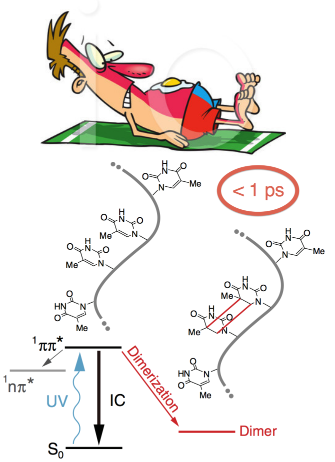
Towards measuring attosecond dynamics with HHG driven by few-cycle long wavelength fields

D. R. Austin, T. Witting, T. Siegel, A. S. Johnson, F. McGrath, P. Hawkins, P. Matía-Hernando, Z. Diveki, S.J. Weber, A. Zaïr, J.W.G. Tisch, J.P. Marangos

Imperial College London

Motivation

- Initial steps of photo-chemistry
  - UV DNA damage



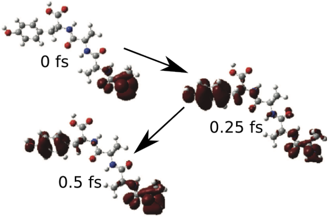
Thymine dimerization in DNA

Schreier WJ, *et al.* *Science* **315**, 625 (2007)

Motivation

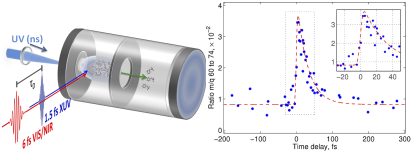
- Initial steps of photo-chemistry
  - UV DNA damage
- Charge migration: beating of multiple electronic states
  - ubiquitous
  - $\approx 1\,\mathrm{fs}$ timescale

Kuleff AI *et al. J. Phys. B: At., Mol. Opt. Phys.* **47**, 124002 (2014)



Charge migration in tetrapeptides

Remacle, *et al. Proc. Natl. Acad. Sci. U. S. A.* **103**, 6793 (2006)



30 fs hole migration in phenylalanine

Belshaw L, *et al. The Journal of Physical Chemistry Letters* **3**, 3751 (2012)

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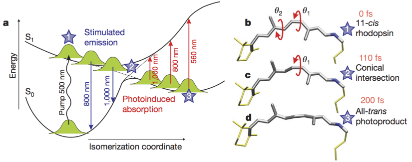
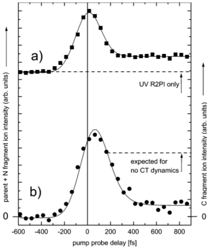
Kuleff AI *et al.* *J. Phys. B: At., Mol. Opt. Phys.* **47**, 124002 (2014)

- Charge transfer: coupling of electronic and nuclear motion



Isomerisation of rhodopsin

Polli D, *et al.* *Nature* **467**, 440 (2010)



Lehr L, *et al.* *J. Phys. Chem. A* **109**, 8074 (2005)

Charge transfer in 2-phenylethyl-N,N-dimethylamine: 80 fs

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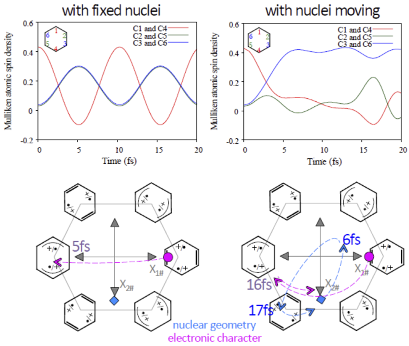
Kuleff AI *et al. J. Phys. B: At., Mol. Opt. Phys.* **47**, 124002 (2014)

- Charge transfer: coupling of electronic and nuclear motion
  - $< 10\,\text{fs}$ when coupled to nuclear motion



Coupled electronic and nuclear motion in benzene cation

Mendive-Tapia D, *et al. J. Chem. Phys.* **139**, 044110 (2013)

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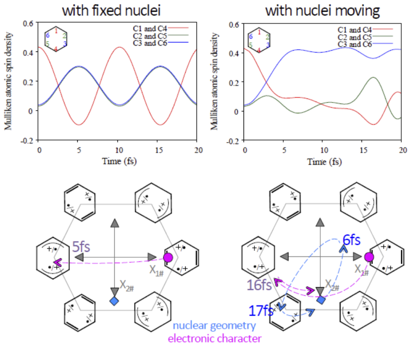

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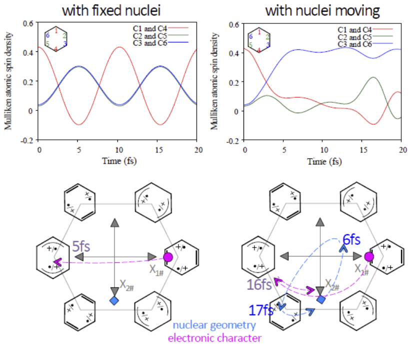
  - ≈ 1 fs timescale

Kuleff AI *et al. J. Phys. B: At., Mol. Opt. Phys.* **47**, 124002 (2014)

- Charge transfer: coupling of electronic and nuclear motion
  - < 10 fs when coupled to nuclear motion



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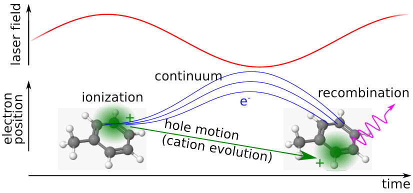
We need pump-probe techniques, capable of resolving hole dynamics with sub-femtosecond temporal resolution.

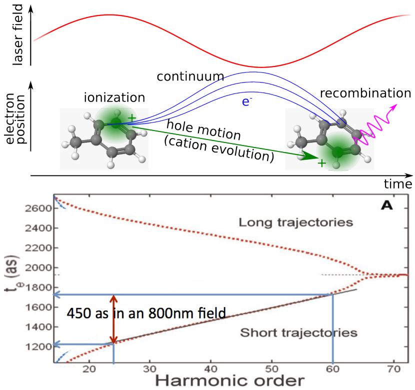
HHG as a pump-probe process

- Pump: strong field ionization
- Probe: recombination and emission of photon



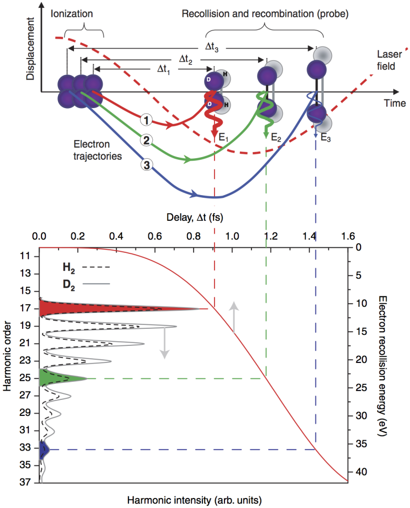
HHG as a pump-probe process

- Pump: strong field ionization
- Probe: recombination and emission of photon
- Delay mapped to harmonic frequency
- Short trajectories: max delay 3/4 optical laser
  - Long trajectories observed qualitatively: Zaïr A, *et al. Chemical Physics* **414**, 184 (2013)



HHG pump-probe spectroscopy

- Comparison with deuterated form
  - Nuclear wavepacket motion in H_2^+



Baker, *et al.* *Science* **312**, 424 (2006)

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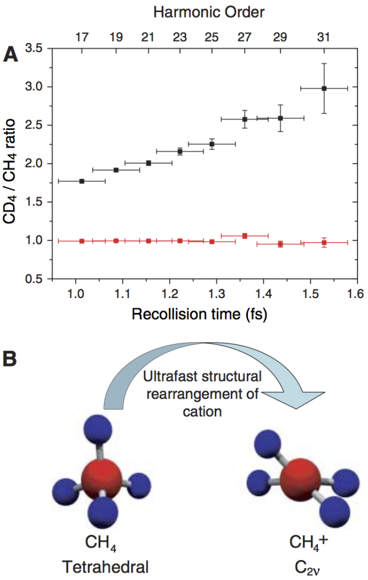

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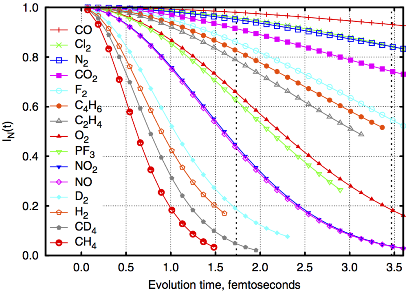

Patchkovskii S. *Phys. Rev. Lett.* **102**, 253602 (2009)

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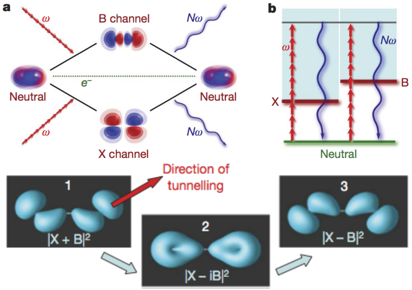
- Theory: this is a general feature of molecular HHG
- Interferometry of aligned and unaligned samples
  - Hole dynamics in CO_2



Smirnova, *et al.* *Nature* **460**, 972 (2009)

1.7 μm HHG spectroscopy in benzene & derivatives

- organic molecules
  $I_p \approx 10\,\mathrm{eV}$
- ionization saturation
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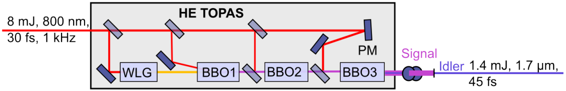

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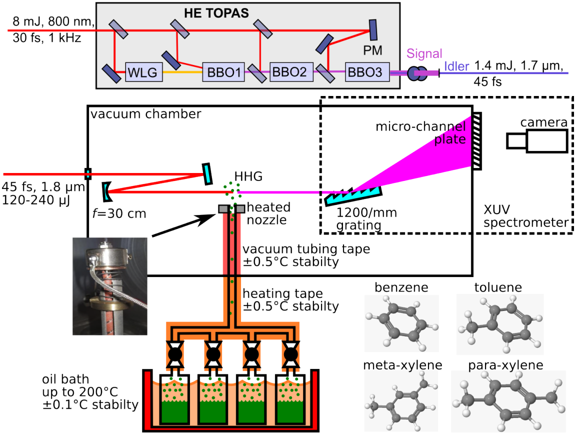
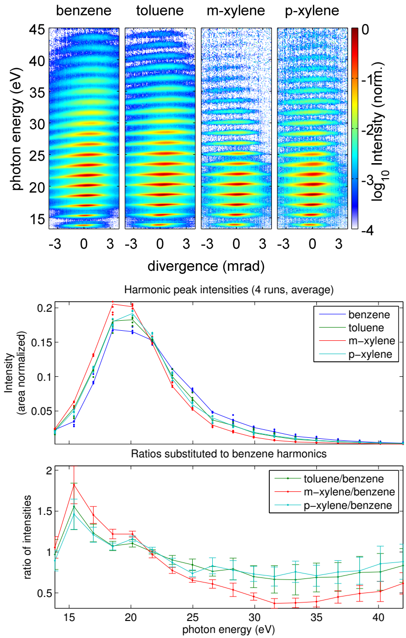
- methyl-substituted benzenes



Results

- Use benzene as reference (analogous to deuterated reference)
- Substituted benzenes harmonics decrease faster versus order
- Effect is greater in m-xylene than toluene and p-xylene



Results

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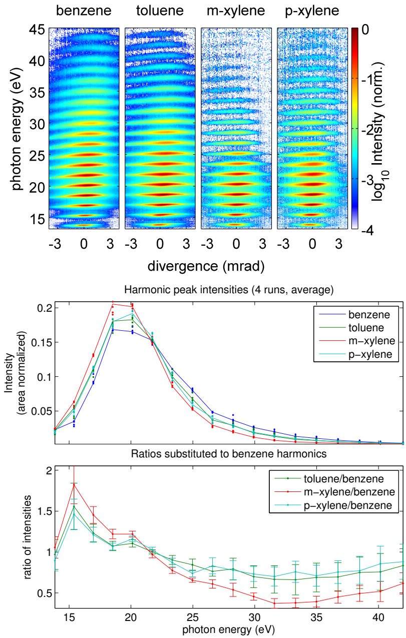
- Substituted benzenes harmonics decrease faster versus order
- Effect is greater in m-xylene than toluene and p-xylene
- Rapid nuclear motion of methyl groups one possible explanation
- Contributions from multiple electronic states are also being considered

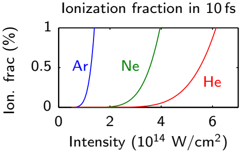


Attosecond transient absorption spectroscopy

- More general than HHG spectroscopy?

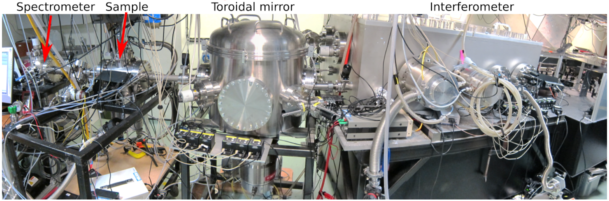
  Wang H, *et al. Phys. Rev. Lett.* **105**, 143002 (2010),Holler, *et al. Phys. Rev. Lett.* **106**, 123601 (2011),Goulielmakis E, *et al. Nature* **466**, 739 (2010)
- Element specificity of inner shell absorption edges
  - carbon K 282 eV, nitrogen K 297 eV, oxygen K 533 eV, chlorine L 204 eV, sulfur L 164 eV
- Need drive wavelength $\gg 800$ nm
  - Helium HHG cutoff: 121 eV at 800 nm vs 460 eV at 1700 nm
- Few-cycle, CEP stable driving laser pulse

$$U_p \propto I\lambda^2$$



Popmintchev, *et al. Proc. Natl. Acad. Sci.* **106**, 10516 (2009)

Yakovlev VS, *et al. Opt. Express* **15**, 15351 (2007)



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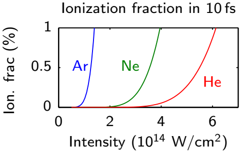

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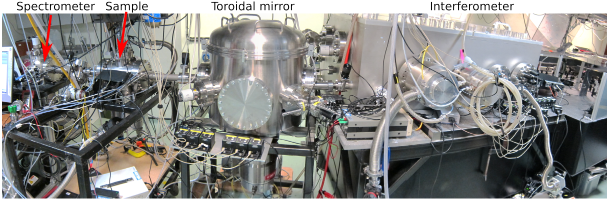
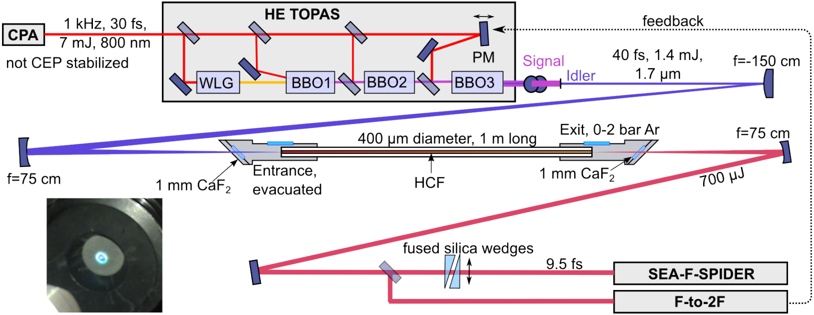


See Zsolt Diveki's talk Seminar 2.6 16.40 (Hall #3 Europe II)

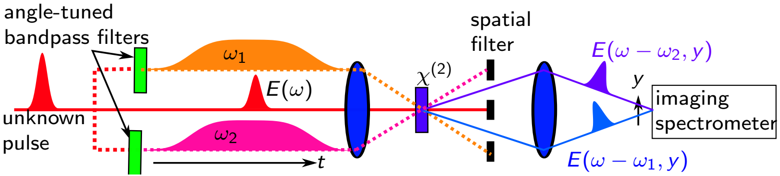
Few-cycle, CEP-stable pulses at 1.7 μm

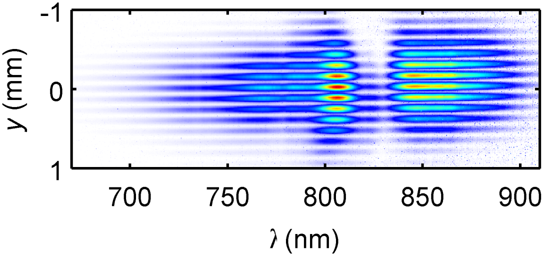


- Anomalous dispersion of SiO_2 compensates self-phase modulation
- $\phi_{\text{idler}} = \phi_{\text{pump}} - \phi_{\text{signal}}$ — "passive" CEP stability
- Transmission: 55-60%
- Power output stability: $\approx 3\%$

Schmidt BE, *et al. Appl. Phys. Lett.* **96**, 121109 (2010),Li C, *et al. Opt. Express* **19**, 6783 (2011)

Characterization with SEA-F-SPIDER





Spatio-temporal across 1D slice (spectrometer slit), single-shot

Witting T, *et al. Opt. Lett.* **34**, 881 (2009),Witting T, *et al. Opt. Express* **20**, 27974 (2012)

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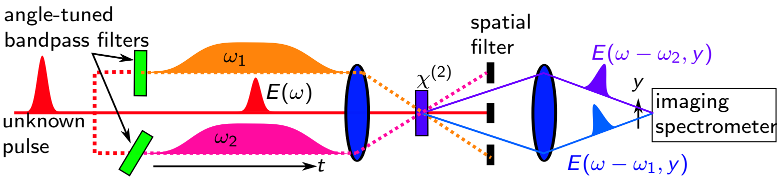

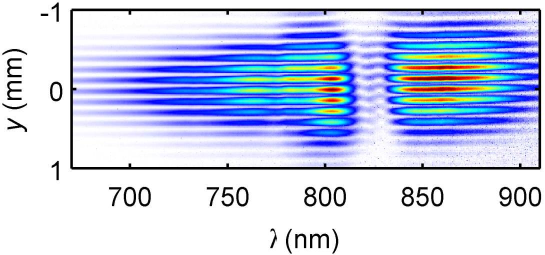

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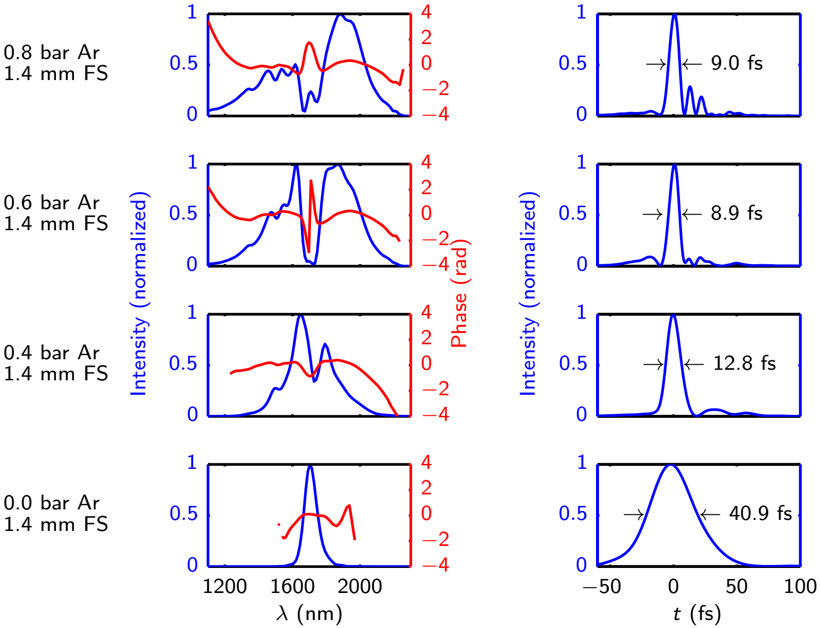
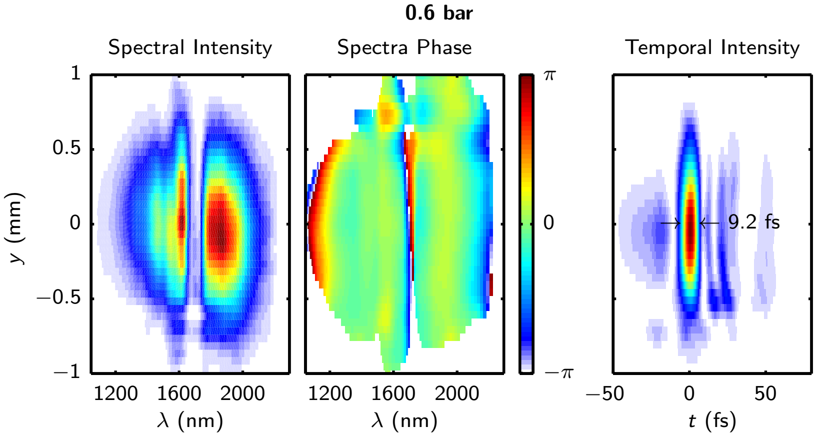
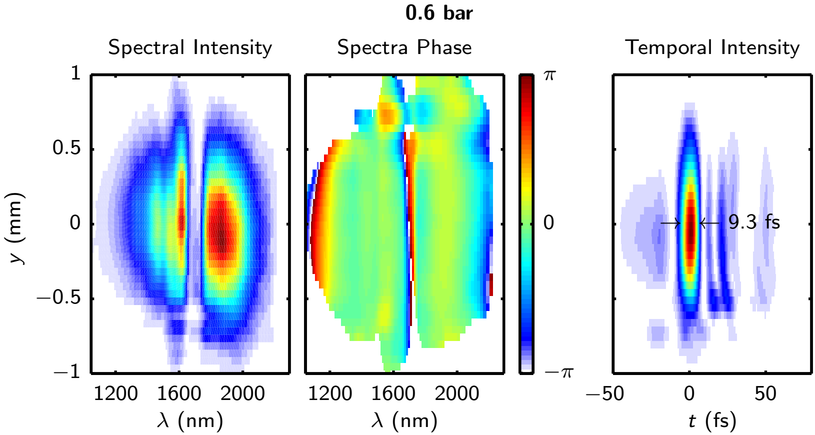
Temporal profiles



Spatially resolved temporal profiles

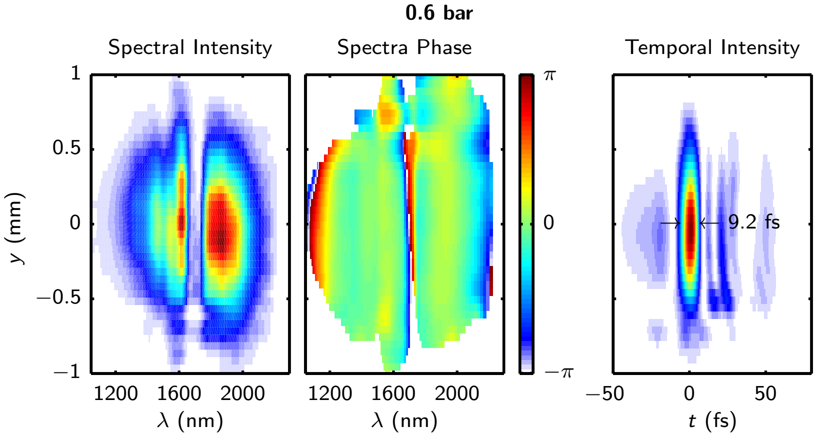


Stability



0.6 bar
Spectral Intensity
Spectra Phase
Temporal Intensity
1
0.5
0
−0.5
−1
y (mm)
1200
1600
2000
λ (nm)
π
0
−π
9.3 fs
−50
0
50
t (fs)

Stability



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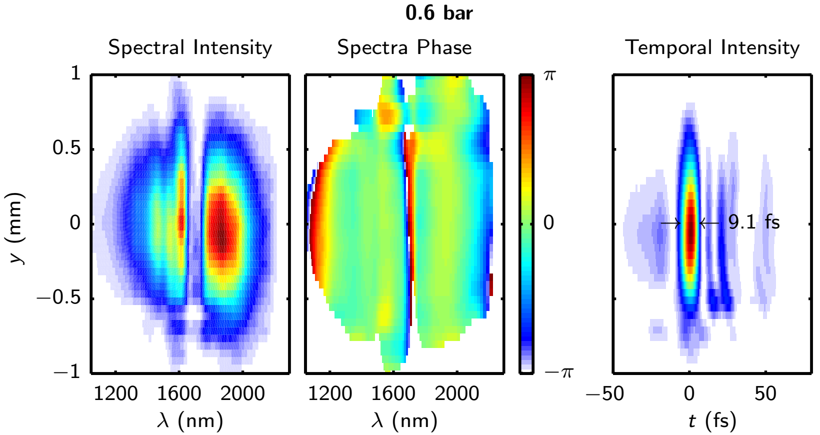
1200
1600
2000
λ (nm)
π
0
−π
9.2 fs
−50
0
50
t (fs)

Stability



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1200
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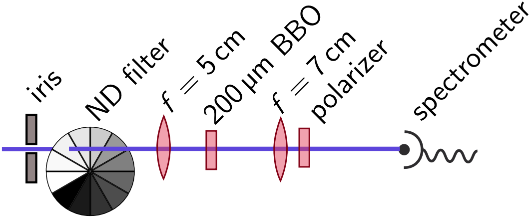
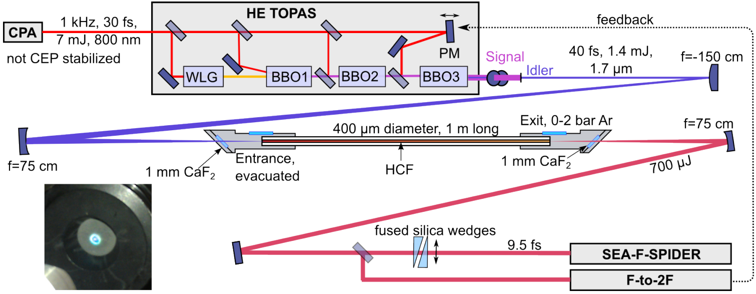
0
−π
9.1 fs
−50
0
50
t (fs)

CEP monitoring and control

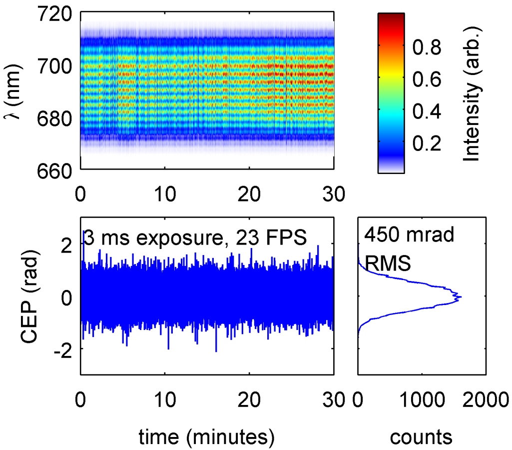
- f-to-$2f$ interferometer, piezo actuator in TOPAS third stage, “slow loop” feedback





CEP monitoring and control

- f-to-$2f$ interferometer, piezo actuator in TOPAS third stage, "slow loop" feedback
- 880 mrad single-shot RMS over 30 minutes



CEP monitoring and control

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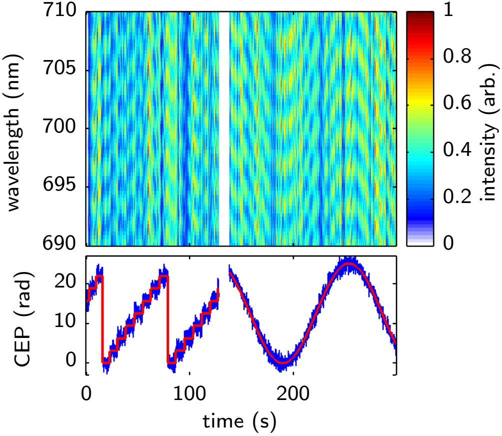
- Arbitrary sequences



Pulse propagation simulation

- Mechanism questions: Self-steepening, higher-order Kerr effect, plasma

  Béjot P, *et al. Phys. Rev. A* **81**, 063828 (2010)

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  Husakou AV *et al. Phys. Rev. Lett.* **87**, 203901 (2001), Couairon A, *et al. Eur. Phys. J. Spec. Top.* **199**, 5 (2011),Courtois C, *et al. Physics of Plasmas* **8**, 3445 (2001)

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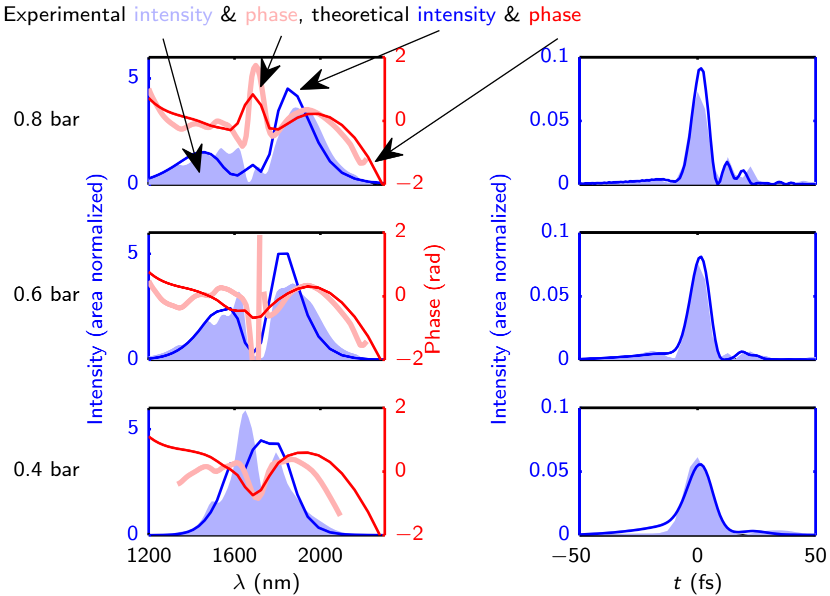
- Free parameters for agreement
  - Additional loss due to fiber imperfections
  - $n_2 \to 1.15 \times 10^{-7}\,\mathrm{cm}^2/\mathrm{TW}$, within literature range

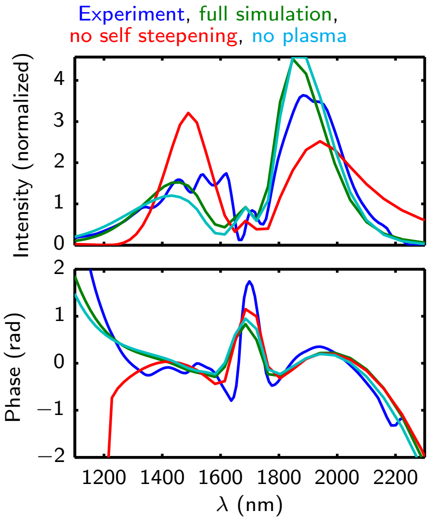
    Wahlstrand JK, *et al. Phys. Rev. A* **85**, 043820 (2012)

Comparison of model with experiment



Model implications

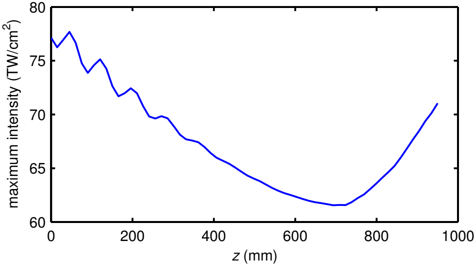
- Self-steepening crucial
- Onset of plasma effects

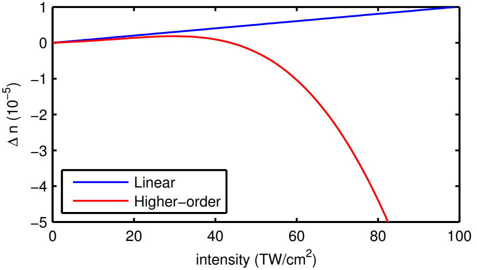


Model implications

- Self-steepening crucial
- Onset of plasma effects
- Kerr+Drude model sufficient — inconsistent with Kerr saturation at ≈ 30 TW/cm^2

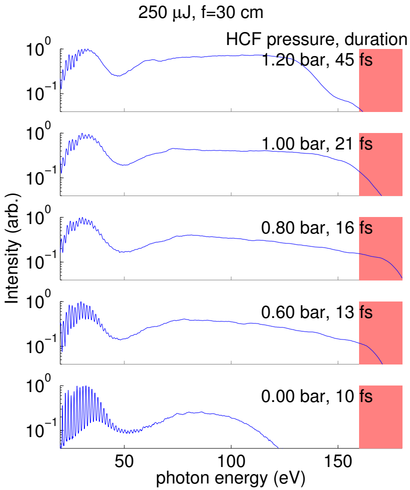
Loriot V, *et al. Opt. Express* **17**, 13429 (2009), Béjot P, *et al. Phys. Rev. Lett.* **110**, 043902 (2013), Wahlstrand JK, *et al. Phys. Rev. Lett.* **109**, 113904 (2012), Brée, *et al. Phys. Rev. Lett.* **106**, 183902 (2011)





HHG continua in argon effusive jet

- Cutoff ≈ predicted (energy, duration, spot size → 103 eV)
- Unresolved harmonics > 40 eV
- Cutoff extension
- Scatter (zeroth-order), calibration → dubious above 150 eV
- Continuum < 40 *eV*
- Cutoff law → 394 eV (10 fs)
- Likely ionization/phase-matching issues
- Spectrometer being upgraded 1200/mm to 2400/mm



Summary & outlook

- HHG spectroscopy with long wavelength fields: substituted benzene molecules
  - Tentative evidence for < 6 fs nuclear motion
  - Theoretical work ongoing
- Spectral broadening of commercial OPA pulses in argon-filled hollow fibre
  - 650 μJ, 9 fs (1.6 optical cycles), 1.7 μm pulses
  - 880 mrad CEP shot-to-shot
  - Consistent with Kerr+Drude model

We are commissioning a multi-stage differentially pumped HHG target for water window HHG.

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EPSRC Pioneering research and skills

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Role of ionization potential

- Ionization saturation doesn't fully explain results

Molecule	Ionization potential (eV)
benzene	9.22
toluene	8.83
m-xylene	8.56
p-xylene	8.44

- Ratio varies fastest in m-xylene