SLAC MeV UED Experience

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UED Opportunities for
Dynamical Imaging of Materials

Santa Fe, NM Nov. 6th-8th, 2023

Outline

- Brief Introduction to MeV UED
- SLAC MeV UED Experimental Setup and Science Highlights
- Perspectives and Comments

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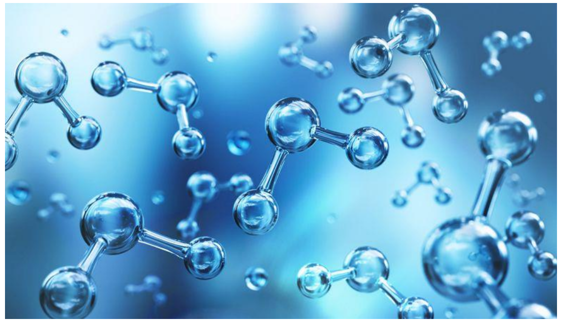
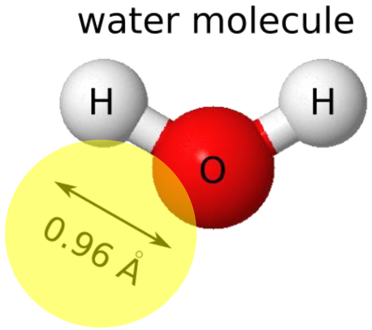
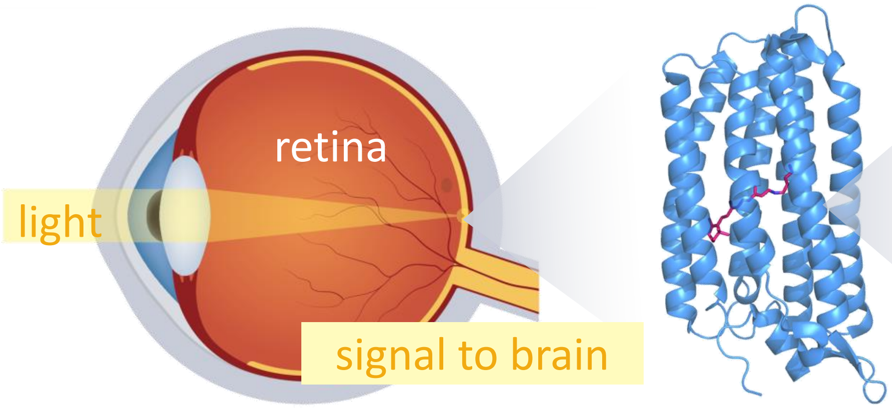
Probing the *ultrasmall* and *ultrafast*

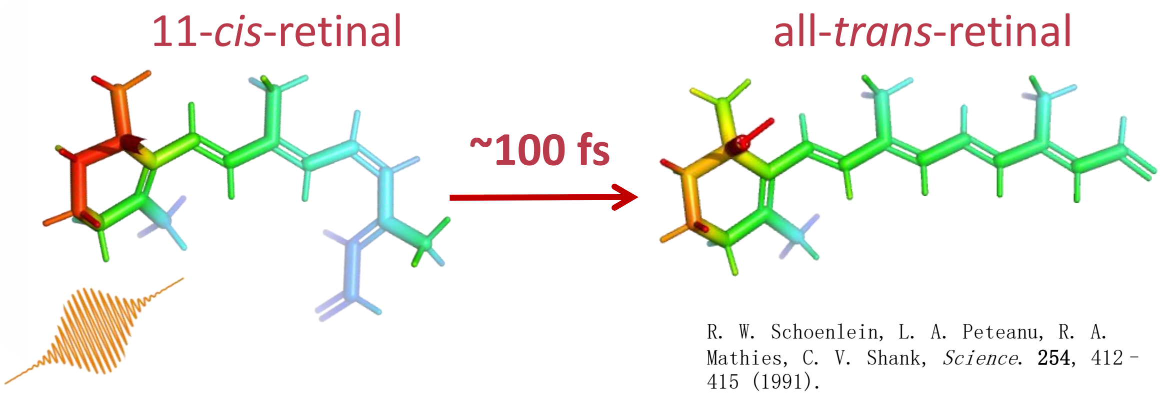
Atomic spatial-temporal scales

- Angstrom (Å, 10^{-10} m)
- femtosecond (fs, 10^{-15} s)







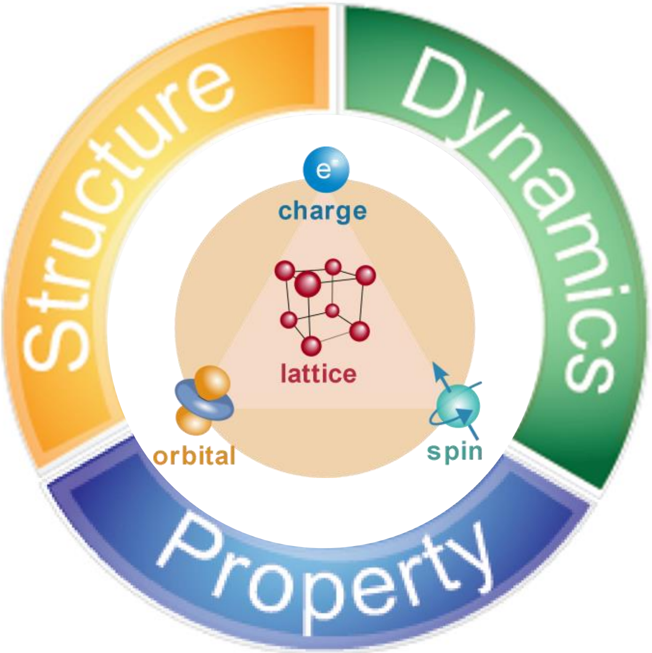


R. W. Schoenlein, L. A. Peteanu, R. A. Mathies, C. V. Shank, *Science*. **254**, 412–415 (1991).

Overall vision process occurs on **0.1 s** time scale

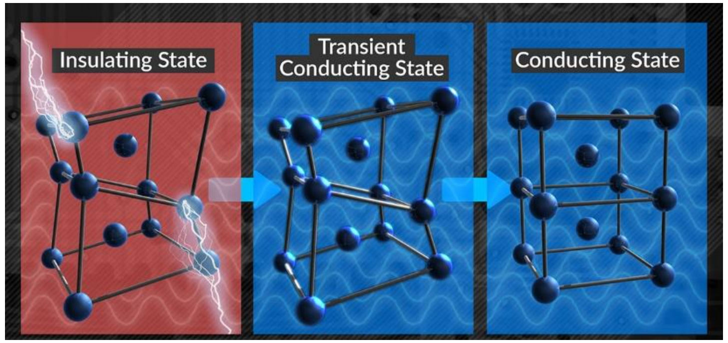
The 1st step of vision process happens over **~100 fs** time scale

Probing the *ultrasmall* and *ultrafast*



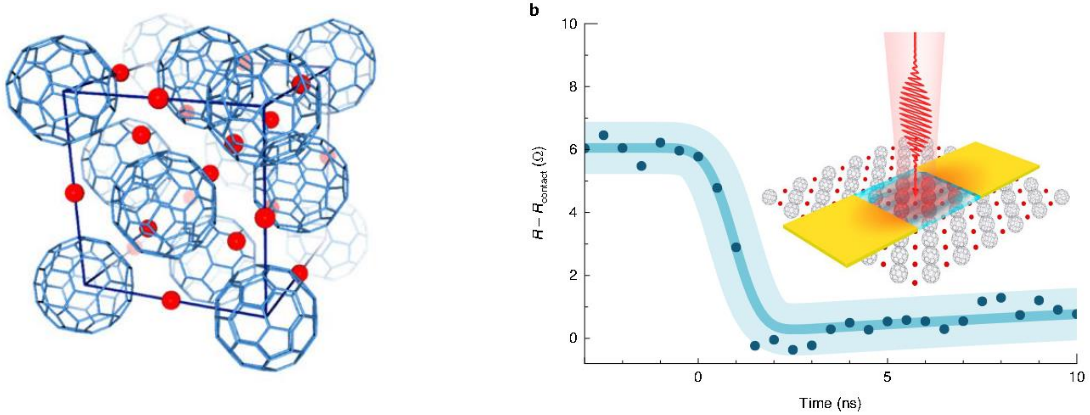
- Atomic structure and phase (function) correlation
- Dynamical process can lead to exotic properties not accessible in thermal equilibrium
- Need **fs-Å** probe to study the correlation

VO_2 insulator-metal phase transition



A. Sood *et al.*, *Science*. **373**, 352–355 (2021).

Room temperature superconductor does exist! – in a transient state over ps-ns scale for K_3C_{60}

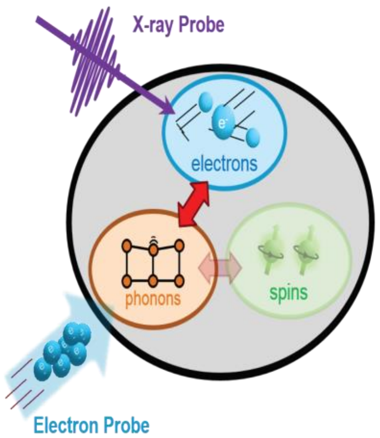


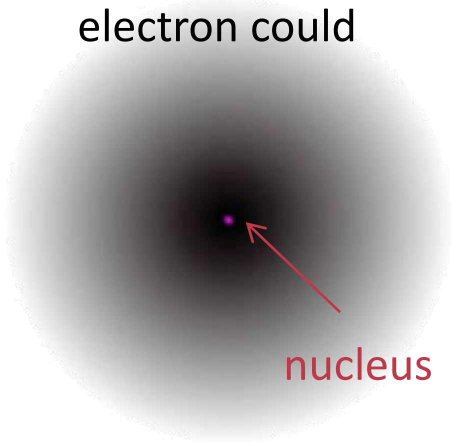
M. Budden *et al.*, *Nat. Phys.* **17**, 611–618 (2021).
E. Rowe *et al.*, *Nat. Phys.* (2023), doi:10.1038/s41567-023-02235-9.

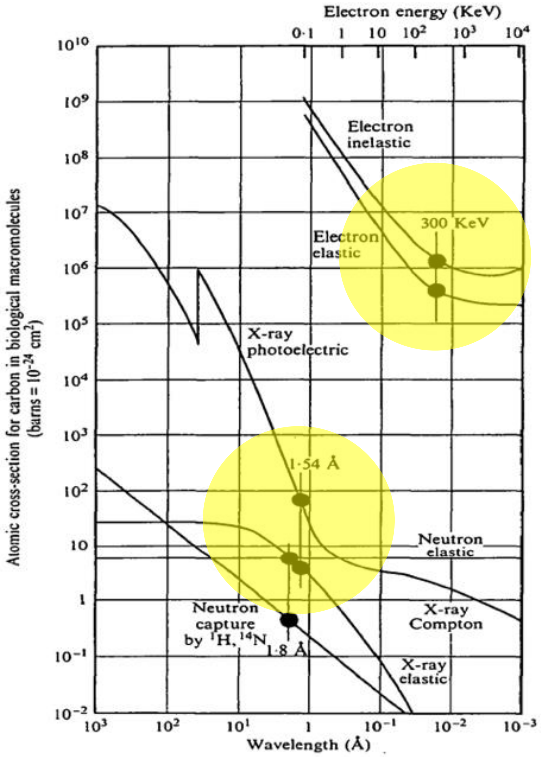
Ultrafast probes – *X-ray* and *Electron*

Unique features of electron compared to x-ray

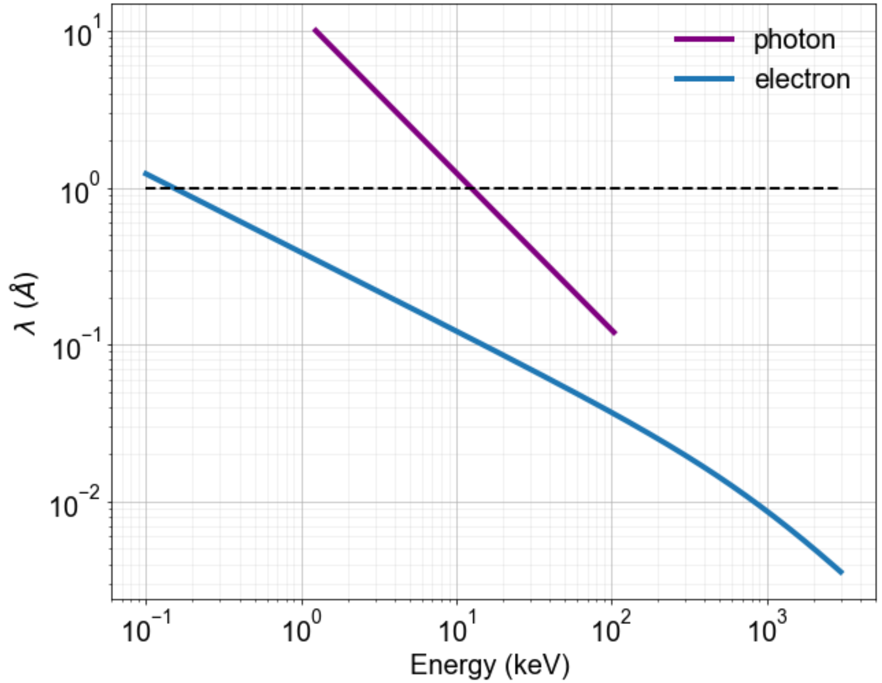
- $10^4 - 10^6$ times larger scattering cross sections
- shorter wavelength, higher spatial resolution
- 10^3 times less radiation damage



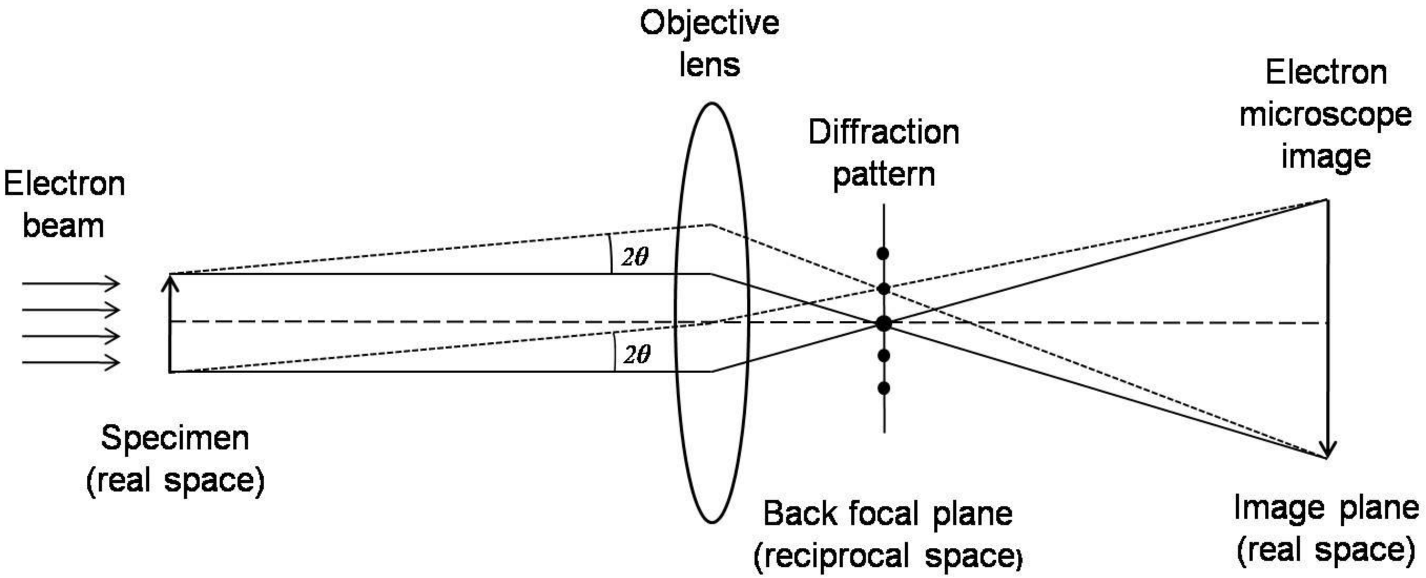


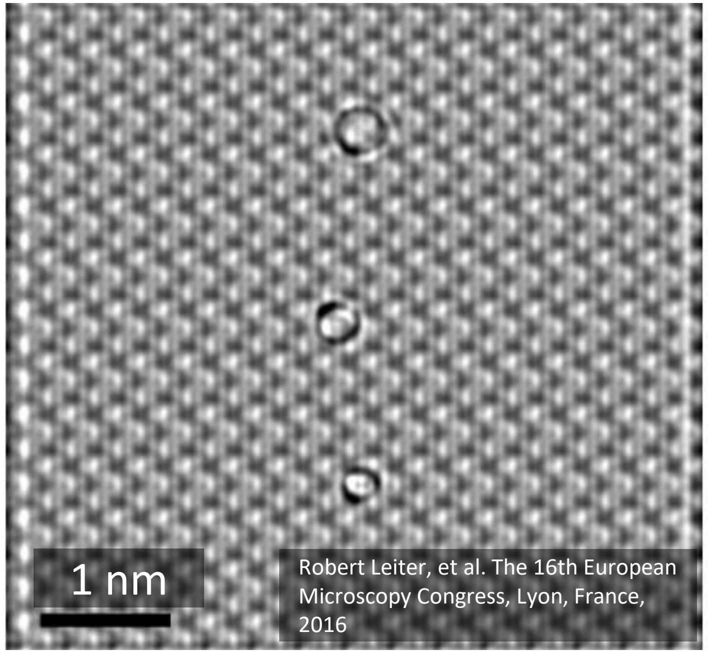


R. Henderson, *Q. Rev. Biophys*. **28**, 171 (1995).



Real space vs diffractive (reciprocal space) imaging



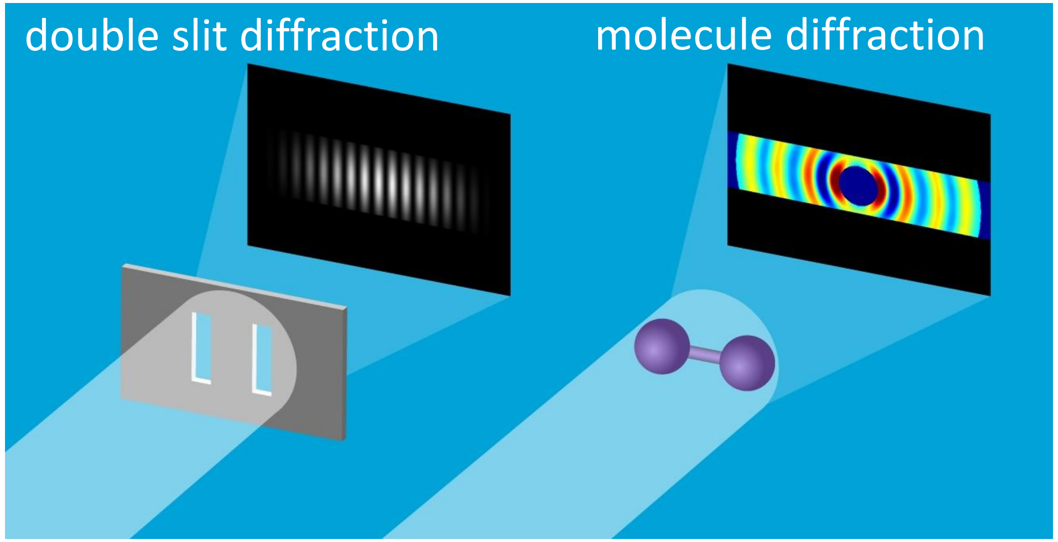


Robert Leiter, et al. The 16th European Microscopy Congress, Lyon, France, 2016

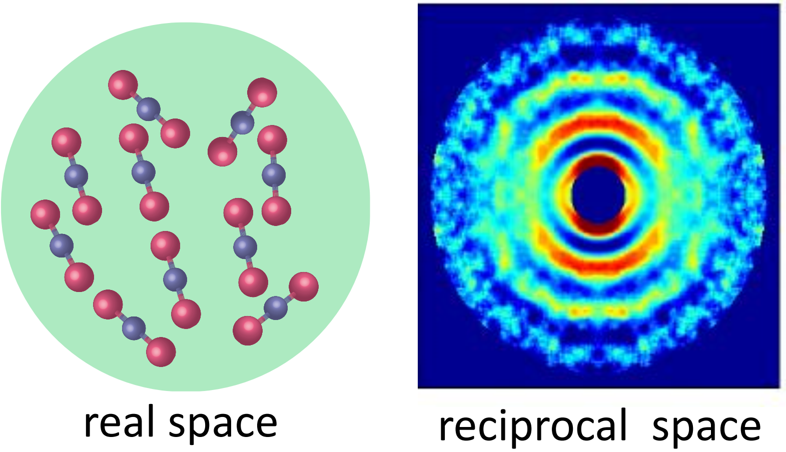
Real space imaging

- Good at resolving local structure, such as defects, dislocations, etc.
- Can reach Å spatial resolution
- Not yet reaching fs temporal resolution at the same time

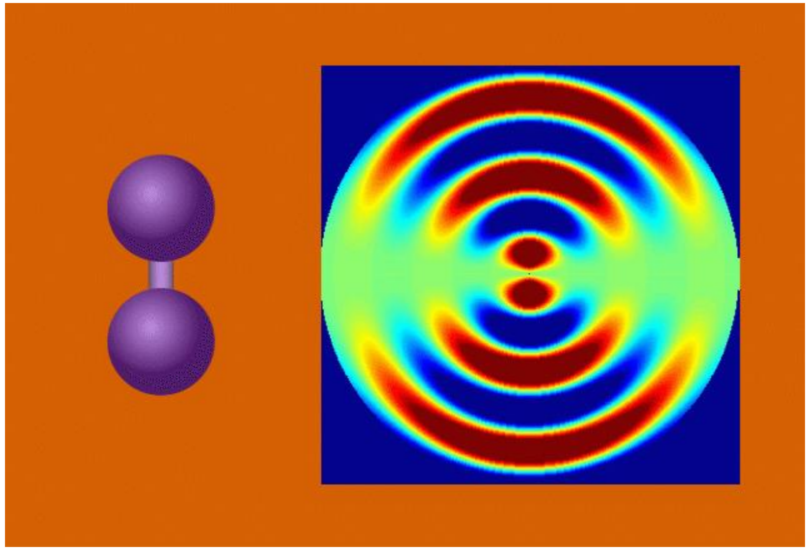
Diffractive (reciprocal space) imaging



Molecule ensemble



- Conjugated to real space imaging
- Ensemble average imaging
- Can reach Å-fs spatial-temporal resolution



Development of keV UED

Prof. Gérard Albert Mourou
recipient of 2018 Nobel Prize for physic "for his invention of chirped pulse amplification"

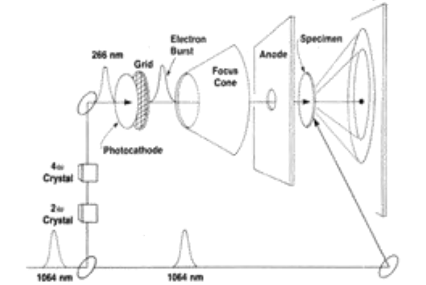
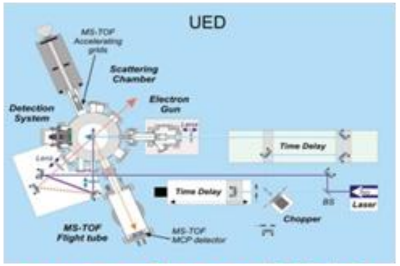


FIG. 1. Schematic of picosecond electron-diffraction apparatus. A streak-camera tube (deflection plates removed) is used to produce the electron pulse. The 25-keV electron pulse passes through the Al specimen and produces a diffraction pattern of the structure with a 20-ps exposure.

First UED

Prof. Ahmed Hassan Zewail
recipient of 1999 Nobel Prize in Chemistry, "for showing that it is possible with rapid laser technique to study in slow motion how atoms in a molecule move during a chemical reaction"



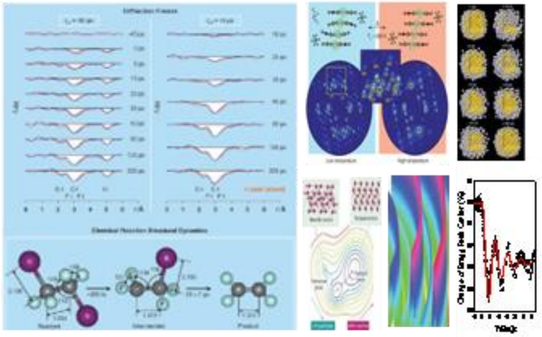
gas phase UED femtochemistry

Prof. R. J. Dwayne Miller
Chemistry and Physics,
University of Toronto

Prof. Bradley J. Siwick
Dept. of Phys. and Chem.
McGill University

Prof. Ralph Ernstorfer
Dept. of Phys. Chem.
Fritz Haber Institute

Prof. Claus Ropers
Max Planck Institute for Multidisciplinary Sciences



M. Chergui and A. H. Zewail, *ChemPhysChem* **10**, 28–43 (2009)

Prof. Jianming Cao
Department of Physics
Florida State University

Prof. Chong-Yu Ruan
Dept. of Phys. and Astron.
Michigan State University

Prof. Peter Baum
Department of Physics
Universität Konstanz

Prof. David Flannigan
Chemical Engineering and Materials Science
University of Minnesota

and many other researchers...

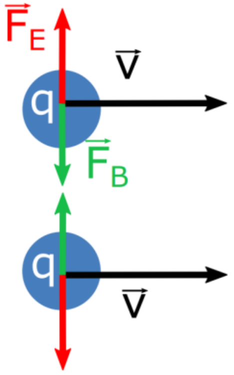
MeV Electrons for UED

Space-charge forces suppression with relativistic electrons

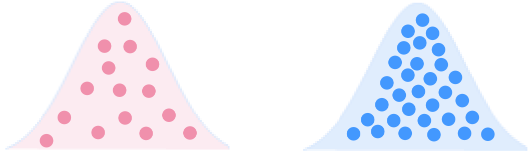
- shorter bunch ⇒ higher time resolution
- more electrons in a bunch ⇒ single shot measurement

Negligible pump-probe velocity mismatch

- Δt_{vm}<10 fs for 3 MeV e beam passing 150 μm gas target

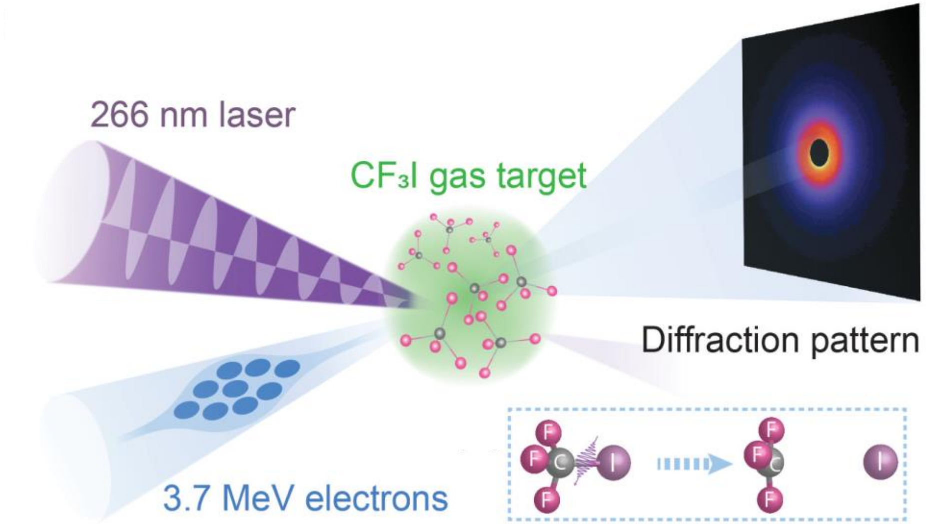


$$F_{sc} \propto \frac{1}{\beta^2\gamma^3}$$



Non-relativistic **Relativistic**

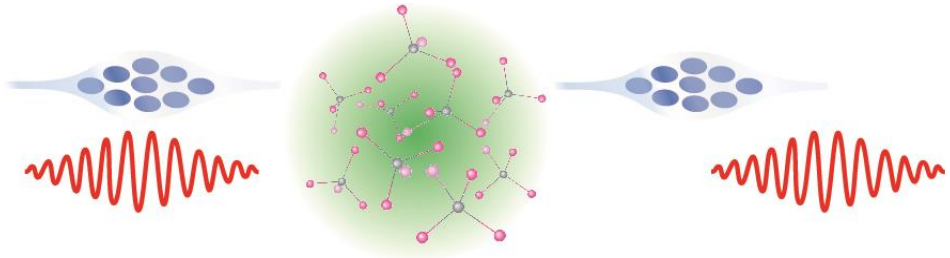
D. A. Edwards and M. J. Syphers, *An Introduction to the Physics of High Energy Accelerators*, Wiley, 1st edn., 1993.



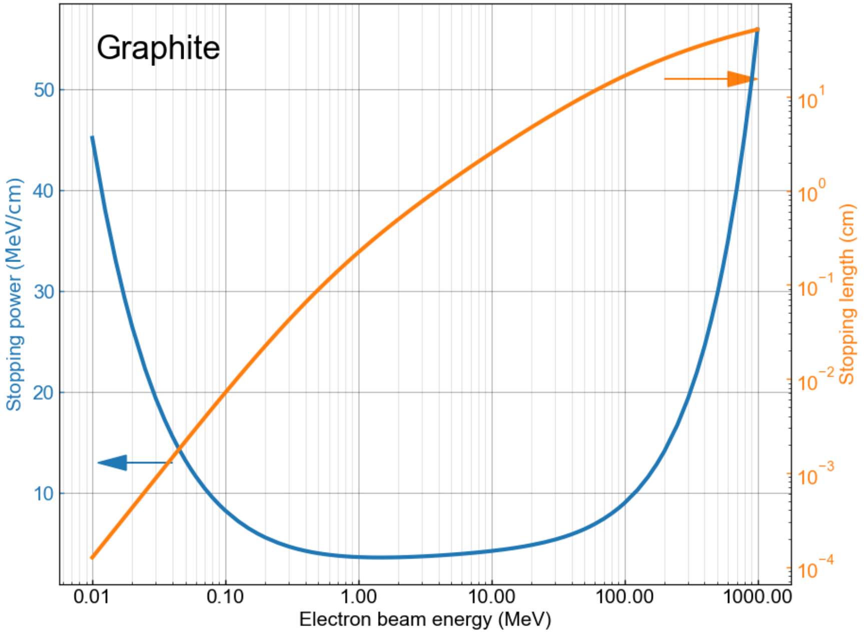
60 keV e^-
$\beta = 0.45$

gas target
150 um

$\Delta t_{vm} > 1\text{ps}$



MeV Electrons for UED



https://physics.nist.gov/PhysRefData/Star/Text/ESTAR.html

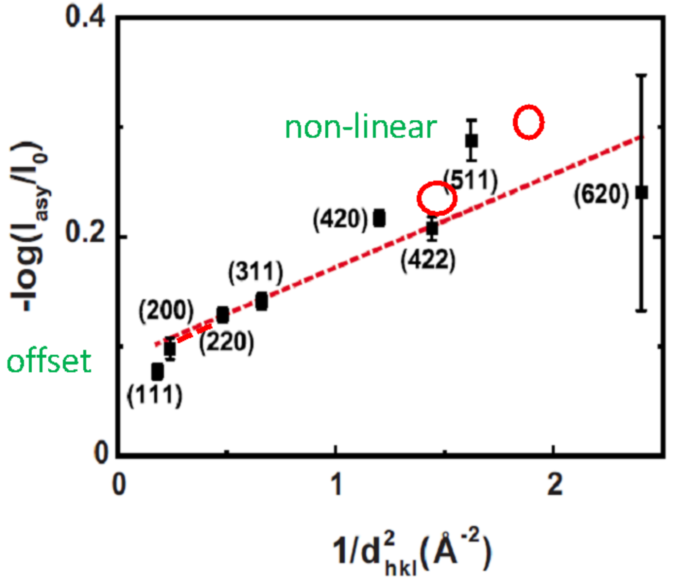
Larger penetration depth

- “thick” sample (100 nm level)
- kinematic diffraction

Debye-Waller effect:

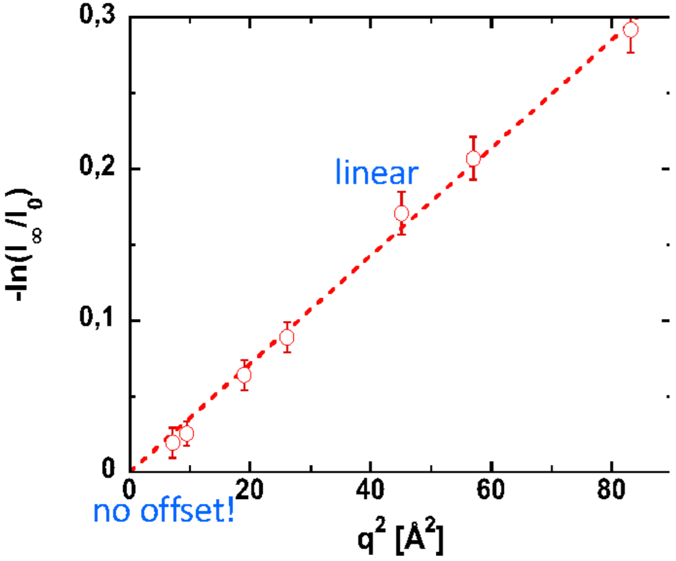
$$\boxed{I_{hkl} = I^0_{hkl} e^{-\frac{1}{2}G^2_{hkl}\Delta\langle u^2\rangle}} \Leftrightarrow \boxed{-\ln\left(\frac{I_{hkl}}{I^0_{hkl}}\right) = \frac{1}{2}G^2_{hkl}\Delta\langle u^2\rangle}$$

20 nm Au, 30 keV



Ligges et al., APL 94, 101910 (2009).

30 nm Au, 4 MeV

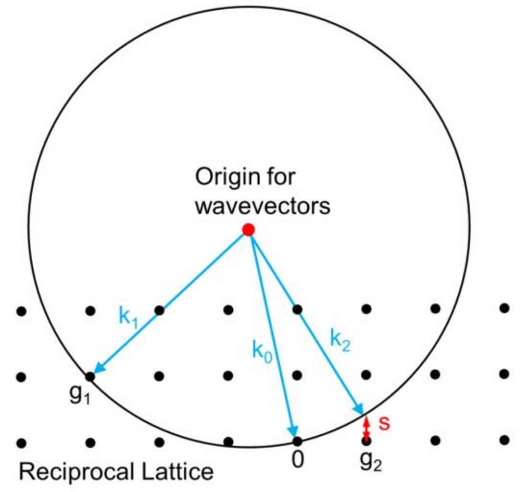


Courtesy of K. Sokolowski-Tinten, University of Duisburg-Essen, Duisburg, Germany

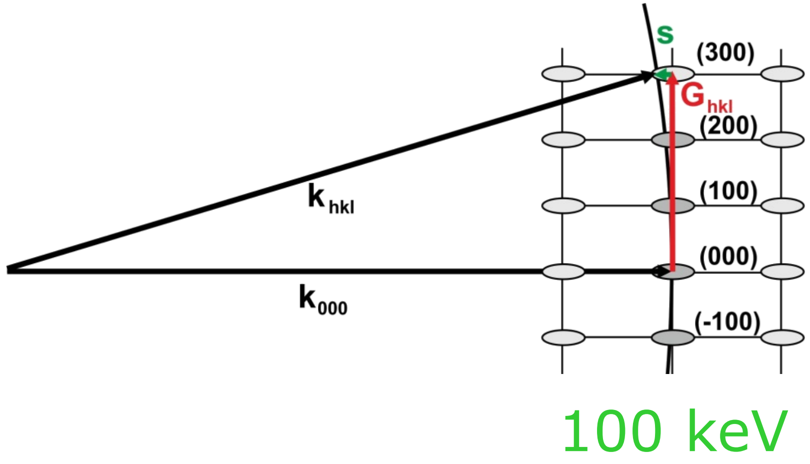
MeV Electrons for UED

Really flat Ewald sphere

Ewald sphere radius $\propto \frac{1}{\lambda} = \frac{p}{h} = \frac{\beta\gamma m_0 c}{h}$

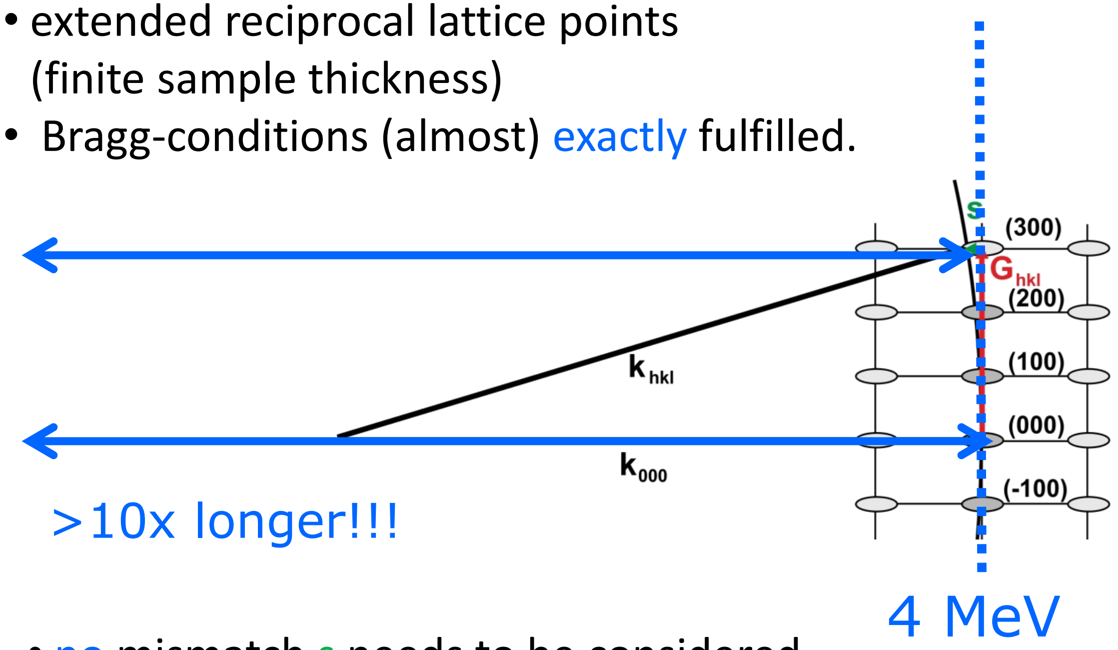


- k >> G ⟶ relativley flat Ewald-sphere
- extended reciprocal lattice “points“ (finite sample thickness)
- Bragg-condition only approximately fulfilled.



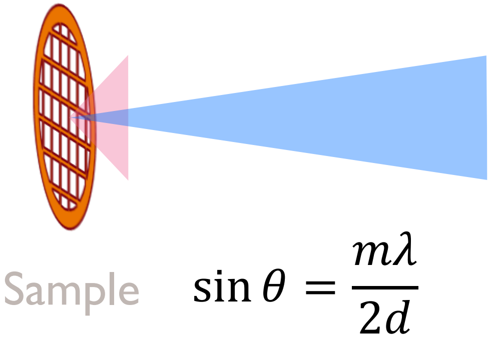
- mismatch s needs to be considered

- k >>> G ⟶ “really“ flat Ewald-sphere
- extended reciprocal lattice points (finite sample thickness)
- Bragg-conditions (almost) exactly fulfilled.

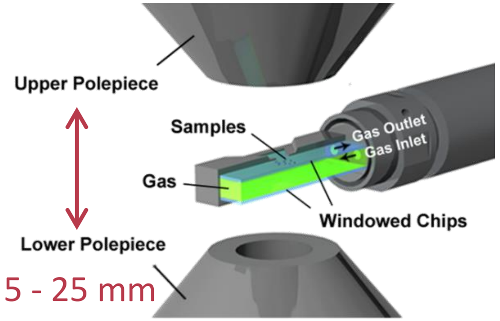


- no mismatch s needs to be considered

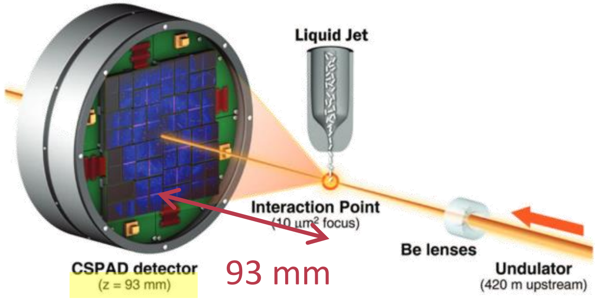
MeV Electrons for UED



$$\sin\theta = \frac{m\lambda}{2d}$$

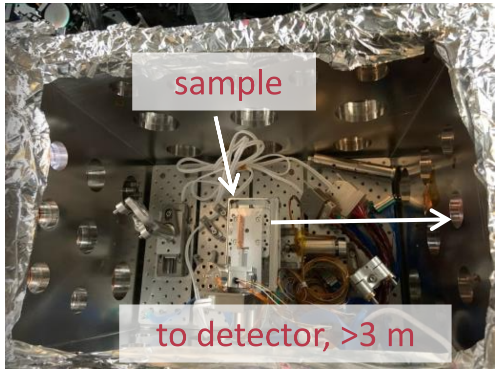


300-keV TEM
λ=0.02 Å



S. Boutet et al., Science. 337, 362–364 (2012).

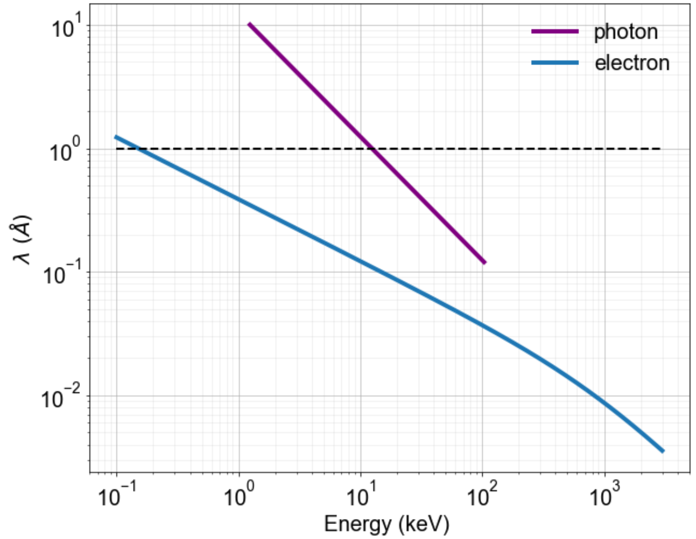
9.4-keV x-ray diffraction
λ=1.3 Å



4-MeV UED
λ=0.003 Å

Shorter wavelength

- higher momentum transfer ⇒ higher spatial resolution
- smaller diffraction angle → longer sample-to-detector distance → flexible environment for “dirty” samples



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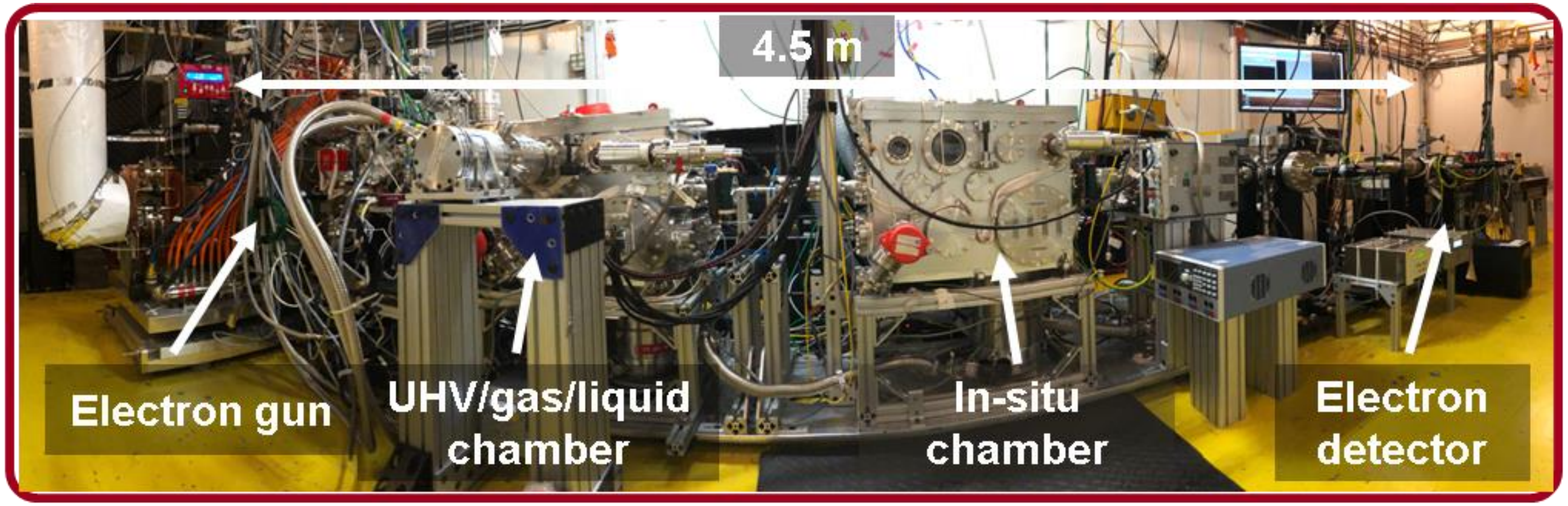
SLAC MeV UED Milestones

Apr 2014	MeV UED program launched
Mar 2015	Gas-phase UED
Mar 2016	Single-shot UED
Jun 2017	THz Mid-IR UED
Jan 2019	Official user facility
Apr 2019	Liquid-phase UED
Jun 2019	User Run01 solid UED
Aug 2020	User Run02 gas UED
May 2021	User Run03 solid UED
Oct 2024	User Run04 gas UED

PENNSTATE
University of Nebraska Lincoln
Stony Brook University
Universität Duisburg Essen
University of York
Universität Potsdam
Stanford University
PULSE
SIMES
Brookhaven National Laboratory
Argonne National Laboratory
MIT

MeV UED instrument overview

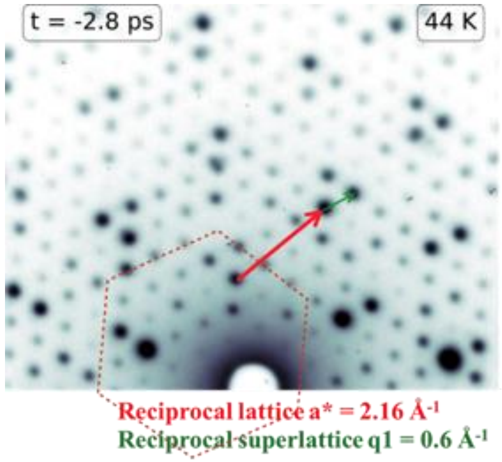


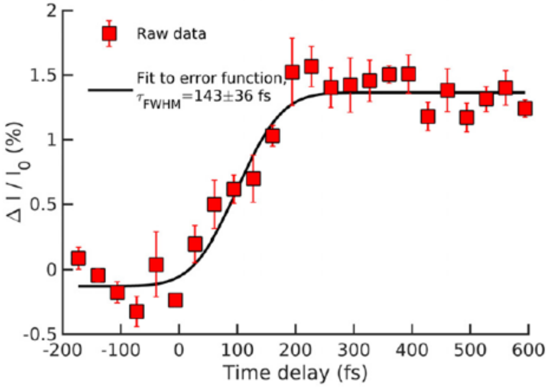
Beam energy	2 – 4 MeV
Energy spread	10^{-4} (200 eV)
Beam charge	$10^4 – 10^6$
Beam emittance	2–20 nm·rad

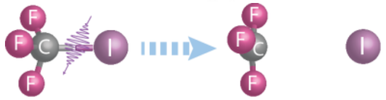
S. Weathersby, *et al.*, *Rev. Sci. Instrum.* **86**, 73702 (2015))

Beam size	100 – 200 µm
Q (2π/d) range/res. (FWHM)	12 $Å^{-1}$ / 0.17 $Å^{-1}$
Time res. (FWHM)	< 150 fs
Repetition rate	SS - 360 Hz

X. Shen, *et al.*, *Struct. Dyn.* **6**, 054305 (2019)







Multifunctional platform for ultrafast science

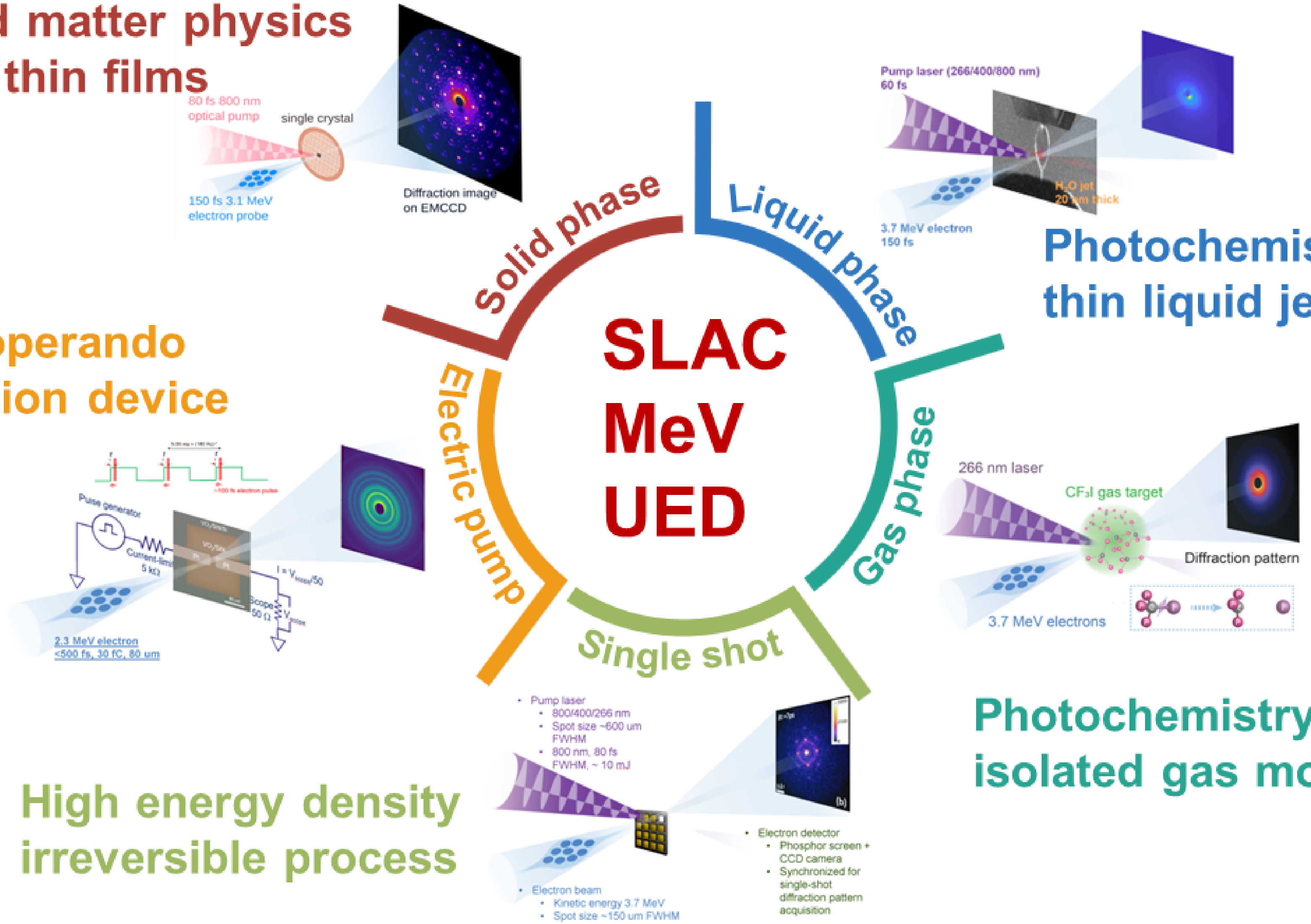
Condensed matter physics
solid state thin films

Photochemistry
thin liquid jet

In-situ/operando
application device

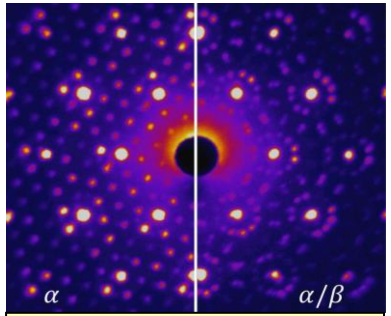
Photochemistry
isolated gas molecule

High energy density
irreversible process

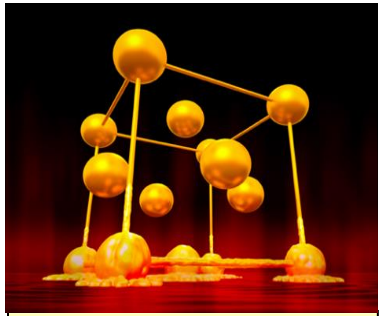


- Pump laser
  - 800/400/266 nm
  - Spot size ~600 um FWHM
  - 800 nm, 80 fs FWHM, ~ 10 mJ
- Electron beam
  - Kinetic energy 3.7 MeV
  - Spot size ~150 um FWHM
- Electron detector
  - Phosphor screen + CCD camera
  - Synchronized for single-shot diffraction pattern acquisition

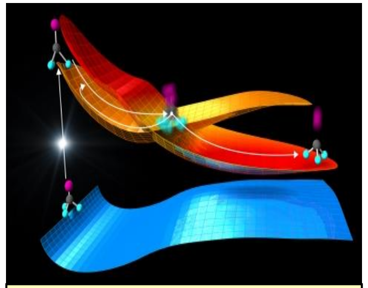
Ultrafast science enabled by MeV UED



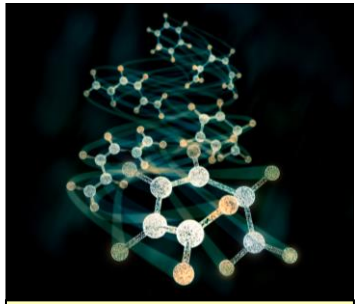
Phase switch with a single flash of light (***Sci. Adv.*** 4, eaau5501 (2018)).



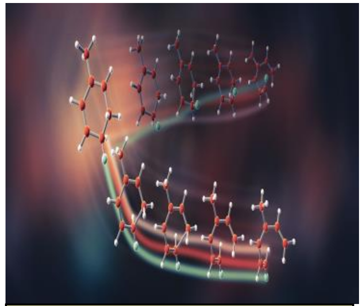
Resolving ultrafast phase transitions (***Science*** *360 1451–1455 (2018))*



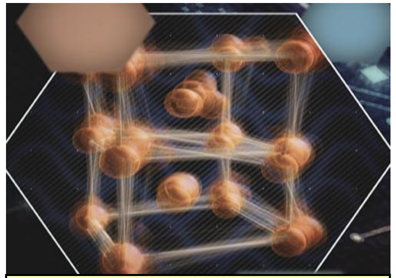
Bond-breaking & nuclear wavepacket passing through conical intersections (***Science*** *361 64–67 (2018)*)



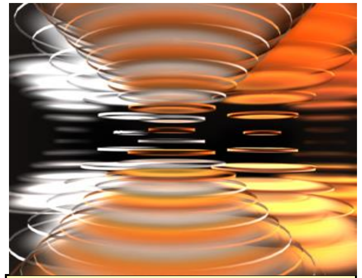
Ring-opening & ground state dynamics (***Nat. Chem.*** 11, 504–509 (2019)).



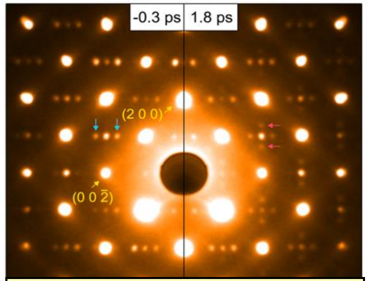
Imaging conformer-specific photochemistry(***Science***. 374, 178–182 (2021).).



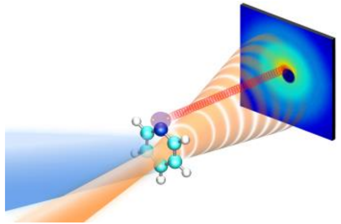
Operando characterization of a quantum electronic device (***Science*** 373, 352–355 (2021)).



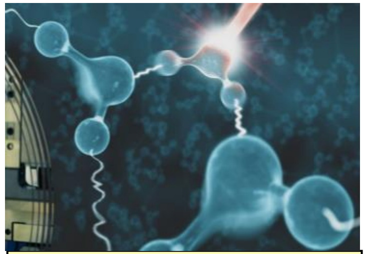
Ultrafast topological switch by Time-varying shear strain (***Nature*** 565, 61-77 (2019))



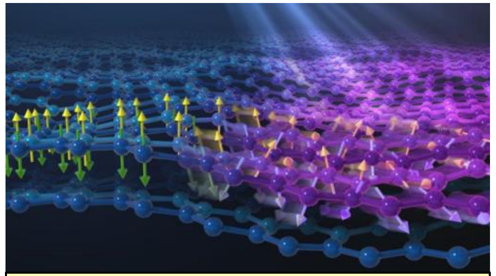
Light induced CDW (***Nat. Phys***. 16, pages159–163(2020)).



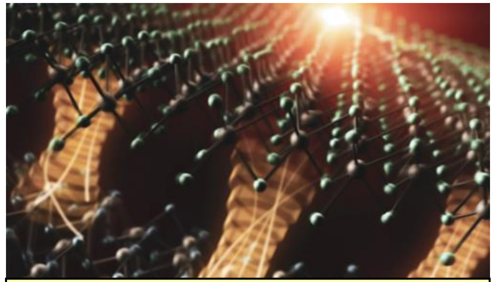
Simultaneous observation of nuclear and electronic dynamics (***Science.*** **368,** 885 (2020)).



Ultrafast hydrogen bond strengthening in liquid water(***Nature***. 596, 531(2021)).

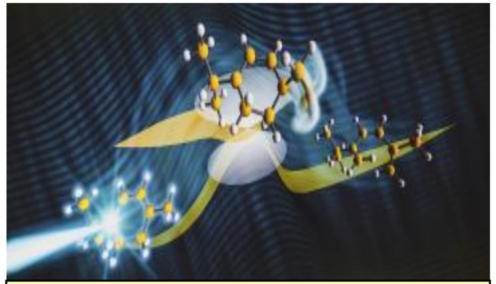


Spin-mediated shear oscillators in a van der Waals antiferromagnet (***Nature***. 620, 988–993 (2023))).



Phonon-mediated heat transfer in a van der Waals heterostructure (***Nat. Nanotechnol.*** 18, 29–35 (2023)).

Incipient plasticity in dynamically compressed matter (***Nat Commun***. 13, 1055 (2022)).



Rehybridization dynmaics into a pericyclic minimum state (***Nat. Commu***. 14, 2795 (2023)).

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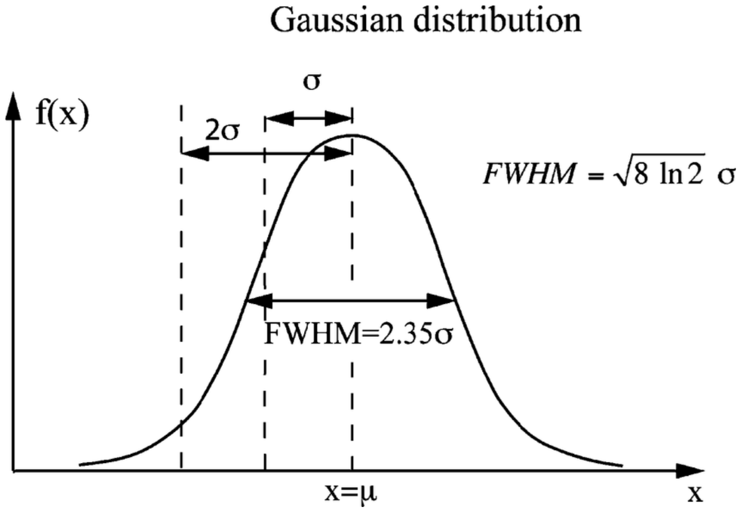
- Perspectives and Comments

On time resolution

- Unit: full-width-at-half-maximum (FWHM) for ultrafast science field, root-mean-square (rms) for accelerator physics field. For a Gaussian beam

  $\tau_{fwhm} = \sqrt{8ln2}\sigma_{rms} = 2.35\sigma_{rms}$
- UED temporal resolution (fwhm)

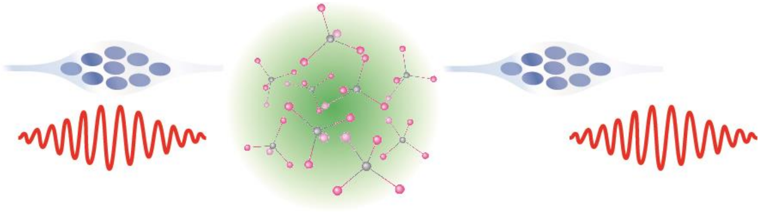
$$\tau = \sqrt{\tau_{laser}^2 + \tau_{electron}^2 + \tau_{VM}^2 + \tau_{TOA}^2}$$

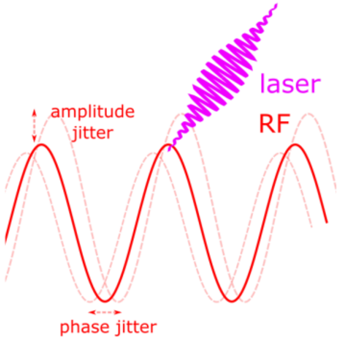


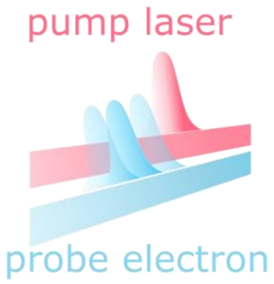
60 keV e⁻
$\beta = 0.45$

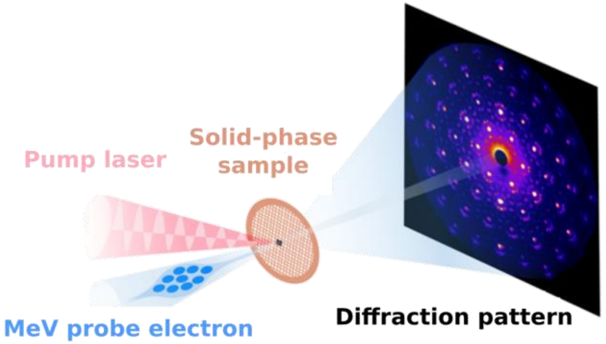
gas target
150 um

$\Delta t_{vm} > 1\text{ps}$







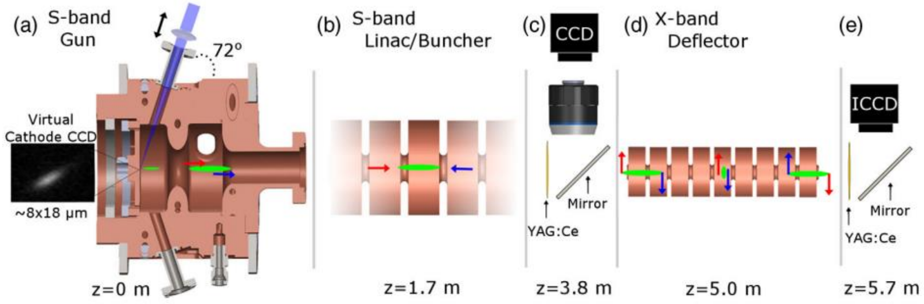


τ_{VM}

τ_{TOA}

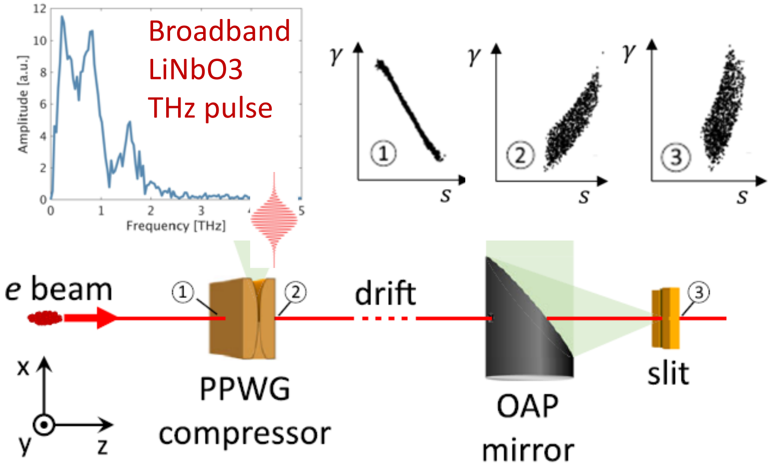
On time resolution

- Temporal compression using RF bunching cavity
    - achieved $\tau_{electron}$ =10 fs (rms)=23.5 fs (fwhm) bunch length
    - Introduced large $\tau_{TOA} > 100$ fs



J. Maxon, et al., *PRL* **118**, 154802 (2017)

- Temporal compression using broadband quasi-single-cycle THz laser
    - achieved $\tau_{electron}$= 39 fs (rms)=91.65 fs (fwhm)
    - reduced τ_{TOA} by a factor of 2 to 31 fs (rms) = 72.9 fs (fhwm)
    - Low THz laser conversion efficiency (~0.1%)
    - Introduced large emittance growth due to the broadband nature the THz pulse

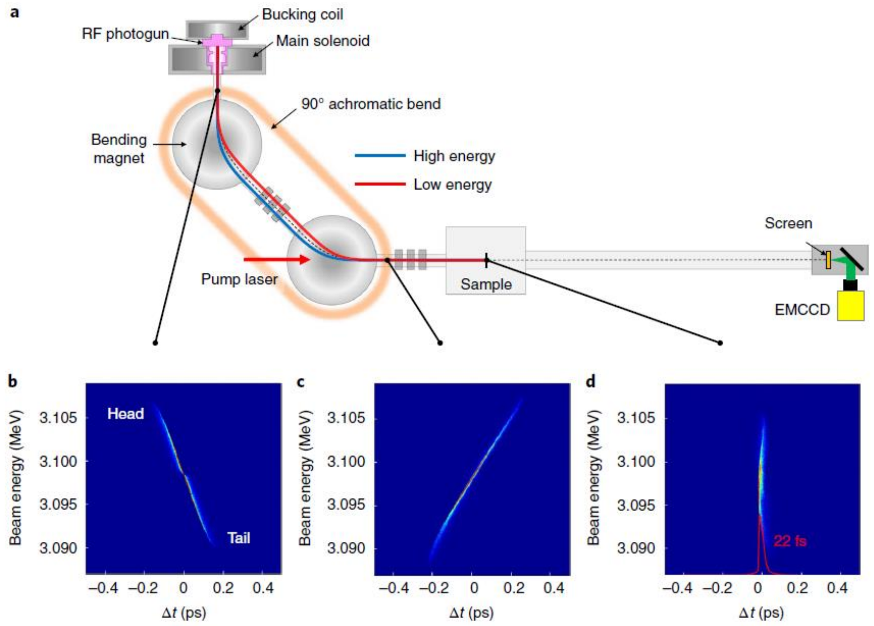


E. C. Snively, *et al. Phys. Rev. Lett.* **124**, 054801 (2020).

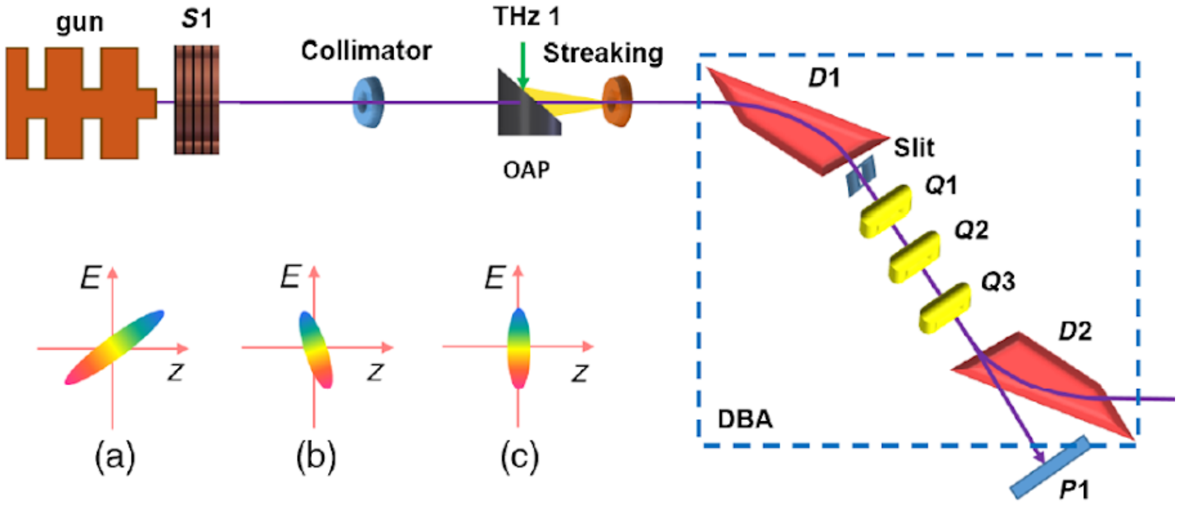
On time resolution

Bunch compression and timing jitter reduction by double bend achromat (DBA)

- Isochronous beamline with DBA to minimize τ_{TOA}
- Use space charge effect to induce correct chirp for $\tau_{electron}$ compression
- Achieved ~30 fs $\tau_{electron}$ and τ_{TOA}
- Fixed energy, fixed high charge, large emittance growth



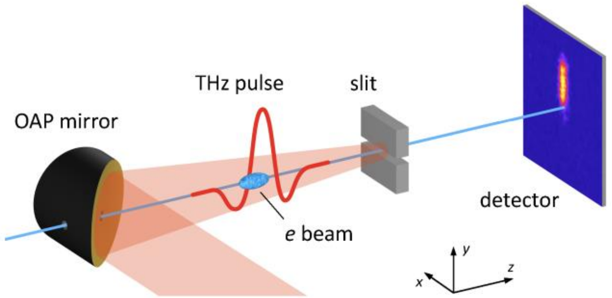
H. W. Kim, *et al. Nat. Photonics* **14**, 245–249 (2020)



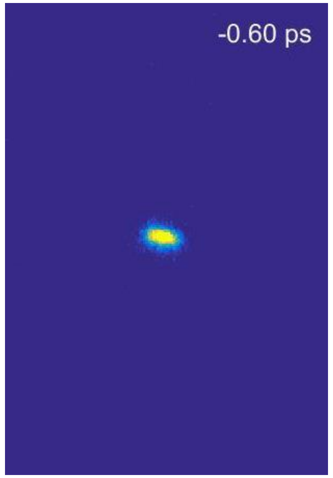
F. Qi, *et al. Phys. Rev. Lett.* **124**, 134803

On time resolution

THz-based arrival time monitor

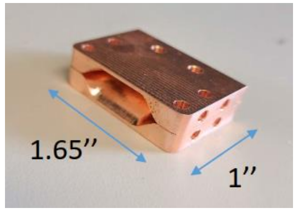


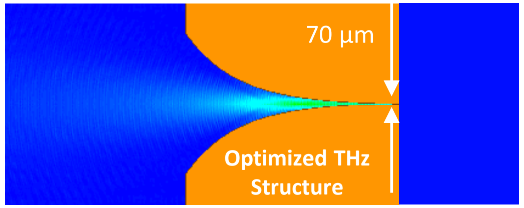
R. K. Li, et al., *Phys. Rev. Accel. Beams* **22**, 012803 (2019).



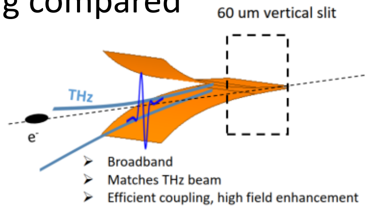
3.1 MeV electrons, 50 um gap, 100 um thick slit

Parallel-plate waveguide for enhanced streaking

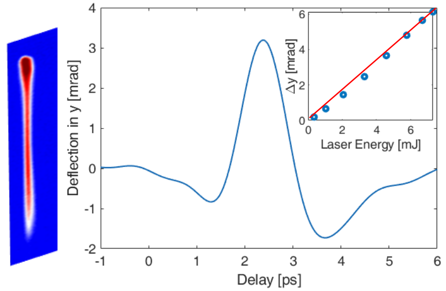




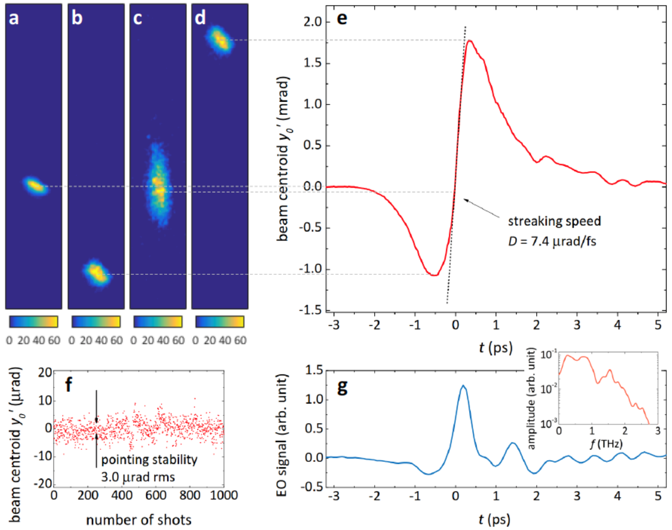
>2 times stronger steaking compared to slit

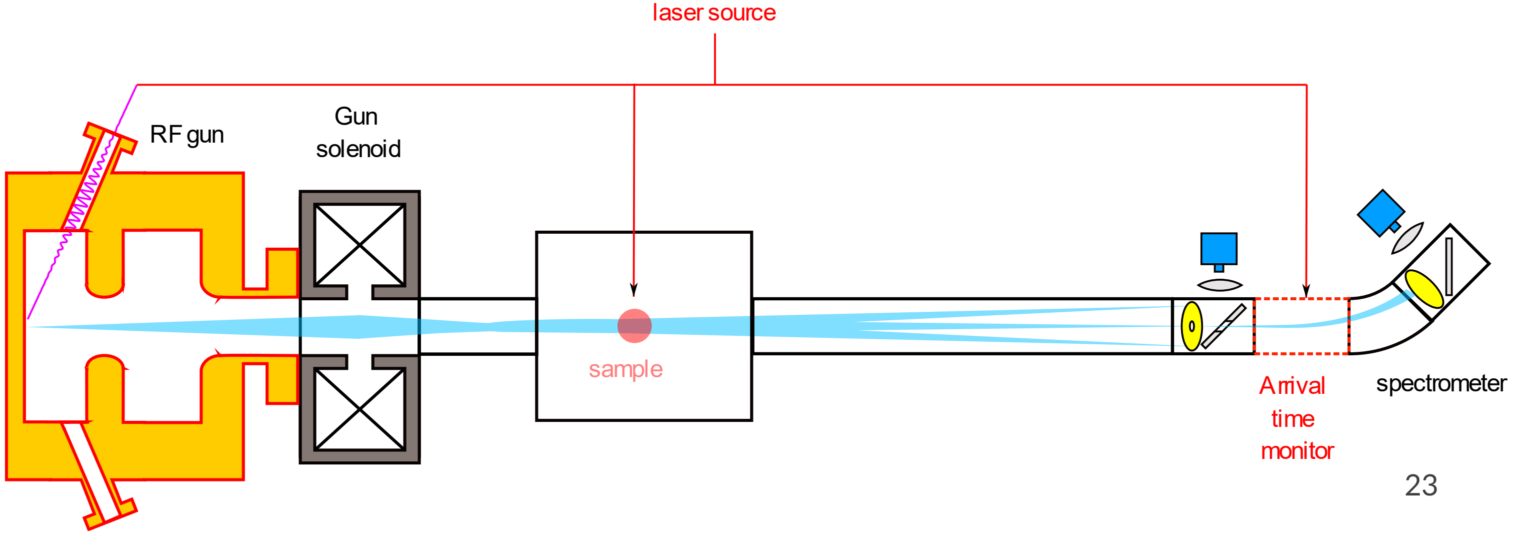


- Broadband
- Matches THz beam
- Efficient coupling, high field enhancement

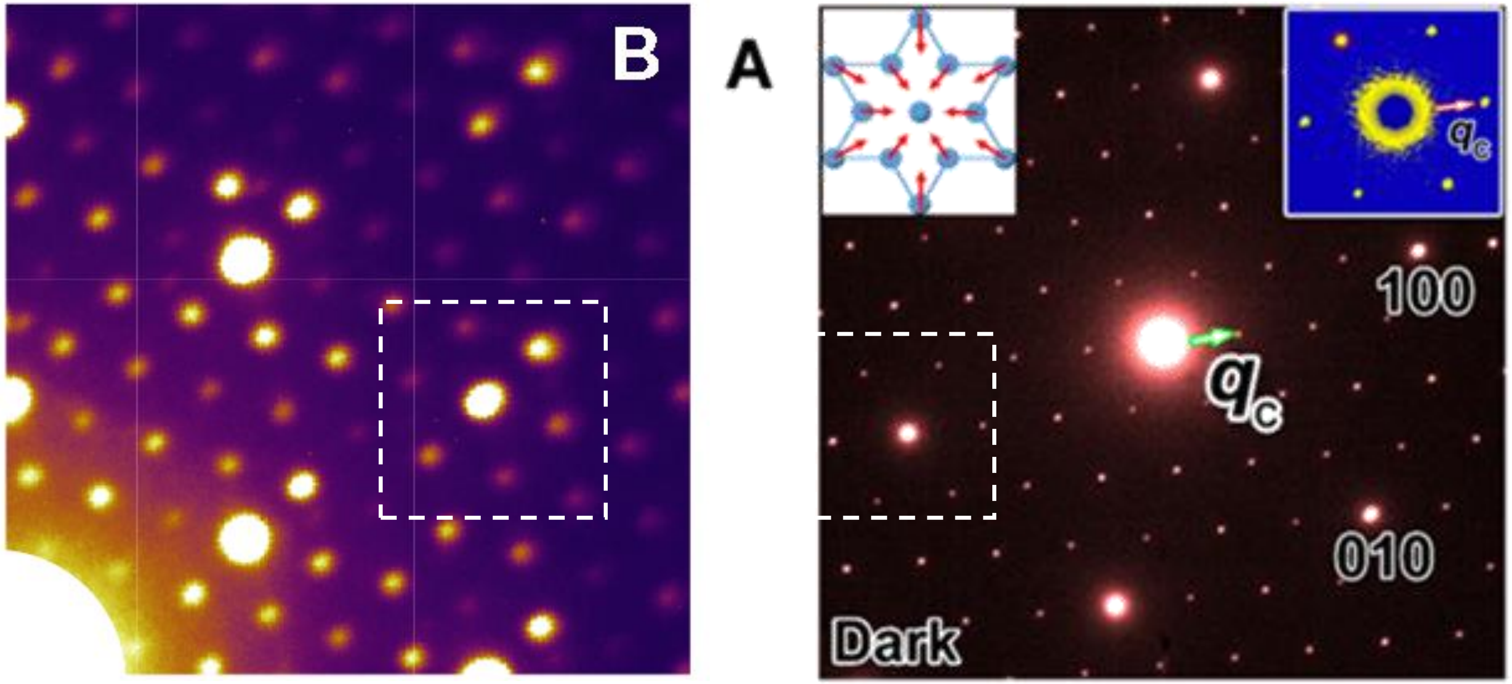


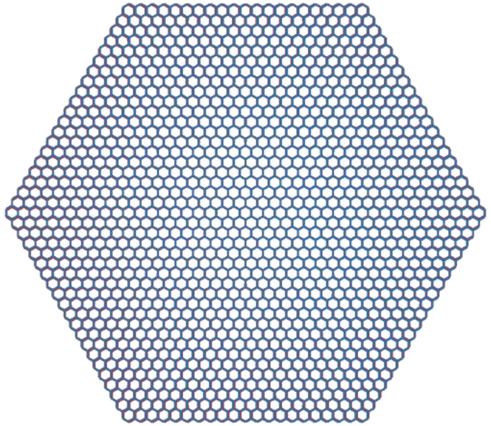
M. A. K. Othman *et al.*, *Opt. Express*. **27**, 23791 (2019).





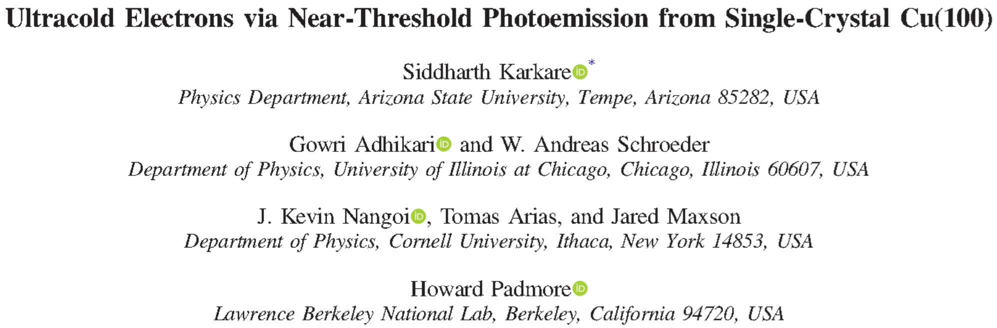
On reciprocal space resolution (spatial coherence)





Moire patterns from twisted bilayer graphene

Higher spatial coherence ⇒ narrower Bragg peak



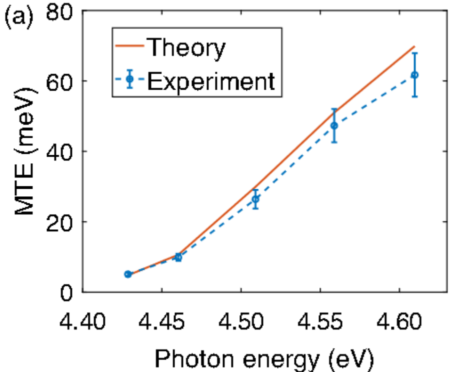
Ultracold Electrons via Near-Threshold Photoemission from Single-Crystal Cu(100)

Siddharth Karkare*
Physics Department, Arizona State University, Tempe, Arizona 85282, USA

Gowri Adhikari and W. Andreas Schroeder
Department of Physics, University of Illinois at Chicago, Chicago, Illinois 60607, USA

J. Kevin Nangoi, Tomas Arias, and Jared Maxson
Department of Physics, Cornell University, Ithaca, New York 14853, USA

Howard Padmore
Lawrence Berkeley National Lab, Berkeley, California 94720, USA



On electron beam brightness

E beam transverse brightness dependence on photoelectron gun gradient

$$B_\perp^{pancake} \propto E, \qquad B_\perp^{cigar} \propto E^{3/2}$$

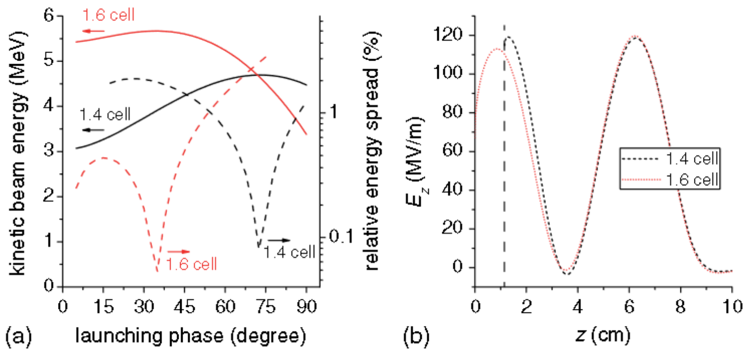


FIG. 1. Comparison of 1.6-cell and 1.4-cell guns for (a) the output kinetic energy and energy spread (10-ps-long, low-charge electron beam) and (b) the acceleration gradient seen by the beam.

R. K. Li, P. Musumeci, *Phys. Rev. Applied*. **2**, 024003 (2014).

High gradient comes with high dark current – exponential dependence!

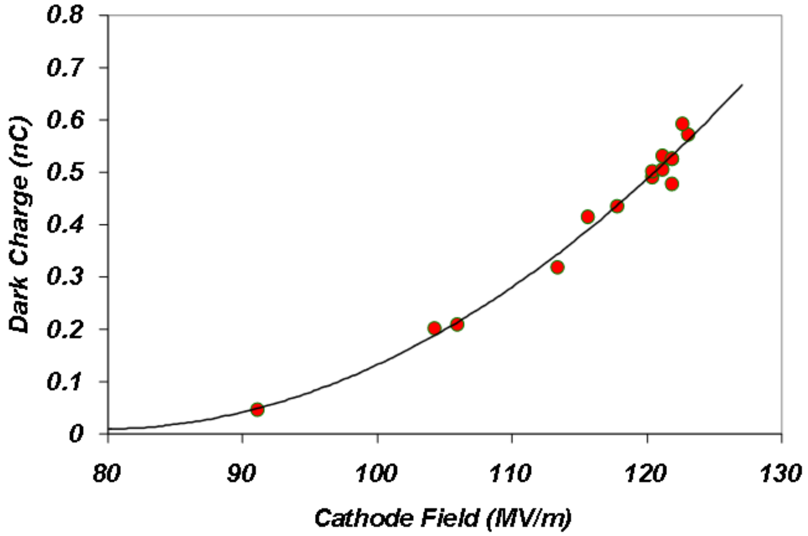
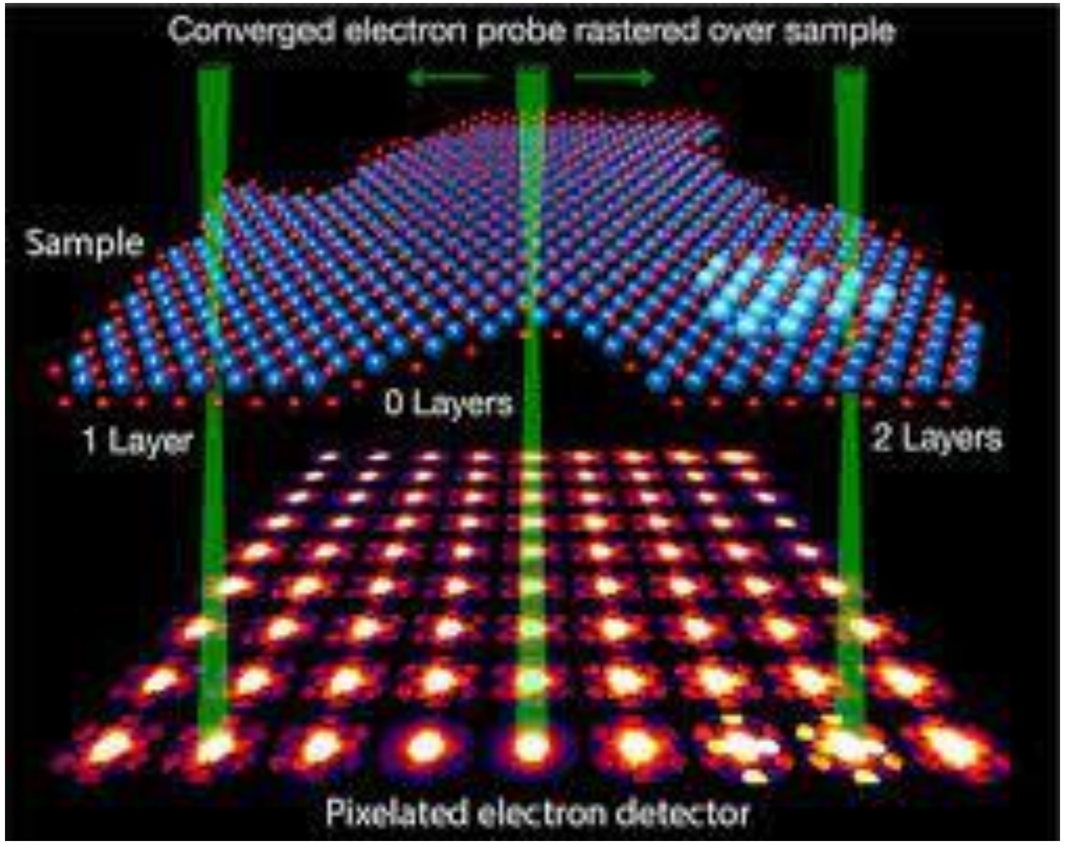


Figure 7: Dark charge vs. the peak cathode field.

On nanoprobe

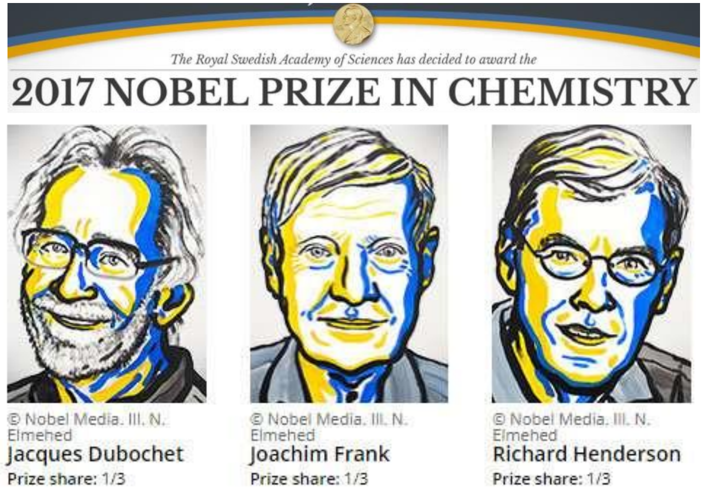
Require low emittance and high brightness e beam
Mechanical vibration and other environmental noises needs to be well diminished



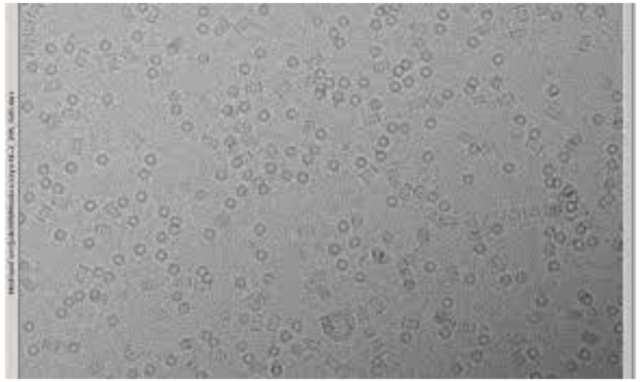
4D (x, y, q_x, q_y) Scanning Transmission Electron Microscopy

On electron beam detector

Every electron counts! Direct electron detectors

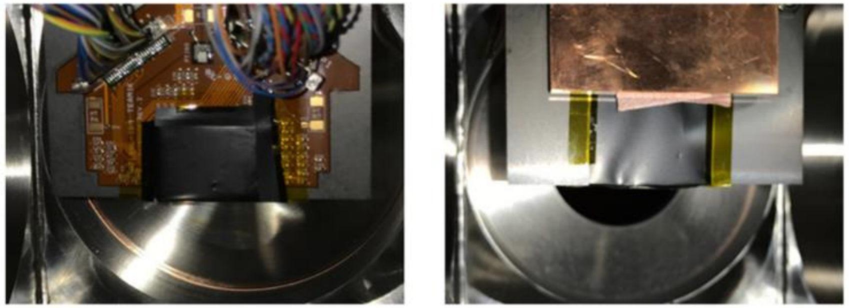


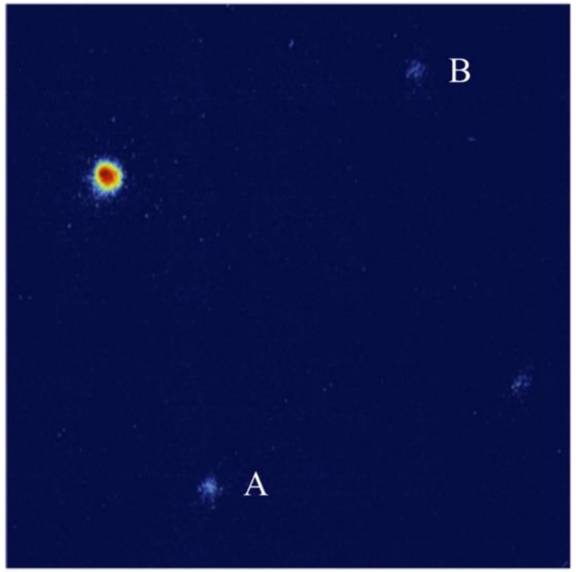
Critical components enabling full capability of cryoEM



Atomic resolution single particle imaging
enabled by direct electron detector

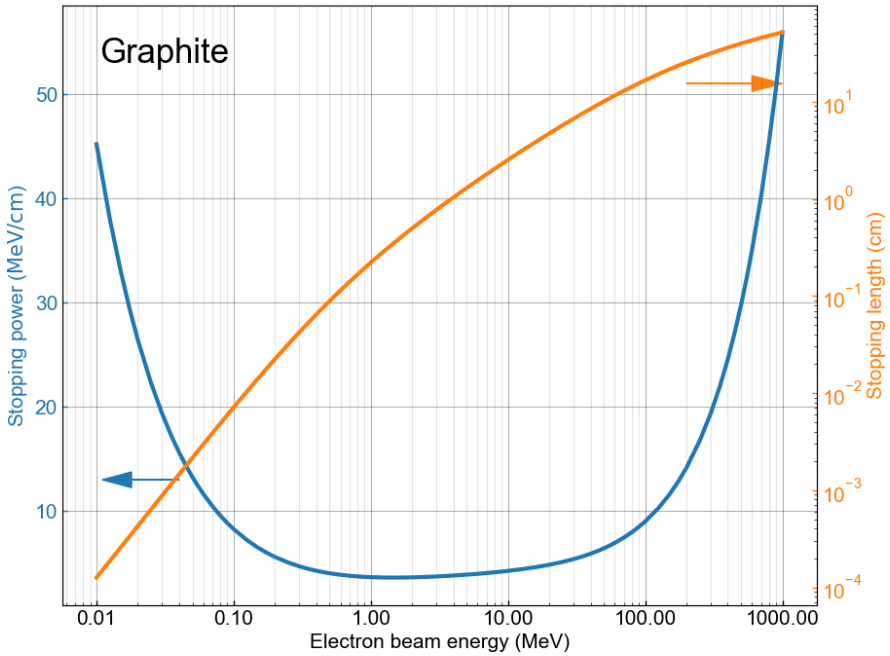
Dr. Peter Denes, LBNL





T. Vecchione *et al.*, *Review of Scientific Instruments*. **88**, 033702 (2017).

Comments



https://physics.nist.gov/PhysRefData/Star/Text/ESTAR.html

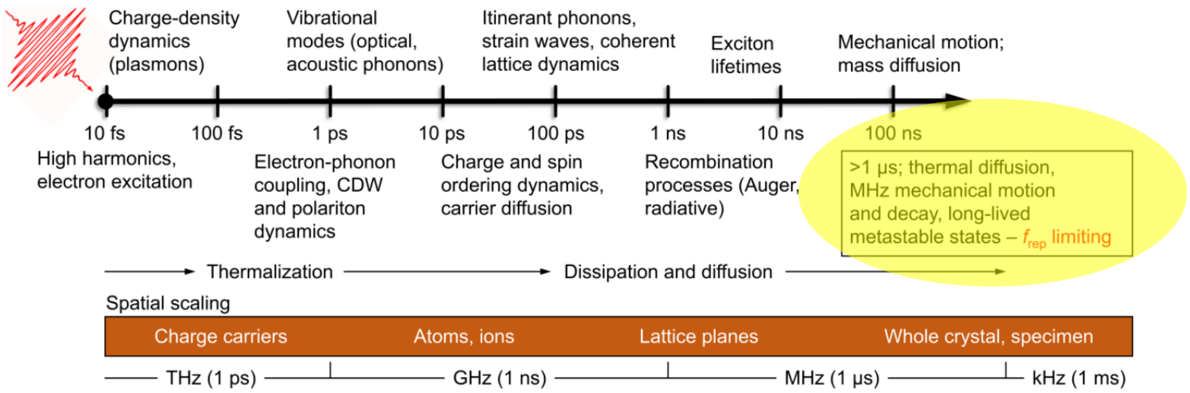


FIG 3. Select zoology of ultrafast photoinduced dynamics and their approximate timescales. Boundary conditions become increasingly important to energy dissipation with increasing time owing to dynamics encompassing the entire specimen (*i.e.*, initiating with charge-carrier dynamics and increasing in scale to whole-crystal/specimen motion and thermal diffusion). Relatively long-lived states and dissipation channels will limit the UEM operational f_{rep} if full recovery is desired, which can take microseconds or longer. For comparison, the f_{rep} (and the associated time between packets) needed to capture full recovery of the discrete examples is shown in relation to the overall chain of events, further illustrating the need for f_{rep} < MHz for full recovery of the broadest range of chemical and materials phenomena.

Flannigan, D. J., The Journal of Chemical Physics **157**, 180903 (2022).

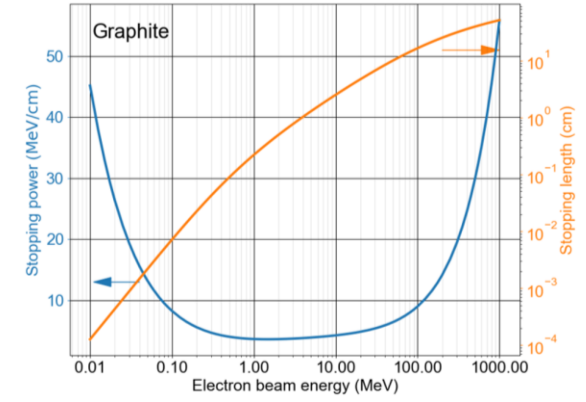
Beam energy: a few MeV
Time resolution: science dependence (fs-ps)
Rep rate: sample dependence
Emittance: the lower the better with reasonable flux
Sample: ~100 nm thin film
Vacuum: gun 1e-10 torr, sample <1e-4 torr, enviornmental TEM possible.

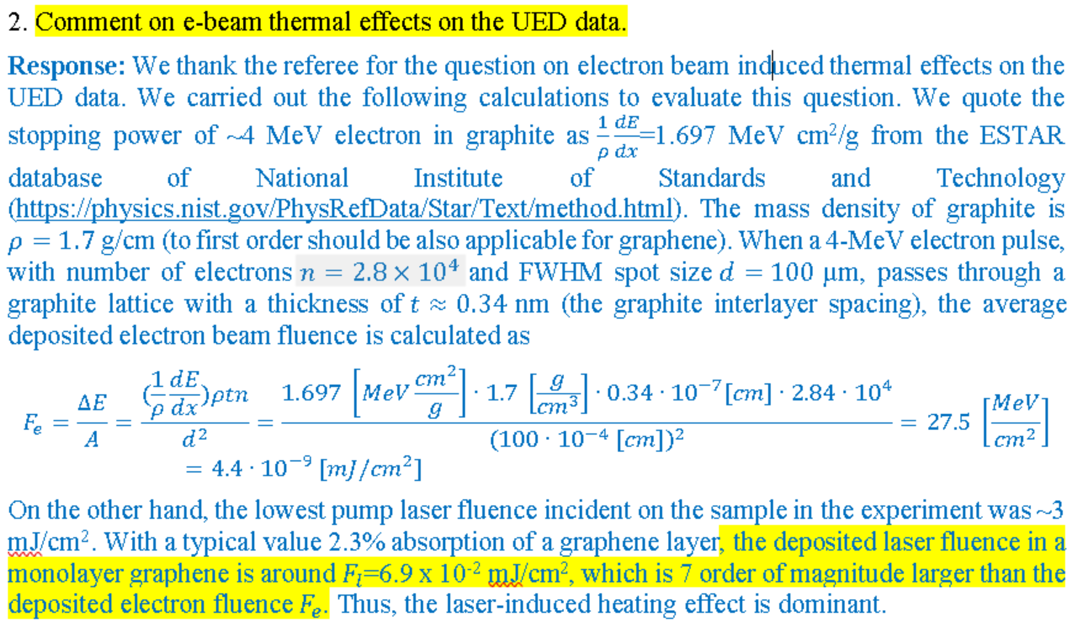
Comments

Multi-modal experiment

Less sample damage

- less energy deposition
- pulsed operation, lower dose rate, less damage to dose rate sensitive matter
- gentle probe for multi-modal



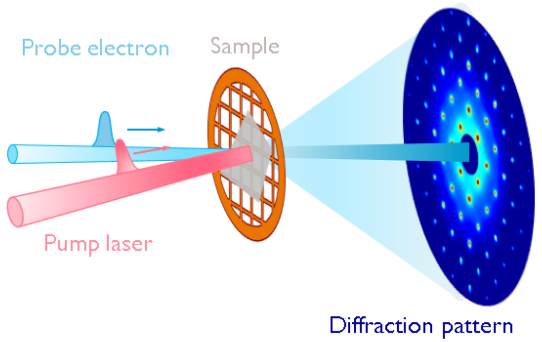


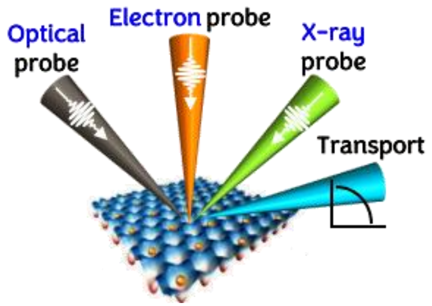
2. Comment on e-beam thermal effects on the UED data.

Response: We thank the referee for the question on electron beam induced thermal effects on the UED data. We carried out the following calculations to evaluate this question. We quote the stopping power of ~4 MeV electron in graphite as $\frac{1}{\rho}\frac{dE}{dx}$=1.697 MeV cm²/g from the ESTAR database of National Institute of Standards and Technology (https://physics.nist.gov/PhysRefData/Star/Text/method.html). The mass density of graphite is $\rho = 1.7$ g/cm (to first order should be also applicable for graphene). When a 4-MeV electron pulse, with number of electrons $n = 2.8 \times 10^4$ and FWHM spot size $d = 100$ µm, passes through a graphite lattice with a thickness of $t \approx 0.34$ nm (the graphite interlayer spacing), the average deposited electron beam fluence is calculated as

$$F_e = \frac{\Delta E}{A} = \frac{(\frac{1}{\rho}\frac{dE}{dx})\rho t n}{d^2} = \frac{1.697\left[MeV\frac{cm^2}{g}\right]\cdot 1.7\left[\frac{g}{cm^3}\right]\cdot 0.34\cdot 10^{-7}[cm]\cdot 2.84\cdot 10^4}{(100\cdot 10^{-4}\,[cm])^2} = 27.5\left[\frac{MeV}{cm^2}\right]$$
$$= 4.4\cdot 10^{-9}\,[mJ/cm^2]$$

On the other hand, the lowest pump laser fluence incident on the sample in the experiment was ~3 mJ/cm². With a typical value 2.3% absorption of a graphene layer, the deposited laser fluence in a monolayer graphene is around F_l=6.9 x 10^{-2} mJ/cm², which is 7 order of magnitude larger than the deposited electron fluence F_e. Thus, the laser-induced heating effect is dominant.





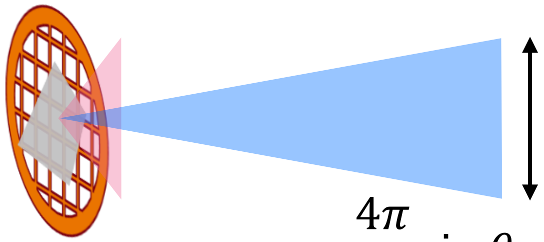
Multi-modal characterization

Comments

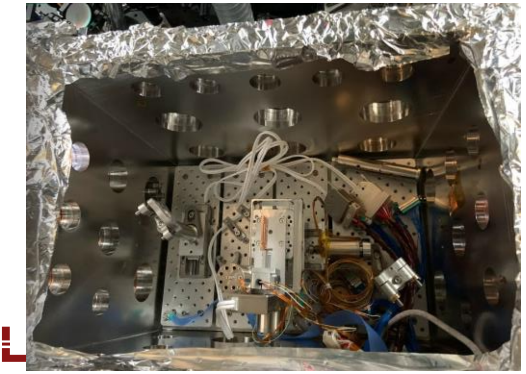
In-situ experiments

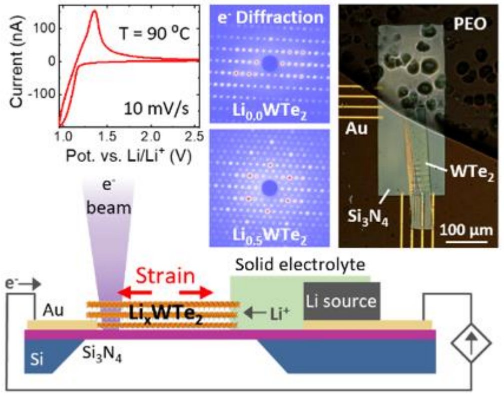
Flexible sample environment

- Bragg's law $m\lambda = 2d\sin\theta \leftrightarrow \sin\theta = \frac{m\lambda}{2d}$
- For MeV electrons, $\lambda \sim 0.001$ Å, $\theta \sim$mrad
- large space chamber, long distance separation between parts

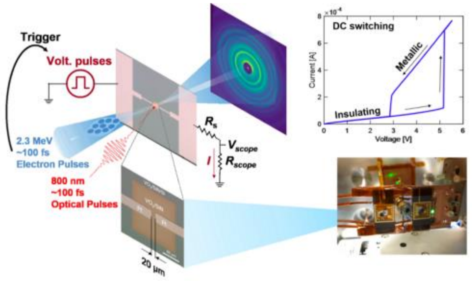


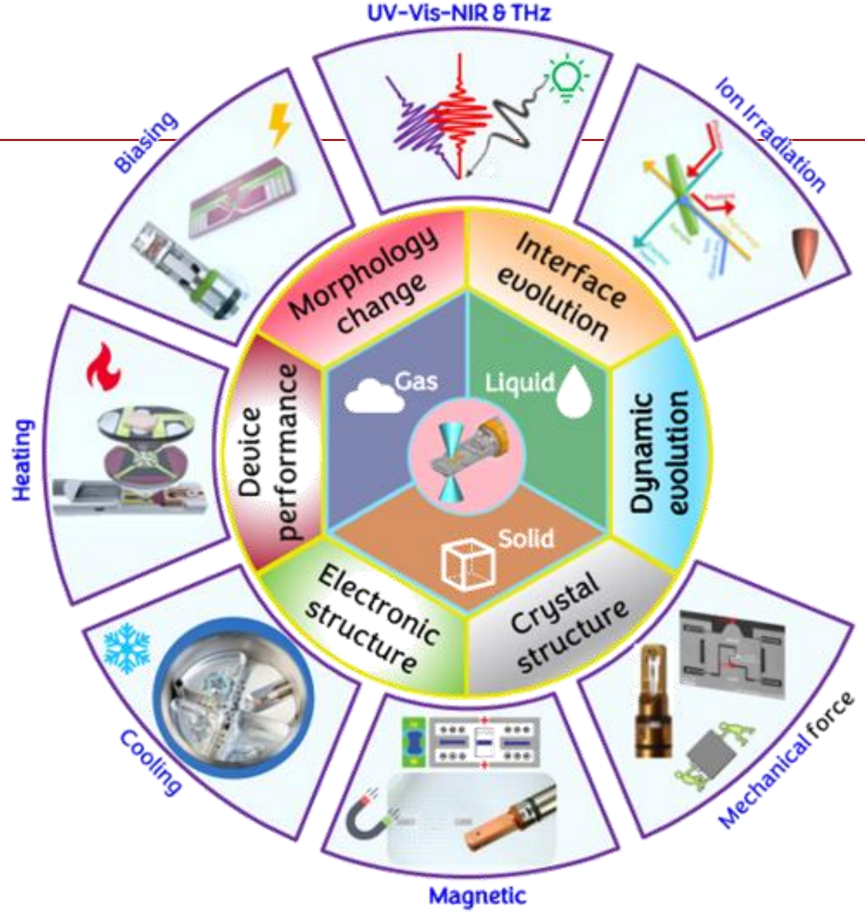
$$q = \frac{4\pi}{\lambda}\sin\theta$$

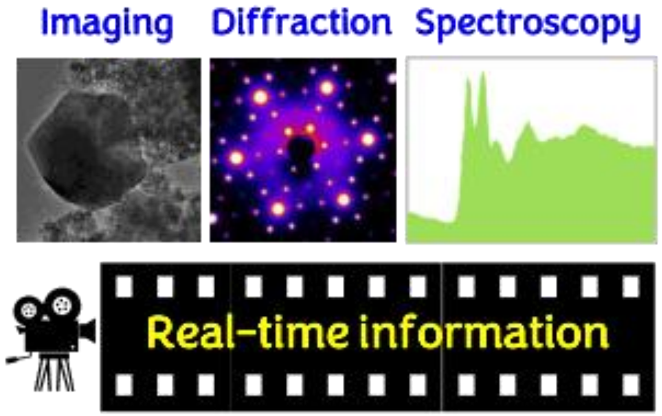




All-solid electron-transparent implementation







Acknowledgments

- SLAC MeV UED team and collaborators
- Strong support from SLAC management. Technical support from SLAC Accelerator Directorate, Technology Innovation Directorate, LCLS Laser Science & Technology Division and Test Facilities Department
- MeV-UED is operated as part of the Linac Coherent Light Source at the SLAC National Accelerator Laboratory, supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76SF00515