PROGRESS OF THE EQUIVALENT VELOCITY SPECTROSCOPY METHOD FOR FEMTOSECOND PULSE RADIOLYSIS BY PULSE ROTATION AND PULSE COMPRESSION

T. Kondoh#, K. Kan, J. Yang, Y. Yoshida, H. Kobayashi, T. Toigawa, K. Norizawa, and A. Ogata, Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan

Abstract

Femtosecond pulse radiolysis is developed for studies of electron beam induced ultra-fast reaction in matter. 98 fs electron pulse was generated by a photocathode RF (radio frequency) gun linear accelerator (LINAC) with a magnetic bunch compressor. However for more fine time resolution, the Equivalent velocity spectroscopy (EVS) method is required to avoid degradation of time resolution caused by velocity difference between electron and analysing light in sample. In the EVS method, incident analysing light is oblique toward electron beam with an angle associated with refractive index of sample, and then, electron pulse is rotated toward the direction of travel to overlap with light pulse. In previous studies, pulse rotation had not been compatible with pulse compression. However, by oblique incident of light to the photocathode, pulse rotation was compatible with pulse compression, and the time resolution was improved by principle of the equivalent velocity spectroscopy.

INTRODUCTION

Radiation chemistry will become more important to next generation lithography and radiation therapy. In the next-generation semiconductor lithography, exposure light source become a shorter wavelength for the miniaturization of nanofabrication. Because it will ionize resist materials, radiation chemistry is important. The initial process of radiation chemistry, such as Ionization, thermalization, geminate ion recombination, dissociative electron attachment, begins with attosecond when high-energy electrons pass through an atom or molecule. In order to elucidate that whole picture, time resolution of a pulse radiolysis is improving with the aim of attosecond.

The pulse radiolysis is a powerful tool for directly observing the time-dependent behavior of the active species in the sample. On the pulse radiolysis of transient absorption spectroscopy, pulsed radiation induce some active species in sample and it is probed by analysis light. The stroboscopic method is used for higher time resolution than that of the detector and oscilloscope. Factors that determine the time resolution of the stroboscopic pulse radiolysis are the pulse width of the electron beam and analysis light, their synchronization jitter and a degradation which due to the velocity difference between the analysis light and electron beam in sample. Pulse width of the electron beam is 98 fs by using the photo-cathode RF gun LINAC with magnetic pulse compressor. Pulse width of the analysis light is about 150 fs by using a femtosecond laser. Timing jitter between the analysis light pulse and the electron beam pulse is about 60 fs in a previous study [1].

There is another important factor in the electron beam pulse radiolysis different from a laser photolysis. Although the high energy electron beam pass through the sample at about the light velocity, the analysis light become slow such as $v_l = c / n$, where v_l, c, n mean the light velocity in sample and vacuum, and the refractive index of sample respectively. The degradation of time resolution is expressed as $g(L) = L/c(1-1/\beta n)$ in the collinear configuration, where L, β is the optical pass length of sample and the velocity ratio of light and electron respectively. Refractive index of water is 1.33, it results in the degradation of time resolution of 11 ps in 1 cm thickness sample. So far the femtosecond time resolution was achieved by reducing this influence using very thin sample. In the absorption kinetic trace measured by the pulse radiolysis, because the horizontal and vertical axis means time and optical absorption respectively, not only the precision and purity of the timing, but also significant changes in the signal which overcome the noise due to the light intensity fluctuation are very important.

The EVS method is the solution of the g(L) problem, it has already been reported[1]. Principle of the EVS method is the overlapped propagation of the electron beam pulse and light pulse at every point in the sample. For more information, please refer to the reference.

There are problems in the previous study. In order to achieve femtosecond time resolution, it is necessary to compress the electron beam to femtosecond region. On the other hand, the electron beam pulse should be rotate toward the travelling direction in the EVS method. In the previous study, in order to rotate the electron pulse, the RF phase of LINAC was shifted from the pulse compression phase, and the electron pulse was modulated in energy. However, because the condition was changed from the pulse compression by this method, the electron beam could not be compressed anymore. For this reason, the time resolution of the EVS pulse radiolysis using a sample of 1 cm water was 6.4 ps.

Therefore, the longitudinal and transverse modulation of electron beam has been devised as a way to rotate the electron beam pulse with the pulse compression. The entrance port which was attached to the cathode by 68 degrees was used on the longitudinal and transverse modulation of electron beam. By obliquely incident, because of the different arrival time by the UV laser pulse to the cathode surface, the electron beam is generated

#t-kondo@sanken.osaka-u.ac.jp

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with angle to the propagation direction. After the compression, the oblique angle of the electron beam was controlled by changing the diameter of the electron beam in the final focus magnet. More information about the longitudinal and transverse modulation of electron beam will be reported elsewhere[2].

In this paper, the rotation angle and pulse width necessary for the EVS methods were measured in the case of using the longitudinal and transverse modulation of electron beam. In order to evaluate the time resolution, the rotation angle of electron beam and the rise time of the absorption of hydrated electron were measured.

EXPERIMENTAL

Pulse radiolysis system with the EVS method which is shown in Fig.1 is consisted of a photo-cathode RF gun LINAC, a magnetic bunch compressor, a femtosecond (fs) laser system for analysis light, time synchronization devices and an optical measurement system.

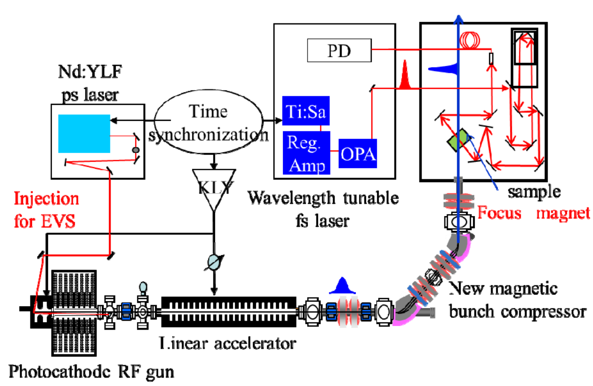

Figure 1: Pulse radiolysis system with the EVS method.

The laser for cathode excitation which is a picosecond Nd:YLF laser by Timebandwidth generate the 262 nm light pulse which is the fourth harmonic generation of 1047 nm. By the UV light pulse irradiated to the copper cathode, the electron beam pulse which has 2 nC/pulse and 5 ps was generated. The electron beam which is generated by the photoelectric effect at the copper surface of the cathode by the UV laser is accelerated by 2856 MHz RF in cavity. Generated electron beam pulse is accelerated up to 32 MeV by the traveling wave accelerator. The energy modulation for pulse compression in the magnetic bunch compressor is performed in the accelerator. By adjusting the electron orbits in the magnetic bunch compressor and by the energy modulation given in the accelerator tube, the incident electron pulse width of 5 ps is compressed to 98 fs. To focus the electron beam in the sample, a final focus magnet was installed. Analysis laser system is composed of a Ti: Sapphire femtosecond laser (by Spectra Physics) and a pulse selector. The pulse selector pick up 20 pulses synchronized with the electron beam pulse of 10 Hz from pulse train of 79.33 MHz. Electron beam irradiated the sample in quartz cuvette in air. In order to measure the optical absorption by the active species, the analysis light is transported to the irradiation point from the clean room. Analysis light is incident with electron beam by a thin aluminium mirror in front of the sample through the optical delay scanning the timing between the light and electron beam. Analysis light was detected by a biased Si-APD S2382 (by Hamamatsu Photonics) via from optical fiber. Double-pulse method of 79.33 MHz which was used the intensity of the analysis light pulse overlaps with the electron beam as I and the intensity of the analysis light pulse before two cycle (25 ns) as Io was performed. Optical density (O.D.) was calculated by the equation as O.D.= ln(Io/I). By using the double-pulse method, it was possible to reduce drift and low-frequency fluctuations such as vibration and airflow.

By using the Cherenkov radiation in air, the rotation angle and pulse width of the electron beam was measured by a streak camera FESCA-200 (by Hamamatsu Photonics). In order to avoid increasing of pulse width due to the dispersion in air, the spectral band pass filter of 480 nm was used. Slit width of the streak camera was the minimum of 15 μm. The time axis and horizontal axis shows the longitudinal and transverse distribution of the electron beam respectively. The time resolution is 200 fs in FWHM. For more detail, please see the reference [1]. Selecting the bunch area of a part of the streak image, the rotation angle was estimated by the method of least squares of light intensity of every pixels in selected region. Pulse width of the oblique electron pulse was determined by the Gaussian fitting to the distance distribution of the electron pulse from the center line of the pulse.

RESULTS AND DISCUSSION

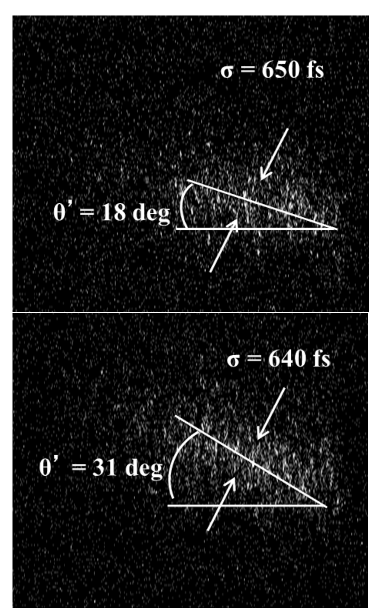

Figure 2: Streak camera images by using the longitudinal and transverse modulation of electron beam.

Figure 2 shows the results of measuring by the streak camera. These images was obtained by converting the raw streak image as for the same scale between the vertical axis and the horizontal axis by using the function Lz = ct for vertical axis. Left picture shows the electron pulse image with minimum angle of 18 degree and pulse width of 650 fs. Right picture shows the electron pulse image with angle of 31 degree and pulse width of 640 fs . The results of the measurements show that the electron beam can be rotated with pulse compression up to 650 fs by the longitudinal and transverse modulation of electron beam.

For the evaluation of the time resolution of the EVS pulse radiolysis, time dependent behavior of the absorption by the hydrated electron was measured. Because the rise time of the optical absorption by hydrated electron is very fast, it is suitable for the evaluation of the time resolution of the EVS pulse radiolysis. Rising behavior of the optical absorption by hydrated electron measured at 800 nm by electron beam irradiation to the 1 mm thickness water sample is shown in Fig.3.

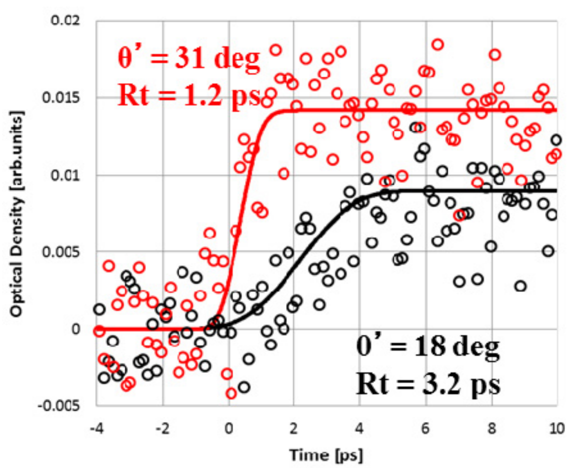

Figure 3: Rising behaviour of the optical absorption by hydrated electron measured at 800 nm.

Black circle means the condition of the 18 degree and 650 fs, and red circle means the case of 31 degree and 640 fs. One point shows averaged data of 50 times measurements. The Rise time of 10-90% is 3.2 ps at 18 degree of pulse angle and 1.2 ps at 31 degree. The time resolution was notably improved. The pulse width is about 650 fs in both cases, improvement of the time resolution is due to the angle of the electron beam. From this result, the time resolution of the EVS pulse radiolysis was improved to 1.2 ps. If the 1cm thickness water sample was used, the time resolution was improved from 6.4 ps to 2.8 ps.

The angle of the electron beam pulse toward the traveling direction versus the pulse width and the rise time of the optical absorption were shown in Fig.4.

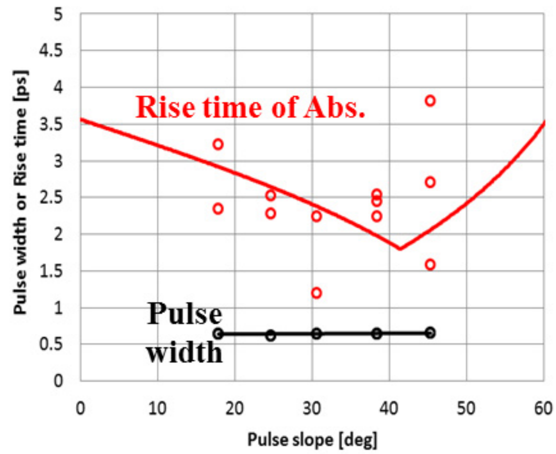

Figure 4: the angle of the electron beam pulse versus the pulse width and the rise time of absorption.

The longitudinal and transverse modulation of electron beam could be rotated with pulse compression. The pulse angle could be changed from 18 degree to 46 degree with the pulse width of about 650 fs. Solid line shows the theoretical degradation of the time resolution g(L). The time resolution is minimized theoretically at 41 degree associated with refractive index of water, obtained rise time vary widely in the experiments, the minimum rise time obtained at 1.2 ps in the case of the angle of 31degree. Behavior of the EVS method was shown in the results as the minimization of the rise time by angle. By the longitudinal and transverse modulation of electron beam, the electron beam pulse was rotated from 18 degree to 46 degree with the pulse compression to 650 fs, and the time resolution of the EVS pulse radiolysis improved to 1.2 ps.

CONCLUSIONS

The pulse rotation of the electron beam with the pulse compression was achieved by using the longitudinal and transverse modulation of electron beam. The time resolution of the EVS pulse radiolysis which had been remained at 6.4 ps was improved to 1.2 ps.

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