MeV Electron Diffraction and Microscopy

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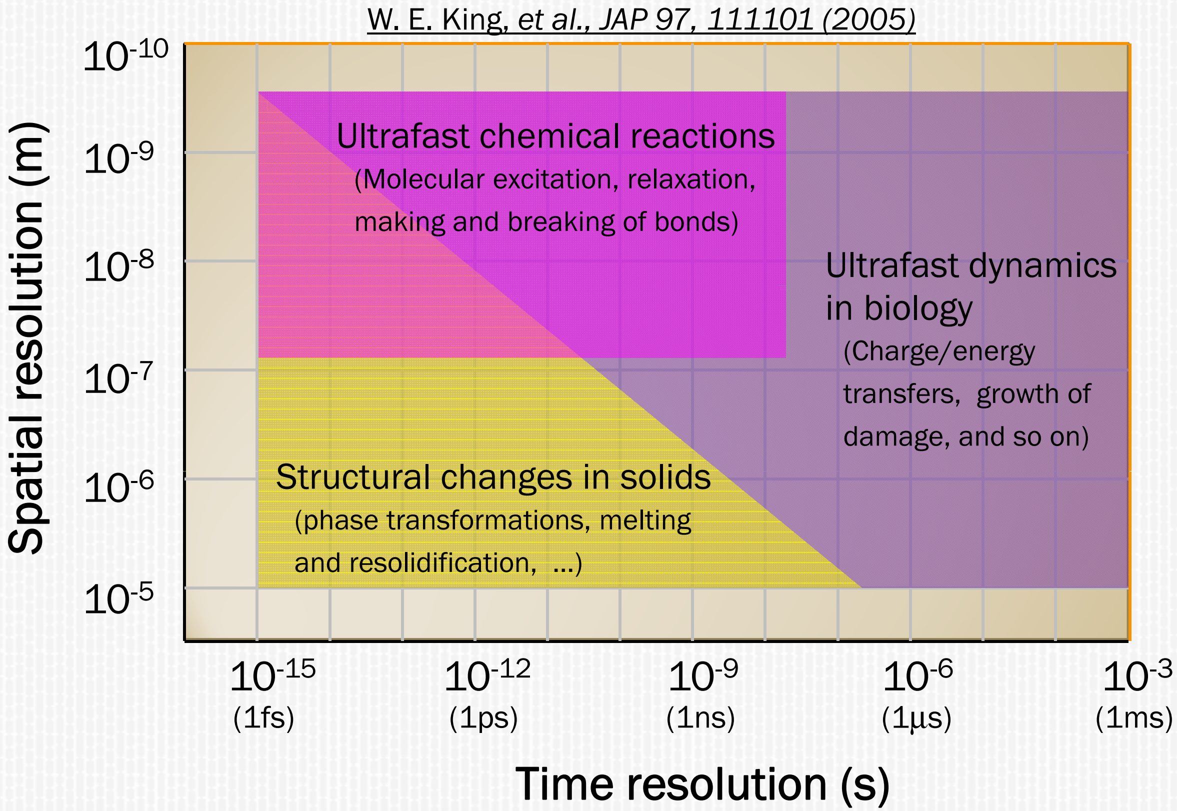
Structural dynamics in matter

✓Ultrafast phenomena or dynamics in materials are occurred on femtosecond time scales over atomic spatial dimensions.



✓An ultrafast measurement with the resolution of 100 fs & sub-Angstrom has long been in goal for the scientists.

Ultrafast detection techniques
for study of ultrafast phenomena and dynamics

1) Ultrafast X-ray diffraction/image

Picosecond/femtosecond X-ray pulses generated from SR, FEL or laser plasmas acceleration have been used. A powerful tool!

→ big experiment/more expensive.

2) Ultrafast electron diffraction (UED)

In UED, a fs laser pulse is used as pump, while a fs or ps e^- bunch is used as probe. Recently, the time resolution has been achieved to 100 fs using DC or RF electron sources, Very compact!

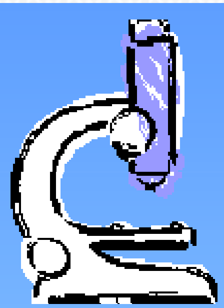
but no spatial resolution!

3) Ultrafast electron microscopy (UEM)

UEM can observe the dynamics of structural transformation in nanometer (even atomic) spatial dimensions.

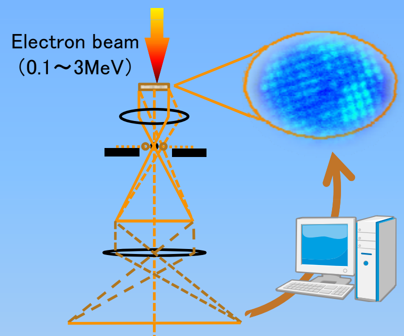
⇒ In UEM, the resolution of has achieved to 10 ns and 10 nm with single-shot measurement.

UEM and its applications



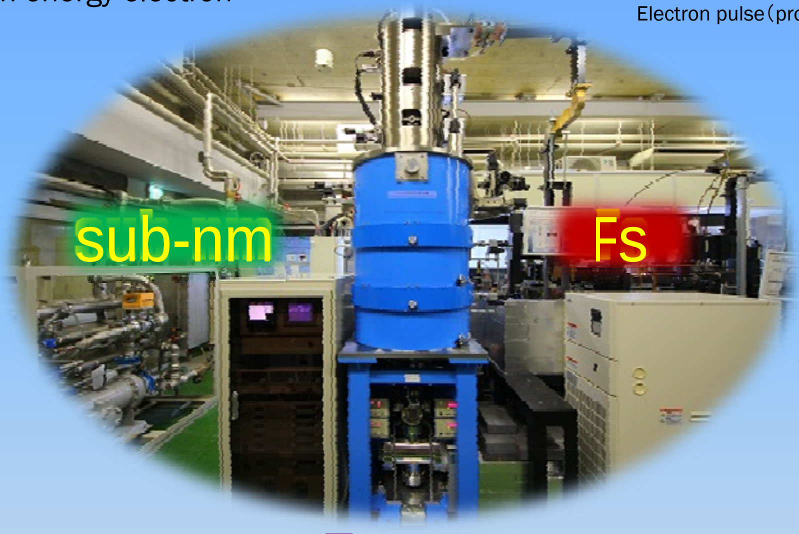
Imaging Technology
イメージング　テクノロジー

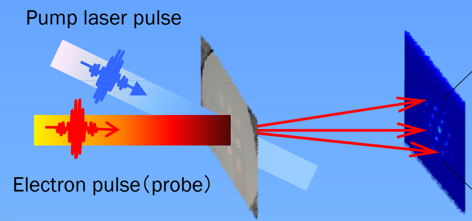
Structural observation and imaging in "real space" with atomic-scale spatial resolution using high-energy electron beam.

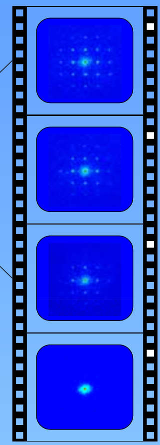


Imaging in "real space"

Methods







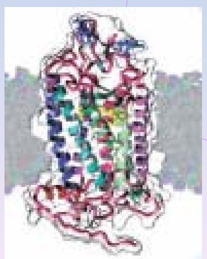
Ultrafast Observation
超高速現象の観察

Observations of fundamental dynamic processes in matter occurring on femto-second time scales over atomic spatial dimensions.

Observation in "real time"

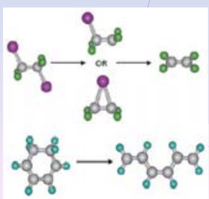
Targets

Protein Structural Dynamics
タンパク質構造ダイナミクス



- Protein structural dynamics
- Macromolecular structure
- Reveal of functioning processes
- New technologies and applications in medical biology.

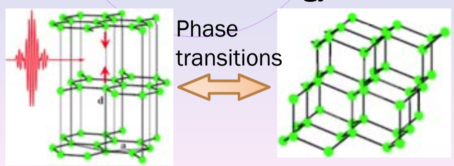
Making Molecular Movie
分子運動の可視化：ー新しい科学ー



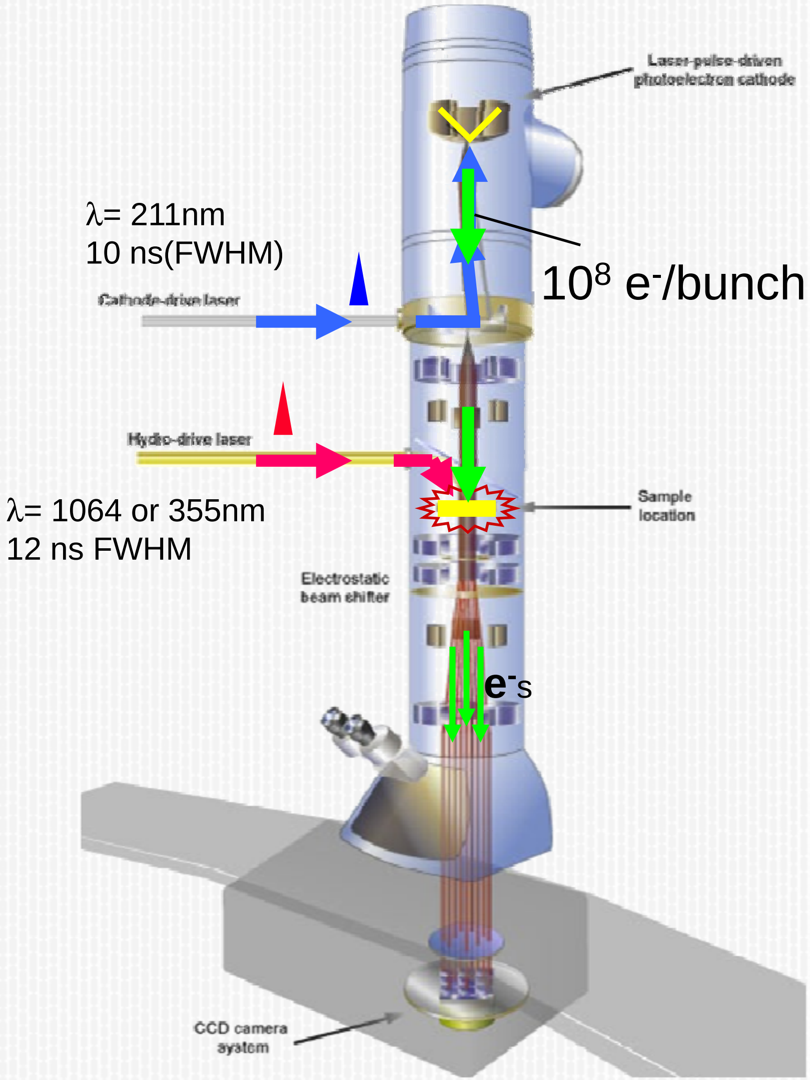
- Observation of single molecule motion.
- Ultrafast chemical reactions
- Solvation dynamics
- Discovery of transition states and reaction intermediates.

Nano-technology/science
ナノテクノロジー・サイエンス

- Transformation dynamics of novel nano-scale materials.
- Creation of new functional materials and devices for nanotechnology.

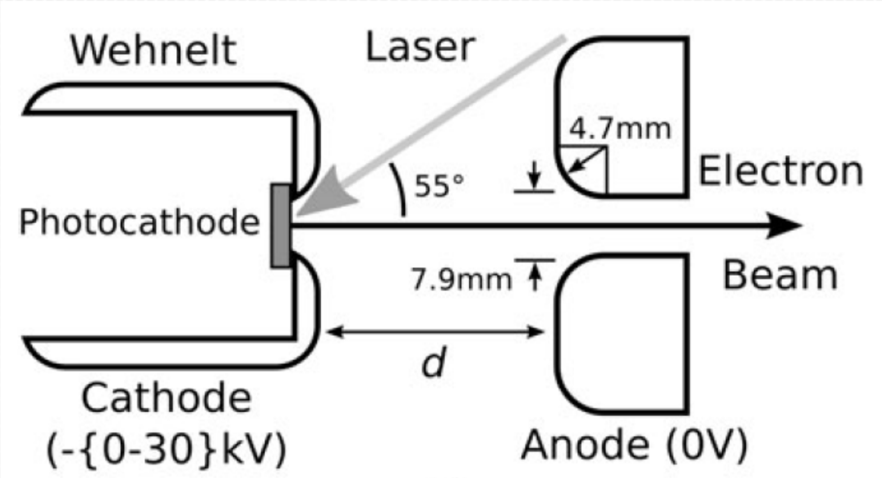


Recent UEMs



B. W. Reed, Workshop on UESDM, 2012

photocathode DC gun in UEMs

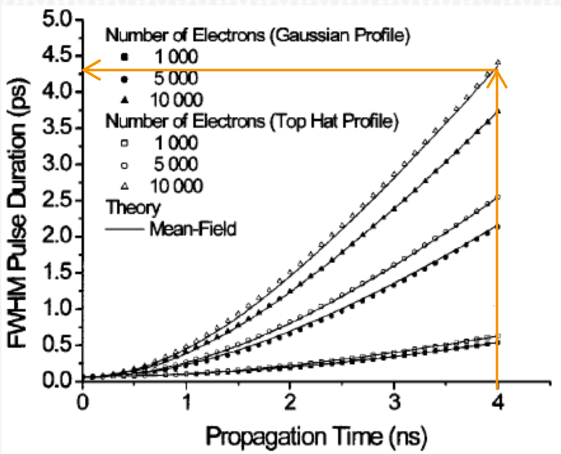


- keV-energy electron pulses with ~ns & 10^8 e^-s/pulse for single-pulse imaging.
- keV-energy single-electron pulses for stroboscopic measurement.

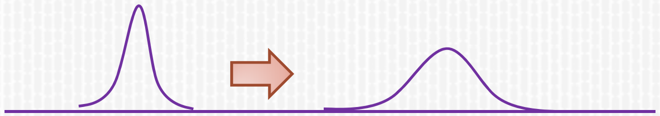
UEM is a powerful technique
for study of structural dynamics!

Space-charge problem in recent UEM

1) Increase of bunch length during beam transport



B. J. Siwick et al., JAP 92, 1643(2002)



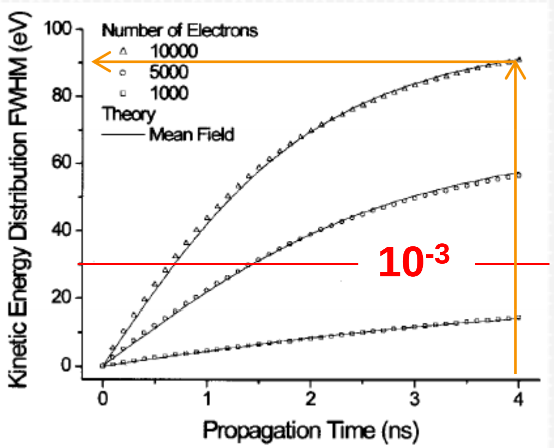
Bunch length increase due to space-charge force

$$\Delta t \approx \Delta t_0 + \frac{Ne^2 t_2}{2\pi r^2 \varepsilon_0 mv}$$

***Δt*=300fs → 4ps during the 40cm transport of 30keV e⁻ beam**

It is difficult to generate a high-intensity and fs-pulse electron beam in keV energy region!

2) Increase of energy spread during beam transport



$$\Delta E_k \approx m v_0 \Delta v = m v_0 \frac{dl}{dt}$$

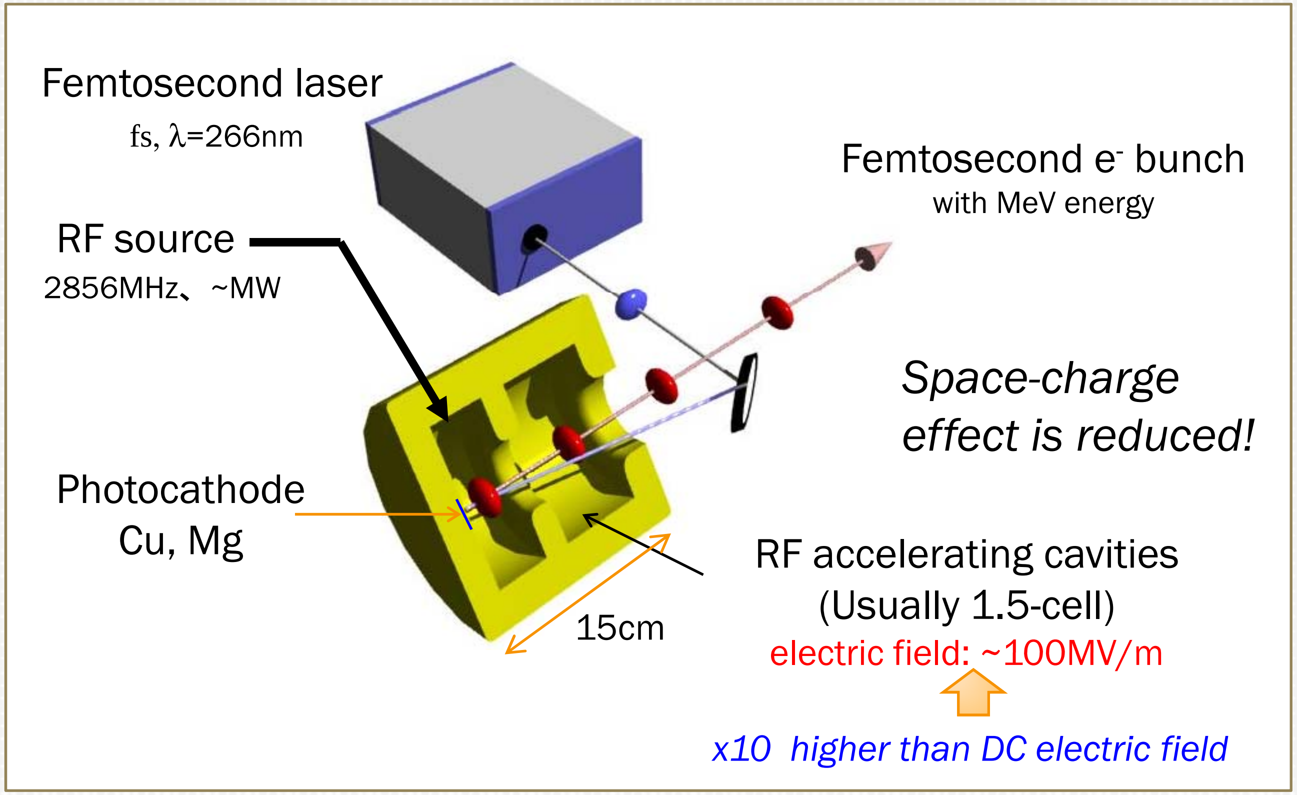
***ΔE/E* → 3×10^{-3} during the beam transport**

⇒ One solution of the space-charge problem is to increase the beam energy!

↓

a choice to use a RF MeV electron gun in UEM

What radio-frequency (RF) electron gun?



The expected beam parameters:

Electron energy:	1~3 MeV
Bunch length:	100 fs
Emittance:	0.1 mm-mrad
Energy spread:	10^{-4} (10^{-5} for challenge)
Charge:	10^{7}~10^{8}e⁻'s/pulse

Key parameters for EM! (Emittance, Energy spread)

How to generate such beam?

Beam dynamics in RF gun

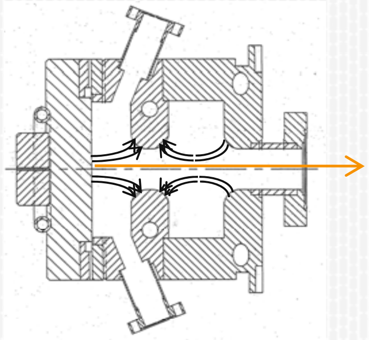
1) Longitudinal dynamics

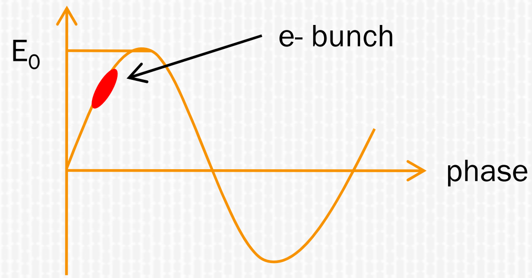
RF field in longitudinal direction:

$$E_z = E_0 \cos kz \sin(\omega t + \phi_0)$$

$$\phi = \omega t - kz - \phi_0 = k\int_0^z \left(\frac{\gamma}{\sqrt{\gamma^2 - 1}} - 1 \right) dz + \phi_0$$

$$\frac{d\gamma}{dz} = \frac{eE_0}{2mc^2}\left[\sin\phi + \sin(\phi + 2kz)\right]$$





Kim, NIM A275, 201-218(1989)
Travier, NIM A340, 26-39(1994)

$$\gamma = 1 + (n + 0.5)\alpha\pi = 1 + 146.8(n + 0.5)\frac{E_0[MV/m]}{f[MHz]}$$

$$\sigma_{\Delta\gamma}(rms) = 2\pi\alpha f \sigma_z = 2.9\times10^{-4} E_0[MV/m]\sigma_z[ps]$$

$$\left.\frac{\Delta E}{E}\right|_{RF}(rms) = \frac{\sigma_{\Delta\gamma}}{\gamma - 1} = 2\times10^{-6}\frac{f[MHz]\sigma_z[ps]}{n + 0.5}$$

$$\alpha = \frac{eE_0}{2mc^2 k} = 46.7\frac{E_0[MV/m]}{f[MHz]}$$

Example:

E_0=25~100MV/m, f=2856MHz, 1.5-cell

Energy: 1~4 MeV

- using 100fs laser,
  $\Delta E/E \sim 10^{-4}$
- using 10fs laser,
  $\Delta E/E \sim 10^{-5}$

Beam dynamics in RF gun

2) Transverse dynamics

Emittance due to space-charge effect:

$$\varepsilon_{x,z}^{sc} = \frac{\pi}{4}\frac{1}{\alpha k}\frac{1}{\sin\phi_0}\frac{I_p}{I_A}\mu_{x,z}$$

Gaussian distribution beam

$$\mu_x = \sqrt{\langle \Gamma_x^2 \rangle \langle x^2 \rangle - \langle \Gamma_x x \rangle^2} = \frac{1}{3\sigma_x/\sigma_z + 5}$$

$$\varepsilon_x^{sc}[mm-mrad] = 3.76\times 10^3 \frac{Q[nC]}{E_0[MV/m](2\sigma_x[mm]+\sigma_z[ps])}$$

Emittance due to RF effect:

$$\varepsilon_x^{rf} = \sqrt{\langle p_x^2 \rangle \langle x^2 \rangle - \langle p_x x \rangle^2} = \alpha k \langle x^2 \rangle \sqrt{\langle \sin^2\phi_f \rangle - \langle \sin\phi_f \rangle^2}$$

$$\phi_f \to \langle \phi_f \rangle + \Delta\phi, \quad \langle \phi_f \rangle = 90^o$$

$$\varepsilon_x^{rf} = \alpha k^3 \frac{\sigma_x^2 \sigma_z^2}{\sqrt{2}}$$

$$\varepsilon_x^{rf} = 2.73\times 10^{-11} E_0 f^2 \sigma_x^2 \sigma_z^2$$

ε_x^{rf} : $mm \cdot mrad$

E_0 : MV/m, f : MHz

σ_x : mm, σ_z : ps

Example:

E_0=100MV/m, f=2856MHz, Q=1pC,

σ_x=0.1mm, σ_z=100fs

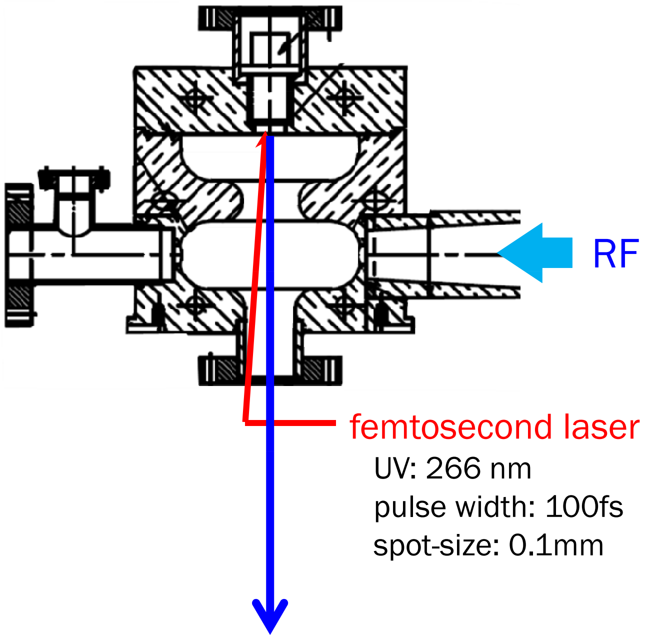
⇩

ε^{sc} ~ 0.1 mm-mrad

ε^{rf} ~ negligible

The emittance is dominated by thermal emittance for low-charge e- beam!

Generation of high-brightness e$^-$ beam with RF gun

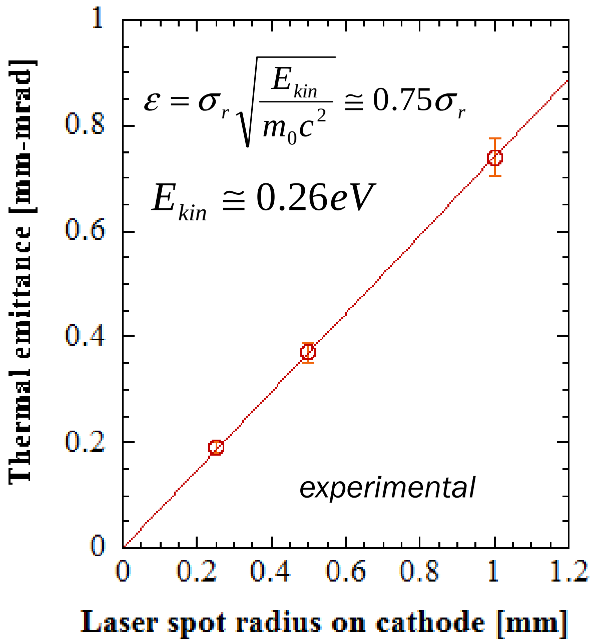


UV: 266 nm
pulse width: 100fs
spot-size: 0.1mm

Femtosecond electron beam

Emittance: 0.1 μm
Energy spread: 10^{-4}

Emittance as a function of laser spot radius on Cu cathode



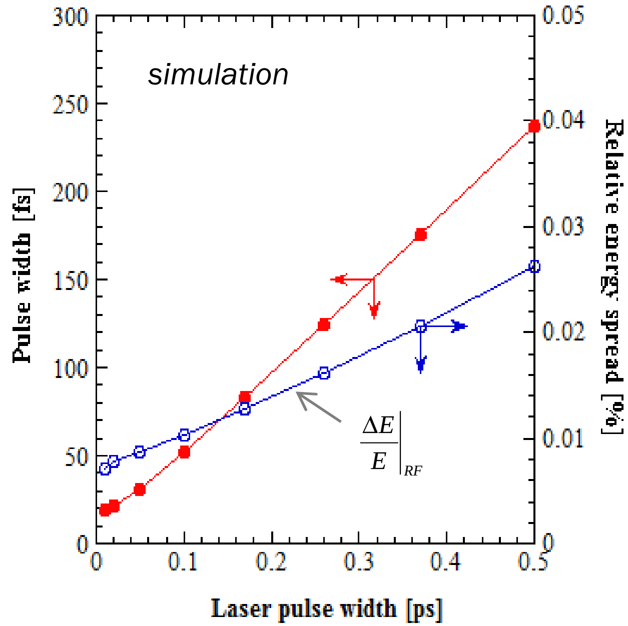
✓reduce thermal emit. with small size laser

ε_{th}~0.1 mm-mrad at σ_r=0.1mm

✓ Initial energy spread emitted from Cu cathode

$$\left.\frac{\Delta E}{E}\right|_{in} = \frac{E_{kin}}{E} \approx 10^{-4}$$

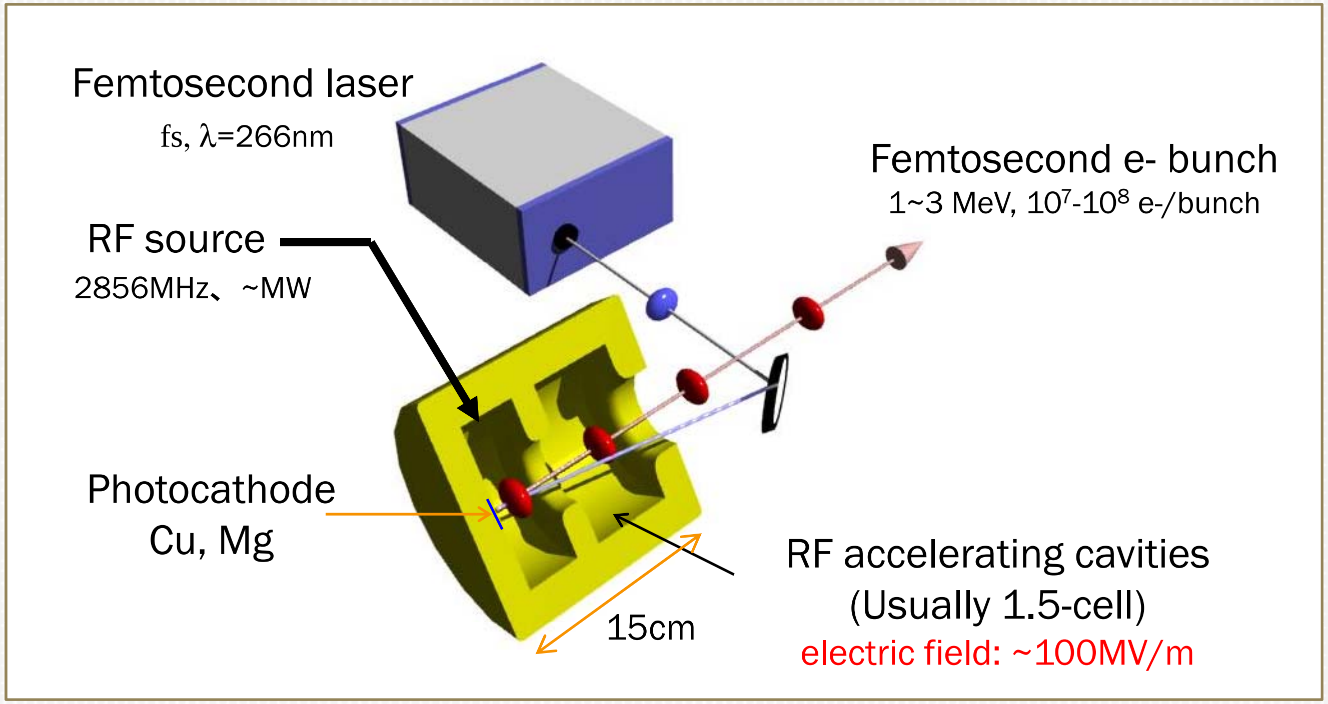
Energy spread as a function of laser pulse width



✓ reduce energy spread due to RF effect with short-pulse laser

ΔE/E ~ 10^{-4} at σ_z<100 fs

RF gun based UED in world



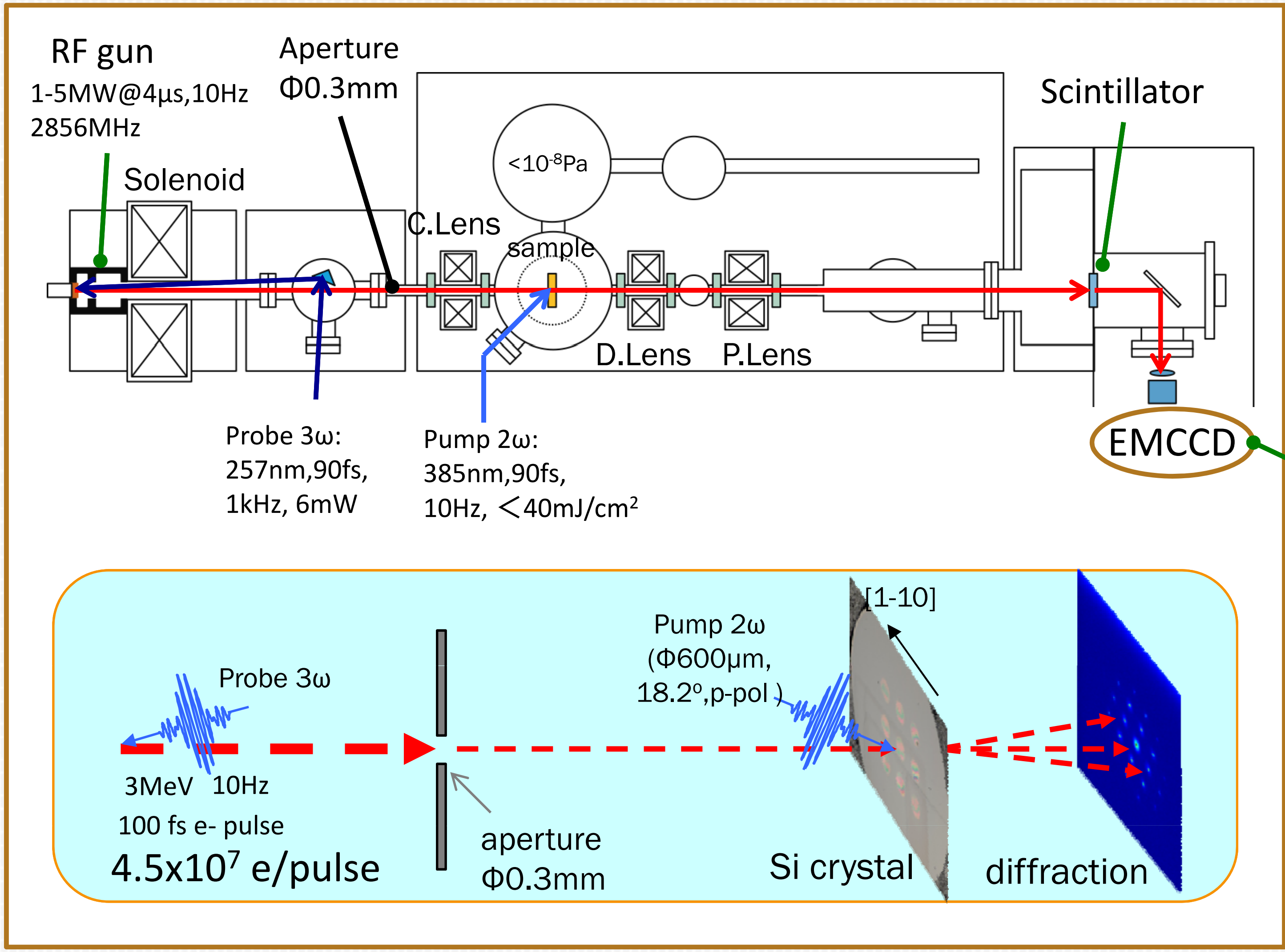
Recently, the RF gun has been successfully used/proposed in UED facilities at BNL, SLAC, UCLA, Tsinghua Univ. , Osaka Univ., DESY, Shanghai Jiaotong Univ., KAERI, ...

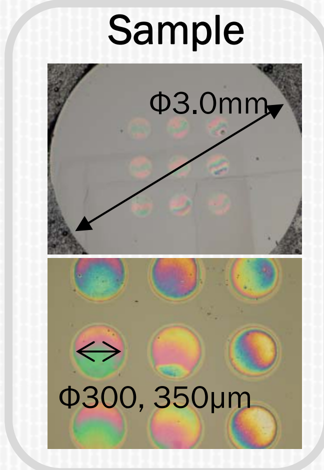
Ultrafast electron diffraction at Osaka University

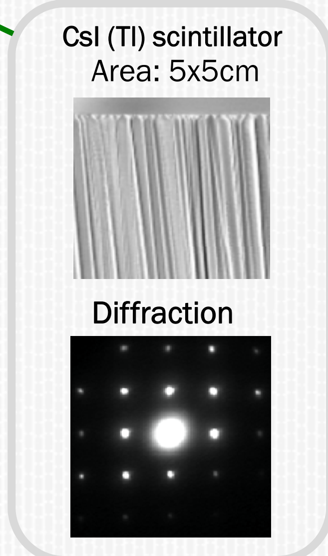
Electron energy: 1～3 MeV
Time resolution: 100 fs

RF gun based MeV UED at Osaka Univ.

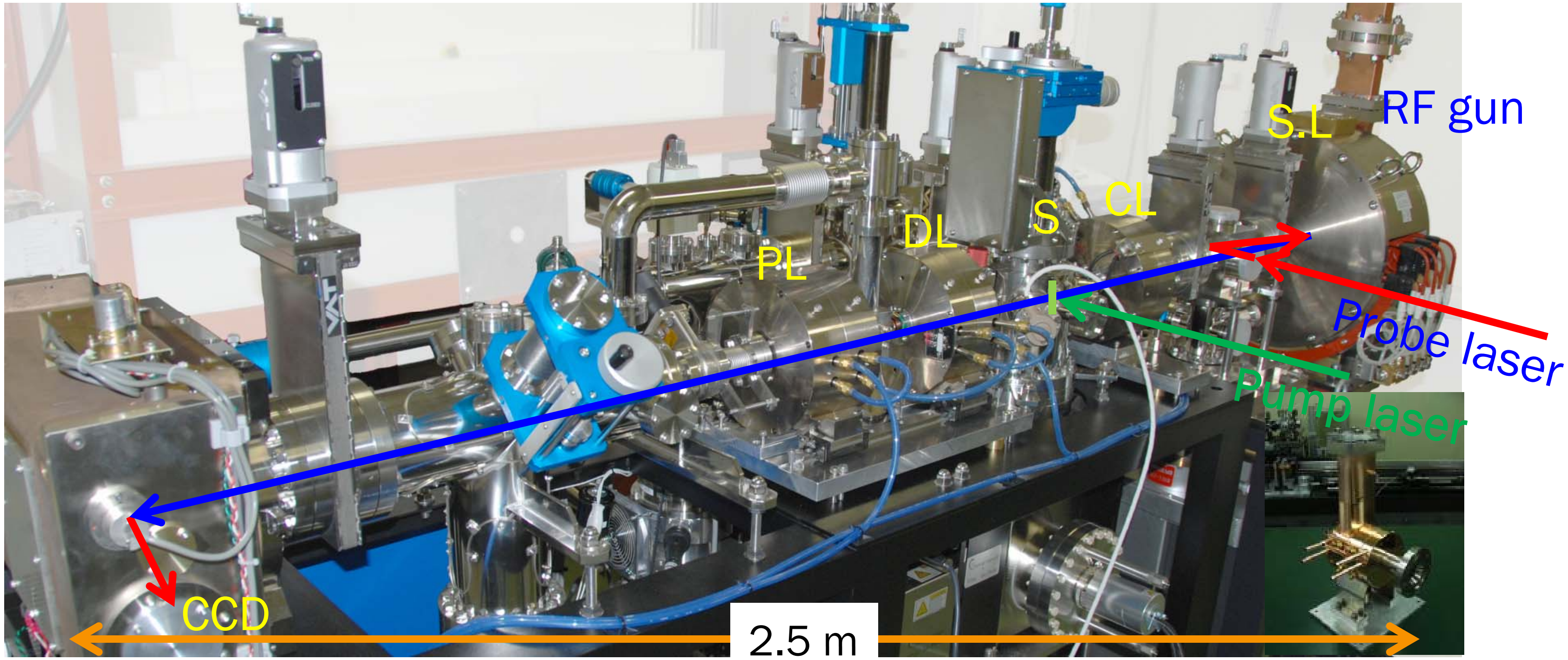
use of electron optical lenses as like in electron microscopy







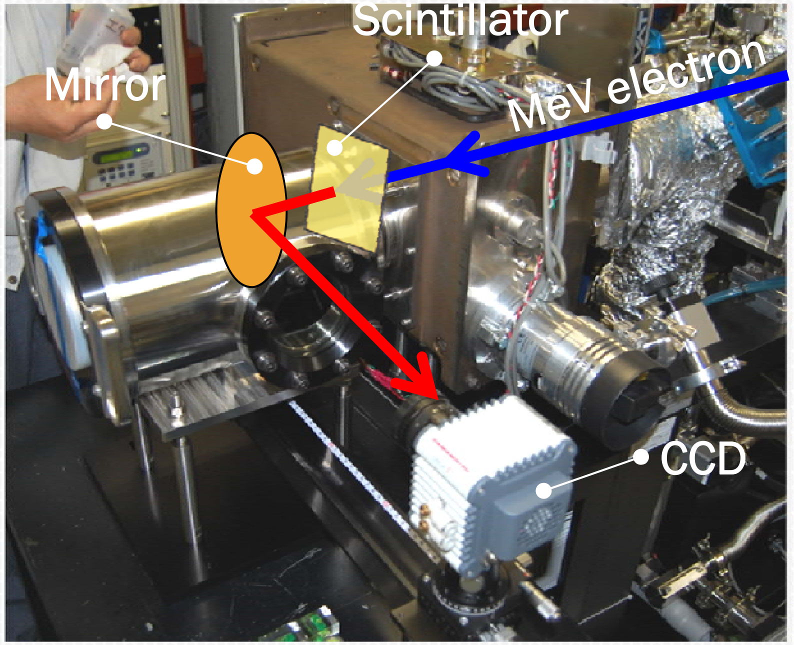
Picture of UED system at Osaka Univ.

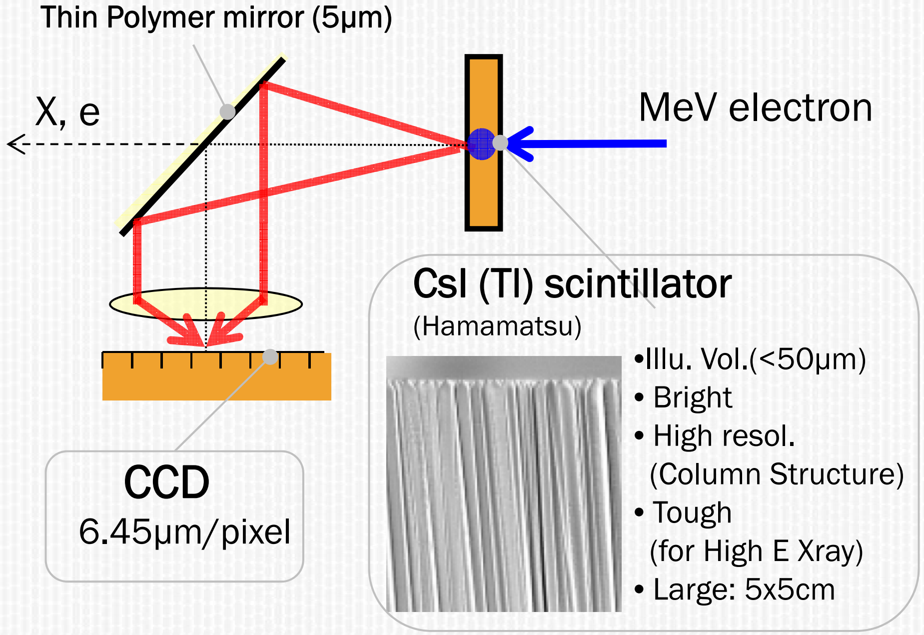


use of electron optical lenses, therefore, compact.

Detection of MeV electron diffraction

Requirements of MeV electron detector: high resolution, high efficiency, no damage





Problems

- Very low current, i.e. ~pA
- Small scattering angle, i.e. 0.1mrad
- Strong X-ray emissions,
  i.e. Backgnd, pixel defect
- Damage by MeV electron,
  i.e. scintillator, fiber
- Diff. Pattern to be magnified/shifted

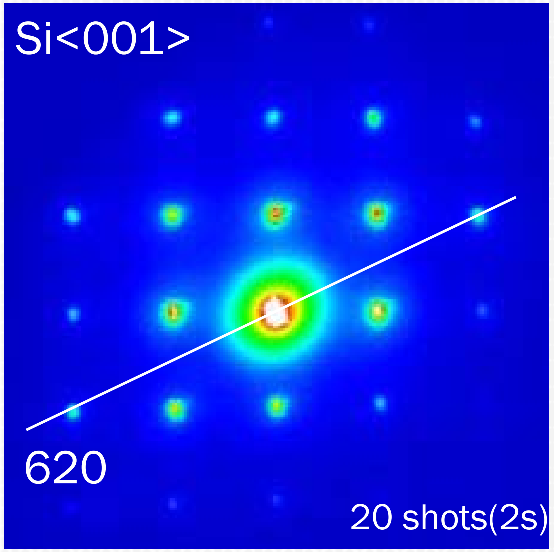
Solution

- CsI: Small Illumination volume
  size-matched to CCD pixel
- Indirect exposure
  Thin mirror + Lens coupling
- No pixel defect observed yet
- Large detection area, i.e. $5x5cm^2$

Quality of MeV electron diffraction

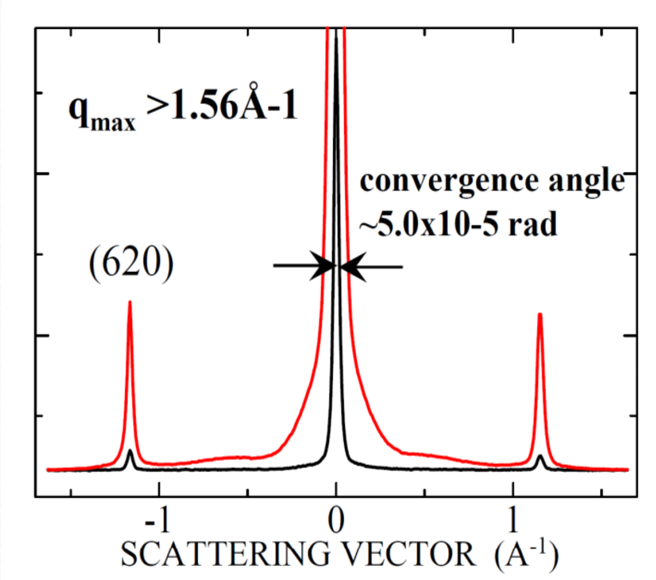
Electron beam: 3 MeV, $8.9\times10^{7} e/cm^{2}/pulse$

Sample: 180nm-thick single crystal Si



A high-quality MeV ED was observed!

Intensity profile of 620 pattern



- Beam convergence angle: 0.05 mrad
- Maximum scattering vector : q_{max} >1.56Å^{-1}
- Requirement of the e^- number: 10^{6-7}

- Bragg law

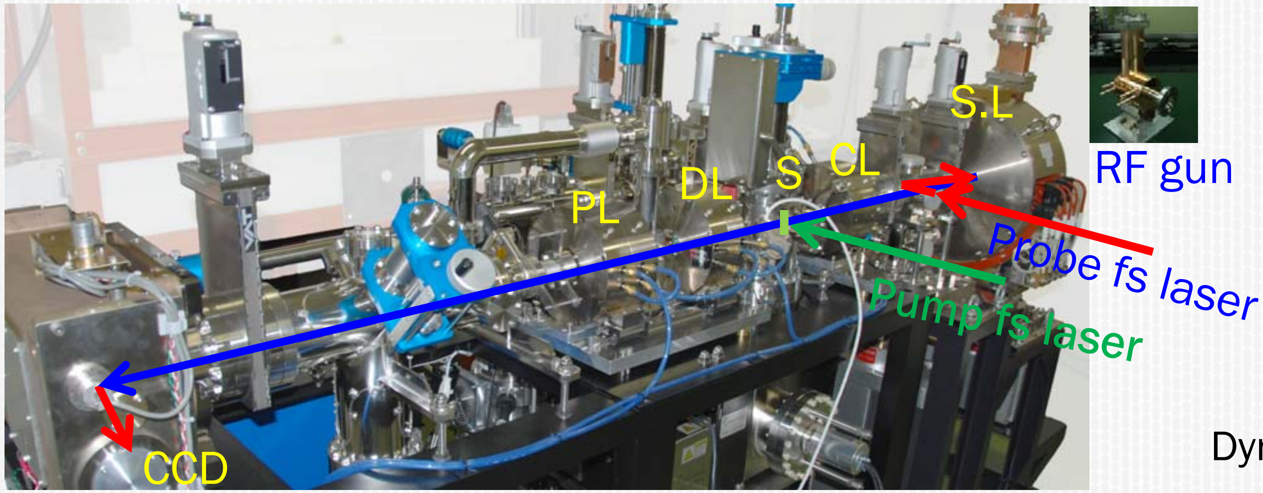
$$2d\sin\theta = n\lambda$$

$$\tan\theta = \frac{D}{L}$$

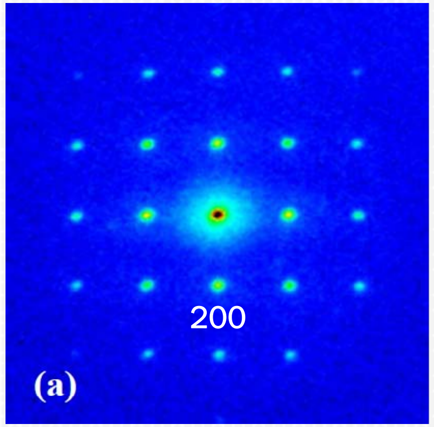
Y. Murooka, et al., Appl. Phys. Lett. 98, 251903 (2011)

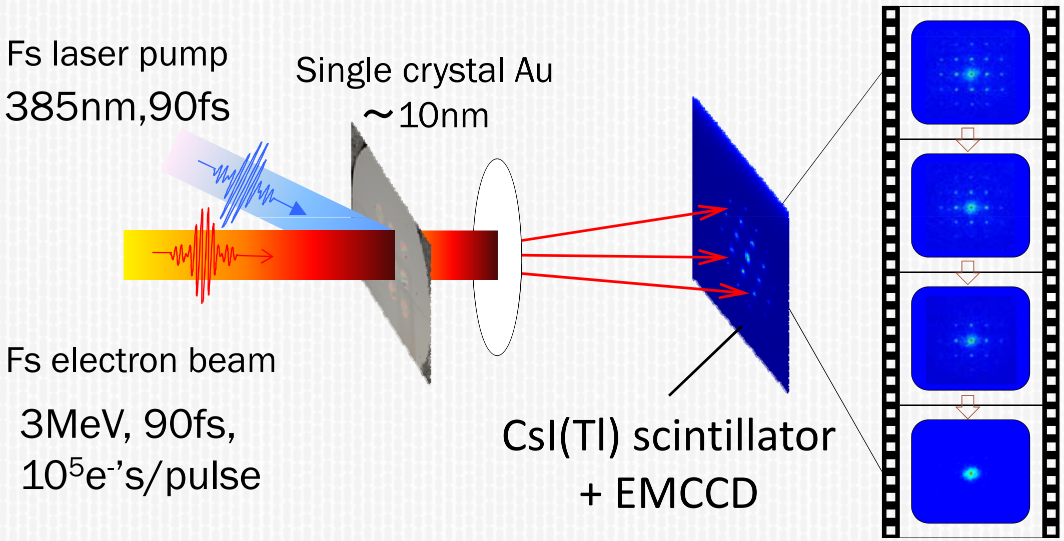
UED$^{\#2}$: Phase transformation on single-crystal Au

Femtosecond MeV electron diffraction at Osaka Univ.

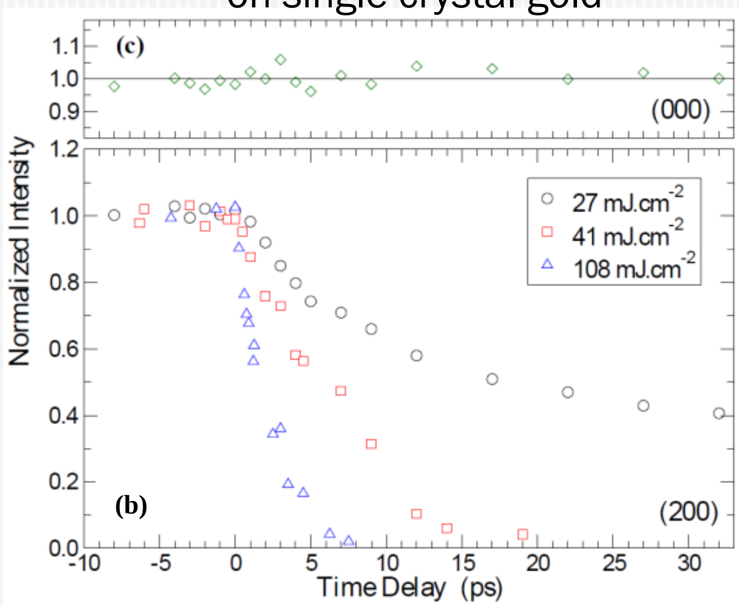


MeV electron diffraction (Single-shot meas.)





Dynamics of laser-induced melting on single crystal gold



PRB 2013; APL accepted.

Concluding remarks in UED

- The UED experiments indicate that the RF gun based MeV UED is useful tool for the study of ultrafast dynamics with time resolution of 100 fs or less.
- However,
  - Can RF gun be used in time-solved electron microscopy?
  - What kind efforts and challenges are needed?

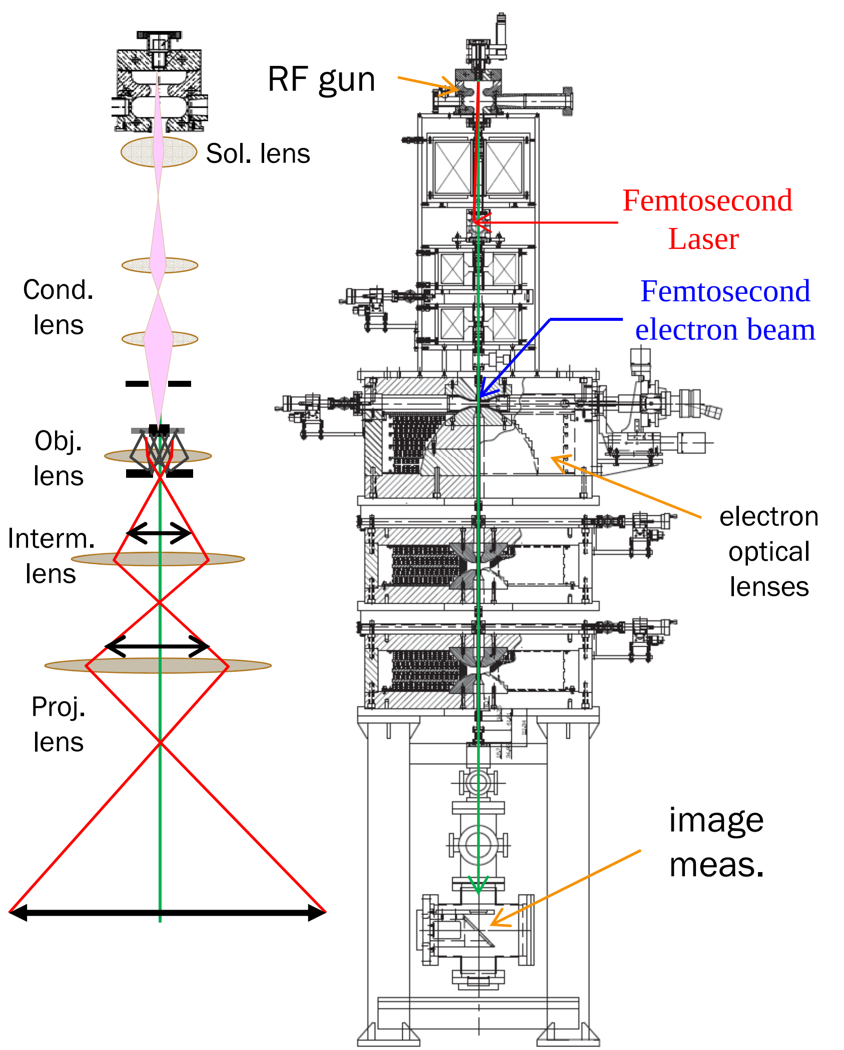
↓

To answer the questions,
we developed a prototype of time-resolved TEM using RF gun.

Femtosecond time-resolved electron microscopy using RF gun

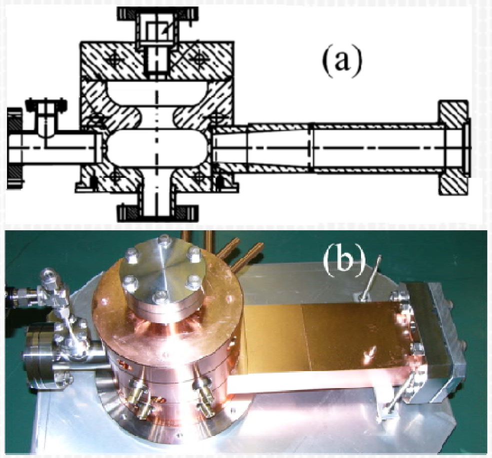
(under development at Osaka Univ.)

Concept of RF gun based electron microscopy



Prototype of MeV UEM

Femtosecond photocathode electron gun



Electron energy：	1~3 MeV
Bunch length ：	100 fs
Emittance：	0.1 mm-mrad
Energy spread：	10^{-4} (10^{-5} for challenge)
Charge：	10^7~10^8e's/pulse

Time resolution: 100 fs
Spatial resolution: 10 nm

Challenge !

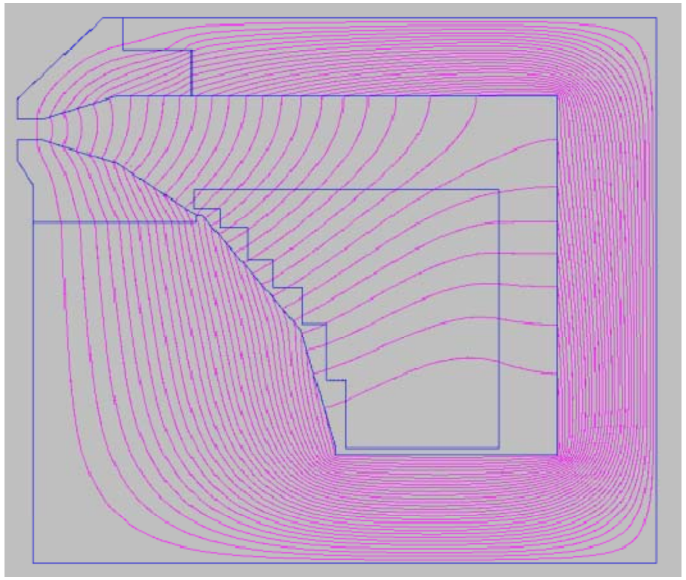
First prototype of RF gun based electron microscopy

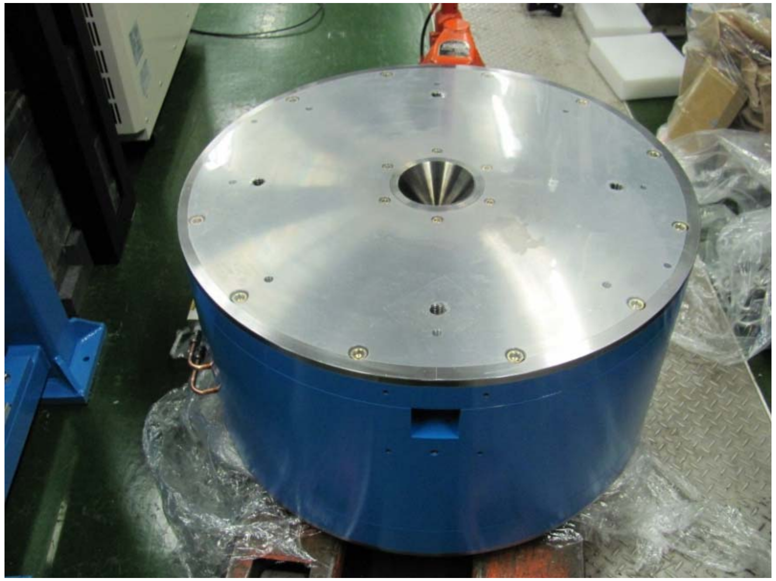
Prototype of MeV UEM at Osaka Univ.

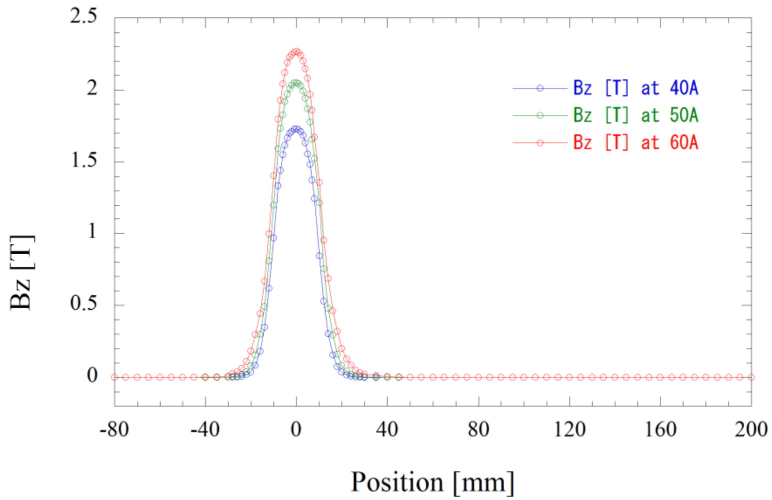
(height: 3m, diameter: 0.7m)

- The prototype was constructed at the end of Oct. 2012.

Design of 2T objective lens







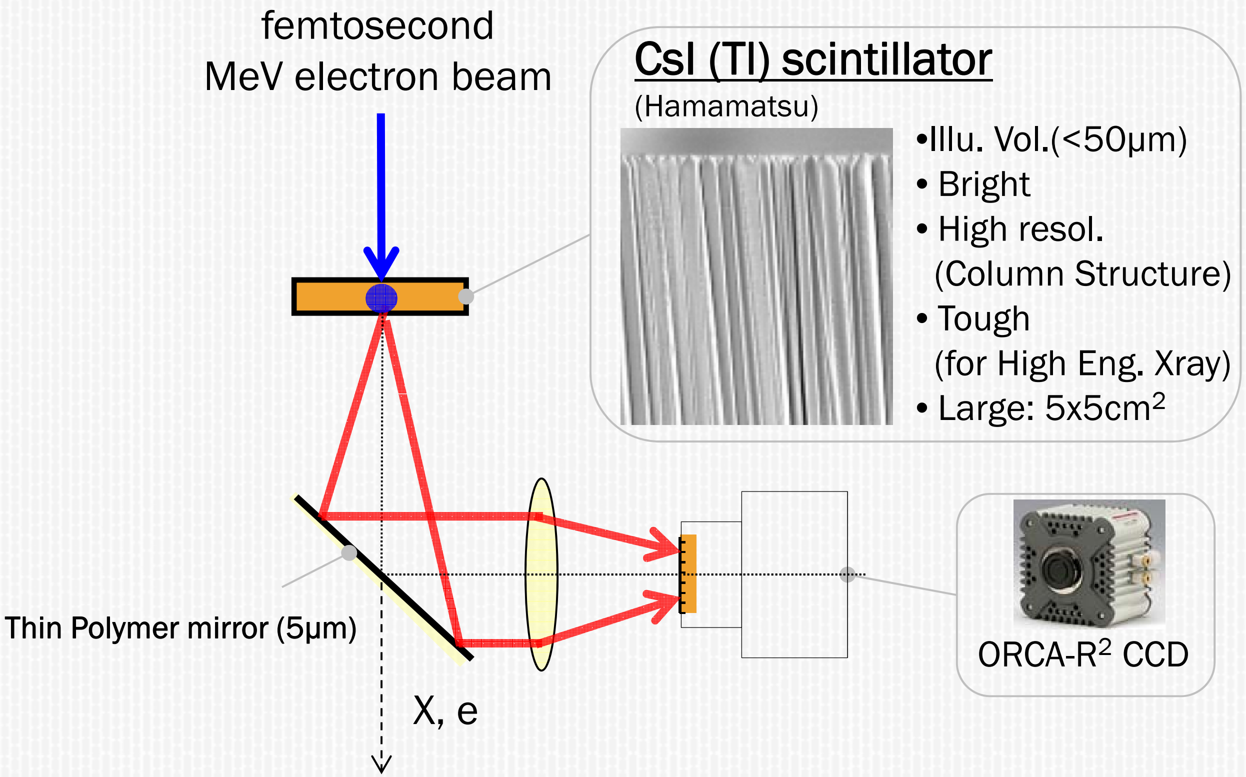
磁極口径	φ13 $^{+0.05}_{0}$ mm
磁極長	350mm
磁場強度	2.2T
起磁力	35000 AT

f_0 = 5 mm for 2MeV e^-

C_s (C_c) ~ 4 mm

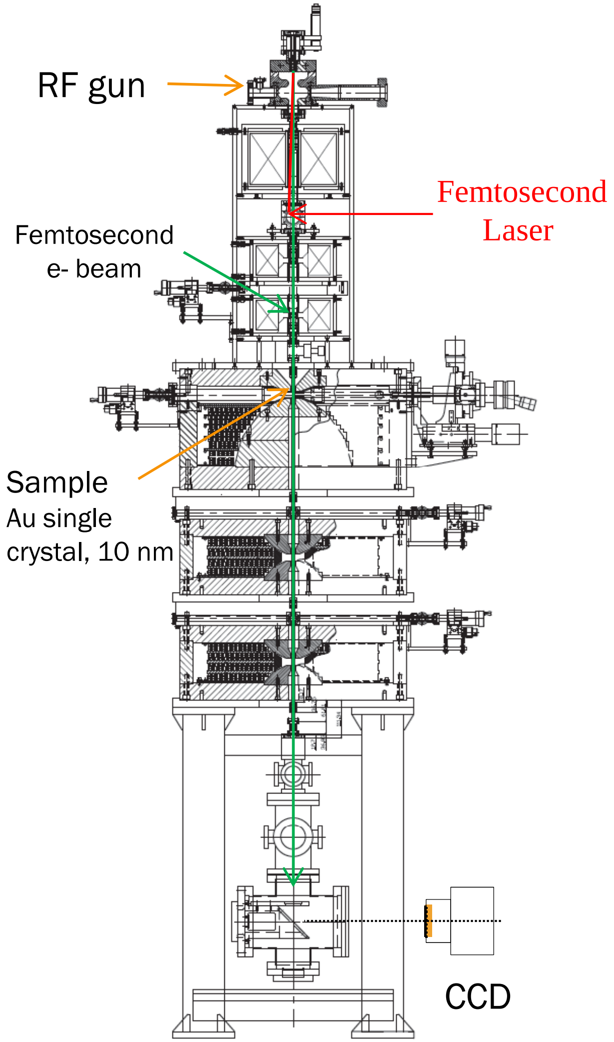
70 cm (D) x 35 cm (H)

Detection of MeV electron images



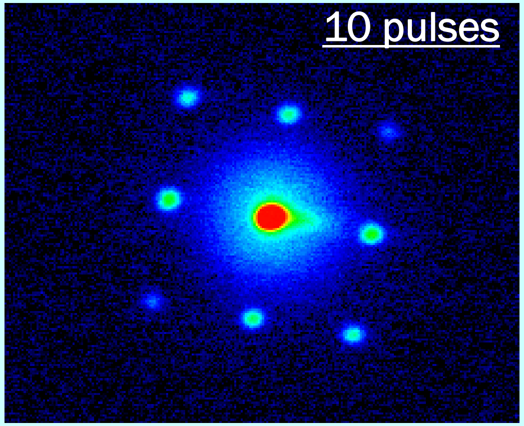
The detection system was successfully used in UED measurement.
(single-shot measurement with 10^5 e^-s/pulse)

Demonstrations of MeV ED/TEM imaging



Prototype of MeV UEM in Osaka Univ.

MeV electron diffraction

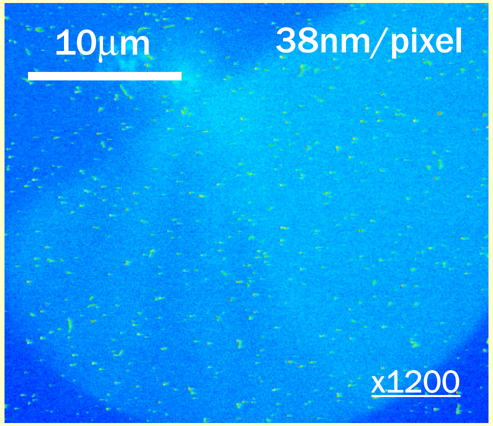


Sample: Au single-crystal film (10nm)

Electron energy: ~ MeV
Electron number:
10^5 e-'s/pulse)

A good-quality MeV ED was observed at 10^6 e-'s/image!

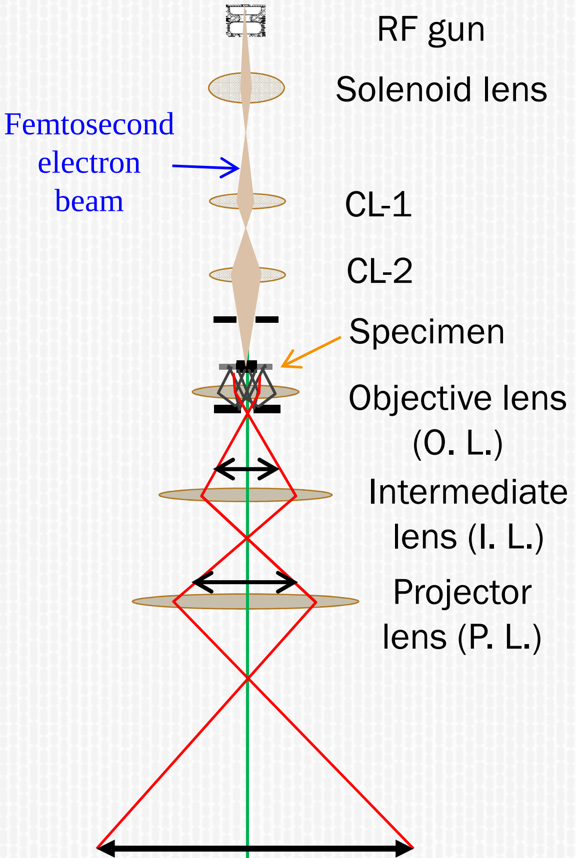
Relativistic-energy TEM image



Electron charge:
~10 fC/pulse
(10^5 e-'s/pulse)
Measurement time:
~10 min

~10^8 e-'s/image

Magnifications & resolutions

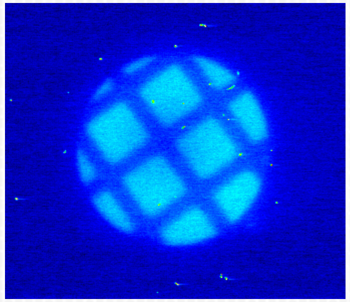


Spatial resolution

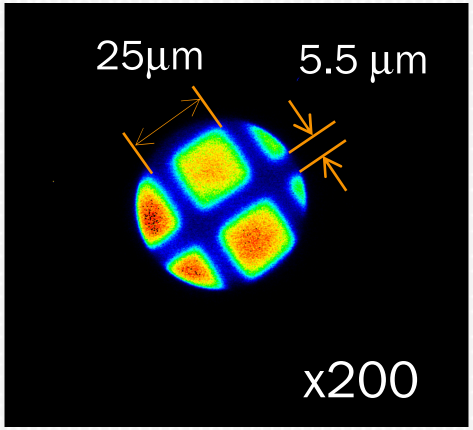
10nm (next step)
x10,000

~1nm (in future)

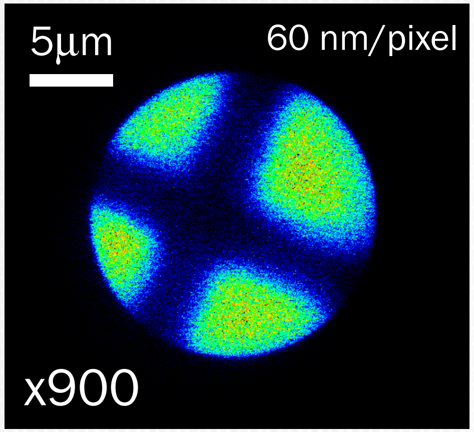
Sample:
Standard Cu grid(1000mesh)



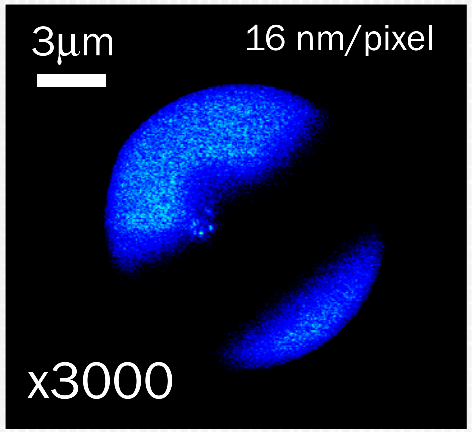
e^- energy: ~ MeV
e^- number: ~10^7/pulse



Using Obj. lens only



Using O.L. and I. L.



Using O.L., I. L. and P. L.

Conclusion and remarks

✓Both RF gun based UED and UEM systems have been constructed at Osaka University.

✓In UED, single-shot and time-resolved measurements have been succeeded. The time resolution was achieved to be 100 fs.

✓In UEM, the MeV electron diffraction and imaging experiment was carried out. The resolution of 30 nm was achieved.

✓Both experiments suggest that RF gun is very useful for ultrafast MeV electron diffraction and is also expected to be used in ultrafast electron microscopy.

However, great efforts and many challenges are required:

- reduce further the emittance (<0.1 μm) and energy spread (10^{-5} or less),
- increase the beam brightness,
- improve the stabilities on the charge and energy,
- develop a detection of very electron with MeV energy, and so on.