Atomic spatiotemporal imaging with MeV bright electron beams

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Acknowledgement

- Helpful discussions with many colleagues and friends in the electron source / accelerator / ultrafast science communities (Pietro Musumeci, Daniele Filippetto, Jared Maxson, and many others)
- R&D on UED key technologies at Tsinghua and UCLA
- UED facility and sciences work at SLAC (PI: Xijie Wang, Team, Collaborators, SLAC AD, TID, and LCLS)
- Many collaborators and users around the world

Outline

- Introduction
- State-of-the-art MeV UED instruments
- Recent scientific outcomes
- Toward better performances
- Summary and outlook

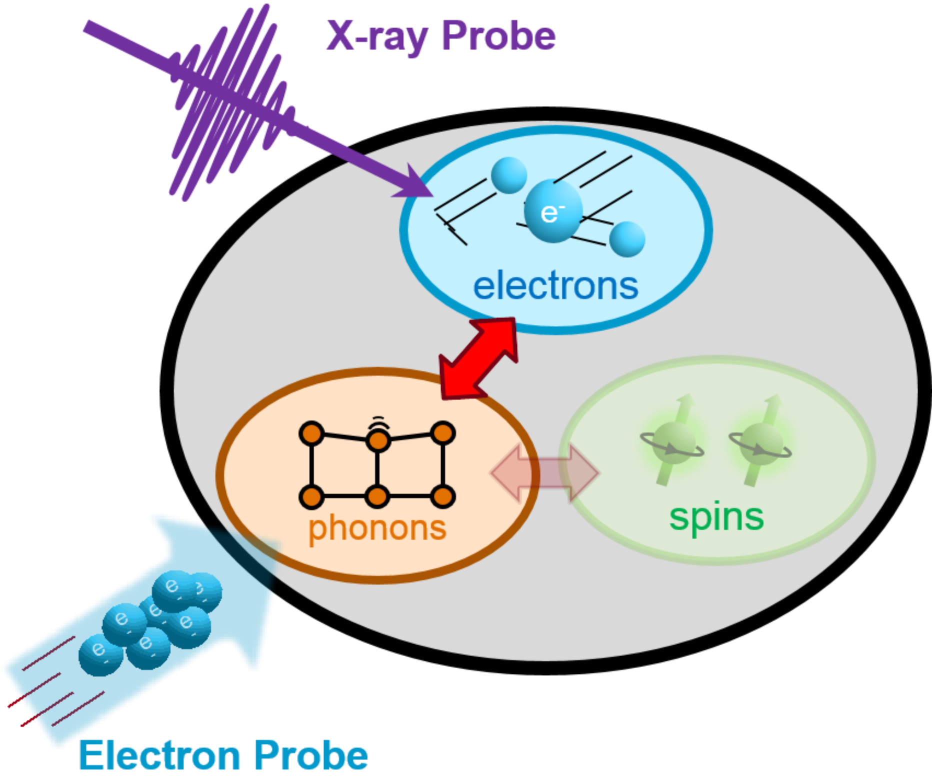
Visualizing the ‘ultrasmall’ and ‘ultrafast’

Ultrasmall:

short wavelength

Ultrafast:

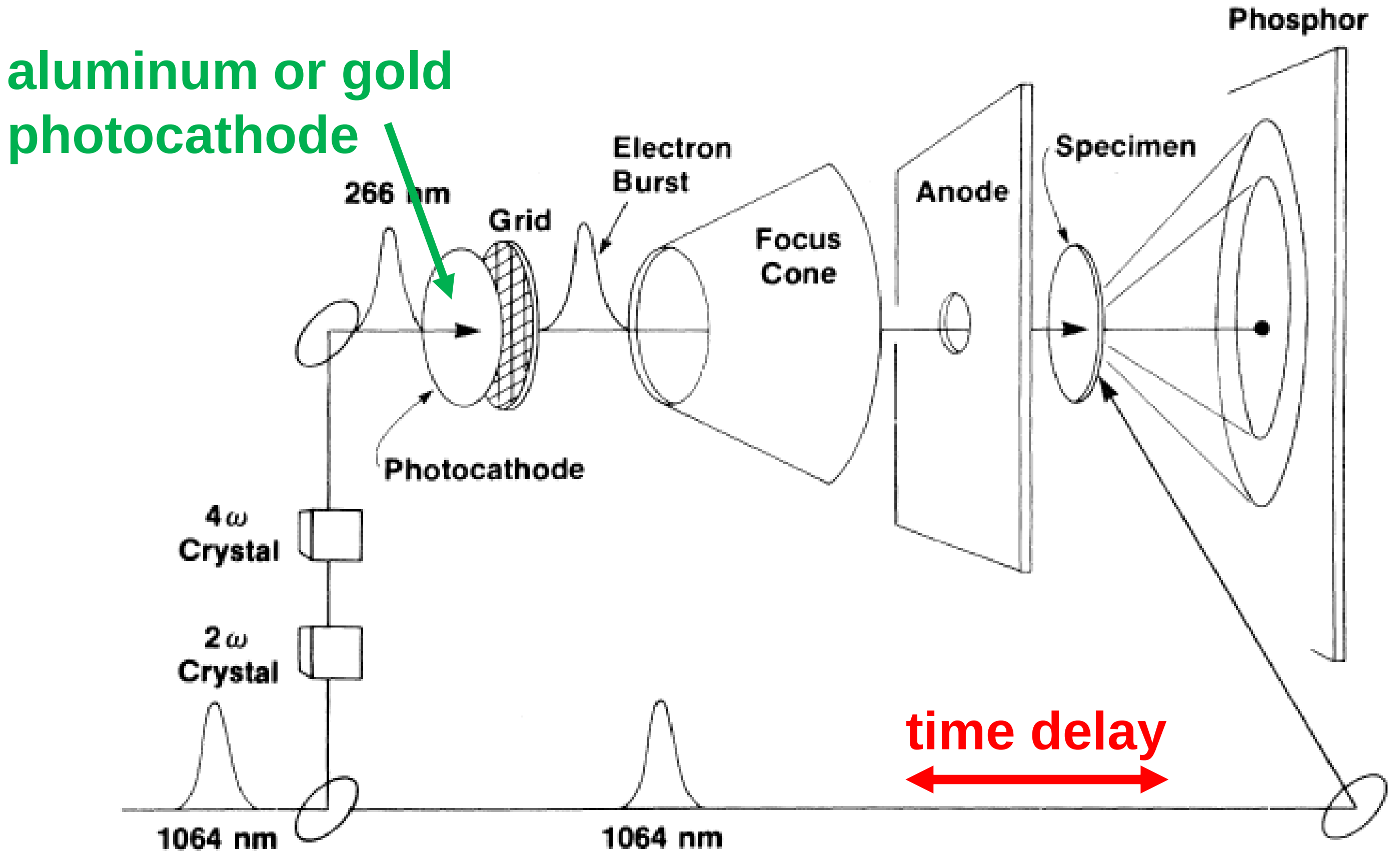
short pulse duration



XFEL and UES are complementary tools towards a complete picture of dynamics

UED – ultrafast electron diffraction

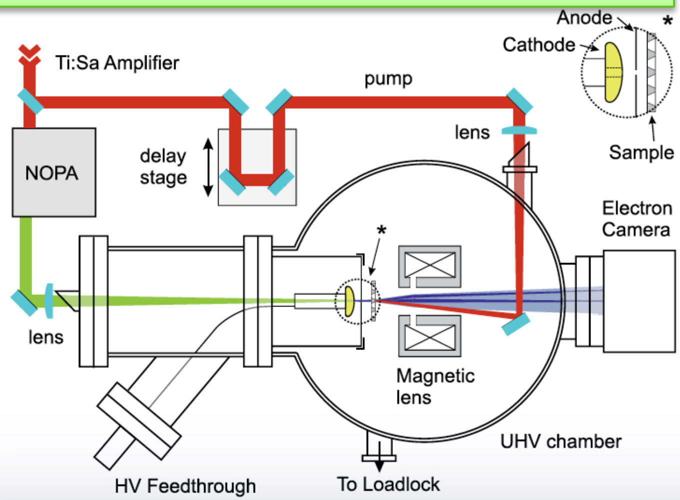
G. Mourou and S. Williamson, APL 41, 44 (1982)
S. Williamson, G. Mourou and J. C. M. Li, PRL 52, 2364 (1984)



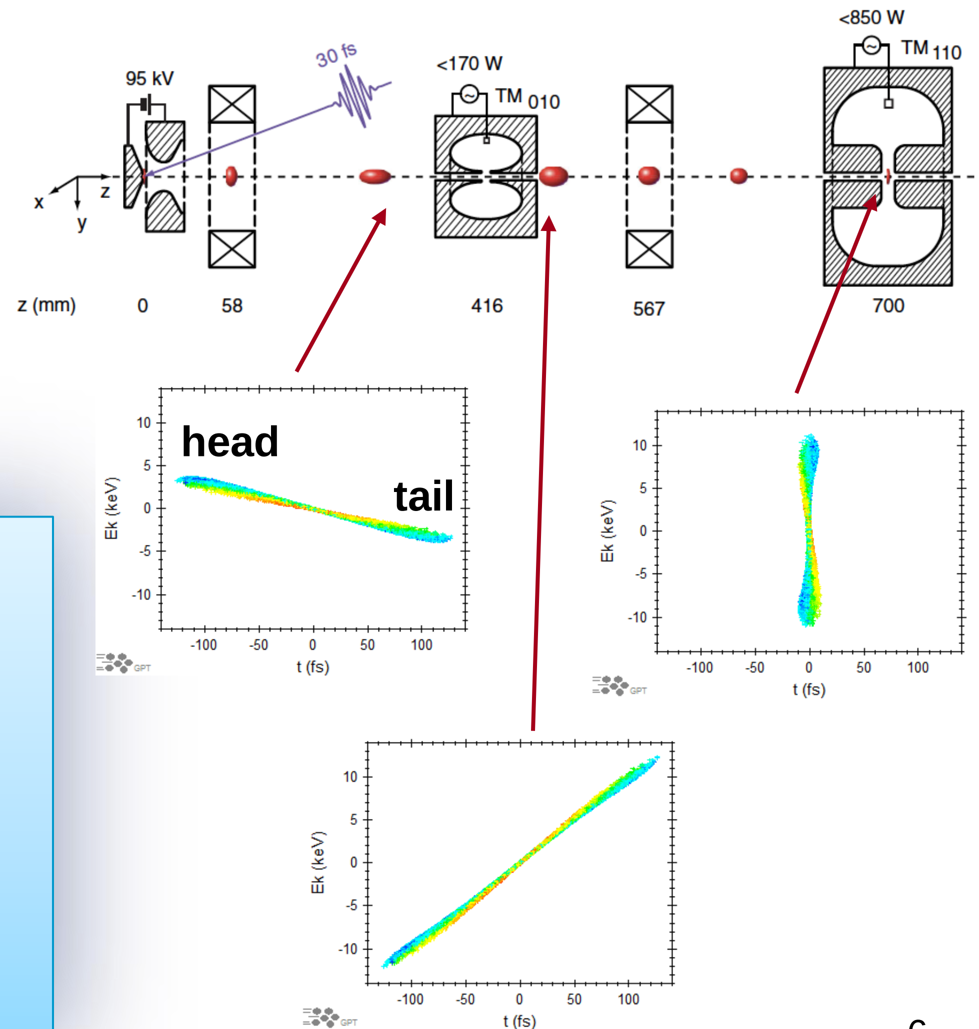
Science outcome: See e.g. M. Chergui and A. H. Zewail, *ChemPhysChem* **10**, 28 (2009); R. J D. Miller, *Science.* **343**, 1108 (2014) and etc.

State-of-the-art keV UEDs

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



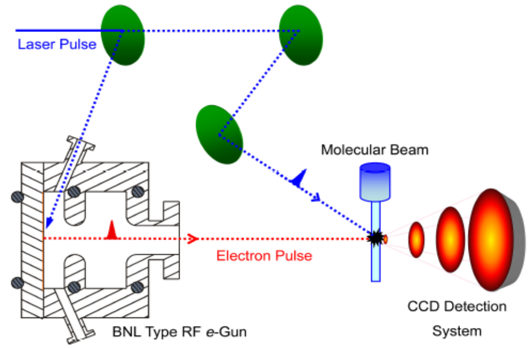
T. van Oudheusden et al., PRL 105, 264801 (2010)



- *Compact DC gun or w/ rf compression*
- *Up to 10^6 e- per pulse, 100-200 fs fwhm pulse duration*
- *Flexible rep-rate (usually kHz to allow samples to relax)*
- *Study solid state (thin nanofilm) samples*

Comparison between keV and MeV

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



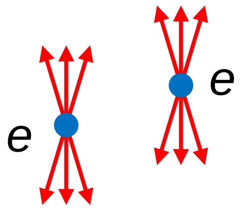
X. J. Wang, Z. Wu, H. Ihee, PAC'03, 420-422 (2003).

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

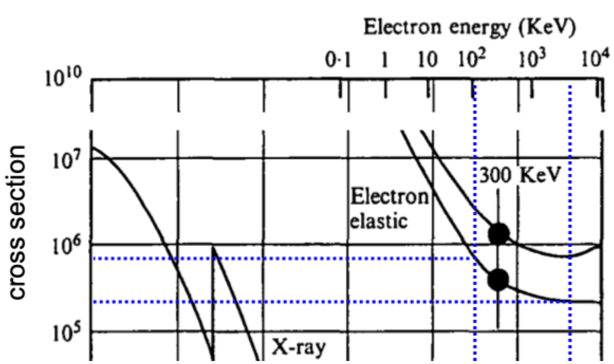
Active R&D field around the world!

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

- ✓ Shorter bunch
- ✓ Higher charge



Penetration/multiple scattering

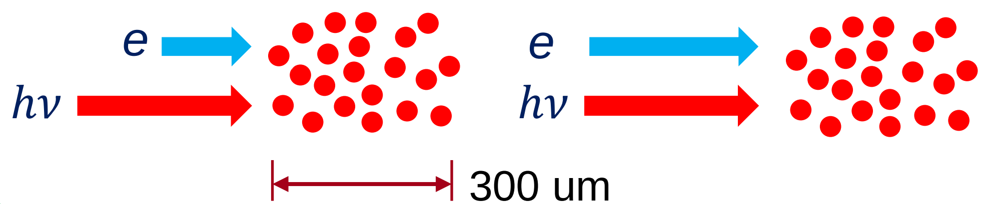


R. Henderson, Q Rev. Biophys. 28, 171 (1995)

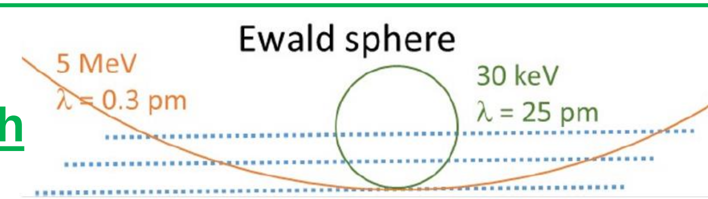
Velocity mismatch

100 keV t_{VM}=0.8 ps

3 MeV t_{VM}=10 fs

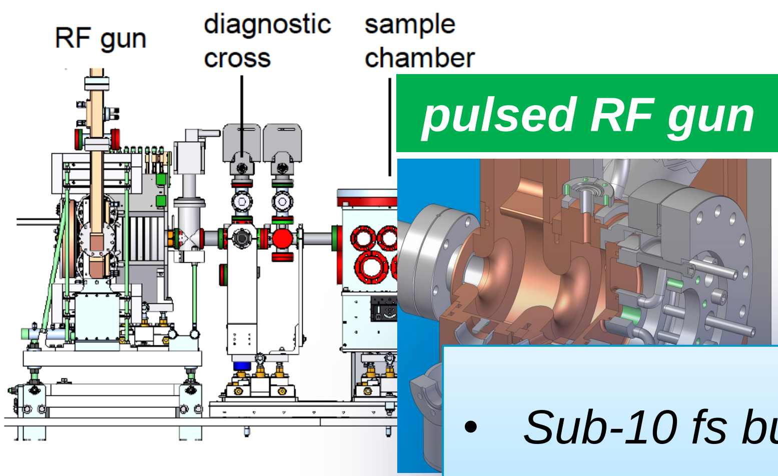


Shorter wavelength

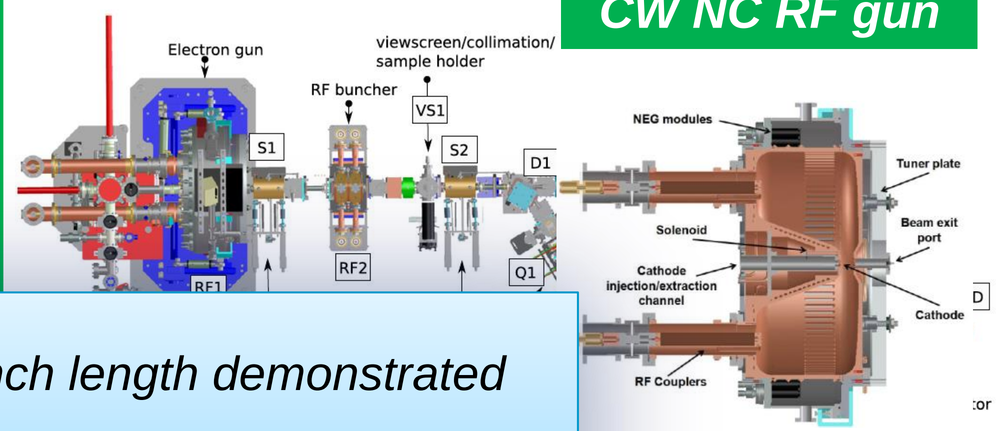


MeV UEDs with rf guns/boosters

pulsed RF gun

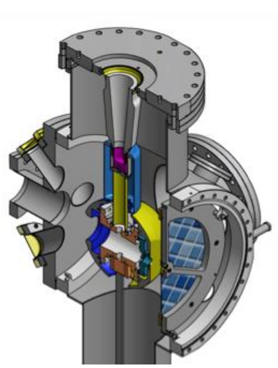


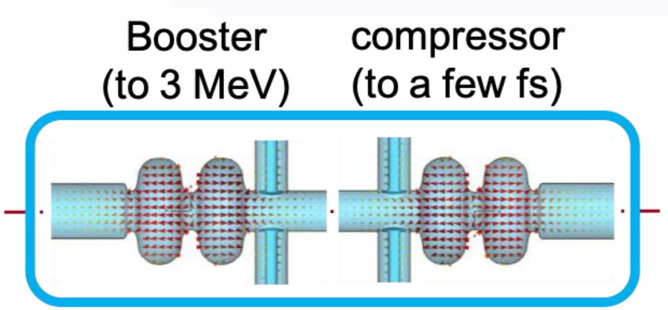
CW NC RF gun



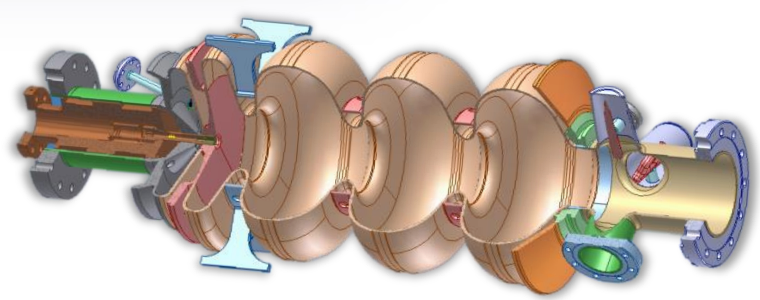
- *Sub-10 fs bunch length demonstrated*
- *need to minimize time-of-arrival jitter*

DC gun + booster



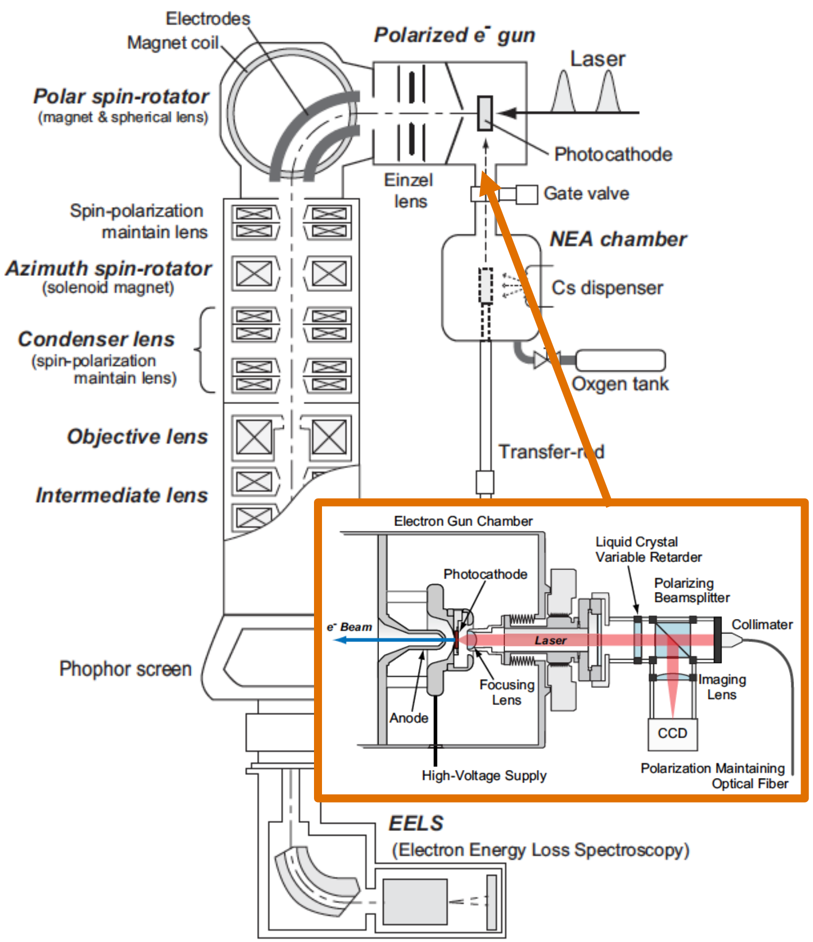


SRF gun



More source options for UED

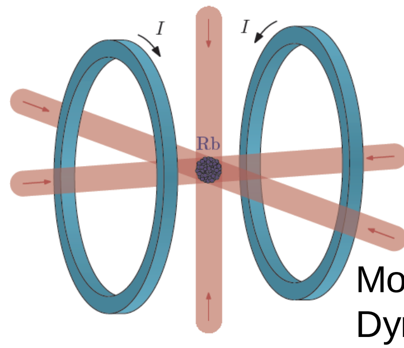
Spin-polarized TEM

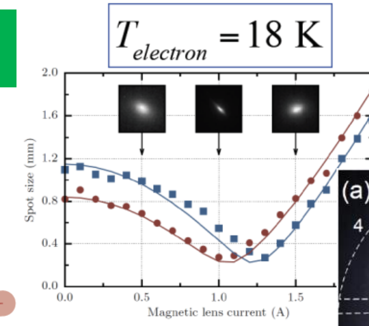


Kuwahara et al. APL 101, 033102 (2012)

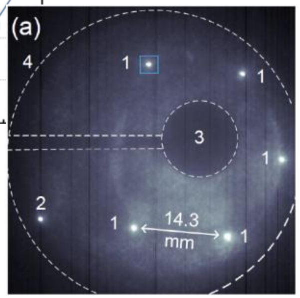
MOT source

Laser cooling & trapping



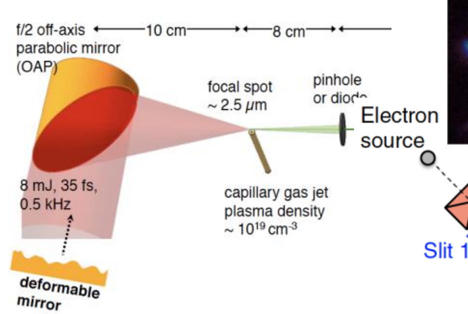


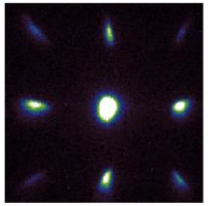
Graphite DP (13.2 keV)

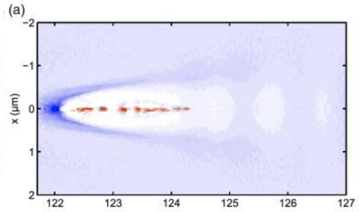


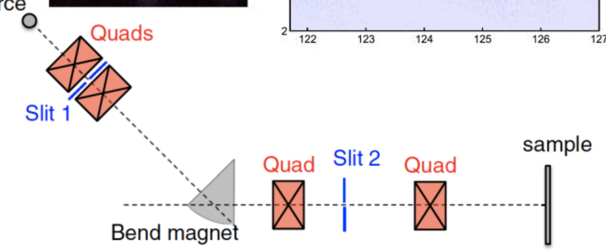
Mourik et al., Struc. Dyn. 1, 034302 (2014).

LPWA source







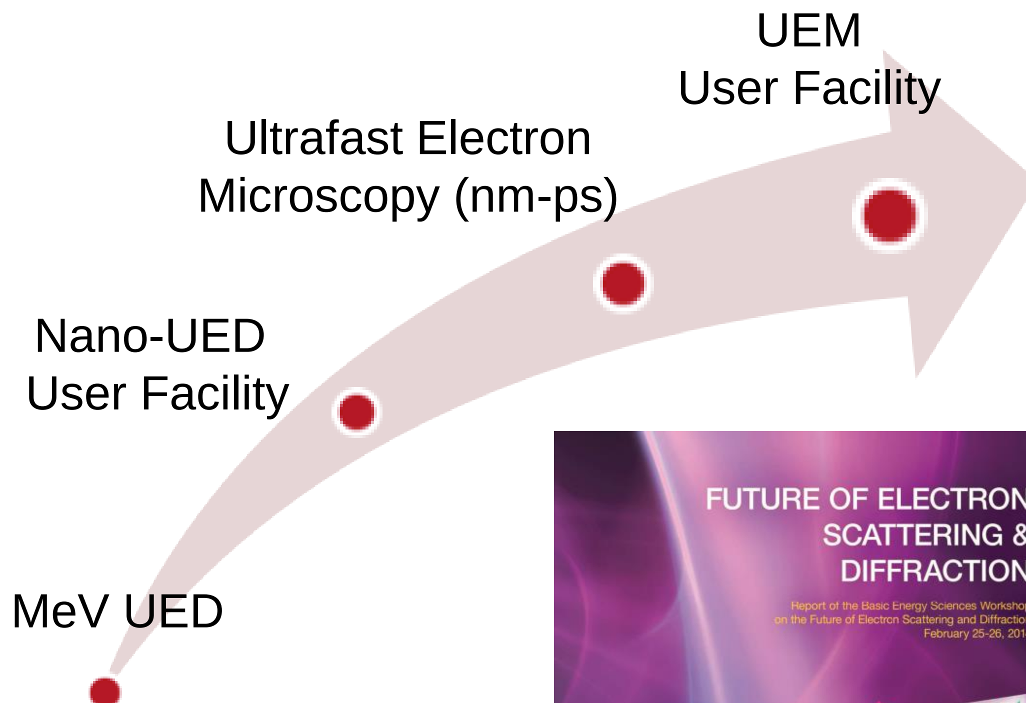


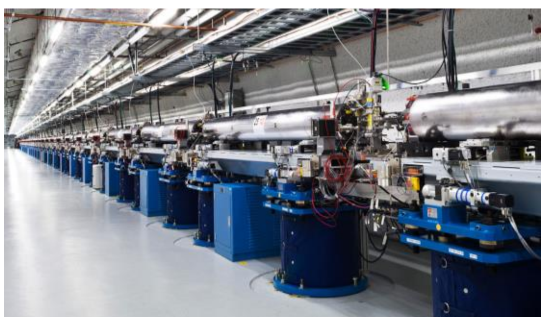
Z. He et al., NJP 15, 05316 (2013)

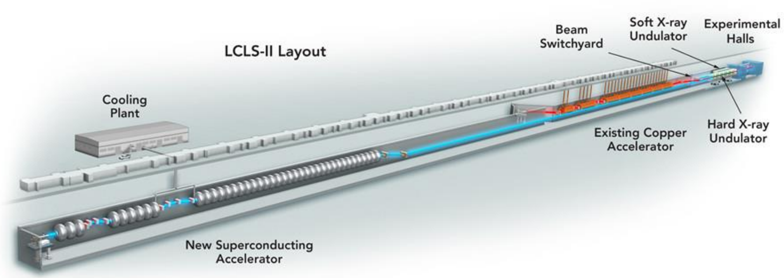
J. Faure et al., PRAB 19, 021302 (2016)

SLAC's vision for UED and UEM

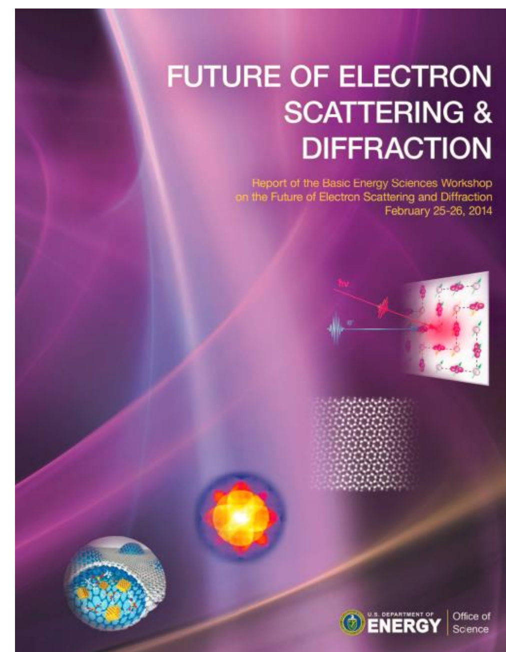
***SLAC UED/UEM Initiative**:*
"… to provide the world's leading ultrafast electron scattering instrumentation."







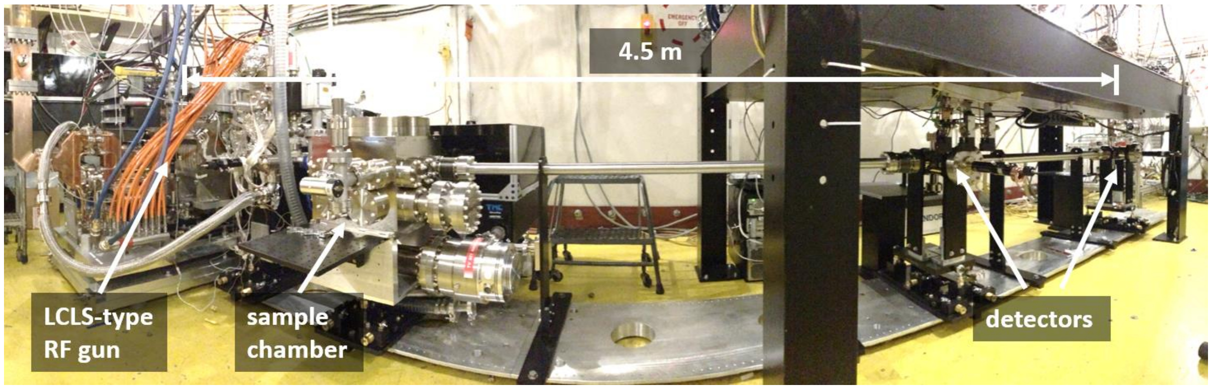
Future of Electron Scattering and Diffraction Workshop February 25-26, 2014



Performance of the SLAC MeV UED system

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

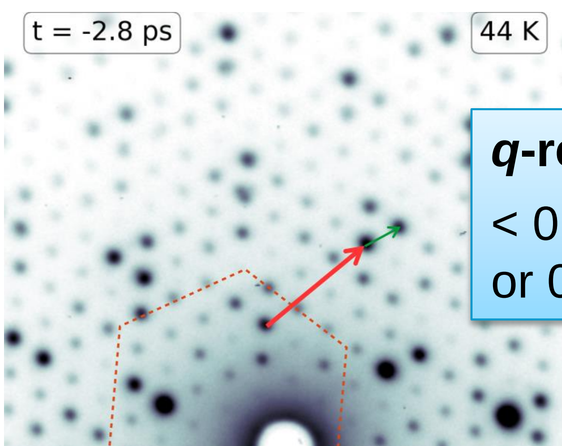
fs laser-rf timing, ultra-stable HV modulator, Ti:Sa laser, etc.



Typical beam parameters

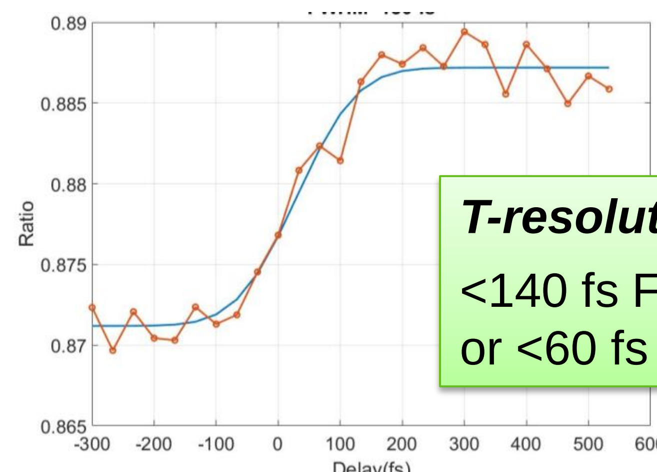
Parameters	Values
rep. rate	SS - 180 Hz
beam energy	2 - 4 MeV
bunch charge	10^4-10^6
emittance	2 - 20 nm
bunch length	<50 fs rms

L. Le Guyader et al.



***q*-resolution**

< 0.17 Å^{-1} FWHM or 0.07 Å^{-1} rms

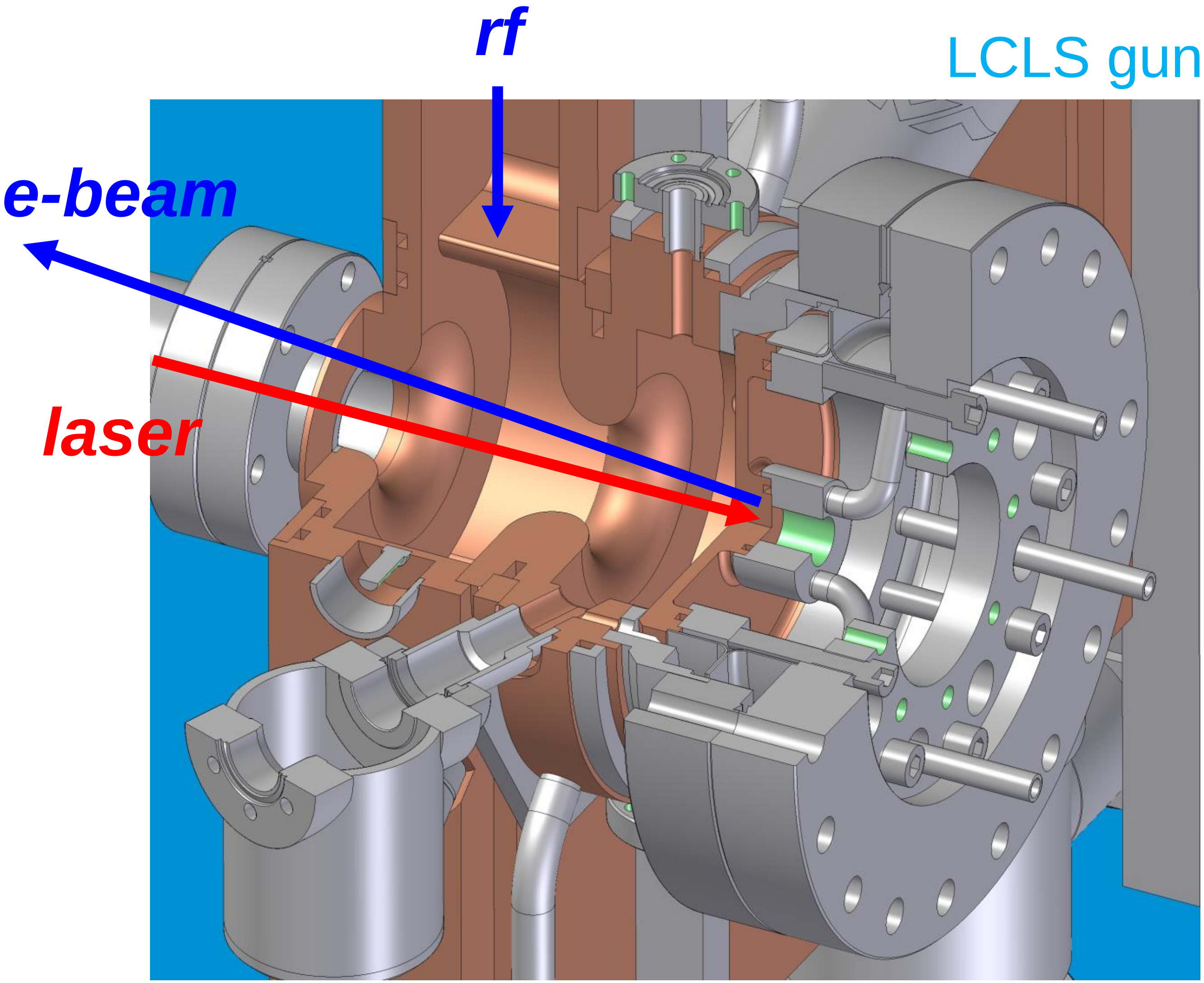


J. Yang et al.

***T*-resolution**

<140 fs FWHM or <60 fs rms

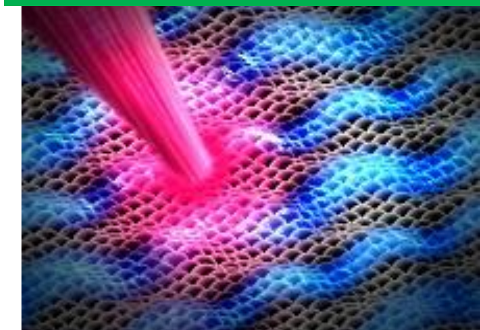
Photocathode rf gun



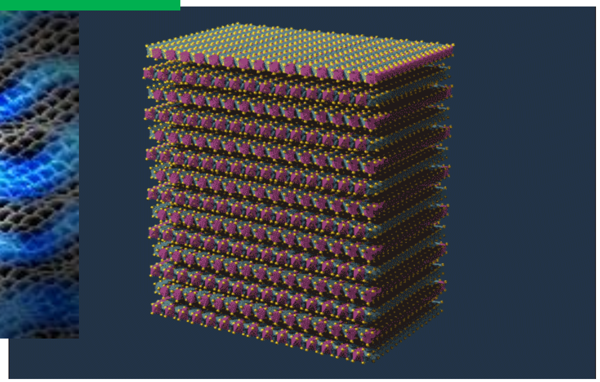
- High brightness
- Enabled XFEL, inverse Compton scattering, etc.
- Extremely flexible $1\text{-}10^9$ e-/pulse
- Reliable

Science outcome from the SLAC UED machine

2D and layered materials

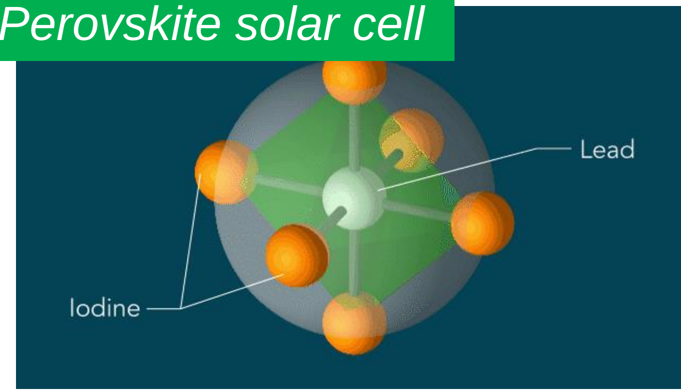


Nano Letters 15, 6889 (2015)



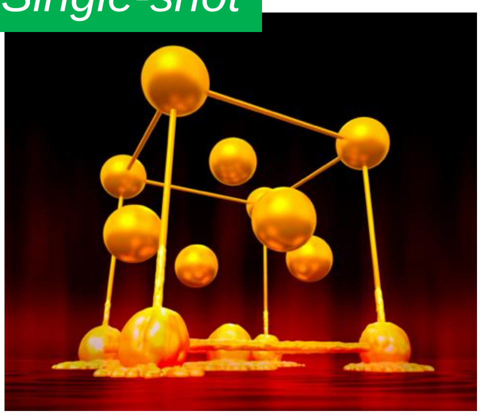
Nature 565, 61 (2019)

Perovskite solar cell



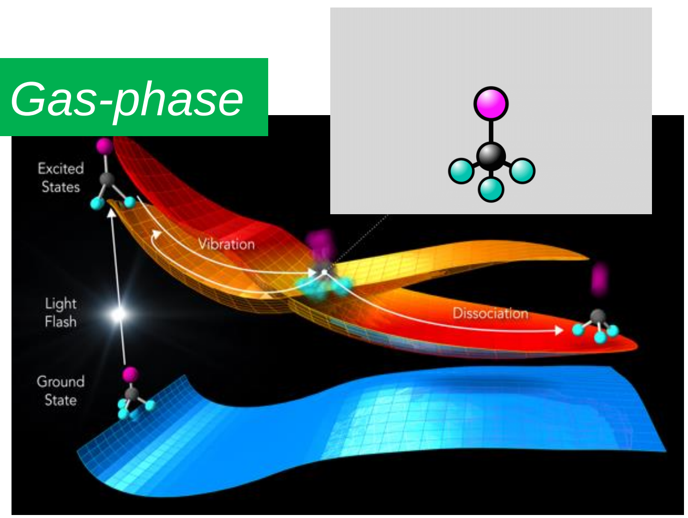
Science Advancve 3, e1602388 (2017)

Single-shot



Science 360, 1451 (2018)

Gas-phase



Science 361, 64 (2018)

30-40 experiments / yr

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

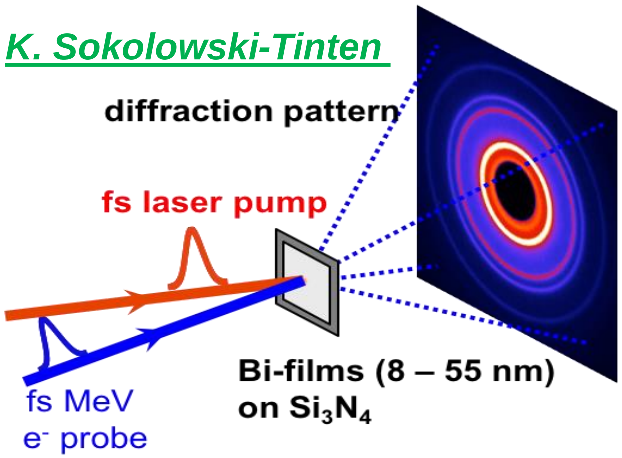
Gas-phase: sequential double-dissociation, roaming reaction, ring opening

Liquid-phase

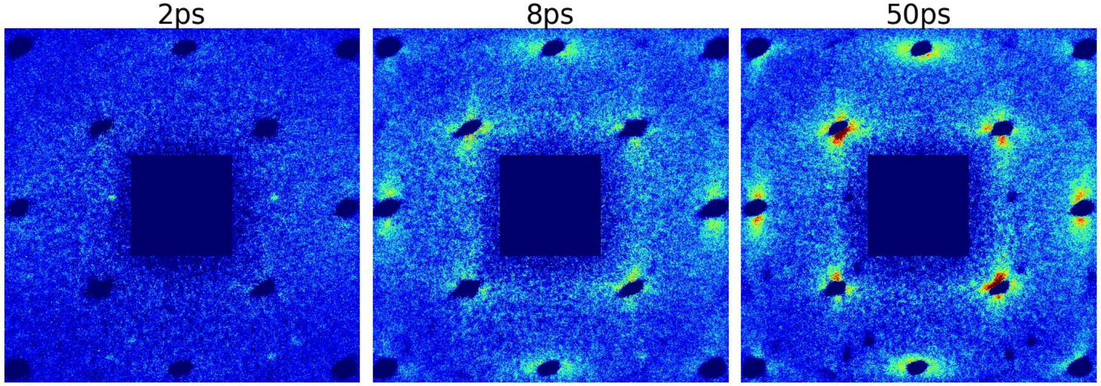
Solid state systems

- ✓ ***Nano-scale materials (Bi, FePt, nanoporous Au, Cr-Cu heterostructures)***
- ✓ ***2D materials (MoS_2, $MoSe_2$, PbI_2)***
- ✓ ***Diffuse scattering (Au, Ni)***
- ✓ ***Strongly correlated system (Bi2212, TaS_2, LSMO)***
- ✓ ***Charge density wave (TaS_2, WTe_2)***
- ✓ ***Functional material (Perovskite, VO_2, high entropy alloys)***
- ✓ ***Warm dense matter (radiation-damaged W, Au)***

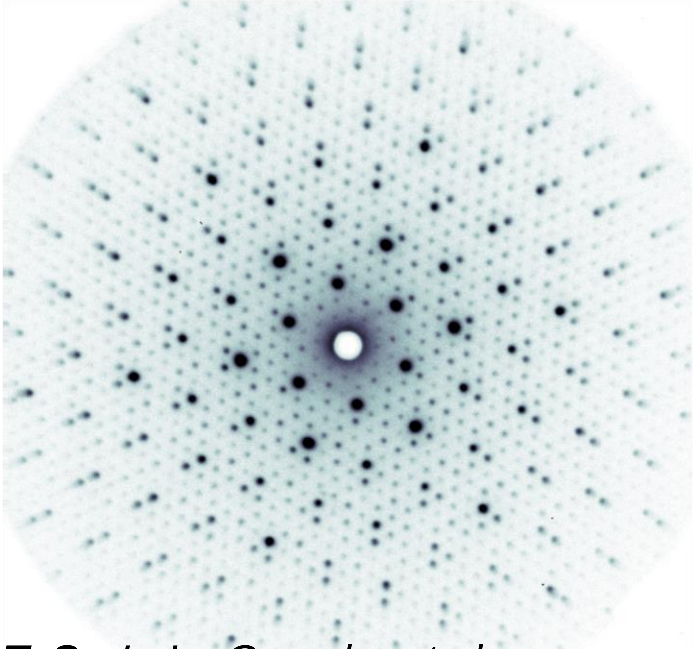
K. Sokolowski-Tinten



Diffuse scattering from single-crystal Au

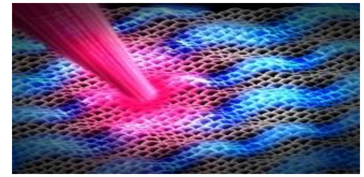


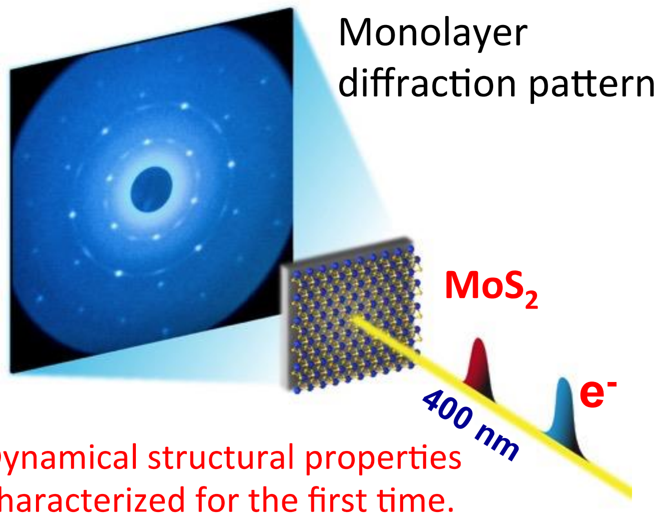
T. Chase et al., APL 108, 041909 (2016)



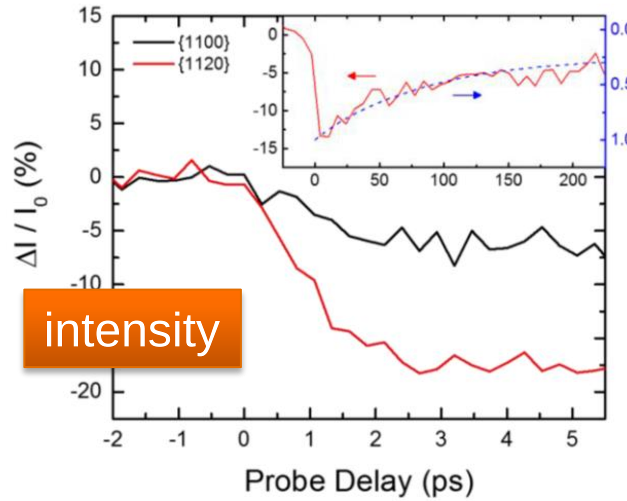
***TaS_2**, L. Le Guyader et al.*

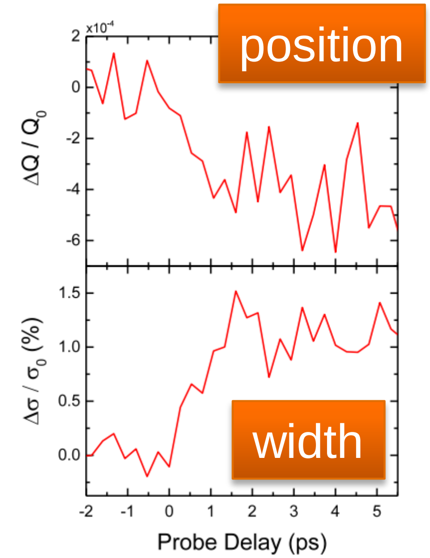
Ultrafast structural deformations in monolayer MoS_2

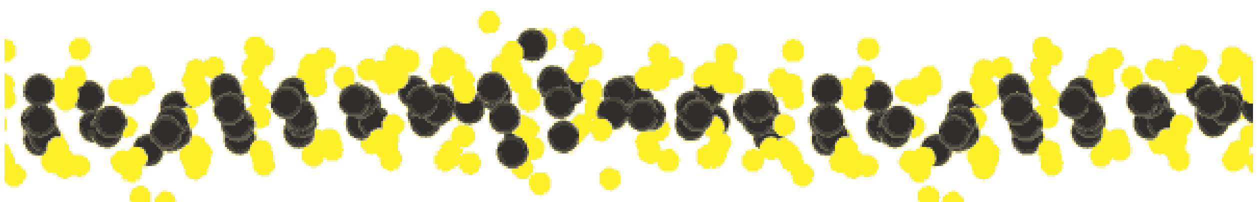




Dynamical structural properties characterized for the first time.



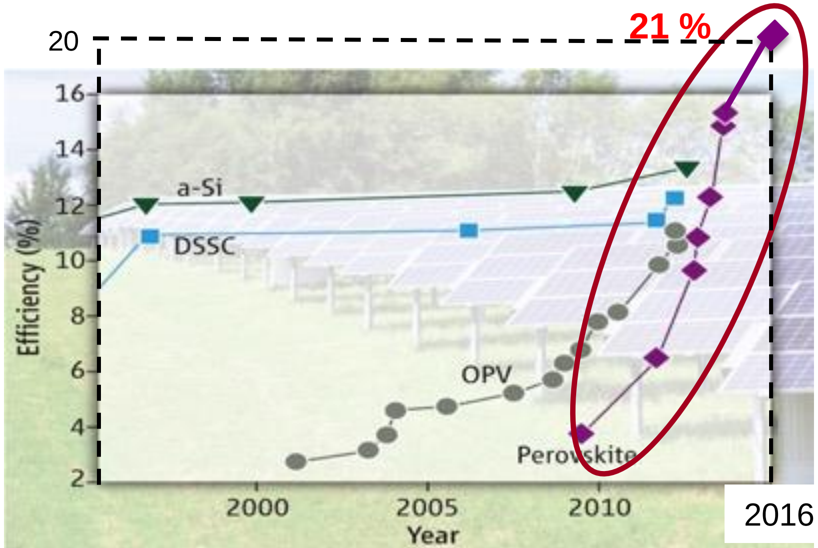


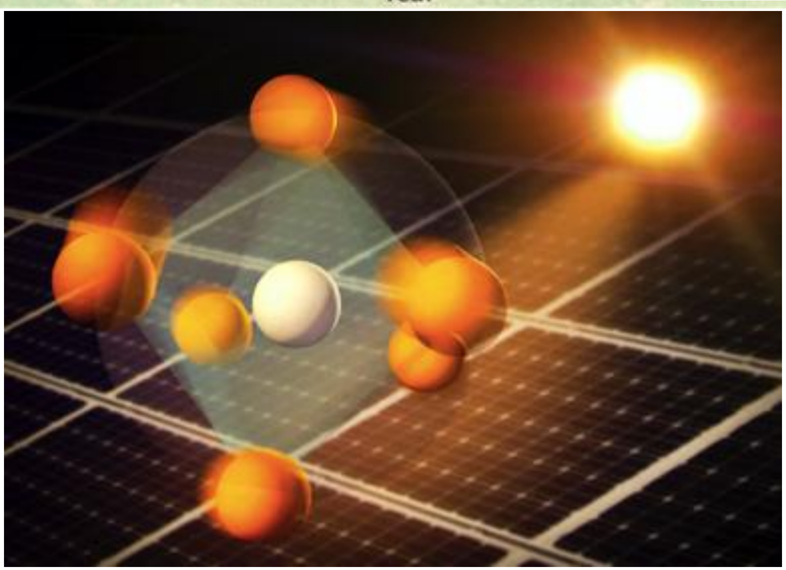


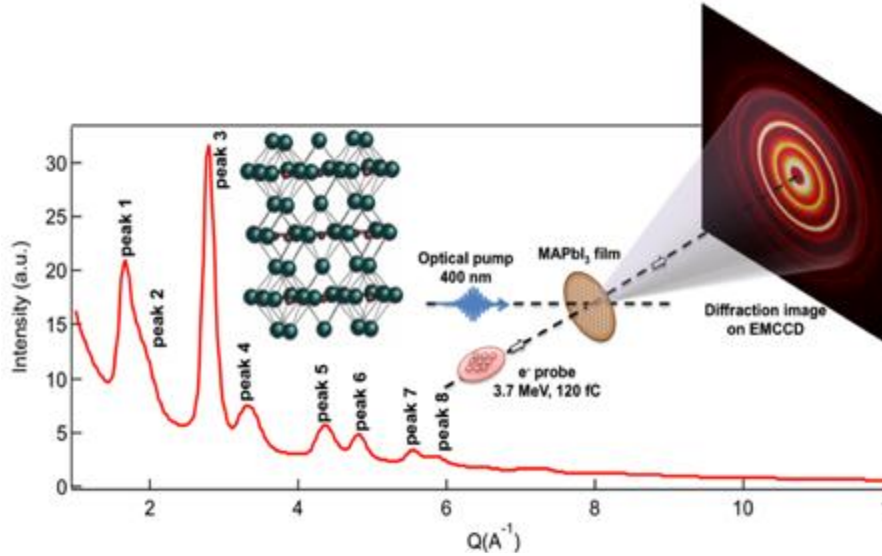
- Large-amplitude in-plane displacements and ultrafast wrinkling of the monolayer
- Time-scale ~1 ps (direct measurements of e-ph coupling time-scales)
- Long-time recovery consistent with interfacial thermal coupling into underlying substrate

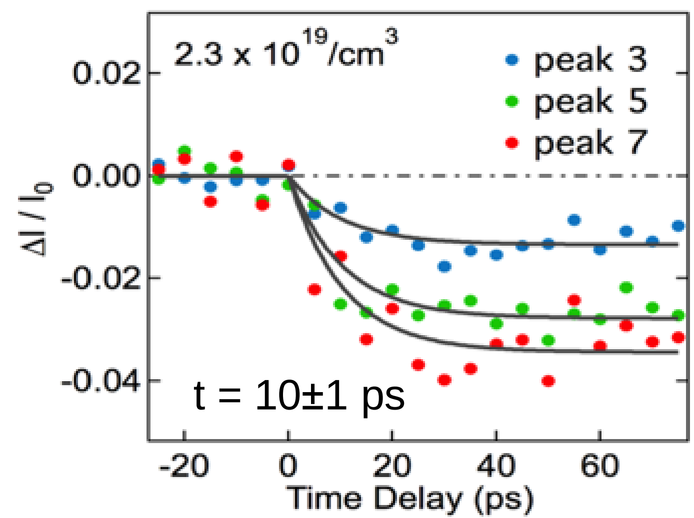
E. M. Mannebach, R. K. Li *et al., Nano Letters* 15, 6889 (2015)

Hybrid perovskites: future of solar cell?



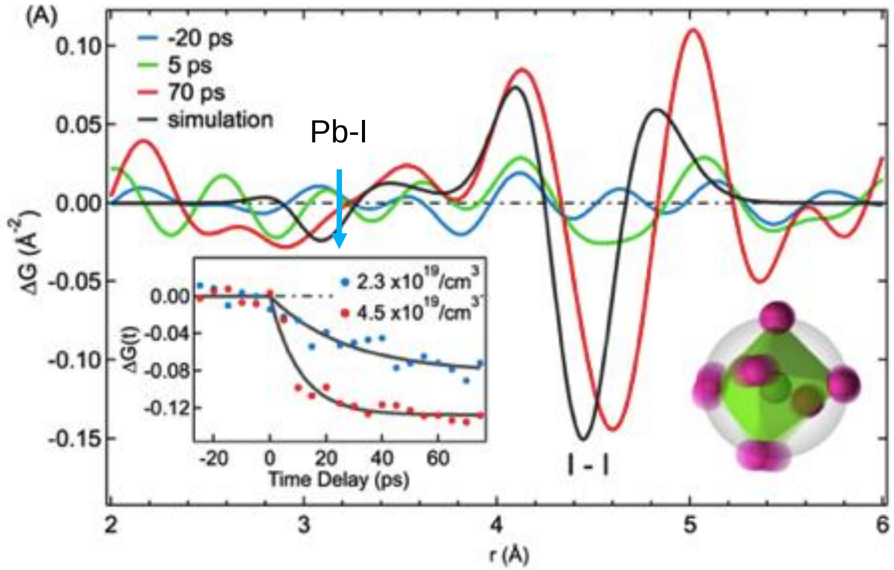


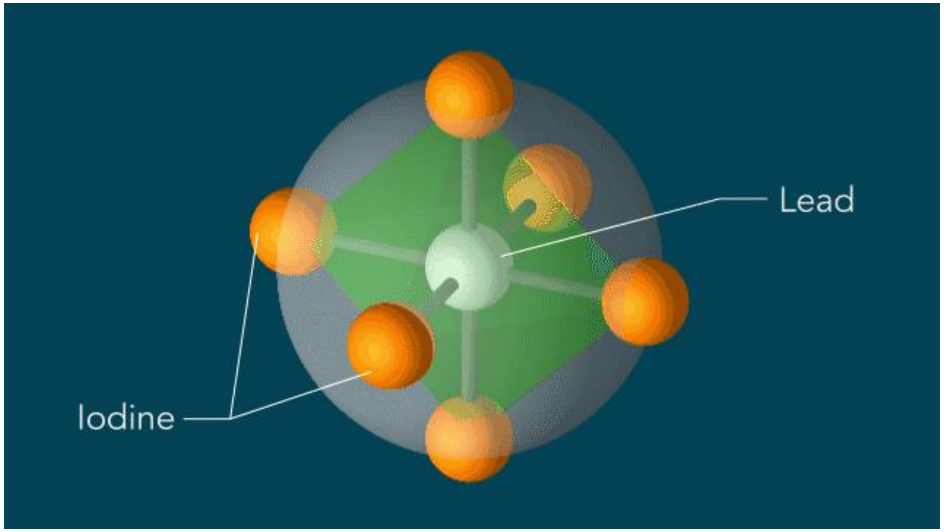




First direct measurements of hot carrier-lattice coupling in the hybrid perovskites

Atomic movies of light-excited perovskite





-Large amplitude lattice displacements observed under weak excitation conditions (~5 K T-jump) – consistent with soft, deformable structure of the hybrid perovskites

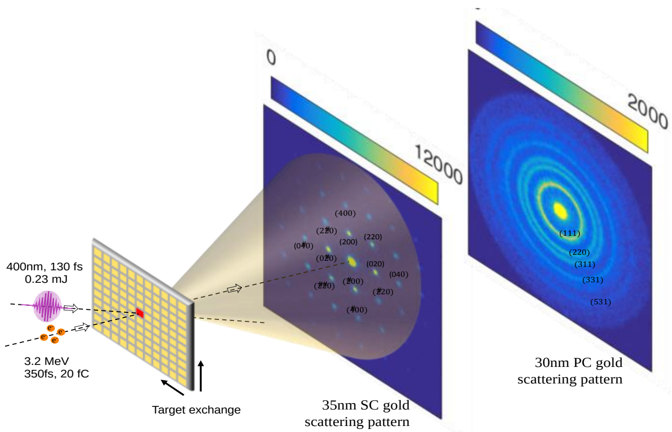
-Differential pair correlation analysis shows that not all the bond lengths are changing. Dominant response is in the iodine octahedra while preserving the Pb-I distance. Indicative of a rotational sub-lattice disordering. First evidence for dynamical response in the inorganic sub-lattice.

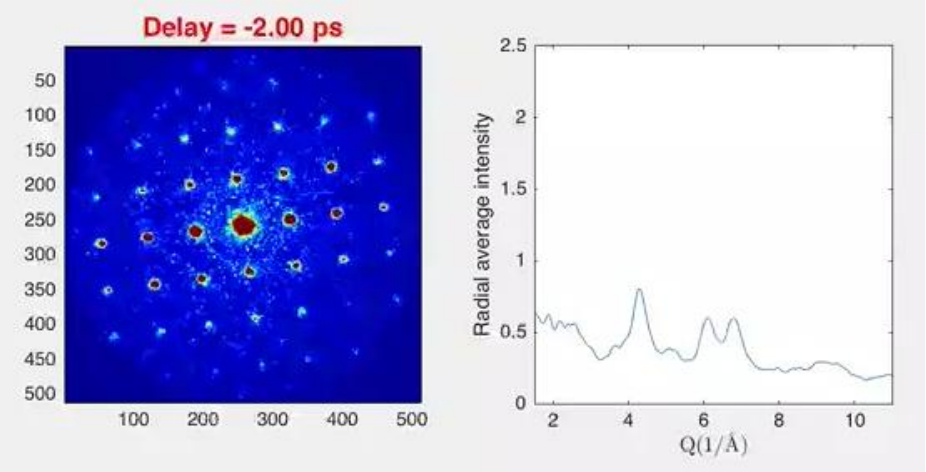
-Dominant contribution from iodines consistent with their role in larger length-scale ionic transport / degradation of PV devices.

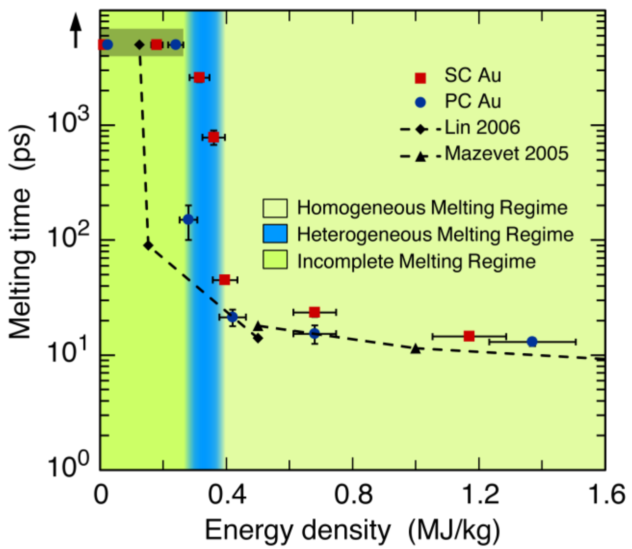
X. Wu et al. Sci. Adv. 3 e1602388 (2017)

Heterogeneous to homogeneous melting transition visualized with UED

M. Z. Mo et al., **Science** 360, 1451 (2018)







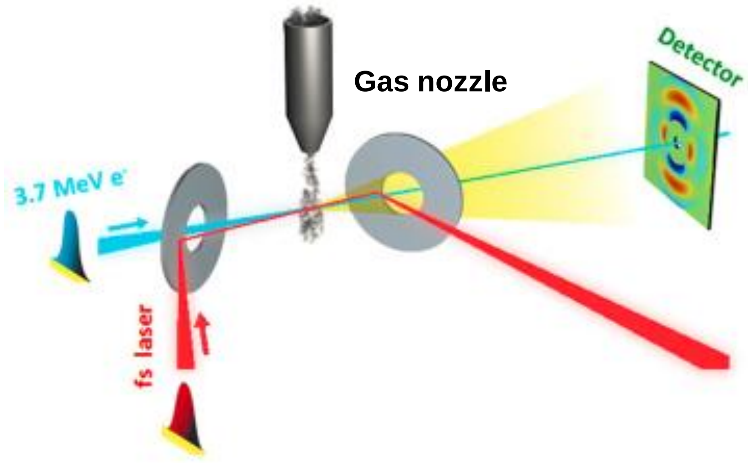
Measured energy density dependence of ultrafast-laser-induced melting mechanisms in Au.

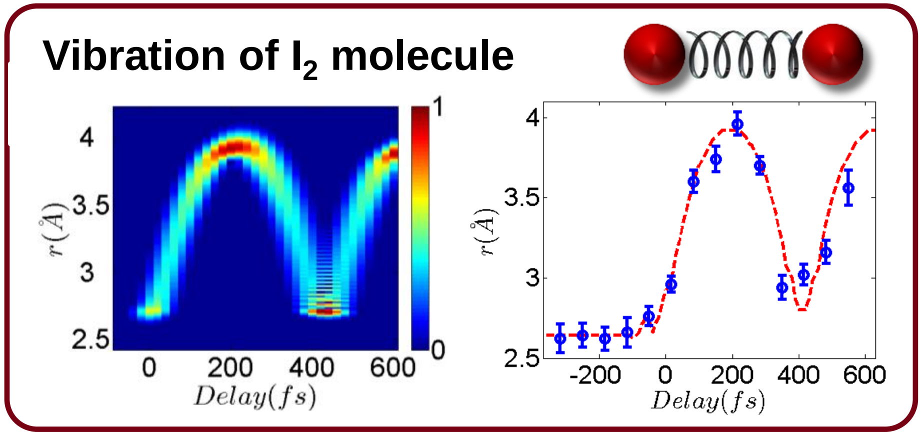
Scientific significance:

- enabled direct comparison with MD simulations and revealed the sensitivity to nucleation seeds for melting.
- Provide critical information to test and improve the kinetic theories of melting.
- Help advance the material processing related to solid-liquid phase transition to atomic level precision.

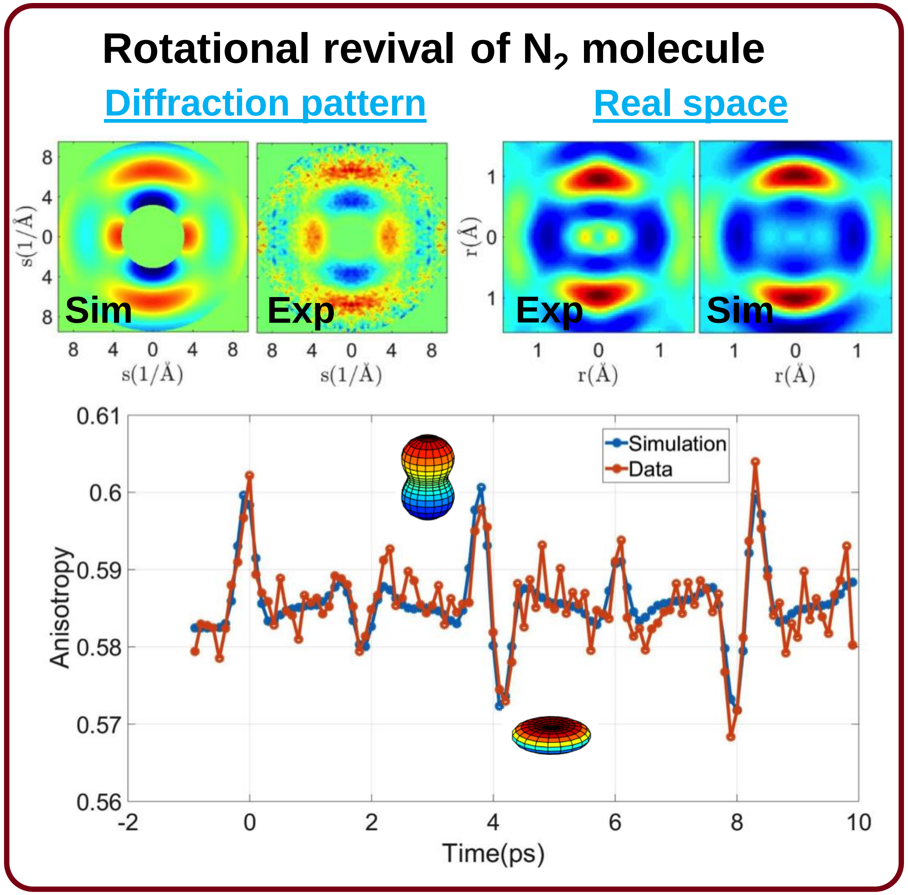
Gas-Phase MeV UED (Year 2015)

- First gas-phase UED with 100-fs temporal resolution





J. Yang et al., PRL 117, 153002 (2016)

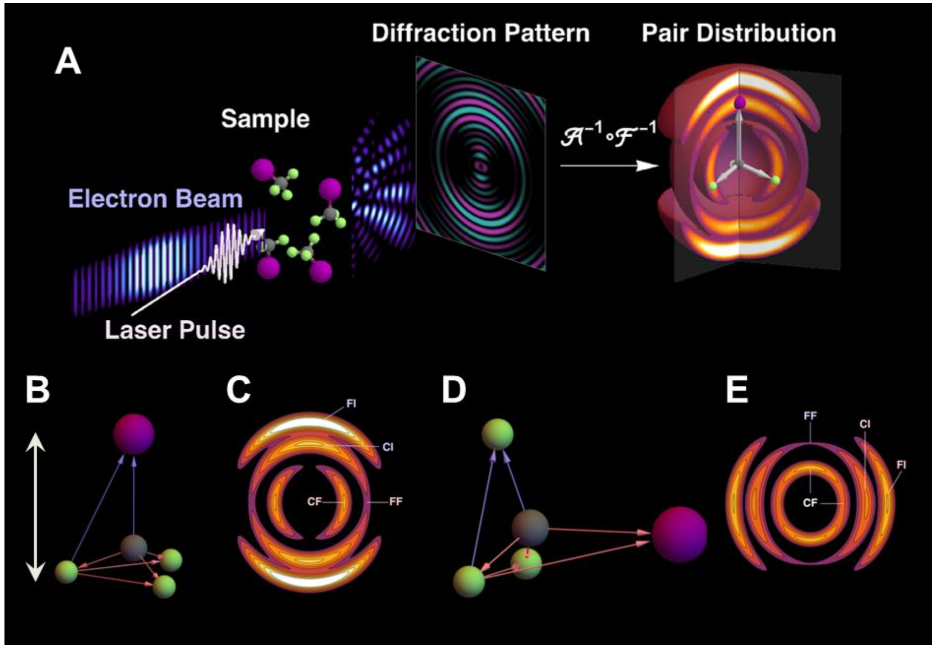


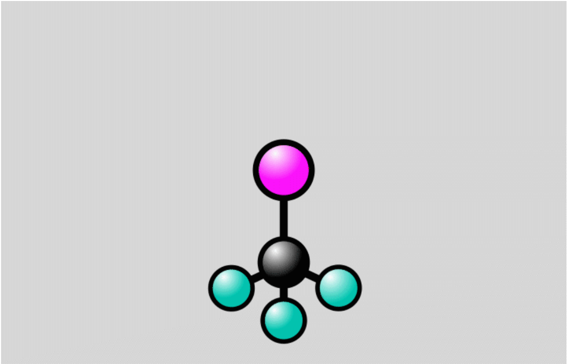
J. Yang et al., Nature Commun. 7, 11232 (2016)

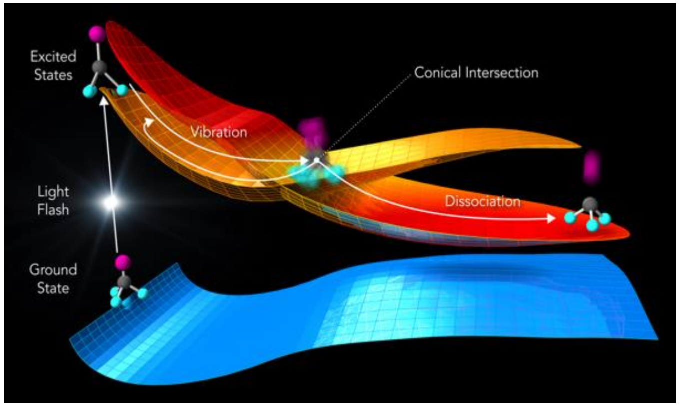
Conical intersection and photodissociation dynamics

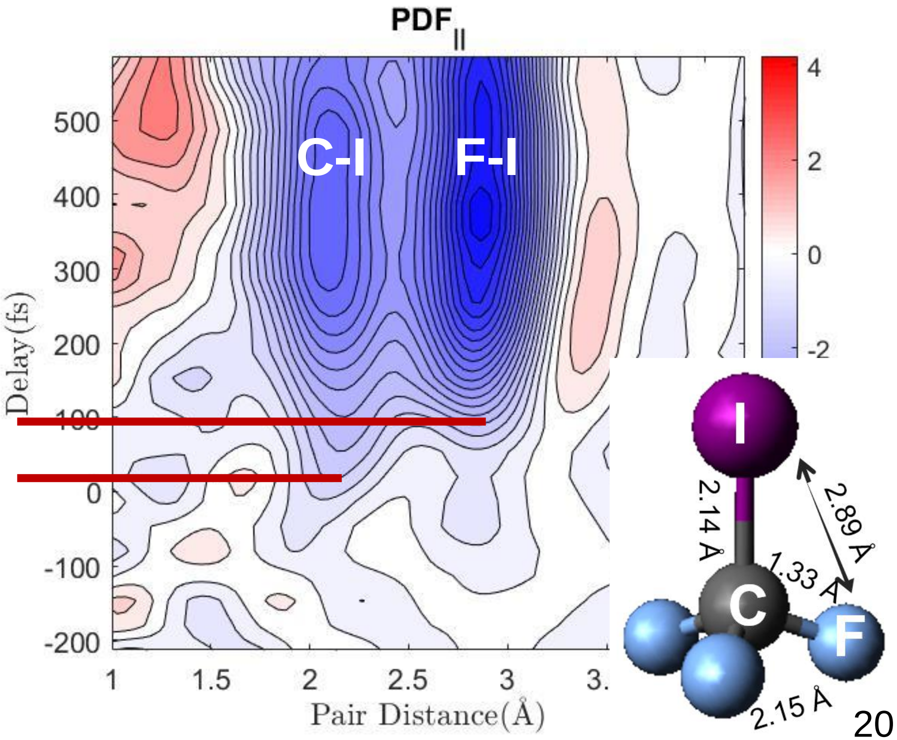
J. Yang et al., **Science** 361, 64 (2018)

CF_3I



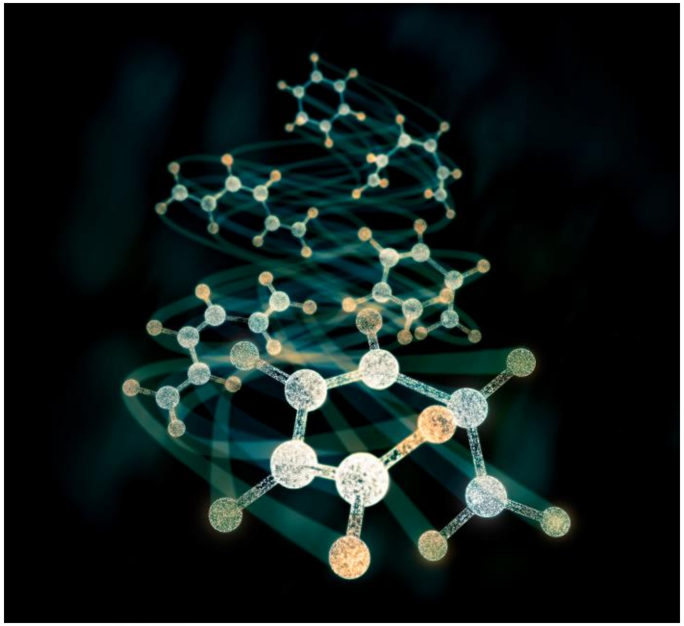




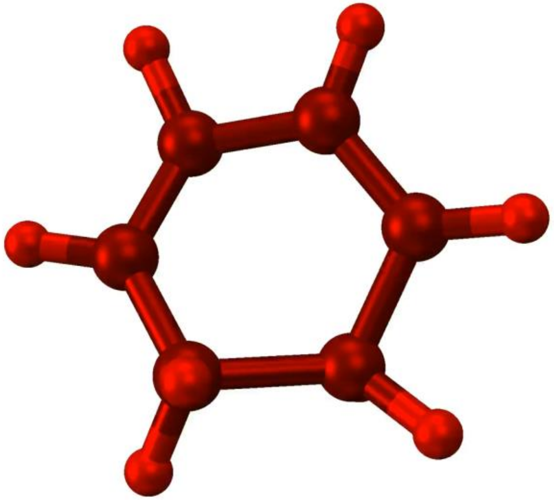


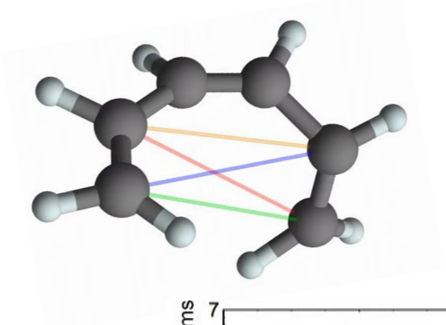
Ring-opening reaction of CHD

- Same reaction has been measured by LCLS
- LCLS has better temporal resolution
- UED has better spatial resolution
- Much shorter wavelength of electrons (signal at large *q*) allows 'direct' interpretation of the structure
- Extremely valuable dataset to benchmark simulation codes



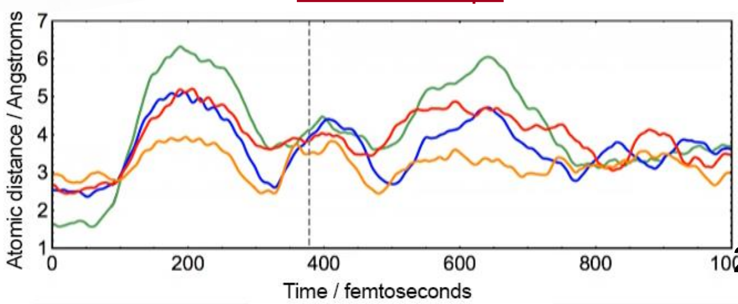
1,3-cyclohexadiene (CHD) to 1,3,5-hexatriene (HT)



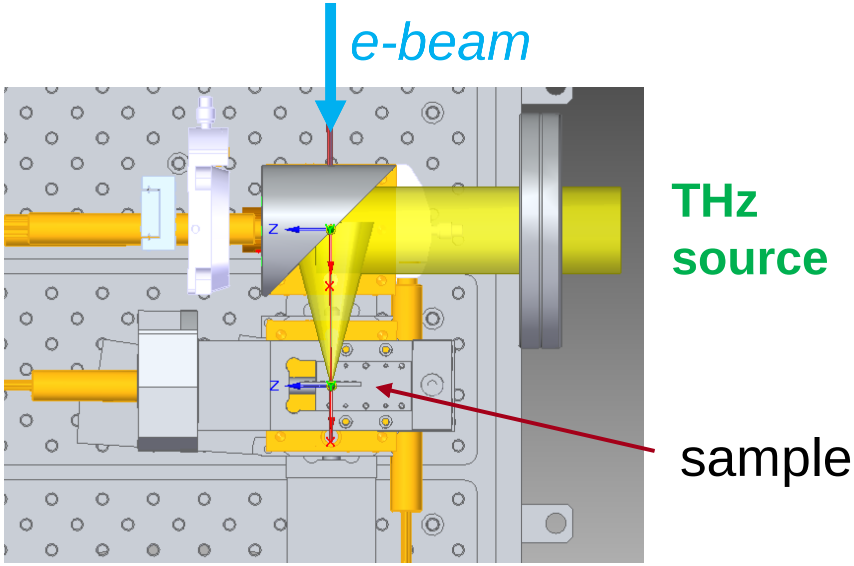


T. J. A. Wolf et al., Nature Chemistry 11, 504 (2019).

https://www6.slac.stanford.edu/news/2019-04-15-slacs-high-speed-electron-camera-films-molecular-movie-hd.aspx

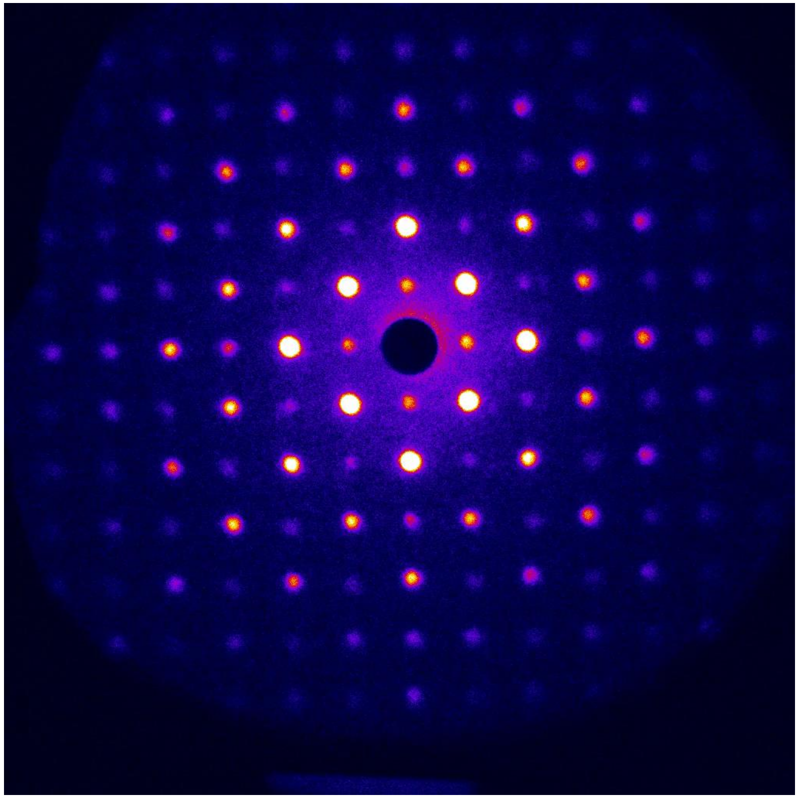


THz pump – electron probe



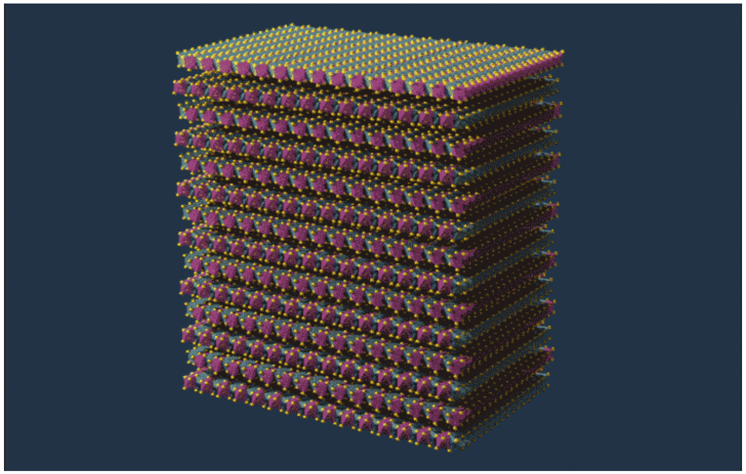
THz scheme	Frequency range	Field strength/Fluence
Optical rectification in $LiNbO_3$ using titled pulse front technique	• 0.8 THz Peak frequency • single cycle pulse	• >10 uJ/pulse (~300 kV/cm)
Rectification in an Organic Crystal	• 2 THz Peak frequency • single cycle pulse	• >5 uJ/pulse (~600 KV/cm)
Difference frequency generation OPA	• Tunable 17-30 THz • 50 fs duration	• >10 uJ/pulse (25 mJ/cm^2)

THz induced, time-dependent kick of MeV electrons



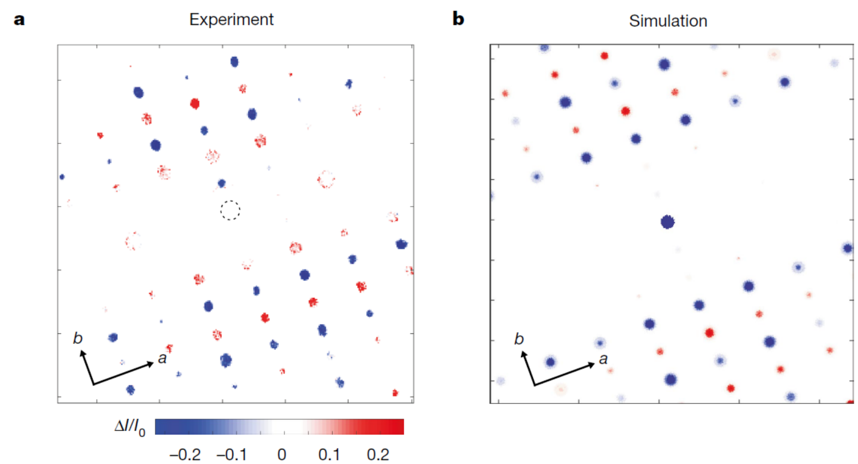
Courtesy of E. Mannebach

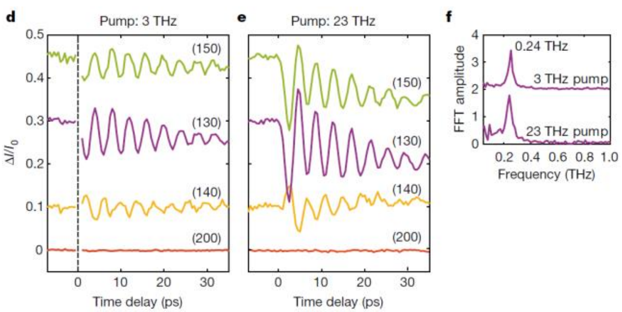
ultrafast symmetry switch in a Weyl semimetal

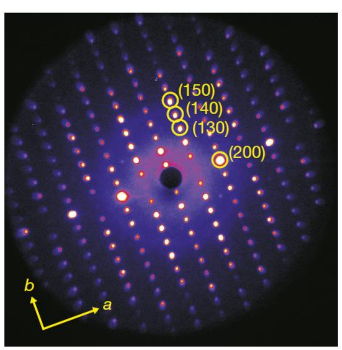


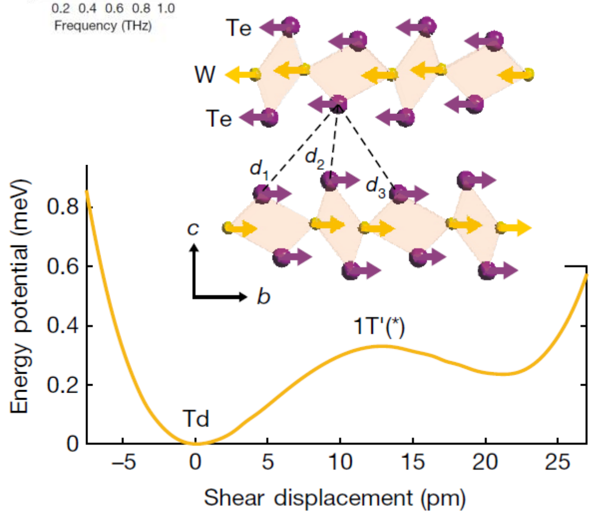
- THz pump, UED probe
- THz-driven shear strain as a mechanism for switching the topology of materials in WTe_2

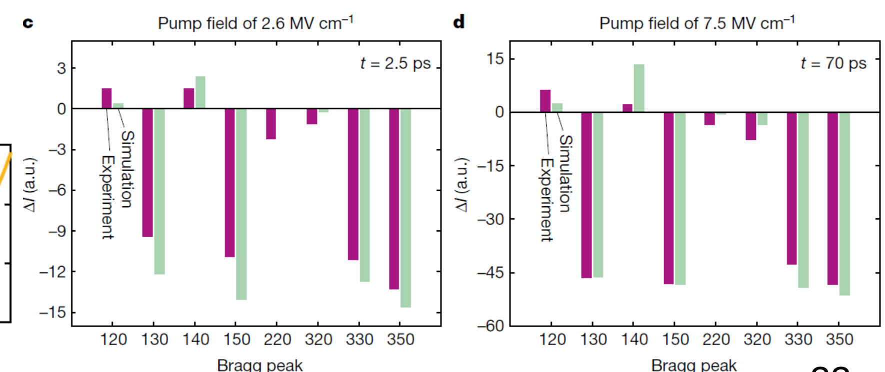
E. J. Sie, C. M. Nyby et al, Nature 565, 61 (2019)







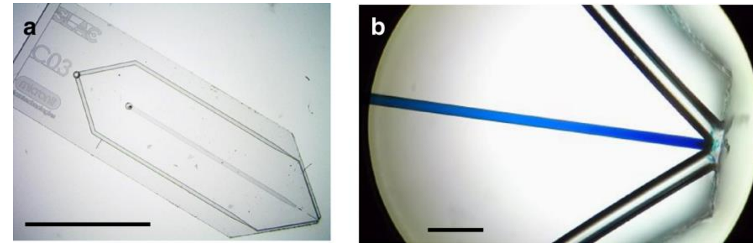


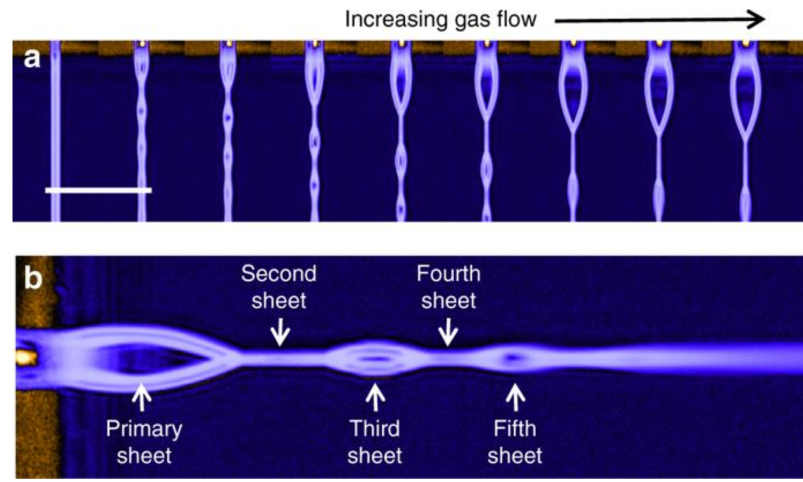


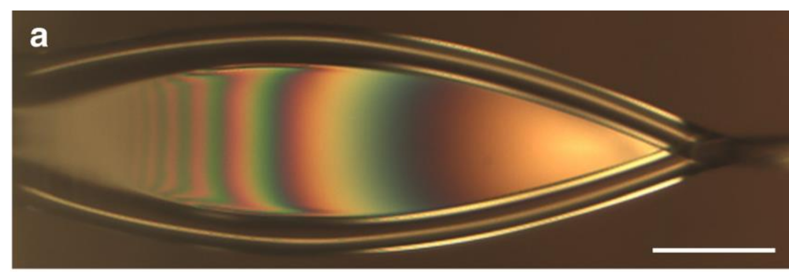
Liquid phase samples

- Many **chemical** and **biological** processes happen in liquid/solution phase
- Liquid sheets jet developed at LCLS (100s nm thick, 100s um wide) suits well UED
- First round experiment April-June 2019

Liquid phase sample chamber under testing at SLAC UED (contact: Xijie Wang)







J. Koralek et a., Nat. Commun. 9, 1353 (2018)

Outline

- Introduction
- State-of-the-art MeV UED based on an rf gun
- Recent scientific outcomes
- Toward better performances (spatial, temporal)
- Summary and outlook

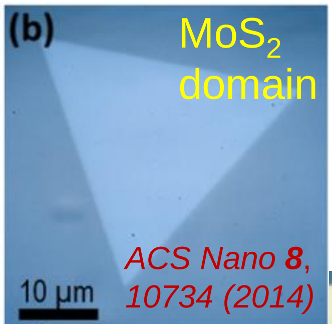
Toward better probe size and q-resolution

$$\epsilon_n = \sigma_x \cdot \gamma\beta\sigma_{x'}$$

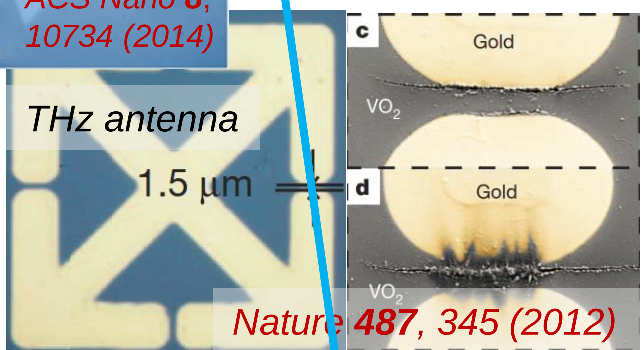
e.g. 0.1 nm-rad = 0.4 um × 0.25 mrad

$$\Delta q = \frac{2\pi}{\lambda_e} \cdot \gamma\beta\sigma_{x'}$$

e.g. $\Delta q = 0.07$ Å^{-1} w/ $\gamma\sigma_{x'}$ = 0.25 mrad

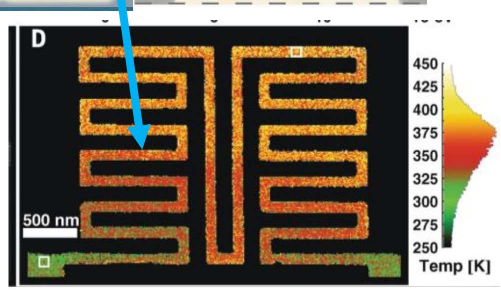


ACS Nano ***8***, *10734 (2014)*

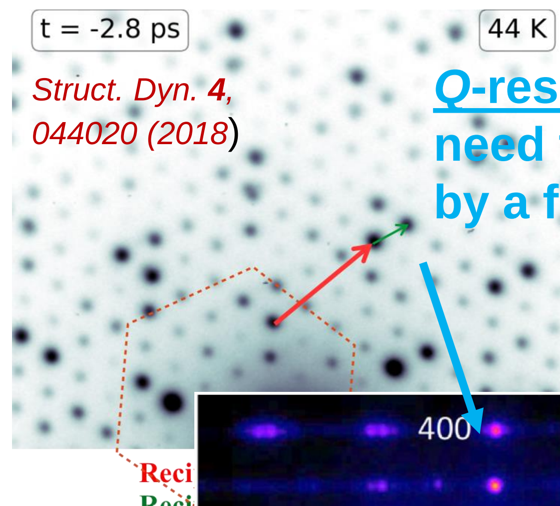


Nature ***487***, *345 (2012)*

Probe size:
10s of um to um to nm

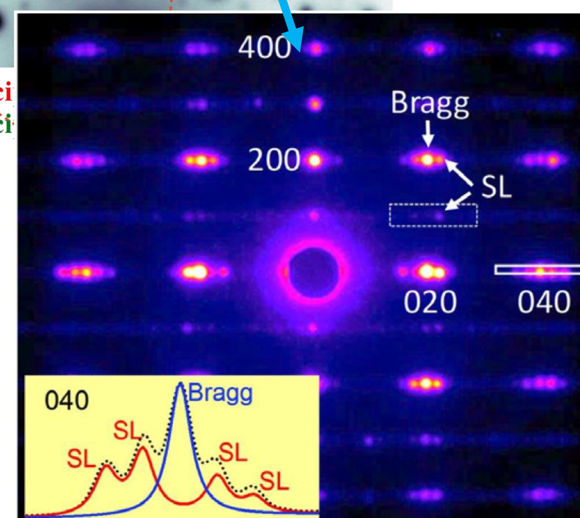


Science ***347***, *629 (2015)*



Struct. Dyn. ***4***, *044020 (2018*)

Q-resolution:
need to improve by a few times

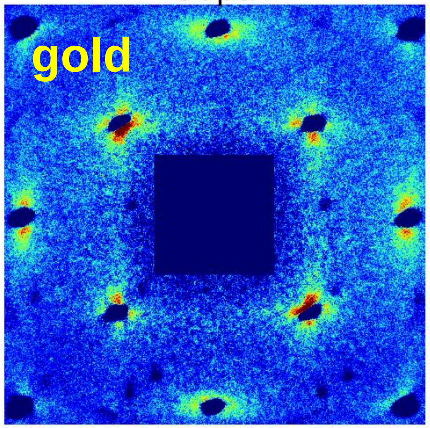


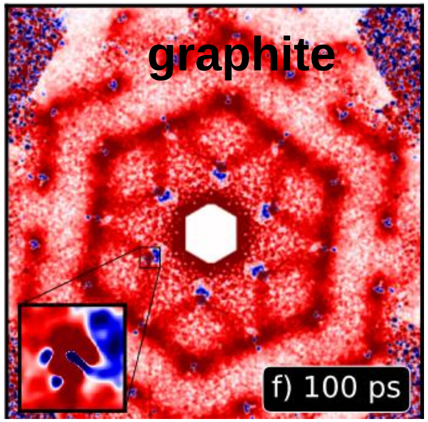
Sci. adv. ***4***, *eaap7427 (2018)*

Better temporal resolution

- Currently best UED temporal resolution ~100 fs fwhm (~50 fs rms)
- ~10 fs level temporal resolution will open up many new opportunities
- Taking advantage of the full optical control to minimize time-of-flight jitter

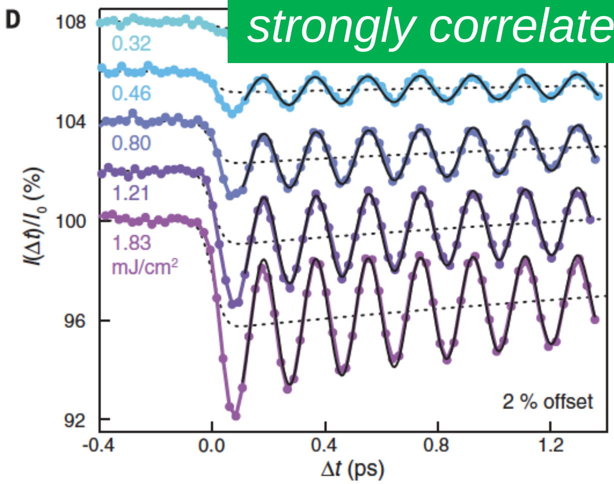
50ps





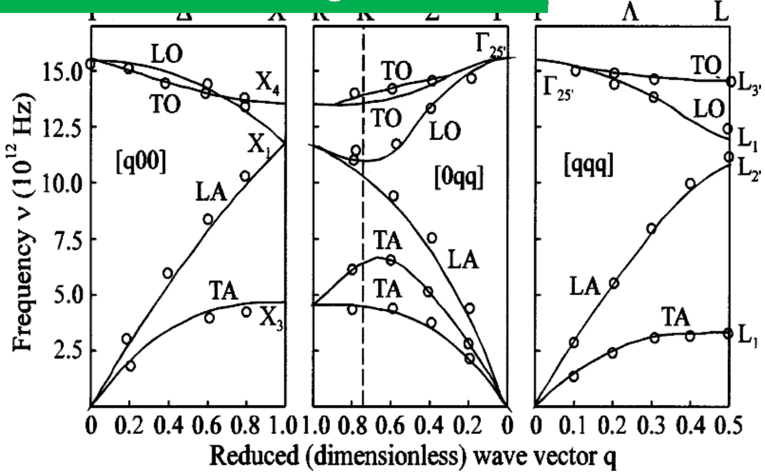
APL 108, 041909 (2016) *PRB 97, 165416 (2018)*

Optical phonon in strongly correlated systems



Gerber et al., Science 357, 71 (2017)

Phonon spectroscopy via diffuse scattering

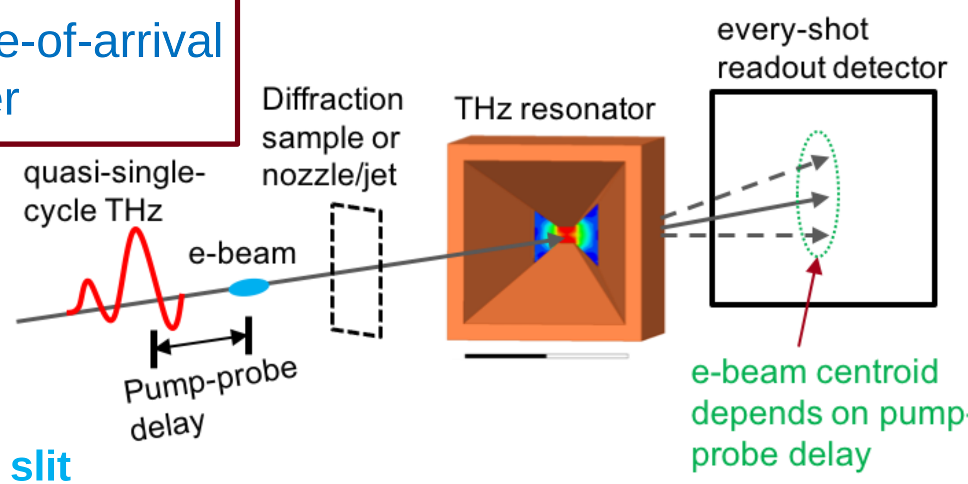


THz stamping to sub-fs accuracy

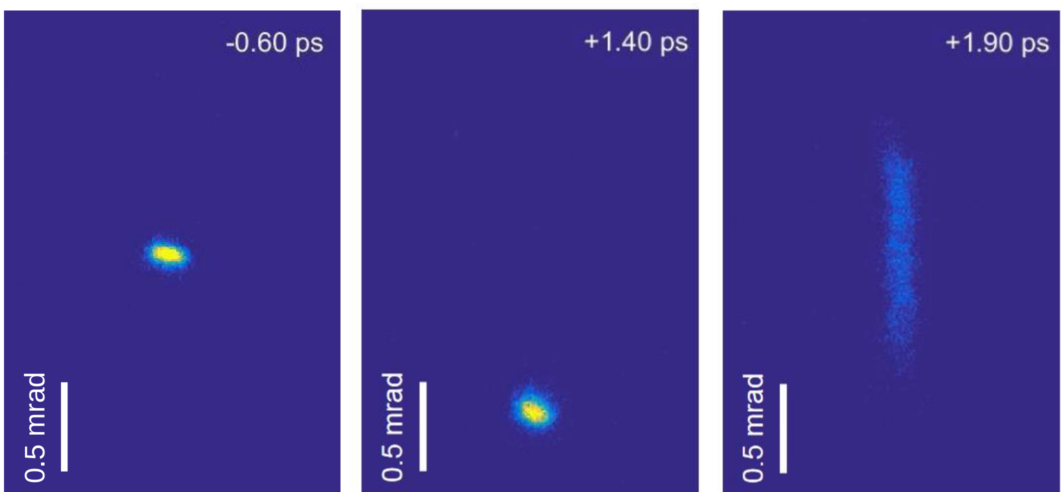
$$\tau^2 = \tau_{\rm ph}^2 + \tau_{\rm e}^2 + \tau_{\rm vm}^2 + \tau_{\rm toa}^2$$

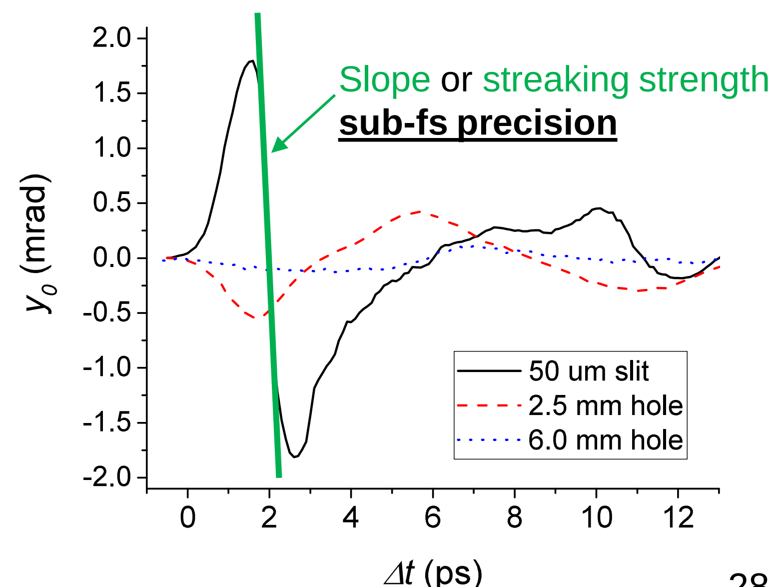
temporal resolution | laser | e-beam | velocity mismatch | time-of-arrival jitter

- characterize e-bunch length
- develop a fs timing tool for UED



3.5 MeV electrons, 50 um gap, 100 um thick slit

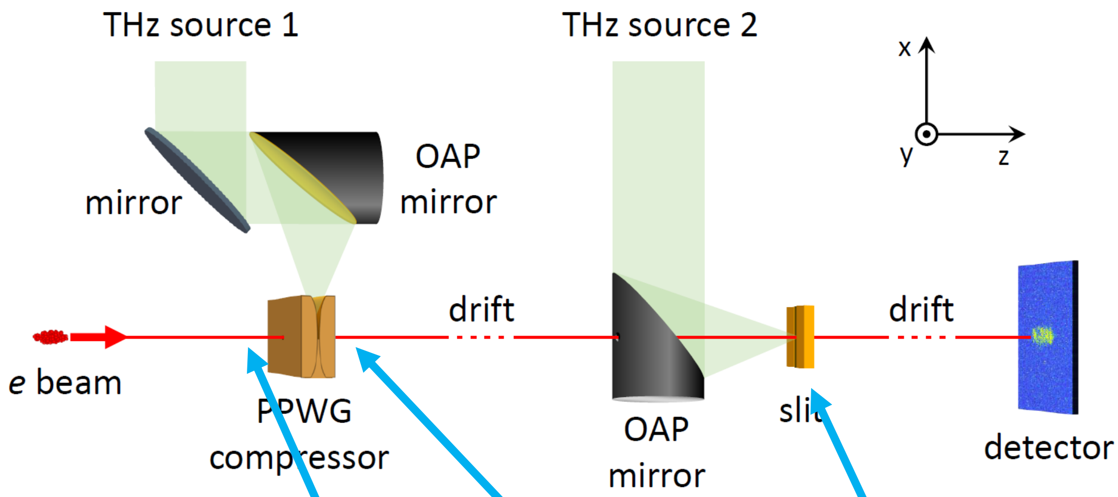


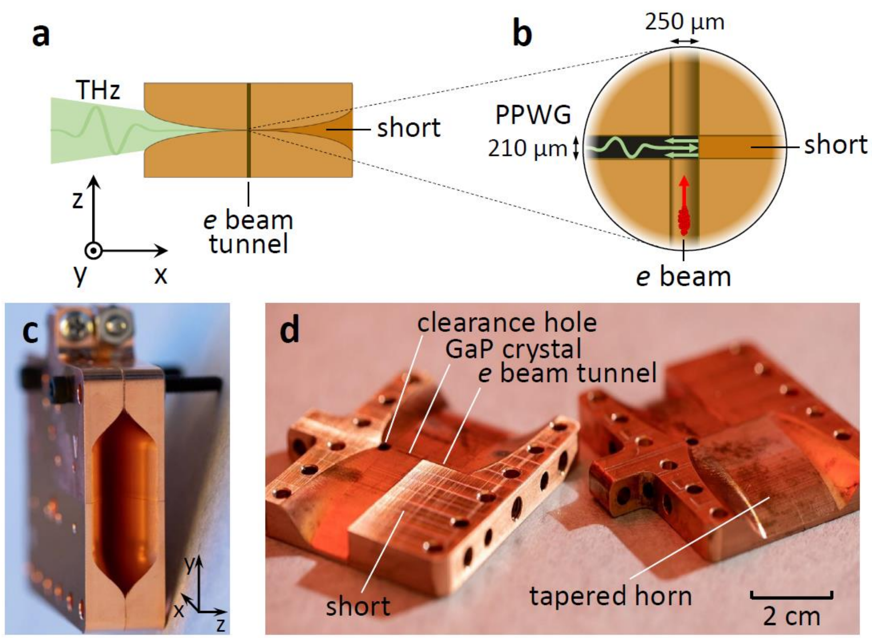


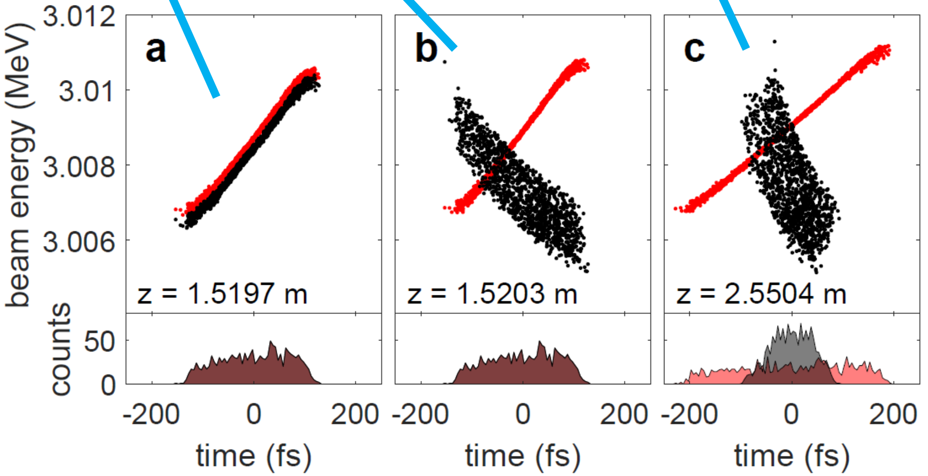
Li, Hoffmann, Nanni et al., PRAB 22, 012803 (2019).

THz compression and jitter suppression

- Laser generated THz can compress the bunch length
- And, stabilize the timing jitter at the same time, relative to the pump laser



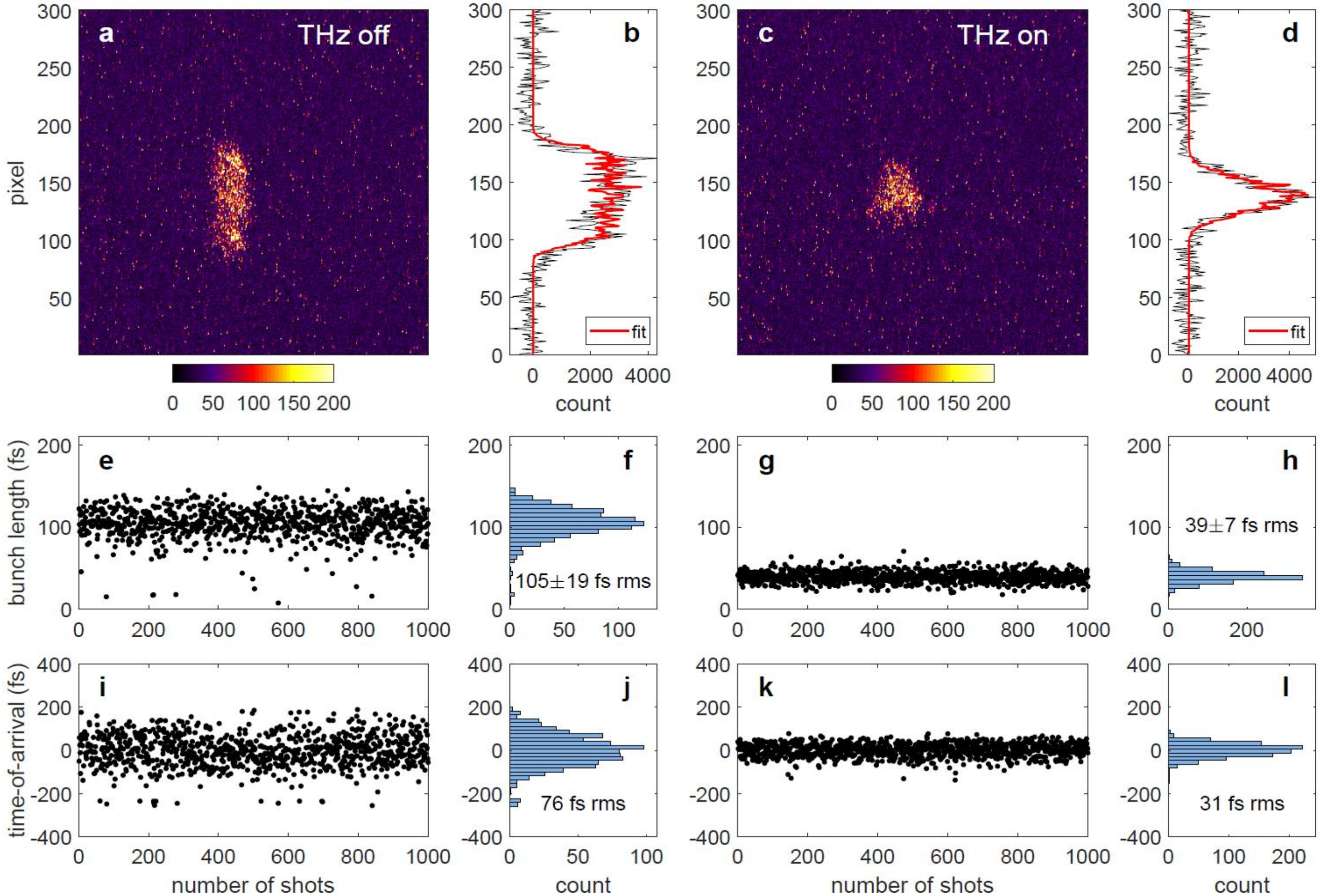




E. Snively et al., arXiv: 1906.03358

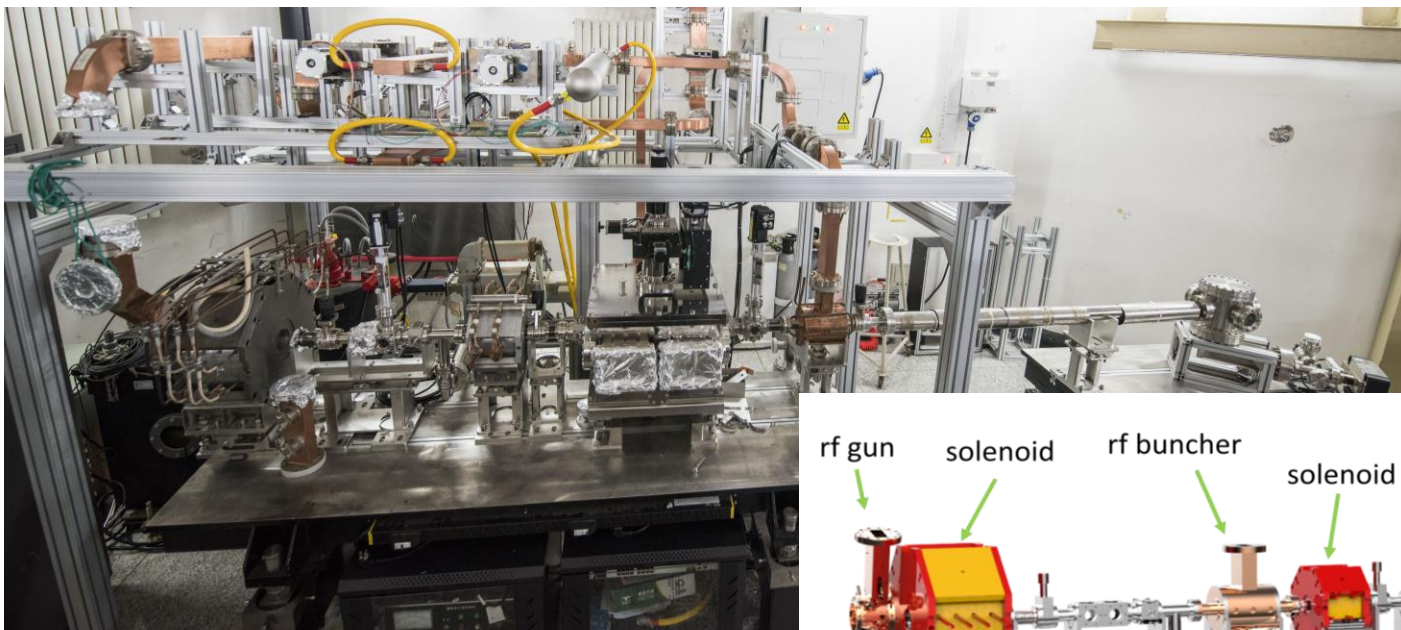
Improved temporal resolution with THz

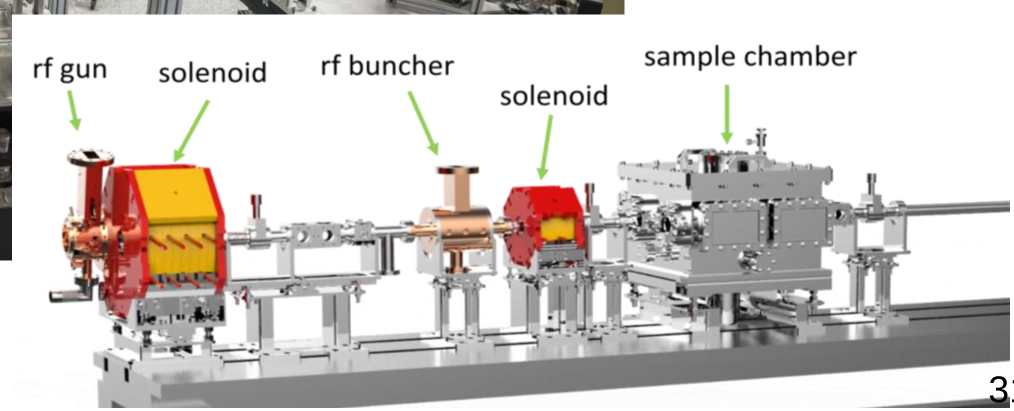
	THz off		THz on
Bunch length	105±19 fs	→	39±7 fs
Timing jitter	76 fs	→	31 fs



Reviving an MeV UED beamline at Tsinghua

- Most hardware in place
- Adding an rf buncher for much higher bunch charge
- Work with material science, chemistry, biology, quantum materials groups on campus and worldwide





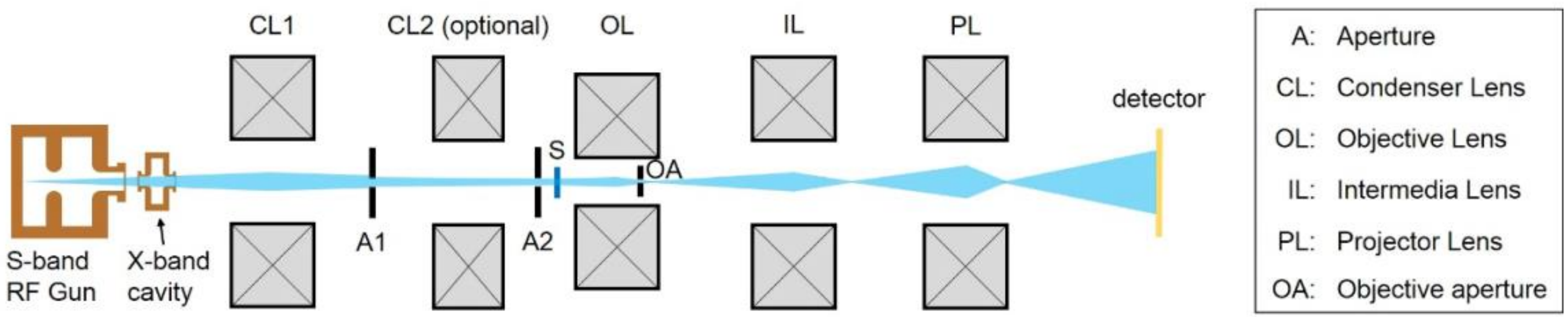
Summary

- MeV UED is a unique and versatile research tool
  - Complementary to XFEL, ARPES, optical spectroscopy, etc.
- Powerful, reliable, cost-effective
  - 24/7 operation, mid-scale and requires good lab environment
- Intensive R&D toward better spatial (*q*-) and temporal resolution
- Laser-generated THz can further improve performances

Thank you for your attention!

Single-shot ps MeV UEM

MeV UEM: 10^3 times improved spatio-temporal resolution



R. K. Li and P. Musumeci, PRApplied 2, 024003 (2014)

- Improved source: higher launch field, higher brightness
- lower beam emittance
- rf amplitude and phase control (at 1×10^{-4} and 0.01 degree level)
- Low energy spread: longi. phase space linearization
- strong electron lens
- numerical studies of aberrations and *e*-*e* interactions