Frontiers in ultrafast electron diffraction instrumentation

P. Musumeci

UCLA Department of Physics and Astronomy

Acknowledgements

- <u>UCLA Pegasus Laboratory group</u>

  E. Cropp, P. Denham, S. Crisp, A. Fisher, M. Lenz, D. Garcia, A. Kulkarni. Graduate students.

  A. Ody. Development engineer

- <u>Collaborators:</u> R. K. Li, A. Kogar, J. Maxson, D. Filippetto, F. Carbone, G. Andonian, A. Murokh, A. Minor, J. Luiten, S. Karkare

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  - NSF Accel Science PHY-1734215
  - GBMF4744 Accelerator on a chip
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Outline

- State-of-the-art MeV UED
- Spatial and reciprocal space resolution
- Sub-10 fs temporal resolution
- Streaked electron diffraction
- Conclusions

Ultrafast Electron Diffraction: Visualizing Dynamic States of Matter

D. Filippetto

Lawrence Berkeley National Laboratory,
Berkeley, California, 94720,
USA

P. Musumeci

Department of Physics and Astronomy,
University of California at Los Angeles,
Los Angeles, California 90095,
USA

R. K. Li

Department of Engineering Physics,
Tsinghua University, Beijing 100084,
China
Key Laboratory of Particle and Radiation Imaging,
Tsinghua University,
Ministry of Education, Beijing 100084,
China

B. J. Siwick and M. R. Otto

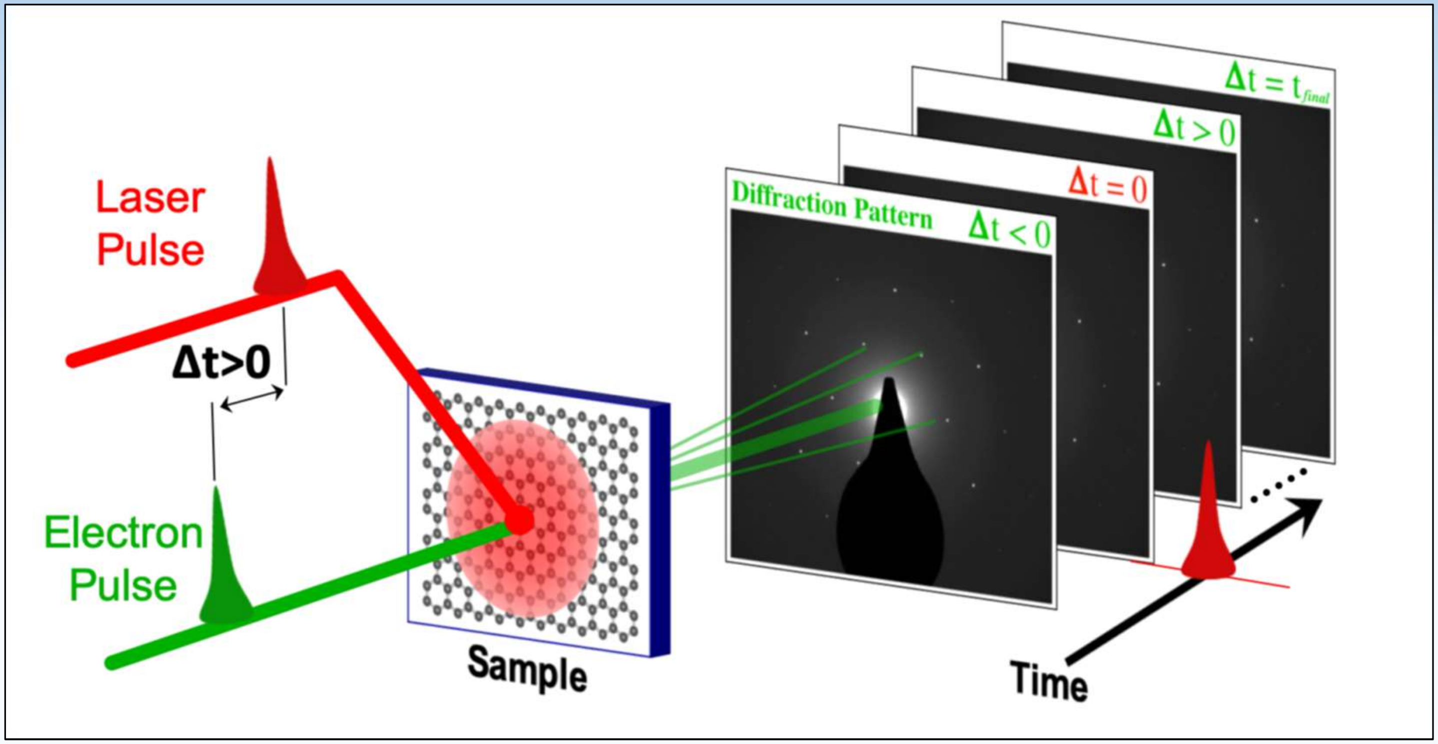
Centre for the Physics of Materials,
Department of Physics and Department of Chemistry,
McGill University, Montreal,
Quebec H3A 2T8,
Canada

M. Centurion and J.P.F. Nunes

University of Nebraska-Lincoln,
855 N 16th Street,
Lincoln, Nebraska 68588,
USA

Review of Modern Physics, 94, 045004 (2022)

Ultrafast Electron Diffraction



Some possible variations on the theme:
Electron beam energy
Excitation pulse type
Repetition rate
Sample phase

High energy ultrafast electron scattering

Unique advantages for MeV electrons

- Single shot diffraction patterns capability
  - Irreversible processes
  - Low repetition rate processes
- Near speed-of-light probe
  - No velocity mismatch (gas, plasma)
- High penetration depth.
  - Kinematic diffraction
  - Thick samples, liquid phases.
- RF compression. Bunch lengths can be shorter than 10 fs !

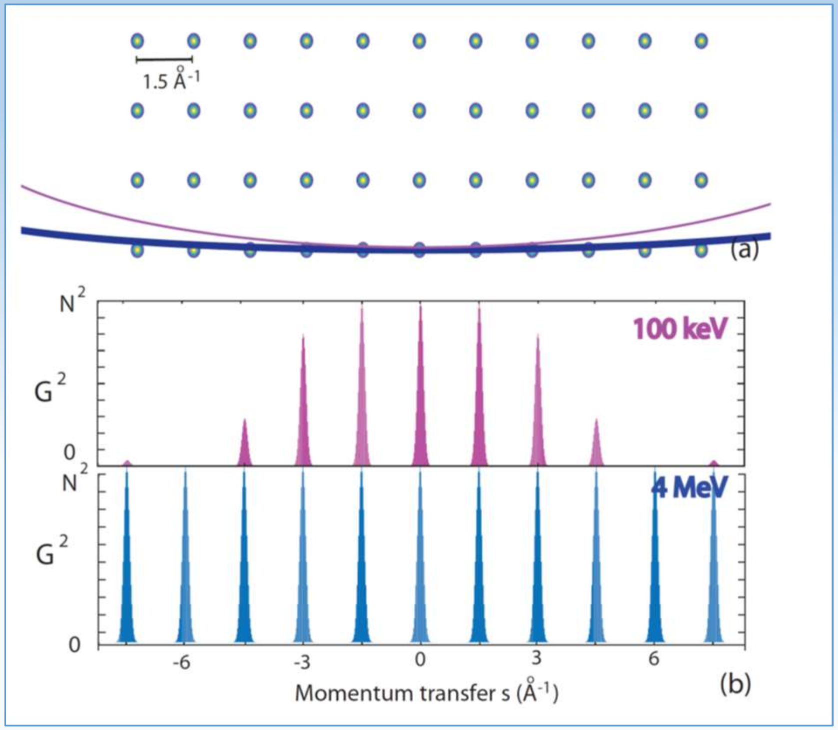
	UED	*MeV ED*
Energy	20-100 KeV	3-5 MeV
Accelerating field	10 MV/m	100 MV/m
# particles	10^4	$10^7 – 10^8$
Pulse length	~200 fs	50 fs
Bragg angle	10 mrad	0.5 mrad
Elastic mean free path (Al)	20 nm	200 nm
Normalized emittance	50 nm	50 nm
Energy spread	<0.01 %	< 0.1 %

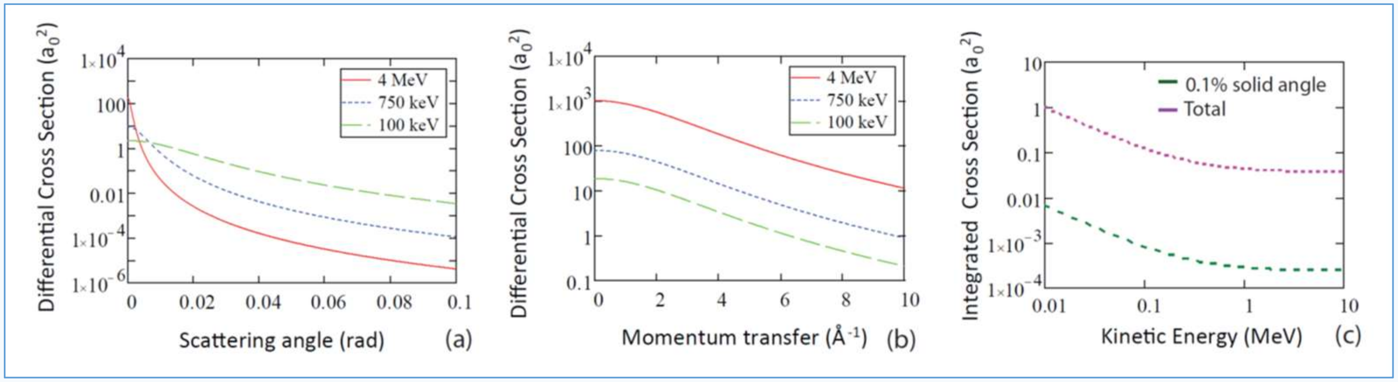
Highlight two important differences

Sensitivity to higher order Bragg reflections $\lambda = h / p$

Difference in cross-section

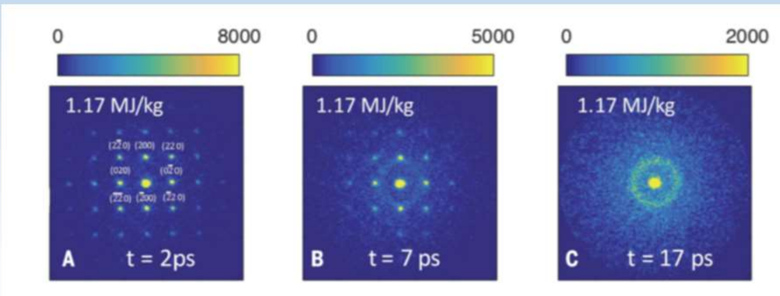
$$\frac{d\sigma}{d\Omega} = \frac{4Z^2}{s^4 a_0^2} \frac{1-\beta^2 \sin^2 \frac{\theta}{2}}{1-\beta^2} \left(1 - F(s)^2\right)^2$$



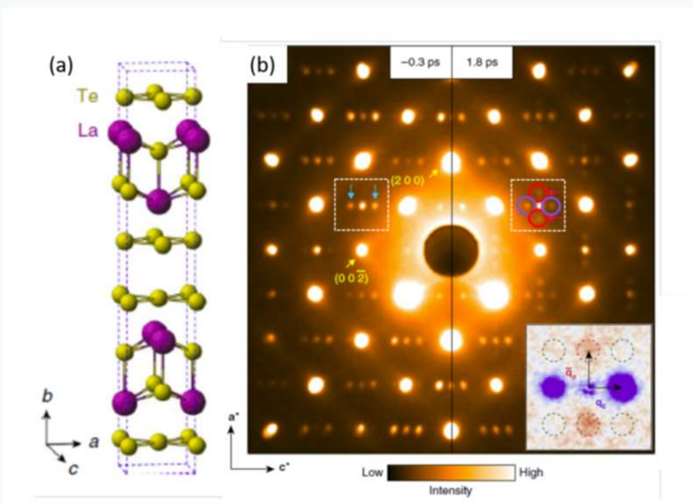


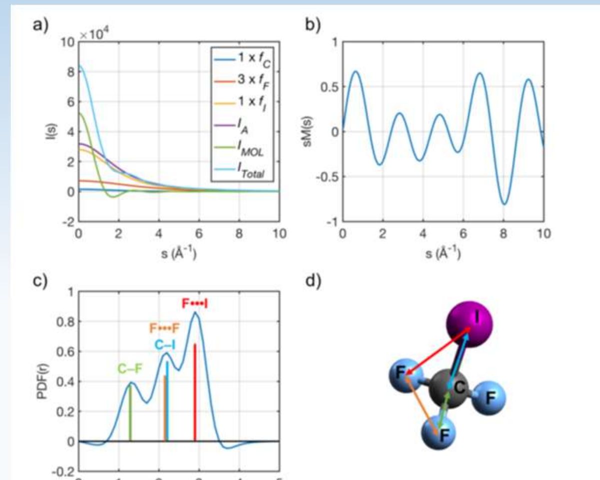
Solid state and gas phase application examples

Ultrafast phase transitions



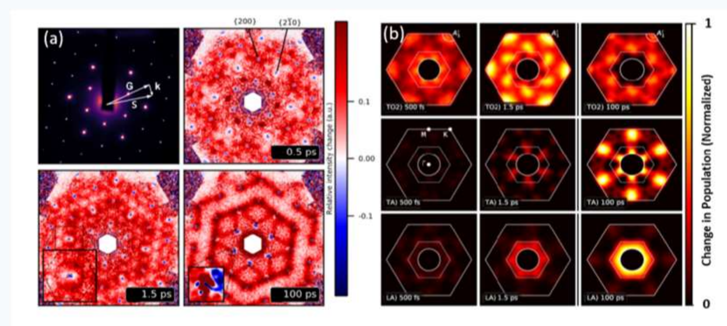
Charge density waves





Molecular dynamics (gas phase)

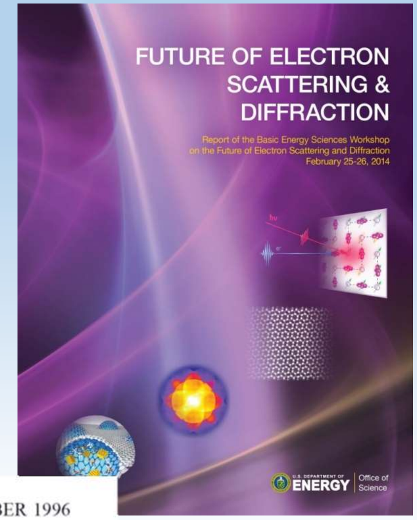
Diffuse scattering: dynamic disorder



RF photoinjector based ultrafast MeV electron diffraction

MeV electron diffraction is a very active and growing field with activities in many national laboratories and universities.

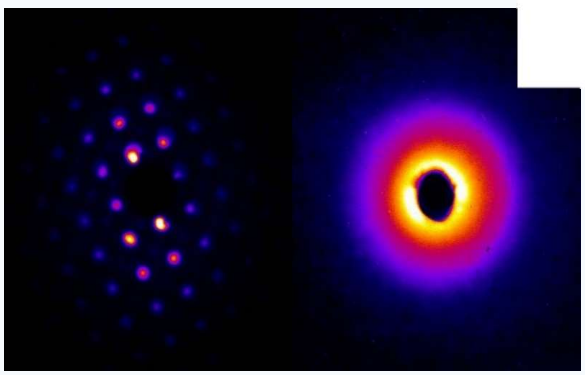
Efforts at UCLA, SLAC, BNL, LBNL
DESY, Shanghai Jiao Tong University, Japan, Korea, UK, Berlin, etc.



PHYSICAL REVIEW E VOLUME 54, NUMBER 4 OCTOBER 1996

Experimental observation of high-brightness microbunching in a photocathode rf electron gun

X. J. Wang, X. Qiu, and I. Ben-Zvi
National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973
(Received 13 February 1996)



Ultrafast electron diffraction patterns of single-crystal gold (left) and nitrogen gas (right) obtained with SLAC's new experimental setup. From Weathersby et al. RSI 2015

J. Hastings et al. Appl. Phys. Lett. 89, 184109 (2006).

P. Musumeci et al., Applied Physics Letters 97, 063502 (2010).

R. Li et al., Rev. Sci. Instrum. 81, 036110 (2010).

Y. Murooka et al., Appl. Phys. Lett. 98, 251903 (2011).

P. Zhu et al., New J. Phys. 17, 063004 (2015).

S. Weathersby et al., Rev. Sci. Instrum. 86, 073702 (2015).

S. Manz et al., Faraday Discuss. 177, 467 (2015)

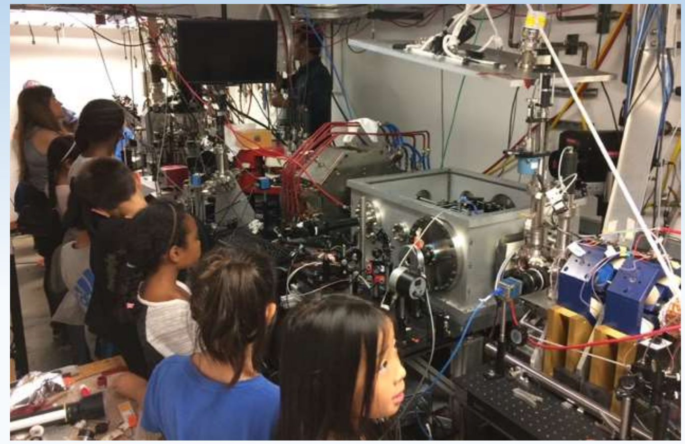
D.Filippetto and H. Qian, J. Atom. and Mol. And Opt. Phys. 49, (2016)

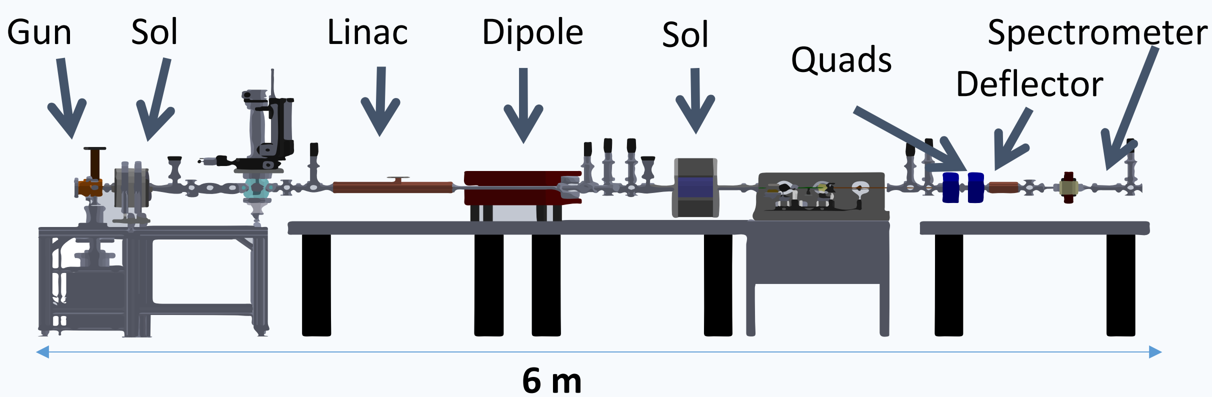
F. Qi et al, Phys. Rev. Lett. 124, 134803 (2020).

H. W. Kim et al., Nature photonics 14, 245 (2020)

High peak brightness beam research at UCLA Pegasus Laboratory

- **3-14 MeV student-run university-size accelerator beamline optimized for sub-pC beams**
- High brightness electron beams:
  - DLA: *few fs acceleration bucket, sub-micron aperture*
  - High resolution ultrafast electron diffraction
  - Single shot imaging / UEM
  - *THz acceleration*
  - *High efficiency THz FEL*





D. Cesar et al., Onset of non-linear effects in high gradient dielectric laser accelerator. **Communications Physics 1, 46 (2018)**

J Maxson, et al., Direct measurement of sub-10 fs relativistic electron beams with ultralow emittance. **Phys. Rev. Lett., 118 154802 (2017)**

D. Cesar, et al., Demonstration of single-shot picosecond time-resolved MeV electron imaging. **Phys. Rev. Lett., 117, 024801 (2016)**

E. Curry et al. Meter-scale THz-driven acceleration of a relativistic electron beam. **Phys. Rev. Lett. 120, 094801 (2018)**

A. Fisher, Y. Park et al. High efficiency single pass THz FEL. **Nature Photonics, 16, 441 (2022)**

What is next for MeV UED? A wish-list for the next 5 years

Instrument characteristic	State-of-the-art	Need to improve	Why
Spatial resolution	5 um	Smaller spot sizes at sample plane	Heterogeneous materials
Absolute and Relative coherence length	10 nm / 100 um < 1e-4	Higher transverse brightness	Larger unit cells, proteins
Temporal resolution	70 fs	Shorter bunch length, jitter	Hard phonons, faster processes
Signal-to-noise ratio	1 A / 10 nA	Peak and average electron current	Weaker signals for reversible/irreversible processes
Stability	100 ppm 0.05^{o} of RF	Low level and high power RF. Laser system	Repeatibility
Pump	IR, NIR, THz, **X-rays**	Synchronized pumps	Different excitation mechanisms

High brightness electron sources

- Better understanding of photocathode physics (condensed matter and material science advances)
- Improved gun technology
  - higher launch field
- Better computation tools
  - Multiobjective optimization
  - 3D electromagnetic solvers
  - Point-to-point space charge algorithms

Relative coherence length

$$l_c = \lambda_c/(2\pi\epsilon_n)$$

https://science.energy.gov/bes/community-resources/reports/

Great potential for breakthrough advances as time resolved electron scattering requires operation in less explored "low charge" regime

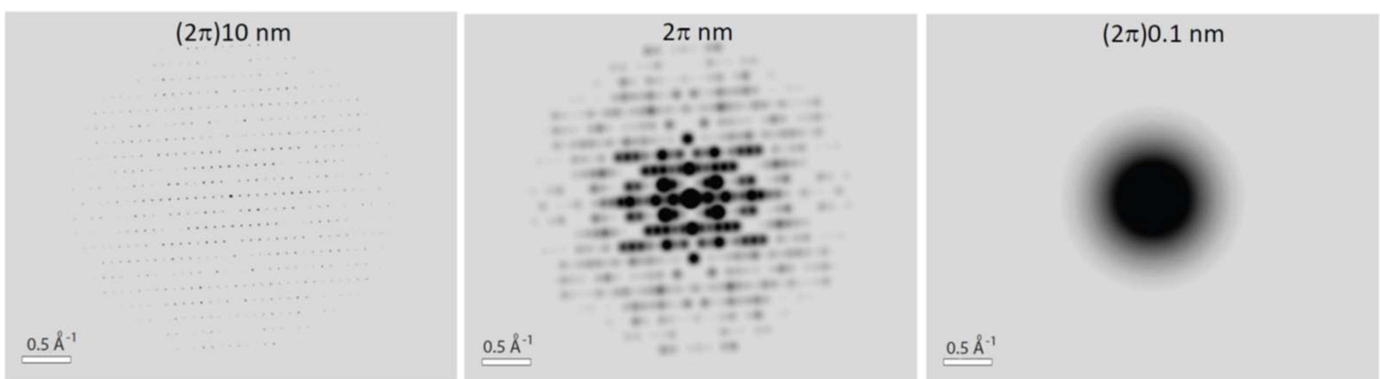
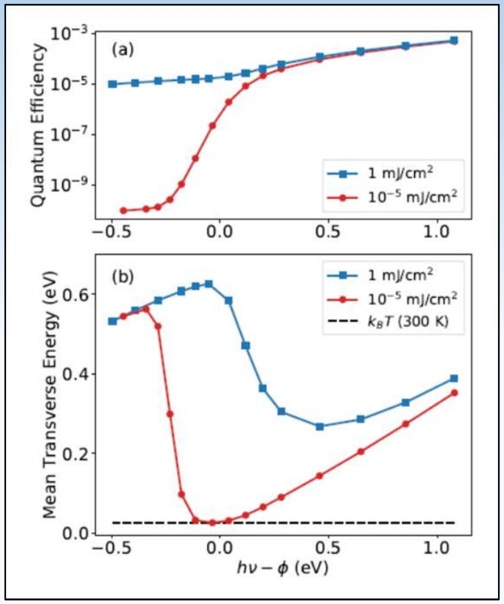


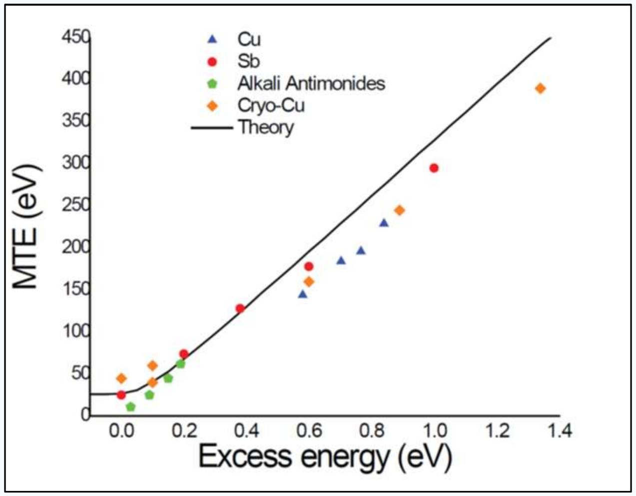
Figure 5 Simulated diffraction patterns of a Salicylic acid (aspirin) crystal for electron probe beams having coherence lengths of (2π) 10 nm, 1 nm and 0.1 nm respectively.

Advanced photocathode research

- Typical operation of Cu cathodes yields MTE of few 100 meV
- 6 meV measured from cooled single crystal Cu
- But…very low QE ~ 10-8. Laser heating when trying to extract more power
- Semiconductor (AA) photocathodes show much promise with potentially more than one order of magnitude increase in coherence length of beam at sample.



J. K. Bae et al. Journal of Applied Physics **124**, 244903 (2018)



Center for BRIGHT BEAMS
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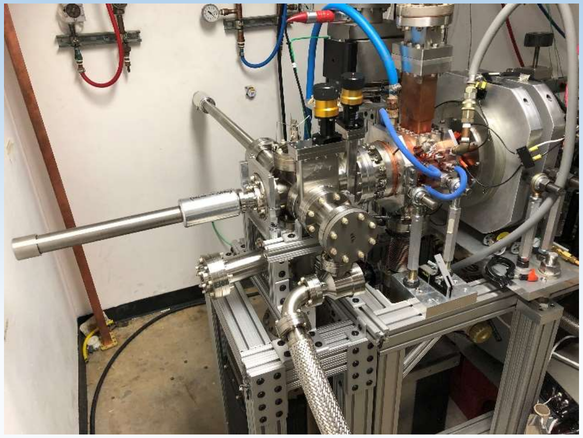
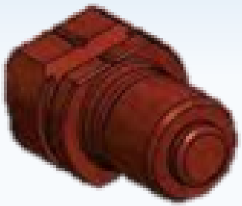
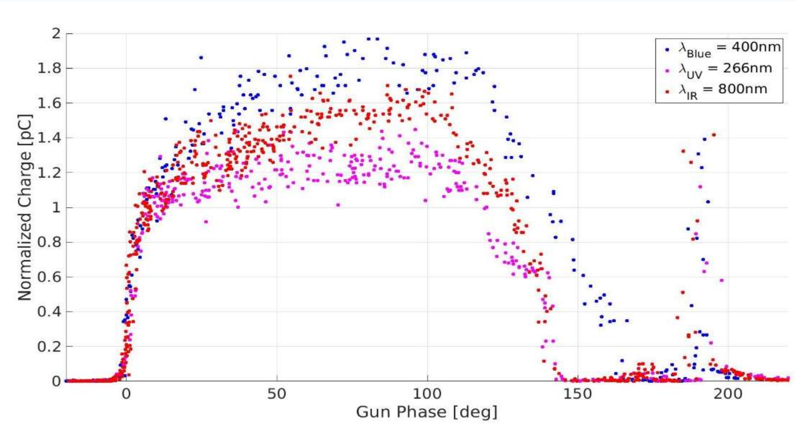
Loadlock system for high gradient photoinjectors

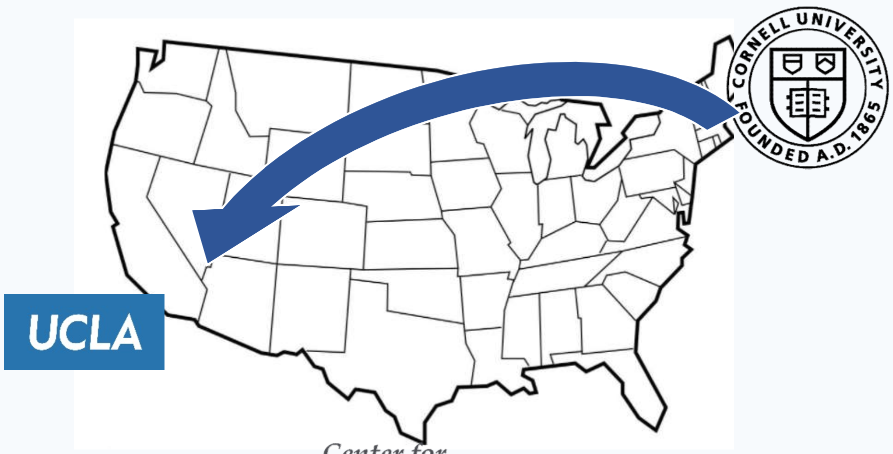
- Advanced photocathodes for S-band RF photoinjectors
- Operating vacuum < 1e-9 (@ 10 Hz) with additional NEG pumping
- Simple cathode transfer setup (copy from LBNL) + INFN-style plug
- Results :
  - 0.5 % level QE (in UV) !
  - Major issues in air-transport
  - Multi-frequency emission studies ongoing
  - Low MTE requires tunable illumination in visible





INFN/DESY/LBNL cathode plug





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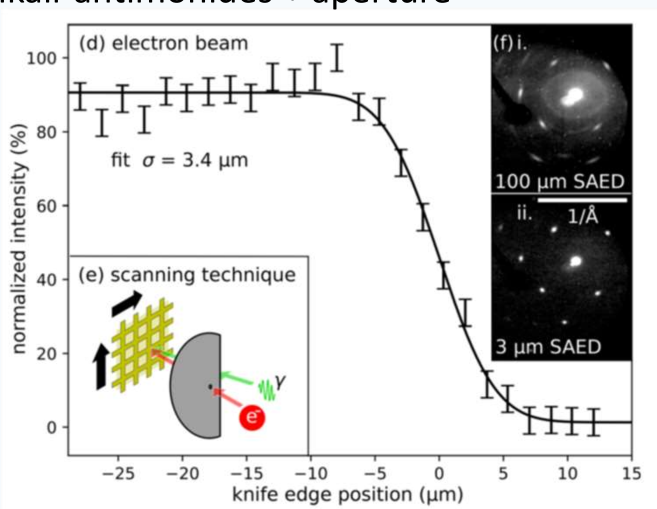
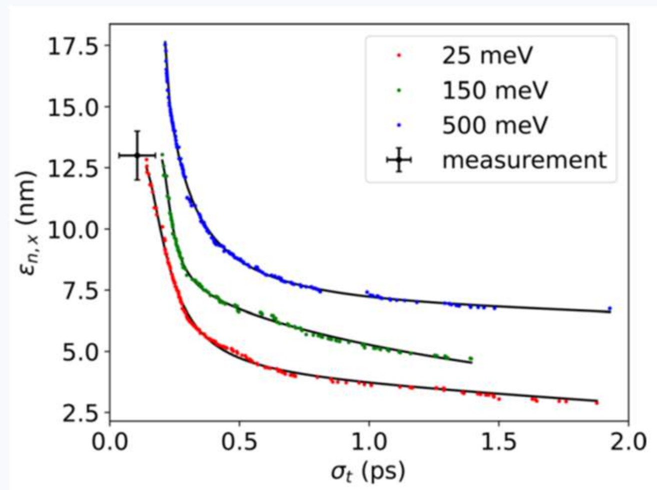
Advanced cathodes can also help non relativistic beamlines : MEDUSA @ Cornell

W. H. Li et al. A kiloelectron-volt ultrafast electron micro-diffraction apparatus using low emittance semiconductor photocathodes featured. Structural Dynamics 9, 024302 (2022)

Alkali-antimonides + aperture

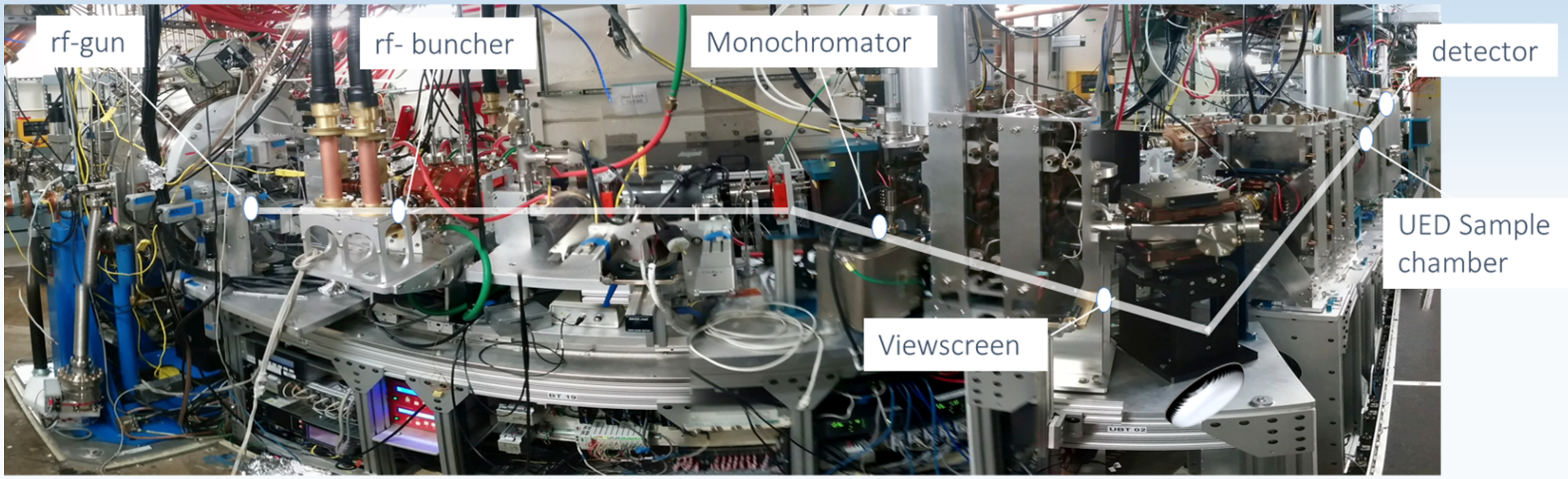


Simulation Pareto front

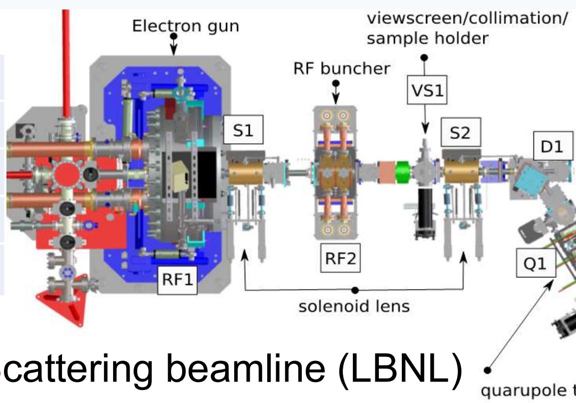


Sub-nm normalized emittance measurements @ APEX

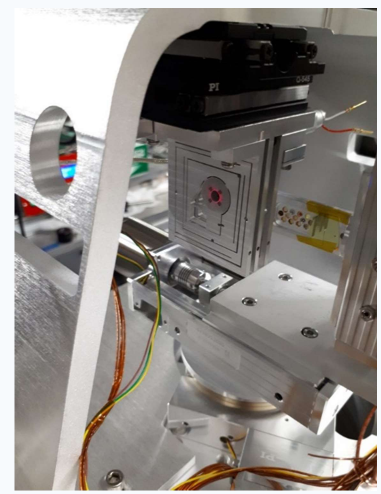
Very small spot sizes and low emittances possible by sacrificing number of electrons per pulse
Compensated by high repetition rate to keep number of electrons per second
PM-based quadrupoles for tight focusing at sample plane



Electron beam energy	0-900 keV
Repetition rate	1-10^6 Hz
Temporal resolution	100 fs-1000 ps
Electrons per pulse	1-10^8
Relative energy spread	10^{-3} 10^{-5}

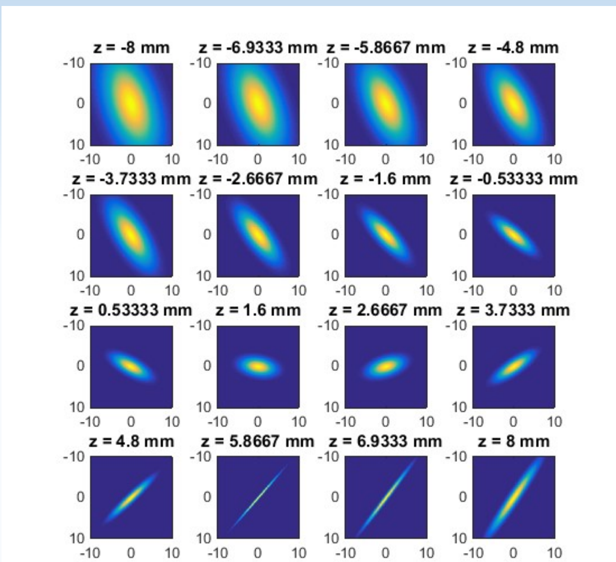


D.Filippetto and H. Qian, JPB: Atom. and Mol. And Opt. Phys. Vol 49, 2016



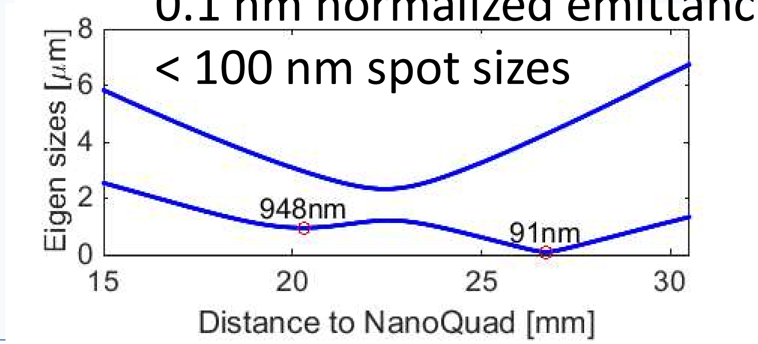
High Repetition Rate Electron Scattering beamline (LBNL)

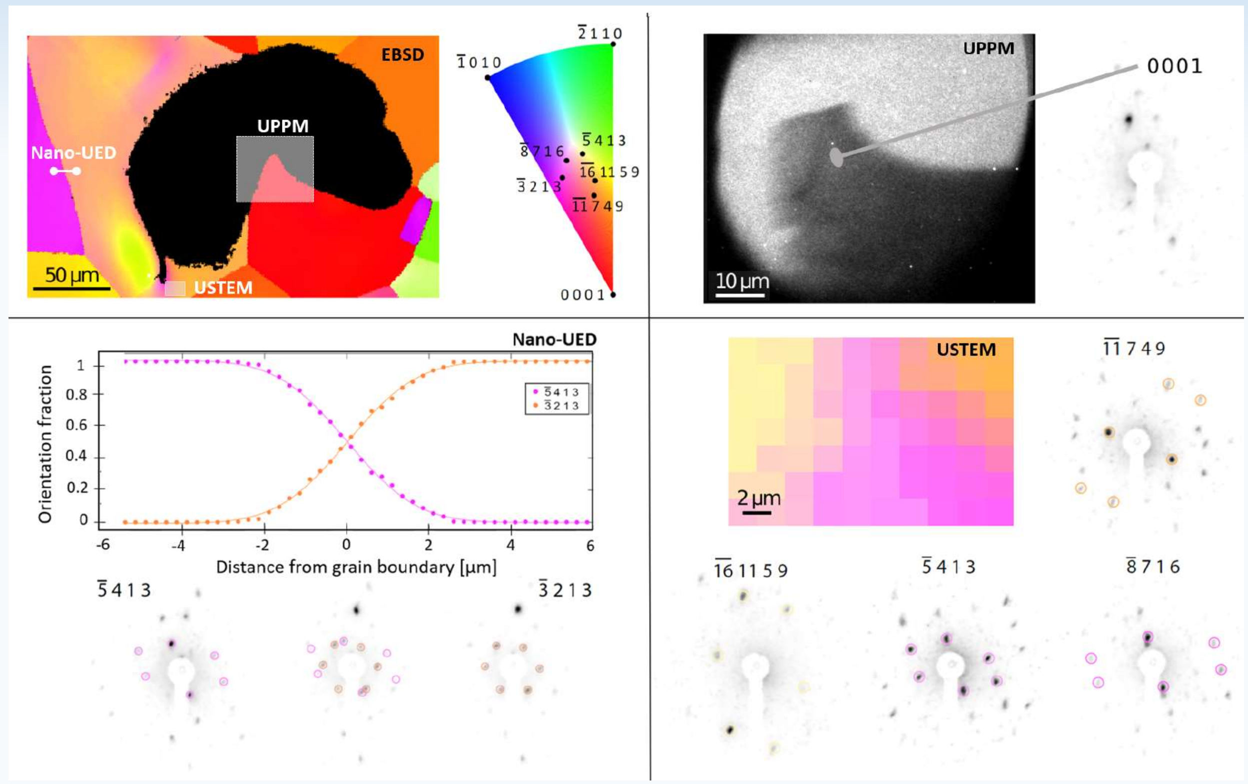
Ultrafast relativistic probes for ultrafast point projection microscopy, nano-UED, and ultrafast STEM



0.1 nm normalized emittances

< 100 nm spot sizes

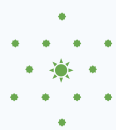


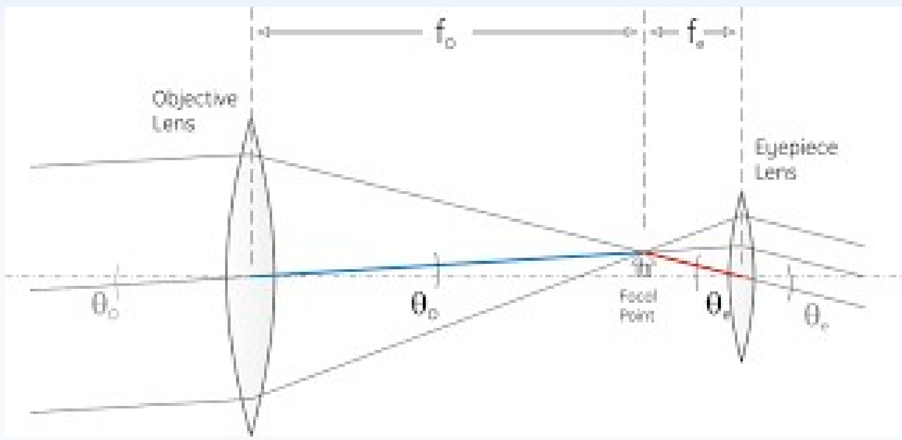


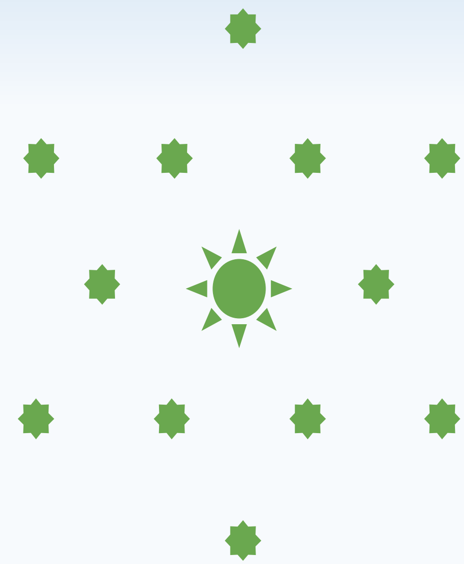
F. Ji, D. Filippetto et al. Communications Physics 2.1-10 (2019)

Improving reciprocal space resolution with variable camera length electron optics

"zoom in" on diffraction features adding magnetic lenses after the sample
telescope configuration for angular magnification ($m = f_0/f_e$)
Improve reciprocal space resolution







Results in angular magnification

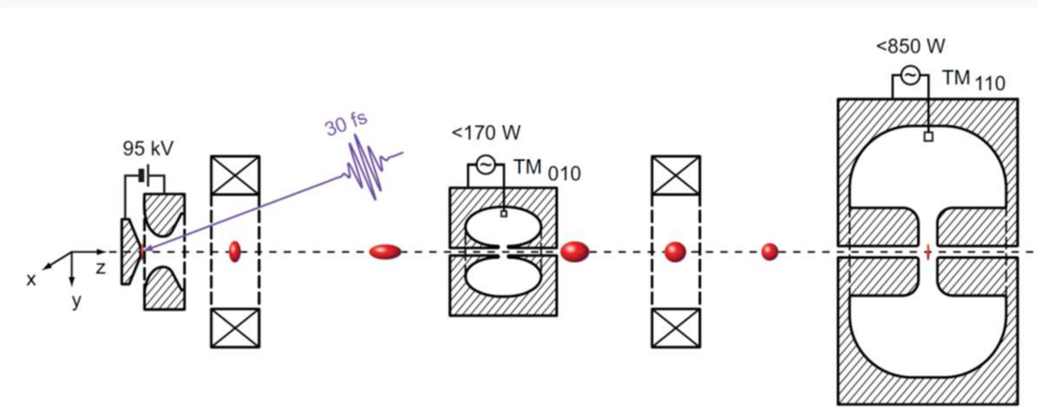
Numerical example
L = 4 m drift *vs.* f_0 = *1 m* ; f_e = 4 cm
R12 goes from 4 m to - 100 m !!!

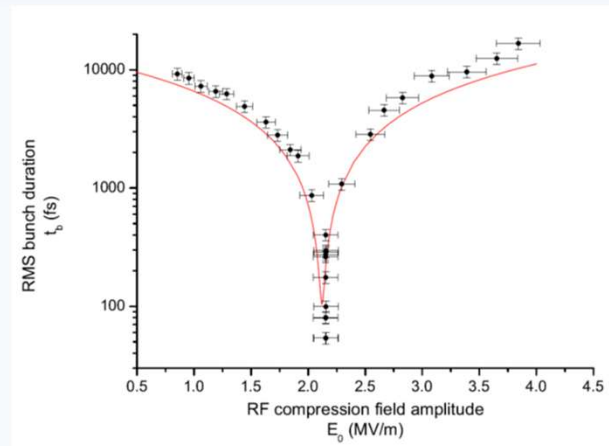
Preliminary results at Cornell
Under evaluation at SLAC, UCLA

V. Guo and P. Denham (UCLA)

RF compression

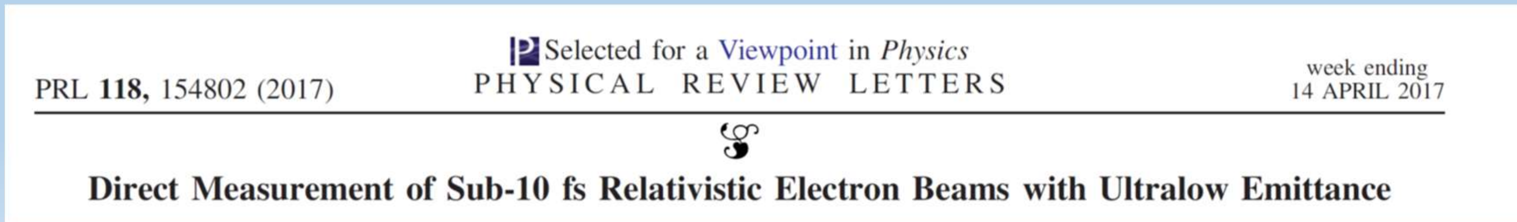
- Standard technique in accelerator and beam physics
- Introduced in UED to compensate longitudinal expansion in keV sources
- Clearly can also be applied to MeV beams





T. van Oudheusden, E. F. de Jong, S.B. van der Geer, W.P.E.M. Op 't Root, B. J. Siwick, O. J. Luiten, J. Appl. Phys. ***102****, 093501 (2007).*

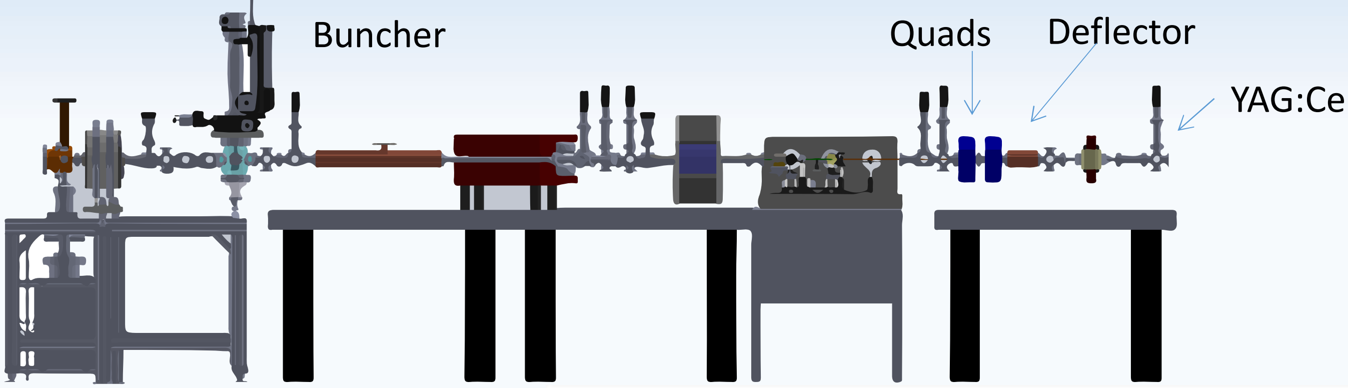
T. van Oudheusden, P. L. E. M. Pasmans, S. B. van der Geer, M. J. de Loos, M. J. van der Wiel, and O. J. Luiten Phys. Rev. Lett. ***105****, 264801 (2010)*

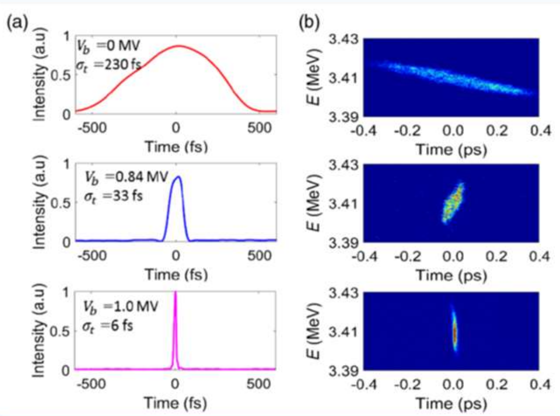

PRL 118, 154802 (2017)
Selected for a Viewpoint in Physics
PHYSICAL REVIEW LETTERS
week ending 14 APRIL 2017

Direct Measurement of Sub-10 fs Relativistic Electron Beams with Ultralow Emittance

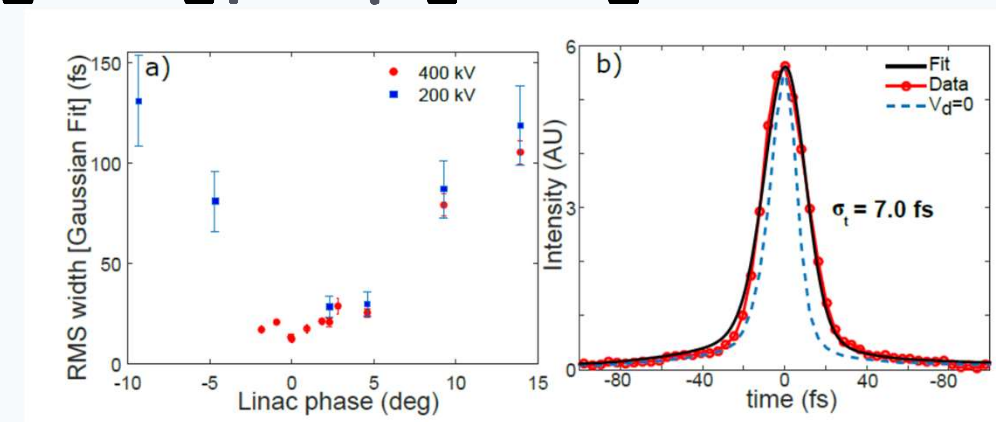
Use short laser pulse (~100 fs) on the cathode to minimize long emittance

- ~5 MeV, 20 fC full beam (no slit), quad focusing on final YAG:Ce screen



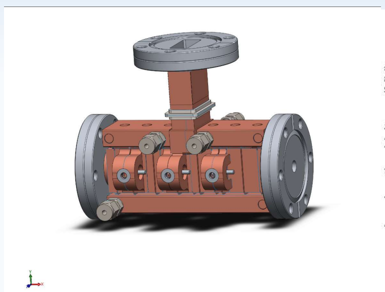


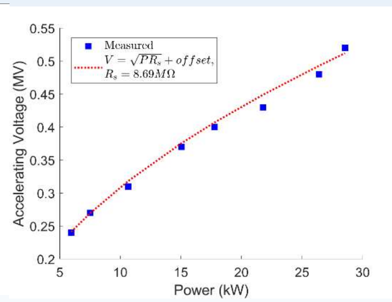
L. Zhao. Phys. Rev. X 8, 021061 (2018)

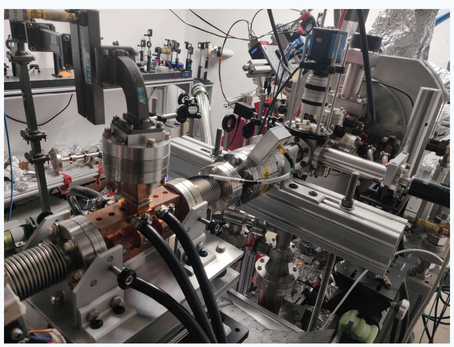


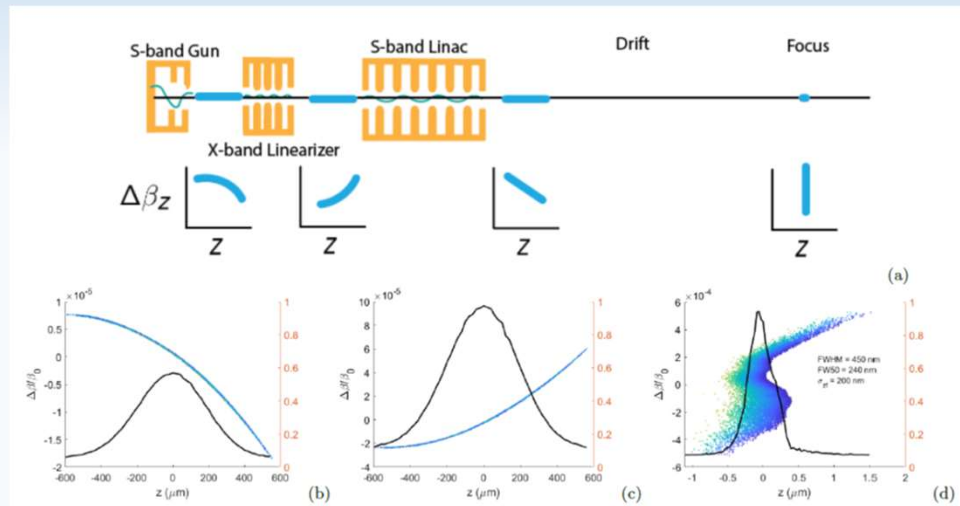
Two frequency RF compression

- Even better performances can be obtained using two cavities at two different frequencies
- Linearize phase space and then compress
- X-band high harmonic cavity can be used to compensate non linearities









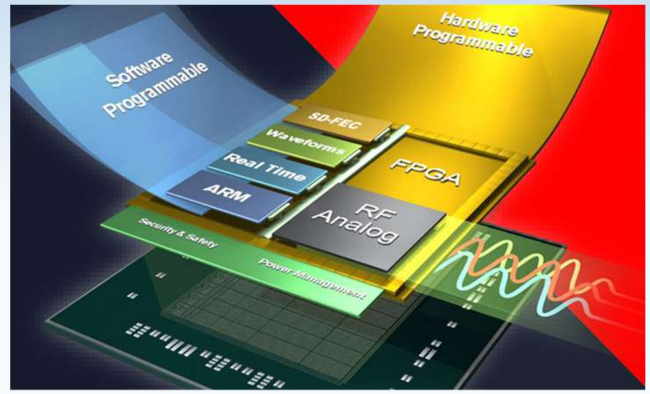
- Measuring sub-fs bunch length is an open challenge
- Phase dependence of laser-driven acceleration signal could be used.

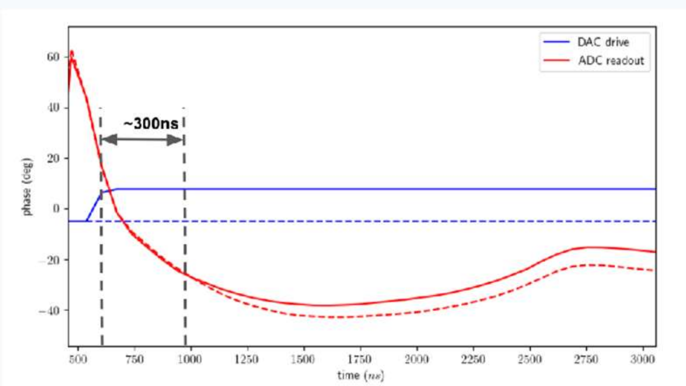
K.Floettmann. Generation of sub-fs electron beams at few-MeV energies. NIM A 740, 34 (2014)

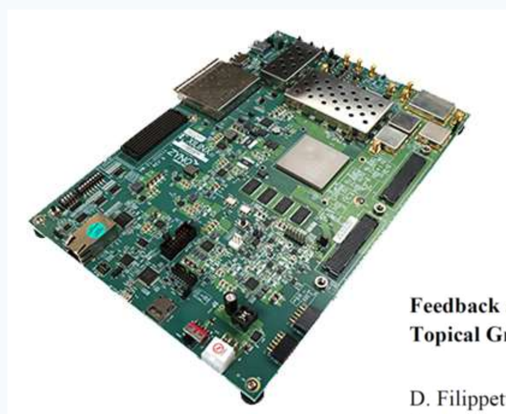
Outstanding challenge for UED : synchronization and timing jitter with pump laser pulse !!!

Improving time-jitter in UED systems

- FPGA-based electronics allow ~100 ns latency feedback loops
  - Up to: 7 GHz BW and 10 GSPS (direct sampling)
  - Not optimized for RF accuracy
  - Developments start now for deployment in accelerators (e.g. UCLA intrapulse RF feedback, ALS-U BPM electronics)
  - These areas would need development for use in LLRF applications







Feedback and control systems for future linear colliders: White Paper for Snowmass 2021 Topical Group AF07-RF

D. Filippetto[1], C. Serrano[1], Q. Du[1], L. Doolittle[1], D. Wang[1], M. Bachtis[2], P. Musumeci[2], A. Scheinker[3], J. Power[4], M. Bellaveglia[5], A. Gallo[5], L. Piersanti[5]

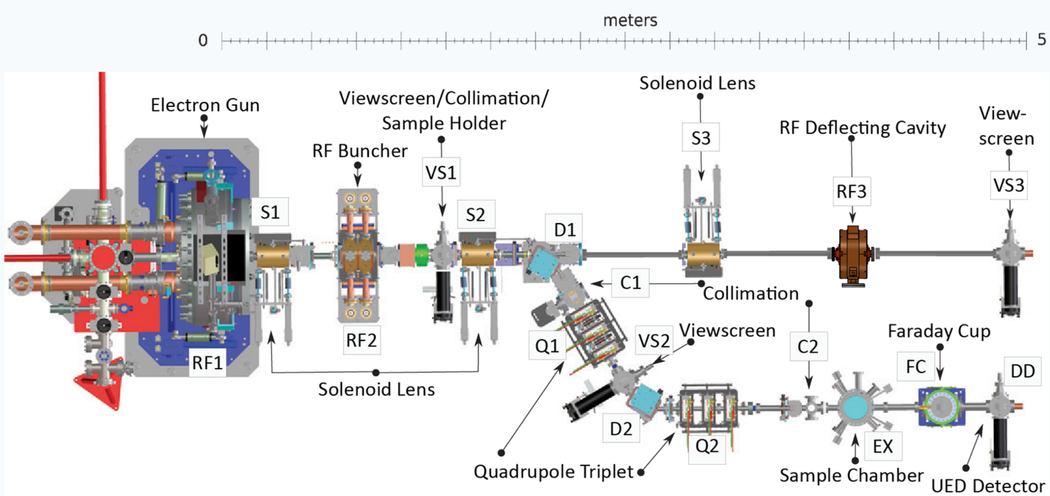
Software-based time-stamping

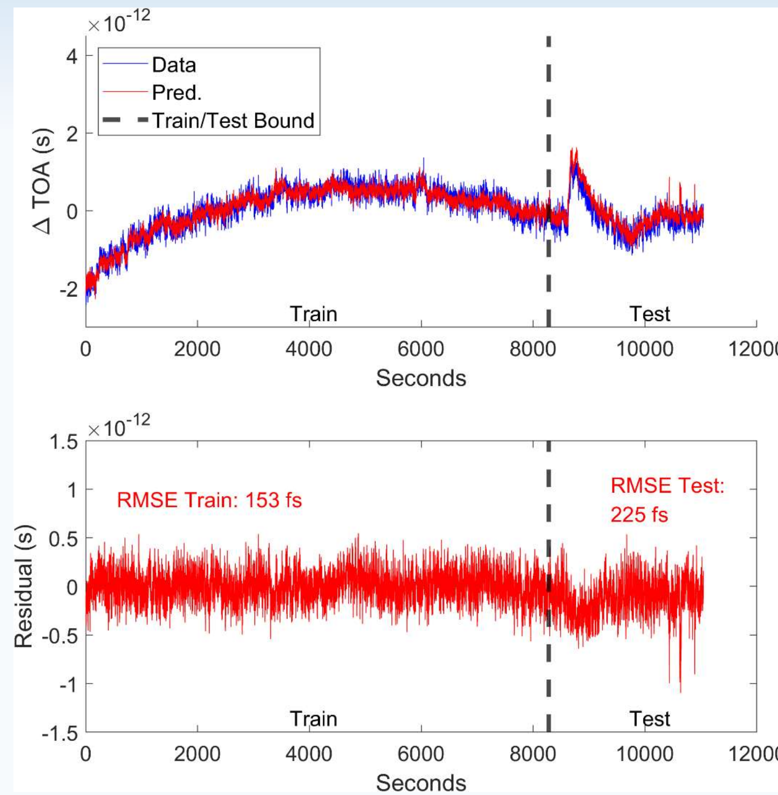

PHYSICAL REVIEW ACCELERATORS AND BEAMS **26,** 052801 (2023)

Virtual-diagnostic-based time stamping for ultrafast electron diffraction

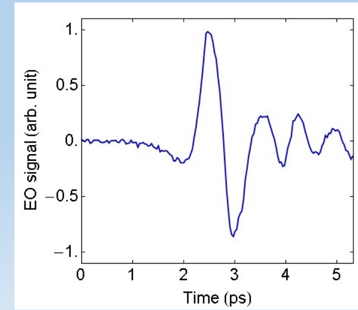
F. Cropp,[1,2,*] L. Moos,[3] A. Scheinker,[4] A. Gilardi,[2] D. Wang,[2] S. Paiagua,[2] C. Serrano,[2] P. Musumeci,[1] and D. Filippetto[2,†]

- Measure all accelerator parameters
- Train neural network to predict time of arrival using data from RF deflecting cavity
- Tag each shot with reconstructed time-of-arrival information
- Demonstrated at LBNL to perform at 150 fs level, better than conventional feedback systems on similar time-scales.





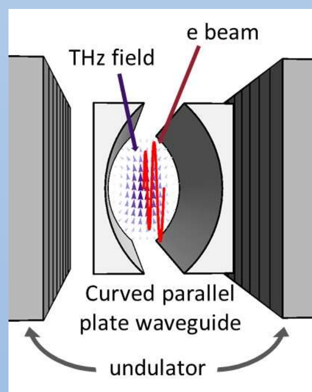
THz-based compression



Velocity bunching driven by laser generated THz field re-sychronizes beam to laser clock

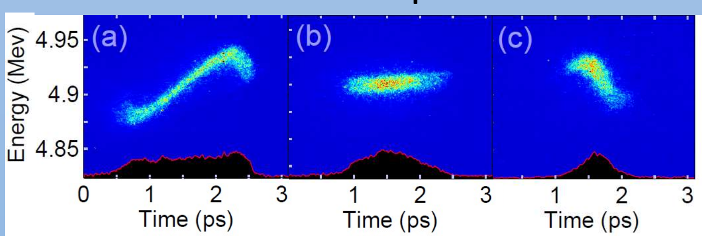
Ponderomotive interaction

- Magnetic undulator
- Coupling to transverse THz field
- Group and phase velocity matching



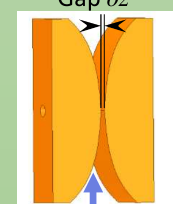
E. Curry, et al. "Meter-scale terahertz-driven acceleration of a relativistic beam." *PRL* 120.9 (2018): 094801.

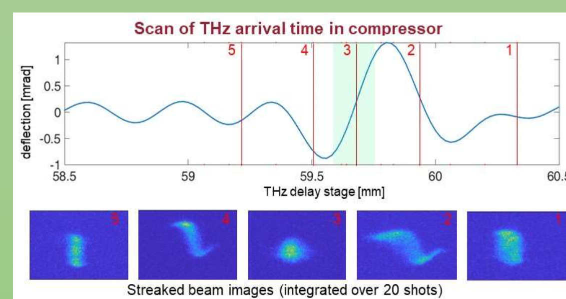
THz- driven compression



TM-mode interaction

- Near-field structure
- Coupling directly to longitudinal particle motion
- Field enhancement
- Short interaction length





Snively, E. C., et al. "Femtosecond compression dynamics and timing jitter suppression in a THz-driven electron bunch compressor." *Physical review letters* 124.5 (2020): 054801.

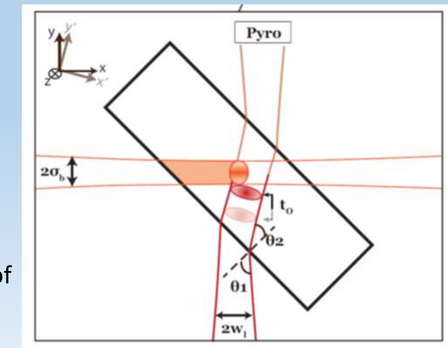
Zhao, Lingrong, et al. "Femtosecond relativistic electron beam with reduced timing jitter from THz driven beam compression." *Physical review letters* 124.5 (2020): 054802.

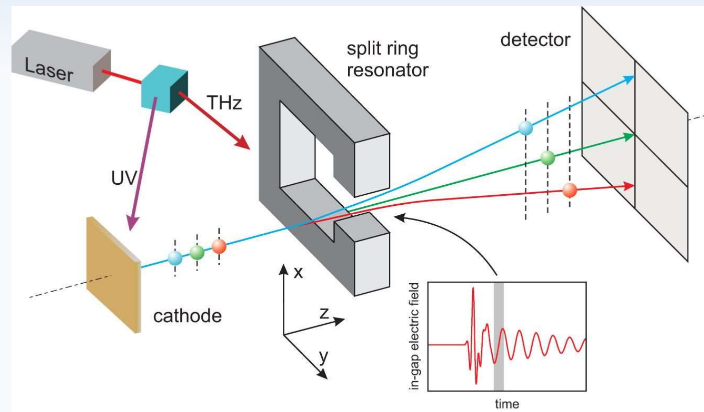
See also Catherine Kealhofer, Waldemar Schneider, Dominik Ehberger, Andrey Ryabov, Ferenc Krausz, Peter Baum. *Science* 352, 429 (2016).

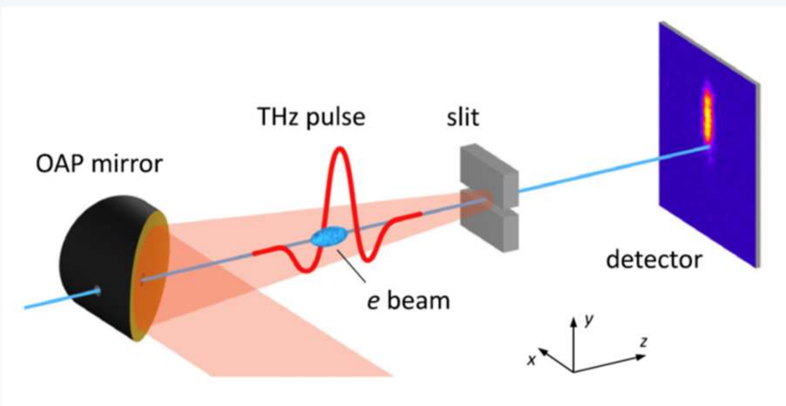
Hardware-based time-stamping

- If temporal jitter can't be avoided, time-stamping is the solution
- EOS or e-beam induced index of refraction changes
- Laser-generated THz can be used to measured time-of-arrival (and bunch length at the same time)

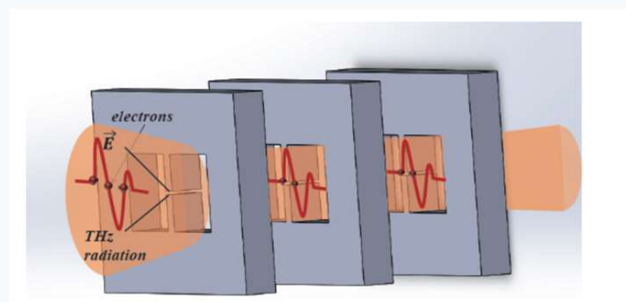
D. Cesar and P. Musumeci. "Ultrafast gating of mid-infrared laser pulses by sub-pC relativistic e-beams", Journal of Applied Physics, 118, 234506 (2015)







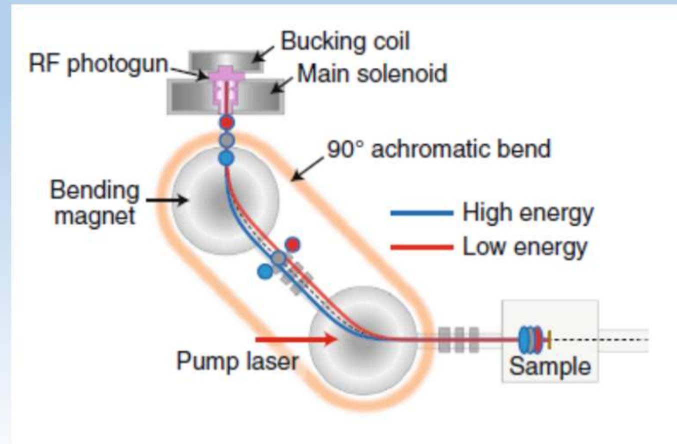
J. Fabiańska et al., Scientific Reports, 4 5645 (2014)
C. Kealhofer et al., Science 352, 429 (2016).
L. Zhao. Phys. Rev. X 8, 021061 (2018)
R. K. Li, Phys. Rev. Accel. Beams **22**, 012803 (2019)
X. Shen et al., AIP Advances **9**, 085209 (2019)



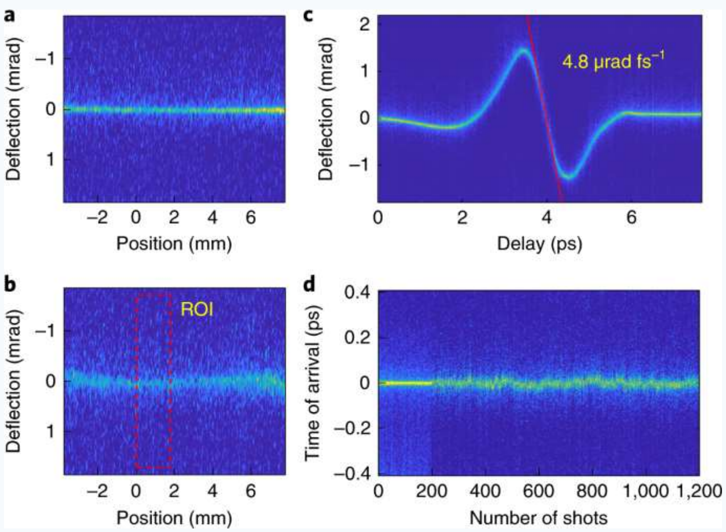
25 urad/fs
10 MV/m THz field

Magnetic compression

- Use double-bend achromat to compress
- Fully passive system. No RF so no additional jitter
- 50 fs temporal resolution demonstrated

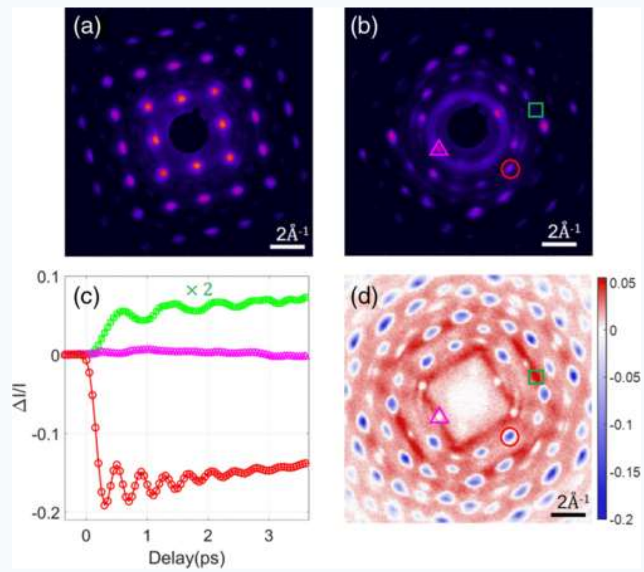


THz streaking



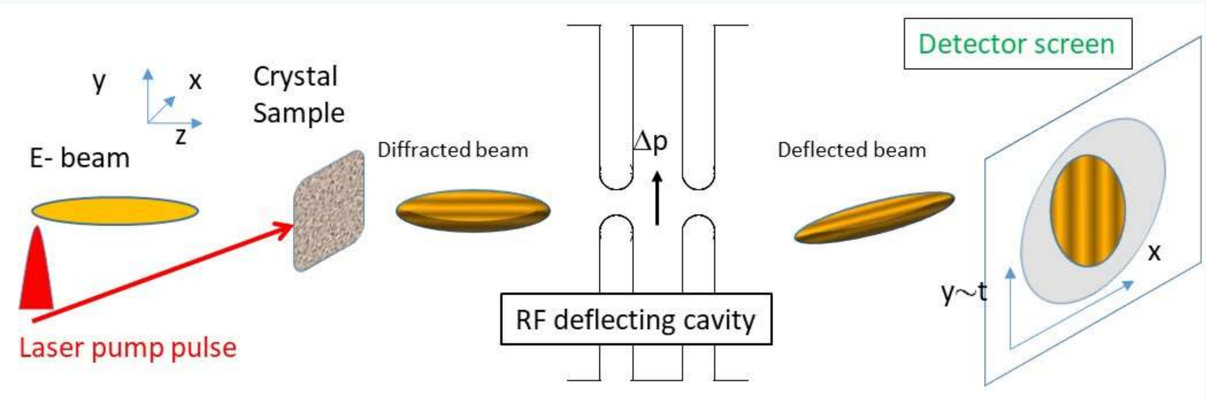
H. W. Kim et al. Towards jitter-free UED technology, Nature Photonics, 14, 245 (2020)

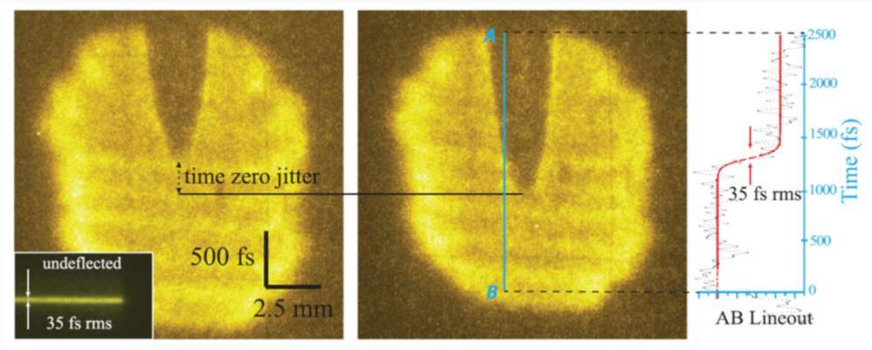
F. Qi et al., Phys. Rev. Lett. 124, 134803 (2020).



Streaked MeV UED

- Capture entire history of ultrafast process in one shot
- RF streak camera based electron diffraction (from Mourou-Williamson original paper on UED)
- Use RF deflecting cavity as a streak camera to time-resolve a relatively long (10s of ps) electron beam after its interaction with the diffraction sample.
- Three significant advantages
  - Free UED by the limitation due to the length of the electron beam.
  - Improve significantly the temporal resolution of the technique.
  - Yield true single-shot structural change studies revolutionizing the approach of the conventional pump-probe experimental procedure.

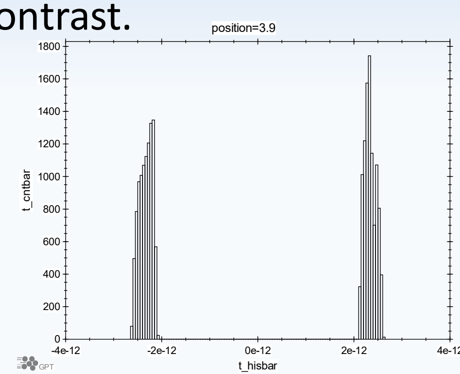




C. M. Scoby et al., APL, 102, 023506 (2013)
P. Musumeci et al. JAP, 108, 114513 (2010)

Double-shot electron diffraction / microscopy

- Use two laser pulse to illuminate the cathode and deflecting cavity to separate images on the screen (similar to ‘movie mode DTEM’).
- Adjustable separation between 1 and 16 ps
- GPT start-to-end simulations use a 8 μm Au disk 25 μm thick. Mass contrast.
- Resolution ~ 200 nm Q = 100 fC per pulse



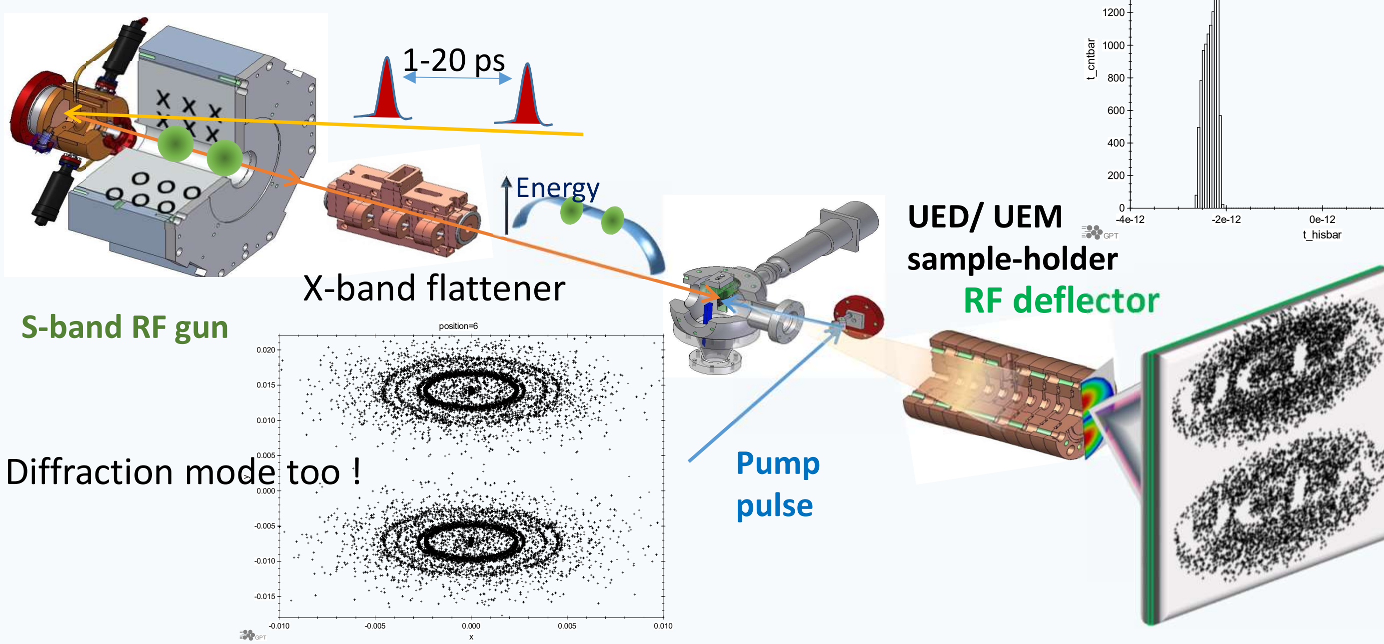
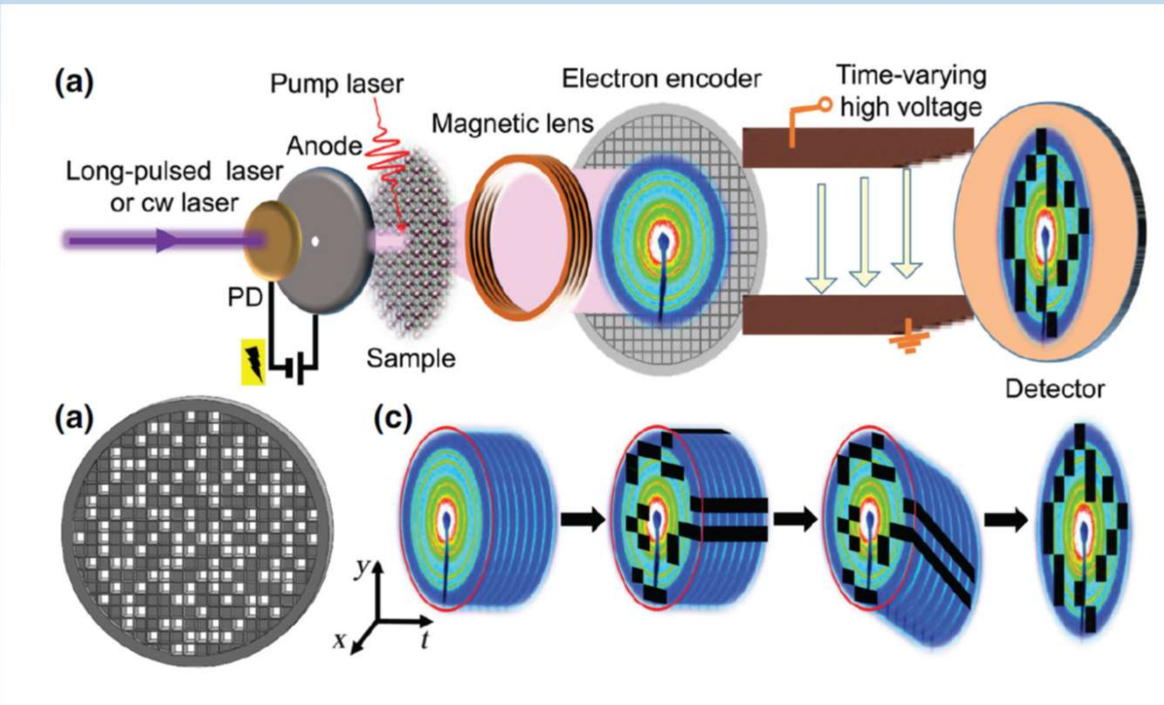


Image reconstruction algorithms and masks can greatly help

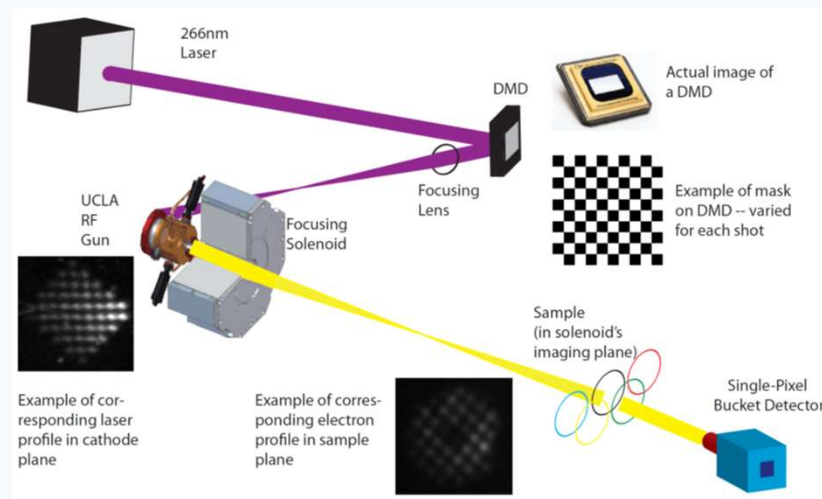


Compressed Ultrafast Electron Diffraction Imaging Through Electronic Encoding
D. Qi et al. **Phys. Rev. Applied 10, 054061 (2018)**

S. Li, F. Cropp, K. Kabra, T. J. Lane, G. Wetzstein, P. Musumeci, and D. Ratner. Electron ghost imaging. **Phys. Rev. Lett. 121, 114801 (2018)** and highlighted in APS Physics**.**

K. Kabra, S. Li, F. Cropp, T. J. Lane, P. Musumeci, D. Ratner. Mapping Photocathode Quantum Efficiency with Ghost Imaging. **Phys. Rev. Accel. Beams 23 022803 (2020).**

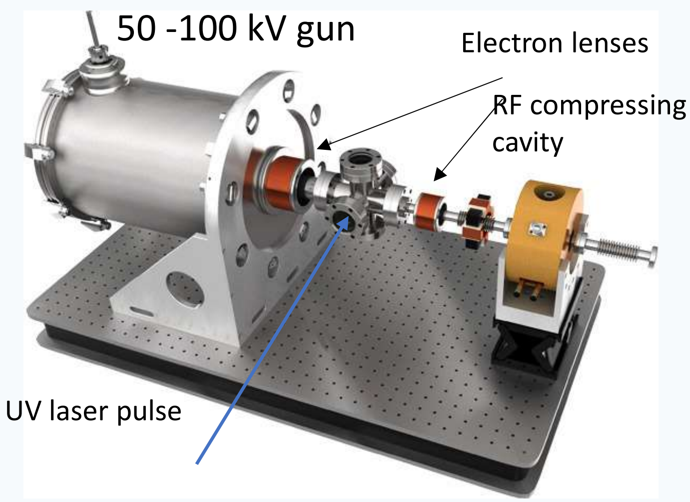
UCLA SLAC NATIONAL ACCELERATOR LABORATORY

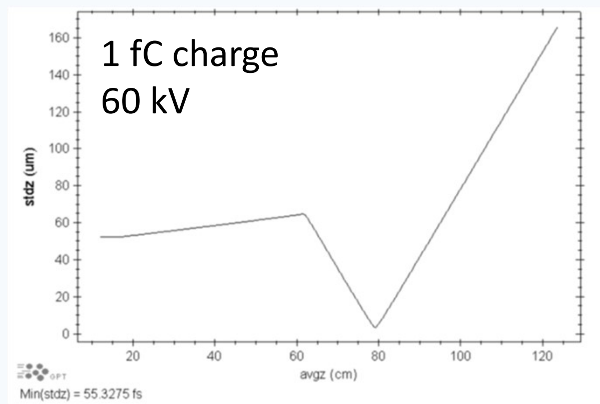


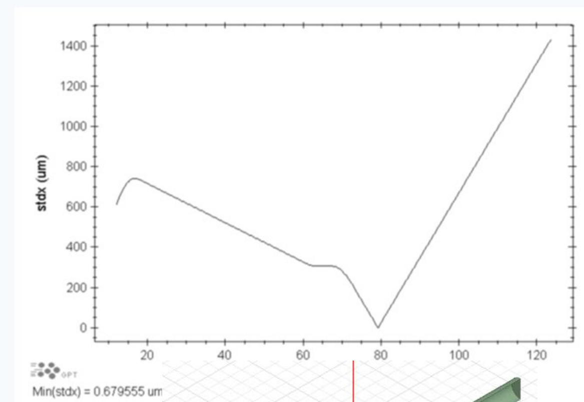
NSF MRI Hy-Res Hybrid modality imaging instrument

- Many experiments can be performed with non-relativistic setups
- Combine X-rays and electrons in one setup:
  - Ultrafast HHG and ED sharing laser and samples. STROBE MRI
  - Preliminary work at FERMI with TUE group
- Use advanced photocathodes and RF compression to obtain ultrahigh brightness ultrashort probes at very high repetition rates (stroboscopic mode)

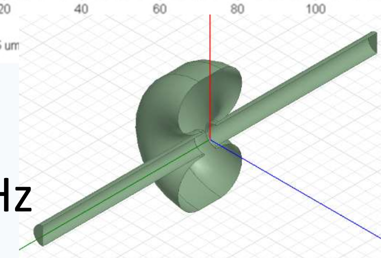
STROBE
NSF SCIENCE AND TECHNOLOGY CENTER



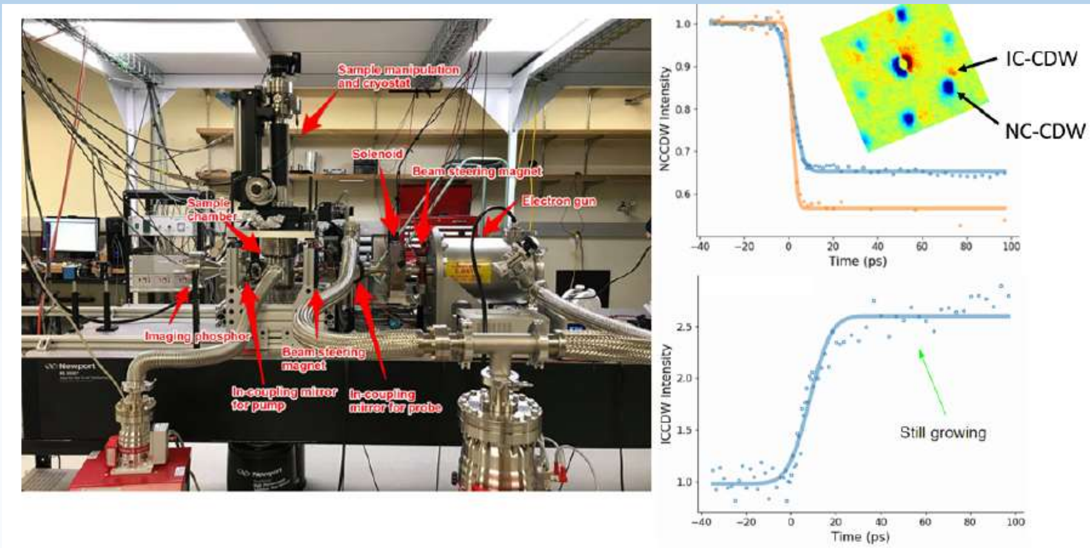




CW RF
Repetition rate set by laser up to 50 KHz

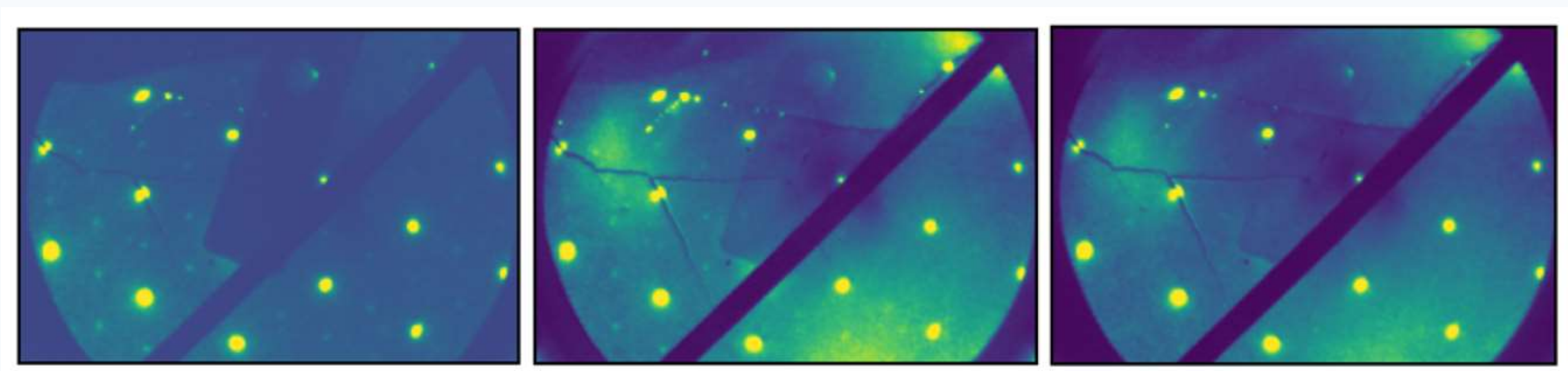


UCLA HyRes non relativistic UED beamline



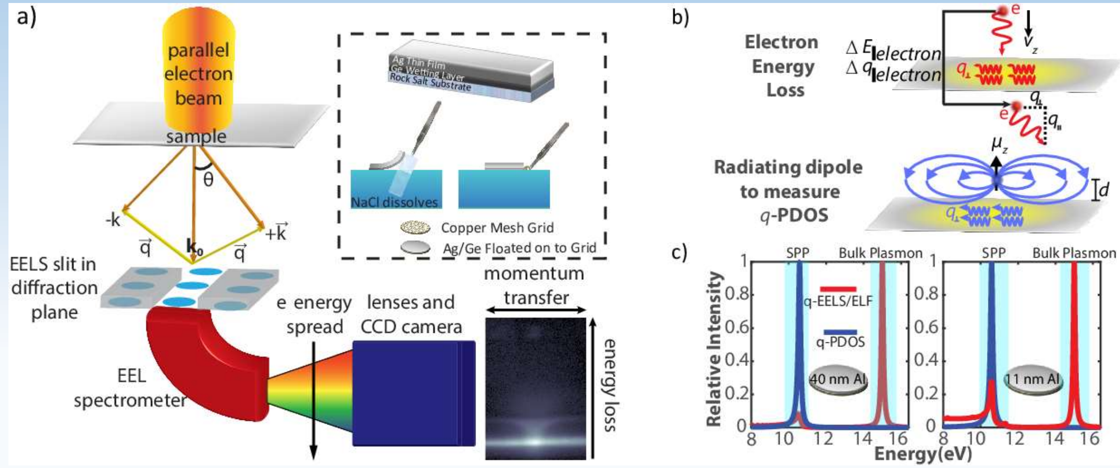
Collaboration with A. Kogar

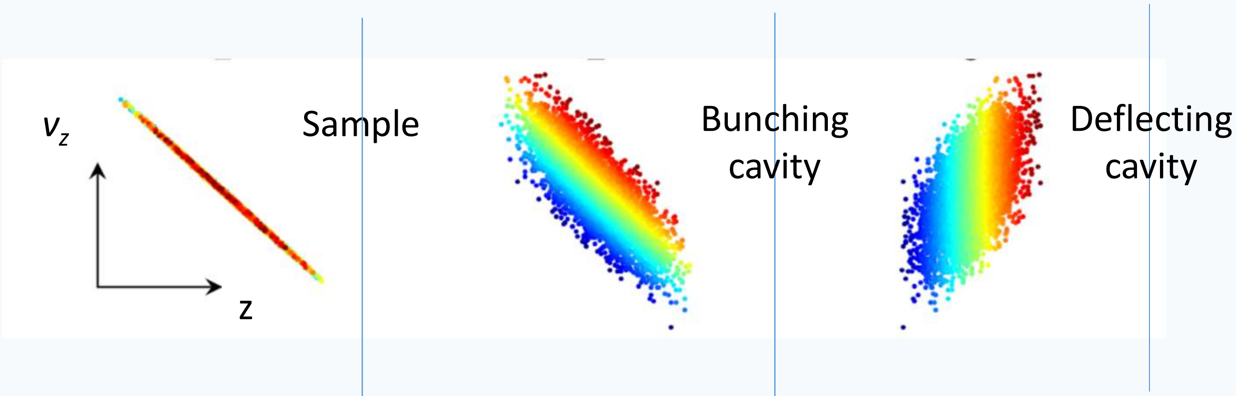
Two lessons learned
Steady state heating -> sample holder
Coherent phonon excitation requires ultrashort pump laser

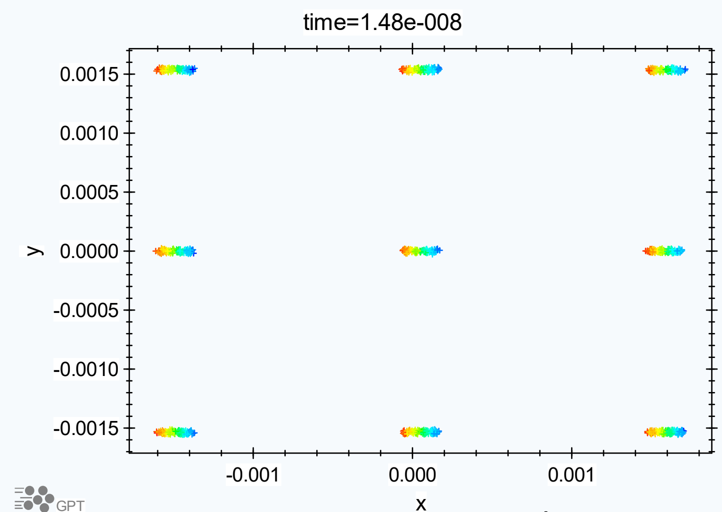


Momentum-resolved electron energy loss spectroscopy

- Add spectrometer to get energy resolution
  - Take advantage of low longitudinal emittance of photoemitted beam
  - Stretch beam at sample to decrease uncorrelated energy spread
    - Spectrometer magnet
    - Time of flight
  - Use image retrieval algorithms to get kx, ky, E





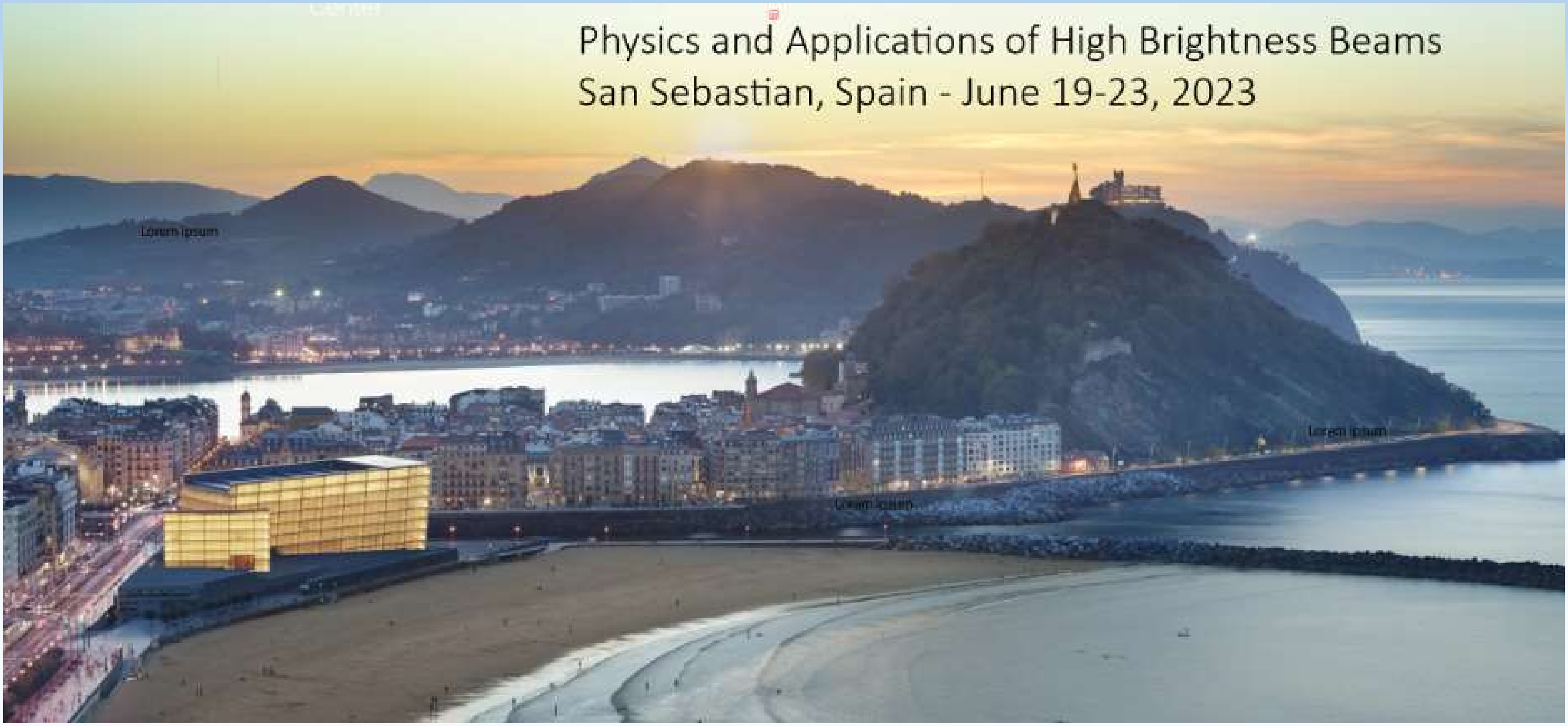


Delta E = 20 eV

Verhoeven, W., et al. "Time-of-flight electron energy loss spectroscopy using TM110 deflection cavities." *Structural Dynamics* 3.5 (2016): 054303.

Conclusions and outlook

- Ultrafast electron sources (MeV AND keV) are pushing the boundary of spatial and temporal resolution in diffraction (but also microscopy/spectroscopy)
- Advanced photocathode and improving beam brightness
  - Longer coherence lengths, nanodiffraction, diffuse scattering
- Sub-10 fs temporal resolution
  - Sub-10 fs electron pulses, THz time stamping, THz compression
- Streaked electron diffraction
  - Highest temporal resolution, no jitter problems, truly single shot, image reconstruction algorithms
- Availability of different pumps
  - THz or X-rays



https://indico.classe.cornell.edu/event/2170/