

Perspectives and outlook for MeV ultrafast electron diffraction

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Disclaimer: This is by all means a personal perspective, shaped by my own involvement in this field. The main focus of the talk is to recount the history of the UED technology (not the UED science), how it started, how it evolved, where we are now and what could be the next steps

UCLA

Outline

- Development of MeV UED
- Spatial and reciprocal space resolution
- Sub-10 fs temporal resolution
- Advanced modalities
- Conclusions

Ultrafast Electron Diffraction: Visualizing Dynamic States of Matter

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B. J. Siwick and M. R. Otto

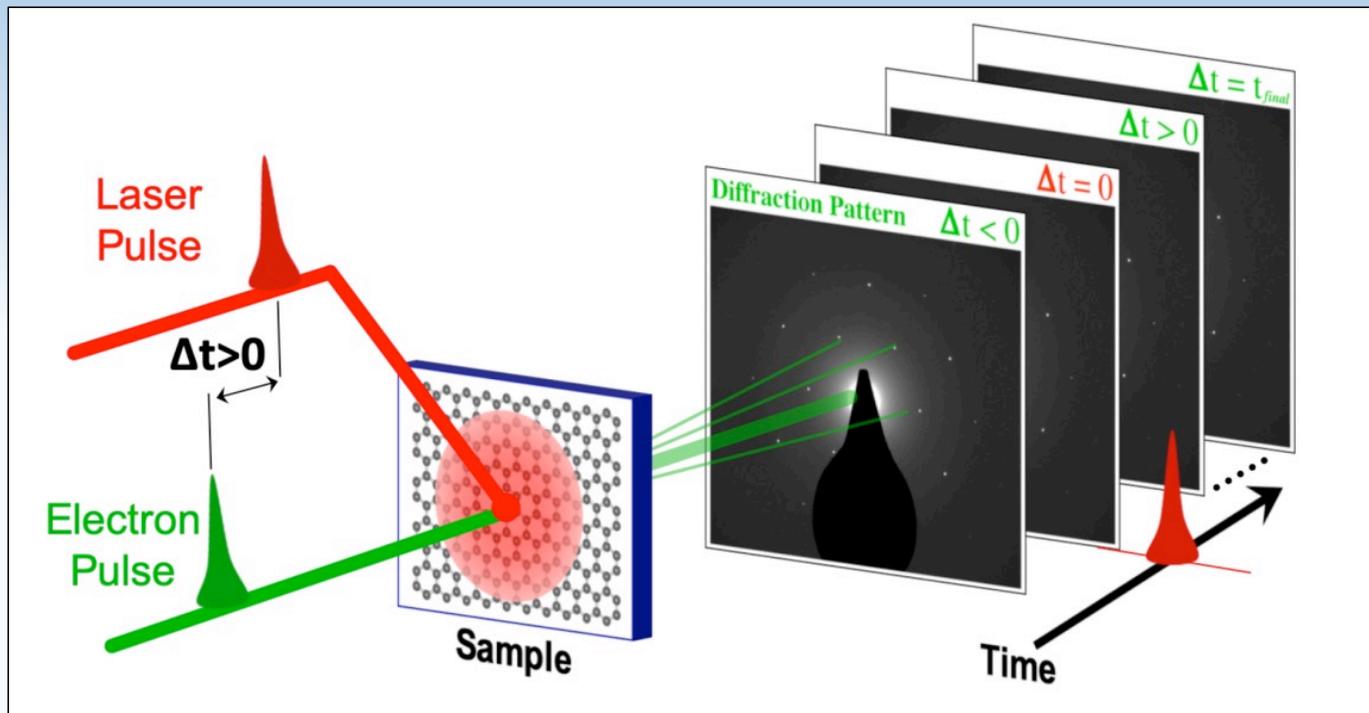
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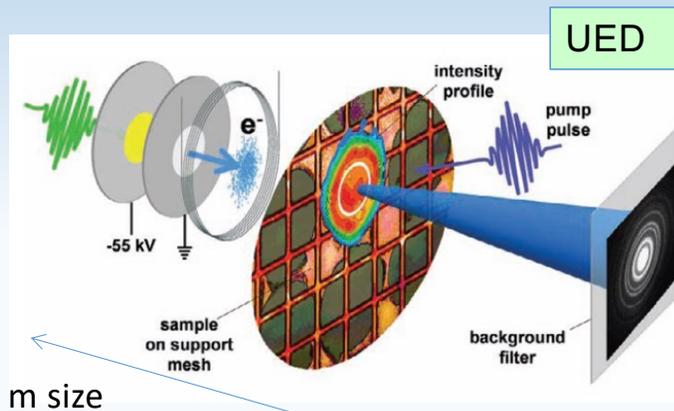
Review of Modern Physics, 94, 045004 (2022)

Ultrafast Electron Diffraction



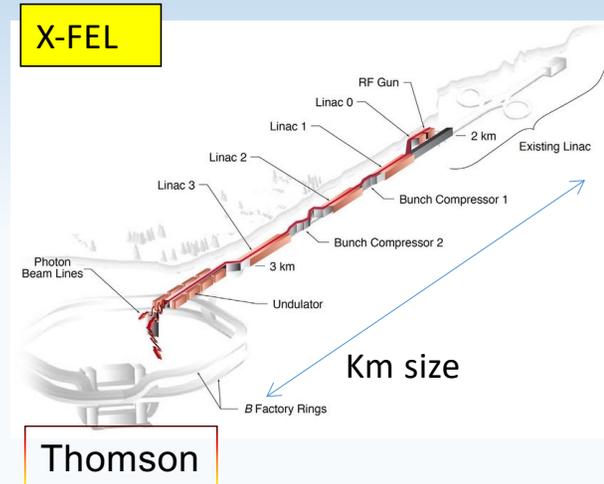
Some possible variations on the theme:
Electron beam energy
Excitation and probe pulse parameters
Sample phase
Repetition rate

Probing matter at its fundamental time and spatial scales photons vs. electrons



Rutherford

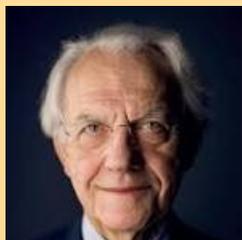
$$\sigma_{el} = \frac{h^2 Z^{4/3} c^2}{\pi E_0^2 \beta^2} = 8.57 \cdot 10^{-20} \text{ cm}^2 \text{ for } C$$



$$\sigma_T = \frac{8\pi}{3} r_e^2 = 6.65 \cdot 10^{-25} \text{ cm}^2$$

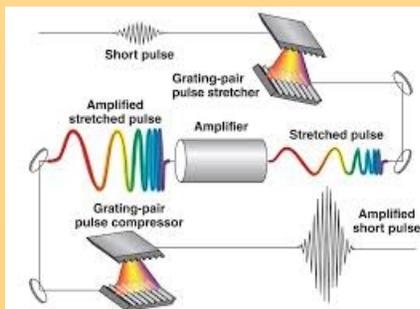
- Electrons provide direct access to structural information with compact tools
- Probing with electrons can be better for surfaces, thin films, gas phase
- Less damage to biological samples (400-1000 times)

Early days of UED: non relativistic electrons



G. Mourou

Developed CPA for ultrafast lasers
Pioneered use of electrons generated by short laser pulses for time-resolved material study

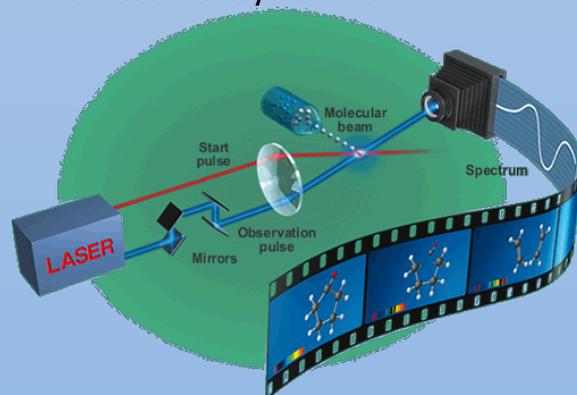


Mourou, Gerard, and Steve Williamson.
"Picosecond electron diffraction." *Applied Physics Letters* 41.1 (1982): 44-45.



A. Zewail

Nobel prize for femtochemistry
Realized that short wavelength probes like electrons are needed for structural dynamics



Zewail, Ahmed H. "4D ultrafast electron diffraction, crystallography, and microscopy." *Annu. Rev. Phys. Chem.* 57.1 (2006): 65-103.



D. Miller

Developed concept of molecular movie
Investigated space charge effects and limits in temporal resolution for keV UED



Siwick, Bradley J., et al. "An atomic-level view of melting using femtosecond electron diffraction." *Science* 302.5649 (2003): 1382-1385.

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 !

| | <i>UED</i> | <i>MeV ED</i> |
|------------------------------------|------------|---------------|
| 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 % |

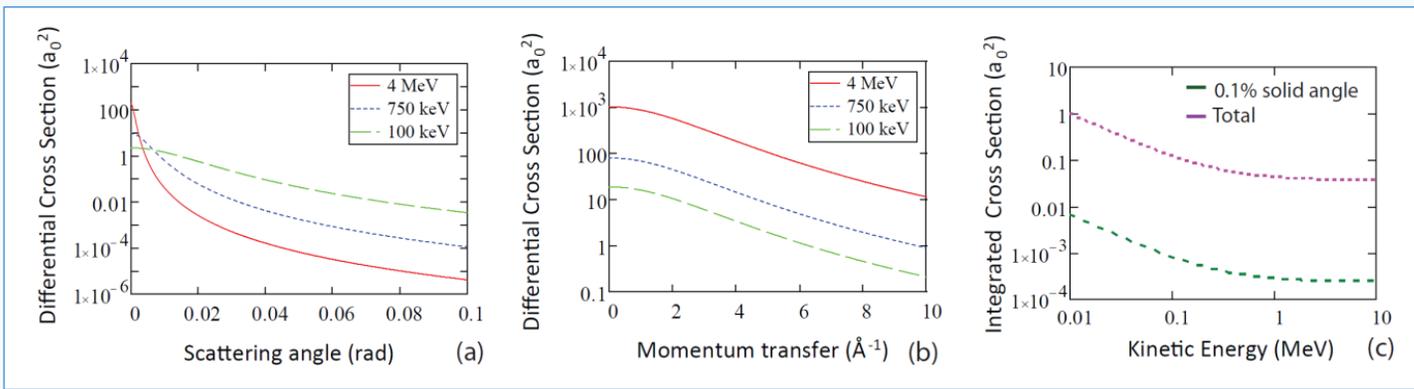
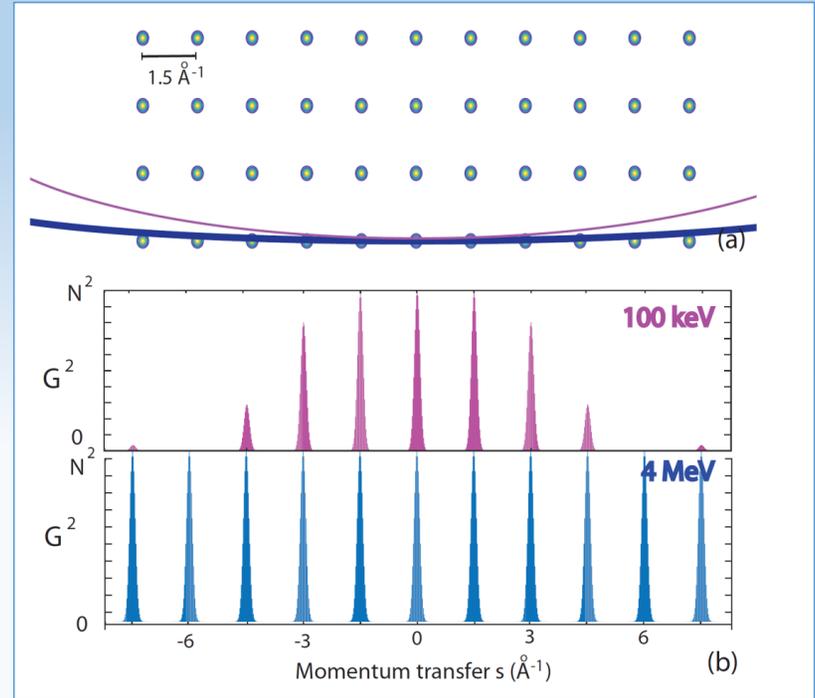
Highlight two important differences for MeV electrons

Sensitivity to higher order Bragg reflections

$$\lambda = h / p$$

Energy dependence of cross-section

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

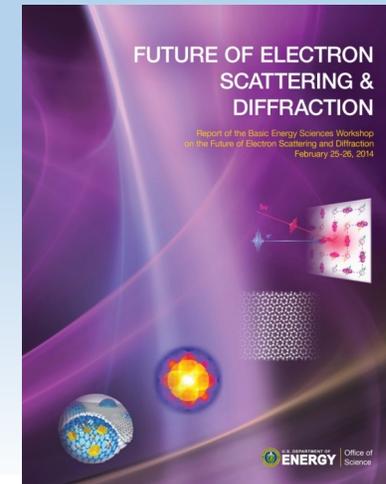


RF photoinjector based ultrafast MeV electron diffraction

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, Tsinghua, Japan, Korea, UK, Berlin, etc.



PHYSICAL REVIEW E

VOLUME 54, NUMBER 4

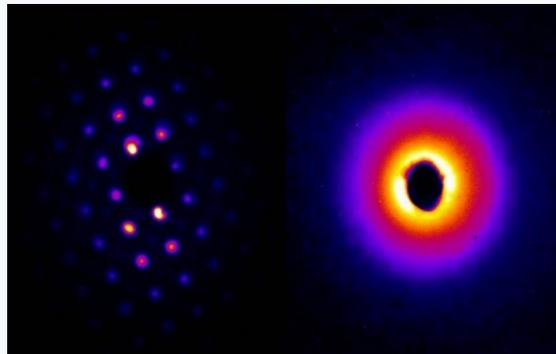
OCTOBER 1996

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)

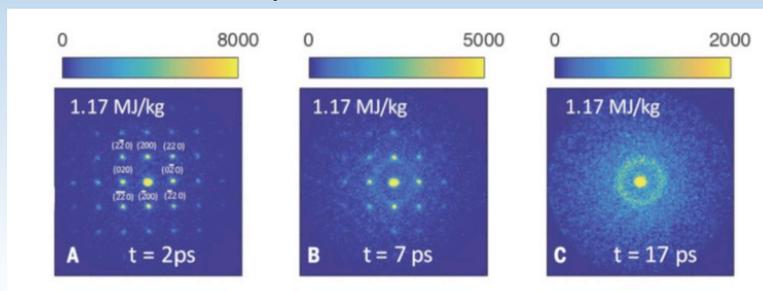
D. Filippetto and H. Qian, J. Atom. and Mol. And Opt. Phys. 49, (2016)

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

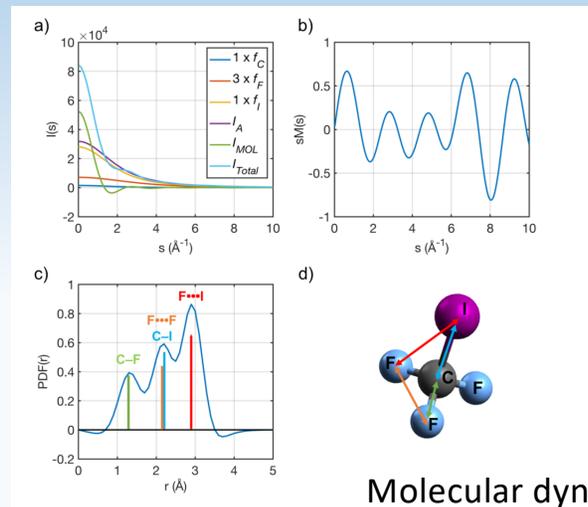
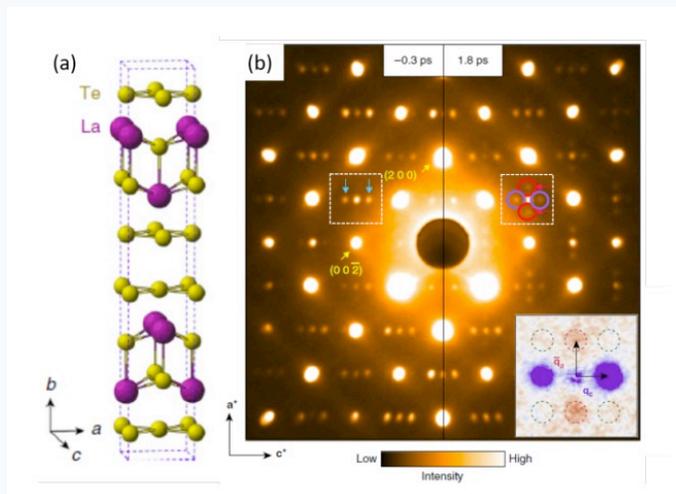
H. W. Kim et al., Nature photonics 14, 245 (2020)

Solid state and gas phase application examples

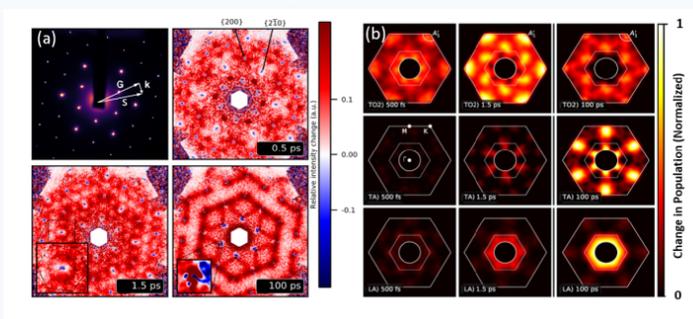
Ultrafast phase transitions



Charge density waves



Diffuse scattering: dynamic disorder



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 μm | Smaller spot sizes at sample plane | Heterogeneous materials |
| Absolute and Relative coherence length | 10 nm / 100 μm < 1e-4 | Higher transverse brightness | Larger unit cells, proteins |
| Temporal resolution | 50 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 | 50 ppm 0.02° of RF | Low level and high power RF. Laser system | Repeatability |
| Pump | IR, NIR, THz, <u>X-rays</u> | Synchronized pumps | Different excitation mechanisms |

High brightness electron sources

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

Relative coherence length

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

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

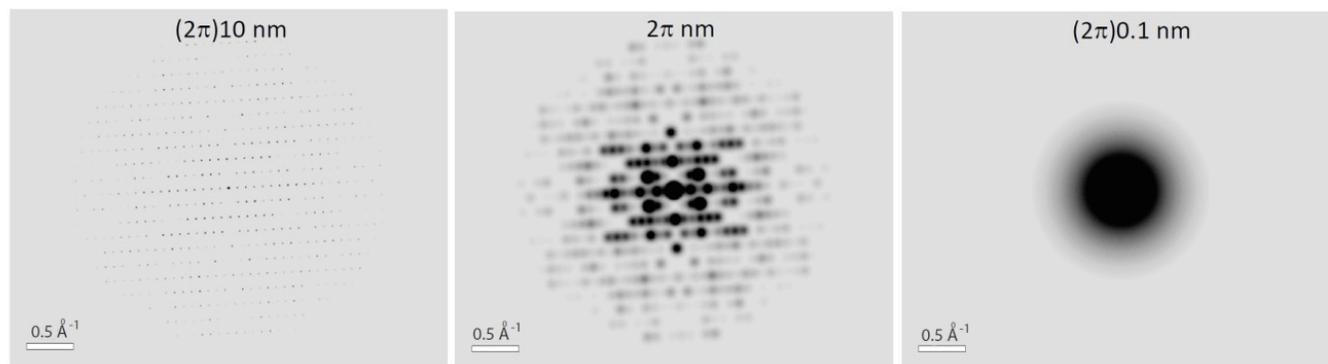
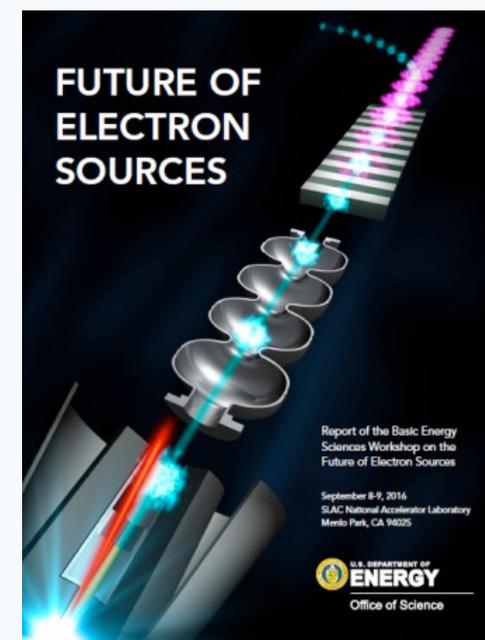


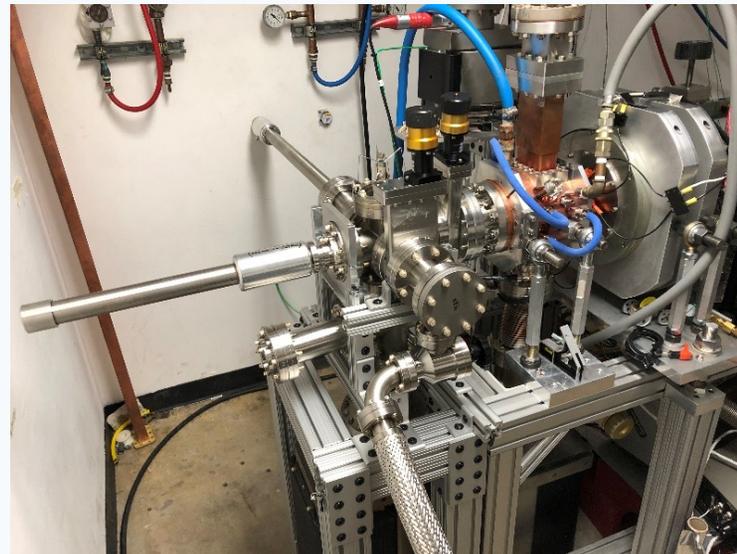
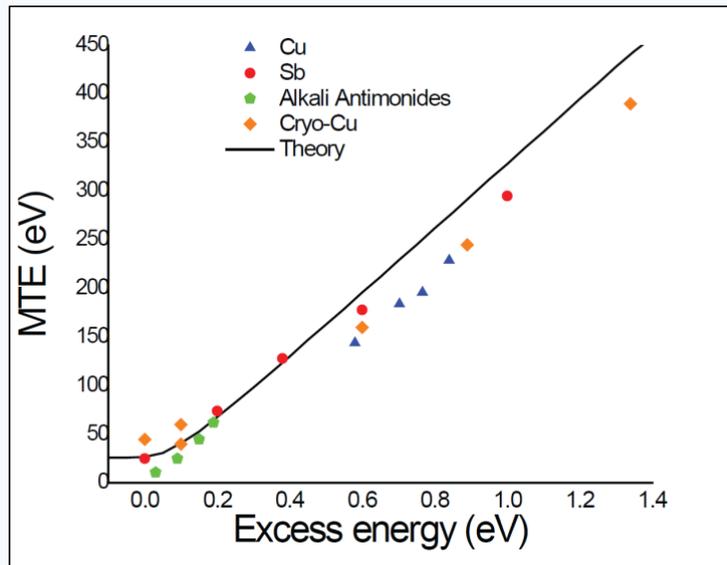
Figure 5 Simulated diffraction patterns of a Salicylic acid (aspirin) crystal for electron probe beams having coherence lengths of (2π) 10 nm, 1 nm and 0.1 nm respectively.

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

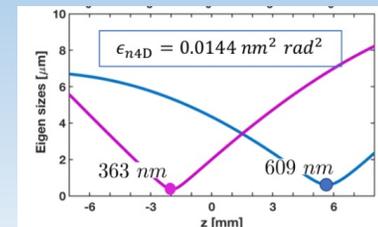
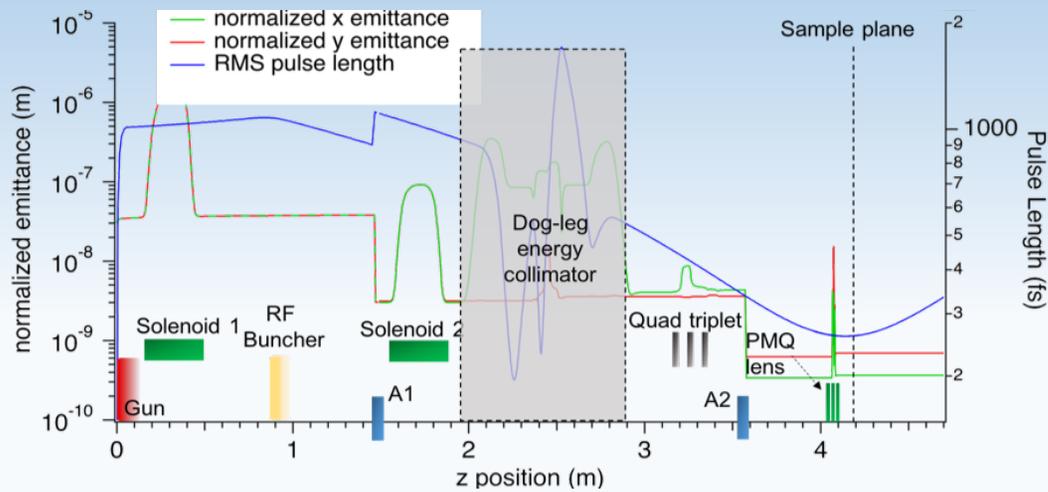


Advanced photocathode research

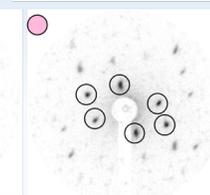
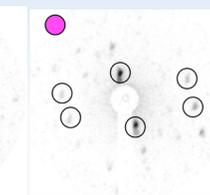
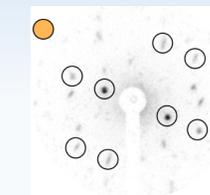
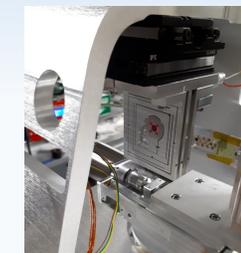
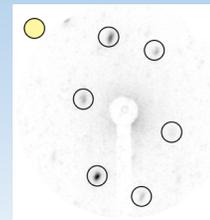
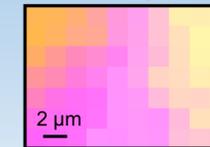
- Typical operation of Cu cathodes yields Mean Transverse Energies of few 100 meV
- 6 meV measured from cooled single crystal Cu...very low QE $\sim 10^{-8}$. Laser heating when trying to extract more power
- Semiconductor photocathodes show much promise with potentially more than one order of magnitude increase in coherence length of beam at sample.
- Need loadlock system and excellent operating vacuum $< 1e^{-9}$ (@ 10 Hz)



Transverse collimation



Scanning UED



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

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

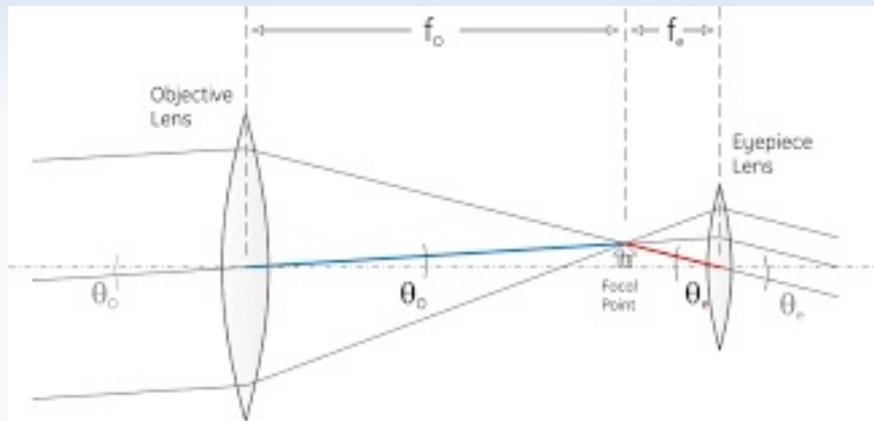
Sub-nm scale emittance in combination PM-based quadrupoles for tight focusing at sample plane

With collimation:

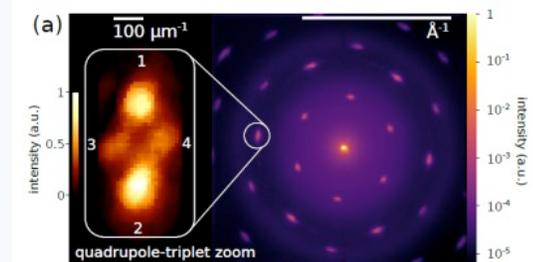
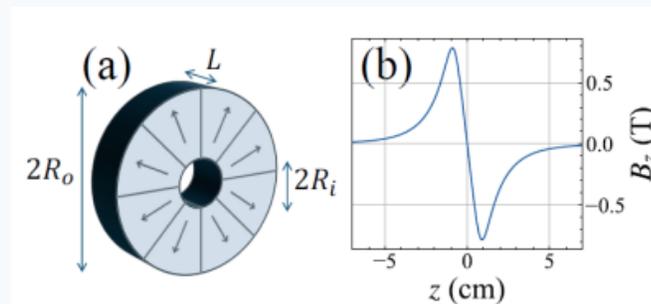
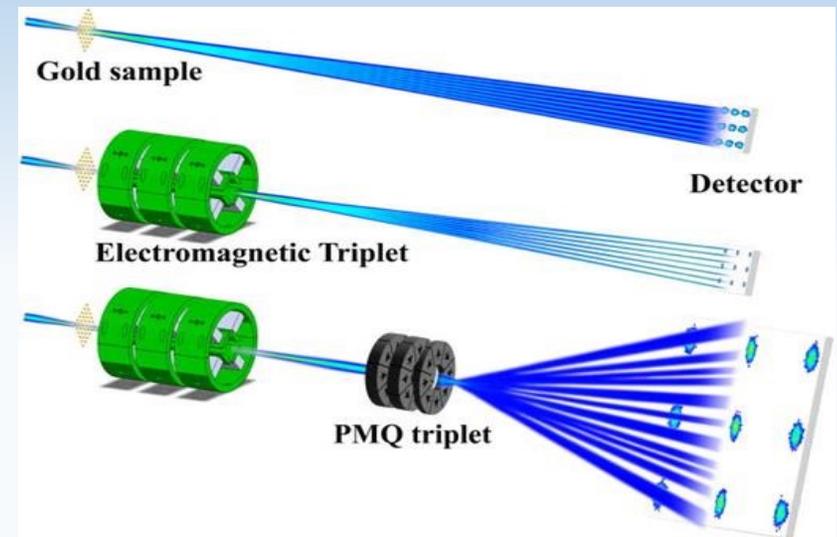
- Brightness stays constant !!!
 - Charge decreased with the square of the collimation radius
 - Emittance decreases linearly with the collimation radius
 - It may get slightly better if collimation is used to select core emittance

Improving reciprocal space resolution

- “Zoom in” on diffraction features adding magnetic lenses after the sample
- Telescope configuration for angular magnification ($m = f_o/f_e$)
- Could tune for variable diffraction camera length



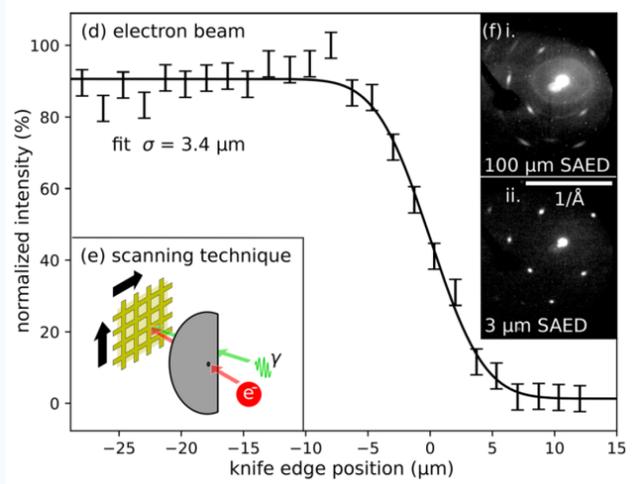
Angular magnification telescope



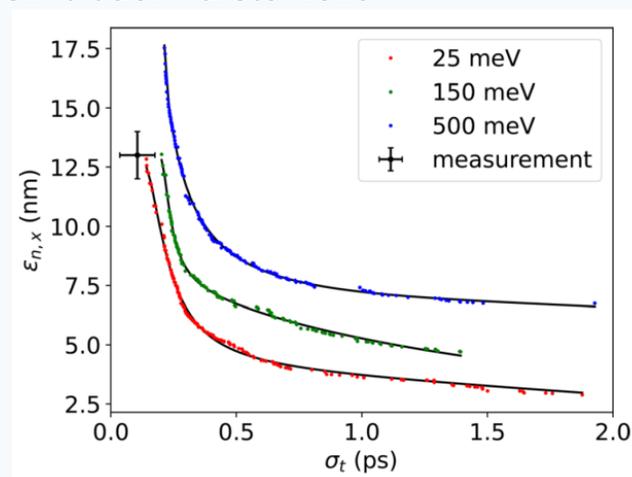
Accelerator and beam physics at non relativistic UED beamlines : MEDUSA @ Cornell

- Importance of beam brightness: photocathode and beam aperture
- Use of post-sample electron optical elements
- High repetition rate detectors

Alkali-antimonides + aperture



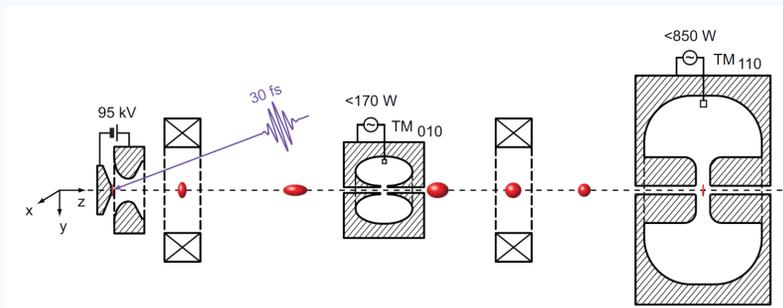
Simulation Pareto front



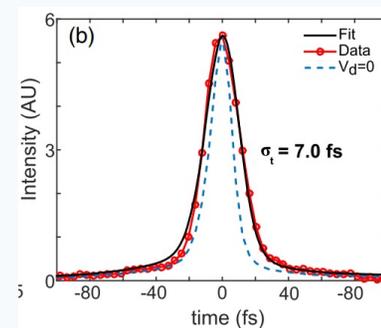
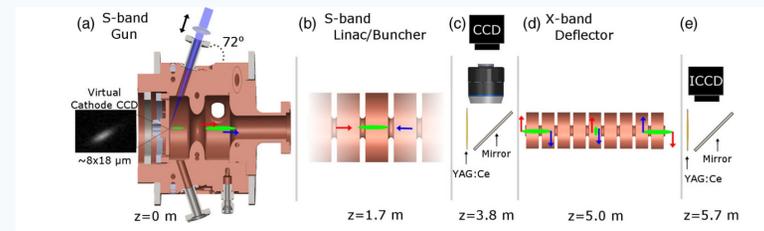
W. H. Li et al. A kiloelectron-volt ultrafast electron micro-diffraction apparatus using low emittance semiconductor photocathodes featured. Structural Dynamics 9, 024302 (2022)

RF compression

- Standard technique in accelerator and beam physics
- Introduced to compensate longitudinal expansion first in keV sources
- Clearly can also be applied to MeV beams
- High harmonic linearization for even shorter bunches
- Suffers from timing jitter
- Laser-based THz compression to synchronize



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)



Maxson et al., PRL 118, 154802 (2017)
 L. Zhao. *Phys. Rev. X* 8, 021061 (2018)

Temporal resolution

For a typical pump and probe experiment the temporal resolution is given by

$$(\Delta t)^2 = (\Delta t_{laser})^2 + (\Delta t_e)^2 + (\Delta t_{VM})^2 + (\Delta t_{jit})^2$$

Pump pulse length
 ≈ 10 fs

Duration of probe
relativistic electrons ≈ 10 fs

Timing jitter
Synchronization and
time of arrival
fluctuations

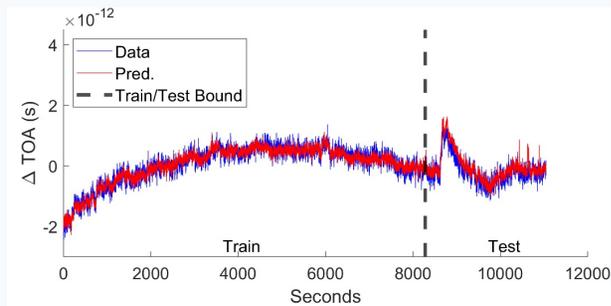
Velocity mismatch
Depends on the geometry of interaction.
Spot sizes and angles of pump and probe beams.
For 5° and $50 \mu\text{m}$ spot size $\Delta t_{vm} \approx 10$ fs

*In early days the probe length was the limiting factor.
But will we be able to take really advantage of shorter probes?*

Time stamping

Software based

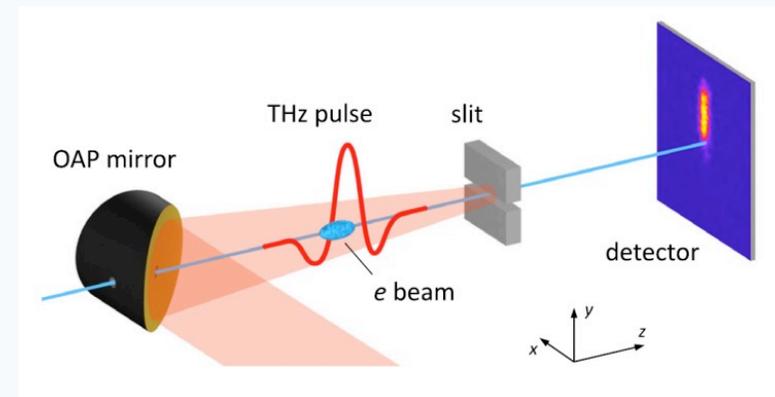
- Measure all accelerator parameters
- Tag each shot with reconstructed time-of-arrival information
- Train neural network to predict time of arrival using data from independent diagnostics
- Demonstrated at LBNL to perform at 150 fs level, better than conventional feedback systems on similar time-scales.



F. Cropp et al., PRAB, 26, 052801(2023)

Hardware based

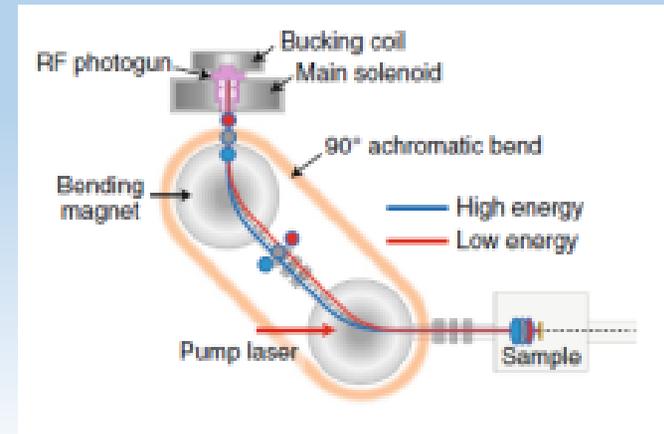
- EOS or e-beam induced index of refraction changes
- Laser-generated THz can be used to measured time-of-arrival (and bunch length at the same time)



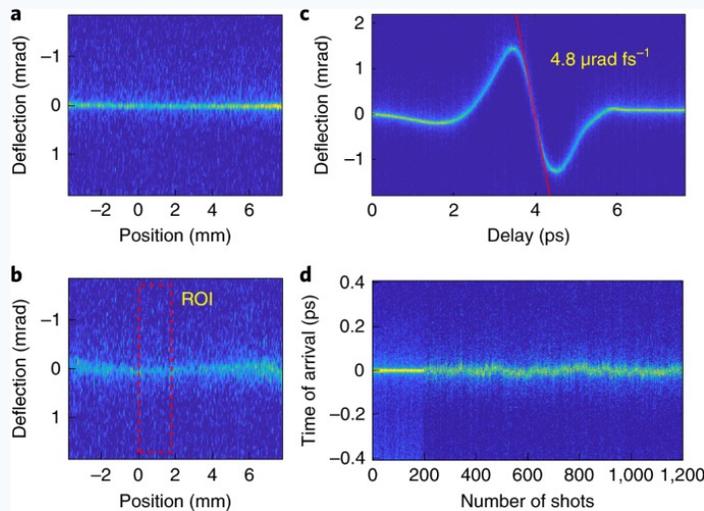
J. Fabiańska et al., Scientific Reports, 4 5645 (2014)
C. Kealhofer et al., Science 352, 429 (2016).
L. Zhao. Phys. Rev. X 8, 021061 (2018)
R. K. Li, Phys. Rev. Accel. Beams **22**, 012803 (2019)
X. Shen et al., AIP Advances **9**, 085209 (2019)

Magnetic compression

- Fully passive system
- Relies on space charge and gun induced chirp
- Multi bend achromat or alpha magnets
- Non linearities can be managed
- Record temporal resolution demonstrated



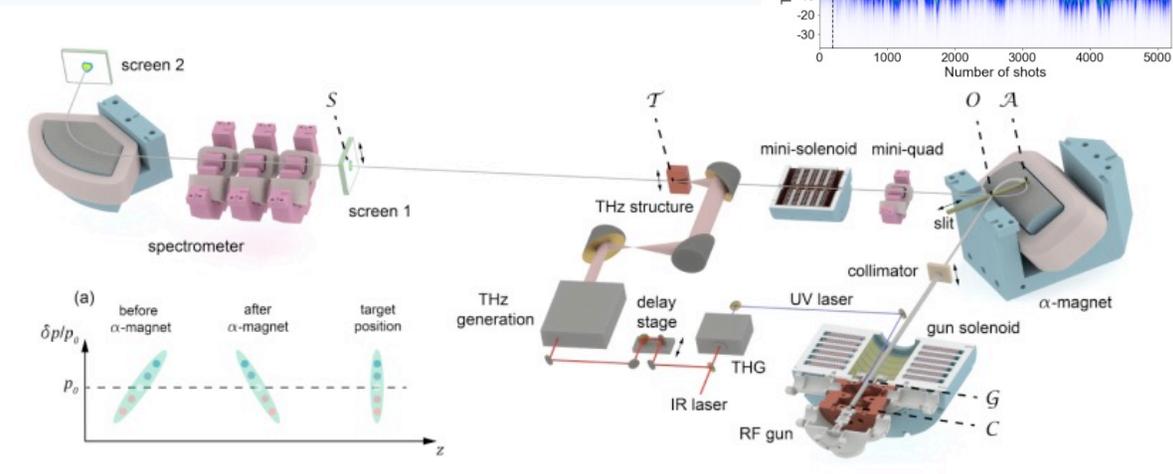
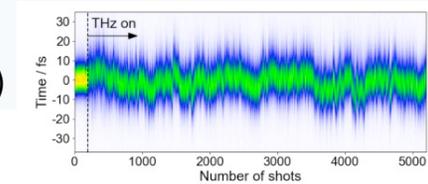
THz streaking



H. W. Kim et al., Nat. Phot., 14, 245 (2020)

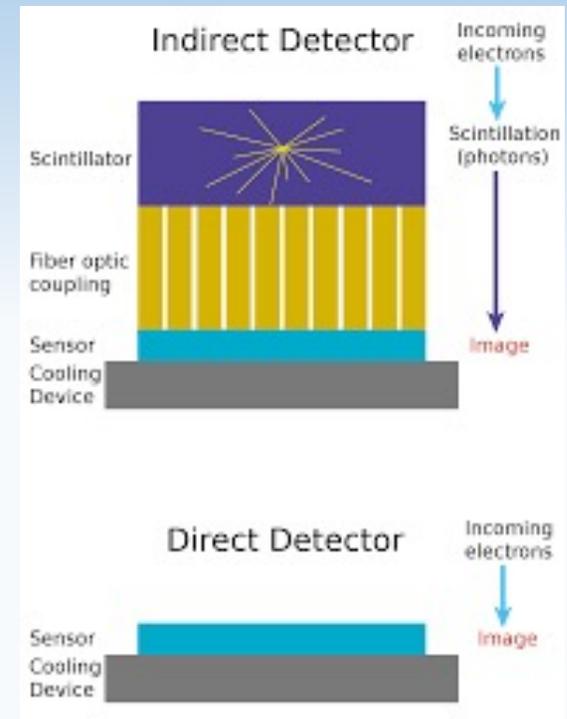
F. Qi et al., Phys. Rev. Lett. 124, 134803 (2020).

Y. Yang et al. <https://arxiv.org/abs/2508.03946> (2025)



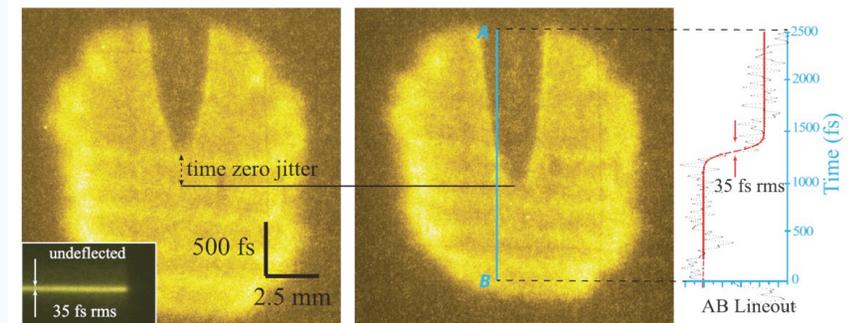
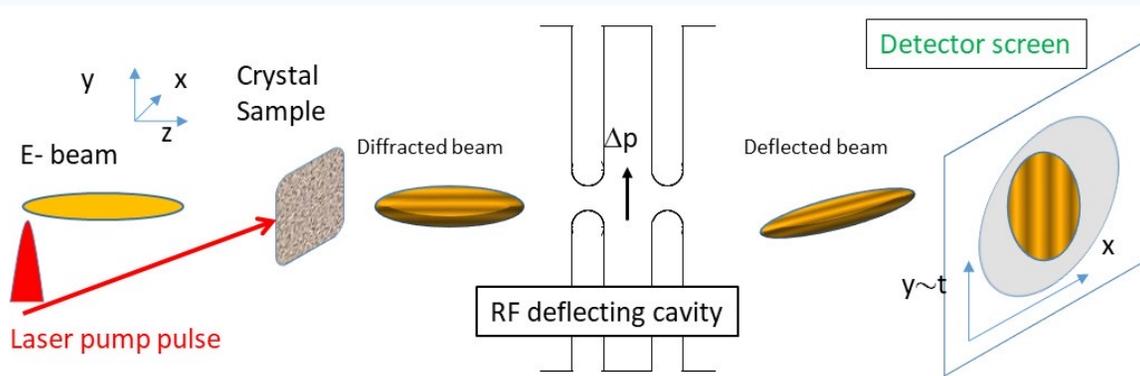
Detectors

- Free-space or fiber-coupled screen + intensified camera
 - Sensitivity vs. resolution
- Direct electron detection
 - > 80 % DQE, large dynamic range, electron counting mode
- Central hole for direct beam diagnostics (charge, time-stamping, energy, pointing)
- High repetition rate readout for laser on/off contrast improvement.
 - Need high rep-rate source



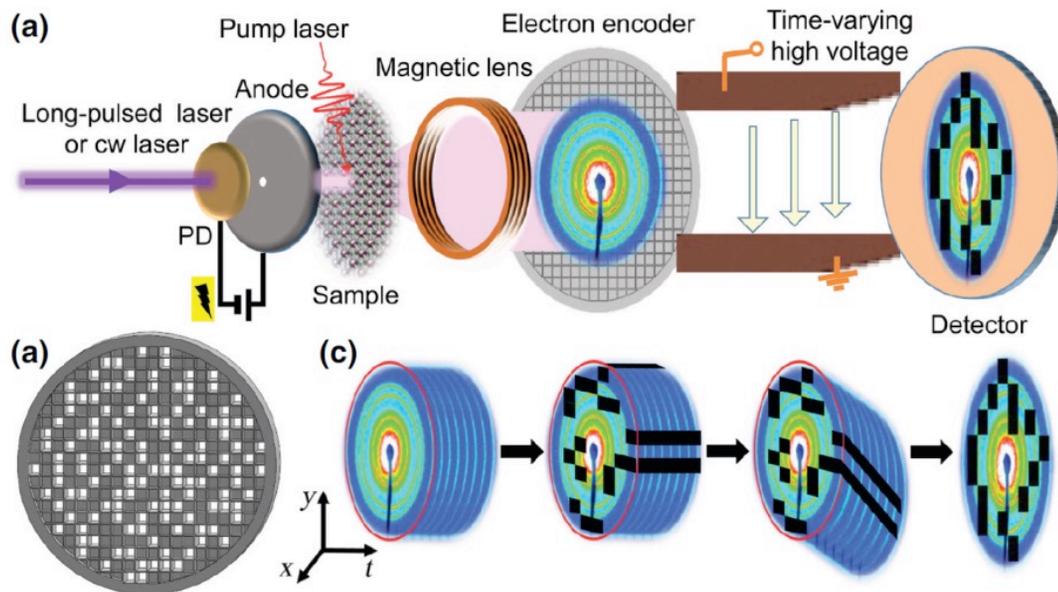
Streaked MeV UED

- Capture entire history of ultrafast process in one shot (full movie in single exposure)
- 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. Soby et al., APL, 102, 023506 (2013)
P. Musumeci et al. JAP, 108, 114513 (2010)

Smart image reconstruction algorithms and masks can greatly help

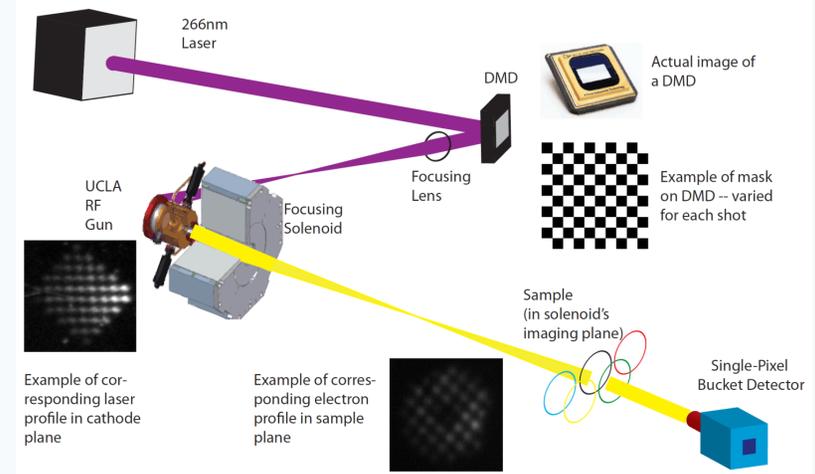


Compressed Ultrafast Electron Diffraction Imaging Through Electronic Encoding
D. Qi et al. **Phys. Rev. Applied** **10**, 054061 (2018)

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.

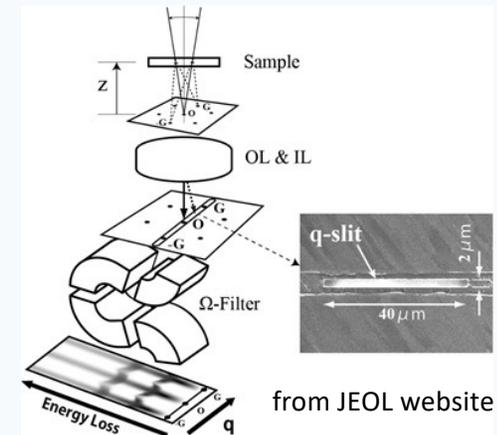
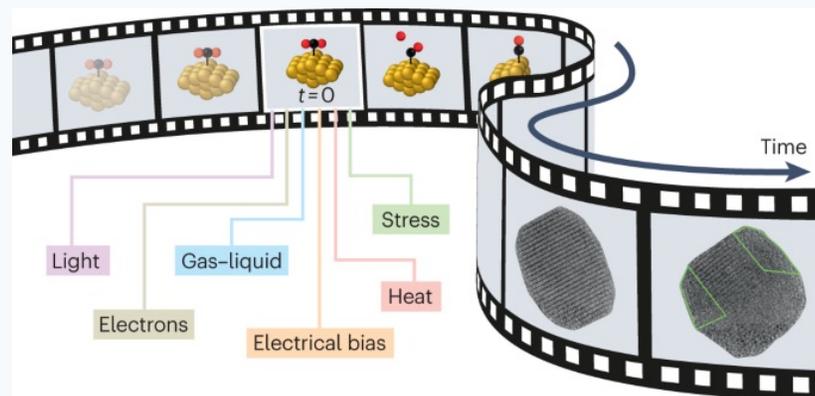
K. Kabra, S. Li, F. Cropp, T. J. Lane, P. Musumeci, D. Ratner. Mapping Photocathode Quantum Efficiency with Ghost Imaging. [Phys. Rev. Accel. Beams](#) **23** 022803 (2020).

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Combining UED with other capabilities/modalities

- FEL pump / UED probe experiments at FERMI FEL
- THz pump / UED probe
- Electron beam pumping ?
- Use postsample optics for real space imaging of grain boundaries (microscopy)
- Energy analyze diffraction patterns for momentum resolved EELS.



Conclusions and outlook

- Ultrafast electron sources (MeV and keV) are pushing the boundary of spatial and temporal resolution in structural dynamics
- Advanced photocathodes and improving beam brightness
 - Longer coherence lengths, nanodiffraction, diffuse scattering
- Towards sub-10 fs temporal resolution
 - Magnetic compression, phase space linearization, THz compression, time stamping
- Streaked electron diffraction
 - Highest temporal resolution, no jitter problems, truly single shot, image reconstruction algorithms
- Multimodal UED
 - Imaging, Spectroscopy, X-rays, THz.