Thin-film studies toward improving the performance of accelerated electron source

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Acknowledgments

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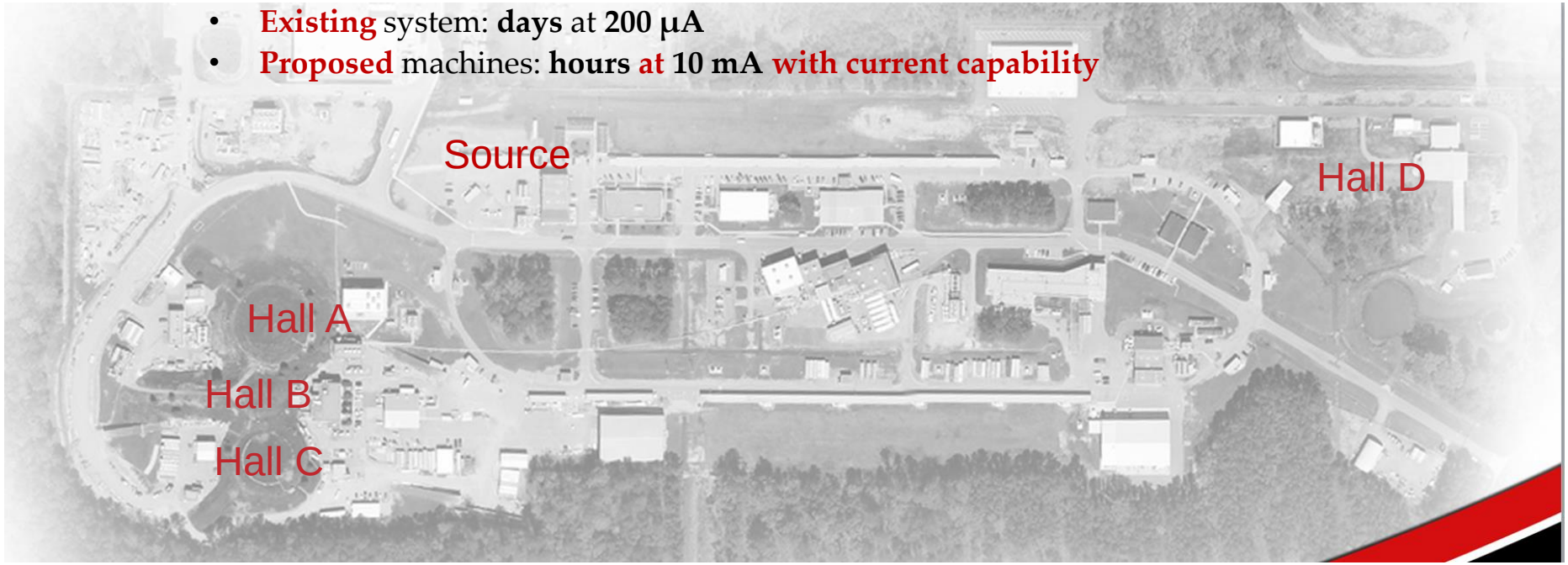
Thomas Jefferson National Accelerator Facility

- 12 GeV electron accelerator for Nuclear Physics
- Up to 90% polarization from DC photoemission source
- Electron currents to 200μA beam (CW) to four experimental halls
  - Recommissioned machine after upgrade to 12 GeV: first beam October 2014
- Lifetime is limited by ionized residual gas accelerated into photocathode
- Future accelerators (e-RHIC, CLIC) require higher currents (> 10 mA)
- Photocathode lifetime
  - **Existing** system: **days** at **200 μA**
  - **Proposed** machines: **hours at 10 mA with current capability**

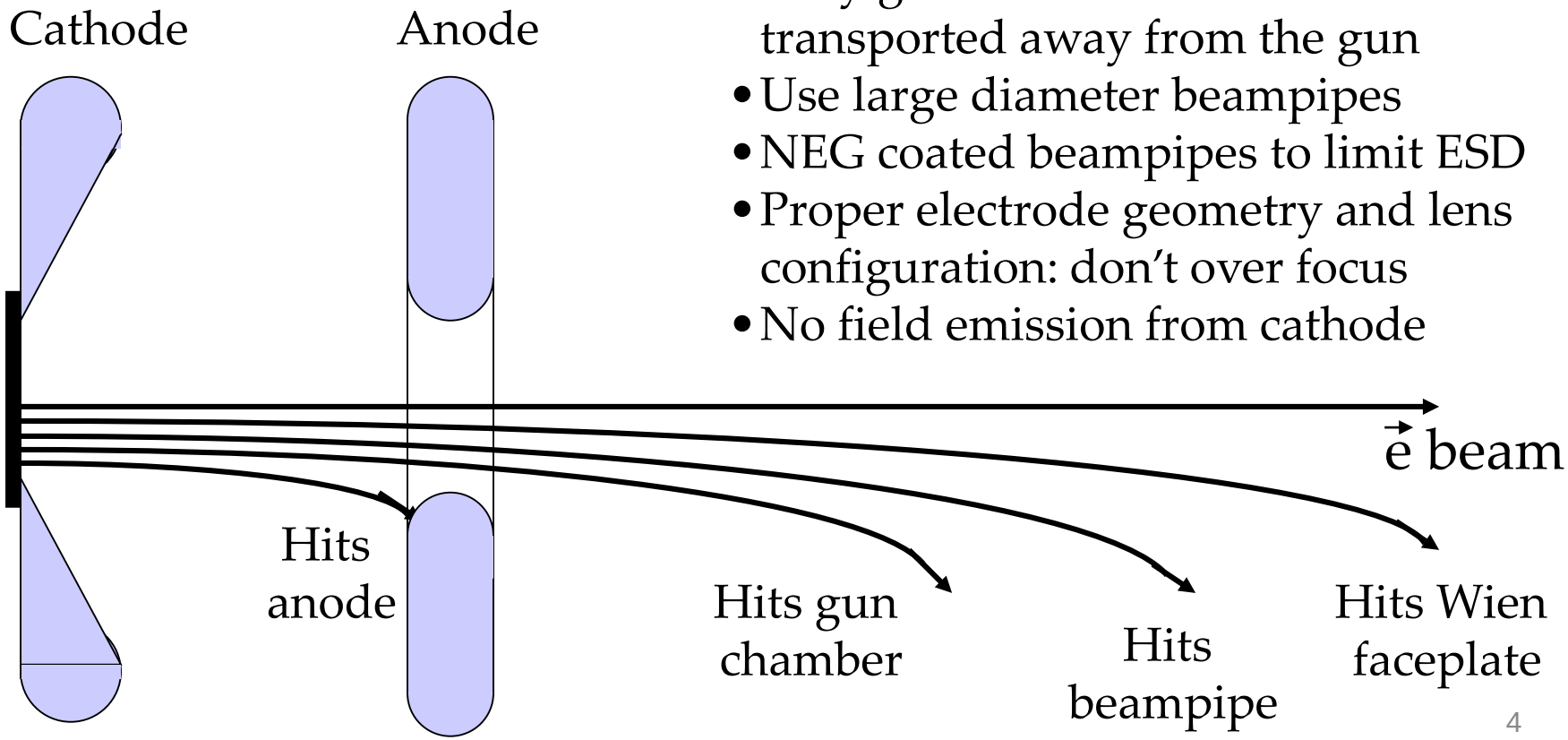


EIC integrated with Relativistic Heavy Ion Collider (eRHIC); Electron-ion Collider (EIC); The Compact Linear Collider (CLIC) ; CW (continuous wave form)

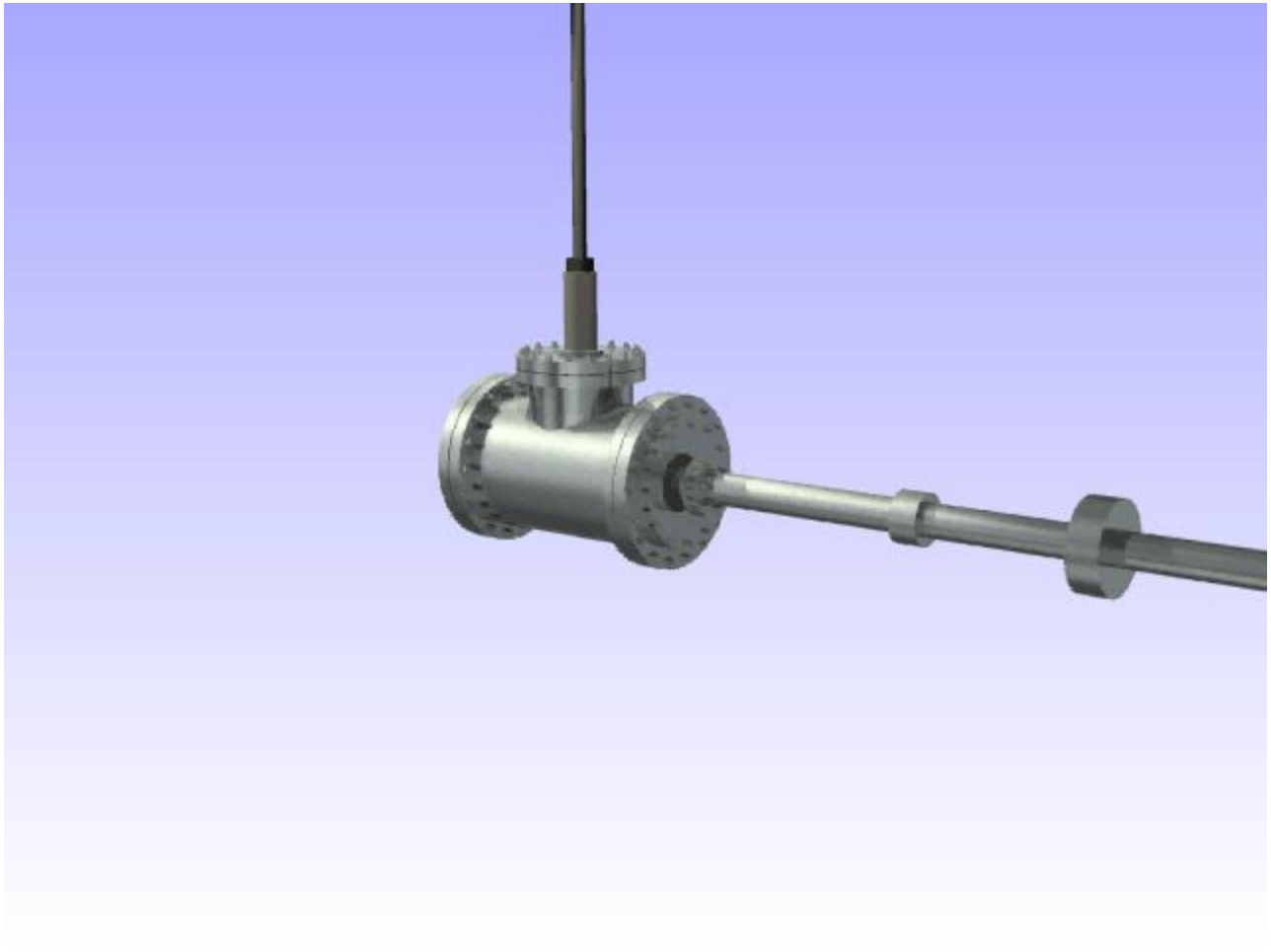
‘Good beam’ versus ‘Bad beam’

The beam that doesn’t make it to the experiment only serves to degrade vacuum. This leads to ion-bombardment and QE decay

- Eliminate stray light
- Only generate electrons that can be transported away from the gun
- Use large diameter beampipes
- NEG coated beampipes to limit ESD
- Proper electrode geometry and lens configuration: don’t over focus
- No field emission from cathode

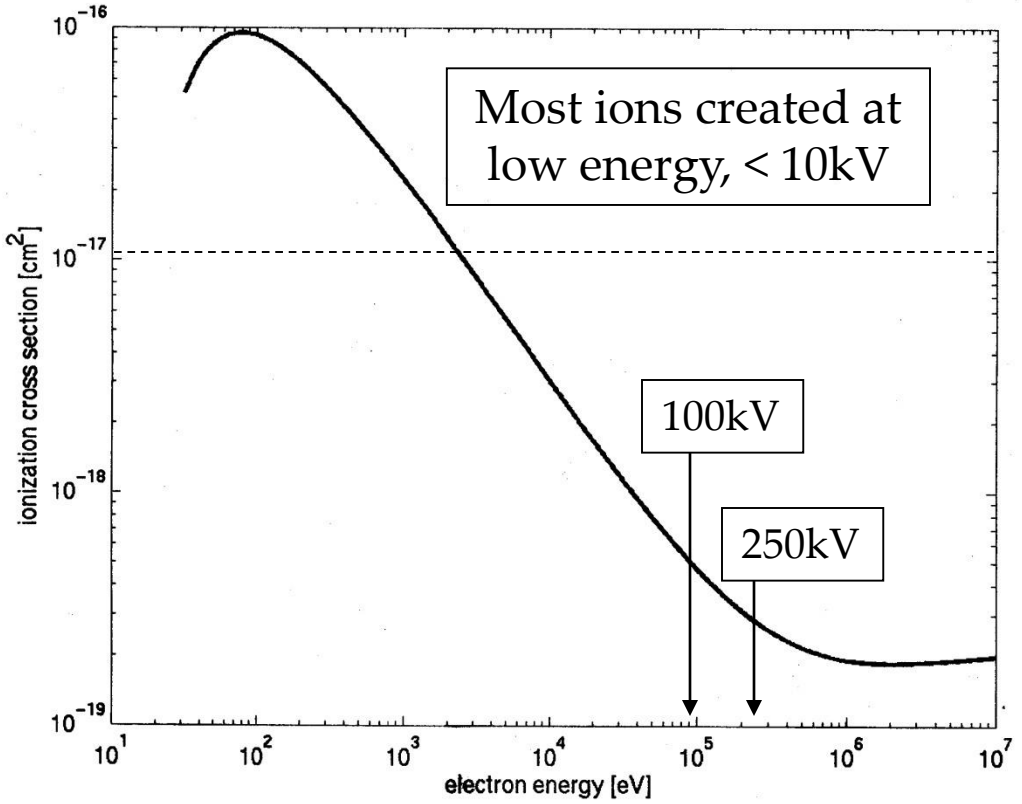


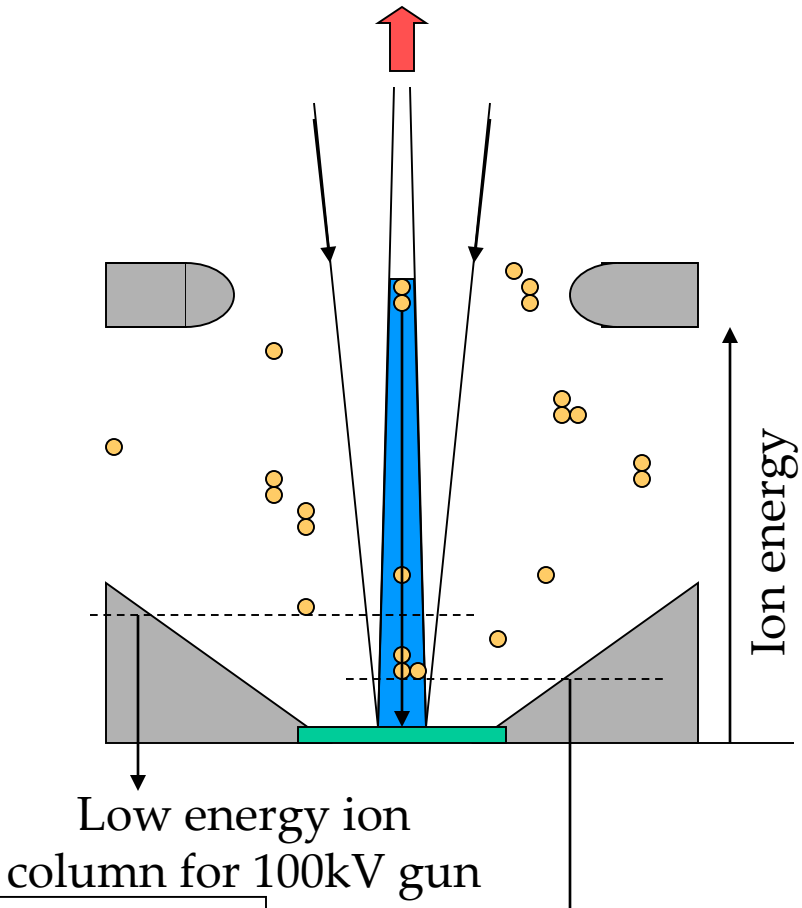
Ion Bombardment



Improve Lifetime with Higher Bias Voltage?

Ionization cross section for H_2





Higher gun voltage means reduced # of "bad" ions, and improved lifetime

Higher gun voltage to prolong charge lifetime?

Higher Gun Voltage:

I. Less ions are created
II. Reduce space-charge emittance growth, maintain small transverse beam profile and short bunch-length; clean beam transport
III. Increase QE by lowering potential barrier (Schottky Effect)
IV. Compact, less-complicated injector

Biggest Obstacle: Field emission and HV breakdown… which lead to bad vacuum and photocathode death

"Charge and fluence lifetime measurements of a DC high voltage GaAs photogun at high average current.," J. Grames, R. Suleiman, et al., Phys. Rev. ST Accel. Beams 14, 043501 (2011)

Overall Goals of this work

To extend operational lifetime in accelerator electron sources and increase current capability through application of thin film(s) for

1. Vacuum improvements
2. Eliminating field emission at DC high voltage
3. Bialkali antimonide photocathode to yield high QE with better lifetime

1. Improving the Vacuum

Improve Vacuum and Improve Lifetime

$$P_{ult} = \frac{Gas\ Load}{Pump\ Speed} = \frac{Q}{S} = \frac{q\,A}{S}$$

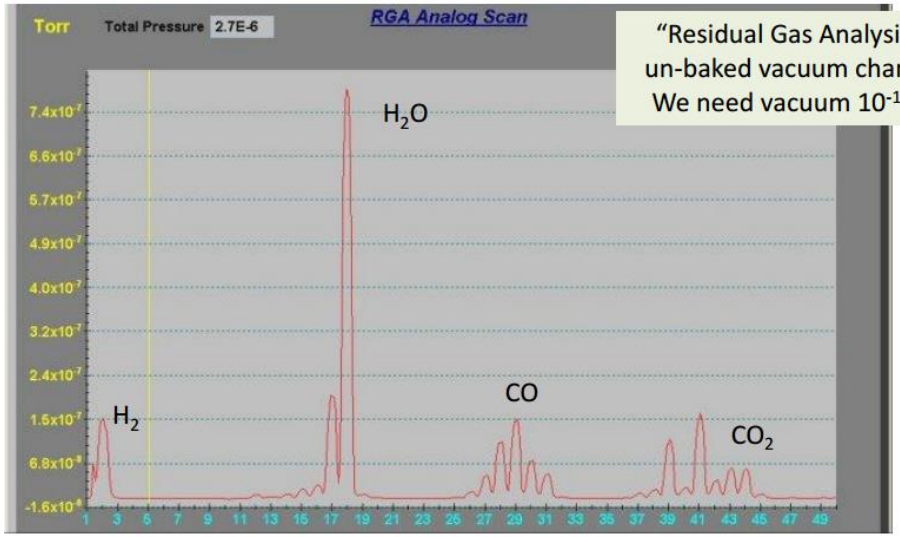
Q= gas load, q= outgassing rate, A = surface area, S = pump speed

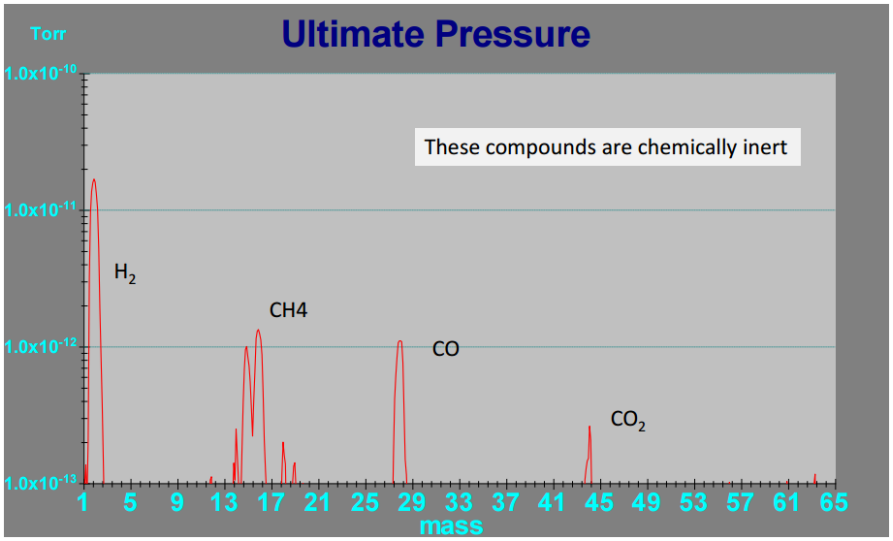
Objective: Make Gas Load small and Pump Speed large

- Gas Load inside a polarized photogun comes from:
  - leaks (real and virtual)
  - outgassing from the vacuum chamber walls and things we put inside the gun
  - from the accelerator beamline
- Limited selection of pumps for UHV and XHV
- Thin film coatings may act as diffusion barrier or pump

What does “Bake” Mean?

It means get rid of water and other chemically reactive substances from inside the photogun… Heat the vacuum chamber at 250 °C for 30 hours





Gas load in vacuum

- Hydrogen dominates at XHV
- Source of H is involved in the production process of metals
- It may be possible that the hydrogen atoms remain mobile even at zero temperature
- Hydrogen is in general dissolved interstitially into metals
- Surface effects and trapping of H at lattice imperfections such as impurities, dislocations, precipitates, grain boundaries, etc. may cause a slowing down of the diffusion
- Hydrogen diffusion also depends on materials structure. Higher diffusivity to the less closely packed structure. Exception, Ti as well as PdCu, the close packed structures for these systems are less dense than the bcc phases.*
- Accelerated diffusion along grain boundaries. Diffusion is faster in polycrystalline materials than single crystal materials.
- a-Si coating is hydrophobic, requires less pump down time to reach good vacuum.

*J. Piper,J.Appl.Phys.37,715 (1966)

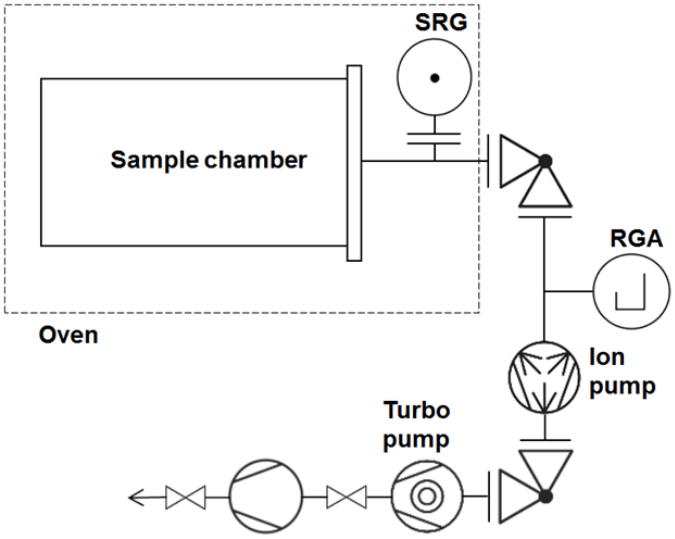
Outgassing rates vs. heat processing and coatings

Identical 304L chambers

- Bare steel (FCC)
- TiN coating (10 μm) (FCC)
- a-Si (SilcoGuard-1000®~800 nm)

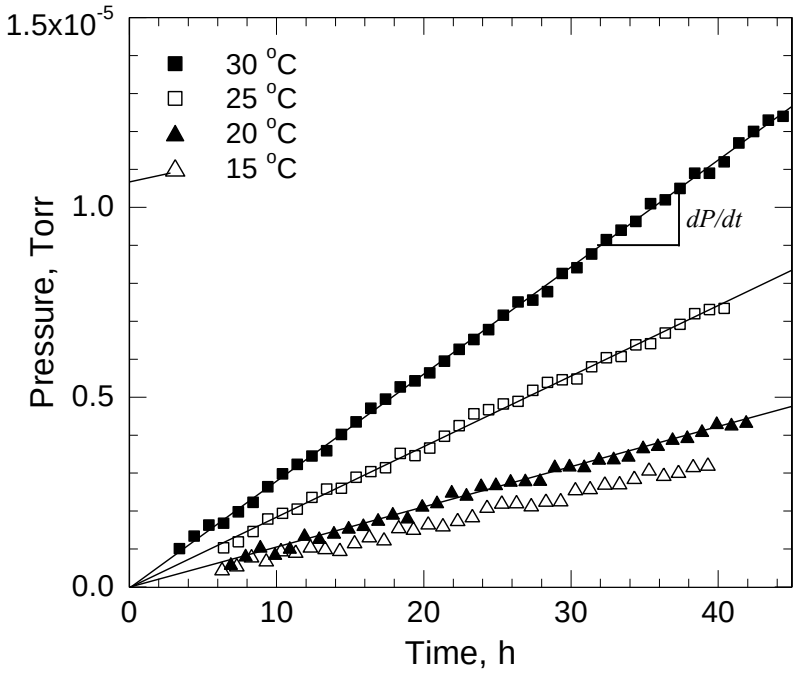
Outgassing rates

- Spinning Rotor Gauge
- Accumulation technique
- Multiple bakes
- Q (T_{room}) after each bake



M.A. Mamun, A.A. Elmustafa, M.L. Stutzman, P.A. Adderley, and M. Poelker, "Effect of heat treatments and coatings on the outgassing rate of stainless steel chambers", J. Vac. Sci. Technol. A 32, 021604 (2014).

Rate of rise measurements



Outgassing Rate:

$$q = \frac{dP}{dt} \times \frac{V}{A} \quad \left(\frac{\text{Torr} \cdot \text{L}}{\text{s} \cdot \text{cm}^2} \right)$$

Key observations

- The outgassing rate for the a-Si chamber is strongly dominated by surface effects rather than by the rate of diffusion of hydrogen in the bulk stainless steel.
- Diffusion of hydrogen from the material during bakes is not significant after a-Si coating (a diffusion barrier at high temp). However, It's hydrophobic nature helps faster pump down.
- TiN acts as NEG with small pump speed, baking helps to degas.
- Worth trying to investigate the a-Si coating on a baked TiN coated SS chamber. May eliminate need of further bake after venting.

Outgassing rate vs. room temperature

- First tests of SilcoGuard-1000™
- Reproduction of SS results @ 400°C bake
- Reproduce and question TiN outgassing

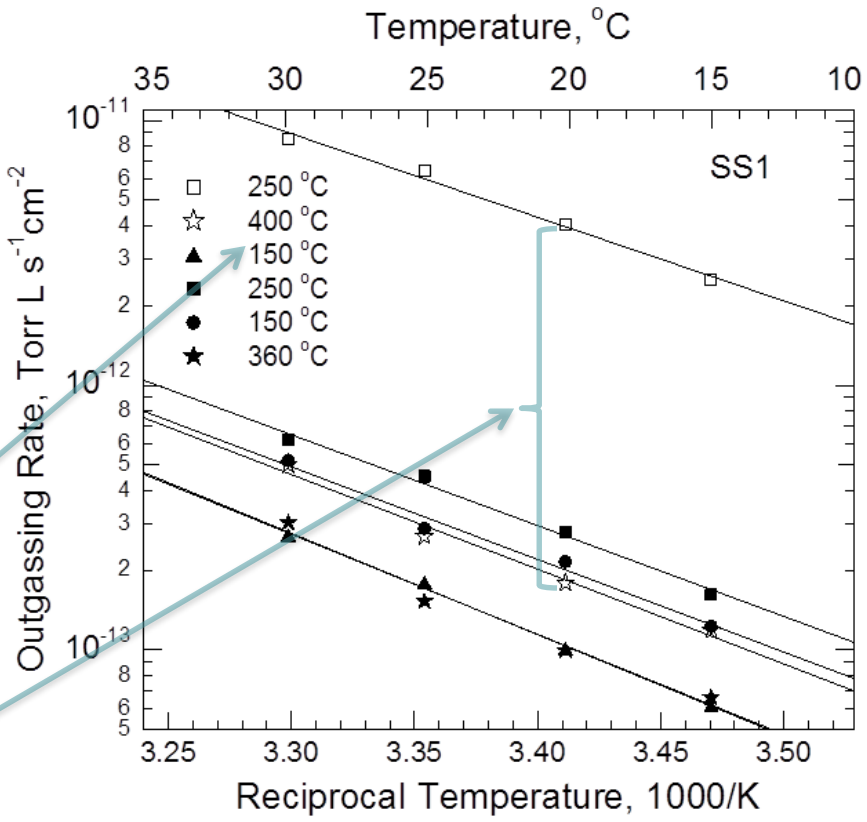
Linearity on Arrhenius plot:

Temperature dependent diffusion activation energy

$$\log q = \log A - \left(\frac{E_D}{R}\right)\frac{1}{T}$$

Bake temperatures, in order

Factor of 20 reduction in outgassing after 400°C heat treatment



Stainless Steel

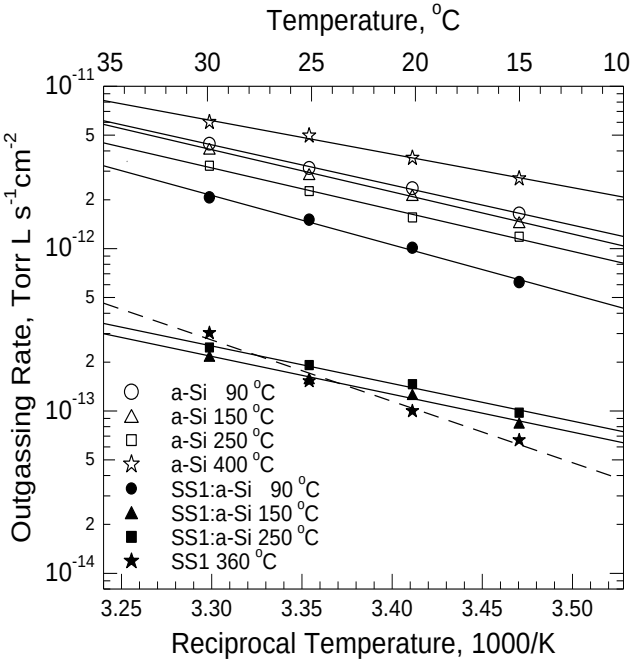
Room temperature dependence: a-Si and TiN coated chambers

Linearity on Arrhenius plot:

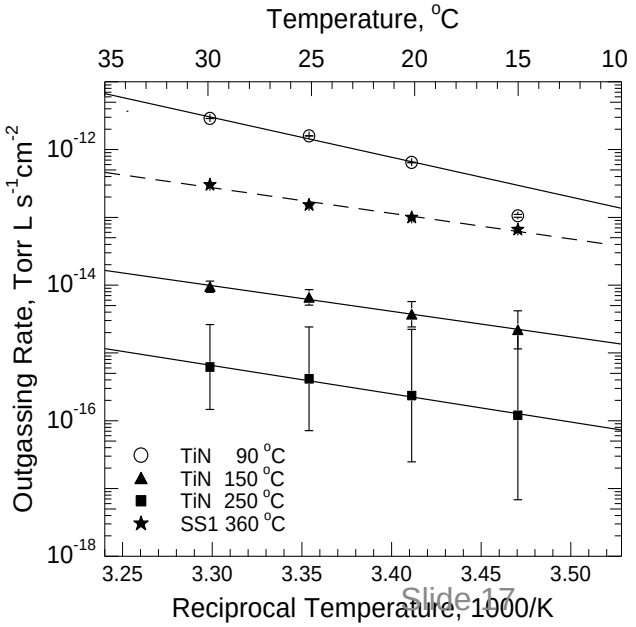
Temperature dependent diffusion activation energy

$$\log q = \log A - \left(\frac{E_D}{R}\right)\frac{1}{T}$$

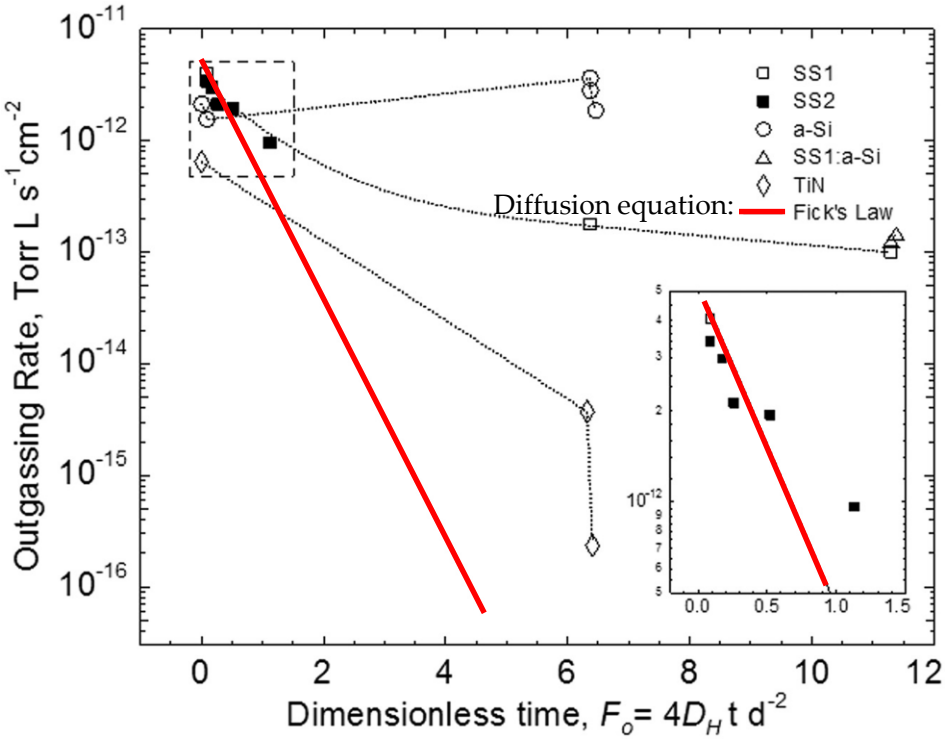
a-Si



TiN



Recombination vs. Diffusion limited outgassing



Outgassing Rate of SS:
- First, limited by diffusion
- Then, by recombination
- Transition occurs near F_0=1.

Outgassing Rate of a-Si:
- Recombination limited

Diffusion constant:

$$D_H(T) = D_0 \exp\left(\frac{-E_D}{RT}\right)$$

Dimensionless Fourier number:

$$F_0 = 4D_H\left(\frac{t}{d^2}\right)$$

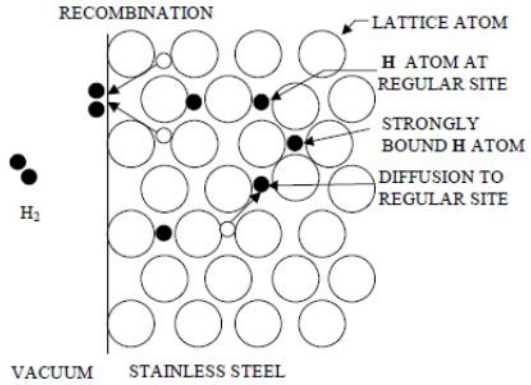
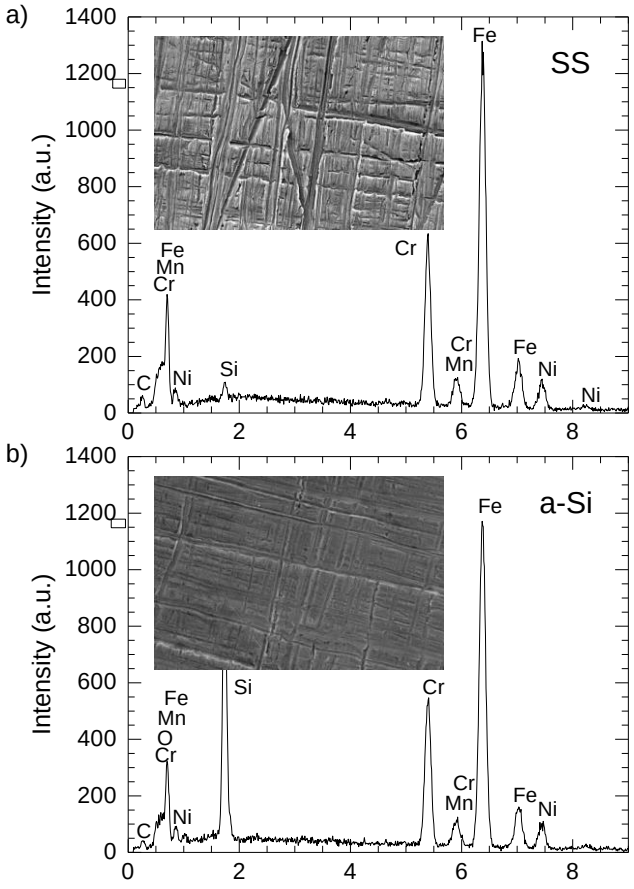
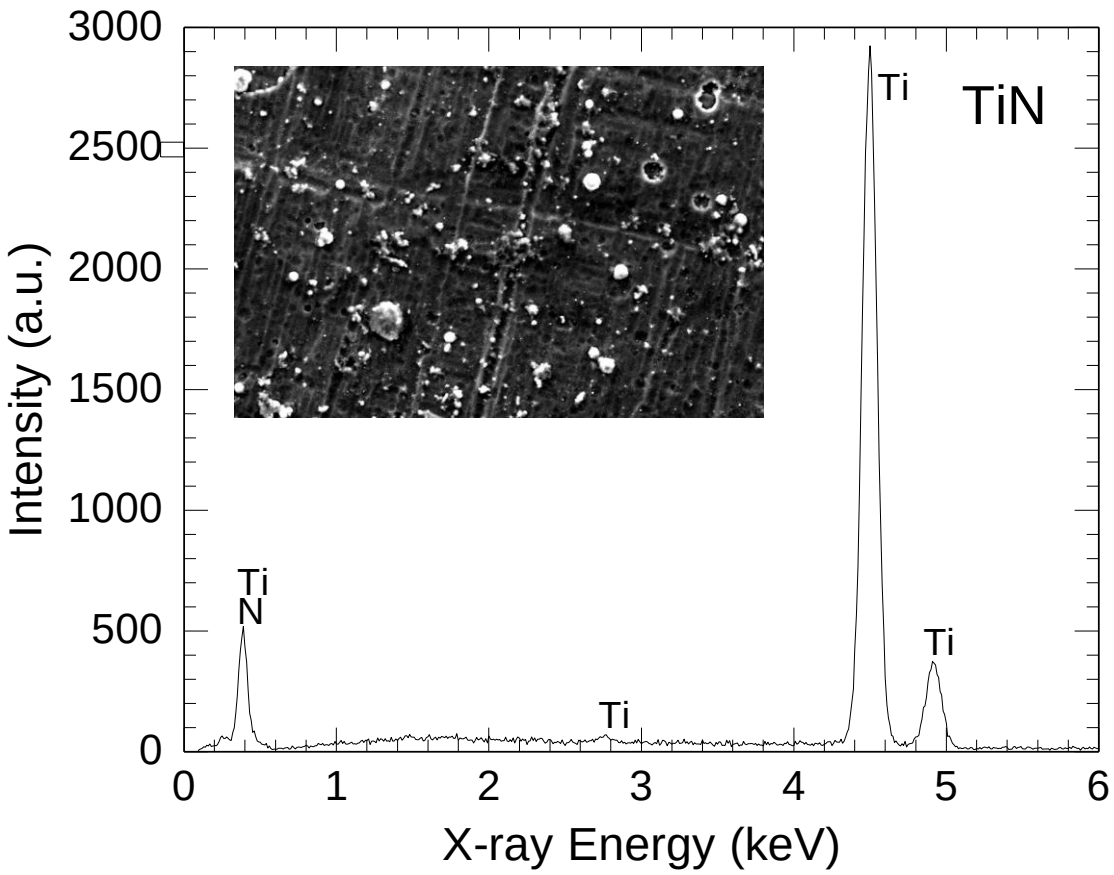


Fig. 3. Schematic diagram of a recombination process at the SS surface.

Yuichi Ishikawa, Vincenc Nemanic, Vacuum 69 (2003) 501–512

Does the TiN coating pump?

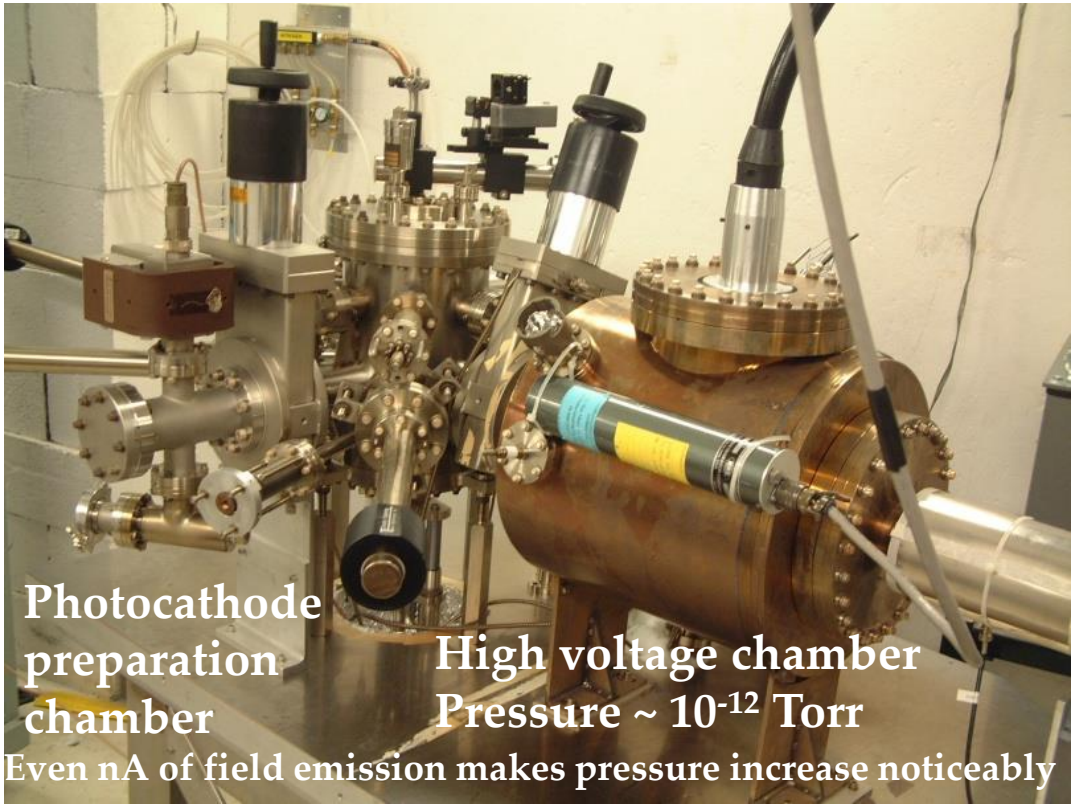


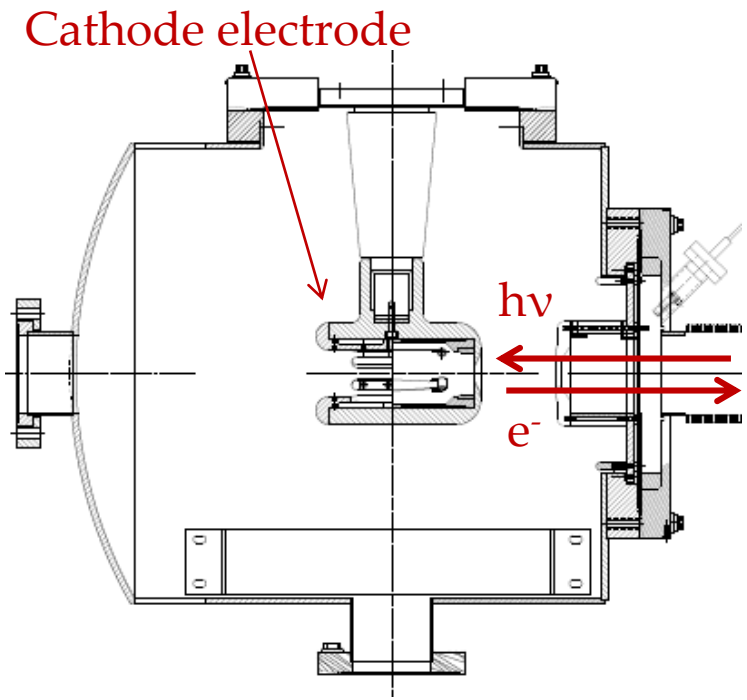


Ti:N ratio ~ 1:1, but inhomogeneous

2. Reduction of Field Emission

Electron beams via photoemission





- Higher voltage = better beam quality
- But higher voltage often leads to field emission
- Field emission degrades vacuum
- Bad vacuum degrades photocathode yield

Sources of field emission

1. Electron emission based mechanism:
   - Micro-protrusions

2. Micro-Particle based Mechanisms:
   - Embedded impurity particles of the polishing medium(Alumina, Diamond,…)
   - Filamentary structures that have been torn from the surface.
   - Dust particles attached to surface by Van der Waals forces from handling or assembling

3. Regenerative Ionization:
   - Local desorption of gas pockets after the bake out

4. Ion Exchange Mechanism
   - Liberating electrons from contaminated areas

Choice of electrode materials

- Traditionally, we choose metals like:
  - Stainless steel, titanium-alloy, or molybdenum
- And Smooth surface
  - Diamond paste polishing typically employed
  - Time consuming

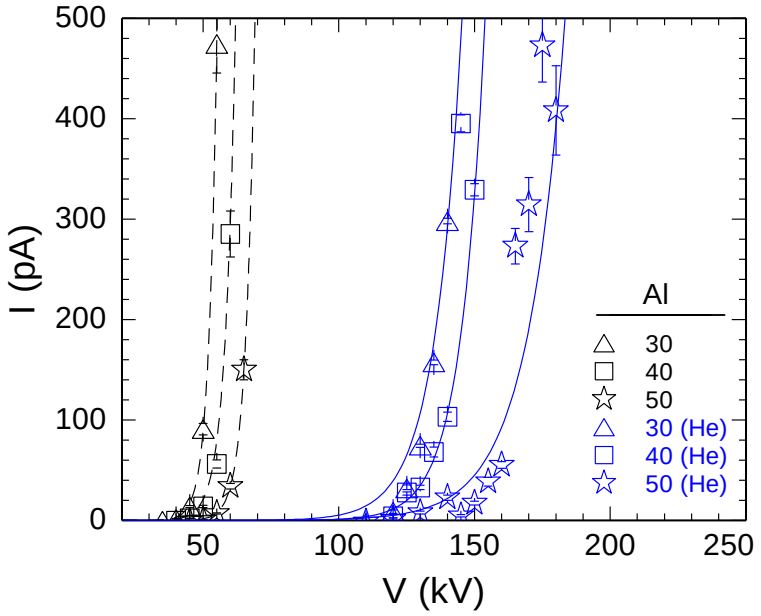
Objective: to eliminate field emission in inexpensive Al electrodes using hard TiN coating that has significantly higher work function than commonly use other electrode materials.

- Aluminum is cheap and easy to machine
- Takes just hours to polish to mirror-like finish with silicon-carbide paper
- Good thermal conductor, compared to steel, a nice feature when gun provides high current
- We used a "boutique" vendor to coat our small electrodes with IBED TiN coating.

High voltage test stand

Materials studied:

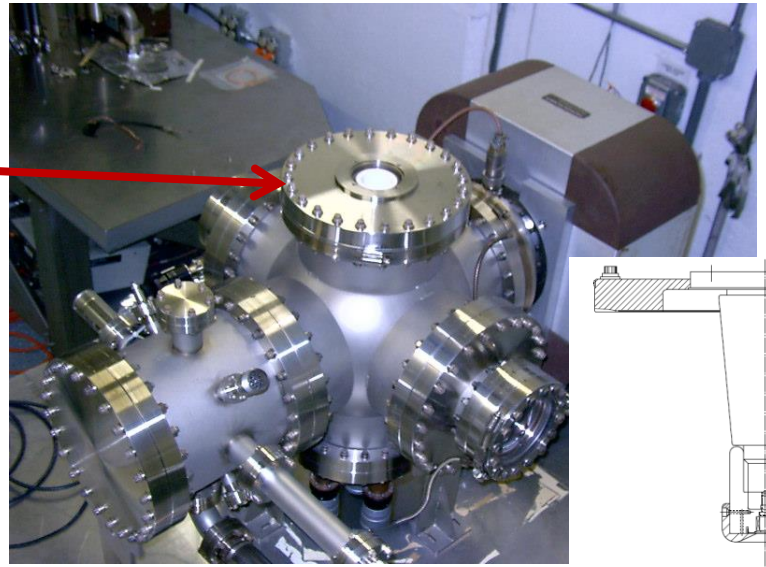
- Ti-alloy (Ti-6AL-4V) DPP
- Aluminum (Al 6061) , 1200 grit polished
- TiN coated Aluminum , 1200 grit polished, 7 nm rms roughness

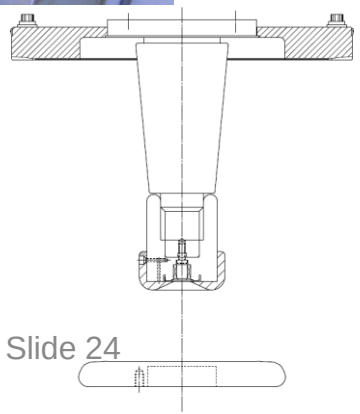


Max.voltage
225 kV

Max. field strength

Gap, mm	F, MV/m
50	12.8
40	13.7
30	15.4
20	18.7
10	28.9





M.A. Mamun, A.A. Elmustafa, R. Taus, E. Forman, and M. Poelker, "TiN Coated Aluminum Electrodes for DC High Voltage Electron Guns", J. Vac. Sci. Technol. A 33, 021509 (2015).

Field emission results

Maximum field strength for <100 pA at different cathode-anode gap:

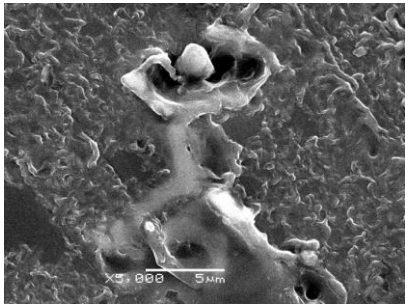
C-A	A1 (He)	A2 (He)	TN1 (He)	TN2 (He)	TN3 (He)	TA1	TA2
gap	1200 grit	1200 grit	800 grit	1200 grit	1200 grit	DPP	DPP
50mm	8.99	9.14	>12.80	>12.80	>12.80	10.90	>12.80
40mm	8.51	9.51	>13.73	>13.73	>13.73	10.50	>13.73
30mm	9.02	11.96	>15.35	>15.10	15.15	10.95	>15.35
20mm	10.00	11.84	17.57	18.35	18.40	12.95	>18.66

Maximum voltage and field strength for <100 pA:

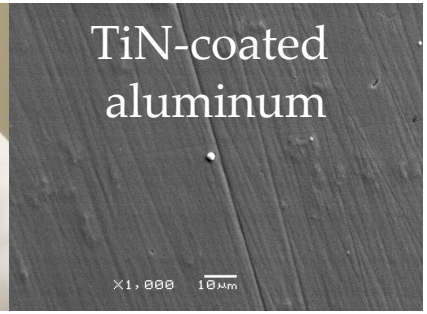
Ti-alloy	Bare Al	TiN/Al	
225 kV	160 kV	225 kV	V_{max} at 50 mm gap
18.4 MV/m	11.8 MV/m	>18.7 MV/m	E_{max} at 20 mm gap

TiN coated electrode:
Field emission **<15 pA** at 175 kV with ~ **22.5** MV/m at **10 mm** gap in the baked chamber without any gas processing.

TiN coated aluminum

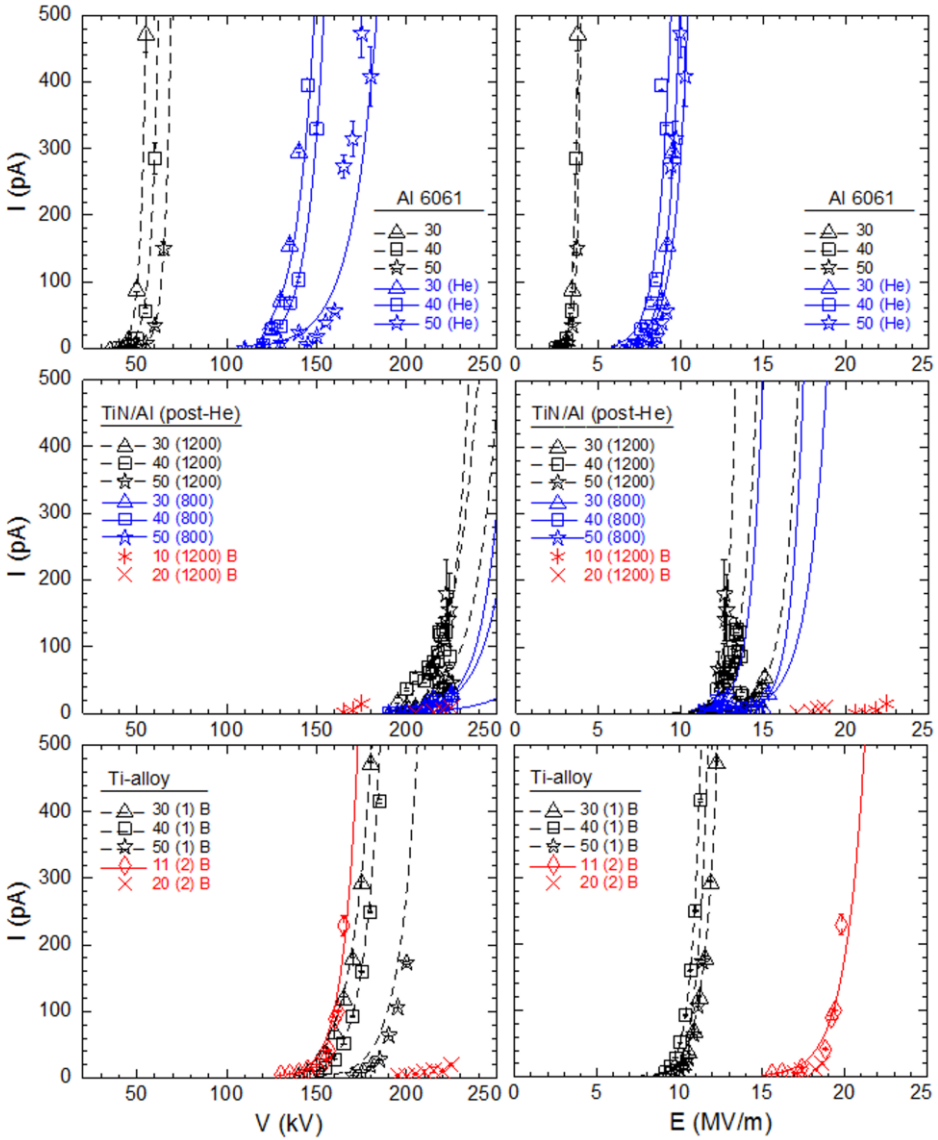


Bare aluminum: easily damaged

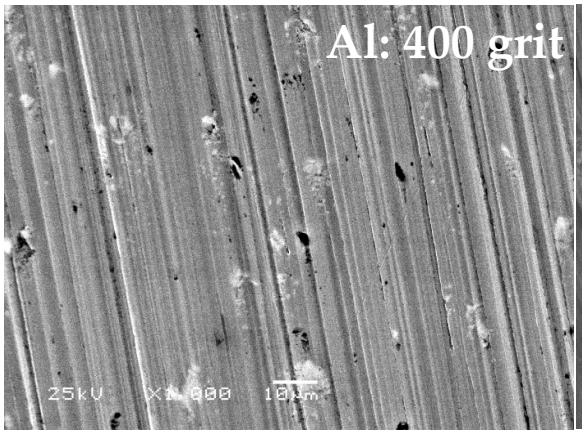


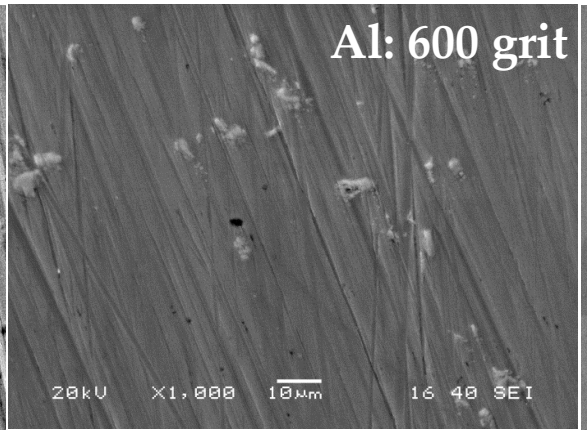
Diamond-paste polished Ti-alloy: a traditional "hard" electrode

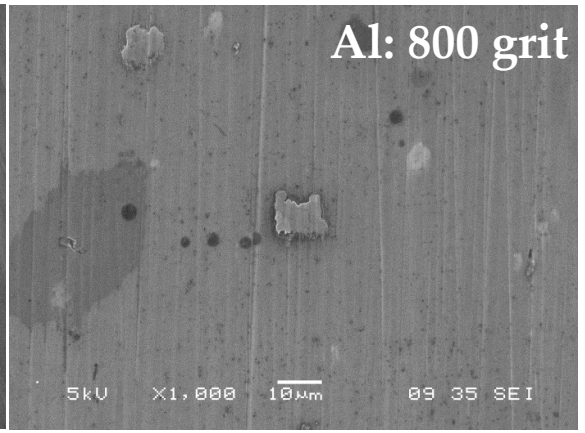
I vs. V and I vs. E

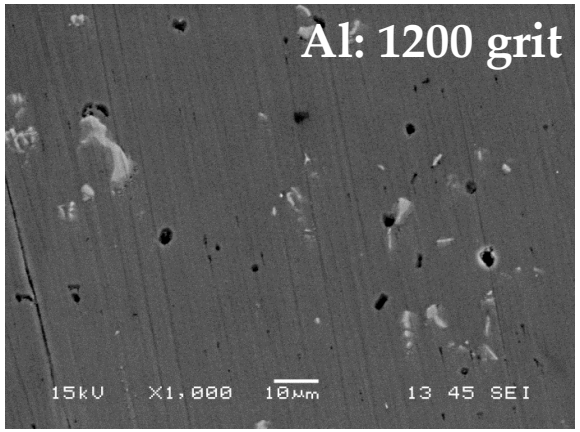


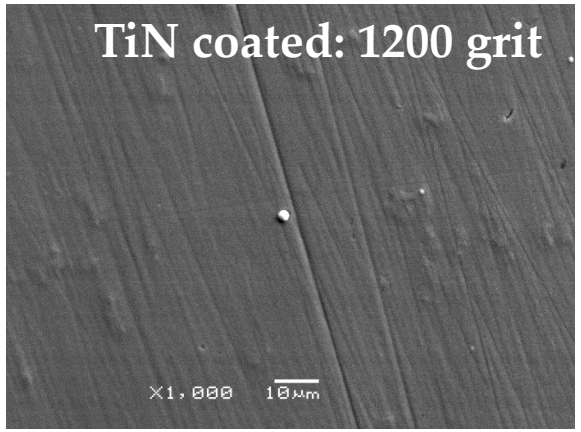
Surface images of Al and TiN-Al





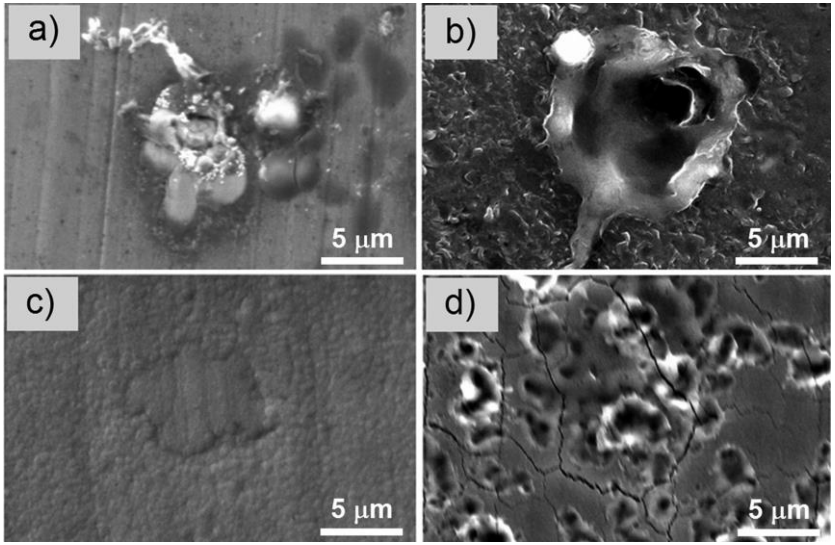






Note, the black spots are voids and defects, not particulate contamination from sand paper polishing

Surface images of Al and TiN-Al

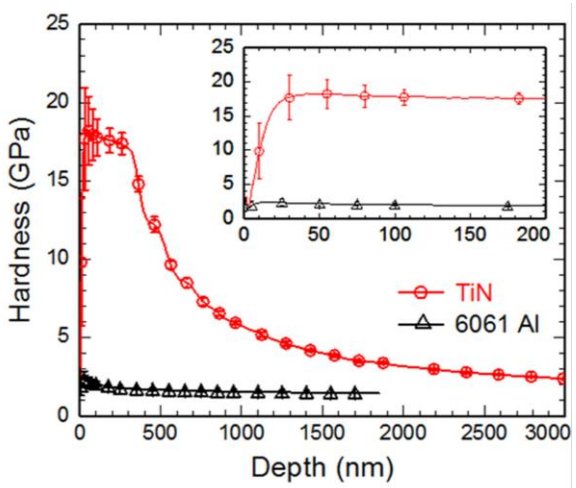


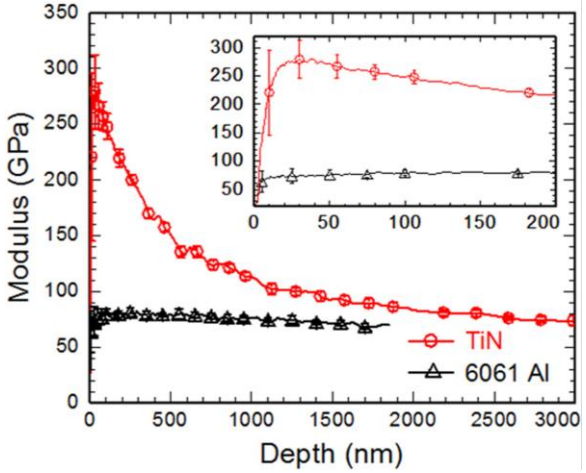
Prior and post HV conditioning: Al (a, b), TiN (c, d)

SEM images of bare and TiN-coated aluminum electrode surface,
(a) bare aluminum electrode surface showing a typical defect,
(b) bare aluminum electrode surface showing a defect produced as a result of helium gas conditioning,
(c) TiN-coated aluminum electrode surface showing a subsurface defect covered by TiN coating, and
(d) TiN-coated electrode surface subjected to high field strength, and damaged postbreakdown

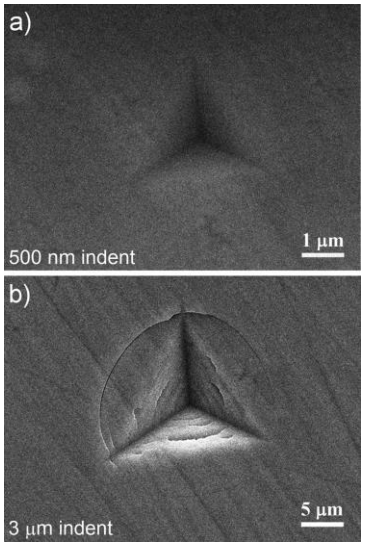
Nanoindentation hardness and modulus

- The TiN coating provided surface finish (7 nm rms roughness) and mechanical properties of the coating (H: 18 GPa and E: 270 GPa) superior to those of stainless steel, titanium-alloy, and niobium electrodes.
- TiN coating is known to have better work function (5 eV) compared to SS (4.5 eV) or Al (3.5 eV).
- These features contributed to the improved high voltage performance of the TiN-coated aluminum electrodes.





- No sign of delamination: ⟹
  a) 500 nm indent
  b) 3 μm indent



3. High QE Photocathode

High QE photocathode for high current photogun

$$QE(\%) = \frac{124}{\lambda_{laser}} \cdot \frac{I}{P_{laser}}$$

$$Q = \lambda \times E \times QE(\%)/124$$

λ = laser wavelength (nm)
P = laser power (mW)
I = instantaneous photo-current (μA)
Q = bunch charge(nC)
E = optical pulse energy (μJ)

High QE at the longest practical wavelength for high average current and high bunch charge applications.

QE is low in metals (<0.003%) because of electron-electron scattering in the conduction band.

QE in semiconductors is typically 1-10% because of scattering between electrons and phonons, i.e. electrons thermalize with the lattice.

Bialkali antimonide photocathode

- NEA GaAs photocathode has a limited lifetime - QE decay is still a puzzle for GaAs – probably due to ion-back bombardment that damages the activated surface
- PEA CsK_2Sb multi-alkali photocathode is based on bulk material properties rather than surface effects: we expect longer lifetime

Cathode Type	Typical λ (nm), (eV)	QE (%)	Vacuum (Torr)	Gap Energy + Electron Affinity	Thermal Emittance (μm/mm (rms))
GaAs (Cs,F)	532, 2.33	10	10^{-12}	1.4	0.44
CsK_2Sb	532, 2.33	10	10^{-10}	1+1.1	0.56

Dowell et al., *NIM-A*, **622**, (2010) 685-697

Objective: In this work our goal is to identify optimal deposition condition for high QE bialkali antimonide photocathode with extended lifetime.

Bialkali antimonide photocathode by codeposition

- Substrate temp: Falling from 120 °C to ° 80 C
- Duration of evaporation varied depending on QE to optimize
- Monitored QE using a 532 nm green laser with cathode biased at V_b=284 V
- K ampoules (1 gram in argon) from Espi-metals, and Cs ampoules (1 gram in argon) from Strem Chemicals

Evaporation of bi-alkali was controlled by adjusting heater power and gas flow through the effusion source., in the following ranges:

- Inlet tube: 381-462 °C
- Dispensing tube: 232-294 °C
- Reservoir tube: 153-281 °C

Partial pressure of alkali vapors was monitored using residual gas analyzer (RGA) during deposition, not the usual quartz crystal thickness monitor. Such as, for 387 °C at inlet tube, 259 °C at dispensing tube , and 184 °C at reservoir:

- Cs PP on RGA: $1.35(\pm0.1)\times10^{-10}$ Torr
- K PP on RGA: $3.15(\pm0.5)\times10^{-11}$ Torr

The duration of deposition, to optimize QE, was also recorded for each case.

Optimizing the growth =>

Deposition time, min	Current to Sb furnace, A	PP of Sb in RGA monitor, Torr	Substrate temperature	Relative Sb qty.	Baked system	Unbaked system
30	32.7±0.2	$5.5(\pm0.5)\times10^{-11}$	200 °C	1	√	√
70	32.7±0.2	$5.5(\pm0.5)\times10^{-11}$	200 °C	2.33	√	√
100	32.7±0.2	$5.5(\pm0.5)\times10^{-11}$	200 °C	3.33	√	√
120	31.2±0.2	$6.0(\pm0.5)\times10^{-11}$	200 °C	4.36	×	√
120	33.7±0.2	$1.0(\pm0.2)\times10^{-10}$	200 °C	7.27	√	√

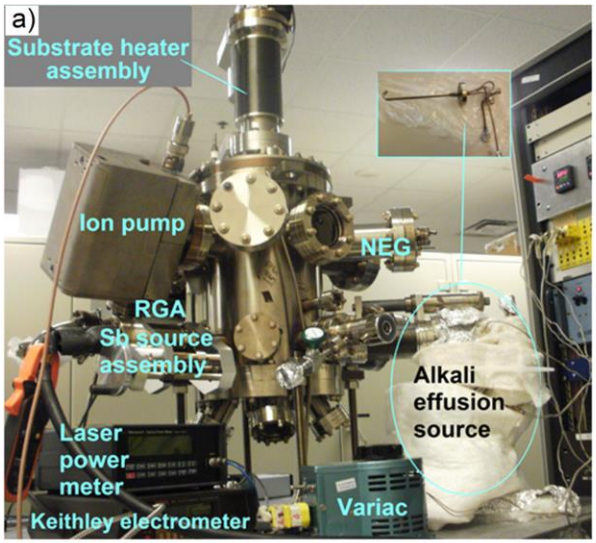
M.A. Mamun, C. Hernandez-Garcia, M. Poelker, and A.A. Elmustafa, "Effect of Sb thickness on the performance of bialkali-antimonide photocathodes," Journal of Vacuum Science & Technology A 34 , 021509 (2016).

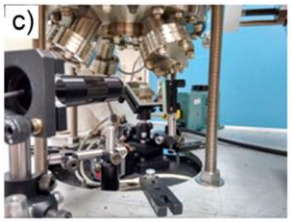
Key observations

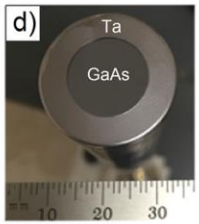
- Thin Sb films are smooth and dense compared to thicker Sb films that exhibit a high degree of roughness and porosity with increasingly higher grain size.
- The required amount of alkali materials for optimum QE depends on the ratio of the effective surface area to the volume of Sb. Thicker layers of Sb served to "store" more alkalis.
- The photocathode with the thickest Sb film (6.7 μm) provided the longest lifetime (~42 days). In stoichiometric photocathode, increasing thickness means increasing ability to replenish the depleted alkali species from surface.
- Highest QE from the Ta and GaAs substrates was 10% and 8% at 532 nm, and 24.2% and 22% at 425 nm, respectively.
- The effectiveness of light detection by a photocathode surface at different wavelength is known to depend on photocathode stoichiometry. Any variation in photocathode stoichiometry is expected to accompany a variation in bandgap energy.
- The QE values varied from ~3% to 10% for photocathodes with Sb layer thickness from <50 nm to ~7 μm. QE increased with crystallite size
- Crystallinity of the Sb film can influence the photocathode sensitivity and that a variation in crystallite sizes can affect the energy bandgap.

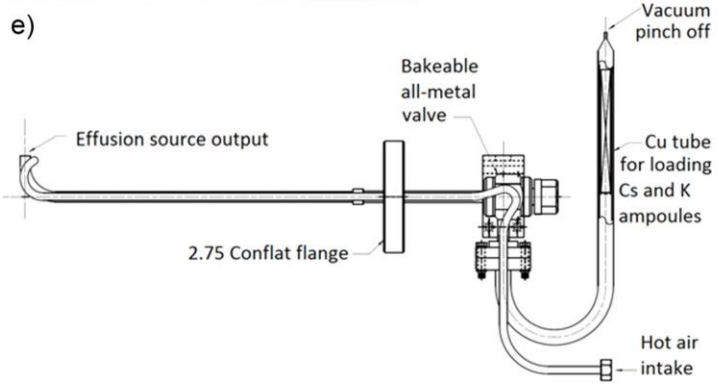
Deposition chamber

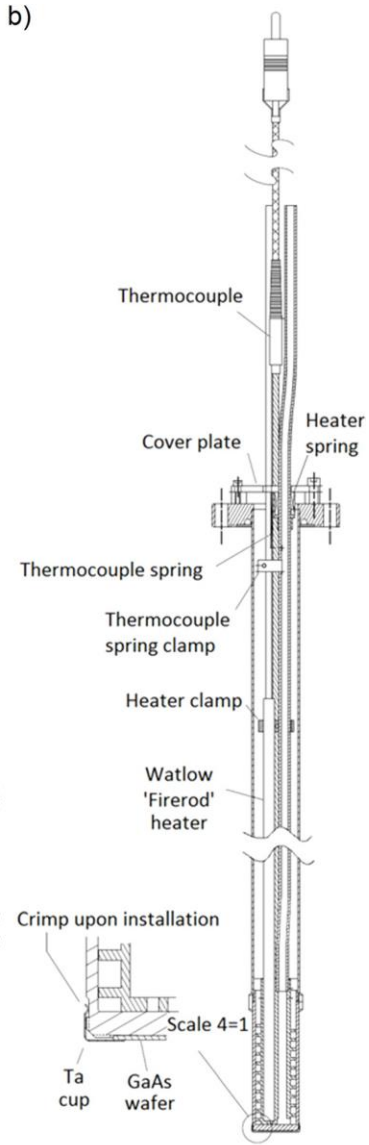
a) Photograph of the bialkali-antimonide photocathode deposition chamber with effusion-type alkali dispenser (shown in the inset),
b) Schematic of the substrate holder assembly with substrate heater,
c) Photograph of the QE scanner system with the mirrors attached to the stepper-motor-controlled translation stages,
d) Photograph of the GaAs substrate secured to the sample holder using an annular Ta cup, and
e) Schematic of the effusion-type alkali dispenser used for coevaporation of K and Cs species (with permission from Lawrence S. Cardman).



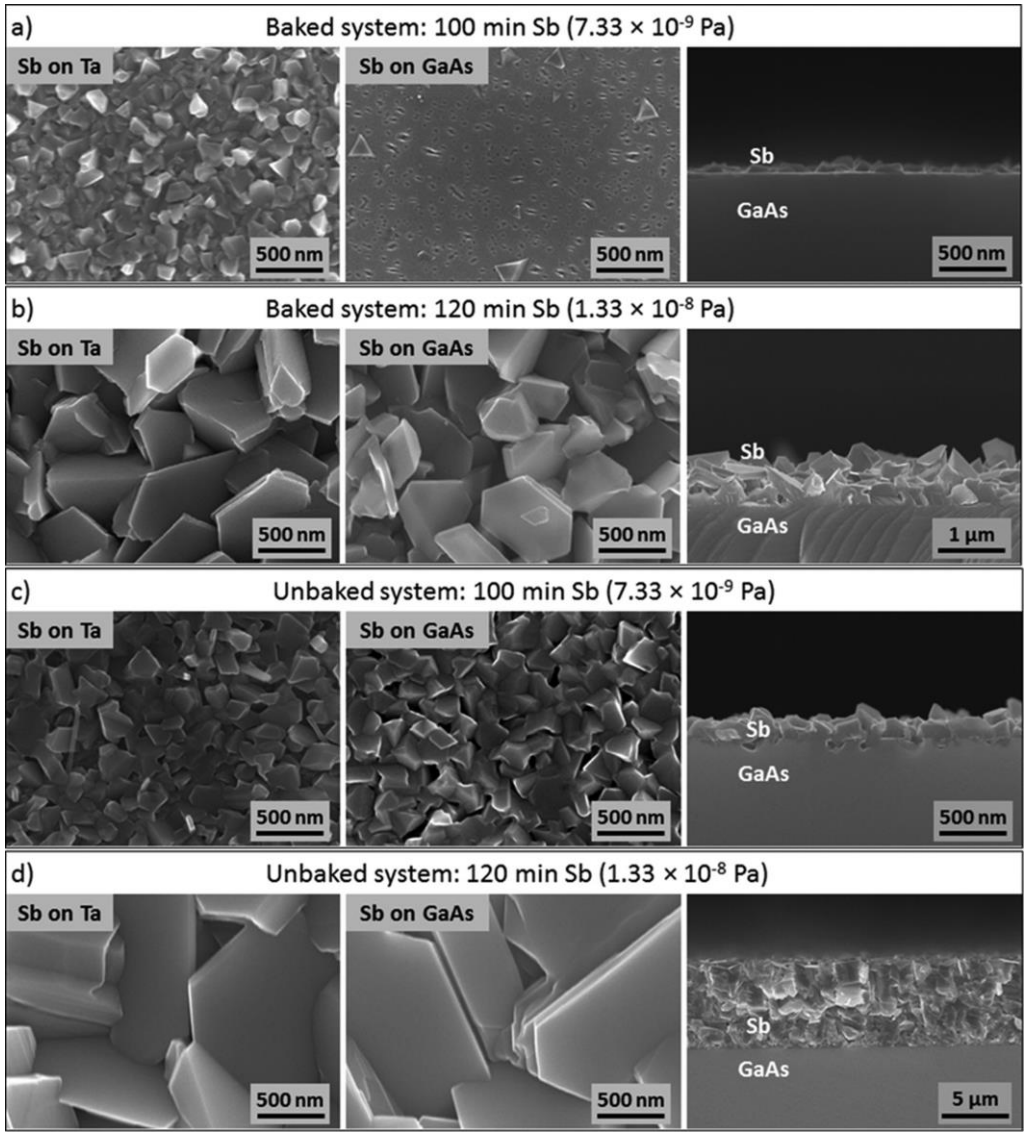








Antimony surface images



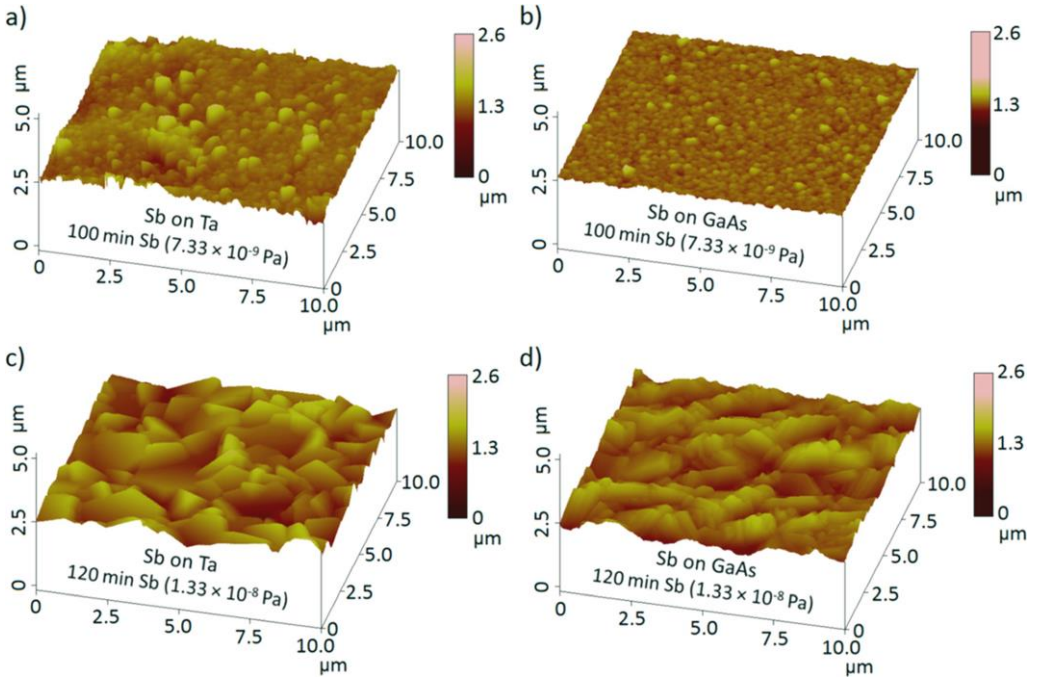
FESEM images illustrating topography and cross-sectional views of Sb films grown on Ta and GaAs substrates for 100 and 120 min deposition times, with 32.7(±00.2) and 33.7(± 0.2) A current, respectively, applied to the Sb crucible heater, under two different vacuum protocols:

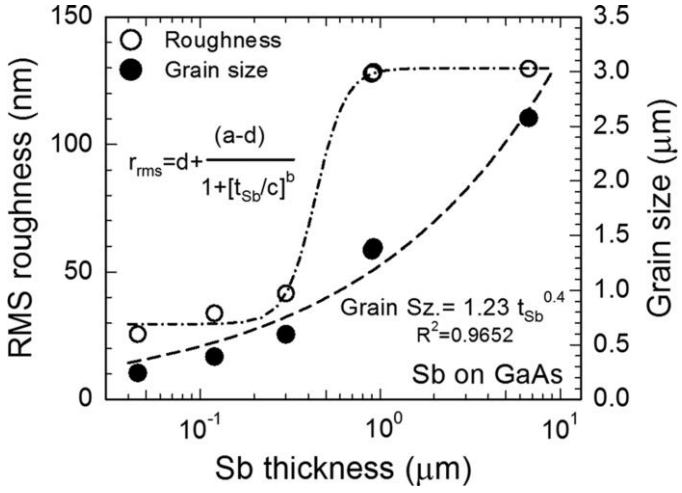
[(a) and (b)] vent-bake and
[(c) and (d)] vent-no bake.

Background pressure:
Vent-bake: ~10^{-7} Pa.
Vent-no bake: ~10^{-6} Pa.

Antimony morphology

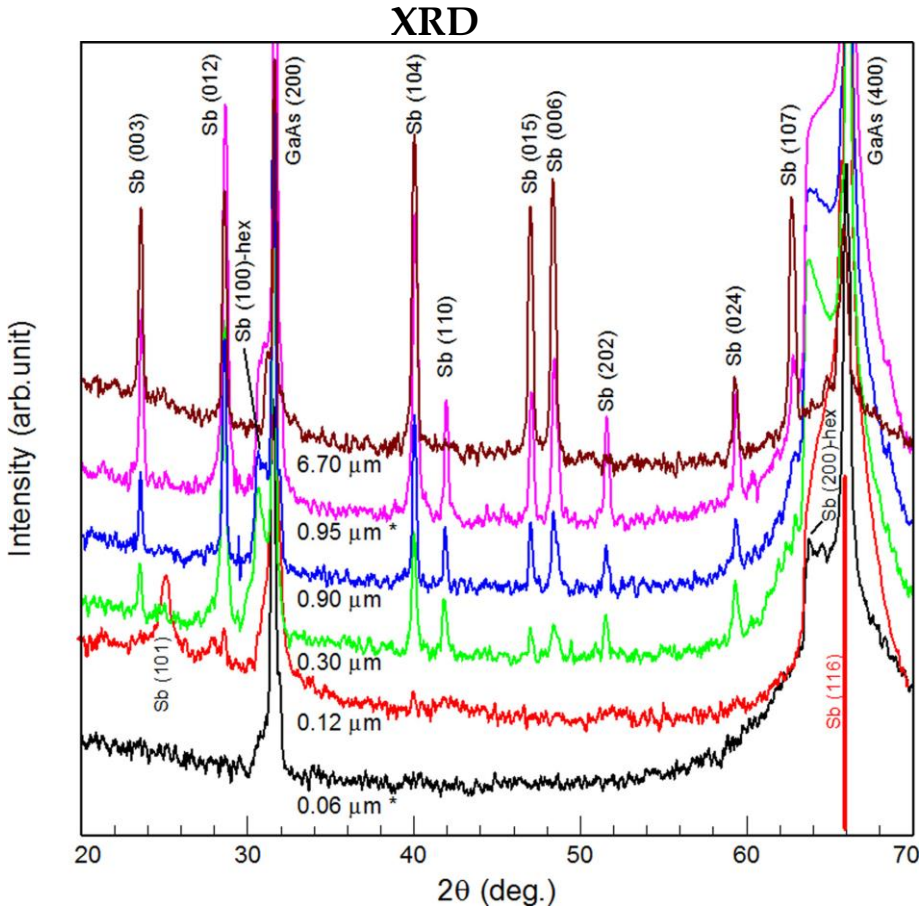
AFM images of Sb films grown on Ta and GaAs substrates for:
[(a) and (b)] 100 min with 32.7(± 0.2) A,
and
[(c) and (d)] 120 min with 33.7(± 0.2) A
current applied to the Sb crucible heater following the vent-no bake protocol.

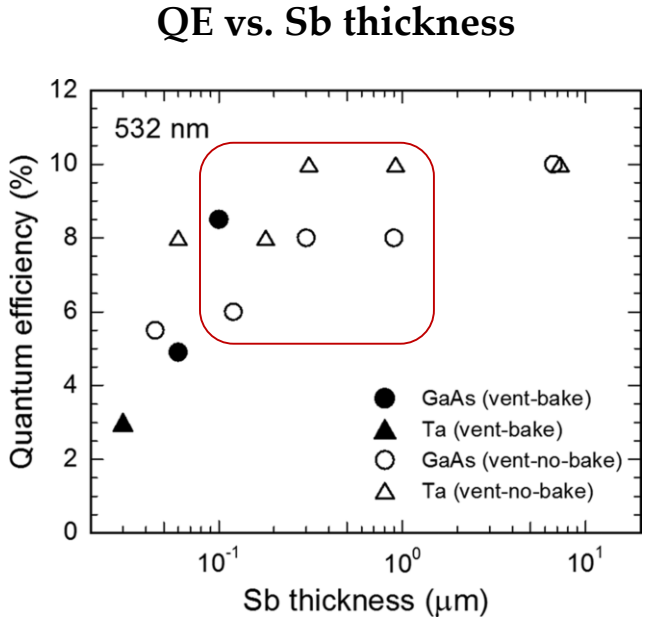




RMS roughness and grain size increased as the Sb thickness increased for Sb films deposited on GaAs substrate

Antimony crystallography and QE





Antimony thickness, µm:	0.06	0.12	0.3	0.9	0.95	6.7
Mean crystallite sizes, nm:	13.6	21.1	19.5	20.3	19.0	53.9

Crystallite size ↑ = e-scattering at grain boundaries ↓ = QE ↑

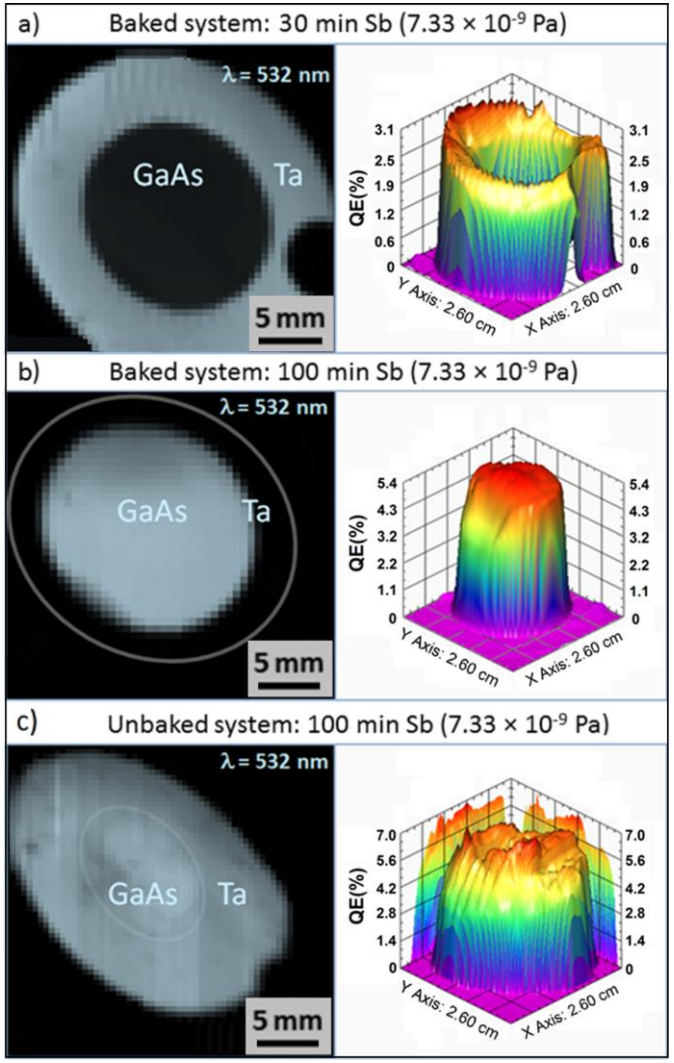
QE distribution

QE (%) map of the photocathodes biased at 284 V with 532 nm (3.96 mW). The photocathodes were prepared by bialkali codeposition following the vent-bake protocol on

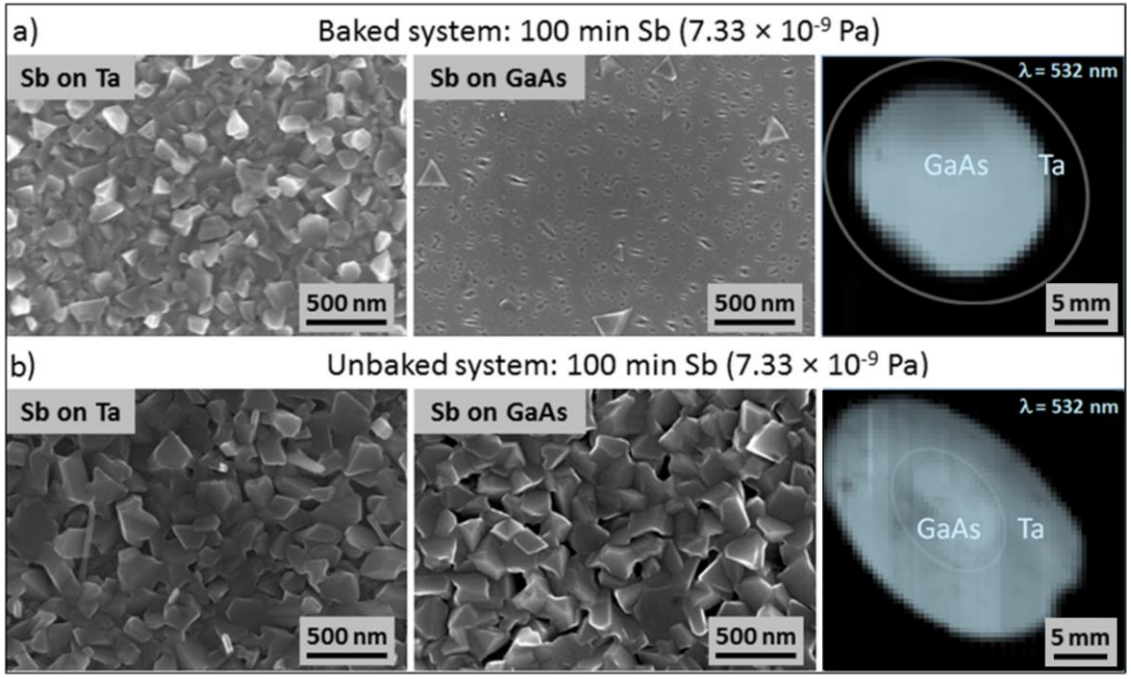
(a) a 30 min grown Sb layer and

(b) a 100 min grown Sb layer; and following the vent-no bake protocol on

(c) a 100 min grown Sb layer. The Z axis in the 3D plot represents QE (%).

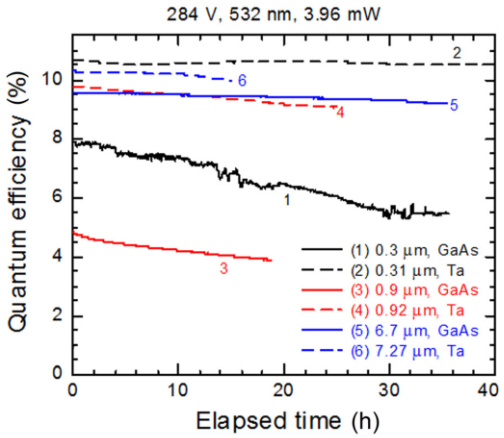


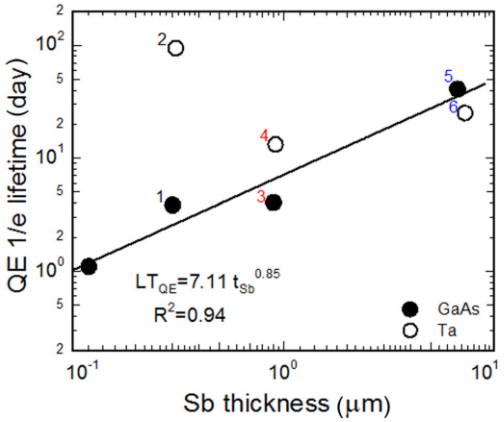
QE and Sb topography

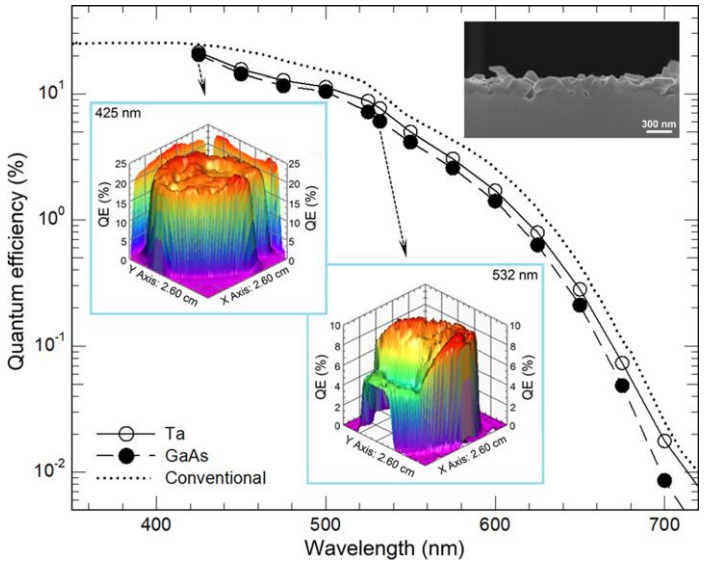


FESEM topography and QE distribution of the photocathodes biased at 284 V with 532 nm (3.96 mW). The photocathodes were prepared by bialkali codeposition following the
(a) vent-bake protocol on a 100 min grown Sb layer and
(b) vent-no bake protocol on a 100 min grown Sb layer

QE lifetime, and spectral response







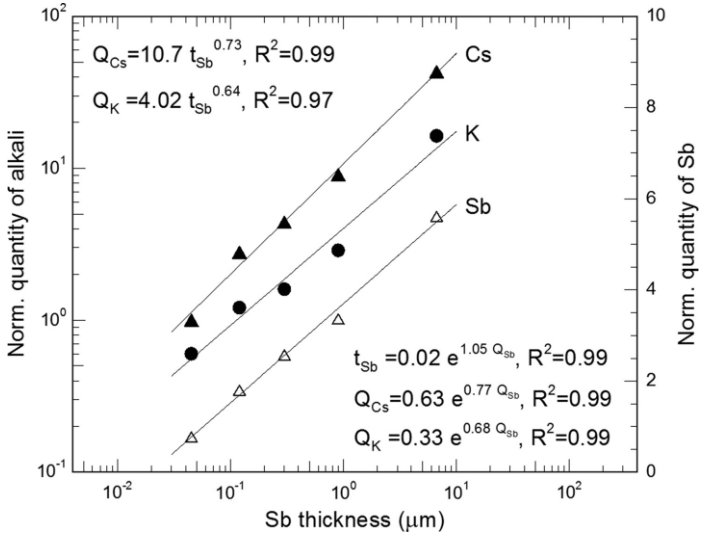
$$\text{Norm } A = \frac{t_{Ai} \cdot PP_{Ai}}{t_{Ao} \cdot PPAo}$$

t = time of deposition

PP = Partial Pressure

i = trial index

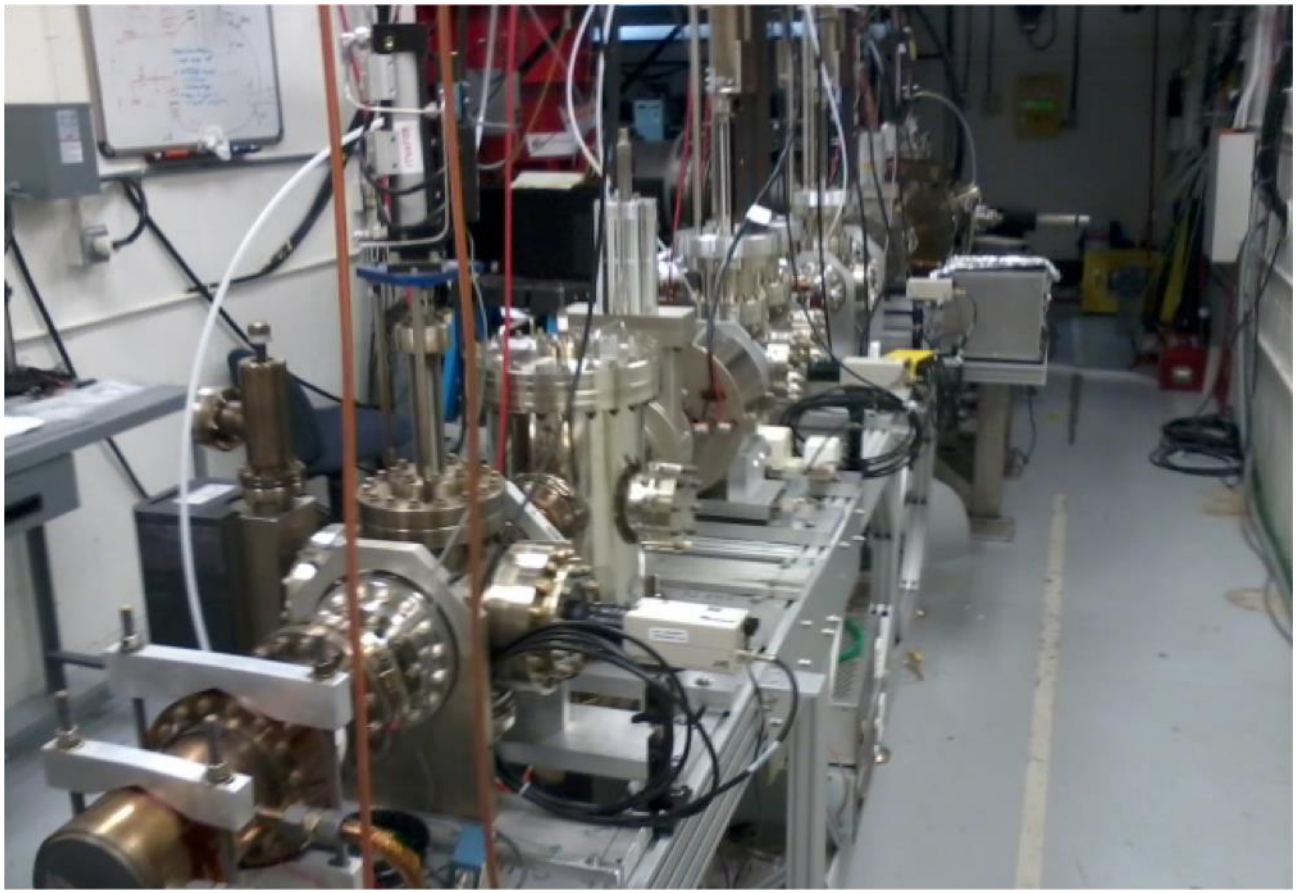
o = benchmark trial's index



Final Remarks

- From the outgassing experiments, a-Si thin film is identified as hydrogen diffusion barrier and TiN film has low pumping speed that can improve vacuum and eliminate high temperature bake of chamber after venting to air.
- From the field emission experiments, TiN thin film on Al electrode is identified to make cheap electrode showing no field emission from cathode electrode under high voltage and electric field strength.
- From photocathode experiments, optimal growth conditions for high QE bialkali antimonide photocathode with longer lifetime are successfully identified.
- A new GTS is constructed at Jefferson Lab to investigate the photocathode performance under real operational condition.
- Implementation of all three studied approaches has potential to meet the next generation photogun requirements by improving vacuum and elimination of field emission at high voltage to ensure extended operational lifetime of a high QE photocathode.

GTS



First beam at GTS

Thank You

Welcome for Q&A