DEVELOPMENT OF FREQUENCY-RESOLVED OPTICAL GATING FOR MEASUREMENT OF CORRELATION BETWEEN TIME AND FREQUENCY OF CHIRPED FEL

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Abstract

A femtosecond infrared-chirped free-electron laser (FEL) is an effective tool of dissociating molecules without the intramolecular vibrational redistribution. The ultrashort FEL pulse with the broadband spectrum is achieved operating the long-pulse electron beam from an energy recovry linac at Japan Atomic Energy Agency. Until now, the broadband spectrum of ultrashort pulse was measured to be $\Delta\omega\ /\omega_0$ = 14% with the central wavelength of 23 μm and the pulse duration of 320 fs at FWHM by an autocorrelation of fringe-resolved second harmonic generation. However the information of pulse shape and variation of frequency depending on the time during the pulse were not obtained. Since it is essential to know both information in the pulse for the dissociation of the molecule, we will measure them by frequency-resolved optical gating (FROG).

INTRODUCTION

A femtosecond infrared-chirped free-electron laser (FEL) is an effective tool of dissociating polyatomic molecules without the intramolecular vibrational redistribution [1,2]. Coherent vibrational climbing proceeds from exciting a molecular vibration transitions by the ultrashort infrared pulse with the broadband spectrum which encompasses several vibrational transitions of the molecule. Indeed, these transition energies become smaller and smaller with climbing the ladder because of molecular anharmonicity. Therefore, the climbing efficiency can be increased dramatically when using a negatively chirped pulse so that its high-frequency components which resonant with the lower transitions of the ladder precede its low-frequency components which resonant with the upper transitions of the ladder. Coherent vibrational climbing can be viewed also as a rapid adiabatic passage leading to efficient excitation of the upper vibrational states with an efficiency that in theory can be close to 100% because of the coherent nature of the interaction.

At Japan Atomic Energy Agency (JAEA), an energy recovery linac (ERL) driven by superconducting accelerators has been constructed to produce a high-power far-infrared FEL(~20μm) [3,4]. Operating this device, it succeeds in generating ultrashort pulses with the broadband spectrum using a long macro pulse of electron beam by JAEA-ERL.

Until now, the broadband spectrum was measured to be $\Delta\omega\ /\omega_0$ = 14% at the central wavelength of 23 μm and the pulse width of 320 fs at FWHM by an autocorrelation of fringe-resolved second harmonic generation (FRSHG) [3]. However the intensity and the variation of frequency depending on the time during the FEL pulse were not obtained. Since it is essential to know both of the intensity and the variation of frequency in the pulse for the dissociation of the molecule, we will measure them by frequency-resolved optical gating (FROG) [5].

FREQUENCY-RESOLVED OPTICAL GATING

Principle of FROG

FROG is a technique to completely determine the intensity and phase versus time or frequency. The apparatus of FROG is only an autocorrelator followed by a spectrometer. In the autocorrelation and related techniques, the ultrashort pulse is measured purely in the time domain (autocorrelator), or in the frequency domain (spectrometer). In all these measurements, detectors can only measure the intensity of the signal. As a result, it is inevitable to lose the phase information, if the measurement is taken only in one domain. The measurement of FROG trace is taken in a hybrid domain: time-frequency domain. As time and frequency are two reciprocal domains connected by Fourier transform, the phase information in time domain is encoded into the intensity information in frequency domain, and vise versa. Therefore FROG trace contains the information of both intensity and phase of ultrashort FEL pulse by doing only intensity measurement in time-frequency domain.

As the time-dependent component of the pulse can be written in

$$E(t) = \mathrm{Re}\left\{\sqrt{I(t)}\exp(i\omega_0 t - i\varphi(t))\right\}, \qquad (1)$$

where $I(t)$ and $\varphi(t)$ are the time-dependent intensity and the variation of frequency, and ω_0 is a carrier frequency, FROG trace is described as

$$I_{FROG}(\omega,\tau) = \left|\int_{-\infty}^{\infty} E_{sig}(t,\tau)\exp(-i\omega t)dt\right|^2, \qquad (2)$$

where a quantity $E_{sig}(t,\tau)$ is a signal field defined by $E(t)g(t\text{-}\tau)$. The function $g(t\text{-}\tau)$ is a gate function with respect to the gate delay τ. Now, consider the Fourier transform of $E_{sig}(t,\tau)$ with respect to τ, Eq. (2) is transformed to

$$I_{FROG}(\omega,\tau)=\left|\int_{-\infty}^{\infty}\hat{E}_{sig}(t,\Omega)\exp(-i\omega t-i\Omega\tau)dtd\Omega\right|^2 . \quad (3)$$

Since Eq. (3) is the 2-dimensional Fourier transform with respect to t and Ω, the signal field $E_{sig}(t)$ implementing I_{FROG} is determined uniquely due to the fundamental theorem of algebra. As a result, the information of $I(t)$ and $\varphi(t)$ in Eq. (1) are obtained from the quantity $I_{FROG}(\omega,\tau)$.

Various non-linearities such as second-harmonic generation (SHG), third-harmonic generation, self-diffraction and polarized-gate are well known as the gate function [6]. In our measurement, SHG autocorrelation is used as the gate for FROG trace.

Simulation of SHG-FROG

In the SHG-FROG geometry, the pulse is split into two pulses which are then spatially overlapped in a piece of frequency-doubling crystal. In the frequency-doubling crystal, the two pulses induce a second harmonic due to the second-order optical non-linearity. Hence the signal field for SHG-FROG is given by $E_{sig}(t,\tau) = E(t)E(t-\tau)$. The main advantage of SHG-FROG is sensitivity: it involves only the second-order nonlinearity. Consequently, for a given amount of input pulse energy, SHG-FROG will yield more signal pulse energy.

In order to evaluate retrieval of the intensity $I(t)$ and phase $\varphi(t)$ of the ultrashort pulse from SHG-FROG trace, a numerical simulation for SHG-FROG was carried out using the JAEA-FEL parameters. As a temporal profile of input intensity, the previous simulation result was used [7]. The carrier frequency ω_0 was determined by an experimental measurement of λ_0 = 21μm. Three types of input variation of frequency were assumed for the simulation.

The simulation results are shown in Fig. 1, in which the top row shows false-color FROG trace for each phase; red means high intensity and violet means low intensity. The images of FROG trace in Fig. 1 are cropped, but the simulation was achieved in double range. The bottom row shows retrieval results of FEL pulse, where red closed (open) circle indicates the retrieval intensity (phase) respectively, and blue lines mean the input shape of FEL pulse. The each column indicates the difference of the

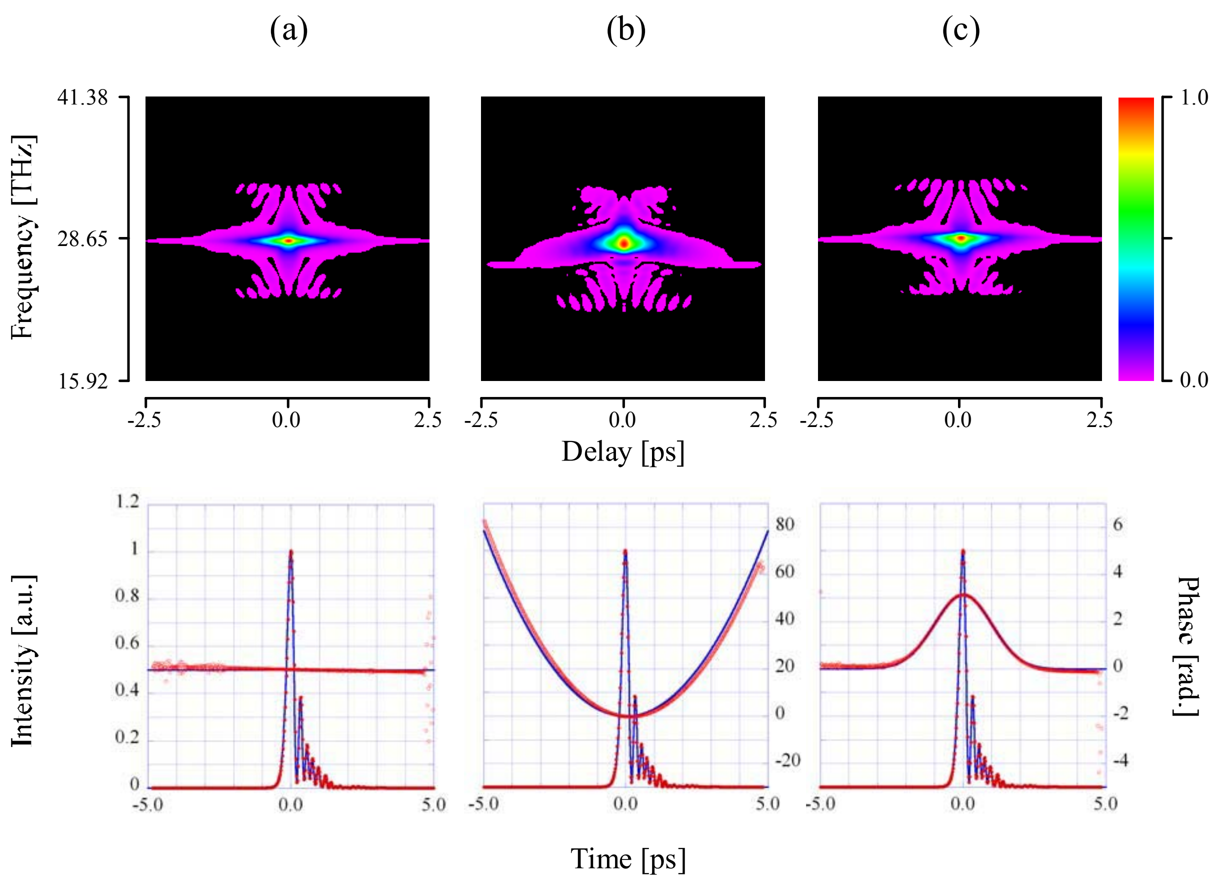

Figure 1: The top row shows false-color FROG trace for each phase; red means high intensity and purple means low intensity. The bottom row shows the retrieval result, where red closed (open) circle indicates the retrieval intensity (phase) and blue line indicates the initial parameter. The each column (a), (b), (c) indicates Fourier transformer-limited, negative chirp, self-phase modulated pulse respectively.

variation of frequency; (a) is a Fourier transformer-limited, (b) is a negative chirp, and (c) is a self-phase modulated pulse respectively. The retrieval intensity and phase were good agreement with the input intensity and phase.

MEASUERMENT SETUP

Focus and transport system

Since the FEL pulse is diverged by optical diffraction due to a ϕ2-mm pinhole on an output coupler mirror of FEL optical cavity, a pair of Au-coated elliptical mirrors is used to parallelize the FEL pulse with large beam size (approximately 5 cm in diameter). This optical focusing system is located near the output coupler. In an experimental room, the transported FEL is focused again by a pair of Au-coated parabolic mirrors. The whole set-up is mounted in vacuum boxes, which are evacuated up to 10^{-7} Torr, in order to avoid distortion of the FEL pulse due to absorption of the infrared radiation by ambient water vapour, and is connected to the 24-m FEL transport ducts. The optics design was determined to be close to 100% optical transport efficiency by a numerical simulation code GRAD [8].

Experimental setup of SHG-FROG

Figure 2 shows a schematic view of SHG FROG apparatus constructed now. As shown in Fig. 2, the incoming FEL pulse is split by a polyethylene terephthalate (PET) film, whose thickness is 23μm, into two beams. One of them is delayed by a movable retro reflector; the other has a fixed path length. The movable reflector can be achieved by a stepper-motor-driven linear stage on which the retro reflector is mounted. A parabolic mirror focuses both pulses onto the frequency doubling crystal consisting of 2mm thickness Tellurium (Te). The second harmonic generated in the crystal when both pulses have a temporal and spatial overlap propagates through a slit to the spectrometer. Finally a mercury-cadmium telluride (MCT) detector detects the intensity of signal.

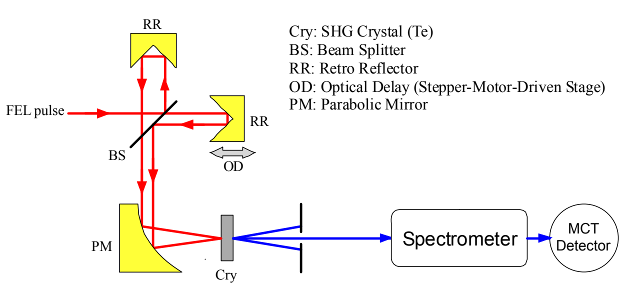

Figure 2: The schematic view of the SHG-FROG setup.

STATUS OF MEASUREMENTS

Spectrum of FEL pulse

Now, the whole optics is being adjusted precisely to provide the SHG signal from the Te crystal, therefore the focus, transport, and measurement system are in atmosphere. The atmospheric transport efficiency was measured to be 40 ~ 50 % due to the absorption by the ambient water vapour, and the average power of FEL pulses was measured to be 500 mW, typically.

Figure 3 shows a spectrum of the FEL pulse measured by a spectrometer. One can find the absorption by the ambient water vapour at 21.1, 21.8 and 22.6 μm.

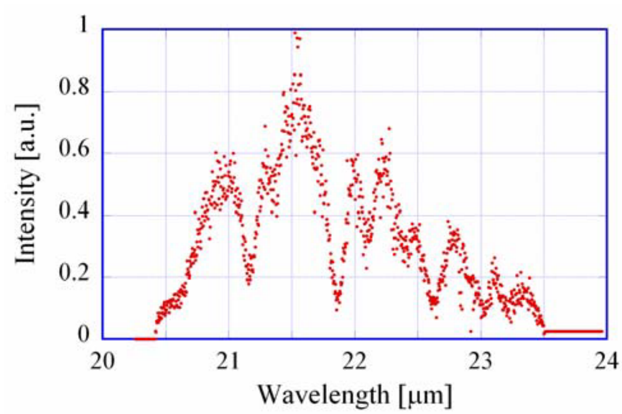

Figure 3: The spectrum of FEL pulse. The absorption of the infrared radiation by the water vapour can be seen at 21.1, 21.8 and 22.6 μm.

Polarization

High efficiency SHG is one of important points to perform SHG-FROG trace. The efficiency depends on an angle between a Te crystal orientation and a polarizing plane, and an input power of the FEL pulse dominantly. On the other hand, the surface of Te crystal is damaged if the pulse is strongly focused to generate the second harmonic. Therefore the angle between the Te crystal orientation and the polarizing plane of FEL pulse should be adjusted precisely.

Figure 4 shows a measurement result of polarizing angle of the FEL pulse at position of Te surface. The measurement was done using a polarizer. In this figure, zero degree corresponds to the horizontal direction. In our case, the FEL pulse has a vertical polarization at the output coupler. At the surface of Te crystal, the polarizing direction rotates to 133 degree due to the refractions via the mirrors in the transport system. The measurement value is good agreement with the designed value.

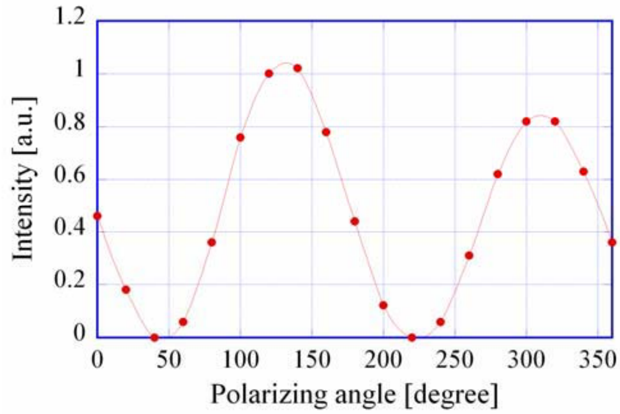

Figure 4: Polarizing angle on the surface of the doubling crystal.

SUMMARY

The femtosecond infrared-chirped FEL is the effective tool of dissociating molecules without the intramolecular vibrational redistribution. JAEA ERL-FEL has been constracted to provide a high-power far-infrared FEL. The ultrashort FEL pulse with the broadband spectrum is achieved operating the long-pulse electron beam from JAEA ERL-FEL. Since it is essential to know both of the intensity and the variation of frequency in the pulse for the high-efficiecny dissociation of the molecule, we consider to measure them by SHG FROG. The numerical simulation of SHG FROG using the FEL parameters can completely retrieves the intensity and phase of the pulse. In the following, we have start to develop the apparatus of SHG FROG. Now we are measuring basic parameters of the FEL pulse (beam size, power, wavelength, polarization and so on) in the experimental room.

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