Overview of photocathode physics*

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Outline

- **Where we come from**
- **Major trends to watch**
- **Some limits & their physics**

(*) Disclaimer: this is not a comprehensive overview, personal biases are injected at will!

Recent workshops

http://www.lns.cornell.edu/Events/Photocathode2012/

PHOTOCATHODE PHYSICS FOR PHOTOINJECTORS
CORNELL UNIVERSITY, OCTOBER 8–10, 2012

Large-Area
Picosecond Photo-Detectors
Project

http://psec.uchicago.edu/workshops/

LIGHT 14
Workshop on the Latest Developments of Photon Detectors
Ringberg Castle, Tegernsee
06 - 10 October 2014

https://conference.mpp.mpg.de/light-14/

P3@LBNL

http://faculty.virginia.edu/PSTP2013

The 2013 International Workshop on
Polarized Sources, Targets & Polarimetry

Photocathode Physics
for Photoinjectors
October 12-14, 2010 • Brookhaven National Laboratory

http://www.bnl.gov/pppworkshop/

EuroFEL
FREE ELECTRON LASERS OF EUROPE
Workshop on Photocathodes for RF Guns
1–2 March 2011, INFN of Lecce, Italy

http://photocathodes2011.eurofel.eu/

Recent overviews & tutorials

D. Dowell et al., “Cathode R&D for future light sources”, NIM A 622 (2010) 685 http://dx.doi.org/10.1016/j.nima.2010.03.104

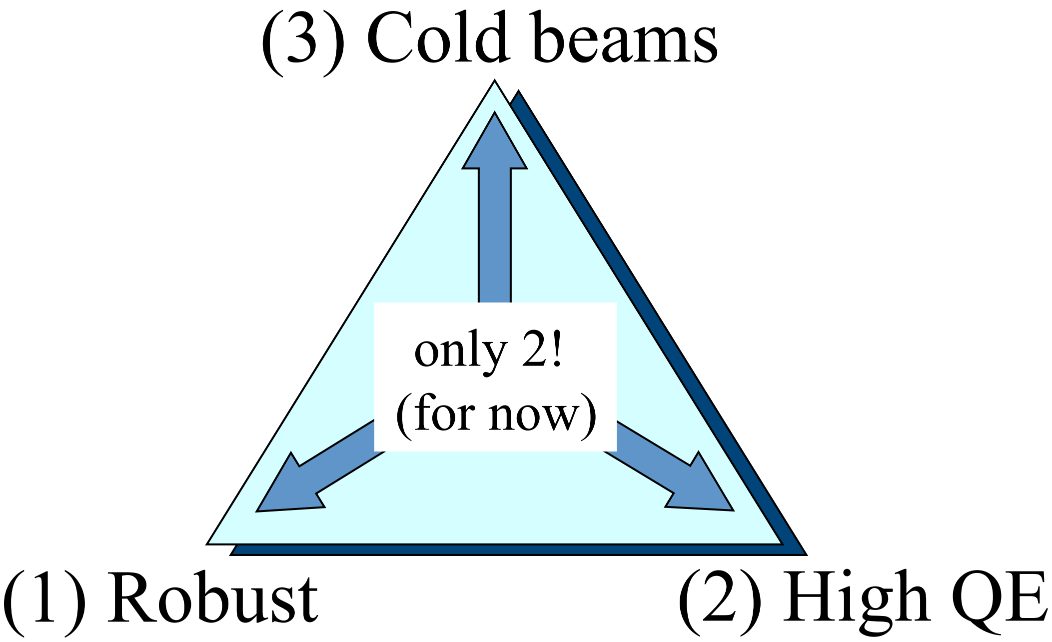
L. Cultrera, “Cathodes for photoemission guns”, PAC2011, http://accelconf.web.cern.ch/AccelConf/PAC2011/papers/thocn1.pdf

J. Smedley and M. Poelker, “Cathode Physics”, USPAS2012 course, http://uspas.fnal.gov/materials/12UTA/UTA_Cathode.shtml

An Engineering Guide to Photoinjectors, eds. T. Rao and D. Dowell (2014) http://arxiv.org/abs/1403.7539

L. Cultrera, “Advances in photocathodes for accelerators”, IPAC2014, http://accelconf.web.cern.ch/AccelConf/IPAC2014/papers/mozb02.pdf

Photocathode research triangle



(3) Cold beams

only 2!
(for now)

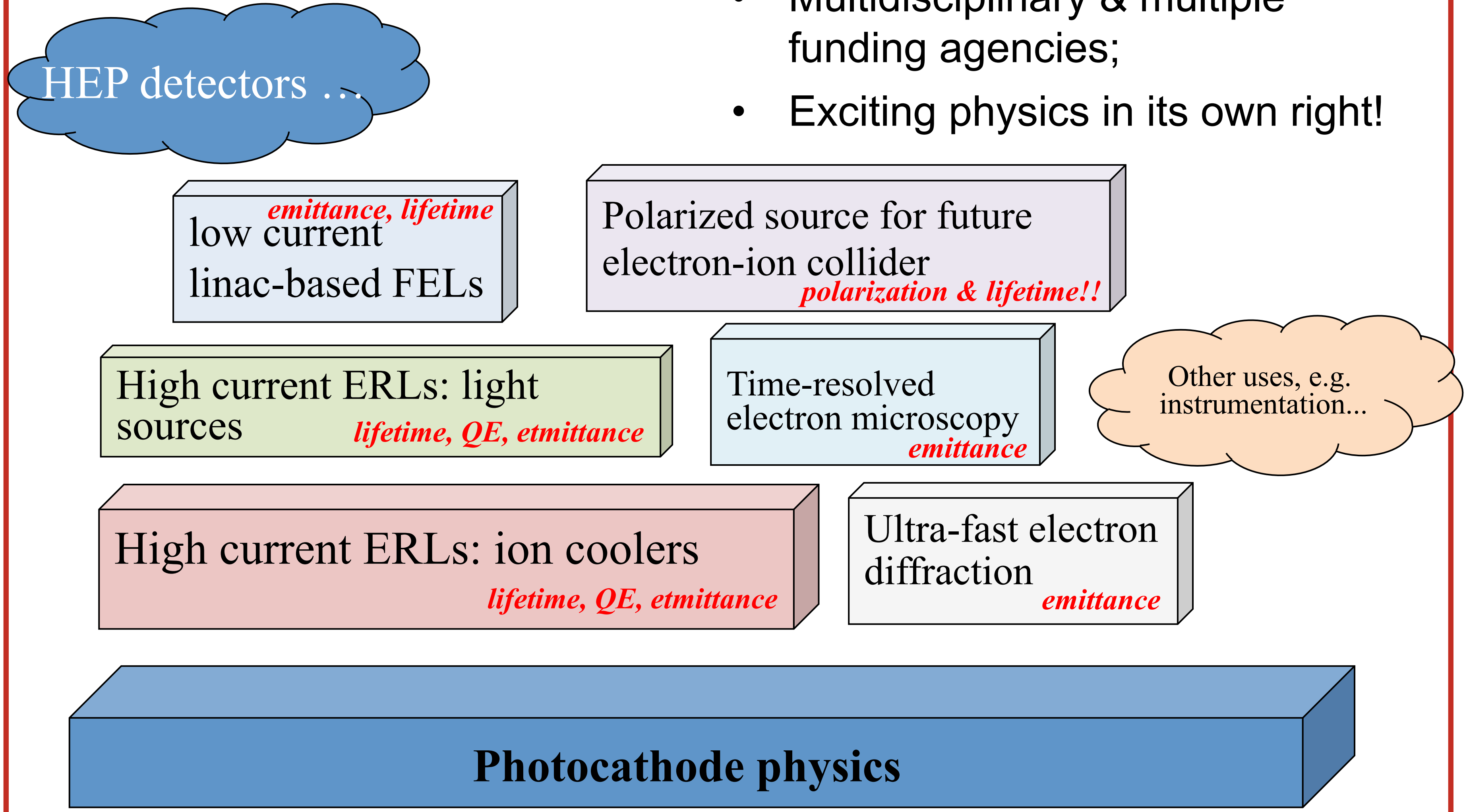
(1) Robust

(2) High QE

Not all application care equally for all three

Know your clientele

- Multidisciplinary & multiple funding agencies;
- Exciting physics in its own right!



HEP detectors …

low current linac-based FELs *emittance, lifetime*

Polarized source for future electron-ion collider *polarization & lifetime!!*

High current ERLs: light sources *lifetime, QE, etmittance*

Time-resolved electron microscopy *emittance*

Other uses, e.g. instrumentation...

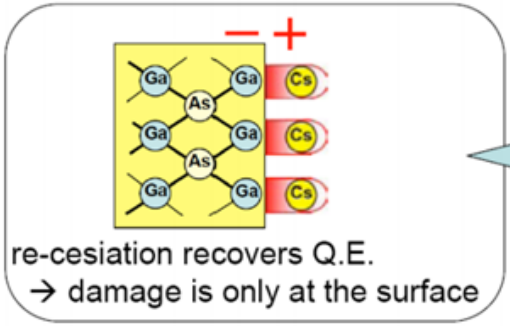
High current ERLs: ion coolers *lifetime, QE, etmittance*

Ultra-fast electron diffraction *emittance*

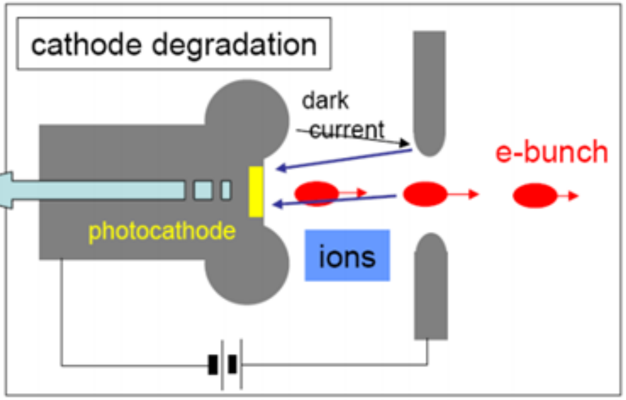
Photocathode physics

(1) Cathode lifetime

- High current tests at Cornell and JLAB: working solutions for DC guns;



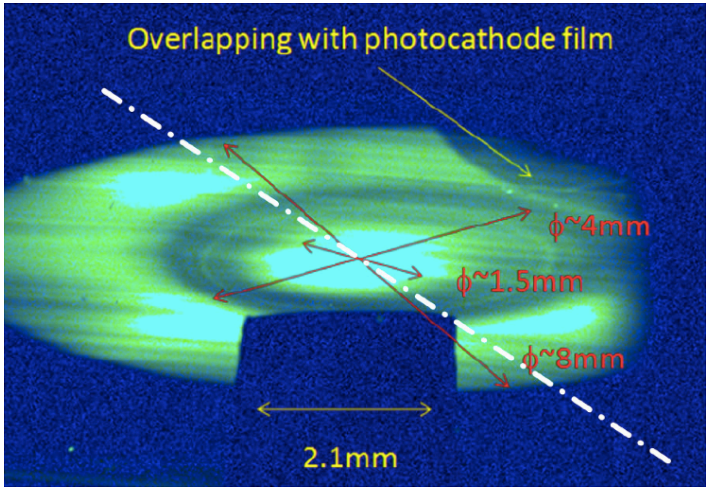
re-cesiation recovers Q.E.
→ damage is only at the surface

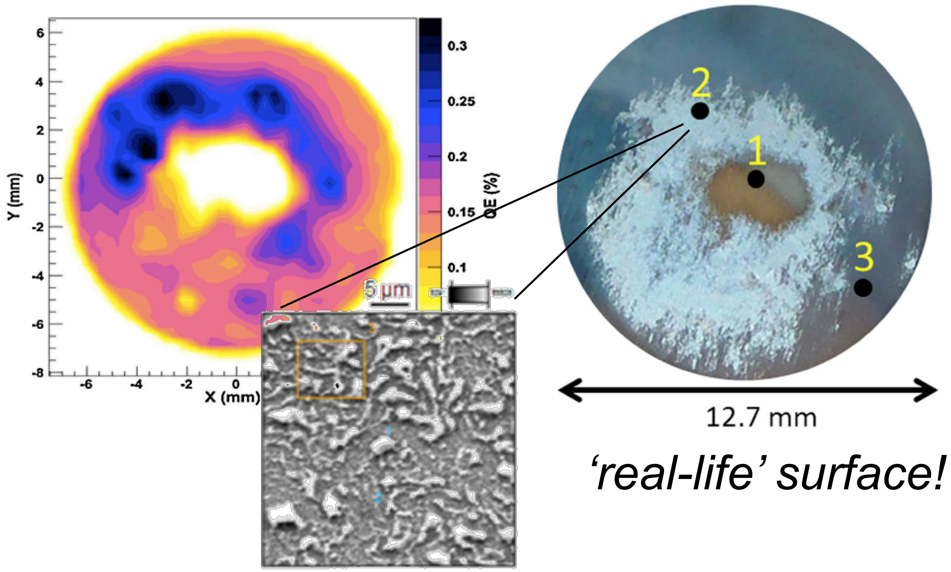


- collision, deposition of residual gases
- dark current (and its enhancement)
- ion back bombardment

- Systematic materials studies afterwards to understand the nature of damage.

Large area structural damage



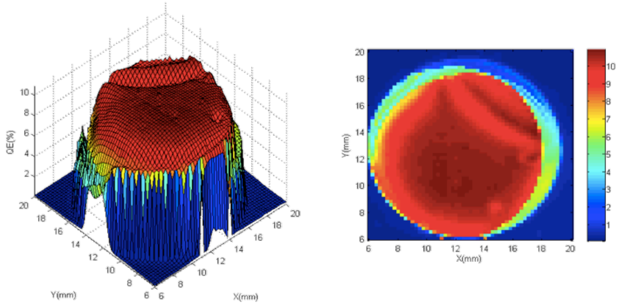


'real-life' surface!

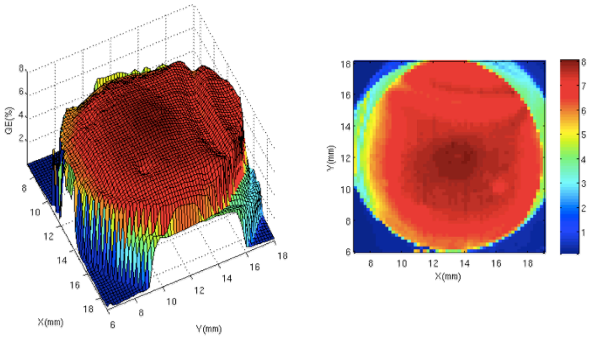
L. Cultrera et al., *PRSTAB* **14** (2011) 120101

R. Mammei et al., *PRSTAB* **16** (2013) 033401

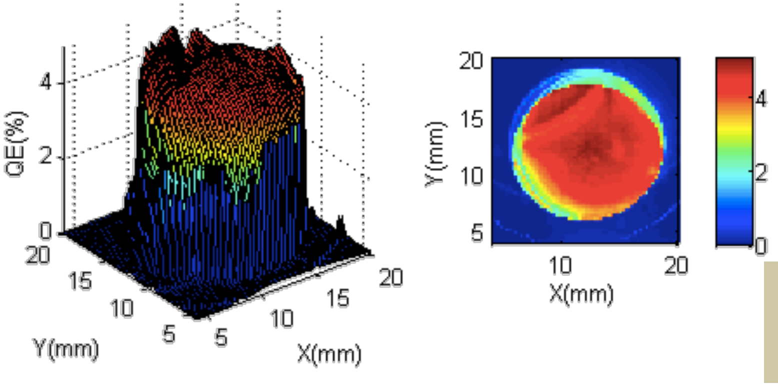
Alkali-antimonides: low current lifetime



QE map on 03 Feb 2012 ~10%



QE map on 03 Apr 2012 ~8%

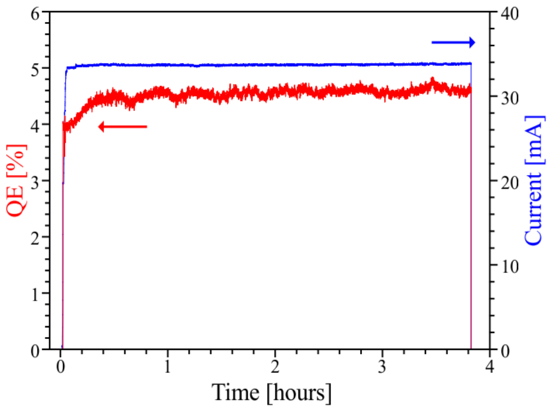


QE map on 02 Oct 2012 ~5%

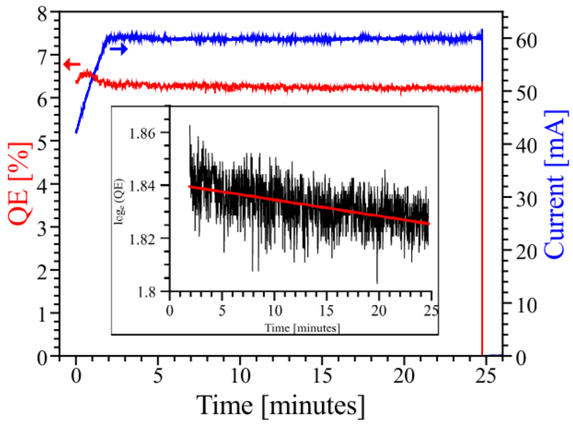
QE 1/e lifetime ~13 months

non-continuous low current (<mA) operation with minor abuses to the gun

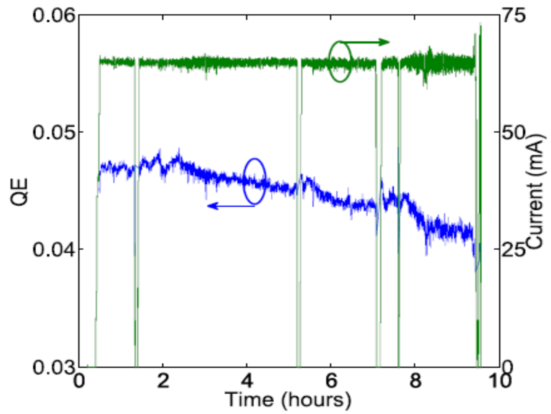
Alkali-antimonides: 30-75 mA current lifetimes



Cs_3Sb
QE @ 520 nm 4%
Max AVG current **33 mA**
Lifetime ≫ 500 C
NO QE DECAY



Cs_2KSb
QE @ 520 nm 6.5%
Max AVG current **60 mA**
Lifetime ≫ 2000 C
1/e QE 30 hr



Na_2KSb
QE @ 520 nm 4.5%
Max AVG current **65 mA**
Lifetime ≫ 2000 C
1/e QE 66 hr

Alklai antimonide based photocathode have been extensively tested in DC gun of the ERL injector prototype at Cornell University. MTEs, response time, QEs and lifetimes at high current are **compatible with the operation of an ERL (user) facility.**

B. Dunham et al., *Appl. Phys. Lett.* **102** (2013) 034105

L. Cultrera et al., *Appl. Phys. Lett.* **103** (2013) 103504

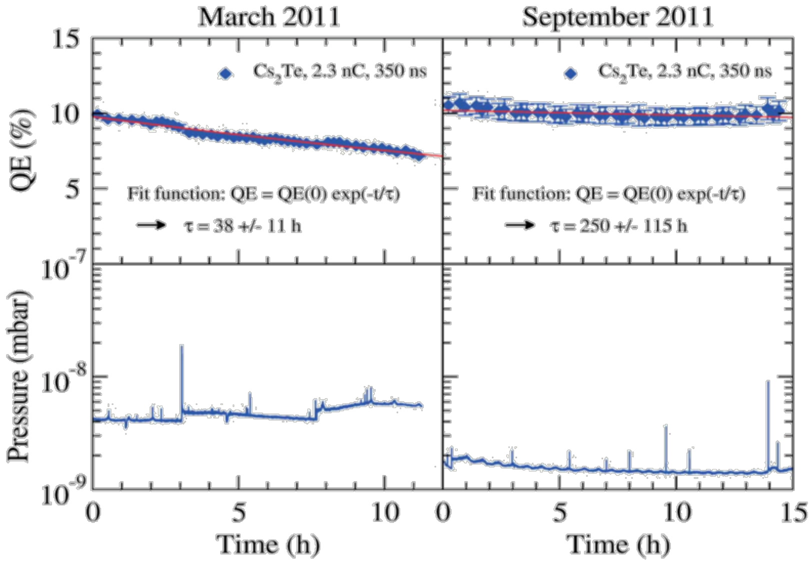
Lifetime challenges

Need to define metrics for 'robustness'!

Cs_3Sb and Cs_2Te Cathodes Prepared and Evaluated in the Same System for the PHIN Photoinjector (Hessler)

- Photocathode planned to replace the nominal thermionic cathode for the CLIC drive beam
- Conclude that Cs_3Sb is about as good as Cs_2Te for this application.
- Vacuum conditions are very important for lifetime, for either photocathode

Impact of Vacuum on Cs_2Te Operating Lifetime



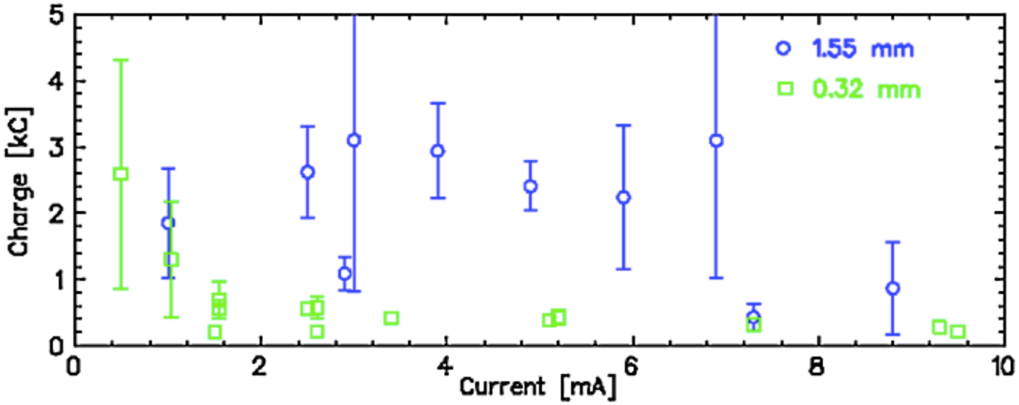
C. Hessler (C. Sinclair), P3@Cornell (2012)

Is lifetime of Cs_2Te really $\gg$ that of Cs_3Sb?

Lifetime challenges (contd.)

- Important for reviews & when comparing performance;
- Robustness metrics: e.g. should specify
  - QE vs. partial pressure of contaminating species;
  - QE vs. temperature of the cathode (bench test);
  - report vacuum (preferably RGA spectrum) for the gun tests;
  - the thickness of the ‘active layer’ & nature of damage;
- Grand challenge: polarized photocathode with similar performance to the antimonides/tellurides?
- Protective coatings?

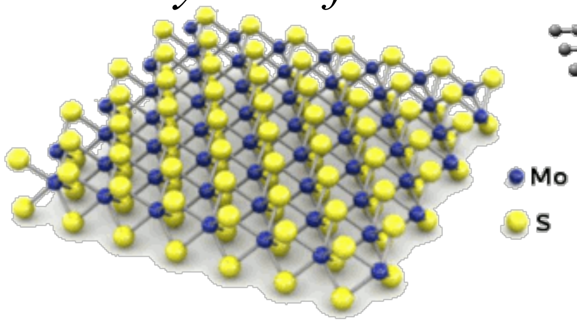
Cs:GaAs measured lifetime



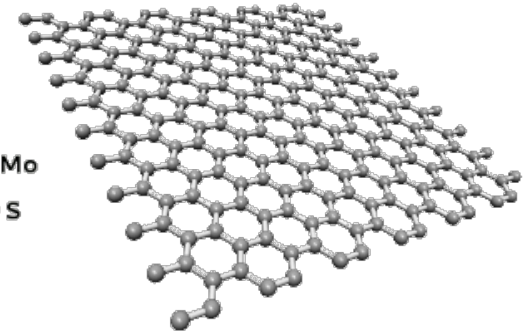
J. Grames et al., *PRSTAB* **14** (2011) 043501

2D materials

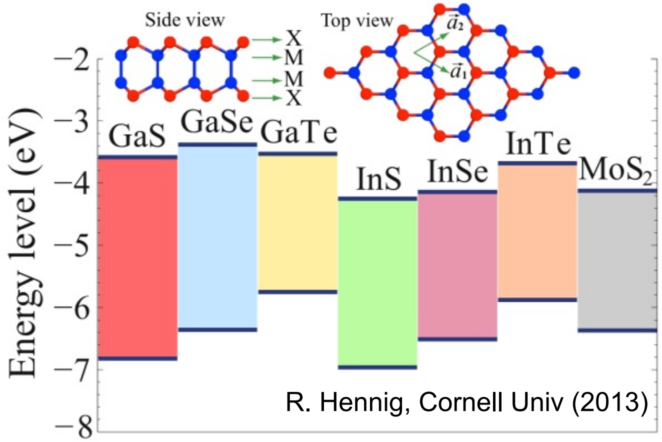
Moly disulfide



Graphene



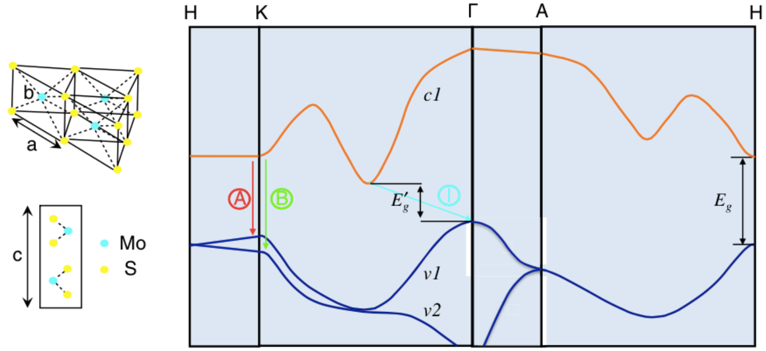
Monochalcogenides



R. Hennig, Cornell Univ (2013)

- Useful to study basic photoemission physics;
  - properties can be tuned to a wide range in heterostructures;
- Protective coatings for traditional photocathodes?

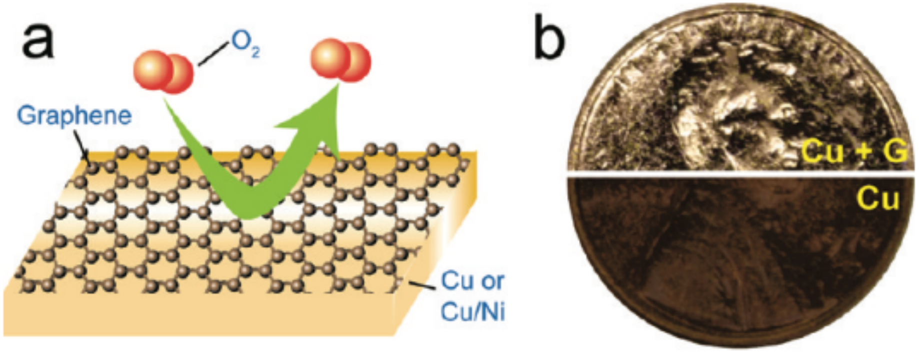
MoS_2: indirect→direct



K. Mak et al., *PRL* **105** (2010) 136805

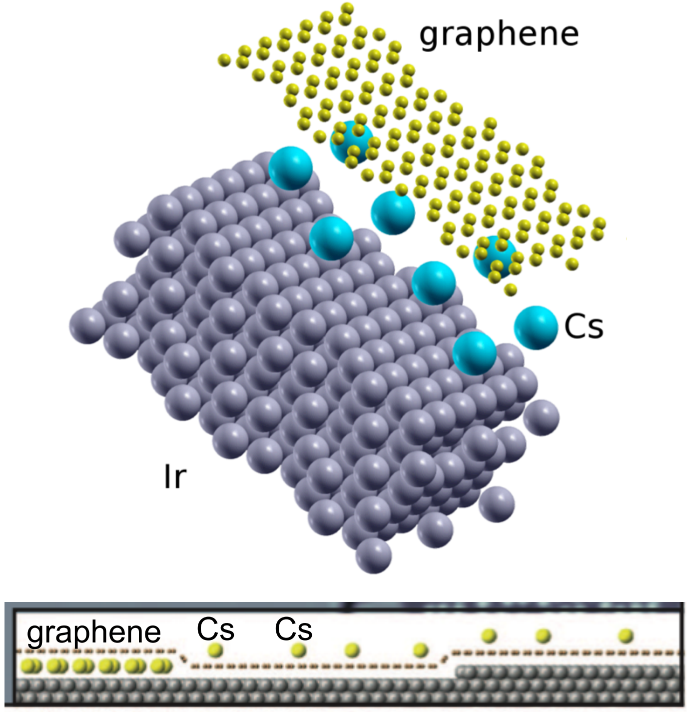
Graphene as a protective layer?

Graphene as chemically inert diffusion barrier



S. Chen et al., *ACS Nano* **5** (2011) 1321

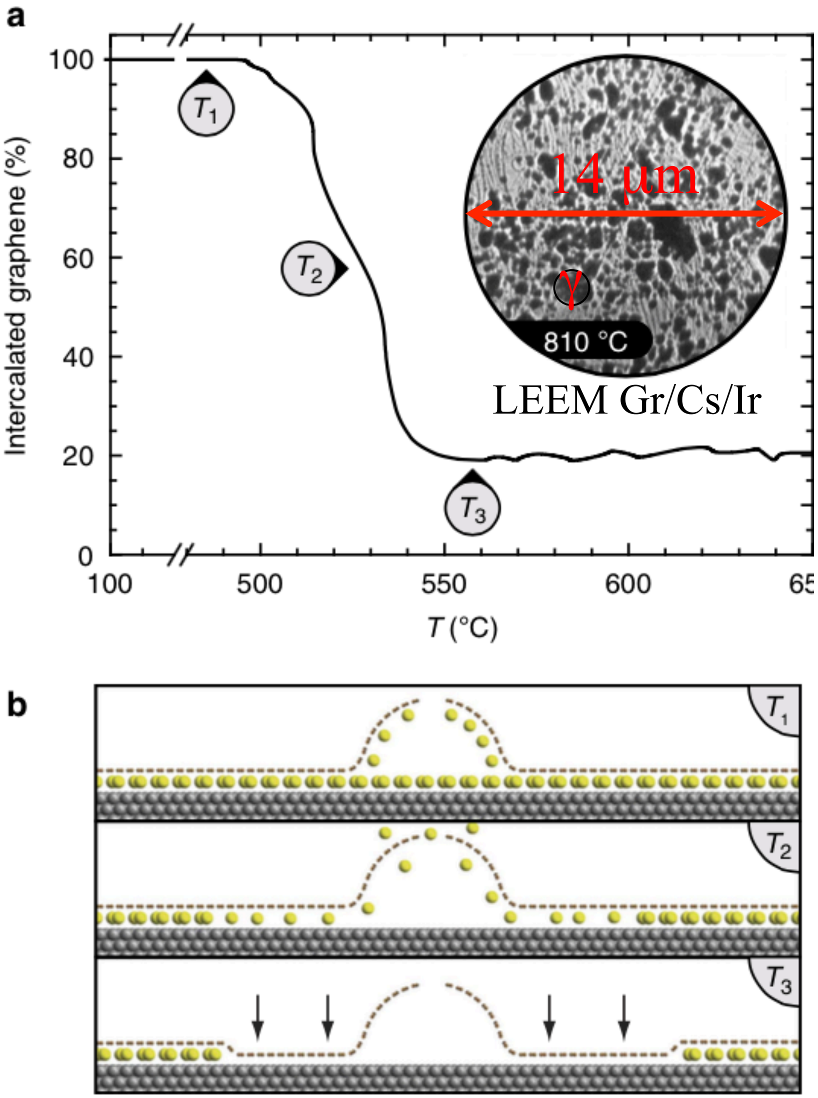
Cesium intercalation of graphene



M. Petrovic et al., *Nature Comm.* **4** (2013) 2772

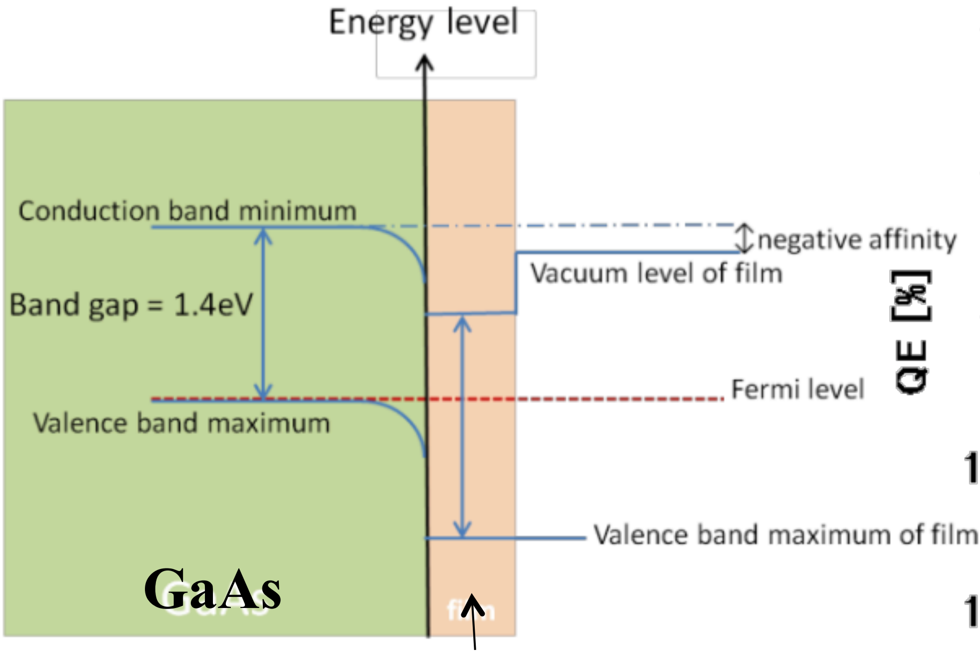
- Graphene wrinkles serve as penetration sites;
- Phase transition vs. Cs coverage with vdW & Coulomb interplay.

Desorption of intercalated Cs at high temperature



M. Petrovic et al., *Nature Comm.* **4** (2013) 2772

Cs$_2$Te + GaAs

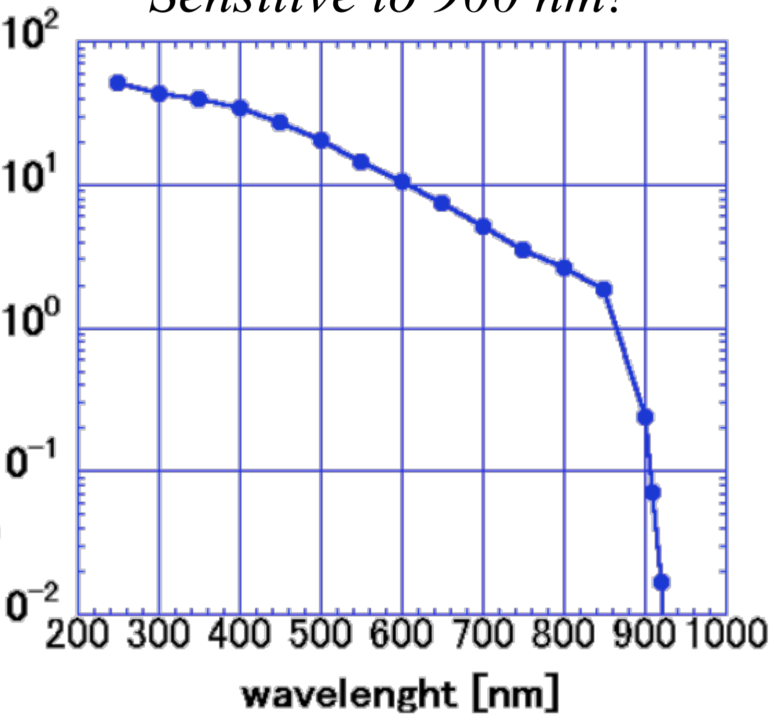


Cs$_2$Te

Te – 0.5 nm

Cs – 200 nm(?)

Sensitive to 900 nm!



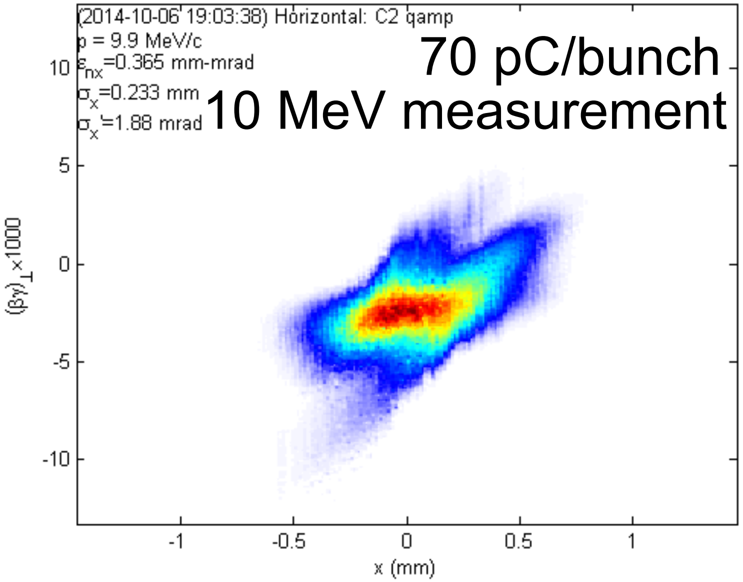
- Exciting result;
- Need to characterize lifetime;
- What will happen to the polarization?

K. Uchida et al., IPAC'14 (2014) 664

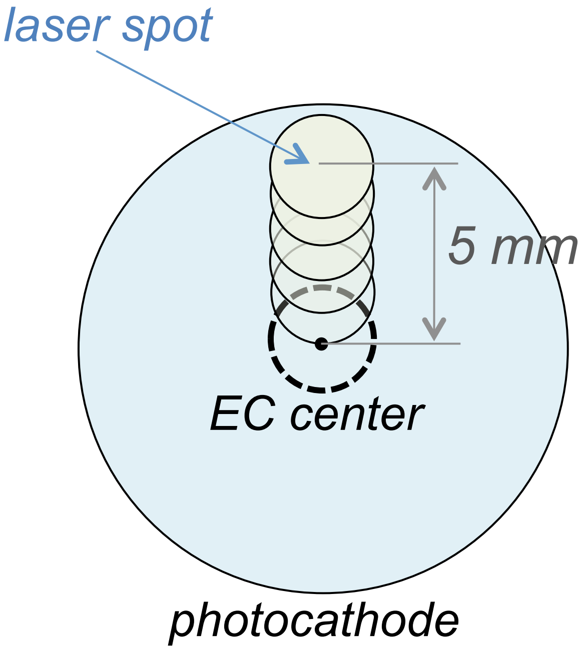
Running laser off-centered in DC gun: no emittance effect

Test: Cornell DC gun photoinjector with laser 0-5 mm off-center

Result: no emittance change 0-4mm, 5% emittance increase at 5mm offset (0.35 → 0.37 mm-mrad)



70 pC/bunch
10 MeV measurement

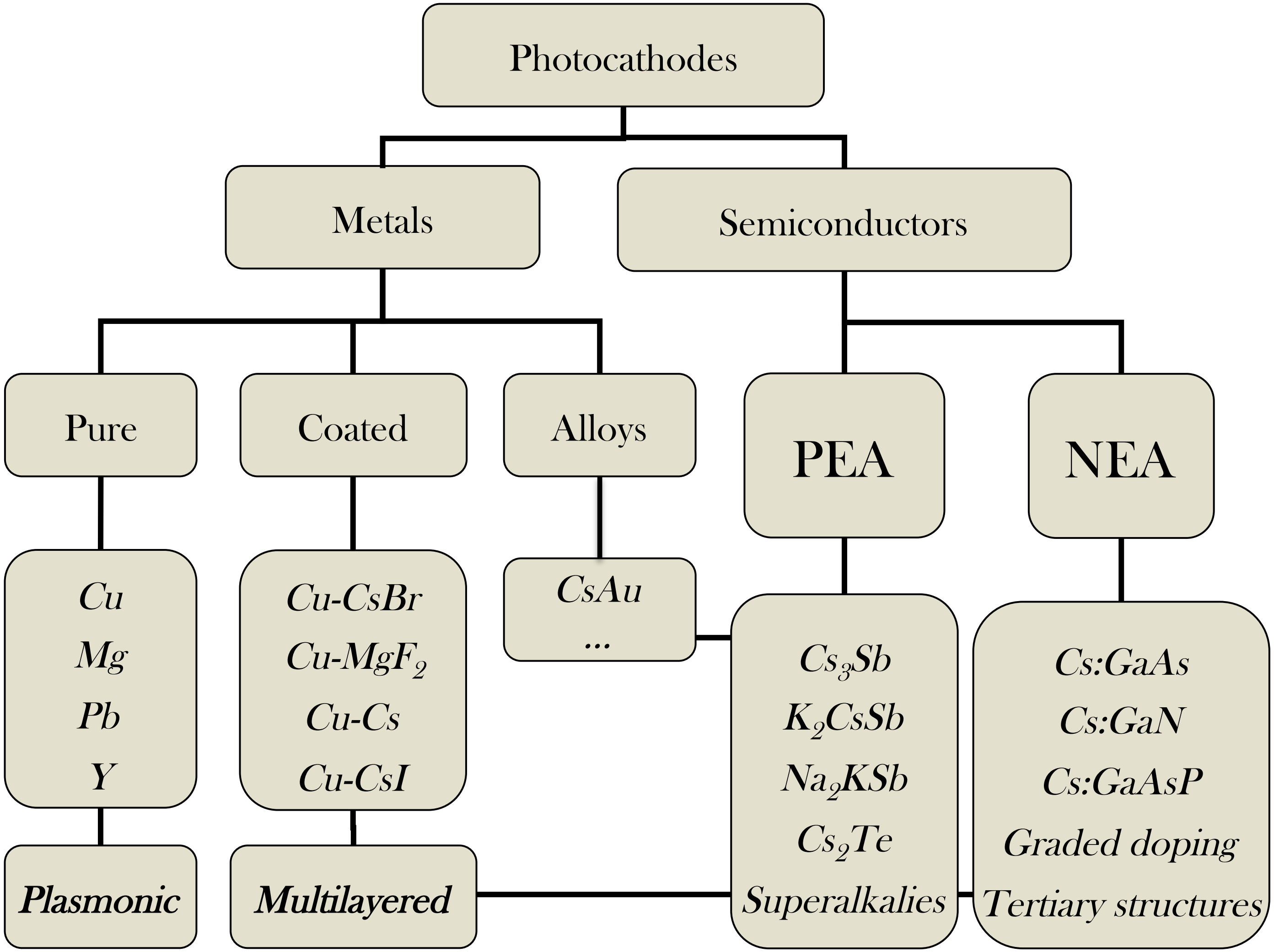


Conclusion: DC gun case is fairly well understood.
Less so for other gun types.

(2) Quantum efficiency & spectral range

- QE does not enter brightness in most of today's beam applications (*only* decides the laser specs);
- A key parameter for some spin-offs (e.g. LAPPD program);
- Still a fundamental parameter to get right (material growth recipes, get to agree with the simulations, etc.);
- Much progress over the last few years in understanding photocathode growth chemistry, refining recipes, manufacturing new structures (e.g. plasmonic, epitaxially grown, etc.).

A Crosscut of Photocathode Materials



Photocathodes

Metals

Semiconductors

Pure

Coated

Alloys

PEA

NEA

Cu
Mg
Pb
Y

Cu-CsBr
Cu-MgF_2
Cu-Cs
Cu-CsI

CsAu
…

Cs_3Sb
K_2CsSb
Na_2KSb
Cs_2Te
Superalkalies

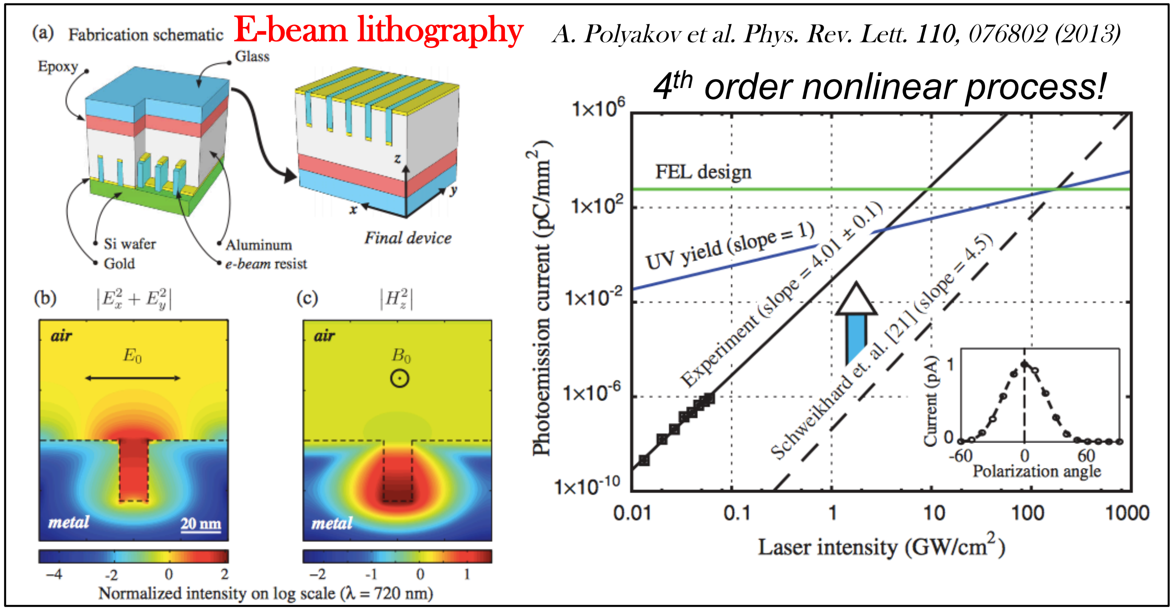
Cs:GaAs
Cs:GaN
Cs:GaAsP
Graded doping
Tertiary structures

Plasmonic

Multilayered

Plasmonic structures: grooves, holes, and such…

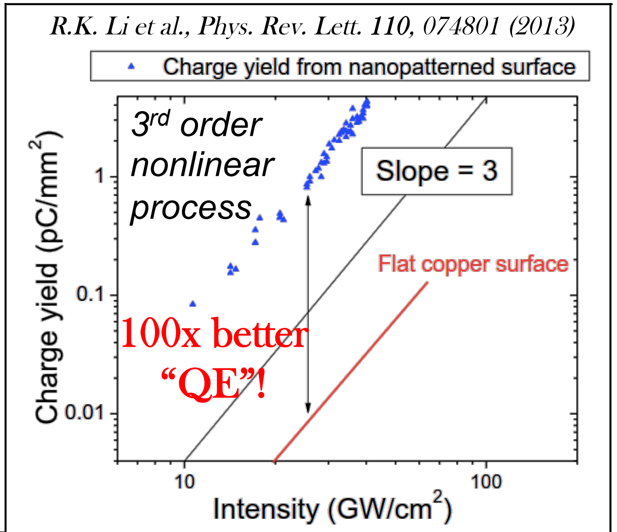
breathing new life into metal cathodes



E-beam lithography

A. Polyakov et al. Phys. Rev. Lett. ***110****, 076802 (2013)*

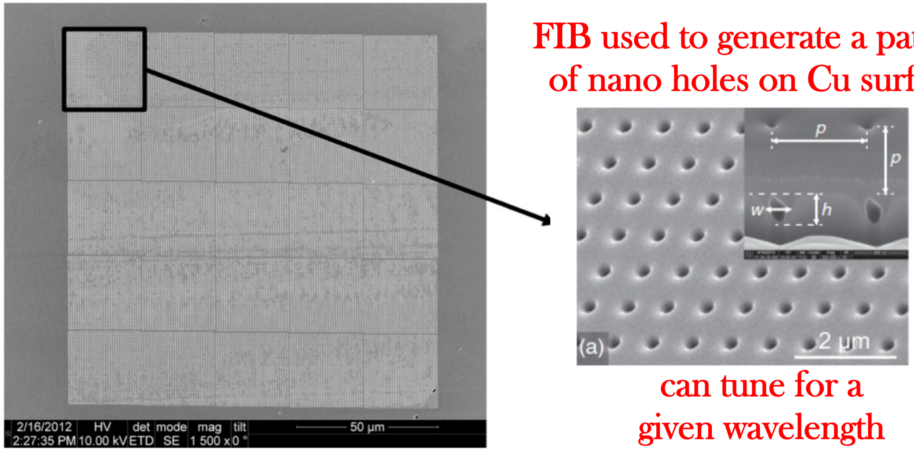
4th order nonlinear process!



R.K. Li et al., Phys. Rev. Lett. ***110****, 074801 (2013)*

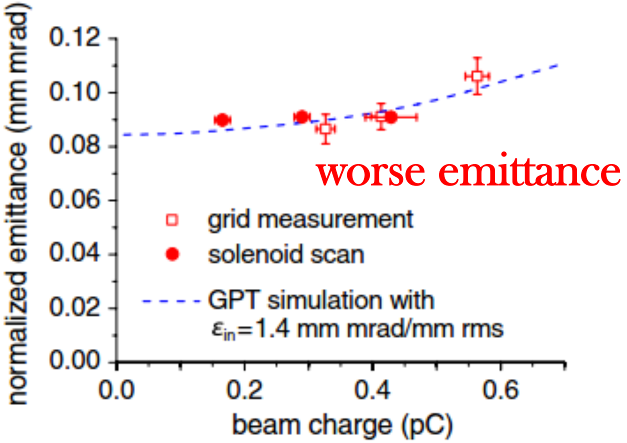
3rd order nonlinear process

100x better "QE"!



FIB used to generate a pattern of nano holes on Cu surface

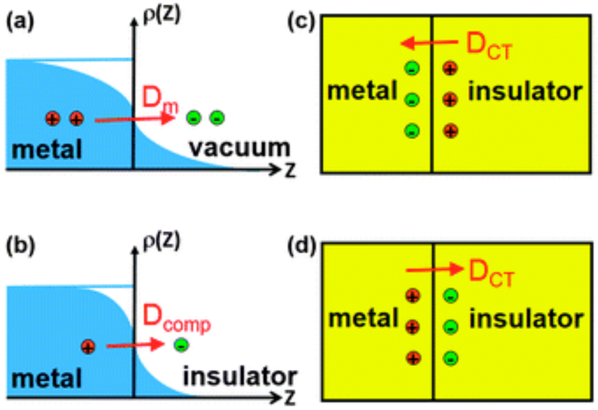
can tune for a given wavelength



worse emittance

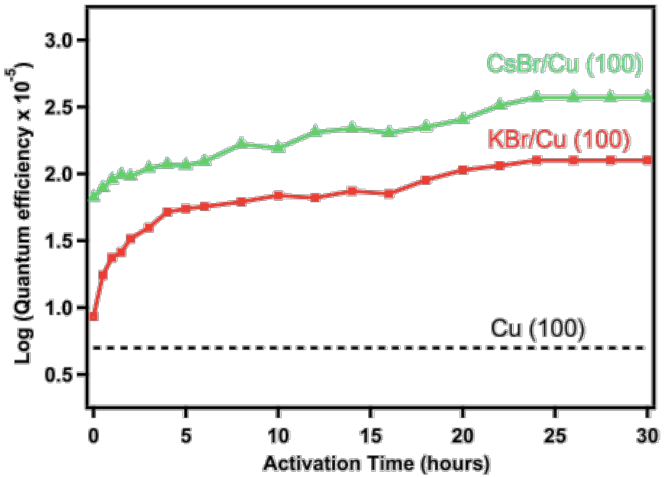
Alkali-halides covered metals: enhancing QE

- Reduce the workfunction;
- Intra-band states of insulating coating;
- Halogen loss over alkali during UV laser activation;

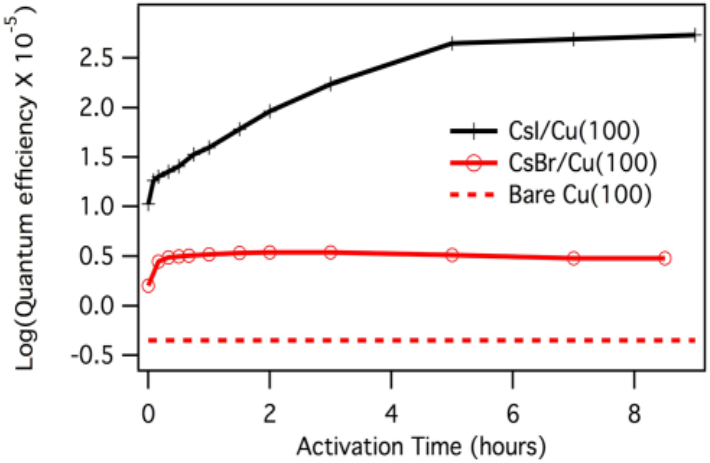


S. Ling et al, *Phys. Chem. Chem. Phys.* **15**, 19615 (2013)

266 nm	KBr	CsBr	CsI
Film thick. (nm)	7	7	8
QE enh. before activation	1.8	14	18
QE enh. after activation	2.6	77	2700
WF before activation (eV)	3.96	3.76	3.68
WF after activation (eV)	3.66	3.41	1.74



W. He et al, *Appl. Phys. Lett.* **102** (2013) 071604

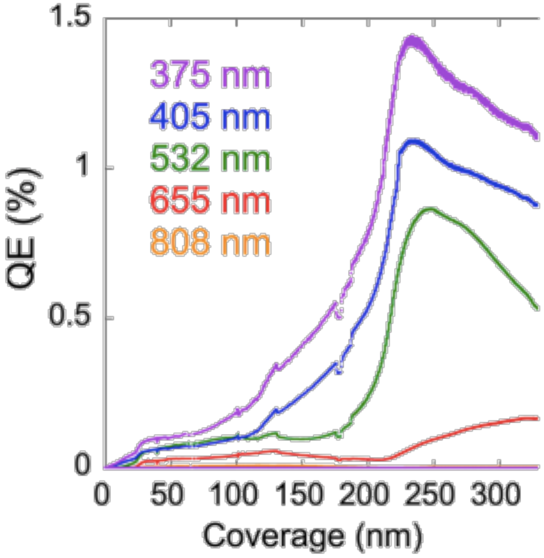


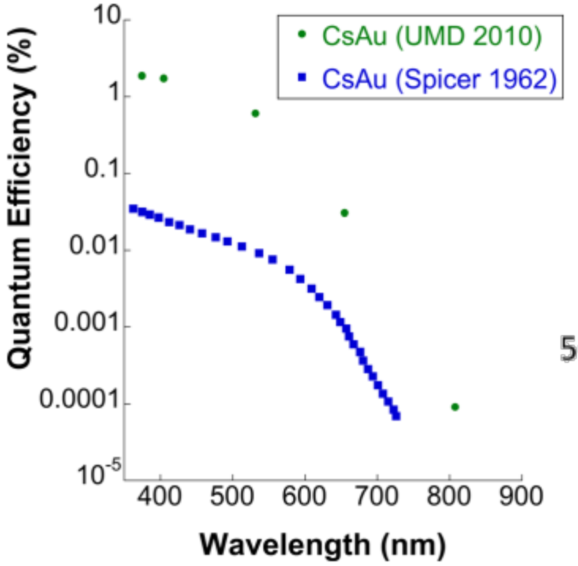
L. Kong et al., *Appl. Phys. Lett.* **104** (2014) 171106

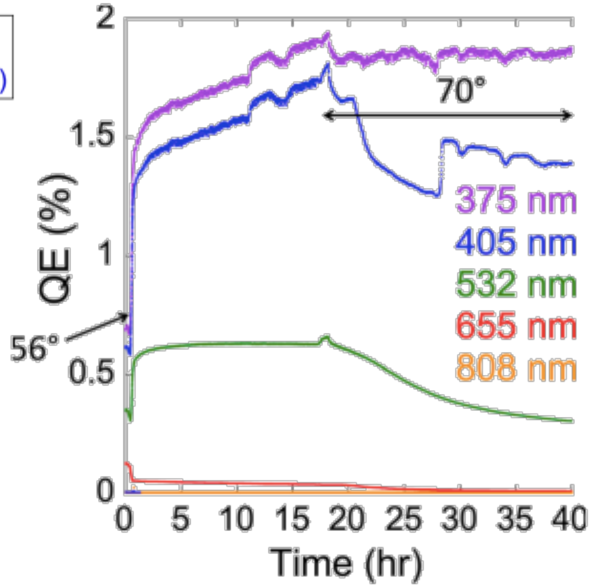
Ceasium Auride

> As was to be expected, the alloys of the Au*M* type are photo-electrically sensitive, but the sensitivity is too low to be of practical importance. An

A. Sommer, Nature **152**, 215 (1943)







S. A. Khan, *J. Vac. Sci. Tech B* 30, 031207 (2012)

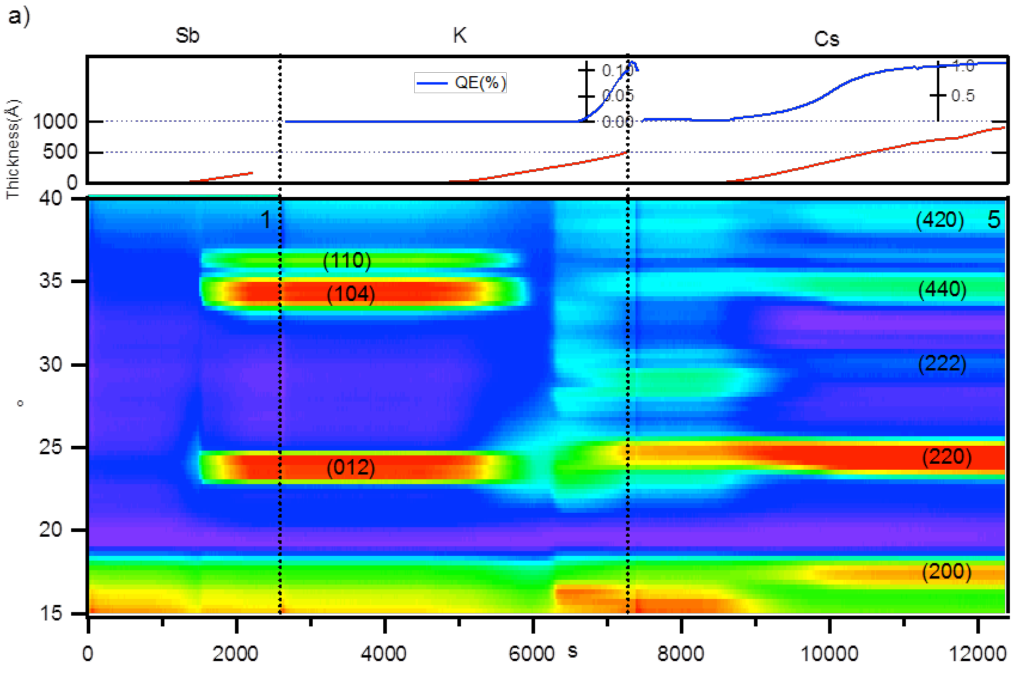
- These results reopen AuM for photocathodes
- QEs in the range of few % can be achieved in the visible
- Lifetime properties at room and moderate temperature are encouraging
- MTE, Response Time yet to be characterized

Alkali Antimonide Family

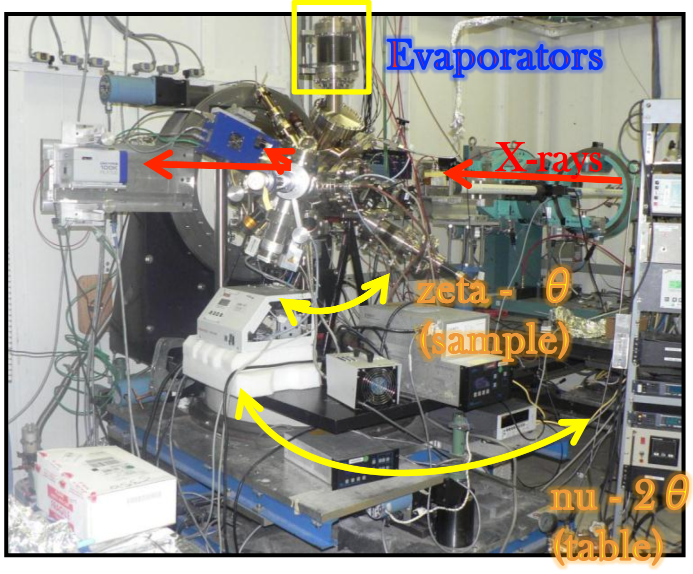
- Most practical choice when high QE is required;
- Well characterized (accelerator performance);
- Interesting challenges ahead:
  - Generating atomically flat surfaces?
  - Getting ultralow emittance (at cryogenic T);
  - Commercialization! (cathode in a can? suitcase next-day shipping?)
- Refining of the recipes and fabrication procedures still on-going:
  - Remove 'human' factor from the growth procedure;
  - Super-alkalies to maximize the QE spectral range to IR for detectors.

Synthesis process now better understood

- 4-axis diffractometer UHV chamber, NSLS & CHESS compatible;
- XRD and GISAXS during growth, high resolution XRD and XRR between growth steps;
- XRD gives reaction chemistry while GISAXS and XRR give roughness.

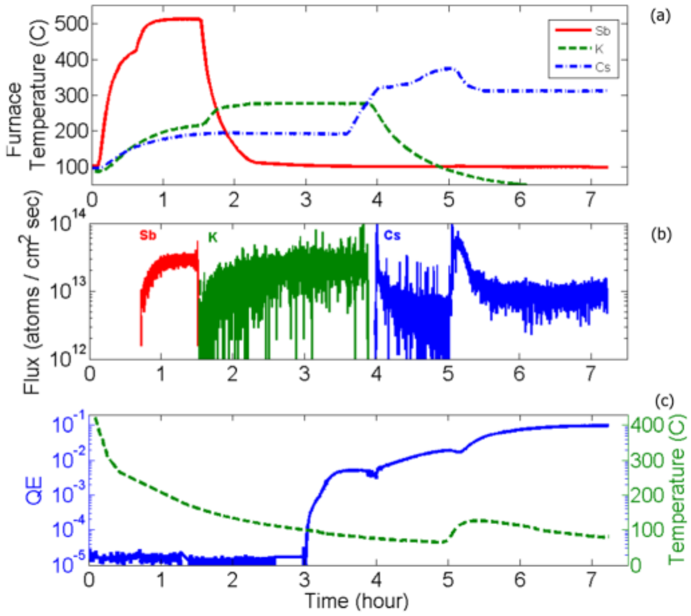


J. Smedley et al., Proceedings of IPAC2013, 464 (2013)



Sb evaporated at 0.2 Å/s
 Room temp,
 Crystallize at 4nm
K deposition dissolves Sb layer
 Film goes amorphous
QE increase corresponds with
 K_3Sb crystallization
Cs increases lattice constant and
 reduces defects

New recipes and sources

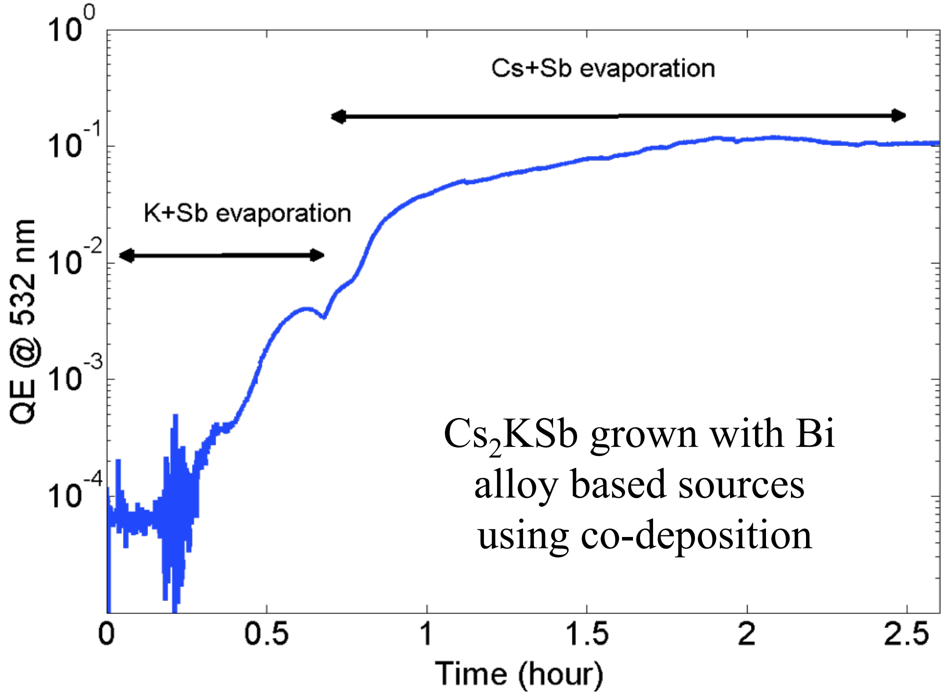


L. Cultrera et al., J. Vac. Sci. Tech. B **32**, 031211 (2014)

- Eutectic alloys;
- Bulk/stoichiometric material sputtering;
- Much progress to be reported at this workshop!

- Alkali Azides (AN_3)

- Pure alkali metals



Cs_2KSb grown with Bi alloy based sources using co-deposition

(3) Cold beam

- Beam ‘coldness’ (temperature $T_\perp$) defined as spread in the transverse momentum^2/m$_e$;
- Mean Transverse Energy (MTE) = same as the momentum spread^2/m$_e$;

$$\frac{\sigma^2_{p_{x,y}}}{m_e} = \langle m_e v_x^2 \rangle = \left\langle \frac{1}{2} m_e v_x^2 + \frac{1}{2} m_e v_y^2 \right\rangle = \text{MTE} = k_B T_\perp$$

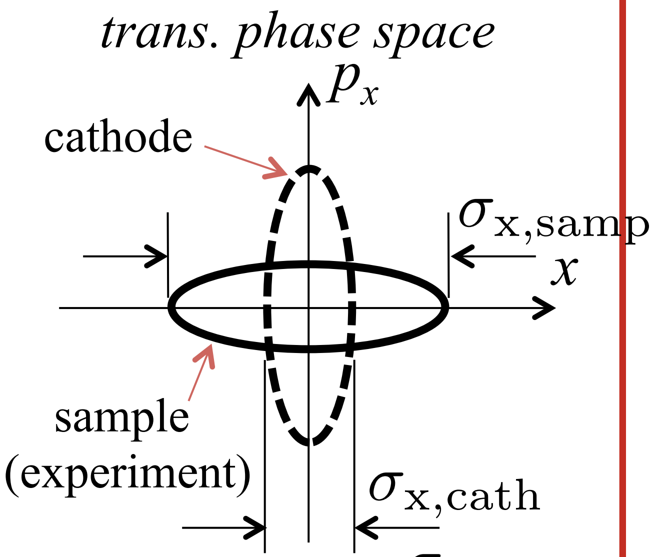
- Normalized emittance:

$$\epsilon_{\mathrm{nx,y}} = \sigma_{\mathrm{x,cath}} \cdot \sigma_{p_{\mathrm{x,y}}} = \sigma_{\mathrm{x,cath}} \sqrt{\frac{k_B T_\perp}{m_e c^2}}$$

- Demagnify beam to reduce $T_\perp$ at the sample:

$$T_{\perp,\mathrm{samp}} = \frac{T_{\perp,\mathrm{cath}}}{M^2}$$

(provided the emittance is conserved)



magnification $M = \dfrac{\sigma_{\mathrm{x,samp}}}{\sigma_{\mathrm{x,cath}}}$

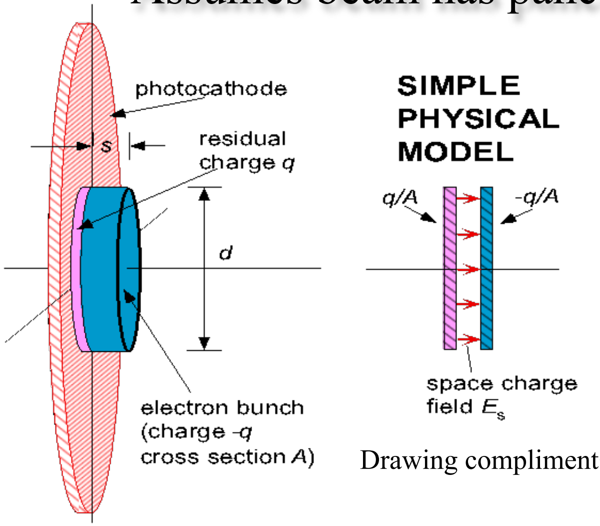
Packing charge into bunches

Assumes beam has pancake shape

- Max charge density for short pulses:

$$\varepsilon_0 E_{\mathrm{cath}}$$

- Max phase space density (a.k.a. transverse brightness):



Drawing compliments of A. Vetter

$$\frac{\text{charge}}{\text{cross-section} \times \text{momentum spread}} = \frac{\varepsilon_0}{m_e c^2} \frac{E^*_{\mathrm{cath}}}{\mathrm{MTE}^*}$$

geometric enhancement factor

$$\mathcal{B}_{4D} \propto \frac{\beta E_{\mathrm{cath}}}{\mathrm{MTE}(\beta E_{\mathrm{cath}})}$$

cathode MTE itself can be a non-trivial function of field, roughness, etc.

Pushing the limits

- Only 2 fundamental parameters to push:

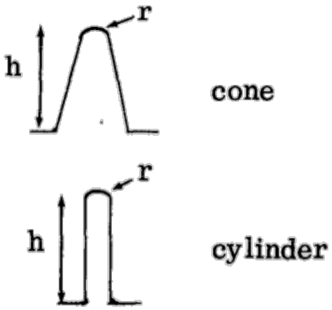
$$\mathcal{B}_{4D} \propto \frac{\beta E_{\mathrm{cath}}}{\mathrm{MTE}(\beta E_{\mathrm{cath}})}$$

MTE ↓

- Can be limited by the non-linear space charge (design your source properly!)
    - ➢ 1-2 meV limit
- Higher $\beta \cdot E_{cath}$ and roughness can be limiting phenomena!
- *Disorder induced heating is the next fundamental limit*
    - ➢ *also on 1-2 meV scale*
- *How does one reliably measure ~meV MTE??*

$\boldsymbol{\beta \cdot E_{cath}}$ ↑

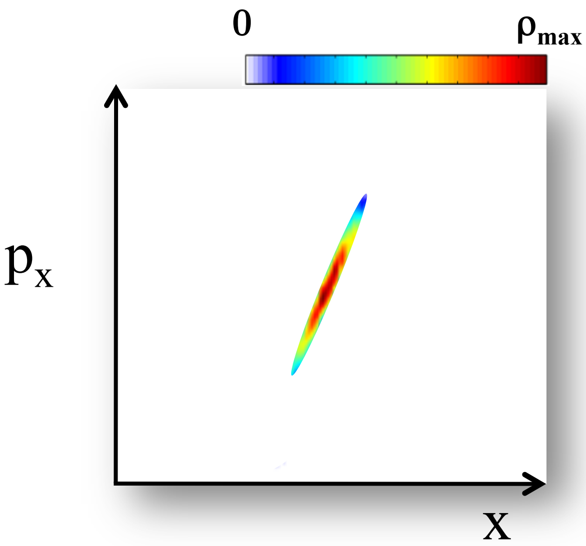
- Geometric enhancement factor: needles & sharp tips;
- A trade-off between photo- and field-emission;
- Needle arrays are great for current, but dilute brightness relative to a single tip.



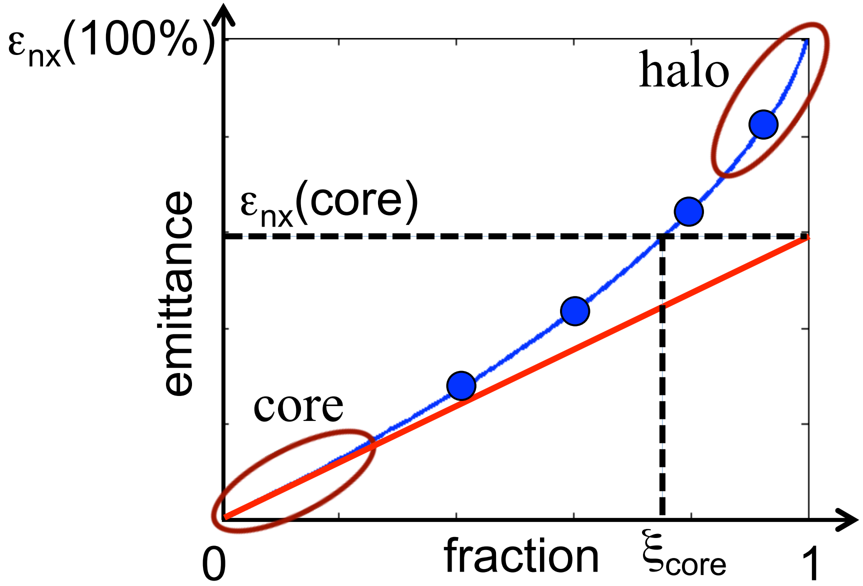
Cylinder: $\beta = \frac{h}{r} + 2$

Cone: $\beta \doteq \frac{1}{2}\frac{h}{r} + 5 \quad \text{for } 20 < \beta < 300$

Core emittance



rms emittance vs. beam fraction: $\varepsilon_{nx}(\xi)$



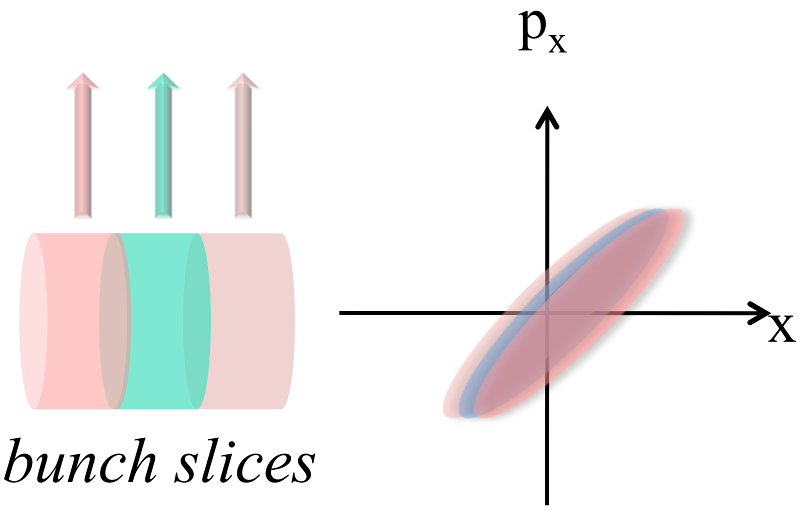
- core emittance is related to the max phase space density
- core fraction: how much of the beam is contained inside the core

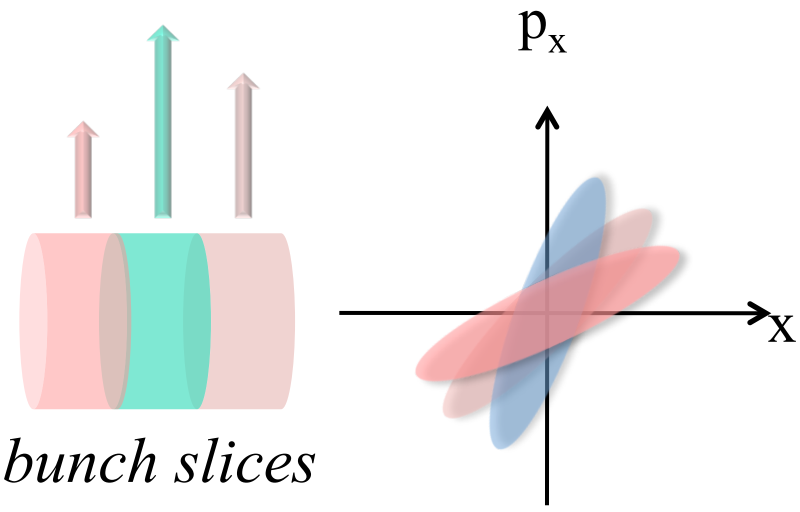
I. Bazarov, PRSTAB 15, 050703 (2012)

C. Lejeune and J. Aubert, Adv. Electron. Phys. Suppl. 13, 159 (1980)

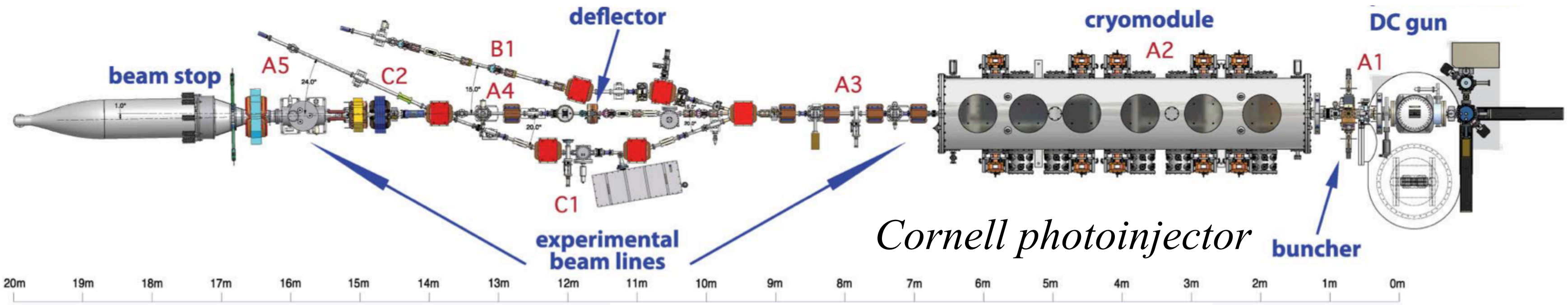
Core emittance invariance

Core emittance (max phase space density) should remain the same even if the forces vary along the bunch slices (e.g. space charge) provided no slice sheering occurs.

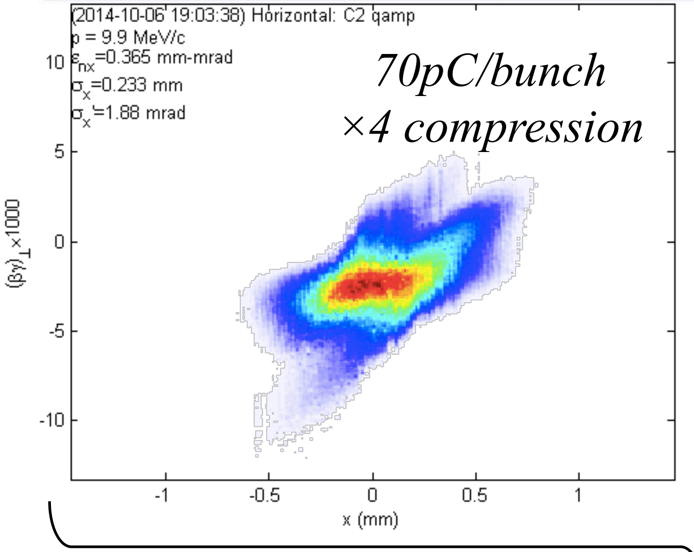




Core emittance determined by the photocathode

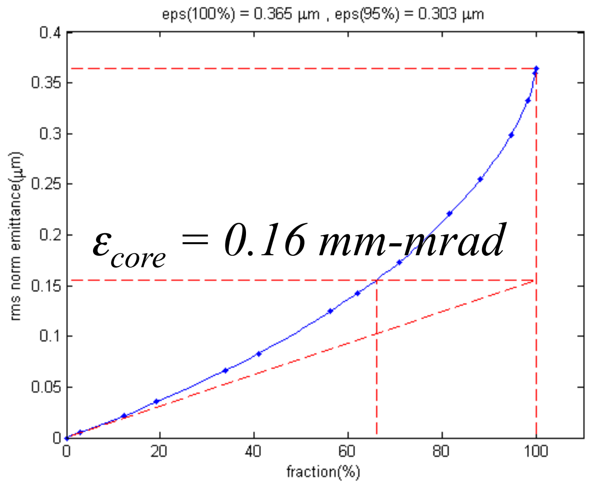


Cornell photoinjector

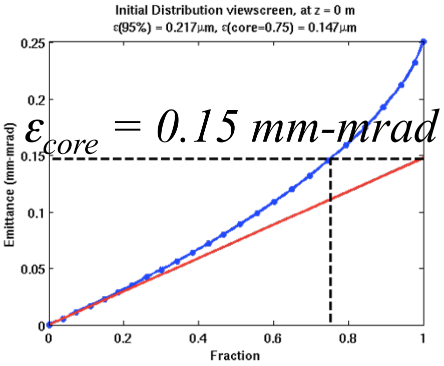


70pC/bunch

×4 compression



ε_{core} = *0.16 mm-mrad*



ε_{core} = *0.15 mm-mrad*

At the cathode (CsKSb, MTE = 190 meV)

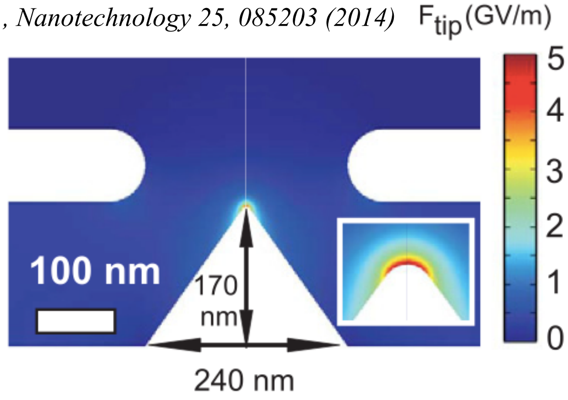
10 MeV (×4 compression)

Core emittance stays the same within experimental uncertainty of 6% or 0.01-0.02 mm-mrad all the way to 300 pC/bunch (highest we measured)

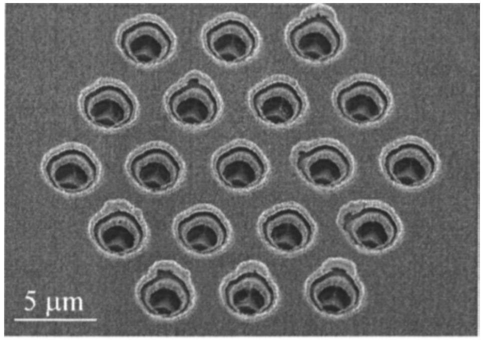
Needle cathodes

- Can tune to *arbitrarily* large fields;
- Arrays enhance current (but reduce brightness);
- Decide between (assisted) photo-/field-emission;

A. Mustonen et al., Nanotechnology 25, 085203 (2014)



E. Kirk et al., J.Vac.Sci.Tech.B 27, 1813 (2009)

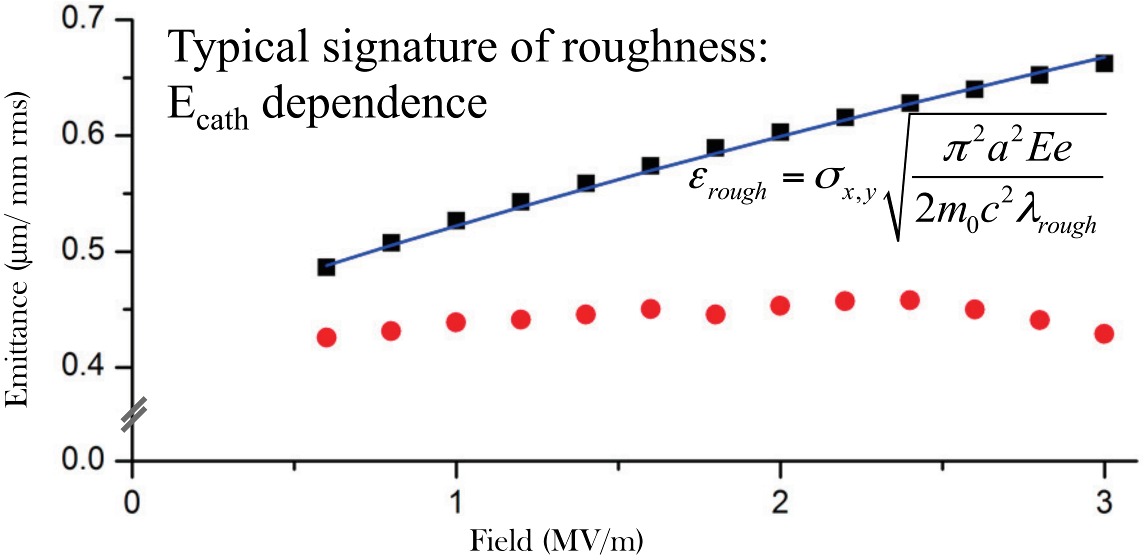


Interesting directions to watch:

- Combine needle cathodes with low MTE materials;
- Ultrashort (attosecond!) electron pulses via enhanced optical field.

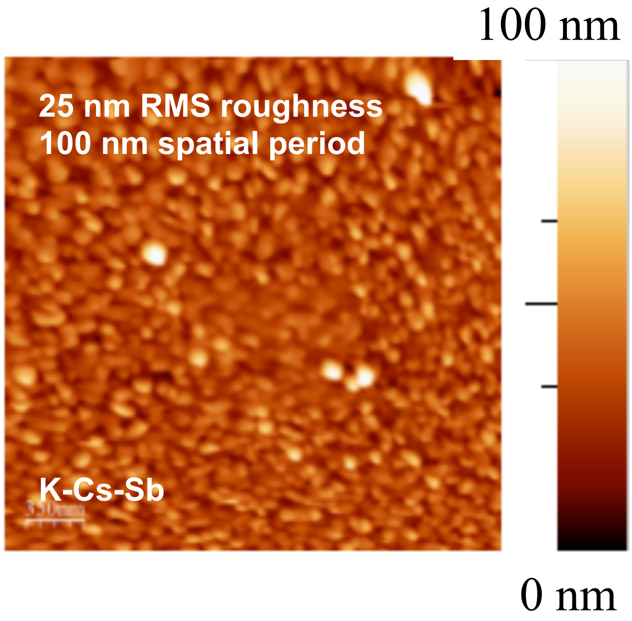
Roughness

- Roughness to become more critical as MTE is pushed lower;
- Some unanswered questions:
  - Roughness-free cathode growth methods;
  - Meso-scale roughness vs. sub-thermal MTE’s (≤10 meV);



Typical signature of roughness: E_{cath} dependence

$$\varepsilon_{rough} = \sigma_{x,y}\sqrt{\frac{\pi^2 a^2 E e}{2 m_0 c^2 \lambda_{rough}}}$$



25 nm RMS roughness
100 nm spatial period

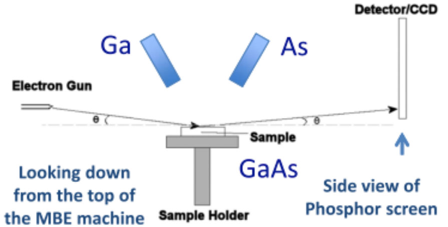
K-Cs-Sb

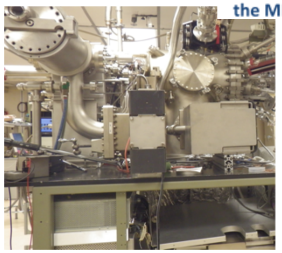
T. Vecchione et al., Proceedings of IPAC2012, 655 (2012)

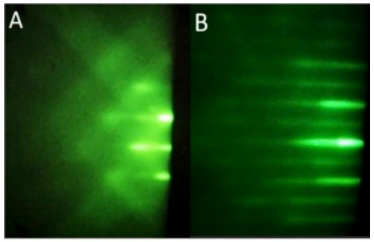
S. G. Schubert et al., *APL Mater.* **1**, 032119 (2013)

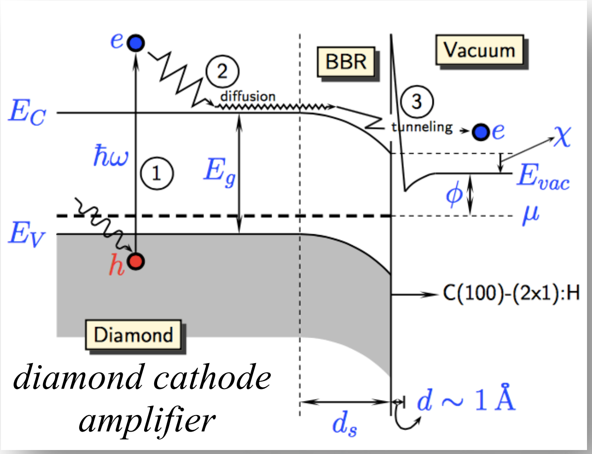
Theory (Monte-Carlo)

- Much progress explaining cathodes where electron transport in the bulk involves scattering;
- E.g. successful modeling of layered III-V's, now attempting more advanced structures.

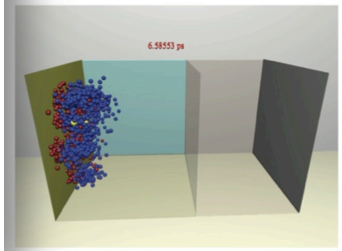


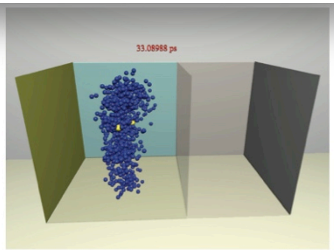


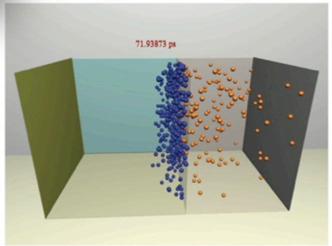


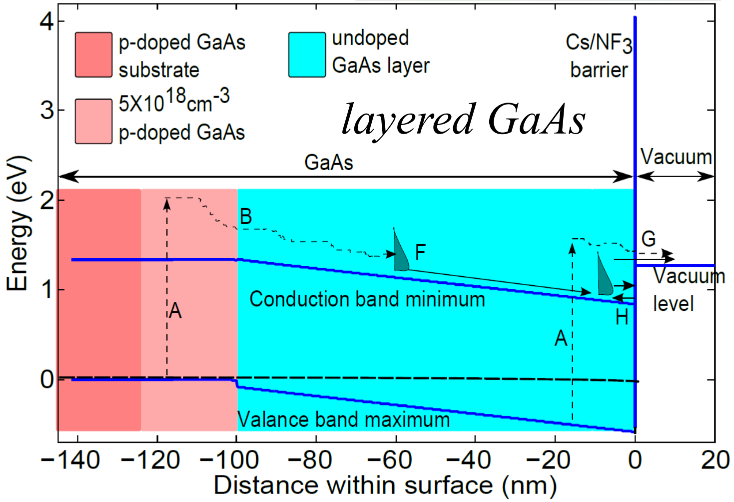


diamond cathode amplifier









layered GaAs

D. Dimitrov et al., IPAC2011, 2307 (2011), X. Chang et al, PRL 105 (2010) 164801

S. Karkare et al., Phys. Rev. Lett. ***112*** *(2014) 097601*

Outstanding questions in transport modeling

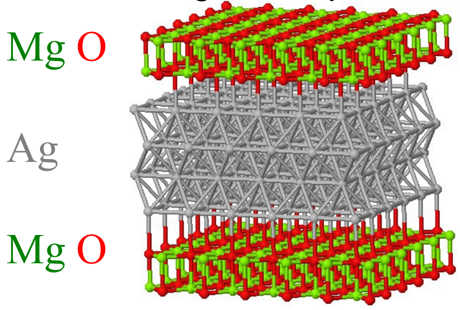
- Lacking self-consistency: modeling that gives best agreement remains a patchwork of several different approaches;
- Time to go fully quantum?
  - The role of the surface in the ultra-low MTE cathodes (e.g. ‘black box’ of photoemission when $m_e^*/m_0 \ll 1$);
  - Incorporating scattering along with QM (via non-equilibrium Green’s or Wigner Dist. Functions?);
  - One-step photoemission model for appropriate materials.

Many interesting physics questions!

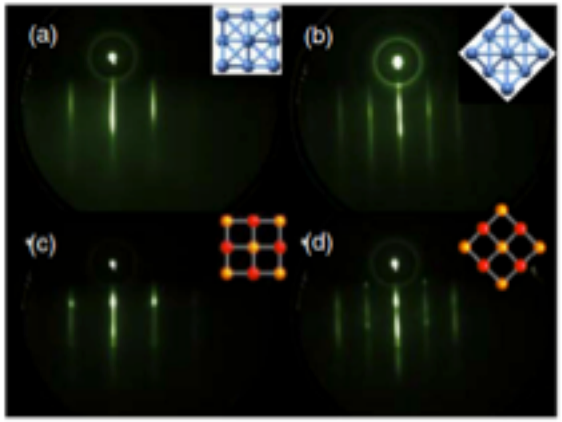
DFT (materials design)

- Goal: design a better photocathode in front of a computer;
- Complimentary goal: explain existing structures/physics.

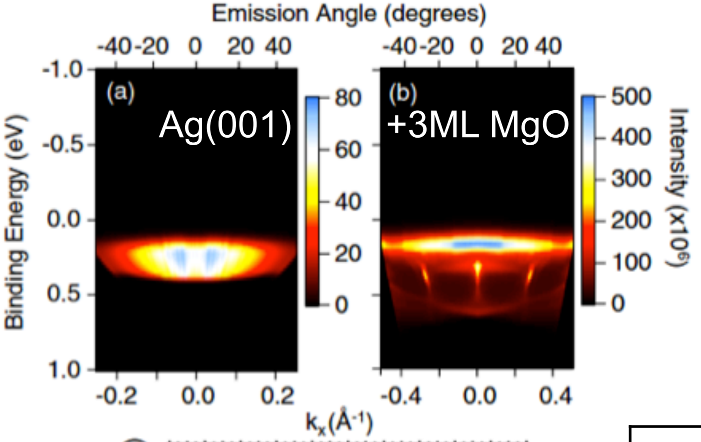
original inspiration

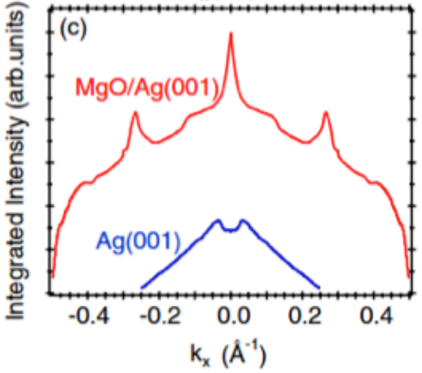


4 ML MBE grown MgO



RHEED indicate a non-perfect layer by layer growth





UV ARPES used to study emission properties and band structure

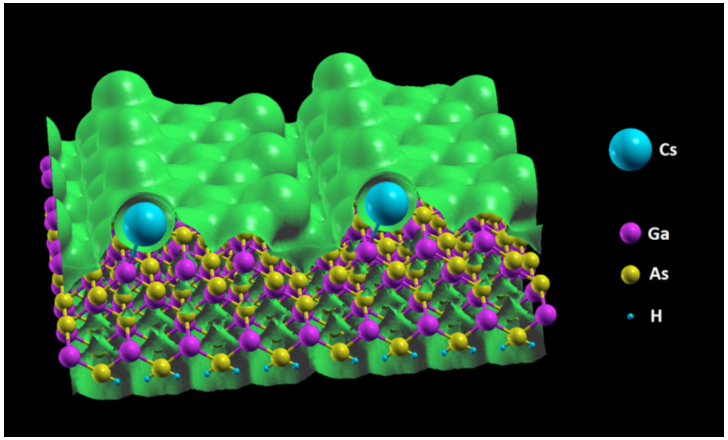
266 nm	Ag(001)	4ML MgO
QE	5×10^{-5}	3.5×10^{-4}
ε_{th} *(μm/mm)*	0.42	0.97

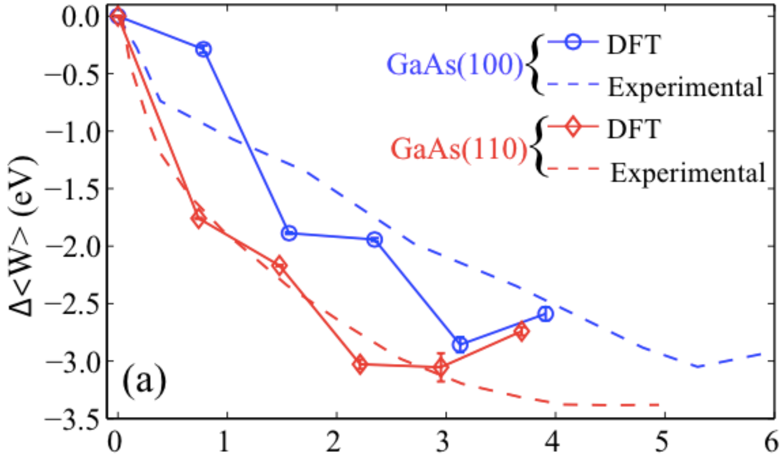
Target ε_{th} = 0.06 mm-mrad/ 1mm-rms (yet to be realized)

K. Németh et.al, Phys. Rev. Lett. 104, 046801 (2010)
*T. Droubay, Phys. Rev. Lett. **112**, 067601 (2014)*

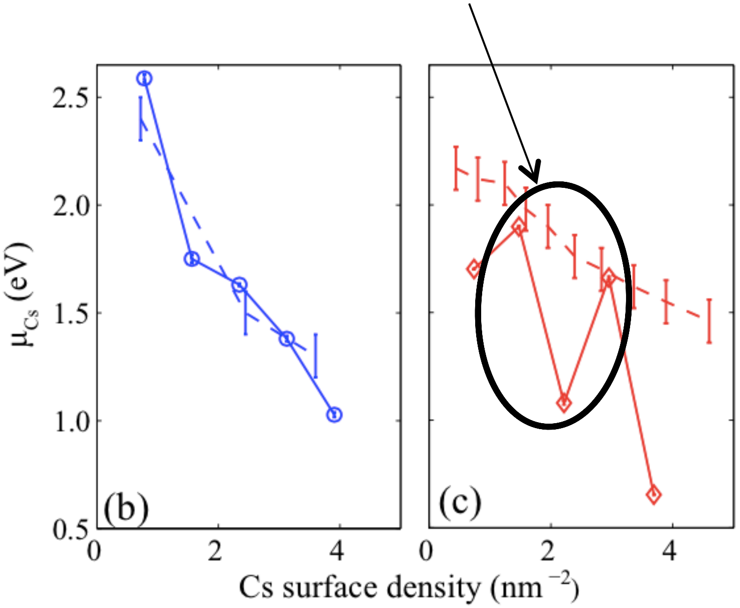
Example: using DFT to understand Cs structure on GaAs

- Can we understand the surprising variety of observed Cs structures on GaAs? *L. Boulet, this workshop*





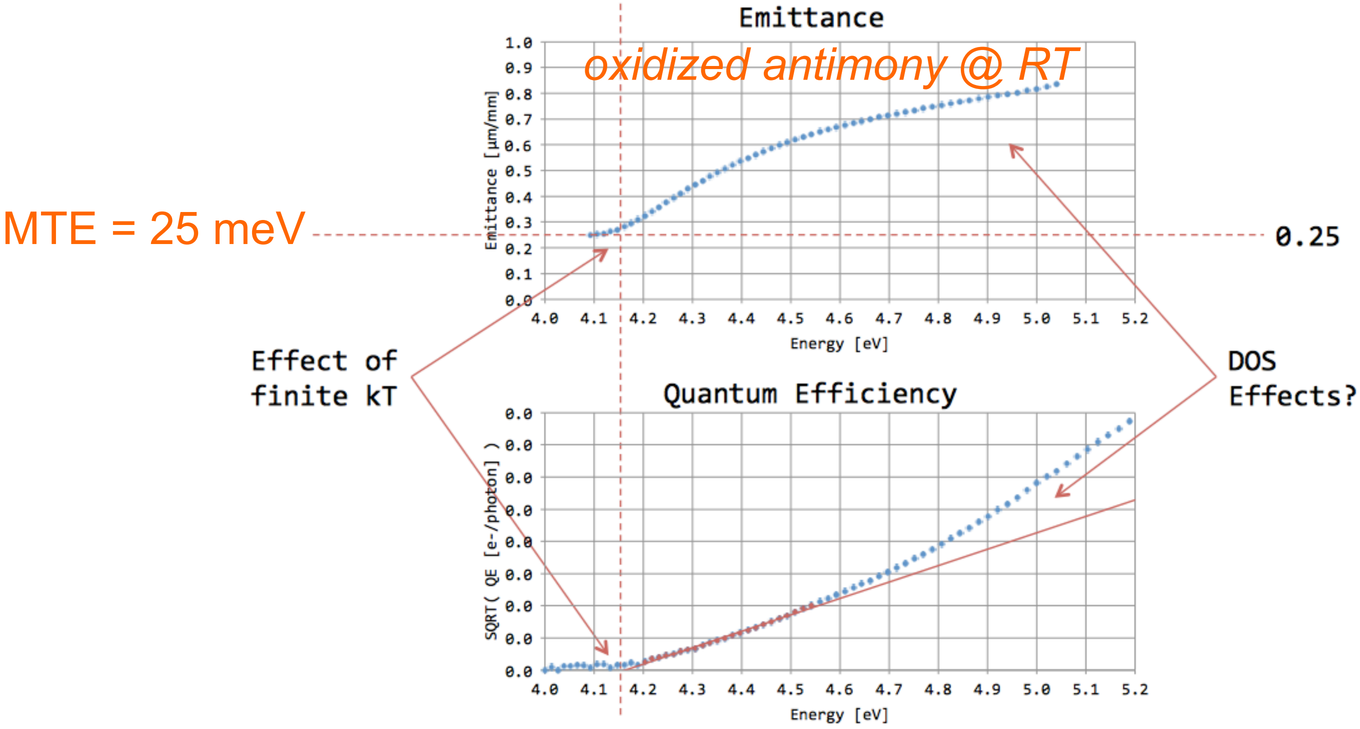
indication of a phase separation!



studied (100) and (110) surfaces

Ultralow MTE from cryogenically cooled photocathodes

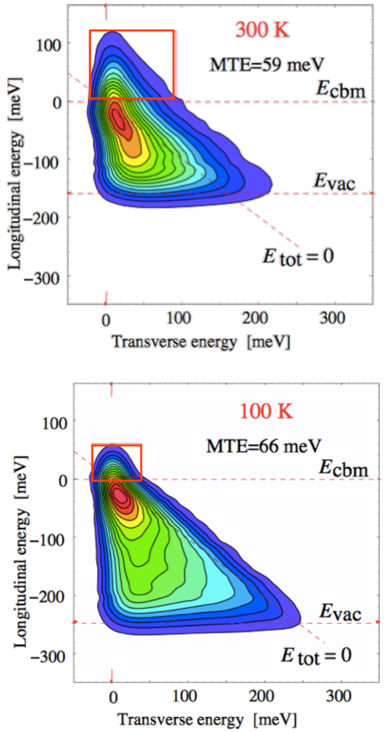
- Find materials with the room temperature MTE and cool them!
- Trading QE for lower MTE (still worth doing so for many apps);
- Try materials with higher density of states;
- Get MTE's similar to ionized ultracold (MOT) atoms.



ϕ = 4.15 eV

T. Vecchione, P3 Cornell workshop (2012)

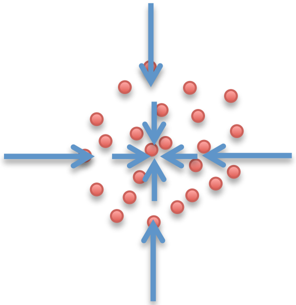
GaAs

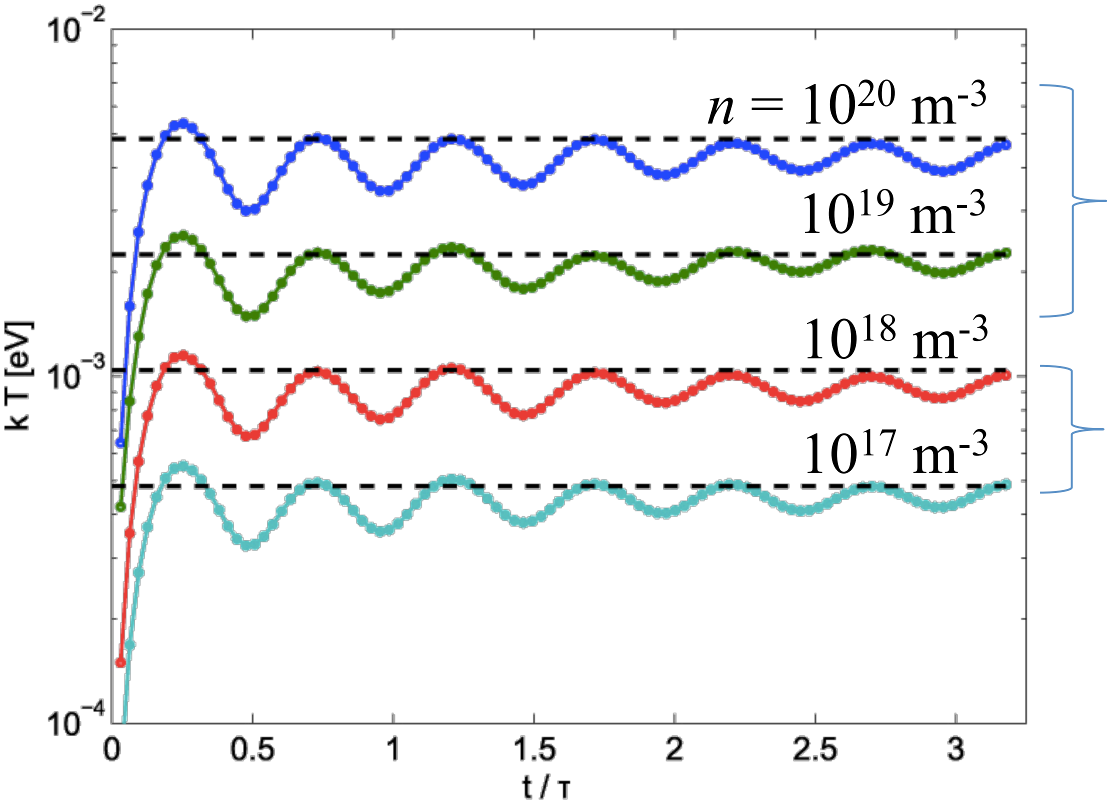


U. Weigel, PhD thesis (2003)

Disorder induced heating limit on MTE

- Potential energy of random electron positions thermalizes just after emission. Not ‘normal’ space charge!
- Density dependent! If $kT_{ini} = 0$, then $kT_{fin} \propto n^{1/3}$
- Reaches maximum in fraction of a plasma frequency.





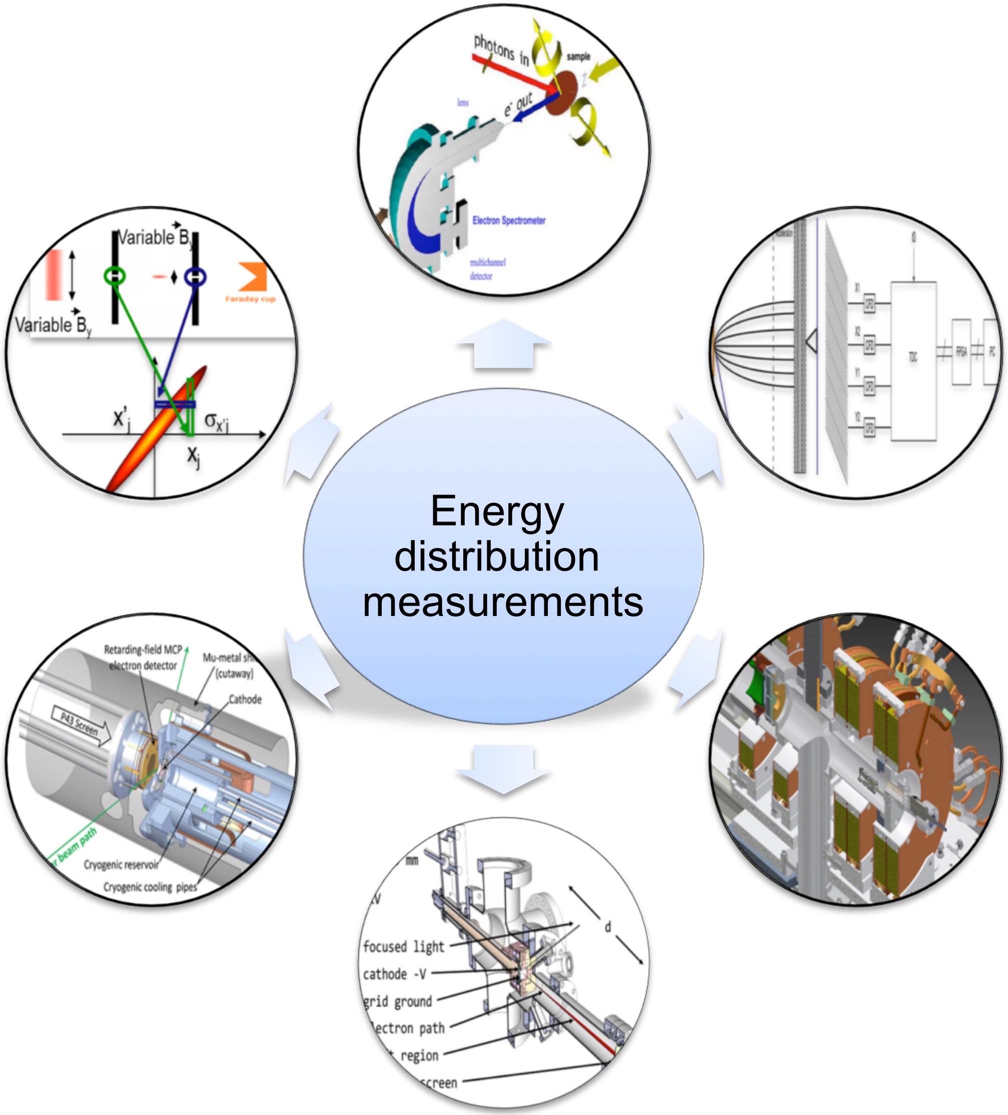
Sources with 10-100 MV/m

Sources with MV/m

Fundamental!

J.M. Maxson et al., New J. Physics ***15*** *(2013) 103024*

Measuring small MTE's is tricky



Energy distribution measurements

- Need to extend methods to higher resolution (~1 meV);
- Stray fields, work function differences, aberrations can dominate the measurement;
- Believe ultra-small MTE's once confirmed by ≥2 independent methods!

Outlook

- Photocathode research to remain vibrant in the upcoming years;
- Mature materials/surface diagnostics and sophisticated theoretical tools are used; link to the accelerator field/practice is essential;
- Provided the effort and funding momentum remains in the field, we should see much progress; breakthroughs possible.

2014…

Lots of great talks & discussions to look forward to!!

Many thanks to the LBNL hosts (especially Howard & Jason) for organizing the workshop