

High Harmonic Frequency Combs Based on an Infrared FEL Oscillator



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National Institutes for Quantum Science and Technology (QST)

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Collaborators



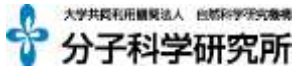
川瀬啓悟/Keigo Kawase (QST)



大垣英明/Hideaki Ohgaki, 全炳俊/Heishun Zen (Kyoto U.)

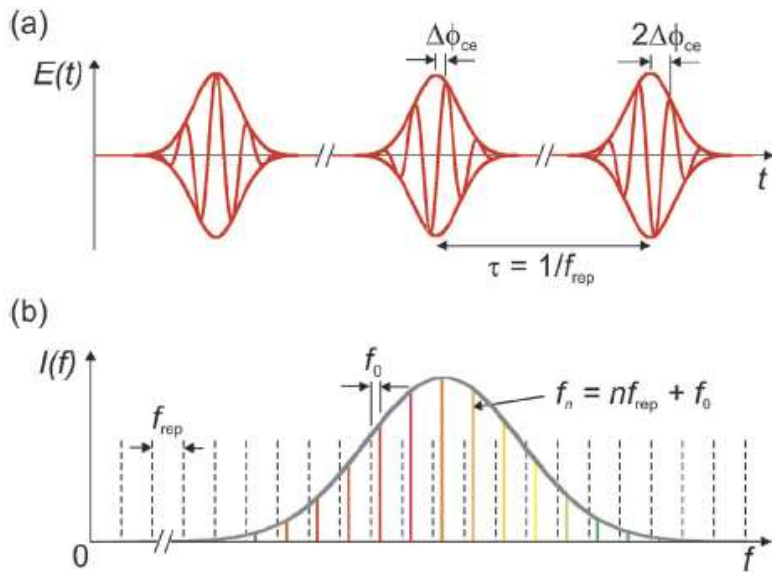


早川恭史/Yasushi Hayakawa, 境武志/Takeshi Sakai (Nihon U.)



金井恒人/Tsuneto Kanai (IMS)

This work was supported by MEXT Quantum Leap Flagship Program (MEXT Q-LEAP) Grant Number JPMXS0118070271 and JSPS KAKENHI (22H03881, 23K25315), Joint Usage/Research Program on Zero-Emission Energy Research, Institute of Advanced Energy, Kyoto University (ZE2021B-28, ZE2022B-23, ZE2023B-20)



H.S. Margolis, Chem. Soc. Rev. (2012)

The Nobel Prize in Physics 2005



Photo: J.Reed
Roy J. Glauber
Prize share: 1/2



Photo: Sears.P.Studio
John L. Hall
Prize share: 1/4



Photo: F.M. Schmidt
Theodor W. Hänsch
Prize share: 1/4

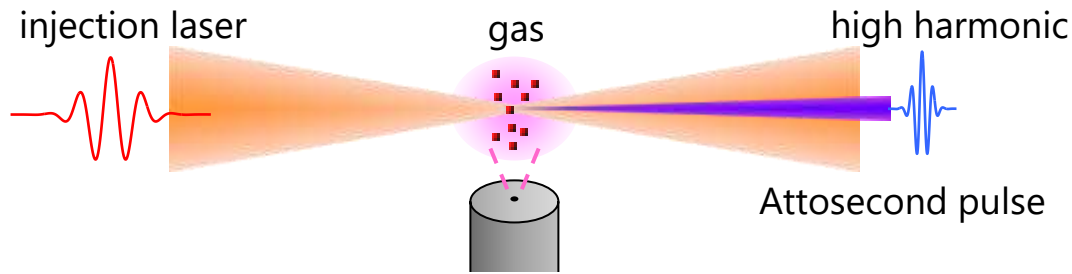
Optical frequency combs

Spectrum made of discrete and regularly spaced lines.
Allows a direct link from radio frequency standards to optical frequency.

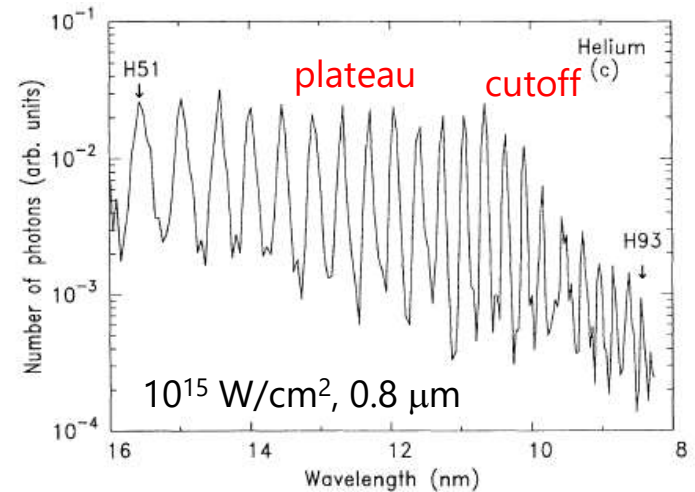
Typical applications.

Precision timekeeping and metrology. [A.D. Ludlow et al., Rev. Mod. Phys. 87, 637 \(2015\).](#)
Spectroscopy. [I. Coddington et al., Phys. Rev. Lett., 100, 013902 \(2008\).](#)

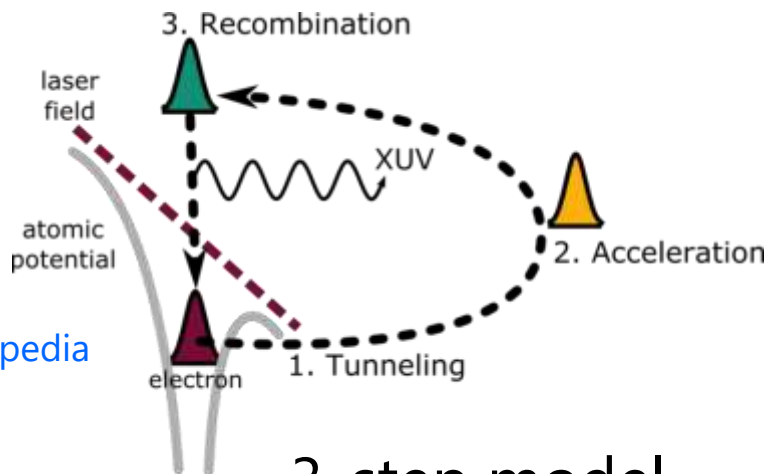
High-Harmonic Generation (HHG)



odd-harmonics with time and spatial coherence



Wahlström et al., *Phys. Rev. A* 48, 4709 (1993)



3-step model

1. tunneling ionization by E_{laser}
2. acceleration of the electron by E_{laser}
3. recombination with the parent atom

The Nobel Prize in Physics 2023

Attosecond science

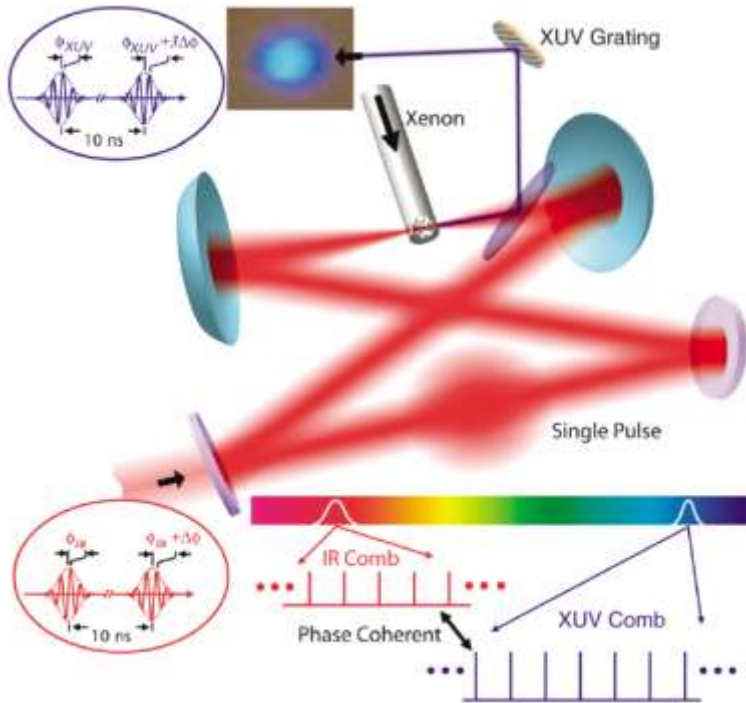


III. Niklas Elmehed © Nobel Prize Outreach
Pierre Agostini

III. Niklas Elmehed © Nobel Prize Outreach
Ferenc Krausz

III. Niklas Elmehed © Nobel Prize Outreach
Anne L'Huillier

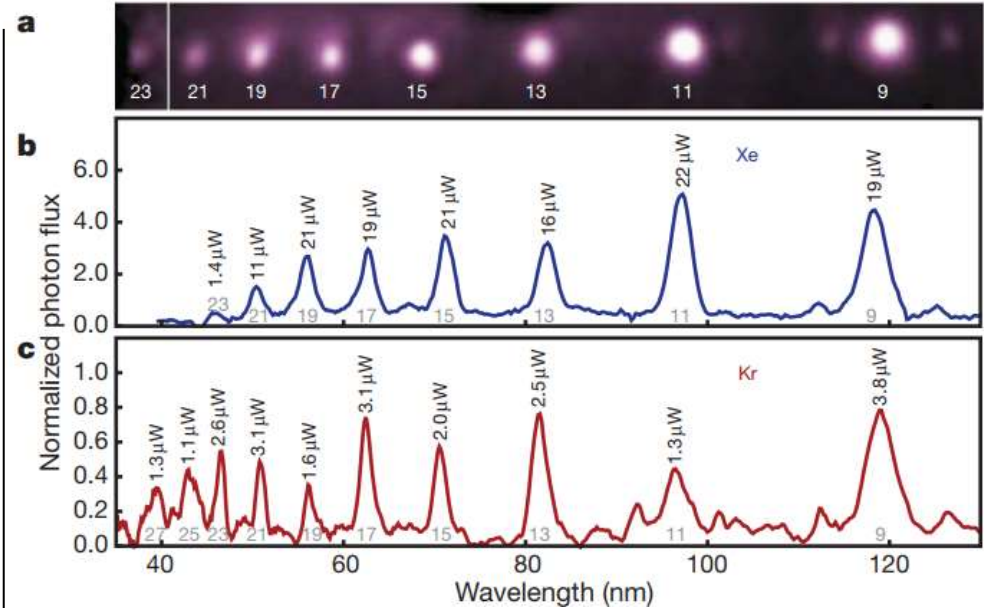
VUV Frequency Combs by HHG



R.J. Jones et al., PRL 94, 193201 (2005).

Demonstration of VUV frequency combs.

The comb structure of the IR laser is fully preserved in the HHG process.



Injection laser power = 30 W
 Intracavity power = 8 kW
 Rep rate = 154 MHz

The combs are powerful enough to determine the absolute frequency of an Ar transition at 82 nm and a Ne transition at 63 nm.

A. Cingoz et al., Nature 482, 68 (2012).

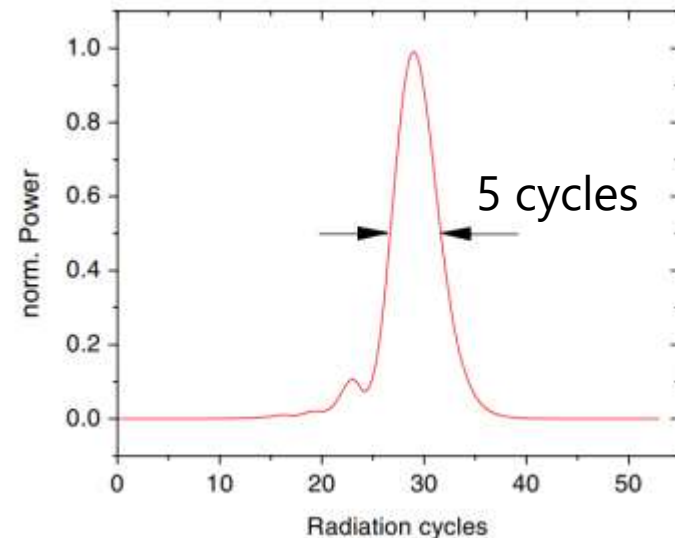
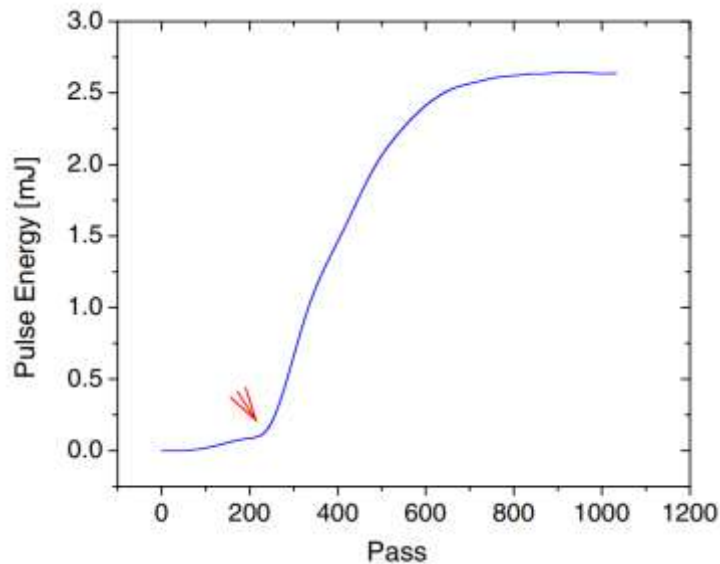
High power coupled midinfrared free-electron-laser oscillator scheme as a driver for up-frequency conversion processes in the x-ray region

M. Tecimer

THz-FEL Group, University of Hawaii at Manoa, Honolulu, Hawaii 96822, USA

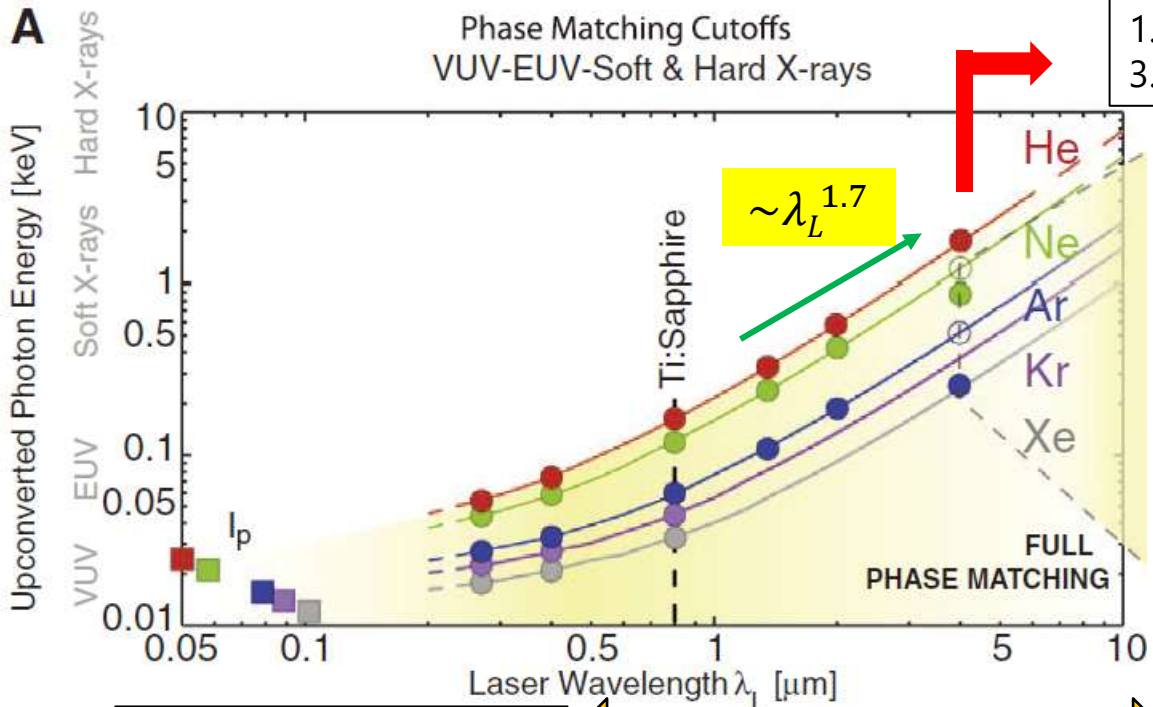
(Received 19 June 2011; published 27 February 2012)

6- μm , 5-cycle, 2.5-mJ FEL pulses from 100-MeV, 80-pC bunches



The FEL pulse can be applied for HHG from a gas target. Utilization of ERL was also discussed.

Advantages of FEL as a HHG driver



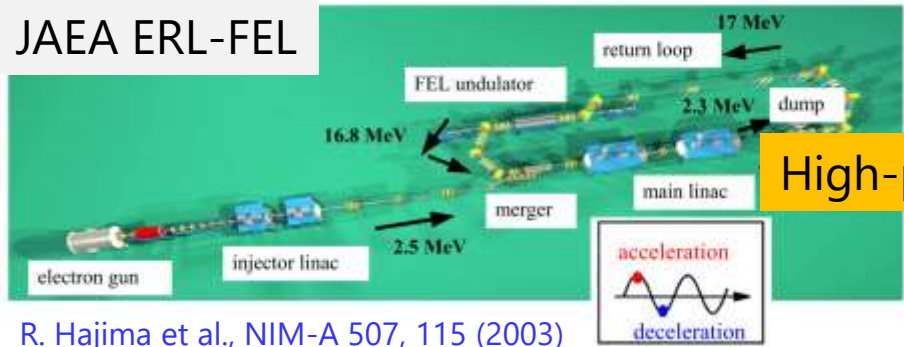
1.6 keV HHG
3.9 μm , 20 Hz solid-state laser

T. Popmintchev et al., Science (2012).

Mid- and long-wave IR pulses are necessary for increasing HHG energies

Wavelength tunability

JAEA ERL-FEL

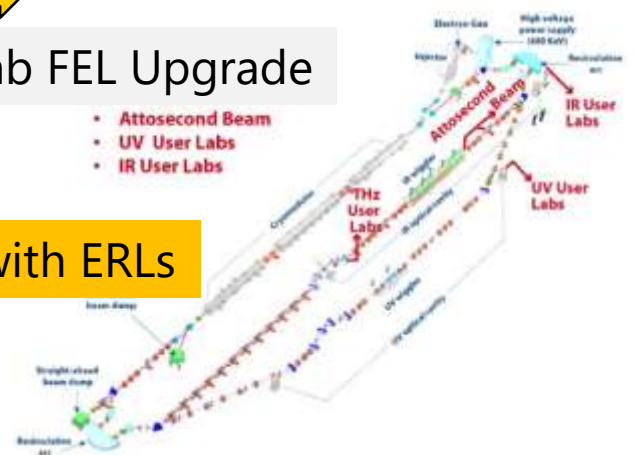


High-power FELs with ERLs

R. Hajima et al., NIM-A 507, 115 (2003)
N. Nishimori et al., FEL-2006 (2006).

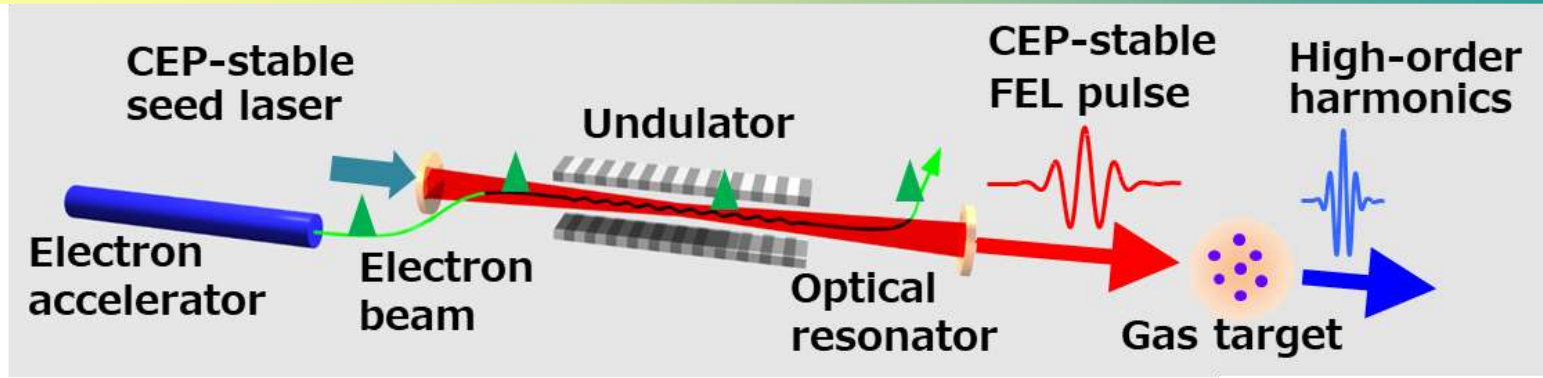
Jlab FEL Upgrade

- Attosecond Beam
- UV User Labs
- IR User Labs



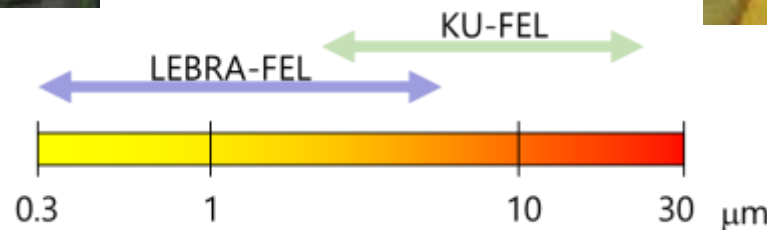
G.R. Neil et al., NIM-A 557, 9 (2006). 7

Research program of FEL-HHG

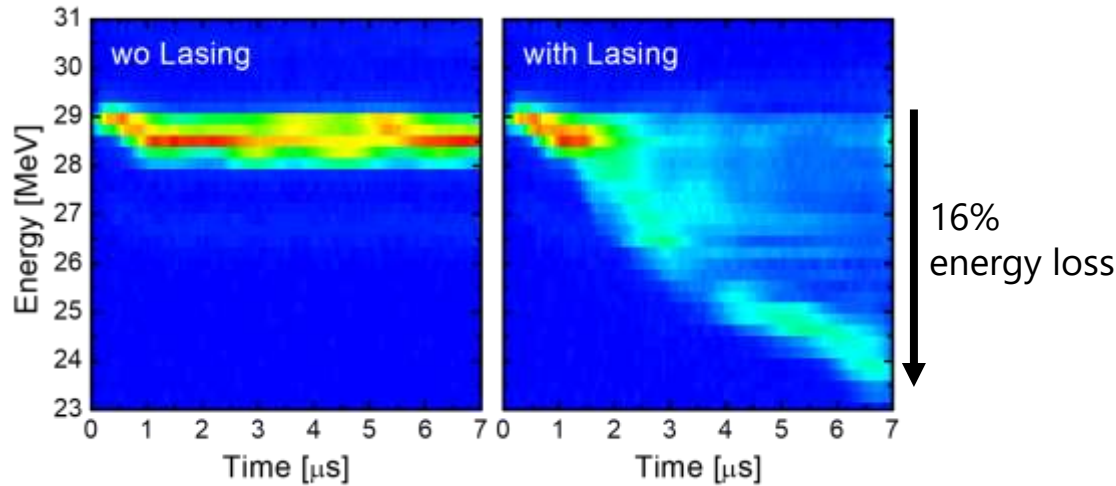


R. Hajima and R. Nagai, PRL 119, 204802 (2017)

A research program has been funded (2018-2027).
We utilize normal-conducting FELs for PoP experiments.



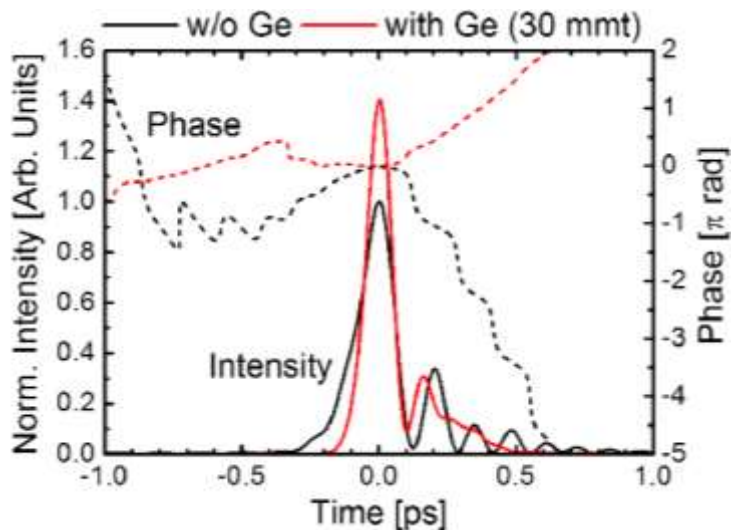
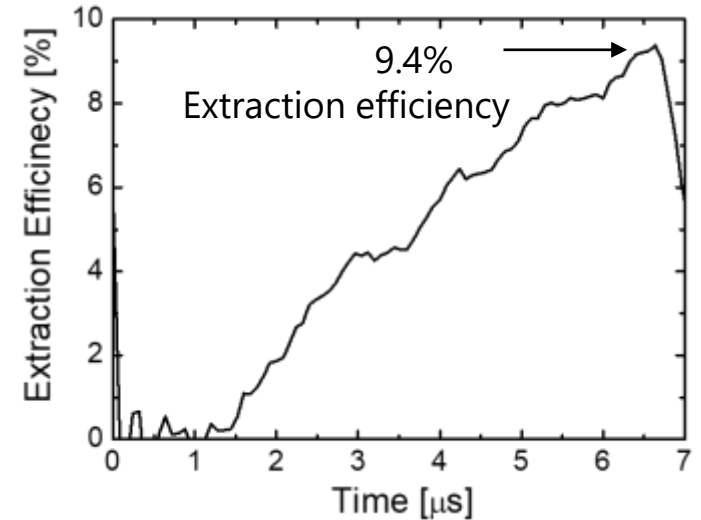
High-efficiency and short pulse at Kyoto U. FEL



Electron energy in a macro pulse

H. Zen, R. Hajima, H. Ohgaki, APEX (2021)

New record of efficiency!

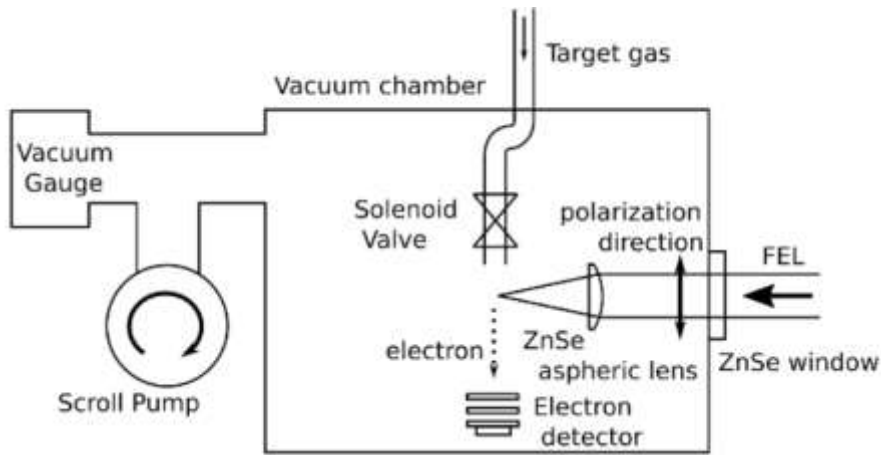


"Few-cycle" lasing

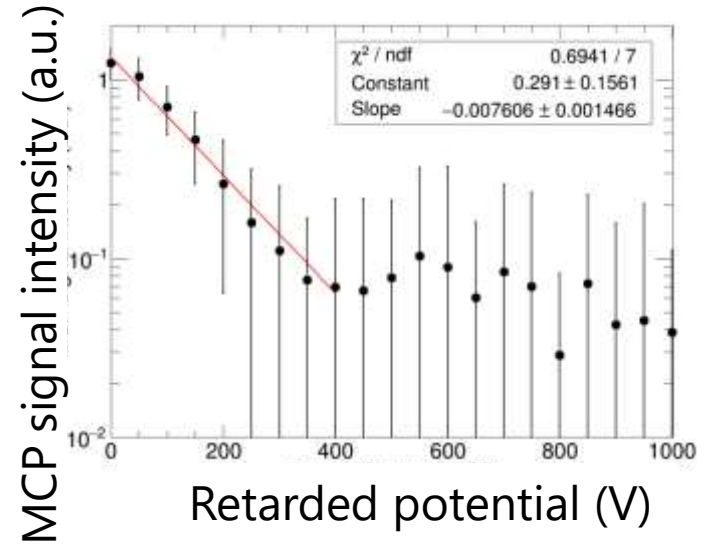
Nonlinear pulse compression by a Ge rod.
5.1 → 3.7 cycle @ 8.7 μm, 40 μJ.

H. Zen, R. Hajima, H. Ohgaki, Optics Express (2023)

Experiment, $\lambda = 9 \mu\text{m}$ @ KU-FEL



Miniature electron detector
-- Retarding field energy analyzer



Electron yield vs retarding potential (N_2 at 0.2 Pa)

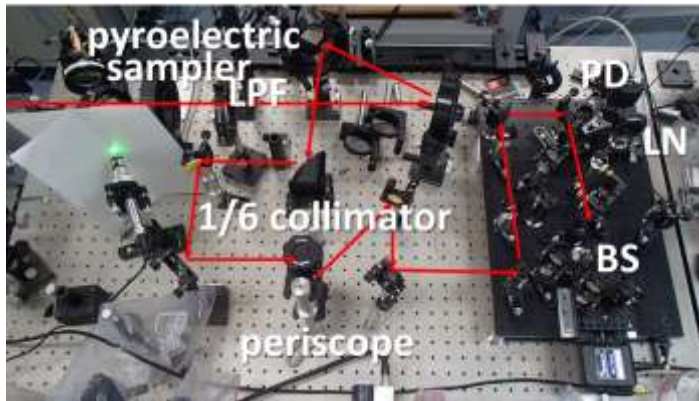
Ponderomotive energy = $563 \pm 37 \text{ eV}$

Laser focal intensity, $I = 7.5 \times 10^{13} \text{ W/cm}^2$

Can be applied to HHG experiments.

K. Kawase et al., Proc. PASJ-2023.

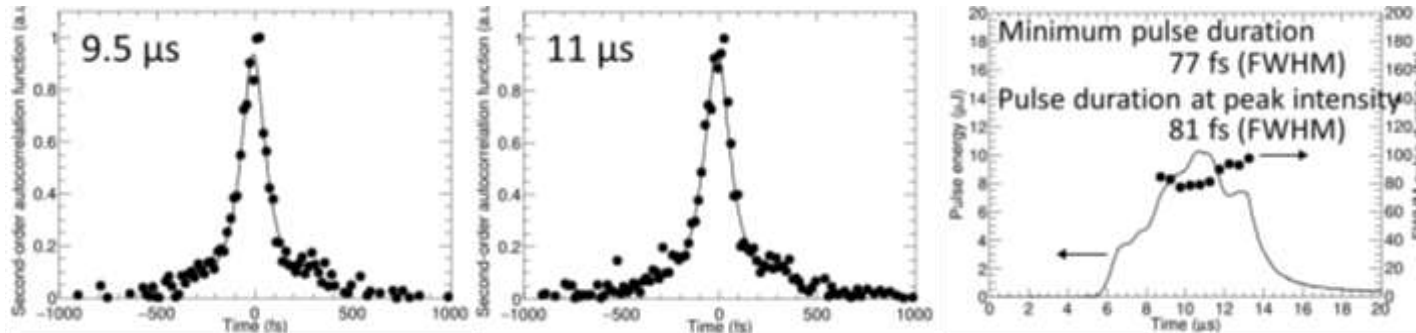
Pulse measurement at LEBRA-FEL



Experiment, $\lambda = 2 \mu\text{m}$ @ LEBRA-FEL

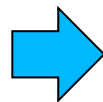
K. Kawase et al., JJAP (2024).

Pulse length measurement by SHG autocorrelation



From a knife-edge measurement

$w = 4.4 \mu\text{m}$ (horizontal)
 $w = 4.3 \mu\text{m}$ (vertical)

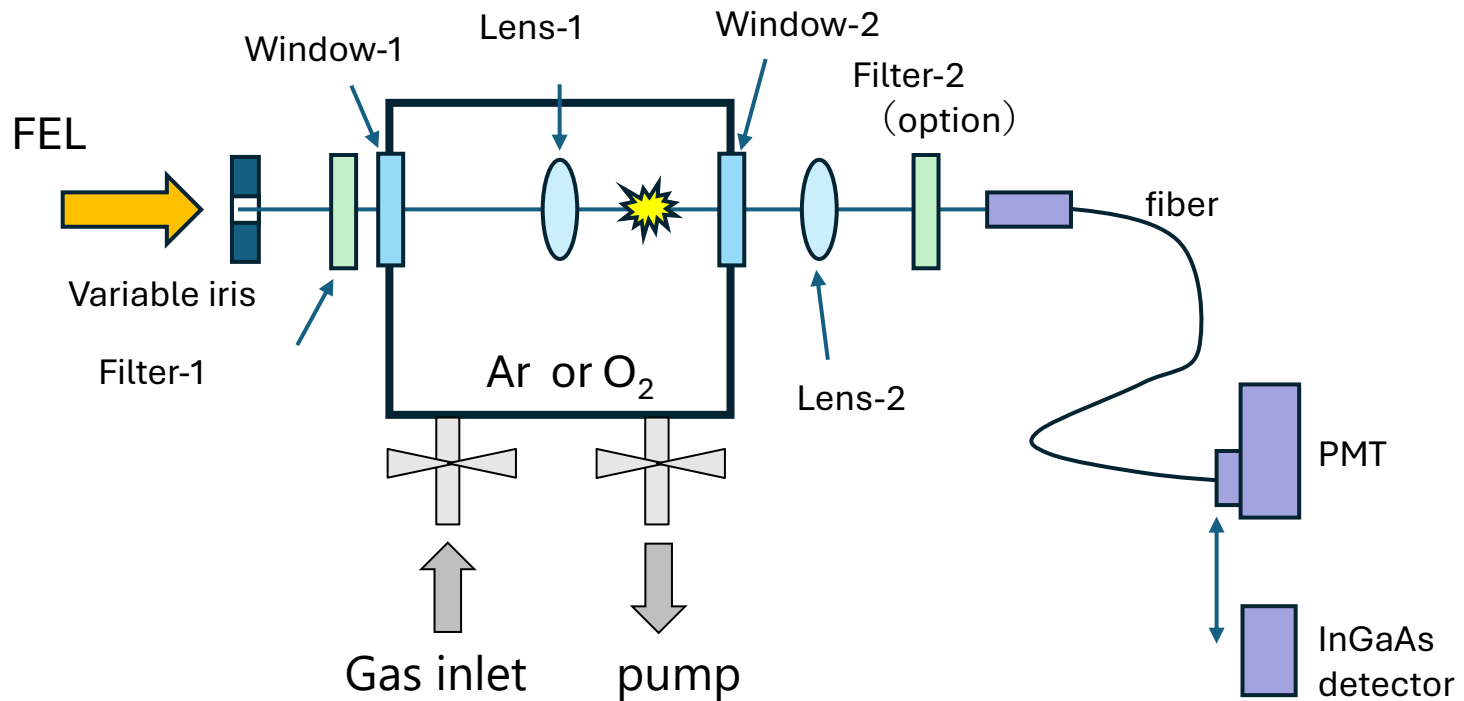


Max. pulse energy	$10.2 \pm 0.6 \mu\text{J}$
Pulse length (FWHM)	$81.2 \pm 0.3 \text{ fs}$
Peak power	$126 \pm 8 \text{ MW}$
Intensity	$(4.2 \pm 0.6) \times 10^{14} \text{ W/cm}^2$
Ponderomotive energy	158 eV



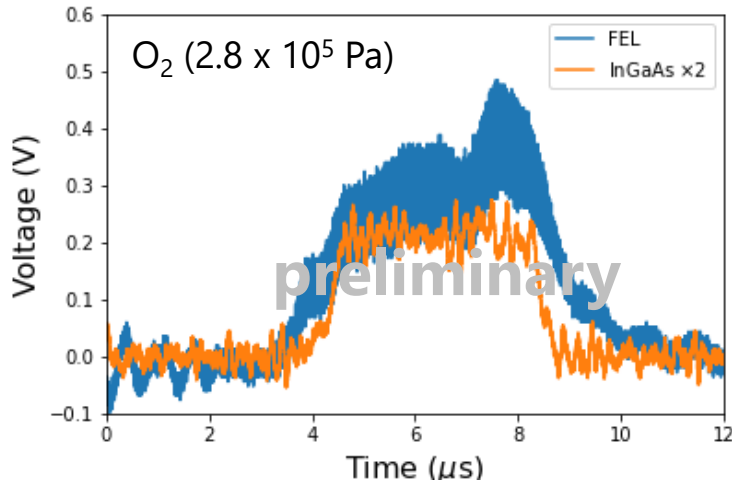
Can be applied to HHG experiments.

HHG experiments at KU-FEL and LEBRA-FEL

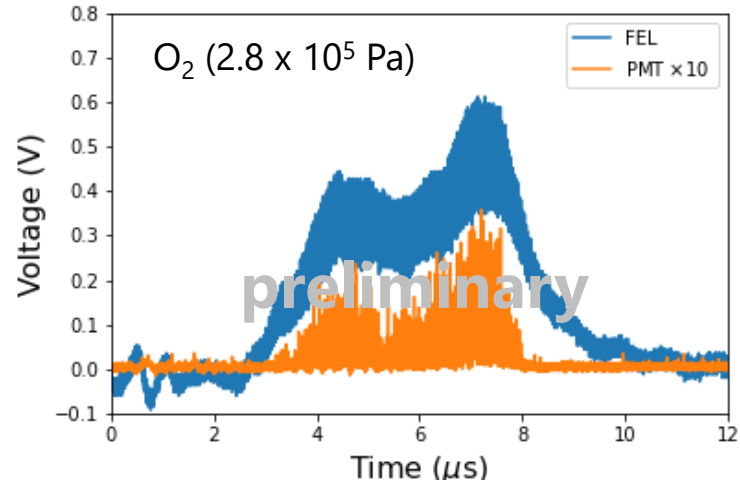


	Filter 1	Window-1	Lens 1	Window 2	Lens 2	fiber
KU-FEL (5 μm)	LPF 4500 nm	CaF ₂	ZnSe f=12.5mm	Sapphire	CaF ₂ f=35 or 50 mm	Use
LEBRA-FEL (2 μm)	LPF 1650 nm	CaF ₂	CaF ₂ f=25 or 35 mm	CaF ₂	Not use	Not use

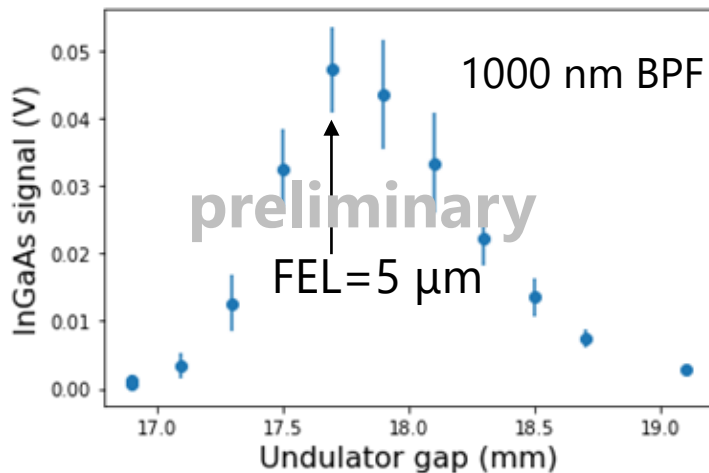
Experiment, $\lambda = 5 \mu\text{m}$, 29.75 MHz @ KU-FEL



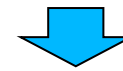
5th harmonic measured by InGaAs
(1000 nm BPF)



7th harmonic measured by PMT
(752 nm BPF)

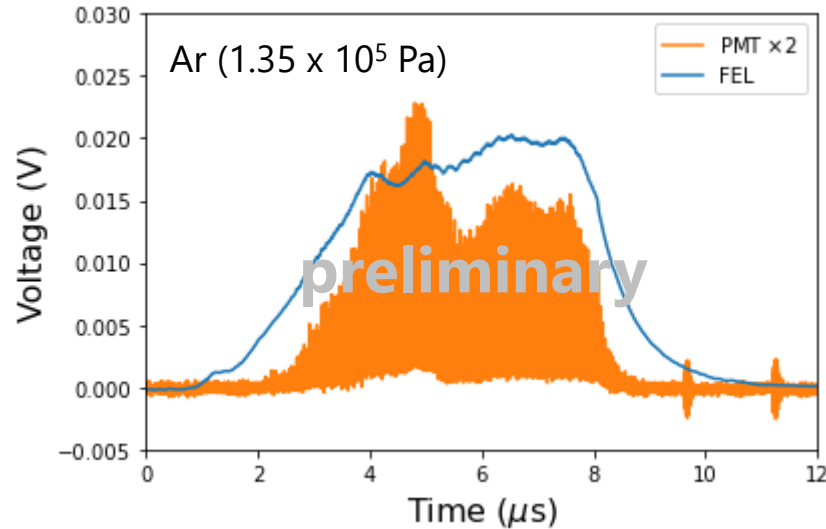


- ✓ The signal varies with the undulator gap.
- ✓ The signal is sensitive to the upstream iris aperture \rightarrow phase matching



HHG from oxygen molecules

Experiment, $\lambda = 2 \mu\text{m}$, 44.625 MHz @ LEBRA-FEL



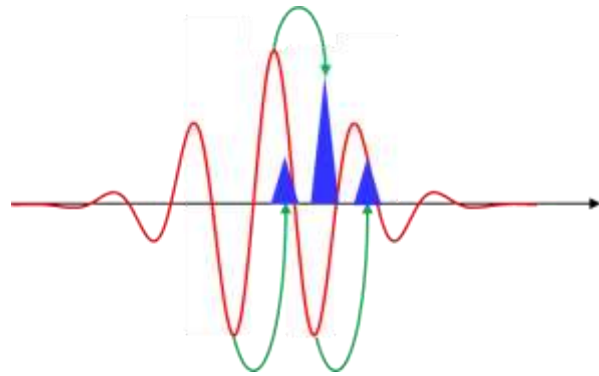
3rd and 5th harmonics
(PMT with SPF 850 nm)

The emission has a linear polarization parallel to the incident FEL.



HHG from argon atoms

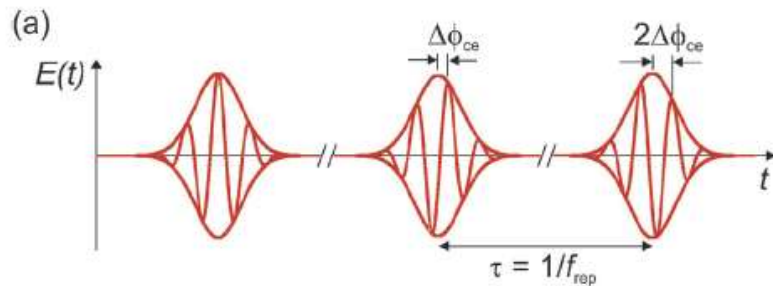
Carrier-Envelope Phase (CEP)



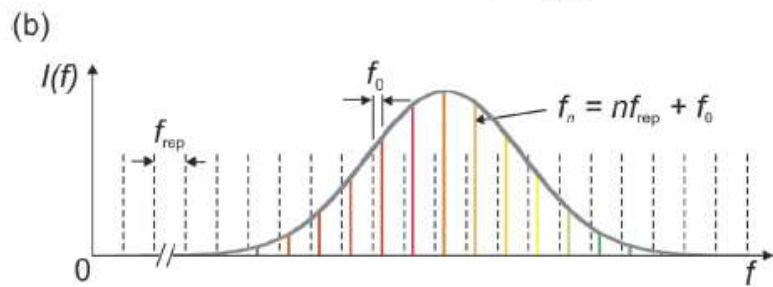
HHG is governed by laser oscillating electric field.

Carrier-envelope phase (CEP) affects the properties of HHG, cut-off energy, spectrum and yield.

CEP must be stabilized for a practical use of HHG.

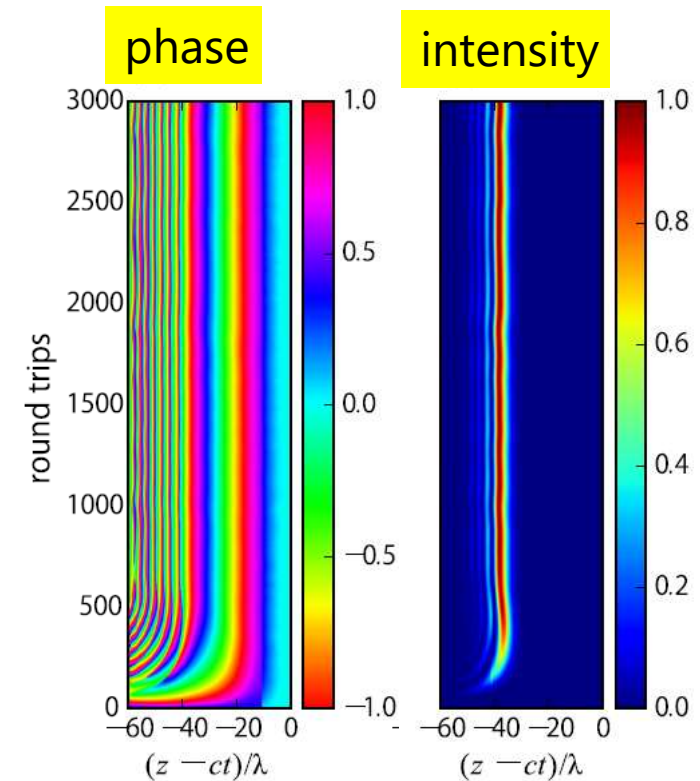
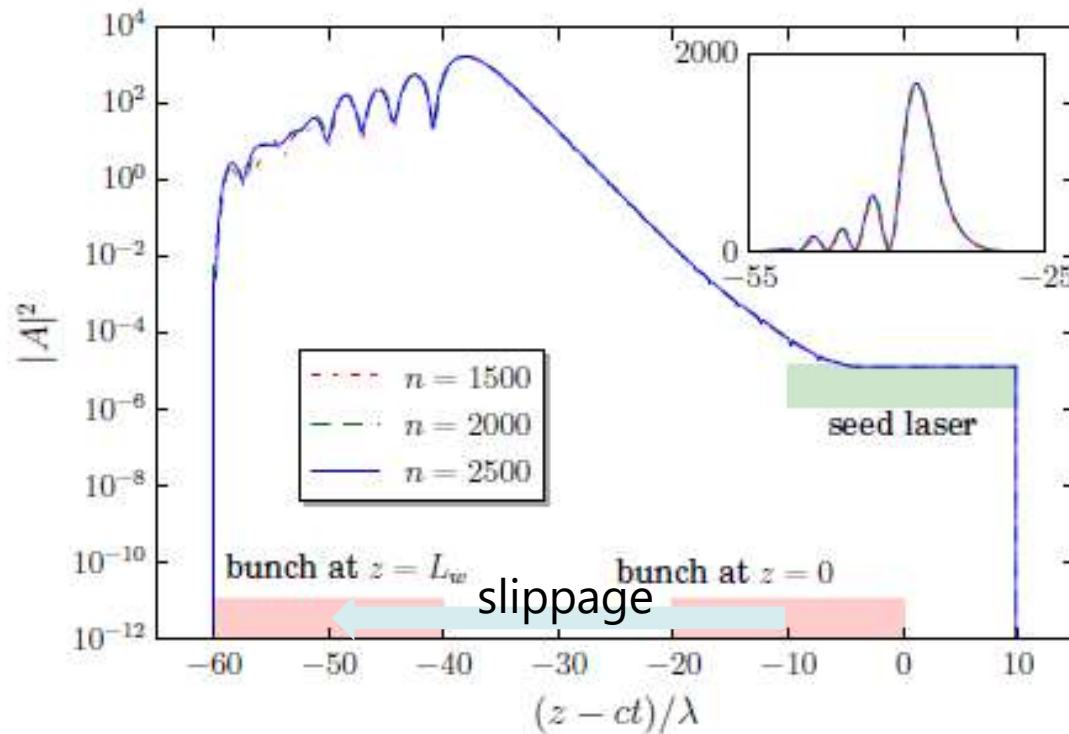


For the optical frequency combs, CEP must be stabilized.



Can a FEL generate CEP-stable pulses?

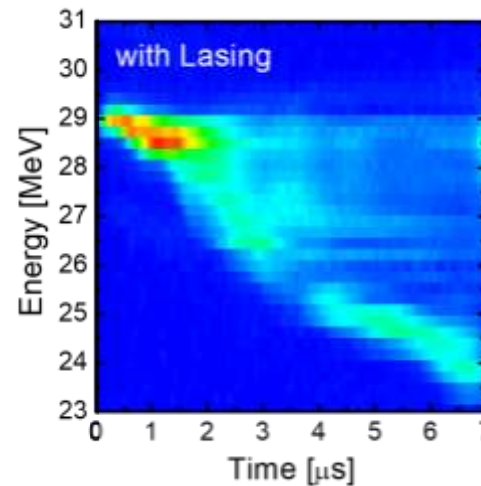
CEP stabilization by an external seed laser



- Introducing a CEP-stabilized seed pulse to overlap the shot-noise part
- The leading edge of FEL pulse has a fixed amplitude and phase.
- The FEL interaction starts with a well-defined optical field.
- Over-all lasing dynamics is stabilized.
- CEP-stabilized few-cycle FEL pulses

Mitigation of a large energy spread

KU-FEL experiment



H. Zen, R. Hajima,
H. Ohgaki, APEX (2021)

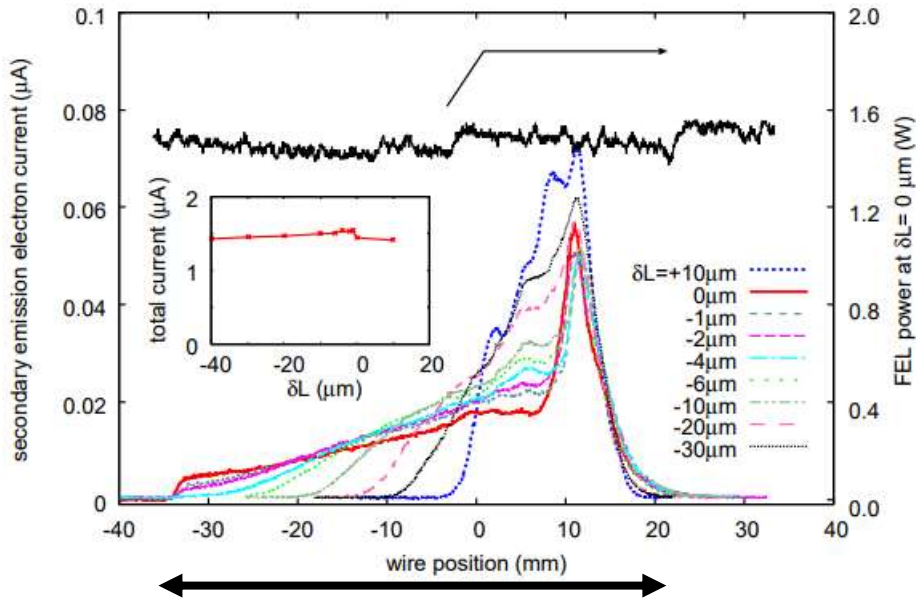
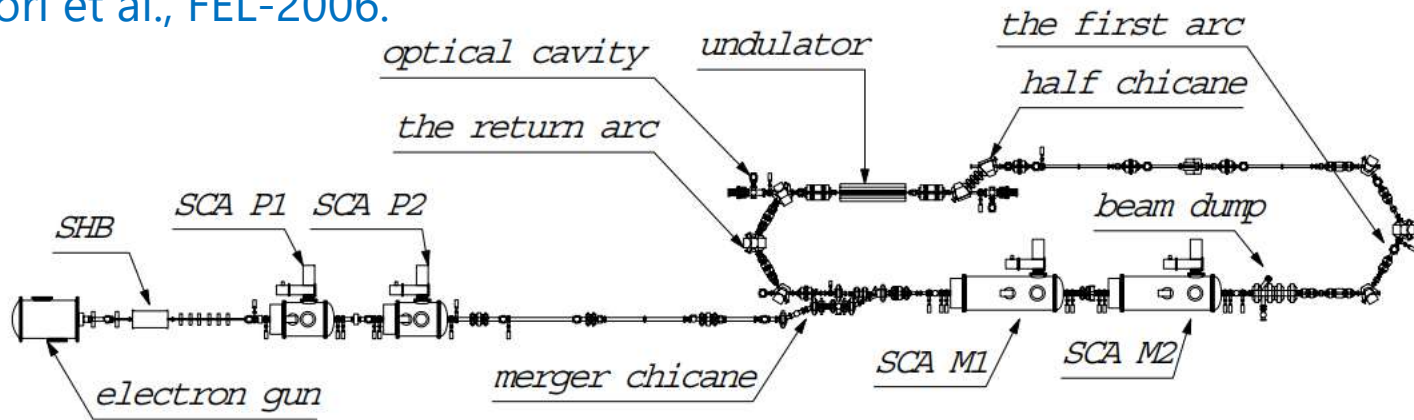
Energy decrease of 16% for an FEL efficiency of 9.4%

A large energy spread is induced in the few-cycle lasing.

Mitigation of the large energy spread is necessary for an ERL-FEL.

Energy spread measurement at JAEA-ERL-FEL

N. Nishimori et al., FEL-2006.



15% energy spread

We measured energy spread by a wire scanner at the return arc.

>15% energy spread for an FEL efficiency of 2.5%.

The energy spread is inconsistent with the KU-FEL experiment.

We need to re-examine the result.

Design Example of an ERL-FEL-HHG

Design parameters for a 4- μm ERL-FEL

Energy	60 MeV
Bunch charge	200 pC
Repetition	20 MHz
Current	4 mA

Bates arc with 15% energy acceptance



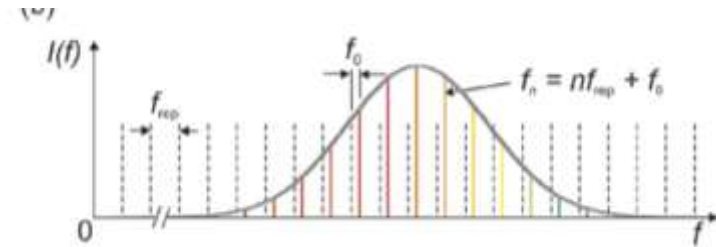
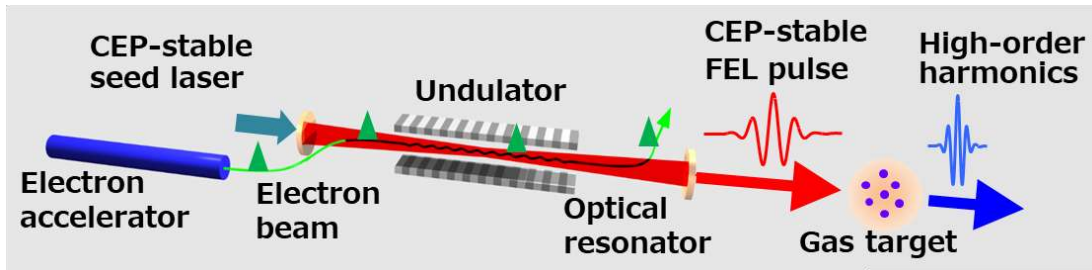
Assuming an extraction efficiency of 5% and an out-coupling efficiency of 50%
FEL pulse energy is

$$60 \text{ MeV} \times 200 \text{ pC} \times 5\% \times 50\% = 300 \text{ } \mu\text{J}$$

$$300 \text{ } \mu\text{J} \times 20 \text{ MHz} = 6 \text{ kW}$$

With an enhancement cavity of finesse ~ 100 , intracavity power $\sim 100 \text{ kW}$.

→ The power is high enough for VUV optical frequency combs applications
and explore beyond VUV frequency combs.



- VUV frequency combs can be realized by HHG from a gas target and have been demonstrated with solid-state lasers at 1 μm .
- FELs operated at mid- and long-wave IR could enable optical frequency combs in VUV and beyond VUV.
- An R&D program to demonstrate FEL-HHG has been launched.
- Harmonic generation from gas atoms was observed at KU-FEL (5 μm) and LEBRA-FEL (2 μm).
- The energy spread is a limitation of the FEL performance when constructing FEL-HHG with an ERL.