PHOTOCATHODE FEMTOSECOND ELECTRON BEAM APPLICATIONS: FEMTOSECOND PULSE RADIOLYSIS AND FEMTOSECOND ELECTRON DIFFRACTION

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Abstract

Both ultrafast time-resolved radiolysis and electron diffraction based on photocathode rf electron guns have been developed in Osaka University to reveal the hidden dynamics of intricate molecular and atomic processes in materials. One of the photocathode rf guns has been used successfully to produce a 100-fs high-brightness electron single bunch with a booster linear accelerator and a magnetic bunch compressor. Pulse radiolysis with time resolution of 240 fs was developed successfully for the observation of femtosecond kinetics in materials. Another photocathode rf gun, which produces directly a near-relativistic 100-fs electron beam, has been developed to construct femtosecond electron diffraction. The megavolt electron diffraction patterns have been observed. The dependences of the emittance, bunch length and energy spread on the radio-frequency (rf) and space charge effects in the rf gun were investigated.

INTRODUCTION

It is well-known that the femtosecond-pulse laser light is an indispensable photon source for the study of ultrafast dynamics in physics, chemistry and material science. The many reactions or phenomena or dynamics in the femtosecond time region were discovered and understood. Ultrashort electron bunches, of the order of 100 fs, are also 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. In pulse radiolysis, a short electron bunch is used as a pump source. The electron-induced reactions are analyzed by an ultrashort probe light such as femtosecond lasers or x-rays using a stroboscope technique. A femtosecond single electron bunch with beam energy of a few tens MeV is very important to be utilized in this technique for observing information of the most basic reaction mechanisms in physics, chemistry and biology (e.g. excitation, ionization, and relaxation of atoms and molecules) on the femtosecond time scale. The ultrafast electron diffraction (UED) or ultrafast electron microscopy (UEM) provides a unique opportunity for a complete determination of the transient structures with atomic level detail. In UED or

UEM, a short electron bunch is used as a probe source. The photon-induced ultrafast structural dynamics or phenomena are observed by monitoring the electron diffraction patterns in the pump and unpump stats.

Here, we present two applications of femtosecond-bunch electron beams: One is a femtosecond pulse radiolysis. The observations of the ultrafast reactions in materials, i.e. solvation processes on the femtosecond time scale, are reported. Another is a time-resolved MeV electron diffraction, which is used a near-relativistic 100-fs electron rf gun, to study the atomic dynamics of phase transitions in solids. The femtosecond electron bunch generation is reported and discussed.

FEMTOSECOND PULSE RADIOLYSIS

Pulse radiolysis is a powerful tool for the study of ultrafast reactions or primary processes in materials. The development of picosecond pulse radiolysis was started in 1970s at the University of Toronto using a picosecond electron micro-bunch produced from an S-band linear accelerator. The Toronto instrument opened firstly the pulse radiolysis study on picosecond time scale, and many important reactions within spurs were observed. After the Toronto experiment, the several picosecond pulse radiolysis facilities were constructed with single-bunch electron accelerators using sub-harmonic bunching and pre-bunching techniques. In the late 1990s, new ultrafast pulse radiolysis facilities have been developed using laser-triggered photocathode rf electron guns, because the photocathode rf gun produces a single picosecond electron bunch with charge of several nano-coulombs using picosecond laser excitation on the photocathode without the need of the sub-harmonic bunching or pre-bunching system. Moreover, the electron beam produced from the rf gun is synchronized with the excited laser. However, the time resolution of pulse radiolysis is still stopped in the picosecond time region (~2 ps), because of the electron bunch length and the time jitter between the electron bunch and the analyzing laser pulse. Until now, the reactions or phenomena of radiation chemistry in the femtosecond time region are not revealed.

Here, we present a femtosecond pulse radiolysis using a femtosecond electron linear accelerator and a time-synchronized femtosecond laser as given in Fig. 1. The femtosecond electron accelerator was constructed with a

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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. In order to obtain a low-emittance electron beam for the femtosecond bunch compression, a single solenoid magnet was installed at the exit of the rf gun to compensate the transverse emittance growth during the beam transport due to space charge effect.

After the emittance compensation, 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 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 [1]. 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.

A Ti:sapphire femtosecond laser was used as a probe light source. The laser pulse was time-synchronized with
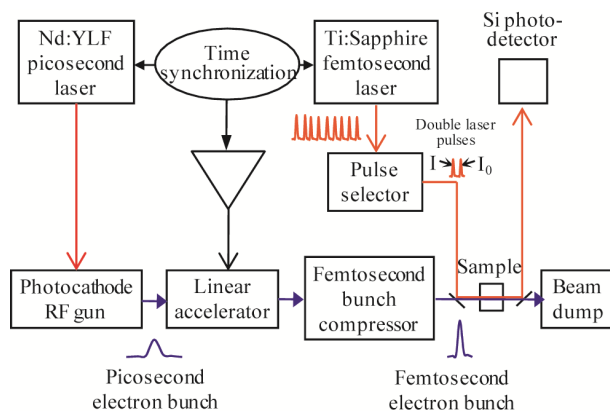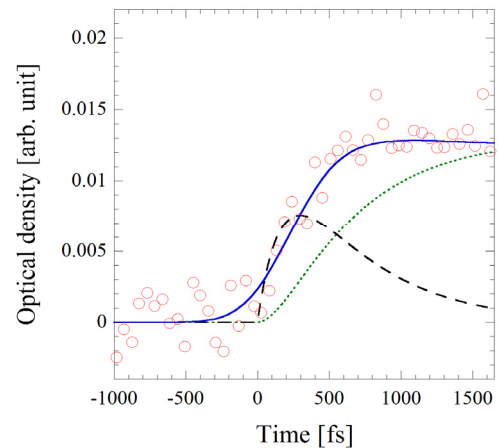

Figure 2: Femtosecond transient absorption kinetics of hydrated electron in water measured by a femtosecond electron bunch and a femtosecond laser light.

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 technique of double-pulse injection [2] was used in the pulse radiolysis to improve the signal to noise ratio due to the fluctuation of the laser intensity caused by long-term drift and mechanical vibration of optics at low frequencies: the continuous laser pulse train of 79.3 MHz, output of the laser, was guided to a pulse selector (Spectra Physics, 3980), which was constructed with an Acousto-optic Modulator (AOM) crystal driven by the rf; Two-pulse laser train , which had same pulse energy with a time interval of 12.6 ns, were then extracted and used respectively as a reference (I_0) pulse and a signal (I) pulse in the pulse radiolysis.

In the system, the laser pulse duration out of the pulse selector is 107 fs in rms. The stability of the laser intensity was 0.9 %. The time jitter between the femtosecond laser output and the reference 79.3 MHz rf signal was measured to be 61 fs in rms. Therefore, we can estimate the time resolution of the pulse radiolysis, due to the electron bunch length (σ_e=201 fs), the laser pulse duration (σ_l=107 fs) and the time jitter (σ_J= 61 fs) between the electron bunch and the laser pulse, to be $\Delta t_{beam} = \sqrt{\sigma_e^2 + \sigma_l^2 + \sigma_J^2}$ = 240 fs. By using the pulse radiolysis system, we measured the transient absorption kinetics of hydrated electron produced by electron-beam-ionization in the 0.18-mm-thick water sample, as shown in Fig. 2. The wavelength of the probe laser was 800 nm with the FWHM bandwidth of 12.5 nm. A build-up of hydrated electron in the femtosecond time region is appeared clearly in the measurement. 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.

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

FEMTOSECOND ELECTRON DIFFRACTION

Most of the conventional UED experiments have been constructed with dc or pulsed photocathode electron guns with beam energy of 30-200 keV. Unfortunately, the space charge effect 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 the electron number of 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, and 400 fs containing 1000 electrons at 60 keV. Alternatively, it is possible to increase the extracting electric field inside the gun, while the space charge effect is reduced. However, this approach is limited by the maximum electric field for vacuum breakdown.

As these reasons, in 2006, a MeV photocathode rf electron gun was firstly considered for UED. The demonstration of methodology was carried out. In the photocathode rf guns, which have been constructed for new development of accelerator physics, the electrons emitted from the photocathode surface are accelerated rapidly with a strong rf electric field (~100 MV/m or more) to reach relativistic speeds within a few millimeters. The increases in the pulse duration, emittance and energy spread due to the space charge effect are thus reduced to the minimum. An ultrashort electron beam at

Figure 3:RF gun based femtosecond electron diffraction

Figure 4:MeV diffraction pattern in single crystal Si foil

the energy of MeV with low emittance and high electron charge is thus generated. As a typical example of a 1.6-cell S-band rf gun developed in Brookhaven National Laboratory, the transverse normalized emittance of 1.2 mm-mrad at bunch charge of 1 nC was obtained [3]. The previous studies indicate that a low emittance femtosecond-bunch electron 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 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: (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 femtosecond laser pulse split from the output of the laser amplifier is also used as a pump pulse. The femtosecond electron beam is propagated to the sample with a magnetic lens 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.

Figure 2 shows the demonstration of MeV electron diffraction measurement in a 100-nm-thick Si sample. The beam energy of the electrons was 2.9 MeV. The electron charge was 0.2 pC/pulse. 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°. An important advantage of using the rf gun to produce femtosecond electron pulse is its capability of compressing the pulse duration as it is being accelerated in the time-dependent rf field. This permits the use of a longer laser pulse and further reduces the space charge effect near the cathode region. The beam simulation indicates that the pulse compression due to the rf is occurred at $<60^o$. A 50-fs electron bunch can be generated at 30° by using a 100-fs laser pulse [4]. The low energy spread of UED also can be obtained at the low laser injection phase, i.e. 10^{-4} at $<30^o$.
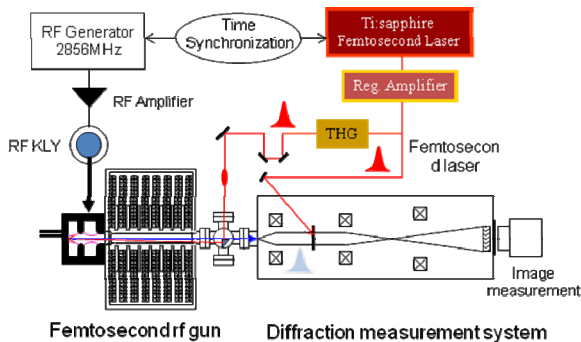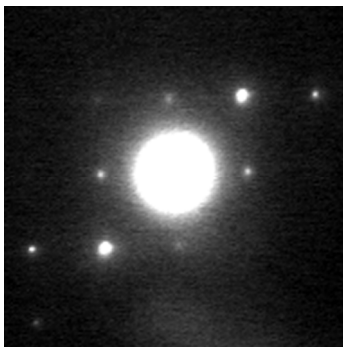
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