SUB-PICOSECOND PULSE RADIOLYSIS PROJECT AT NERL, UNIV. OF TOKYO

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Abstract

Pulse radiolysis is a method of time-resolved absorption of the chemical spieces. It is able to measure the reactions in the time range from sub-ns to ms. The highest time resolution has been achieved up to 100 ps. Many primary processes in the time scale of ps or even sub-ps are still remained to be investigated. A new sub-picosecond pulse radiolysis project at Nuclear Engineering Research Laboratory (NERL), University of Tokyo, is under construction. Generation of a sub-ps electron single pulse and a fs Ti:Sapphire laser pulse, and the synchronization of them within 1ps will be inevitable for this purpose.

1 INTRODUCTION

It is widely recognized that the pulse radiolysis technique is very useful and powerful to study chemical reactions in addition to laser photolysis. A large number of the rate constants of the reactions has been measured and much knowledge has been accumulated in liquids, especially in aqueous solutions. Therefore, the pulse radiolysis method has been applied to various subjects such as radical reactions in biology, medicine and atmospheric chemistry, and radiation effects in nuclear technology. At NERL, Univ. of Tokyo, intensive work using the pulse radiolysis method has been carried out not only in aqueous solutions but also in organic liquids and polymeric systems [1, 2].

There are two electron linear accelerators (see Fig. 1), both of which can provide single ps and sub-ps electron pulses. Much effort has been done to reach the higher time resolution. The time resolution of the pulse radiolysis method has attained to 100 ps. Many primary processes, which characterize radiation induced reactions, such as solvation of e_{aq}^-, anions and cations, geminate ion recombination and the formation of the excited states in organic liquids etc., are still remained to be investigated. These reactions take place in the time scale of ps, even sub-ps. Thus, in order to investigate the elementary processes of radiation induced phenomena, the development of the experimental system with much higher the time resolution has been expected.

In this paper, the present pulse radiolysis system and new system with time resolution of ps and sub-ps is presented which under construction at NERL. Details of the new system is presented taking into consideration several technical subjects which affect the time resolution.

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2 PULSE RADIOLYSIS SYSTEM

2.1 Conventional system

The pulse radiolysis system consists of pulsed radiation and analyzing light sources together with a synchronization system for them. In conventional systems, the 35 MeV linac is used as the pulse of electron source. The analyzing lights are cw and pulsed which are selected according to the time region of target reactions. The electron pulses and analyzing light are synchronized by the electrical devices including the delay units and phase shifters. Change of the analyzing light after passing the sample solution and a monochromator is detected by a photodetector and displayed on the oscilloscope combined with the computer for data acquisition and analysis. Absorption of the analyzing light obeys the Lambert-Beer law,

$$log_{10}\,(I_0\,/I) = \varepsilon\, C\, l \qquad (1)$$

where I_0, I, ε, l and C are the intensities of the analyzing light without and with absorption, a molar absorption coefficient, the length of the sample cell and the concentration of species of interest, respectively.

The time resolution of this system is determined by the speed of the detection system, the photodetector and the oscilloscope. This system can achieve to sub-ns only when the fast detector and oscilloscope are utilized. Therefore, a

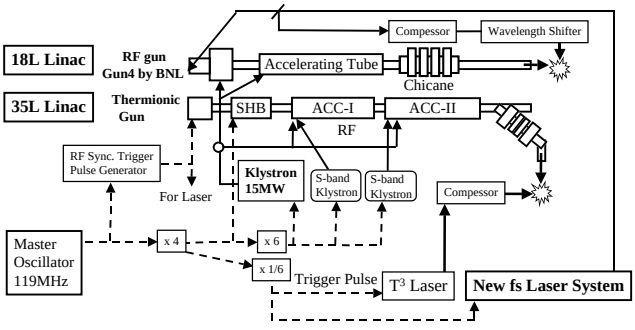


Fig. 1. Synchronization system

stroboscopic method, so called a pump & probe method, should be introduced to perform the experiments in a ps scale.

2.2 Present system

The stroboscopic method (see Fig. 2) was developed by Hunt in 1968 for the first time. In this method Cherenkov pulses associated with electron multi-pulses were used for the analyzing lights and the precise synchronization between Cherenkov and electron pulses is automatically fulfilled. Here, the electron and Cherenkov pulses can be assumed as pump and probe ones, respectively. The intensity of I_0 can be obtained by using a lead block in front of the sample to avoid irradiation and I is taken under irradiation after the optical delay line which can adjust the time delay [5, 6].

As mentioned before, single ps electron pulses are available and the single 65 fs (rms) light pulse is also fed by the Ti:Sapphire laser at NERL. In addition, the synchronization of the electron and laser pulses with the time jitter of less than 4 ps (rms) is attainable as shown in Fig. 3 [3]. As the preliminary step, the picosecond pulse radiolysis system was set up as shown in Fig. 1 before introducing the new one which is discussed later. The preliminary experiments were done to check the feasibility of the new system.

The fundamental light pulse of 790 nm from the Ti:Sapphire laser was splitted into two pulses used for a reference and an analyzing light detected by the two PIN photodiodes. The first measurement is to watch the temporal behavior of the hydrated electron at 790 nm. Another was a measurement of the spectrum of the hydrated electron using a optical multi-channel analyzer (OMA) based on after the generation and splitting of the white light produced by the focusing of the fundamental light in a water cell. Here, the two white light pulses were used as the reference and analyzing lights, which were fed into two optical fibers. The OMA consisting of the two photodiode arrays which are sensitive to the wavelength region of from 200 to 1000 nm is a kind of imaging spectrograph.

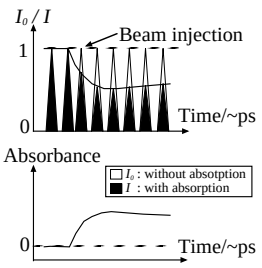


Fig.2. Stroboscopic method

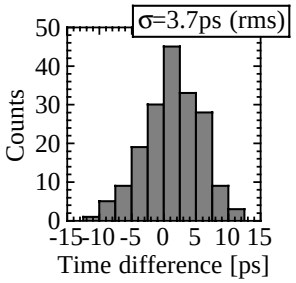


Fig.3. Synchronization of 35L linac & T^3 laser

Obtained results are shown in Figs. 4 and 5. In spite of the not enough S/N ratio, the formation of the hydrated electron having the peak at 720 nm was confirmed. The experiences obtained at this system are useful to the new system. The delay was adjusted by the delay line of the cable. The timing between the laser and electron pulses was adjusted by monitoring both the laser pulse and the Cherenkov radiation pulse associated with the electron pulse with the femtosecondstreak camera (FESCA200, Hamamatsu). The total time resolution is evaluated as around 10 ps (rms), which is determined by the widths of the electron (5ps(rms)) and laser pulses(65fs(rms)), the timing jitter of the electrical synchronization devices (about ps), the time delay between the electron and light pulses after passing through the 1.8 cm water cell.

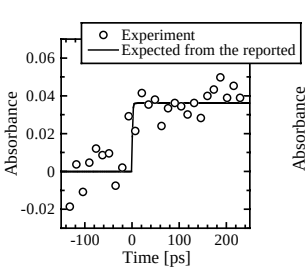


Fig. 4. Time profile of e_{aq}^-

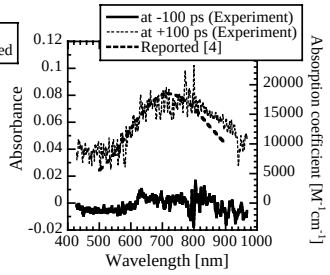


Fig. 5. Optical spectra of e_{aq}^-

3 NEW PULSE RADIOLYSIS SYSTEM

3.1 Synchronization system (see Fig. 1)

For further development to achieve higher time resolution, several technical subjects which govern the time resolution should be considered. These factors are classified into the two; the widths of the electron and analyzing pulses, and the timing jitter between them. As for the width of electron pulse, ps and sub-ps pulses are available by using the SHB buncher and the magnetic compressing system, respectively, at present. The analyzing light is fed from the newly installed fs Ti:Sapphire (44fs(rms), 0.3TW, 790nm) laser.

In the present system the synchronization devices provide the trigger pulses to the thermionic electron gun and the Ti:Sapphire fs laser so that its timing jitter hardly becomes down to 1 ps. Therefore, the new synchronization method is required. The fs laser light from the new Ti:Sapphire laser is splitted into two pulses, which are used for electron pulse generation at the laser photocathode rf-gun [7] and analyzing light. As the two laser lights are produced from the same source, the timing jitter should be rather small.

Next, the timing jitter from other sources is considered. The first is the jitter from the accelerating process in the linac, namely the fluctuation of the accelerating rf. In the present system, the two klystrons are used and the fluctuation from the two klystron attributes the timing jitter. In the new system, only one klystron with higher power (15MW) Is employed. This can reduce mutual rf fluctuation of the two klystrons. The stability of the modulator voltage of the klystron also contributes to the timing jitter. It can be said that the timing jitter of the new system would be ramarkably suppressed compared to that of the present system. Of course, precise inspection

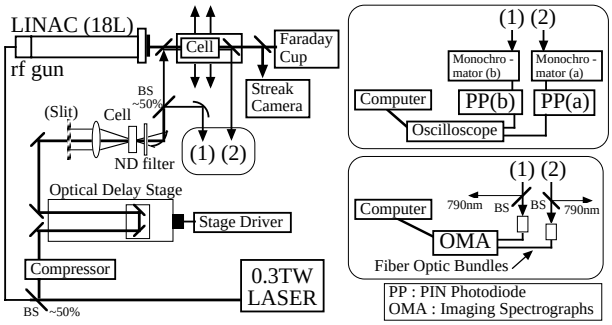
should be done after the construction.

3.2 Evaluation of the timing jitter

The timing jitter including the pump- and probe-sources was evaluated. Fluctuations of rf phase and power at the 15 MW klystron were measured to be 0.2 deg (p-p) and 0.02 dB (p-p), respectively. With these parameters, the timing jitter of the electron pulse in the acceleration process was simulated by PARMELA code to be 50fs (rms). Considering the timing jitter at Ti:Sapphire laser oscillator to be 100 fs (rms), the total timing jitter between the electron and laser pulses is expected to be less than 200 fs (rms) in the new experimental system.

3.3 Measurement system (Fig. 6)

Fig. 6. Measurement system of sub-ps pulse radiolysis



Total time resolution is determined by both the timing jitter of synchronization and the error in the measurement devices. Since the white light for probe is obtained by the non-linear conversion from the fundamental light (44fs(rms), 790nm), the jitter would be 100 fs (rms) at most. The accuracy of the delay line such as the reproducibility of the mechanical stage causes additional fluctuation. Furthermore, the time delay between the electron and laser pulses after passing through a sample cell because of the difference of their travelling speeds in the medium must be considered. It is clear from the equation (1) that the thinner sample cell introduces the smaller signal of absorbance. Therefore, it must be selected the optimal cell length.

4 CONCLUSION

A new sub-picosecond pulse radiolysis project at NERL, Univ. of Tokyo was introduced comparing with conventional and present system. By this system, direct measurement of the phenomena in the time range from sub-ps to ns, for example, electron solvation, geminate ion recombination in organic liquids and migration of electrons between organic and aqueous phase etc., is expected.

5 REFERENCE

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