

## AN ADVANTAGE OF THE EQUIVALENT VELOCITY SPECTROSCOPY FOR FEMTOSECOND PULSE RADIOLYSIS

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### Abstract

For studies of electron beam induced ultra-fast reaction process, femtosecond (fs) pulse radiolysis is under construction. To realize fs time resolution, fs electron and analyzing light pulses and their jitter compensation system are needed. About a 100fs electron pulse was generated by a photocathode RF gun LINAC and a magnetic pulse compressor. Synchronized Ti: Sapphire laser have a pulse width about 160fs. And, it is significant to avoid degradation of time resolution caused by velocity difference between electron and analyzing light in a sample. In the 'Equivalent velocity spectroscopy' method, incident analyzing light is slant toward electron beam with an angle associated with refractive index of sample. Then, to overlap light wave front and electron pulse shape, electron pulse shape is slanted toward the direction of travel. As a result of the equivalent velocity spectroscopy for hydrated electrons, using slanted electron pulse shape, optical absorption rise time was about 1.4ps faster than normal electron pulse shape. Thus, the 'Equivalent velocity spectroscopy' is effective for femtosecond pulse radiolysis.

### INTRODUCTION

Electron beam and extreme ultraviolet attract attention as light sources of top-down nano-processing nowadays. Chemical amplification resists are used for electron beam lithography. High energy electron beam causes ionization and generation of secondary electron in the resist. Secondary electron generates acid at the place distant from irradiated point. This effect causes unclear pattern on electron beam lithography. On the scale aimed at, this ambiguity cannot be disregarded any longer. Development and research of resists and additive agents having short diffusion length of secondary electron are necessary. For this reason, it is necessary to clarify initial process of electron beam induced reactions and phenomena.

Electron beam pulse radiolysis is a powerful method for that. In electron beam pulse radiolysis, excitation electron beam pulse generates a reaction intermediate in a sample, and then delayed analyzing light pulse carried out time re-solved spectroscopy measurement. Reaction process of a intermediate induced by electron beam can

be measured.

Femtosecond electron beam pulse radiolysis system is developed in the Institute of Scientific and Industrial Research (ISIR), Osaka University, Japan. To realize femtosecond time resolution, some important factors must be achieved.

- (1) Generation of excitation electron beam pulses which have femtosecond time duration,
- (2) Use of a femtosecond laser for analyzing light pulses,
- (3) Timing jitter compensation between femtosecond electron pulse and femtosecond analyzing laser light pulse with femtosecond accuracy[1,2],
- (4) Avoid degradation of time resolution caused by velocity difference between a high energy electron pulse and a laser light pulse in a sample.

For the generation of femtosecond electron beam pulse, a photo-cathode RF gun LINAC with magnetic pulse compressor was used. An electron beam pulse of duration about 100fs (R.M.S.) was generated [3]. For the analyzing light source, a Ti: Sapphire femtosecond laser of pulse width about 160fs was used (using pulse selector). The timing jitter between an electron beam pulse and an analyzing laser light pulse is measured and corrected by a femtosecond streak camera.

Degradation of time resolution by the velocity difference between an electron beam and an analyzing light in a sample is a very serious problem. To avoid this degradation problem, new pulse radiolysis technique: 'Equivalent velocity spectroscopy' was thought up. And then a measurement system of equivalent velocity spectroscopy was constructed.

The purpose in this study is improvement in the time resolution of electron beam pulse radiolysis system. For this purpose, establishment of the equivalent velocity spectroscopy is necessary. As a performance test, time profiles of the transient optical absorption of the hydrated electron were measured at 800nm. Advantages of equivalent velocity spectroscopy and results of the performance test are reported in this paper.

### EXPERIMENTAL

Excitation electron beam pulses were generated by a laser photo-cathode RF gun s-band LINAC at ISIR in Osaka University. This photo-cathode was made of Oxygen-Free Copper. Fourth harmonic generation (FHG: 262nm) of Nd: YLF laser (Timebandwidth) with 5ps pulse duration was injected into a photo-cathode

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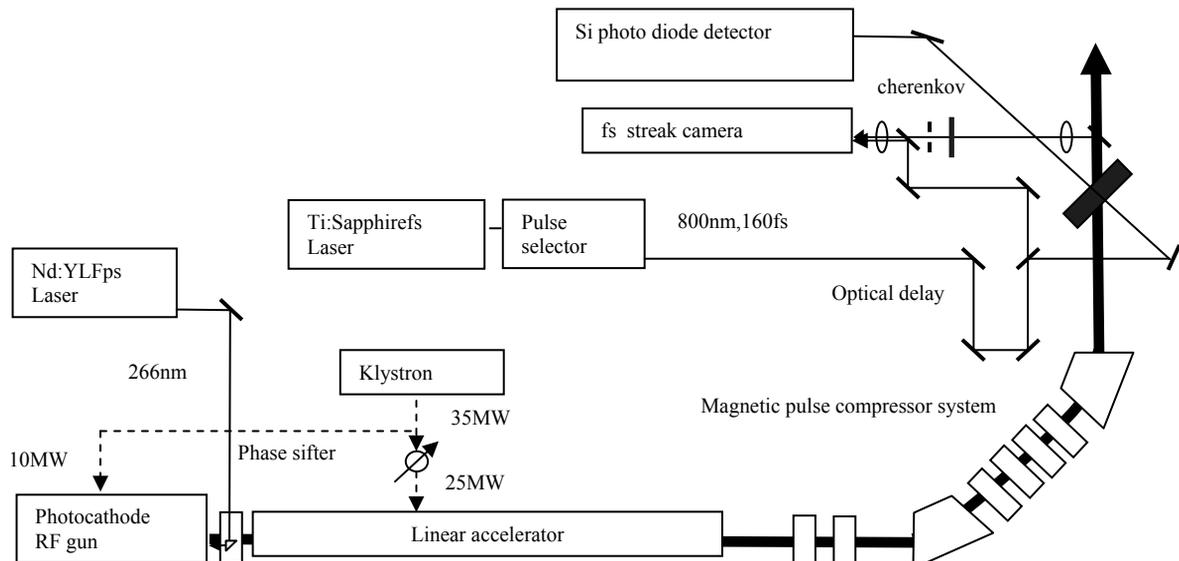


Figure 1: Experimental equipment of a new femtosecond electron pulse radiolysis system with equivalent velocity spectroscopy: photo-cathode RF gun s-band LINAC, Ti: Sapphire fs laser, fs streak camera, optical system of equivalent velocity spectroscopy

perpendicular toward a copper surface. Electron beam pulse was generated by 1.6 cell cavity which operated by 2856MHz RF. Then, this electron pulse was accelerated to about 35MeV by 2m travelling wave accelerate tube with energy modulation. Accelerated electron pulse was injected into the magnetic pulse compressor and compressed to femtosecond region finally.

Normalized transverse emittance was about 10mm-mrad at 2nC. Minimum electron pulse duration was 100fs (r.m.s.) at 100pC/pulse. Cherenkov light which was emitted in air by an electron pulse at exit of magnetic pulse compressor were measured and monitored by a femtosecond streak camera (HAMAMATSU, FESCA-200, C6138). This detail is reported other[3].

Timing synchronized Ti: Sapphire femtosecond laser (Spectra Physics, Tsunami) was used for analyzing light source. Repetition rate of a laser was 79.33MHz at 1/36 of master oscillator frequency (2856MHz). A pulse selector was used for picking out only synchronized laser pulse with 10Hz electron pulse. Analyzing laser light pulse has wavelength of 800nm and pulse duration of 160fs. Analyzing laser light was transported to the irradiation position through a tube (40m). Then, through an optical delay, analyzing light pulse was injected into the sample with incident angle of 45degree. Signal light through the sample was detected by a high speed Si photodiode.

### EQUIVALENT VELOCITY SPECTROSCOPY

Light velocity in a sample of refractive index  $n$  is  $v_L=c/n$ . The velocity of a High energy electron is nearly equal to the light velocity  $v_e \sim c$ . By the velocity difference

between an excitation electron beam pulse and an analyzing light pulse, if their incident to a sample at same time, degradation of time resolution occur.

When an electron beam pulse and an analyzing laser pulse incidence parallel in a sample, an optical length may be shortened to avoid this degradation of time resolution in a sample. For femtosecond time resolution, an optical length must be much shortened. However, if an optical length is shortened, optical density become very small, thus optical absorption can not be measured.

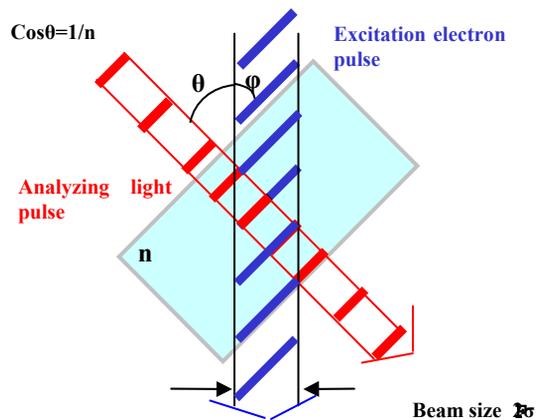


Figure 2: Concept of Equivalent velocity spectroscopy  
Refractive index of a sample :  $n$ , Incident angle :  $\theta$ , Slant angle of electron beam pulse :  $\phi$

To avoid this crucial problem, we thought up the equivalent velocity spectroscopy. Fig.2 is the concept figure of the equivalent velocity spectroscopy. The velocity component of the electron beam toward the analyzing light propagation direction and the velocity of

light in a sample are made to become equal. Thus, if the refractive index of a sample is  $n$ , an incident angle  $\theta$  between the electron beam and analyzing laser light is expressed as  $\theta = \arccos(1/n)$ . Furthermore, the shape of the electron beam pulse is controlled so that an analyzing light pulse and an electron pulse overlap in a sample. Degradation of time resolution is avoided by above thing.

Moreover an optical pass length in the equivalent velocity spectroscopy was decided by the crossing region of an electron beam and an analyzing light. Beam size is new important factor, optical pass length independent on a sample cell length. By the equivalent velocity spectroscopy, degradation of time resolution is avoided, and optical absorption measurement is possible.

### RESULTS AND DISCUSSIONS

The transient optical absorption of the hydrated electron was measured at 800nm for performance evaluation of the equivalent velocity spectroscopy. A sample cell length was 2mm. Excitation electron beam size was  $\sigma=0.7\text{mm}$ . Charge of a pulse was 0.85nC. Electron beam Energy was about 35MeV.

The Results is shown in Fig.3. Triangle points in Fig.3 show a time profile of an optical absorption by ordinary slanted incident case. Rise time of optical absorption by the hydrated electron was estimated about 5.5ps. For the equivalent velocity spectroscopy, we tried to control the pulse shape of an electron beam pulse. Square points in Fig.3 show a time profile of an optical absorption by equivalent velocity spectroscopy. The rise time of this case was estimated about 4.0ps. The rise time of the optical absorption by the equivalent velocity spectroscopy

is 1.5ps faster than the case of ordinary slanted incident.

This result shows the improvement of time resolution by the equivalent velocity spectroscopy. By the equivalent velocity spectroscopy, the measurement at faster time region will become possible.

Based on the equation from simple geometry consideration, the wave front of an electron pulse was estimated about  $\phi=70\text{degree}$ . However, this value has little credibility. Experimental data of a controlled electron pulse shape is not obtained yet. It is necessary that an accurate method of measuring the shape of electron pulse is developed.

### CONCLUSIONS

A new technique of a pulse radiolysis measurement was proposed as the equivalent velocity spectroscopy. For the equivalent velocity spectroscopy, analyzing light pulse is slanted and incident on a sample with the angle determined by the refraction index of a sample toward electron beam. Furthermore, the shape of the electron beam pulse is controlled so that the analyzing light pulse and the electron pulse overlap in a sample. The equivalent velocity spectroscopy is a new powerful method which can measure optical absorption and avoid degradation of time resolution in a sample. Rise times of optical absorption of the hydrated electron were measured for performance test. The rise time of the optical absorption by the equivalent velocity spectroscopy is 1.5ps faster than the case of ordinary slanted incident. To investigate the electron beam induced ultra fast phenomena in femtosecond region, the equivalent velocity spectroscopy is need.

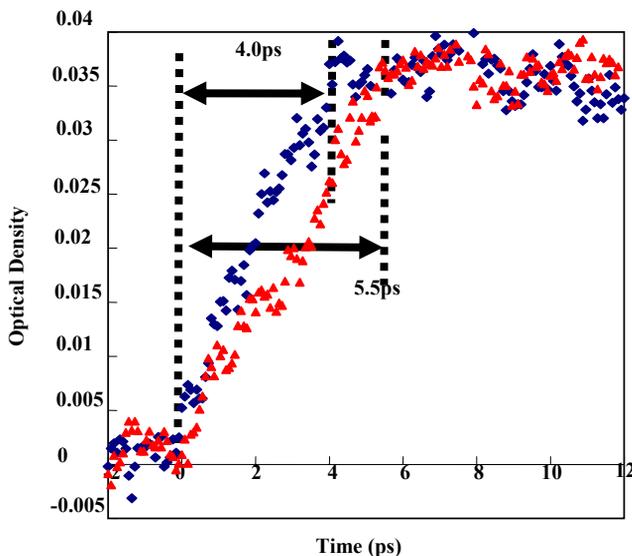


Figure 3: Time profile of optical absorption by the hydrated electron, Sample cell length: 2mm, Wavelength of analyzing light:800nm, Electron beam size:  $\sigma=0.7\text{mm}$ , Charge:0.85nC/pulse, Energy:35MeV

- ( $\blacktriangle$ ) Normal electron beam pulse shape
- ( $\blacklozenge$ ) Try to control electron beam pulse shape

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