MeV ultrafast electron diffraction and microscopy for the scientific frontier

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The quest of resolution



Galaxy by Hubble Space Telescope





Molecule by super resolution fluorescence microscopy

Spatial resolution: imaging and diffraction



 λ : wavelength of incident beam $n \cdot \sin(\alpha)$: Numerical Aperture

> **Resolution:** $\lambda/2$ (~200 nm for optical light)





Crystallography

Diffraction





Coherent diffraction imaging

Temporal resolution: pump-probe technique



- A 'pump' pulse drives the system out of equilibrium state to initiate a dynamic process
- A 'probe' pulse measures the transient state following the excitation at a given delay time
- Changing time delay to map out the whole process by which the system relaxes back
- Pulsed x-ray or electron 'probe' beams provide both high spatial and temporal resolution
- Ultrafast Electron Diffraction (UED): laser-pump electron-probe technique

Ultrafast electron diffraction (UED): complementary to FELs



UED application: phase transition



UED temporal resolution



Electron beam pulse width limited by space charge effect





APL 83, 1044 (2003)

UED and UEM go relativistic





- *v* = 0.99*c* for 3 MeV electron beam
- The electric force and magnetic force largely cancel out
- MeV beam from a photocathode rf gun
- High acceleration gradient increases beam brightness
- High acceleration gradient reduces cost and size





- Introduction
- MeV UED and UEM developments
- Breaking ~50 fs resolution barrier
- Summary

Accelerator-based MeV UED: world-wide efforts



UCLA



















- First demonstration of MeV UED
- Single-shot diffraction obtained with a 5.4 MeV / 3 pC /500 fs beam
- No collimator to improve S/N ratio

APL 89, 184109 (2006)

UCLA, 2010





- For the first time MeV UED is used in a pump-probe experiment for studies of ultrafast dynamics
- Laser-induced Debye-Waller effect in Gold
- Collimator to improve S/N ratio
 APL 97, 063502 (2010)



Osaka U, 2011



Compact diffractometer



Ultrafast electron microscope

APL 98, 251903 (2011) APL 103, 253107 (2013)

Microscopy 67, 291 (2018)

DESY/MPSD, 2015







- Static diffraction obtained
- Buncher and deflector to improve resolution

Faraday Discuss., 177, 467 (2015)

BNL



NJP 17, 063004 (2015); Sci. Adv., 4: ap7427 (2018)

Separate optical and acoustic phonons

SLAC



- State-of-the-art
- 180 Hz / 150 fs resolution (FWHM)
- Optical to THz pump pulse
- Solid / gas sample
- 30 K 400 K temperature control
- 5-axis sample manipulation
- Now a DOE user facility



RSI 86, 073702 (2015); PRL 117, 153002 (2016); Science 360, 1451 (2018) / 361, 64 (2018); Nature 565, 61 (2019)

SJTU



3 MeV ultrafast electron diffraction and microscope

SJTU



High quality electron diffraction





Single-shot with ~100 fs beam

Diffuse scattering with ~10¹⁰ electrons



data

data fitted curv

 $10 \times I_{CDW}$

fitted curv





LBNL: High Repetition Rate Electron Scattering beamline (HiRES)



Electron beam energy	700-900 keV
Repetition rate	1-10 ⁶ Hz
Temporal resolution	200 fs-1000 ps
Electrons per pulse	1-10 ⁸
Relative energy spread	10-3 10-4







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break ~50 fs resolution barrier

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

rf compression + time stamping

Produce short beam and correct timing jitter

Compression with space charge force Produce short beam with small timing jitter





SJTU, 3 MeV ~200 fs -> 6 fs



- C-band buncher
- C-band deflector (2 MV)

Measure and sort



Achieving few-femtosecond time-sorting at hard X-ray free-electron lasers



7 papers published in Nature Photonics

THz deflector



- THz pulse in tight synchronization with laser
- Time-dependent angular kick
- E-field is not in phase with B-field in the gap
- Field enhancement in the narrow gap

Sci Rep 4, 5645 (2014)

THz streaking at SLAC



- 4 mm X 50 µm slit
- Single oscillation

PRAB 22, 012803 (2019)

THz streaking at SJTU





Deflection vs time delay

- 250 µm X 10 µm slit
- multipole oscillation
- 6 fs beam with jitter corrected with 1.5 fs accuracy

PRX 8, 021061 (2018)

THz oscilloscope for recording time information



- Small aperture
- Limited dynamic range

PRL 122, 144801 (2019) (Editors' Suggestion and Featured in Physics)

Large aperture + large dynamic range

break ~50 fs resolution barrier

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

rf compression + time stamping

Produce short beam and correct timing jitter

• Compression with space charge force

Produce short beam with small timing jitter

Compression with space charge force



- Test beam in the middle of two drive beams
- Negative energy chirp
- Centroid energy will not change
- Test beam will be compressed with negligible timing jitter
- The front drive beam pushes the test beam backward
- The back drive beam pushes the test beam forward
- The test beam stays in the middle with its pulse width reduced

PRL 120, 044801 (2018) (Editors' Suggestion and Featured in Physics)

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PRL 120, 044801 (2018) (Editors' Suggestion and Featured in Physics)





Space charge force induces positive energy chirp without changing beam centroid energy



- Isochronous: R₅₆¹+ R₅₆²+ R₅₆³=0
- Gun amplitude jitter won't convert to arrival timing jitter
- Space charge induced positive energy chirp leads to bunch compression (R₅₆²+ R₅₆³<0)
- Jitter free bunch compression

~30 fC beam compressed to ~30 fs (FWHM) with <5 fs timing jitter



Manuscript in preparation

KAERI



SJTU



Before installation

New DBA beamline



THz source for measuring electron bunch length and jitter



THz-electron interaction chamber









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Summary

- Growing interest in using MeV UED/UEM to solve the grand challenges in probing matter at ultrafast temporal and ultrasmall spatial scales
- Compact facility, yet with rich physics and great potential
- Potential of MeV UED/UEM fully demonstrated
- Still room to improve and lots of possibilities to explore

Thanks!