Three ways to choose between two attosecond pulses

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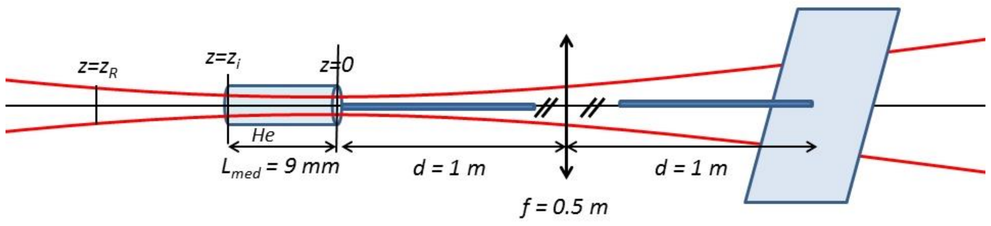
Abstract: Based on numerical calculations, we propose a new experimental beamline for high-order harmonic generation (HHG) of mid-infrared femtosecond laser pulses. The advantage of the proposed configuration is that it allows the generation of two successive (high-harmonic) attosecond pulses having separate spectral content, but both in the XUV regime. We demonstrate that the two attosecond pulses have naturally separated spectral content, which results from macroscopic HHG in Helium gas medium. With the proposed experimental parameters the gap between the two spectral domains is close to the onset of the water window (282–533 eV). Coherent light sources in the water window are important for the ultrafast dynamic imaging of biological samples in water medium. The spectral separation of successive attosecond pulses is an example of space-time coupling in nonlinear optics, good candidate to be further explored and exploited both theoretically and experimentally.

Proposed configuration



General parameters of the case study:

- → λ = 1700 nm; E = 1 mJ pulse energy; τ = 6 fs duration FWHM;
- → focused with f = 50 cm mirror into L =9 mm He static cell at p = 200 torr;
- → z = [-9; 0] mm, laser beam waist of w_0 = 90 μm and z_R = 15 mm Rayleigh range;
- → $I_{peak} = 9 \cdot 10^{14}$ W/cm² at the cell entrance;
- → 2.3% ionization of the neutral He population.

Model

1. Propagation of the laser pulse through the gas medium

$$\nabla^2 E_1(r,z,t) - \frac{1}{c^2}\frac{\partial^2 E_1(r,z,t)}{\partial t^2} = \frac{\omega^2}{c^2}(1-\eta_{eff}^2)E_1(r,z,t)$$

$$\eta_{eff}(n_a,n_e,r,z,t) = \eta_0(n_a) + \eta_2(n_0)I(r,z,t) - \frac{\omega_p^2(n_e,r,z,t)}{2\omega^2}$$

2. Single-atom response in laser-atom interaction

Lewenstein (1994) Strong Field Approximation

$$P_{nl}(t) = 2Re\left\{i\int_{-\infty}^{t} dt' d^*[p_{st}(t',t) - A(t)]\, E_1(t') exp\left[-\int_{-\infty}^{t'} w(t'')dt''\right] exp[-iS_{st}(t',t)] \cdot d[p_{st}(t',t) - A(t')]\right\} \cdot [n_0 - n_e(t)]$$

3. Harmonic field propagation

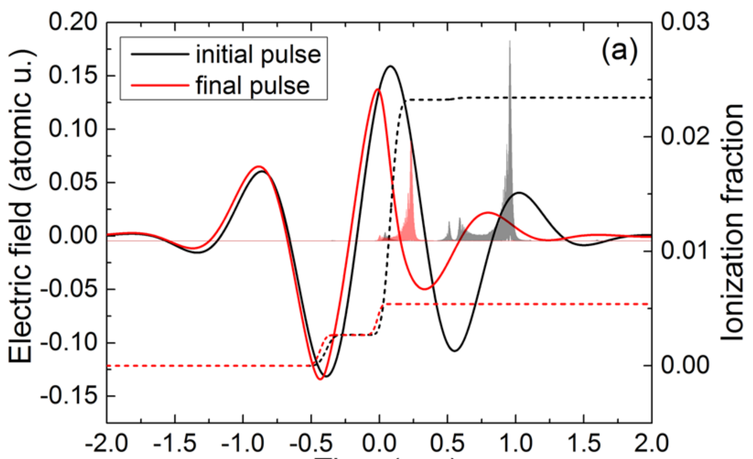
$$\nabla^2 E_h(r,z,t) - \frac{1}{c^2}\frac{\partial^2 E_h(r,z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P_{nl}(r,z,t)}{\partial t^2}$$

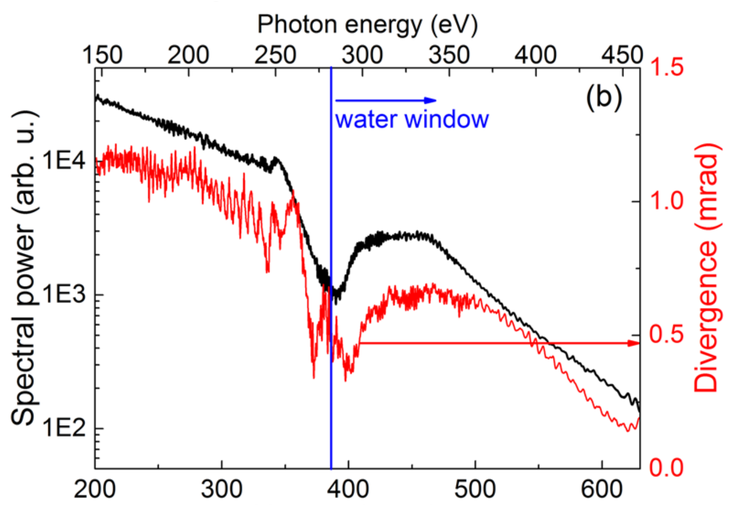
Fourier tr.

4. Transport the XUV frequency components from near-field to far-field: Hankel transform

$$E_\omega(r,z) = -\frac{2\pi}{\lambda B} exp\left[ik\left(z + \frac{Dr^2}{2B}\right)\right]\int_0^a E_\omega(r_1, z_1 = 0) exp\left(\frac{ikAr_1^2}{2B}\right) J_0\left(\frac{krr_1}{B}\right) r_1 dr_1$$

Driving pulse propagation and harmonic spectrum

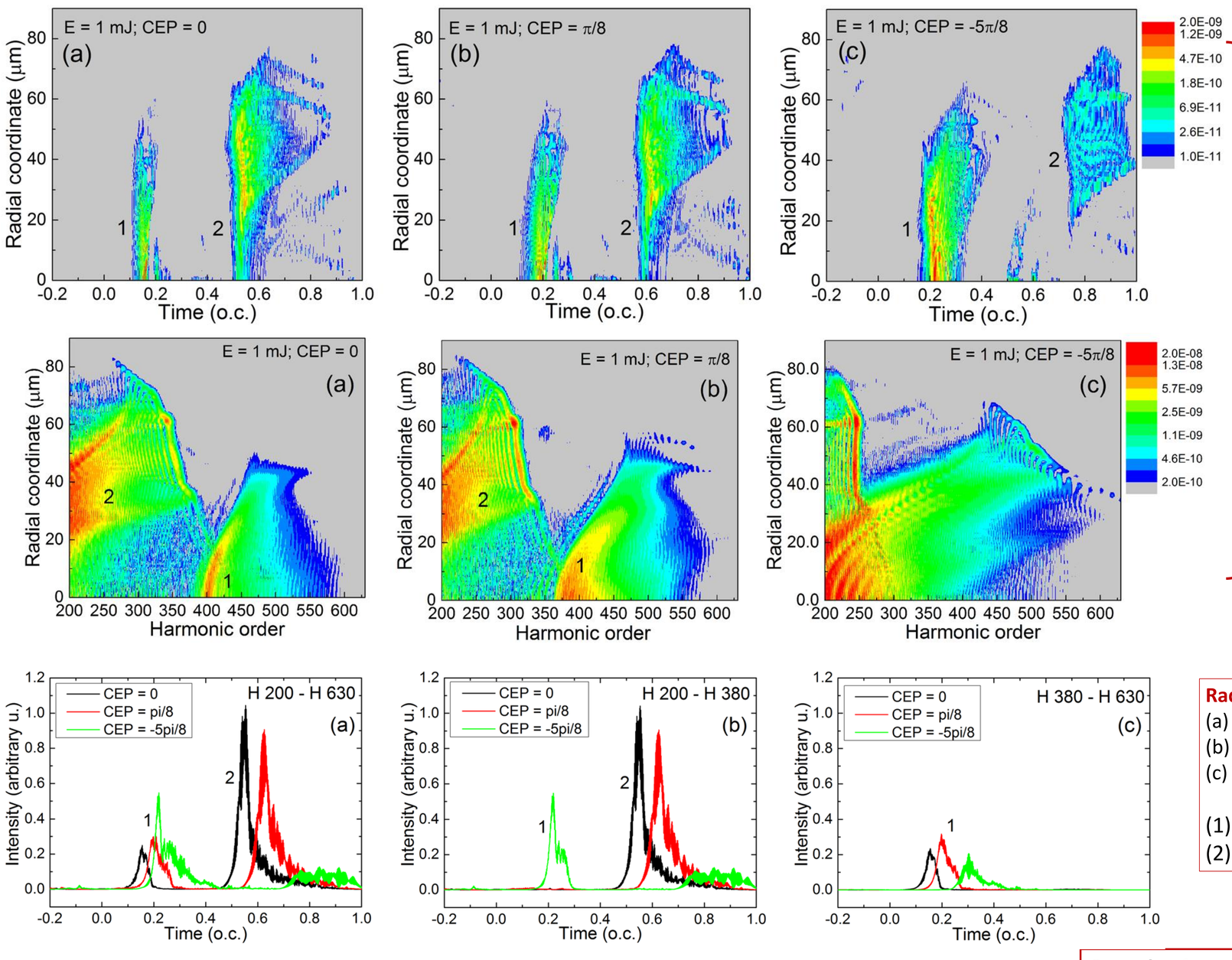




(a) Driving pulse shape (solid line) along with the temporal ionization dynamics (dashed line) at the beginning (black) and at the end (red) of the propagation. Shaded in the background we show the dipole radiation filtered between harmonic orders H200–630.
→ Steep ionization front causes self-phase modulation and intensity decrease.
→ Two elementary dipole emission events: (1) T=0.2 o.c. (2) T=0.6 o.c.

(b) (black curve, left axis) Radially integrated harmonic power spectrum at the end of the interaction region.
(red curve, right axis) Radially averaged divergence of harmonics at the medium exit.
Spectral gap @ the beginning of the water window (~280 eV)

Mechanism of the temporal AND spectral attosecond pulse separation

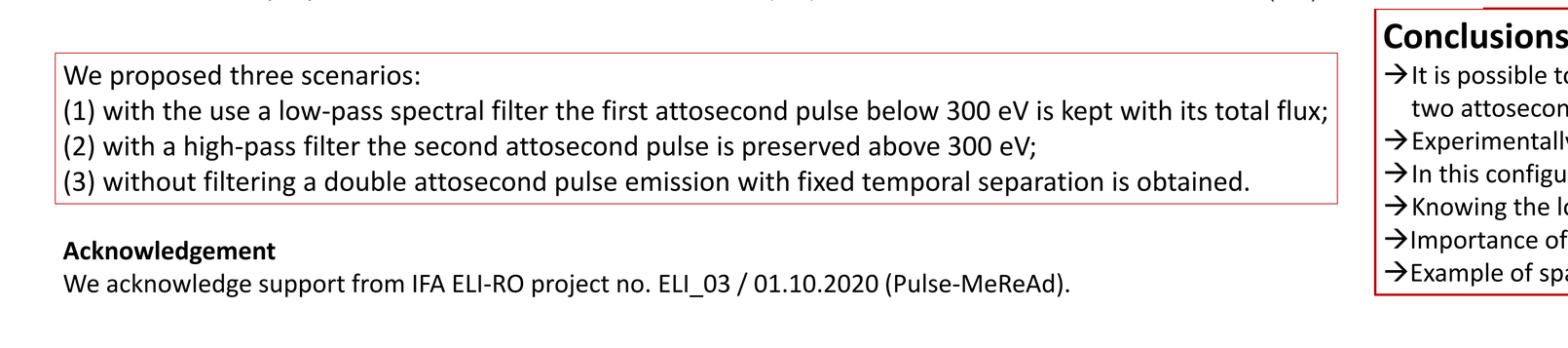


Temporal–radial maps of the attosecond pulses in the H200–H630 spectral domain. Snapshots are taken at the exit of the interaction region.
Color scale spans two orders of magnitude, logarithmic scale, arbitrary units.
In panel (a) CEP = 0; (b) CEP = π/8; (c) CEP = −5π/8.

Emission (1): earlier in time, concentrated on-axis;
Emission (2): later in time, mostly off-axis;

Spectral-radial maps of the harmonic radiation at the exit of the interaction region.
Snapshots are taken at the exit of the interaction region.
Color scale spans two orders of magnitude, logarithmic scale, arbitrary units.

Emission (1): contains high-energy spectral components >H400;
Emission (2): contains spectral components up to ~H350



Radially integrated attosecond pulses in the far-field.
(a) Full spectrum between H200–H630.
(b) The lower part of the spectrum between H200–H380.
(c) The high part of the spectrum between H380–H630.

(1) the spectral separation is CEP-dependent;
(2) the spectral gap can be controlled by the CEP of the driving pulse

We proposed three scenarios:
(1) with the use a low-pass spectral filter the first attosecond pulse below 300 eV is kept with its total flux;
(2) with a high-pass filter the second attosecond pulse is preserved above 300 eV;
(3) without filtering a double attosecond pulse emission with fixed temporal separation is obtained.

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Conclusions

→ It is possible to create the conditions for the natural temporal AND spectral separation of two attosecond pulses
→ Experimentally feasible parameters
→ In this configuration one can keep the total flux of one emission, which is an advantage
→ Knowing the low efficiency of the HHG process especially with increasing driving wavelength
→ Importance of the macroscopic propagation effects in shaping the final XUV emission
→ Example of space-time coupling in nonlinear optics