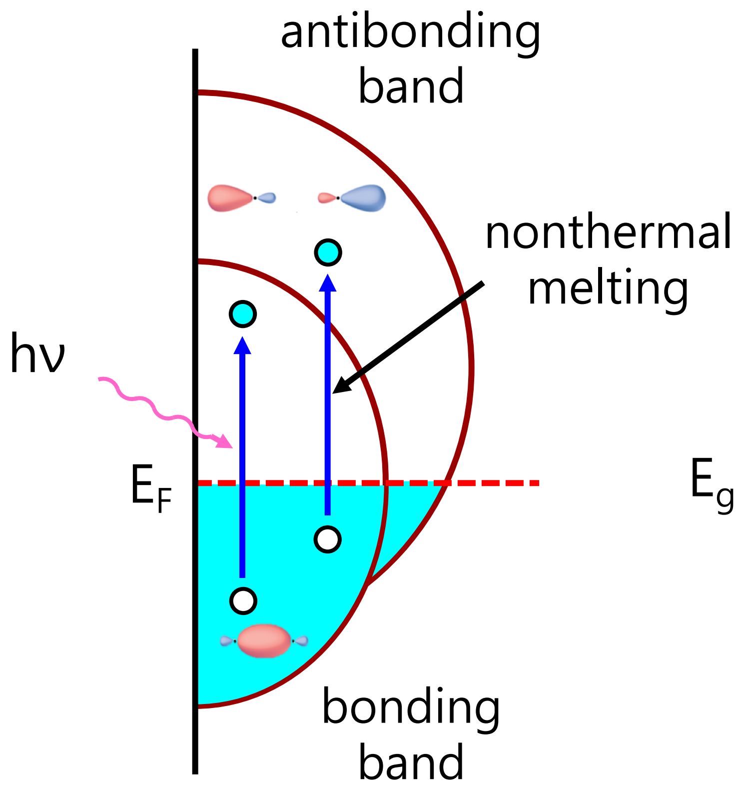
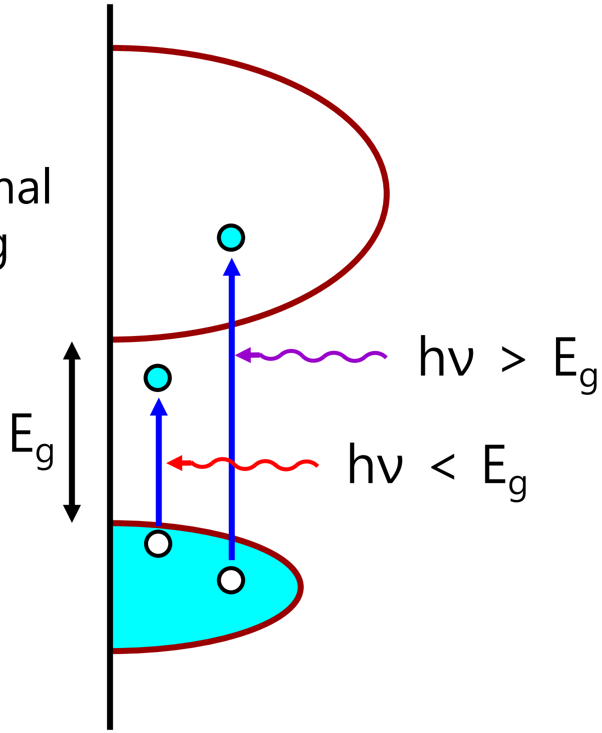
Ultrafast dynamics in solids, liquid, and gas





- Using pump-probe techniques

(1) diffraction (x-ray, electron)

(2) x-ray spectroscopies: XAS, XES, RIXS

(3) electron spectroscopies: ARPES, XPS, AES, EELS

(3) optical measurements: reflectivity, photoluminescence, MOKE

• • • • • •

- To investigate

(1) Order-melting dynamics: lattice, charge, spin, orbital, …

(2) Photo-induced phase transition

(3) Roles of intermediate states

• • • • • •

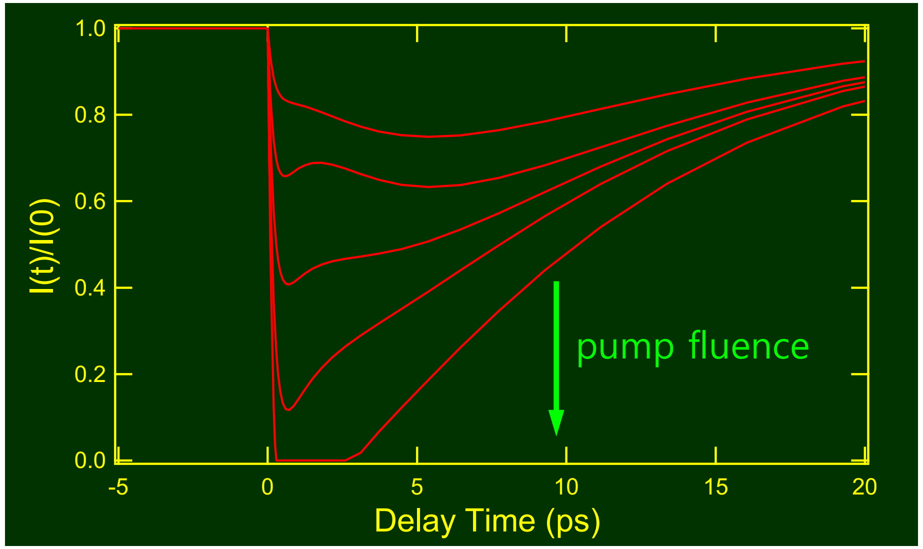
- Usual dynamics in solids

(1) fast order-melting/emerging by creation of e-h pairs

(2) fast order-recovering by electronic recombination

(3) slow order-melting by thermal processes

(4) slow order-recovering by cooling



Ultrafast Electron Diffraction (UED)

- De Broglie wavelength of a relativistic electron

$$\lambda = \frac{h}{p} = \frac{hc}{pc}$$

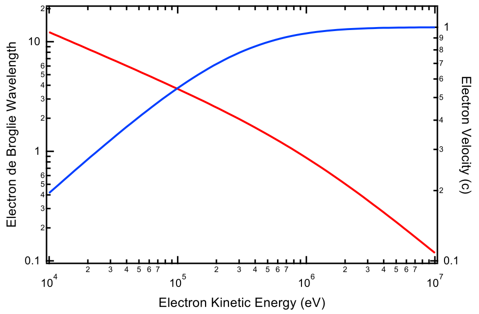
$$pc = \sqrt{(E_k + m_e c^2)^2 - (m_e c^2)^2}$$

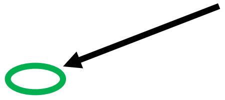
$$= \gamma \beta m_e c^2$$

$$hc = 1.24 \text{ MeV} \cdot \text{pm}$$

$$m_e c^2 = 0.51 \text{ MeV}$$

$$\beta = v/c, \ \gamma^{-1} = \sqrt{1 - \beta^2}$$

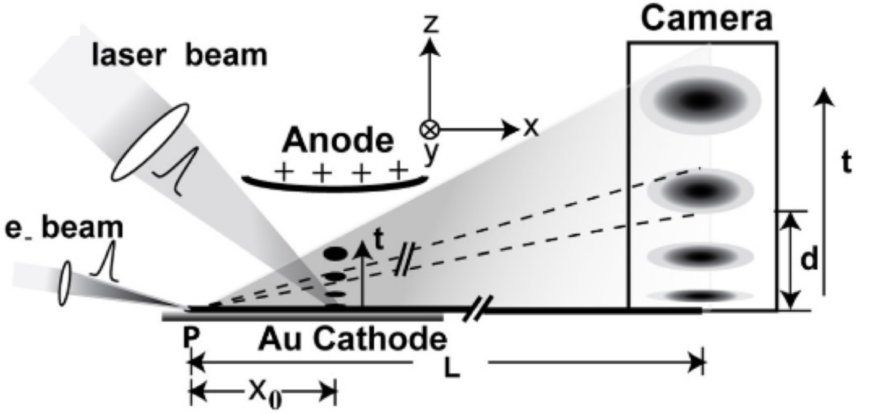


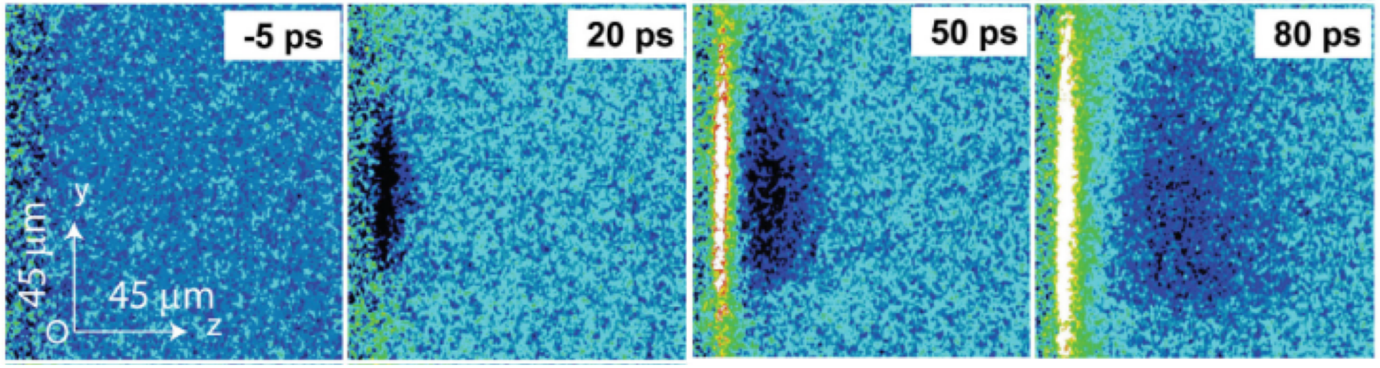


v/c ~ 0.99

1,000 times smaller than
typical unit cell size

- Space charge effects → poor temporal resolution





Z. Tao et al., J. Appl. Phys. **111**, 044316 (2012)

Why UED?

1. $10^4 - 10^6$ times larger scattering cross sections,
   good to study much smaller samples or gas & liquid.

2. Elastic mean-free-path is similar to optical pumping depth,
   good for pump-probe experiments.

3. 10^3 times less radiation damage per elastic scattering event.

4. Easily manipulated by EM lenses.

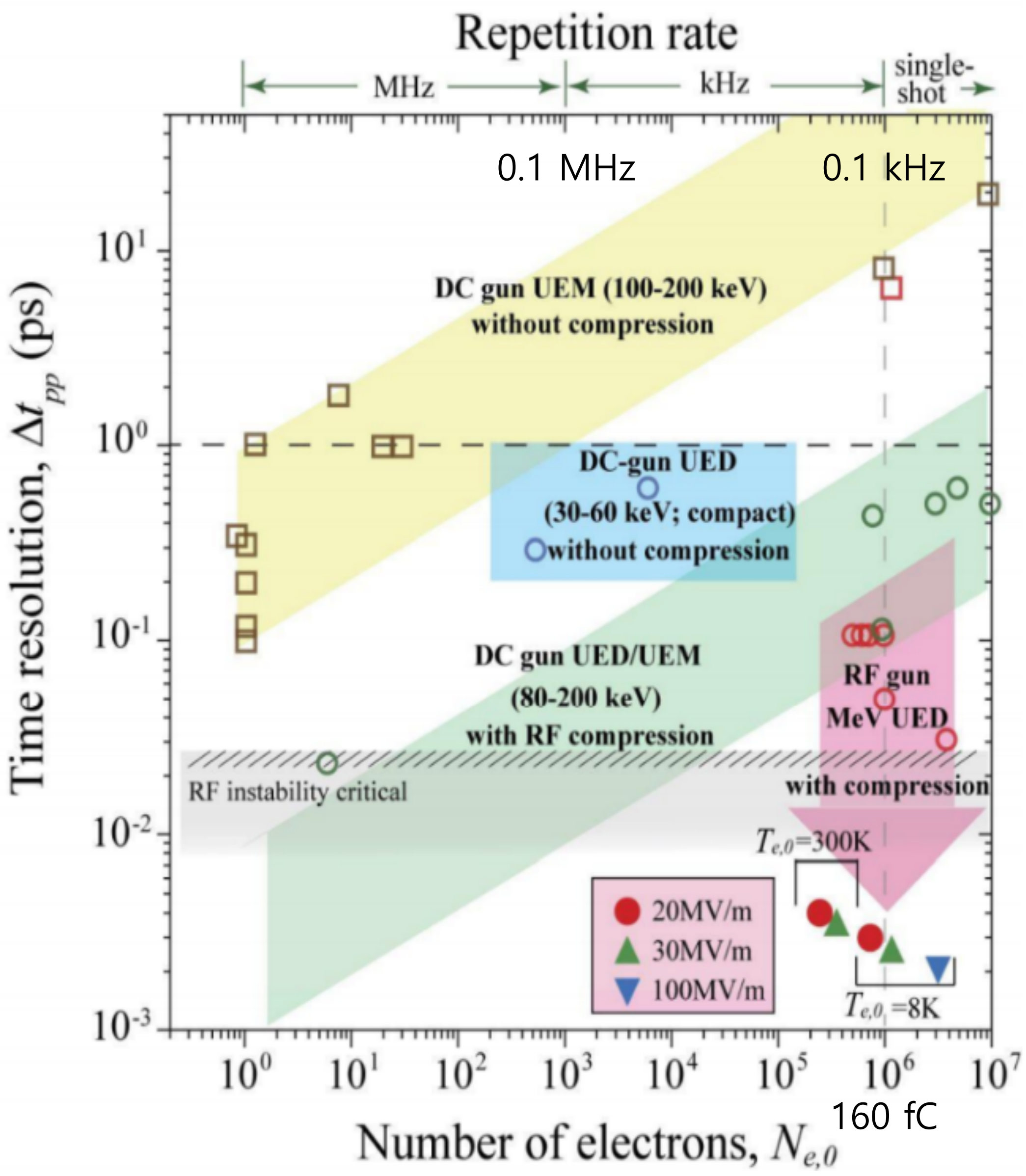
MeV UED vs keV UED

1. Minimize emittance and bunch length growth.

2. Naturally solve the velocity mismatch issue ($v_e < v_{ph}$).

3. RF bunch compress

→ Much better temporal resolution down to 30 fs
(500 fs with keV UED).

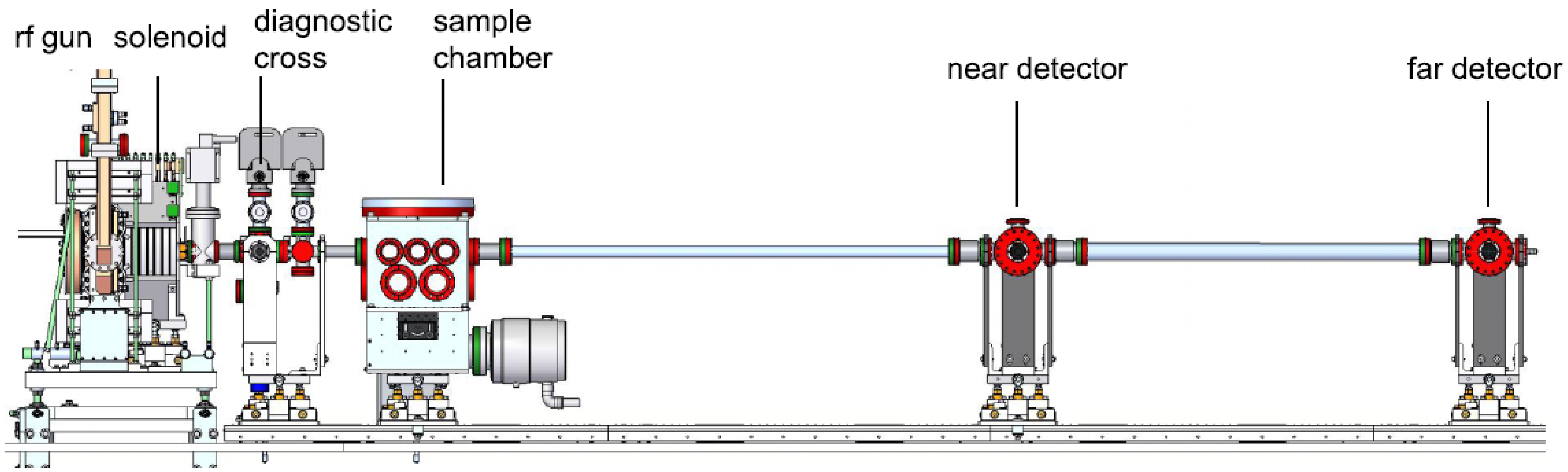


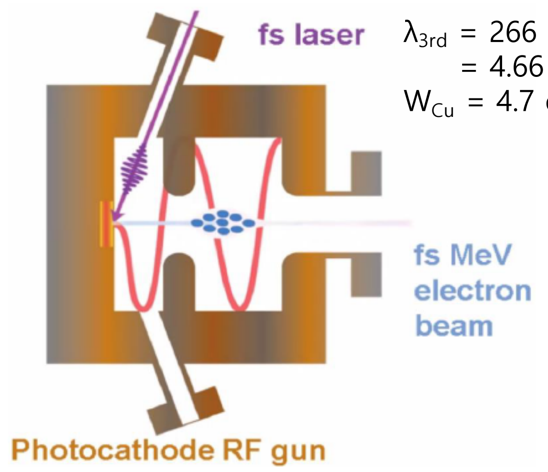
X. Sun et al., arXiv:2108.04860 (2021)

MeV UED @ SLAC

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

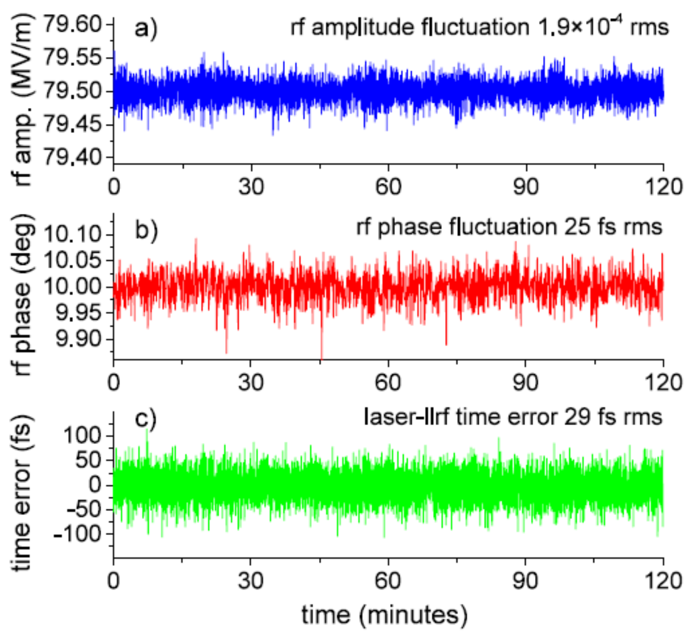
- Schematic of MeV UED beamline @ SLAC



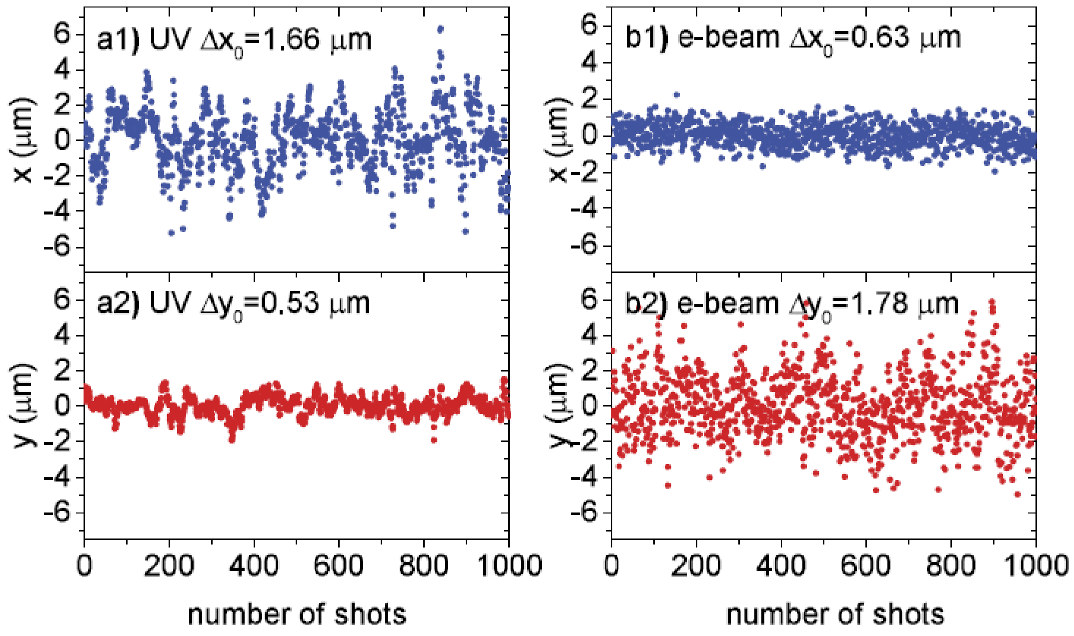


λ_{3rd} = 266 nm
= 4.66 eV
W_{Cu} = 4.7 eV

- RF quality



- UV & e-beam quality



MeV UED @ SLAC

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

- Sample chamber
  - Sample manipulator for translation & rotation
  - Sample holder for TEM grids or SiN windows
- Electron detector

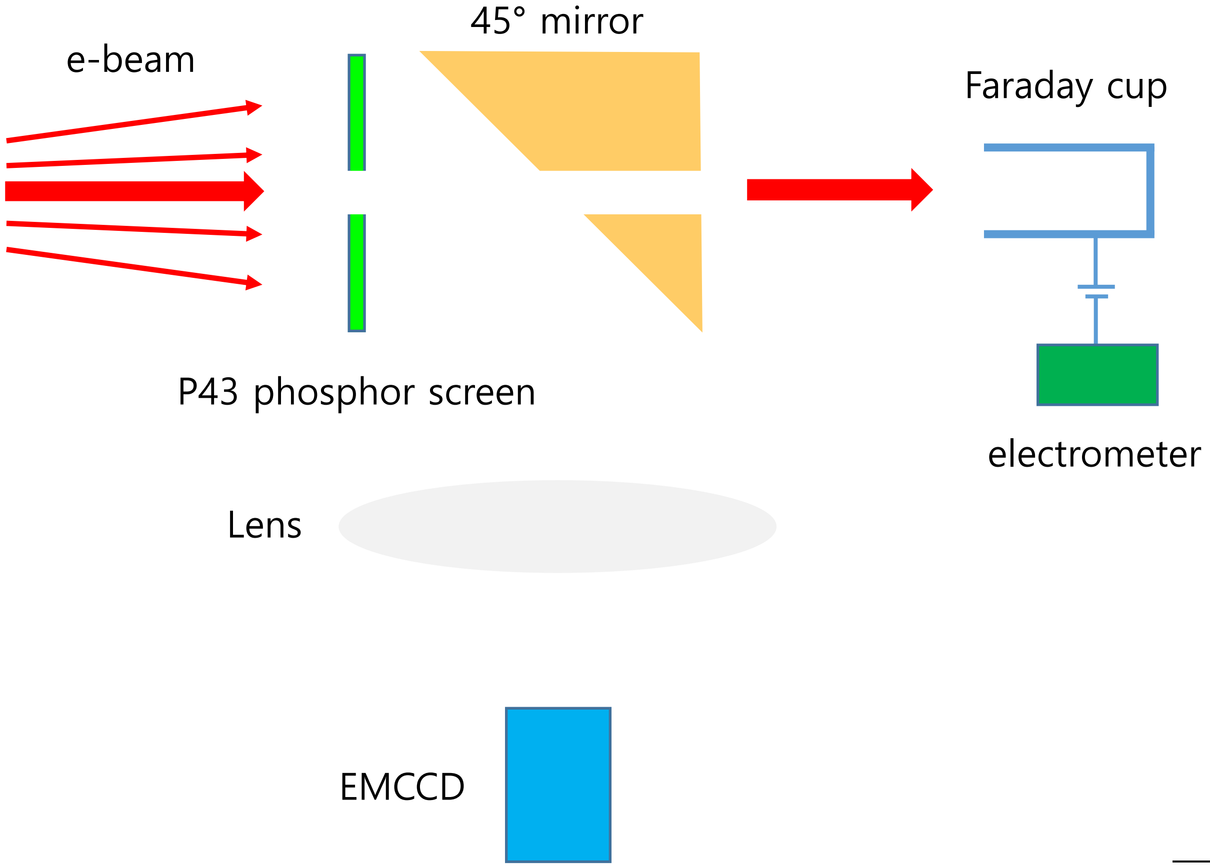


TABLE I. Typical machine and beam parameters of the MeV UED system.

Parameters	Values
Repetition rate	120 Hz
Gun gradient	79.5 MV/m
Launching phase	10°
Solenoid strength	0.314 kG-m
UV spot size, rms	40 μm
UV pulse duration, FWHM	60 fs
UV energy stability, rms	2.5%
Initial beam charge	75 fC
Intrinsic emittance	0.5 mrad
Collimator diameter	500 μm
Beam charge	60 fC
Beam size (diameter)	400 μm
Normalized emittance	18 nm-rad
Bunch length, rms	102 fs
Kinetic beam energy	3.68 MeV
Relative energy spread, rms	6.6×10^{-4}
IR pump spot size (diameter)	1.5 mm
IR pump pulse duration, FWHM	60 fs

1st Chamber: Cryo and Quantum Materials

Techniques

- Time resolution diffraction
- Momentum resolved scattering

Cryogenic Environment (upgrade)

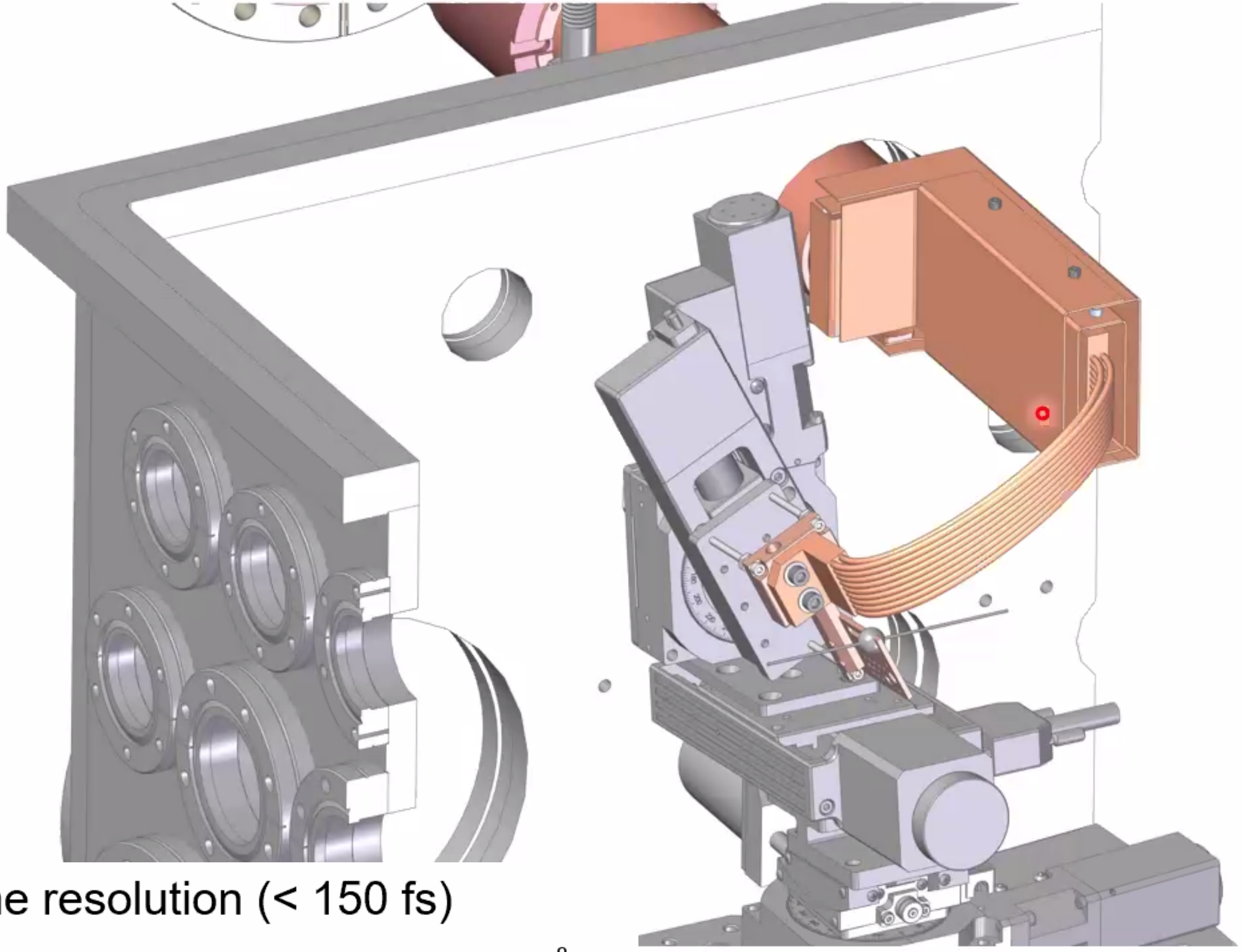
<20 K – 300 K *   *Temperature in the range 10 – 20 K may be achievable in certain circumstances, please discuss feasibility with UED staff

Sample Card

- <10 TEM style samples
- 6 axis motion (X,Y,Z, pitch, yaw, roll)

Laser

800, 400, 266 nm, OPA (UV – 2 um), THz*

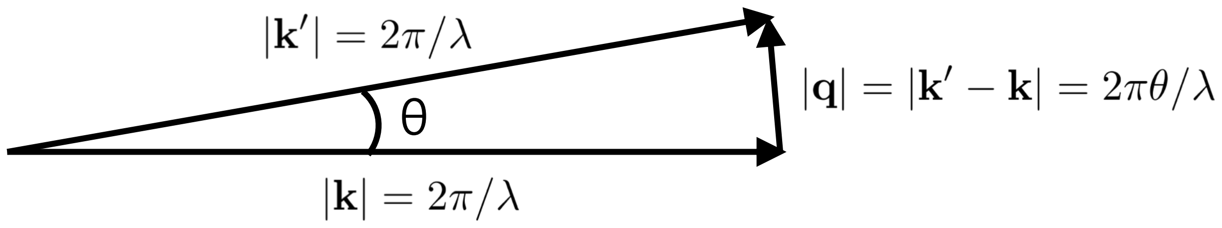


- Best time resolution (< 150 fs)
- Best momentum resolution (< 0.17$Å^{-1}$)

MeV-UED @ SLAC

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

- Reciprocal space resolution



q-resolution

$$\Delta q = 2\pi\frac{\Delta\theta}{\lambda} = 2\pi\frac{\Delta r/L}{\lambda} \approx 2\pi\frac{\sigma_\theta}{\lambda} = \frac{2\pi}{\lambda_C}\frac{\epsilon_n}{\sigma_x} \qquad (\epsilon_n = \gamma\beta\sigma_x\sigma_\theta)$$

Δr : rms width of the diffraction spot

L : distance between sample & detector

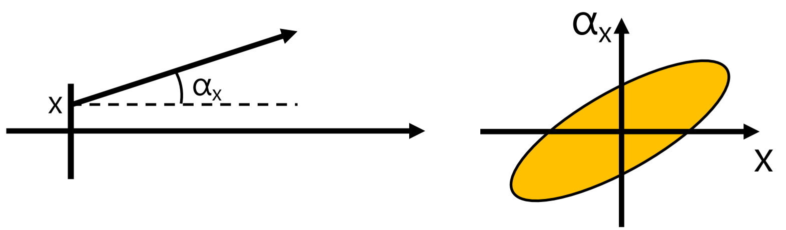
σ_θ : beam divergence at the sample

σ_x : beam size at the sample

ε_n : normalized emittance

λ_C : Compton wavelength

Emittance ε = (area of particle distribution in x and α_x)

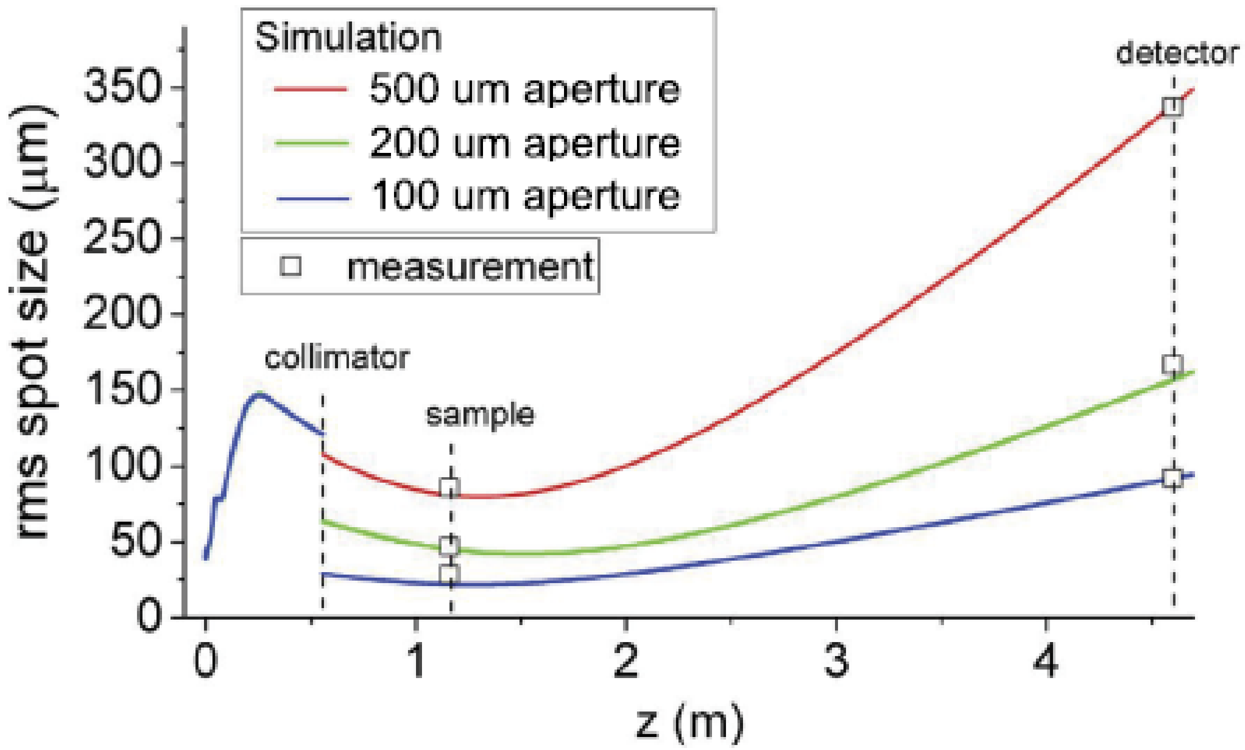


q-resolution is determined by ε_n when σ_x is fixed.

MeV UED @ SLAC

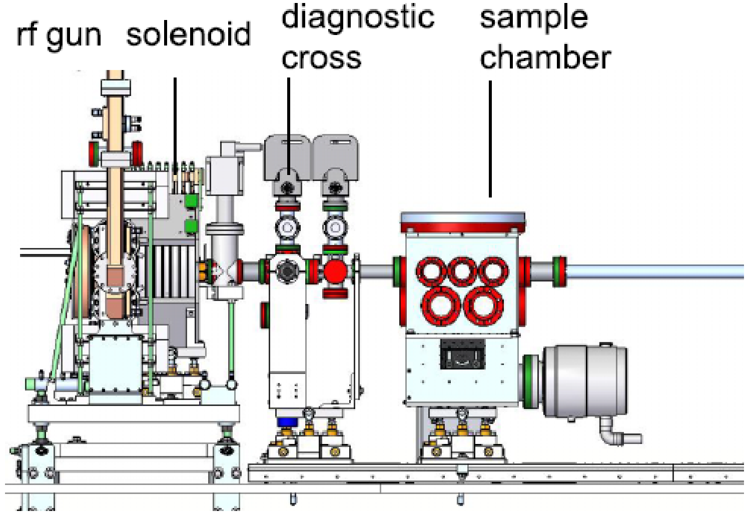
S. P. Weathersby et al., Rev. Sci. Instrum. 86, 073702 (2015)

- Beam size & normalized emittance control varying collimator diameter



Collimator diameter (μm)	measured Q (fC)	measured σ_x (μm)	simulated ϵ_n (nm-rad)	calculated Δs (Å $^{-1}$)
500	61	86	18	0.054
200	11	47	7.1	0.039
100	1.4	29	2.1	0.019

The solenoid is the only focusing element and is tuned to deliver the sharpest diffraction features to the detector.



※ Phosphor screen resolution

$$\Delta q = 2\pi \frac{\Delta r / L}{\lambda} = 2\pi \frac{10^{-4}/3}{0.33 \times 10^{-2}} \text{ Å}^{-1} \approx 0.06 \text{ Å}^{-1}$$

perovskite a = 4 Å → k_{BZ} = π/a = 0.8 Å^{-1}

MeV UED @ SLAC

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

- Temporal resolution

$$\tau = \sqrt{\tau_e^2 + \tau_{ph}^2 + \tau_{\mathrm{TOA}}^2 + \tau_{\mathrm{VM}}^2}$$

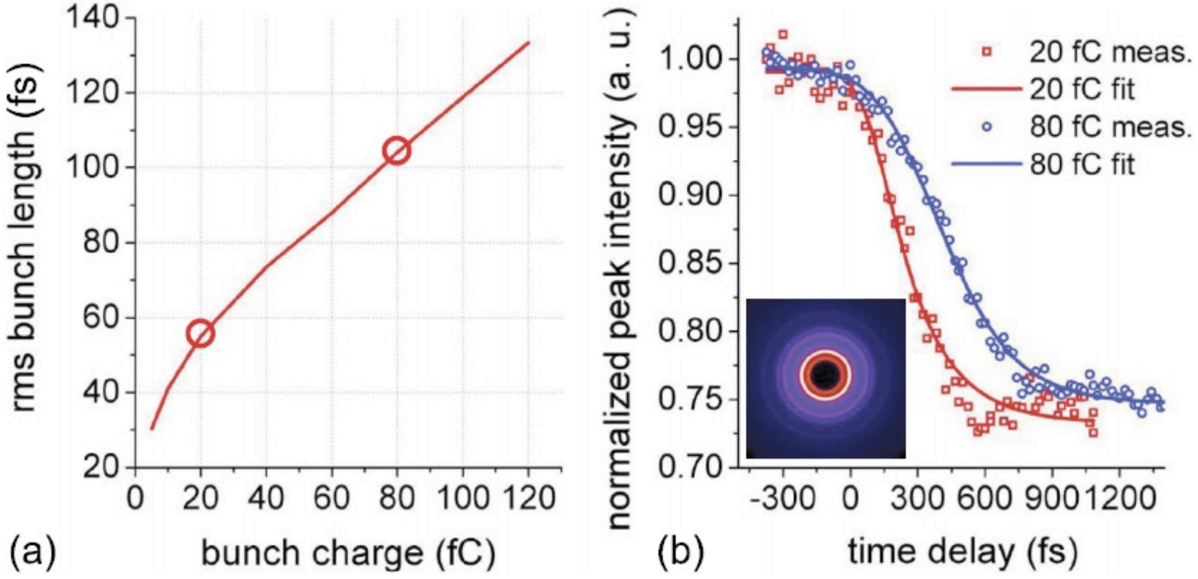
τ_e : pulse duration of the probe pulse → 10° launching phase for shortest bunch length at the sample

τ_{ph} : pulse duration of the pump pulse

τ_{TOA} : time-of-arrival jitter between the pump and probe pulses < 50 fs (slide 3, rf quality)

τ_{VM} : velocity mismatch (due to intersection angle between pump and probe pulses)

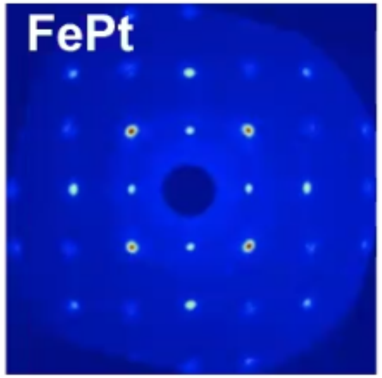
From the intensity of the (410) ring of 25 nm thick Bi(111)

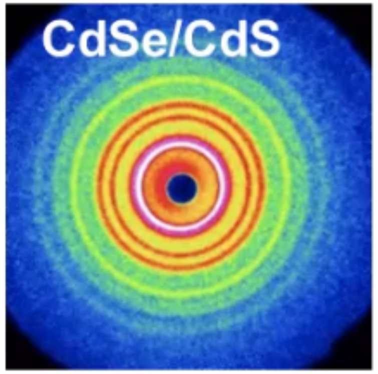


MeV UED @ SLAC

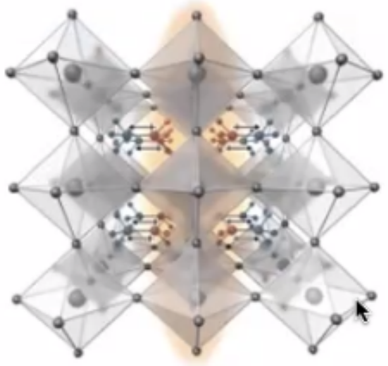
S. P. Weathersby et al., Rev. Sci. Instrum. 86, 073702 (2015)

- UED Gallery

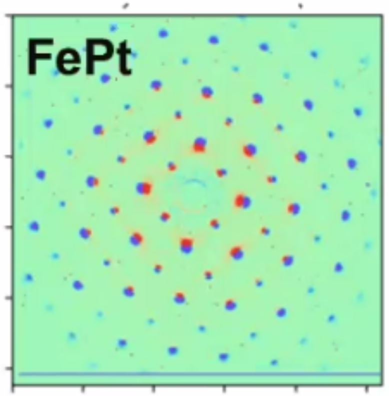


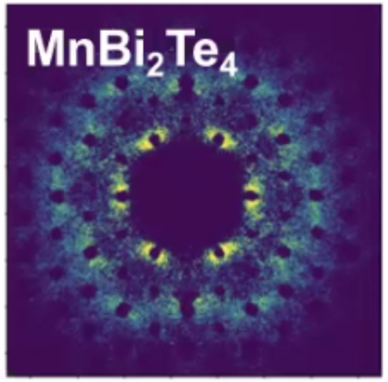


Nano materials

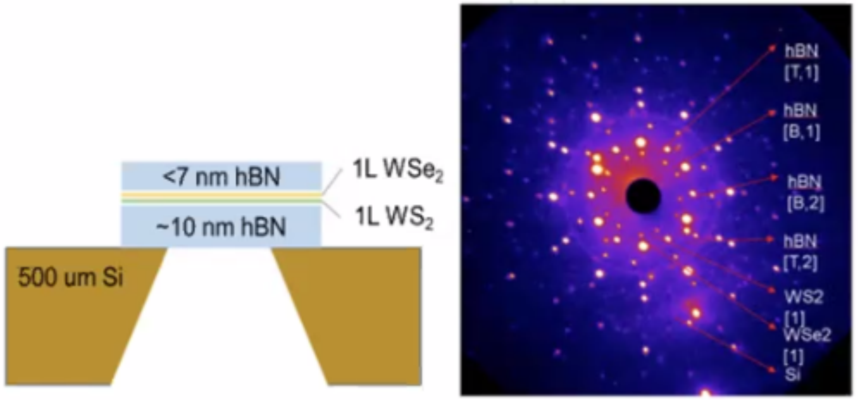


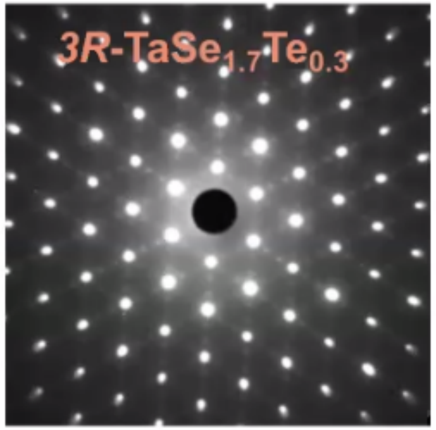
Materials for photovoltaic

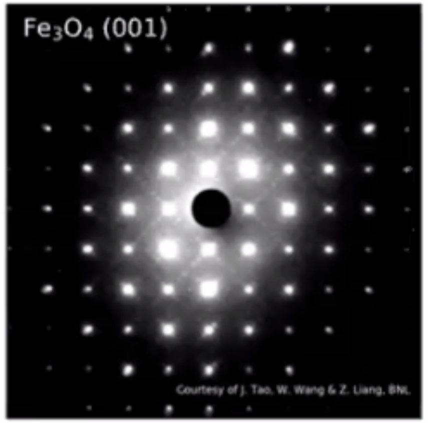


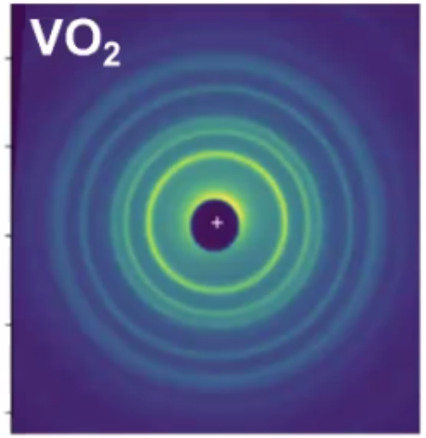


Diffuse scattering



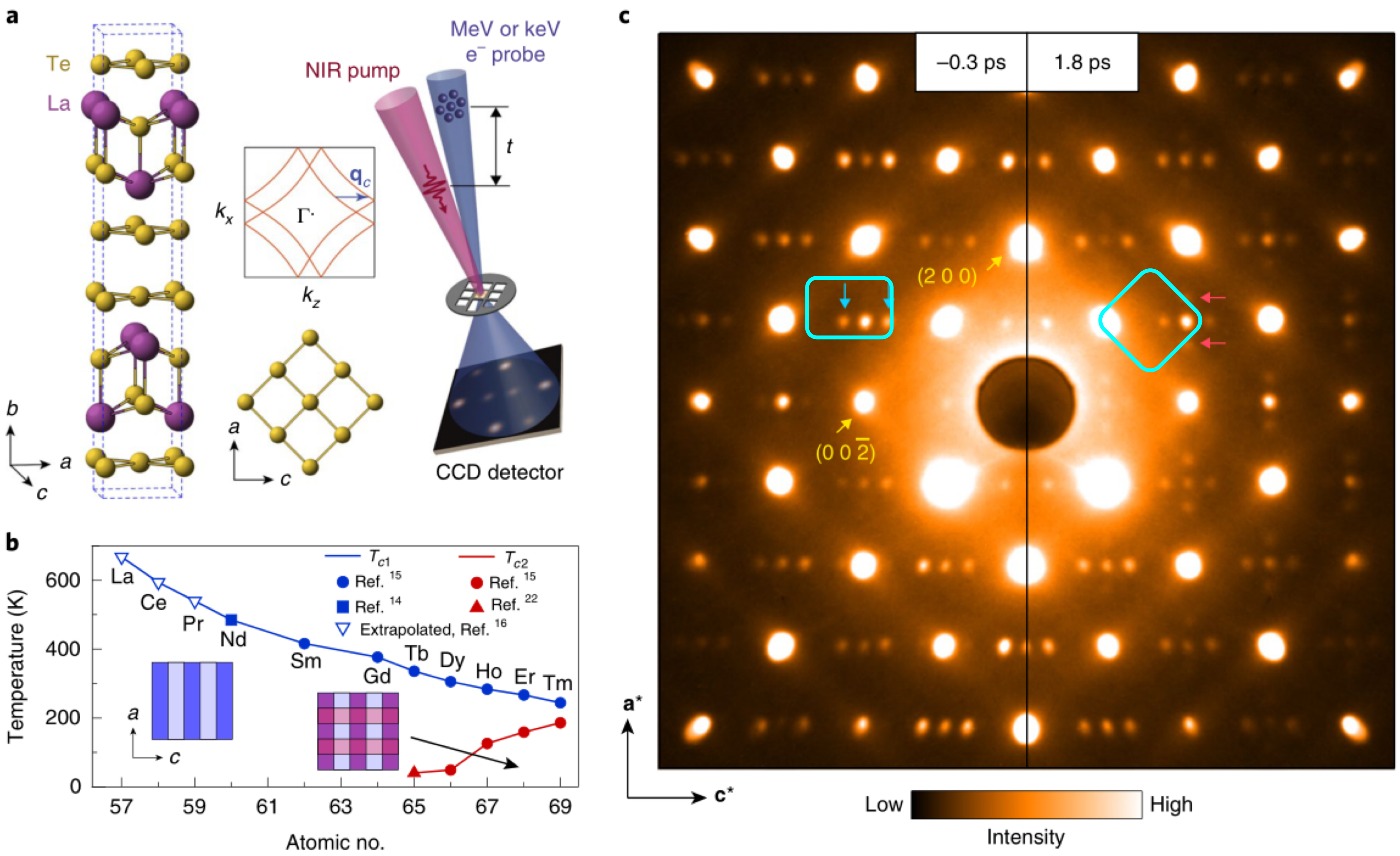




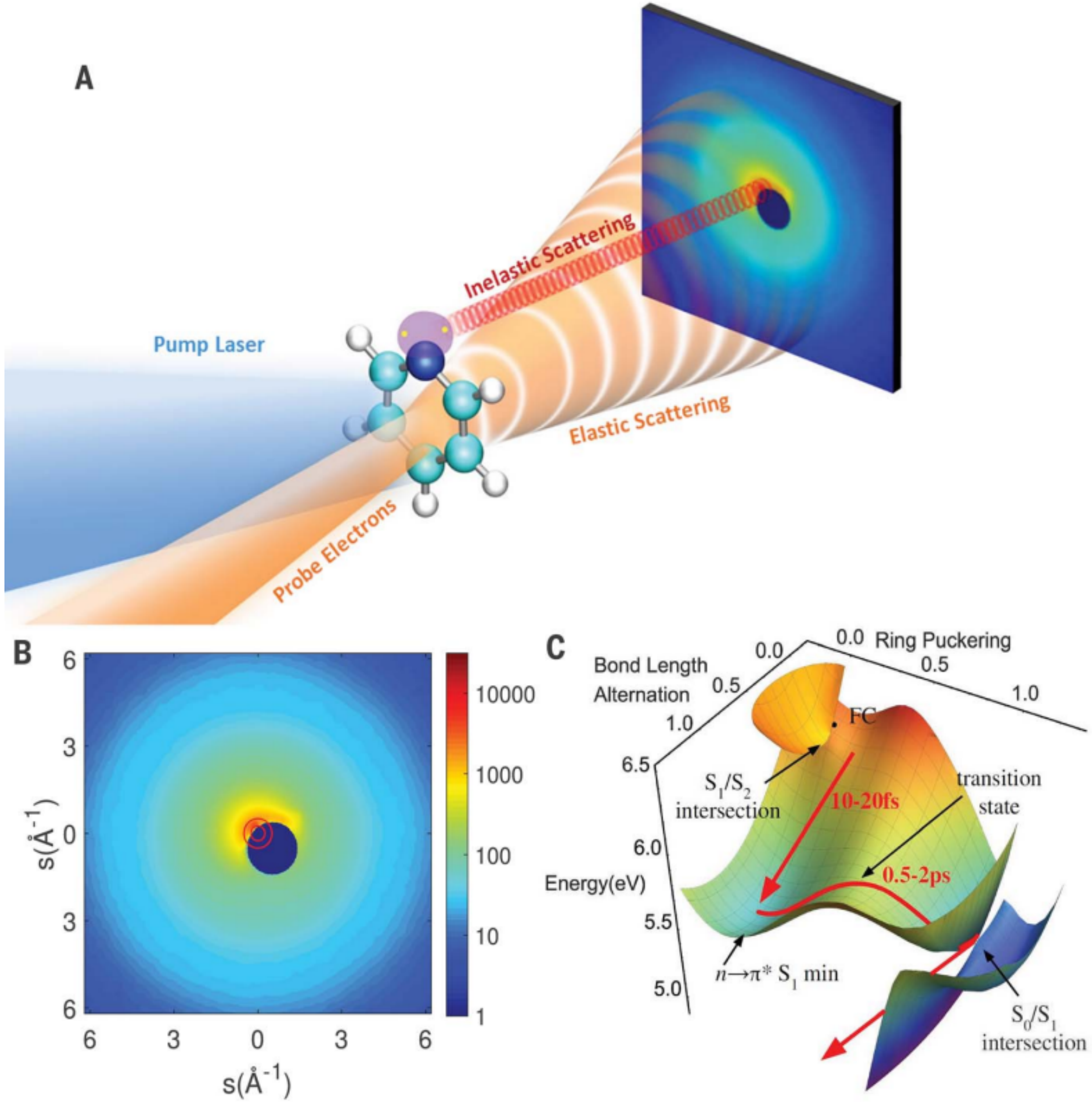


Light-induced charge density wave in $LaTe_3$

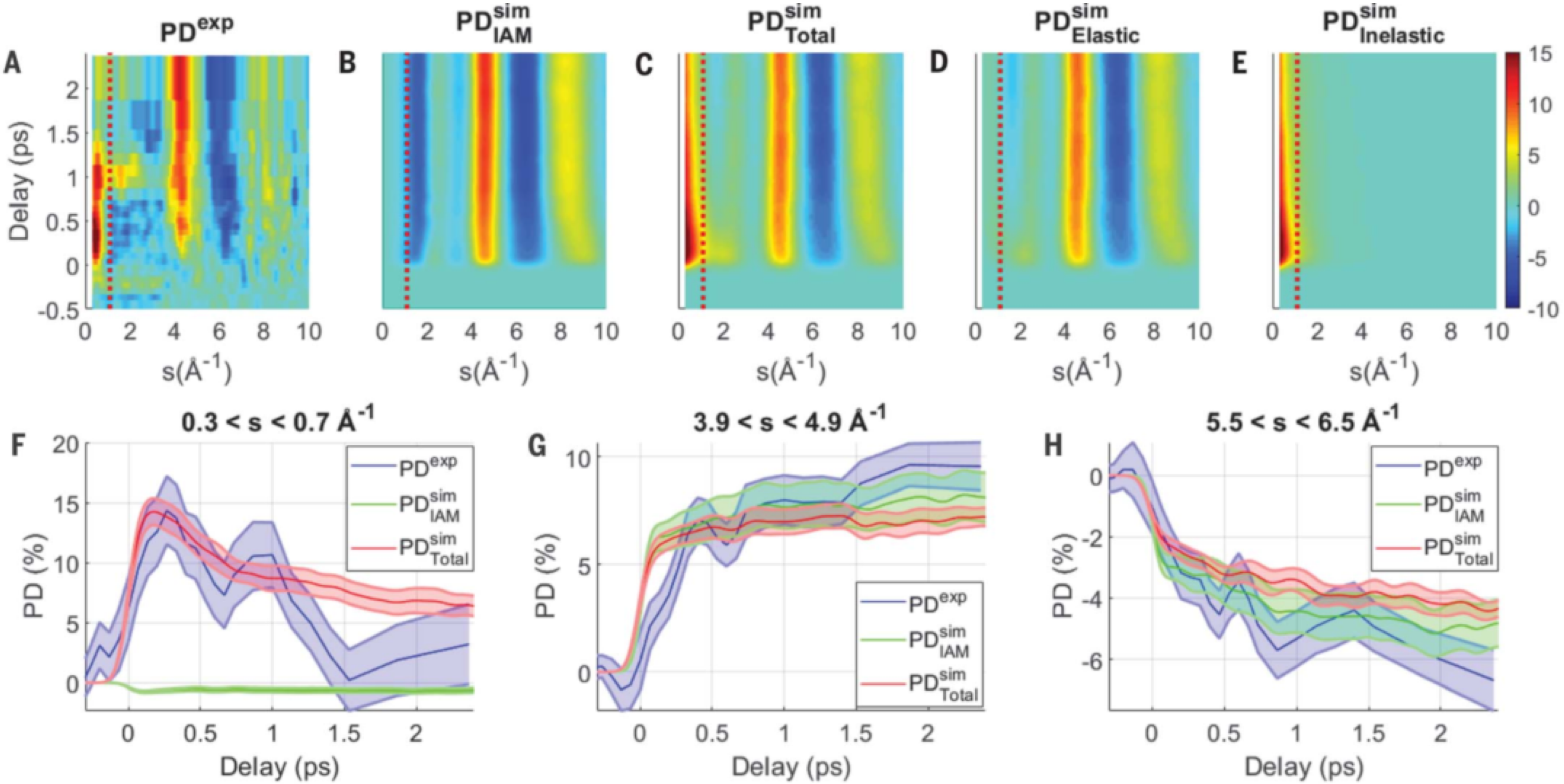
Gedik group, Nat. Phys. **16**, 159 (2020)



Simultaneous observation of nuclear and electronic dynamics by UED Science **368**, 885 (2020)

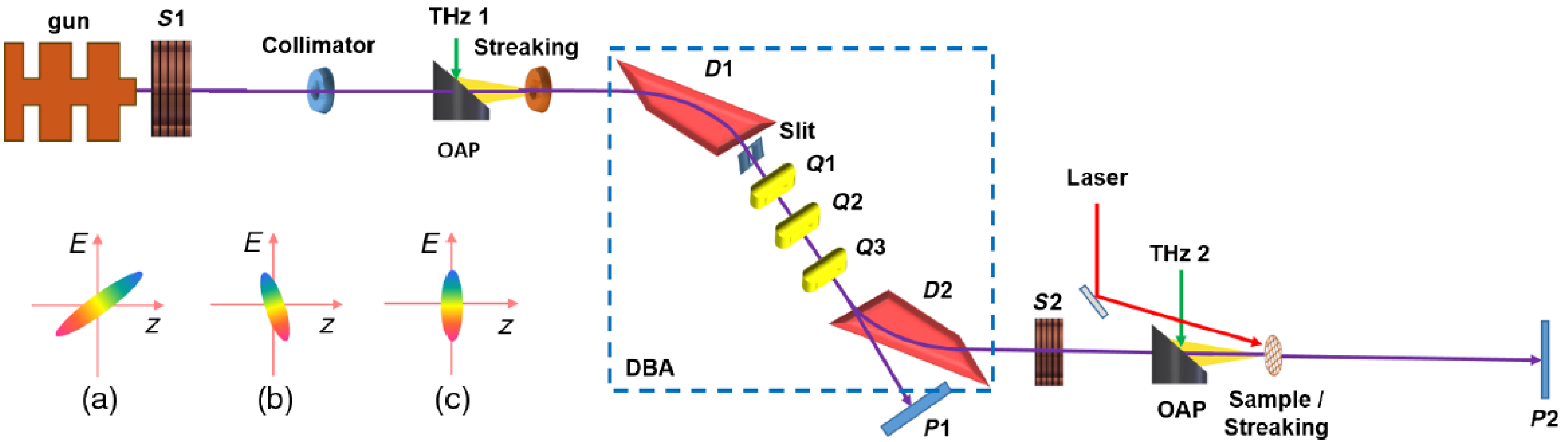


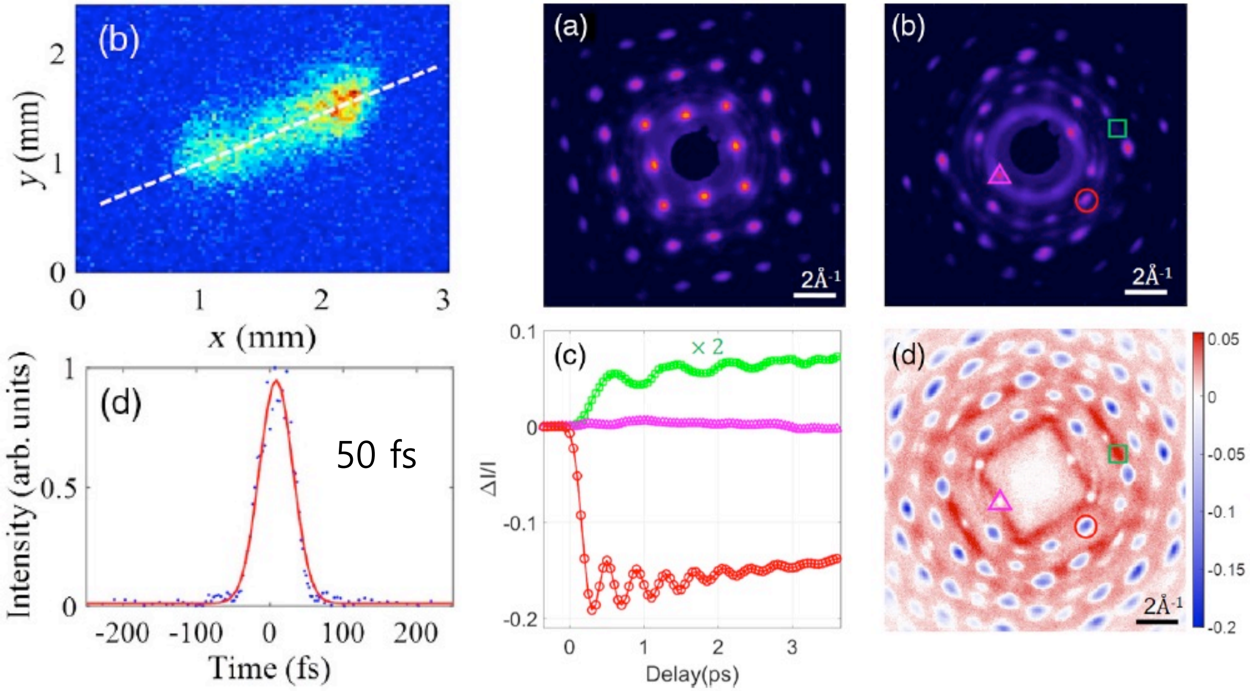
Simultaneous observation of nuclear and electronic dynamics by UED Science **368**, 885 (2020)



Toward better temporal resolution

上海交通大學, Phys. Rev. Lett. **124**, 134803 (2020)

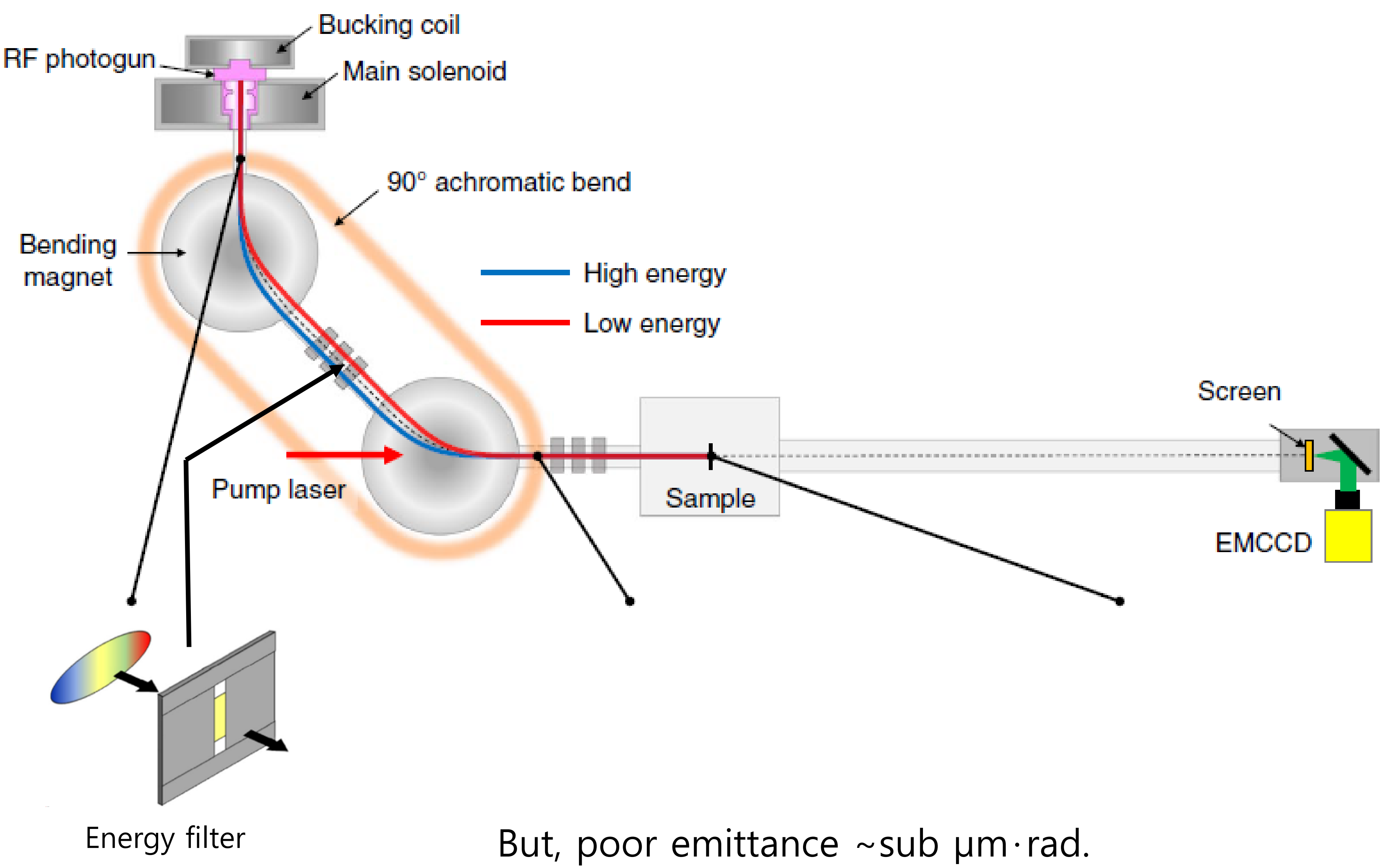


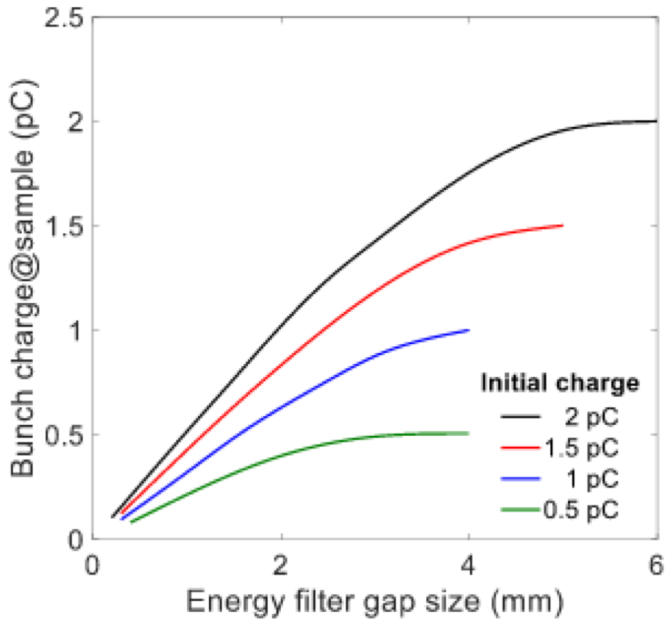


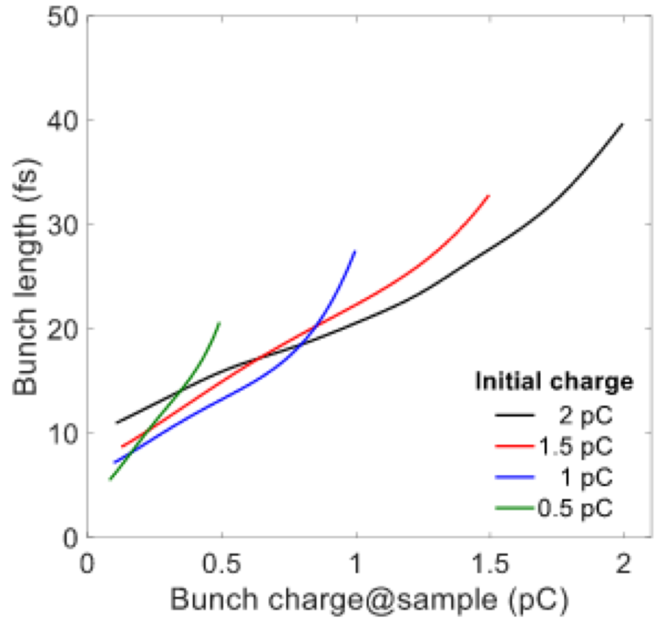
No considerable degradation of beam emittance and jitter.

Toward better temporal resolution

한국원자력연구원, Nat. Photon. **14**, 245 (2020); Struct. Dyn. **7**, 034301 (2020)







But, poor emittance ~sub μm·rad.