DEVELOPMENT OF FEMTOSECOND AND ATTOSECOND PULSE RADIOLYSIS BY USING LASER PHOTOCATHODE RF GUN S-BAND ELECTRON LINAC

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Abstract

Femtosecond pulse radiolysis system based on linear accelerator was developed in Osaka University for study of radiation-induced ultrafast physical and chemical reactions. A 32 MeV single electron pulse with pulse width of 98 fs was generated by using a laser photocathode rf gun s-band linac with a magnet pulse compression system. A femtosecond laser synchronized with the linac was used as analyzing light. An equivalent velocity spectroscopy, as a new method to get high time-resolution, was developed for measuring transient absorption in sample. By using the spectroscopy, we have started the preliminary experiment on attosecond pulse radiolysis. A new concept of double-decker electron linac was proposed to improve the time resolution to attosecond. The femtosecond double-decker electron beams were successfully generated with a laser driven photocathode rf gun.

INTRODUCTION

The geminate ion recombination has been studied as an important reaction in the primary process of radiation chemistry [1, 2]. The initial distributions and geminate decay were observed by using picosecond pulse radiolysis, and the recombination process was analyzed by the Smoluchowski equation base on the diffusion theory.

The picosecond pulse radiolysis, which is one of the promising methods to measure such a fast reaction, has been developed all over the world by using a picosecond electron pulse. Previously, the measurement of reactions that occur within 30 ps had been difficult because of a low time resolution for a few decades. Recently, a new picosecond pulse radiolysis system, in which a femtosecond laser was used, was proposed. The synchronization of the femtosecond laser with the sub-picosecond electron single pulse was succeeded at the Institute of Scientific and Industrial Research, Osaka University [3]. Very recently, a new pulse radiolysis system was developed to obtain a higher resolution by using a sub-picosecond electron pulse and a jitter compensation system [3]. We re-examined the geminate process on sub-picosecond time scale. However, only tail part of the geminate decay could be observed, because the time resolution of the pulse radiolysis was limited on a few picoseconds or sub-picosecond.

In order to investigate the reactions and the processes in femtosecond time region, a new femtosecond pulse radiolysis is developed by using a femtosecond electron pulse produced with a photocathode RF gun linac. An equivalent velocity spectroscopy was developed by an oblique incidence of the probe light to reduce the degradation of velocity difference between the electron and the laser light in samples.

An attosecond pulse radiolysis without the analysis laser light has been also studied. A new concept of double-decker electron linac was proposed to improve the time resolution to attosecond.

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FEMTOSECOND PULSE RADIOLYSIS

Figure 1 shows the femtosecond pulse radiolysis system. The system consists of a femtosecond electron linac as an irradiation source, a femtosecond laser as an analyzing light, and a jitter compensation system. The sample was irradiated by the femtosecond electron single pulse. The radiation-induced reactions were measured by the synchronized femtosecond laser light

Femtosecond Electron Source

The femtosecond electron single pulse was generated by a photocathode linear accelerator with a magnetic pulse compressor. A 1.6-cell S-band (2856MHz) RF gun was used for the electron pulse generation. The copper cathode used in the system was driven by an all solid-state LD-pumped Nd:YLF picosecond laser. The pulse width of the laser light (262nmUV) was 5 ps in FWHM. A single solenoid magnet was mounted at the exit of the RF gun to compensate the space charge emittance. The electron beam produced by the RF gun was accelerated with a 2 m long S-band travelling-wave linear accelerator. The linac was located at a distance of 1.2m from the cathode surface. The energy of the electron pulse was also modulated by adjusted the RF phase for magnetic pulse compression, as described below.

The magnetic bunch compressor was constructed with two 45°-bending magnets and four quadrupole magnets (two pairs), which provides the necessary path length dependence on energy. The picosecond electron bunch, which was produced in the linac with an energy-phase correlation, was compressed into femtosecond by rotating the bunch in the longitudinal phase space distribution. 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. Finally, the compressed bunch length was obtained by measuring Cherenkov radiation emitted from the electron bunch in air at the exit of the compressor with a femtosecond streak camera (HAMAMATSU, FESCA-

200, C6138). The time resolution of the streak camera in the measurement time region of 20 ps was 183 fs in FWHM and 78 fs in rms. An ultrashort electron bunch with 98 fs in rms was obtained at charge of 0.17 nC [4].

Femtosecond Analysis Light Source

A mode-locked Ti:Sapphire femtosecond laser was used as an analysis light source in the pulse radiolysis measurement. The Ti:Sapphire laser oscillator output was phase-locked with the 79.3 MHz RF (the 36th sub-harmonic of the 2856MHz accelerating RF). The laser pulse width was 60 fs.

In order to obtain a good s/n ratio, the femtosecond oscillator light was selected by a pulse selector based on an acoust optic modulator (AOM). A pulse train including 7 pulses with time interval of 12.6 ns from pulse to pulse was used to compensate the laser energy fluctuation during the transport of the laser light, in which the fourth pulse (I) was used as the reference light (I_0), and fifth pulse was used for probe of the absorption (I), called as "double pulse method". The intensities of the both laser pulses were measured by a Si photodiode. The optical density was obtained as $\log(I_0/I)$. The time delay was adjusted by guiding the laser light into an optical delay. The resolution in the optical delay was 80 fs.

Time Jitter Compensation

The time jitter between the electron pulse and the laser pulse also limits the time resolution of pulse radiolysis. In the presented system, we used a time jitter compensation technique based on a femtosecond streak camera. That is, we measure the temporal profiles of both the electron pulse and the laser pulse with the femtosecond streak camera, and calculate the precious time interval between the laser pulse and the electron pulse from the images in the camera. The time interval is then used as the time delay between the laser pulse and the electron pulse in the pulse radiolysis. The time jitter of few picoseconds can be compensated in the system.

Equivalent Velocity Spectroscopy

Finally, the time resolution of the pulse radiolysis is limited with the degradation of time resolution due to the velocity difference between the electron and the light in sample. An equivalent velocity spectroscopy was developed by an oblique incidence of the probe light to reduce the degradation of velocity difference between the electron and the laser light in samples. Figure 2 gives the concept of the equivalent velocity spectroscopy. The electron beam and the laser light were injected into sample with an incident angle θ. We set the incident angle as $\cos\theta=1/n$, where n is the refractive index of sample. If the electron pulse shape is also rotated with a same angle θ, the shape of electron beam pulse is overlapped with the laser pulse at all times. Therefore, the degradation of time resolution due to the velocity difference between the electron and the light in sample with refractive index of n is avoided.

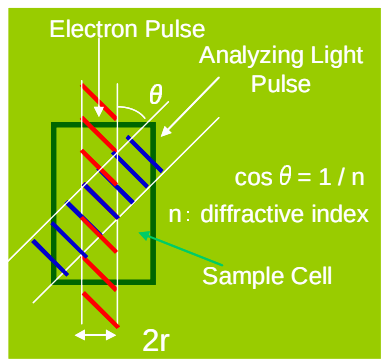

Fig.2 Concept of equivalent velocity spectroscopy

With the equivalent velocity spectroscopy, optical absorption of the hydrated electron was measured at 800nm, as shown in Fig. 3. Rise time of absorption by rotating the electron pulse shape is 1.4ps faster than that

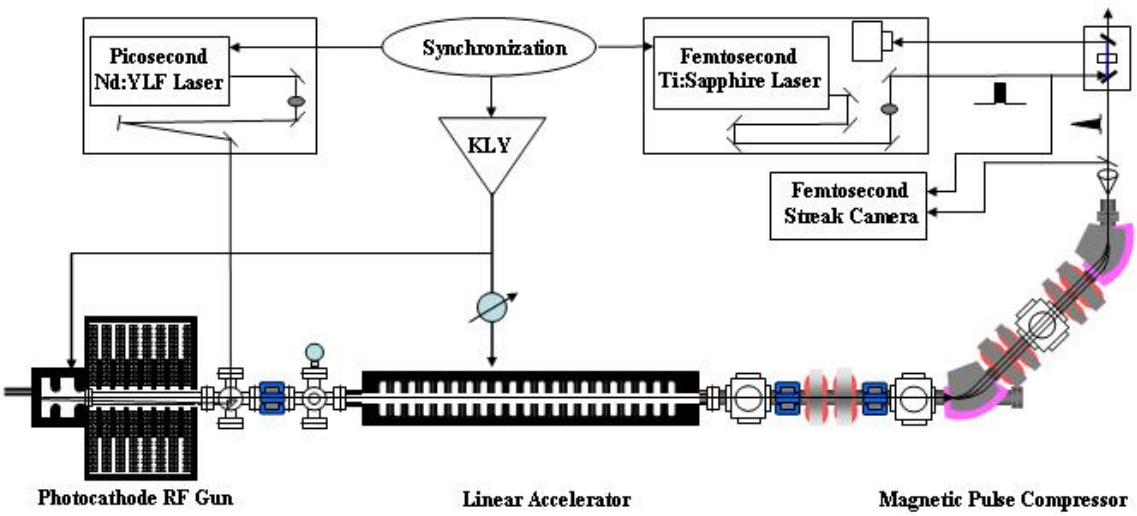

Figure 1: Block diagram of femtosecond pulse radiolysis system.

without the electron pulse shape rotation. The data indicates that the equivalent velocity spectroscopy is a useful tool for improvement of time resolution by the pulse radiolysis.

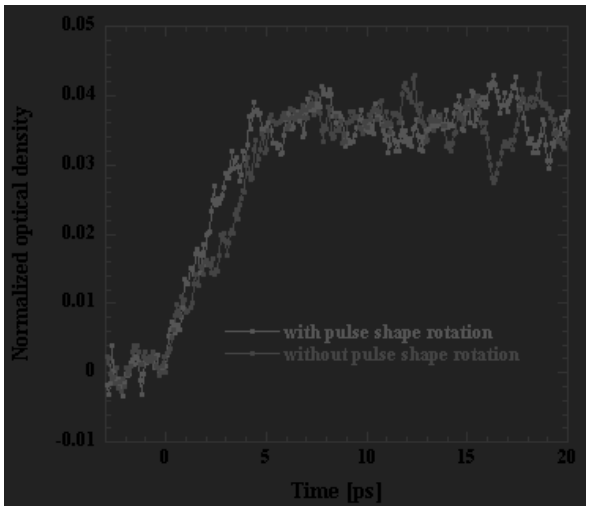

Fig. 3, optical absorption of hydrated electron in water measured with equivalent velocity spectroscopy at 800nm.

ATTOSECOND PULSE RADIOLYSIS

In order to improve the time resolution to attosecond, we are going to develop an attosecond pulse radiolysis system based on a new double-decker electron beam generation technique. Figure 4 shows the attosecond pulse radiolysis system. In the system, both the irradiation source and the analyzing light are generated by a double-decker electron beam linac.

The double-decker electron beam linac was consisted of a 1.6-cell S-band photocathode rf gun, a booster linac and a magnetic pulse compressor, which was same as the femtosecond electron pulse generation system described above. An injection optics system of double laser beams was used for double electron beam generation in the photocathode rf gun. As shown in Fig. 4, the incident UV laser beam produced by a Nd:YLF picosecond laser was divided into two by a beam splitter. The time interval of the two pulse should be the integral multiple of 350ps, because the RF gun is operated by 2856 MHz (1period =350ps) rf. The time delay was adjusted to 1.4ns in the experiment by guiding one laser beam into an optical delay. Finally, the UV light was injected on the cathode surface at an incident angle of approximately 2^{o} along the direction of the electron beam using a prism placed downstream of the gun. The diameter of the beam size at the cathode surface was 1 mm for both the beams.

Figure 4 gives the beam profiles of the double electron bunches at the exits of the rf gun, the linac and the bunch compressor. The beam profiles at the exit of the bunch compressor were measured in air. The profiles show that the double electron beams were generated in the rf gun and accelerated in the linac with same distance between two beam, which was called “double-decker beam”. However, the operation of the bunch compressor may be not optimal, resulting in the distance increase between the two beams at the exit of the compressor. The relative energy spread of the double beams downstream of the linac was measured to be 0.08-1.2% for both the beams. The normalized emittance of the double beams was obtained to 2.5 mm-mrad and 3.6 mm-mrad. The compressed pulse width was obtained to 380 fs in rms., seen ref. [5] for detail.

One of the double-decker electron beams will be used as a pump source, while another is used to produce a probe light source with Cherenkov radiation. The double-decker electron beams were generated in one accelerator and one laser. The effect of time jitter between the pump and probe sources on the time resolution is negligible.

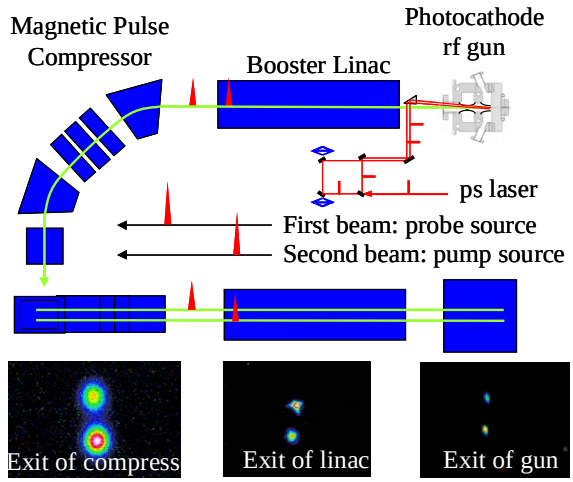

Fig. 4 A double-decker electron beam generation system.

CONCLUSION

Femtosecond and attosecond pulse radiolysis based on a photocathode RF gun linac were presented for the study of the ultrafast radiation-induced reactions. An equivalent velocity spectroscopy, as a new method to get high time-resolution, was developed for measuring transient absorption in sample. A new concept of double-decker electron linac was proposed and successfully used to generate femtosecond double-decker electron beams with a laser driven photocathode rf gun. The double beams are expected in the pulse radiolysis system for the analysis of ultrafast physical and chemical reactions on femtosecond /attosecond time scale.

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