フェムト秒時間分解MeV電子顕微鏡の開発

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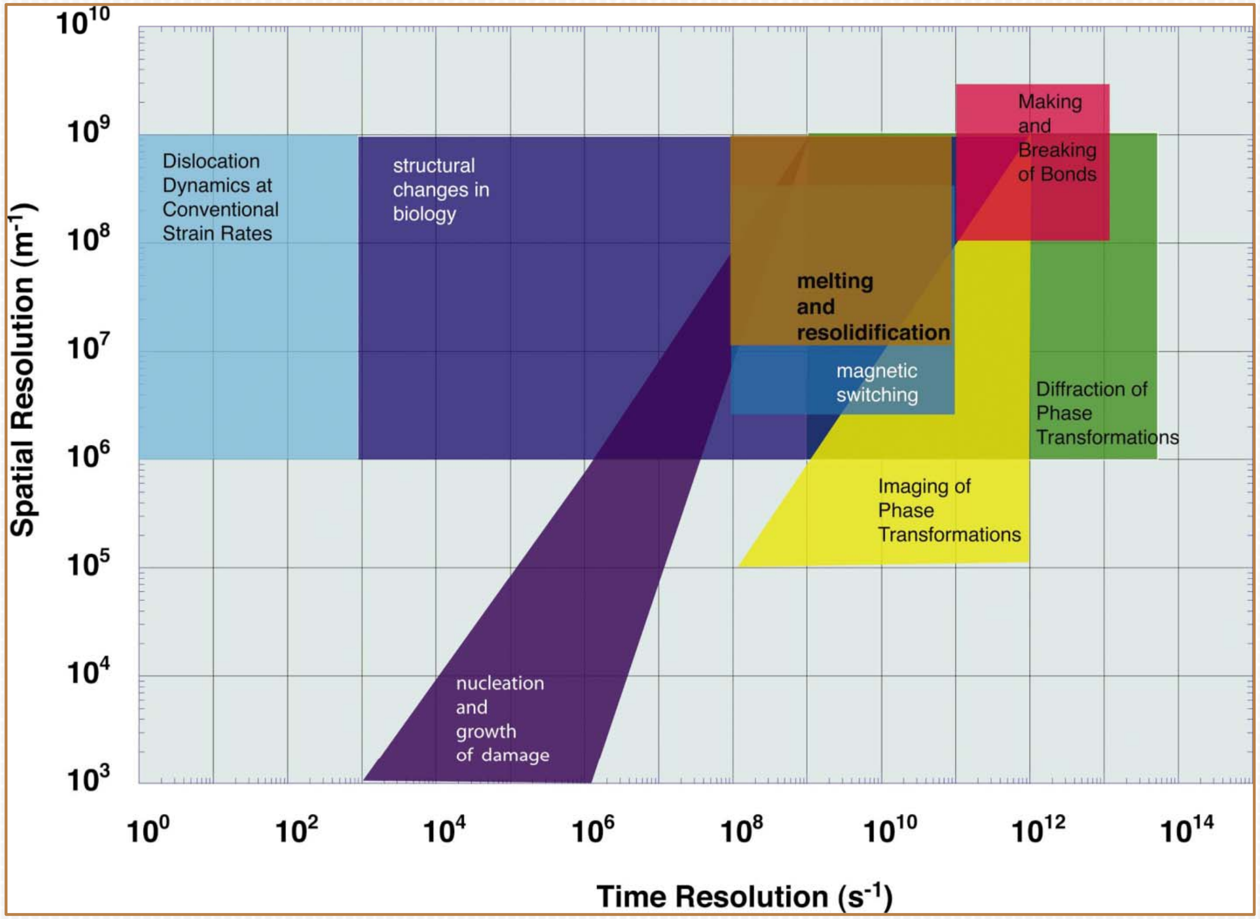
（産総研）黒田隆之助

構造変化による物質・化学現象



W. E. King, *et al., JAP 97, 111101 (2005)*

構造変化による物質の高速物理・化学現象はフェムト秒時間、ナノメートル空間のスケールで進行している。

これらの現象を解明するために、

~ 100 fs and
sub-Angstrom

の原子レベルの時空間分解能を有する測定技術が不可欠である。

原子・分子レベルの超高速測定技術

1） 高速X線回折／顕微鏡

Picosecond X-ray pulses from SR are used to observe the structure transformation.

Femtosecond X-ray pulses from X-ray FEL or laser plasmas acceleration will be used.
→ low scattering cross section, large energy deposited, ...
big experiment/measurement

2） 高速電子線回折 (UED)

time-resolved electron diffraction measurement using femtosecond electron beam and femtosecond laser. The time resolution has been achieved to 400~600fs using DC gun, and to 100 fs using photocathode RF gun.

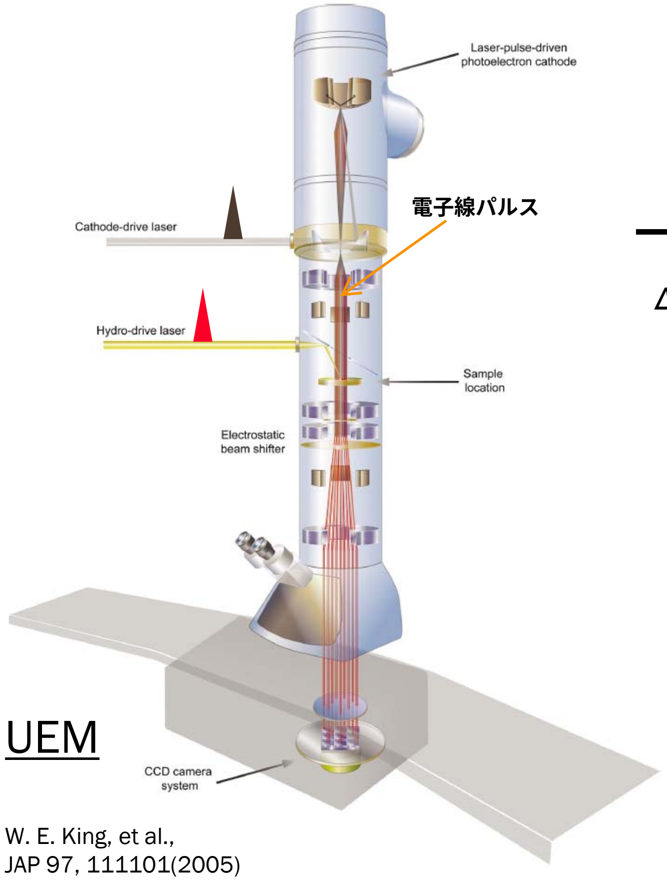
but no spatial resolution!

3） 時間分解電子顕微鏡 (UEM)

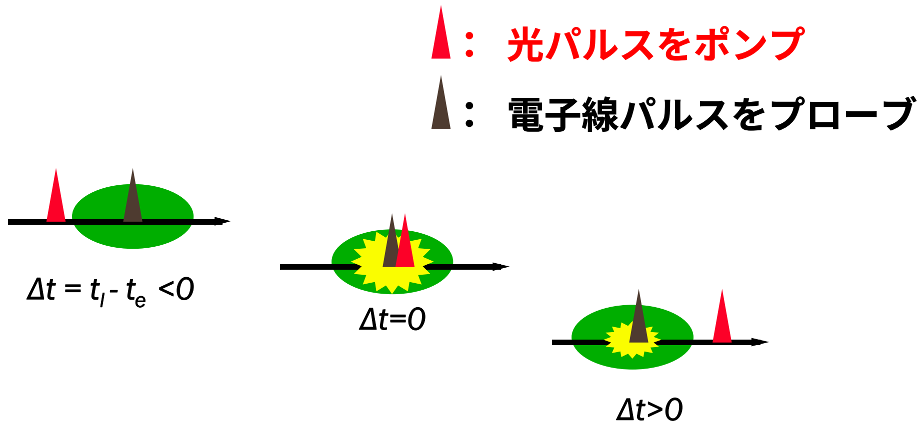
It can be observed the dynamics of structure transformation in real space.

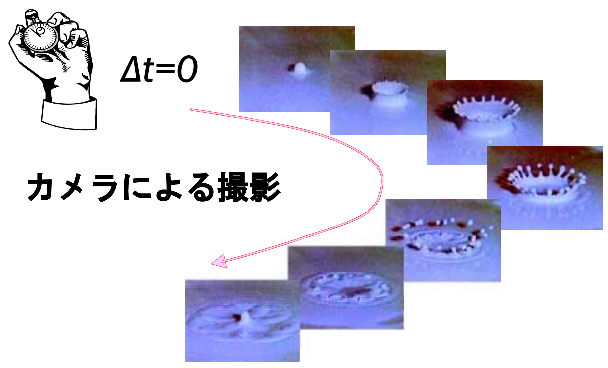
However, the resolution of UEM is stopped at ns-nm or ps-μm due to the electron bunch length, ...

時間分解電子顕微鏡の測定原理



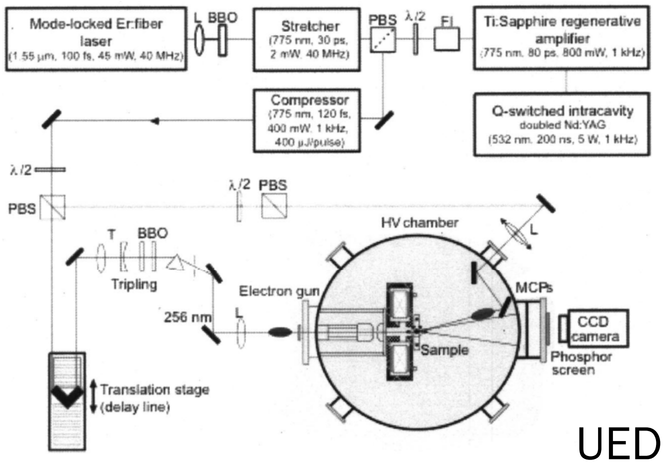
W. E. King, et al.,
JAP 97, 111101(2005)



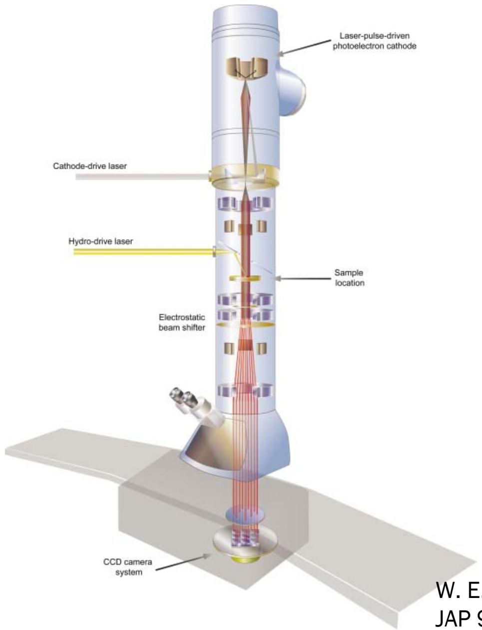


ポンププローブ技術と電子顕微鏡技術の融合

Why RF gun in UED & UEM?



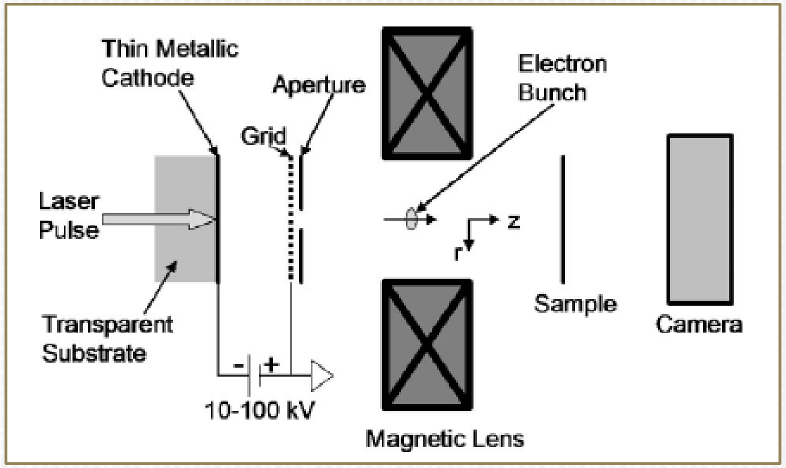
UED



UEM

W. E. King, et al.,
JAP 97, 111101(2005)

<u>Photocathode DC gun in UED & UEM</u>



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

<u>高速電子線回折</u>:

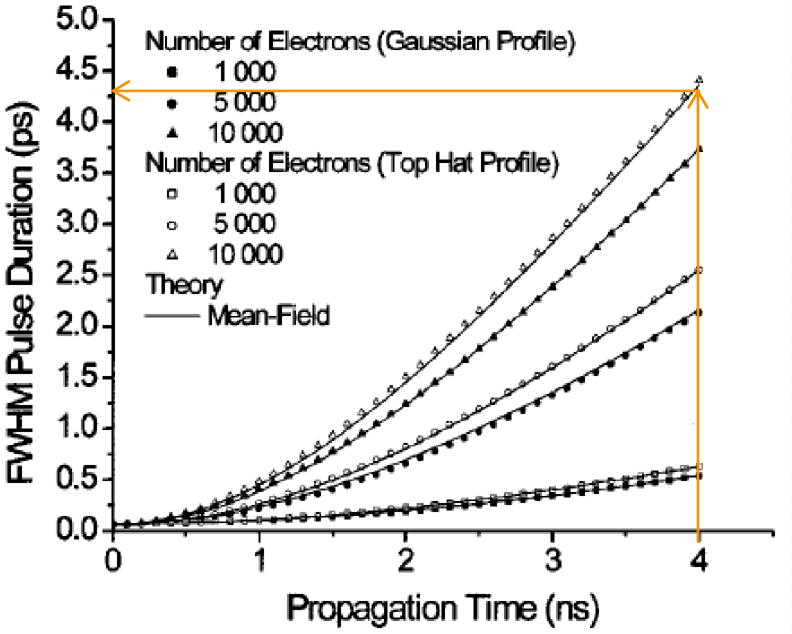
Bunch length: 400~600 fs
Electron charge: 10^3~10^4 e^-/pulse
Distance between sample and cathode: <5 cm

<u>時間分解電子顕微鏡の現状</u>：

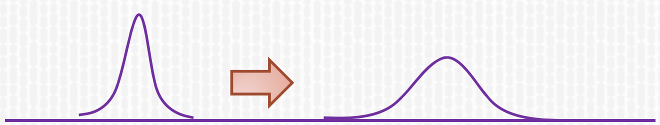
Bunch length: 10 ns
Electron charge: 10^8 e^-/pulse

Why RF gun in UED & UEM?

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



t=4ns → 40cm for 30KeV



空間電荷によるパルス幅の増大

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

Problems using DC electron gun:

加速電場(~10MV/m)が低いため、

空間電荷効果

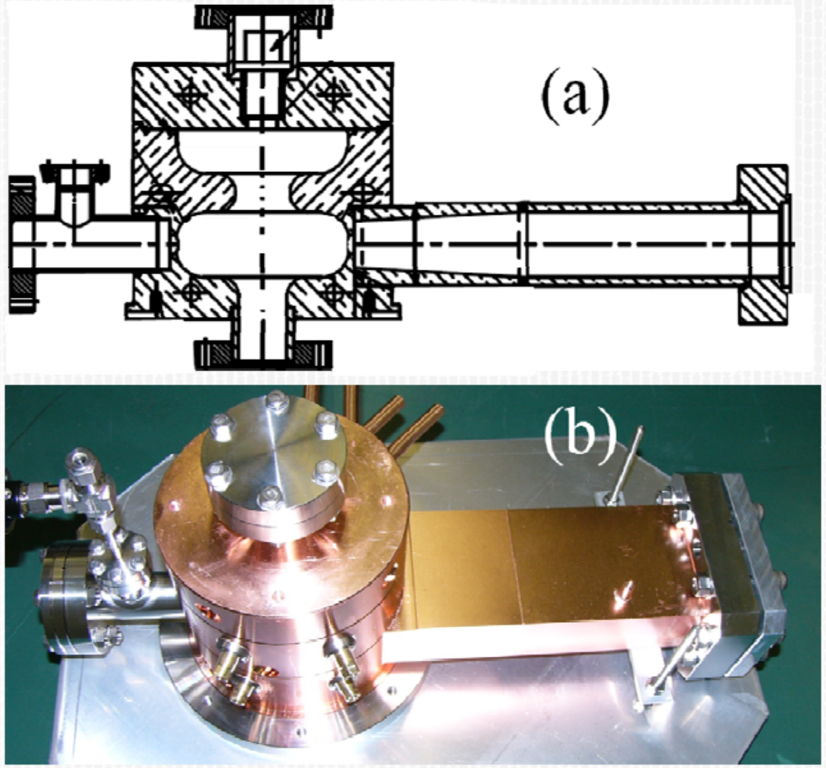
電子線パルス幅、エネルギー幅を増大

フェムト秒の時間分解能、ナノメートルの空間分解能を同時に実現することが非常に難しい。

⇒ 解決策として、
加速電圧を上げるしかない。

例えば、RF電子銃の使用

New femtosecond RF gun for UEM



developed in 2011
under the collaboration with KEK

Improvements for high quality:

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

Challenge !

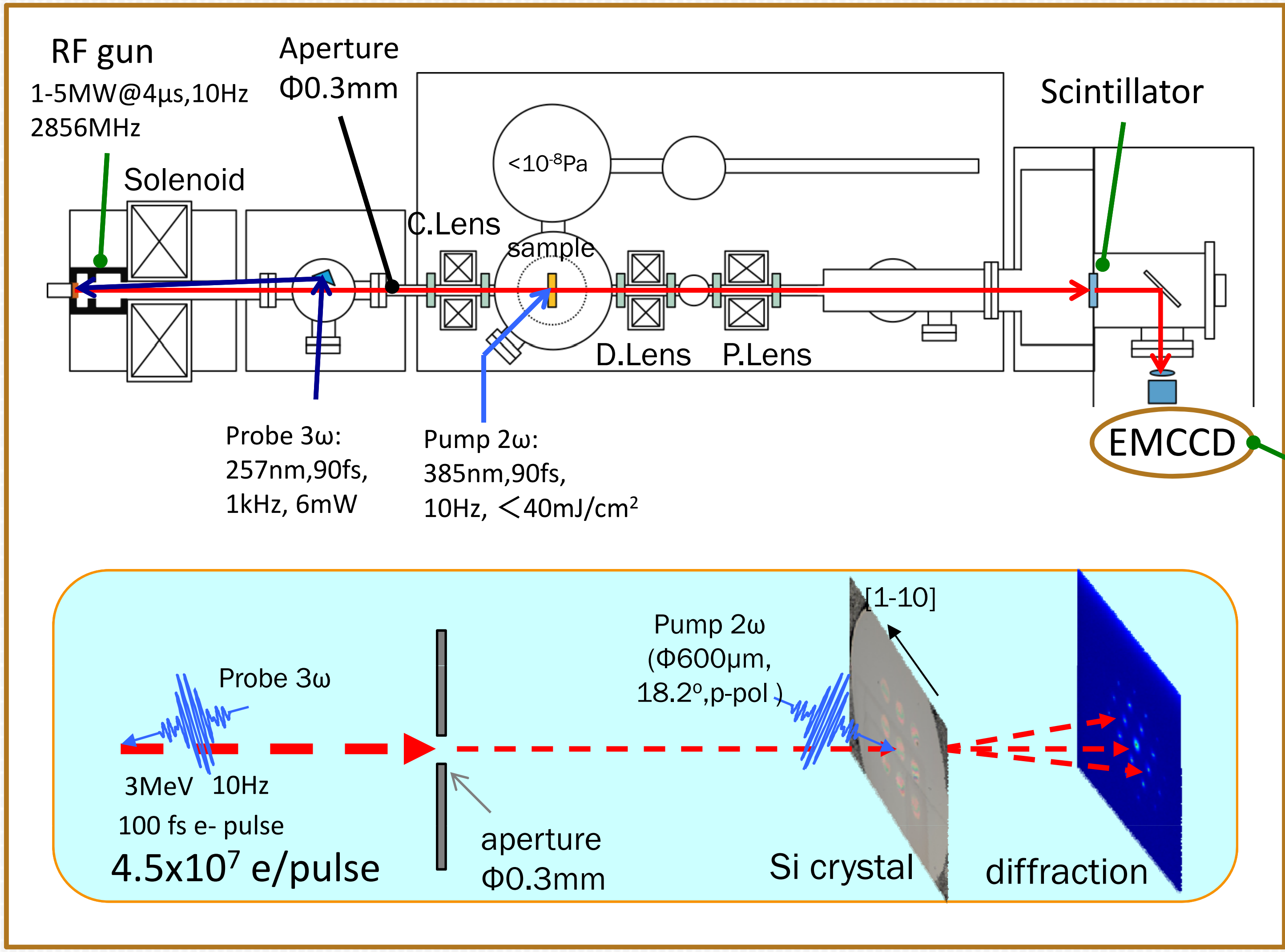
Electron energy：	1~3 MeV
Bunch length ：	≦100 fs
Emittance：	< 0.1mm-mrad
Energy spread：	10^{-4} (10^{-5} for challenge)
Charge：	10^{7}~10^{8} e^{-}'s/pulse

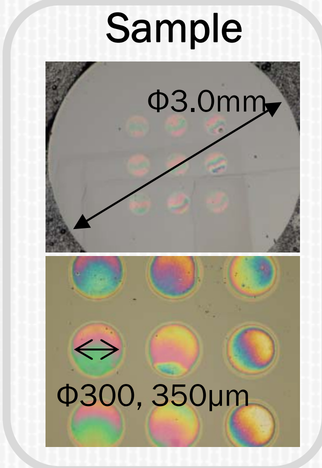
RF gun based
MeV electron diffraction at Osaka Univ.

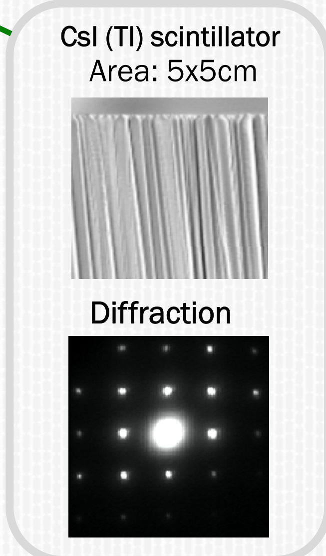
Electron energy: 1～3 MeV
Time resolution: 100 fs

RF gun based MeV UED at Osaka Univ.

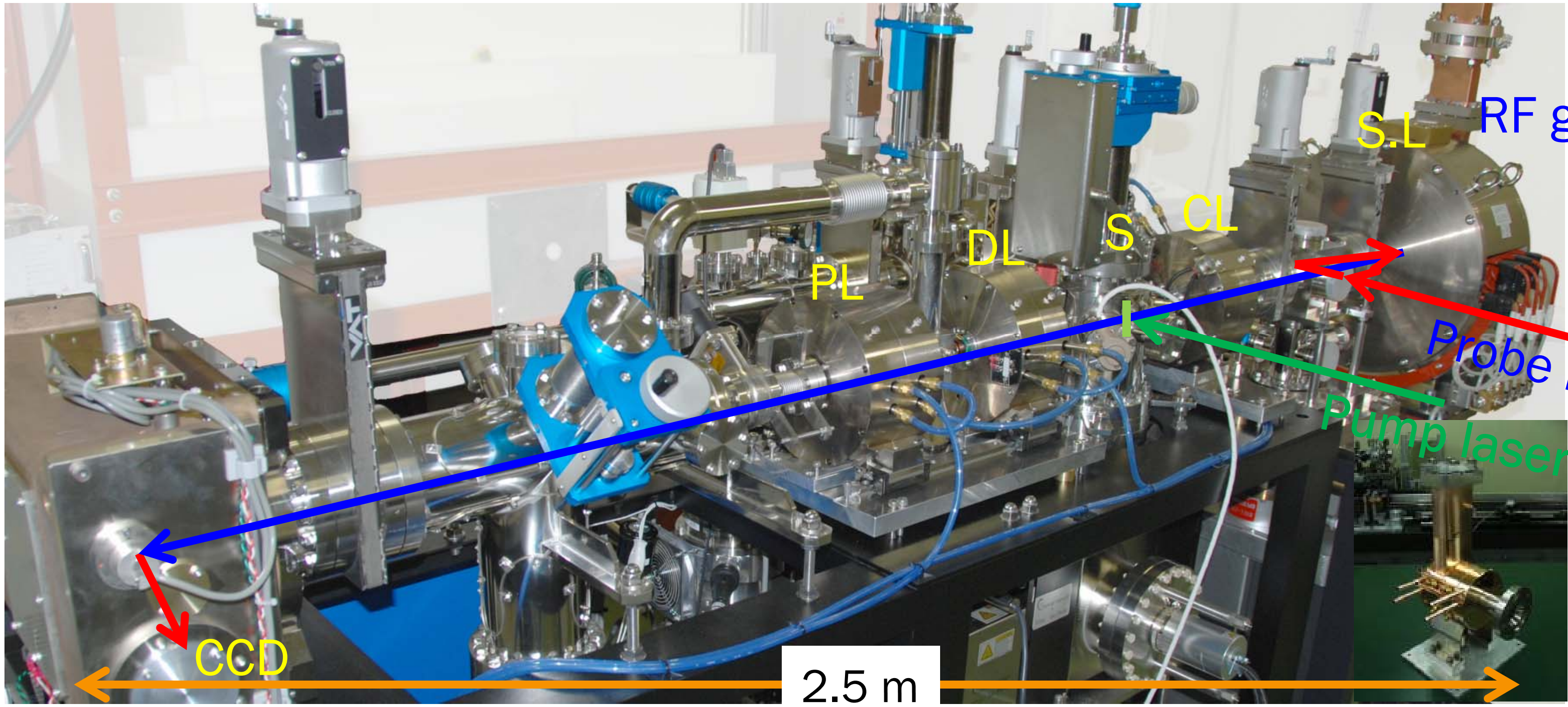
use of electron optical lenses as like in electron microscopy







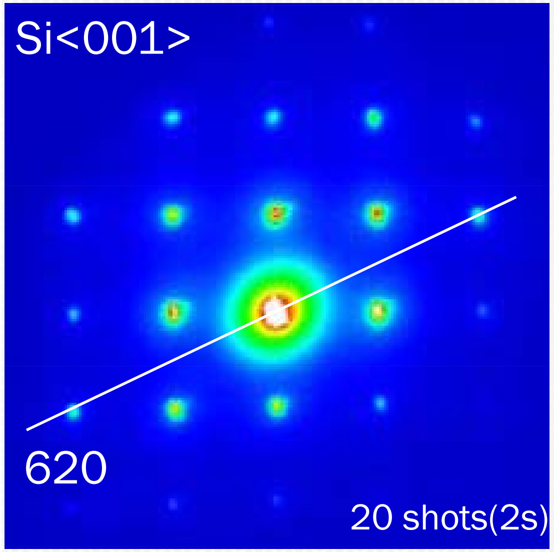
Picture of UED system at Osaka Univ.



use of electron optical lenses, therefore, compact.

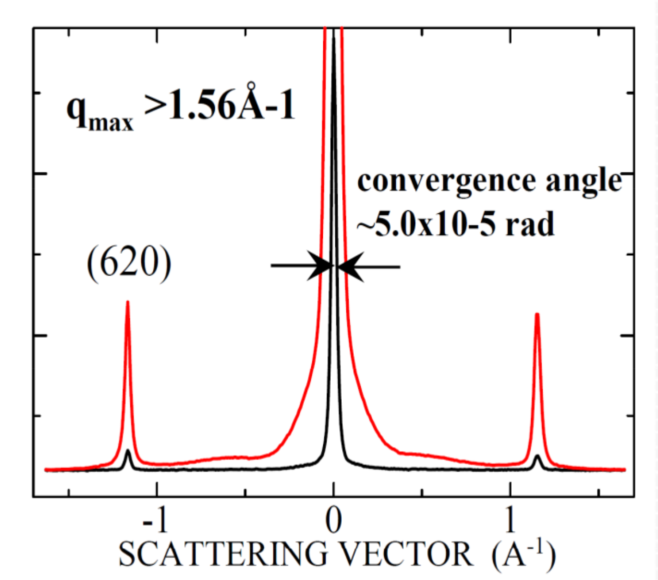
Quality of MeV electron diffraction

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} >1.56$Å^{-1}$
- Requirement of the e^- number: 10^{5-6}

- Bragg law

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

$$\tan 2\alpha = \frac{D}{L}$$

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

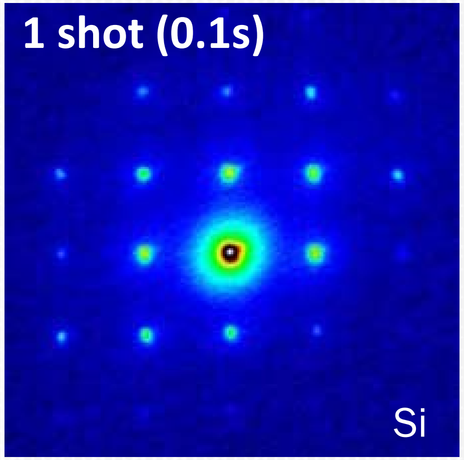
Power of the technique: static diffractions

- Single-shot measurement

Si
single crystal
Thickness: 180nm

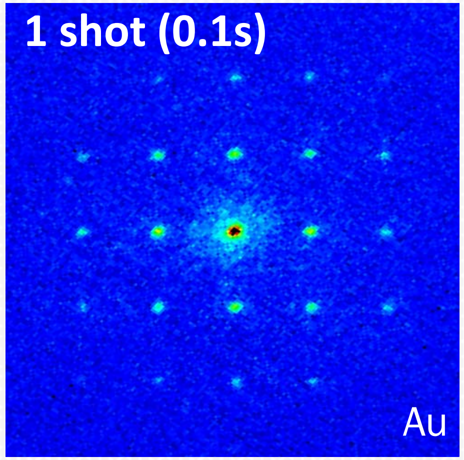
e- energy: 3MeV

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

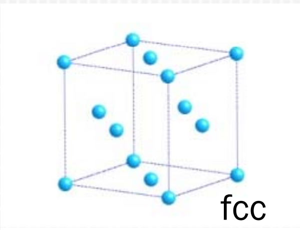


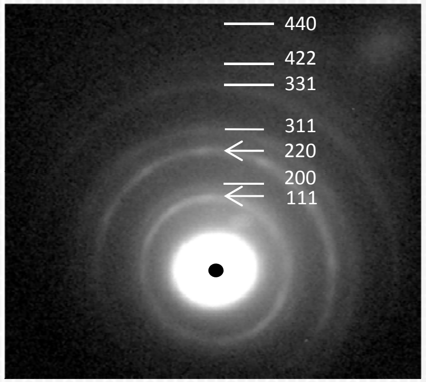
Au
single crystal
Thickness: 20nm

e- energy: 3MeV



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

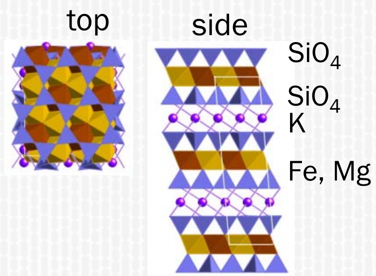


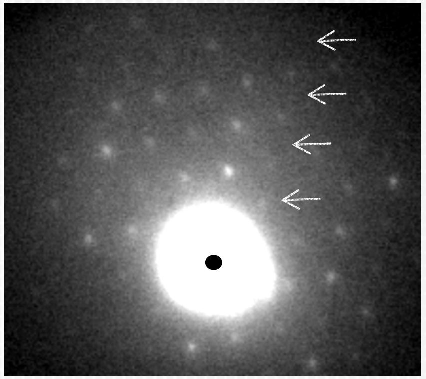


Large scattering vector
q_{max}

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

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

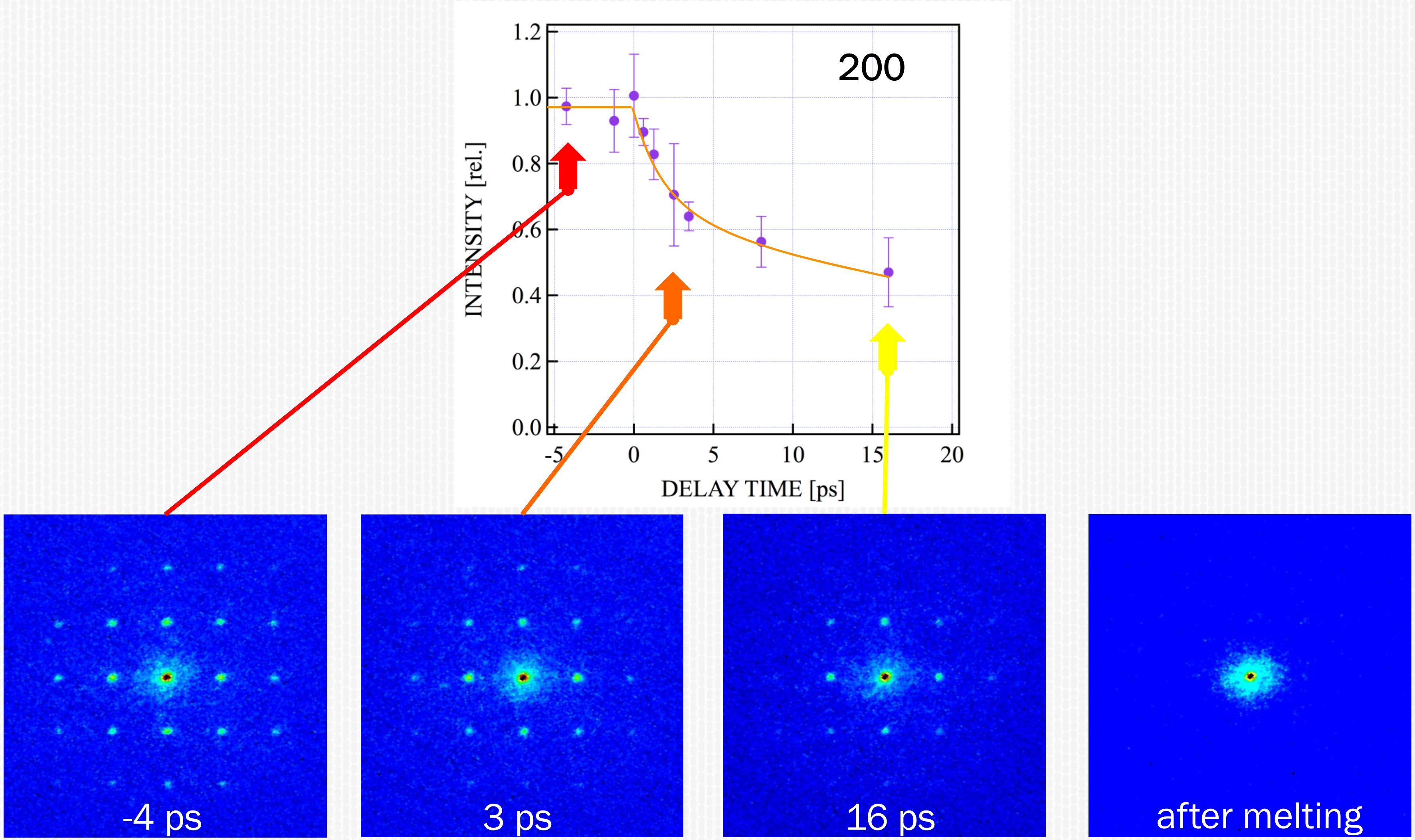




No charging effect
(Difficult at Low Voltage)

Time-resolved measurement #2

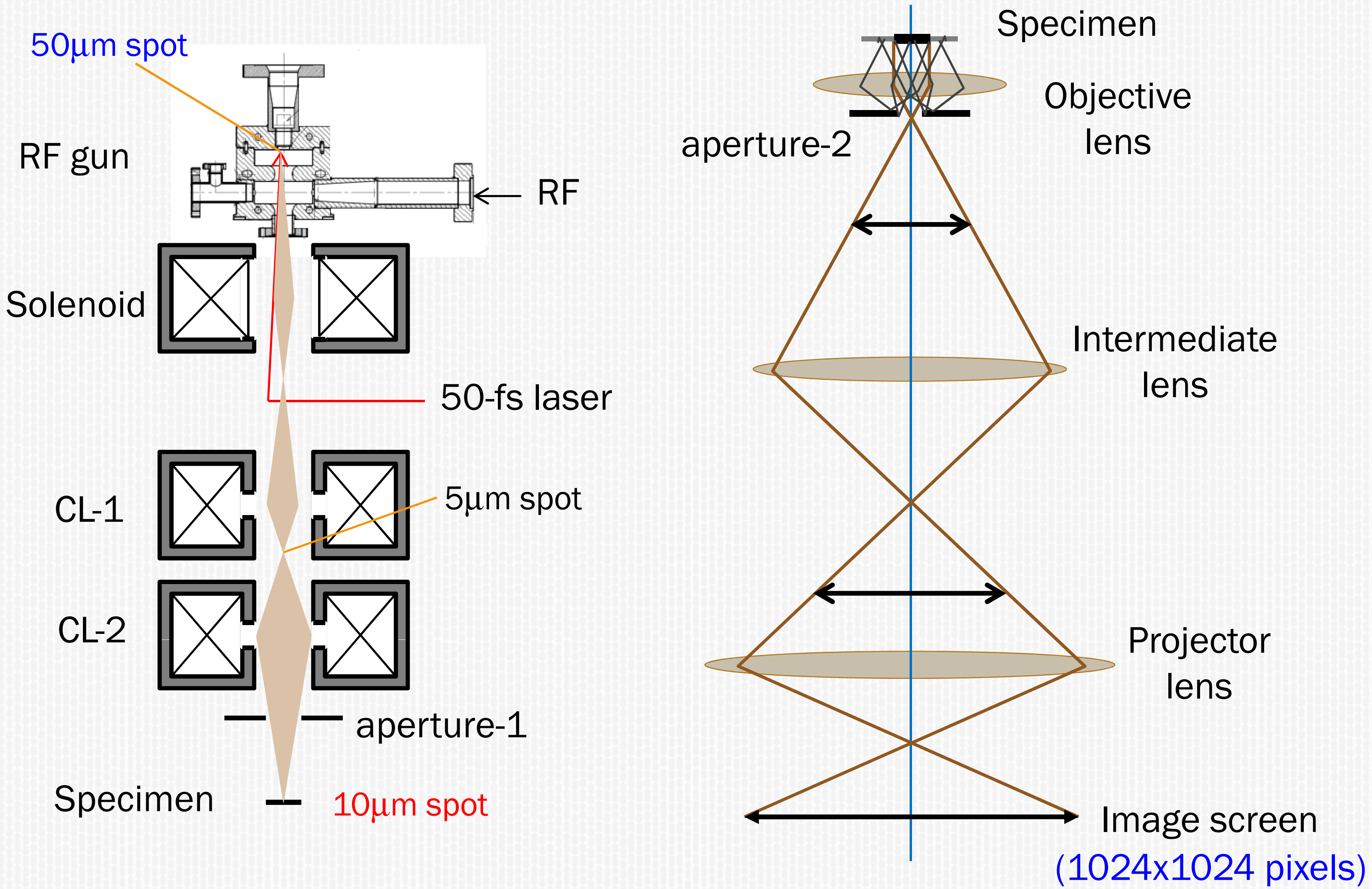
Laser heating and melting dynamics of single crystal Au



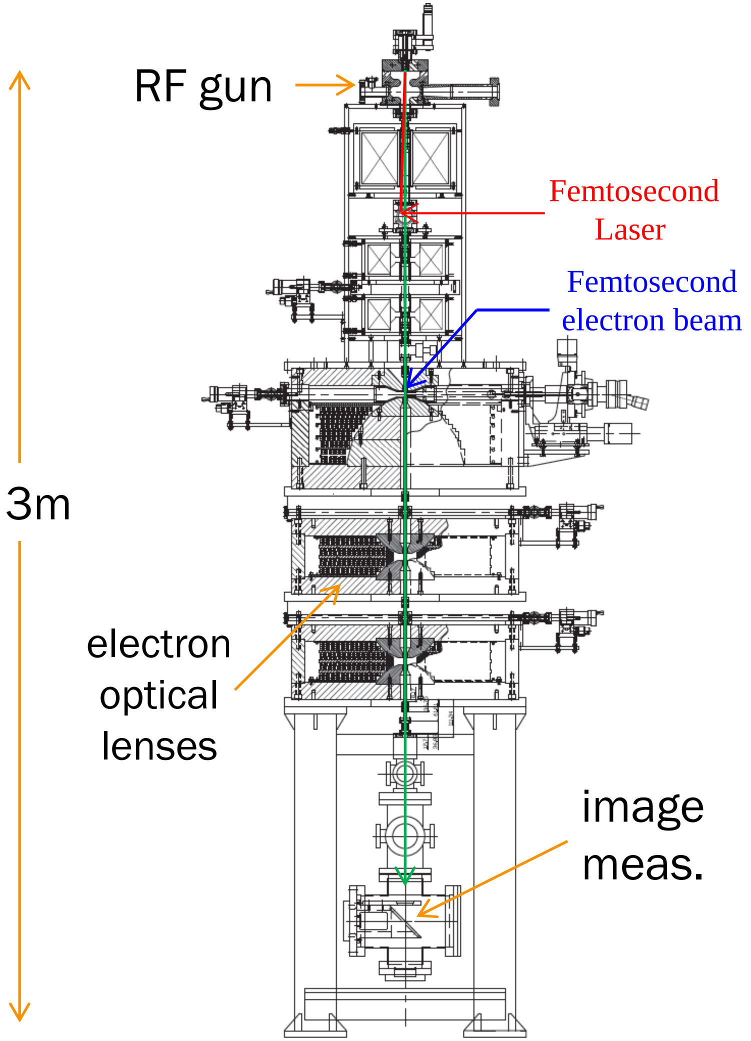
RF gun based transmission electron microscopy

Electron beam energy:	1～3 MeV
Temporal resolution:	100 fs
Spatial resolution:	10 nm

Concept of MeV UEM

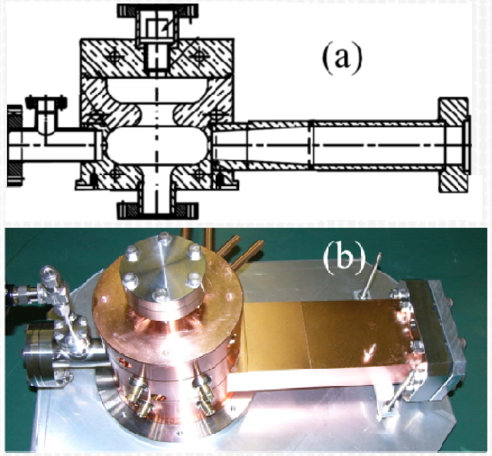


Prototype of
RF gun based MeV electron microscopy



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^{8}e's/pulse

Time resolution: 100 fs

Spatial resolution: 10 nm

Challenge !

Construction Processes of MeV UEM

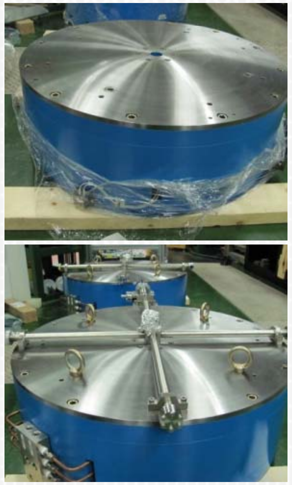
RF gun based injector

2010

2T objective lens

2011

Interm. & proj. lenses



2012

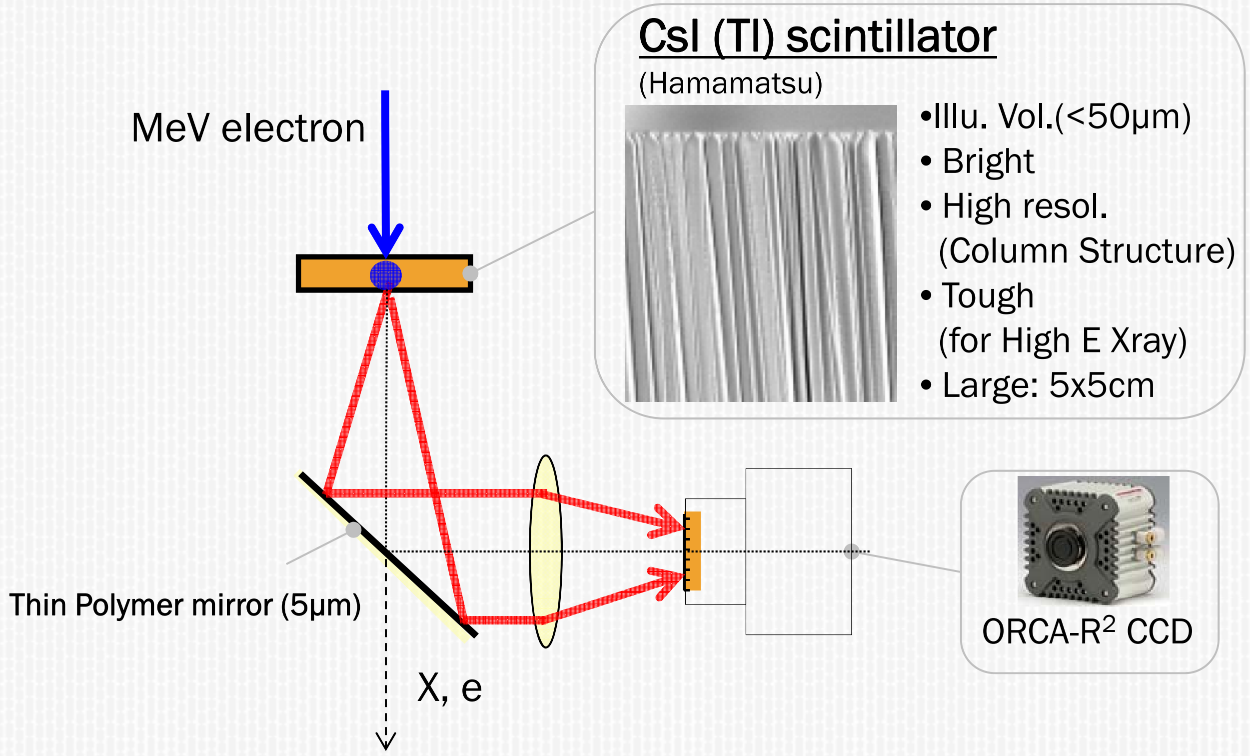
Image recording system

Prototype of
RF gun based MeV electron microscopy

Prototype of MeV UEM
(height: 3m, diameter: 0.7m)

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

Detection of MeV electron images

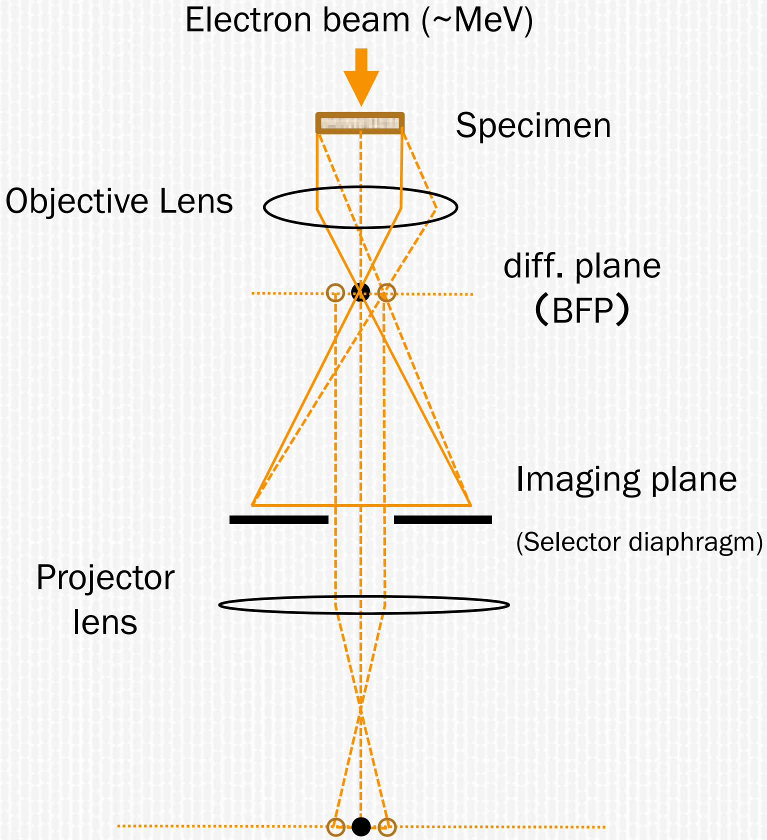


The detection system was successfully used in UED measurement.
(single-shot measurement with 10^5 e^-s/pulse)

MeV電子線回折の実験

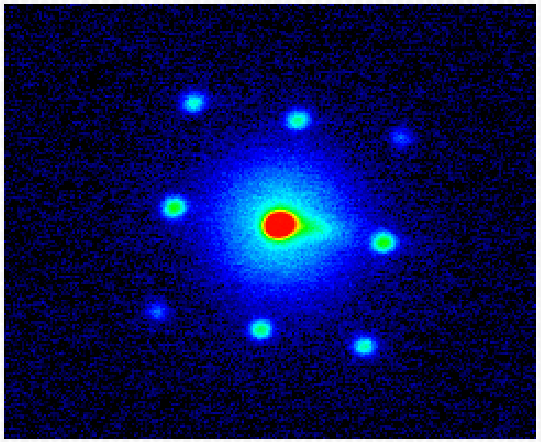
MeV electron diffractions observed in prototype of RF gun based UEM

First exp. at Nov. 9, 2012

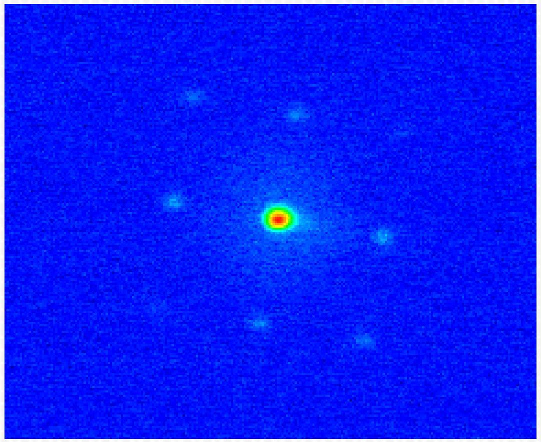


Sample： Au single crystal, ~15 nm
Electron charge： ~100 fC/pulse

10 pulses

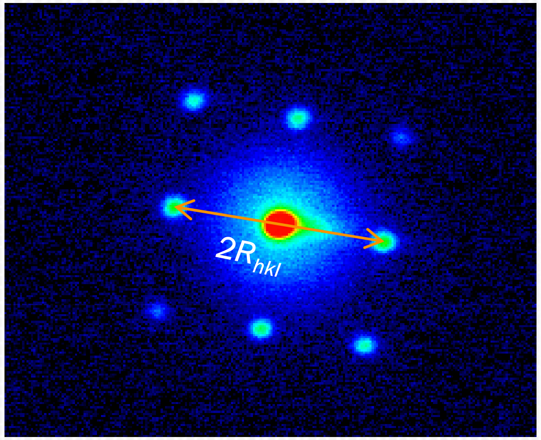


Single-shot



電子線回折によるビーム診断

回折パターンから ビームエネルギーの評価



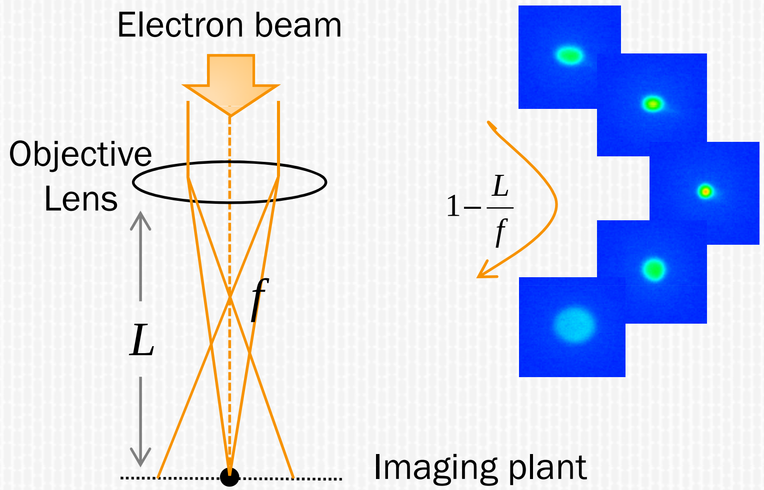
$$R_{hkl} d_{hkl} = \lambda L$$

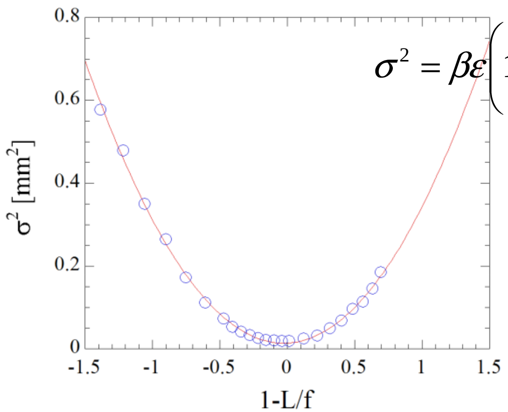
$$d_{hkl} = \frac{a}{\sqrt{h^2 + k^2 + l^2}}$$

L = 1 m: **カメラ長**
a=4.0788 A: Au**の格子定数**

$\lambda \rightarrow$ **ビームエネルギー**(E)

磁気レンズによる ビームエミッタンスの測定





$$\sigma^2 = \beta\varepsilon\left(1-\frac{L}{f}\right)^2 - 2L\alpha\varepsilon\left(1-\frac{L}{f}\right) + L^2\gamma\varepsilon$$

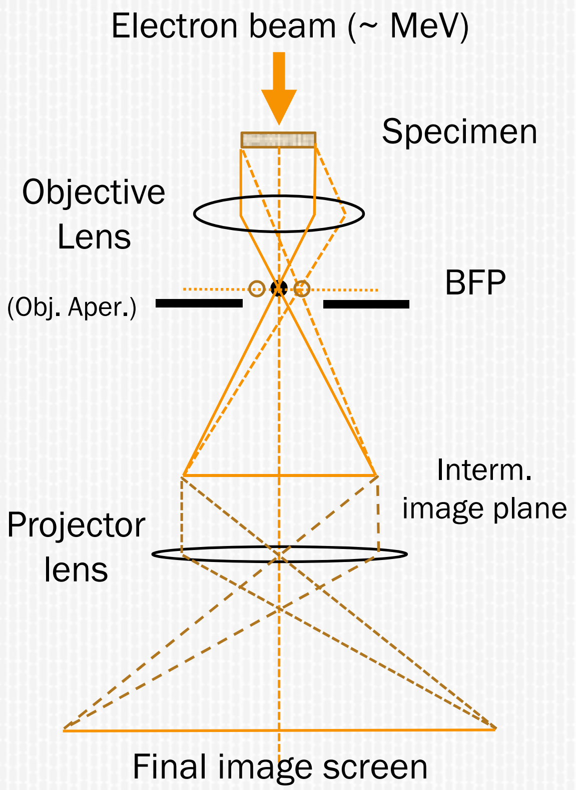
ε = 0.2 mm-mrad

Laser spot radius on cathode:
0.2 mm

MeV電子線イメージングの実験

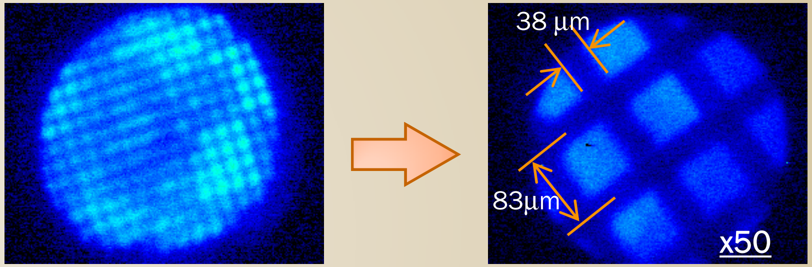
MeV electron imaging
in prototype of UEM

First exp. at Nov. 10, 2012



Sample： Au single crystal, ~15 nm
Electron charge： ~10 fC/pulse

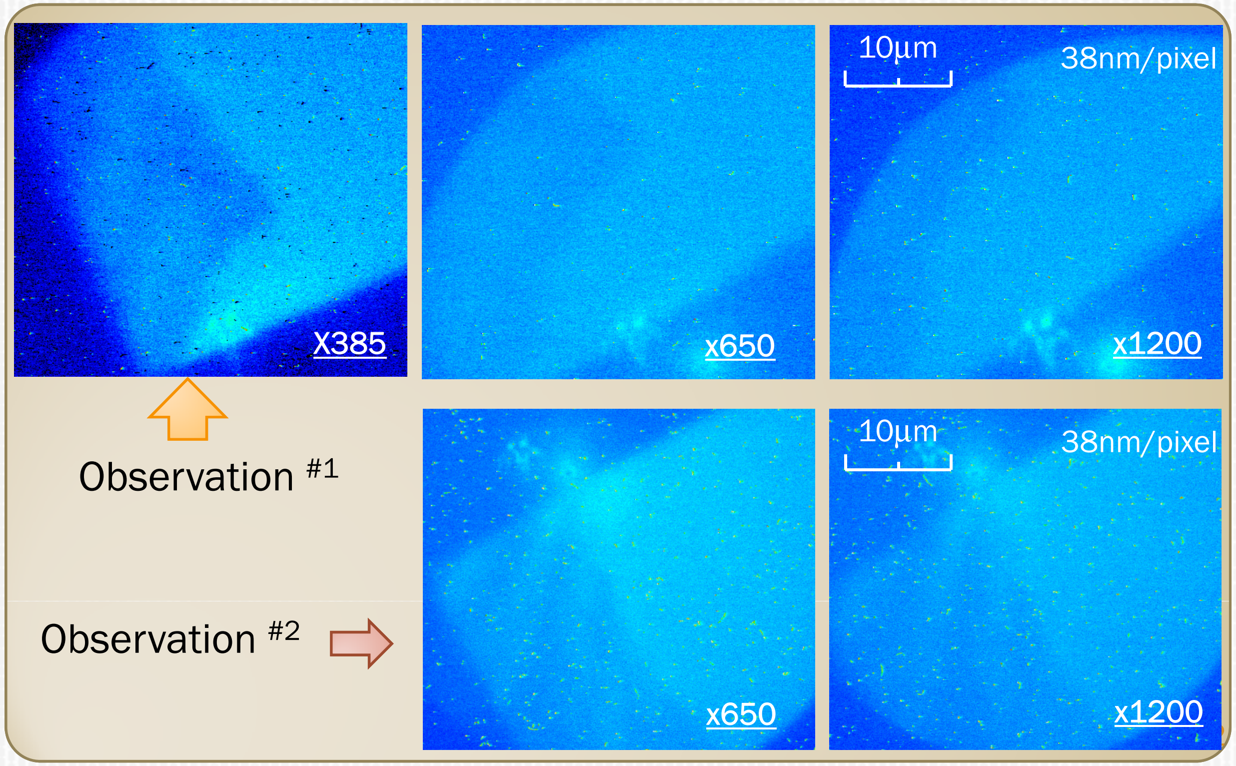
Images of
Au film



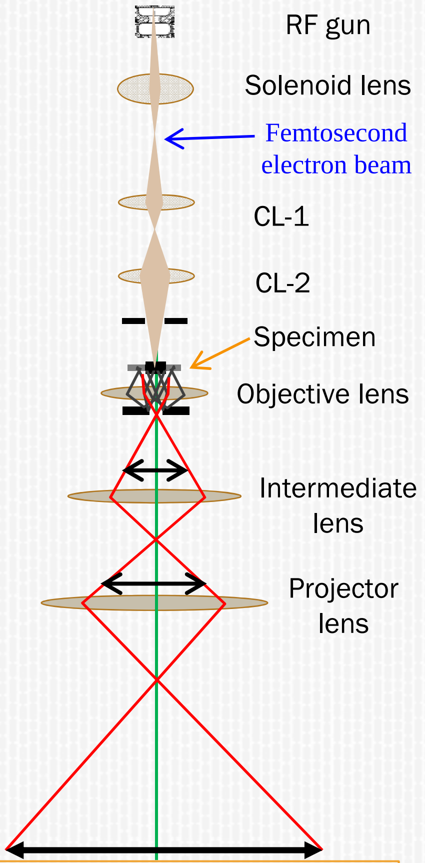
Exposure time：2s

Exposure time：60s

Observed images of Au film



拡大倍率と空間分解能



<u>Spatial resolution</u>

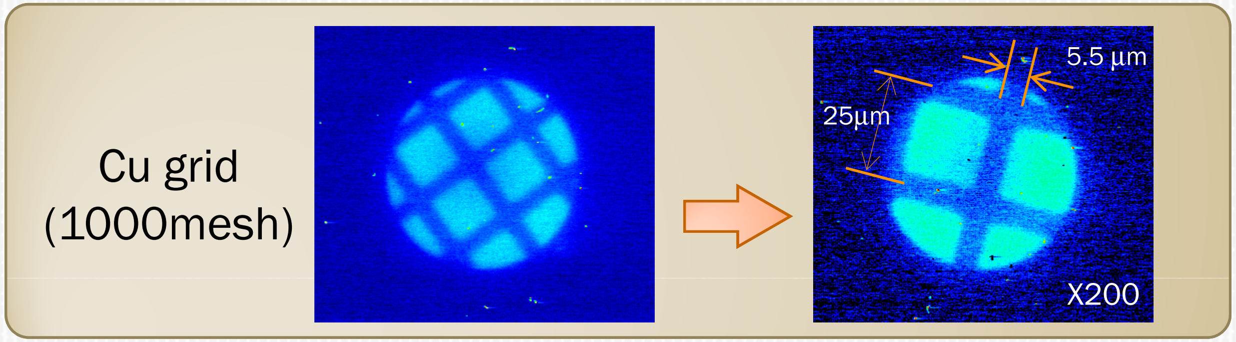
300 nm (present)

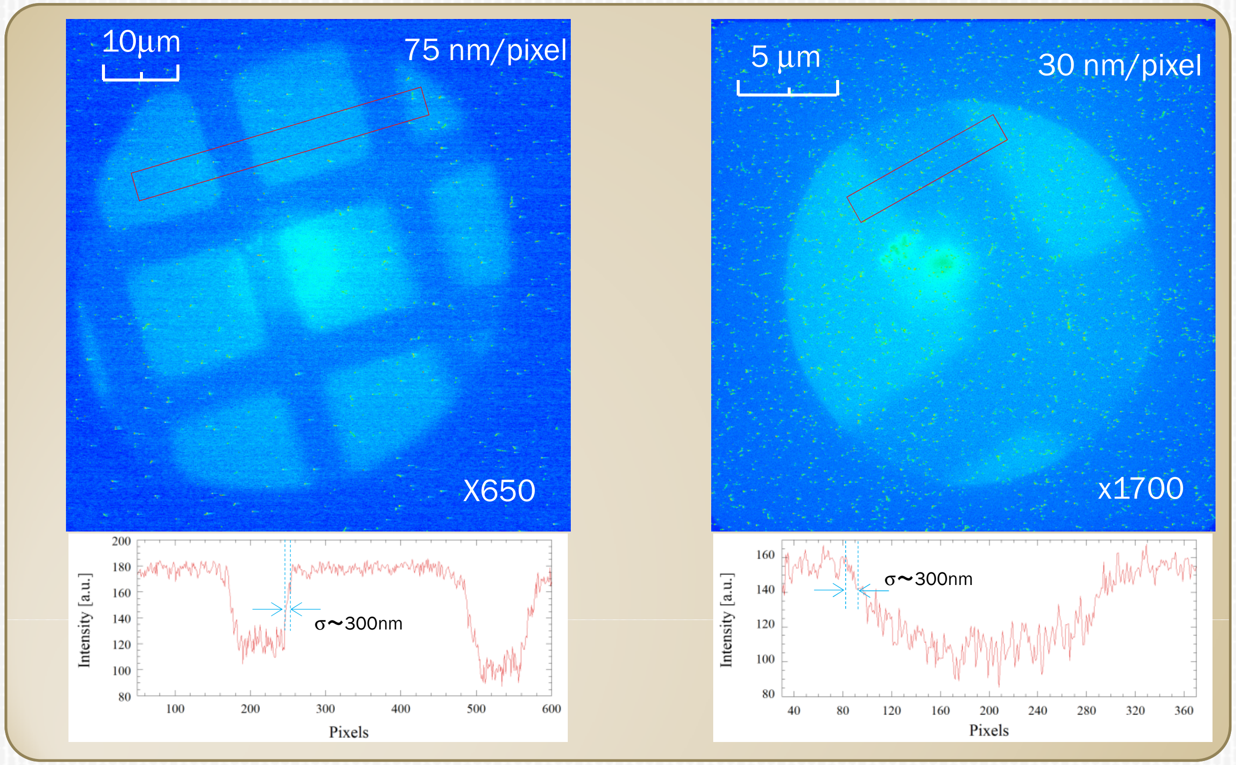
⇩

10nm (next step)

⇩

<1nm (in future)



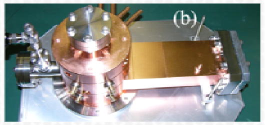


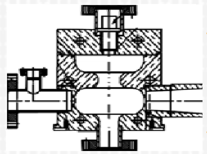
Next TEM: “Dream TEM”

10m

3m

Photocathode RF gun





15cm

- Electron energy : 1~3 MeV
- Bunch length : 100 fs
- Emittance : 0.1 mm-mrad
- Energy spread : 10^{-4}~10^{-5}
- Bunch charge : 10^{7}~10^{8} e^{-}

Compact High-voltage Transmission Electron Microscopy

3m

Next TEM

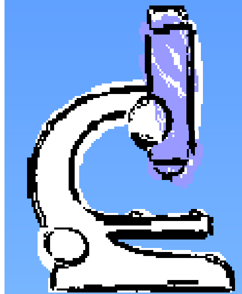
- high-voltage TEM function (nm or sub-nm, MeV)

+

- time-resolved function (femtosecond)

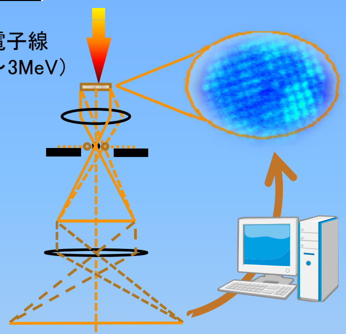
Dream TEM!

時間分解電子顕微鏡が拓く新しい科学技術



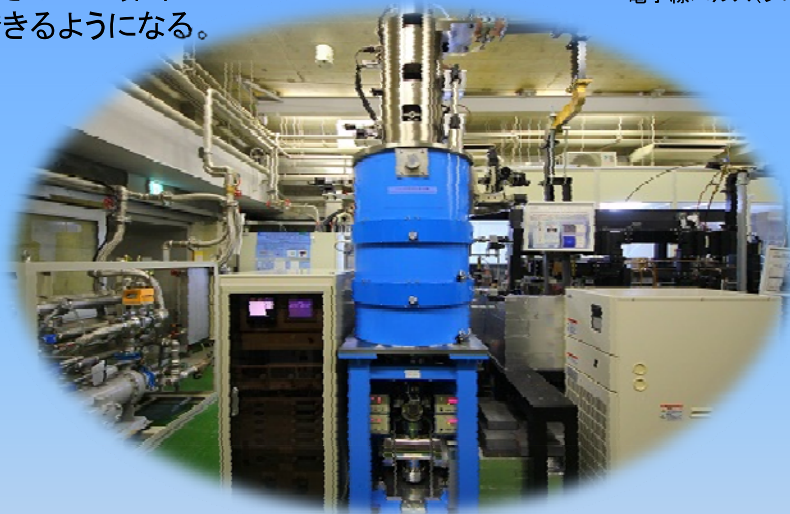
Imaging Technology
イメージング　テクノロジー

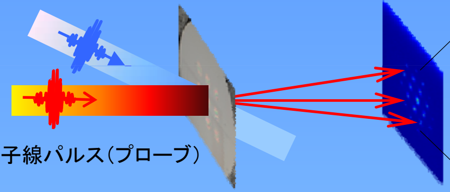
高エネルギー電子を用いると、さまざまな対象物質を原子の大きさほどの非常に細かい分解能で観察できるようになる。

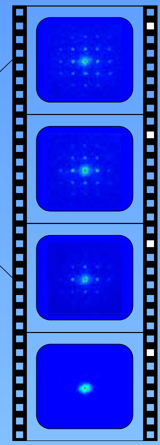


小型超高圧電子顕微鏡の誕生

Methods







Ultrafast Observation
超高速現象の観察

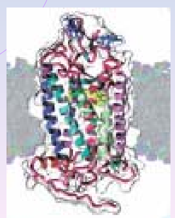
100フェムト秒(10兆分の1秒)以下という短い電子線パルスを用いると、超高速で起こる物質の構造変化を観測することができる。

原子・分子動画技術の実現

Targets

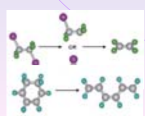
Protein Structural Dynamics
タンパク質構造ダイナミクス

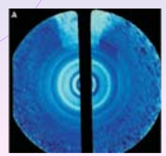
構造ダイナミクスの解析によって、生物学、医療における新しい機能材料が生み出される。



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

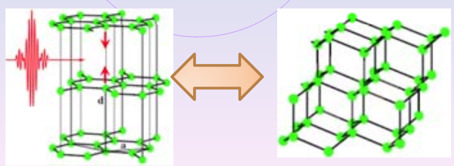
フェムト秒短パルス電子ビームを用いると、分子の運動や生まれ変わりをリアルタイムで見ることができる。





Nano Technology
ナノテクノロジー

構造変化の可視化によって、新機能の発見、新しいデバイスの創製に重要な役割を果たす。



まとめ

✓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. In UEM, the MeV electron imaging experiment was carried out.

✓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 region.