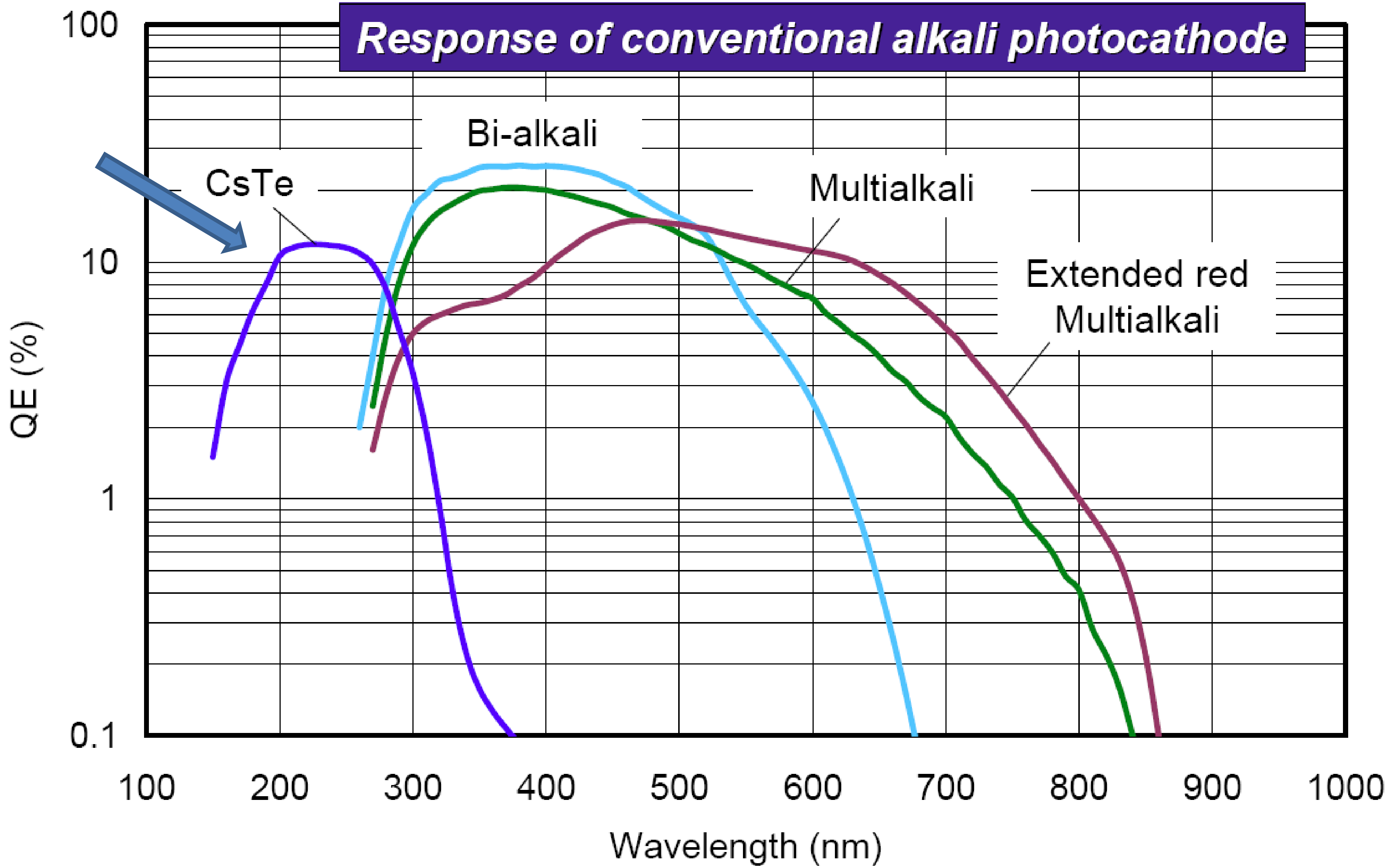
Computational physics models and tools such as XRR, XRF, XRD, XPS, RHEED

Can deliver…

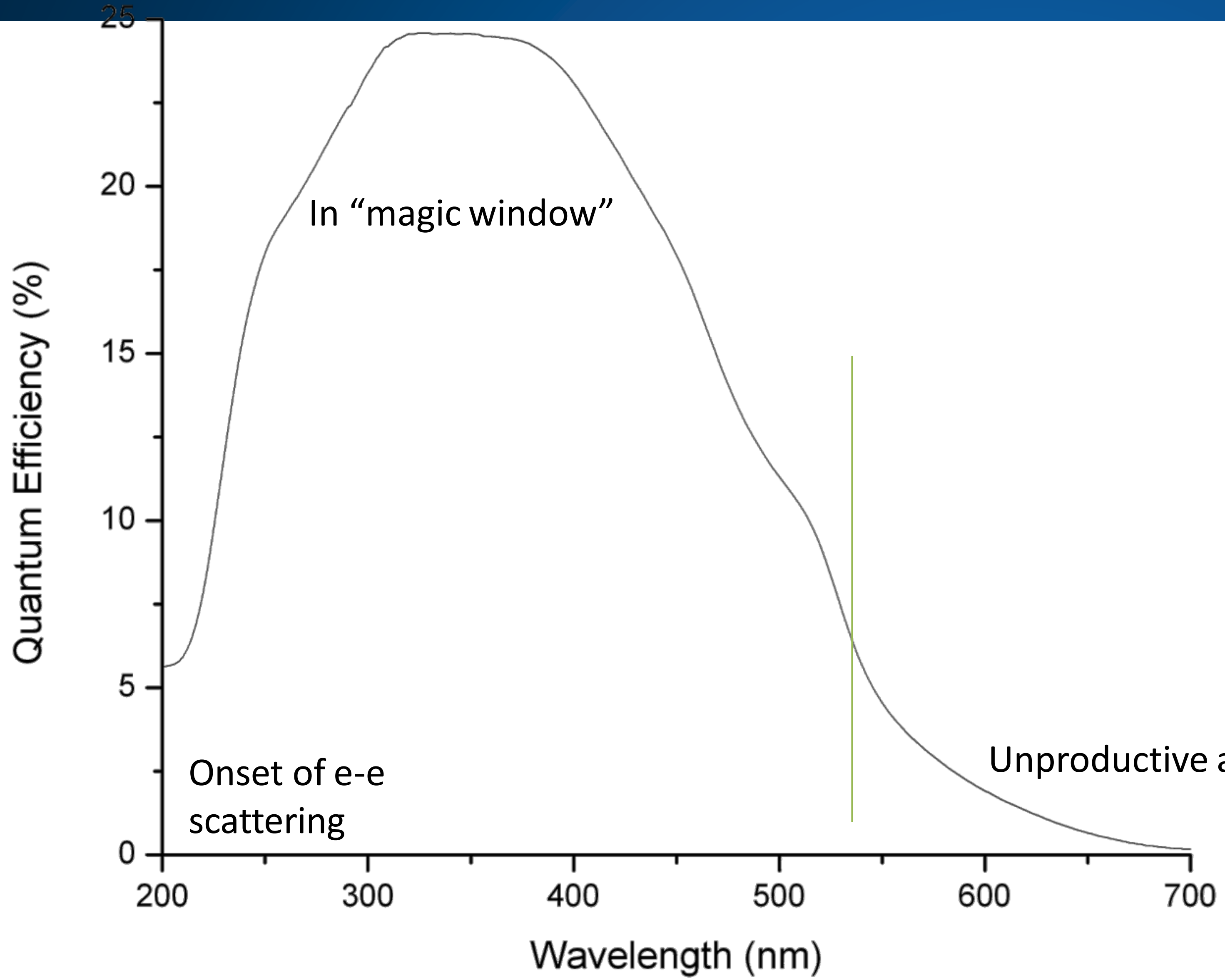
- Higher QE
- Low Thermal Emittance
- Long lifetime
- Fast Response

- ***In Situ*** **or rapid feedback is required to optimize for material properties other than QE**
- **Modeling is needed to guide growth and understand properties**

Response of conventional alkali photocathode



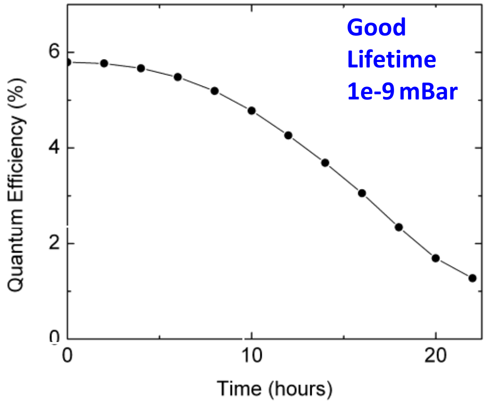
K_2CsSb: A Good Candidate



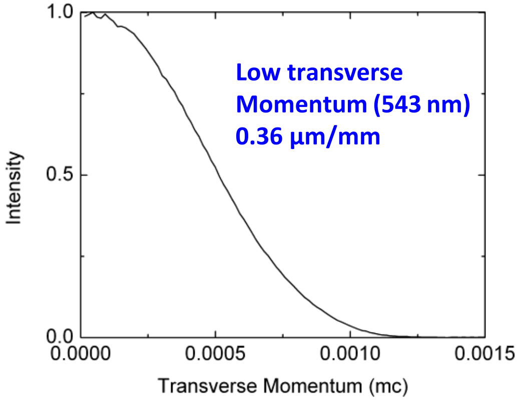
In “magic window”

Onset of e-e scattering

Unproductive absorption



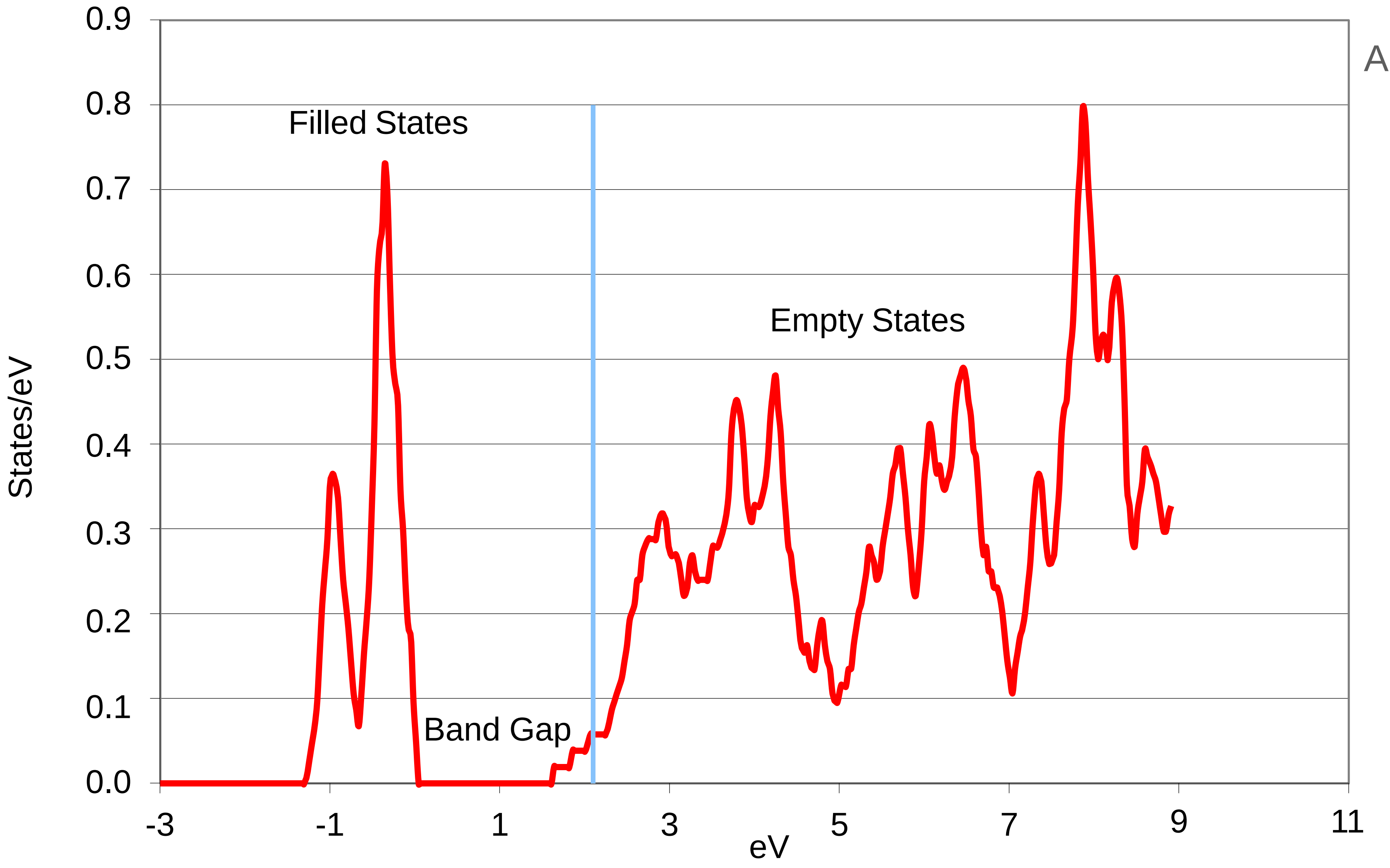
Good Lifetime 1e-9 mBar



Low transverse Momentum (543 nm) 0.36 μm/mm

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

K_2CsSb Density of States



Advantages of the Antimonides

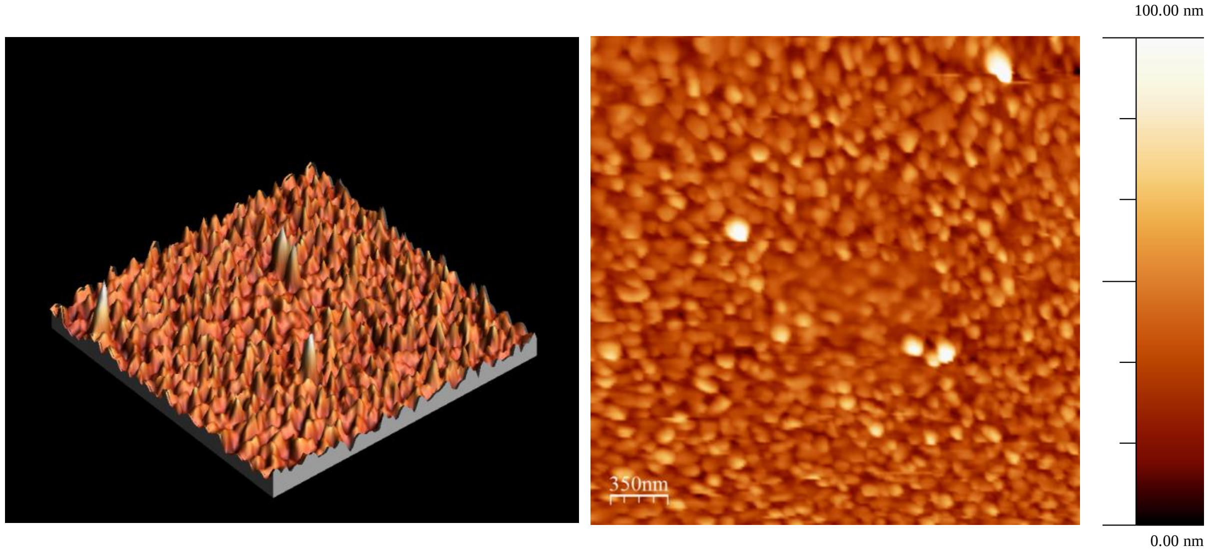
- Narrow Valence Width
- High Optical Density
- Fast (for semiconductors)

A.R.H.F. Ettema and R.A. de Groot, Phys. Rev. B **66**, 115102 (2002)

Deposition Methods

- **Sequential deposition via thermal evaporation**
  - Easy to control (one element at a time)
  - Recipe to maximize QE
- **Co-evaporation via thermal evaporation or effusion cells**
  - Much harder to control stoichiometry
  - Potentially much better films
- **ALD – difficult as no alkali precursors**
- **Sputtering and Pulsed Laser Deposition**
  - Difficult to control stoichiometry, does not sputter stoichiometricly
  - Bulk targets are a pain (require custom fabrication and vacuum load lock)
  - Can fix stoichiometry with Cs evaporation: M. Gaowei, et al., Synthesis and x-ray characterization of sputtered bi-alkali antimonide photocathodes. APL Materials, 2017. 5(11): p. 116104
  - PLD of Sb can lead to precision control of deposition – Digital Growth!

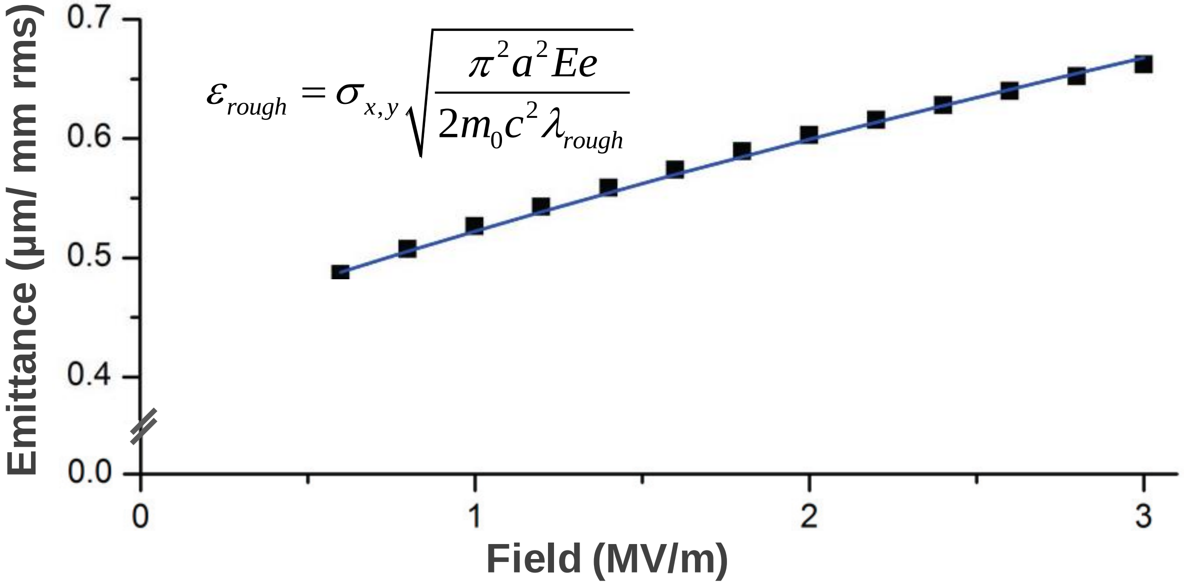
Traditional Sequential Deposition of K2CsSb High QE and Rough Surface



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

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

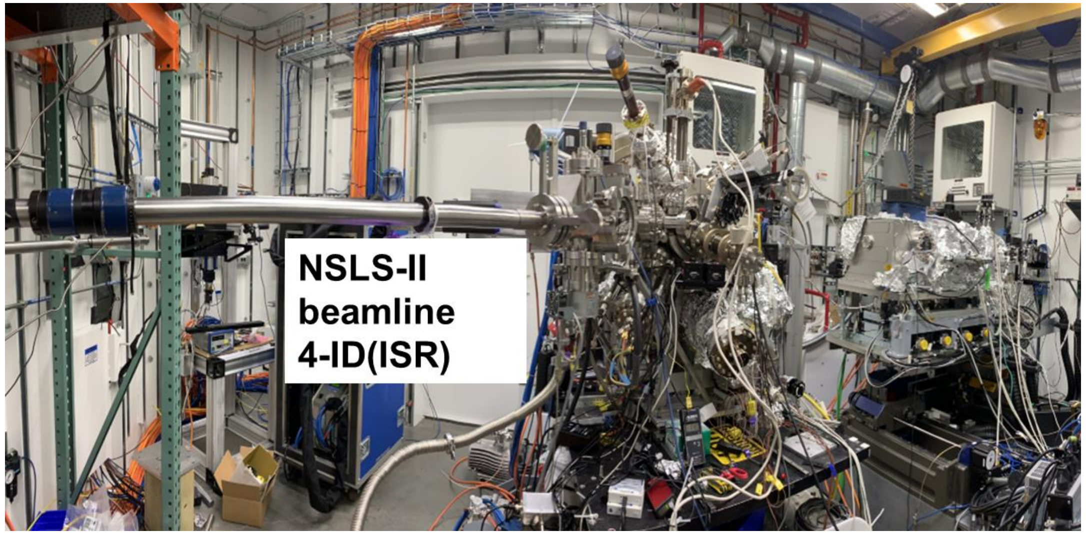
**Emittance vs field
measured with
Momentatron, 532 nm light**

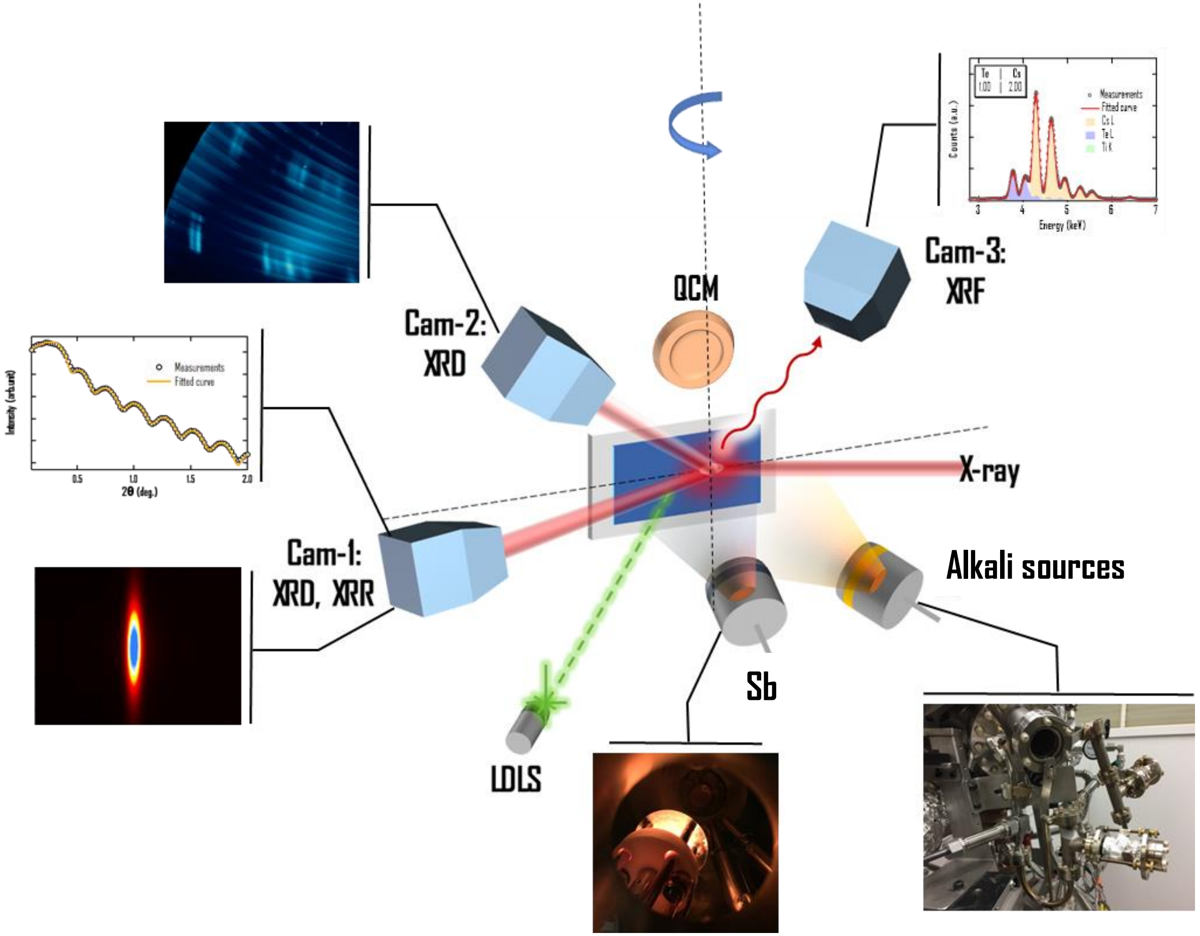


$$\varepsilon_{rough} = \sigma_{x,y}\sqrt{\frac{\pi^2 a^2 E e}{2 m_0 c^2 \lambda_{rough}}}$$

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

Cathode Material development @BNL : In situ and real time x-ray characterization



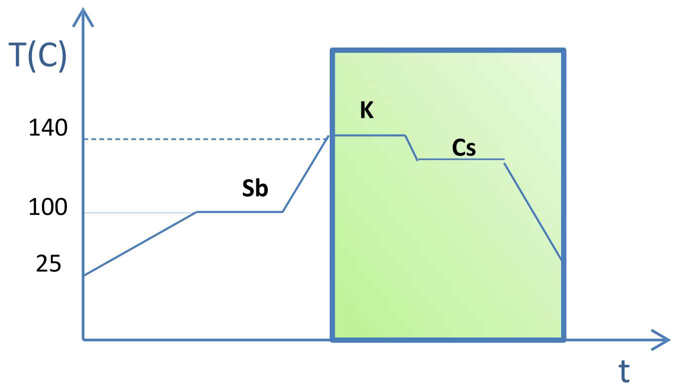


Growth controls:
☐ T_{sub}
☐ Flux rate

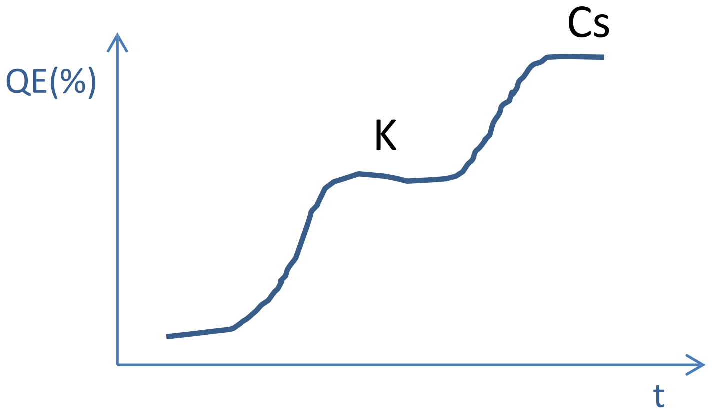
Characterization:
☐ QCM
☐ XRD
☐ XRR
☐ XRF
☐ QE

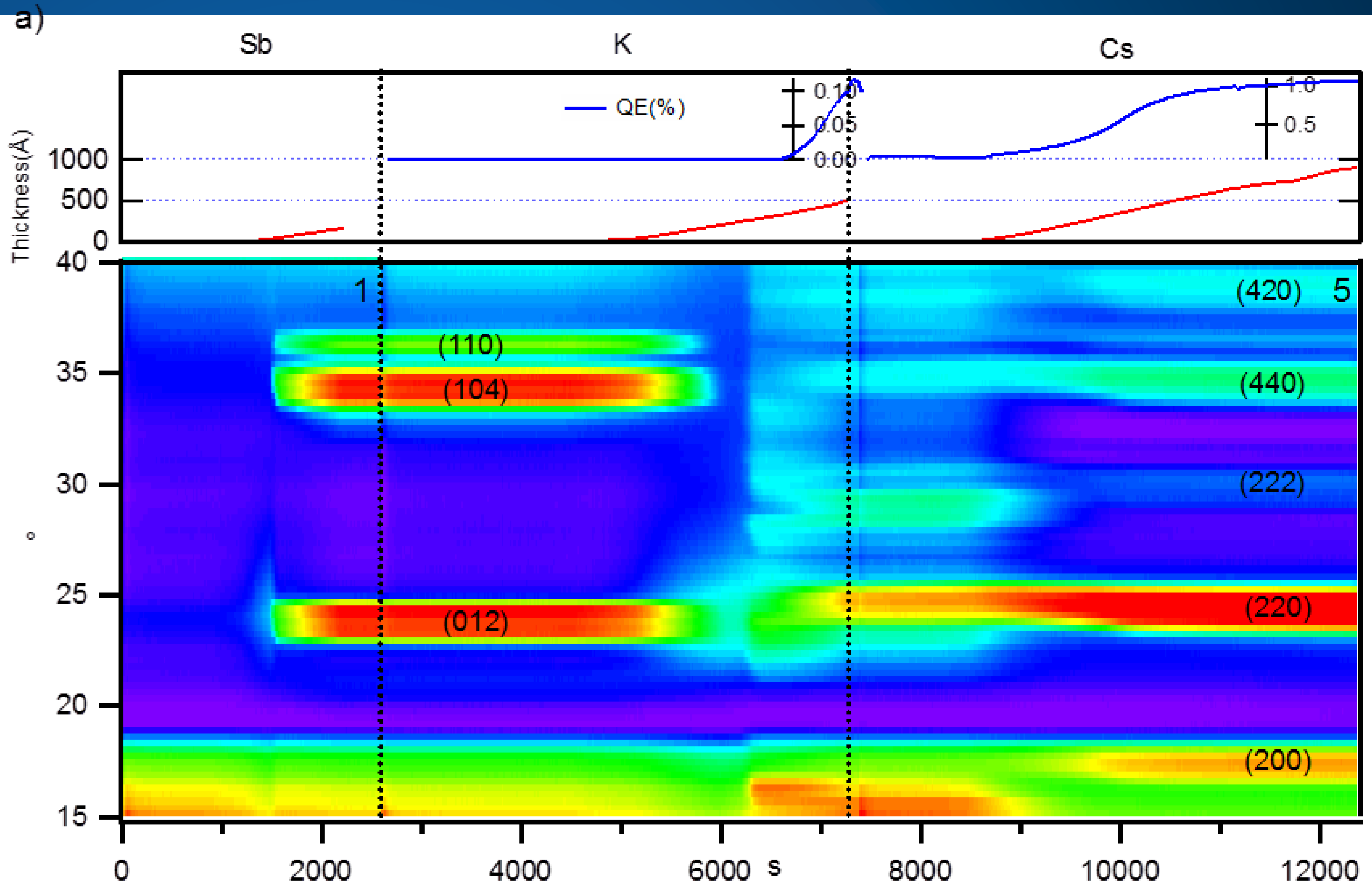
Sequential Evaporation Reaction Dynamics

Recipe:



QE during growth (532 nm laser)





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

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

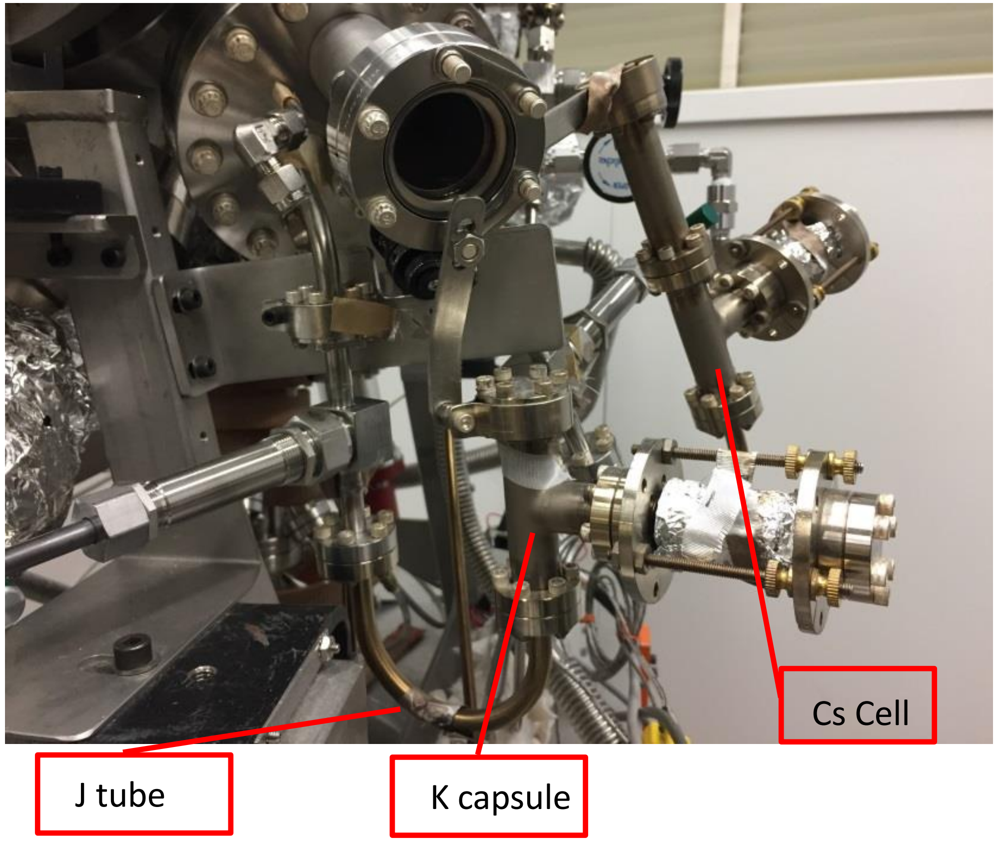
QE increase corresponds with K_xSb crystallization

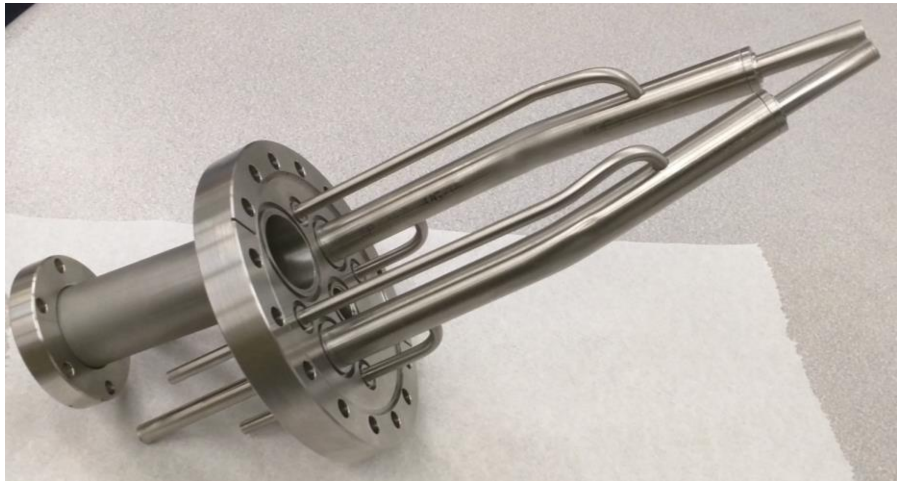
Cs increases lattice constant and reduces defects

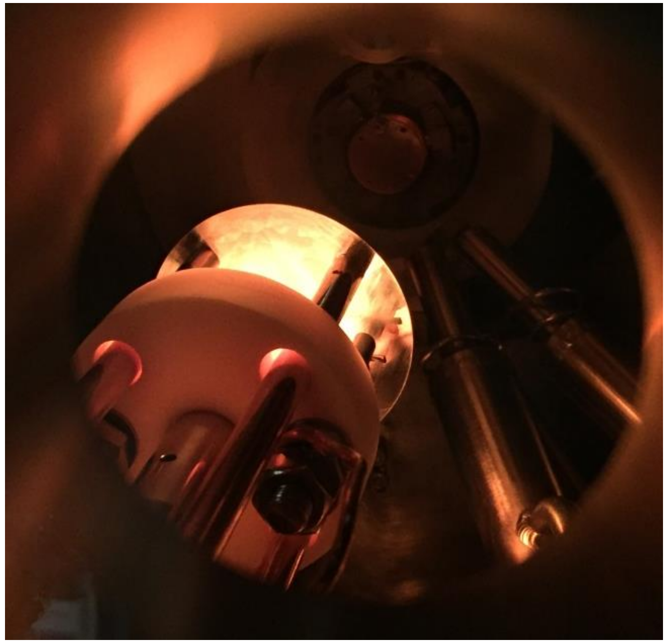
M. Ruiz-Osés et al., APL Mat. 2, 121101 (2014)

Ternary Co-evaporation

Simultaneously evaporate from Sb evaporator and K,Cs effusion cells







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

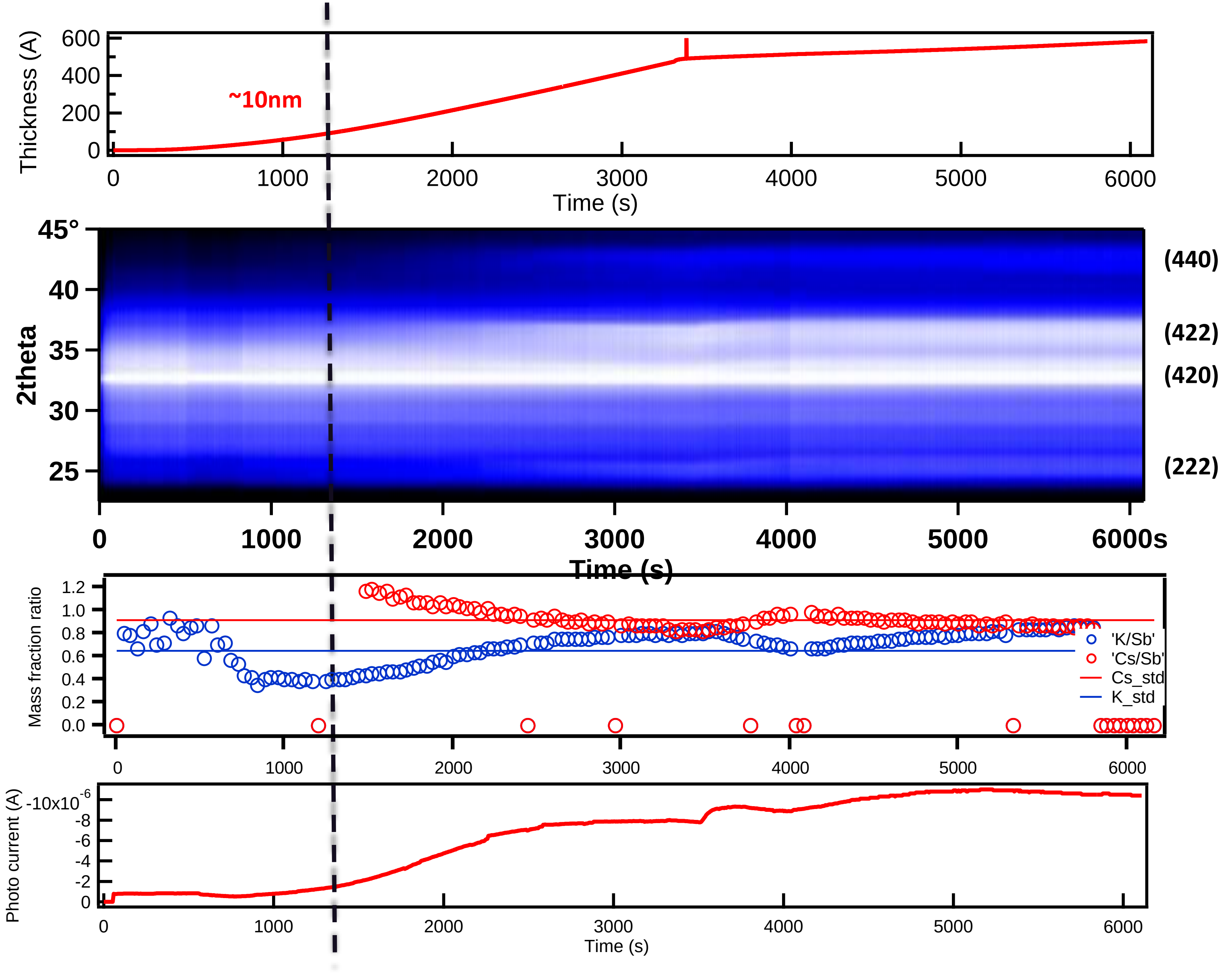
Stoichiometry controlled by real time XRF

Thickness

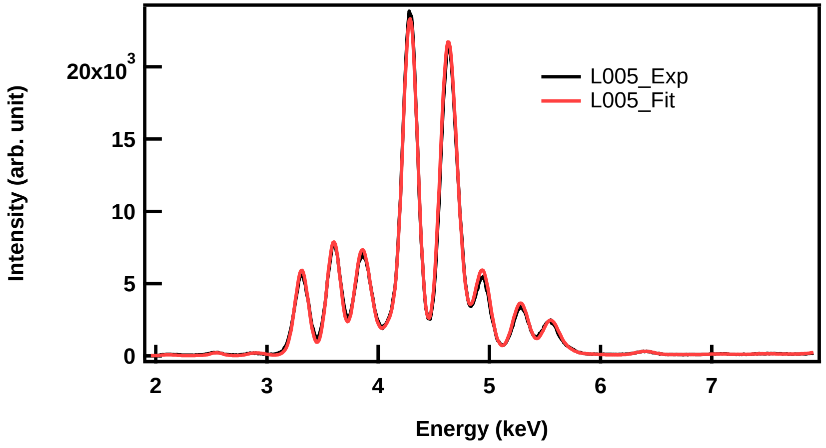
Real time XRD

Real time Fluorescence

QE



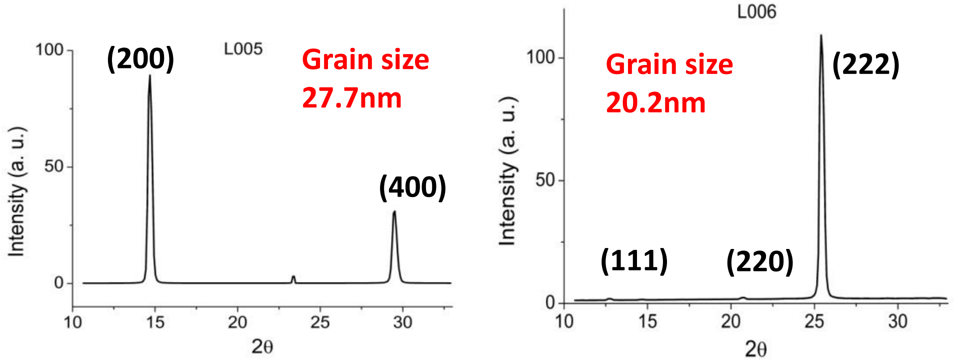
Stoichiometry & Structural Analysis



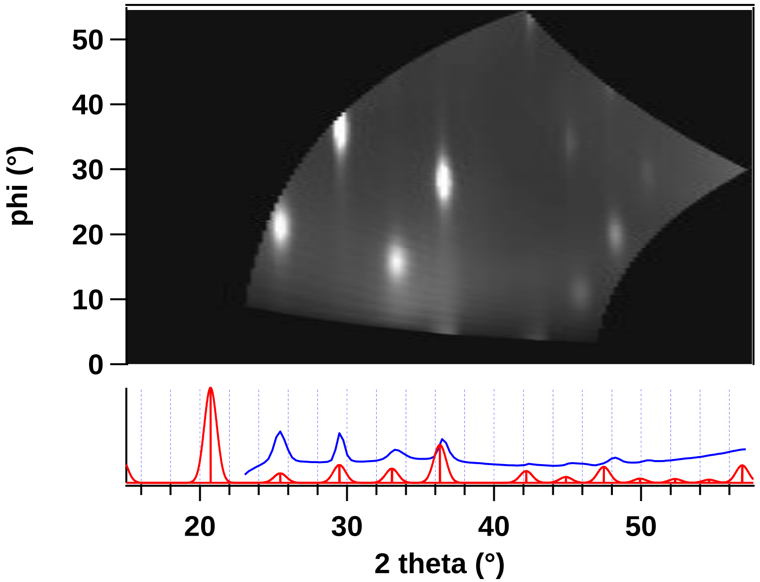
	K	Sb	Cs
L004 Si	2.50	1.00	1.16
L005 Si	2.37	1.00	0.91
L006 Si	2.21	1.00	0.95
L011 Si	2.07	1.00	0.94
L012 MgO	1.98	1.00	0.88

Good K/Cs/Sb ratio!

Camera 1



Camera 2

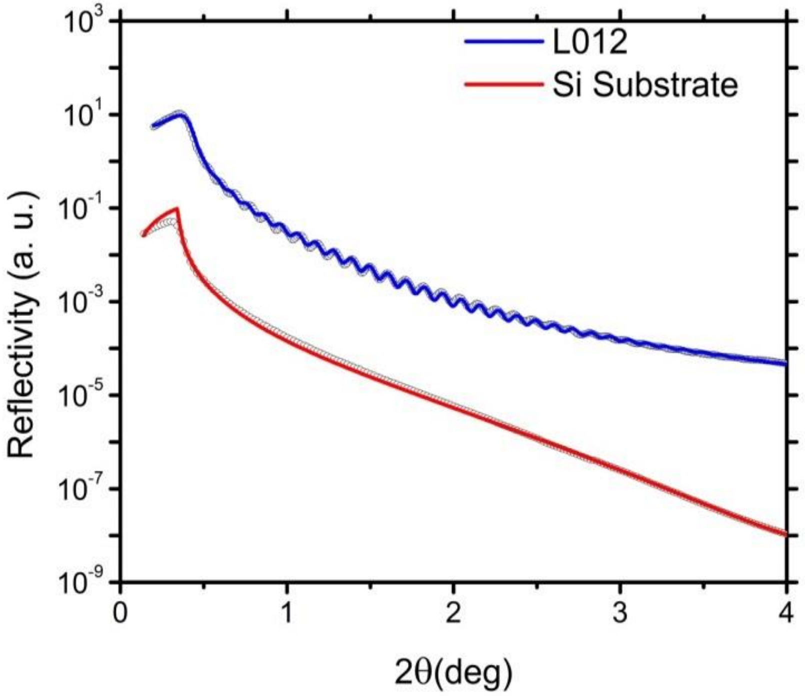


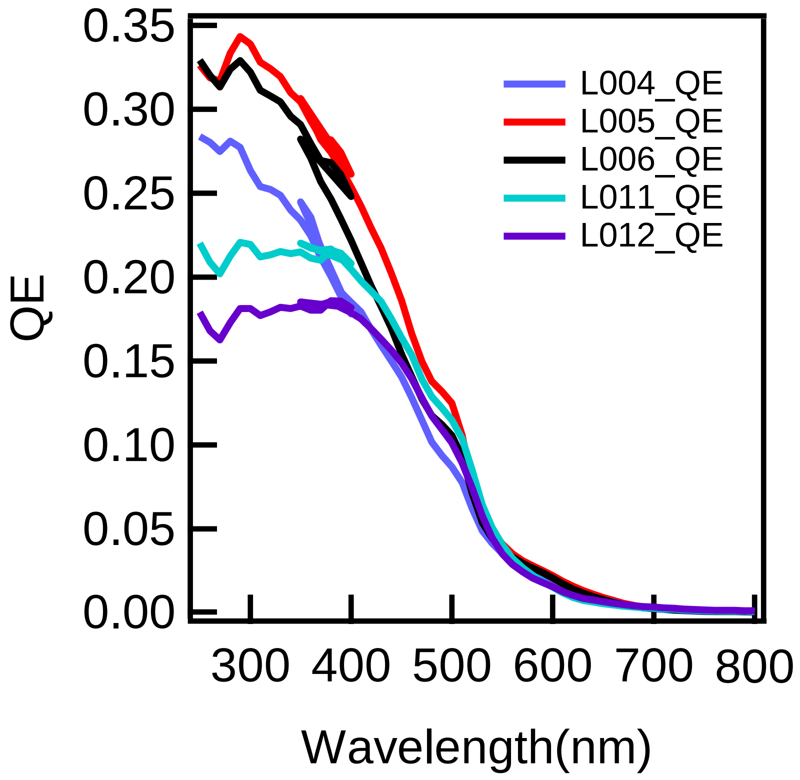
Highly textured K_2CsSb phase!

mm size lateral grains

Works for the entire Alkali antimonide family – we've created highly textured films with a wide range of stoichiometries

Surface Roughness & QE





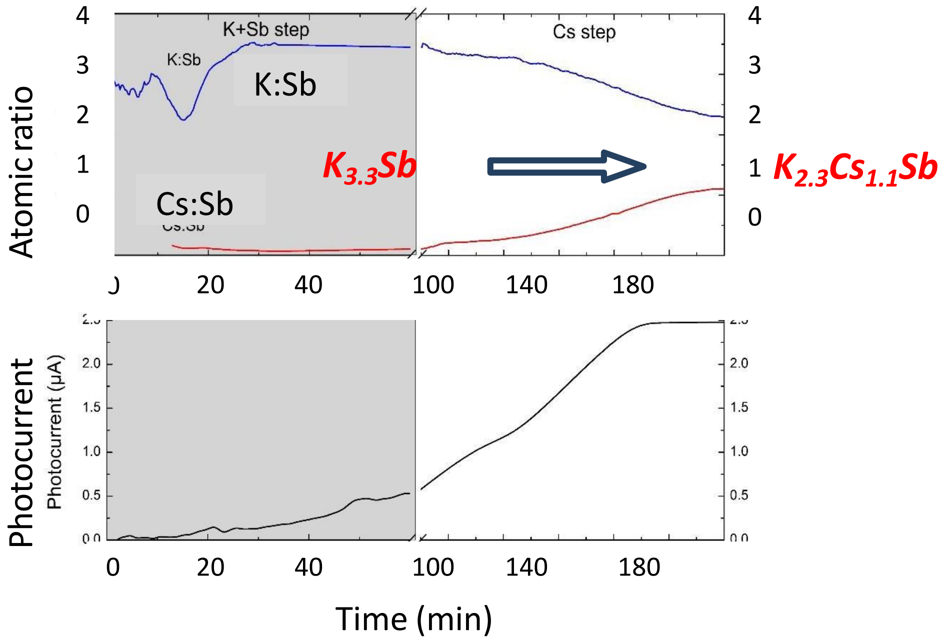
	QE@532nm(%)	Roughness(A)	Thickness (A)	Grain size (A)
L004 Si	4.9	3.5	234	155
L005 Si	5.8	11.5	815.3	277
L006 Si	5.4	13.8	757.5	202

Simultaneous evaporation of all constituents results in no crystal phase transformation
Smooth, reproducible and **ultra-high QE**. Highly Crystalline!

2-step Co-evaporation

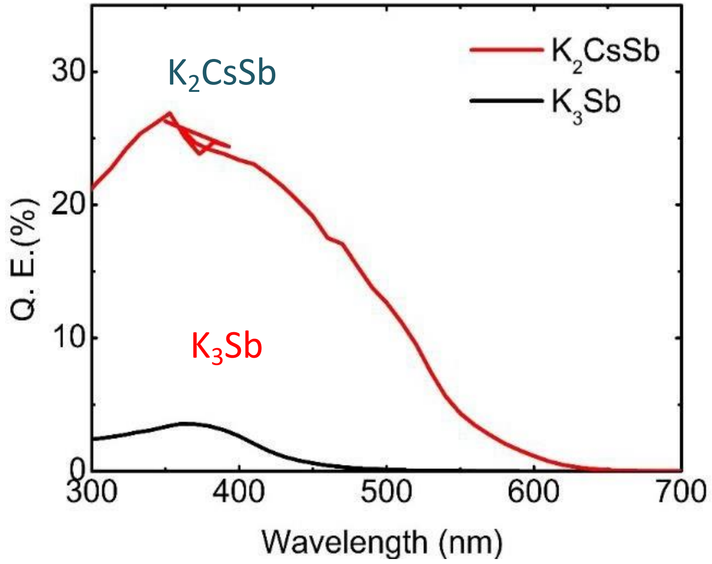
Two-step recipe:

1. K+Sb co-deposition, maximize QE at 380 nm, @ ~120℃
2. Cs deposition on top @ ~100℃ until QE (530 nm) maximizes



K:Sb becomes close to 3:1 shortly after K-Sb co-deposition starts

Spectral response:



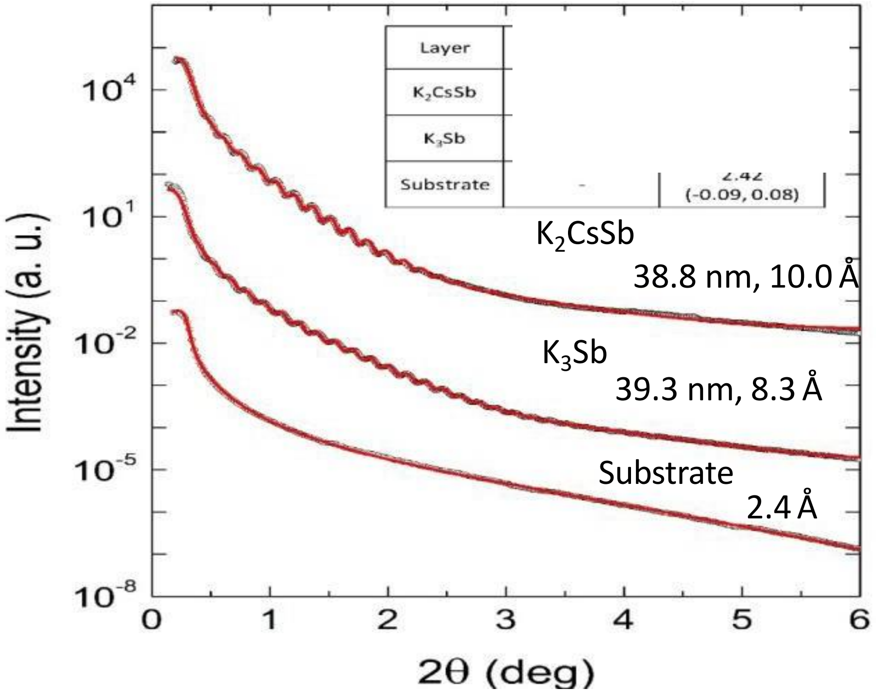
- K_3Sb layer: peak 3.5% at 360 nm;
  0.047% at 530 nm
- After Cs: peak 26% at 360 nm;
  7% at 530 nm

Most of the performance advantages of Ternary co-evaporation, but MUCH easier

2-step Co-evaporation

XRR

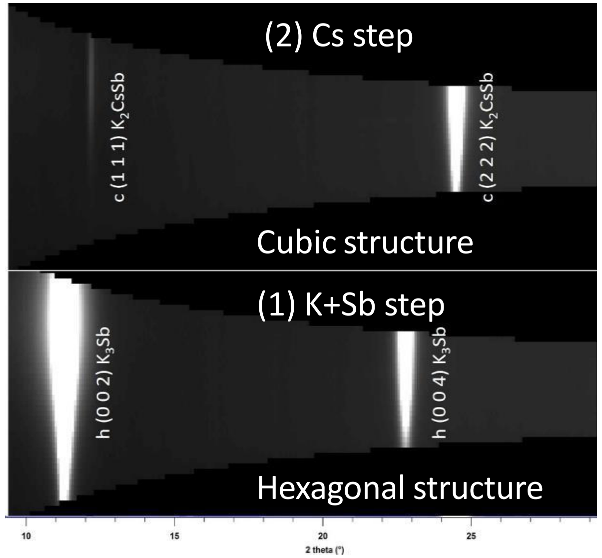
Sub-nm roughness
Similar thickness, roughness from K_3Sb to K_2CsSb



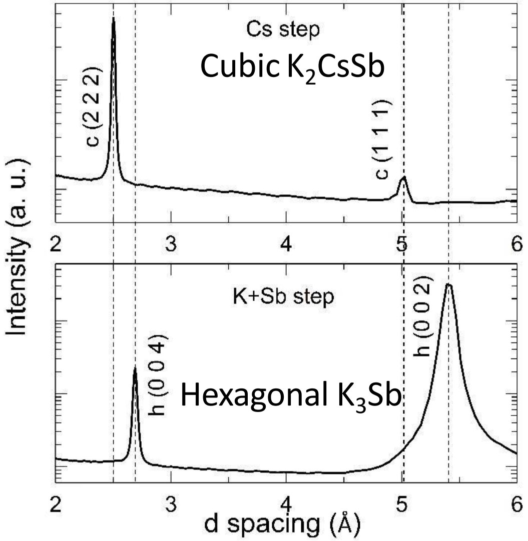
XRD

Full conversion from hexagonal K_3Sb to perfect cubic K_2CsSb
Textured XRD pattern

θ-2θ scan



θ-2θ scan

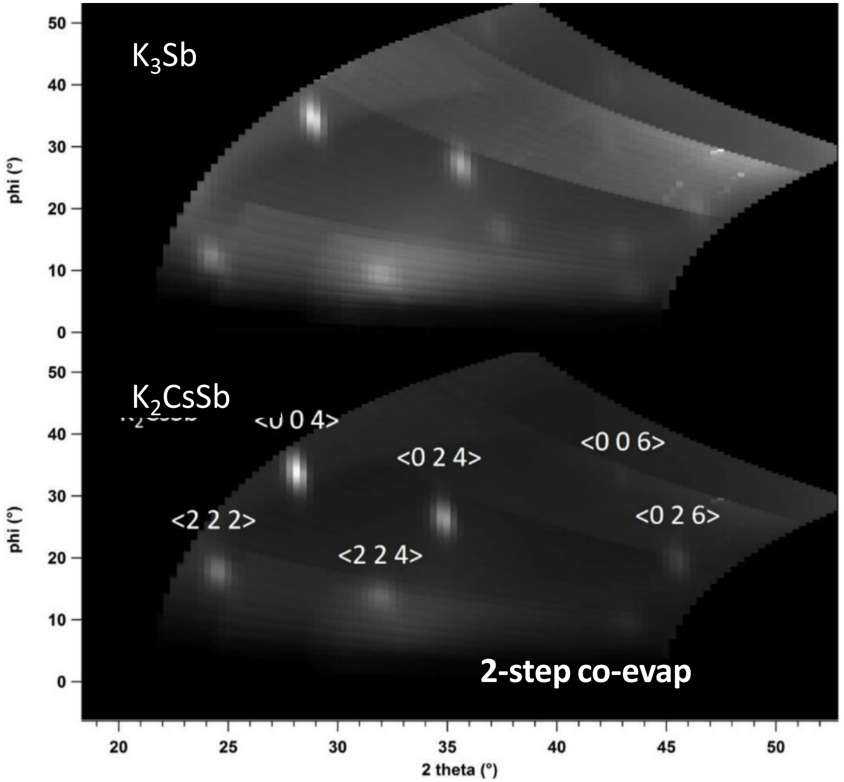


Diffraction arcs – textured film (both on Si (1 0 0) & Si (1 1 1))

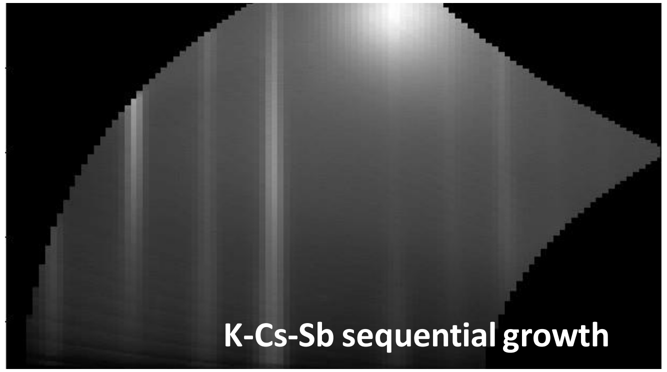
Crystal Quality

Wide angle X-ray diffraction patterns

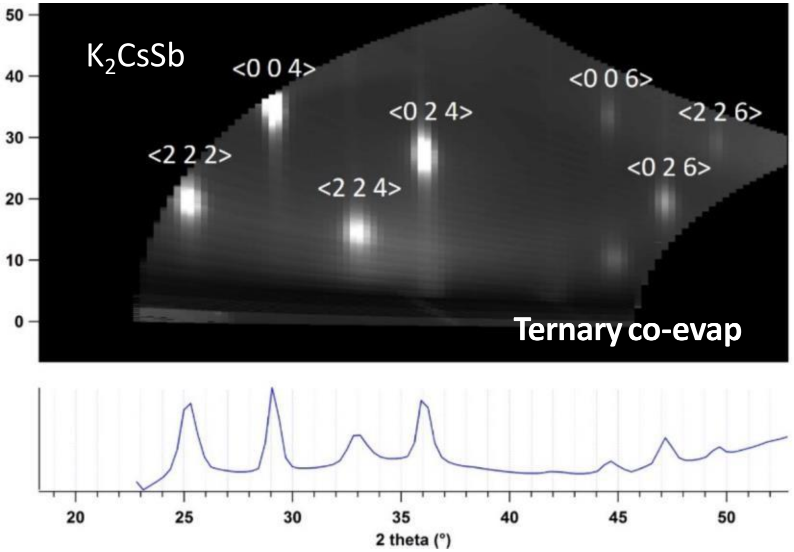
Two-step recipe



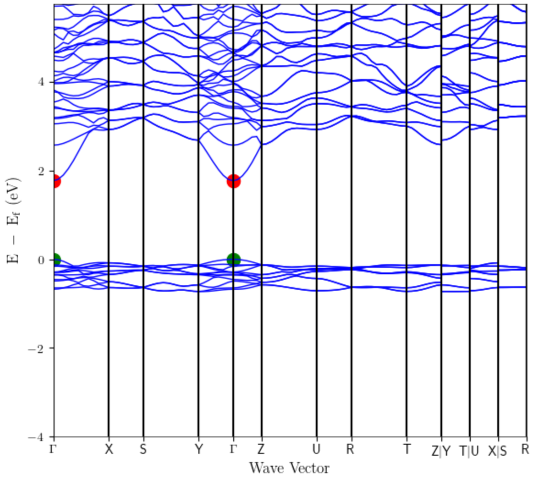
Large Grain, highly textured



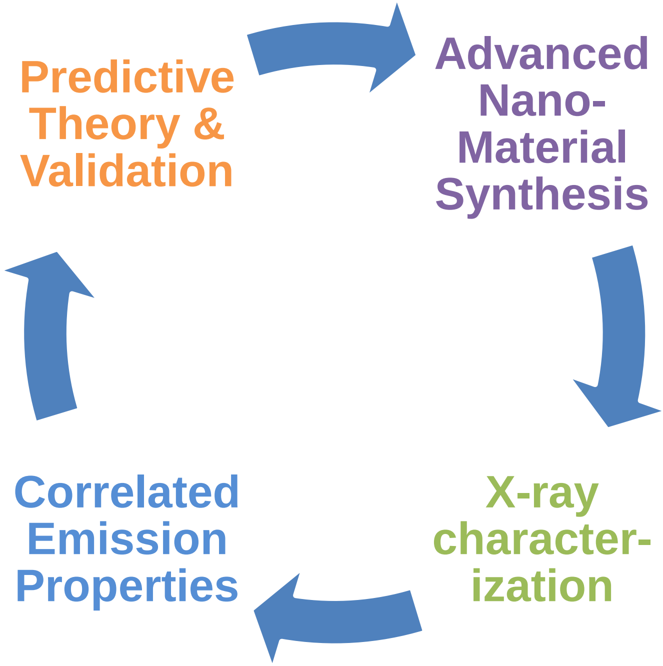
Ternary co-deposition



Example of success in one study being applied to adjacent problem: studies of K_2CsSb enabled growth of Cs2Te



HKL	Theory_d spacing	Exp_d spacing
222 Cs_2Te	2.307	2.315
111 Cs_2Te	4.613	4.615

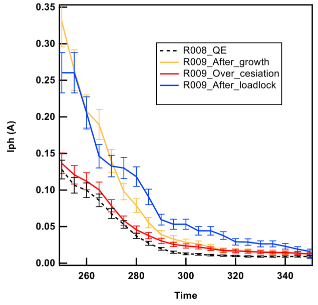


Predictive Theory & Validation

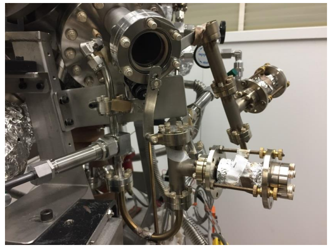
Advanced Nano-Material Synthesis

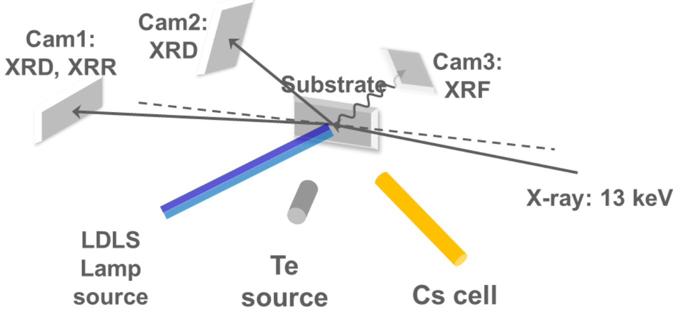
X-ray character-ization

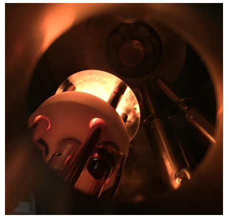
Correlated Emission Properties

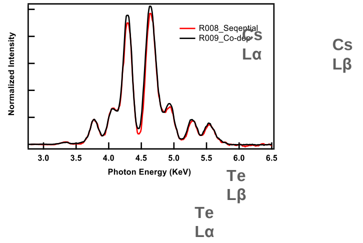


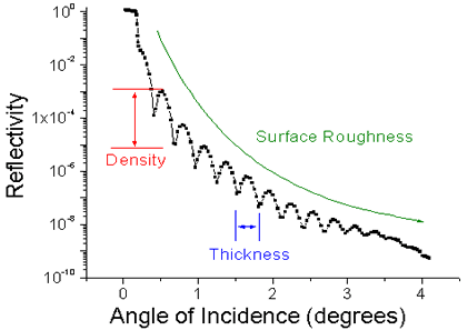
Next step: transfer to procedure not requiring synchrotron light source

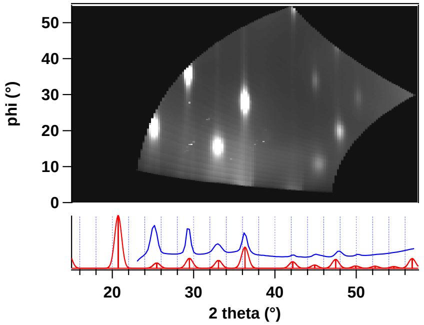




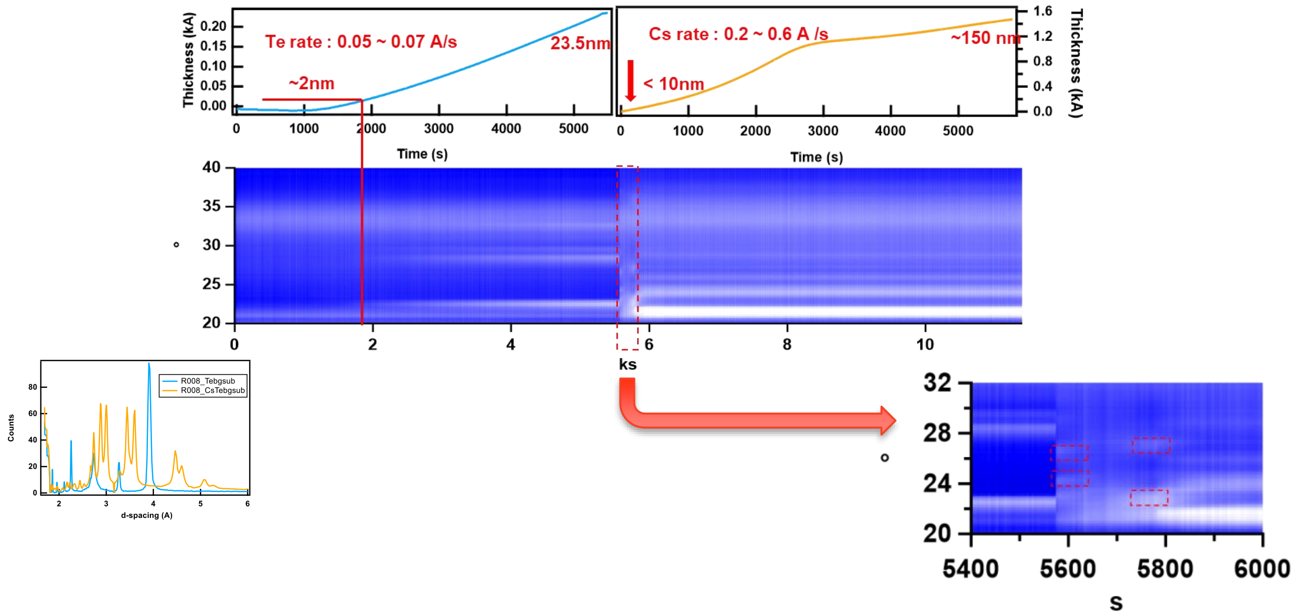


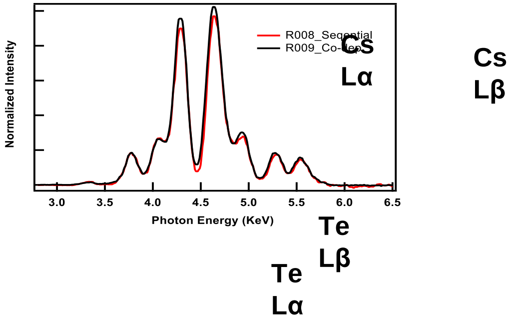


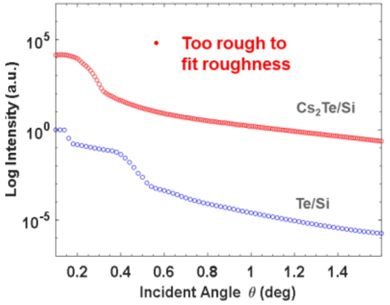




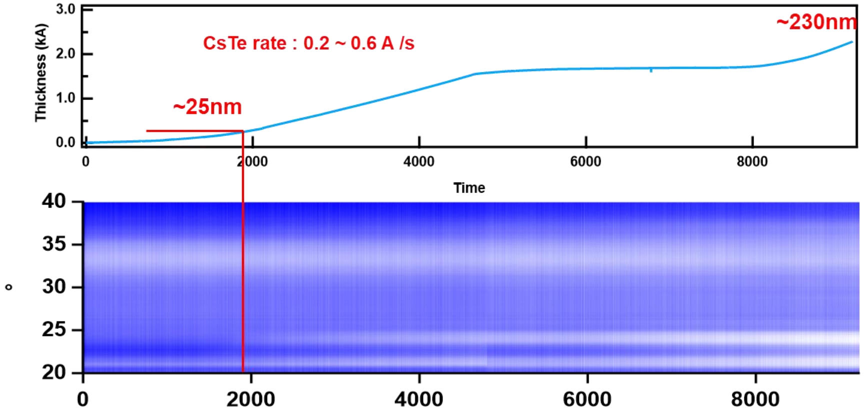
Results presented by Mengjia Gaowei at P3(2018) show the efficacy of the materials by design approach: co-deposition gives smooth single phase

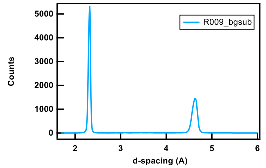


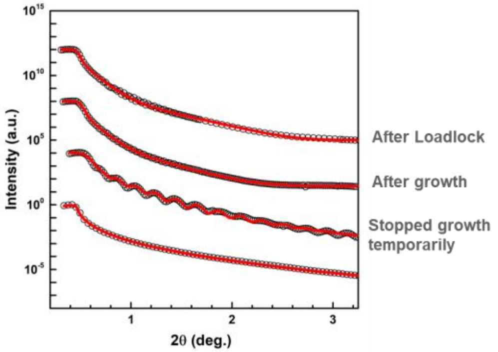




- Nearly all of the crystalized Te has dissolved
- Low counts in diffraction peaks
- Multiple phase of Cs-Te compound co-exist



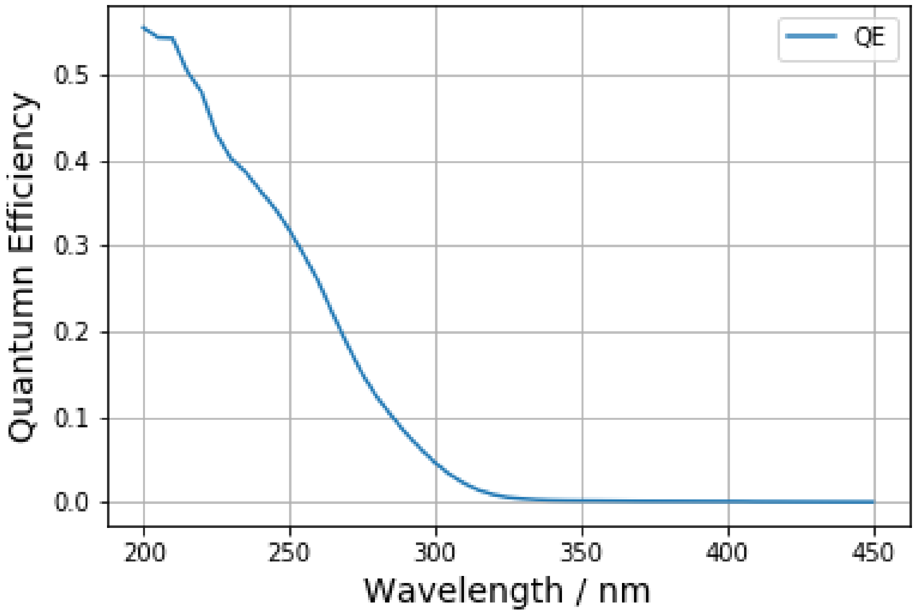


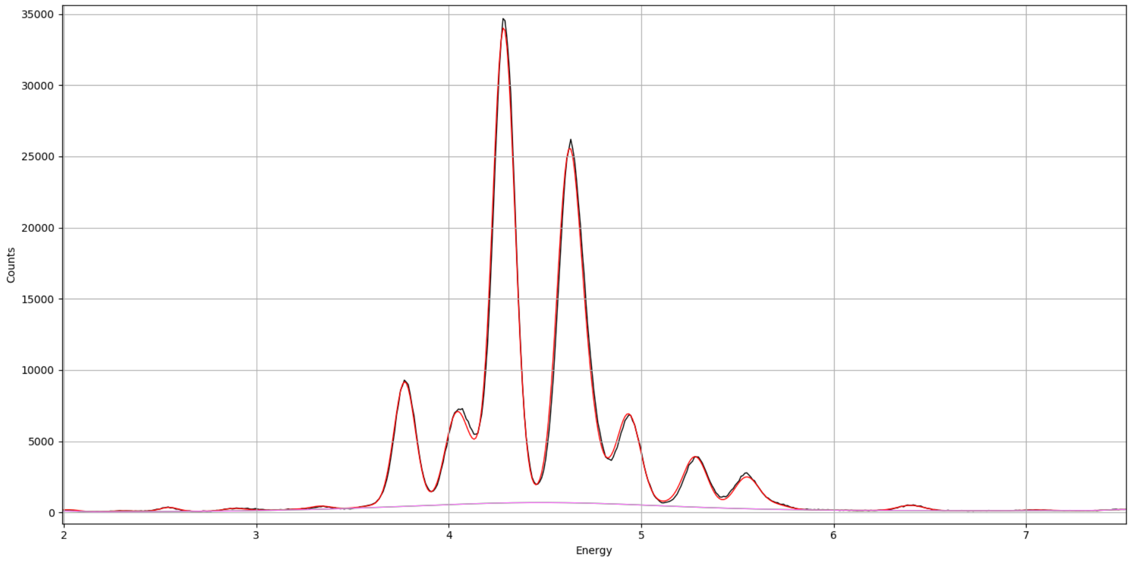


- Well crystalized
- Single phase

CsTe/Si: QE & XRF

CsTe	Te	Cs
Q007	1	1.98





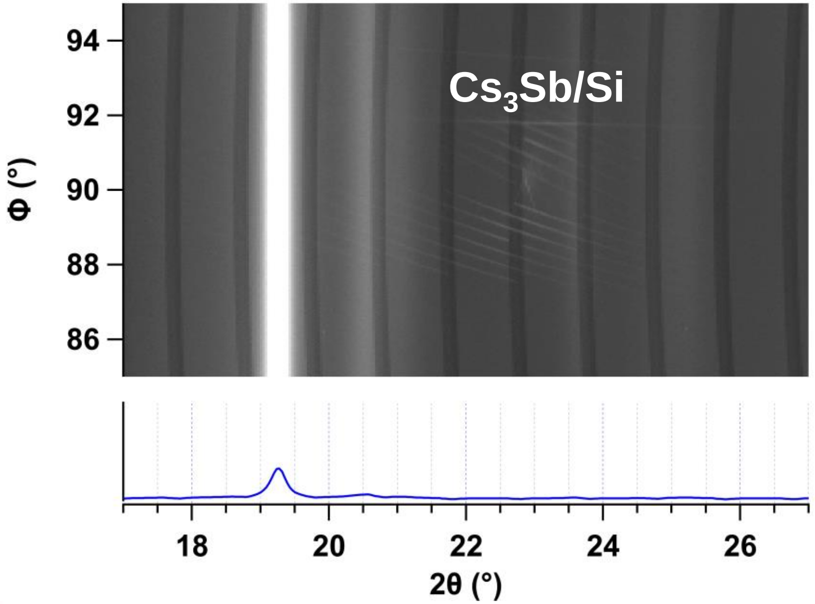
M. Gaowei, J. Sinsheimer, D. Strom, J. Xie, J. Cen, J. Walsh, E. Muller, and J. Smedley
Phys. Rev. Accel. Beams 22, 073401

Epitaxy

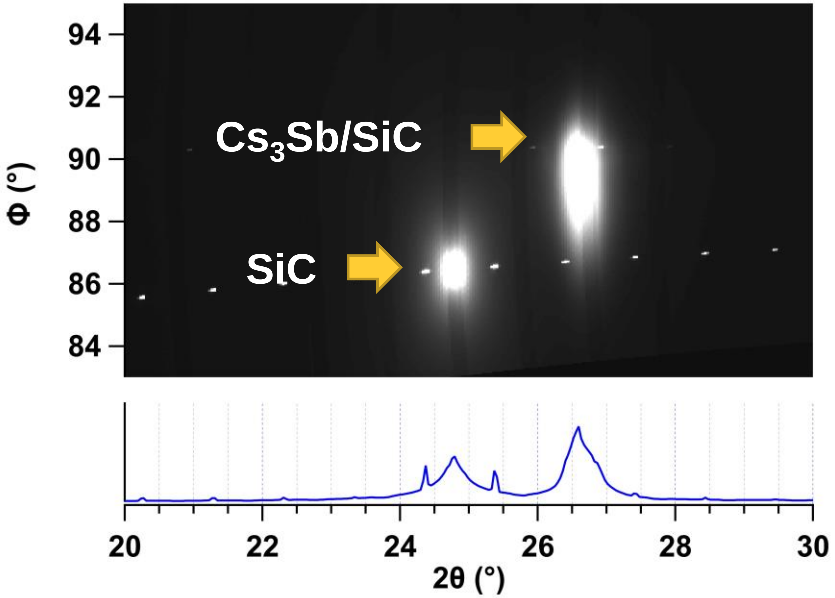
- Epitaxy – alignment of crystals to a substrate, to create a pseudo-single crystal
- Gateway to complex materials engineering and heterostructuring
- X-rays are a good tool, but in this case electron diffraction is vital

Co deposition of Cs_3Sb on 4-H SiC: Post growth Characterization

X-ray Diffraction

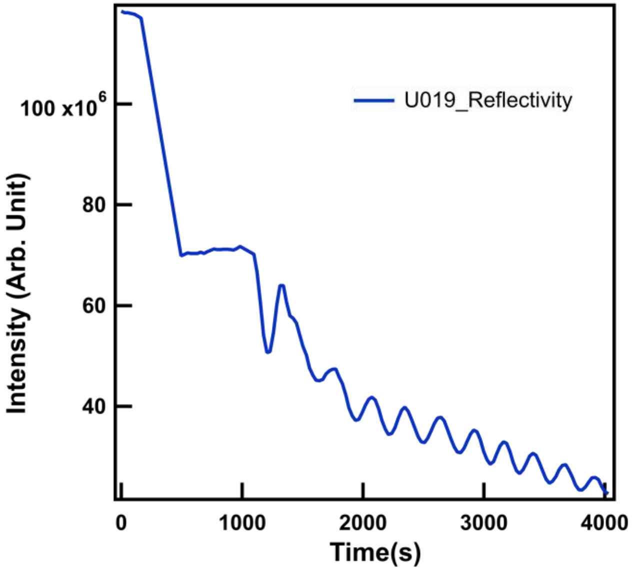


Film thickness 48.1 nm



Film thickness 20.5 nm

X-ray reflectivity during growth

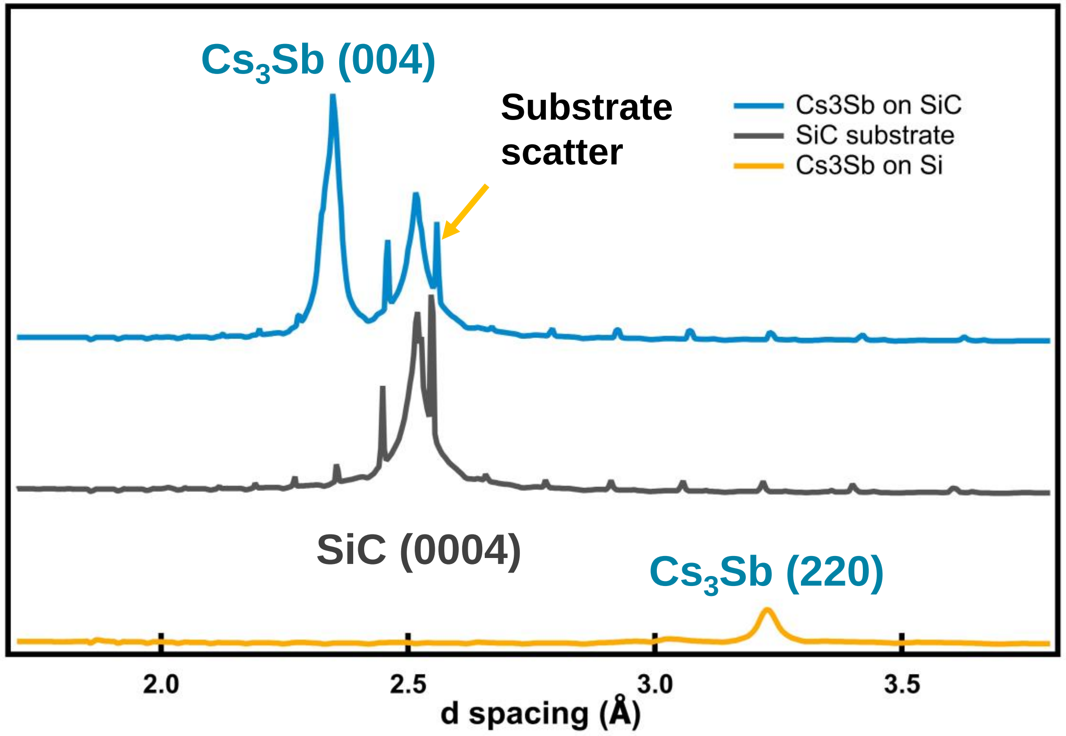


- Smooth film during growth
- Highly textured on SiC
- Possible epitaxy?

Brookhaven National Laboratory

Cs_3Sb on 4-H SiC:
Post growth Characterization

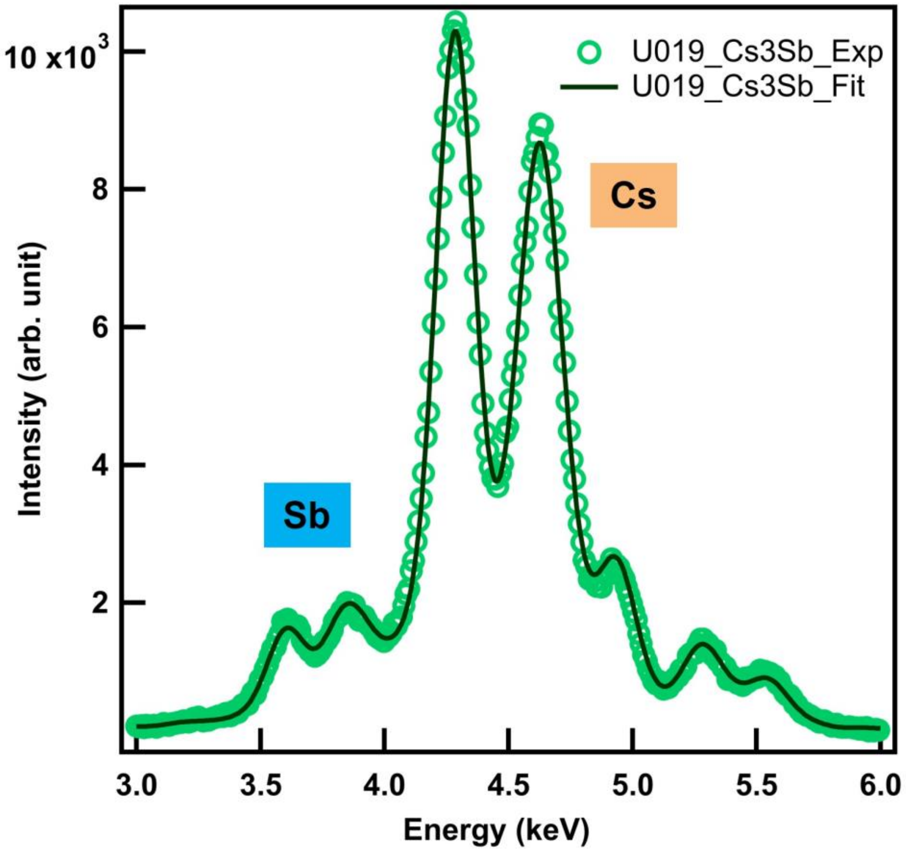
X-ray Diffraction



Diffraction peak	D spacing (Å)
Cs3Sb (004)	2.34
SiC (0004)	2.51
Cs3Sb (220)	3.23

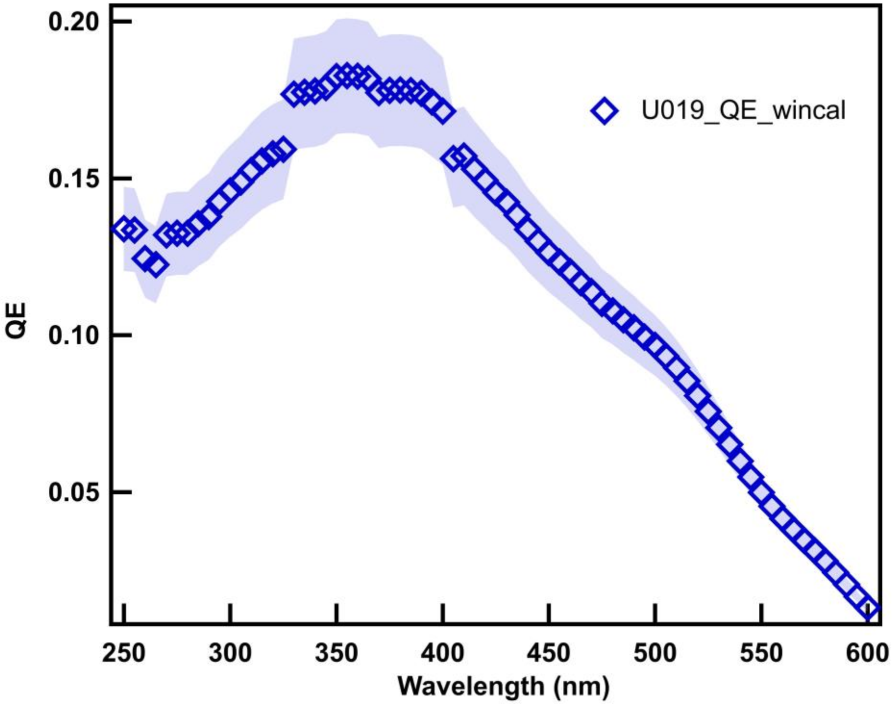
Cs_3Sb on 4-H SiC: Post growth Characterization

X-ray Fluorescence



	K/Sb	Cs/Sb
Cs3Sb/4H-SiC	/	2.95

Spectral Response



- 350 nm (peak): 18.2%
- 530 nm: 7%

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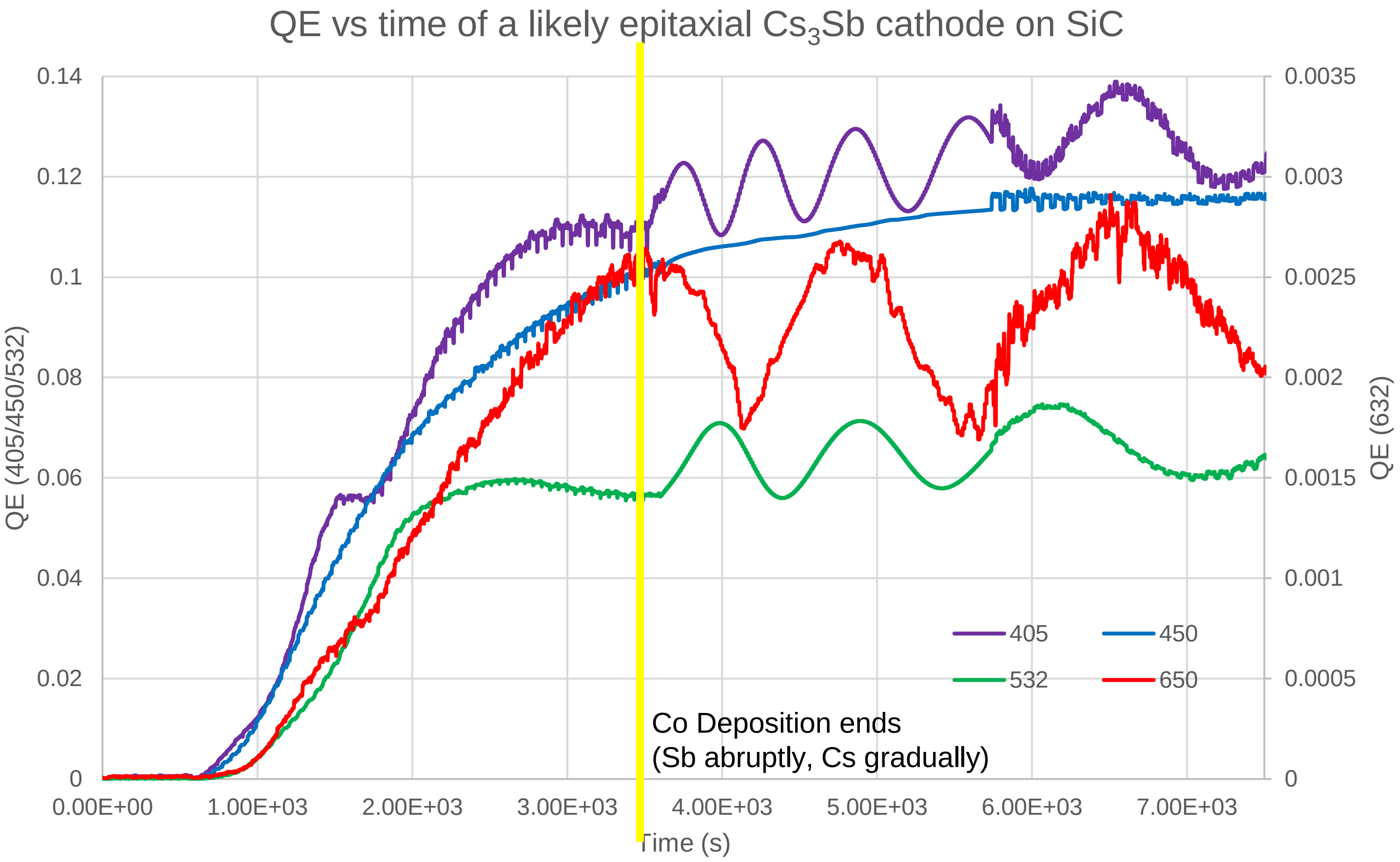
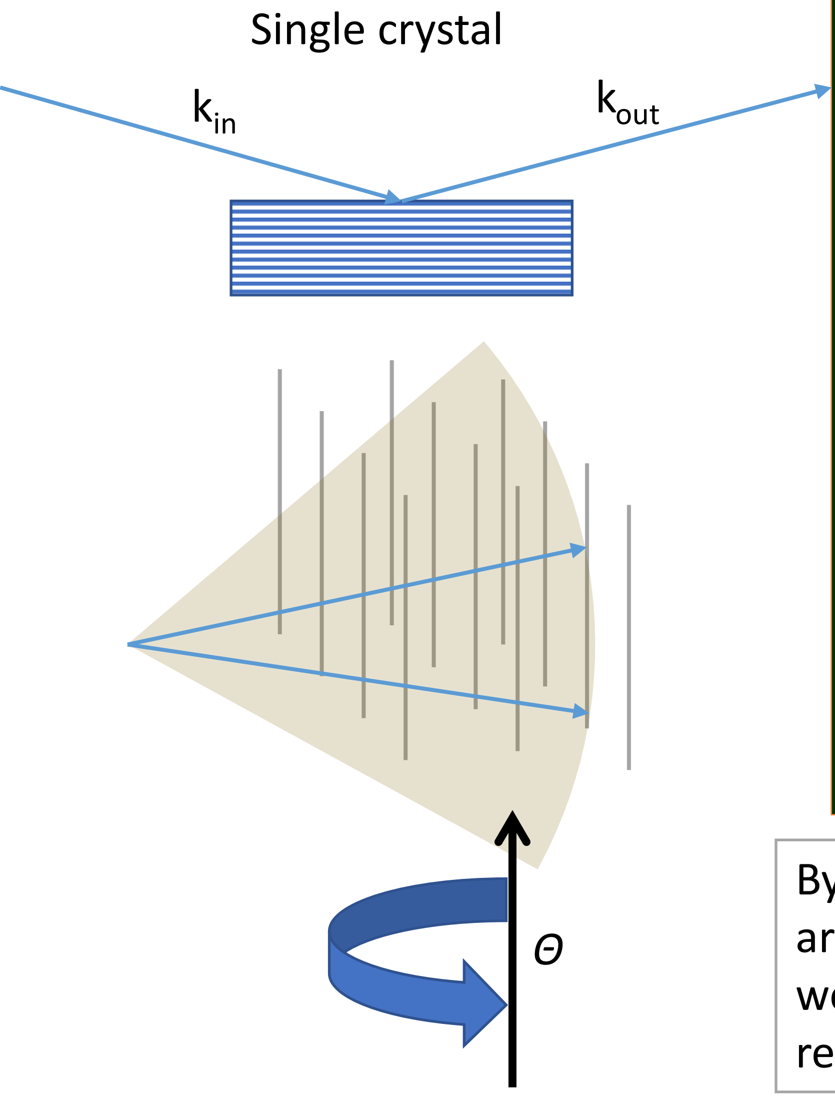
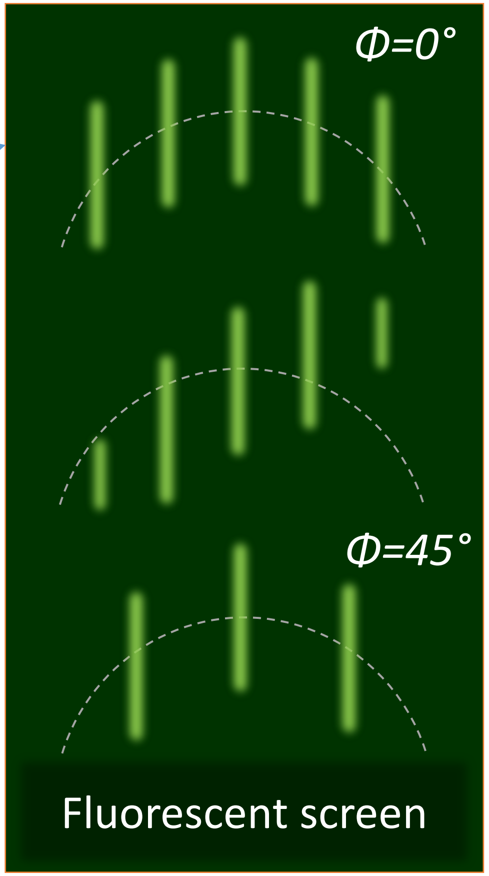
Cs_3Sb on 4-H SiC: QE and optical etalon effect



- Cathode is evolving after growth is stopped and substrate cooling down.
- Change of index of refraction
- Loss of material

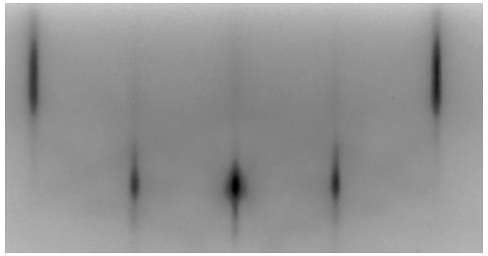
Information provided by RHEED



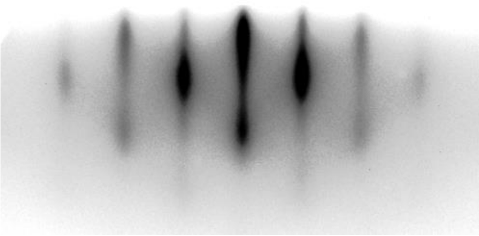


By rotating the sample around its surface normal, we intersect different sets of reciprocal space rods

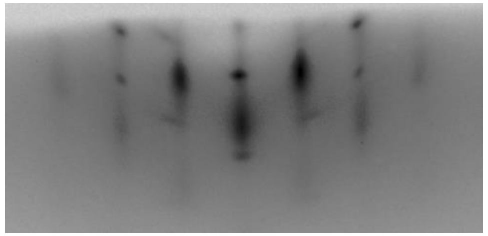
- Real-time
- Sub-ML sensitivity
- Qualitative probe of surface roughness and crystallinity



Single crystal
High coherence



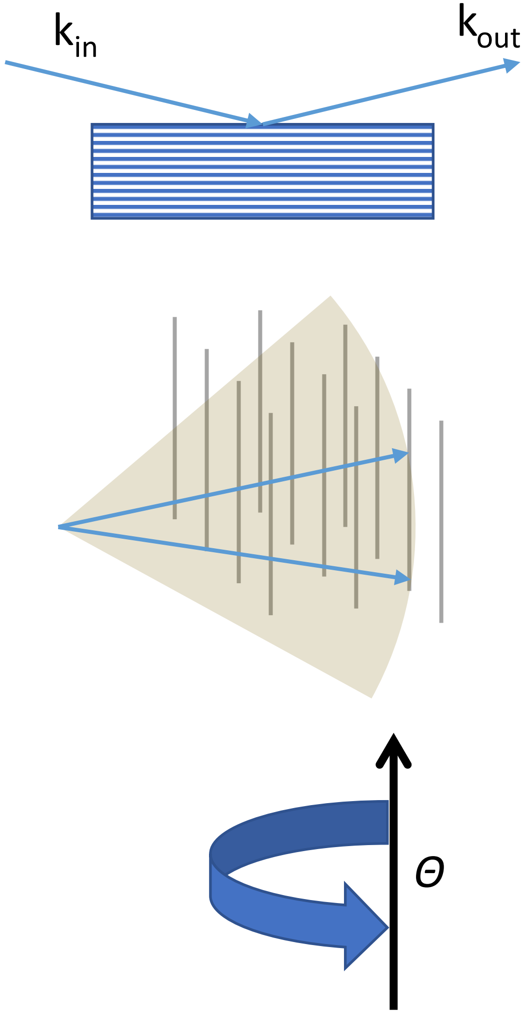
Film
Reduced coherence
Roughened surface

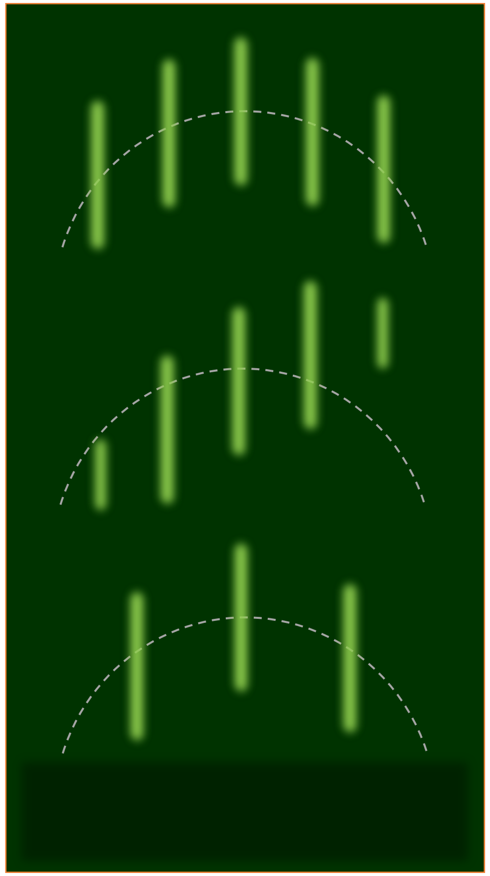


Film
Polycrystalline domains/impurities

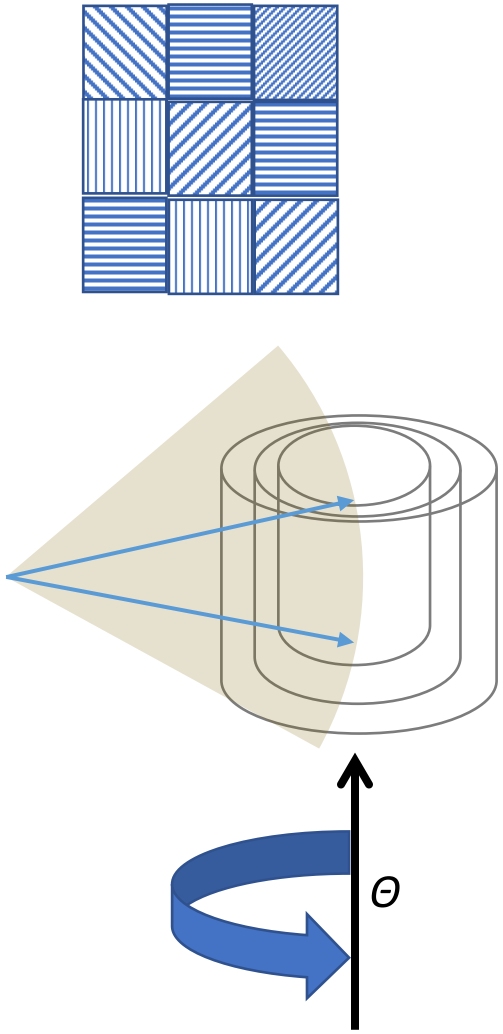
Information provided by RHEED

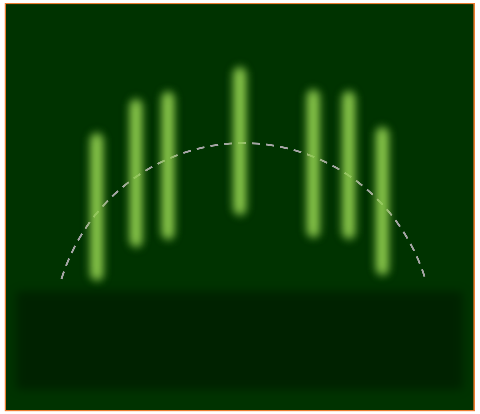
Single crystal





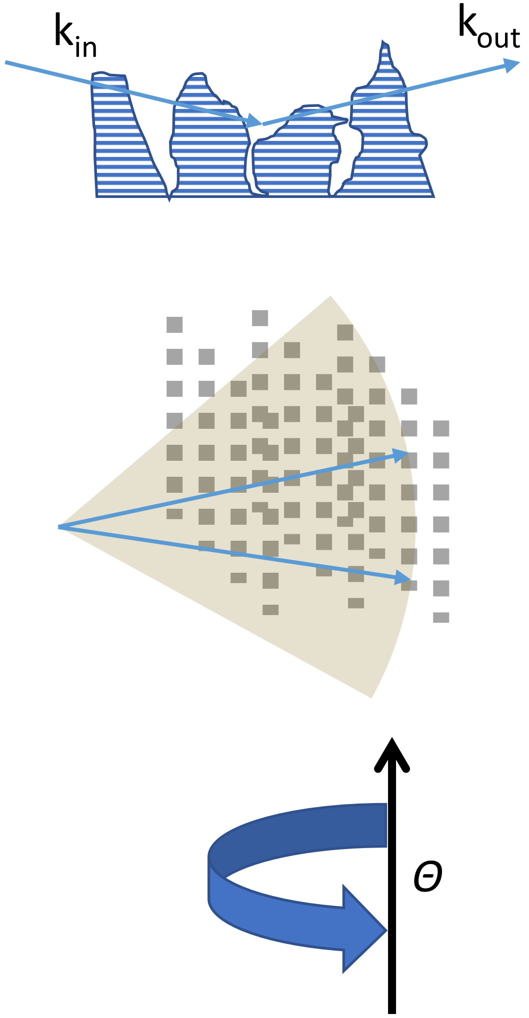
Fiber texture

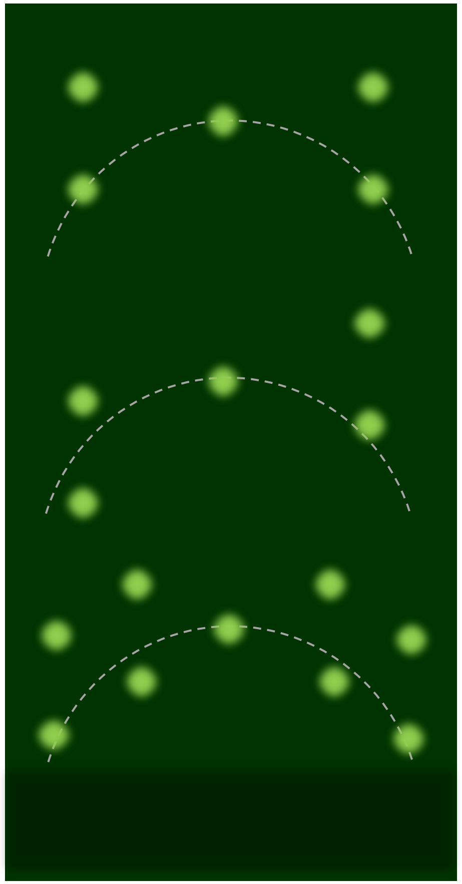




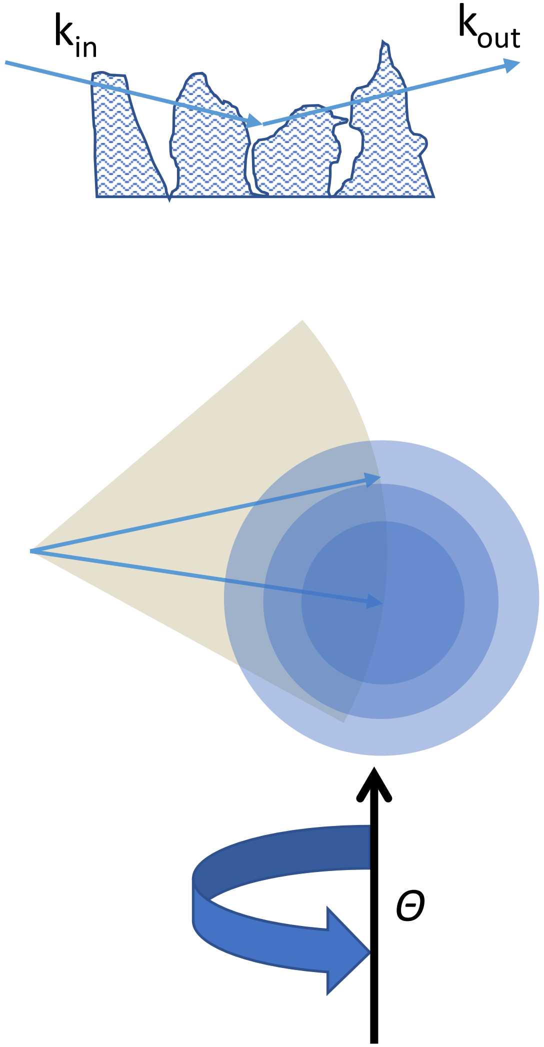
Information provided by RHEED

Single crystal islands





Polycrystalline islands



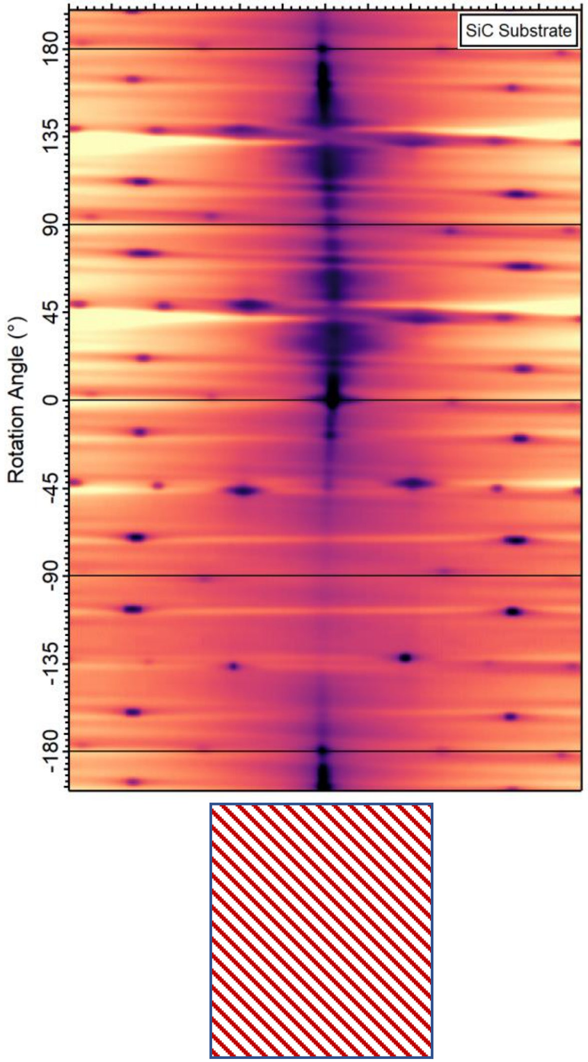
No texture

Textured film

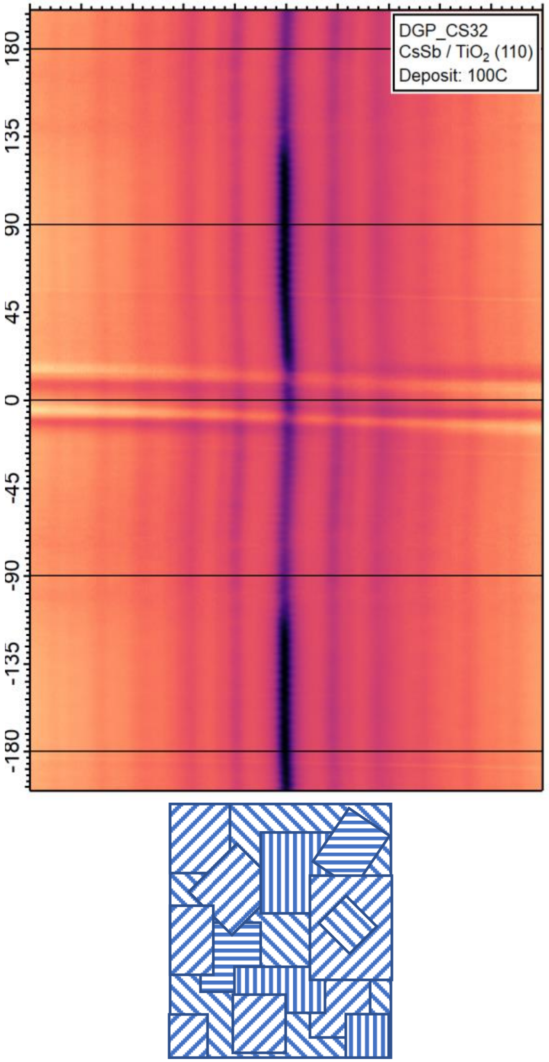
No rotation dependence if the texture axis is out-of-plane (uniaxial)
Rotation dependence if the texture axis is in-plane (biaxial)

Epitaxial Relationship

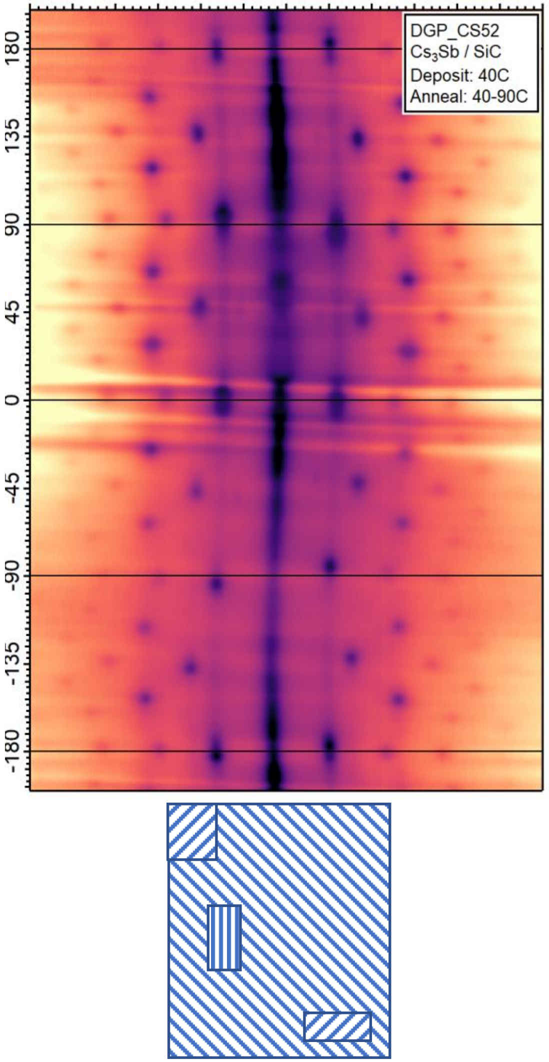
Substrate
Fully Ordered



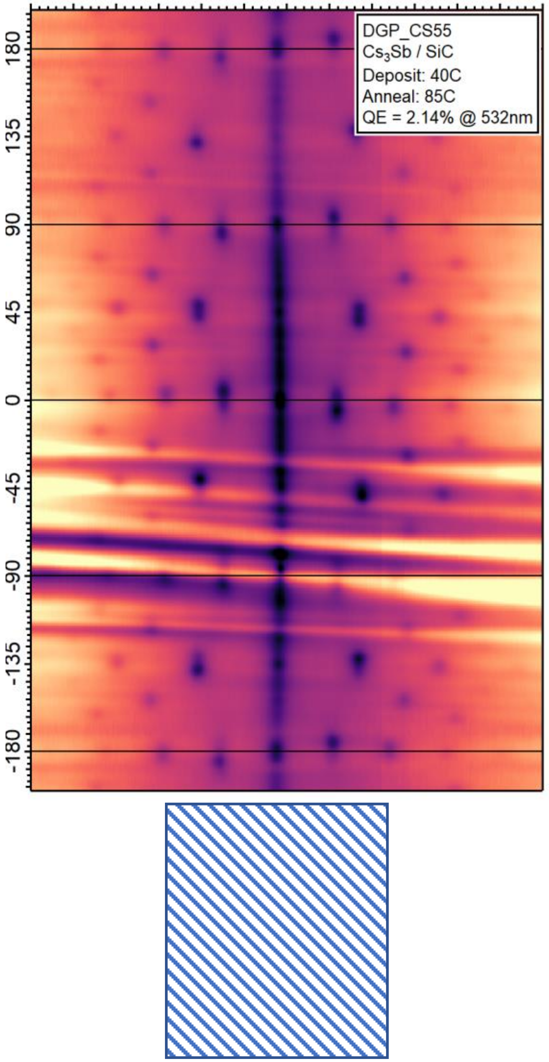
Fiber Textured Film
Only c-axis oriented



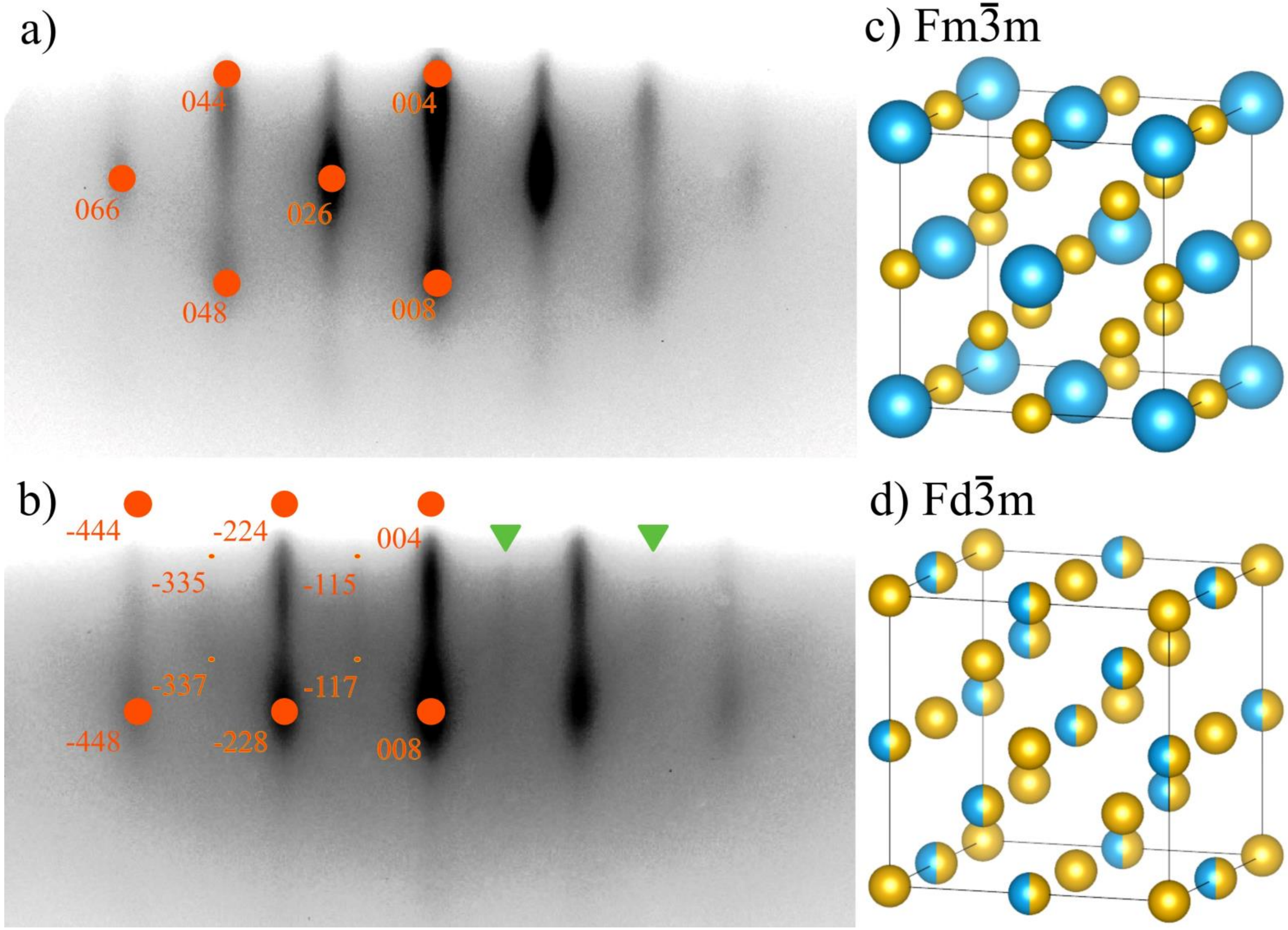
Partially Ordered Film



Epitaxial Film



RHEED of epitaxial Cs_3Sb



To prove that indeed that the epitaxial relationship is [100]SiC//[100]Cs3Sb I plot the reciprocal lattice of Fd-3m Cs3Sb superimposed to the RHEED patterns.

Half period streaks are predicted to appear in the [11] azimuth, as we observe.

Our data is compatible with any of the two literature reported structures.

Final Thoughts

- The materials science community has developed a robust toolkit for optimizing thin films for various applications
- For materials where “dual uses” exist (GaAs), nearly perfect crystals, with heterostructuring, doping gradients and such exist
- For the PEA semiconductors, we still have work to do, but it is an exciting time. The dawn of epitaxy in these systems heralds a revolution in optimization.

To-Do list:

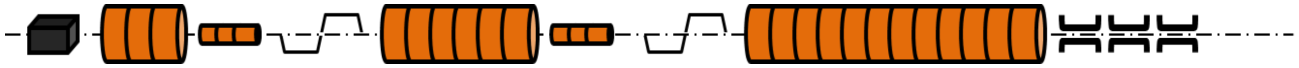
  Cross-correlate X-ray techniques with non-synchrotron measurements

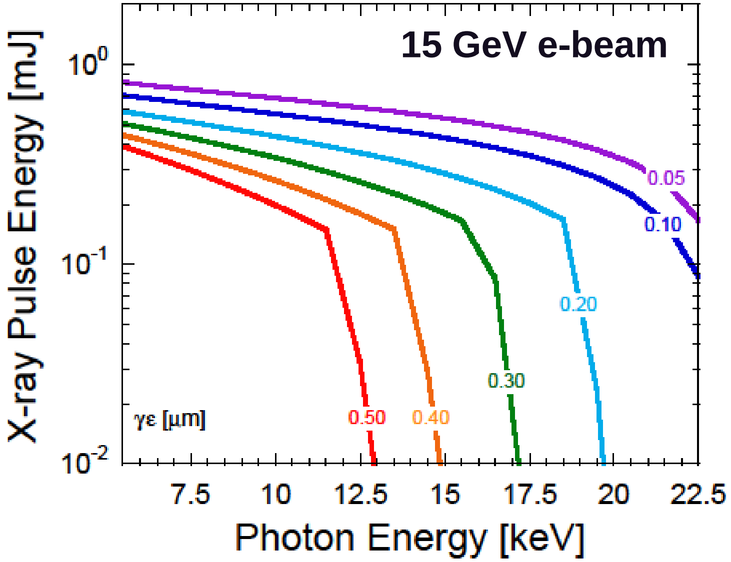
  Develop epitaxy for Ternary systems and Cs_2Te

  Demonstrate functional heterostructures

  Measure conductivity vs temperature – better crystals = less resupply

Emittance leverage for XFELs: Electron source sets ultimate limits on achievable electron beam quality, which sets ultimate limits on photon beam





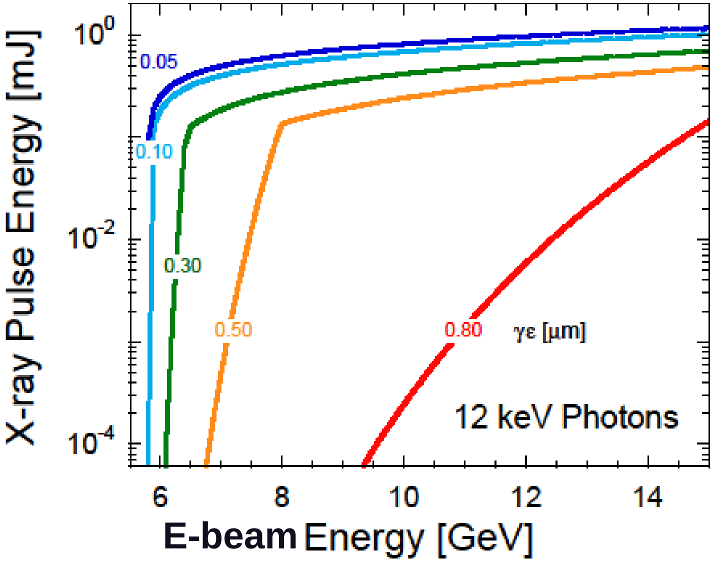
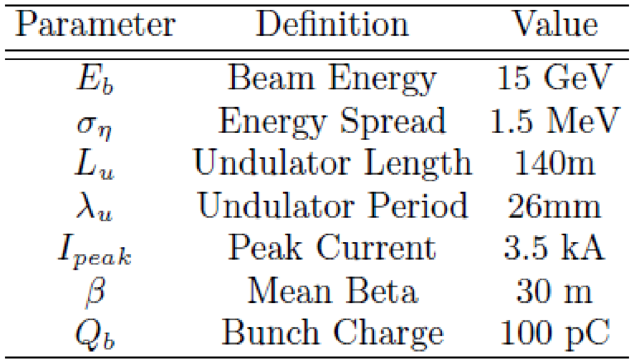


TABLE II. LCLS-II/HXR Case Study Parameters



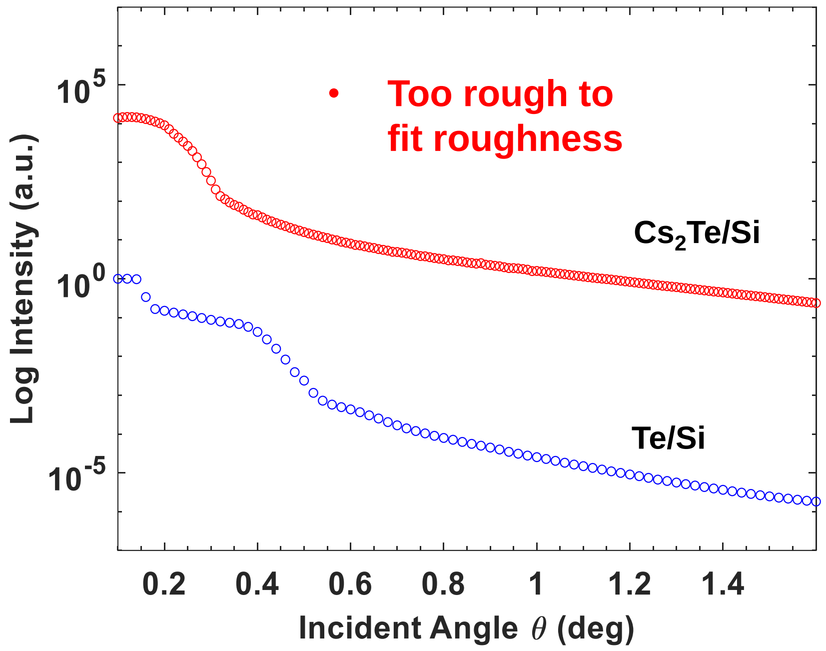
Parameter	Definition	Value
E_b	Beam Energy	15 GeV
σ_η	Energy Spread	1.5 MeV
L_u	Undulator Length	140m
λ_u	Undulator Period	26mm
I_{peak}	Peak Current	3.5 kA
β	Mean Beta	30 m
Q_b	Bunch Charge	100 pC

- ***Photocathode upgrade presents a low-cost investment for improved performance of existing machines***
- ***Emittance suppression at the cathode enables machine designs with lower emittance budget***

Moody, N.A., et al., *Perspectives on Designer Photocathodes for X-ray Free-Electron Lasers: Influencing Emission Properties with Heterostructures and Nanoengineered Electronic States.* Physical Review Applied, 2018. **10**(4): p. 047002.

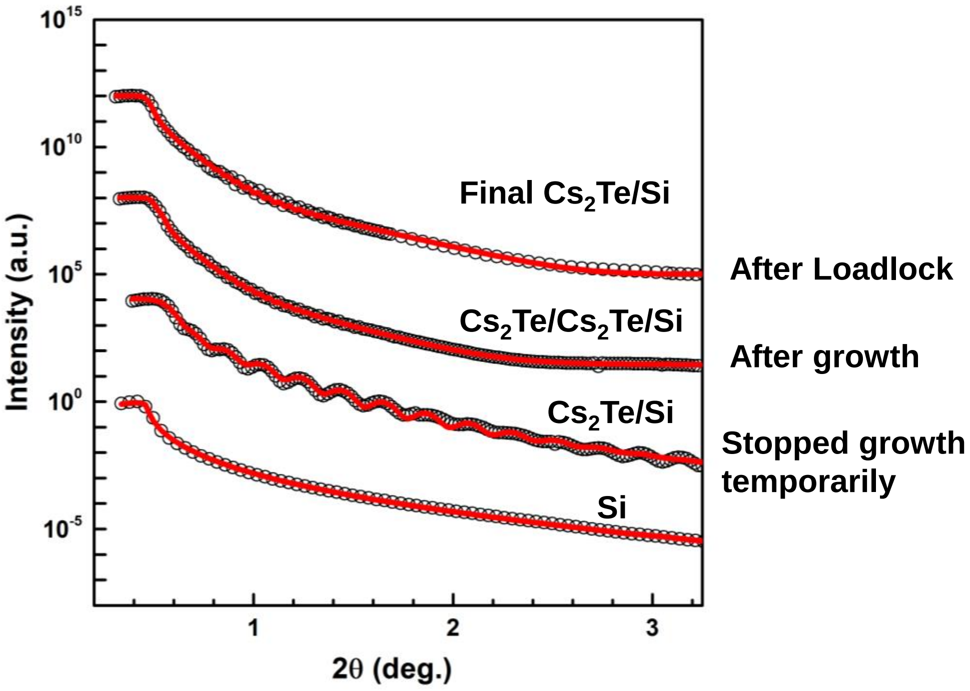
CsTe cathode surface roughness: XRR analysis

R008 Sequential - CsTe on Si



- Too rough to fit roughness

R009 Co-dep - CsTe on Si



After Loadlock

After growth

Stopped growth temporarily

	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