

Generation of keV harmonics and ultrashort waveforms driven by midinfrared lasers

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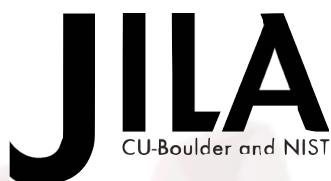


KITP, August 13th 2014



People involved

*'Generation of keV harmonics and ultrashort waveforms
driven by midinfrared lasers'*



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Tenio Popmintchev, Margaret M. Murnane, and Henry C. Kapteyn

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Luis Plaja

Grupo de Investigación en Óptica Extrema, Universidad de Salamanca, Spain

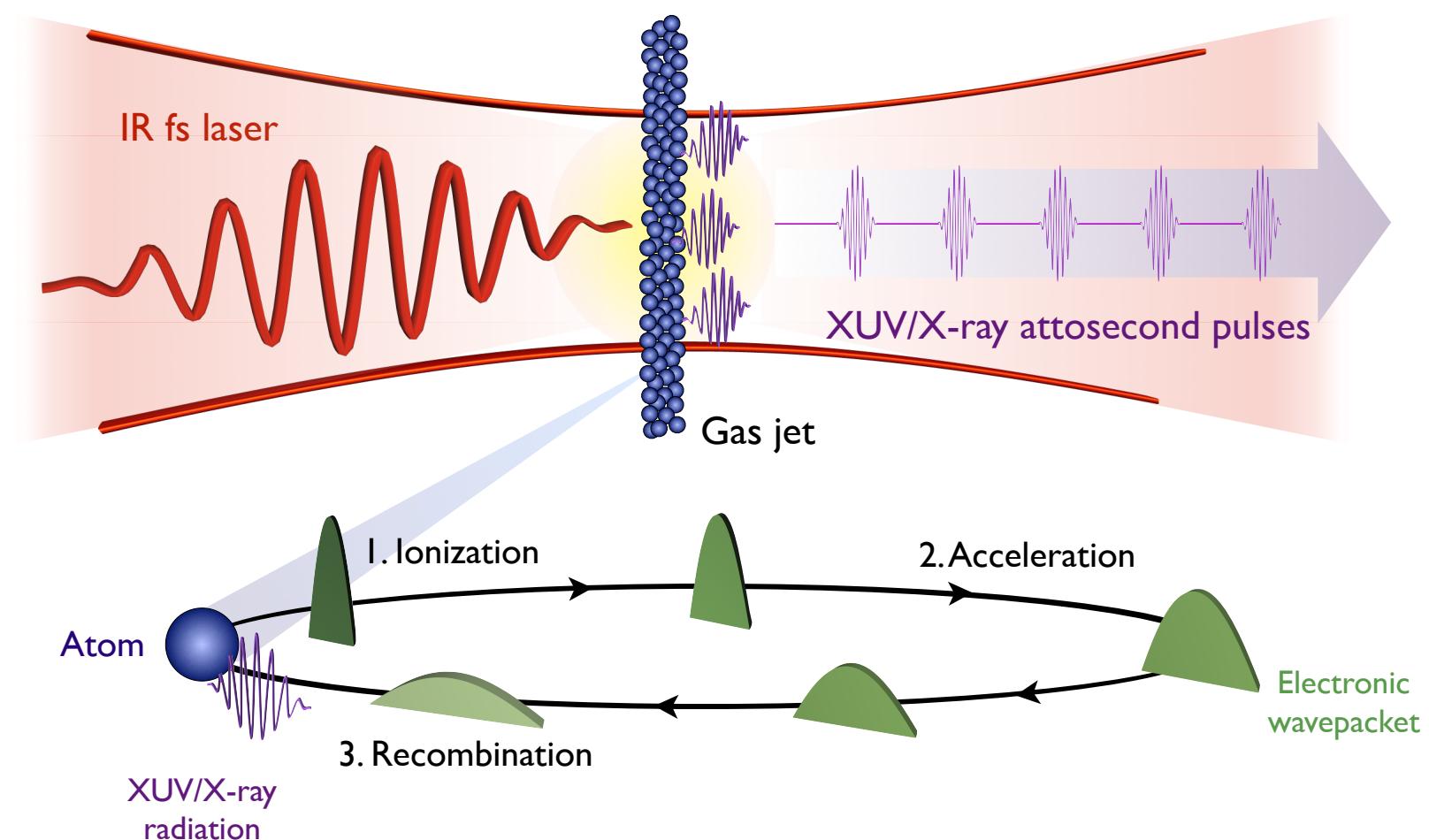
J.A. Pérez-Hernández

Centro de Láseres Pulsados, CLPU, Salamanca, Spain



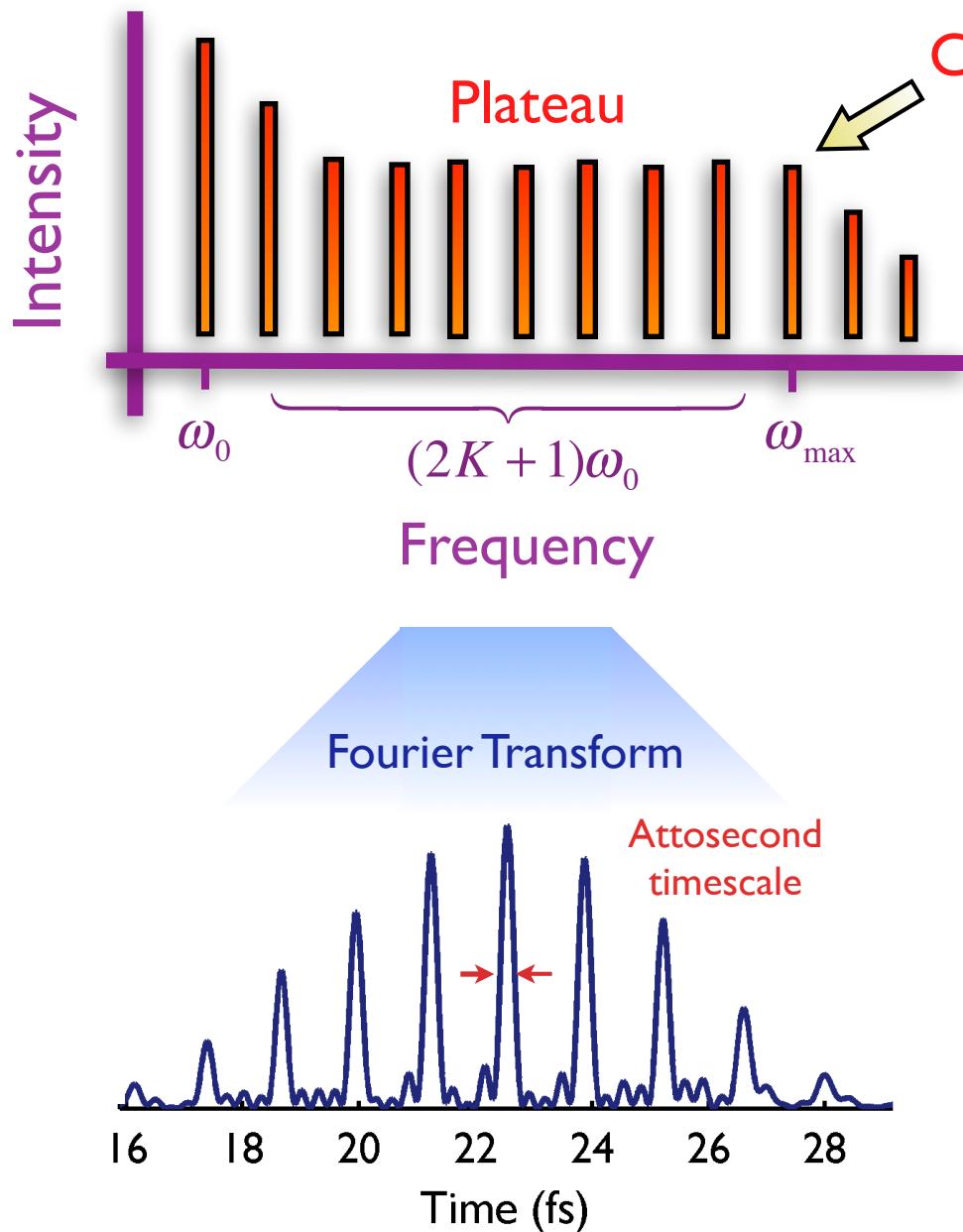
supported by DOE, NSF,
Janus supercomputer (Univ. Colorado)

High-order Harmonic Generation (HHG)



Corkum, PRL 71, 1994 (1993)
Schafer et al. PRL 70 1599 (1993)

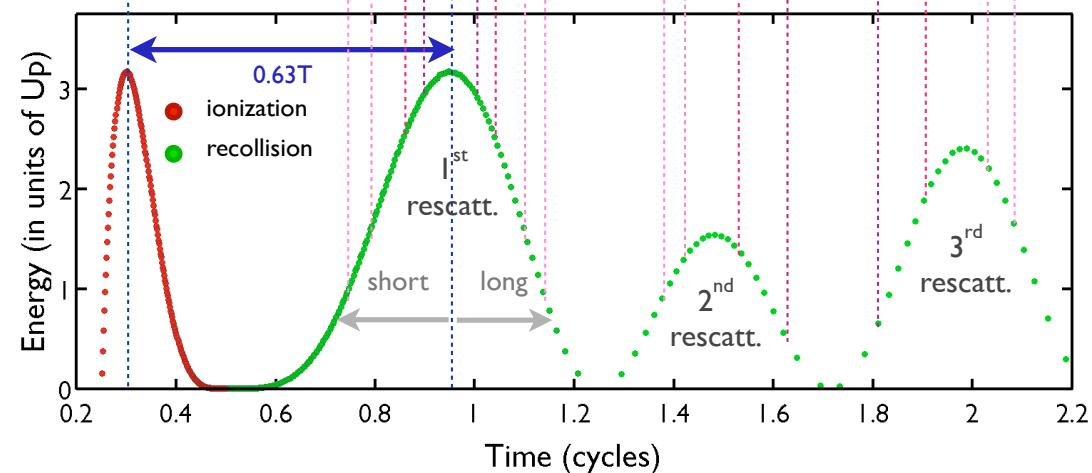
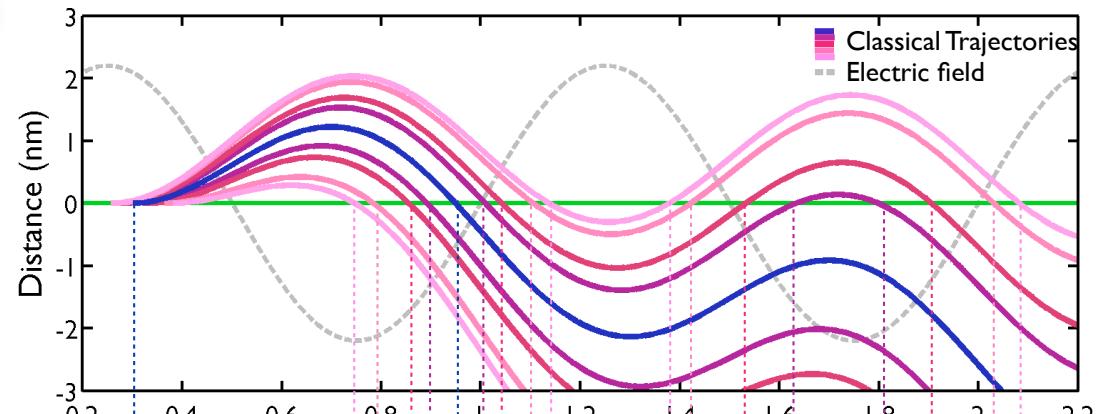
High-order Harmonic Generation



Cut-off

$$\hbar\omega_{\max} \simeq I_p + 3.17U_p \propto I\lambda^2$$

$$U_p = \frac{E_0^2}{4\omega_0^2}$$



Macroscopic Phase-matching

Macroscopic phase matching

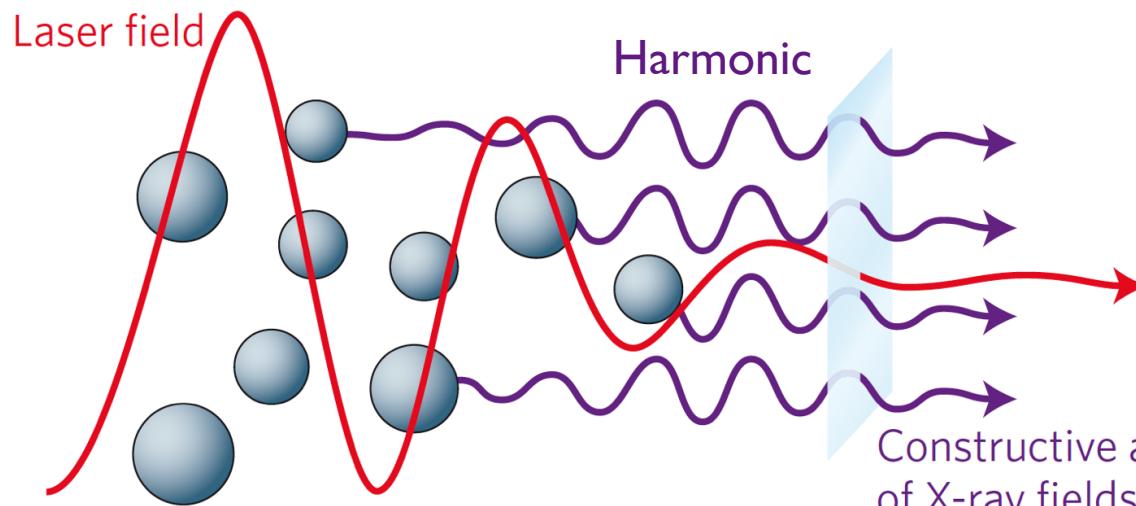


Figure courtesy of
T. Popmintchev

Phase mismatch function: $\Delta k_q = k_q - qk_1$

Perfect
phase-matching
 $\Delta k_q = 0$

P. Salières, A. L'Huillier, and M. Lewenstein, PRL, 74, 3776 (1995)

P. Balcou, P. Salières, A. L'Huillier, and M. Lewenstein, PRA, 55, 3204 (1996)

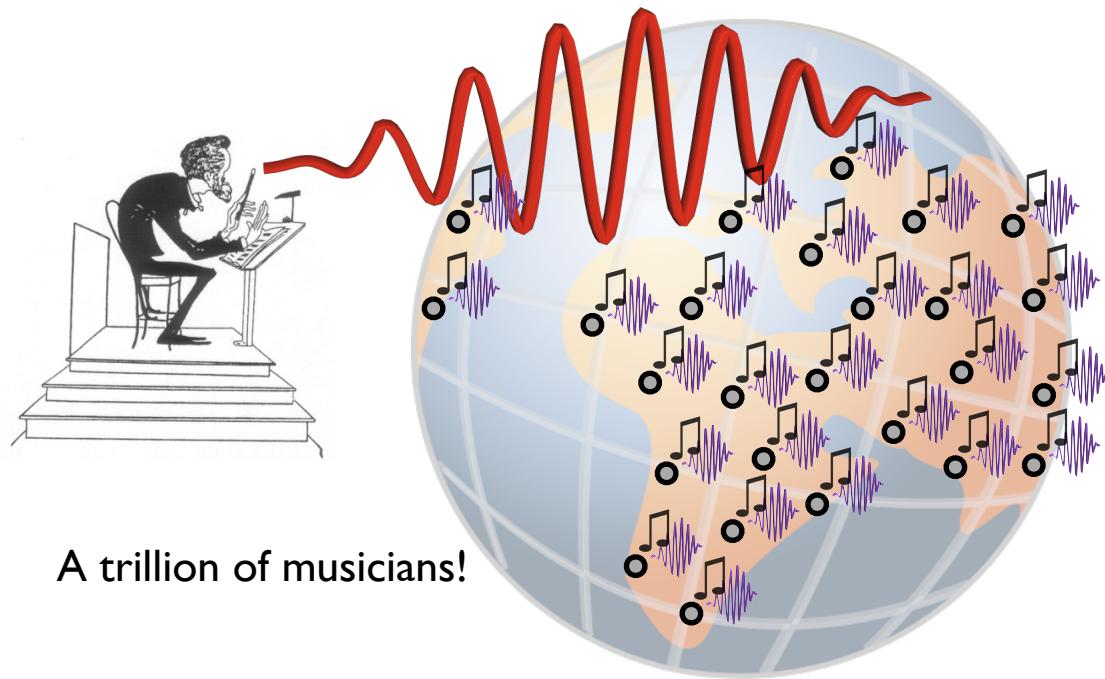
A. Rundquist et al, Science 280, 1412 (1998)

M. B. Gaarde, J. L. Tate, and K.J. Schafer J. Phys. B: At. Mol. Opt. Phys. 41 (2008)

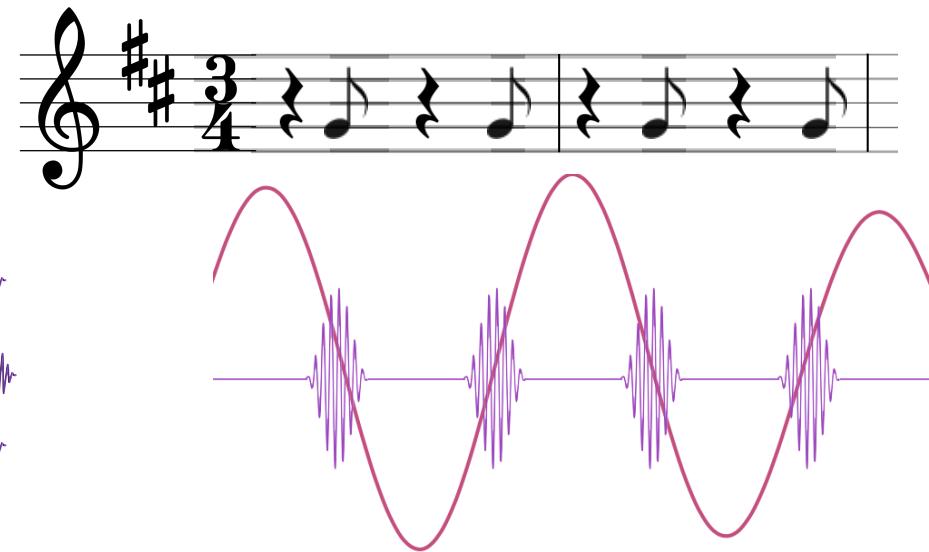
T. Popmintchev et al, PNAS 106, 10516 (2009)

The harmonic orchestra

Laser as the orchestra director...



... and each atom of the gas as a musician, emitting
attosecond pulses synchronized with the rhythm
dictated by the laser



As a result, a melody of attosecond pulses
is emitted coherently.

Outline

*'Generation of keV harmonics and ultrashort waveforms
driven by midinfrared lasers'*

- Microscopic HHG: single-atom
 - Method: SFA+
 - Multiple rescatterings to produce zeptosecond waveforms
- Macroscopic HHG: phase-matching
 - Method: Discrete Dipole Approximation
 - Time-gated phase-matching: isolating X-ray attosecond pulses
- Conclusions

Microscopic HHG

Single-atom



Quantum description of HHG

Harmonic emission spectrum is proportional to the power spectrum of dipole acceleration

$$I(\omega) \propto |\vec{a}_d(\omega)|^2 \quad \vec{a}_d(\omega) = \int \vec{a}_d(t) e^{i\omega t} dt$$

$$\vec{a}_d(t) = \frac{1}{m} \langle \psi(t) | \vec{\nabla} V + \vec{E} | \psi(t) \rangle$$

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = H(t) |\psi(t)\rangle$$
$$H = \frac{\vec{p}^2}{2m} + V + e\vec{E} \cdot \vec{r}$$

Exact solution: Time Dependent Schrödinger Equation (TDSE) for $\psi(t)$

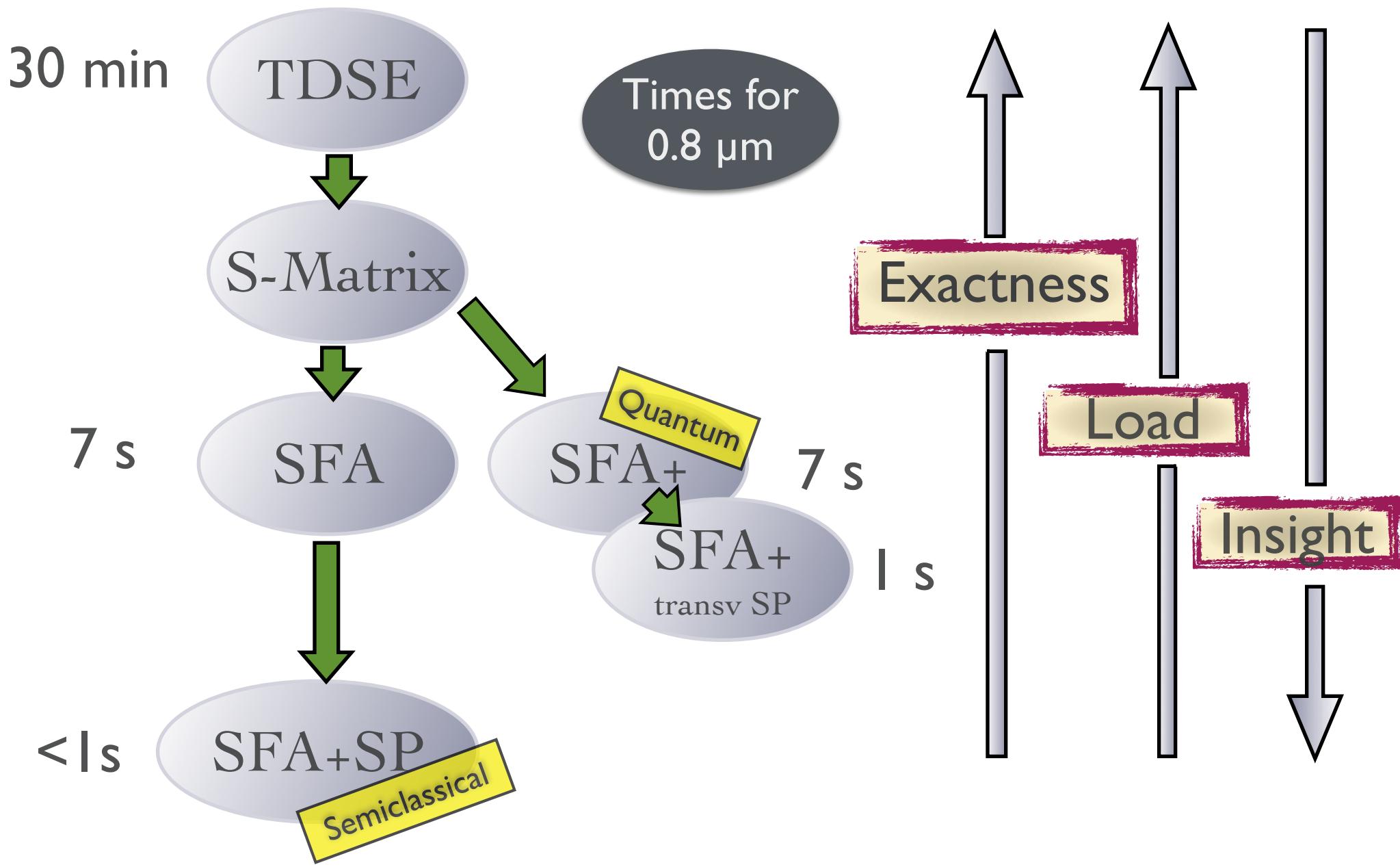
TDSE is very challenging when:

1. Considering phase-matching (involves lot of single-atom calculations)
2. Long wavelengths (huge spatio-temporal grid)

We have to use approximations:

- ✓ The standard way: SFA and saddle point M. Lewenstein et al. PRA 49, 2117 (1994)
- ✓ Our choice: SFA+ J.A. Pérez-Hernández, et al, Opt. Express 17, 9891 (2009)

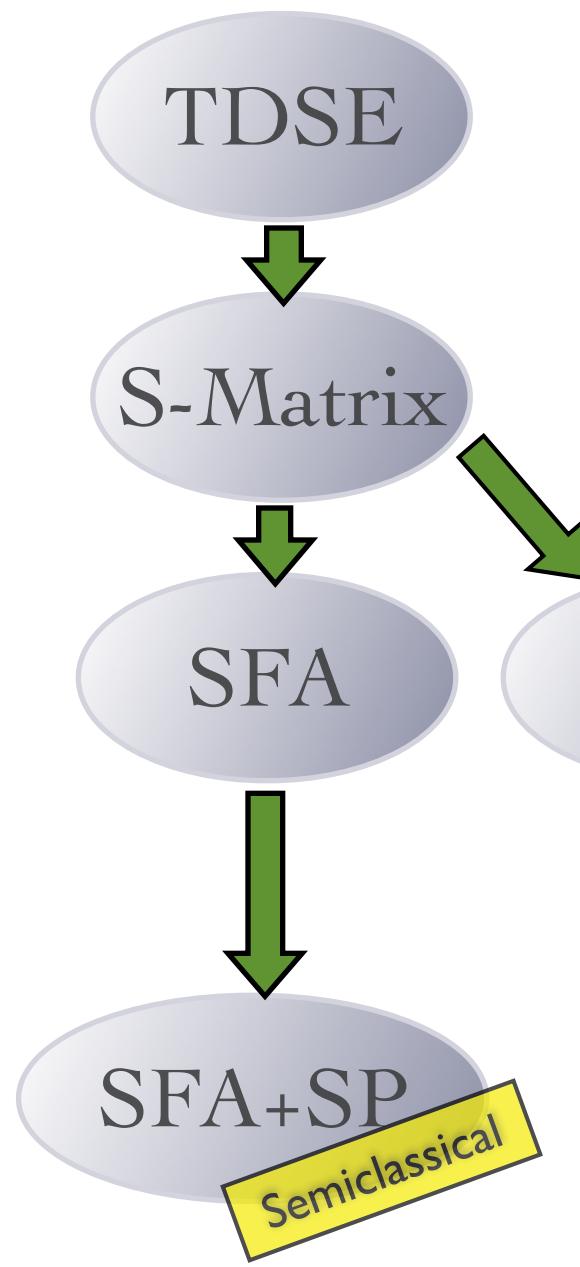
Strategies for computing HHG



Strategies for computing HHG

I

I/2



Times for
2 μm

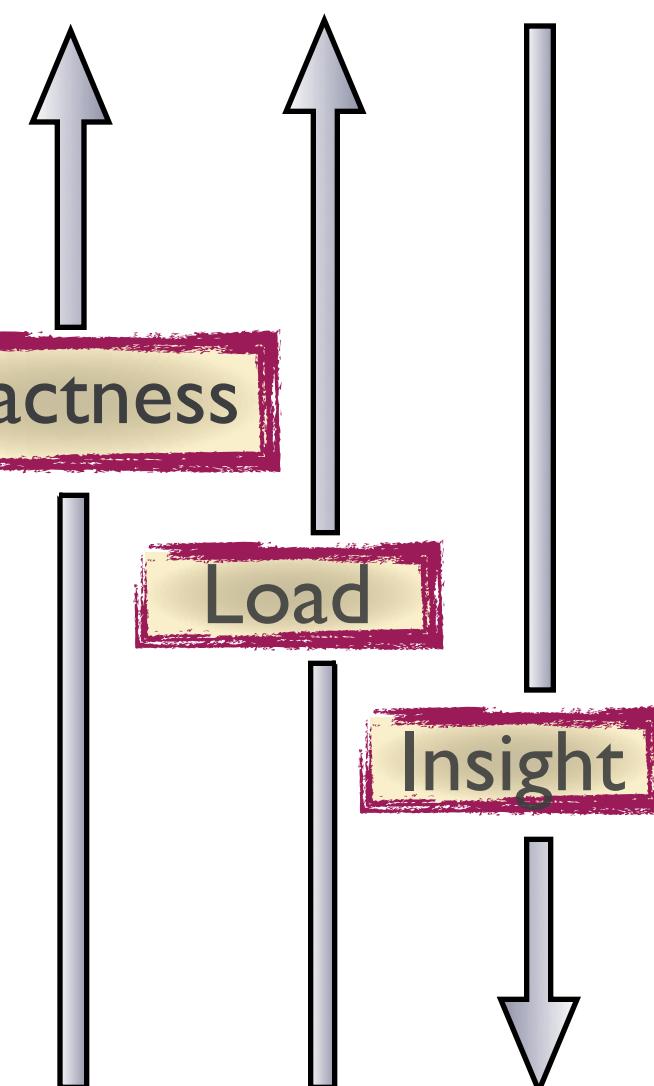
I/2 day

20 min

Exactness

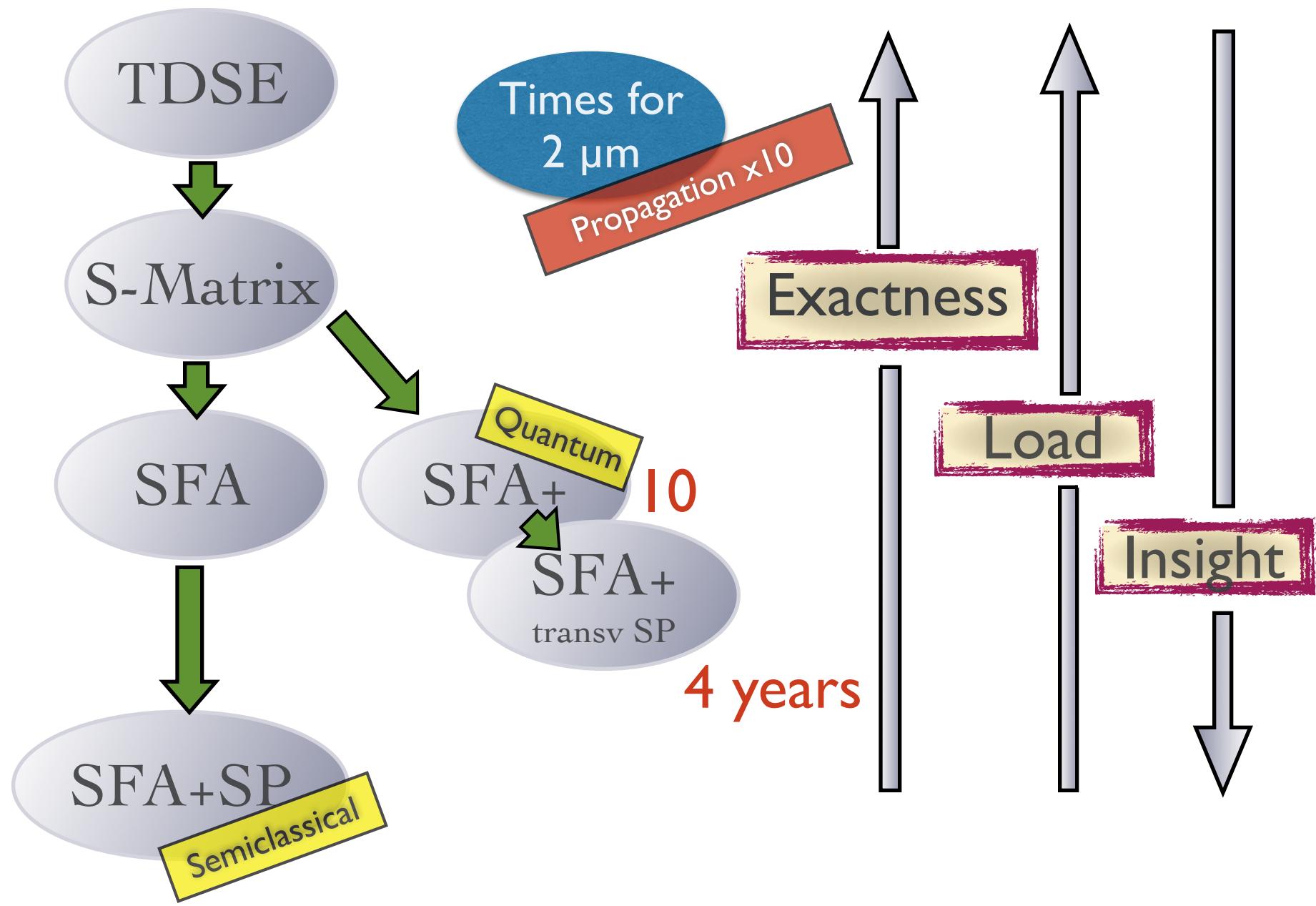
Load

Insight

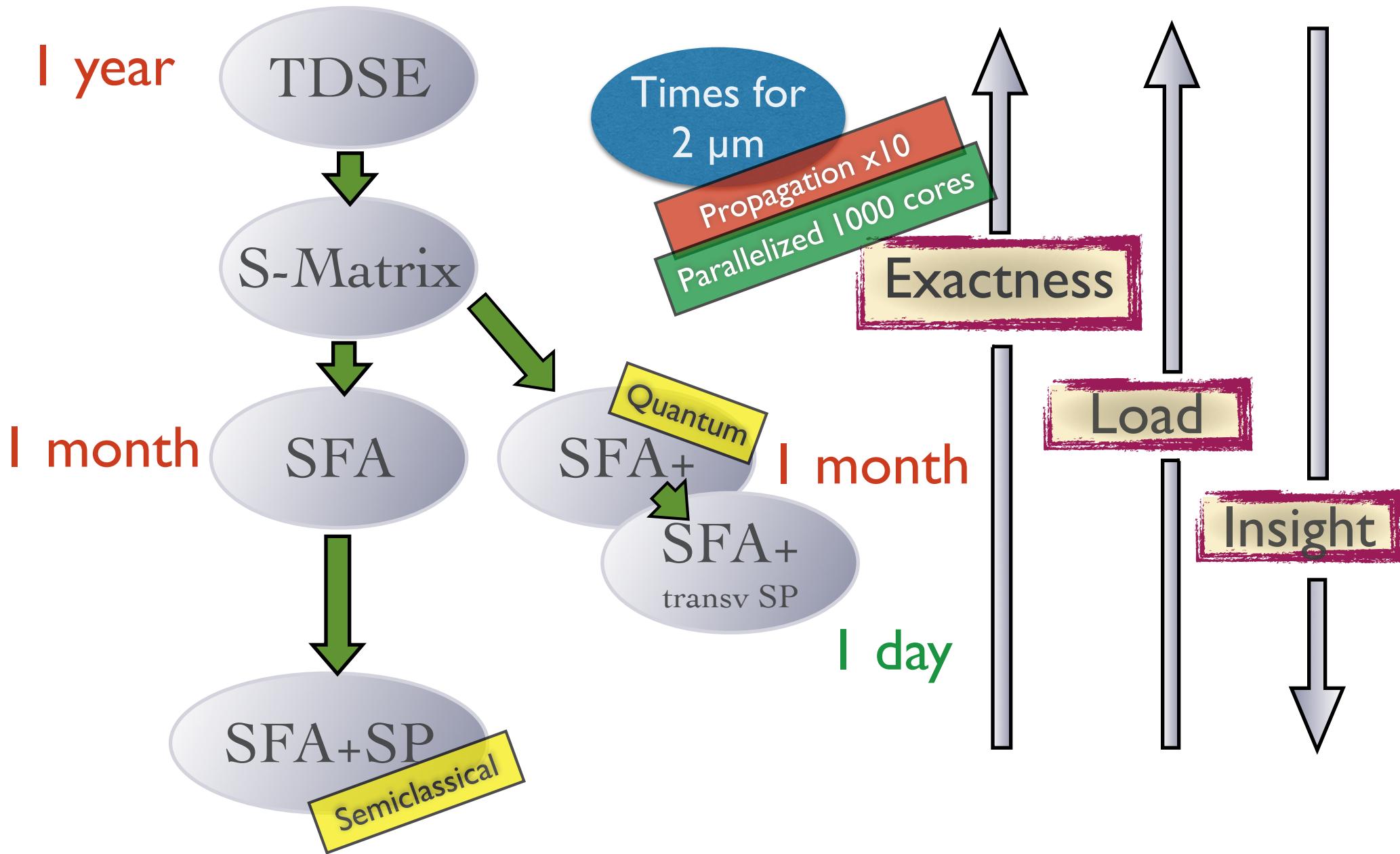


Strategies for computing HHG

10



Strategies for computing HHG



Principles of SFA+

Strong Field Approximation (SFA)

1. Considering the electrons promoted to the continuum to have no possibility to recombine.
2. Neglecting the bound-state excitations (field-dressing).
3. Considering the electrons as free particles in the continuum.

$$a(\mathbf{k}, t) = -\frac{i}{\hbar} C_F \int_{t_0}^t dt_1 \langle \phi_0 | \hat{a} | \mathbf{k} \rangle e^{-\frac{i}{\hbar} S(\mathbf{k}, t, t_1)} V_i(\mathbf{k}, t_1) \langle \mathbf{k} | r^{-n} | \phi_0 \rangle$$

Transition back to
ground state Transition into the
continuum at time t_1
 Evolution as a free electron

$$S(\mathbf{k}, t, t_1) = \int_{t_1}^t [\epsilon(\mathbf{k}, \tau) - \epsilon_0] d\tau$$

$$V_i(t) = -(q/mc)A(t)p_z + q^2/(2mc^2)A^2(t)$$

$$\epsilon(\mathbf{k}, \tau) = \hbar^2 k^2/2m - (\hbar q/mc)A(\tau)k_z + q^2 A^2(\tau)/(2mc^2)$$

Extended Field Approximation (SFA+)

To include the possibility of atomic bound-state excitations (field-dressing).

$$a(t) = a_b(t) + a_d(t) + c.c.$$

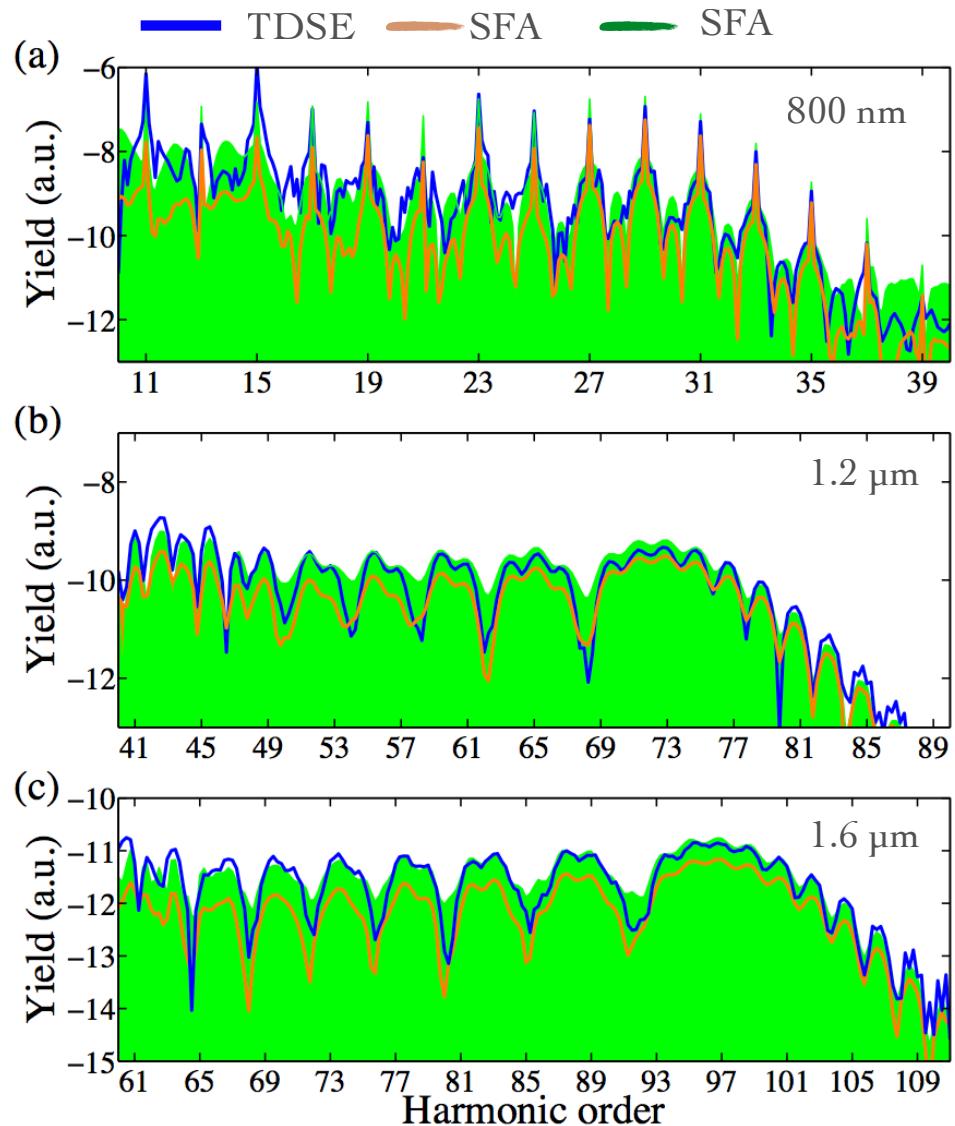
$$a_d(\mathbf{k}, t) \simeq - \left[1 + \frac{k^2/2m - \epsilon_0}{\Delta_s} \right] a_b(\mathbf{k}, t)$$

$$\Delta_s = \langle V_i(t) \rangle = (1/\delta t_s) \int_{t-\delta t_s}^t [-(q\hbar/mc)A(\tau)k_z + (q^2/2mc^2)A^2(\tau)] d\tau$$

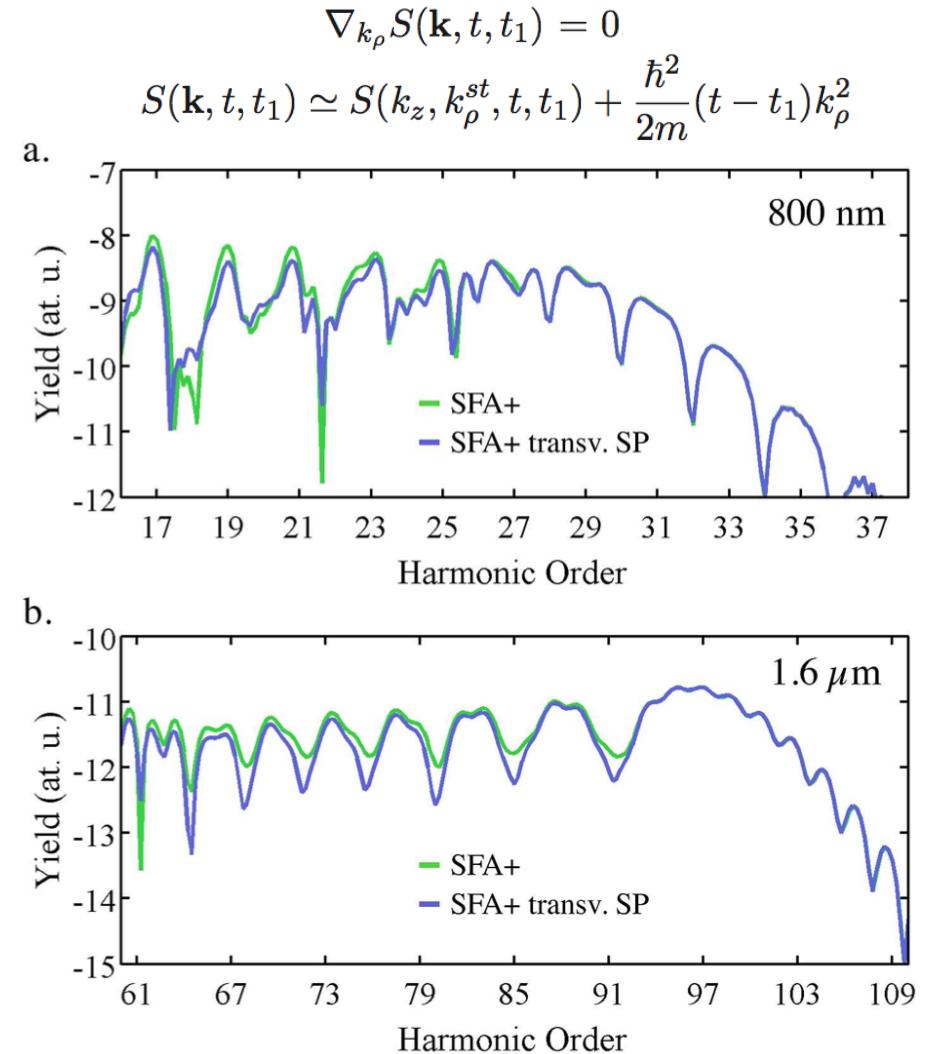
J.A. Pérez-Hernández, L. Roso and L. Plaja, Opt. Express 17, 9891 (2009)

J.A. Pérez-Hernández, C. Hernández-García, J. Ramos, E. Conejero Jarque, L. Plaja, and L. Roso, Springer Series in Chemical Physics Vol. 100 (Springer, New York, 2011), Chap. 7, p. 145

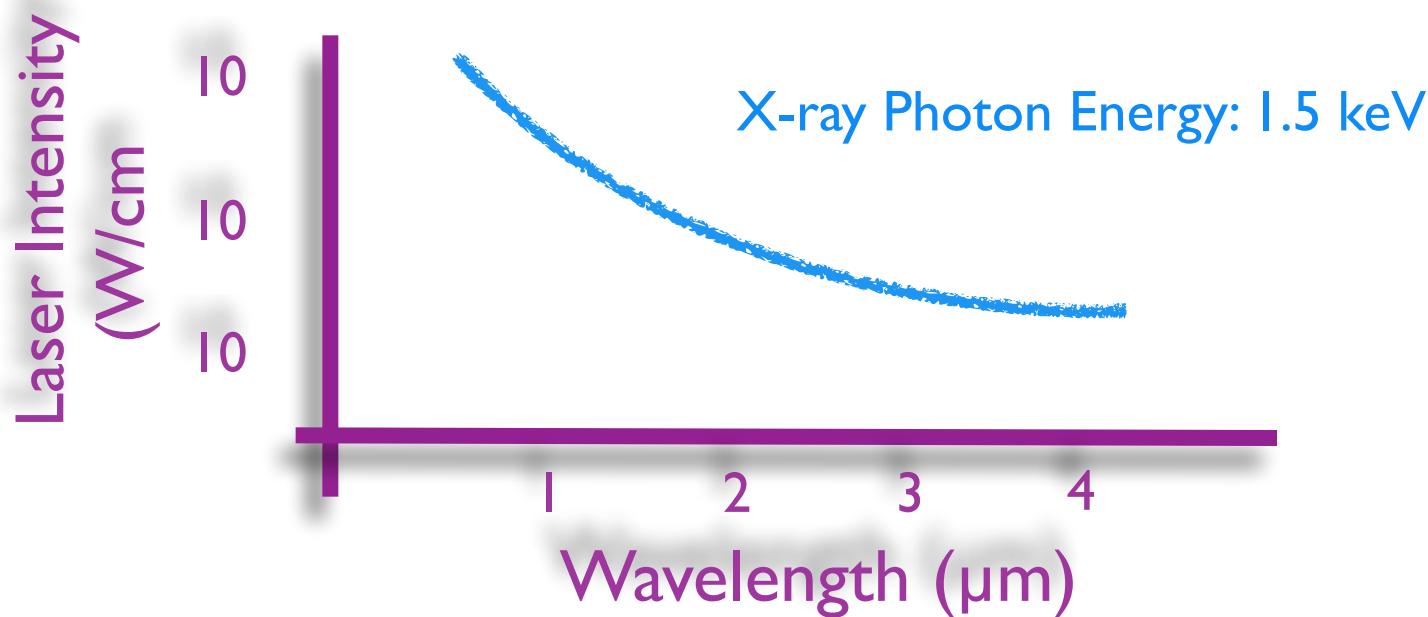
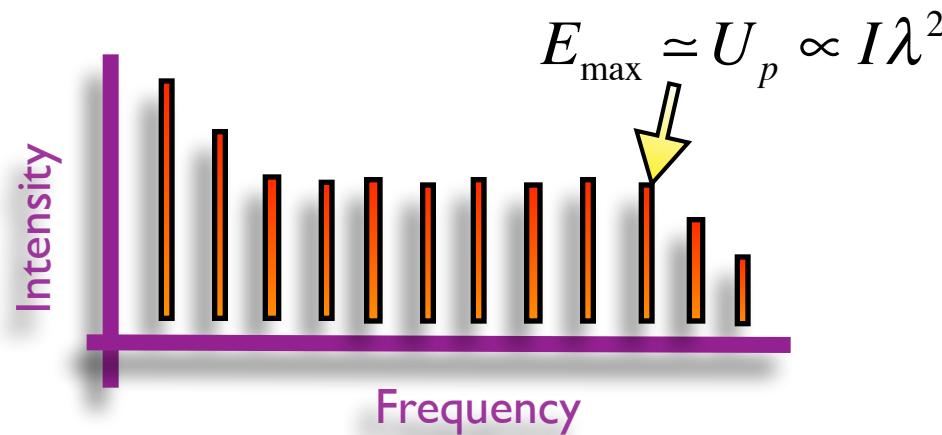
Principles of SFA+



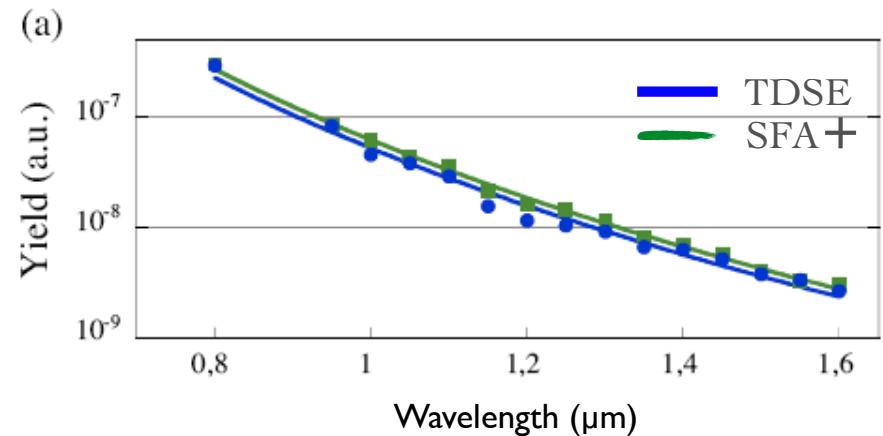
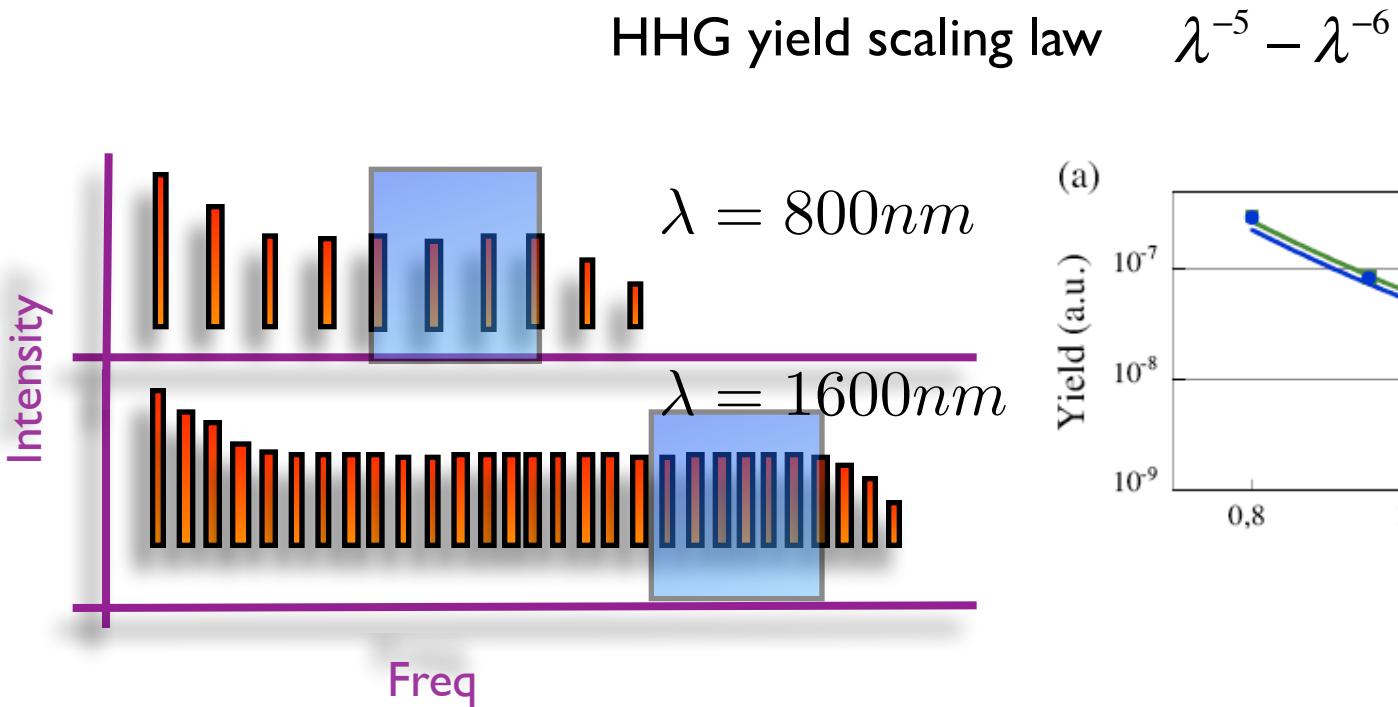
SFA+ with transversal saddle-point



Obtaining X-rays from HHG

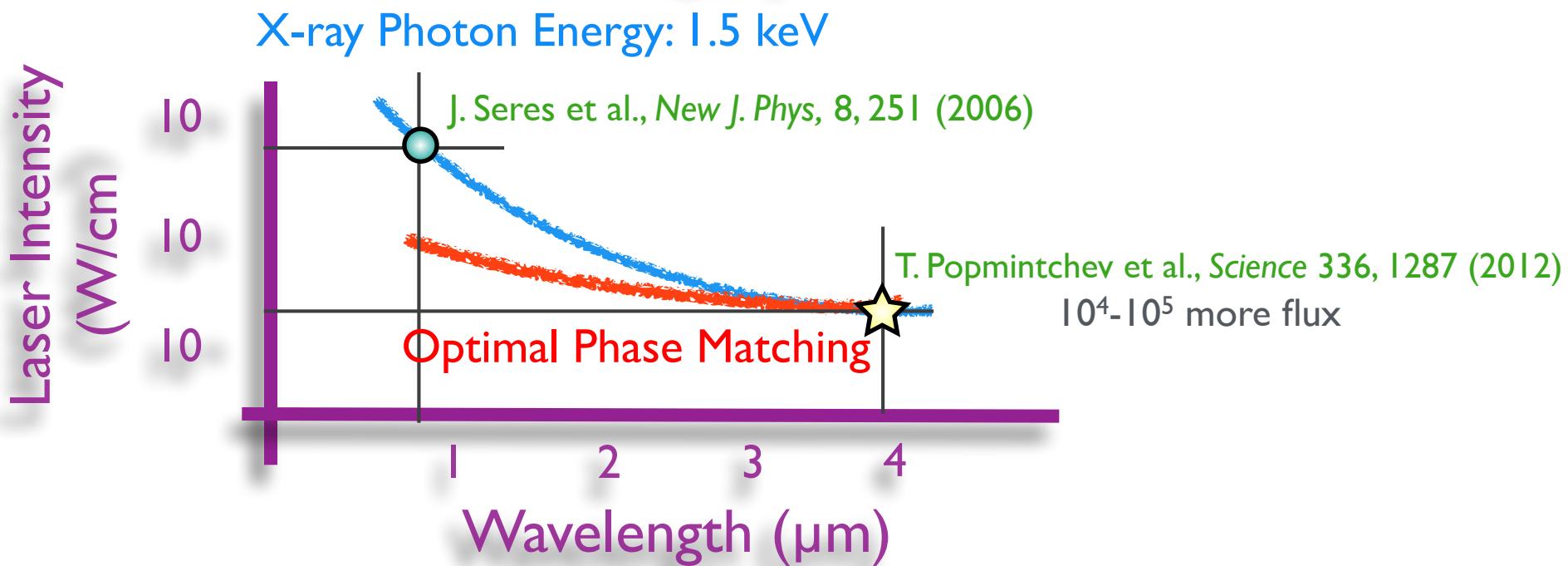
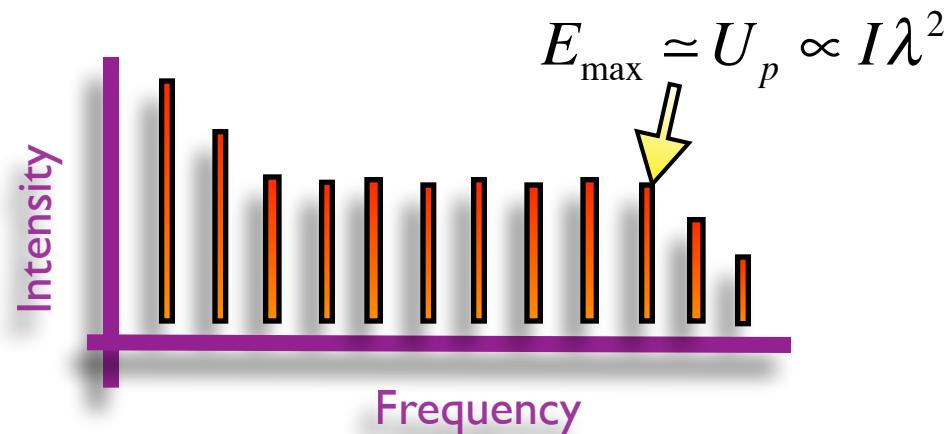


Unfavorable yield scaling law with wavelength



- J. L. Tate, T. Auguste, H. G. Muller, P. Salières, P. Agostini, and L. F. DiMauro, PRL 98, 013901 (2007)
K. Schiessl, K.L. Ishikawa, E. Person, and J. Burgdörfer, PRL 99, 253903 (2007)
M.V. Frolov, N.L. Manakov, and A. Starace, PRL 100, 173001 (2008)
J.A. Pérez-Hernández, L. Roso and L. Plaja, Opt. Express 17, 9891 (2009)

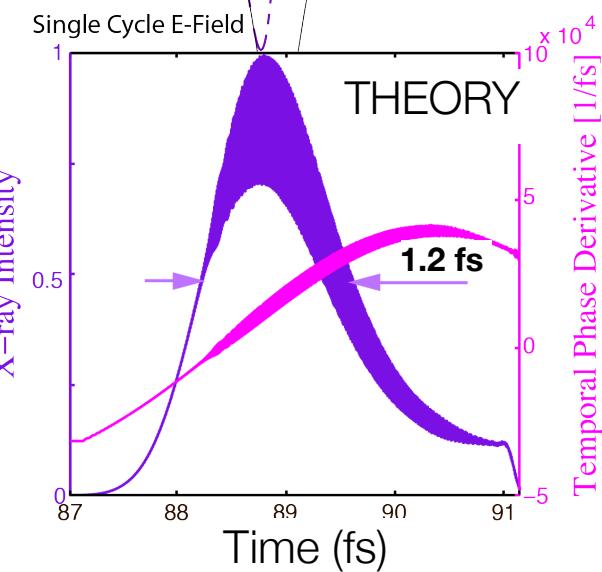
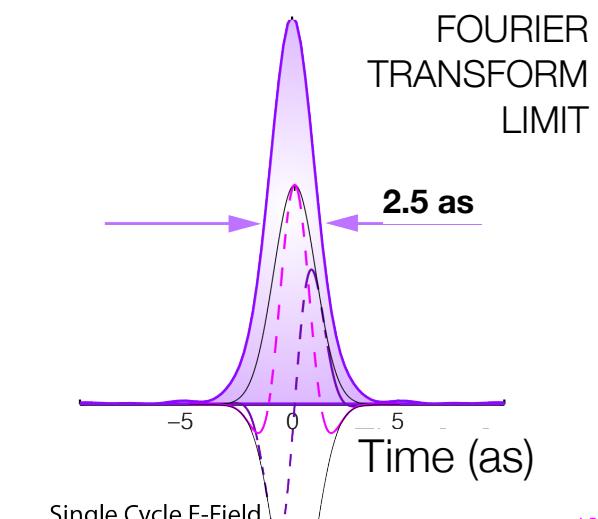
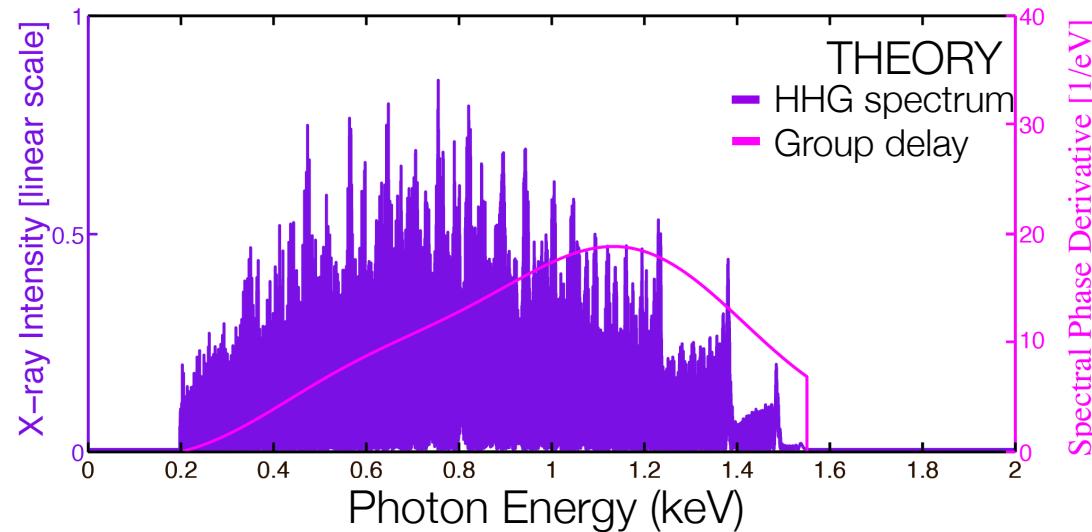
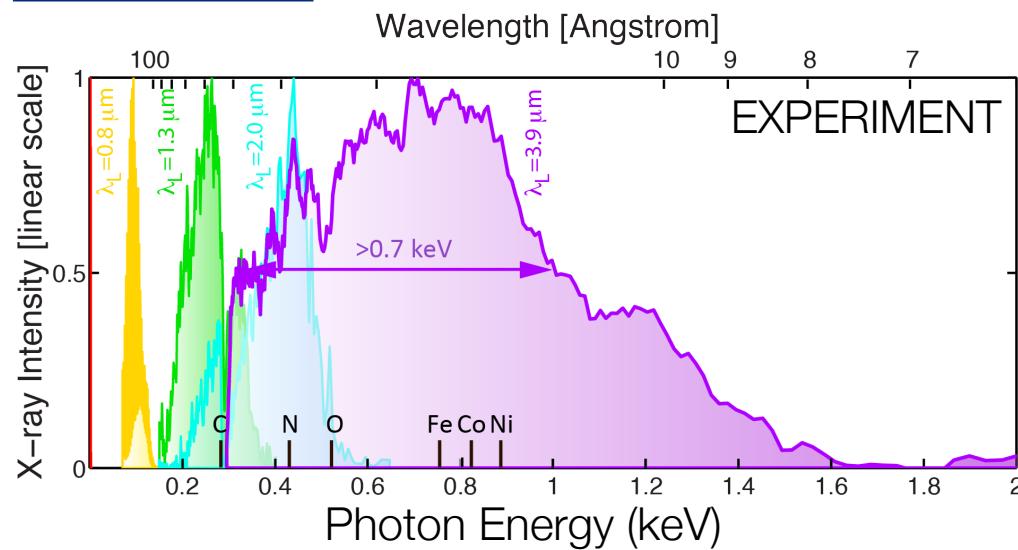
Obtaining X-rays from HHG



Phase-matched X-rays



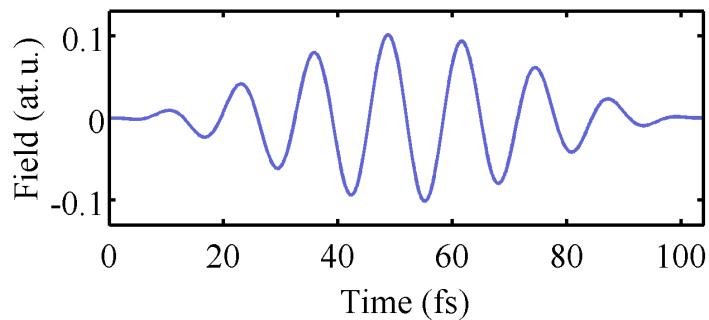
Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers
 Tenio Popmintchev *et al.*
Science **336**, 1287 (2012);
 DOI: 10.1126/science.1218497



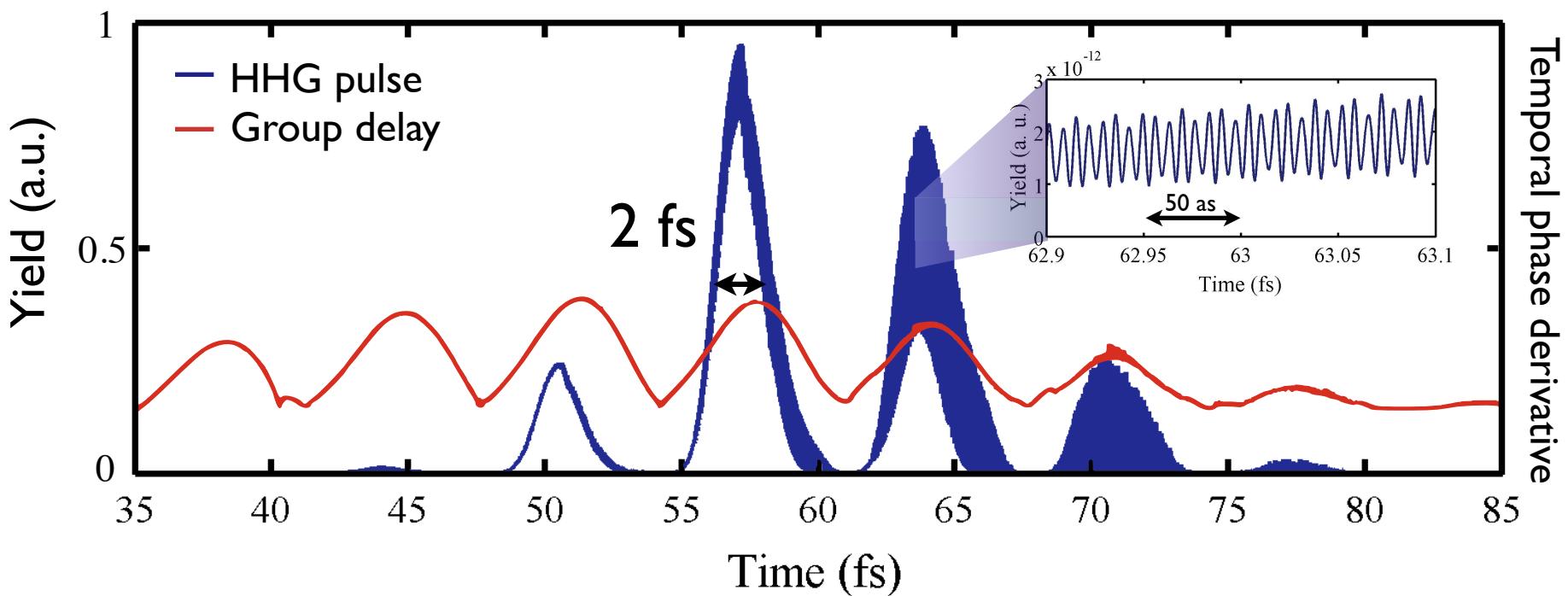
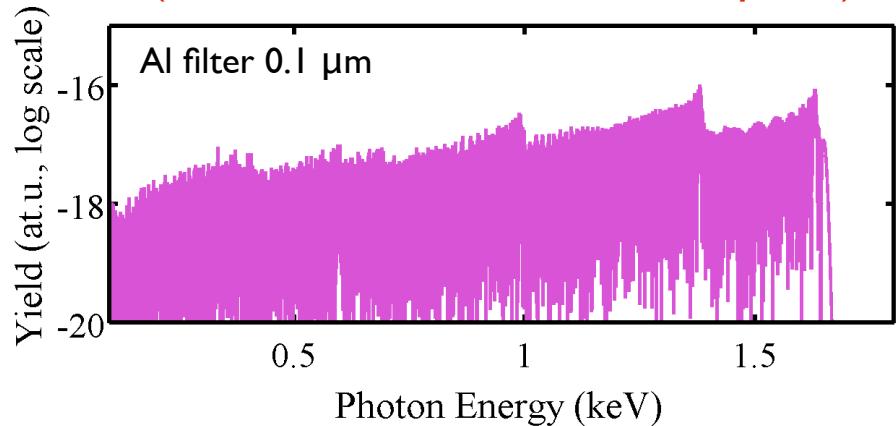
Single atom calculations at 4 μm

$$\lambda = 3.9 \mu\text{m}$$

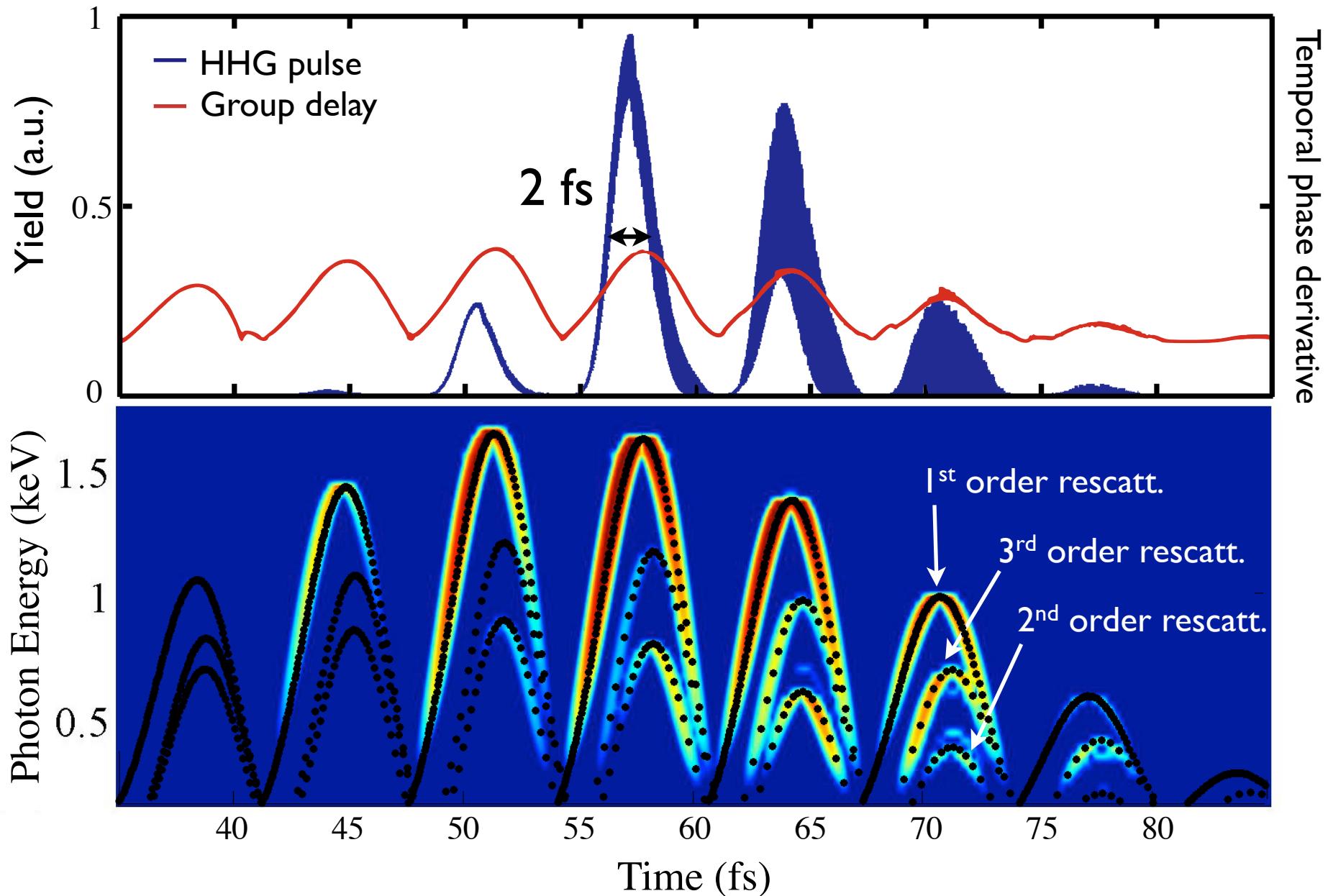
Laser field: $I = 3.6 \times 10^{14} \text{ W/cm}^2$
3 cycles (38 fs)



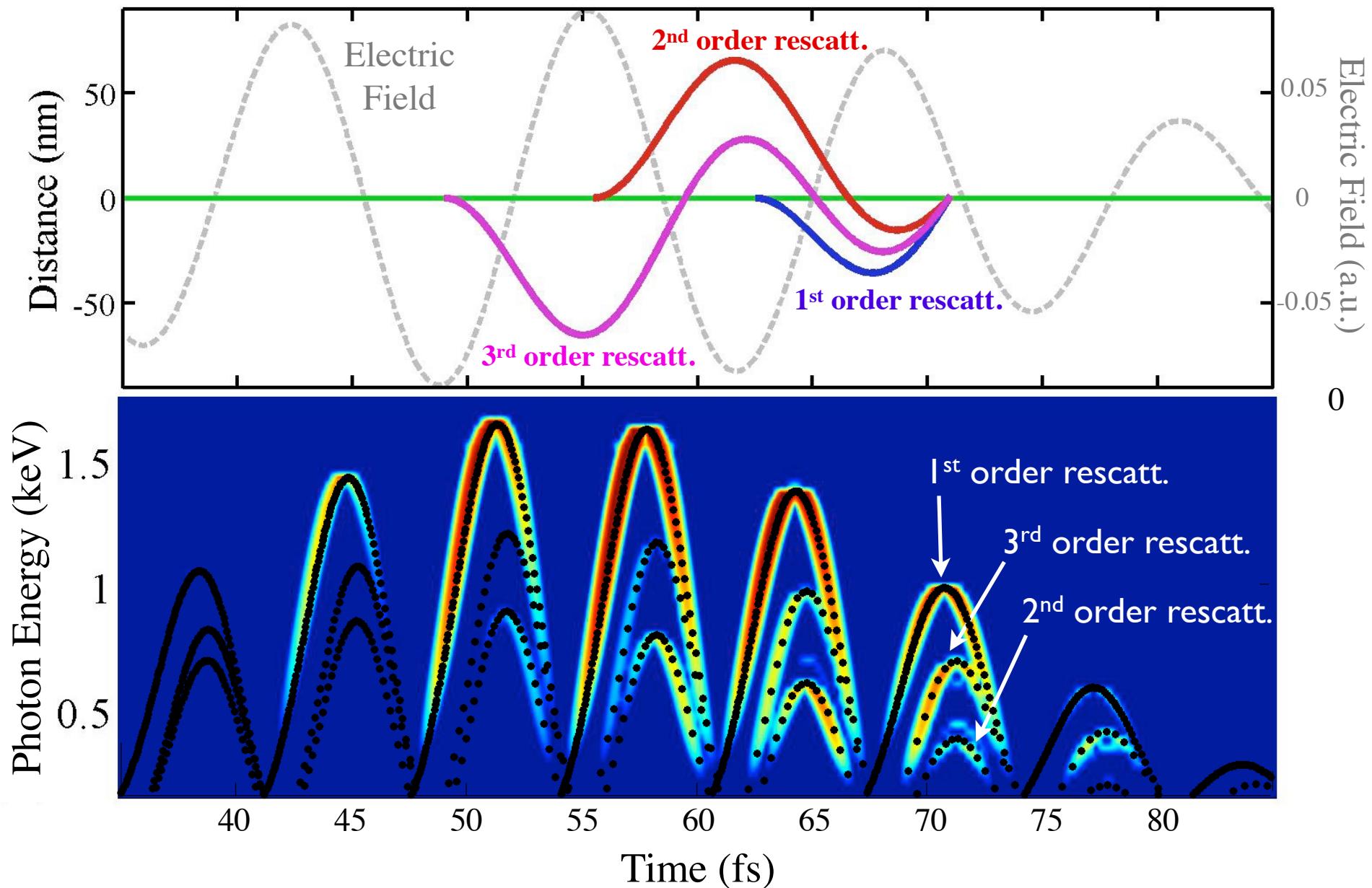
HHG spectrum
(SFA+ and transversal saddle point)



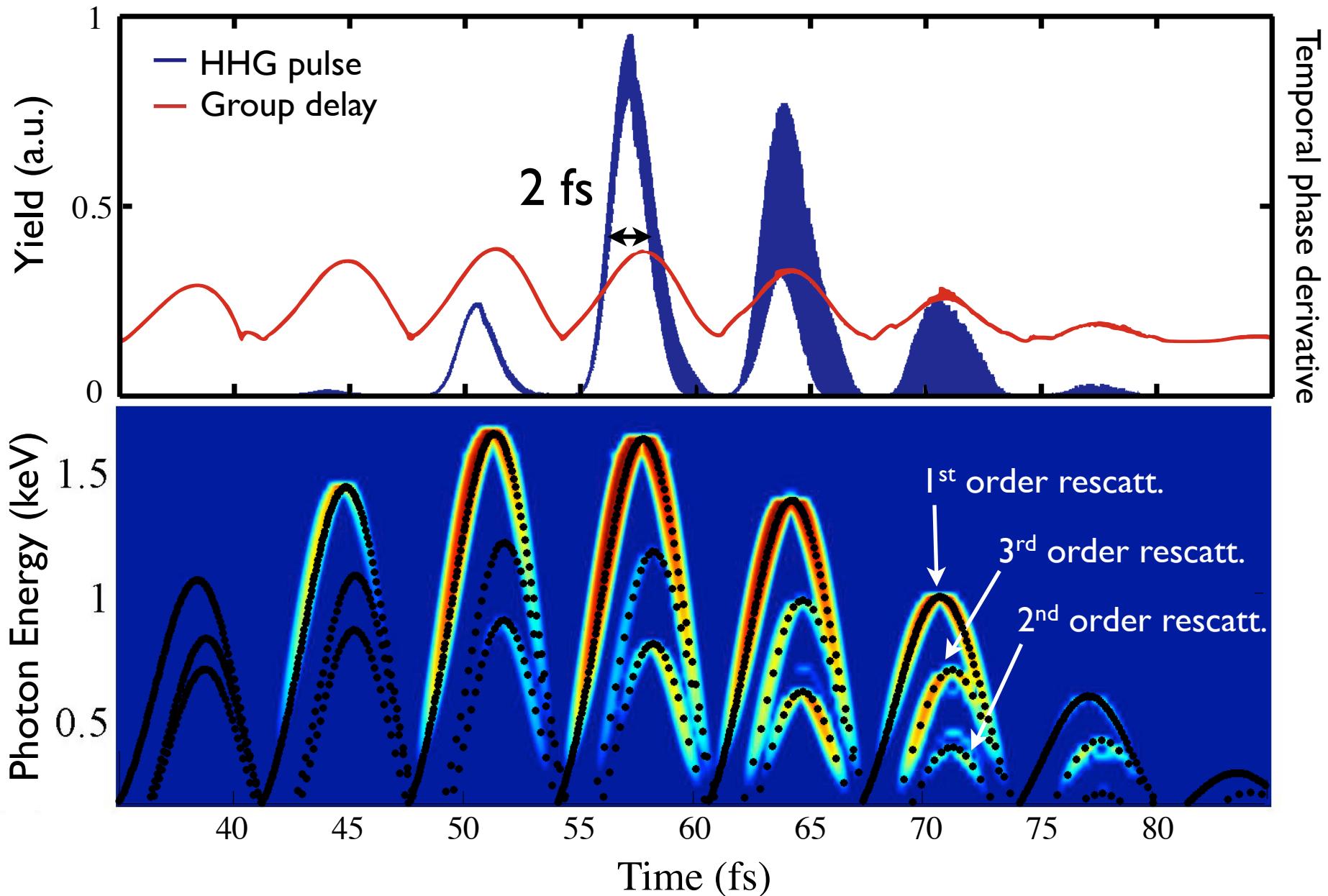
High-order rescattering effects



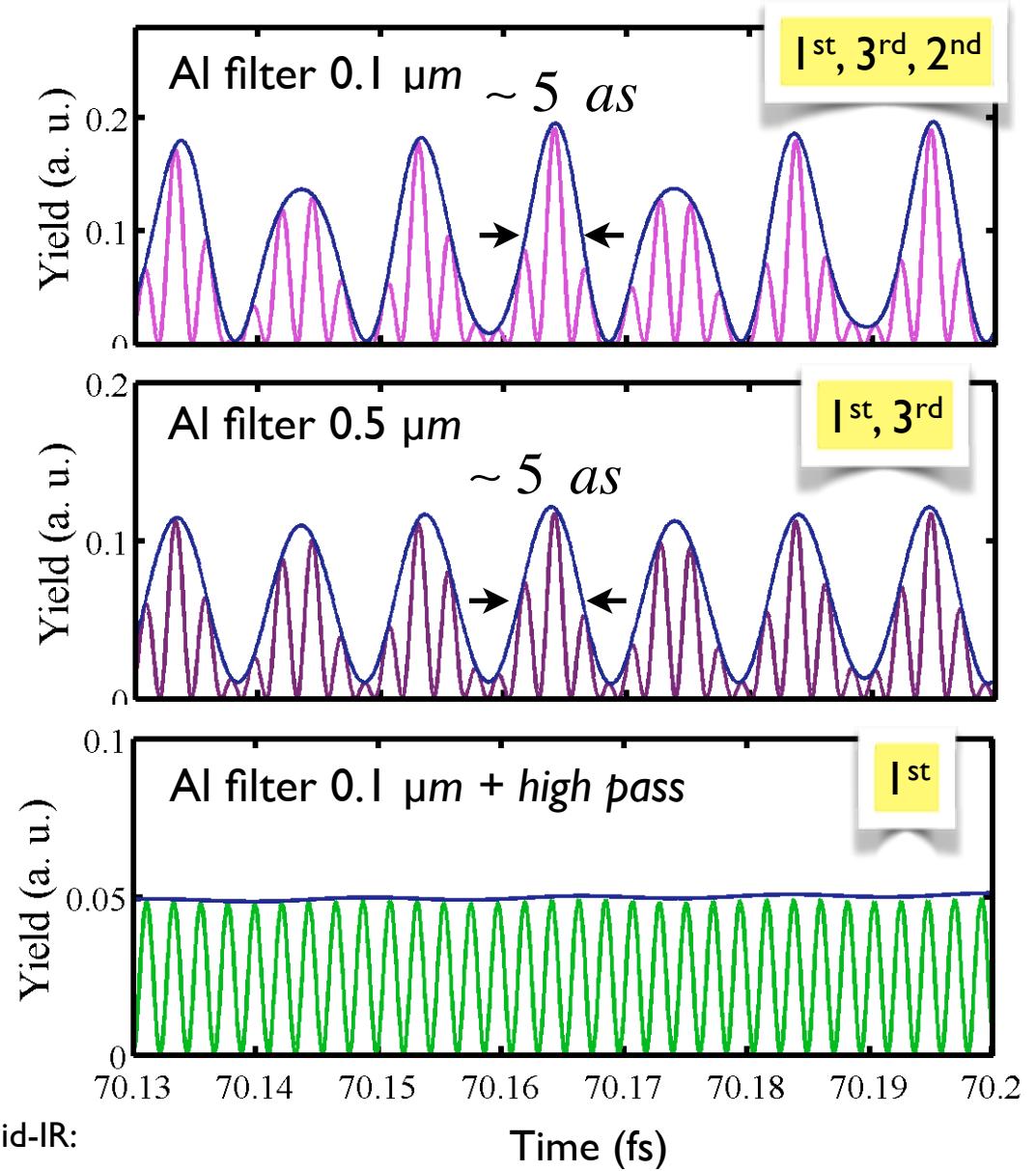
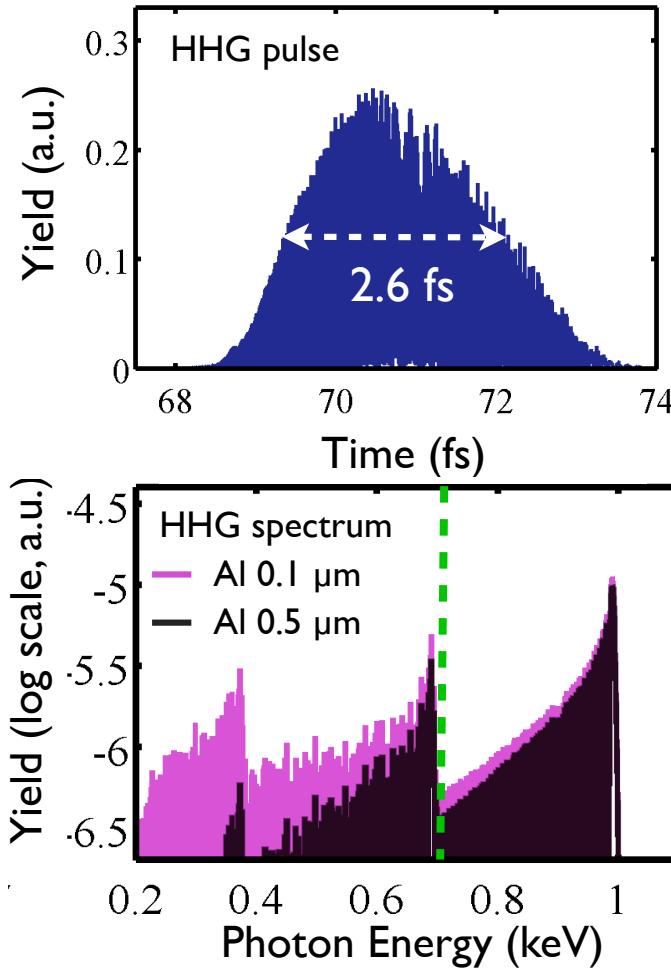
High-order rescattering effects



High-order rescattering effects



Attosecond pulse trains in the keV



C. Hernández-García, J.A. Pérez-Hernández, T. Popmintchev, M. Murnane, H. Kapteyn, A. Jaron-Becker, A. Becker, and L. Plaja, PRL 111, 033002 (2013)

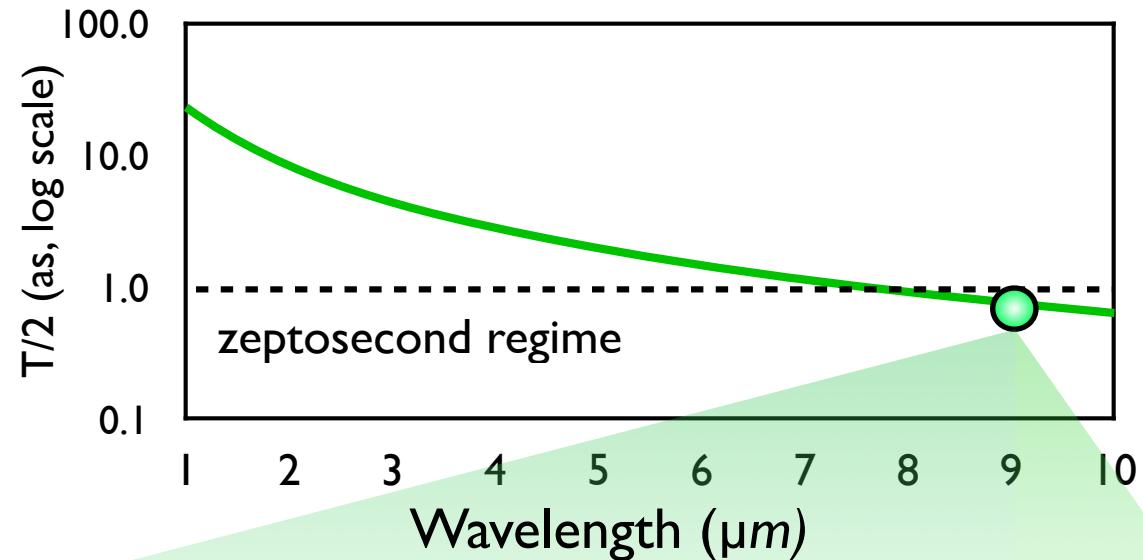
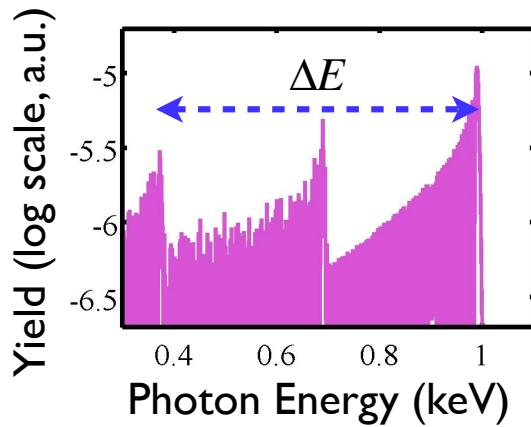
Previous works involving multiple rescatterings using mid-IR:

J. L. Tate, et al. PRL 98, 013901 (2007)

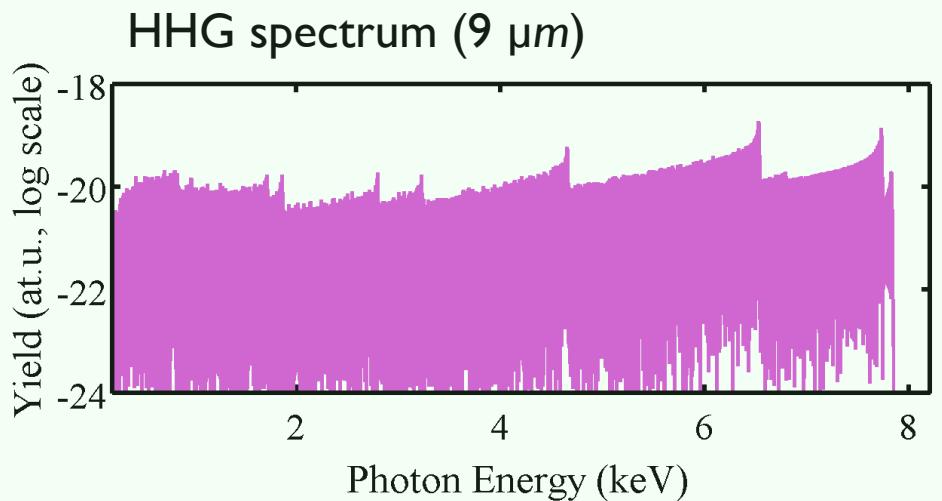
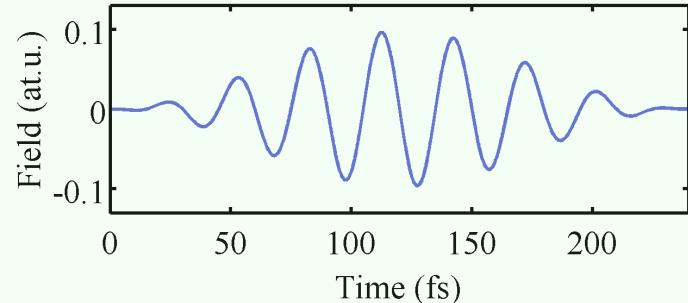
D. Hickstein et al. PRL 109, 073004 (2012)

Towards zeptosecond pulse trains

$$T = \frac{2\pi}{\Delta E}$$

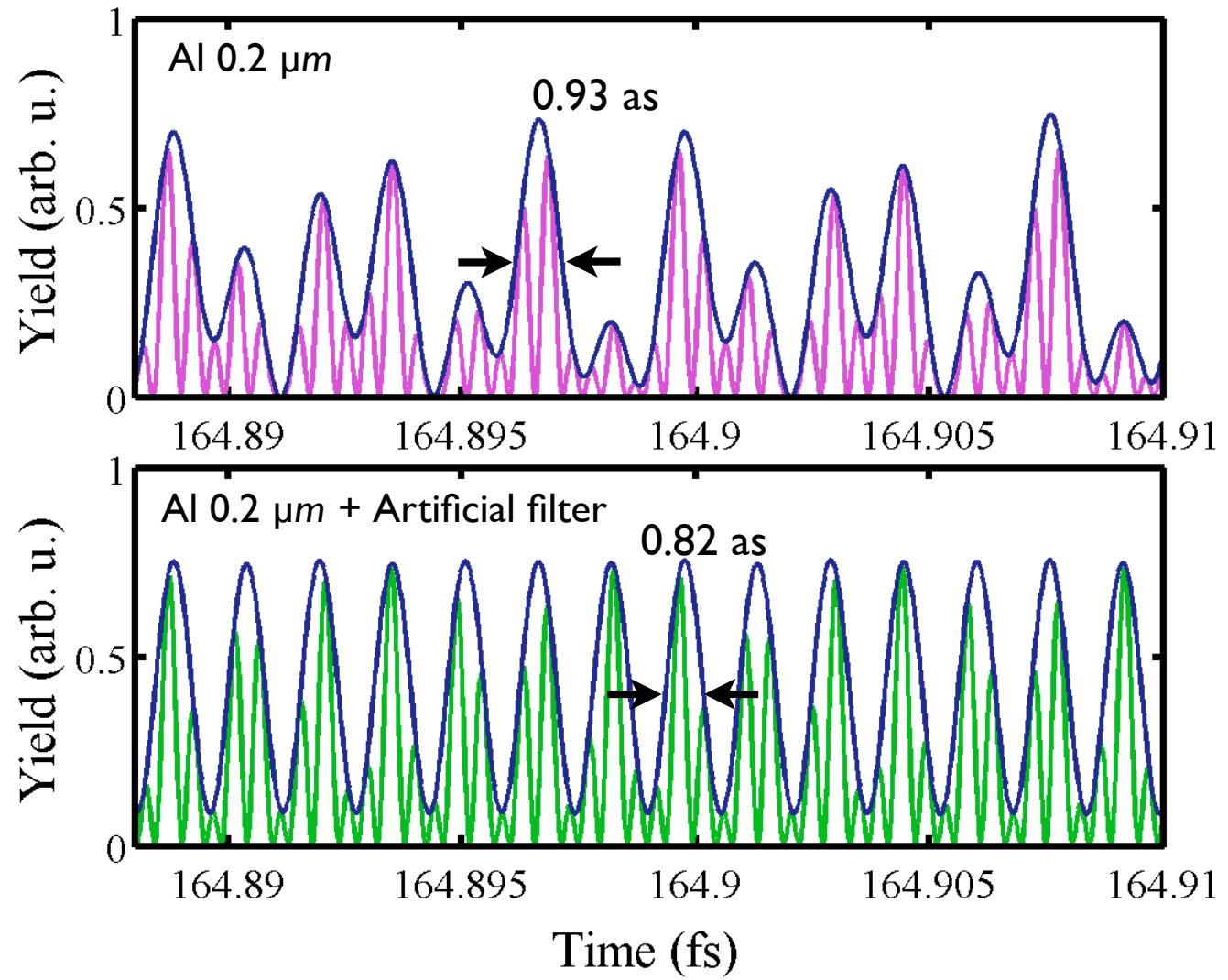
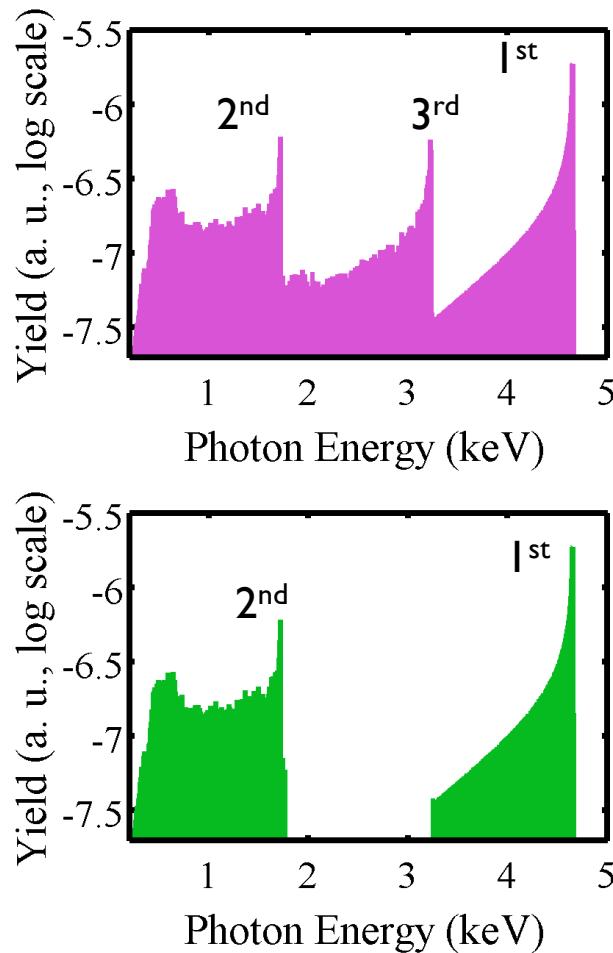


Laser field: $\lambda = 9 \mu\text{m}$
 $I = 3.16 \times 10^{14} \text{ W/cm}^2$
3 cycles (90 fs)



Towards zeptosecond pulse trains

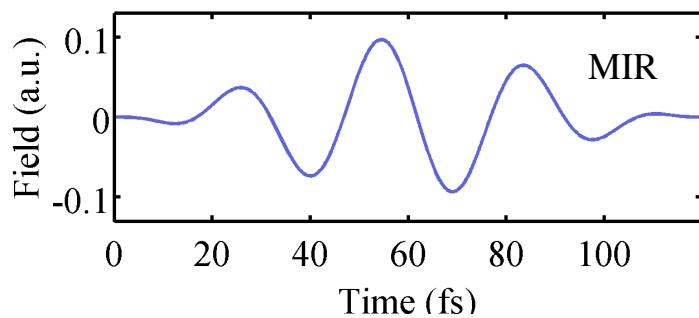
$\lambda = 9 \mu m$



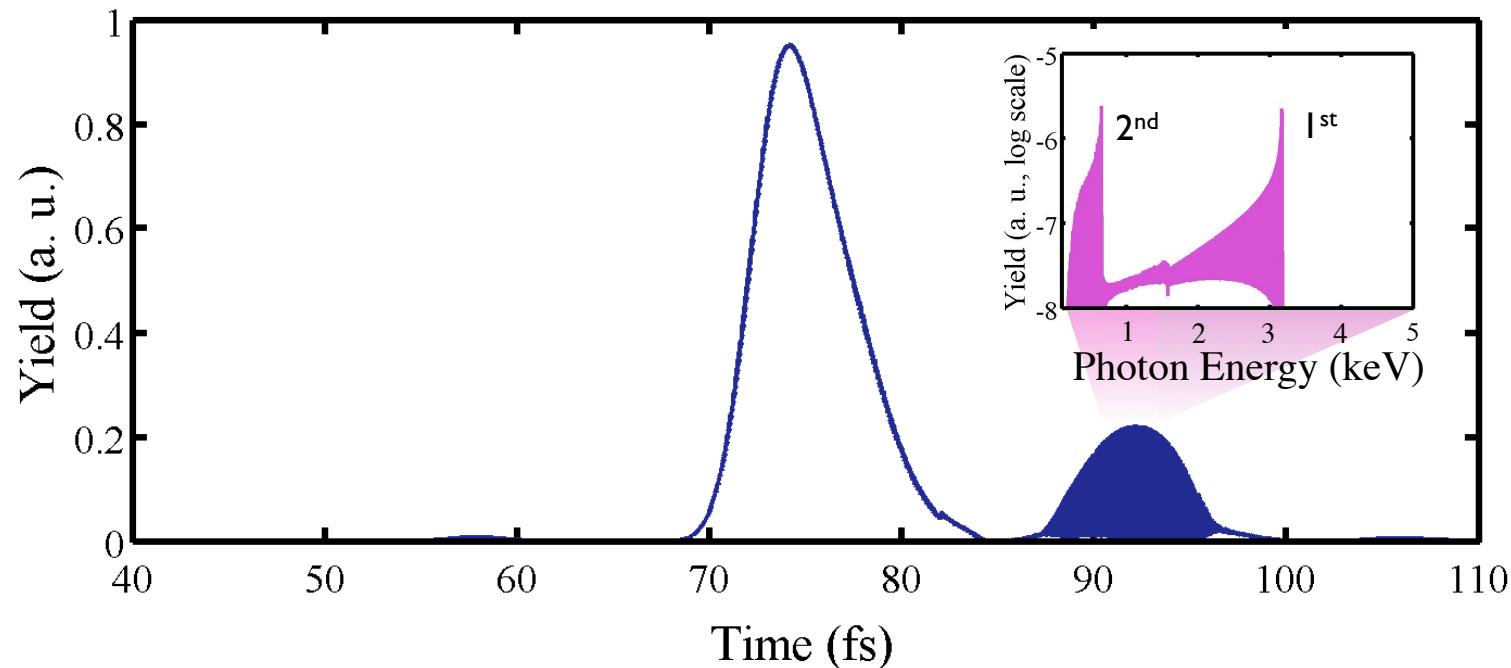
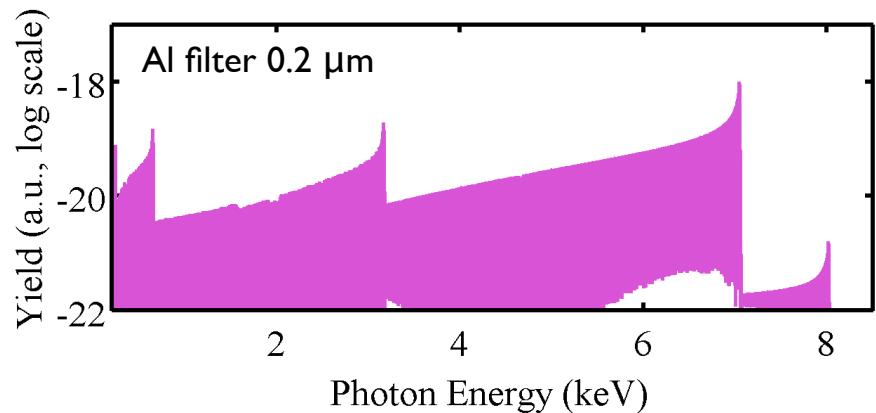
Zeptosecond waveforms

$$\lambda = 9 \text{ } \mu\text{m}$$

Laser field: $I = 3.4 \times 10^{14} \text{ W/cm}^2$
1.44 cycles (43fs)

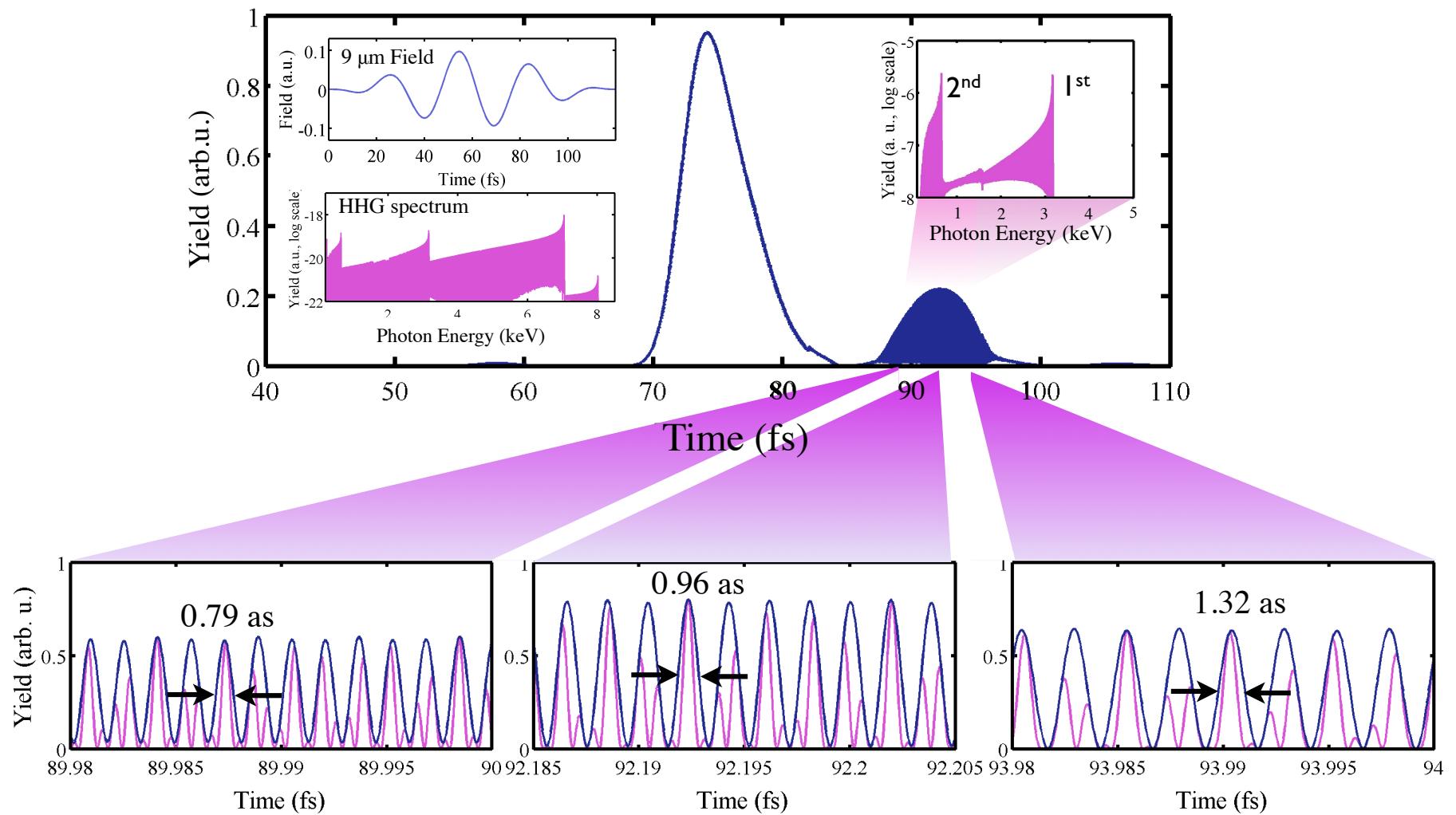


HHG spectrum



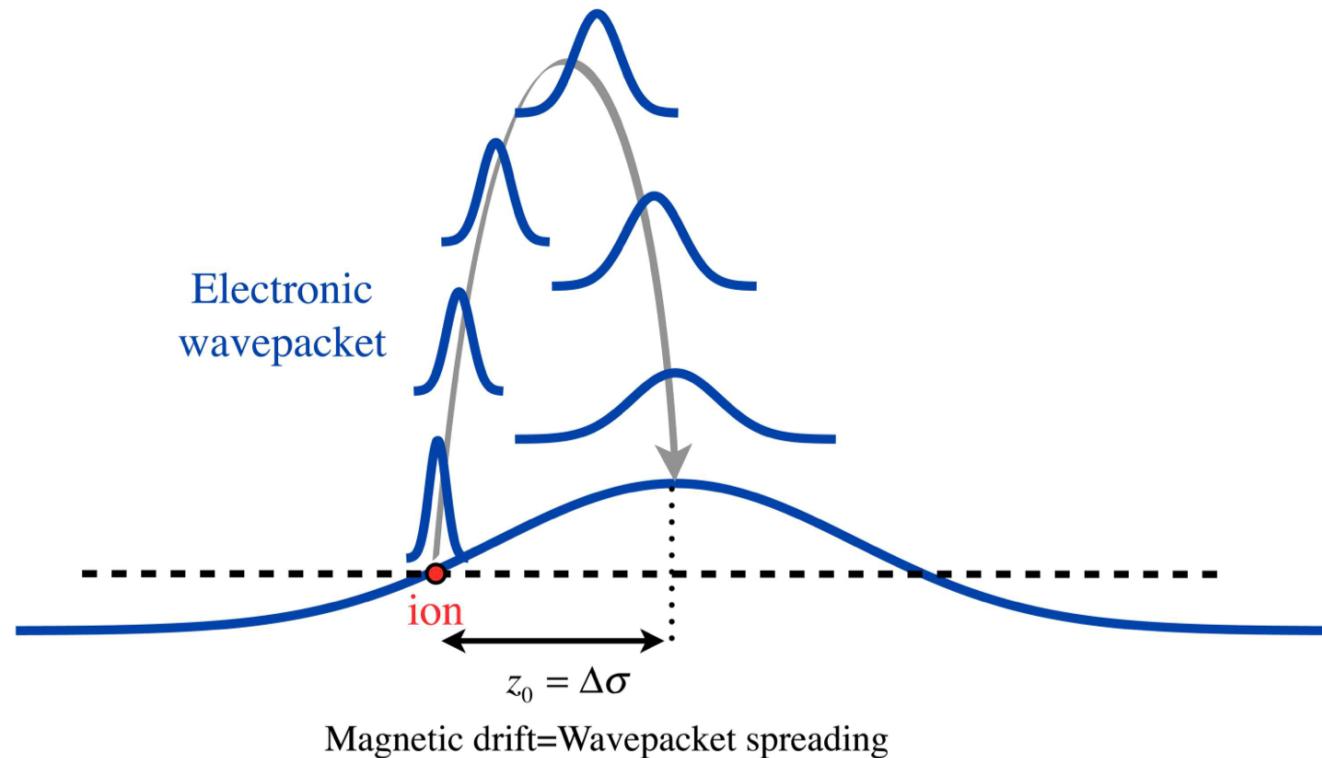
Zeptosecond waveforms

$\lambda = 9 \mu m$



Why not even longer wavelengths?

The magnetic drift mitigates the harmonic yield



The yield would drop by an order of magnitude at 10 μm

Macroscopic HHG

Phase-matching effects



Macroscopic Phase-matching

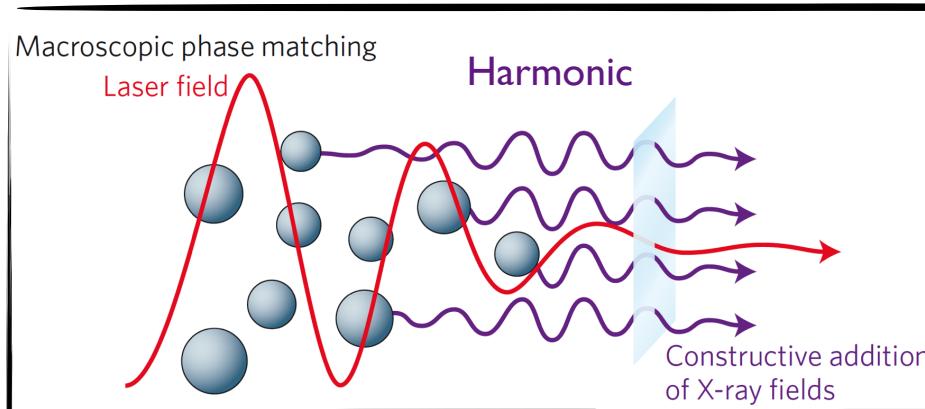


Figure courtesy of
T. Popmintchev

Electric field of the qth-order harmonic:

$$E_q(\mathbf{r}) \propto \left[|d_q^j(\mathbf{r})| e^{i\phi_q^j(\mathbf{r})} \right] e^{i(\mathbf{r}_d - \mathbf{r})\mathbf{k}_q}$$

$|d_q^j(\mathbf{r})|$ spectral amplitude of the single-atom dipole, and
 $\phi_q^j(\mathbf{r})$ its phase

Phase mismatch: $\Delta\mathbf{k}_q = \mathbf{k}_q - \nabla\phi_q^j = k_q \mathbf{e}_z - \frac{\partial\phi_q^j}{\partial z} \mathbf{e}_z - \frac{\partial\phi_q^j}{\partial\rho} \mathbf{e}_\rho$

Perfect
phase-matching
 $\Delta k_q = 0$

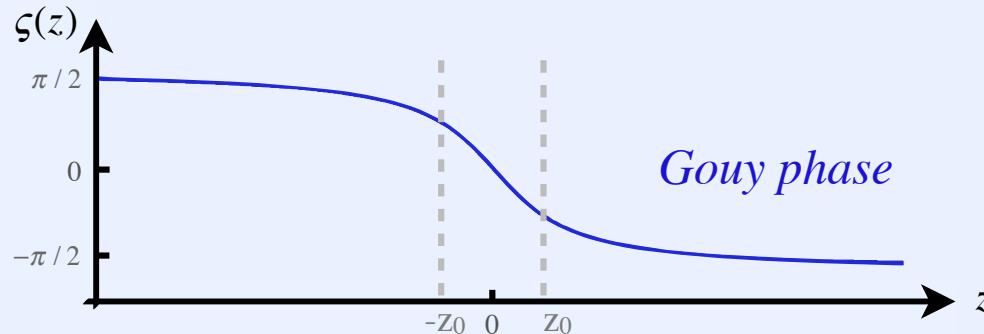
$$\Delta k_q^\parallel = k_q - \frac{\partial\phi_q^j}{\partial z} \approx q \frac{\omega}{c} [n_r(q\omega_0) - n_r(\omega_0)] + (q-1) \frac{\partial\zeta(z)}{\partial z} + \alpha_q^j \frac{\partial I(z)}{\partial z} \quad \text{Longitudinal PM}$$

$$\Delta k_q^\perp = -\frac{\partial\phi_q^j}{\partial\rho} \approx -q \frac{\omega}{c} n_r(\omega) \frac{\rho}{R(z)} + \alpha_q^j \frac{\partial I(z)}{\partial z} \quad \text{Transversal PM}$$

Contributions to phase-matching

$$\Delta k_q = k_q - qk_1 \approx \Delta k_q^{geom} + \Delta k_q^{int} + \Delta k_q^f + \Delta k_q^b$$

Geometrical: Gouy + angle of detection



$$\Delta k_q^{geom} \approx \frac{q-1}{z_0} - \frac{q\omega_0}{2c}\theta^2$$

Intrinsic phase $\phi_q^i = \frac{1}{\hbar} S(\vec{k}^{st}, t^{st,i}, t_{ion}^{st,i}) + \omega t^{st,i}$ $\Delta k_q^{int} \approx \alpha_q^i \frac{\partial I(z)}{\partial z}$

Free electrons and neutrals $n_r \approx 1 + 2\pi(\chi_f + \chi_b)$

$$\Delta k_q^{f,b} = \frac{4\pi^2}{\lambda_q} [\chi_{f,b}(\lambda_q) - \chi_{f,b}(\lambda_0)] \approx \frac{4\pi^2}{\lambda_q} \chi_{f,b}(\lambda_0)$$

Computing Harmonic Propagation

I) The standard way: to solve the Wave equation

$$\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial}{\partial t^2} \vec{E} = \frac{4\pi}{c^2} \frac{\partial}{\partial t} \vec{J}$$

A. L'Huillier et al, J. Opt. Soc. Am. B 7, 527 (1990).
A. L'Huillier, et al, Phys. Rev. A 46, 2778 (1992).

- ✓ Slowly Varying Envelope Approximation: $\partial^2/\partial t^2 \ll \omega \partial/\partial t$
- ✓ Paraxial Approximation: $\partial^2/\partial z^2 \ll k \partial/\partial z$

2) Our choice: to use the formal solution of the Maxwell's equations for an elementary radiator:

$$\vec{E}_i(\vec{r}, t) = -\frac{1}{c^2} \int d\vec{r}' \frac{1}{|\vec{r} - \vec{r}'|} \left[\frac{\partial}{\partial t'} \vec{J}(\vec{r}', t') \right]_{ret}$$

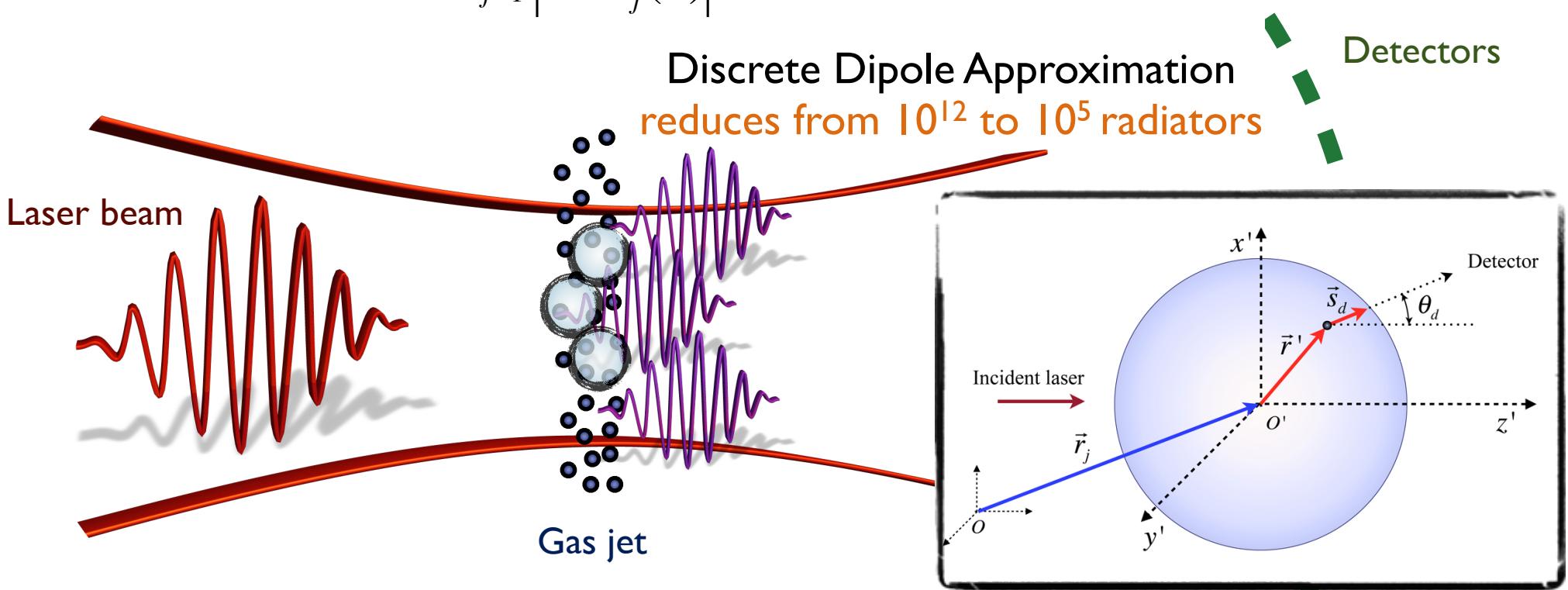
without Slowly Varying Envelope and Paraxial Approximations.

C. Hernández-García, et al, Phys. Rev. A 82, 0033432 (2010)

Sum of elementary radiators

We decompose the integral into a discrete sum of elementary contributions:

$$\vec{E}_i^j(\vec{r}, t) = -\frac{1}{c^2} \sum_{j=1}^N \frac{q_j}{|\vec{r} - \vec{r}_j(0)|} \vec{s}_d \times \left(\vec{s}_d \times \vec{a}_j \left(t - |\vec{r} - \vec{r}_j(0)|/c \right) \right)$$



C. Hernández-García, J.A. Pérez-Hernández, J. Ramos, E. Conejero
Jarque, L. Roso, and L. Plaja, Phys. Rev. A 82, 0033432 (2010)

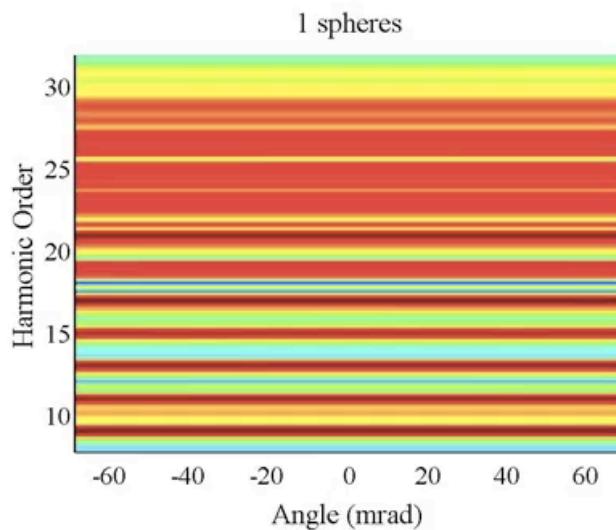
$$\vec{E}_{i,j}(\vec{r}_d, \omega) \propto -N(\vec{r}_j) \vec{s}_d \times \left(\vec{s}_d \times \vec{a}_j(\vec{r}_j, \omega) e^{-i \frac{\omega}{c} |\vec{r}_d - \vec{r}_j|} F(\theta_d, \omega) \right)$$

DDA - Convergence

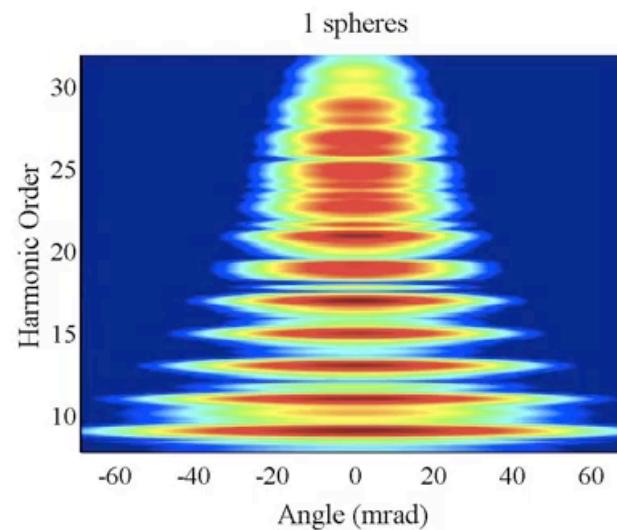
Without DDA

With DDA

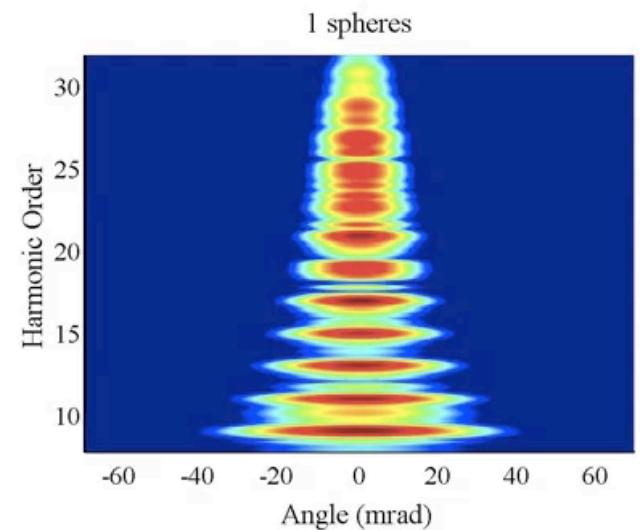
Point Atoms



1 μm Spheres

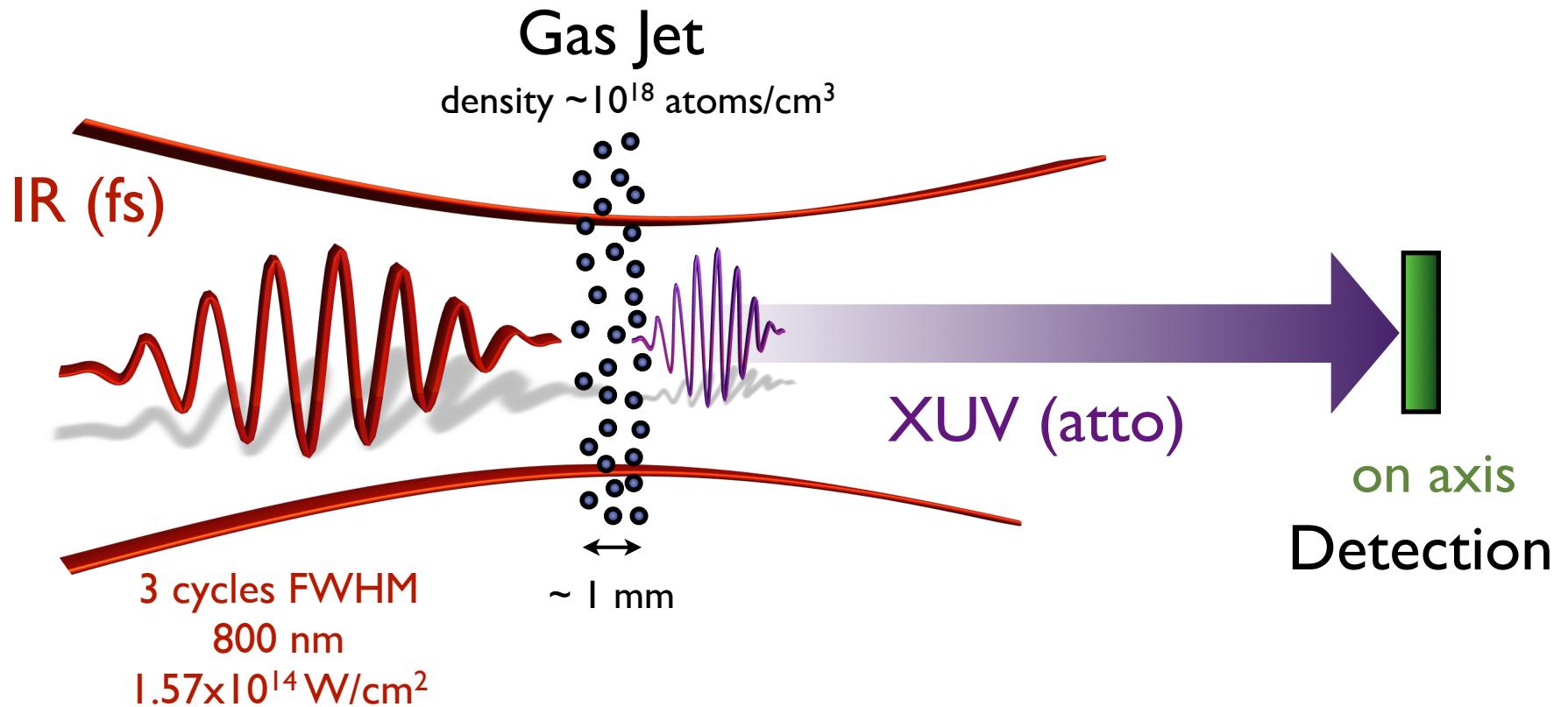


2 μm Spheres



C. Hernández-García, J.A. Pérez-Hernández, J. Ramos, E. Conejero
Jarque, L. Roso, and L. Plaja, Phys. Rev. A 82, 0033432 (2010)

On-axis phase-matching in a gas jet

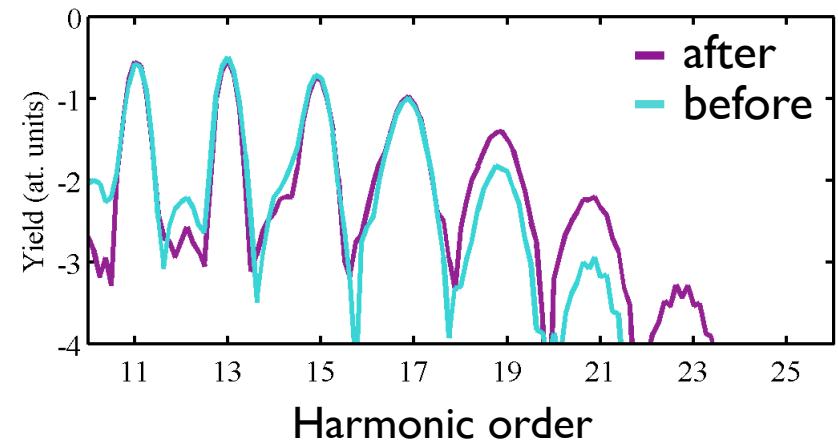
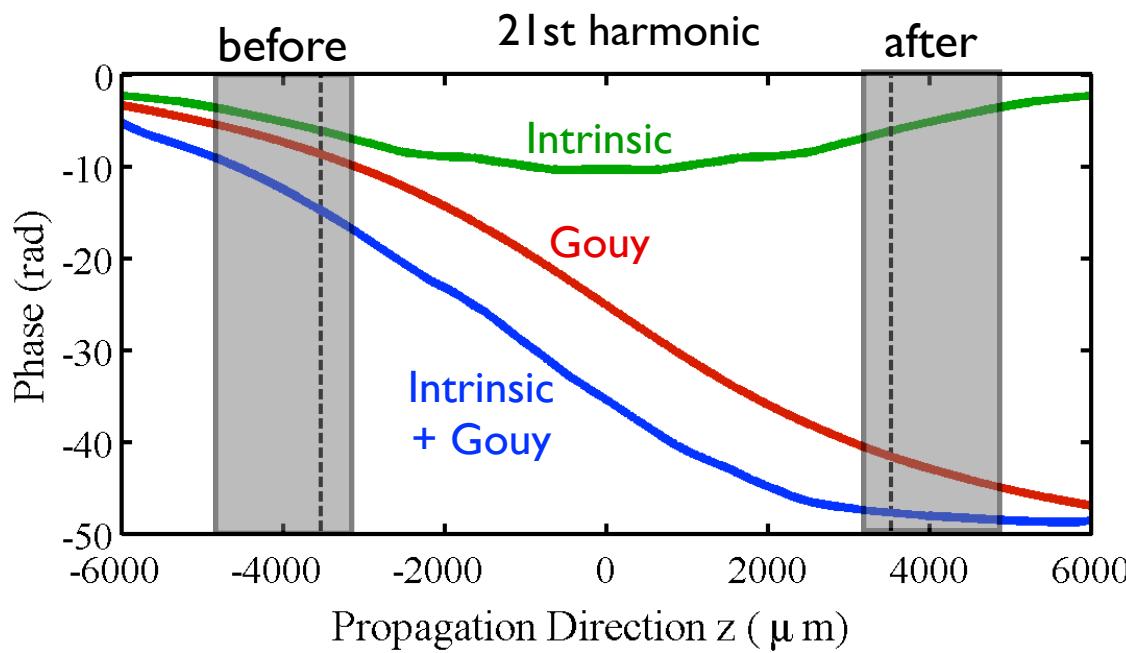


$$\Delta k_q \simeq \Delta k_q^{Gouy} + \Delta k_q^{\text{int}} + \Delta k_q^f + \cancel{\Delta k_q^b} \simeq \frac{q-1}{z_0} + \alpha_q^i \frac{\partial I(z)}{\partial z} + q \frac{e^2 n_f \lambda_0}{mc^2}$$

On-axis phase-matching in a gas jet

$$\Delta k_q \simeq \Delta k_q^{Gouy} + \Delta k_q^{\text{int}} + \Delta k_q^f + \Delta k_q^o \simeq \frac{q-1}{z_0} + \alpha_q^i \frac{\partial I(z)}{\partial z} + q \frac{e^2 n_f \lambda_0}{mc^2}$$

	>0	>0
before the focus	>0	<0



Already demonstrated:

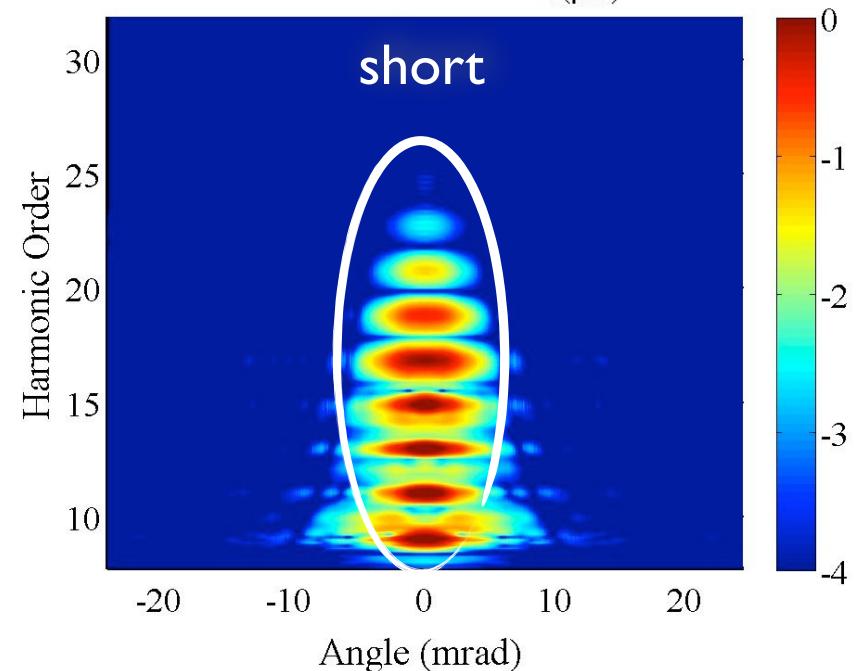
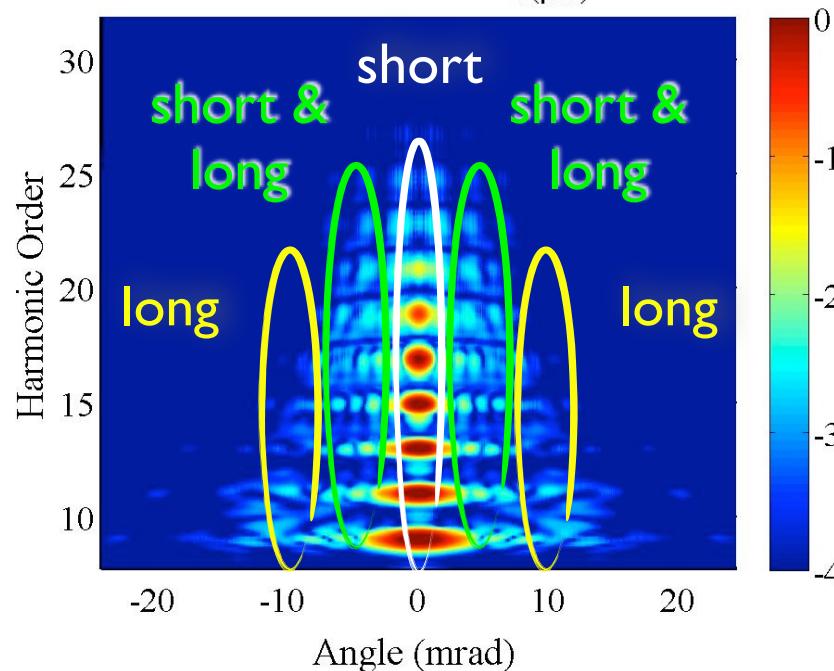
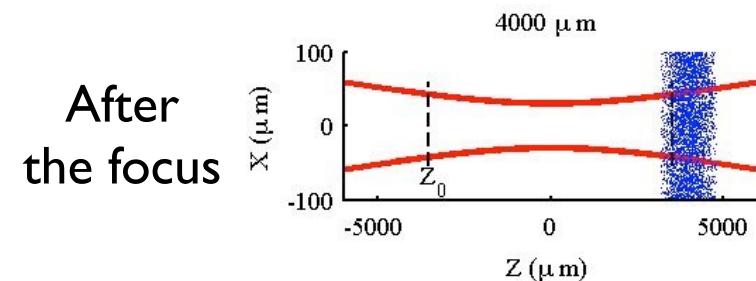
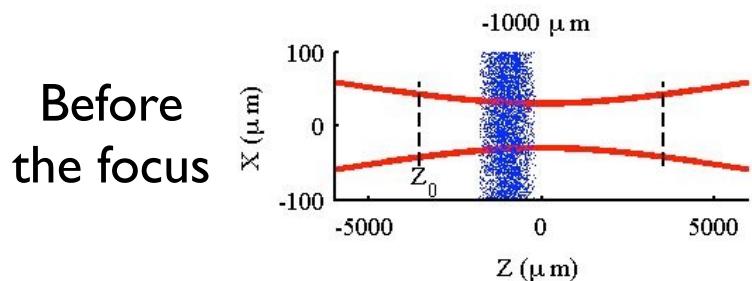
P. Salières, A. L'Huillier, and M. Lewenstein, PRL, 74, 3776 (1995)

P. Antoine , A. L'Huillier, and M. Lewenstein, PRL, 77, 1234 (1996)

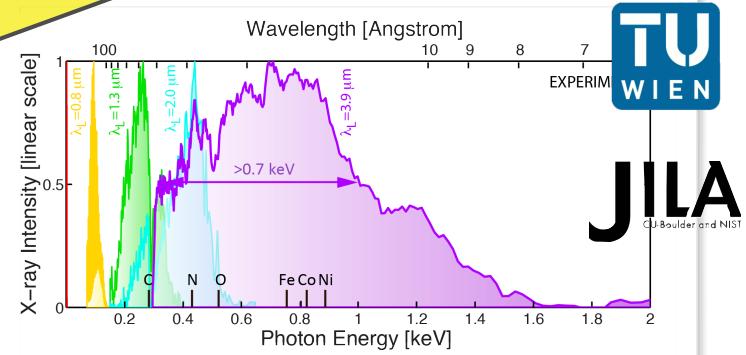
M. B. Gaarde, J. L. Tate, and K.J. Schafer J. Phys. B: At. Mol. Opt. Phys. 41 (2008)

Off-axis phase-matching in a gas jet

$$\Delta k_q \simeq \Delta k_q^{Gouy} + \Delta k_q^{\text{int}} + \Delta k_q^f + \Delta k_q^{\text{angle}} \simeq \frac{q-1}{z_0} + \alpha_q^i \frac{\partial I(z)}{\partial z} + q \frac{e^2 n_f \lambda_0}{mc^2} - \frac{q\omega_0}{2c} \theta^2$$

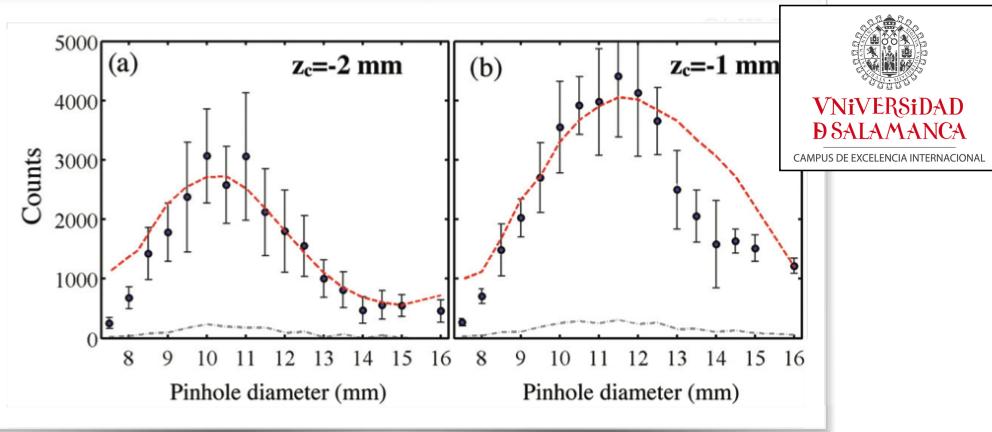


Deliverables of our
Phase-matching code



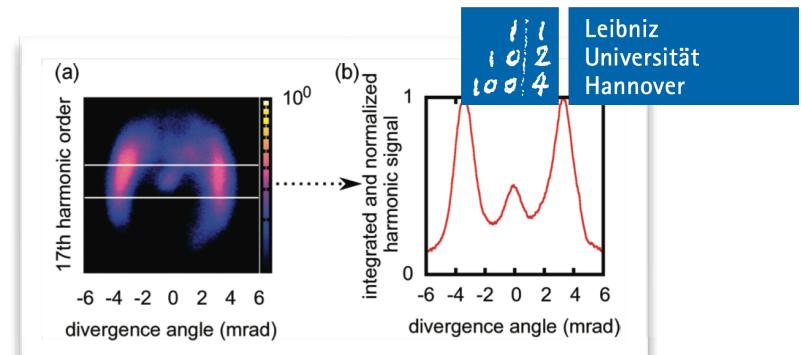
1.6 keV HHG X-rays

T. Popmintchev, et al *Science* 336, 1287 (2012)



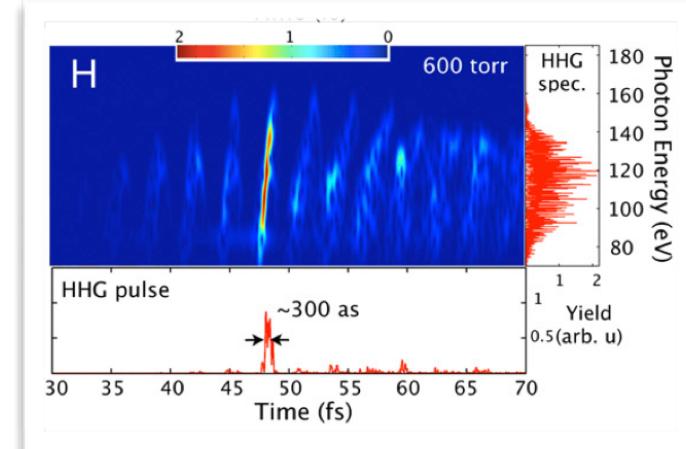
Role of transversal phase-matching

C. Hernández-García, et al *Phys. Rev.A* 88, 043848 (2013)



XUV harmonics in gas cell

M. Kretschmar, et al *Phys. Rev.A* 88, 013805 (2013)



JILA

CU Boulder and NIST

Isolated X-ray attosecond pulses

M.-C. Chen, et al *PNAS* 111, E2361-E2367 (2014)

XUV harmonics from few-cycle pulses

W. Holgado et. al. (in preparation) (2014)

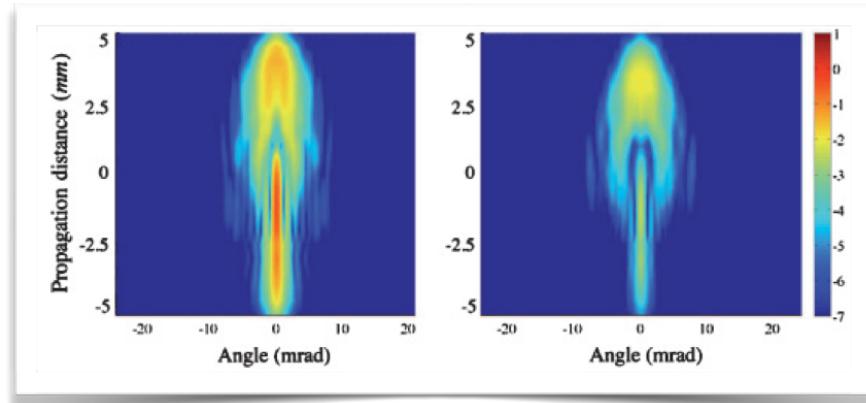
U. PORTO

VNIVERSIDAD
DE SALAMANCA

CAMPUS DE EXCELENCIA INTERNACIONAL

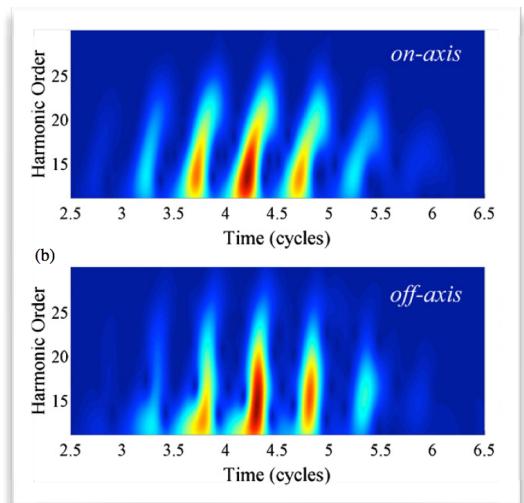
Deliverables of our
Phase-matching code

New theoretical findings



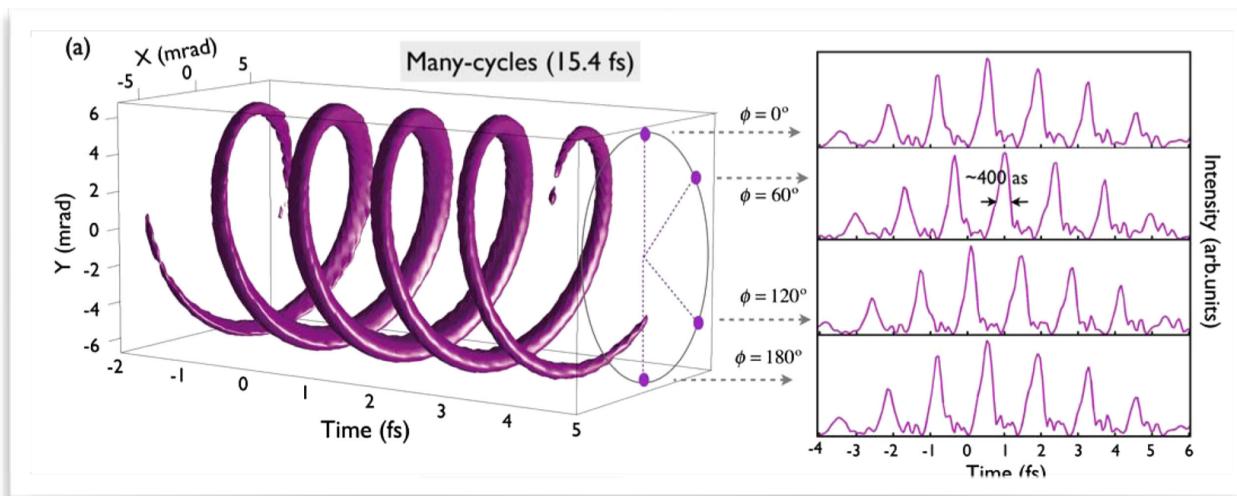
Phase-matching conditions along the propagation direction

C. Hernández-García, et al, Phys. Rev. A 82, 0033432 (2010)



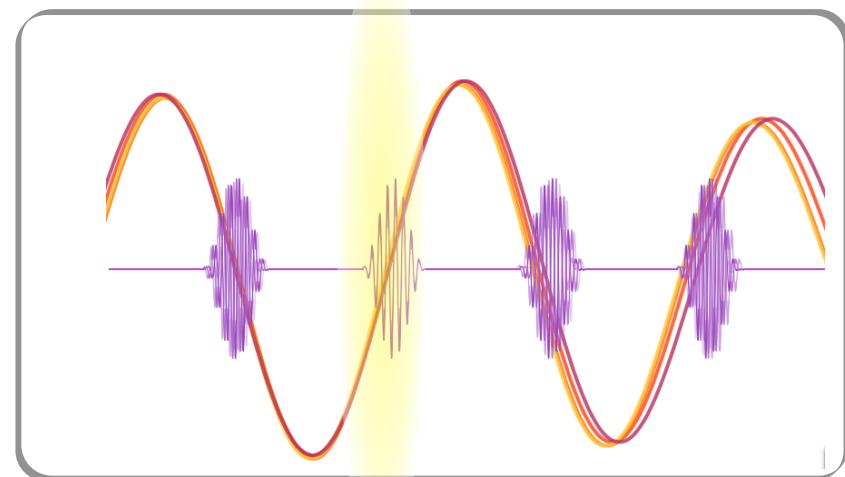
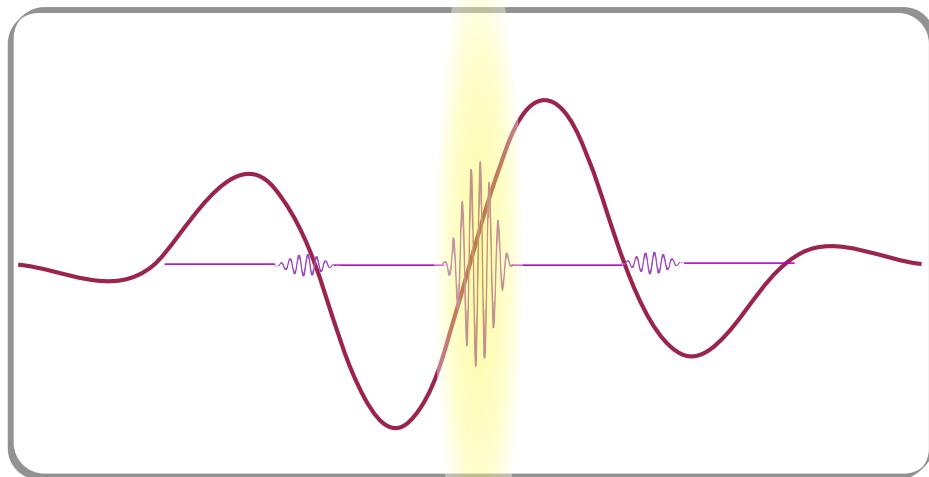
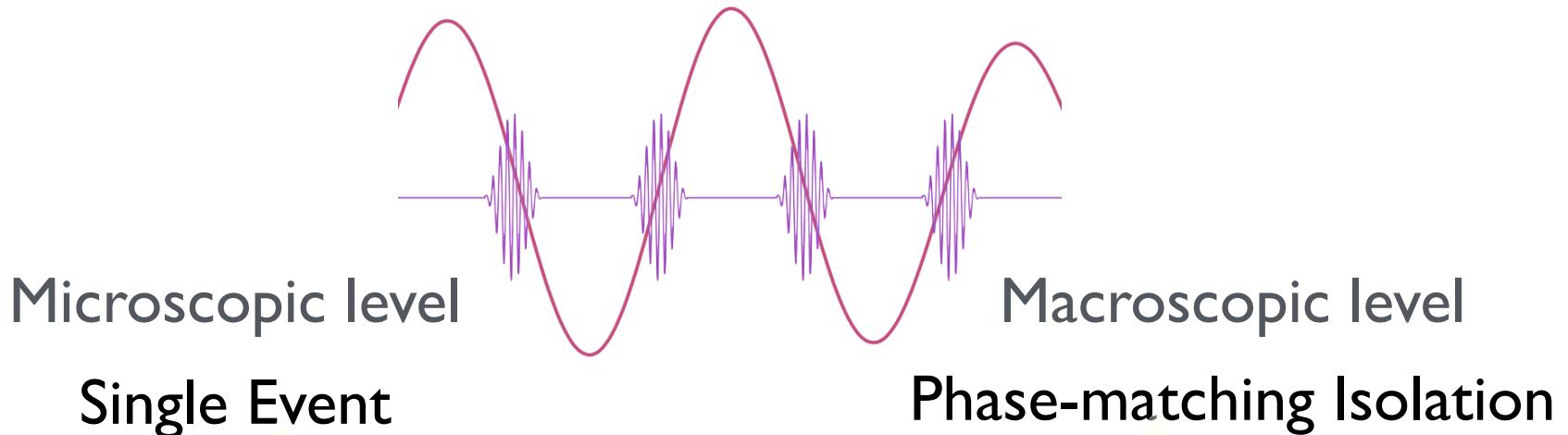
Off-axis compensation of
the atto-chirp

C. Hernández-García, and L. Plaja,
J. Phys. B: At. Mol. Opt. Phys. 45,
074021 (2012)



XUV vortices: conservation of orbital angular momentum (OAM) in HHG
C. Hernández-García, A. Picón, J. San Román , and L. Plaja, Phys. Rev. Lett. 111, 083602 (2013)

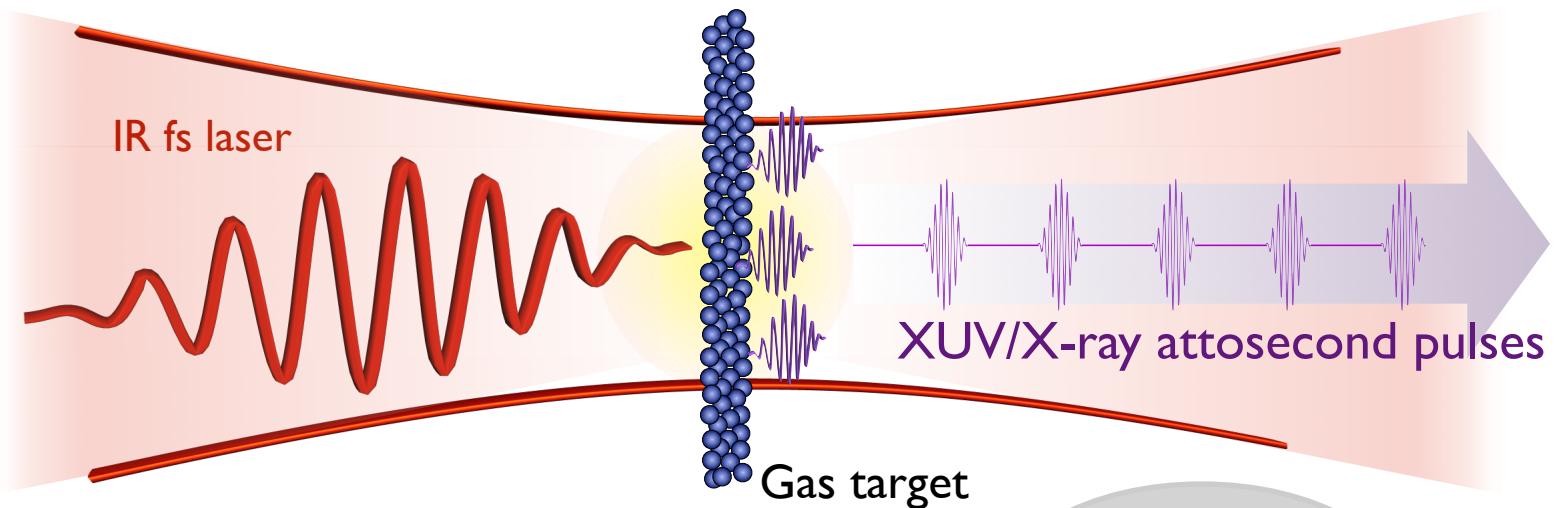
Isolating an attosecond pulse



E. Goulielmakis et al., *Science* **320**, 1614–1617 (2008).
B. Shan et al., *J. Mod. Opt.* **52**, 277 (2005).
I. J. Sola et al., *Nature Physics* **2**, 319–322 (2006).
G. Sansone et al., *Science* **314**, 443–446 (2006).
E. J. Takahashi, et al., *Phys. Rev. Lett.* **104**, 233901 (2010).
Zhao et al. *Opt. Lett.* **37**, 3891 (2012)

M. J. Abel et al., *Chem Phys* **366**, 9–14 (2009)
I. Thomann et al., *Opt. Express* **17**, 4611 (2009)
F. Ferrari, F. Calegari, M. Lucchini, C. Vozzi, S. Stagira, G. Sansone, and M. Nisoli, *Nature Photonics* **4**, 875–879 (2010)
M.-C. Chen, C. Mancuso, C. Hernández-García, F. Dollar, B. Galloway, D. Popmintchev, B. Langdon, A. Auger, P.-C. Huang, B.C. Walker, L. Plaja, A. Jaron-Becker, A. Becker, M. M. Murnane, H. C. Kapteyn, T. Popmintchev, *PNAS* **111** (23), E2361-E2367 (2014)

Optimal phase-matching



$$\text{Phase-mismatch: } \Delta k_q = k_q - qk_1$$

Perfect
phase-matching
 $\Delta k_q = 0$

$$\Delta k_q \simeq \cancel{\Delta k_q^{geom}} + \cancel{\Delta k_q^{int}} + \Delta k_q^f + \Delta k_q^b$$

$$\text{Optimal phase-matching: } |\Delta k_q^f| = |\Delta k_q^b| \rightarrow \Delta k_q = 0$$

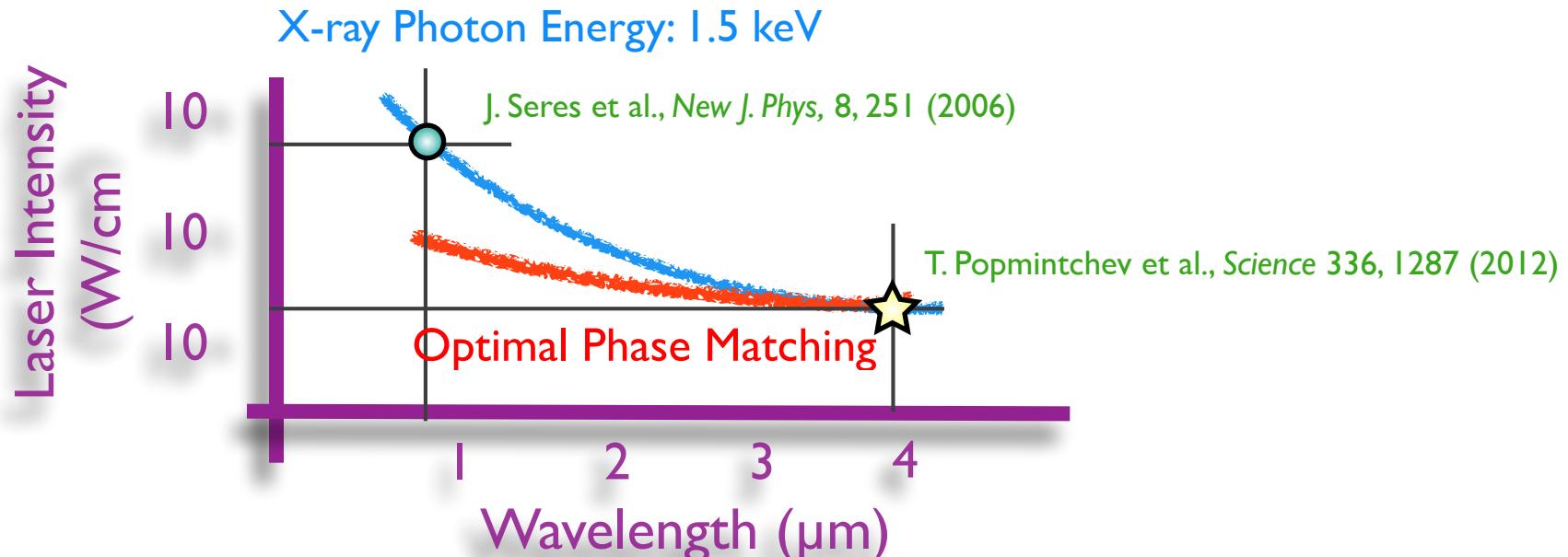
Optimal phase-matching

$$\Delta k_q = k_q - qk_1 \simeq \Delta k_q^{geom} + \Delta k_q^{int} + \Delta k_q^f + \Delta k_q^b$$

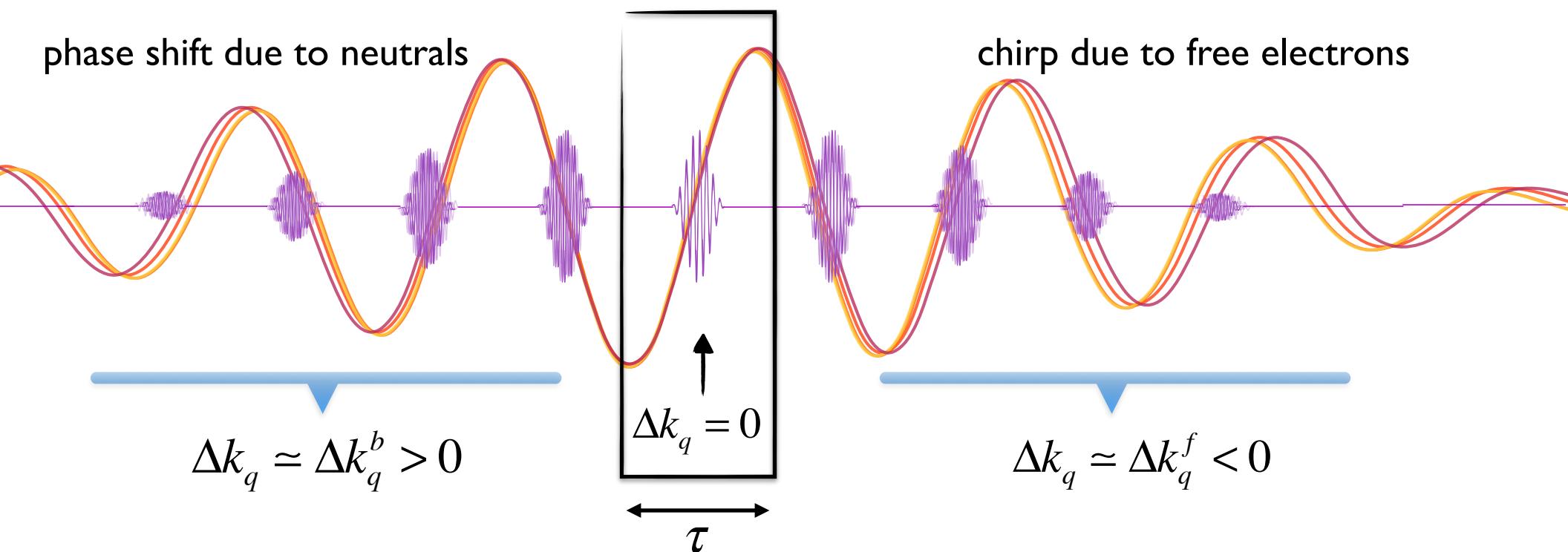
$$\Delta k_q \simeq -\frac{4\pi^2}{\lambda_q} [\chi_f(\lambda_0) + \chi_b(\lambda_0)] \simeq q \left[\frac{P_f n_0 e^2 \lambda_0}{mc^2} - \frac{4\pi^2 (1 - P_f) n_0 \chi_b^0}{\lambda_0 \eta_0} \right]$$

$$\Delta k_q = 0 \rightarrow P_f^{opt} \approx \frac{\chi_b^0}{\eta_0} \left(\frac{e^2 \lambda_0}{4\pi mc^2} - \frac{\chi_b^0}{\eta_0} \right)^{-1}$$

Optimal ionized population,
 $P_f^{opt} \rightarrow (\lambda_0, I)$



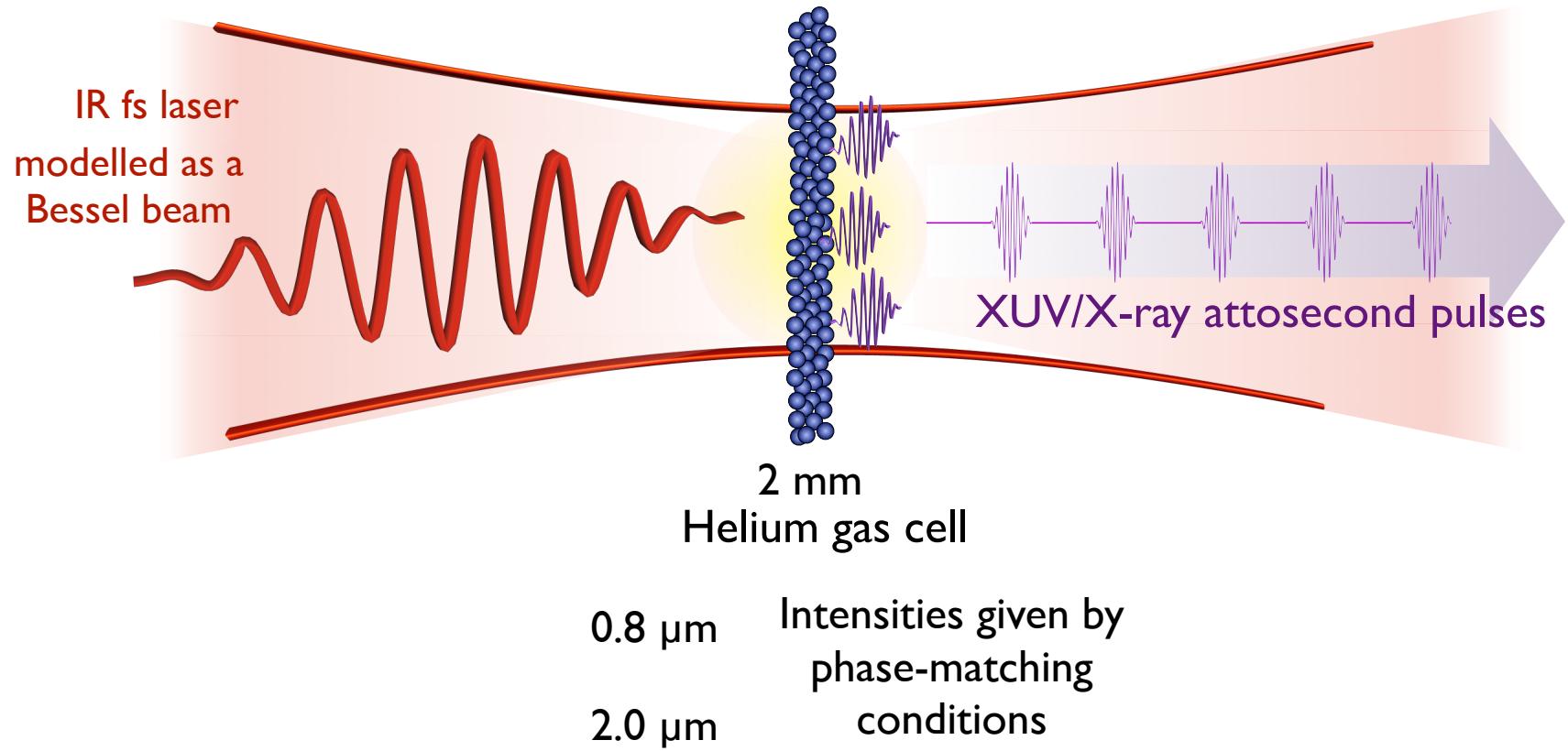
Time gated phase-matching



Size of the phase-matching window

$$\frac{1}{\tau} \propto \frac{\partial \Delta k_q}{\partial t} \propto \lambda_0^{1.5} P$$

3D simulations



Theoretical Method:

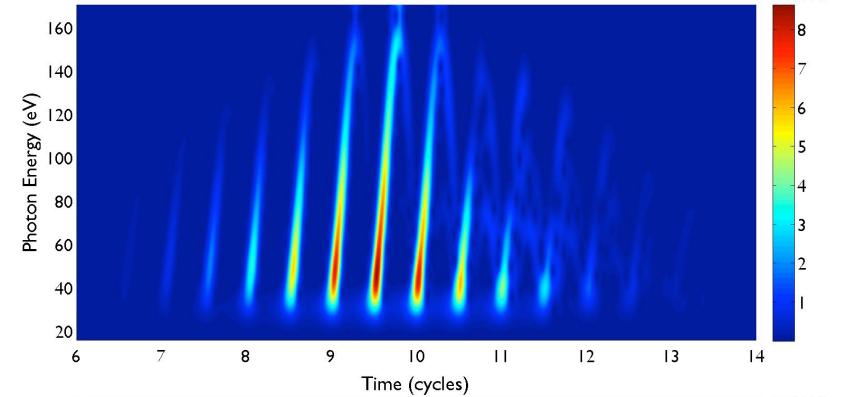
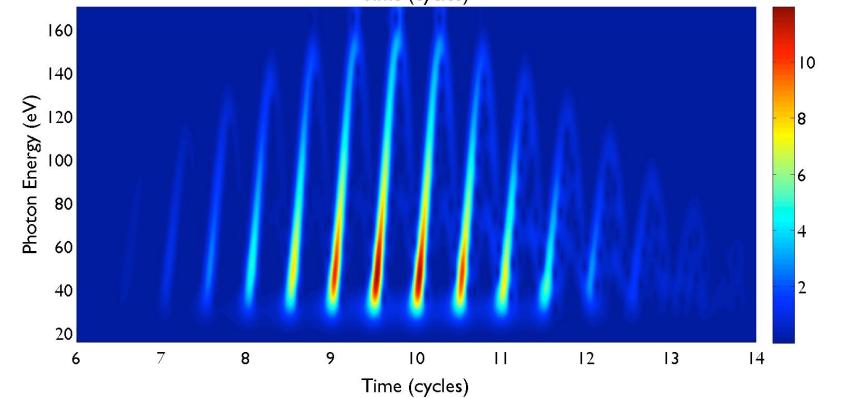
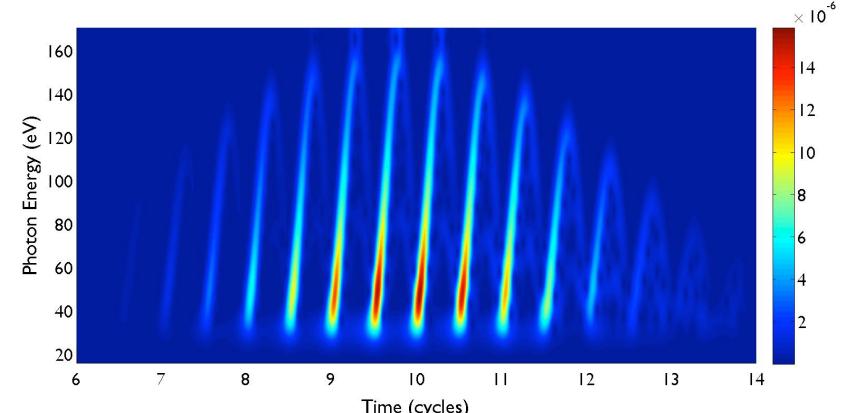
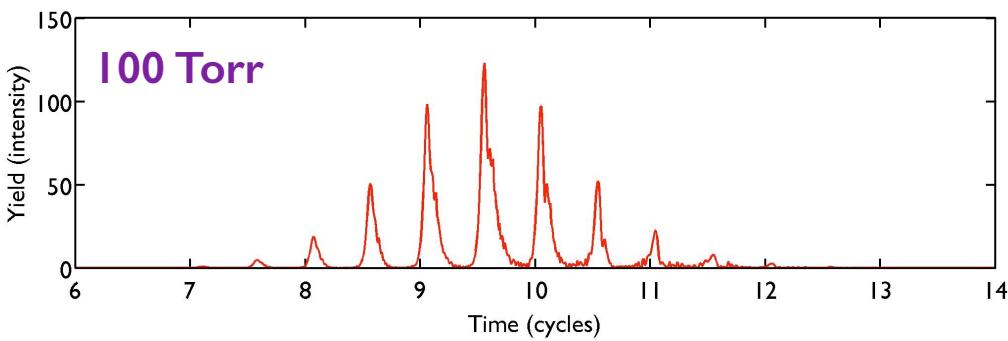
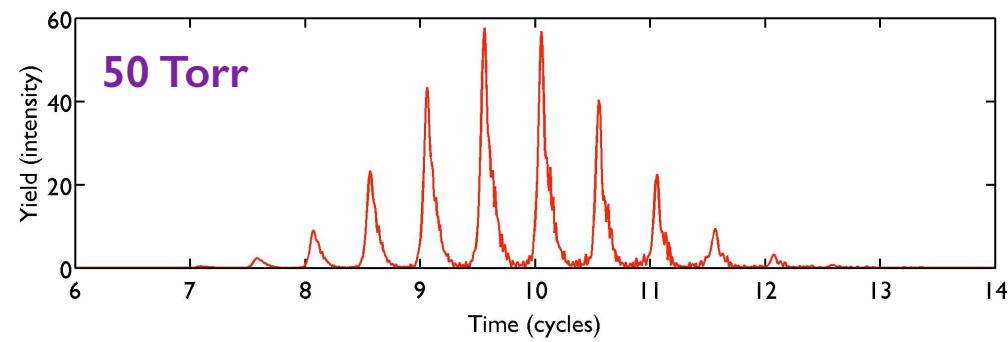
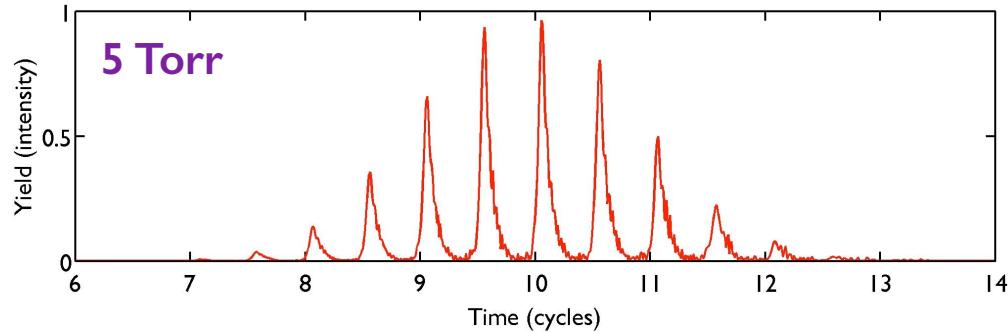
- High-Harmonic Generation: SFA+
- Propagation: Discrete Dipole Approximation using Maxwell equations

C. Hernández-García, et al. Phys. Rev.A 82, 033432 (2010)

3D simulations: 800 nm, 5.8 cycles

2 mm He gas cell, peak intensity $6.6 \times 10^{14} Wcm^{-2}$

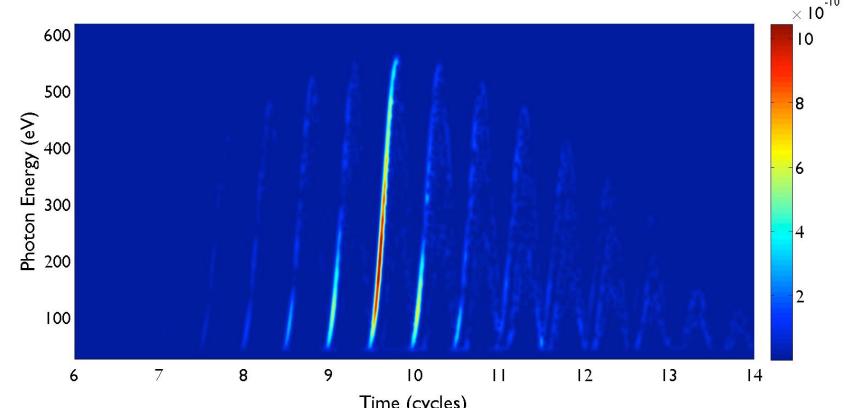
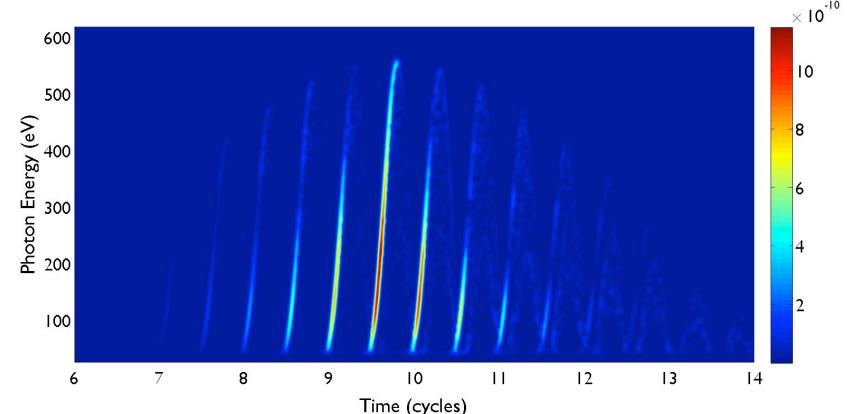
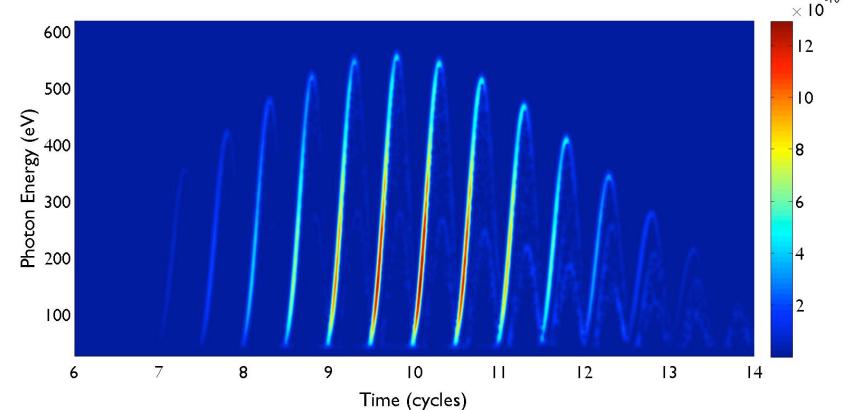
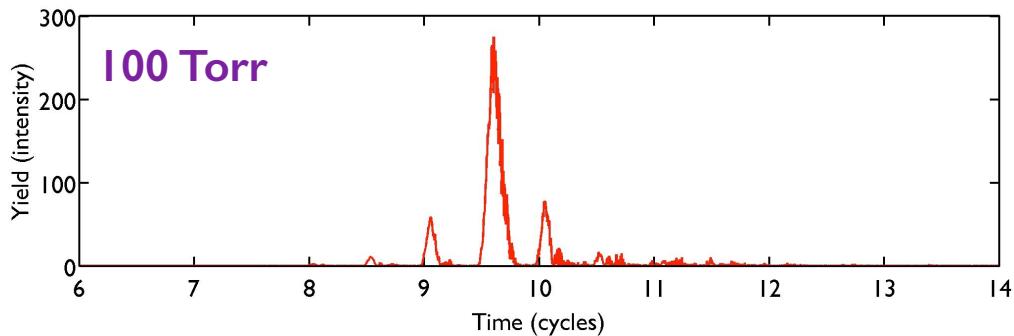
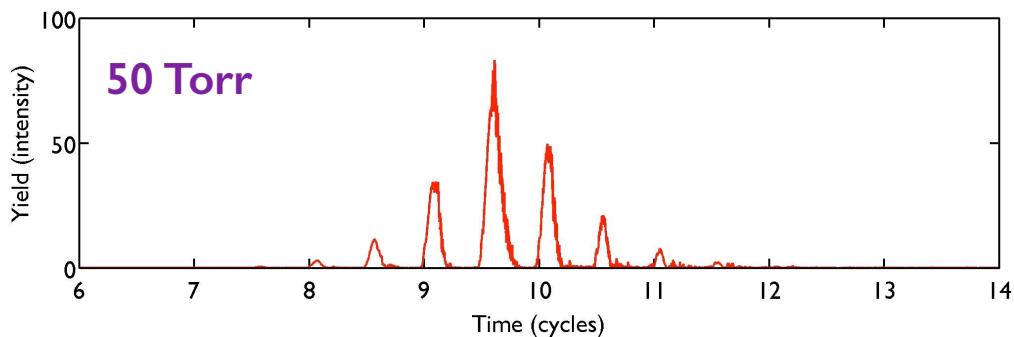
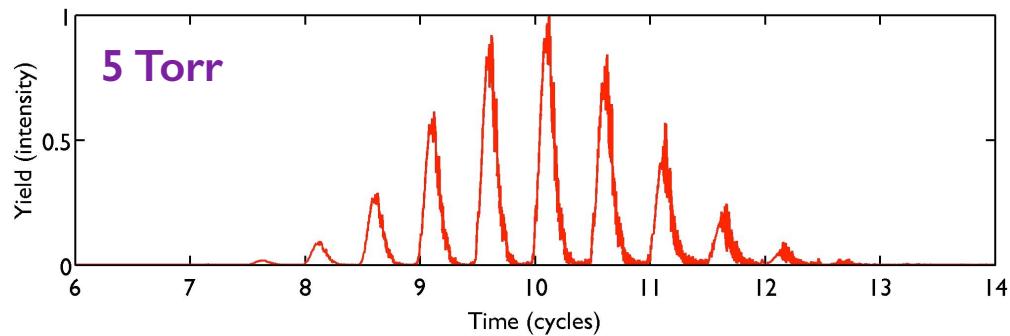
$$\frac{\partial \Delta k_q}{\partial t} \propto \lambda_0^{1.5} P$$



3D simulations: 2.0 μm , 5.8 cycles

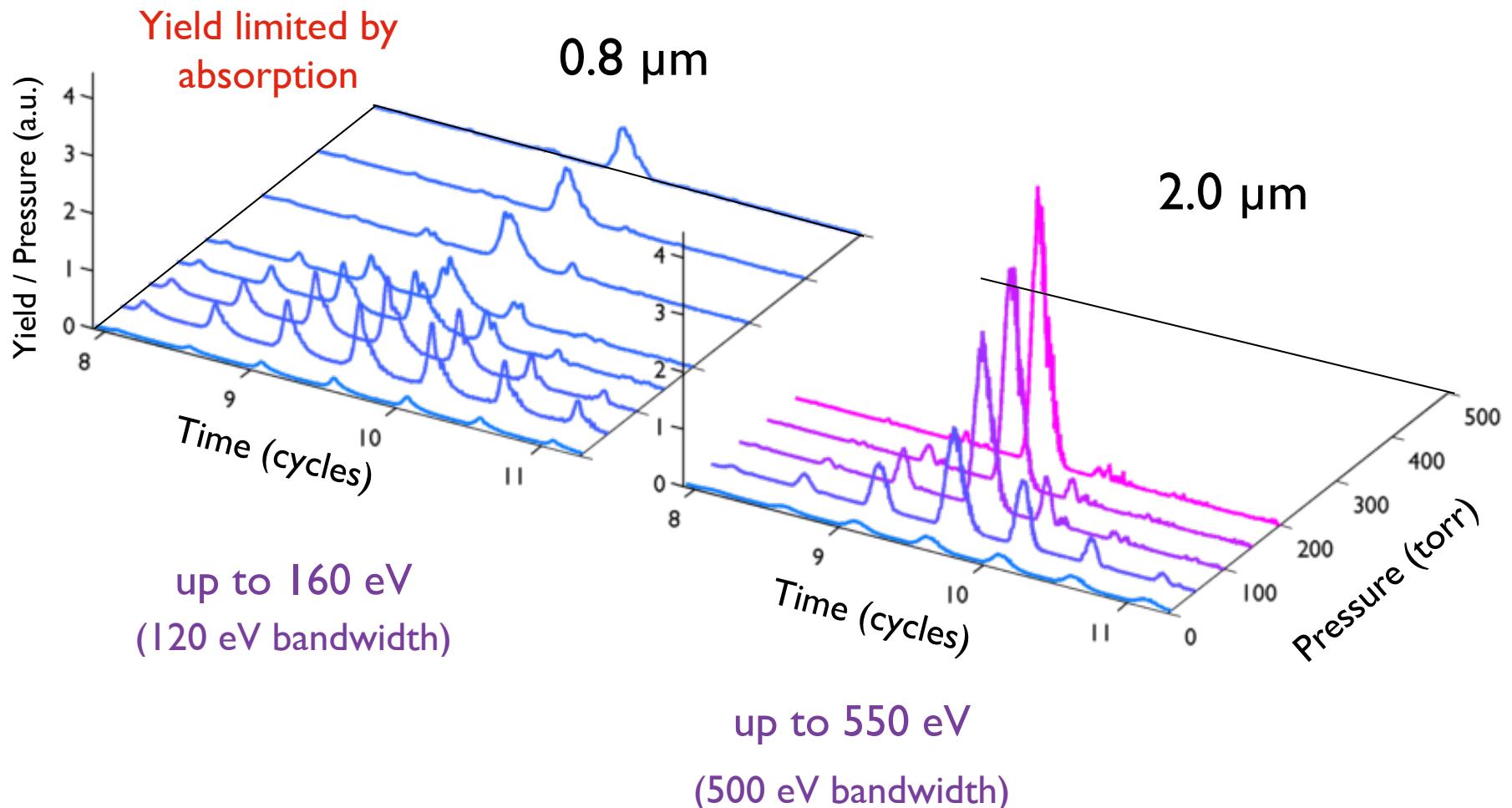
2 mm He gas cell, peak intensity $4.4 \times 10^{14} \text{ W cm}^{-2}$

$$\frac{\partial \Delta k_q}{\partial t} \propto \lambda_0^{1.5} P$$

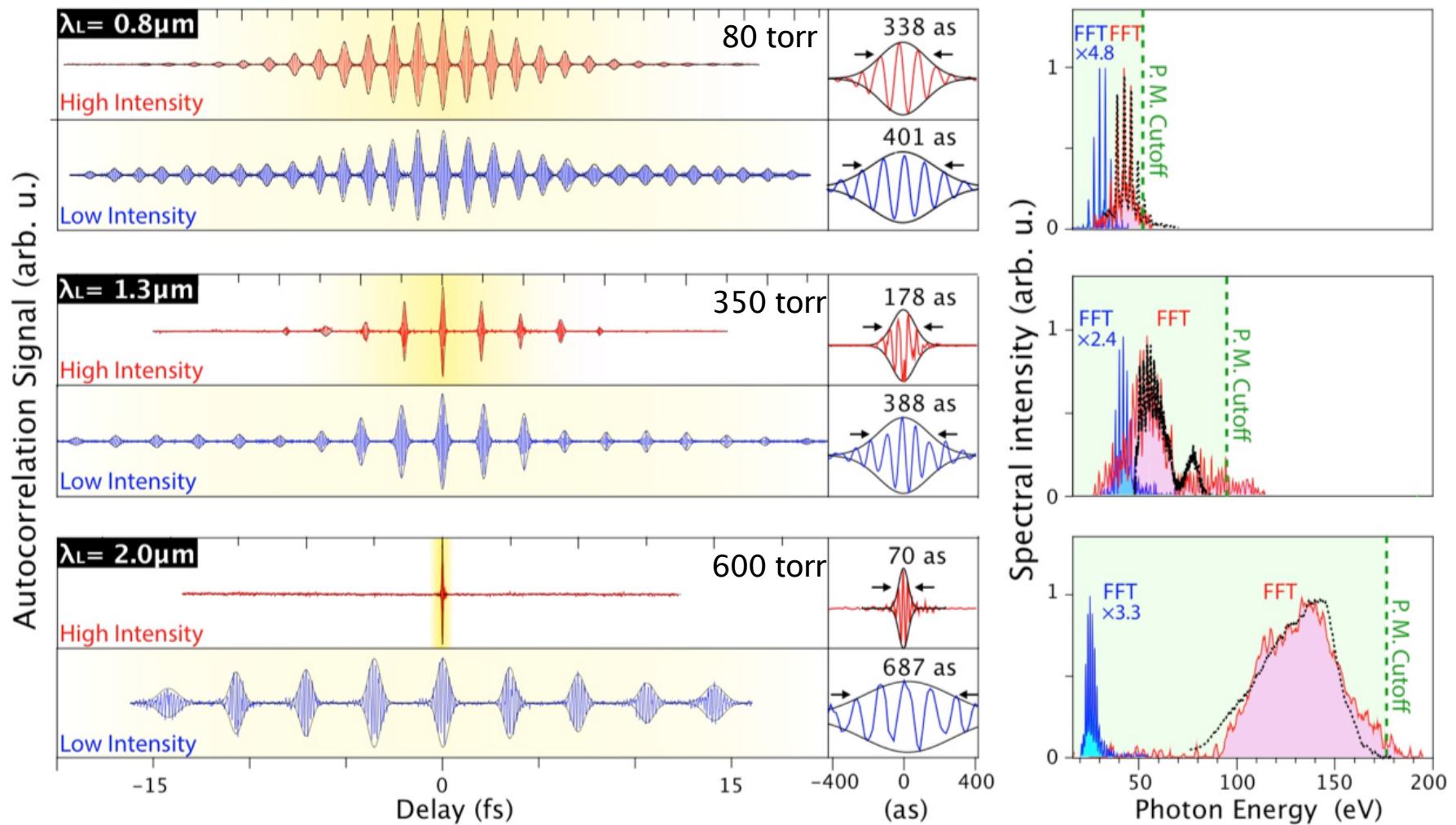


Phase-matching with multi-cycle pulses

2 mm He gas cell

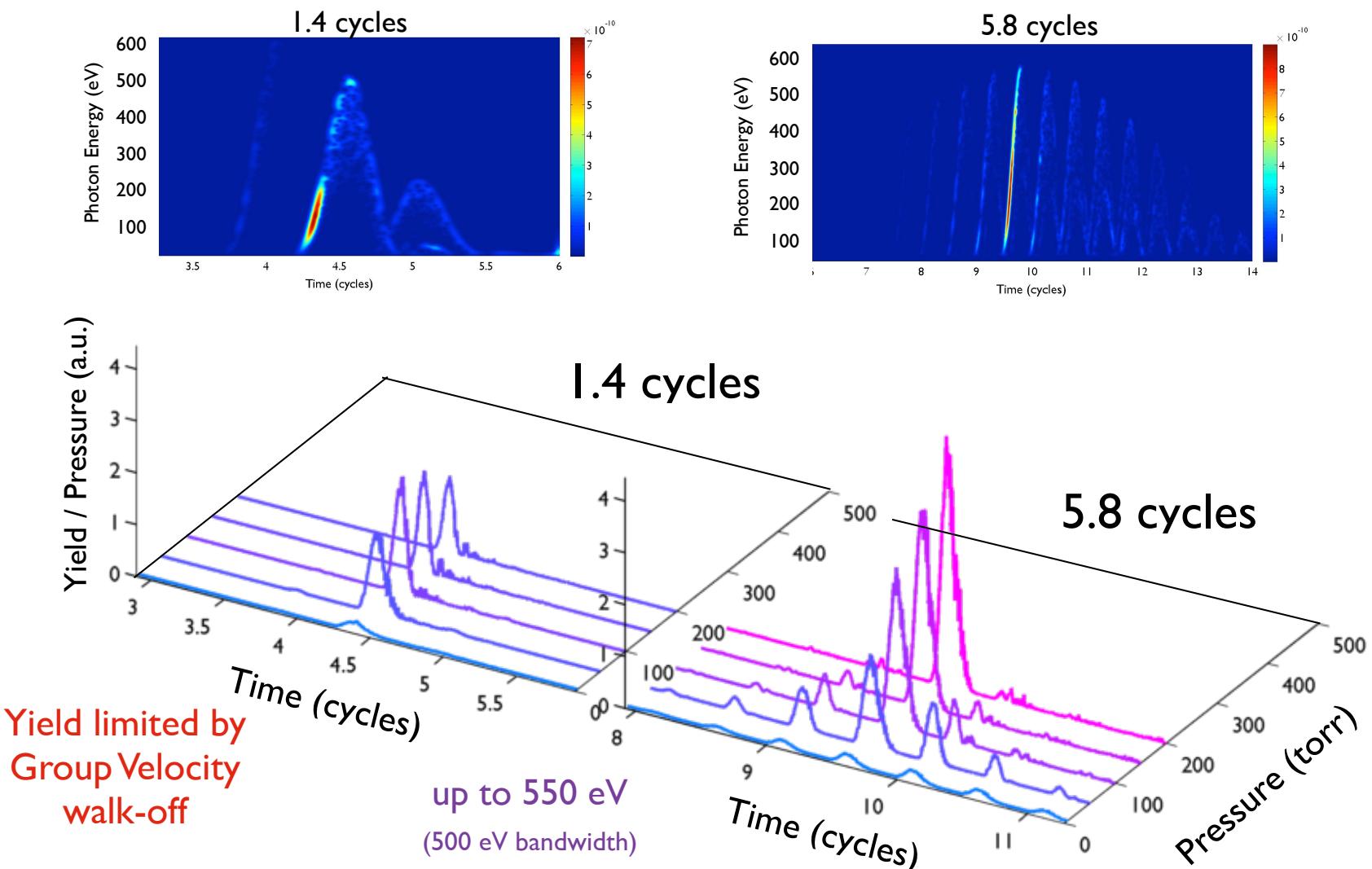


Experimental results

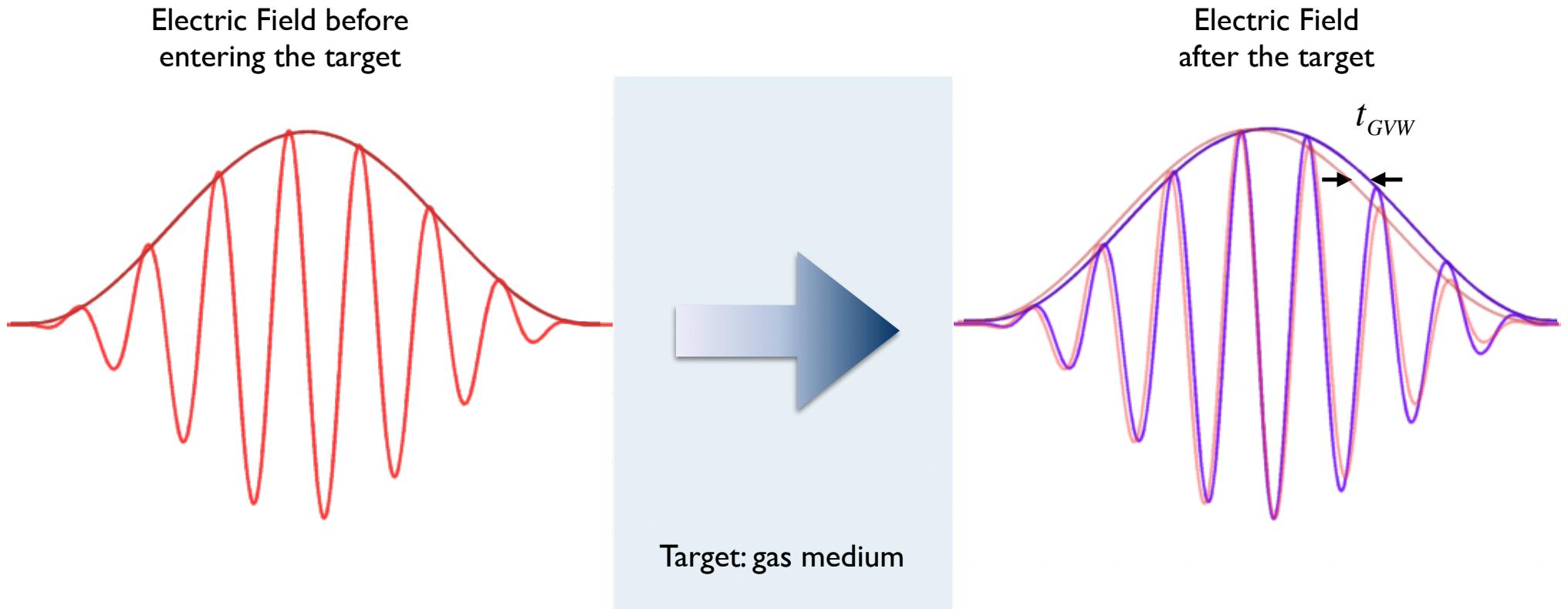


M.-C. Chen, C. Mancuso, C. Hernández-García, F. Dollar, B. Galloway, D. Popmintchev, B. Langdon, A. Auger, P.-C. Huang, B.C. Walker, L. Plaja, A. Jaron-Becker, A. Becker, M. M. Murnane, H. C. Kapteyn, T. Popmintchev,
PNAS 111 (23), E2361-E2367 (2014)

Few-cycle vs Multi-cycle at 2 μ m



Group velocity walk-off



Group velocity:

$$\frac{1}{v_g} - \frac{1}{v_{ph}} = \frac{\omega}{c} \frac{\partial n(\omega)}{\partial \omega}$$

Temporal delay due to
group velocity walk-off

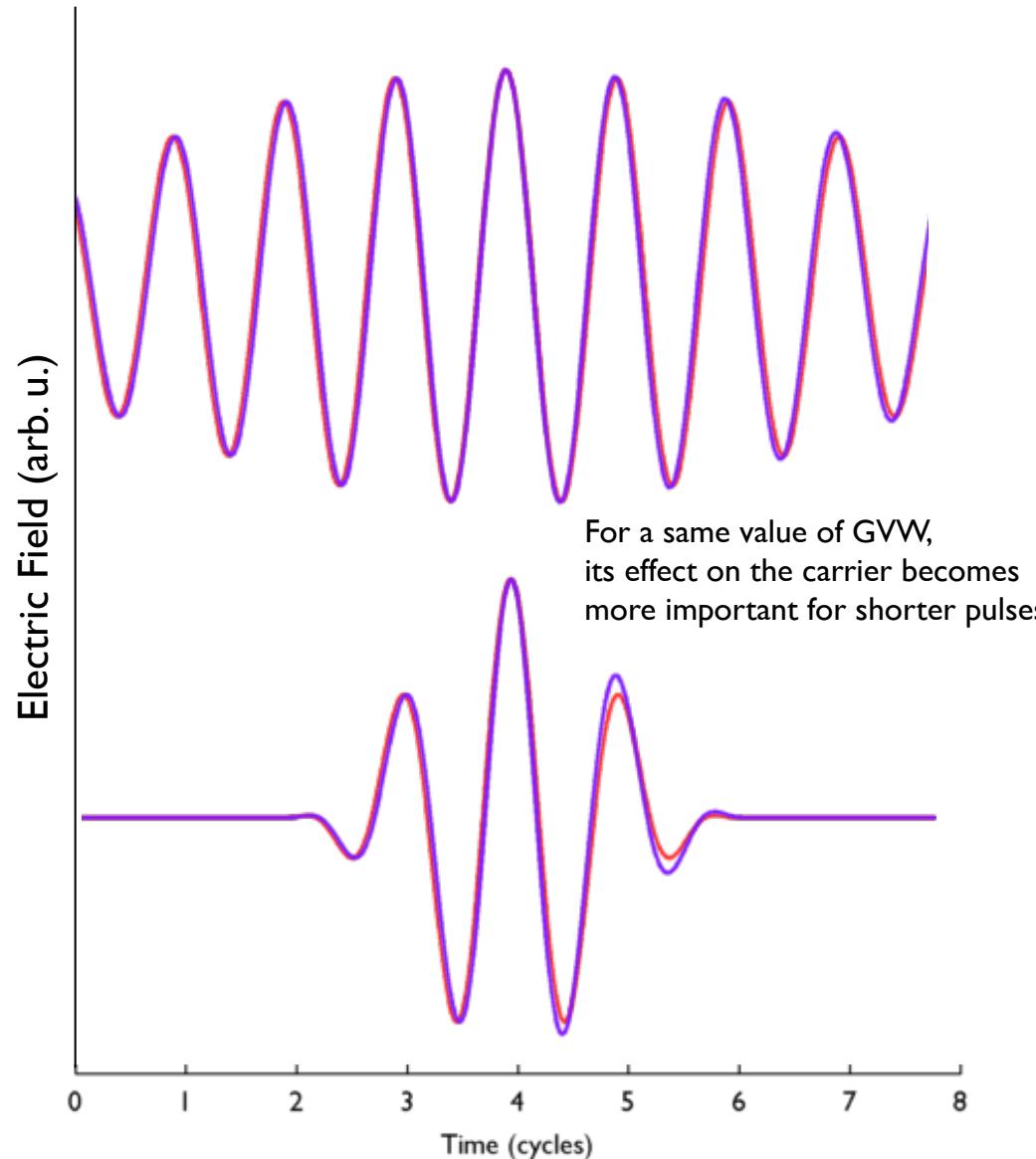
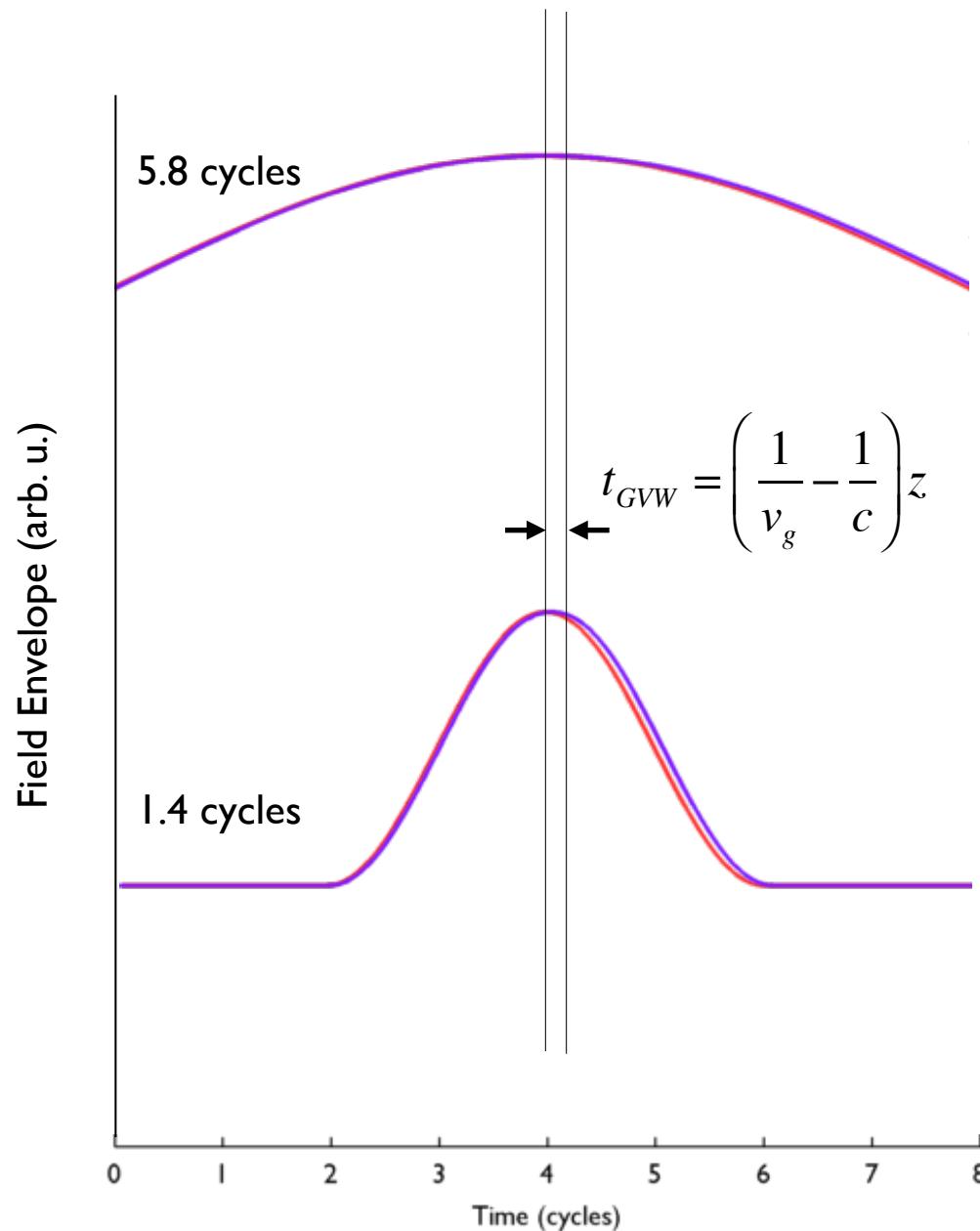
$$t_{GVW} = \left(\frac{1}{v_g} - \frac{1}{c} \right) z$$

Group velocity matching:

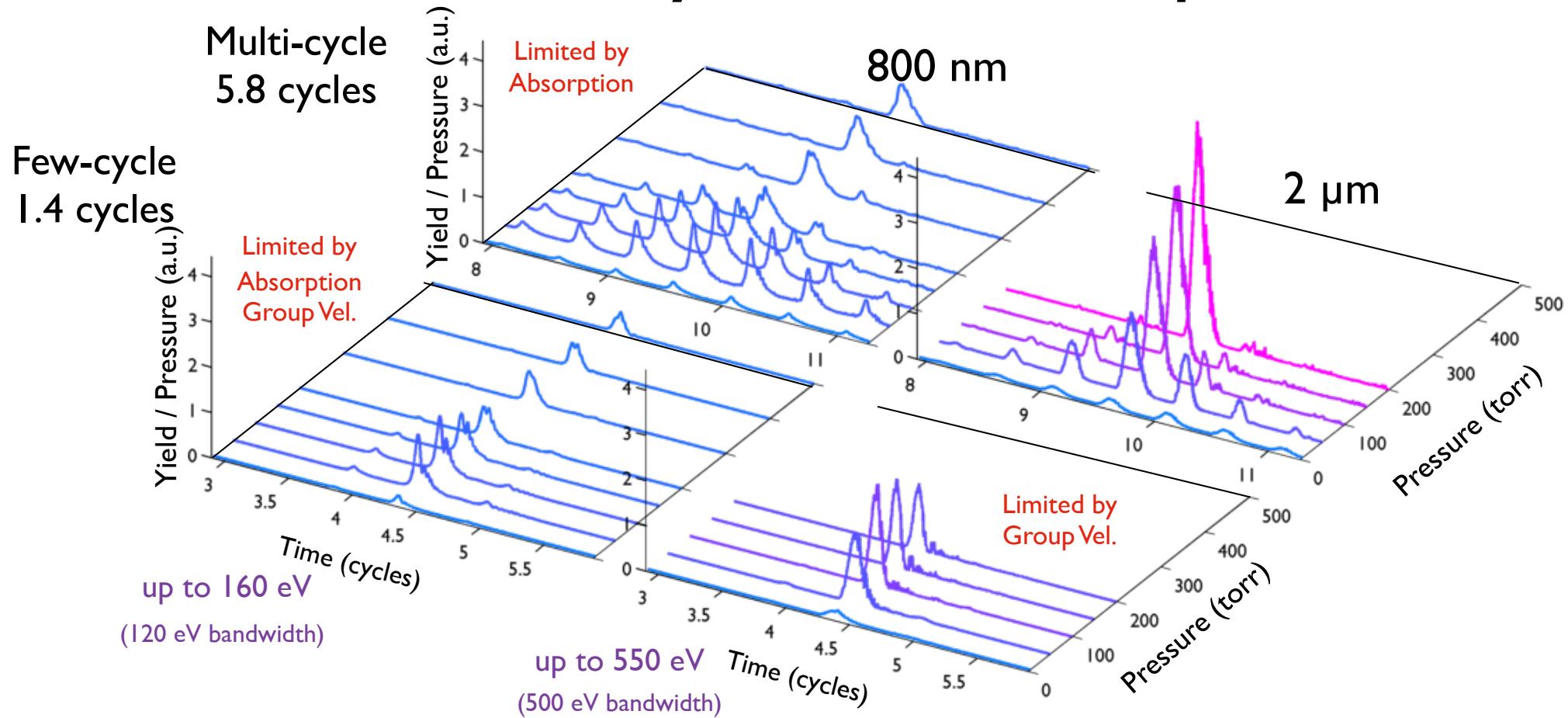
Fundamental field travels with $v_g(\omega_0)$

Harmonic field travels with $v_g(q\omega_0) \approx c$

Effect of the group velocity walk-off on the carrier



Isolated X-ray attosecond pulses



- We derive a route for **extending isolated attosecond pulses to the x-ray regime** by using mid-IR wavelengths.
- **Multi-cycle laser pulses** show better yield scaling due to group velocity walk-off.

Conclusions

- We propose a route for producing zeptosecond waveforms in the keV regime, driving HHG by mid-IR laser pulses.
- We demonstrate the generation of isolated attosecond pulses in the X-ray regime using mid-IR multi-cycle laser pulses.