ISBN: 978-3-95450-210-3 doi:10.18429/JACoW-FEL2019-TUD02

APPLICATION OF INFRARED FEL OSCILLATORS FOR PRODUCING ISOLATED ATTOSECOND X-RAY PULSES VIA HIGH-HARMONIC GENERATION IN RARE GASES

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Abstract

High harmonic generation (HHG) in rare gases is now becoming a common technology to produce attosecond pulses in VUV wavelengths. So far HHG sources have been realized by femtosecond solid-state lasers, not FELs. We propose a FEL-driven HHG source to explore attosecond pulses at photon energies above 1 keV with a MHz-repetition, which is difficult with solid-state lasers. A research program has been funded to establish technologies for the FEL-HHG, which covers generation and characterization of few-cycle IR pulses in a FEL oscillator, stacking of FEL pulses in an external cavity, and a seed laser for stabilization of carrier-envelope phase in a FEL oscillator. In this paper, we present the scheme of FEL-HHG and the status of the research program.

INTRODUCTION

Recent development of solid-state laser technologies has realized a generation of isolated attosecond pulses in ultraviolet and X-ray wavelengths via high harmonic generation (HHG) in rare gas and solid-state targets [1]. As such attosecond photon sources are routinely available in laboratories, attosecond science is becoming an active research field, in which ultrafast dynamics in atoms and molecules is investigated in detail.

Combination of HHG and a free-electron laser (FEL) was examined at a SASE FEL, where a UV pulse from HHG was used as a seed laser to stabilize the shot-to-shot variation in spectrum and energy of the FEL pulses [2]. The experiment can be called HHG-seeded FEL or HHG-FEL. To the contrary, an optical pulse generated from an infrared FEL can be used, in principle, to drive HHG, that is FEL-HHG [3]. However, such an experiment has never been considered seriously.

In the present paper, we describe a research project towards the FEL-HHG to generate attosecond VUV and X-ray pulses from an infrared FEL oscillator.

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FEL-HHG

Advantage of FEL-HHG

There are several trends in the development of HHG photon sources. One of the major trends is increasing the photon energy (or decreasing the photon wavelength) available in HHG photon sources. The highest photon energy so far demonstrated is a generation of a bright supercontinuum that spans from the ultraviolet to more than 1.6 keV [4]. The experiment utilized a mid-infrared laser pulse, a wavelength of $\lambda = 3.9\ \mu m$, as a driver of HHG. Theoretical and experimental studies revealed that the HHG cut-off energy scales as $\lambda^{1.7}$ in the phase-matched condition [4]. Pushing the cut-off energy above 1.6 keV has not realized mainly due to the lack of mid-infrared laser pulses satisfying conditions for HHG, wavelength, pulse energy, pulse duration, and repetition. Thanks to wavelength tunability and high-repetition availability, a FEL oscillator is a potential driver of HHG photon sources complementary to conventional solid-state femtosecond lasers.

Few-Cycle Pulse Generation

Generation of isolated attosecond pulses from HHG is available in several schemes [5]. The simplest one is utilizing carrier-envelope-phase-stable (CEP-stable) few-cycle pulses as a driver of HHG.

In a FEL oscillator, few-cycle lasing has been studied experimentally and theoretically. In normal conducting linac FELs, few-cycle lasing was observed in a transient regime before the onset of saturation, when the FEL oscillator was operated at a small negative detuning of the cavity length [6]. This lasing behavior was analytically studied in the context of supermode theory and recognized as a multisupermode regime that occurs in the limit where all the supermodes converge toward a unique degenerate supermode [7]. After the onset of saturation, the lasing shifts to a chaotic regime and a few-cycle solitary pulse is no longer available.

Another type of few-cycle lasing appears in a FEL oscillator operated with a long macro pulse from a superconducting linac [8]. This steady-state few-cycle lasing is accompanied by high extraction efficiency and occurs at the condition of a

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perfectly synchronized cavity, or zero detuning length, with high-gain and small-loss parameters [9]. We consider this lasing can be applied for driving a HHG photon source.

Stabilization of Carrier-Envelope Phase

In the generation of isolated attosecond pulses from few-cycle laser pulses, photon yield and cut-off energy depend on carrier-envelope phase (CEP) [10]. Therefore, CEP-stable laser pulses are necessary for a practical use of isolated attosecond pulses from HHG photon sources. In solid-state lasers, generation of CEP-stable few-cycle pulses is now a well-established technology.

However, CEP stabilization in FEL oscillators has never been demonstrated because the evolution of FEL pulses is initiated by the shot noise. In the steady-state few-cycle lasing at the perfectly synchronized cavity, the leading part of the optical pulse contains incoherent shot noise with random amplitude and phase. The amplitude and phase in the entire FEL pulse are governed by the interaction between the electrons and the radiation initiated by the shot noise in the leading part. Consequently, the carrier frequency and phase of the FEL pulses are not stabilized.

We propose that the above shot-noise-induced fluctuation can be completely stabilized by an external CEP-stable seed laser to lock the phase and amplitude of the leading part of the FEL pulse [11]. Simulations including the seed laser pulse show that a CEP-stable few-cycle FEL pulse can be generated at a FEL oscillator and the jitter of CEP is small enough with a realistic accelerator parameters.

PROPOSED CONFIGURATION OF FEL-HHG

Figure 1 shows a schematic view of the proposed FEL-HHG. The FEL wavelength should be determined from the requirements of attosecond pulse generation in HHG. For a generation of attosecond pulses whose wavelength covers from VUV to X-ray above 1 keV, we assume the FEL wavelength of 2-6 μm from the scaling law of HHG cut-off energy. A superconducting linac is adopted to deliver a CW pulse train to the FEL-HHG.

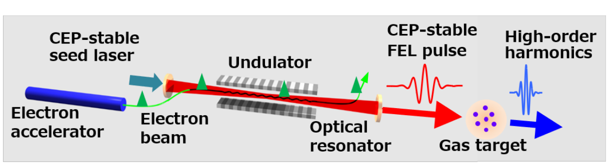

Figure 1: Schematic view of high-harmonics generation driven by an infrared FEL oscillator (FEL-HHG).

Example sets of FEL-HHG parameters are listed in Table 1, where the FEL wavelength is 2 μm and 6 μm and the parameters are chosen so that FEL pulse energy is 0.5 mJ. We consider such FEL oscillators can be constructed with existing technologies: a photo-cathode electron gun [12] and superconducting RF cavities [13].

Table 1: Example Parameters of FEL Oscillators for FEL-HHG

	(A)	(B)
Electron beam		
energy (MeV)	85	50
bunch charge (pC)	60	100
norm. emittance (x/y) (mm-mrad)	12/12	12/12
bunch length (ps)	0.27	0.4
peak current (A)	220	250
bunch repetition (MHz)	10	10
undulator		
undulator parameter (rms)	1.34	1.25
pitch (cm)	4.0	4.5
number of periods	80	40
FEL		
wavelength (μm)	2	6
Rayleigh length (m)	0.92	0.52
FEL parameter, ρ	0.0030	0.0052
cavity loss	6%	4%

RESEARCH PROGRAM

Overview of the Program

A 10-year research program (2018-2027) has been funded to establish basic technologies for the FEL-HHG. In the program, we are conducting research and development towards the FEL-HHG at two FEL facilities, KU-FEL at Kyoto University and LEBRA-FEL at Nihon University, both of which are infrared FEL oscillators driven by normal conducting linear accelerators. These facilities of normal conducting FELs can be exploited for basic technologies development and a following proof-of-concept experiment of FEL-HHG, whereas FEL-HHG for a full-scale application of attosecond VUV and X-ray pulses should be realized by a superconducting linac FEL oscillator.

The research subjects to be conducted are (1) generation and characterization of few-cycle mid-infrared pulses from FEL oscillators, (2) enhancement of FEL pulse energy by an external optical cavity, (3) scheme for CEP stabilization of FEL pulses including the development of a mid-infrared seed laser. We plan to explore these subjects in the first 6 years, 2018-2023, and proceed to an experimental demonstration of the FEL-HHG at one of two facilities. A seed laser for the CEP stabilization and a HHG gas target will be developed at National Institutes for Quantum and Radiological Science and Technology (QST).

In the following sections, we describe the status of our research program at each laboratory.

KU-FEL

KU-FEL at Kyoto University is operated to provide FEL pulses from 3.4 to 26 μm for user experiments [14]. We recently achieved lasing with high-extraction efficiency at the perfectly synchronized cavity length at KU-FEL and a

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numerical simulation reproducing the experimental result suggests a generation of a few-cycle FEL pulse [15]. For realizing such lasing within a limited macro pulse duration, 8 μs, at KU-FEL, we applied dynamic cavity desynchronization (DCD), in which RF phase for the klystron input signal was modulated to change the electron bunch interval during a macro pulse [16]. We optimized parameters for the DCD, timing and depth of the modulation, to maximize the extraction efficiency. A measurement of spent electron beam energy indicated a FEL extraction efficiency of 5% at the end of a macro pulse as shown in Fig.2. In this experiment, electron beam energy was 27 MeV, bunch charge was 40 pC, and FEL wavelength was 11.6 μm.

In our research program, we plan to study a scaling law of FEL extraction efficiency and pulse duration with respect to the FEL gain and the cavity round-trip loss. Previous studies show that efficiency is a function of the cavity round-trip loss normalized to the FEL small gain integrated over the slippage distance [17].

The FEL lasing of 5% extraction efficiency was realized with 40-pC electron bunches from a 4.5-cell RF gun operated at a thermionic cathode mode. The bunch charge can be increased to 120 pC by changing the gun operation to a photocathode mode [18]. A drive laser for the photo-cathode operation is now ready and FEL lasing with 120-pC bunches will be soon carried out.

The FEL cavity of KU-FEL is equipped with two gold-coated copper mirrors, one of which has an on-axis small hole for out coupling. In order to reduce the cavity loss, the mirrors will be replaced by dielectric mirrors. After the replacement, we expect the cavity loss can be reduced from 3% with the hole-coupling to 1% with a dielectric mirror. Inverse square root of the normalized cavity loss, $1/\sqrt{\alpha}$, for FEL lasing at 10μm can be varied from 13 (with thermionic cathode and metal mirrors) to 38 (with photocathode and dielectric mirrors), which is beyond the JAERI-FEL parameter to demonstrate extraction efficiency of 9%, $1/\sqrt{\alpha} = 27$ [9]. For the further increasing of FEL pulse energy, we also plan to install a new 1.6-cell RF gun to provide a train of 1-nC bunches.

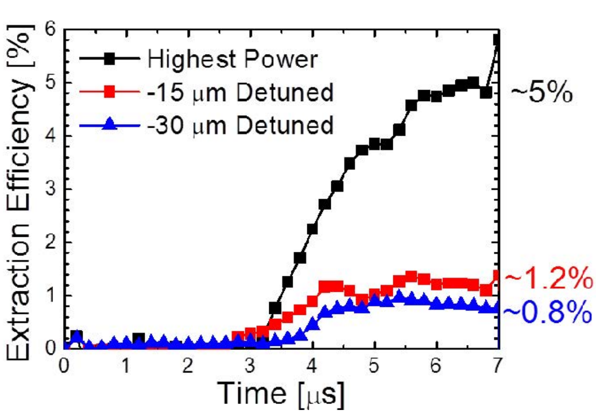

Figure 2: Temporal evolution of extraction efficiency under the relative cavity length of 0, -15 and -30 μm. [15]

LEBRA-FEL

The FEL oscillator at LEBRA covers 0.827-6.1 μm with a macro-pulse of 20μs [19]. We plan to explore FEL pulse stacking in an external optical cavity utilizing advantage of the relatively long macro pulse.

Stacking laser pulses in an external cavity is a common technology to enhance the energy of pulses from a mode-locked laser for HHG [20] and other applications [21]. Generation of high-harmonics from a gas target is possible either using stored pulses in an external cavity or using a laser pulse dumped from a cavity. Such cavity dump can be done with semiconductor photo switches.

An experiment of FEL pulse stacking was conducted at the superconducting linac FEL at Stanford. They demonstrated accumulation of micropulses with more than 75 times the energy of the incident FEL pulses [22]. The Stanford FEL was operated at a quasi-CW mode and the external cavity could work in the steady-state mode, in which the injected pulse energy was balanced with the cavity loss. In normal-conducting-linac FELs, pulse stacking in the transient mode is suitable for maximizing the stored pulse energy [23]. In LEBRA-FEL, we will optimize the external cavity to realize FEL pulses for HHG experiments.

For the pulse stacking experiment, we set up an external cavity at an experimental room of LEBRA-FEL. The cavity is a bow-tie shape and the frequency of the cavity is chosen at 44 MHz, double of the FEL cavity frequency. The cavity length must be controlled precisely to stack successive pulses coherently. The FEL pulse train of 20 μs is, however, not long enough to tune the cavity length. Thus, we use a fiber laser oscillator for the cavity tuning. As a preliminary tuning of the cavity, we injected laser pulses from the fiber oscillator and confirmed multiple recirculation of injected laser pulses in the cavity (Fig. 3). Further tuning of the external cavity is in progress.

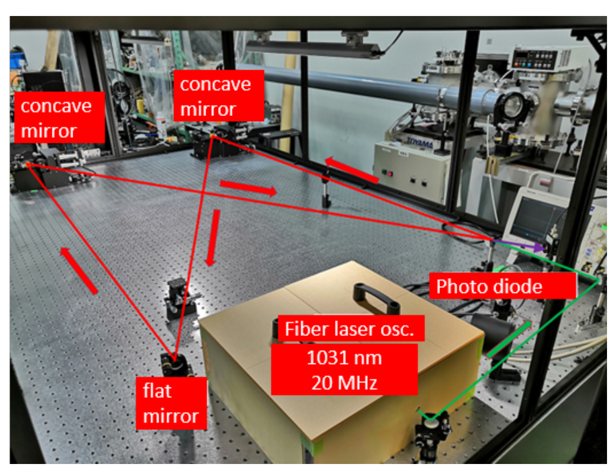

Figure 3: The external cavity for the FEL pulse stacking experiment at LEBRA-FEL.

In parallel with the experimental work, we are making simulation studies to predict a performance of the pulse stacking with optimized cavity parameters, transmittance

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of the input coupler and recirculation path length. In the simulations, we use a 3-dimensional FEL code, GENESIS, coupled with a wave propagation code to calculate iterative interaction of a FEL pulse and electron bunches [24].

QST

A seed laser for the CEP-stabilization in a FEL oscillator must provide CEP-stable laser pulses with a moderate pulse energy, ~ 1 nJ. In addition, the laser pulse should be synchronized to the electron bunch repetition. Since generation of such laser pulses below 3 μm is well-established, we focus our efforts on the development of seed lasers at wavelength longer than 3 μm. We design a laser system comprising a mode-locked fiber oscillator and a 30-W class fiber amplifier followed by a difference frequency generation between the light pulse of the fiber laser and its wavelength shifting. Figure 4 shows a fiber oscillator developed as the first stage of the laser system. The oscillator produces laser pulses at a repetition rate of 23.5 MHz and an average power of 150-300 mW. The second stage of the system, a 30-W amplifier, is now under fabrication.

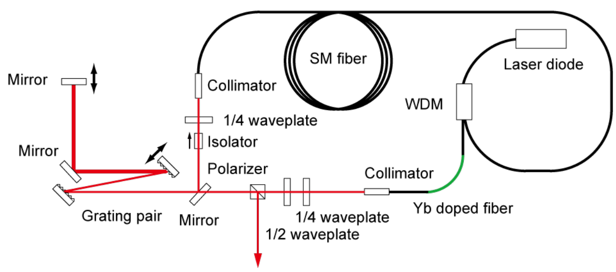

Figure 4: Schematic drawing of a fiber oscillator developed for a mid-IR seed laser. Output laser pulses are delivered to an amplifier followed by a difference frequency generation.

SUMMARY

We have proposed an attosecond photon source based on high-harmonic generation driven by mid-infrared FEL oscillators (FEL-HHG). The FEL-HHG is able to provide attosecond VUV and X-ray pulses with a MHz repetition. A research program has been funded for developing basic technologies for the FEL-HHG and realizing a proof-of-concept experiment of the FEL-HHG. In this research program, two FEL facility, KU-FEL and LEBRA-FEL, are exploited as platforms for the technology development.

ACKNOWLEDGEMENTS

This work was supported by MEXT Quantum Leap Flagship Program (MEXT Q-LEAP) Grant Number JPMXS0118070271. The authors thank Xiao-Min Tong, Jiro Itatani, Nobuhisa Ishii and Keisuke Nagashima for helpful discussion.

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