Centre - Énergie Matériaux Télécommunications

INRS Université d'avant-garde

Outlines

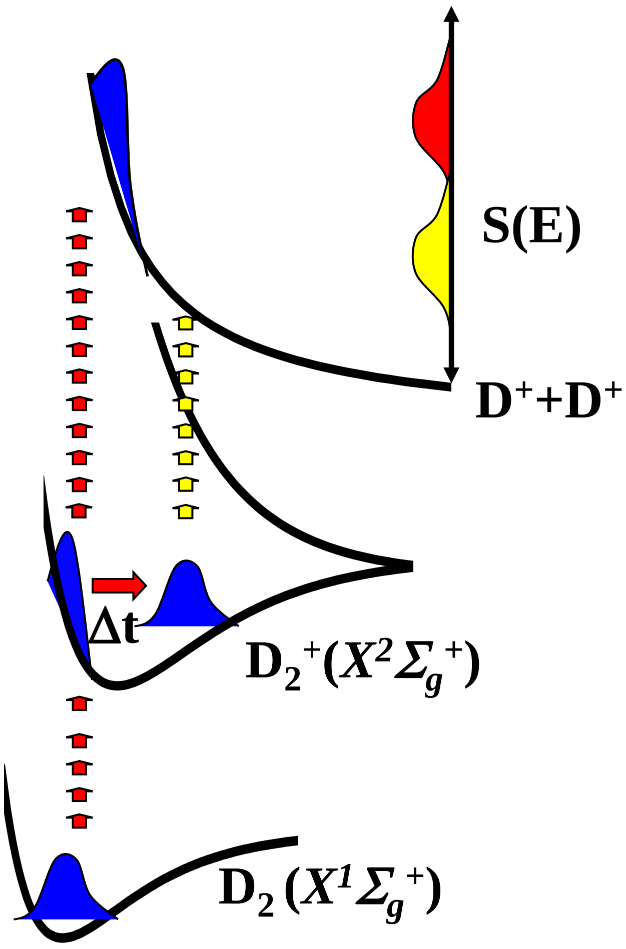
- Imaging proton migration with strong field ionization.
- Recent laser development at ALLS and new perspectives for ultrafast science – Frequency domain Optical Parametric Amplification.

François Légaré, legare@emt.inrs.ca

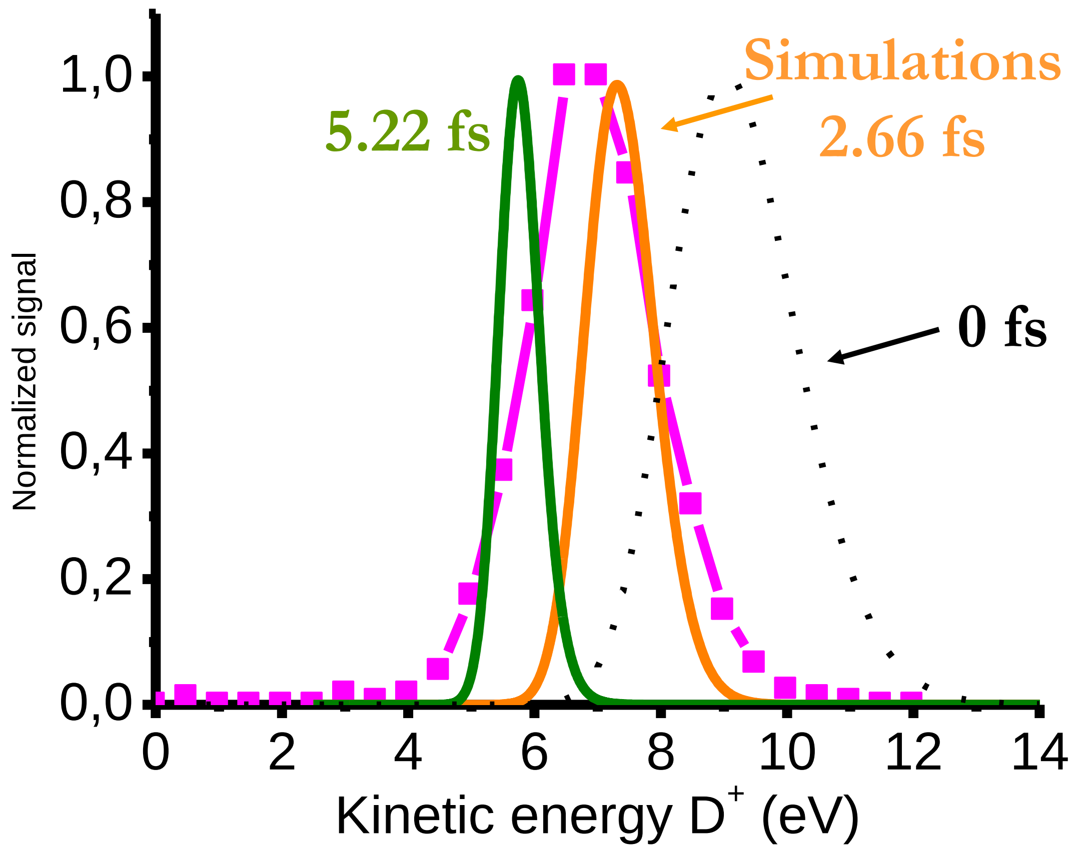
ALLS Advanced Laser Light Source

Imaging proton migration with strong field ionization

Strong field ionization induced Coulomb Explosion

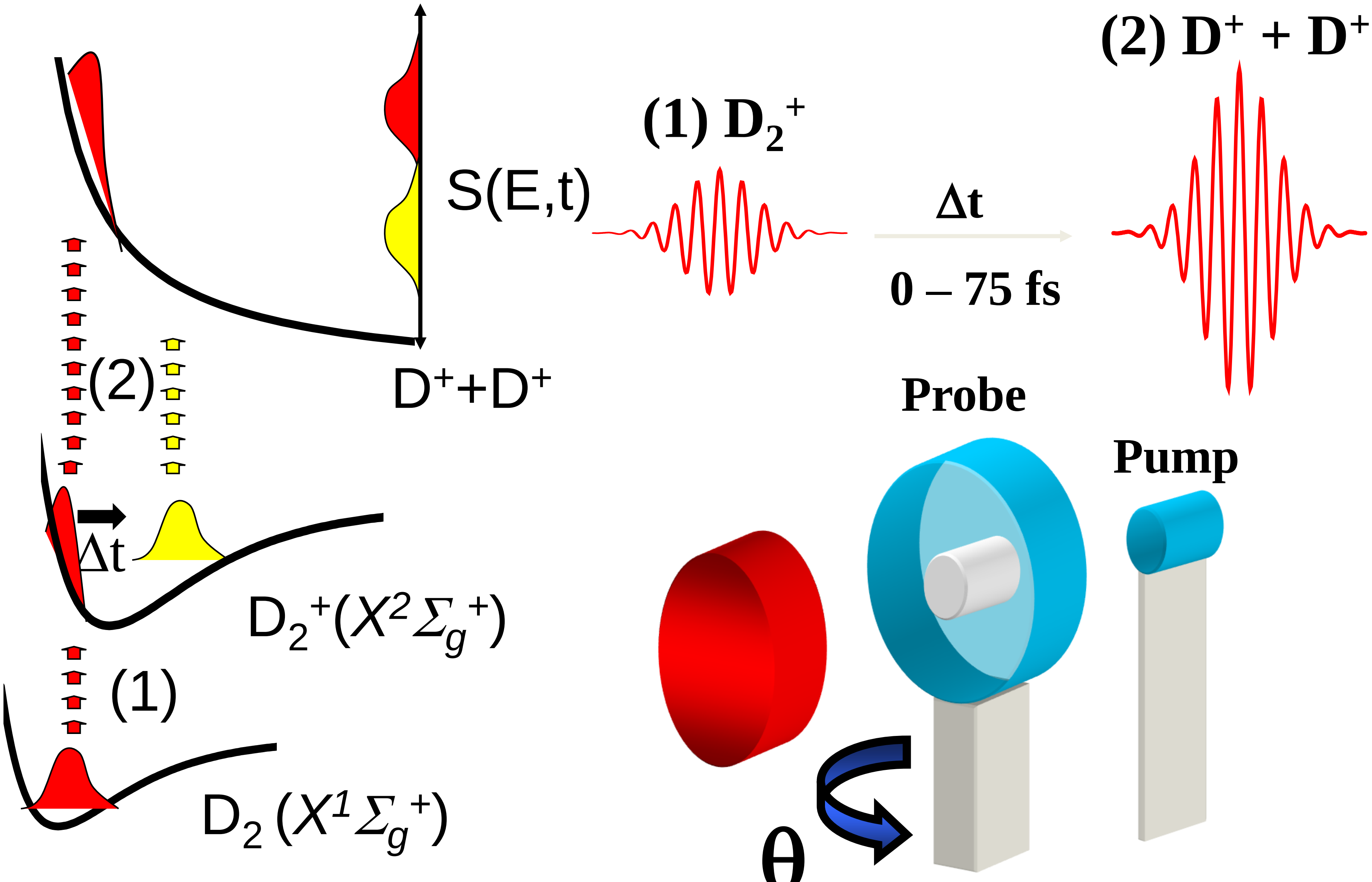


<u>**Sub-8fs: ~2x10^{15} W/cm^2**</u>

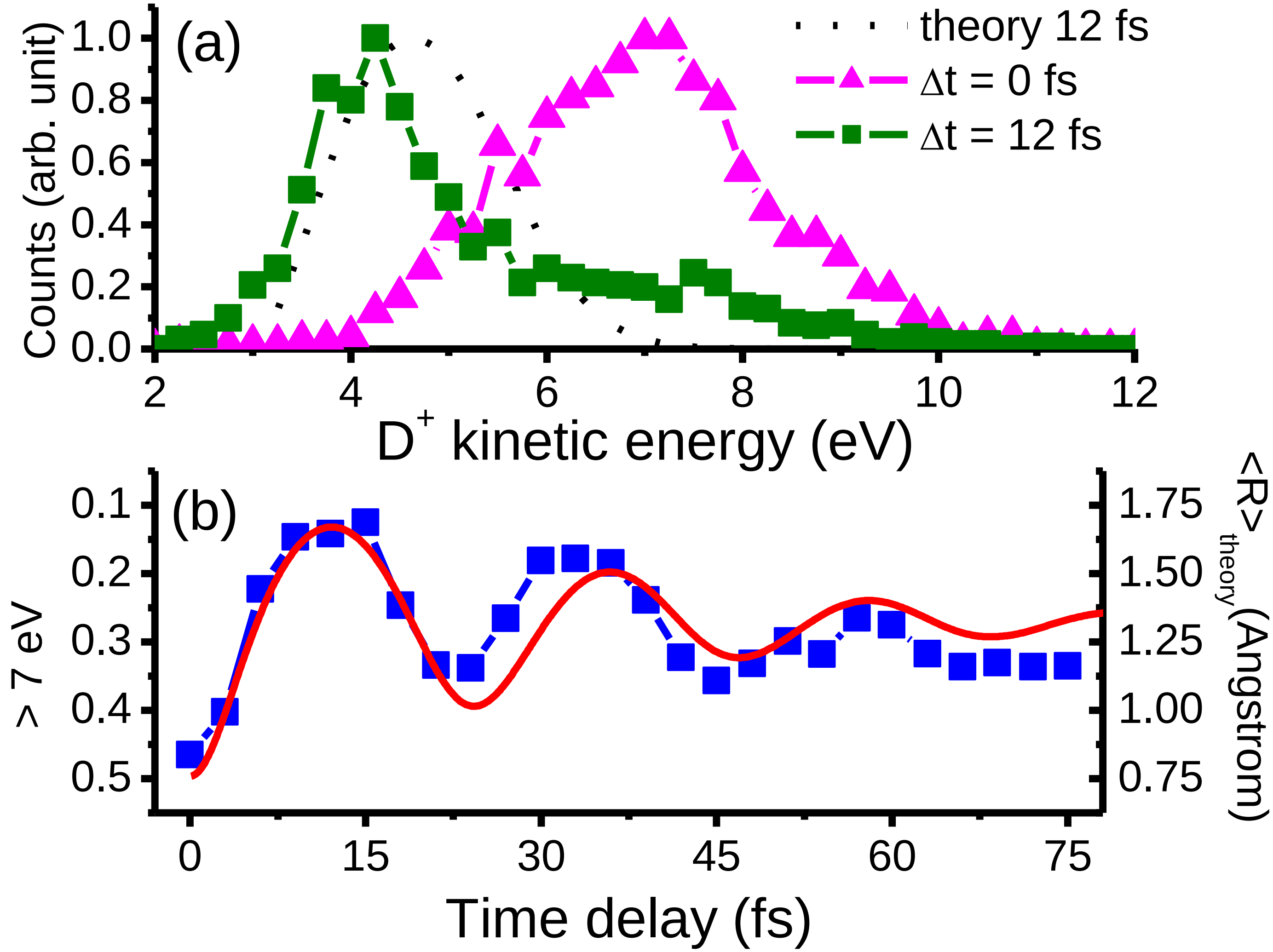


F. Légaré et *al. Phys. Rev. Lett.* **91**, 093002 (2003).

Coulomb Explosion Imaging of simple dynamics



D_2^+ dynamics

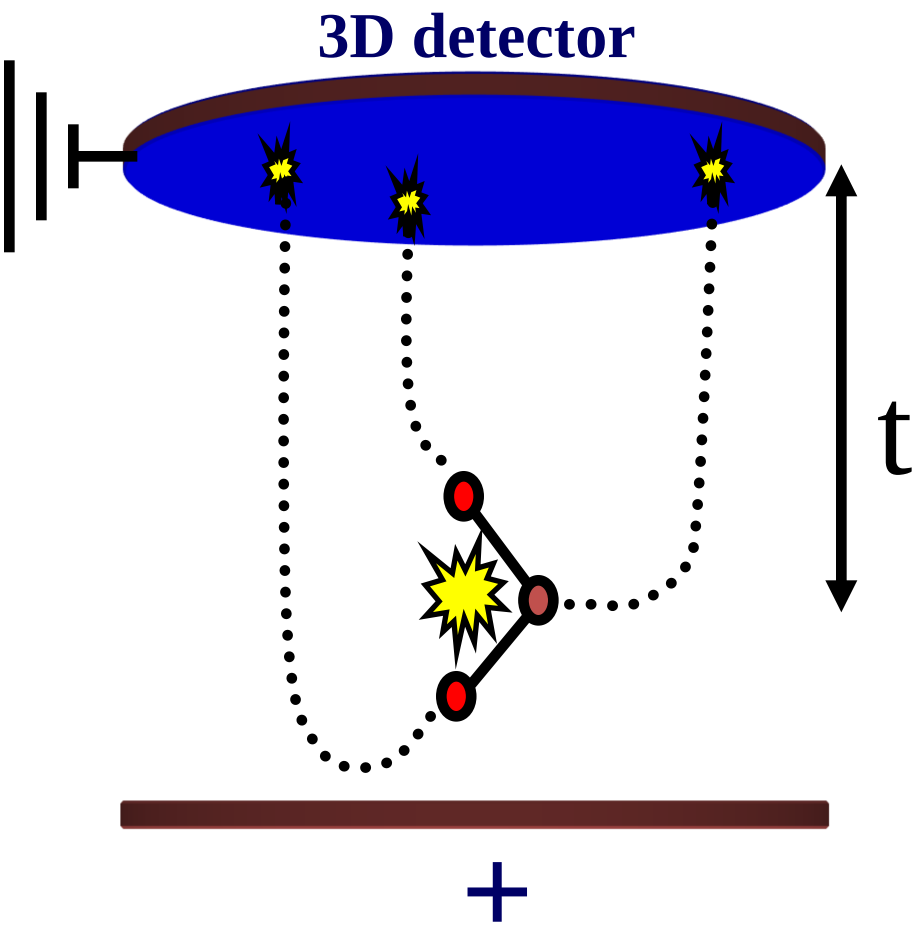


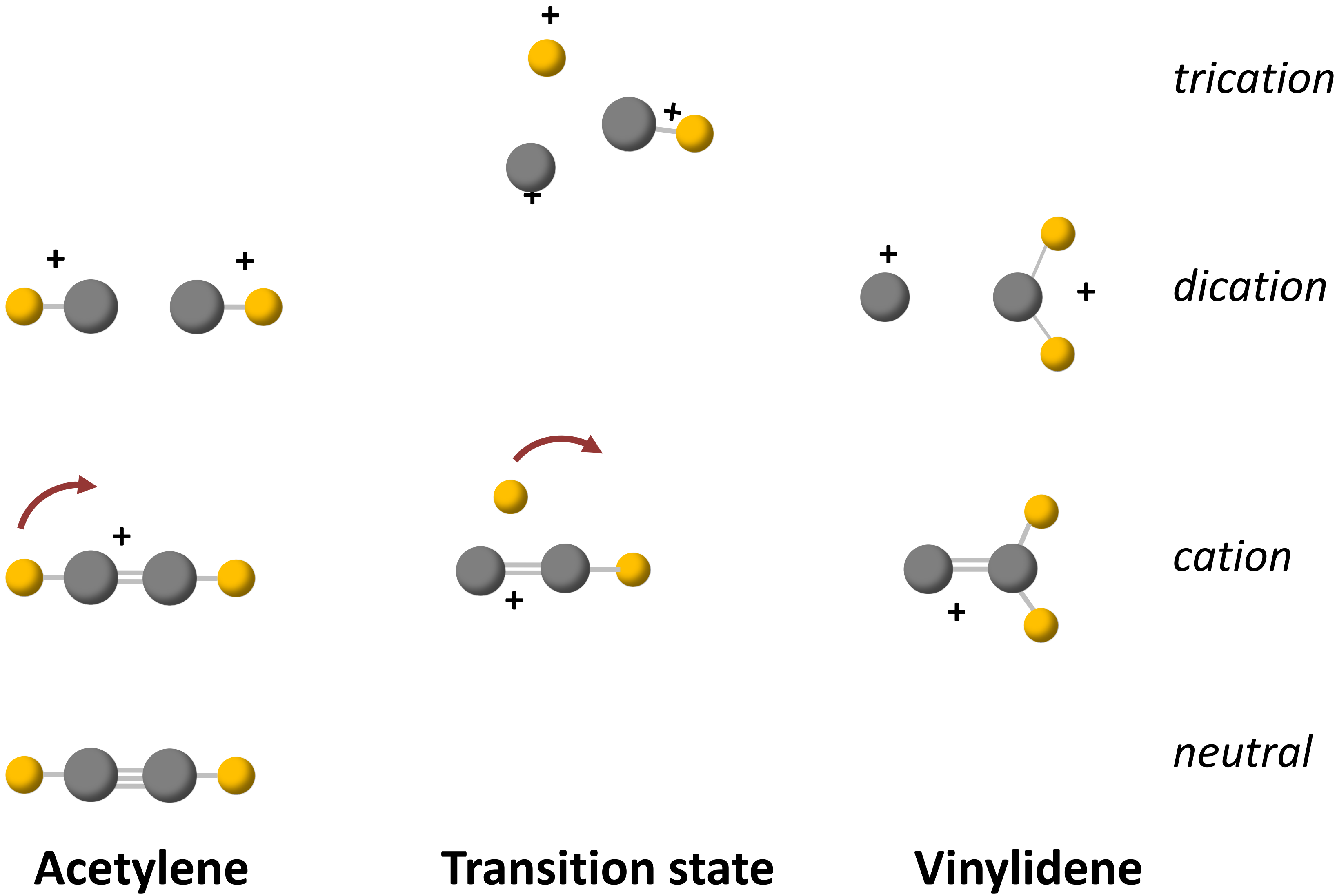
- Sub-5 fs temporal resolution.
- By using coincidence imaging spectroscopy, one can extend this technique to polyatomics.

F. Légaré et *al. Phys. Rev. A* **72**, 052717 (2005).

Coincidence imaging spectroscopy

- Experimental: P_i^{exp}
- $U(X_i)$ - Coulombien orab initio
- $dP_i/dt = -\partial U(X_i)/\partial X_i$
- Initial conditions: $X_i(t=0) = X_i^0$
- Solving: $X_i(t)$, $P_i(t)$, $P_i^{\infty} = P_i(t \rightarrow \infty)$
- Varying X_i^0 to minimize $\sum|P_i^{\infty} - P_i^{exp}|$
- **Optimal structure**

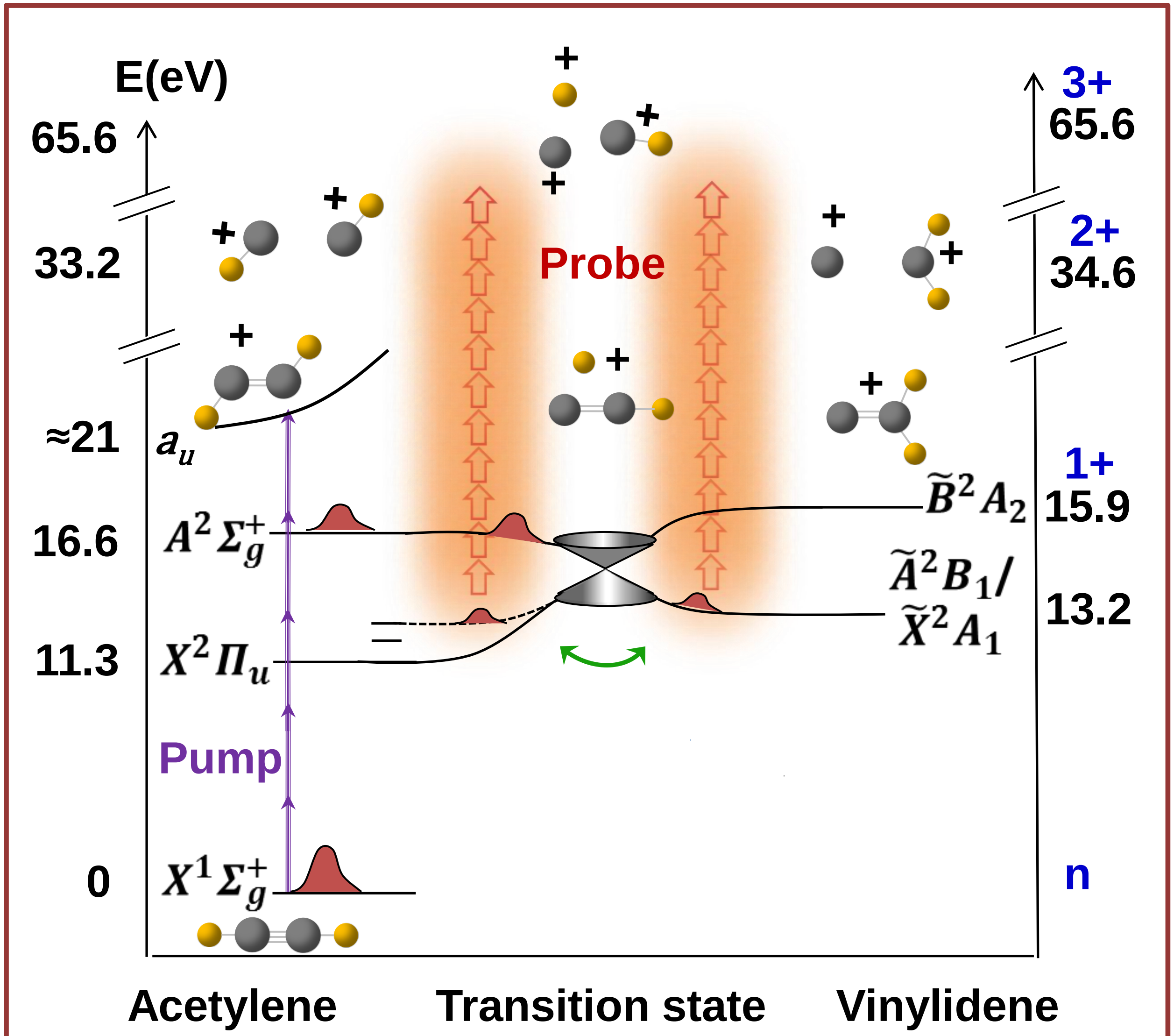




Hishikawa, et al., PRL **99,** 258302 (2007)

Jiang, et al., PRL **105,** 263002 (2010)

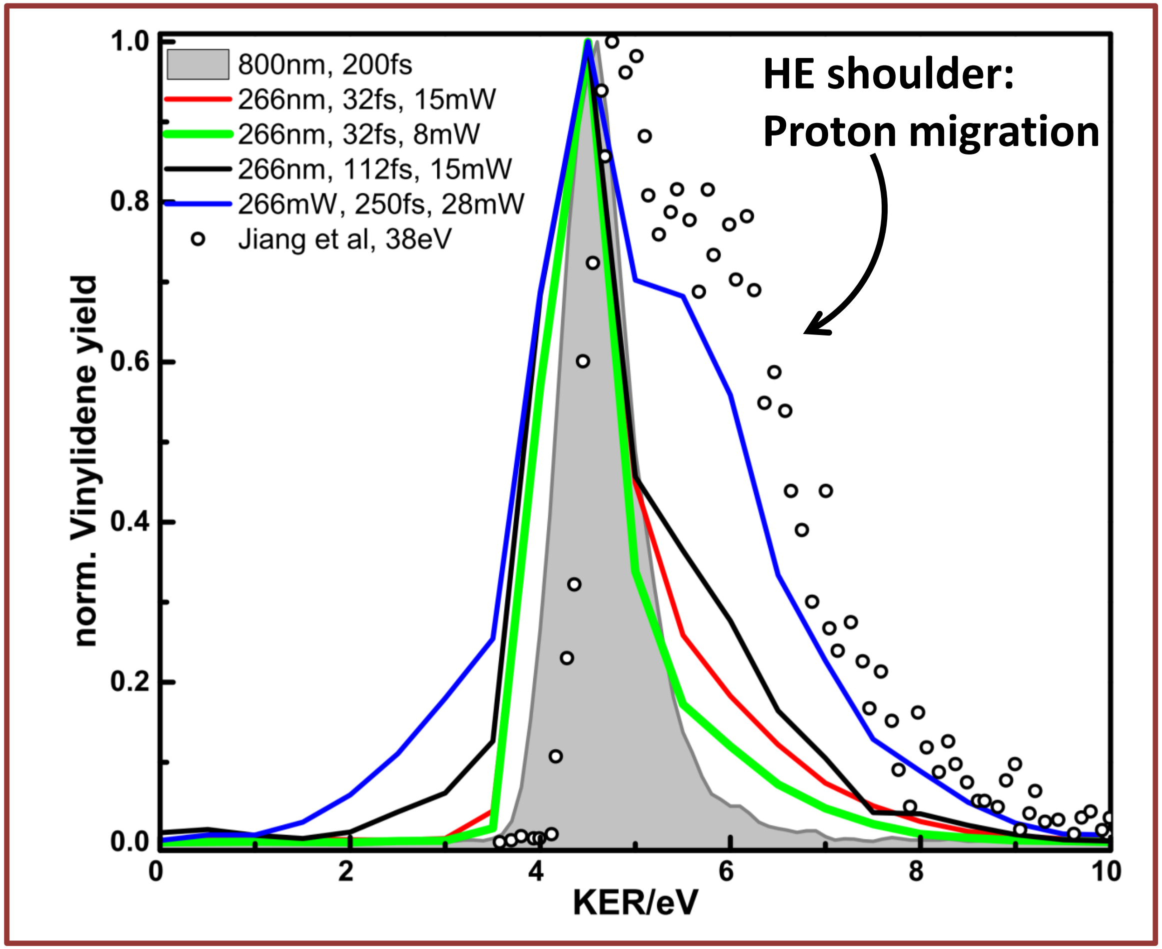
Experimental scheme



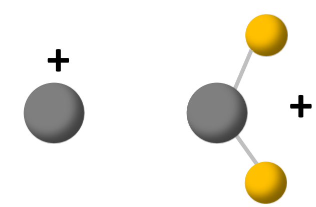
How do we distinguish channels?

Wavelength dependence

Single pulse experiment



Vinylidene channel C^+ CH_2^+ correlation

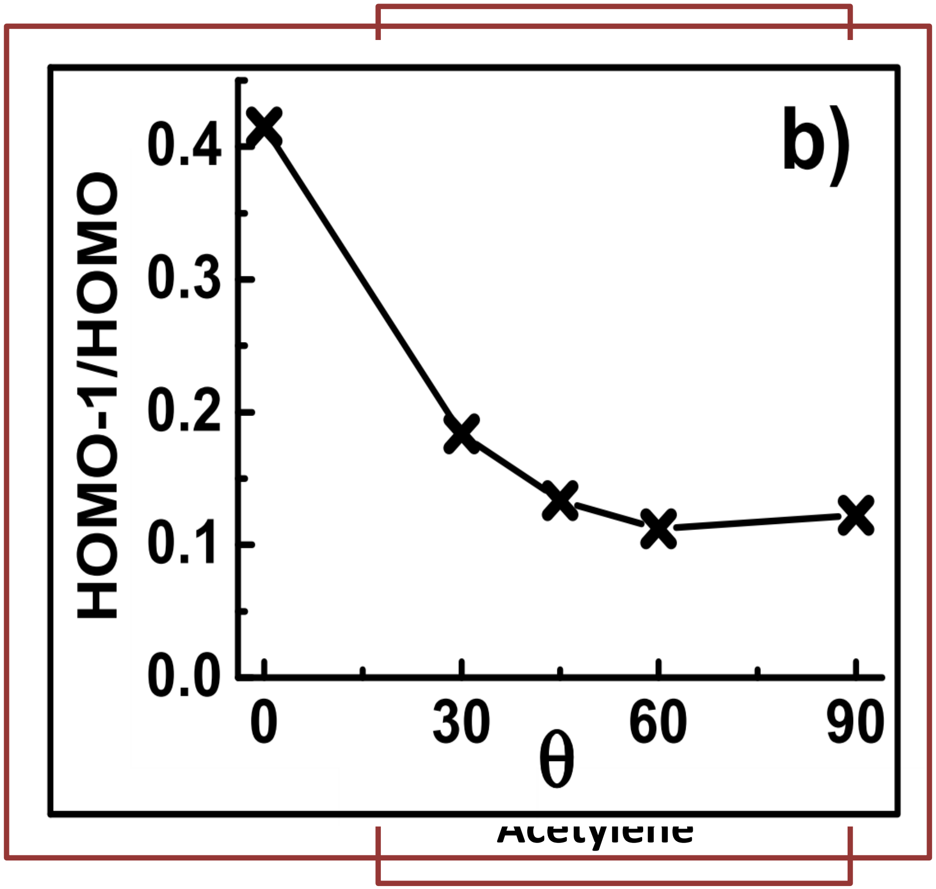


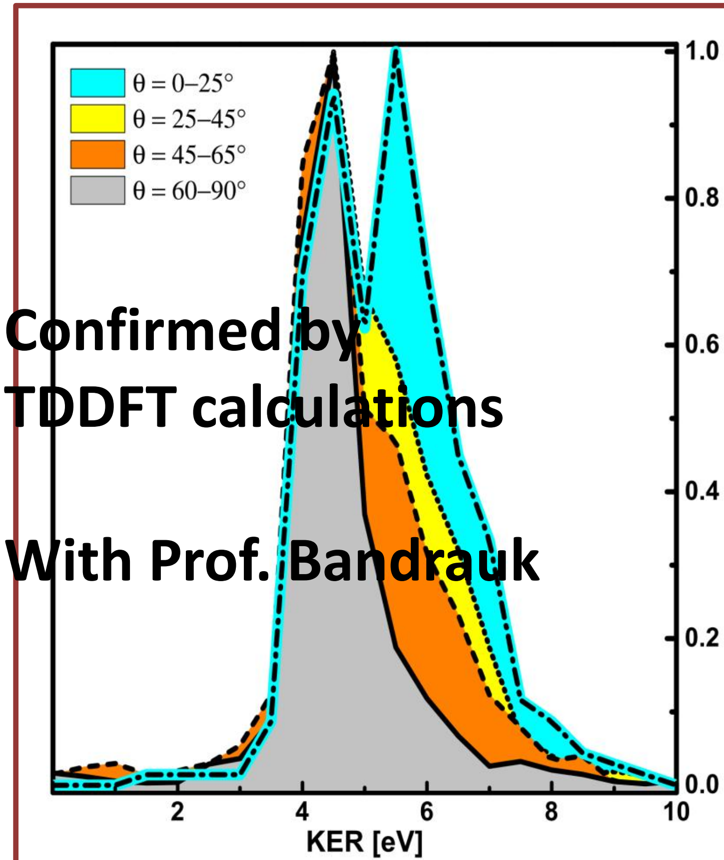
Jiang, et al., PRL **105,** 263002 (2010)

State assignment: angular dependence

Single pulse experiment

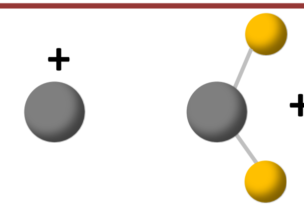
266 nm, 110 fs





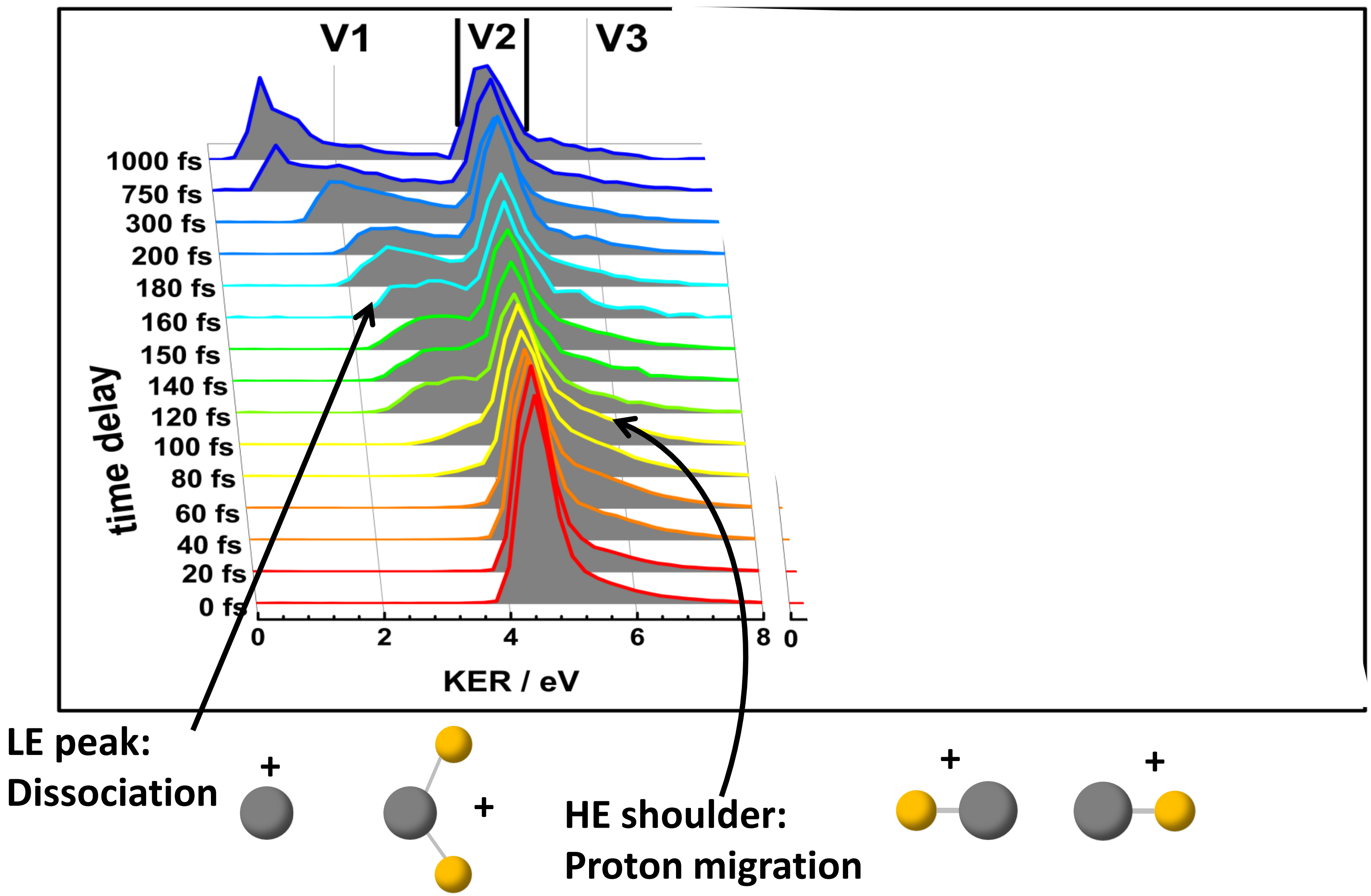
Confirmed by
TDDFT calculations

With Prof. Bandrauk



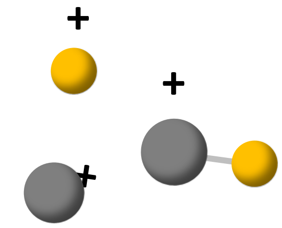
2 body breakup KER

Pump probe experiment

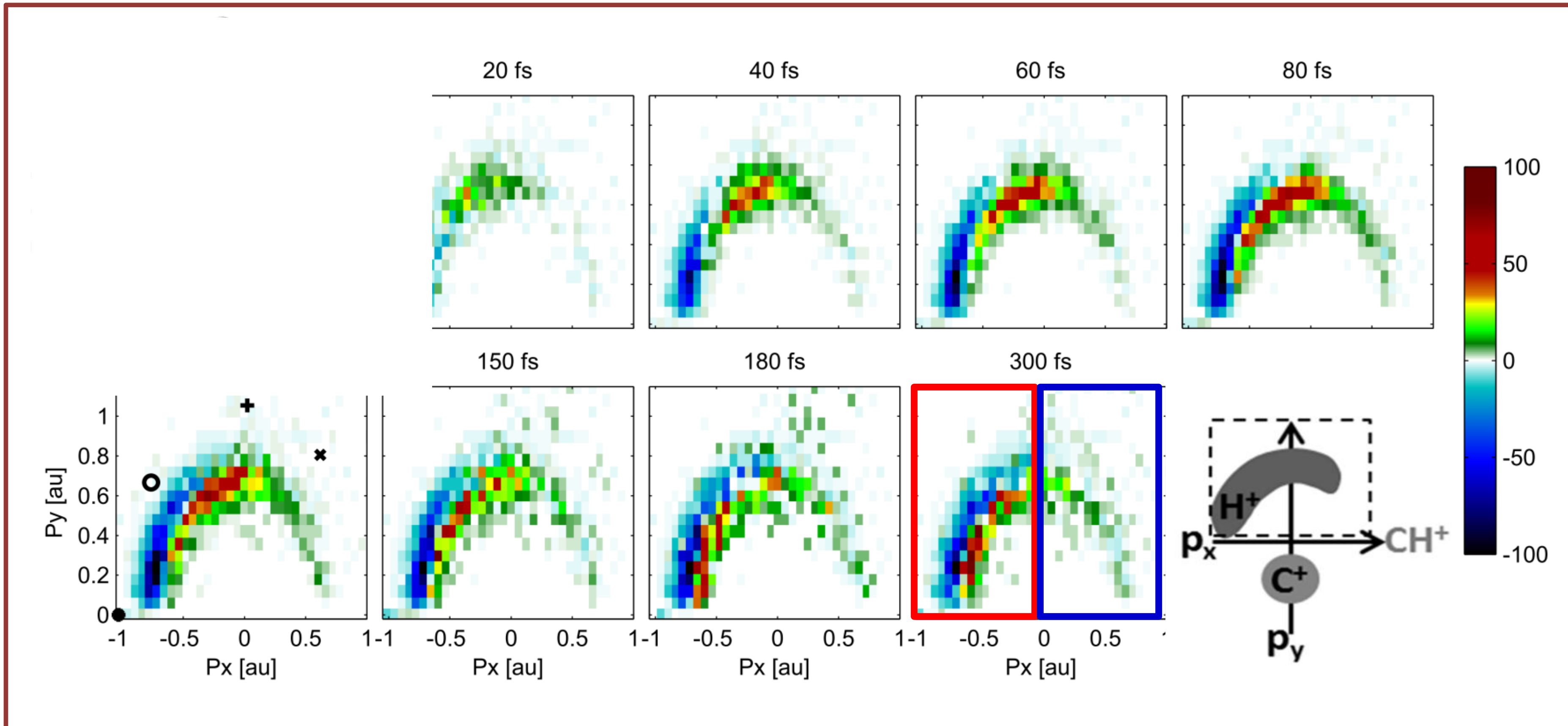


Molecular movie

Newton plot – movie frames

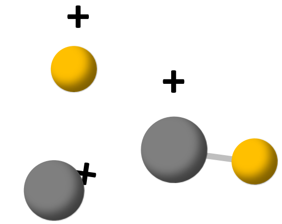


Pump probe experiment

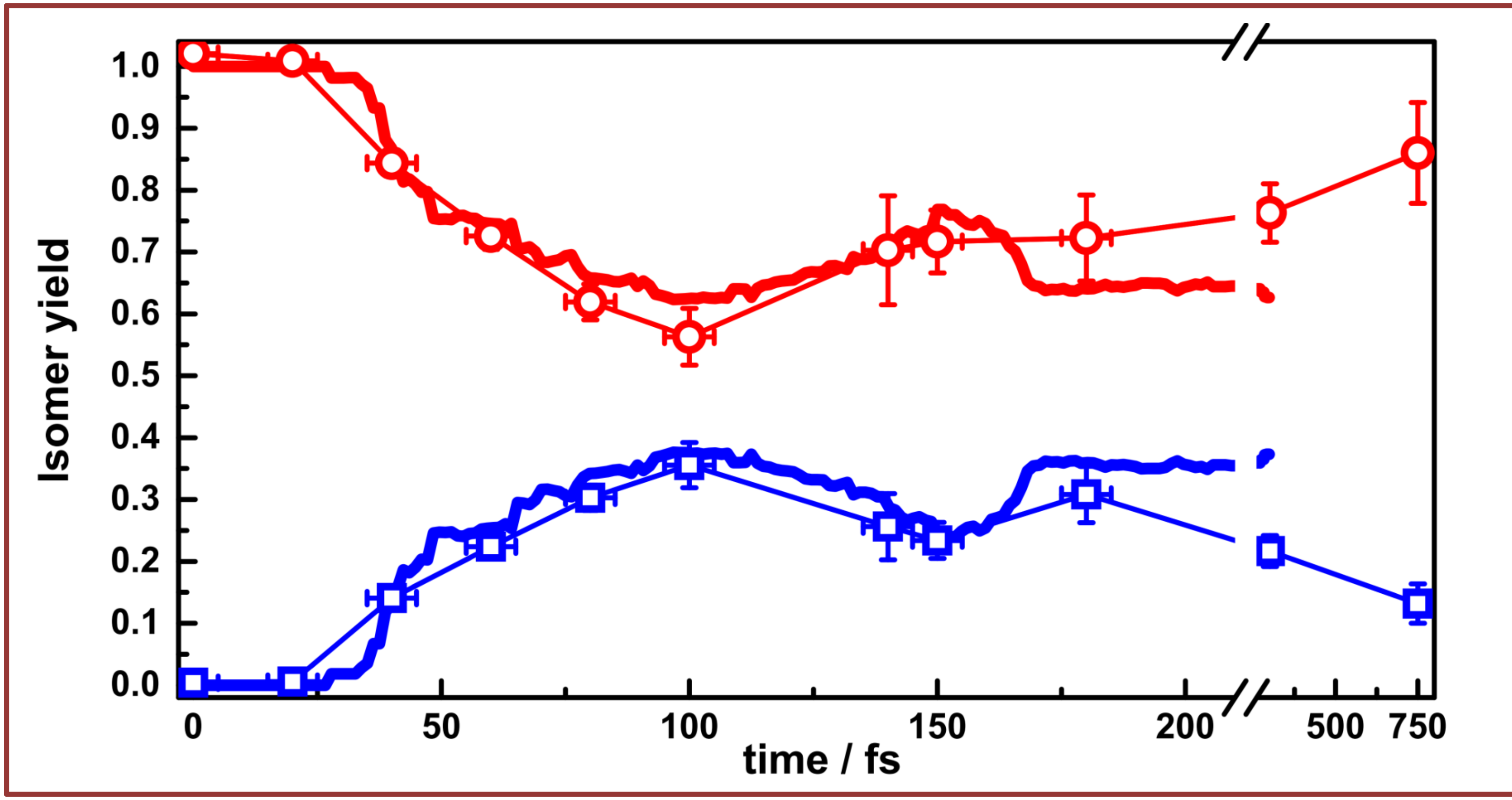


Fragment momenta in molecular frame correspond well to position of atoms: H^+ is much lighter than C^+

Comparison with theory



Pump probe experiment



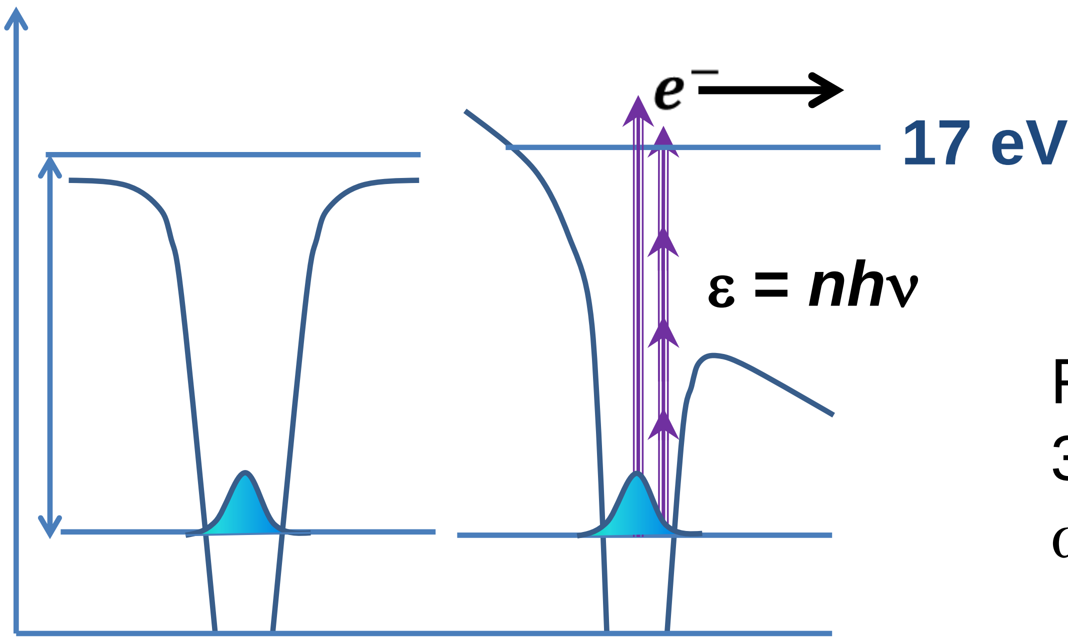
Acetylene exp. ○ theory —

Vinylidene exp. □ theory —

M. Schuurman

Ibrahim, H. et al. Nat. Commun. 5:4422 doi: 10.1038/ncomms5422 (2014)

1 photon vs. multiphoton ionization



FEL : 38 eV	→	1 p
3ω : 4.6 eV	→	4 p
ω : 1.5 eV	→	12 p

If energy gap of ionization from HOMO vs. HOMO-1 is too large to be overcome by 800 nm (and one has no VUV source at hand) multiphoton ionization is a very efficient way to launch dynamics.

Future directions

- Isotopic effect – C_2D_2
- Other systems – C_2H_4, C_3H_4

IOP Publishing — Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. 42 (2009) 081002 (5pp) — doi:10.1088/0953-4075/42/8/081002

FAST TRACK COMMUNICATION

Femtosecond isomerization dynamics in the ethylene cation measured in an EUV-pump NIR-probe configuration

J van Tilborg[1], T K Allison[1,2], T W Wright[1], M P Hertlein[1], R W Falcone[1,2], Y Liu[1], H Merdji[3] and A Belkacem[1]

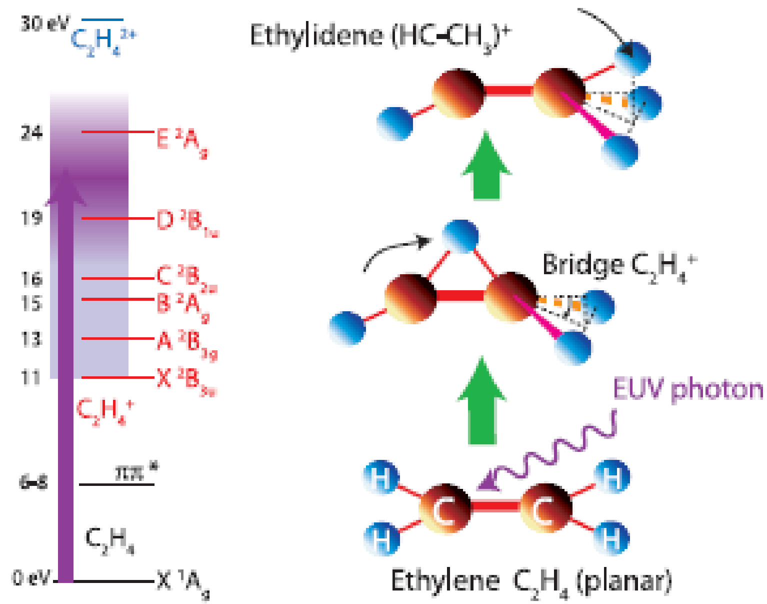
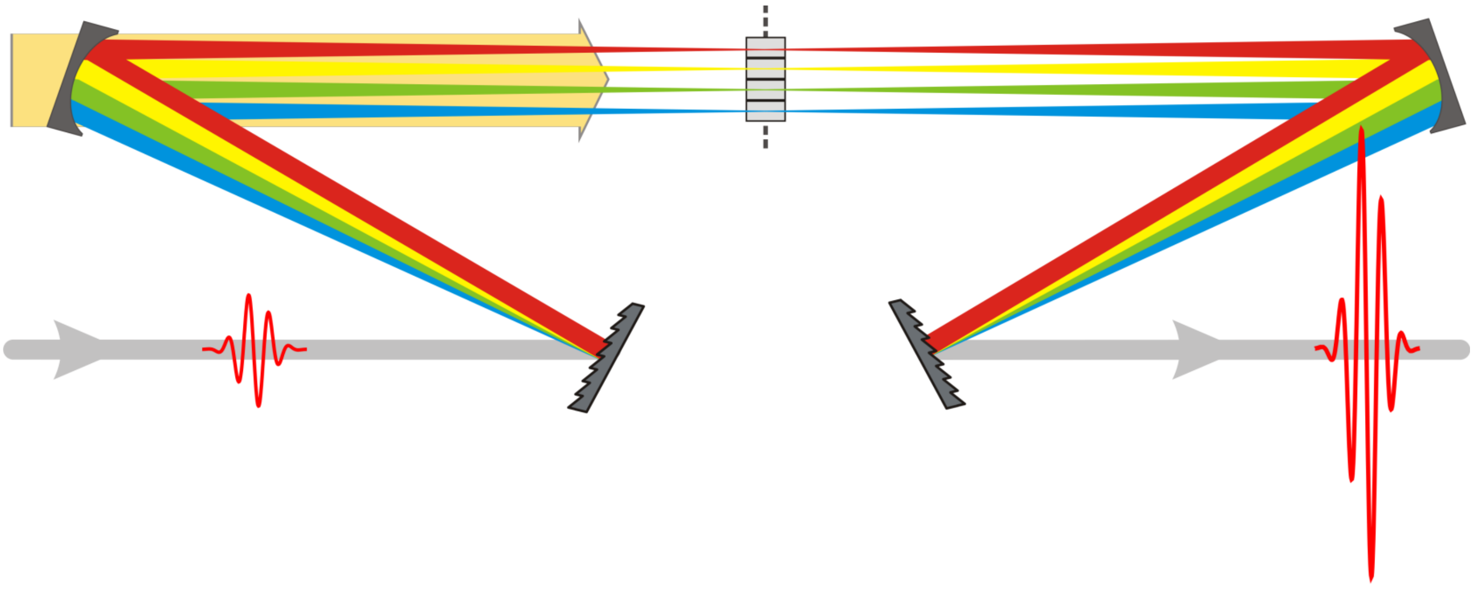
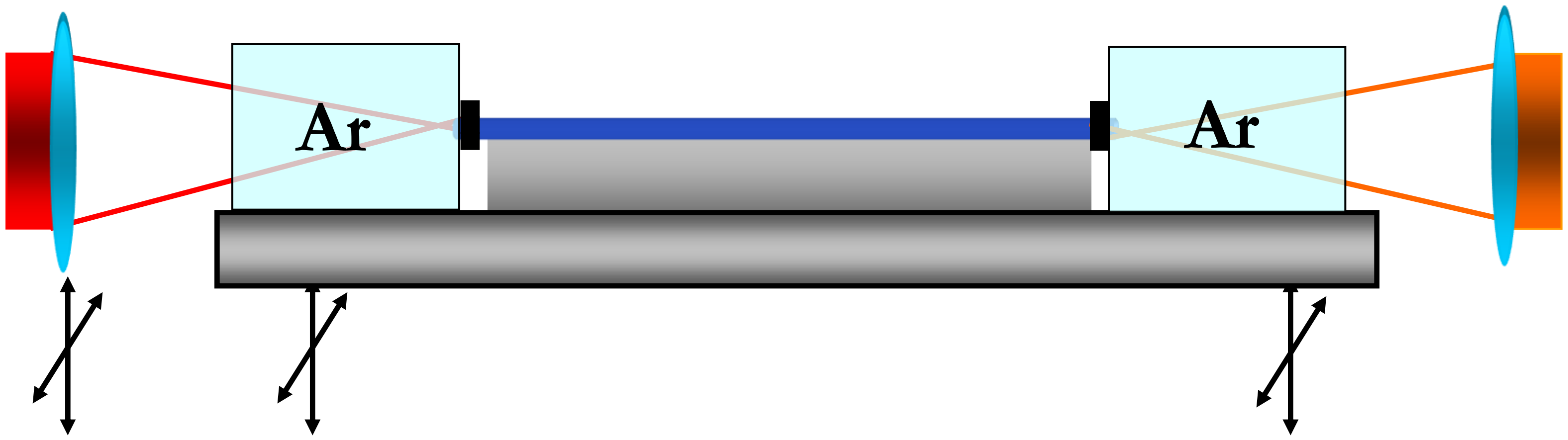


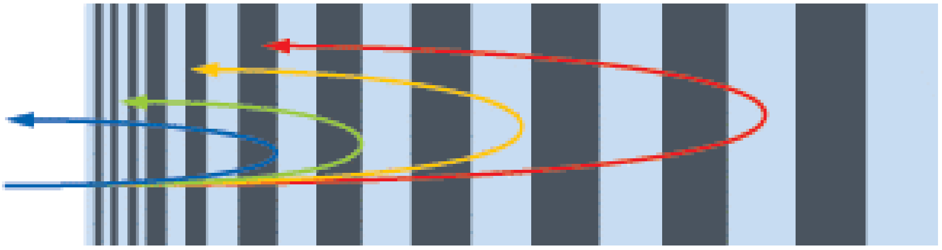
Figure 1. The left diagram shows the energy levels of the ground and excited states of neutral ethylene, the cation and the dication. On the right is a sketch of a predicted [3, 4, 17] isomerization sequence following EUV-driven population of the excited states of the cation. Note the ethylidene configuration, which is believed to be followed by a H_2 stretching configuration.

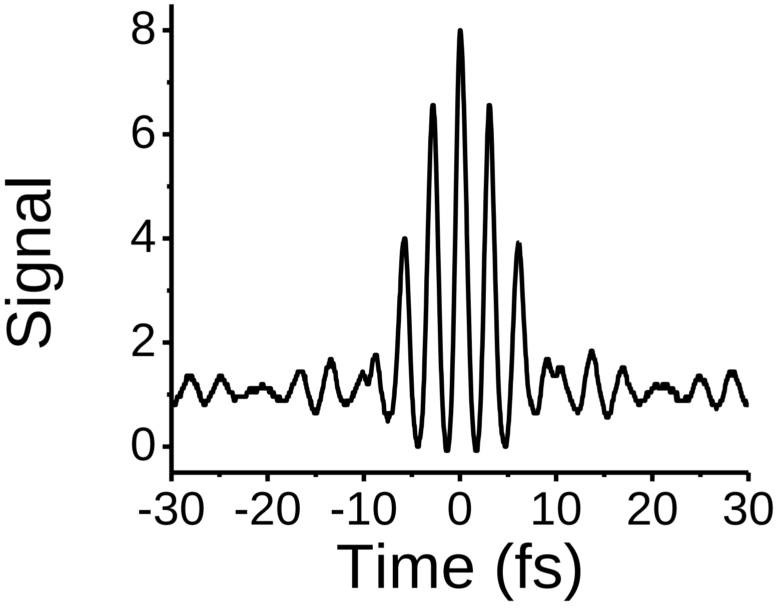
Frequency domain Optical Parametric Amplification



Few-cycle laser pulses @ 800 nm

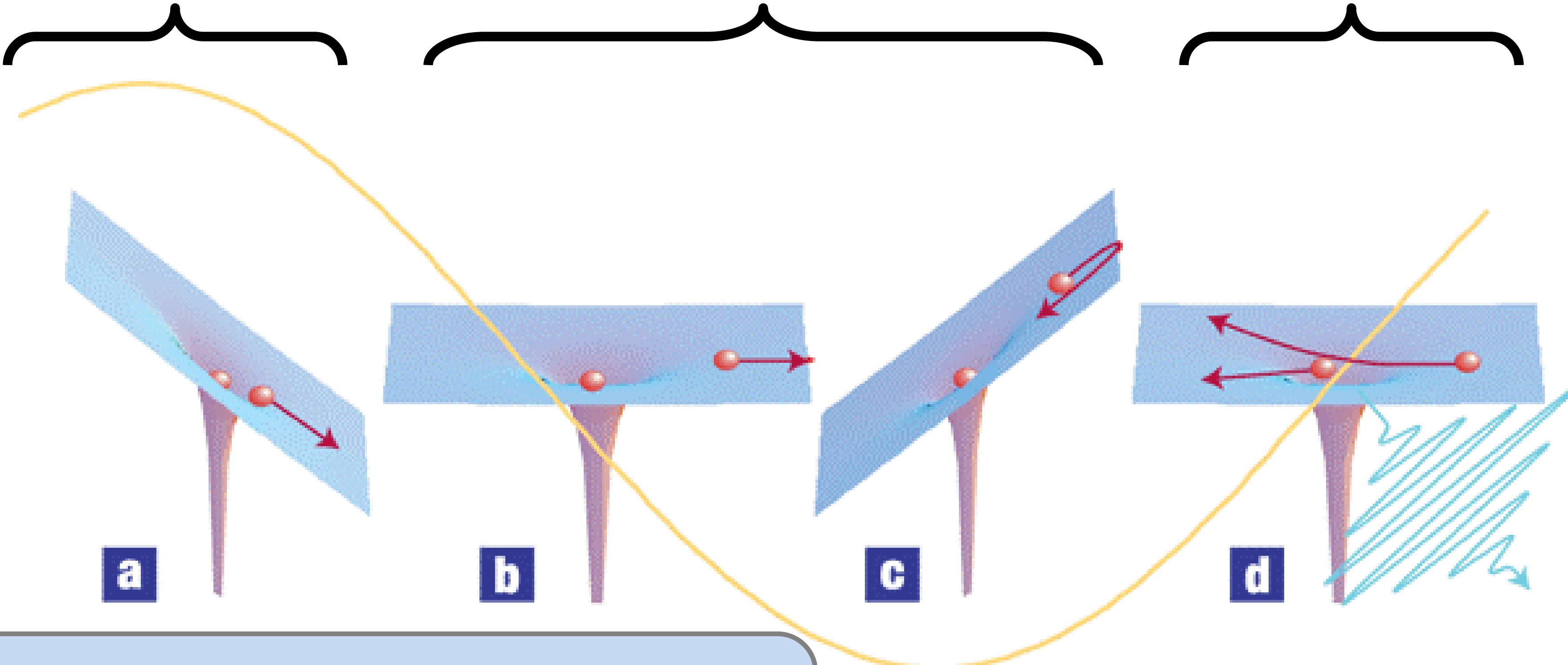






High Harmonic Generation

Tunnel Ionization | Electron acceleration | Recombination & X-ray generation

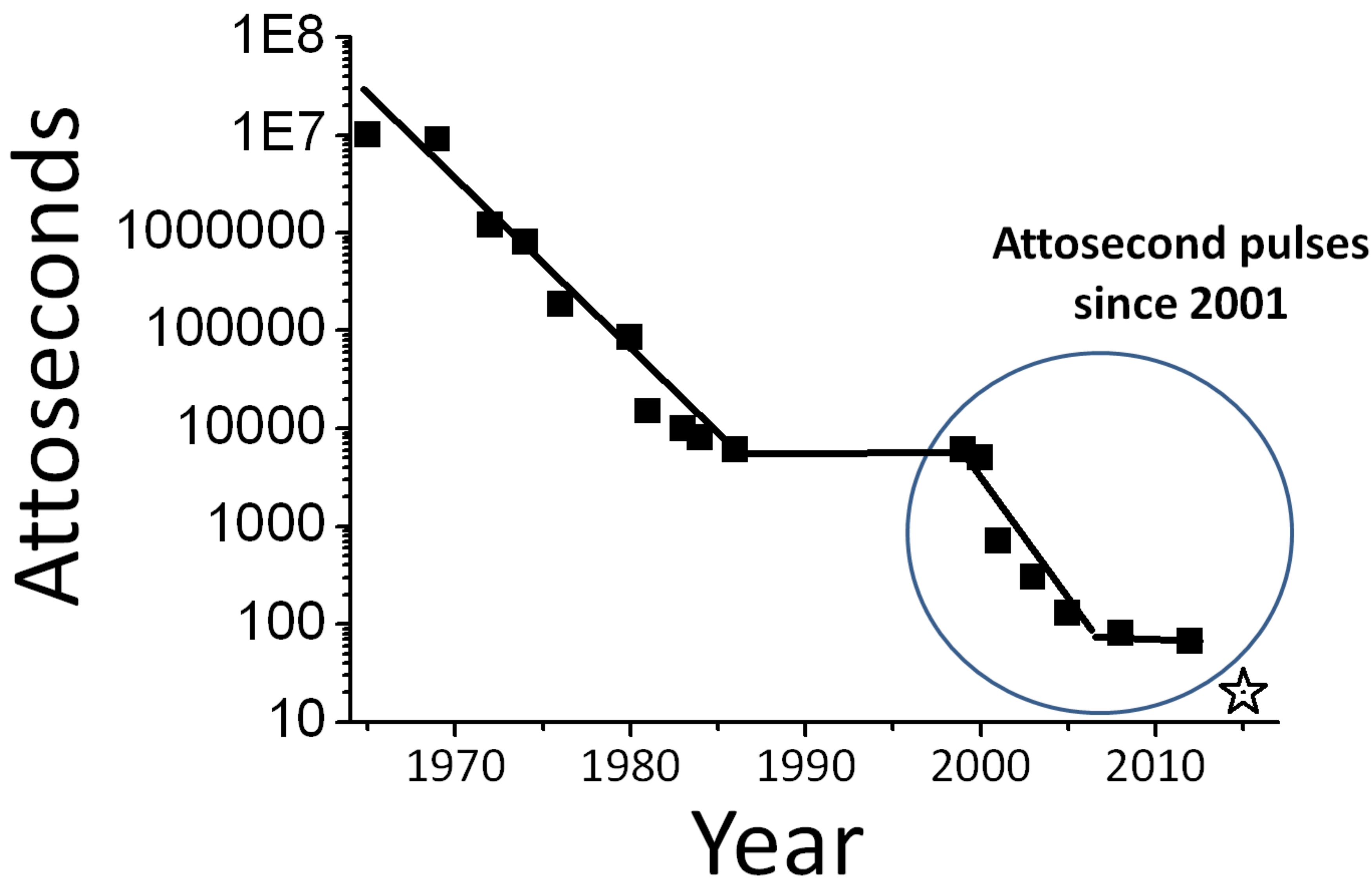


$Up = I\lambda^2$; $h\nu = Ip + 3.17Up$

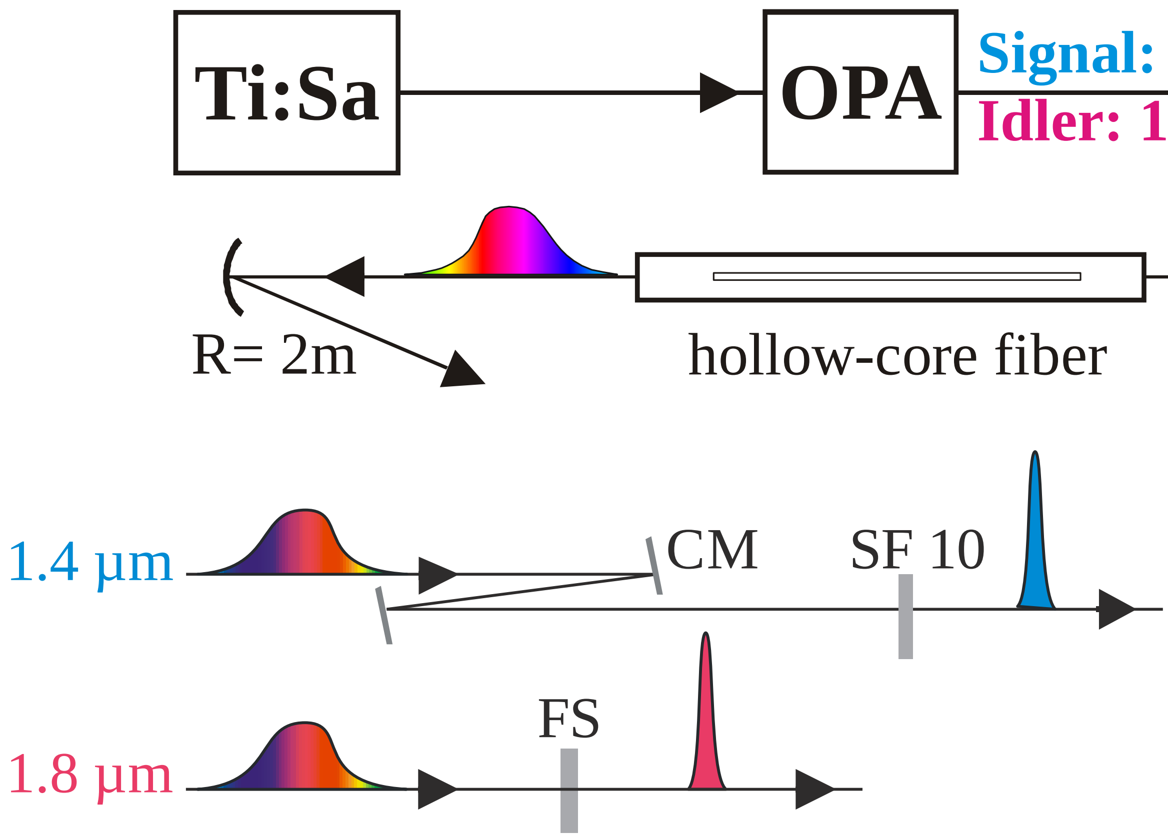
$Up = 300$ eV @1800 nm 10^{15} W/cm^2

Corkum and Krausz, Nat. Phys. 2007.

Laser pulse duration since the 60s



Experimental setup for few-cycle IR

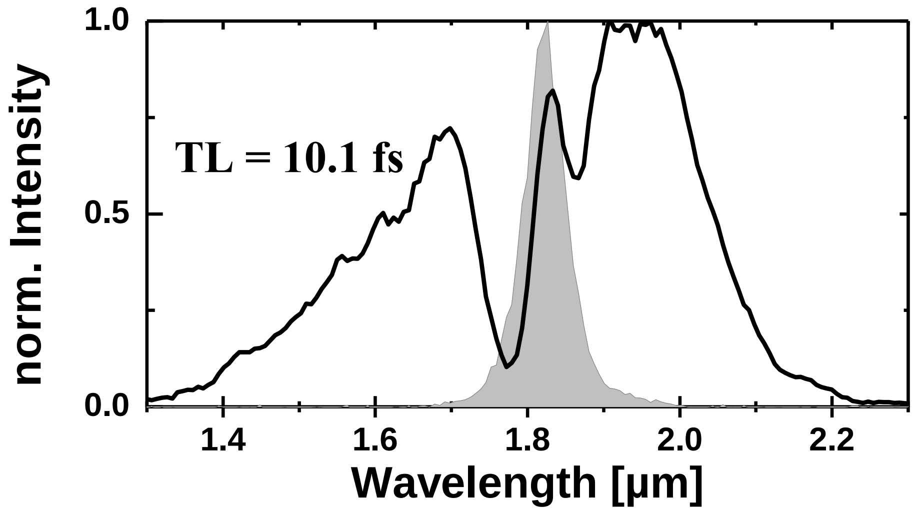


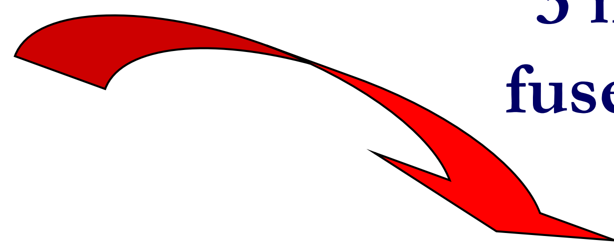
Sub-three cycles:
Opt. Lett. **34**, 1894 (2009).

Sub-two cycles:
APL **96**, 071111 (2010)

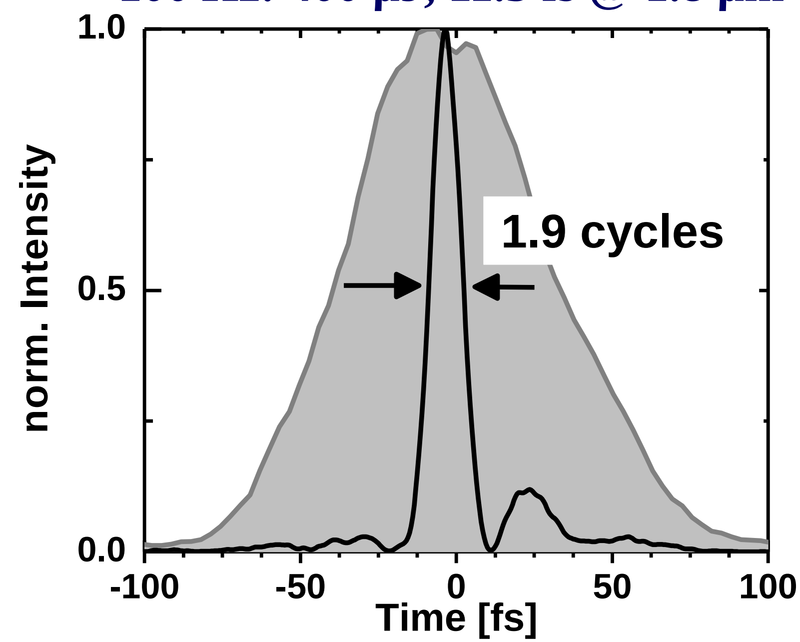
CEP stable 1.6 cycles:
Opt. Exp. **19**, 6858 (2011)

Idler pulse compression – 1.8μm



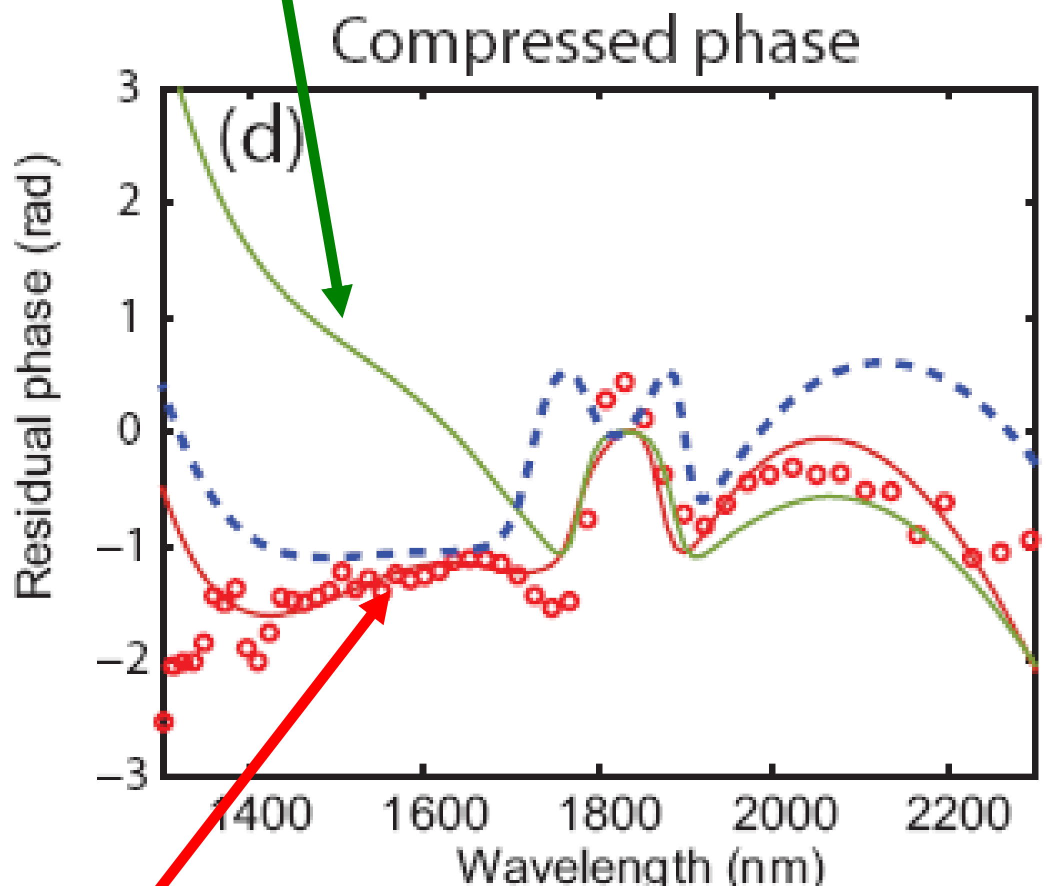


100 Hz: 400 μJ, 11.5 fs @ 1.8 μm



B. E. Schmidt et al. APL **96**, 121109 (2010).

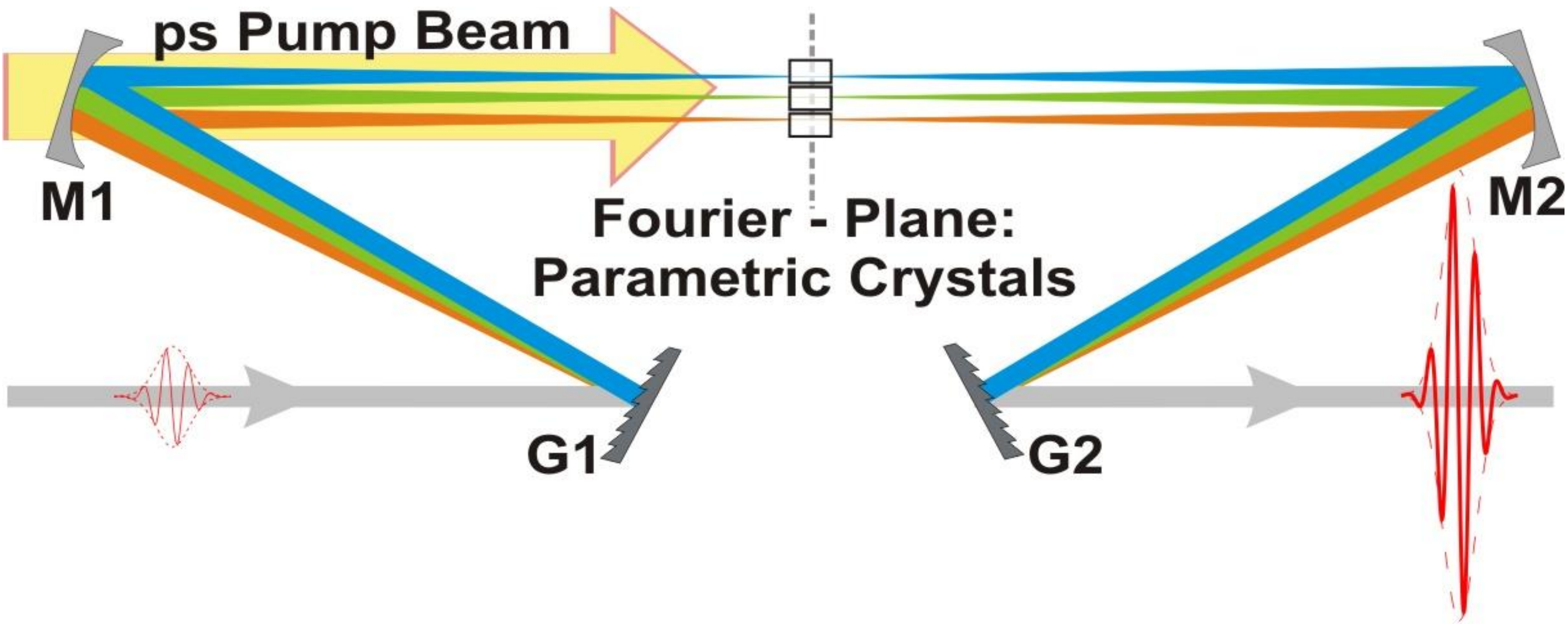
Only Self-Phase Modulation (SPM)

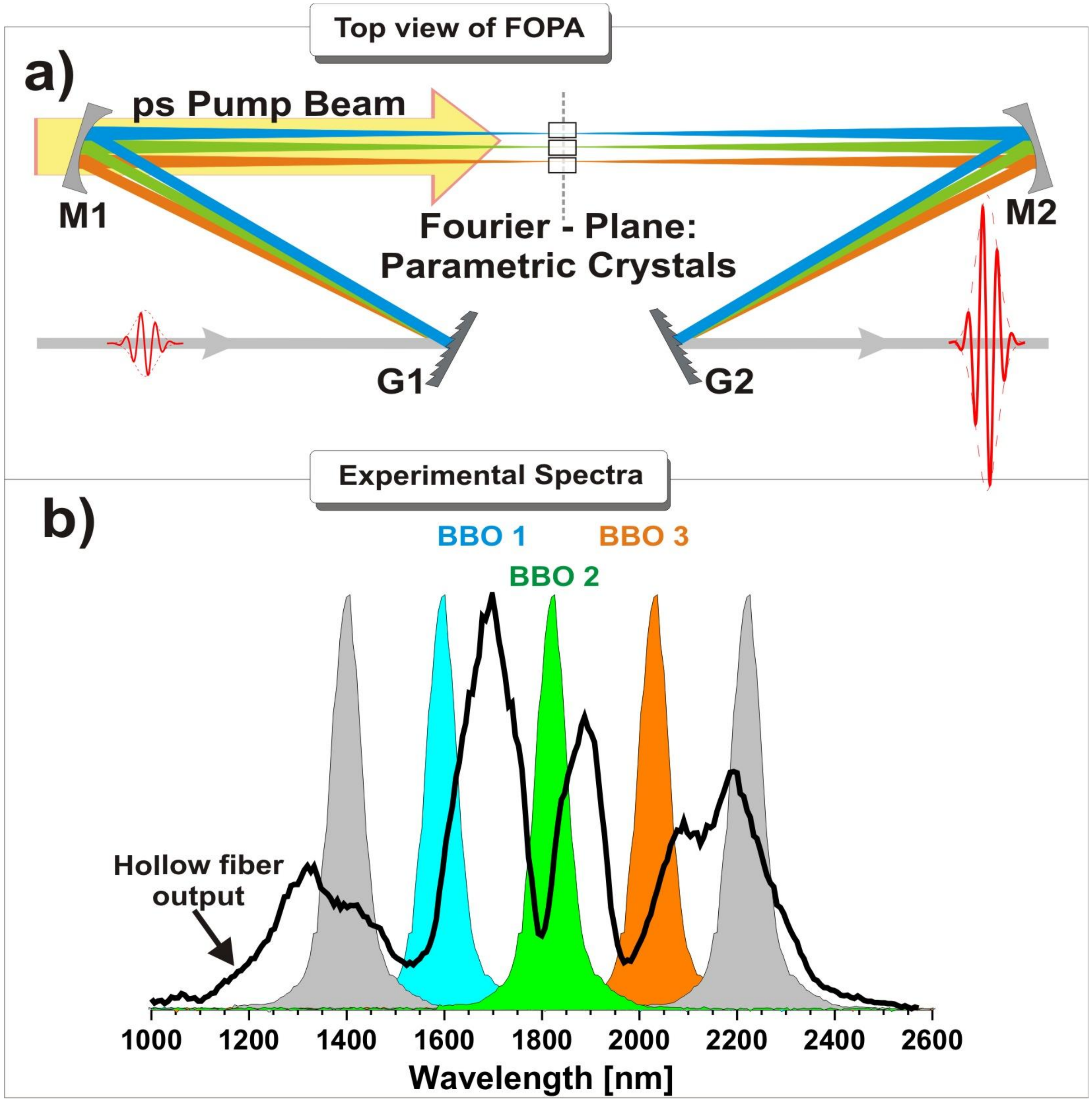


Full model: SPM + self-steepening

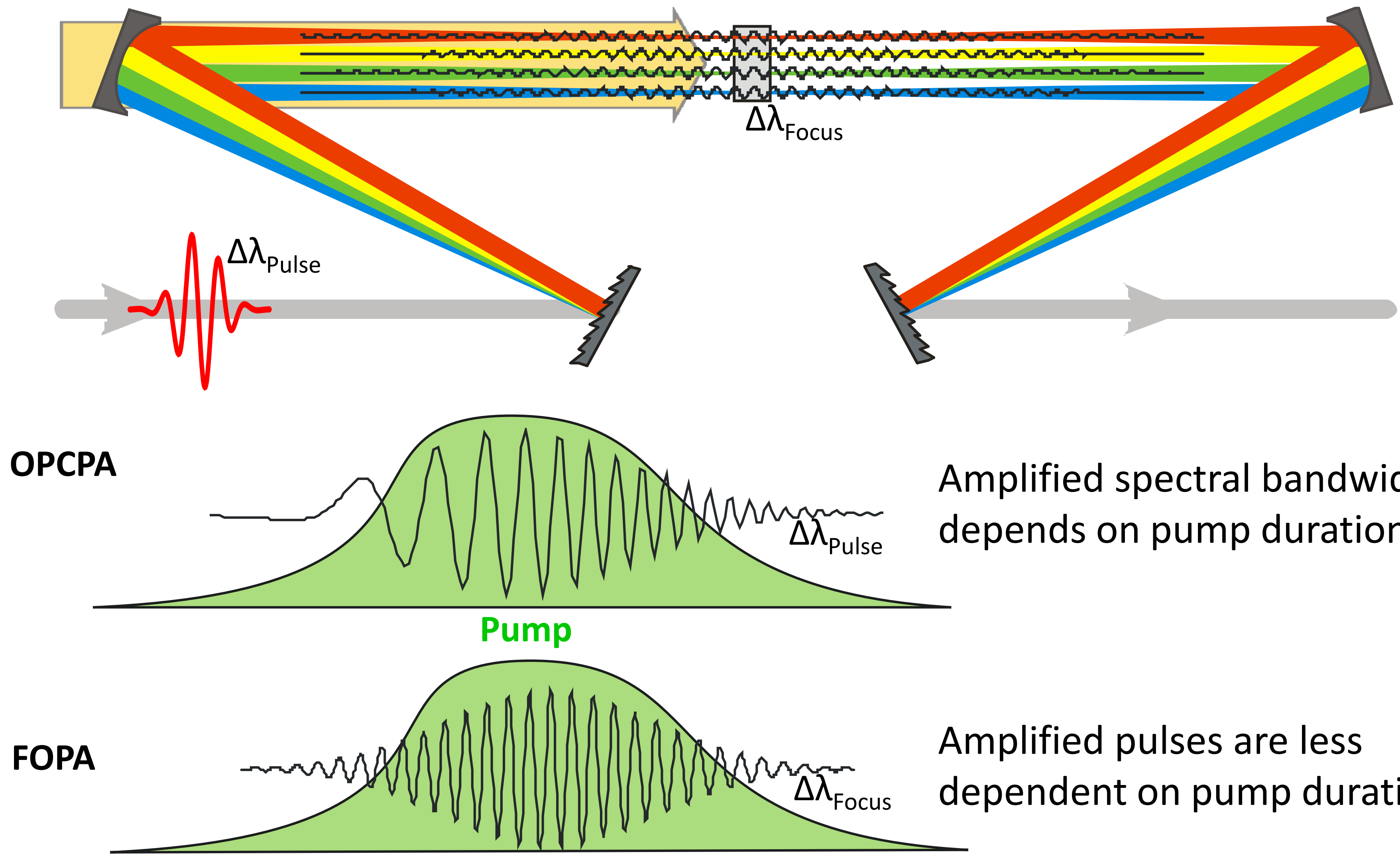
B. E. Schmidt et *al.* APL **96**, 121109 (2010).
P. Béjot et al. PRA **81**, 063828 (2010).

Frequency domain OPA




Top view of FOPA
a)
ps Pump Beam
M1
M2
Fourier - Plane:
Parametric Crystals
G1
G2
Experimental Spectra
b)
BBO 1
BBO 3
BBO 2
Hollow fiber
output
1000
1200
1400
1600
1800
2000
2200
2400
2600
Wavelength [nm]

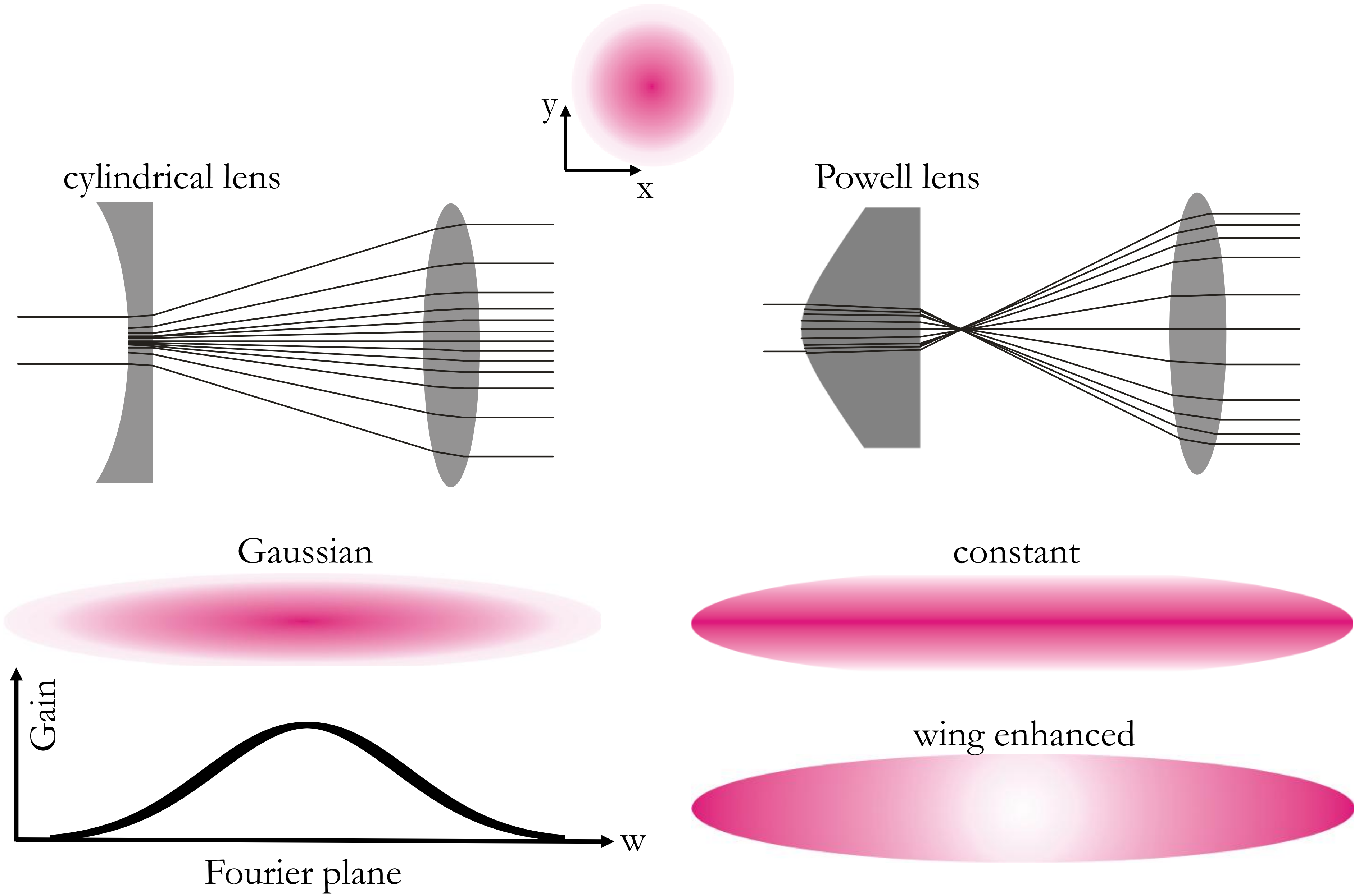
Transform Limited seed Pulses



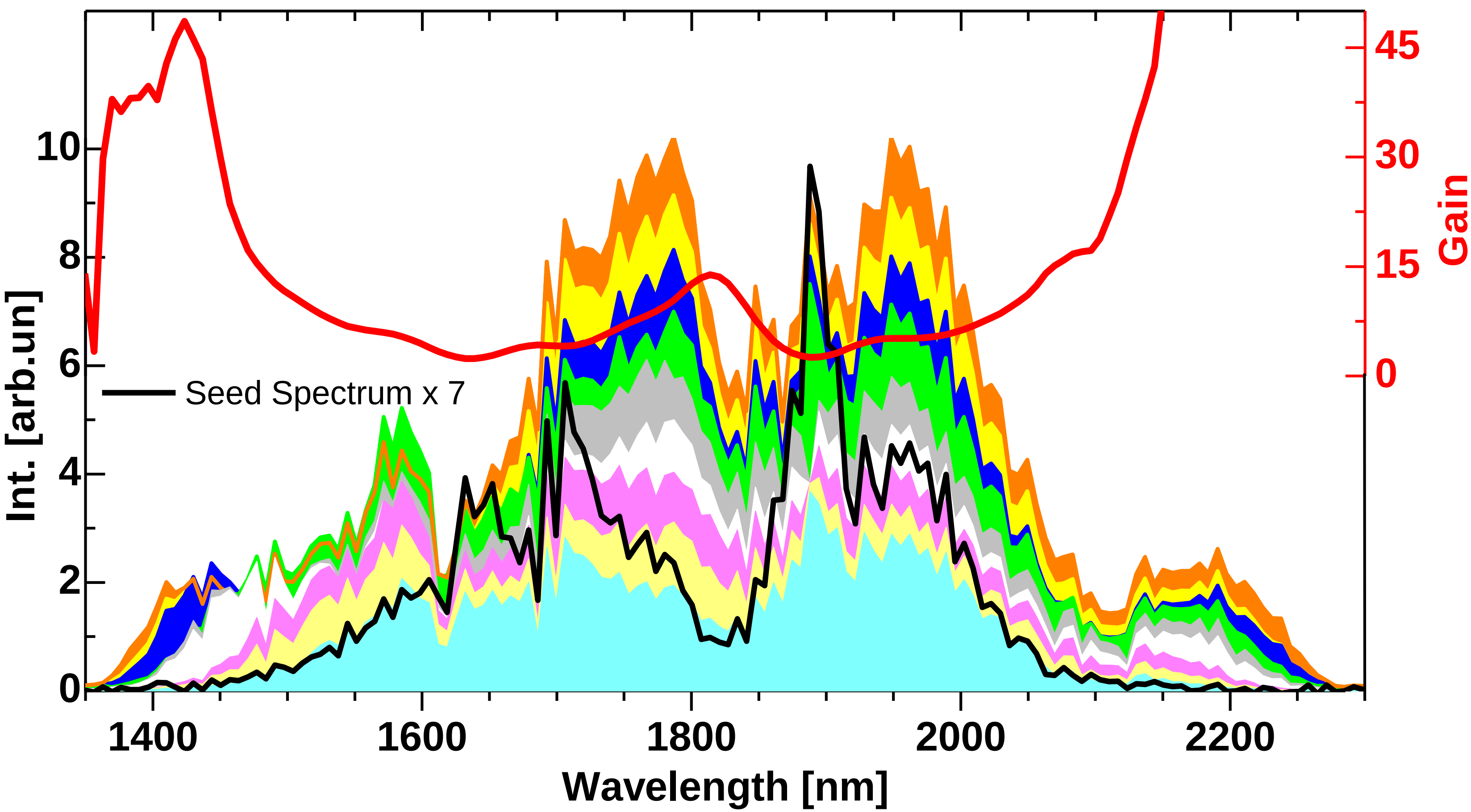
Amplified spectral bandwidth depends on pump duration

Amplified pulses are less dependent on pump duration

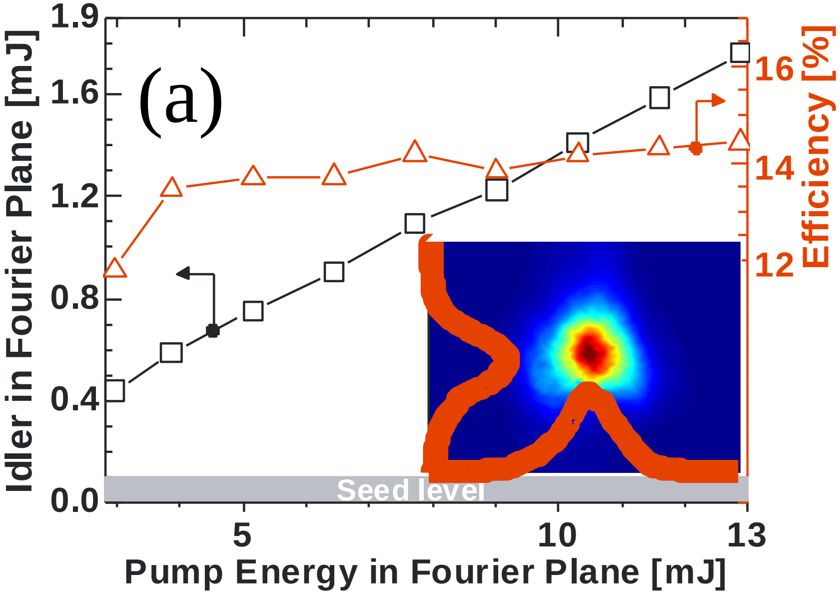
Gain Tailoring

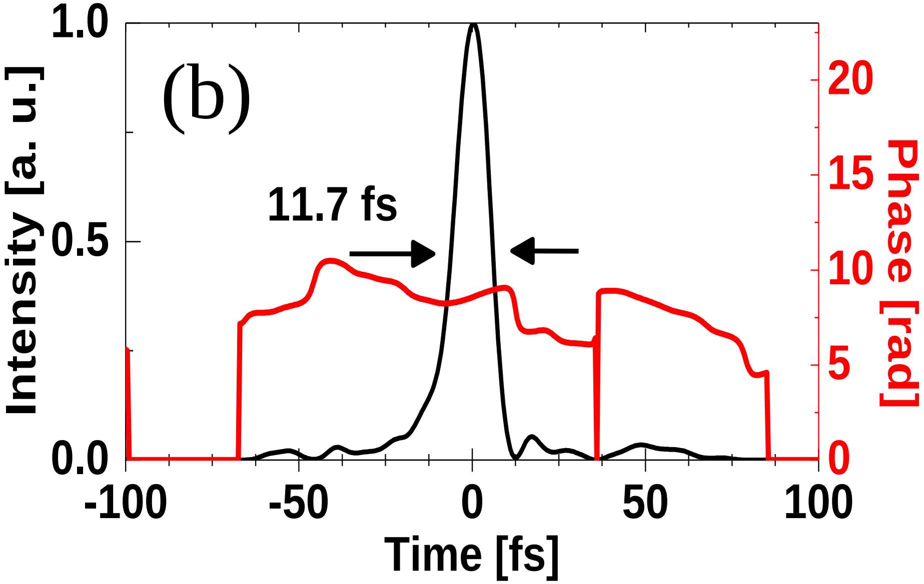


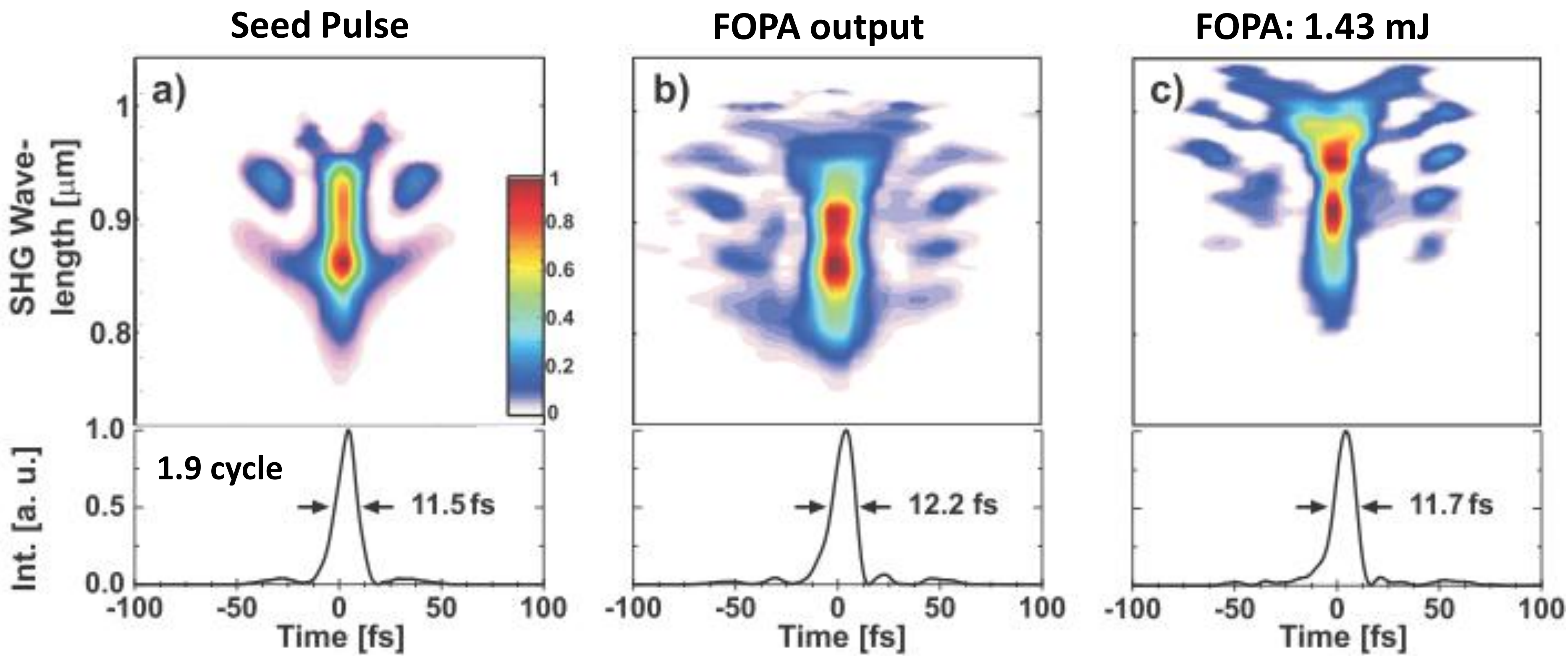
Amplified spectra as a function of pump energy

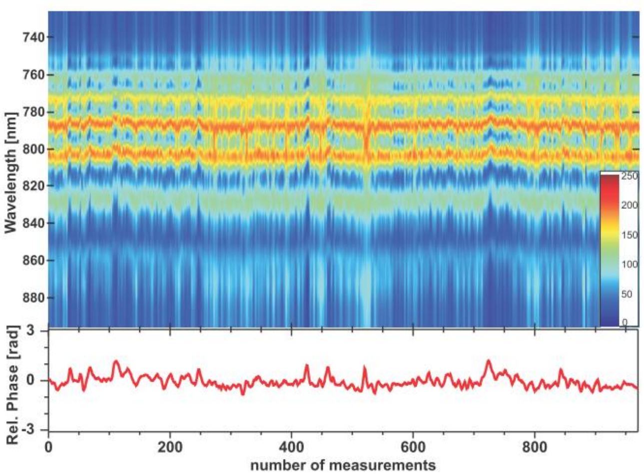


Energy per pulse as a function of pump energy









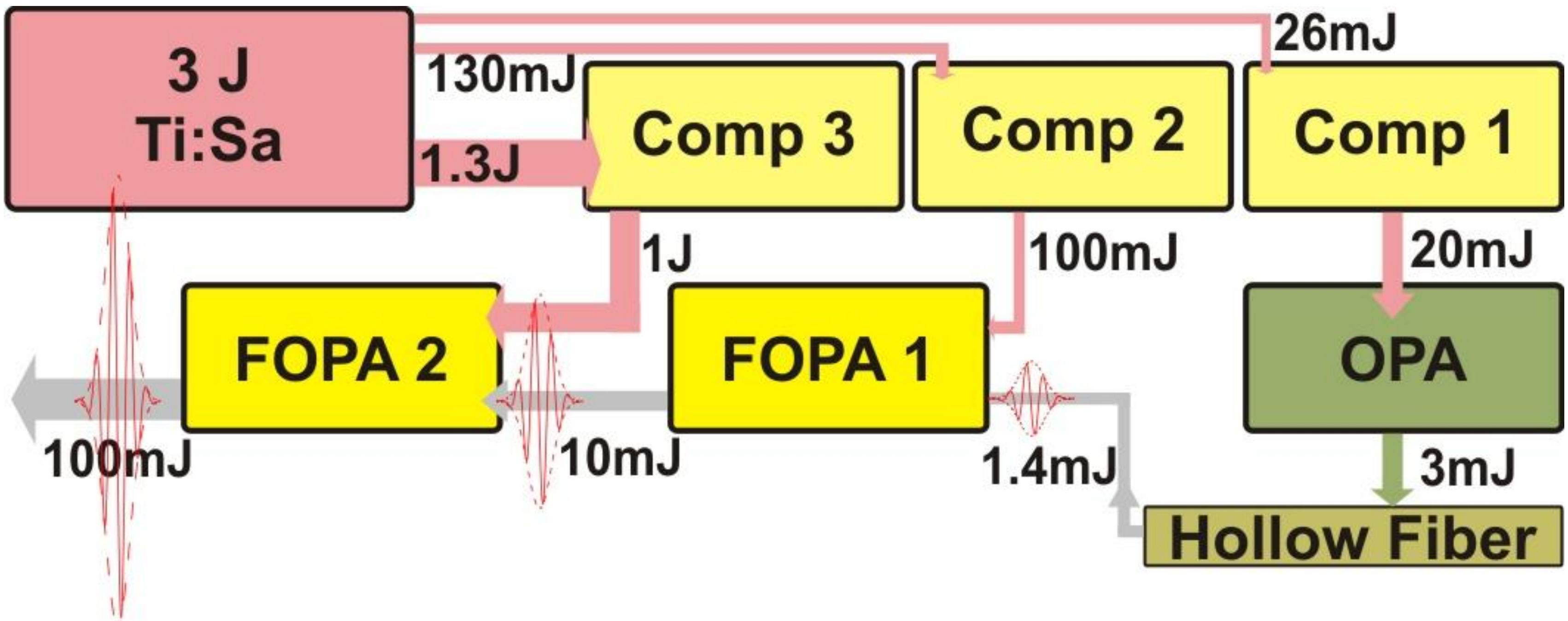
<u>CEP stability</u> at 50% amplification:
460 mrad measured over 1000sec,
each shot 10 times average
(350 mrad stability at input)

B. E. Schmidt et al. Nature Comm. **5**, doi:10.1038/ncomms4643.

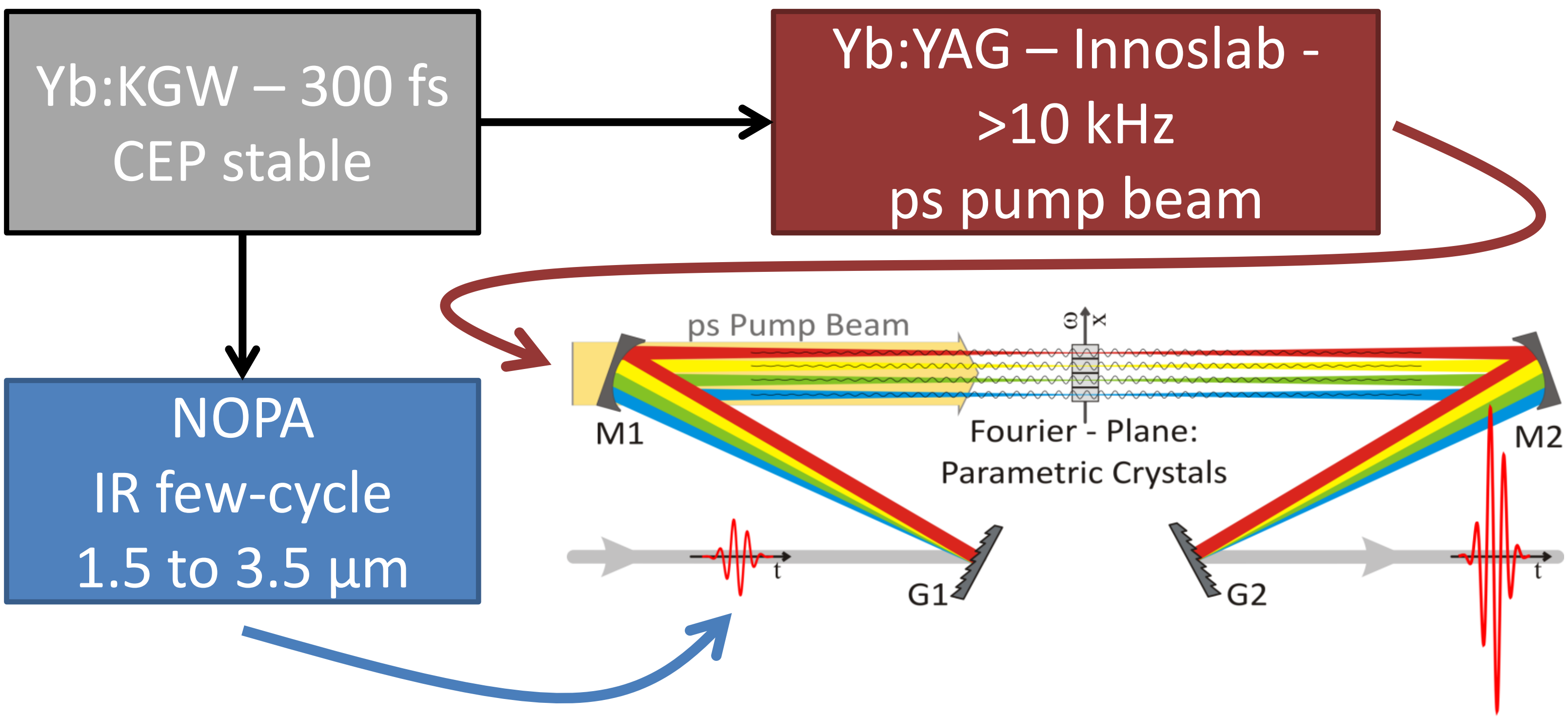
Advantages of FOPA

- Amplification of two cycle, 1.5mJ pulses
- Simultaneous upscaling of peak power and spectral bandwidth
- Upscaling not limited by aperture size of crystals
- No extra stretcher / compressor needed
- Absence of superfluorescence in the output beam
- No need for clean pump beam profile
- No Pulse Shaper for dispersion compensation
- Combination of many pump beams – no need for coherent synchronization.
- Amplification of arbitrary pulse shape possible

Short term plan – with Prof. Z. Chang



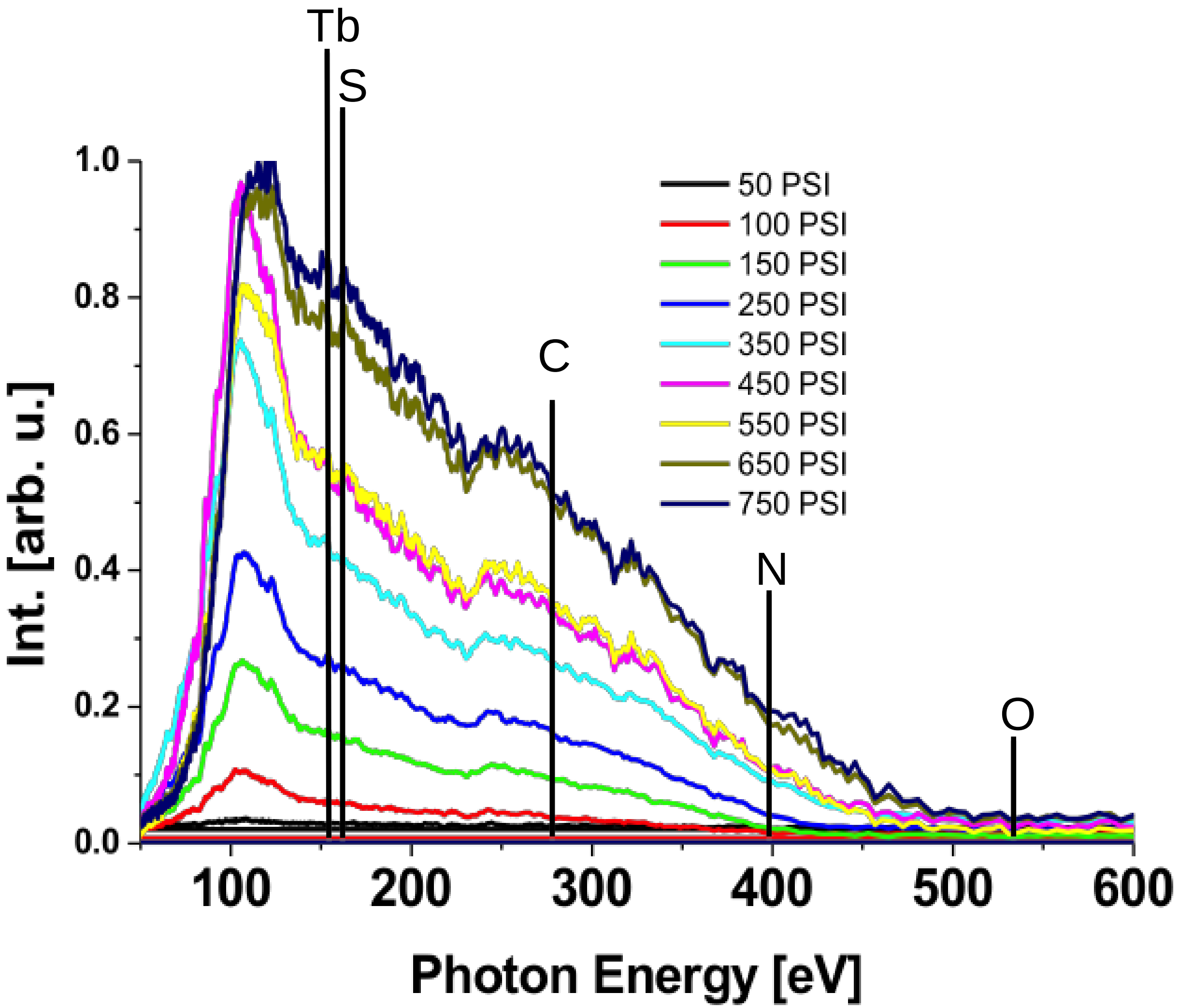
FOPA based on Yb (>10 kHz)



- **Expected output:** 75 W at 1.5 µm, 35 W at 3.5 µm. Few-cycle pulses. CEP stability. FOPA based on KTA crystals.
- **Future:** Scaling the average/peak power by multiplexing pumping in the Fourier plane.

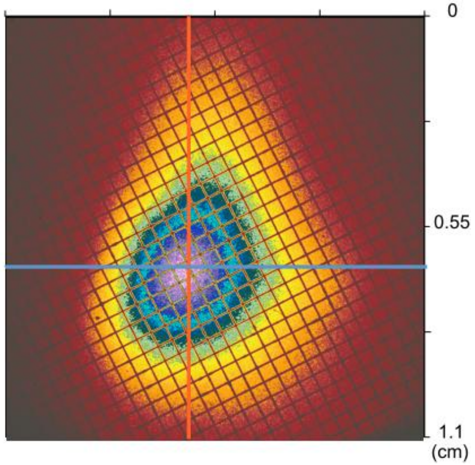
High brightness attosecond soft X-ray pulses

Scaling HHG (He with 1.8 μm)



Absolute flux

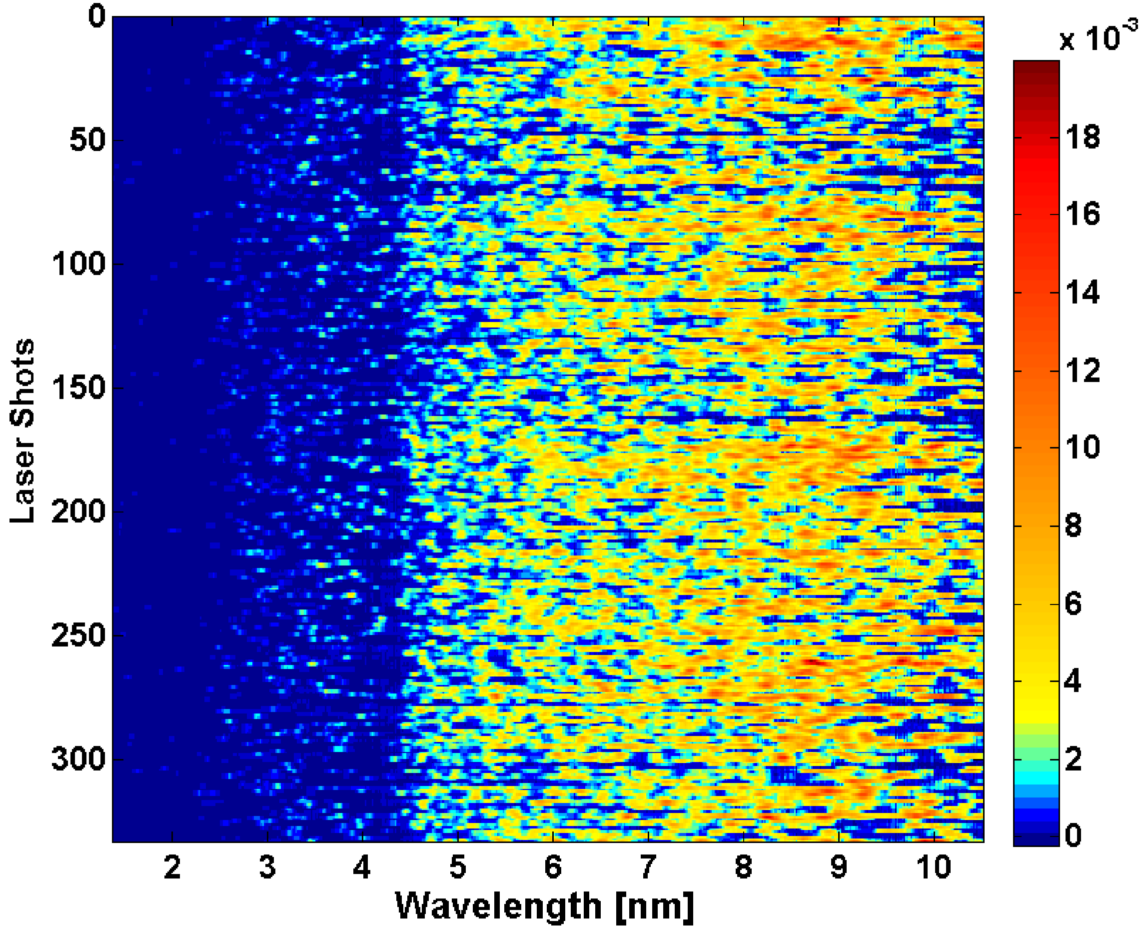
> $2\cdot10^5$ photons/shot beyond carbon K-edge



div < 2mrad

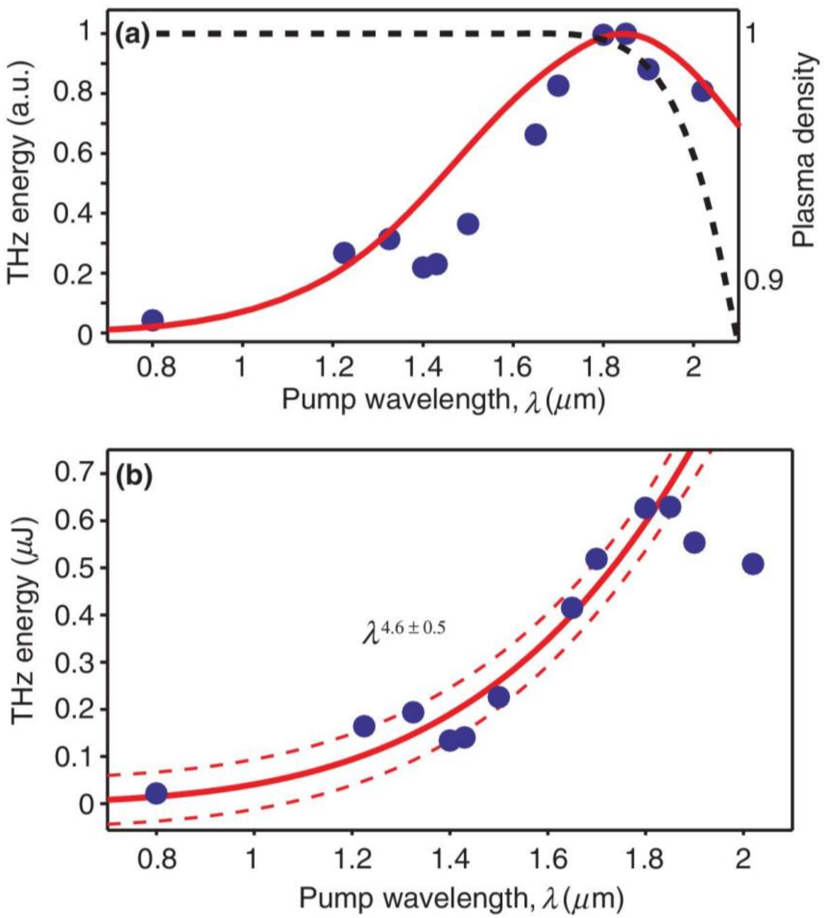
B. E. Schmidt et *al.* (in preparation).

Single shot carbon k-edge



Other applications of the IR beamline

THz generation with ω/2ω



Wavelength dependent parameter:

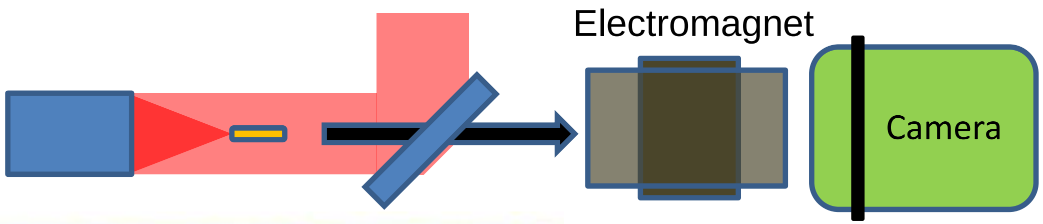
- Transverse plasma photocurrent $\propto \lambda^2$
  Proportionnal to the ponderomotive force ($I\lambda^2$)
- Plasma volume $\propto \lambda^3$
  Rayleigh range
  Beam diameter at focus

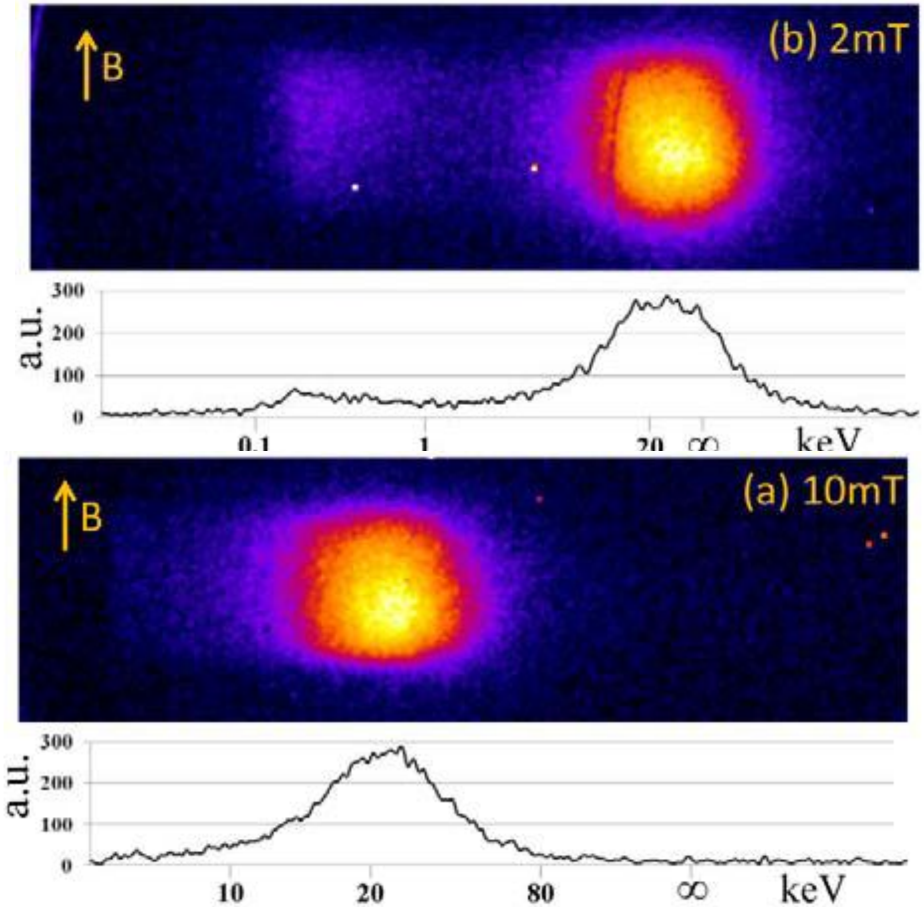
$$I_{THz}(\lambda) \propto \lambda^{4.6 \pm 0.5}$$

Single cycle THz pulses with 4.4 MV/cm

M. Clerici et al. *Phys. Rev. Lett.* **110**, 253901 (2013).

Longitudinal electron acceleration

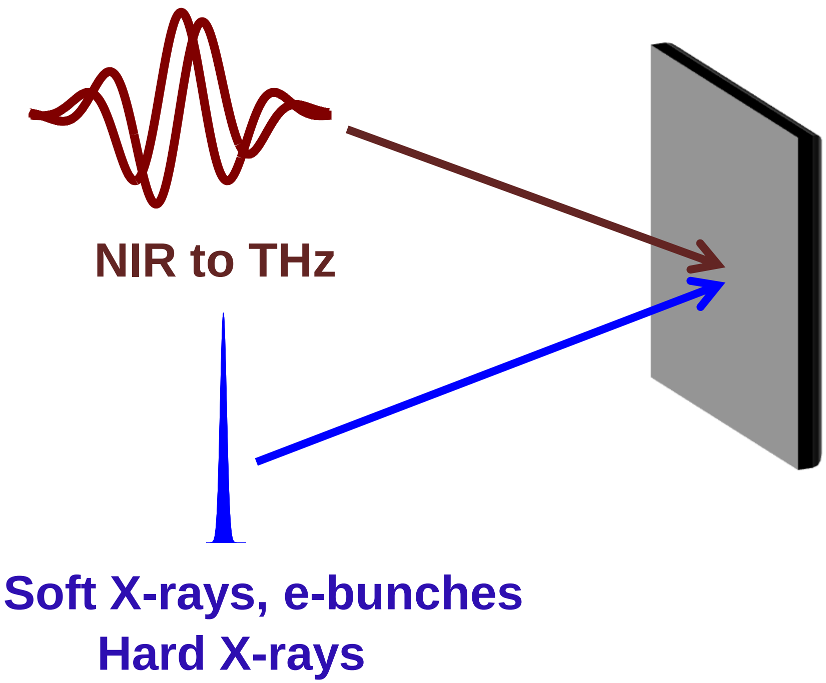




15 fs 0.5 mJ 1.8 µm
Peak @ ≈ 23 keV (9%c) 10^6 e^-/pulse
Simulations: ≈ 125 keV if 5 mJ/pulse

S. Payeur et *al. Appl. Phys. Lett.* **101**, 041105 (2012).

Dynamic imaging platform



- Pumping with tunable CEP stable pulses from NIR to THz.
- Probing using time-resolved soft X-ray measurements
  - absorption
  - reflectivity
  - diffraction
  - photoelectrons
- Ultrafast electron diffraction imaging
- Ultrafast hard X-ray diffraction imaging

J. Weisshaupt et *al.* CLEO: 2014, OSA Technical Digest (online) (Optical Society of America, 2014), paper SM1F.5.

Thanks to the team

Fonds de recherche
Nature et
technologies
Québec

Conseil de recherches en sciences naturelles et en génie du Canada

Natural Sciences and Engineering Research Council of Canada

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ICIP
CIPI

INRS Université d'avant-garde

ALLS Advanced Laser Light Source

NRC·CNRC uOttawa JASLab

Joint Laboratory for Attosecond Science Laboratoire commun de la science de l'attoseconde