EQUIVALENT VELOCITY SPECTROSCOPY BASED ON FEMTOSECOND ELECTRON BEAM ACCELERATOR

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Abstract

A new femtosecond pulse radiolysis system, which is called as "Equivalent Velocity Spectroscopy (EVS)" based on a photocathode rf linear accelerator and a femtosecond laser, is developed in Osaka University for the study of primly process and ultrafast electron-induced reactions for the nanofabrication. In order to achieve a time resolution on femtosecond scale, a femtosecond electron beam bunch produced by a photocathode accelerator and a synchronized femtosecond laser were used. The electron bunch and laser pulse were injected with an angle determined by the refractive index of the sample. The electron bunch was also rotated with a same angle, resulting in the time resolution degradation due to the velocity difference between light and the electron in the sample is thus avoided. A jitter compensation technique with a femtosecond streak camera was used to reduce the time jitter between the electron bunch and laser pulse. Moreover, in EVS, a technique of double laser pulse injection was used successfully to improve the signal to noise ratio due to the fluctuation of the laser intensity during the measurement.

INTRODUCTION

Pulse radiolysis is a powerful tool for studying chemical kinetics and primary processes or reactions of radiation chemistry. In the pulse radiolysis, a short electron beam, which is almost produced by radio-frequency (rf) electron linear accelerator (linac) with energy from a few MeV to a few tens MeV, is used as a pump or irradiative source. The electron-induced reactions or phenomena in matter are analyzed by a short-pulse analyzing light (e.g. synchronized lasers or Cherenkov light emitted from the electron beam) with the time-resolved stroboscopic technique.

The electron bunches on the order of a few nanoseconds are in use worldwide for pulse radiolysis to study the reactions on the nanosecond time scale. To study ultrafast chemical kinetics or reactions, a picosecond pulse radiolysis system was developed at the University of Toronto in the late 1960s [1]. A 10 ps-long electron micro-bunch within the 35 ns electron pulse produced by the rf linac was used as the irradiative pulse, while Cherenkov radiation generated from the later electron bunch was used as the analyzing light. The Toronto instrument opened firstly a picosecond time region for pulse radiolysis, and many important reactions within spurs were observed. After the Toronto experiment, several groups (e.g. Argone National Laboratory, the University of Tokyo) have constructed new picosecond pulse radiolysis systems based on picosecond single-bunch electron linear accelerators. However, despite many innovations and modifications of the techniques that have been developed, the time resolution has remained at about 20 ps since the 1960s. Over the past a few years, several picosecond pulse radiolysis facilities based on a picosecond or sub-picosecond single-bunch electron pulse and a synchronized femtosecond laser light were developed at Osaka University [2,3], at Brookhaven National Laboratory [4], and at the University of Tokyo [5]. At Osaka University, a 28 MeV single-bunch electron beam with a bunch length of about 30 ps was produced by an L-band (1300MHz) electron linac system which was constructed with a grid-gated thermionic electron gun, a sub-harmonic pre-bunching system, and a L-band linac. After acceleration, a magnetic bunch compressor was used to compress the electron bunch from 30 ps to <1 ps. A synchronized Ti:Sapphire laser with pulse width of 60 fs was used as analyzing light source. Moreover, a time jitter compensation technique with a femtosecond streak camera was developed and used to reduce effect of time jitter between the sub-picosecond electron bunch and a femtosecond laser light on the time resolution of pulse radiolysis. Finally, the time resolution of pulse radiolysis was achieved to 2.0 ps.

At Brookhaven National Laboratory and the University of Tokyo, a photocathode rf electron gun linac and a femtosecond Ti:Sapphire laser were used in the pulse radiolysis. The photocathode rf gun produces a single picosecond electron bunch with charge of several nano-coulombs by using picosecond laser excitation on the photocathode without the need of a sub-harmonic pre-bunching system. The accelerator is compact comparing with the thermionic electron gun linac such as the L-band linac system. The electron beam produced from the rf gun is synchronized with the excited laser. It would be easy to synchronize the electron beam with the analyzing femtosecond laser light in picosecond time precision.

In the photocathode rf gun, the electrons are emitted from the photocathode surface with a strong rf electric field (~100 MV/m). An electron beam with low space-charge induced emittance can be generated from the rf gun. As a typical example of a 1.6-cell rf gun developed in Brookhaven National Laboratory [6-10], the transverse normalized rms emittance of 2.3 mm-mrad at bunch charge of 1 nC was obtained by a 11 ps long Gaussian laser pulse, and the emittance was reduced to 1.2 mm-mrad by using a square laser pulse. The low emittance beam produced from the photocathode rf gun provides not only a high brightness beam with small beam spot size on the sample, but also a choice to compress the electron bunch from picosecond to femtosecond for opening the

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next pulse radiolysis on the femtosecond time scale.

However, the time resolution of pulse radiolysis is not only dependent on the electron bunch length and the analyzing light pulse width, but also determined by degradations due to velocity difference between light and the electron in the sample because of the refractive index, and time jitter between the electron bunch and the analyzing light. In this paper, we presented a new pulse radiolysis system based on a technique of Equivalent Velocity Spectroscopy (EVS) was proposed. In EVS, a femtosecond electron beam produced by a photocathode rf linear accelerator was used, and a femtosecond laser was used as the analyzing light source. A jitter compensation technique with a femtosecond streak camera was used to reduce the time jitter between the electron bunch and laser pulse. A technique of double laser pulse injection was used successfully to improve the signal to noise (S/N) ratio due to the fluctuation of the laser intensity during the measurement.

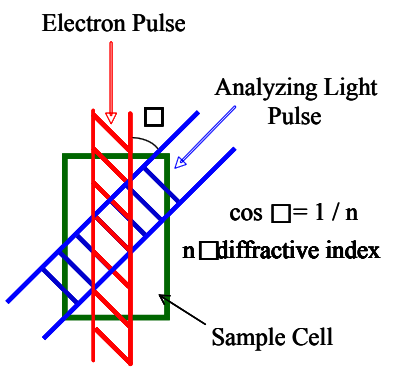
EQUIVALENT VELOCITY SPECTRO-SCOPY

Equivalent Velocity Spectroscopy (EVS) was constructed by a femtosecond single-bunch electron and a femtosecond laser light, as shown in Fig. 1. In order to avoid the degradation of the time resolution caused by the velocity difference between the light and the electron beam in sample, the electron beam and the laser light were injected into sample with an angle (θ), which is determined by the refractive index (n) of the sample. The electron bunch was also rotated with a same angle to make an overlap of the electron bunch with the laser pulse.

In EVS, the time for the light passing through the sample is given by $\Delta t_l = Ln/c$ because the velocity of the light is reduced to c/n, where L is the sample thickness (the optical path length). On the other hand, the time for the electron passing through the sample can be obtained as $L/v \cdot \cos\theta$, where v is the velocity of the electron in sample (we can assume v=c for a few tens MeV electron beam). Therefore, the degradation of the time resolution caused by the velocity difference between the light and the electron beam was calculated as

$$g(L) = Ln/c - L/v \cdot \cos\theta. \qquad (1)$$

As shown in Eq. (1), we can obtained g(L)=0 by adjusting the incident angle to $\cos\theta = 1/n$. However, the rotation of the electron bunch would be indispensable in EVS. To rotate the electron bunch, a magnetic bunch compressor, which was constructed with two 45°-bending magnets and four quadrupole magnets (two pairs) to provide a necessary path length dependence on energy, was used. The low-emittance electron beam generated from the photocathode rf gun was accelerated by a booster linear accelerator up to 32 MeV with energy-phase modulation in the bunch. The energy-phase correlation of the bunch was obtained and optimized by adjusting the accelerating rf phase (using the curvature of the rf waveform in the linac). Finally, the necessary rotation of the electron bunch was achieved by passing the electron beam through the compressor and optimizing the magnetic fields of the quadrupole magnets.



Equivalent Velocity Absorption Spectroscopy

Figure 1: Equivalent Velocity Spectroscopy

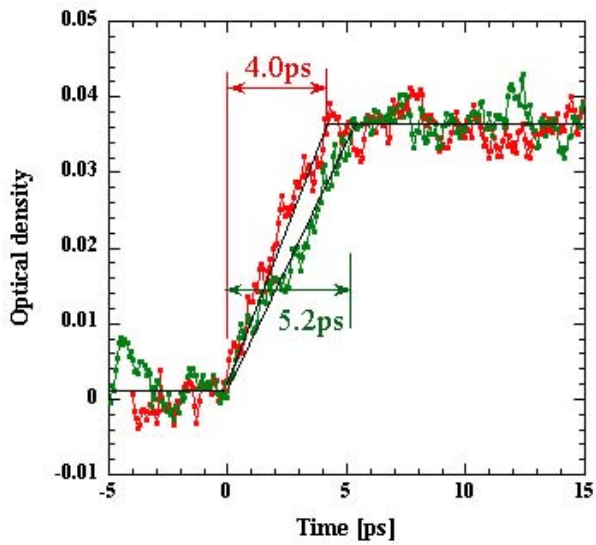


Figure 2: Bunch length vs. bunch charge. The red and green data were measured with and without the rotation of the electron bunch. The incident angle was fixed at 41° between the electron beam and the laser light.

Figure 2 gives the transient absorption kinetics of hydrated electrons measured in water at wavelength of 800 nm. The electron charge in bunch was 0.85 nC. The optical length of the sample was 1.5 mm. The time step of the optical delay was 80 fs in the measurement. The optical density at each time step was obtained by averaging 32 shots to reduce the time jitter between the electron bunch and the laser pulse. The rise time of the hydrated electron in water was observed to be 5.2 ps without the rotation of the electron bunch. It was improved to 4 ps by rotating the electron bunch in EVS. The remained time resolution was mainly limited by the time drift of the electron bunch due to the phase drift of the accelerator rf system during the electron acceleration. However, the experimental results indicate that EVS is a powerful tool to improve the time resolution of pulse radiolysis. Moreover, the optical density in EVS is

independent on the optical path length. The higher optical density can be obtained at low-charge electron beam.

JITTER COMPENSATION TECHNIQUE

In pulse radiolysis, the analyzing laser light is synchronized with the electron beam by mode-locking the laser oscillator to the accelerating rf. However, the synchronized time jitter of a few picoseconds or a few hundreds femtosecond between the electron beam and the analyzing laser light is occurred in the pulse radiolysis due to the accuracy of the mode-locked technique in the laser and the time drift between the electron beam and the laser light. To reduce the time jitter between the electron bunch and laser pulse, a jitter compensation technique based on a femtosecond streak camera was proposed in EVS.

In the compensation, both the laser light and Cherenkov radiation emitted from the electron bunch when it passes through air at the compressor exit were guided into a femtosecond streak camera (HAMAMATSU, FESCA-200, C6138). The time interval of the electron bunch and the laser pulse was measured shot-by-shot, and used as the time delay of pulse radiolysis. The time resolution of the streak camera under the measurement time region of 20 ps was 78 fs in rms. The degradation of the time resolution due to the time jitter can be expected to be improved to <100 fs in EVS.

DOUBLE-PULSE INJECTION TECHNIQUE

A technique of double laser pulse injection was used in EVS to improve the signal to noise (S/N) ratio due to the fluctuation of the laser intensity caused by long-term drift and mechanical vibration of optics at low frequencies. In EVS, a Ti:Sapphire femtosecond laser (Spectra Physics, Tsunami) was used as a probe light source. The laser pulse was time-synchronized with the electron beam by adjusting the oscillator's cavity length to phase-lock the laser output with the phase of the 79.3 MHz rf produced as the 36th sub-harmonic of the 2856MHz accelerating rf. The output energy of the laser pulse was 10 nJ. The wavelength of the laser light was 800 nm with FWHM bandwidth of 20 ns. The pulse width of the laser measured by using an autocorrelator was 80 fs in FWHM. The continuous laser pulse train of 79.3 MHz, output of the laser, was guided to a pulse selector (Spectra Physics, 3980), which was constructed with an Acousto-optic Modulator (AOM) crystal driven by the rf. Seven-pulse laser train was extracted by adjusting the pulse width of the rf. The fourth pulse of the seven-pulse laser train was used as a reference pulse, while the fifth pulse coming 12.6 ns later was used as a signal pulse. The optical density is obtained by $OD=\log(I_o/I)$, where I_o and I are the areas of the reference pulse (fourth pulse) and the signal pulse (fifth pulse), respectively.

Figures 3 gives the I_o/I ratios measured in the cases of the single-pulse injection and the double-pulse injection. The intensity of the laser pulse was measured by a Si photodiode. The data at each time step was obtained in the single-shot measurement. In the case of single-pulse injection, one pulse of the 79.3 MHz laser pulse train was used for the measurement. The value of I_o was measured without the electron beam, while the value of I was measured with the electron irradiation. In the case of the double-pulse injection, an optical delay was used to adjust the fifth pulse in the seven-pulse laser train overlapping the electron bunch. The I_o/I ratios in the both cases were measured in air without any sample in the cell. As shown in Fig. 3, the fluctuation of the I_o/I ratio was reduced from 4.7% to 1.8% (rms) by using the double-pulse injection technique. The double laser pulse injection was used successfully to measure the transient absorption kinetics of hydrated electrons in Fig. 2.

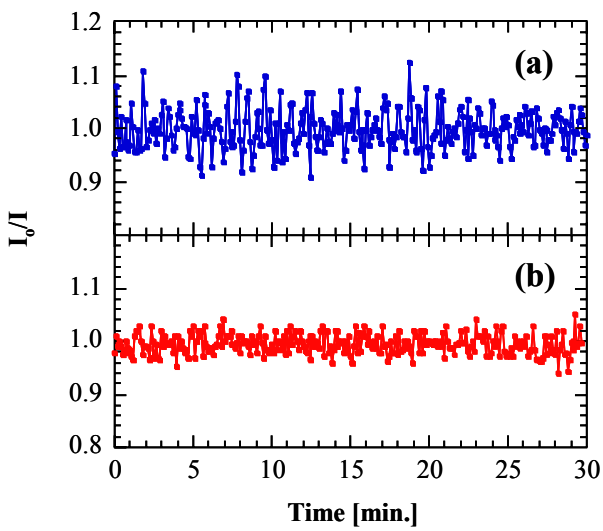


Figure 3: Fluctuations of I_o/I ratios in the cases of the single-pulse injection (a) and the double-pulse injection (b), seeing in ref. [7] for details.

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