Ultrafast Laser Science

Light is very common in daily life, on the other hand, light has many interesting physical properties, for example, constancy of velocity, wave-particle duality, etc. The study of light is still important in modern physics.

Light is electro-magnetic field, same as radio wave, however, the measurement of the waveform of light is not easy task even in the 21st century. The difficulty comes from the extremely fast oscillation of the light wave. The oscillation frequency of light wave is the order of hundred terahertz (THz = 10^{12} Hz), in other words, the oscillation period of light wave is the order of femtosecond (fs = 10^{-15} s).

In 2013, we have developed a new method for the measurement of light wave. It is called FROG-CEP, frequency-resolved optical gating capable of carrier-envelope determination. Our method does not need attosecond pulses, even self-referencing is possible. The electric field oscillation of infrared light with the period of several femtoseconds were clearly measured with the method as is shown in Figure 1.

Currently, amplitude modulation and phase modulation are common encoding techniques in optical communication. If we can encode information in the shape of the light wave itself, the communication speed becomes 3 orders of magnitude faster. We believe that our method, FROG-CEP, becomes very important to realize such communication technology.

Other than FROG-CEP, ultrabroadband mid-infrared continuum generation through filamentation and single-shot detection of ultrabroadband mid-infrared spectra has been realized in our laboratory. We are developing such cutting edge technologies for ultrafast laser science.

Figure 1. Infrared light waveforms measured with FROG-CEP. The phase difference between the two infrared pulses was clearly measured.
1. Attenuated Total Reflectance Spectroscopy with Chirped-Pulse Upconversion\(^1,2\)

Attenuated total reflectance Fourier-transform infrared spectroscopy (ATR-FTIR) is a powerful tool to study liquid or solid samples in various scientific and industrial fields. However, the time resolution of the Fourier-transform infrared spectroscopy (FTIR) with the rapid scan mode is limited by the speed of the FTIR device which needs time (at least several milliseconds) for scanning the delay of the interferometer.

Chirped-pulse upconversion\(^1\) is a noteworthy method to improve the time resolution of IR spectroscopy. In this method, an infrared beam is upconverted into a visible beam and the spectrum is measured with a high performance visible dispersive spectrometer. Here, we report attenuated total reflectance (ATR) spectroscopy connected with chirped-pulse upconversion. Single-shot IR absorption spectrum measurement of liquids from 200 to 5500 cm\(^{-1}\) has been realized.\(^2\)

We performed a proof-of-principle experiment by continuously recording IR spectra while exchanging the liquid on the ATR prism from water to aceton and vice versa. The rapid solution exchanging system is a useful tool to study some biological samples at advanced time-resolved ATR-FTIR spectroscopy. It is possible to monitor the IR spectrum in real-time with initiating some chemical reactions by exchanging the solutions on the ATR crystal by using two pneumatic drive pump systems, which are generally used in a stopped flow system.

We show the absorption change at 1230 and 3400 cm\(^{-1}\) in the time domain in Figure 2. At 169 and 422 ms only water and aceton, respectively, should be on the ATR prism. 182 ms is the timing at which the exchange occurs. It is clear that the exchange is finished within 10 ms.

![Figure 2. Measured dynamics of exchange of liquids with the rapid solution-exchange ATR-CPU system. Upper figure: Absorption spectra at each timing. Lower figure: Absorption change at each frequency.](image)

2. Controlling the Carrier-Envelope Phase of Single-Cycle Mid-Infrared Pulses with Two-Color Filamentation\(^5\)

The rapid development of ultrafast laser technology in the last decade has made it possible to study dynamics of electrons in atoms and molecules on attosecond time scale. One of the most famous schemes of attosecond time-resolved spectroscopy is based on attosecond streaking, which is direct waveform measurement of few-cycle electric field using attosecond pulses. In this method, both an extreme ultraviolet attosecond pulse and the target pulse are focused into a noble gas, and measurement of the kinetic energy of the photoelectrons produced by the extreme ultraviolet pulse reflects the vector potential induced by the target field.

In 2013, Fuji et al. has developed a new waveform measurement scheme, frequency-resolved optical gating capable of carrier-envelope phase determination (FROG-CEP).\(^3,4\) The concept is based on a combination of frequency-resolved optical gating (FROG) and electro-optic sampling (EOS), which enables us to determine not only the intensity and (relative) phase profile but also carrier-envelope phase (CEP) at the same time. There are a lot of advantages of the new scheme, such as all-optical method, variety of nonlinear interactions, and possibility of single-shot measurement.

We applied the method for investigation of the phase of the single-cycle pulses generated through multi-color laser filamentation.\(^5,6,7\) We have experimentally found that the CEP variation depends on the frequency of the generated pulses. The phase of high frequency components of the generated pulses continuously and linearly changes with the relative phase between the two-color input pulses, on the other hand, the phase of the low frequency components takes only two discrete values. We have numerically simulated the CEP variation based on the two different models, four-wave mixing and photocurrent. Eventually, both the models are consistent with the experimental results.\(^8\)

References


Award

FUJI, Takao; NOMURA, Yutaka; SHIRAI, Hideto; Laser Research Development Award, the Laser Society of Japan (2015).

* IMS International Internship Program