ISIR SUBPICOSECOND PULSE RADIOLYSIS SYSTEM

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Abstract

The highest time resolution of the pulse radiolysis had remained about 30 ps since the late 1960's. To make clear the primary processes in the radiation chemistry and physics within 30 ps, we developed a stroboscopic pulse radiolysis system for the absorption spectroscopy with the time resolution of 2.0 ps (10-90% rise time). The time resolution of 2.0 ps was estimated from the time-dependent behavior of the hydrated electrons. The system consists of a subpicosecond electron linac as an irradiation source, a femtosecond laser as an analyzing light and a jitter compensation system.

1 INTRODUCTION

Pulse radiolysis is a very powerful method to detect and observe transient phenomena in radiation-induced reactions. The first experiment in the picosecond regime was carried out by the picosecond pulse radiolysis system of Toronto University with a time resolution of about 30 ps in the late 1960's [1]. Since then, several types of picosecond pulse radiolysis systems were developed [2-4] and many researches have been reported on ultrafast phenomena in radiation chemistry, physics, biology and applied fields such as material science.

Recently, a new picosecond pulse radiolysis system, in which a femtosecond laser was used as an analyzing light instead of the Cherenkov light, was proposed and developed at the Institute of Scientific and Industrial Research (ISIR), Osaka University [5], [6]. This system covered the wide wavelength from ultraviolet to infrared region by using second harmonic generation, third harmonic generation, optical parametric oscillation techniques and so on. The time resolution was several tens picoseconds. The primary processes in radiation chemistry of water, materials for microelectronics and so on have been investigated using this system.

Our pulse radiolysis system by using a femtosecond laser synchronized to the electron linac has a potential to detect ultrafast phenomena in the femtosecond region because the pulse width of analyzing light is less than 100 fs. We attempted to construct a higher resolution system for the investigation of the primary processes of the radiation chemistry and physics within 30 ps.

2 STROBOSCOPIC TECHNIQUE AND TIME RESOLUTION

To detect ultra fast phenomena in the time region of picosecond, a so-called stroboscopic technique is used [1-6]. The short-lived intermediates produced in a sample by very short radiation such as electron beams are detected by measuring the optical absorption of very short analyzing light such as Cherenkov radiation (femtosecond laser in our system). The time profile of the optical absorption can be obtained by changing the delay between the electron beam and the analyzing light.

The time resolution of the stroboscopic pulse radiolysis depend on the length of irradiation pulse, that of analyzing light and the jitter between them (Fig. 1). It does not depend on the time resolution of detection system such as a photodiode, an oscilloscope and so on.
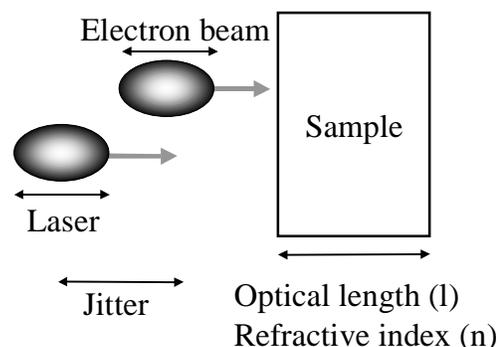
Fig. 1 Factors which limit the time resolution of the stroboscopic pulse radiolysis
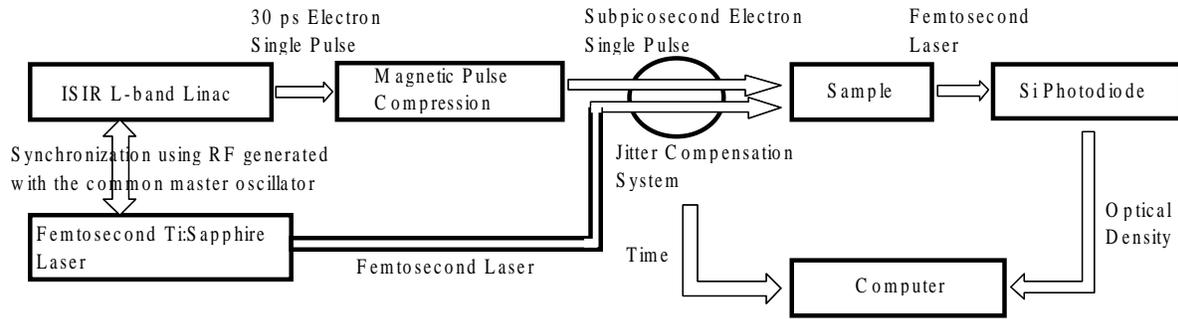
Fig.2 Subpicosecond pulse radiolysis method

For the development of subpicosecond pulse radiolysis system, a subpicosecond electron pulse, a subpicosecond analyzing light and precise synchronization are necessary.

Furthermore, the time resolution is limited by the difference between the velocity of the light and that of the electron pulse in a sample. The passing time of the electron pulse through a sample is given by $l/(\beta c)$, where l is an optical length of a sample, β the ratio of the velocity of electron to that of light in the vacuum and c the velocity of light in the vacuum. On the other hand, in the case of light, the passing time is given by ln/c, where n is a refractive index of a sample. Therefore, the time resolution is limited by the thickness of a sample. For example, the time resolution is limited to >1.8 ps (10-90% rise time) at $n = 1.33$ and $l = 2$ mm.

3 SUBPICOSECOND PULSE RADIOLYSIS SYSTEM

Figure 2 shows the subpicosecond pulse radiolysis system. The system consists of a subpicosecond electron linac as an irradiation source, a femtosecond laser as an analyzing light, and a jitter compensation system. A sample was irradiated by a subpicosecond electron single pulse. The time-resolved optical absorption was detected with a femtosecond laser which was synchronized to the electron pulse. The intensity of the laser pulse was measured by a Si photodiode. The timing between the electron pulse and the laser pulse was controlled by radio frequency (RF) system. The time profile of the optical absorption could be obtained by changing the phase of the RF with an electrical phase shifter. All equipment described below was controlled by a personal computer. The acquisition time was 1 second per one shot.

The ISIR linac consists of an thermionic electron gun, two 108 MHz subharmonic bunchers (SHBs), a 216 MHz SHB, a 1300 MHz accelerating

tube and a focusing system [7]. The magnetic pulse compressor consists of two 45° sector magnets, four quadrupole magnets and a vertical beam slit as shown in Fig. 3. The longitudinal energy distribution of the electron pulse was modulated so that the energy of electrons in the early phase of the pulse was higher than that in the later phase of the pulse. The phase of accelerating electric field was 70°. The peak energy of accelerated pulse was 26.5 MeV. The energy spread after the modulation was 9.4 %. The pulse length was approximately 30 ps at the end of the accelerating tube. In the magnetic pulse compressor, high energy electrons in the early phase take a long path and low energy electrons in the later phase take a short path. By translating the energy dispersion into the difference of the trajectory length, the electron pulse is compressed at the end of the magnetic pulse compressor. This system can compress the 30 ps electron single pulse to subpicosecond [8].

A modelocked Ti:Sapphire laser (Tsunami, Spectra-Physics Lasers, Inc.) was synchronized to the ISIR L-band Linac using a commercially available phase lock loop. The frequency of the laser was 81 MHz. On the other hand, the ISIR L-band Linac was driven by 108 MHz RF. The frequency of 27 MHz, which is the greatest common divisor, was used as a common master
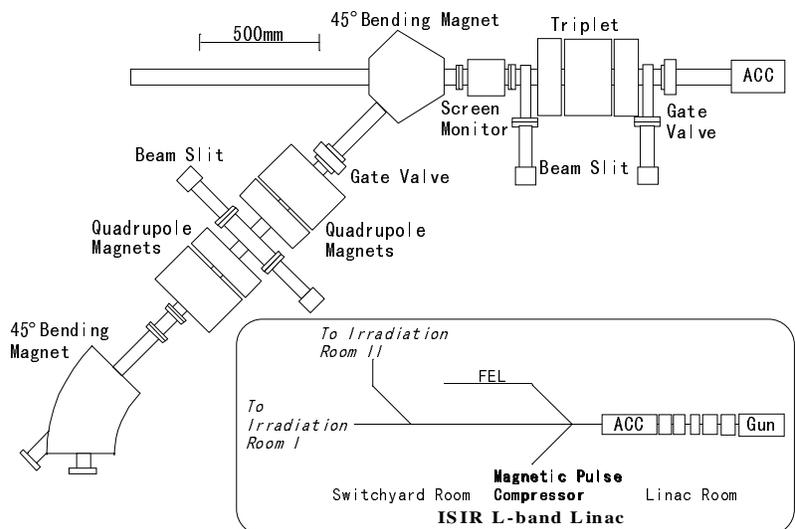
Fig. 3 Magnetic pulse compressor

oscillator. The jitter between the laser pulse and the electron pulse was several picoseconds from the measurement using a streak camera (C1370, Hamamatsu Photonics Co. Ltd.).

In order to avoid effects of the jitter between the electron pulse and the laser pulse on the time resolution, a jitter compensation system was designed as shown in Fig. 4. The time interval between the electron pulse (Cherenkov light) and the laser pulse was measured by the streak camera. The Cherenkov radiation was emitted by the electron pulse in air at the end of the beam line. The laser pulse was separated from the analyzing light by a half mirror. The precise time interval could be obtained by the analysis of the streak image.
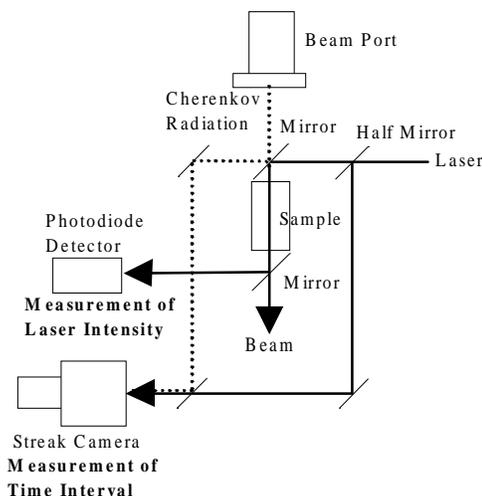
4 PULSE RADIOLYSIS EXPERIMENT

Radiation induced reactions in the water was observed. Distilled water was used as a sample. The water was deaerated in a quartz cell with the optical length of 2.0 mm. It is known that a broad absorption due to hydrated electron with the maximum at the wavelength of 720 nm is observed in neat water at an irradiation of electron beam [9]. The formation process of hydrated electrons have been investigated by femtosecond laser flash photolysis. It has been reported that the hydrated electron is formed on a subpicosecond time scale [10-14]. Figure 5 shows the time-dependent behavior of the hydrated electron obtained in the pulse radiolysis at the wavelength of 780 nm. The 10-90% rise time was 2.0 ps. It is found that the time resolution of this system is less than 2.0 ps.

5   CONCLUSION

The direct observation of radiation-induced reactions with the time resolution of 2.0 ps (10-90% rise time) has been succeeded using the subpicosecond electron pulse, the femtosecond laser and the jitter compensation system.

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Fig. 4 Jitter compensation system for pulse radiolysis.

Fig. 5 Time-dependent behavior of hydrated electron obtained in the subpicosecond pulse radiolysis, monitored at the wavelength of 780 nm.

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