RF GUN BASED MEV HIGH-VOLTAGE ELECTRON MICROSCOPY

Jinfeng Yang [#,A)], Koichi Kan[A)], Nobuyasu Naruse [A)], Takafumi Kondoh [A)], Yoichi Yoshida [A)], Katsumi Tanimura [A)], Toshikazu Takatomi[B)], Jyunji Urakawa[B)]

[A)] The Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan

[B)] High Energy Accelerator Research Organization , 1-1 Oho, Tsukuba, Ibaraki, Japan

Abstract

The direct visualization of fundamental dynamic processes in matter occurring on femtosecond time scales over sub-nanometer (even atomic) spatial dimensions has long been a goal in science. The electron microscopy is a powerful tool to observe directly the image from specimen with high spatial resolution, however generally without time resolution. The photocathode radio-frequency (RF) gun is a short-pulse electron source to generate a high-brightness femtosecond electron beam with energies of 1~3 MeV. The RF gun based relativistic ultrafast electron diffraction is a promising new technique that has the potential to probe directly structural changes at the atomic scale with sub-100 fs temporal resolution. In this paper, the development of a time-resolved relativistic electron microscopy based on the RF gun is reported. The requirements and limitations of the beam parameters used in this technique are discussed.

INTRODUCTION

The direct visualization of fundamental dynamic processes in matter occurring on femtosecond time scales over sub-nanometer (even atomic) spatial dimensions has long been a goal in science. Ultrafast electron diffraction (UED) provides a direct measure ("real-time" probe) of structural dynamics in matter by recording the change in the characteristics of electron diffraction peaks (position, intensity, width) in the pump state and the unpump state. The UED technique has the key advantage with respect to X-ray diffraction of a much larger interaction cross section in the matter. A large number of important results, i.e. phase trans-formations, melting, ultrafast breaking of chemical bonds and ultrafast reactions, have already been obtained with picosecond or sub-picosecond temporal resolution. Most widely used UED instruments are based on laser-driven photoemission DC guns, generating typically 50-100 keV electron beams [1,2]. The time resolution is achieved by operating in the non-space-charge-limited regime with thousands of electrons per pulse, because of space-charge broadening during propagation in nonrelativistic beams. For example, in the theoretical calculation for a 30 keV femtosecond electron bunch with an electron number of 10,000 per pulse, the length of the electron bunch increases to ~4 ps by propagating with a distance of 40 cm [3]. The energy spread also increases due to the space charge effect during propagation. To overcome the problem, one is to minimize the propagation distance by placing the sample in close proximity of the electron source (~ 4.5 cm); another is to decrease the number of electrons in the beam, i.e. 1,000 e-'s in a 600 fs bunch. However, the technique of UED cannot observe the structural dynamics in real-space (imaging over atomic spatial dimensions).

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 material science, physics, chemistry and biology. 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 [4]. In order to obtain high time resolution, a stroboscopic imaging of periodically driven processes was preformed. In this configuration, a time resolution of 200 ps was achieved for processes up to 100 MHz, which called the single-electron-pulse method [5]. These techniques were applied to ultrasonically driven disruption of crystals and magneto-elastic effects and magnetic-field-induced oscillations of the domain magnetization of domain walls and of their substructures. However, there is no resolution to achieve the femtosecond-temporal and sub-nanometer (even atomic) spatial resolution in the recent UEM, because of the long electron bunch length and low bunch charge due to the space-charge effect.

It is well known that the photocathode RF electron gun have definitely been a fundamental technological breakthrough in the last 20 years as relativistic electron sources and are in fact the preferred elect. The RF gun is also a significant benefit in UED, because it can generate a femtosecond relativistic electron pulse reducing the space-charge limitation. Recently, the single-shot MeV electron diffraction measurement and the time-resolved measurement have been also succeeded by research groups in the world [6-11]. The studies suggest that the photocathode RF gun is useful for the diffraction measurement with high time resolution. In this paper, we report the design and construction of a new time-resolved near-relativistic electron microscopy using a photo-cathode RF gun to study the atomic dynamics of phase transitions in solids. The project was started from 2010. A new structure femtosecond electron RF gun was developed in 2010. The beam study and the RF gun based

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

UED experiments have been carried out in 2011. In 2012, a prototype of time-resolved near-relativistic TEM has been constructed.

PROTOTYPE OF TIME-RESOLVED RELATIVISTIC ELECTRON MICROSCOPY

The concept of time-resolved relativistic TEM is shown in Fig. 1. It consists of a photocathode rf gun, an injection system, imaging optical lens system and image measurement system. The TEM is not only compact, but also the temporal and spatial resolutions of 100 fs and <10 nm are expectable to be achieved.

Femtosecond RF gun

The electron source required for TEM has to be able to generate a low emittance and low energy spread beam with ultrahigh stabilities on charge and energy, and low dark current. For this reason, we have developed a new structure RF gun under the KEK/Osaka University collaboration in 2010 [12]. At the level of 0.1 mm-mrad or less, the emittance can be affected by a number of small contributions like field asymmetries, the structure and fabrication of the RF cavity and so on. To reduce these effects, the new RF gun has been developed with following optimum design and improvements:

(1) A new structure of the RF cavity was used. The shape of the RF cavity wall is near to the ideal wall contour to produce an optimum electric field reducing the Fourier coefficients of all higher harmonics. The Q-value of the new RF cavity was 16,300, which is 1.4~1.6 times higher than the old RF gun.

(2) The conventional laser injection ports in the half cell were removed for good field symmetry. The field asymmetries not only lead to an asymmetrical emittance resulting in the emittance growth, but also cause a distortion on UED image.

(3) A new wall turner system was designed to adjust precisely the electric field balance in the half and full cells. The dark current produced from the turner antennas in old gun is also avoided.

(4) The field emission due to the strong electric field between the cathode plate and the half-cell cavity is the biggest problem in the old type RF gun. To minimize the field emission, a new insertion function of the photocathode was designed in the new gun as shown in Fig. 5. The cathode plate was blazed on the half cell cavity without the use of the helicon flex vacuum shield without the use of the helicon flex vacuum shield. The dark current from the new gun was greatly suppressed to <0.1 pC/pulse.

(5) The photocathode in the new gun is removable. Finally, the RF gun is driven by a 90-fs Ti:Sapphire laser. A copper cathode is used in the electron microscopy experiment. The expected beam parameters are listed 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 or less
Energy spread	10^{-4} (10^{-5} for challenge)
Bunch charge	10^7 ~ 10^8 e^-/pulse

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 ~$C_s\theta^3$, where C_s is a spherical aberration coefficient and θ is the convergence angle of beam on the sample. For MeV TEM, C_s ~ a few mm. The previous experiment suggests 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 can control the convergence angle $\theta < 2$ mrad on the sample. The

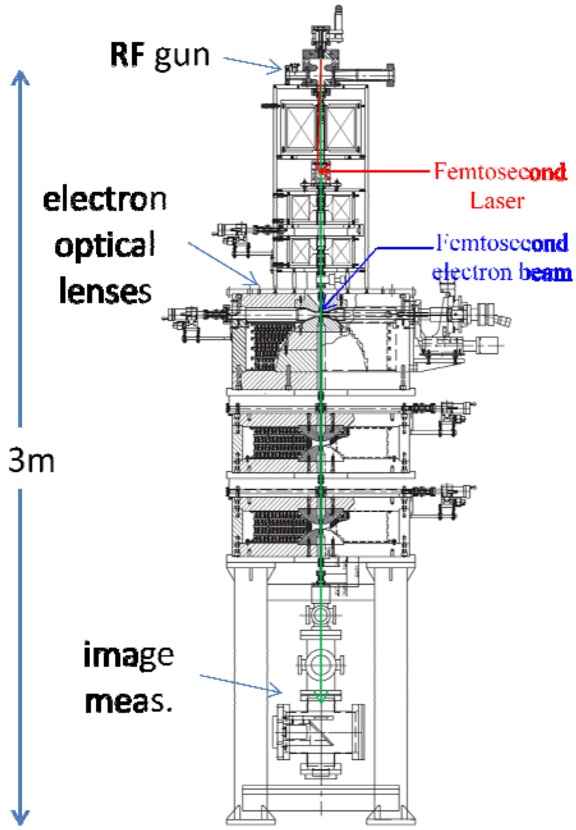

Figure 1: Concept of time-resolved relativistic transmission electron microscopy (TEM) using RF gun

limitation due to the spherical aberration is less than $C_s\theta^3 < 0.1$ nm.

Imaging optical Lens 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 fabricated precisely. The lenses can provide the maximum magnetic field of 2.4 T in the center of the pole pieces with the maximum Ampere-turns of 38,000 AT.

The magnification (M) of the image in the prototype is 5,000~125,000 for the electron 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 I.

Image Measurement System

To achieve high sensitivity and a high damage threshold of MeV electrons, a scintillater of CsI(Tl) equipped with fiber optic plates was used to convert the diffraction pattern into an optical image with a spatial resolution of 50 μm. The size of the scintillater is 50 x 50 mm^2. 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 the CCD sensor.

Figure 2: Prototype of RF gun based time-resolved relativistic TEM. The height is 3 m and the diameter is 0.7 m.

The detecting system has been used successfully in the relativistic UED experiments [8]. The single-shot diffraction patterns with 5 x 10^5 ~ 1 x 10^6 electrons are able to be recorded using the detector.

CONCLUDING REMARKS

The RF gun is a high-brightness femtosecond electron source and is used successfully in the relativistic UED experiments. It is also a significant benefit in the development of next time-resolved TEM and compact relativistic TEM. However, many developments and improvements are needed to challenge. One is required to reduce further the transverse emittance and 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. However, once again, space charge will limit the minimum beam size to ~ 50μm. The damage problem on cathode should be considered for the use of such small UV laser. New cathode materials with small excess kinetic energy E_k are needed to develop to obtain the low emittance with the use of a large laser spot size. The spatial resolution is determined by the energy spread. Especially for the electron microscopy, $\Delta E/E \sim 10^{-5}$ has been used to obtain a sub-nanometer resolution. 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 10 fs laser. However, a drastically different approach is required to solve this problem. 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.

ACKNOWLEDGEMENT

This work was supported by a specially promoted research (Grant No. 22246127) of Grant-in-Aid for Scientific Research from the MEXT, Japan. The authors acknowledge Professor H. Kobayashi and Professor A. Ogata for valuable suggestions and discussions.

REFERENCES

[1] B.J. Siwick, et al., Sicence 302,1382 (2003).
[2] W. E. King, et al., J. Appl. Phys. 97, 111101(2005).
[3] B.J. Siwick, et al., J. Appl. Phys. 92,1643 (2003).
[4] T. LaGrange, et al., Ultramicroscopy 108, 1441-1449 (2008).
[5] B. Bariwick, et al., Science 322, 1227-1231 (2008).
[6] P. Musumeci, et al., Ultramicroscopy 108, 1450(2008).
[7] R. Li, et al.,, Rev. Sci. Intrum. 80, 083303 (2009).
[8] Y. Murooka, et al., Appl. Phys. Lett. 98, 251903 (2011).
[9] J. Yang, et al., Nucl. Instrum. Methods A 637, S24 (2011).
[10] X. J. Wang, et al., PAC'03, 420-422 (2003).
[11] http://regae.desy.de/
[12] J. Yang, et al., Proceedings of 8th Particle Accelerator Meeting in Japan, 2011.