PHOTOCATHODE FEMTOSECOND ELECTRON LINAC AND ITS APPLICATIONS

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

Abstract

Photocathode rf electron linac facilities have been developed in Osaka University to reveal the hidden dynamics of intricate molecular and atomic processes in materials. One of the linacs was developed using a booster linear accelerator and a magnetic bunch compressor. This linac was successfully produced a 100-fs high-brightness electron single bunch and initiated the first experimental study of radiation chemistry in the femtosecond time region. Another was constructed with a photocathode rf gun to generate a near-relativistic 100-fs electron beam with a beam energy of 1~4 MeV. A time-resolved MeV electron diffraction was successfully developed with this gun to study the ultrafast dynamics of structure change in materials.

INTRODUCTION

Ultrashort electron bunches, of the order of a few 100 fs with energy of a few or tens MeV, are essential to reveal the hidden dynamics of intricate molecular and atomic processes in materials through experimentations such as femtosecond electron diffraction and femtosecond pulse radiolysis. Pulse radiolysis, which is an important method for studying the primary processes of radiation chemistry, has been constructed in a large number of facilities around the world. In pulse radiolysis, a short-pulse electron beam is used as the irradiation source, while short-pulse light is used as the analyzing source. Over the past 10 years, several ultrafast pulse radiolysis facilities involving the use of a laser-triggered electron accelerator with a photocathode rf gun have been constructed at Brookhaven National Laboratory [1], the University of Tokyo [2], the University of Paris-Sud [3], Sumitomo Heavy Industries [4] and Osaka University [5]. A photocathode rf gun can generate a single picosecond electron bunch with a charge of several nano-coulombs by picosecond laser excitation of the photocathode without the need of sub-harmonic pre-bunching system. The electron bunch produced by the rf gun is synchronized with the excited laser. It is possible to synchronize the electron beam with the analyzing laser light in the picosecond or subpicosecond time region. Moreover, the electrons are emitted from the photocathode surface in the presence of a strong rf electric field (~100 MV/m). A low-emittance electron beam can be generated by the reduction of the space-charge effect in a short-bunch electron beam. Typical parameters of a 1.6-cell S-band (2856 MHz) rf gun developed at Brookhaven National Laboratory [6,7] are follows: transverse normalized emittance, 1.2 mm-mrad; bunch charge, 1 nC [8]. A low-emittance electron beam helps achieve high brightness with a small beam spot size on the sample, resulting in a good signal-to-noise ratio at a low charge. However, the time resolution is still limited to the picosecond time scale, because of the electron pulse duration, time jitter, and the effect of group velocity mismatch (GVM) of the electron beam and the light in the sample. Our group has developed a "double-decker electron beam accelerator" to reduce the time jitter between the electron beam and the analyzing light [9], and has also developed "equivalent velocity spectroscopy (EVS)" for pulse radiolysis to overcome the effect of GVM on the time resolution [10]. Here, we present femtosecond pulse radiolysis with the aim of initiating an experimental study of the primary processes of radiation chemistry in the femtosecond time region. The dependence of the radiolysis signals on the electron charge and the optical path length is investigated [11, 12].

In UED, a short electron bunch is used as a probe source. The ultrafast phenomena initiated with ultrashort light pulses are observed by monitoring the electron diffraction patterns in the pump and unpump stats. Most of the conventional UED experiments[13,14] have been constructed by using dc or pulsed photocathode electron guns with beam energy of 30-60 keV. Unfortunately, the space charge effect (Coulomb repulsion) within the pulse for low energy, and the initial kinetic energy distribution of the photoelectrons act to broaden the electron pulse as it propagates. Those made it difficult to obtain pulse much shorter than 1 ps containing large numbers of electrons (10^4 or more). To generate a femtosecond electron beam for UED, one could use low electron charge, i.e. ~1000 electrons per pulse or less. Recently, the shortest width of the electron pulses used in UED is 600 fs containing 6000 electrons at 30 keV. Alternatively, it is possible to increase the extracting electric field inside the electron gun, while the space charge effect is reduced. However, this approach is limited by the maximum electric field for vacuum breakdown (12 and 25 MV/m for the dc and pulsed, respectively).As these reasons, in 2006, a MeV photocathode radio-frequency (rf) electron gun was firstly considered for UED [15]. The rf gun can generate a low emittance femtosecond-bunch electron

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

beam with megavolt beam energy by injecting a femtosecond laser light on the cathode is achievable by using the rf gun. It provides a big choice to construct megavolt electron diffraction. Here, we approach femtosecond time-resolved electron diffraction using the rf electron gun, with the aim of obtaining single-shot diffraction patterns on a 100 fs time scale, to study the atomic dynamics of phase transitions in solids. Here, we report the developments of a near-relativistic 100-femtosecond rf electron gun and femtosecond megavolt electron diffraction system, and the studies of beam dynamics of femtosecond electrons in the rf gun. The growths of the emittance, bunch length and energy spread due to the rf and space charge effects are discussed.

FEMTOSECOND TIME-RESOLVED PULSE RADIOLYSIS

Figure 1 shows the presented femtosecond pulse radiolysis using a femtosecond electron linear accelerator and a time-synchronized femtosecond laser. The femtosecond electron accelerator was constructed with a 1.6-cell BNL-type photocathode rf gun, a 2 m-long boost linac and a magnetic femtosecond bunch compressor. The rf gun generated a picosecond electron bunch on a copper cathode driven by an Nd:YLF picosecond laser. The laser pulse was time-synchronized with the electron beam by phase-locking the laser output with the 79.3 MHz rf produced as the 36th sub-harmonic of the 2856MHz accelerating rf. The linac accelerated the electron bunch up to 32 MeV with an energy-phase correlation in the bunch for the compression. Finally, the electron bunch was compressed into femtosecond by rotating the bunch in the longitudinal phase space distribution in the magnetic bunch compressor. The compressor was constructed with two 45°-bending magnets and four quadrupole magnets (two pairs), which provides the necessary path length dependence on energy. In order to obtain a short bunch length, all magnets were carefully

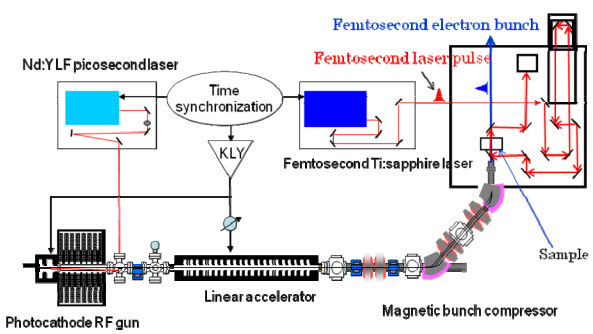

Figure 1: Femtosecond pulse radiolysis using a femtosecond electron beam and a time-synchronized femtosecond laser.

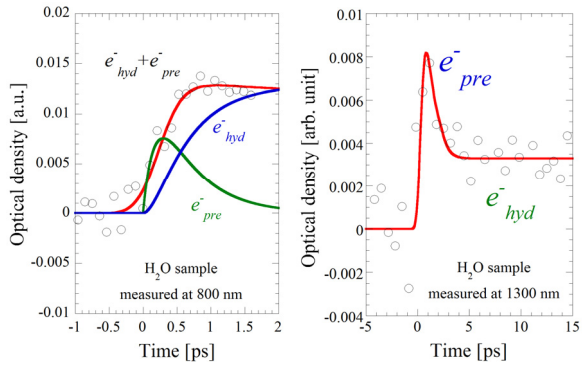

Figure 2: The transient absorption kinetics of solvated and pre-solvated electrons in water sample. The data was observed in pulse radiolysis at the wavelength of the probe laser was 800 and 1300 nm.

installed with the minimum lattice error to reduce the aberrations in the phase space distribution. However, during the bunch compression, the second-order dispersion causes a nonlinear deformation of the longitudinal phase space, which increases the final bunch length. To reduce the nonlinear effects, a nonlinear energy correlation along the bunch length, which introduced by re-phasing the linac away from the zero-crossing of the rf (i.e. away from the linear slope), was used. The shortest bunch was obtained to be 98 fs in root main square (rms) at bunch charge of 0.2 nC by re-phasing the accelerating rf to 94° off crest of the rf waveform [11]. In the presented pulse radiolysis experiment, the rms bunch length was 201 fs at 0.4 nC. The repetition rate of the operation of the accelerator was 10 Hz in the experiment [10]. A Ti:sapphire femtosecond laser was used as a probe light source. The laser pulse was time-synchronized with the electron beam as well as the picosecond laser as described as above. The output energy of the laser pulse was 10 nJ. The wavelength of the laser light was 800 nm with bandwidth of 12.5 nm in full width in half of maximum (FWHM). A regenerative amplifier amplifies the pulse energy up to about 0.8 mJ with a repetition rate of 1 kHz. Finally, the amplified laser pulse was guided to OPA to change the light wavelength. The wavelength range from 400 to 1600 nm was used in the pulse radiolysis experiment.

Figure 2 gives an example of the transient absorption measurement in water sample. The wavelength of the probe laser was 800 and 1300 nm. A build-up of hydrated electron in the femtosecond time region is appeared clearly in the measurement. The transient absorption kinetics suggests that the hydrated electron is formed via an intermediate state (“pre-hydrated” electron) after the thermalization process in the water radiolysis. Numerical analysis of the experimental data indicates that the hydrated electron is formed from a pre-hydrated electron state in water radiolysis. The formation time is 550 ± 50 fs that is consistent with the photoionization process.

FEMTOSECOND MEV ELECTRON DIFFRACTION

We have developed successfully a femtosecond MeV electron diffraction using the rf electron gun as shown in Fig. 3, with the aim of obtaining single-shot diffraction patterns on a 100 fs time scale, to study the atomic dynamics of phase transitions in solids. In the system, a new 1.6-cell S-band rf gun has been developed under the KEK/Osaka University collaboration with many improvements [12,16]: (1) the conventional laser injection ports in the half cell were removed to reduce field asymmetries; (2) a new turner system was designed to adjust precisely the electric field balance in the half and full cells; and (3) a new insertion function of the photocathode was installed to reduce the field emission. The rf gun is driven by a time-synchronized femtosecond Ti:Sapphire laser. The theoretical studies indicate that a 100-fs electron beam with the energy of 1.5-4 MeV, the transverse emittance of 0.02-0.1 mm-mrad and the relative energy spread of 10^{-3}-10^{-4} at bunch charge of 0.1-2 pC(10^{6}-10^{7} electrons per pulse) is achievable at the gun phase of 30^{o} using a 200-fs laser pulse.

The femtosecond electron beam produced from the rf gun for obtaining the diffraction patterns is propagated to the sample with a magnetic lens besides the solenoid magnet. This magnetic lens, which is located at a distance of 1 m from the cathode, is used to make a parallel electron beam with the minimum divergence on the sample. The sample is located at a distance of 25 cm from the magnetic lens. The diffraction patterns in the sample are magnified with two magnetic lenses downstream of the sample. The two magnetic lenses are installed downstream of the sample: the first lens (diffractive lens) is located at the position of 25 cm from the sample, while the second lens (projective lens) is located downstream of the diffractive lens. The magnified diffraction patterns are measured by an emulsion plate which is located at a distance of 1.4 m from the sample.

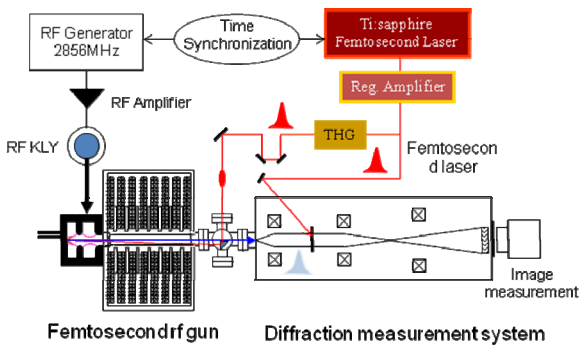

Figure 3: RF gun based femtosecond electron diffraction.

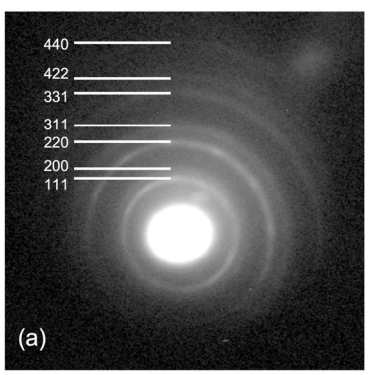

Figure 4: MeV diffraction pattern of polycrystalline Al foil.

Figure 4 shows the demonstration of MeV electron diffraction measurement in a 70-nm-thick polycrystalline Al foil. The beam energy of the electrons was 2.9 MeV. The electron charge was 2 pC. The normalized emittance was less than 0.8 mm-mrad. The dark current in the diffraction measurement was less than 0.06 pC. The laser injection phase was fixed to 30^{o}. The demonstration suggests that the photocathode rf gun is a powerful tool for femtosecond time-resolved electron diffraction.

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