Science Frontiers of Megaelectronvolt Ultrafast Electron Probes

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SLAC National Accelerator Laboratory

10th International Particle Accelerator Conference
MELBOURNE CONVENTION & EXHIBITION CENTRE, AUSTRALIA
19 – 24 MAY 2019

U.S. DEPARTMENT OF ENERGY | Stanford University

IPAC19

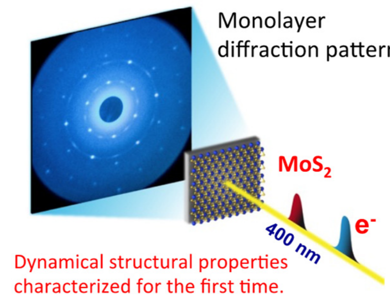
SLAC NATIONAL ACCELERATOR LABORATORY

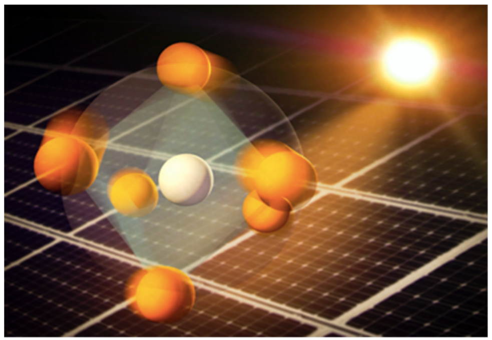
MeV-UED is pushing the frontiers of ultrafast Science

SLAC

✓ *Structure dynamics in 2-D materials*

- ❖ Dynamic structural response and deformations of monolayer MoS2 (Nano Lett.15 6889 (2015)).
- ❖ Ultrafast non-radiative dynamics of atomically thin $MoSe_2$ *(Nature Communications* ***8*** *(2017)*.



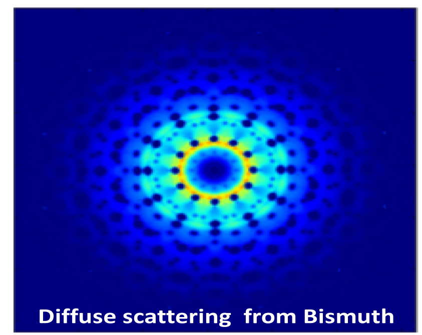


✓ *3-D structure Dynamics & Atomic Movie*

- ❖ Atomic movies may help explain why perovskite solar cells are more efficient (*Sci. Adv.* ***3*** *e1602388 (2017)).*
- ❖ Atomic view of magnetostriction *(Nature Comm. 9 (2018)).*
- ❖ *Heterogenous to homogenous melting observed (Science 360 1451–1455 (2018)).*

✓ *Diffuse scattering & energy relaxation*

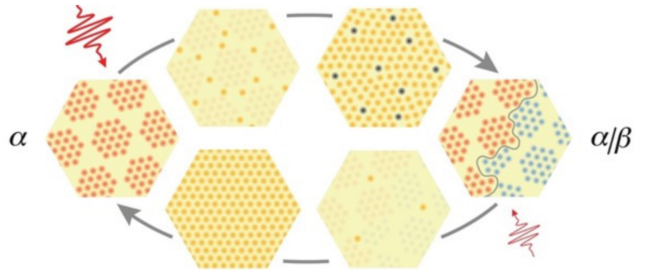
- ❖ Optical excitation and thermalization of phonons in Au *(APL108 041909 (2016)).*
- ❖ Energy transfer and relaxation in nano-scale heterostructure (*Struct. Dyn. 4, 054501 (2017)).*
- ❖ Energy flow in the high-Tc cuprate superconductor *(Sci. Adv. 2018;4:eaap7427).*

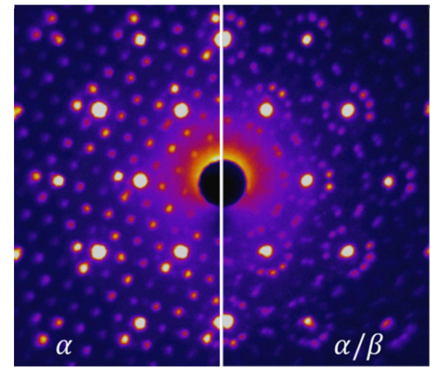


SIMES · Stanford University · Argonne · BROOKHAVEN NATIONAL LABORATORY · MIT Massachusetts Institute of Technology · PENNSTATE · UNIVERSITÄT DUISBURG ESSEN · MAGICS

MeV-UED is a discovery tool: discovering novel quantum phases through coherent light-matter coupling

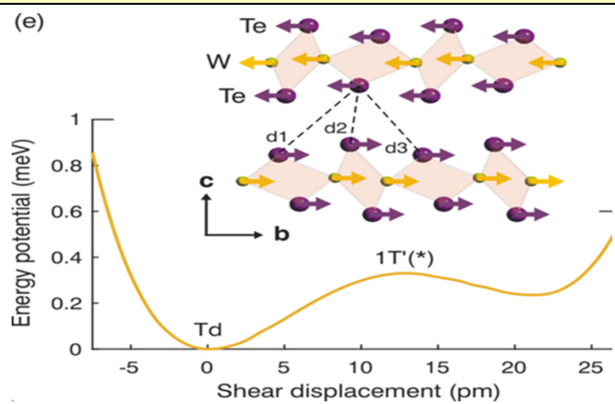
CDW material TaS_2 switch from one state to another with a single flash of light

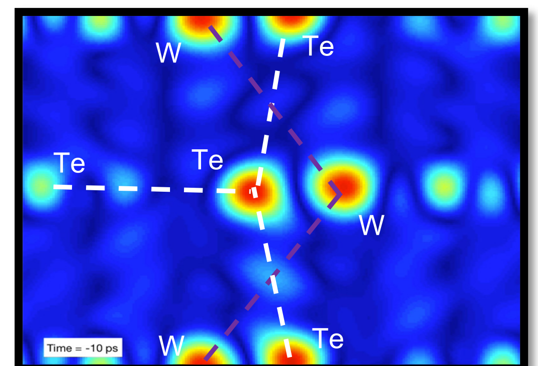




Zong & Shen et al, ***Science Advances***
19 Oct 2018:Vol. 4, no. 10, eaau5501

Time-varying shear strain as an ultrafast topological switch in a Weyl semimetal





E. Sie et al. ***Nature*** 565, 61-77 (2019)

SLAC MeV-UED: A tool for ultrafast science communities

SLAC

- Femto-sec MeV UED with ***24/7*** operation.
- > ***50*** ultrafast material science experiments.
- ***15*** molecules investigated.
- ***8*** single-shot warm dense matter UED experiments.
- **Time-resolved electron diffuse scattering.**
- **Femto-second gas phase/liquid UED.**
- ***30 publications (2 Science, 1 Nature &Nature Chem., 4 Science Adv., 3 Nat. Comm., 1 PRL).***

https://oraweb.slac.stanford.edu/apex/slacprod/slacesaf.pubs_ued

LCLS | Linac Coherent Light Source

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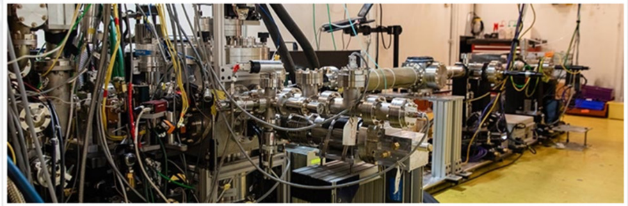
SLAC MeV-UED

INSTRUMENTS

- AMO - Atomic, Molecular & Optical Science
- CXI - Coherent X-ray Imaging
- MEC - Matter in Extreme Conditions
- MFX - Macromolecular Femtosecond Crystallography
- SXR - Soft X-ray Materials Science
- XCS - X-ray Correlation Spectroscopy
- XPP - X-ray Pump Probe
- SLAC MeV-UED
  - MeV-UED Specifications
  - MeV-UED Schematics
  - MeV-UED Endstations
  - MeV-UED Proposals
  - MeV-UED Proposal Review Process
  - MeV-UED Publications
- LCLS-II Instruments (L2SI)



SLAC Megaelectronvolt Ultrafast Electron Diffraction Instrument: MeV-UED

Short Description

The MeV-UED instrument, part of the LCLS User Facility, is a powerful "electron camera" for the study of time-resolved, ultrafast atomic & molecular dynamics in chemical and solid-state systems. Taking advantage of the LCLS-II upgrade period, MeV-UED has been established based on the successful research program at SLAC using MeV electron diffraction. This instrument has demonstrated the following properties: high spatial resolution (< 0.5 Å), large momentum-transfer range (0.5 to 12 Å-1), high elastic scattering cross sections, high temporal resolution (< 150 fs FWHM), with the additional benefits of relatively large penetration depths (> 100 nm) and negligible sample damage. See current **specifications**.

Since 2014, the SLAC UED instrument has been developing robust methods in the pursuit of time-resolved measurements to control and understand molecular structural dynamics and the coupling of electronic and nuclear motions in a variety of material and chemical systems. Driven by a broad array of collaborating teams at SLAC and around the world, the UED instrument has produced an impressive array of high impact **publications**. Examples range from imaging the motions of monolayer MoS_2, to atomic level movies of light-induced structural distortion in perovskites solar cells, along with molecular movies of chemical bond breaking, ring-opening, and nuclear wavepacket at

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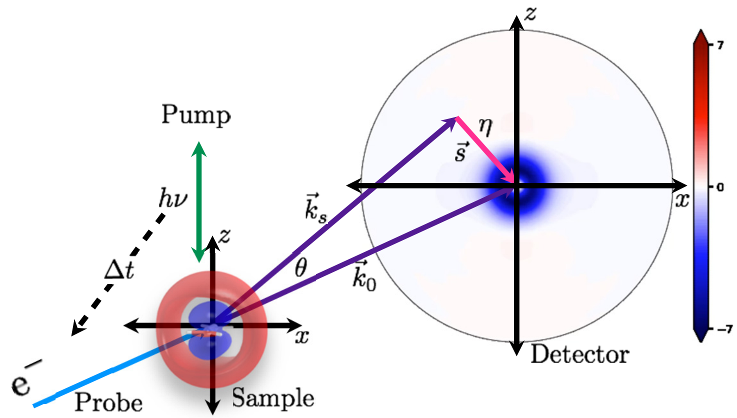
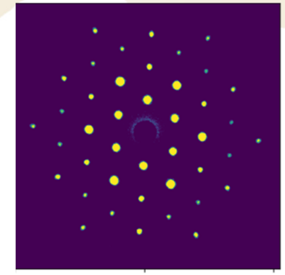
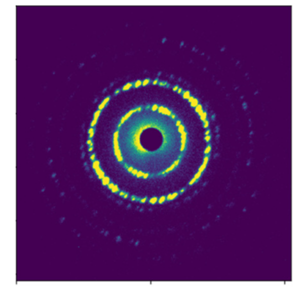
mfucb@slac.stanford.edu

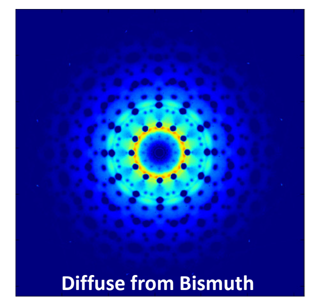
Good electron beam is necessary, but not sufficient for MeV-UED



Courtesy of Rob Parrish

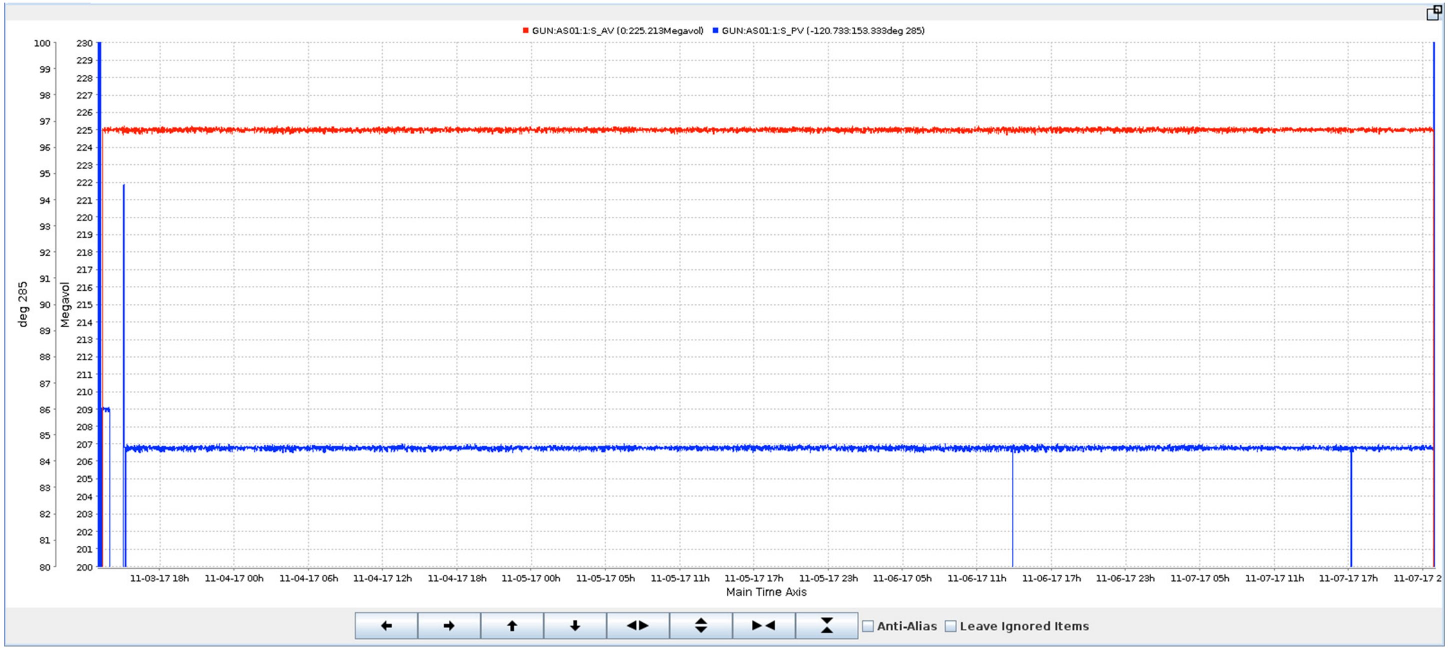




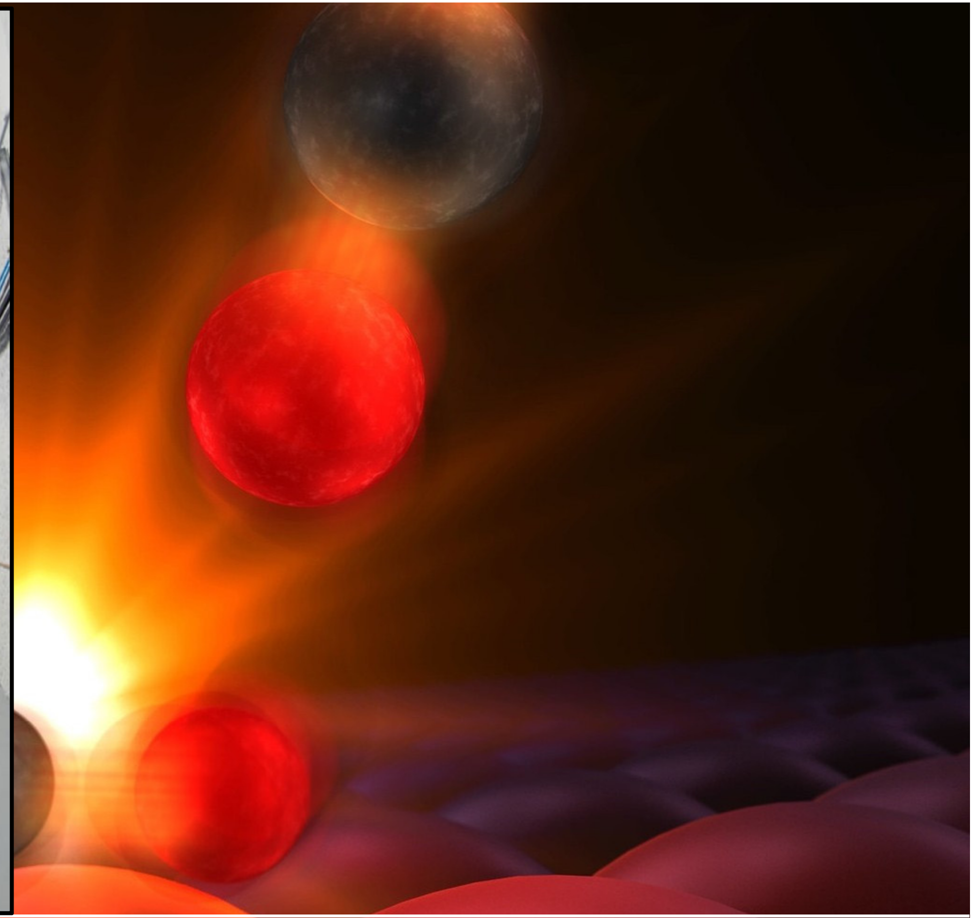


MeV Ultrafast Electron Diffraction (MeV-UED)

SLAC MeV-UED operation – first 4-day continuous operation



MeV-UED has transformed from laboratory toy into a **Research Tool!**



Ultrafast Electron Diffraction(UED)

Melting of Thin-Film Al (20 ps)

Mourou *et al.*, Appl. Phys. Lett. **41**, 44 (1982)

Williamson *et al.*, Phys. Rev. Lett. **52**, 2364 (1984)

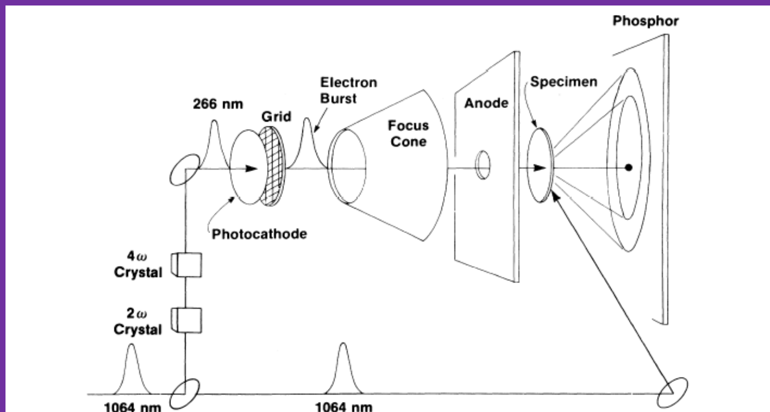
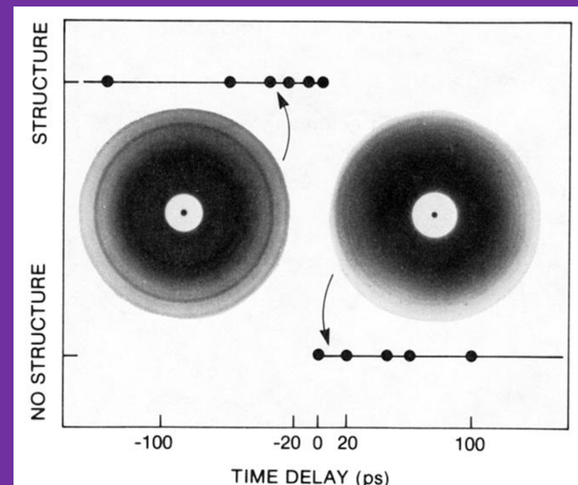


FIG. 1. Schematic of picosecond electron-diffraction apparatus. A streak-camera tube (deflection plates removed) is used to produce the electron pulse. The 25-keV electron pulse passes through the Al specimen and produces a diffraction pattern of the structure with a 20-ps exposure.



1. "Photocathode RF gun and Its Applications", the Workshop on the Marriage of High Intensity Lasers with Synchrotrons, February 21 – 22, 2000, University of Michigan, Ann Arbor, MI 48109, USA.

MeV Electron Beams for UED/UEM

PHYSICAL REVIEW E VOLUME 54, NUMBER 4 OCTOBER 1996

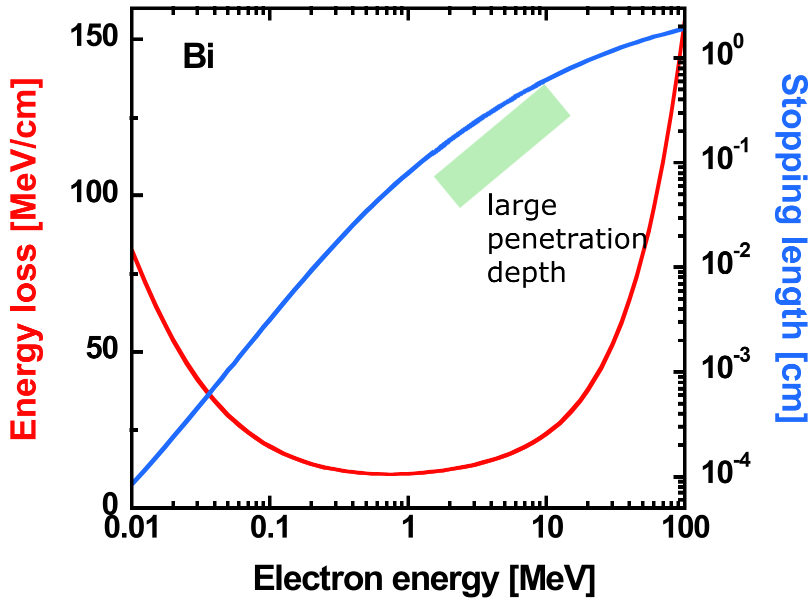
Experimental observation of high-brightness microbunching in a photocathode rf electron gun

X. J. Wang, X. Qiu, and I. Ben-Zvi
National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973
(Received 13 February 1996)

We report the measurement of very short, high-brightness bunches of electrons produced in a photocathode rf gun with no magnetic compression. The electron beam bunch length and the charge distribution along the bunch were measured by passing the energy chirped the electron beam through a momentum selection slit while varying the phase of the rf linac. The bunch compression as a function of rf gun phase and electric field at the cathode were investigated. The shortest measured bunch is 370 ± 100 fs (at 95% of the charge) with 2.5×10^8 electrons (170 A peak current); the normalized rms emittance of this beam was measured to be 0.5π mm mrad and the energy spread is 0.15%. [S1063-651X(96)51110-4]

[1] J. C. Williamson and A. H. Zewail, Proc. Natl. Acad. Sci. USA **88**, 5021 (1991).
[2] J. C. Williamson and G. Mourou, Phys. Rev. Lett. **52**, 2364 (1984).
[3] H. E. Elsayed-Ali and J. W. Herman, Appl. Phys. Lett. **57**, 1508 (1990).
[4] K. J. Kim, S. Chattopadhyay, and C. V. Shank, Nucl. Instrum. Methods Phys. Res. A **341**, 351 (1994).
[5] G. P. Gallerano *et al.*, Nucl. Instrum. Methods Phys. Res. A **358**, 74 (1995).
[6] P. Kung, H. Lihn, and H. Wiedemann, Phys. Rev. Lett. **73**, 967 (1994).
[7] B. E. Carlsten and S. J. Russell, Phys. Rev. E **53**, R2072 (1996).

Why diffraction with MeV electrons?



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

Space charge effect: $(1/(\beta^2\gamma^3)$

"thick" samples

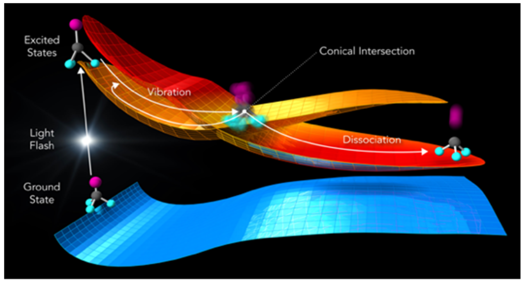
kinematic diffraction

@ 3.2 MeV, MFP ~ 20 nm
@ 55 KeV, MFP ~ 3 nm

"really" flat Ewald-sphere

MeV-UED made it possible (easier) to access broad ultrafast experiments and quantitively understand what is observed!

Science enabled by MeV-UED

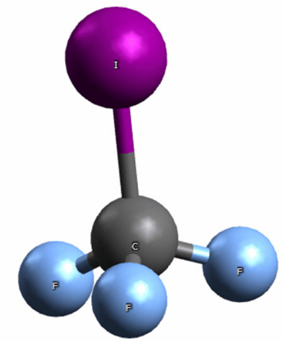


Bond-breaking & nuclear wavepacket passing through conical intersections (*Science 361 64–67 (2018)*)

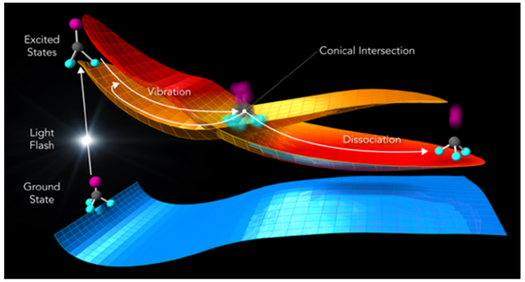
CHEMICAL PHYSICS

Imaging CF_3I conical intersection and photodissociation dynamics with ultrafast electron diffraction

Jie Yang[1,2*], Xiaolei Zhu[2,3], Thomas J. A. Wolf[2], Zheng Li[2,4,5], J. Pedro F. Nunes[6], Ryan Coffee[2,7,8], James P. Cryan[2], Markus Gühr[2,9], Kareem Hegazy[2,8], Tony F. Heinz[2,10], Keith Jobe[1], Renkai Li[1], Xiaozhe Shen[1], Theodore Veccione[1], Stephen Weathersby[1], Kyle J. Wilkin[11], Charles Yoneda[1], Qiang Zheng[1], Todd J. Martinez[2,3*], Martin Centurion[11*], Xijie Wang[1*]



Science enabled by MeV-UED



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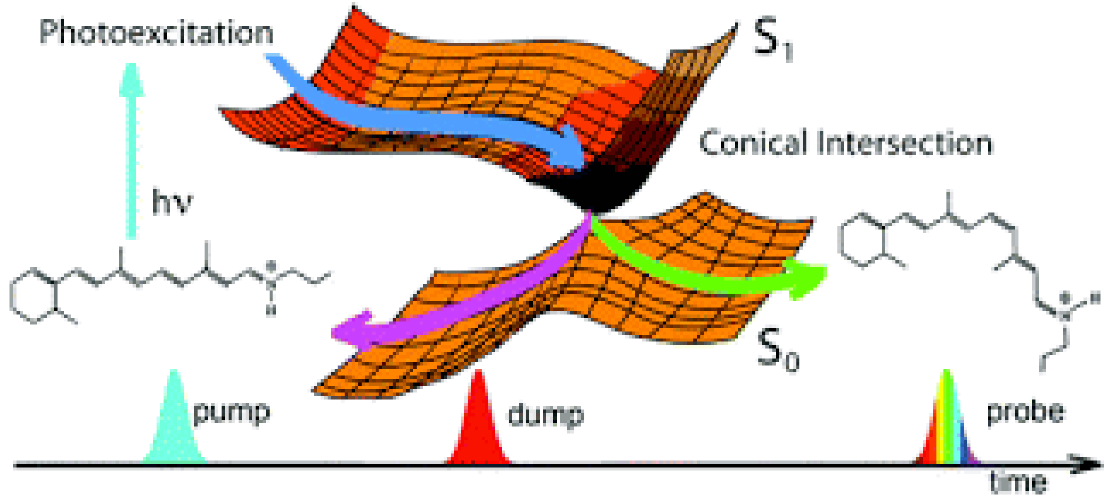

Acc. Chem. Res. **1995,** *28,* 119–132 **119**

Direct Observation of the Transition State

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Department of Chemistry, University of Toronto, Toronto, Ontario, Canada M5S 1A1

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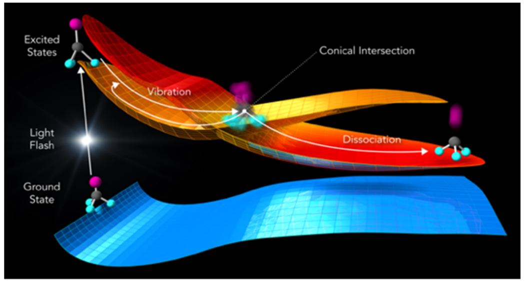
Received December 14, 1994

Science enabled by MeV-UED



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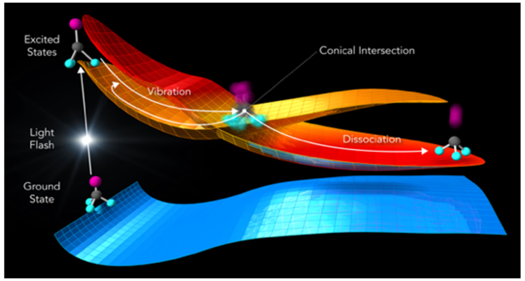
I. Introducing the Transition State

The direct observation of the "transition state" for chemical reaction clearly merits a place in this series devoted to the "Holy Grails" of chemistry. The transition state is neither one thing, namely, chemical reagents, nor the other, reaction products. Instead it illustrates the mystical event of trans-substantiation.

The time required for the electronic process of bond rupture and bond formation would be unmeasurably short were it not for the fact that the electronic transformations are triggered by substantial changes in internuclear separation. This became evident in the early years of quantum mechanics when it was possible, for the first time, to calculate the binding energy between an atomic pair, BC, as a function of internuclear separation.[1]

Science enabled by MeV-UED



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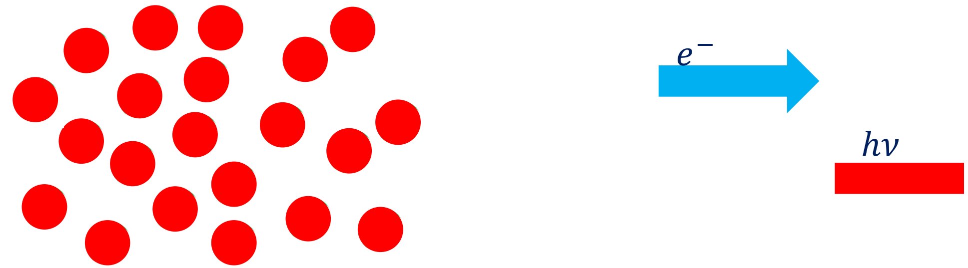
60keV UED

$V_e = 0.446c$

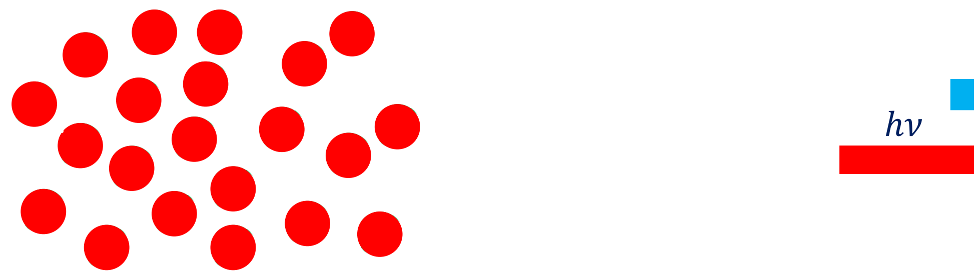
$\Delta t_{VM} > 1ps$



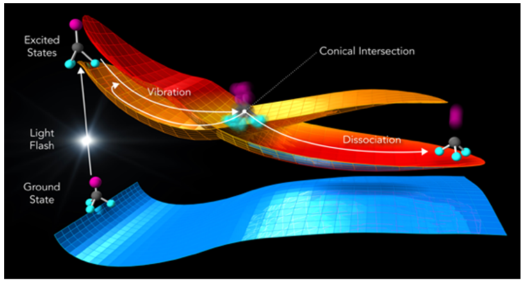
3MeV UED

$V_e = 0.989c$

$\Delta t_{VM} < 10fs$



Science enabled by MeV-UED



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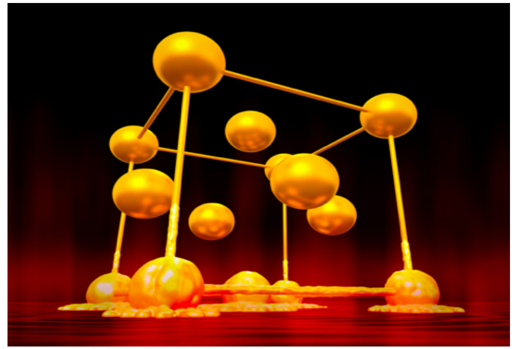


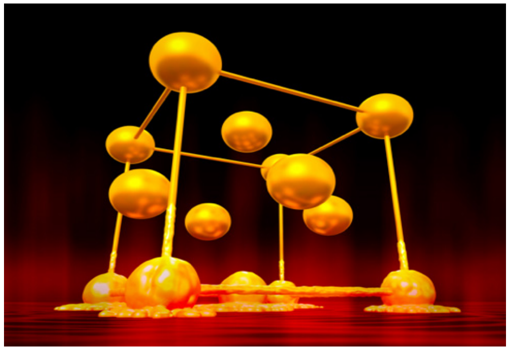
Resolving ultrafast phase transitions *(Science 360 1451–1455 (2018))*

CONDENSED MATTER

Heterogeneous to homogeneous melting transition visualized with ultrafast electron diffraction

M. Z. Mo[1*†], Z. Chen[1†], R. K. Li[1], M. Dunning[1], B. B. L. Witte[1,2], J. K. Baldwin[3], L. B. Fletcher[1], J. B. Kim[1], A. Ng[4], R. Redmer[2], A. H. Reid[1], P. Shekhar[5], X. Z. Shen[1], M. Shen[5], K. Sokolowski-Tinten[6], Y. Y. Tsui[5], Y. Q. Wang[3], Q. Zheng[1], X. J. Wang[1], S. H. Glenzer[1*]

Science enabled by MeV-UED



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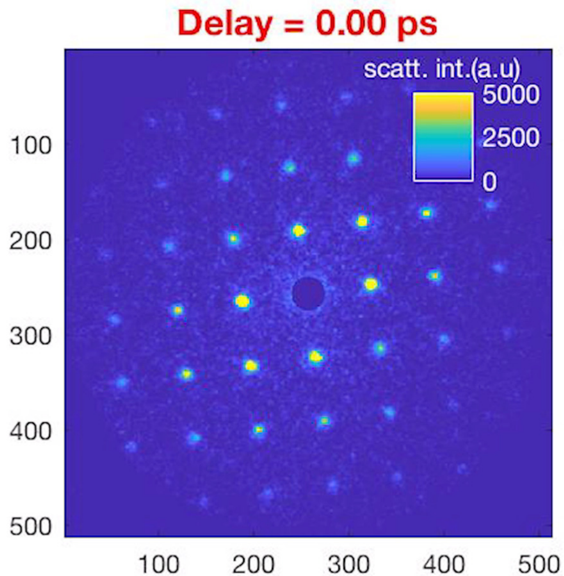
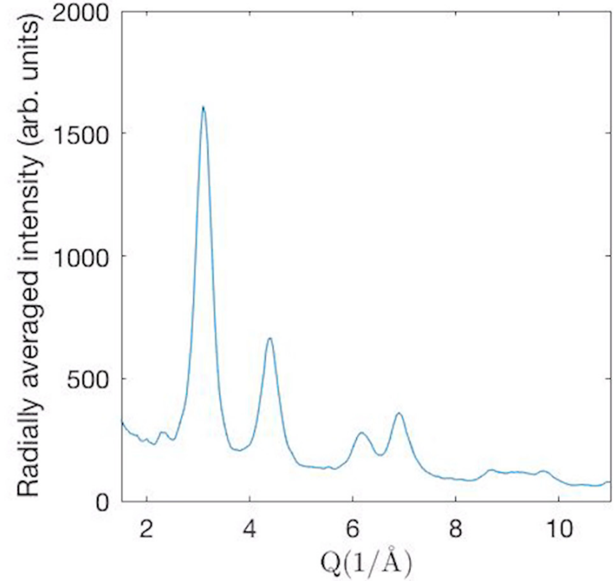
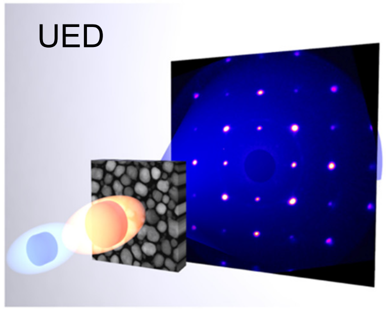
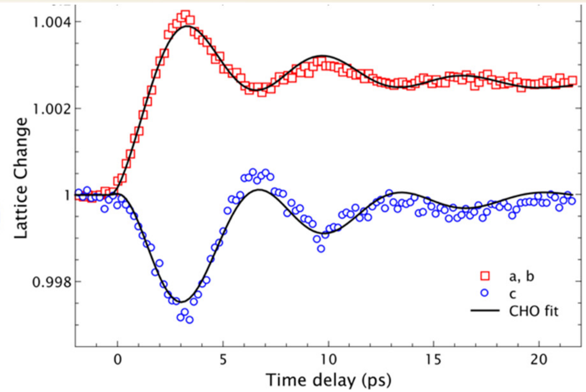


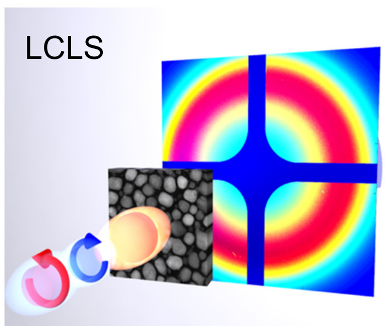


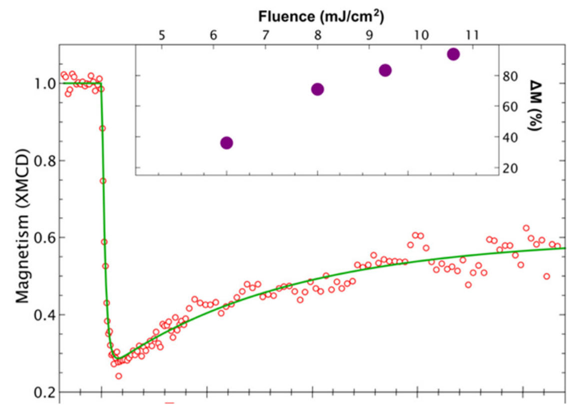
3-D Structure Dynamics& Multi-modal probe (UED/LCLS)

Beyond a phenomenological description of Magnetostriction









A. H. Reid et al, Nature Communications **9,** 388, DOI: 10.1038/s41467-017-02730-7 (2018).

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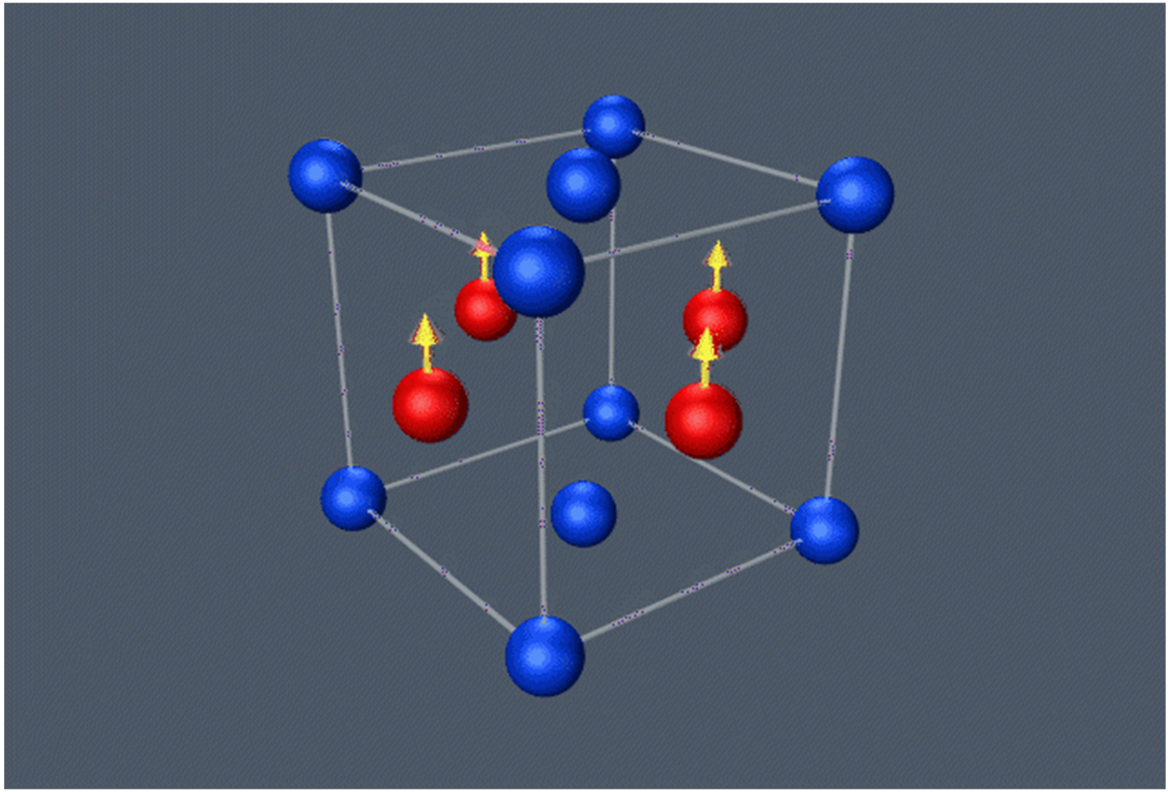
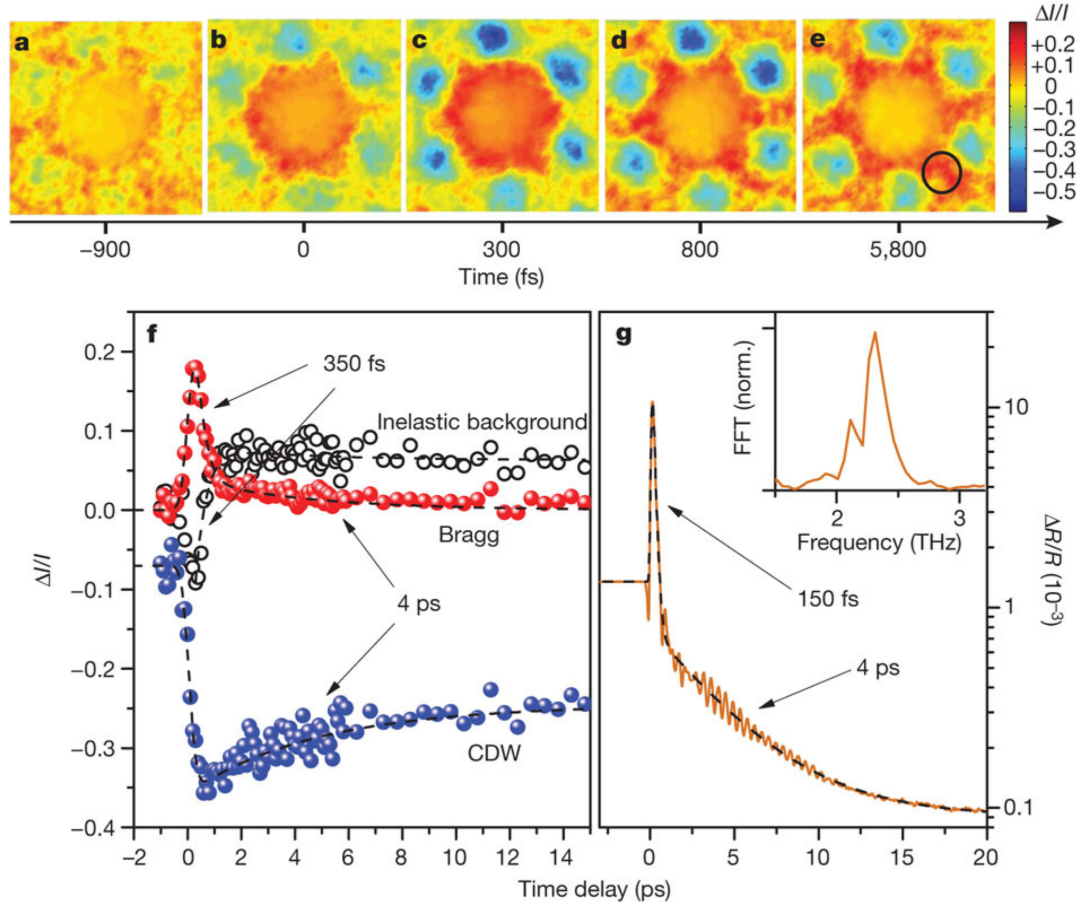
SLAC



A. H. Reid et al, Nature Communications **9,** 388, DOI: 10.1038/s41467-017-02730-7 (2018).

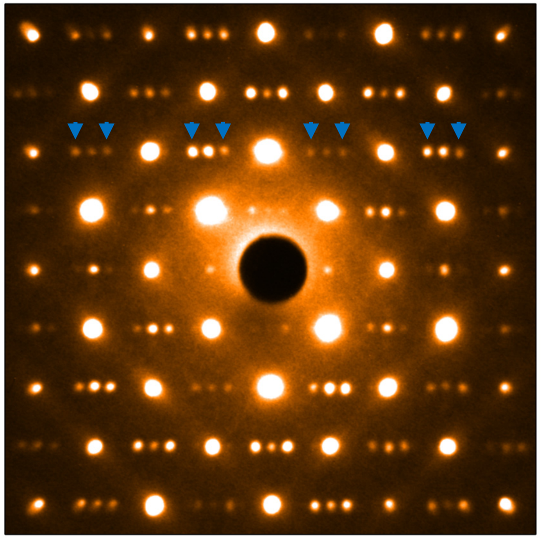
Snapshots of cooperative atomic motions in the optical suppression of charge



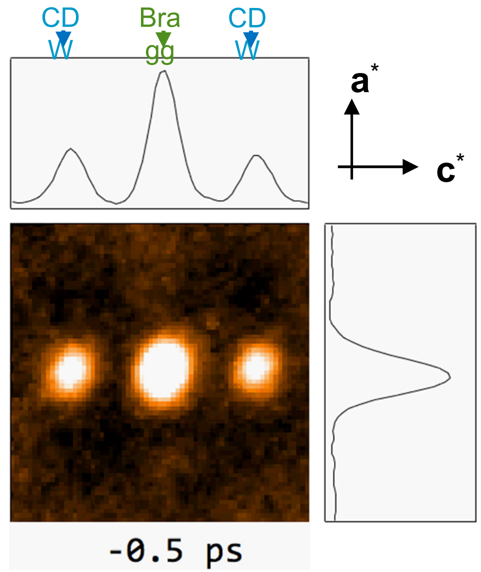
M Eichberger *et al. Nature* **000**, 1-4 (2010) doi:10.1038/nature09539

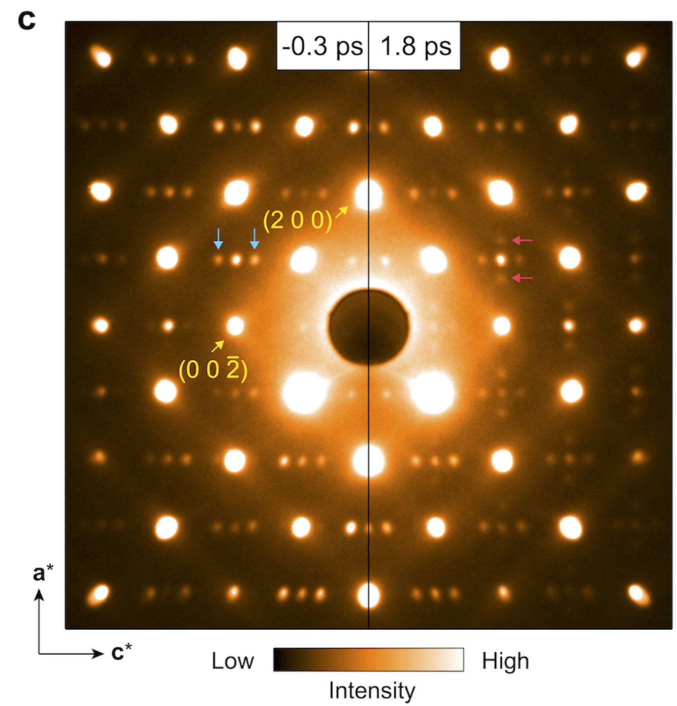
nature

Light induced CDW in $LaTe_3$



3.1 MeV electron kinetic energy





Anshul Kogar, Alfred Zong *et al*, under review, arXiv:1904.07472 [cond-mat.mtrl-sci]

Academically – non-adiabatic dynamics are a grand challenge

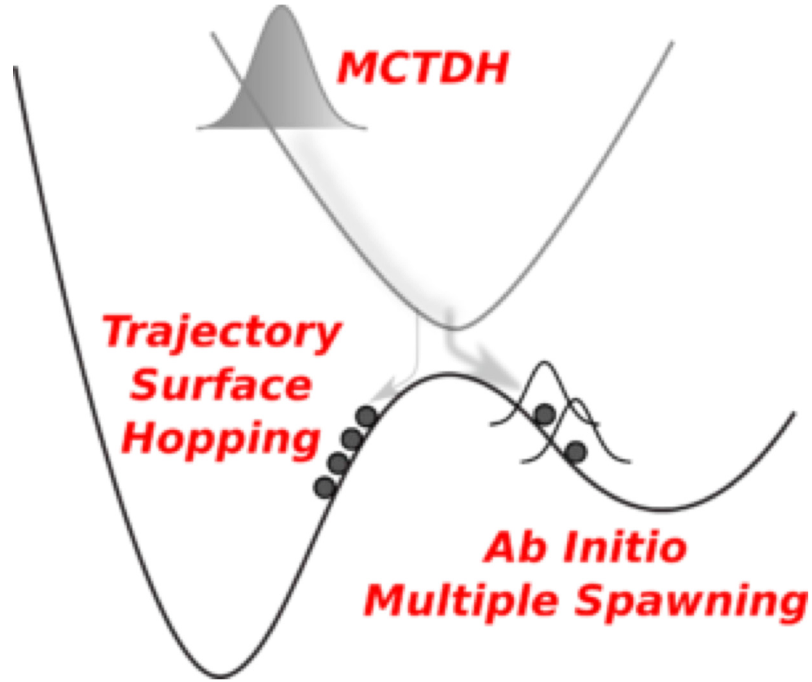
Potential energy surface is the central concept for understanding chemical reactions.

Adiabatic (Born-Oppenheimer) PES a 3N hypersurface is a function of all nuclear positions – mapped w/ high accuracy for larger & larger systems.

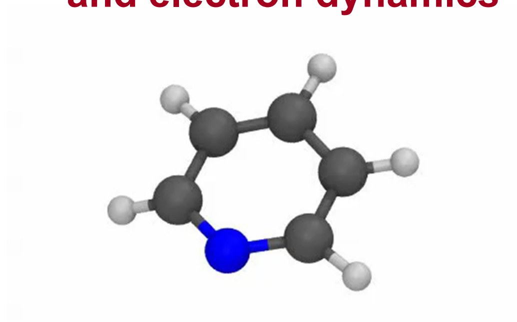
BUT – many events cannot be described w/ BO

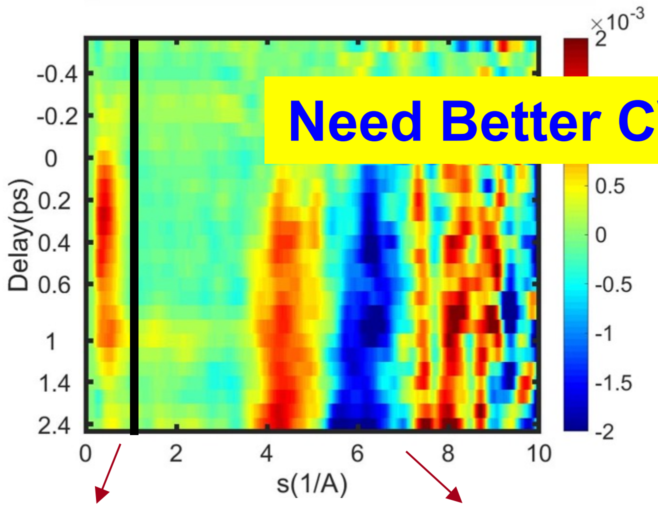
Non-adiabatic electronic transitions between PESs are ubiquitous and important …

- conical intersections
- intersystem crossing
- internal conversion
- electron transfer ….



Pyridine—Simultaneous nuclear and electron dynamics

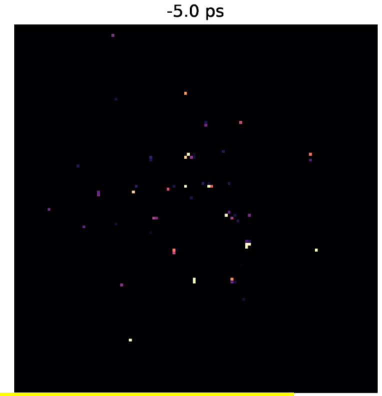




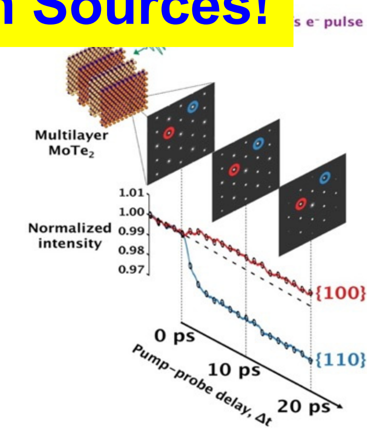
Electronic dynamics **Nuclear dynamics**

Jie Yang et al, to be summitted.

Optical control of non-equilibrium phonon dynamics



Need Better CW Electron Sources!



Krishnamoorthy et al, under review

The Nobel Prize in Chemistry 2017

© Nobel Media. Ill. N. Elmehed

Jacques Dubochet

Prize share: 1/3

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Joachim Frank

Prize share: 1/3

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Richard Henderson

Prize share: 1/3

The Nobel Prize in Chemistry 2017 was awarded to Jacques Dubochet, Joachim Frank and Richard Henderson *"for developing cryo-electron microscopy for the high-resolution structure determination of biomolecules in solution"*.

The Nobel Prize in Chemistry 2017



© Nobel Media. Elmehed

Jacques Dub

Prize share: 1/3

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The Nobel P

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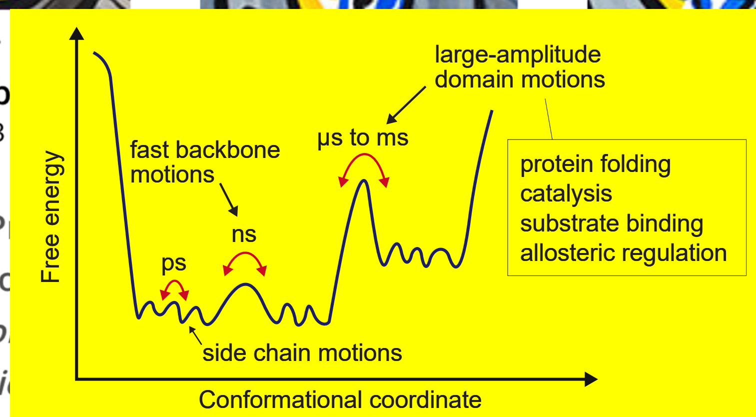


Sample Damage

Ultramicroscopy 2 (1977) 199–203

ADVANTAGES OF MEGAVOLT ELECTRON MICROSCOPY IN BIOLOGICAL RESEARCH

Gaston DUPOUY
Laboratoire d'Optique Electronique du Centre National de la Recherche Scientifique, Toulouse, France

Received 21 December 1976

New electron microscopes, operating within the megavolt range have opened important prospects for scientific research. These microscopes were, at first, mainly used by physicists and metallurgists; but nowadays more and more biologists are interested in high voltage electron microscopy: they have obtained important and significant results.

The present paper gives some information concerning experiments that have been achieved in Toulouse with our two big instruments working at 1 million volts (1 MV) and three million volts (3 MV).

Sample Damage

by the electron beam. This problem is particularly interesting when the specimen is an organic or biological material. The first experiments in this matter were carried out by Kobayashi and Sakaoku [12] on organic polymers. The important result of their experiments is the decreasing of radiation damage when the electron energy is increased. Similar experiments have been done by Thomas [15] and Glaeser and Thomas [11] with valine and glycine.

In collaboration with F. Perrier we extended these experiments up to 3 MV; they were carried out with Thymine, which is one of the components of D.N.A. We operated at four voltages: 0.5 MV, 1 MV, 2 MV and 3 MV.

The results of these experiments are following: radiation damage is about four times less at 3 MV than at 100 kV.

Single-shot MeV Ultrafast Electron Microscope (UEM)

To capture processes in materials science and biology, such as direct imaging of biologically important conformational transitions of macromolecules and glass phase transitions in real time, not possible with existing transmission electron microscopes (TEMs)

- **Atomic spatial resolution (0.3 nm)**
- **Sub-nanosecond temporal resolution (~ 100 ps)**

SLAC proposed single-shot UEM will be based on the 200 MHz superconducting RF gun transferred to SLAC. SLAC single-shot MeV UEM will also addresses the fundamental limit of the current cryo-EM:

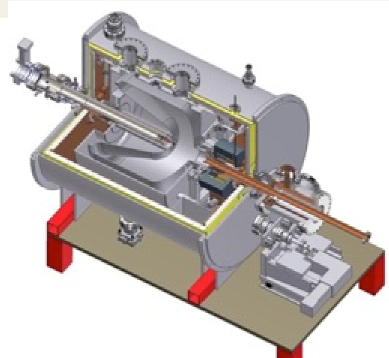
- Beam induced sample movements (~µs).
- Fast backbone protein movements (~ ns).
- Sample in its native environments.
- Large (thicker) samples.

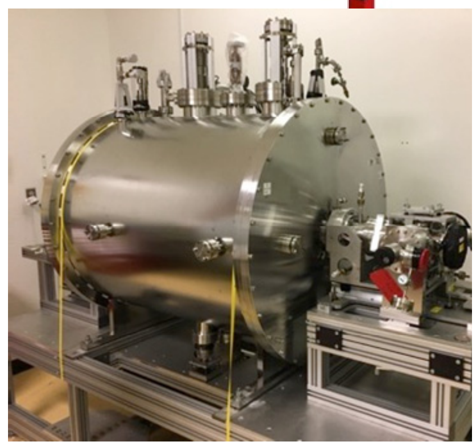
VHF SRF gun for MHz UED & single-shot UEM

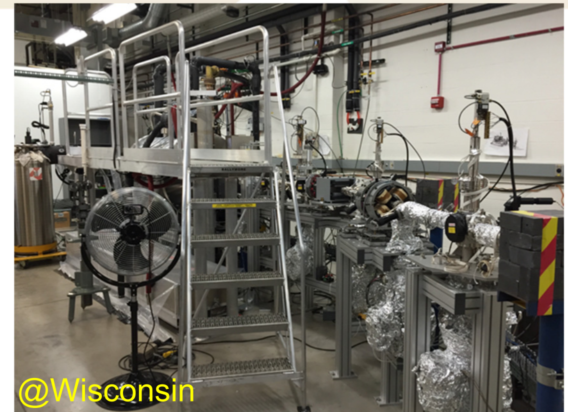
SLAC

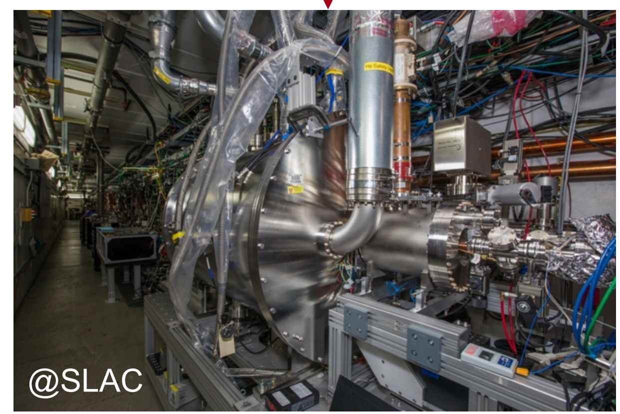
- **Stability**: toward 10^{-5} amplitude and 10 fs phase stability
- **High gradient**: >25 MV/m, high peak beam brightness
- **High beam energy**: 4 MeV, SC suppression, 10 fs t-resolution
- **High rep-rate**: up to 200 MHz for **nano-UED**
- **Flexibility**: deliver up to 100 ps bunch length for **UEM**

VHF SRF gun synergy with LCLSII-HE !







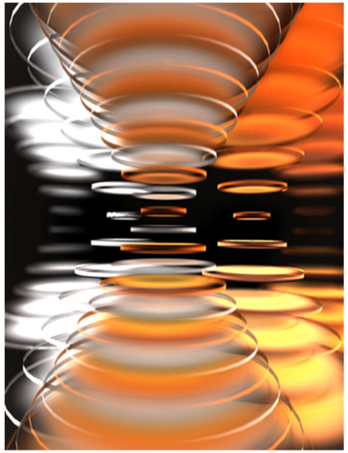


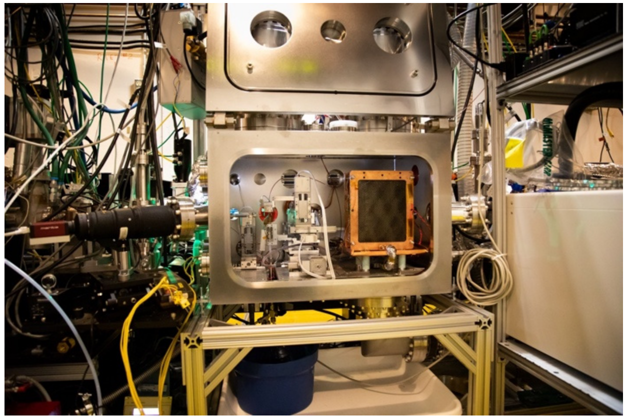
Summary

- ✓ SLAC MeV-UED has demonstrated high temporal and spatial resolutions required for ultrafast science. It is capable of operating from single-shot to 360 Hz.

- ✓ Large scattering cross section & q-range, negligible multiple scattering and versatile sample environment, enabled ultrafast science in material science, chemical dynamics and warm dense matter physics.

- ✓ Future MeV-UED require more stable and better electron sources.





Thank You!

Acknowledgement

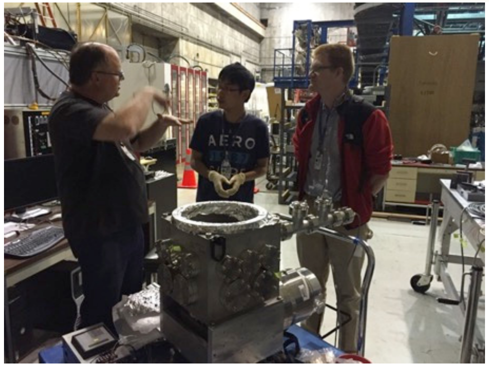
SLAC

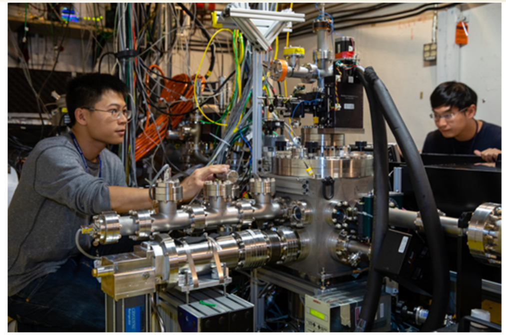
- **SLAC UED program is supported in part by the U.S. Department of Energy (DOE) Office of Basic Energy Sciences Scientific User Facilities Division – Accelerator & Detector R&D Program .**

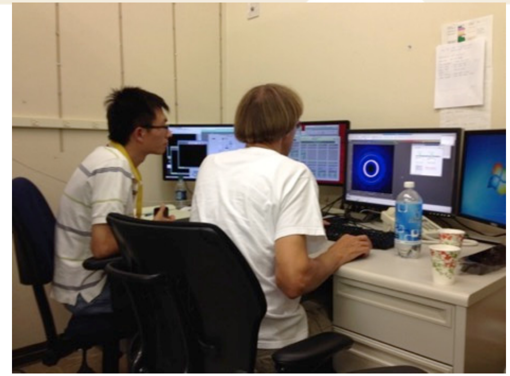
- **SLAC UED/UEM Initiative Program Development Funds (SLAC program development and AD program development), and Laboratory Directed Research & Development (LDRD) fund.**

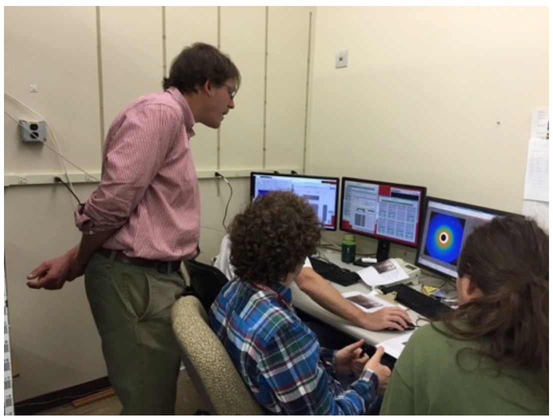
- **The technical support provided by SLAC Accelerator Directorate, Technology Innovation Directorate, and LCLS is grateful acknowledged.**

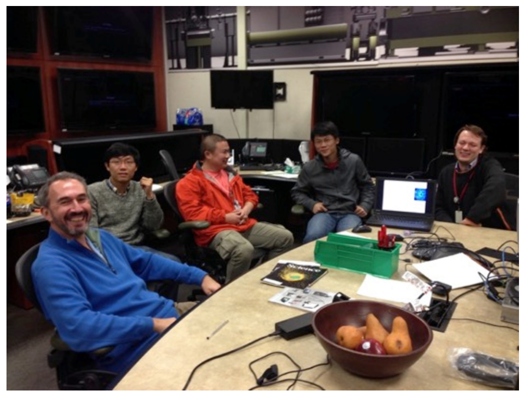
SLAC MeV-UED collaborators

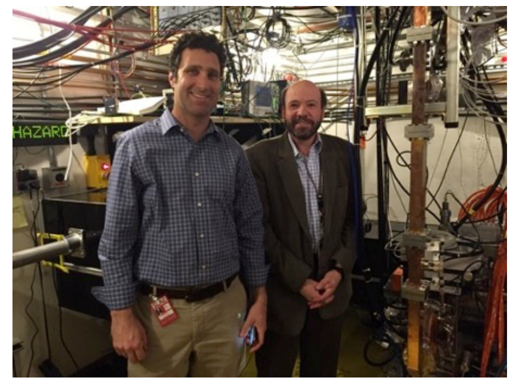


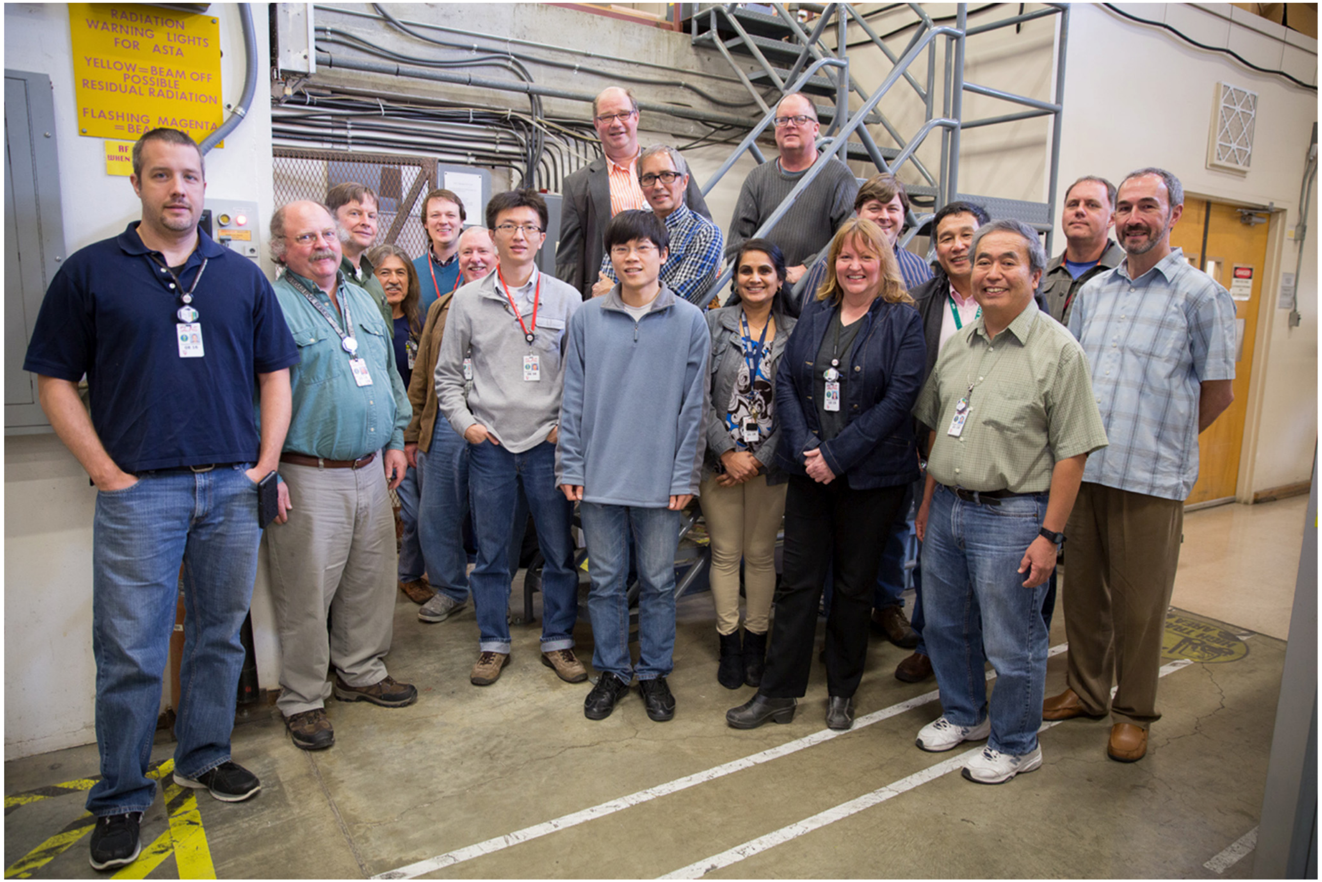












SLAC's UED/UEM team. From left to right: Theodore Vecchione, Eric Bong, Jeff Corbett, Bobby McKee, Alexander Hume Reid, Perry Anthony, Renkai Li, Carsten Hast, Xiaozhe Shen, Stephen Weathersby, Shanta Condamoor, Keith Jobe, Margery Morse, Garth Brown, Xijie Wang, Charles Yoneda, James Lewandowski, Hermann Dürr., **Not shown: Ryan Coffee, Juan Cruz, Juan, John Eichner, Nick Hartmann , Josef Frisch, Bo Hong, Erik Jongewaard, Justin May, Doug McCormick, Minh Nguyen, Dentell Reed, Daniel Van Winkle & Juhao Wu**