



Photocathode Physics for Photoinjectors (P3)

October 15-17, 2018
Santa Fe, New Mexico

Photocathode requirements and challenges for ultrafast electron scattering

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Oct 15, 2018, Santa Fe, NM

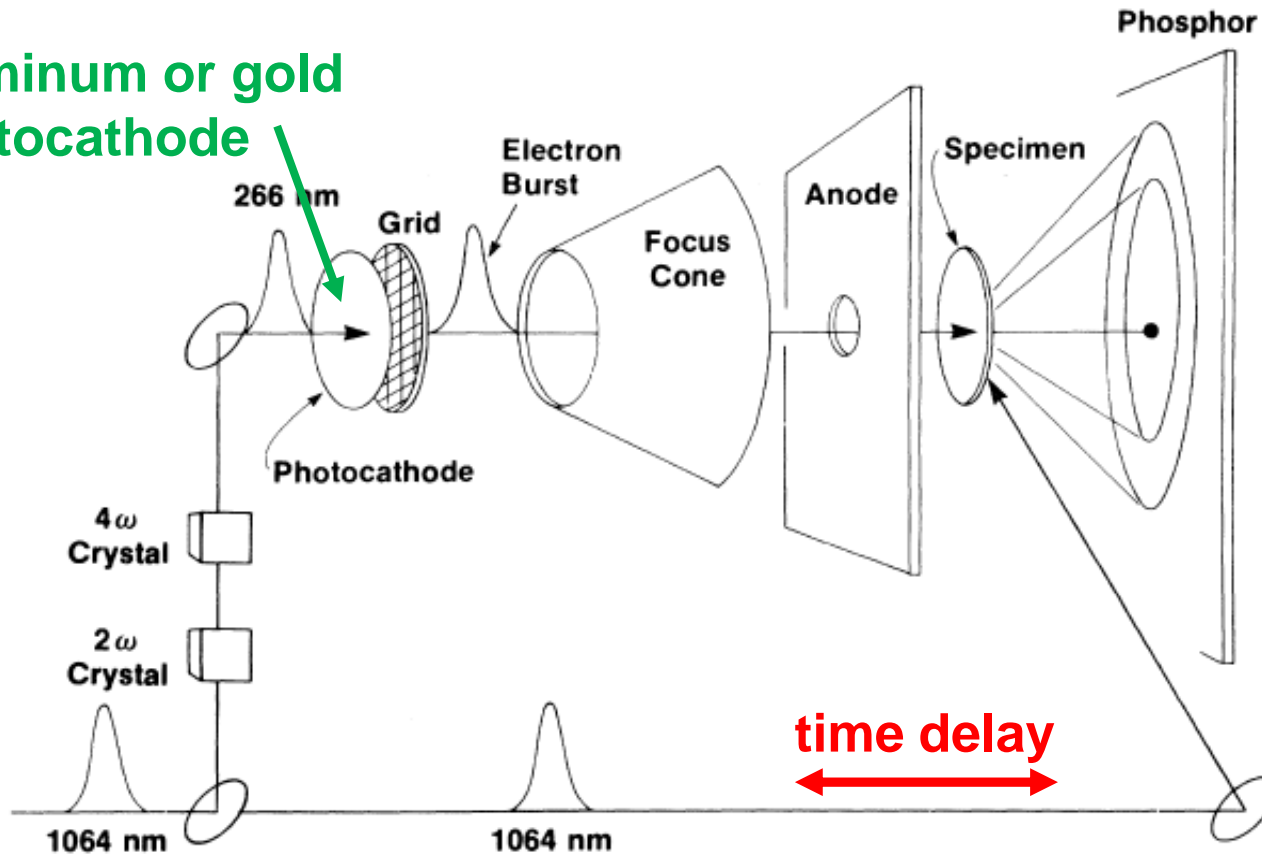
- Introduction and status of UED
- Photocathode requirements and challenges for UES
 - UED: single-shot, high rep-rate
 - UEM: extremely low emittance in SRF
 - fs EELS: pushing the longitudinal emittance
- Summary and outlook

UED – ultrafast electron diffraction

G. Mourou and S. Williamson, APL 41, 44 (1982)

S. Williamson, G. Mourou and J. C. M. Li, PRL 52, 2364 (1984)

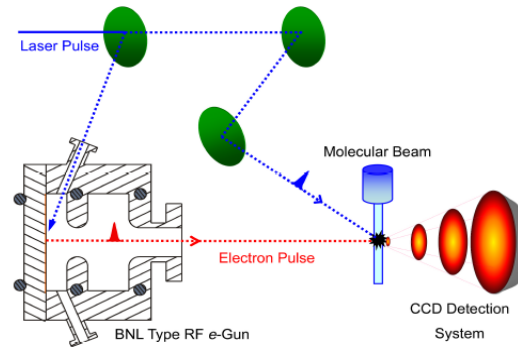
aluminum or gold
photocathode



Science outcome: See e.g. M. Chergui and A. H. Zewail, *ChemPhysChem* **10**, 28 (2009); R. J D. Miller, *Science*. **343**, 1108 (2014) and etc.

Benefits with MeV electrons

- Tremendous advances with keV UEDs in the past decades
- Significant Benefits with MeV e-**



Active R&D field around the world!

X. J. Wang, Z. Wu, H. Ihee, PAC'03, 420-422 (2003).

P. Musumeci and R. K. Li, ICFA BD Newsletter No. 59 (2012).

Space charge effects $\frac{1}{\beta^2 \gamma^3}$

- ✓ Shorter bunch
- ✓ Higher charge

Penetration/multiple scattering

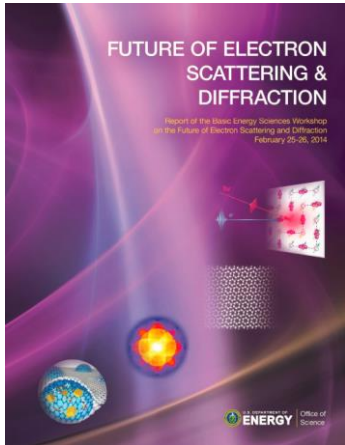
R. Henderson, Q Rev. Biophys. 28, 171 (1995)

Velocity mismatch

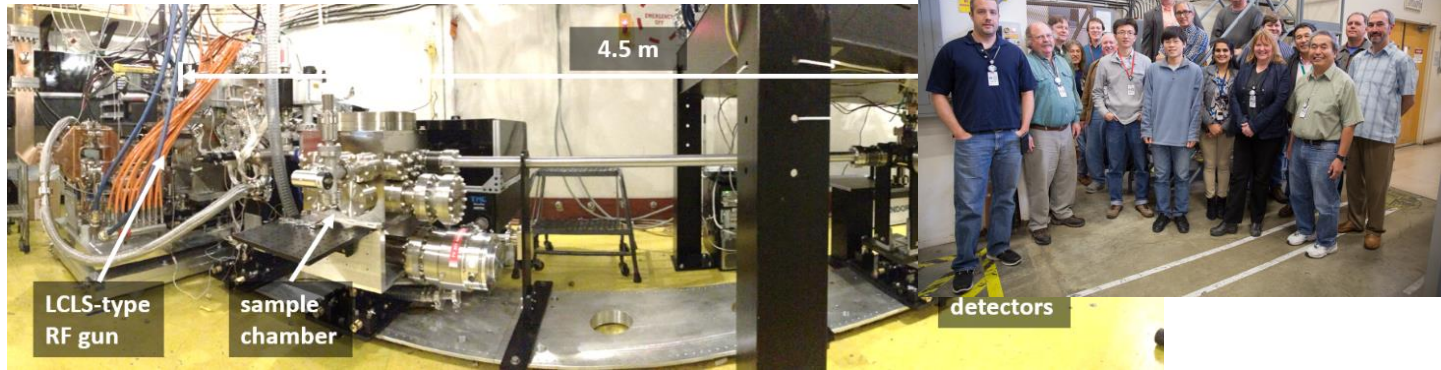
100 keV	$t_{VM}=0.8 \text{ ps}$	3 MeV	$t_{VM}=10 \text{ fs}$
$e \rightarrow$		$e \rightarrow$	
$h\nu \rightarrow$		$h\nu \rightarrow$	
300 um			

Shorter wavelength

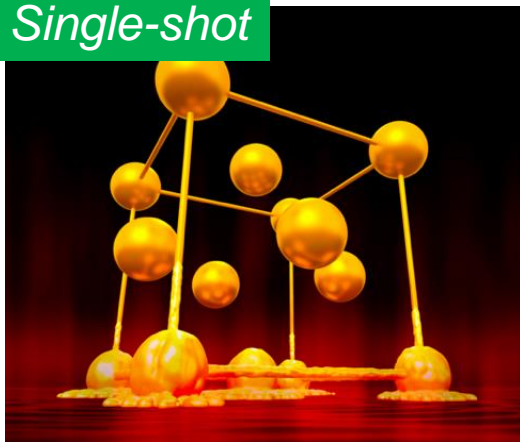
MeV UED at SLAC



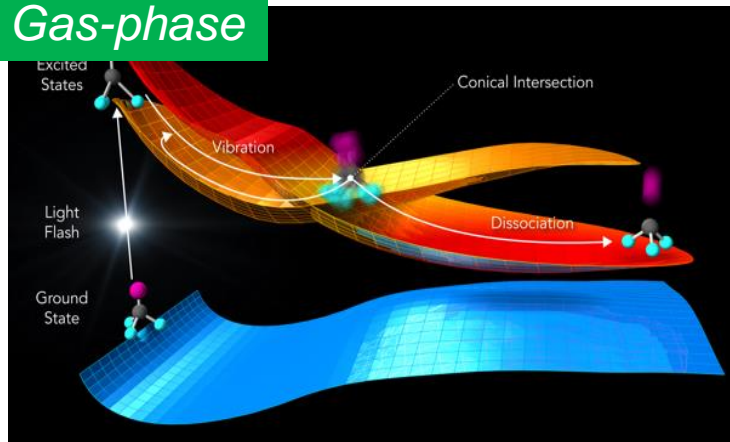
S. Weathersby et al., *RSI* **86**, 073702 (2015)



Single-shot



Gas-phase



30-40 experiments / yr

Solid state: nano-scale, 2D materials, diffuse scattering, strongly correlated system, functional material

Gas-phase: sequential double-dissociation, roaming reaction, ring opening

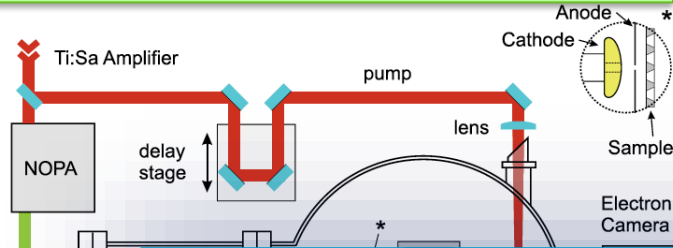
Liquid-phase: soon

Resolving ultrafast phase transitions -
Science **360**, 1451 (2018)

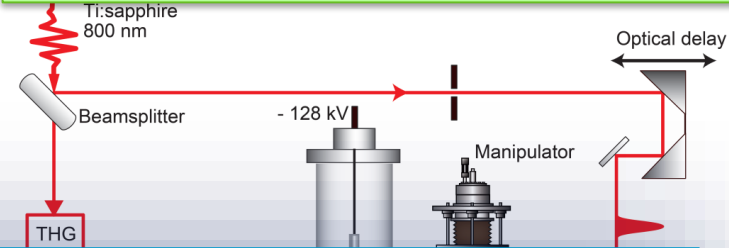
Bond-breaking & nuclear wavepacket passing through conical intersections
- *Science* **361**, 64 (2018)

State-of-the-art keV UEDs

L. Waldecker et al., JAP 117, 044903 (2015)



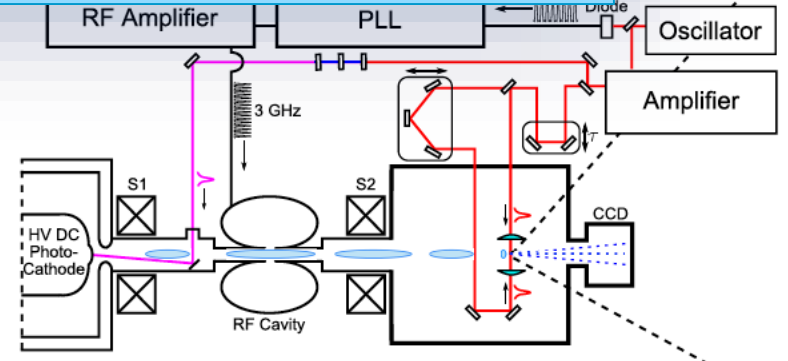
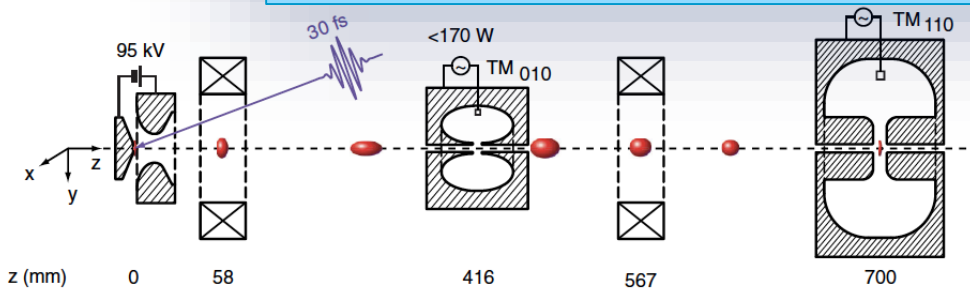
S. Ideta et al., Sci. Adv. 4, eaar3867 (2018)



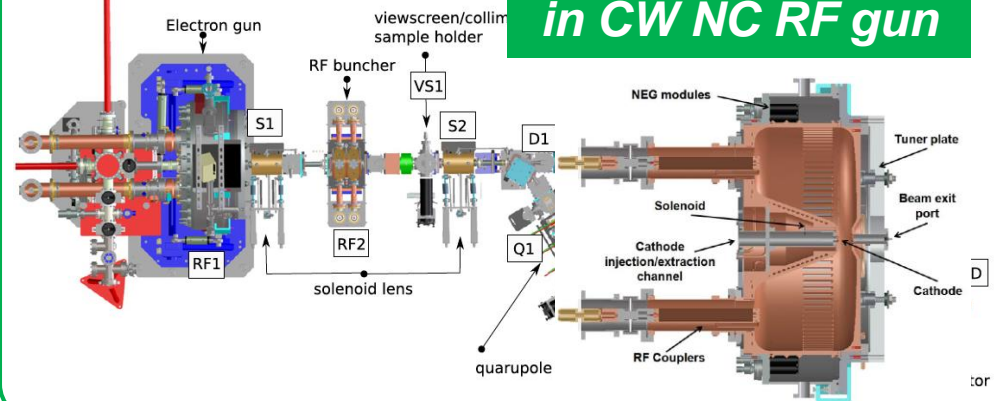
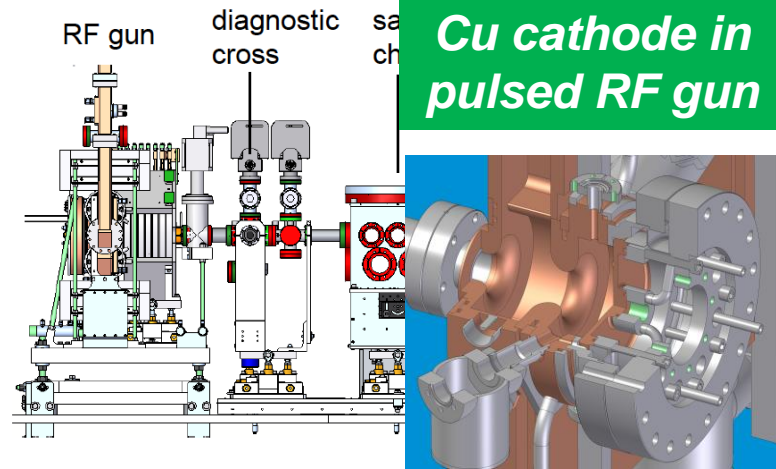
- Simple **thin film** or **bulk** metallic cathodes (Au, Cu)
- Up to 10^6 e- per pulse w/ fs laser and ~ 100 μm emission area
- Up to kHz rep-rate (allow samples to relax)

T. van Oudh...

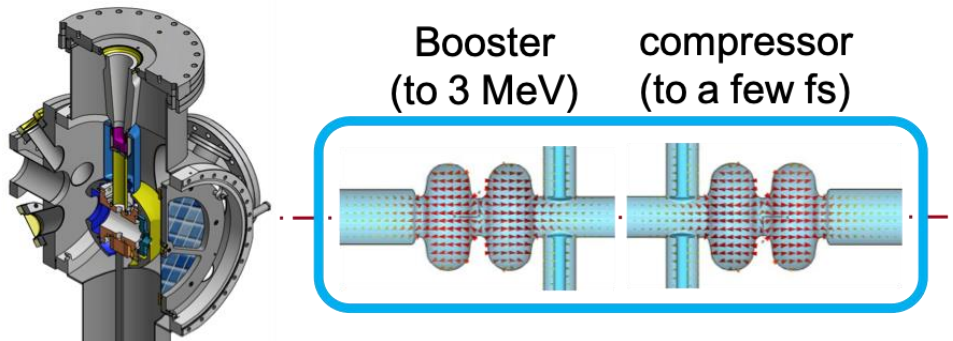
01 (2012)



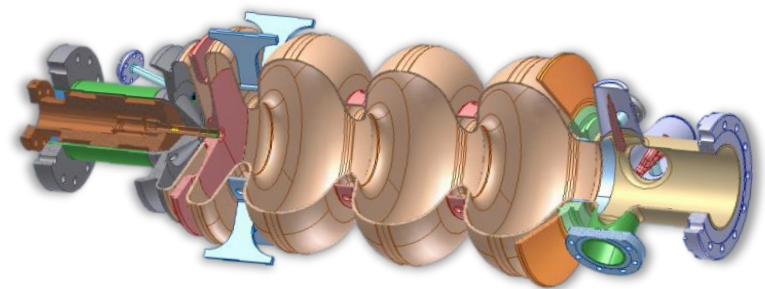
Guns and photocathodes for MeV UEDs



Cooled semiconductor cathodes in DC gun + booster



Metallic or semiconductor cathodes in SRF gun



Requirements for UED

Signal level roughly scales *linearly* with beam brightness

$$B_{6D} \propto \frac{N}{(\sigma_x \cdot \gamma \sigma_{x'})^2 \cdot \sigma_t \cdot \sigma_\gamma} \cdot f_{\text{rep}}$$

← **signal level** (points to N)
← **rep. rate, depends on samples and processes** (points to f_{rep})
← **sample size** (points to σ_x)
10s of μm or less
← **q-resolution** (points to $\gamma \sigma_{x'}$)
1 mrad or smaller
↑ **bunch length** (points to σ_t)
100 to few fs
← **energy spread** (points to σ_γ)
10⁻³ or smaller

Processes	f_{rep}	Photocathodes
Irreversible, high-energy density	Single-shot	to reach highest B_{6D}/f_{rep}
Reversible, relatively strong pump	Up to kHz	pulsed gun
Reversible, weak pump, gas/liquid flow, nano-diffraction	Up to MHz	CW gun

Reaching atomic-level spatial resolution is the priority

$$B_{6D} \propto \frac{N}{(\sigma_x \cdot \gamma \sigma_{x'})^2 \cdot \sigma_t \cdot \sigma_\gamma} \cdot f_{\text{rep}}$$

illumination area
1 μm or less

related to contrast & Cs, 1 mrad or smaller

signal level

Somehow relaxed

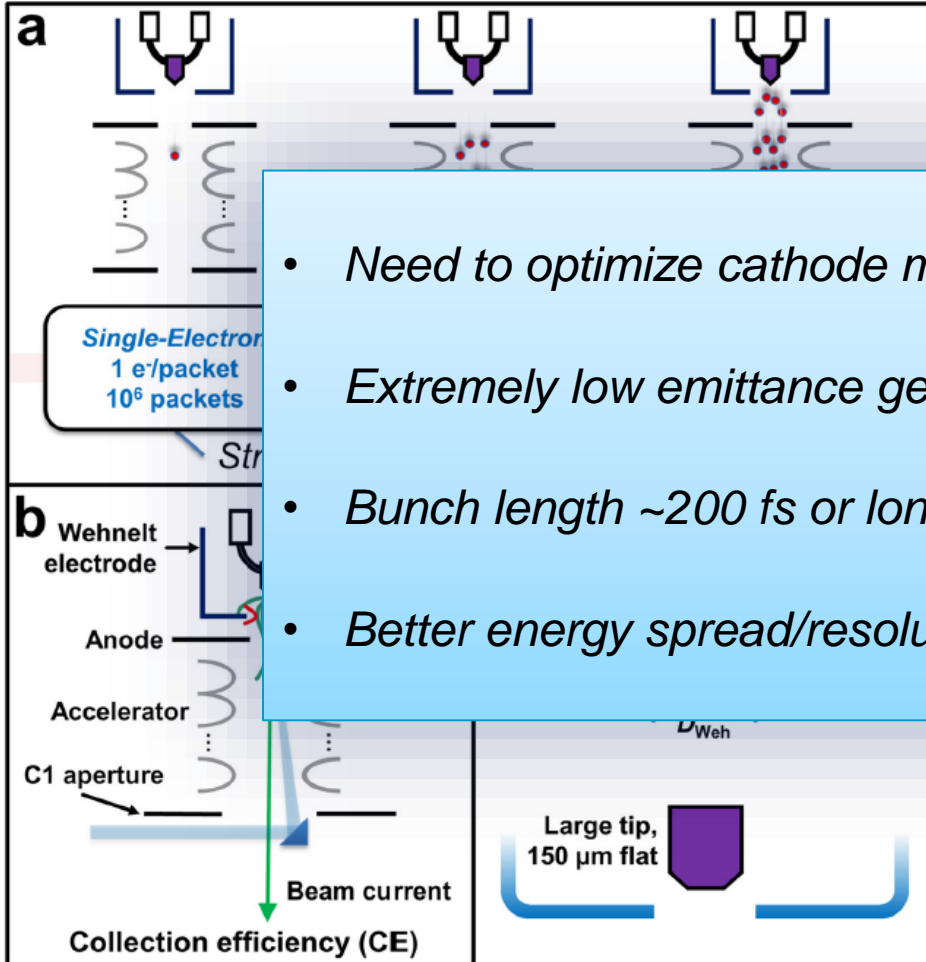
related to Cc 10⁻⁵ level

rep. rate stroboscopic or single-shot

Operation mode	Beams
stroboscopic	200-300 kV UEM: ~1 e- per pulse, $\epsilon_n < 1$ nm-rad, 200 fs pulse MeV UEM: similar charge and ϵ_n , can reach 10 fs
single-shot	10 ⁶ -10 ⁸ e- per pulse, similar ϵ_n

Cathodes for keV UEM

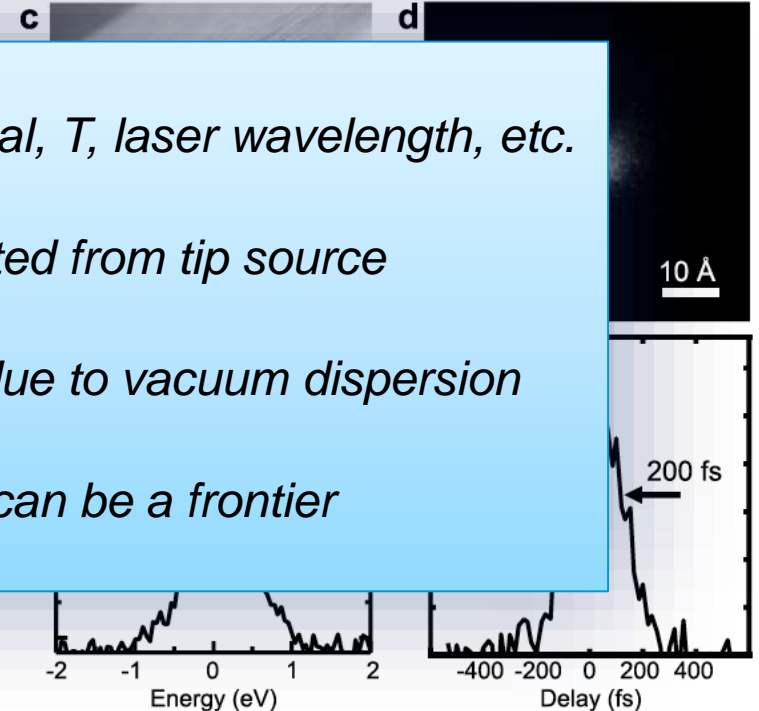
LaB₆ source in commercial TEMs



Tip field emission source

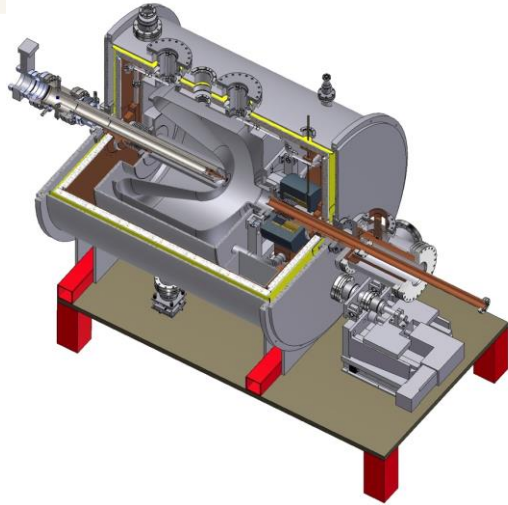
ZrO/W(100) tip emitter
0.5-1 V/m field, 1800 K

- Need to optimize cathode material, T , laser wavelength, etc.
- Extremely low emittance generated from tip source
- Bunch length ~ 200 fs or longer due to vacuum dispersion
- Better energy spread/resolution can be a frontier



Stable operation >48 h

A. Feist et al. Ultramicroscopy 176, 63 (2017)



VHF SRF gun as a promising candidate for UEM w/ atomic spatial resolution

- Requires $\epsilon_n < 1$ nm-rad
 - e.g. < 5 μm spot size and 0.2 $\mu\text{m}/\text{mm}$ intrinsic emittance
- > 1 pC charge in single-shot mode
 - 1 pC from few μm spot: need high QE to avoid heating effect

Stability: toward 10^{-5} in amplitude

High gradient: up to 40 MV/m

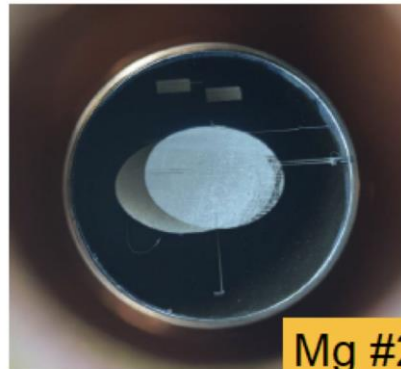
High beam energy: 4 MeV

Flexibility: ~ 100 ps bunch length

High rep-rate: up to 200 MHz

QWR geometry, 4.5 K operation

Challenges: gradient, cathode



Mg #214

Reliable operation of Mg cathode in SRF Gun II at ELBE HZDR

R. Xiang et al,
THPMF039,
IPAC2018.

Requirements for fs EELS

Pushing the *longitudinal emittance* of photocathodes

$$B_{6D} \propto \frac{N}{(\sigma_x \cdot \gamma \sigma_{x'})^2} \cdot f_{\text{rep}} \cdot \sigma_t \cdot \sigma_\gamma$$

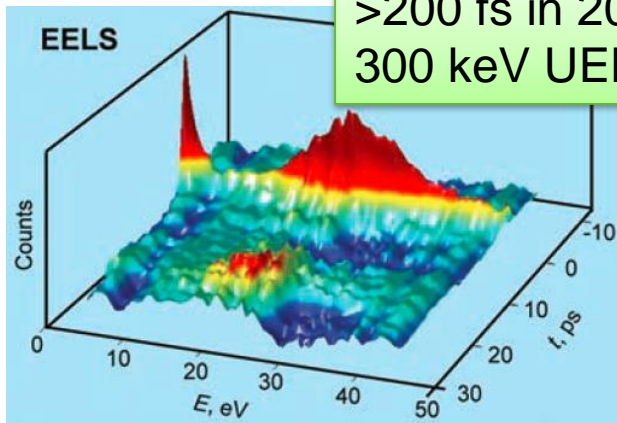
1 e- per pulse to control space charge

rep. rate

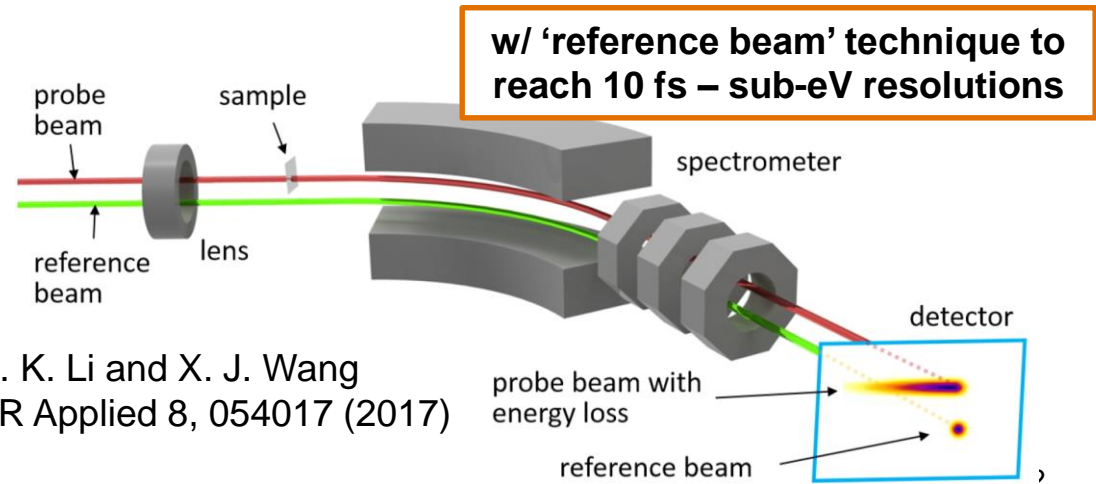
Probe size
nm level

related to spectrometer resolution, < 1 mrad

Towards 10 fs
Stay effectively at sub-eV level



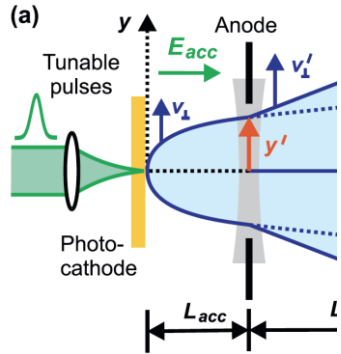
>200 fs in 200-300 keV UEMs



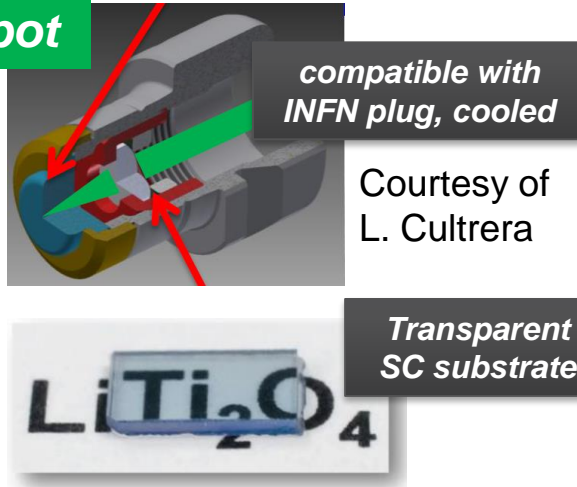
R. K. Li and X. J. Wang
PR Applied 8, 054017 (2017)

Sub-nm-rad emittance from small emission area

tight focus to um spot

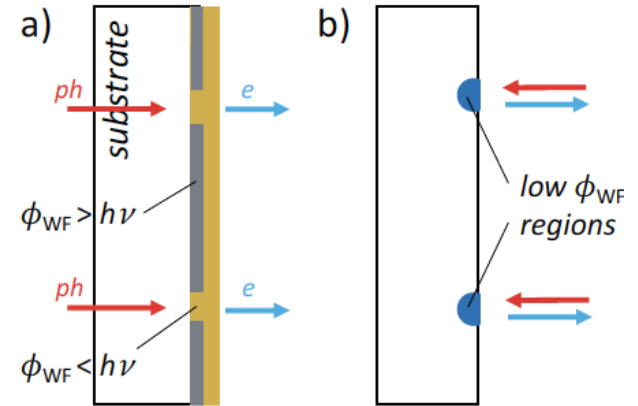


P. Baum et al., PNAS 107, 19714 (2010).



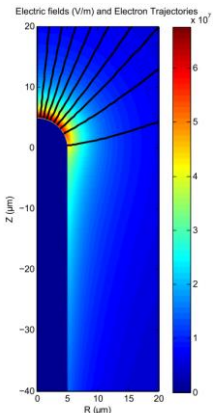
R. Inagaki et al., IPAC'14, MOPRI035

Sub-um flat cathode

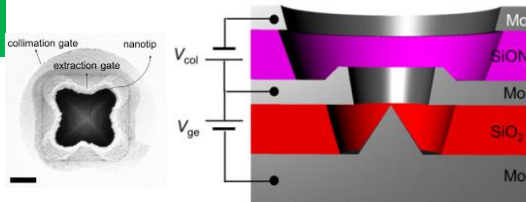


R. K. Li and X. J. Wang, PR Applied 8, 054017 (2017)

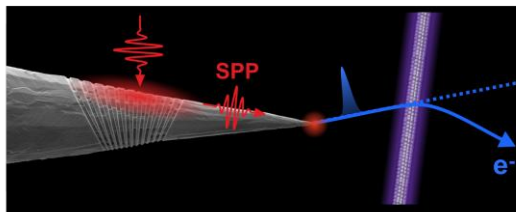
Tip sources



Courtesy of J. Maxson



C. Lee et al., APL 113, 013505 (2018)



M. Muller et al., ACS Photonics 3, 611 (2016)

- Minimize spot size × divergence
- Control MTF with T, λ, E, etc.
- Laser heating
- Surface contamination

Cathodes in extremely high fields

$$\mathcal{B}_{4D} \propto \frac{E_z}{\text{MTE}}$$

for pancake beam

Bazarov et al., PRL 102, 104801 (2009)

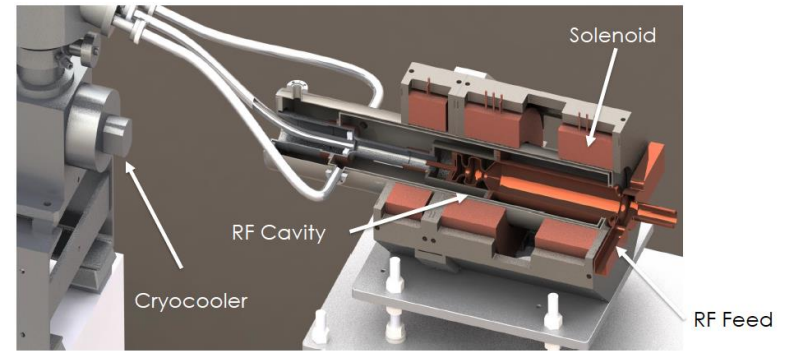
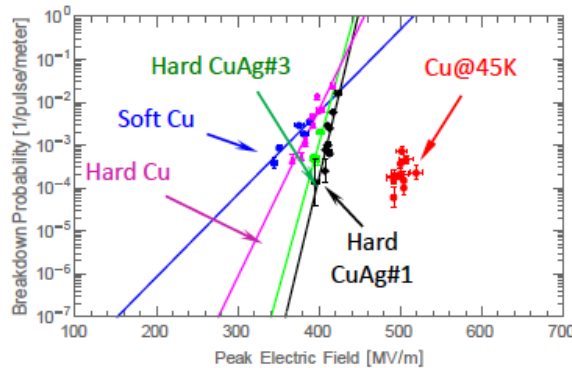
$$\propto \frac{E_z^{3/2}}{\text{MTE}} \frac{\sigma_t}{\sigma_x^{1/2}}$$

for cigar beam

Filippetto et al., PRAB 17, 024201 (2014)

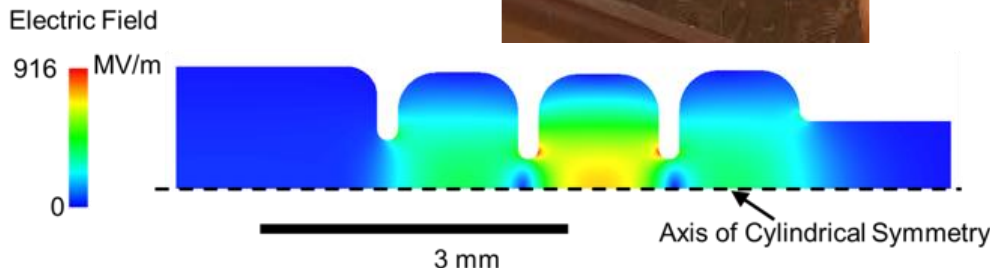
>250 MV/m field
with cryo-cooled Cu gun

Courtesy of A. Cahill, J. B. Rosenzweig, et al.



GV/m field in
THz gun

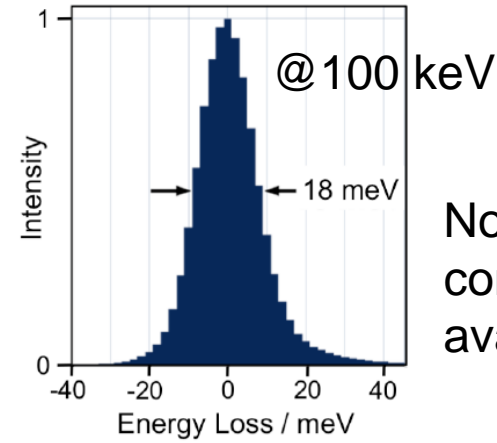
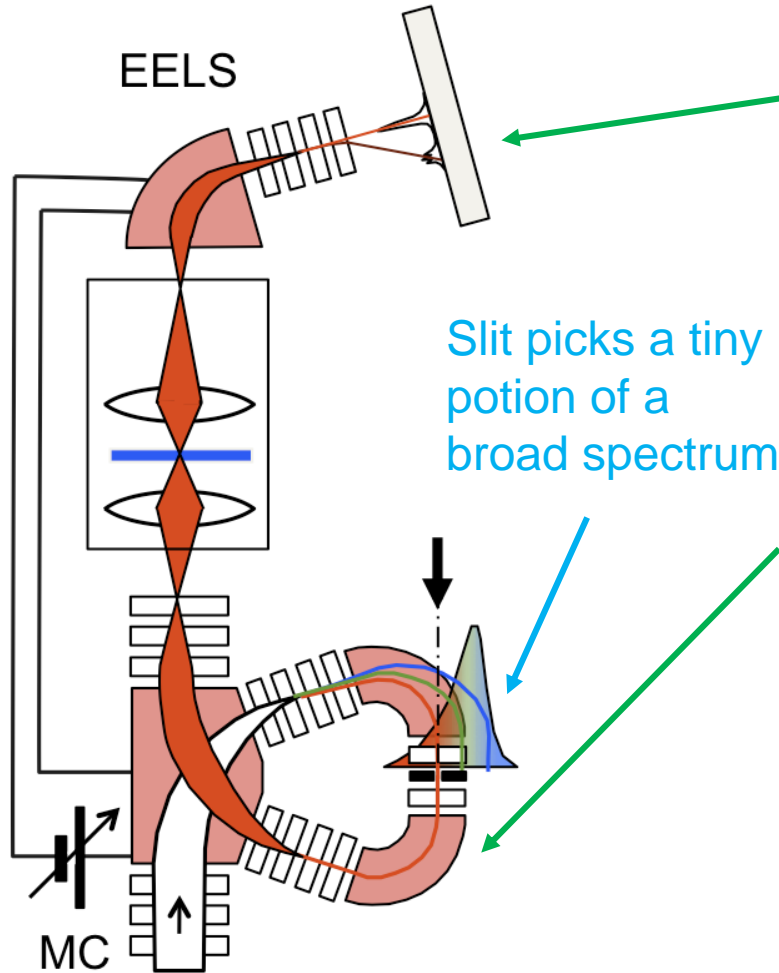
Courtesy of E. A. Nanni



- Need compatible cathodes
- Can they survive?
- Modified MTF due to fields, surface morphology, etc.

meV energy resolution

MC+EELS change



Now sub-10-meV commercially available

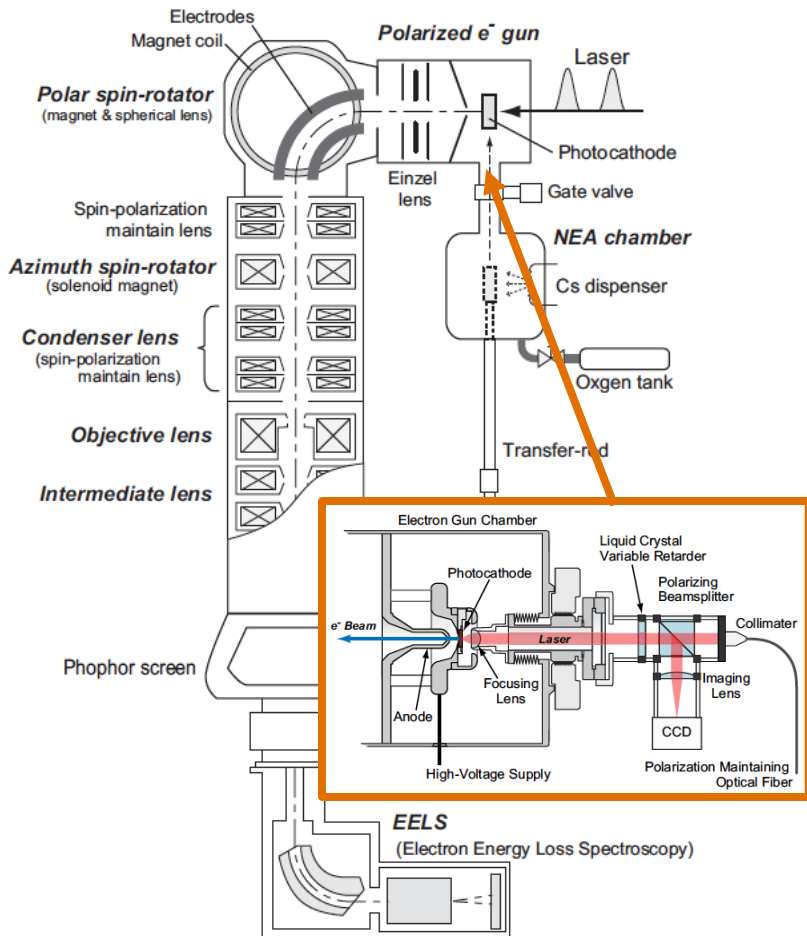
ΔE (eV) @100 kV	LaB ₆	Schottky FEG	Cold FEG
ΔE	1-2	0.3-0.7	0.2-0.7

Reimer and Kohl, TEM, 2007

Cryogenic photocathodes will significantly advance ultra-resolution EELS!

More cathode options for UED

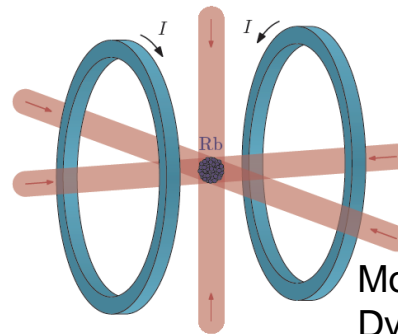
Spin-polarized TEM



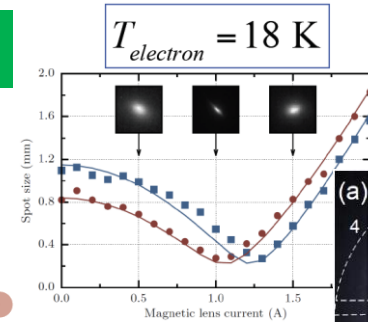
Kuwahara et al. APL 101, 033102 (2012)

MOT source

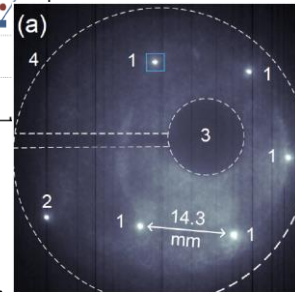
Laser cooling & trapping



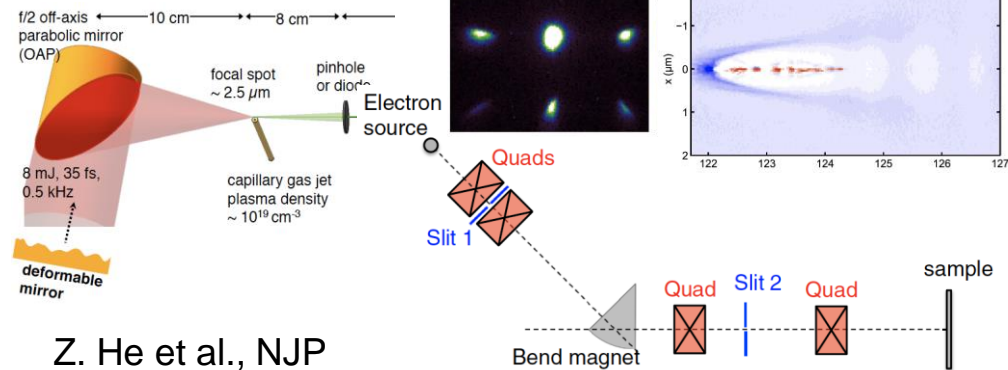
Mourik et al., Struc. Dyn. 1, 034302 (2014).



Graphite DP (13.2 keV)



LPWA source



Z. He et al., NJP 15, 05316 (2013)

J. Faure et al., PRAB 19, 021302 (2016)

Acknowledgement

- Helpful discussions with Pietro Musumeci, Daniele Filippetto, Jared Maxson, Theo Vecchione and many others
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- Work supported in part by the U.S. Department of Energy Contract No. DE-AC02-76SF00515 and the SLAC UED/UEM Initiative Program Development Fund.



- UES are demonstrated tools with transformative impacts, complementary and synergistic with XFEL
- Photocathode improvements will benefit UES

A few near-term items on the wish-list: In extremely high launching field, in high gradient SRF gun, extremely small initial emittance, meV energy spread

- New capabilities will be enabled by new photocathodes

Thank you for your attention!

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