Development of Subpicosecond Pulse Radiolysis System (II)

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Abstract

For the development of femtosecond pulse radiolisys system, we have investigated and developed the generation technique of ultra short electron pulse and the detection system for the ultra fast phenomena. The detection system with the femtosecond electron pulse generated by the magnetic pulse compression method achieved the time resolution of 800 fs. For the further improvement of the time resolution and the S/N ratio, a new algorithm has been applied in the system, which has improved the S/N ratio of the 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 [2-5] 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 [6], [7]. Then, we attempted to produce an ultra short electron pulse [8] and apply it to the pulse radiolysis. The biggest problem to achieve higher time resolution is timing jitter between a laser pulse and an electron pulse. In order to reduce the timing jitter, it is necessary to reduce the environmental factor such as mechanical vibration and fluctuation of coolant temperature, room temperature and power supply. However, it does not seem feasible to reduce such factors of the whole accelerator system because the accelerator system consists of many pieces of equipment and occupies large space. We opted to compensate the effects of timing jitter [9] and achieved the high time resolution of 800 fs. However, the number of observable phenomena has been limited due to its poor S/N ratio. We attempted to improve the S/N ratio in order to achieve the higher time resolution and pursue the investigation of the primary processes of the radiation chemistry and physics within 30 ps.

2. Subpicosecond Pulse Radiolysis System

Figure 1 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 timing detection 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

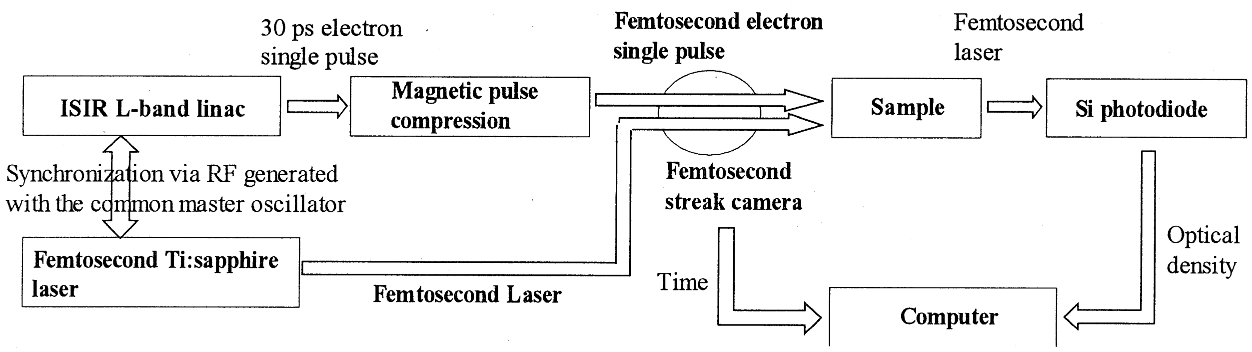

Figure 1. Diagram of subpicosecond pulse radiolysis system

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 [10]. The magnetic pulse compressor consists of two 45° sector magnets, four quadrupole magnets and a vertical beam slit as shown in Fig. 4. 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 subpicoseond [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 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 timing detection system was designed. The time interval between the electron pulse (Cherenkov light) and the laser pulse was measured by the streak camera at every shot. 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.

3. Improvement of S/N ratio

It is necessary to use a thin sample in high time resolution pulse radiolysis. The use of thin sample leads to the degradation of S/N ratio, because the signal intensity is proportional to the optical length of sample. Therefore, achieving the high time resolution requires the measuring system with good S/N ratio. In pulse radiolysis experiments, the optical density is generally calculated with the intensity of probe light passing through sample with an electron beam present (I) and that of probe light passing through sample with an electron beam absent (I0).

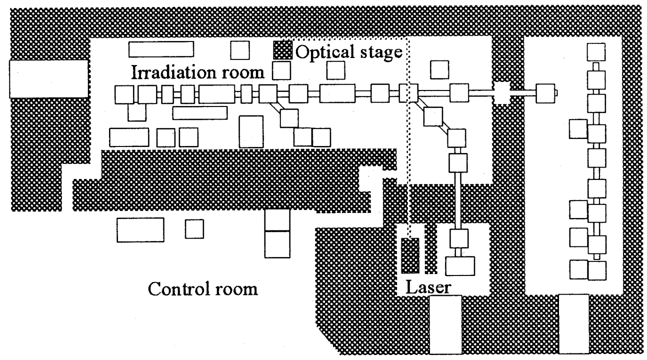

Figure 2. Layout of Experimental Apparatus

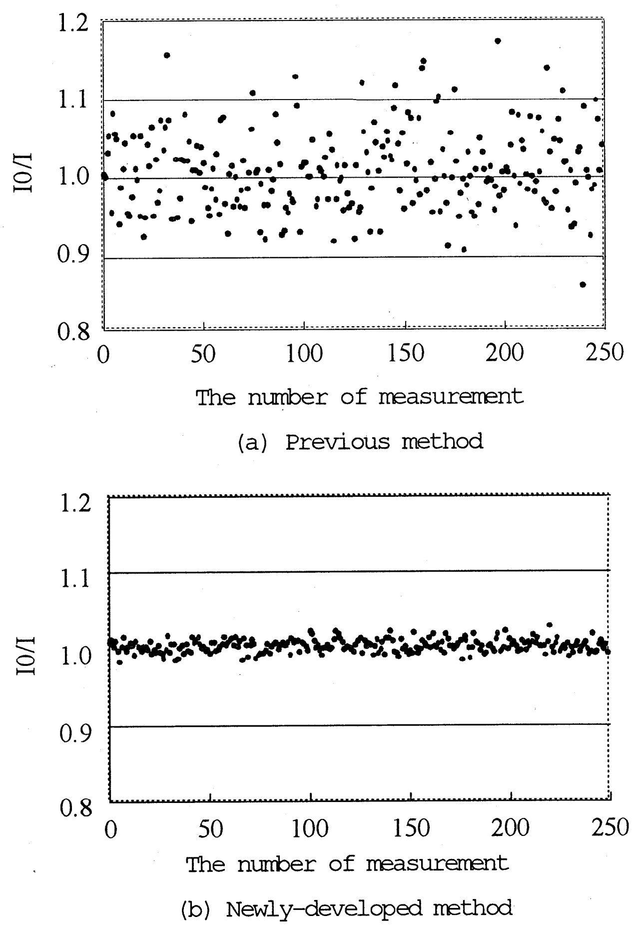

Figure 3. The fluctuation of I0/I

If there is no fluctuation of laser intensity, we have only to measure I0 once. Actually, the envelope of laser pulses changes in the relatively long-term range (typically from several minutes to a few tens minutes) due to the change of environmental factors such as room temperature, coolant temperature and so on. There is also an intensity jitter caused by the mechanical vibration of mirrors, the timing jitter of electronics and so on. Furthermore, in our system, the laser system is located away from the beam port in order to avoid the radiation damage of the laser system. The layout of the subpicosecond pulse radiolysis system is shown in Fig. 2. The laser system was located about 15 m away from the beam port. Later, it was moved into a clean room, which was installed in the control room to keep the room temperature constant and to keep the laser system clean. The distance is still about 10 m. Therefore, it is predicted that a slight tilt of a mirror placed upstream causes a displacement of laser pulse at the downstream position where photo detector is placed.

The fluctuation of I/I0 is shown in Fig. 3(a), measured at the position where the photo detector is located. The fluctuation of I/I0 was 5 %. A new algorithm to reduce the intensity jitter caused by mechanical vibration was applied to the subpicosecond pulse radiolysis system. The fluctuation of I/I0 is shown in Fig. 3(b). It is reduced to 0.8 % with the newly-developed method. The time-dependent behaviors of solvated electrons in methanol are shown in Fig. 3 and 4 as an example of experimental result. Two components which have different decay times are observed in the result obtained with the new method. The fast component is unobservable in the result obtained with the previous method because of its poor S/N ratio. The fast and slow components are thought to be due to a presolvated electron and a solvated electron, respectively.

4. Conclusion

The fluctuation of I/I0 was reduced from approximately 5 % to less than 1 %. The S/N ratio of the system was improved. It enabled further experiment on the radiation induced reactions in materils.

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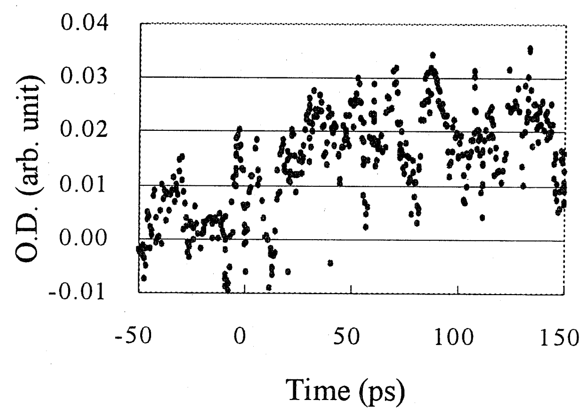

Figure 4. The time-dependent behavior of intermediates generated in neat methanol, measured with the previous subpicosecond pulse radiolysis system at the wavelength of 790 nm.

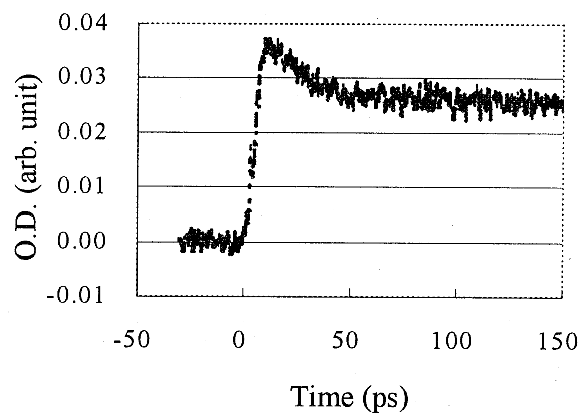

Figure 5. The time-dependent behavior of intermediates generated in neat methanol, measured with the newly-developed subpicosecond pulse radiolysis system at the wavelength of 790 nm.

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