A New Approach to Photocathode Development: From the Recipe to Theory Inspired Design

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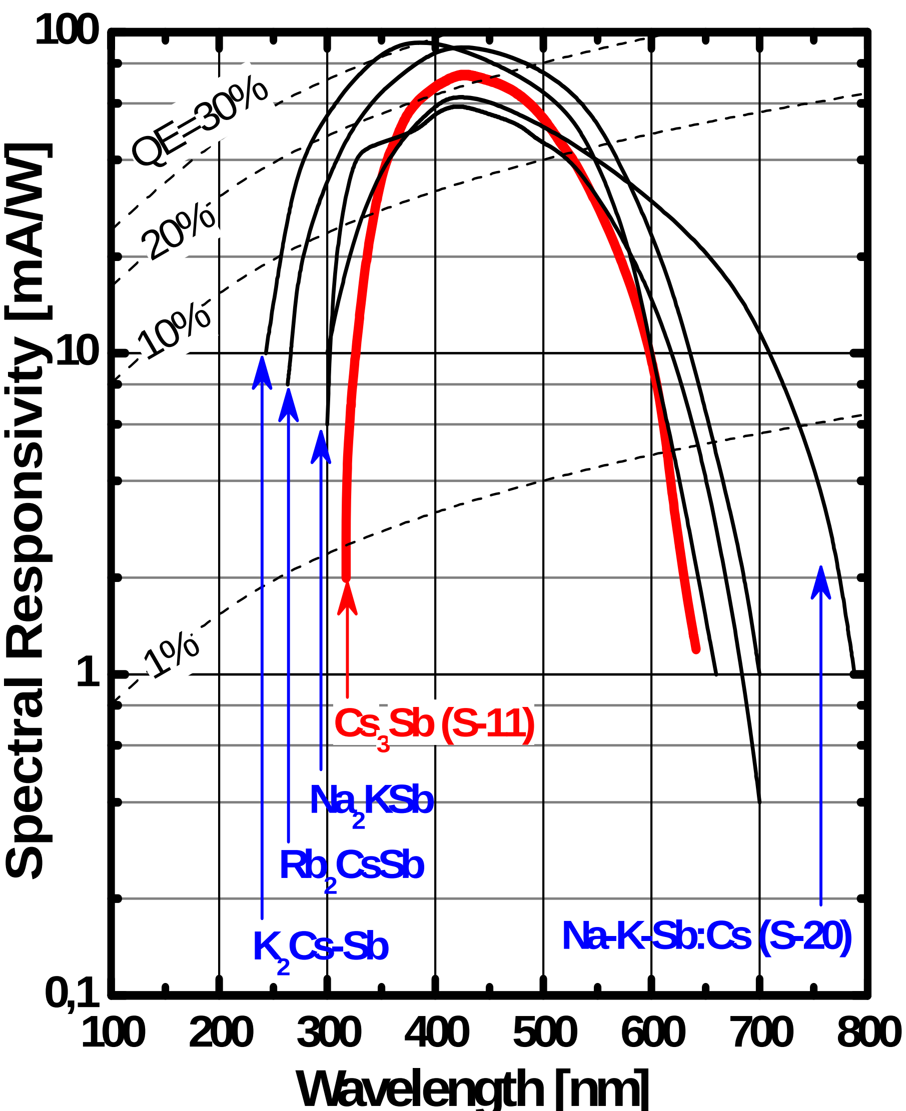
Outline

- Motivation
- What is “theory inspired growth”
- X-rays, a tool to visualize the structure of films
- Conclusion

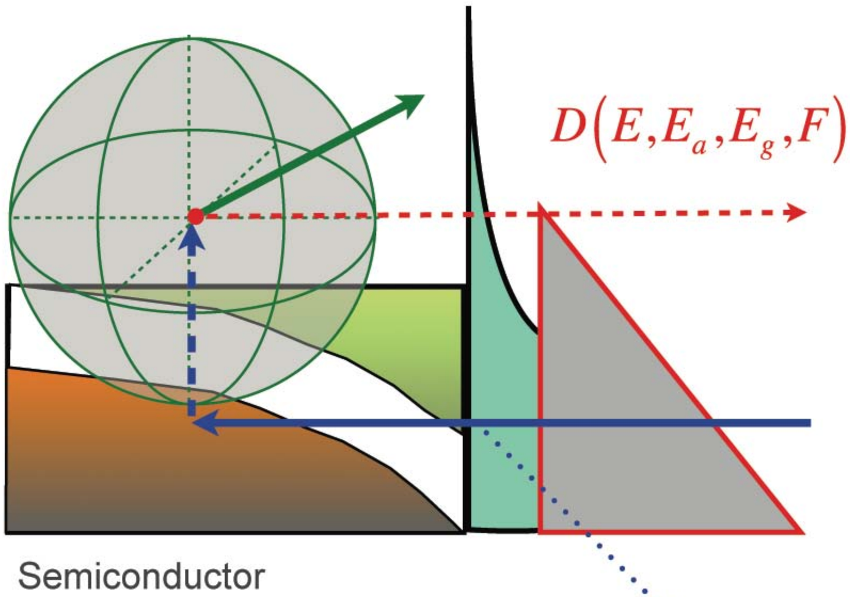
Selection of Cathode Material



- Multi-alkali are
  - "Obvious" choice in the wavelength range around 400nm
  - Most cost-efficient to produce (Thin-film-technology)
- Selection criteria:
  - Process compatibility
  - Wavelength response
  - Conductivity (large area)
  - High Quantum efficiency
  - Low dark current
  - Robustness (device life time)
- Options:
  - CsK_2Sb
  - KNa_2Sb
  - Cs_3Sb

A. Lyashenko

What Determines the Quantum Efficiency



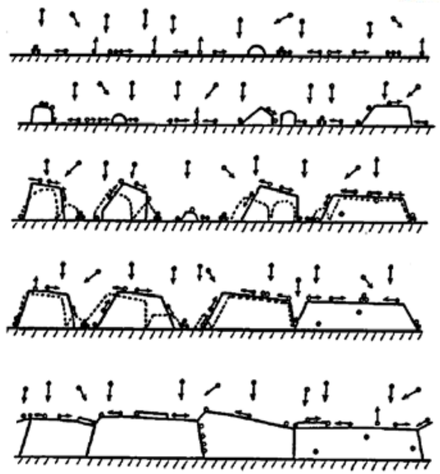
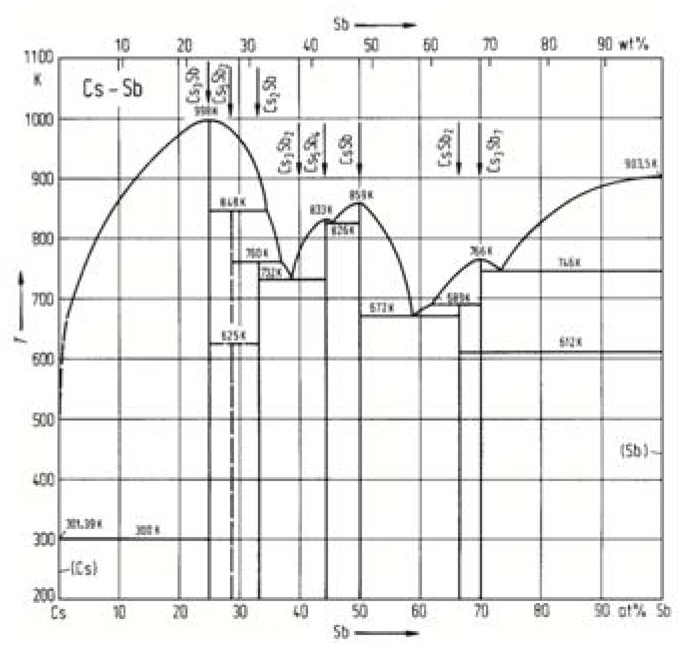
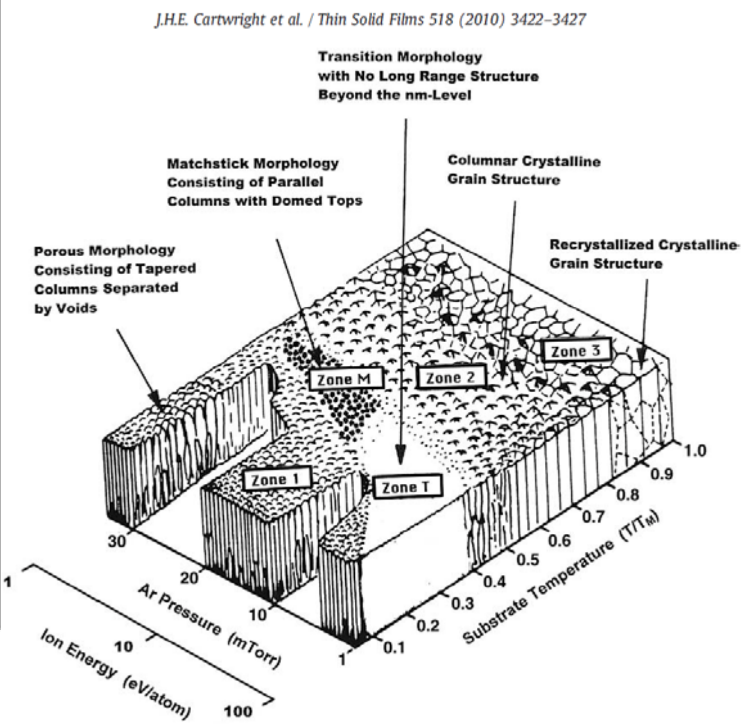


FIG. 1. Schematic diagram illustrating fundamental growth processes controlling microstructural evolution: nucleation, island growth, impingement and coalescence of islands, grain coarsening, formation of polycrystalline islands and channels, development of a continuous structure, and film growth (see Ref. 9).

- Three step model:
  - Absorption
  - Transport to the surface
  - Emission through the surface barrier
  - (reflection losses)
- Ways to manipulate the material:
  - Absorption (band gap & DOS):
    - Band structure by composition variations
  - Transport (scattering):
    - Electron-electron scattering negligible (if not highly doped)
    - Electron-phonon scattering; very difficult to manipulate
    - Electron-impurity scattering; fully growth related
    - Symmetry break (electric fields)
  - Emission properties
    - Surface composition
    - morphology

A Few Thoughts about Thin-Film Growth





- Film morphology is responsible for
  - Lateral and transversal diffusion rate
  - Impurity scattering
  - Speciation distribution trough out the film

Recipe parameters and film structure are strongly correlated

Examples for band-gap variations: K_3Sb

Eg: 1.1eV, 1.3eV, 1.4eV (dependent on crystalline phase)

Roadmap to Optimize Growth Recipe

- **Correlation between functionality and structure**
  - The absorption of photons and generation of photoelectrons
  - The transport of the electrons from the point of generation to the surface
  - The escape from the surface
- **Correlation between Recipe and Structure**
  - Temperature / growth rate
  - Composition of materials
  - Grain size and thickness of the film
- **In - situ Visualization Tool of Microscopic Structure**
  - **X - ray Diffraction:** Crystallographic structure, chemical composition, grain size, crystalline orientation
  - **X - ray Reflectivity:** Control of thickness, various defects, surface roughness

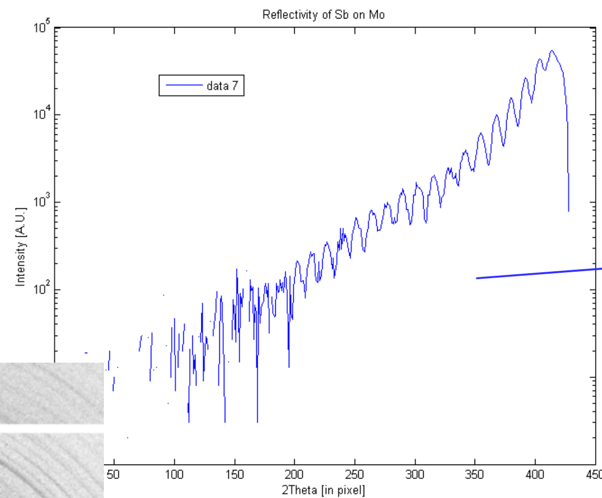
X-rays, a tool to visualize the structure of films

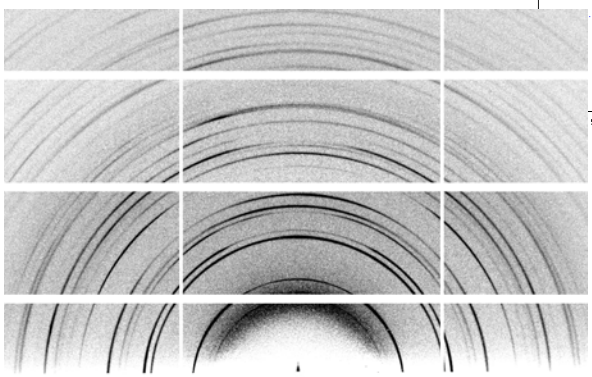
In-situ X-ray Scattering

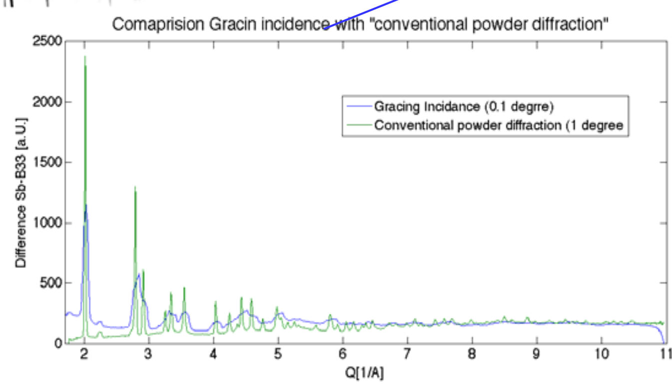
Movie like characterization during the growth:

- Macroscopic film properties
  - Film thickness
  - Roughness
- Microscopic composition
  - Which phases are present
  - Lateral and transversal and homogenuity
  - Crystalline size
  - Preferential crystal growth

Surface composition
  - Local workfunction
  - Chemical composition



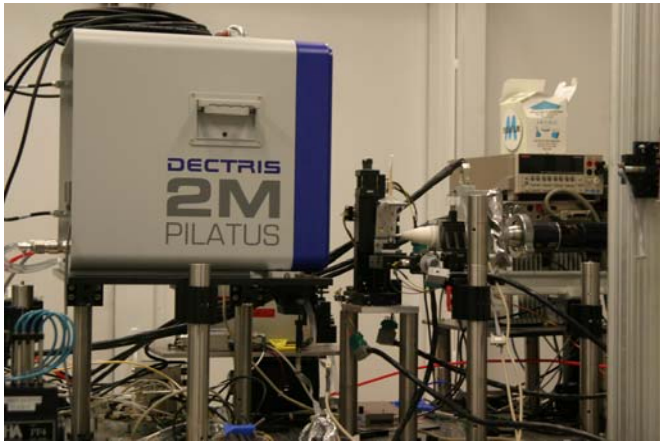




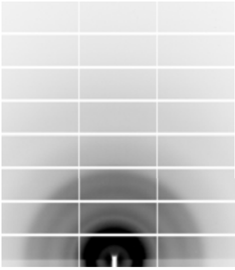
PEEM: collaboration with Howard Padmore (ALS)

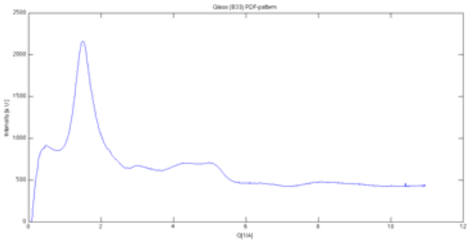
Data-Acquisition & Processing

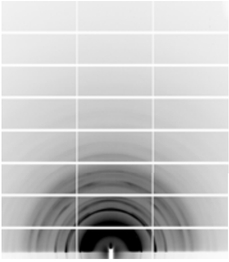
Azimuthal Integration and fit of peak positions

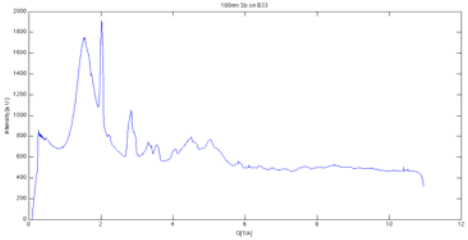


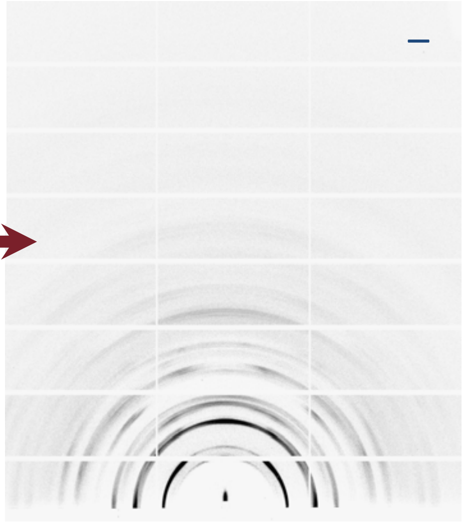
- Single shot reveals full structure
  - Time resolution ~ 100ms
  - Perfect for insitu experiments
  - However: evaporator similar to production condition
- Data-Analysis:
  - Calibration with known standard (CeO_2)
  - “Empty”-pattern (B33)
  - Sb-on B33 pattern
  - Result: Difference showing only Sb-film and changes on glass-substrate

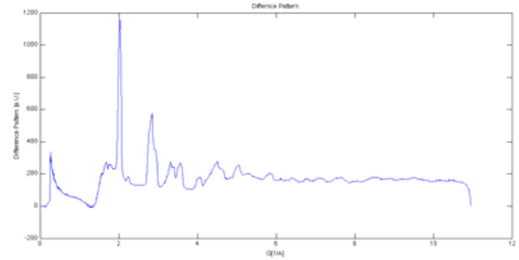




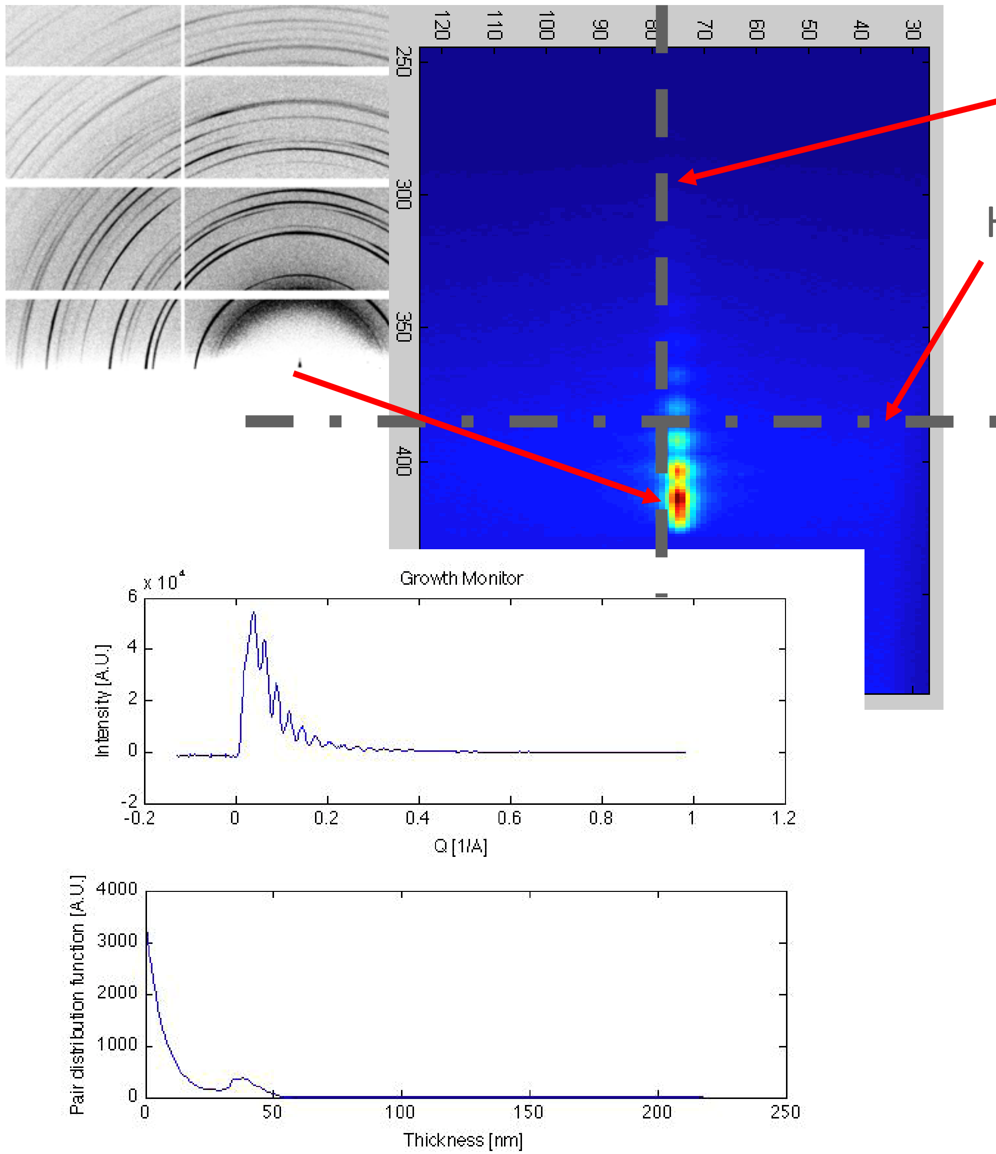








Thickness Measurement of the Film in Real-Time

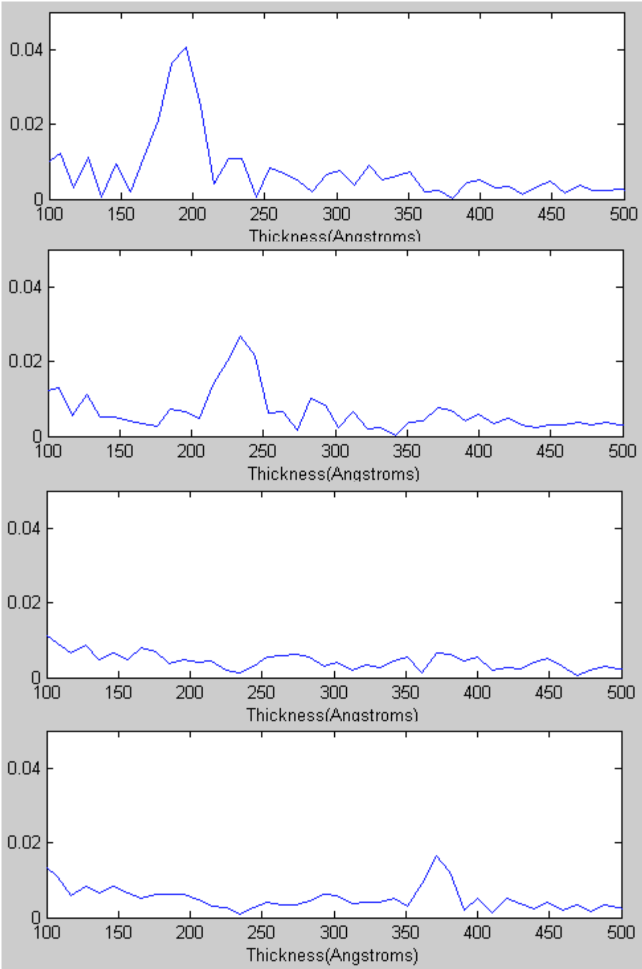


Vertical cut reveals thickness and vertical roughness of film (FFT)

Horizontal cut reveals lateral island-size

- Film thickness and roughness was determined for
  - Sb-fims (different substrate conditions)
  - Sb-film and subsequent K-evaporation
- Currently systematic investigations of
  - different growth conditions
  - surface preparation (interlayer)
- Extracted information (with about 1s time resolution)
  - Thickness
  - Roughness
  - In-plane “partical” size distribution

K-growth on Sb: One Example (Boing-recipe)



Sb-film (18nm)

K-evaporation started

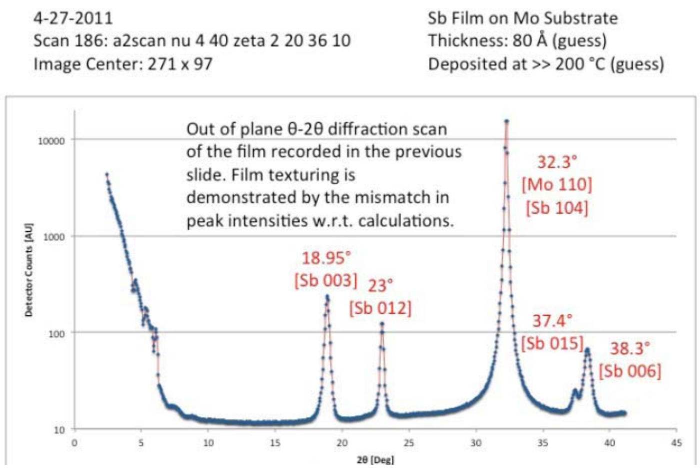
K-evaporation goes on

Final film

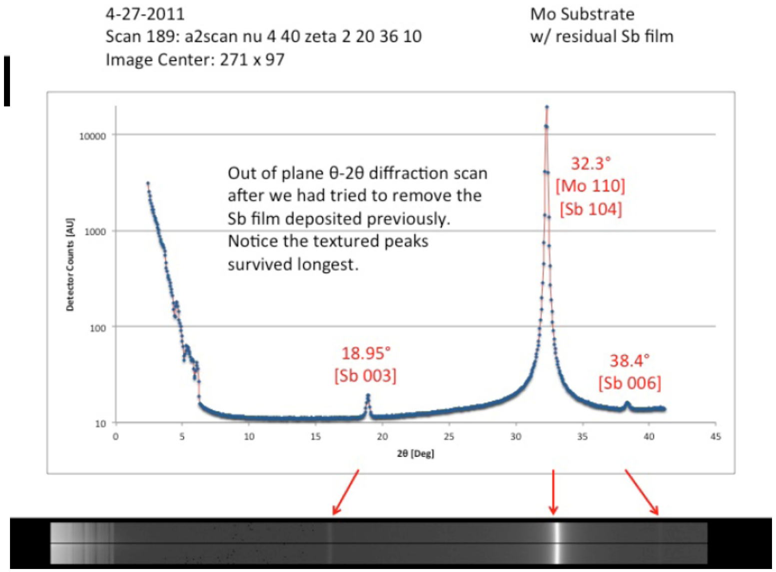
- Sb film starts with about 3 nm (consistent with AFM measurements)
- K-evaporation starts:
  - Roughness is increasing
  - Single peak shows that K instantaneously (1s) reacts and intermixes
- Ongoing evaporation of K:
  - Film shows extreme strong roughness (no peak)
- Final Film:
  - Film roughness decreases with increasing evaporation

Currently in progress: More quantitative analysis and systematic recipe variations

Crystal growth behavior of Sb-film
(Wide Angle X-ray Scattering WAXS)



Sb film as deposited

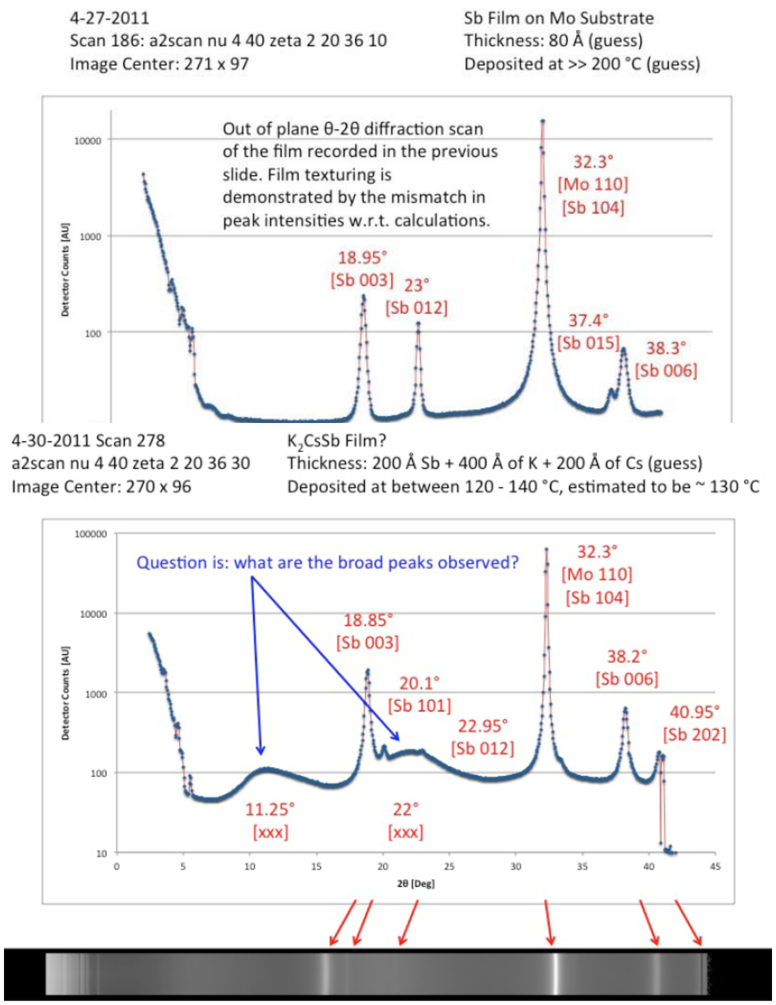


Sb film after heating

- Peak-ratio reveals texturing (pre-orientation of crystallites [Sb 003-orientation])
- After heating:
  - Different crystal orientation show different sticking coefficient
  - Textured crystals "survive" longer

Is multiple applications of heating and evaporations a way to increase the texture and tailor crystal size of the original Sb-film (key to ultra high QE)?

What Happens after K- and Cs-Evaporation



Original Sb-film

Cathode after K- and Cs-Evaporation

- High indexed reflections disappear:
  - Indication that crystallite gets smaller or very strained
- Main reflections show reduced intensity
  - Smaller crystals but still Sb-phase present
- Strong background observable:
  - Not clear which $Cs_xK_ySb_n$-phases are produced
  - Is active cathode material in amorphous phase?

Is K- and Cs- reacting on the grain boundaries with strongly increased strain (like Fe-oxidation?)

Conclusion:
Is Theory Inspired Growth Possible

- **Correlation between functionality and structure**
  - In principle known
  - Phenomenological model available (no first principle calculations)
- **Correlation between Recipe and Structure**
  - Large parameter space is available
  - Recipe shows strong correlations between individual steps
- **In - situ Visualization Tool of Microscopic Structure**
  - All necessary microscopic film parameters can be visualized in real time

The next big steps:

• Proof of principle has been performed and shown.

• Understanding correlations between process parameters and microscopic structures of known recipes.

• Developing new strategies in thin-film technology to create high quantum efficiency cathodes.