ULTRAFAST ELECTRON MICROSCOPY USING 100 FEMTOSECOND RELATIVISTIC-ENERGY ELECTRON BEAM*

Jinfeng Yang#, Koichi Kan, Katsumi Tanimura, Yoichi Yoshida, ISIR, Osaka, Japan
Junji Urakawa, KEK, Ibaraki, Japan

Abstract

An ultrafast detection technique on 100 fs time scales over sub-nanometer (even atomic) spatial dimensions has long been a goal for the scientists to reveal and understand the ultrafast structural-change induced dynamics in materials. In this paper, the generation of femtosecond electron pulses using the RF gun and the first prototype of femtosecond time-resolved relativistic-energy ultrafast electron microscopy (UEM) are reported. Finally, both relativistic-energy electron diffraction and image measurements in the UEM prototype are presented.

INTRODUCTION

Ultrafast electron microscopy (UEM), which has not only spatial resolution and but also has temporal resolution, would be the strongest tool for the study of ultrafast dynamics in materials. Currently, all of UEMs are using a photocathode DC electron gun to generate two kind pulse electron beams: One is a long-pulse electron beam with high bunch charge such as 10 ns and 10^8 electrons in pulse. Lawrence Livermore National Laboratory has succeeded to achieve 10-ns and 10-nm time-spatial resolutions with single-shot observation in UEM [1]. Another is a single-electron pulse beam with high repetition-rate, i.e. 80 MHz, which is developed typically in California Institute of Technology, USA [2]. They have improved the time resolution to 700 fs. 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. However, the DC electron guns in the present UEMs are very difficult to generate a femtosecond-pulse electron beam with high bunch charge, because of the space-charge limitation for the non-relativistic-energy electron beam.

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 [3-9]. In 2012, a first prototype of RF gun based UEM has been constructed at Osaka University [8]. The resolutions of 1 nm and 100 fs in spatial and temporal respectively will be challenged. In 2014, we succeeded to generate a 100-fs-pulse electron beam with energy of 3.1 MeV using the RF gun driven by a femtosecond laser [9]. Using such electron pulses, we succeeded to observe excellent-quality electron diffraction patterns under the single-shot observation. In 2015, we upgraded the lens system and the detection system for the transmission electron microscopy (TEM) measurement. In 2016, we succeeded to observe TEM images of polystyrene particles with the diameters of 1.1 and 0.5 µm using the 3.1-MeV-energy femtosecond electron pulses. Other material imaging have been also demonstrated.

THE FIRST PROTOTYPE OF RF GUN BASED UEM

The first prototype of RF gun based UEM has been constructed in the end of 2012 at Osaka University. The prototype is consisted of an S-band photocathode RF gun, femtosecond electron pulse injection lens system, a TEM imaging system and a MeV image detection system, as shown in Fig.1.

To achieve the aim resolutions of 1 nm and 100 fs in spatial and temporal, an electron gun has to be able to generate a low-emittance and low-energy-spread beam, i.e. 0.1 mm-mrad for the 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 in 2014 and 2015 with following optimum considerations and improvements, i.e. new-shape RF cavities, a large iris between the half cell and the full cell to reduce the electric field on the cavity surface and to reduce both the transverse emittance and energy spread. The new RF gun was designed for the operation under 1 kHz in future. The details have been reported on the last IPAC15 [9].

The injection lens system is consisted of two condense lenses and an aperture with three pin holes to control and focus the electron beam on the sample. The diameters of the pin holes are 0.5, 1 and 2 mm. In order to generate a lowest emittance electron for UEM, we reduced the thermal emittance firstly by focusing UV laser on the photocathode. The copper cathode was used. The laser spot size was 0.3 mm in diameter or less. Moreover, we collimated the large-divergence electrons after the RF gun using the aperture to reduce furthermore the beam emittance. As a result, we succeeded to generate a 0.14 mm-mrad electron beam using the present injection lens system. Please see the proceedings of IPAC15 for the details of the low-emittance generation [7].

In the TEM imaging system, we used three magnetic lenses, i.e. an objective lens, an intermediate lens and a projective lens as the simplest imaging lens system in the microscopy. The objective lens can generate a strong magnetic field up to 2.2 Tesla. The focal length is around 5 mm for the electron beam with the energy of 2 MeV. The

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#yang@sanken.osaka-u.ac.jp

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TEM imaging system is able to make a TEM image with the magnification of 10,000 times for the 2 MeV electron beam.

Figure 1: The first prototype of UEM using RF gun, which was constructed at Osaka University in 2012 and upgraded in 2014 and 2015.

Finally, the relativistic-energy TEM image is detected by a scintillator of Tl doped CsI equipped with fiber optic plates with an EMCCD camera. The detection area of the scintillator is 50x50 mm^2, and the spatial resolution is 50 μm. 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 and the time-resolved electron diffraction measurement as described in refs. 5, 6 and 7.

DEMONSTRATIONS OF MEV-ENERGY ELECTRON DIFFRACTION AND MICROSCOPY IMAGING IN UEM

Figure 2 gives the relativistic-energy electron diffraction patterns observed in the prototype UEM with single-shot and 10-pulse accumulating measurements using 3.1 MeV femtosecond electron pulses. The sample was a single-crystal gold with the thickness of ~10 nm. The beam emittance was 0.14 mm-mrad and the bunch charge was about 1 pC. The most important result is the success of the single-shot observation. It provides us a possibility to study the non-reversible dynamics in materials. The diffraction images indicate that high-quality electron diffraction patterns are able to be obtained in our UEM with the diffraction measurement mode. It suggests that the electron beam quality is high and the electron lenses in UEM have high performance and work very well.

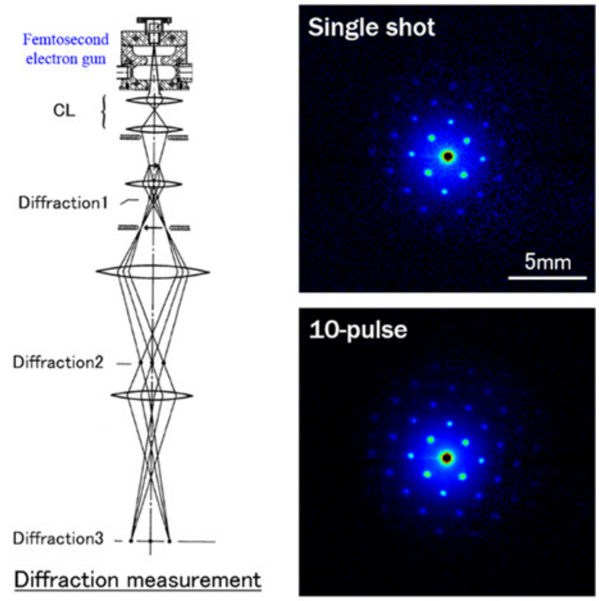

Figure 2: Electron diffraction images of single-crystal gold obtained in our UEM using a 3.1 MeV femtosecond-pulse electron beam with single-shot and 10-pulse accumulating measurements.

Figure 3 gives the first demonstration of TEM imaging using the 3.1 MeV femtosecond-pulse electron beam in our UEM. The sample is a standard TEM copper grids with 1,000 meshes. The top three images were observed with the beam normalized emittance of 0.6 mm-mrad and the bunch charge of 7 pC (the brightness is $6x10^9$ e$^-$/mm^{2}mrad^2). The mid three images were observed with the beam emittance of 0.3 mm-mrad and the bunch charge of 4 pC (the brightness is $1.4x10^{10}$ e$^-$/mm^{2}mrad^2), and the bottom three images were observed with the beam normalized emittance of 0.14 mm-mrad and the bunch charge of 1 pC (the brightness is $1.6x10^{10}$ e$^-$/mm^{2}mrad^2). The left, mid and right images were obtained with single-shot, 10 and 100 pulses accumulating measurements, respectively. All of the images were detected with the magnification of 500 times.

Firstly, the low-emittance electron beam is very essential in the electron microscopy. The contrast of the images is very dependent on the beam emittance. Presently, the demonstrations indicate that a good contrast image is able to be observed with the low-emittance beam, i.e. 0.14 mm-mrad. Anyway, the normalized emittance of 0.1 mm-mrad or less is necessary to reduce the spherical aberration from the electron lenses. The low-emittance electron beam has also a high brightness even the bunch charge is small, because the beam is able to be focus on the sample.

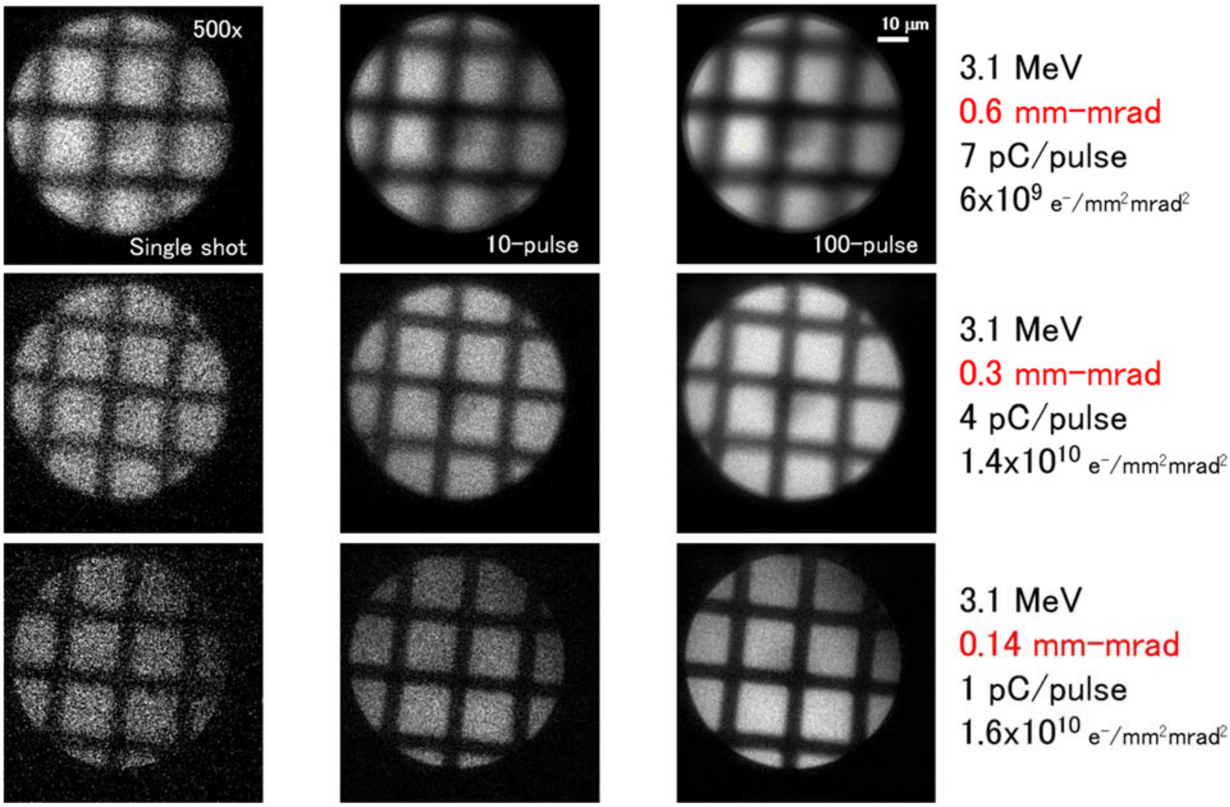

Figure 3: Transmission electron microscopy images of standard copper grids obtained in our UEM as a function of emittance using a 3.1 MeV femtosecond-pulse electron beam with single-shot, 10-pulse and 100-pulse accumulating measurements.

Secondly, at the observation with the low magnification such as 500 times, there is a possibility of the single-shot imaging in our UEM. Of cause, the brightness is still not enough in the demonstrations. Anyway, it is very expected if we can improve furthermore the beam emittance.

CONCLUSION

The first prototype of relativistic-energy ultrafast electron microscopy using a photocathode RF gun has been developed. The demonstrations of both electron diffraction and microscopy imaging have been succeeded using a 100-fs-pulse electron beam with energy of 3.1 MeV. In the electron diffraction measurement, the single-shot observation is succeeded. In the electron microscopy measurement, the dependence of image contrast on the emittance was investigated. The data indicates that, under the condition of low-magnification observation, the single-shot imaging is very expected in our UEM using 3. MeV femtosecond-pulse electron beam. Other material imaging have been also demonstrated. Our development and demonstrations suggest that the photocathode RF gun is a high-brightness femtosecond electron source 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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