

Attosecond pulse  
generation & application:

Theoretical perspectives  
from strong field physics

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# What do we mean by strong field physics?

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## Phenomena:

- Above threshold ionization
- High harmonic generation
- Seq. & non-seq. multiple ionization
- Coulomb explosion of molecules
- X-rays from clusters

# What do we mean by strong field physics?

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## Attributes:

- Ionization driven processes
- Non-perturbative:  $H_I \sim H_{at}$
- Electron wave packet dynamics (non-trivial!)

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  - Recollision model
  - Quantum orbits

# What do we mean by strong field physics?

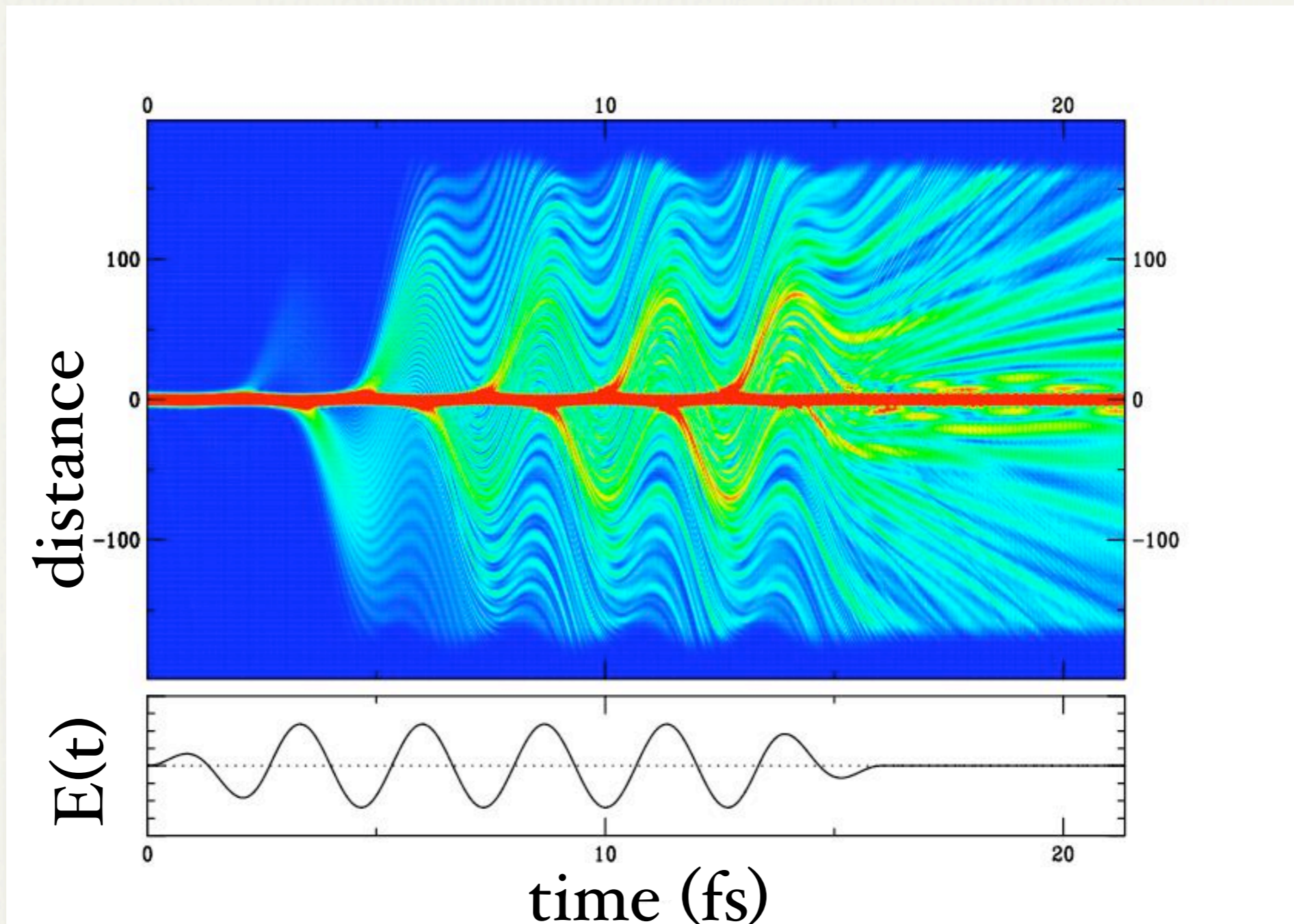
## Attributes:

- Ionization driven processes
- Non-perturbative:  $H_I \sim H_{at}$
- Electron wave packet dynamics (non-trivial!)
- Recollision model
- Quantum orbits

$$d_q(\mathbf{I}) = \sum_{\text{traj}} A_\tau(\mathbf{I}) \exp[-iS_\tau(\mathbf{I})]$$

Sum over  
paths

# Visualizing quantum orbits



1-d quantum model

# Strong field physics in attoscience

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- Generation
- Metrology
- Applications



# Strong field physics in attoscience

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- **Generation**
  - Single atto pulses from cutoff harmonics
  - Trains of atto pulses from plateau harmonics
  - Compression of APT pulses

# Strong field physics in attoscience

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- **Generation**

- Single atto pulses from cutoff harmonics

- Trains of atto pulses from plateau harmonics

- Compression of APT pulses

- New paradigm for single pulse production

- Trains of atto pulses with one pulse per IR cycle

# Strong field physics in attoscience

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- **Metrology**
  - SAPs - Ponderomotive streaking
  - APTs - Two color interference - RABBIT

# Strong field physics in attoscience

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- **Metrology**
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  - APTs - Two color interference - RABBIT
    - Absolute phase effects in few cycle XUV pulses

# Strong field physics in attoscience

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- **Applications**

- Combine attosecond pulse with strong IR field.
- Above threshold attosecond electron wave packets.

# Strong field physics in attoscience

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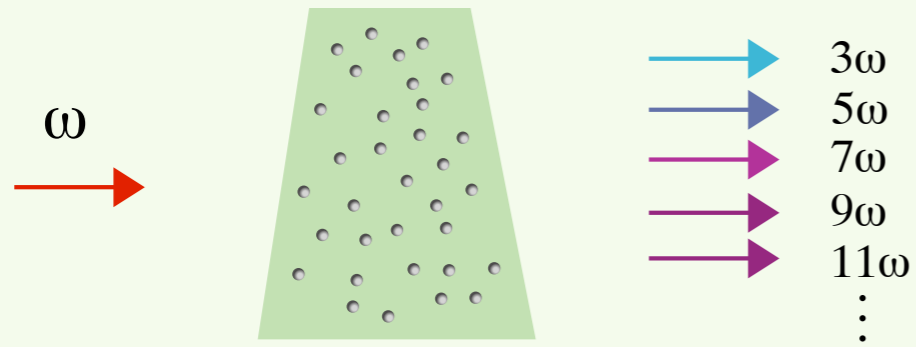
- **Applications**

- Combine attosecond pulse with strong IR field.
  - Above threshold attosecond electron wave packets.
    - Attosecond electron wave packet interferometry
    - Near threshold AEWPs - quantum path selection.
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# How are attosecond light pulses made?

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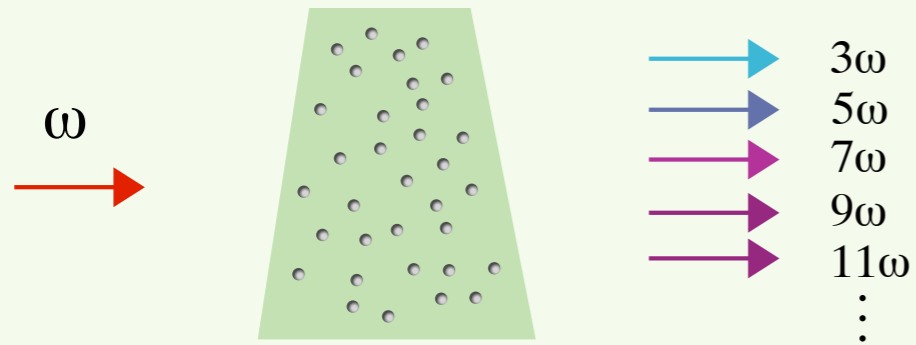
## High harmonic generation



Up conversion of IR laser light to coherent, short pulsed XUV light

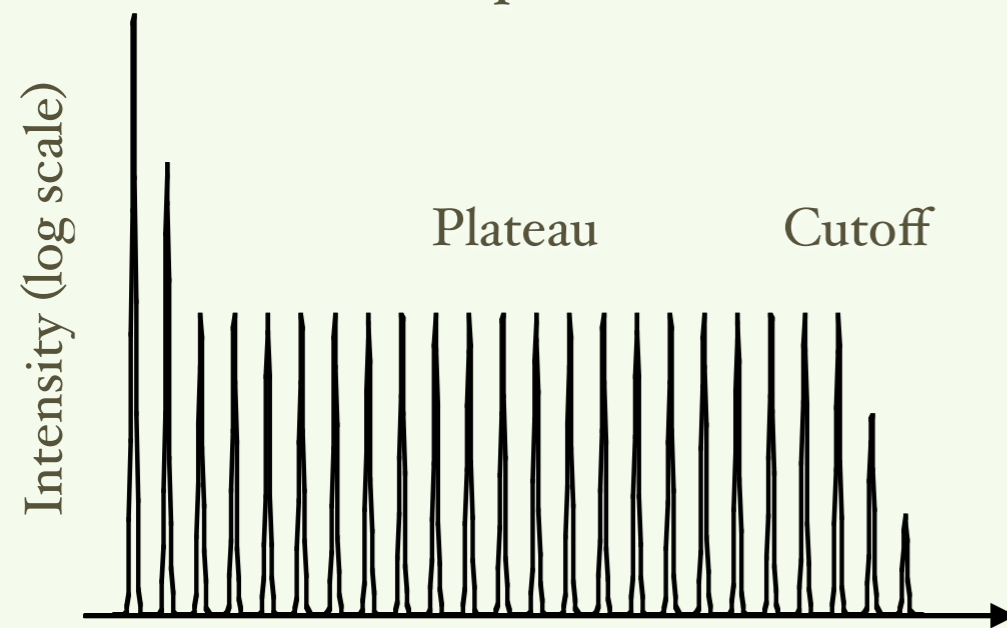
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## High harmonic generation



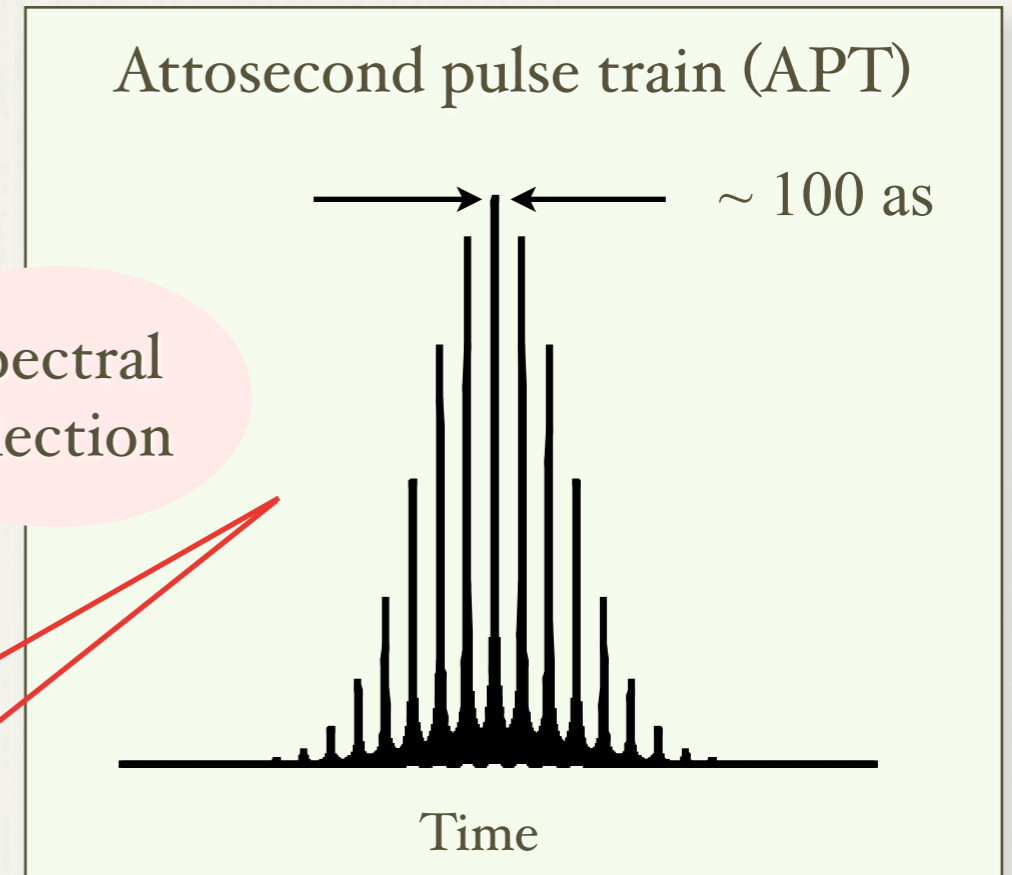
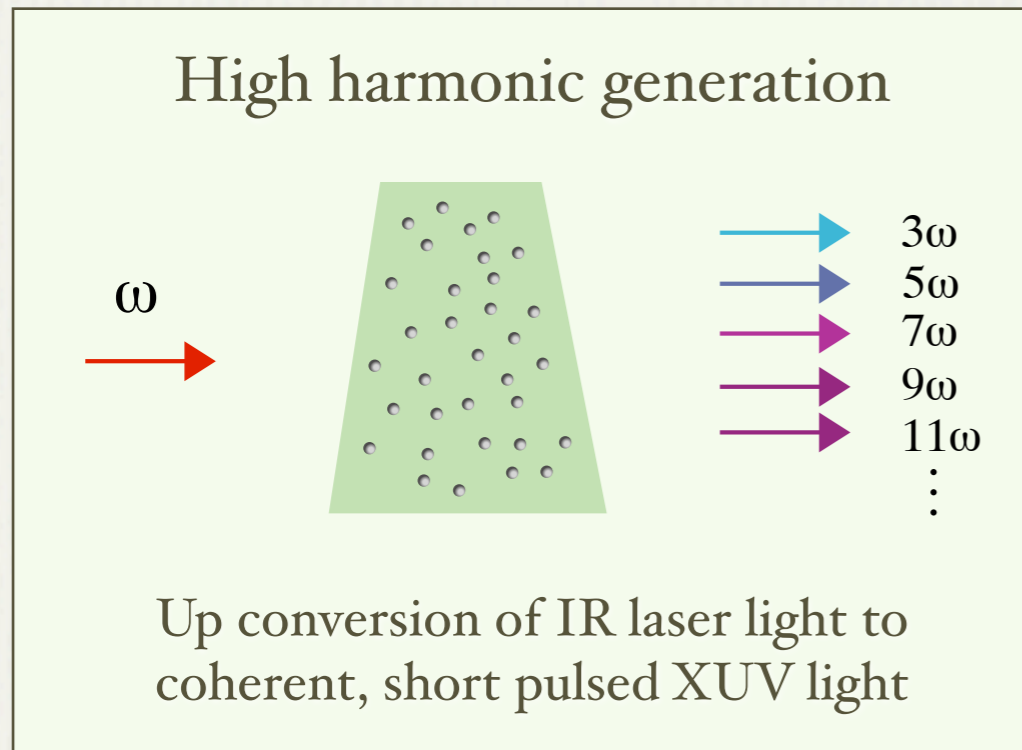
Up conversion of IR laser light to coherent, short pulsed XUV light

## Harmonic spectrum

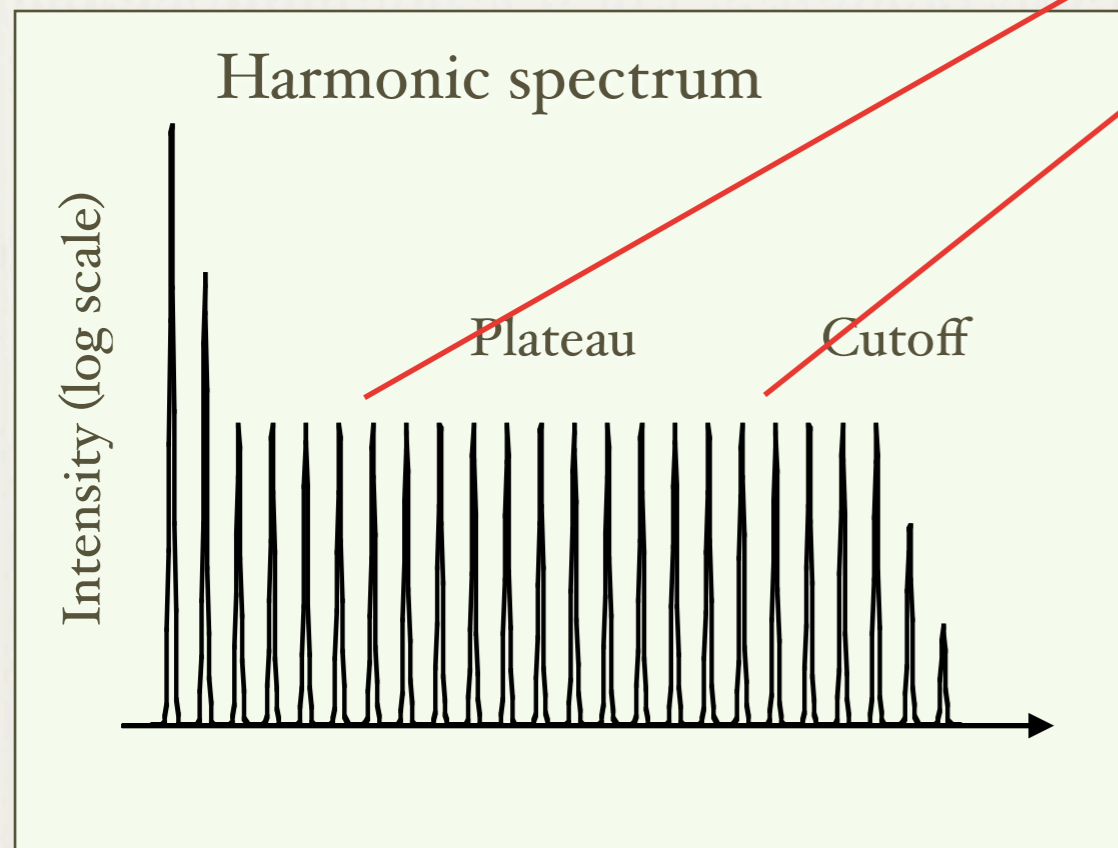




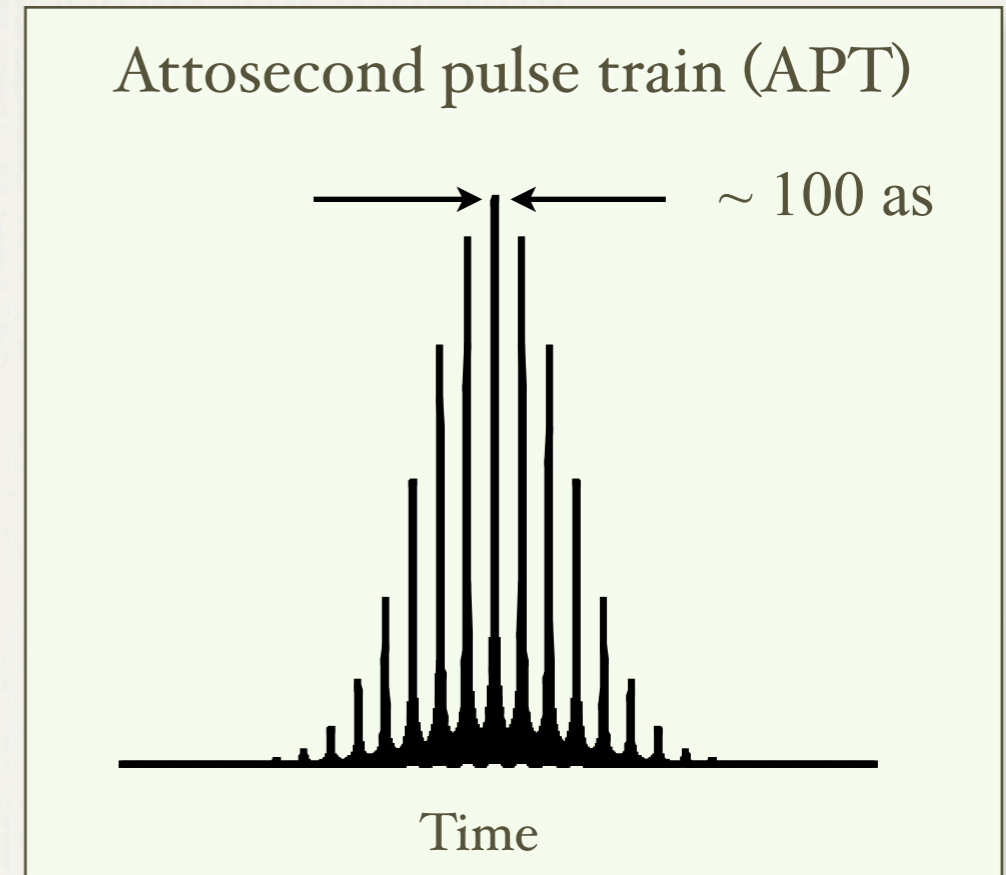
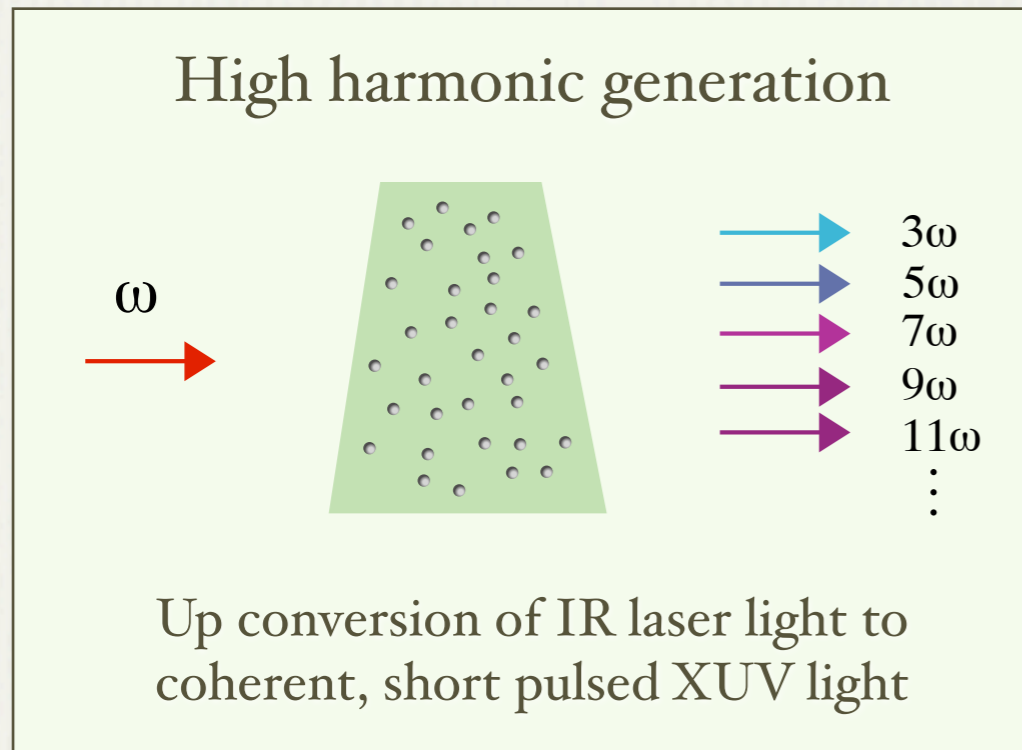
# How are attosecond light pulses made?



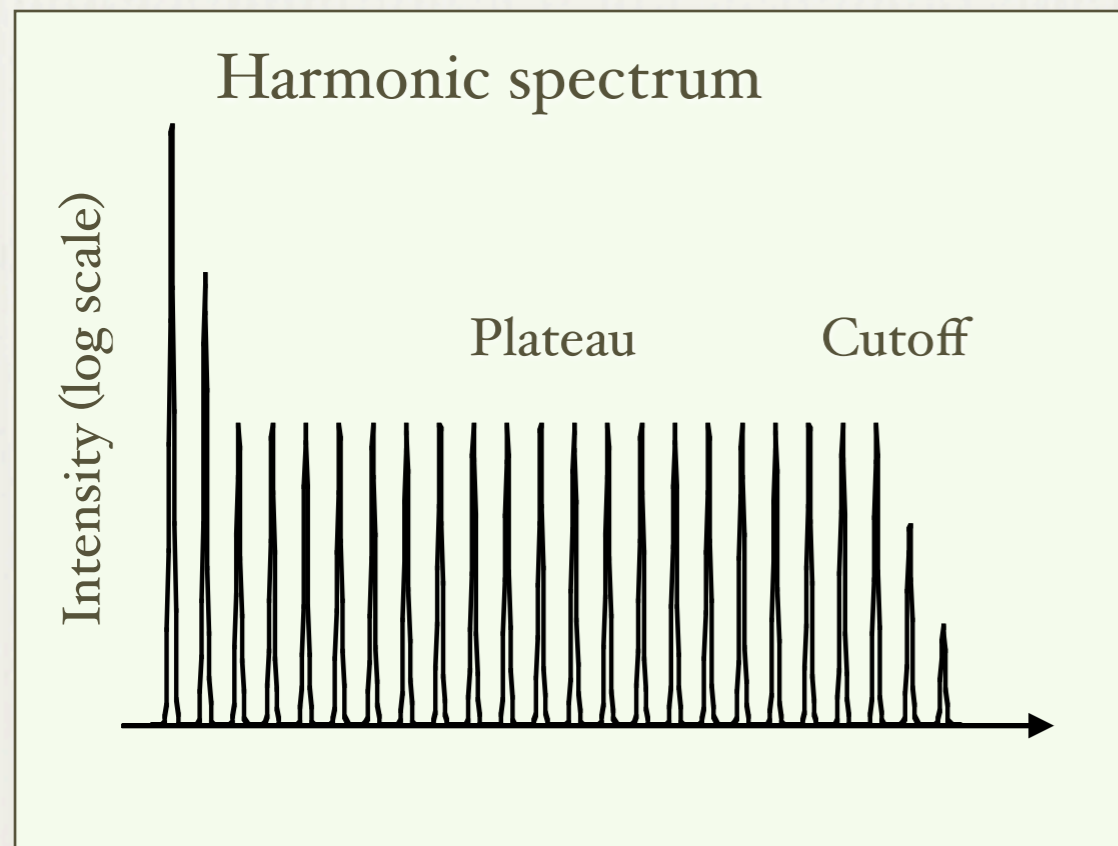
Paul et al, Science 2001



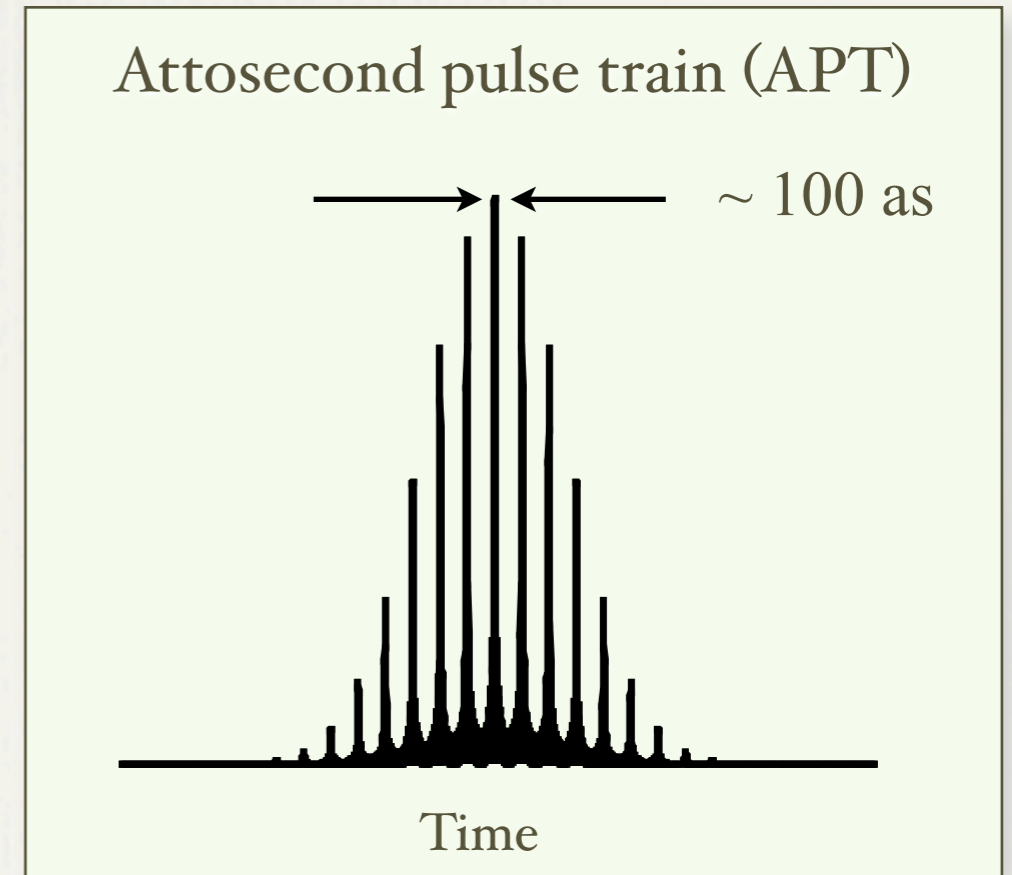
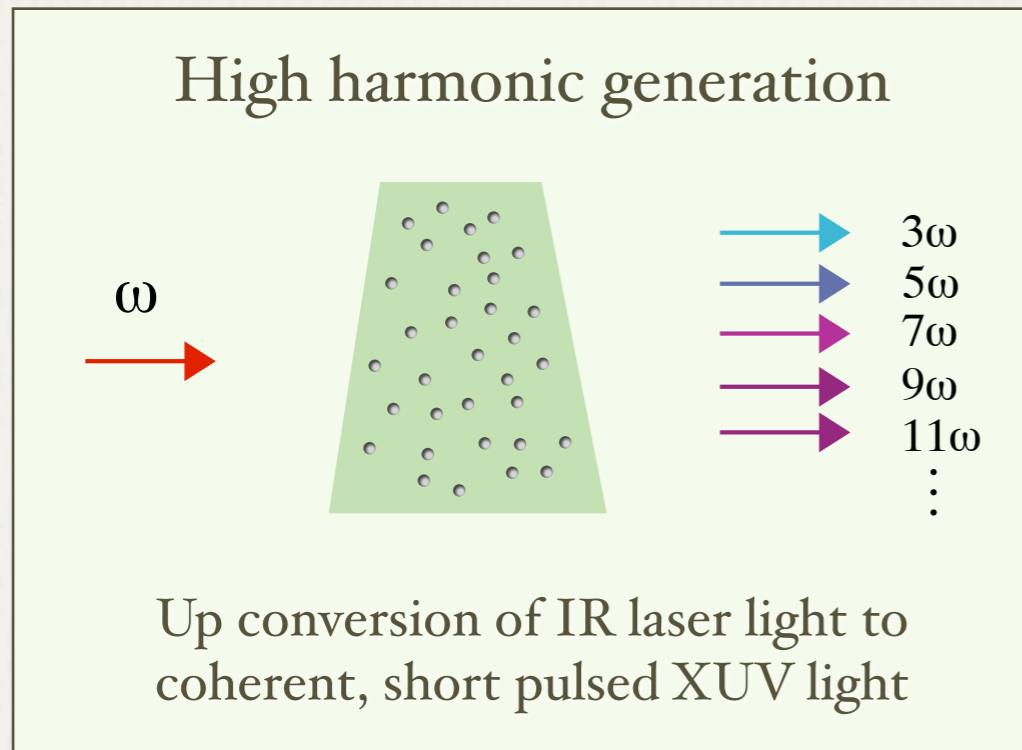
# How are attosecond light pulses made?



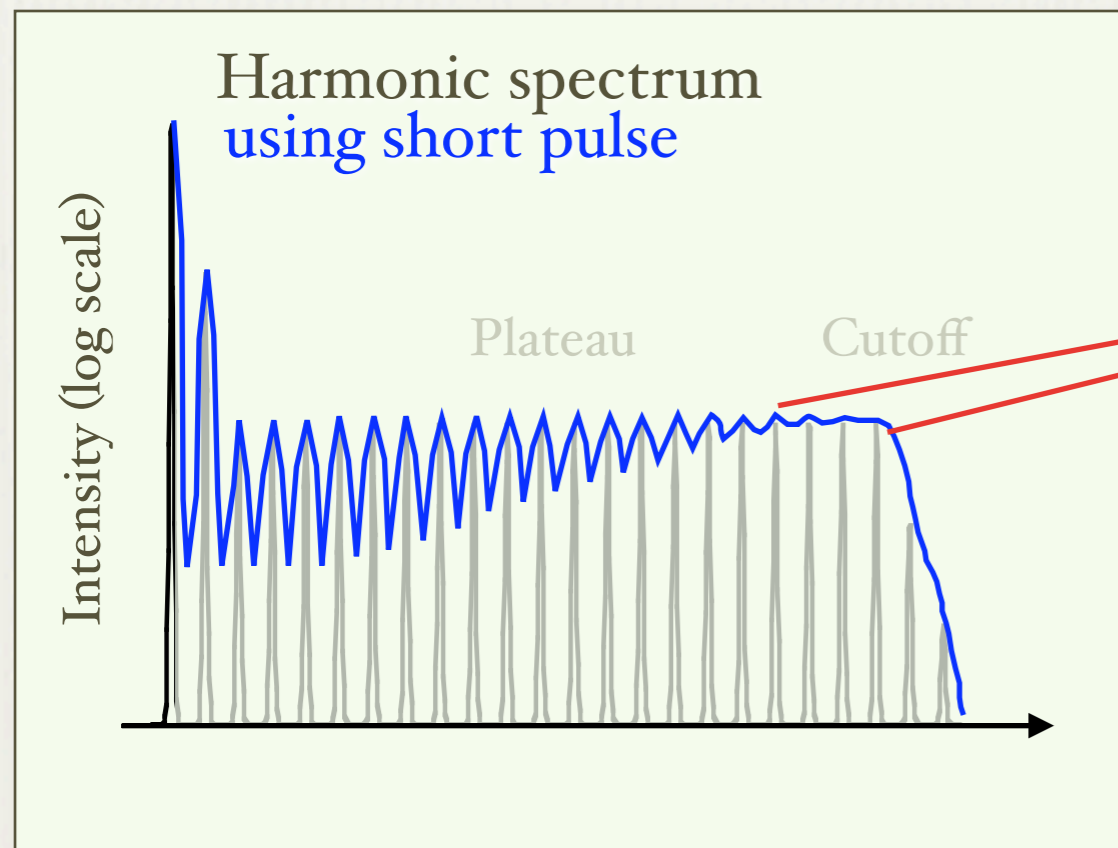
Paul et al, Science 2001



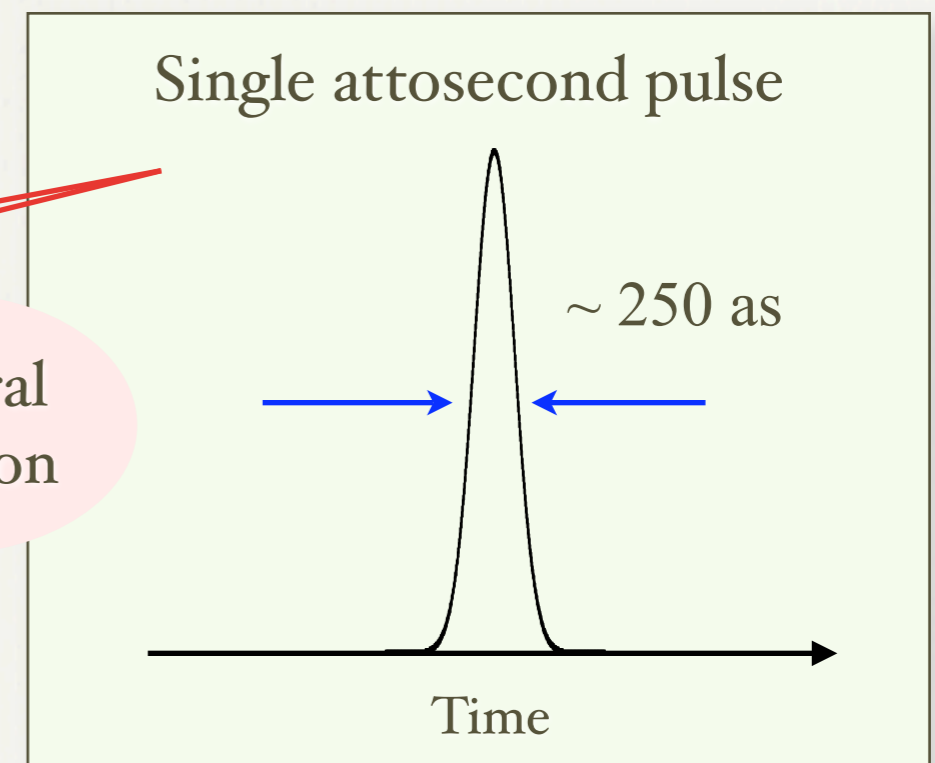
# How are attosecond light pulses made?



Paul et al, Science 2001



Spectral selection

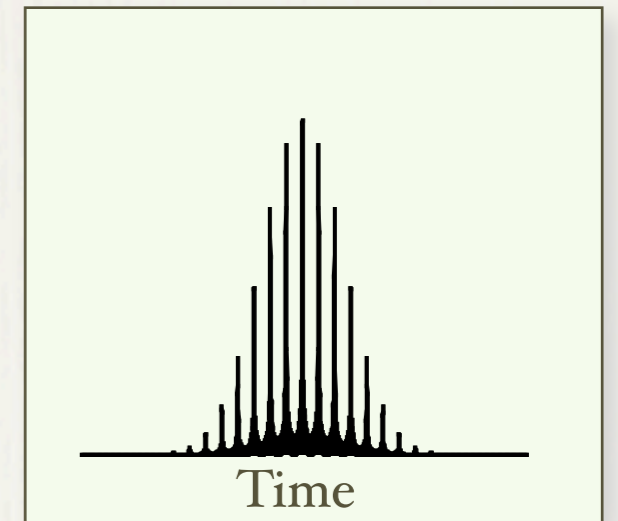


Krausz, et al.

# APTs: Phase locking

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$$\mathcal{E}_x(t) = \sum_q \mathcal{E}_q \sin(q[\omega_1 t - \phi_q] + \varphi)$$

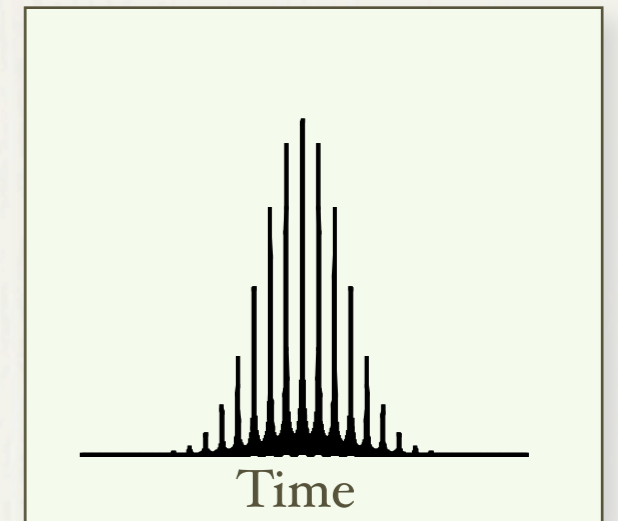


# APTs: Phase locking

$$\mathcal{E}_x(t) = \sum_q \mathcal{E}_q \sin(q[\omega_1 t - \phi_q] + \varphi)$$

q-dependent phase

C-E phase

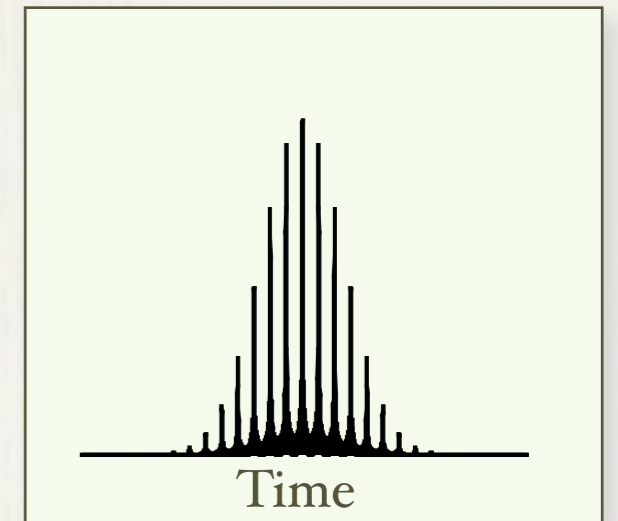


# APTs: Phase locking

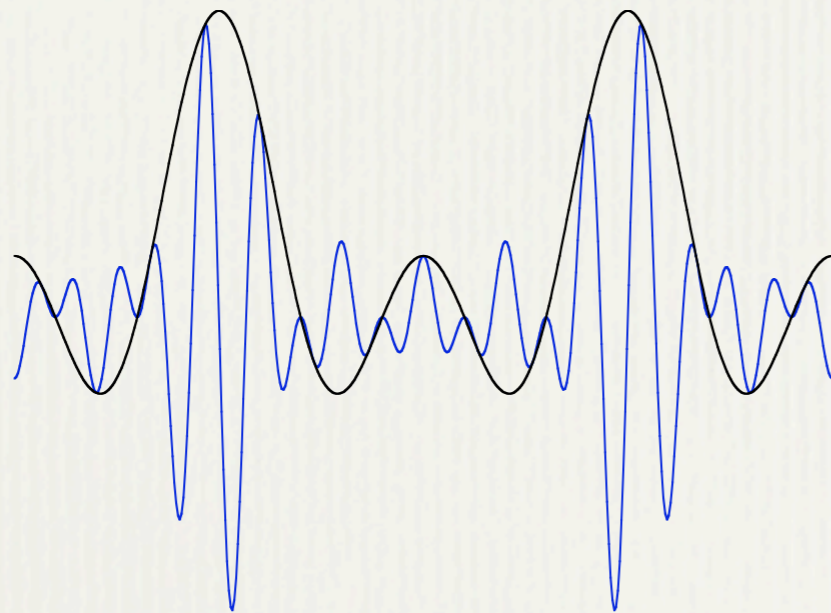
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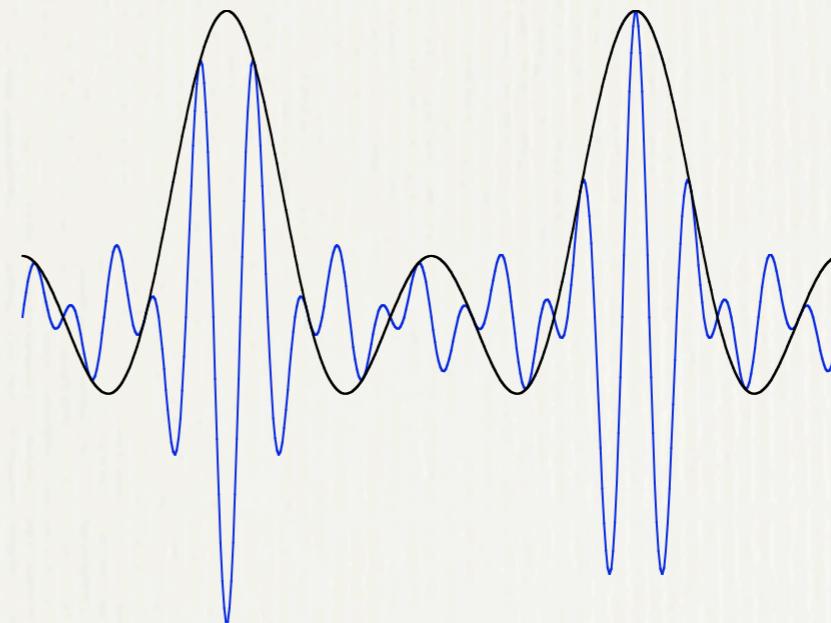
C-E phase



**APT electric fields**



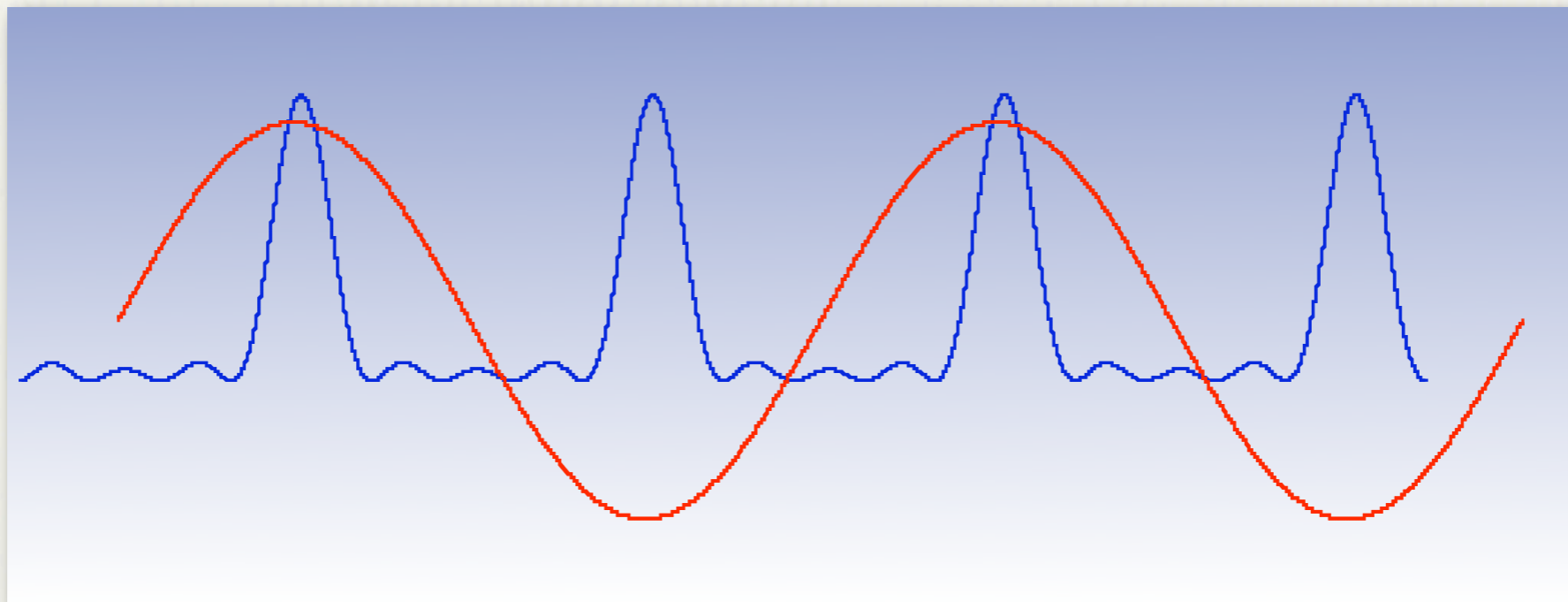
$\varphi = 0$



$\varphi = \pi/2$

## Example: Quantum Path Selection

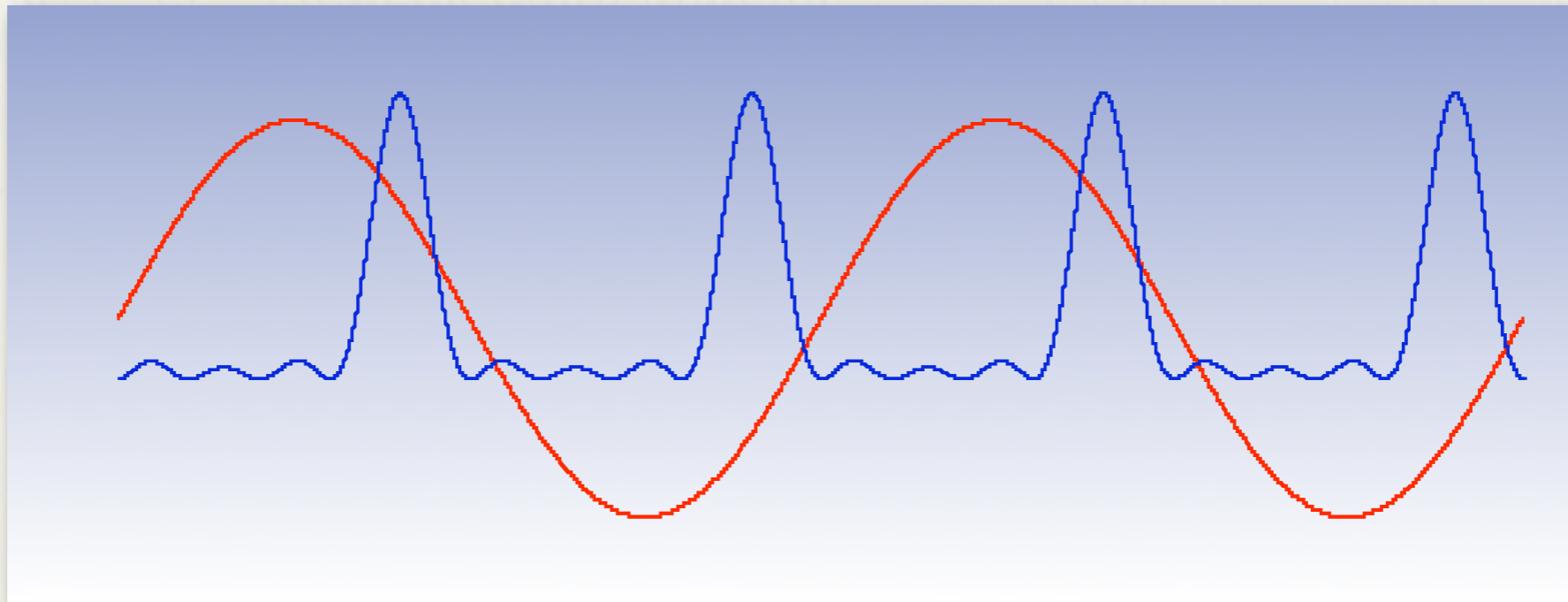
Idea = By controlling the time of ionization, we can choose one of several interfering contributions to a strong field process.



In general, we can hope to better understand and perhaps control strong field processes via this method.

## Example: Quantum Path Selection

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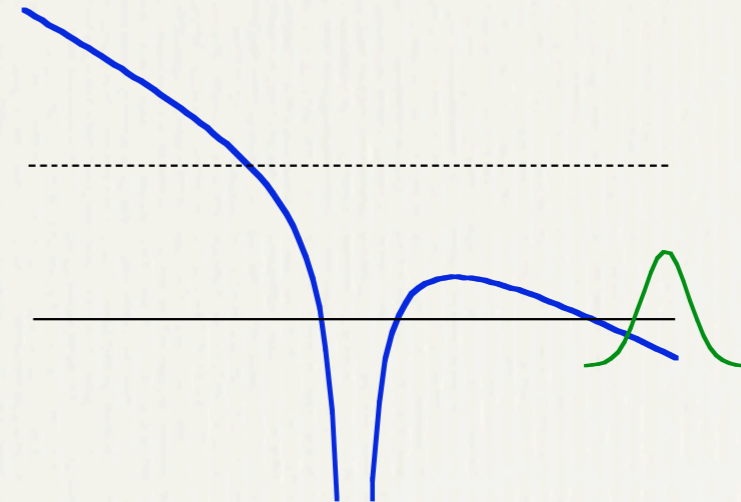


In general, we can hope to better understand and perhaps control strong field processes via this method.

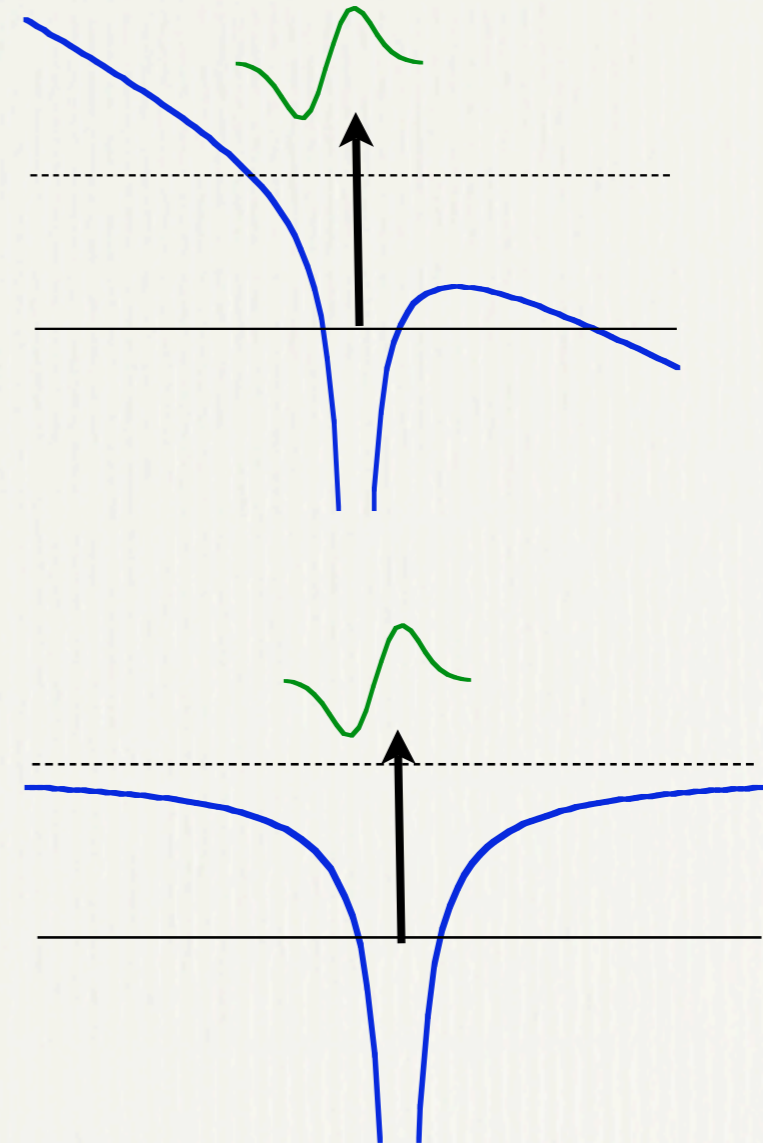


# Tunneling electron wave packets

- All tunneling WPs are "identical"
- Born near peak of IR cycle
- Duration depends on IR intensity
- Born outside potential well, with initial velocity  $v_0 = 0$
- Ionization step is highly non-linear  
- limits "low-IR" experiments

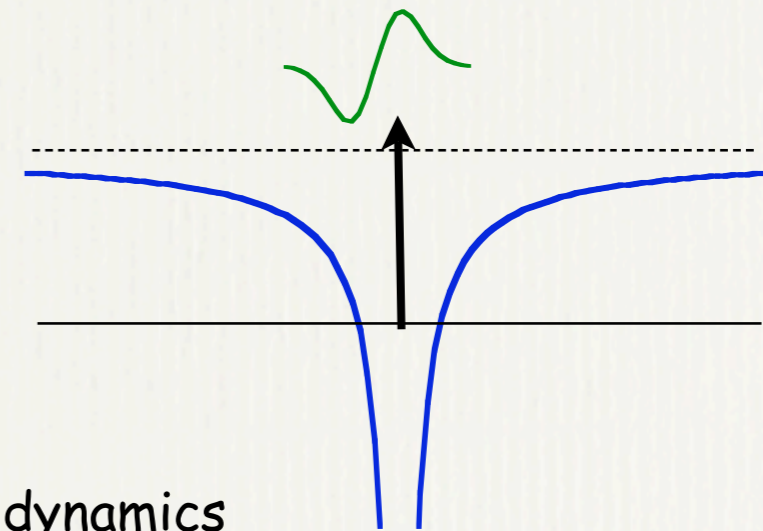
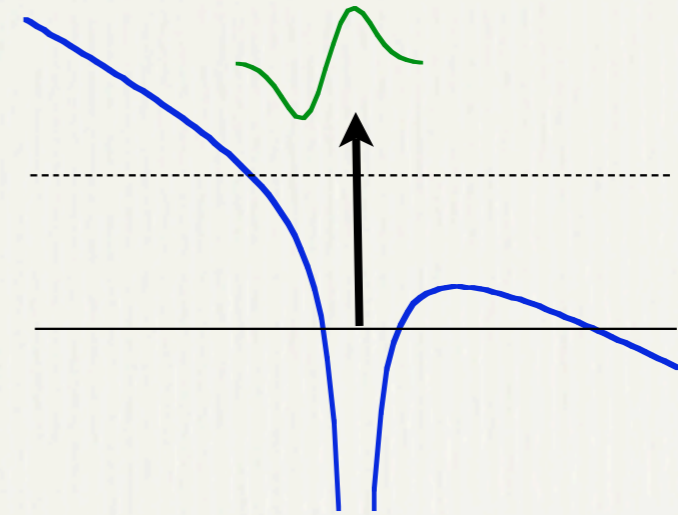


# APT electron wave packets



# APT electron wave packets

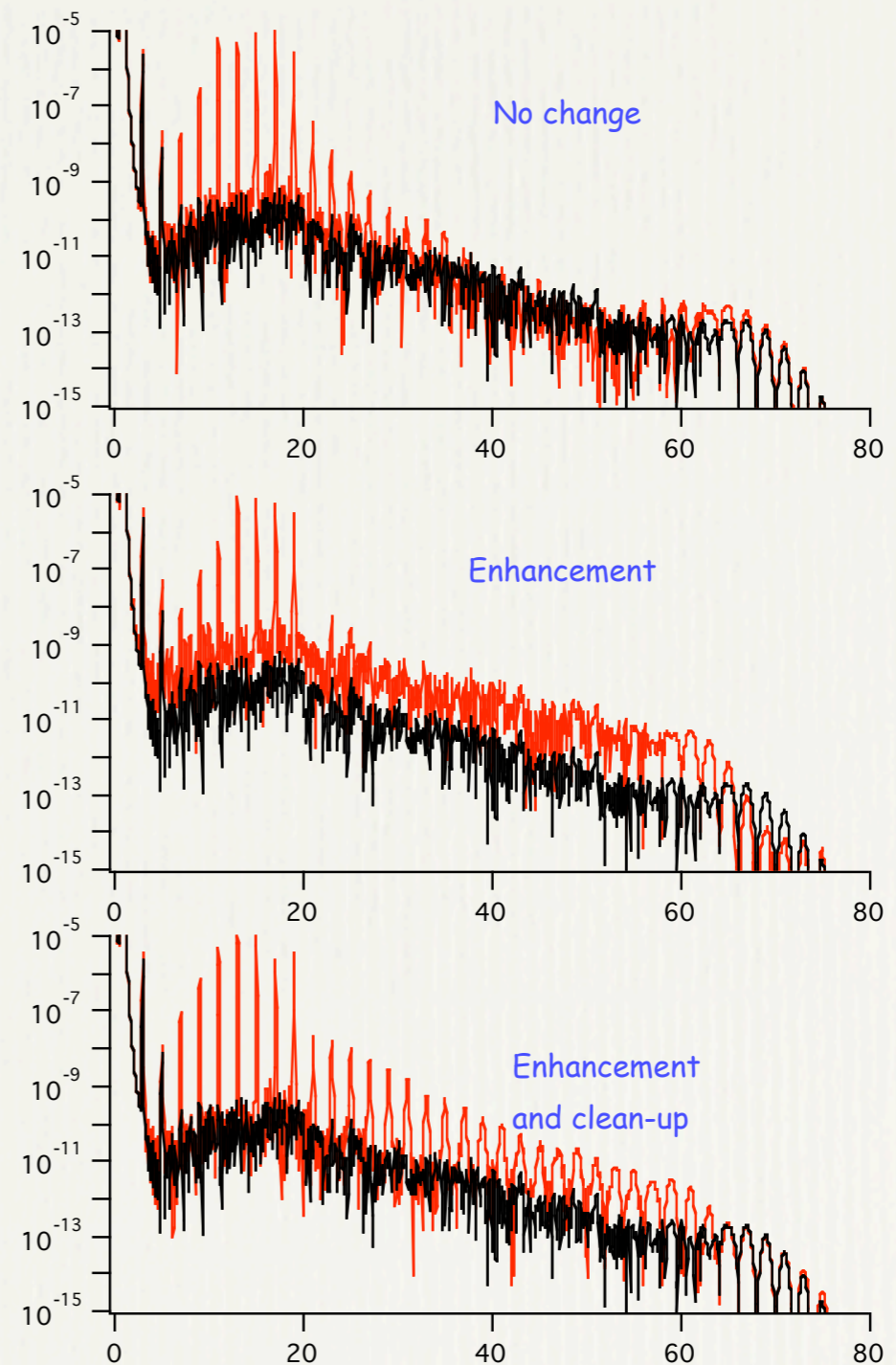
- Time of release determined by APT
- Properties can be tailored by APT
  - Duration
  - Non-zero  $v_0$  - can be left or right
  - Chirp (on attosecond scale)
- Can be born near or even below threshold
- Ionization step separate from (IR) strong field dynamics
  - study "weak" IR or "strong" IR dynamics



# Quantum Path Selection

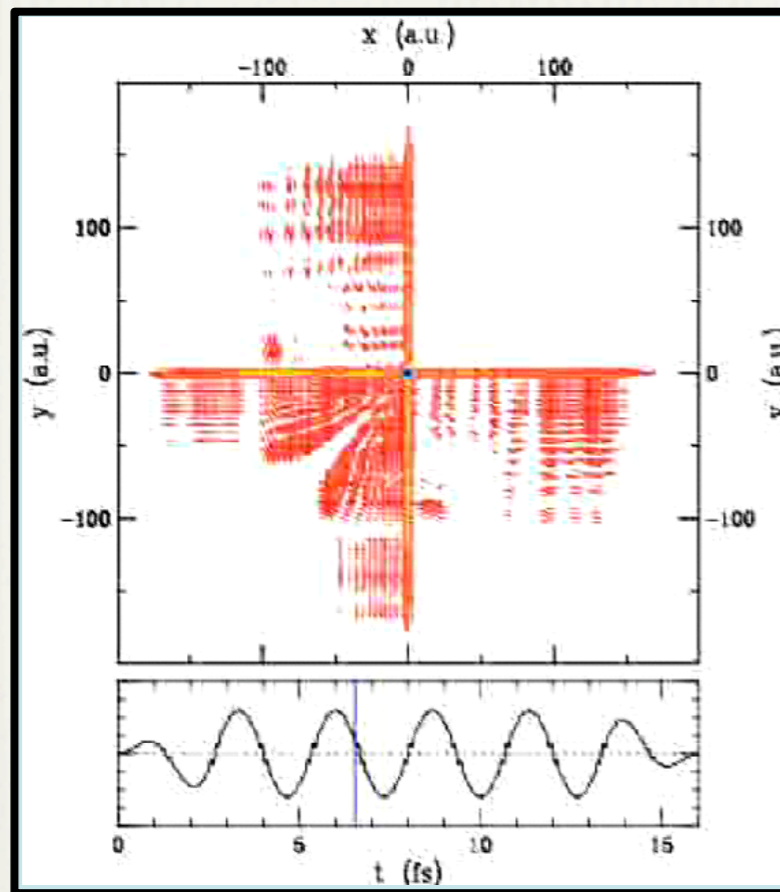
A previous theoretical study showed that QPS via APT driven ionization could be used to select a single contribution to the high harmonic spectrum.

Schafer et al, PRL 2004



# Example: Non-sequential double ionization

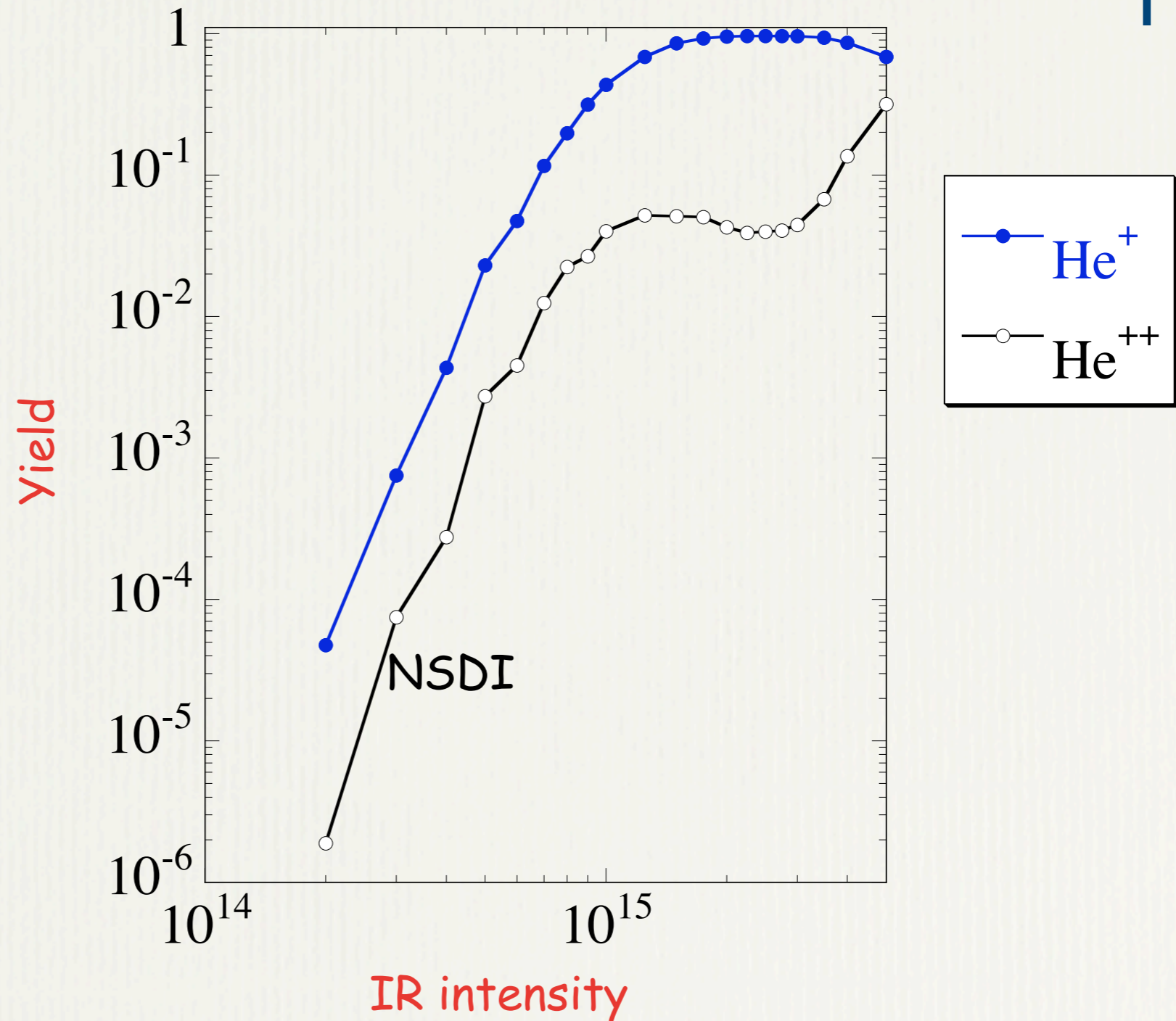
## The “Aligned” Helium Atom



	2-d He	6-d He
$E_0$ neutral	-2.898 au	-2.902 au
$E_0$ ion	-1.920 au	-2.000 au
1st I.P.	0.978 au	0.904 au

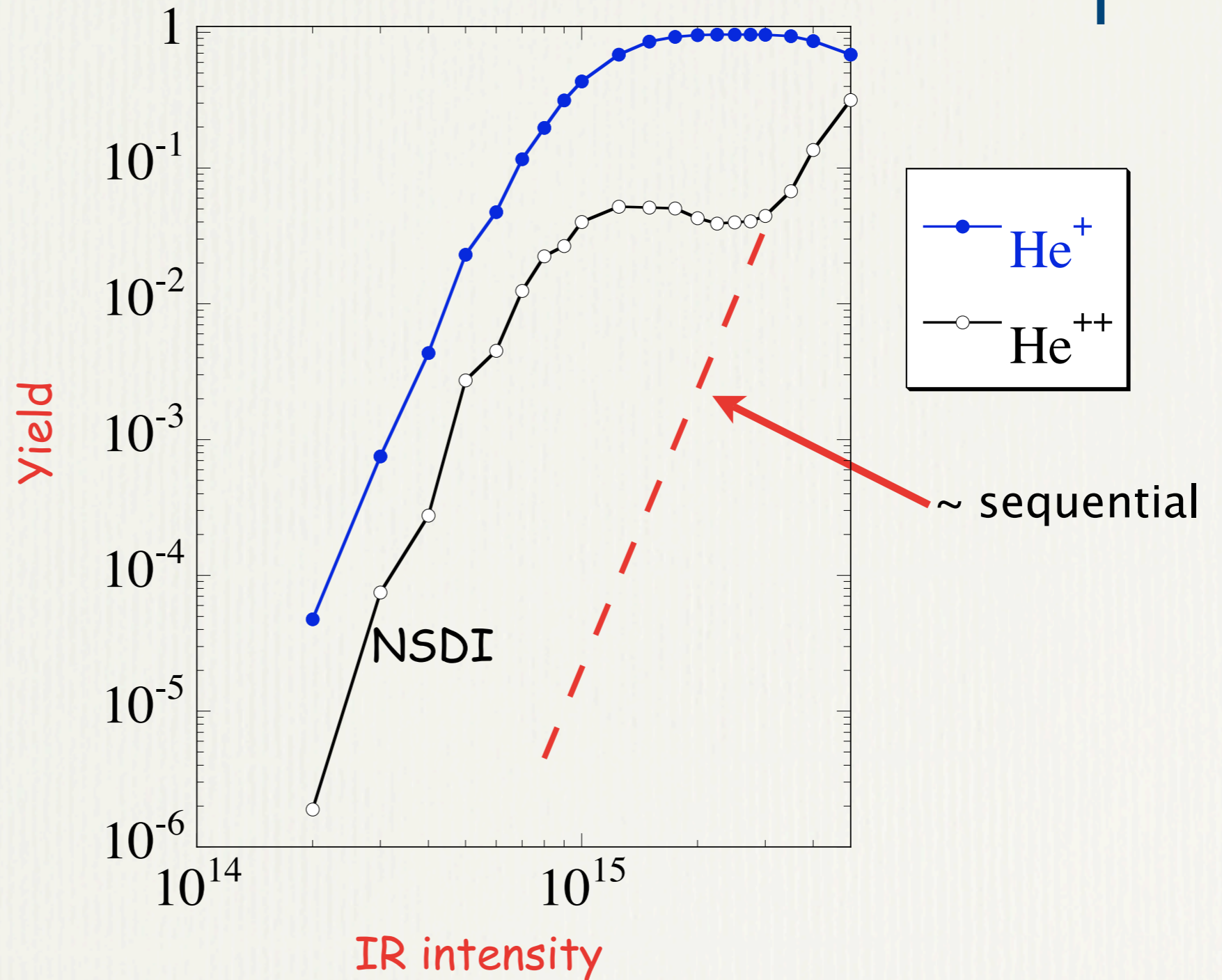
# Example: Non-sequential double ionization

The AEM reproduces the NSDI "knee" structure

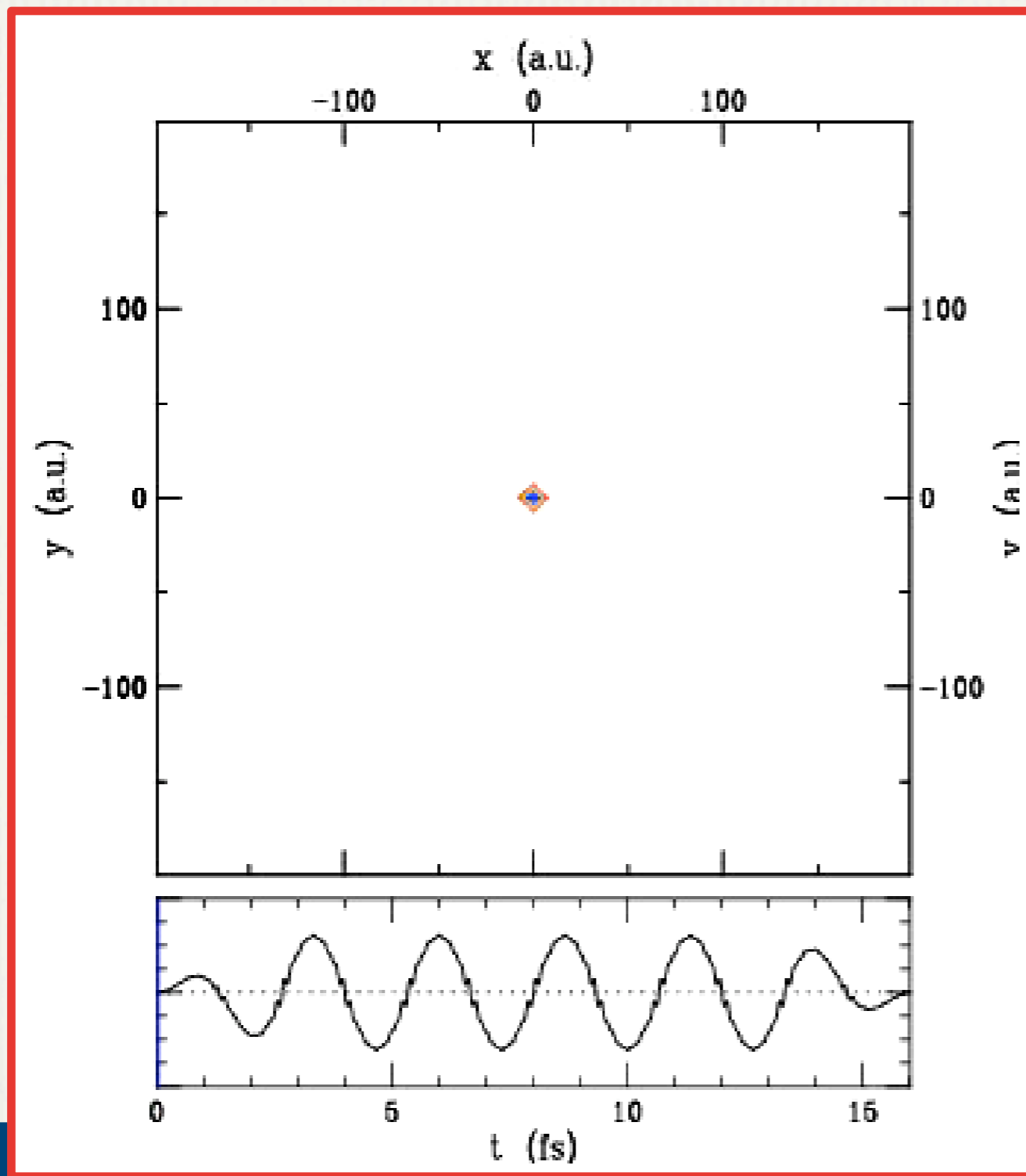


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# Example: Non-sequential double ionization



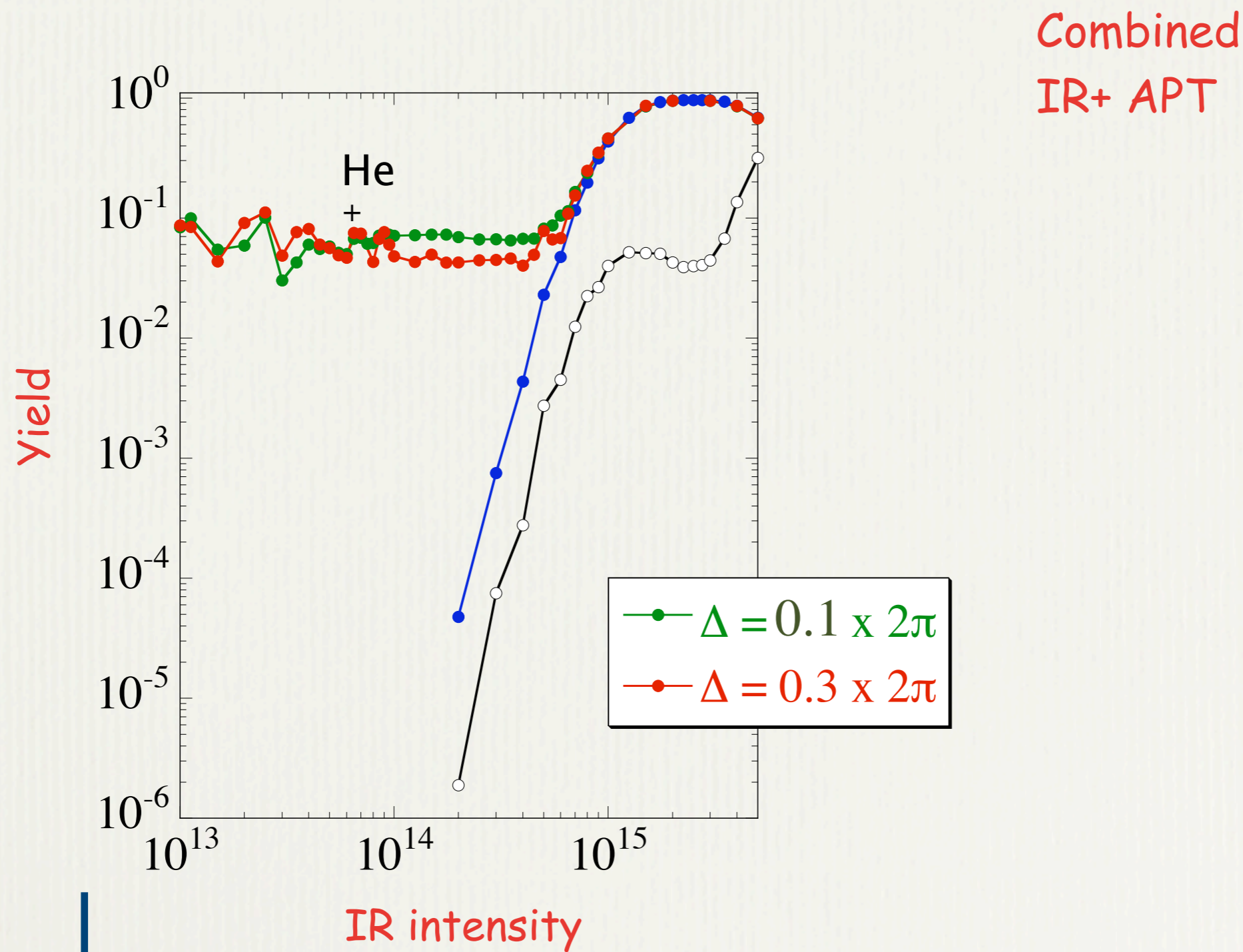
APT = harmonics 11-19

$$I_r = 5 \times 10^{14} \text{ W/cm}^2$$

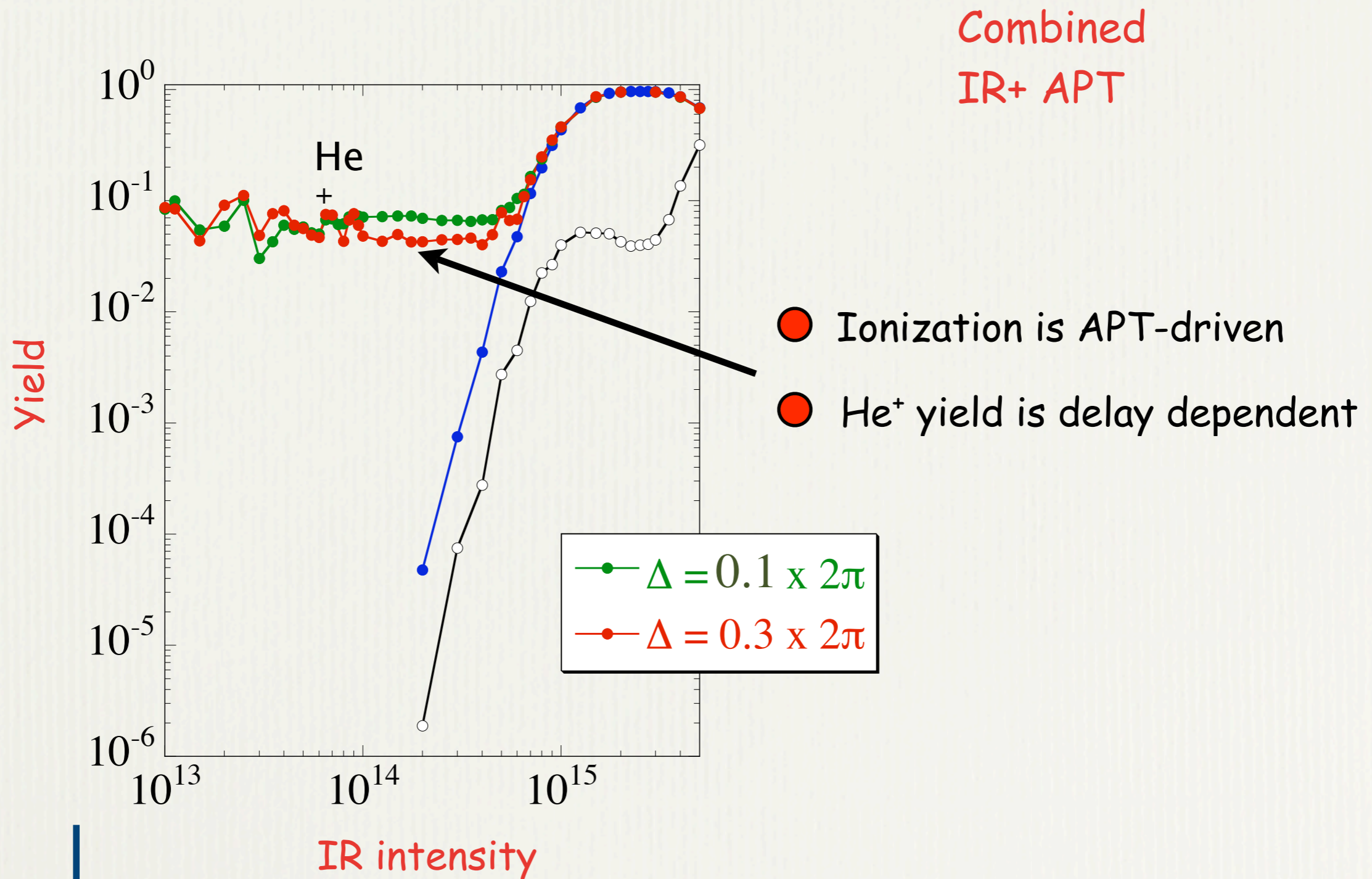
$$\text{APT} = 1 \times 10^{13} \text{ W/cm}^2$$



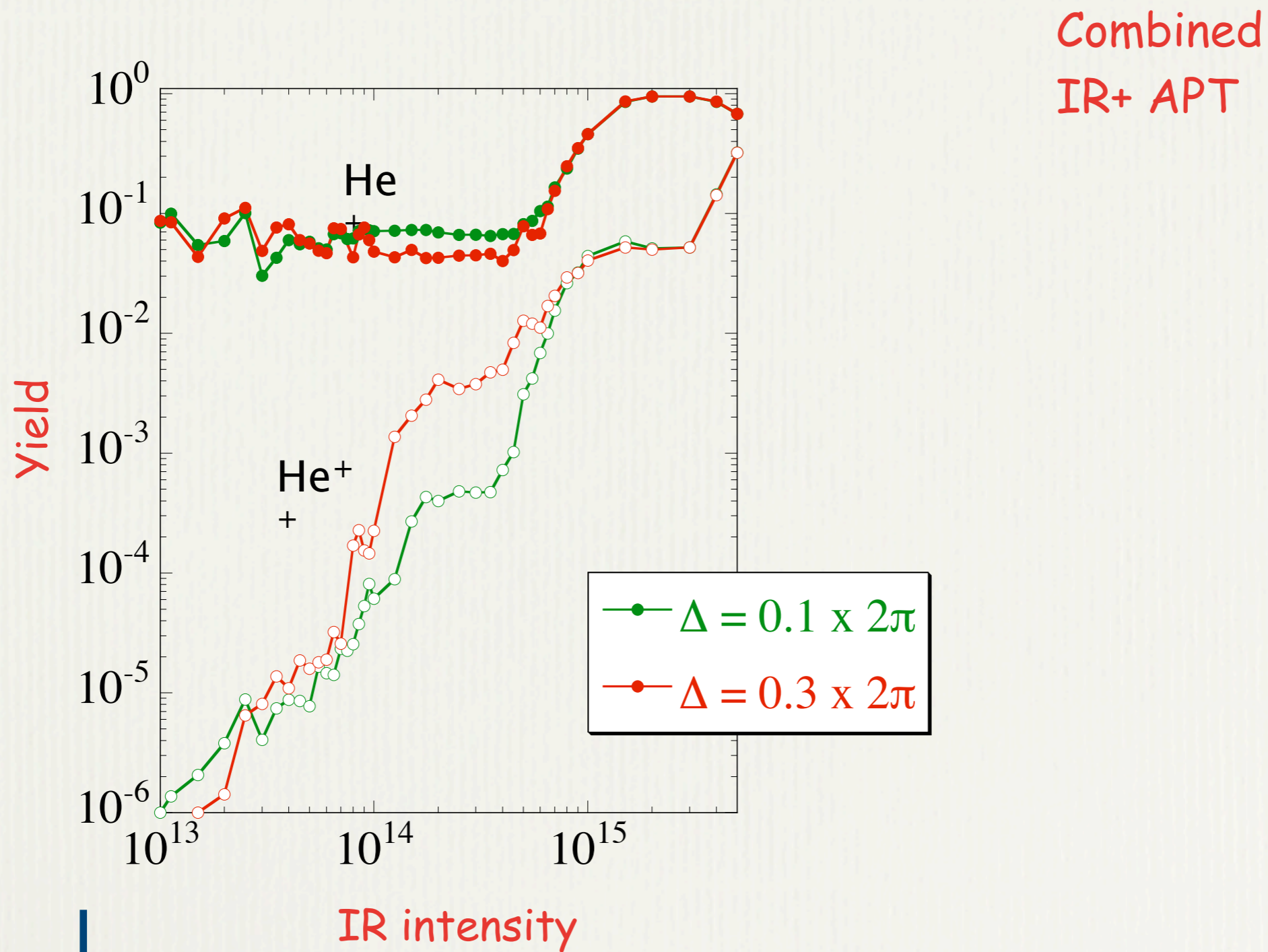
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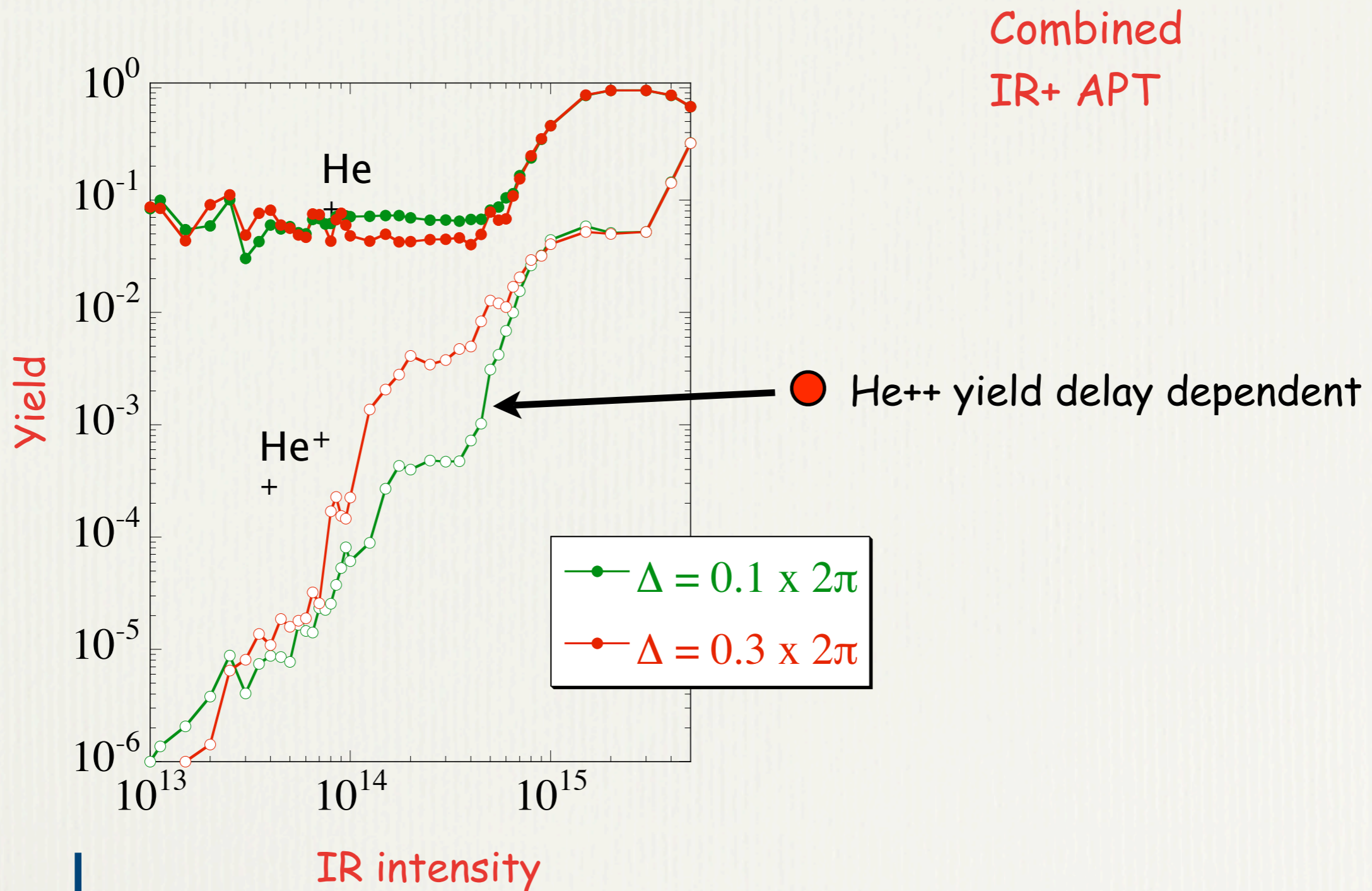
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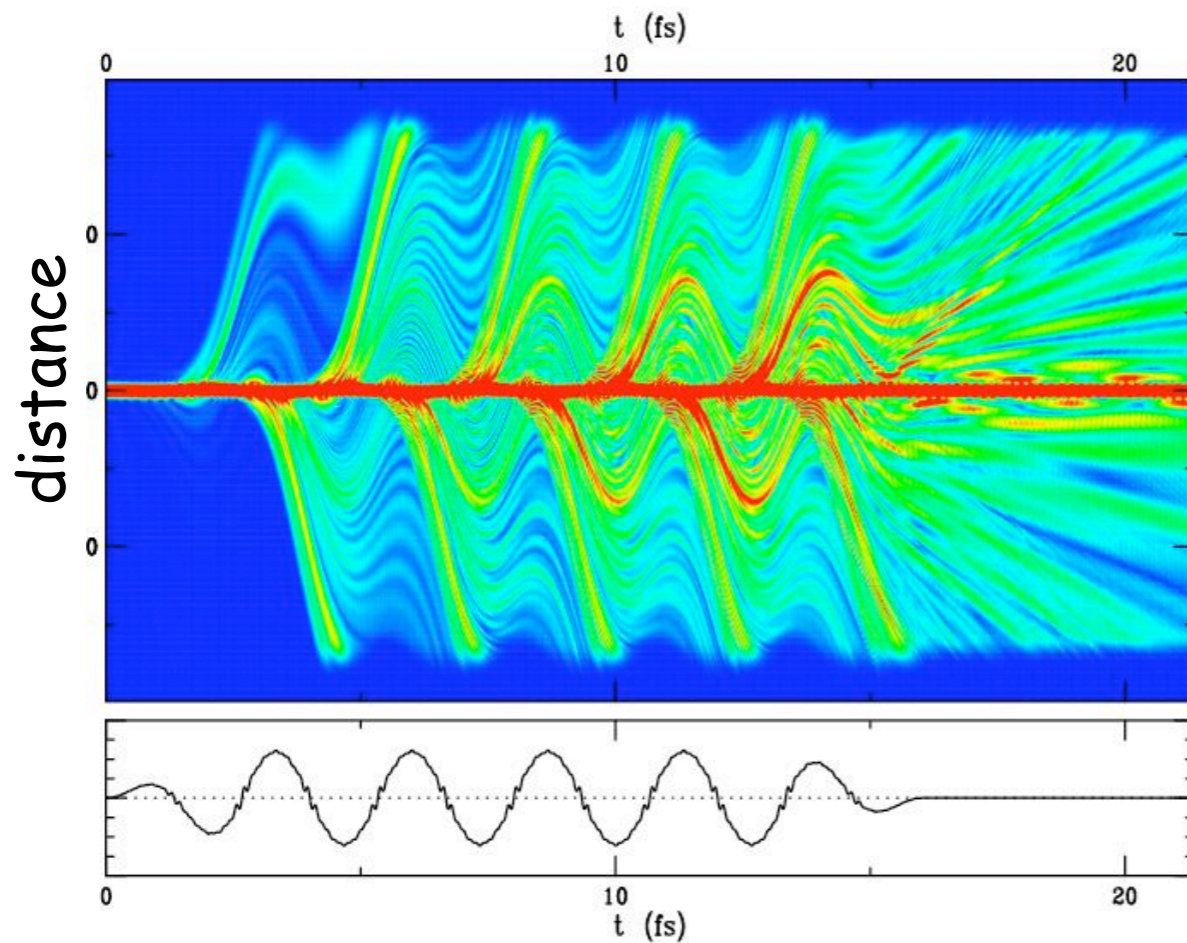


# Example: Non-sequential double ionization

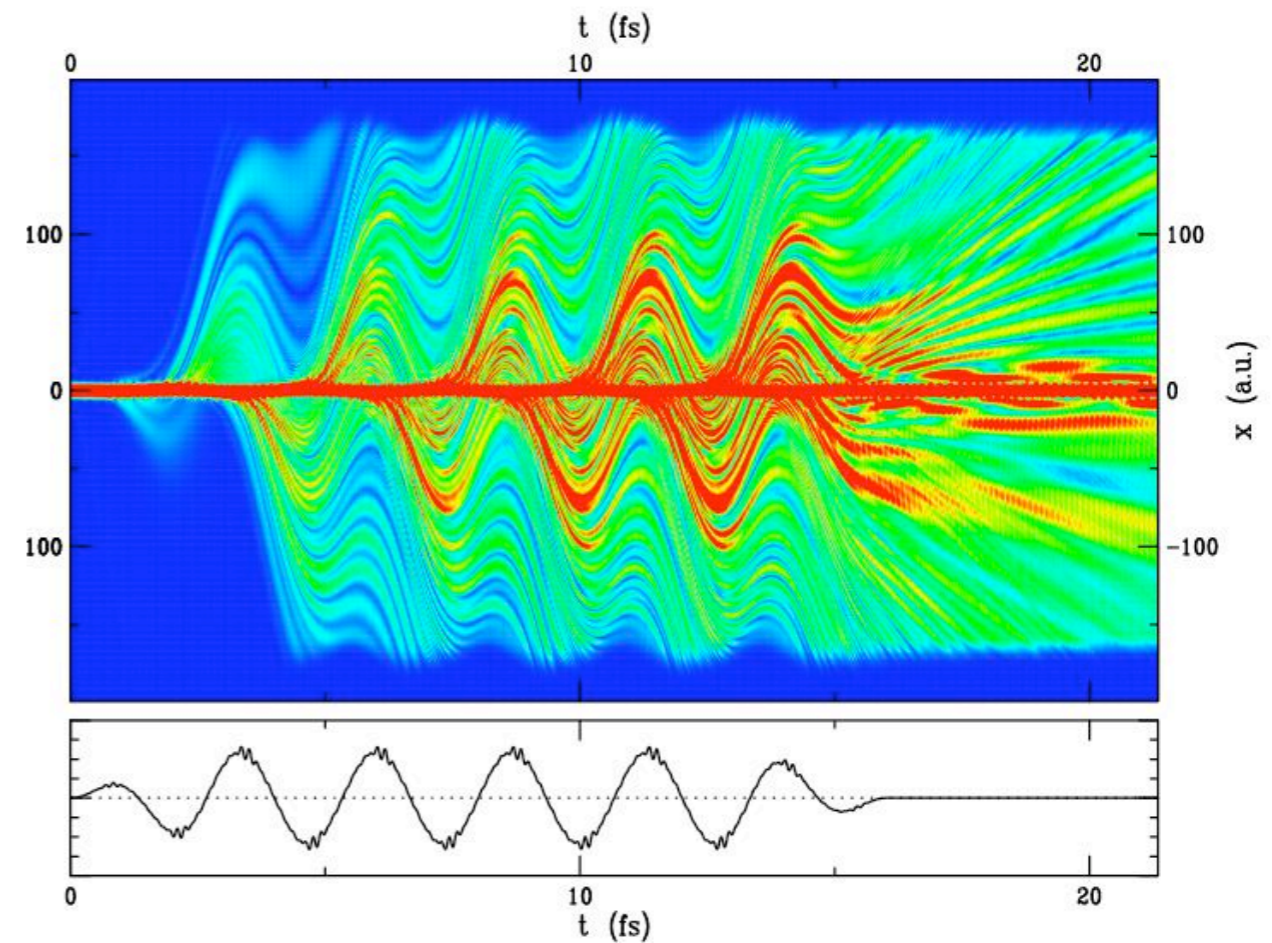


# Example: Non-sequential double ionization

Comparing electron trajectories from 2 different IR-APT delays

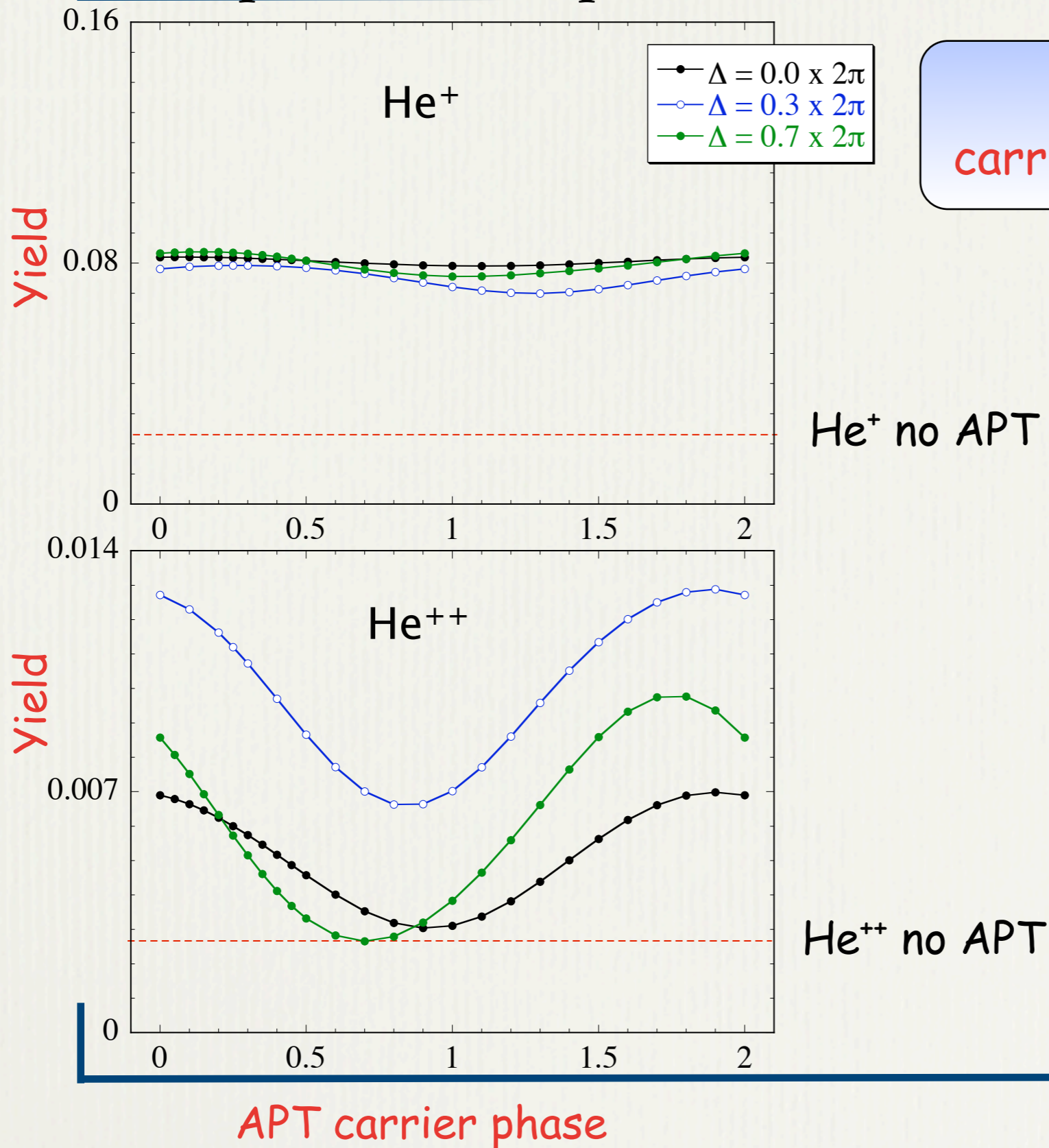


$$\Delta = 0.0$$



$$\Delta = 0.3$$

# Example: Non-sequential double ionization



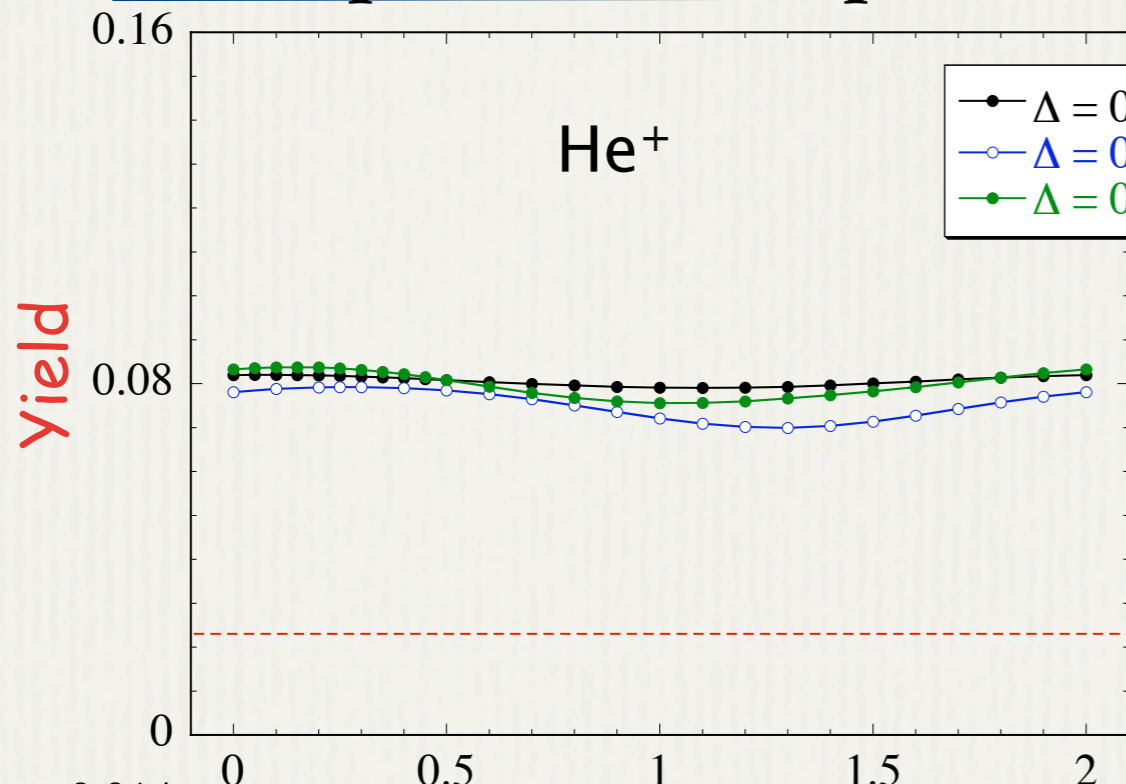
Effect of carrier envelope phase

$\text{He}^+$  no APT

$\text{He}^{++}$  no APT

APT carrier phase

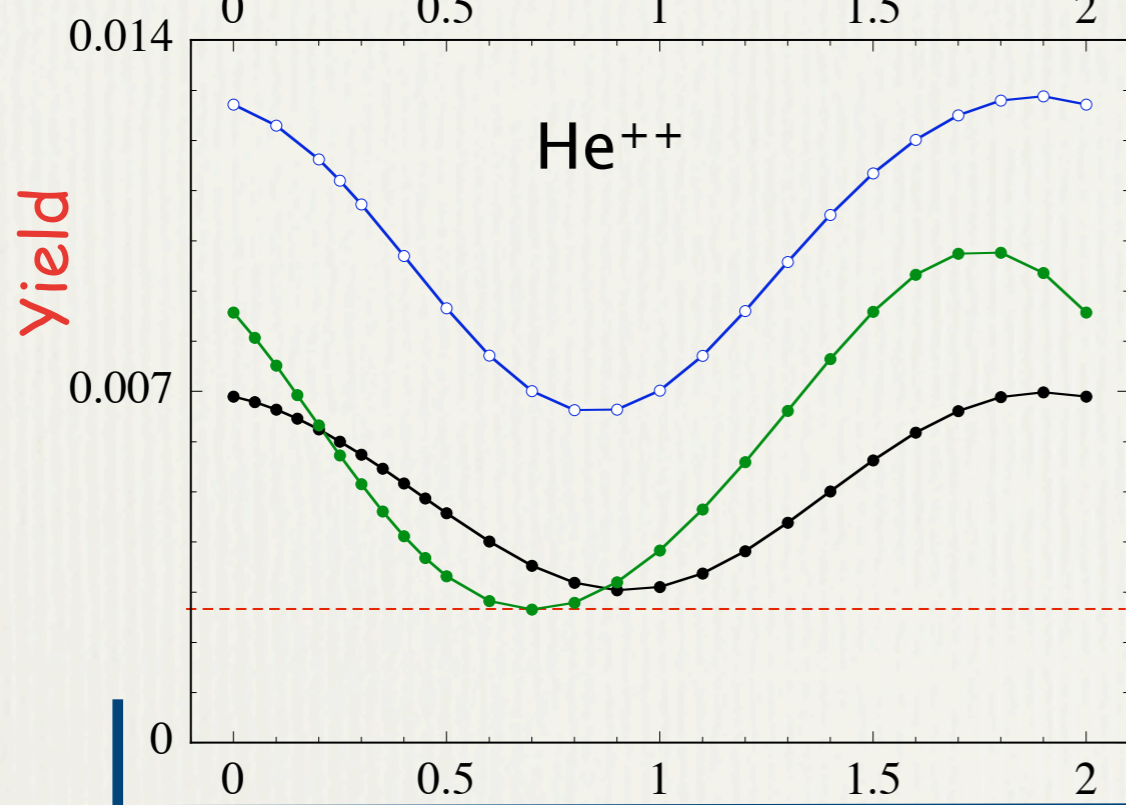
# Example: Non-sequential double ionization



Effect of carrier envelope phase

● He<sup>+</sup> insensitive to APT CEP

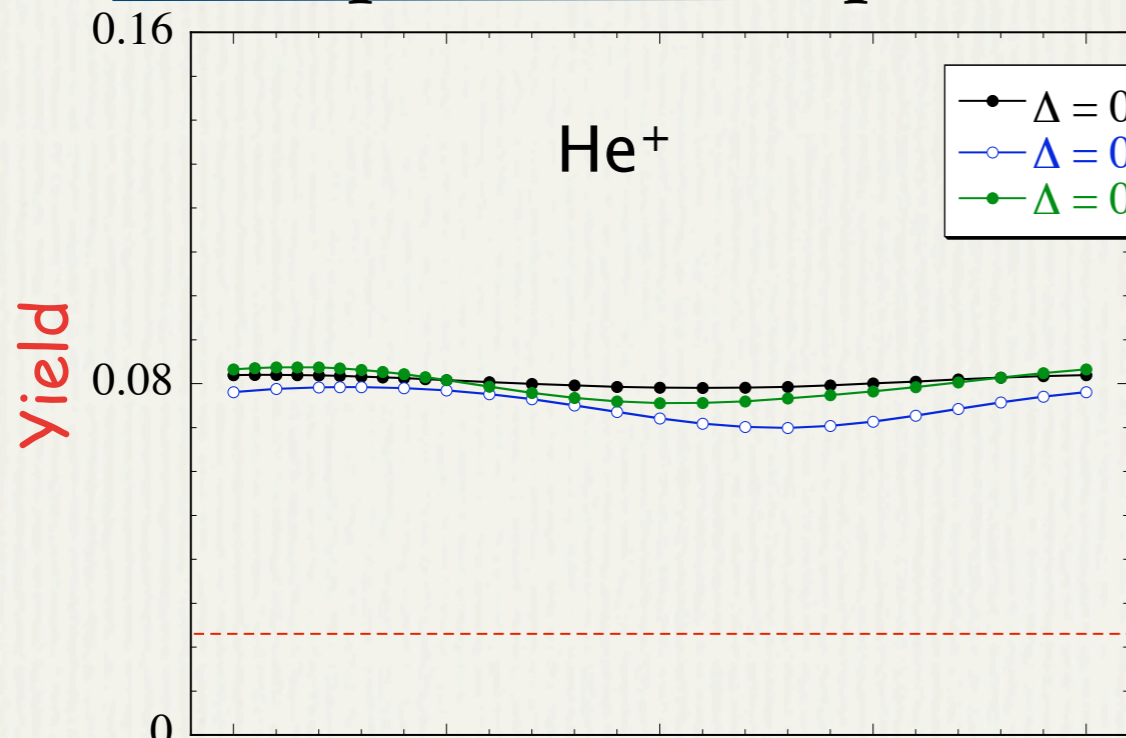
He<sup>+</sup> no APT



He<sup>++</sup> no APT

APT carrier phase

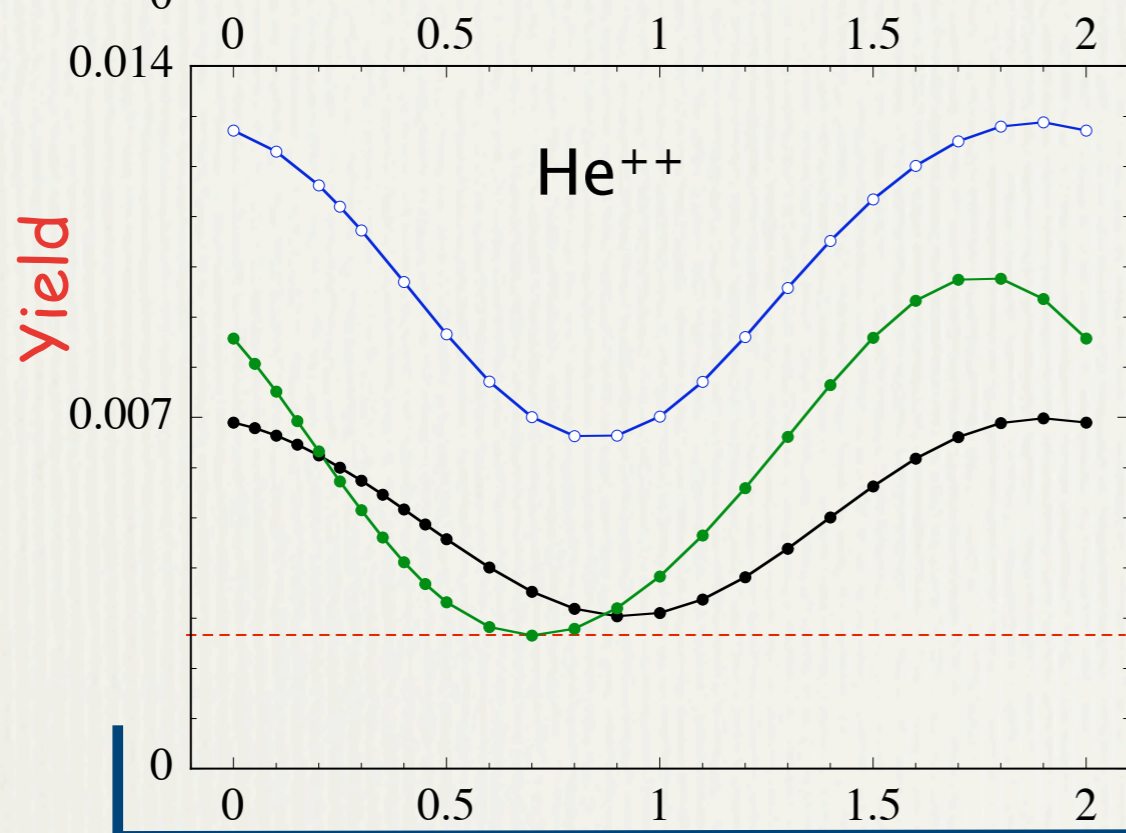
# Example: Non-sequential double ionization



Effect of carrier envelope phase

● He<sup>+</sup> insensitive to APT CEP

He<sup>+</sup> no APT



● He<sup>++</sup> very sensitive to APT CEP

He<sup>++</sup> no APT

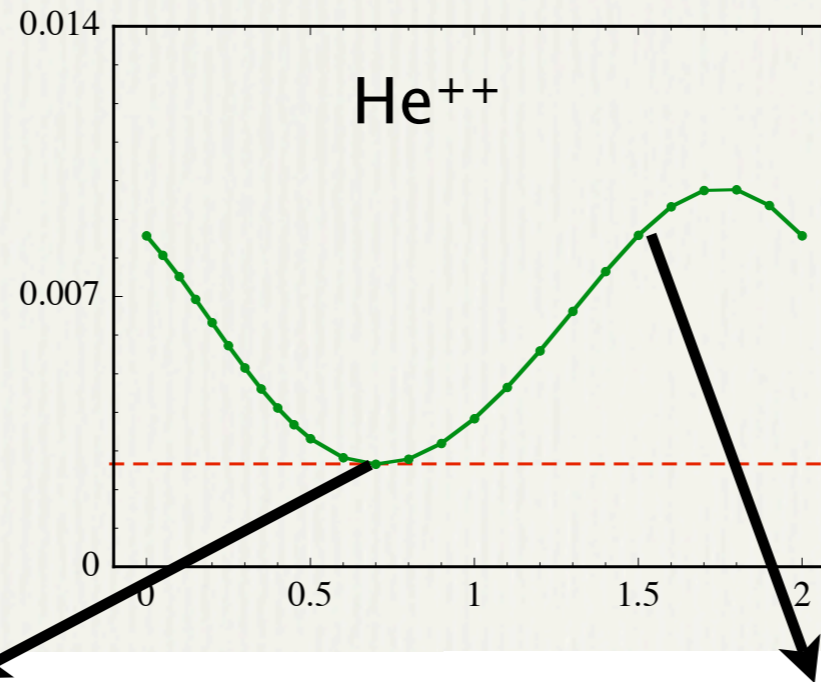
APT carrier phase



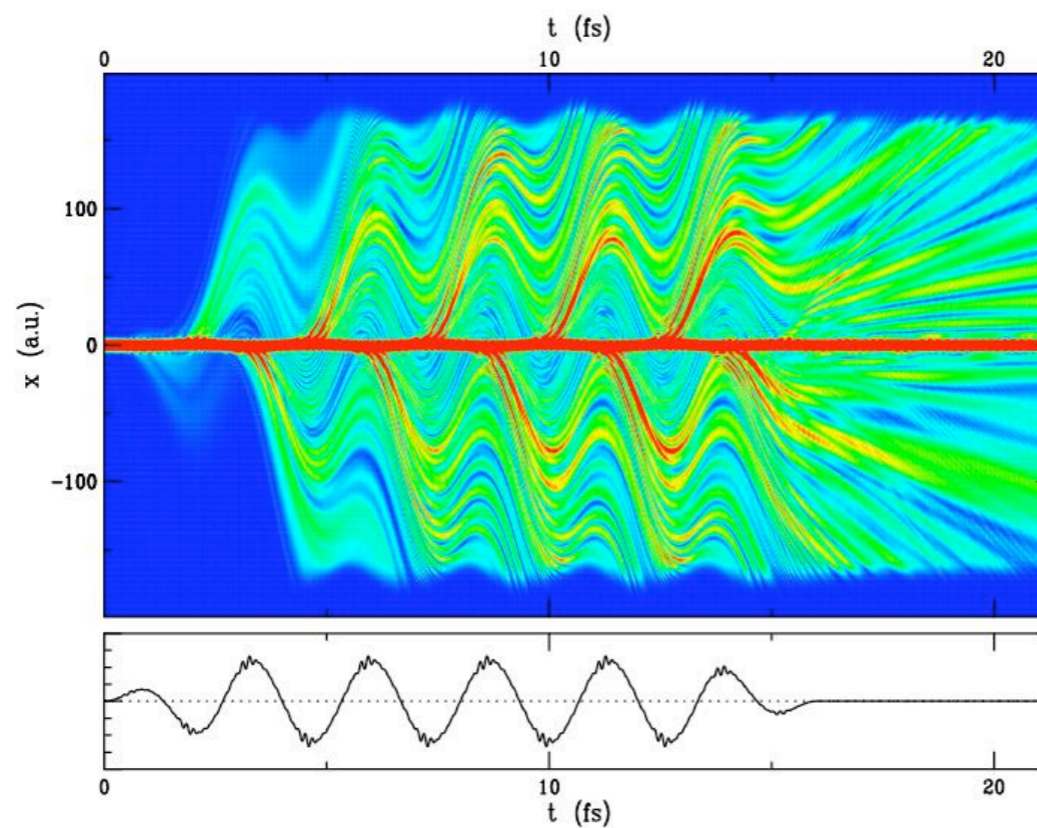
# Example: Non-sequential double ionization

$$\Delta = 0.7$$

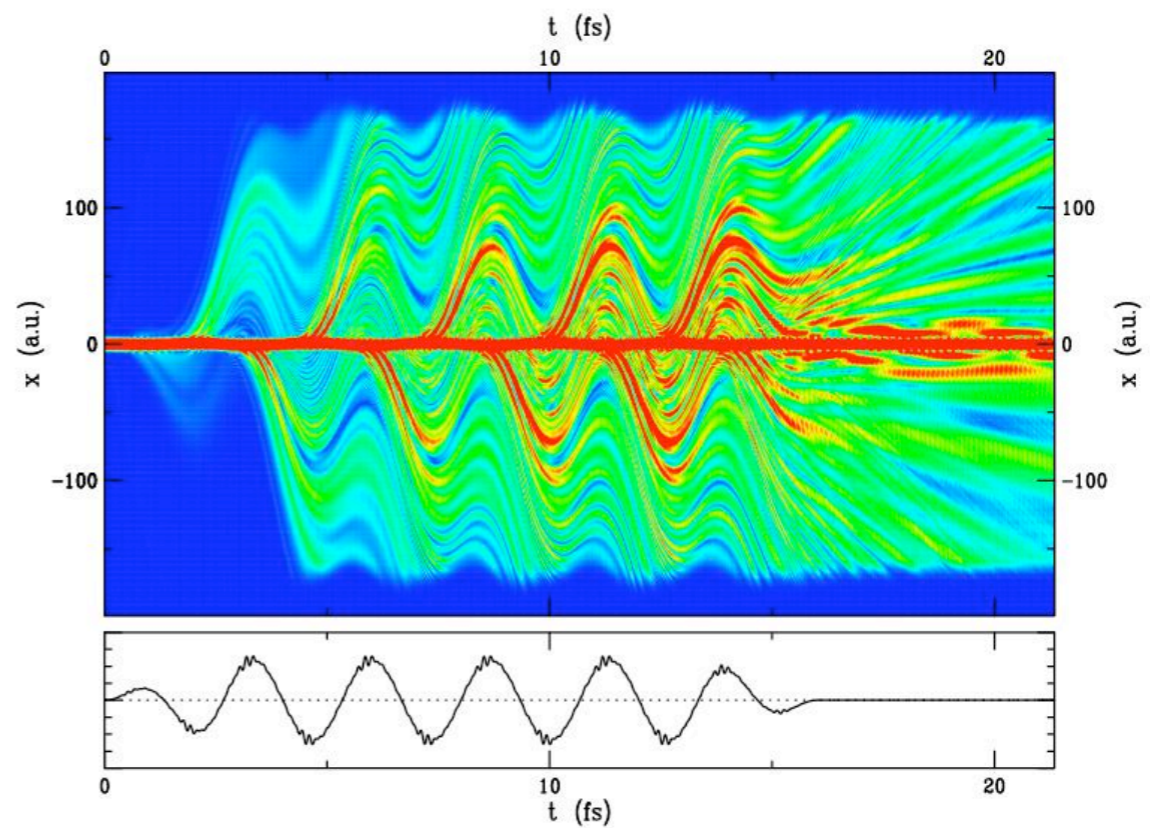
Yield



Single ionization



$$\varphi = 0.7\pi$$



$$\varphi = 1.7\pi$$

# Ionization drives HHG

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## Single atom response

- Electron WP must return to core:
  - Ellipticity dependence
  - Saturation

# Ionization drives HHG

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## Single atom response

- Electron WP must return to core:
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  - Saturation

## Propagation

- Free electrons reduce the refractive index.
    - Happens in space: fundamental defocused.
    - Happens in time: frequency modulation.
-

# Calculating the single atom and macroscopic response

One atom, strong field



Dipole radiation  
Ionization probability

# Calculating the single atom and macroscopic response

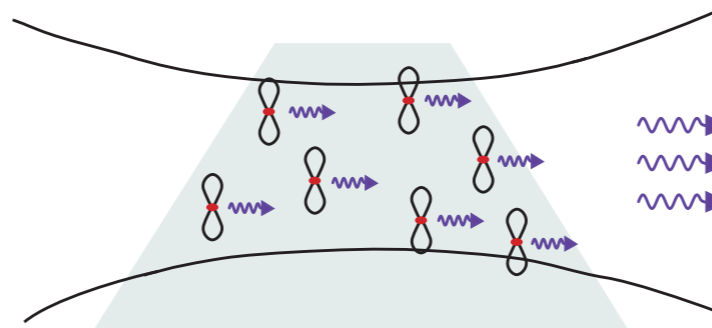
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Many atoms, focused laser beam



Propagation  
Phase matching

# Calculating the single atom and macroscopic response

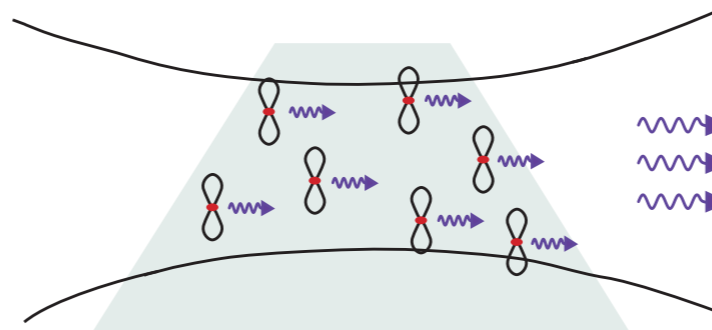
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# Calculating the single atom and macroscopic response

Non-adiabatic, coupled solutions of wave equation and time-dependent Schrödinger equation.

$$\nabla_{\perp}^2 E_1(\omega, \mathbf{r}) + \frac{2i\omega}{c} \frac{\partial E_1(\omega, \mathbf{r})}{\partial z} = G(\omega, \mathbf{r})$$

$$\nabla_{\perp}^2 E_h(\omega, \mathbf{r}) + \frac{2i\omega}{c} \frac{\partial E_h(\omega, \mathbf{r})}{\partial z} = -\omega^2 \mu_0 P_{nl}(\omega, \mathbf{r}) - \frac{i\omega}{c} \alpha(\omega, \mathbf{r}) E_h(\omega, \mathbf{r})$$

SEWA: Brabec and Krausz, Rev. Mod. Phys 2001

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SEWA: Brabec and Krausz, Rev. Mod. Phys 2001

## Laser field: $G(\omega, \mathbf{r})$ from ionization

- Free electrons change refractive index in space and time. Leads to defocusing and self-phase modulation (frequency shift).
- Loss of energy to ionization process
- Use ADK rates, scale ionization probability to fit numerical solution of TDSE

Gaarde et al, PRL submitted

## Harmonic fields

$$P_{nl}(t, \mathbf{r}) = N_{atom}(t, \mathbf{r}) d(t, \mathbf{r})$$

- Use SFA, non-adiabatic
- Absorption  $\alpha(\omega, \mathbf{r})$



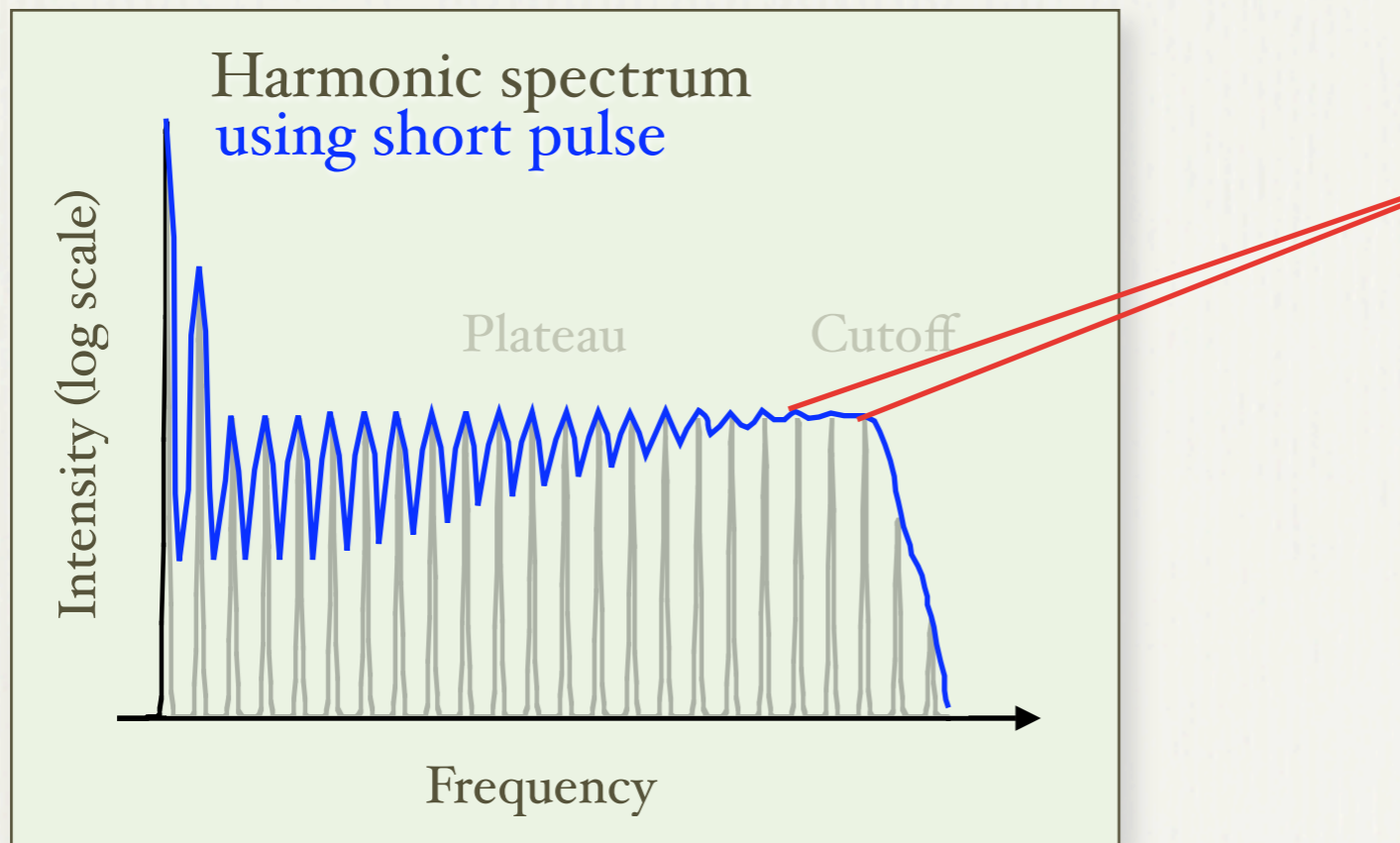
# Single attosecond pulse production

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SAP production is usually understood as a single atom effect.

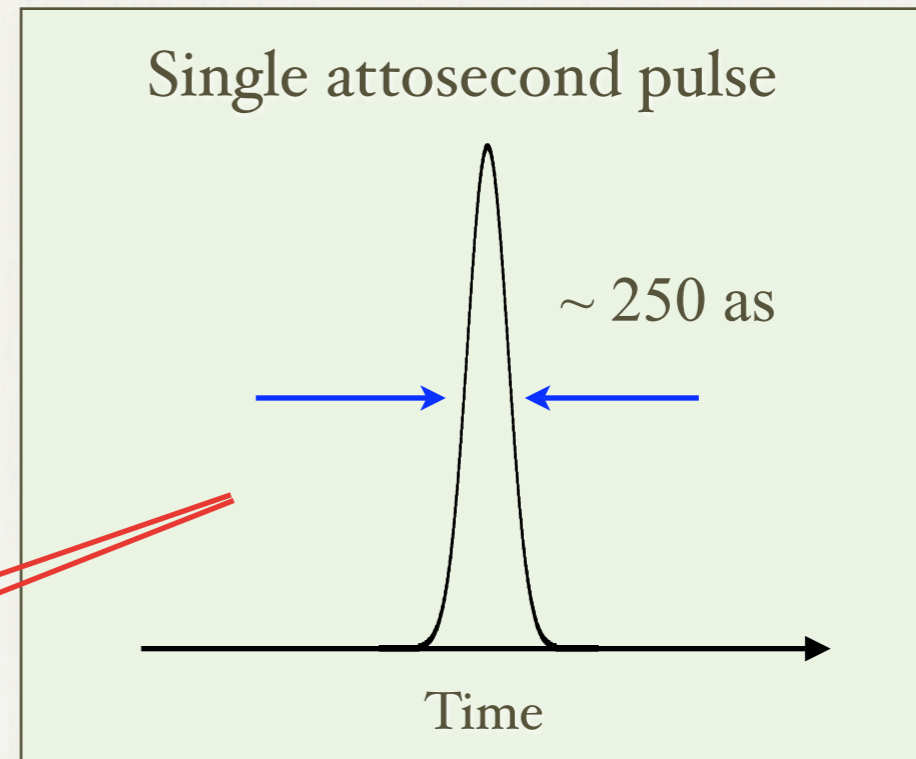
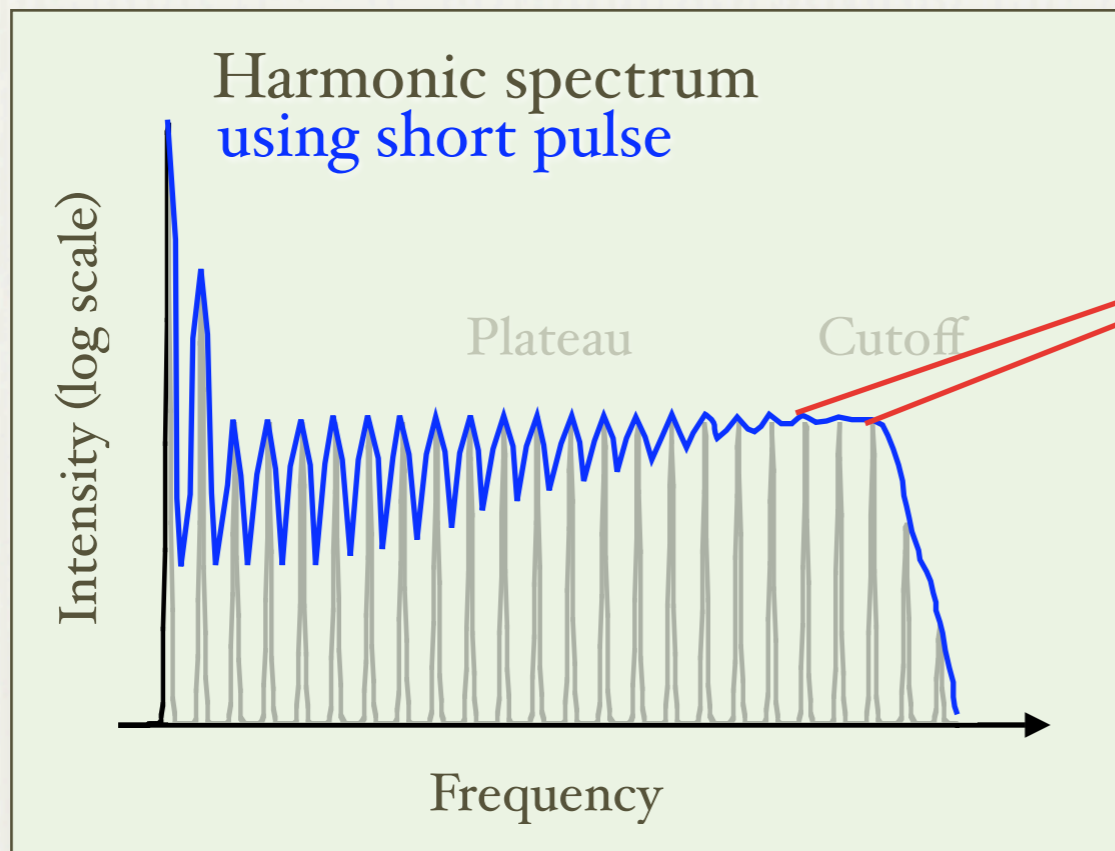
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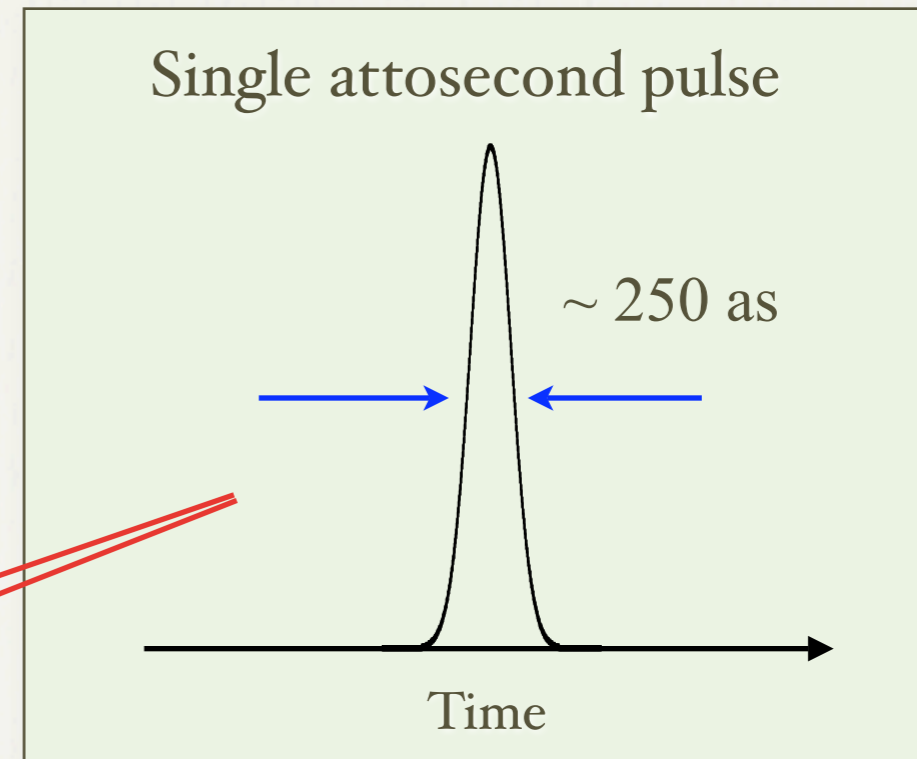
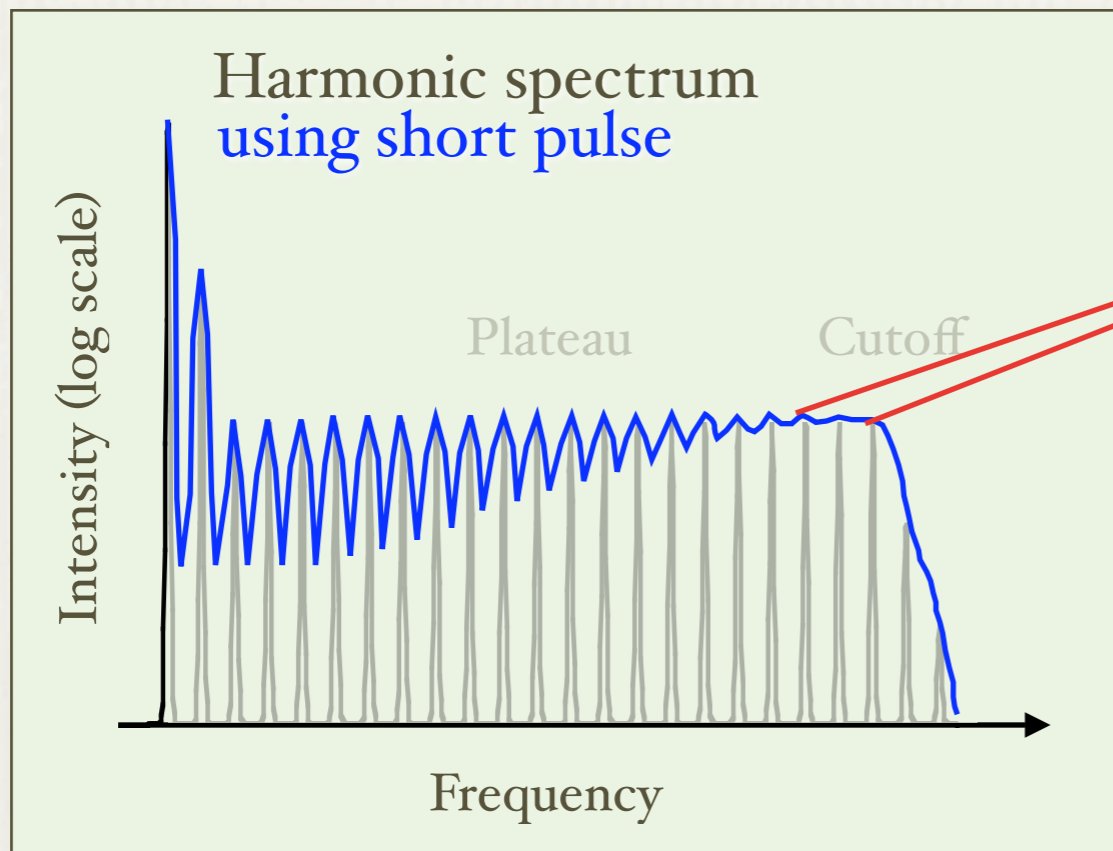
# Single attosecond pulse production

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# Single attosecond pulse production

SAP production is usually understood as a single atom effect.



- Two cycle (5-6 fs) driving pulse.
- Spectral selection.
- Carrier envelope phase control.

# Few cycle C-E control of HHG

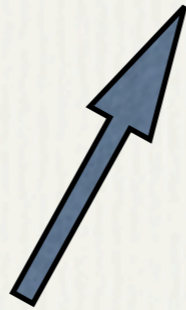
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$$E(t) = f(t) \cos(\omega_0 t + \phi)$$

# Few cycle C-E control of HHG

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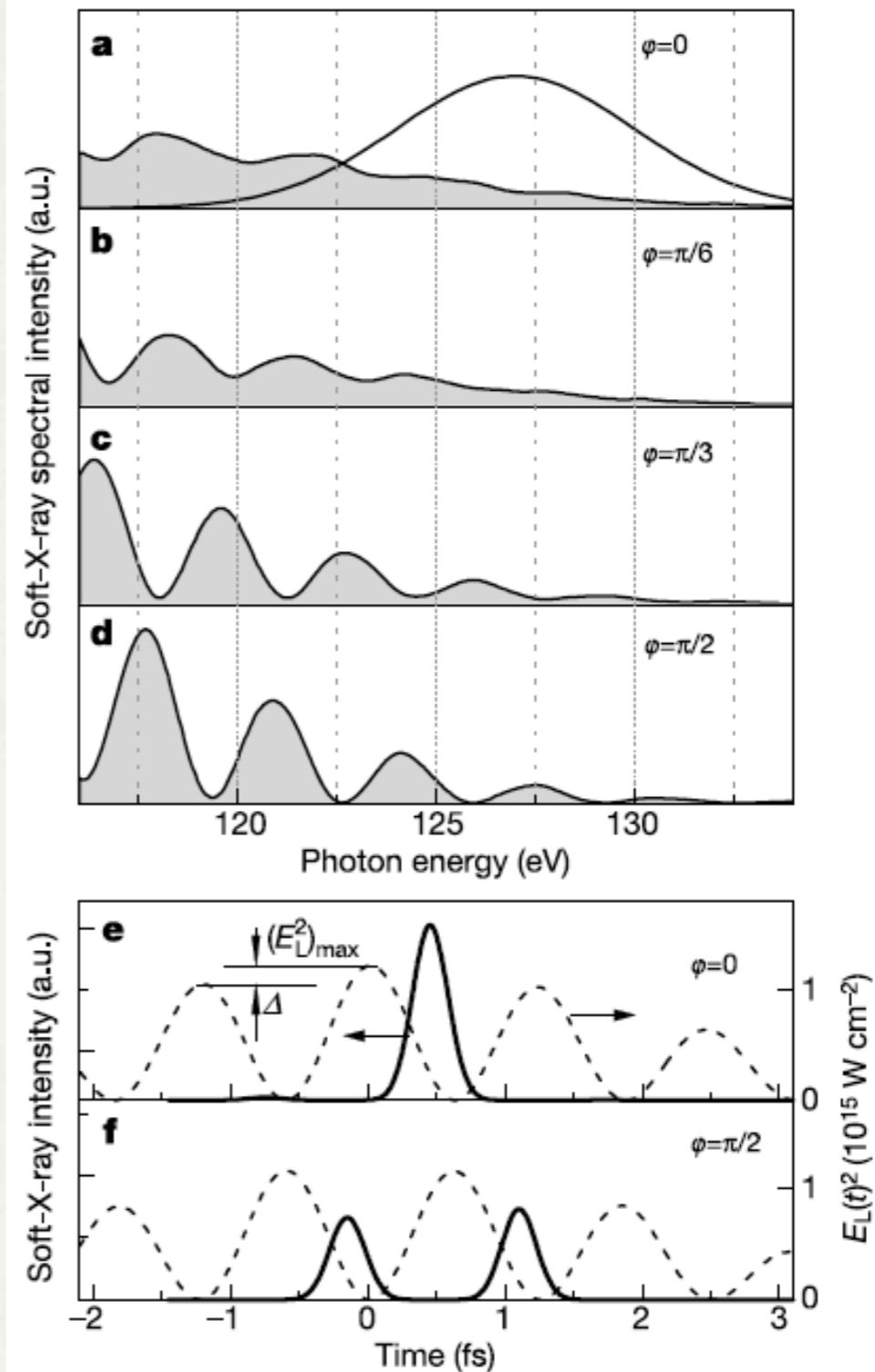


Carrier envelope phase

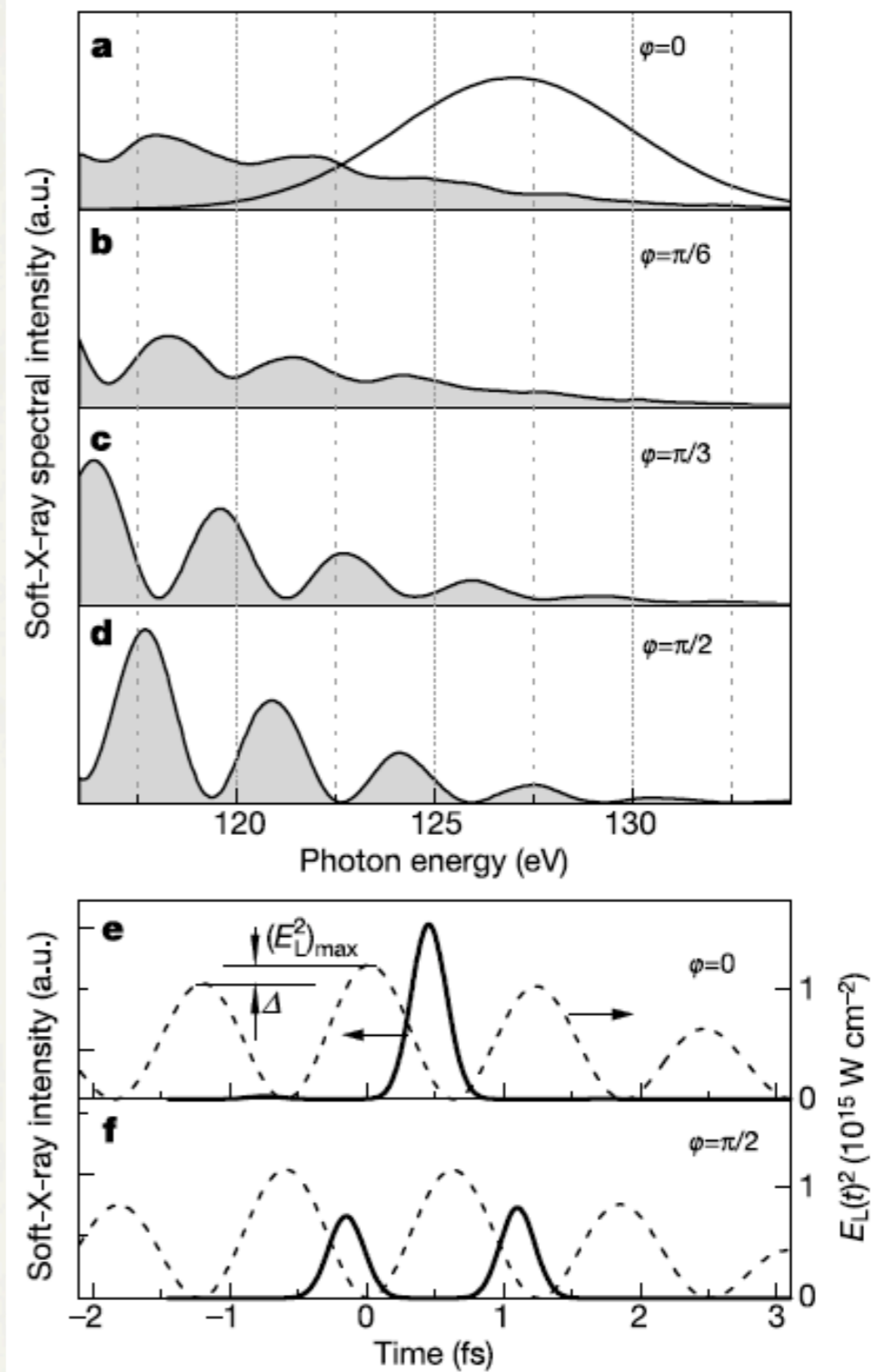
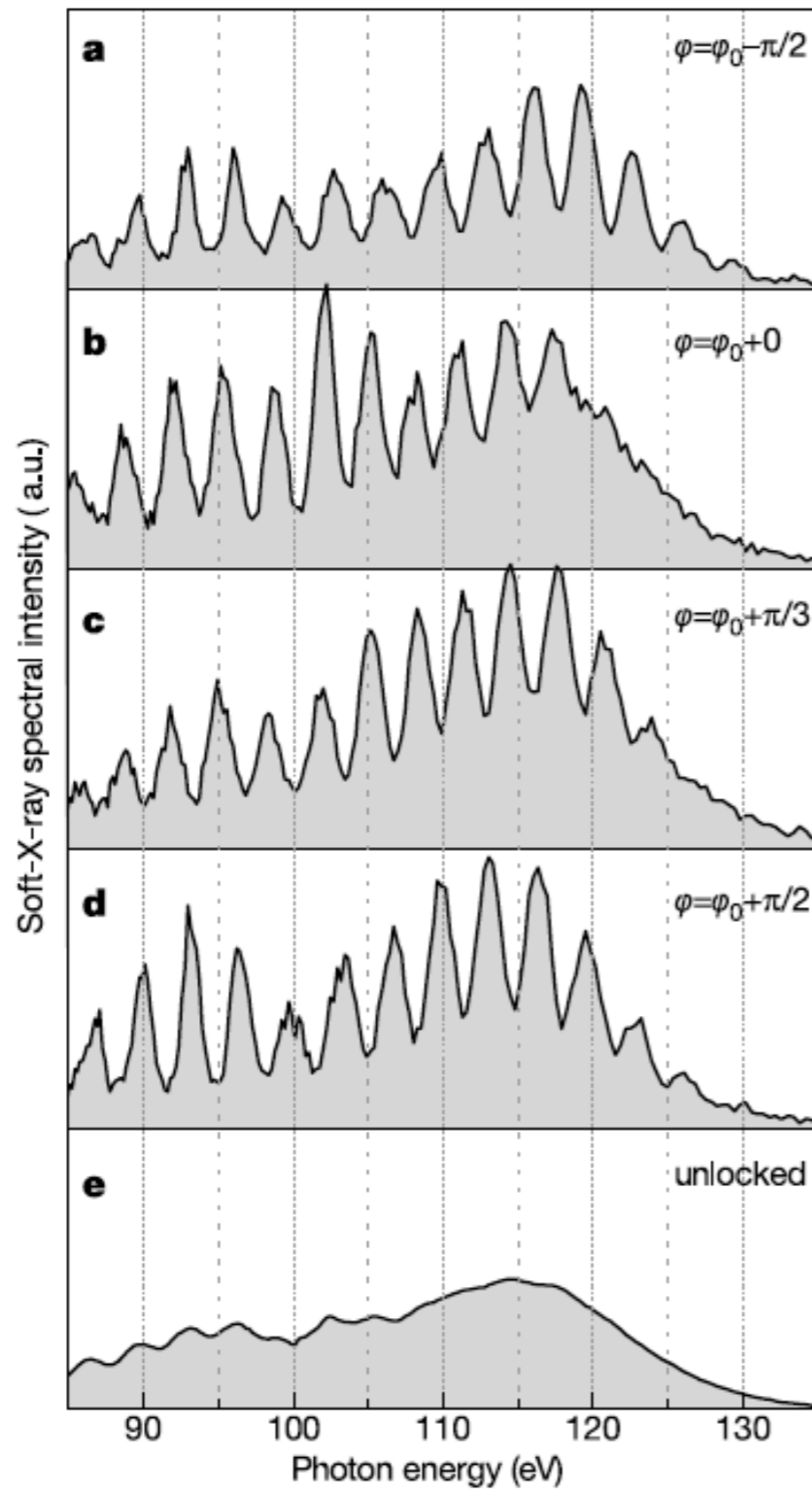
# Few cycle C-E control of HHG

$$E(t) = f(t) \cos(\omega_0 t + \phi)$$

Carrier envelope phase

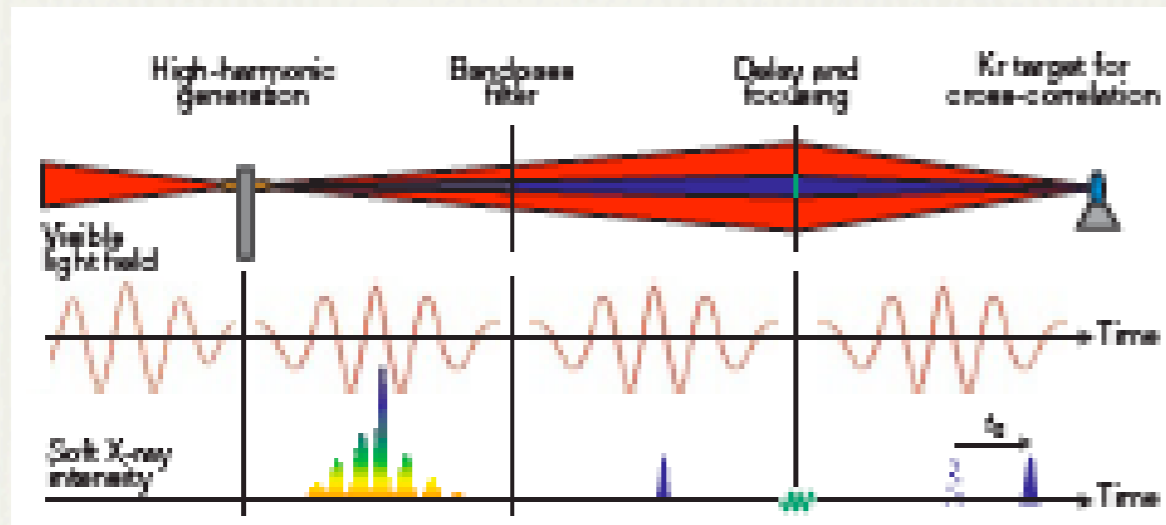


# Few cycle C-E control of HHG





# Attosecond pulse selection

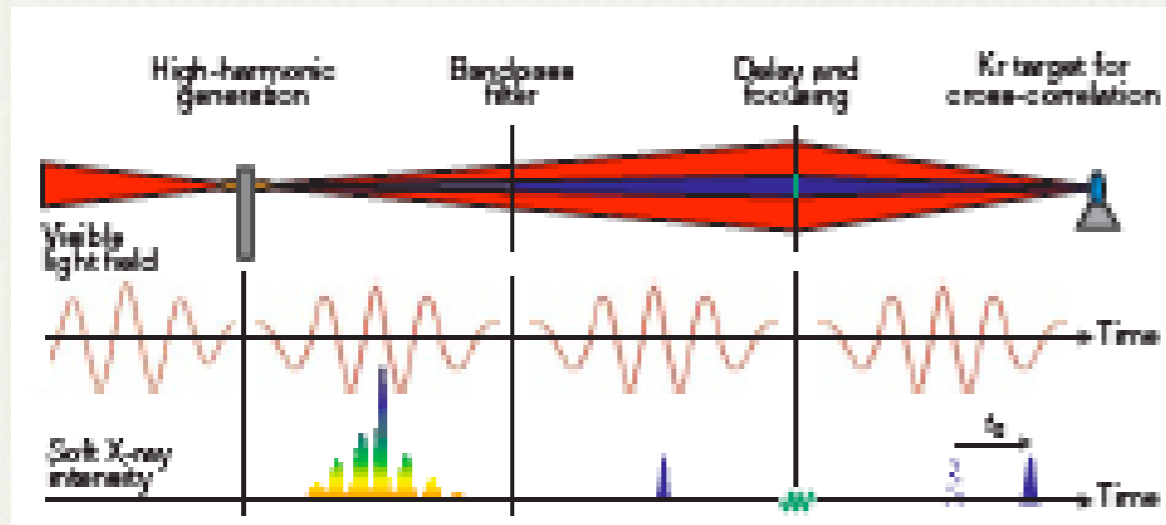


● The pulse is too long (3 cycles).

● The C-E phase was not controlled.

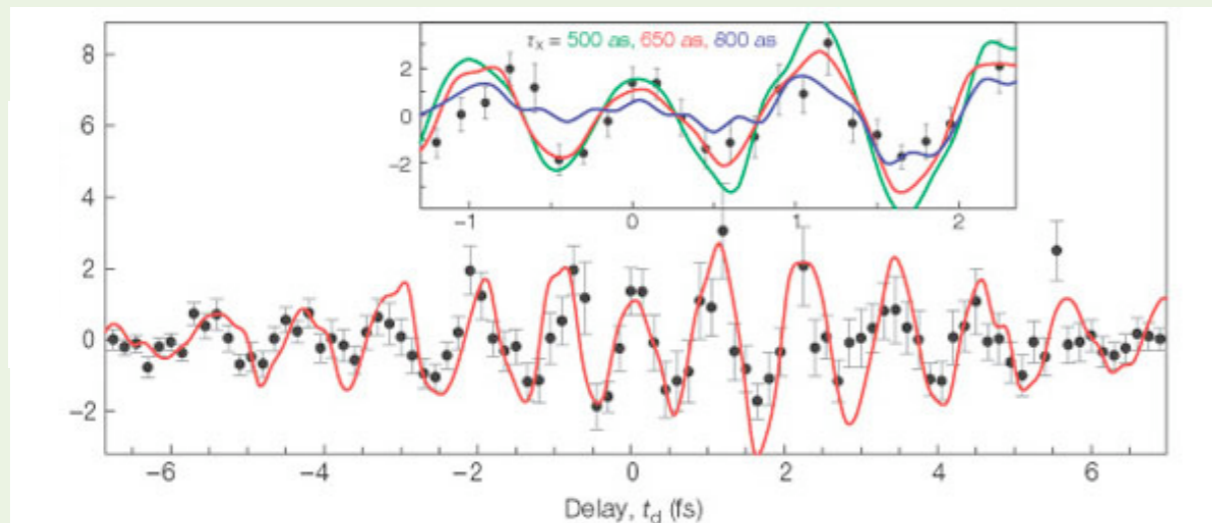
Hentschel, *et al*, Nature 2001.

# Attosecond pulse selection



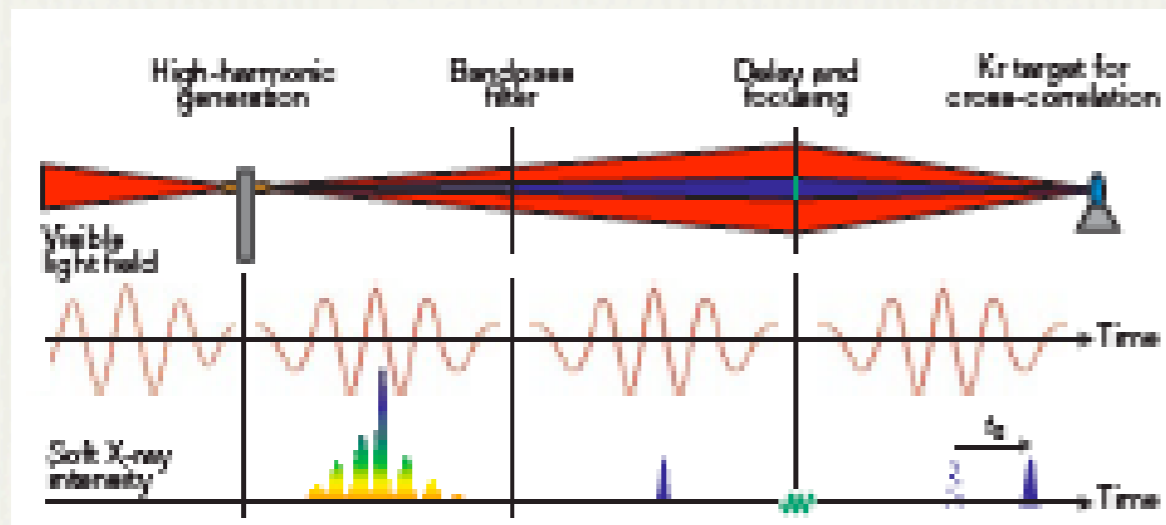
- The pulse is too long (3 cycles).
- The C-E phase was not controlled.

Hentschel, *et al*, Nature 2001.



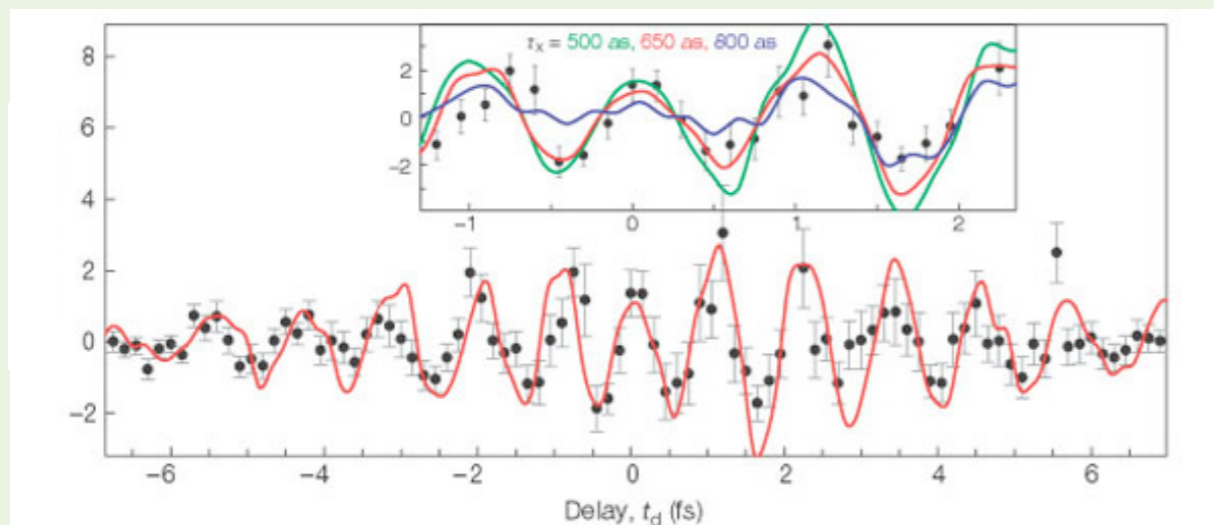
- 750 nm, 7 fs pulses,  $I = 9 \times 10^{14}$  W/cm<sup>2</sup>  
→ Isolated 630 as pulses at 90 eV
- Measured absolute phase of laser field:  
Large dynamical blue shift, up to 35%  
Result of rapid ionization dynamics?

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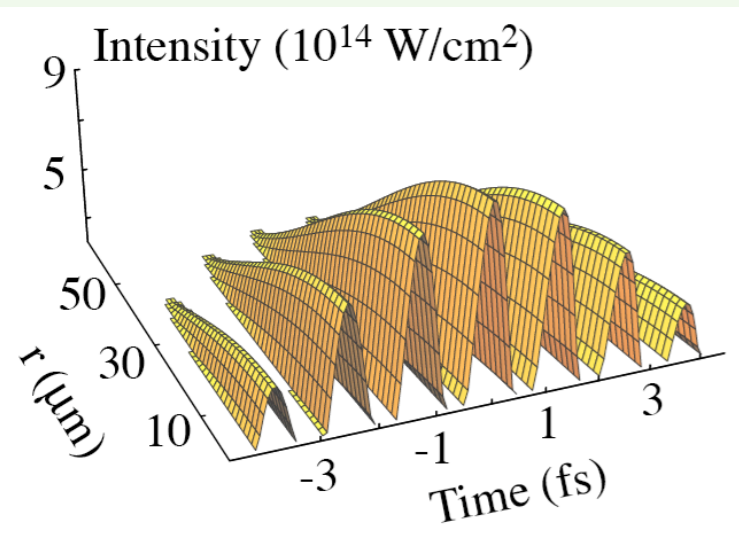


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## Selection of single attosecond pulse in farfield

# Ionization-driven spatiotemporal reshaping

Initial laser pulse

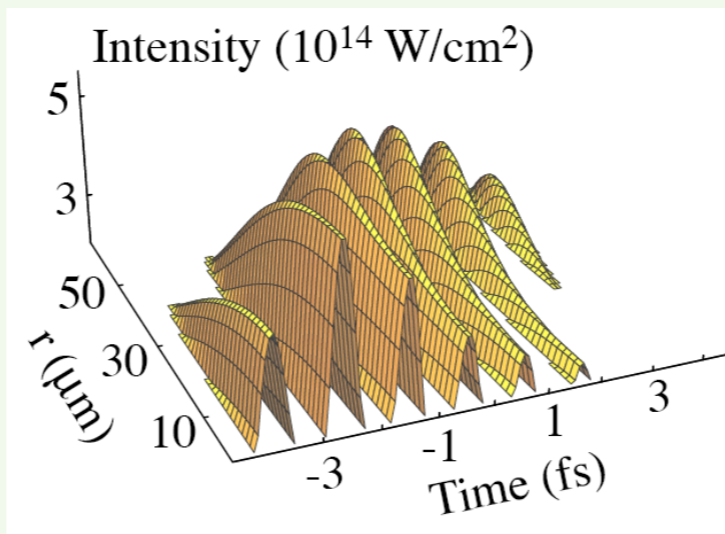


neon  
3 mm



180 mbar

After propagation



Calculation

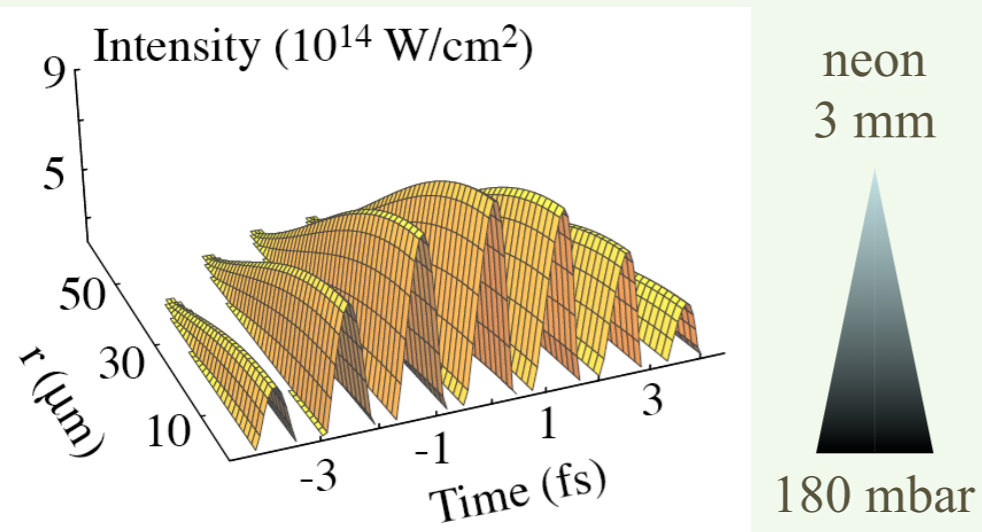
using parameters from 2001 exp.

- (Much) lower peak intensity
- Strongly divergent wave front  
Intensity peaks at different times for different radial pos.

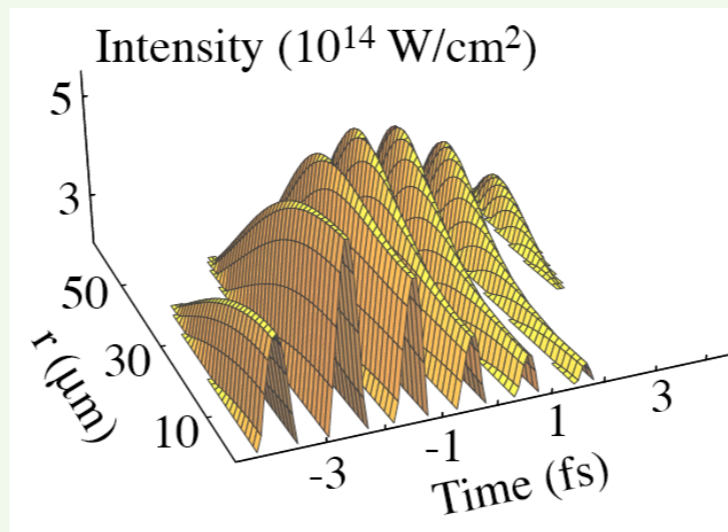
Gaarde et al, PRL submitted

# Ionization-driven spatiotemporal reshaping

### Initial laser pulse



### After propagation



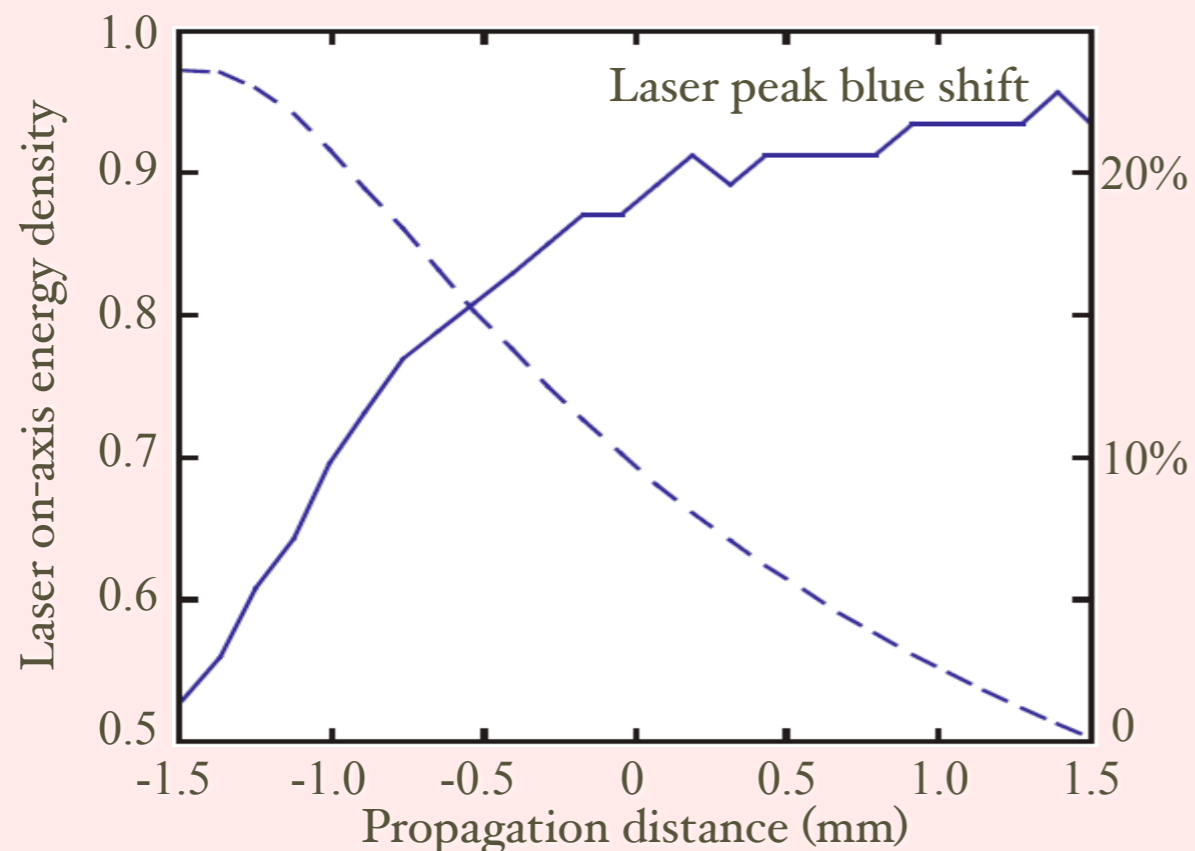
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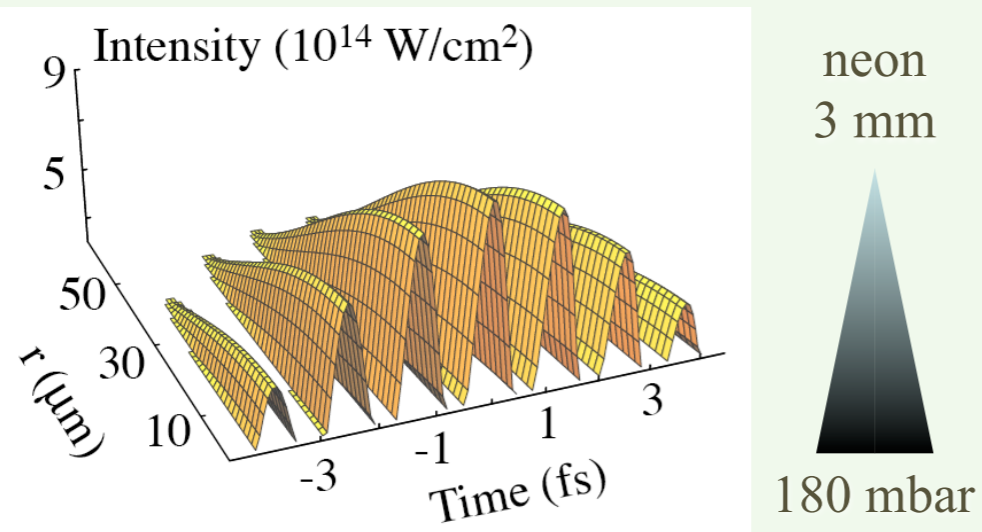
### Defocusing and frequency modulation



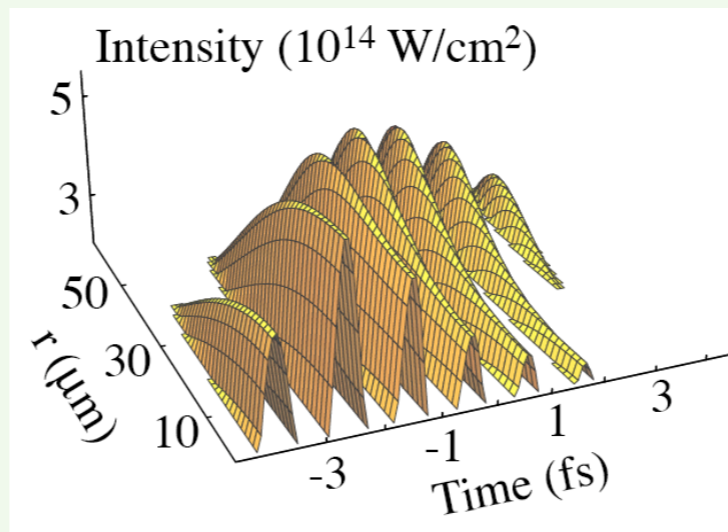
- Blue shift builds up rapidly in first half of gas while intensity is still high
  - Defocusing reduces on-axis intensity  
Consistent with 90 eV cutoff energy
- Slightly different laser focusing reproduces 35% frequency shift and 90 eV cutoff

# Ionization-driven spatiotemporal reshaping

Initial laser pulse



After propagation



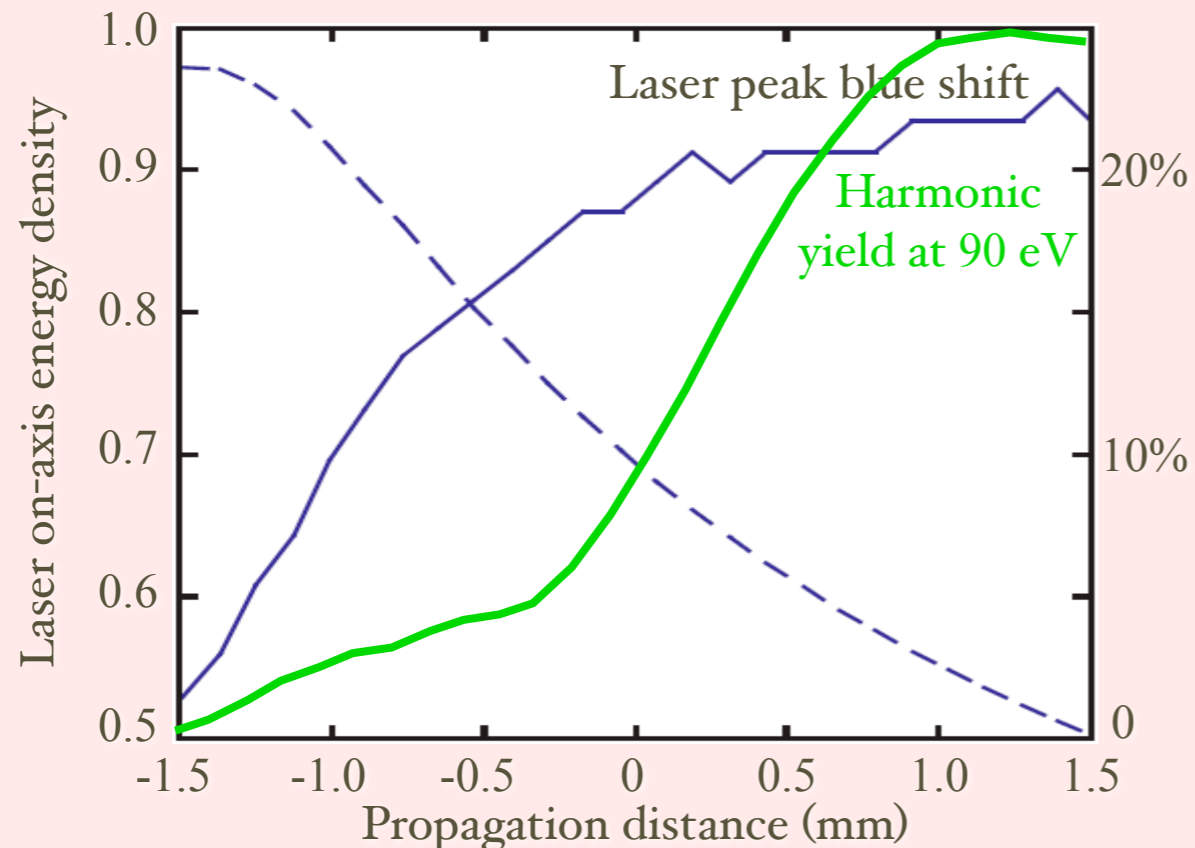
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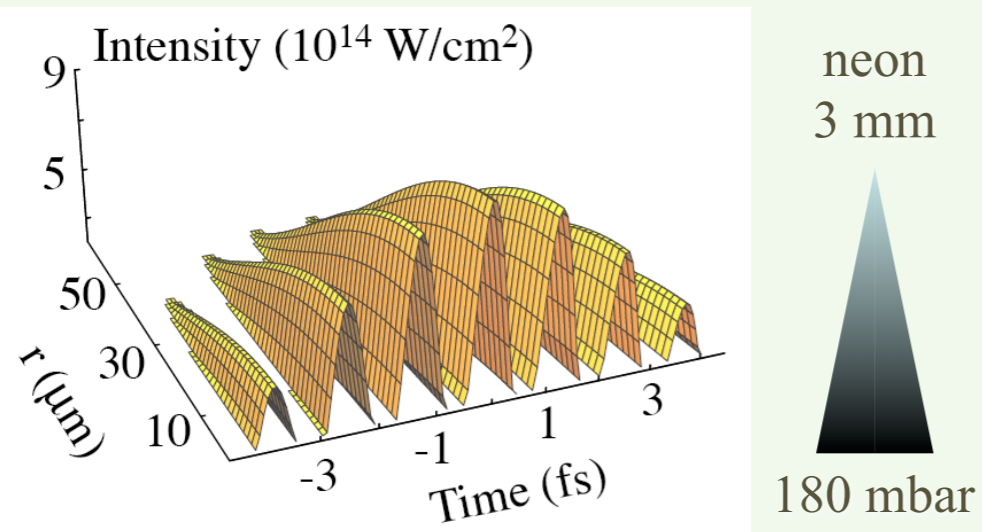
Defocusing and frequency modulation



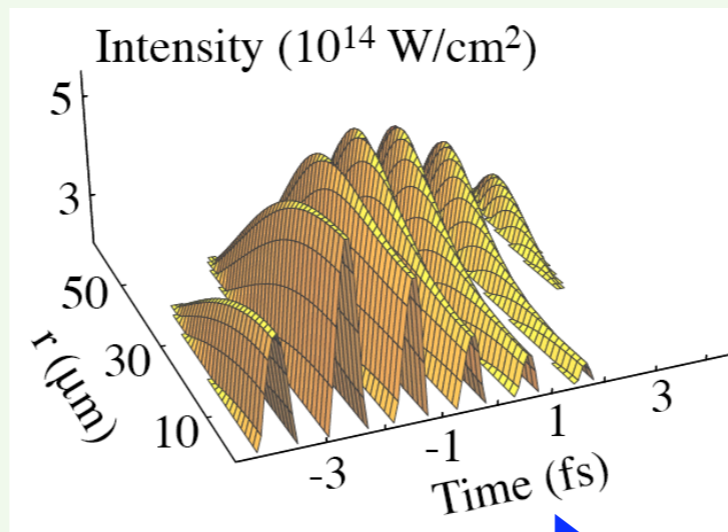
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Slightly different laser focusing reproduces 35% frequency shift and 90 eV cutoff
- Harmonics around 90 eV are generated in second half of medium

# Ionization-driven spatiotemporal reshaping

Initial laser pulse



After propagation



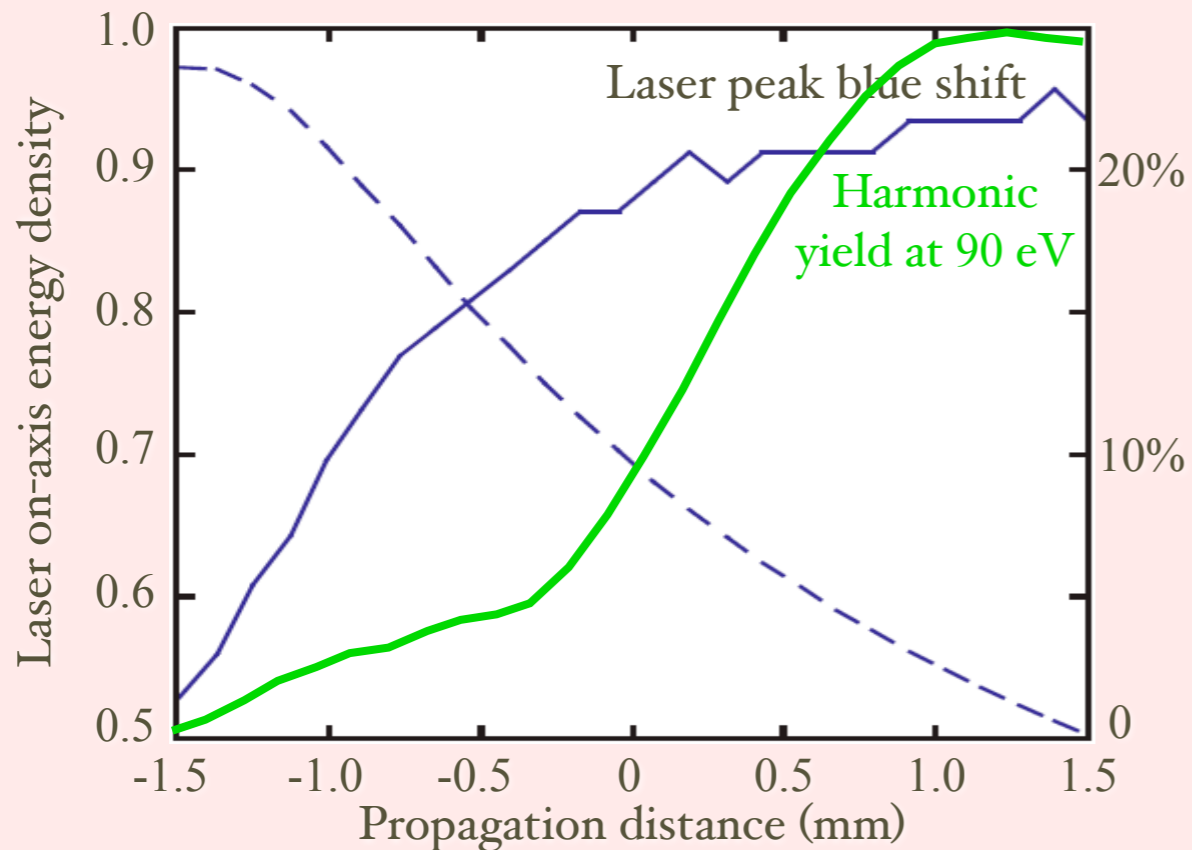
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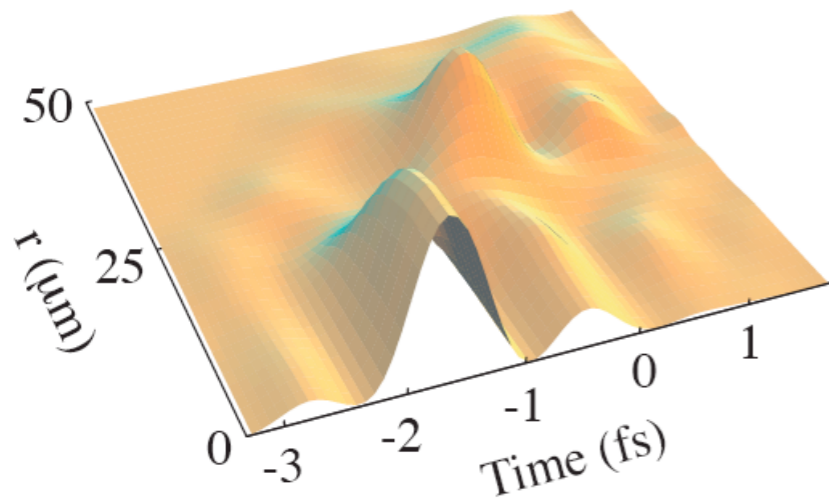


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# Harmonic generation and attosecond XUV pulses

Gaarde and Schafer, in preparation

Spatiotemporal intensity profile of 90 eV radiation



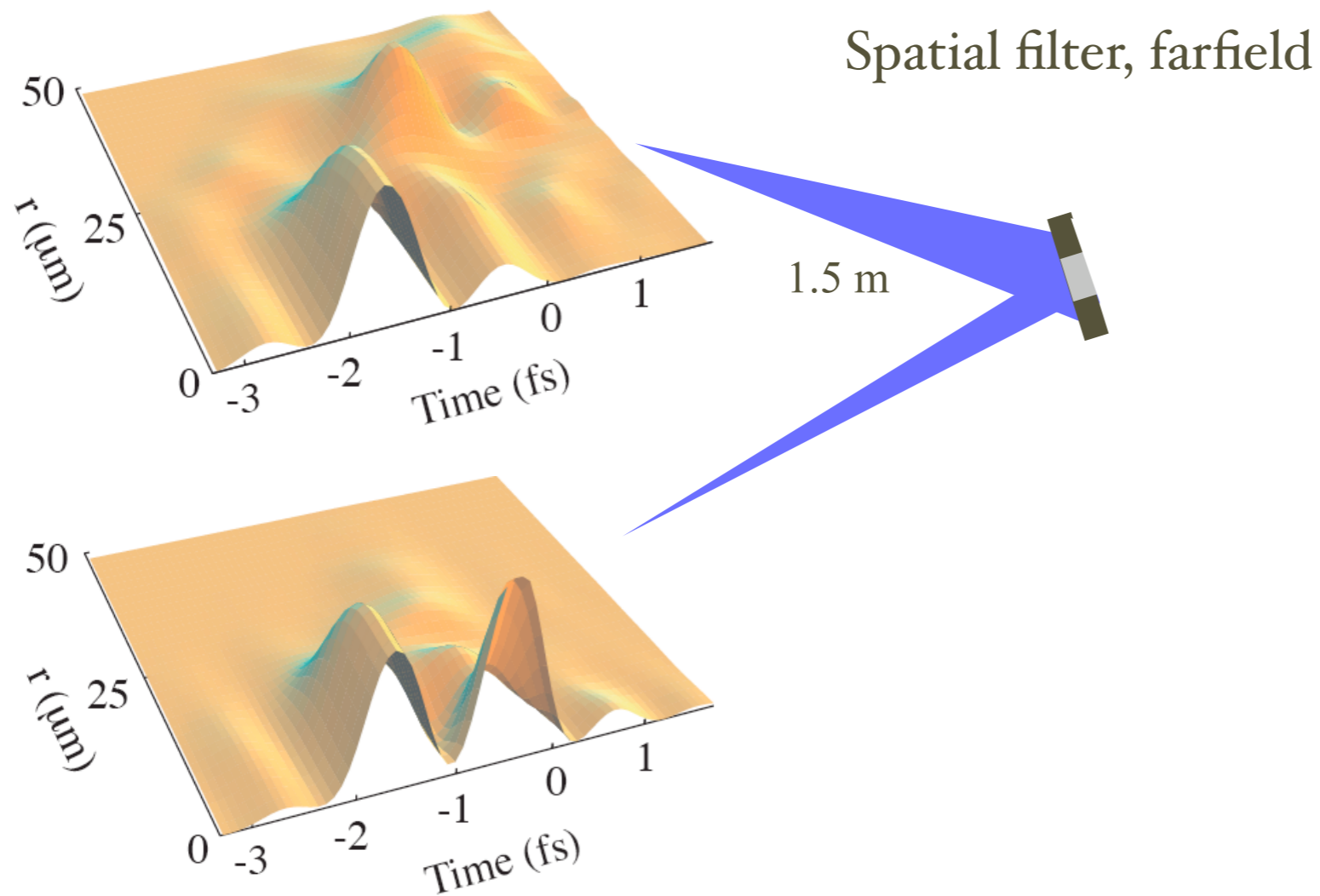
- Multiple pulses in nearfield
- Reshaped driving laser pulse means they are generated with different divergences



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Spatiotemporal intensity profile of 90 eV radiation

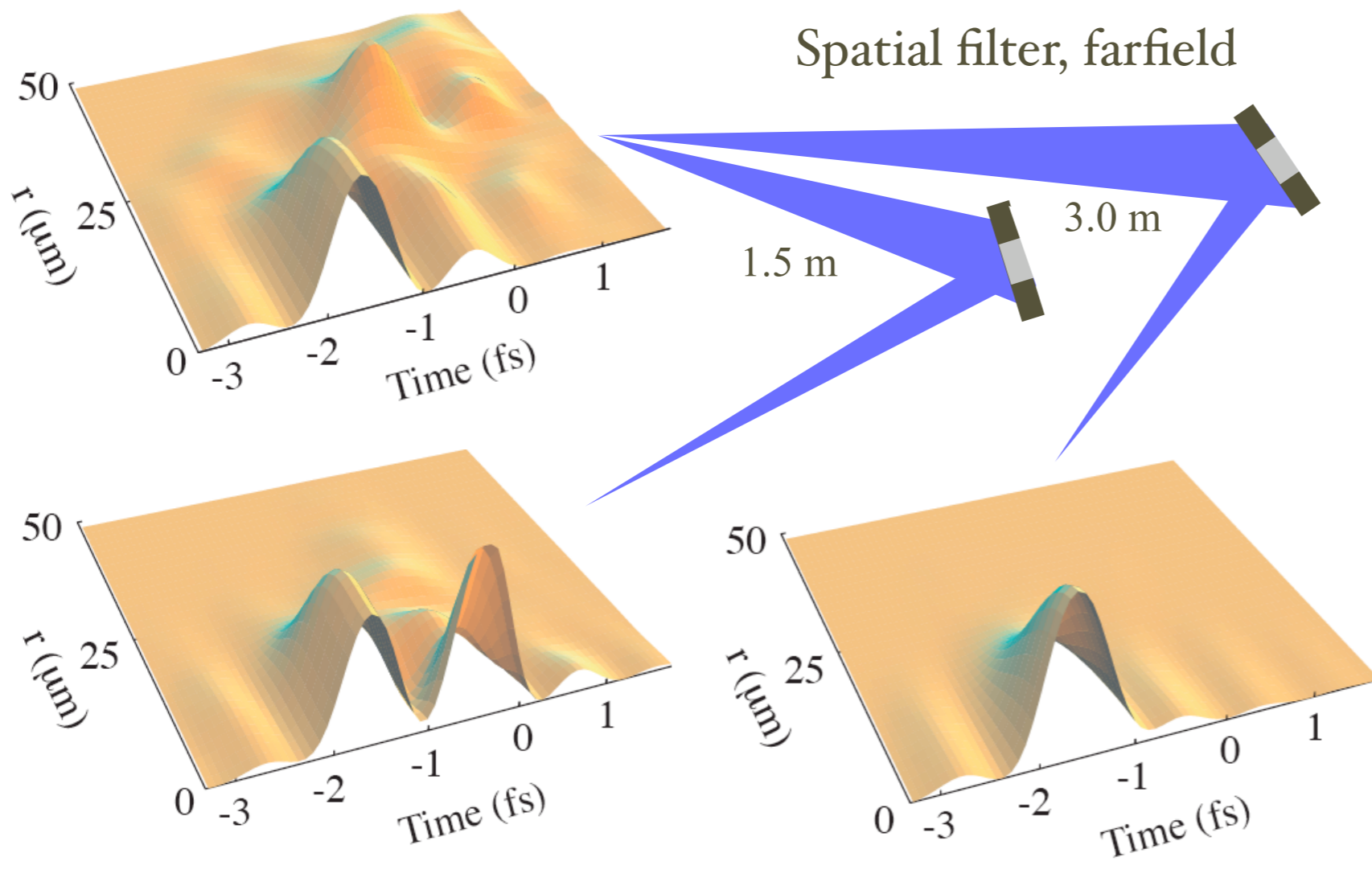


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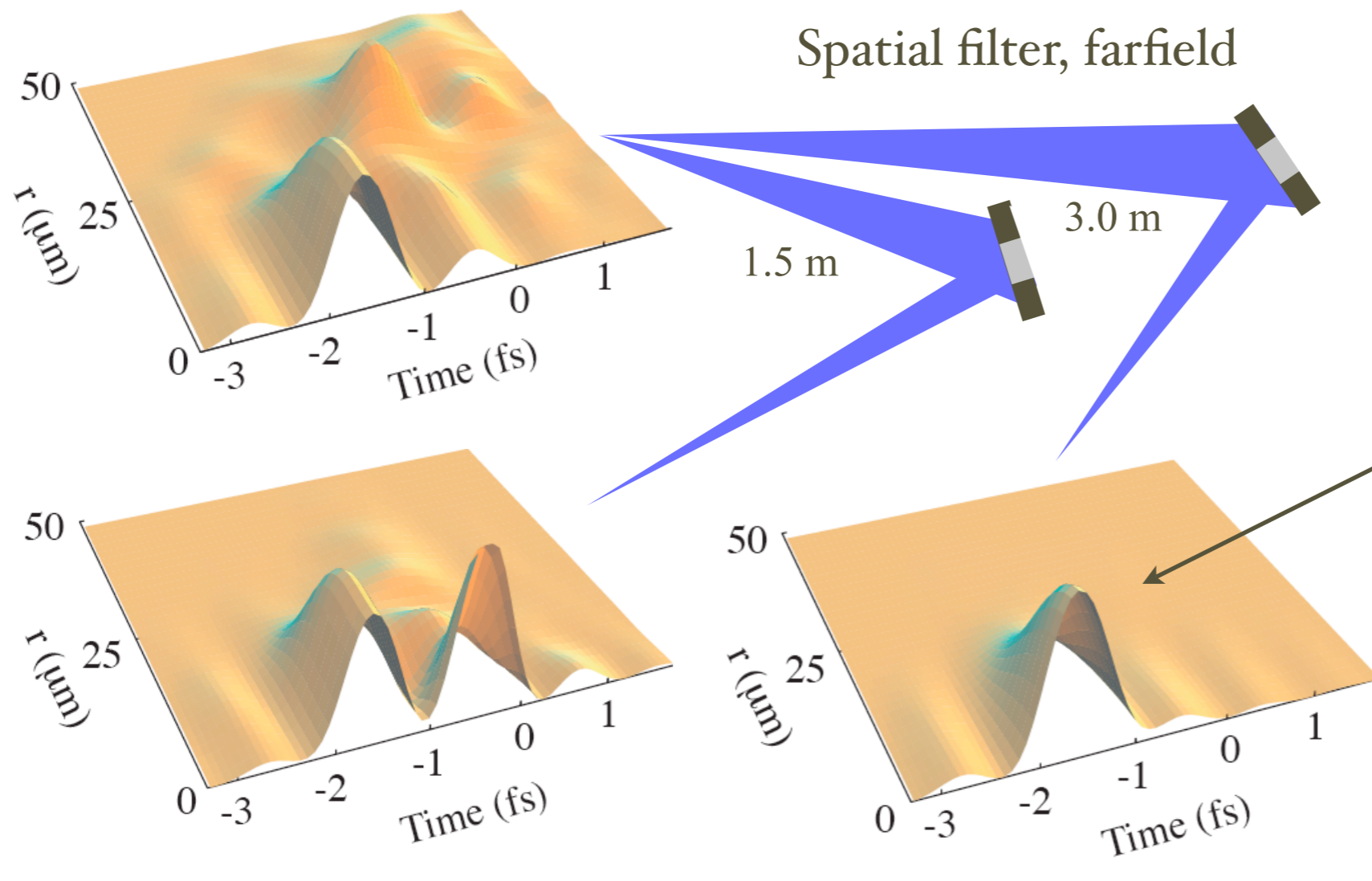


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Gaarde and Schafer, in preparation

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- Multiple pulses in nearfield
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630 as, 3 pJ

Spatial filter in farfield works as time gate  
Isolate a single attosecond pulse among several

Spatial filter is XUV mirror with a 2 mm diameter, as in Hentschel *et al*, Nature 2001 (placed 2.5 m from source)

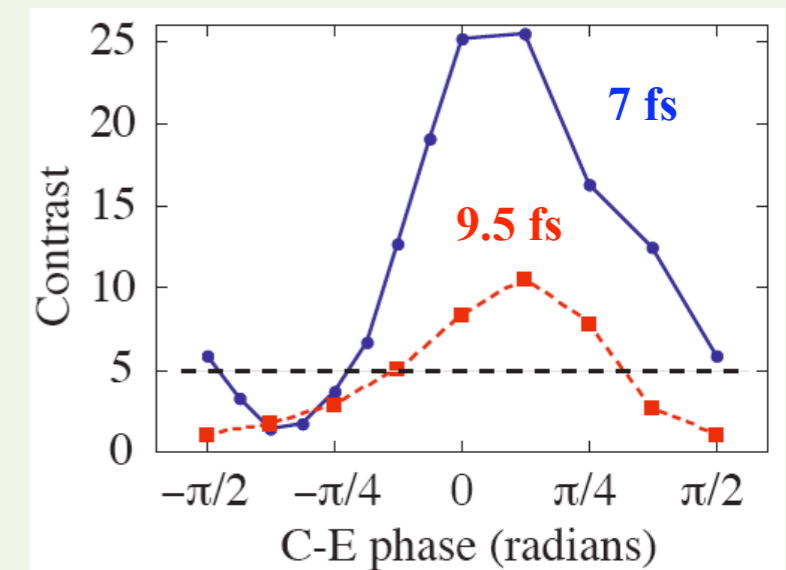
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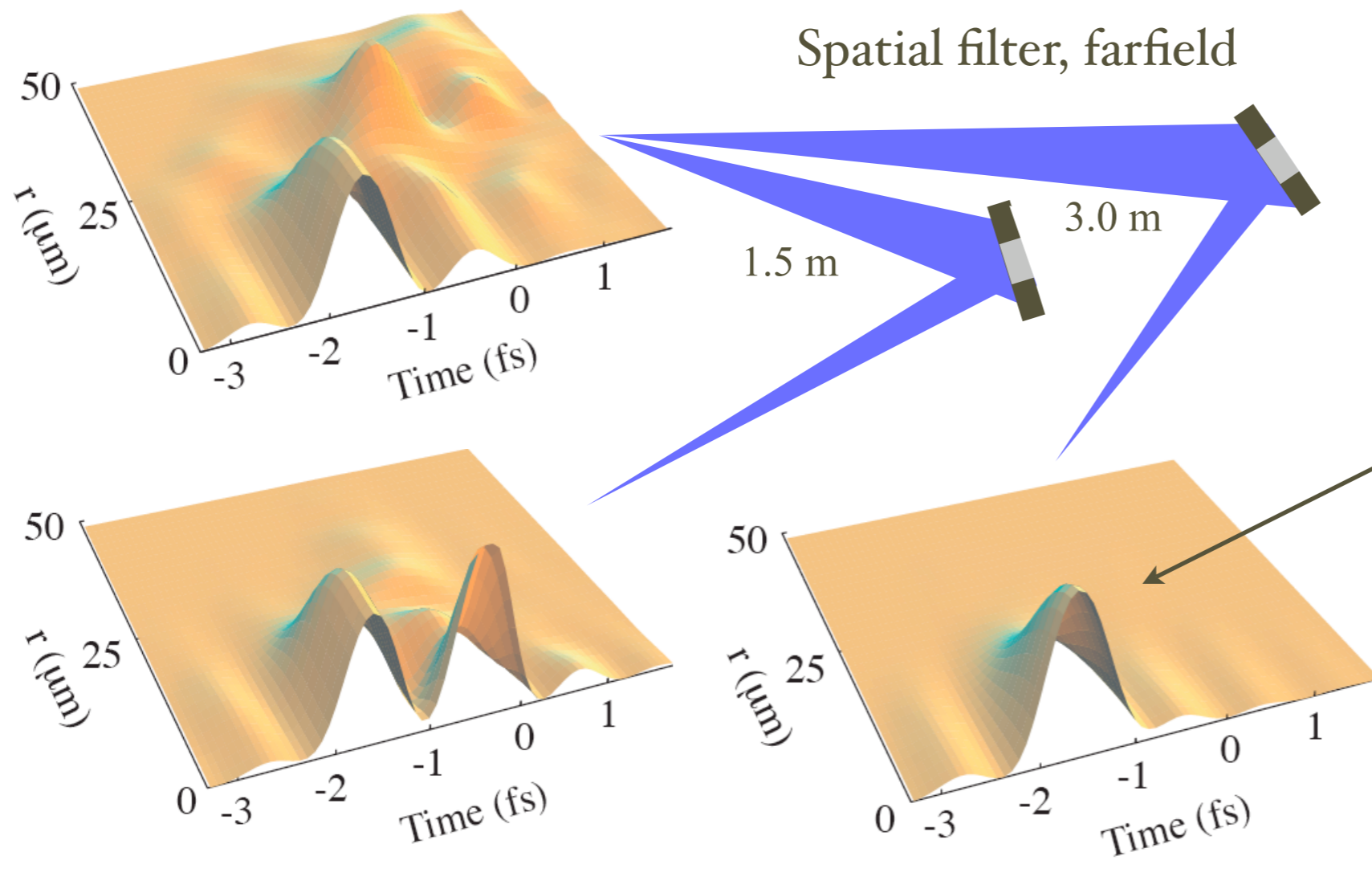
630 as, 3 pJ

Works for even longer pulses:



- 9.5 fs driving pulse gives contrast ratio of 10:1
- 11 fs driving pulse gives contrast ratio of 6:1

Spatiotemporal intensity profile of 90 eV radiation



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Isolate a single attosecond pulse among several

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# Strong field physics in attoscience

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- Theory challenges

- What we know we don't know:

- Beyond single active electron model,  
multiple electrons in full dimensionality.

- What we don't know:

- Where are multi electron effects important?

- We need better TDSE/MWE solvers.

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# Strong field physics in attoscience

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● Take home message:

Attosecond pulses are an exciting new tool to increase the precision with which we can study and control strong field processes. This, in turn, can lead to new attosecond sources and metrologies.