

Ultrafast relativistic-energy electron microscopy

Jinfeng Yang

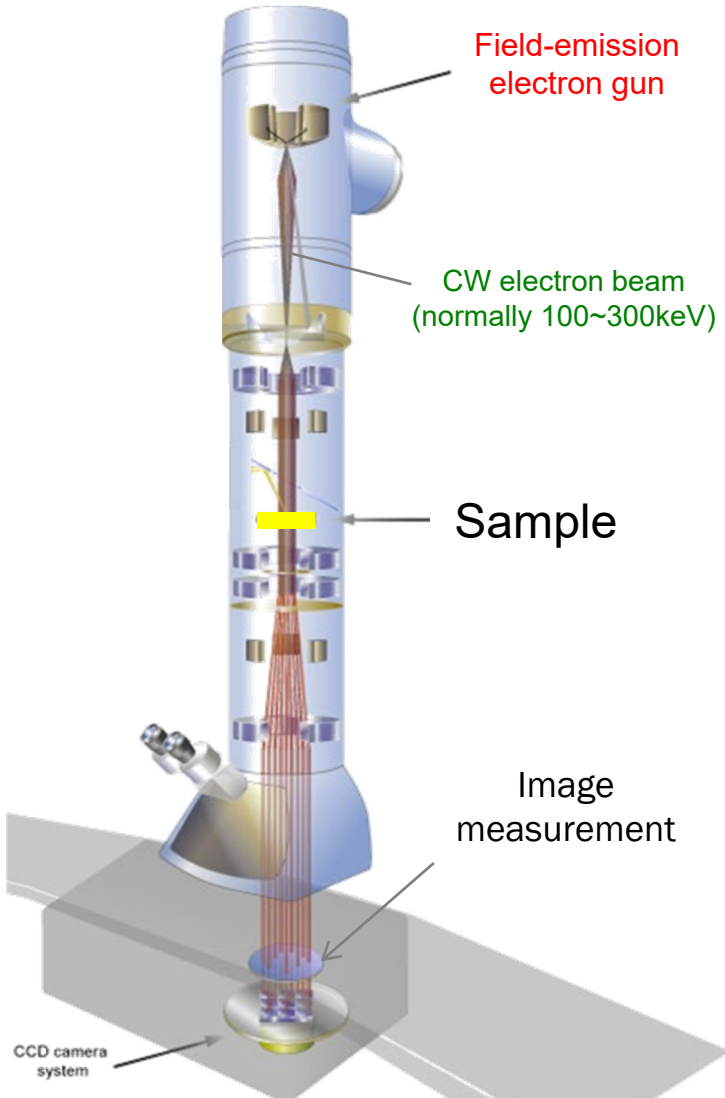
The Institute of Scientific and Industrial Research (ISIR)
Osaka University



Motivations

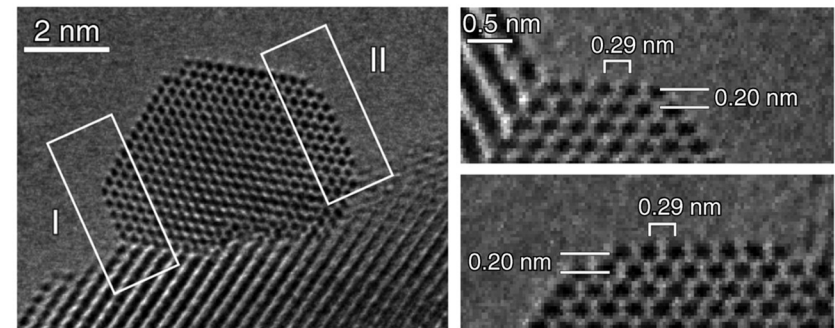
(1) Commercial Electron Microscopy

- A powerful tool to observe the **static image** of atomic structures in materials
- Spatial resolution: **<0.1 nm**



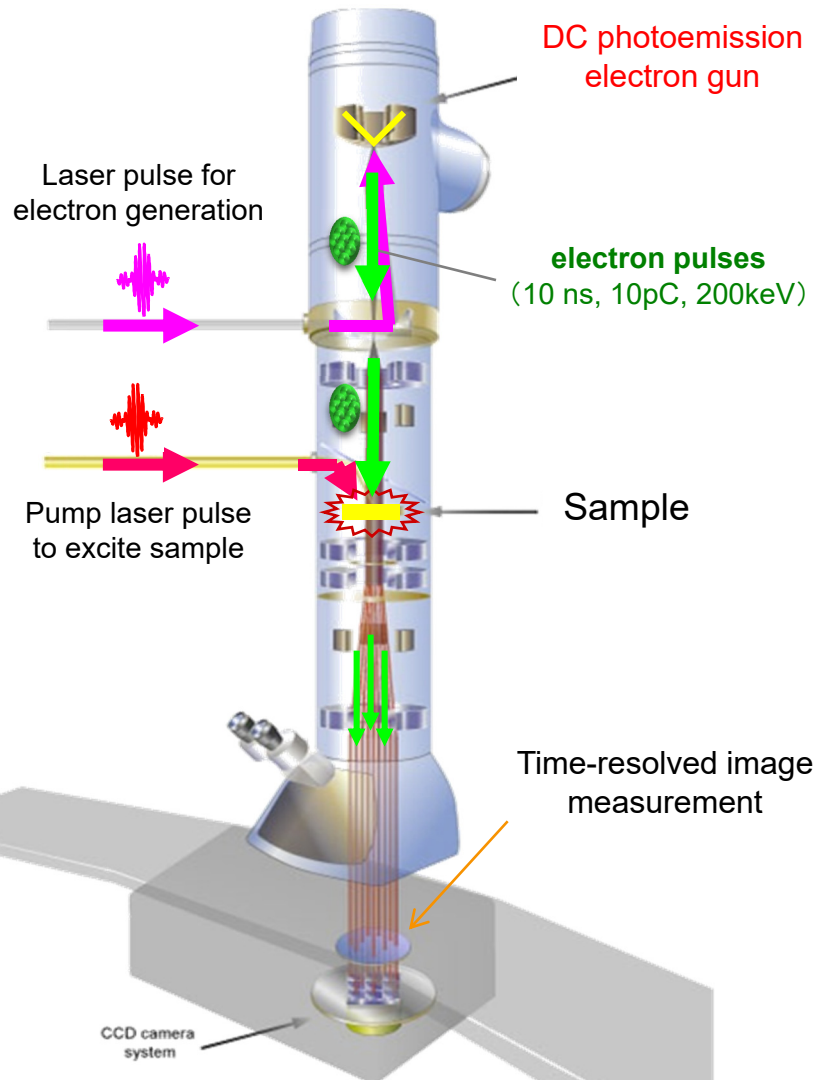
JAP 97, 111101 (2005)

Ex.: Images of Au atoms in vacuum observed by 300-keV TEM



Science 335, 317, 2012

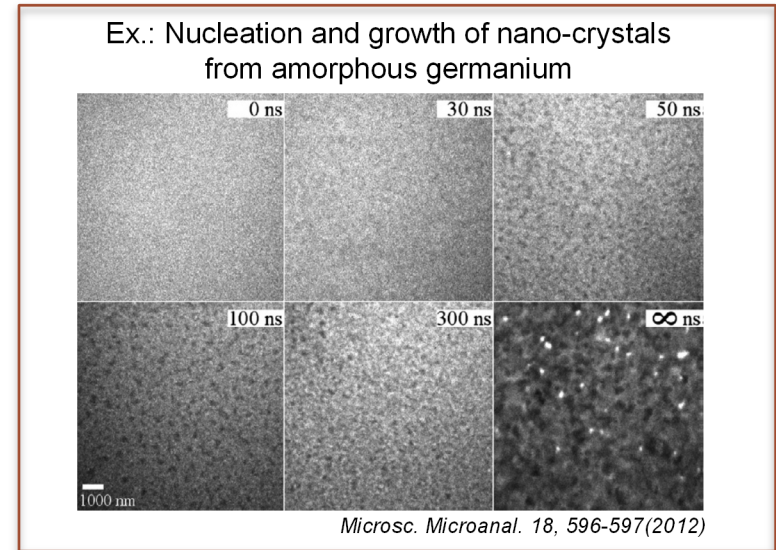
Motivations



JAP 97, 111101 (2005)

(2) Ultrafast Electron Microscopy(UEM)

- UEM can observe a **structural movie** for study of dynamics in materials.



- However, the temporal resolution of single-shot measurement is

~10 ns

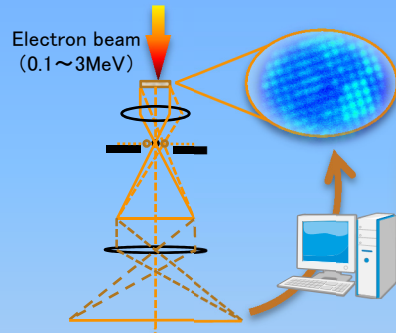
which is limited by the electron pulse length under the low-energy region.

UEM and its applications



Imaging Technology

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



Imaging in “real space”

Methods



nm

fs

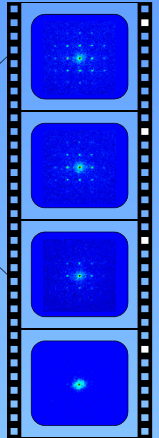
Pump laser pulse

Electron pulse (probe)

Ultrafast Observation

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

Observation in “real time”

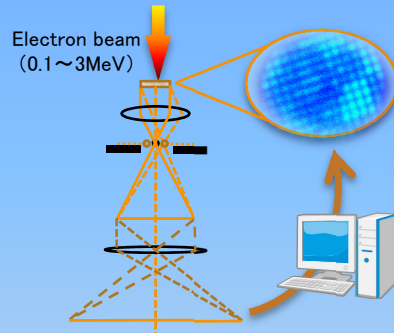


UEM and its applications



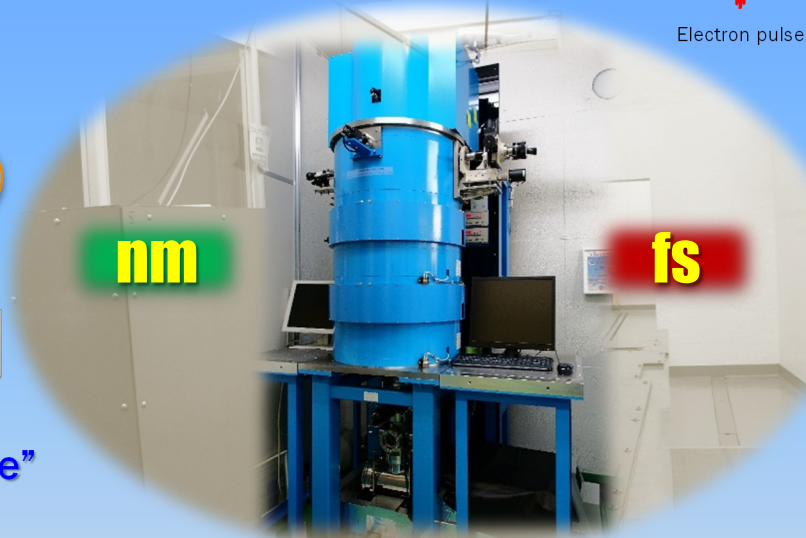
Imaging Technology

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



Imaging in “real space”

Methods



nm

fs

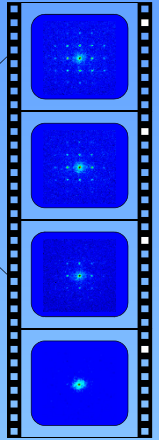
Pump laser pulse

Electron pulse (probe)

Ultrafast Observation

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

Observation in “real time”



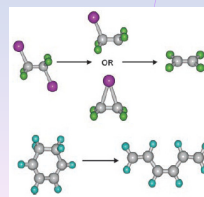
Protein Structural Dynamics



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

Targets

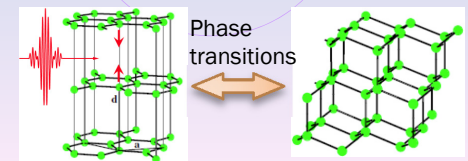
Making Molecular Movie



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

Nano-technology/science

- Dynamics of phase transients in solid-state materials.
- Creation of new functional materials and devices for nanotechnology.



Why use RF gun in microscopy?

The ultrashort electron pulse is a space-charge dominated particle beam!

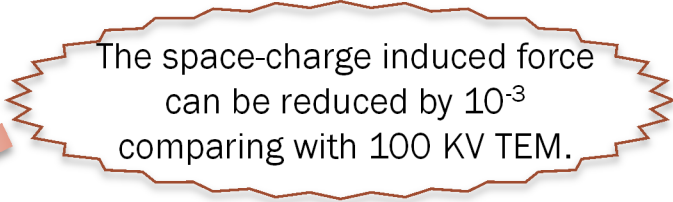
Why use RF gun in microscopy?

The ultrashort electron pulse is a space-charge dominated particle beam!

✓ Space-charge induced force can be reduced.

$$\vec{F}^{sc} \cong e\vec{E}^{sc} / \gamma^2$$

Beam energy using RF gun: 3 MeV



The space-charge induced force can be reduced by 10^{-3} comparing with 100 KV TEM.

In our experiment, 100-fs electron pulses at 3 MeV and 1~10 pC are possible.

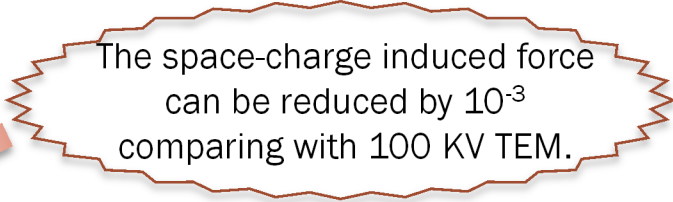
Why use RF gun in microscopy?

The ultrashort electron pulse is a space-charge dominated particle beam!

✓ Space-charge induced force can be reduced.

$$\vec{F}^{sc} \cong e\vec{E}^{sc} / \gamma^2$$

Beam energy using RF gun: 3 MeV



The space-charge induced force can be reduced by 10^{-3} comparing with 100 KV TEM.

In our experiment, 100-fs electron pulses at 3 MeV and 1~10 pC are possible.

✓ Multi-scattering of MeV e⁻'s in sample is negligible.

- Easy to understand the dynamics in materials
- Thick sample (~μm)

Why use RF gun in microscopy?

The ultrashort electron pulse is a space-charge dominated particle beam!

✓ Space-charge induced force can be reduced.

$$\vec{F}^{sc} \cong e\vec{E}^{sc} / \gamma^2$$

The space-charge induced force can be reduced by 10^{-3} comparing with 100 KV TEM.

Beam energy using RF gun: 3 MeV

In our experiment, 100-fs electron pulses at 3 MeV and 1~10 pC are possible.

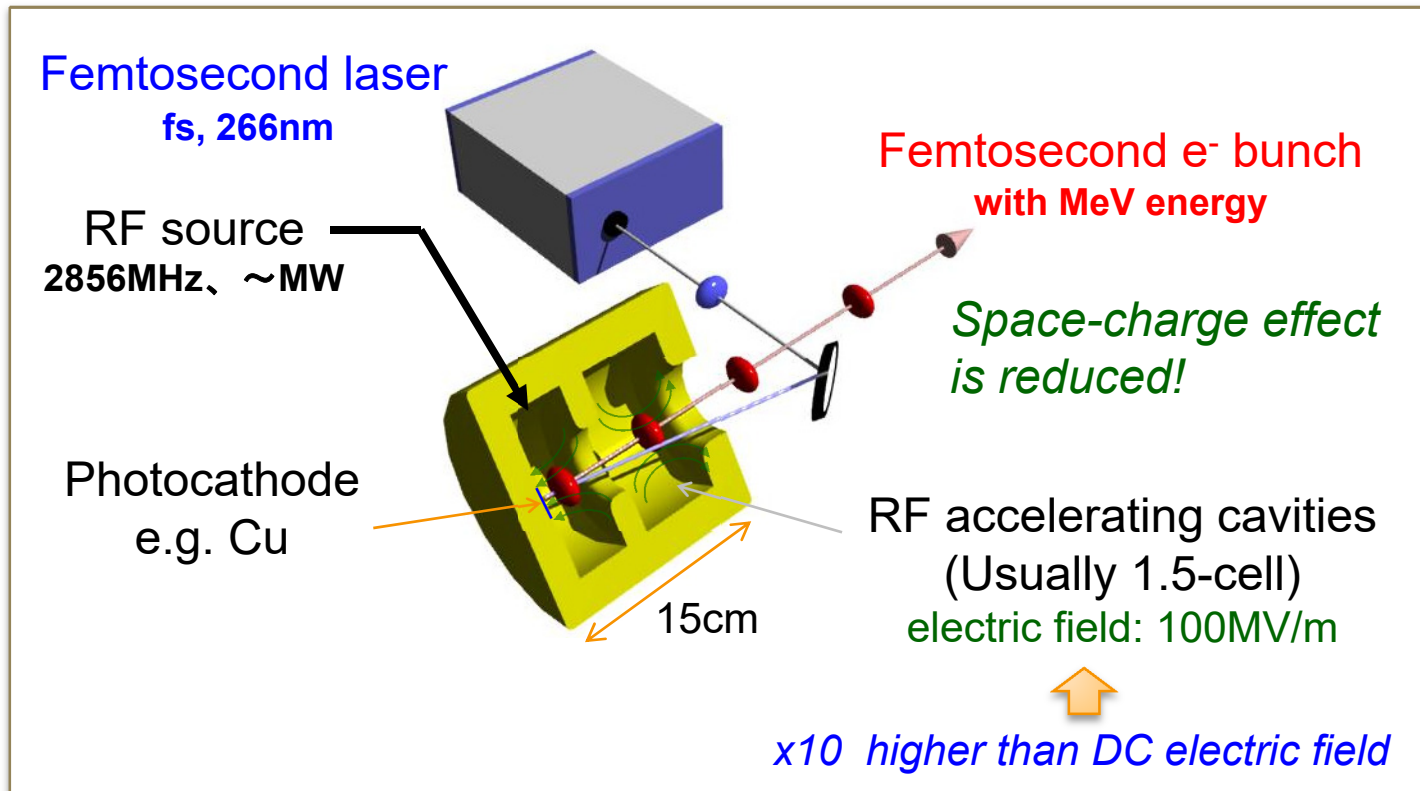
✓ Multi-scattering of MeV e-'s in sample is negligible.

- Easy to understand the dynamics in materials
- Thick sample ($\sim\mu\text{m}$)

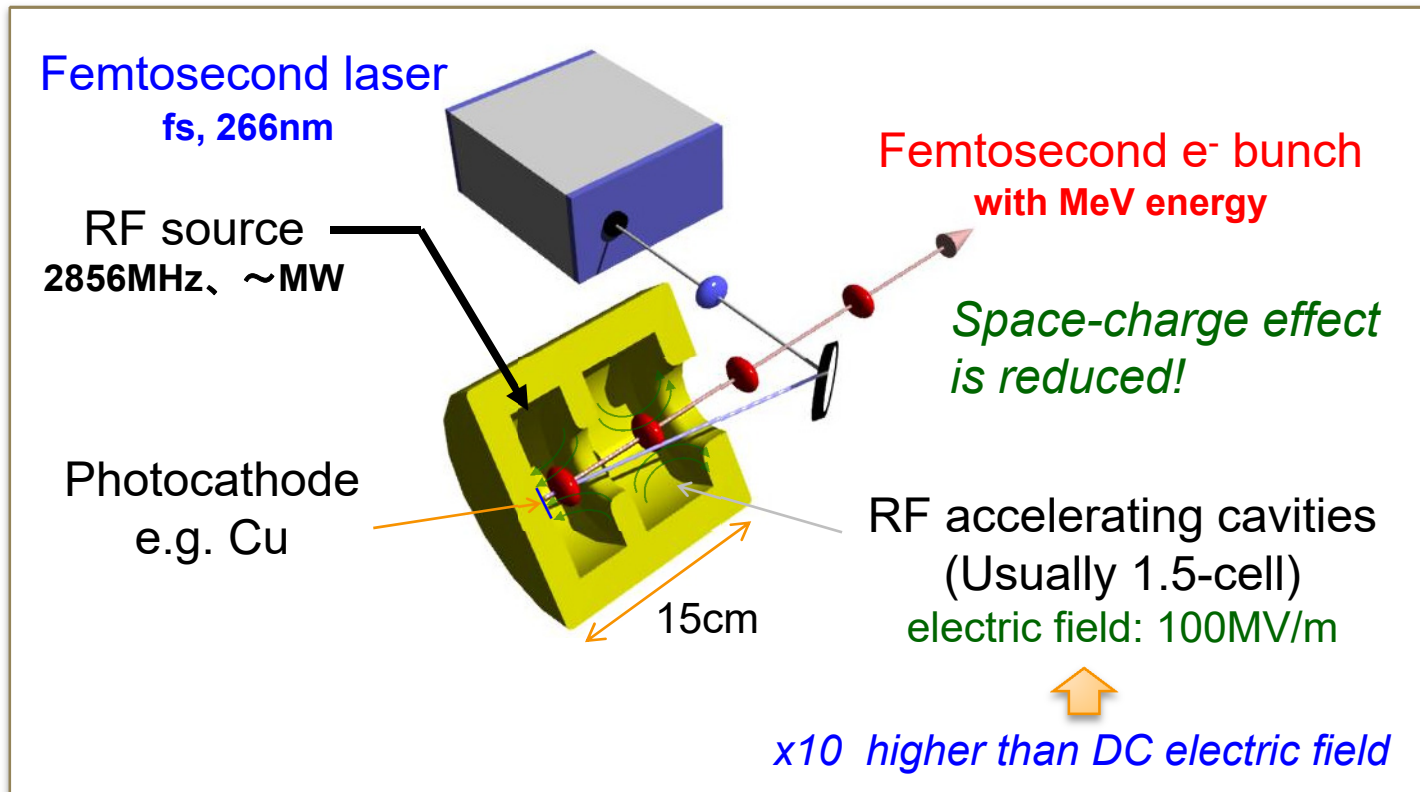
✓ Femtosecond single-shot imaging is realizable.

- Possibility for study the irreversible phenomena or reactions
- Observation before damage for biomolecules

What is RF gun?



What is RF gun?

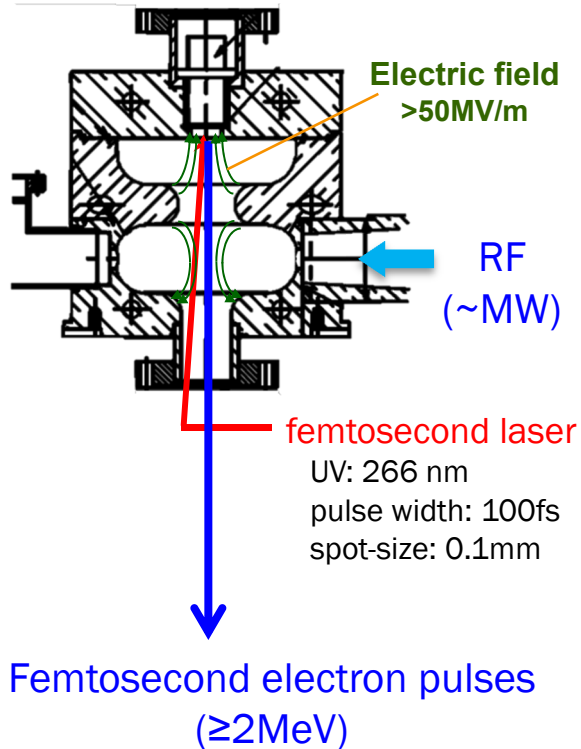


The expected beam parameters:

Electron energy:	1~3 MeV
Bunch length:	100 fs
Norm. emittance:	0.1 mm-mrad
Energy spread:	10 ⁻⁴ (10 ⁻⁵ for challenge)
No. of electrons:	10 ⁷ ~10 ⁸ e ⁻ /pulse

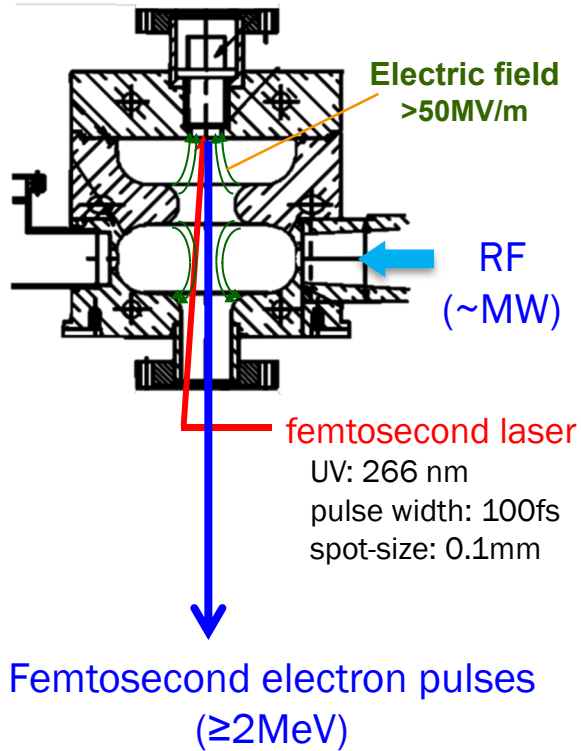
} essential parameters for EM!

100 fs electron pulse generation



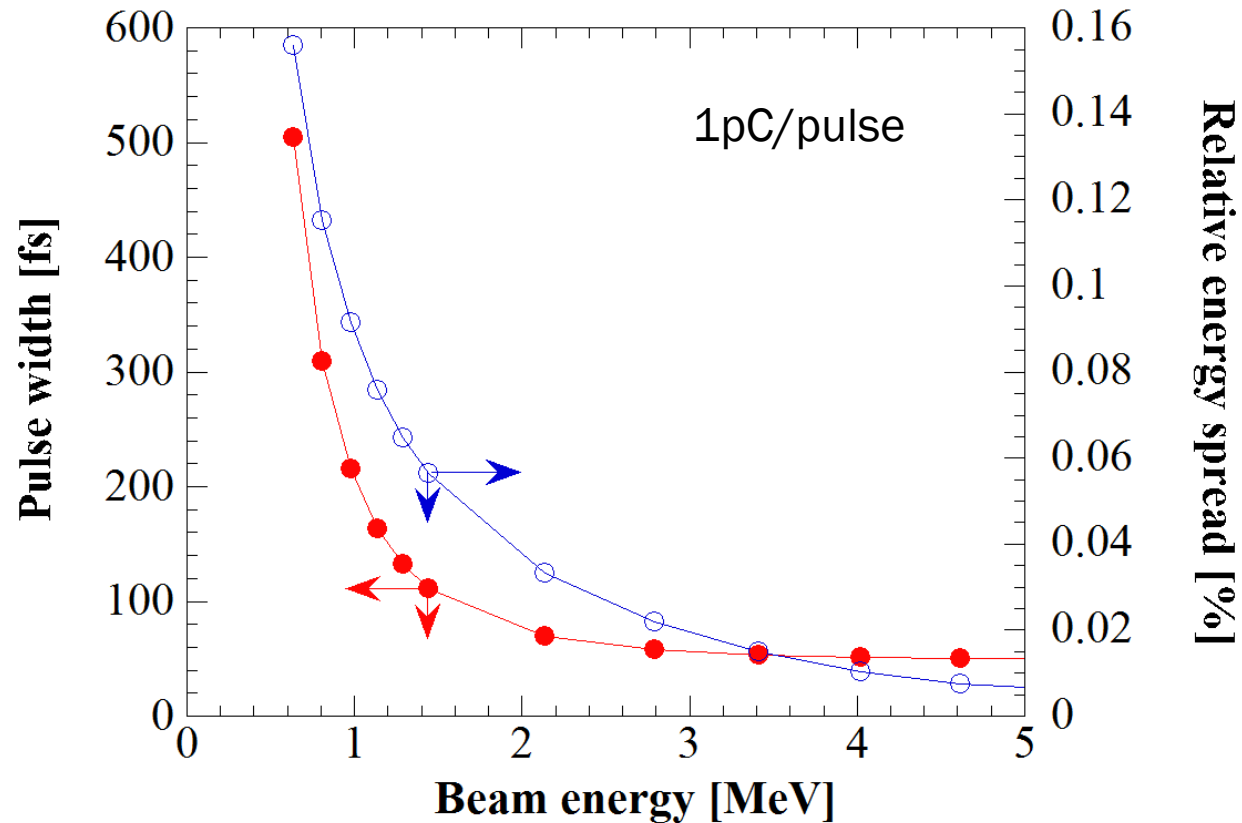
Bunch length is dominated by space-charge effect!

100 fs electron pulse generation



Bunch length is dominated by space-charge effect!

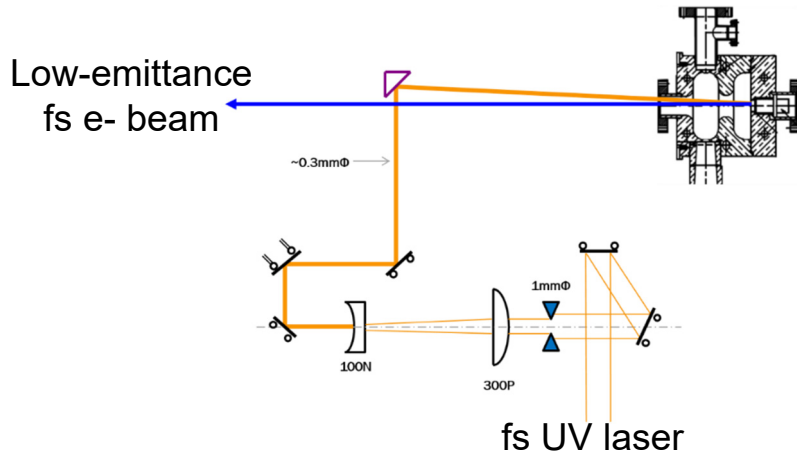
Simulation results



A 100-fs electron pulse with low energy spread of 10^{-4} can be generated at beam energy of ≥ 2 MeV.

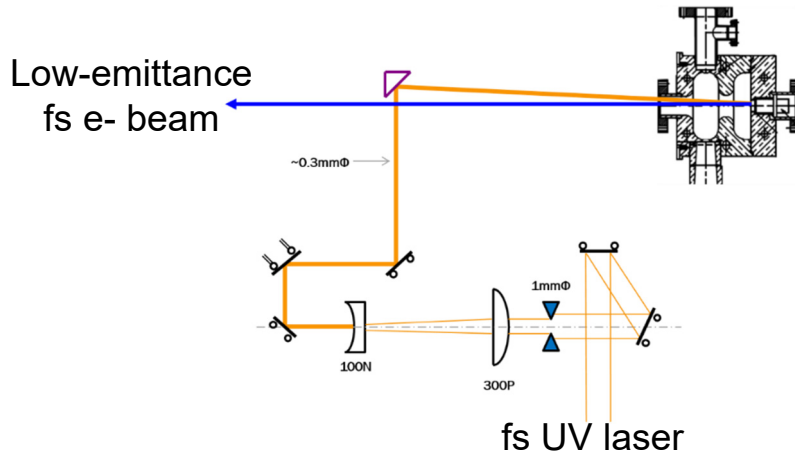
0.1 μm - emittance & fs-bunch electron beam

(1) reduce the thermal emittance by focusing laser on the cathode

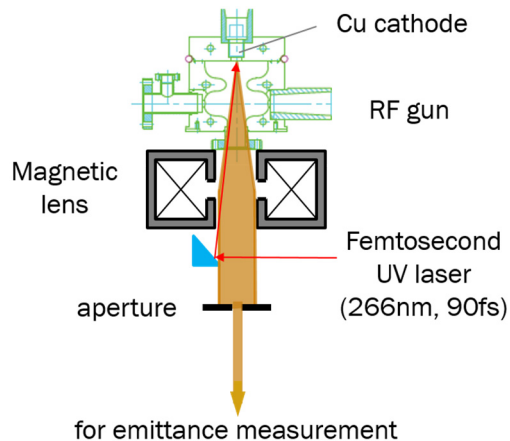


0.1 μm - emittance & fs-bunch electron beam

(1) reduce the thermal emittance by focusing laser on the cathode

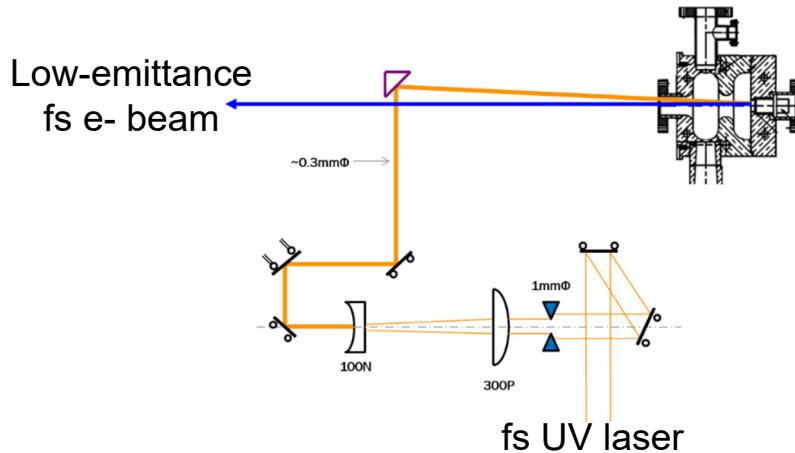


(2) collimate emittance after RF gun

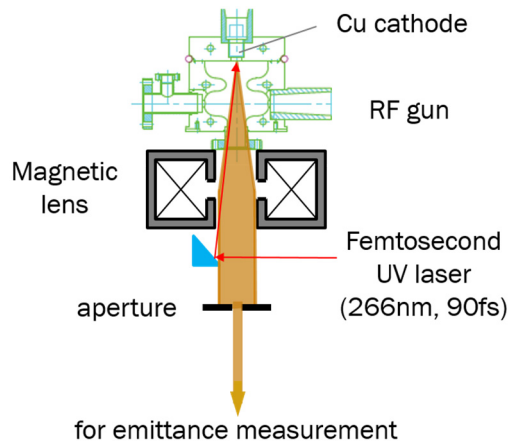


0.1 μm - emittance & fs-bunch electron beam

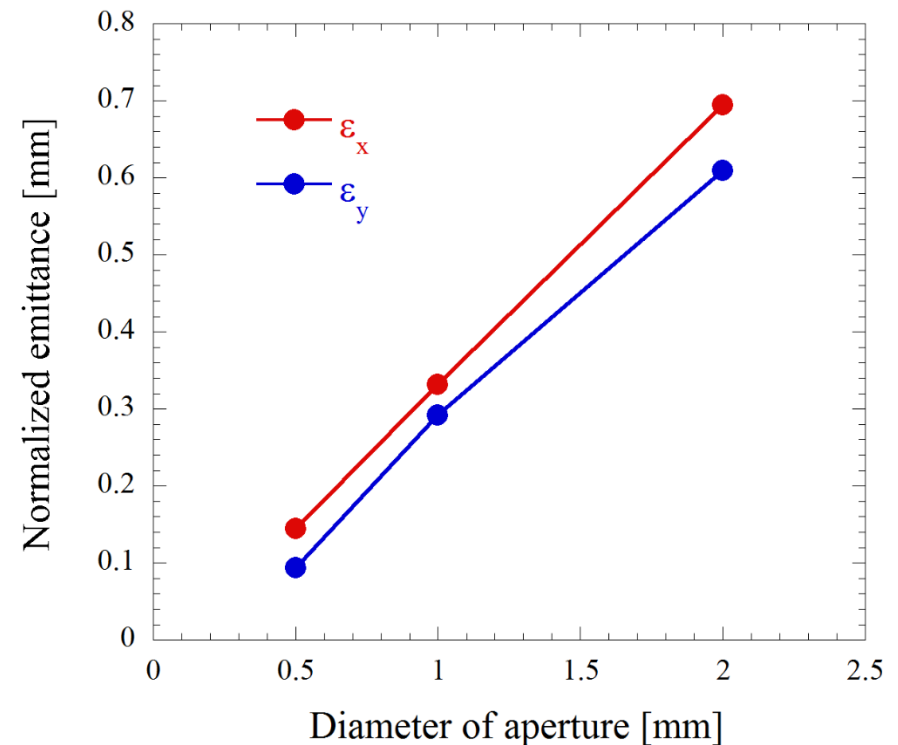
(1) reduce the thermal emittance by focusing laser on the cathode



(2) collimate emittance after RF gun



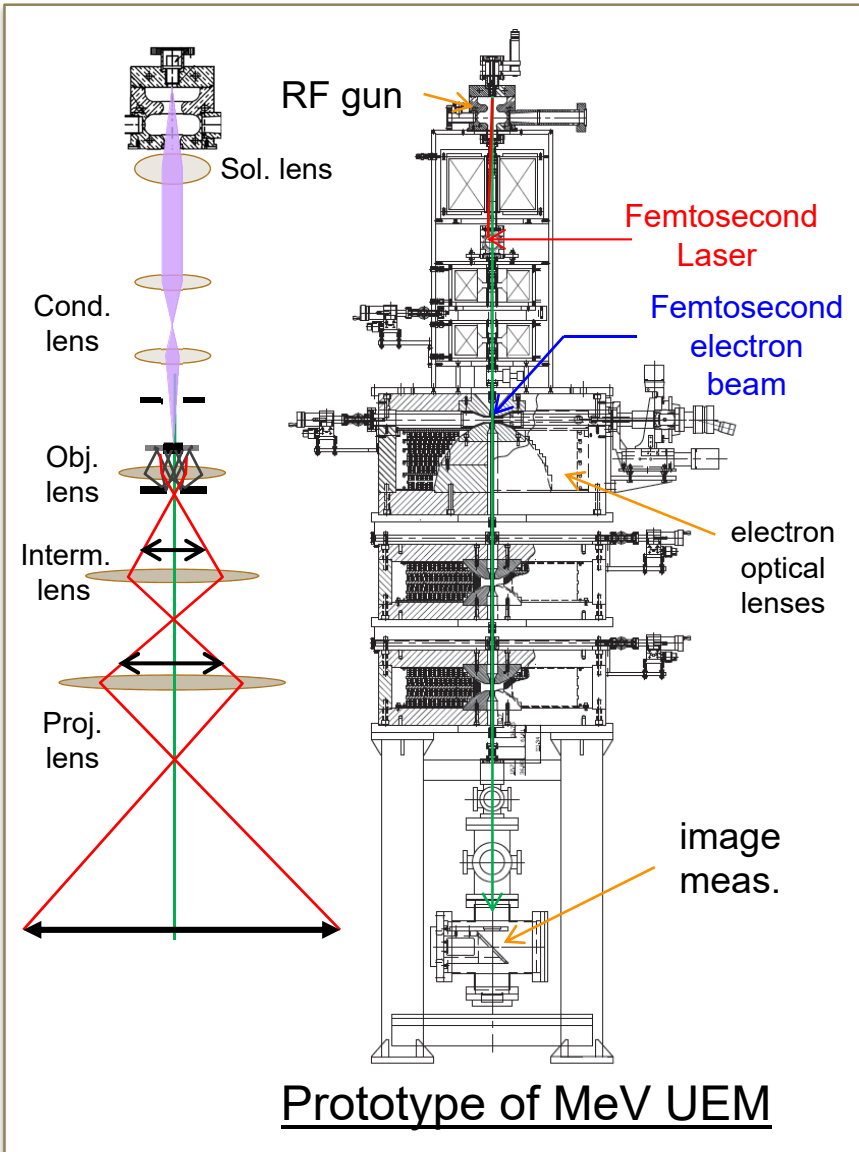
Measurement results of emittance as a function of aperture diameter



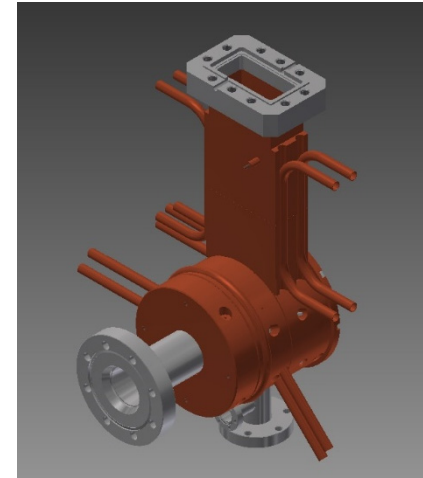
0.1 mm-mrad using a small aperture!

RF electron gun based
Ultrafast Electron Microscopy (UEM)
(The first prototype in world)

Concept of RF gun based 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 : $10^7 \sim 10^8 e^-$'s/pulse

Time resolution: 100 fs
Spatial resolution: 10 nm

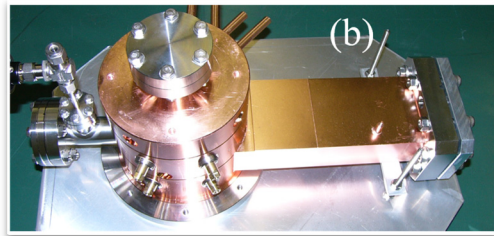
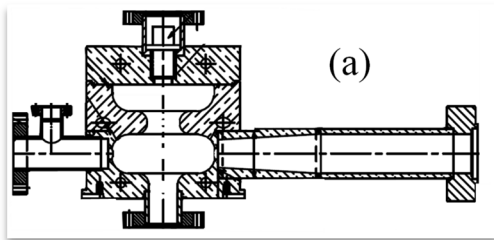


First prototype of RF gun based MeV electron microscopy



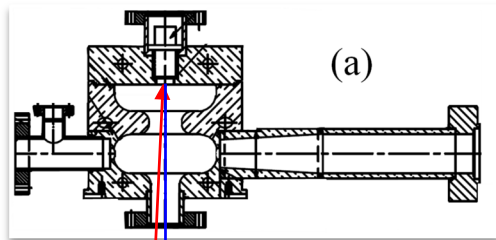
Size: 3m(H) x 0.7m(D)

First prototype of RF gun based MeV electron microscopy



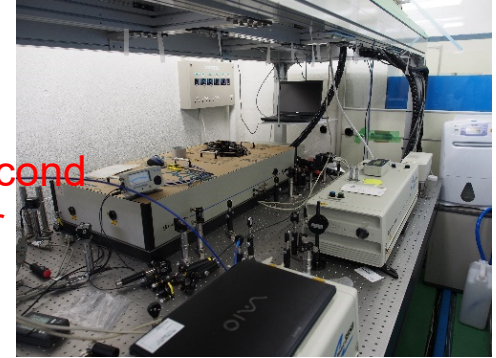
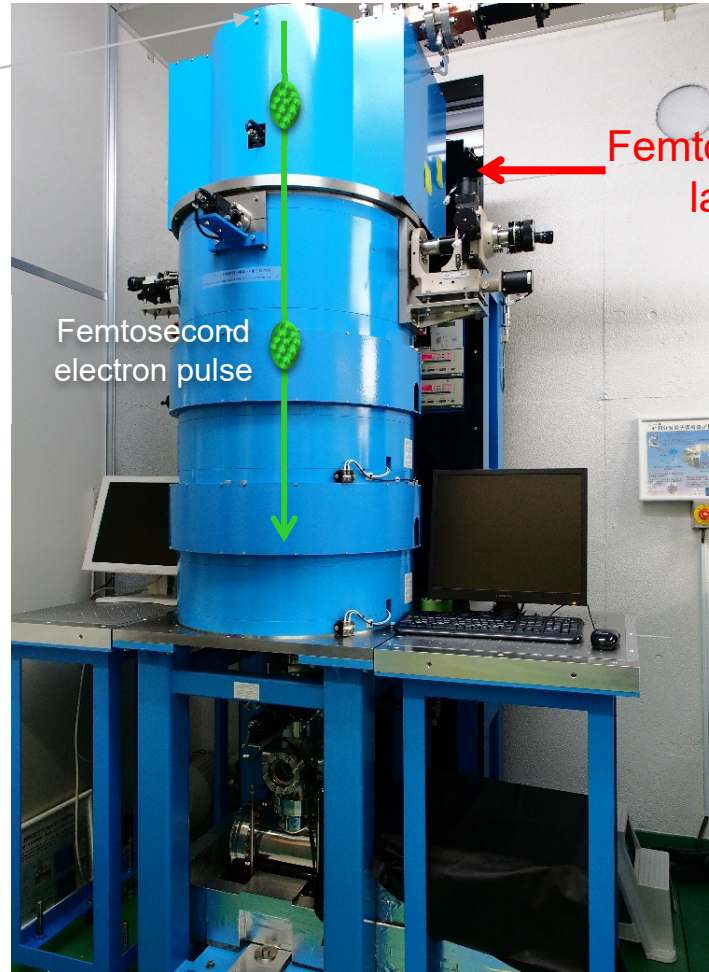
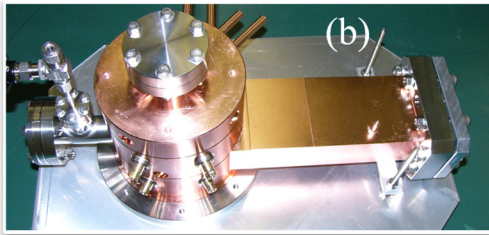
Size: 3m(H) x 0.7m(D)

First prototype of RF gun based MeV electron microscopy



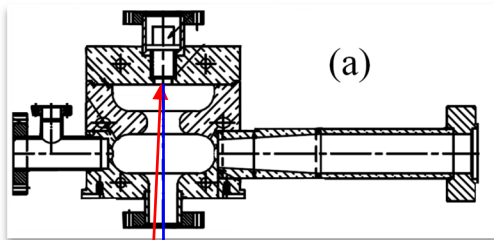
fs laser

MeV-energy
fs electron pulses

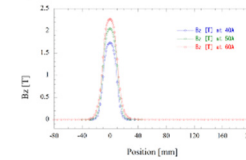
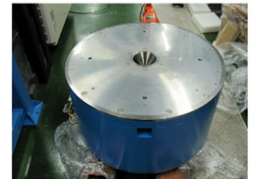
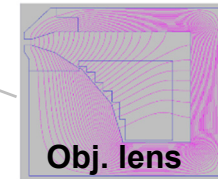
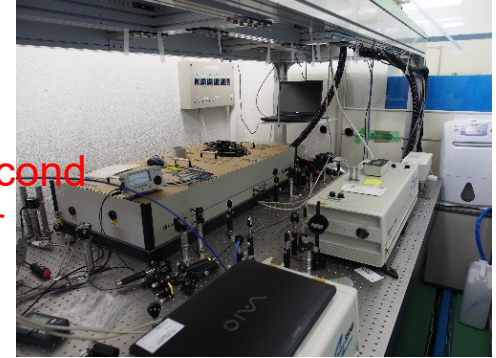
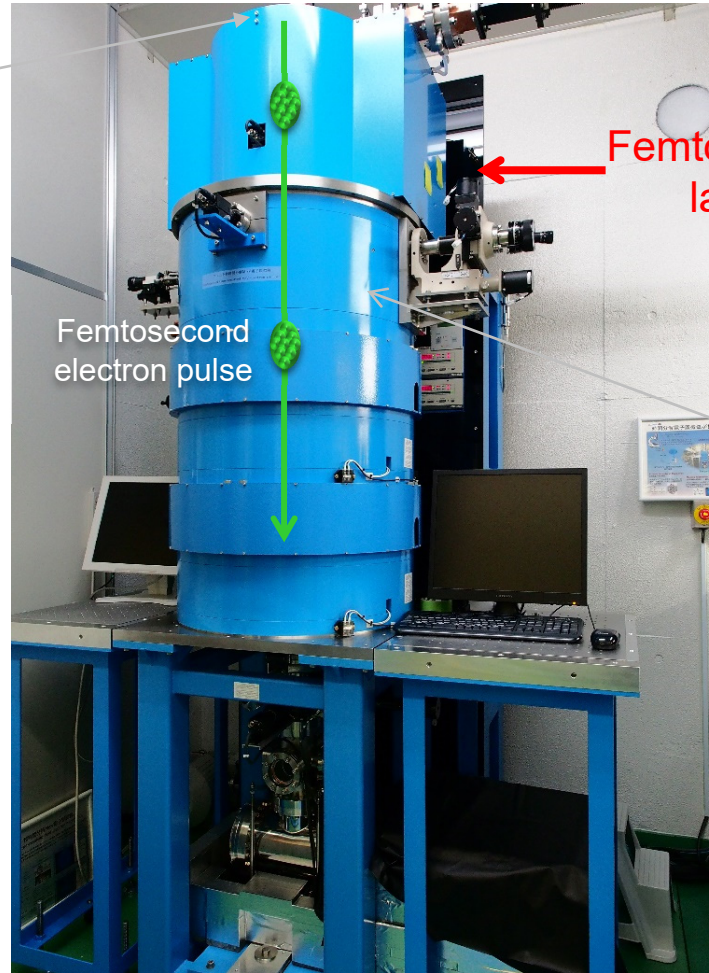
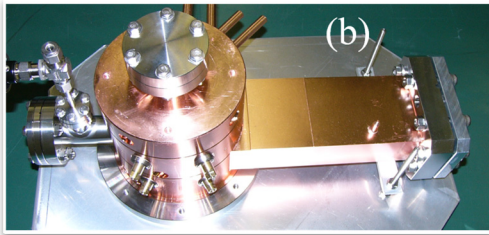


Size: 3m(H) x 0.7m(D)

First prototype of RF gun based MeV electron microscopy



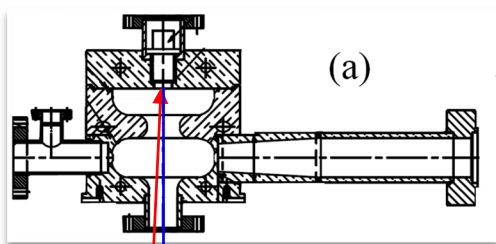
fs laser
MeV-energy
fs electron pulses



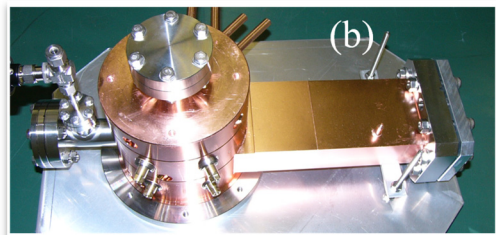
Bore diameter: 13mm
Magnetic field: 2.2Tesla
Ampere-turn: 35,000
 $f = 5$ mm for 2MeV e^-
 $C_s(C_c) \sim 4$ mm
70 cm (D) x 35 cm (H)

Size: 3m(H) x 0.7m(D)

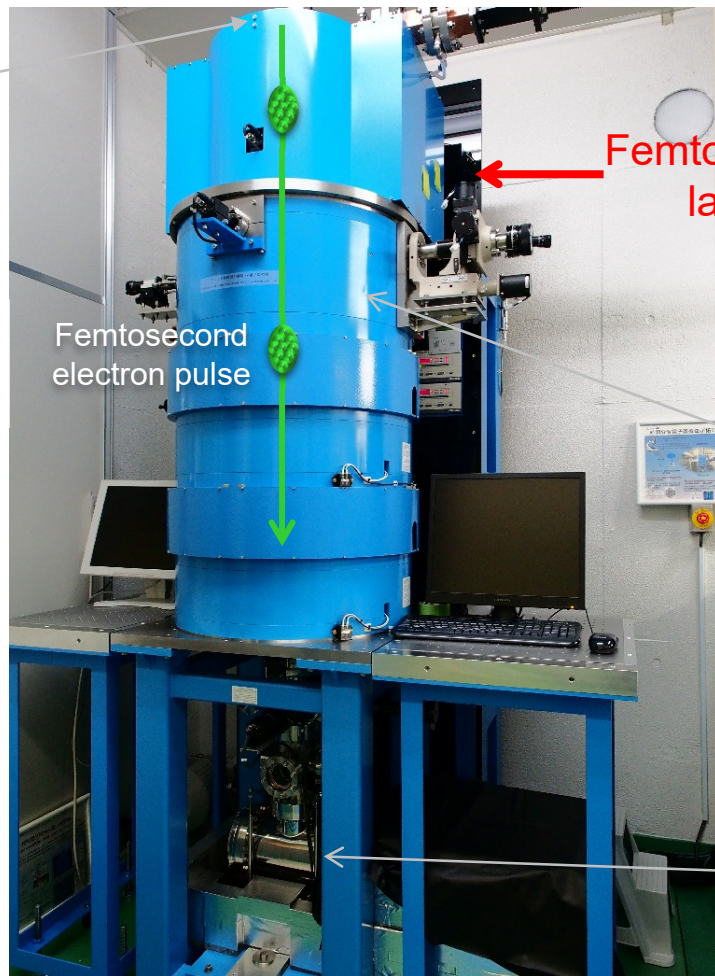
First prototype of RF gun based MeV electron microscopy



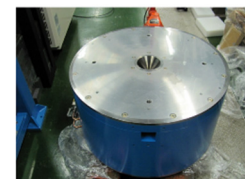
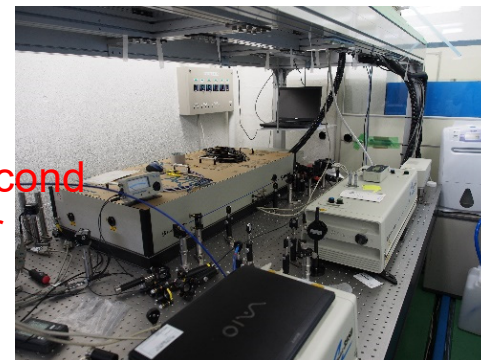
fs laser
MeV-energy
fs electron pulses



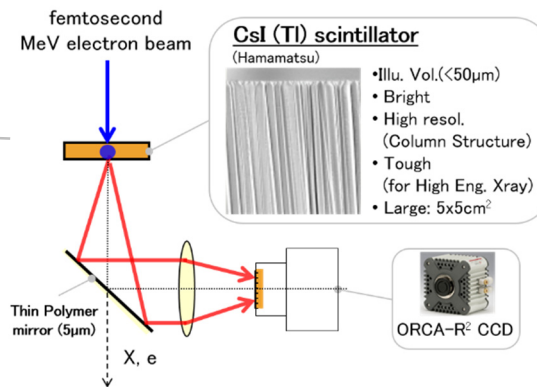
- The prototype was constructed at the end of Oct. 2012.
- The prototype was upgraded in 2016.



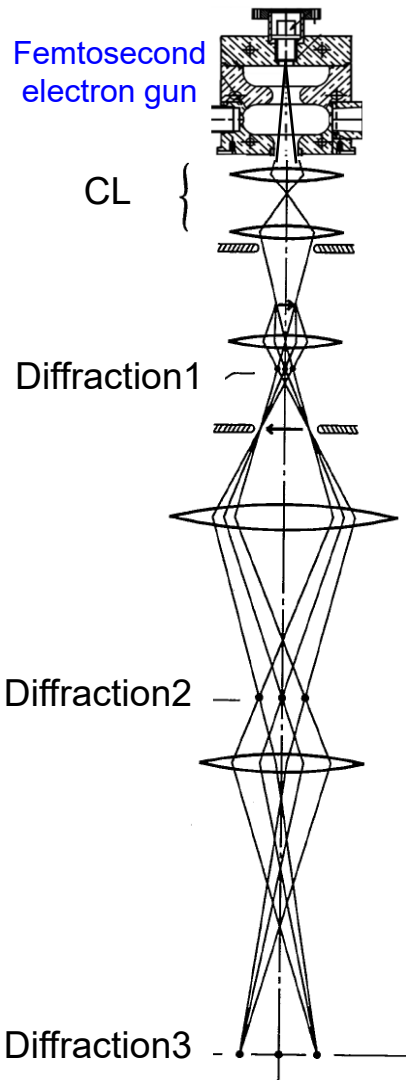
Size: 3m(H) x 0.7m(D)



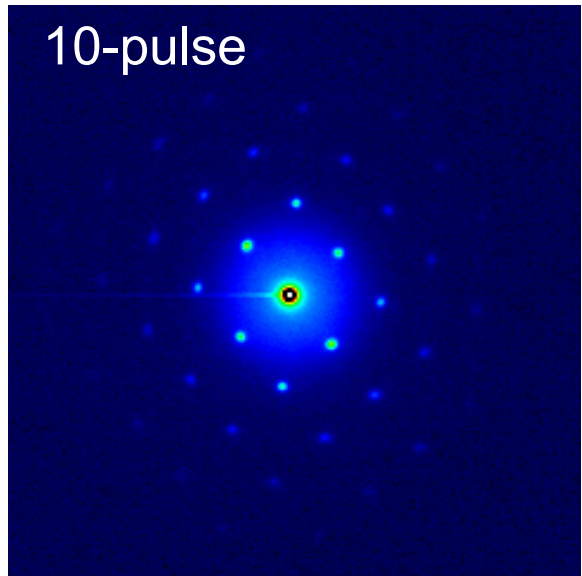
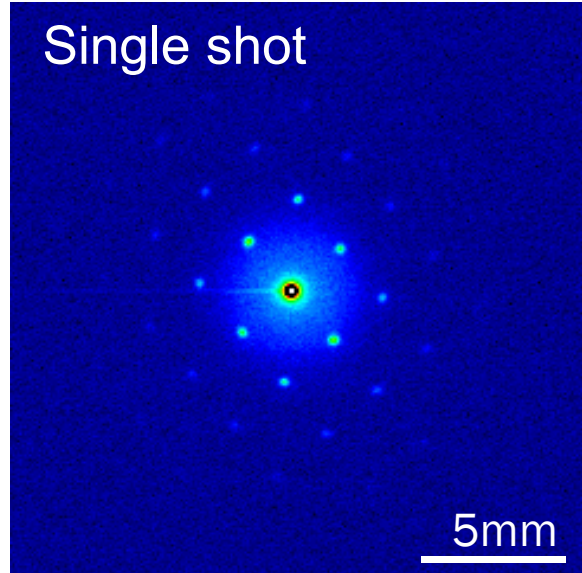
Bore diameter: 13mm
Magnetic field: 2.2Tesla
Ampere-turn: 35,000
 $f = 5 \text{ mm}$ for 2MeV e^-
 $C_s (C_c) \sim 4 \text{ mm}$
70 cm (D) x 35 cm (H)



MeV ED imaging using fs e- pulses



Diffraction measurement



Electron diffraction imaging in UEM

Sample:

Single-crystal Au

Thickness: ~10nm

Electron beam:

Energy: 3.1 MeV

Pulse length: 100 fs

e- charge: ~1pC/pulse

Results:

High-quality electron diffraction patterns can be observed with MeV-energy single e- pulse.

Ultrafast dynamics: Laser induced melting in Au

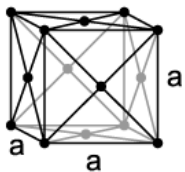
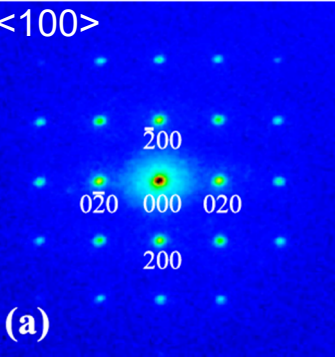
Pump laser pulse
385nm, 90fs

Single crystal Au
~10 nm

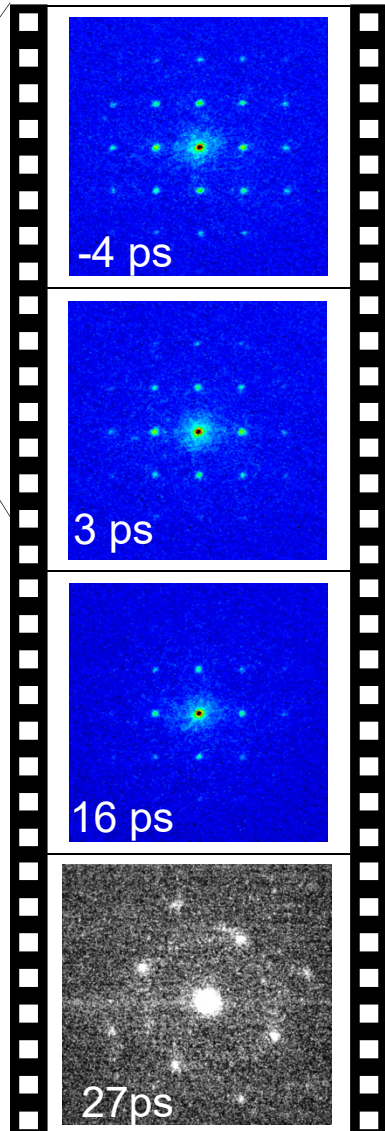
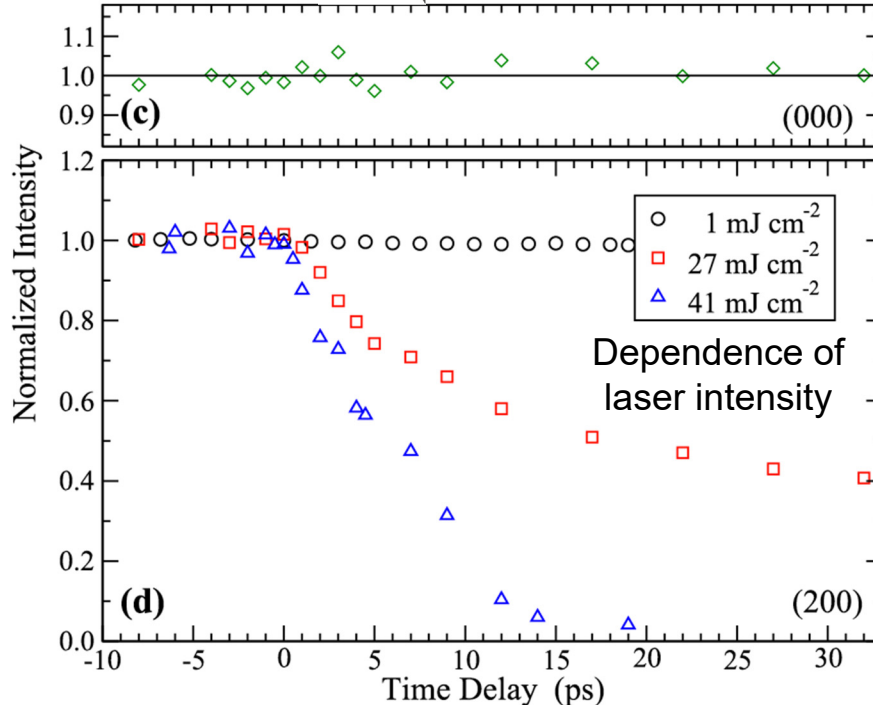
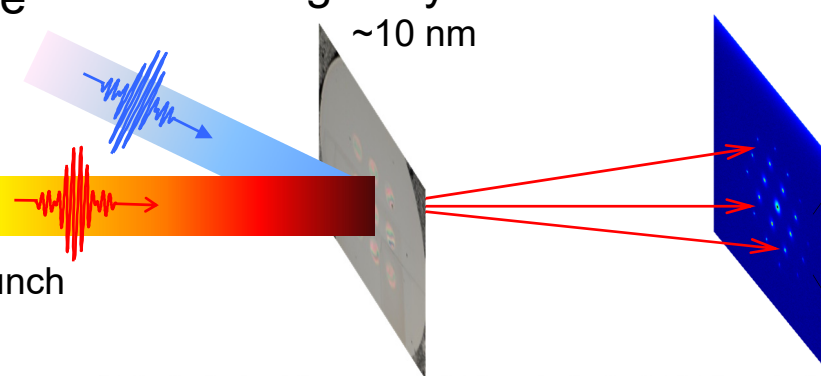
Movie of laser melting on Au

Electron pulse
3 MeV, 90fs, $10^5 e^-/\text{bunch}$

ED image of
single-crystal Au



$a = 4.07864 \text{ \AA}$

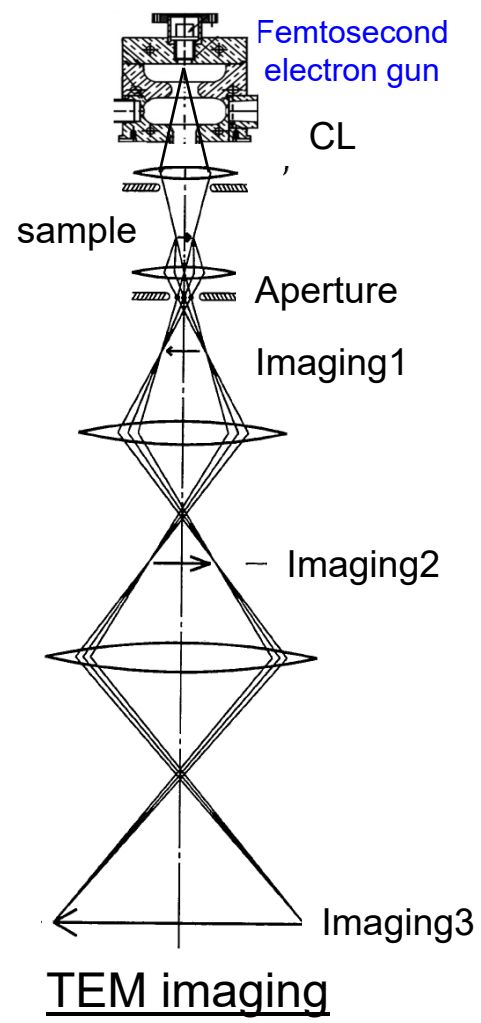


solid



liquid

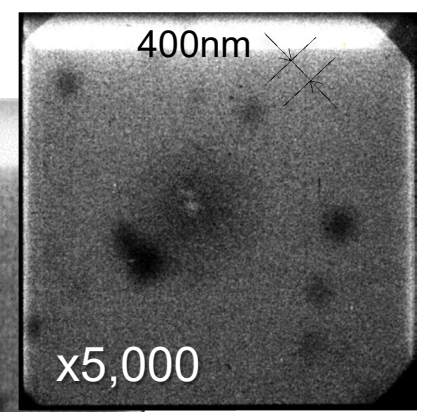
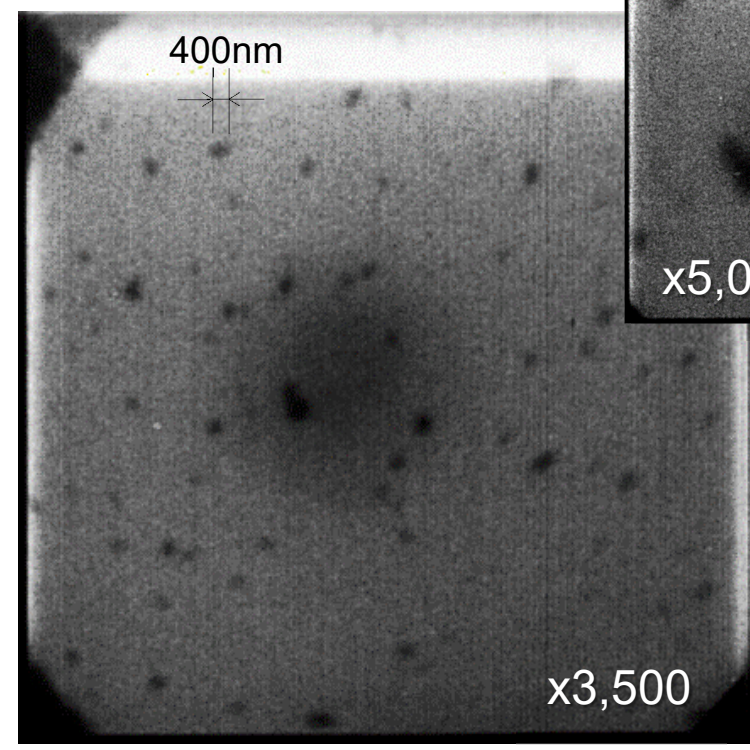
MeV EM imaging using fs e- pulse



Electron beam: 3.1 MeV, 100 fs, ~1pC/pulse

Sample: **Au nanoparticles** (diameter: 400nm)

TEM observed with 2000 pulses



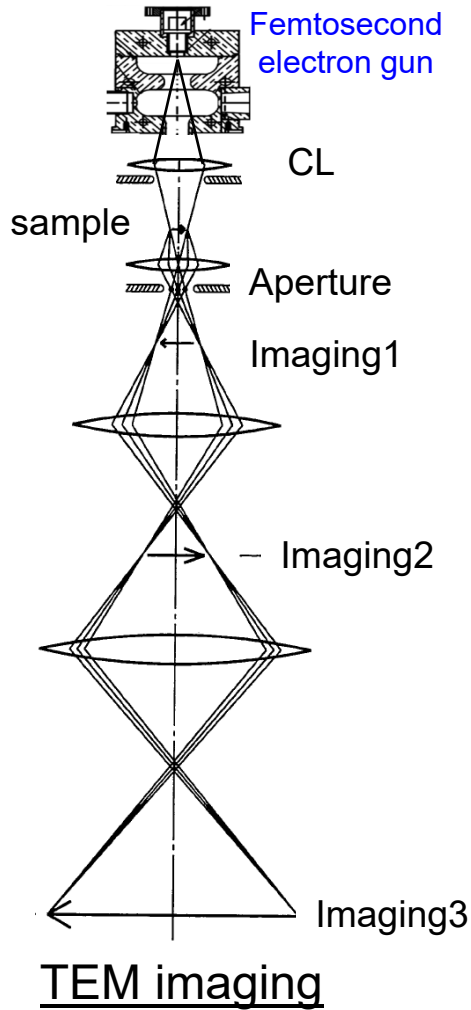
19nm/pixel

First demonstration of EM imaging using MeV femtosecond electron beam!

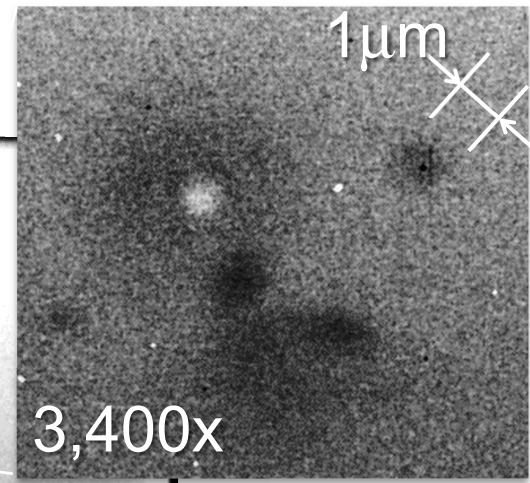
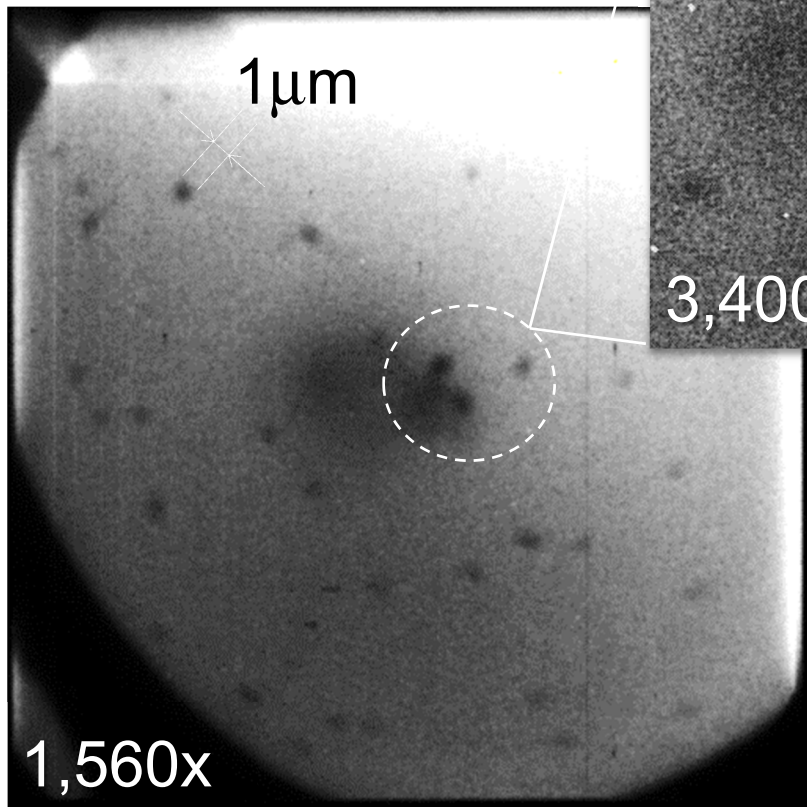
MeV EM imaging using fs e- pulse

Electron beam: 3.1 MeV, 100 fs, ~1pC/pulse

Sample: polystyrene latex particles
(diameter: 1.09 μ m)



TEM observed with 10,000 pulses



Single-shot and accumulated imaging

Dependence on No. of electron pulses

e- beam: 3.1 MeV, 100 fs, ~ 1 pC/pulse

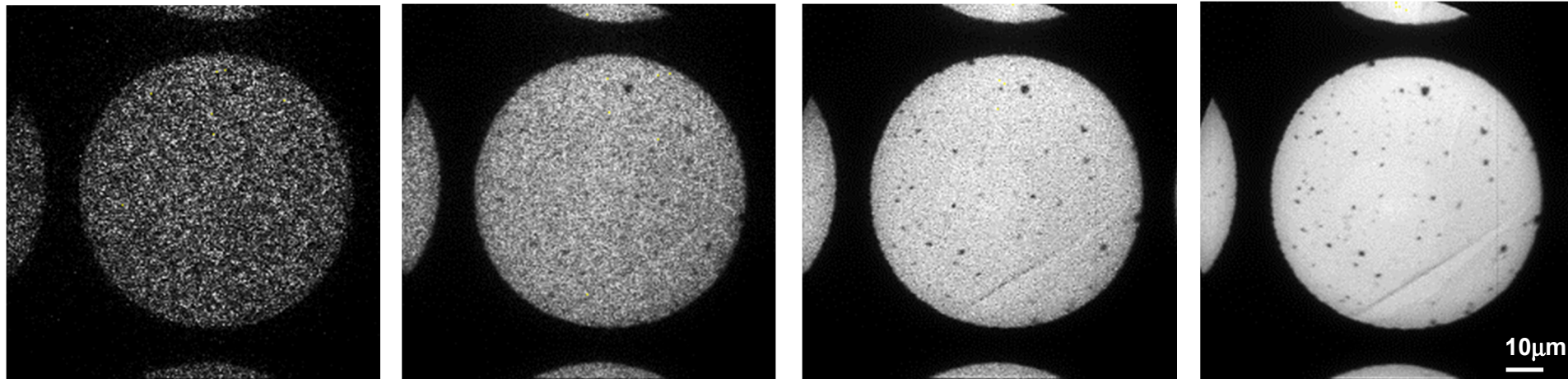
Sample: Au nano-particles (diameter: ~ 400 nm)

1-pulse

10-pulse

100-pulse

1000-pulse



- Under the low-magnification observation, the single-shot imaging using a femtosecond electron pulse is available in future.

Single-shot and accumulated imaging

Dependence on No. of electron pulses

e- beam: 3.1 MeV, 100 fs, ~1pC/pulse

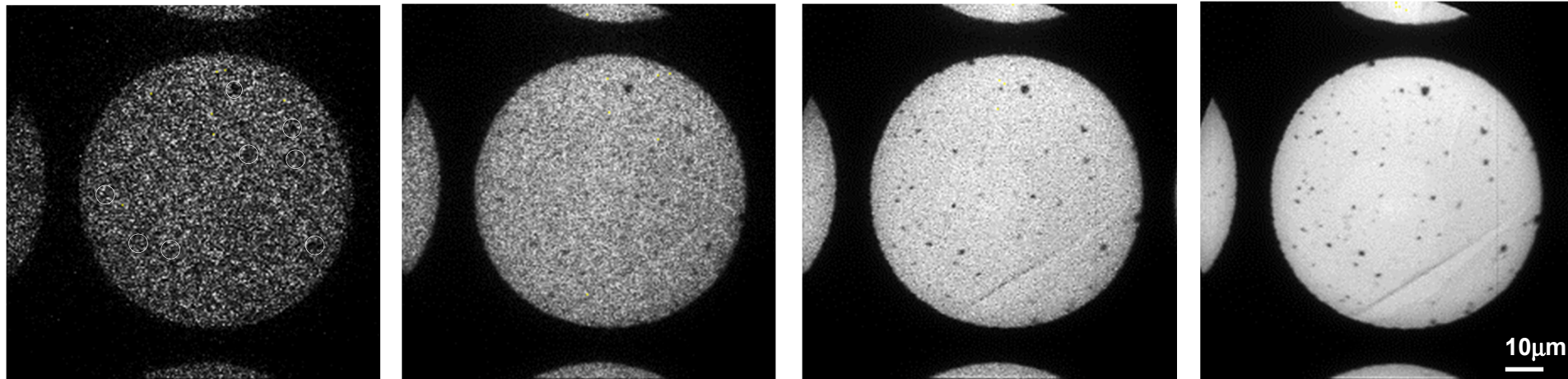
Sample: Au nano-particles (diameter: ~400nm)

1-pulse

10-pulse

100-pulse

1000-pulse



- Under the low-magnification observation, the single-shot imaging using a femtosecond electron pulse is available in future.

Conclusions

- RF gun based ultrafast electron microscopy (UEM) have been constructed at Osaka University.
 - In UED exp., single-shot and time-resolved measurements have been succeeded. The time resolution was achieved to be ~ 100 fs.
 - In UEM exp., the demonstrations of MeV electron imaging were carried out.
- Both experiments suggest that RF gun is very useful for ultrafast 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 ($< 0.1 \mu\text{m}$) and energy spread (10^{-5} or less),
- increase the beam brightness overcoming space-charge effect,
- improve the stabilities on the charge and energy,
- improve the spatial resolution, ...

Acknowledgment

- (Osaka Univ.) R. Asakawa, K. Kan, T. Kondoh, M. Gohdo, Y. Yoshida
H. Yoshida, S. Taketa, K. Tanimura
- (KEK) T. Takatomi, N. Terunuma, J. Urakawa
- (Shiga Univ.) N. Naruse
- (UCL) Y. Giret, A. Shluger

Work is supported by Grant-in-Aid from MEXT Japan for
Scientific Research(A): Grant No. 22246127 (2010~2012);
Scientific Research(A): Grant No. 26246026 (2014~2016);
Challenging Research: Grant No. 16K13687 (2016~2017);
Scientific Research(A): Grant No. 17H01060 (2017~2019)

