Perspectives and challenges of accelerator-based ultrafast electron diffraction(UED) and microscopy(UEM)

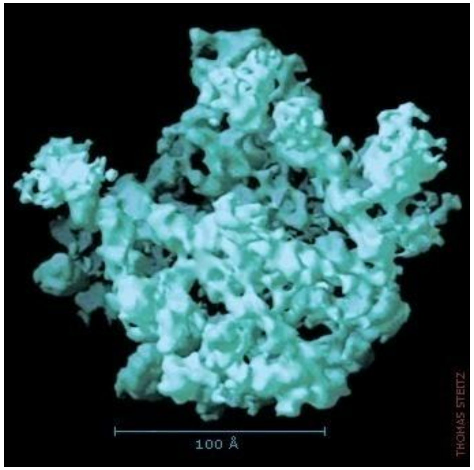
Dao Xiang(向导)
Shanghai Jiao Tong University

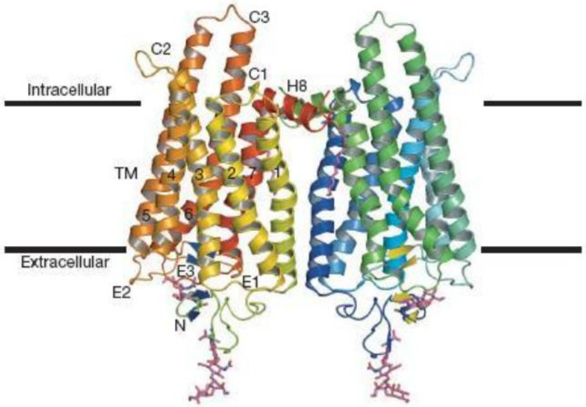
10/29/2015

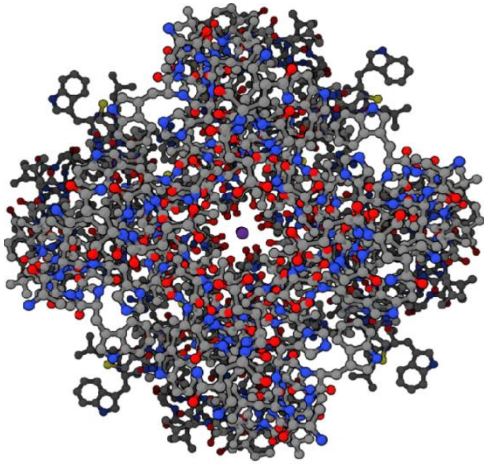
Outline

- Introduction
- keV UED/UEM: why and how
- MeV UED/UEM: why and how
- Challenges of MeV UED/UEM
- UED/UEM center at SJTU
- Summary

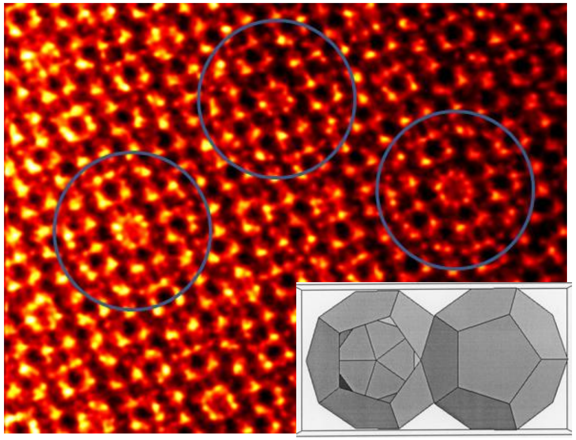
The most important part of science: observation

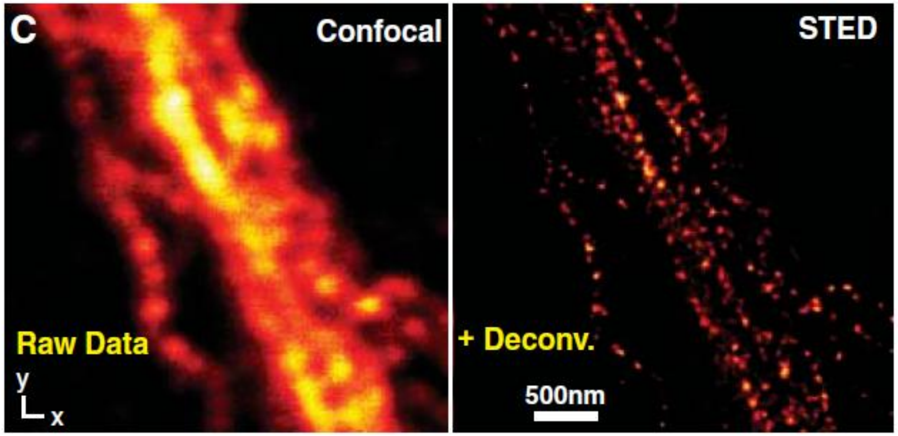






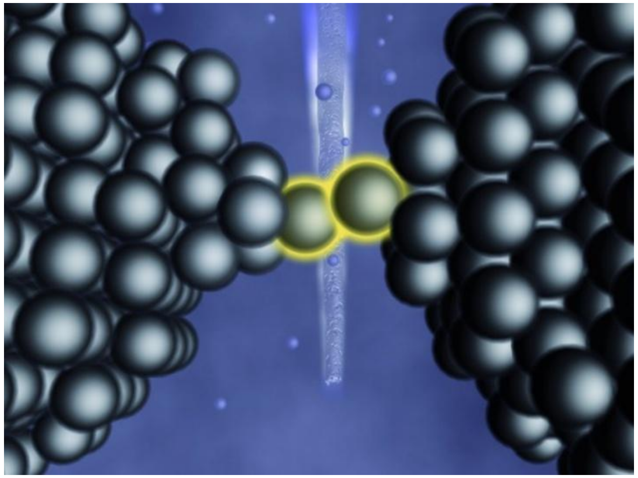
Ribosome, ATP, GPCR etc.;
1997, 2003, 2006, 2009, 2012 Nobel prize in Chemistry

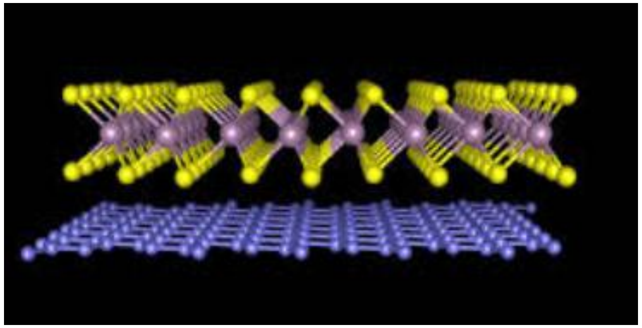




Quasi-crystal, super-resolved fluorescence microscopy;
2011 and 2014 Nobel prize in Chemistry

The most important part of science: observation





Galaxy

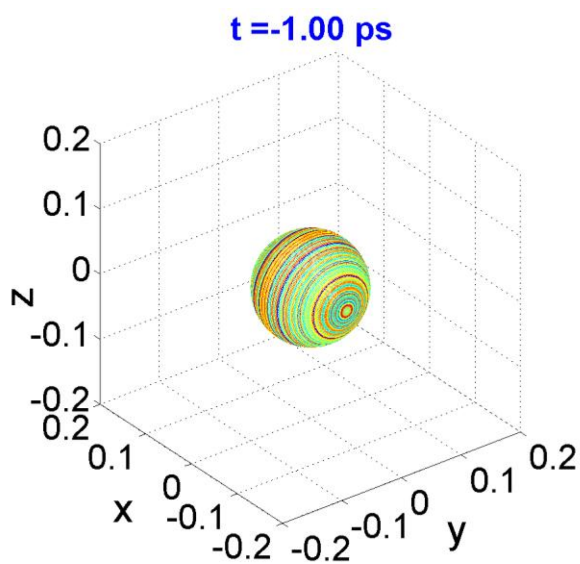
Atom

The 3D static world is no longer a mystery to us

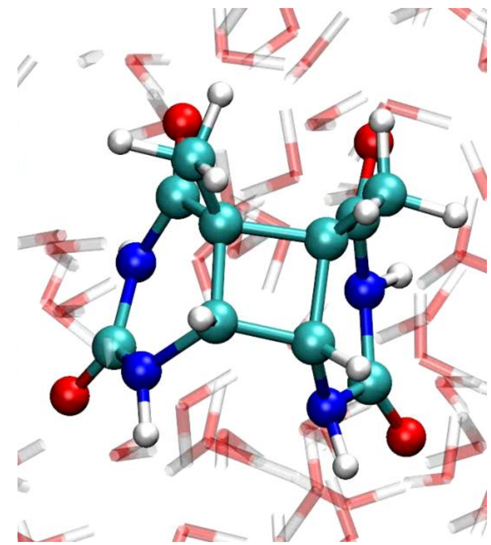
The BIG challenge

- All matter around us consists of atoms and electrons; their structure determines its properties.
- Any reaction or process is essentially defined by movement paths on an atomic level.
- Understanding the functionality requires observation with **both high spatial and temporal resolution.**

Horse galloping
(Muybridge/Stanford, 1878)

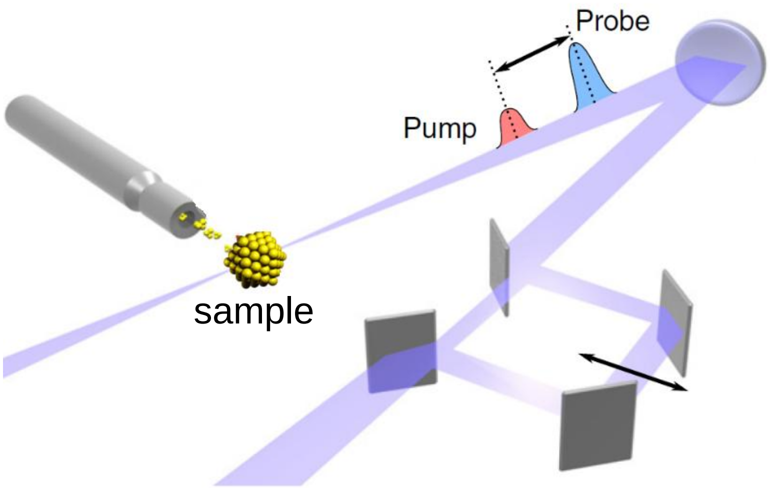


Molecular rotation
(Wu, ECNU)

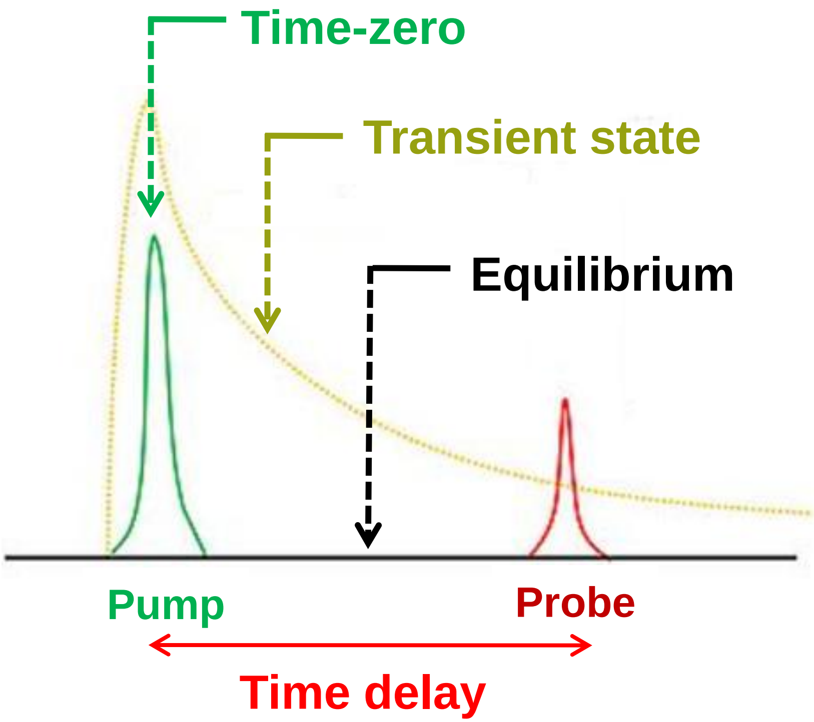


Thymine dimer splitting
(Zhong, OSU)

Pump-probe technique for ultrafast science



Schematic of a pump-probe experiment



- A pumping pulse to initiate a dynamical process
- A pumping pulse drives the system out of equilibrium state
- Probing pulse measures the response
- Pumping pulse defines time zero
- Changing time delay to obtain a complete picture

Three probes in ultrafast science

Laser

- ✓ A few femtosecond
- ✓ Fully coherent
- ✓ No radiation damage

- × Poor resolution in imaging

X-ray

- ✓ Atomic resolution
- ✓ Attosecond possible
- ✓ Fully coherent possible
- ✓ Large penetration depth

- × Small cross section
- × Massive and expensive
- × Serious radiation damage

Electron

- ✓ Atomic resolution
- ✓ Large cross section
- ✓ Small facility

- × Space charge effect
- × Poor coherence

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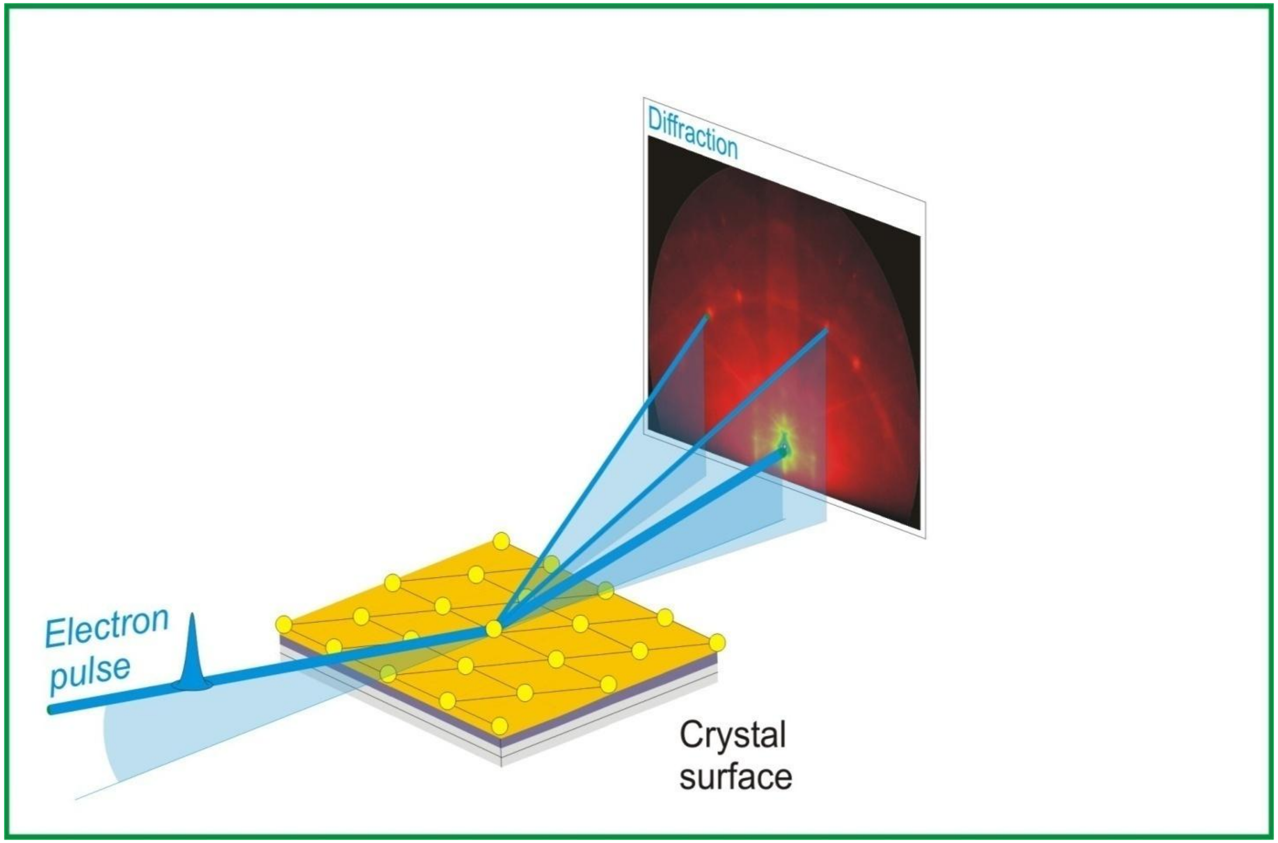
Ultrafast electron diffraction

- **J.J. Thomson: electrons are particles (1906); G.P. Thomson: electrons are NOT particles (1937).**
- **Structure information encoded in the diffraction pattern**



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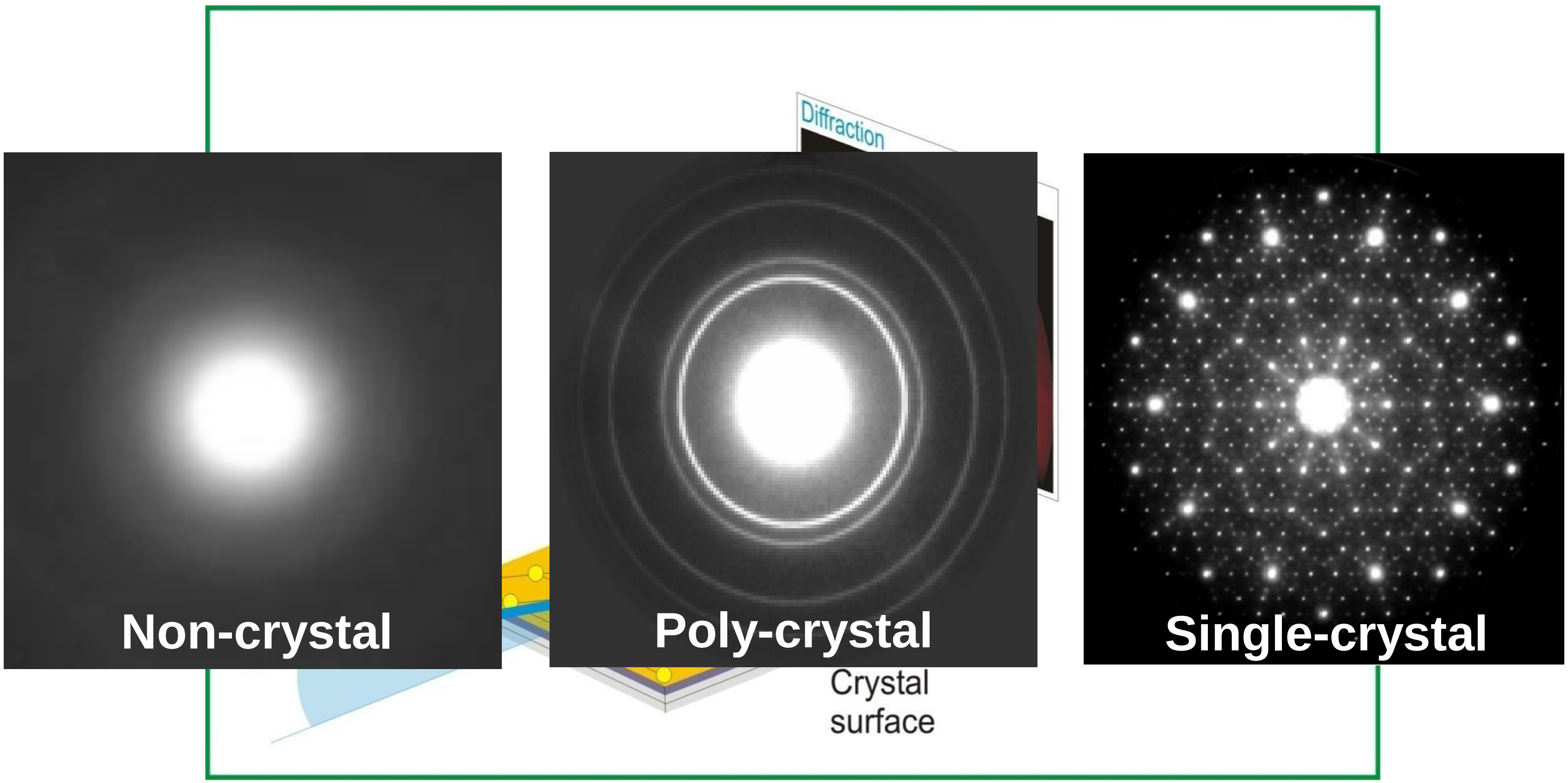

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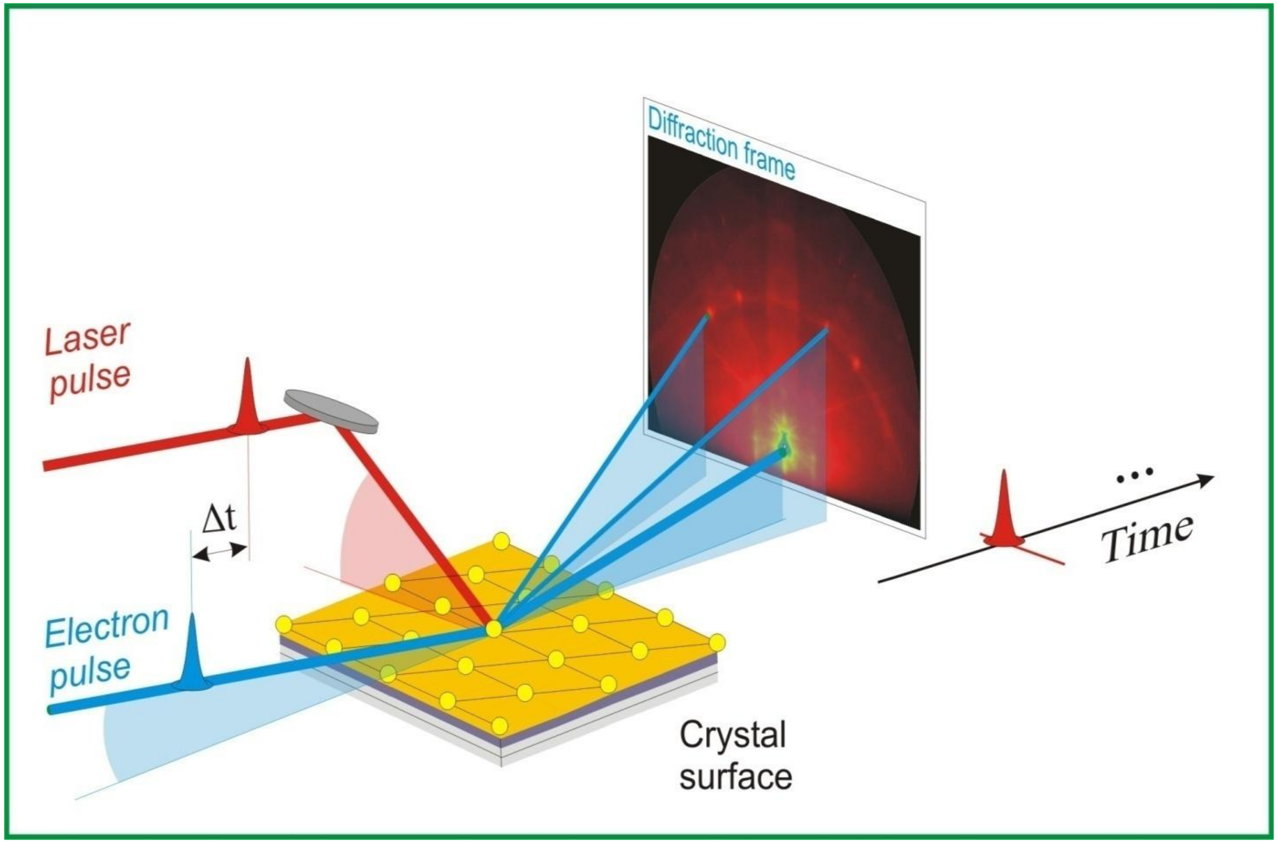

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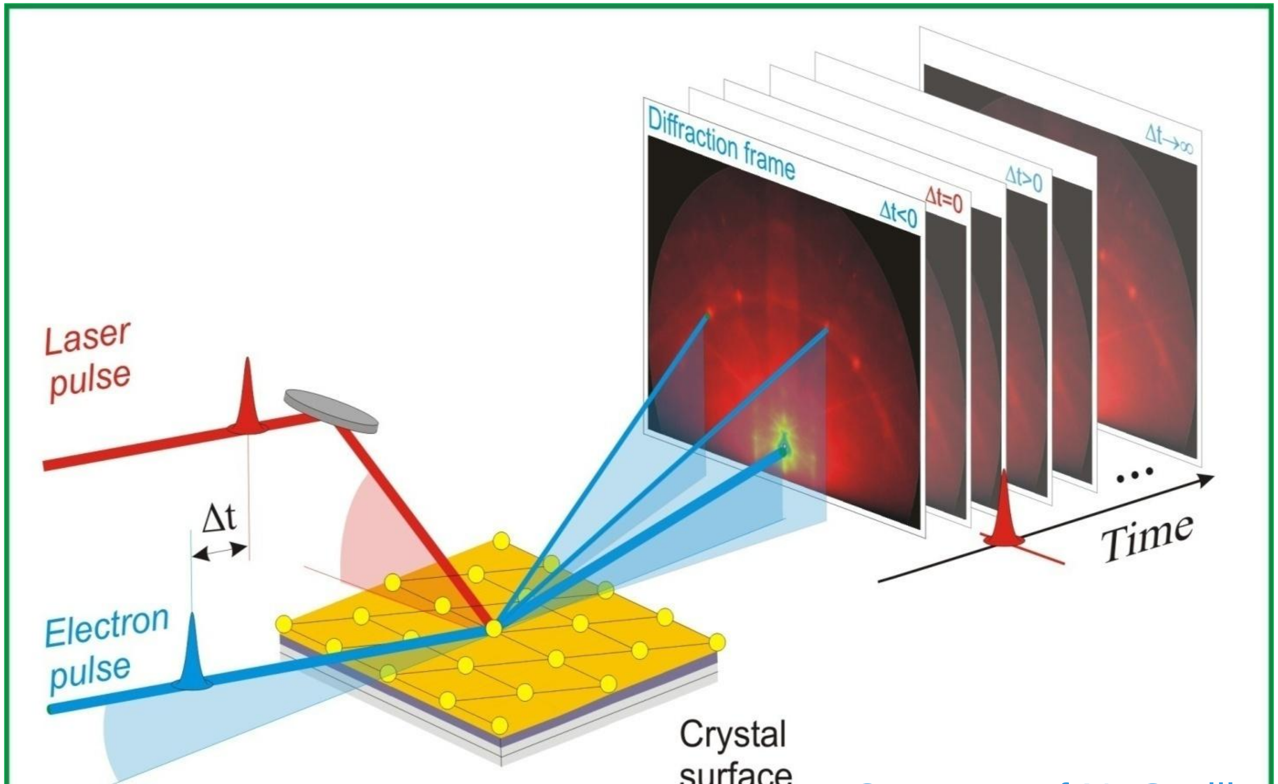
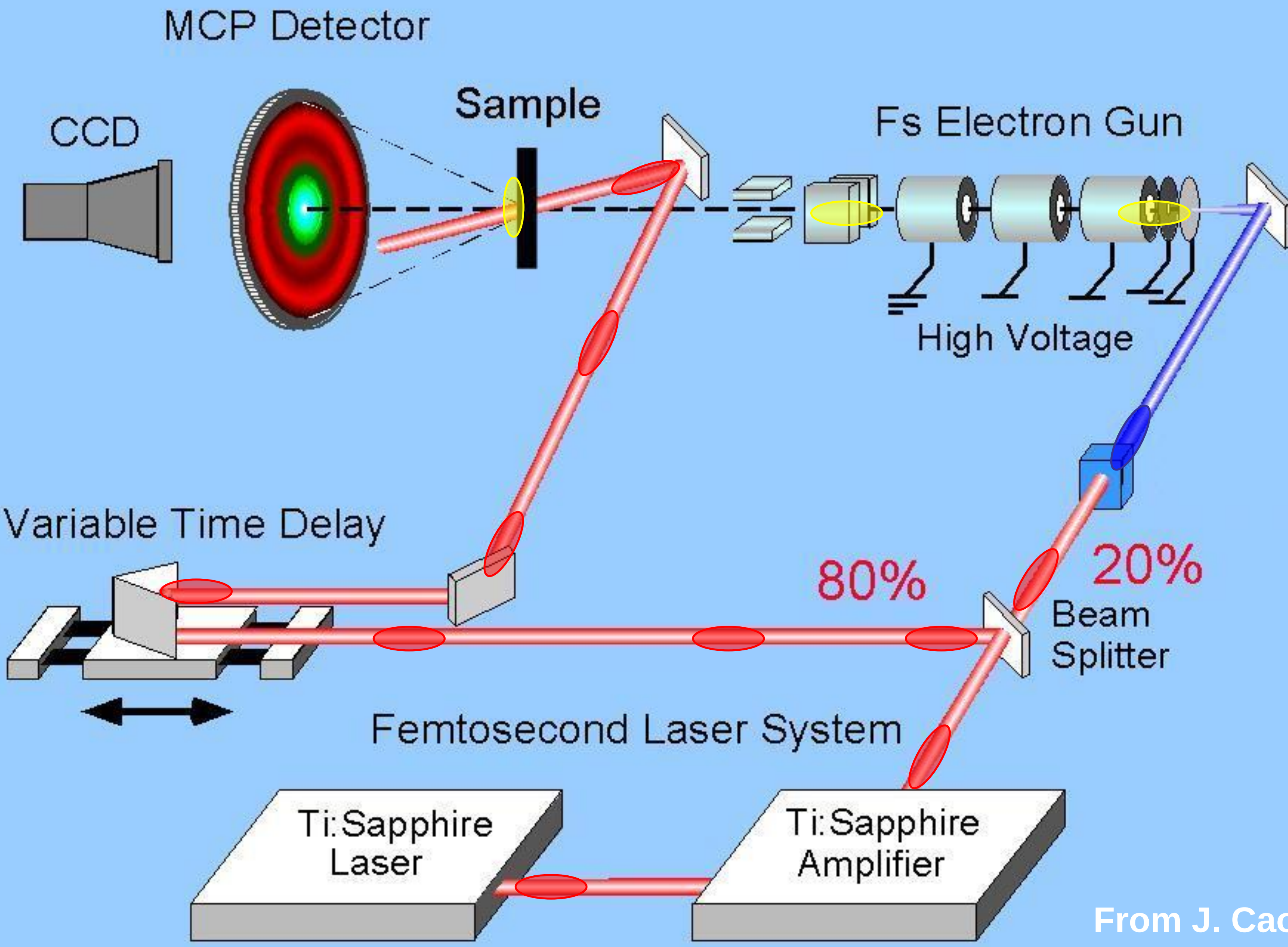


Courtesy of N. Gedik

With positions of the ball and sportsman recorded, one can reproduce the process of jump-serving in volleyball.

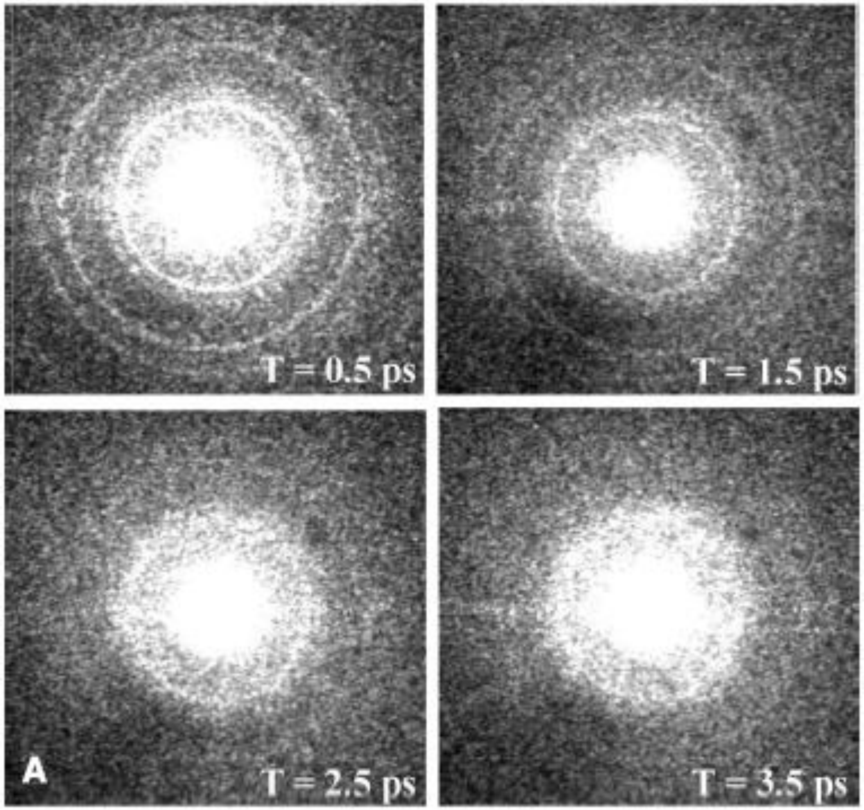
Schematic of a keV UED



From J. Cao (FSU)

UED applications

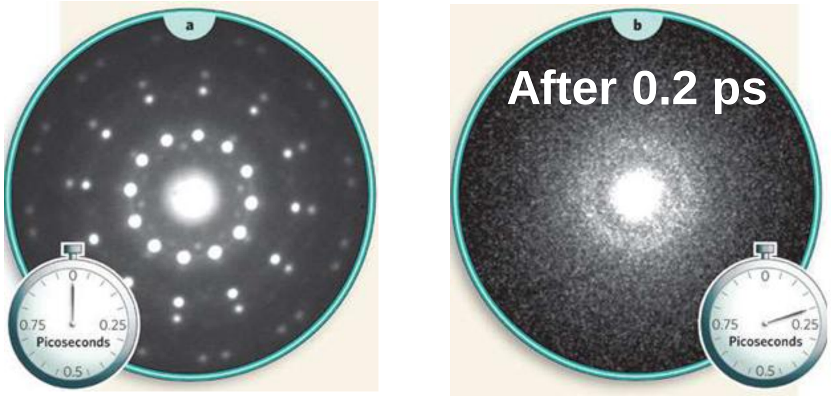
Phase transition



An atomic view of phase transition in Aluminium

Science (2003)

Melted in a flash!



"How does the laser-excited, ordered solid know how to become disordered without letting its atoms bounce around a couple of times to find the new ground state?"

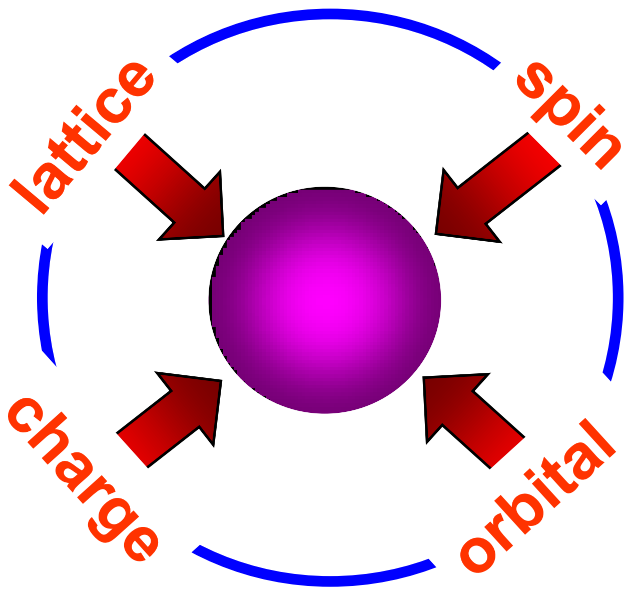
Disorder & weak bond

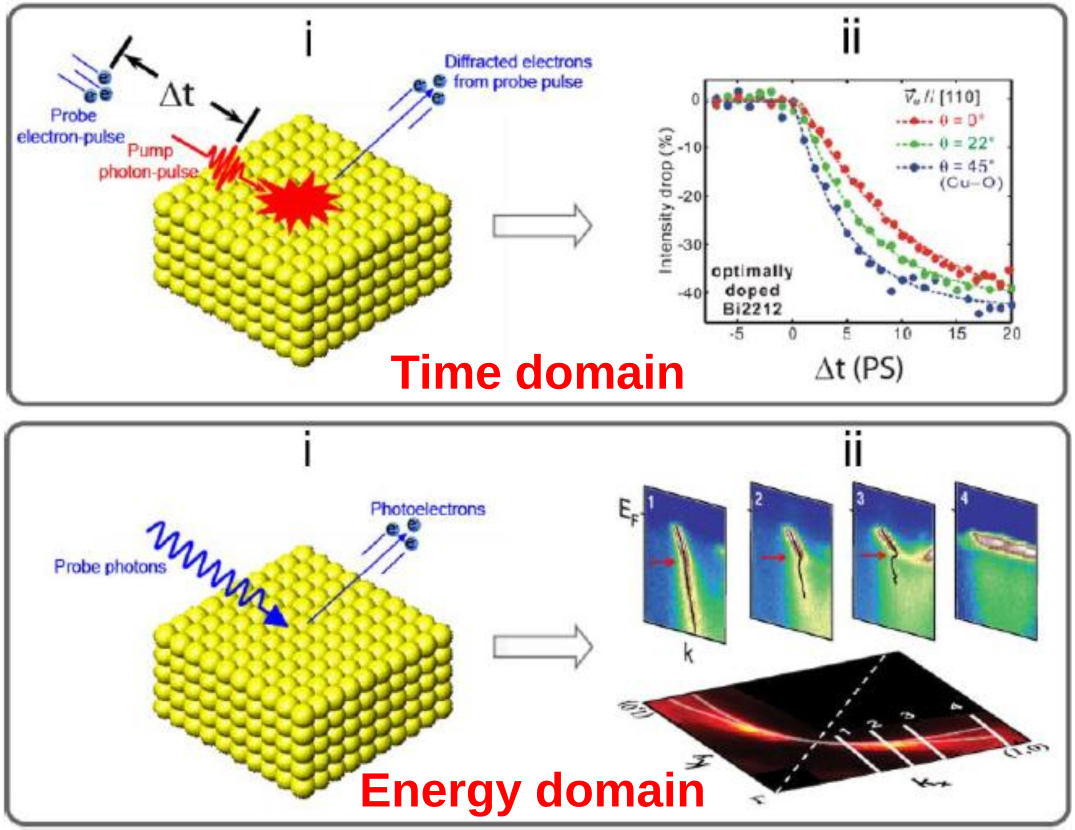
Bismuth

Nature (2009)

UED applications

Strongly correlated electron materials





Zewail et al., PNAS 105, 20161 (2008)

A step closer to visualizing the electron–phonon interplay in real time

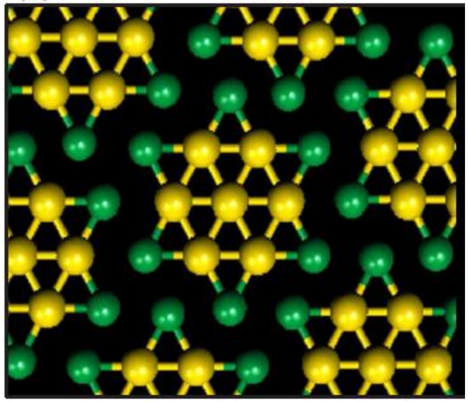
Y. L. Chen, W. S. Lee, and Z. X. Shen[1]
Departments of Applied Physics and Physics, and SLAC Photon Science, Stanford University, Stanford, CA 94305

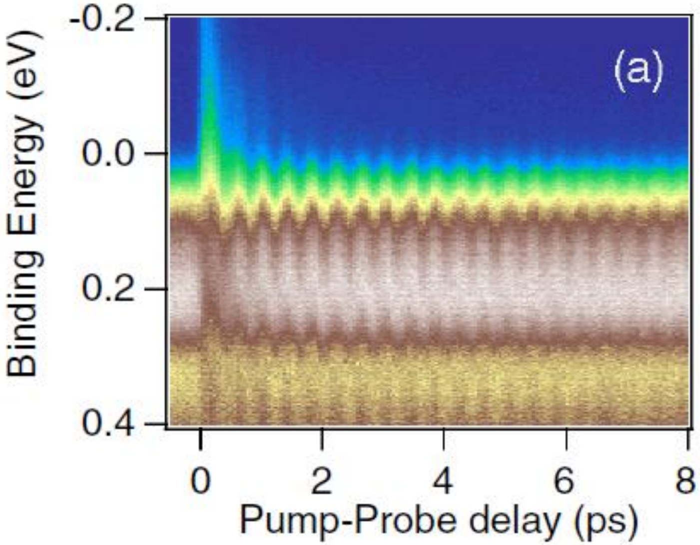
Bi2212: anisotropy electron–phonon coupling within the CuO2 plane

UED applications

Strongly correlated electron materials

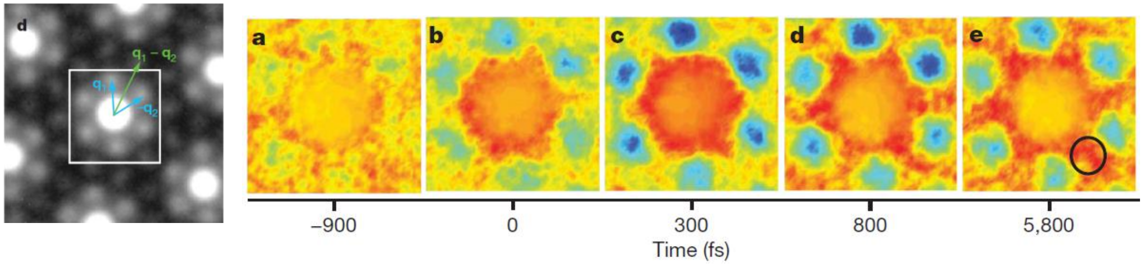
1T-TaS2, tr-ARPES

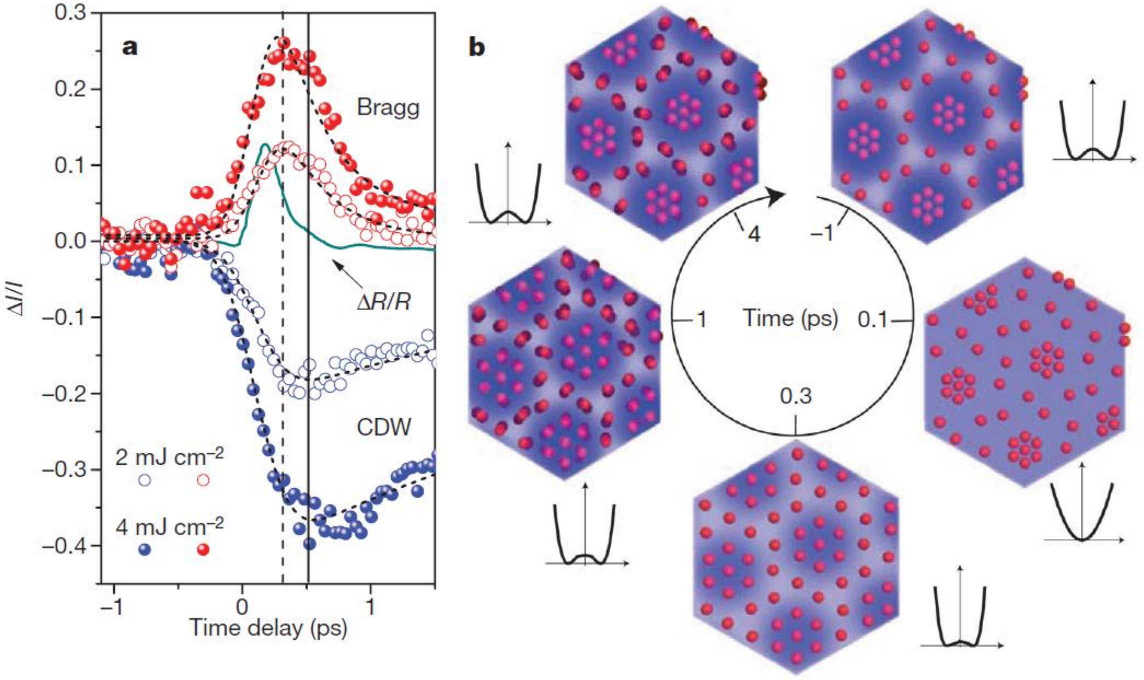




PRL 97, 067402 (2006)

1T-TaS2, UED





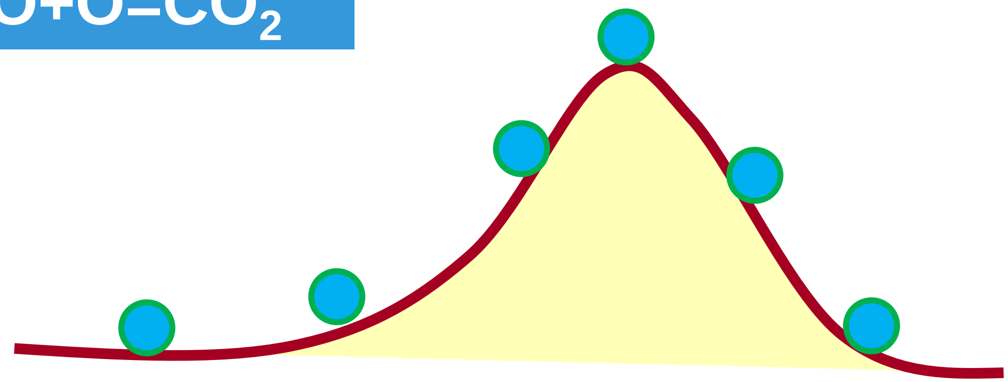
Nature 468, 799 (2010)

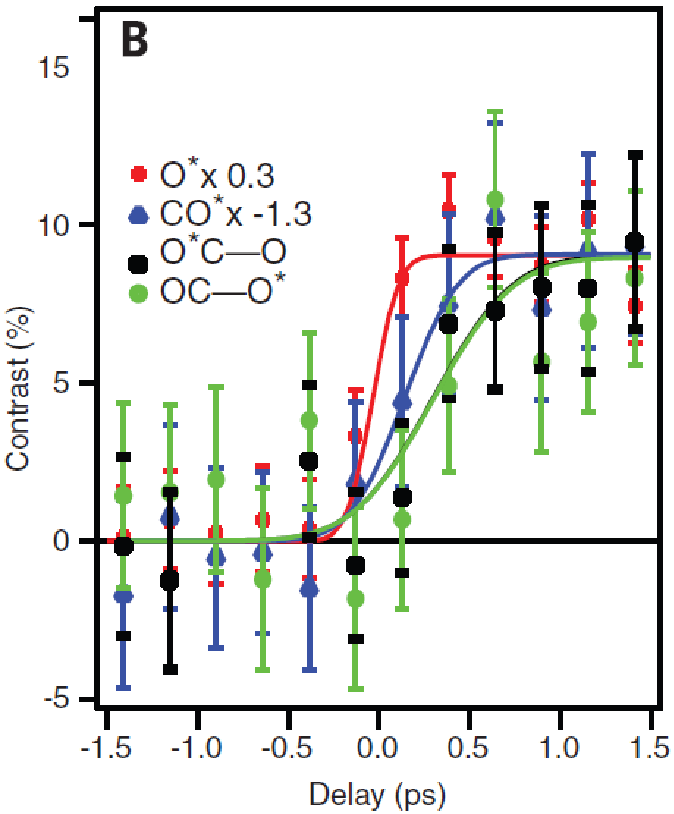
UED applications

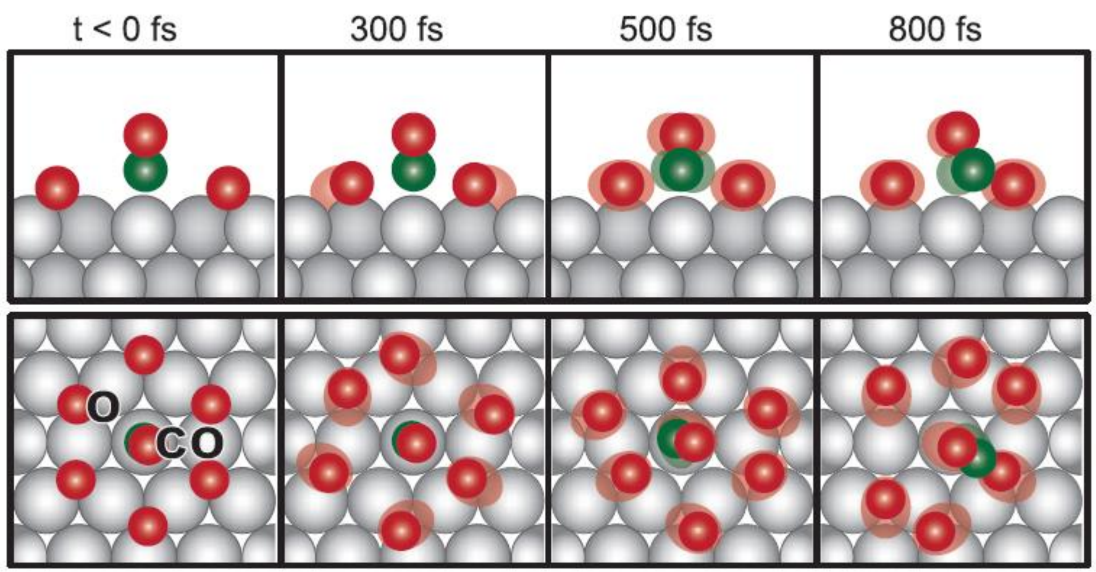
Chemical bond forming and breaking

Carbon monoxide oxidation reaction on Ru initiated by an optical laser pulse

$CO+O=CO_2$



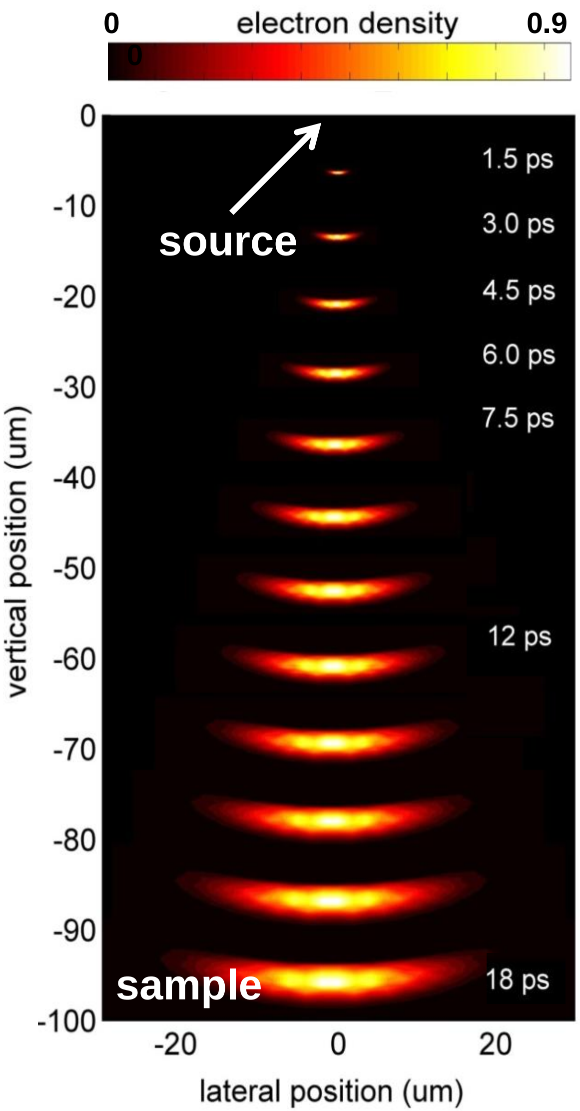


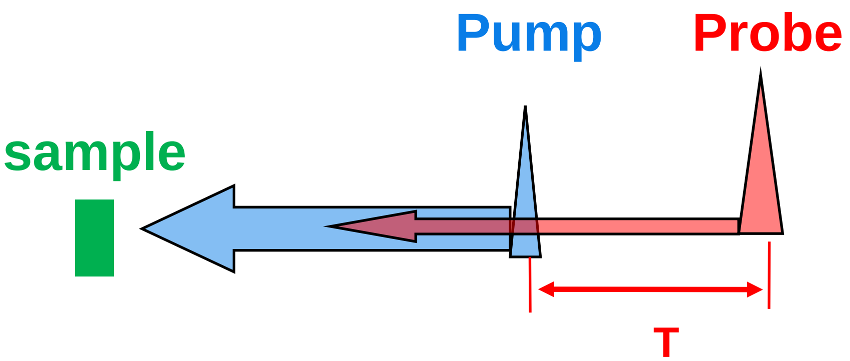


Science (2015)

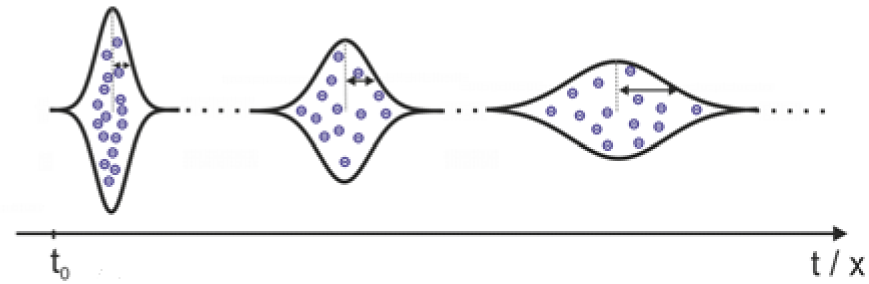
keV UED resolution limit

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





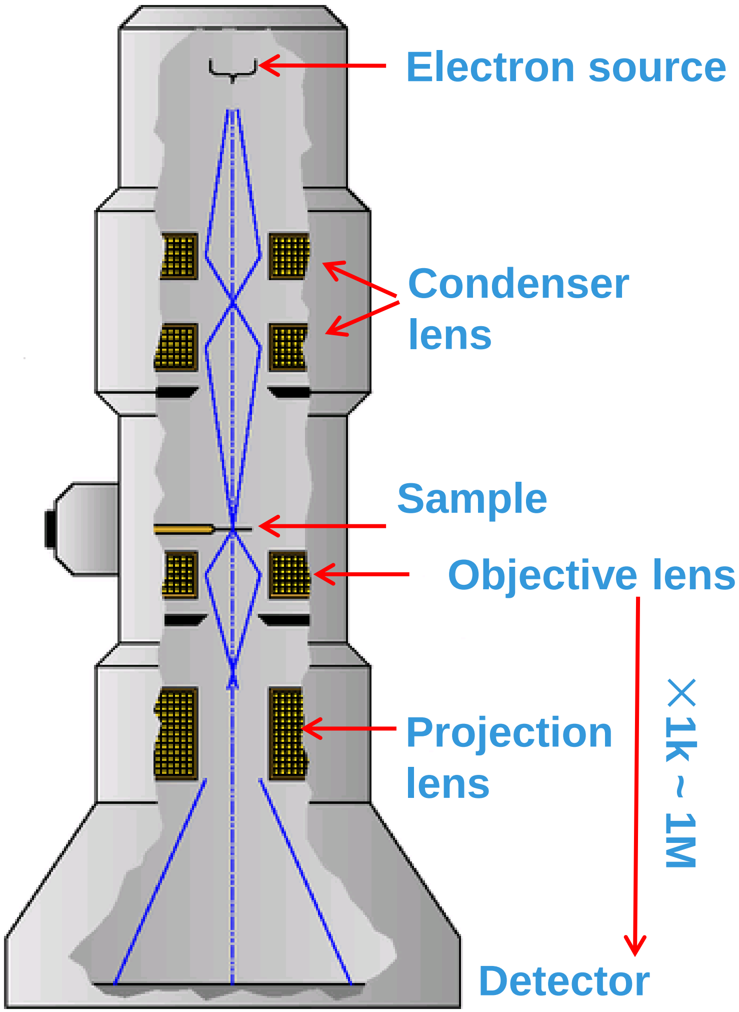
Space charge



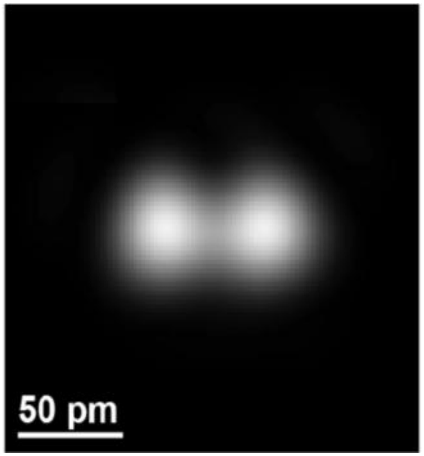
- **e-beam bunch length limited by space charge and dispersion effects**
- **Velocity mismatch limited resolution for studies of gas phase samples (for 60 keV electrons, *v* ≈ 0.45 c)**

UEM: Ultrafast electron microscope

Transmission EM

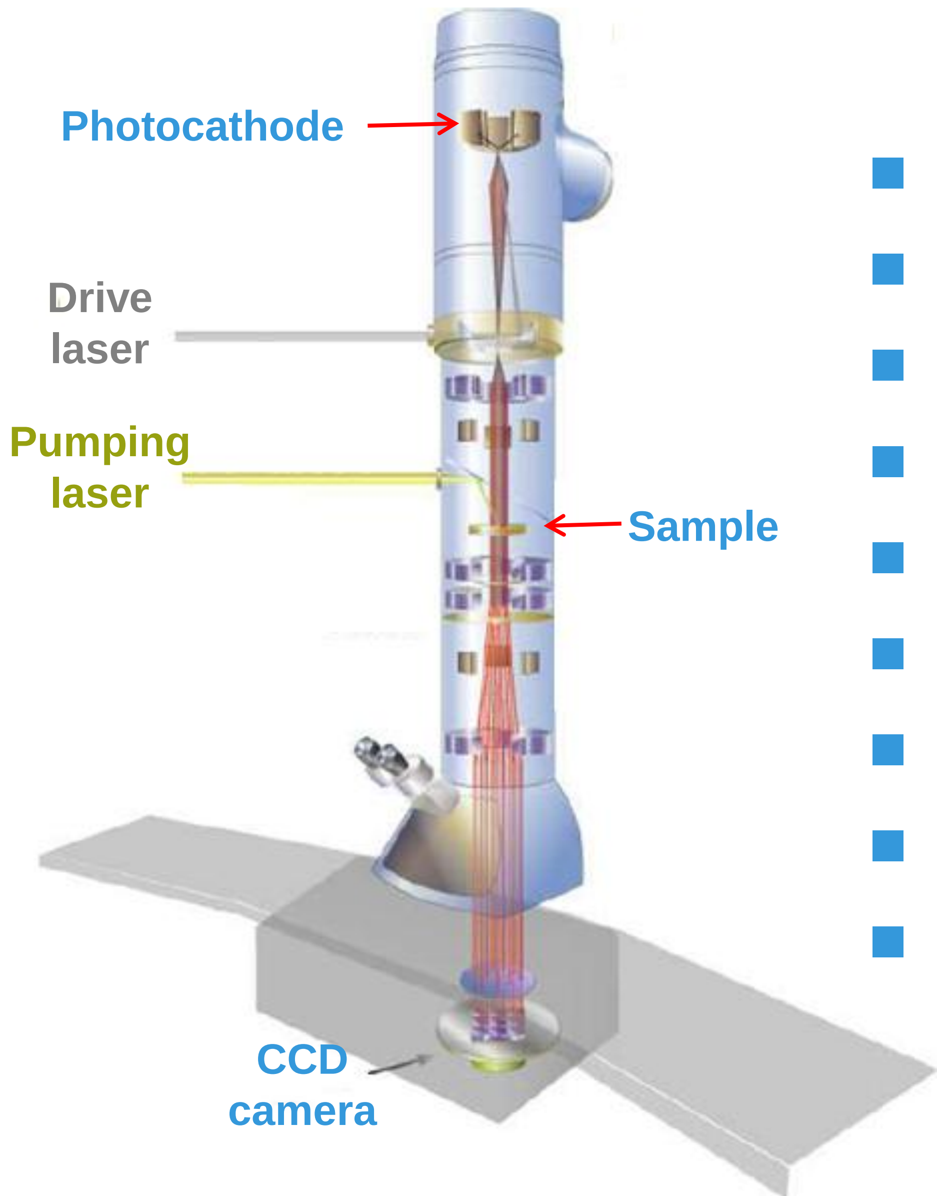


- Gun: thermionic or field emission
- Acceleration with DC field
- Voltage: <200 kV (routine); 200 ~ 500 kV (medium energy); 500 kV ~ 3 MV (high voltage)
- Spatial resolution: down to 50 pm



5[th] order aberration corrected TEM (2009)

UEM: Ultrafast electron microscope



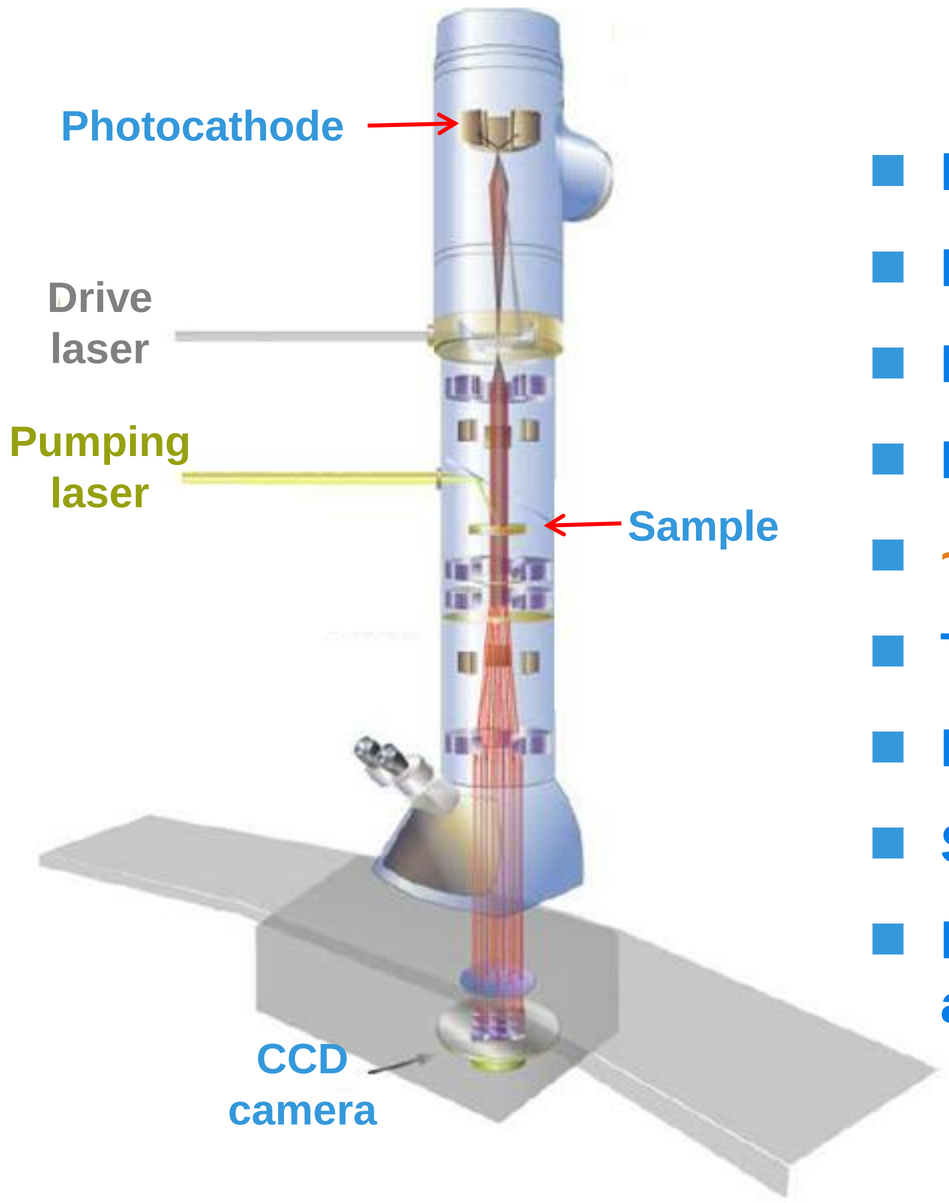
Prof. A. Zewail's group

Stroboscopic

- Modified from a commercial TEM
- Beam energy: <200 keV
- Laser triggered photocathode gun
- Pumping laser
- ~ 1 electron per pulse (multiple shot)
- Temporal resolution: ~100 fs
- Spatial resolution: ~0.1 nm
- Integration over 10^8 shots
- Perfectly reversible process

Caltech's 4D-EM

UEM: Ultrafast electron microscope

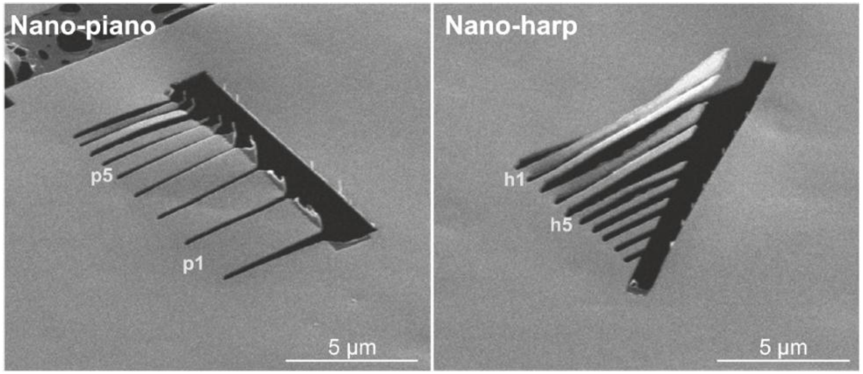


Single shot

- Modified from a commercial TEM
- Beam energy: ~200 keV
- Laser triggered photocathode gun
- Pumping laser
- ~ 10^8 electrons per pulse
- Temporal resolution: ~10 ns
- Peak current: ~1 mA
- Spatial resolution: ~10 nm
- Resolution limited by space charge and beam brightness

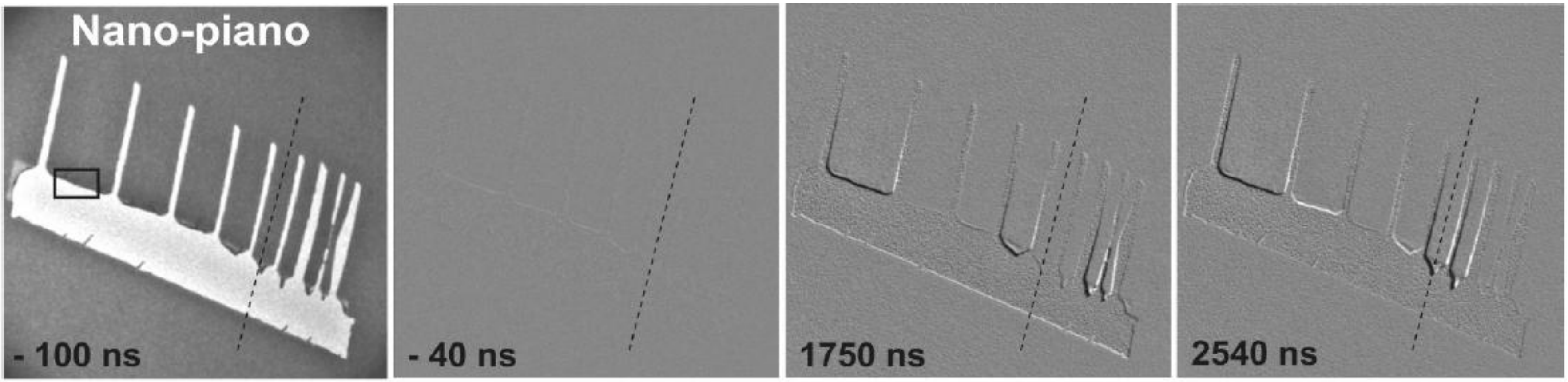
LLNL's DTEM

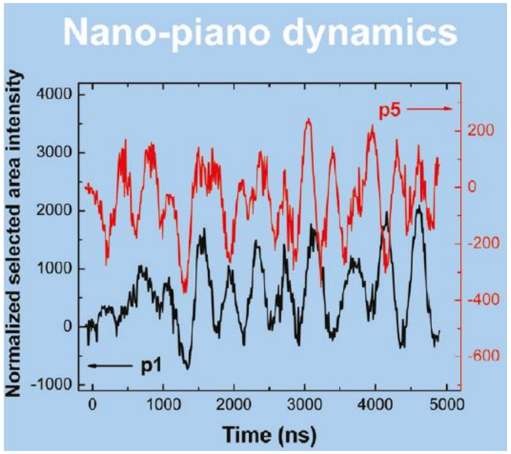
UEM applications

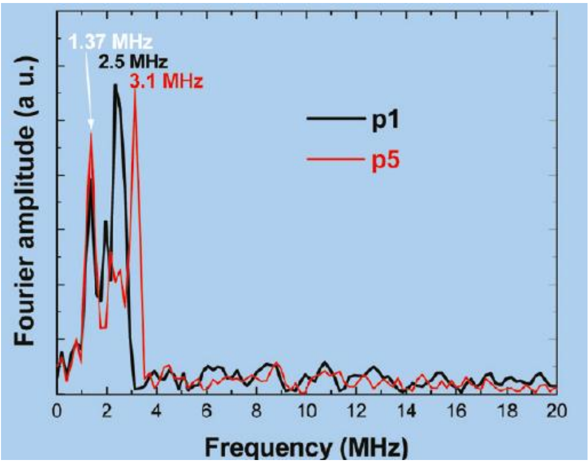


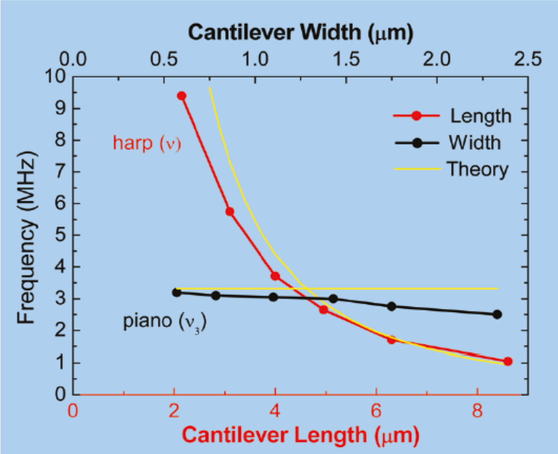
Real-time imaging of mechanical motions of nano-structures (MEMS/NEMS)

Nano Lett. 11, 2183 (2011)









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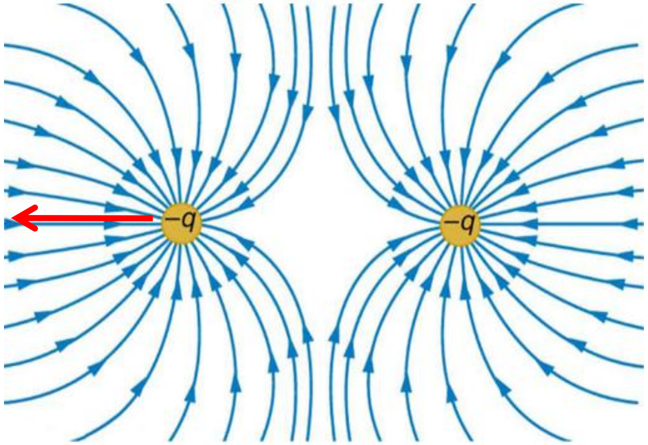
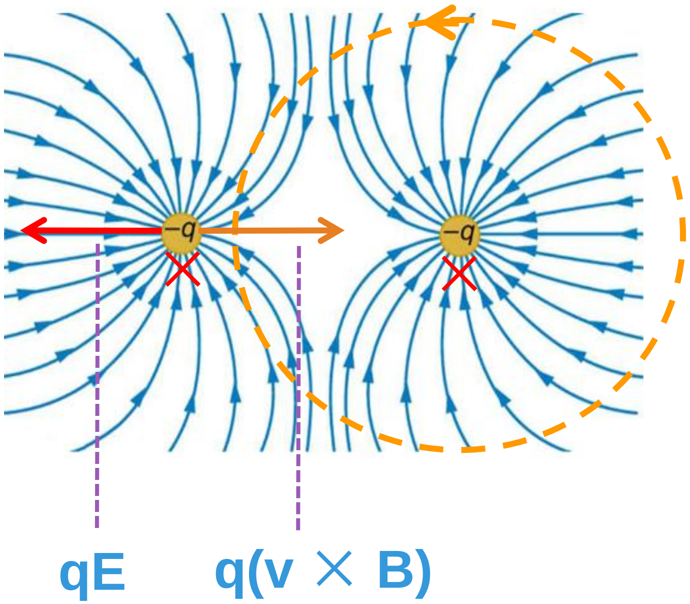
UED/UEM goes relativistic!



The electric force and magnetic force cancel if v=c

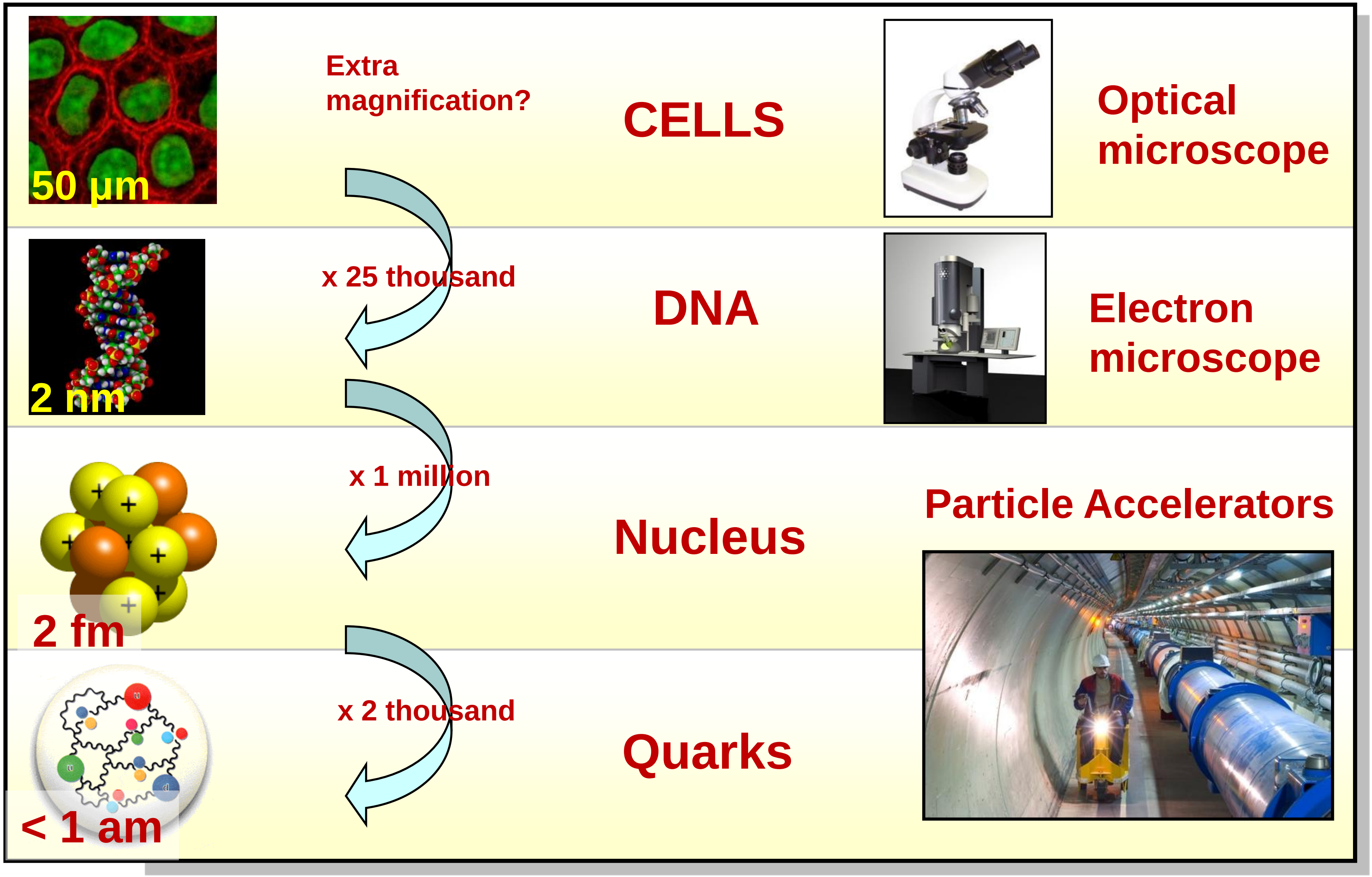
3 MV TEM@ Osaka

F=q[E+(v×B)]



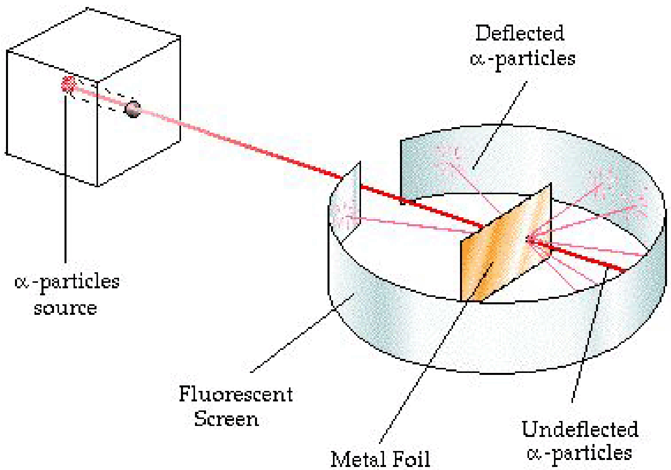
1 MV TEM@ Beijing

The quest for higher resolution



What is an accelerator

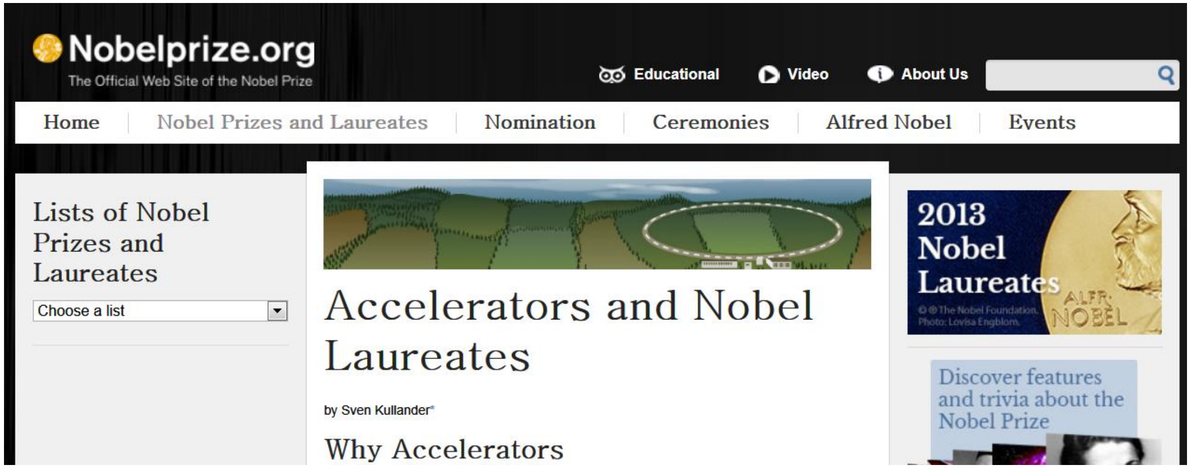
Use electromagnetic fields to propel charged particles to high speeds



I have long hoped for a source of positive particles more energetic than those emitted from natural radioactive substances.

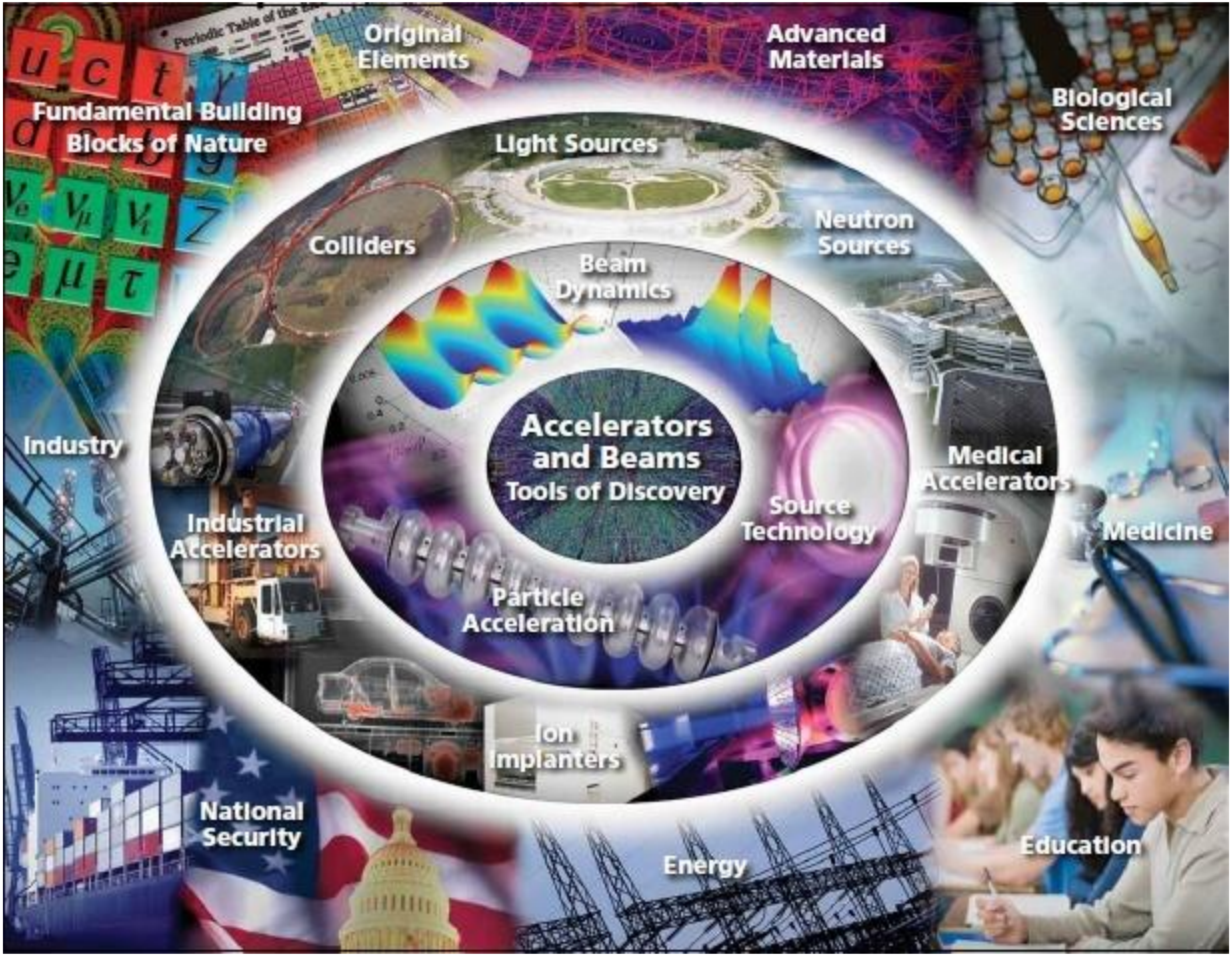
Ernest Rutherford, 1928

Rutherford's alpha scattering experiment



On average accelerator science has contributed to a physics Nobel Prize winning research every 2.9 years since 1938.

Applications of accelerators



Accelerator-based MeV UED

First proposals and feasibility studies

Journal of the Korean Physical Society, Vol. 48, No. 3, March 2006, pp. 390∼396

Potential of Femtosecond Electron Diffraction Using Near-Relativistic Electrons from a Photocathode RF Electron Gun

X. J. Wang
National Synchrotron Light Source, Brookhaven National Laboratory, Upton, NY 11973, USA

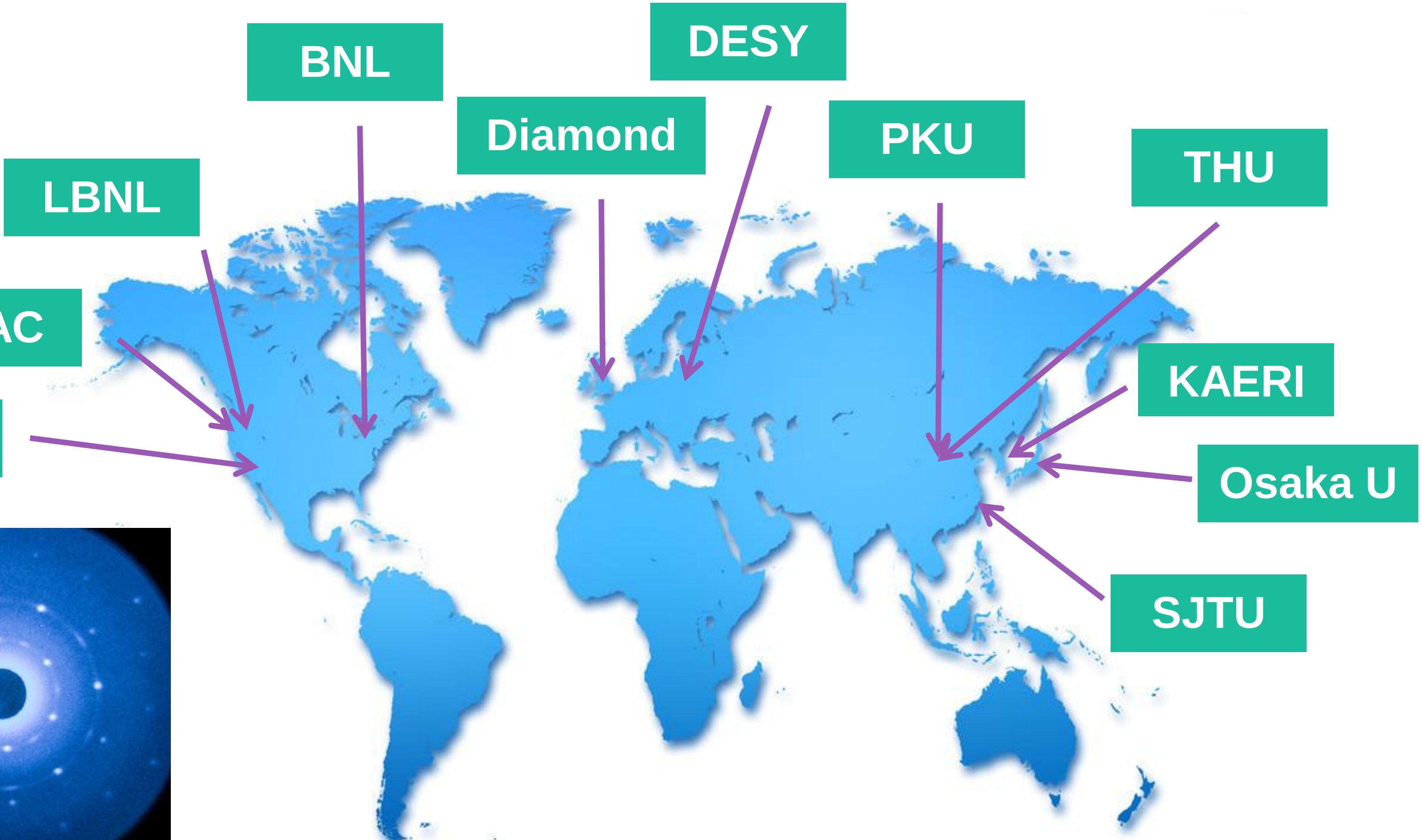
D. Xiang
Department of Engineering Physics, Tsinghua University, Beijing, China

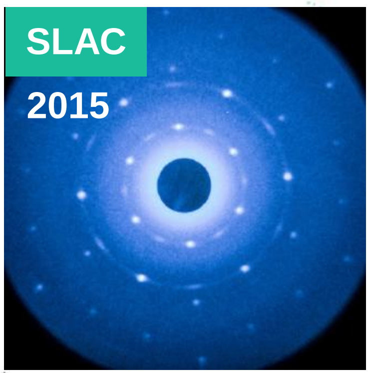
T. K. Kim and H. Ihee*
Department of Chemistry and School of Molecular Science (BK21),
Korea Advanced Institute of Science and Technology, Daejeon 305-701

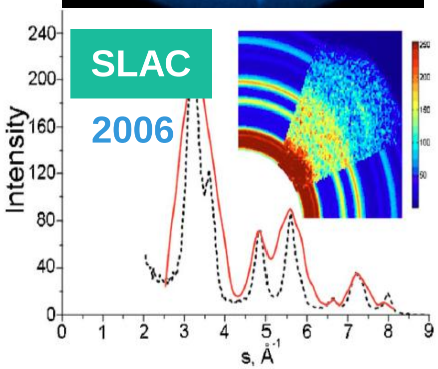
(Received 20 December 2005)

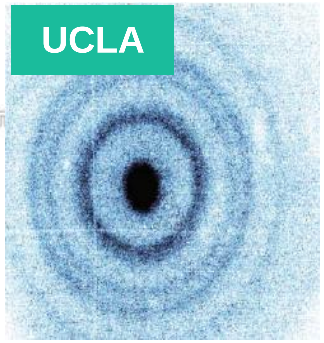
Wang, BNL LDRD, 2000; Wang et al., PAC03; Xiang et al., PAC05; Wang et al., 2006

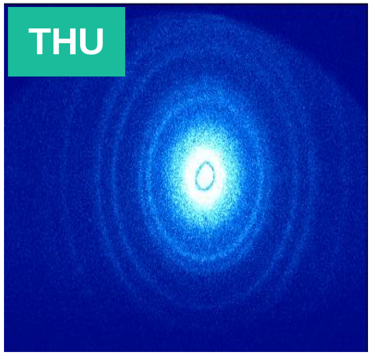
Accelerator-based MeV UED: world-wide efforts

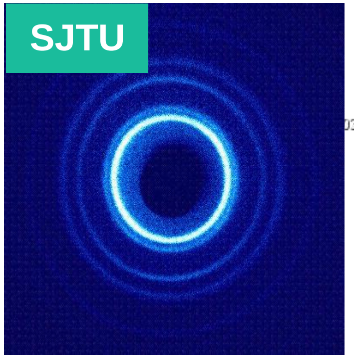


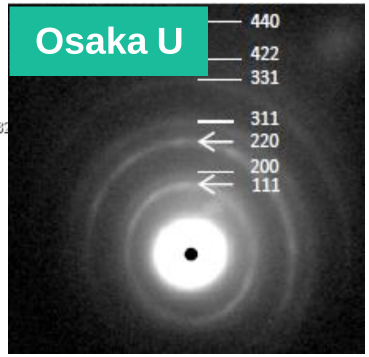






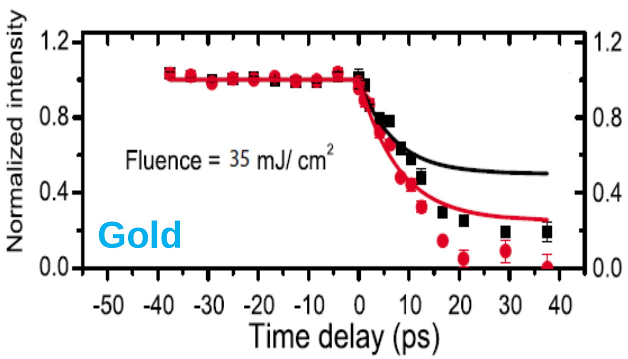


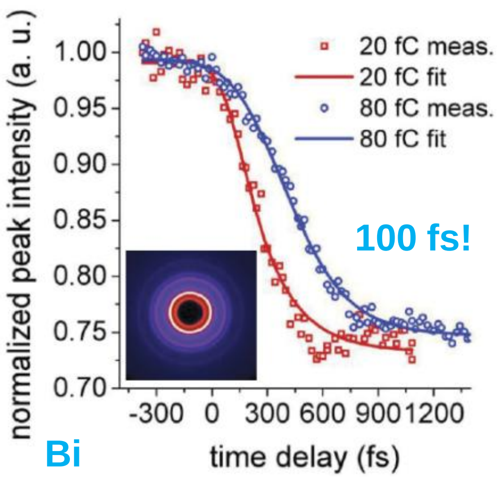




Accelerator-based MeV UED: applications

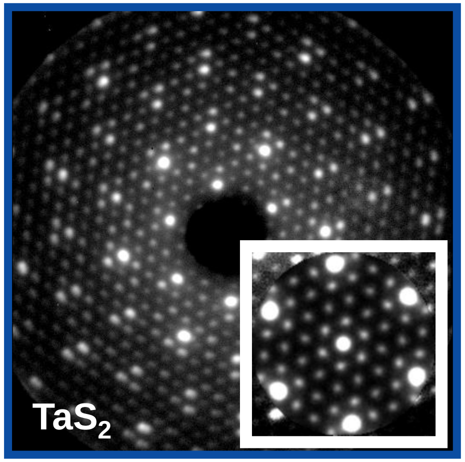
Phase transition

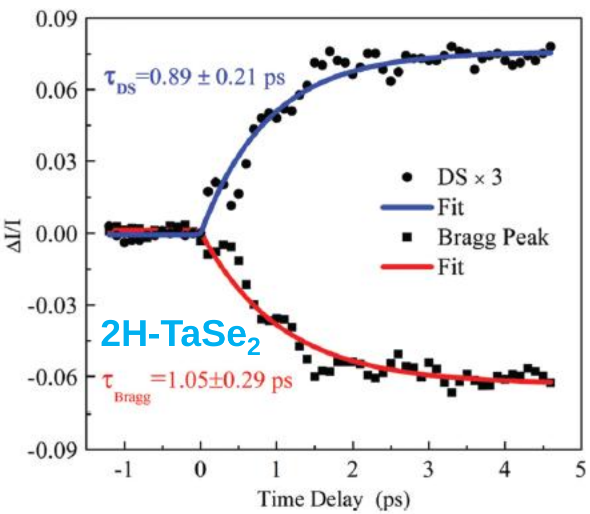




UCLA, 2010; Osaka U, 2013; SJTU, 2014; THU, 2014; SLAC, 2015

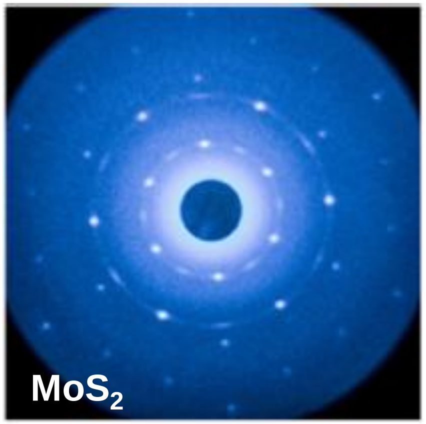
Superlattice

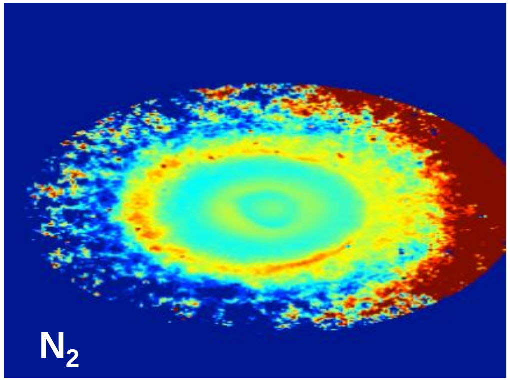




BNL-FSU-SJTU Collaboration, 2013

2D material and gas phase sample

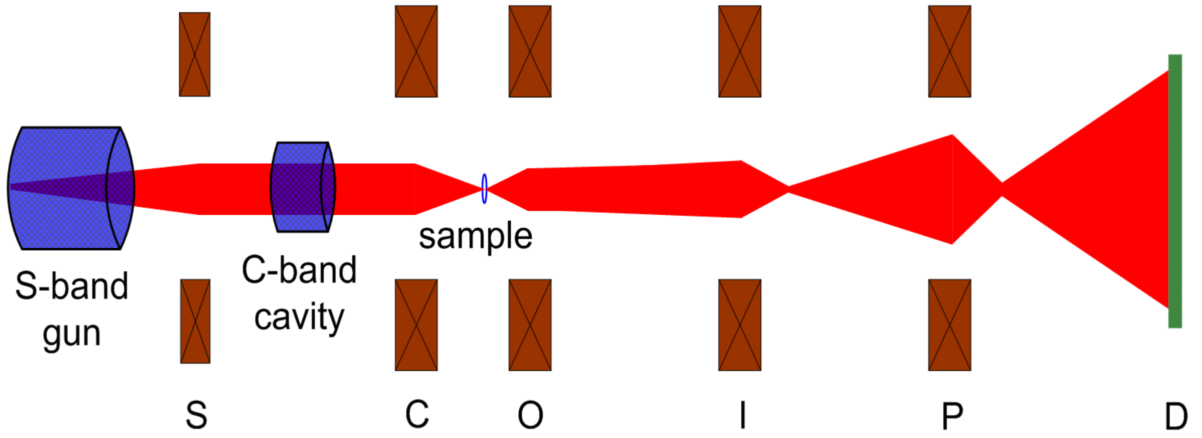




SLAC, 2015
~100 fs resolution

Accelerator-based MeV UEM

Conceptual design with superconducting solenoids

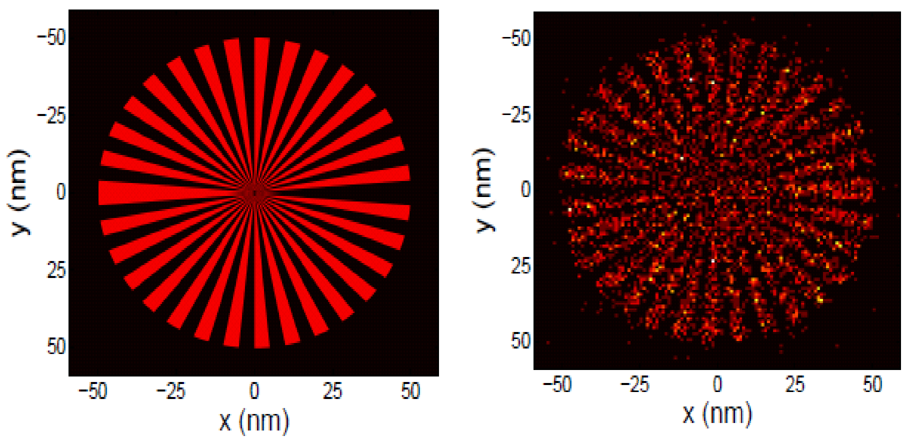


- S: solenoid
- C: condenser lens
- O: objective lens
- I: intermediate lens
- P: projection lens
- D: detector

Formulation with accelerator terminology

- Imaging condition: $R_{12}=R_{34}=0$
- Chromatic aberration: T_{126}
- Spherical aberration: U_{1222}

$$T_{ijk} = \sum_{m=1}^{6} R^{(2)}_{im} T^{(1)}_{mjk} + \sum_{m,n=1}^{6} T^{(2)}_{imn} R^{(1)}_{mj} R^{(1)}_{nk}$$



Sample　　Image

Xiang et al., Nucl. Instrum. Methods A, 759, 74 (2014)

Outline

- Introduction
- keV UED/UEM: why and how
- MeV UED/UEM: why and how
- Challenges of MeV UED/UEM
- UED/UEM center at SJTU
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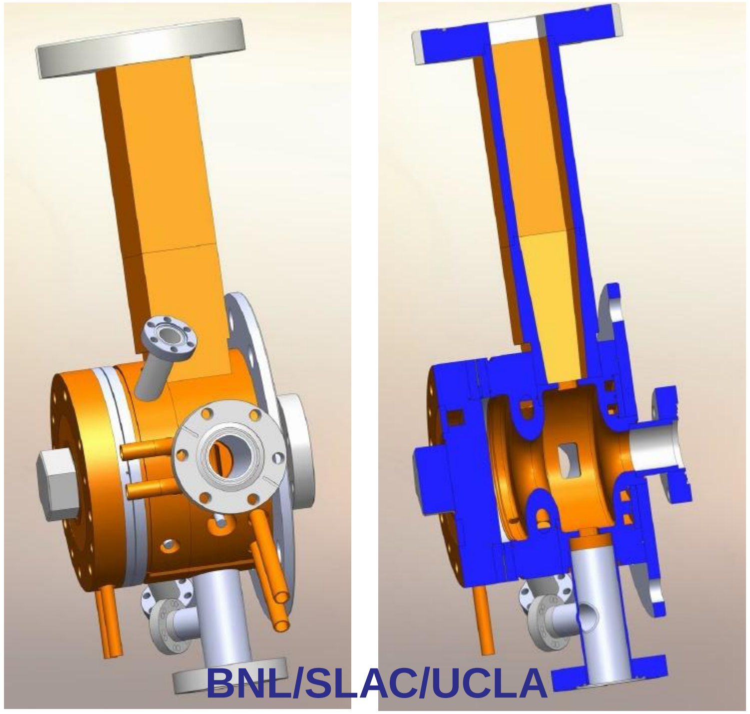
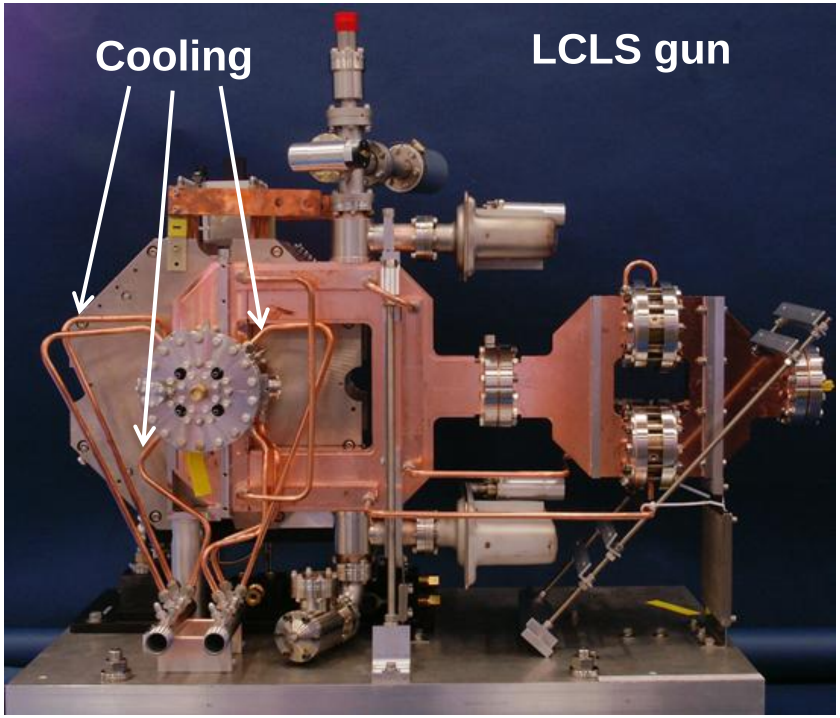
Challenges of MeV UED/UEM

1. Increase rep-rate to 1 kHz and beyond

- 1 kHz pumping laser widely available
- keV UED typically operates at 1 kHz
- Reduce data acquisition time and enable new science





LCLS gun designed for 120 Hz at 120 MV/m; ~500 Hz with 60 MV/m; Roughly a factor of 2 to go.

Challenges of MeV UED/UEM

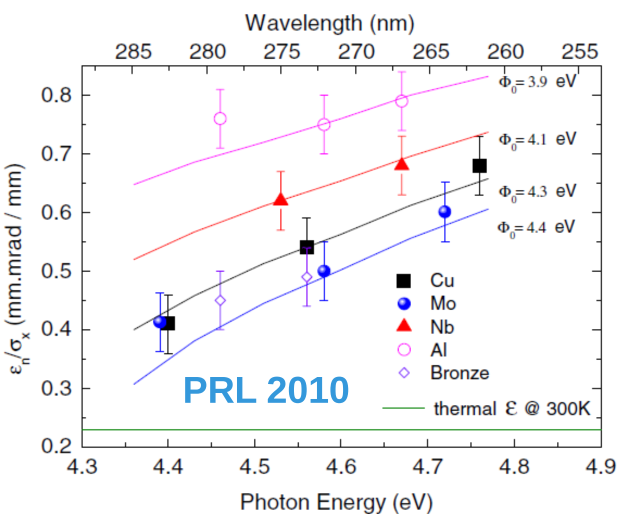
2. Reducing beam emittance to ~10 nm

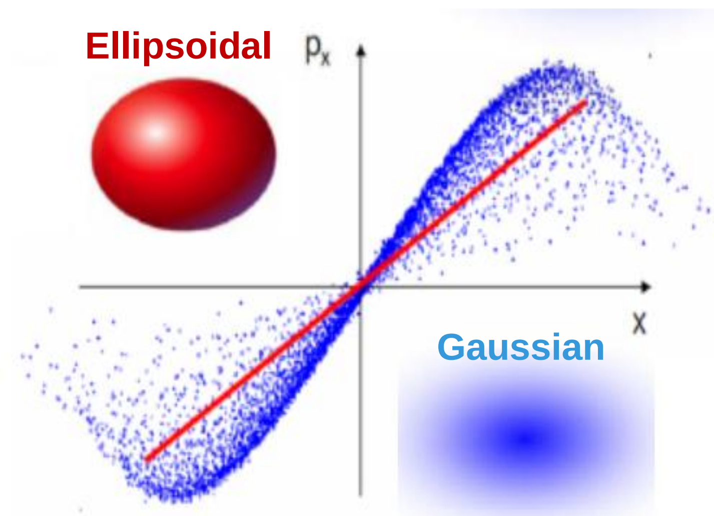
$\triangle r = C_s\theta^3$; $\triangle r = C_c\theta\triangle E/E$; $\varepsilon = \sigma_x\sigma_{x'}$

- Reduce thermal emittance
- Reduce space charge induced emittance growth

$$\varepsilon_n = \sigma_x\sqrt{\frac{2\langle E_{kin,x}\rangle}{m_0c^2}}$$

$$\phi(x,y,z) = \frac{1}{4\pi\varepsilon_0}\int\frac{\rho(x',y',z')dx'dy'dz'}{[(x-x')^2+(y-y')^2+(z-z')^2]^{1/2}}.$$





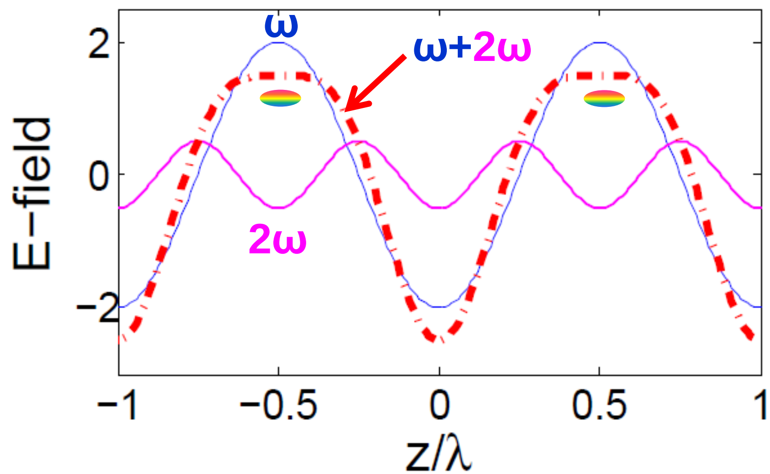
Vary cathode and laser parameters to control the photoelectron distribution

Laser shaping to produce uniform (ellipsoidal) beam

Challenges of MeV UED/UEM

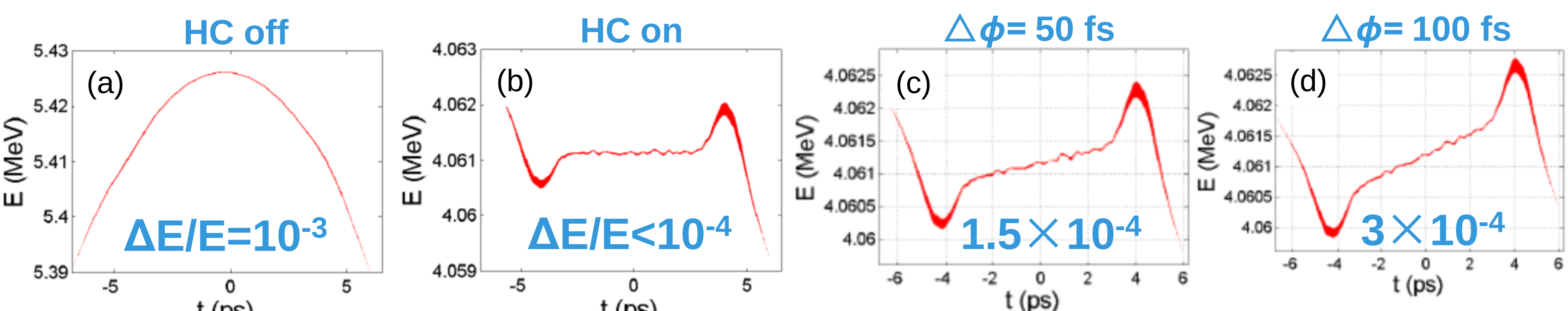
3. Reducing beam energy spread to <10^{-4}

- Remove quadratic energy chirp with a harmonic cavity ($\cos 5^0=0.996$)
- Excellent RF and Low-level RF system



$$E(z) = E_i + E_s\cos(\phi_s + k_s z) + E_x\cos(\phi_x + k_x z)$$

$$\frac{\Delta E(z)}{E_0} = -\frac{E_s k_s \sin\phi_s}{E_0} z + \frac{1}{2}\frac{E_x k_x^2 - E_s k_s^2 \cos\phi_s}{E_0} z^2 + \Theta(z^3) + \ldots$$

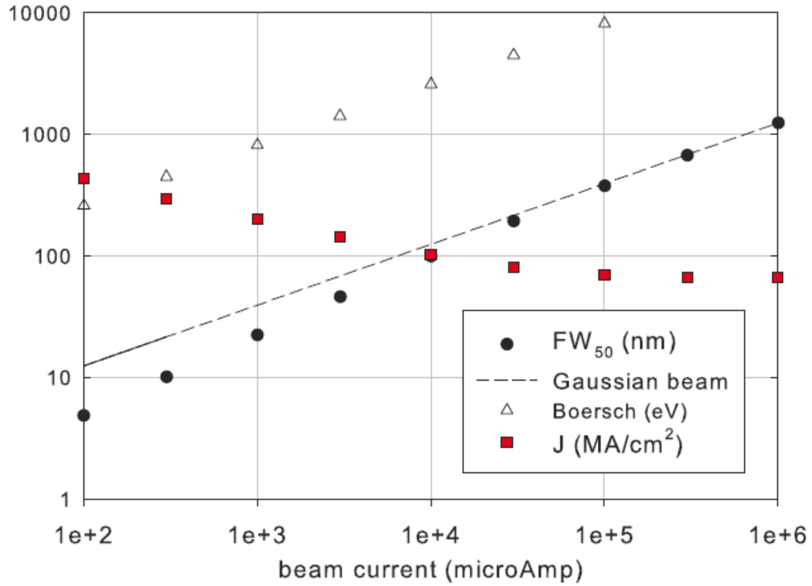


Hemsing, Stupakov, Xiang, Zholents, Rev. Mod. Phys. 86, 897(2014)

Challenges of MeV UED/UEM

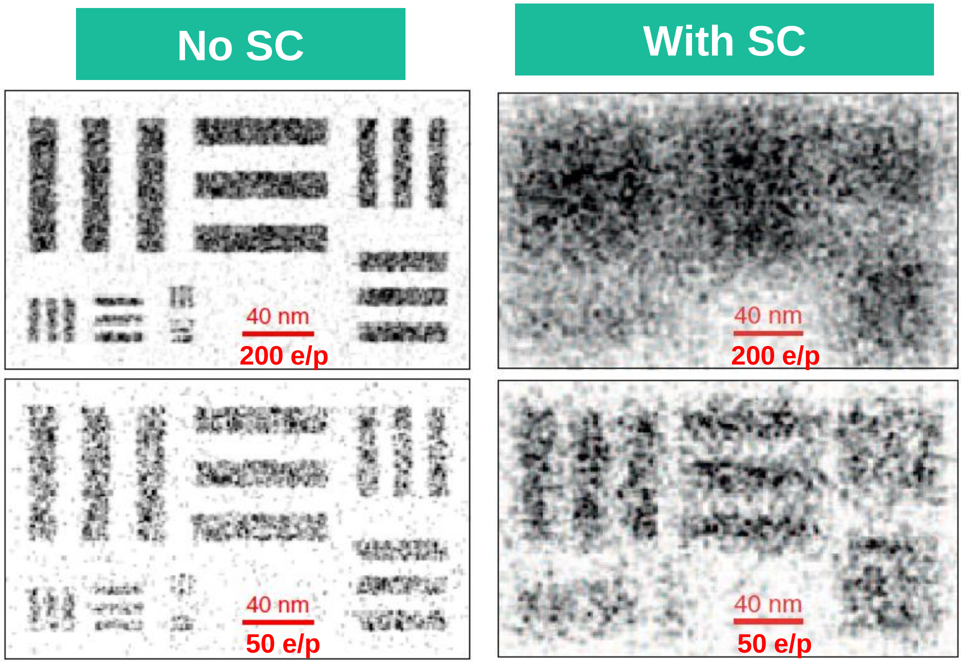
4. High beam density and space charge effect

- Rose theorem: >100 electrons/pixel to make a useful image
- Ultimate beam density (e^-/nm^3) limited by space charge (SC) effect
- Spatial resolution also limited by stochastic space charge effect



FW_{50}~$I^{1/2}$; J_{max}=50 MA/cm² for 2.5 MeV

Handbook of charged particle optics



Li and Musumeci, PRA, 2, 024003 (2014)

10 nm & 10 ps seems to be achievable; extremely challenging to push spatial resolution to ~1 nm while keeping ps temporal resolution.

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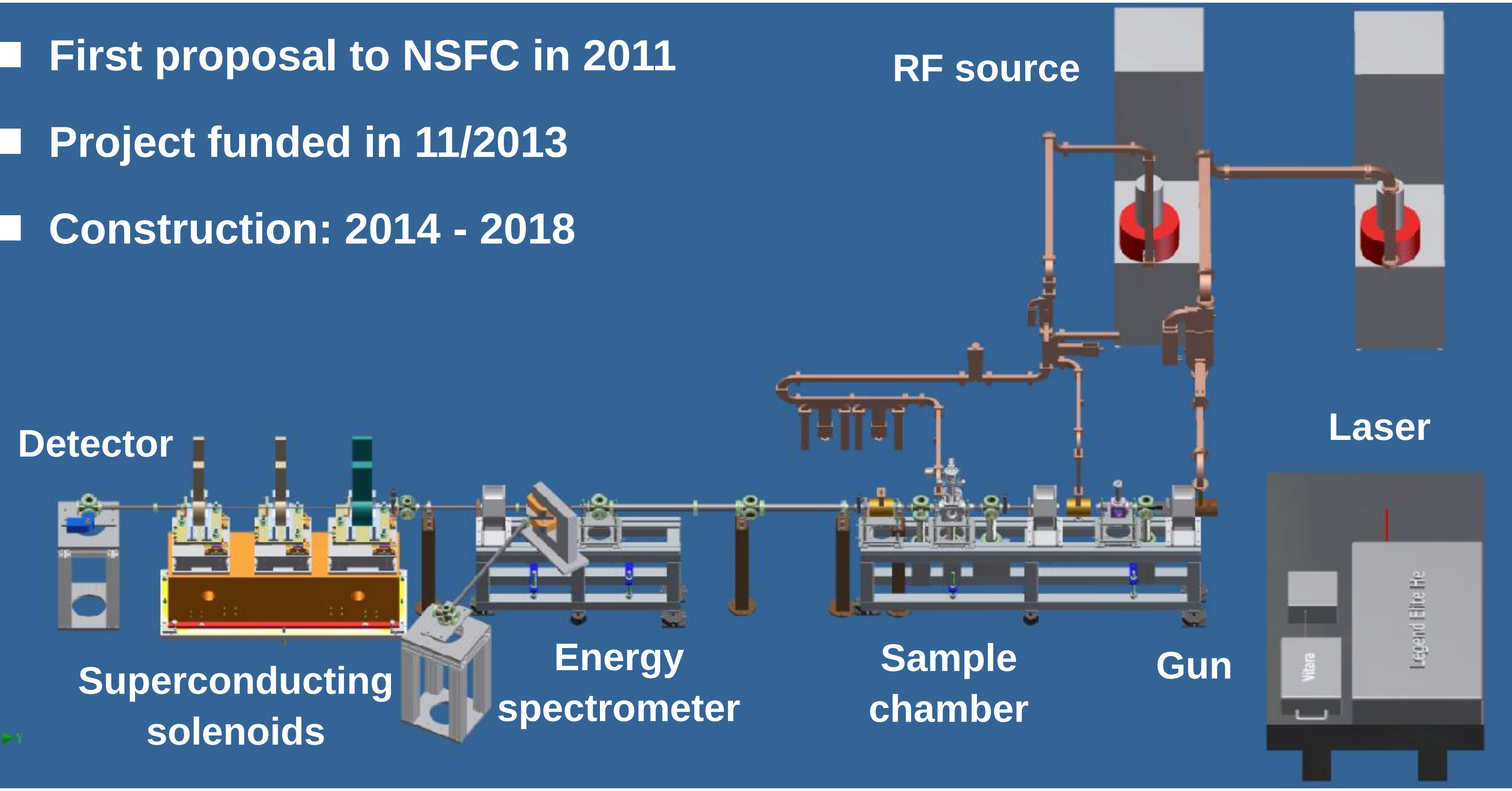
MeV UED/UEM center at SJTU

- First proposal to NSFC in 2011
- Project funded in 11/2013
- Construction: 2014 - 2018



Mode	Diffraction	Microscopy	CDI	Rep-rate
State-of-the-art	~200 fs	10 ns/10 nm	\	10~100
SJTU Goal	~50 fs	10 ps/10 nm	1 ps / 1 nm	~1000

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Construction of a new experimental hall

06/2014

08/2014

09/2014

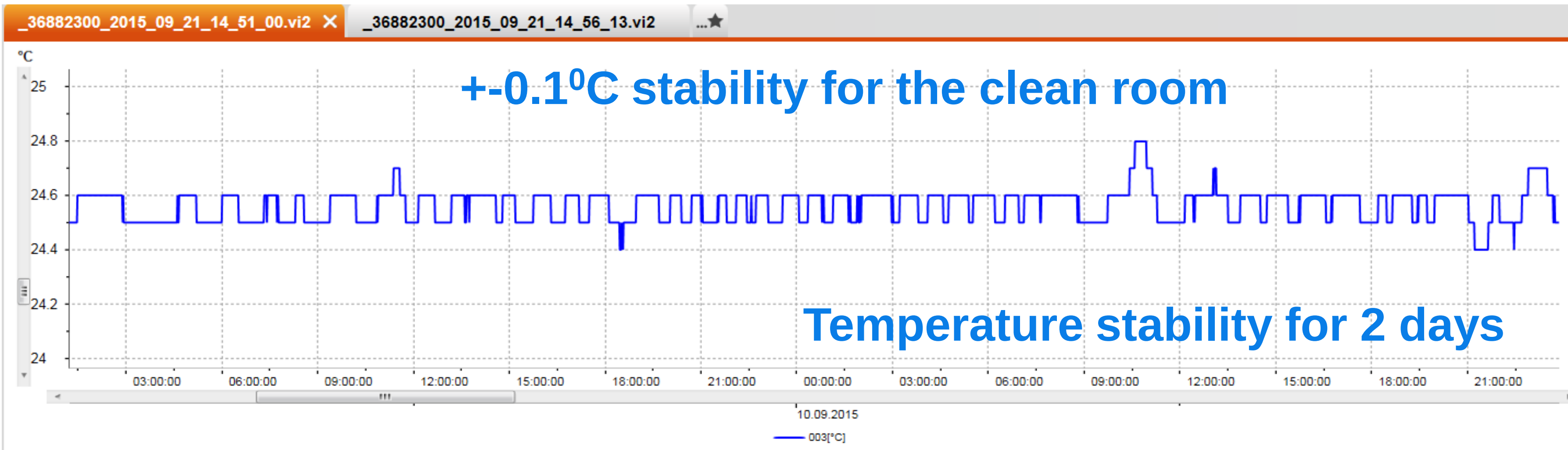
10/2014

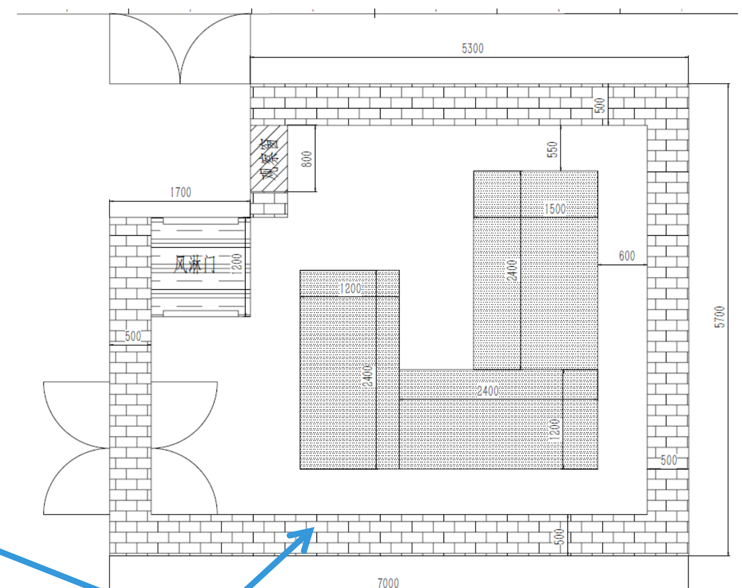
11/2014

12/2014

Construction of a new experimental hall

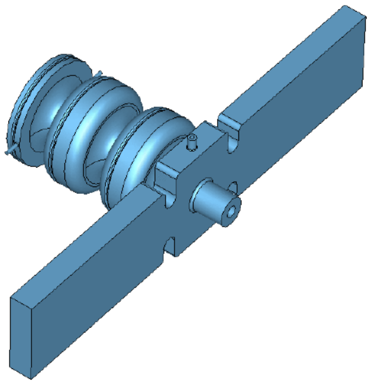
Construction of a new experimental hall

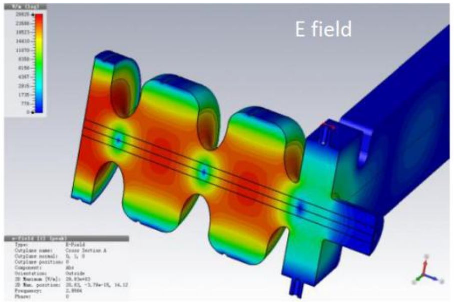


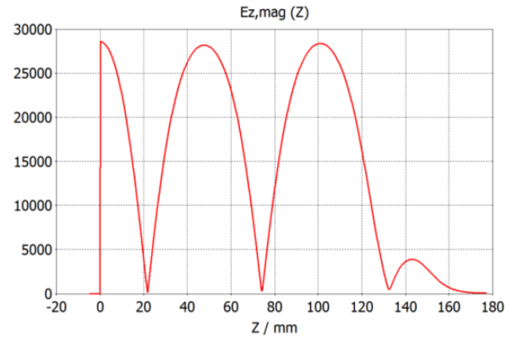


Buffer region (+-1^{0}C)

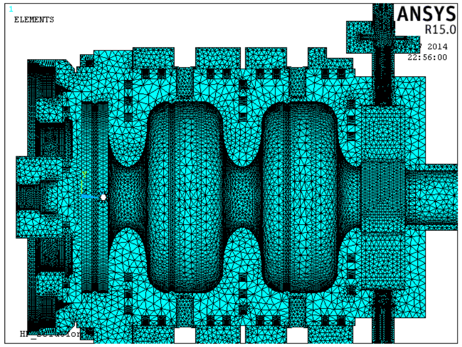
Development of 1 kHz photocathode rf gun

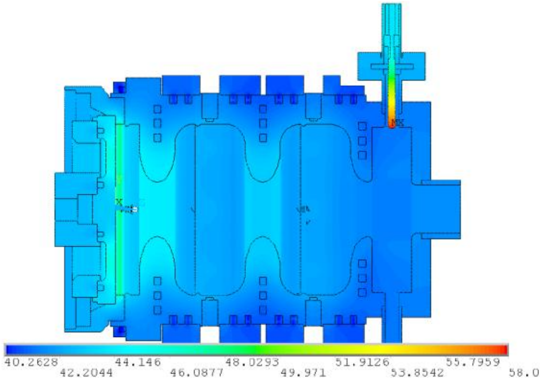




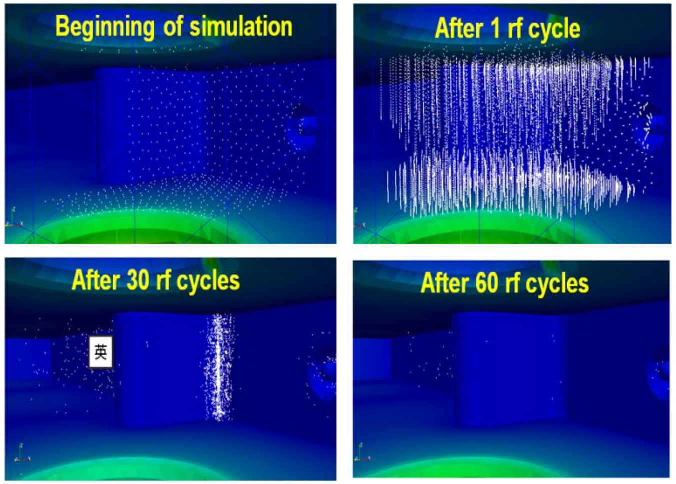


Field distribution





- 2.4 cell gun
- 9 Cooling channels
- Max temperature rise 18^{0}C
- Multipacting OK

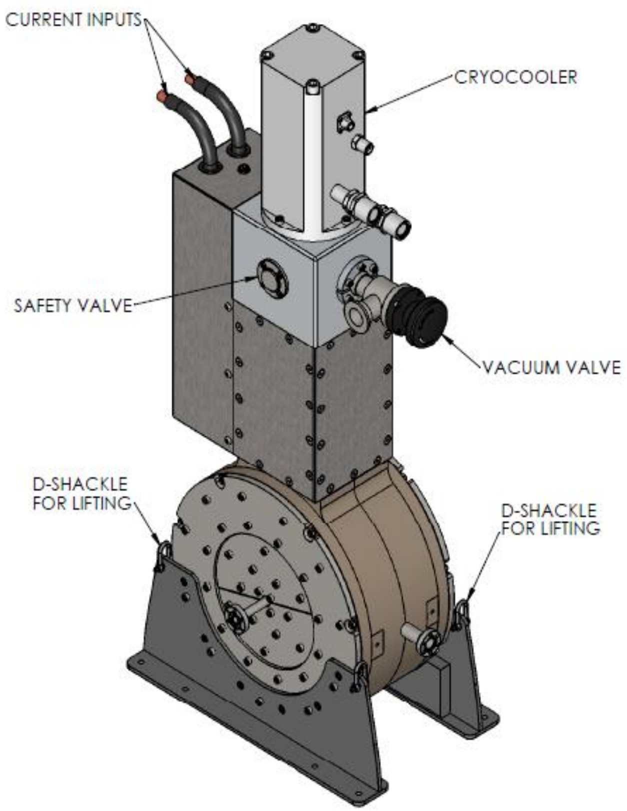


SJTU/SINAP/ANL/LBNL Collaboration

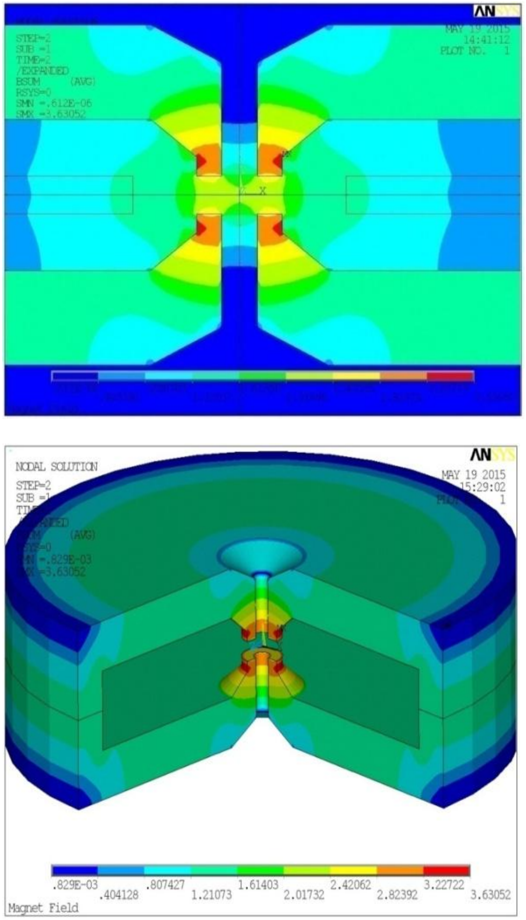
Development of high-field solenoid lens

$C_c \sim C_s \sim f$

$$\frac{1}{f} = \frac{e^2}{4\gamma^2 m^2 v_z^2} \int B_z^2 \mathrm{d}z$$

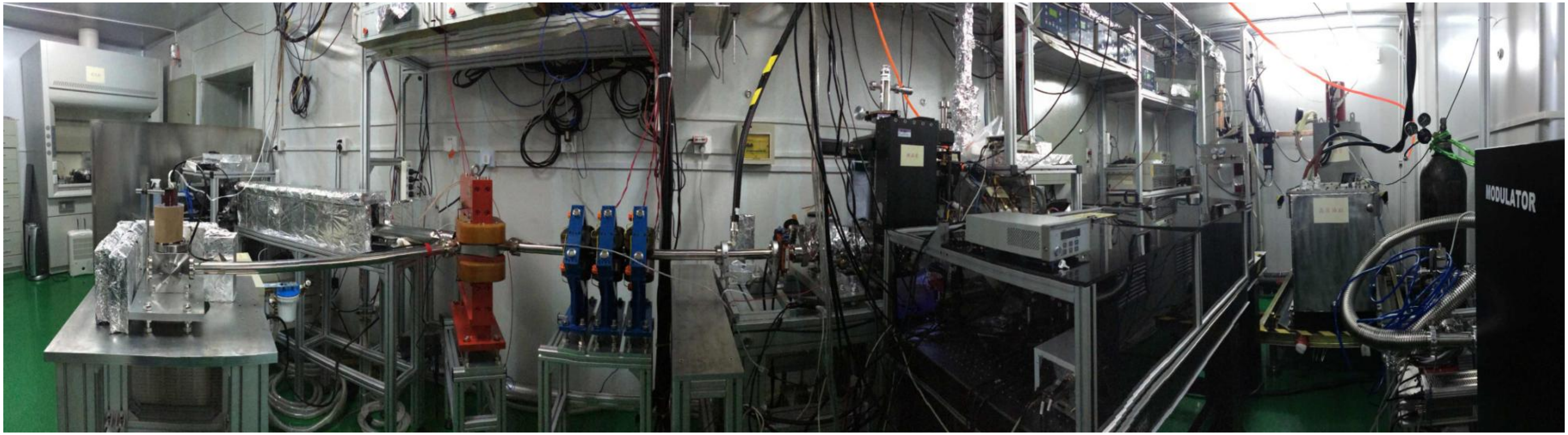


Schematic of the HT_c superconducting lens

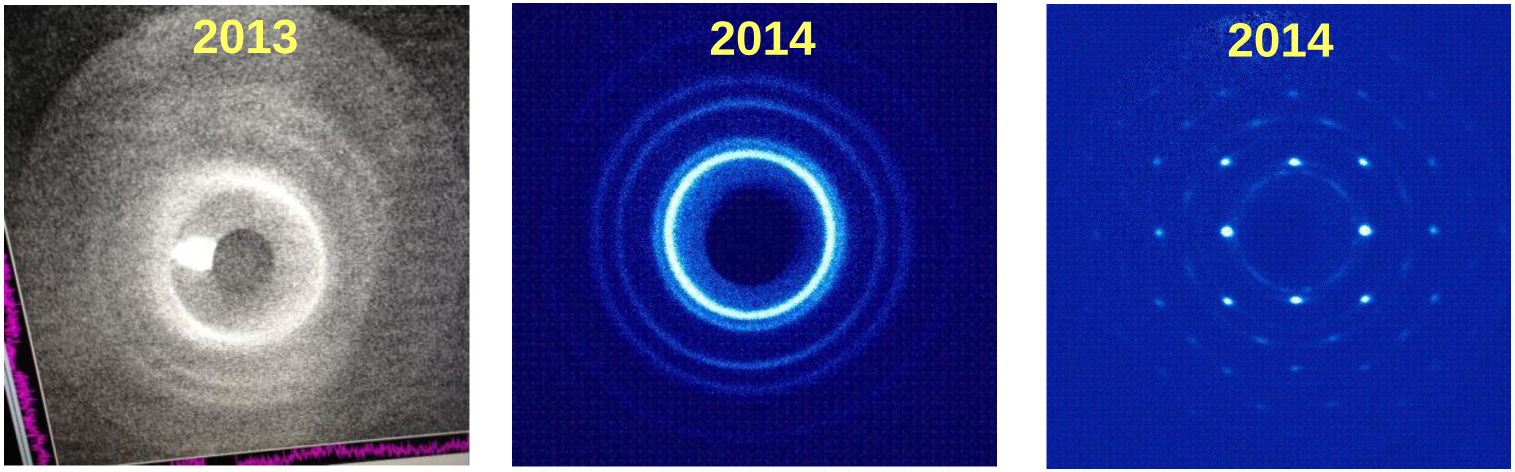


HTS lens by IHEP

Commissioning the test facility

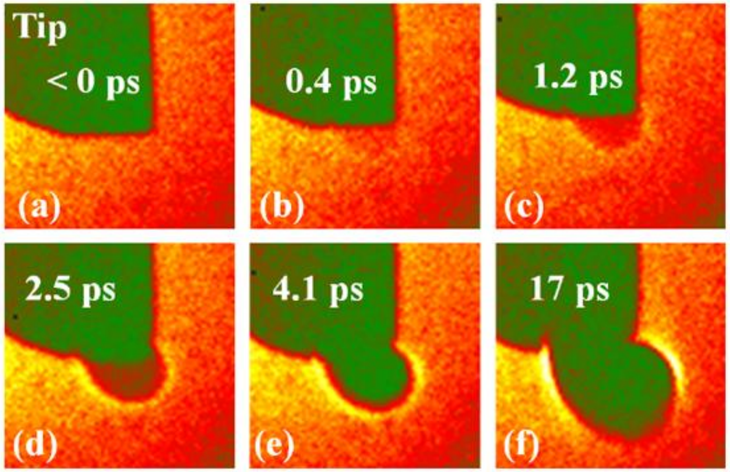


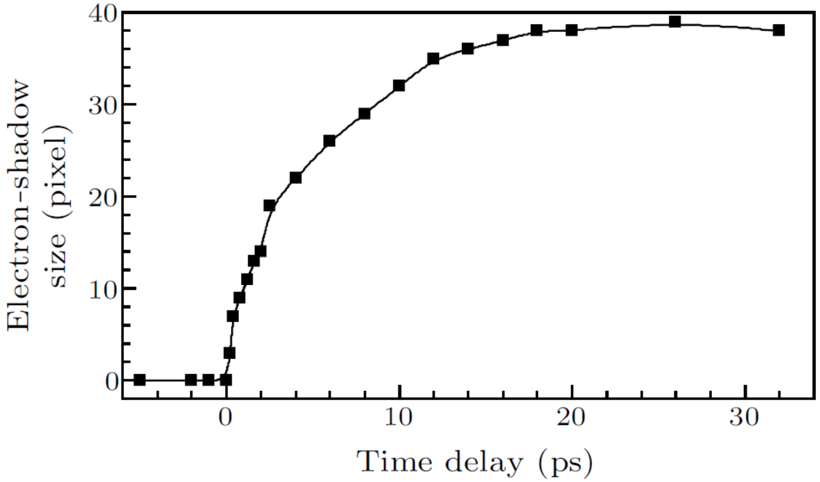
Photocathode rf gun based MeV UED test facility



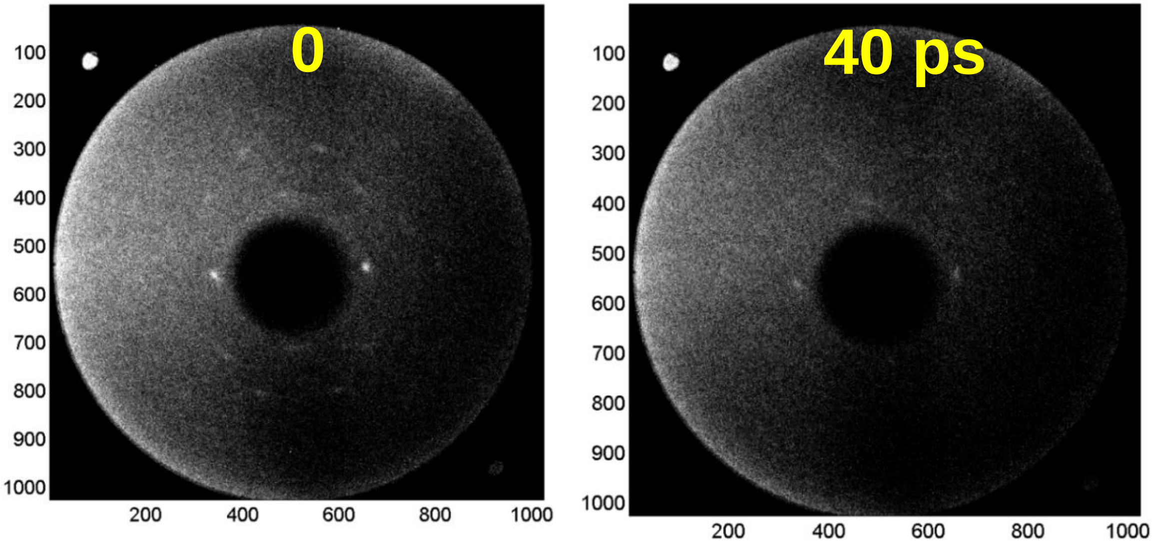
High quality Al and Au diffraction pattern (Fu et al., Rev. Sci. Instru. 2014)

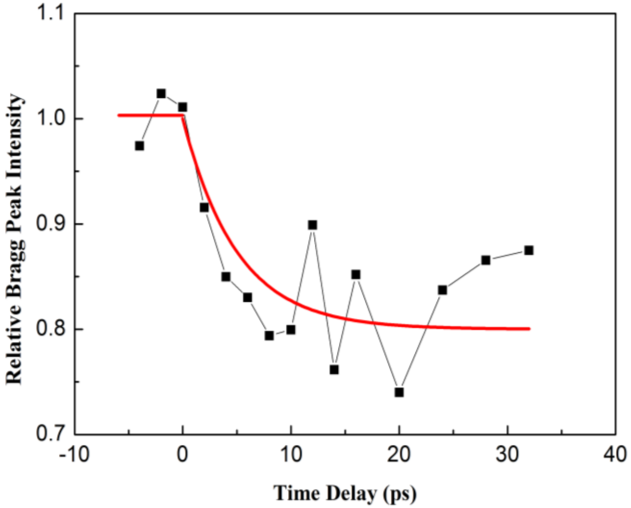
Commissioning the test facility





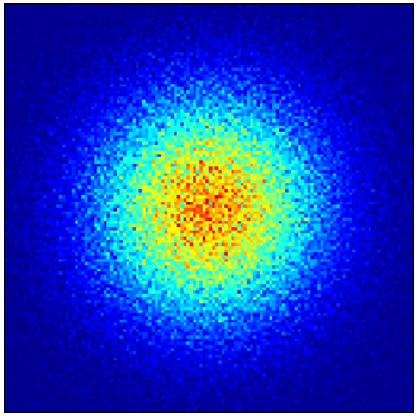
Finding time-zero with the perturbation from laser-induced plasma



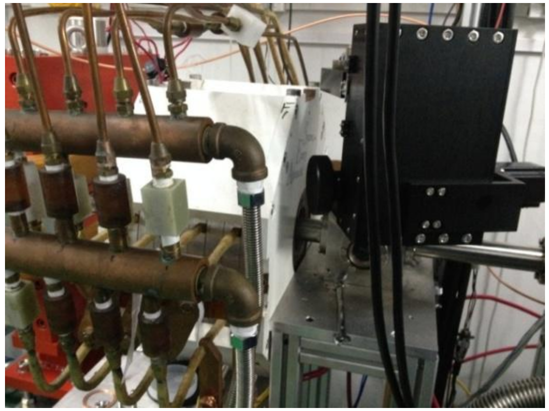


MeV UED pump-probe experiment (Zhu et al., CPL, 2014)

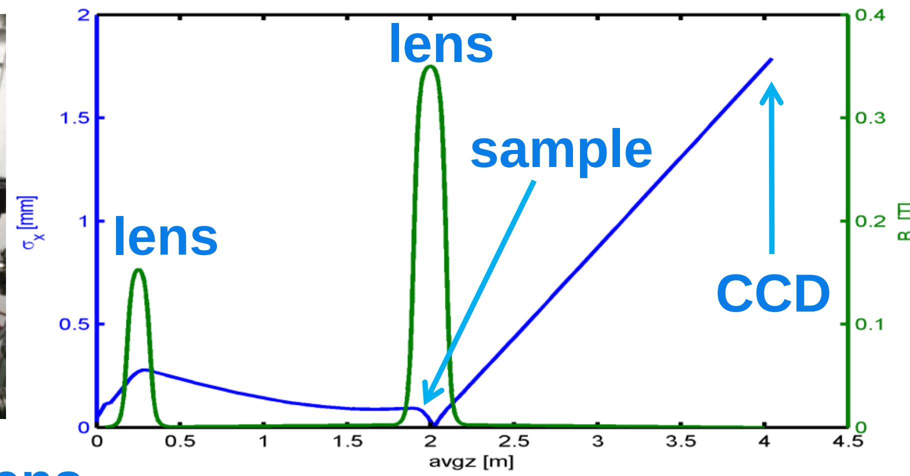
Commissioning the test facility

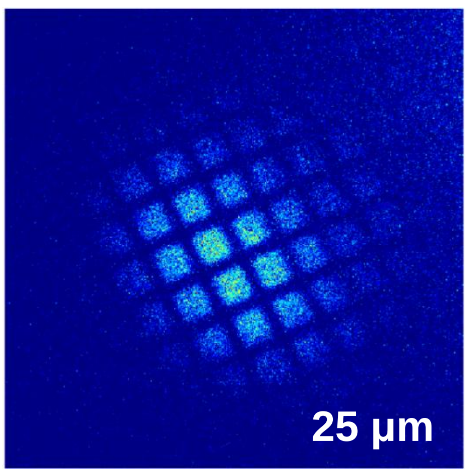


Simulation

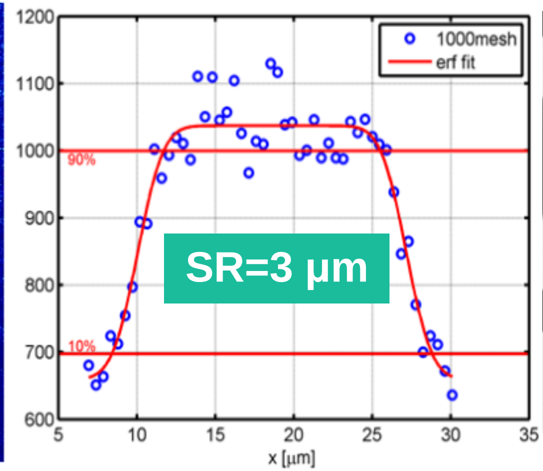


Condenser-objective lens

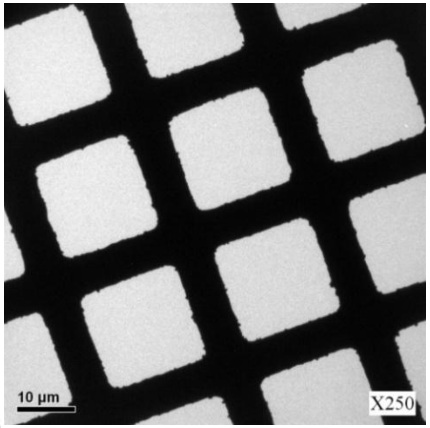




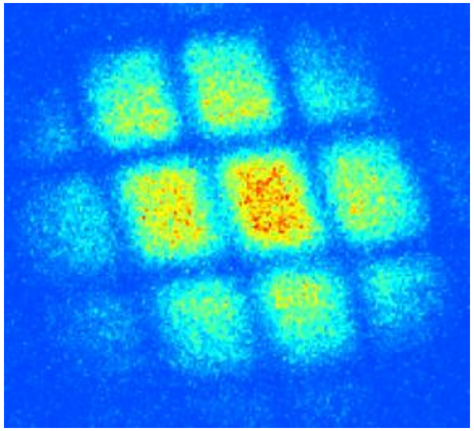
MeV UEM



Resolution



keV TEM

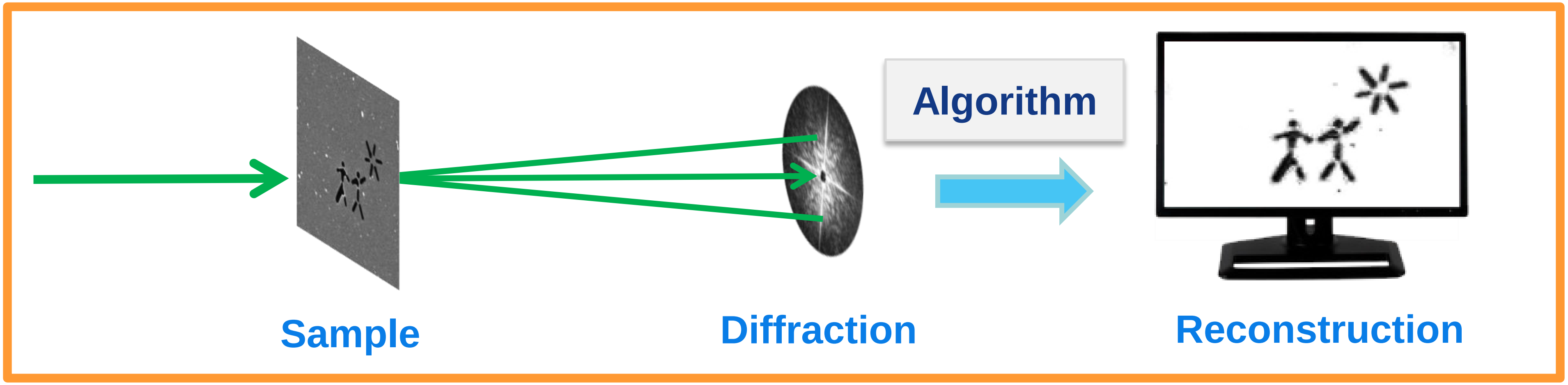


Space charge

Temporal-spatial resolution (300 fs*3μm=10^{-18} s*m);

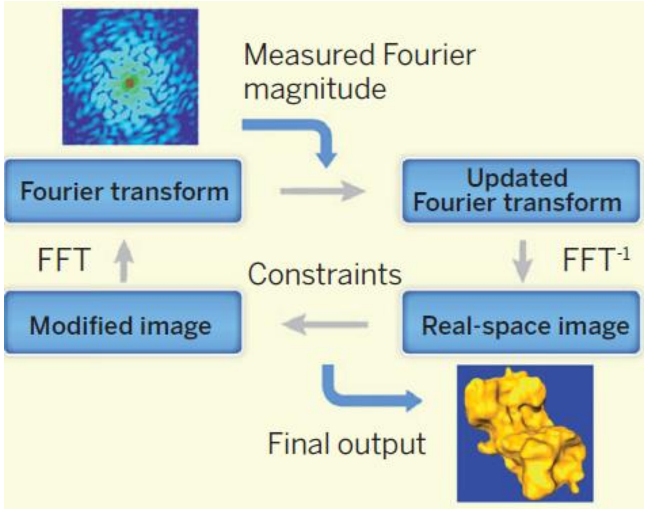
Shoot for 10^{-19} s*m with our user facility

Coherent diffraction imaging



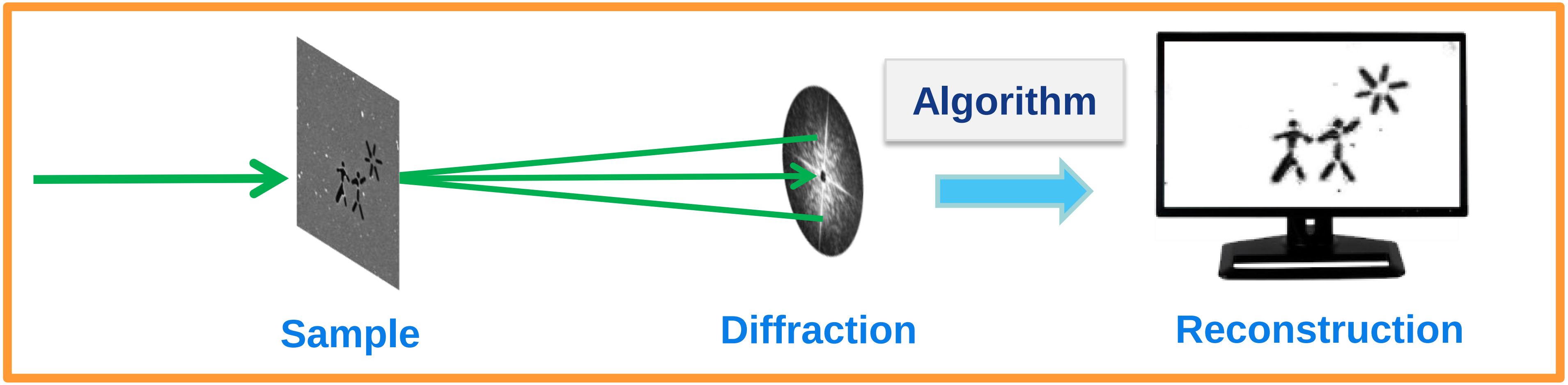
Schematic layout of coherent diffraction imaging

- Start with a random phase set;
- Combine this random phase set with the measured Fourier magnitude;
- Apply an inverse FFT to get an initial image;
- Apply constraints to get an updated image;
- Apply FFT and replacing its magnitude with the measured data;
- Repeat.

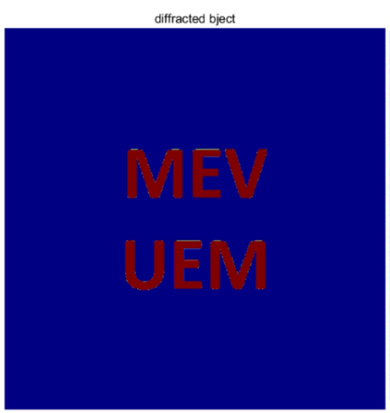


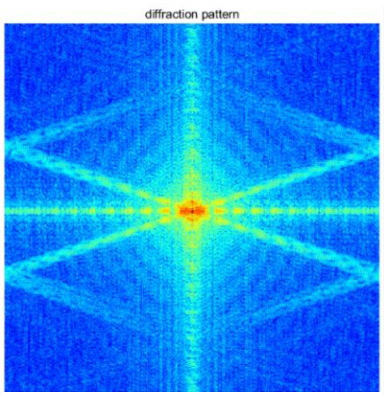
Algorithm

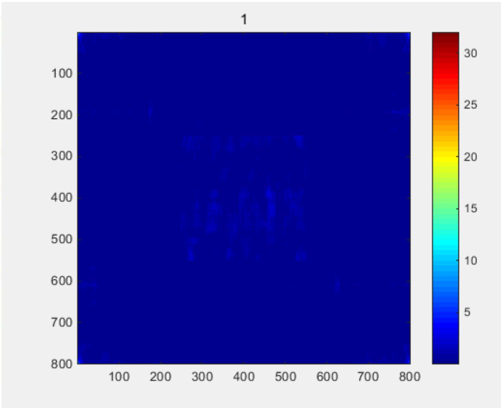
Coherent diffraction imaging

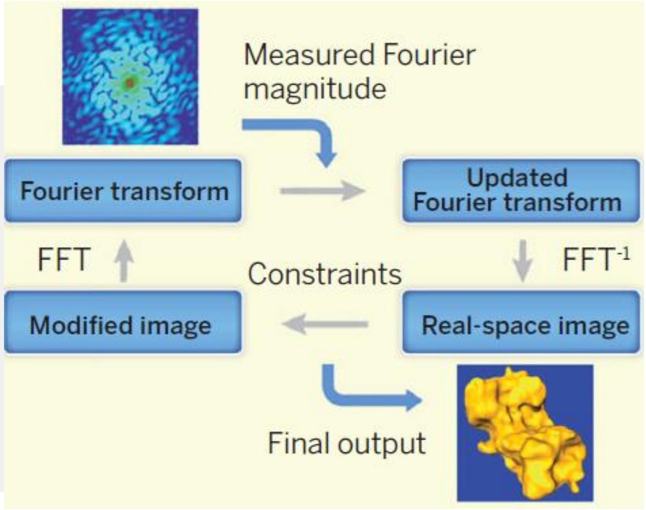


Schematic layout of coherent diffraction imaging



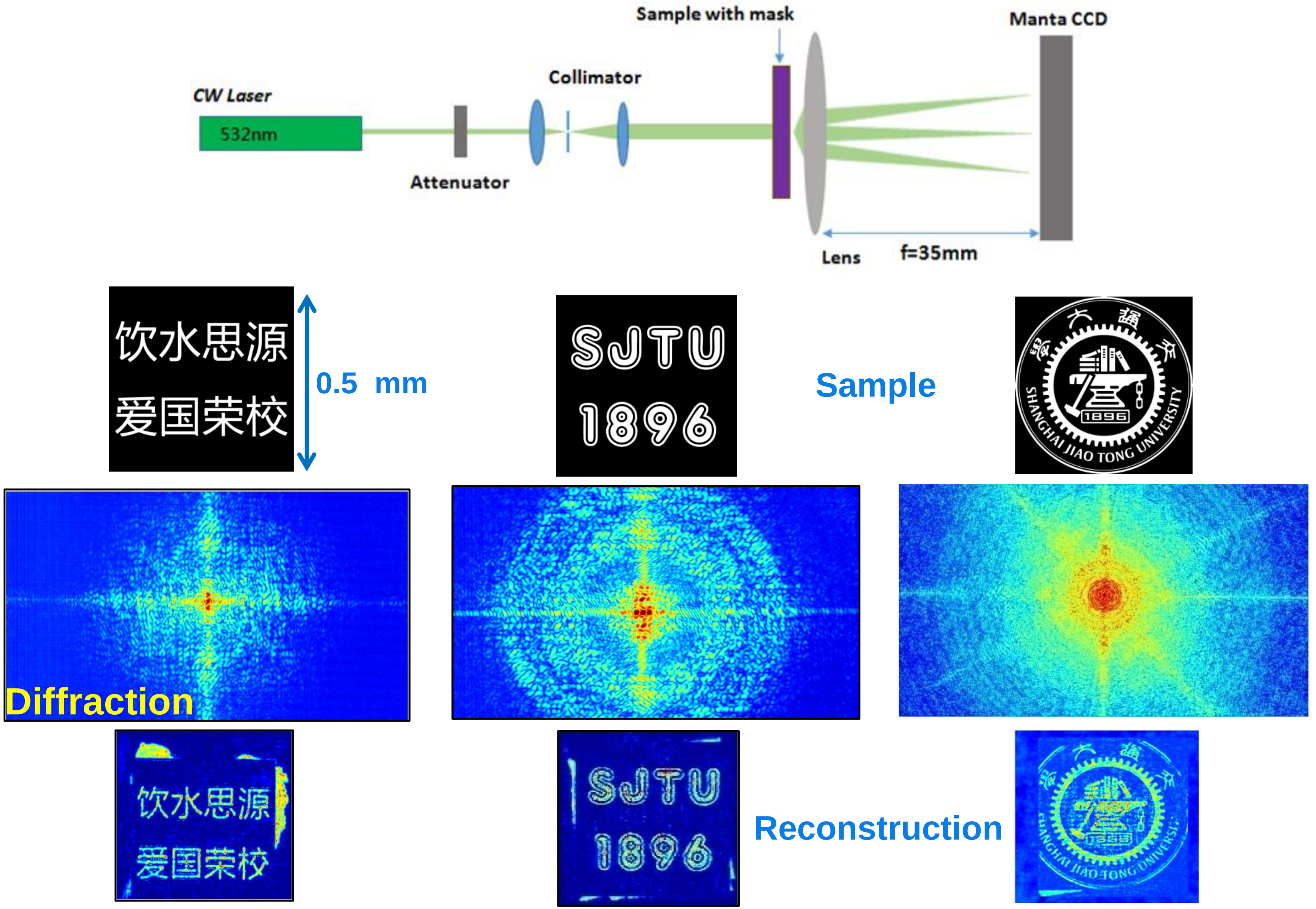






Sample | Diffraction | Reconstruction | Algorithm

Coherent diffraction imaging



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Summary

- Historically particle accelerators are instrumental for high energy physics and photon sciences.
- Accelerator based MeV UED/UEM hold great potential in solving the challenges in probing matter at ultrafast temporal and ultrasmall spatial scales.
- Potentially better performance than keV UED/UEM.
- Compact facility, yet with rich physics and grand challenges.
- Several MeV UED user facilities are being built.
- Initial results are very encouraging.
- The fun just begins!


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Thanks!
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