Bright Sources for Accelerators

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Overview

Beam Parameters and Overview of cathode types

Why the source matters… and sometimes doesn't

Brief intro to 3-step model as it applies to semiconductors and metals

Where do we need to be? What is limiting us now?

In situ materials analysis during cathode formation

How to grow smoother cathodes (and why you might want to)

Reference Material

Some of this talk comes from a course on Cathode Physics Matt Poelker and I taught at the US Particle Accelerator School

http://uspas.fnal.gov/materials/12UTA/UTA-Cathode.shtml

Modern Theory and Applications of Photocathodes
W.E. Spicer & A. Herrera-Gómez
SAC-PUB-6306 (1993)

Great Surface Science Resource:
http://www.philiphofmann.net/surflec3/index.html

What matters?

- For Colliders and electron coolers, it is often Luminosity (particle density) that matters
- For light sources and electron diffraction/microscopy, it is Brightness: $B = \frac{N_e}{\varepsilon_{nx}\varepsilon_{ny}\varepsilon_{nz}}$
  - large number of electrons in a small volume of phase space
  - Phase space volume is a conserved quantity under forces which conserve energy and particle number
- Some applications depend on emittance (FEL, coolers)
  - Determines the electron energy required for an X-FEL at a given wavelength $\varepsilon \approx \frac{\lambda}{4\pi} \Rightarrow \frac{\varepsilon_n}{\beta\gamma} \approx \frac{\lambda}{4\pi}$
- High Quantum Efficiency, High Average Current, Long Operational Lifetime, Vacuum requirements, Response time

Does the particle source matter?... Sometimes

- The electron beam properties determine the photon beam properties
  - Pulse duration, degree of coherence, flux
- In all light sources through 3rd generation, the phase space is determined by the ring

- In X-ray free electron lasers (LCLS II, XFEL, MaRIE), this will change – the electron source will determine the beam properties
- The highest brightness sources available are photoinjectors, which use a laser on a photocathode to control the spatial and temporal profile of the emitted electron beam

Applications at the State of the Art

Electron cooling of ion machines

Requires high current with long operational life, other requirements are modest (~50 mA with 5μm emittance)

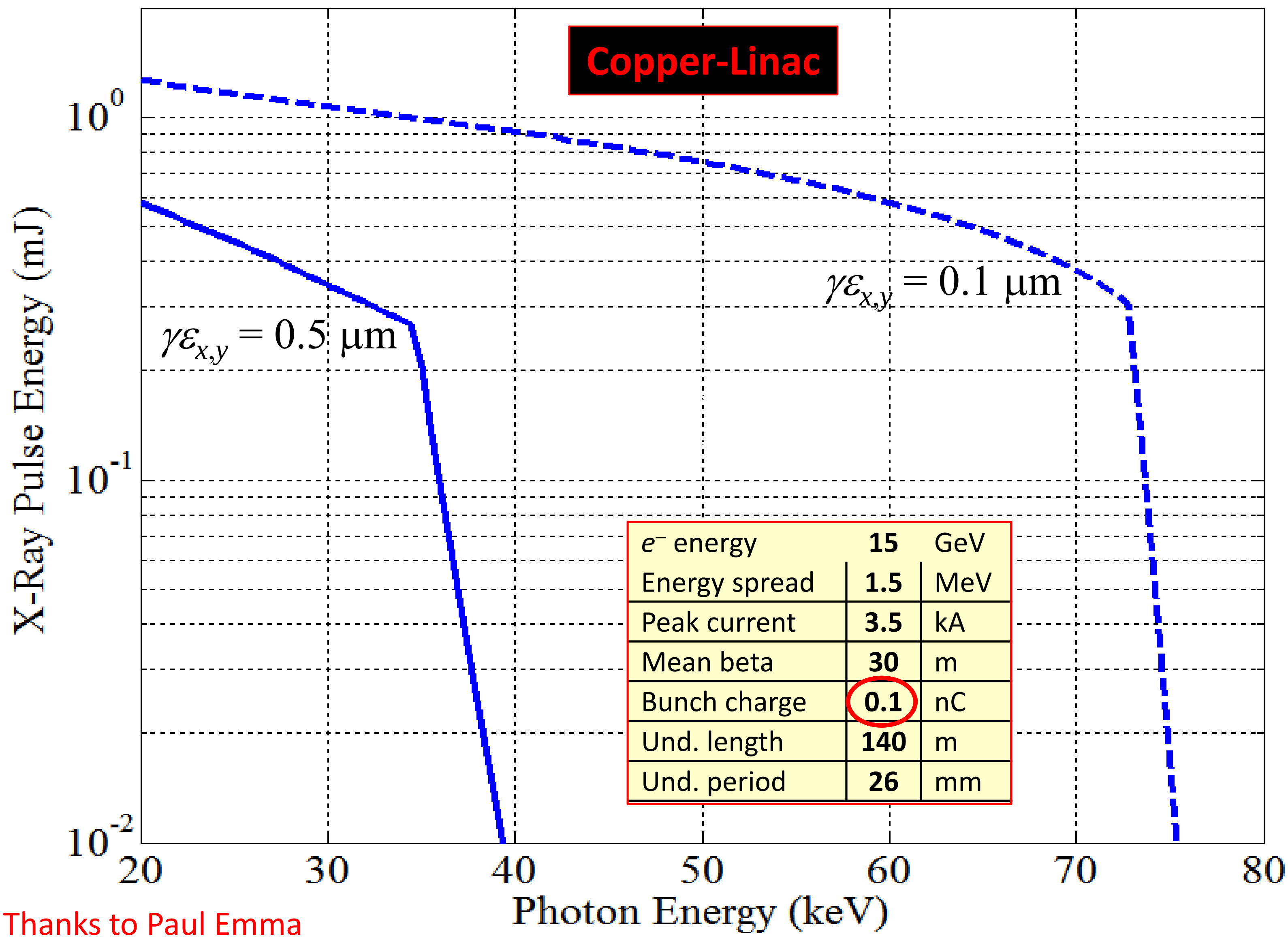
FEL sources

Going to moderate currents (still under 1 mA); emittance improvement is a big deal (ideally 0.1 μm)

Ultrafast Electron Diffraction/Microscopy

High brightness! Ideally a factor of 100 from current photoinjectors. Very low current. Short pulse duration (100 fs at sample, less for some applications)

Emittance Leverage at LCLS-II/HXR (15 GeV, 120 Hz)



e^- energy	**15**	GeV
Energy spread	**1.5**	MeV
Peak current	**3.5**	kA
Mean beta	**30**	m
Bunch charge	**0.1**	nC
Und. length	**140**	m
Und. period	**26**	mm

Thanks to Paul Emma

Where are we? Where do we want to be?

QE: Typically few % for semiconductors, ~10^{-5} for metals

- Room for improvement for both

Average current: >75 mA (meets current needs)

- Appl. Phys. Lett. **102**, 034105 (2013)

Average current density: ~20 mA/mm^2 (limiting)

- Phys. Rev. ST Accel. Beams **16**, 033401 (2013)

Peak current density limited by Child's Law/space charge

Lifetime: Days for high current; unlimited for lower current

- Ion Bombardment, Chemical Contamination, Thermal

Response time:

- Metals: ~fs, faster than current UED requirements, even from cathode
- PEA semiconductors: <1ps; May need to improve for UED
- NEA semiconductors: ps to 100ps, depending on wavelength

Emittance lower limit set by disorder induced heating (not there yet)

- New Journal of Physics 15,103024 (2013)

LCLS-II parameters met: APL 106, 094101 (2015)

https://science.energy.gov/~/media/bes/pdf/reports/2017/Future_Electron_Source_Worskhop_Report.pdf

Parameters, and how to affect them

Increasing the electron MFP will improve the QE. Phonon scattering cannot be removed, but a more perfect crystal can reduce defect and impurity scattering:

$$\frac{1}{\lambda_{MFP}} = \frac{1}{\lambda_{el-el}} + \frac{1}{\lambda_{ep}} + \frac{1}{\lambda_{ep,ems}} + \frac{1}{\lambda_{ep,abs}} + \frac{1}{\lambda_{impurity}} + \frac{1}{\lambda_{defect}} + \frac{1}{\lambda_{boundary}}$$

Choice of wavelength affects emittance and QE:

$$E_{excess} = h\nu - \phi_{threshold} \qquad MTE = \frac{E_{excess}}{3} \qquad \frac{\varepsilon_n}{\sigma_x} = \sqrt{\frac{MTE}{mc^2}} \qquad \text{BUT} \qquad QE \propto (h\nu - \phi)^2$$

Control of surface roughness is critical to minimizing the intrinsic emittance – epitaxial growth?

A question to consider: Why can CsI (another ionic crystal, PEA cathode) achieve QE>80%?

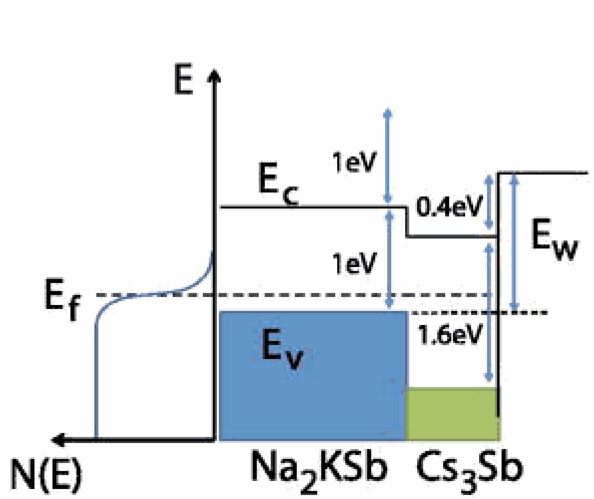
T.H. Di Stefano and W.E. Spicer, Phys. Rev. B **7**, 1554 (1973)

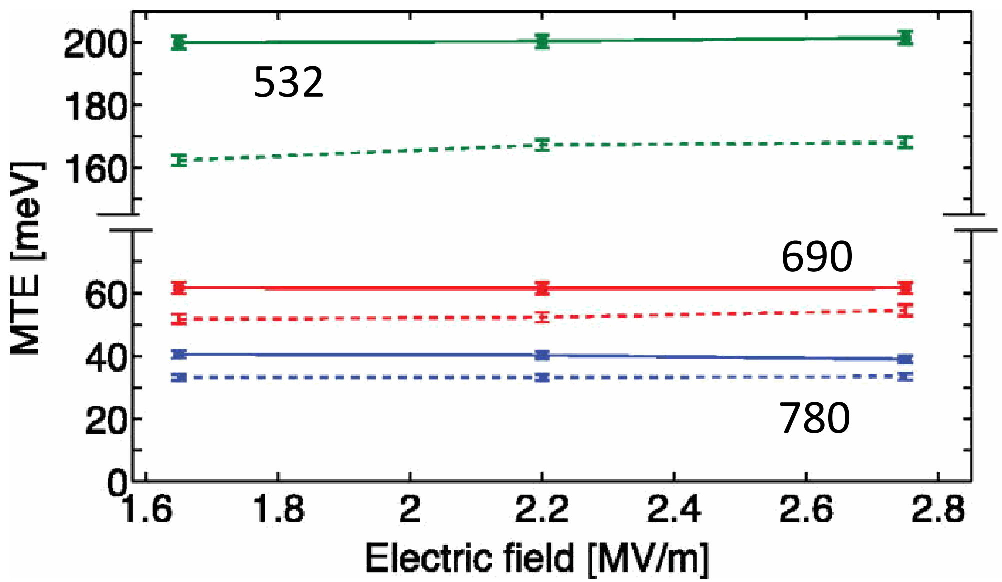
Large band gap and small electron affinity play a role, but, so does crystal quality.

Scattering

Phonon scattering can be helpful

- Well known in thermalization of electrons in GaAs and Diamond
- Luca & company recently demonstrated in PEA materials:
- 20% reduction in MTE for transmission mode operation





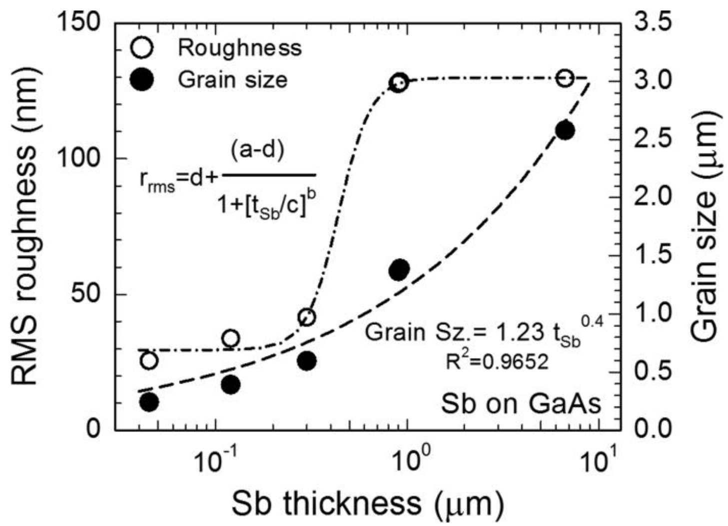
150 nm Na_2KSb (with few nm surface layer)

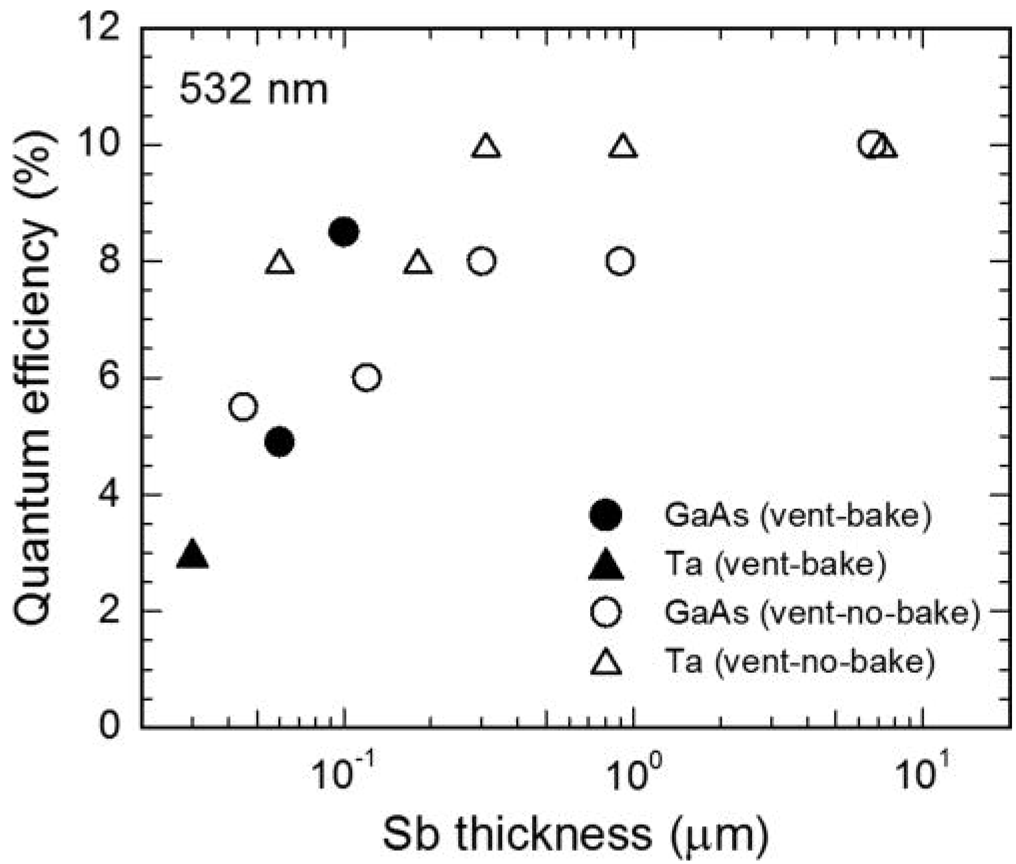
Appl. Phys. Lett. **108**, 124105 (2016)

Grain Boundaries

Grain boundary scattering: just bad

- Large grains or Epitaxy
- One solution: REALY thick cathodes (50 optical absorption lengths)
- Another: Stay tuned





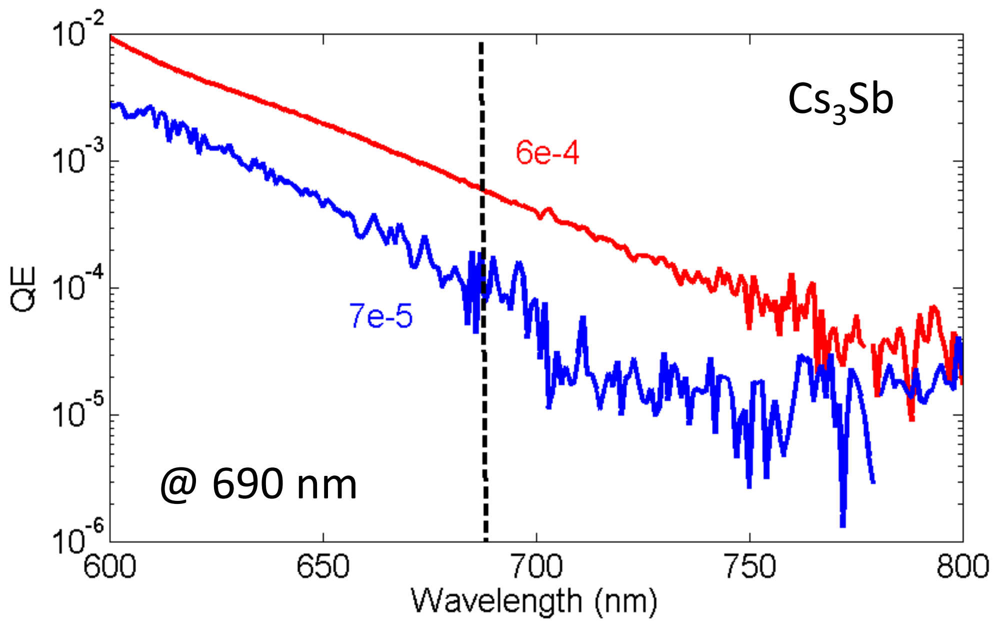
J. Vac. Sci. Technol. A **34**, 021509 (2016)

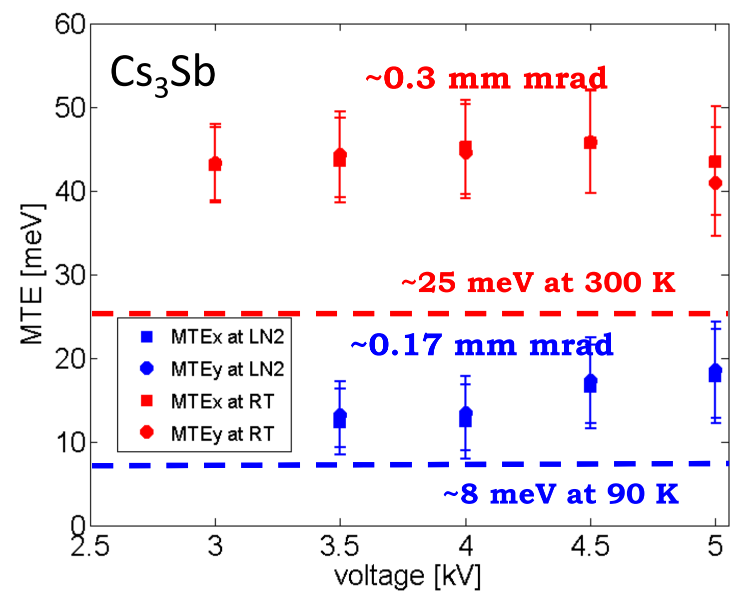
Lattice Temperature

For E_{excess}<0, MTE=kT, and lattice temperature becomes important

Photoemission in this domain relies on defect states

- May depend on crystal quality
- Material conductivity will as well



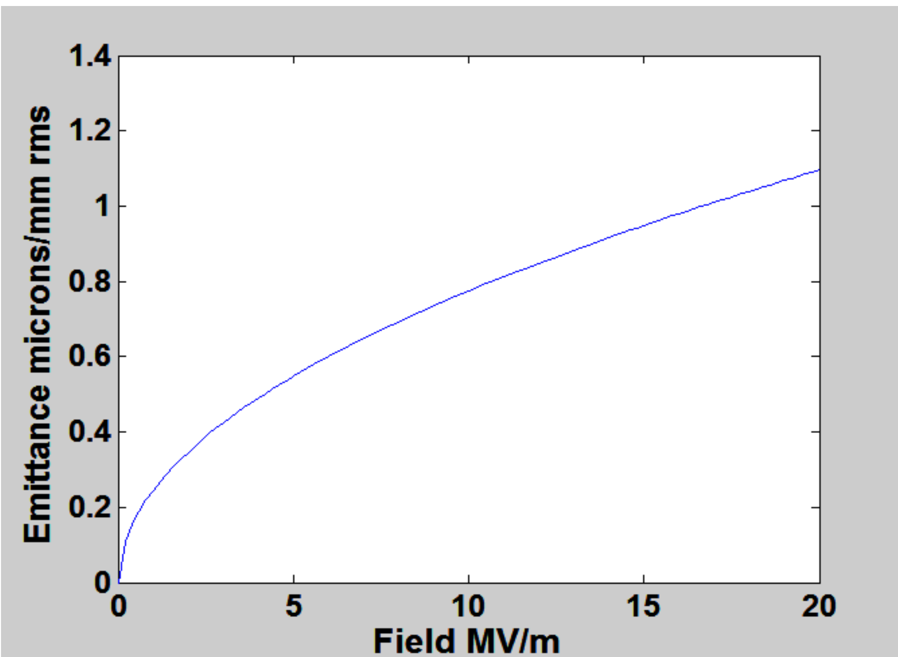


Phys. Rev. ST Accel. Beams **18**, 113401

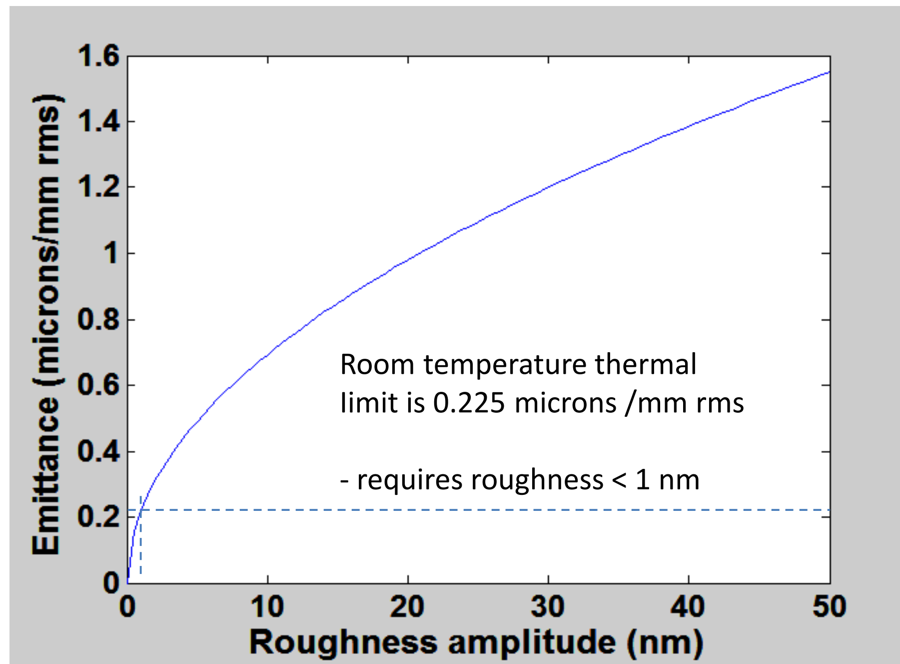
Roughness and Emittance

$$\varepsilon_{rough} = \sigma_{x,y}\sqrt{\frac{\pi^2 a^2}{2m_0 c^2 \lambda} Ee}$$

D. Xiang et al. Proceedings of PAC07, Albuquerque, New Mexico, USA



Field dependent emittance growth: 20 nm amplitude, 80 nm period



Emittance growth at 20 MV/m, period 4 x amplitude.

Calculations by H. Padmore

The Old Standby - Metals

Normal conducting RF photoinjectors often use metal cathodes, either Cu (simplicity) or Mg (higher QE)

The good points:

- Basically unlimited lifetime (with occasional laser or ion cleaning)
- Tolerant of poor (nTorr) vacuum
- Prompt response time (fs)
- Low field emission

However

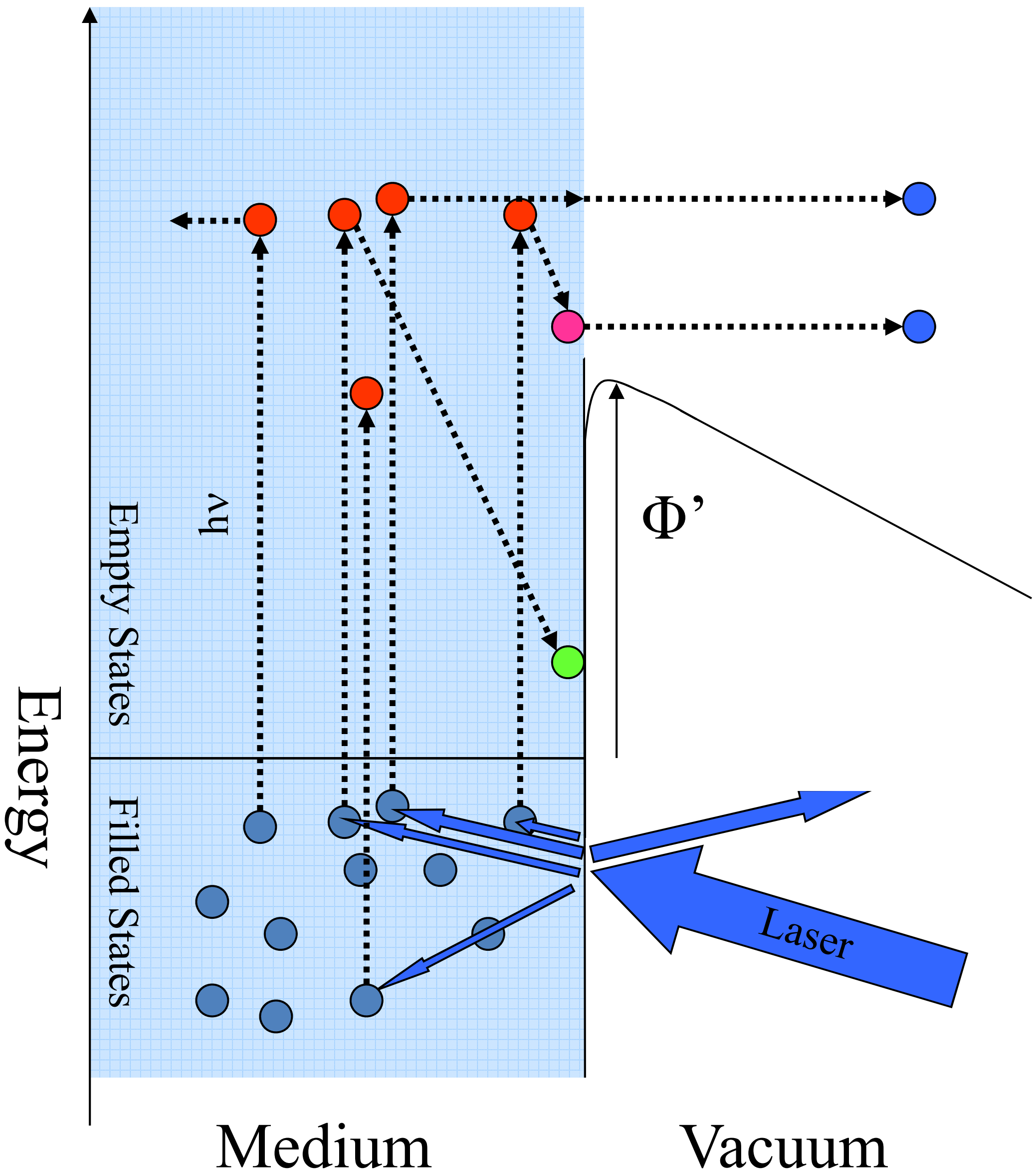
- Require UV laser
- Typical QE of 10^{-5} to 10^{-3}

} Not suitable for >1 mA injectors

Magnesium QE @ 266 nm = 0.2% @ 266nm

W.F. Krolikowski and W.E. Spicer, Phys. Rev. 185, 882 (1969)
D. H. Dowell *et al.*, Phys. Rev. ST Accel. Beams 9, 063502 (2006)
T. Srinivasan-Rao *et al.*, PAC97, 2790

Three Step Model of Photoemission - Metals

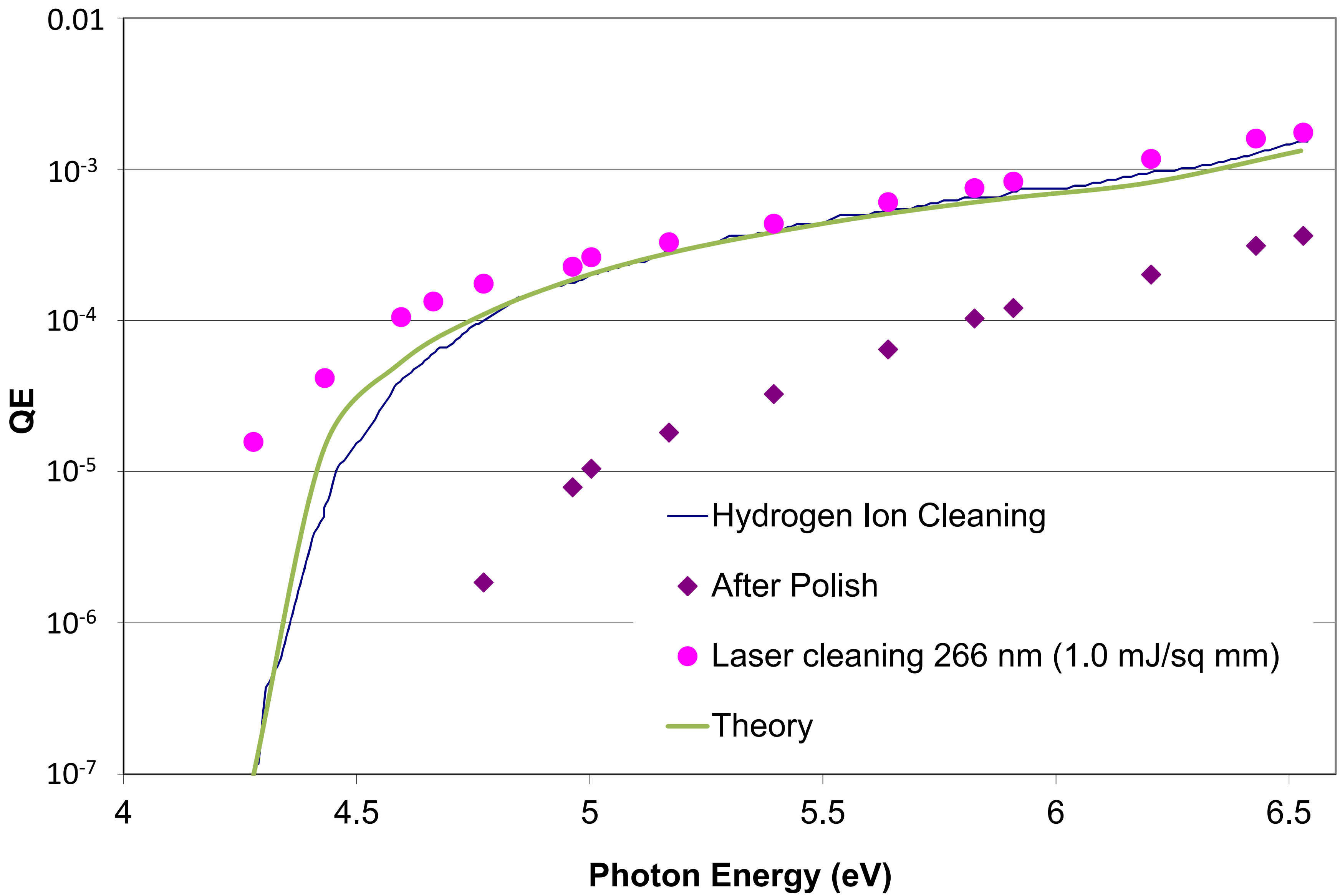


1) Excitation of e^- in metal
   - Reflection
   - Absorption of light
   - Energy distribution of excited e^-
2) Transit to the Surface
   - e^--e^- scattering
     - mfp ~50 angstroms
   - Direction of travel
3) Escape surface
   - Overcome Workfunction
   - Reduction of Φ due to applied field (Schottky Effect)

Integrate product of probabilities over all electron energies capable of escape to obtain Quantum Efficiency

M. Cardona and L. Ley: Photoemission in Solids 1, (Springer-Verlag, 1978)

LCLS Copper Cathodes



D. H. Dowell *et al.*, Phys. Rev. ST Accel. Beams 9, 063502 (2006)

Superconducting Photocathodes

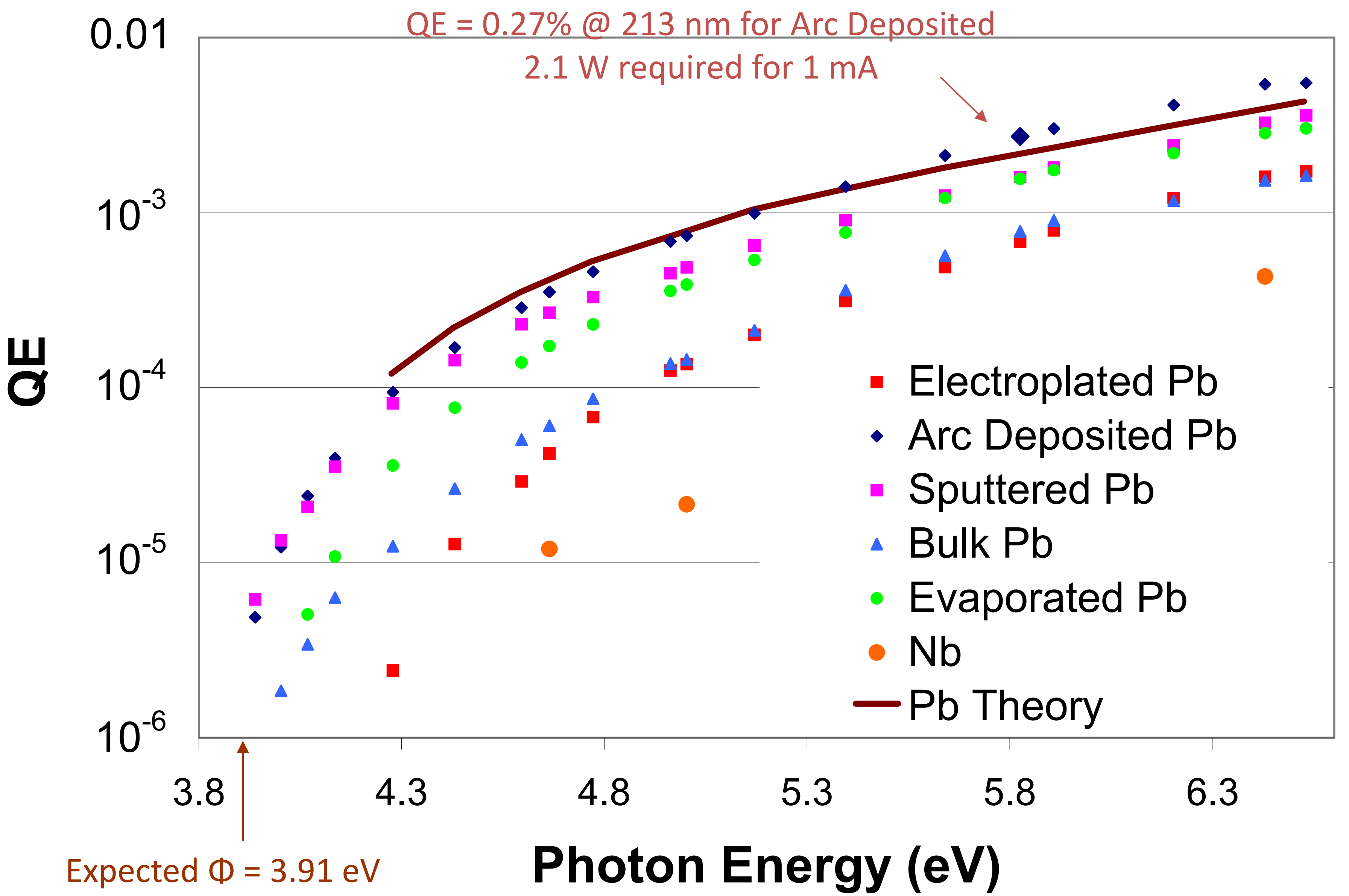
- The cathode-cavity interface is the most difficult part a superconducting injector
- Using a superconductor as a cathode removes the need for a RF choke, and may allow higher gradients
- Niobium is a poor photocathode -> use Lead
- Two ½ cell cavities (1.3 & 1.42 GHz) have been tested
  - Both reached 40 MV/m; RF performance unaffected by lead
  - Lead cathode QE comparable to room temperature values
  - Peak laser power of 3 MW/cm^2 (@ 248 nm) did not quench the cavity

J. Smedley, T. Rao, and Q. Zhao, J. Applied Physics 98, 043111 (2005)

J. Smedley, T. Rao , J. Sekutowicz, Phys. Rev. ST Accel. Beams 11, 013502 (2008)

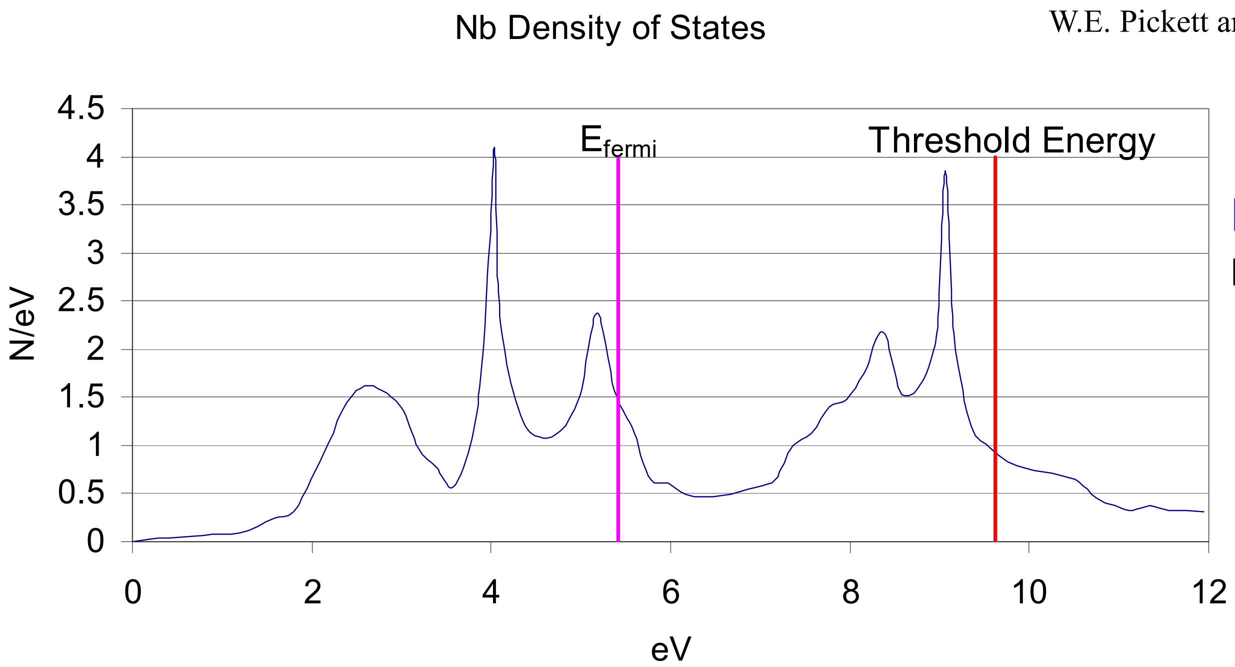
J. Smedley *et al.*, PAC07, 1365; J. Sekutowicz *et al.*, PAC07, 962

DC Room Temperature Photoemission Results



J. Smedley, T. Rao , J. Sekutowicz Phys. Rev. ST Accel. Beams **11**, 013502 (2008)

W.E. Pickett and P.B. Allen; Phy. Letters **48A**, 91 (1974)

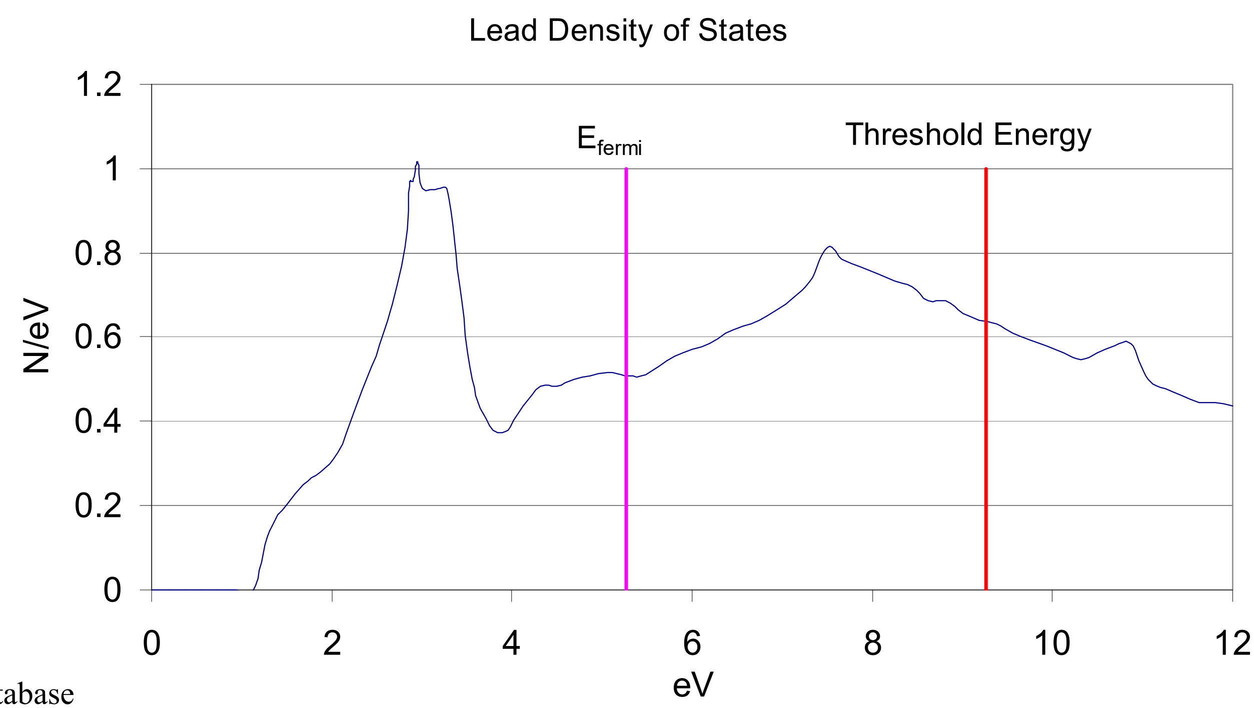


Density of States for Nb

Large number of empty conduction band states promotes unproductive absorption

Density of States for Lead

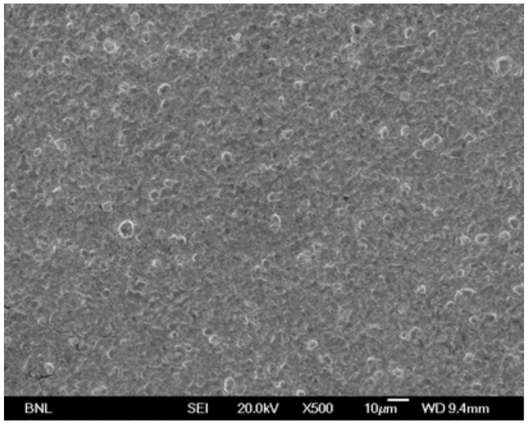
Narrow p-band limits unproductive absorption at higher photon energies



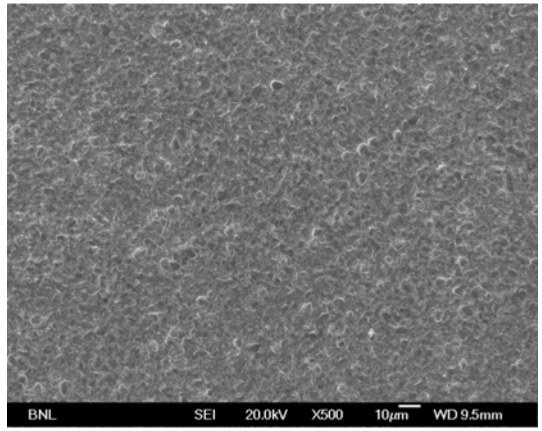
NRL Electronic Structures Database

Lead Surface Finish and Damage Threshold

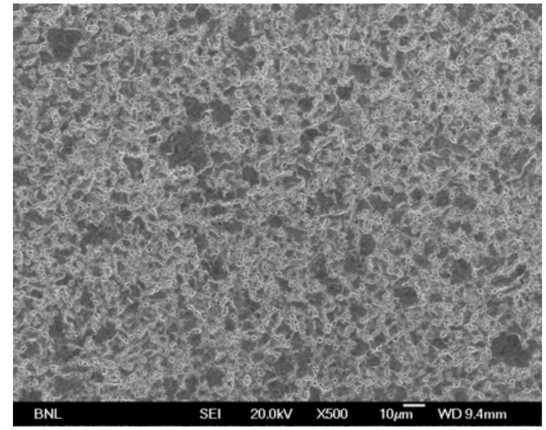
Electroplated Lead



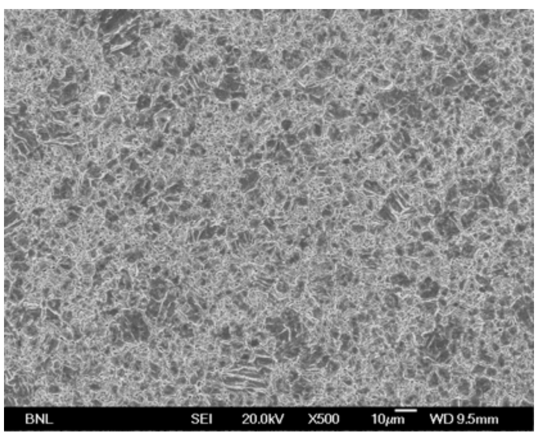
Prior to Laser Cleaning



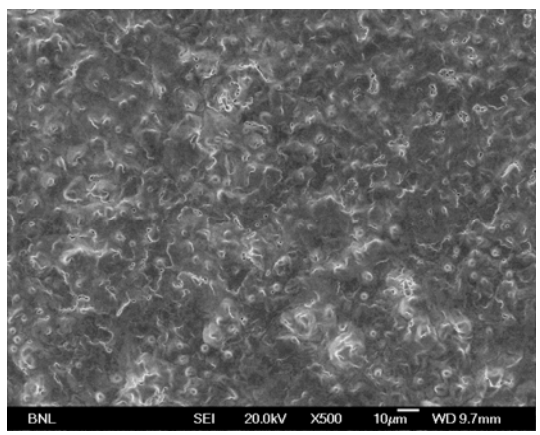
0.11 mJ/mm^2



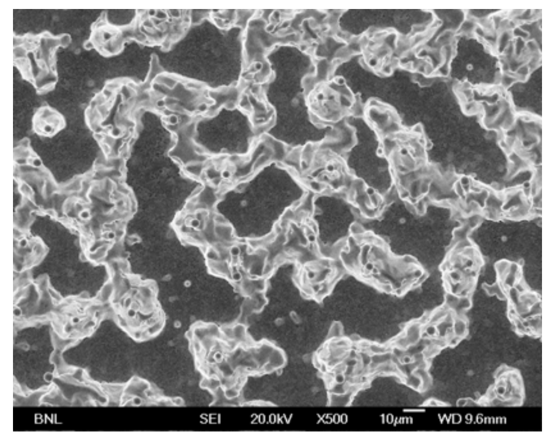
0.26 mJ/mm^2



0.52 mJ/mm^2



1.1 mJ/mm^2

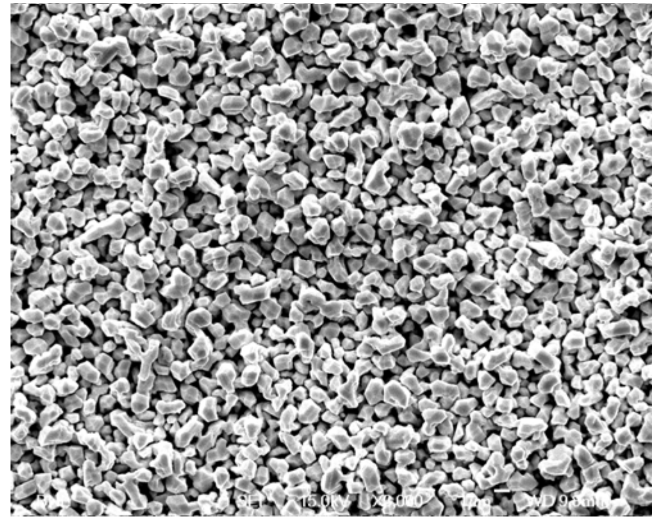


1.8 mJ/mm^2

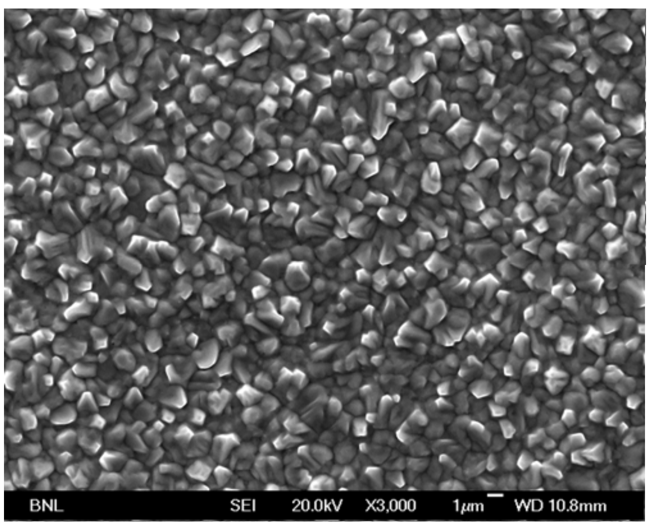
Surface Uniformity

Arc Deposited

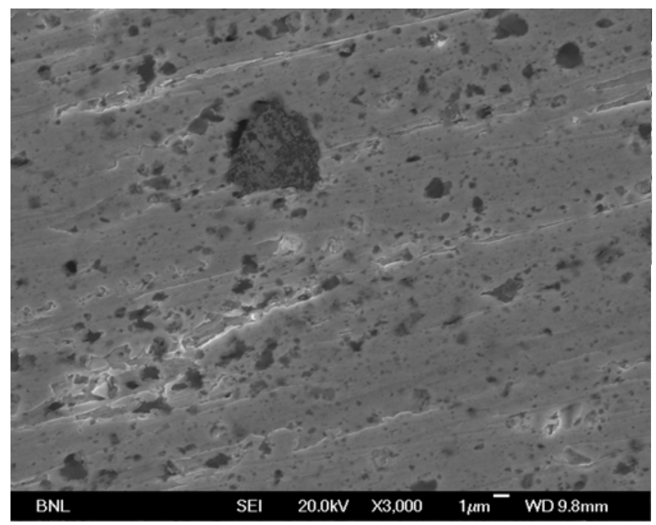
Sputtered



Vacuum Deposited



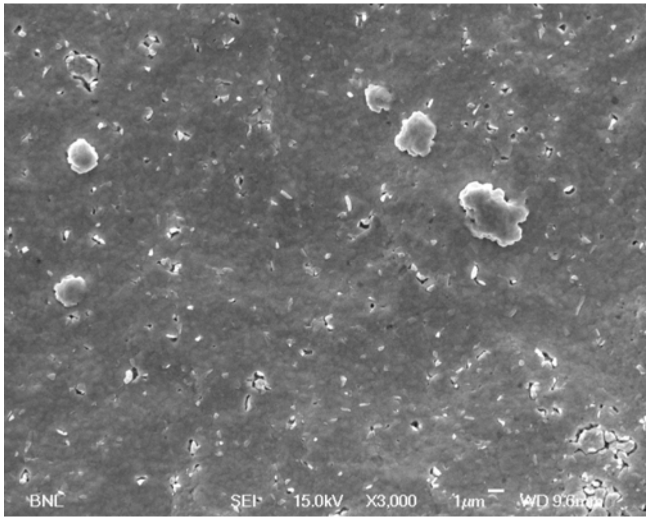
Solid



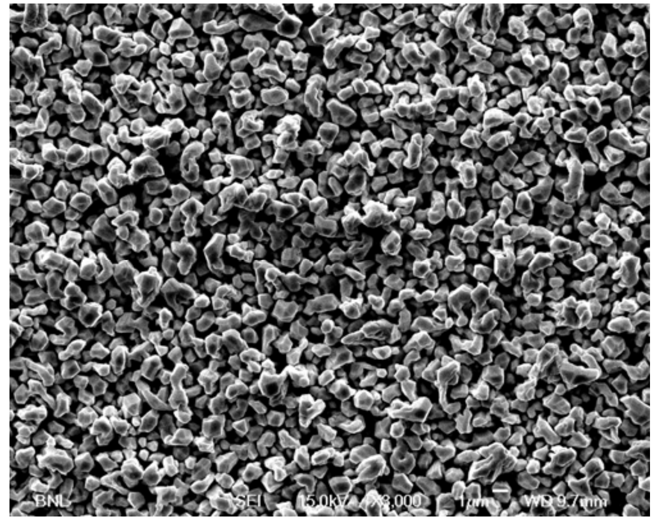
10 µm

Surface Uniformity

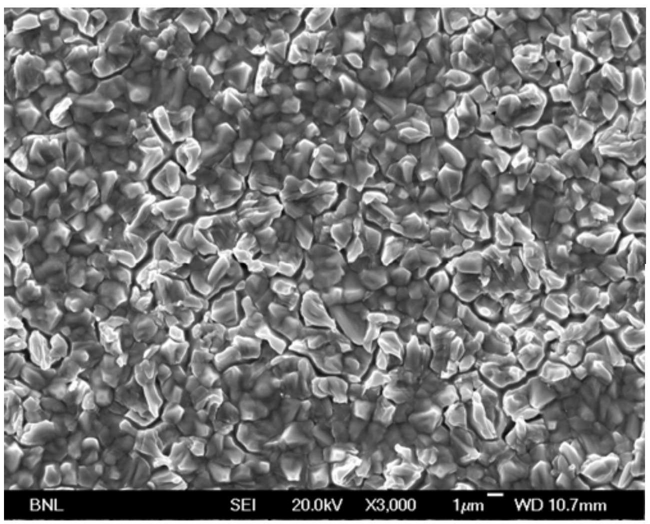
Arc Deposited



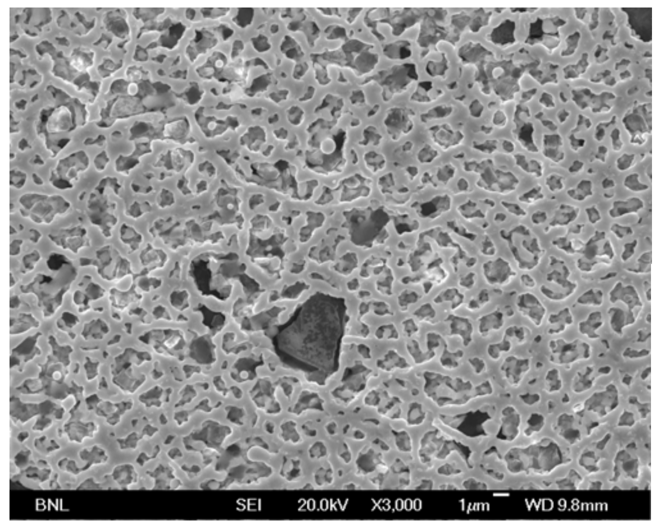
Sputtered



Vacuum Deposited



Solid



10 µm

All cathodes laser cleaned with 0.2 mJ/mm^2 of 248nm light

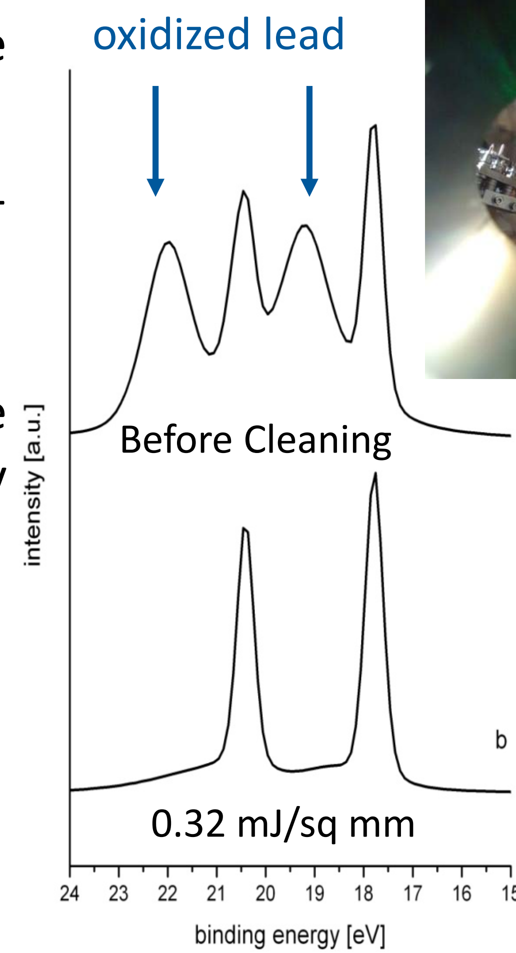
How does laser cleaning work?

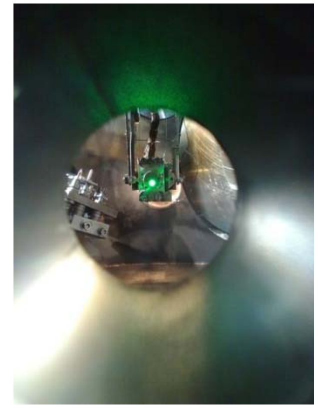
Initial lead QE is generally two orders of magnitude below theoritical predictions

Exposure to 0.2 mJ/sq mm of 248 nm light for ~1 min at 300 Hz is sufficient to achieve expected QE

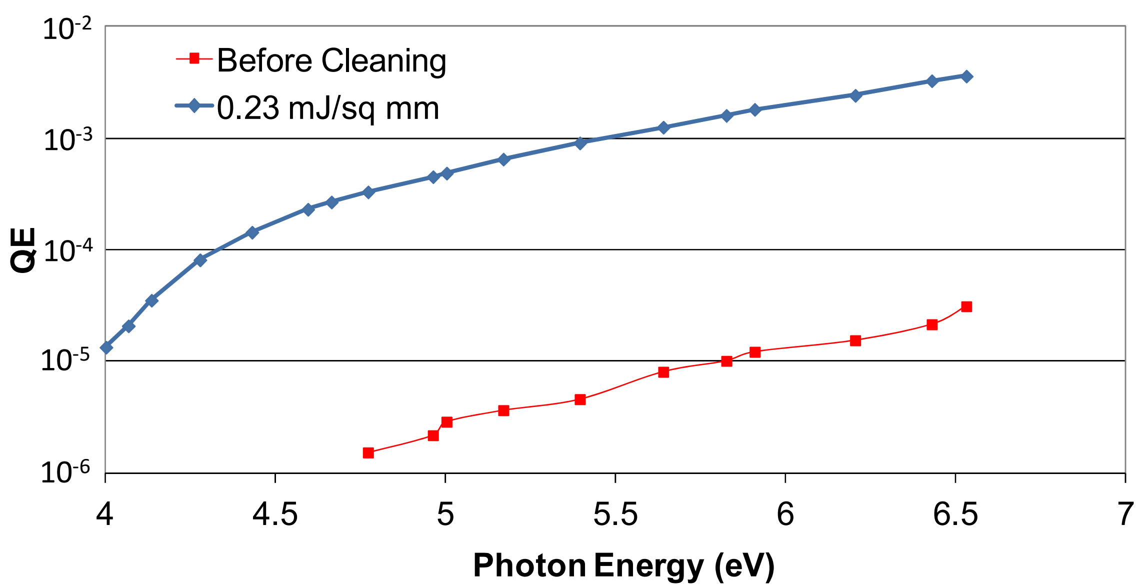
CW UV light leads to a small improvement in QE

XPS shows that Pb Oxide beaks begin to decrease at 0.1 mJ/sq mm, and have totally dissapeared by 0.3 mJ/ sq mm





Lead XPS (arc depositied)



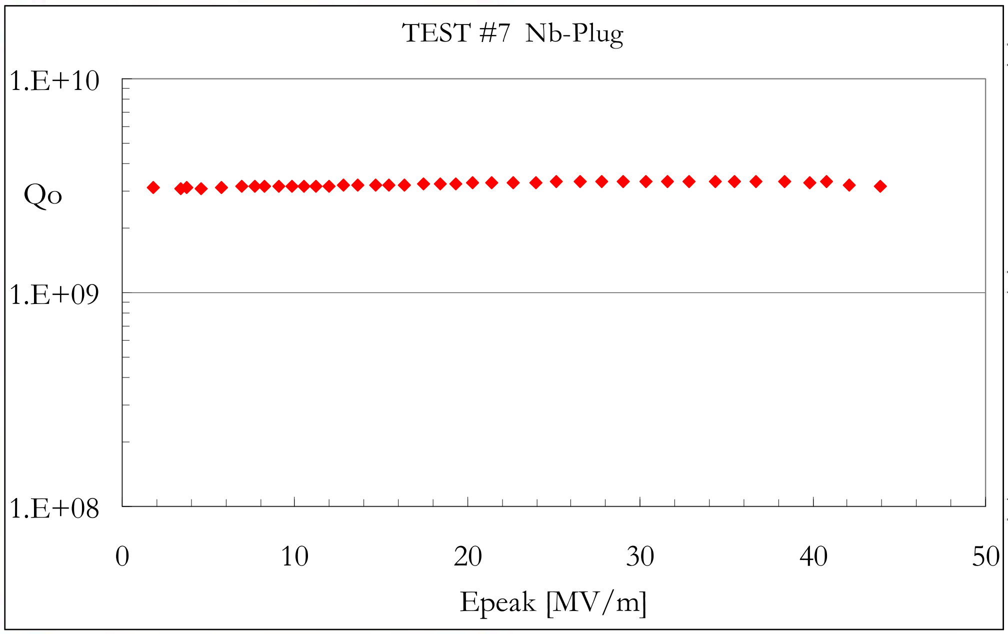
Spectral response (sputtered)

Hybrid Cavity Options

Plug Gun (Jlab)

1.42 GHz niobium cavity w/ removable plug

Q_0=3x10^9 w/ Nb Plug

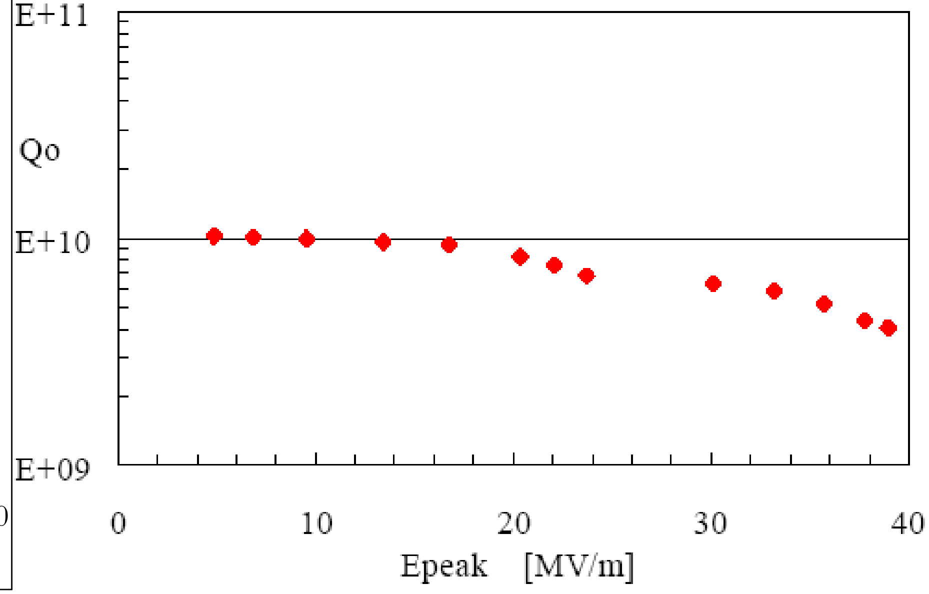


P. Kneisel, et al., PAC 05

DESY Gun

1.3 GHz niobium cavity

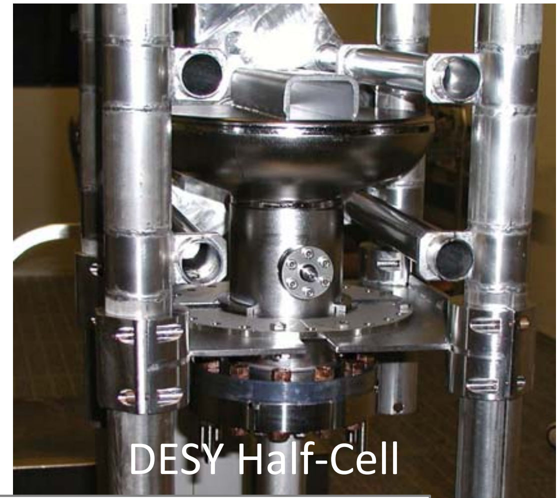
Q_0=1x10^{10} w/o Lead Plating

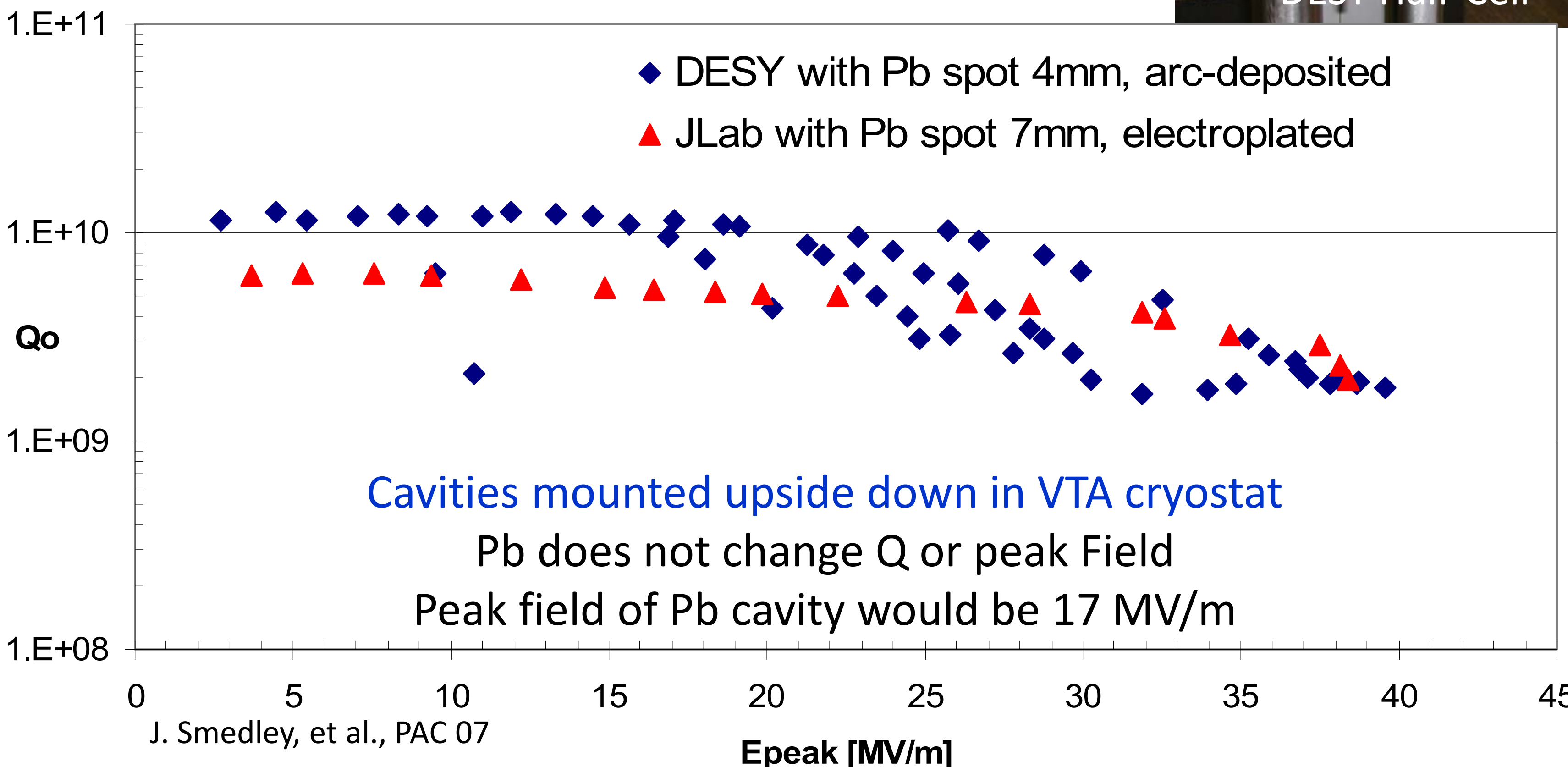


Cavity Tests

Nb-Pb Half-cells

Status March 07



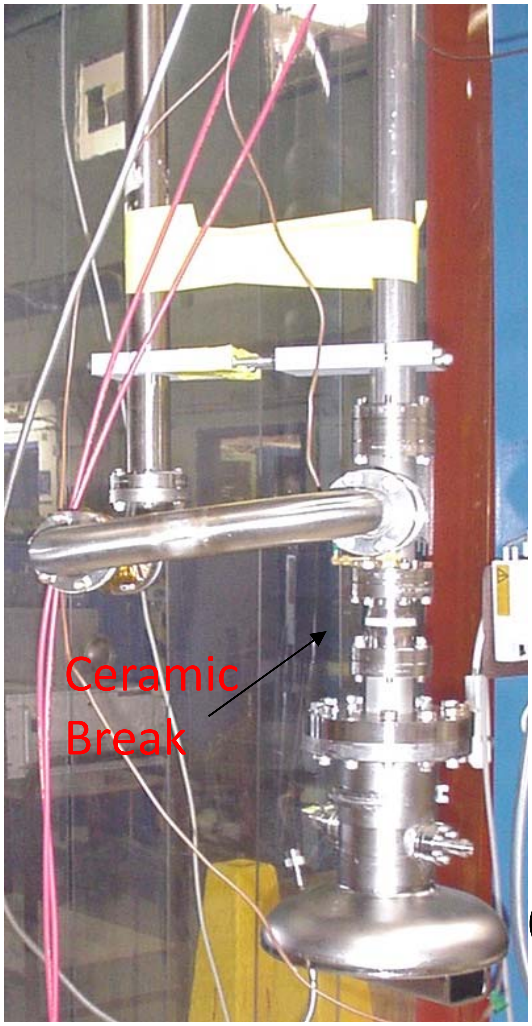


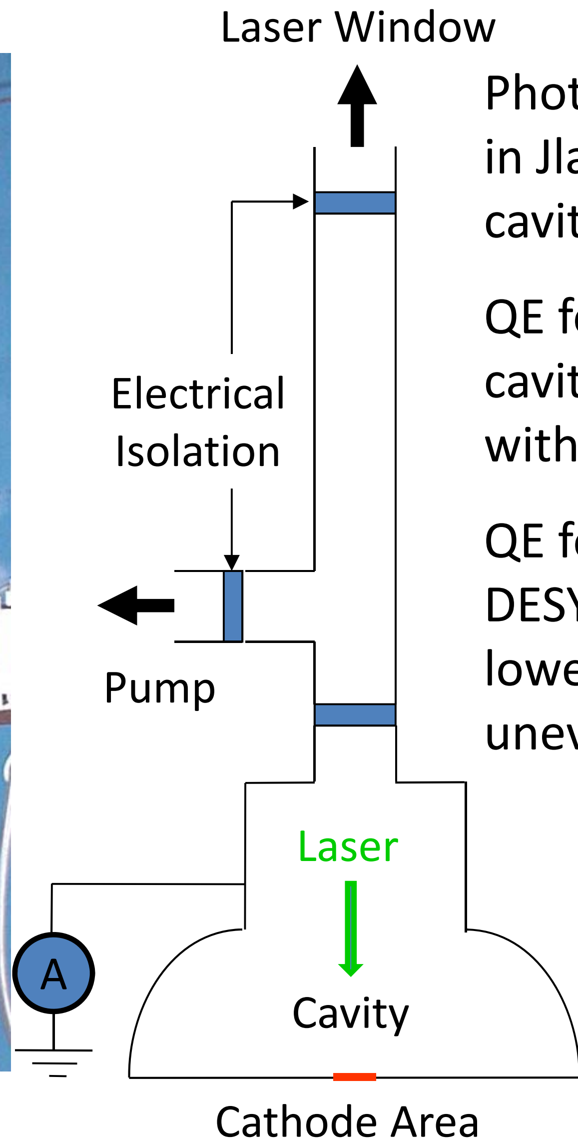
J. Smedley, et al., PAC 07

J. Smedley, et al., ERL 07

J. Sekutowicz, et al., PAC 07

Charge Measurement



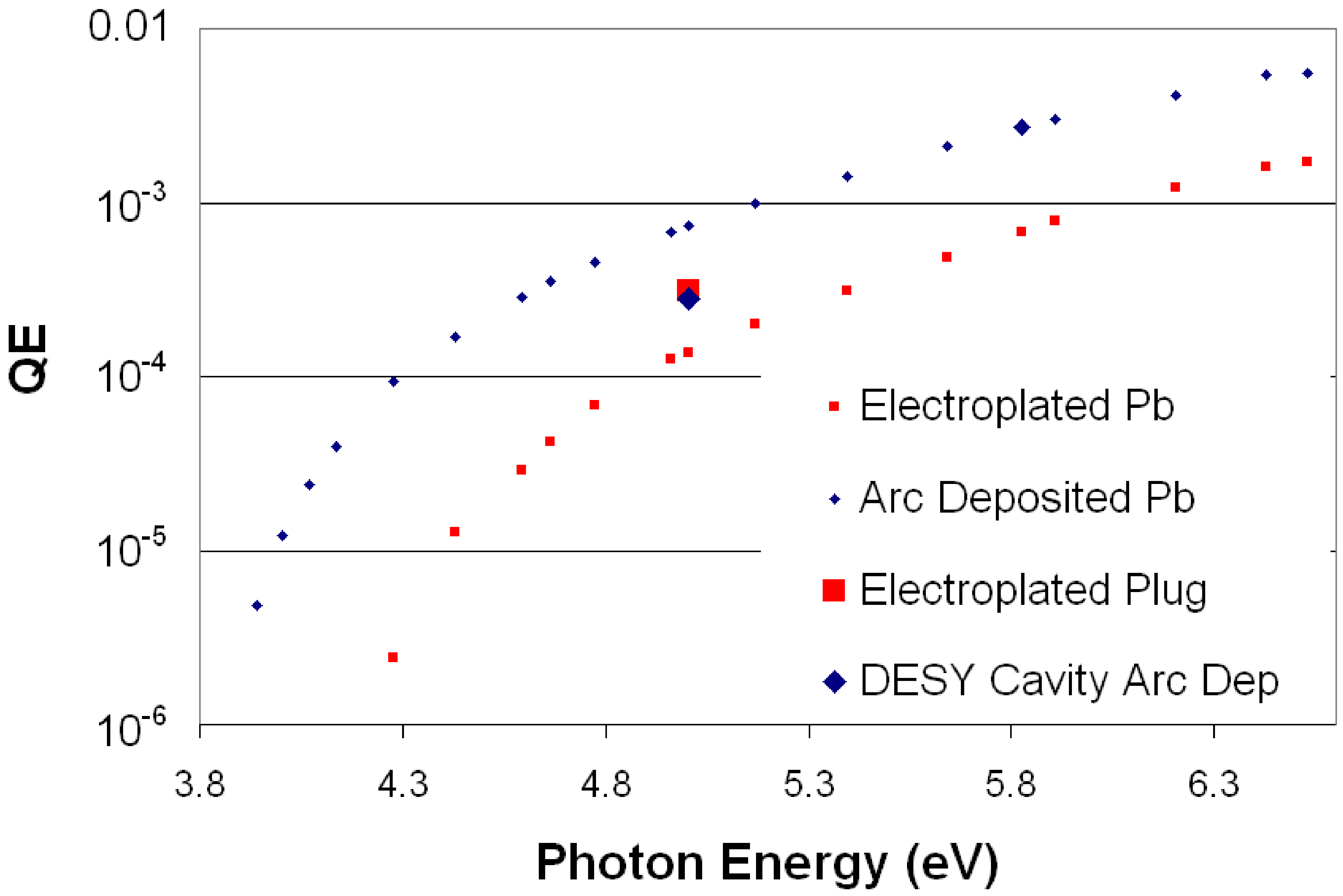


Photocurrent for both cavities measured in Jlab Vertical Test Area, by isolating the cavity and monitoring the current leaving

QE for electroplated lead plug in Jlab cavity was 1.6×10^{-4} (@248nm), in line with expected performance

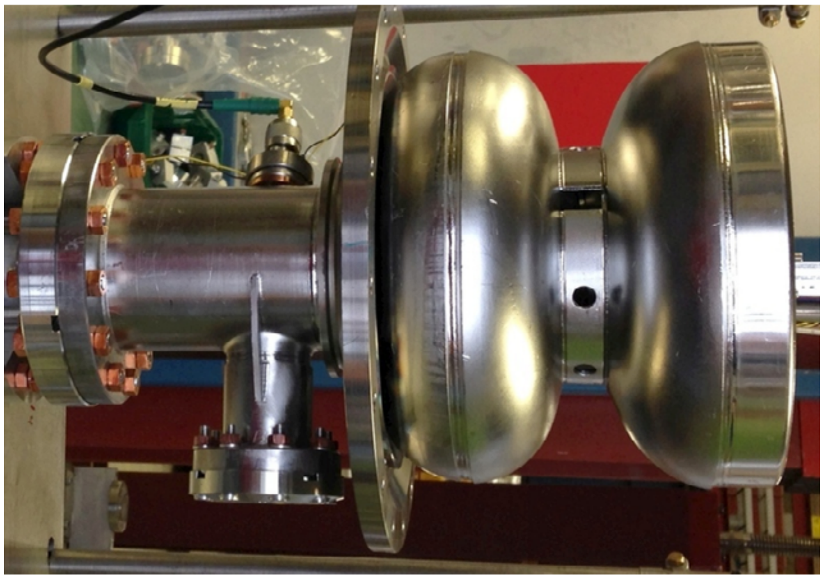
QE for Arc deposited lead cathode in DESY cavity was 1.4×10^{-4} (@248nm), lower than expected, possibly due to uneven lead coating

Comparison to Room Temp DC



New 1.5 cell Plug Gun

The 1.5-cell gun cavity prototype was built at TJNAF. The present plug version has very effective cooling of the cathode.

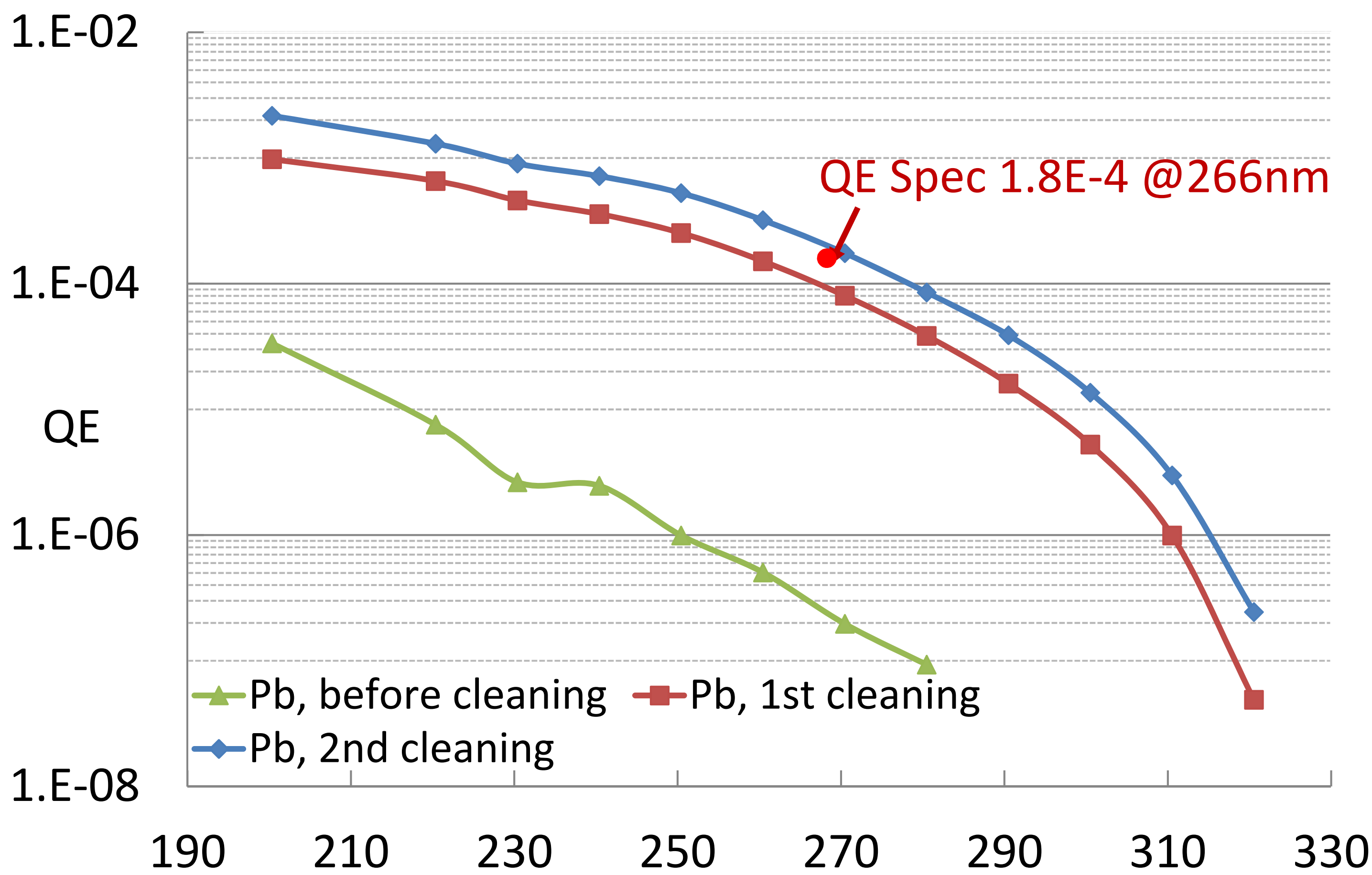


1.5-cell , 1.3 GHz gun cavity

Plug with LHe channels

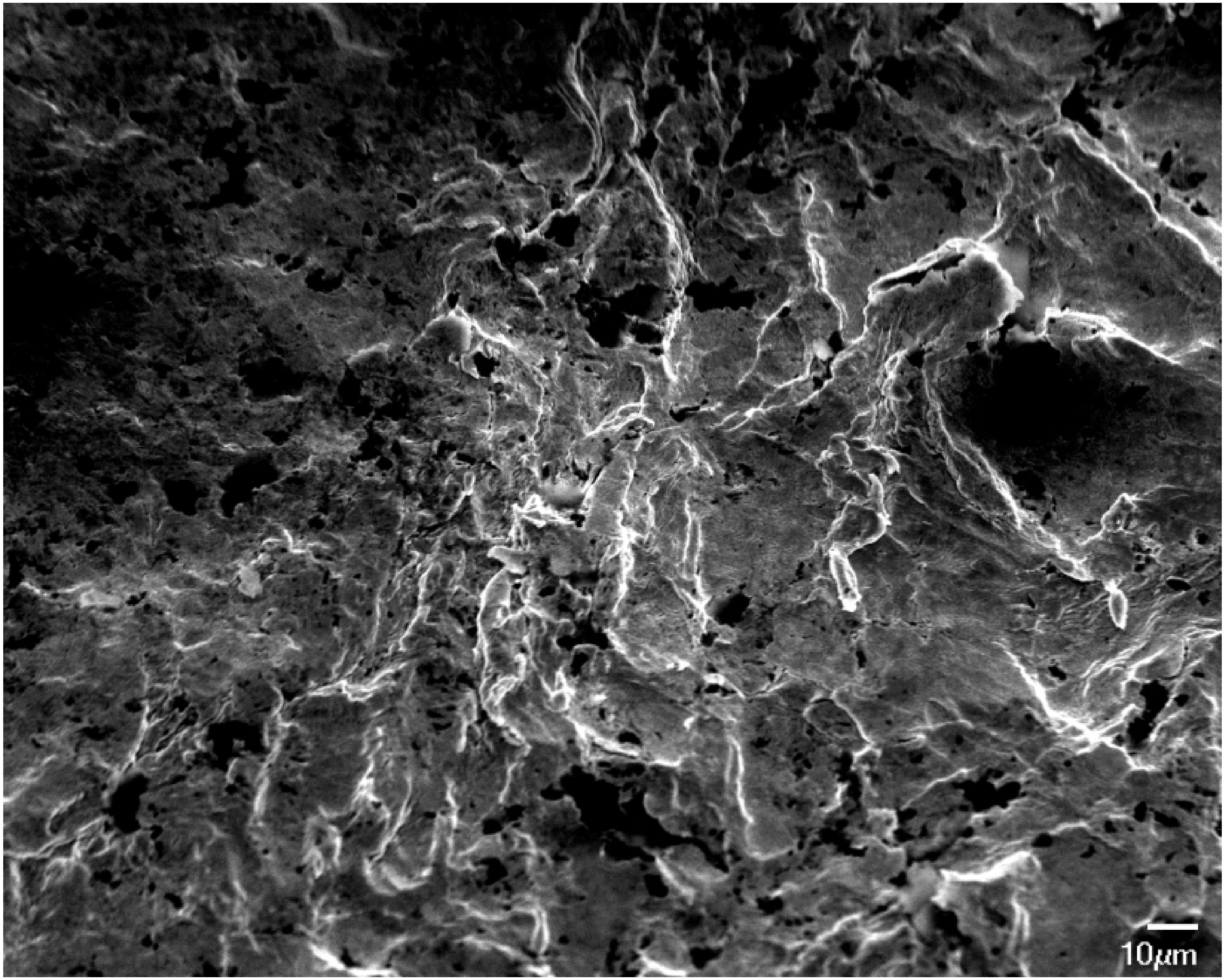
Nb/Pb cathode

QE test at BNL of the Pb coating on Plug

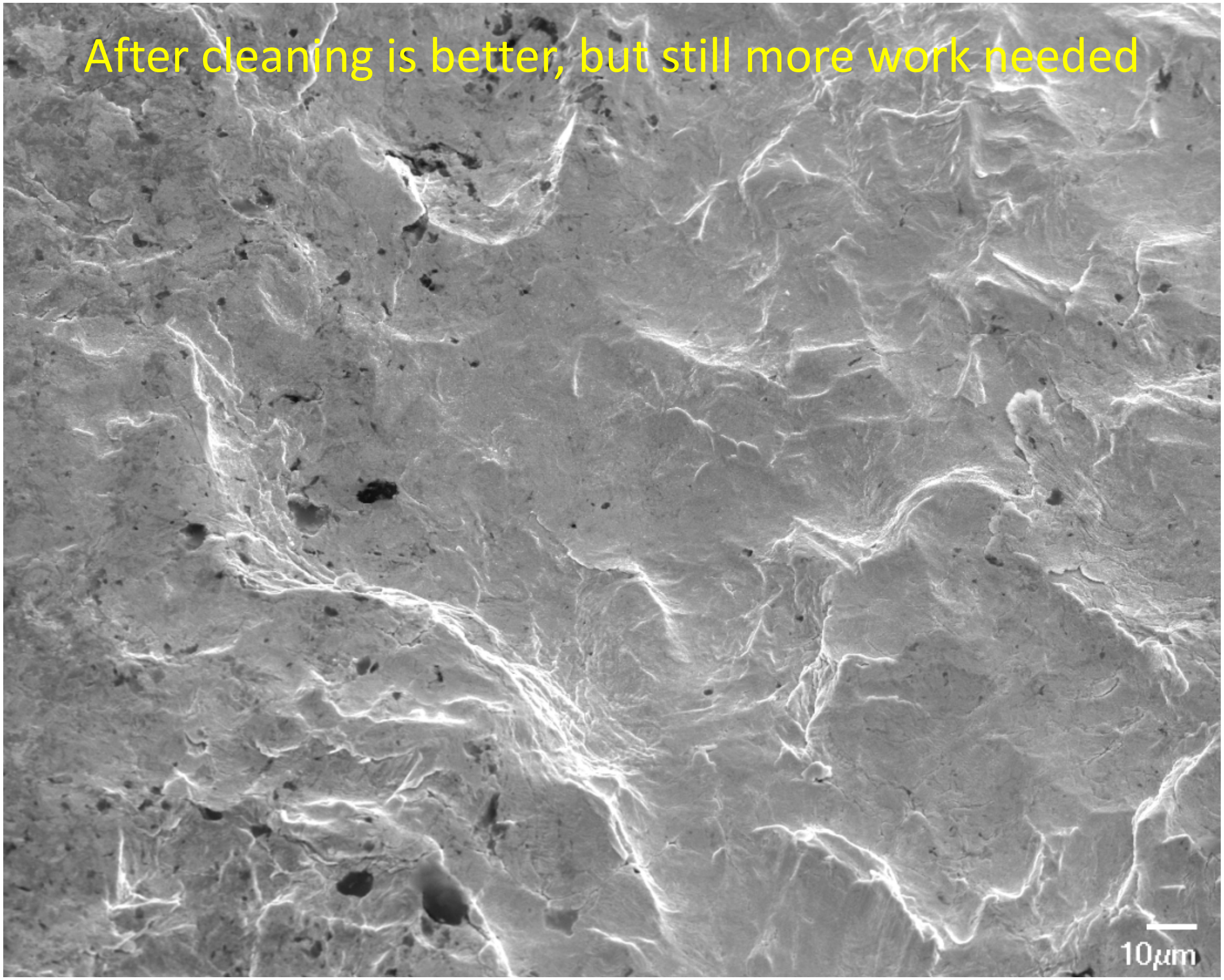


Laser cleaning: 1^{st} 1000 shots with 0.06 mJ/mm^2,
2^{nd} 10000 shots with 0.06 mJ/mm^2, all at 248nm

Uniformity and Roughness of Lead on Plug



Uniformity and Roughness of Lead on Plug



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:

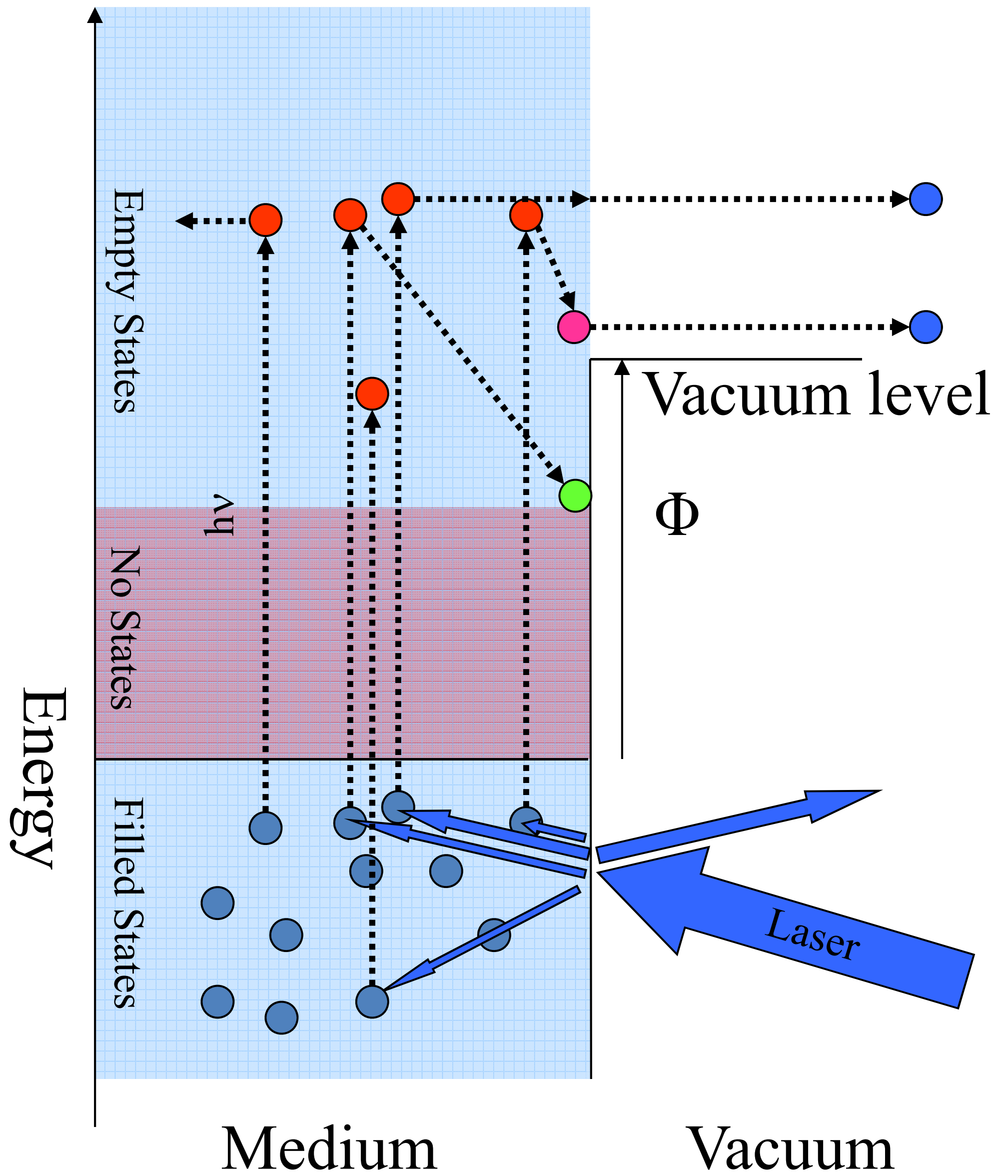
Cs_2Te – QE ~7% @ 262 nm, Lifetime 1000's of hrs

K_2CsSb – QE >4% @ 532 nm, Lifetime <10 hrs (Dowell)

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



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

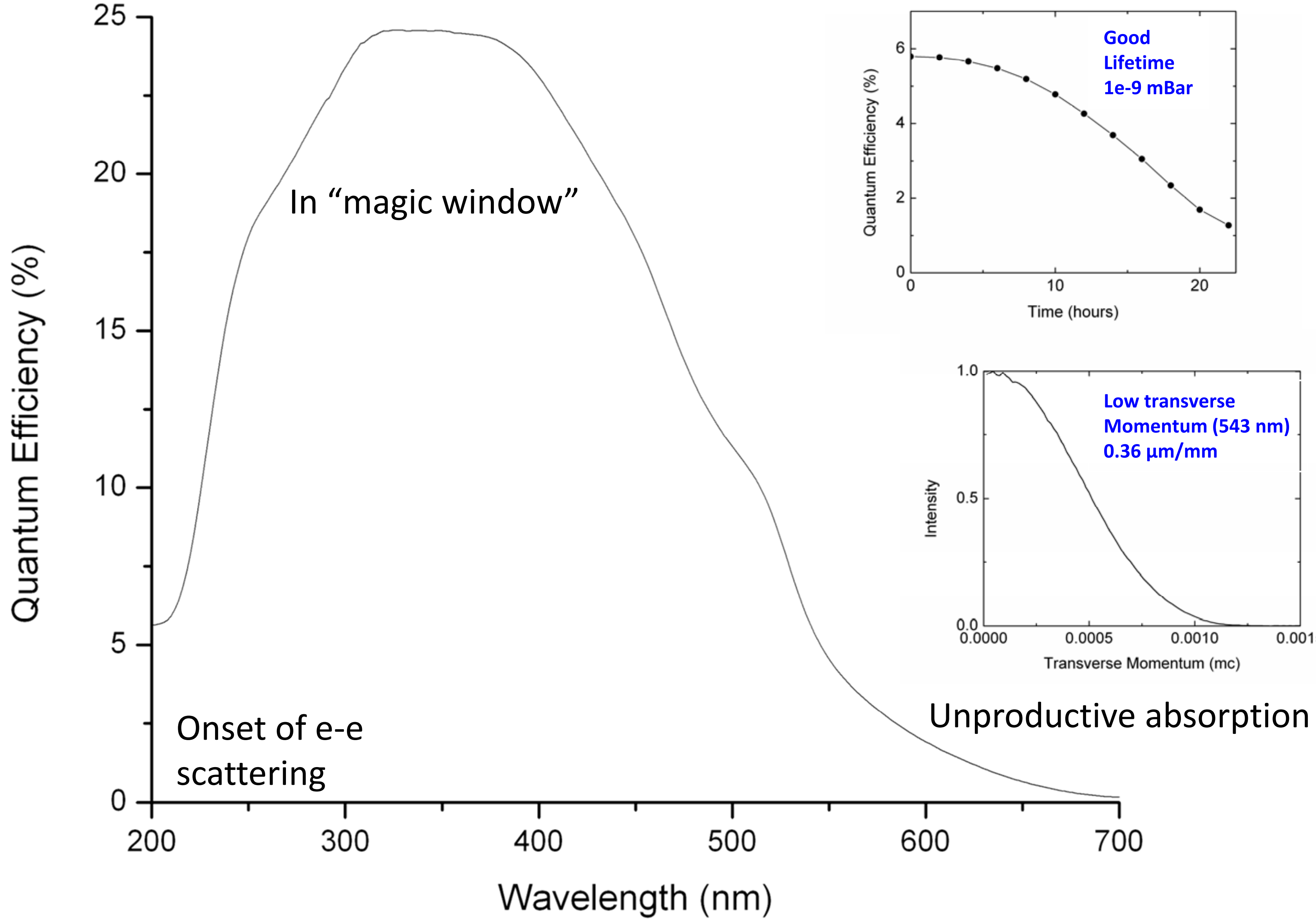
3) Escape surface

 Overcome Workfunction

 Multiple tries

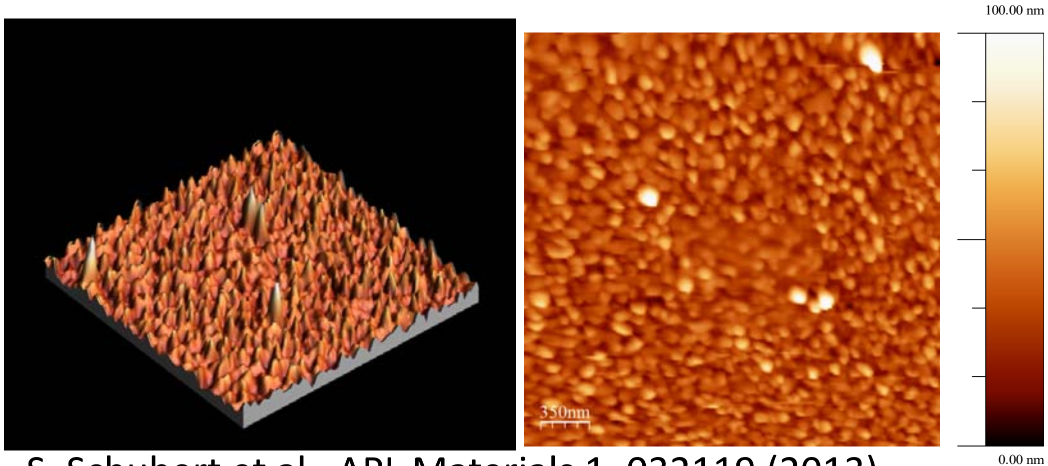
Need to account for Random Walk in cathode suggests Monte Carlo modeling

K_2CsSb: A Good Candidate



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

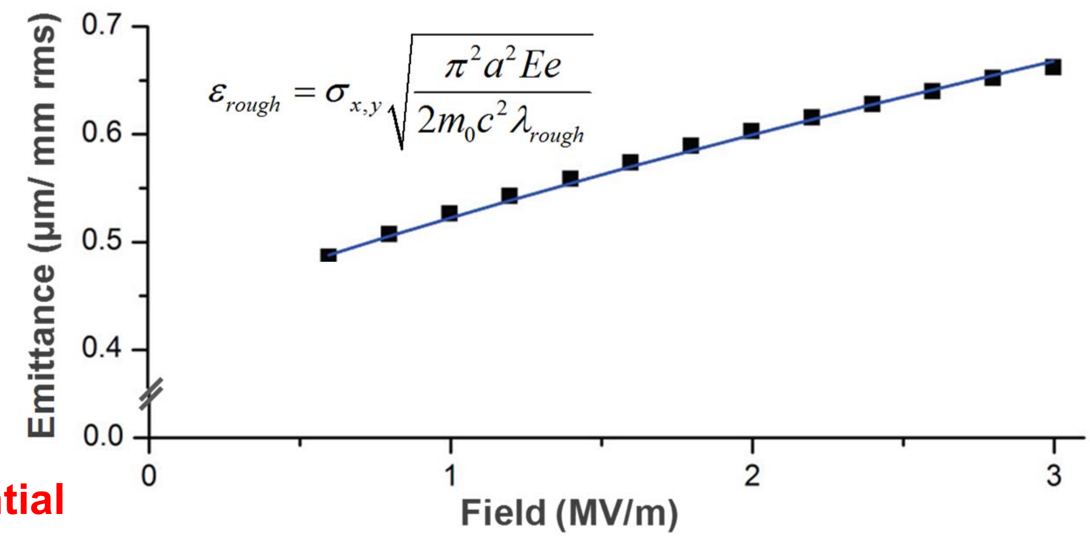
Roughness and Emittance



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

25 nm roughness, 100 nm spatial period

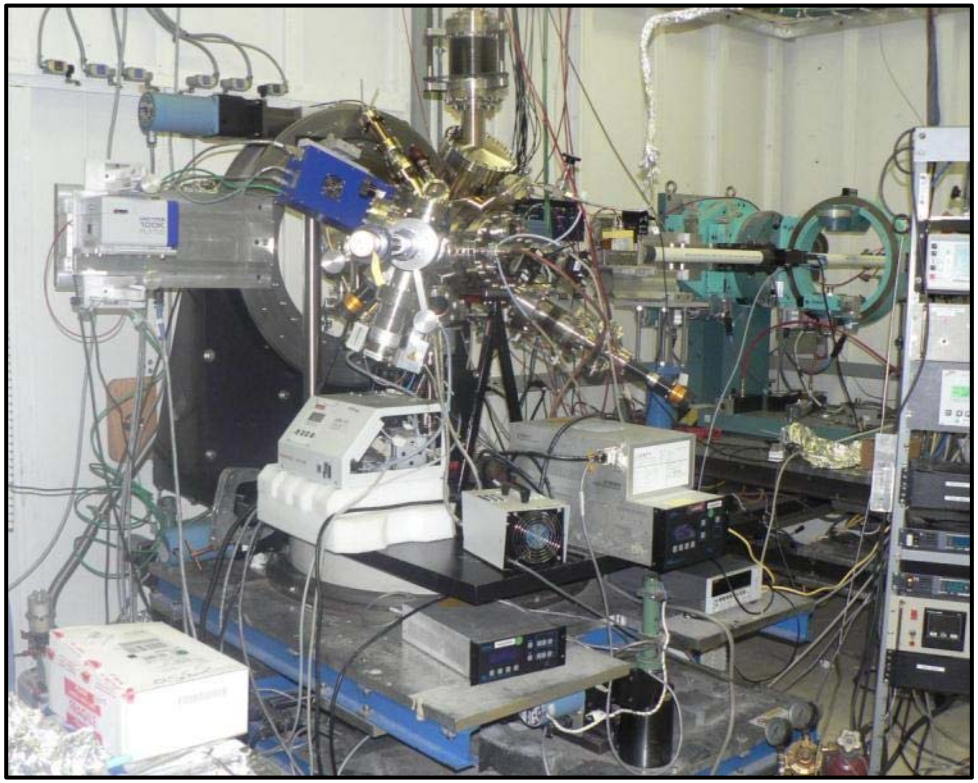
Emittance vs field measured with Momentatron, 532 nm light



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

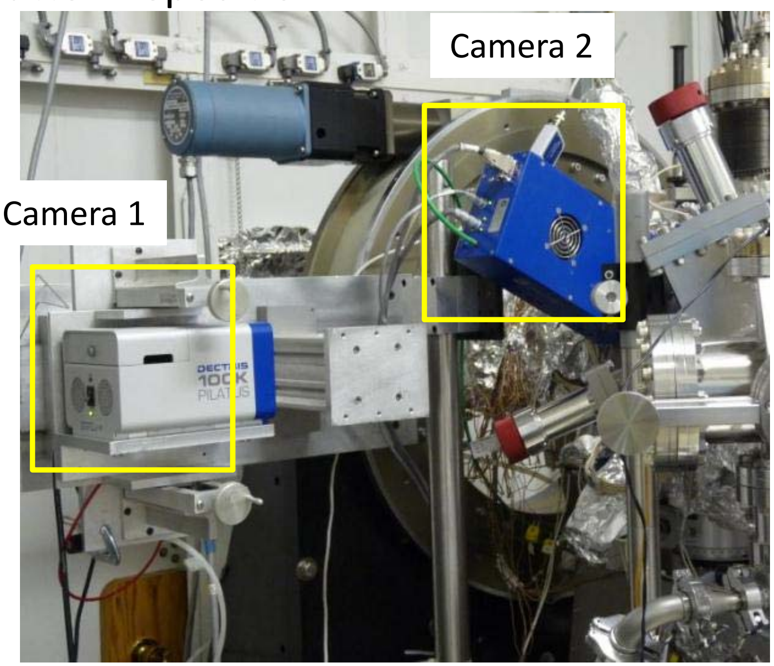
We now understand why sequential growth causes roughness, and can achieve near-atomic roughness with Alkali antimonides!

In operando analysis during growth
(setup at NSLS/X21 & CHESS G3 – ISR soon)



- UHV system (0.2 nTorr base pressure)
- Residual Gas Analyzer (RGA)
- Heating/cooling substrate/cathode
- Load lock
    - fast exchange of substrates
    - gun transfer
- Horizontal deposition of Sb, K and Cs.
- Sputter Deposition!

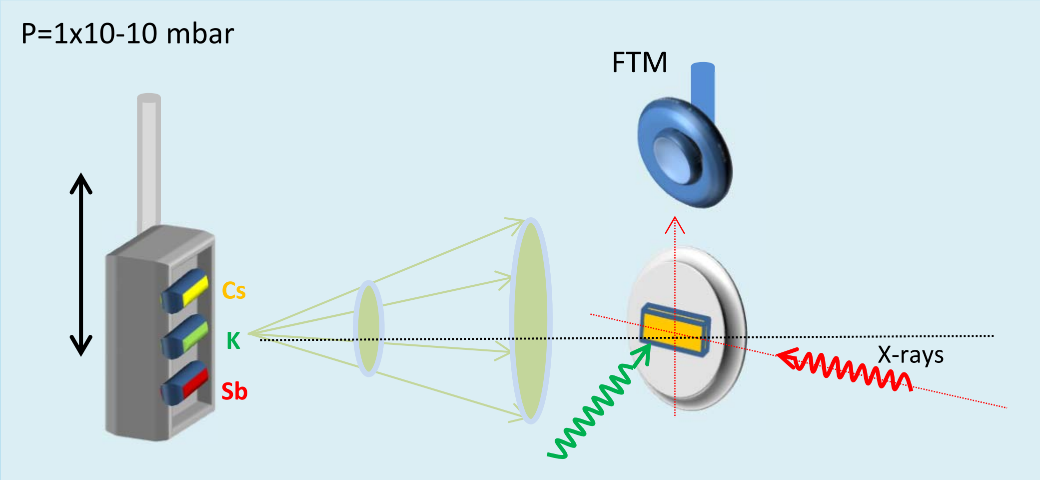
Two 2D detectors (Pilatus 100K) ⇨



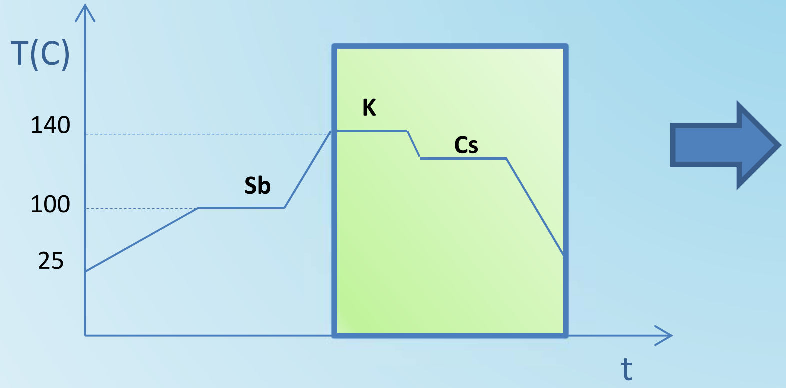
XRF, XRD, XRR, GISAXS, QE

Experimental set up: K_2CsSb cathode growth

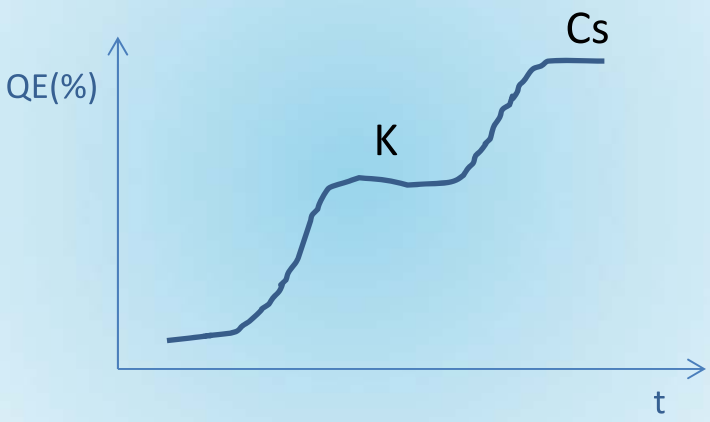
Horizontal evaporation of three sources:



Recipe:

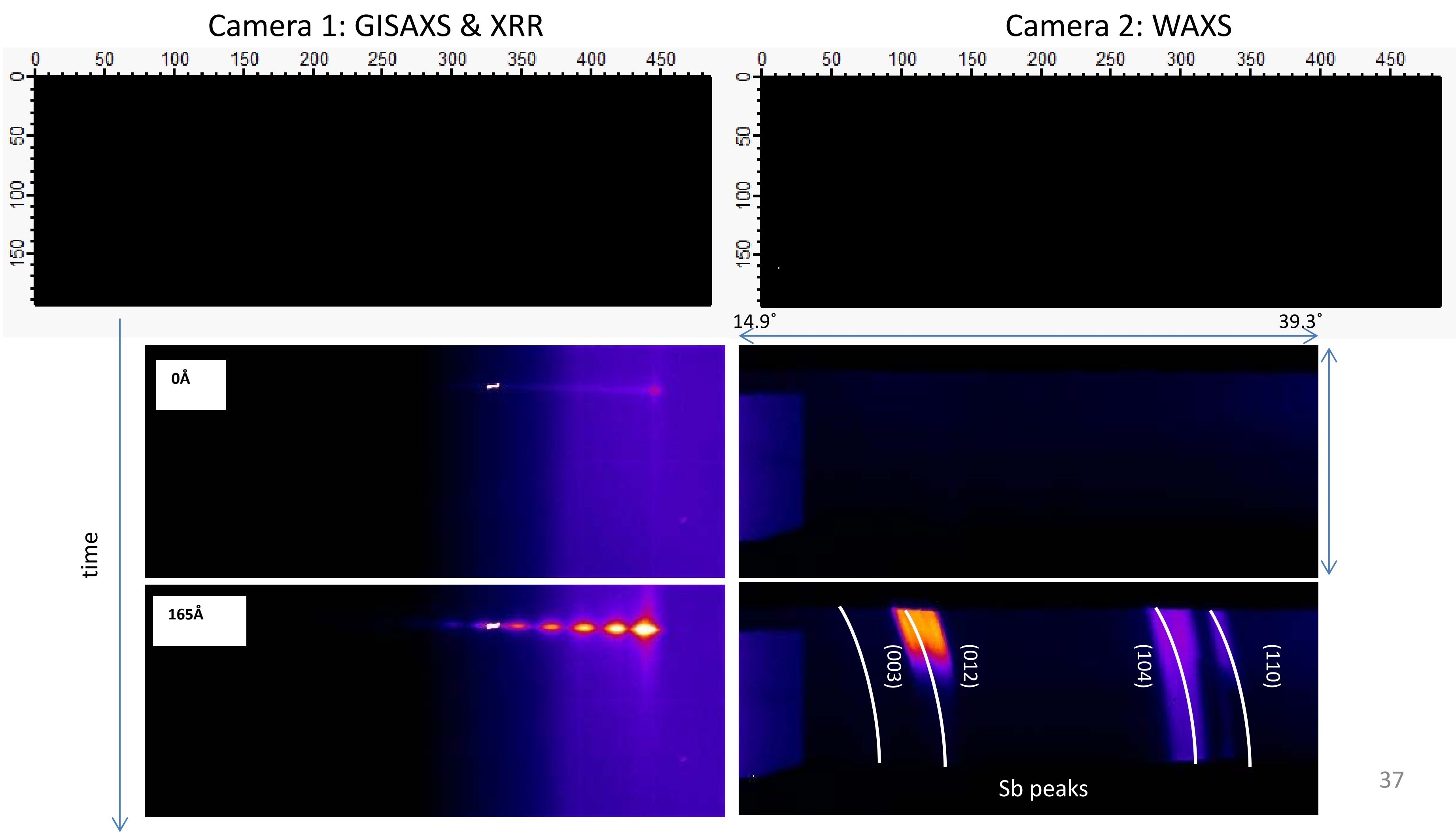


QE during growth (532 nm laser)

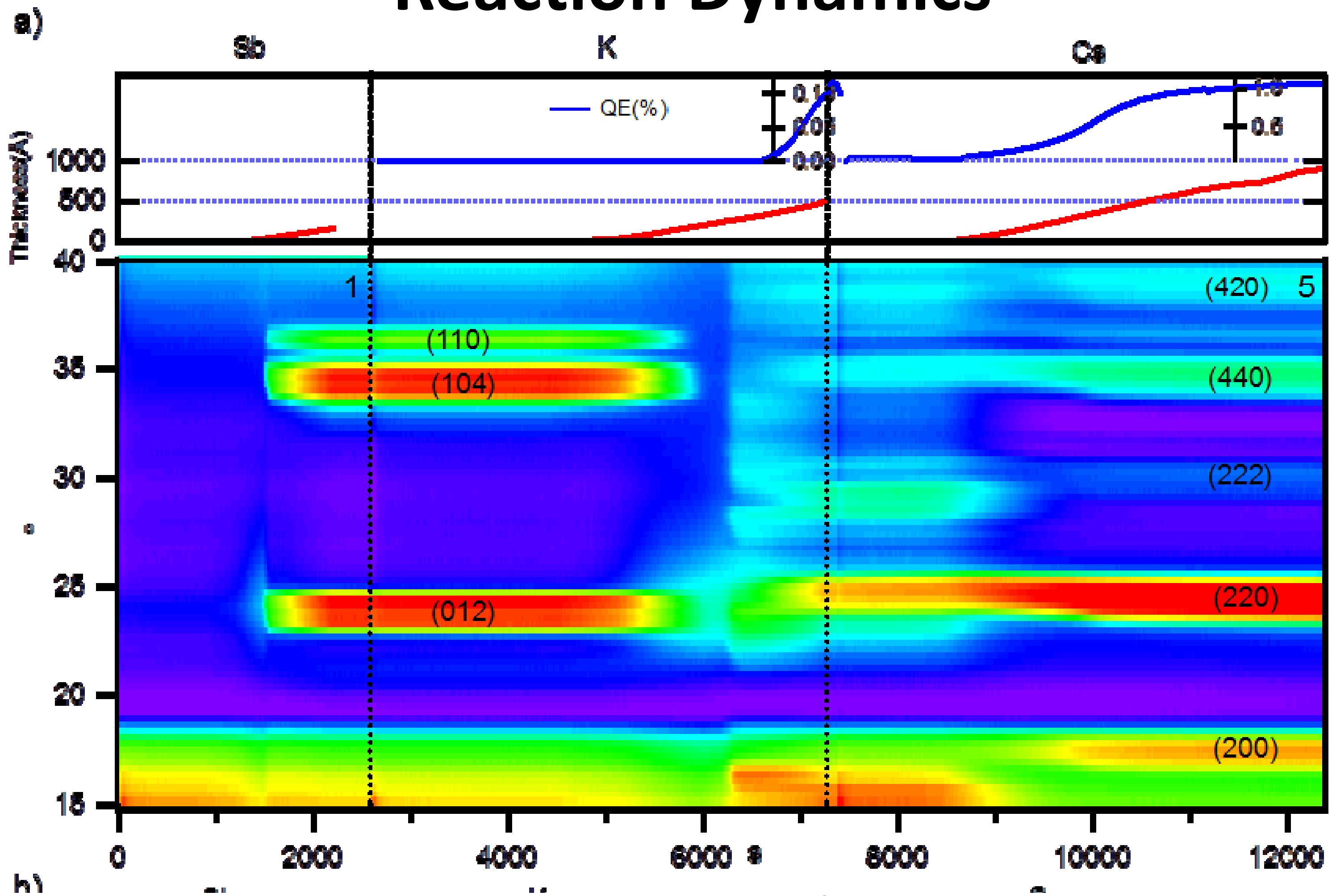


Simultaneously Acquire XRD and GISAXS

- Understanding reaction dynamics through crystalline phase evolution
- Map the thickness and roughness evolution of the cathode
- Is there a correlation between reactivity, QE and roughness?



Reaction Dynamics



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

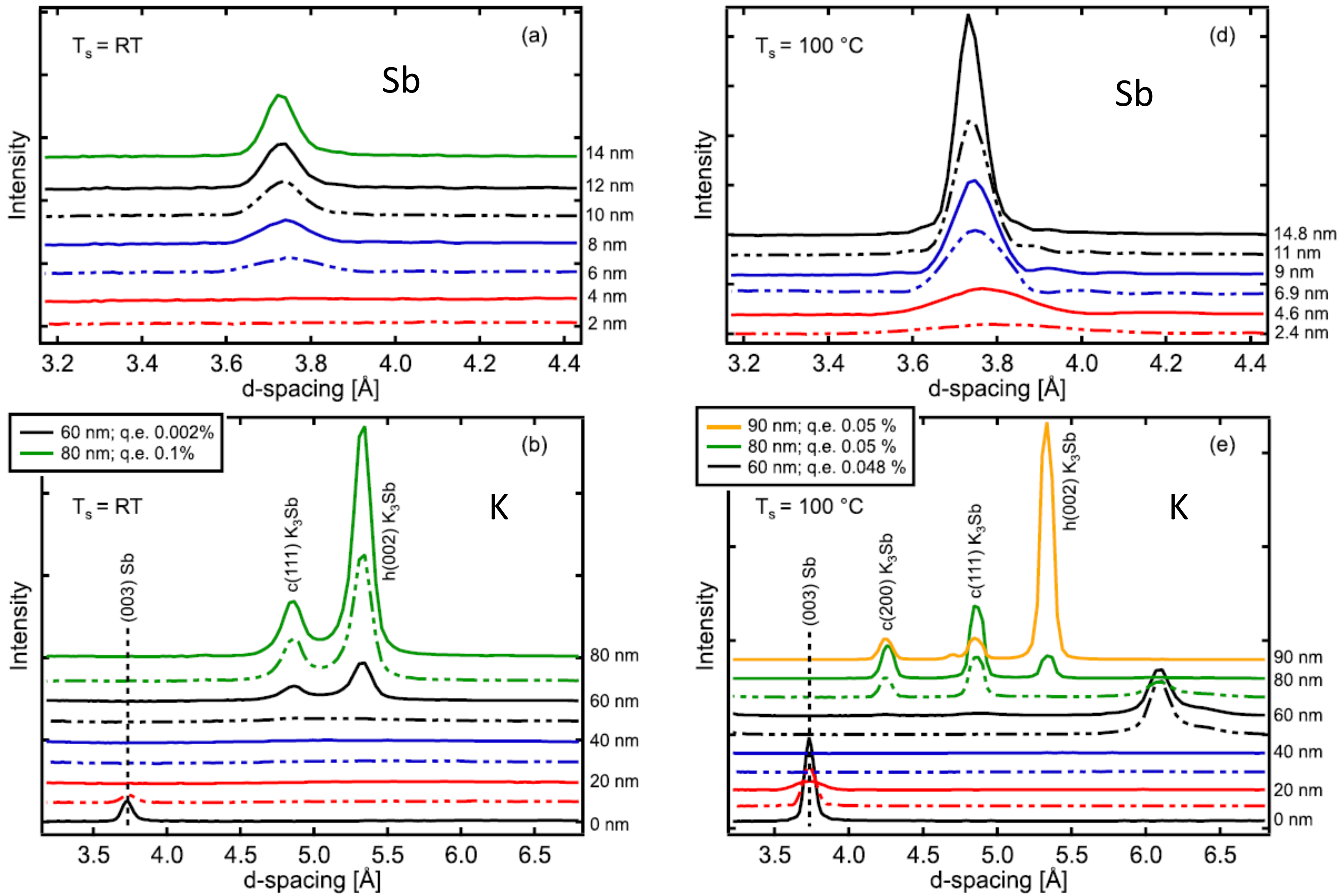
K deposition dissolves Sb layer - This is where roughening occurs!

QE increase corresponds with K_xSb crystallization

Cs increases lattice constant and reduces defects

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

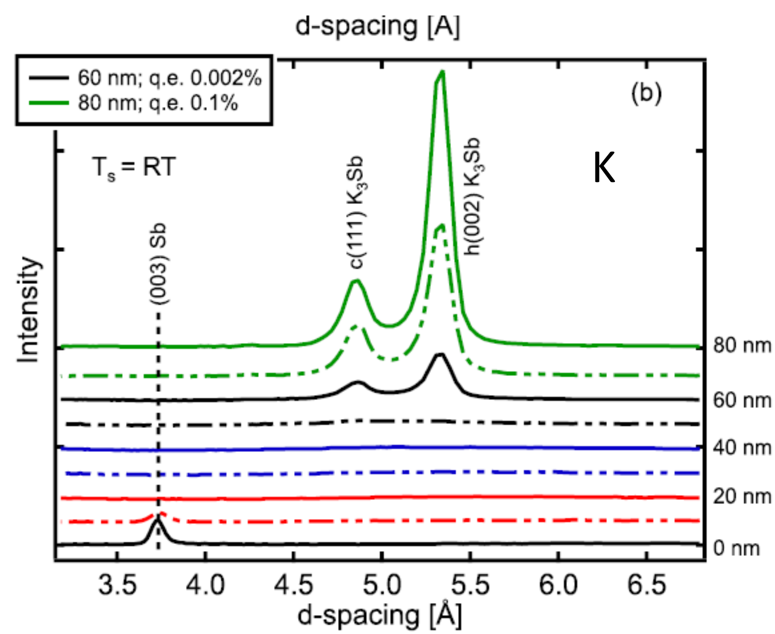
Stepwise High Resolution XRD
A little bit of Potassium goes a long way…

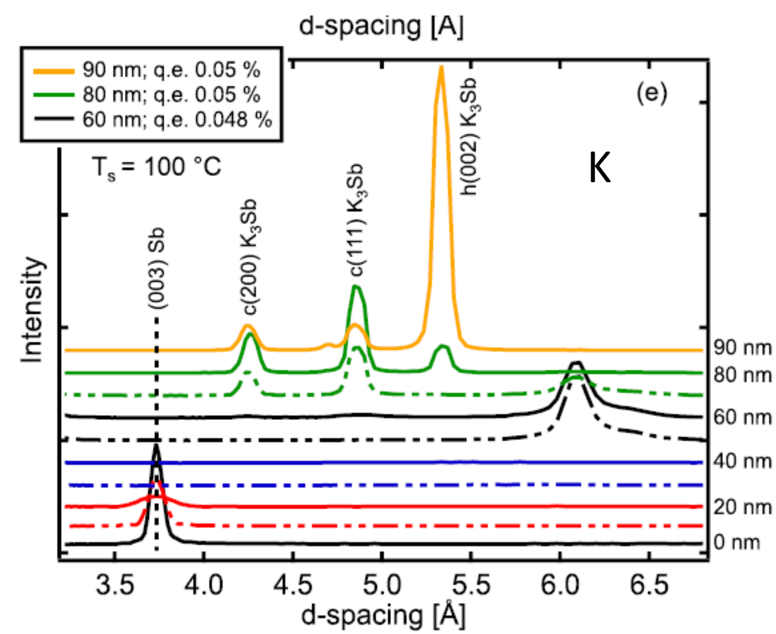


S. Schubert et al., J. Appl. Phys. 120, 035303 (2016)

Stepwise High Resolution XRD
A little bit of Potassium goes a long way…

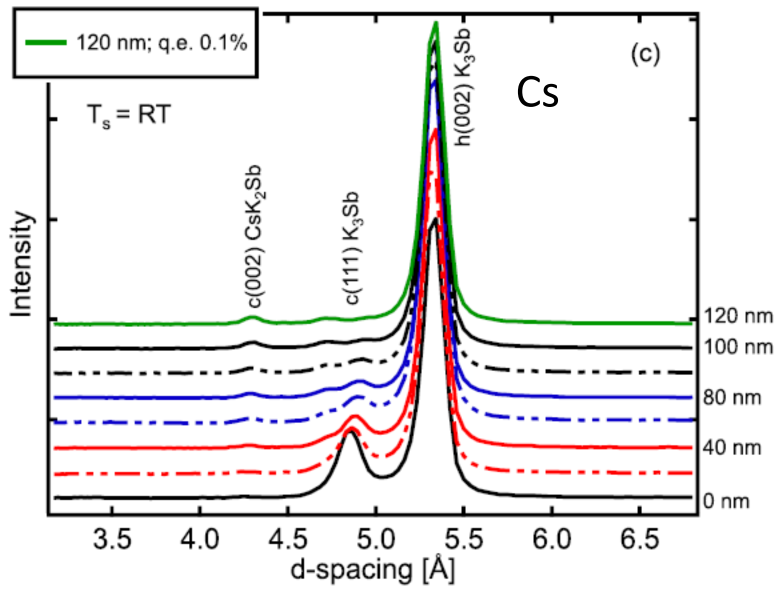
Room Temperature	100C Substrate
Recrystallization to K_3Sb	Recrystallization to K_xSb
Better QE of K_3Sb	Cubic K_3Sb first
Principally Hexagonal	Eventually Hex appears (oops!)



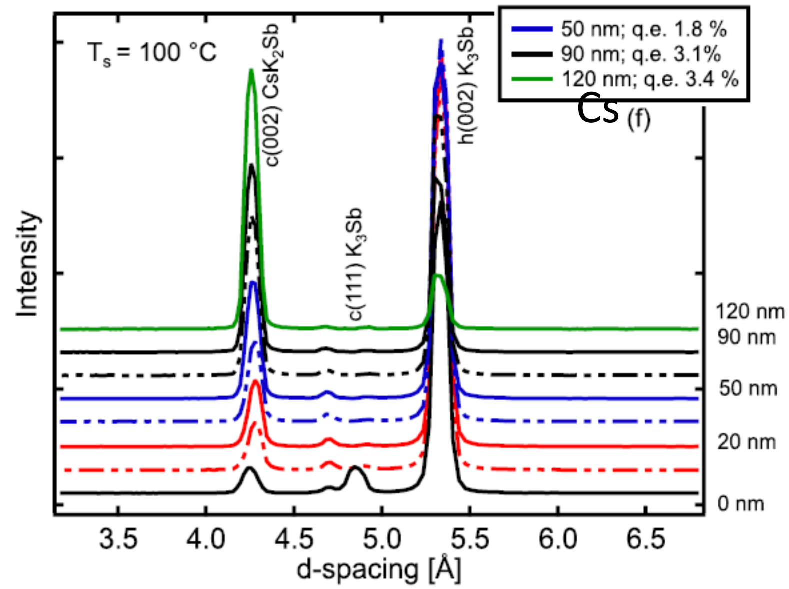


S. Schubert et al., J. Appl. Phys. 120, 035303 (2016)

Stepwise High Resolution XRD



Room Temperature



100C Substrate

K_3Sb resists Cs incorporation

QE never improves

Cubic K_3Sb converts quickly

Hex K_3Sb mostly converts

S. Schubert et al., J. Appl. Phys. 120, 035303 (2016)

Stepwise High Resolution XRD

100C without "too much" K

15 nm Sb, 70 nm K

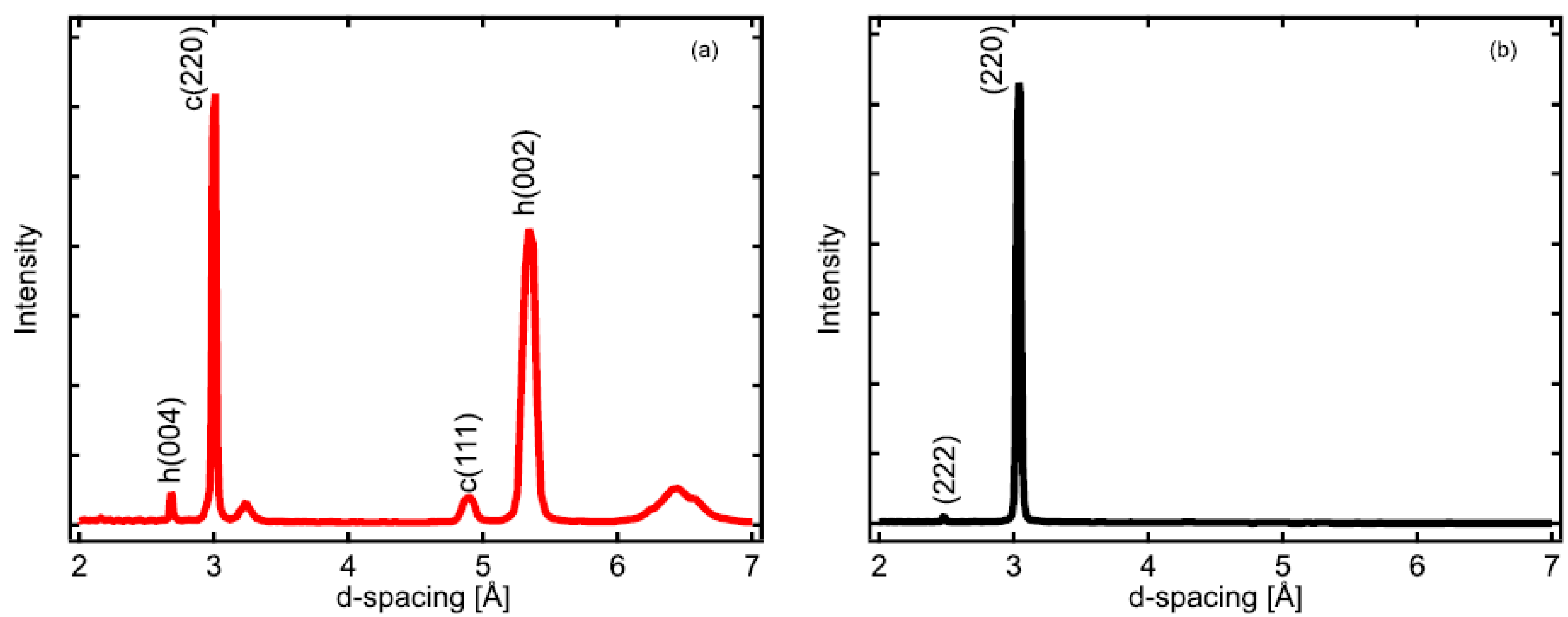
Stop at "mixed phase" K_xSb

lower QE of K_3Sb

90 nm Cs sufficient

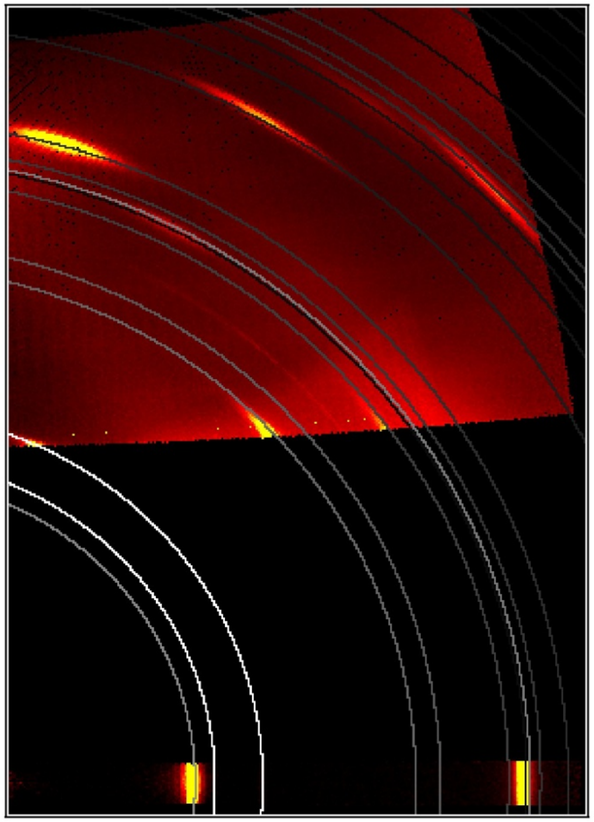
Full conversion to CsK_2Sb

QE = 6.7% at 532 nm

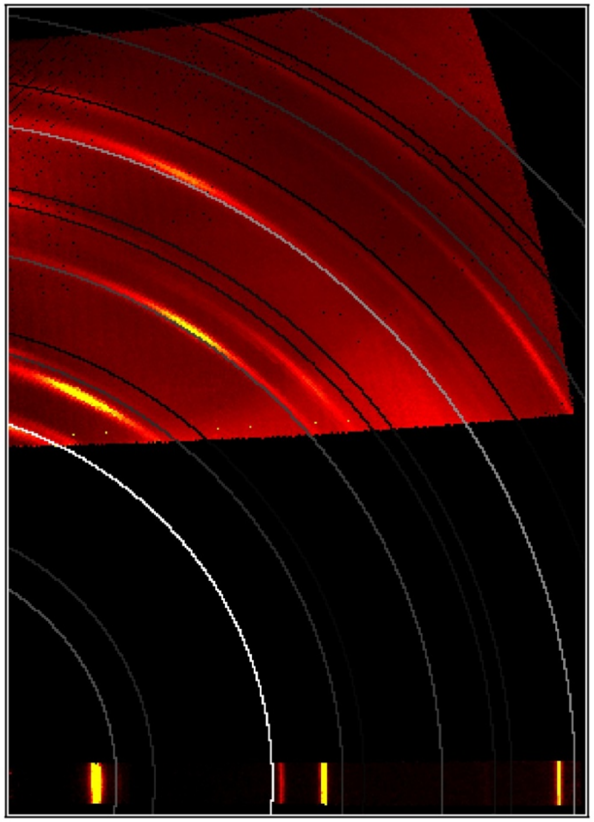


S. Schubert et al., J. Appl. Phys. 120, 035303 (2016)

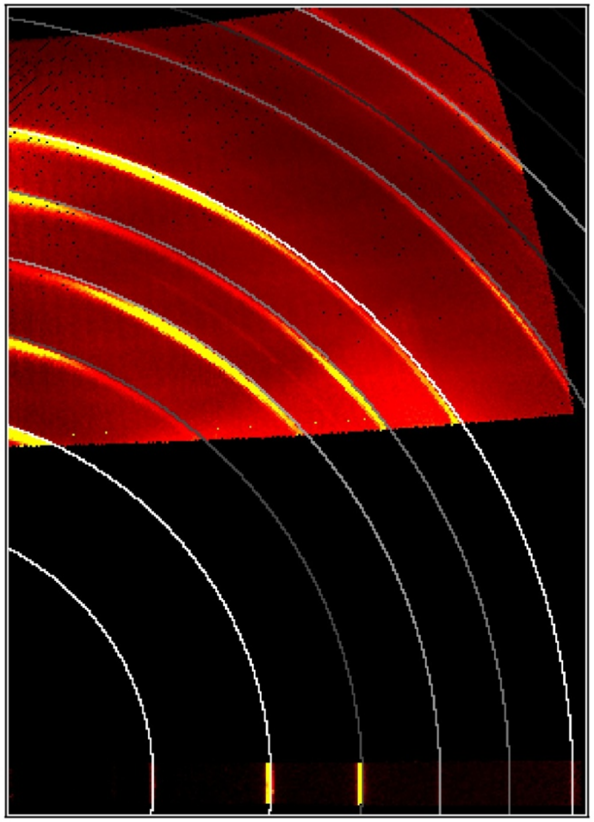
Cathode Texture



Sb evaporated at RT
Clear [003] texture



Add Potassium at 140C
Textured final film
But not K_3Sb



Add Cesium at 140C
Textured final film
Both [220] & [222]
(domains?)
Final QE 7.5% @ 532nm

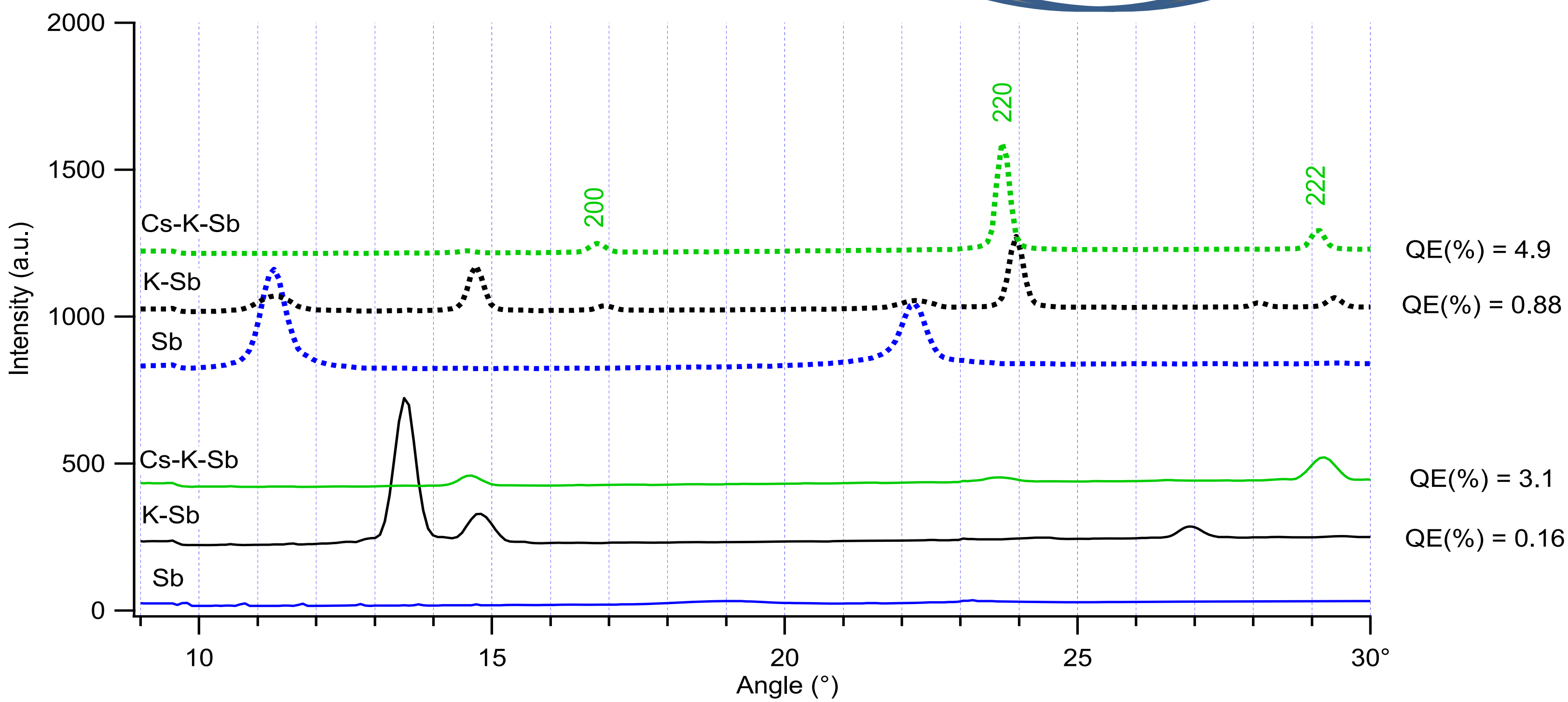
Engineering a Smoother Cathode

Idea: Never let Sb crystalize

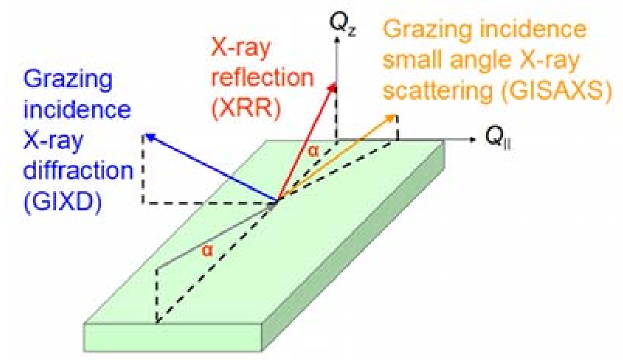
Substrate: Si (100) → 100 C, +3nm Sb → Sb → 125 ℃, +K until photocurrent peaks → K_xSb → 125 ℃, +Cs until photocurrent peaks → CsK_2Sb

2nd layer: +5nm Sb

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



X-ray reflectometry (XRR) provides in-situ thickness monitoring

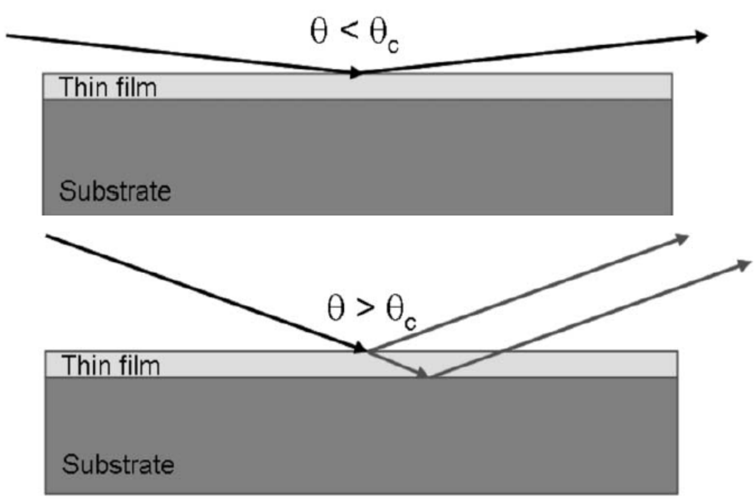


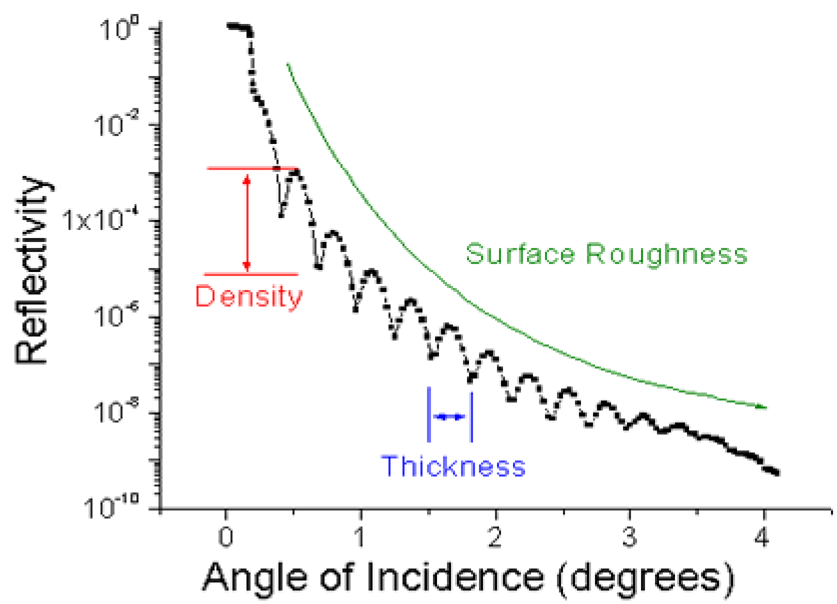
Understand 'sticking' coefficient of materials to substrates at various temperatures

Observe the intermixing vs layering of materials

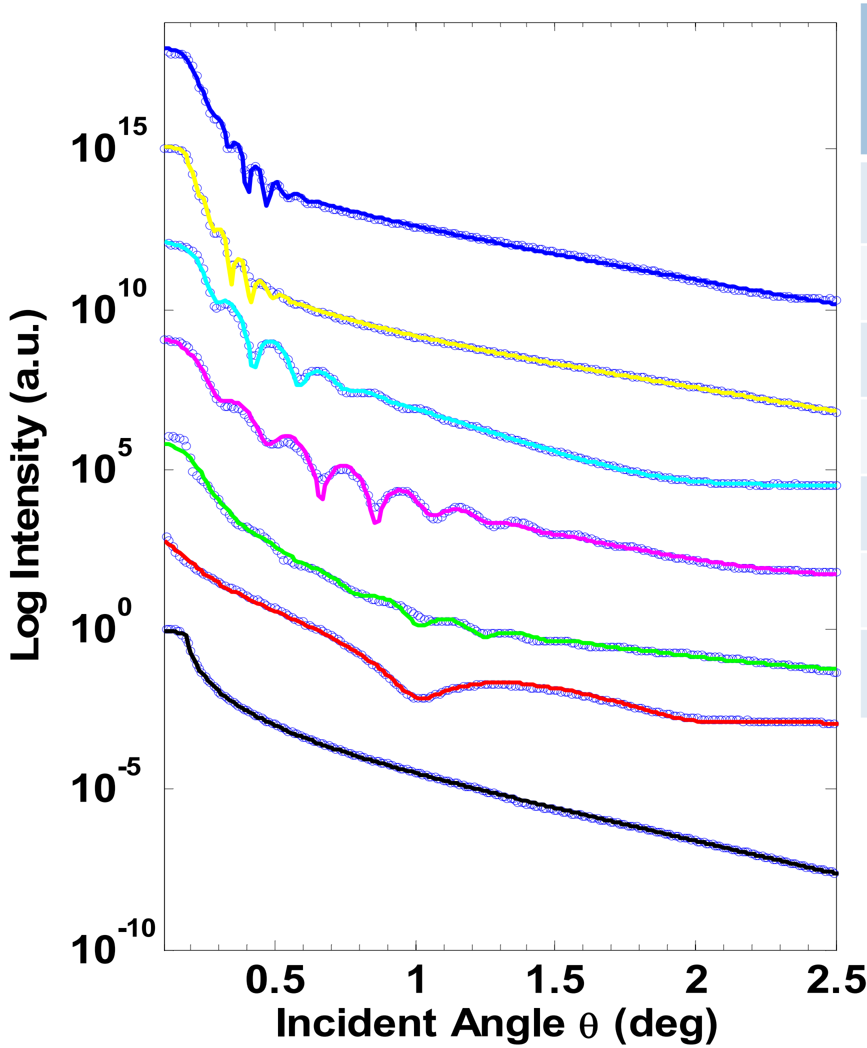
Observe the onset of roughness

$$\theta_c = \arccos(n_{medium} / n_{air})$$





XRR shows roughness evolution

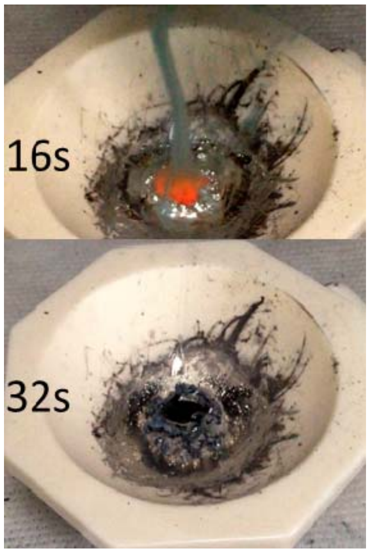


Deposited Layers	Total Thickness (Å)	Roughness (Å)
Cs-K-Sb-Cs-K-Sb/Si	469	32
K-Sb-Cs-K-Sb/Si	449	36
Sb-Cs-K-Sb/Si	200	21.3
Cs-K-Sb/Si	174	13.2
K-Sb/Si	141	10.5
Sb/Si	35	2.9
Si Substrate	-	3.1

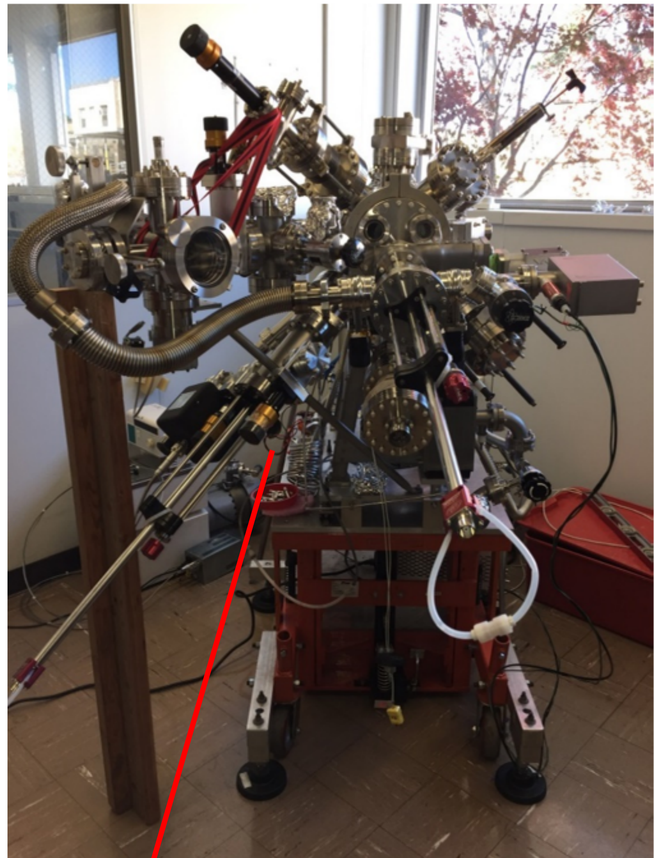
The substrate fit includes 1.5 nm of SiO_2

Multi-layer subcrystalline film is smoother,
At slight loss of QE

Sputter growth of Bi-alkali photocathode



Sputter target and sputter gun was product of RMD. Inc. Photos of sputter target prep are contributed by H. Bhandari



K_2CsSb sputter gun

Before

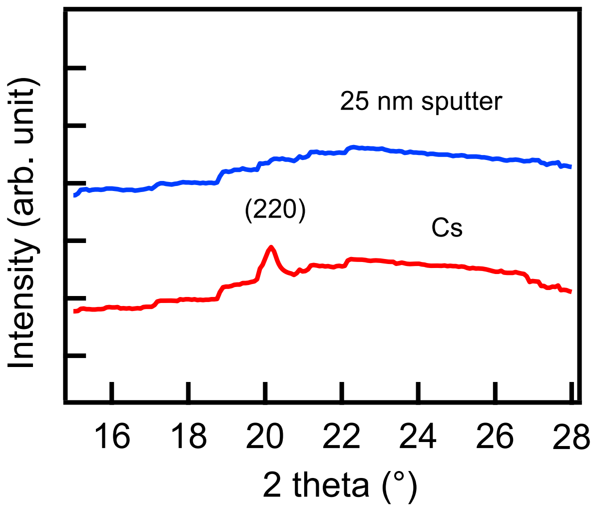
Sputtering

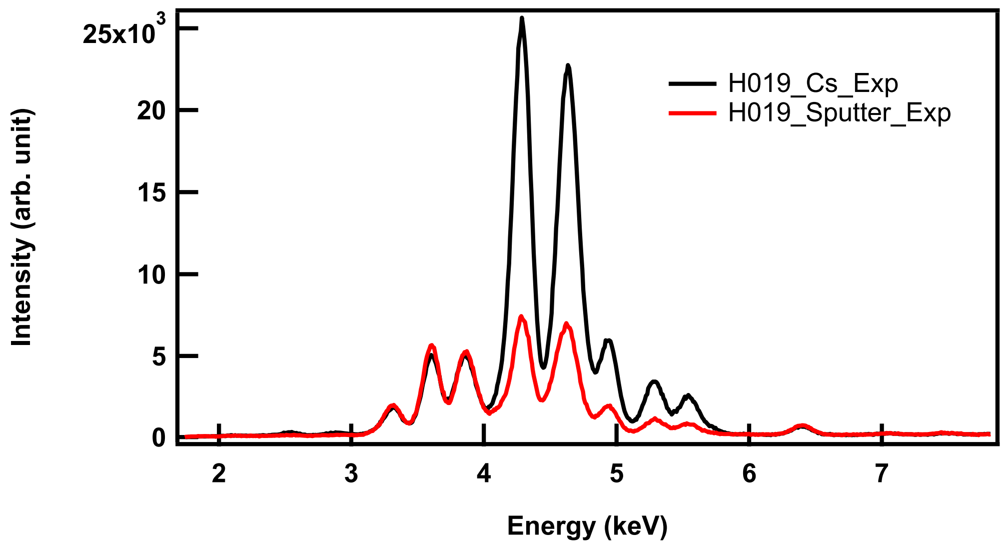
Sputter Growth

25 nm K_2CsSb + layers of (total 30 nm) Cs evap.

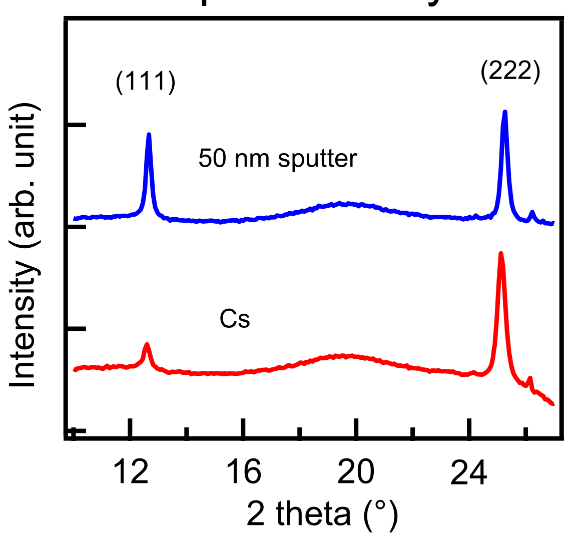
Silicon substrate at 90 C, layer barely crystalline

25 nm sputtered layer + Cs



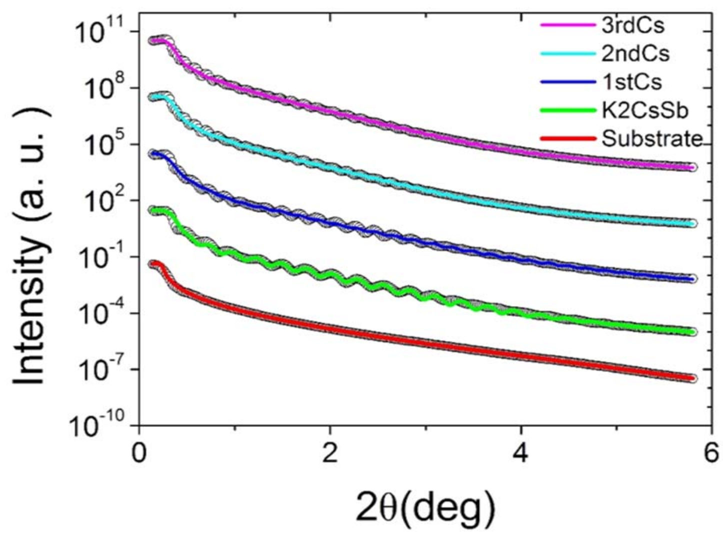


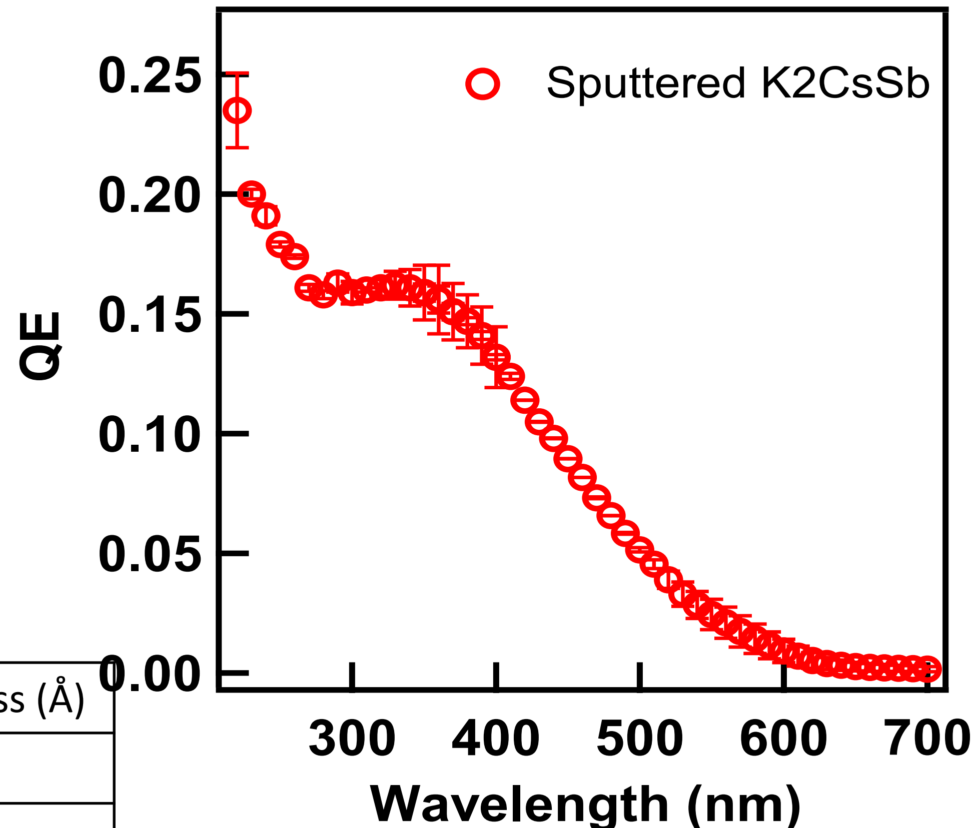
50 nm sputtered layer + Cs



layer	K (±0.1)	Sb (±0.05)	Cs (±0.05)	K/Cs
K_2CsSb sputter	0.85	1.00	0.41	2.08
Cs	0.84	1.00	1.75	0.48

Surface roughness & QE of Sputtered Photocathodes

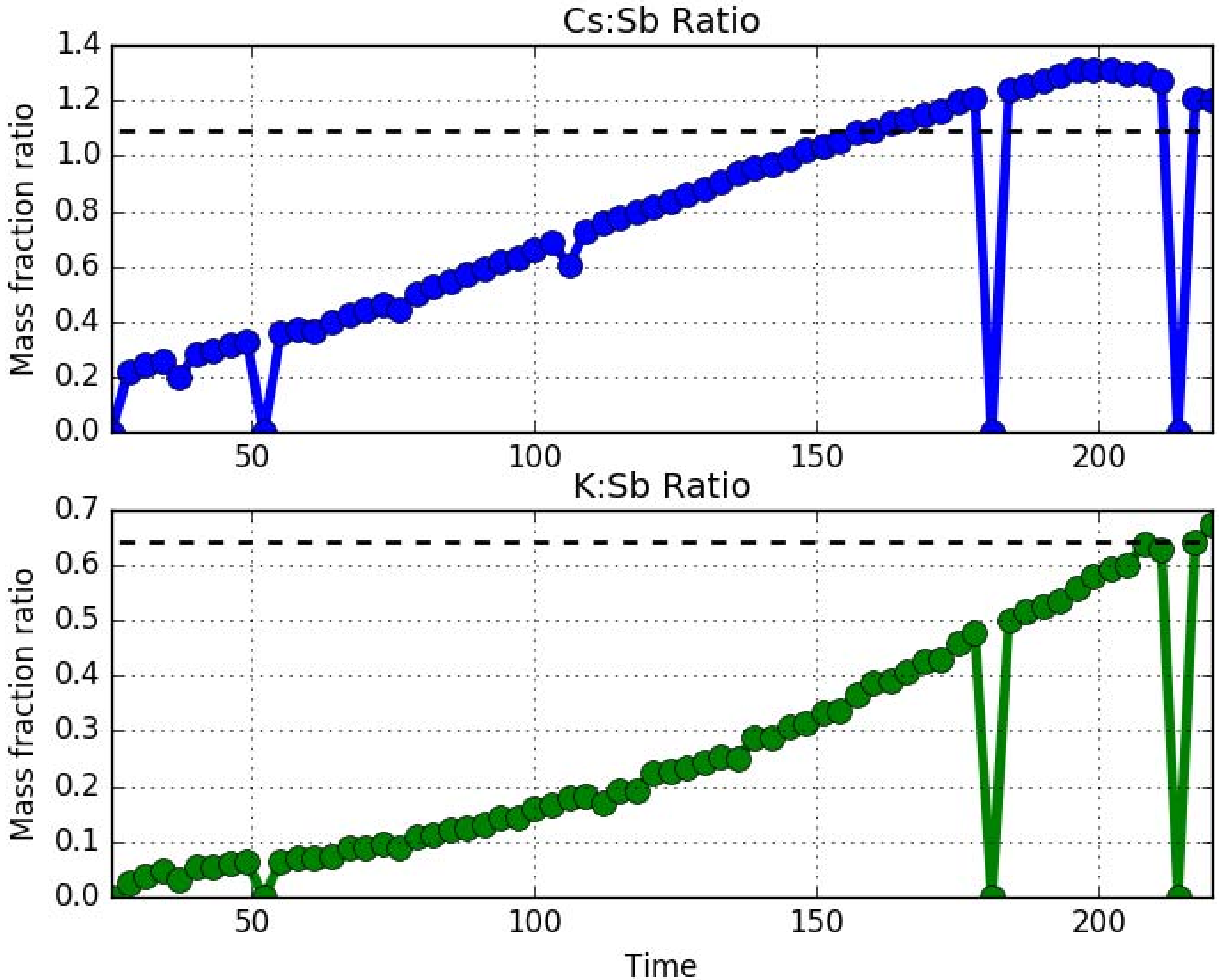




	Thickness (Å)	Roughness (Å)
3rd Cs	416.0	5.67
2nd Cs	341.3	4.94
1st Cs	249.5	4.91
sputter K_2CsSb	234.2	5.17
SiO_2	10.24	3.27
Substrate (Si)	---	3.75

- Peak QE > 20%
- Green QE: 4.3%

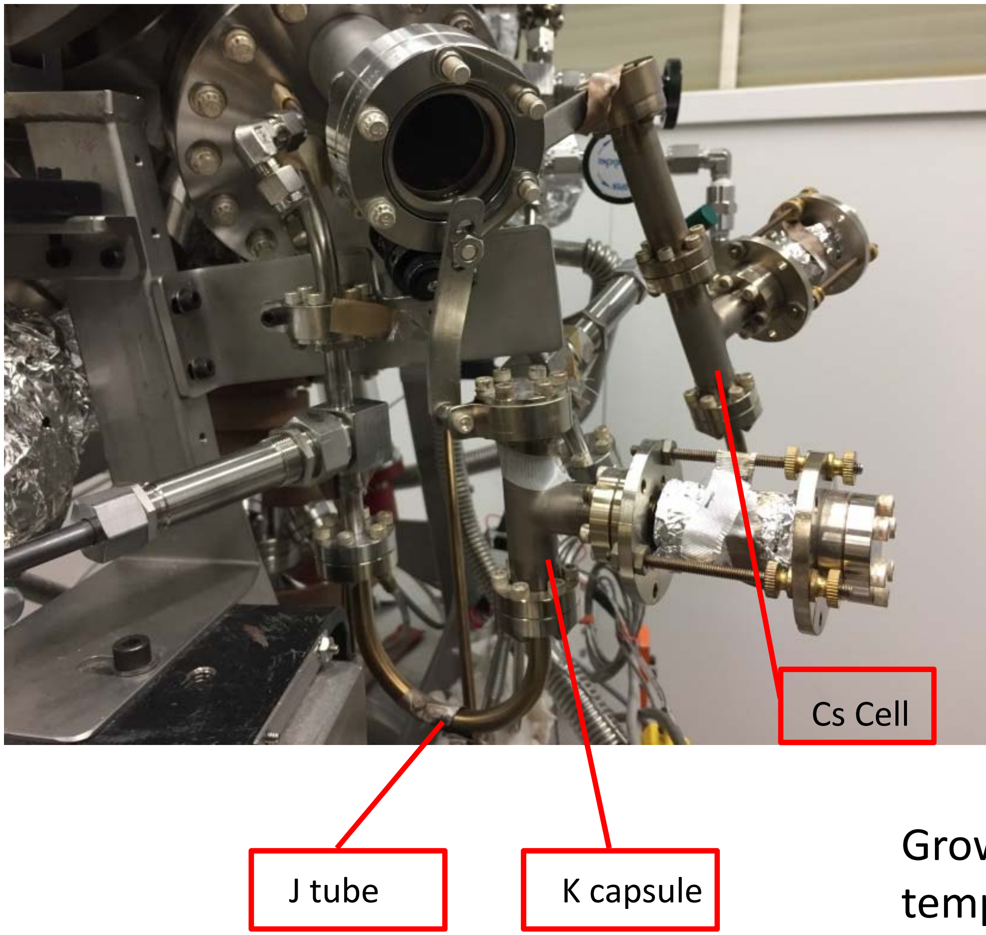
Software Upgrades - Real Time XRF during growth

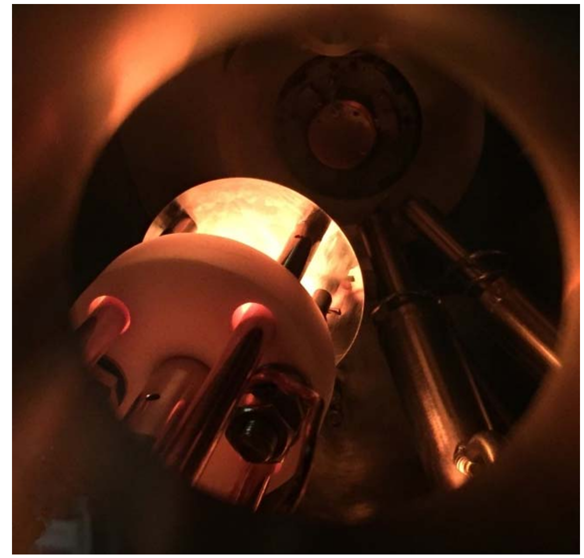


Currently using to regulate ternary co-evap

Ternary Co-evaporation

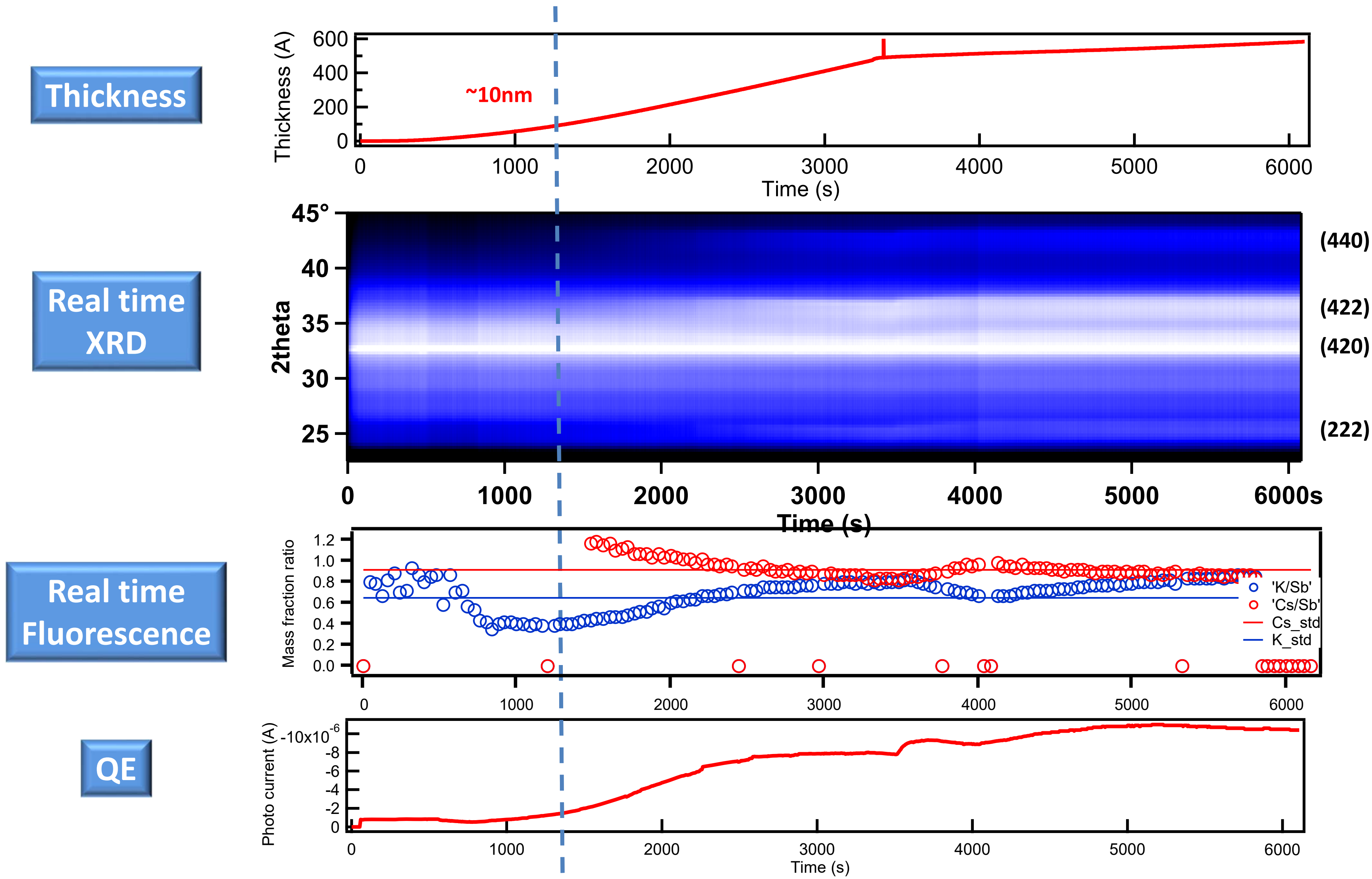
Simultaneously evaporate from Sb evaporator and K,Cs effusion cells





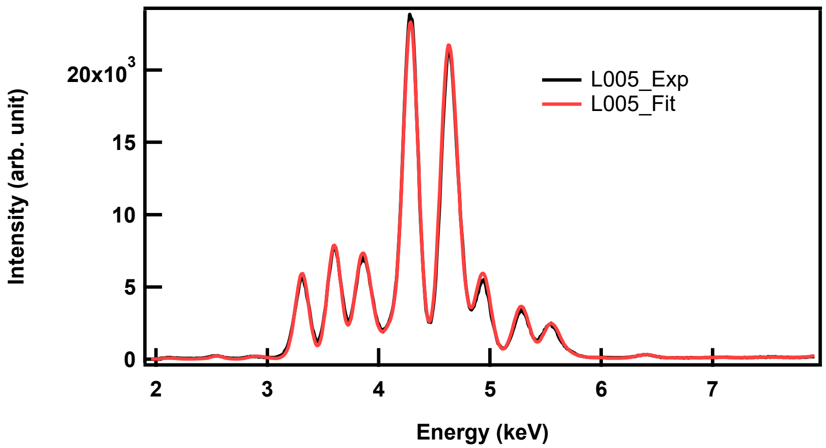
In situ, In operado XRR, XRF, XRD & Quantum efficiency (QE) measurement

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

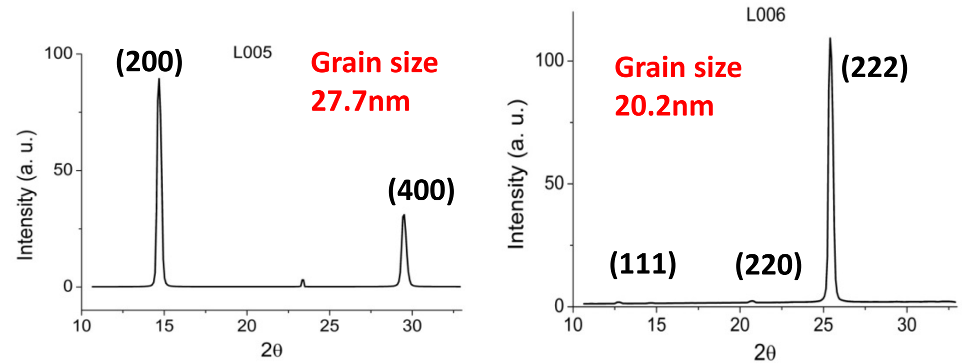


Thickness
Real time XRD
Real time Fluorescence
QE
Thickness (A)
~10nm
Time (s)
2theta
45°
(440)
(422)
(420)
(222)
6000s
Time (s)
Mass fraction ratio
'K/Sb'
'Cs/Sb'
Cs_std
K_std
Photo current (A)
-10x10^-6
Time (s)

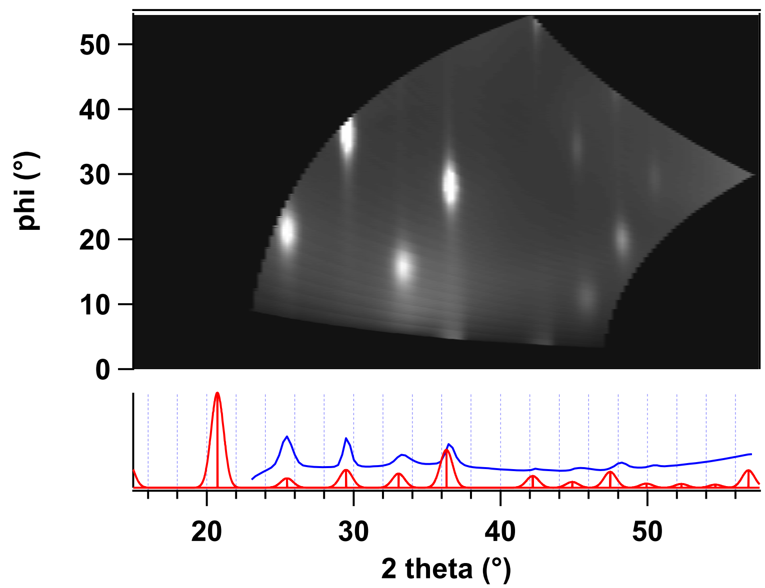
Stoichiometry & Structural Analysis



Camera 1



Camera 2

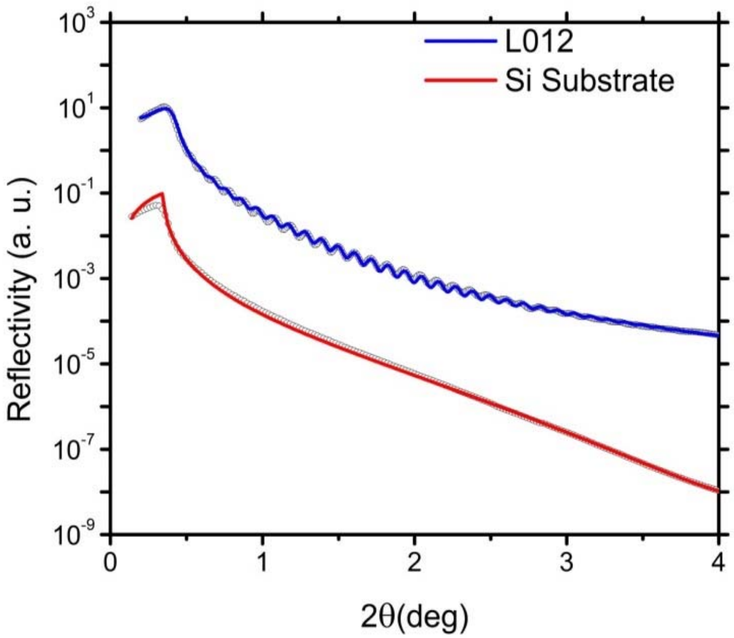


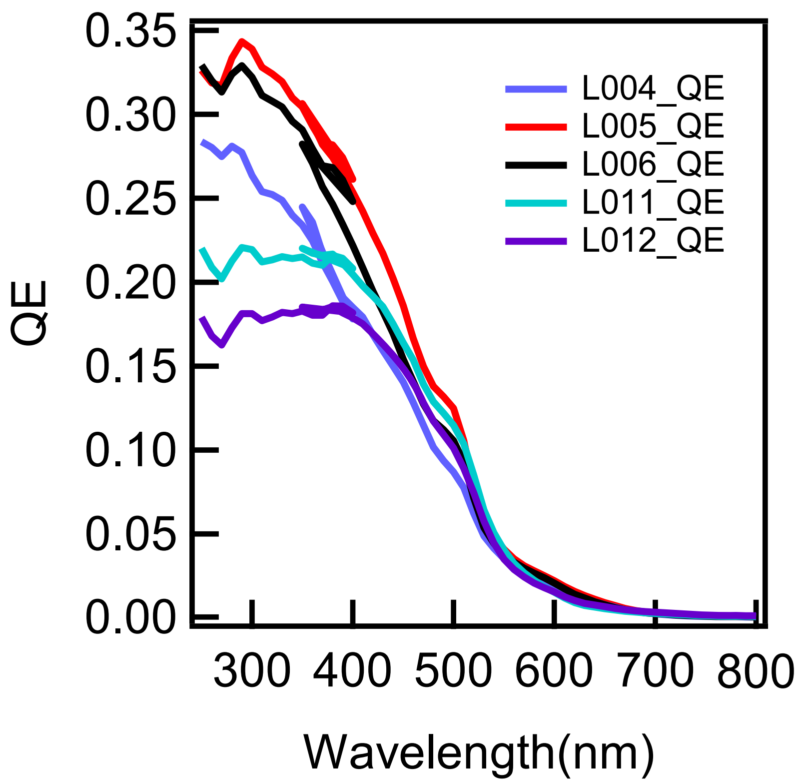
Highly textured K_2CsSb phase!

	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!

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
L011 Si (HT Si)	6.4	79.0	905.2	208
L012 MgO	5.7	8.6	566.4	193

Summary

Alkali Antimonides

We now have a tool which is capable of optimizing growth parameters for figures of merit other than Quantum Efficiency

We understand the formation chemistry of these materials, and why traditional deposition results in rough cathodes

RMS roughness down almost 2 orders of magnitude, to ~atomic scale

Avoiding crystalline Sb helps, as does co-evaporating alkali

Sputter deposition is good – easy to do, covers large area, almost atomically smooth even for thick films – but alkali poor

Real time XRF feedback provides option of ternary coevaporation, producing best cathode

Lead

Significantly superior QE performance compared to Nb

Modest laser cleaning provides the theoretical QE

Lead does not affect cavity RF performance, and the laser does not cause quenching

More work required to achieve a smooth enough surface for realistic low emittance operation

Thanks for your attention!

Thanks to Jacek Sekutowicz

P. Kneisel, K. Attenkofer, S. Schubert, M. Ruiz Oses, J. Xie, J. Wang, H. Padmore, E. M. Muller, M. Gaowei, J. Walsh, J. Sinshiemer, Z. Ding, R. Nietubyc

DOE Office of Science – Basic Energy Science