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Stoichiometry control and automated growth of alkali antimonide photocathode films by molecular beam deposition

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AFFILIATIONS

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ABSTRACT

We report on a method of photoemissive film growth that controls stoichiometry in real time. We show that stoichiometry control using a

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  Alexander Scheinker
  Ryan L. Fleming
  Anna Alexander
  Mark A. Hoffbauer
  Nathan A. Moody

ACERT
APPLIED CATHODE ENHANCEMENT AND ROBUSTNESS TECHNOLOGIES

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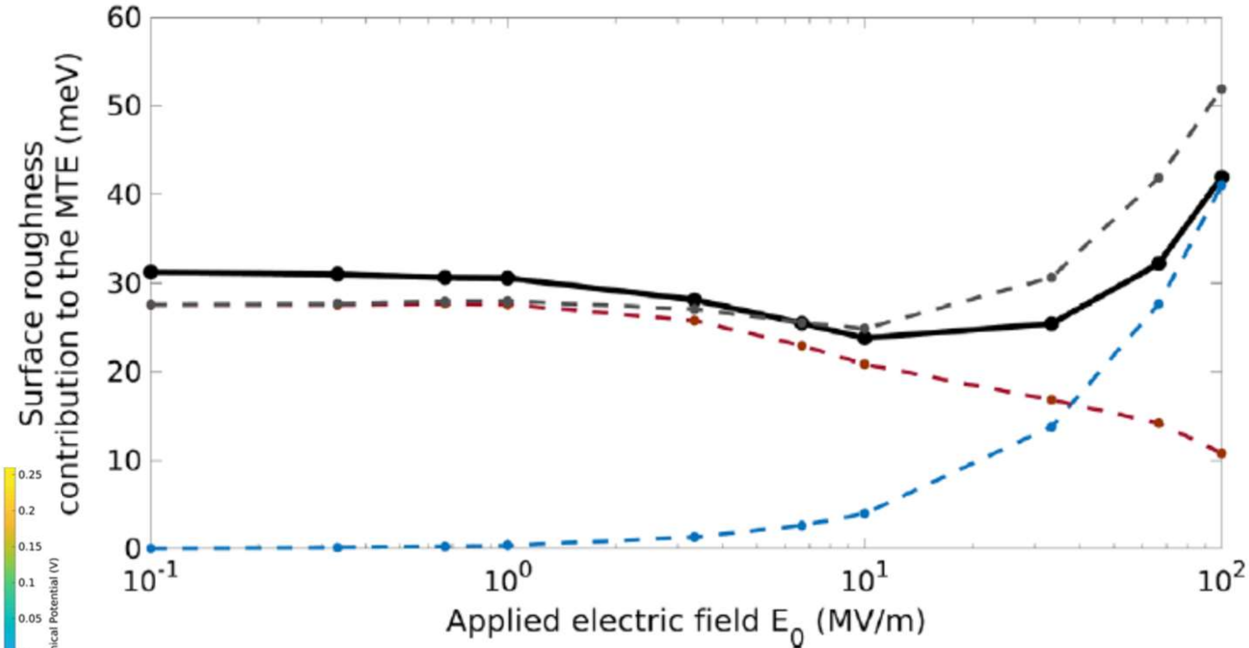
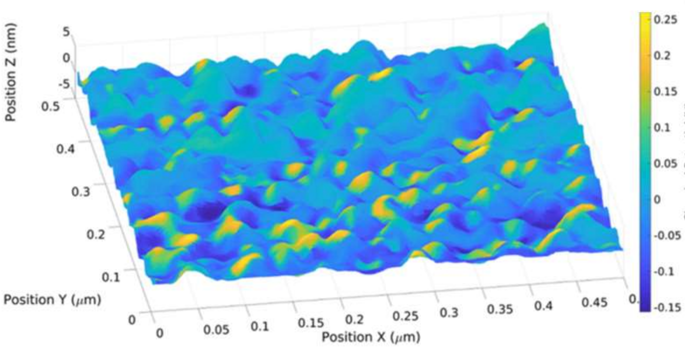
INTRO: Roughness is always bad for MTE

kT+





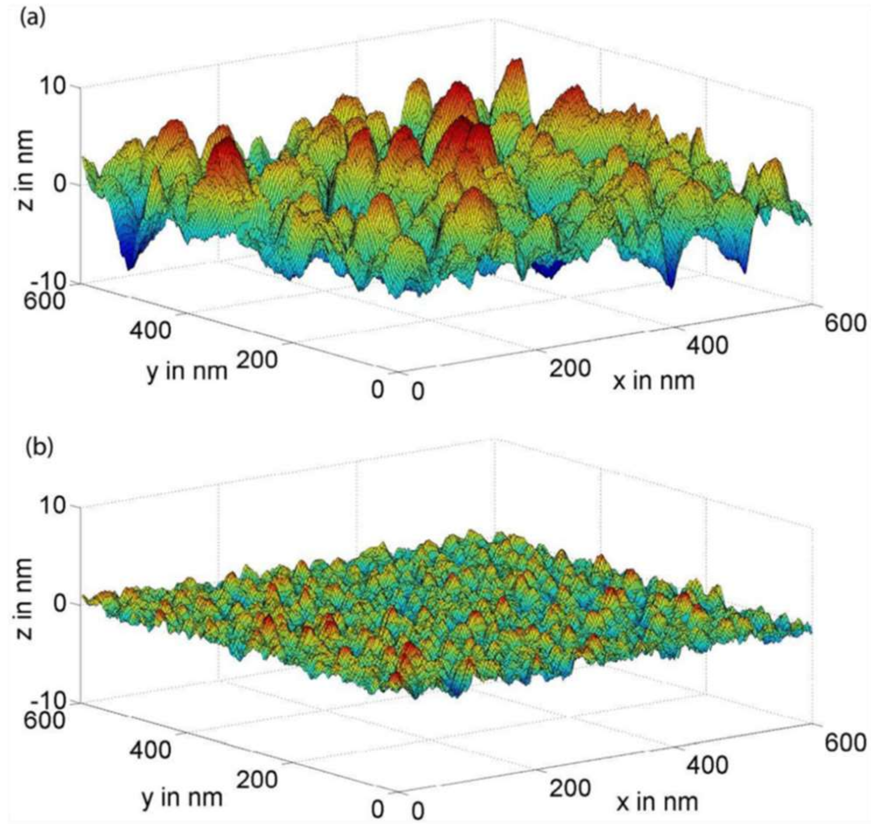
From: G. S. Gevorkyan, S. Karkare, S. Emamian, I. V. Bazarov, and H. A. Padmore, Effects of physical and chemical surface roughness on the brightness of electron beams from photocathodes, Phys. Rev. Accel. Beams **21**, 093401 (2018)

INTRO: Co-deposition produces smoother films

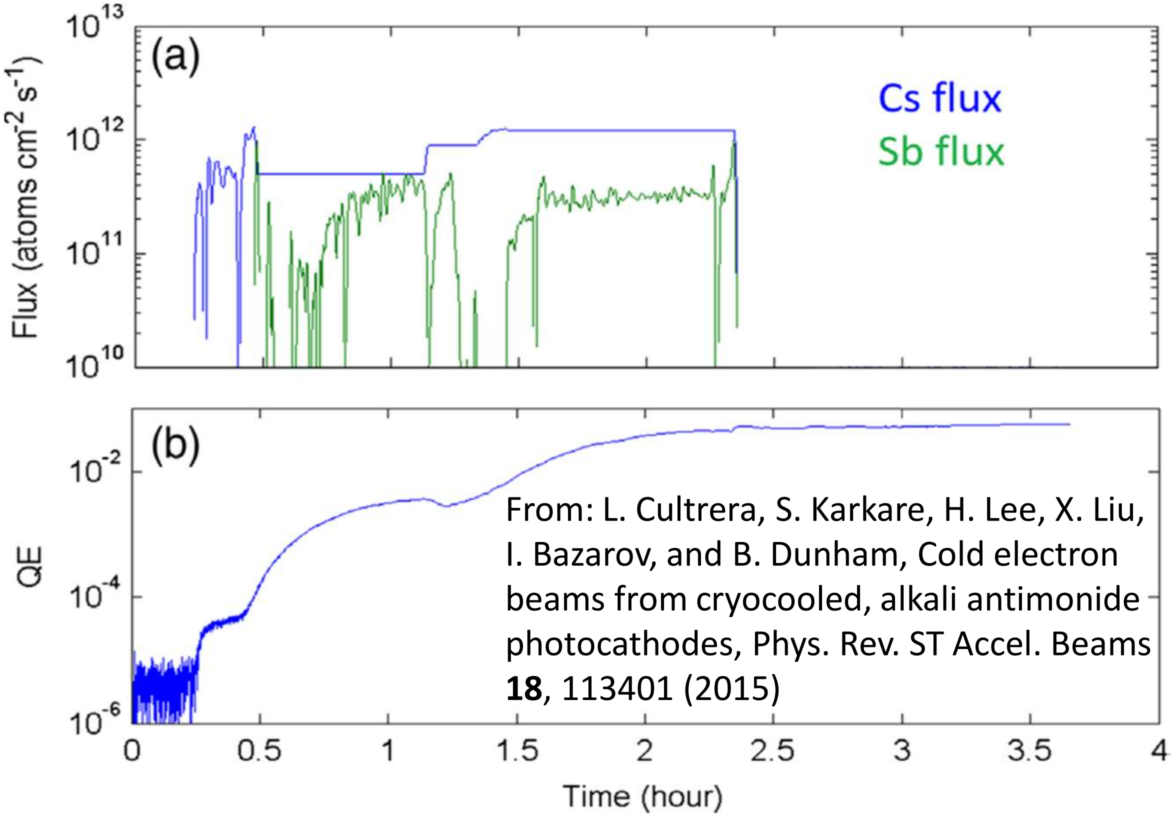
From: Jun Feng, Siddharth Karkare, James Nasiatka, Susanne Schubert, John Smedley, and Howard Padmore, Near atomically smooth alkali antimonide photocathode thin films, Journal of Applied Physics **121**, 044904 (2017)

sequential deposition

co-deposition



INTRO: Real-world co-deposition of mono-alkalis



From: L. Cultrera, S. Karkare, H. Lee, X. Liu, I. Bazarov, and B. Dunham, Cold electron beams from cryocooled, alkali antimonide photocathodes, Phys. Rev. ST Accel. Beams **18**, 113401 (2015)

Fluxes are being adjusted, what is the algorithm?

"… many growers adjust their reactant fluxes to maximize the quantum efficiency (QE) of the growing photocathode at a convenient wavelength."

From: Alice Galdi, William J. I. DeBenedetti, Jan Balajka, Luca Cultrera, Ivan V. Bazarov, Jared M. Maxson, and Melissa A. Hines, The effects of oxygen-induced phase segregation on the interfacial electronic structure and quantum efficiency of Cs_3Sb photocathodes, J. Chem. Phys. **153**, 144705 (2020)

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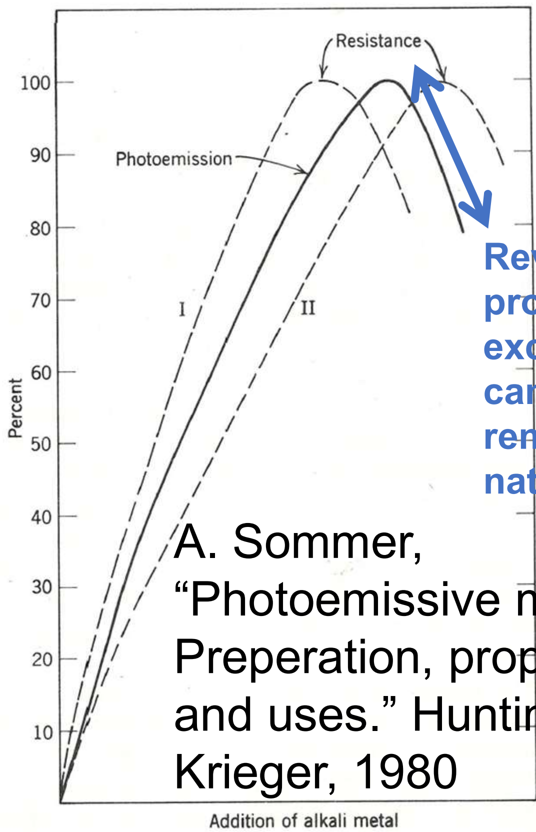
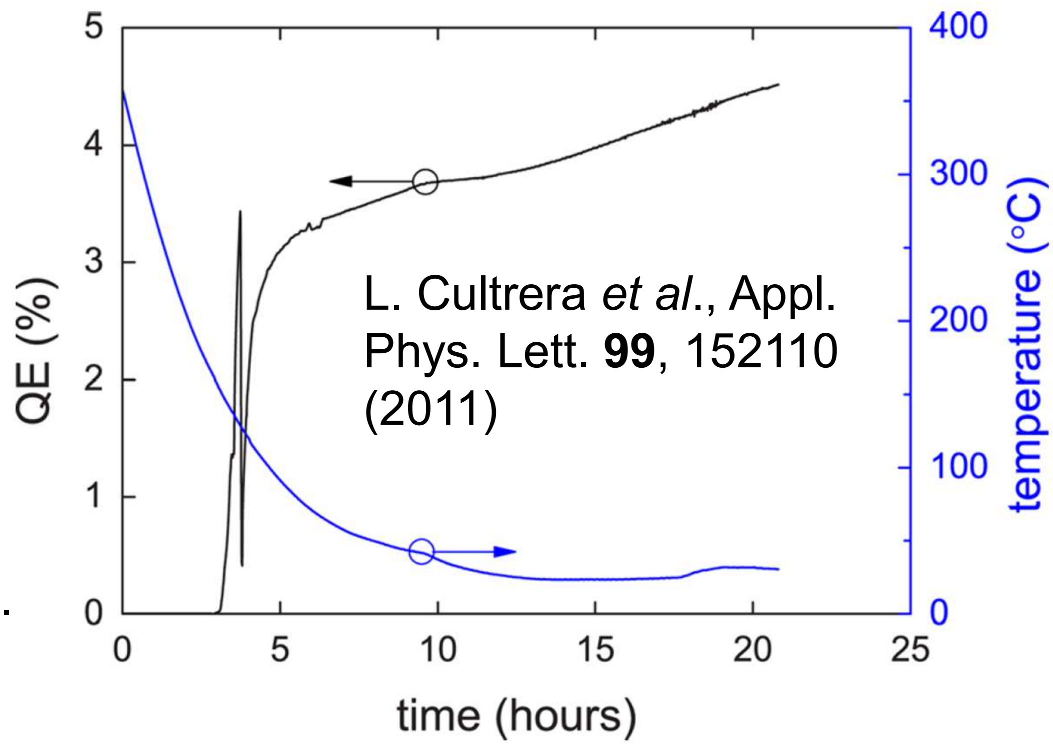
INTRO: What does maximizing QE mean?



Reversible process, excess alkali can be removed naturally

A. Sommer, “Photoemissive materials. Preperation, properties and uses.” Huntington: Krieger, 1980

Figure 21. Variation of resistance and photoemission during formation of alkali antimonide photocathodes: curve I for *n*-type materials and curve II for *p*-type materials.



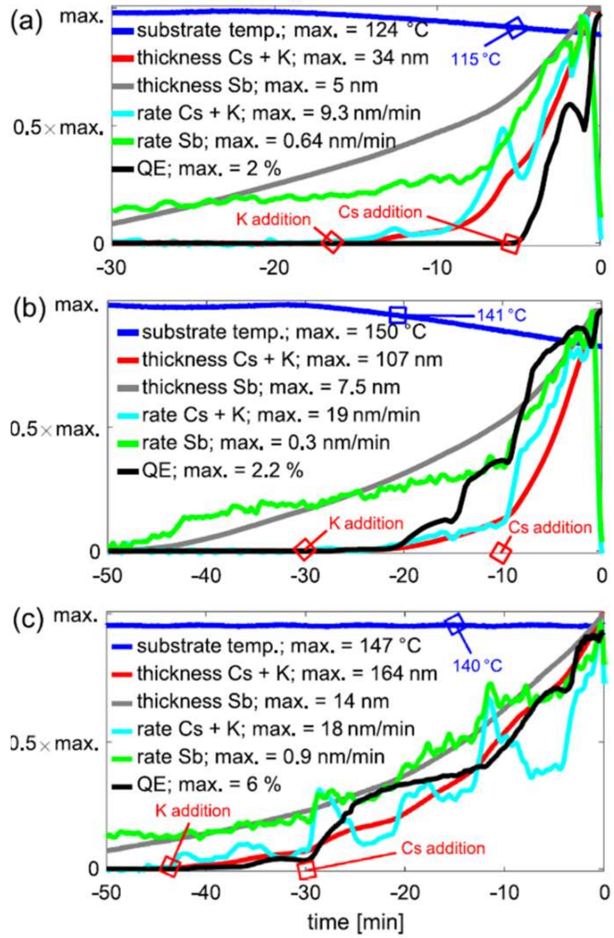
L. Cultrera *et al.*, Appl. Phys. Lett. **99**, 152110 (2011)

Max QE = Stable “Stoichiometric” Compound

But QE is a function of film thickness

INTRO: Real-world co-deposition of bi-alkalis

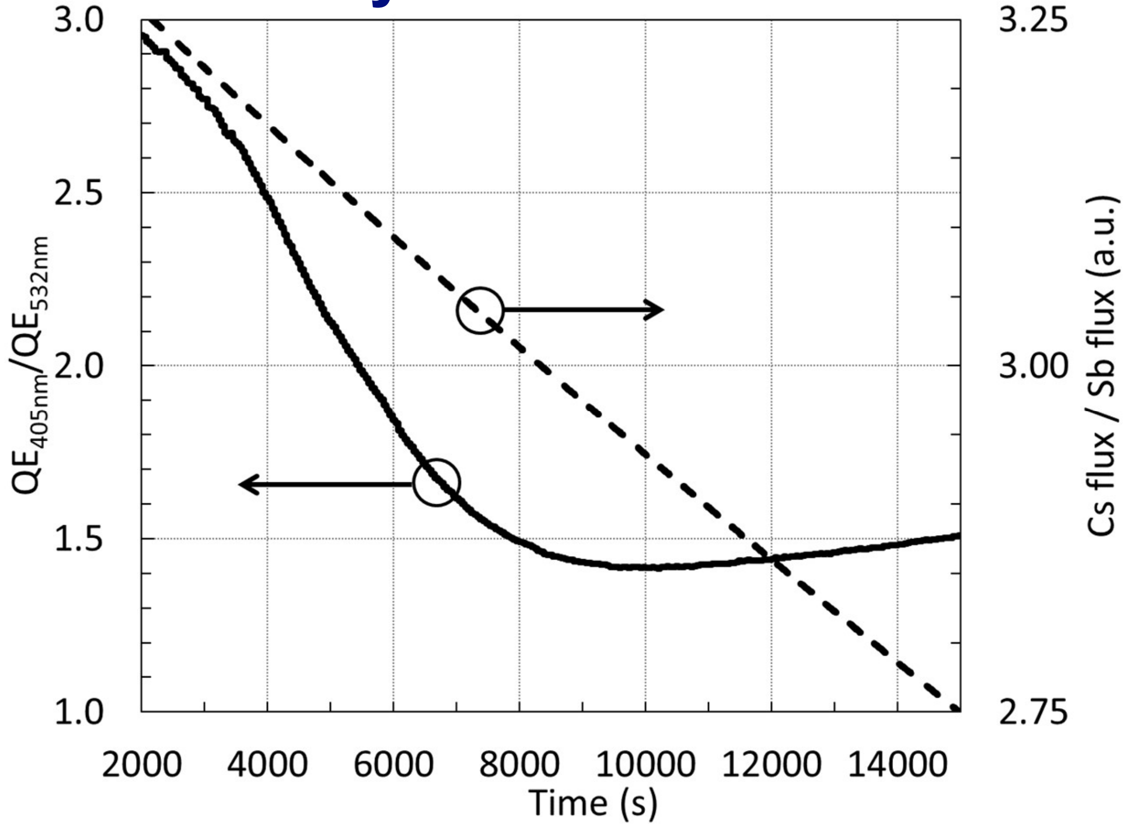
From: H. Panuganti, E. Chevallay, V. Fedosseev, M. Himmerlich, Synthesis, surface chemical analysis, lifetime studies and degradation mechanisms of Cs-K-Sb photocathodes, Nuclear Inst. and Methods in Physics Research, A **986** (2021) 164724



Algorithm description:
“slope of the *in situ* photocurrent as the driver for the growth process”

Human-mediated algorithms typically do not work well.

Thickness-independent photoemission parameter(s) vs stoichiometry



***Corroboration**: excess Cs removal experiments & thermal decomposition experiments*

- Time axis effectively represents stoichiometry
- Cs/Sb atomic flux ratio close to 3 for low substrate temperature
- Minimum of QE_{405nm}/QE_{532nm} approximately coincides with QE maximum
- To facilitate process control, need to remain is Cs-rich growth mode, otherwise need ***elaborate** extremum seeking*

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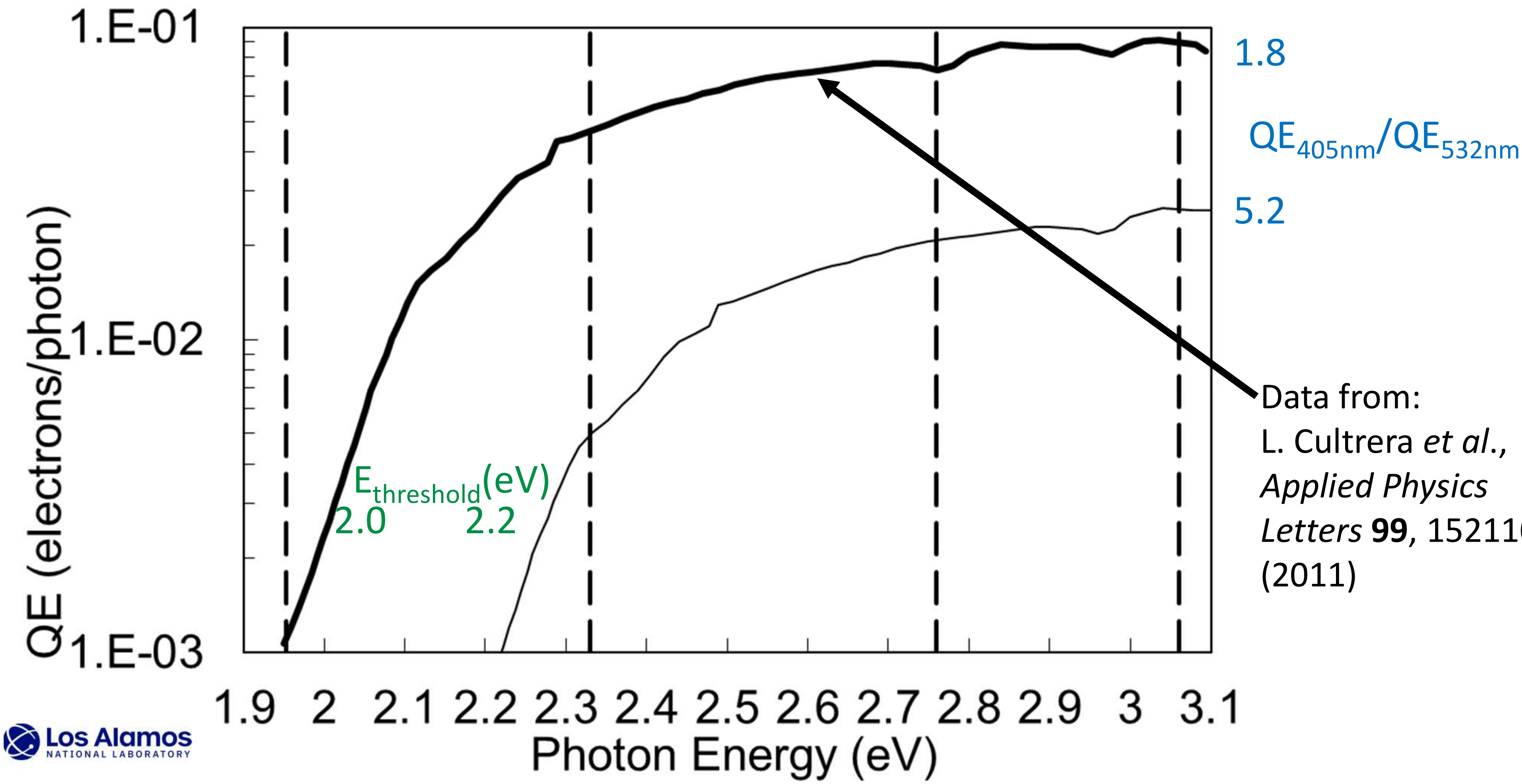
Why does QE_{405nm}/QE_{532nm} change when stoichiometry changes?



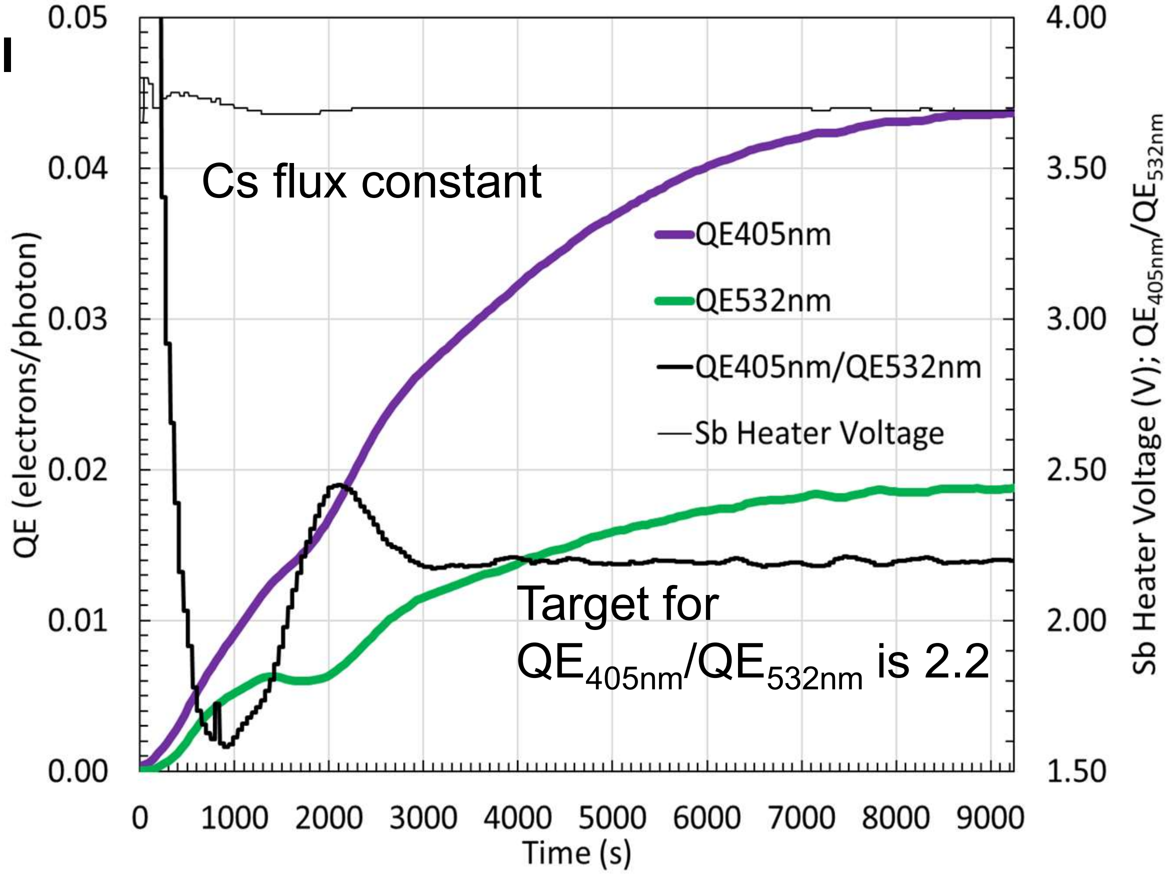
Data from:
L. Cultrera *et al.*, *Applied Physics Letters* **99**, 152110 (2011)

Process control: stabilizing thickness-independent photoemission parameter

Feedback loop is an essential part of process control.

PREREQUISITES:

- Reasonably stable calibrated sources
- Cs-rich growth mode
- Software PID feedback loop with pre-determined gains (Ziegler-Nichols method) or other properly tuned algorithm



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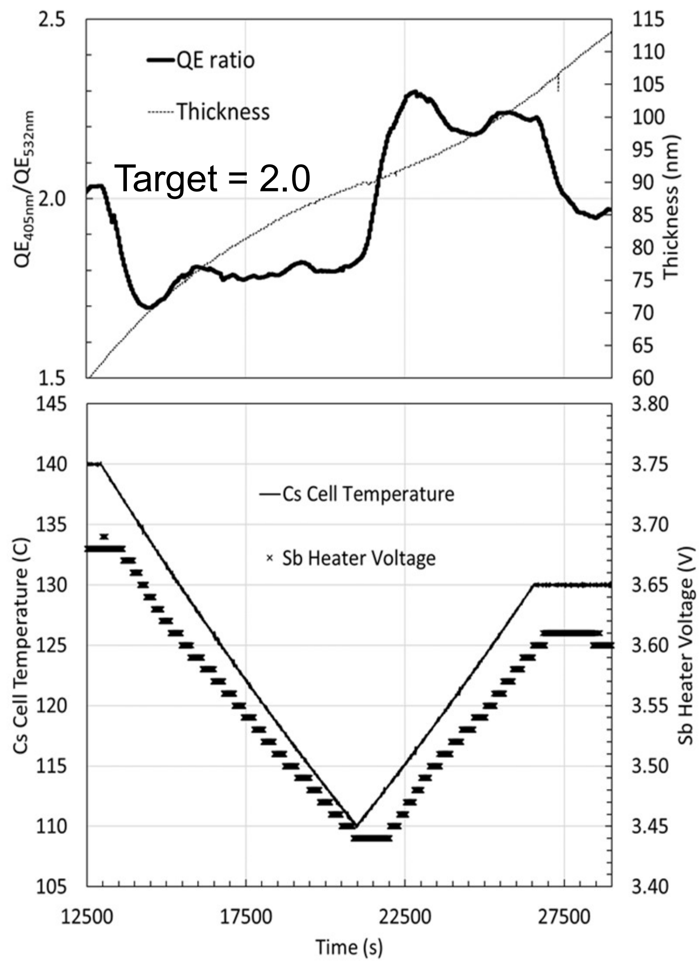
Process control: stress test

Sb flux
"chasing"
Cs flux



- Sb flux keeps up with Cs flux!
  (Sb flux "knows" nothing about Cs flux value or derivative, it is merely a function of film's photoemission)
- Time lag is about 500 s, including significant instrumental factor
- Ratio of fluxes and stoichiometry (both inferred) are maintained with a few % precision
- Max/min growth rate here is about 2.5, practically achievable range much larger
- Excellent tool for more accurate estimates of starting fluxes

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Thin film growth recipes that we can share

- **Recipes for photoemissive materials**
  - Substrate temperature (low, difficult to calibrate but solvable)
  - Growth rate (proxy such as Sb flux), including **variable**
  - Stoichiometric offset(s) based on photoemission

Calibration uncertainties do not matter too much.

Accurate QE measurements do not require cross-calibration between the labs.

Future Plans: Technology Maturation and Transfer

LANL ACERT APPLIED CATHODE ENHANCEMENT AND ROBUSTNESS TECHNOLOGIES automated photocathode growth technology

Radiation Monitoring Devices, Inc. (RMD)

RMD A Dynasil Company

Sputter deposition tech & targets

$2K_3Sb + Cs_3Sb \rightarrow 3K_2CsSb$

euclid BEAMLABS

Thank you for your attention!

Questions?