ASTA

MEV-UED INSTRUMENT

Application of Low-Emittance Electron Beams for MeV UED

Joel England (Head of UED Accelerator Operations Group)
Alex Reid (UED Instrument Lead), Mike Minitti (UED Scientific Director), Xiaozhe Shen (UED R&D Lead)

Physics and Applications of High Brightness Beams Workshop, San Sebastian, Spain (2023)

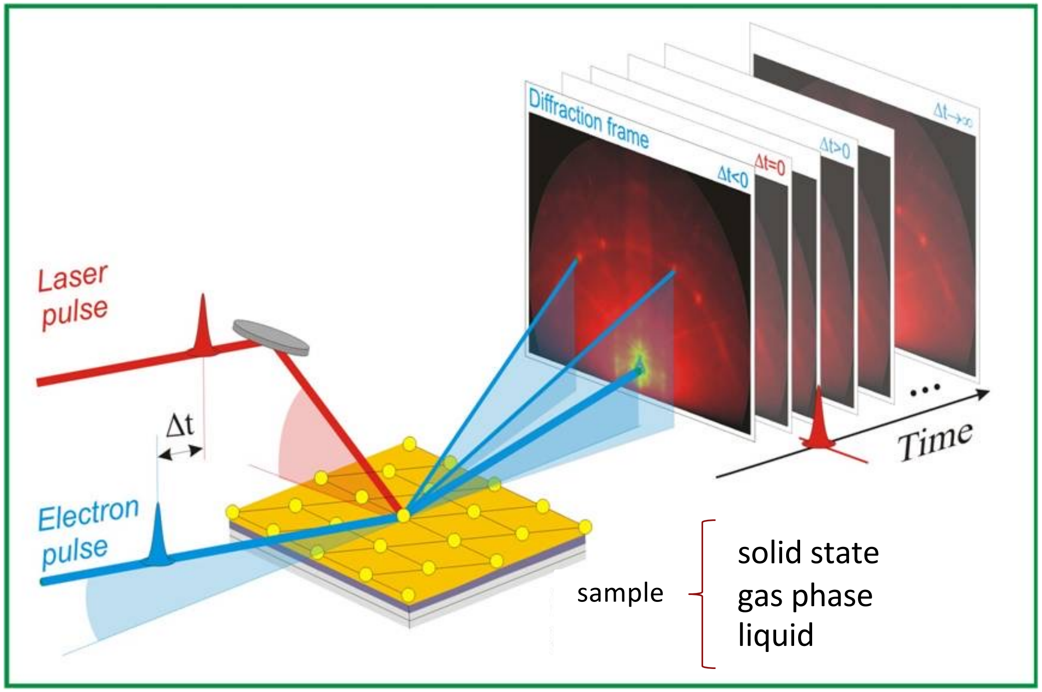
June 19-23, 2023

SLAC NATIONAL ACCELERATOR LABORATORY

Stanford University | U.S. DEPARTMENT OF ENERGY

Ultrafast Electron Diffraction

UED enables measurement of dynamic processes with unprecedented spatial (Å) and temporal (fs) resolution to visualize the ultrafast and ultrasmall



adapted from F. Vigliotti, et al., Angewandte Chemie, 43 (20), 2585 (2004)

Dynamic Processes

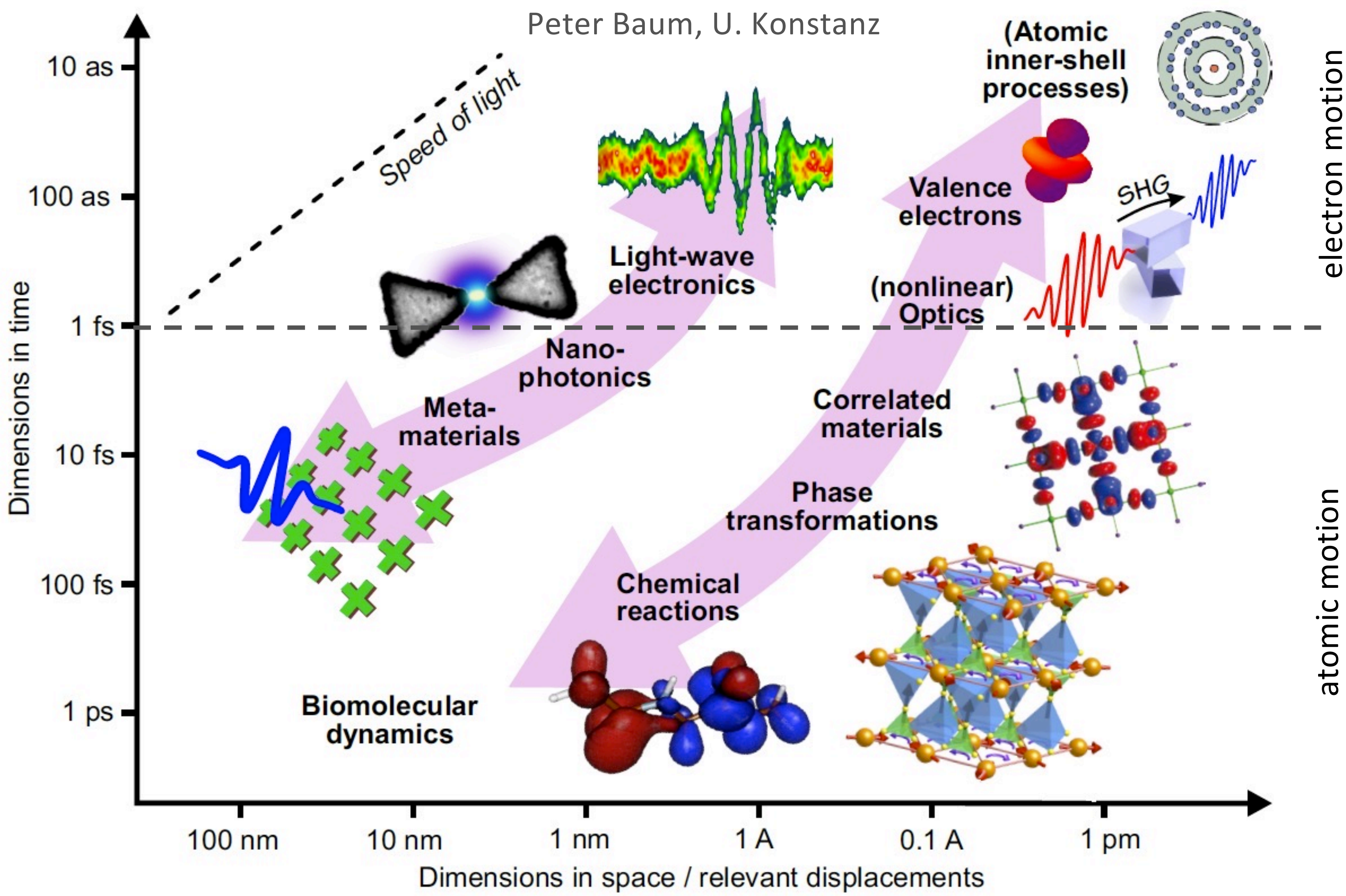
Solid-state: nano-scale, 2D materials, diffuse scattering, strongly correlated systems, functional materials

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

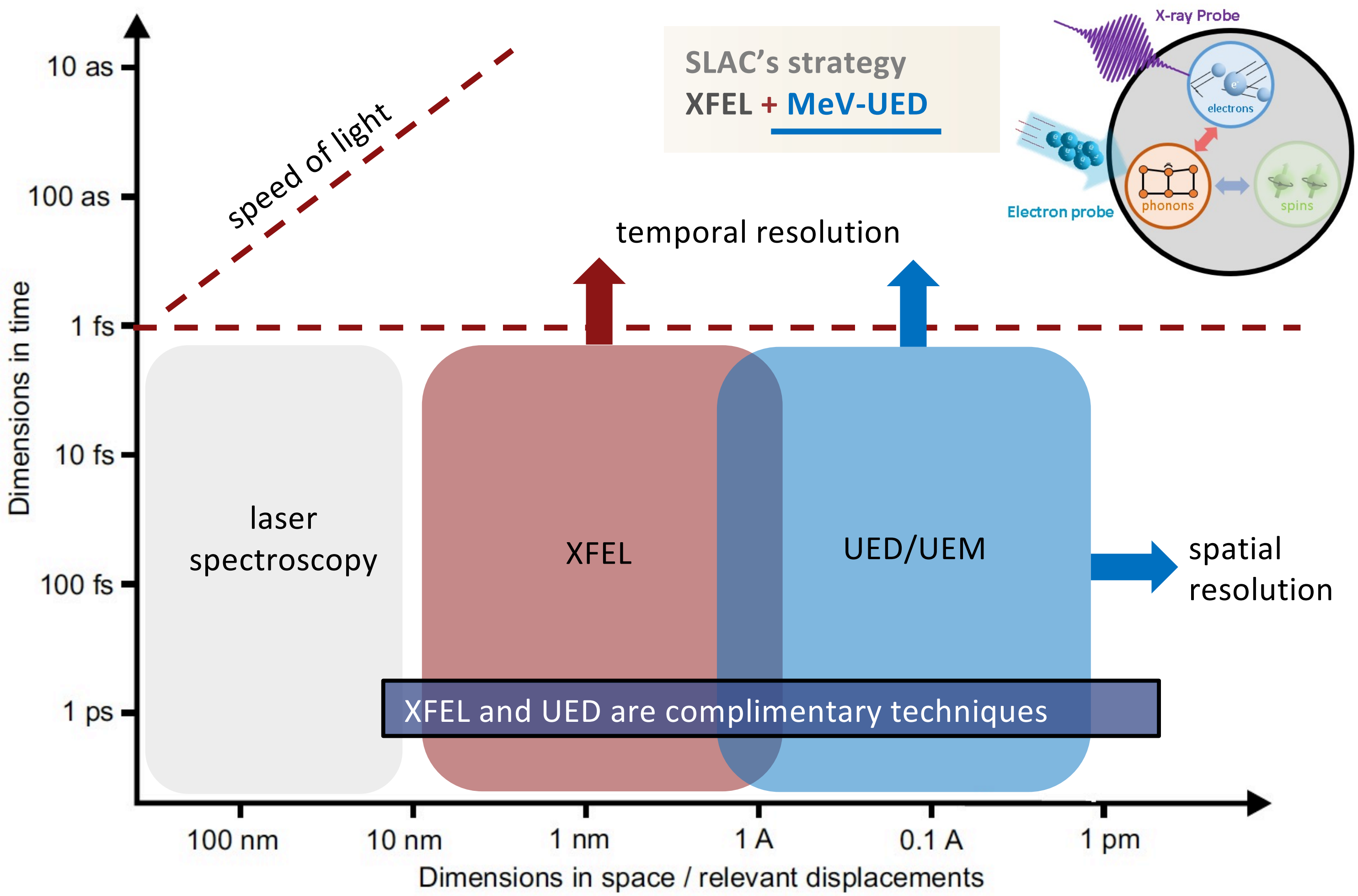
Liquid-phase: photochemistry, photosynthesis, DNA photodamage

Upcoming user run 4 will focus on gas phase dynamics; some liquid phase experiments may be fielded as limited collaborative efforts.

Light-driven Electronic and Atomic Motion



Light-driven Electronic and Atomic Motion

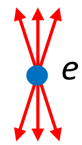


Benefits of MeV vs. keV Electrons

Space charge effects

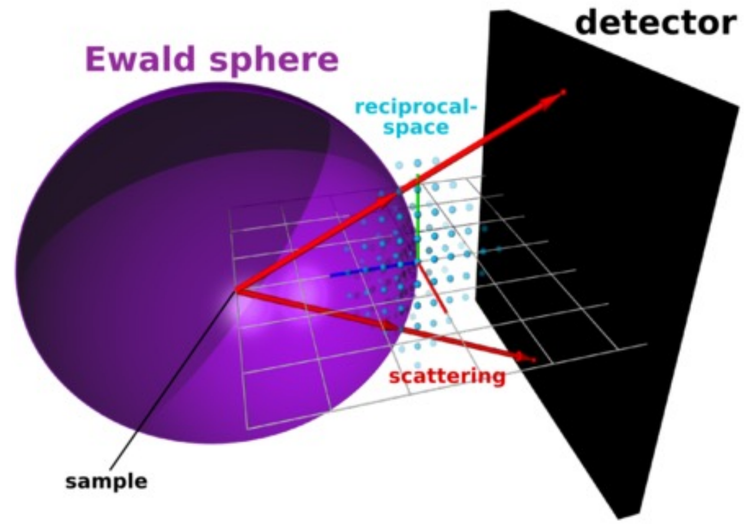
Coulomb repulsion is reduced at higher energy, allowing for higher charge density (and thus shorter bunches)

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



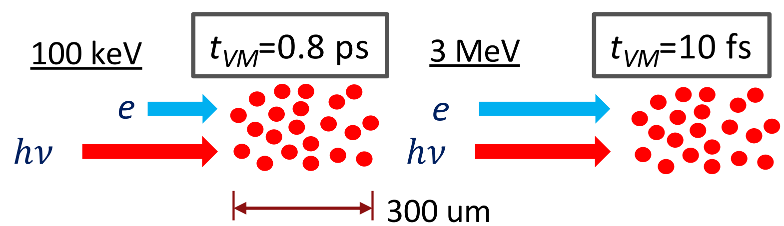
Larger Ewald Sphere

Due to reduced DeBroglie wavelength, Ewald sphere is “flatter” at MeV energies, encompassing more diffraction peaks.



Reduced velocity mismatch

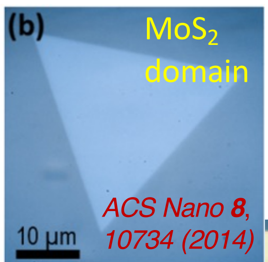
Reduced e- velocity mismatch compared to speed-of-light pump laser improves temporal resolution



Q-Resolution: High-Brightness Beams

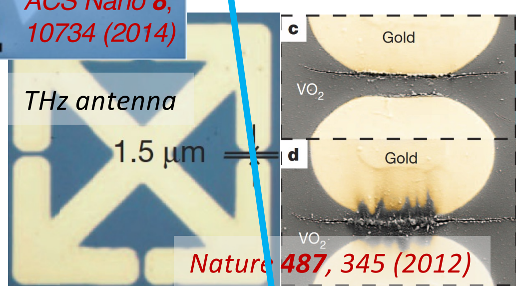
$$\epsilon_n = \sigma_x \cdot \gamma\beta\sigma_{x'}$$

e.g. 0.1 nm-rad = 0.4 um × 0.25 mrad

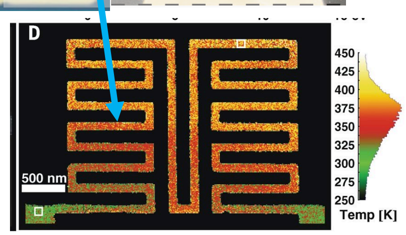


ACS Nano ***8****, 10734 (2014)*

Probe size:
10s of um to um to nm



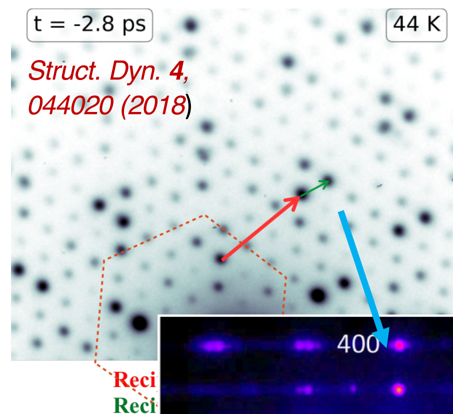
Nature ***487****, 345 (2012)*



Science ***347*** *(6222), 629 (2015)*

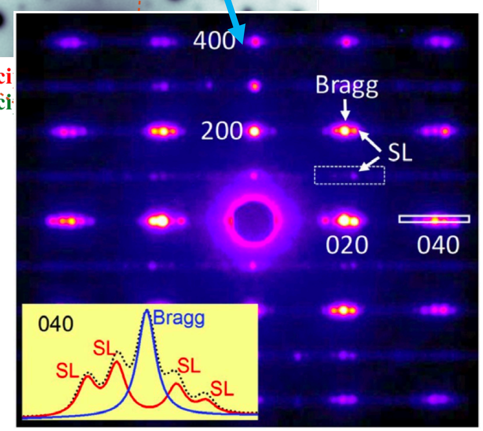
$$\Delta q = \frac{2\pi}{\lambda_e} \cdot \sigma_{x'}$$

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



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

Q-resolution:
need to improve by a few times



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

adapted from slide courtesy Renkai Li

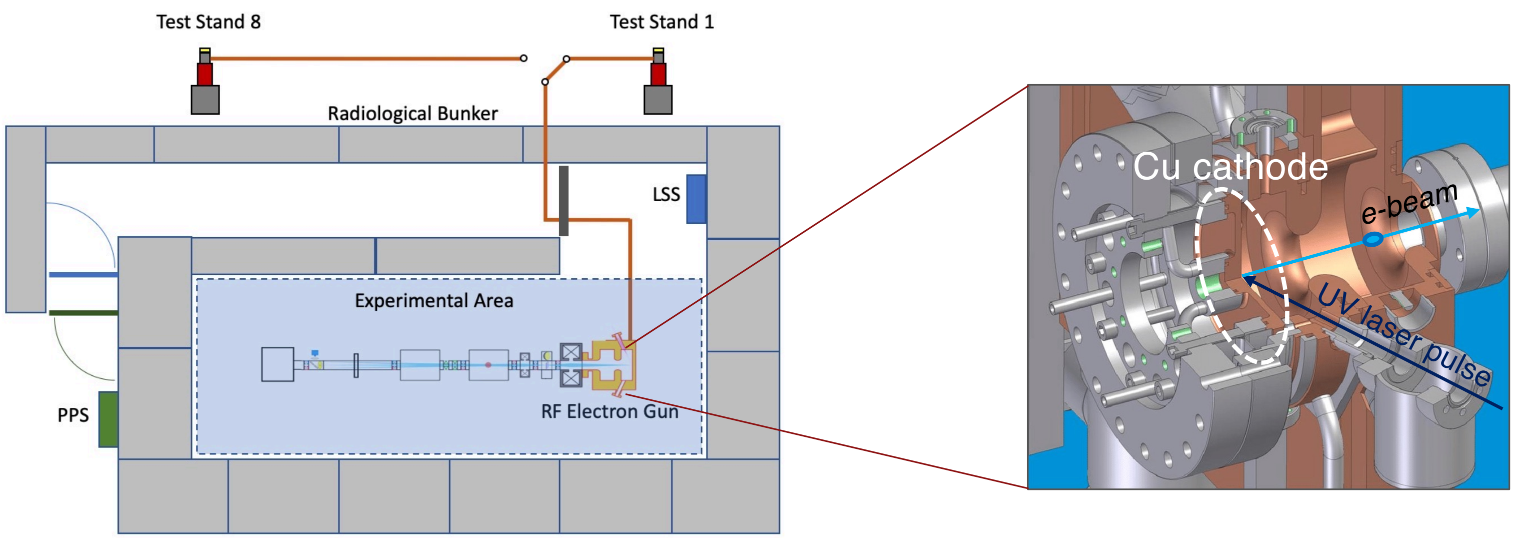
ASTA Photocathode Electron Gun

The radio-frequency (RF) photoinjector is the current state of the art in high-brightness electron beam generation:

→ Can reach relativistic energy in 1.5 RF wavelengths – "strong" acceleration

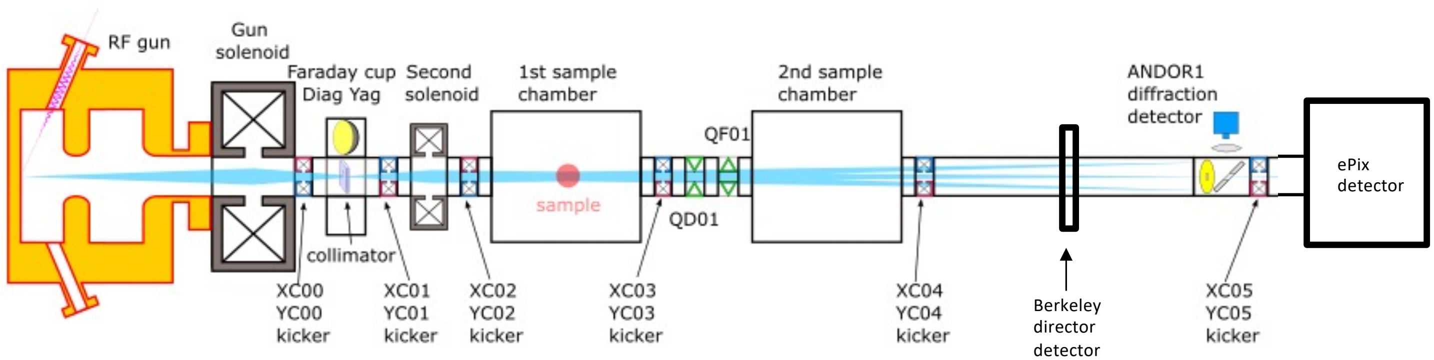
→ Allows emittance compensation of space-charge emittance growth*

The ASTA gun is based on the design for the LCLS injector, operating at 360 pps with an accelerating gradient of 80 MeV/m and exit beam energy of 4.2 MeV.



*Serafini, Rosenzweig, Phys. Rev. E **55**, 7565 (1997)

ASTA UED Experimental Area



S. Weathersby, et al., Rev. Sci. Instr. 86, 073702 (2015), X. Shen et al., Struct. Dyn. 6 054305 (2019)

***q*-resolution**

< 0.17 Å^{-1} FWHM

time-resolution

<150 fs FWHM

L. Le Guyader et al., Struct Dyn. 4 (4), 044020 (2017)

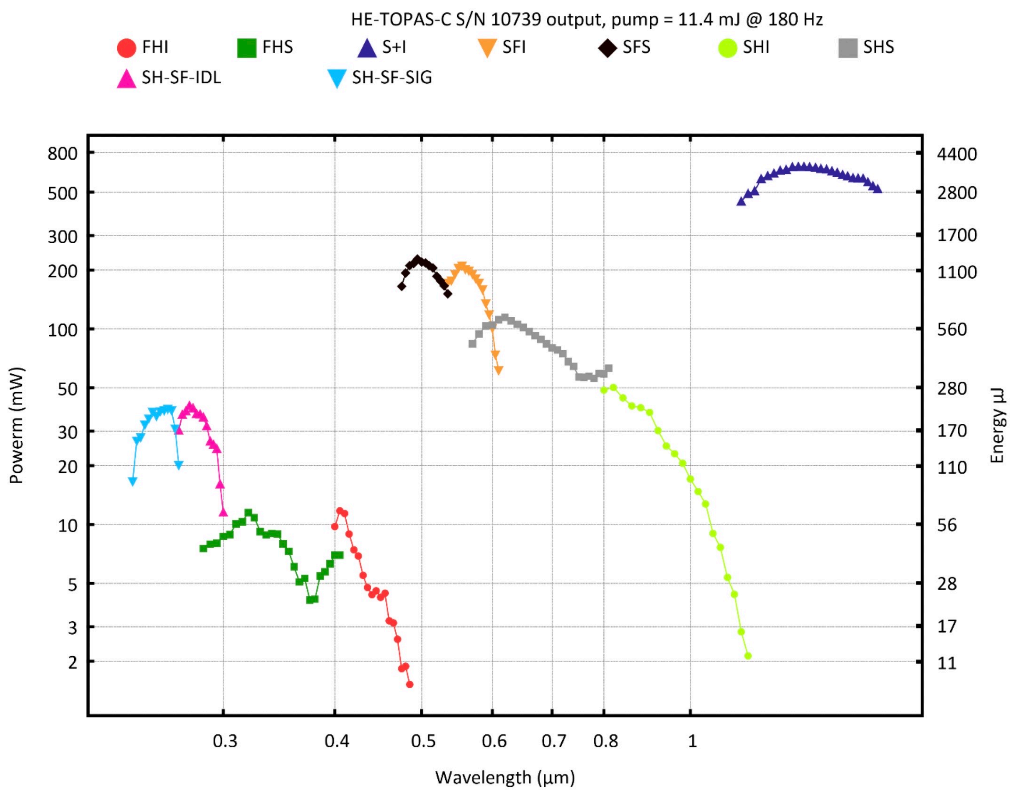
J. Yang et al., Science 368, 885 (2020)

stable over multi-day operation
(typical 5 days continuous 24 hr/day)

Current beam parameters

Parameters	Values
rep. rate	single shot to 360 Hz
beam energy	4.2 MeV
q-range	12 Å^{-1}
bunch charge	10^2-10^6
emittance	0.3 - 20 nm
bunch length	<120 fs FWHM

Pump Laser Wavelengths



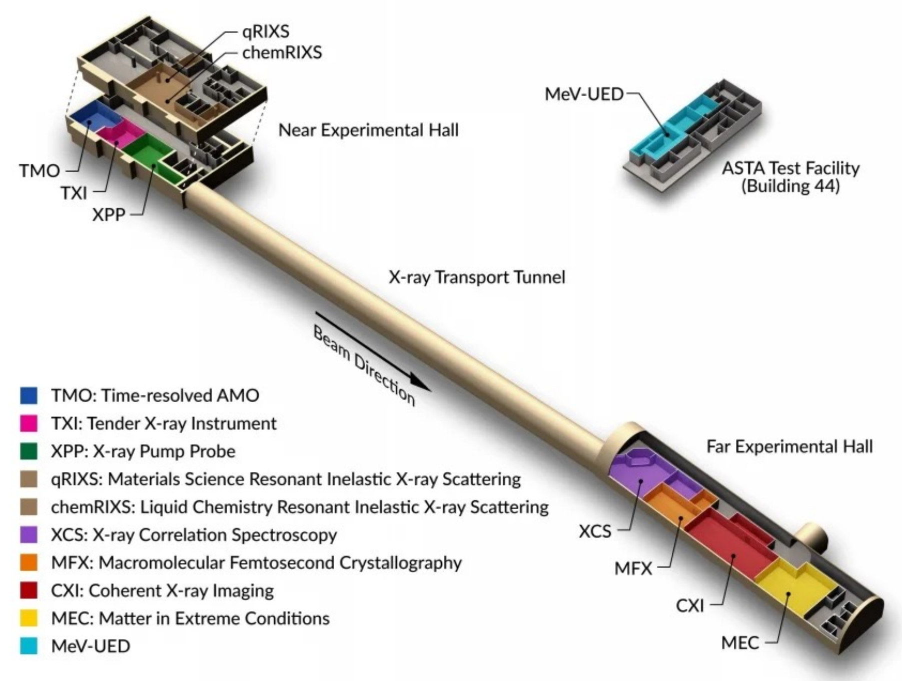
Optical Laser Properties

Parameter	Value
Repetition rate	Single shot → 360 Hz
Wavelength range	200 nm, 266 nm, 400 nm, 800nm, 240 nm - 2 μm tunable, 3-15 μm tunable
Typical optical pulse energy	> 8 mJ @ 800 nm > 0.8 mJ @ 400 nm > 0.08 mJ @ 266 nm ~ 16 μJ @ 200 nm For pulse energies at specific wavelengths (e.g. 260 nm – 750 nm) or other details please contact **Patrick Kramer**
Nominal pulse duration	75 fs (FWHM)
Optical delay	0 - 3 ns (physical delay stage)
Optical spot size	200 - 1500 μm (FWHM)

currently have a gap from 200 to 240nm; DFG solution being developed under OIP funds

MeV-UED: Transition to a User Facility

- Leveraging the strengths of LCLS, became a user facility in 2019
- 3 user runs fielded; Run 4 planned this year
- Modeled operations to focus on science delivery on the big stage
- Large part of MeV-UED publications are classified as DOE defined high impact journals publications. https://oraweb.slac.stanford.edu/apex/slacprod/slacesaf.pubs_ued



MeV-UED has produced high user science output since 2016, including 9 PhD theses and 37 pubs in high-impact journals (IF > 7).

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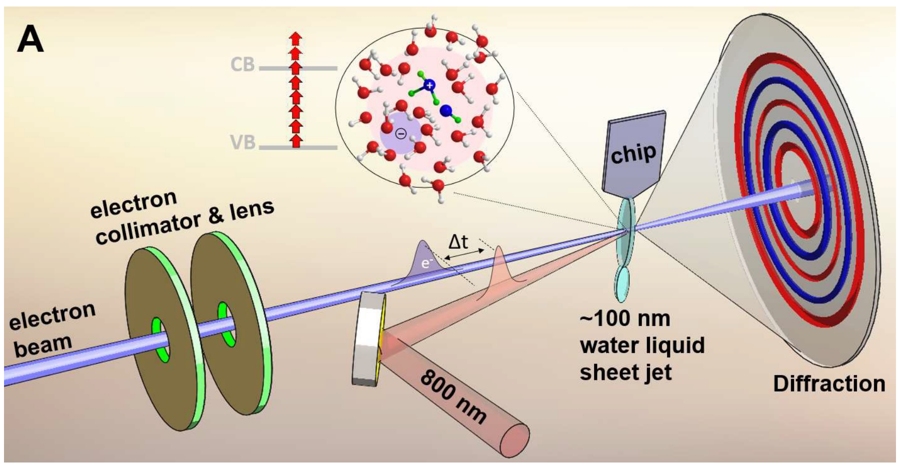
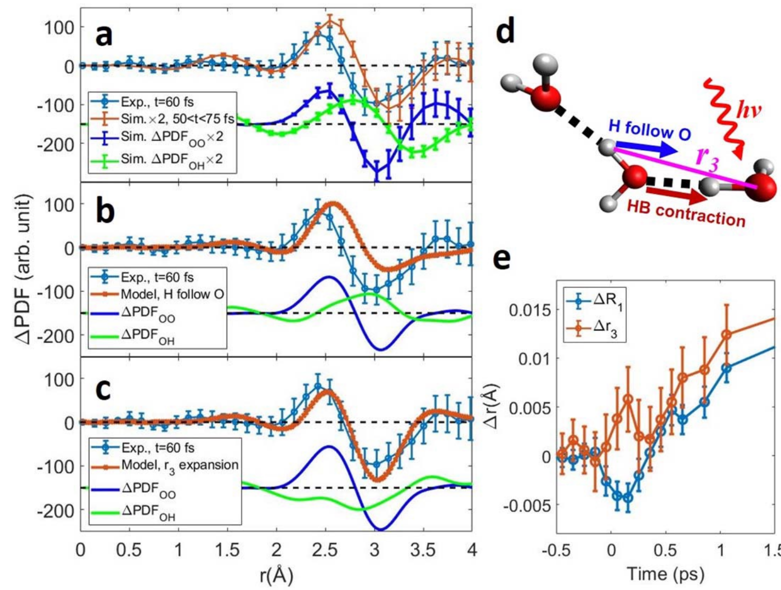
Recent User Science Highlights

M.F. Lin, et al., Imaging the short-lived hydroxyl-hydronium pair in ionized liquid water, *Science, 374*.(6563), 91-95 (2021)



J. Yang, et al., Direct Observation of Ultrafast Hydrogen Bond Strengthening in Liquid Water, *Nature*, 596, 531-535 (2021)



Recent advances in liquid-phase ultrafast electron diffraction techniques make it possible to observe what has only been theoretically presumed to occur at short times upon interaction of ionizing radiation with liquid water.

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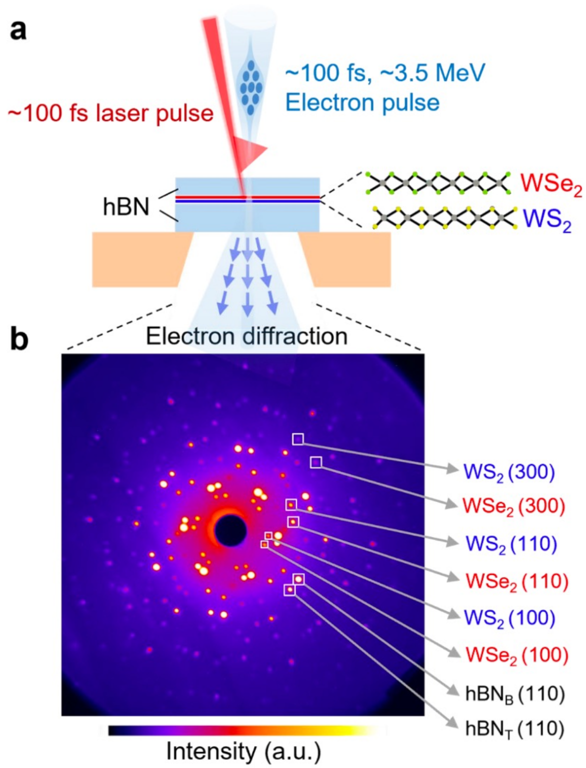
Ultrafast charge and energy transport across two-dimensional atomic junctions



Top: Experimental setup probing dynamic response of 2D van der Waals heterostructures. Bottom: Electron diffraction pattern of heterostructure showing how selective probing of each atomic layer is enabled.

SIMES BERKELEY LAB MOLECULAR FOUNDRY SLAC

Scientific Achievement

We present a new understanding of the microscopic origins of ultrafast charge and energy transport across two dimensional heterostructures

Significance and Impact

This work explains how heat dissipates during ultrafast charge transfer at nanoscale interfaces. This understanding will help with the design of future devices for microelectronics, quantum science and other applications.

Research Details

- The SLAC UED facility was used to selectively probe energy transfer in a photoexcited bilayer of WSe_2 and WS_2 via diffraction techniques.
- Following impulsive photoexcitation of WSe_2, we observed an unexpected concurrent heating of both layers, showing that energy was shared on ultrafast time-scales.
- In combination with first principles theory, we found that this could be understood via a new type of phonon-assisted interlayer charge transfer, involving hybridized electronic states across the bilayer.

A. Sood., J.B. Haber, J. Carlström, E.A. Peterson, E. Barre, J.D. Georgaras, A.H.M. Reid, X. Shen, M.E. Zajac, E.C. Regan, J.Yang, T. Taniguchi, K. Watanabe, F. Wang, X. Wang, J.B. Neaton, T.F. Heinz, A. Lindenberg, F. H. da Jornada, A. Raja, Nat. **Nature Nano. 18, 29 (2023)**

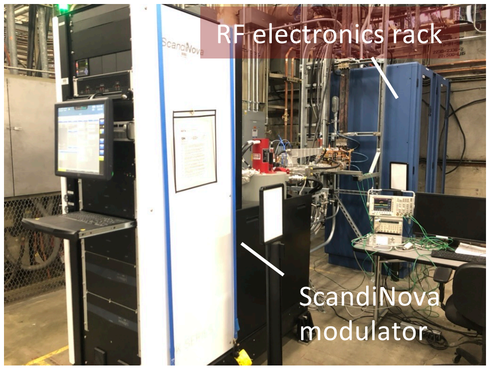
Other MeV-UED facilities are coming online around the world

Country	Facility Name	Electron beam energy range (MeV)	Charge per pulse (fC)	Electrons per pulse	Bunch length (fs)	Repetition rate (Hz)	Start date
Germany	DESY - REGAE	2 - 5	<100 - 1000	10^4 - 10^6	7 - 30	50	2011
China	Tsinghua Thomson scattering X-ray source (TTX*)	2 - 4	10^4 - 10^5	10^8 - 10^9	1000 - 3000	5 - 10	2013
	SHINE**	0.75	1000 - 10^4	10^8 - 10^9	1000	10^6	2025
United States	SLAC MeV-UED	2 - 4	1 - 100	10^4 - 10^6	<150***	1 - 360	2014
	SLAC MeV-UED	2 - 3	1 - 100	10^4 - 10^6	<150***	1000	2023
*	https://indico.cern.ch/event/577810/contributions/2479863/attachments/1424734/2185126/2017-CLIC_WS-Thomson_Scattering_X-ray_Source_at_Tsinghua_University.pdf						
**	http://linac2018.vrws.de/talks/mo2a01_talk.pdf (FEL Source)						
***	Nominal. Beam charge dependent						

Growing international interest in MeV-UED with new facilities coming online. Not a comprehensive listing. Other UED programs at BNL, Berkeley, UCLA, LANL, and Daresbury (UK).

ASTA / MeV-UED: Current Status / Recent Progress

Upgrade to 1080 Hz Pump-Probe Acquisition Rate

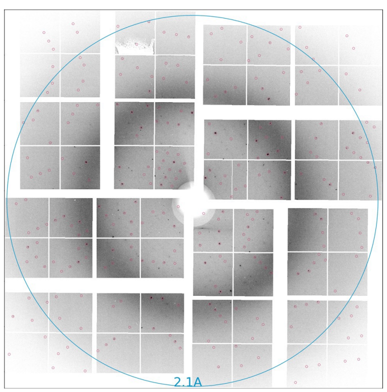


ScandiNova RF Klystron (S-band 2.856 GHz @ 1080 pps)

- Fully installed and in state of technical readiness; conditioned to full power on an RF load in 2022
- Further testing at 1 kHz planned with electron beam characterization following ARR and approval to restart.

Ti:Sapphire Coherent Legend Elite Duo + SPA

- Infrared laser upgrade completed in April 2021 (regen + single pass amp).
- Successfully operated through Run 3.



ePix Direct Electron Detector

- Upgrade components for 1 kHz ePix have been delivered; will include e-beam thru-hole
- Working with detector group to implement as an independent detector (anticipated 6 month time scale)
- Existing 360 Hz version was commissioned in Run 3 and will be on offer in Run 4 on a by-request basis while the upgraded version is being built

MeV-UED Instrument Retreat

- Brought together the UED instrument team and other stakeholders for a one-day focused workshop

- **Three focused sessions:**
  Science, Research & Development, and Operations
  3-member committees set program for each session
- **Charge:**
  "Define the science, development and operational priorities that will drive continued scientific excellence at the MeV UED Instrument in the LCLS-II era"
- **Participants:**
  Instrument science experts
  UED machine development experts
  UED operations experts
  SRD leadership
- **Followed-up with report & recommendations for the instrument**

MeV-UED Instrument Retreat

Thursday March 2nd – Sycamore Room Bld 40 R195

Agenda

8:20	Welcome and Retreat Charge	Alex Reid
8:30	**Science on Session: Introduction**	Yusong Liu
8:35	UED Gas phase science	Thomas Wolf
9:05	UED Solid phase science	Alex Reid
9:35	UED Liquid phase science	Ming-Fu Lin
10:05	UED Warm Dense Matter science	Mianzhen Mo
10:35	Wrap up	Yusong Liu
10:40	Coffee	
11:00	**Research and Development Session: Introduction**	Xiaozhe Shen
11:10	High-density radical source for UED gas phase photochemistry	Ming-Fu Lin
11:35	Enhanced laser sources and beam delivery	Patrick Kramer
12:00	Electro-Optic, mm-wave timing tool for UED	Stephen Weathersby
12:25	Intelligent beamline R&D for MeV-Ultrafast Electron Diffraction	Fuhao Ji
12:50	Wrap up	Xiaozhe Shen
13:00	Lunch at SLAC Café	
14:00	**Session on Operations: Introduction**	Antonio Gilardi
14:05	UED Facility Operations and Deliverables	Mike Minitti
14:30	UED from a POC prospective	Xinxin Cheng
14:55	UED Safety and Work Planning	Stephen Weathersby
15:20	UED Controls & Data Outlook	Antonio Gilardi & Silke Nelson
15:45	UED Engineering Outlook	Mason Landrum
16:10	Wrap up	Stephen Weathersby
16:15	Coffee	
16:30	Open Discussion Session	
17:10	Closing Remarks	Alex Reid

In-person RSVP: Alex Reid alexhmr@slac.stanford.edu
Zoom: https://stanford.zoom.us/j/6030938400?pwd=QmMyMG5RSktwL1pvRDFXRGRSRDlGQT09

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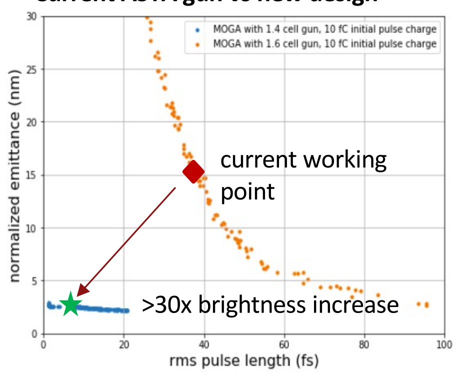
ASTA / MeV-UED: Mid-Term (1-2 Year) R&D Plans

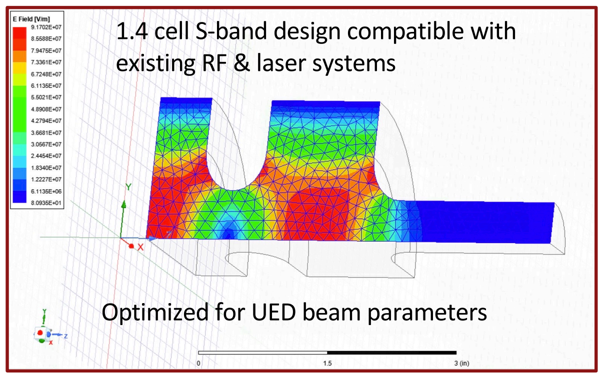
Critical Mid-Term R&D goals identified from recent UED strategic planning efforts:

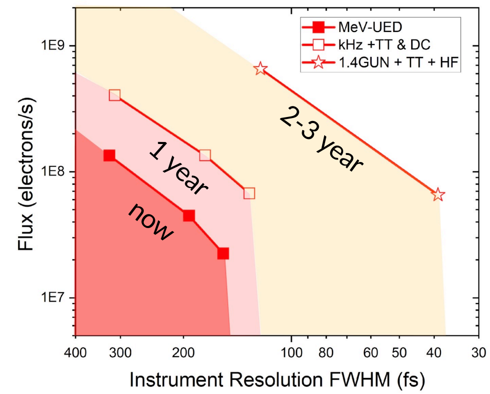
- Improve instrument time resolution towards 50 fs
- Increase electron flux to > 1e8 electrons/sec
- Improve tranverse emittance to reach $\Delta q = 0.01$ Å^{-1}

To meet future user requirements on electron flux, spot size, and time resolution, a new higher brightness, lower emittance electron source is needed

Current ASTA gun vs new design







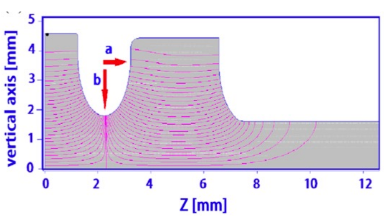
Mid-Term (1-2 Year) Goals / Milestones	Funding	Key Personnel/Responsibilities
Prototype of 1.4-cell S-band gun design	TBD	UED AD with Test Fac and TID RFAR support
Dedicated online (shot-to-shot) THz time-tool	TBD	UED AD & LCLS team with Laser & Nanni Groups
Upgrade of the ePix detector to kHz rep rate	DOE-BES	UED AD & LCLS team with TID Sensor Group
Laser DFG wavelength extension for UV pump	OIP	UED LCLS with Laser group support

Prior art: 1.4 Cell S-band RF gun

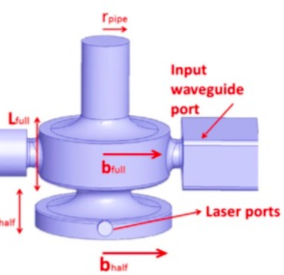
S-band 1.4 cell photoinjector design for high brightness beam generation

E. Pirez [a], P. Musumeci [a], J. Maxson [a], D. Alesini [b,*]

[a] Department of Physics and Astronomy, UCLA, Los Angeles, CA 90095, USA
[b] INFN-Laboratori Nazionali di Frascati, Via Enrico Fermi 40, 00044 Frascati, Rome, Italy



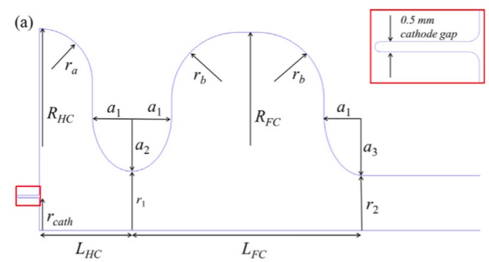
r_{pipe}	18mm
L_{full}	53.04mm
L_{half}	26.17mm
b_{full}	43.745mm
b_{half}	43.0618mm

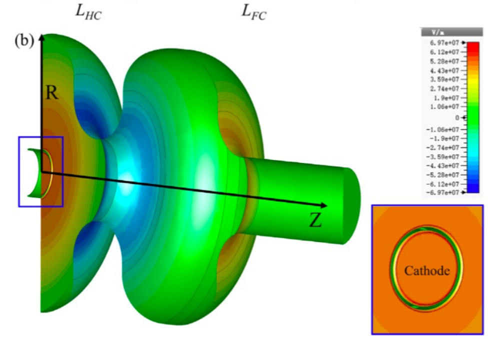


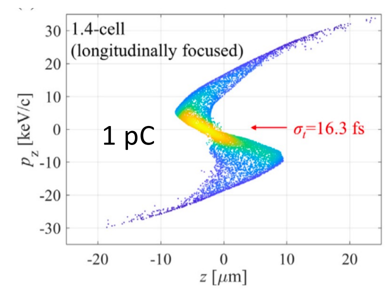
$$\mathcal{B}_{4D} \propto \frac{E_z}{\mathrm{MTE}} \qquad A = \frac{\sigma_x m_e}{\sigma_t^2 E_z e}$$

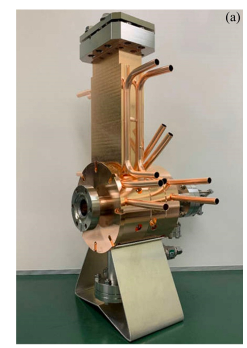
for pancake (A>>1) beam

I. Bazarov, B. Dunham, and C. Sinclair, *PRL 102, 104801(2009)*









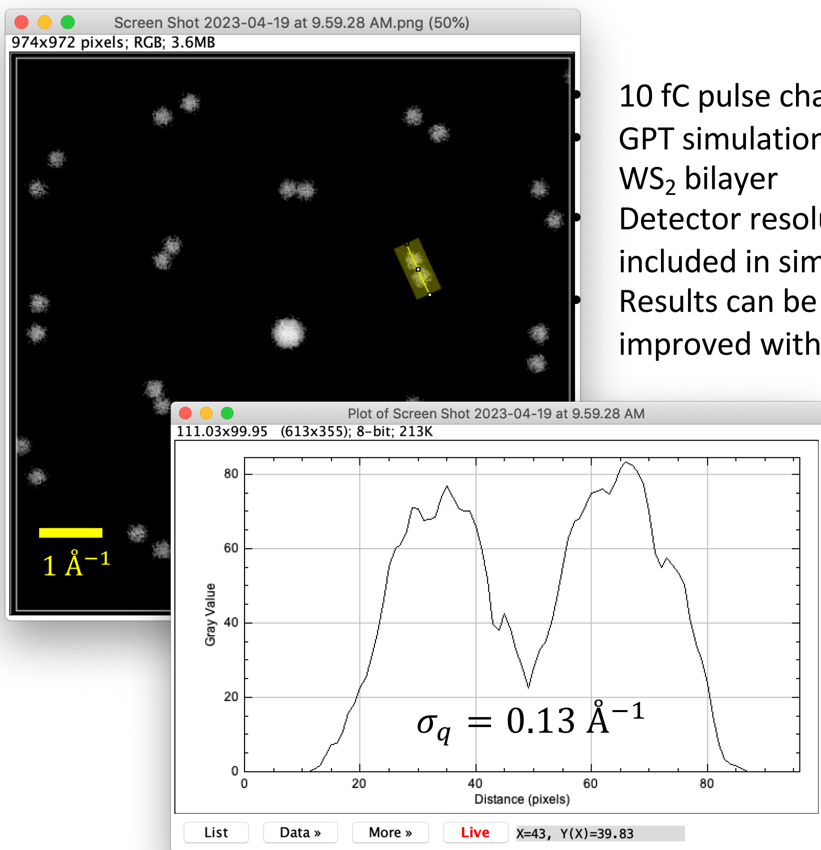
Y. Song, et al., Nuclear Inst. and Methods in Physics Research, A 1031 (2022) 166602.

- For 1.6 cell gun, launching phase is ~37 degree -> 54 MV/m acc field at cathode
- Shorter cathode cell yields larger acc field at cathode

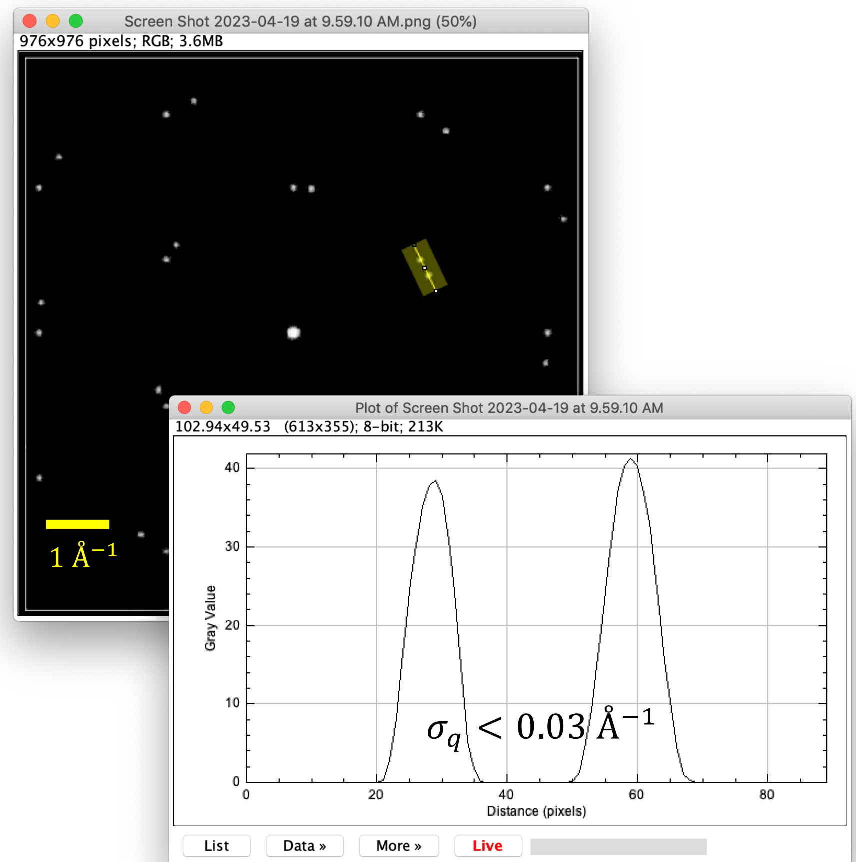
ASTA / MeV-UED: Mid-Term (1-2 Year) R&D Plans

Simulated q-resolution enhancement with new high brightness gun design

Current ASTA 1.6 Cell Gun → **Alternative 1.4 Cell Gun**



- 10 fC pulse charge (both cases)
- GPT simulation of 7 deg rotated WS_2 bilayer
- Detector resolution not included in simulation
- Results can be further improved with collimation



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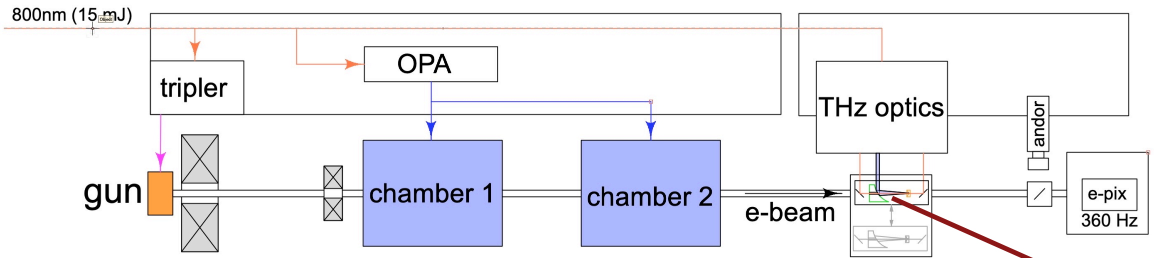
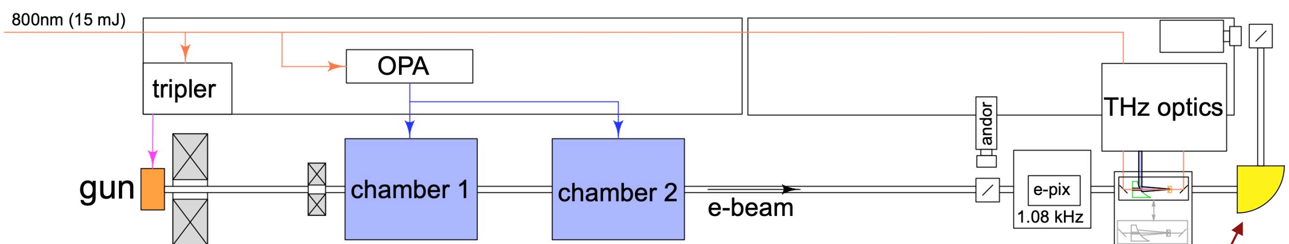
ASTA / MeV-UED: Mid-Term (1-2 Year) R&D Plans

Implementation of Dedicated THz Time-of-Arrival Diagnostic

Phase I: Interruptive Diagnostic



Phase II: Shot-to-Shot Diagnostic

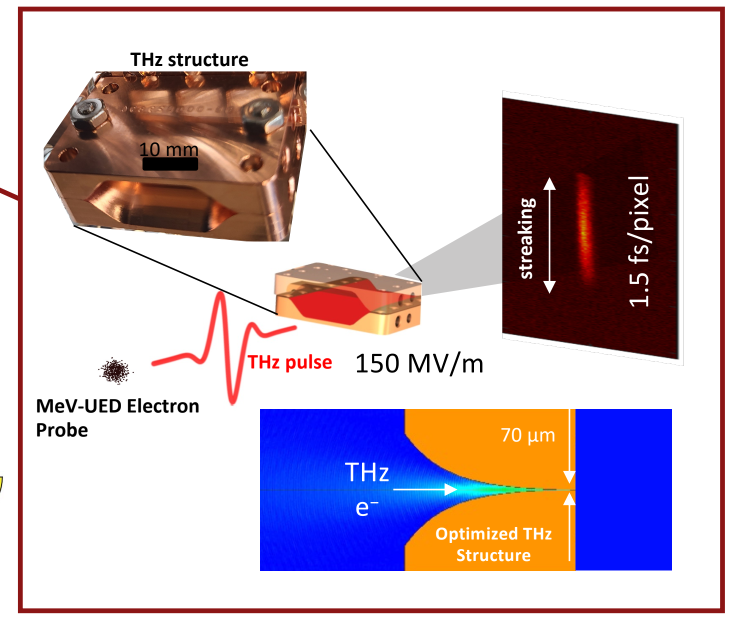


new ePix with thru-hole

spectrometer magnet

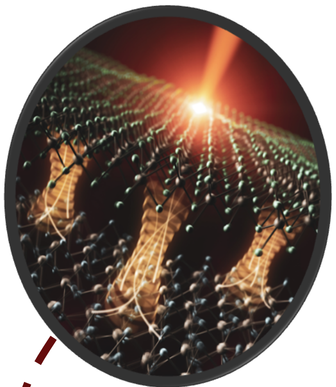
Proof of Principle demonstrated in prior runs:

- Enables simultaneous fs arrival time and bunch length measurement
- Developed in collaboration with Nanni group
- Robust performance under insert/remove actuation

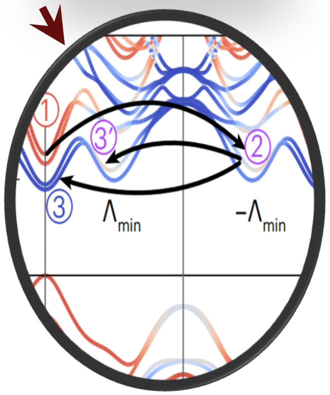


M. Othman, et al., Efficient THz Time Stamping of Ultrafast Electron Probes, 47th IRMMW-THz Conference (2022).

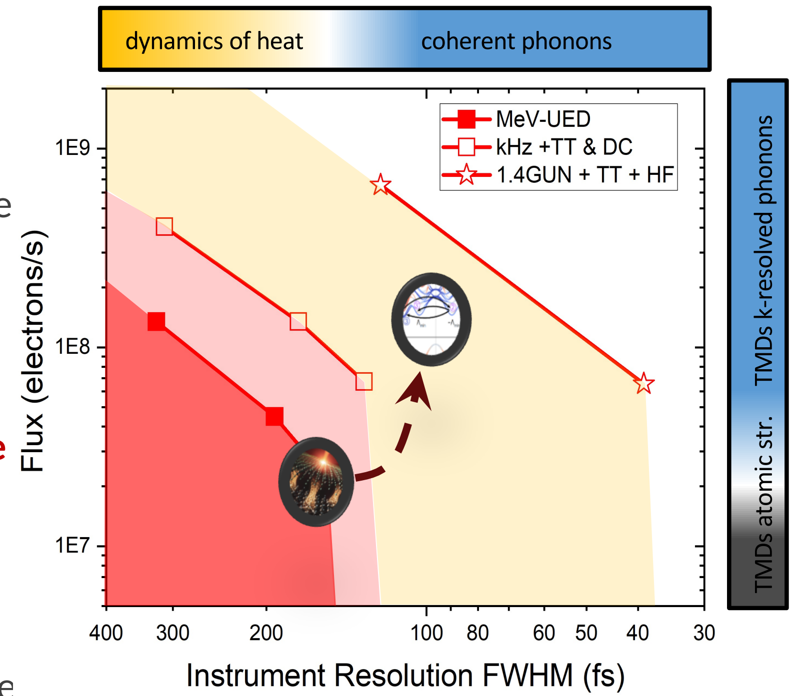
MeV-UED for 2D-materials based Microelectronics



towards



- **UED Highlight – Heat transfer across van der Waals heterostructure – Sood *et al.* Nature Nano. 18, 29 (2023)**
- **UED – 150 fs, ~2e7 electrons/s**
- Benchmarked thermal transport across a monolaye WSe_2/WS_2 heterostructure using Debye Waller effect
- Uncovered ultrafast *thermal bridge* via phonon-assisted charge transfer
- **Breakthrough – *Directly map phonons* enabling the charge transfer process for microscopic understanding**
- **UED – 100 fs, ~1.3e8 electrons/s**
- A new high-brightness UED enables diffuse scattering from monolayers for momentum- & time resolved phonon measurement
- Microscopic verification of intervalley mediated transfer hypothesis



- Diffuse-scattering phonon measurements are only possible for films 10s nm in thickness (e.g. 20 nm Ni). A ten-fold increase in electron flux enables measurements in monolayers.
- Temporal resolution of 100 fs enables separation of the interlayer transfer from e-ph scattering within a layer
- MeV penetration depth paired with low energy deposition per scattering event enable future in-operando studies

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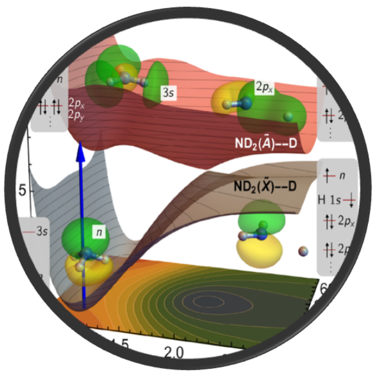
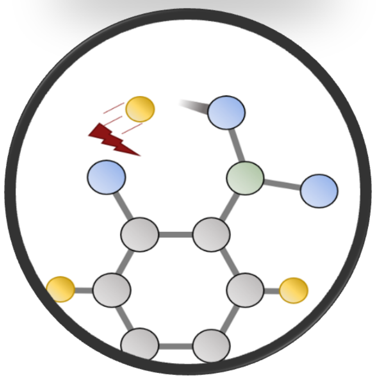
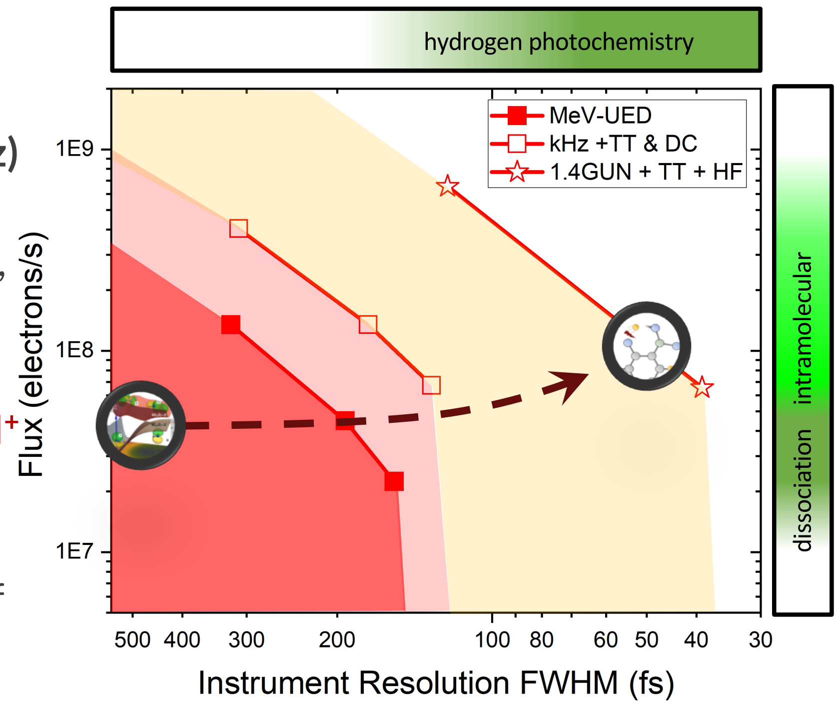
MeV-UED for understanding ultrafast photochemistry





- **UED Highlight –Photodissociation in Ammonia E. Champenois *et al.* arXiv:2303.03586**
- **UED – ~500 fs, ~4e7 electrons/s (180 Hz)**
- Clear signature from loss of a nitrogen-deuterium bond in ND_3 photodissociation, but dissociation not yet resolved in time
- **Breakthrough – Resolve intramolecular H^+ transfer in 2-nitrophenol**
- **UED – 50 fs, ~1e8 electrons/s**
- Map electronic and structural dynamics of proton transfer in a single measurement
- Electronic dynamics from inelastic scattering
- Structural dynamics from elastic scattering



- 70 fs temporal resolution required to follow H+ photodissociation
- ~2 x electron flux required to resolved intramolecular H+ dynamics

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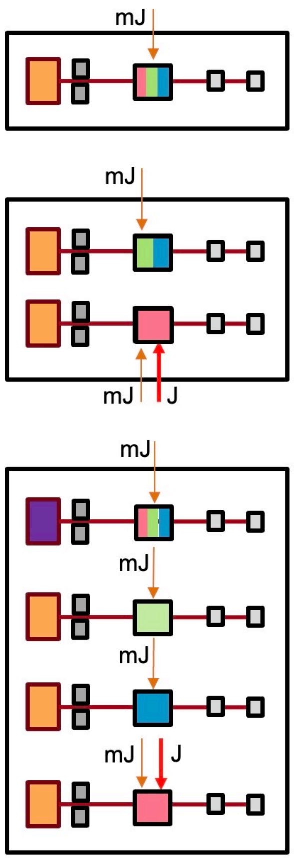
ASTA / MeV-UED: Long-Term (5-10 Year) Roadmap*



Near Term

Long Term

Single Multipurpose Beamline (1-2 years)

Solid → Gas → Liquid, Bio

Improved temporal and q-resolution:

- Single shot THz streaking diagnostic
- Single electron detection for high SNR
- High repetition **RF** operation (**kHz**)
- Development of new 1.4 cell gun prototype

Additional Beamline (3-5 years)

- Testing/commissioning of 1.4 cell gun
- Gun R&D → User operations
- Two lines (Gas + Liquid/Bio) & (Solid + MEC)
- Increased R&D and User capacity

UED Farm (5-10 years)

- Dedicated instruments for Solid/Liquid/Gas
- Dedicated lasers: IR, TW/Joule-class, THz
- Diversified beam lines for User + R&D
- **SRF** or APEX-like **VHF** gun (**kHz** → **MHz**)

*R. J. England, A. Reid, X. Shen, M. P. Minitti, *White Paper: Expansion of MeV UED Facility at SLAC, March 23, 2022*

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Summary & Conclusions

- Critical needs identified from UED strategic planning efforts and user feedback:
  - Improved electron source performance to achieve ≤0.01 $Å^{-1}$ momentum resolution, ≤50 fs temporal resolution, and ≥1e8 e-/sec flux imply need for higher beam brightness
  - Increase user support capacity with diversified beamlines for solid, liquid, gas phase, and MEC experiments and more efficient R&D development

- Machine development R&D to meet the above needs:
  - Near-Term (1-2 years): upgrade existing beamline with shot-to-shot THz TOA and kHz RF and detector rep rates
  - Mid-Term (3-5 years): additional user beamline based on optimized 1.4 cell S-band gun for improved beam brightness
  - Long-Term (5-10 years): multi-beamline facility using combination of S-band and SRF or VHF sources (leveraging LCLS-II-HE gun development)

- Note on broader interests:
  - MeV-UED facilities coming online at DESY REGGAE, Tsinghua TTX, SHINE, BNL
  - MeV-UED directly complements LCLS-II HE (sensitivity nuclear and electronic distributions, low energy deposition, and high cross section)

- Synergy and Collaboration:
  - Seeking methods and techniques to maintain MeV-UED as a prominent pump-probe technique.
  - Strong synergy with other SLAC programs leveraging LCLS injector, laser, and detector development.

Acknowledgements

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The MeV-UED team


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Alex Reid


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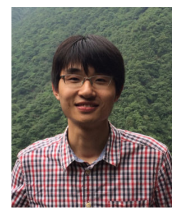
Xiaozhe Shen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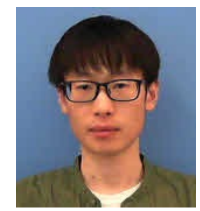
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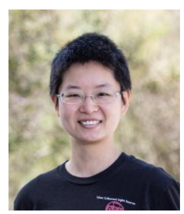
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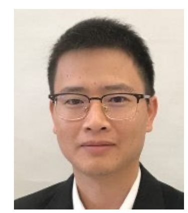
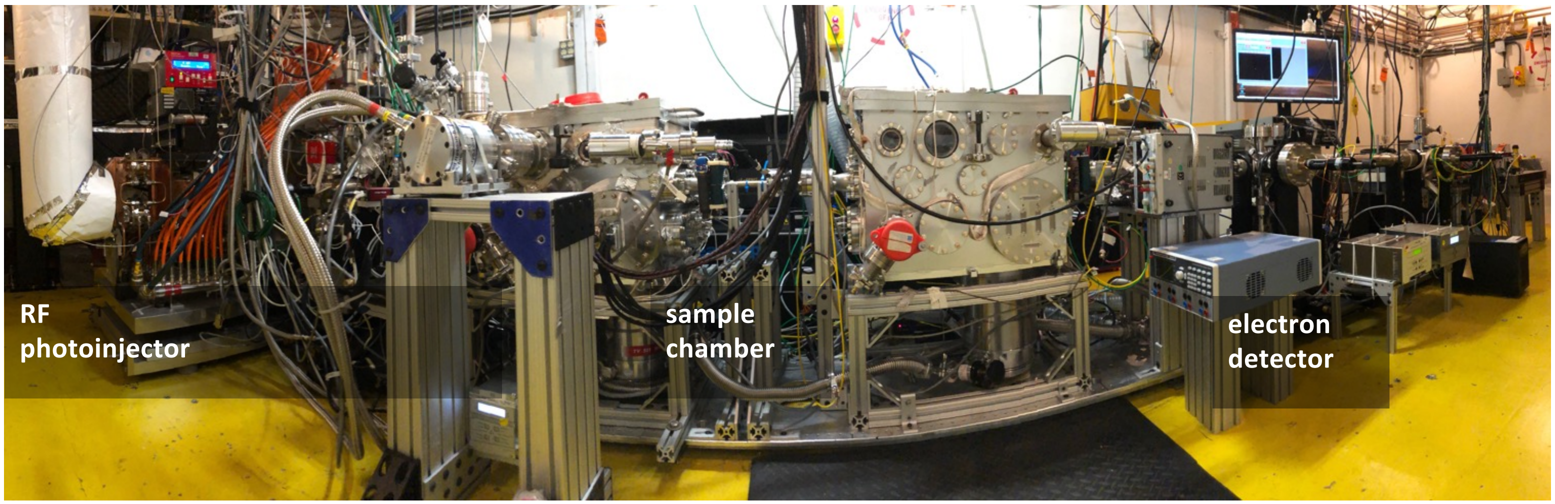

Mianzhen Mo


Mason Landrum

Thank You!



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