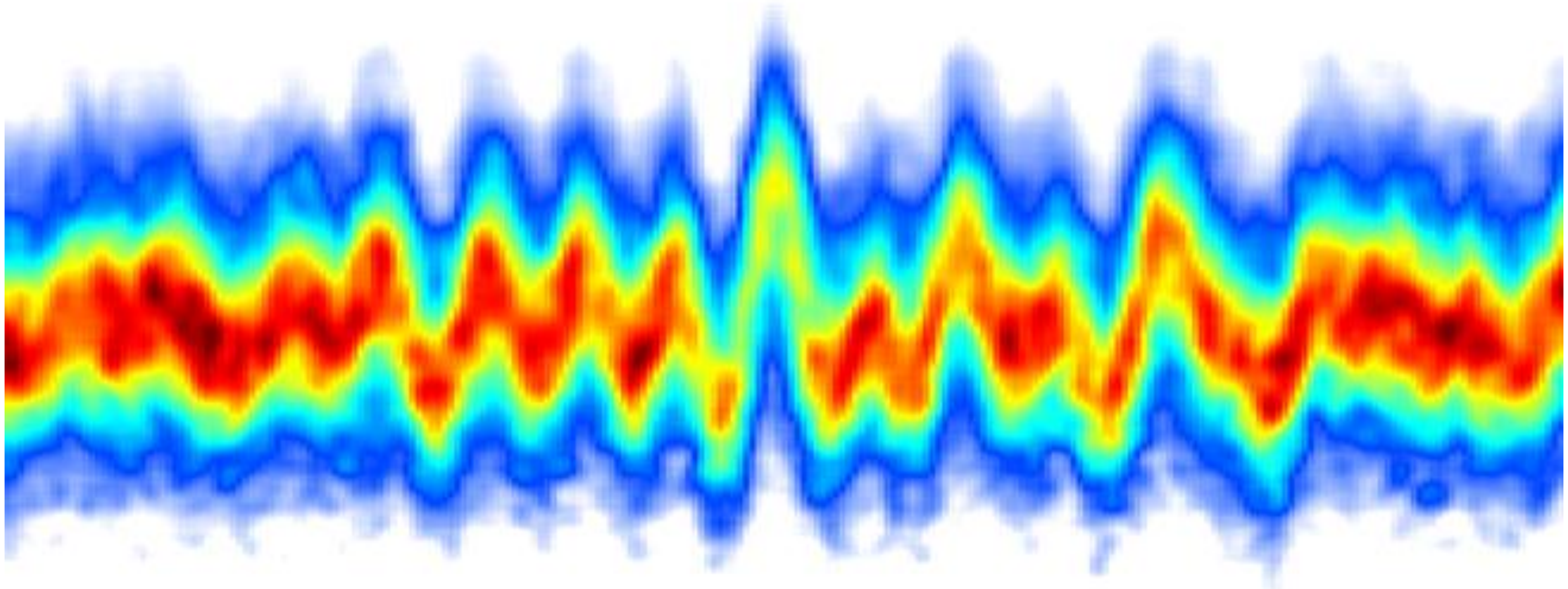
Laser Pulses shorter than one Light-Field Oscillation

DESY Science Day 2020

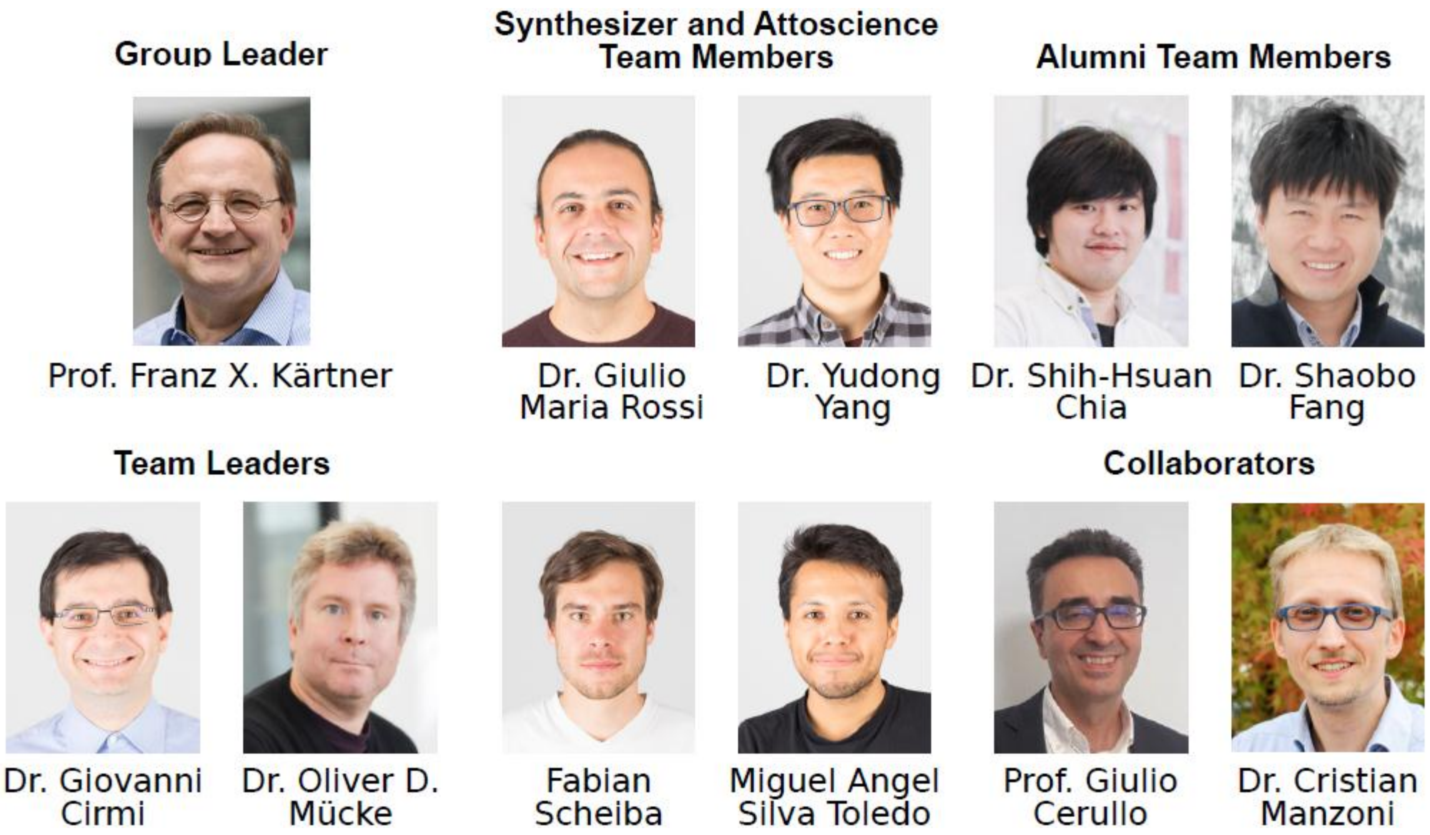


Roland E. Mainz
Hamburg, 2nd December 2020

Group of Ultrafast Optics and X-Rays

At CFEL, Prof. Franz X. Kärtner

Our Team

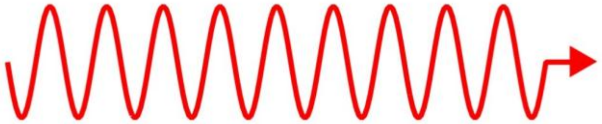


Center for Free-Electron Laser Science

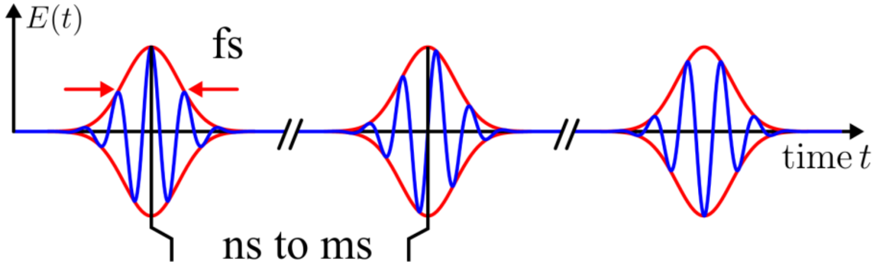
60 Years of Laser Development

Gaining Full Control of the Light Field

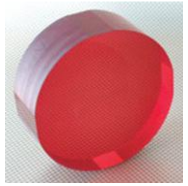
Continuous Wave Laser



Femtosecond Pulses via Modelocking

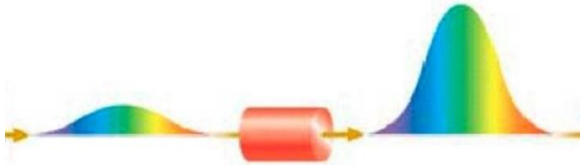


Broadband Laser Media



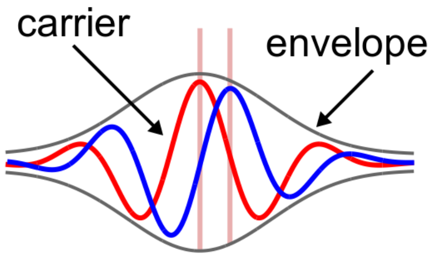
Ti:Sapphire

Chirped-Pulse-Amplification



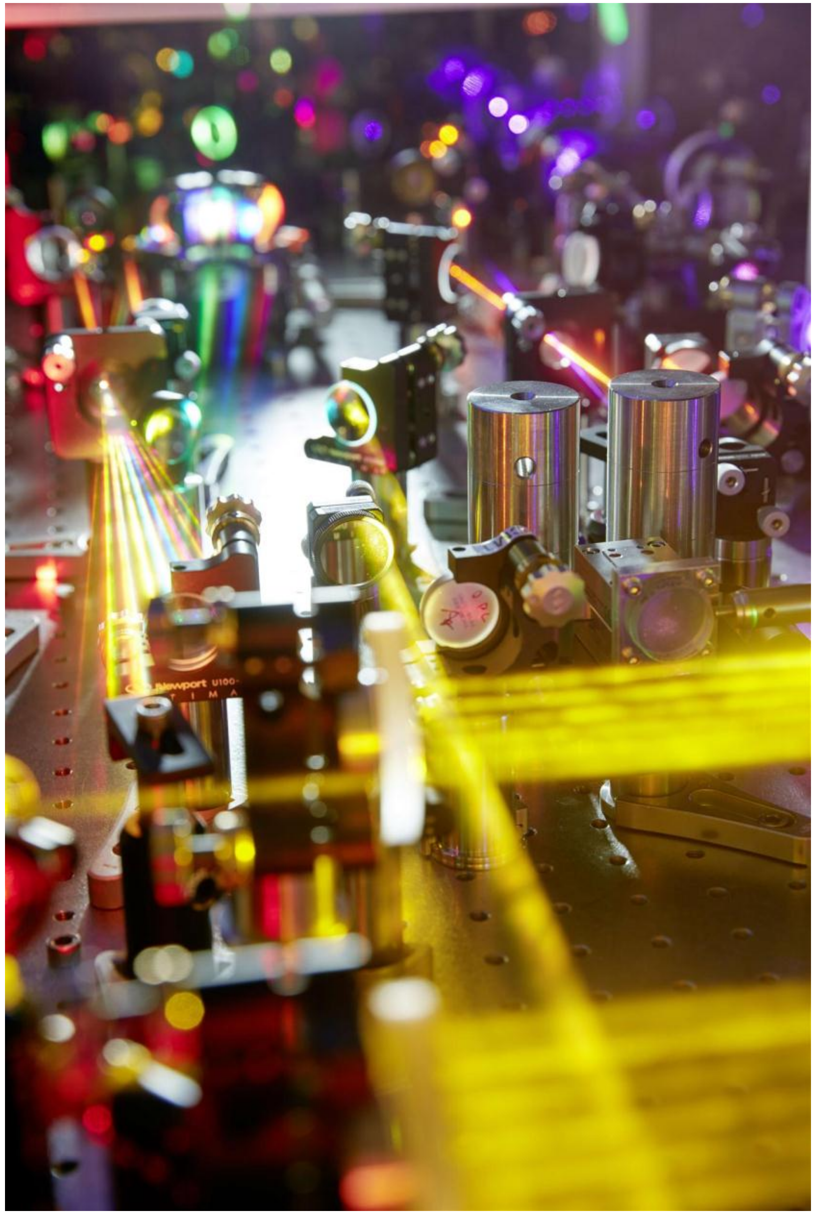
G. Mourou & D. Strickland, Nobel Prize 2018

Controlling the Carrier-Envelope-Phase (CEP)



J. L. Hall & T. W. Hänsch, Nobel Prize 2005

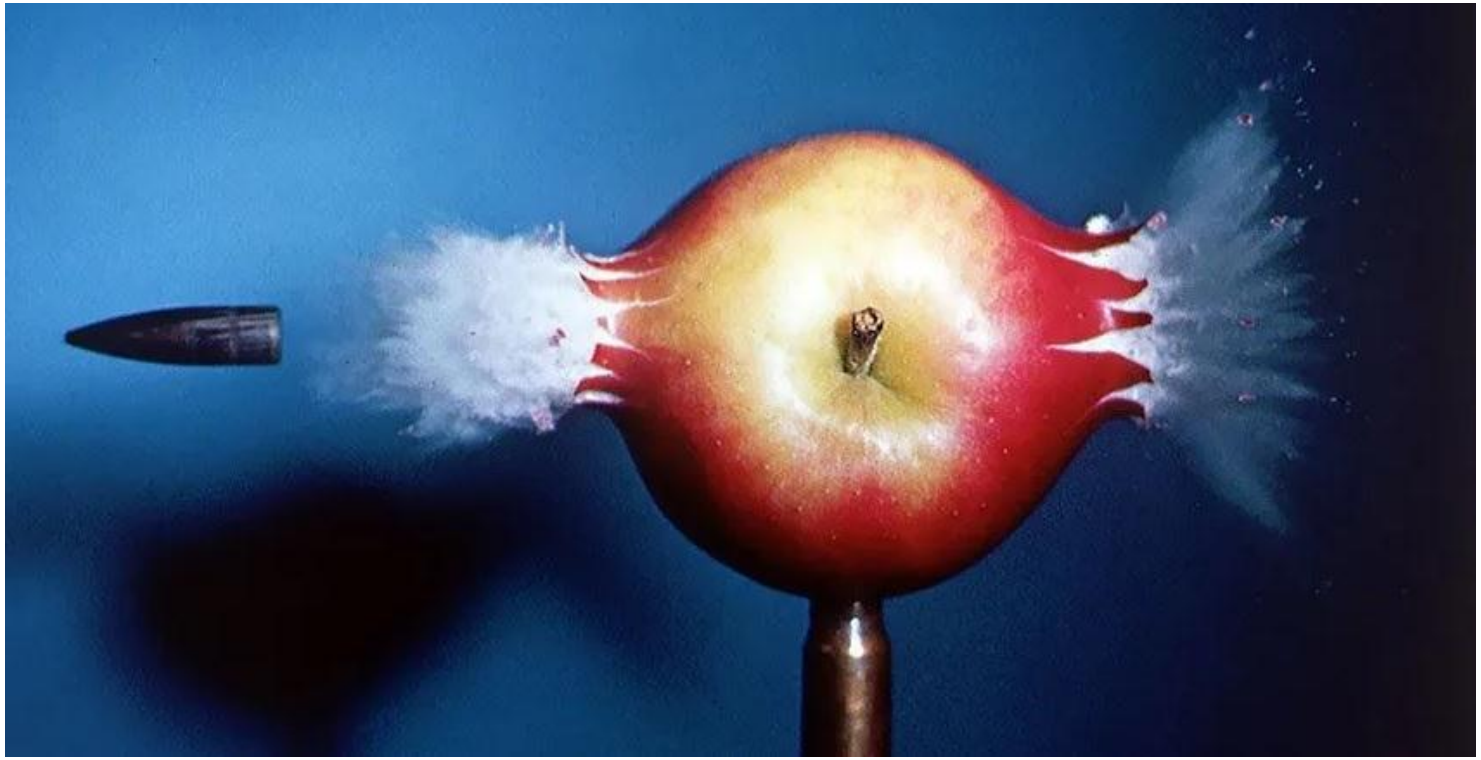
View of Our Experiment



Light at Extreme Scales

Listening to ultrafast processes

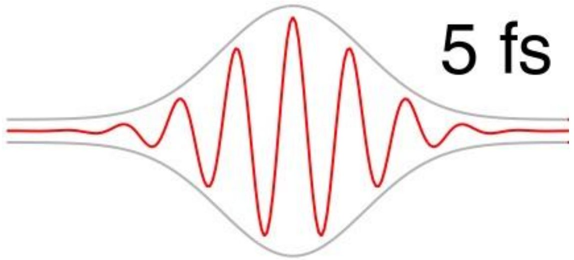
Freezing Time with Electronic Flash Photography



by H. Edgerton

- The shortest flashes of light for the highest time resolution

Few-Cycle Laser Pulse



- Allows to study the timescale of molecular dynamics

Light at extreme scales:
1 fs = 0.000 000 000 000 001 s
1 PW = 1 000 000 000 000 000 W

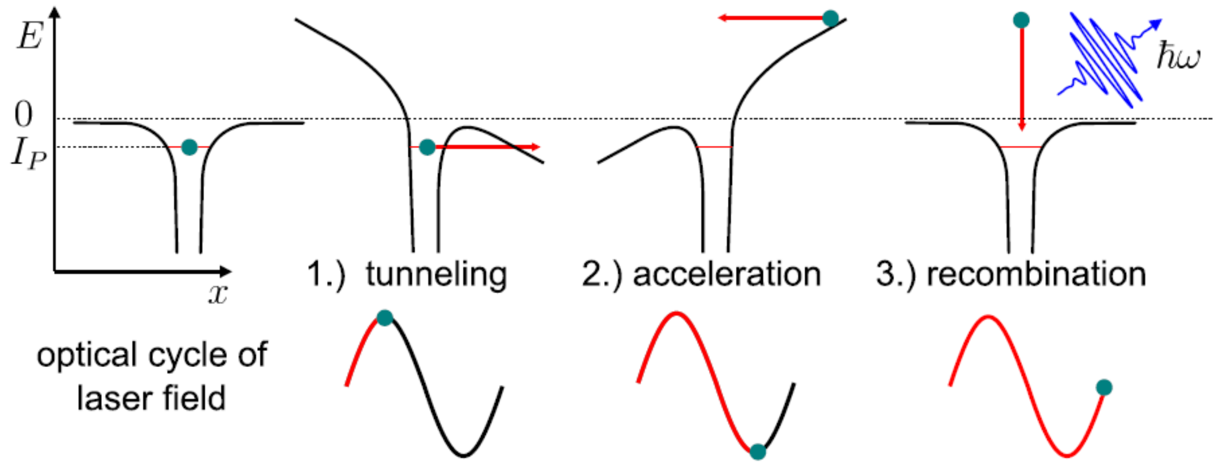
Entering the Realm of Attoseconds

High Harmonic Generation (HHG) at the forefront of ultrafast science

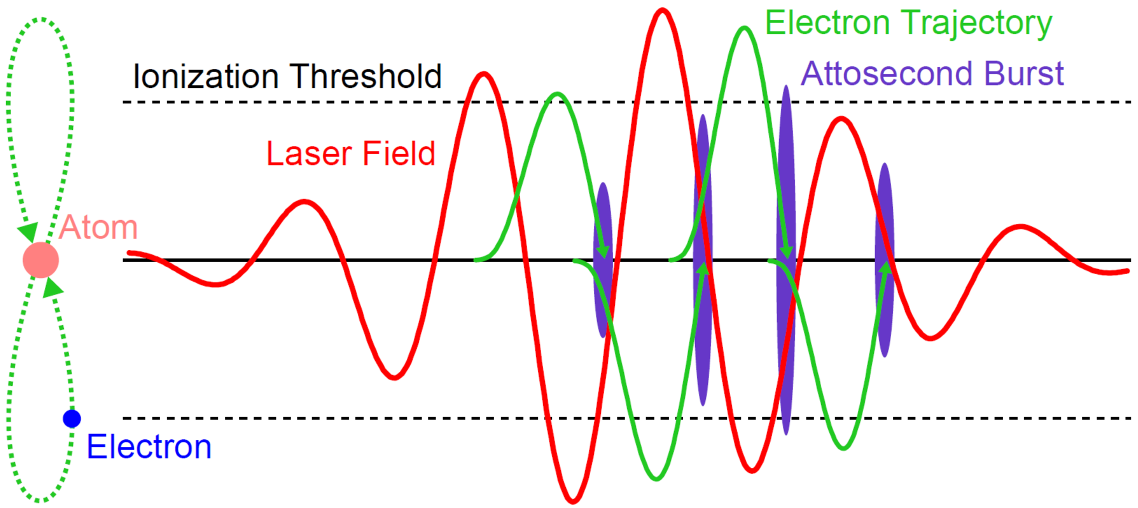
HHG provides:

- Pulses in the XUV to soft X-ray
- Pulse shortening (~ 50x)
- BUT: low yield: 10^{-8} to 10^{-5}
- up to 1.6 keV photon energy

3-Steps of HHG:



HHG driven by Few-Cycles:



Entering the Realm of Attoseconds

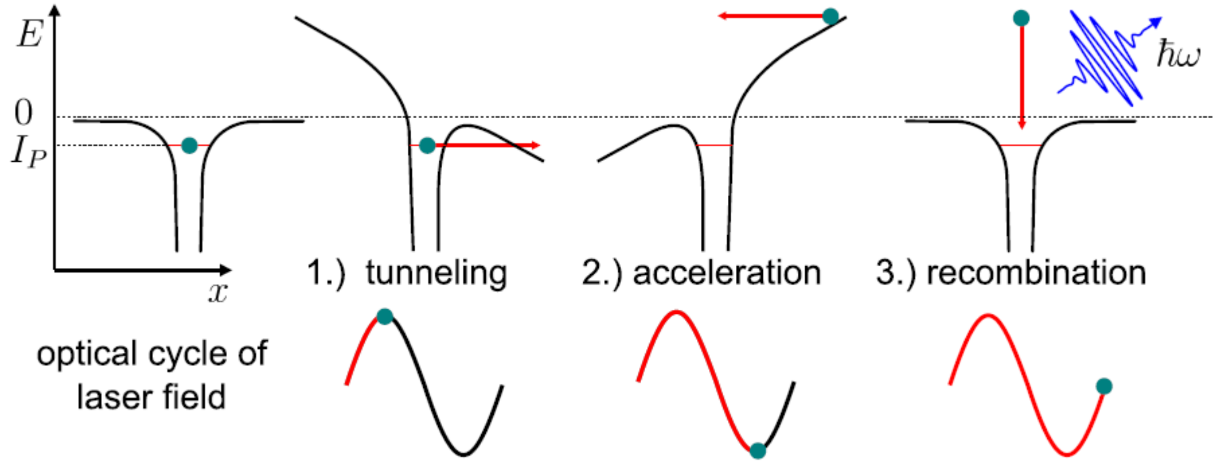
High Harmonic Generation (HHG) at the forefront of ultrafast science

HHG provides:

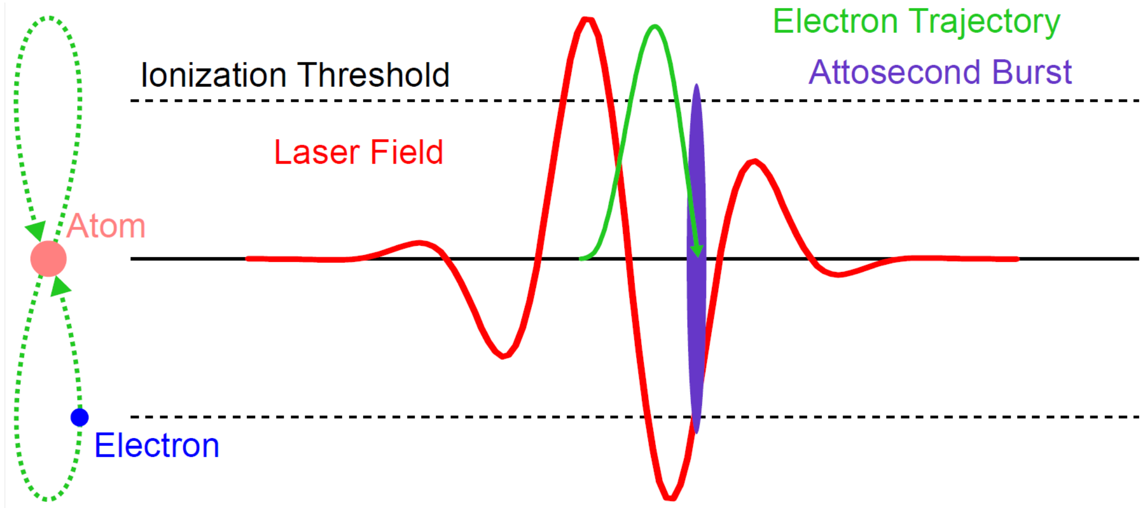
- Pulses in the XUV to soft X-ray
- Pulse shortening (~ 50x)
- BUT: low yield: 10^{-8} to 10^{-5}
- up to 1.6 keV photon energy

Sub-cycle pulses eliminate the need for gating techniques and promise to increase the brightness of the isolated attosecond pulse

3-Steps of HHG:



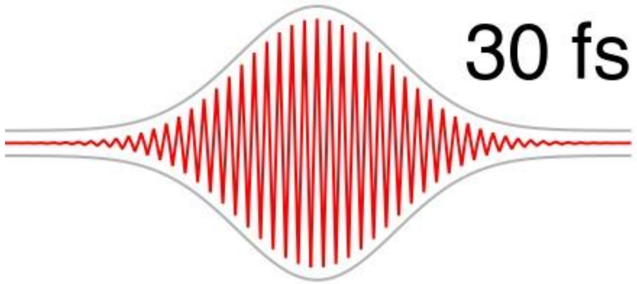
HHG driven by Few-Cycles:



Pulse Shortening Techniques

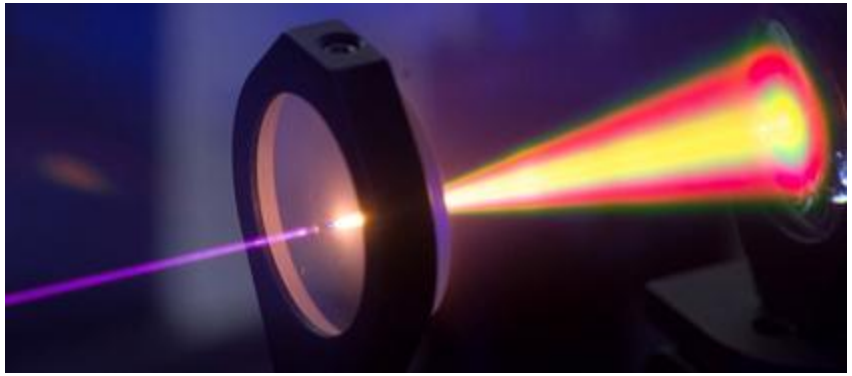
Spectral broadening for a short pulse in time

Input Pulse



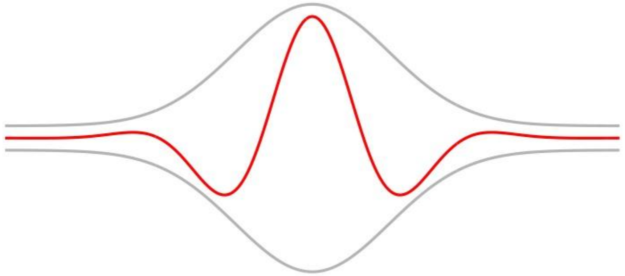
White-light generation in bulk

~nJ, >2 octaves



https://en.wikipedia.org/wiki/Supercontinuum

Sub-Cycle Output Pulse



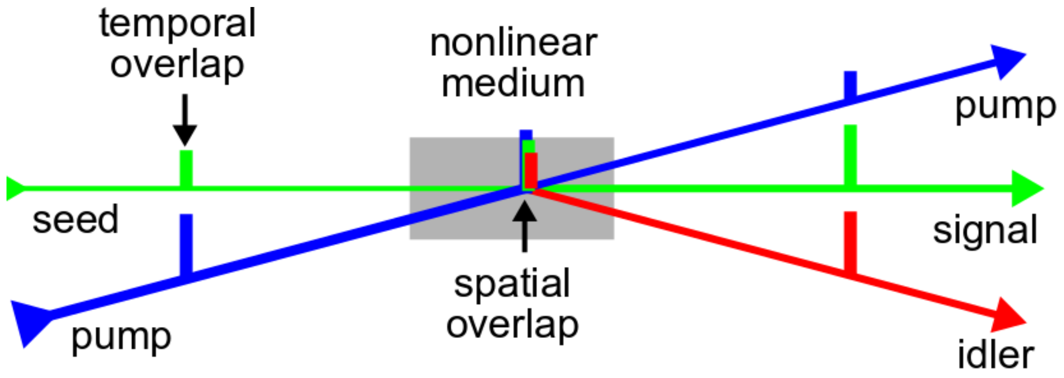
Broadband Amplification with OPAs

Optical Parametric Amplifiers

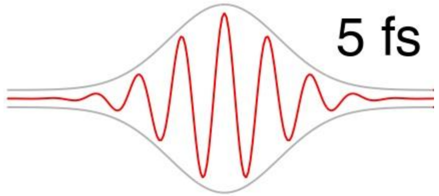
- High gain 10x – 10.000x
- Suitable for UV to mid-IR
- Allows broadband amplification

  (up to 1 octave)

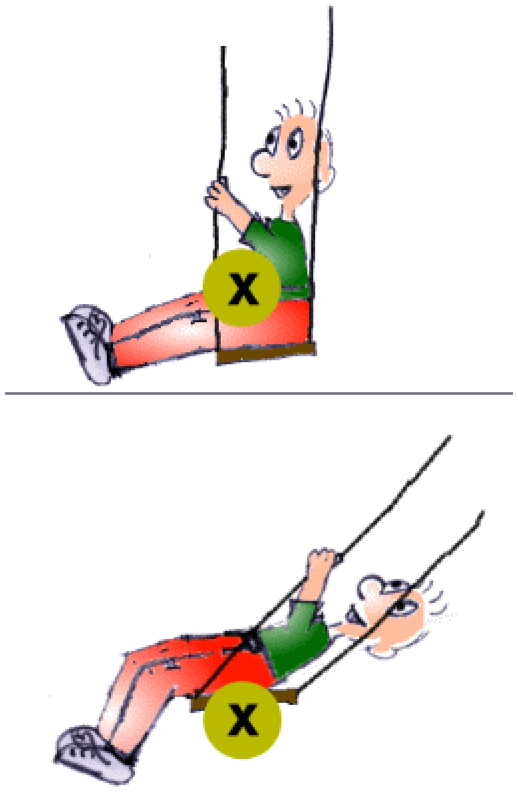
OPA setup



Few-Cycle Pulse



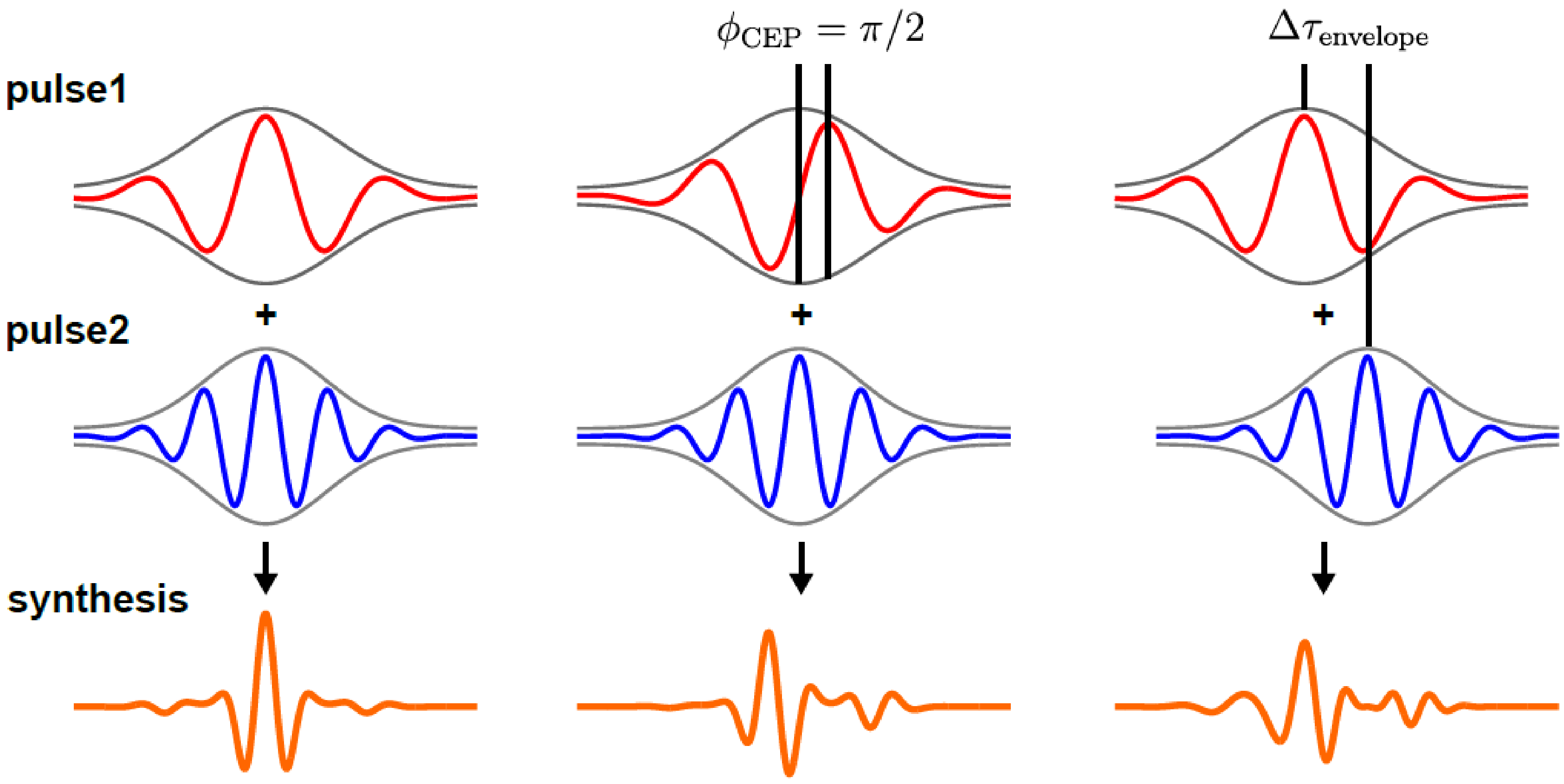
Intuitive Analogy



Shifting of center-of-mass twice per swing period

The Synthesis of Few-Cycle Laser Pulses

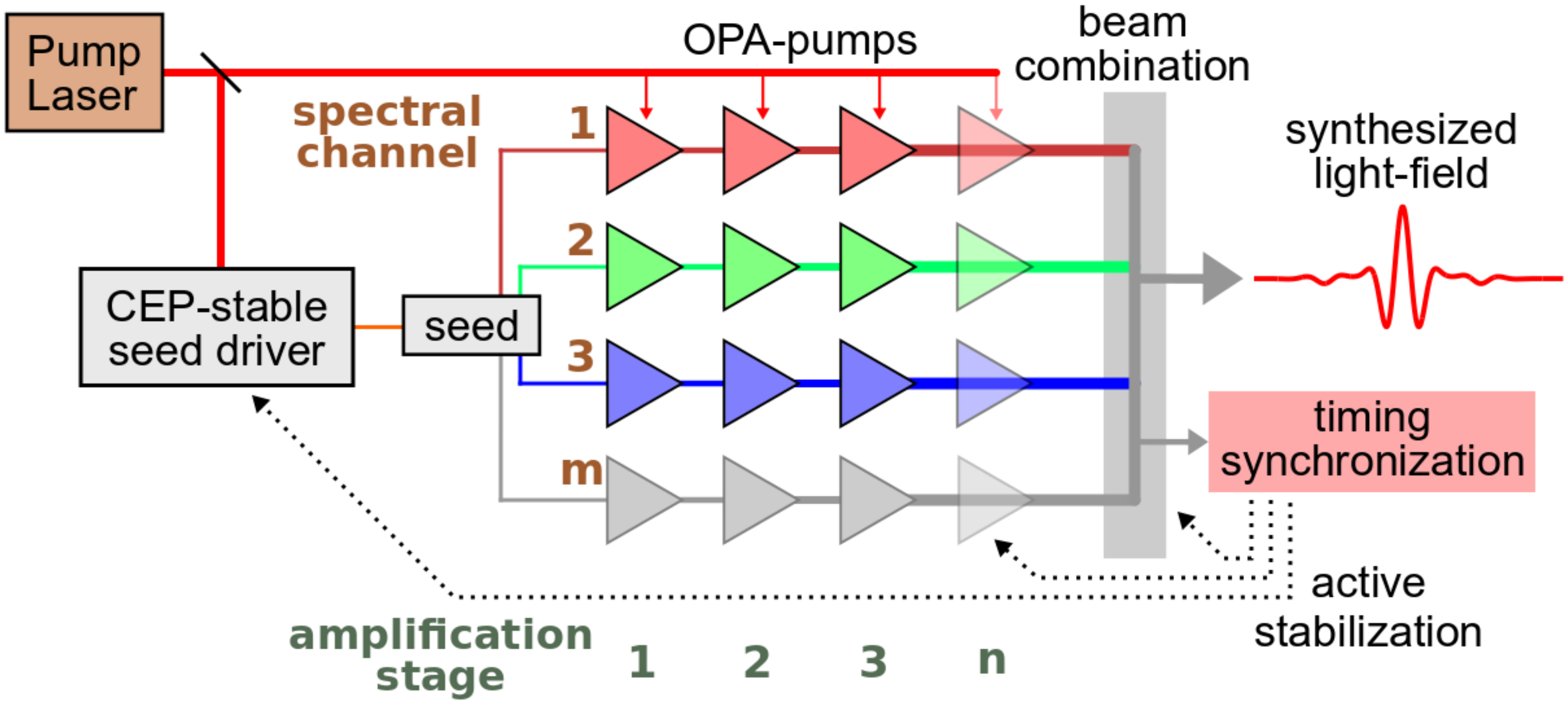
Crafting laser pulses shorter than one optical cycle



Scheme for OPA-based Synthesis

Coherent combination of ultrashort laser pulses

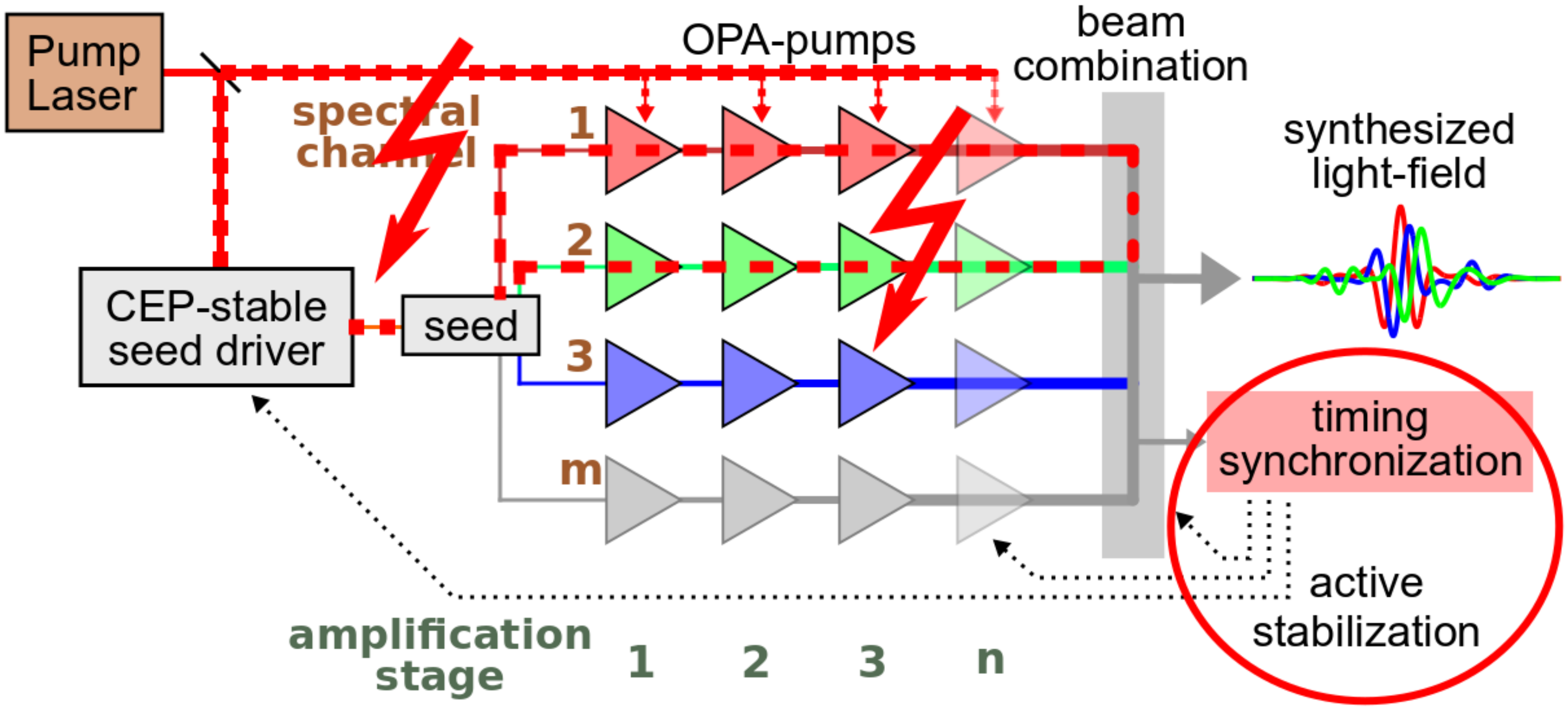
- Scheme is Scalable in:
  - Bandwidth
  - Pulse Energy
  - Output Power
- CEP-stable broadband seed generation is crucial



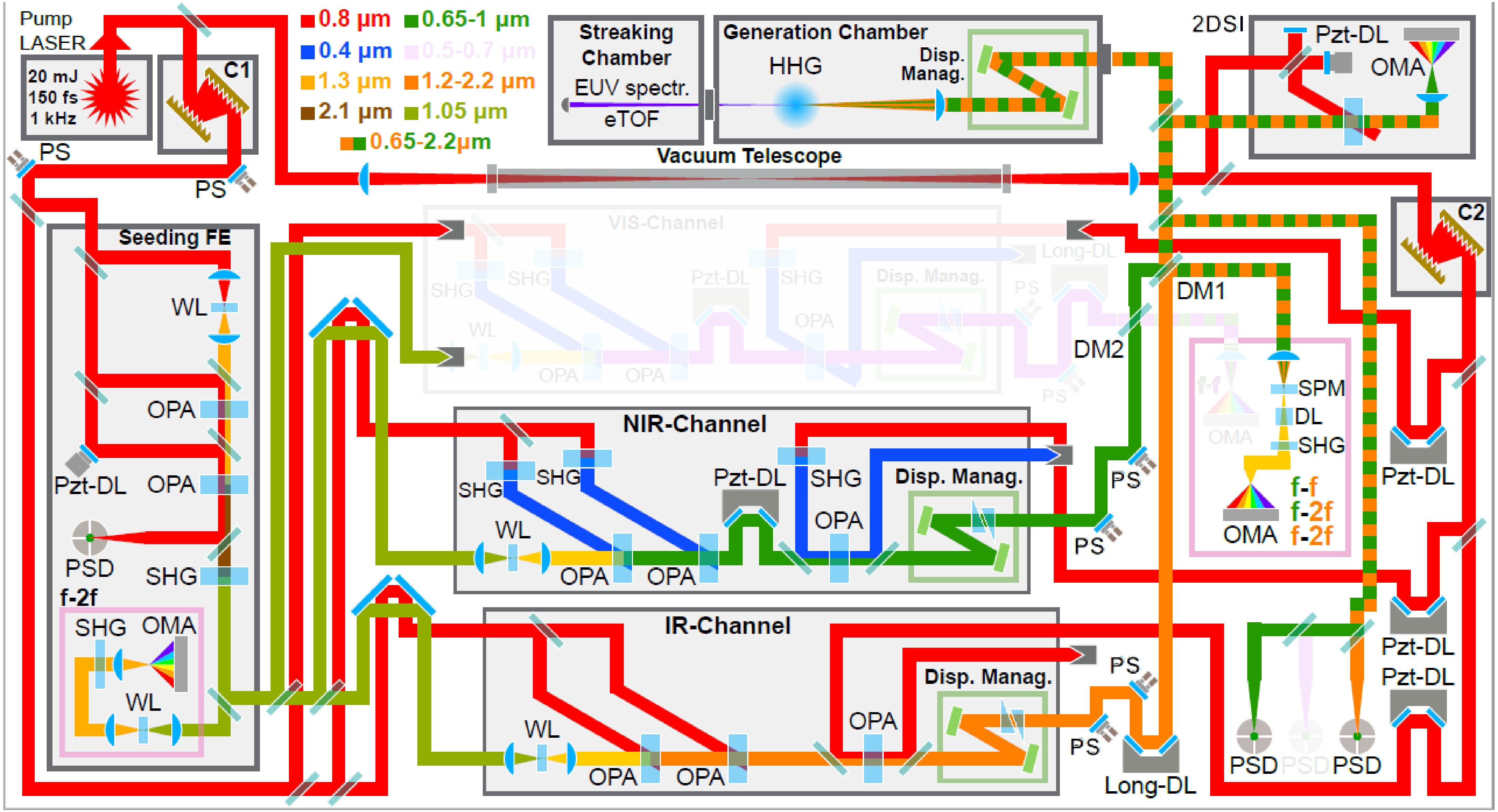
Scheme for OPA-based Synthesis

Coherent combination of ultra-short laser pulses

- Scheme is Scalable in:
  - Bandwidth
  - Pulse Energy
  - Output Power
- CEP-stable broadband seed generation is crucial
- **Requires active attosecond synchronization**



Our Current Implementation

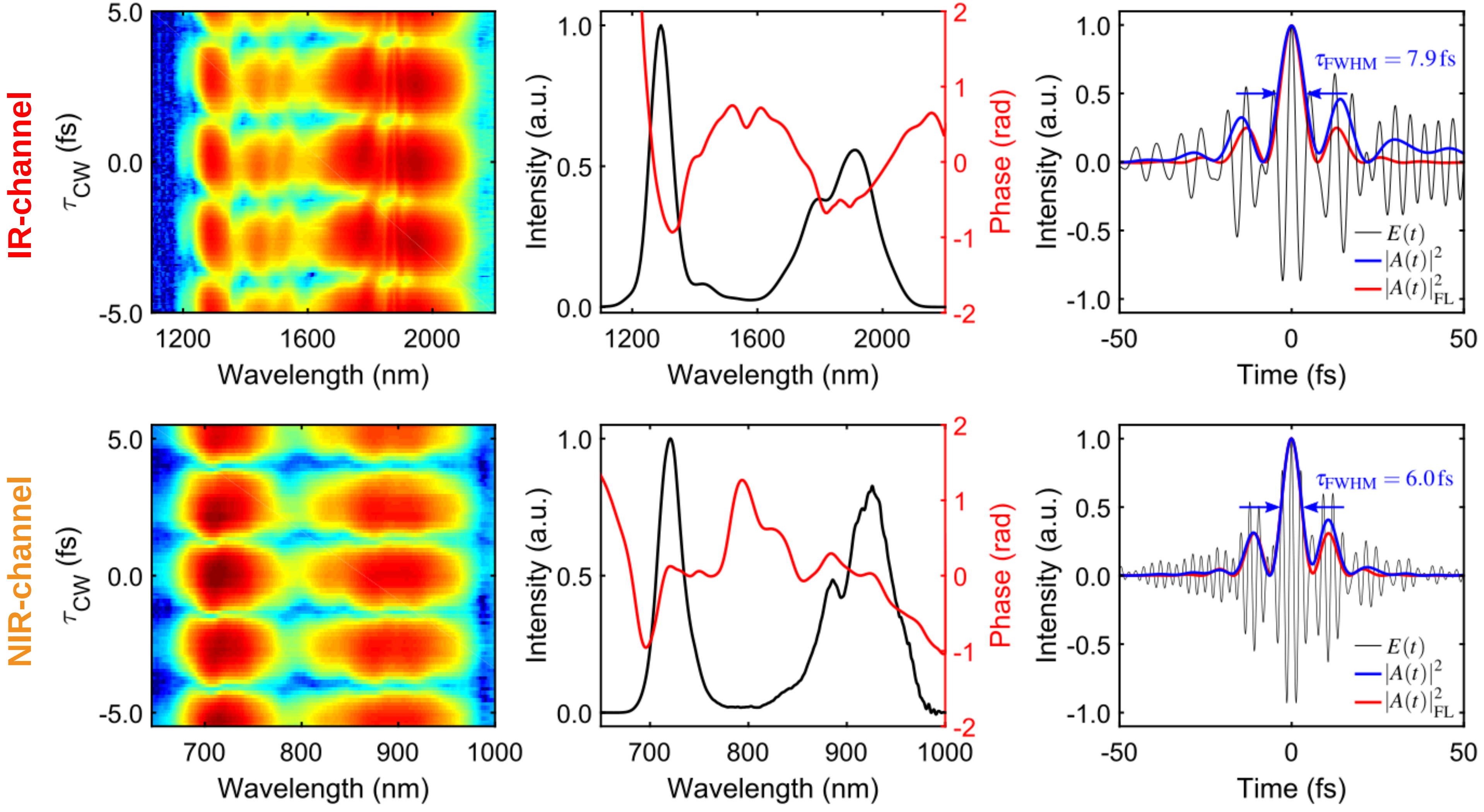


Rossi et al., Nature Photonics 2020

Few-Cycle Pulses from each Spectral Channel

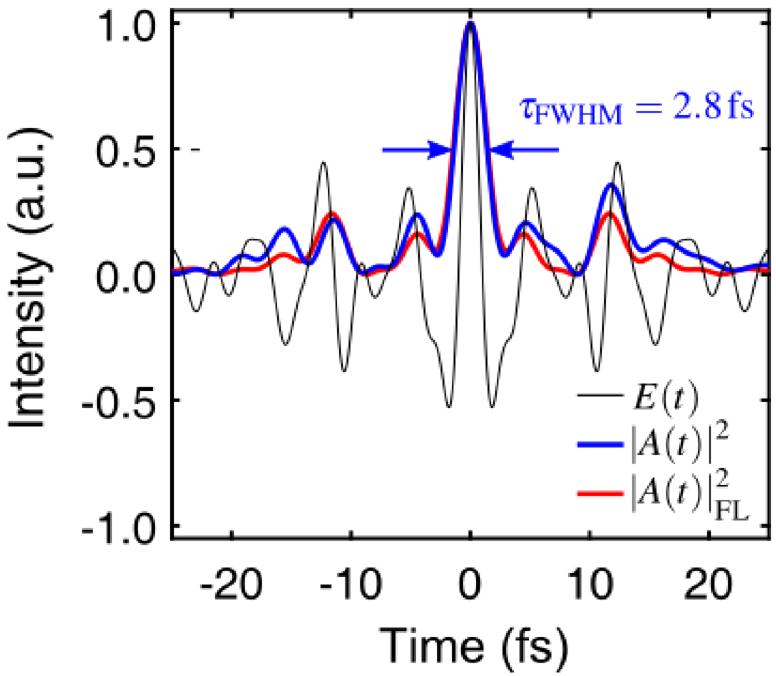
Pulses characterized via 2-dimensional spectral shearing interferometry

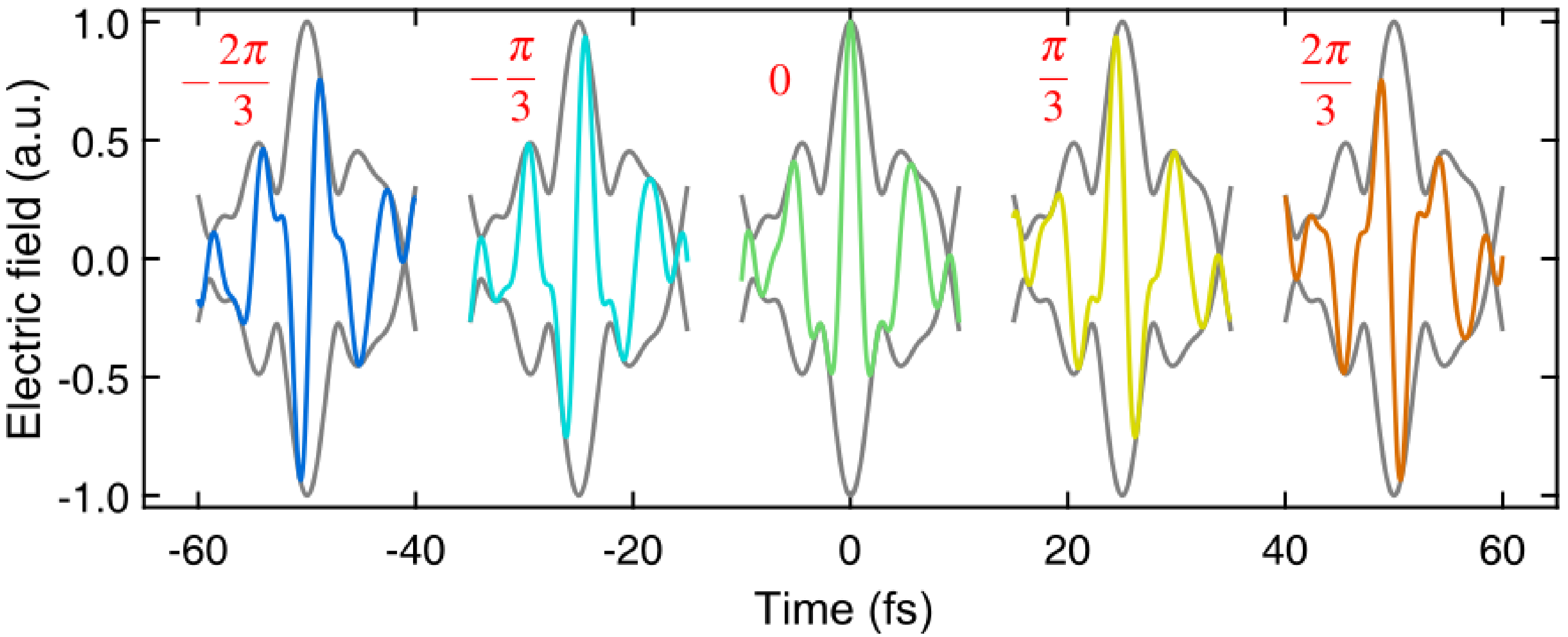
- **IR-channel:**
  - 1200-2200 nm
  - 7.9 fs
  - 500 µJ
- **NIR-channel:**
  - 650-1000 nm
  - 6.0 fs
  - 100 µJ
- **(VIS-channel:)**
  - 500-700 nm
  - ~6 fs
  - 150 µJ



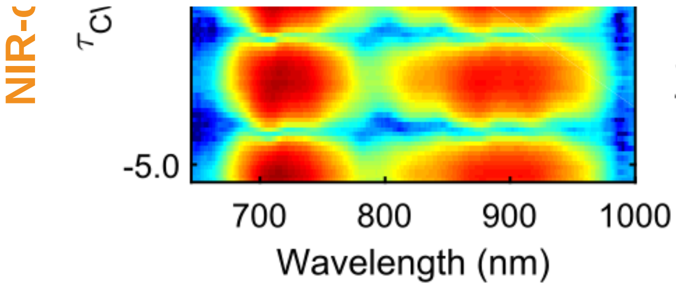
Few-Cycle Pulses from each Spectral Channel

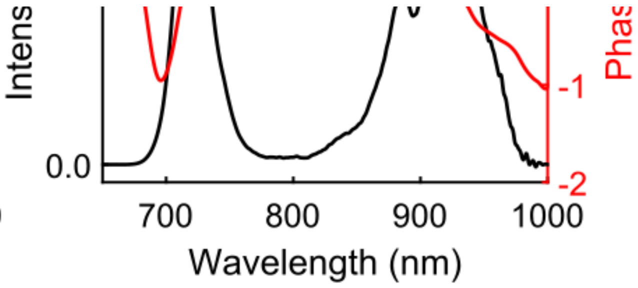
Pulses characterized via 2-dimensional spectral shearing interferometry

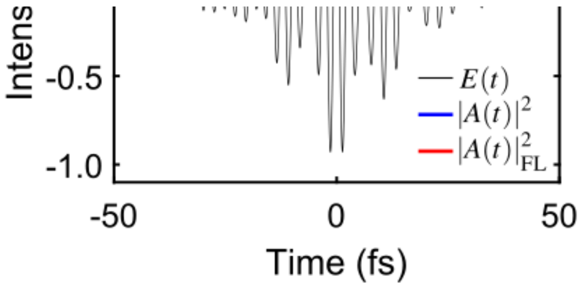




- 500-700 nm
- ~6 fs
- 150 µJ







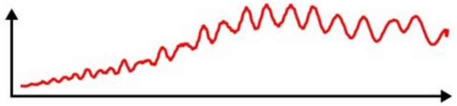
Stabilizing and Controlling the Synthesized Waveform

.... with attosecond precision

Synchronization System:

- In-Line Dual Phase Meter with single-shot spectrometer (right)
- FPGA-based feedback system
- Several timing actuators:
  - short- and long-range stages
  - affect CEPs/RP/Delays

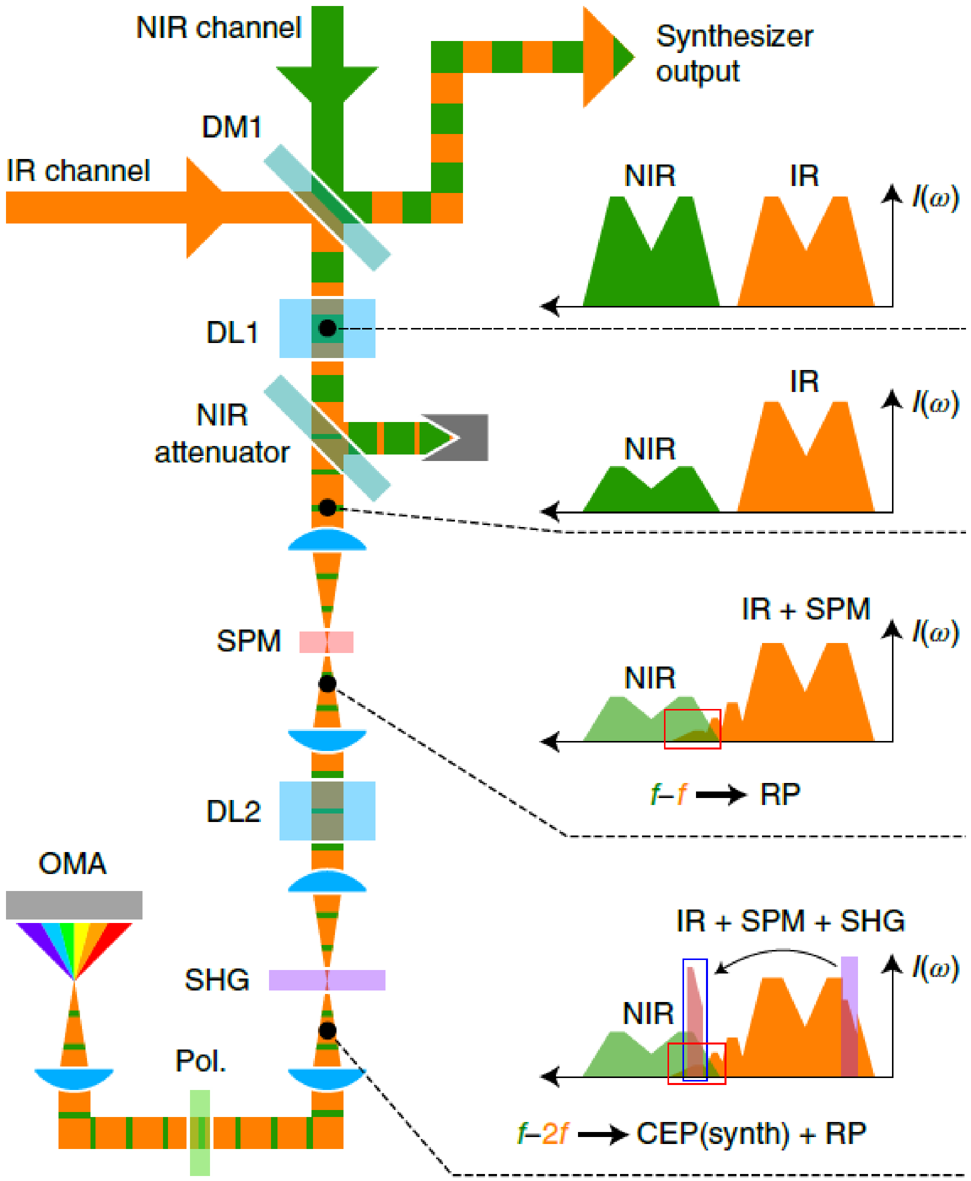
observed spectral interference signal



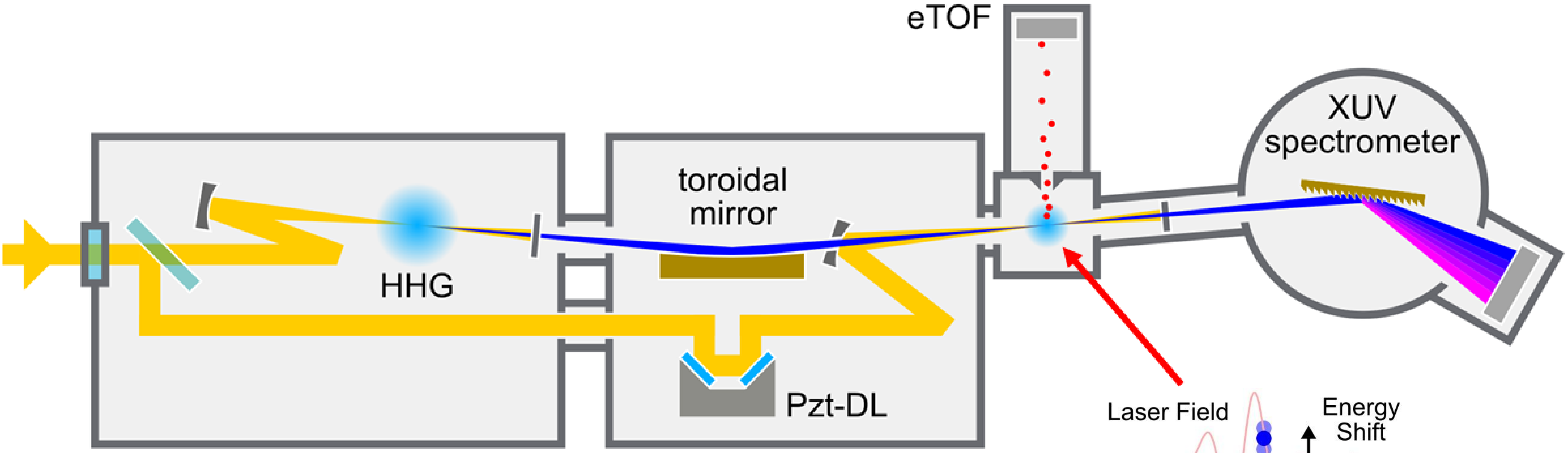
CEP-noise: 250 mrad rms

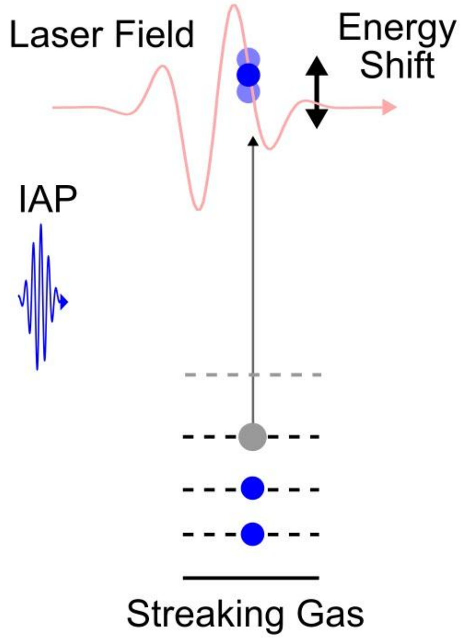
RP-noise: 80 mrad rms

- (1-4 % of waveform period)



Pulse Measurement via Attosecond Streaking

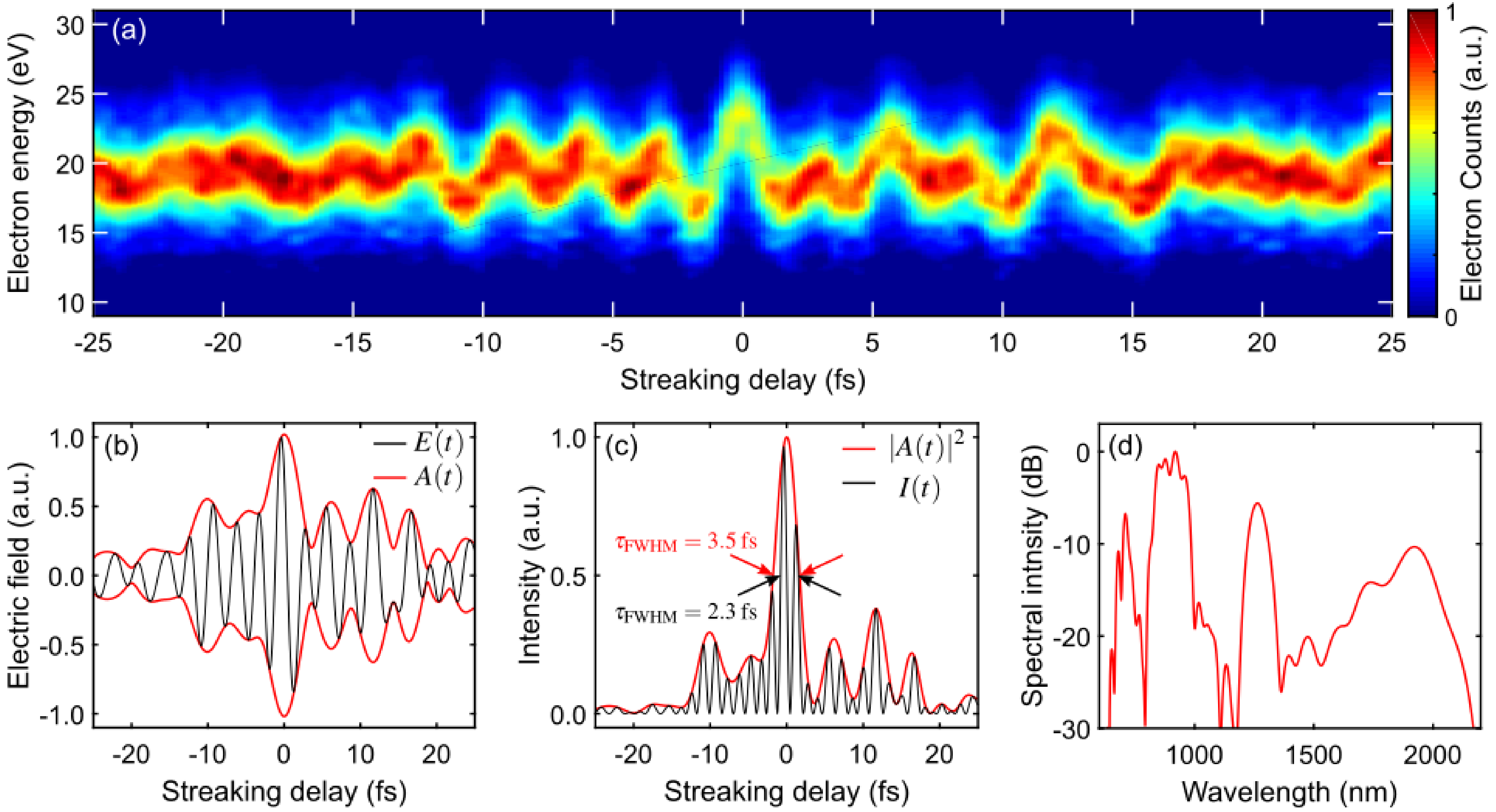




Attosecond Streaking Setup:

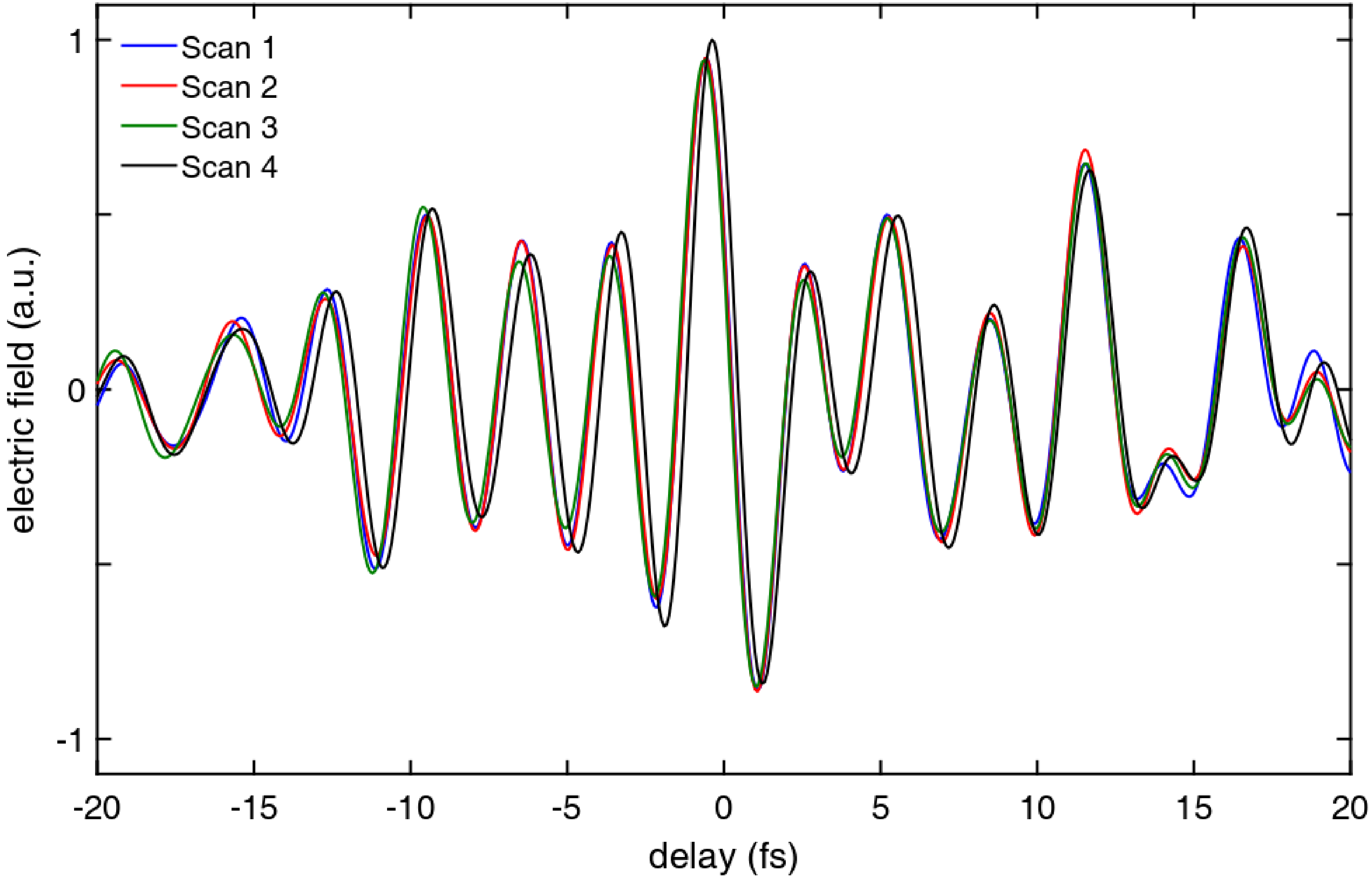
- Attosecond pulse releases photo-electrons in streaking gas
- Photo-electron energy modulated by sub-cycle pulse
- Full time-domain reconstruction of synthesized pulses

Attosecond Streaking of a Sub-Cycle Pulse



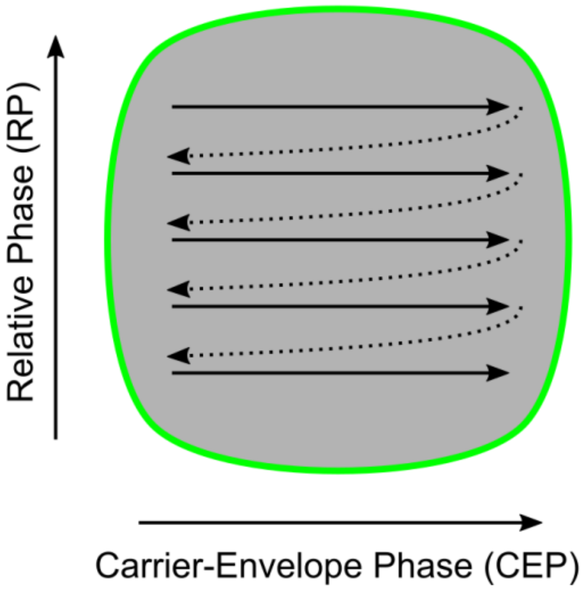
Rossi et al., Nature Photonics 2020

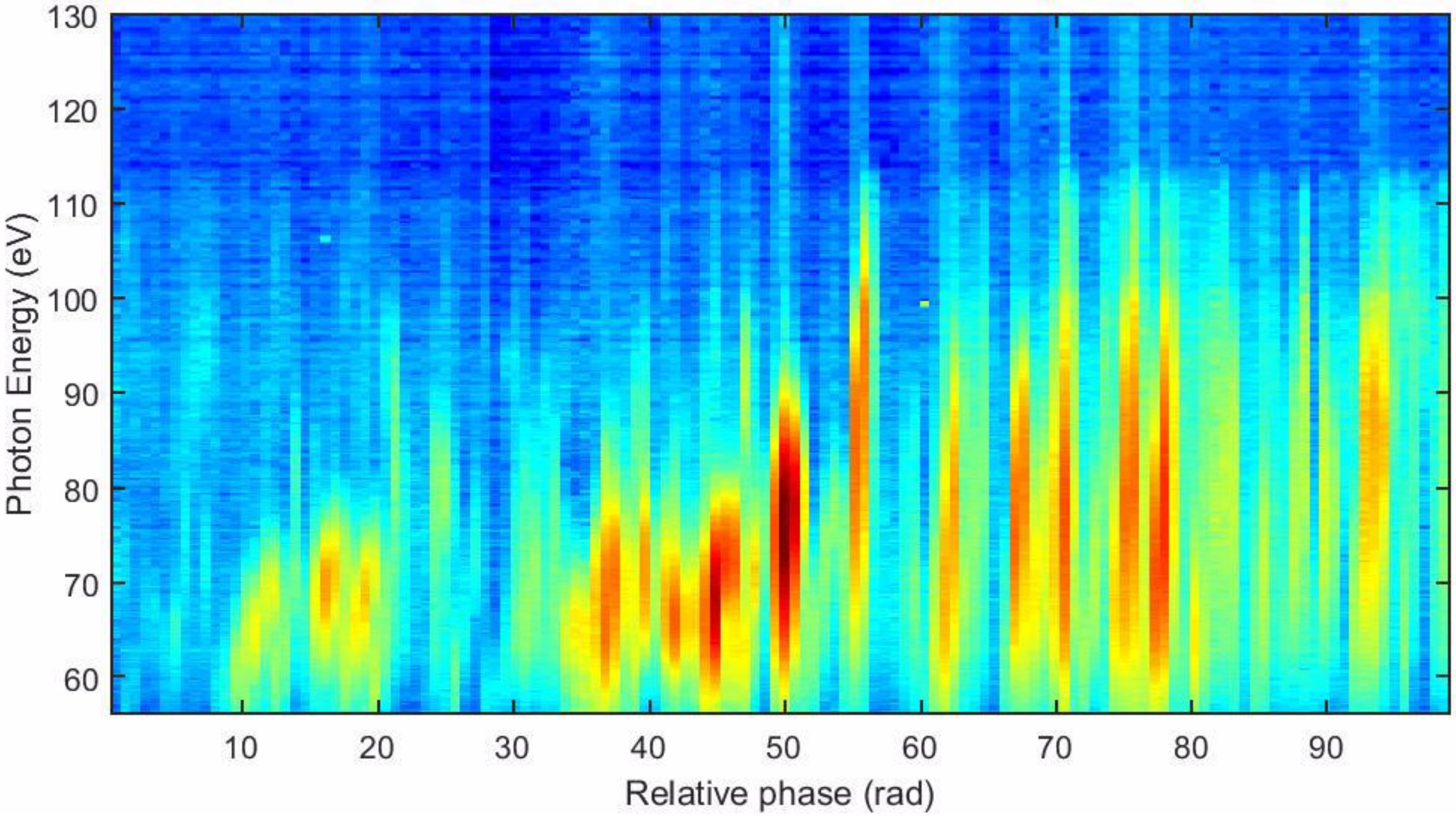
Repeatability of the Synthesized Waveform



HHG during Synthesis Parameters Scans

Online Scanning of CEP and RP while observing the HH-spectrum

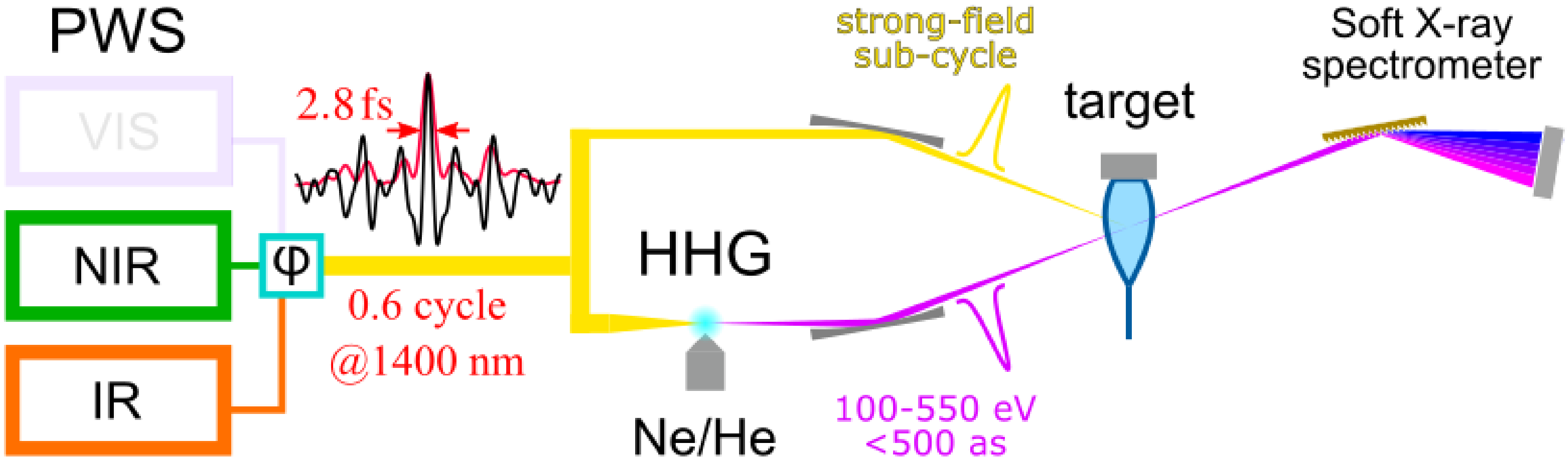




We have control on:

- Spectral Shape/Bandwidth
- Central Energy
- HHG Yield

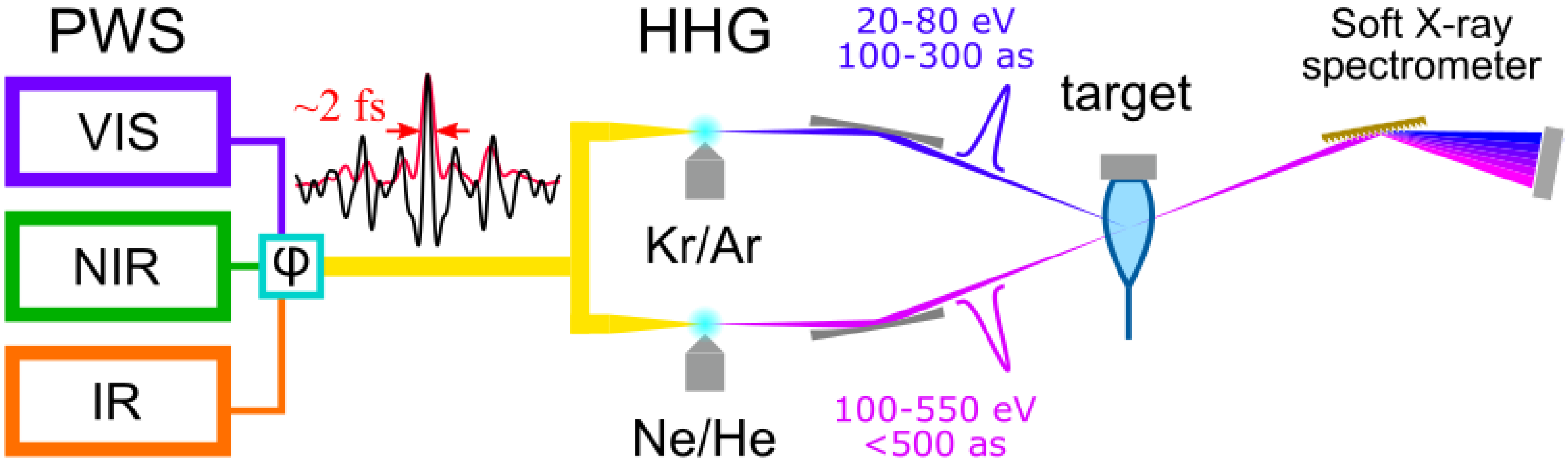
Towards Attosecond Pump-Probe Spectroscopy



Strong-Field-Attosecond pump-probe spectroscopy:

- Exploiting the sub-cycle nature of our synthesizer to achieve a *hybrid attosecond resolution*
- Strong-field excitation could potentially induce extrinsic dynamics

Towards Attosecond Pump-Probe Spectroscopy

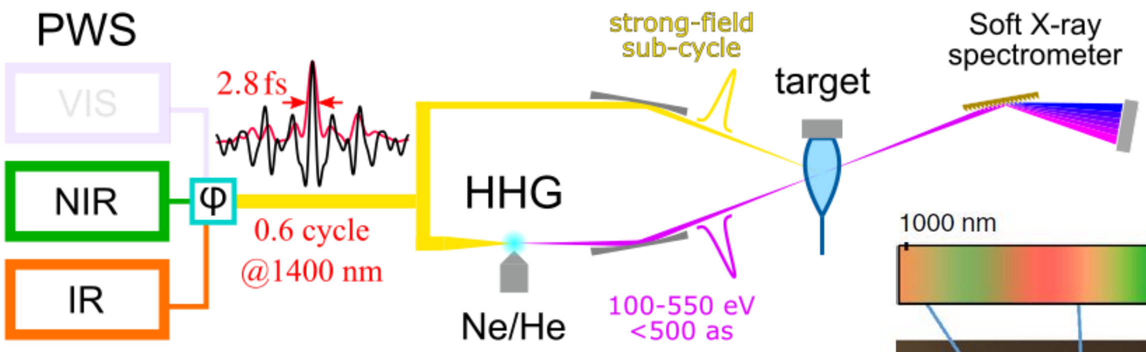


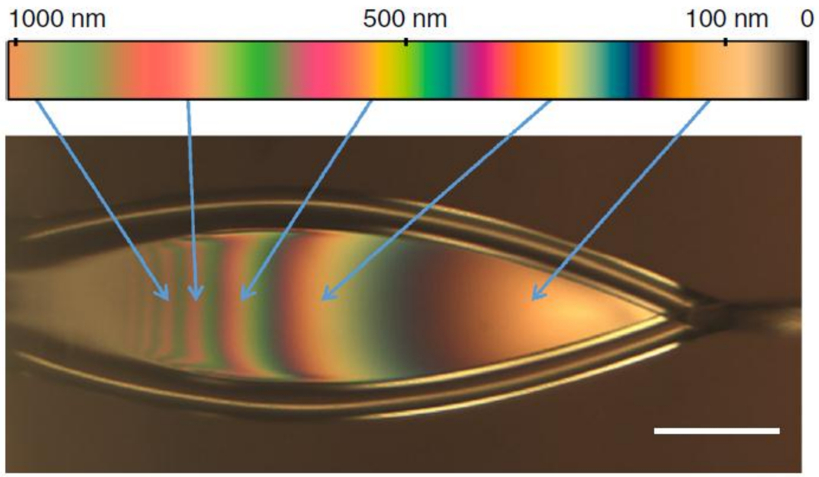
Attosecond-Attosecond pump-probe spectroscopy

- Different energy ranges via different HH-source gases
- **Very efficient HHG + tight focusing strictly required**
- Direct attosecond resolution

Towards Attosecond Pump-Probe Spectroscopy

- **Sample: Thin Liquid-Jet**
  - Defined confinement
  - Variable thickness
  - Sample: liquid or dissolved agent

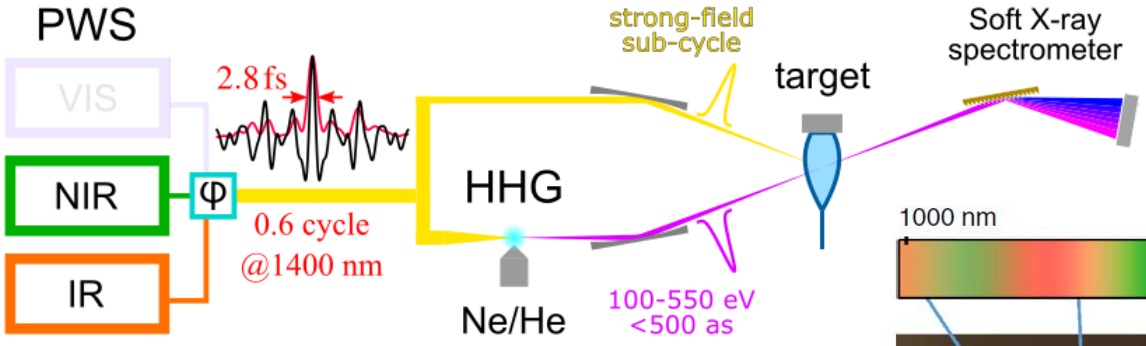


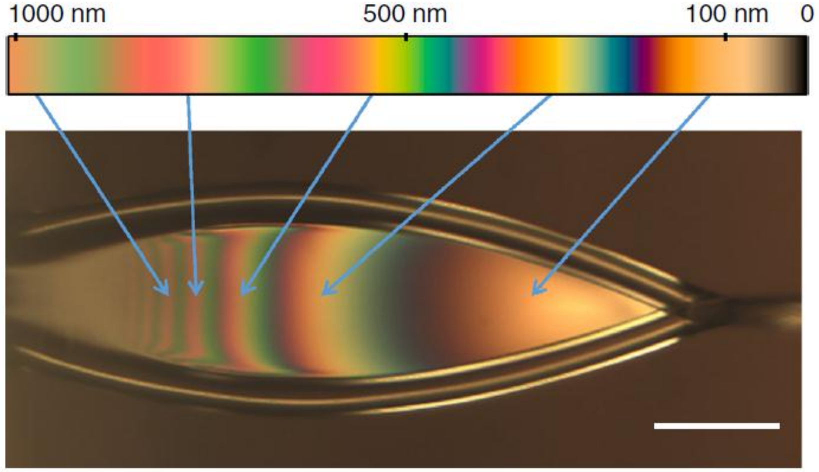


D. Koralek et al., Nat.Comm. Vol. 9, (2018)

Towards Attosecond Pump-Probe Spectroscopy

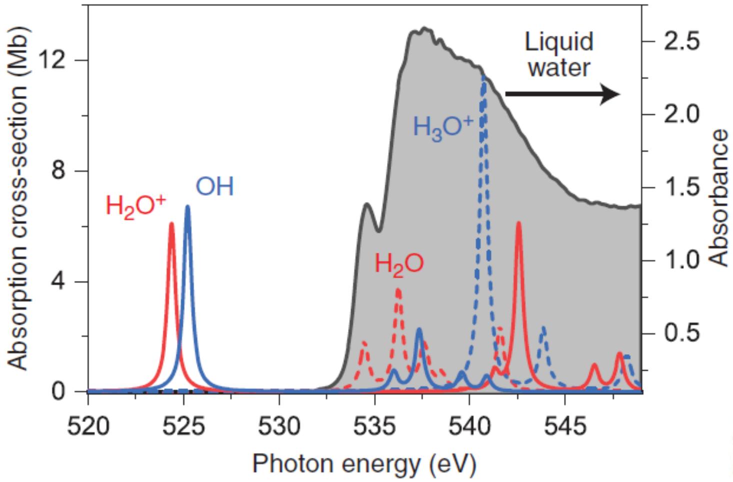
- **Sample: Thin Liquid-Jet**
  - Defined confinement
  - Variable thickness
  - Sample: liquid or dissolved agent





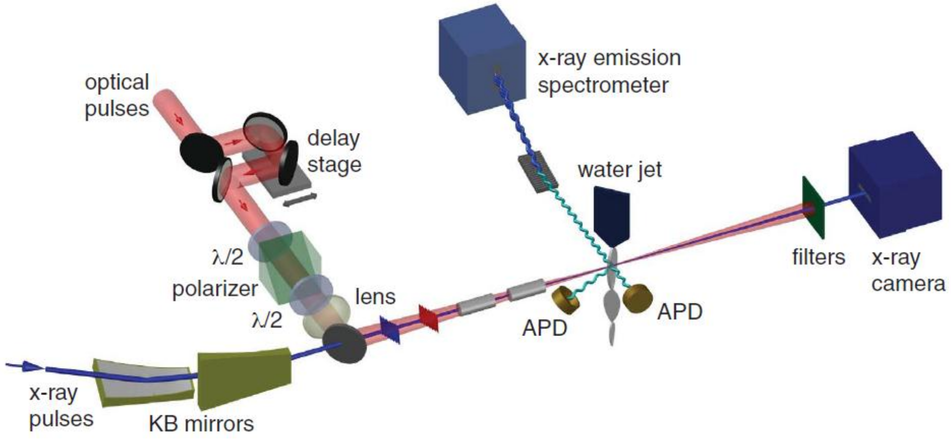
D. Koralek et al., Nat.Comm. Vol. 9, (2018)

$$H_2O^+ + H_2O \rightarrow OH + H_3O^+$$



Loh et al., Science 367, 179-182 (2020)

- **Studying Water Dissociation**
  - Manifold anomalies of water
  - Simple molecule of high relevance
  - Complex hydrogen-bond network dynamics

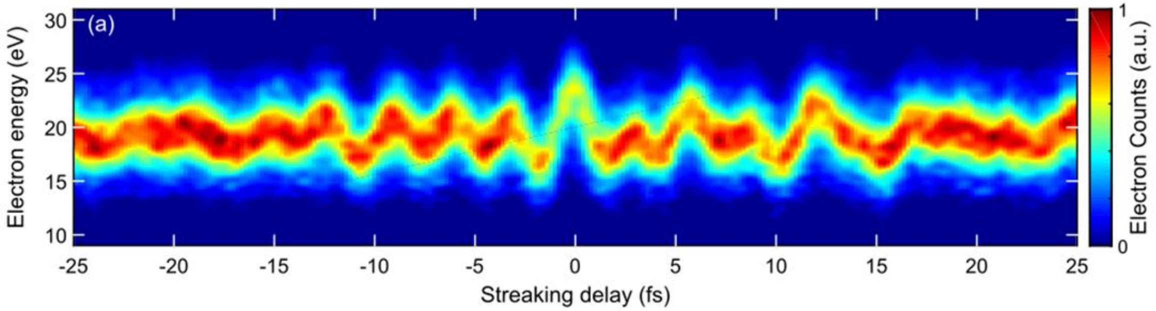


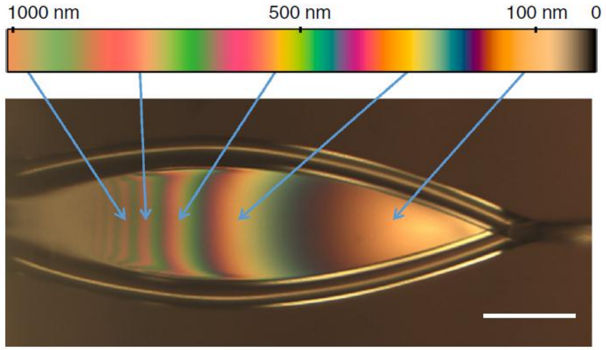
Conclusion and Outlook

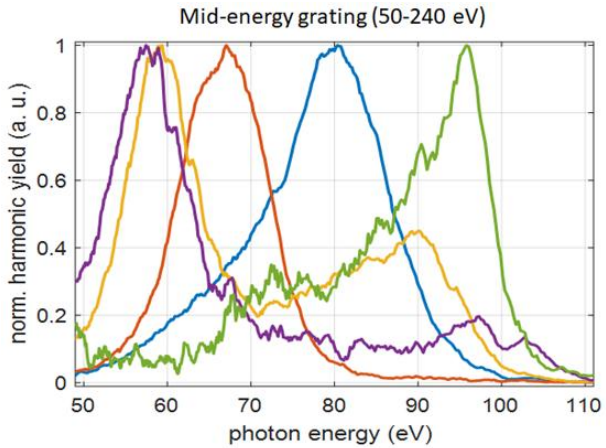
Demonstration of a novel laser technology and the implications for attosecond-resolved experiments

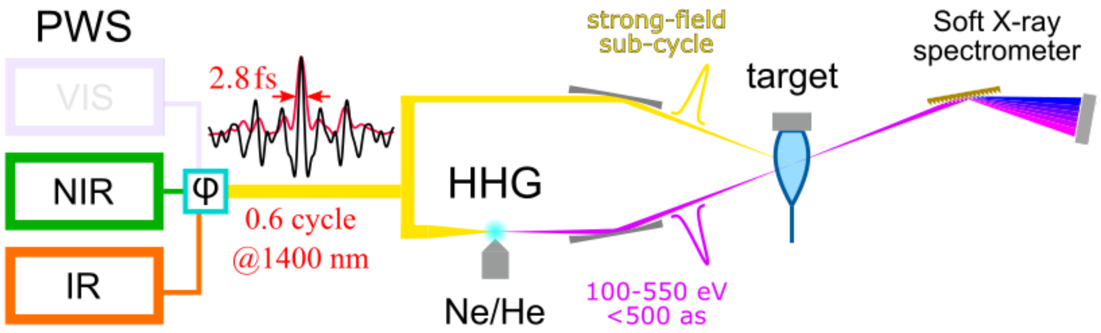
- Stable Sub-Cycle Pulse Generation
- Direct generation of isolated attosecond pulses via HHG
- Manifold Shaping of the attosecond pulses
- Attosecond Streaking for full reconstruction of the synthesized field

- *HHG reaching the water-window*
- *Sub-Cycle/Attosecond pump-probe experiments*









Thank you for your attention!