論文の内容の要旨

Development of a terawatt sub-10-fs laser system and its application in high-order harmonic generation

（テラワットサブ **10 fs** レーザーシステムの開発とその高次高調波発生への応用）

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1) Introduction

By focusing a high-power femtosecond laser pulse into a rare gas medium, high-order harmonic (HH) fields in the extreme ultraviolet (XUV) and soft X-ray spectral range can be generated. The whole setup is compact and can be applied in various cutting-edge experiments including soft X-ray nonlinear optics and attosecond (10^{-18} s) science. Application of attosecond pulses yielded by high-order harmonic generation (HHG) has paved the way to observation and control of different ultrafast physicochemical phenomena such as the electron dynamics in the atoms to open up the frontiers of science.

HHG can be explained by a semi-classical three-step model. A bound electron is tunneling ionized by a strong laser field and travels in a trajectory and recombines with the parent ion to emit XUV or soft X-rays. A two-color laser field synthesized from the fundamental field and its relatively weak second harmonic (SH) field can control the electron trajectory, which results in controlling the HH spectra. This method is considered as a good candidate in intense isolated attosecond pulse (IAP) generation. Specially, a detuned SH field can bring up new frequency components in the HH spectra and relax the conditions of IAP generation. Moreover, a two-color laser field can be even applied in in-situ control of HHG processes. By changing the delay between the two fields, it has become possible to measure and control the birth of attosecond pulses.

Previous research results show that for intense IAP generation, a sub-10-fs laser pulse as the fundamental field with pulse energy of over 10 mJ is necessary. So far, neither an intense IAP that can be applied in soft X-ray nonlinear optics has been generated, nor generation technique of the necessary fundamental field been fully established. A good candidate for this purpose is the chirped pulse amplification (CPA) system of Ti:sapphire laser which is widely applied in many scientific communities. Yet, it has been difficult to amplify sub-10-fs pulses directly in a terawatt CPA laser system owing to the fact that spectral width is restricted by gain narrowing of the laser medium and narrow bandwidth of high-damage-threshold (HDT) mirrors.

With the goal of intense IAP generation, this research first aims to get over these obstacles and develop a terawatt sub-10-fs Ti:sapphire CPA laser system. Then we will apply it in HHG experiments by a two-color laser field synthesized from the fundamental and its detuned SH field for in-situ control of HHG processes. We will also numerically analyze the experimental results and optimize the wavelength of a detuned SH field for relaxing the conditions of IAP generation, through another numerical simulation.

2) Development of a terawatt sub-10-fs laser system

There are three obstacles in developing a terawatt Ti:sapphire CPA laser system in the sub-10-fs regime. The first one is gain narrowing, which is compensated by inserting a partial mirror called a gain narrowing compensator (GNC) in a regenerative amplifier. This GNC reflects the spectral components near 800 nm that have a very high gain and lets the other spectral components get amplified (Fig. 1-(a)). The second obstacle is spectral narrowing due to narrow bandwidth of HDT mirrors. To avoid it in the regenerative amplifier and to compensate for group delay dispersion (GDD) oscillation, a pair of HDT chirped mirrors are used. They hold a sufficiently broad bandwidth for amplification of sub-10-fs pulses and have a low dispersion (Fig. 1-(b)). Also, hybrid broadband HDT laser mirrors are used in a multi-pass amplifier. The third obstacle is high-order dispersion, to compensate which we applied a liquid-crystal spatial light modulator (LC-SLM) holding 640 channels.

The developed laser system (Fig. 1-(c)) consists of a commercial 7-fs Ti:sapphire laser oscillator. The oscillator is followed by an Öffner type pulse stretcher. The stretcher is followed by the LC-SLM. The 200-ps output pulse is injected into the regenerative amplifier pumped by an 11.5-mJ Q-switched green laser at 1 kHz repetition rate. We could successfully compensate for the spectral and gain narrowing by using the HDT chirped

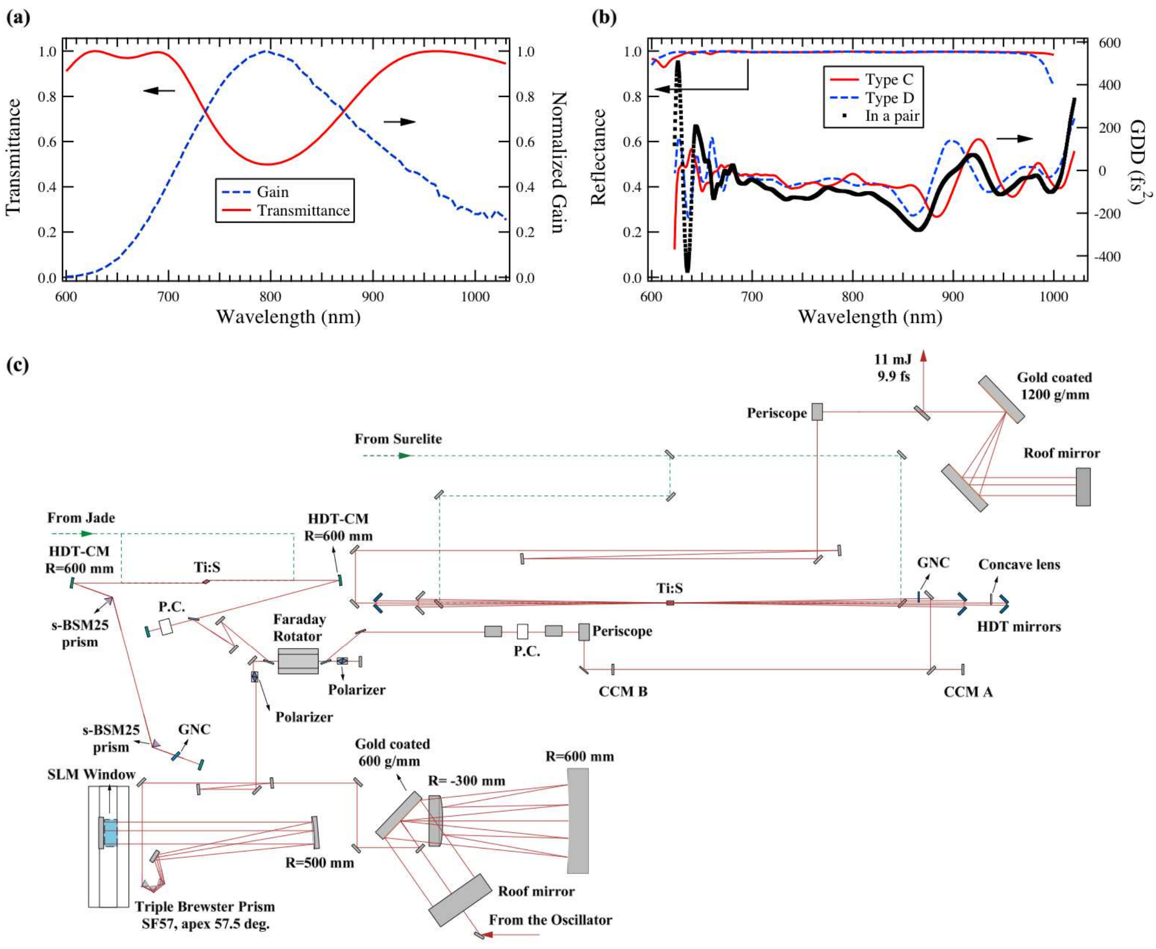

Fig. 1: (a) Calculated transmittance of the GNC (solid curve) compared to gain profile of Ti:sapphire crystal (dashed curve). (b) Spectral characteristics of the HDT chirped mirror pair. The upper curves show the calculated reflectance and the bottom curves show the measured GDD of these mirrors. The dots show the total GDD measured in a pair. (c) Schematic of the developed terawatt sub-10-fs Ti:sapphire CPA laser system.

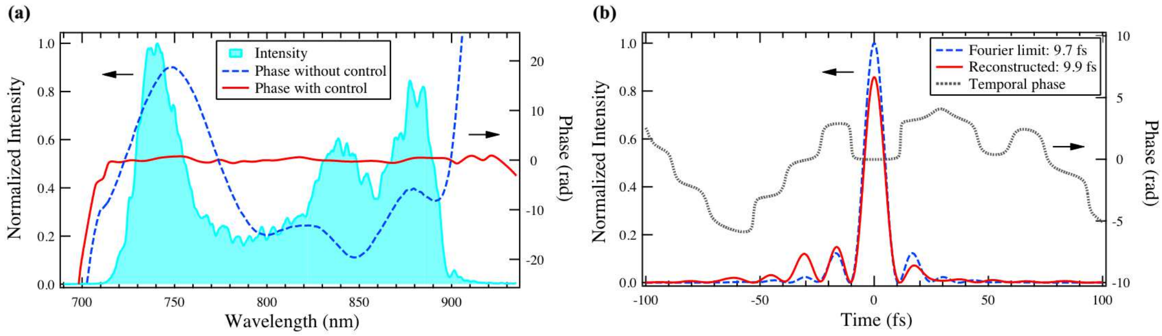

Fig. 2: (a) Spectrum behind the compressor (hatched area) and the initial phase (dashed curve). The solid curve shows the spectral phase with control by the LC-SLM. (b) Fourier-limit temporal profile (dashed curve) and the reconstructed temporal profile (solid curve). The dotted curve shows the phase in the temporal domain.

mirrors and the GNC. The obtained pulse energy and bandwidth were 27 μJ and 230 nm, respectively. The amplified pulse is sent to a pulse slicer to isolate the two amplifiers and to reduce the repetition rate to 10 Hz. After 5-pass amplification in a bowtie multi-pass amplifier consisting of hybrid HDT laser mirrors and being pumped by the SH of a Q-switched YAG laser with pulse energy of 180 mJ, an amplified 33-mJ pulse was obtained.

The amplified pulse is sent to a pulse compressor, the spectrum behind which is shown in Fig. 2-(a) by the hatched area with a bandwidth of 210 nm. The Fourier-limit pulse duration is 9.7 fs. Without using the LC-SLM, the measured spectral phase (dashed curve in Fig. 2-(a)) by the SPIDER technique, has a high modulation. We produced the opposite of this phase by the LC-SLM and applied it to the laser pulse and successfully achieved an almost flat spectral phase (solid curve in Fig. 2-(a)). The reconstructed temporal profile (solid curve in Fig. 2-(b)) calculated by the spectrum and the flat spectral phase has an FWHM of 9.9 fs. The pulse energy after compression was 11 mJ and therefore yields a peak power of 1.1 TW. As a result, we have achieved direct amplification of 9.9 fs pulses in a terawatt Ti:sapphire CPA laser system, for the first time.

3) In-situ control of HHG processes

As an application, we generated HH fields by a two-color laser field synthesized from the fundamental field and a detuned SH field of this laser system. By controlling the electron trajectory, which happens at a sub-fs time scale, we performed a high-resolution experiment to reveal the effects of delay change on the HH spectra to observe a previously unknown characteristic of HH fields. To do this, we have used a 15-mJ fundamental field with a pulse duration of ~15 fs yielded by a brief modification in the GNC to realize a higher pulse energy. The detuned SH field has been generated by a Type-I BBO crystal to result in a stretched ~40-fs pulse at a central wavelength of 417 nm detuned from the exact SH field (400 nm). This shift of wavelength plays an important role in the observed phenomenon. The SH field and the residual fundamental field were sent into a two-color interferometer to synthesize the two-color laser field.

Using a broadband dichroic mirror, the two-color laser field is reflected into a focusing chamber consisting of an Al-coated off-axis parabolic mirror (f=500 mm) and a 12-mm gas cell. The HH spectra are measured by an XUV imaging spectrograph. The pulse energy of the fundamental field was 8.5 mJ and that of the SH field was

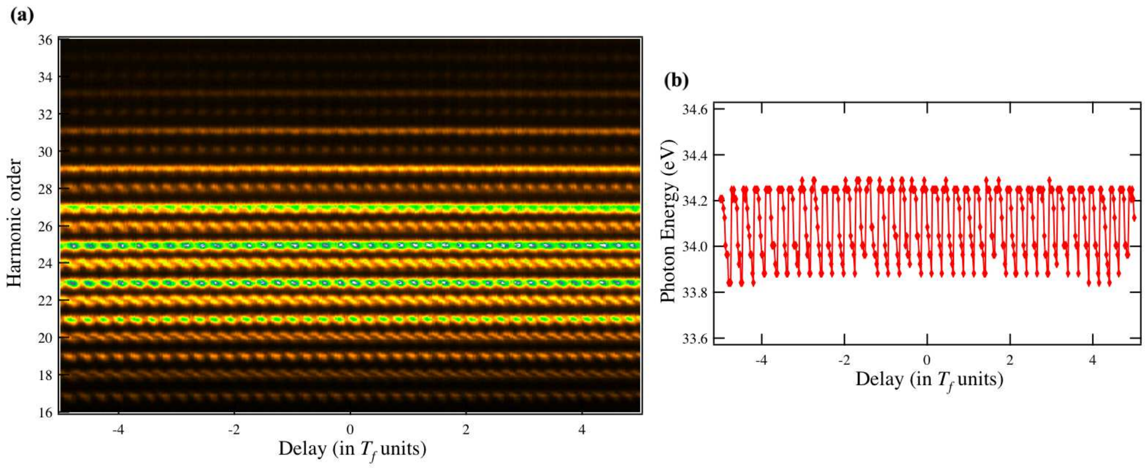

Fig. 3: (a) Spectrogram showing the effects of delay on the HH spectra. T_f indicates the optical period of the fundamental field. (b) Peak frequency of the 22nd harmonic field against the delay, showing a modulation amplitude of ~0.4 eV.

125 μJ. The two fields have the same beam spot at the focus (~100 μm) to get a high yield. The target gas was argon (Ar) with a backing pressure of ~20 torr. By changing the delay in 20 nm steps, HH spectra for every 100 laser shots were measured, which show intensity modulation of not only the even-order harmonic fields but also the odd-order harmonic fields (Fig. 3-(a)). Moreover, we also find modulation of the frequency (photon energy) of the even-order harmonic fields with an amplitude up to ~0.4 eV for the 22nd harmonic field as shown in Fig. 3-(b). Further investigations showed that this phenomenon could be controlled by the intensity and chirp of the fundamental field to result in a larger amplitude by a fundamental field with a higher intensity and a small amount of minus chirp (GDD≅-75 fs^2). As a result, we have been able to observe periodical frequency modulation of HH fields, for the first time to add another degree of freedom to the control parameters of HHG processes.

As a further step towards understanding this phenomenon, we numerically solved the time-dependent Schrödinger equation (TDSE) on an Ar atom within the single-active-electron approximation, which has been already developed in a previous study. One of the parameters we could not widely consider during the experiment was the wavelength of the detuned SH field. Hence, we investigated the effects of the detuned SH wavelength as well as those of the intensity and chirp of the fundamental field on the frequency modulation of HH fields. The numerical analysis revealed that an exact SH field (400 nm) is not able to bring up frequency modulation of HH fields, even though the fundamental field is chirped and the intensity is increased. While the same conditions with the 417-nm detuned SH field results in frequency modulation of HH fields with an amplitude getting larger by increasing the intensity and chirp of the fundamental field.

4) Simulation of IAP generation with a two-color laser field

By numerically solving the TDSE, we also calculated HH spectra from a neon atom obtained by a two-color laser field synthesized from a 10~15-fs fundamental field (3.2×10^{14} W/cm^2) and a 40-fs detuned SH field (7.2×10^{12} W/cm^2). By variation of the wavelength of the detuned SH field when the delay is optimized, continuous HH spectra can be obtained when the wavelength is shorter than 400 nm (λ_{DSH}≤400 nm) as shown in Fig. 4-(a). The HH spectra in the cut-off region are selected with a broadband super-Gaussian filter and inverse

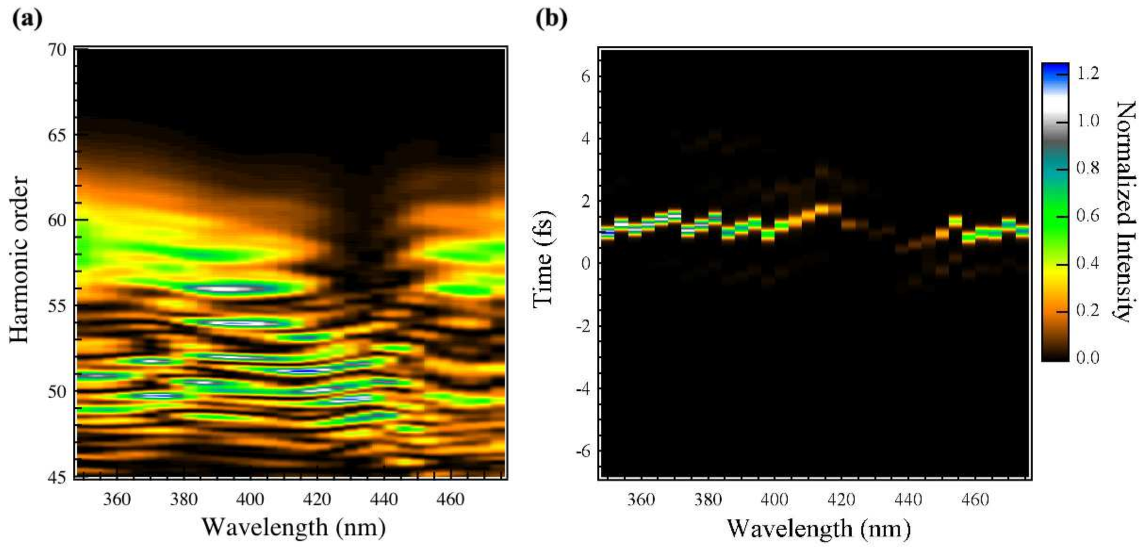

Fig. 4: (a) Calculated HH spectra obtained by a two-color laser field synthesized from a 10-fs fundamental field and a 40-fs detuned SH field in the range of 350 nm~474 nm. (b) The attosecond pulses calculated after filtering the cut-off region.

Fourier transformed to calculate the attosecond pulses shown in Fig. 4-(b). Likewise, IAPs are obtained with $\lambda_{DSH}\leq$400 nm, while low-intensity attosecond pulse trains are obtained otherwise. Investigating the same effects with only increasing the pulse duration of the fundamental field (δt), numerical analysis has revealed that an IAP can be obtained easier provided that $\delta t\leq$12 fs and λ_{DSH}<380 nm, while with a 15-fs fundamental field an IAP can not be obtained. We also investigated the effects of carrier-envelope phase on the obtained IAP with the two-color laser field and compared the results to those obtained by a 5-fs fundamental field alone with the same peak field amplitude.

5) Conclusion

By establishing the spectral narrowing, gain narrowing and high-order dispersion compensation techniques in the sub-10-fs regime, we have successfully developed a 1.1-TW 9.9-fs Ti:sapphire CPA laser system, for first time. As an application, HH fields have been generated by a two-color laser field synthesized from the fundamental and the detuned SH field of this laser system with a modified GNC to verify the effects of delay on the HH spectra for in-situ control of HHG processes. As a result, periodical frequency modulation of HH fields has been observed, for the first time. We have also investigated the effects of several other laser parameters on this phenomenon and have shown that it can be controlled by the intensity and chirp of the fundamental field. Numerical analysis by solving the TDSE highly approves of the experimental results and shows that this phenomenon can be also controlled by the wavelength of the detuned SH field. In another simulation, we have optimized the wavelength of the detuned SH field to relax the conditions of IAP generation and have shown that an IAP can be relatively easier obtained by a 10~12-fs fundamental field superposed to a detuned SH field shorter than 380 nm. All of these results are promising in intense IAP generation using the developed laser system.