FABRICATION OF HUNDREDS- AND TENS-FEMTOSECOND PUMP-AND-PROBE ANALYSIS

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Abstract

Femtosecond ultrafast quantum phenomena research facility was constructed at Nuclear Engineering Research Laboratory (NERL), University of Tokyo, in 1999. 100 fs 0.3 TW laser system can be synchronized with a femtosecond electron linear accelerator (linac) with the total timing jitter of 330 fs. Using pump-and-probe scheme, the system is now applied to the subpicosecond pulse radiolysis for radiation chemistry of pure and super-critical waters. 50 fs 12 TW laser system is utilized for a time resolved x-ray diffraction and a plasma cathode that is supposed to generate ~10 fs electron pulse.

1 NUCLEAR ENGINEERING RESEARCH LABORATORY (NERL)

Since an electron linac at NERL generated 10 ps single pulse in 1977, the facility has focused on the generation, the diagnostics and the control of the ultrashort single electron pulse, and its application [1-3]. In 1995, a single electron pulse of 700 fs duration was produced by the 35 MeV linear accelerator which consists of a thermionic electron gun, a sub-harmonic buncher (SHB), two accelerating tubes and an arc-type magnetic pulse compressor. After that in 1997, a laser photocathode rf-gun and a chicane-type magnetic pulse compressor were introduced to the 18 MeV linac then the single electron pulse was compressed to be as short as 240 fs. Further, in 1999, 100 fs 0.3 TW and 50 fs 12 TW Ti:Sapphire lasers, which were constructed by B.M. Industries, were installed to the facility. Using such ultrashort pulses from these four apparatus, it is now possible to perform ultrafast pump-and-probe experiments, such as pico- and subpicosecond pulse radiolysis, a time-resolved x-ray diffraction and a generation of 10 fs electron pulse via plasma cathode. The schematic view of the facility is illustrated in Fig. 1.

The main feature of the 100 fs 0.3 TW laser is the oscillator (Mira900, Coherent Inc.), which utilizes the technique of Kerr lens modelocking. The optical cavity is specifically designed to utilize changes in the spatial profile of the beam produced by self-focusing from the optical Kerr effect in the Ti:Sapphire crystal. The cavity length is controlled by the timing stabilizer, which monitors the laser output and an external frequency source coming from the linear accelerator. After passing through a regenerative amplifier, a multipass amplifier, 50 m transport line, and an optical compressor, the laser is split by two. One of them generates third harmonics (265 nm) and it is irradiated to a laser photocathode rf-gun [4]. The other is used as a probe light. Then the laser system can be synchronized with the linac in the femtosecond regime as will be represented below.

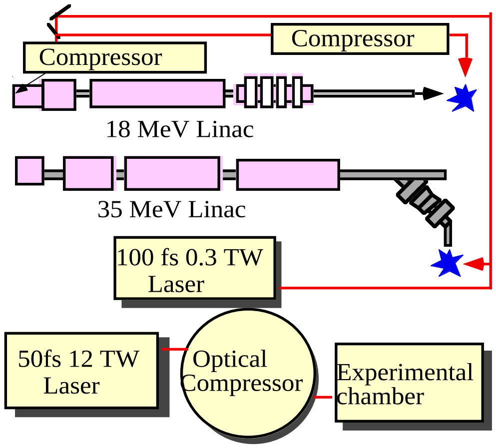

Fig. 1: Schematic view of the facility

50 fs 12 TW laser consists of the 20 fs laser oscillator (FemtoSource20, FEMTOLASERS) and a chirped pulse amplification system with an amplification factor of about 10^9. The system provides a very high peak power and stable laser pulse with relatively easy operation. The oscillator of the 50 fs 12 TW laser also utilizes the Kerr lens modelocking and the timing stabilizer. However the laser is usually used without the synchronization with the linac. The main purpose is to make plasma in a gas and generate femtosecond electron, ion THz radiation pulses via the wavebreaking of the plasma oscillation and the wakefield acceleration.

All apparatus are driven by the rfs that are higher- or sub-harmonics of the master clock 119 MHz.

2 SYNCHRONIZATION

The time resolution of the pump-and-probe system is decided by the durations of pump and probe pulses, and the timing jitter between them. In the case that the femtosecond electron and laser pulses are used as a pump and a probe pulses, the durations of them are much smaller that the timing jitter. Hence we have estimated and decreased the timing jitter for the sake of higher time resolution.

The total timing jitter is composed of those coming from the electron linac and the laser system. It is known that there are several major contributions to the timing jitter of each apparatus (see Table.1). In the case of the electron linac, the fluctuations of the voltage and the phase of the rf in the klystron cause most of the total jitter [5], even though the photoelectrons or thermal electrons are generated with some timing difference in a gun. We had used two klystrons of 6 MW power and two accelerating structures were supplied with rf from each klystron. It is supposed that our previous timing jitter of ~3 ps (rms) resulted from the fluctuations of each klystrons and mutual jitter between them. Hence a new klystron of 15 MW that can supply all accelerating structures with rf are expected to decrease the timing jitter. Now the fluctuations of the power and the phase are 0.5 % (rms) and 0.2 deg (rms), respectively. The numerical simulation using PARMELA code represents that the timing jitter from the electron linac is 300 fs. While in the case of the laser, almost all jitters originate in the vibration of the mirror. In particular, the vibration of the cavity in the laser oscillator cannot be neglected, since the slight change of the cavity has a large effect on the timing of the laser. Therefore the modelocking and the timing stabilizer are the most important factors for the lower jitter system. The Kerr lens modelocking, or passive modelocking, intrinsically generates lower jitter rather than the positive modelocking. Further the timing stabilizer that monitors 9th harmonics of the rf and the laser output has a good influence on the stability of the cavity length, since the fluctuations of the voltage and the phase of the rf can be separated much easily in the higher harmonics rather than in the fundamental [6]. We roughly make an estimate of the timing jitter from the oscillator around 100 fs as an ideal value, considering the achievement of other facilities [7]. Finally it seems that the time resolution of our system is $\left(300^2+100^2\right)^{1/2}=320\,[fs]$.

Table 1: Timing jitter of each component

Main Contributions	**estimation of jitter** (rms)
Klystrons	300 fs
Mutual between KLYs	negligible
Laser oscillator	100 fs
Mutual between lasers	none
Other components in the laser system	negligible

When we measure the total timing jitter between the electron linac and the laser system, we observe the variation of the time differences between electron and laser pulses by the femtosecond streak camera (FESCA-200, Hamamatsu Photonics Co., Ltd.). Figure 2 indicates a series of the variation of the time difference. In the previous case [cf. Fig. 2(a)], where the laser oscillator was actively modelocked, the fluctuation of the timing interval is in the picosecond regime and accordingly the timing jitter was 3.5 ps. In the present case [cf. Fig. 2(c)], on the other hand, the timing intervals fluctuate in the fraction of picosecond. The jitter 1.9 ps was affected not by the actual timing jitter but by the drift, which is caused by a shift of the temperature of the accelerating cavity. The shift of 1 ˚C corresponded to ~10 degree of the rf phase. The jitter separated from the drift in Fig.2(c) is 330 fs, which shows good agreement with the estimated value 320 fs. After we obtained the results that are shown in Fig.3, we replaced the water cooling device of which stability is within 0.01 ˚C. We will measure the total timing jitter in near future. You note that the 9th harmonics lock [cf. Fig. 2(c)] makes the timing more stable than the fundamental lock [cf. Fig. 2(b)].

When only the laser is used as a source of pump and probe lights, the synchronization is much better than that in the case that the electron linacs is synchronized with the laser. In the case, the time resolution of the pump-and-probe experiment is in the order of the pulse duration, which is usually less than 100 fs.

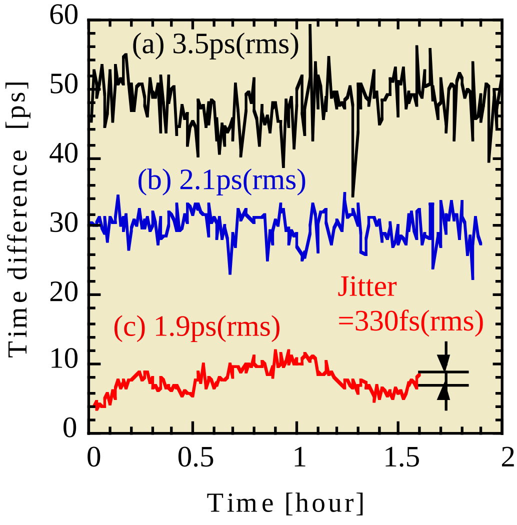

Fig. 2: Series of the time differences between the electron pulse and the laser pulse

3 APPLICATIONS

3.1 Pulse radiolysis

In the field of radiation chemistry, there are two main pump-and-probe analysis methods; a laser photolysis and a pulse radiolysis. The former utilizes only laser then the highest time resolution is now a few femtosecond or less. The latter is composed of the laser and the accelerator and then the time resolution is now at the border between picosecond and femtosecond. In our facility, the pulse radiolysis system, where 100 fs 0.3 TW laser are synchronized with the 18 MeV linac, has been developed [8]. The time resolution of the pulse radiolysis system depends not only on the timing jitter of the system but also on the characteristics of the experimental setup, such as a thickness of the target. The experimental result is indicated in Fig. 34. In the experiment, the electron pulse of the 7 ps duration (pump) excited a transitional chemical reaction and 100 fs laser pulse were monitored as a probe light. Pure water and 1 M $HClO_4$ were put into the cell as a sample. The thickness of the cell was 18 mm, which leaded the deterioration of the time resolution. Because the velocity of the electron and that of the photon in a water cell are different. It is known that the rise time of the reaction is less than 1 ps, while the experimental result showed 30 ps. Consequently, the time resolution of the pulse radiolysis system is estimated to be 30 ps, which is appropriate value considering the pulse durations and the thickness of the cell.

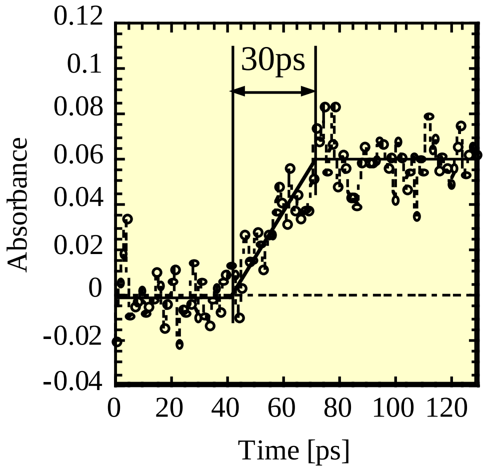

Fig. 3: Experimental result of pulse radiolysis

3.2 *Time resolved x-ray diffraction*

In recent years, there has been increasing interests in the use of ultrashort x-ray pulses. In the facility, 50 fs 12 TW laser is irradiated to the solid target to generate a picosecond laser plasma x-ray pulse for the time-resolved x-ray diffraction experiment. A part of the laser pulse is split before the irradiation as a pump and excites the lattice vibration on the surface of the crystal. The time resolution of the experiment is ~10 ps, since the duration of probe pulse are ~10 ps that are much longer than the duration of the pump pulse and the timing jitter. Static images of the x-ray diffraction were taken as shown in Fig. 4. The pump-and-probe experiment is going on.

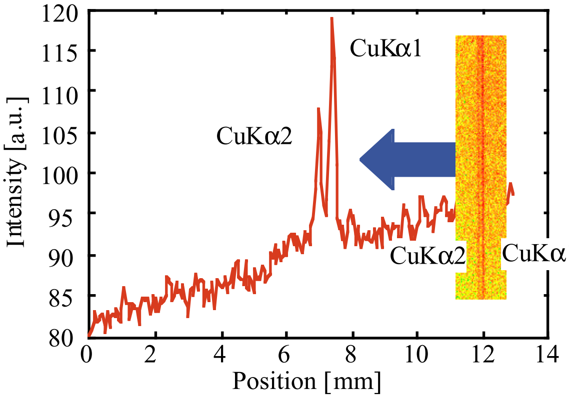

Fig. 4: Static image of laser plasma x-ray diffracted by GaAs (111)

3.2 *Laser Plasma cathode*

Laser plasma cathode is based on the idea of nonlinear wake wavebreaking and is supposed to generate 10 fs relativistic electron pulse. 50 fs 12 TW laser introduced to the gas jet excites the plasma oscillation in the gas then a number of electrons get on the accelerating phase of the wakefield via wavebreaking of the plasma. There are several advantages of the technique, such as the generation of extremely short pulse, the small timing jitter, and the compactness of the apparatus. The numerical results indicate that it is possible to generate ~10 fs electron pulse (see Fig. 5) [9]. The experiment is now under way.

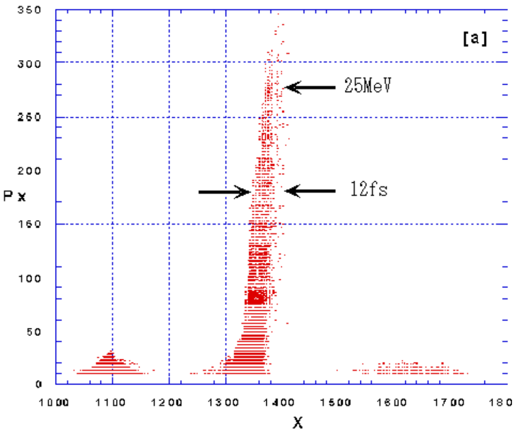

Fig. 5: Simulated result of the generation of ~10 fs electron pulse

4 SUMMARY

In the facility, four ultrafast pulses, two femtosecond electron pulses and two femtosecond TW laser pulses, are now available and they are precisely synchronized with one another. Several application experiments of ultrafast pump-and-probe analysis are in progress making use of the four ultrashort pulses. We intend to observe ultrafast quantum phenomena from viewpoints both of the chemistry and the physics then visualize their appearances using computer graphics.

REFERENCES

[1] M. Uesaka et al., Phys. Rev. E 50 (1994) 3068.
[2] M. Uesaka, T. Watanabe et al., Proc. of the 6th European Particle Accelerator Conference (EPAC98), Stockholm, Sweden (1998) 776.
[3] T. Watanabe et al., Nucl. Instrum. Meth. A 437 (1999) 1.
[4] T. Kobayashi et al, in the proceedings.
[5] A. Takeshita, et al., Nucl. Instrum. Meth. A 421 (1999) 43.
[6] D. von der Linde, Applied Physics B 39 (1986) 201.
[7] For example, G.M.H. Knippels et al, Optics Lett., 23 (1998) 1754., private communication with K. Kabayashi, FESTA, Japan.
[8] Y. Muroya et al., Proc. of Trombay Symposium on Radiation and Photochemistry (2000) 1.
[9] H. Nasr et al., Nucl. Instrum. Meth. A, in press.