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Our goal:

- Use an all optical technique to investigate tunneling dynamics; map attosecond dynamics onto a femtosecond time scale
- Move away from photoelectron spectroscopy– enable experiments on bulk

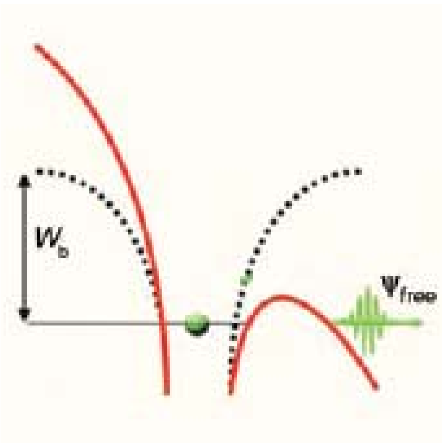
Outline

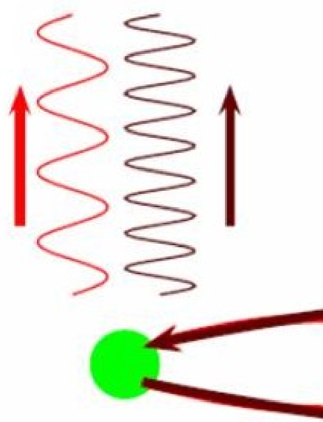
- How to read out tunneling ionization with an optical pulse?

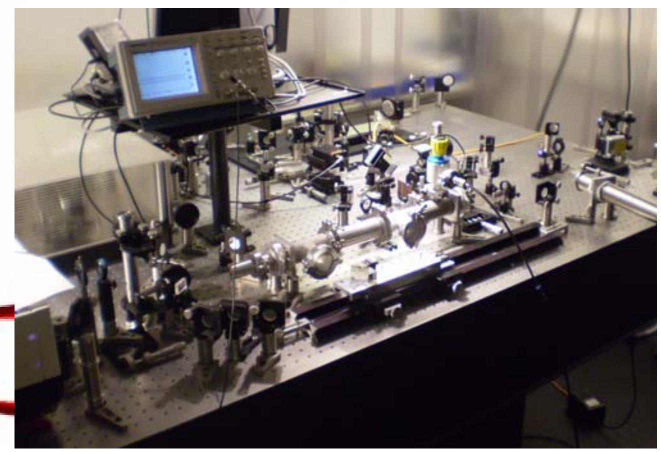
- Observation of recollision-free (Brunel) harmonics:
  from noble gas
  from bulk transparent solids

- Brunel mixing with two-color fields

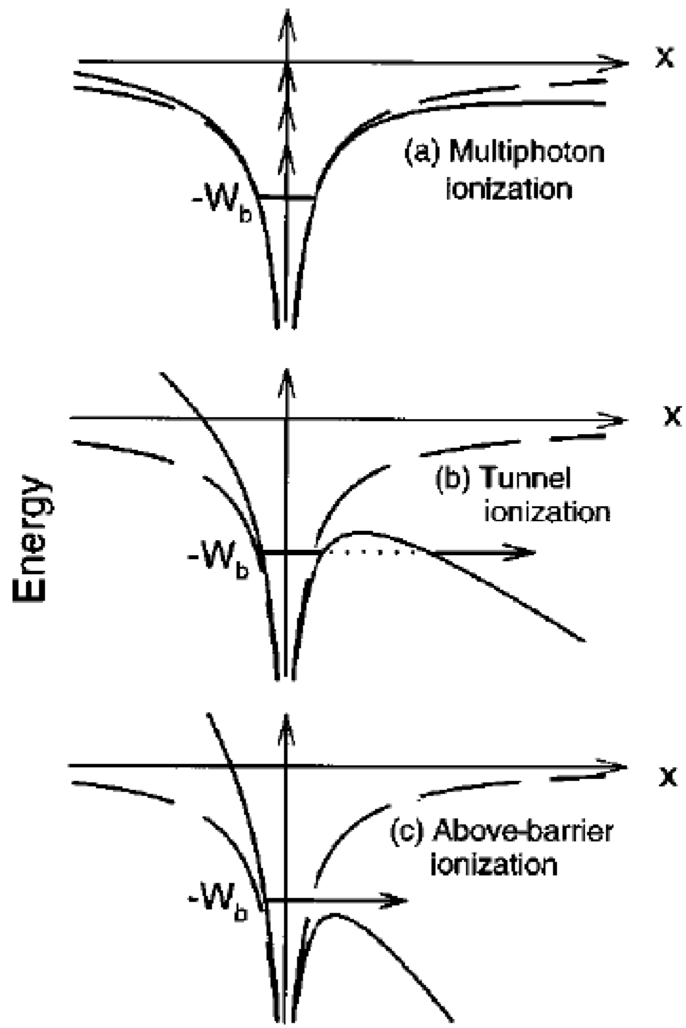
- Future: use 1.6 µm IR CEP OPA for bulk







Ionization Regimes



$$\gamma = \frac{\omega_L \sqrt{2mW_b}}{eE_0}$$ - Keldysh parameter

Multi-photon (MPI)

plasma-induced spectral blue-shift

Tunnel (TI)

harmonics generation

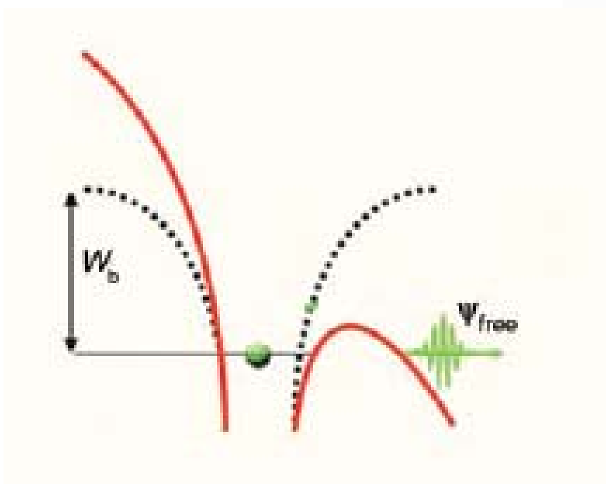
$\gamma>1$ – tunneling rate slower than laser period
Multiphoton ionization

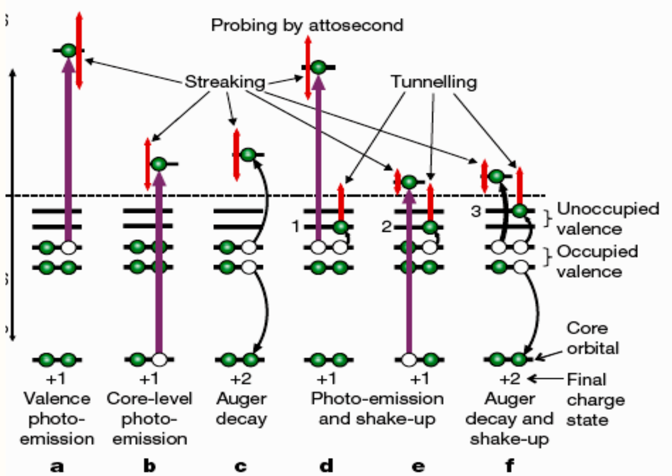
$\gamma<1$ – tunneling rate faster than laser period
Tunnel ionization

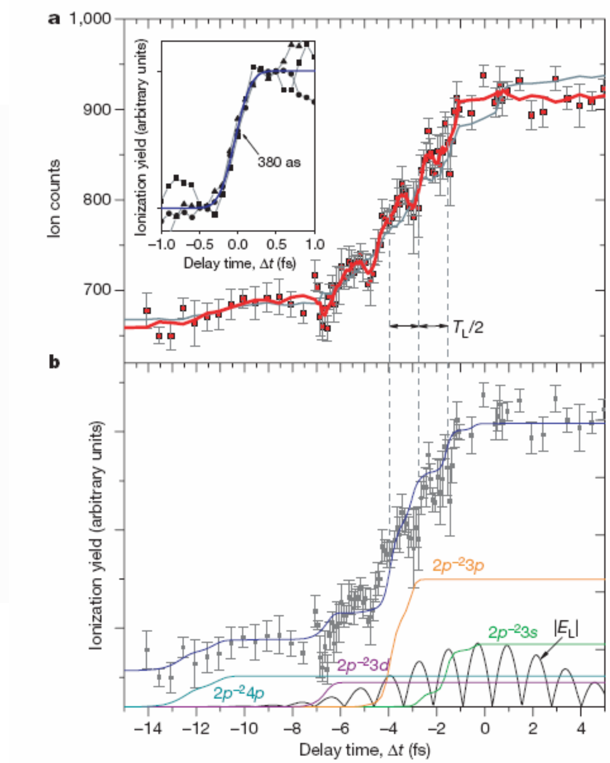
Keldysh (1965)

Motivation

- Real-time observation of tunneling ionization dynamics
  - Uiberacker *et al.*, *Nature* **446**, 627 (2007)





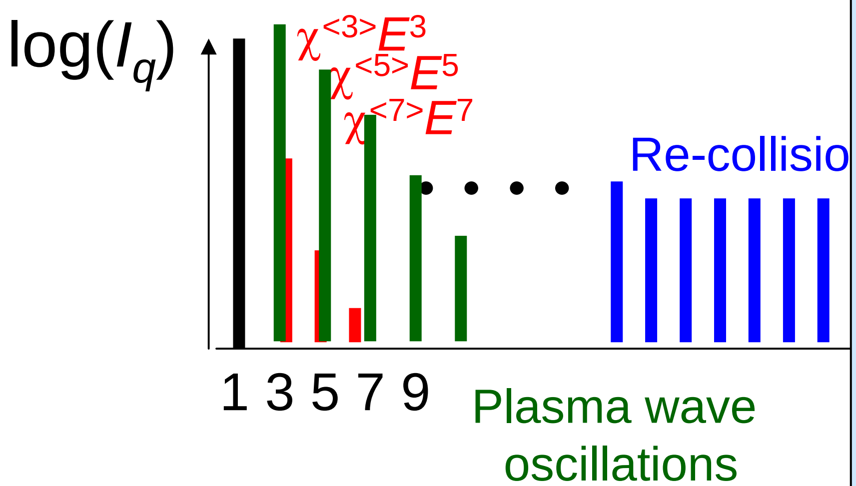


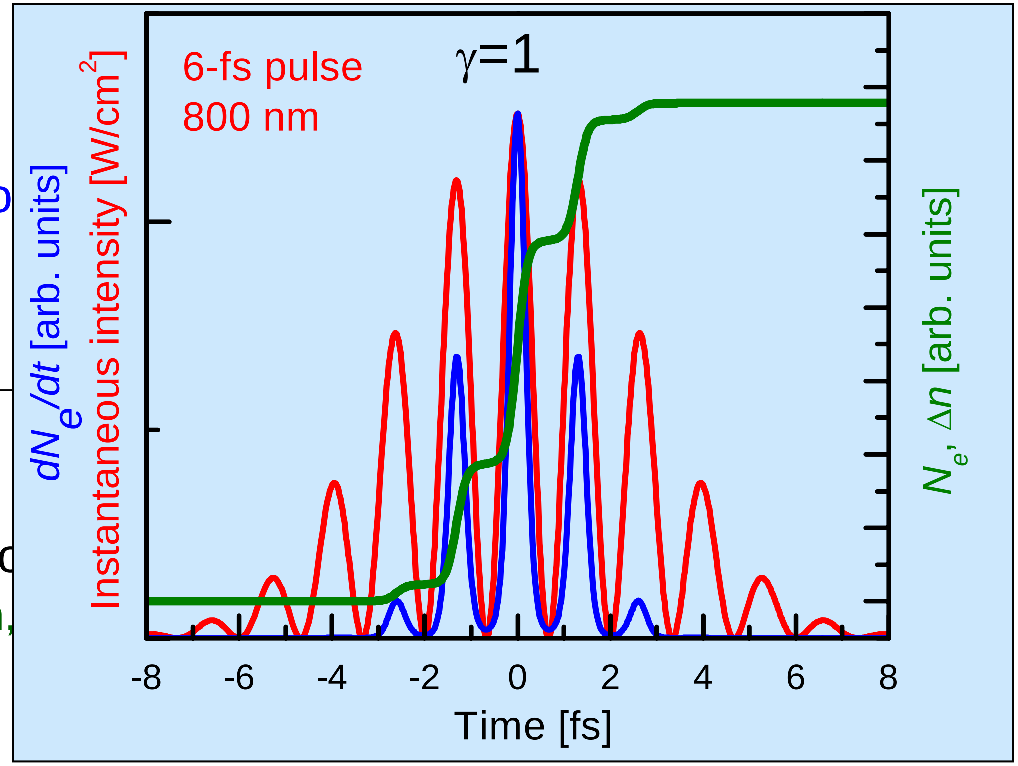
  - Attosecond angular streaking using circular polarized light and COLTRIMS

    P. Eckle *et al.*, *Nature Physics* **4**, 565 (2008)

We set out to develop an optical read-out technique that can work with bulk solids!

Mechanisms of Higher-Order Harmonic Generation





High orders (**Corkum**): Tunnel ionizatio

P.B. Corkum,

Lowest orders:

$\chi^{<3>}$, $\chi^{<5>}$, $\chi^{<7>}$ …

Brunel: twice-per-cycle tunnel ionization ➔ step-wise plasma concentration increase ➔ transverse plasma current $J_\perp = en_e\mathbf{v}$

F. Brunel, *JOSA B* **7**, 521 (1990)

Origin of new frequency components due to the Brunel mechanism: rapid time-domain phase modulation!

Harmonics signal is independent of the final state of electrons!

Previous Work

Classical models for harmonic generation based on high-frequency variation of the tunnel ionization current:

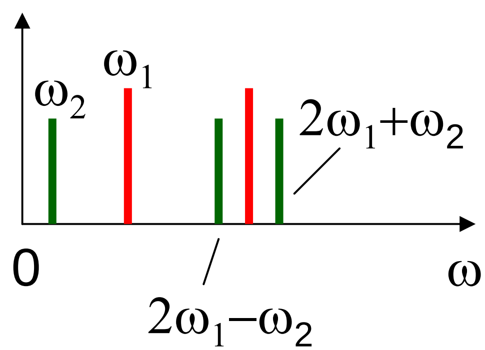
Theory:

F. Brunel, *JOSA B* 7, 521 (1990)
S. Rae and K. Burnett, *J. Phys. B* **26**, 1509 (1993)

These models predict the right magnitude of 3rd and 5th harmonics, but no plateau:

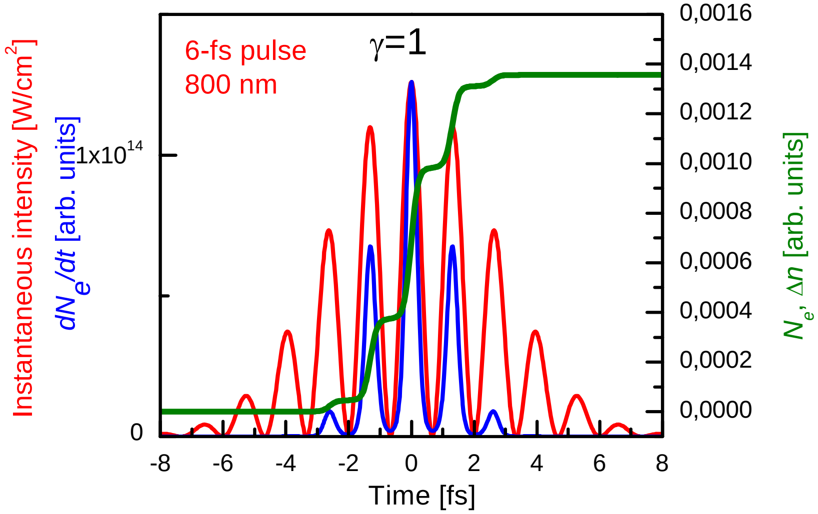
N. Burnett, C. Kan, P.B. Corkum, *PRA* **51**, R3418 (1995)

Experiment:



"Brunel mixing" in gas.
C.W. Siders *et al.*, *PRL* **87**, 263002 (2001)
Driven by laser ω_1, n_e oscillates at $2\omega_1$
$\omega_3 = 2\omega_1 \pm \omega_2$ → for $\omega_1 = \omega_2$ predicts THG and THz emission on the leading pulse edge. (Similar to THz emission via 4 wave mixing $2\omega-\omega-\omega$
(D. Cook, et al. *Opt. Lett.* **95**, 1210 (2000),
M. Kress, et al. Nat. Phys. **2**, 327, (2006))

Time Dependent Refractive Index Modulation



Analytical expression for nonadiabatic tunnel ionization:
G. L. Yudin and M. Yu. Ivanov, *PRA* **64**, 013409 (2001)

3-D TDSE in good agreement with the Yudin-Ivanov formalism:
Uiberacker *et al.*, *Nature* **446**, 627 (2007), Suppl. Information

Refractive index change:

$$\Delta n_p \approx -\frac{\omega_p^{\ 2}}{2\omega^2}$$

$$\omega_p^2 \propto n_e(t)$$

→

Time-dependent phase shift:

$$\Delta\varphi_p(t) \propto \frac{1}{\omega^2}$$

→

Can be read out only by an optical field

Not accessible in the XUV!

Tunneling Dynamics in Bulk Solids

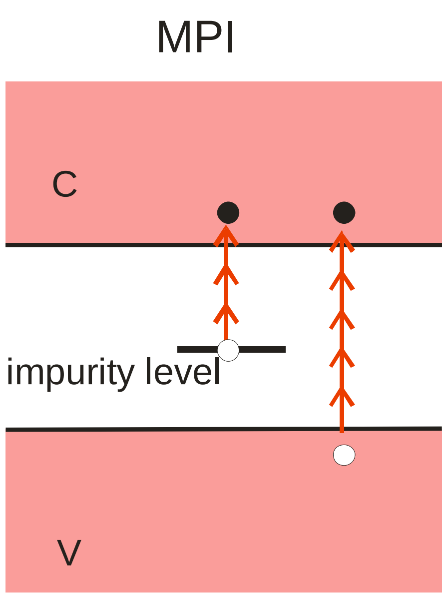
Proof of principle experiment in gas phase

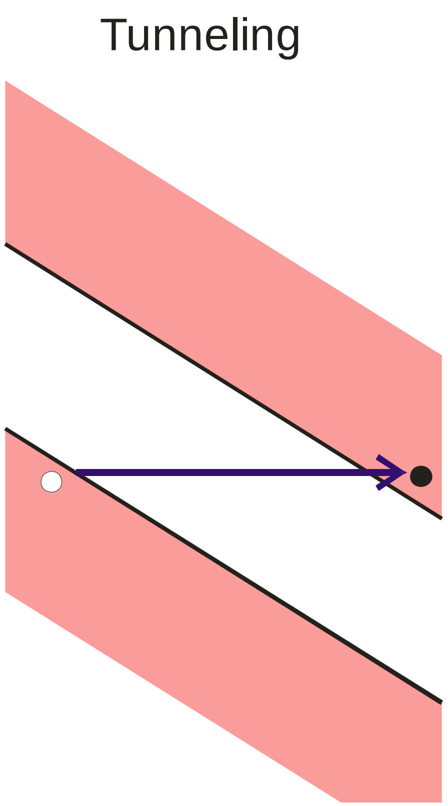
Strategic goal: To develop a technique for investigation of TI dynamics in bulk solids

Ionization is the starting point for all strong field phenomena…

It is also starting point for optical breakdown

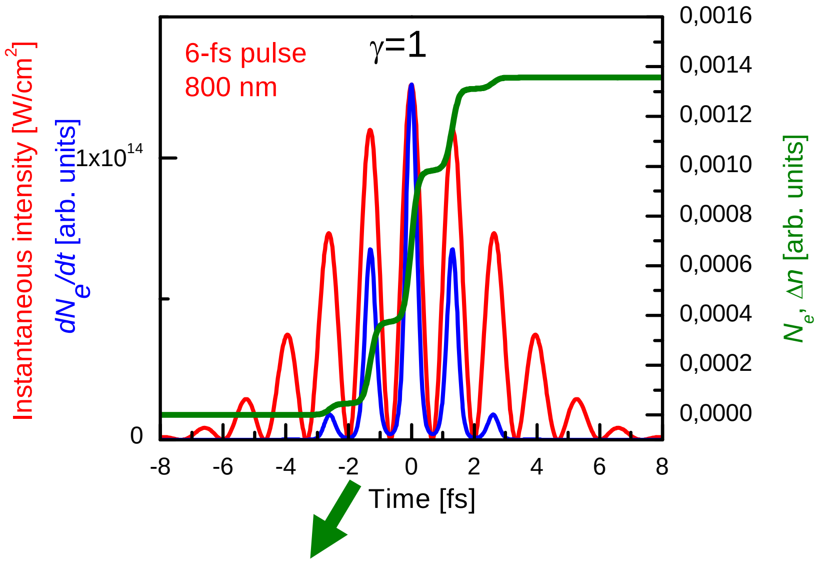
Photo-electrons cannot be observed from bulk material !

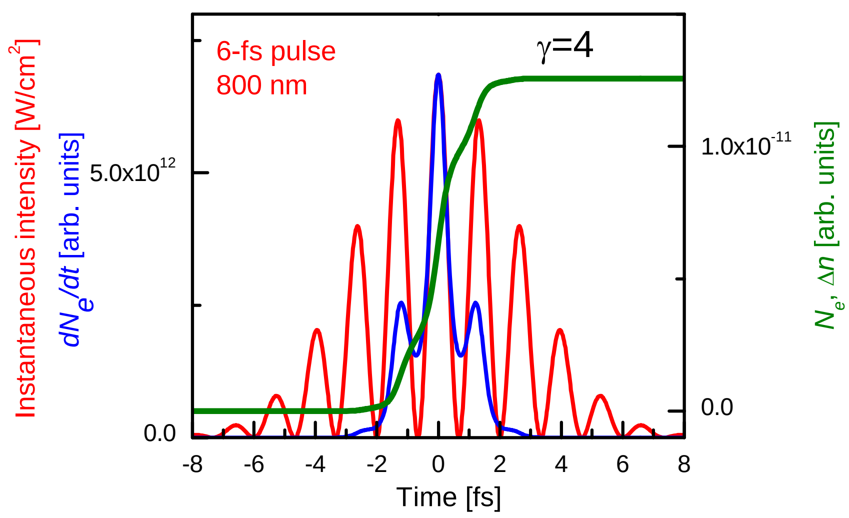




Stepwise *vs.* Smooth Refractive Index Modulation?

$$\gamma = \frac{\omega_L \sqrt{2mW_b}}{eE_0}$$ - Keldysh parameter





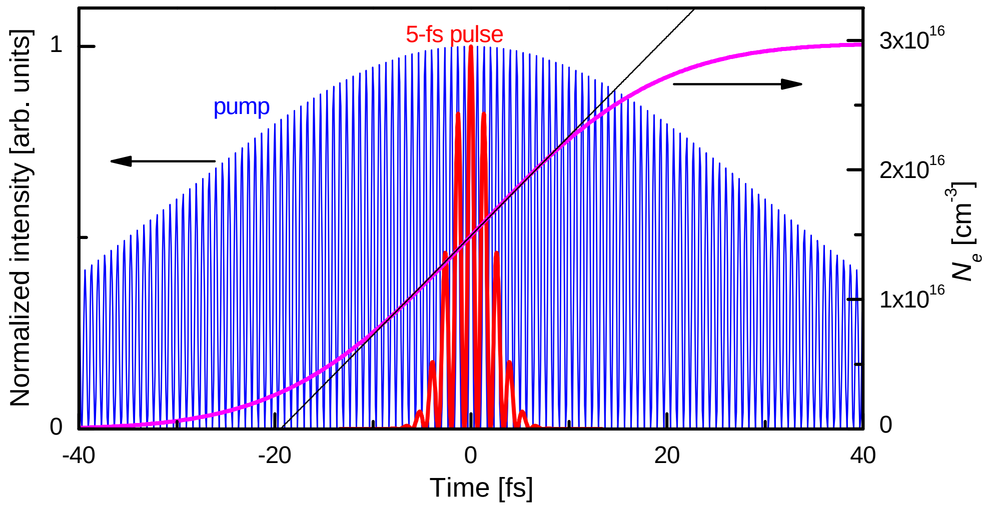
Attosecond time structure due to strong dependence of ionization probability on the field strength

TI $\gamma \longrightarrow$ **MPI**

Smooth ramp due to MPI

(follows intensity envelope)

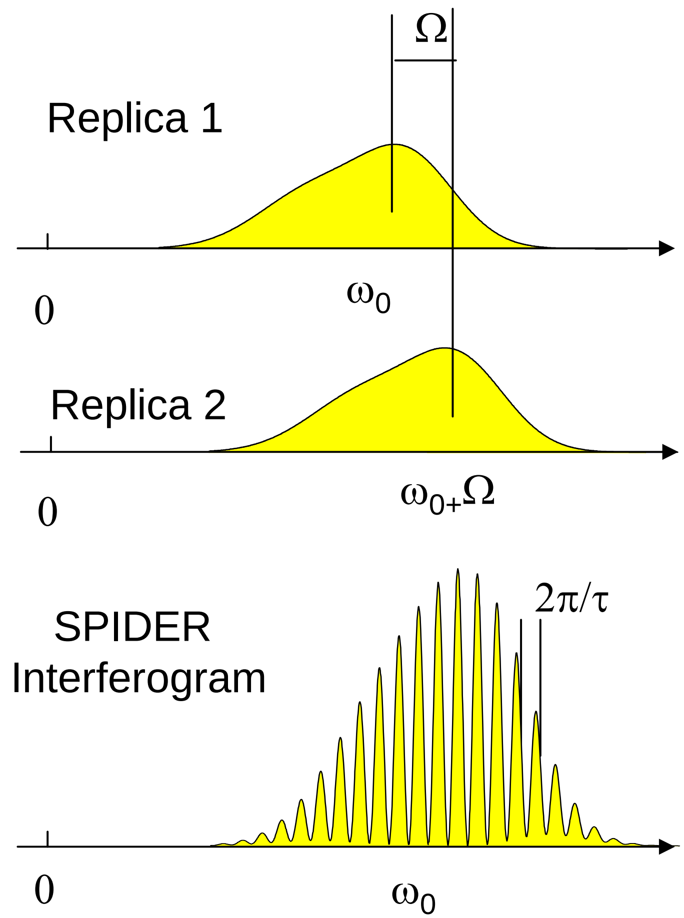
Quasi-Linear MPI Ramp



i(onization)-Spider

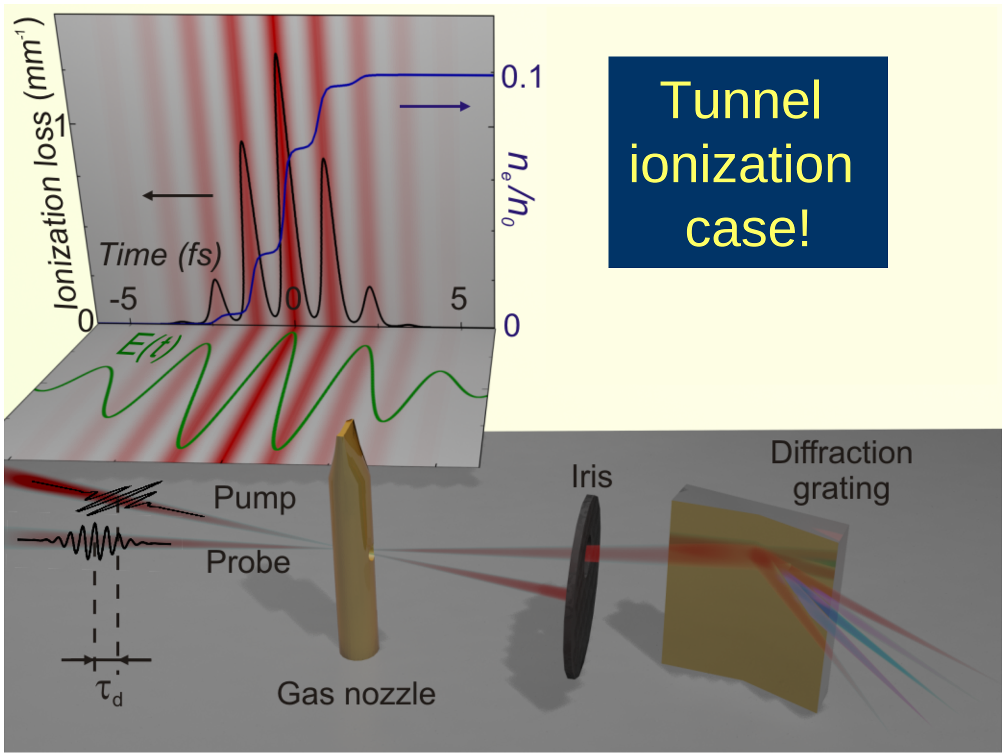
Plasma-Blue-Shift spectral shear interferometry for characterization of ultimately short optical pulses

Base harmonic frequency shift



A. Verhoef et al., *Opt. Lett. 34, 82* (2009)

Attosecond Phase Mask



Advantage:

Brunel harmonics do not depend on the final state of electrons

Harmonic frequencies

$N \times 2\omega_{pump}+\omega_{probe}$, N=1,2,3…

Time domain:
Attosecond phase mask
Showing tunnel ionization dynamics

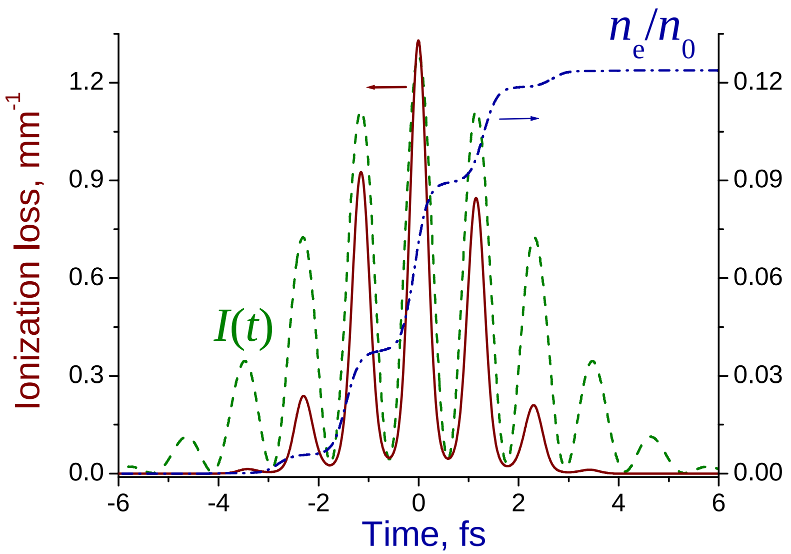
⇄

Frequency domain:
Harmonic spectra

Question: How badly is the attosecond time-domain phase mask distorted by pulse propagation?

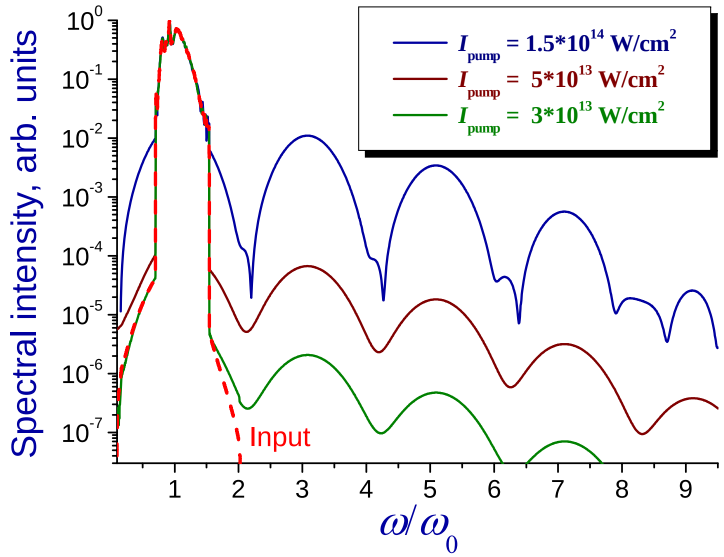
Spectral Response to Temporal Phase Modulation

Formation of time-dependent attosecond phase mask



Formation of harmonic spectrum

"Spectral scattering"

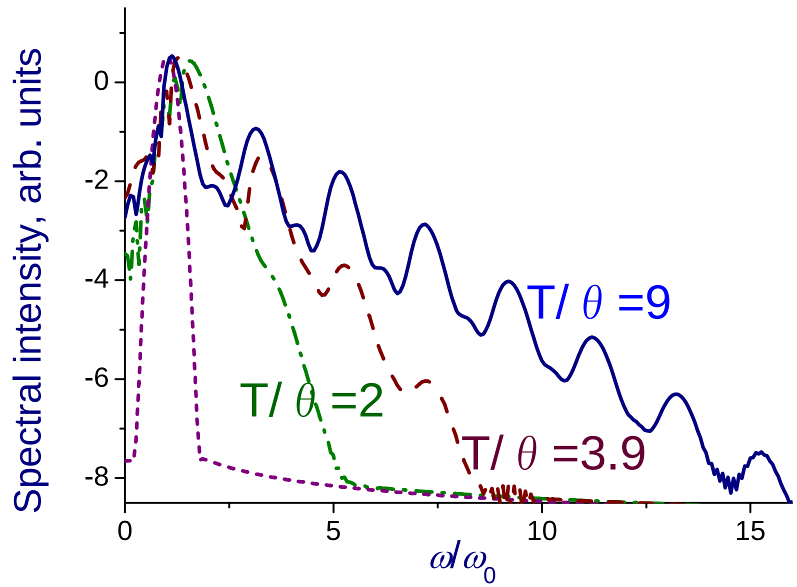


Interpreting Spectral Signatures

Model:

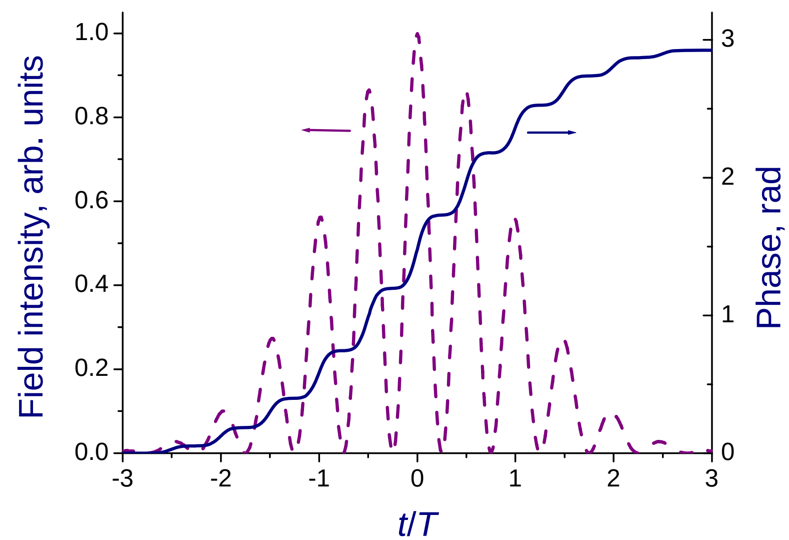
Phase mask Φ(t) with different finite rise time θ

This work: A. Zheltikov and E. Serebryannikov, 3-D propagation code for Brunel and Kerr harmonics based on Yudin-Ivanov formalism



Spectra of the resulting laser field for different phase masks

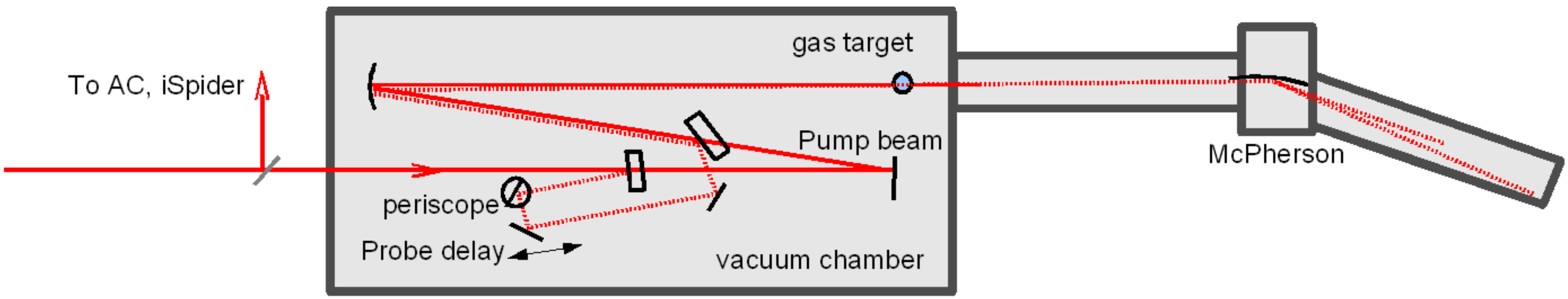
Phase mask for T/ θ = 9



T – laser period

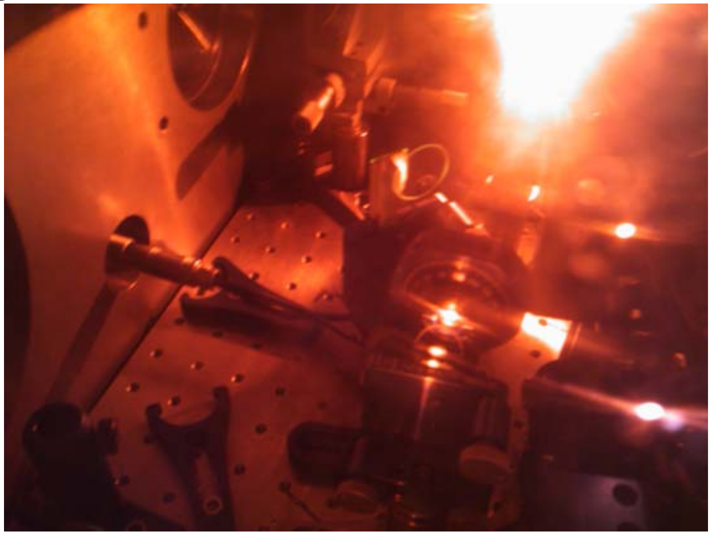
Power ratio between adjacent harmonics orders depends on the speed of electron density release (Δn step sharpness)

Experimental Setup



λ_c=750 nm

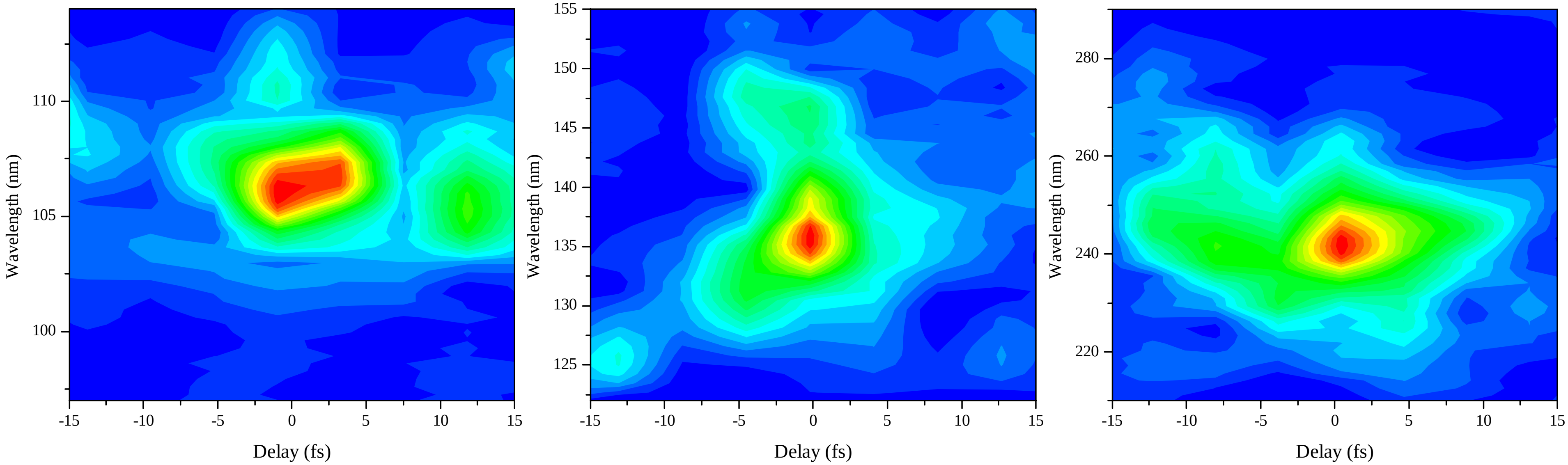
- Pump: 5 fs, 200 μJ; Probe: ~20 fs, 2 μJ.
- The harmonics are detected in the direction of the weak, cross polarized chirped probe pulse. The pump beam is blocked before the entrance slit of the spectrometer.
- ω_{probe} may differ from ω_{pump} to see the effect of the phase mask.

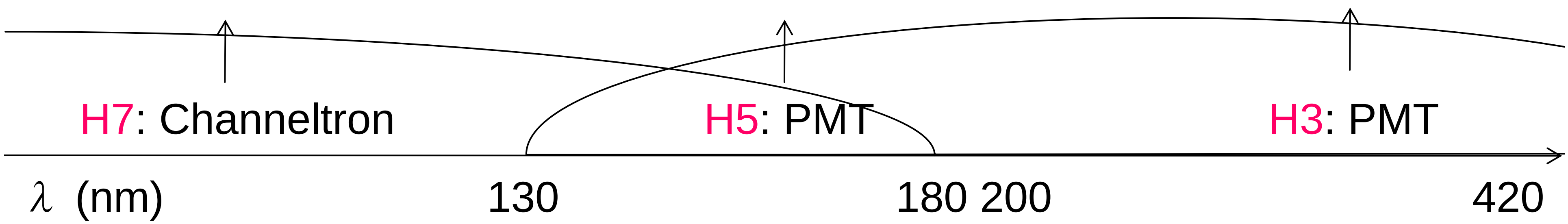


Results

Harmonics

experiment

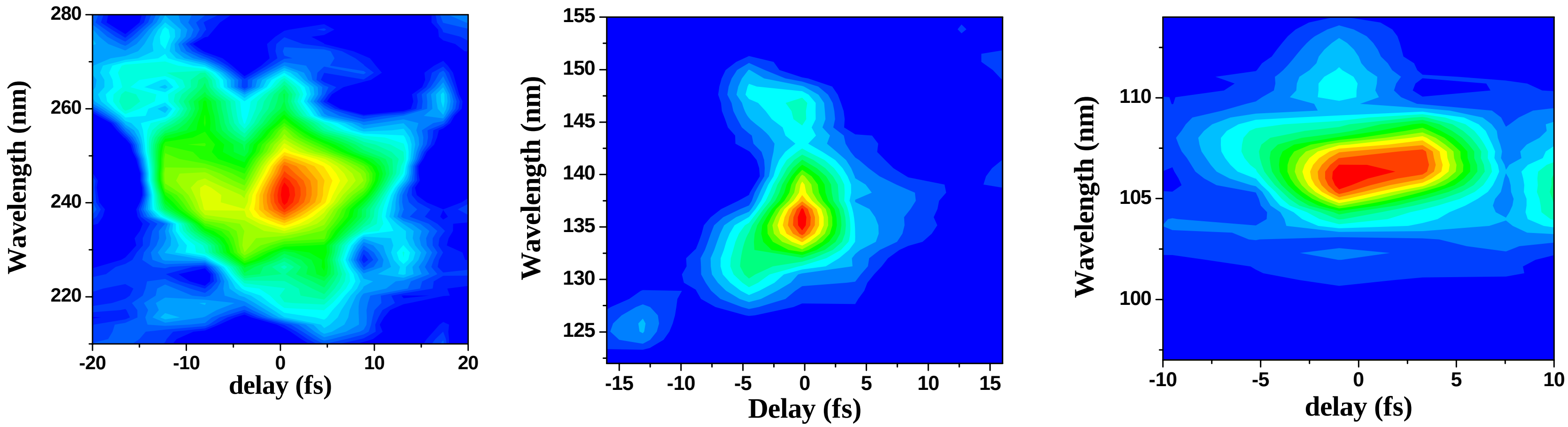




signal detected in the direction of a weak cross polarized chirped probe pulse

Experiment vs. Simulation

experiment

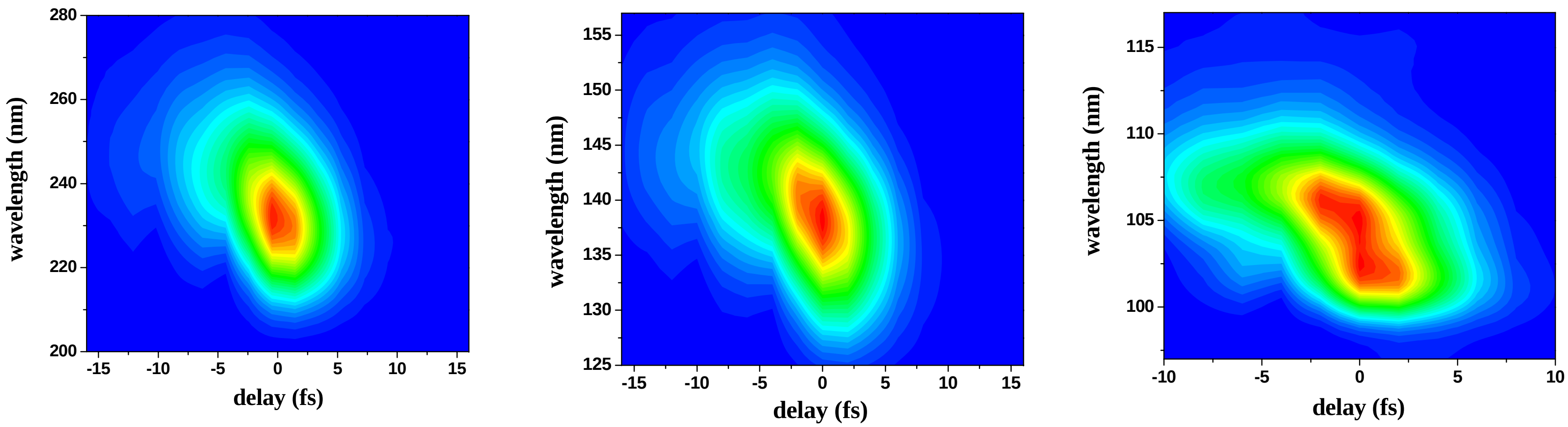


H3

H5

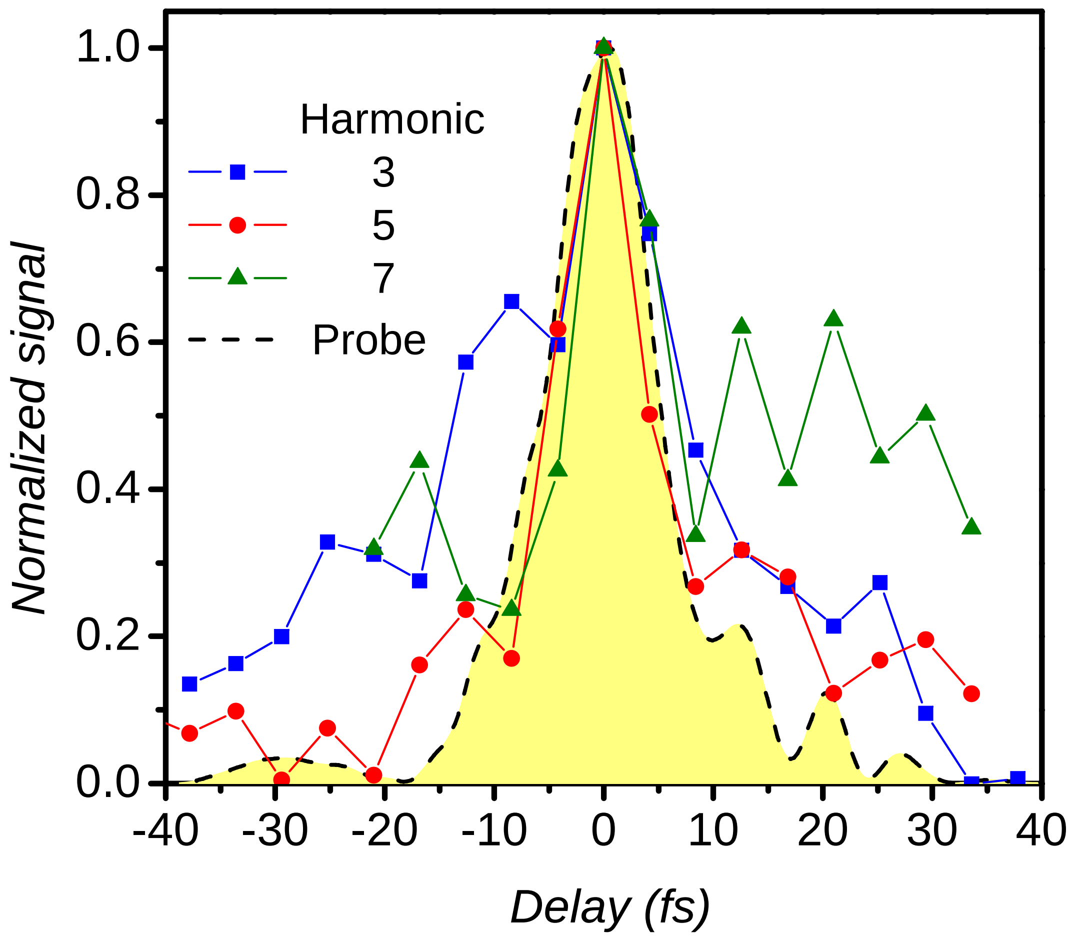
simulation

H7



300 mBar Kr

Cross-Correlations



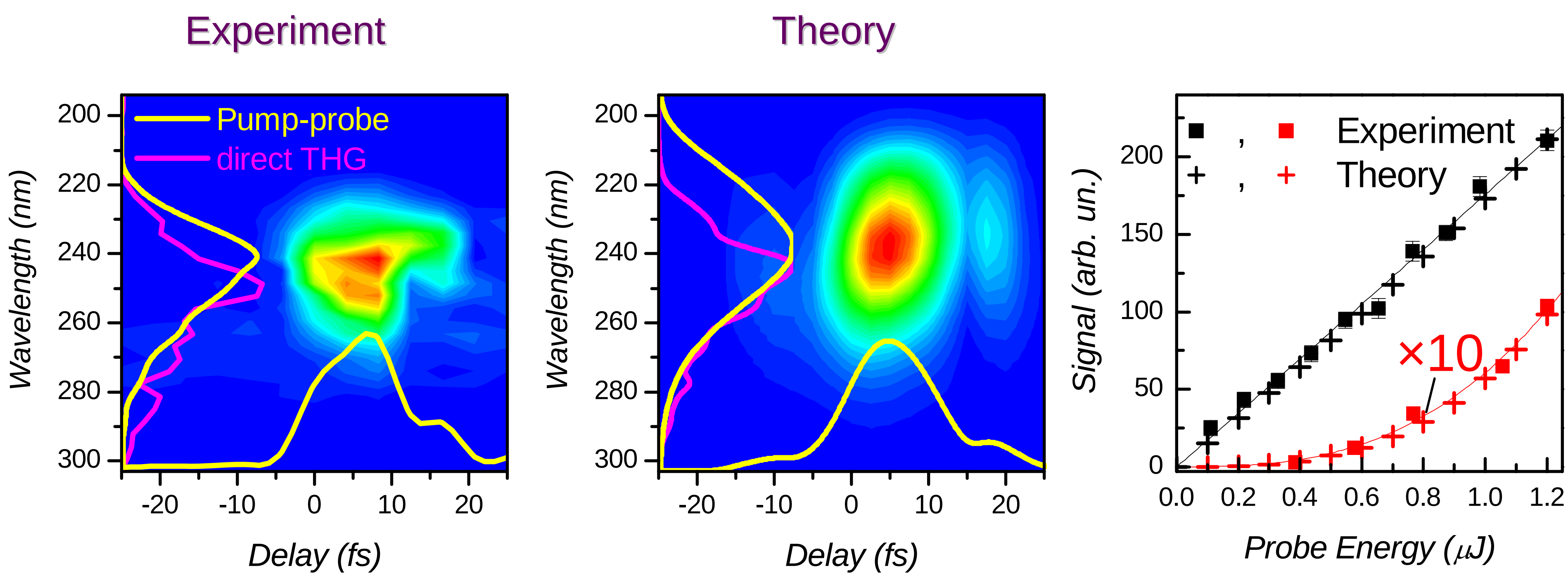
Temporal marginal:

H3,H5,H7 maps integrated over spectrum.

All harmonics follow the same time structure.

Linear Dependence on Probe Intensity

3rd Harmonic, 1mm Argon target



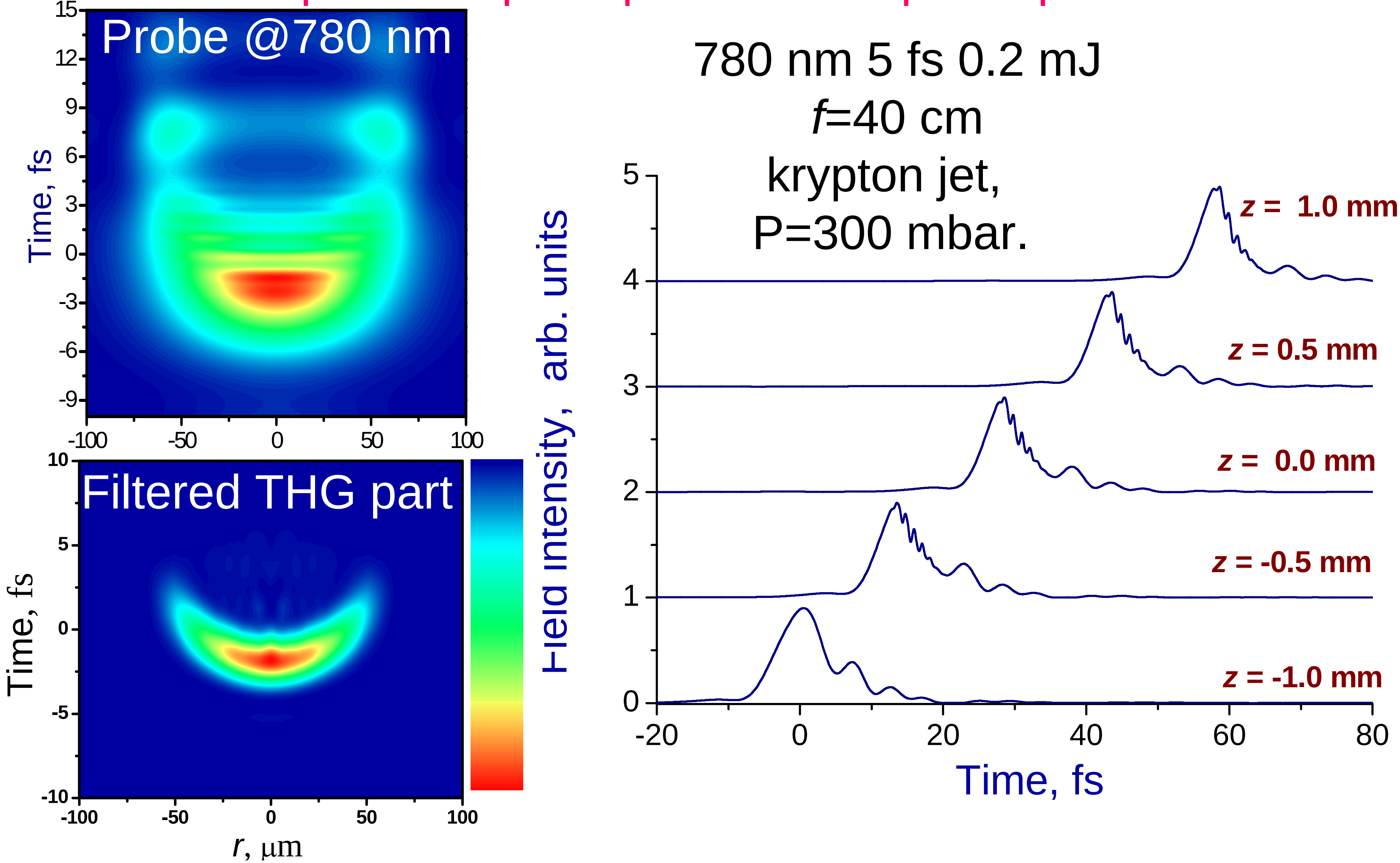
Proof of separation of $\chi^{(3)}$ from Brunel-harmonics:

Linear intensity dependence of H3 on probe intensity!

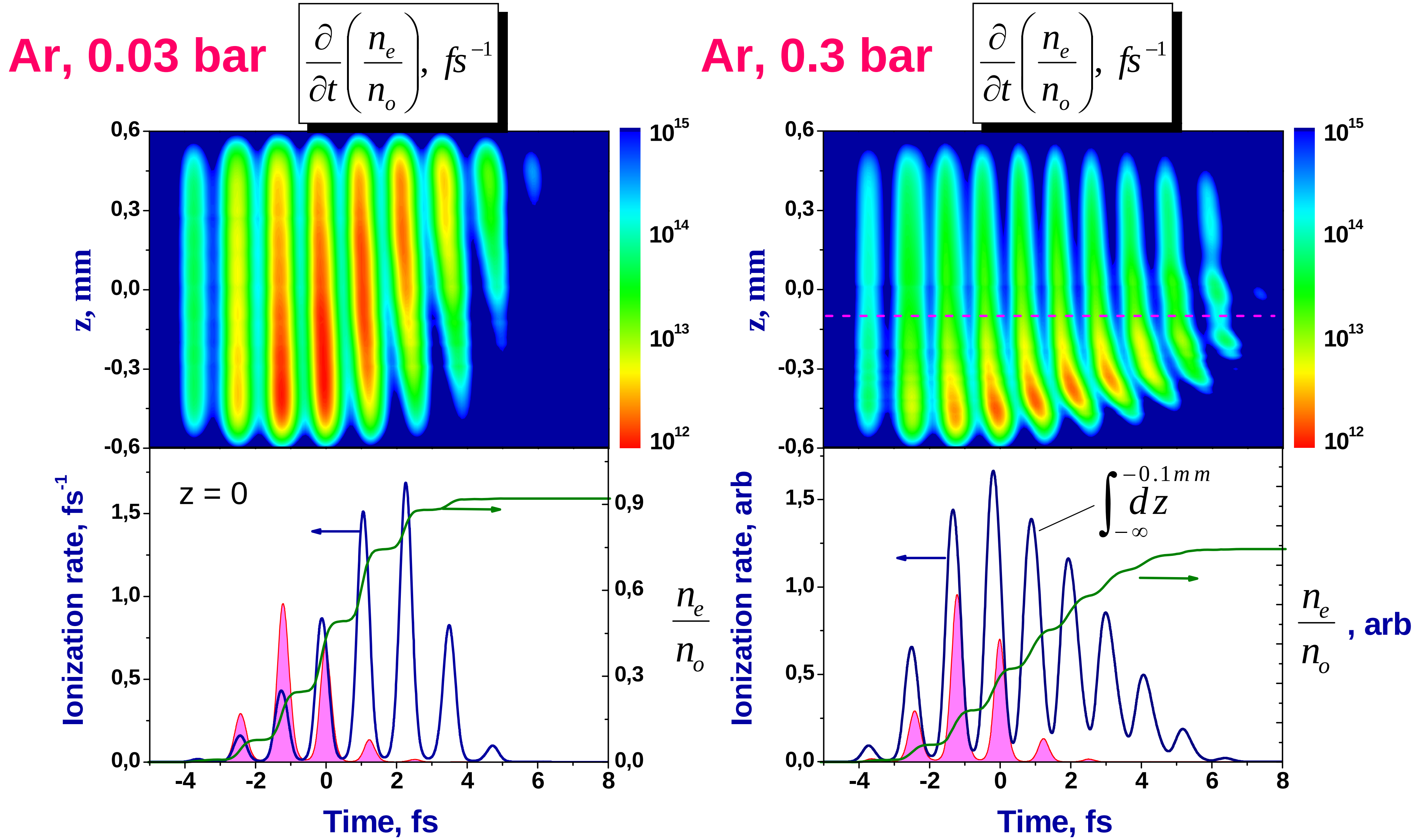
THG spectra measured with pump on are blue-shifted!

Propagation Effects

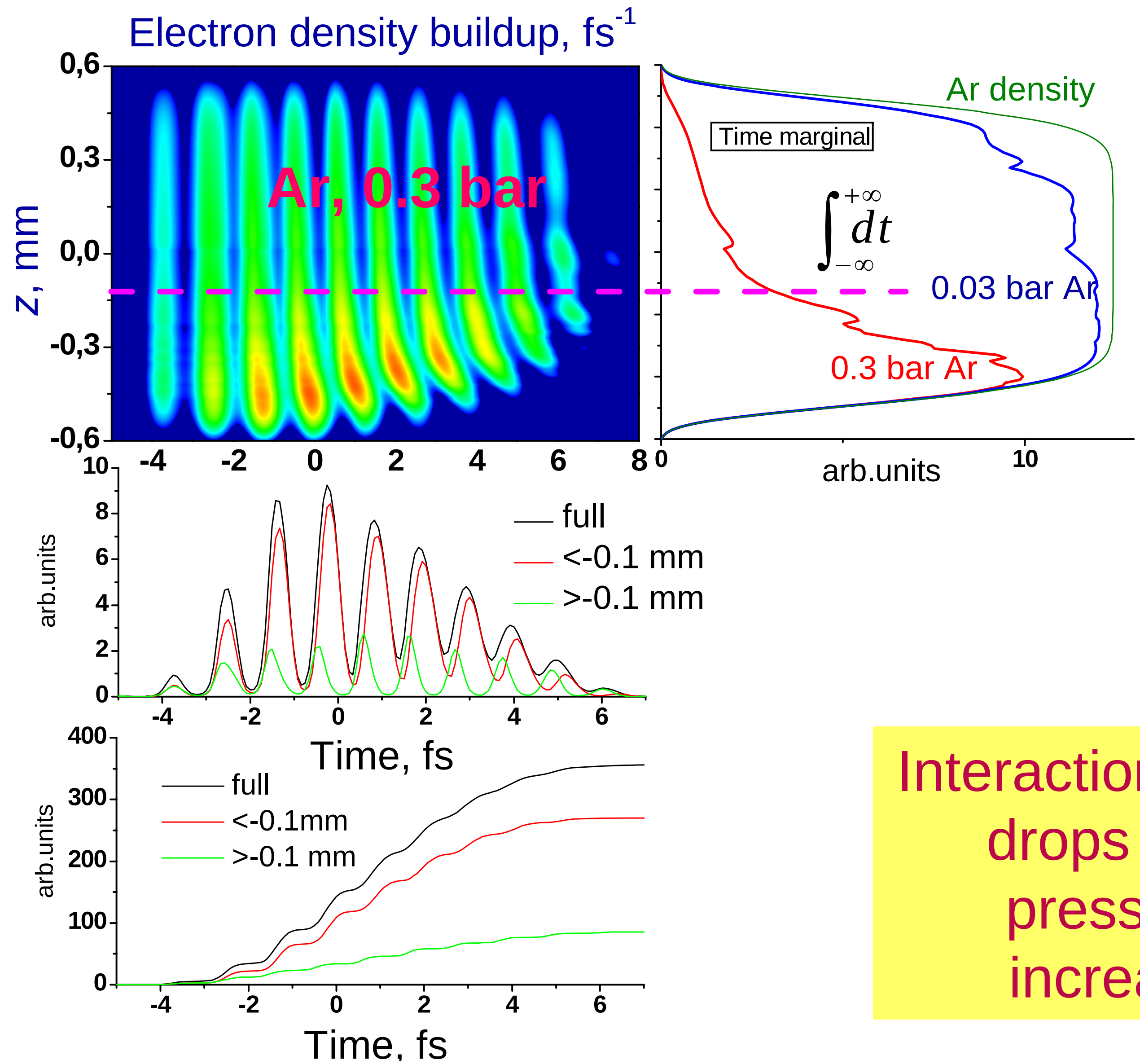
Spatio-temporal profile of the probe pulse



Distortion by Propagation



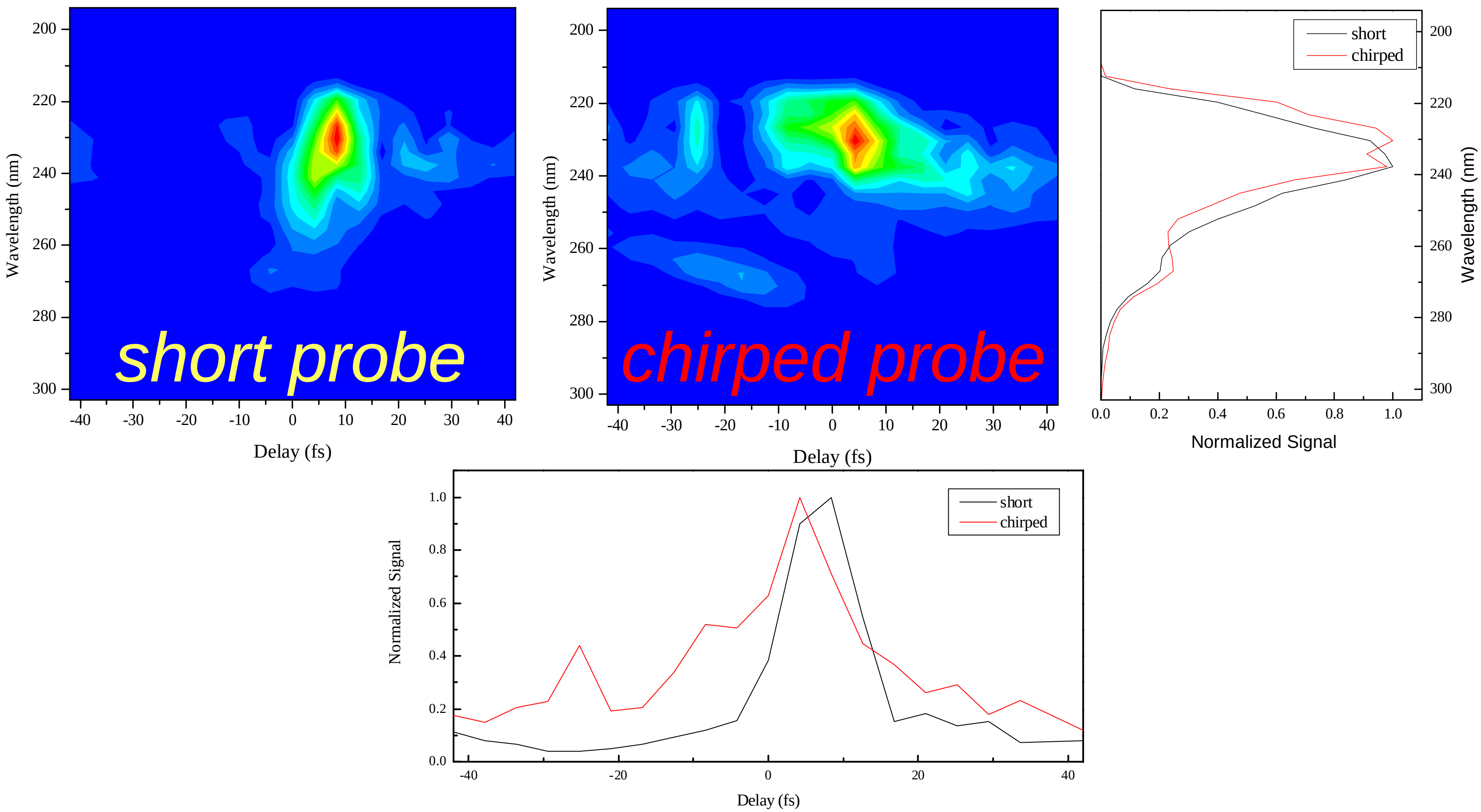
Interaction Length *vs.* Pressure



Interaction length drops with pressure increase

Chirp of Probe Pulse

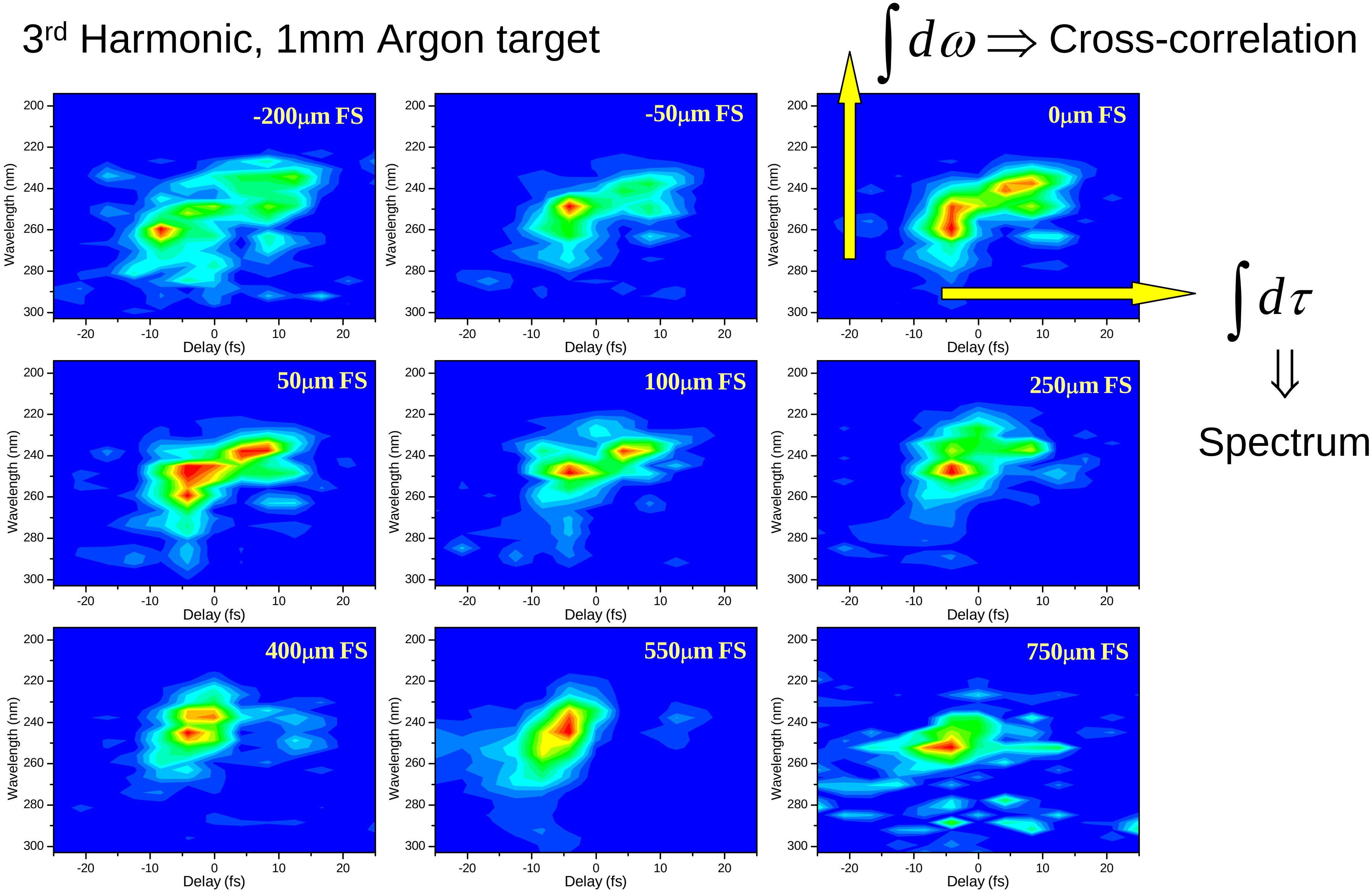
3^{rd} Harmonic, 1mm Argon target



The spectral marginal does not depend on the chirp of the probe pulse.

Coherent Control with Pump Pulse

3^{rd} Harmonic, 1mm Argon target

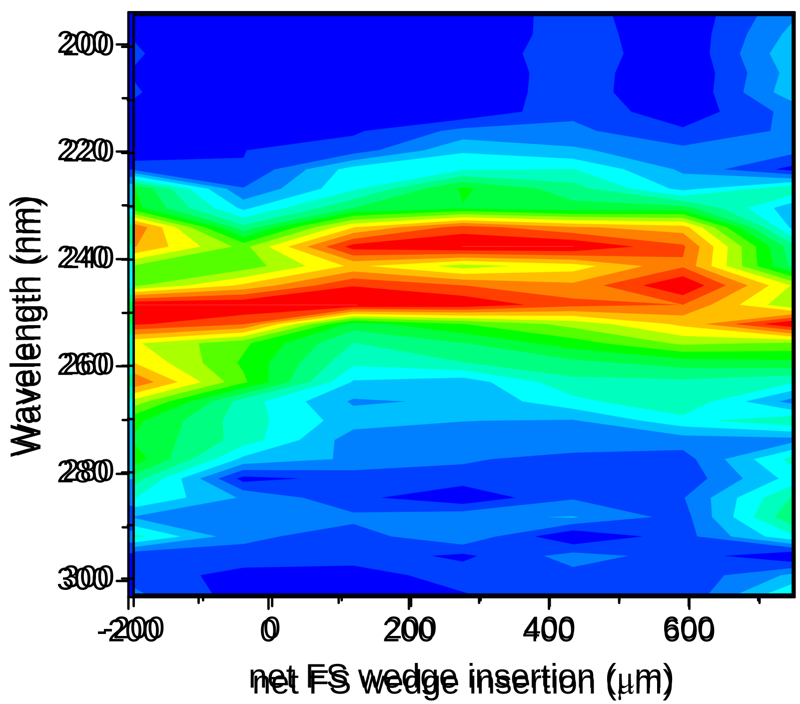


Chirp of Pump Pulse

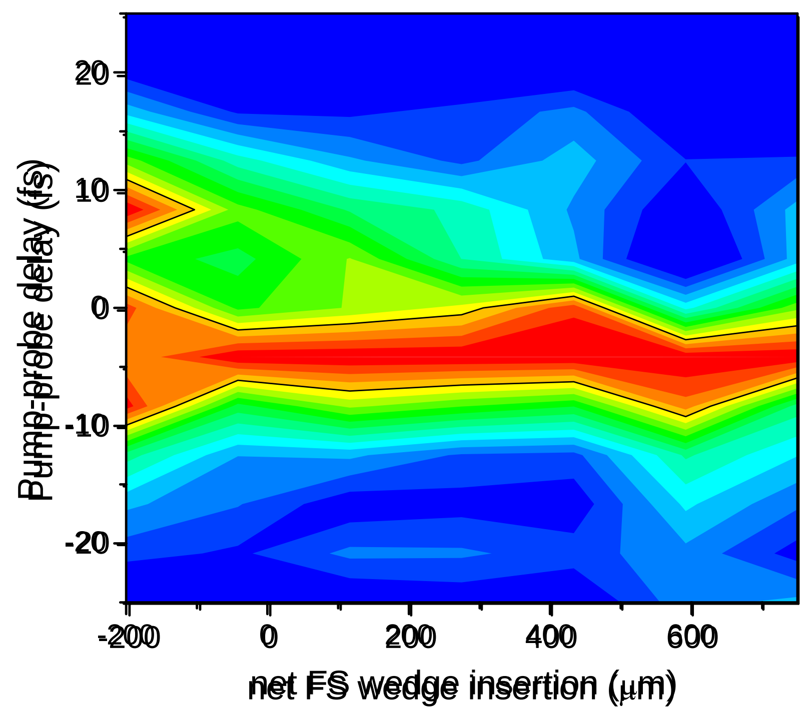
3rd Harmonic, 1mm Argon target

Each marginal is normalized to its own maxima

spectral marginal vs. pump chirp



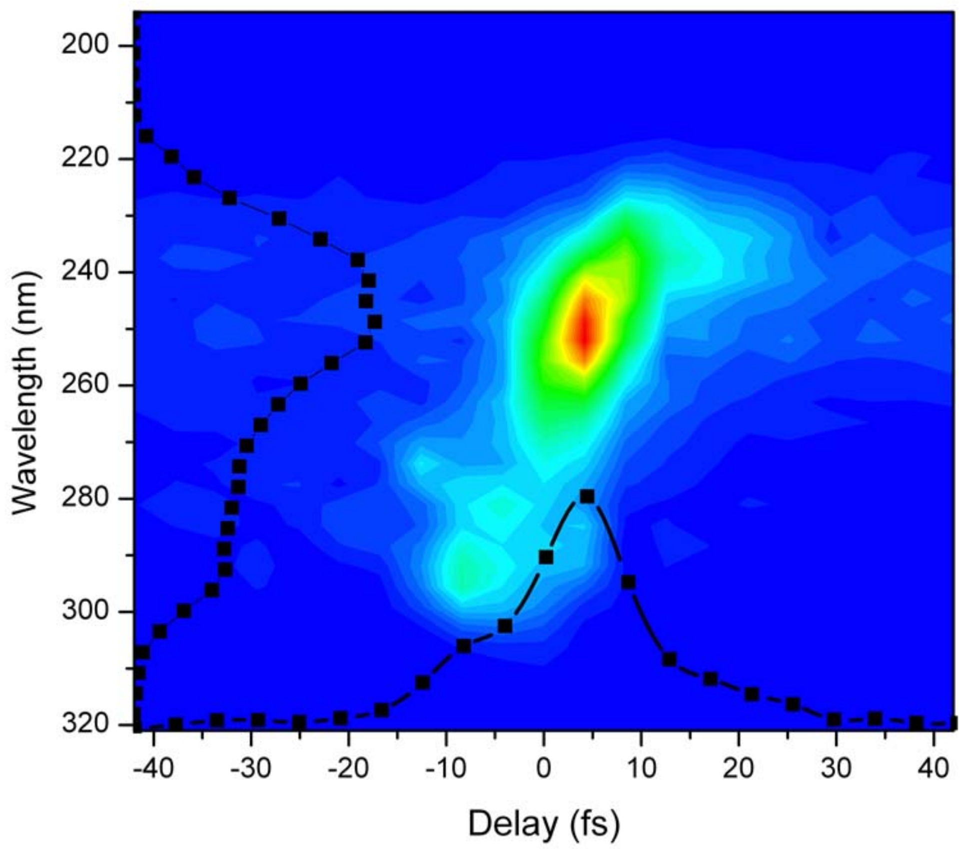
cross correlation vs. pump chirp



Spectrum broadens even for very little chirp

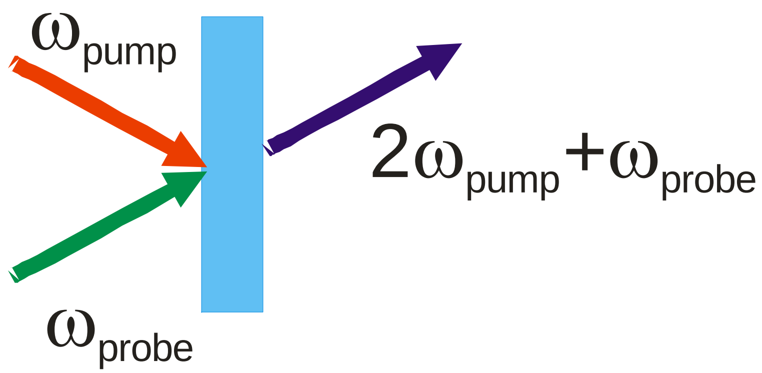
the signal decreases fast with increasing pump chirp

Investigation of TI in bulk solids



Experiment in bulk:

Brunel type harmonic in glass



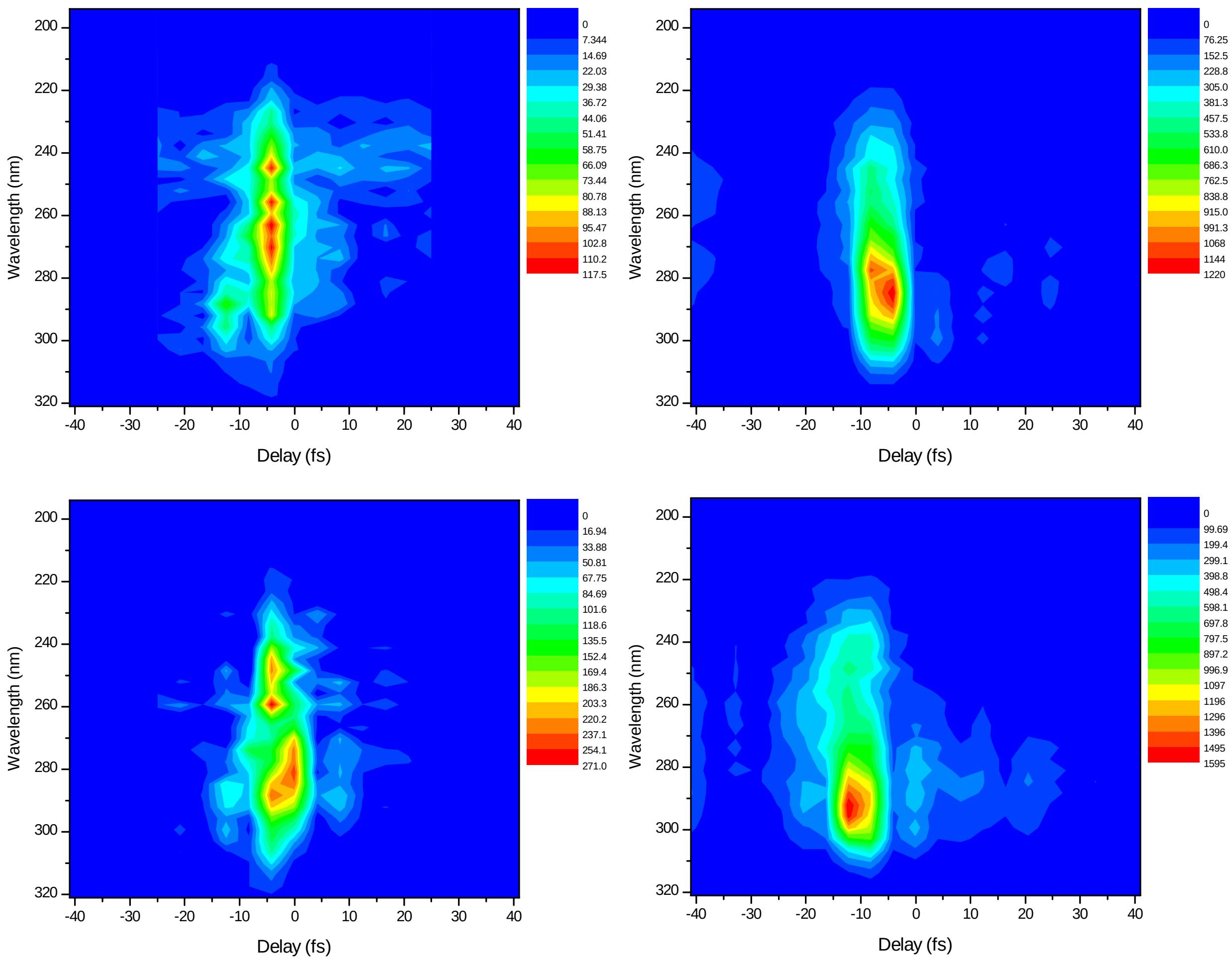
For measuring several harmonics:

Use an OPA system at 1.5 μm

Are We There Yet?

Linear Polarized Pump

Circular Polarized Pump

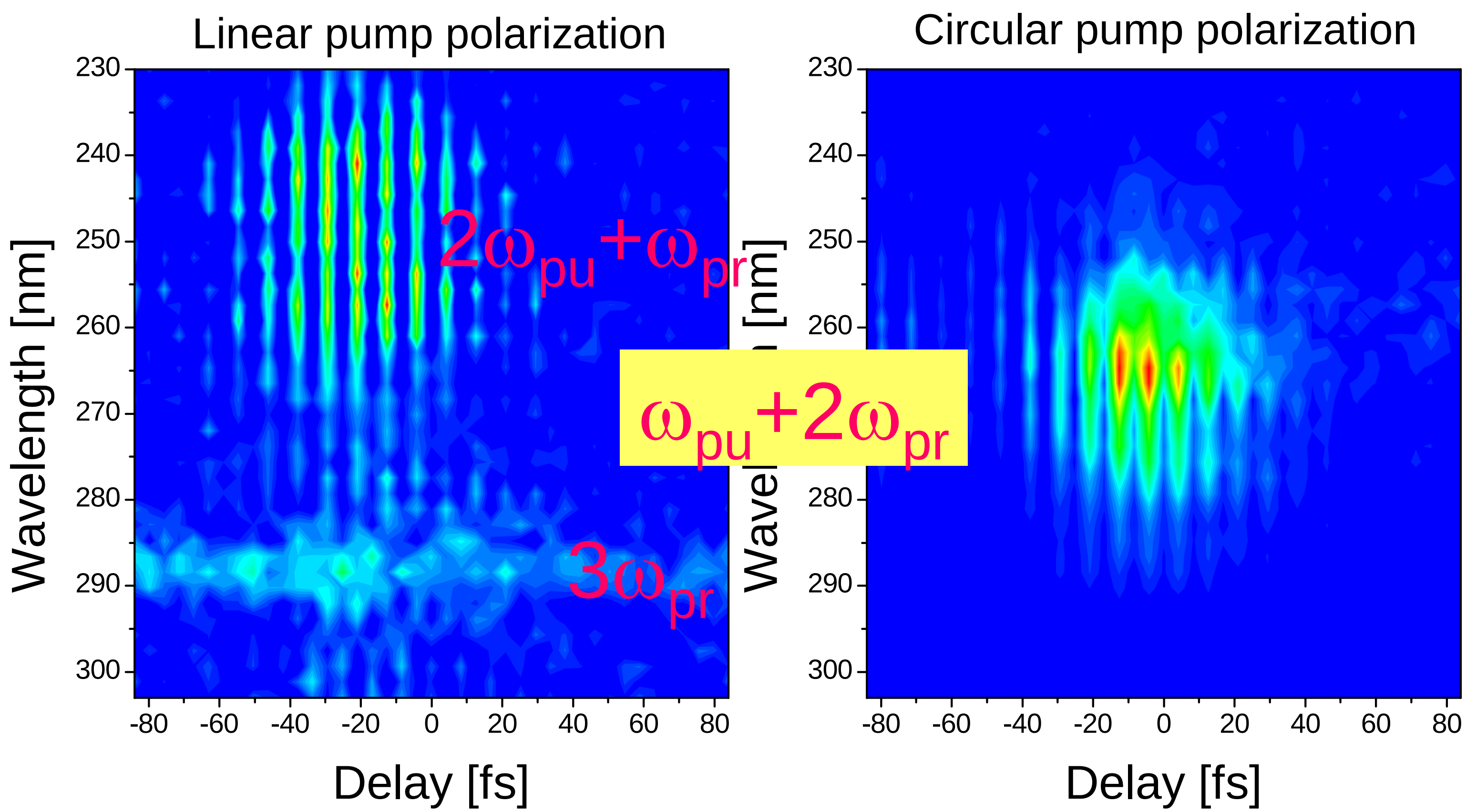


Low Probe Power/Intensity

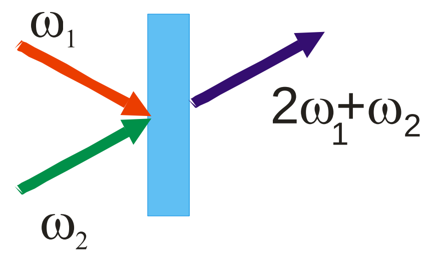
High Probe Power/Intensity
direct probe THG (background)

Target: fused silica
Q: Why is there a signal with circular polarization?

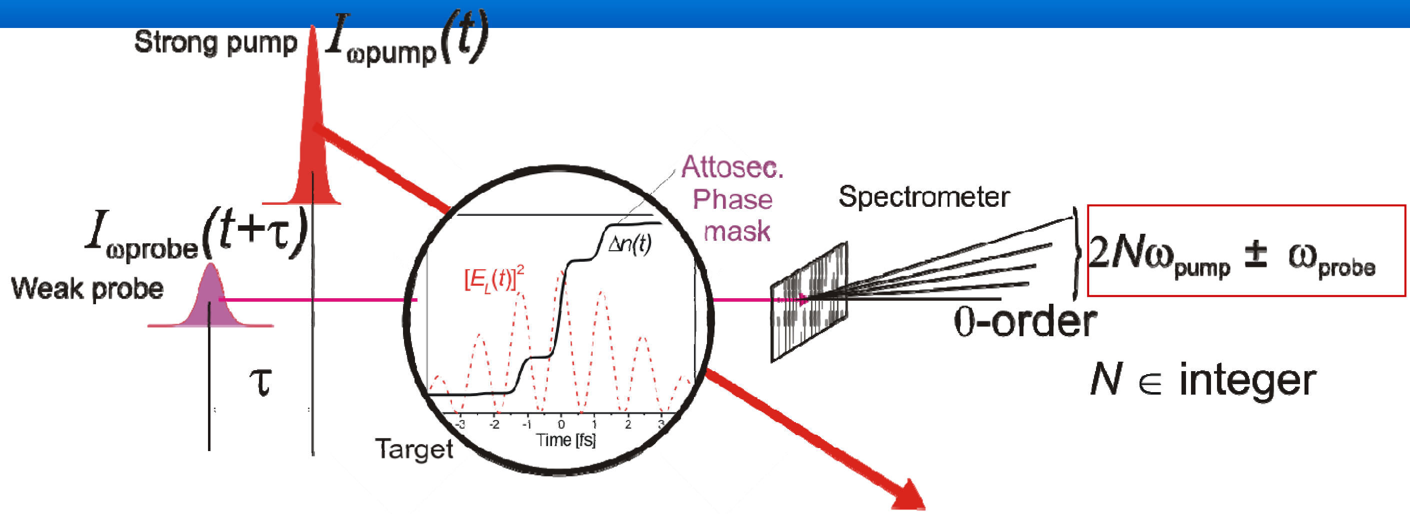
Two Color Experiment



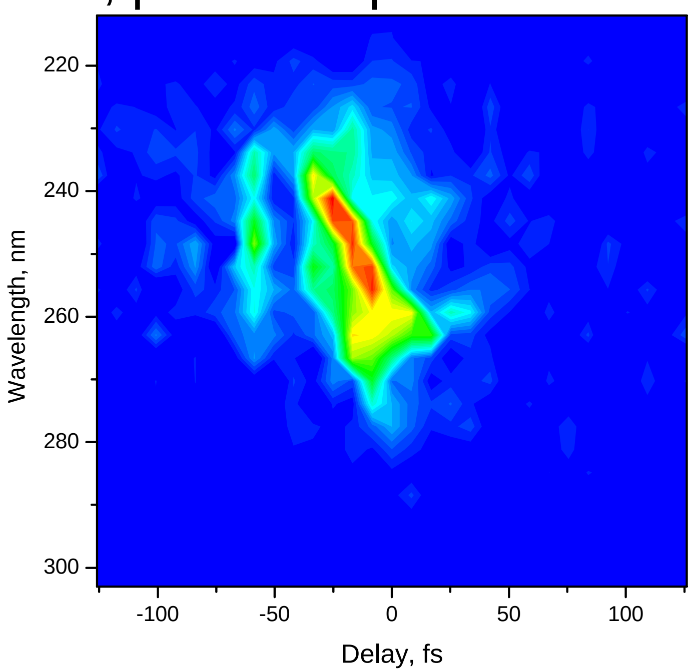
$$\omega_{pump} \neq \omega_{probe}$$



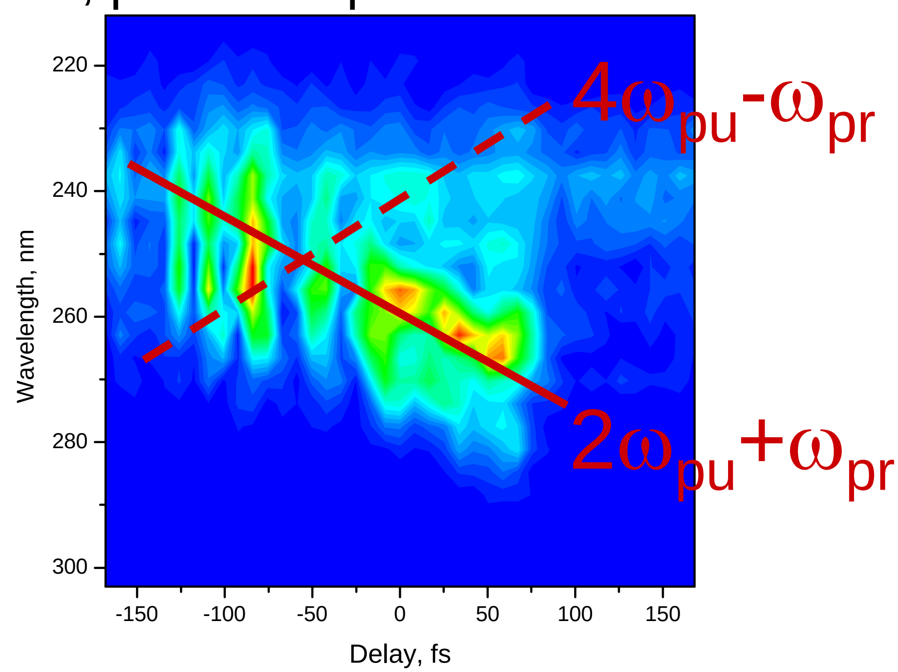
Looking for Beat Modes



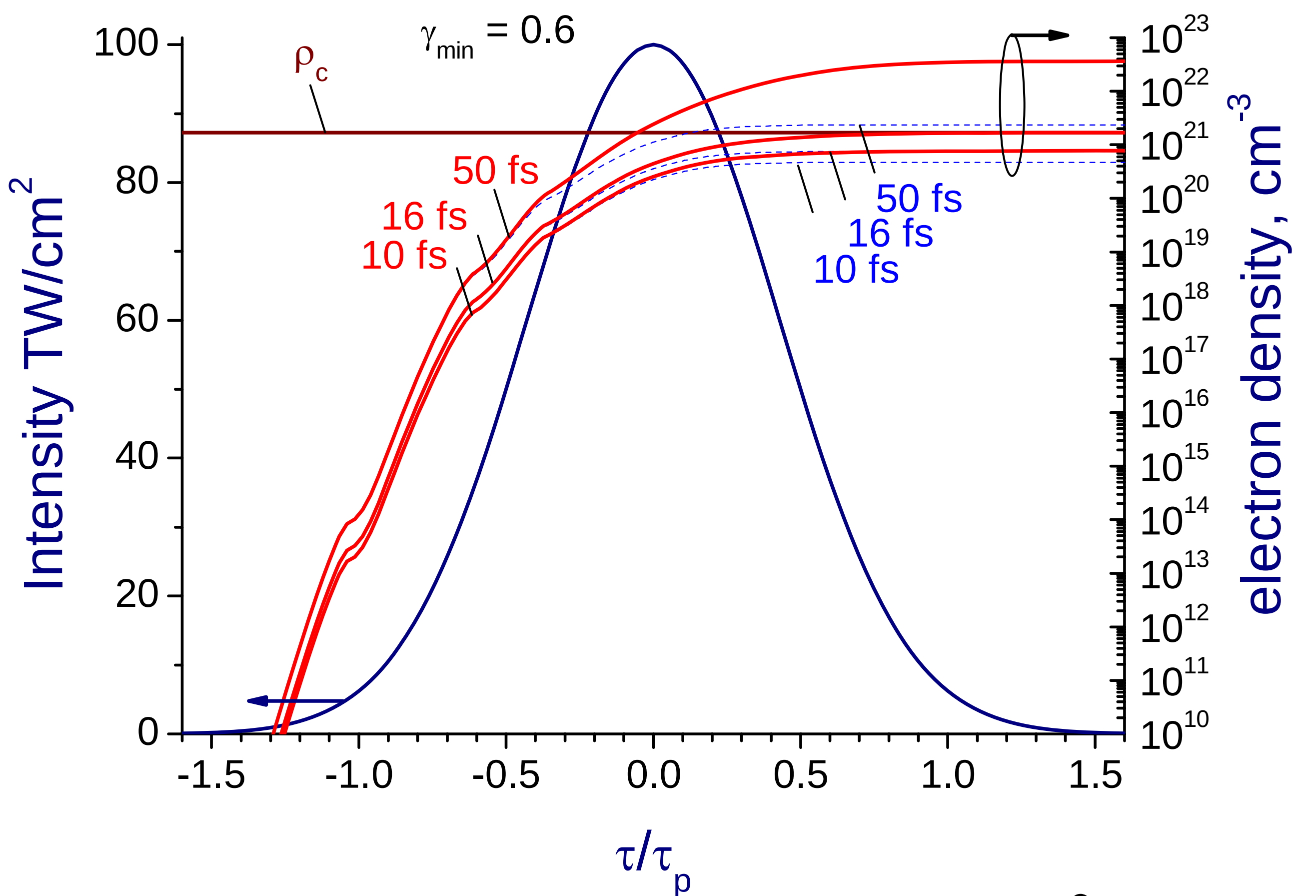
THG, probe chirped with 3 mm of FS



THG, probe chirped with 6 mm of FS



Ionization in Quartz

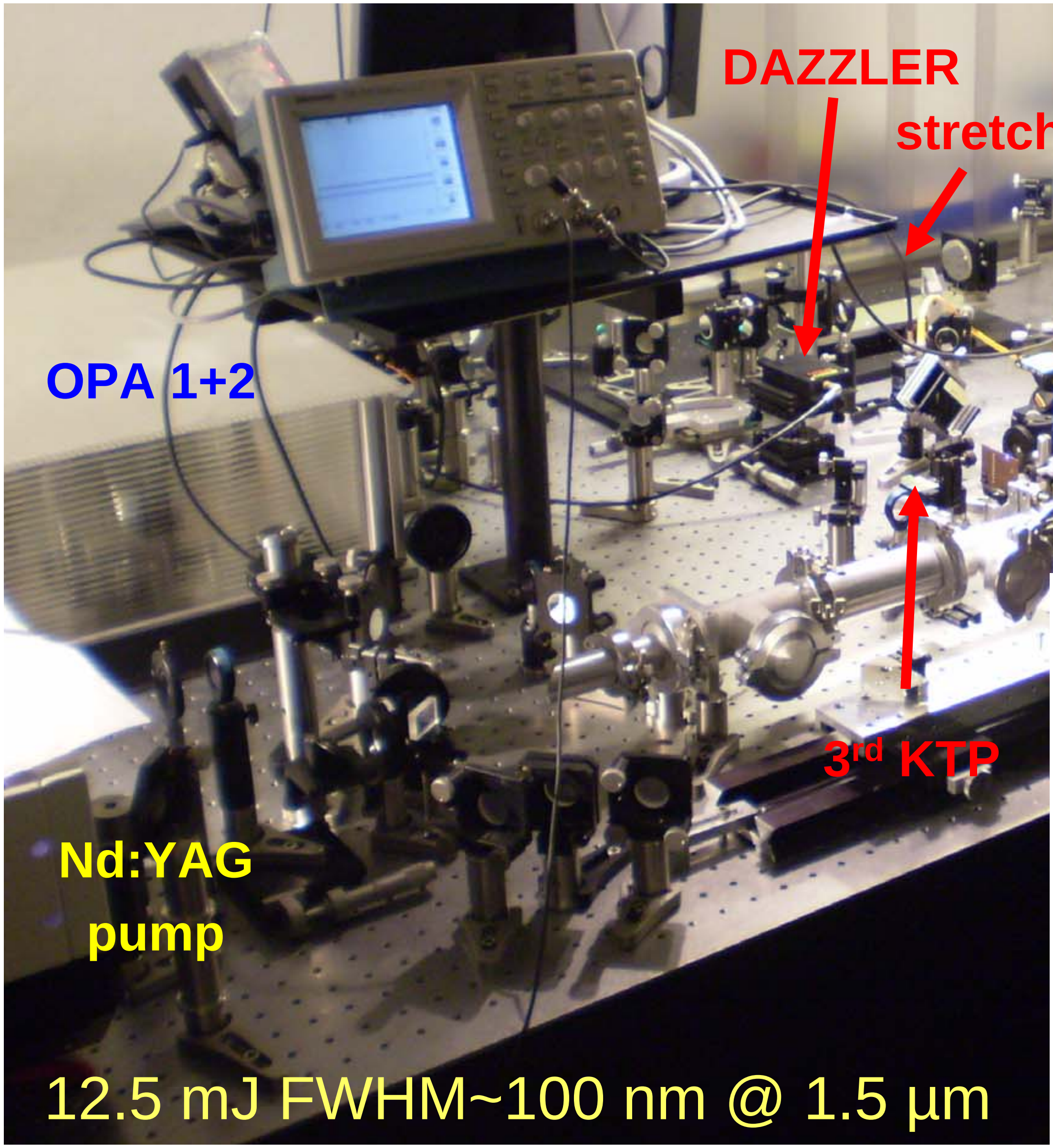


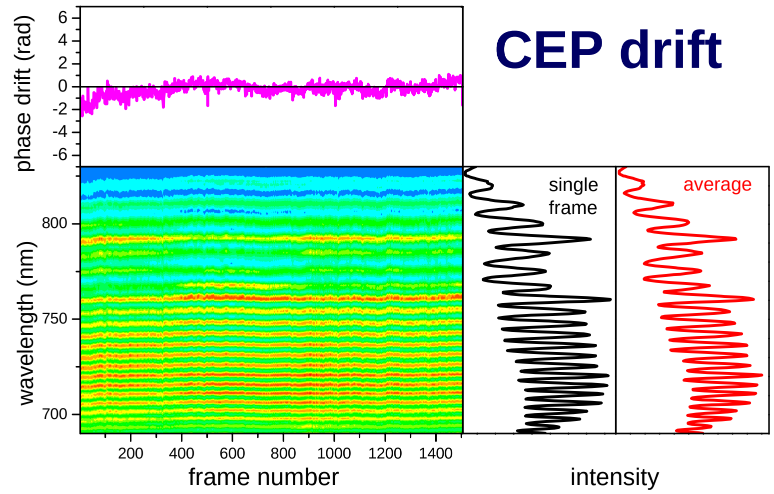
$$\frac{\partial \rho}{\partial t} = W_{PI}\left(|\mathcal{E}|\right) + \frac{\sigma}{U_i}\rho|\mathcal{E}|^2 - \frac{\rho}{\tau_r}$$

Red curves: with avalanche ionization
Dotted blue: without

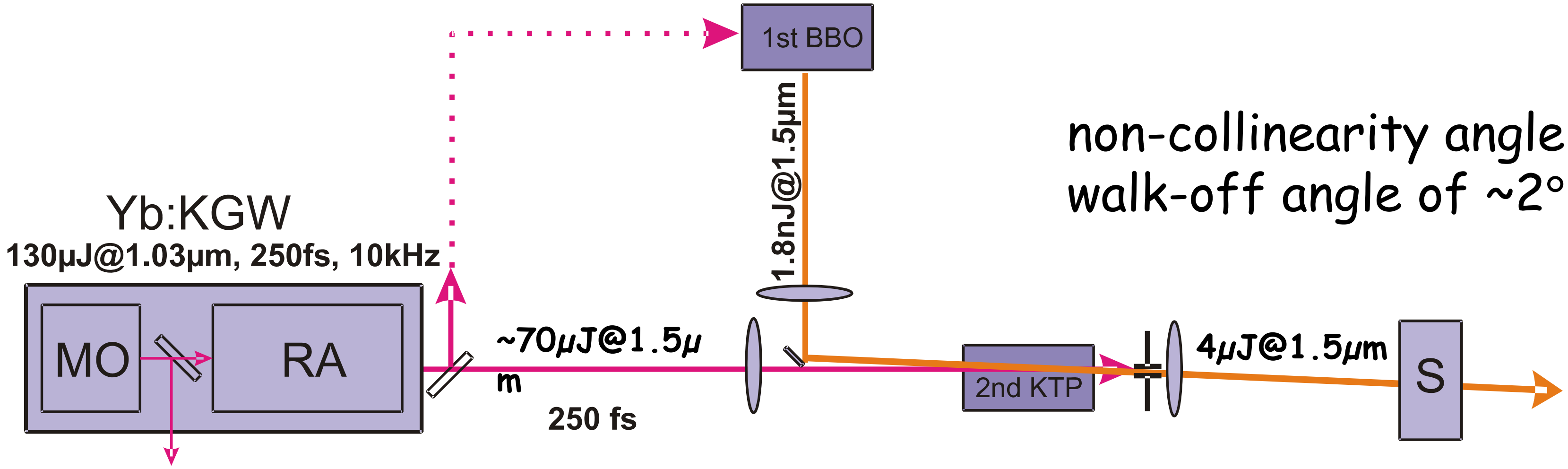
CEP stable 20-Hz IR OPCPA

O.D. Mücke, A. Alisauskas, D. Sidorov
A. Pugzlys, A. Verhoef

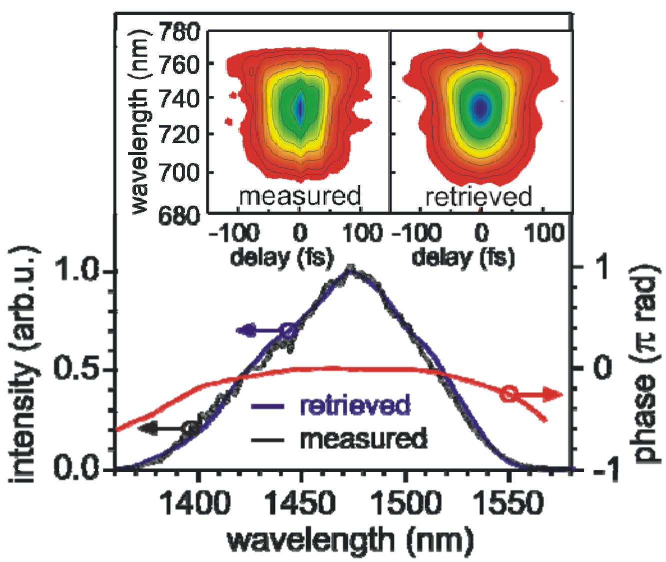


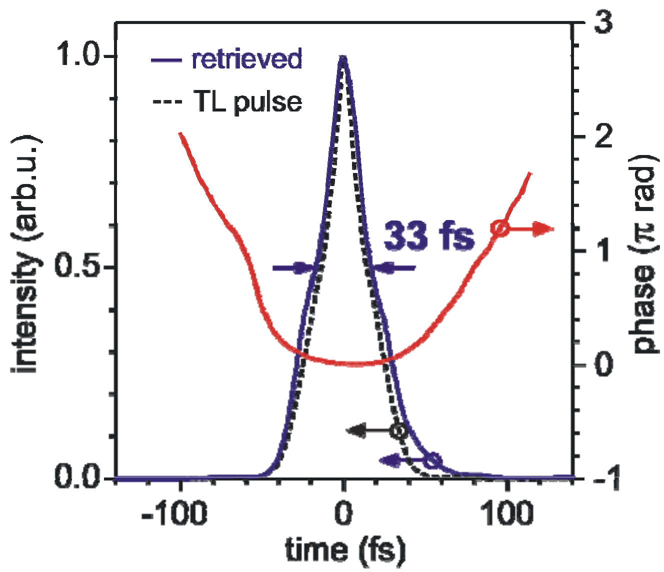


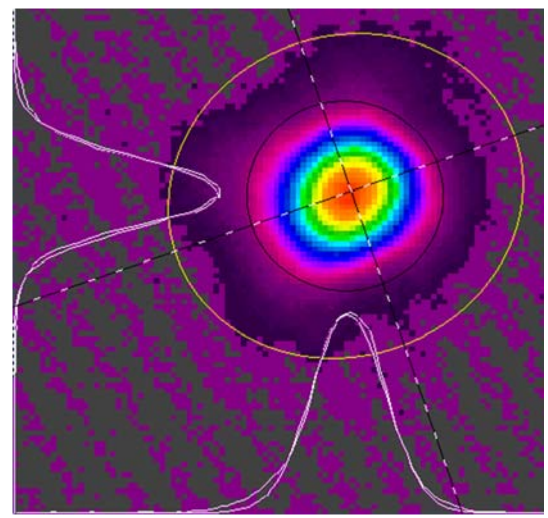
2^{nd} OPA Stage (10 kHz)



non-collinearity angle:
walk-off angle of ~2°



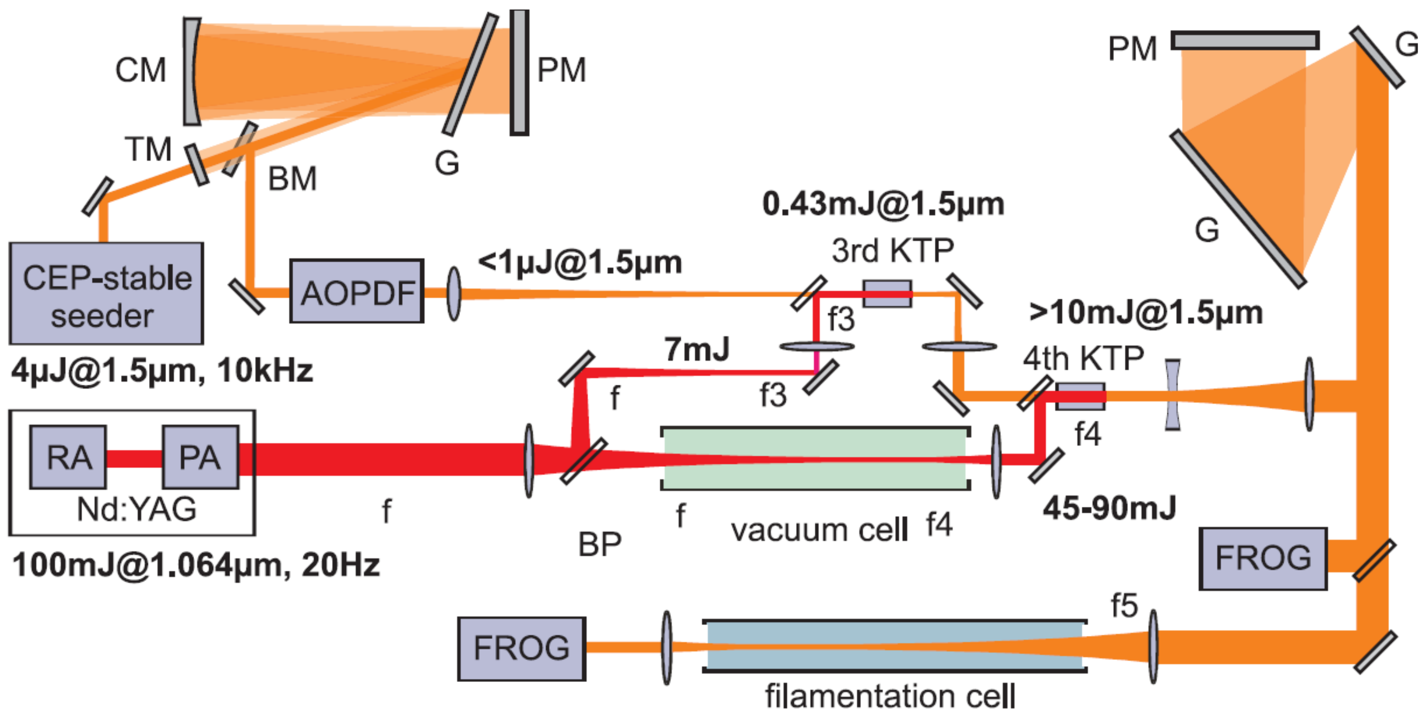




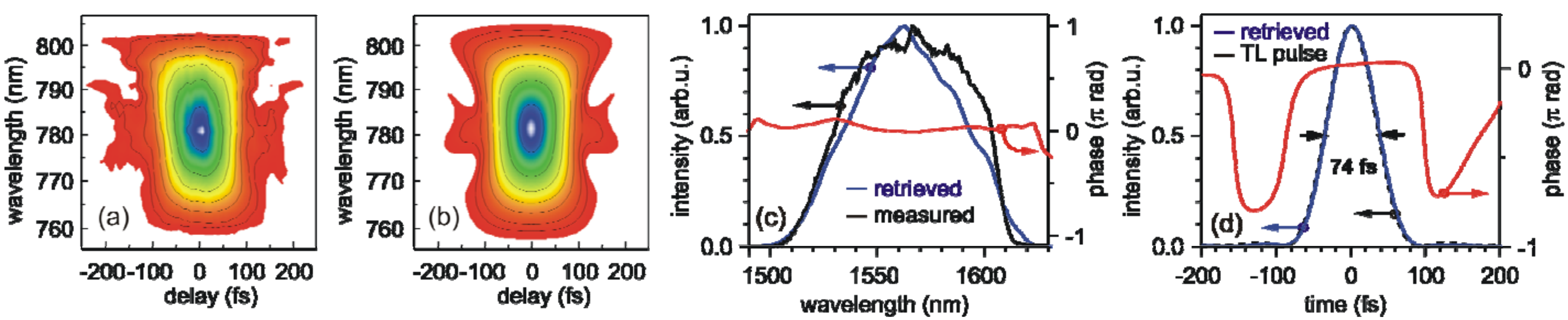
$M^2 = 1.13 \pm 0.04$ (2^{nd}-stage signal)

$M^2 < 1.2$ (Yb:KGW pump)

IR OPCPA (20 Hz)

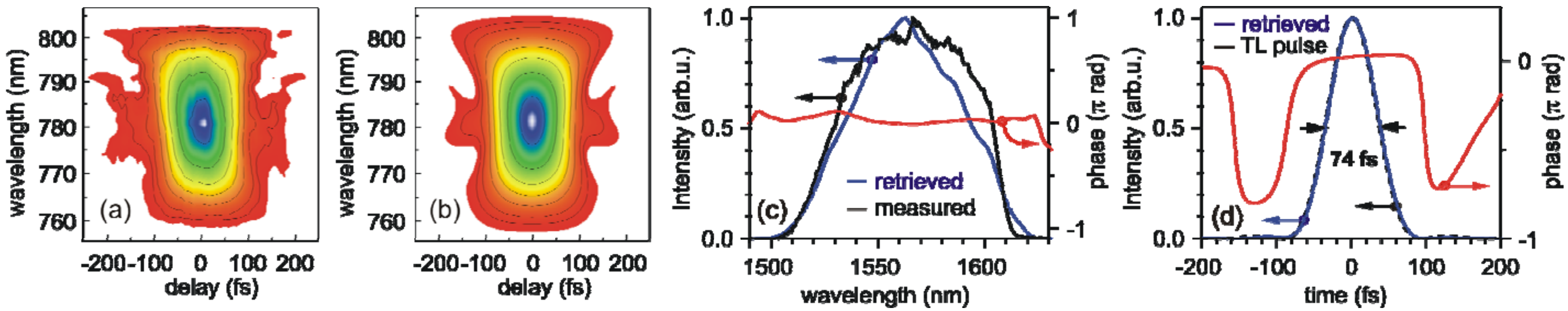


Pulses after the 4th stage

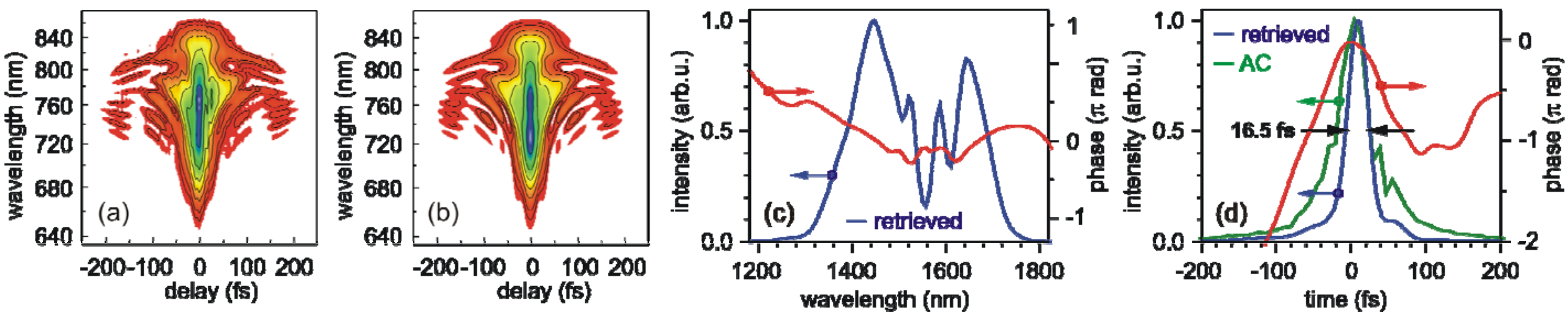


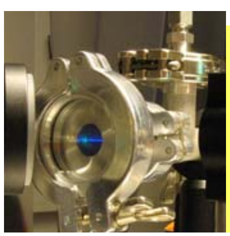
4-Fold Self-Compression of mJ IR Pulses

Pulses after the 4^{th} stage

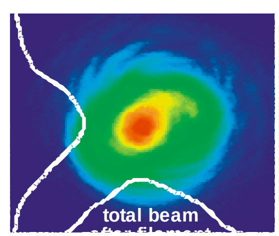


Self-compressed 1.5-µm pulses: >1.5 mJ, 3 optical cycles





Gas: Ar, 5 bar,
Cell length: 140 cm
Filament length: 12-15 cm



Optimal compression:
E_{in}: 2.2 mJ, E_{out}=1.5 mJ
Throughput: 66%

Summary

- **First direct experimental observation of Brunel harmonics in gas and bulk.**
- **Attosecond ionization dynamics can be mapped onto a spectral response that is free of recollision contribution.**
- **Attosecond phase mask is not intuitive but quite robust.**
- **It is feasible to develop an optical technique instead of registering photo-ionization fragments ⇒ <u>attoscience in bulk</u>.**
- **Future experiments on bulk: use the CEP 1.5(signal)/3.5(idler) µm OPCPA.**

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The Group

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