Attosecond technology - quantum control of high harmonic generation for phase matching

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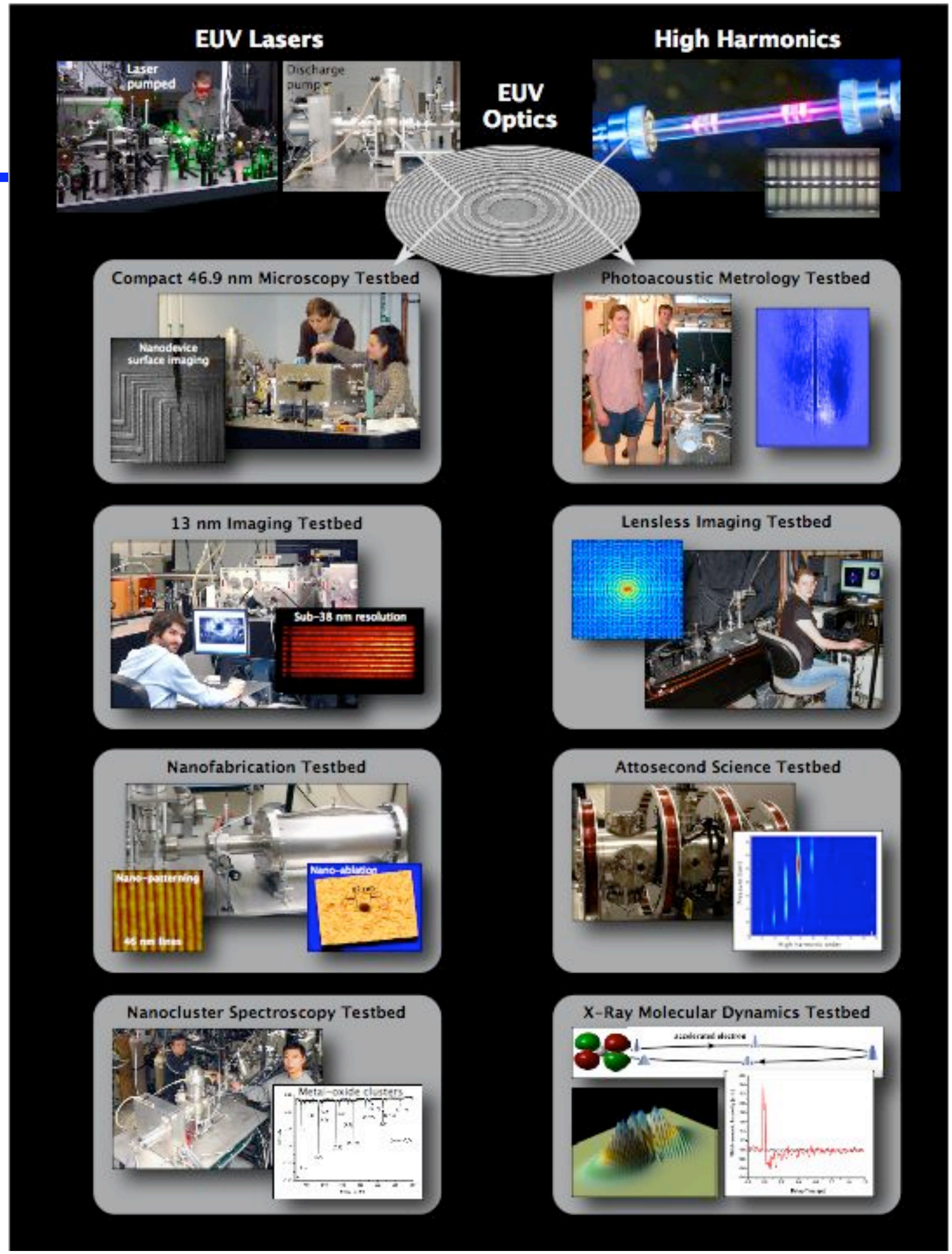
EUV

An NSF Engineering Research Center

Objective:

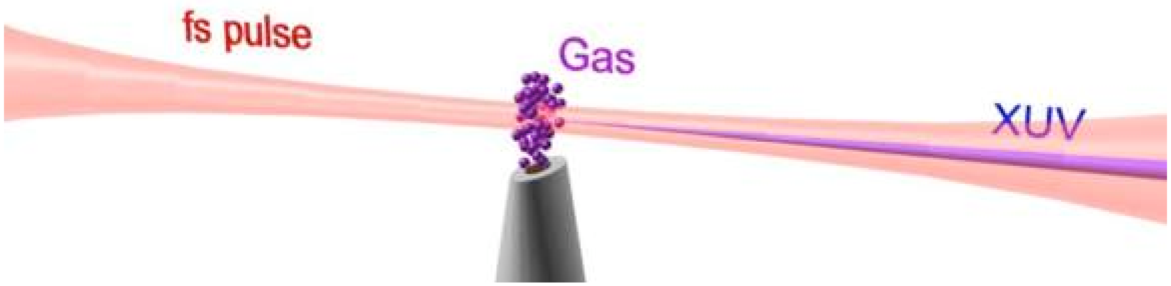
- Many critical questions important to scientific and technological progress can be addressed using ultrafast coherent short wavelength sources
  - Nanoscale electron dynamics (e.g. heat transport)
  - Nano imaging
  - Control and manipulation of atoms and electrons in molecules

- Barrier to overcome - increasing the flux and wavelength range



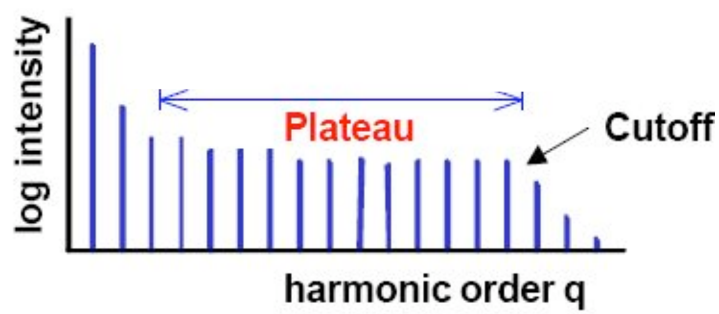
Coherent x-ray generation using HHG

- *Coherent* EUV is generated by focusing an intense laser into a gas



- Origin of HHG work: 3HG, 5HG, FWM work using nanosecond lasers
  - *S.E. Harris et al, J. Reintjes*
  - $P(3\omega) \propto \chi^{(3)} EEE$ *etc.*

- Nonperturbative nature of HHG using ps, fs pulses was a *discovery*
  - L'Huillier
  - Rhodes
  - CO_2 laser HHG



Frequency

The birth of Nonlinear Optics

- P.A. Franken et al, Physical Review Letters 7, p. 118 (1961)

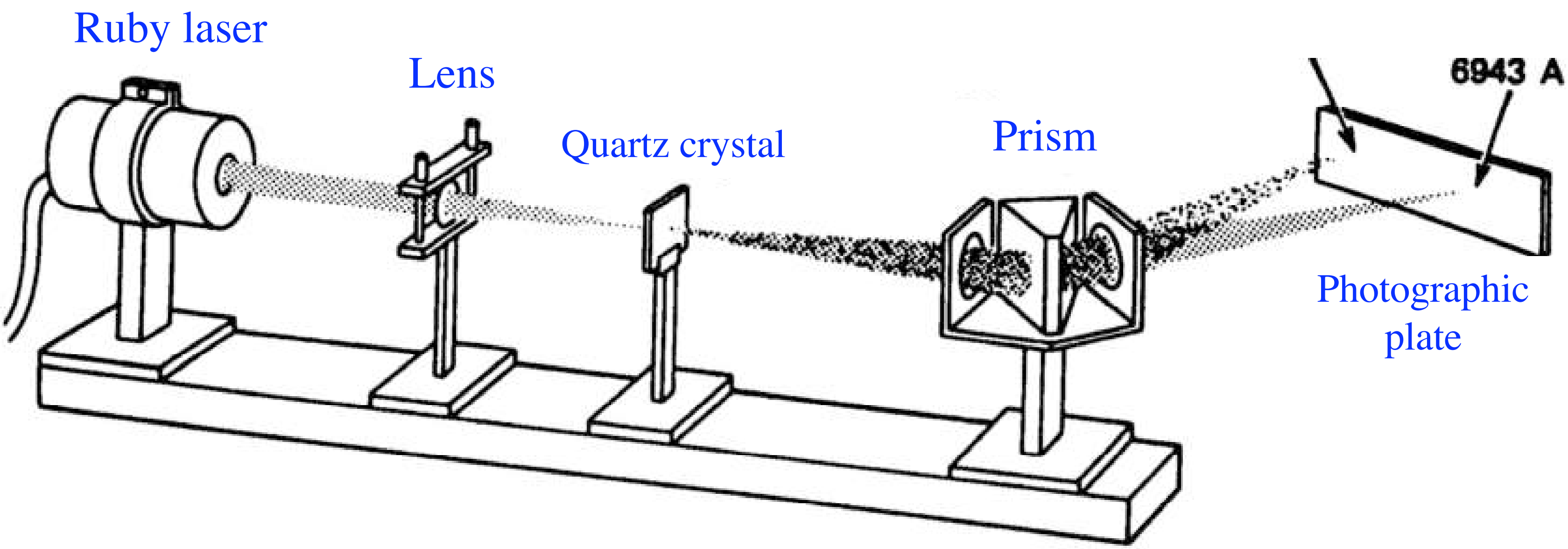
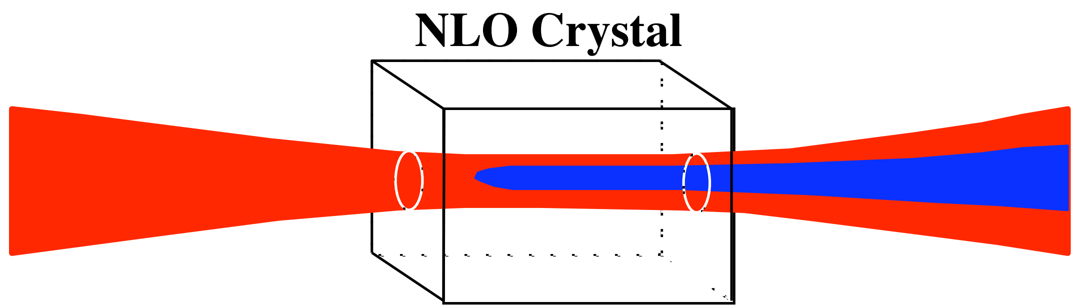
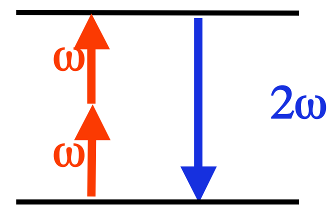


Figure 12.1. Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694\ \mu m$ is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347\ \mu m$. The two beams are then separated by a prism and detected on a photographic plate.

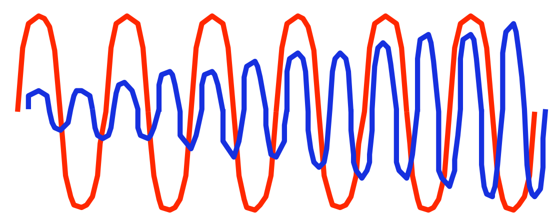
NLO technology has seen widespread use



- Second Harmonic generation

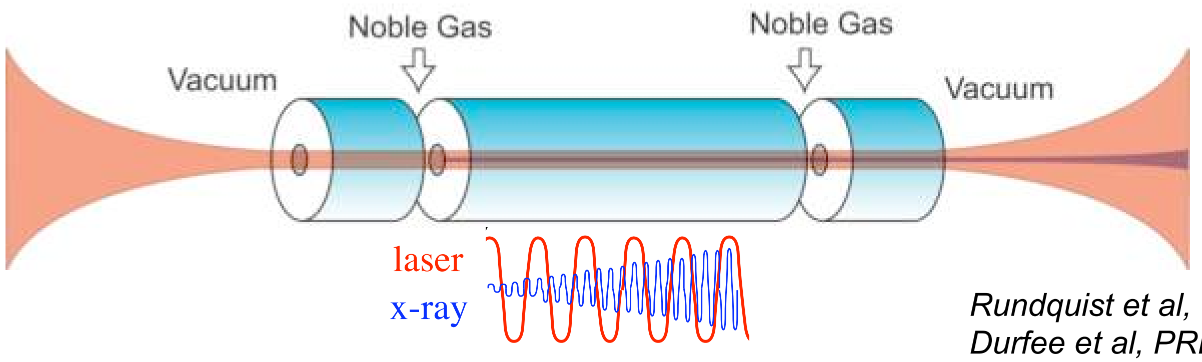


- Need **phase-matching** for good efficiency
  - $v_{phase}(\omega) = v_{phase}(2\omega)$



- Problem in case of HHG: crystal based phase-matching does not apply
- Neither do methods based on resonant dispersion!

Waveguides for high harmonic generation



Rundquist et al, Science ***280****, 1412, 1998*
Durfee et al, PRL ***83****, 2187, 1999*

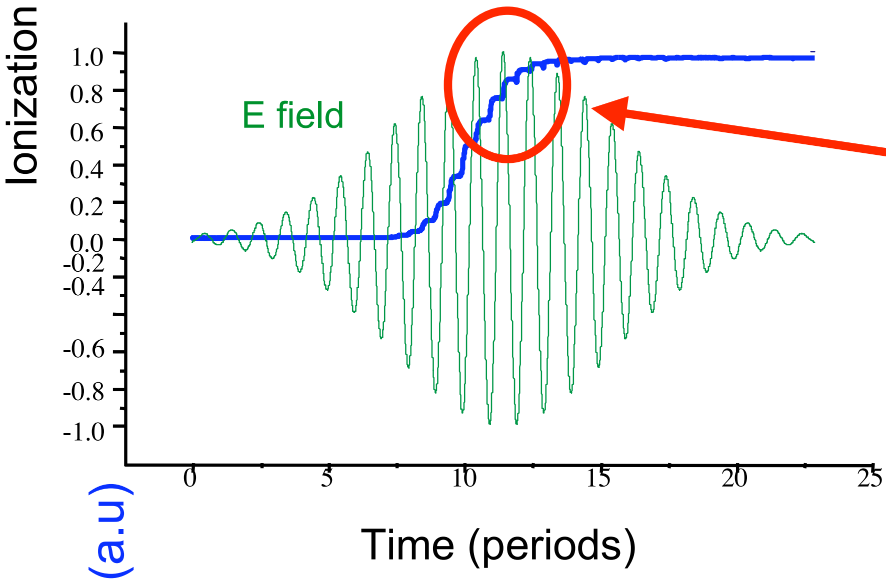
$$\Delta k = qk_f - k_q = 0$$

$$\Delta k = q\left\{\left(\frac{u_{11}^2 \lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

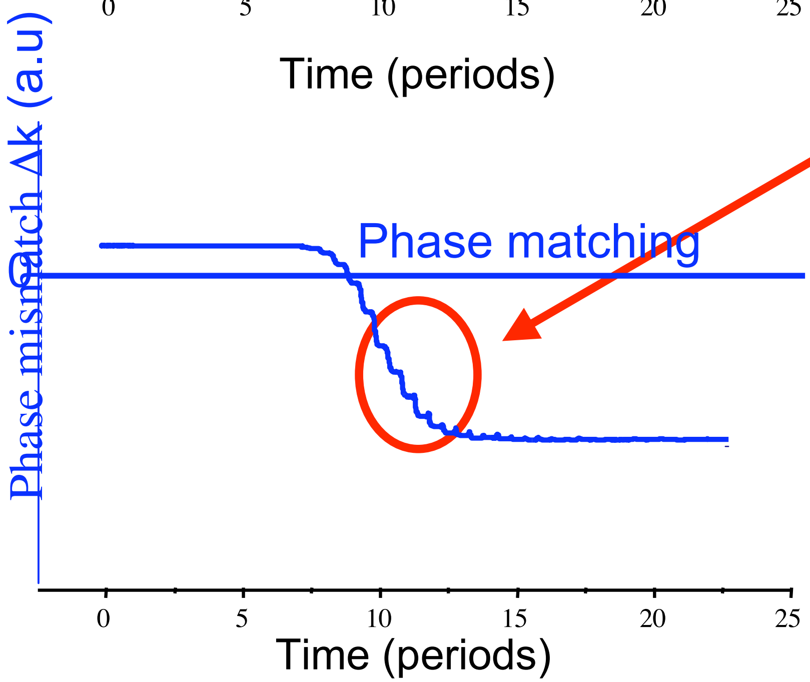
Waveguide **Neutrals** **Plasma**

- Use structure *surrounding* NLO medium to control phase matching!

Limitation: HHG is coupled to ionization



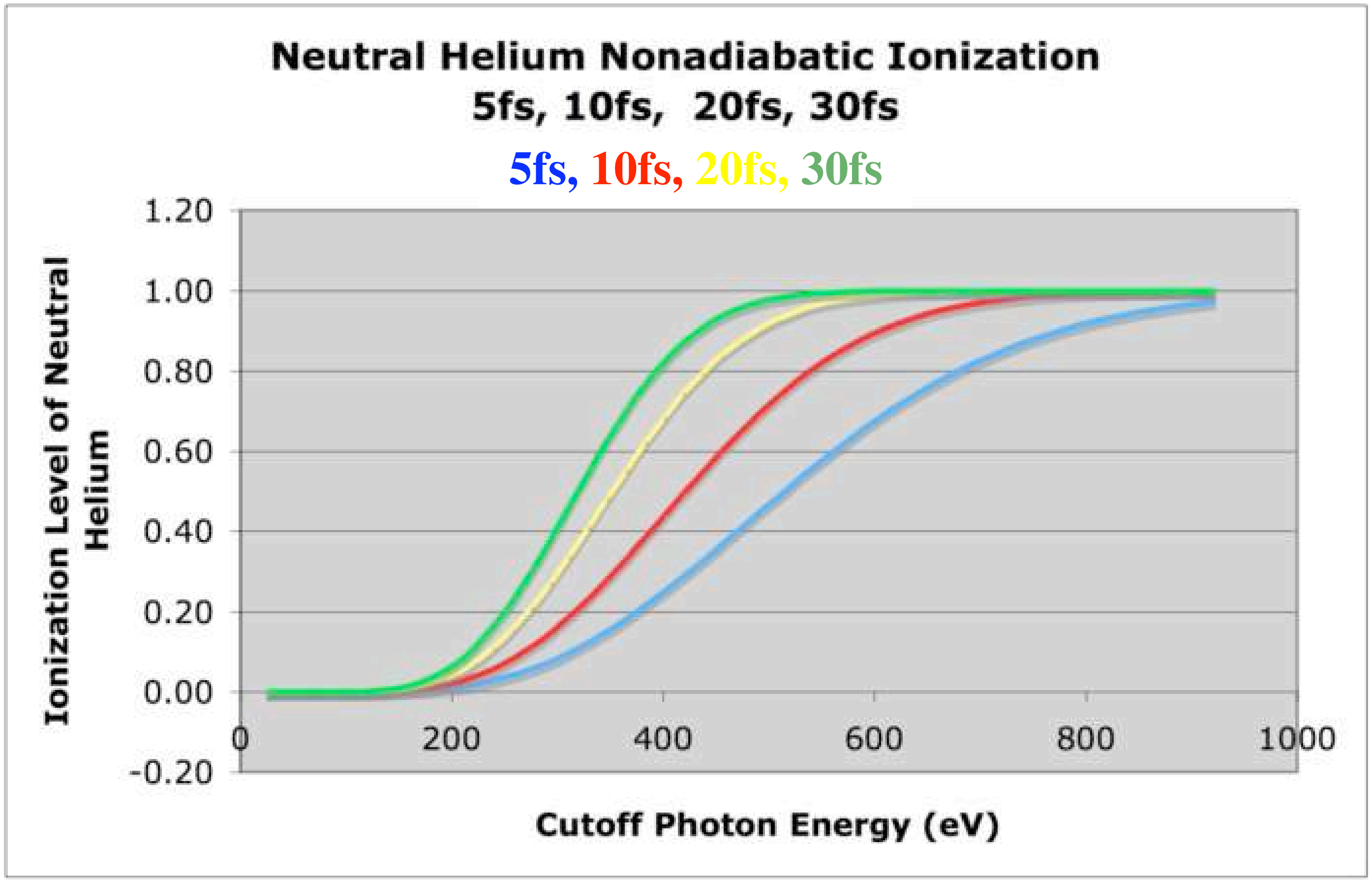
- Since higher harmonics are generated at higher laser intensities and ionization levels, impossible to phase match above $\eta_c \approx 0.5$ - 5% or E < 100eV



$$\Delta k = q\left\{\left(\frac{u_{11}^2 \lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

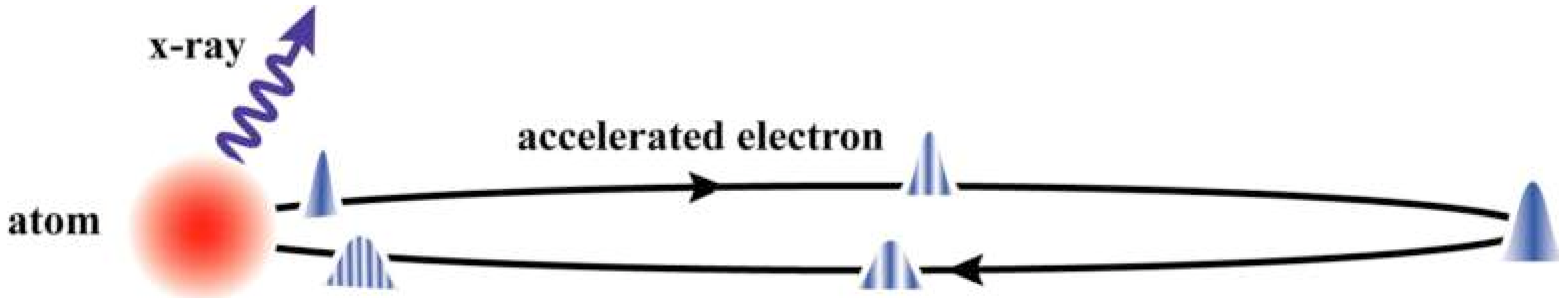
Waveguide **Neutrals** **Plasma**

Beating ionization using shorter pulses



- Impossible to phase match above 150eV
- Need phase corrective technique to compensate for ionization

The quantum phase--



- HHG depends on phase of *recolliding* electron

- A *non-instantaneous*, but still purely electronic, NLO response!!!

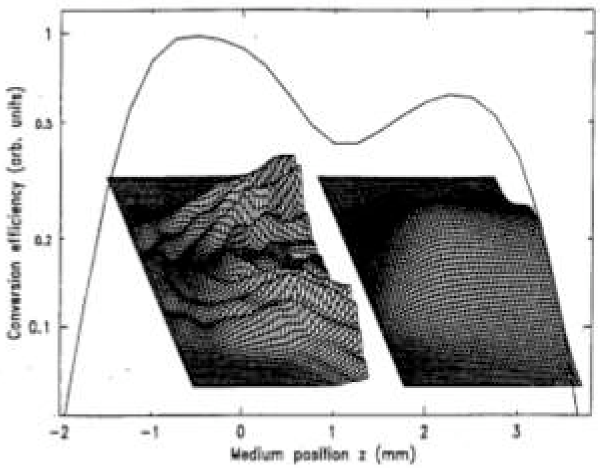
$$\varphi_{EUV} = \frac{2\pi}{h}\int_{t_o}^{t'}\left(\frac{p^2}{2m}+I_p\right)dt$$

$$\approx \frac{2\pi}{h}U_p(t'-t_o) = \frac{2\pi}{h}U_p\tau$$

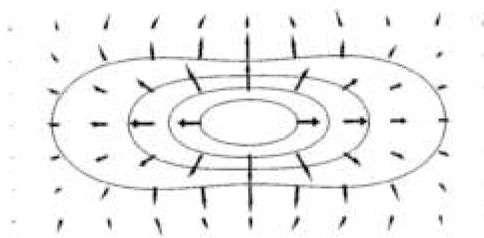
$$\propto \frac{2\pi}{h}I_p\lambda^2\tau$$

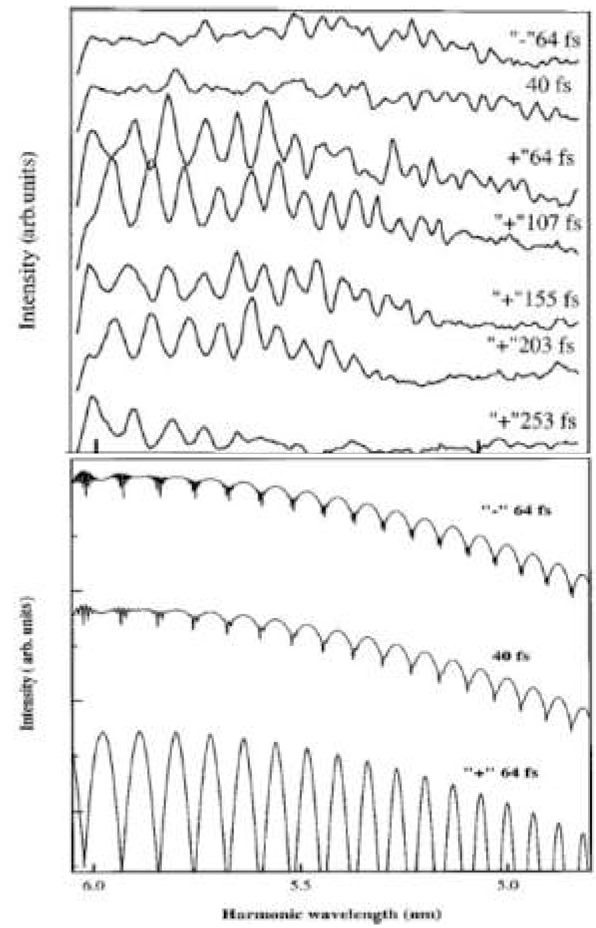
- M. Lewenstein, et al., Physical Review A 49 (3), 2117 (1994).
- Z. Chang et al., Physical Review A 58 (1), R30 (1998).

Experimental evidence of quantum phase



(b)



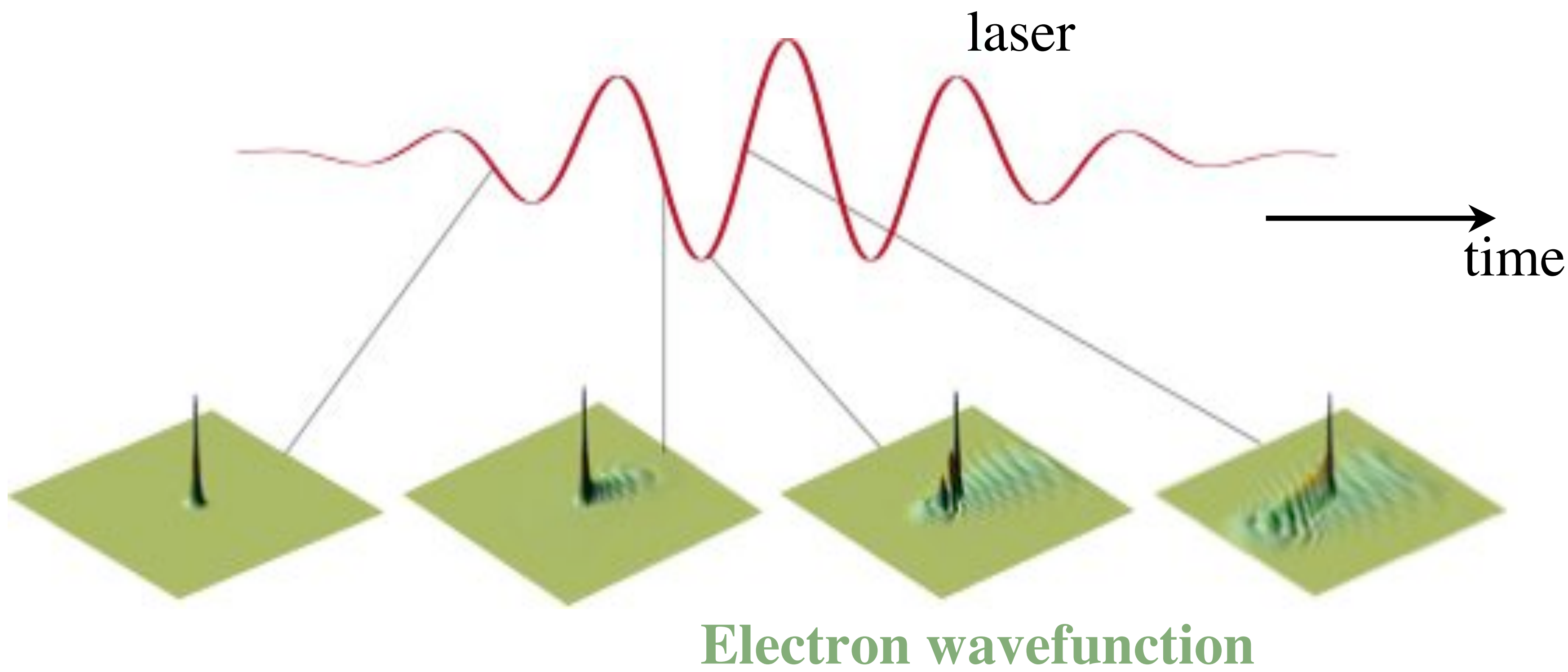


- Propagation, "spatial" gradients
  - P. Saliéres et al PRL 74, 3776 (1995)
  - P. Balcou et al, PRA 55, 3204 (1997)

- Chirp dependence of spectrum
- Single atom, time-domain
  - Z. Chang, et al., PRA 58 (1), R30 (1998)

Quantum picture (2D) of electron in strong field

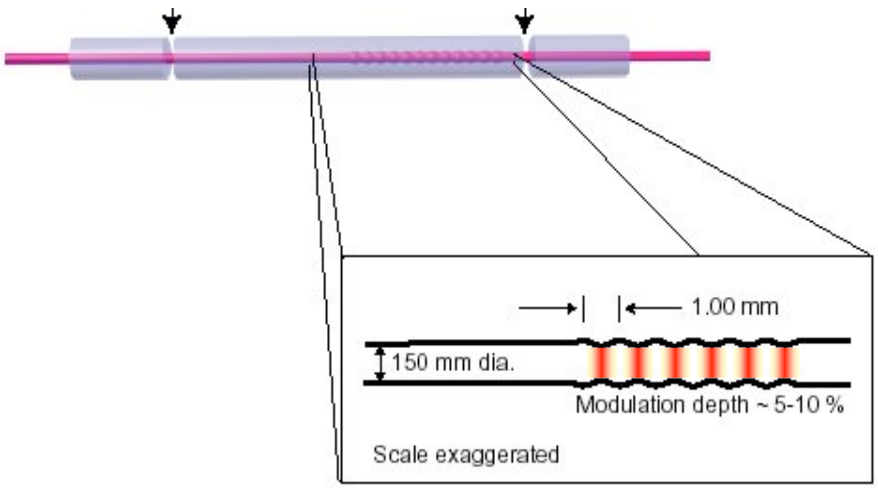
- Phase shift φ~1 rad / harmonic order

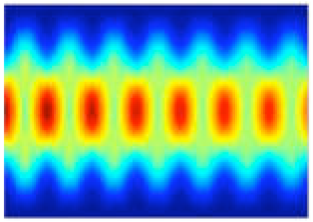


Physics Today, Kapteyn et al. March 2005

"EUV photonics" : Quasi Phase matching

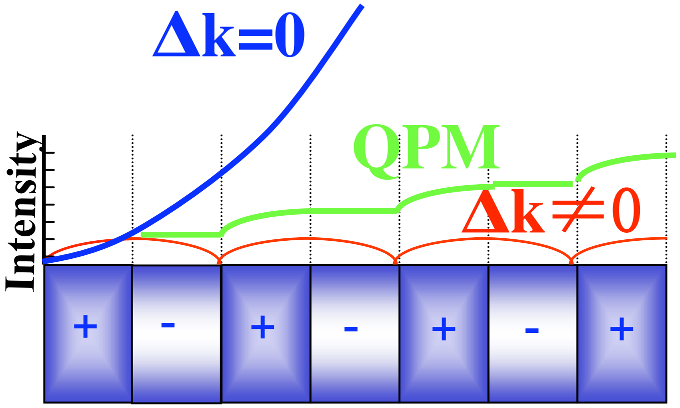
- Modulate the driving field by modulating a waveguide
- HHG is modulated because it is sensitive to phase and amplitude of driving laser





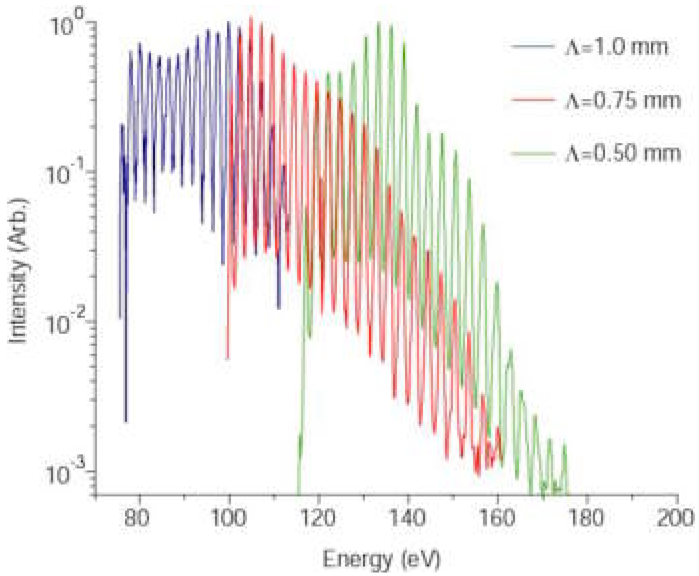
$$\Delta k_{QPM} = qk_f - k_q + \frac{2\pi}{\Lambda} = 0$$

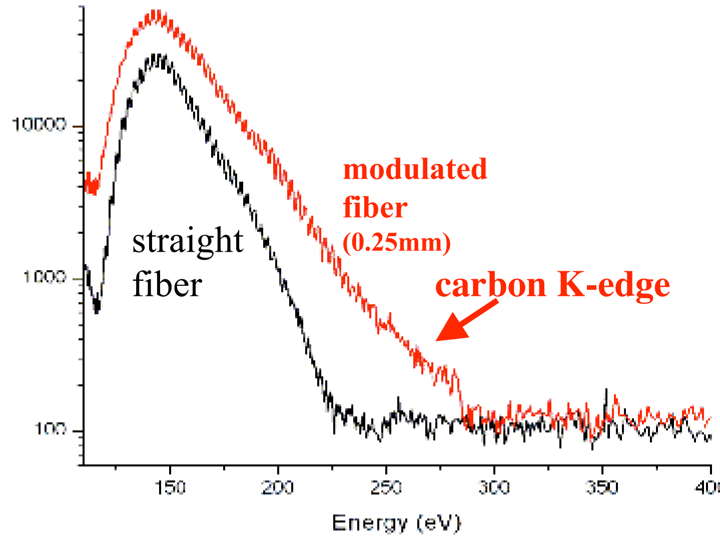
Λ= Periodicity of nonlinear medium

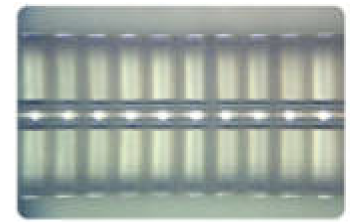


$L_{coh}=\pi/\Delta k$

QPM using modulated waveguides



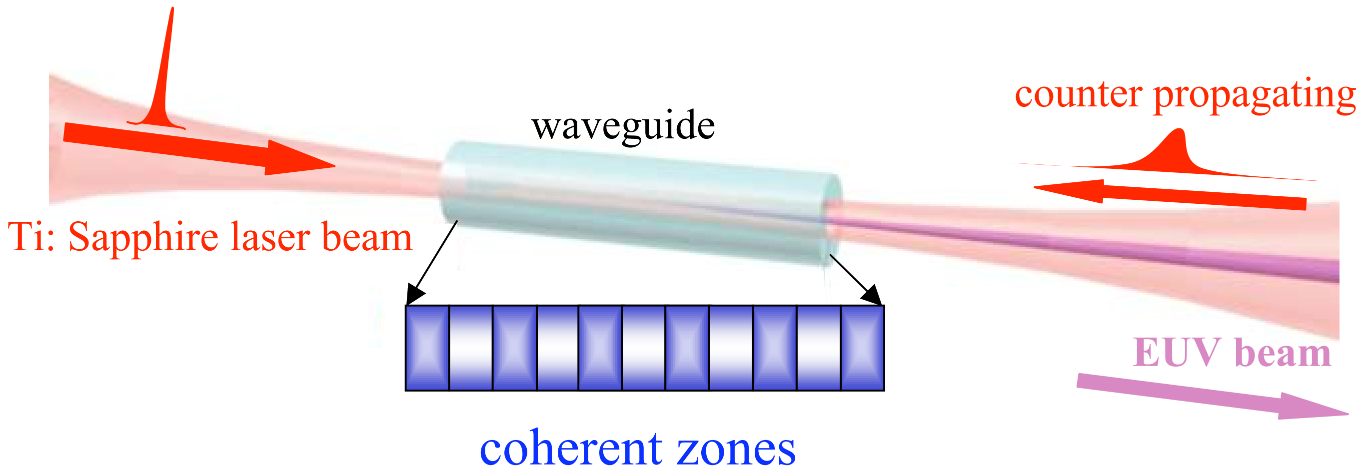




Nature ***421****, 51 (2003)*
Science ***302****, 95 (2003)*

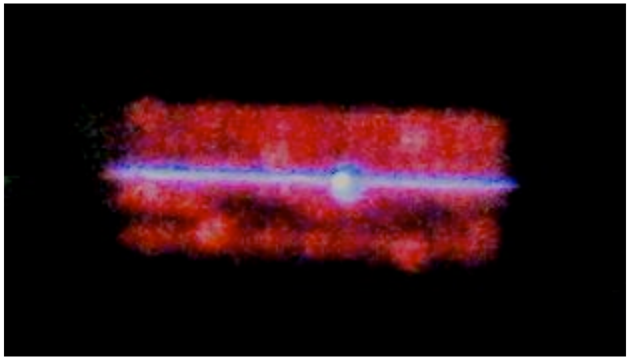
- First quasi phase matching technique to work in highly-ionized gas
- Pathway for more efficient higher harmonics (up to keV)
- BUT:
  - Limited (~10-100x) enhancement because of varying coherence length
  - To design the modulation, need to know coherence length
  - Periodicity limited to ~diameter
  - Plasma and waveguide help with quasi random QPM

Counterpropagating pulses

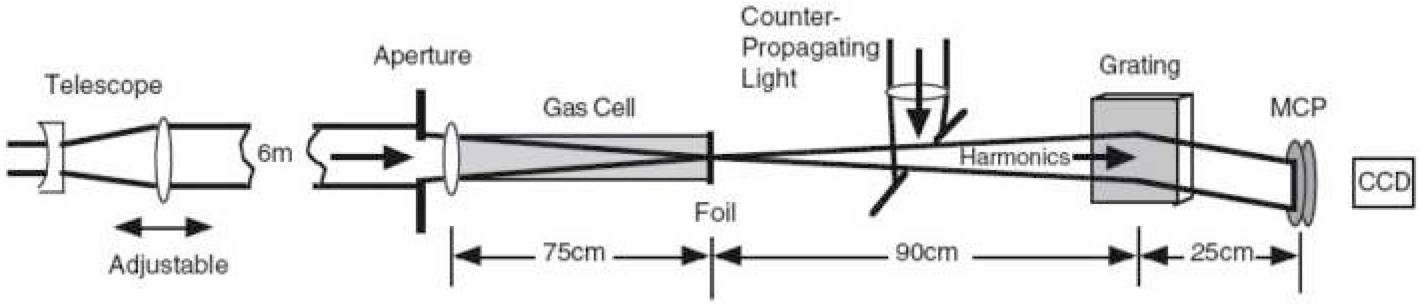


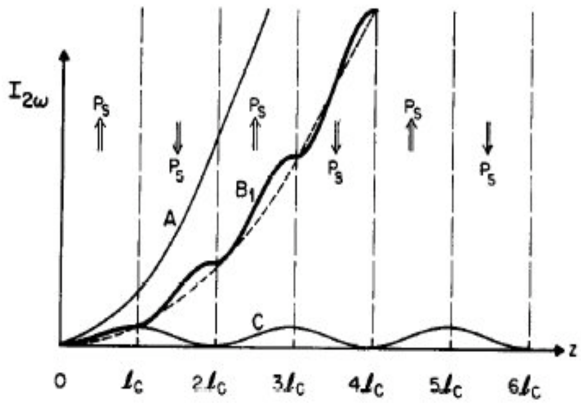
- Counterpropagating beam can probe coherence

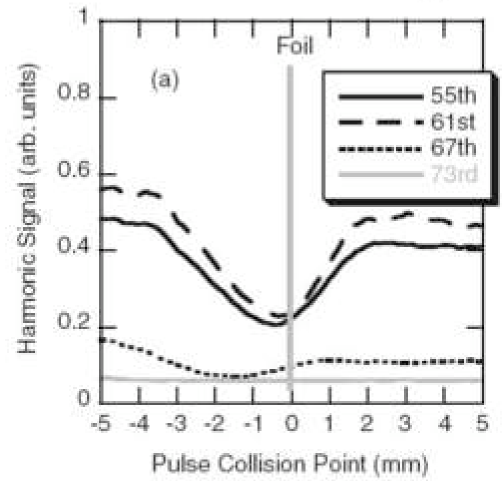
- Pulse train can implement QPM



Counterpropagating beams in a gas cell



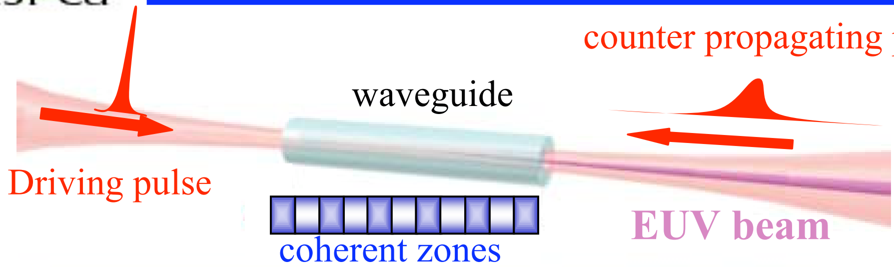


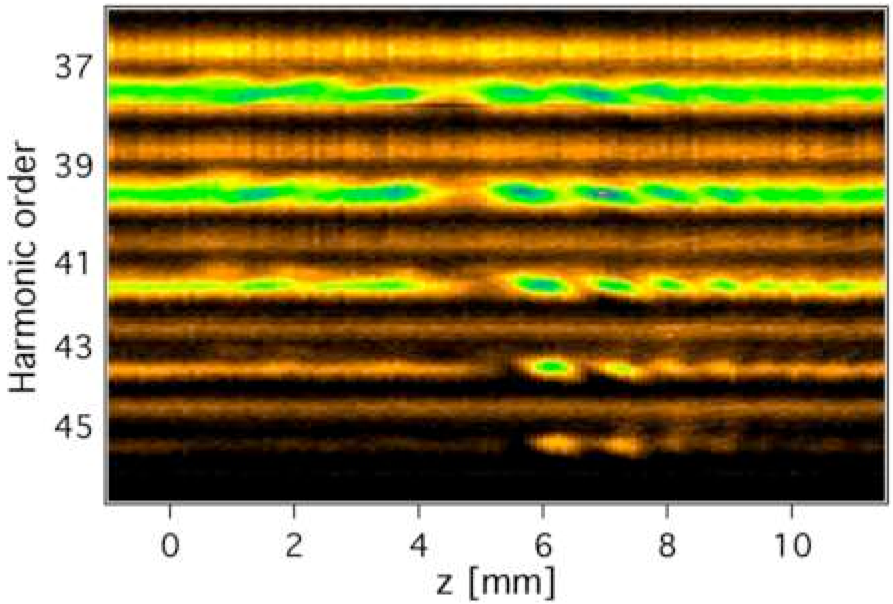


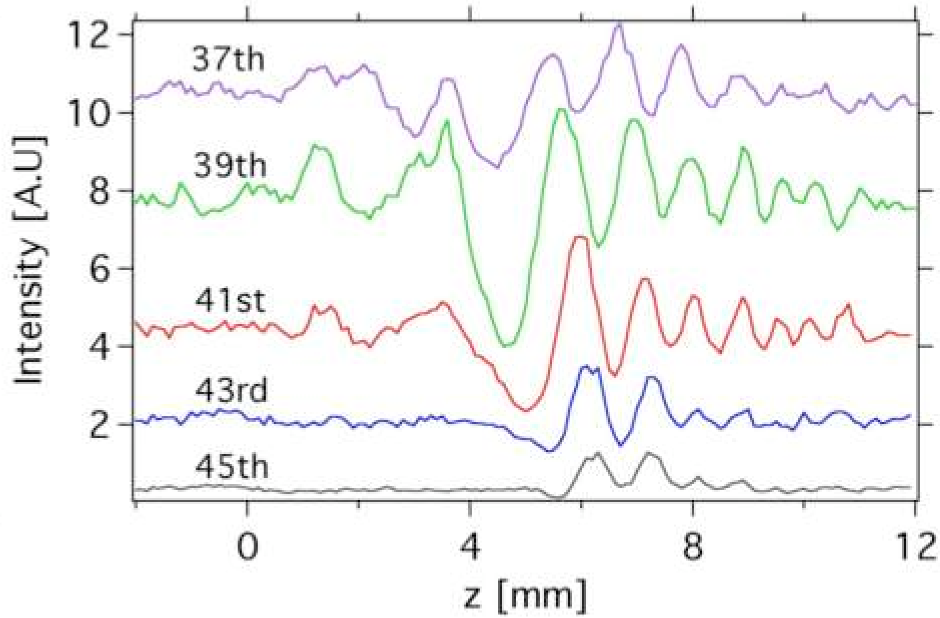
- Presence of counterpropagating field disrupts HHG
- Observe suppression of HHG
  - Peatross et al. PRL 84, 2370 (2000); Opt. Exp. 12, 4430 (2004)

- Should work better in hollow waveguide
  - long, uniform, interaction length
  - pressure-controlled phase matching

Single counterpropagating pulse in waveguide

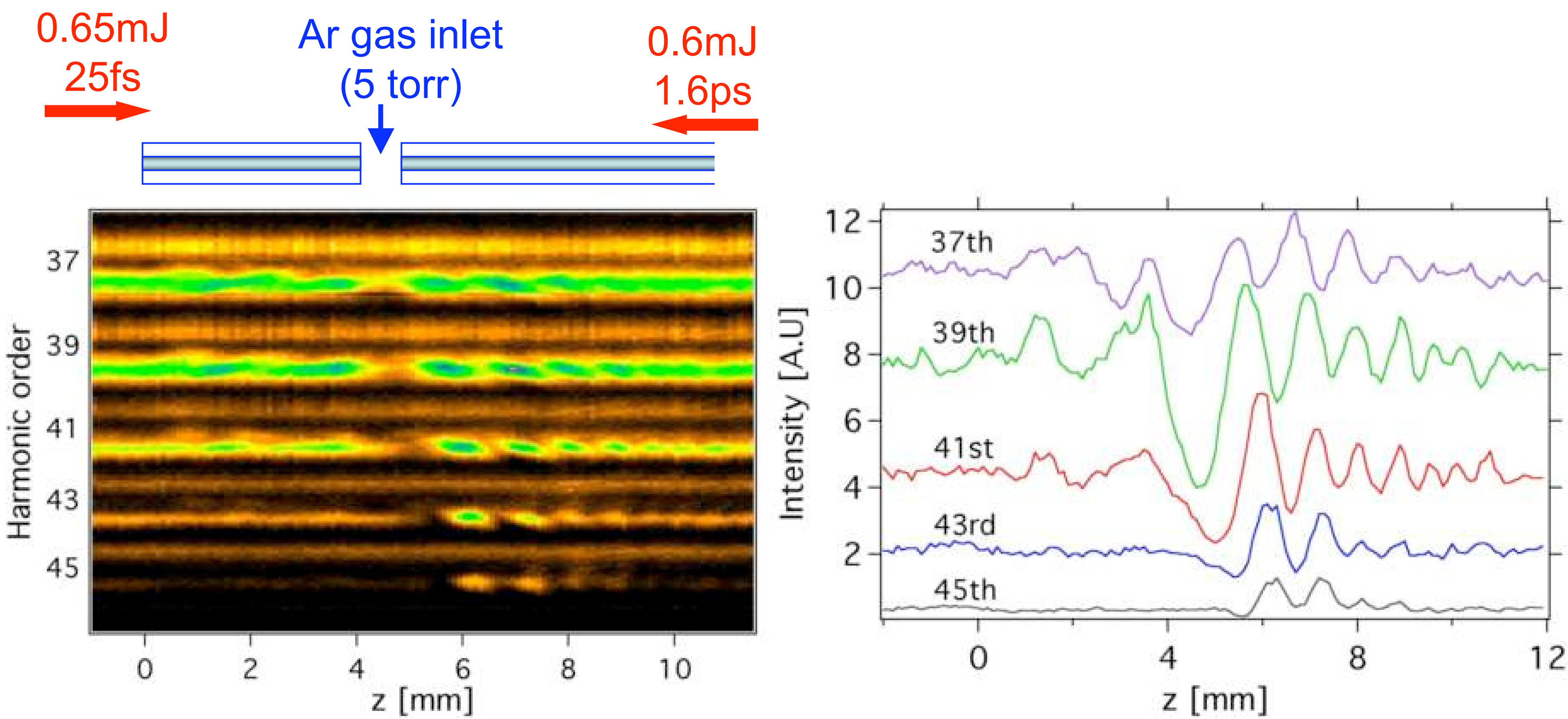






- Use low pressure, non-phase matched regime
  - "standard" phase matching in waveguide is ~30 torr for H23-31
- General method for mapping coherence
  - coherence length corresponds to 1/2 fringe period

Single counterpropagating pulse in waveguide



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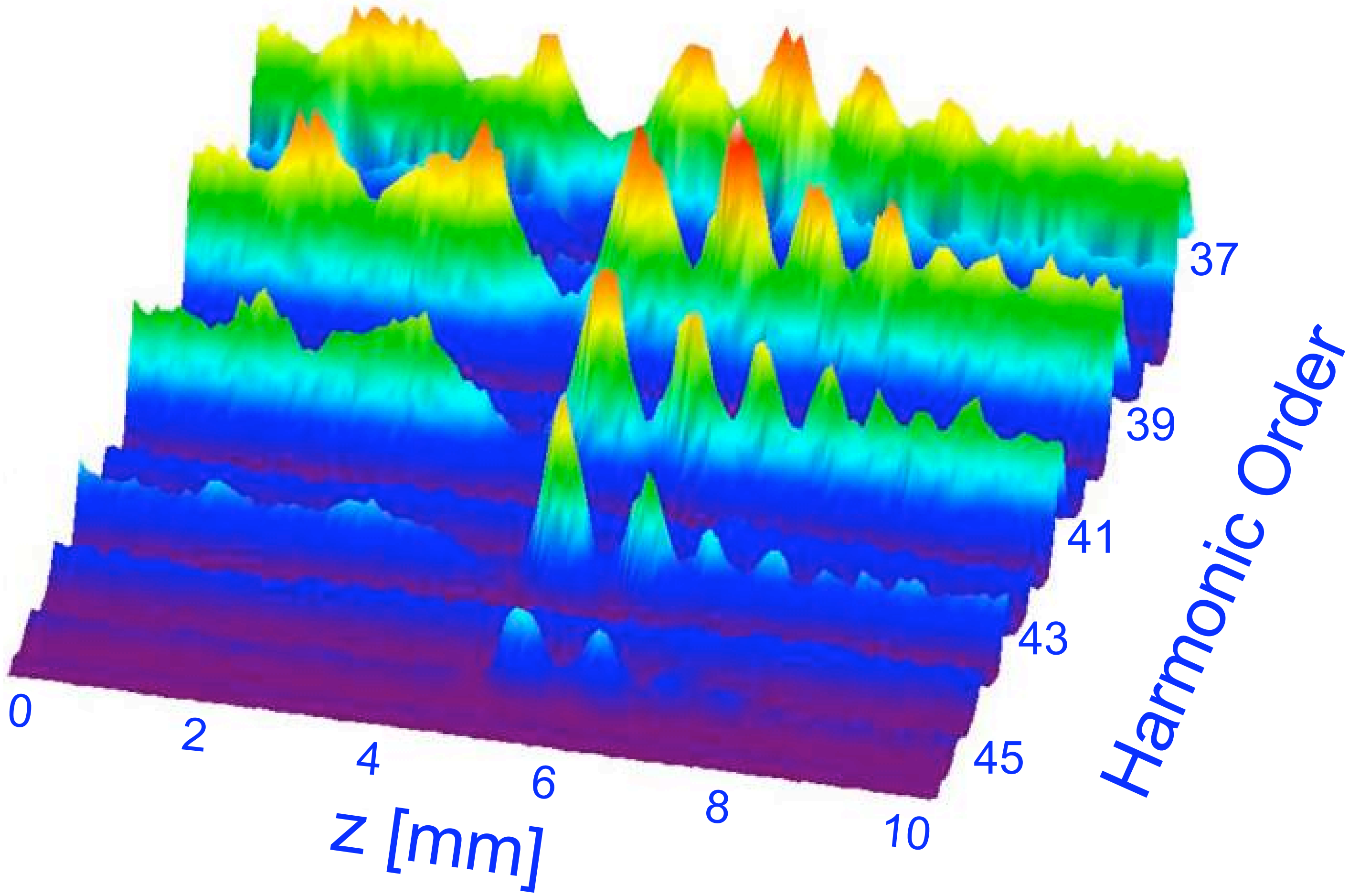
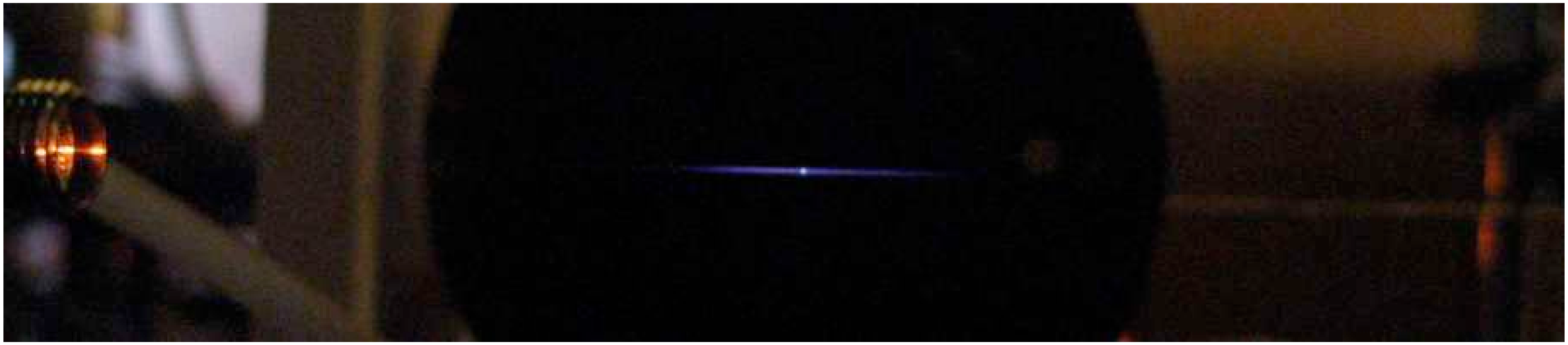
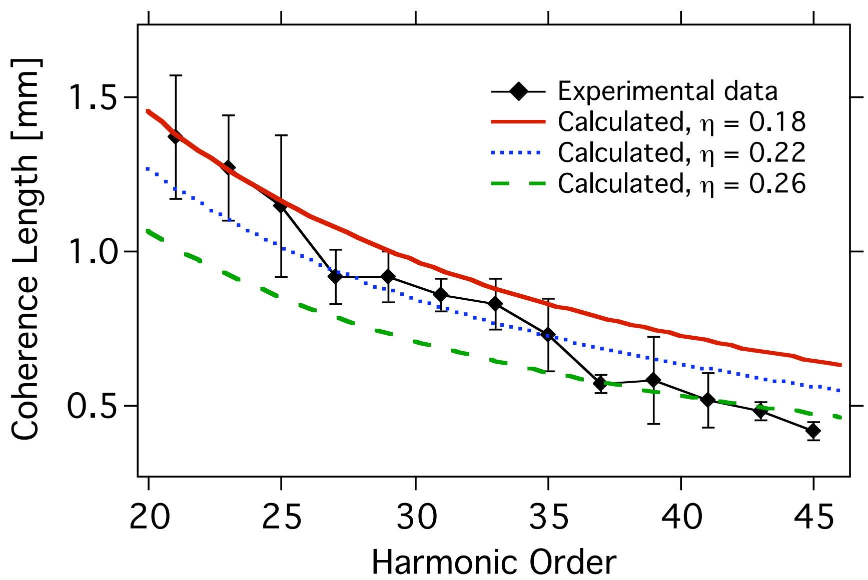
Single counterpropagating pulse

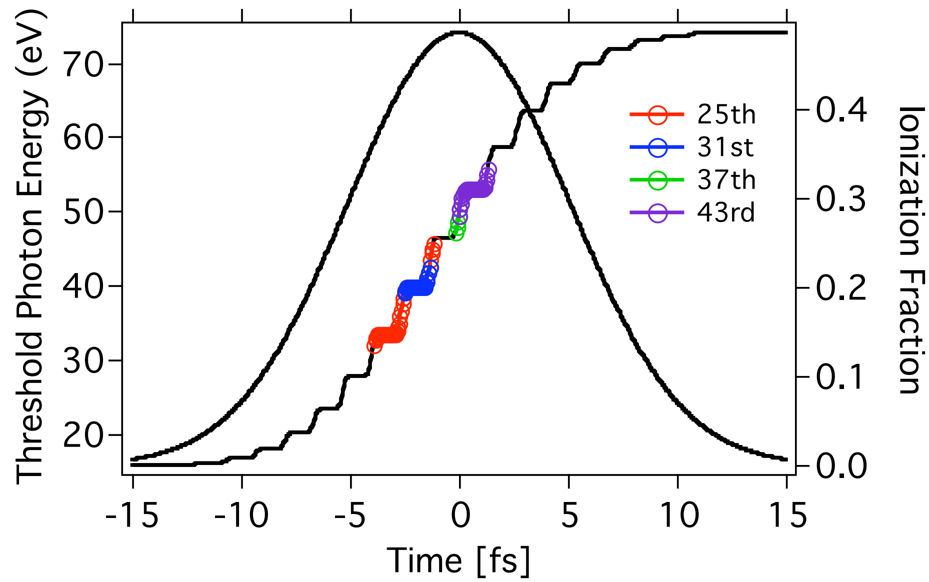


Colliding pulses



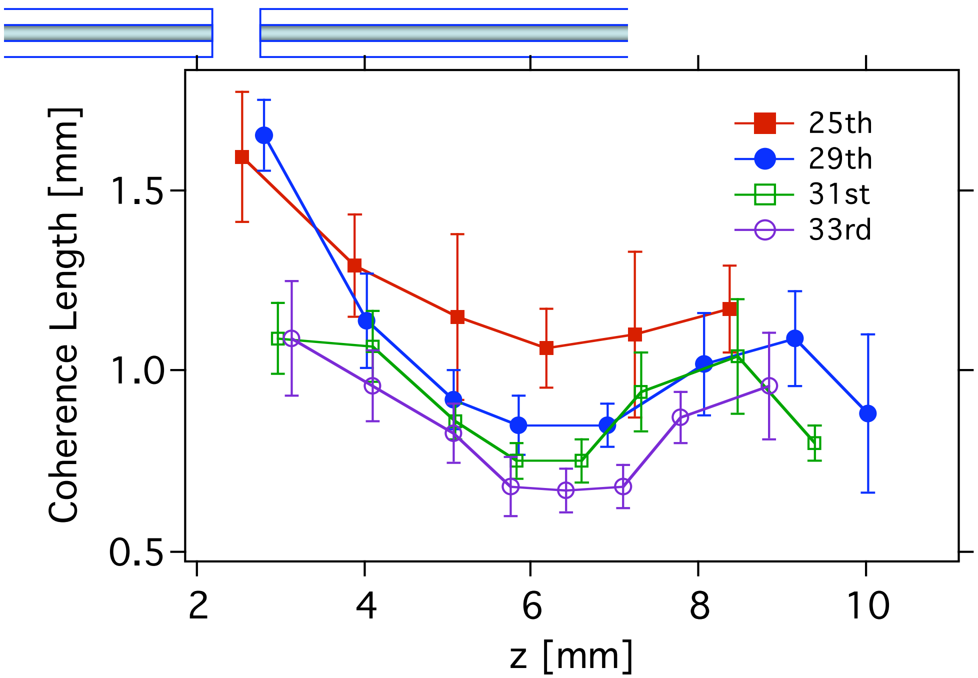
Coherence length vs order, position

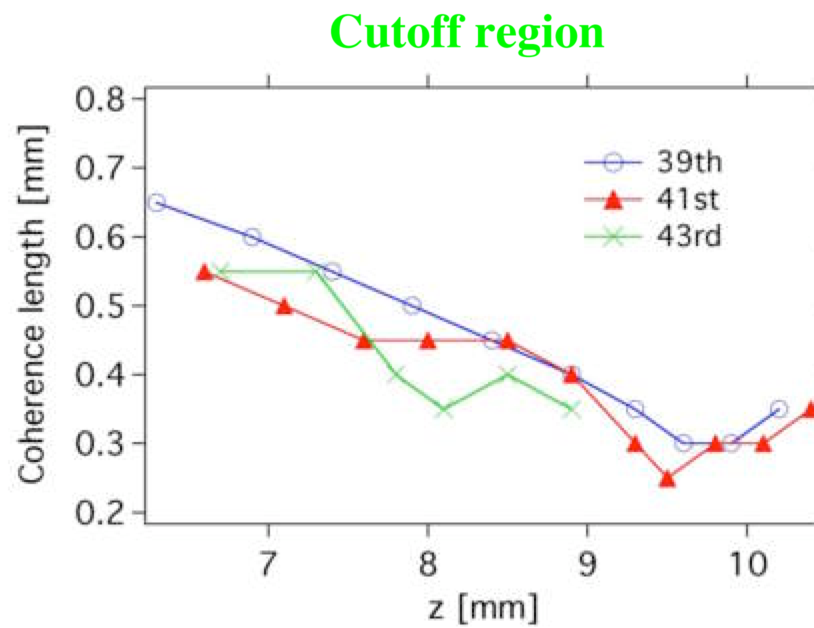




- L_c decreases with increasing harmonic order
- At high ionization, near cutoff, $L_c \sim 1/q^2$
- ADK and L_c can be used to identify at which ionization levels different harmonics are generated

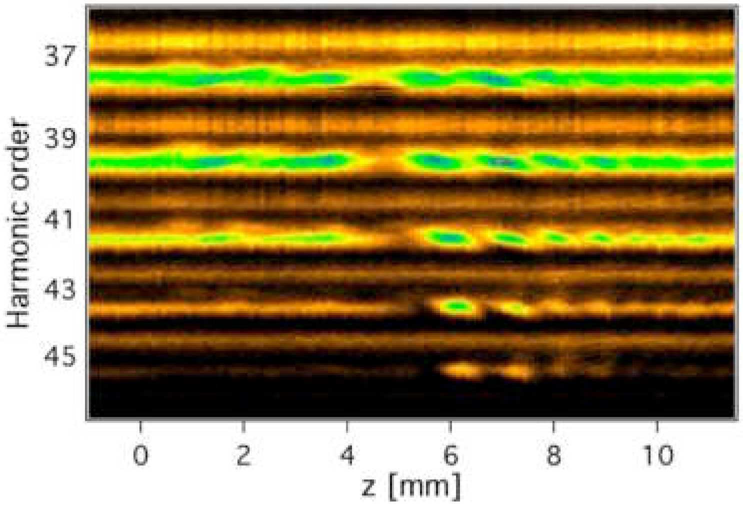
Coherence length in waveguide





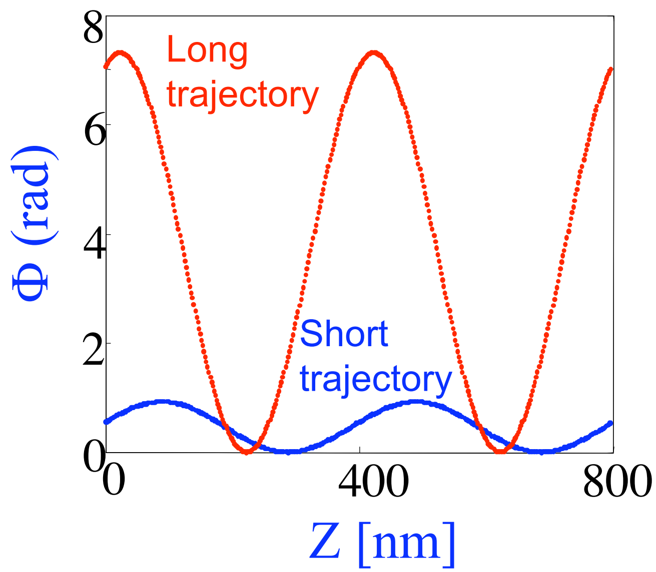
- Loss in waveguide decreases ionization, increases L_c toward exit
- Varying L_c limits number of fringes observed for fixed counterpropagating pulse duration
- Evidence of mode beating?

Quantum path control

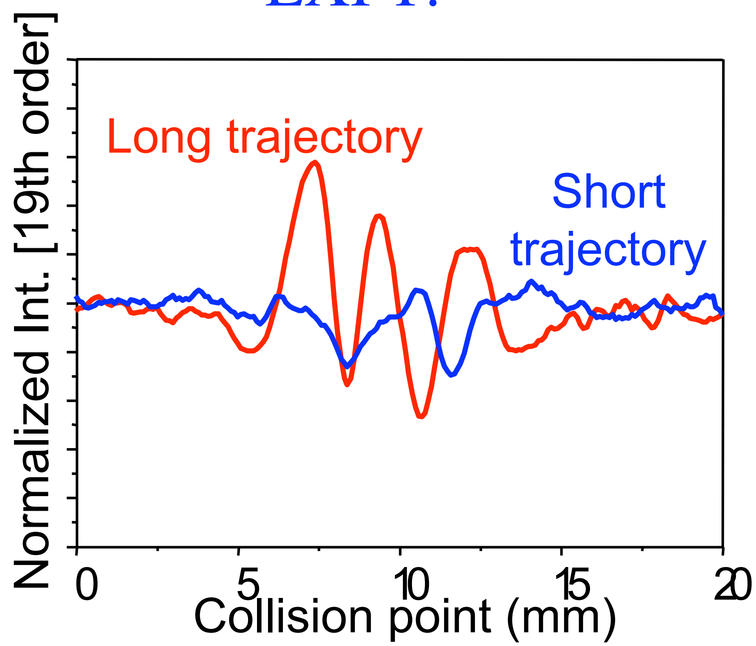


- Observe HHG from long and short trajectories
- Long trajectories strongly modulated, while short trajectories need higher energies

THEORY

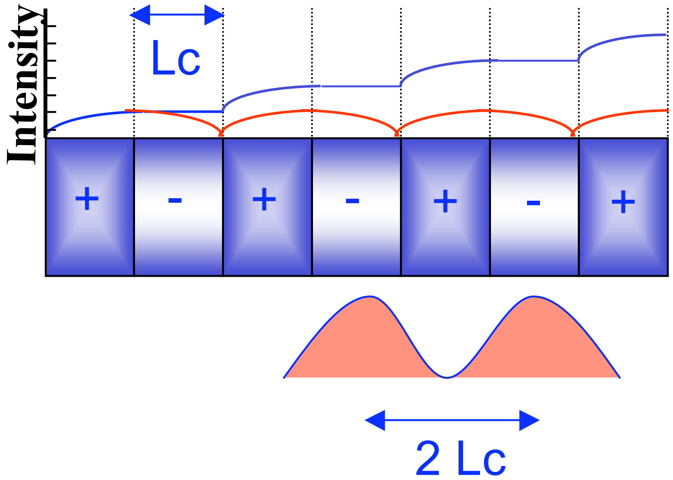


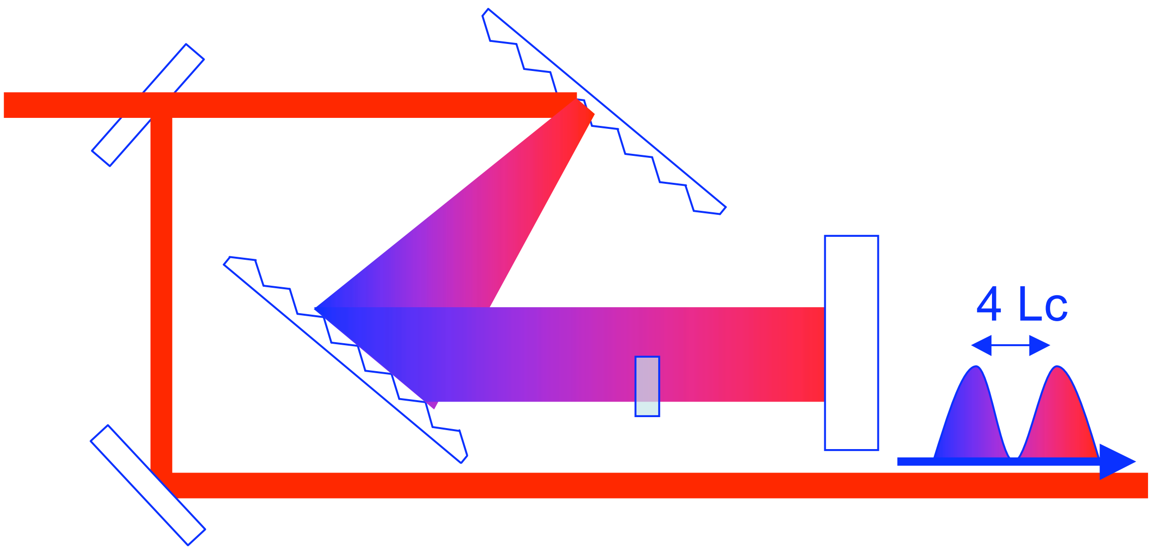
EXPT.



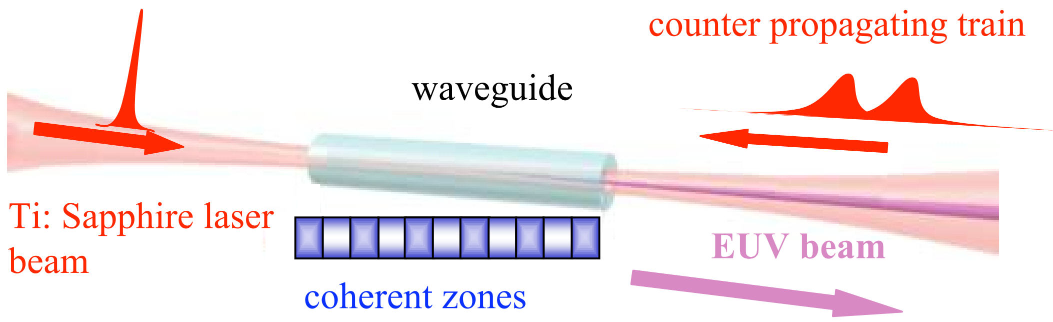
QPM using a TWO pulse "train"

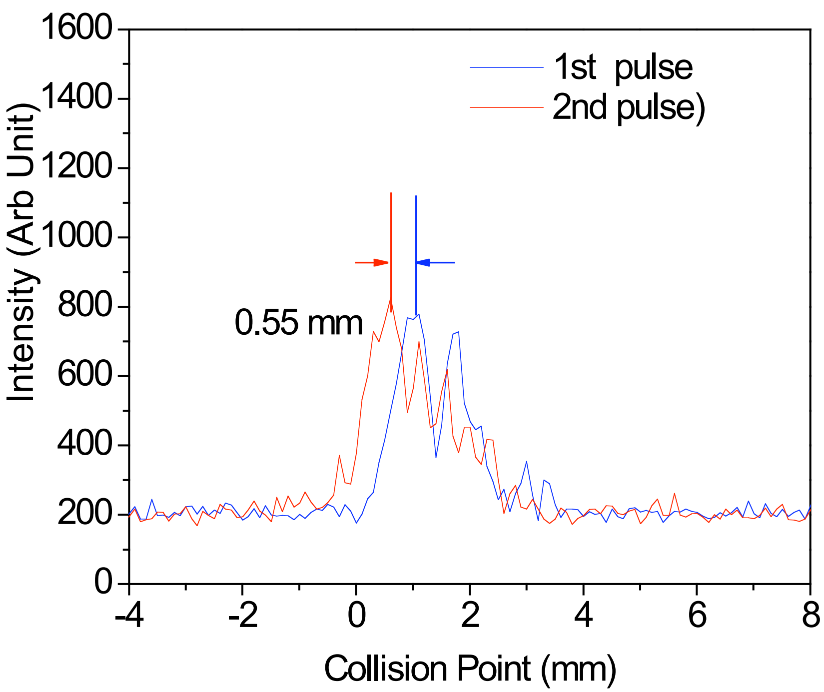
- N pulses give (N+1)2 enhancement (expect factor of 9)
- Glass plate in stretcher splits pulse, allows independent control





Modulation due to each pulse, separately

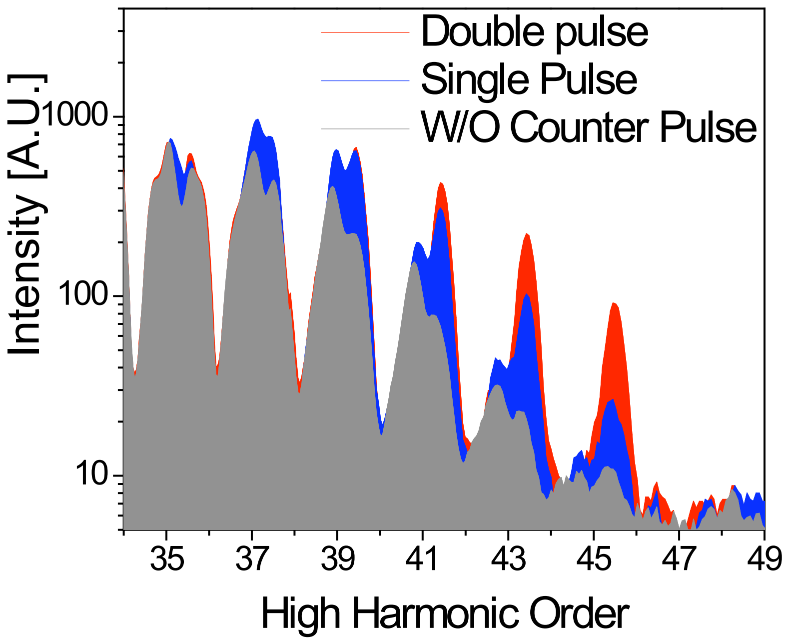


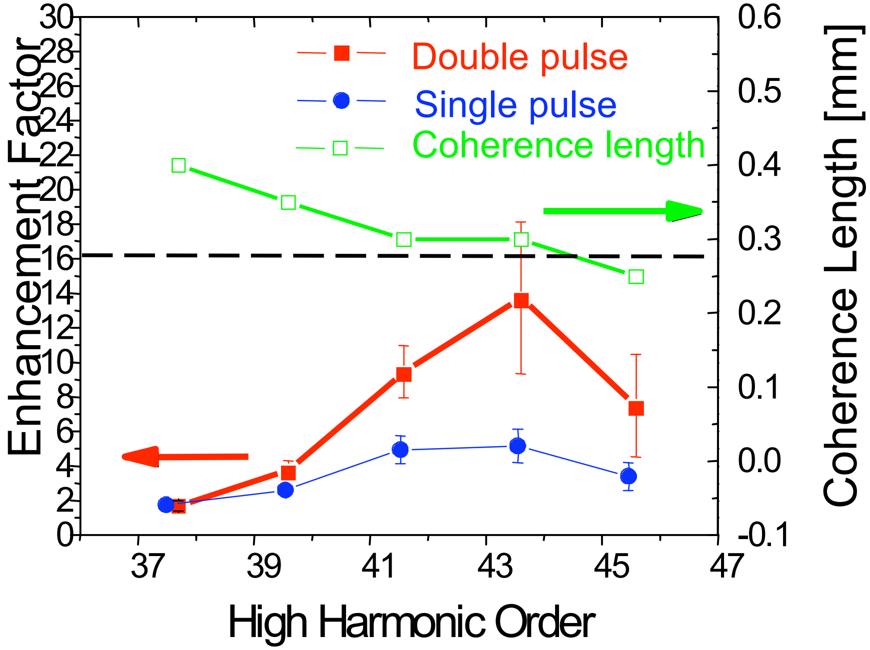


- Pulse sequence adjusted so each counter propagating pulse causes modulation

- Can measure and adjust the pulse separation - in this case ≈ 1.1mm

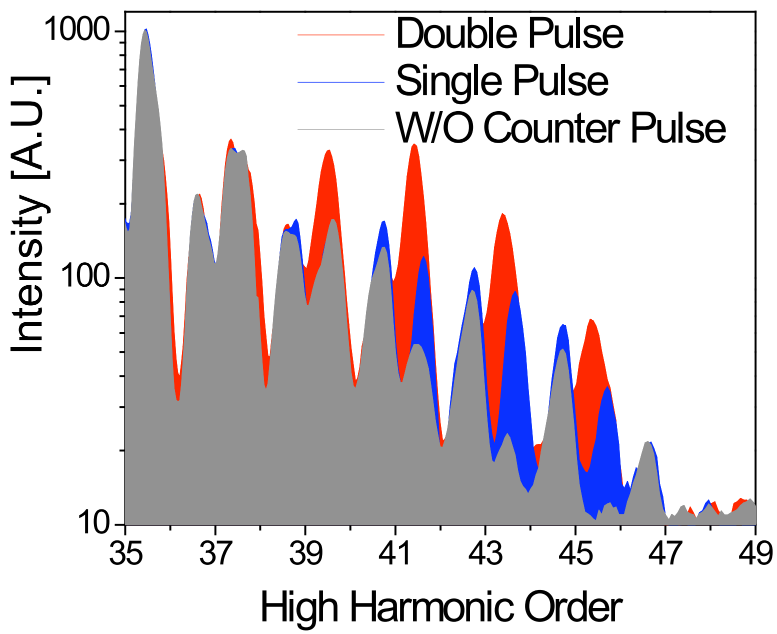
QPM using two pulses

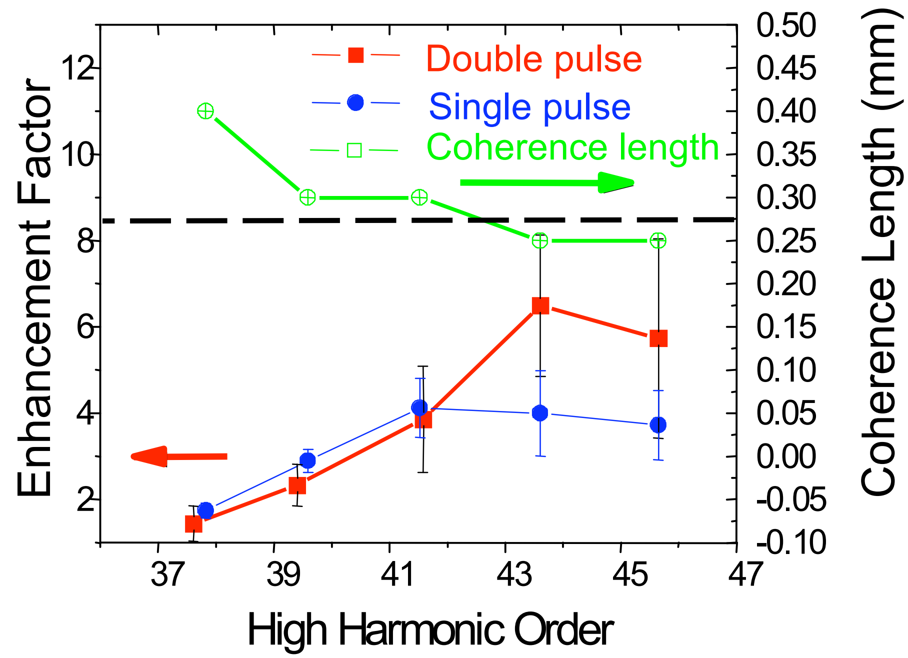




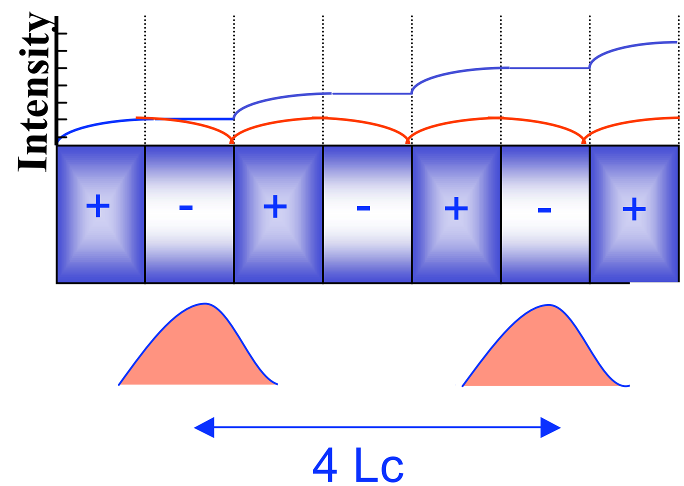
- Use 1.1 mm pulse separation
- When L_c = 1/4 pulse separation, largest enhancement
- H43 is closest to QPM period => shows largest (x14) enhancement!!

Higher-order QPM

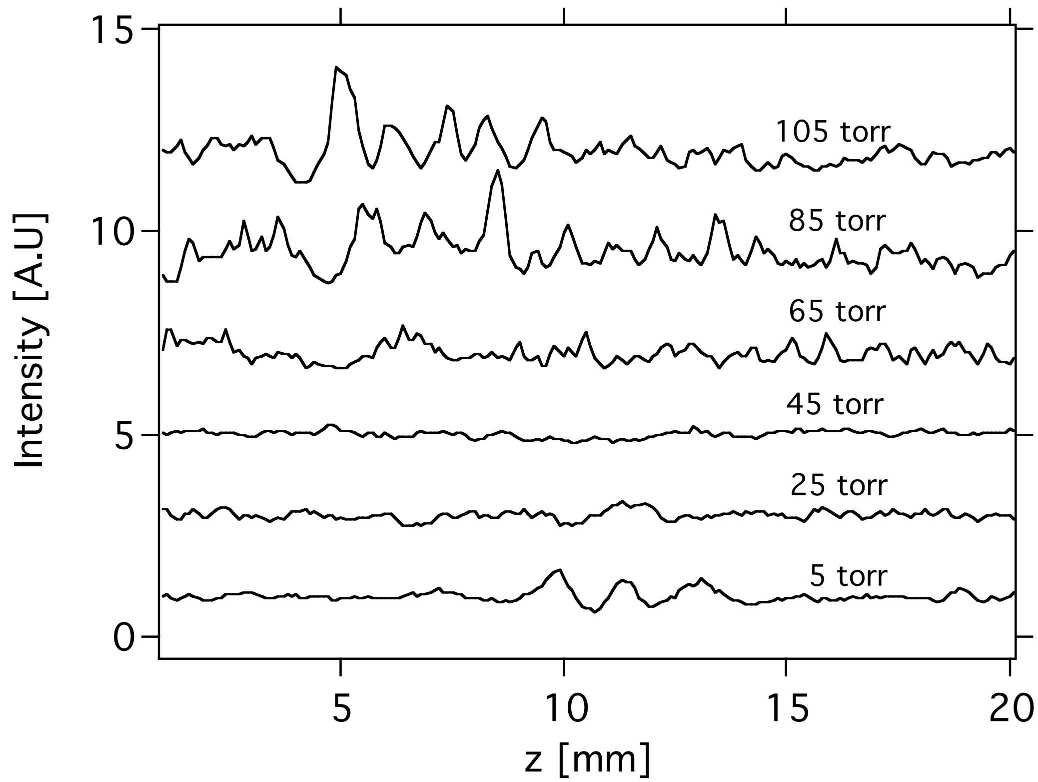


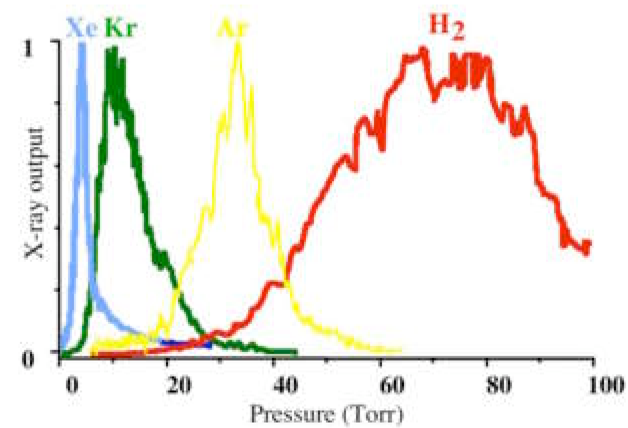


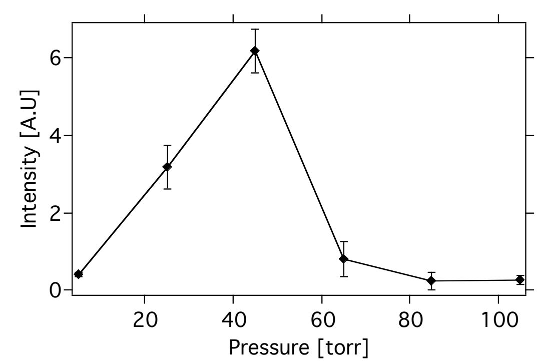
- Pulse separation 2.2 mm
- m=2 QPM is obtained at H43 with 7x enhancement



Directly observe pressure-tuned phase matching

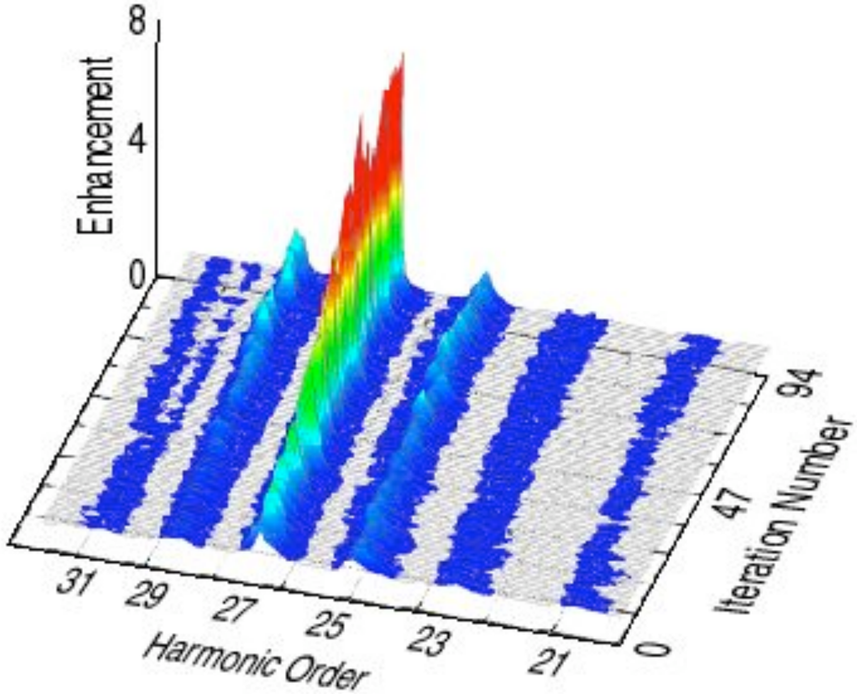






- H29 (in standard phase-matching regime!)
- For phase matching (~40 torr) L_c is longer than counterpulse
- For lower and higher pressures, observe finite coherence length

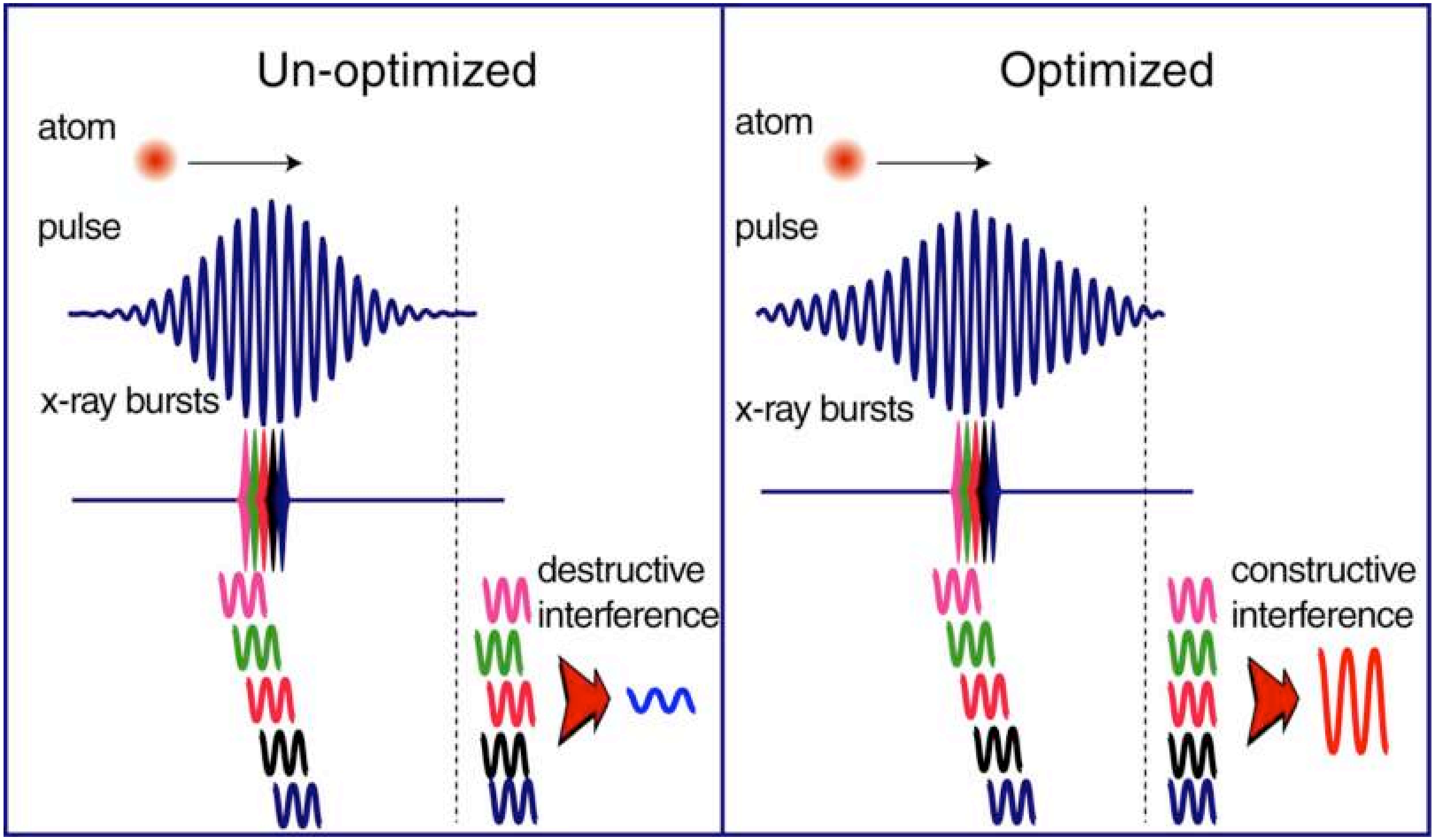
Excellent p-m in WG reveals atto dynamics



Bartels et al., Nature ***406****,164 (2000)*

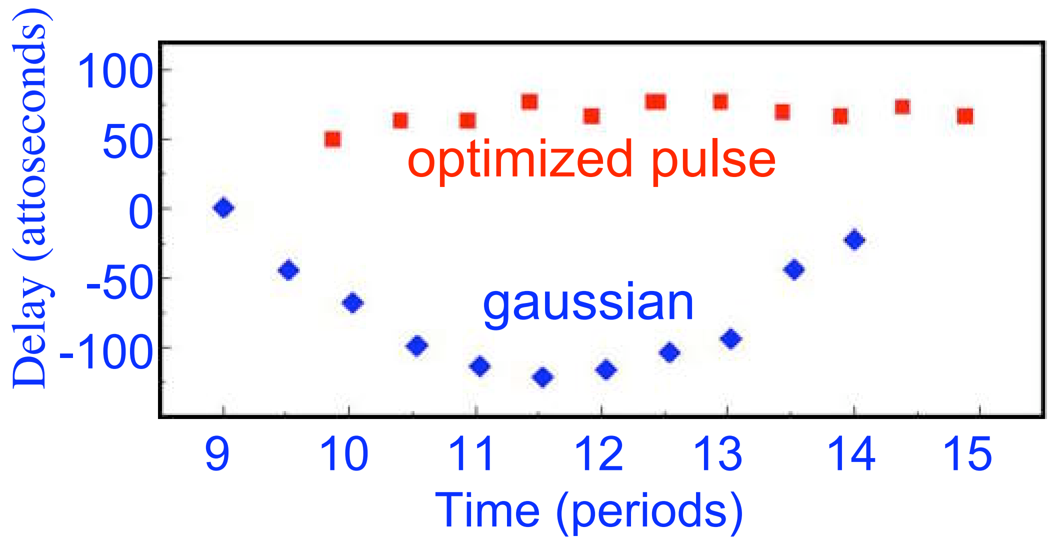
- Phase matching: signal from all emitters adds in phase
  - ∴ signal reflects *single atom* dynamics
- 1st demonstration of learning control on a very high-order quantum nonlinear system
- Learning algorithm discovered new science!

Controlling interfering pathways

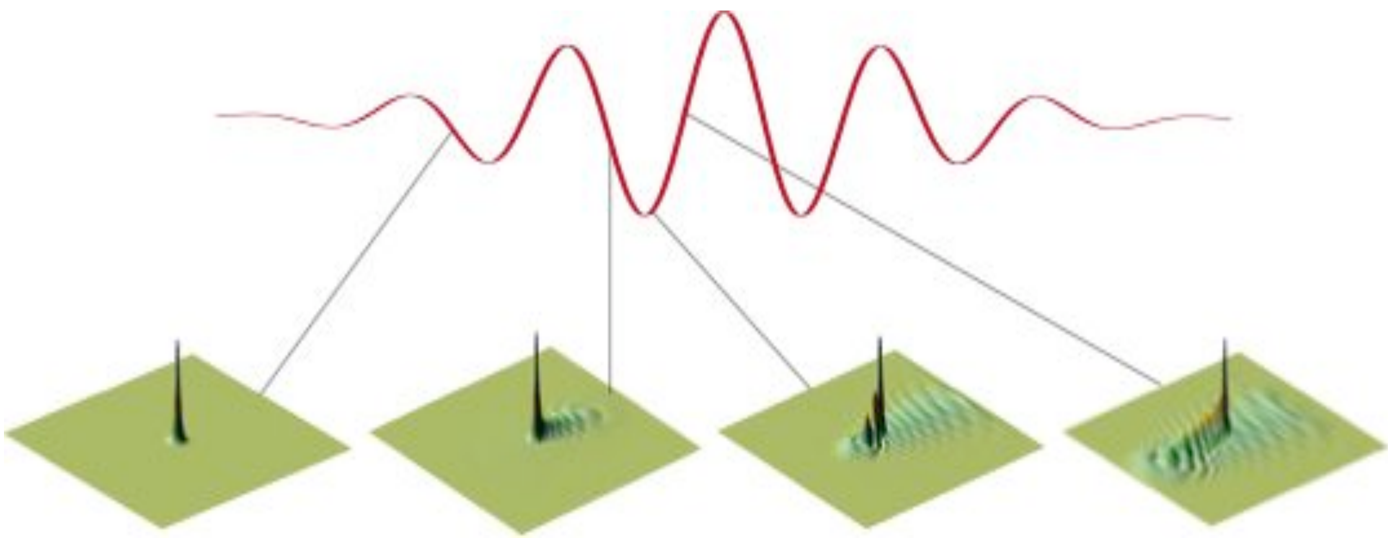


Christov et al, PRL ***86****, 5458 (2001)*
Bartels et al. Chem. Phys. ***267****, 277 (2001)*
Bartels et al. PRA ***70****, 112409 (2004)*

Attosecond control



- Use algorithm to optimize theory
- For optimized laser pulse, all harmonics in phase within **25 attoseconds**!
- Experiment feasible using *few-cycle* pulses

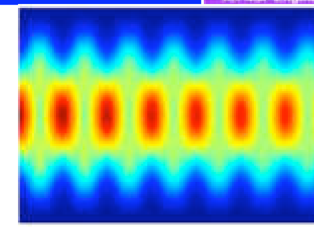


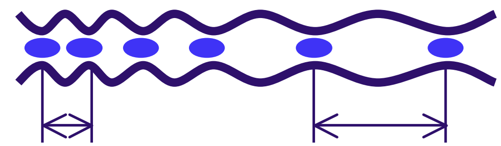
How far can we go?

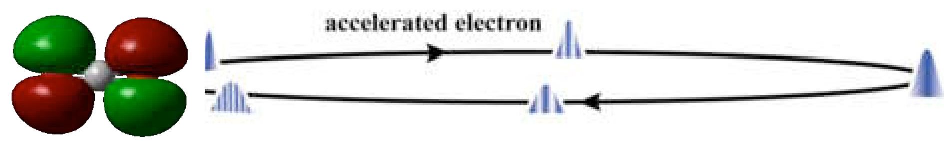
- HHG in Helium in the water window
  - 5 torr L_c ~100 μm
  - Absorption depth @ 300 eV: 10 *meters*
  - *Possible enhancement:* $\left(\frac{10}{10^{-4}}\right)^2 \sim 10^{10}$
- Neon
  - L_c~ 100 μm
  - Abs depth ~0.5 meters
- Limitations: defocusing, waveguide propagation, group velocity slip

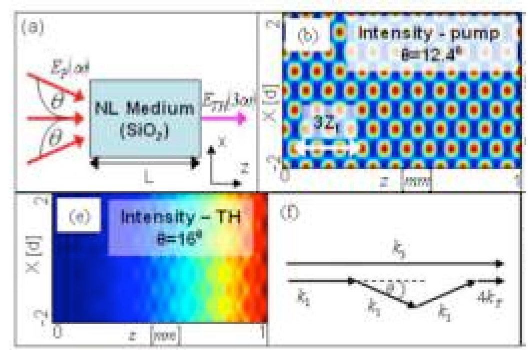
Future plans for HHG sources

- Better quasi phase matching techniques
  - pre-formed, tailored and modulated discharges
  - counterpropagating pulsetrains
  - chirped, tapered, waveguides
  - 1-D waveguide to increase flexibility of structures
  - quasi phase matching using two-color laser fields
  - HHG from molecules





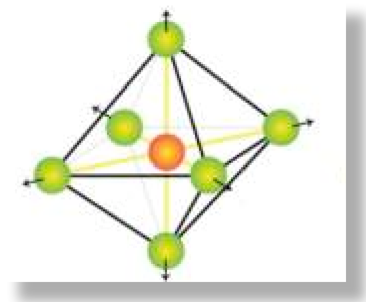


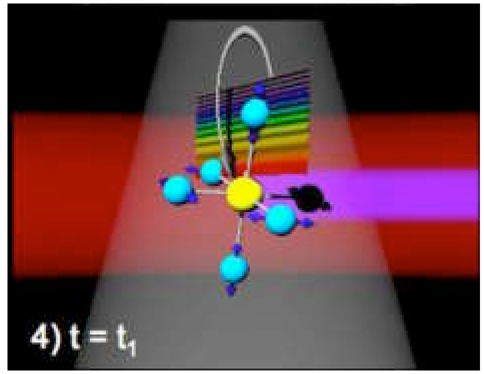


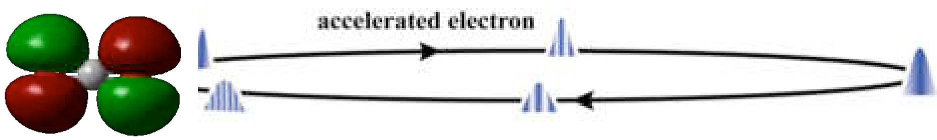
- More-extensive modeling to understand laser propagation in plasma-filled waveguides

- Multi kHz repetition rate lasers

Ultrafast Coherent Spectroscopy and Imaging





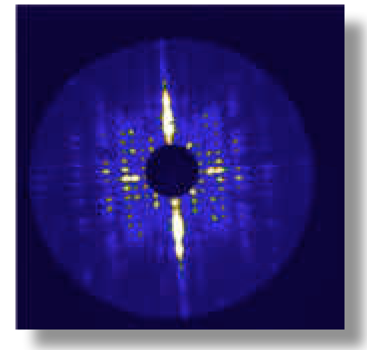


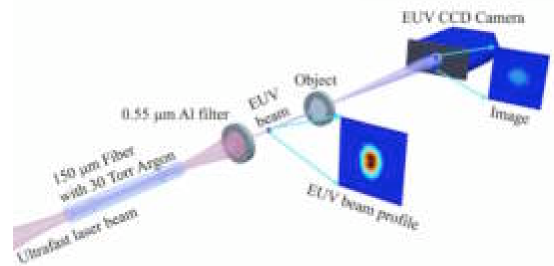
High Order X-Ray Raman Probes of Molecules

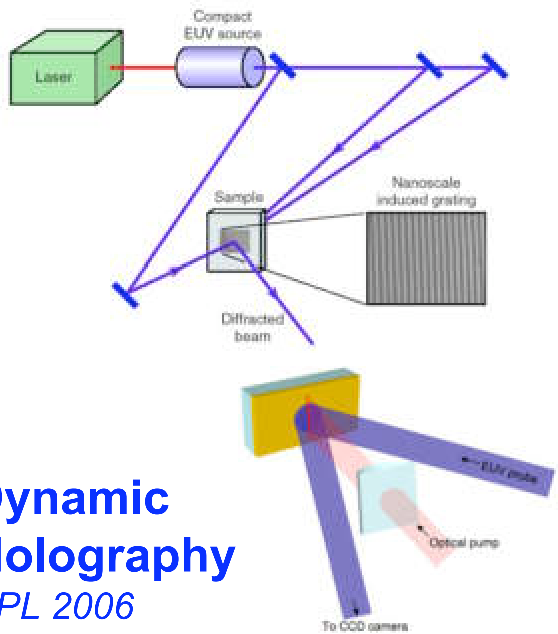
PNAS Sept 2006

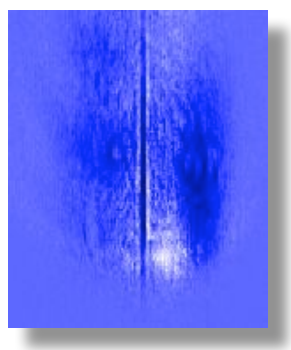
Lensless Imaging

Optics Lett. submitted



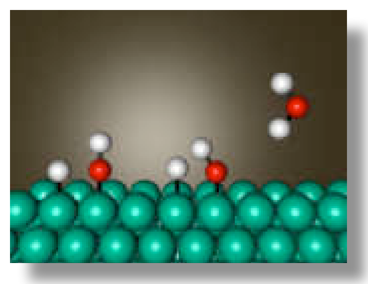


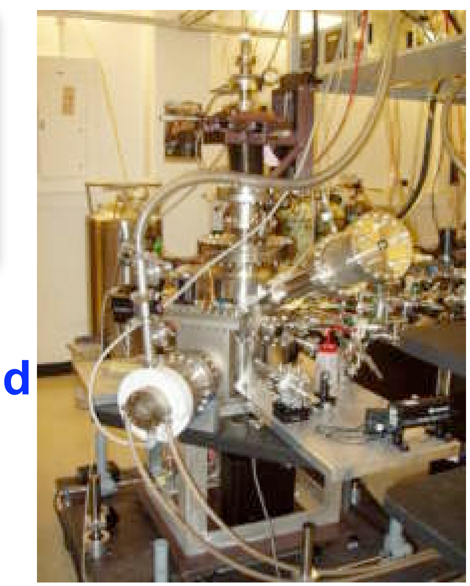




Dynamic Holography

APL 2006

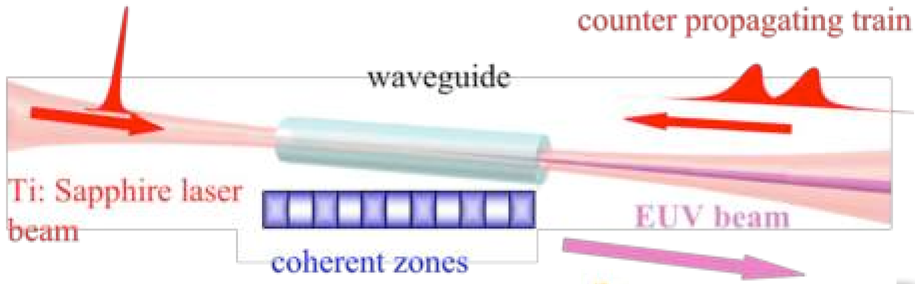


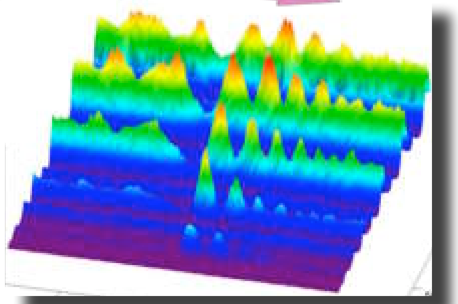


Attosecond solid state dynamics

PRL Sept 2006

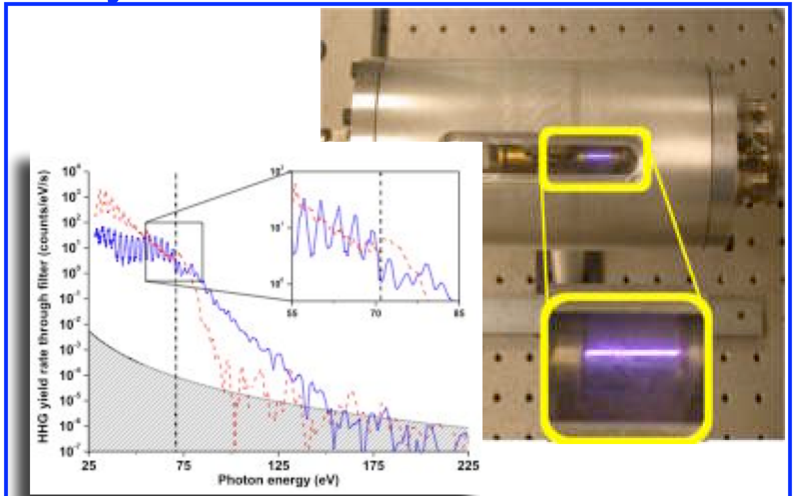
Generating Bright Ultrafast X-Ray Pulses



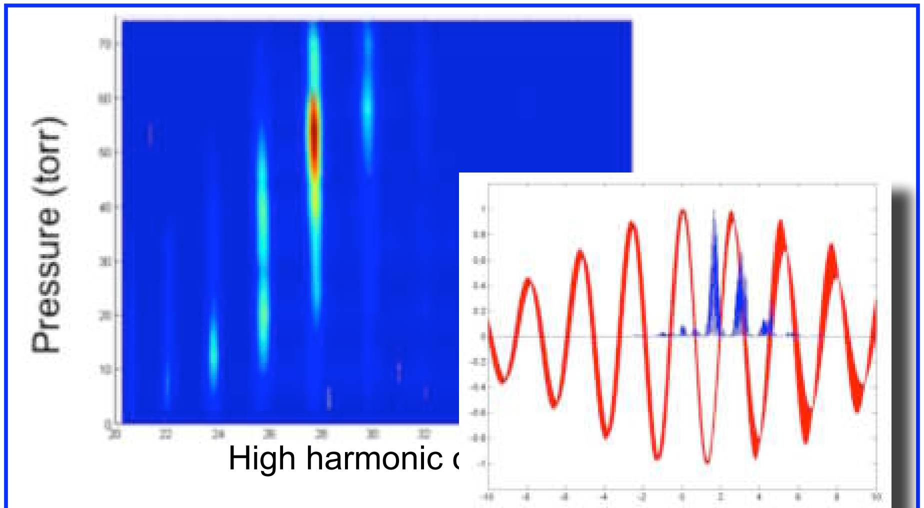


Quasi Phase Matching using Light

Postdeadline, Ultrafast Phenomena, Aug. 2006

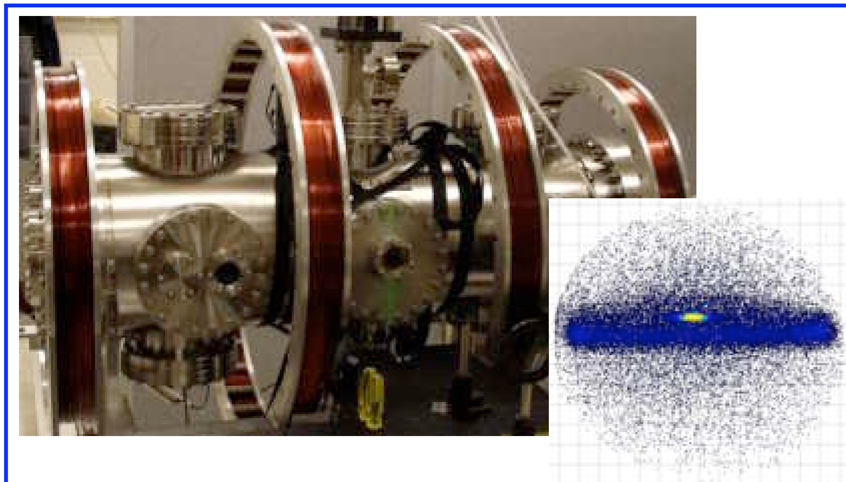


HHG from ions (plasma discharge) *PRL, May 2006*



Bright, sub-cycle, EUV pulses

PRL submitted



COLTRIMS - attosecond reaction microscope

Conclusion

- HHG does not simply “happen”-- it can be manipulated and optimized in sophisticated ways (!!)
- It involves the fastest *coherent*, controllable dynamics yet encountered by man
  - Complex, yet decipherable, spatial-spectral-temporal couplings
- The attosecond quantum dynamics of rescattering provides the basis for a new technology of extreme nonlinear-optics, with *many* possibilities
  - Shaped pulse optimization
  - Engineered waveguide structures
  - Counterpropagating fields
  - ???