Ultrafast Electron Diffraction
present status & future advances

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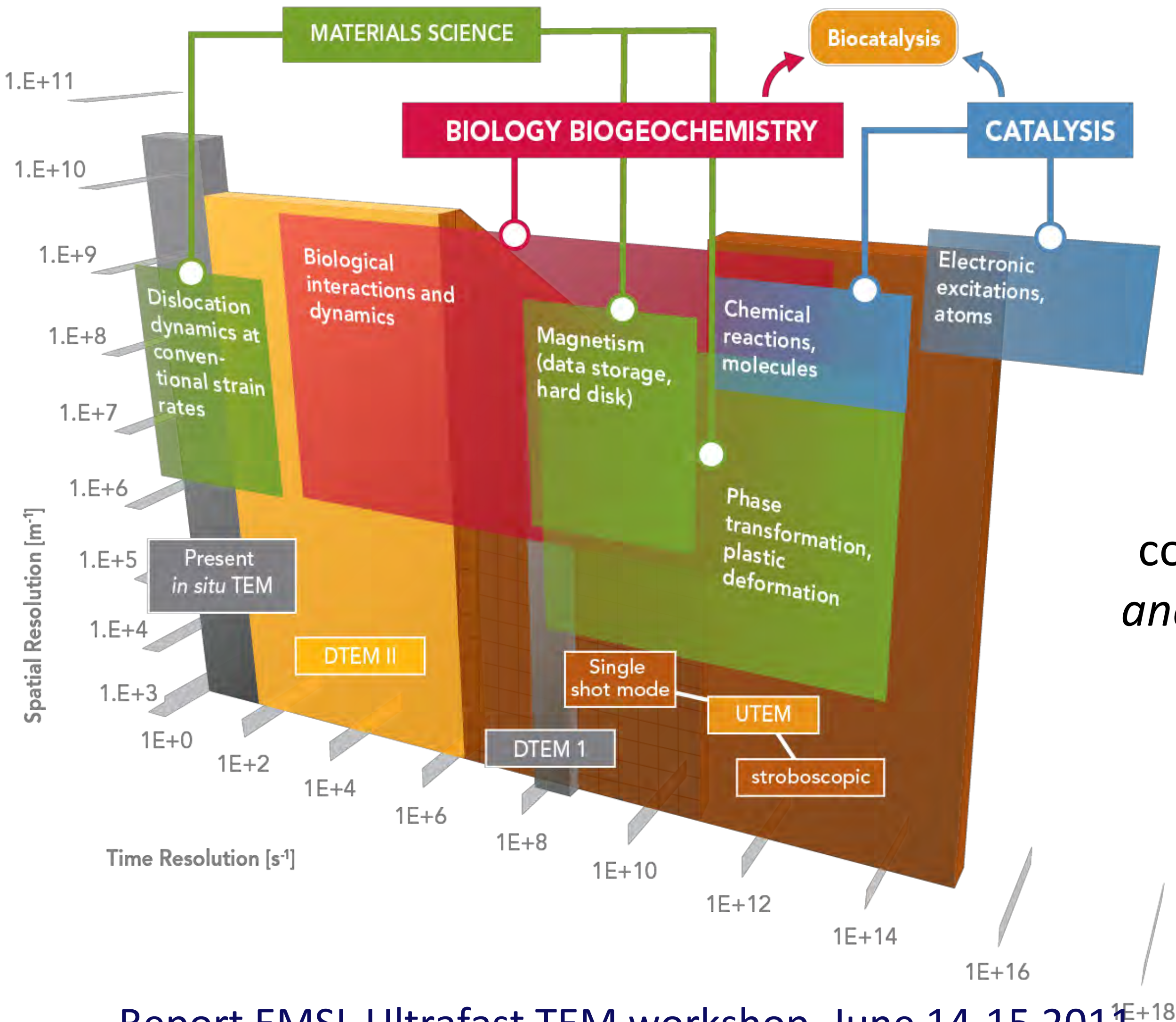
IPAC '15, April 7th 2015

Ultrafast structural dynamics

TU/e

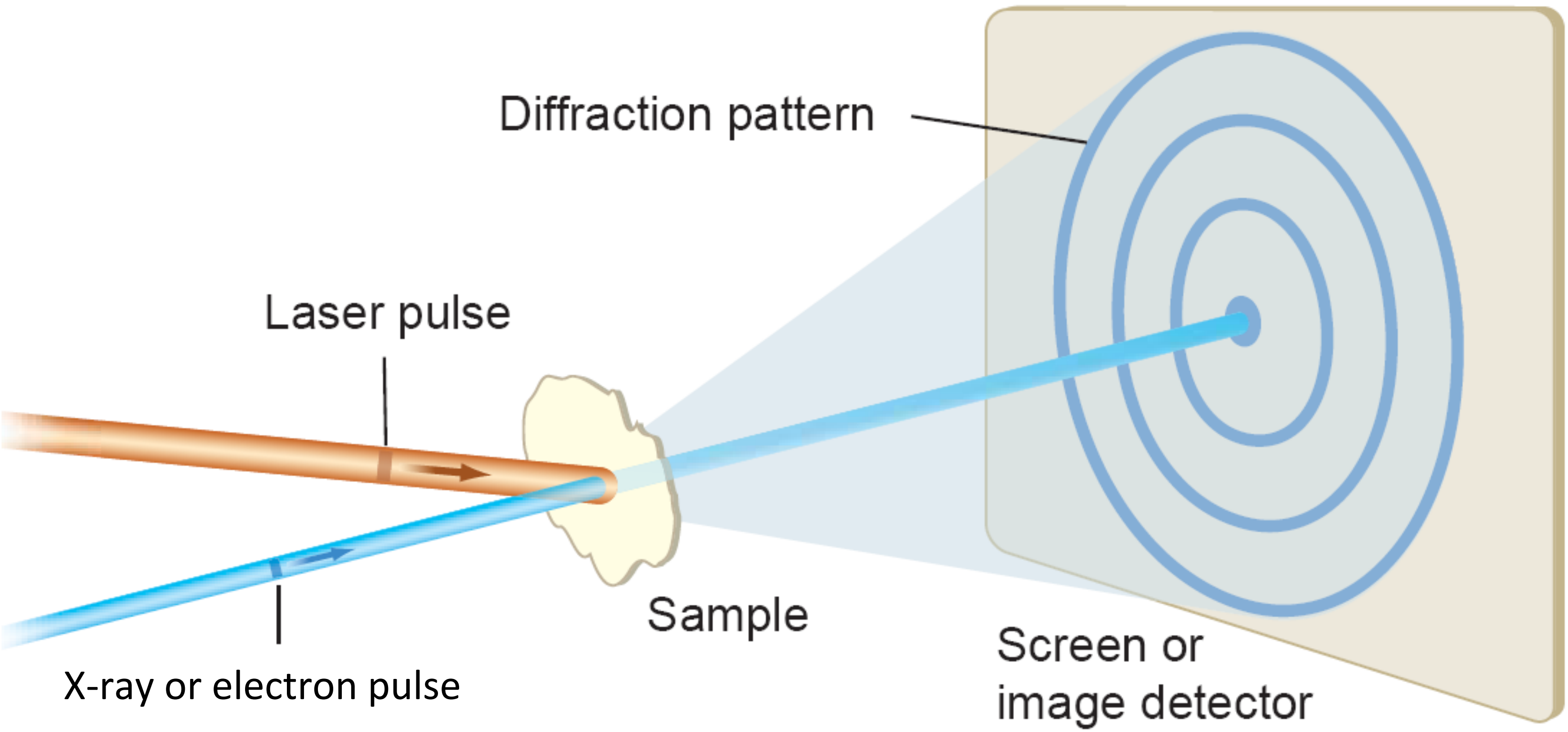


The challenge:
combine atomic spatial *and* temporal resolution, 0.1 nm *and* 0.1 ps

Report EMSL Ultrafast TEM workshop, June 14-15 2011

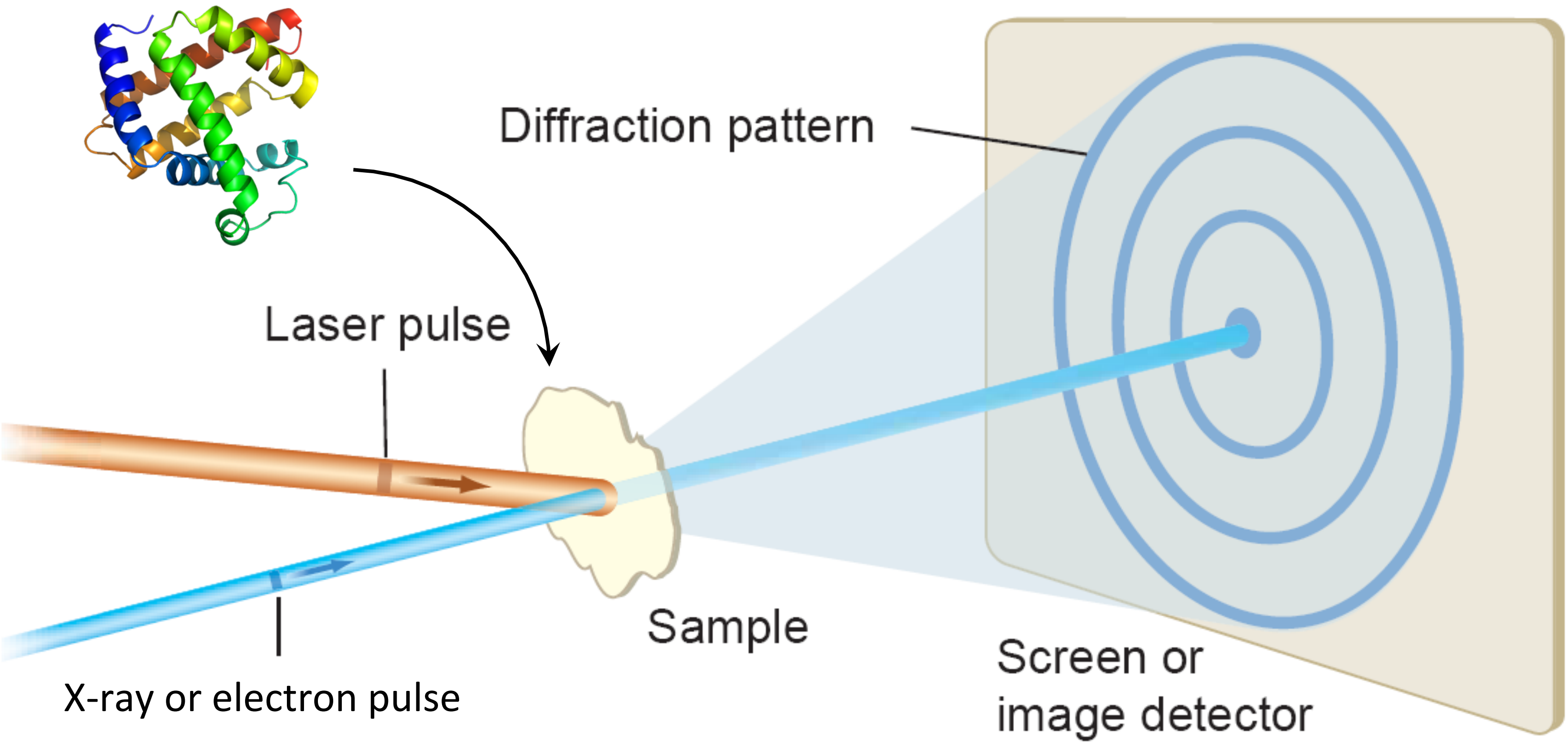
Pump-probe method

TU/e



ultrafast diffraction

Pump-probe method

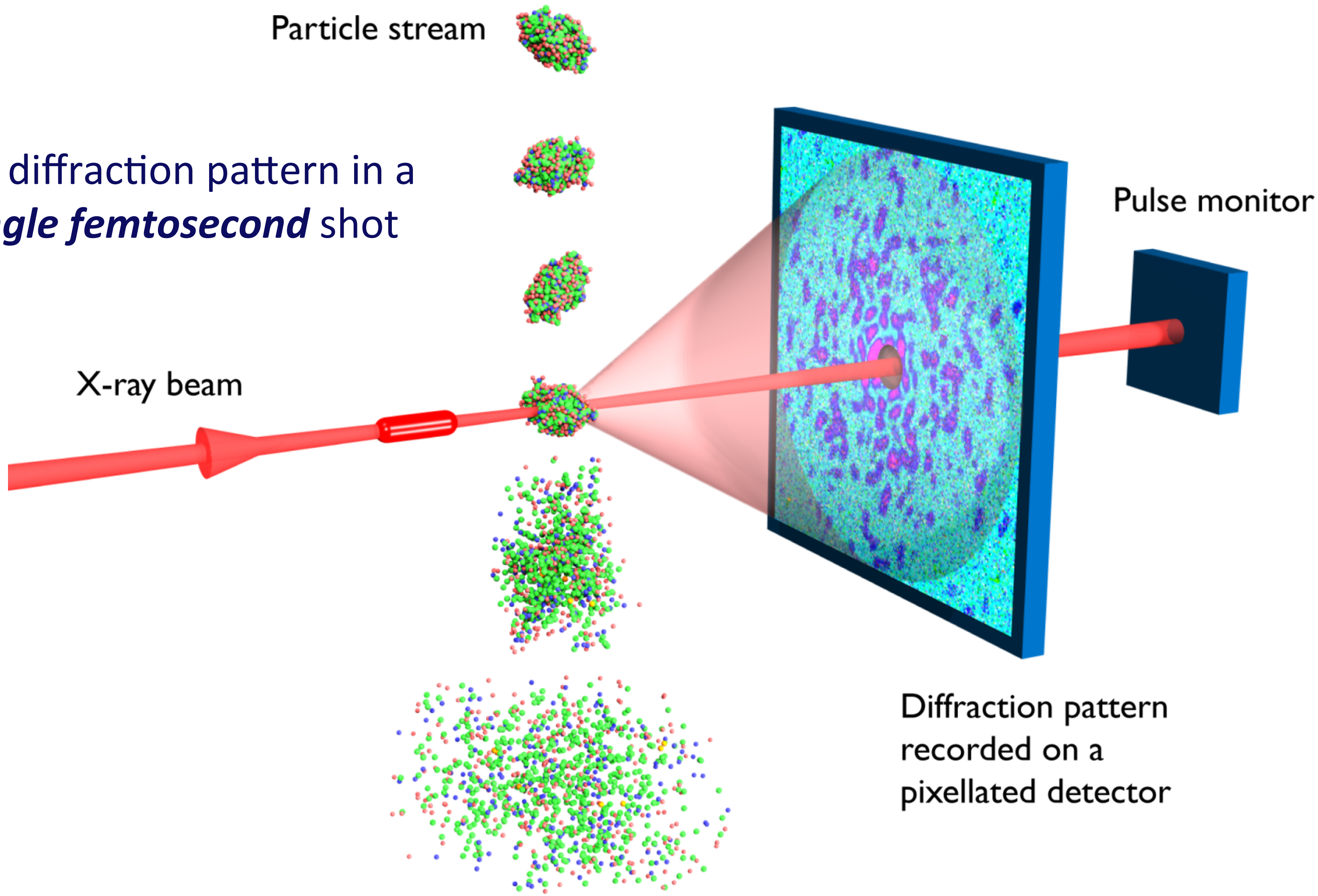


ultrafast diffraction

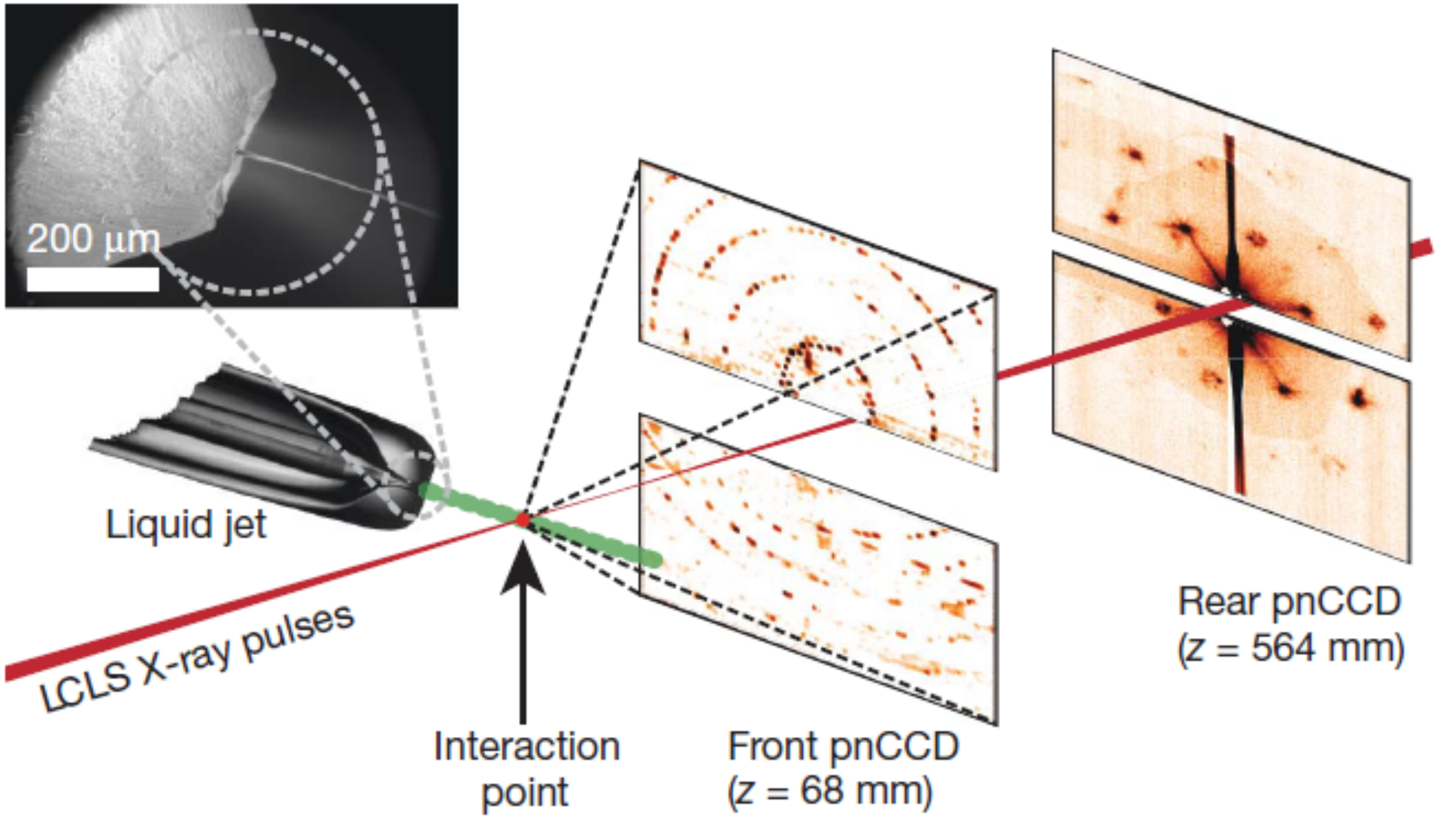
radiation damage, repeatability → *single-shot!*

Since 2009: XFEL

Full diffraction pattern in a ***single femtosecond*** shot

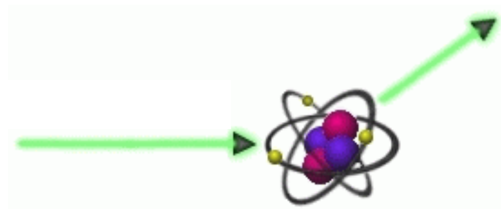


Since 2009: XFEL



Femtosecond, single-shot X-ray diffraction of protein nanocrystals
(Chapman et al., Science 2011)

X-rays vs electrons



X-rays:

Thomson scattering

$$\sigma_T = 6.6\times10^{-29}\ \mathrm{m}^2$$

high density, bulk

3D protein crystals

Electrons:

Rutherford scattering

$$\sigma_R > 10^{-24}\ \mathrm{m}^2$$

gas phase, surfaces

2D membrane proteins

Complementary information!

X-rays vs electrons

Property	Electrons (100 keV)	Hard X-rays (10 keV)
Wavelength	0.04 Å	1.2 Å
Beam control	Charged particle optics, BUT space charge effects	Optics challenging
Beam coherence	$< 10^{-4}$ (EM)	>>1 (XFEL)
Ratio (inelastic/elastic) scattering	3 (carbon)	10
Energy deposited per elastic event	1	>1000
Elastic mean free path	1	$10^5 – 10^6$

X-rays vs electrons

TU/e

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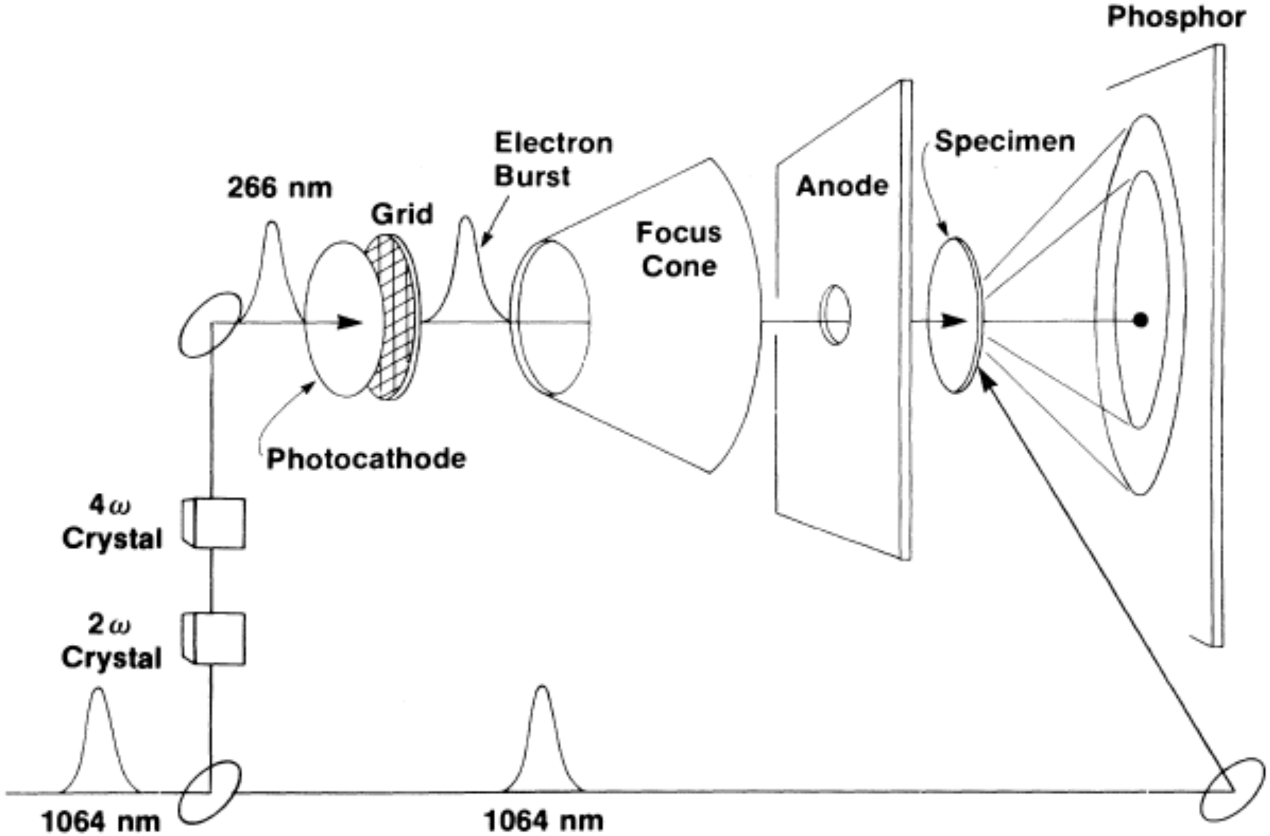
10^6-10^7 (0.1-1 pC) electrons sufficient for single-shot diffraction!

Ultrafast Electron Diffraction (UED)

first generation

TU/e

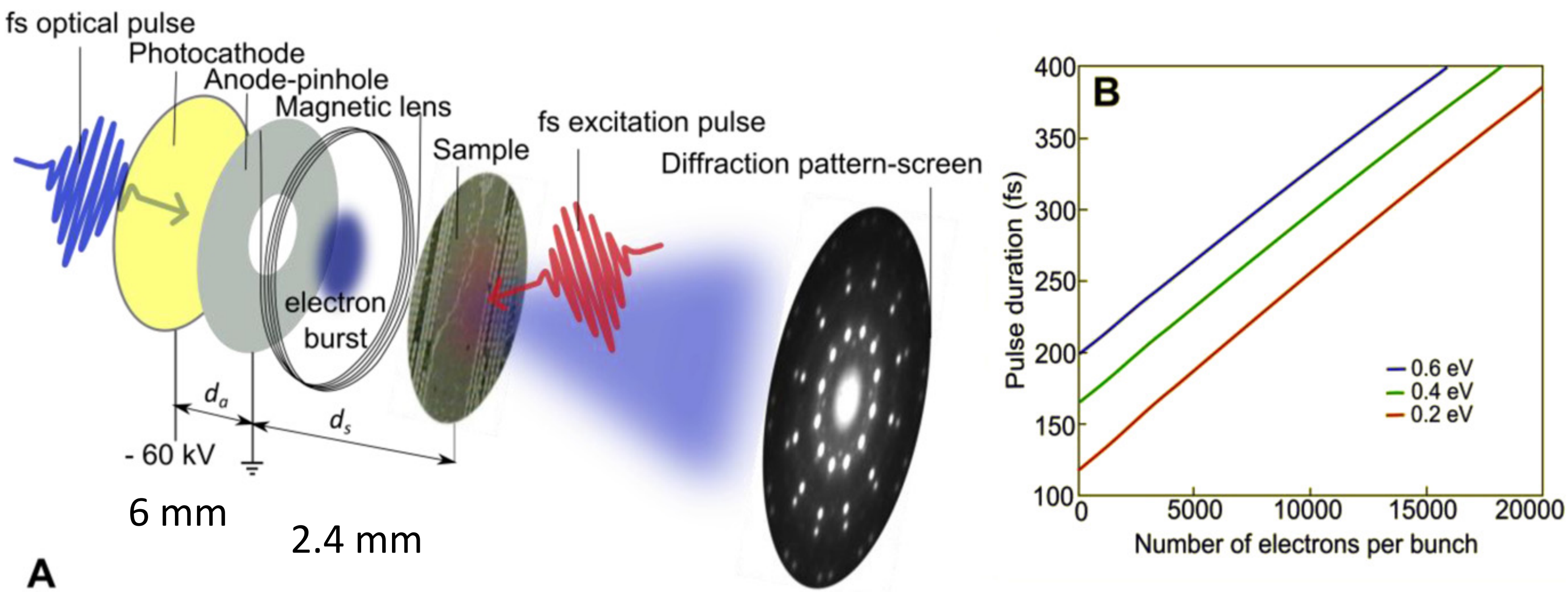
First demonstration with ps pulses by Mourou & coworkers in 1980s



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

UED – first generation

>1990s: Zewail group at CalTech & Miller group at Toronto University



Few 10 keV beam, 10 MV/m at cathode
~10^3 e/pulse @ 1 kHz; ~300 fs resolution

UED – first generation

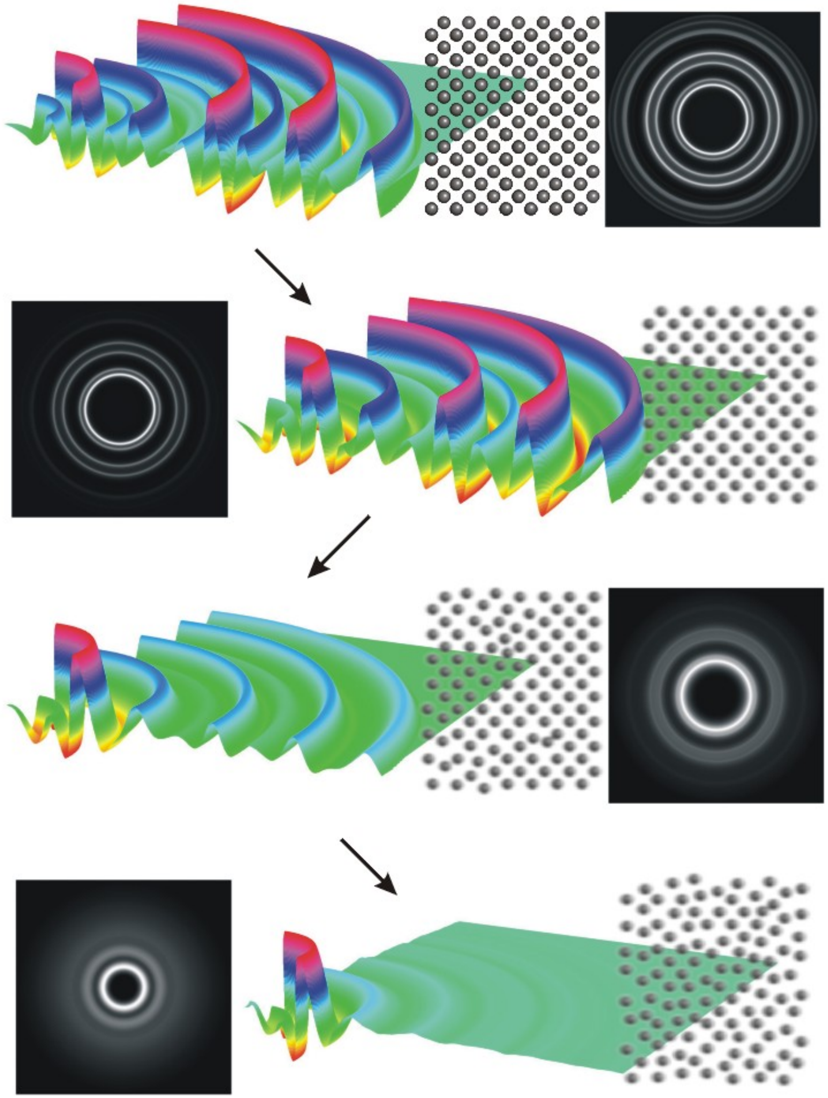
TU/e

0.5 ps

1.5 ps

2.5 ps

3.5 ps

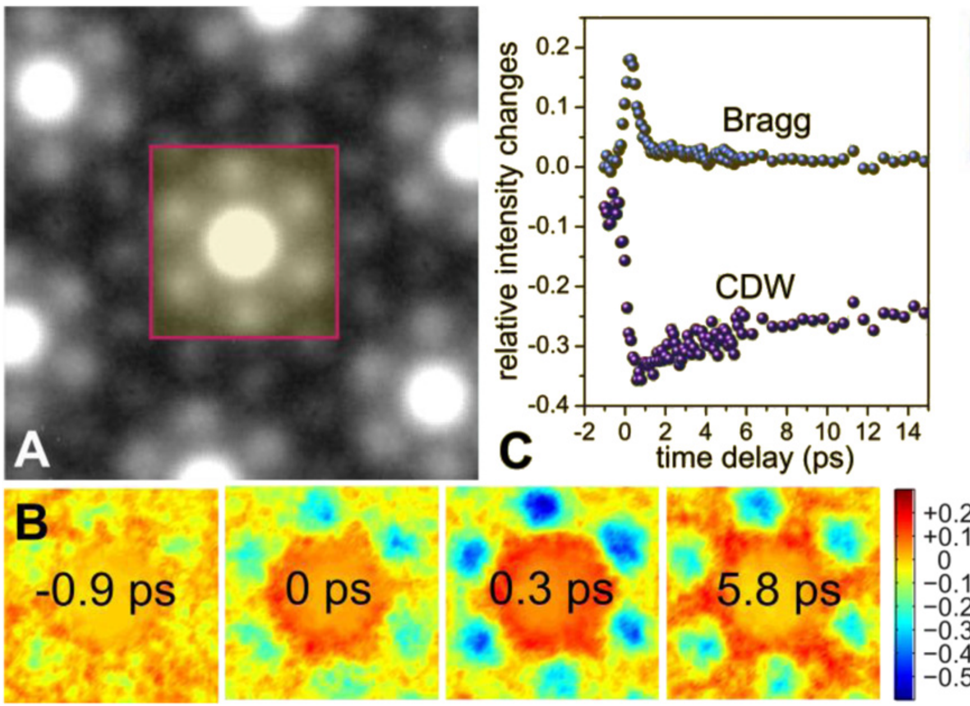


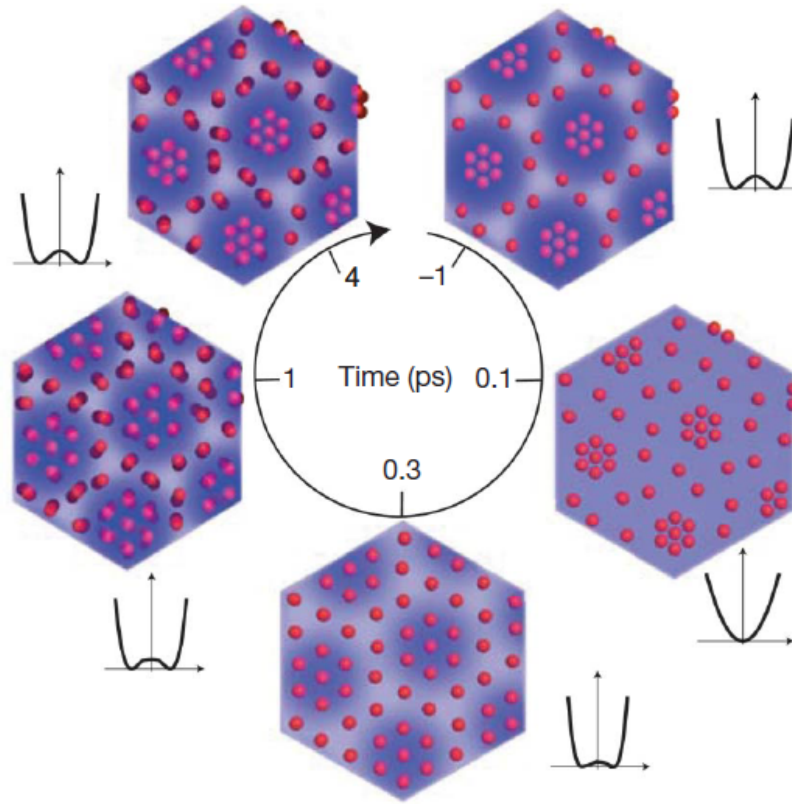
laser induced melting of aluminum

Siwick et al., Science 2003

UED – first generation

Charge Density Waves in 1T-TaS_2

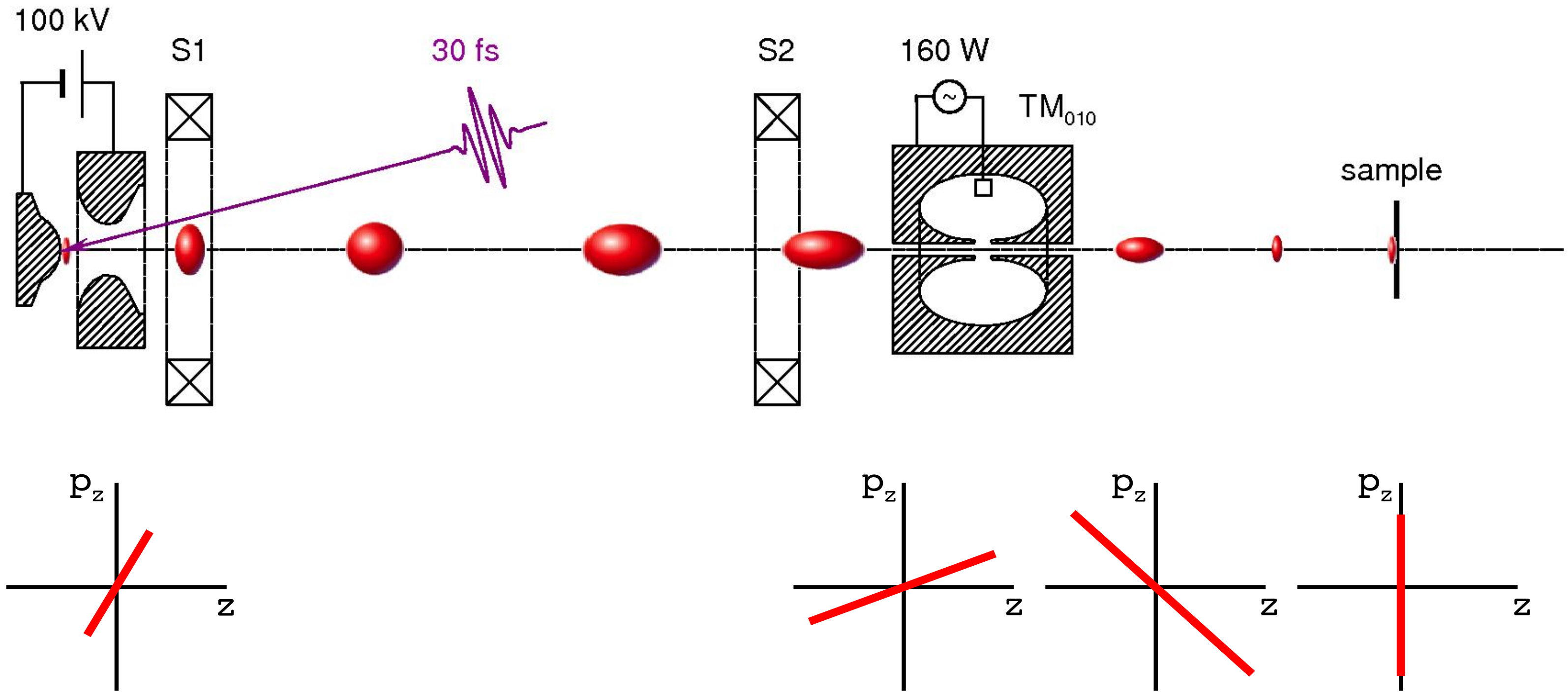




Eichberger et al., Nature 2010

UED – second generation

Radiofrequency techniques → femtosecond, single-shot electron diffraction

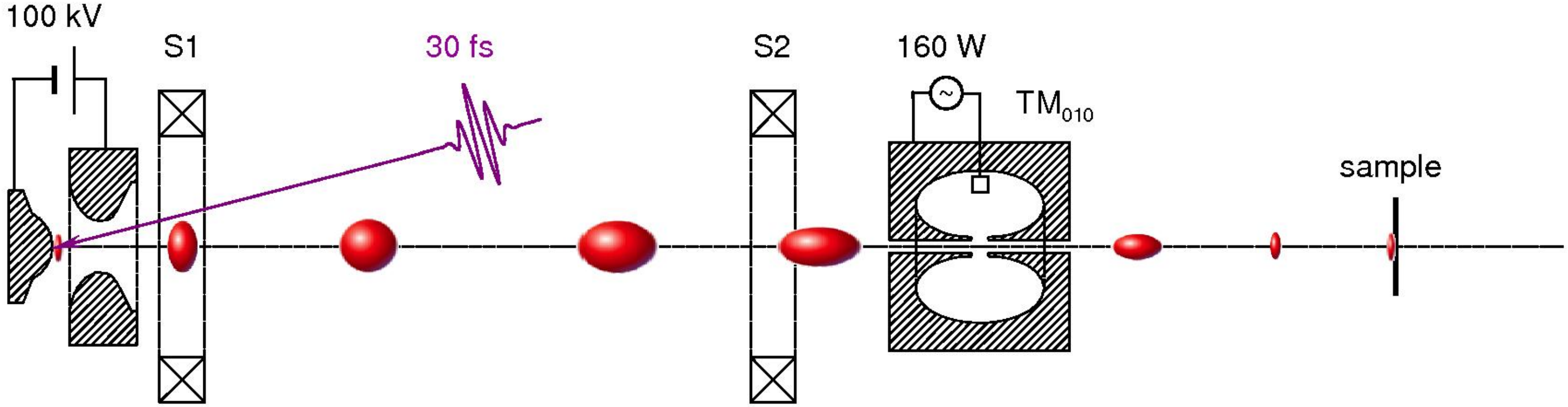


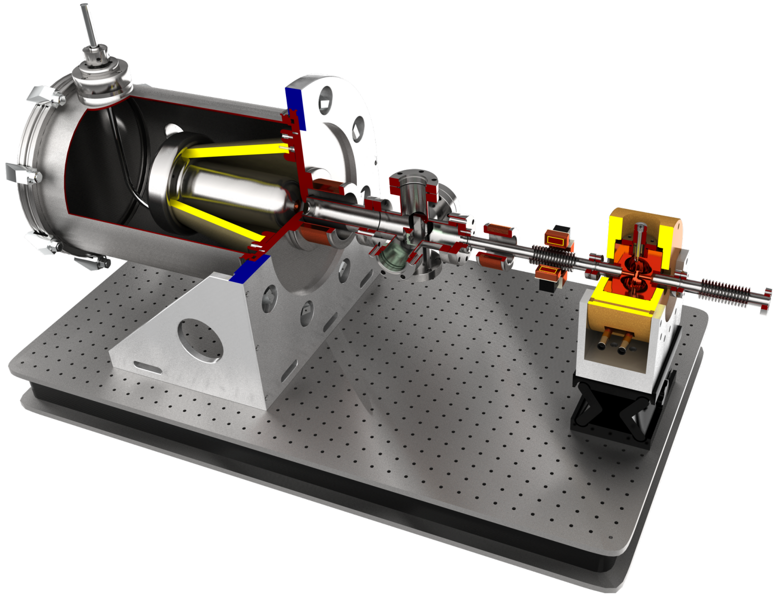
100 keV beam, 10 MV/m at cathode, RF compression
10^6-10^7 e/pulse @ 1 kHz; 100 fs resolution

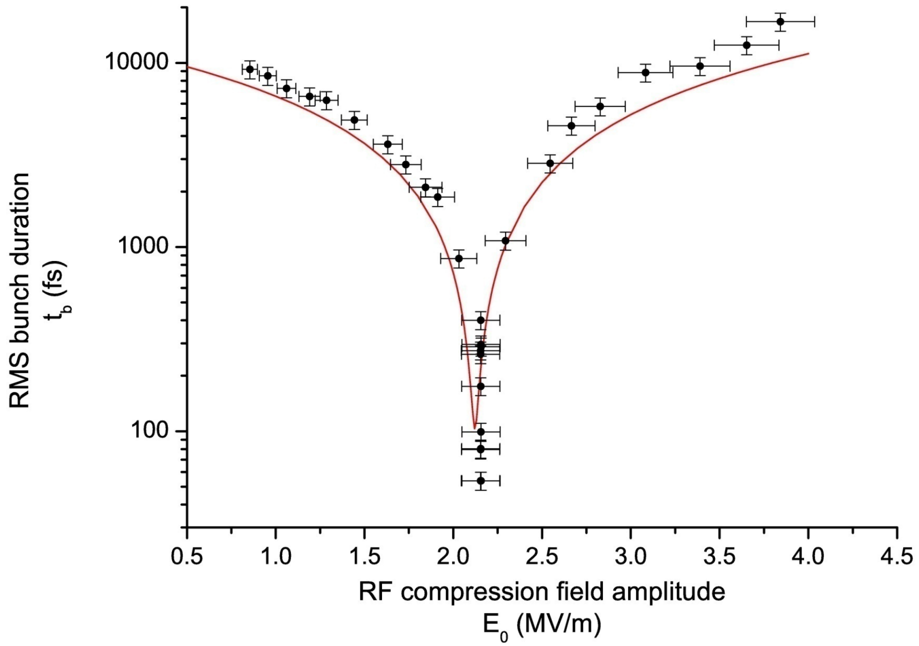
Van Oudheusden et al., PRL **105**, 264801 (2010)

UED – second generation

Radiofrequency techniques ➔ femtosecond, single-shot electron diffraction



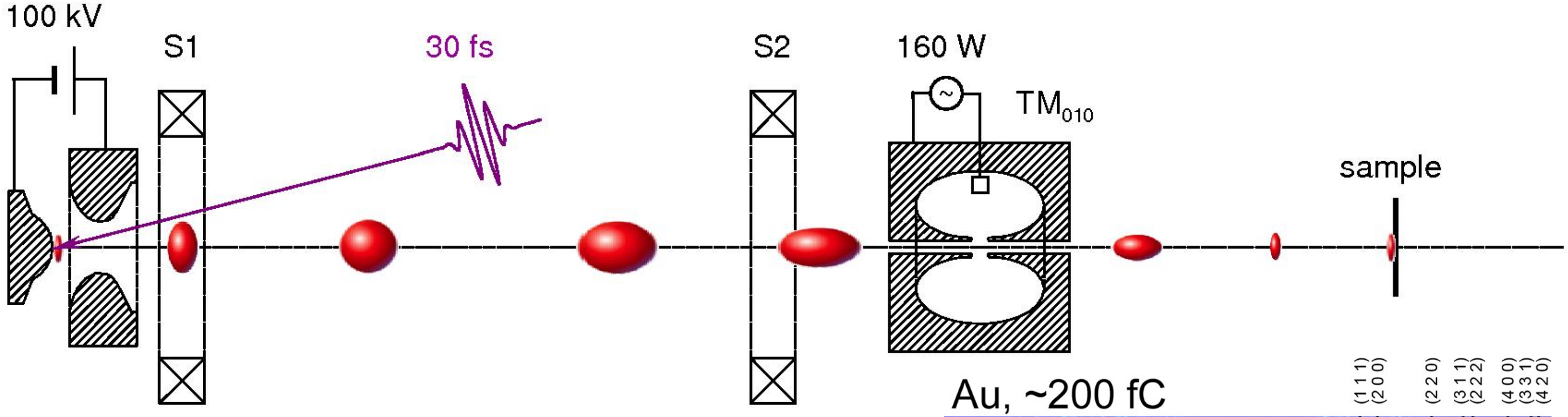


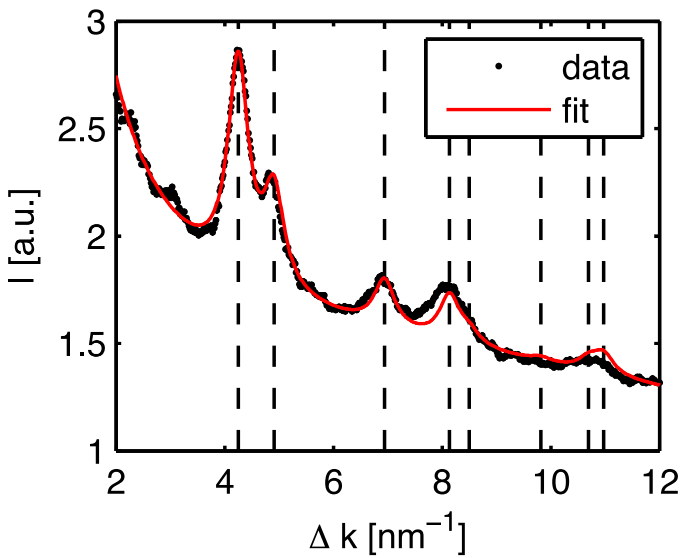


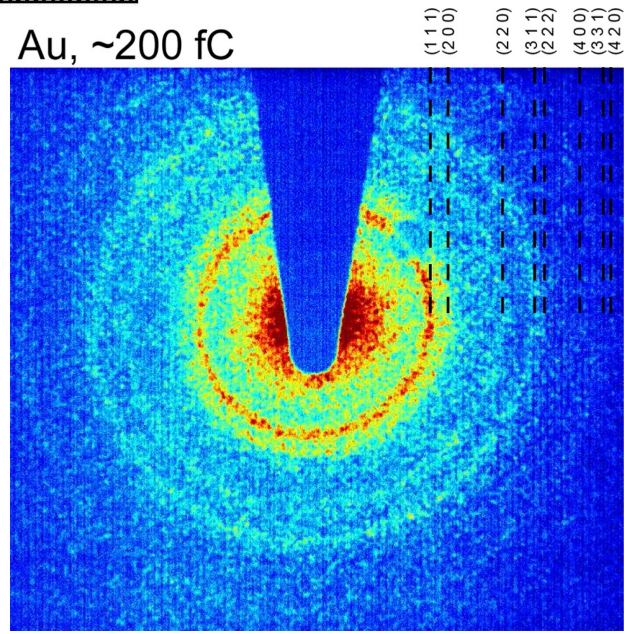
Van Oudheusden et al., PRL **105**, 264801 (2010)

UED – second generation

Radiofrequency techniques ➔ femtosecond, single-shot electron diffraction





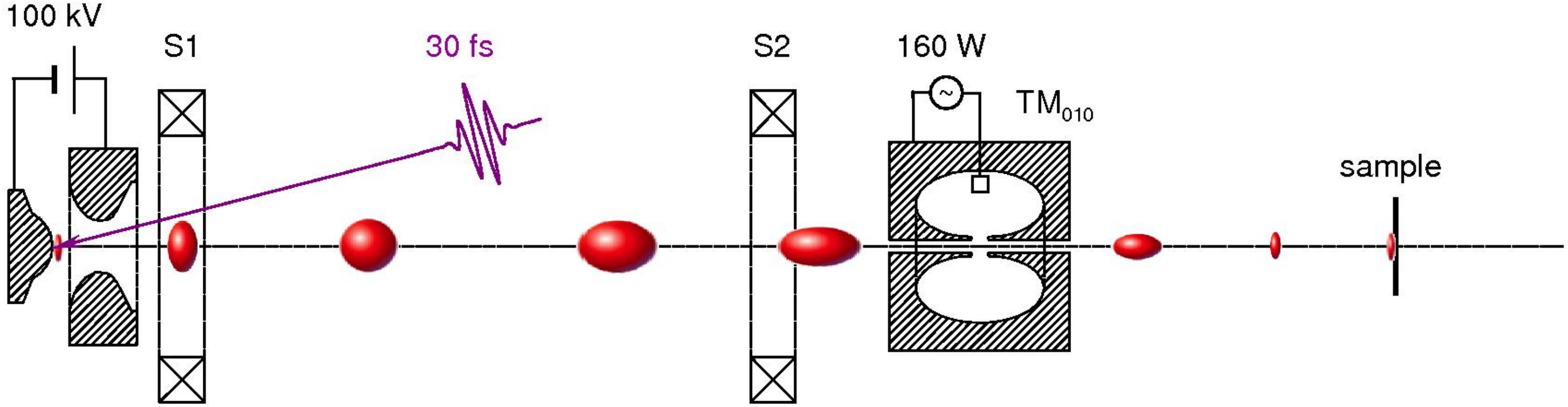


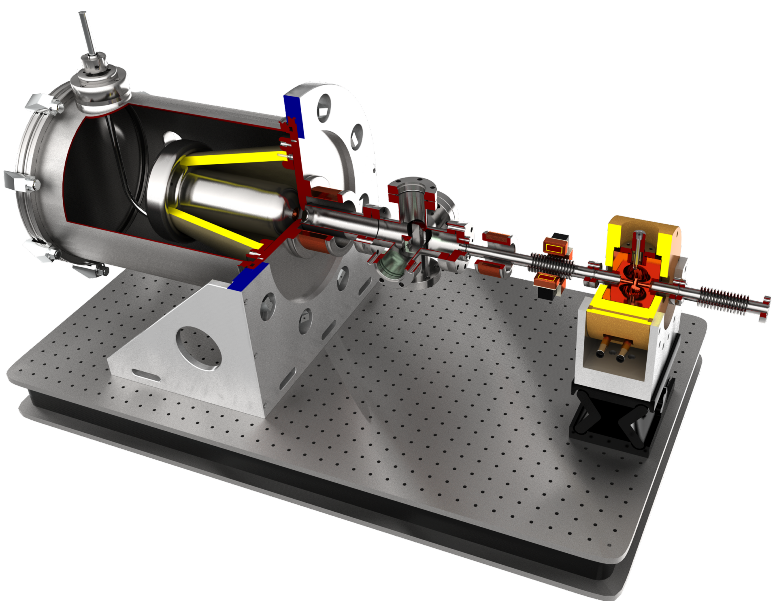
Single-shot diffraction pattern

UED – second generation

TU/e

Radiofrequency techniques → femtosecond, single-shot electron diffraction





$$Q = 0.2 \text{ pC}, \ \varepsilon_n = 40 \text{ nm} \cdot \text{rad}$$

$$\Rightarrow B = \frac{Q}{\varepsilon_n^2} \approx \frac{100 \text{ pC}}{(1 \ \mu\text{m} \cdot \text{rad})^2}$$

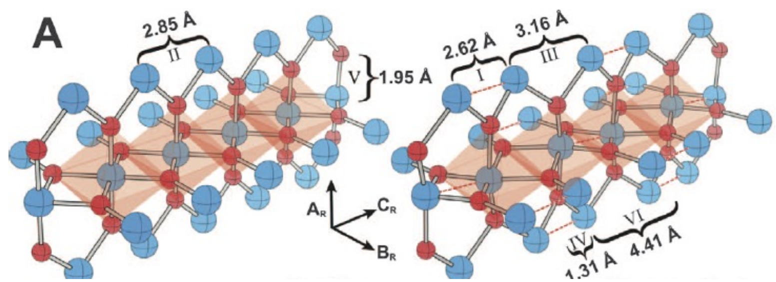
RF photogun Brightness!

UED – second generation

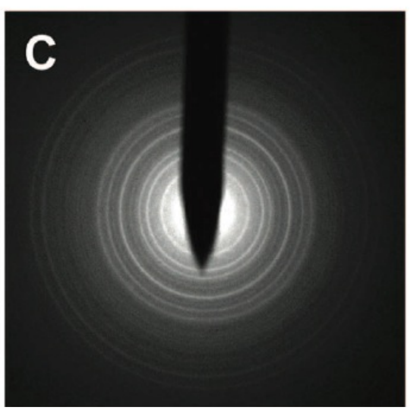
TU/e

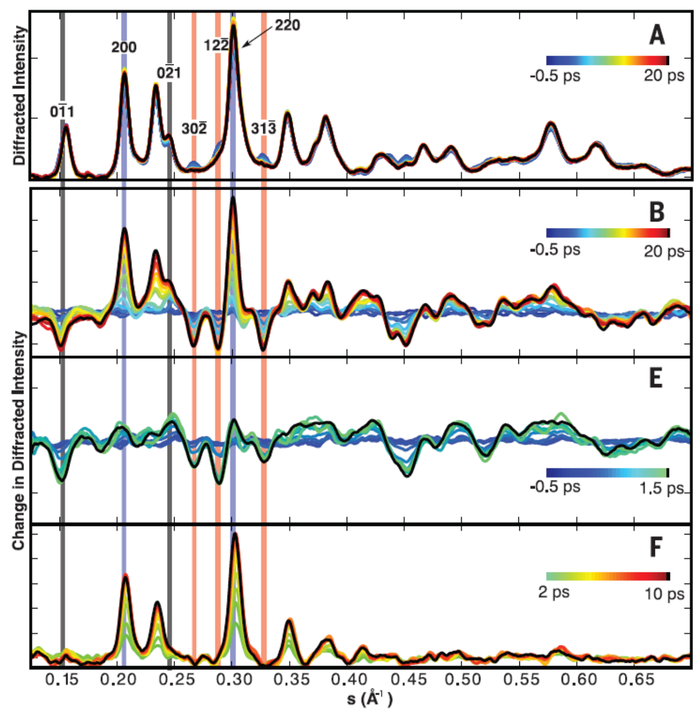
A photoinduced metal-like phase of monoclinic VO_2 revealed by ultrafast electron diffraction
Vance R. Morrison *et al.*
Science **346**, 445 (2014);
DOI: 10.1126/science.1253779

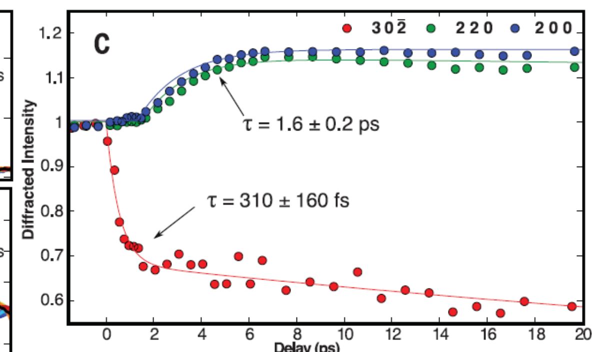
T>343 K: metallic

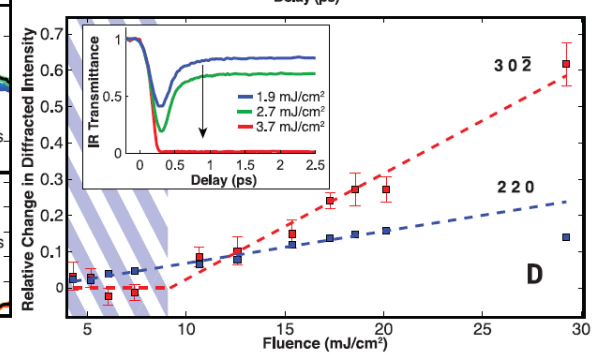


T<343 K: semiconductor









UED – second generation

TU/e

LETTER 18 APRIL 2013 | VOL 496 | NATURE | 343

doi:10.1038/nature12044

Mapping molecular motions leading to charge delocalization with ultrabright electrons

Meng Gao[1,2]*, Cheng Lu[1], Hubert Jean-Ruel[1,2], Lai Chung Liu[1,2], Alexander Marx[2], Ken Onda[3,4], Shin-ya Koshihara[5,6], Yoshiaki Nakano[7], Xiangfeng Shao[7]†, Takaaki Hiramatsu[8], Gunzi Saito[8], Hideki Yamochi[7], Ryan R. Cooney[1,2], Gustavo Moriena[1,2], Germán Sciaini[1,2]* & R. J. Dwayne Miller[1,2]

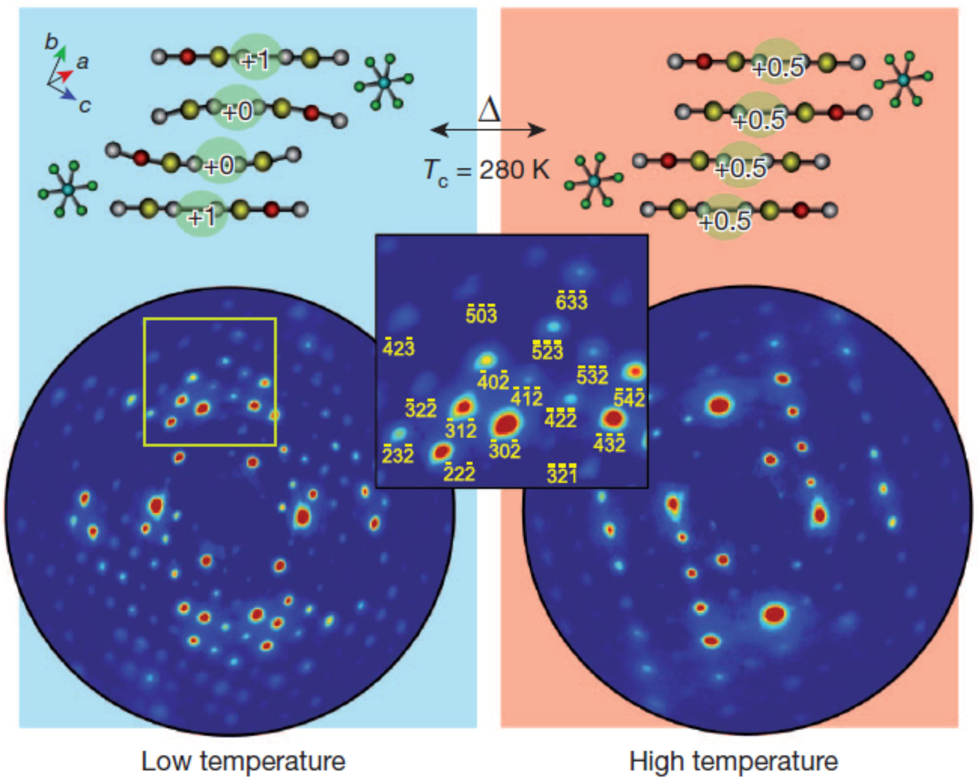


Photo-induced insulator-to-metal phase transition of the organic salt $(EDO\text{-}TTF)_2PF_6$, monitored with 300 fs temporal resolution

UED – second generation

TU/e

To *c* or not to *c*...?

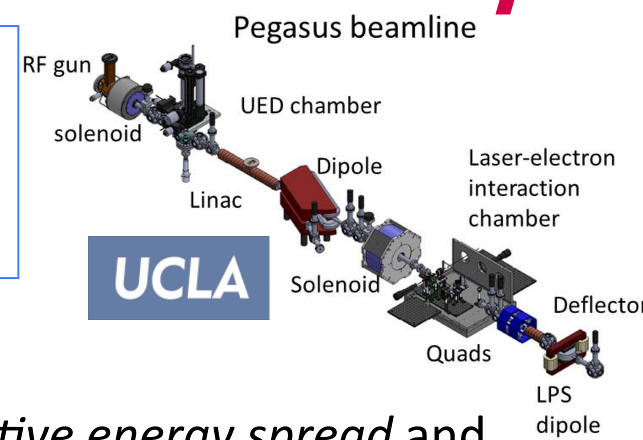
Relativistic beams: better temporal resolution

- higher energy ➔ shorter pulses for the same *relative energy spread* and *longitudinal emittance*
- relativistic suppression *space charge* effects ➔ shorter pulses
- minimization *pump-probe velocity mismatch* for thick (gas phase) samples

Towards <10 fs temporal resolution!

UED – second generation

UED with **RF photoguns**:
2-5 MeV beam, 50-100 MV/m at cathode
~10^7 e/pulse, single-shot diffraction



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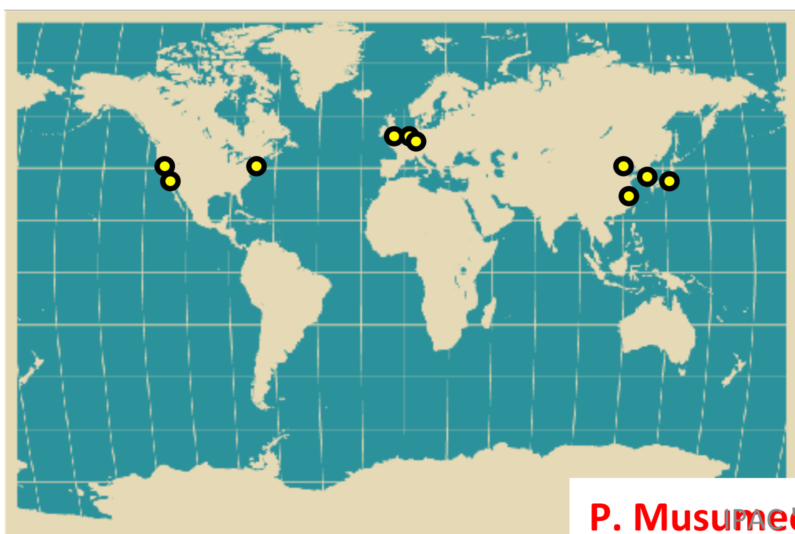


A rapidly growing field:

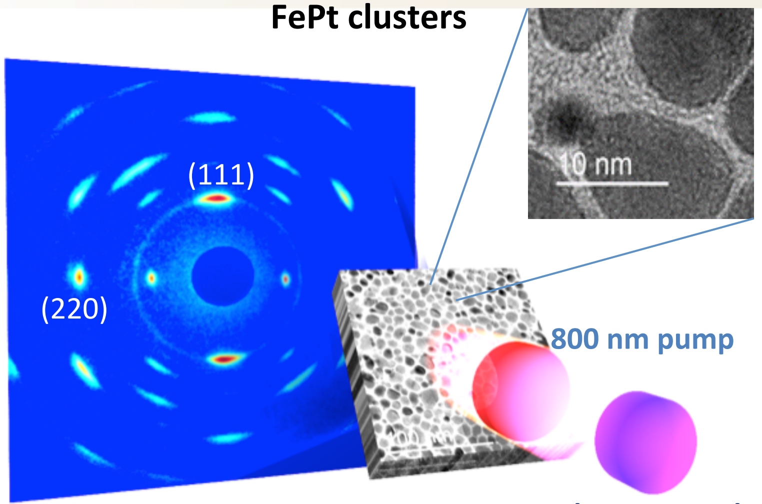
Efforts at SLAC, LLNL, UCLA, Tsinghua Univ., Osaka Univ., BNL, DESY, POSTECH, Diamond UK, Shanghai Jiaotong Univ., etc.

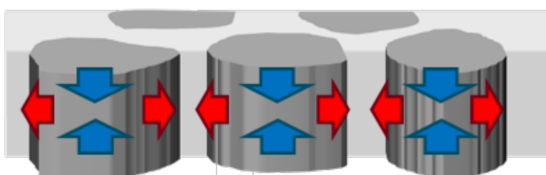
P. Musumeci and R. K. Li, ICFA Newsletter No. 59, pp. 13-33.

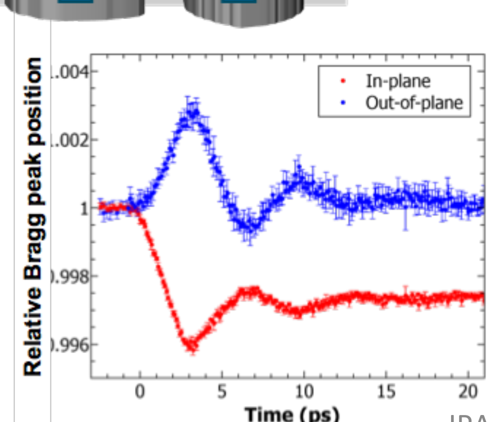
First Ultrafast Materials Science Experiments @ SLAC

SLAC

Large-q of electrons allows to simultaneously measure in-plane and out-of-plane motion in FePt clusters

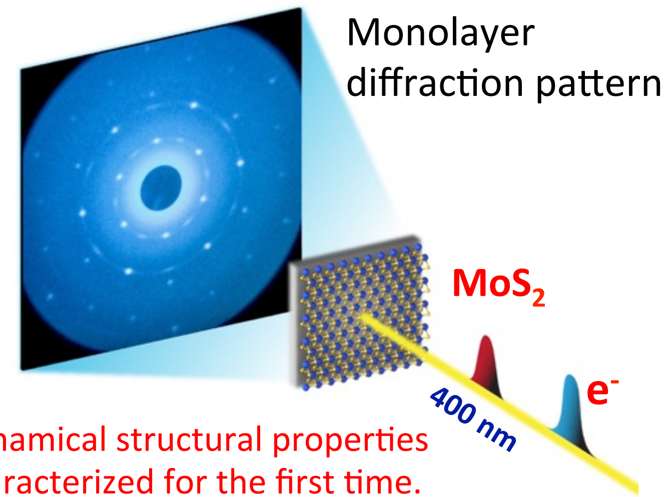






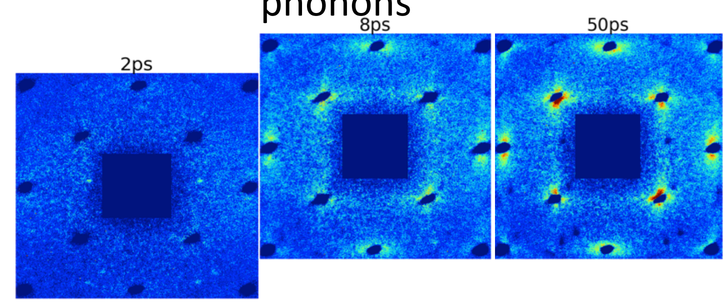
Observation of anisotropic volume expansion and volume conserving breathing mode in FePt clusters

Large scattering cross section allows to probe individual atomic layers (top) and diffuse scattering from phonons (bottom)



Dynamical structural properties characterized for the first time.

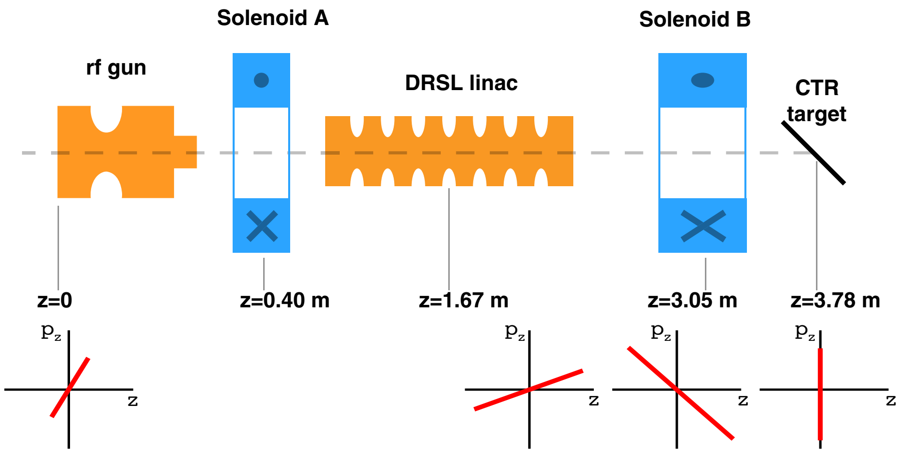
Diffuse scattering from non-equilibrium phonons

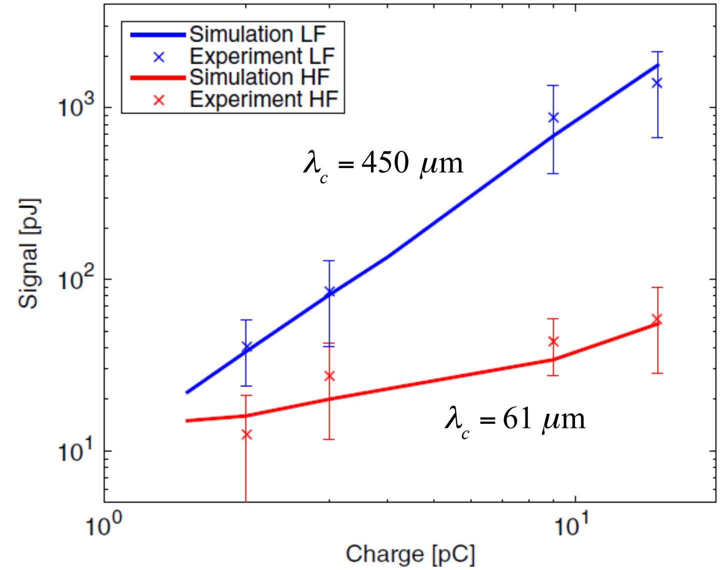


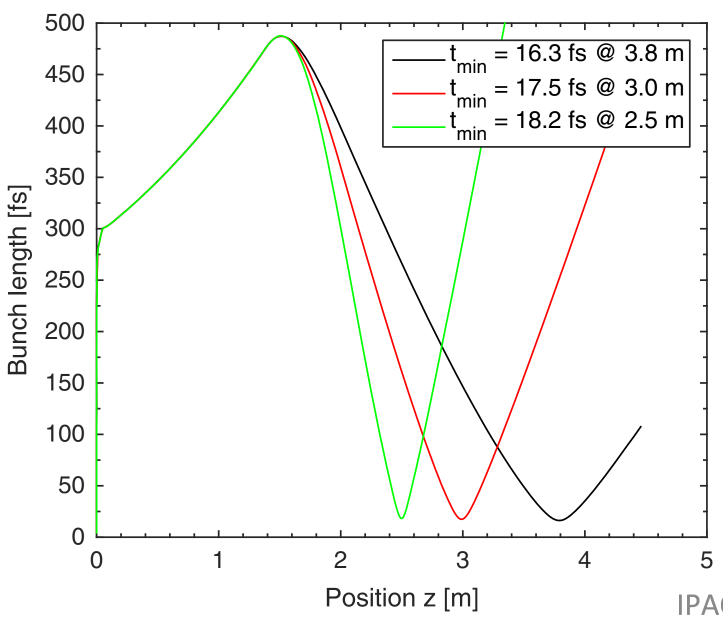
Probe electron-phonon coupling and phonon thermalization of phonons in 20nm Au films

Courtesy of R.K. Li and X.J. Wang

RF compression of MeV electron bunches for Ultrafast Electron Diffraction







CTR spectrum based diagnostic

TABLE III. Measured electron pulse duration.

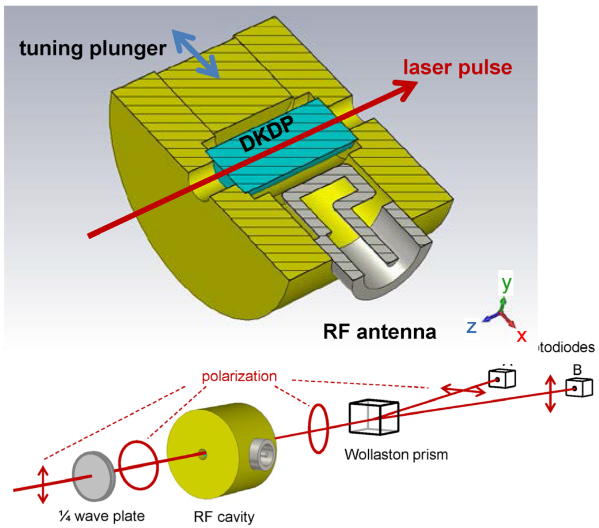
Charge	Bunch length	Simulation
2 pC	33 fs–51 fs	35 fs
3 pC	18 fs–44 fs	38 fs
9 pC	50 fs–63 fs	71 fs

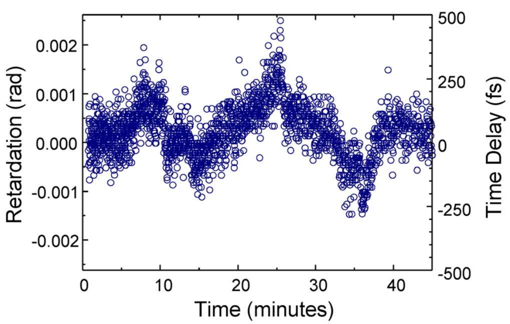
X. Lu et al., PRSTAB 2015

Beyond time-jitter limited resolution

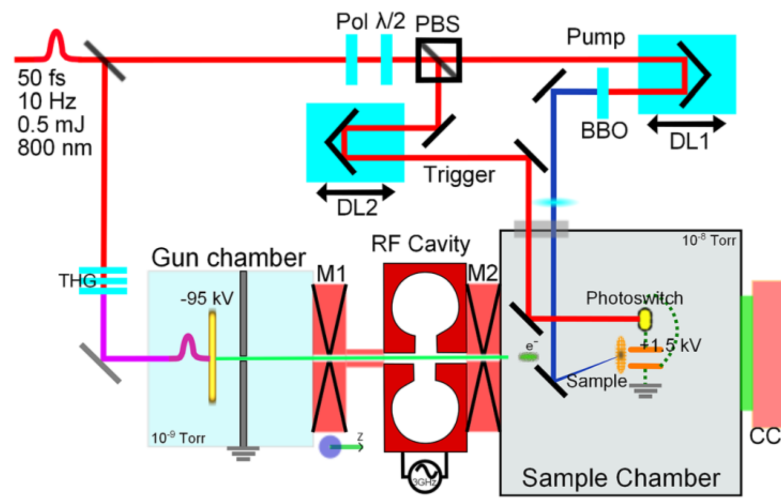
RF-fs laser timing jitter is at this point the main limitation in temporal resolution

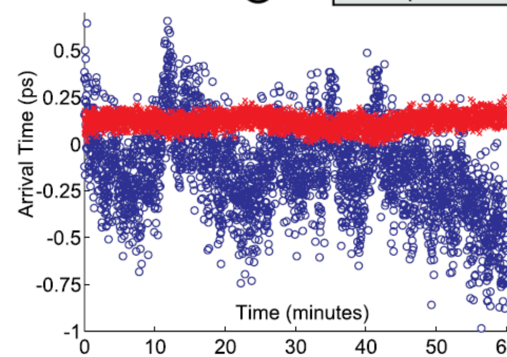
Time stamp RF phase in RF cavity using electro-optic method





Time stamp arrival time electron bunch using laser-triggered streak camera





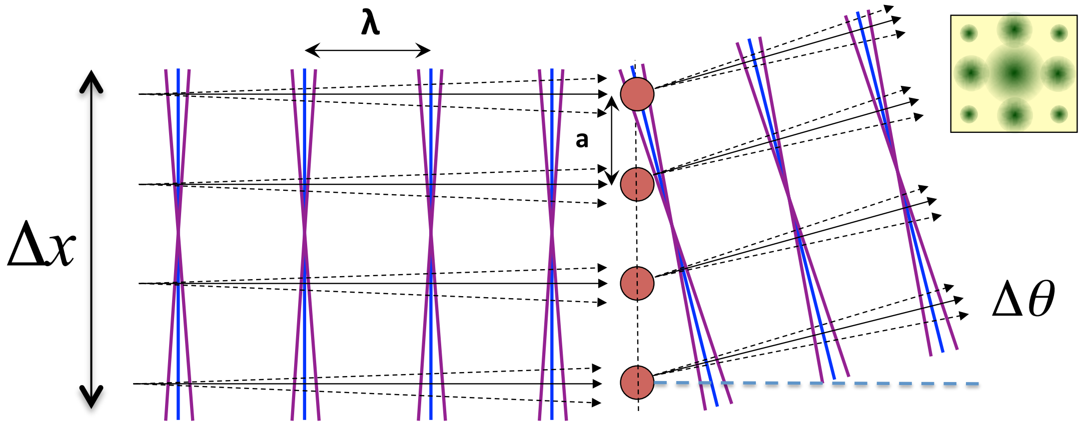
- Few-10 fs resolution;
- < 10 fs possible;
- more methods being developed.

G.J.H. Brussaard et al,. APL 2013

M. Gao et al., APL 2013

UED – what is next?

Not yet possible: single-shot UED of macromolecular crystals



a = lattice constant
d = crystal size

Requirements:

- Visibility diffraction pattern: $\Delta\theta << \lambda / a$ $\Rightarrow \varepsilon_n << \frac{\hbar}{mc}\frac{d}{a} = (0.4\ \text{pm}\cdot\text{rad})\frac{d}{a}$
- Not waste any electron: $\Delta x \leq d$

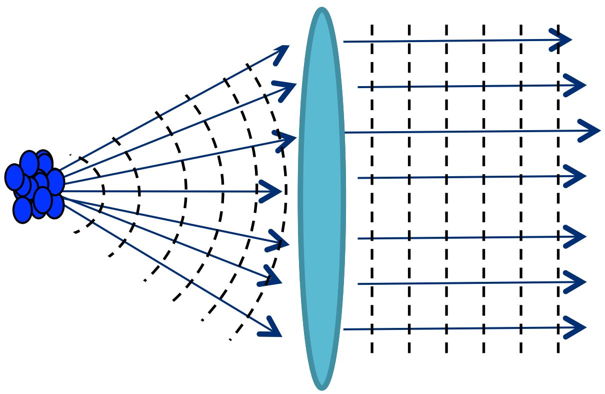
Protein crystals: $\frac{d}{a} \approx 10^4 \Rightarrow \varepsilon_n << 4\ \text{nm}\cdot\text{rad}$

Single-shot UED of protein crystals: $Q = 0.1 - 1\ \text{pC} \Rightarrow B = \frac{Q}{\varepsilon_n^2} >> \frac{10\ \text{nC}}{(1\ \mu\text{m}\cdot\text{rad})^2}$

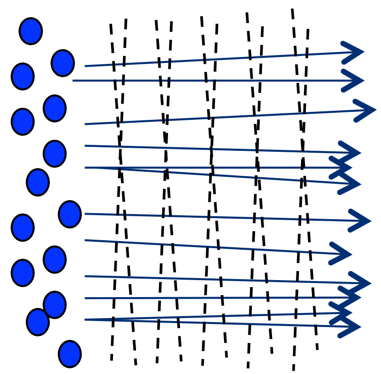
UED – what is next?

TU/e

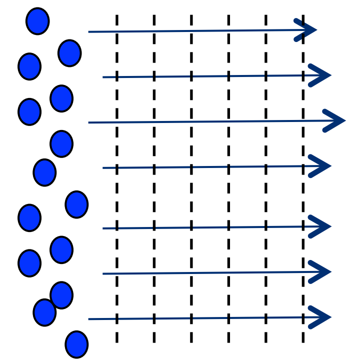
$$\varepsilon_n = \sigma_x \sqrt{\frac{kT}{mc^2}}$$



high extraction field
point-like source



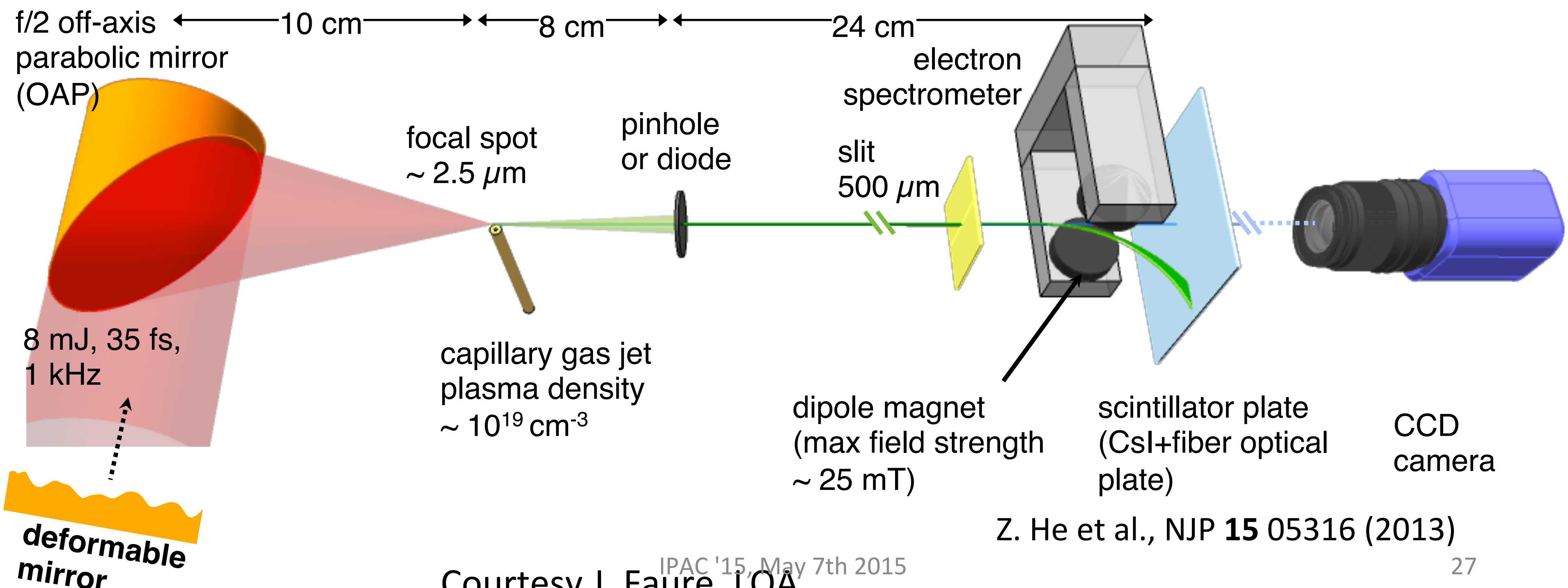
conventional extended
photoemission source



extended
ultracold source

kHz laser-plasma accelerator for UED

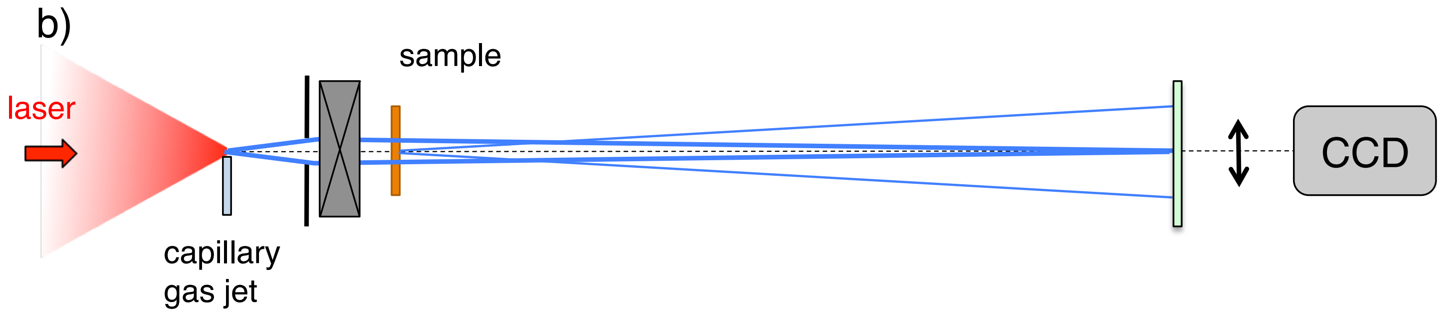
- 100 GeV/m accelerating gradient
  - MeV electron bunches
  - Mitigates space charge
- Few femtosecond duration possible (O. Lundh et al., Nat. Phys. 2011)
- Accelerating structure is generated by the laser pulse
  - Perfect synchronization
  - No jitter in pump-probe experiment



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

Courtesy J. Faure, LOA

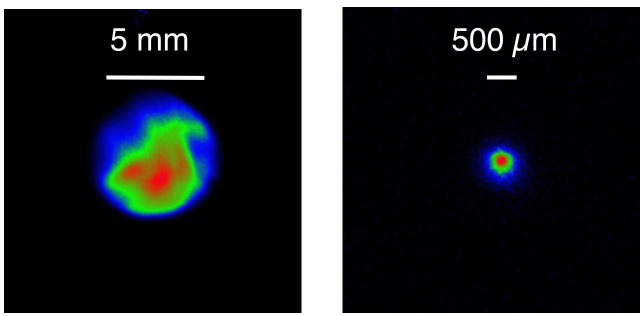
kHz laser-plasma accelerator for UED



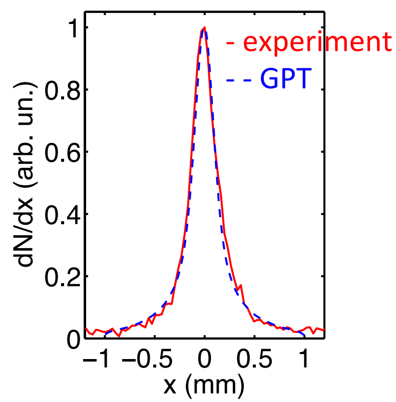
Stable kHz electron beams

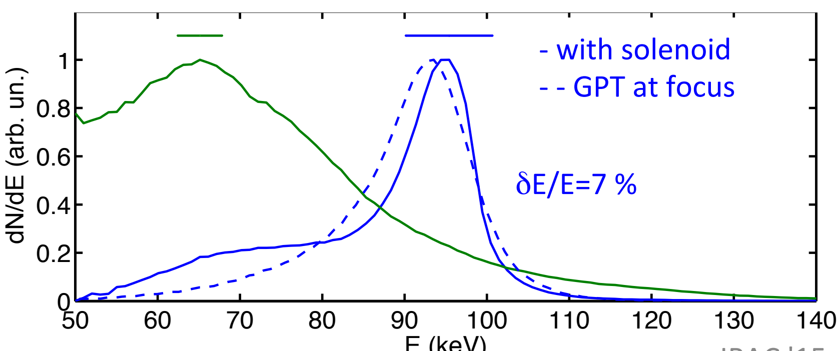
after pinhole

focused with solenoid



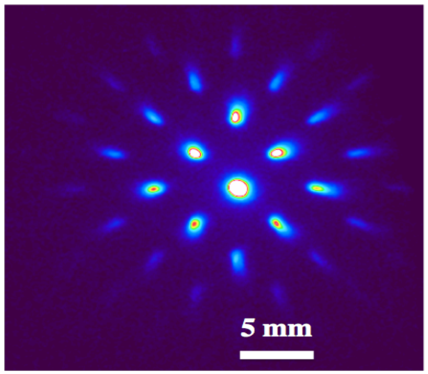
280 µm FWHM at focus





Courtesy J. Faure, LOA

Au diffraction pattern



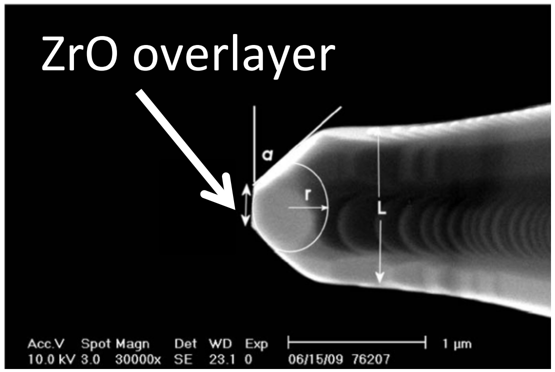
$$Q = 5\ \mathrm{fC}$$

$$\varepsilon_n = 20\ \mathrm{nm}\cdot\mathrm{rad}$$

$$\Rightarrow B = \frac{Q}{\varepsilon_n^2} \approx \frac{10\ \mathrm{pC}}{(1\ \mu\mathrm{m}\cdot\mathrm{rad})^2}$$

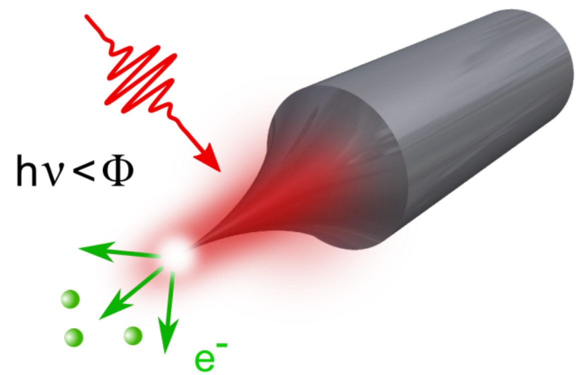
Z. He et al., Appl. Phys. Lett. 102, 064104 (2013)

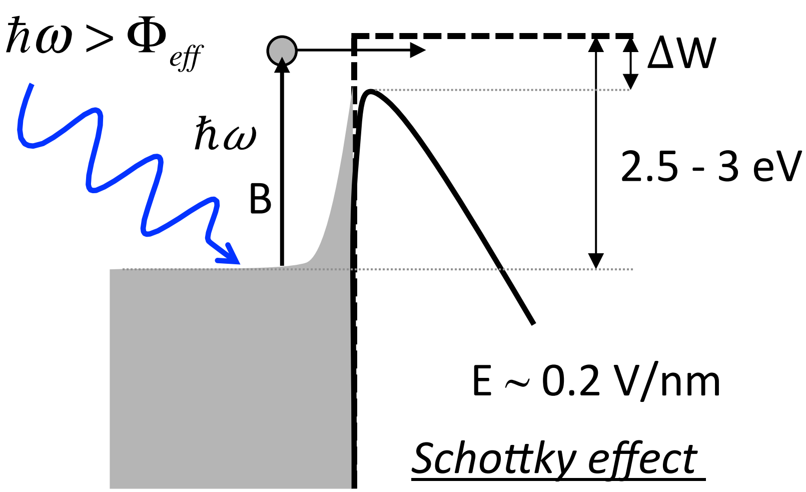
Localized electron emission from needle-shaped photocathodes



Liu et al., J. Vac. Sci Tech. (2010).

Localized Photoemission
(Multiphoton PE,
localized single-photon PE)





photon energy: 3.1 eV (λ = 400 nm)

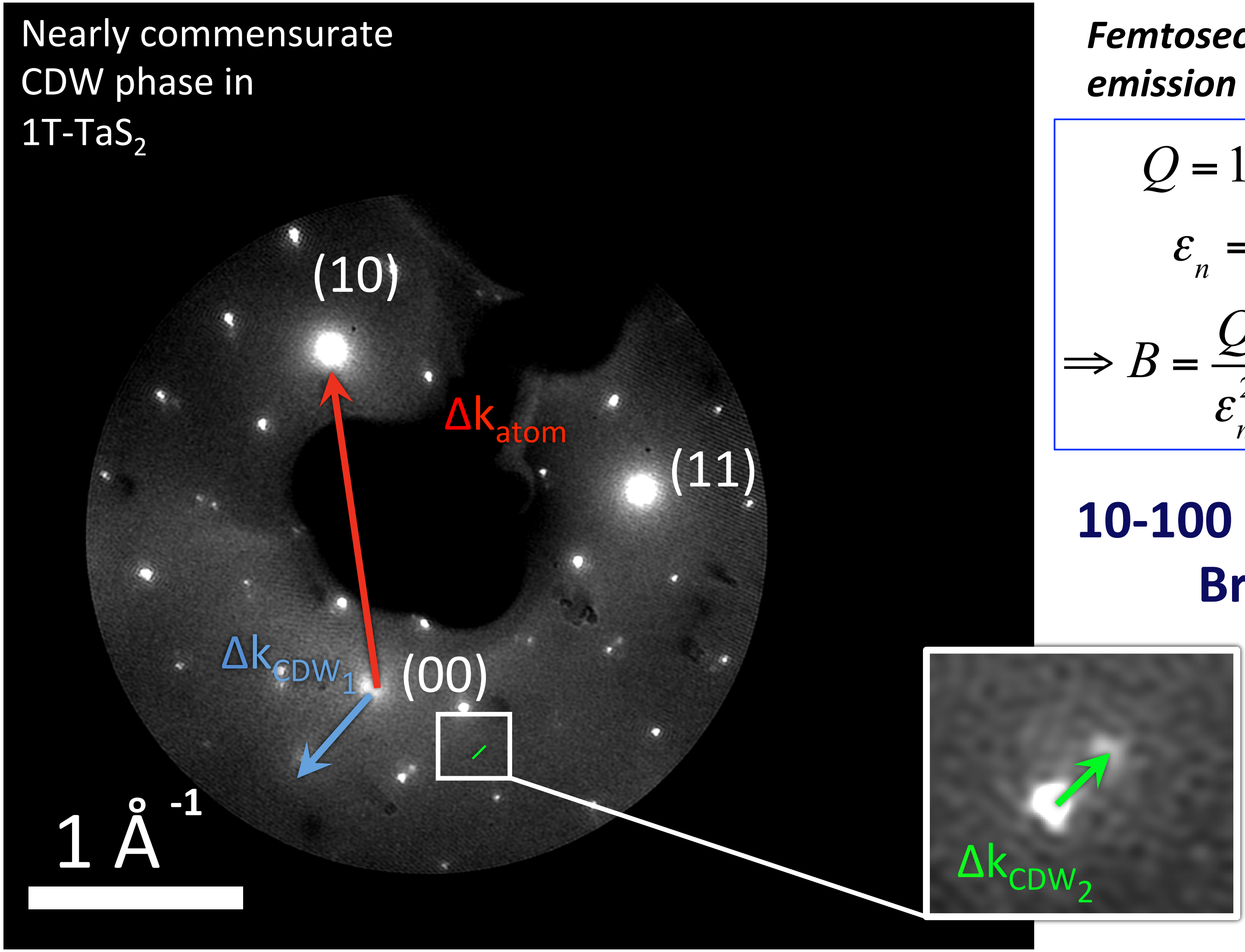
1PPE localization by

- by Schottky effect
- by facet-specific workfunction
- by optical field.enhancement

Courtesy S. Schäfer, C. Ropers, Univ. Göttingen

Ultrafast *Low Energy* Electron Diffraction (ULEED)

E_{kin} = 80 eV , 20 s integration time



Femtosecond single-electron emission near quantum limit

$$Q = 1.6 \times 10^{-19}\ \text{C}$$

$$\varepsilon_n = 4\ \text{pm} \cdot \text{rad}$$

$$\Rightarrow B = \frac{Q}{\varepsilon_n^2} = \frac{10\ \text{nC}}{(1\ \mu\text{m} \cdot \text{rad})^2}$$

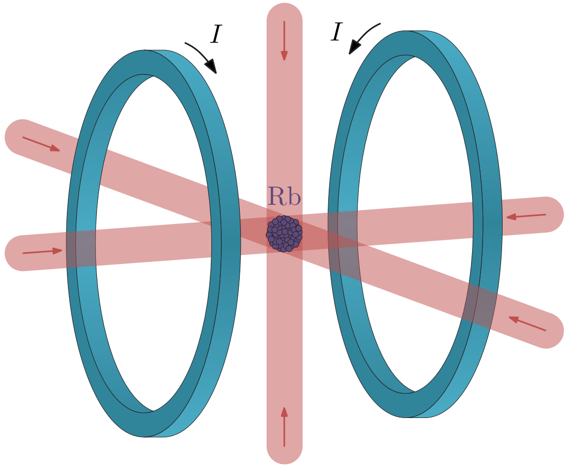
10-100 × RF photogun Brightness!

transverse coherence length: >25 nm

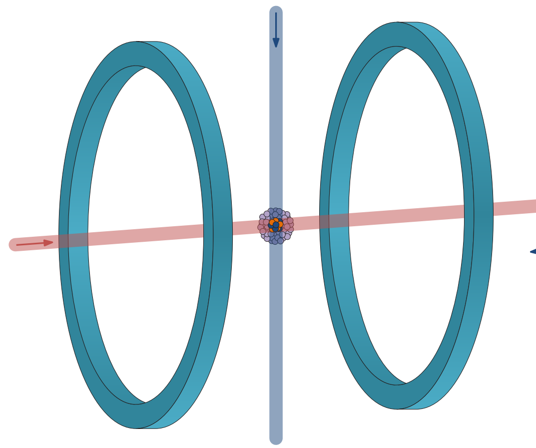
Courtesy S. Schäfer, C. Ropers, Univ. Göttingen

Laser-cooled electron source

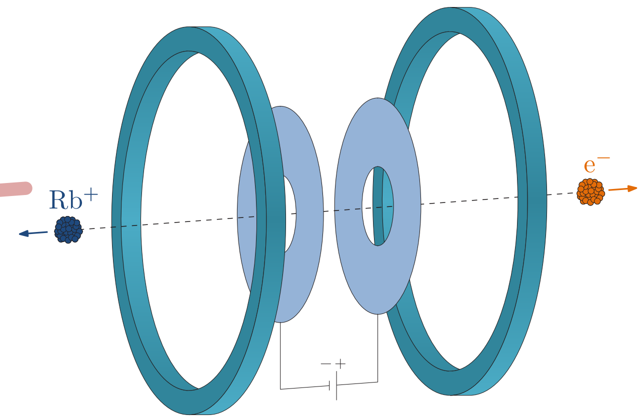
Laser cooling & trapping

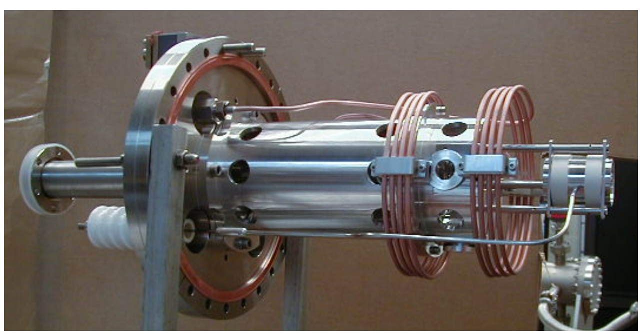


Near-threshold fs photoionization

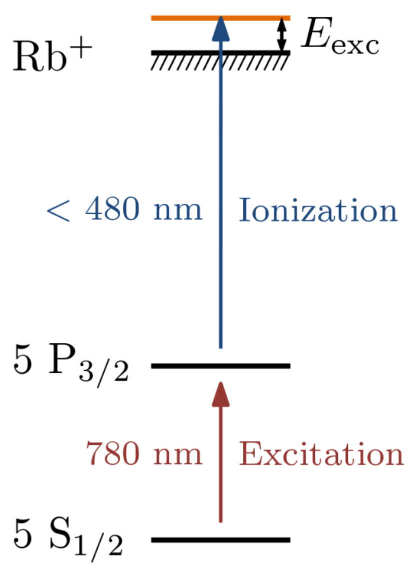


Acceleration

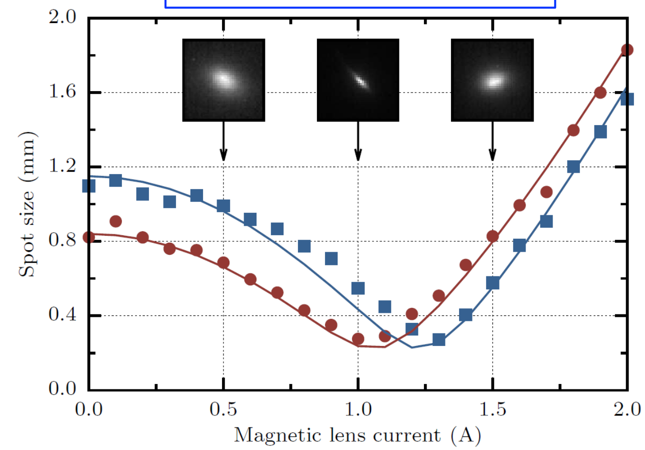




Magneto-Optical Trap (MOT) inside coaxial accelerator



$T_{electron} = 18\ \mathrm{K}$



G. Taban et al., PRSTAB 2008

Engelen et al., Nat. Comm. 2013

Laser-cooled electron source

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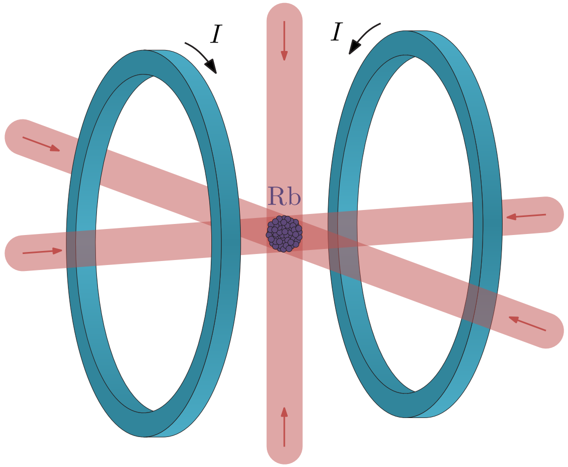
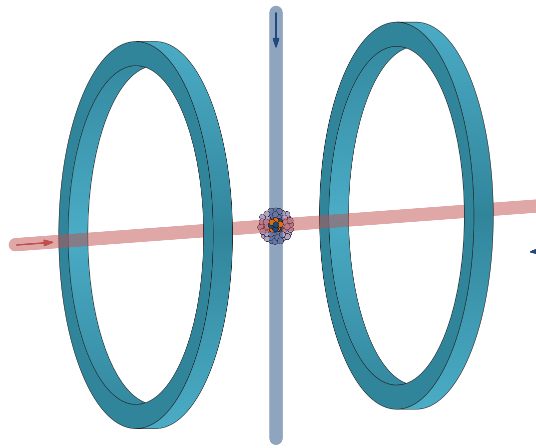
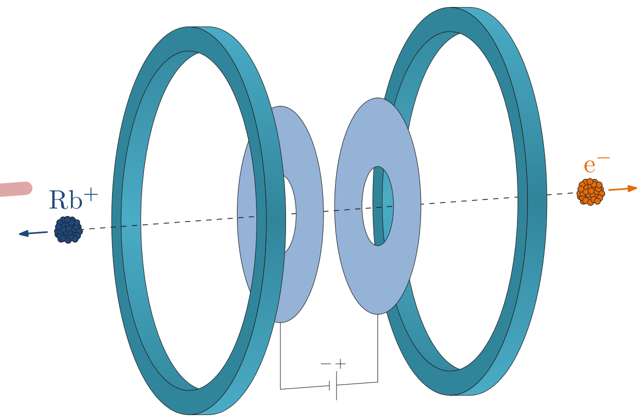
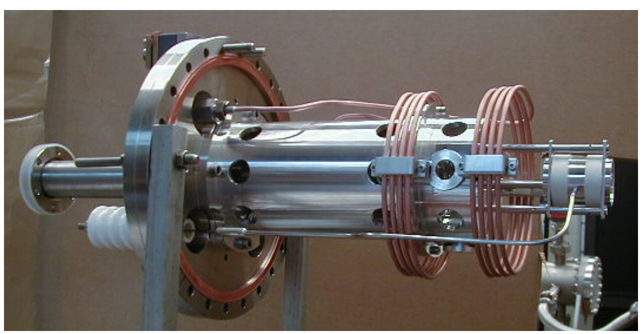


Near-threshold fs photoionization



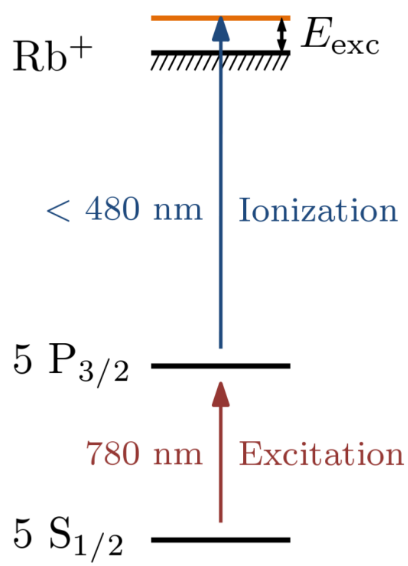
Acceleration



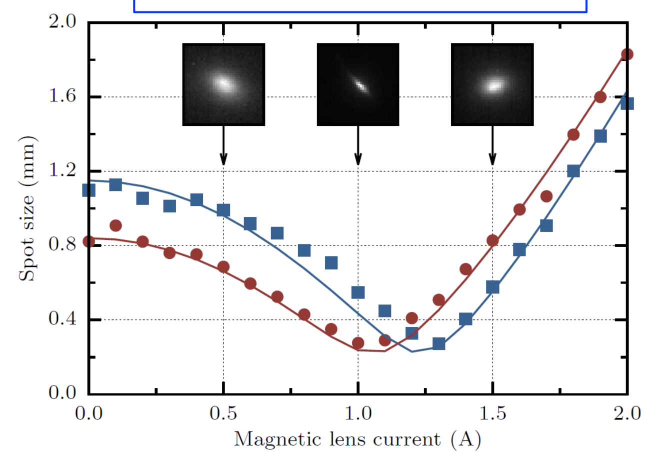


Magneto-Optical Trap (MOT) inside coaxial accelerator

G. Taban et al., PRSTAB 2008

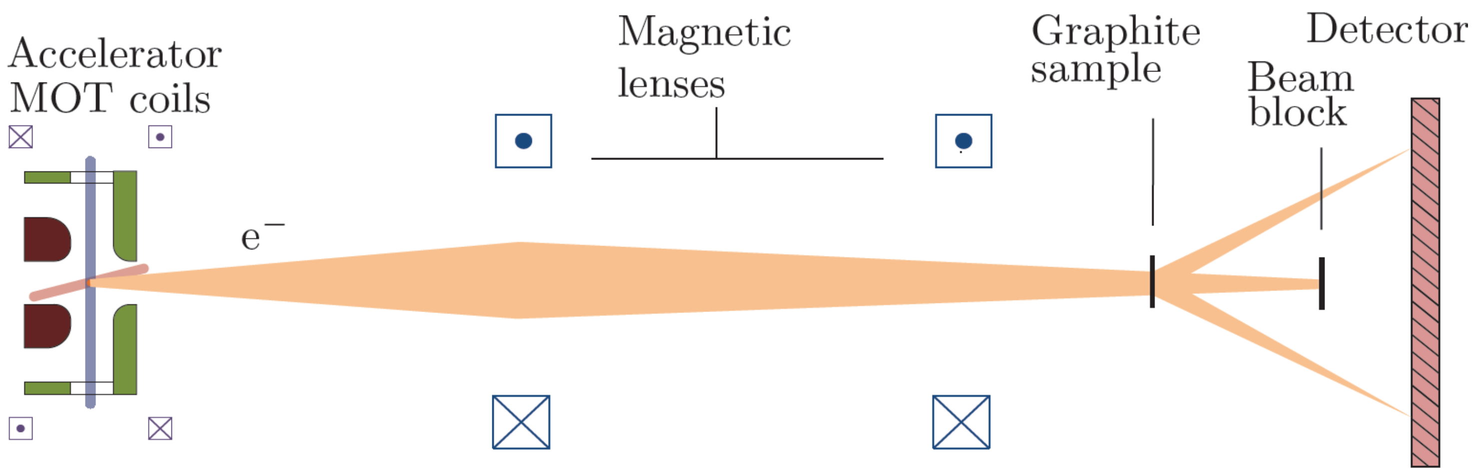


$$\varepsilon_n = 1.4 \text{ nm} \cdot \text{rad}$$

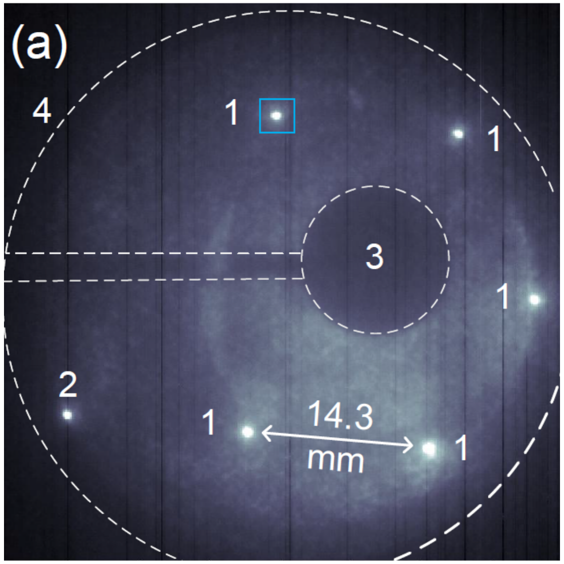


Engelen et al., Nat. Comm. 2013

Laser-cooled electron source



Graphite diffraction pattern (13.2 keV)



$$Q = 0.1 - 0.2 \text{ fC}$$

$$\varepsilon_n = 1.4 \text{ nm} \cdot \text{rad}$$

$$\Rightarrow B = \frac{Q}{\varepsilon_n^2} \approx \frac{100 \text{ pC}}{(1 \ \mu\text{m} \cdot \text{rad})^2}$$

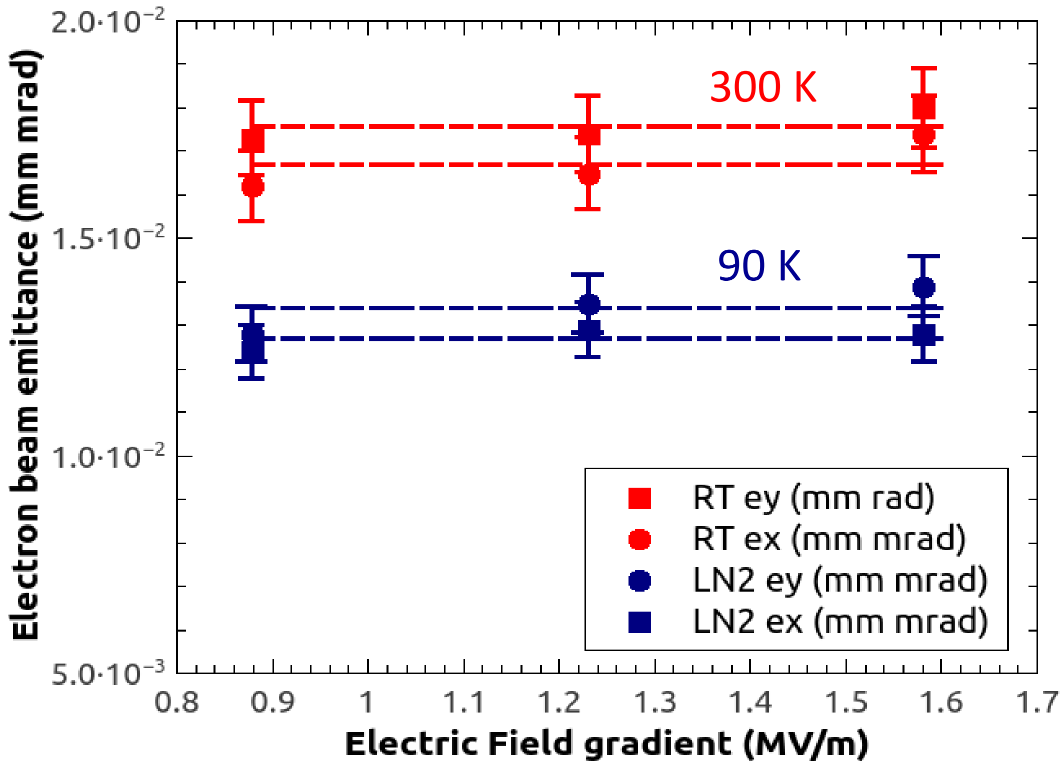
No space charge effects (yet).

Next:

- Bunch charge 10^2-10^3 × higher;
- acceleration to >100 keV with RF cavities.

Van Mourik et al., Struct. Dyn. 2014
Physics Today, July 2014

Cryo-cooled semiconductor cathode



Cs_3Sb photocathode

RMS laser spot size:

$\sigma_x = 60\ \mu\text{m},\ \sigma_y = 64\ \mu\text{m}$

$\varepsilon_n / \sigma_x = 0.2\ \mu\text{m} \cdot \text{rad/(mm rms)}$

- Thermal emittance @90 K 4× better than Cu cathode;
- emittance larger than expected – probably surface roughness;
- high bunch charge operation not yet demonstrated.

Cultrera et al., arXiv:1504.05920

Summary

- Field of Ultrafast Electron Diffraction developing rapidly
- 100 fs, single-shot UED of simple systems possible
- Relativistic UED will allow <10 fs temporal resolution
- Single-shot UED of macromolecules requires substantially higher source brightness
- Ultra-high-field-strength point-like source and/or ultracold source may provide required brightness