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FEMTOSECOND ELECTRON RF GUNS FOR ULTRAFAST ELECTRON DIFFRACTION

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Outline

1. Introduction
2. Femtosecond RF guns

   Requirements / Limitations / experimental results
3. RF gun based MeV ultrafast electron diffraction (UED)

   First MeV UED experiment at SLAC

   MeV UED facilities around world

   development of RF gun based MeV electron microscopy
4. Concluding remarks

Introduction

Ultrafast dynamic processes

Ultrafast dynamic processes in materials, i.e.

Phase transition/structure transformation in solid state,

Chemical reactions in molecules,

Energy transfer in biology, and so on,

are occurred on femtosecond time scales over nanometer (even atomic) spatial dimensions.

- The direct observation of such ultrafast dynamic processes has long been a goal in science.
- Ultrafast techniques with femtosecond time resolution are required.

Ultrafast Techniques

1） Ultrafast X-ray diffraction/image

Picosecond X-ray pulses from SR & femtosecond X-ray pulses from FEL or laser plasmas acceleration have been used.

→ big experiment/measurement,
large energy deposited → large damage to specimens.

2） Ultrafast electron diffraction (UED)

A fs laser pulse is used as pump, while a fs or ps e^- bunch is used as probe

Short keV e^- beam with pulse length of 400-600 fs have be used in low-energy UED.

Recently, the time resolution has been achieved to 100 fs or less using RF gun.

3） Ultrafast electron microscopy (UEM)

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

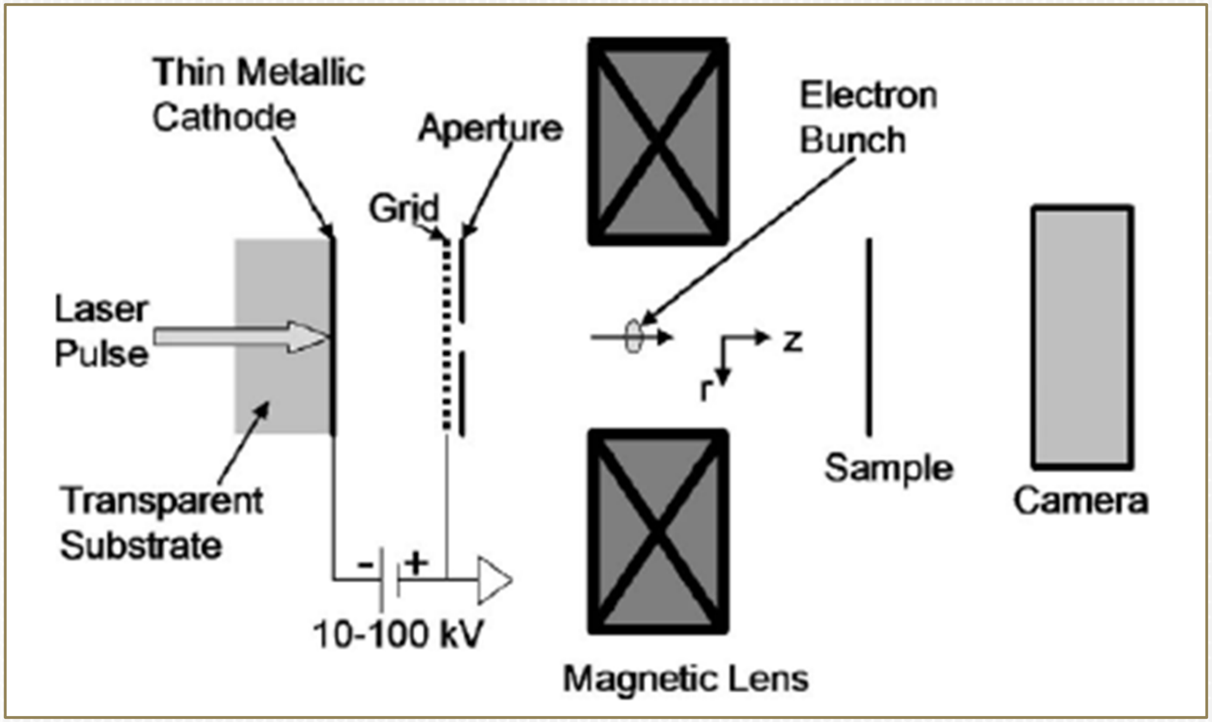
→ *Recently, the resolution of ns-nm or ps-μm has been achieved in 100-keV TEM.*

MeV UEM using RF gun is being developed at Osaka University.

2. Femtosecond RF gun

Why RF gun in UED & UEM?

Most of UED systems are used the photocathode-based DC gun.



✓Beam energy : 30~100keV（Max. DC field=10~12 MV/m）

✓Bunch length : 400~600fs at e^- number of 10^3~10^4 e^-/pulse

Why RF gun in UED & UEM?

Problem#1: Strong space-charge force in low-energy e^- bunch.

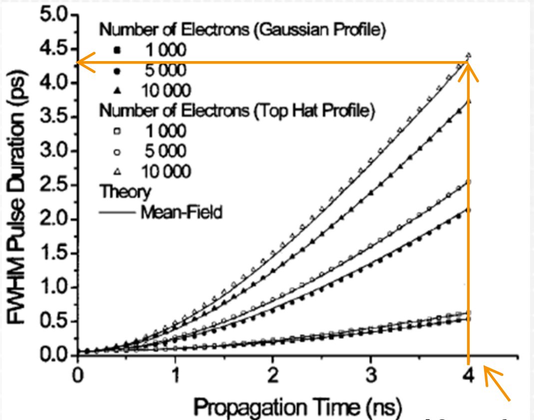
1) Increase of bunch length during beam transport

$$\frac{d^2 l}{dt^2} = \frac{Ne^2}{m\varepsilon_0 \pi r^2}\left(1 - \frac{l}{\sqrt{l^2+4r^2}}\right)$$

l: pulse length, N: number of e^-,
t: propagation time
r: electron beam radius

If we transport a 30 keV e- beam to a distance of 40 cm, the bunch length is increased from fs to a few ps.

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

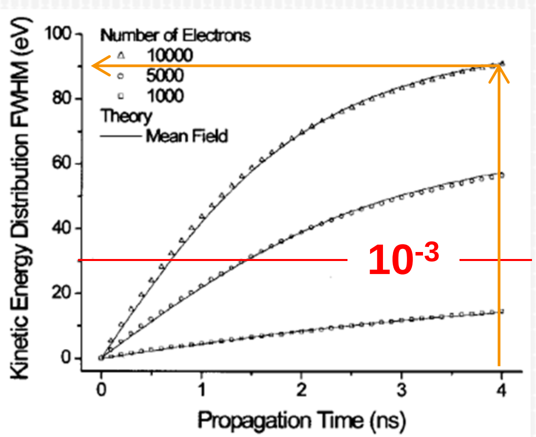


40cm for 30KeV

2) Increase of energy spread during beam transport

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

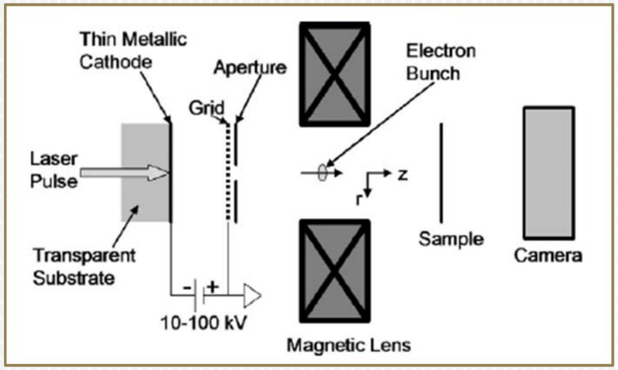
$\Delta E/E \rightarrow 3\times10^{-3}$ for transporting to 40 cm.



It is difficult to obtain a 100 fs e- bunch with energy spread of $\Delta E/E<10^{-3}$ using DC guns.

Why RF gun in UED & UEM?

Problem#2: Small number of electrons.



To reduce the space-charge effect in low-energy UED system,

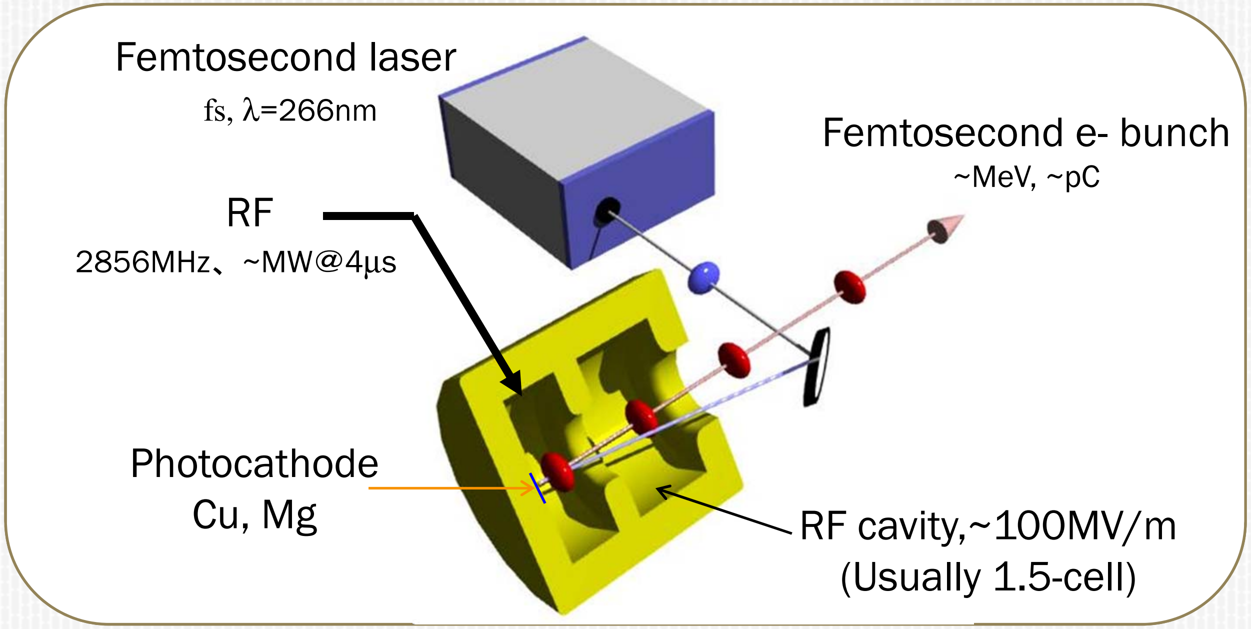
- minimize the distance between sample and cathode: 4~5 cm
- decrease the number of electrons in bunch

400 fs at 1,000 e^-/bunch

✓It is difficult to observe the ultrafast dynamics with single-shot measurement.
✓The studies of low-energy UED are limited to the reversible processes.

Photocathode RF gun is a good choice
to generate a 100 fs e- beam with large e^- number in bunch.

Femtosecond photocathode RF gun



The expected beam parameters:

Bunch length :	100 fs
Beam energy :	1~3 MeV
Emittance :	~0.1mm-mrad
Energy spread :	~ 10^{-4} (10^{-5} for challenge)
e- number :	$10^{7\text{-}8}$ e^{-}'s/bunch (1~10 pC)

Beam dynamics in RF gun

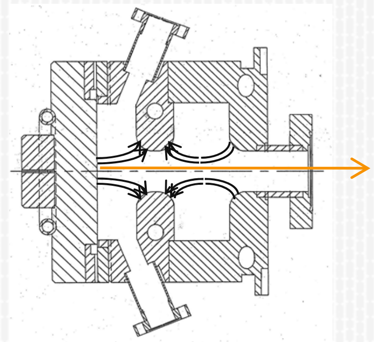
1) Longitudinal dynamics

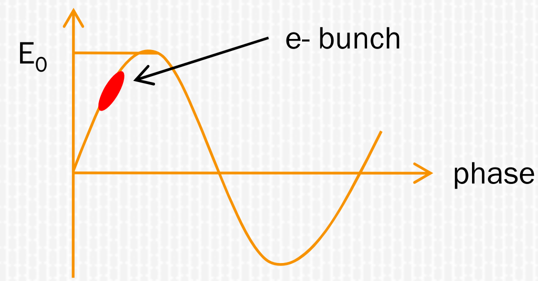
RF field in z-axial :

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

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

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





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

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

$$\sigma_{\Delta\gamma}(rms) = 2\pi\alpha f \sigma_z = 2.9\times 10^{-4} E_0[MV/m]\sigma_z[ps]$$

$$\frac{\Delta E}{E}(rms) = \frac{\sigma_{\Delta\gamma}}{\gamma - 1} = 2\times 10^{-6}\frac{f[MHz]\sigma_z[ps]}{n+0.5}$$

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

<u>Example:</u>

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=200μm, σ_z=100fs

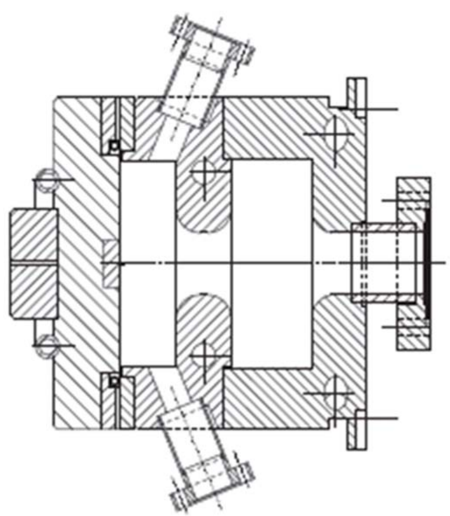
ε^{sc} < 0.1 mm-mrad

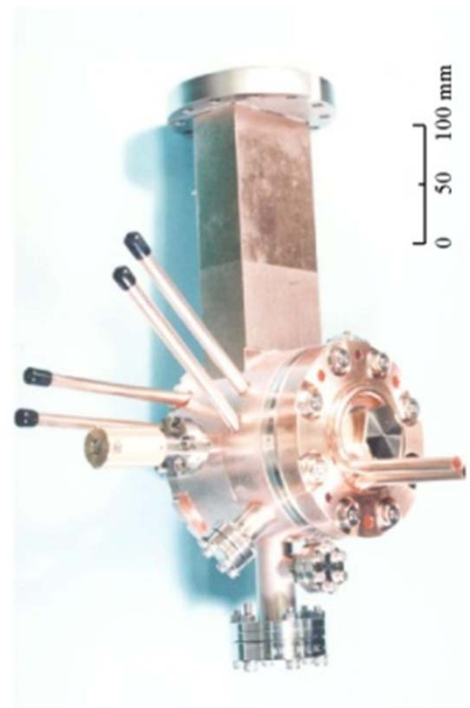
ε^{rf} ~ negligible

New femtosecond RF gun at Osaka Univ.

➢A typical 1.6-cell S-band RF gun (BNL type gun-IV) is used in the most of MeV UED facilities.

<u>BNL type gun-IV</u>

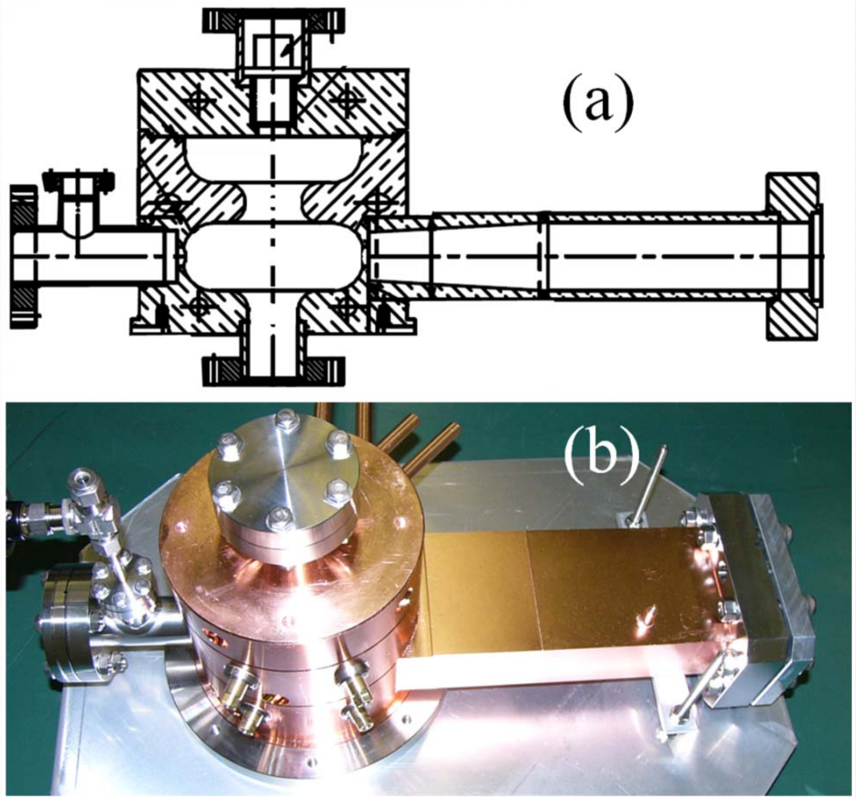




New femtosecond RF gun at Osaka Univ.

➢A typical 1.6-cell S-band RF gun (BNL type gun-IV) is used in the most of MeV UED facilities.

➢To reduce dark current and make a high-quality RF cavity, a new femtosecond RF gun was developed in 2010 at Osaka Univ. under the collaboration with KEK.



Improvements:

- **remove two laser injection ports**
- **a new turner system**
- **new structure cavities**
- **a new insertion function of photo-cathode**
  (The photocathode is removable)

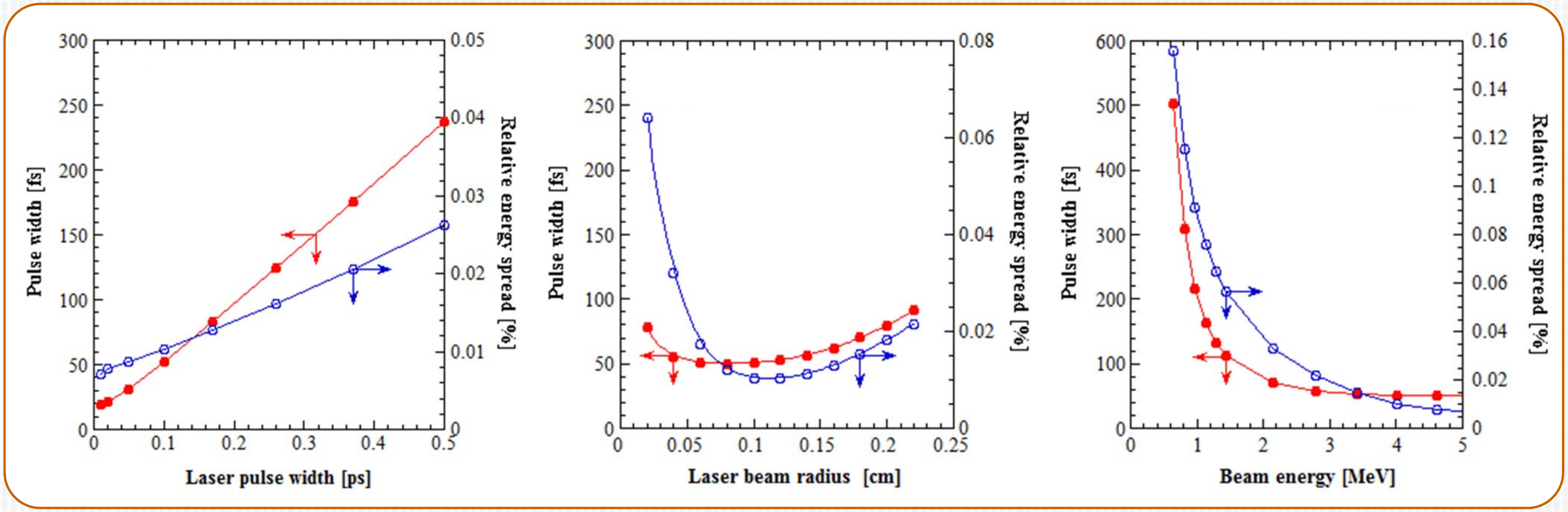
The mode separation between π mode and 0 mode is increased up to 8.5 MHz.

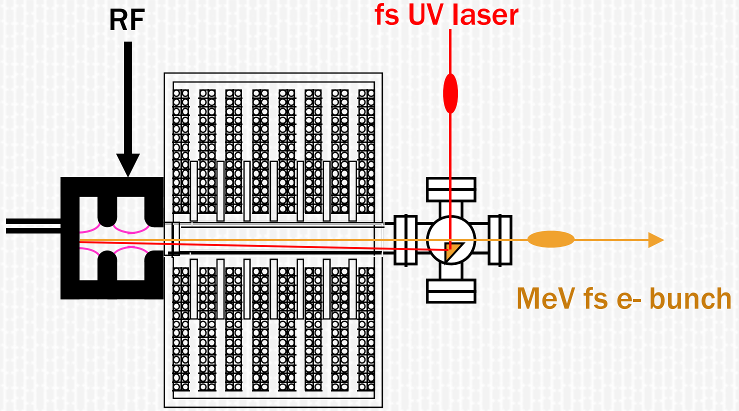
The Q value is increased up to 14,500.

Femtosecond e⁻ bunch generated from RF gun

Simulation studies of low-charge e-beam in RF gun

(Q=0.1pC/bunch)



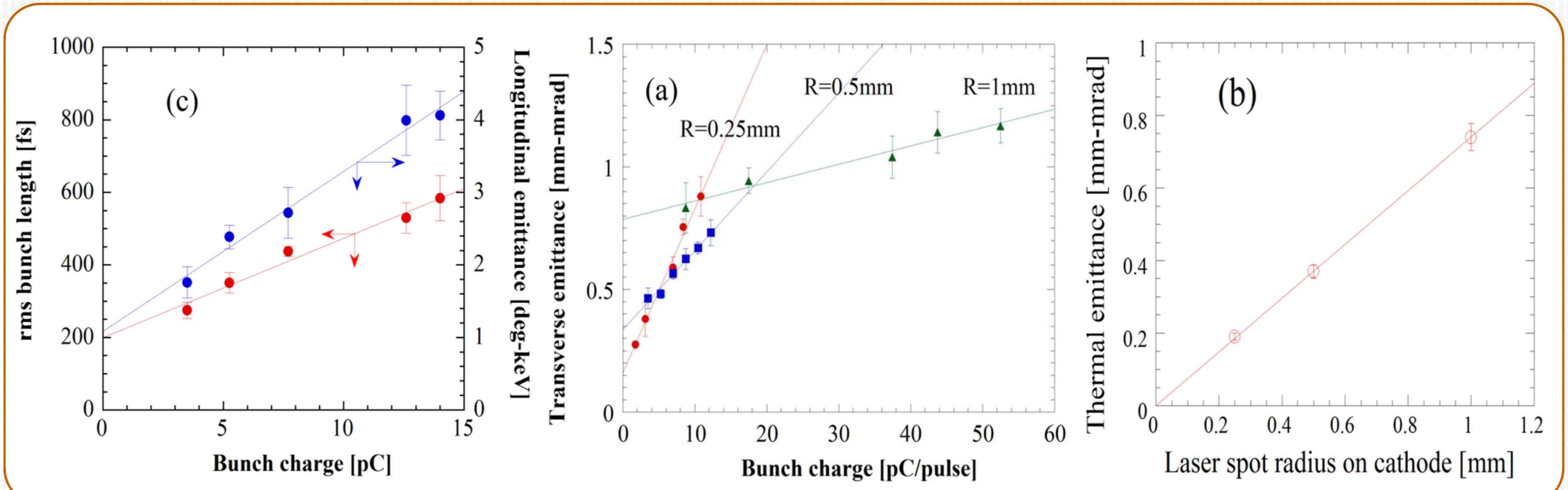


- At ≥2 MeV, we can obtain a 100 fs e^- bunch with energy spread of 10^{-4}.
- The laser spot size does not effect the bunch length and energy spread at low-charge.

Femtosecond e$^-$ bunch generated from RF gun

<u>Experiment studies of femtosecond e- beam in RF gun</u>

(exp. conditions: 200 fs UV laser, 30º gun phase, 3MeV)



- The bunch length, longitudinal and transverse emittance of femtosecond e- beam are dominated by the bunch charge, if we increase to >1 pC.
- The thermal emittance at Cu cathode increases linearly with the laser spot size.

- For a copper cathode, the thermal emittance can be

$$\varepsilon_{th} = \sigma_r \sqrt{\frac{E_{kin}}{m_0 c^2}} \cong 0.18mm - mrad$$

$$E_{kin} = h\nu - \phi + \alpha\sqrt{\beta E_0 \sin\theta} \cong 0.26eV$$

- Reducing laser spot size to 0.1mm,

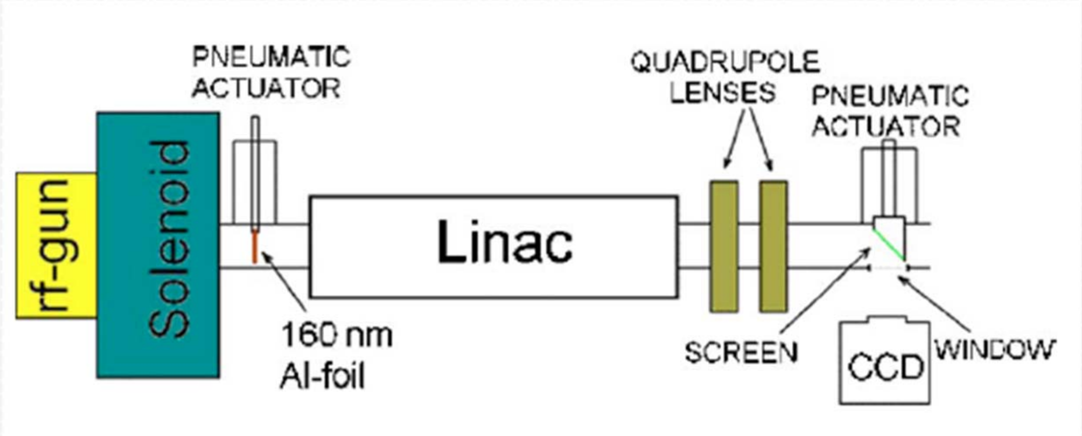
$$\varepsilon_x \leq 0.1mm - mrad$$

It is possible to generate a 100fs e- bunch with energy spread of 10^{-4} and emittance of 0.1 μm using RF gun.

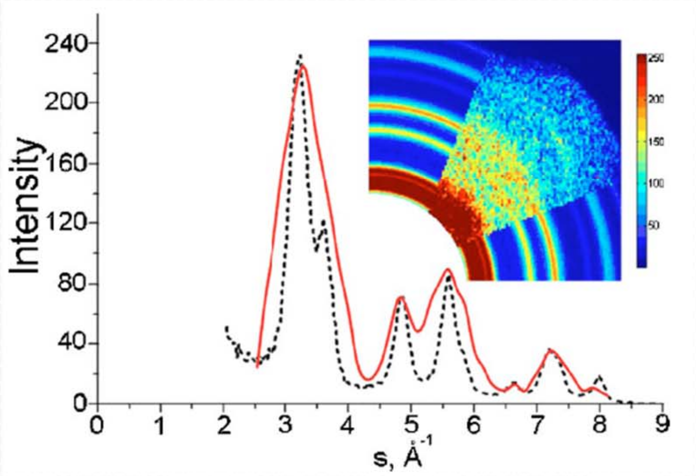
3. RF gun based MeV electron diffraction

First UED demonstration using RF gun

First MeV UED experiment at SLAC in 2006: Hastings, *et al.* APL 89, 2006



MeV e[-] diffraction from 160-nm Al



Beam energy: 5.4MeV

Bunch charge: 2.9pC

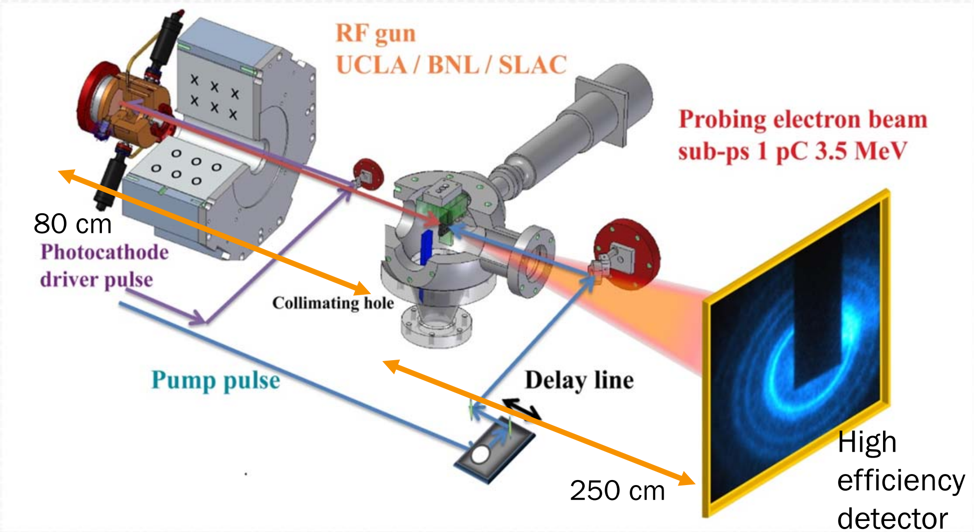
Emittance: 0.85mm-mrad

Energy spread: 0.65%

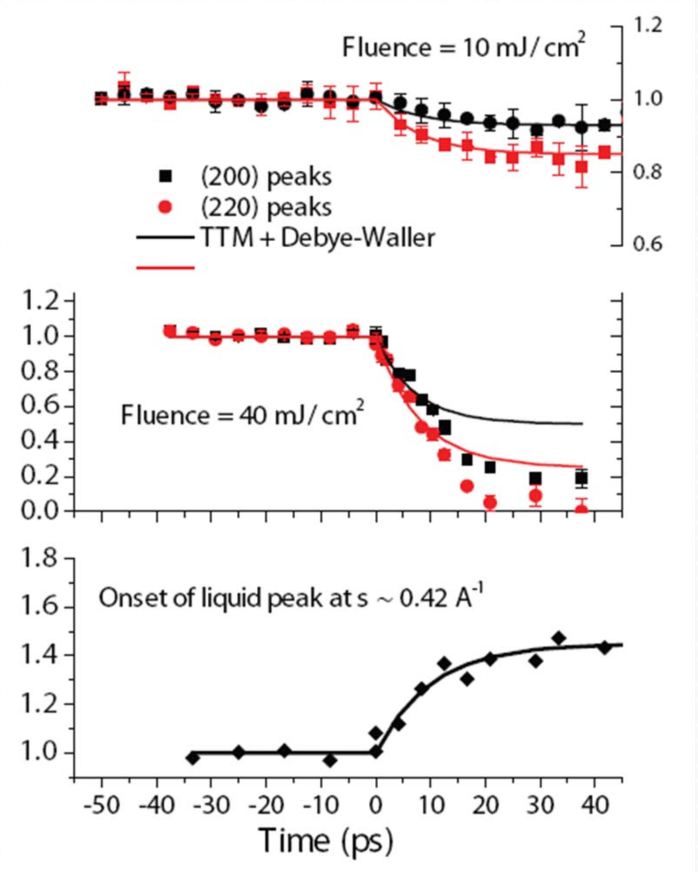
RF gun based MeV UED in UCLA

Courtesy of Pietro Musumeci

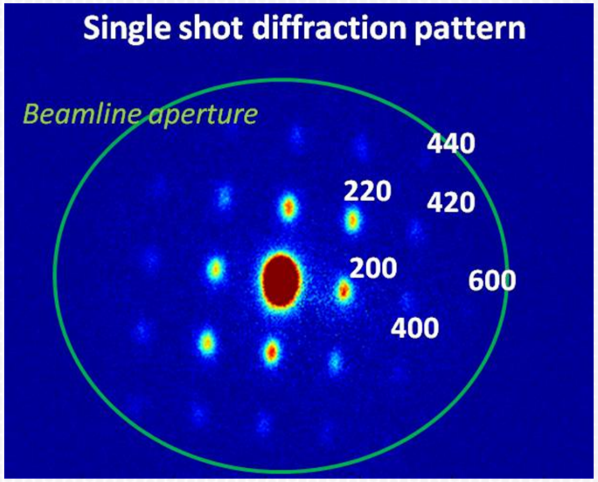
Schematics of UCLA Pegasus setup



Laser induced heating and melting of single crystal gold samples (APL, 2010)



- ✓80 MV/m field in RF gun,
- ✓3.5 MeV electron energy,
- ✓10^6-10^7 e-'s in bunch
- ✓<100 fs at sample
- ✓Single-shot meas.
- ✓RF streaking cavity

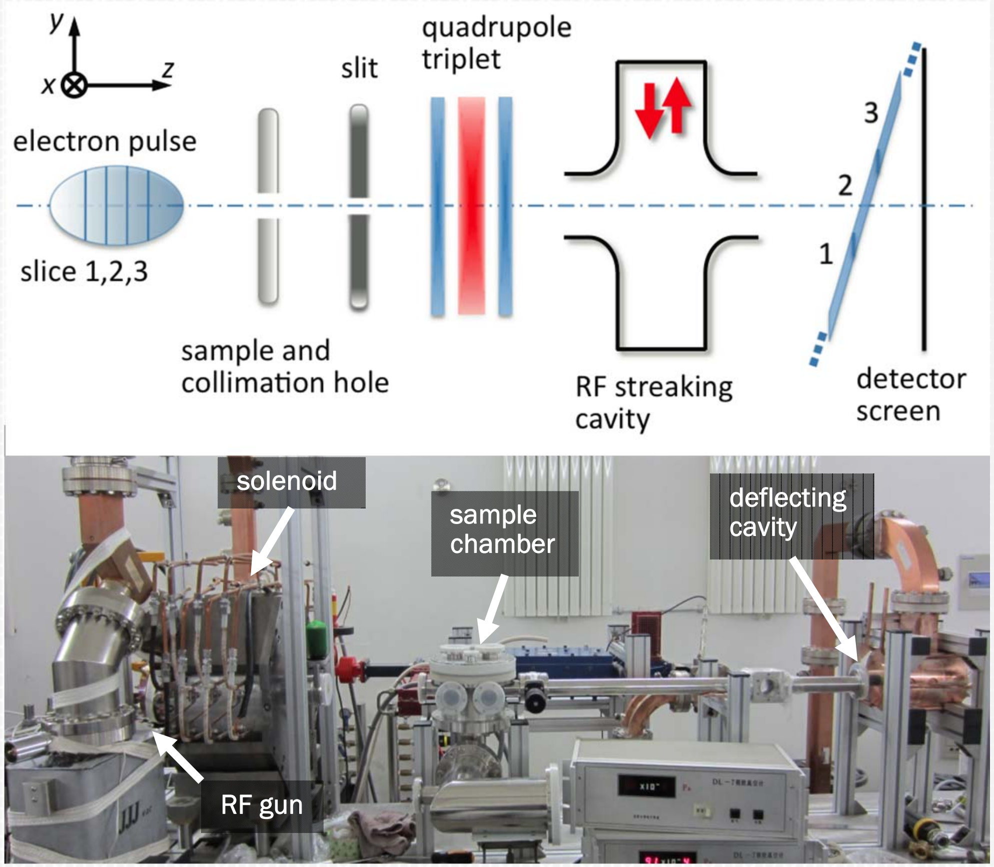


Two-temperature model tests. Electron-phonon coupling constant.

RF gun based MeV UED at Tsinghua Univ.

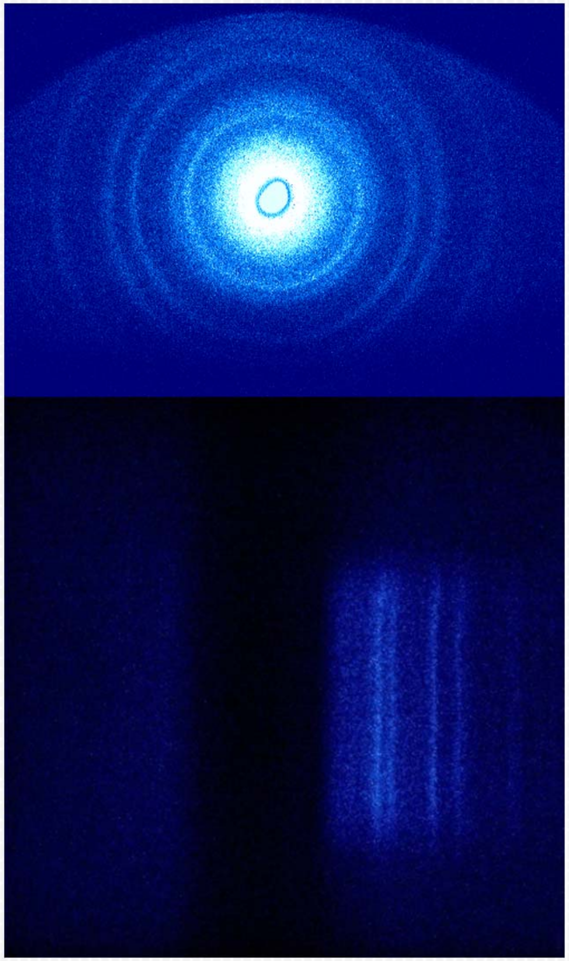
Courtesy of W. H. Huang, C. X. Tang

RF gun based UED at Tsinghua Univ. since 2009



- ✓2.5 MeV electron energy, 2.8 mm-mrad,
- ✓3.6x10^6 e-'s in bunch (0.58 pC)
- ✓Single-shot measurement
- ✓Time-resolved measurement using RF streaking cavity technique

Laser heating and melting of gold samples

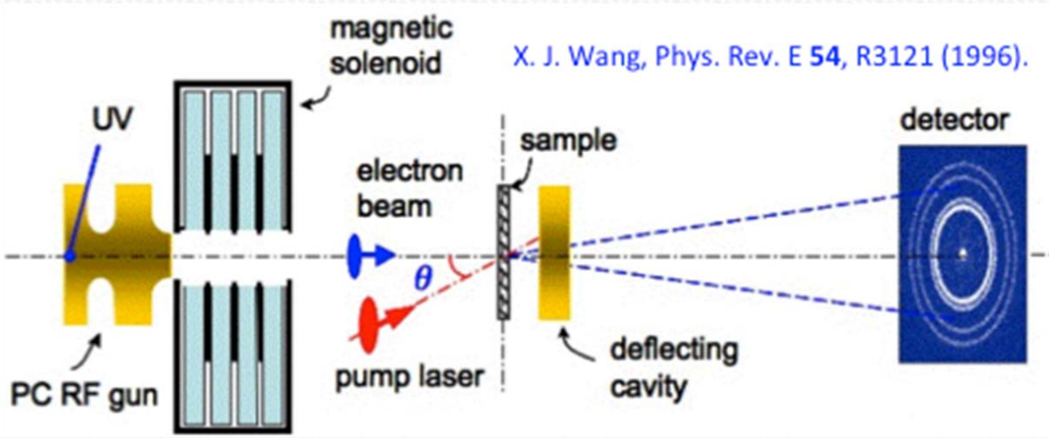


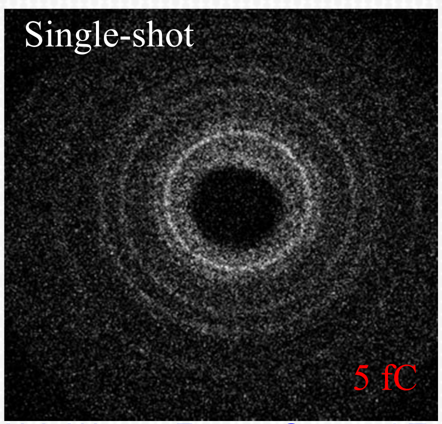
Measured with RF streaking cavity

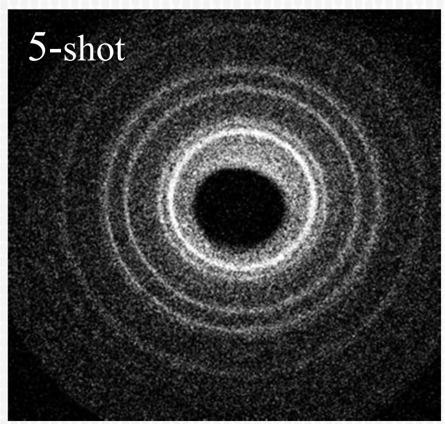
RF gun based MeV UED in BNL/Tuaotong Univ.

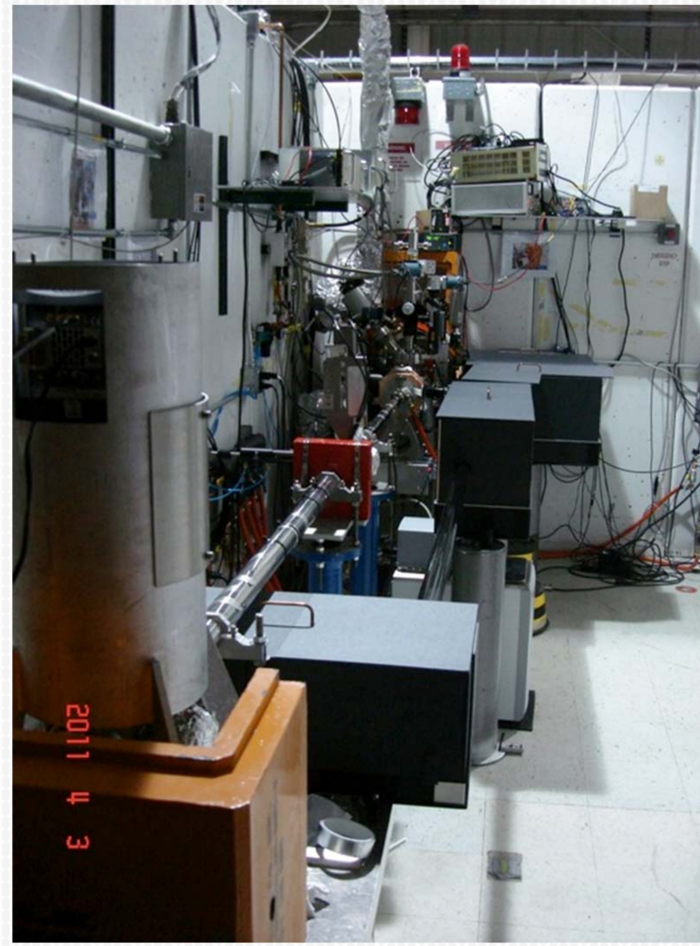
Courtesy of X. J. Wang

Schematics of UED in BNL/Shanghai Juaotong Univ.









- X.J. Wang, Femto-Second Transmission Electron Microscope Based on Photocathode RF Gun, BNL LDRD 01-39(2000).
- X. J. Wang et al, PAC'03, 420-422 (2003).

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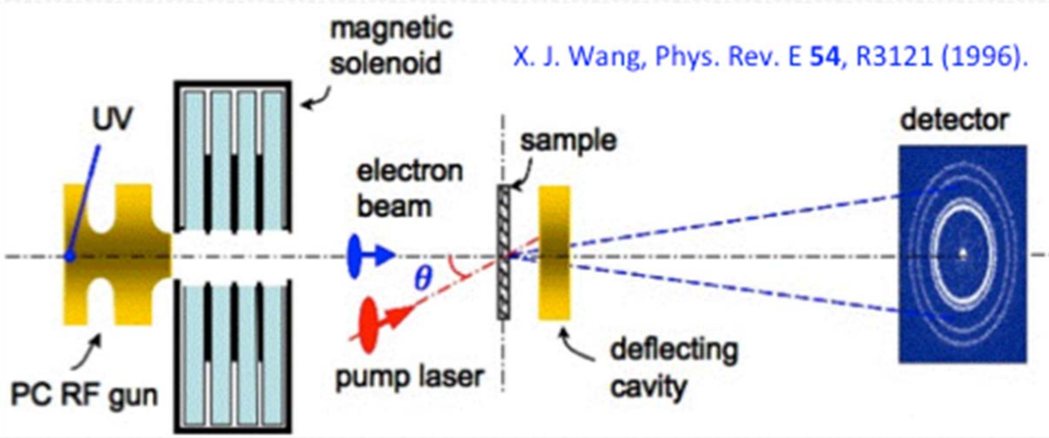
上海交通大學 SHANGHAI JIAO TONG UNIVERSITY

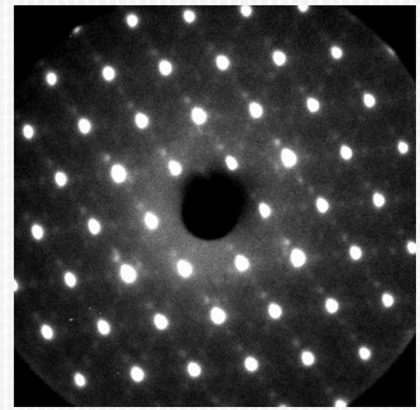
RF gun based MeV UED in BNL/Tuaotong Univ.

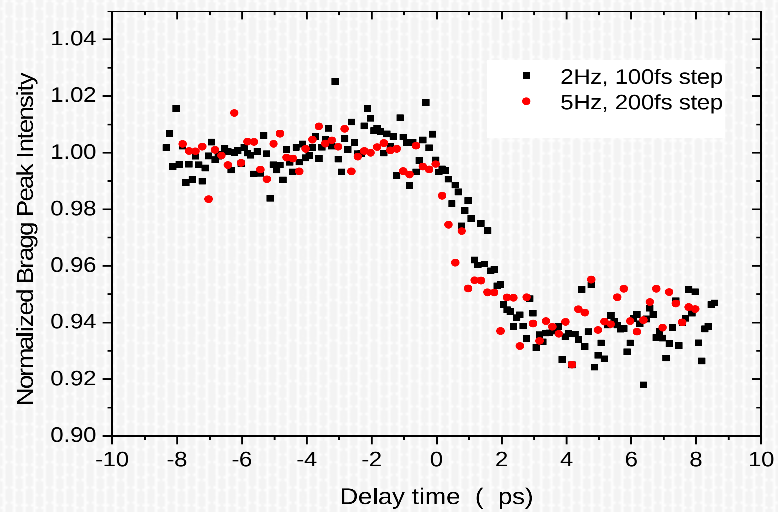
Courtesy of X. J. Wang

<u>Schematics of UED in BNL/Shanghai Juaotong Univ.</u>



$TaSe_2$ Super-Lattice





- ✓ Single-shot diffraction with 5 fC.
- ✓ Signal to noise ratio > 200.
- ✓ Timing jitter ~ 100 fs
- ✓ Pump-probe experiment with ~100s fs time resolution.

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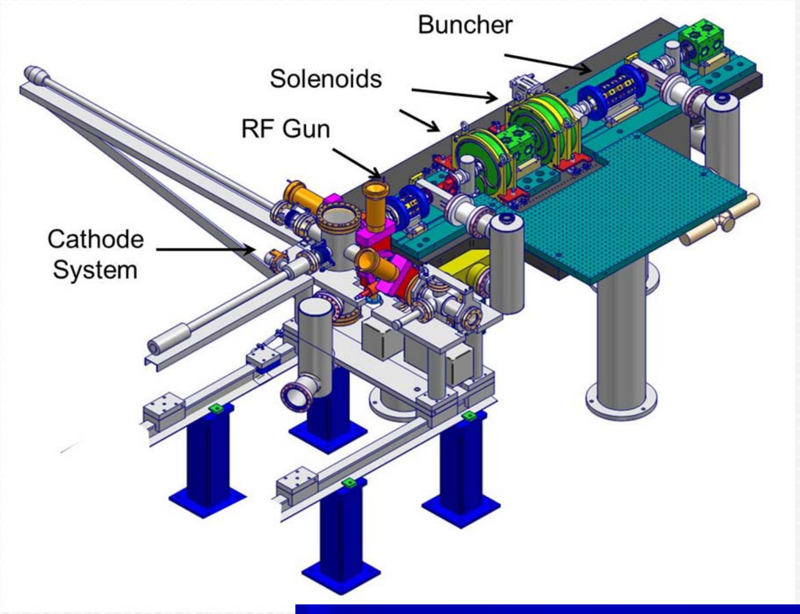
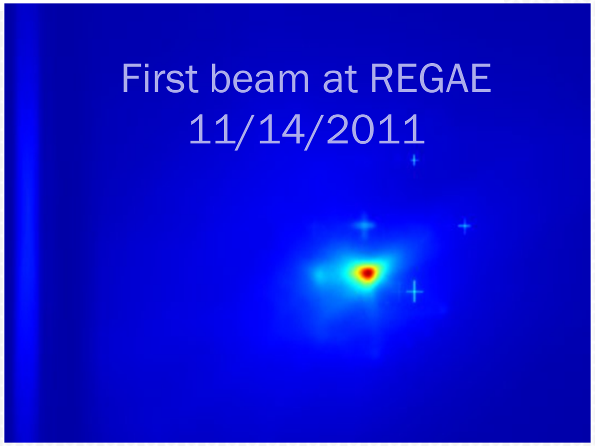
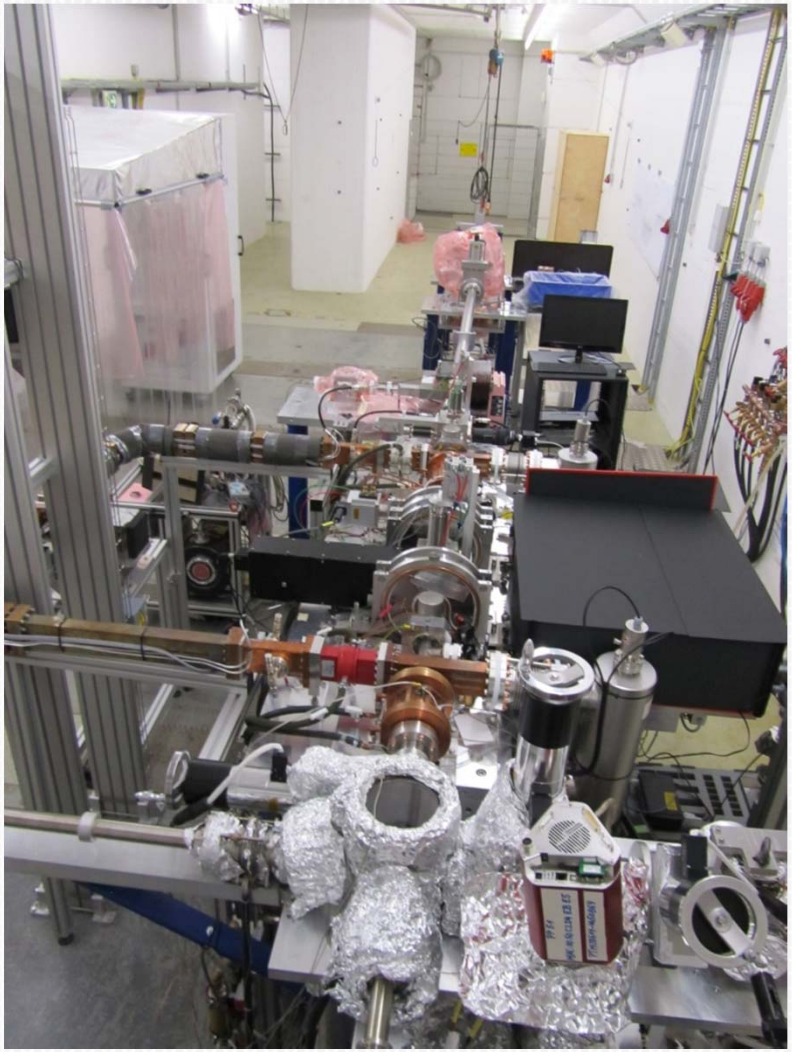
RF gun based MeV UED in DESY

Courtesy of Klaus Floettmann

REGAE: The Relativistic Electron Gun for Atomic Exploration

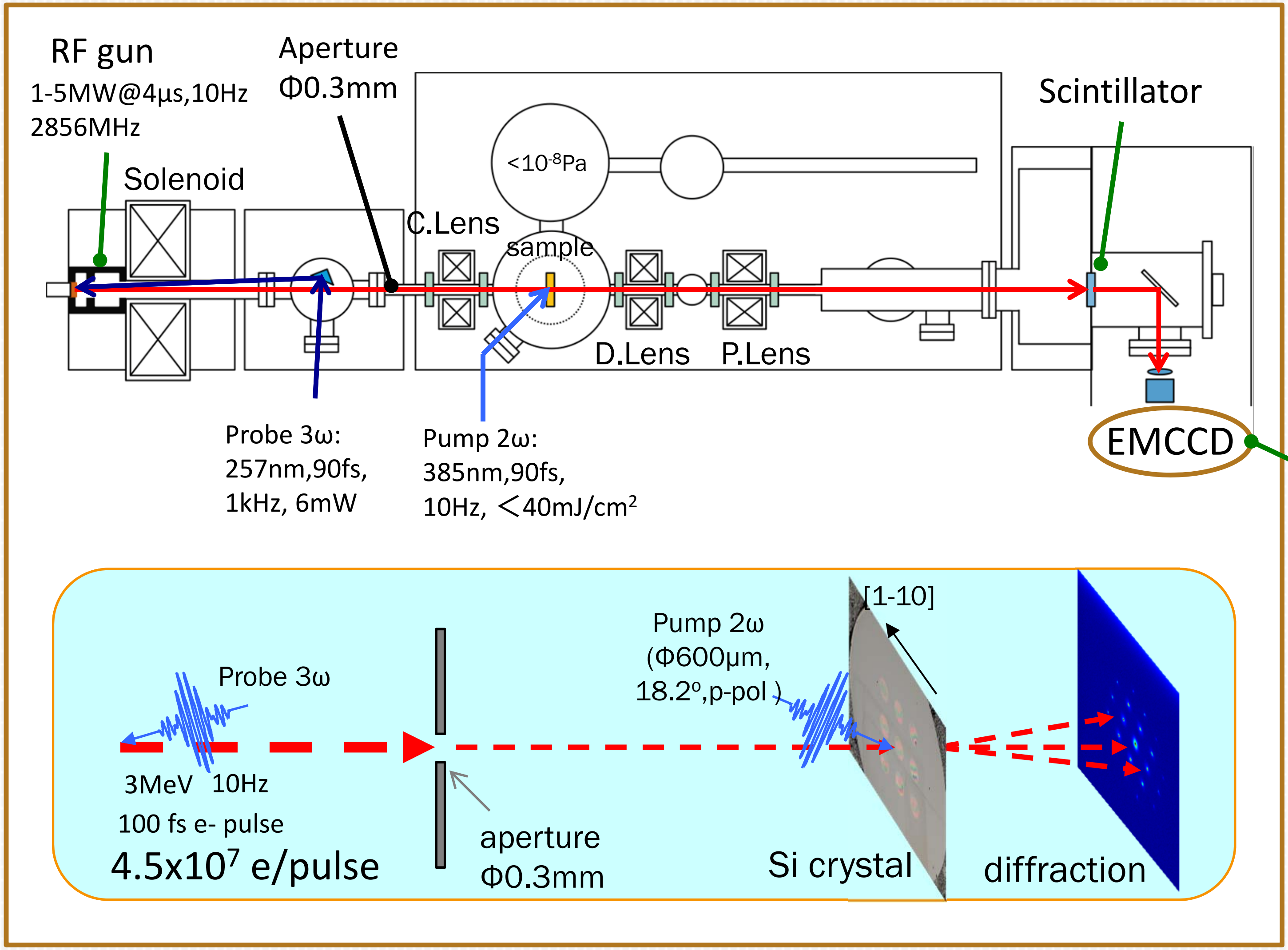


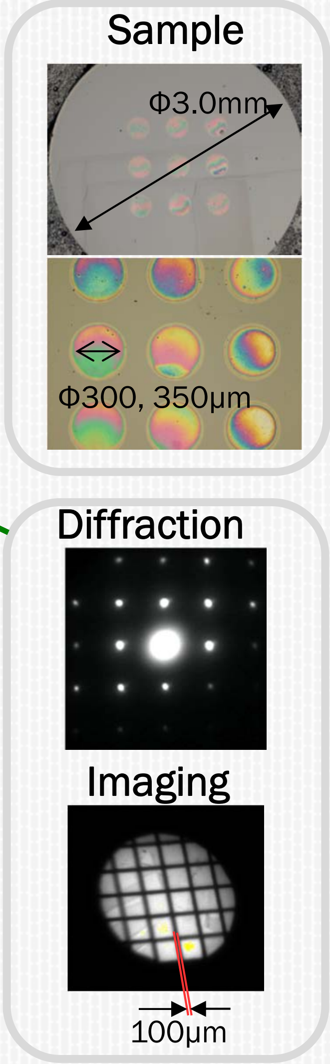




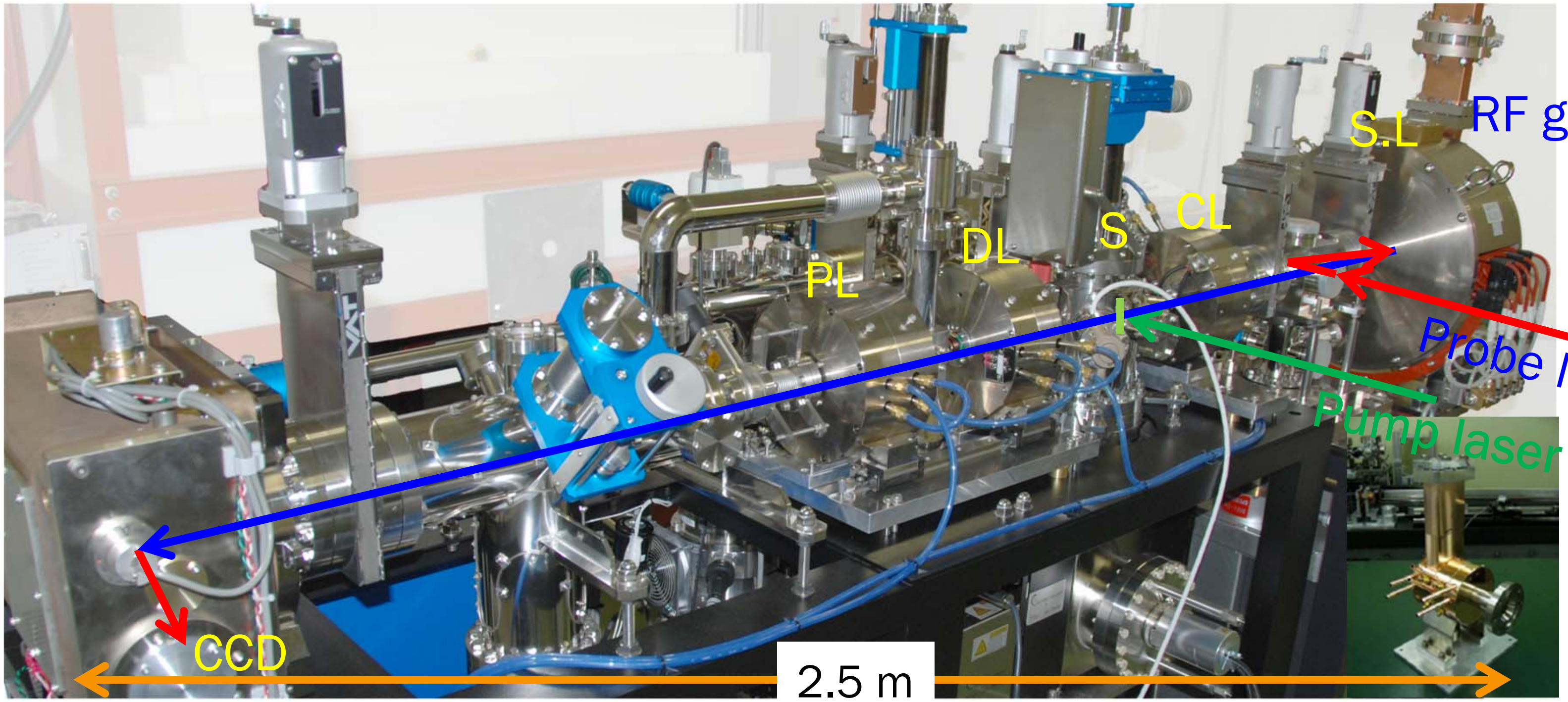
RF gun based MeV UED at Osaka Univ.

use of electron optical lenses as like in electron microscopy





Picture of fs MeV electron diffraction

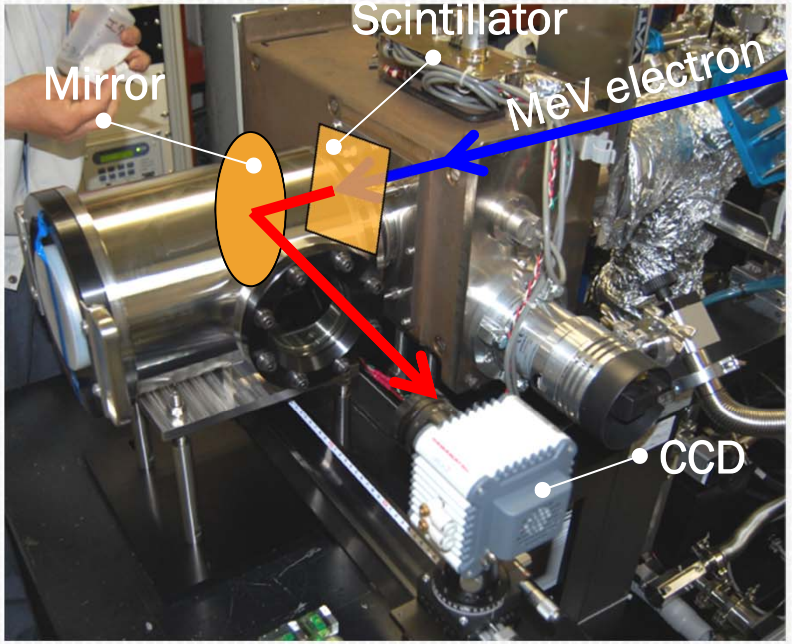


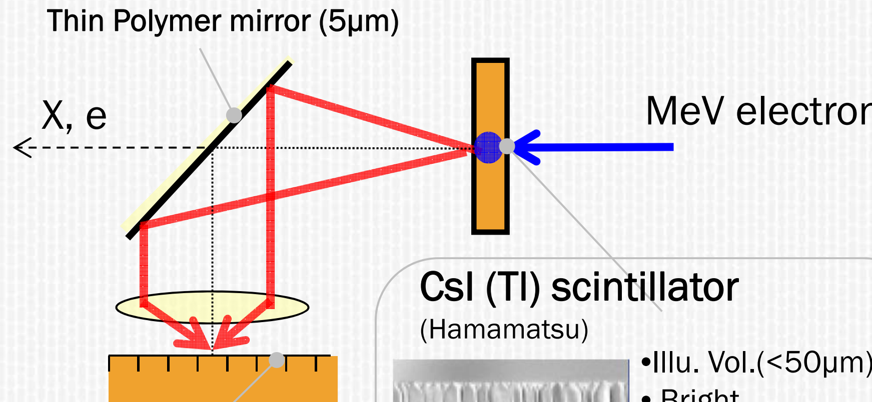
Difference with other UED facilities (i.e. UCLA, Tsinghua Univ., BNL, DESY):

use of electron optical lenses, therefore, compact.

Detection of MeV electron diffraction

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





CCD
6.45μm/pixel

CsI (Tl) scintillator
(Hamamatsu)

- Illu. Vol.(<50μm)
- Bright
- High resol. (Column Structure)
- Tough (for High E Xray)
- Large: 5x5cm

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

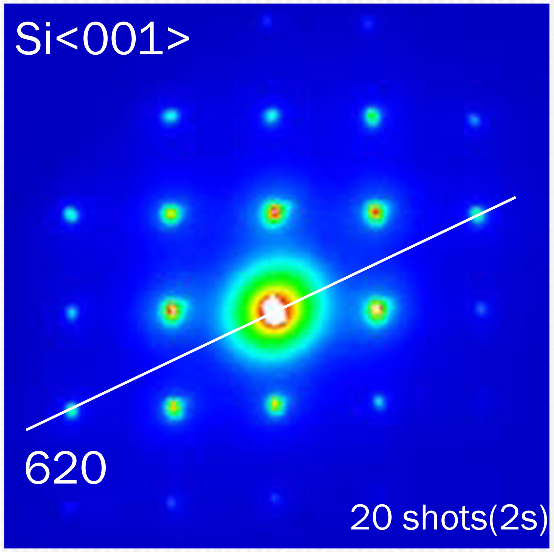
Solution

- 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

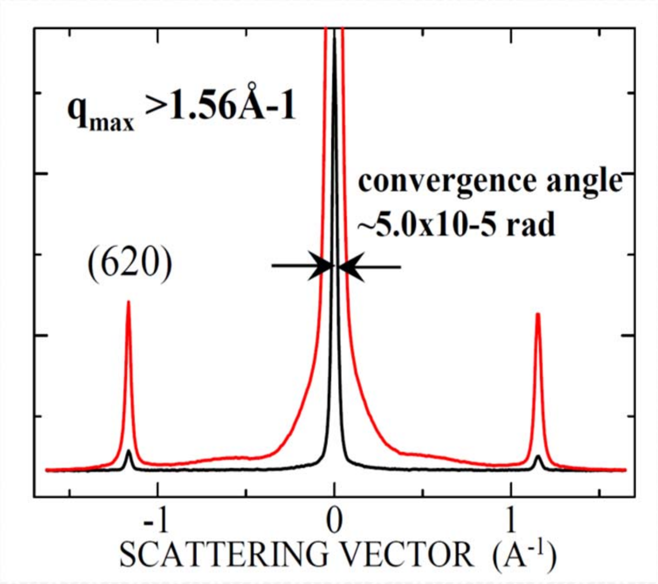
Quality of MeV electron diffraction

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

Sample: 180nm-thick single crystal Si



Intensity profile of 620 pattern



A high-quality MeV ED was observed!

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

- Bragg law

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

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

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

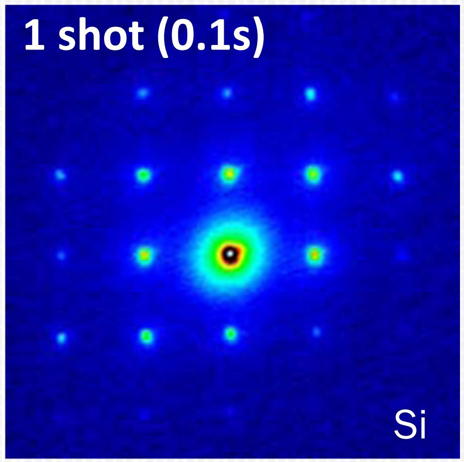
Power of the technique: static diffractions

- Single-shot measurement

Si
single crystal
Thickness: 180nm

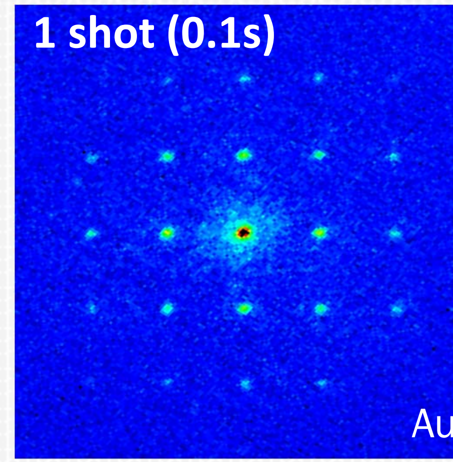
e- energy: 3MeV

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

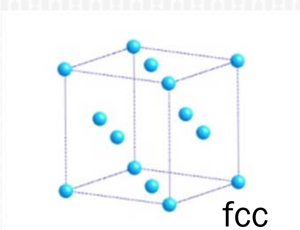


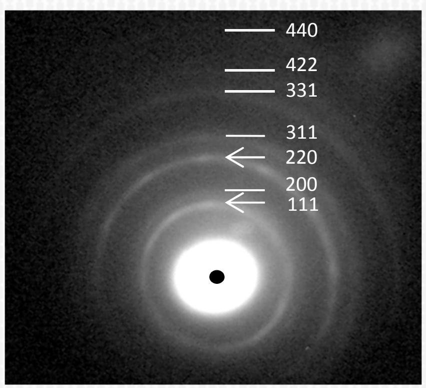
Au
single crystal
Thickness: 20nm

e- energy: 3MeV



- **Metal** (Al)
  - polycrystal (100nm)

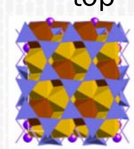


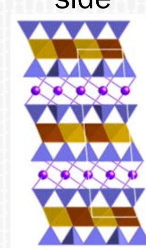


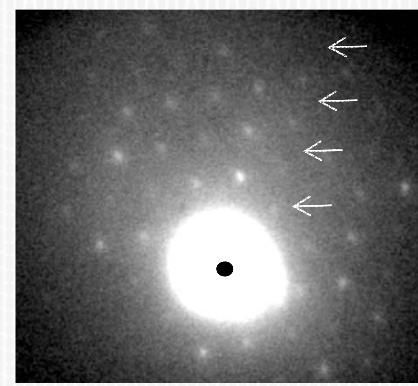
Large scattering vector
q_{max}

- **Insulator** (Mica)
  - Single crystal (~100s nm)

$K(Fe,Mg)_3(AlSi_3O_{10})(OH,F)_2$



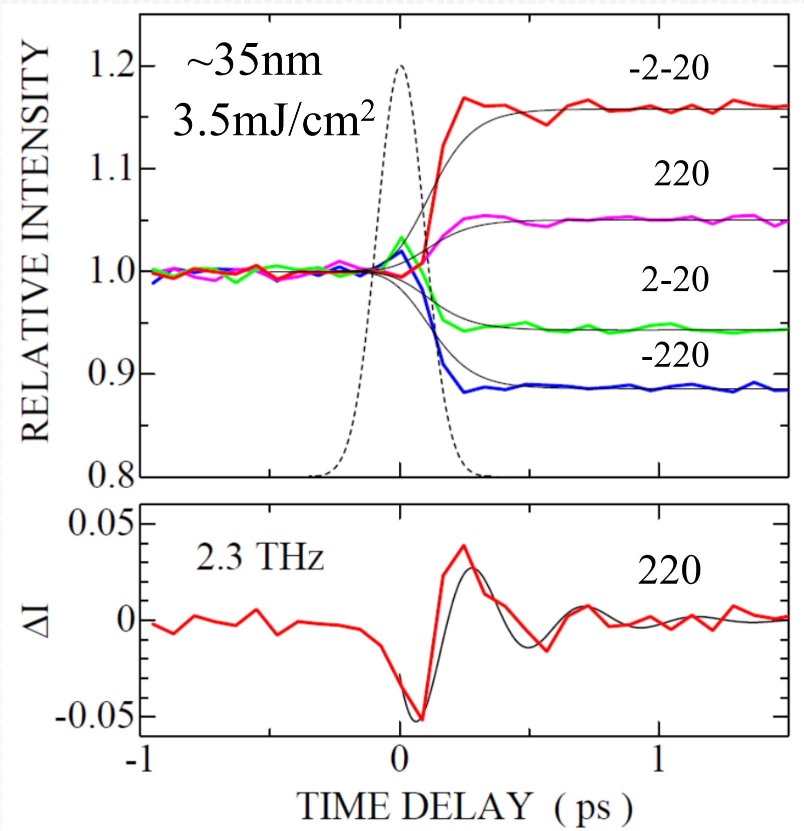




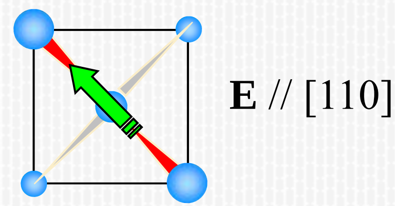
No charging effect
(Difficult at Low Voltage)

Time-resolved measurement #1

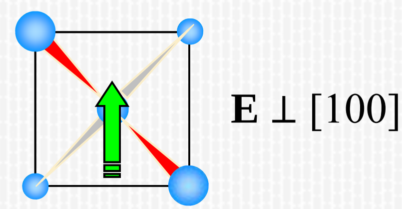
Dynamics of phase transition in single-crystal Si

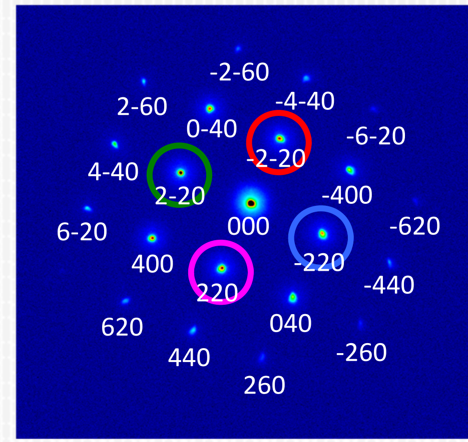


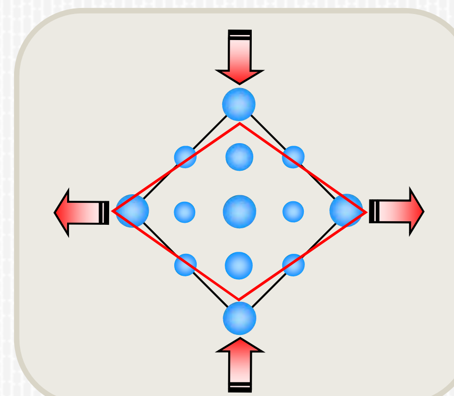
The intensity of diffraction pattern increases due to the lattice heating, if



The intensity of diffraction pattern decreases due to the lattice heating, if

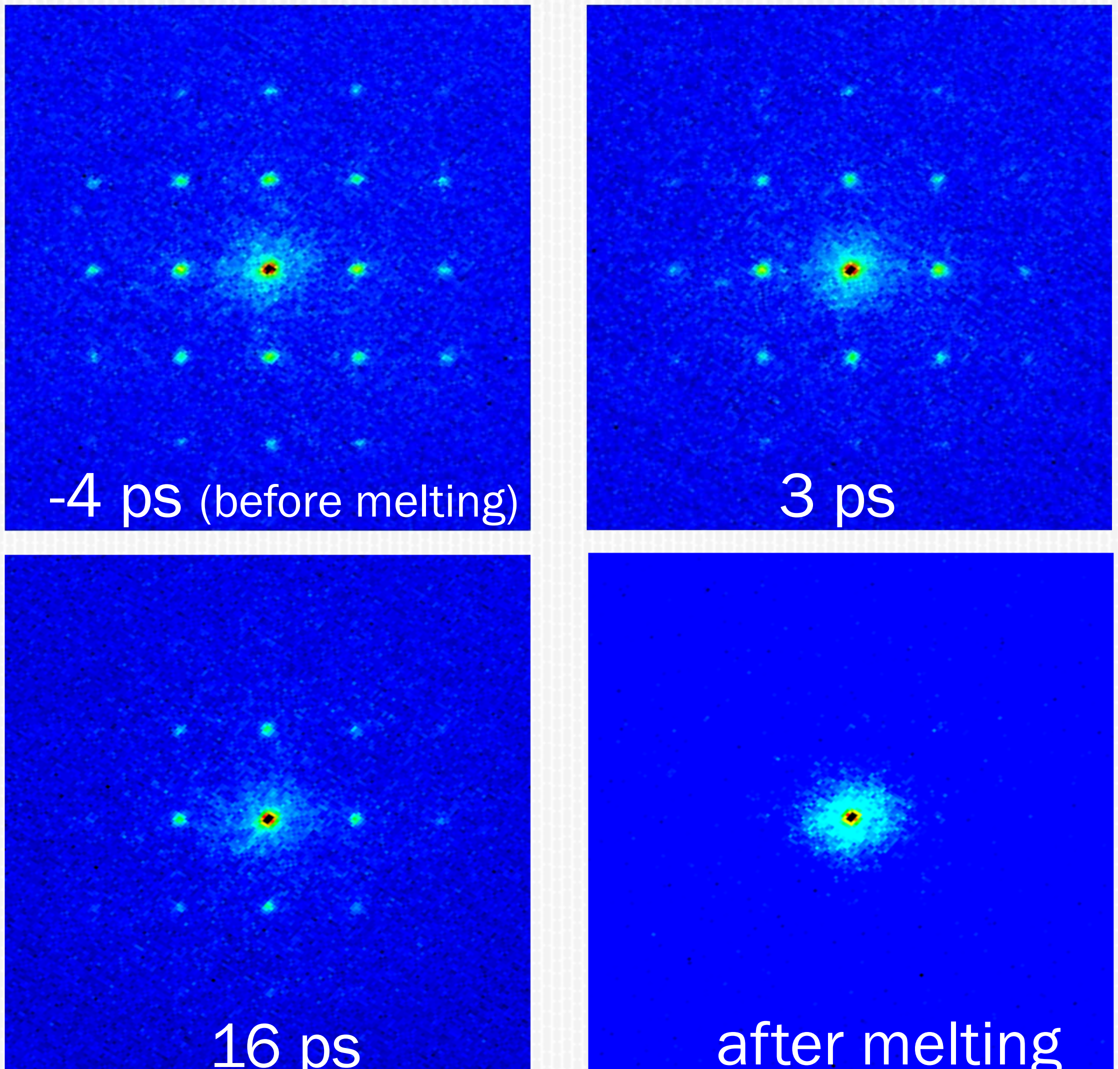






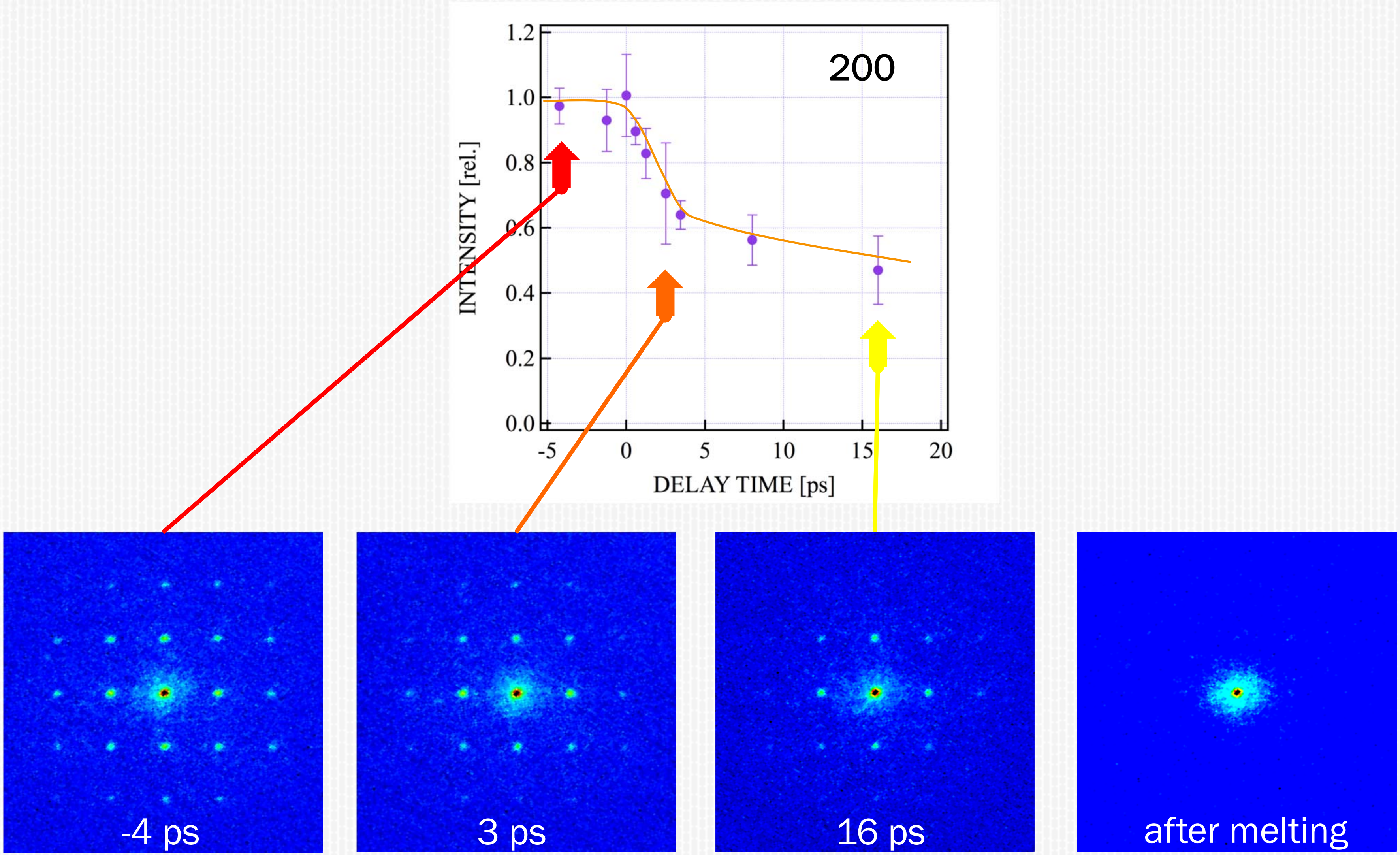
Time-resolved measurement #2

Laser heating and melting dynamics of single crystal Au



Time-resolved measurement #2

<u>Laser heating and melting dynamics of single crystal Au</u>



- The UED experiments indicate that the RF gun based MeV UED is powerful tool for the study of ultrafast dynamics with time resolution of 100 fs or less.

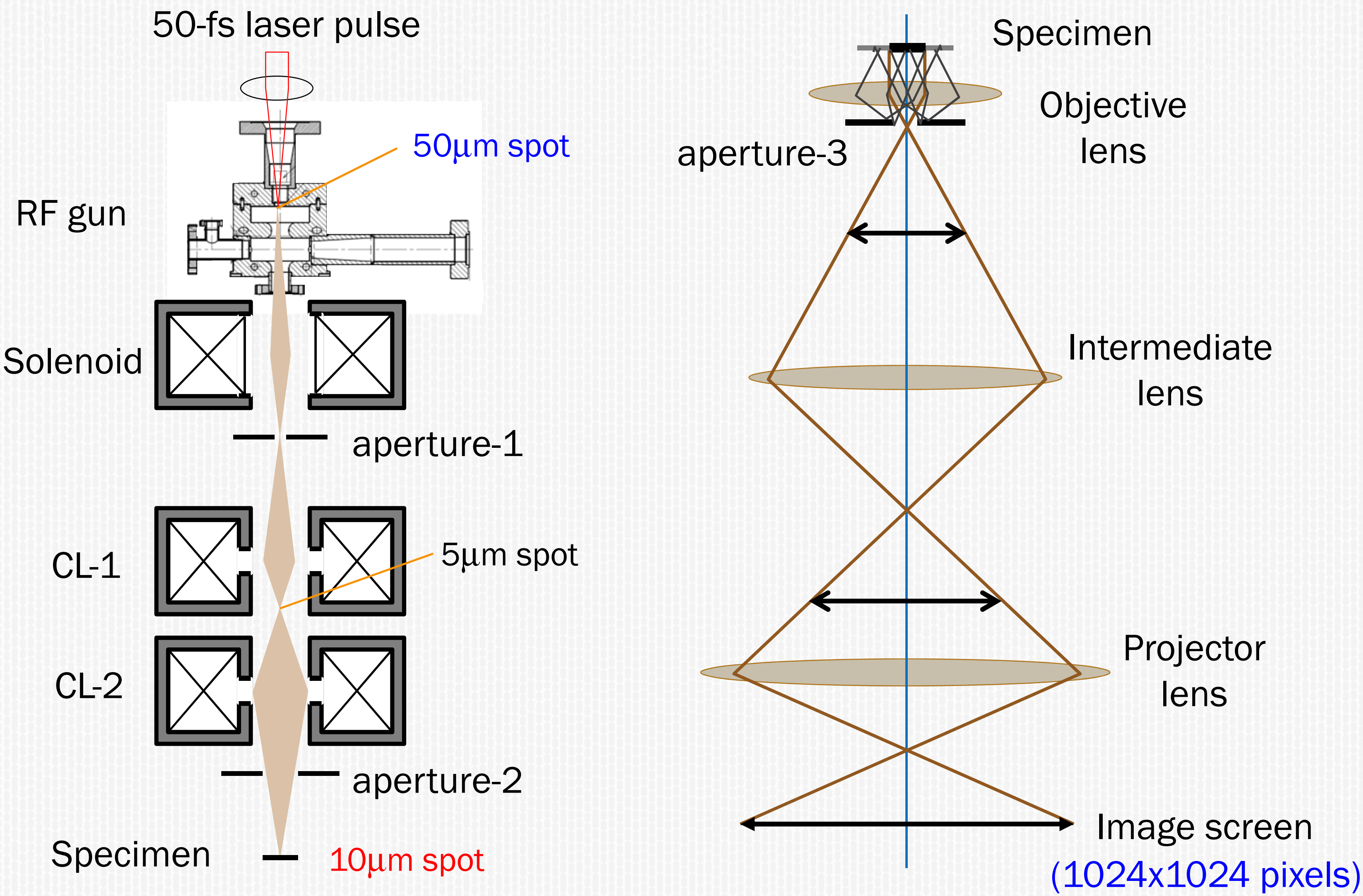
- However, there is no spatial resolution in UED

- To achieve both the time and spatial resolutions, i.e. fs-nm, a time-resolved electron microscopy is required.

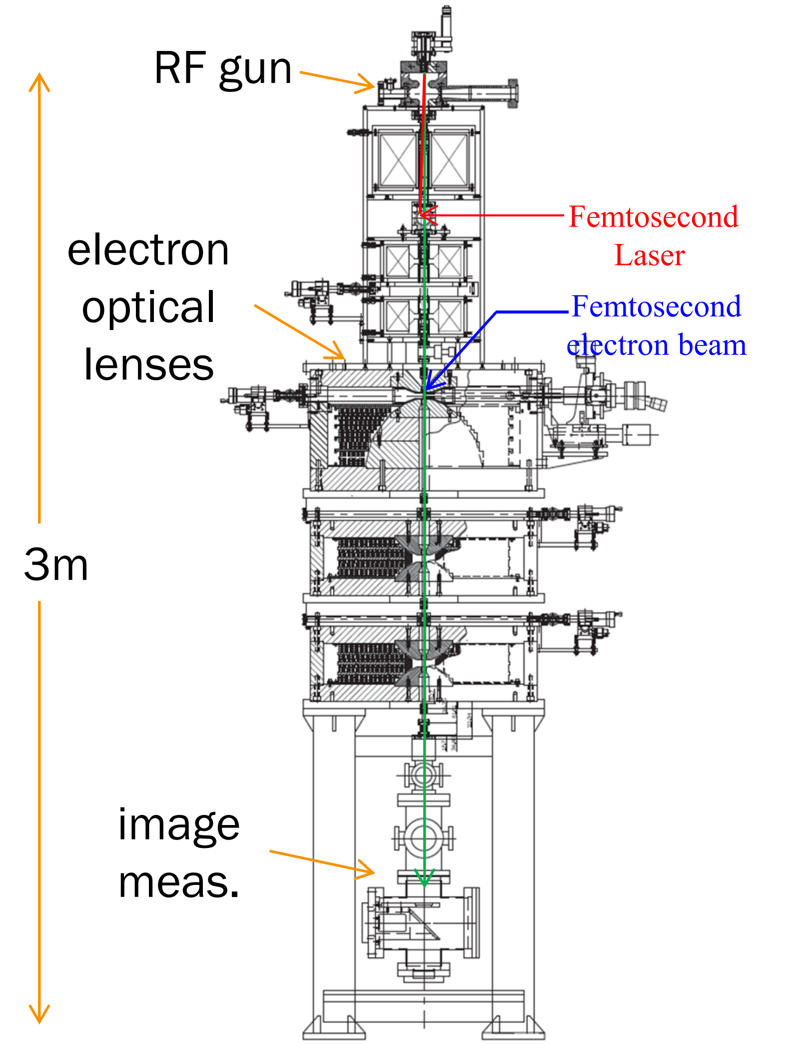
Femtosecond MeV electron microscopy using RF gun
(MeV UEM)

(under development at Osaka Univ.)

Concept of MeV UEM

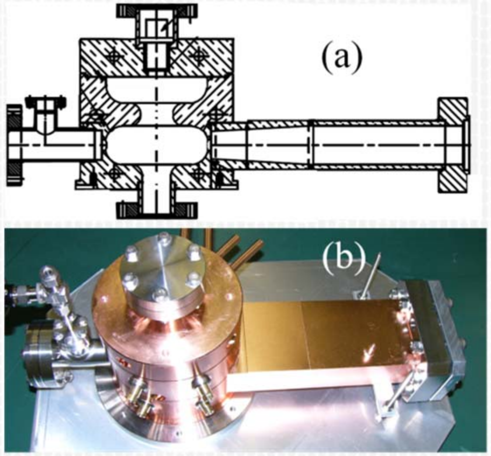


Prototype of femtosecond MeV transmission electron microscopy



Prototype of MeV UEM

Femtosecond photocathode electron gun



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

Time resolution: < 1 ps

Spatial resolution: ~10s nm

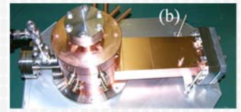
Challenge !

Next TEM

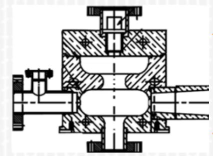

Standard 3-MeV TEM
at Osaka Univ.

10m

3m



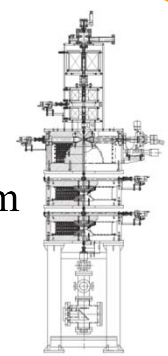
photocathode
RF gun



15cm

First compact
MeV Transmission Electron Microscopy

3m



Next TEM

With functions of

TEM (nm or sub-nm, MeV)

+

time resolved (femtosecond)

Dream electron microscopy!

Concluding remarks

✓The photocathode RF gun is a powerful source to generate directly a 100 fs electron beam with emittance of ~0.1 μm

✓The femtosecond RF gun is very useful for ultrafast MeV electron diffraction.

✓It is expected to be used in high-voltage time-resolved electron microscopy.

However, great efforts and many challenges are required:

- ➢reduce further the emittance (<0.1 μm) and energy spread (10^{-5} or less),
- ➢improve the stabilities on the charge and energy,
- ➢reduce the synchronized time jitter,
- ➢develop a detection of very electron with MeV energy region.

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