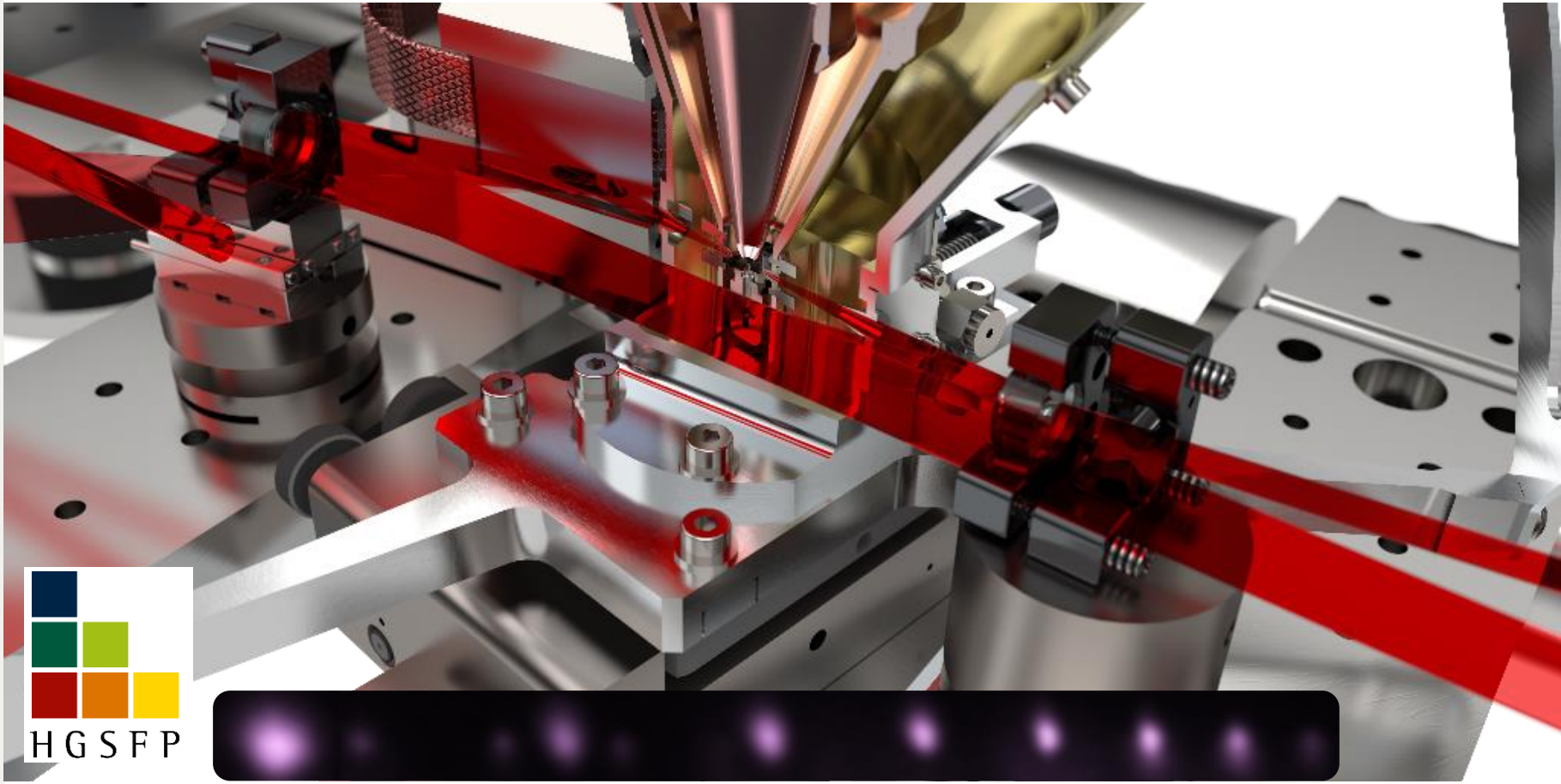
An extreme ultraviolet frequency comb for highly charged ion metrology



Janko Nauta

Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Currently part of:

ALPHA collaboration, Department of Physics, Swansea University Swansea, UK

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HGSFP

TC FS

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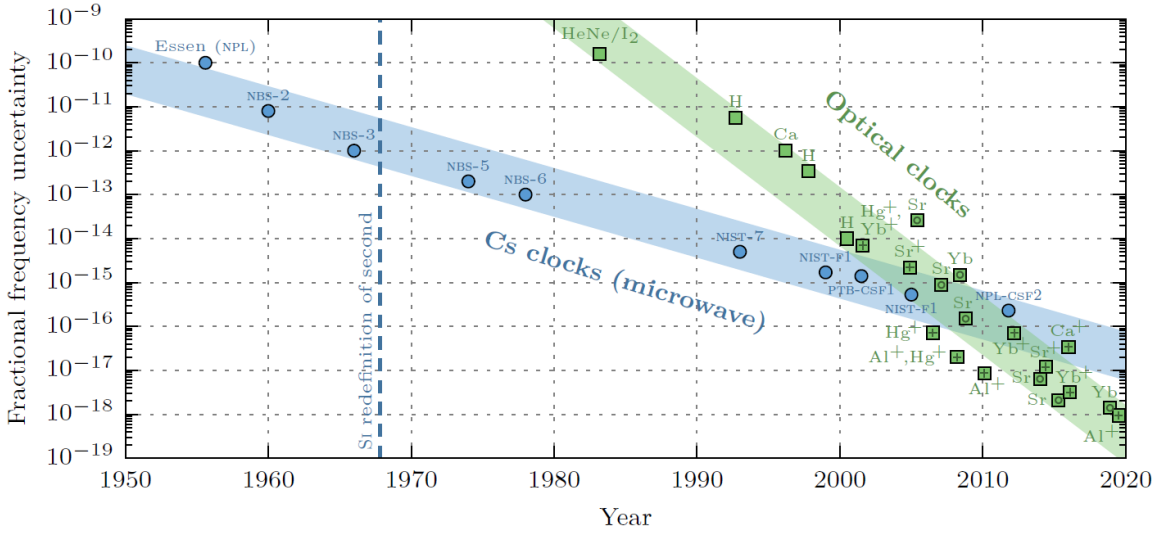
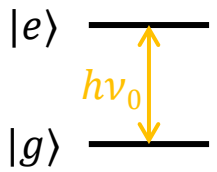
Frequency determinations





Nowadays:
$\Delta\nu/\nu < 10^{-18}$

Alan deviation: $\sigma_y \propto \frac{1}{\nu_0\sqrt{NT_R t}}$

Transition frequency

N – number of references

t – total measurement time

T_R – probe time

Goal: **perform first ultra-high precision laser spectroscopy in the extreme ultraviolet (XUV) region**

Requirements:

1. Atomic system that withstands such high energy radiation: Highly Charged Ions (HCI)

2. Coherent XUV source: XUV frequency comb

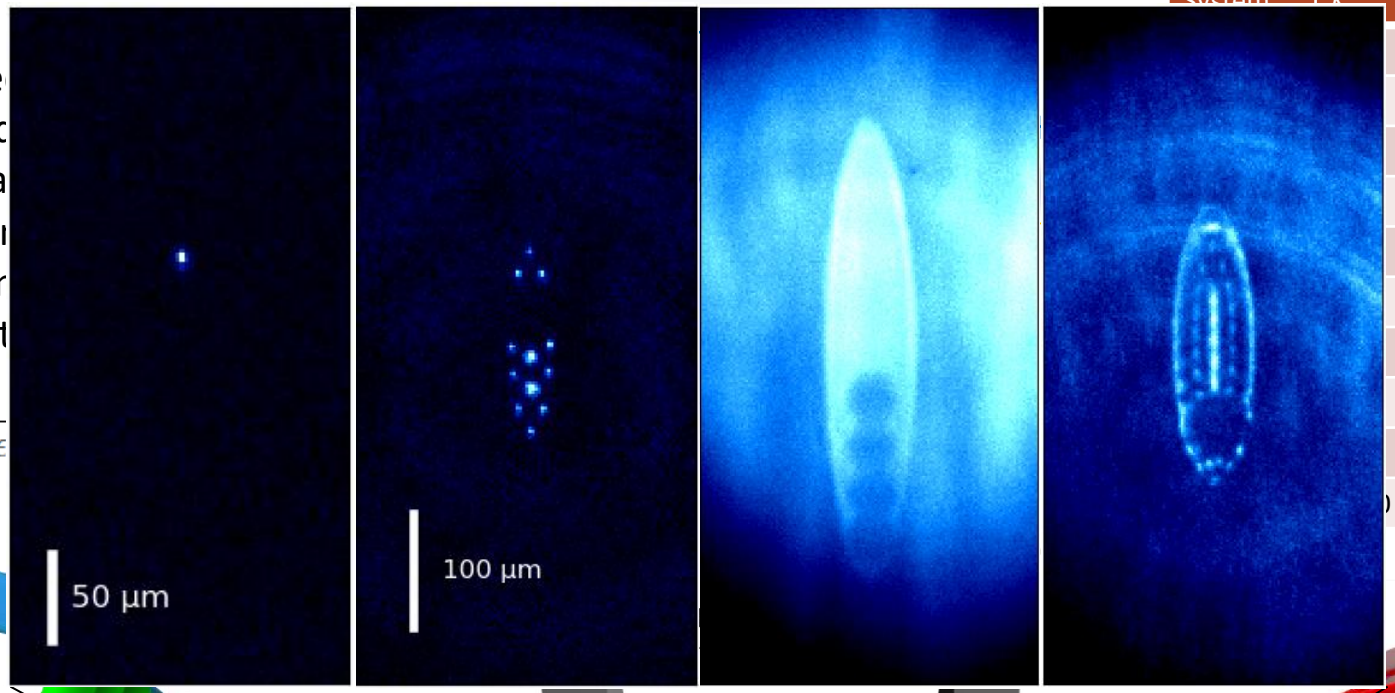
Highly Charged Ions (HCI) for frequency metrology

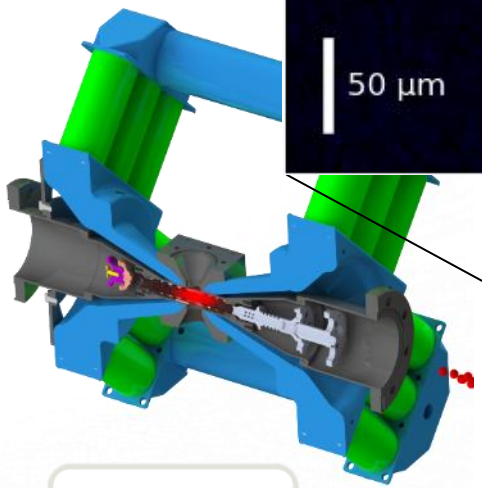
HCI:

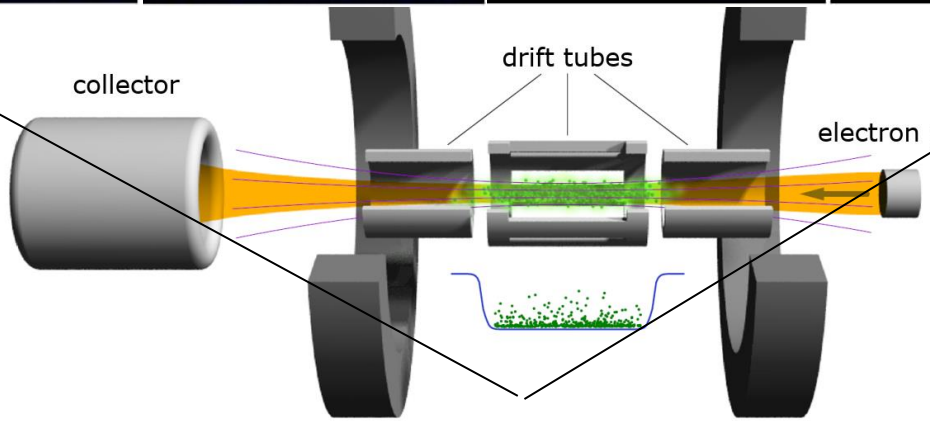
- Propose
  standarc
  inaccura
- Most se
  variatior
  constant

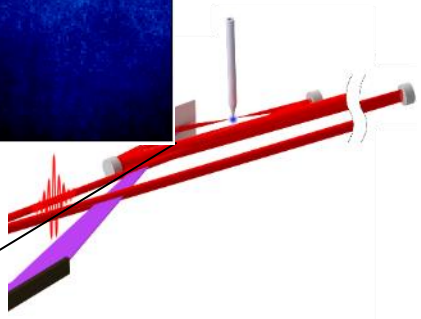
$$\alpha = \frac{1}{4\pi\epsilon}$$

System	K	λ (nm)
		699
		436
		467
		281.5
		267
		ca. 280
		ca. 1980
		ca. 775
		ca. 535
		ca. 150









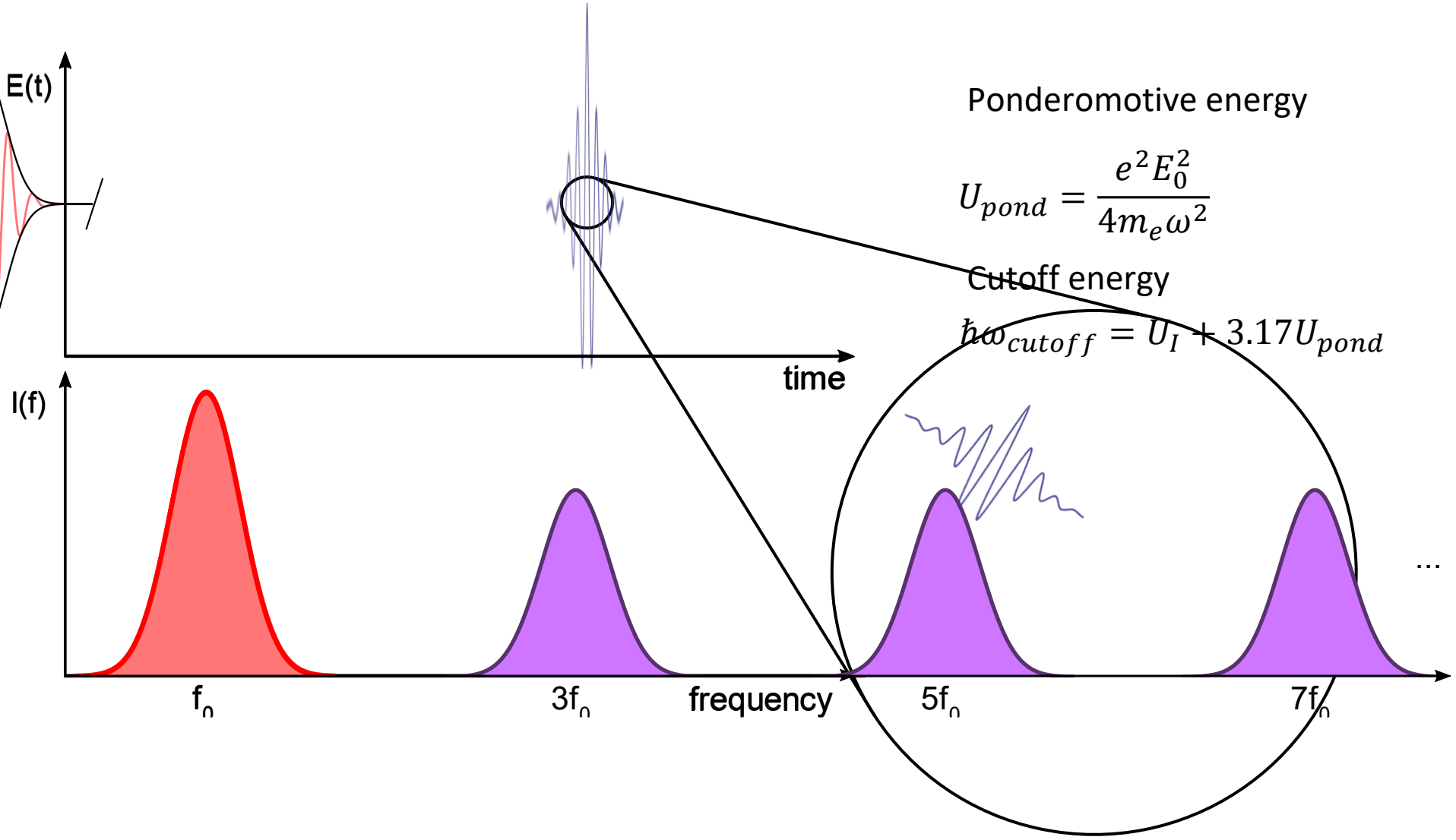
EBIT:
HCI production

dece

P. Micke et al., Nature **578**, 60 (2020)
J. Stark et al., RSI **92,** 083203 (2021)

M. Safronova et al., RMP **90**, 025008 (2018)
L. Schmöger et al., Science **347**, 6227 (2015)

High harmonic generation (HHG)



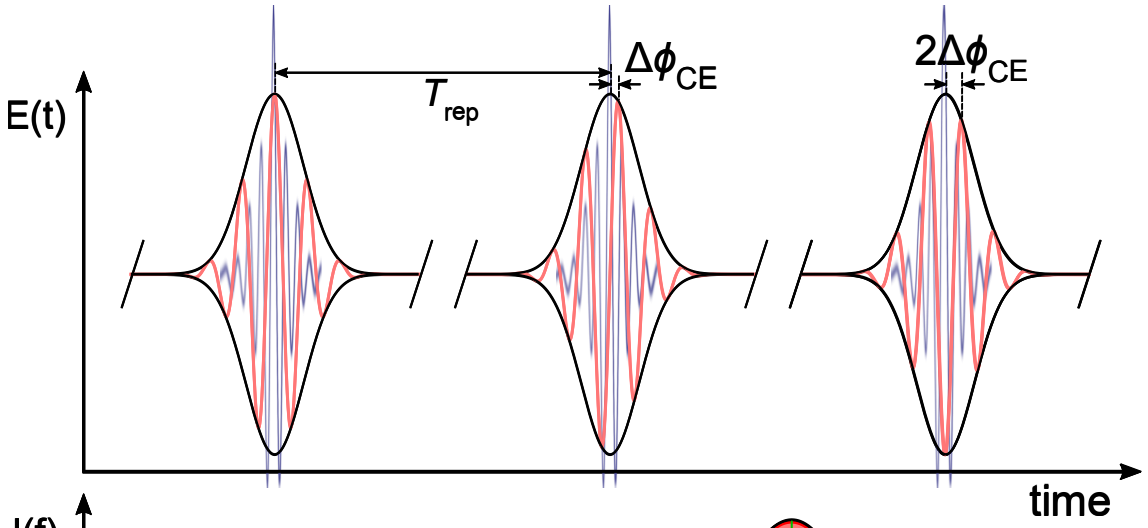
Ponderomotive energy

$$U_{pond} = \frac{e^2 E_0^2}{4 m_e \omega^2}$$

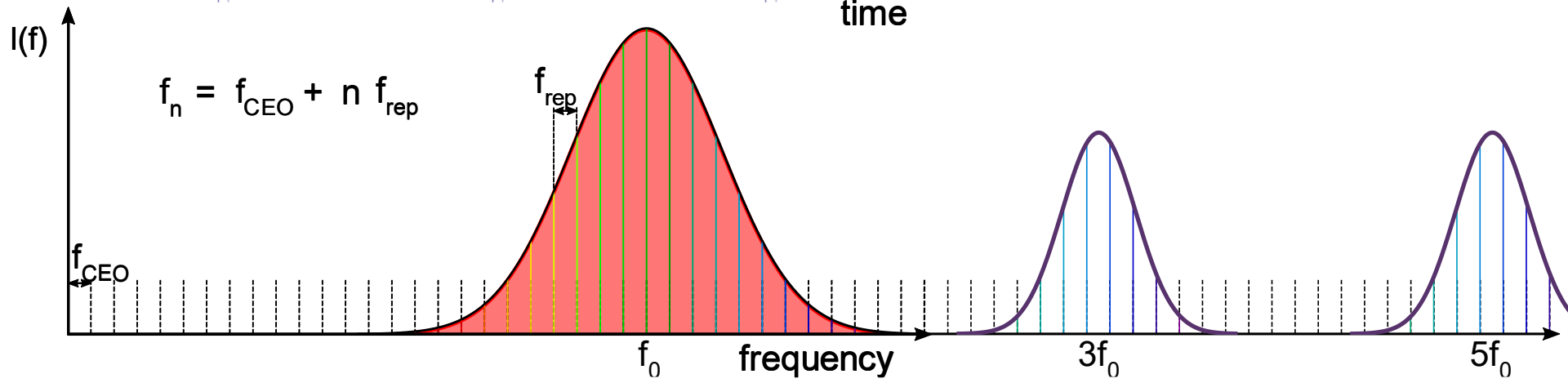
Cutoff energy

$$\hbar\omega_{cutoff} = U_I + 3.17 U_{pond}$$

HHG of a pulse train: XUV frequency comb



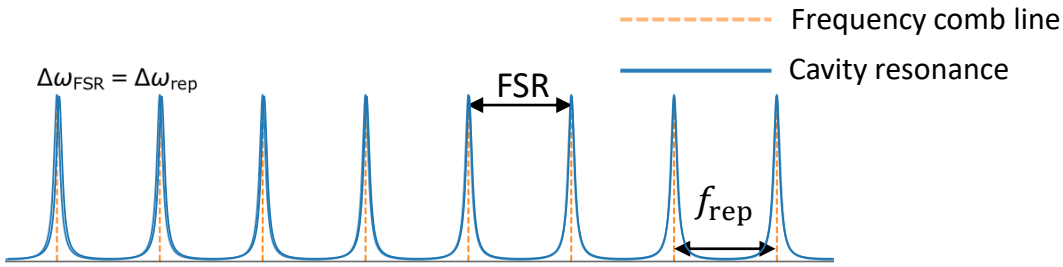
- Generate odd higher harmonics of fundamental light
- Phase coherent transfer of IR comb to XUV



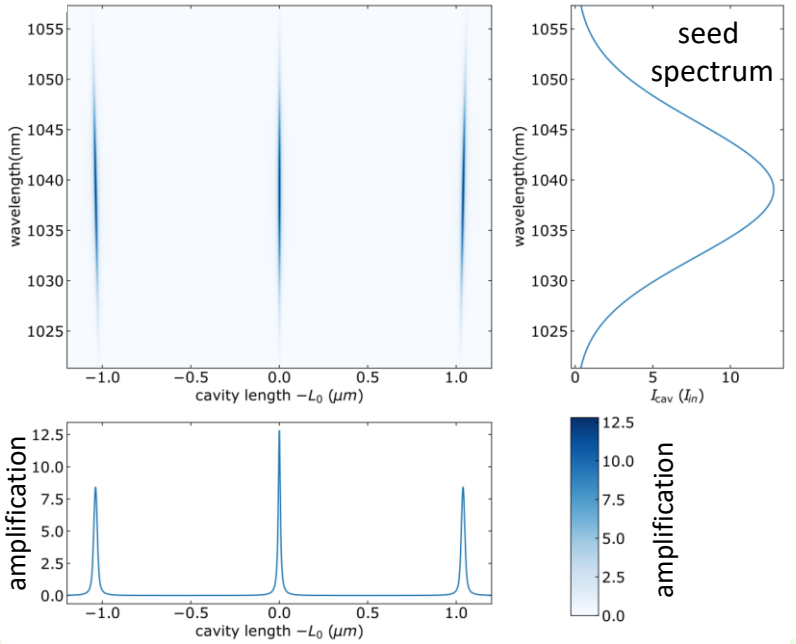
- Laser intensity > 10^{13} W/cm^2 needed in every pulse
  - usually achieved by kHz laser systems, but spectroscopy requires f_{rep}>> 1 MHz

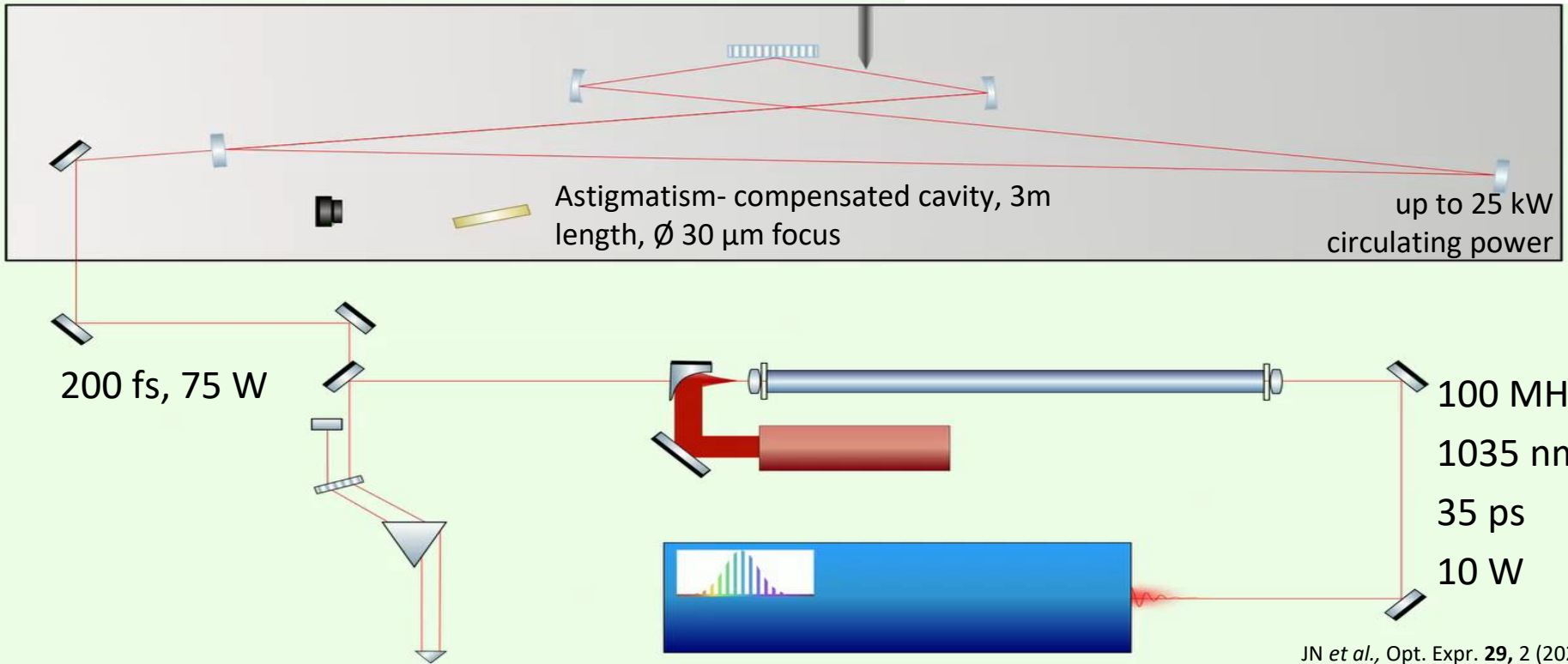
  → additional amplification necessary

Femtosecond enhancement cavity



- Frequency comb repetition rate has to match cavity free spectral range
- Frequency comb offset phase has to match cavity roundtrip phase

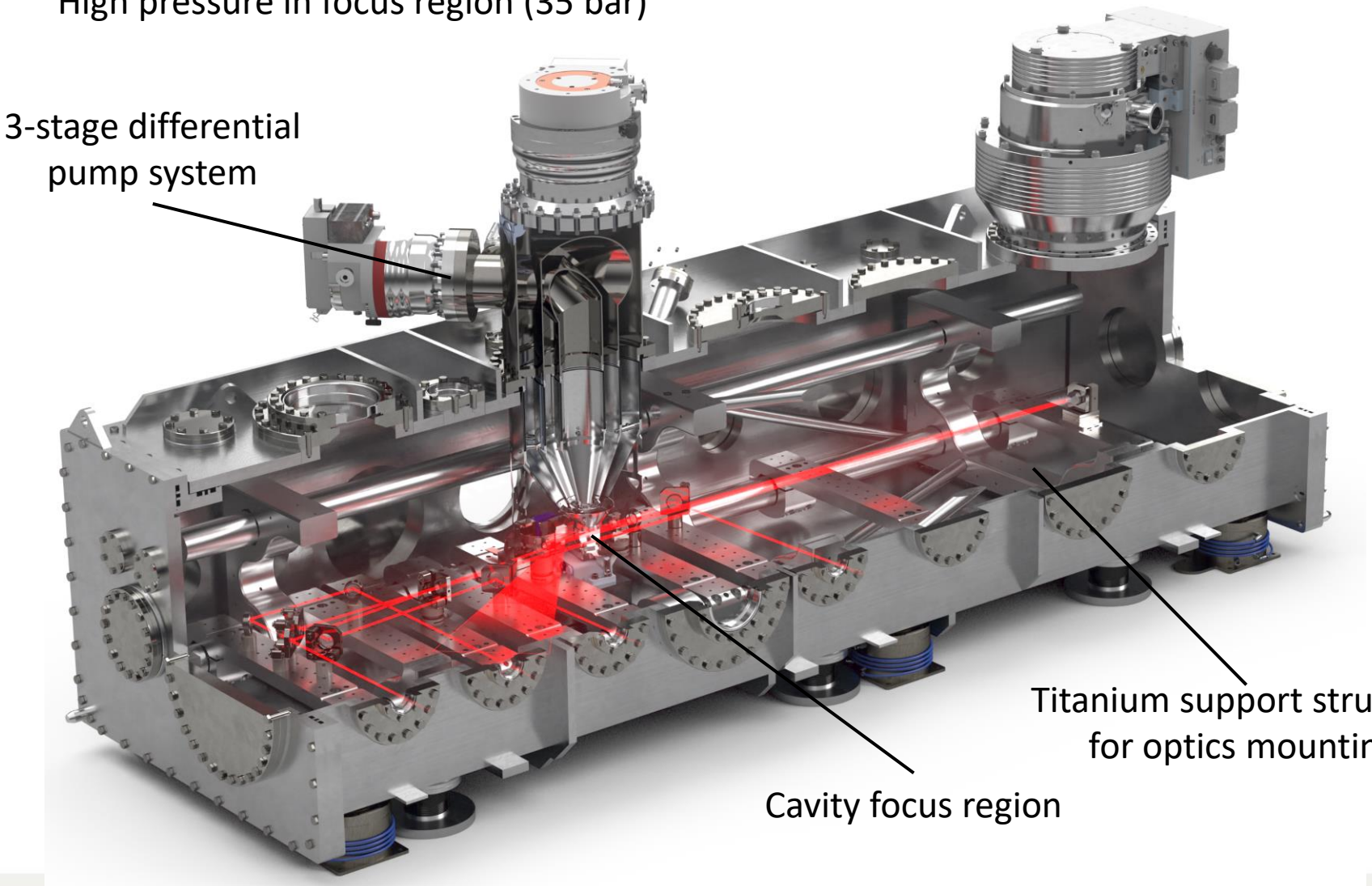




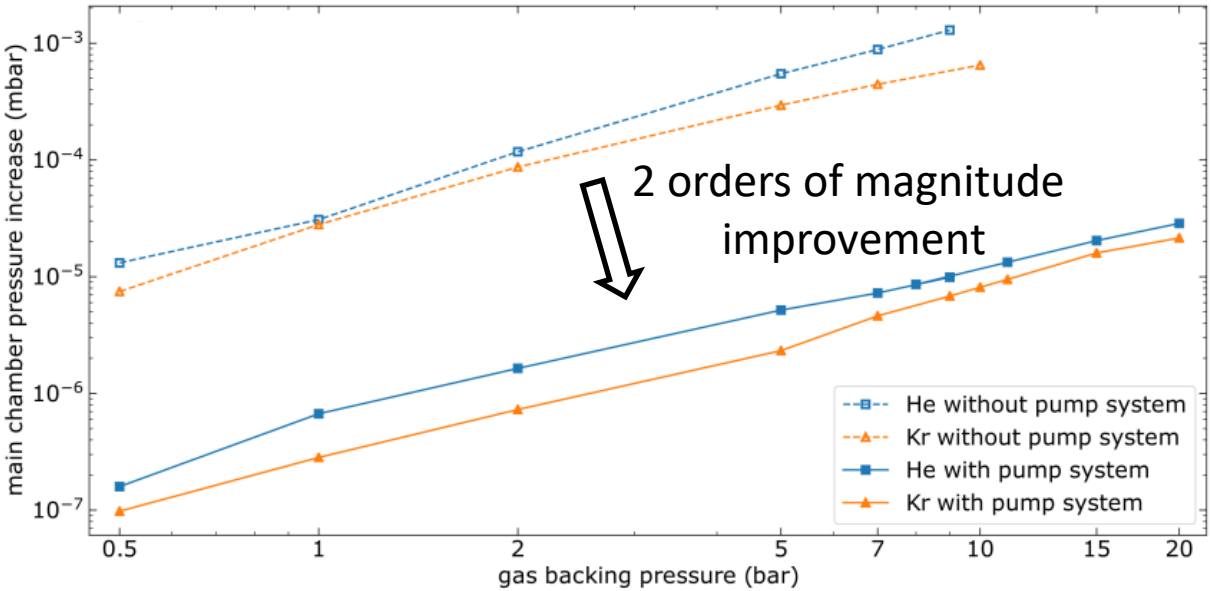
JN *et al.,* Opt. Expr. **29,** 2 (2021)

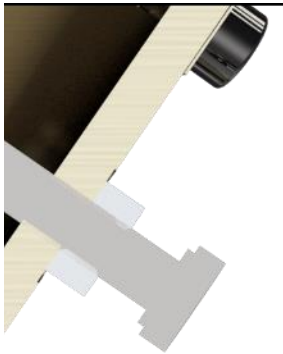
Vacuum system

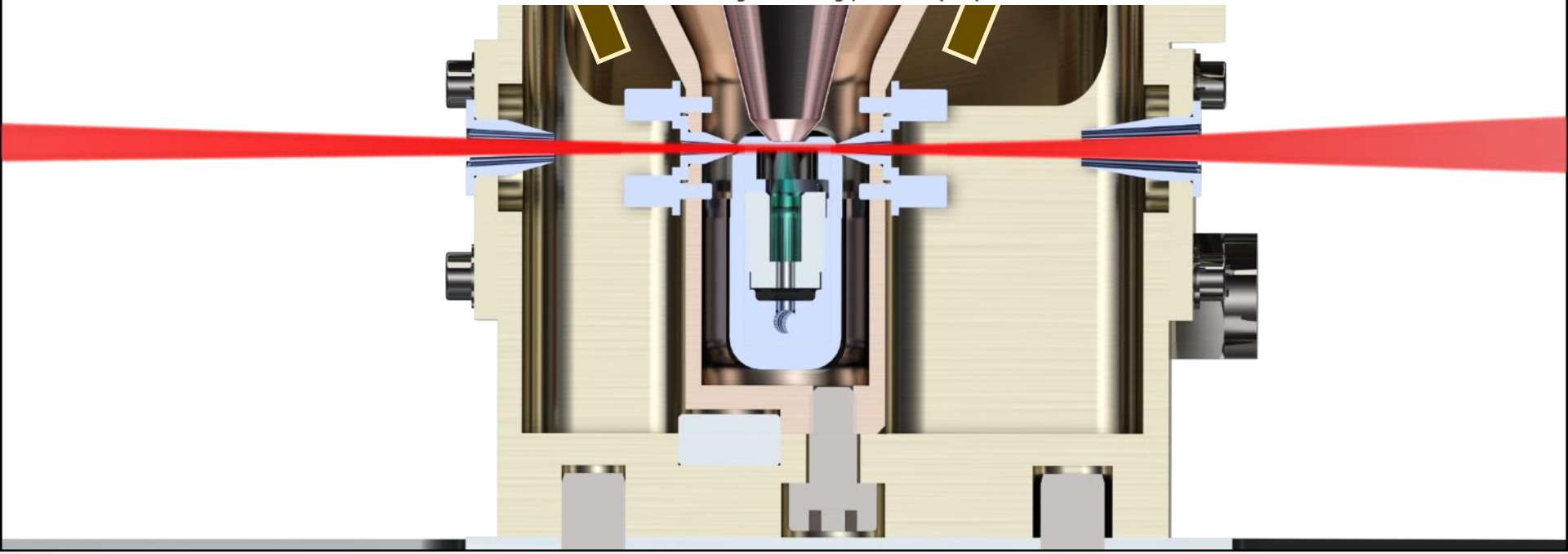
- UHV conditions in chamber (10^{-8} mbar)
- High pressure in focus region (35 bar)



Differential pump structure

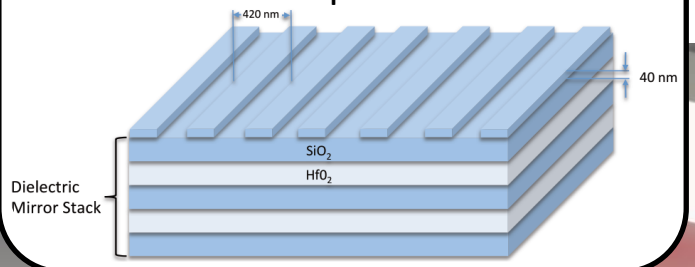


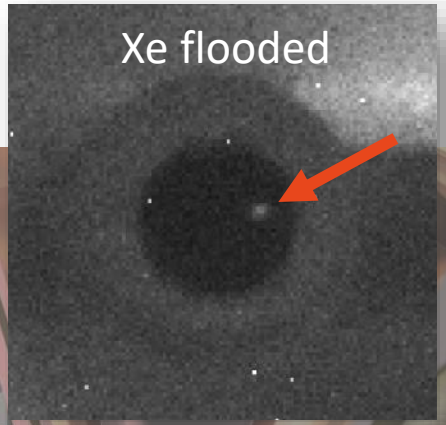


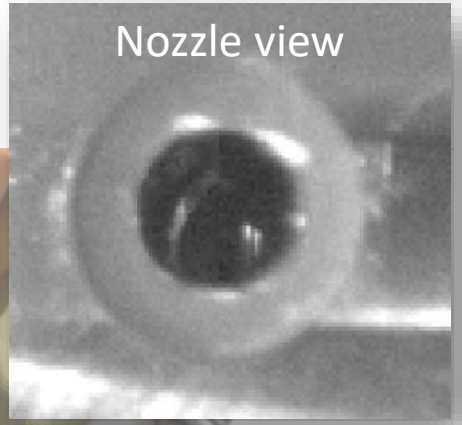


XUV generation & detection

Nanograting etched in top layer of HR mirror separates XUV & IR







grating

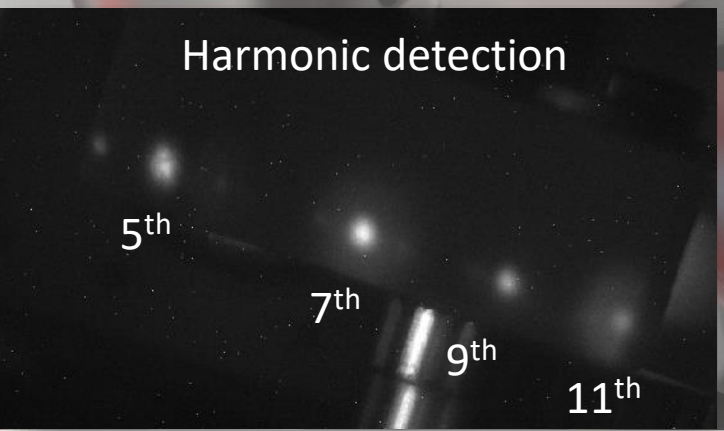
IR

IR + XUV

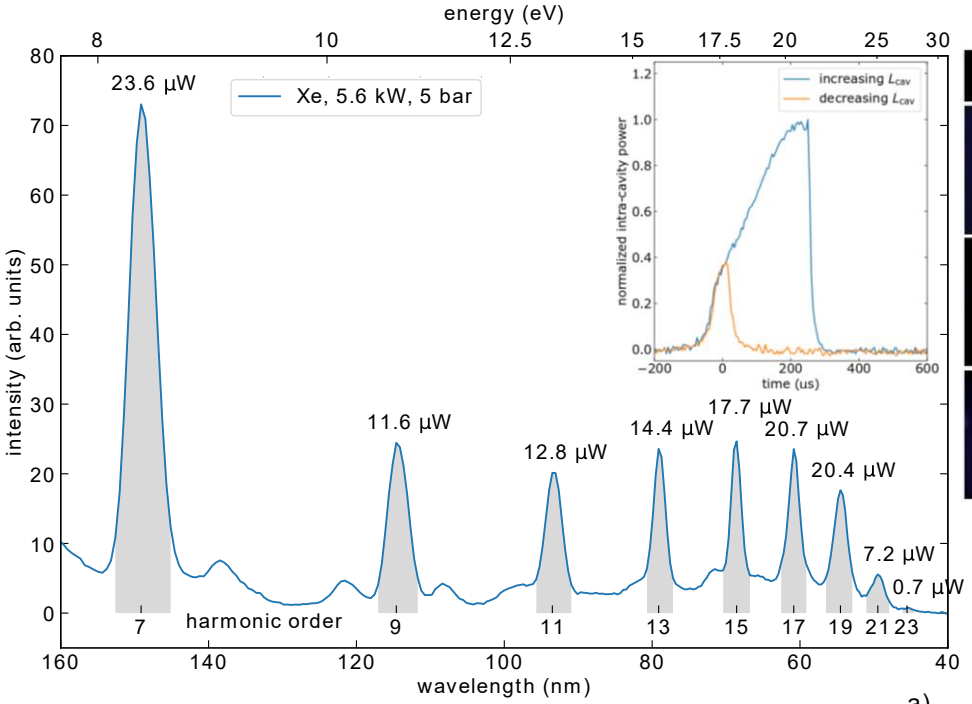
IR

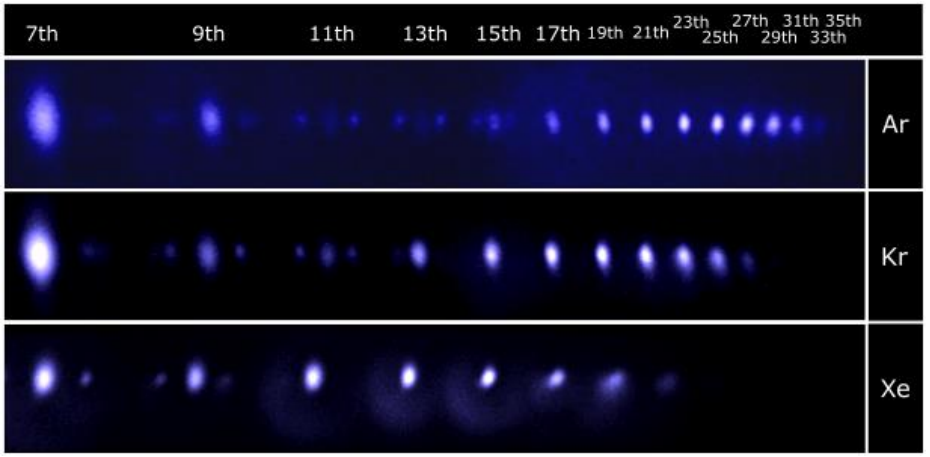
11th
9th
7th
5th
3rd harmonic

XUV



Harmonic generation & phase-matching improvements

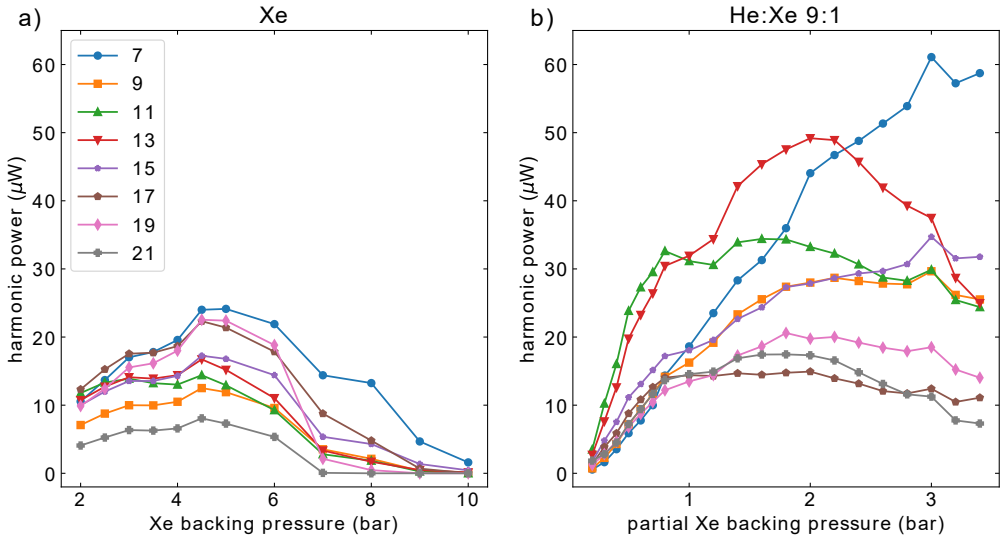




$$\Delta \mathbf{k} = \Delta \mathbf{k_g} + \Delta \mathbf{k_d} + \Delta \mathbf{k_n} + \Delta \mathbf{k_p} = 0$$

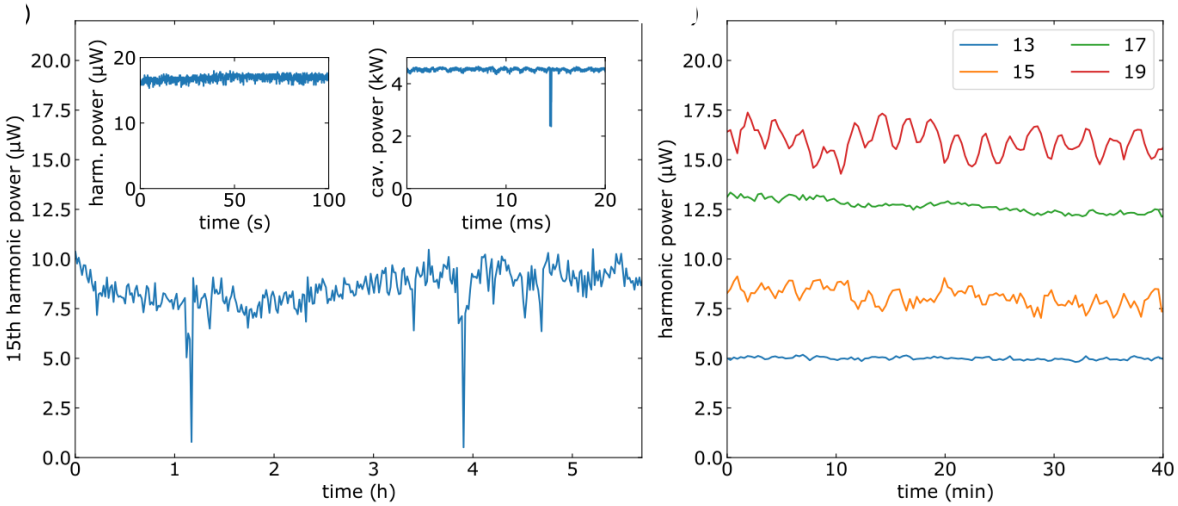
$$v_{\mathrm{gas}} = \sqrt{5RT/M_{\mathrm{avg}}}$$

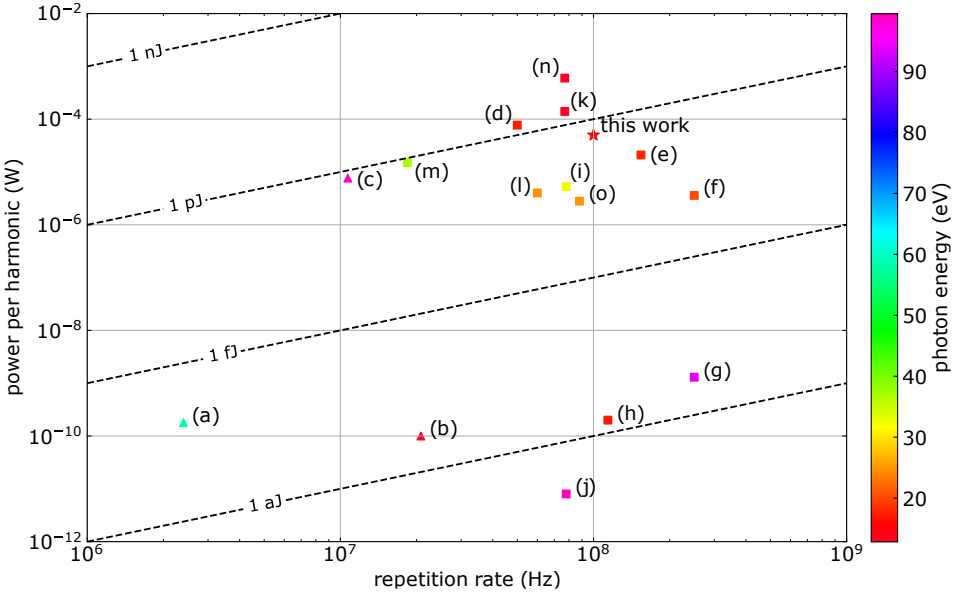
- Cut-off energy increases with ionization potential
- 35th harmonic visible using Argon at 42 eV (30 nm)
- Large steady-state plasma fraction at cavity focus
- Increasing the gas speed by using a He:Xe mixture reduces the amount of plasma
- XUV yield significantly improved



Continuous XUV output

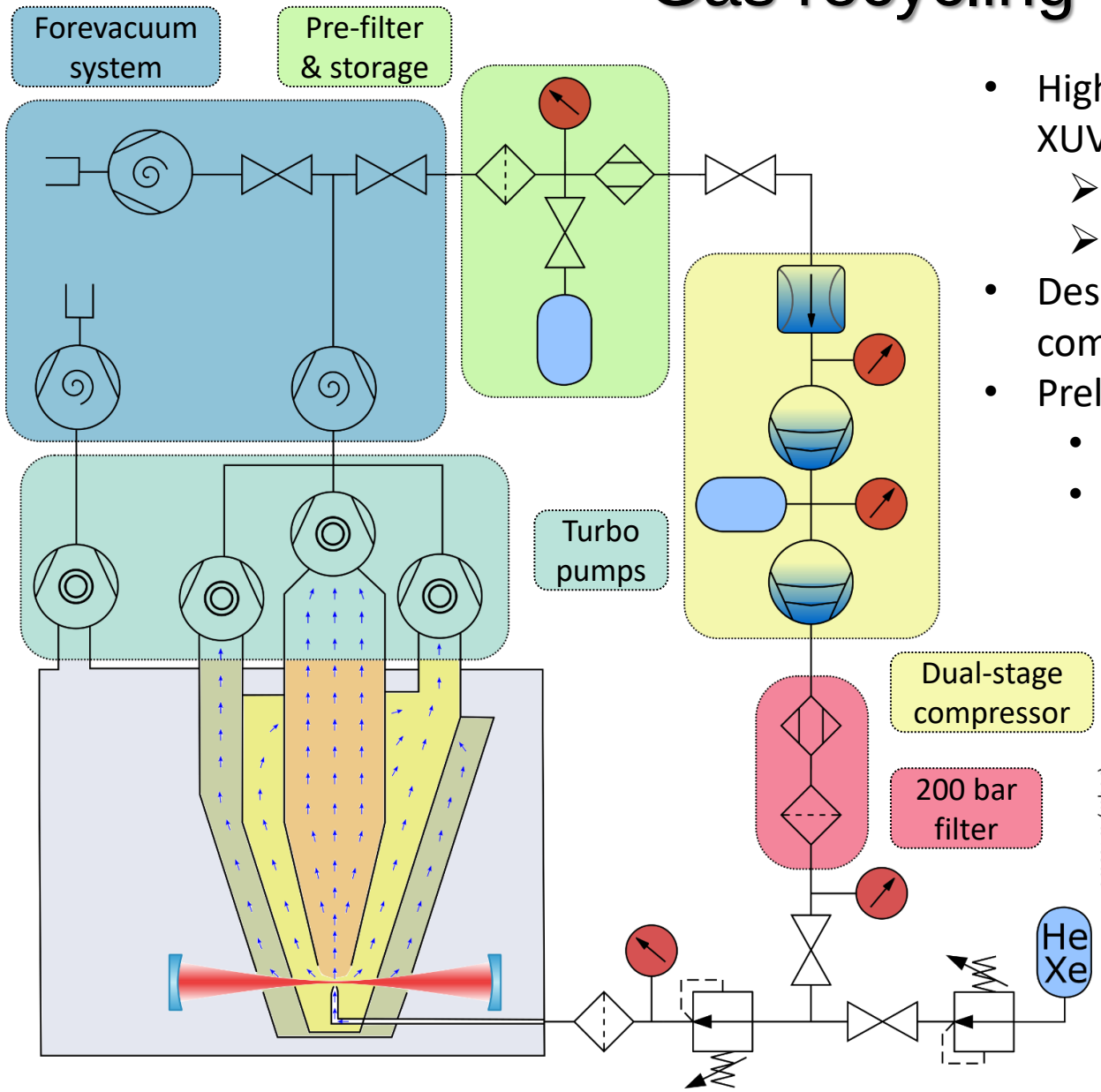
- Continuous operation over 5 hours demonstrated
- No signs of cavity mirror degradation observed
- Newly built system is among the most powerful worldwide
- Sufficient power & stability for first XUV spectroscopy on HCI



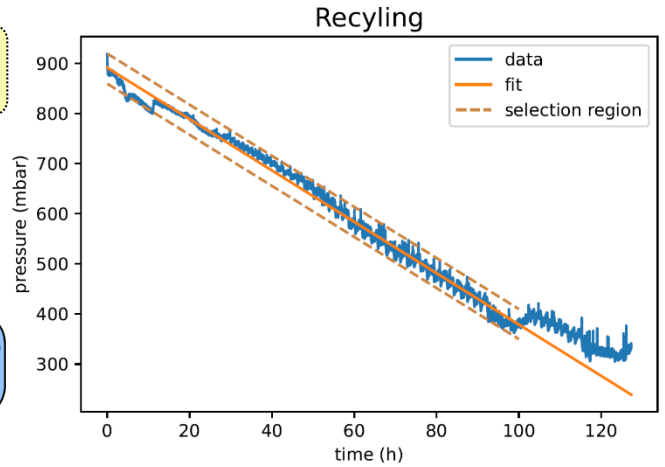


(a) Emaury *et al.*, *Optica* ***2****, 980 (2015)*
(b) Vernaleken *et al.*, Opt. Lett. **36**, 3428 (2011)
(c) Hädrich *et al.*, *Light. Sci. Appl.* ***4****, e320 (2015)*
(d) Lee *et al.*, *Opt. Express* ***19****, 23315-23326 (2011)*
(e) Cingöz *et al.*, *Natutre* ***482****, 68-71 (2012)*
(f) and (g) Carstens *et al.*, *Optica* ***3****, 366 (2016)*
(h) Gohle *et al.*, *Nature* ***436,*** *234-237(2005)*
(i) and (j) Pupeza *et al.*, *Nat. Photonics* ***7****, 608-612 (2013)*
(k) Porat *et al.*, *Nat. Photonics* ***12****, 387-391 (2018)*
(l) Mills *et al.*, *Rev. Sci. Instrum.* ***90****, 083001 (2019)*
(m) Saule *et al.*, *Nat. Commun.* ***10,*** *458 (2019)*
(n) Zhang *et al.*, *arXiv: 2003.02429v1 (2020)*
(o) Corder *et al.*, *Struct. Dyn.* ***5****, 054301 (2018)*

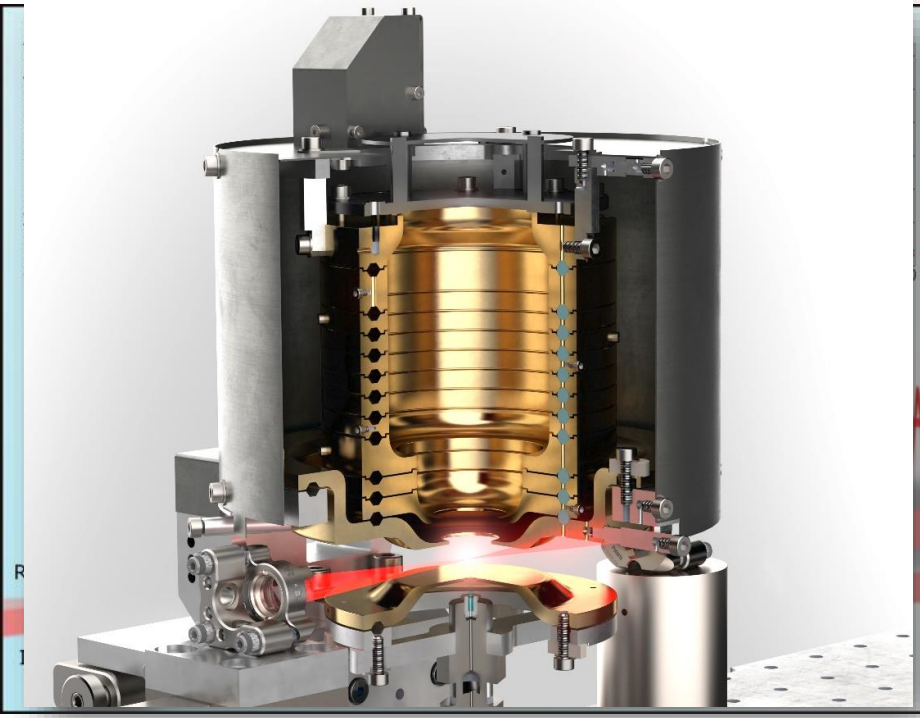
Gas recycling

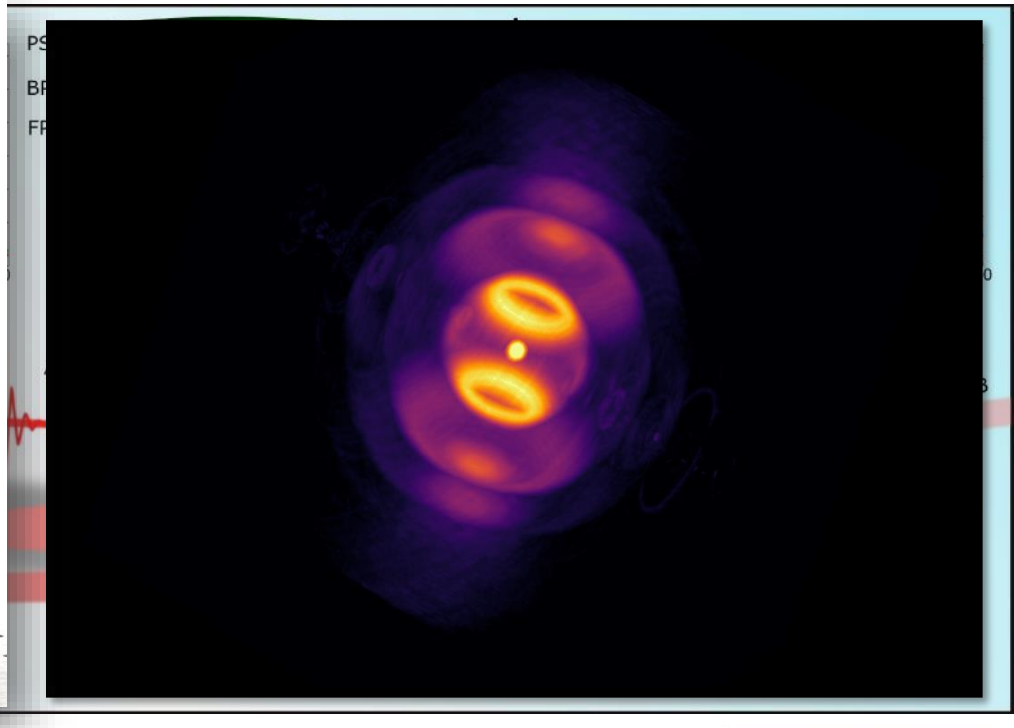


- High Xe pressure needed for large XUV yield
  - ➢ Xe consumption: 5 L/hour
  - ➢ 10-day measurement: €17.000
- Designed gas recycling system with compression of Xe
- Preliminary results
  - Xe consumption: 0.1 L/hour
  - 10-day measurement: €430

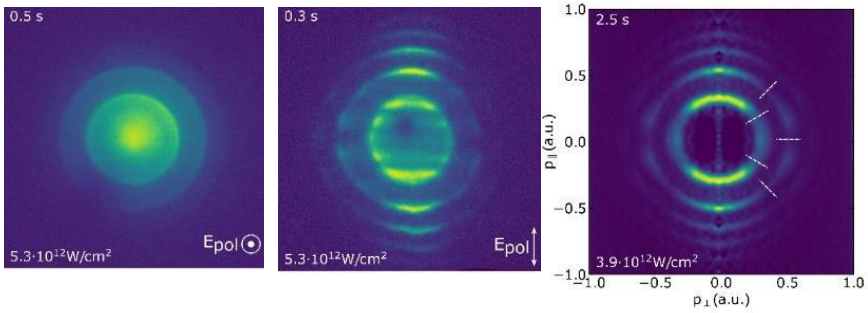


Cavity-enhanced comb application: Velocity Map Imaging (VMI)





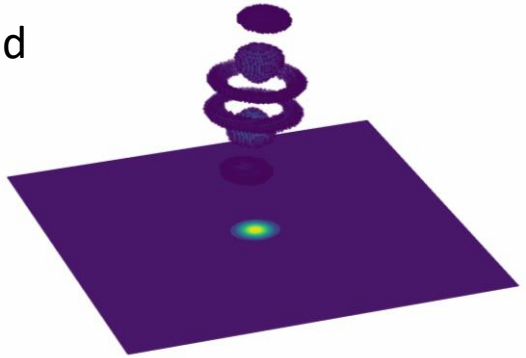
- Imaging of Xenon electrons ionized by absorption of many IR photons in the cavity focus region
- First VMI at a repetition rate of 100 MHz, much higher count-rate compared to kHz laser systems
- Allows for studies at lower intensities and at higher precision
- Dedicated enhancement cavity for VMI has been designed and built
- Rotating polarization allows 3D electron tomography

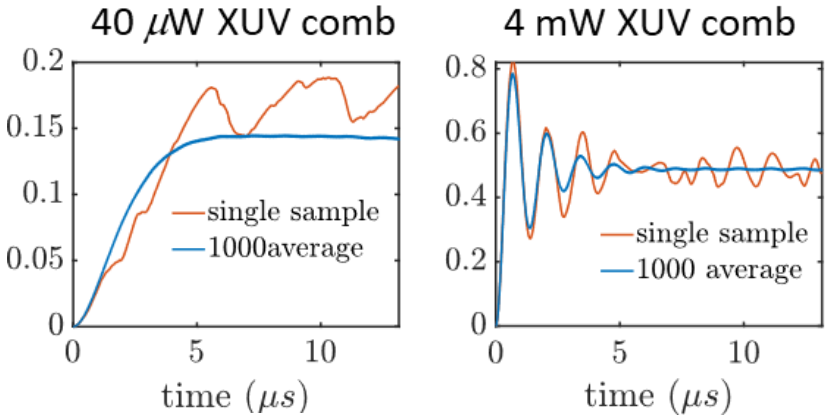


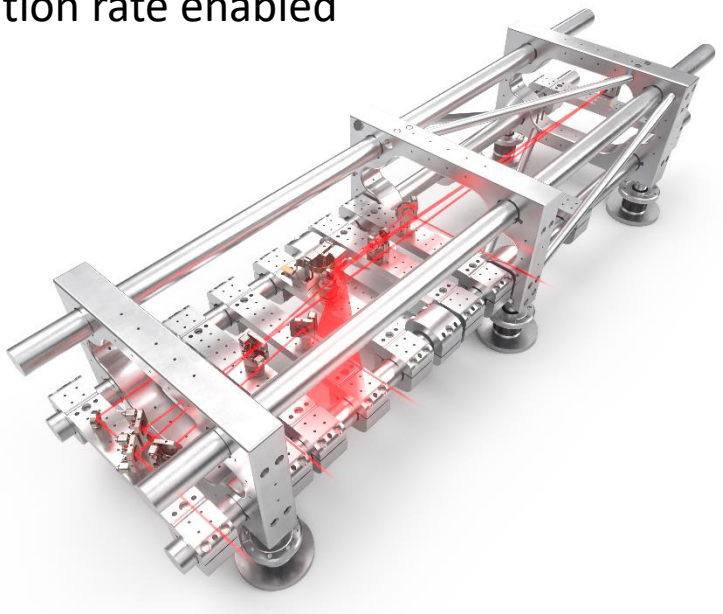
JN *et al.*, Opt. Lett. ***8,*** 2156 (2020)

Summary & Outlook

- A new, unique XUV frequency comb system has been developed
- High harmonics up to the 35th order were observed at an energy of 42 eV (30 nm)
- Stable operation for many hours demonstrated
- Yield of 50 μW is sufficient for driving HCI transitions with kHz excitation rates
- Important milestone towards the very first HCI metrology in the XUV region
- As an interesting new avenue, the high repetition rate enabled multi-photon studies at a very low intensity







Lyu *et al.*, Phys. Rev. Lett. **125**, **093201** (2020)

Thanks for your attention!

José Crespo Lopéz-Urrutia & Thomas Pfeifer

XUV comb

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