

Spectral interferometry with high harmonics, isolated attosecond pulses from laser plasmas on solid targets

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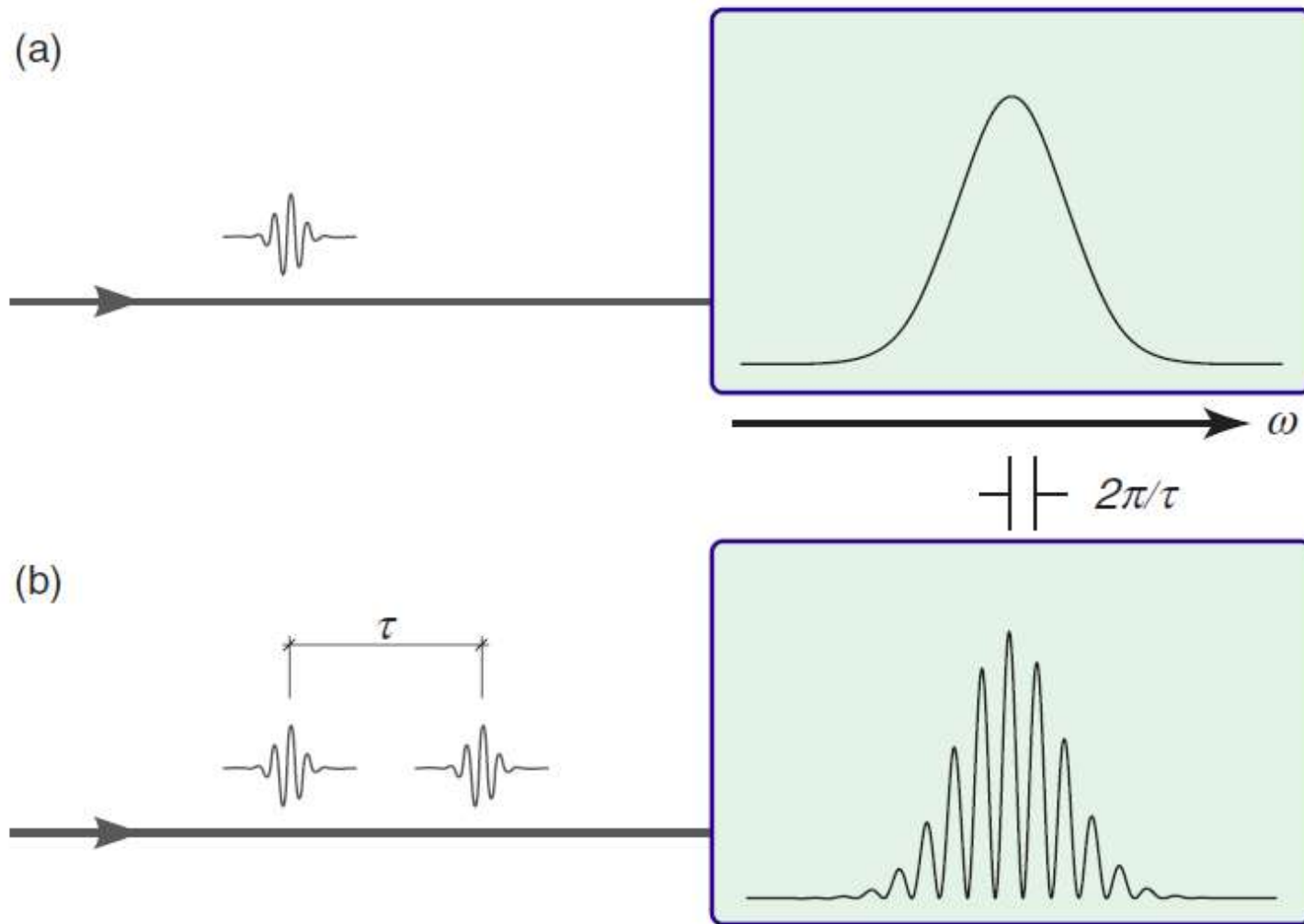
Collaboration with D. Kormin, A. Borot, G. Ma, W. Dallari, M. Aladi, B. Bergues, R. Bolla,
G. D. Tsakiris, F. Krausz

The experiments were carried out in the Max Planck
Institute of Quantum Optics, Garching, Germany

Content

- Spectral interferometry
- High Harmonic Generation (HHG) from laser plasmas on solid surfaces (plasma mirrors)
- The LWS-20 laser
- Measuring the carrier-envelope phase (CEP)
- Experimental arrangement
- Analysis of HHG spectra with spectral interferometry
- Plasma denting and isolated attosecond pulses
- Conclusion

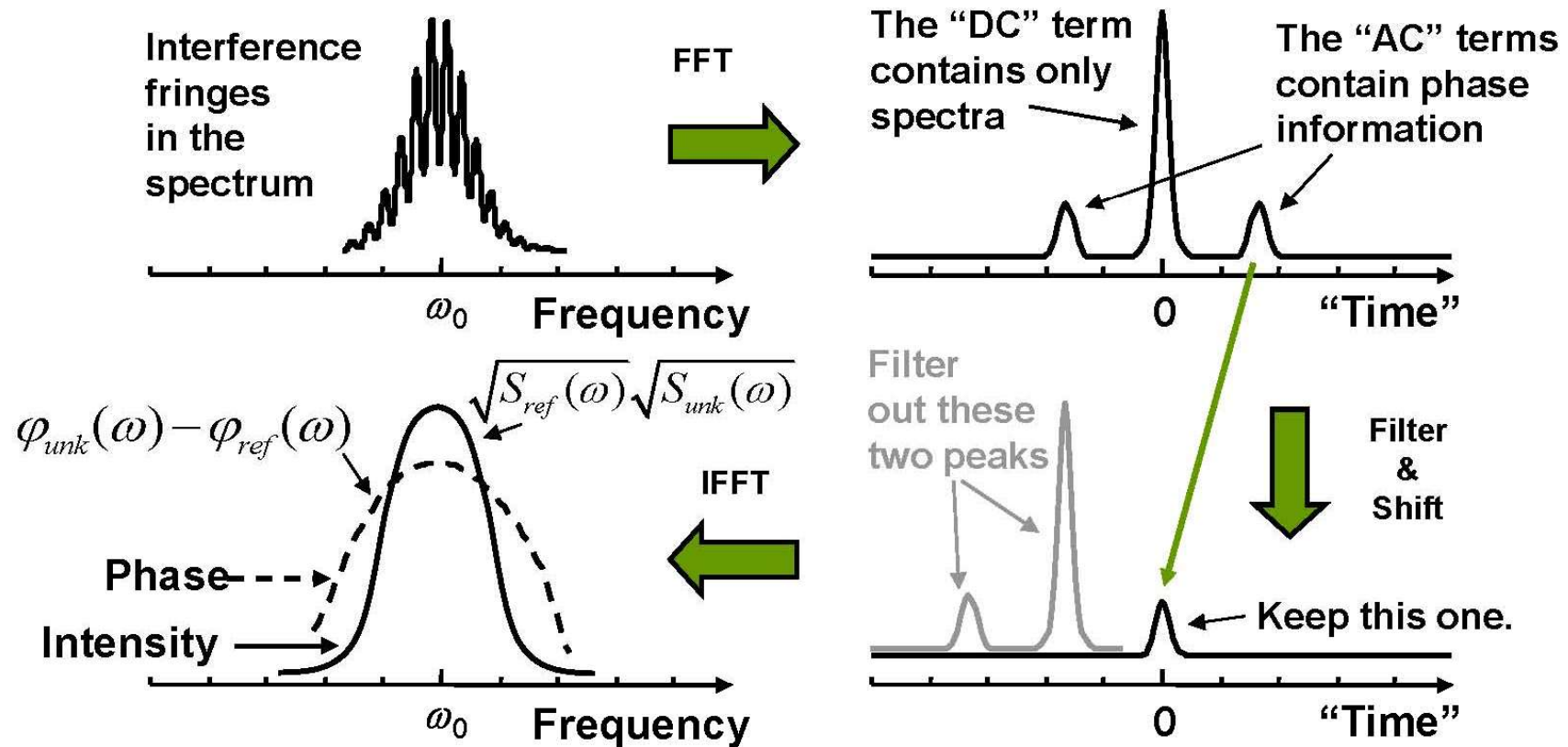
Spectral interferometry (Rick Trebino)



The spectrum in case of a known and an unknown pulse:

$$S_{SI}(\omega) = S_{ref}(\omega) + S_{unk}(\omega) + 2\sqrt{S_{ref}(\omega)}\sqrt{S_{unk}(\omega)}\cos[\varphi_{unk}(\omega) - \varphi_{ref}(\omega) + \omega T]_B$$

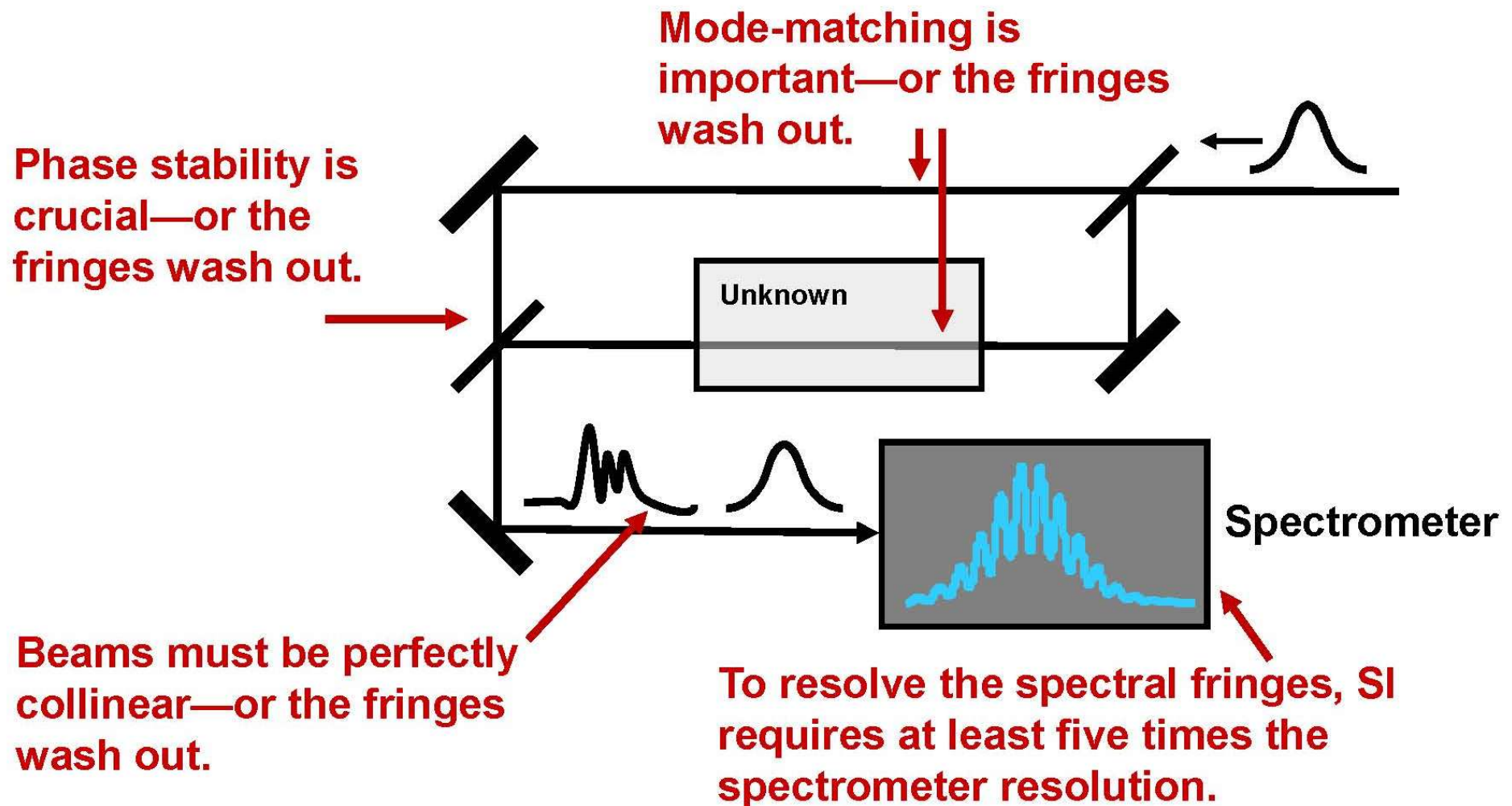
Retrieving the pulse in spectral interferometry



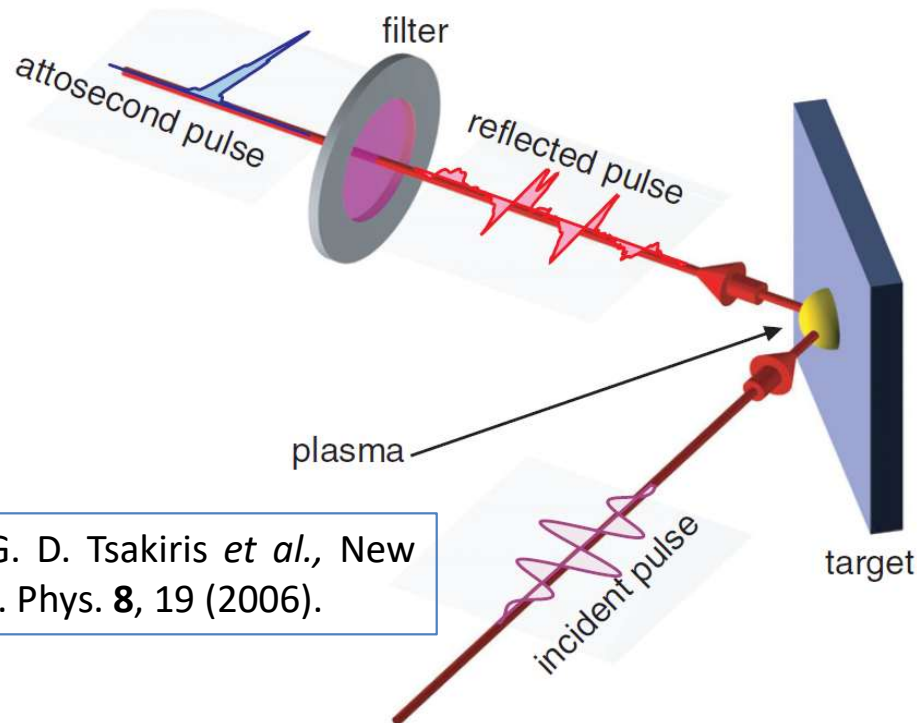
This retrieval algorithm is quick, direct, and reliable.
It essentially uniquely yields the pulse.

Spectral Interferometry: Experimental Issues

The interferometer is difficult to work with.



High harmonics from plasma mirrors



G. D. Tsakiris *et al.*, New J. Phys. **8**, 19 (2006).

Source of radiation:

Critical layer oscillates due to the $\mathbf{v} \times \mathbf{B}$ field (relativistic intensity)
Doppler-shift of reflected radiation results in harmonics

- No intensity limit
- No upper limit for the order of harmonics
- Sub-attosecond pulses are possible
- High conversion efficiency

Relativistic threshold (ROM):

- Oscillatory velocity of electron:
 $v \sim c$
- Oscillation amplitude larger than laser wavelength
- Magnetic field is not negligible:
- $(\mathbf{v} \times \mathbf{B} \sim e\mathbf{E})$

Relativistic (ROM) harmonics

Incoming laser: $E_{inc}(t) \sim \sin(\omega_L t)$

$\mathbf{v} \times \mathbf{B}$ Lorentz force causes the oscillation of the critical surface,
as $\mathbf{v} \sim \sin(\omega_L t)$ and $\mathbf{B} \sim \sin(\omega_L t)$

Position ($\sim$ ponderomotive force): $X(t') = A_m \sin(2\omega_L t' + \phi)$

Assuming a stepwise function for the plasma: $X(t)$

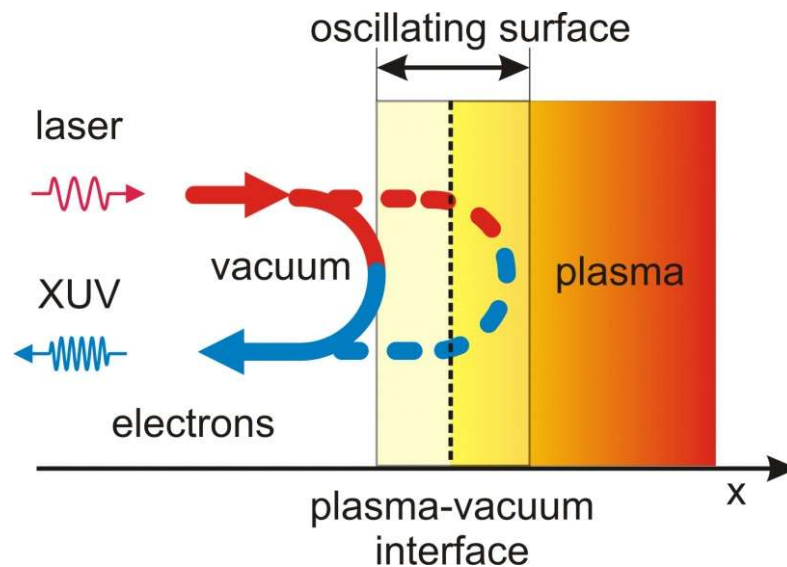
Retarded reflected pulse: $t' = t + X(t')/c$

For the full roundtrip of reflected light:

$$t + 2(t' - t) = t + 2(R + x(t'))/c,$$

thus the reflected field: $E_{refl}(t) = \sin(\omega_L t + 2k_L X(t'))$

Neglecting the R phase factor, this is the source of harmonics.



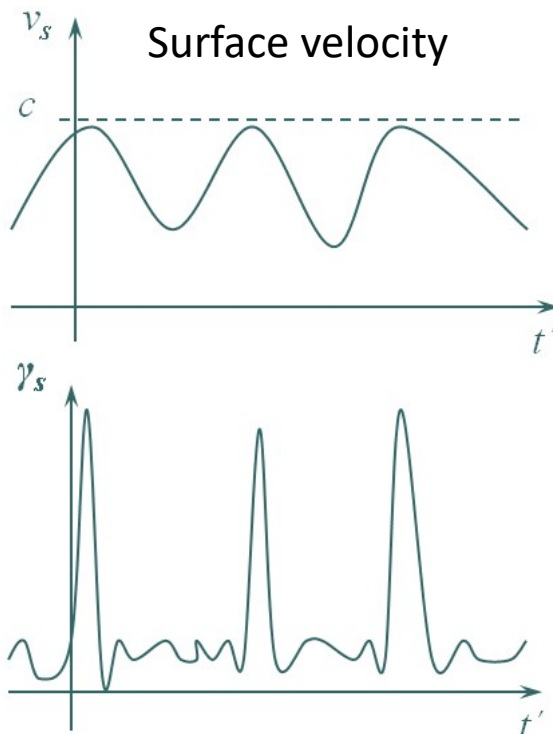
Theory of
Lichters and Bulanov
+PIC simulations

Relativistic harmonics

Alternative analytical model:

γ -spiking model

(Similarity solution of the relativistic Vlasov equation)



$$\gamma_s = \frac{1}{\sqrt{1 - (v_s/c)^2}}$$

**High harmonics
are generated
at the peaks.**

$$\eta(n) \sim n^{-8/3}$$

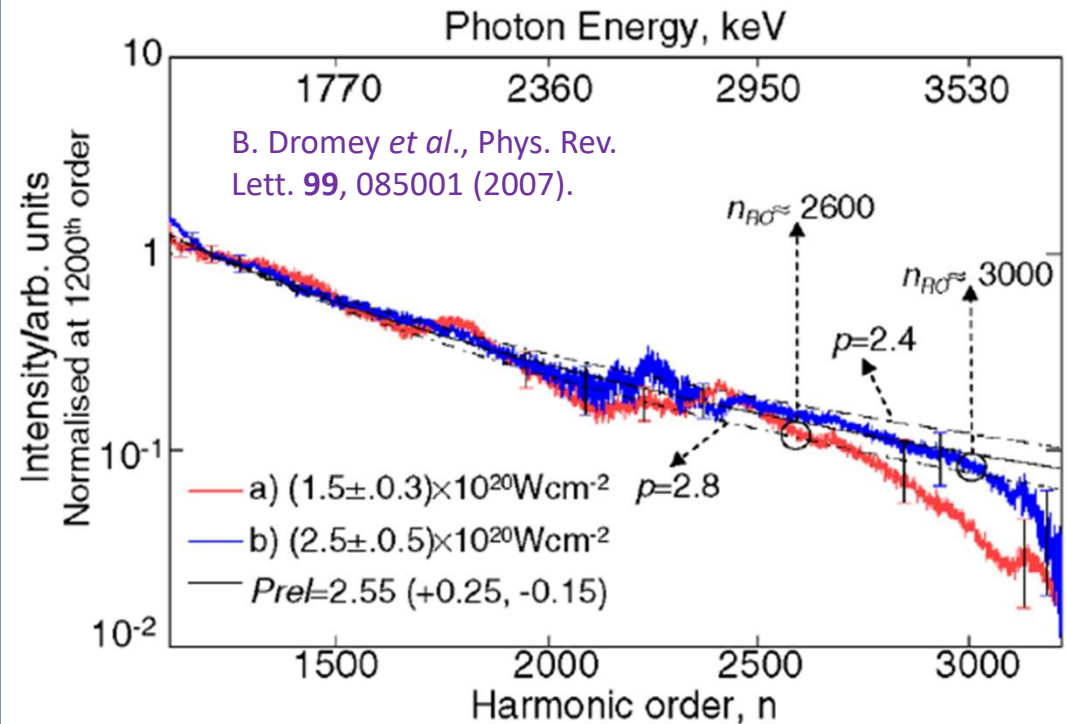
$$n_{\text{cutoff}} \sim \gamma_{\text{max}}^3$$



γ spiking causes the short pulse
duration!

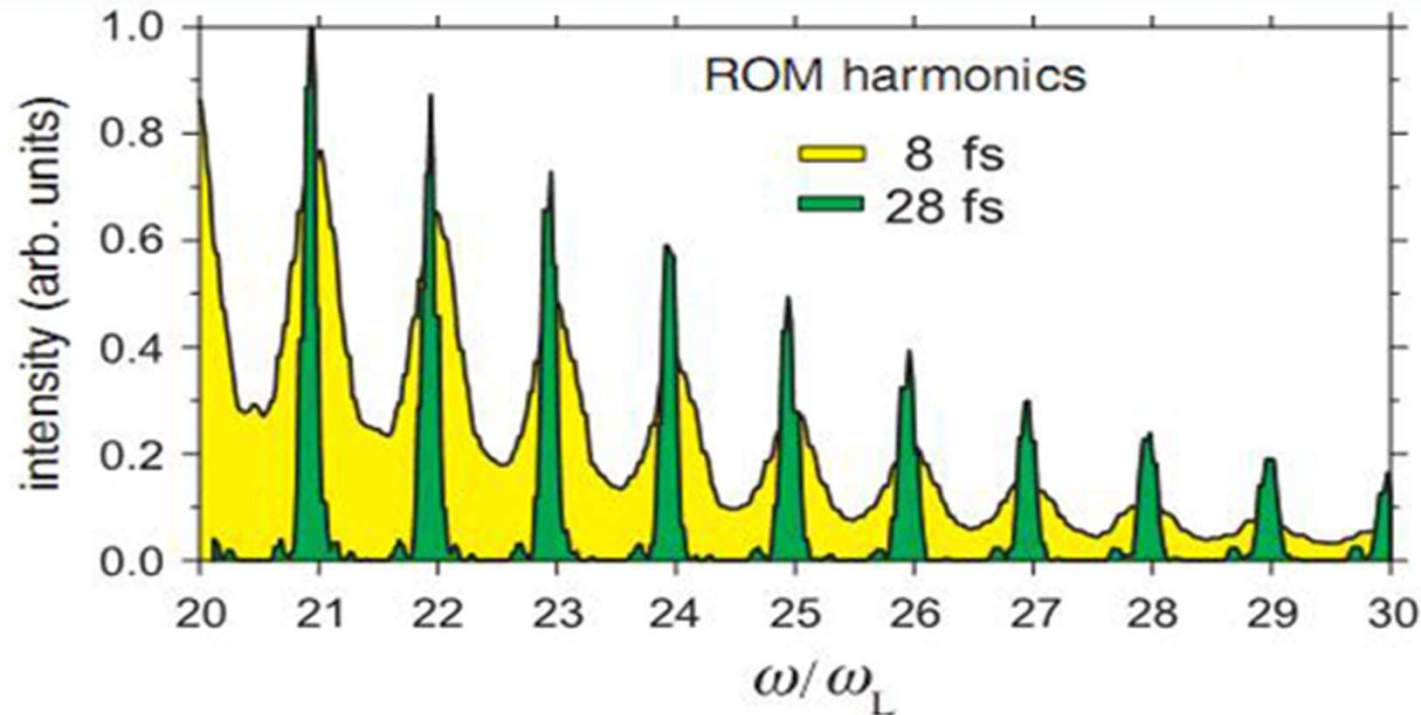
T. Baeva *et al.*, Phys. Rev. E **74**, 065401(R) (2006).

Harmonics up to several keV energy



- $n > 3200 \rightarrow 3.8 \text{ keV}$
- $\eta \sim n^{-2.5} - n^{-3}$ scaling law
- Beaming within $\sim 4^\circ$
- 10 keV zeptosecond pulses are - in principle - possible

Aim: isolated attosecond pulses



Harmonics spectra broadens when laser pulse is shorter



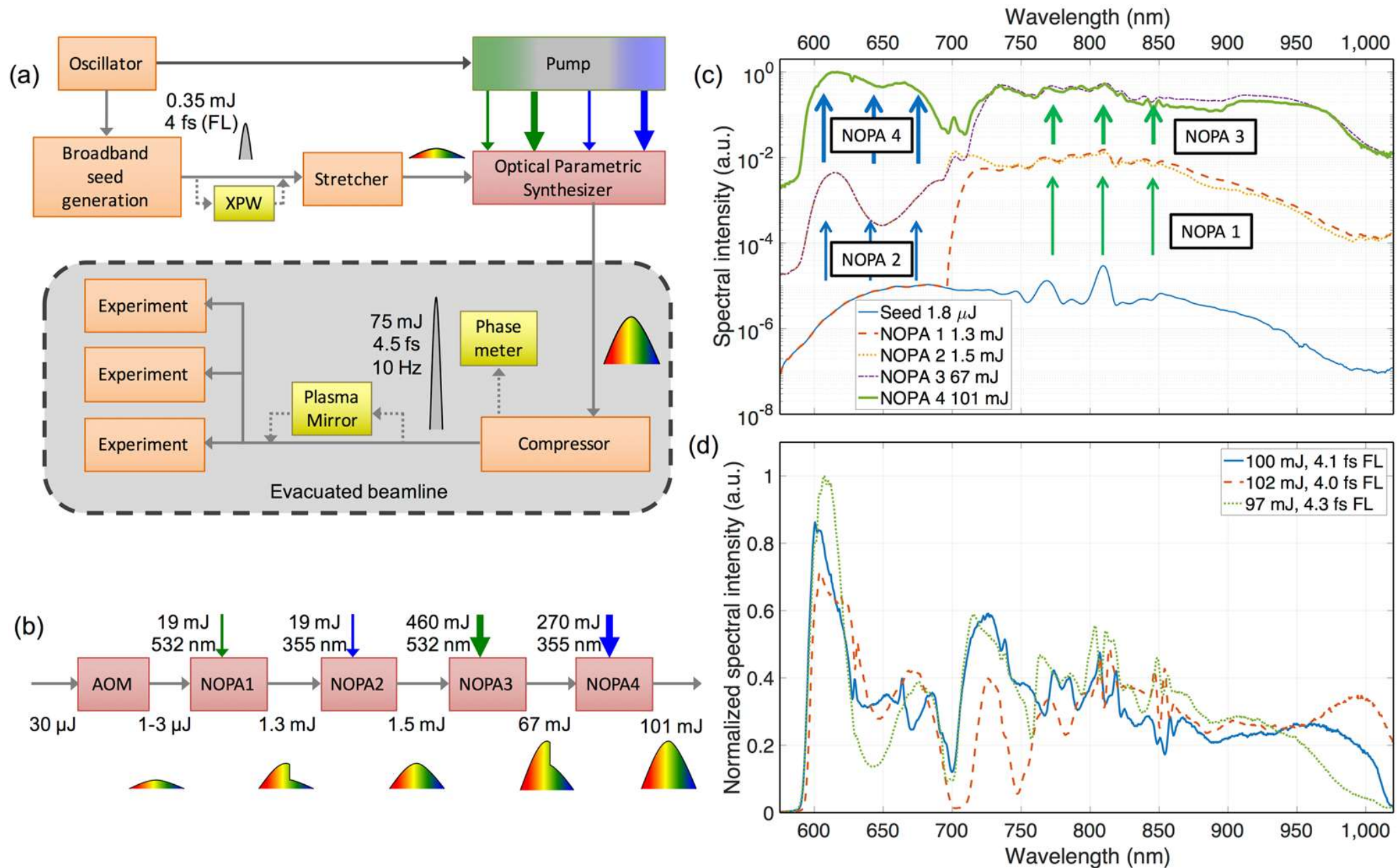
Continuum expected for 4.5 fs pulses



Generation of isolated attosecond pulses

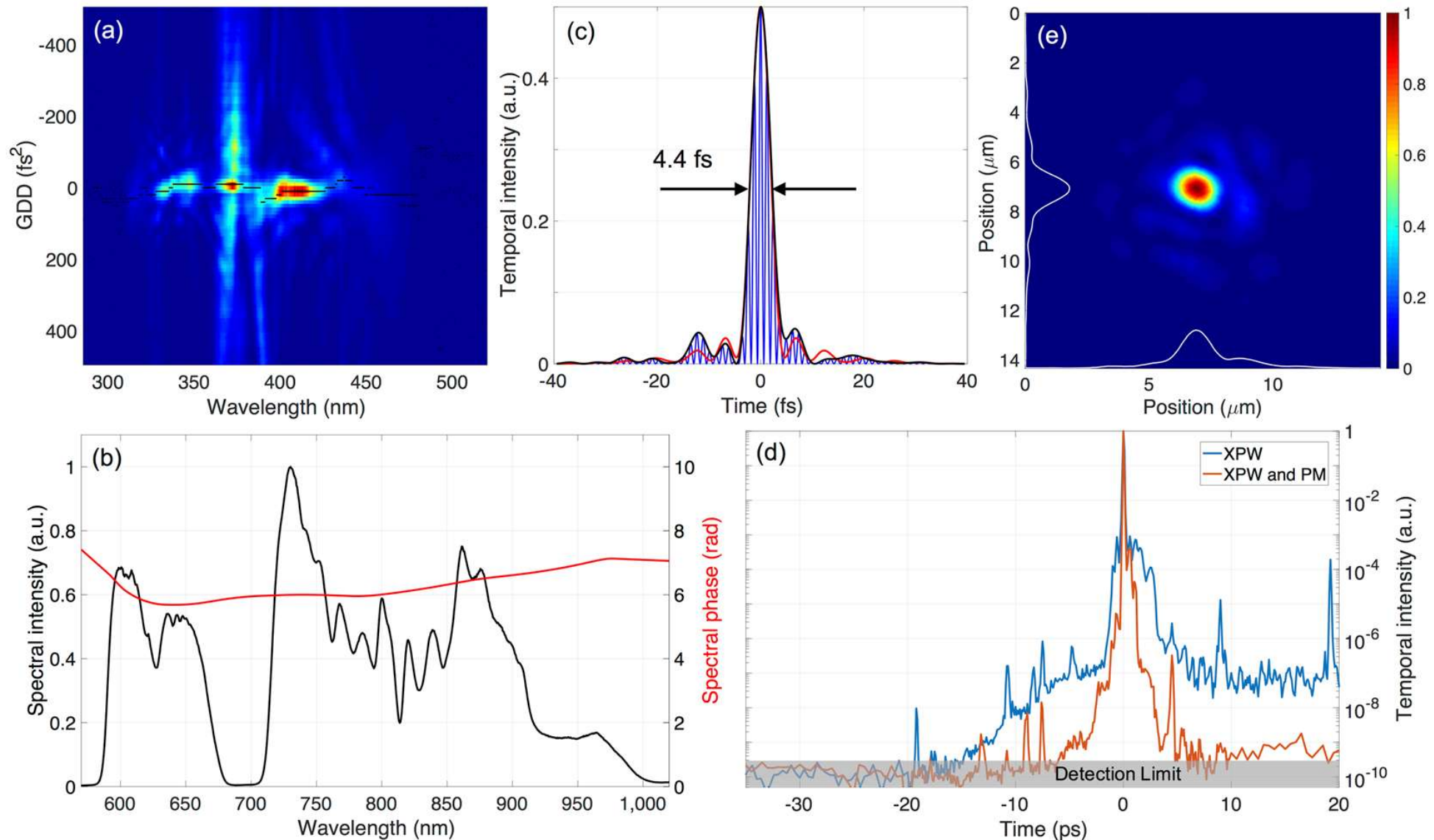
Motivation!

Az LWS20 laser



Beam characterization

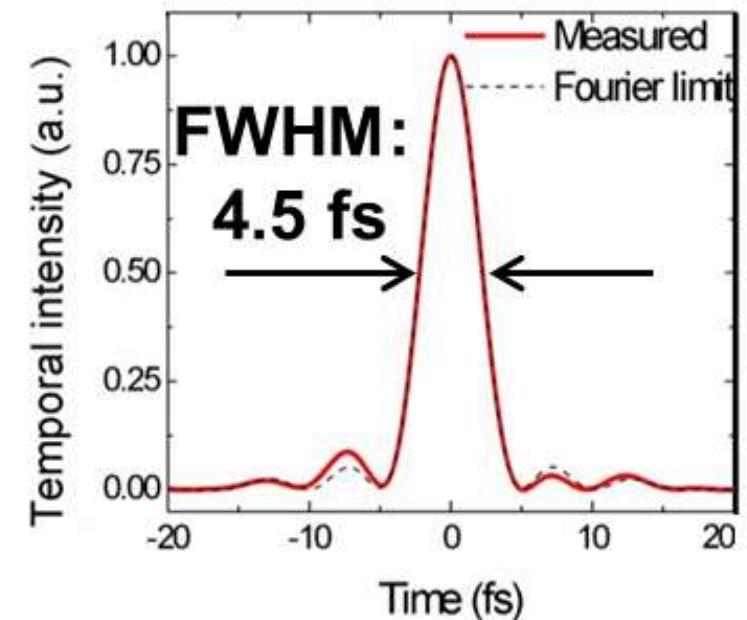
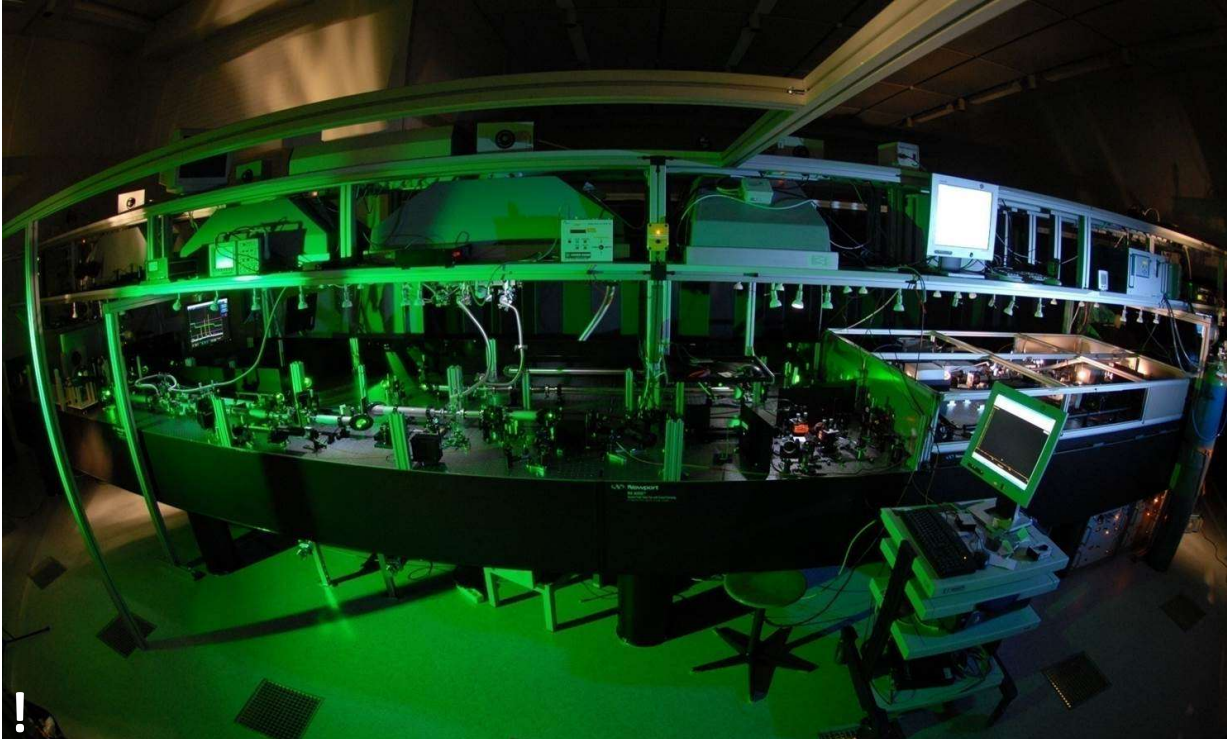
Measuring chirp from 2ω generation



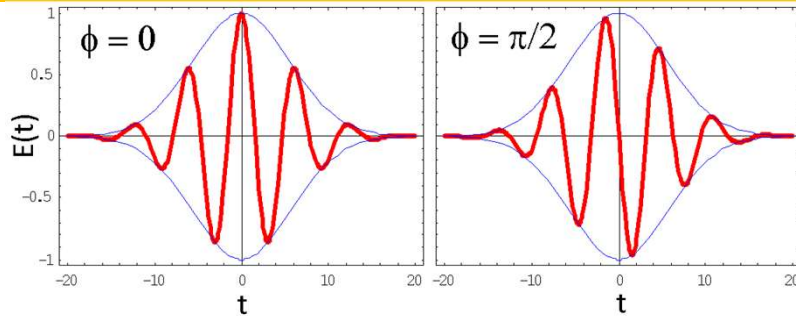
High contrast with XPW plus plasma mirror

Light-Wave-Synthesizer (LWS) laser parameters

- Spectrum: 580 – 1020 nm
- Reprate: 10 Hz
- Focusability: 1.3 μm
- Intensity: $> 10^{20} \text{ W/cm}^2$
- Energy: 40 mJ on target
- Pulse duration: 4.5 fs (FWHM)
- Power: 16-18 TW
- Contrast: $> 3 * 10^{-8}$ (4 ps), 10^{-19} (30 ps)

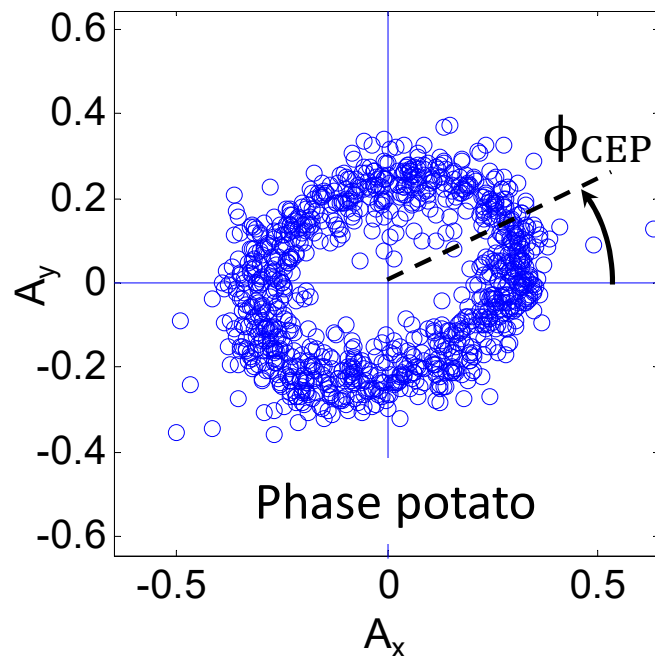


Measuring CEP phase

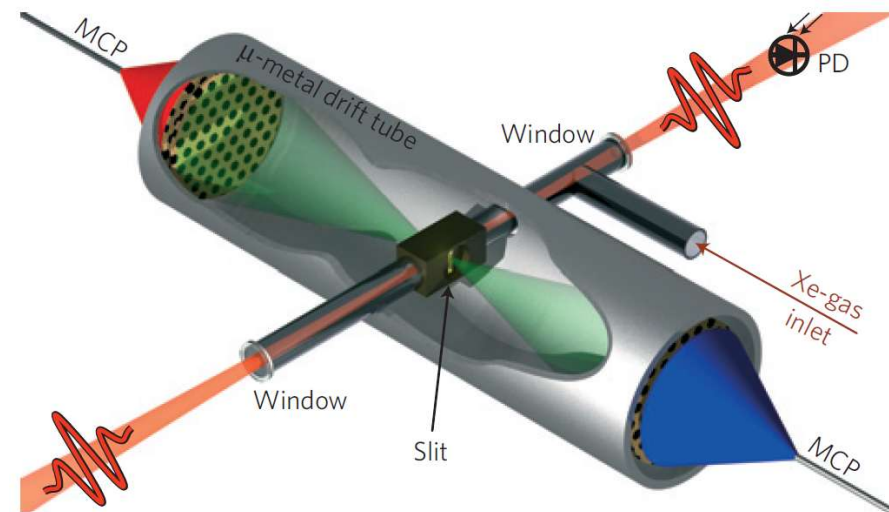


$$E(t) = E_0(t)\cos(\omega t + \phi)$$

CEP tagging:



- Detecting electrons generated by above threshold ionizations (ATI) for each laser shot.
- ATI spectra of few cycle ATI is strongly phase dependent.
- Spectra in counterpropagating directions are asymmetric.

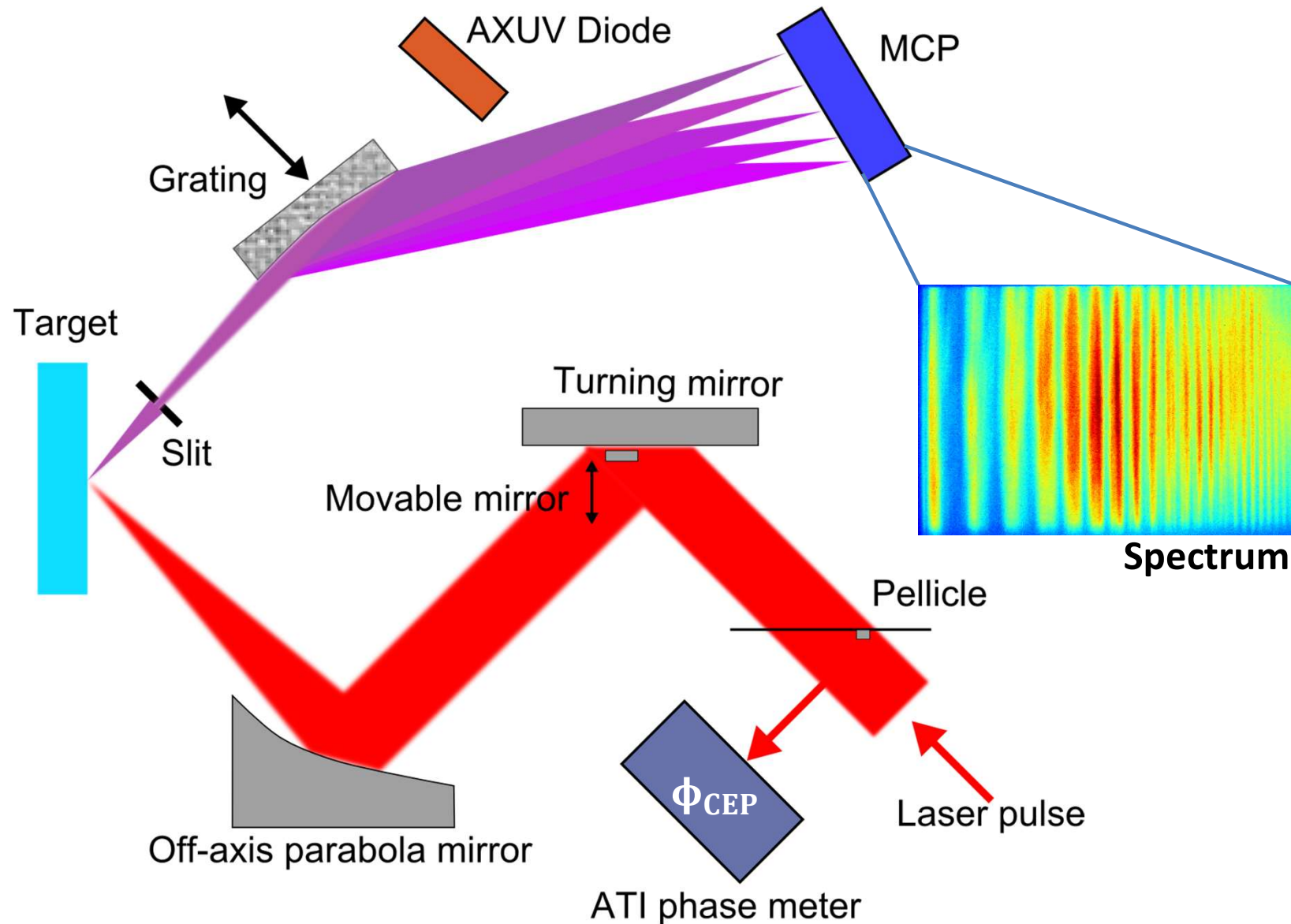


T. Wittman *et al.*, Nature Physics **5**, 357 (2009).

A_x and A_y : phase asymmetry parameters from certain ranges of ATI spectra.

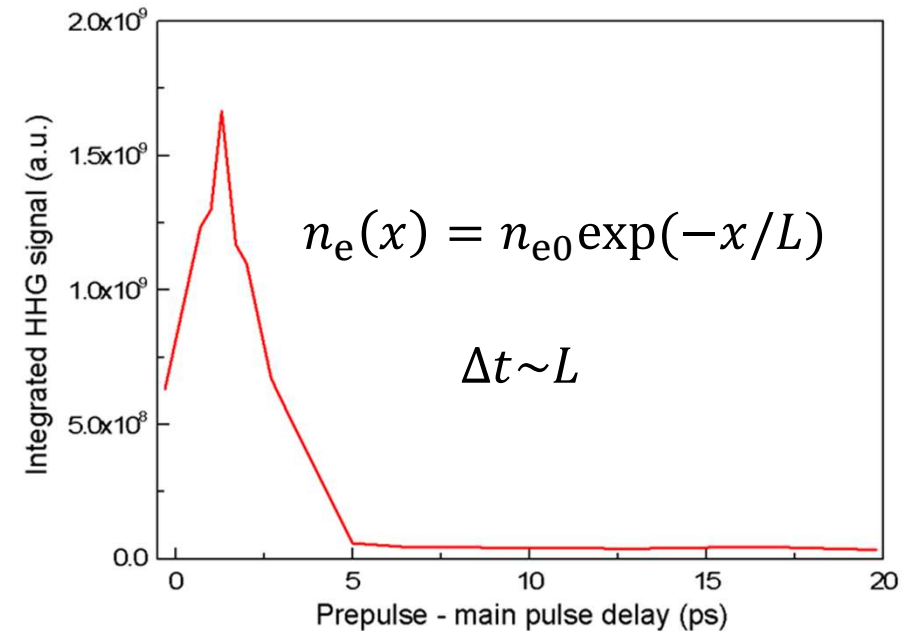
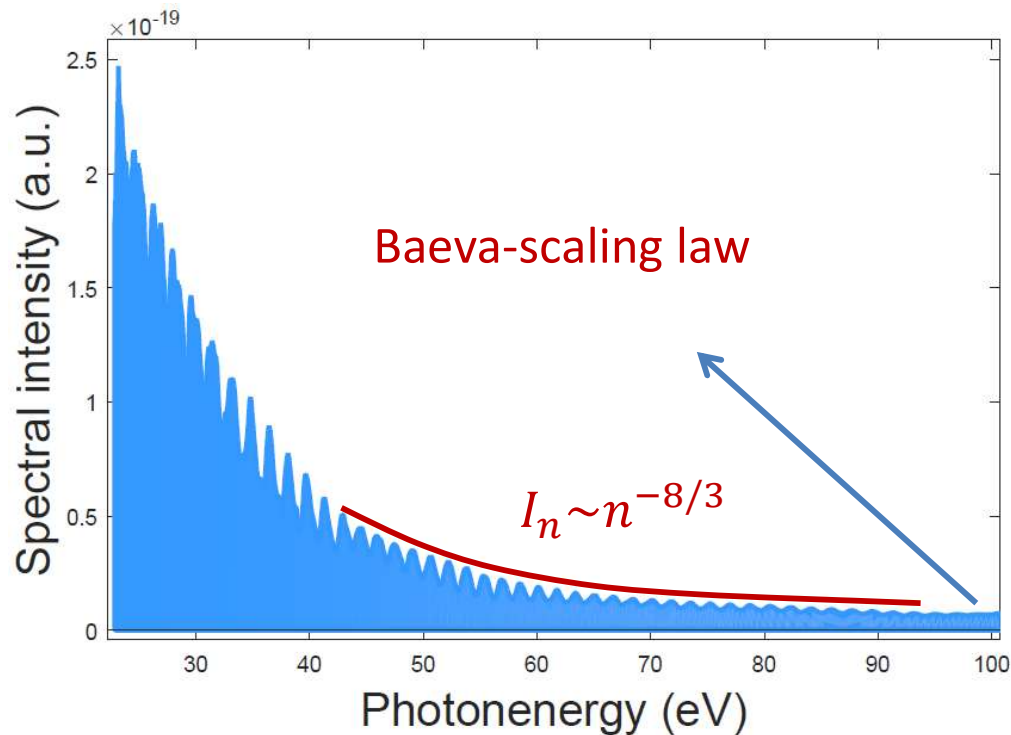
ϕ_{CEP} value: 1-DI TDSE (time-dependent Schrödinger equation) simulations.

Experimental arrangement

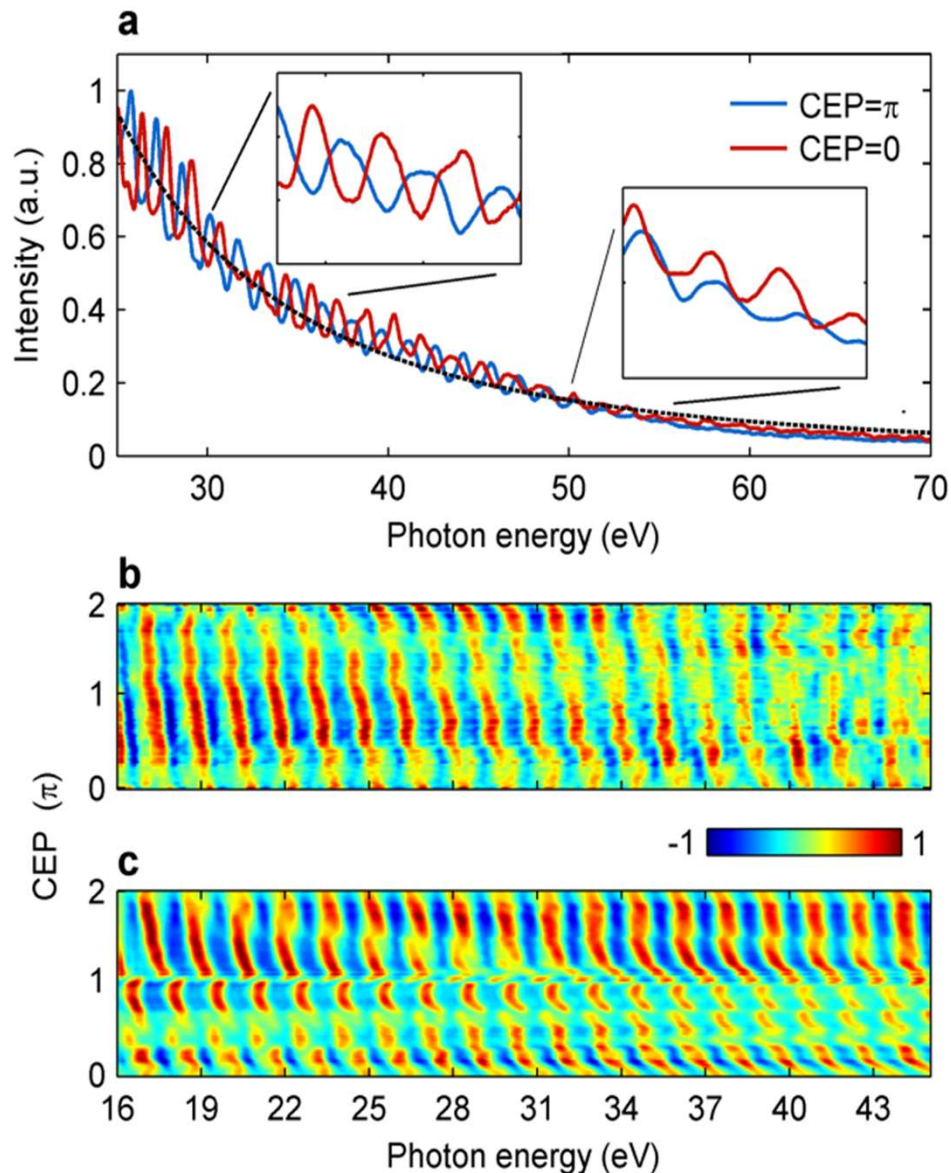


Harmonics up to 100 eV energy

- HHG optimization with prepulse
↓
The laser contrast is „too good“
- In this case no plasma mirror was used



CEP-dependent XUV spectrum



- a) Measured spectra shifted by π phase
Dotted line: Fit with -2.62 exponent.
- b) CEP dependence of measured spectrum
- c) PIC simulations for $L/\lambda=0.13$ plasma length

Measured harmonics are NOT integer multiples of the laser frequency.

They are shifted often with a half period.

Increasing plasma scalelength the CEP dependence is more complicated.

It is a proof that spectra of relativistic harmonics depend on the electric field and not on the intensity.

Spectra contain the interference of the attosecond pulse train.

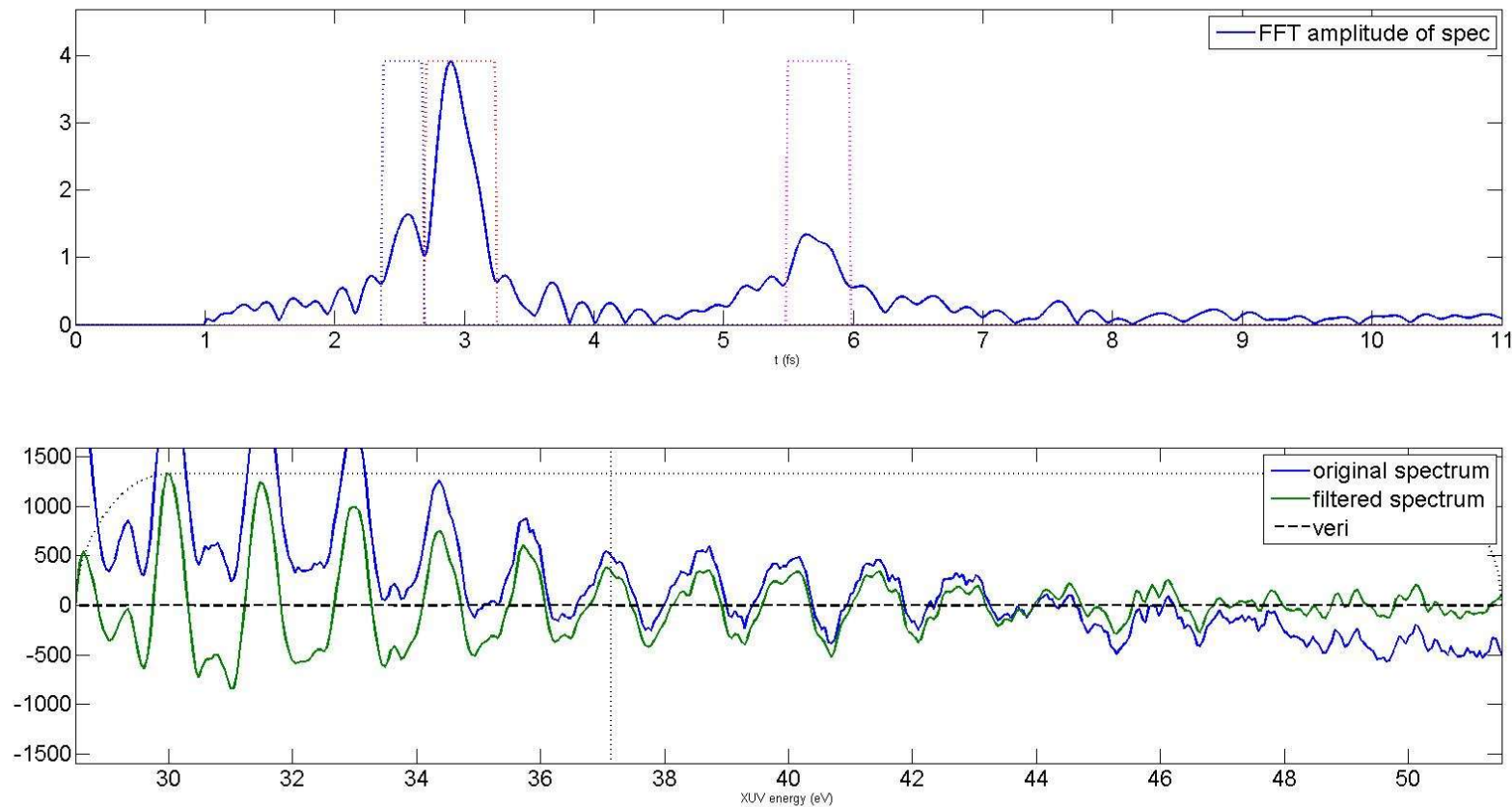
Spectrum analysis therefore may lead to the demonstration of isolated attosecond pulses.

Fourier transformation of spectra

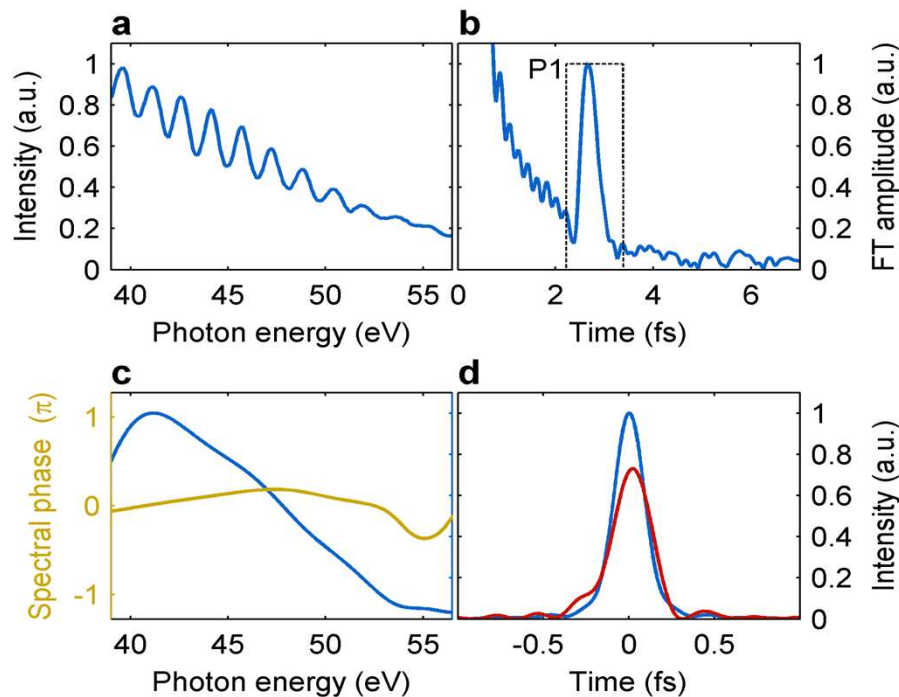
Fourier transformed spectra without time 0.

3 peaks: 3 attosecond pulses generated by the ~ 5 fs laser pulse produce 3 peaks. Interference of peaks 1-2, 1-3, 2-3.

It is the basis of spectral interferometry. CEP dependent.



Spectral interferometry



Simulation:

$\phi_{\text{CEP}}=0$: ~ 2 attosecond pulses.

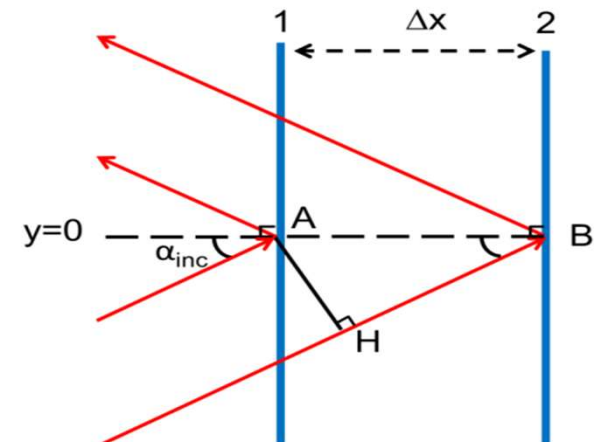
Exp.: (a) Interference of 2 pulses.

b): Fourier transformation gives 1 peak (P1), which contains amplitude and phases.

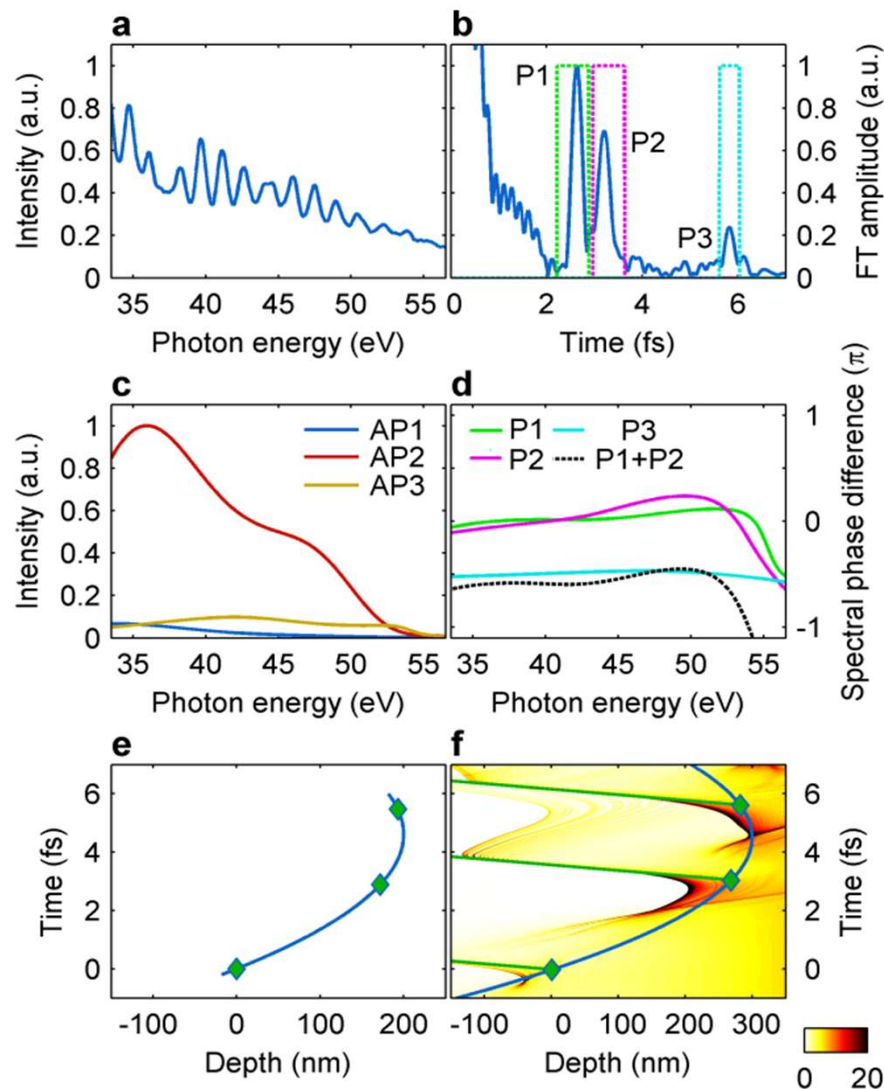
c) Phase difference: $S = \sqrt{S_{AP1} S_{AP2}}$
Assuming 1st pulse with constant phase (sim).

d) Attosecond pulse is not sensitive to phase (red curve is with phase).

Experimental time difference between the
2 attosecond pulses: $2.7 \text{ fs} > T_L = 2.55 \text{ fs}$.
Simulation: Radiation pressure pushes the plasma.
Attosecond pulses originate from different depths.
Delay: 150 as
Spatial difference: 40 nm (perpendicular to target).



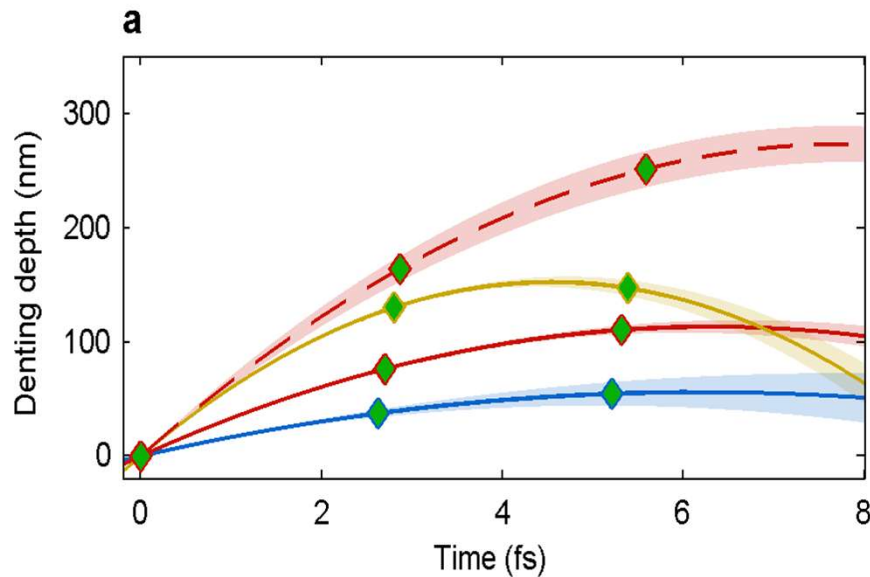
Spectral interferometry of 3 attosecond pulses



- a) Spectrum for $\phi_{\text{CEP}} = \pi$ and $L/\lambda = 0.5$
 b) FT in range 33-57 eV: P1, P2 and P3
 Interference of 3 pulses:
 AP2&AP3, AP1&AP2 és AP1&AP3, respectively.
 Dotted lines: limits of inverse FT .
 c) Reconstructed spectra of 3 pulses.
 d) Spectral phase difference neglecting linear contribution.
 e) Reconstructed plasma surface dynamics from timing of 3 attosecond pulses.
 f) Electron density from 1D LPIC simulation ($\phi_{\text{CEP}} = \pi$ and $L/\lambda = 0.4$).
 Green diamonds: Time of XUV generation.
 Blue line: Parabolic interpolation from the 3 points, describing surface motion.
 Assumption: At least 1 pulse has a flat phase.

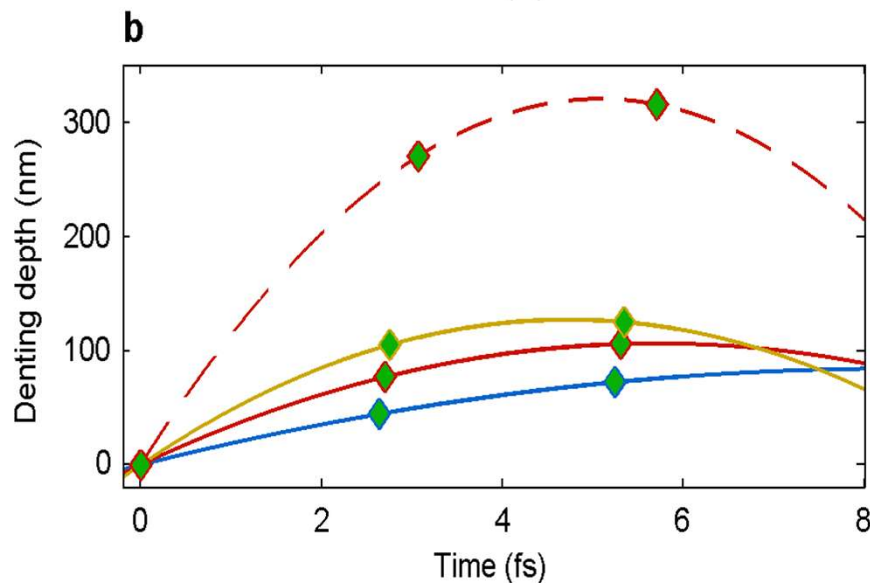
The results prove that the attosecond pulses do not have atto-chirp.

Plasma (critical) surface denting can be mapped



a) Experiment and b) simulation, $L/\lambda=0.13$:
| $\varphi=7\pi/6$, | $\varphi=\pi/2$, | $\varphi=5\pi/3$
Red dashed: $L/\lambda=0.5$, $\varphi=7\pi/6$. Different scale

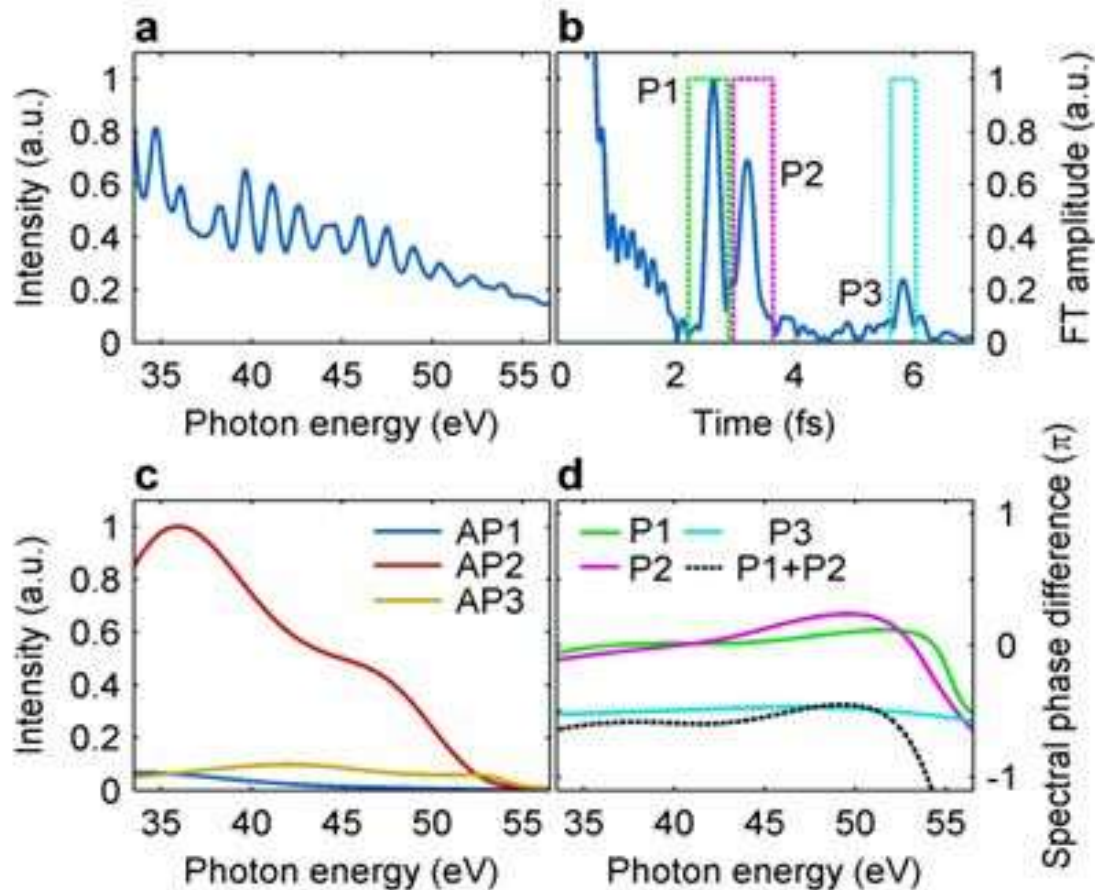
Longer plasma: larger motion.
Phase are changed so,
that P3 is above noise level



Good agreement with simulations (b).
Deviations due to 3D effects in the λ^3 region.

CEP dependence of the shape of parabolas:
Dynamics of plasma surface is field dependent.
It is not only intensity dependent
as in case of longer pulses.

Application of the method for the generation of isolated attosecond pulses

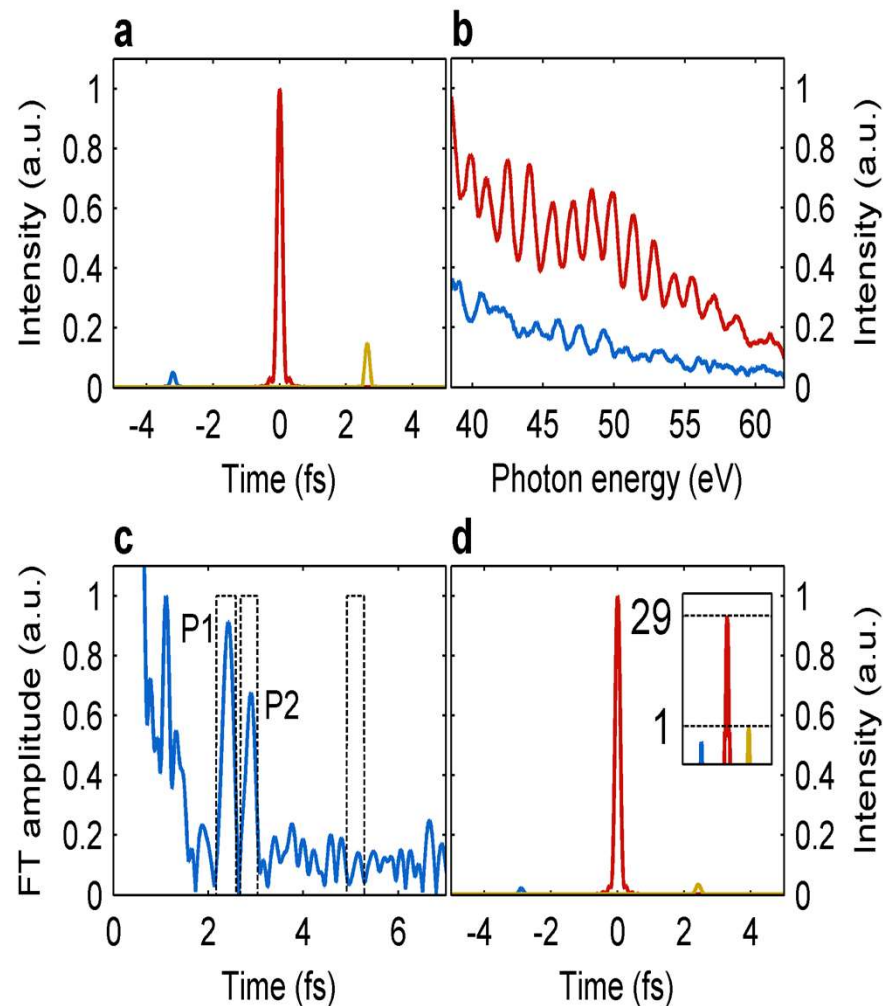


If the central attosecond pulse is dominant, it is ~ 5 times more intense than the first and third pulse.

According to the calculations the dominance is stronger if P1 and P2 peaks are much stronger than P3.

Let us find such spectrum and carry out the procedure!

Isolated attosecond pulse



a) Shows the previous case.

b) The blue spectrum for $\varphi_{\text{CEP}} = 11\pi/6$ and $L/\lambda=0.5$ compared with the red spectrum, $\varphi_{\text{CEP}} = \pi$ which gives 2 peaks of equal sizes.

c) Fourier transformed XUV spectrum. P1 and P2 is the interference of the strongly isolated pulse with the weak side pulses. P3 is below noise level because side pulses are weak.

d) Reconstructed time dependence, with an isolation degree larger than 30.

The isolation degree of 30 was the first demonstration of isolated attosecond pulse generated from laser plasmas on solid surfaces (plasma mirrors). Spectral interferometry proved to be well applicable for the analysis of attosecond pulse trains for <2 cycle laser pulse.

Conclusion

- High harmonics were generated in laser plasmas on solid surfaces as a dependence of tagged CEP monitoring.
- Spectral interferometric analysis showed that isolated attosecond pulses were generated for a certain CEP value.
- Temporal and spectral phases of the pulses were using spectral interferometry.
- The second and third order spectral phases of attosecond pulses are similar, the compression is the same, there is no attochirp.
- Spectral analysis reveals the spatial and temporal motion of the reflecting plasma surface.
- Motion of the plasma surface depends on the electric field of the laser pulse, not only on the intensity.

CEP stabilized direct experiments will be able to generate directly isolated attosecond pulses. LWS100 makes it possible.

<https://www.nature.com/articles/s41566-025-01720-2>

Thank you for your attention!

Questions?