FEMTOSECOND ELECTRON GUNS FOR ULTRAFAST ELECTRON DIFFRACTION*

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Abstract

The photocathode radio-frequency (RF) gun is a powerful electron source to generate a high-brightness femtosecond electron beam with energies of 1~3 MeV. The RF gun based relativistic ultrafast electron diffraction (UED) 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, we analyze the requirements and limitations of the beam parameters used in this technique. The characterization of the performance of femtosecond RF guns, and the experimental instruments of the RF gun based relativistic UED are reviewed. Finally, some demonstrations of relativistic UED measurements are reported.

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, it is difficult to obtain a 100fs electron bunch and to improve the time resolution to 100 fs or less. The small number makes it also difficult to observe the ultrafast dynamics with the single-shot measurement. The studies of nonrelativistic UED are limited to the reversible processes.

In order to achieve sub-100 fs temporal resolution, a unique approach is constituted by the use of relativistic electrons to suppress the increase of the electron bunch length due to the space-charge forces. For a relativistic beam, a larger number of electrons, i.e. 10^7 to 10^8, can be packed in a single 100 fs long bunch, which provides us observing ultrafast dynamics with single shot diffraction measurement for the study of irreversible ultrafast processes. The relativistic energy electrons have also been a fundamental breakthrough on the limitation of elastic mean free path in materials, and provide transmission electron diffraction measurements to straightforward structural information, which greatly increase the applicability for many interesting phenomena.

Photocathode radio-frequency (RF) gun is a powerful candidate as a possible relativistic electron source for UED. RF guns have definitely been a fundamental technological breakthrough in the last 20 years as relativistic electron sources and are in fact the preferred electron source for advanced accelerator and free electron laser. High accelerating gradients in RF guns, ≥100 MV/m, accelerate the photoemission electrons to relativistic energies in few cms of propagation distance, as shown in Fig.1. The space-charge forces are greatly suppressed. An ultrashort electron beam with low emittance and low energy spread can be expected.

First experimental demonstration of RF gun based UED was carried out in 2006 at SLAC [4]. From 2006, several groups at UCLA [5], Tsinghua University [6], Osaka University [7,8], BNL/Shanghai Juaotong University [9] and ZESY [10] have developed new MeV UED systems using the photocathode RF guns. Ultrashort electron bunches at the energy of 1~3 MeV, the emittance of < 1 mm-mrad, and the charge of few pC have been used. Recently, both the single-shot MeV electron diffraction measurement and the time-resolved measurement have been successful. The experimental results suggest that the RF gun is a useful source for relativistic UED.

Here, we analyze the requirements and limitations of the beam parameters used in UED. The characterization of the performance of RF guns, and RF gun based relativistic UED system are reviewed.

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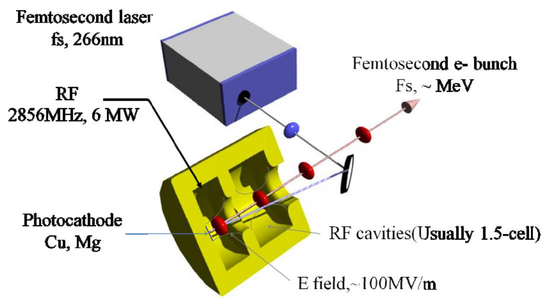

Figure 1: Concept of femtosecond photocathode RF gun.

REQUIREMENTS AND LIMITATIONS OF RF GUN FOR UED

The beam parameters required for UED can be determined as the characteristics: charge, energy, emittance (spatial size and divergence) and bunch length of the ultrashort electron beam.

Electron Charge

For the single-shot measurement with electron microscopy, the Rose criterion indicates that an average detector signal strength of ~100 electrons/pixel is required for adequate gray scaling of the image. For a 1k x 1k CCD camera, this implies an incident electron pulse with N~10^8 for imaging in microscopy.

In UED, the beam charge is determined by the requirement of having enough scattered electrons to be able to detect the Bragg peaks. The previous researches indicate that, typically, about 10^6 electrons/pulse as significantly less signal is required to determine the position of discrete single-crystal Bragg diffraction spots or diffraction rings for polycrystalline specimens. In the relativistic case, the detectors have not been fully developed yet to yield single-electron detection, and some improvements should be expected from further research. Nevertheless, that limit is not very far and as an example at Osaka University acceptable diffraction images with 5 x 10^5 ~ 1 x 10^6 electrons have been recorded using a scintillater of CsI(Tl) equipped with fiber optic plates and an electron multiplying CCD camera, as described below.

Beam Energy

The electron energies from 2 to 3 MeV is more suitable for the relativistic UED when considering both the temporal resolution and the spatial quality of the patterns. The De Broglie wavelength associated with the beam ($\lambda=h/p$, where h is the Planck constant and p is the beam momentum) is 0.6-0.4 pm for 2-3 MeV beam, which is more than 10 times shorter than the one for non-relativistic electrons.

The beam energy generated in the RF gun can be varied easily by adjusting the acceleration field gradient. However, both the bunch length and the energy spread increase largely with decreasing the field gradient, especially at beam energy of <2 MeV. The space charge force is dominant in this energy region, as given in Fig. 2. To reduce the growths, one could use a high-field gradient in the RF gun. The theoretical studies indicate that a 100-fs electron beam with the energy of 2 MeV or more, and the relative energy spread of 10^{-4} at bunch charge of 0.1-1pC (10^6-10^7 electrons/pulse) is achievable. For the higher energy, i.e. >3 MeV, other problems of decreasing the quantum efficiency into the detector and magnetic field saturation of electron optics are occurred.

Beam Emittance

A key parameter determining the performance of UED is the normalized beam emittance, which includes both the divergence and spatial size of the electron source. To obtain a good quality diffraction image with enough spatial resolution in UED, the resolving power of the diffraction camera should be $P=R/\Delta R > 10$, where R is the radius of the diffraction rings on the detector screen and ΔR is the smallest distance between two neighbouring rings that can be barely discriminated. The limiting distance between two rings that can just be separated is $\Delta R \sim 2\sigma_x$, and hence $P=R/2\sigma_x = \lambda/(2d\sigma_\theta)$, where σ_x is the radius of the undiffracted beam size at the detector, σ_θ is beam divergence angle and d is an inter-atomic distance, typically $d \sim 2$ Å. To keep $P > 10$ with a wavelength $\lambda =$ 0.4 pm for 3 MeV electrons and $d \sim 2$ Å, it is necessary to have $\sigma_\theta < 0.1$ mrad at the target.

For the time-resolved study in UED, a typical pump laser focusing spot size at sample is ~1 mm^2 or less with intensity on the order of tens of mJ/cm^2. By considering a same beam size of the probe electrons at the sample, the desired normalized transverse emittance is about 0.1 mm-mrad. Assuming then N = 1 x 10^6 electrons in an rms bunch length of σ_z = 100 fs, and an rms normalized emittance of ε_n = 0.1 mm-mrad at 3 MeV energy, the source has to be able to generate a beam with normalized

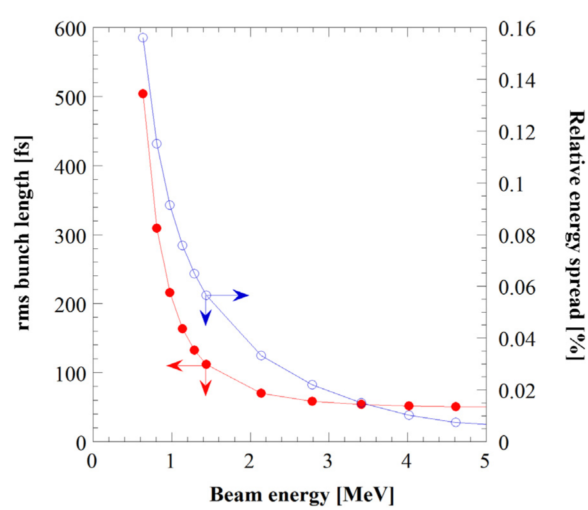

Figure 2: Simulation results of bunch length and energy spread as function of beam energy in the RF gun at the bunch charge of 0.1 pC.

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brightness of

$$B_n = \frac{2N}{\sigma_z \varepsilon_n^2} = 3 \times 10^{14}\ \mathrm{A/m^2}.$$

Although the desired value is critical, a small divergence angle of $\sigma_\theta \sim 50$ µrad has been achieved in the RF gun based UED experiment, as will be shown below.

For the RF gun operated by ultrashort bunch at low charge, i.e. 1 pC or less, the RF and space charge effects are not as significant. The beam emittance is, therefore, dominated by "thermal" emittance at the photocathode. The thermal emittance can be described by an angular spread due to the excess kinetic energy E_k arising from the difference between the energy of the driver laser photon energy and the cathode work function,

$$\varepsilon_{th} \cong \sigma_r \sqrt{\frac{E_{kin}}{m_0 c^2}}\ .$$

The thermal emittance is also proportional to the rms laser spot size σ_r. Figure 3(a) gives the normalized transverse emittance as functions of the bunch charge and the rms laser spot radius on a copper cathode, which measured in a 1.6-cell S-band (2856 MHz) RF gun. The gun phase was 30°. The pulse width of the UV light (266 nm) illuminated on the cathode was 200 fs in rms. The thermal emittance, which is the emittance at zero charge in Fig. 3(a), is plotted in Fig. 3(b). The data shows that the thermal emittance increases linearly with the laser spot radius with a rate of 0.73 mm-mrad/mm. From the data, we can obtain the spread in divergence angle $(E_k/m_0c^2)^{1/2} \sim 0.74$ mrad, and the kinetic energy $E_k = 0.26$ eV on the copper cathode. The desired normalized emittance of 0.1 mm-mrad can be obtained with the laser spot radius of 130 µm or less using the copper cathode.

However, a small increase of the transverse emittance due to the space charge effect at the charge of < 2 pC is occurred during the propagation. The increase rate depends on the laser spot size in the cathode, i.e. 0.067 mm-mrad/pC at $\sigma_r = 0.25$mm.

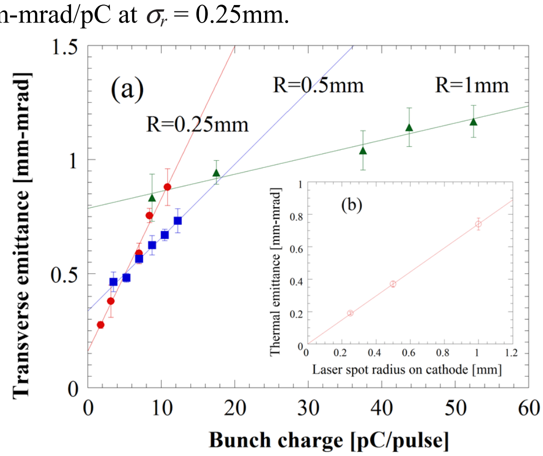

Figure 3: (a) transverse emittance as a function of bunch charge, (b) thermal emittance as a function of laser spot radius on the copper cathode.

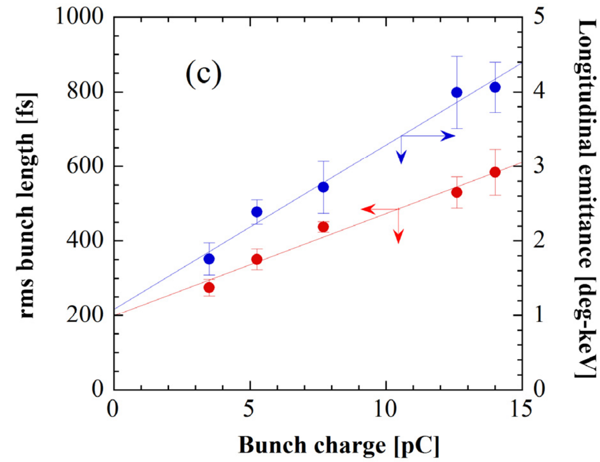

Figure 4: Bunch length and longitudinal emittance measured as a function of bunch charge.

Bunch length

To improve the temporal resolution, the length of the beam could be reduced, for example, by reducing the laser pulse length at the cathode, but the bunch broadening occurs during the propagation due to the space charge force in the bunch. In Fig. 4, we report the measurement results of the bunch length and longitudinal emittance generated from the RF gun where we changed the bunch charge from 3 to 14 pC. The incident UV laser pulse width was 200 fs. At < 14 pC, the bunch length and the longitudinal emittance increase linearly with the bunch charge by rates of 27.4fs/pC and 0.22 deg-keV/pC, respectively. Both the bunch length of 200 fs and the longitudinal emittance of 1.1deg-keV at zero charge are due to the incident laser pulse width. The energy spread can be calculated to ~2x10^{-3}, which is not enough in UED measurement with a good spatial resolution. Anyway, it is possible to reduce the energy spread to 10^{-4} by using a short laser pulse at the cathode. A laser pulse of ≤ 100 fs has been used in most of RF gun based UED systems.

The rms energy spread of the beam generated from the RF gun can be estimated by Kim's theoretical model, for a 1.5-cell RF gun, $\Delta E/E = 0.13 f \sigma_z$, where f in GHz is the accelerating radio-frequency (f = 2.856GHz for S-band) and σ_z in ps is the electron bunch length ($\sigma_z \sim \sigma_r$ at low charge), i.e. $\Delta E/E \sim 10^{-4}$ using a 100 fs UV laser on the cathode. In future development, the use of a short-pulse UV laser (i.e. ~ tens fs) would be essential to achieve a low energy spread of 10^{-5} for the challenge of RF gun-based MeV electron microscopy.

NEW FEMTOSECOND RF GUN

The electron source required for electron diffraction and microscopy 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 femtosecond RF gun under the

KEK/Osaka University collaboration, as shown in Fig. 5. 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. 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 UED 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

RF GUN BASED UED AT OSAKA UNIVERSITY

Figure 6 shows a schematic of RF gun based UED system at Osaka University. A 1.6-cell S-band rf gun is used with a copper cathode, which illuminated by the third harmonic of a Ti:Sapphire laser, with a 90 fs pulse full width at half maxima (FWHM). The electron energy is 3.0 MeV. The operation repetition rate is 10 Hz. Laser pulses were time-synchronized with rf by adjusting the oscillator cavity length to phase-lock the laser output with the 79.3 MHz rf generated as the 36th subharmonic of the 2856 MHz accelerating rf. The time jitter of 144 fs FWHM between laser output and 79.3 MHz rf has been demonstrated by this technique. After the RF gun, a solenoid and a condenser lens are used to make a parallel electron beam with the minimum divergence on the sample. The sample is located at a distance of ~ 1 m from the RF gun. The diffraction patterns in the sample are magnified with two electron lenses downstream of the sample: a diffraction lens (DL) to provide a back-focal plane for expanded diffraction images, and a projection lens (PL) to display the diffraction patterns with desired fashion on the detector. To achieve high sensitivity to MeV electrons and a high damage threshold, 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 optical image 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. By using the electron lenses, the UED system is hence very compact, i.e. just 2.8 m long from photocathode to detector. The maximum number of electrons in the UED measurement was 4.5 x 10^7 electrons/bunch (~3 pC). The diffraction images with 5 x 10^5 ~ 1 x 10^6 electrons are able to be recorded using the detector.

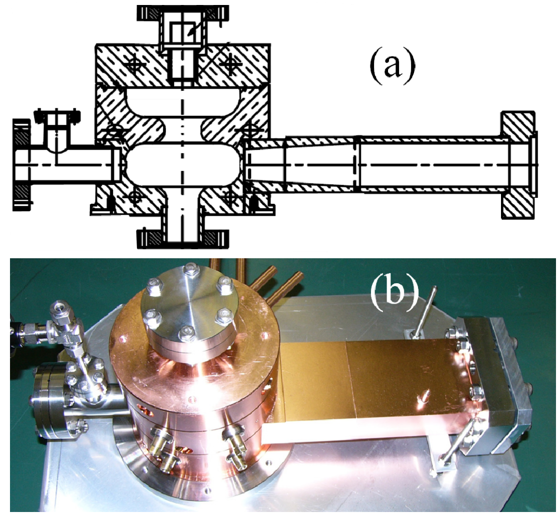

Figure 5: New femtosecond photocathode rf gun for ultrafast electron diffraction and microscopy.

Here, we give the measurement results of 180-nm-thick single crystalline Si using the UED facility. Figure 6 gives the experimental results: a 20-shot averaged image, a single-shot image, the intensity profiles of diffraction patterns, and the ultrafast dynamics observed by time-resolved measurement. Not only the lowest Bragg peaks but also higher-order peaks are clearly visible. The excellent quality of the single-shot image for 180-nm-thick samples is comparable to the pioneering data in low-energy TED for 30-nm-thick samples. From the intensity profile, the maximum scattering vector is obtained to be more than 1.56 Å^{-1}. The FWHM of the zeroth-order spot is 0.02 A^{-1}, showing an excellent spatial resolution for MeV diffracted beams. The width of the zeroth-order spot

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corresponds to the convergence angle. We obtained a small angle of σ_θ= 50 μrad in MeV UED, which is 10 times smaller than that in nonrelativistic UED.

The dynamical phenomena in Si, i.e. phase transition, have been successfully observed by recording the change in the intensity of electron diffraction spots (i.e. 620 spot) when changing the time delay of the pump laser pulse and the probing electron bunch with time step of 80 fs. The ultrafast dynamics with a time constant of 800 fs may be caused by the phonon interactions in the lattices. We have also observed successfully other ultrafast dynamics in metals and insulators using the UED facility, i.e. the laser heating and melting in gold and aluminium. The experimental results strongly that the RF gun is very useful for relativistic UED.

CONCLUDING REMARKS

The RF gun is a high-brightness femtosecond electron source for UED. It can generate a sub-100 fs electron bunch with normalized emittance of ≤0.1 mm-mrad, energy spread of $\leq 10^{-3}$ and energy of 1~3 MeV. A large number of 10^6 ~ 10^7 in the relativistic-energy bunch provides us to make a single-shot measurement for the study of irreversible ultrafast processes. The UED with MeV electron pulses can be a powerful tool to study structural dynamics in wider range of materials with 100 fs or sub-100 fs temporal resolution.

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, $\Delta E/E$ ~ 10^{-6} has been used to obtain a sub-nanometer resolution. For the RF gun, the energy spread is limited to $\Delta E/E$ ~ 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.

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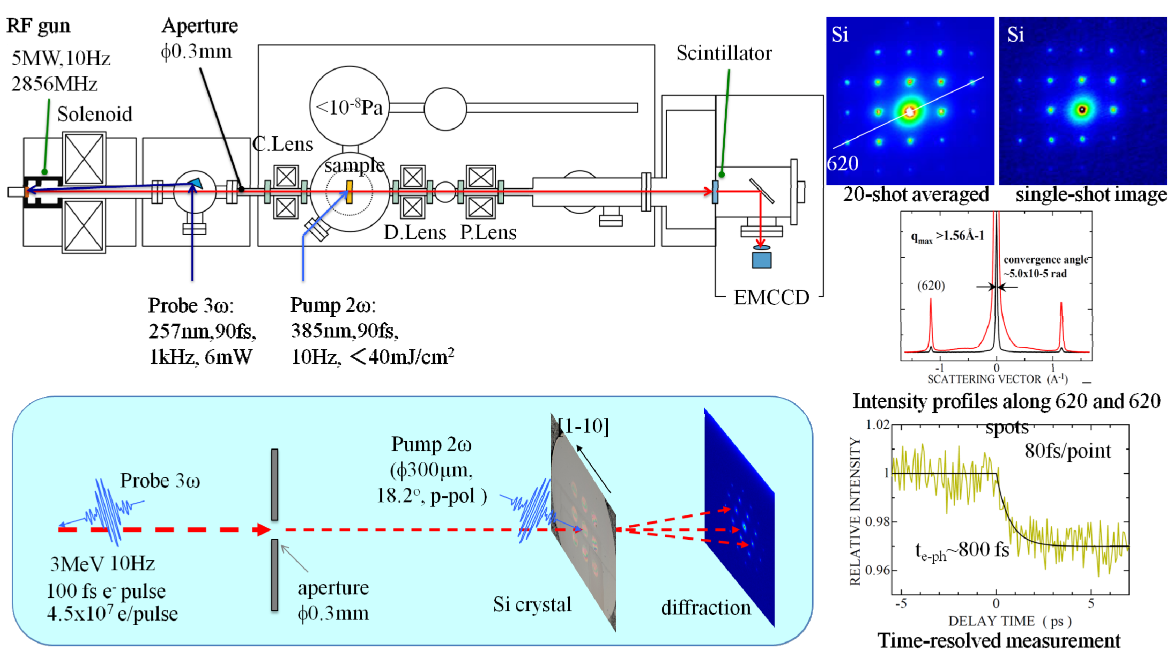

Figure 6: RF gun based UED facility at Osaka University and the experimental results in single-crystal Si: a 20-shot averaged image, a single-shot image, intensity profiles, and ultrafast dynamics observed by time-resolved measurement.