FEMTOSECOND TIME-RESOLVED TRANSMISSION ELECTRON MICROSCOPY USING RF GUN*

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Abstract

The first prototype of RF gun based relativistic-energy electron microscopy has been constructed at Osaka University to study ultrafast structural dynamic processes in matter. The RF gun driven by a femtosecond laser has generated a 100-fs-pulse MeV electron beam with emittance of 0.1 mm-mrad and energy spread of 10^-4. Both the electron diffraction and image measurements have been succeeded in the prototype using the femtosecond electron beam. In the diffraction measurement, an excellent quality of diffraction pattern was acquired with electron number of 10^6. The single-shot measurement is available in the prototype. In the image measurement, the TEM image was acquired with a total electron number of 10^8. The magnification was 3,000 times. In the next step, we will reduce further the emittance to increase the beam brightness on the sample, and then improve the spatial resolution to <10 nm.

INTRODUCTION

Photocathode radio-frequency (RF) electron gun is a useful source to directly produce a relativistic-energy femtosecond-bunch electron beam at the low bunch charge of pC or less. Recently, several facilities of ultrafast relativistic-energy electron diffraction (UED) using RF gun have been constructed or proposed in the world to study ultrafast structural dynamics or phenomena in matter occurring on femtosecond time scales [1-7]. The time resolution of UED has been achieved to 100 fs using a 100-fs-bunch MeV-energy electron beam. Although UED can be used to understand many ultrafast dynamics such as laser heating, melting and so on, however, the UED studies are limited to the periodical phenomena in the materials. The UED data are insufficient to clarify the structural dynamic processes concerning the motion of individual atom or molecule, because of no spatial resolution in UED observation. If the electrons in matter are excited by the laser photons or X-rays, the excited electron relaxes, and then transfers its energy to the atoms or molecules. Finally, the new structure will be created by phase transformation if the sufficient energy transfer is occurred. Therefore, to elucidate such structural dynamics, the spatial resolution over the atomic scale are indispensable. The resolution of 100 fs and sub-angstrom has long been in goal for the material scientists.

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 called ultrafast electron microscopy (UEM), also called dynamic transmission electron microscopy, (DTEM), would be the strongest tool for the study of ultrafast dynamics in materials [8-10]. Currently, the DTEM with the 15 ns time resolution has been achieved in conventional TEM through the use of photo-activated electron source driven by a nanosecond laser [9]. A large number of important phenomena, i.e. phase transforma-tions, melting, resolidification, nucleation and growth of damage, have been investigated. Most widely used UEM instruments are based on laser-driven photoemission DC guns, generating typically ~100 KeV electron beams [10]. The time resolution is achieved by operating in the non-space-charge-limited regime with ns-long pulse or single-electron pulse, because the electron pulse length is increased due to the space-charge broadening during propagation in nonrelativistic beams. The energy spread also increases to be larger than 10^{-3} due to the space charge effect during propagation, resulting in the degradation of spatial resolution. There is no resolution to achieve the time-spatial region of femtosecond and sub-nanometer in the recent UEM or DTEM.

To overcome the problems, we have designed and constructed a femtosecond time-resolved relativistic-electron microscopy using a photocathode RF gun at Osaka University. The project was started from 2010. A new structural-cavity femtosecond-pulse electron RF gun was developed in 2011. In 2012, a prototype of relativistic-energy TEM has been constructed using the RF gun. Both the relativistic-energy electron diffraction and image measurements have been succeeded in the prototype [11].

THE FIRST PROTOTYPE OF TIME-RESOLVED RELATIVISTIC-ENERGY TEM

The photo of femtosecond time-resolved relativistic-energy TEM is shown in Fig. 1. It consists of a photo-cathode RF gun, an injection column, imaging optical lenses and an image measurement system. The temporal and spatial resolutions of 100 fs and <10 nm are expectable to be achieved.

A Femtosecond Photocathode RF Gun

The electron source required for TEM has to be able to generate a low emittance and low energy spread beam with low dark current and ultrahigh stabilities on charge and energy. For this reason, we have developed a new

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structure RF gun with following optimum design and improvements:

- A shape of the RF cavity wall is designed near to the ideal wall contour to produce an optimum electric field reducing the Fourier coefficients of all higher harmonics.
- The conventional laser injection ports in the half cell were removed for good field symmetry.
- A new wall turner system was designed to adjust precisely the field balance in the half and full cells.
- 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. A new insertion function of the photocathode was designed. 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 <0.1 pC/pulse. The expected beam parameters is given in Table 1.

Table 1: The Expected Beam Parameters

Beam energy	1~3 MeV
Bunch length	100 fs or less
Emittance	0.1 mm-mrad
Energy spread	10^{-4} (10^{-5} for challenge)
Bunch charge	$10^7 \sim 10^8$ e^-/pulse

Figure 1: Prototype of femtosecond time-resolved relativistic-energy electron microscopy using RF gun.

An Injection System

The electron beam generated from the RF gun is propagated to the specimen through the injection system. The injection system constructed with a solenoid magnet and two condenser lenses. The solenoid magnet is used to compensate the emittance growth during the beam propagation due to space-charge effect. The condenser lenses with a diaphragm precisely control and collimate a small-size and small-convergence-angle beams on the sample. The effect of spherical aberration, as a limitation of spatial resolution, is a significant problem in TEM. It can be described by $\sim C_s\theta^3$, where C_s is a spherical aberration coefficient and θ is the convergence angle of beam on the sample. For relativistic TEM, C_s ~ a few mm. The previous experiments [4] suggest a 0.1 mm-mrad low emittance electron beam can be achieved in the RF gun by reducing the laser spot size of 0.2 mm on the copper cathode. By using the injection system, we have controlled the convergence angle $\theta < 1$ mrad on the sample by adjusting the focusing strength of the condenser lenses and by collimating the beam with the condenser aperture. The limitation due to the spherical aberration is $C_s\theta^3 << 0.1$ nm.

An Imaging System

The imaging system in the prototype of TEM consists of an objective lens, an intermediate lens and a projector lens: the objective lens (OL) to provide a back-focal plane (BFP) for expanded images, and the intermediate lens (IL) and the projector lens (PL) to display the images with desired fashion on the detector. The pole pieces in the three lenses are made by the Fe-Co alloy materials and are fabricated precisely. The lenses can provide the maximum magnetic field of 2.2 T in the center of the pole pieces with the maximum Ampere-turns of 38,000 AT. The designed magnification of the image in the prototype is 5,000~125,000 for beam energy of 1~3 MeV. The spatial resolution of <10 nm is achievable using the lens system with the given electron beam parameters in Table 1.

To achieve high sensitivity and a high damage threshold of MeV electrons, a scintillater of CsI(Tl) equipped with fiber optic plates was used. The detection area of the scintillater is 50x50 mm^2, and the spatial resolution is 50 μm [4]. The optical image from the scintillator is then reflected at 45° into an electron multiplying CCD camera while passing the electron beam through the mirror to prevent electron and X-ray irradiation of CCD sensor.

EXPERIMENTAL RESULTS

MeV Electron Diffraction

Figure 2 gives the relativistic-energy electron diffraction patterns observed from a 15-nm-thick single-crystal gold film with single-bunch and 10-bunch averaging measure-ments in the prototype. A 100-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 condenser aperture collimation. The excellent quality of diffraction pattern was acquired by averaging 10 electron bunches. The observed sharp diffraction patterns suggest that a low-emittance and low-energy-spread beam generated from the RF gun.

The single-shot diffraction measurement is also available in the prototype TEM. However, the S/N ratio is insufficient in the observation, because we used a normal CCD camera with no amplification function in order to improve spatial resolution and to reduce x-ray noises. Since these radiation noises were found to be very low in our detection system, in the next step, we can enhance the sensitivity more than a factor of 10 or 100 by replacing the camera with one equipped with a state-of-art on-chip multiplication function, i.e. EMCCD camera which used successfully in another relativistic-energy UED facility [4].

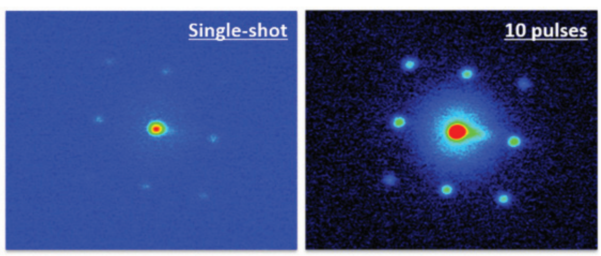

Figure 2: Relativistic-energy electron diffraction patterns of single-crystal gold observed with single-bunch and 10-bunch averaging measurements in the prototype.

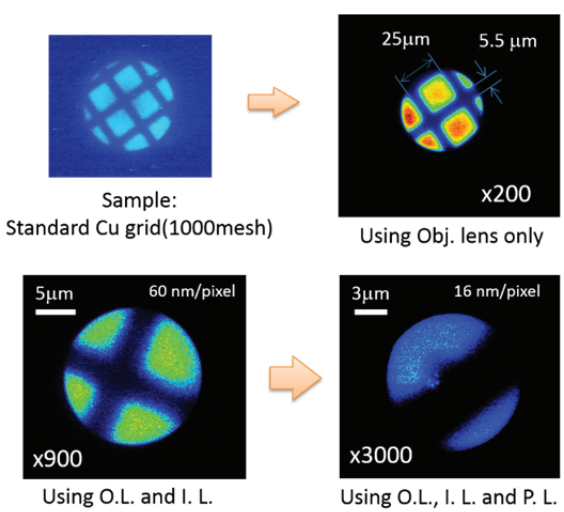

Figure 3: Relativistic-energy TEM images observed by the prototype.

MeV-Energy TEM Image

In the TEM image observations, a standard copper grid with 1000 meshes was used to determine the magnification. Figure 3 shows the relativistic-energy TEM images observed under the different conditions. The emittance of the electron beam was 0.2 mm-mrad. The electron beam was not focused on the sample to avoid the space-charge effect. The beam size at the sample was 1 mm in diameter. 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 TEM lenses. The best resolution was 16 nm/pixel. The data suggest the sufficient-quality image would be acquired with a total electron number of 10^8. In the next step, we will reduce 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 100,000 and then to improve the spatial resolution to <10 nm.

CONCLUSION

The 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 next femtosecond time-resolved TEM. It is will-known that the RF gun is very compact relativistic-energy electron source. The RF gun based TEM must be very compact, and would be a “dream” TEM for the next electron microscopy if both the temporal-spatial resolutions of 100 fs and angstrom can be achieved. However, many efforts and challenges are required:

- One is required to reduce further the transverse emittance and the energy spread. For a low charge beam, the emittance can be minimized by reducing the laser spot size on the cathode, i.e. 50 μm or less. For the RF gun, the energy spread is limited to $\Delta E/E \sim 10^{-4}$ using a 100 fs laser. It is possible theoretically to reduce to 10^{-5} using a few tens femtosecond-long laser.
- Finally, the stabilities (charge and energy) and the synchronization of the laser with accelerating RF are needed to be improved.
- The detection of every electron is also essential in future developments because of small signal levels.

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