MeV Ultrafast Electron Microscopy

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Department of Physics and Astronomy

Acknowledgements

- R. K. Li (now at SLAC) (*Phys. Rev. Applied* **2**, 024003, 2014)
- Graduate students: **D. Cesar** and E. Pyrez

- Collaborators: D. Alesini, G. Andonian, F. Carbone, D. Filippetto, J. Luiten, J. B. Rosenzweig, J. Spence
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radiabeam TECHNOLOGIES

NSF

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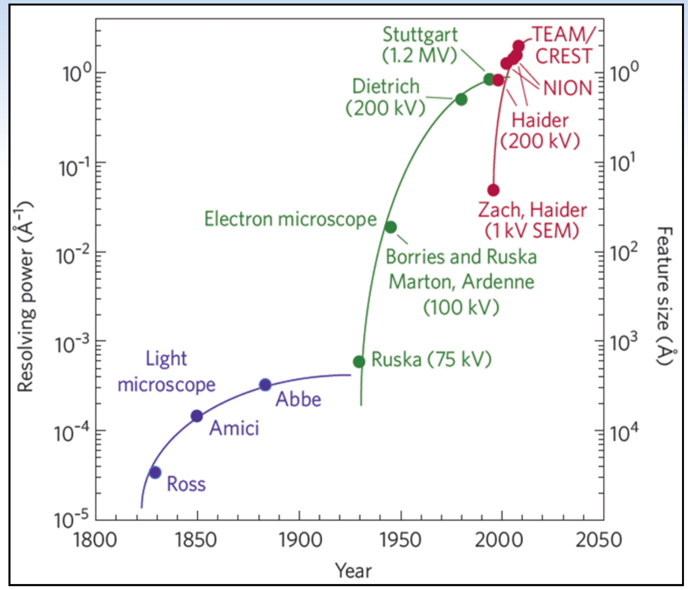
Outline

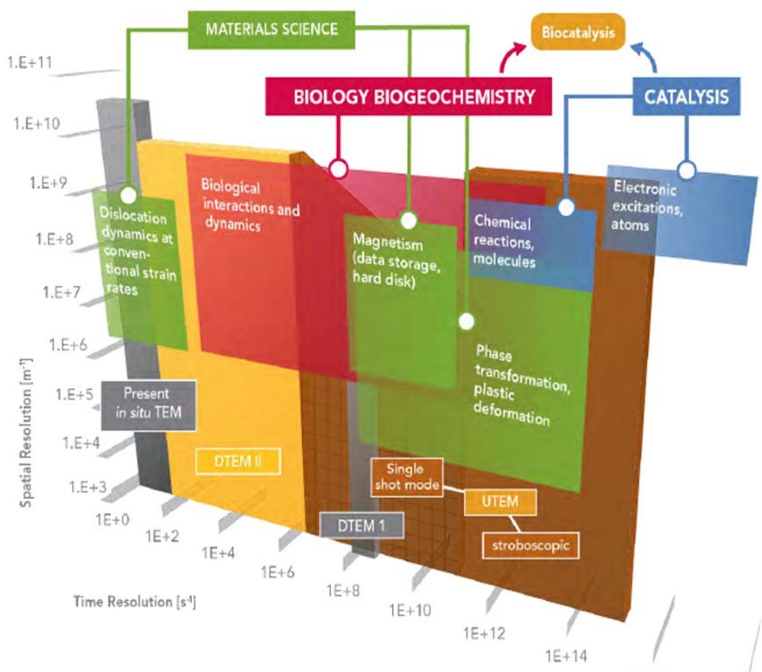
- Background
  - DTEM, UEM and UED
- MeV time resolved TEM Design study
  - High brightness. Low thermal emittance. Cold Cathodes.
  - RF gun
    - Clamped gun commissioning
    - 1.4 cell design
  - Energy spread linearizer. X-band structure design
  - Condenser stage
    - Two-solenoids scheme
    - Pegasus beamline measurements. 30 nm 100 fC 3 um rms spot size
  - Lenses
    - PMQ aberration comparison
    - MEMS-based quadrupoles. Prototype results
  - Space charge effects. Stochastic and mean field contributions to spatial resolution
- Conclusions

Add 4th dimension: Time resolved TEMs

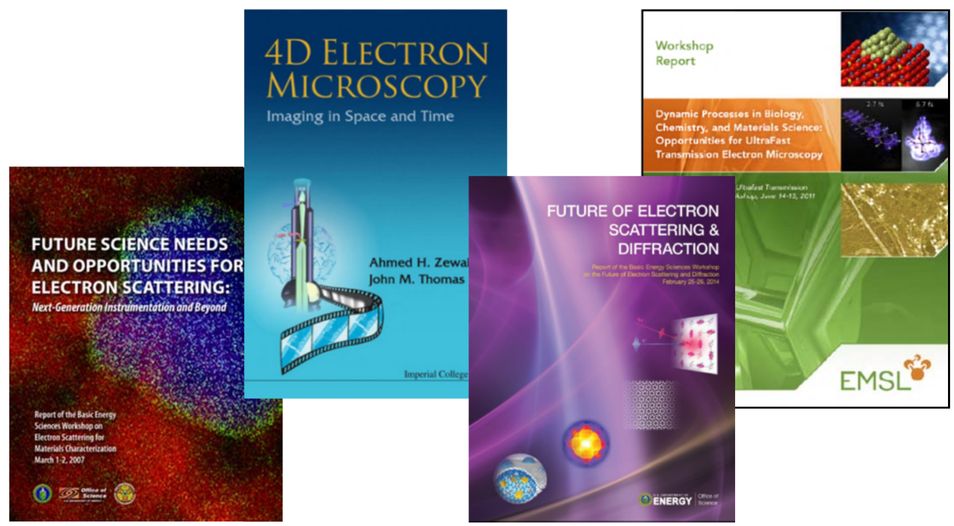
- *Conventional TEMs* primary tool for material science, chemistry, physics, biology, and industry
- *need millisecond to second exposure time*
- sub-Ångström spatial resolution with aberration correction
- *Many reasons to see structural* ***changes*** *in time, rather than static images*



D. A. Muller, Nat. Mater. 8, 263 (2009)
Adapted from H. H. Rose (2009)

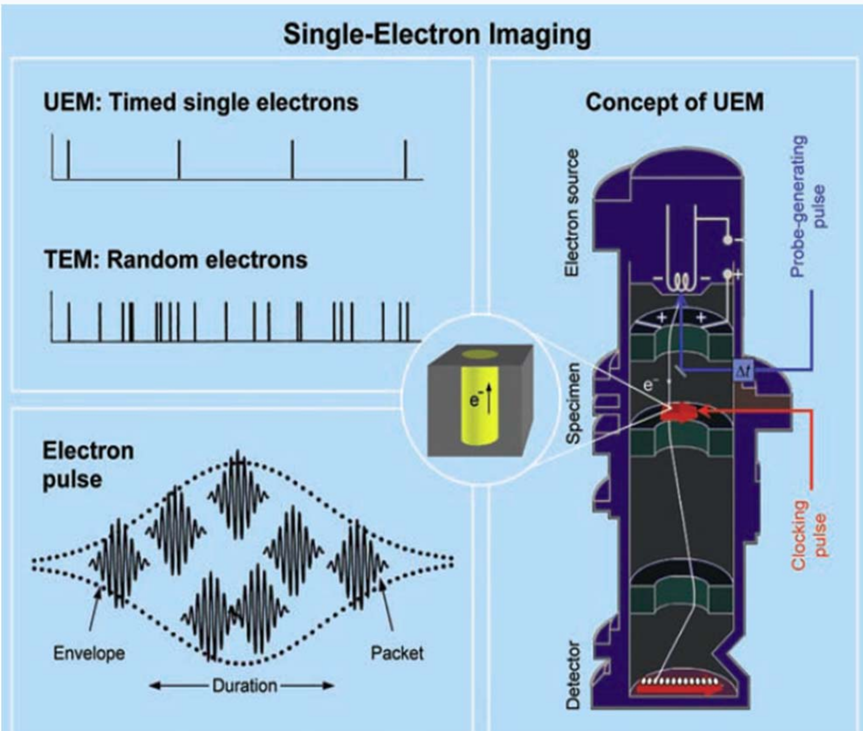


EMSL Ultrafast TEM Workshop Report, June 2011



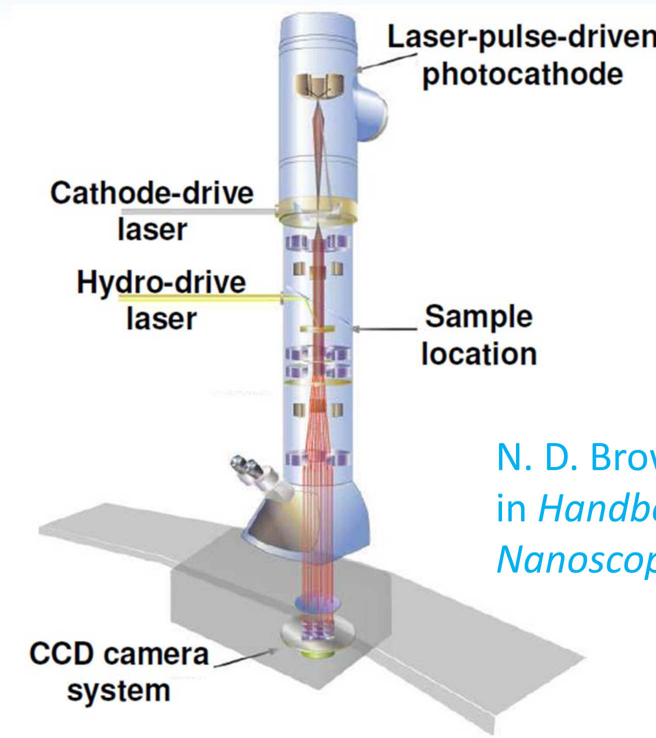
Ultrafast TEM: stroboscopic and single-shot

Stroboscopic



A. H. Zewail, Science 328, 187 (2010)

Single-shot



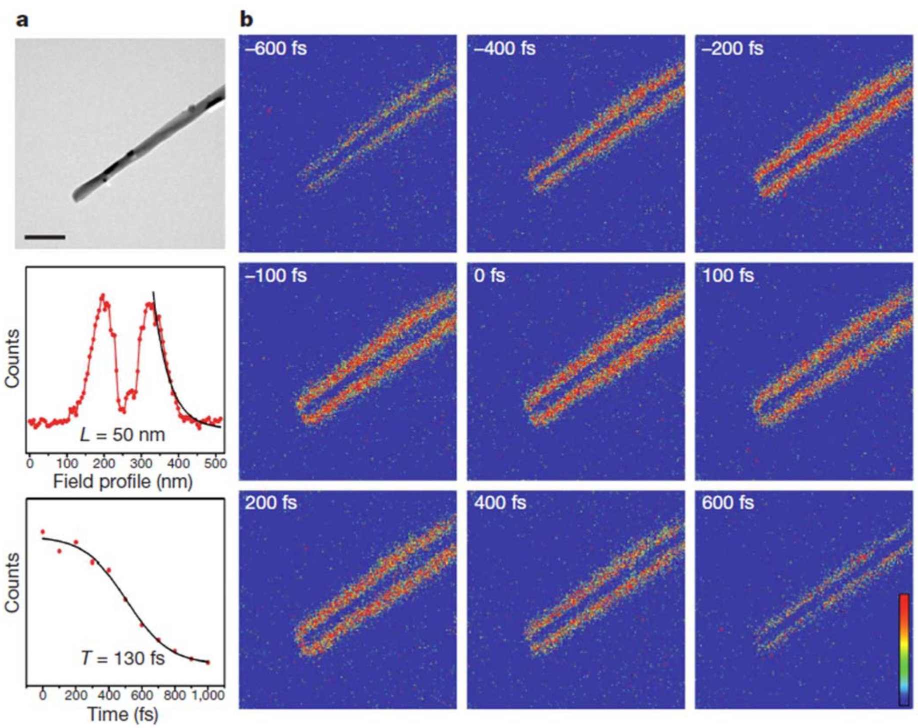
O. Bostanjoglo, in *Advan in Imag Elect Phys*,121 (2002)

N. D. Browning et al., in *Handbook of Nanoscopy*, 2012

- *use photocathode in conventional 200 kV TEMs, pump-probe scheme*
- *Stroboscopic: 1 e^-/pulse, sub-ps, atomic scale resolutions*
- *Single-shot: 10^8 e^-/pulse, 10 nm – 15 ns resolution*

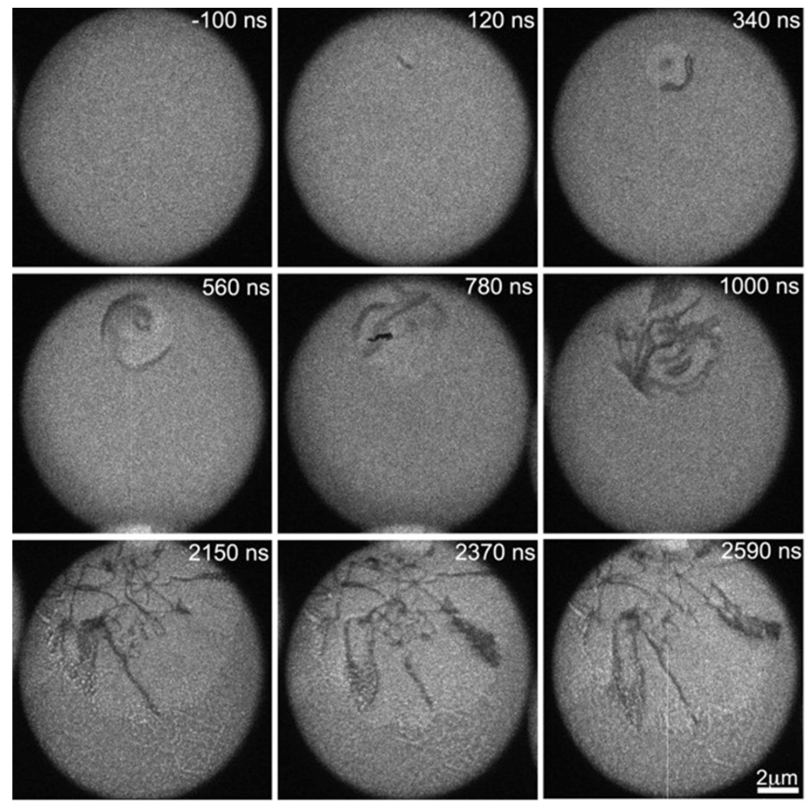
Science opportunities using UEM

PINEM using UEM at Caltech



A. H. Zewail, Science 328, 187 (2010)

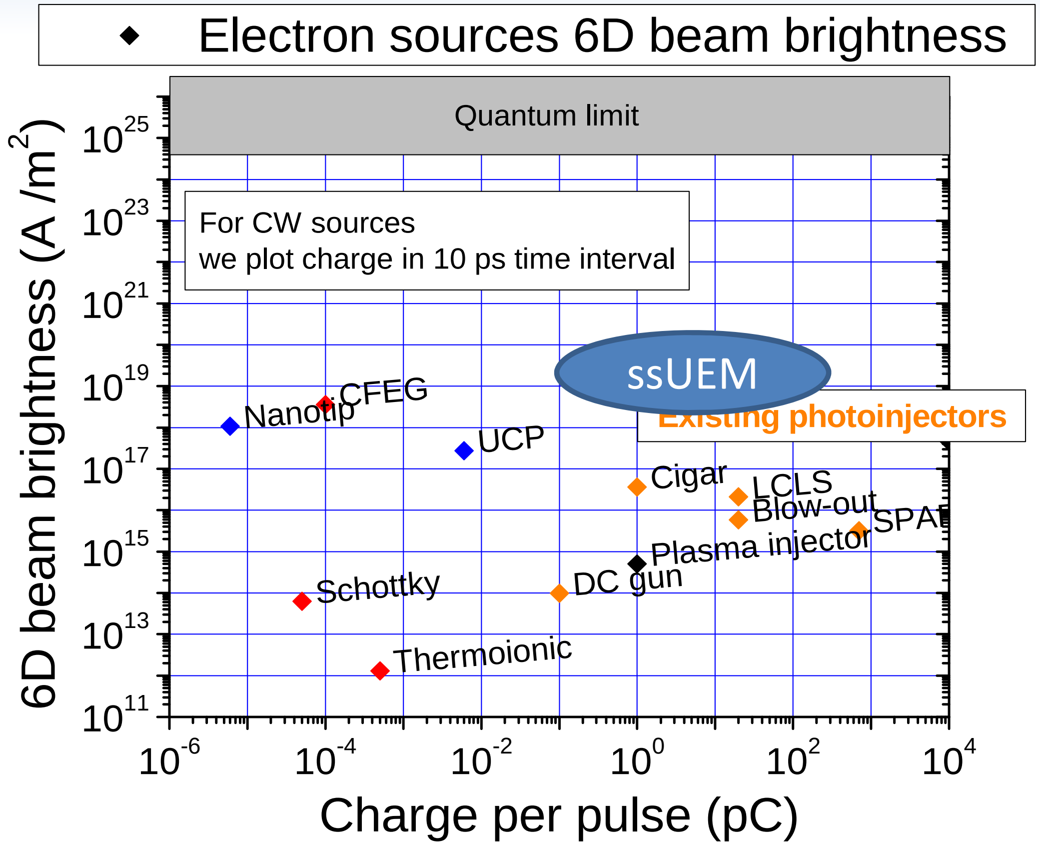
DTEM at LLNL



N. D. Browning et al., in *Handbook of Nanoscopy*, 2012

High beam brightness requirements for single shot UEM

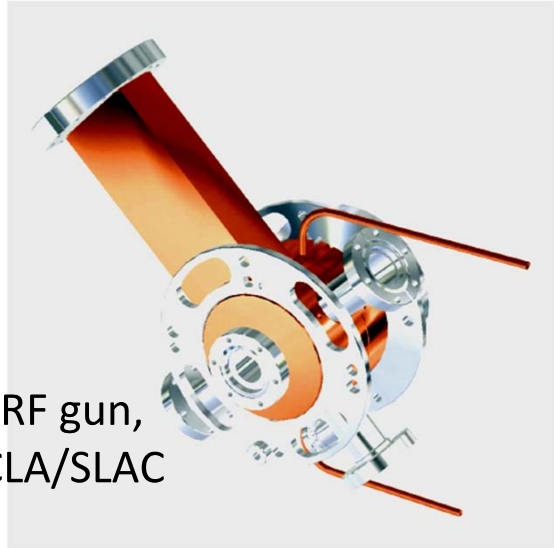
- *temporal resolution (a few ps):*
  - *bunch length:* $\Delta t \sim 1 - 10\ ps$
  - *Timing and synchronization between laser and electron beams*
- *spatial resolution (a few tens of nm):*
  - *high flux at the sample (Rose criterion):* $N \sim 10^{7-8},\ \sigma_x \sim 1\ \mu m$
  - *low angular divergence (related to scattering angle and Cs):* $\sigma_{x'} \sim 1\ mrad$
  - *low energy spread (related to chromatic aberration):* $\frac{\Delta\gamma}{\gamma} < 10^{-4}$



$$B_{6D,\ norm} \sim 10^{19}\ A/m^2$$

The RF photoinjector

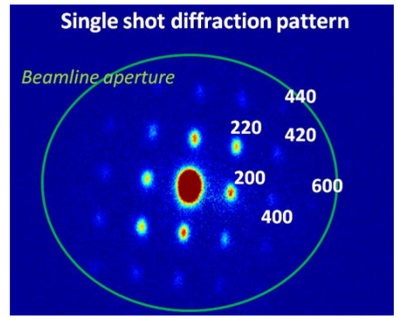
- *State-of-the-art high brightness electron beam source*
  - o Developed for advanced accelerators & FELs
  - o Applications to MeV UED (X. J. Wang, 1996, PRE 54 R3121)
- Photo-emission inside ultrahigh field RF cavity
  - o Peak field $E_0 > 100$ MV/m
  - o 3-5 MeV output energy
- Well characterized – Mature technology
  - – Sub-ps beams possible (response time from metal cathodes is <50 fsec)
  - – Flexible charge (few fC – few nC)
  - – Very low emittance
- Space charge effects suppression
  - – High field at the cathode
  - – High final energy
  - – Beam distribution shaping

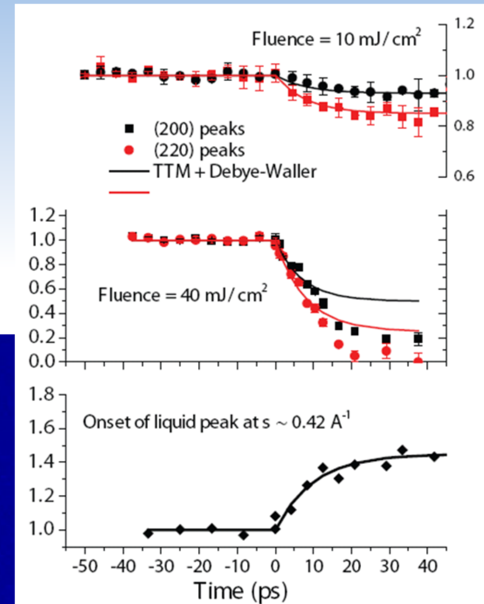


1.6 cell RF gun, BNL/UCLA/SLAC design

MeV UED science

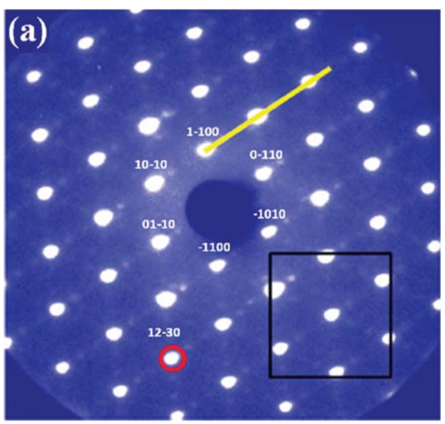
- Time-resolved science with RF photoinjector based UED
- Each example points to a specific MeV advantage
  - ❖ Single shot
  - ❖ Thickness of sample
  - ❖ < 100 fs temporal resolution
- ➢ Recent results from SLAC

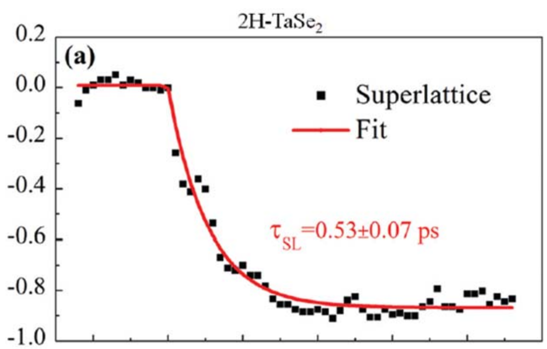


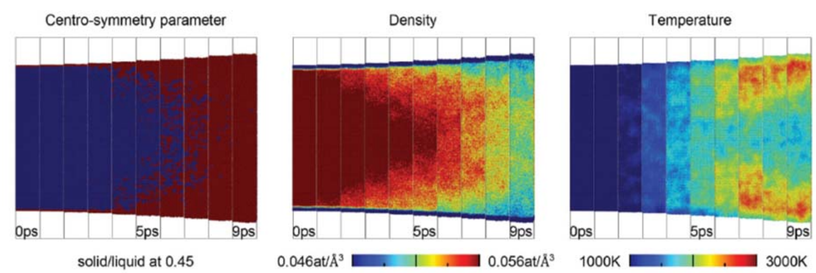


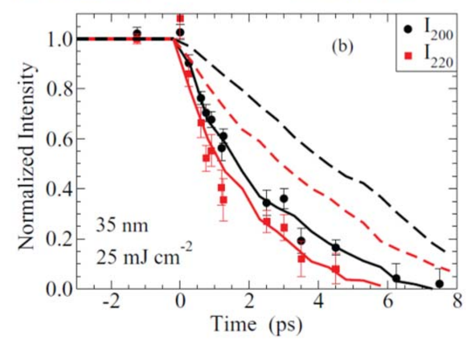
P. Musumeci et al. Applied Physics Letters, 063502 (2010)

P. Zhu et al. Appl. Phys. Lett. 103, 071914 (2013)





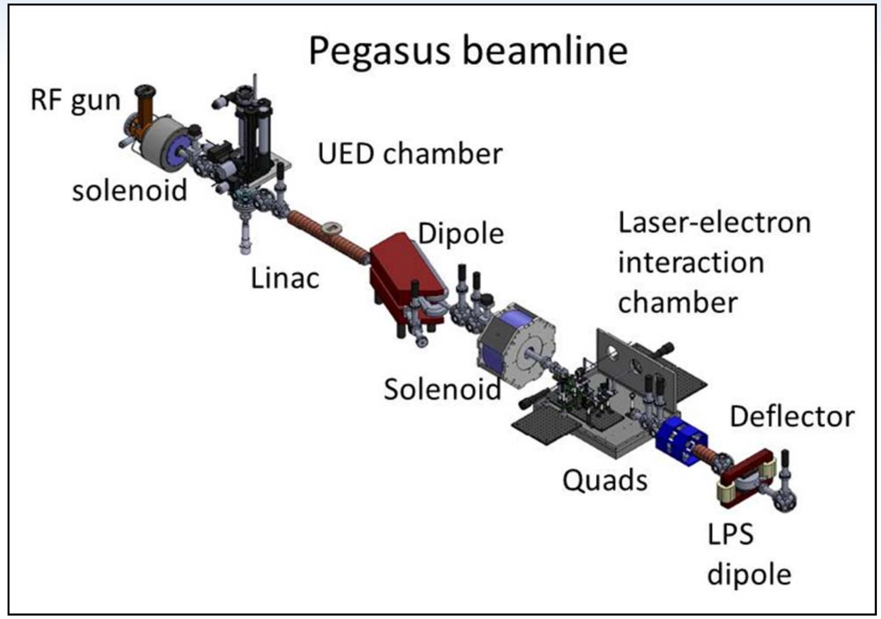




S. Daraszewicz et al.
Phys. Rev. B 184101 (2013)

Compression of MeV electron beams for Ultrafast Electron Diffraction

- Operate photoinjector in blow-out regime
- Use high shunt impedance RF linac to invert longitudinal space charge expansion
- Simulations
- Beam too short for RF deflection resolution
- Develop spectrum-based CTR diagnostics



PHYSICAL REVIEW SPECIAL TOPICS - ACCELERATORS AND BEAMS 18, 032802 (2015)

Generation and measurement of velocity bunched ultrashort bunch of pC charge

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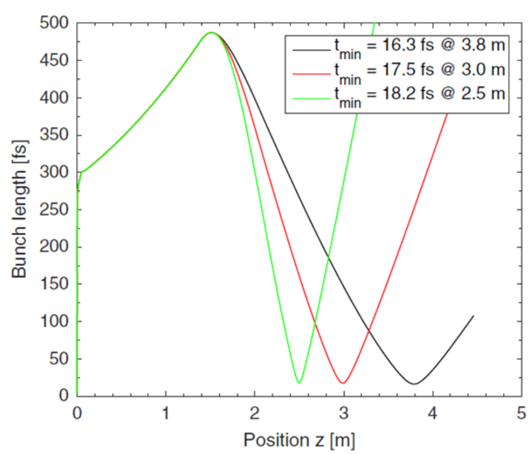
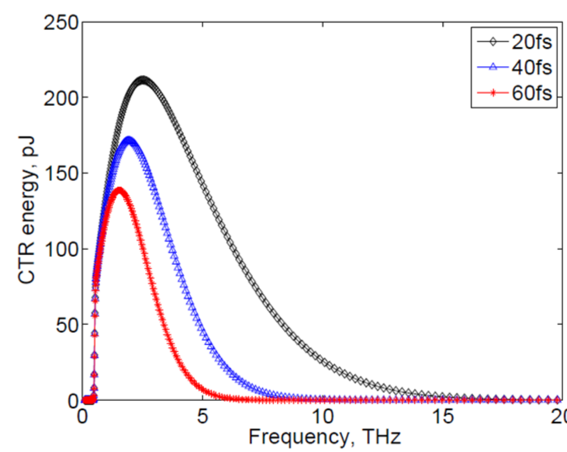
Department of Physics and Astronomy, UCLA, Los Angeles, California 90095, USA
(Received 6 August 2014; published 18 March 2015)





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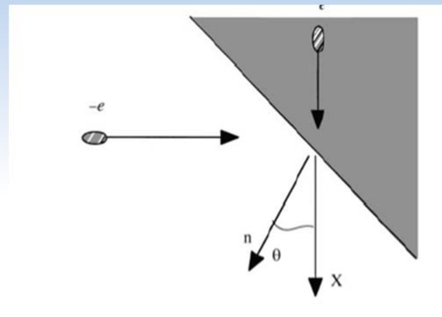
Sub-50 fs bunch length diagnostic

- Use Coherent Transition Radiation
  - pJ of energy expected
- Liquid He cooled bolometer detector
- Maximize CTR signal with linac phase
- Strong dependence on transverse spot size
- Use two filters at 1 THz and 5 THz to reconstruct spectral content
- Very sensitive to RF – laser jitter !

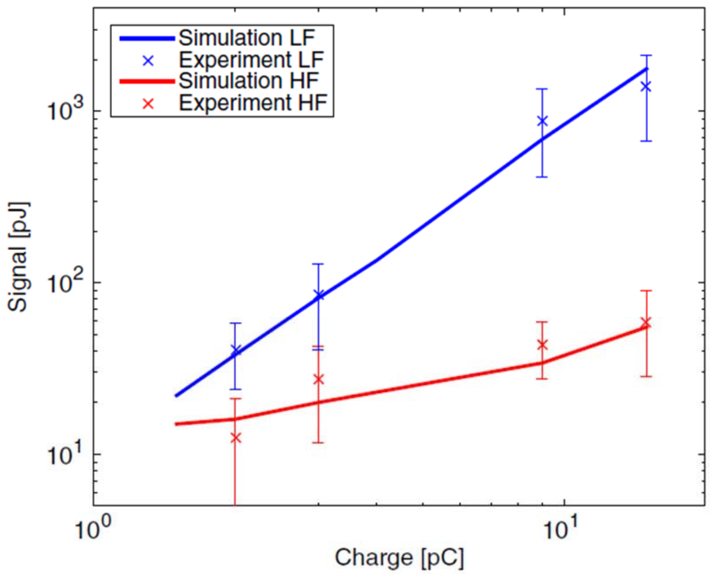


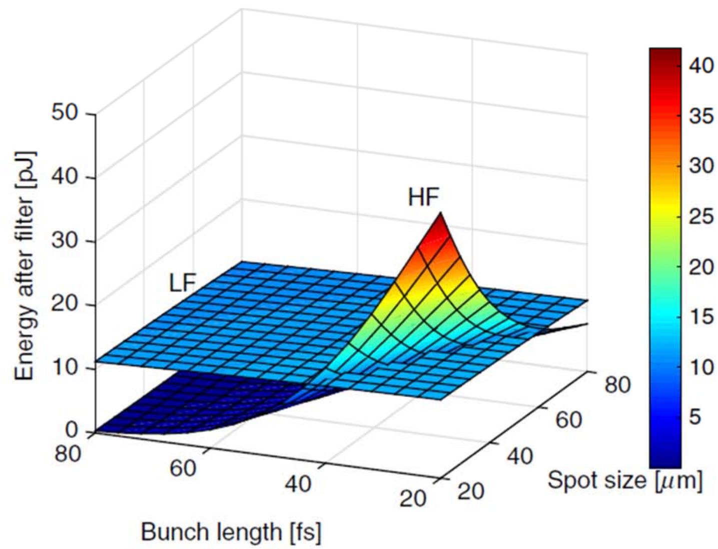
A. Murokh et al./Nucl. Instr. and Meth. in Phys. Res. A 410 (1998) 452—460

He cooled bolometer

TABLE III. Measured electron pulse duration.

Charge	Bunch length	Simulation
2 pC	33 fs–51 fs	35 fs
3 pC	18 fs–44 fs	38 fs
9 pC	50 fs–63 fs	71 fs





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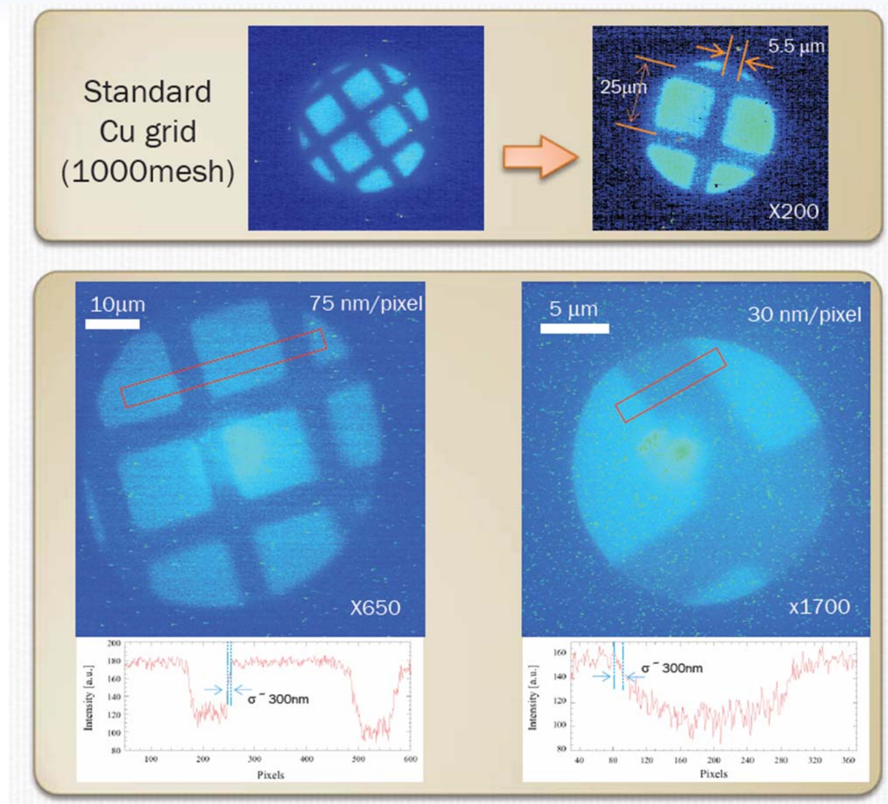
Worldwide R&D on MeV UEM: following MeV UED steps

Prototype MeV UEM at Osaka Univ.

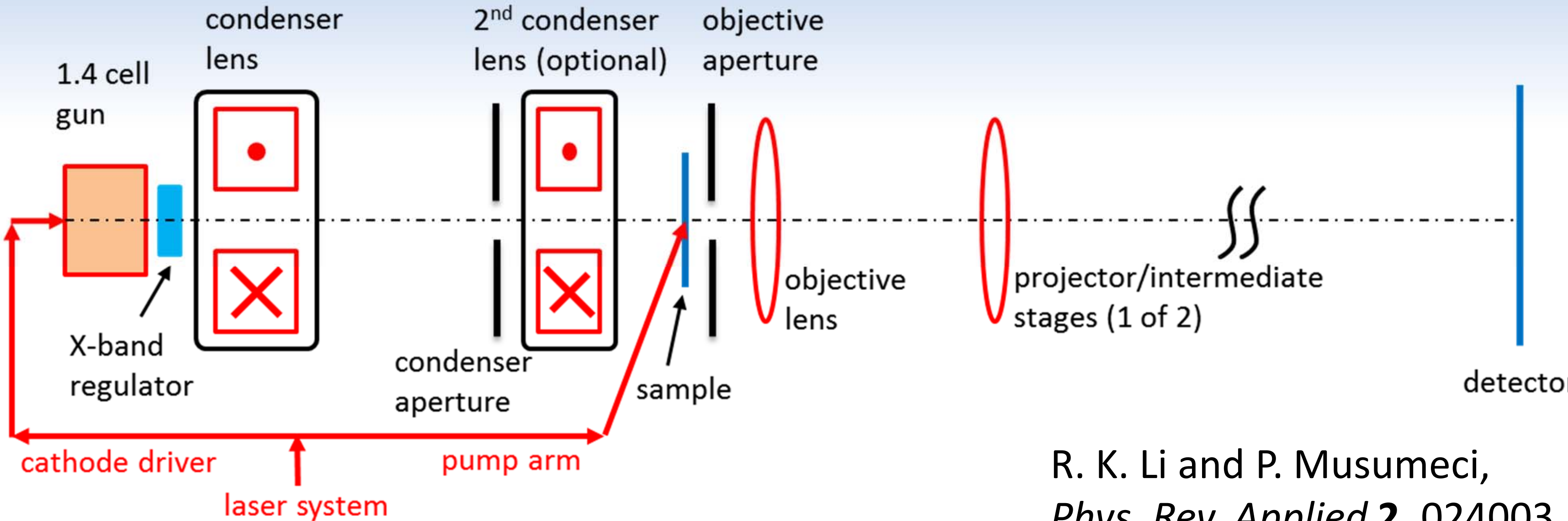
Courtesy of Jinfeng Yang



See also D. Xiang, et al., Nucl. Instrum. Methods Phys. Res., Sect. A 759, 74 (2014).

similar UEM/UED projects in China and Germany! Now SLAC and LBNL

UCLA concept for MeV-ps microscopy



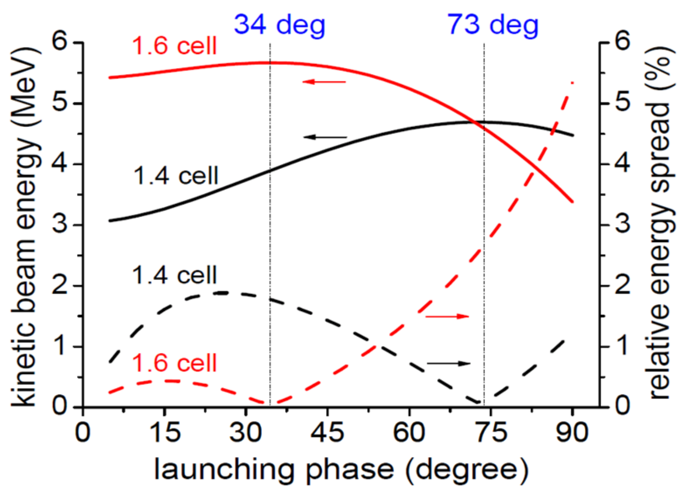
R. K. Li and P. Musumeci, *Phys. Rev. Applied* **2**, 024003, 2014

- *high gradient S-band rf gun (1.4 cell gun)*
- *cigar-shape low emittance beam (small spot UV laser)*
- *X-band rf regulation cavity for energy spread silencing*
- *strong PMQ- based electron lenses*
- *high efficiency detection of MeV electrons R. K. Li et al., JAP 110, 074512 (2011)*
- *rf amplitude and phase control (at 1×10^{-4} and 0.01 degree level)*
- *Full start-to-end numerical studies of aberrations and e-e interactions*

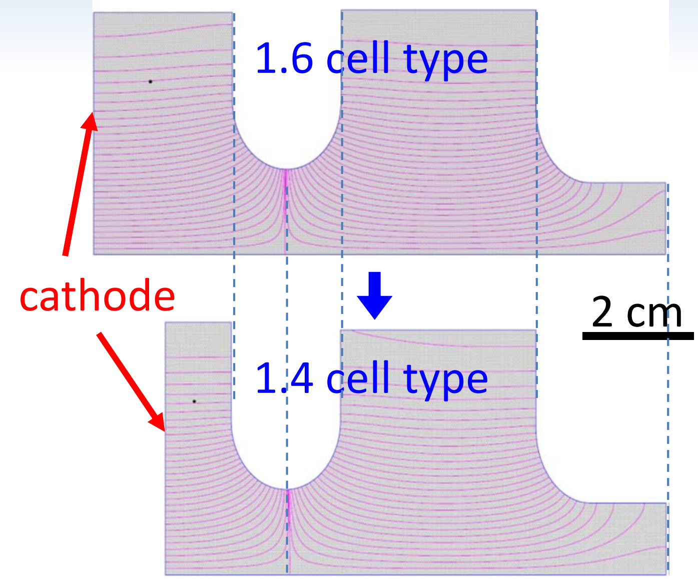
Maximize extraction field in the gun

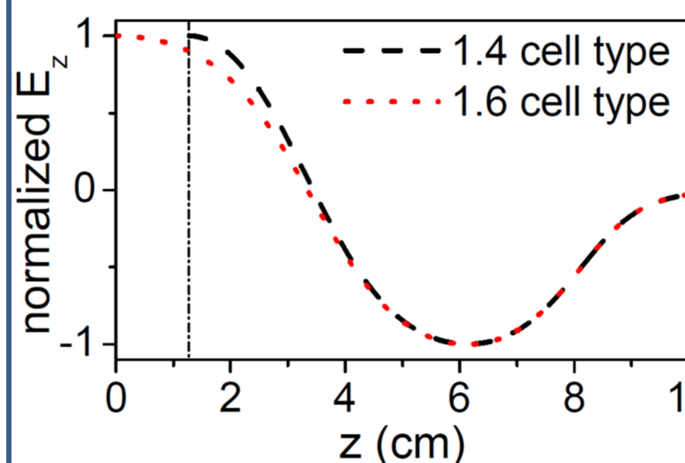
- Traditional (1.6 cell) photocathode rf gun optimized for high charge, high final output energy
- Optimal launch phase for 1.6 cell gun is 30˚
- E-field at extraction (critical to beam brightness) is only half (sin30˚=0.5) of the available acceleration gradient of the gun
- Shortening the photocathode cell ⟹ higher launch phase, e.g. 70˚, (sin70˚=0.94), ⟹ ×2 improvement in brightness

Comparing longitudinal dynamics



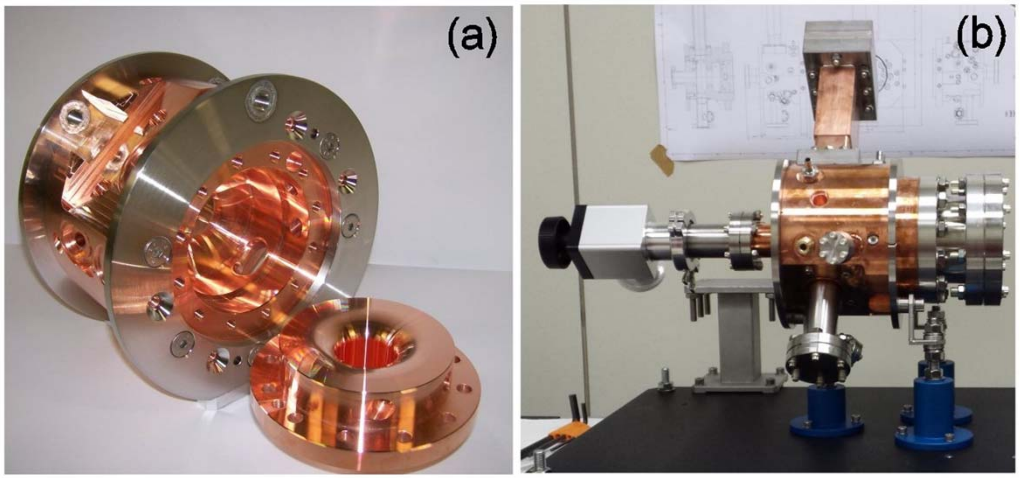
Shorten the photocathode cell



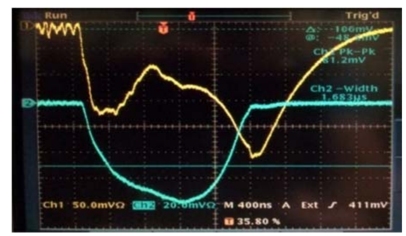


1p4 cell gun development at UCLA

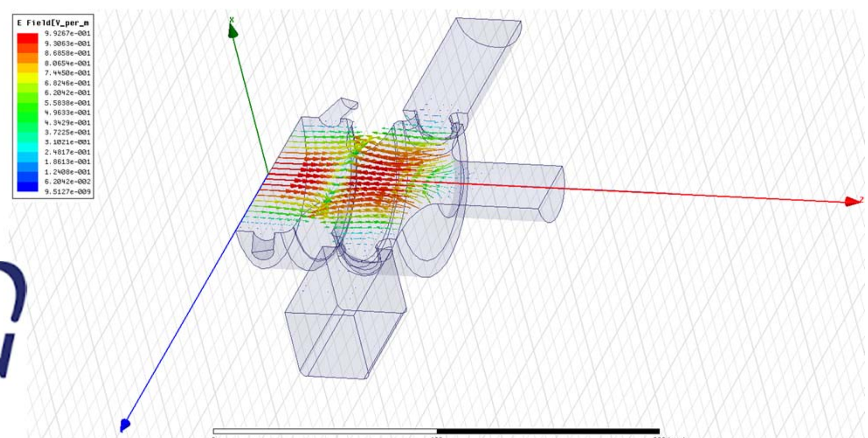
- Clamped RF gun by INFN

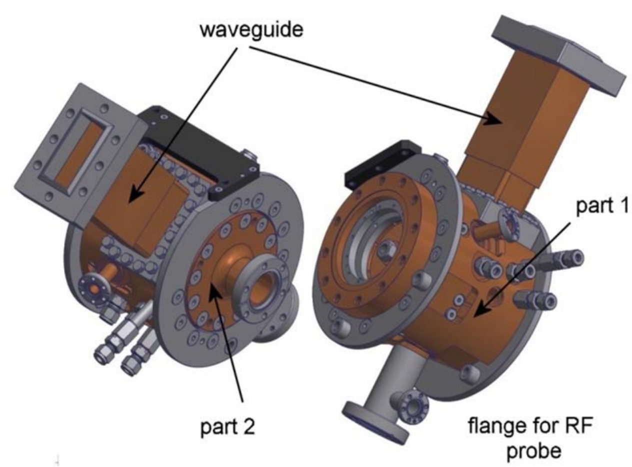


- ✓ Commissioned at UCLA Pegasus in June 2014
- ✓ ~95 MV/m. Limited by available RF power
- ✓ Reliable operation
- ✓ Replaceable cathode



- 1p4 cell gun development



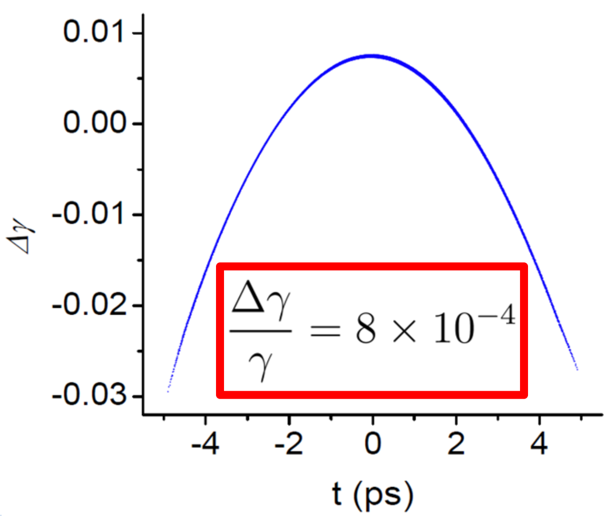


Ultralow energy spread: *rf-based compensation*

- Beam rf curvature from rf guns
  - beam energy depends on launching phase (time)
  - beam energy spread dominated by the rf curvature
  - slice energy spread much smaller

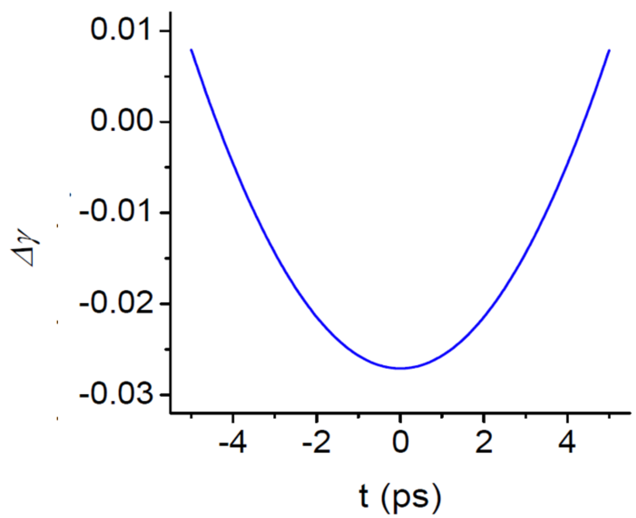
γ-t at S-band gun exit

$$\gamma(t) = \gamma_0 \cos\omega_0 t = \gamma_0 (1 - \frac{\omega_0^2 t^2}{2})$$



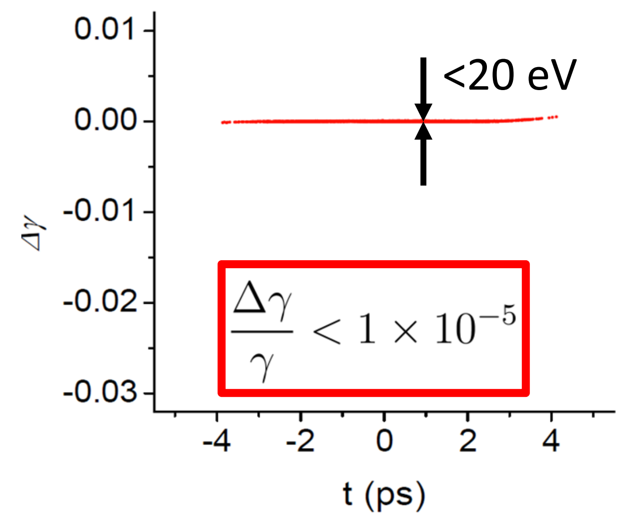
deceleration in X-band cavity

$$\gamma_X(t) = -\gamma_X (1 - \frac{k^2 \omega_0^2 t^2}{2})$$



final γ-t distribution

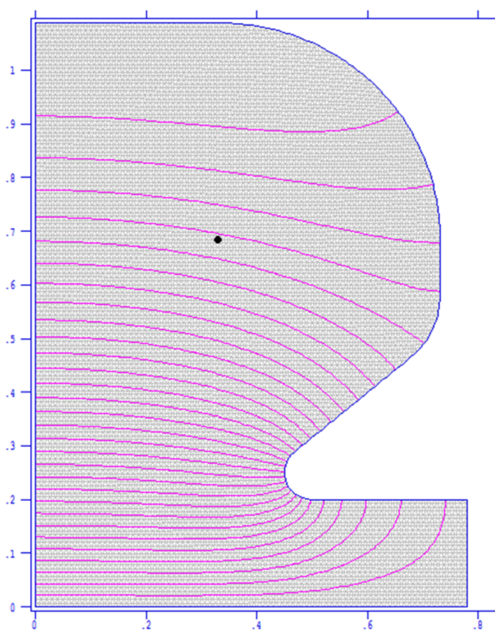
$$\gamma_X = \gamma_0 / k^2$$

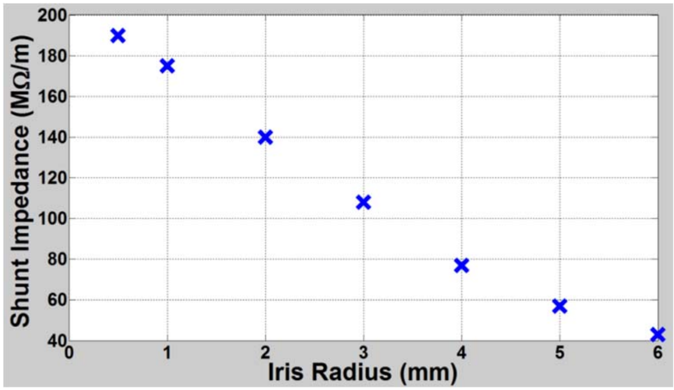


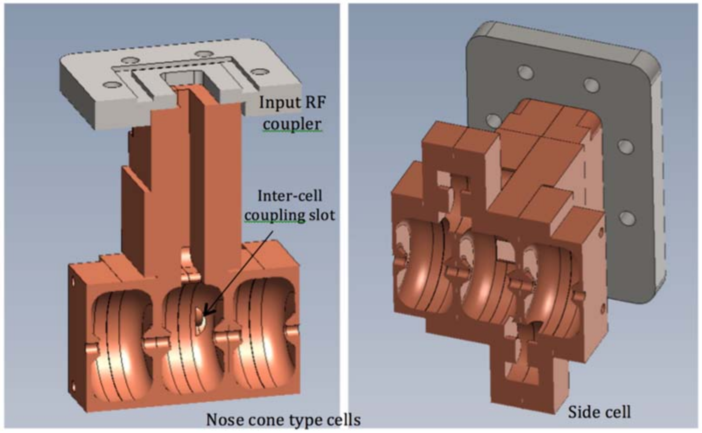
- *larger $k \equiv \omega_X / \omega_0$ works better (less deceleration, less power)*
- *performances limited by rf amplitude and phase stabilities*

High shunt impedance RF linearizer design

- 9.6 GHz 50 kW power source available
- **3 cells with 4 mm iris diameter = > 400 kV**
- Main challenge is two-frequency phase stability



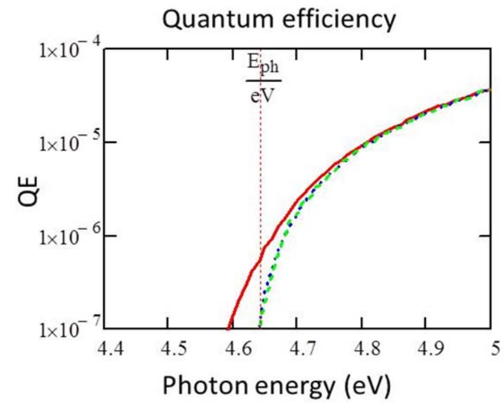


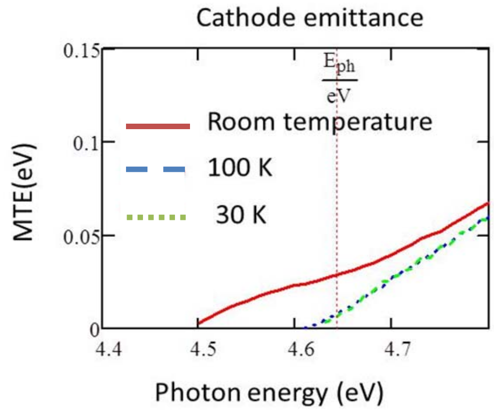


Cell length at 9.6 GHz ~ 1.56 cm

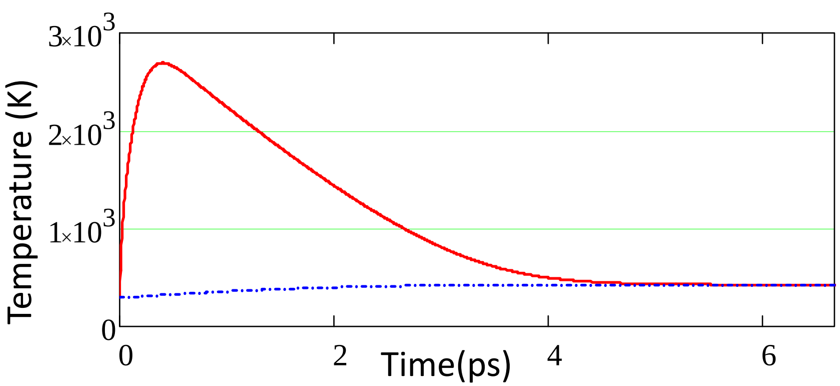
Cold metal cathodes

- A solution for high density low thermal emittance?
- Recent results from Cornell (L. Cultrera et al., arXiv 1504.05920 2015) indicate record low emittances for alkali-antimonide
- Dowell and Schmerge (*PRST-AB 12, 074201 (2009)*) formulas suggest similar benefits for metals





- **Ultrafast heating of metals**
  - What is the electronic temperature at emission?
  - Long cigar aspect ratio takes advantage of electron-lattice coupling



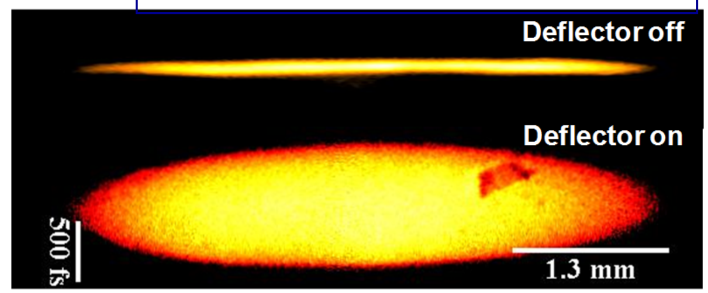
QE 10^{-5}

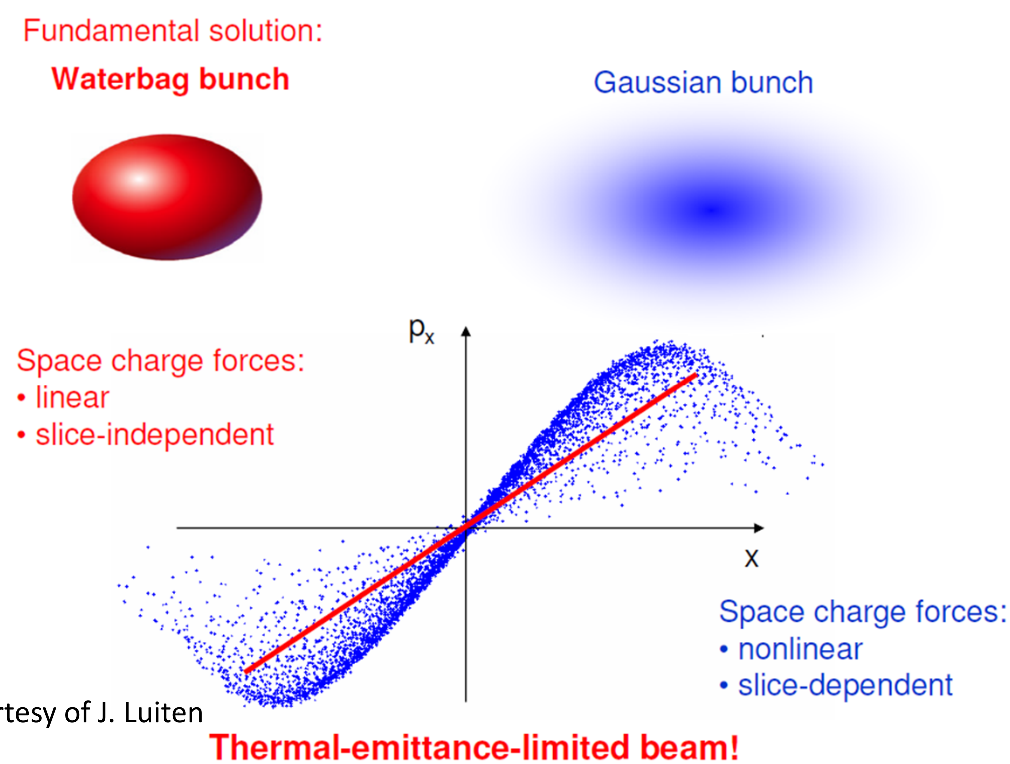
Q = 10 pC

Preserve brightness during transport

- Uniformly filled ellipsoidal distribution is the ideal case.
- How to create these distributions:
  - Illuminate cathode with short pulse and use longitudinal space-charge expansion (PANCAKE)
  - Illuminate cathode with long and skinny pulse and use transverse space-charge expansion (CIGAR)
  - Direct ellipsoidal laser pulse shaping (not yet demonstrated)

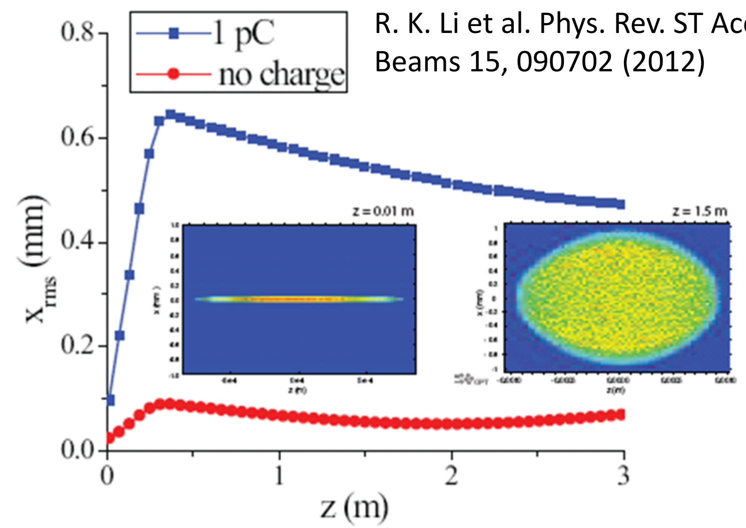
P. Musumeci et al. *Phys. Rev. Lett.* **100**, 244801 (2008)





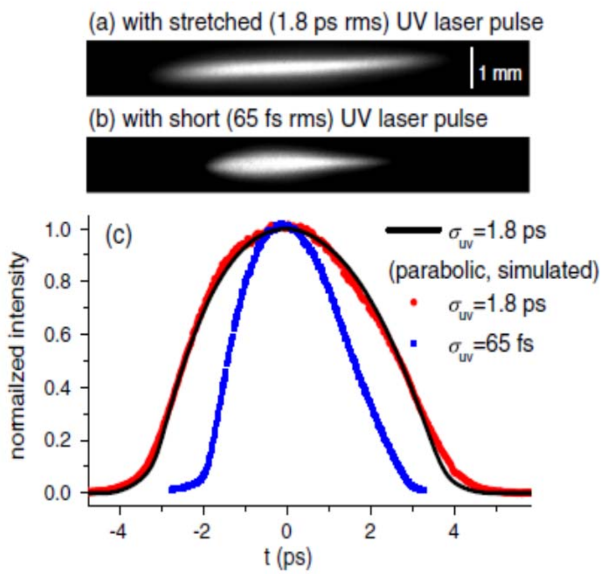
Courtesy of J. Luiten

R. K. Li et al. Phys. Rev. ST Accel. Beams 15, 090702 (2012)

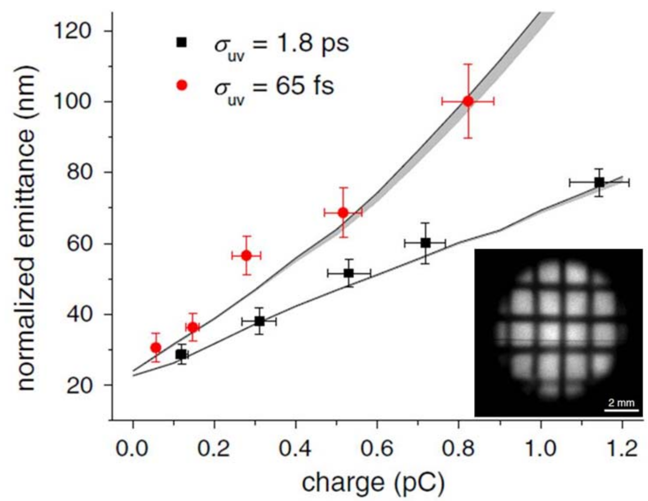


Cigar-beam: pC charge, ultralow emittance

- Cigar-beam: long (10 ps) and narrow (<30 μm spot size)
  - the aspect-ratio can beat the ‘virtual cathode limit’ D. Filippetto et al., PRSTAB
  - very small intrinsic emittance from the cathode Claessens et al., PRL (2005)
  - transverse and longitudinal dynamics are essentially decoupled



R. K. Li et al, PRSTAB 15, 090702 (2012)



‘grid method’ measure very low (~1 nm) geometric emittance

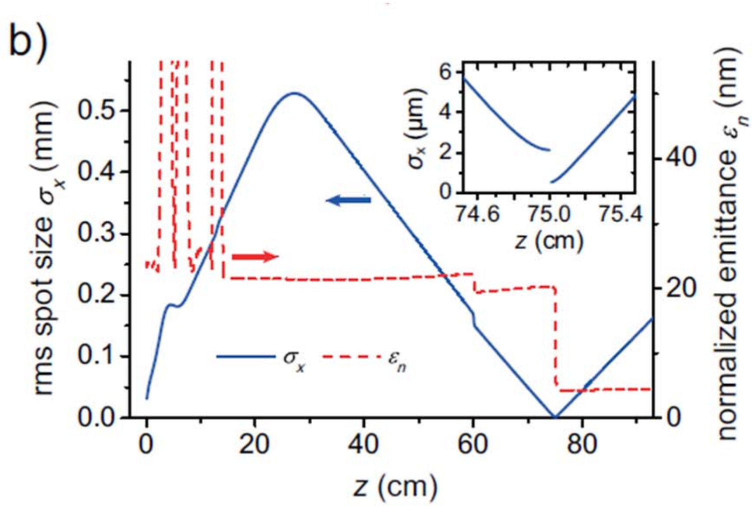
Simulations shows we can generate 5 MeV, 1 pC charge, 3 ps rms width, <10 nm normalized emittance, <1 um rms spot size at the sample.

RF photoinjector based condenser stage

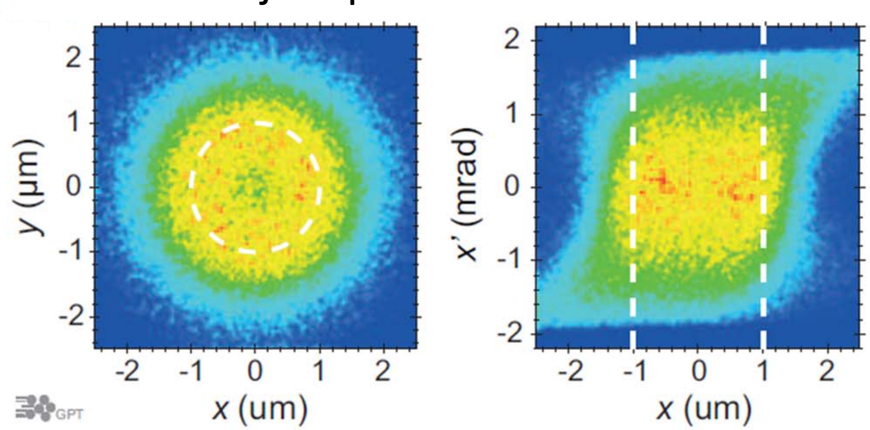
- Could be used for nanodiffraction
  - Tune phase of cavity to compression
- Optimization:
  - Higher gun field
  - Cathode thermal emittance
  - Space charge induced dynamics
- More flexibility with two solenoids

TABLE II. Parameters for the operation of the electron source and the electron beam qualities at the sample.

Parameters	Values
gun gradient	120 MV/m
gun phase	73 degree
initial beam charge	6.0 pC
UV spot size, rms (spherical)	20 μm
UV pulse duration, FWHM	10 ps
X-band cavity gradient	43.3 MV/m
At the sample (within the 2.0 μm diameter area)	
beam charge	1.0 pC
transverse momentum spread $\gamma\sigma_\theta$, rms	9.5×10^{-3}
bunch length, FWHM	10 ps
kinetic beam energy	4.4 MeV
relative energy spread $\delta\gamma/\gamma$, FW50	1.5×10^{-5}

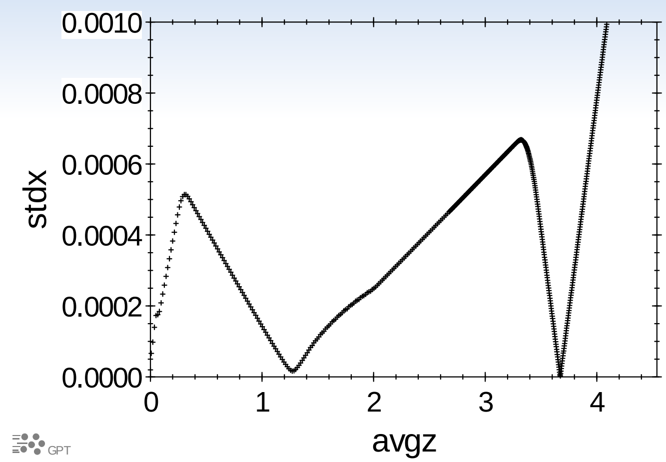


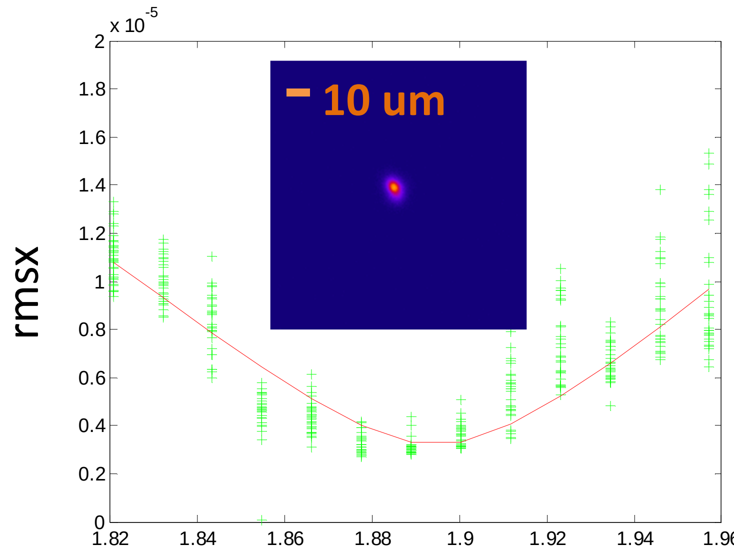
Object-plane distribution



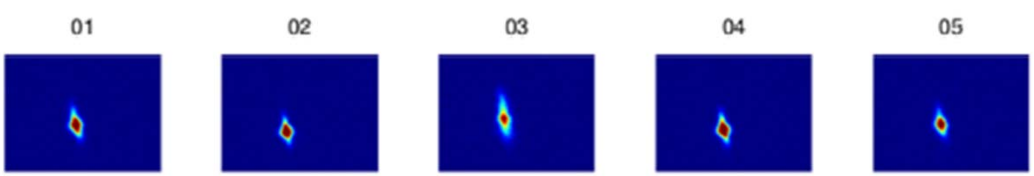
Experiments at UCLA

- Two-solenoid condenser stage
- **3 um x 3 um** beam demonstrated.
  - Limited by depth of focus
- Solenoid scan yields
  - **30 nm normalized emittance @ 100 fC**
- Addition of pinhole to shape distribution and improve stability
  - RF amplitude - $1.5 \cdot 10^{-3}$
  - RF phase - 0.1 degrees
  - laser pointing on virtual cathode - 3 μm
- Microscope objective + 30 micron thick YAG screen enable high resolution spatial profile measurements





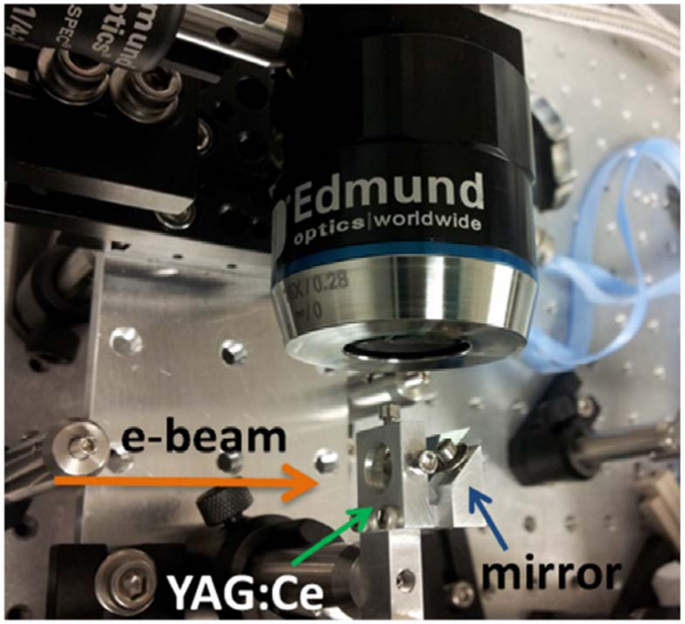
Solenoid field



5 μm rms pointing stability

Micron beam size diagnostic development

- In-vacuum microscope objective.
  - Thinner fluorescent screen
  - TR?
- USE TEM-2000 grid as a knife-edge / emittance measurement
- Sub ps-beam point-projection microscope with micron resolution



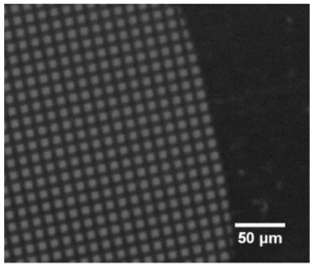
$$R = \frac{0.61\,\lambda}{NA}$$

$$NA = 0.28$$

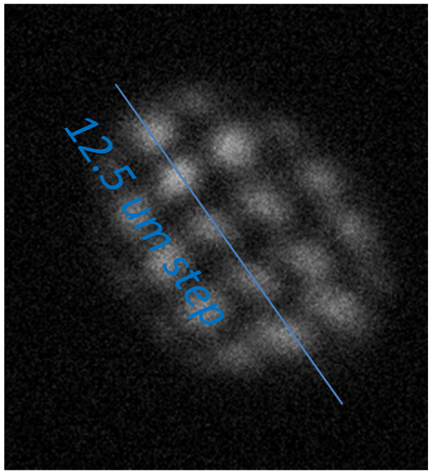
$$\lambda_{YAG:Ce} = 550\ nm$$

$$\Rightarrow R = 1.2\ \mu m$$

Optical image of TEM 2000 grid



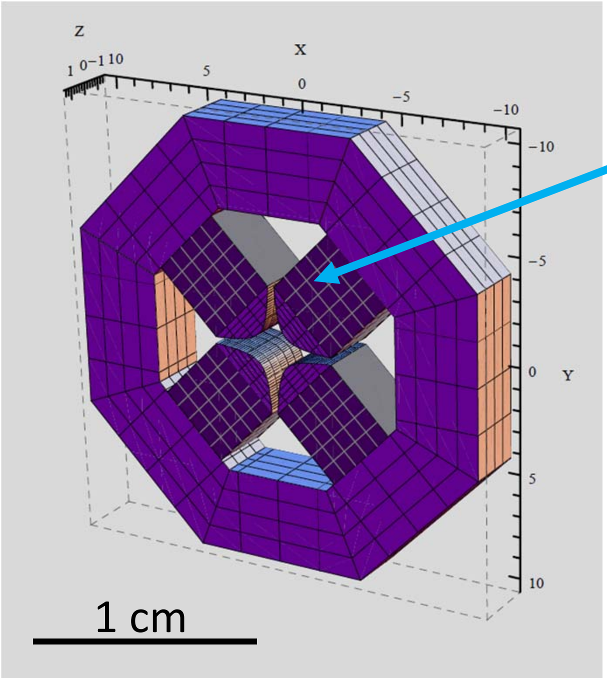
150 nm per pixel



Strong lenses for MeV beams

- *for conventional TEMs (<300 keV), solenoid lenses are used*
- *solenoid is symmetric, but very ineffective for high energy electrons*
- *Cs and Cc are roughly equal to the focal length*
- *heavy and bulky NC solenoid (≤2.2 T), or SC solenoid can be used*
- *PMQ quadrupoles are strong and compact*
- *Compare aberrations for different designs*

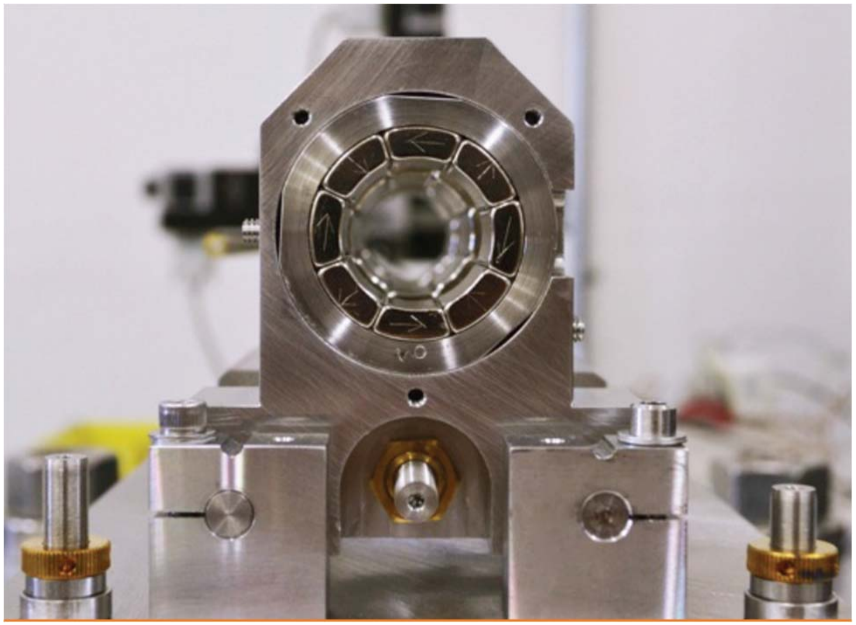
Cube design



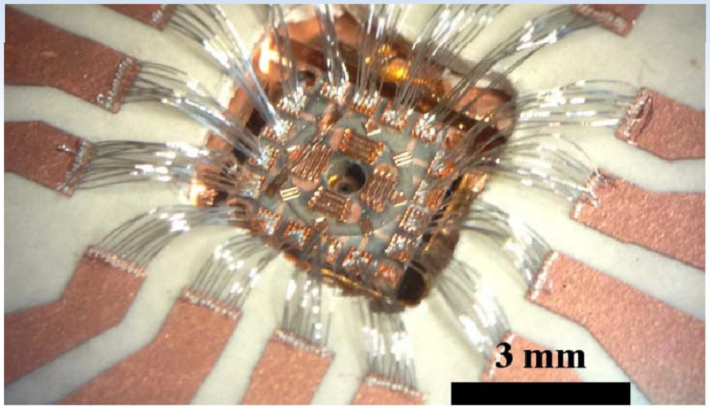
permanent magnets, B_0=1.40 T

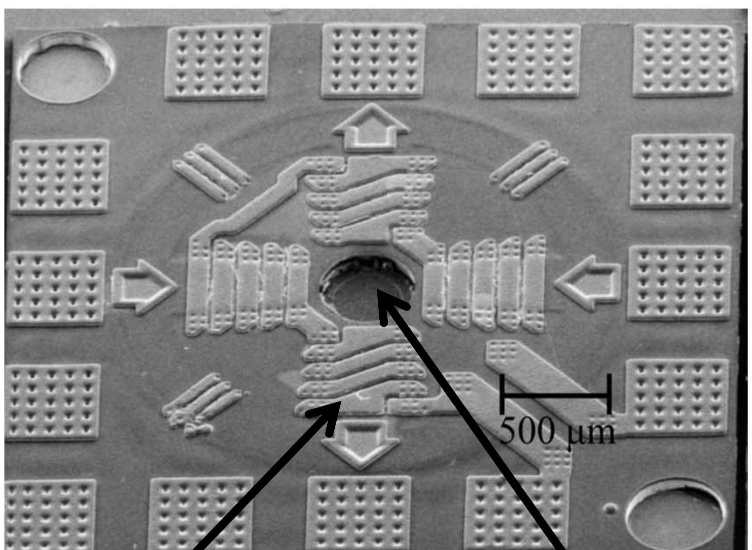
Gradient G~500 T/m

Halbach design



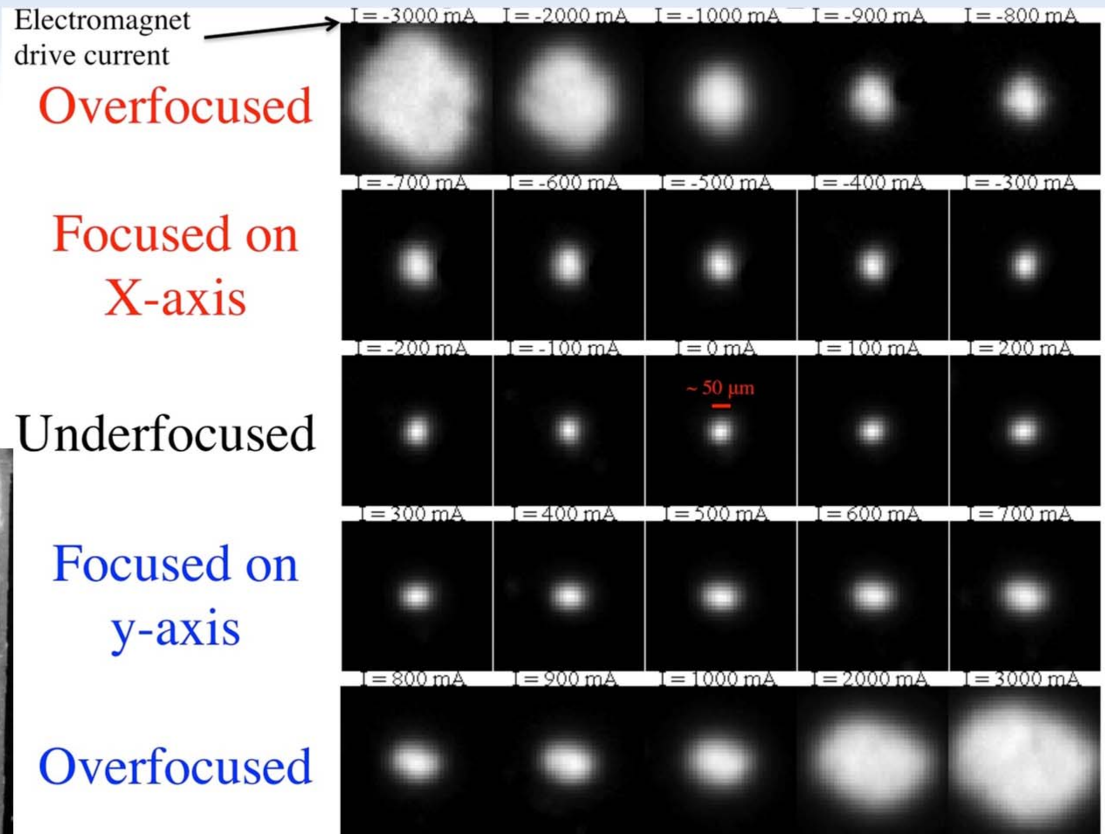
Microfabricated quadrupoles





Windings (top)

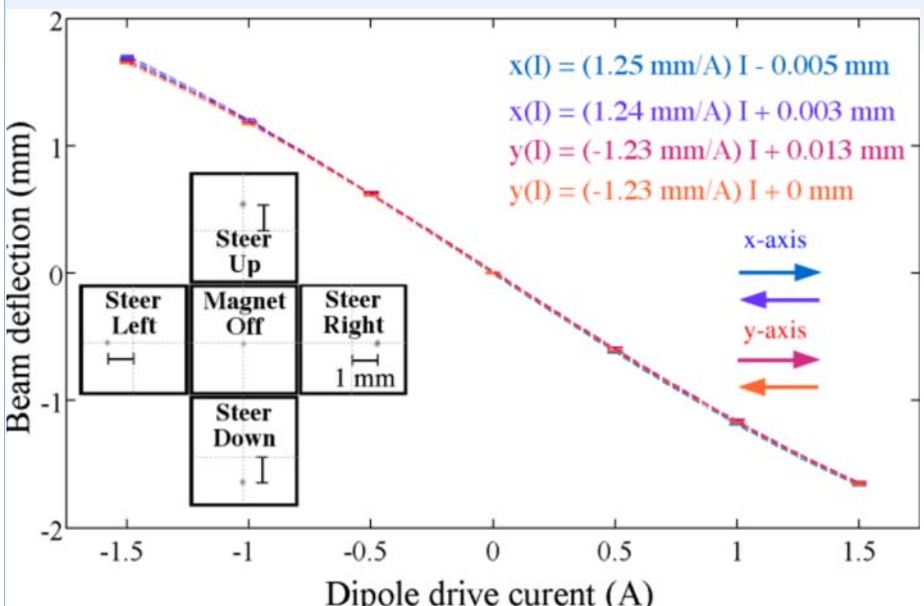
Bore (not yet etched)

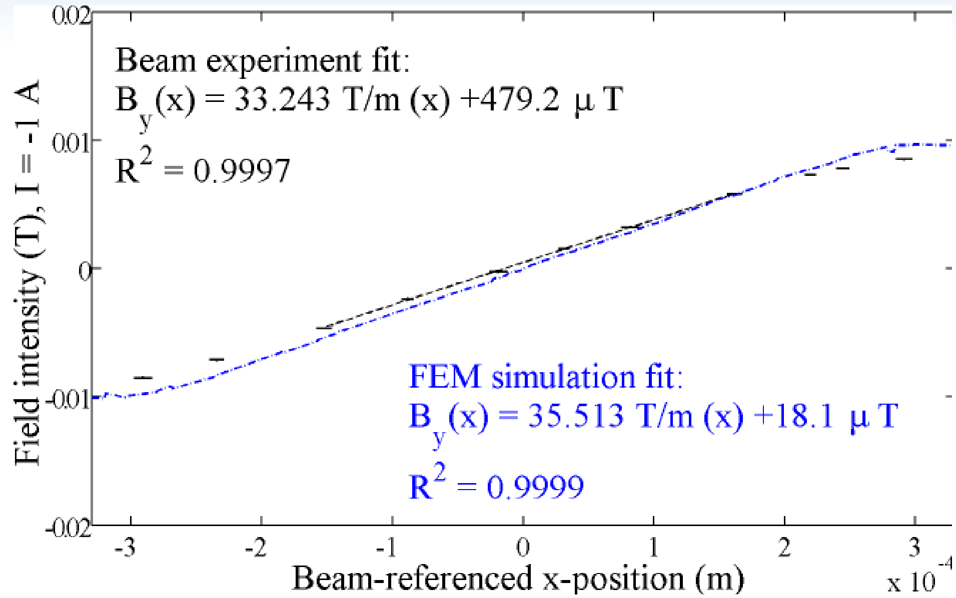


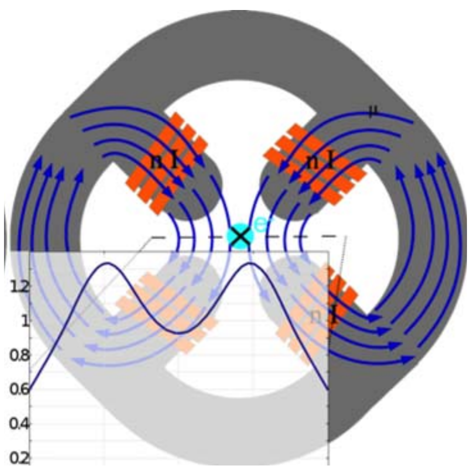
J. Harrison et al., Phys. Rev. ST Accel. Beams 18, 023501 (2015)

J. Harrison et al. J. Microelectromech. Syst. 23, 505 (2014)

First MEMS quads work!
(new batch to improve focusing strength)

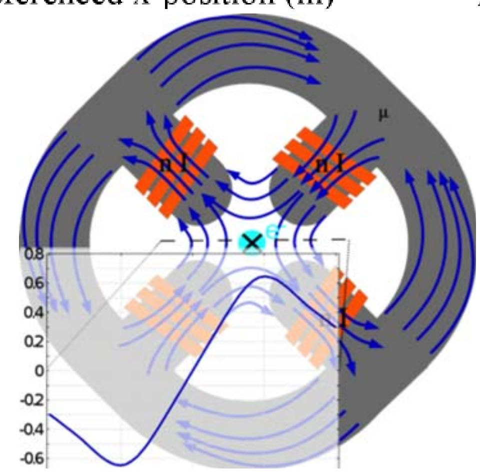






Dipole mode

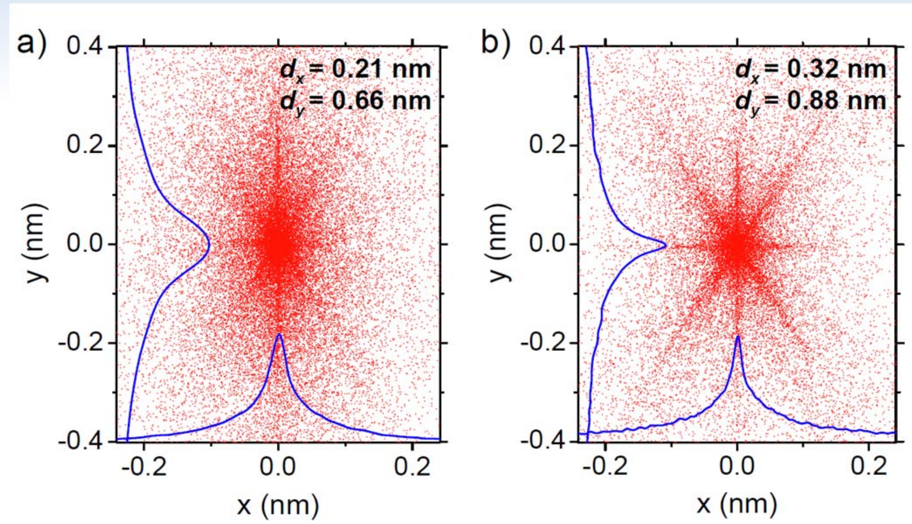
Magnet gap	Yoke length	Pole taper	Peak field gradient	Magnetic length	Winding current
0.6 mm	**0.055 mm**	**7.5°**	**253 T/m**	**0.477 mm**	**4.6 A**
0.6 mm	0.2 mm	35°	1.0 kT/m	0.504 mm	11.0 A
0.4 mm	0.2 mm	40°	2.2 kT/m	0.387 mm	9.6 A
0.2 mm	0.2 mm	30°	6.1 kT/m	0.281 mm	6.1 A
0.1 mm	0.2 mm	30°	10.0 kT/m	0.245 mm	2.9 A



Quadrupole mode

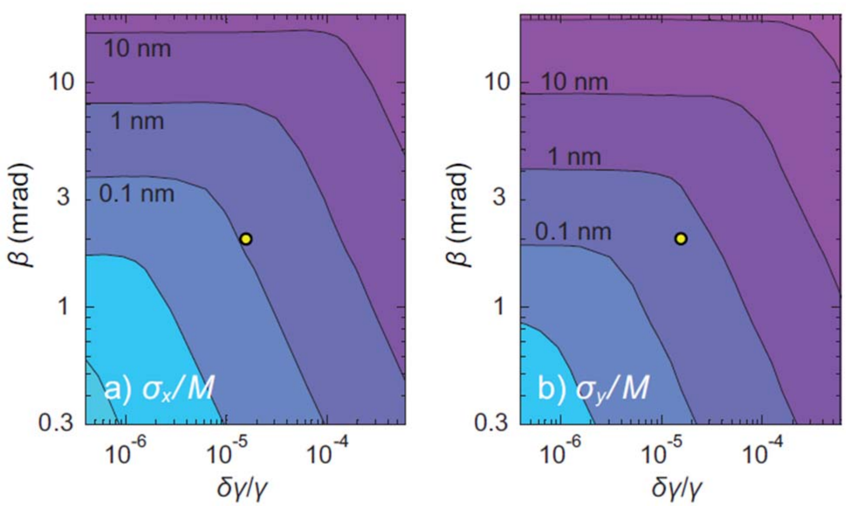
Spatial resolution limits

ideal hard-edge PMQ **3D PMQ field map**



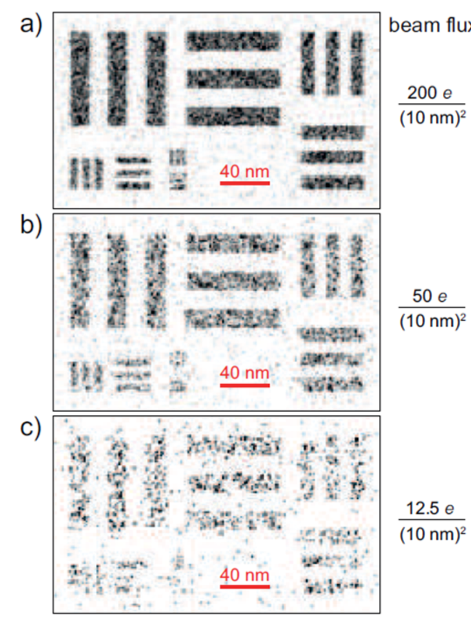
- *resolution determined by*
  - *- aberrations*
  - *- electron flux*
  - *- e-e interactions*
- *need reliable numerical tool*

Chromatic and spherical aberrations:
Thin quadrupoles case



Rose criterion:

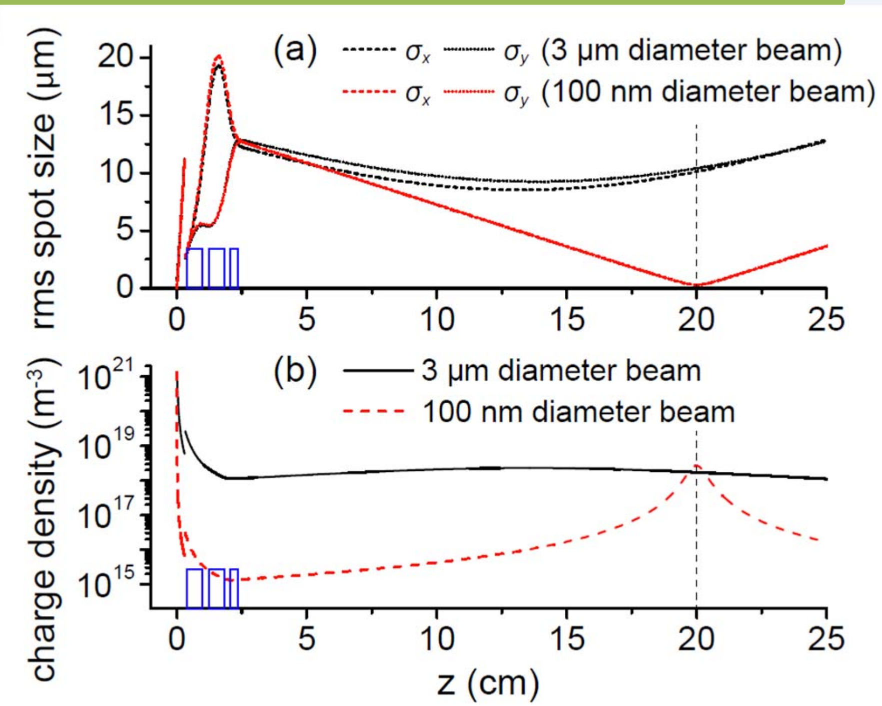
- ❑ Beat Poisson noise on detector
- ❑ > 100 e- / d^2

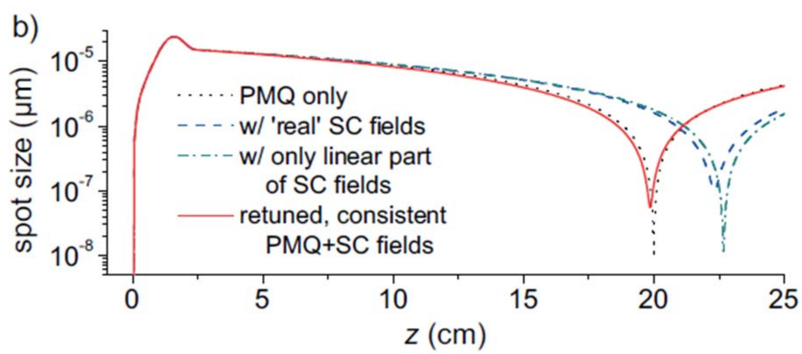


Space charge effects in transmission electron microscope column

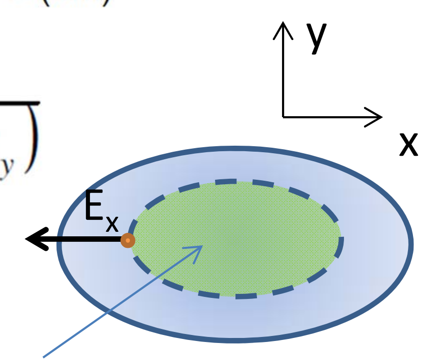
- *Space charge: smooth + stochastic part*
- *Full scale simulation cumbersome and not easy to optimize*
- *Scaled simulation is usually incorrect !!!*
- *Novel algorithms to estimate space charge effects on resolution*
- *Separate smooth space charge contribution*
  - *Linear effect: retune lenses*
  - *Non linear aberrations*

Example: same divergence, 1/30 spot size





$$E_x \approx \frac{e\lambda}{2\pi\varepsilon_0}\frac{x}{\sigma_x\left(\sigma_x+\sigma_y\right)}$$

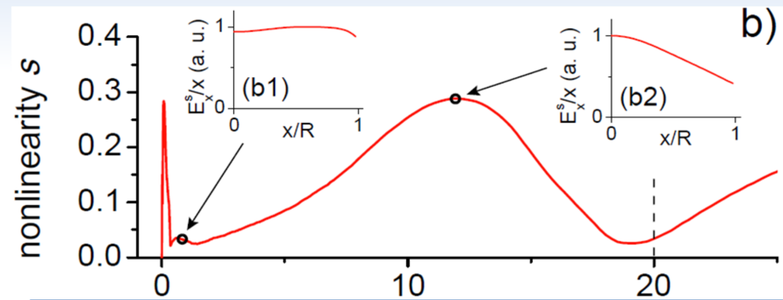


λ enclosed current

Non linear space charge aberrations

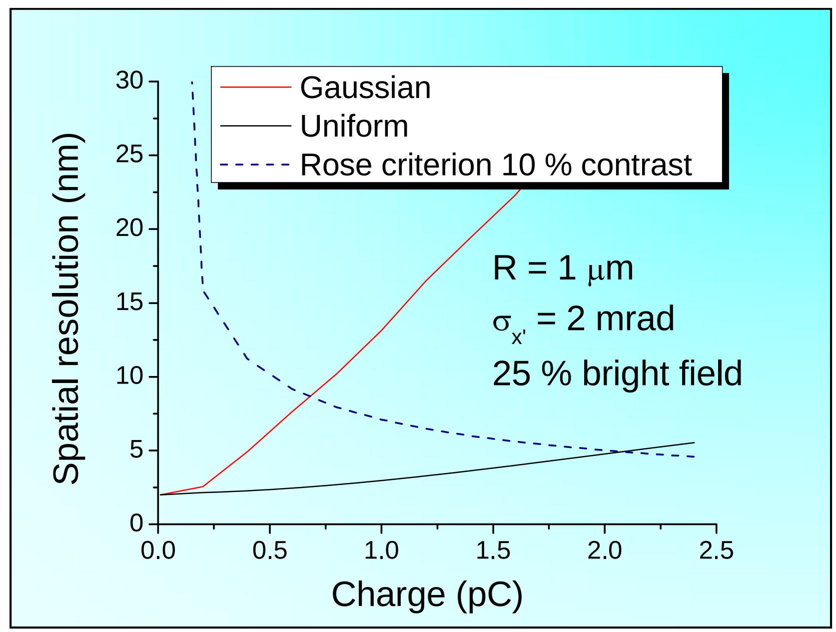
- GPT – based:
  - Pre-calculate space charge field using fewer macroparticles
  - Track full particle numbers into external field map
- Matlab-based microscope design tool: *SC-TEMlab*
  - Design beam line using matrix formalism with linear space charge approximation
  - Particle tracking
    - Effect of aberrations using particle tracking
    - Effect of Poisson noise on detector
    - Simulation of full image
    - Can take into account stochastic scattering !
    - Fast !
- Depend on angular distribution at samples
- Optimum charge level to maximize Rose resolution and minimize space charge aberrations.

Track beam shape and SC field profile



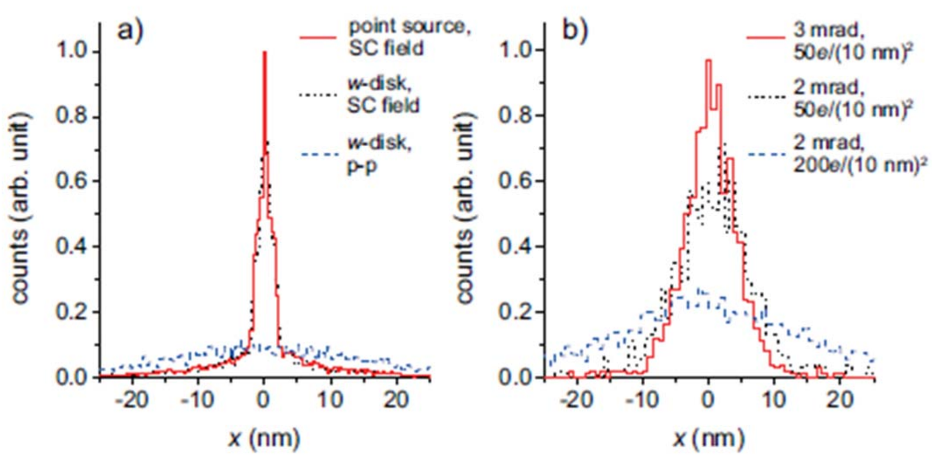
SC forces are only linear when charge density is uniform (only at object and image planes)!

Resolution can be further optimized by alternative beam optics design introducing dynamics higher order corrections.



Point spread function and spatial resolution estimate

- Two different ways to calculate:
  - o Size of a point source at the image plane
  - o w-disk of correlation function (final position vs. magnified initial position i.e. *FW50*$(x_f - M\,x_i)$
- Point-to-point stochastic scattering effects are a major contribution.
  - In GPT point-to-point using Barnes-Hut algorithm
  - In SCTEMlab. Stochastic heating effect



Object plane

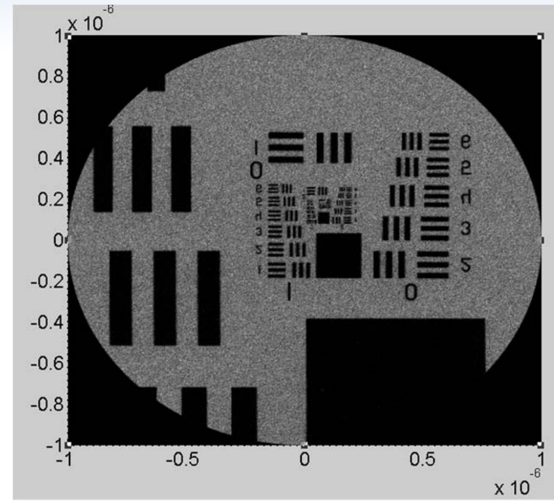
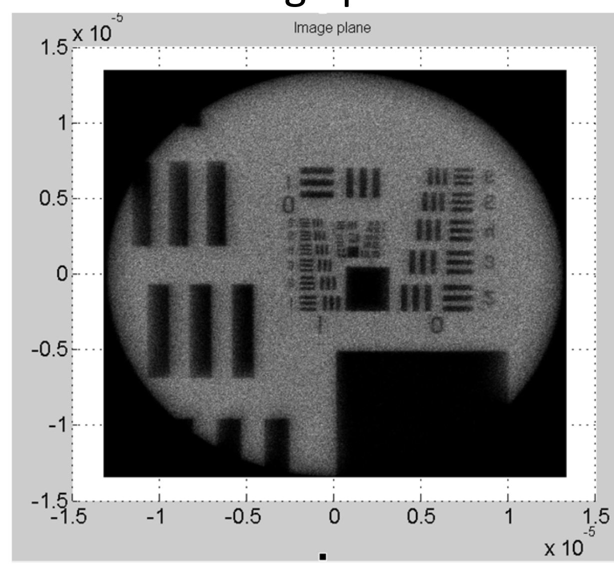


Image plane



Conclusions

- MeV time-resolved microscopy is an exciting novel application of high brightness beams
- Push to the limits RF photoinjectors as high brightness electron sources
- Condenser stage can be used for femtosecond relativistic electron nanodiffraction !
- Electron optics development opportunities
  - PMQ quads
  - MEMS quads
- Space charge effects in microscope column