5th International Particle Accelerator Conference IPAC2014, Dresden, Germany JACoW Publishing
ISBN: 978-3-95450-132-8 doi:10.18429/JACoW-IPAC2014-MOPRI036

PULSE RADIOLYSIS USING TERAHERTZ PROBE PULSES

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Abstract

Pulse radiolysis, which utilizes a pump electron beam and a probe pulse, is a powerful tool that can be used for the time-resolved observation of ultrafast radiation-induced phenomena. Recently, double-decker pulse radiolysis using visible probe pulses were demonstrated based on a photocathode RF gun driven by two UV pulses, which enabled synchronized pump electron beam and visible probe pulses. In this study, pulse radiolysis using terahertz (THz) probe pulses which were realized by the “double-decker” electron beams and dynamics of transient quasi-free electrons in semiconductors are presented.

INTRODUCTION

Short electron bunches with durations of picoseconds to femtoseconds are key elements for the development of high-quality and intense light sources for applications in accelerator physics such as free-electron lasers [1,2] and laser-Compton X-rays [3,4]. Such electron bunches can be also applied to time-resolved pump-probe experiments involving the application of techniques such as ultrafast electron diffraction [5] and pulse radiolysis [6-9] for improving the time resolution of each system. Pulse radiolysis, which utilizes a pump electron beam and a probe pulse, is a powerful tool that can be used for the observation of ultrafast radiation-induced phenomena that are studied in physics, chemistry, and biology. Pulse radiolysis based on stroboscopic method generally utilizes optical probe pulses from a laser. Recently, double-decker pulse radiolysis, which does not utilize a laser for probe pulses, has been demonstrated by double-decker electron beams. The double-decker electron beams were generated by 2 separated ultraviolet (UV) pulses for a photocathode electron gun [9,10] driven by high-power radio-frequency (RF). One of the beams was converted into an optical probe pulse by Cherenkov radiation in the air, whereas the other was used as a pump electron beam. From the viewpoint of electrodynamics, Pulsed electron beams are also applied to electro-magnetic radiation production in terahertz (THz) range because of the inverse of 1 ps corresponding to the frequency of 1 THz. Schemes of THz generation using electron beams are investigated in coherent transition radiation (CTR) [11-14], Smith-Purcell radiation [15], and coherent Cherenkov radiation [16,17] for several applications, e.g., beam diagnoses, pump sources, and probe sources. Recently, THz generation based on a modulation of electron beam are investigated to obtain narrowband THz pulses [18,19]. The combination of pulse radiolysis technique and THz generation based on electron beams would expand the wavelength of probe pulses and observation of transient species different from the case using optical probe pulses from lasers.

In this study, pulse radiolysis using pump electron beams and probe THz pulses based on the double-decker electron beams was investigated. Transient spectral transmittance of a sample was induced by the pump electron beams. Time- and frequency-resolved THz transmittance in the sample was observed by utilizing a probe delay and Michelson interferometer.

EXPERIMENTAL SETUP

Generation of Femtosecond Double-Decker Electron Beams

Figure 1 shows the generation of the femtosecond double-decker electron beams based on the photocathode RF gun linac. The linac consisted of a photocathode RF gun, linac, and a magnetic bunch compressor. A Nd:YLF picosecond laser was used for driving a photocathode with a copper cathode. The output of the picosecond laser was 180 μJ/pulse of UV pulse (262 nm) with a pulse width of 5 ps at 10 Hz. The UV pulse was separated by a beam splitter (BS1). The separated 2 UV pulses were injected on the photocathode with spatial and time separations adjusted by an optical delay (OD1) and mirrors with actuators. The double-decker electron beams, i.e., precisely synchronized 2 electron beams, were accelerated in the gun and linac up to 32 MeV at 10 Hz because of the repetition rate of klystron feeding RF to the gun and the linac. Finally, the accelerated electron beams were compressed to femtosecond by a magnetic bunch compressor to improve the time resolution in pulse radiolysis and obtain broadband THz probe pulses. The bunch compressor is composed of bending, quadrupole, and sextupole magnets. In the present case, the spatial and temporal separations of the UV pulses were ~3 mm and 2.1 ns, respectively, on the cathode. Bunch charges of front and back electron beams were 180 and 120 pC/pulse, respectively. Bunch length was estimated as <200 fs in rms according to the measurement using an interferometer based on a filtered model [13].

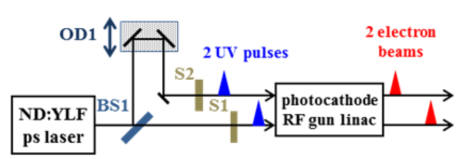

Figure 1: Generation of femtosecond double-decker electron beams. BS: a beam splitter; OD: an optical delay line; S: a shutter.

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Pulse Radiolysis Using THz Probe Pulses

Schematic diagram and pictures of pulse radiolysis using THz probe pulses are shown in Fig 2. In pulse radiolysis, change of transient spectral transmittance of high-resistivity silicon (HRSi) induced by irradiation of the pump electron beam was investigated. The thickness of the sample was 380 µm. The front electron beam (L) of the double-decker beams was converted into a probe THz pulse by CTR on a plane mirror (M1) as shown in Fig. 2 (a). The probe pulse was collimated by an off-axis parabolic mirror (OAP1) with a focal length of 191 mm from a viewpoint of opening angle of CTR at the plane mirror. The probe THz pulse traveled through an optical delay (OD2) to enable time-resolved measurement. The back electron beam (B), which was used as a pump electron beam, reached the sample with a delay of 2.1 ns with respect to the front beam. The probe pulse was injected on the sample after travelling a longer path than the back electron beam. The spatial separation between the beams was set to 6 mm in the vertical direction to avoid the beam used as the probe pulse from penetrating through medium such as the mirrors and sample which scatters electron beams. The probe pulse was focused in the sample by OAP2 and collimated again by OAP3 to be transported to a Michelson interferometer [14,17, 20] which enabled frequency-resolved measurement, i.e., spectral transmittance. In the interferometer, THz probe pulse was separated by a beam splitter (BS2) made of HRSi. One of the THz pulse was reflected by a moving mirror (OD3), whereas the other was reflected by a fixed mirror (M4). Finally, the separated THz pulses were superposed at a liquid-helium-cooled silicon bolometer (general-purpose 4.2-K system, Infrared Laboratories). The frequency spectrum of THz pulse was calculated by the Fourier transform (FT) of an interferogram, which was the bolometer output as a function of the moving mirror (OD3) position. These optical delays were used for the measurement of transient spectral transmittance of HRSi induced by the back electron beam. The electron beams were introduced to the pulse radiolysis system in a lower vacuum chamber after passing through titanium foil (with a 20-µm thickness), which separated the high-vacuum beam line from the system as shown in Fig. 2 (b). The electron beams spatially separated in the vertical direction travelled near the sample as shown in Fig. 2 (c).

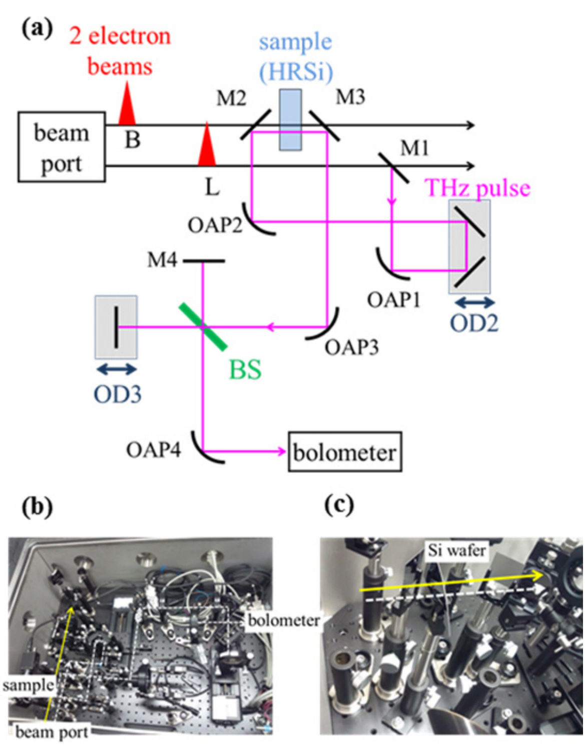

Figure 2: (a) Pulse radiolysis using THz probe pulses. Beams of "L" and "B" were used for generation of THz probe pulses and irradiation of a sample, respectively. OAP: an off-axis parabolic mirror, M denotes a plane mirror, and BS denotes a beam splitter. (b) Picture of a low-vacuum chamber. Lines denote paths of beams and probes. (c) Picture of the sample irradiated by the pump beam (B, solid line) and path of the beam for the probe-generation (L, dashed line).

RESULTS AND DISCUSSION

Figure 3 shows the bolometer outputs as a function of the optical delay (OD2) in the pulse radiolysis. In this case, the optical delay in the interferometer (OD3) was fixed at the position of 4 ps away from the centerburst. The data corresponds to the total intensity of detected THz probe pulses. Data for *L* and *B* denote the outputs for usage of only "probe Light" generated by CTR and only "Beam" from the pump source, respectively. Data for *LB* denote the output for the combination of *L* and *B*. Each data set corresponds to the conditions of the shutters as shown in Fig. 1. Values obtained by averaged data of 5 single-scans of the optical delay are shown. Only when the probe pulse went through the sample of silicon following the pump beam, transmittance decreased due to quasi-free electrons induced by the irradiation of the pump beam. The quasi-free electrons would enhance dielectric loss resulting in absorption of THz probe pulse. As a result, the plot for *LB* decreased at a time of ~20 ps.

Figure 4 shows the time- and frequency-resolved measurement in THz region on silicon. Transient spectral transmittance could be analyzed by the Michelson interferometer with and without the irradiation of the pump beam. Figure 4(a) and 4(b) show interferograms near the centerburst in the cases of 0 and 50 ps of OD2, respectively. Each data set corresponds to the condition of electron beam. Decrease in the intensity of an interferogram of *LB* was observed when the probe pulse went through the sample after the irradiation of the pump beam, i.e., OD2= 50 ps, where transient transmittance decreased as expected from Fig. 3. According to the linearity of the FT, transient spectral transmittance of *T* can be expressed as follows:

$$I = \mathcal{F}[LB - B], \quad (1)$$

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$$I_0 = \mathcal{F}[L], \quad (2)$$

$$T = \frac{I}{I_0}, \quad (3)$$

where I and I_0 denote the frequency spectra with the irradiation of the pump beam and that without the irradiation, respectively. $\mathcal{F}$ denotes the FT of time-domain data. In this present case, frequency spectra with and without the irradiation using Eq. (1) and (2) were calculated. When THz probe pulses travelled through the sample before the irradiation, the frequency spectra of I and I_0 do not have much difference [Fig. 4 (c)]. Conversely, the frequency spectra have difference because of quasi-free electrons induced by the irradiation of the pump beam [Fig. 4 (d)]. Time- and frequency-resolved measurement in THz region was indicated based on the double-decker electron beams. Pulse radiolysis using optical pulses are used for the observation of transient species such as hydrated electrons and cation radicals [7,9]. In this case, analysis using the Drude model, which treats imaginary part of refractive index,

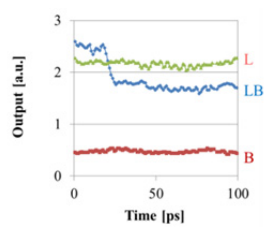

Figure 3: Bolometer output as a function of the optical delay for the probe (OD2). *LB*, *L*, and *B* denotes the shutters (S1/S2) conditions for front/back electron beams of Opened/Opened, Opened/Closed, and Closed/Opened, respectively. The transient decrease in transmittance was observed at a time of ~20 ps.

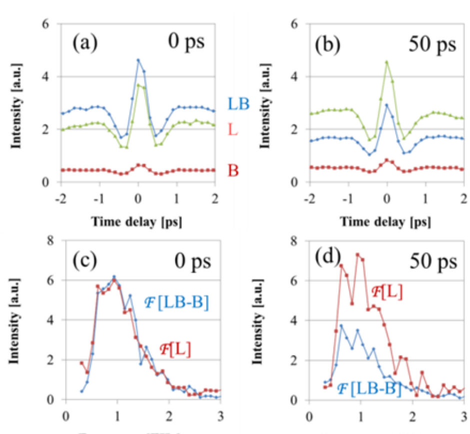

Figure 4: Time- and frequency-resolved measurement on silicon. Interferograms in the cases of (a) 0 and (b) 50 ps of OD2 are shown. Frequency spectra of I and I_0 in the cases of (c) 0 and (d) 50 ps of OD2 are shown.

would be useful for the transient spectral transmittance. Measurement at a frequency of ~0.5 to ~2 THz would be possible due to the intensity of probe pulse in this system.

CONCLUSION

Pulse radiolysis using pump electron beams and probe THz pulses based on the double-decker electron beams of was investigated. Electron beams with charges of 180 and 120 pC/pulse were used at 32 MeV. Transient spectral transmittance induced by quasi-free electrons was measured by the system using Michelson interferometer. In the present case, measurement at a frequency of 0.5 to 2 THz would be possible.

ACKNOWLEDGMENT

We thank the staff of the research laboratory for quantum beam science at the Institute of Scientific and Industrial Research, Osaka University, for the experimental setup. This work was supported by KAKENHI (21226022, 23109507, and 25870404).

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