Extreme Laser Science Laboratory（2023）

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(0) Research fields

CPR Subcommittee**:** Physics

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(1) Long-term goal of laboratory and research background

Laser is highly controllable light, and by using various laser technologies, we can create optical pulses with arbitrary electric field shape, pulse duration, and beam shape. The main research goal of our laboratory is to extend the wavelength range of coherent light sources up to hard x-ray by using the extreme nature of laser light, such as ultra-short pulse duration, ultra-broadband wavelength, and ultra-high intensity. Recently, we have been advancing research aimed at achieving sub-10 fs pulse duration in the mid-infrared laser through the utilization of femtosecond laser technologies, and targeting the single- and sub-cycle of the laser electric field contained within its intensity envelope. This effort not only leads to the shortening the temporal duration of attosecond soft x-ray pulses generated by high-order harmonic generation but also serves as fundamental research for post-attosecond laser, namely zeptosecond laser sources.

(1) Current research activities (FY2023) and plan

(A-1) Development of a single-cycle laser using DC-OPA

High power lasers in the near to mid-infrared region are expected to have a wide range of applications from basic science to industrial use, and there has been active development of lasers with shorter pulses and higher intensity. Among them, single-cycle laser, which have only one cycle in a pulse, are useful in attosecond science and are expected to be applied to various physical property measurements. We have developed a single-cycle laser using the DC-OPA (dual chirped optical parametric amplification) method in FY2023, and used it to generate attosecond pulses by high harmonics generation.

Single-cycle laser has a bandwidth of about one octave, and for high intensity, it is necessary to amplify this wide bandwidth at the same time. We used a combination of two types of crystals (BiBO and MgO:LN) to amplify 1.4 μm-2.4 μm with BiBO and 2.4 μm-3.0 μm with MgO:LN, and succeeded in amplifying a bandwidth exceeding one octave in total at one time. Finally, a laser with 1.05 cycles (8.58 fs), 53 mJ pulse energy, and 6 TW peak power at a center wavelength of 2.44 μm was generated, and this laser was used for higher harmonic generation. Figure 1 shows the carrier envelope phase (CEP) dependence of the high harmonic spectrum when Ar gas is used as the nonlinear medium. It can be seen that high harmonic spectrum changed over a very broadband from 80 eV to 160 eV with the single-cycle laser as the pump laser. This CEP-dependent spectral region can be used as an isolated attosecond pulse. The pulse width calculated from the spectral intensity is 50 as and the period is 33 as, suggesting that 1.5 cycles of isolated attosecond pulses can be generated if we can compensate the dispersion.

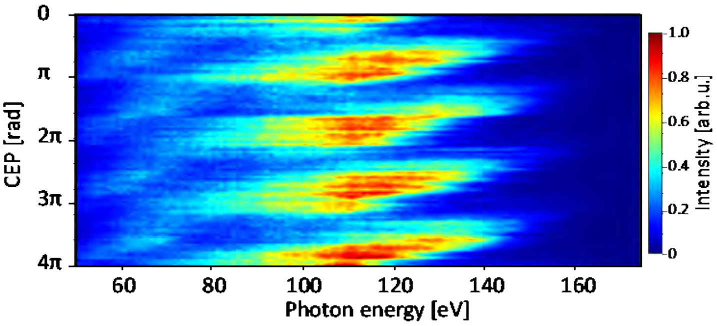

Figure 1. CEP dependence of HH spectrum

Future plan, A single-cycle laser with a peak power of 6 TW was developed in FY2023, and we will develop a 10 TW-class sub-cycle laser (0.75 cycle) in FY2024. To realize the amplification of sub-cycle lasers, it is necessary to amplify a bandwidth of 1.8 octaves at the same time and compensate for its dispersion. To cover this very wide bandwidth, one BiBO crystal and two MgO:LN crystals are used for amplification, and two AOPDFs with different wavelength bands are used for dispersion compensation. This will generate a 0.75-cycle laser with a center wavelength of 2.47 μm, pulse energy of 65 mJ, and pulse width of 6.2 fs, which will be applied to high harmonic generation generation.

(B-1) Development of Co:MgF$_2$ amplifier for mid-infrared broadband pulses

Our team have been developing ultrashort and intense laser pulses in mid-infrared region aiming at generation of intense coherent attosecond pulse in soft X ray region utilizing high harmonic generation process. Broadband amplifier scheme covering octave bandwidth of 1.5-3 μm is strongly desired to obtain higher repetition rate and higher photon flux. Here, we propose a hybrid master oscillator power amplifier (MOPA) scheme with different kinds of gain crystals, as shown in Fig.2 (a). In this wavelength region, Cr:ZnS or Cr:ZnSe crystal is popular owing to their broadband gain in 2-3 μm and the availability of high-power pump sources such as Ho:YAG lasers. To compensate 1.5-2 μm region, we focused on Co:MgF$_2$ crystal which are used as laser material at 1.5-2.3 μm.

In this year, we evaluated the gain of a Co:MgF$_2$ crystal in this wavelength region. Because Co:MgF$_2$ crystal has absorption peak around 1.3 μm, we first developed 1.3 μm Nd:YAG Q-switched laser as the pump laser. Since the emission cross section of Nd:YAG at 1064 nm is much higher than that of 1.3 μm, we employed specially designed mirrors to suppress the laser emission at 1064 nm. Then we achieved a pulsed laser with the repetition of 1 kHz, pulse energy of 1.5 mJ, and pulse duration of 120 ns. We also prepared the seed pulses covering 1.5-3 μm by using intra-pulse difference frequency generation from ultra-broradband Ti:Sapphire laser pulses which are spectrally broadened by a gas-filled hollow core fiber. Then, we measured one-pass gain of Co:MgF$_2$ crystal by focusing both pump and seed lasers on the crystal, and measuring the change of the transmitted seed pulse, as shown in Fig. 2 (b). We have observed 10 % gain signal under the pump fluence of 8 J/cm^2 as shown in Fig. 2 (c).

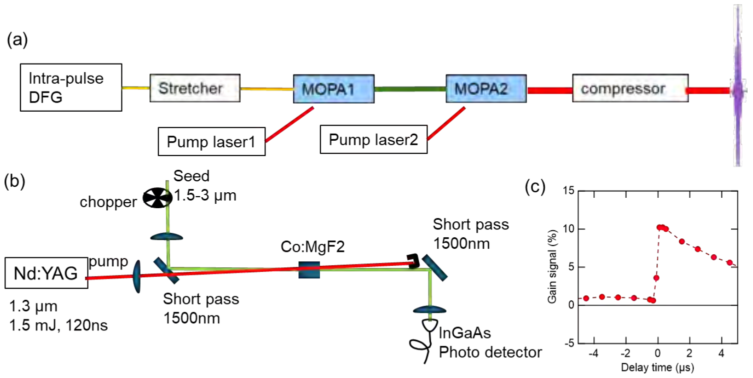

Fig.2 (a) The concept of hybrid master oscillator power amplifier (MOPA) system. (b) A schematic for evaluation of one-pass gain in a Co:MgF$_2$ crystal. (c) Delay-time dependence of gain signal.

<u>Future plan</u>, In FY2024, we will examine the possibility of the hybrid amplifier system combining Cr:ZnSe and Co:MgF$_2$ crystals. For the Co:MgF$_2$ amplifier, development of Nd:YAQ Q-switched pump laser with higher pulse energy is necessary for scaling up. The optimization of dope density and quality of Co:MgF$_2$ crystal would be also important. For the Cr:ZnSe amplifier, we have already purchased Ho:YAG Q-swtiched laser for pumping in FY2023. We will evaluate the gain of Cr:ZnSe, and will develop a regenerative amplifier or a multi-pass amplifier.

(C-1) Construction of sub-μm focusing optics for soft x-ray isolated attosecond pulses

Soft x-ray has been widely utilized for applications, including the observation of surrounding atomic and electronic information from absorption spectral structures and its use in element-selective microscopy and processing. Recently, isolated attosecond pulses of soft x-ray with high spatio-temporal coherence and peak power in the GW class has been obtained using high harmonic generation pumped by ultrashort laser pulses and are used for attosecond nonlinear optical phenomena in gas media. In this study, to generate attosecond nonlinear optical phenomena in solids, which are characterized by high atomic density and band structure formation, we generate soft x-ray isolated attosecond pulses with a high peak intensity of PW/cm2 class. This is expected to lead not only to academic developments such as the observation of peculiar nonlinear electron dynamics in solids in the real-time domain which has not been seen in gases, but also to practical applications such as the development of high-power devices utilizing the high atomic density of solids and PHz optical switch elements based on saturable absorption.

In FY2023, to obtain a high peak intensity of PW/cm^2 class, a focusing optics was constructed using a spheroid mirror for soft x-ray (Fig. 3(a)), which was fabricated in cooperation with the Ultrahigh Precision Optics Technology Team. The spheroid mirror is precisely adjusted to remove aberrations using a five-axis stage, including two rotation axes. A mechanism to hold the sample at the focal point has been fabricated, and position control using a 3-axis piezo stage enables knife-edge measurements of soft x-ray isolated attosecond pulses with a focal beam diameter of 1 μm or less to be performed. An x-ray CCD camera is installed downstream of the focus, allowing the pulse energy and the beam shape of the soft X-ray isolated attosecond pulses to be observed. Experiments were carried out to focus a He-Ne laser using this focusing optics, and a focused beam diameter of 8 μm (1.3 times the diffraction limit) was achieved (Fig. 3(b)). The diffraction limit of the soft x-ray isolated attosecond pulses with the focusing optics is 0.59 μm, suggesting the generation of a soft X-ray isolated attosecond pulses with a focused beam diameter of 0.77 μm and a peak intensity of 3 PW/cm2. Soft x-ray isolated attosecond pulses are in the process of the observation (Fig. 3(c)) and will soon be focused to less than 1 μm.

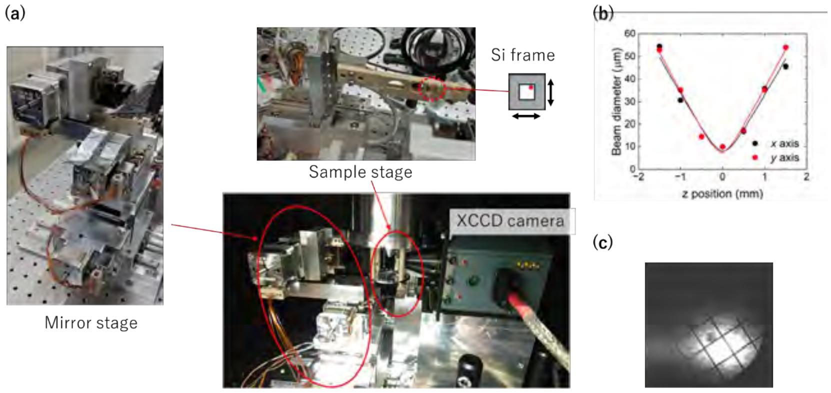

Fig. 3 (a) Focusing optics for soft x-ray isolated attosecond pulses. (b) Knife-edge measurement with He-Ne laser. (c) Observation of soft x-ray isolated attosecond pulses with x-ray CCD camera.

Future plan, In FY2024, the generation of soft x-ray isolated attosecond pulses with a peak intensity of PW/cm2 class and the observation of nonlinear optical phenomena in solids using the pulses will be carried out. The former is expected to be achieved by optimizing the generation of soft x-ray isolated attosecond pulses by high harmonic generation and aligning the focusing optics. The latter involves second harmonic generation and saturable absorption in solid-state nonlinear optical phenomena. Although there were reported cases with femtosecond light pulses in both cases, the relatively long pulse widths were not sufficient to capture electron dynamics. In this study, observation of nonlinear electron dynamics in solids will be achieved by using attosecond light pulses, which is in the time scale of electron motion.

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(4) Representative research achievements

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Laboratory Homepage
https://www.riken.jp/en/research/labs/chief/els/index.html