

Making Molecular Movie with MeV Electrons

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on behalf of SLAC UED team

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Outline

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- Introduction: SLAC UED/UEM Initiative
 - Why MeV electrons
- Status and performance of the SLAC MeV UED system
 - Key components: rf-laser timing system, stable high-power rf source, etc
 - Temporal resolution
- Highlights of recent science results
 - Materials science experiments: MoS₂, FePt, diffuse scattering...
 - Gas-phase experiment: N₂ alignment, I₂ vibrational dynamics
- Outlook and future development

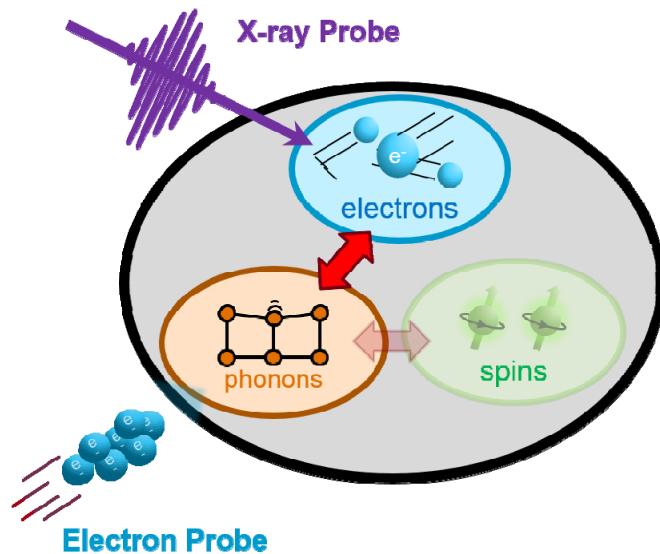
Visualizing the ‘ultrasmall’ and ‘ultrafast’

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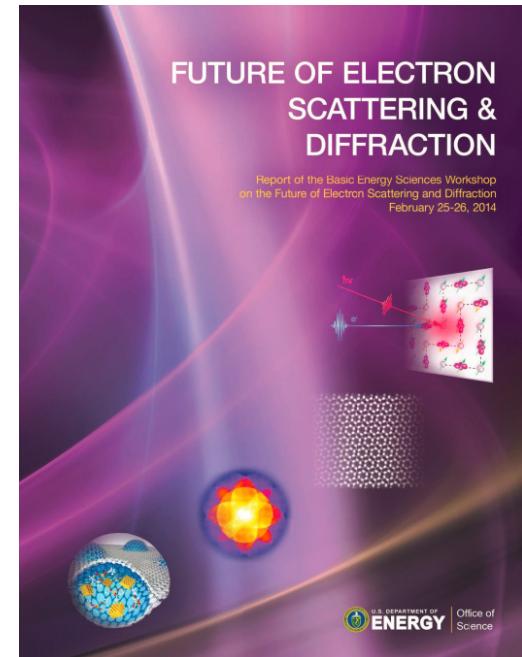
- *Ultrasmall – atomic length scale, Å*
- *Ultrafast – atomic time scale, 100 fs*

One of the Critical Instruments –
**Ultrafast Electron Diffraction
and Microscopy (UED/UEM)**

Y. Zhu, Report to BESAC, July 2014



X-ray (XFEL) and electron (UED/UEM) probes are *complementary tools* towards a *complete picture* of the fundamental processes.



Future of Electron Scattering
and Diffraction Workshop
February 25-26, 2014

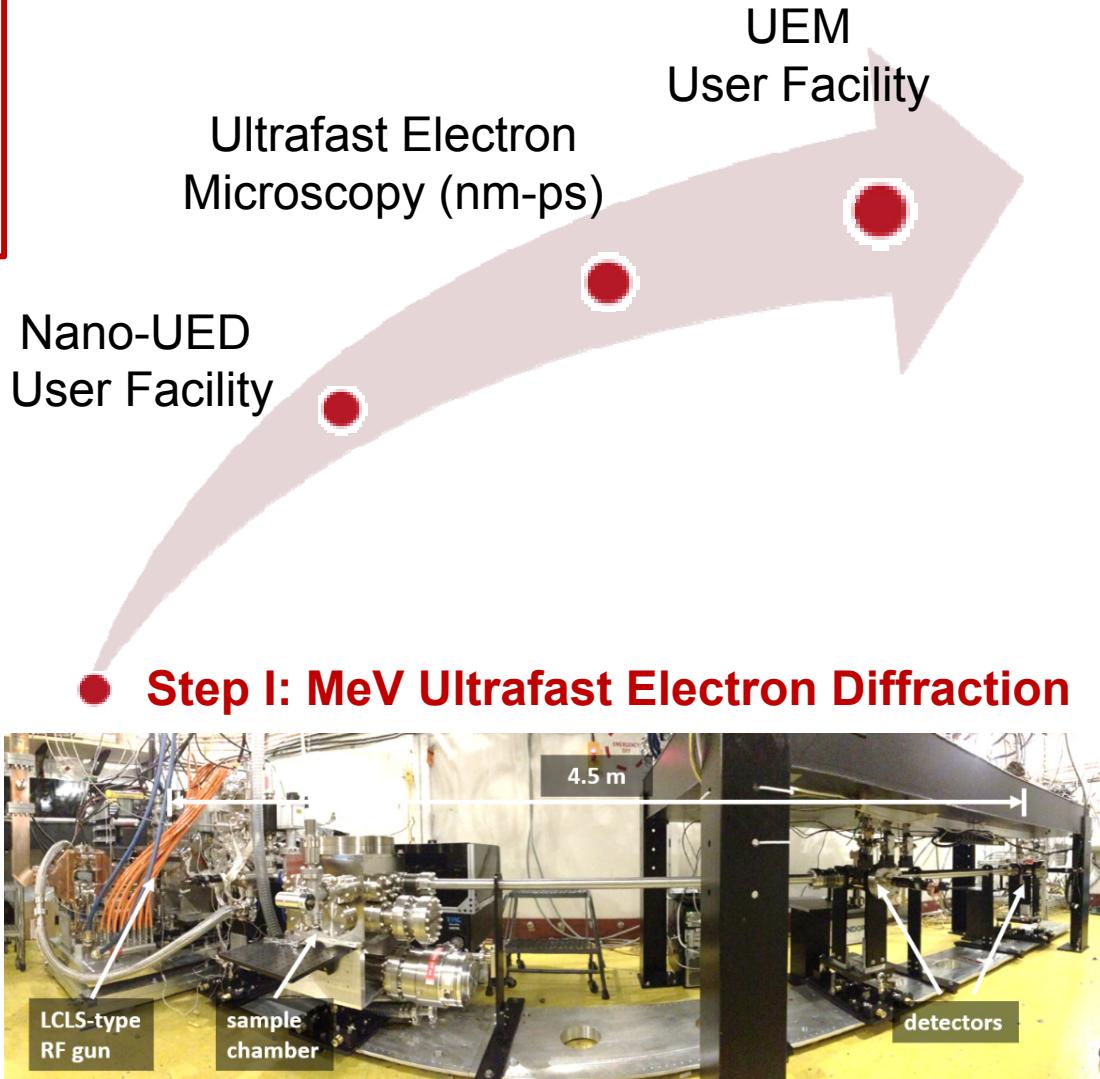
SLAC's vision for UED and UEM

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SLAC UED/UEM Initiative:

“... to provide the world’s leading ultrafast electron scattering instrumentation.”

- Complementarity with LCLS
- Ultrafast science community
- Expertise in electron beam physics and technology
- Pumping capabilities: ultrafast laser, extremely intense THz from FACET, and X-ray from LCLS



Why MeV electrons?

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1. MeV electrons effectively suppresses space charge effect $\propto \frac{1}{\gamma^3}$ -> more electrons packed into a shorter bunch Δt_e
2. In gas sample, eliminate velocity mismatch Δt_{vm} between pump laser and probe electrons

60keV UED

$$V_e = 0.446c$$



3MeV UED

$$V_e = 0.989c$$



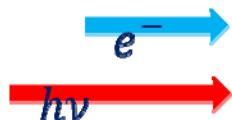
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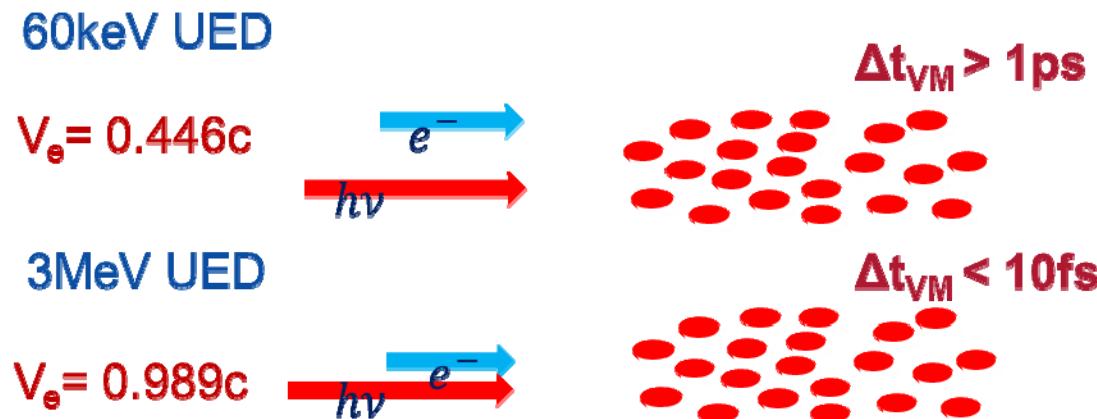
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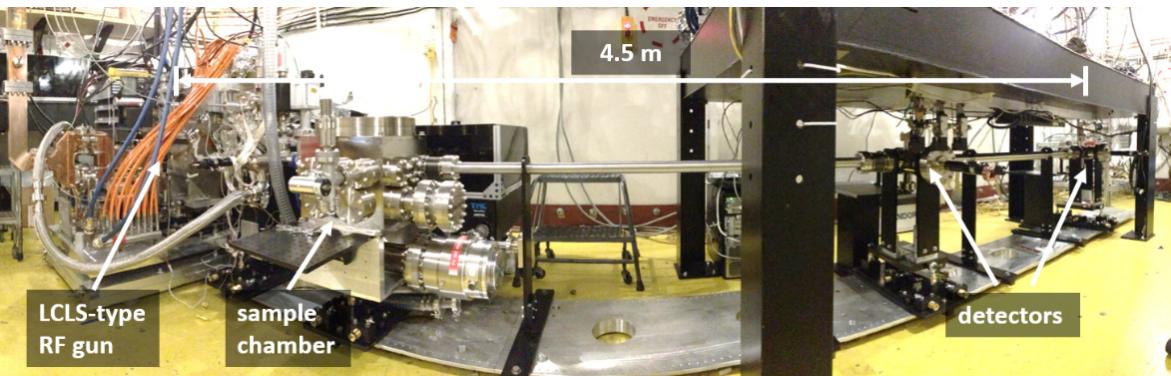
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Performance of the SLAC MeV UED system

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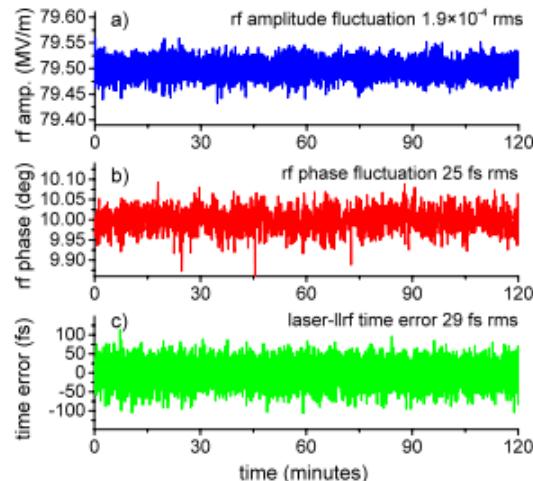


- LCLS-type S-band RF gun
 - Ultra-stable HV modulator
 - 5 mJ, 40 fs Ti:Sapphire laser system
 - fs laser-LLRF synchronization system
 - Sample stage with 5-axis motion and cryo-module to 34K
 - High efficiency electron detector

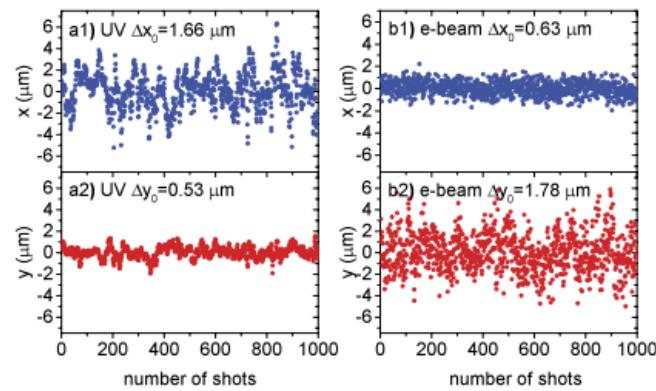
Typical operation parameters

Parameters	Values
rep. rate	180 Hz
beam energy	4 MeV
bunch charge	50 fC
emittance	<20 nm-rad
bunch length	<100 fs

ultra-stable high power rf system and rf-laser timing



μm-level e-beam pointing



Temporal resolution

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

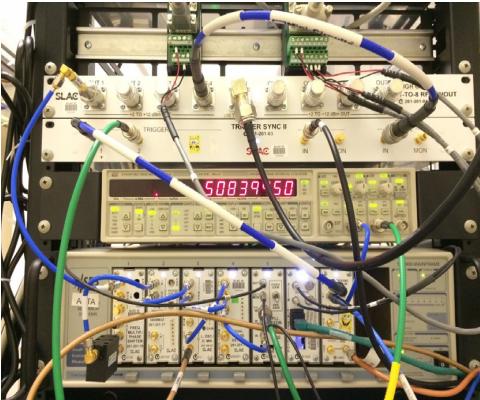
temporal laser pulse e- pulse velocity
resolution length length mismatch TOA jitter

Time-of-arrival jitter, due to the amp. and phase jitter of the rf field, used to be a major challenge for MeV UED

Two major upgrades: TOA jitter < 50 fs rms

SLAC laser-IIrf timing system

ultra-stable modulator and high power rf source



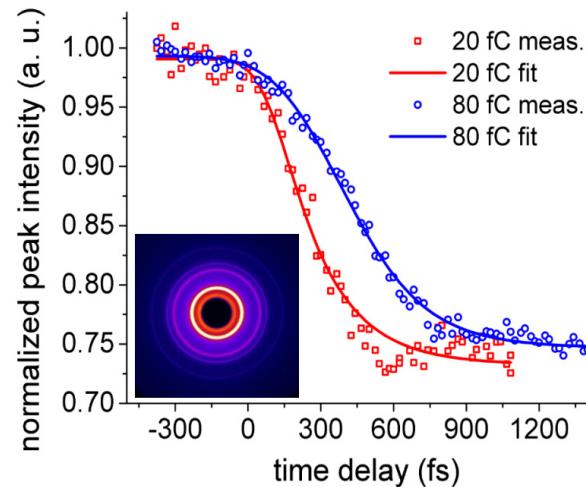
laser-IIrf timing jitter <30 fs rms



phase jitter <30 fs rms,
amplitude jitter <2×10⁻⁴

temporal resolution ~100 fs rms

based on the fast decay of (410) ring of 25 nm Bi films after laser excitation, assuming the intrinsic decay time is 150 fs rms.

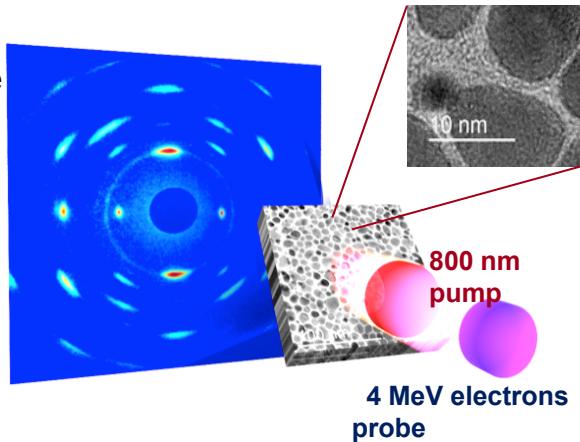


Material Science MeV UED Experiments

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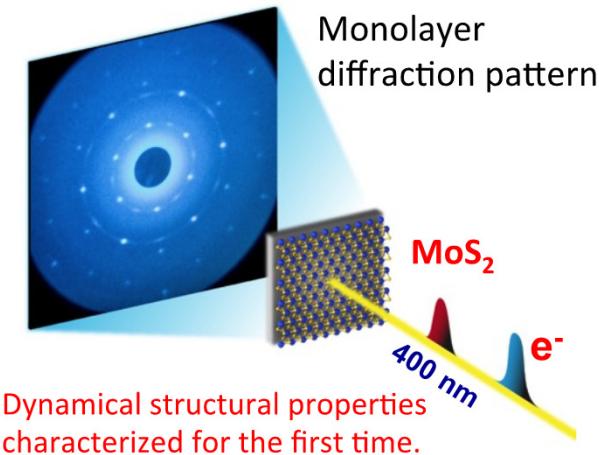
Lattice dynamics of FePt nanoparticles

- Large q-range
- Access to multiple Bragg reflections
- Solve in-plane and out-of-plane lattice dynamics with tilted sample geometry



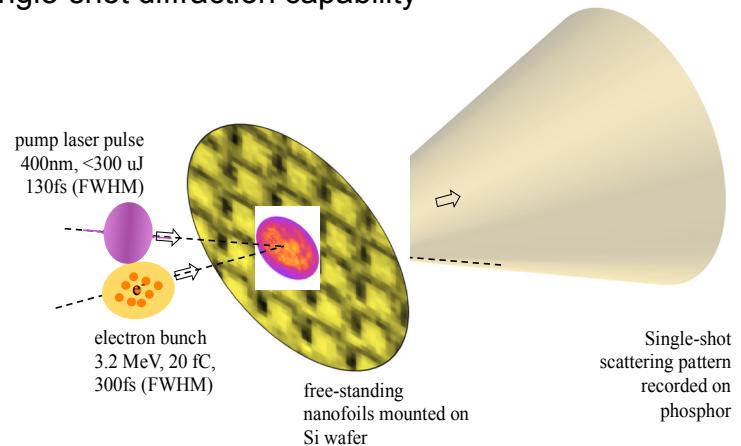
Lattice dynamics of monolayer MoS₂

- High SNR diffraction pattern from monolayer materials



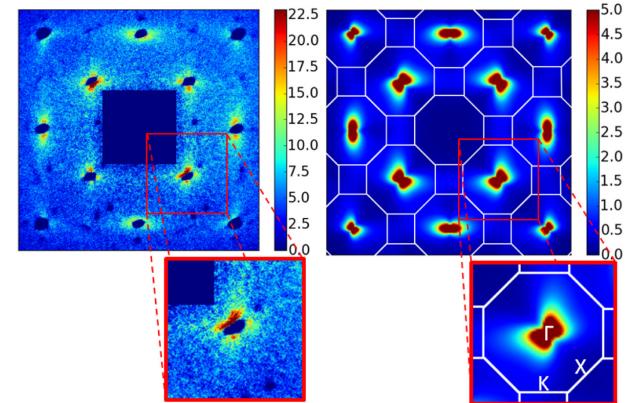
Ultrafast melting of single crystal Au

- Single-shot diffraction capability



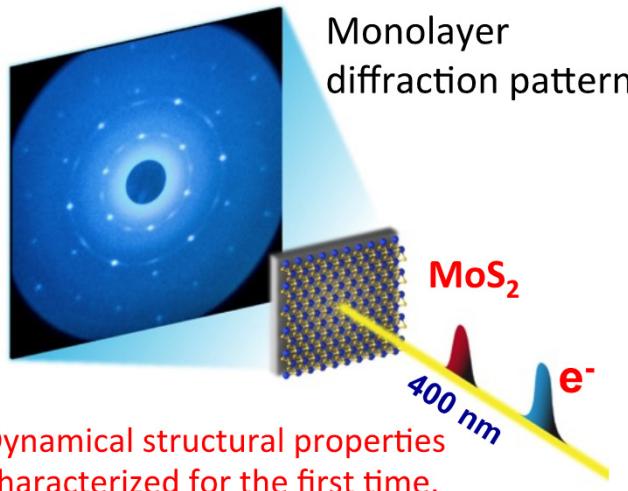
Single crystal Au diffuse scattering

- Reduced impact of multiple and inelastic scattering



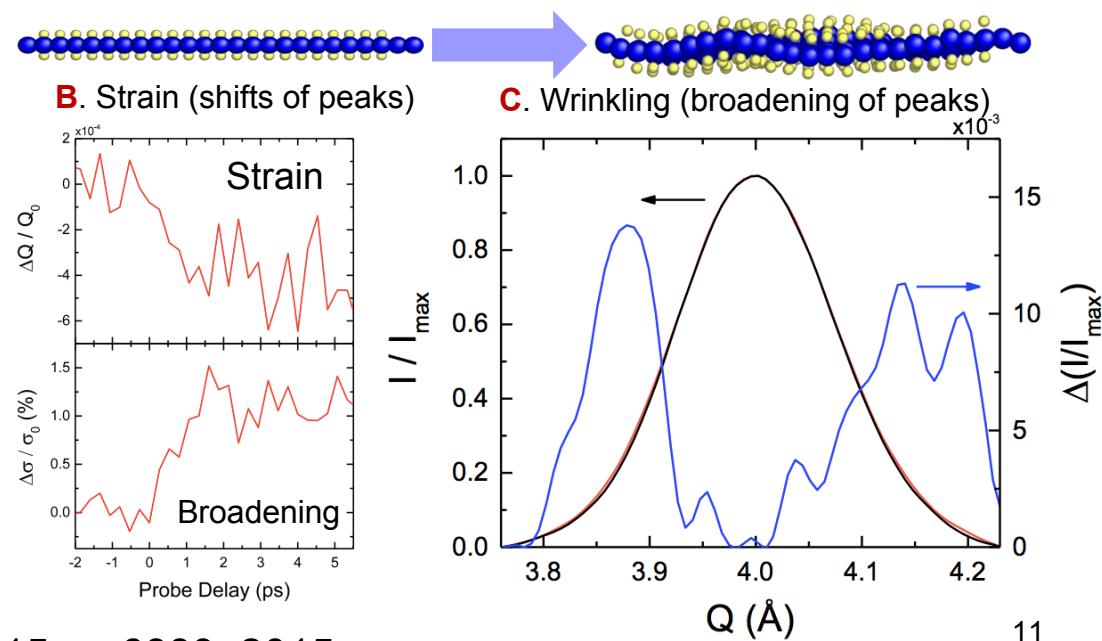
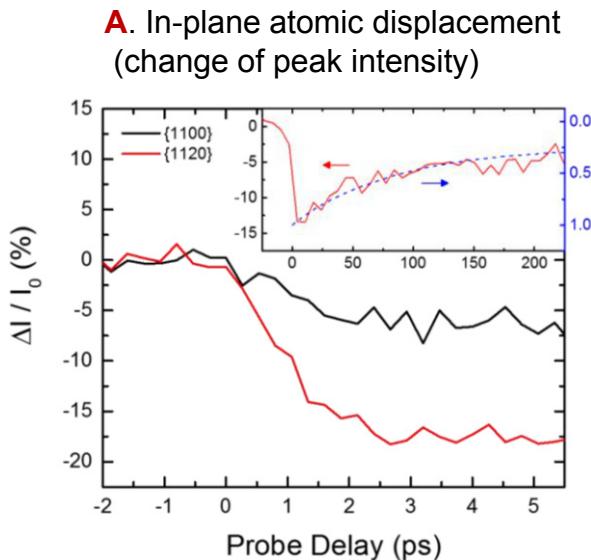
Dynamic structural deformations of monolayer MoS₂

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Dynamical structural properties characterized for the first time.

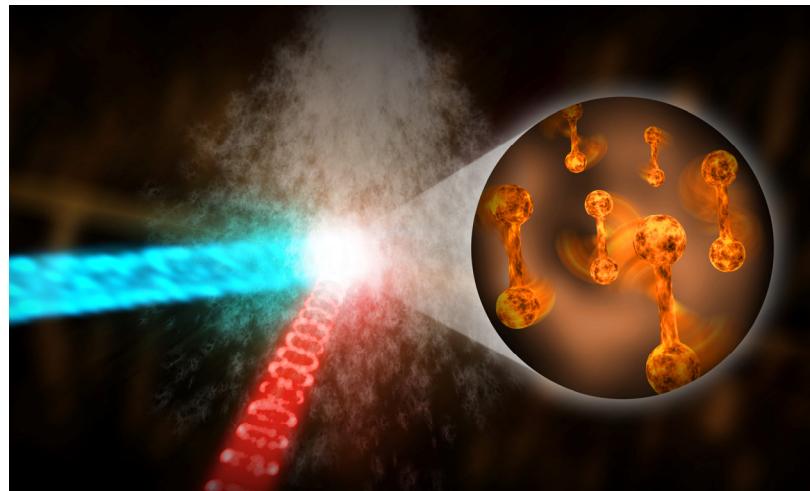
- Femtosecond electron diffraction measurements on **monolayer MoS₂**
- Optical excitation induces large-amplitude (**A**) **in-plane atomic displacement** as well as ultrafast (**B**) **strain** and (**C**) **wrinkling** of the monolayer on nanometer length-scales and picosecond time-scales
- New possibilities of all-optical dynamic control of electronic and optical responses of such materials



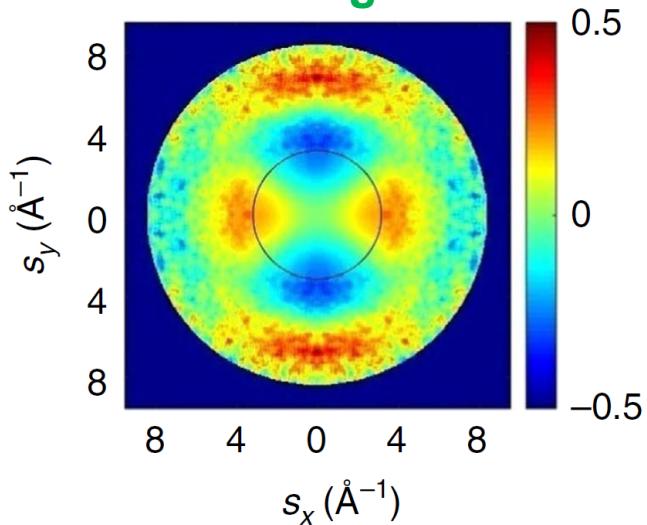
Gas-phase MeV UED

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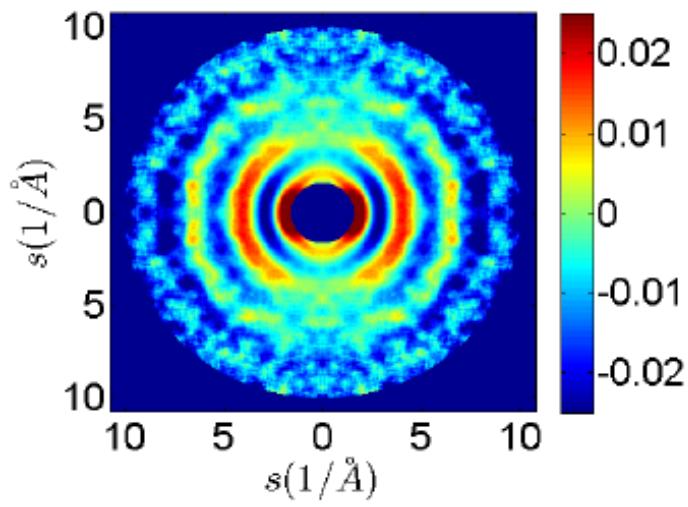
- **Advantage:**
 - strong interaction
 - much larger q-range
 - negligible velocity mismatch
 - 100 fs temporal resolution
- **Challenges:**
 - long exposure->high machine stability
 - manage vacuum level: 10^{-10} Torr at the gun and 10^{-4} Torr in the sample chamber



Rotational dynamics
of laser-aligned N₂

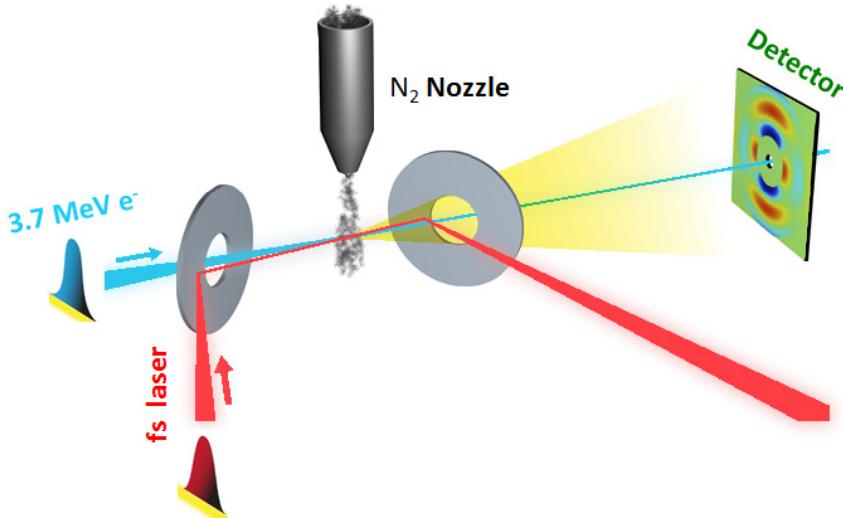


Vibrational dynamics of I₂



Ultrafast rotational dynamics of laser-aligned N₂

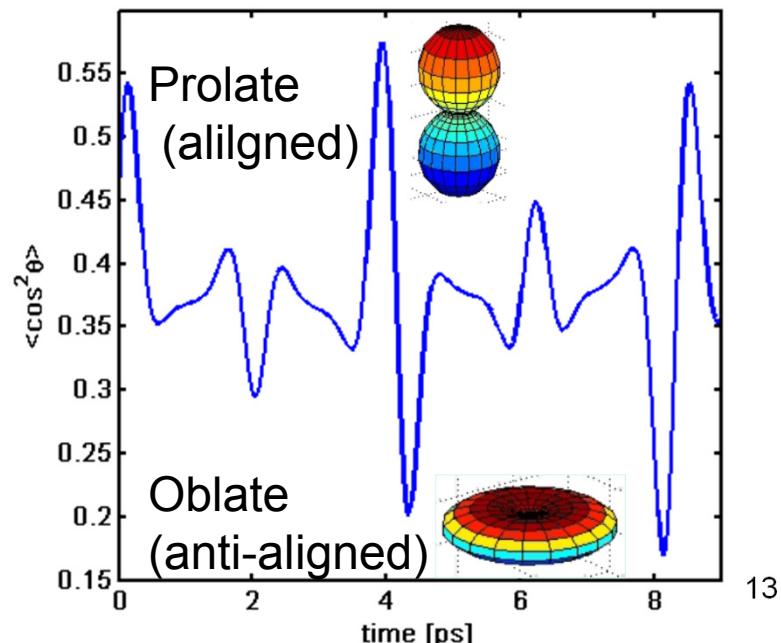
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- N₂ jet from pulsed nozzle
- Rotational alignment by intense ($\sim 10^{13}$ W/cm²) laser pulse
- Ultrafast rotational dynamics probed by MeV electrons

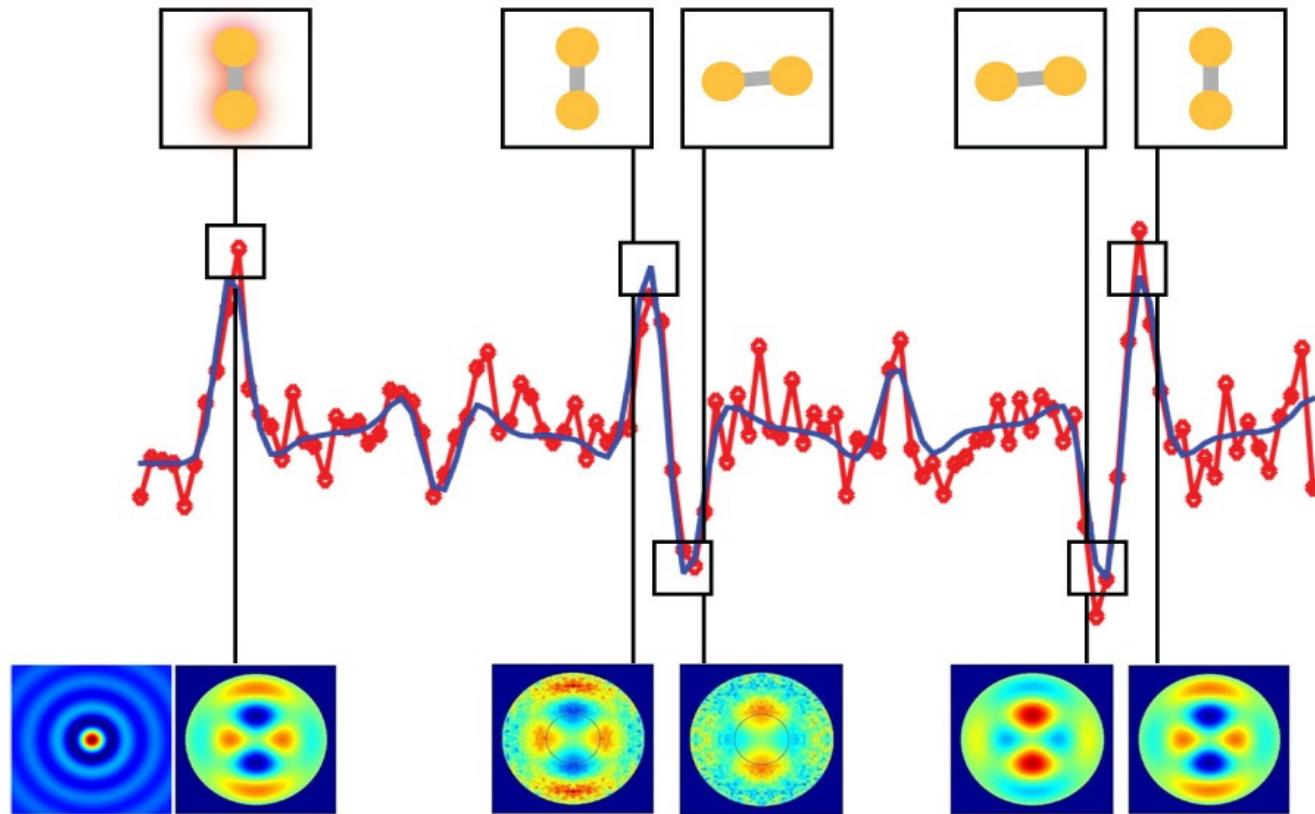
Physical picture

- Laser induces dipole along molecular axis
- Dipole aligns with electric field
- Quantum mechanics calculation shows periodic molecule distribution revivals after 4 and 8 ps



Direct measurement of N₂ rotational revival

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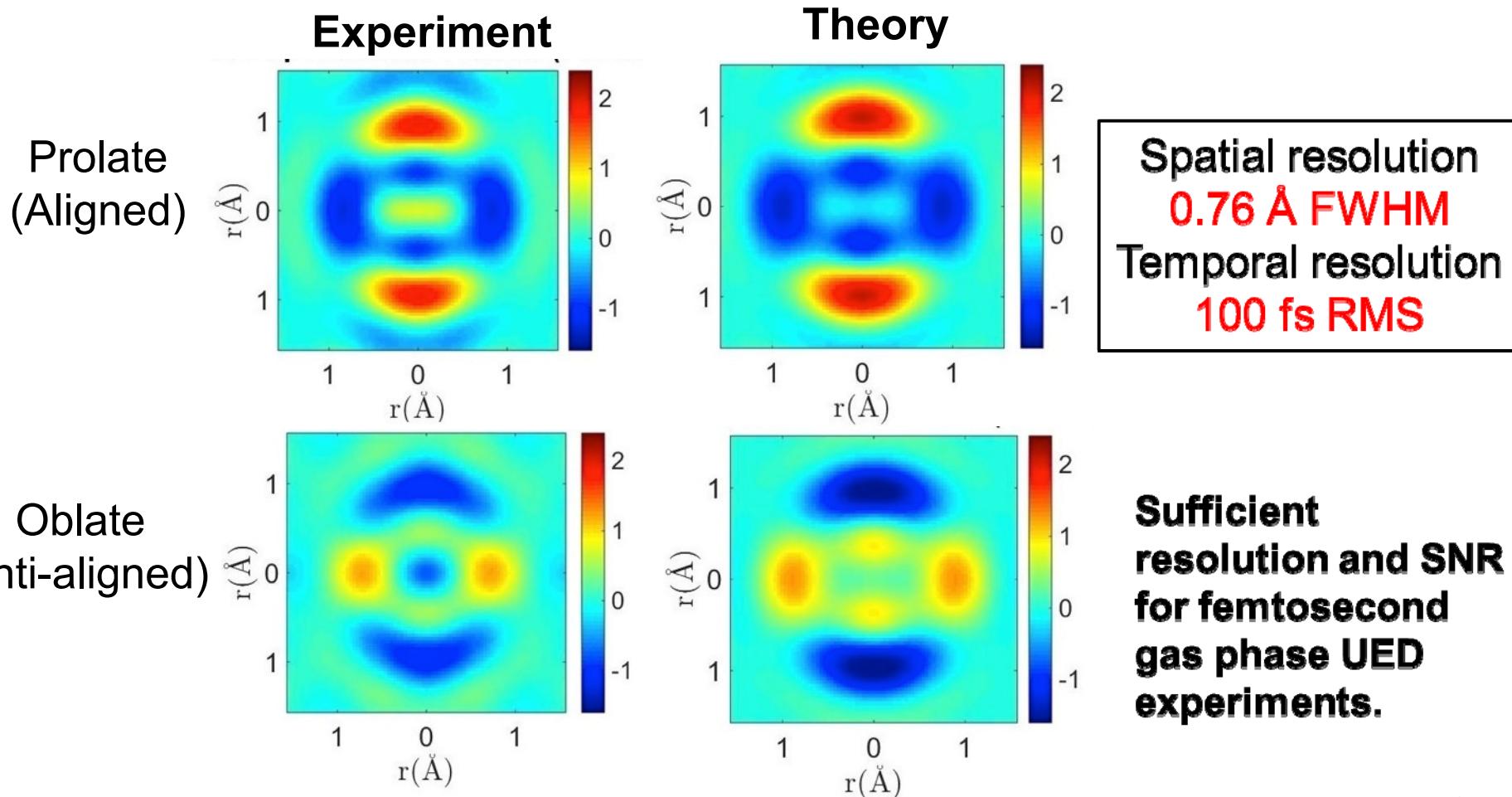


- Experimental (red) vs simulation (blue) results of N₂ rotation revival
- Experimental diffraction pattern in good agreement with simulation results

Summary of N₂ Experiment

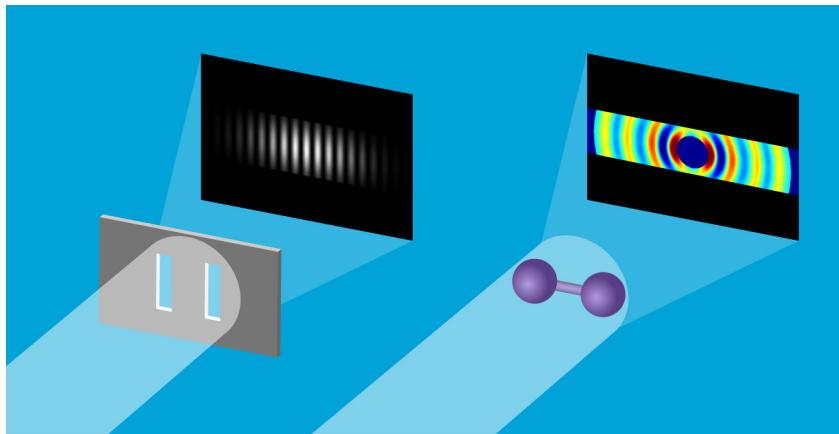
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Molecular Images from FT of Diffraction Difference Patterns

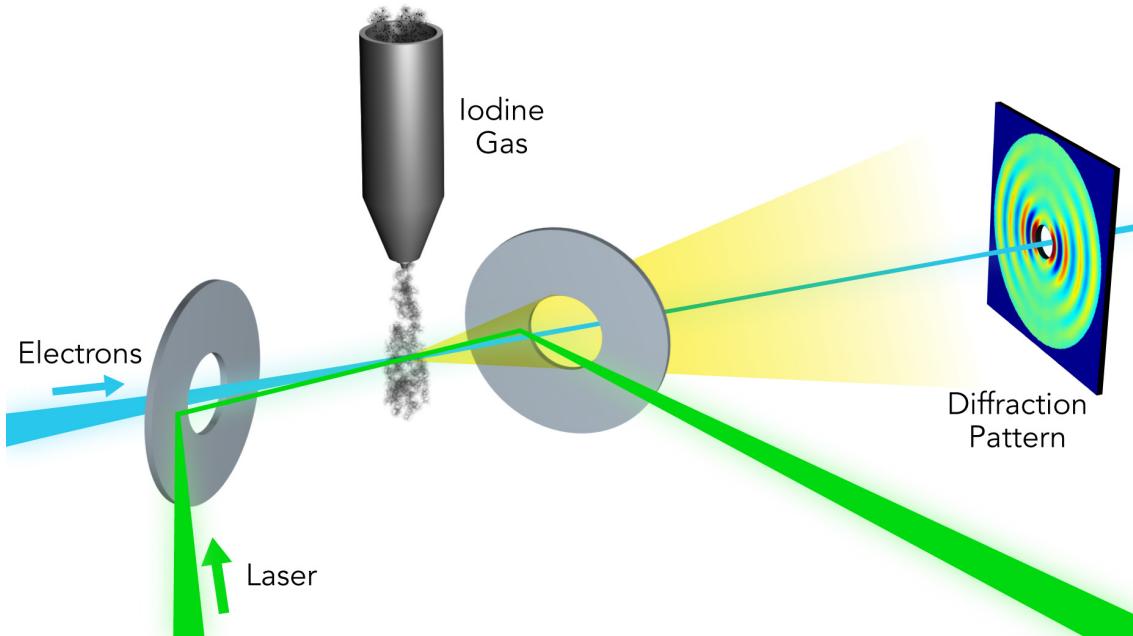


Ultrafast vibrational dynamics in I_2

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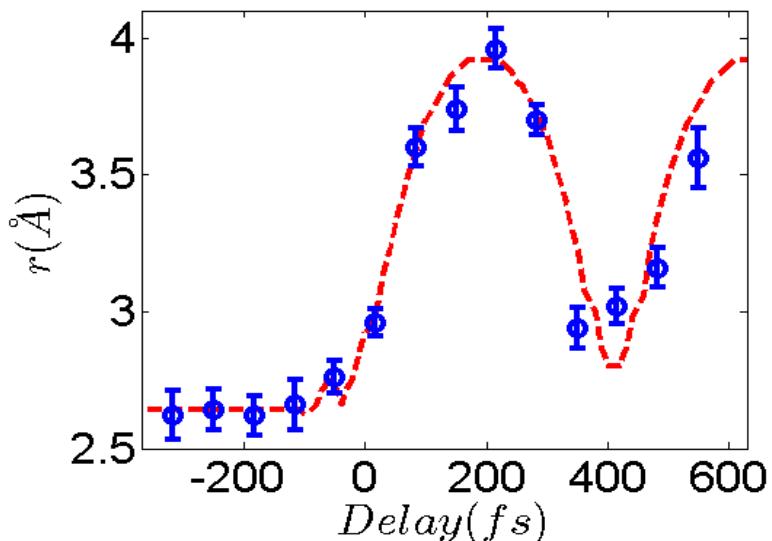
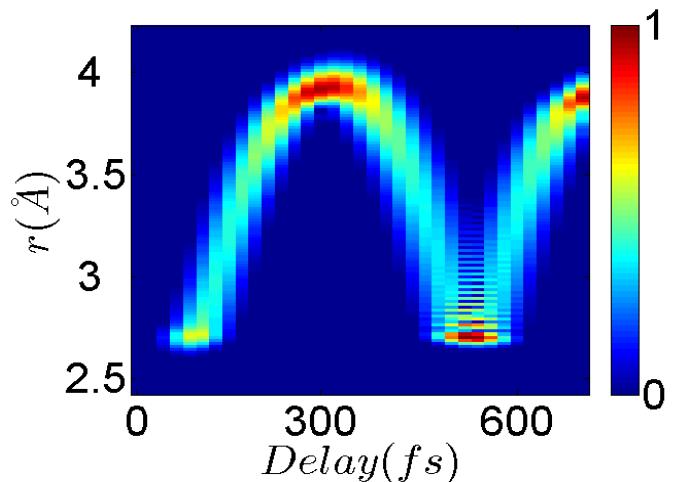
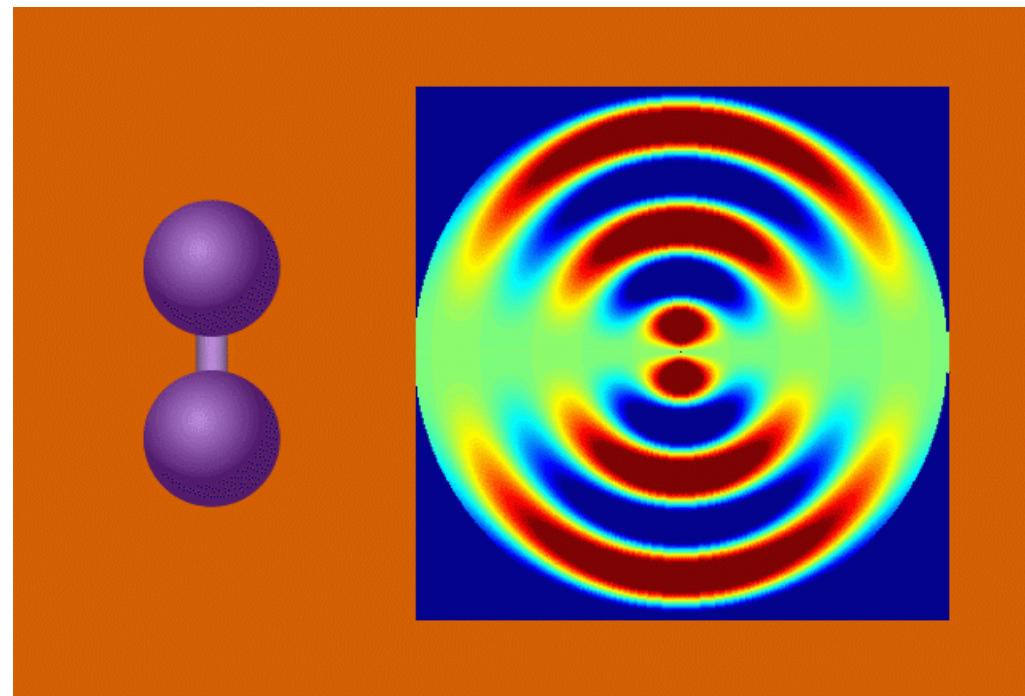
- Diatomic I_2 molecule acts like a double-slit in diffraction
- Bond length of I_2 in a vibrational state oscillates with a period about 400 fs
- Ultrafast vibrational dynamics was captured by MeV UED



- I_2 gas jet from heated pulsed nozzle
- 530 nm laser resonantly excites I_2 molecules to vibrational state
- Ultrafast vibrational dynamics probed by MeV electrons

Direct measurement of I₂ vibrational dynamics

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- Simulated diffraction pattern as a function of I₂ bond length
- Experimental results in good agreement with theories.

Summary and outlook



An MeV UED system has been constructed and commissioned, which is now serving science experiments, as well as development of next generation ultrafast electron scattering instruments.

Ongoing and future instrument developments:

- **nano-focused UED (nano-UED)**: reduce the probe size to a few um (reach 5 um), and eventually sub-um (to study single grains and nanoscale features).
- **sample environment and pump capability**: cryo-sample stage (reach 34 K). FIR and THz pump.
- **detector testing in collaboration with LBL**: direct-detection detector (under commission)
- **10s fs temporal resolution UED**: rf bunching cavity for beam compression, time stamping techniques.

Acknowledgement

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- SLAC UED/UEM 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
- Helpful discussions and suggestions by many colleagues
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Thank you for your attention!

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**