State-of-the-Art Photocathodes and New Developments

Applying the tools of modern material science to cathode design

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Free Electron Laser Conference (FEL2019)

University of Hamburg

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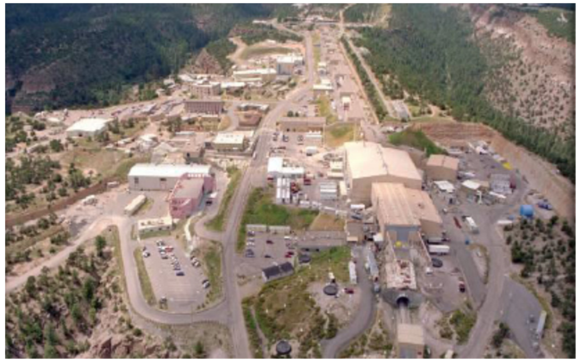
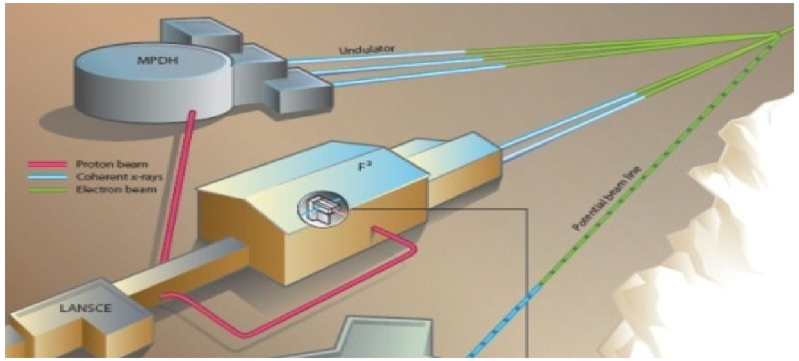
Talk Outline

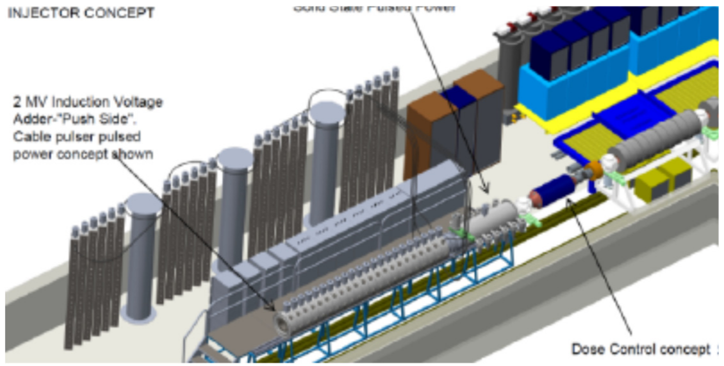
- **Introduction**
- **Motivation for continued progress**
- **Overview of the approach**
  - Improved synthesis techniques
  - *In operando* x-ray analysis
  - Reduction to practice
  - Target compatibility with working injectors
  - Extend photo-physics capability to other applications
- **Examples of recent results**
- **Summary**

Why does Los Alamos care about electron source design?

- **Accelerator R&D is crucial for delivering LANL mission AND preparing for tomorrow:**
  - LANL anticipates an advanced dynamic mesoscale material science capability and advanced cathodes are an enabling technology
  - Closes technology gaps to enable accelerator upgrades to other LANL accelerators
  - Important for recruitment/retention of leaders in the accelerator field
  - National / international stature translates to sponsor confidence
- **Investments in electron source design are consistent with being both a <u>material science laboratory as well as NNSA's primary accelerator capability</u>**







Alignment between BNL and LANL affords new opportunities, leveraging of programs, long-term sustainability, and equipping of future leaders

N. Moody; J. Smedley; K. Jensen; H. Yamaguchi; F. Liu; J. DeFazio; M. Gaowei; C.W. Narvaez Villarrubia; J. Xie; J. Sinsheimer; D. Strom; V. Pavlenko; A. Mohite; E. Batista; S. Lambrakos; A. Shabaev; M. Hoffbauer; J. Pietryga; I. Robel; S. Schubert; J. Wong; J. Feng; Siddharth Karkare; H. Padmore; J. Power; M. Conde; E. Muller; Z. Ding; K. Attenkofer; X. Liang; J. Xie; and J. Kühn

Ongoing Goal: understand and exploit correlations between material science and cathode performance to gain control parameters for designer cathodes

Material Science Design Choices

- Substrate choice
- Substrate orientation
- Synthesis methods
- Growth rate
- Duration of growth
- Substrate temperature
- Coating composition
- Coating thickness
- Surface termination

Material Science Observables

- Real-time stoichiometry
- Real-time thickness
- Roughness
- Texturing
- Grain boundaries
- Phase purity
- Heterostructure
- Reactivity
- Photoconductivity
- Optical transmission

Electron Source Design Choices

- Applied field gradient
- Drive laser wavelength
- Laser time / spatial structure
- Ambient environment (gases, etc.)
- Operating temperature

Electron Source Observables

- Quantum efficiency
- MTE
- Response time
- Dark current
- Lifetime
- “Quality” parameter or cost function

Linked by physics models and x-ray tools such as XRR, XRF, XRD, XPS

LANL hosted the P3 2018 workshop

https://indico.cern.ch/event/759878/

Photocathode Physics for Photoinjectors (P3)

October 15-17, 2018

Santa Fe, New Mexico

Where are the frontier applications in photoemission sources?

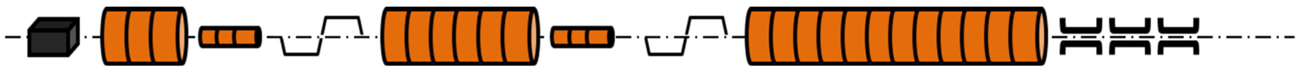
- **XFELs (LCLS-II, MaRIE-class)**
  - moderate currents (still under 1 mA);
  - emittance improvement is a limiting/enabling technology (ideally ~0.1 μm)
  - the electron source will determine the beam properties

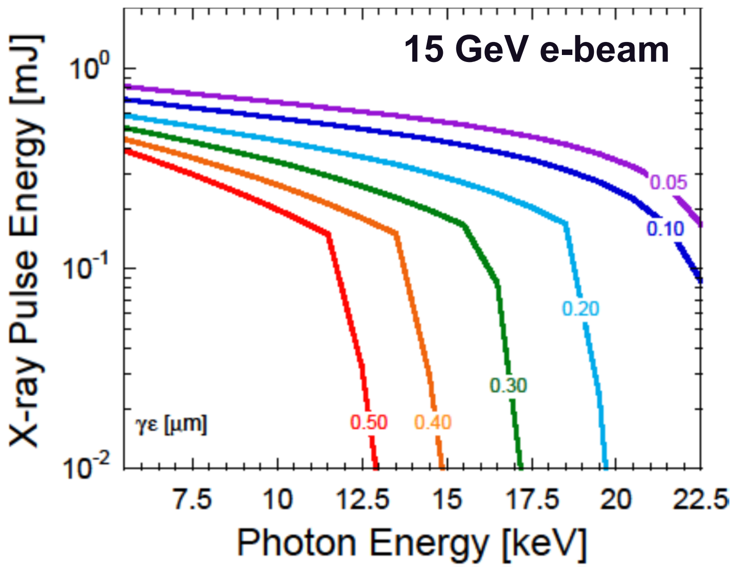
- **Ultrafast Electron Diffraction / Microscopy**
  - High brightness! Ideally a factor of 100 from current photoinjectors. Very low current. Short pulse duration (100 fs at sample, less for some applications)

- **Electron cooling of ion machines**
  - Requires high current with long operational life, other requirements are modest (~50 mA with 5μm emittance)

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

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





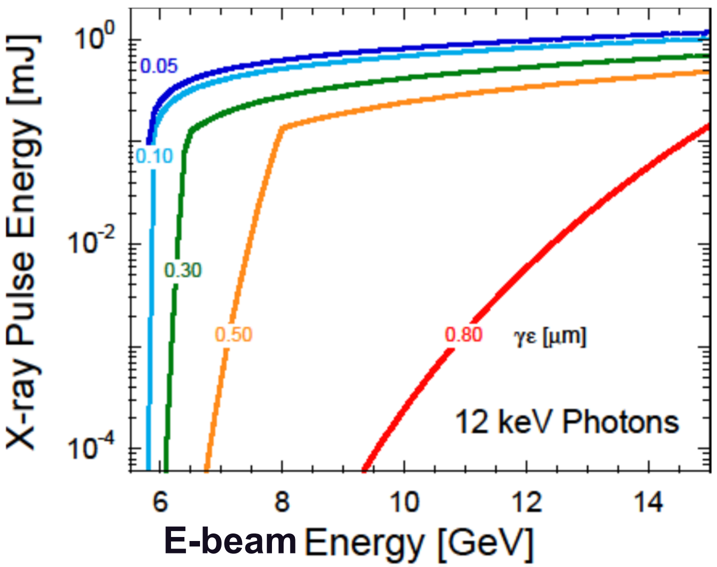


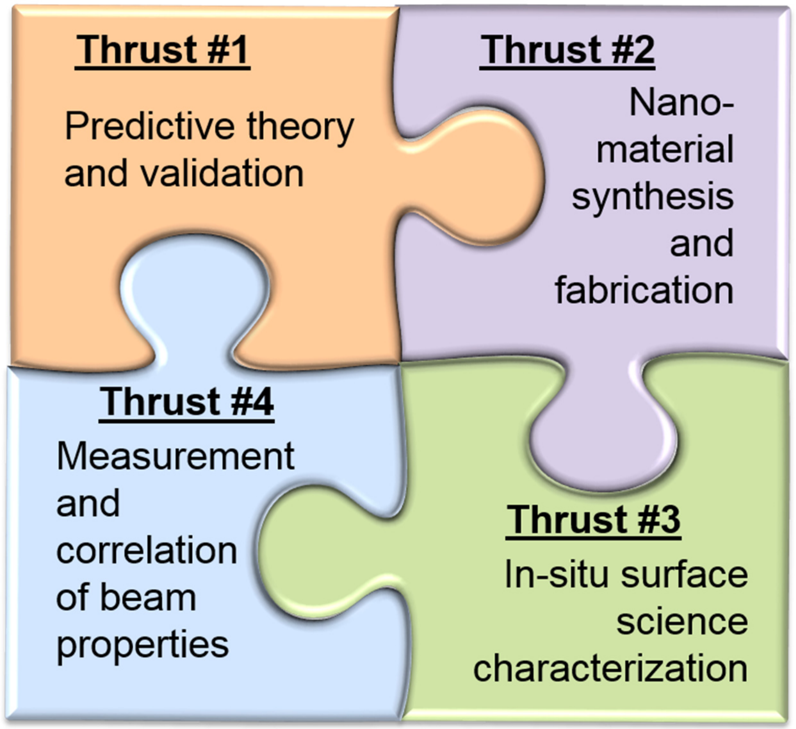
TABLE II. LCLS-II/HXR Case Study Parameters

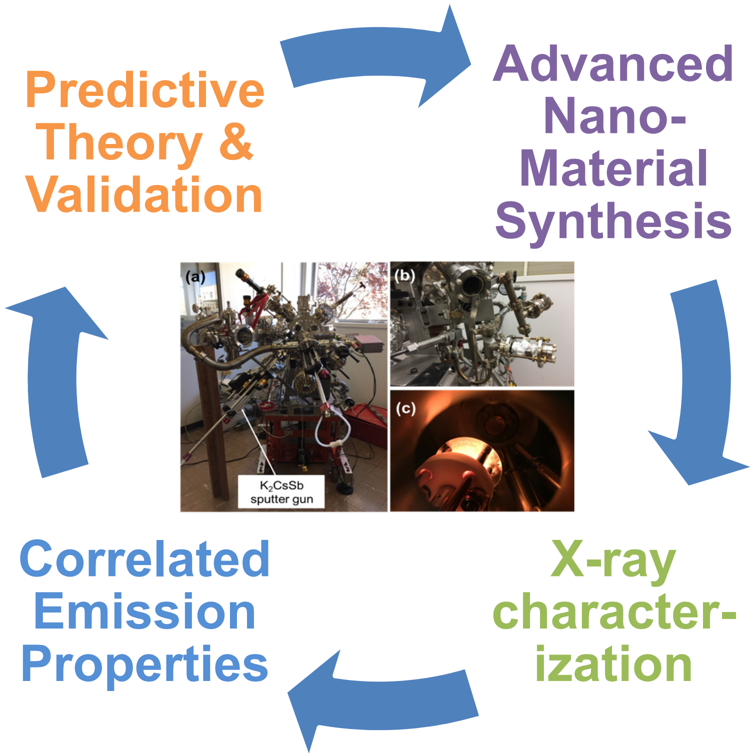
Parameter	Definition	Value
E_b	Beam Energy	15 GeV
σ_η	Energy Spread	1.5 MeV
L_u	Undulator Length	140m
λ_u	Undulator Period	26mm
I_{peak}	Peak Current	3.5 kA
β	Mean Beta	30 m
Q_b	Bunch Charge	100 pC

- ***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.

LANL’s approach consists of four pillars and advances the TRL of new techniques for specific needs and applications





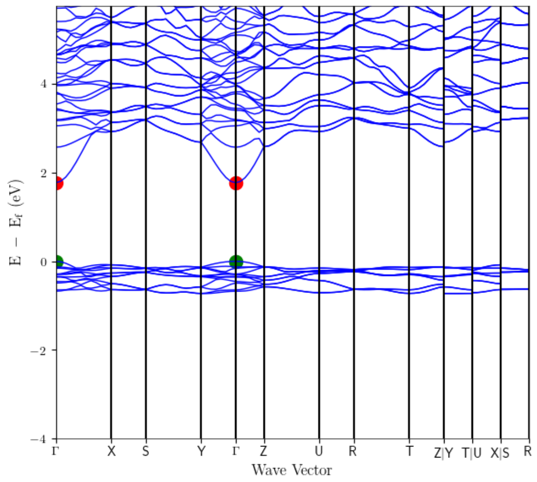
Research		Development		Demonstration	Production
Basic	Applied	Exploratory	Advanced		

Technology Readiness Levels (1-9)

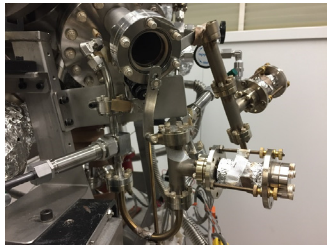
1	2	3	4	5	6	7	8	9
Basic Principles	Technology Concept	Proof of Concept	Component Validation	Field Validation	Prototype Demo.	System Demo.	System Complete	Product in use

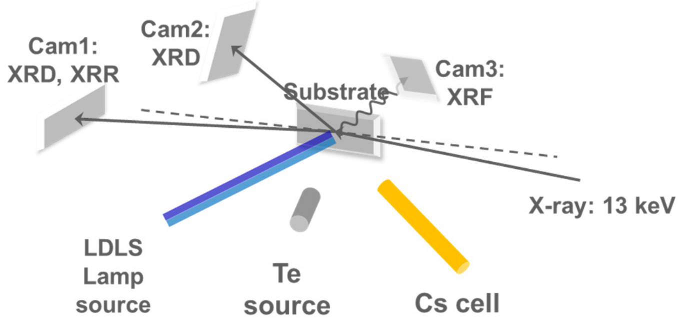
Goal is to develop and perfect the technologies using x-ray characterization and transition it to groups without such tools.

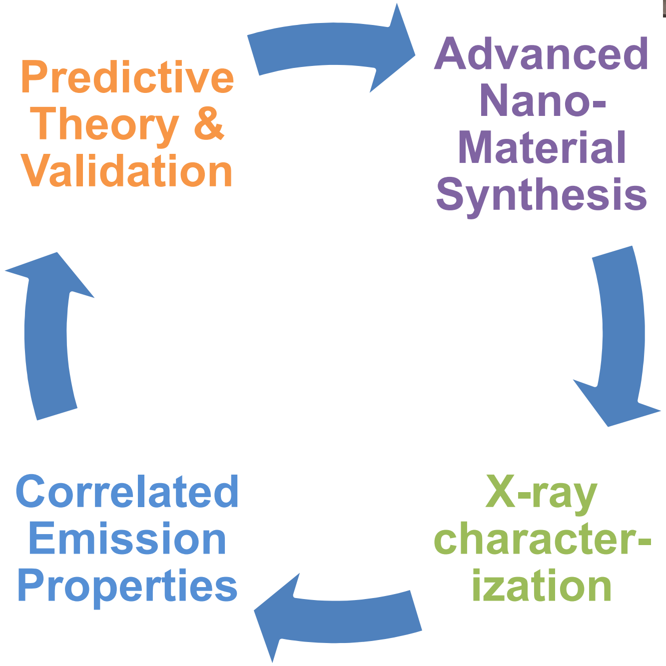
Example of success in one study being applied to adjacent problem: studies of K_2CsSb enabled growth of Cs2Te

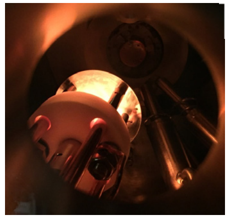


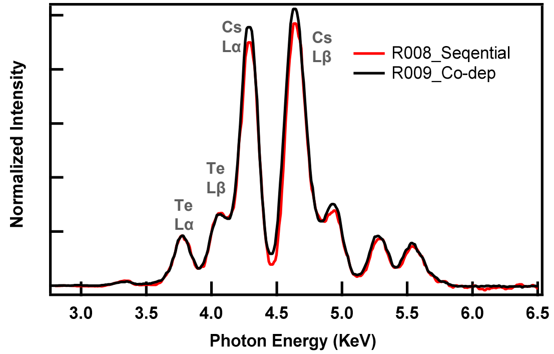
HKL	Theory_d spacing	Exp_d spacing
222 Cs_2Te	2.307	2.315
111 Cs_2Te	4.613	4.615

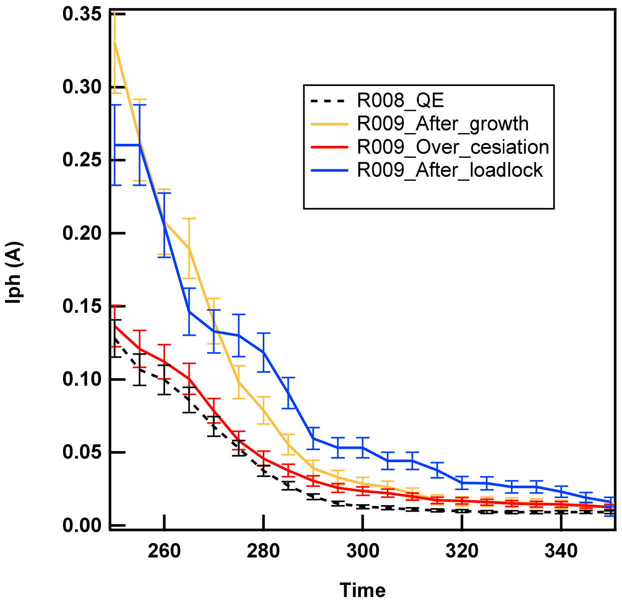




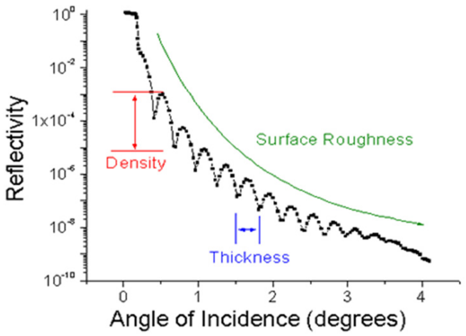


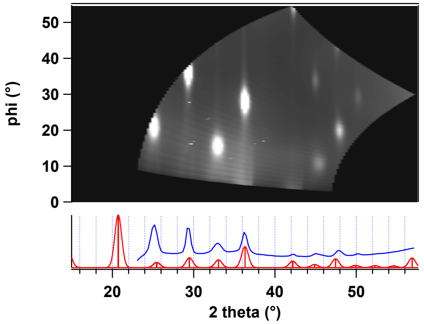






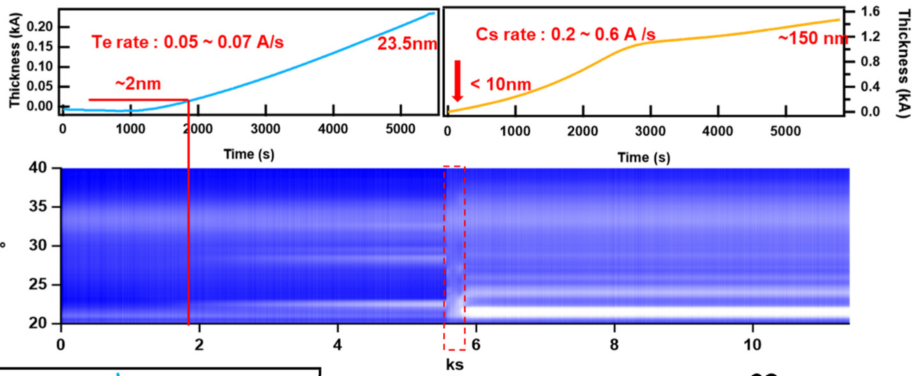
Next step: transfer to procedure not requiring synchrotron light source

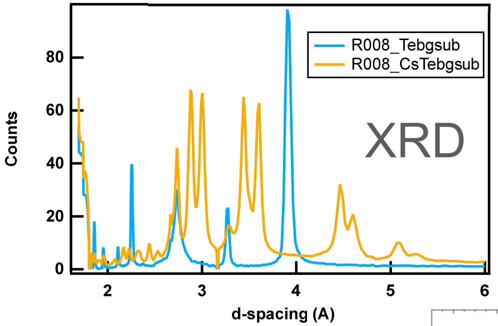


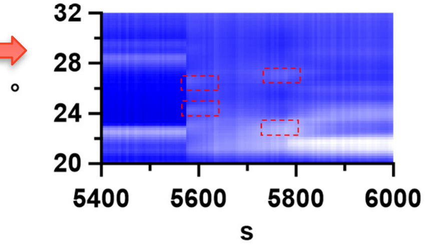


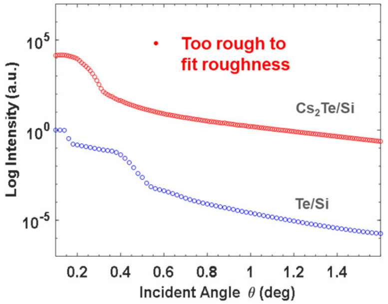
Cs-Te Results (presented by Mengjia Gaowei at P3-2018) show the efficacy of real-time characterization: co-deposition gives smooth single phase

Sequential growth

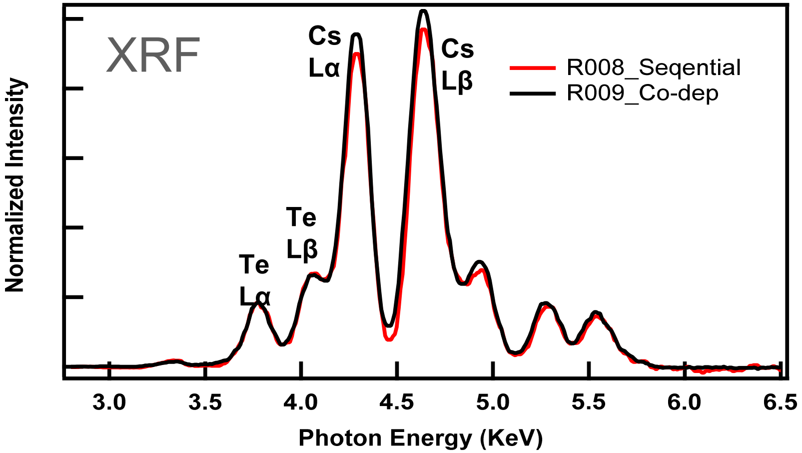




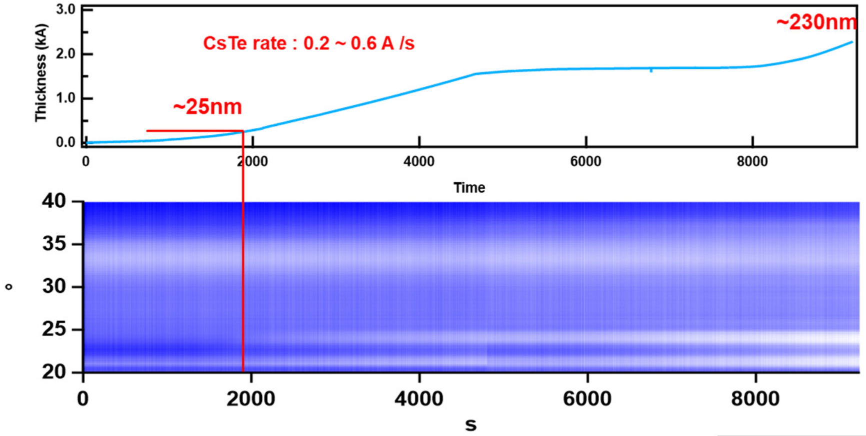


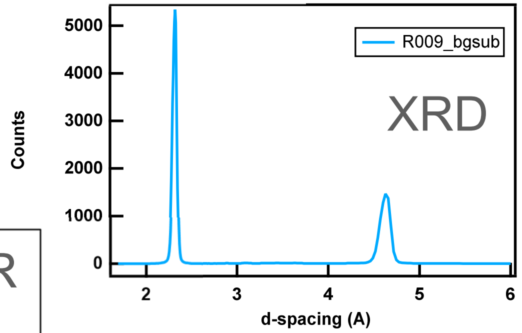


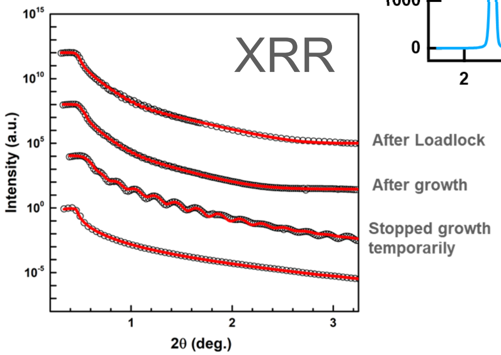
- Nearly all of the crystalized Te has dissolved
- Low counts in diffraction peaks
- Multiple phase of Cs-Te compound co-exist



Co-deposition growth



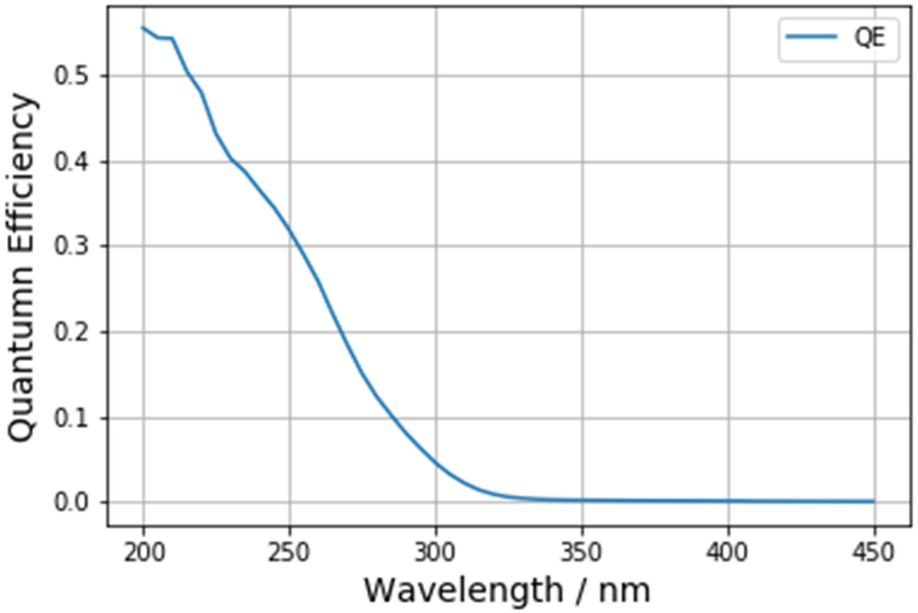


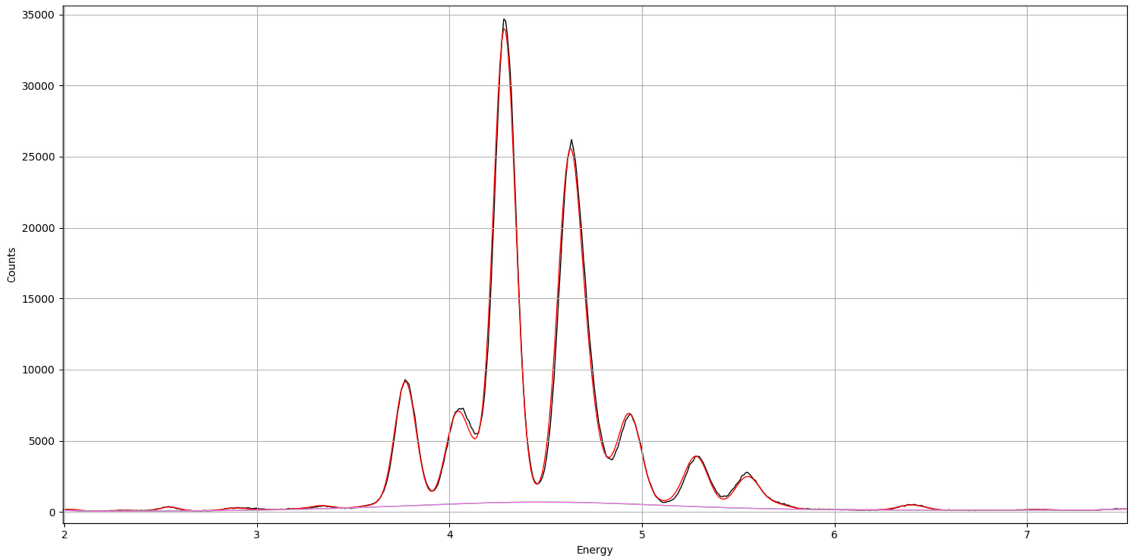


- Well crystalized
- Single phase, smooth

Real-time supervision of Cs-Te growth: record QE and perfect stoichiometry

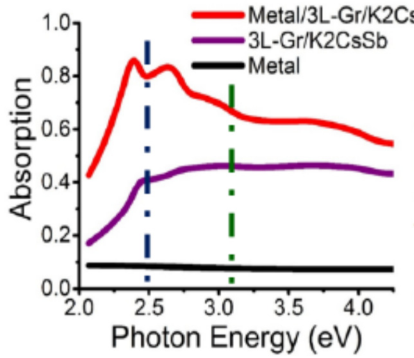
CsTe	Te	Cs
Q007	1	1.98



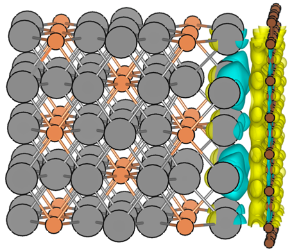


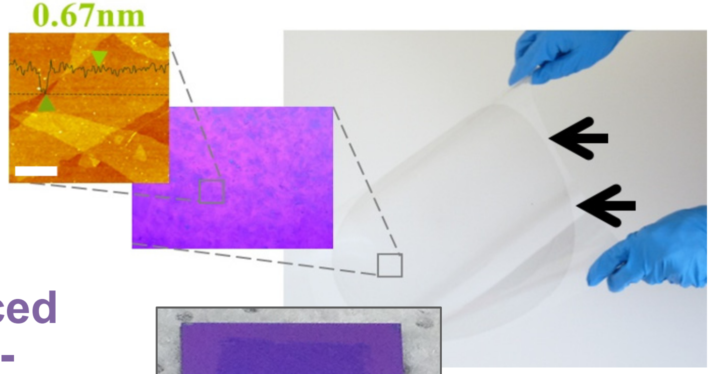
M. Gaowei, J. Sinsheimer, D. Strom, J. Xie, J. Cen, J. Walsh, E. Muller, and J. Smedley
Phys. Rev. Accel. Beams 22, 073401

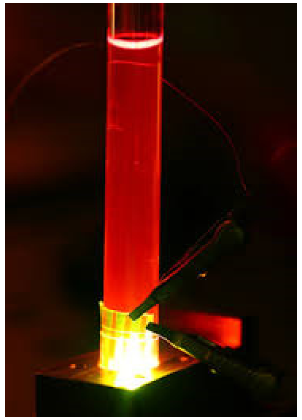
Progress toward cathode integration with 2D coatings for protection and enhancement



NKD Gen software (University of Barcelona) based on conventional transfer matrix methodology was used.





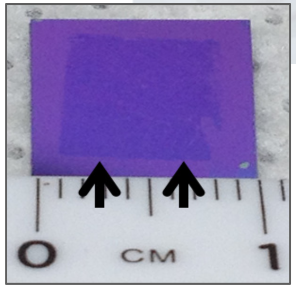


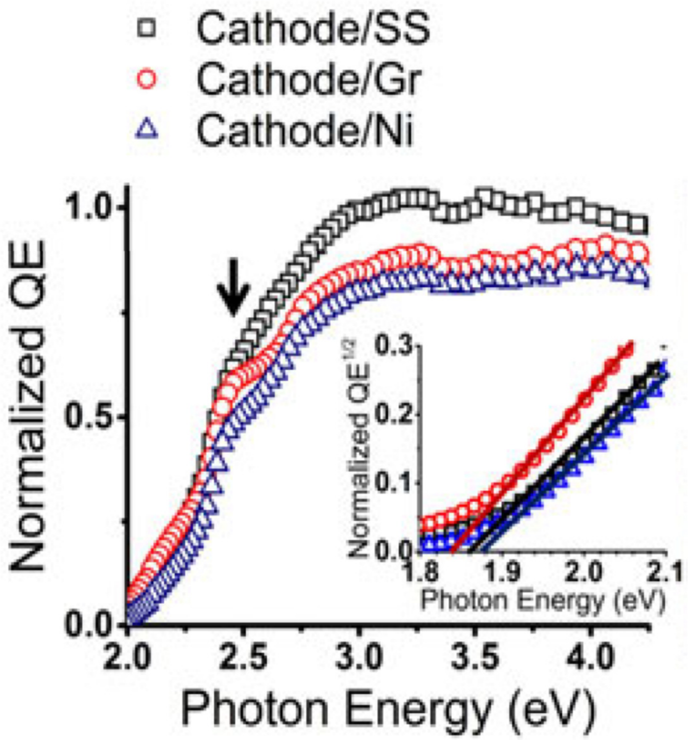
Predictive Theory & Validation

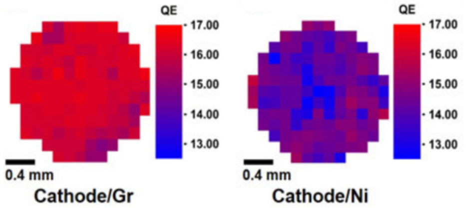
Advanced Nano-Material Synthesis

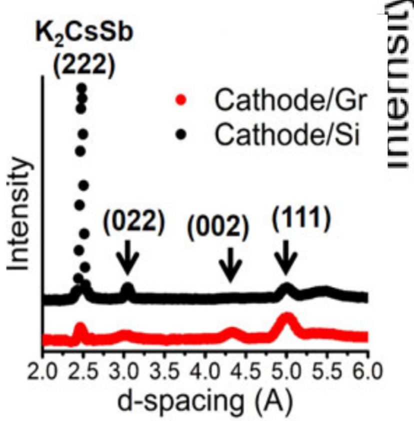
X-ray character-ization

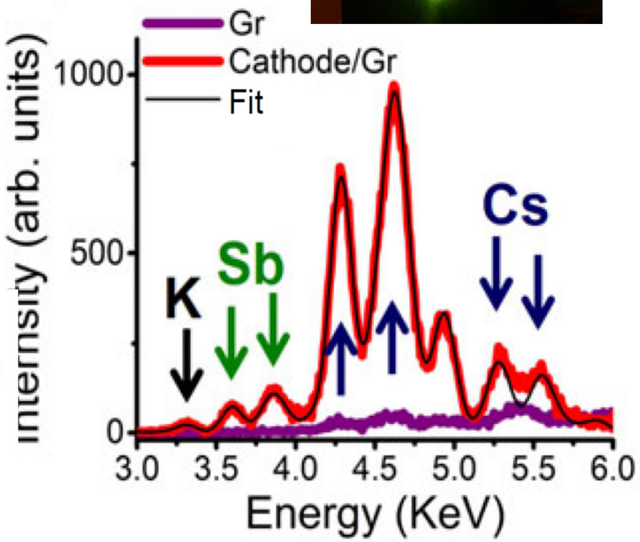
Correlated Emission Properties



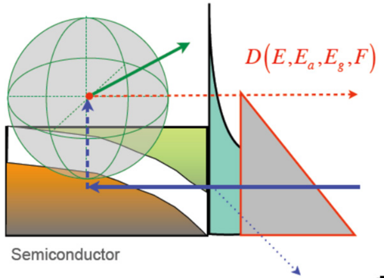


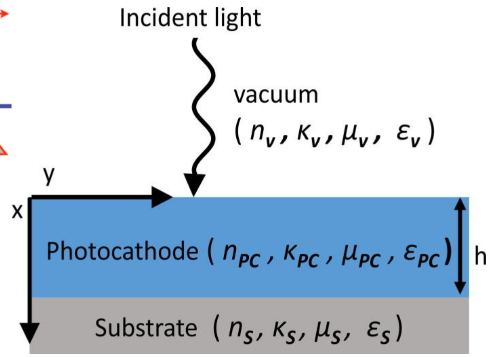






Development of ‘Etalon’ (wave interference) thin film cathodes allows for design of fast response cathode with reasonable QE



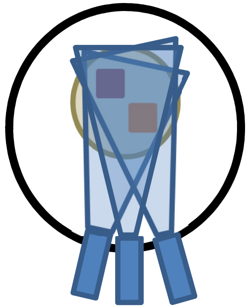


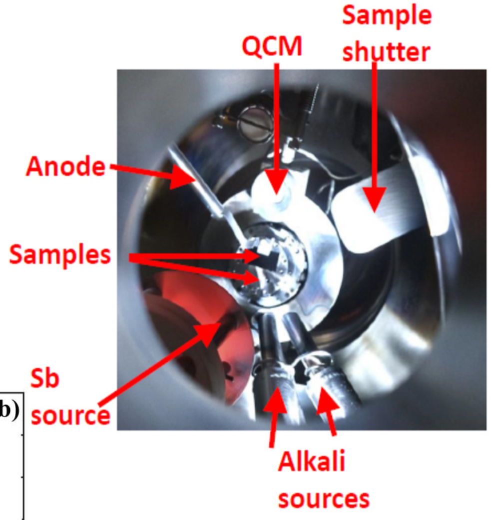
$$a(x) = \frac{P_a(x)}{P_{in}}$$

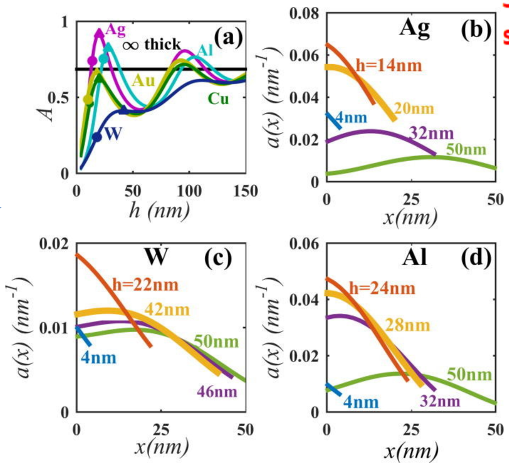
$$= -\left(\frac{\kappa_{PC}\, k\tilde{n}_{PC}}{\mu_o \mu_{PC}}\right) |E_o(1 + r_{V-PC})|^2 e^{-x/\lambda_{opt}}$$

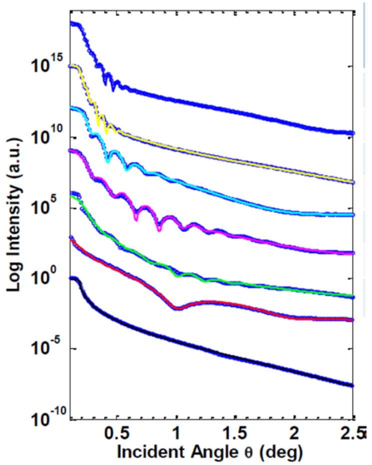
Predictive Theory & Validation

Advanced Nano-Material Synthesis

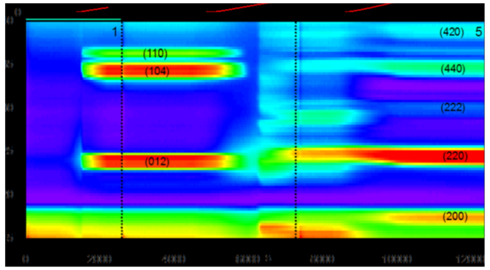




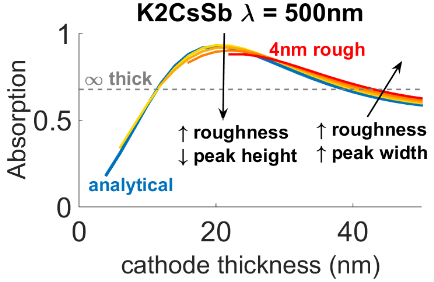




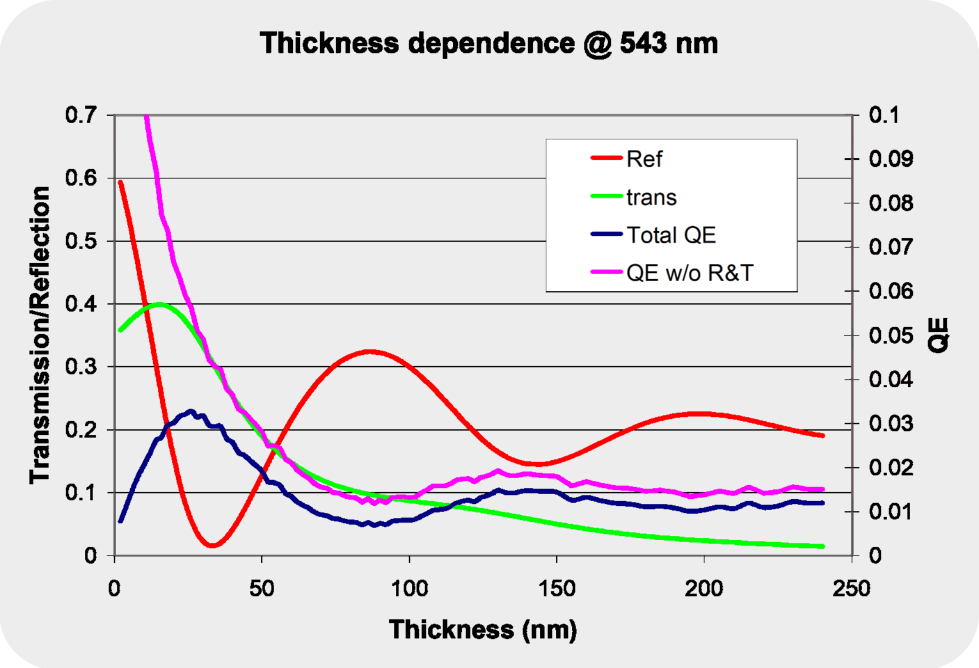
X-ray characterization

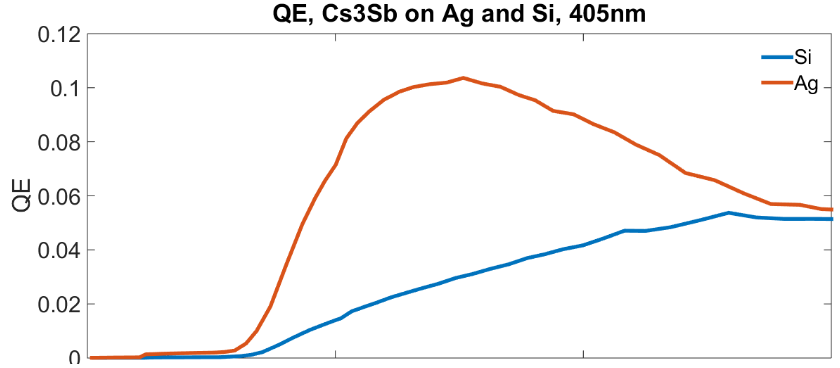


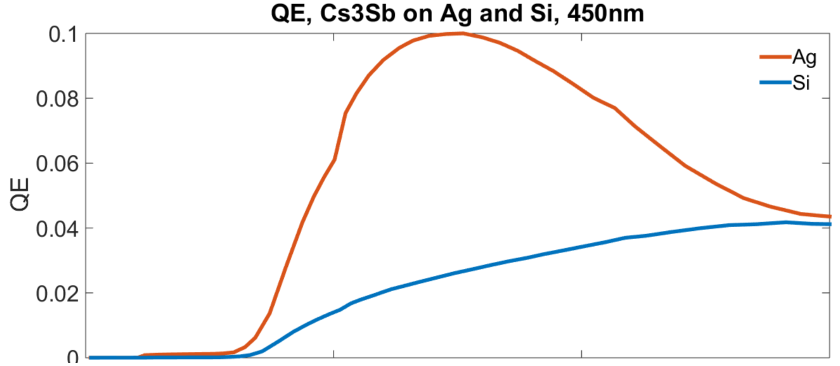
Correlated Emission Properties

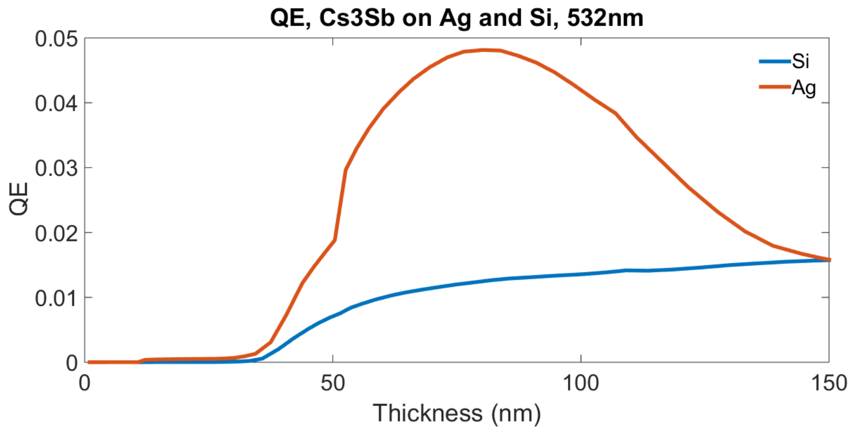


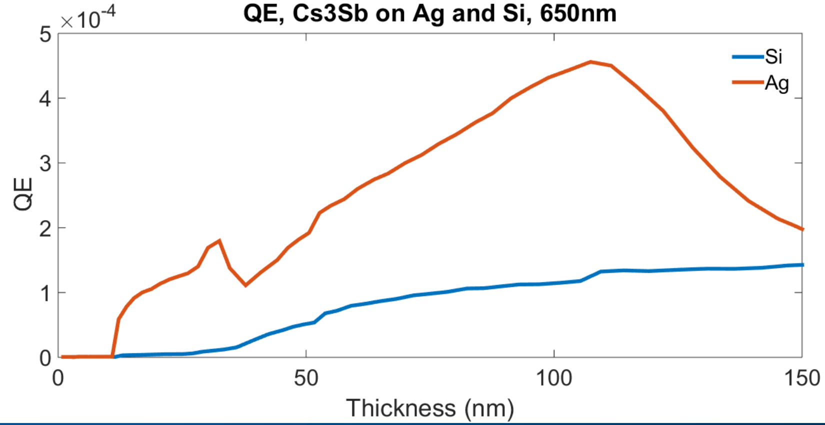
Etalon effect predicts QE enhancement in Cs3Sb and experiments show this at various combinations of substrate, thickness, wavelength











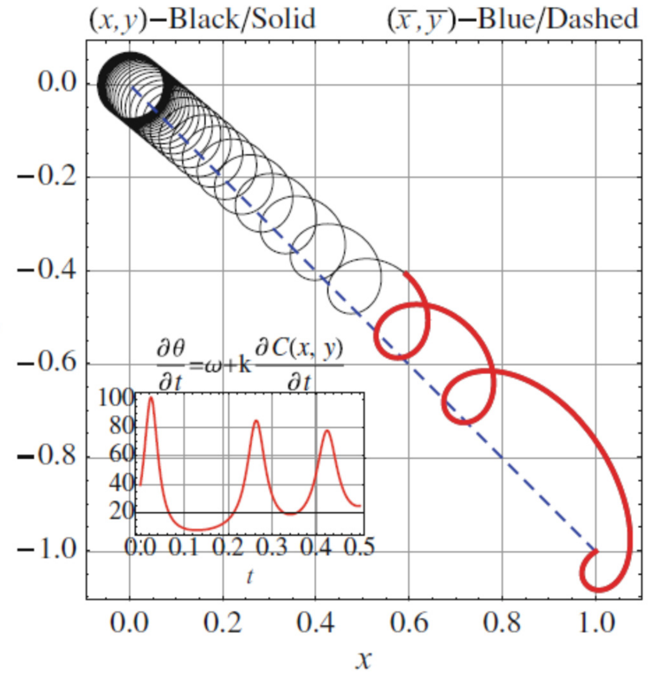
Towards automated growth: control and supervision of material growth without a synchrotron (at least not every time)

- **Goal:**
  - Automatically identify ideal growth conditions
  - Maintain ideal growth in dynamic conditions where “correct” parameters may shift with time

- **Algorithm: extremum seeking (ES)**
  - Applies sinusoidal variation to inputs
  - Minimizes cost parameter with (multiple) inputs
  - Works by spending more time near minimum than at higher values

- **ES algorithm:**
  - $\frac{dx}{dt} = \alpha\omega \cdot \sin(\omega t + kC)$
  - X: input value
  - α, k, ω are tuning parameters
  - C is the function to be minimized

- **Next step: characterize the material using x-ray toolset**



Starting point

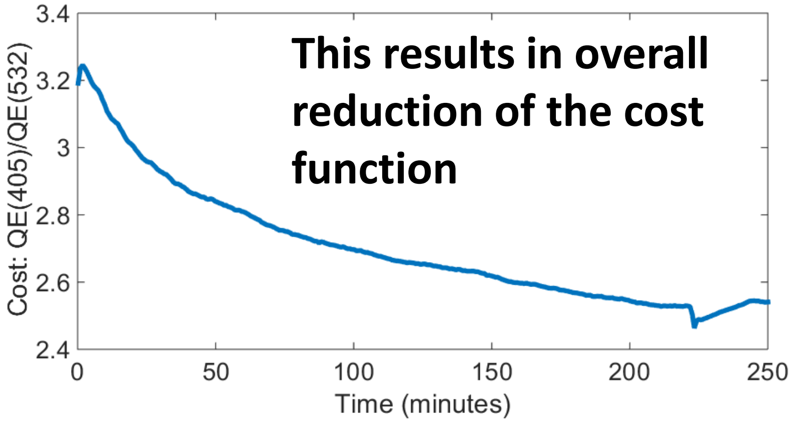
Example of ES for motion where

$$\frac{d\theta}{dt} = \omega + \frac{dC}{dt}$$

$$C = x^2 + y^2$$

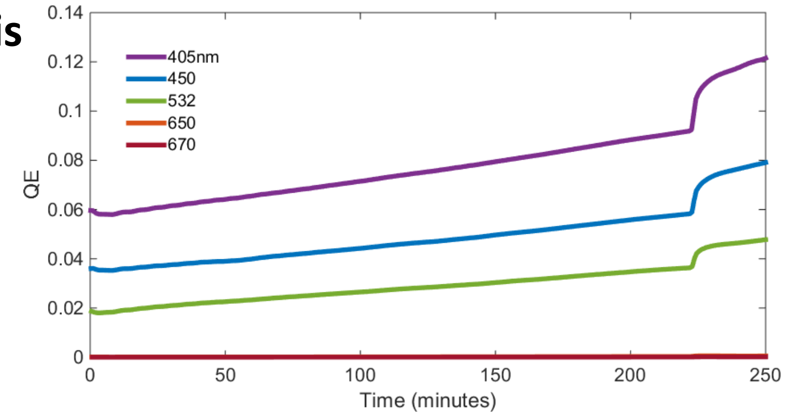
First results using ES growth algorithm are encouraging: Cs_3Sb with max QE before/after cooling 9% / 15%

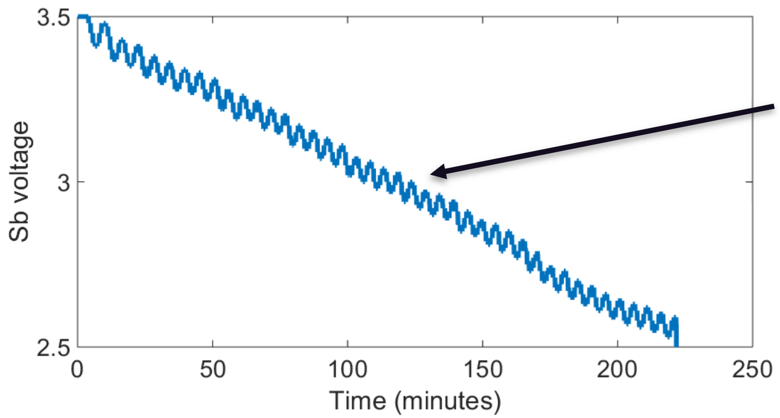
- **Growth conditions:**
  - Sample temperature: 125° C
  - Cs temperature: 180 ° C
  - Initial Sb flux ~ .01 A/s
- **Parameter optimized: Sb flux/voltage**
- **Cost function minimized: QE(405)/QE(532)**
- **Next steps:**
  - More materials: K2CsSb, CsTe
  - More parameters optimized
  - Better performance/stability for thin cathodes



This results in overall reduction of the cost function

If the cost function is chosen correctly, this increases the QE



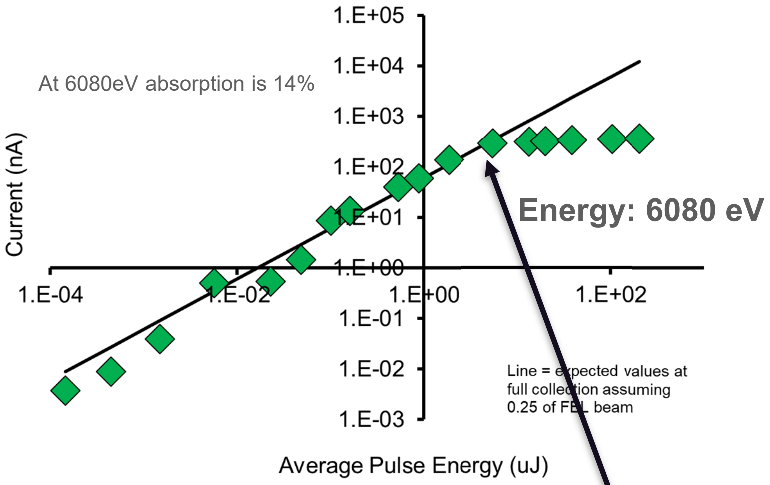


Algorithm applies sinusoidal variation to input, spending more time where cost function is lower

Expanding the scope of photophysics beyond photoemission: x-ray detector development via BNL/LANL collaboration

- **Goal: to achieve linearity over full gain curve (10^4 – 10^{11} ph/pulse)**
  - Improved beam targeting (< beam size)
  - Diagnostics for future FELs (DMMSC) enabling full gain curve optimization
  - Beam profiling: shot to shot profiles (flux, position, horizontal and vertical distribution)
- **So far: Modified detector with additional capacitance, achieves linearity up to 3% of full beam**
  - Detector performance unchanged by FEL beam (no damage)
  - Limit caused by geometry and charge pileup – we have engineering solutions in mind

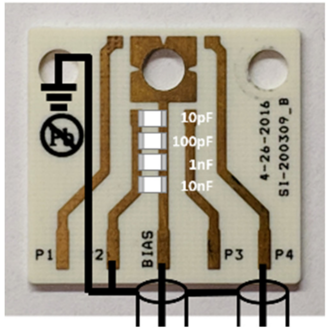
Flux Linearity (SwissFEL Bernina Measurements)



Response linear to ~3% of full beam

Looking for collaborations to continue this development and calibration effort

J. Bohon, P. Juranic, E. Muller, M. Zou, A. Sumant, U. Jastrow, K. Tiedtke, T. Tanaka, S. Owada, R. Follath, C. Arrell, Y. Arbelo, M. Degenhardt, A. Sorokin, J. Smedley



Circuit board design

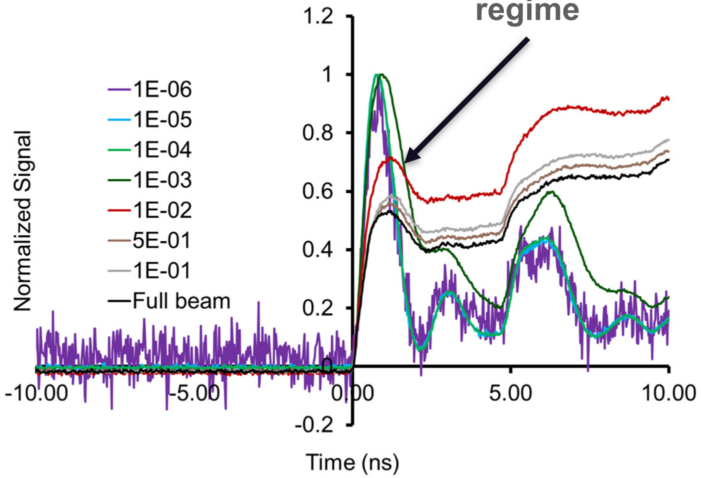
External case (similar)

Diamond Properties

Low X-ray Absorption
High Thermal Conductivity
Mechanical Strength
Radiation Hardness
Indirect bandgap (no radiative recombination)
Fast - carrier mobility 4500/3800 $cm^2V^{-1}s^{-1}$ (e/h)

This result is the first demonstration of synchrotron detectors applied to FEL applications.

Individual pulse measurement, pulse broadening beyond linear regime



The ACERT collaboration published recent advances in many aspects of electron source design

- **Time-dependent x-ray analysis of photocathode growth**
  - Schubert, S., et al., *Bi-alkali antimonide photocathode growth: An X-ray diffraction study*. Journal of Applied Physics, 2016. **120**(3): p. 035303
  - Ding, Z., et al., *In-situ synchrotron x-ray characterization of K2CsSb photocathode grown by ternary co-evaporation*. Journal of Applied Physics, 2017. **121**(5): p. 055305.
  - Gaowei, M., et al., *Synthesis and x-ray characterization of sputtered bi-alkali antimonide photocathodes*. APL Materials, 2017. **5**(11): p. 116104.
- **Etalon wave interference effects: ultra-fast photocathode and recovered QE for near-threshold emission**
  - Alexander, A., N.A. Moody, and P.R. Bandaru, *Enhanced photocathode performance through optimization of film thickness and substrate*. JVST-B, 2017. **35**(2): p. 022202
- **Tunneling effects of single-layer and multi-layer 2D barriers on a photoemitter**
  - Wang, G., N. Moody, et al., *Overcoming the quantum efficiency-lifetime tradeoff of photocathodes by coating with atomically thin two-dimensional nanomaterials*, Nature npj 2D Materials **2**(17), 2018.
- **Enhanced quantum efficiency using plasmons**
  - Alexander, A., N.A. Moody, and P.R. Bandaru, *Enhanced quantum efficiency of photoelectron emission, through surface textured metal electrodes*. Journal of Vacuum Science & Technology A, 2016. **34**(2): p. 021401.
- **Effect of quantum confinement on electron emission**
  - Makarov, N.S., et al., *Quantum Dot Thin-Films as Rugged, High-Performance Photocathodes*. Nano Letters, 2017. **17**(4): p. 2319-2327.
- **Lifetime enhancement using 2D films (hBN and graphene)**
  - Yamaguchi, H., et al., Active bialkali photocathodes on free-standing graphene substrates. npj 2D Materials and Applications, 2017. 1(1): p. 12.
  - Liu, F., et al., Single layer graphene protective gas barrier for copper photocathodes. Applied Physics Letters, 2017. 110(4): p. 041607.
- **First principles model of cathode degradation and new lifetime metric**
  - Wang, G., et al., Degradation of Alkali-Based Photocathodes from Exposure to Residual Gases: A First-Principles Study. The Journal of Physical Chemistry C, 2017. **121**(15): p. 8399-8408.
  - Pavlenko, V., et al., Kinetics of alkali-based photocathode degradation. AIP Advances, 2016. **6**(11): p. 115008

Summary of what we have learned across multiple collaboration efforts

- Real-time x-ray characterization tool which is capable of optimizing growth parameters for figures of merit other than Quantum Efficiency, and to specifically target material properties.
- 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 constituent metals
- Conformal coating of structured surfaces possible
- Real time XRF feedback provides option of ternary co-evaporation, producing best cathode
- Can consider ultra thin (under 10 nm) cathodes to improve response time but not suffer catastrophic loss in QE, thanks to Etalon optimization of substrate, cathode, thickness, wavelength
- Progress in surface coatings, some of which may enhance emission while protecting surface
- Hypothesized 'cost function' or quality parameter that may guide growth without synchrotron
- Progress toward automated growth with anticipated validation studies (XRF, XRD, XRR, etc.)
- Applications of photophysics extend well beyond photoemission (detectors, sensors, power sources, etc.)

Summary: Changes to the LANL / BNL photocathode programs are a strategic response to evolving needs and opportunities within NNSA

- **The Materials by Design strategy engages the broader material science communities at both LANL and BNL**
- **Progress has been non-linear with time and investment**
  - Techniques honed for one application can be rapidly applied to many others
  - Example: Cs_2Te following successes of smooth K_2CsSb
- **We enter a cycle of using accelerator-based tools to improve accelerators**
  - providers of advanced tools of material science (accelerator based x-ray light sources) are now availing themselves of the same capabilities to improve the next-generation machines
- **We are looking to define future collaborations**
  - Make our solutions available and partner to tackle new problems

www.lanl.gov/acert