FEMTOSECOND PHOTOINJECTOR AND RELATIVISTIC ELECTRON MICROSCOPY

J. Yang[#1], K. Kan[1], N. Naruse[1], Y. Murooka[1], Y. Yoshida[1], K. Tanimura[1], J. Urakawa[2]
[1]The Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan
[2]The High Energy Accelerator Research Organization, 1-1 Oho, Tsukuba, Ibaraki, Japan

Abstract

A new structure femtosecond rf gun has been developed under the KEK/Osaka University collaboration. An rf gun based relativistic electron microscopy has being constructed in Osaka University for the study of ultrafast dynamics of intricate molecular and atomic processes in materials. In this paper, the design of the new gun and the beam dynamics of femtosecond electron bunch in the rf gun were reported to achieve a low-emittance and low-energy-spread. The some demonstrations of the rf gun based MeV electron diffraction/ imaging measurements were discussed.

INTRODUCTION

Many intricate atomic processes or ultrafast chemical/ physical reactions in materials initiated with atomic motions on spatial-temporal scale of single atomic vibrational period, i.e. on nanometer or sub-nanometer spatial scale and in femtosecond time region. The revealing and understanding of these phenomena are essential not only in material science, physics, chemistry and biology, but also for new material development, new device fabrication and other applications. Most of researches into ultrafast dynamic processes are investigated by a visible-ultraviolet-infrared optical spectroscopy based on a femtosecond laser. The non-imaging optical spectroscopy exclusively measures frequency or time-domain quantities, and is only indirectly connected to atomic positions in condensed matter through model-dependent response function [1-2].

The X-ray or electron diffraction provides a direct measurement of the radial distribution function for atomic spacings in materials. When coupled with time resolution, diffraction yields a "real-time" probe of structural dynamics. Recently, two techniques of ultrafast electron diffraction (UED) with femtosecond time resolution and ultrafast X-ray diffraction with picosecond time resolution have been developed. The many ultrafast phenomena initiated with ultrashort light pulses, i.e. phase transformations, melting, resolidification and ultrafast breaking of chemical bonds in sub-picosecond or femtosecond time region, have been observed in UED [3-5]. In the ultrafast X-ray diffraction, a picosecond or femtosecond X-ray produced from synchrotron radiation or X-ray FEL (XFEL) are used or proposed. Many ultrafast dynamics or processes in proteins have been revealed. The time resolution in both the techniques can be expected to be ~100 femtosecond using a relativistic femtosecond electron pulse generated from the rf gun [6-9] or a femtosecond X-ray pulse produced from XFEL. However, there are not spatial resolutions.

Electron microscopy 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), would be the strongest tool for the study of ultrafast dynamics in material science, physics, chemistry and biology. Currently, the UEM with the nanosecond time resolution has been achieved in conventional TEM through the use of photo-activated electron source driven by a nanosecond laser [10]. 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 [11]. 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 nanometer/sub-nanometer-spatial resolution in UEM, because of the long electron bunch length and low bunch charge due to the space-charge effect.

A MeV photocathode rf electron gun may be a significant benefits in UED, because it can generate a femtosecond relativistic electron pulse reducing the space-charge limitation. The rf gun based UED was considered firstly at SLAC in 2006 [6]. Recently, the single-shot MeV electron diffraction measurement and the time-resolved measurement have been also succeeded by other three research groups [7-9]. 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 fabrication of a new near-relativistic femtosecond electron rf gun for UEM to study the atomic dynamics of phase transitions in solids. The considerations and design of a MeV UEM system using the rf gun is presented. The results of beam diagnostics of femtosecond electrons in the rf gun and the demonstrations of MeV electron diffraction and imaging measurements are reported.

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

NEW FEMTOSECOND RF GUN

Figure 1 gives the new femtosecond rf gun developed under the KEK/Osaka University collaboration. 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 shap and structure of the rf cavity and so on. To reduce these effects, the new rf gun has been developed with many improvements:

(1) A new structure of the rf cavity was used in the rf gun. 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 to reduce field asymmetries. The field asymmetries not only lead to an asymmetrical emittance resulting in the emittance growth, but also cause a distortion of the UEM 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 reduced.

(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. 1. 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 <0.1 pC/pulse using a copper photocathode.

(5) The photocathode in the new gun is removable. A transmission photocathode was proposed in UEM.

Table 1: The Expected Beam Parameters

Beam energy	1~3 MeV
Bunch length	100 or 200 fs
Emittance	0.1 mm-mrad or less
Energy spread	10^{-4}
Bunch charge	1.6 pC (10^7 e^-'s)

The rf gun is driven by a femtosecond Ti:Sapphire laser. In the laser system, the Ti:Sapphire oscillator (Tsunami, produced by Spectra-Physics Co.) is mode-locked with a frequency of 79.33 MHz, the 36th sub-harmonic of the 2856 MHz accelerating rf. The outputs of the oscillator laser pulses are captured by a Pockels cell and amplified up to 1mJ in a regenerative amplifier. The repetition rate of the amplifier is 1 kHz to amplify the laser pulse with a high-stability in the pulse energy. However, a part (10Hz) of the amplified pulses is used to produce the electron beam. The amplified pulse is converted to the ultraviolet (UV) light using two nonlinear crystals with the maximum energy of 40 µJ. The femtosecond UV light is injected on the cathode surface at an incident angle of

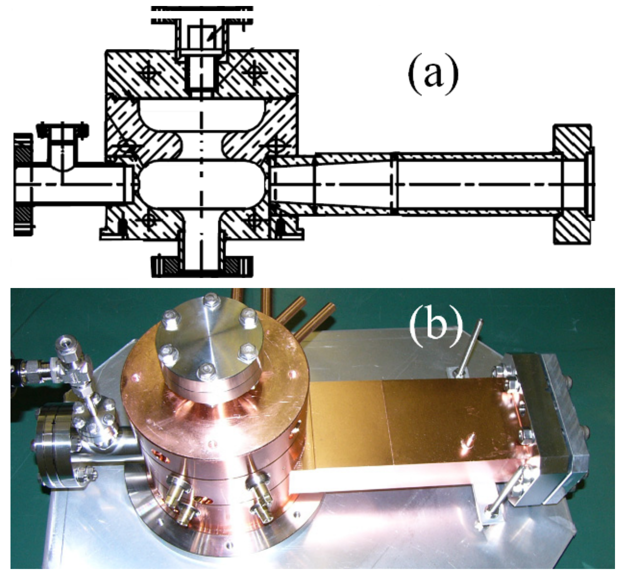

Figure 1: New femtosecond photocathode rf gun for UEM.

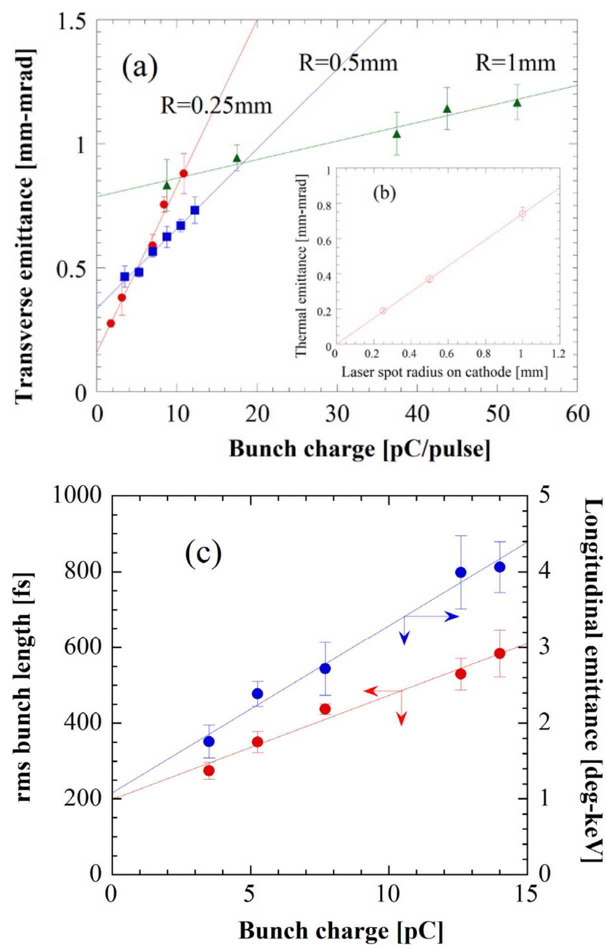

Figure 2: (a) transverse emittance as a function of bunch charge, (b) thermal emittance as a function of laser spot radius on the copper cathode, (c) longitudinal emittance and bunch length as a function of bunch charge.

approximately 2^{o} along the direction of the electron beam using a prism placed downstream of the gun. The expected beam parameters are listed in Table 1.

In the beam dynamics study, the pulse width of the UV light used in rf gun was 200 fs in full-width at half-maximum (FWHM). The laser spot radius on the copper cathode was R=0.25 to 1 mm. Figure 2(a) gives the normalized transverse emittance as a function of the bunch charge. The gun phase was 30^{o}. It was found that the increase of the transverse emittance due to the space-charge effect is 0.067 mm-mrad/pC at R=0.25mm. The thermal emittance, which is the emittance at zero charge in Fig. 2(a), increases linearly with the laser spot radius with a rate of 0.73 mm-mrad/1mm (Fig. 2(b)). In the case of the bunch charge of <2 pC, the emittance is dominated by the thermal emittance. The data indicates that a low thermal emittance of 0.1 mm-mrad can be obtained with the laser spot radius of 100 µm or less. Figure 2(c) gives the longitudinal emittance and the bunch length as a function of the bunch charge. At the bunch charge of <15 pC, the rms bunch length and the longitudinal emittance increase linearly with the bunch charge, i.e. 27.4fs/pC and 0.22 deg-keV/pC. Both the bunch length of 200fs and the longitudinal emittance of 1.1deg-keV at zero charge are due to the injection laser pulse width. The energy spread can be calculated to 2 x 10^{-3}. Anyway, it is possible to reduce the energy spread to 10^{-4} through the injection electron optics of TEM with a condense beam aperture. The use of a short-pulse UV laser (i.e. <100 fs) is also essential to achieve such low energy spread of 10^{-4}.

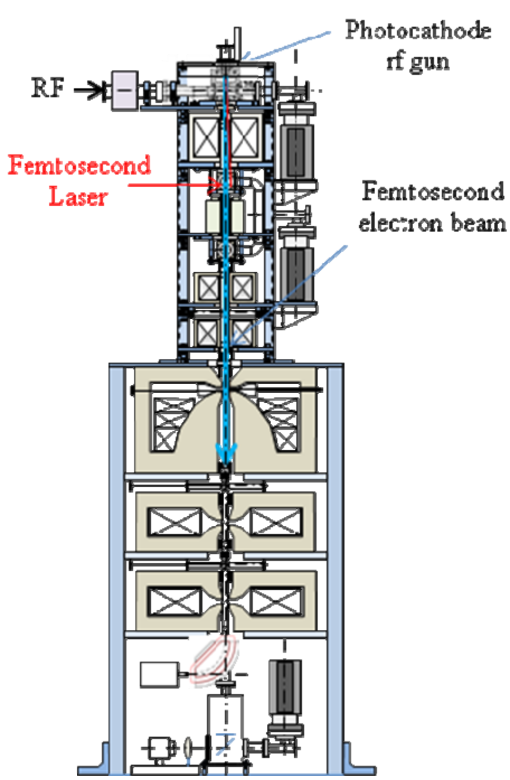

Figure 3: The rf gun based ultrafast transmission electron microscopy

RF GUN BASED MEV UEM SYSTEM

The prototype system of MeV UEM using the rf gun in Osaka University is given in Fig. 3. After the rf gun, two condense lenses is used to transported and controlled the electron beam on the specimen. An objective lens, an intermediate lens and a projector lens are used to form and project an imaging. Finally, the MeV imaging is measured by a scintillater and an efficient 1000 x 1000 pixel charge-couple device (CCD) camera with 25 µm pixels. To achieve high sensitivity to MeV electrons and a high damage threshold, a scintillater of CsI(Tl) equipped with fiber optic plates (Hamamatsu photonics FOP11) was used. For the TEM operation, the following requirements of the electron beam should be satisfied:

(1) According to the Rose criterion (~100 electrons/ pixel), the number of electrons in the bunch can be considered to N~10^{8} for the single-shot imaging and N~10^{6} for the single-shot diffraction.
(2) The normalized emittance of the electron beam generated from the rf gun is required to less than 0.14 mm-mrad for obtaining a focal spot size of 10 µm on the specimen.
(3) The energy spread should be of order of 10^{-4} to reduce the effects of spherical and chromatic aberrations in the electron optics.
(4) The final requirement is low dark current.

As the experiences, there is no problem to generate the required electron number with low dark current (<0.1pC) using the new rf gun. To obtain the low-emittance beam, we can use a small laser spot size on the cathode (R=0.1 mm or less). For the low energy spread, in addition to use a short-pulse laser, the rf compression in longitudinal phase space in the rf gun operated at low gun phase is also useful, i.e. 10^{-4} at <30^{o} with 100fs laser injection. However, at the low injection phase, the effective electric field at the cathode decreases, resulting in the increase of space-charge effect on the transverse emittance.

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