

RF GUN BASED ULTRAFAST ELECTRON MICROSCOPY*

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Abstract

Ultrafast electron microscopy (UEM) would be a powerful tool for the direct visualization of structural dynamic processes in matter. The resolutions of the observation on femtosecond time scales over sub-nanometer (even atomic) spatial dimensions have long been a goal in science. To achieve such resolutions, we have designed and constructed a femtosecond time-resolved relativistic-energy electron microscopy using a photocathode radio-frequency (RF) electron gun (RF based UEM). The RF gun has successfully generated a high-brightness electron beam with bunch length of 100 fs and emittance of near 0.2 mm-mrad, which are essential beam parameters for the achievement of nm-fs space-time resolution in the microscopy. Both the static measurements of both relativistic-energy electron diffraction and image have been succeeded. In this presentation, the activities on RF based UEM are introduced. The requirements and limitations of the beam parameters are reviewed. The concept and design of RF based UEM are reported. Finally, some demonstrations of the relativistic-energy UEM images are reported.

INTRODUCTION

Transmission electron microscopy (TEM) is a powerful tool to observe directly the image from specimen with high spatial resolution. When coupled with time resolution, it, which also called ultrafast electron microscopy (UEM), would be the strongest tool for the study of ultrafast dynamics in materials. Currently, the UEM with the time-spatial resolution of nanosecond and nanometer has been achieved in conventional TEM through the use of photo-activated electron source driven by a nanosecond laser in the non-space-charge-limited regime with ns-long pulse length. A large number of important phenomena, i.e. phase transformations, melting, re-solidification, nucleation and growth of damage in nanosecond time region, have been investigated. To achieve a high time resolution overcoming the space-charge limitation, we have proposed and designed a femtosecond time-resolved relativistic-energy electron microscopy using a photocathode radio-frequency (RF) electron gun. In 2009 [1,2], we have developed a RF gun to generate a low-emittance femtosecond-bunch electron beam: 100 fs and 0.1 mm-mrad, which are essential for the achievement of nm-fs space-time resolution in future. In 2010, we constructed successfully an instrument of ultrafast relativistic-energy electron diffraction (UED) using the RF gun [3-5]. The time resolution of 100 fs has

been achieved. In 2012, a first prototype of RF gun based relativistic-energy TEM (which is called *rf-UEM*) has been constructed at Osaka University [6,7]. The resolutions of 1 nm and 100 fs in spatial and temporal respectively will be challenged. In 2014, a new RF gun with the highest repetition rate of 1 kHz was designed and produced to generate a further low-emittance and low-energy-spread electron beam in *rf-UEM*. Both the static measurements of relativistic-energy electron diffraction and image have been succeeded in the prototype. In this poster, the activities on UED and UEM are introduced. The concept and design of the new RF gun and the prototype of RF gun based relativistic-energy electron microscopy are reported. The beam dynamics and challenges in femtosecond RF gun will be discussed. Finally, some demonstrations of the relativistic-energy TEM images, the single-shot and time-resolved UED measurements are reported.

1 kHz NORMAL CONDUCTING RF GUN

To achieve the aim resolutions of 1 nm and 100 fs in spatial and temporal, an electron source in *rf-UEM* has to be able to generate a low emittance and low energy spread beam, such as 0.1 mm-mrad for the normalized emittance and 10^{-4} or 10^{-5} for the energy spread. In addition, low dark current and ultrahigh stabilities on charge and energy are also required. For these reasons, we have designed and fabricated a new structure RF gun with following optimum considerations and improvements:

- New shapes of the structures in both the RF cavity wall and the iris between the half cell and the full cell are designed near to the ideal contour to reduce the nonlinear electric fields. A large aperture of the iris was used to reduce the electric field on the cavity surface and to reduce both the transverse emittance and energy spread.
- The conventional laser injection ports in the half cell were removed for good field symmetry. A new insertion function of the photocathode in the cathode plate was designed. It will be used to develop a transmission photocathode to generate a further low thermal emittance beam from the RF gun.
- New wall-structural turners were designed in the half and full cells to adjust precisely the field balance.
- The field emission due to the strong electric field between the cathode plate and the half-cell cavity is the biggest problem in old type RF gun. In the new RF gun, the cathode plate was blazed on the half-cell cavity without the use of the helicon flex vacuum shield. The dark current from the new gun was greatly suppressed to <0.01 pC/pulse. The

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photocathode is also removable to change the cathode materials.

- The cooling system was improved in the new RF gun. The water cooling channels were increased from 3 to 4 channels. The heats on the cavity wall in the half cell can be removed by the new cooling channel outside of the half cell. The waveguide is also cooled. The heats near the coupling hole between the waveguide and the full cell are expected to be removed.

Figure 1 shows the cavity structure and the cooling system of the new RF gun. The elliptical shape and a large aperture were used in the new gun not only to reduce the electric field on the cavity surface, but also to reduce the energy spread due to the RF effect and the transverse emittance in the case of operation at the higher gun phase. The mode separation of π -mode and 0-mode was increased to be 15.2 MHz. The new RF gun was designed to be operated under the peak RF power of 3 MW, the pulse duration of 1 μ s, and the repetition rate of 1 kHz. The beam energy is ~ 3 MeV, which is suitable for the electron microscopy.

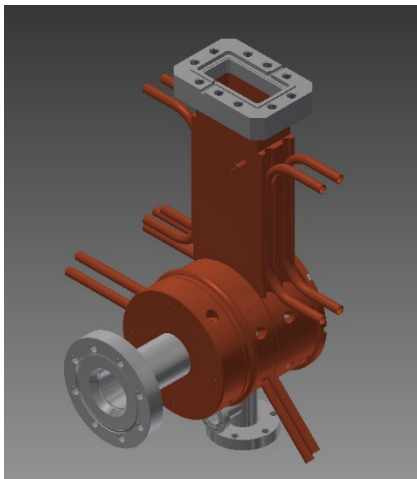
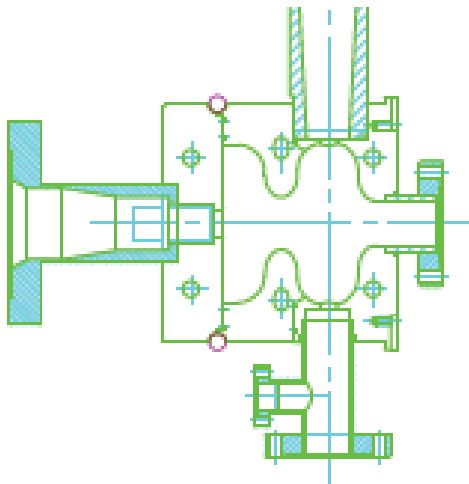


Figure 1: New photocathode RF gun for electron microscopy.

The electron beam generated from the RF gun is propagated to the specimen through a condenser lens and an aperture. The condenser lens and the aperture precisely control and collimate a small-size and small-convergence-angle beams on the sample to reduce the effect of spherical aberration in *rf*-UEM. We measured the normalized transvers beam emittance as a function of the aperture diameter, as shown in Fig. 2. In the measurement, the laser spot size on the copper cathode was 2 mm in diameter. The laser pulse width was near 90 fs in FWHM. The beam energy was 3.0 MeV. The previous experiments suggest a 0.1 mm-mrad low emittance electron beam can be achieved using an aperture diameter of 0.5 mm or less. This value of the beam emittance would be required to achieve the spatial resolution of \sim nm in the electron microscopy.

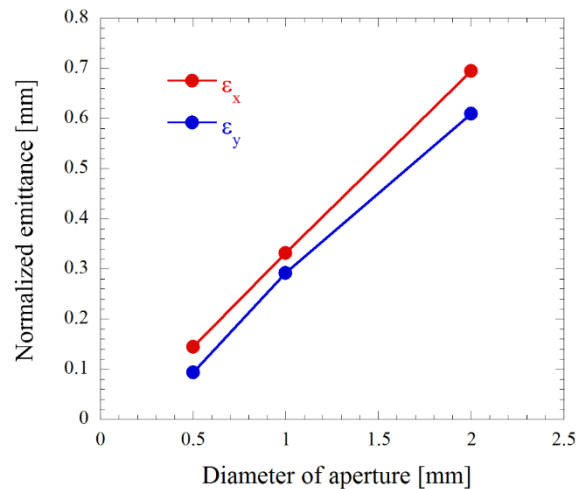


Figure 2: The normalized transverse emittance of the femtosecond electron beam generated from the RF gun as a function of the aperture diameter.

PROTOTYPE OF RF GUN BASED RELATIVISTIC-ENERGY ELECTRON MICROSCOPY

Figure 3 gives the picture of the prototype of RF gun based relativistic-energy electron microscopy, which was constructed in 2012 [6,7] at Osaka University and improved in 2014. The new RF gun as described above was used in the prototype. After the RF gun, we used a condenser lens and an aperture to precisely control and collimate a small-size and small-convergence-angle beam on the sample. The image of the sample is formed with three kind electron lenses: an objective lens, an intermedia lens and a project lens. The details of the electron lenses were reported and published in IPAC2014, as shown in refs. 6 and 7. Finally, the image is detected by a scintillator of Tl doped CsI equipped with fiber optic plates with a CCD camera. The detection area of the scintillator is 50x50 mm², and the spatial resolution is 50 μ m. The optical image from the scintillator is reflected at 45° into the CCD camera while passing the electron beam through the mirror to prevent electron and X-ray

irradiation of CCD sensor. The detection system was succeeded for the single-shot ultrafast electron diffraction measurement with the electron number of 10^5 in bunch, as described in refs. 3, 4 and 5.

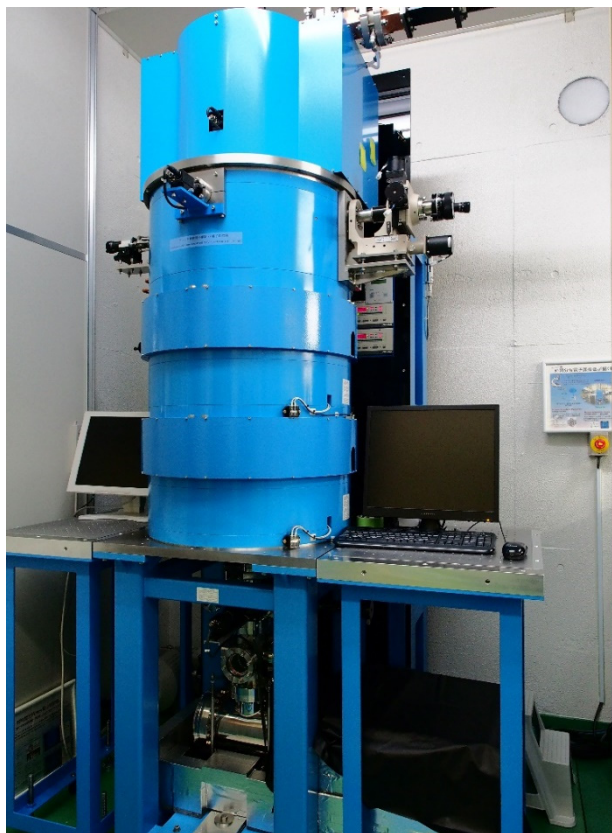


Figure 3: The prototype of RF gun based relativistic-energy electron microscopy which was constructed at Osaka University in 2012 and improved in 2014.

For the first demonstration using the prototype, we observed the relativistic-energy electron diffractions from a 10-nm-thick single-crystal gold film and a near 100-nm-thick single-crystal Mica ($\text{K}(\text{Fe,Mg})_3(\text{AlSi}_3\text{O}_{10})(\text{OH,F})_2$) with single-pulse, 10-pulse and 100-pulse averaging measurements, as shown in Fig. 4. A near 90-fs-bunch MeV electron beam generated from the RF gun was used in the observations. The electron charge was 0.1 pC ($\sim 10^6$)/bunch at the sample after the collimation with a 0.5-mm-diameter condenser aperture. The excellent quality of diffraction patterns were acquired by averaging 10 electron pulses. The single shot measurement is also available in the prototype TEM. The observed sharp diffraction patterns suggest that a low-emittance and low-energy-spread beam after the aperture collimation has been generated from the RF gun.

In the TEM image observations, the magnifications of images were obtained to 200 times using the objective lens only, 900 times using the objective and intermediate lenses, and 3,000 times using the three kind lenses. The best resolution was 16 nm/pixel, which were reported and published in IPAC14 [7]. In the next step, we will reduce further the thermal emittance of the electron beam by

focusing the UV laser spot on the cathode, and focus the electron beam to micrometre at the sample to increase the beam brightness. Finally, we will increase the image magnification up to 10,000 and then to improve the spatial resolution to <10 nm.

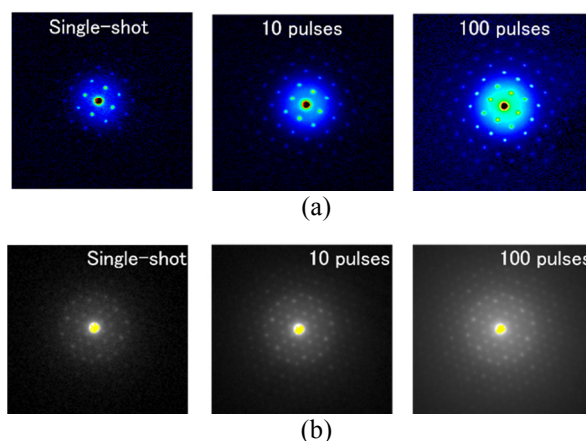


Figure 4: Relativistic-energy electron diffractions observed from single-crystal gold (a) and single-crystal Mica (b) with single-pulse, 10-pulse and 100-pulse averaging measurements.

CONCLUSION

The femtosecond laser driven photocathode RF gun is a high-brightness femtosecond electron source and is used successfully for the relativistic-energy UED to study the structural dynamics in matter. It is also very expected to be a significant benefit in the development of a femtosecond time-resolved electron microscopy. However, many efforts and challenges are required: (1) it is required to reduce further the transverse emittance and the energy spread. (2) The stabilities on the charge and the energy would be improved, and (3) the detection of every electron is also essential in future developments because of small signal levels.

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