Automated growth of photocathode films: from the basics of process control towards artificial intelligence

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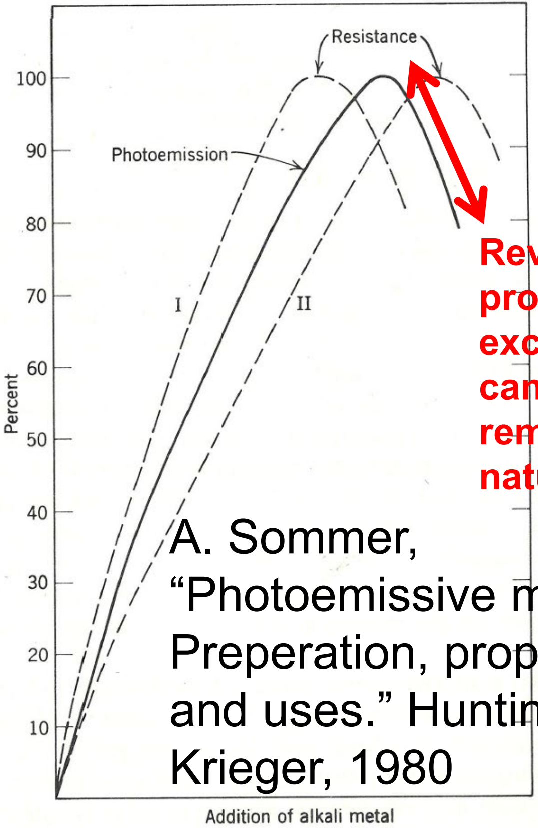
Outline

- Alkaline antimonide stoichiometry and QE
- Choice of optimization parameter for process control
- Thin film growth control for photoemissive materials
- Future plans
- Conclusions

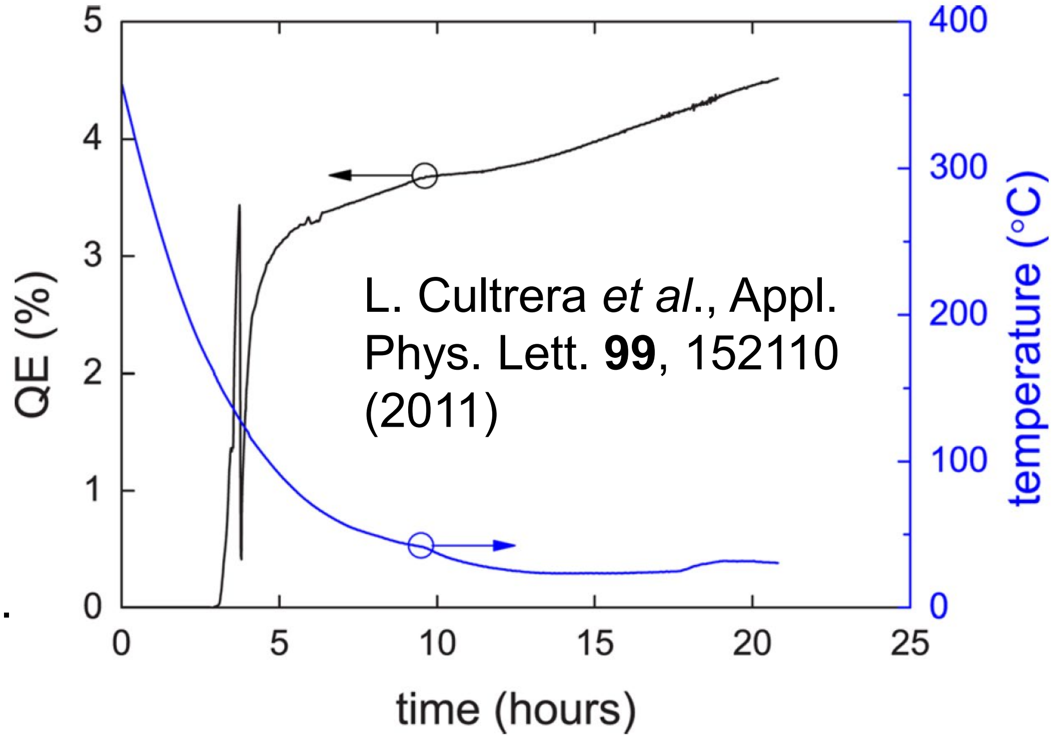
QE vs stoichiometry



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 = Target Compound

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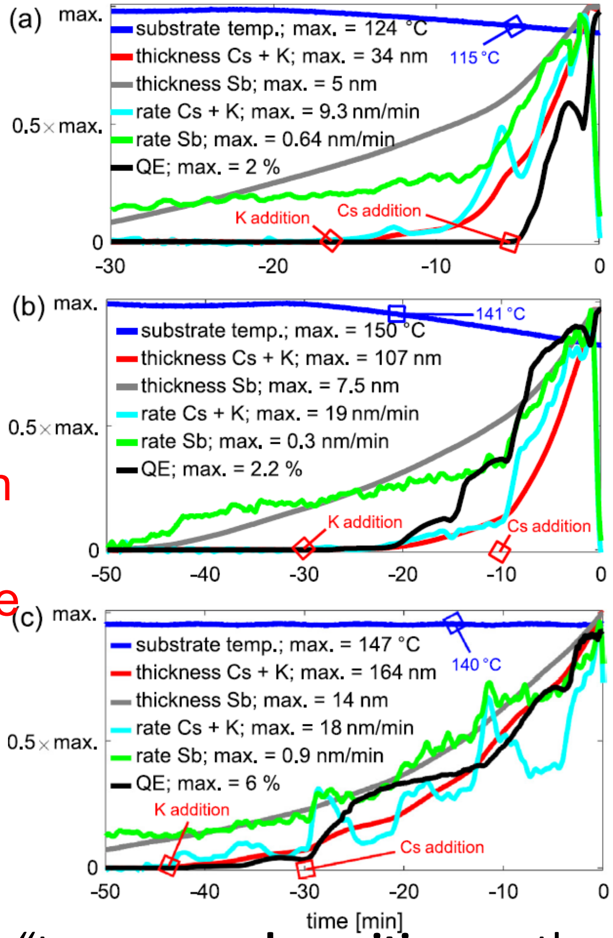
Optimizing co-deposition: can we use QE maximization?

Stated goal vs Reality

"slope of the *in situ* photocurrent as the driver for the growth process"

Long chain of adjustments and guesses: too much or not enough of this and that?.. The film, consequently, grows with large "swings" in stoichiometry.



"ternary **co-deposition** method"

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

+CATCH: QE vs time slope is the highest for sequential deposition.

If at all possible, QE slope maximization must be very difficult to implement…

TERMINOLOGY: what's the definition of co-deposition?

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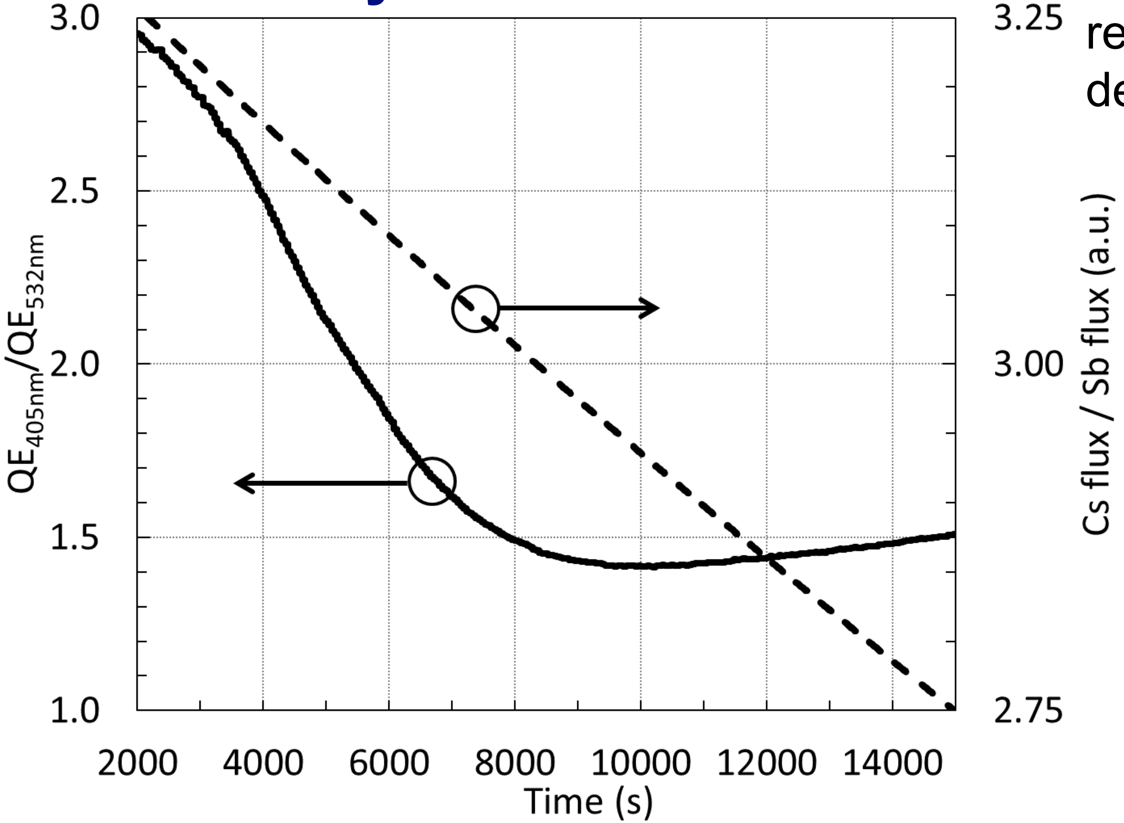
Thickness-independent photoemission parameter(s) vs stoichiometry



Corroboration: excess Cs removal experiments & thermal decomposition experiments

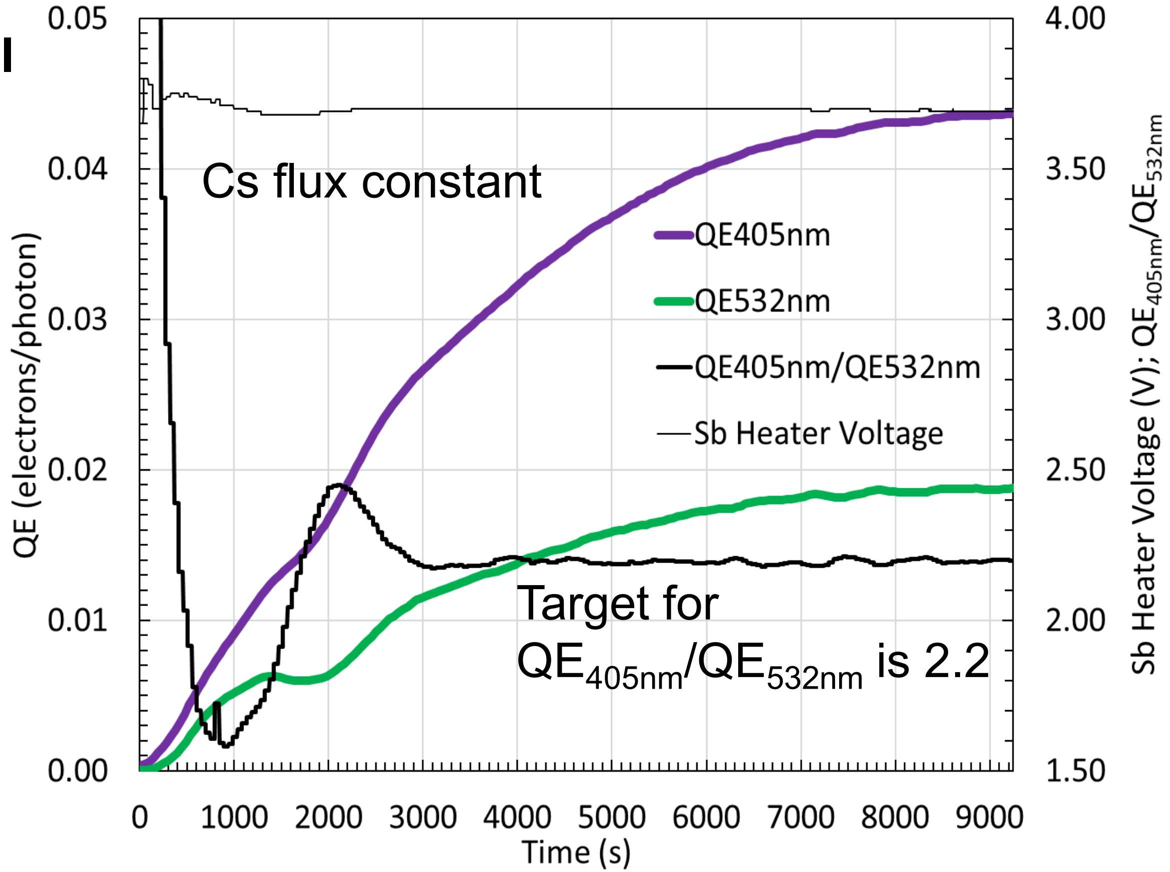
- Time axis effectively represents stoichiometry
- Cs/Sb atomic flux ratio close to 3 for low substrate temperature
- Minimum of QE_{405nm}/QE_{532nm} approximately coincides with QE maximum
- To facilitate process control, need to remain is Cs-rich growth mode, otherwise need ***elaborate*** *extremum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

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

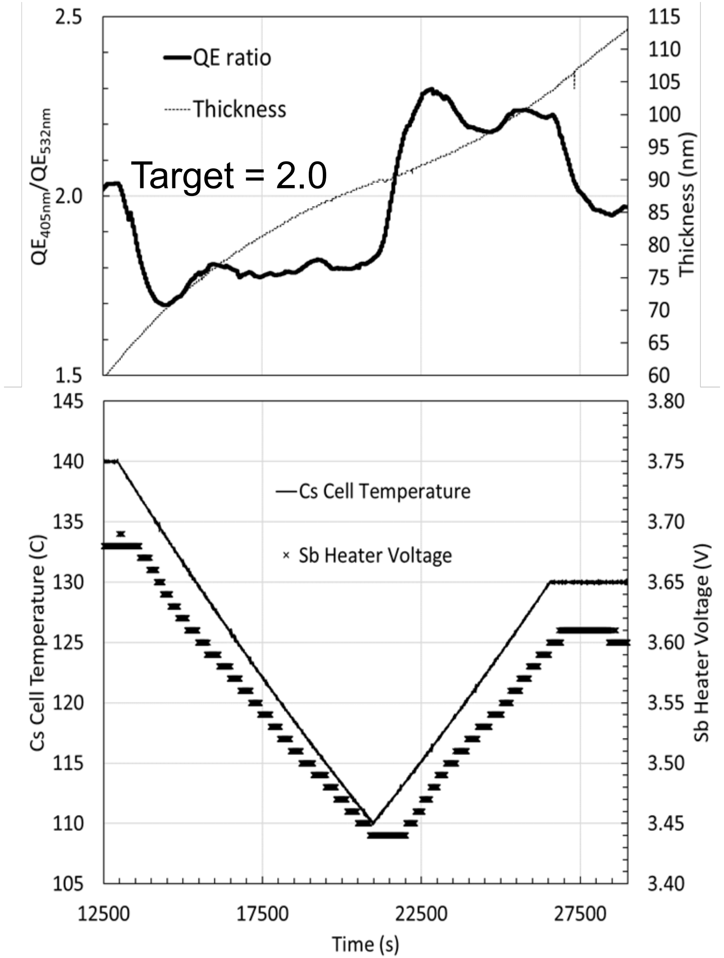


Process control: intermediate conclusions

- Yes, it is possible to grow a uniform film – automatically
- Very slow growth rates can be used
- Different algorithms can used – we verified by seeking (and finding) extremum of ABS(QE_{405nm}/QE_{532nm} – Target)
- Deviations from stoichiometry are apparent, due to experimental artifacts, not optimal algorithm type or tuning, “inaccurate” estimates for starting fluxes – all of those issues can (and should be) be mitigated
- Necessary flux adjustments are of a few % scale
- It should be possible to accomplish uniform growth with ultra-stable constant fluxes, but how one would find the values for those fluxes manually within reasonable amount of time?
- OK, you managed to grow a uniform film manually after numerous cycles of trial and error, now can you match this (next slide)?..

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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New type of film growth recipes

- **Traditional recipes (e.g., MBE)**:
  - Substrate temperature (usually high, 500+ degrees C, easy to calibrate)
  - Flux A
  - Flux B
  - (Flux C)

+TWEAKING of FLUXES

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

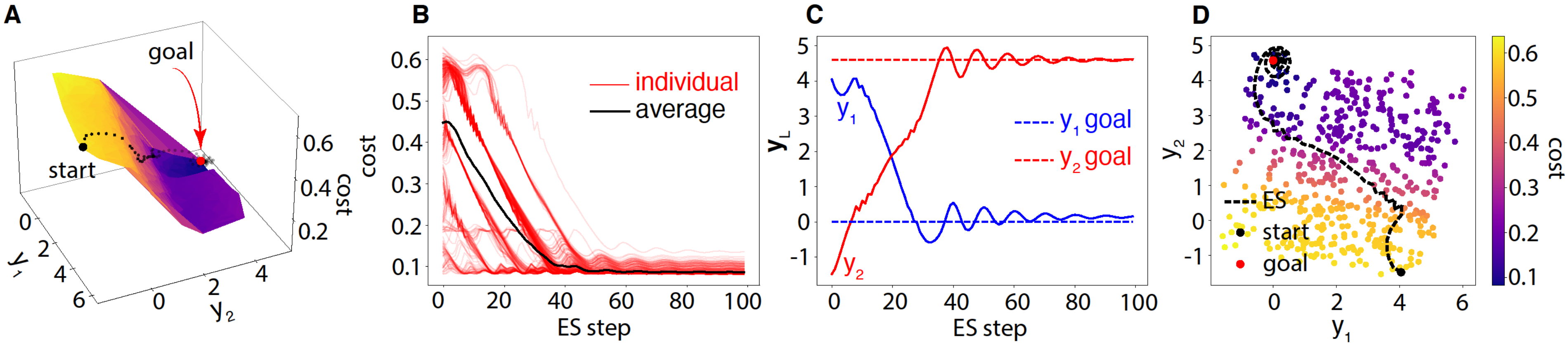
NO TWEAKING of FLUXES NECESSARY

We need to solve this problem

Technology status and costs

- Exists within LANL ACERT lab
- MBE-like deposition system + extra features for alkali metals & photoemission
- Highly flexible & modular integrated software-hardware system
- Costs in addition to MBE system are small
- Functional in the lab environment but not ready for deployment; critical components are not available commercially (custom-built)
- Development continues…

Future development: what about AI?



A convolutional neural network (CNN) is used to approximate an analytically unknown cost function surface as shown in (**A**). Model-independent adaptive feedback can then be used to traverse the approximated surface to get a guess for the correct initial parameter conditions, which can then be used for feedback-based optimization of the actual unknown system. Cost function evolution is shown for 100 random initial conditions (**B**). A single example trajectory of the two parameters y1 and y2 is shown in (**C**) and is superimposed on a top-down view of the surface in (**D**). The optimization trajectory is also shown as it evolves on the surface in (**A**).

A. Scheinker. "Adaptive machine learning for time-varying systems: low dimensional latent space tuning." *Journal of Instrumentation* 16.10 (2021): P10008.
https://doi.org/10.1088/1748-0221/16/10/P10008

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Future development: collaboration

We would like to…

Give

out/share the technology, at least its current generation. And start exchanging the recipes.

Provide platform for custom model-validating experiments.

Get

predictions from the theory/modeling perspective of how small deviations from the optimal stoichiometry affect photoemissive properties, especially for the case of bi-/multi-alkali antimonides.

Conclusions

- Automated growth of Cs_3Sb demostrated, based on real-time stoichiometry control method
- Modest technology costs
- Future efforts: technology maturation and extension onto Cs_2Te and bi-/multi-alkali antimonides