Reshaping of ultrafast light pulses through interaction with atomic structure

Mette Gaarde, Louisiana State University

KITP lecture, September 2014

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Funding
NSF, DOE BES, LONI (computing time)

Outline

Theme: what happens to the XUV light as a result of it's interaction with the atomic system (here: structure)

- Example, HHG: Cooper minimum in argon reshapes asec pulses

  Collaboration Ohio State group

- Example, transient absorption: Macroscopic propagation can also change absorption line shape in presence of IR

  Collaboration Arizona group

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Theme: what happens to the XUV light as a result of it’s interaction with the atomic system (here: structure)

- Introduce theoretical methods, coupled TDSE-MWE
- Example, HHG: Cooper minimum in argon reshapes asec pulses

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- Example, transient absorption: Macroscopic propagation can also change absorption line shape in presence of IR

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Methods

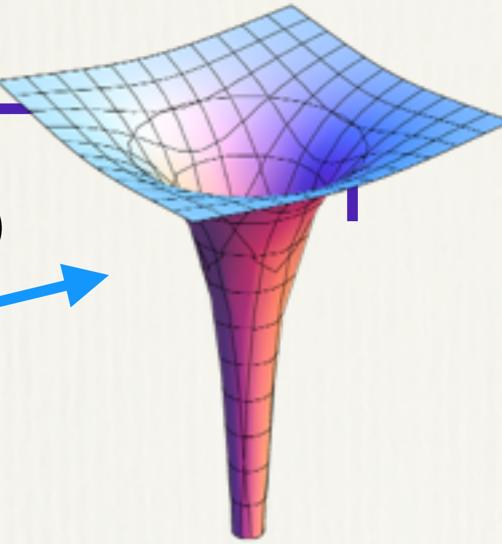
Schrödinger equation, in SAE:

$$i\frac{\partial}{\partial t}\psi(\vec{r},t) = [-\frac{1}{2}\nabla^2 + \mathcal{E}(t)z + V(\vec{r})]\psi(\vec{r},t)$$

Maxwell wave equation:

$$\nabla_\perp^2\tilde{\mathcal{E}}(\omega) + \frac{2i\omega}{c}\frac{\partial\tilde{\mathcal{E}}(\omega)}{\partial z} = -\frac{\omega^2}{\epsilon_0 c^2}\tilde{P}(\omega)$$

Methods



Pseudo potential V(r,l)

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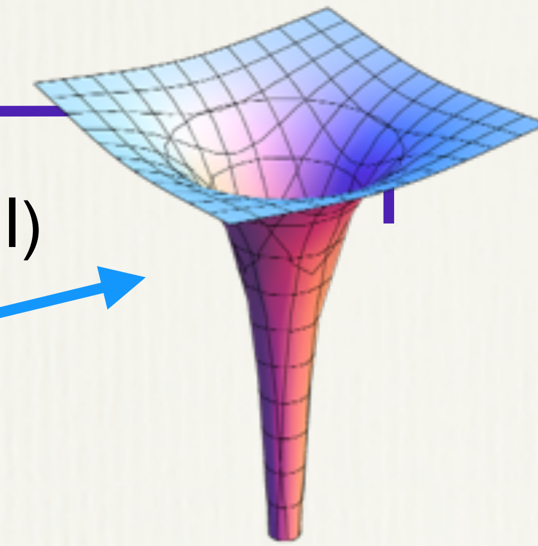

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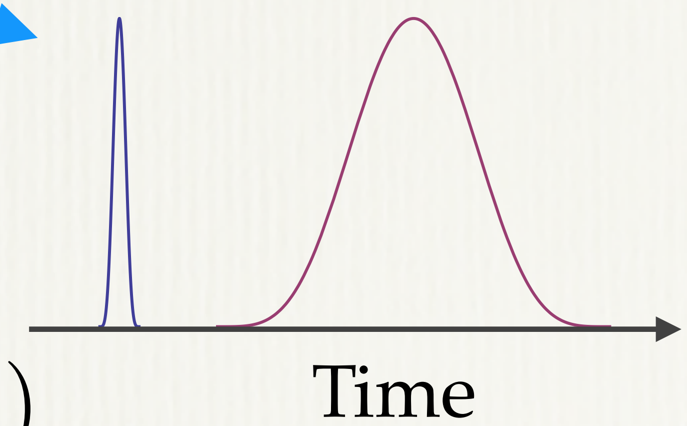

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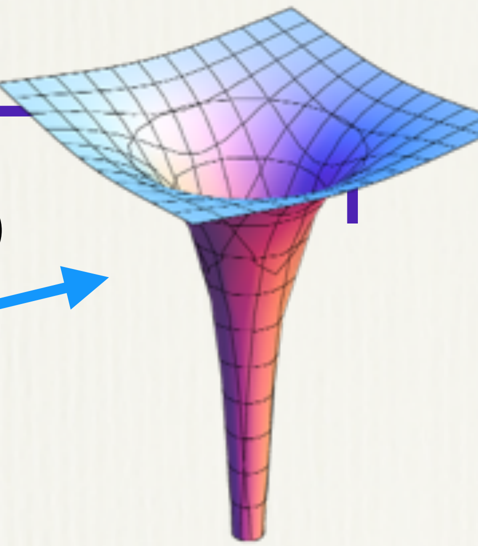

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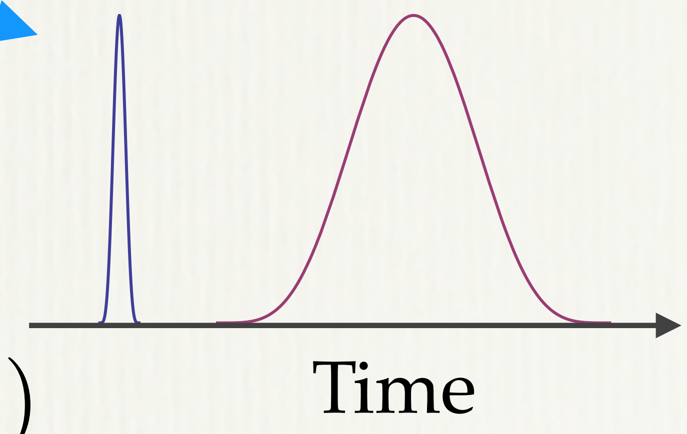

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$$\tilde{P}(\omega) = \rho\tilde{d}_{SAE}(\omega)$$

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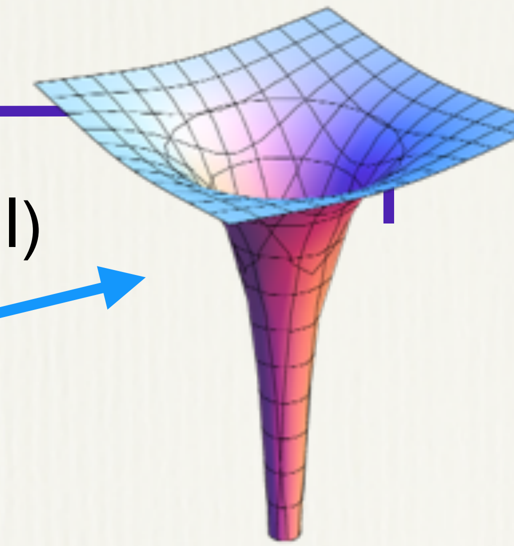

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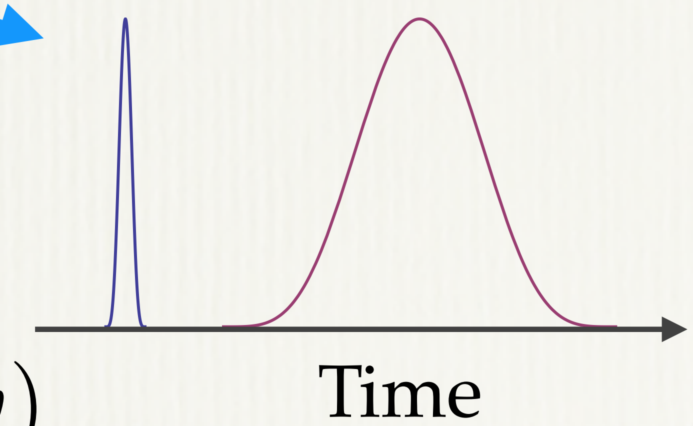

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FT of time-dependent dipole

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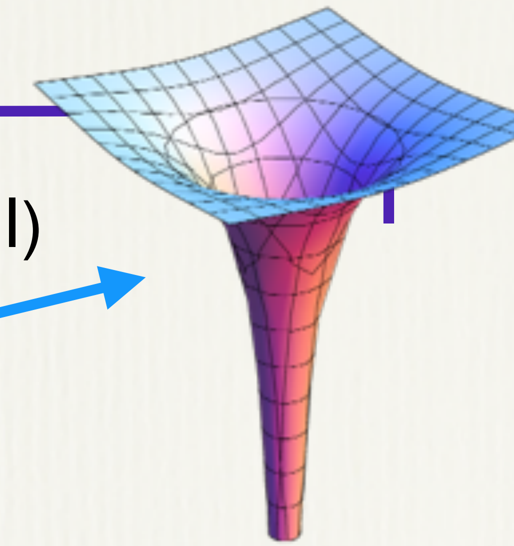

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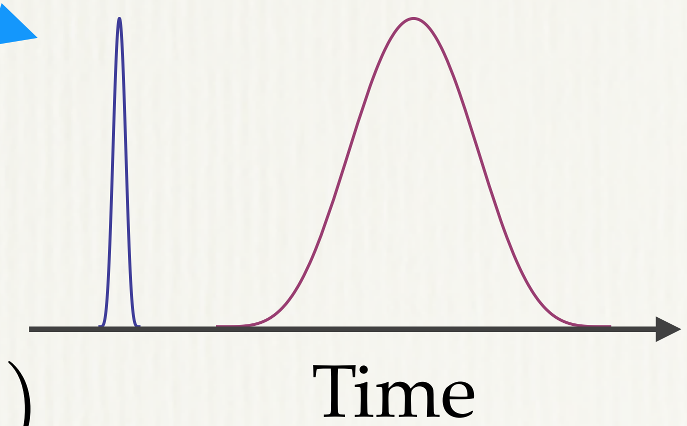

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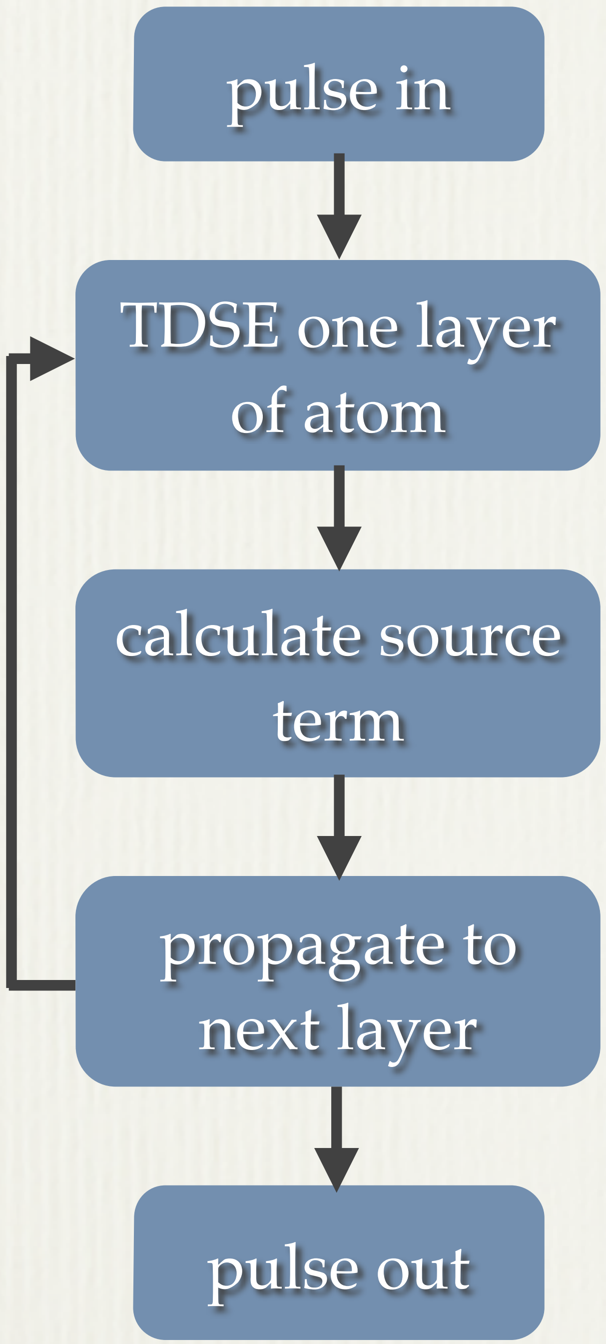
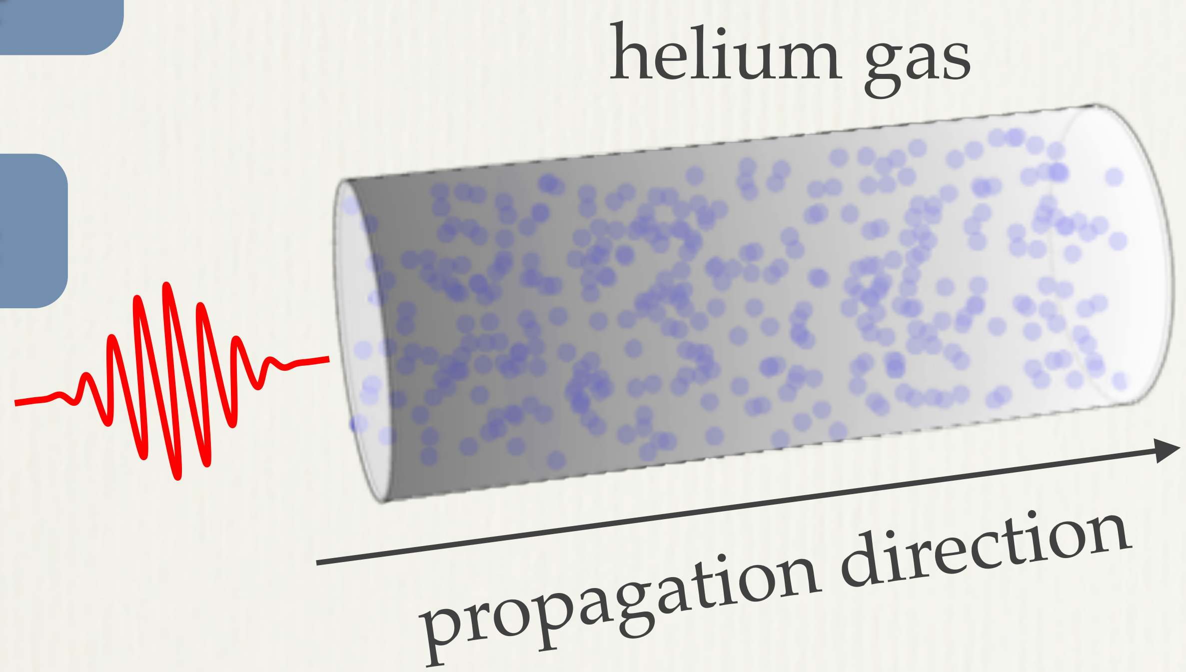
$$d(t) = -\langle\psi(\vec{r},t)|z|\psi(\vec{r},t)\rangle$$

FT of time-dependent dipole

Numerical solution of MWE-TDSE





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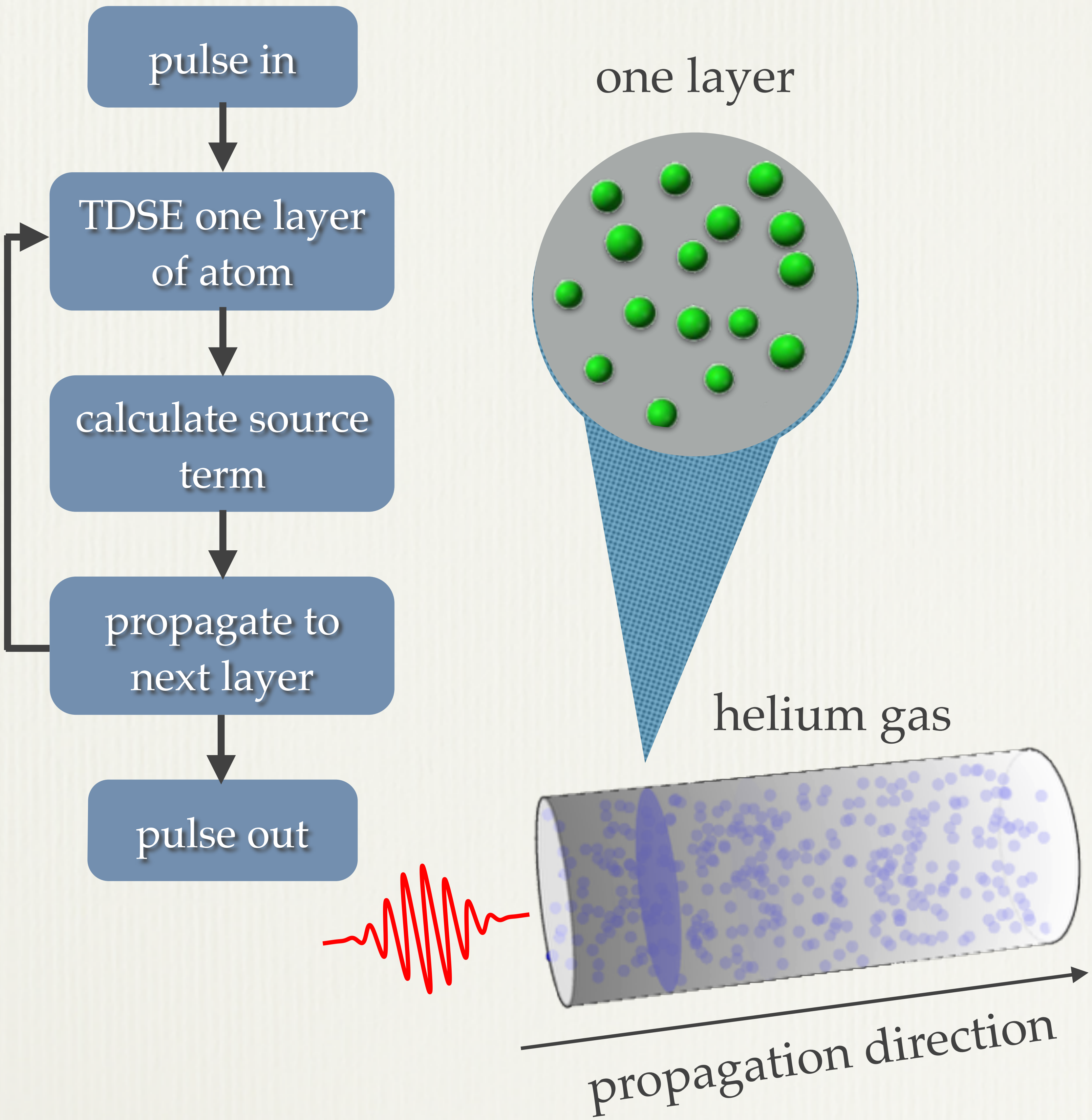

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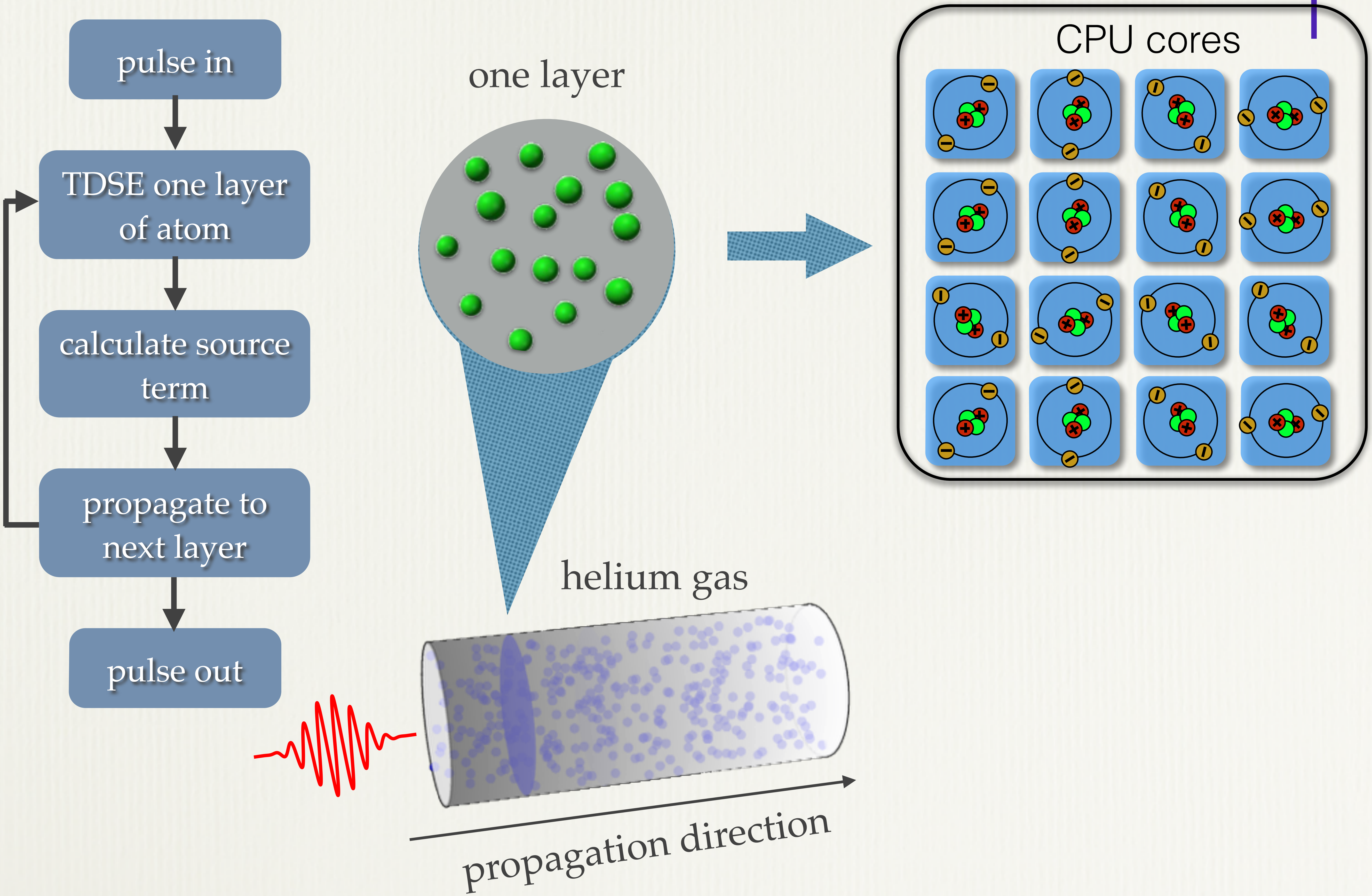

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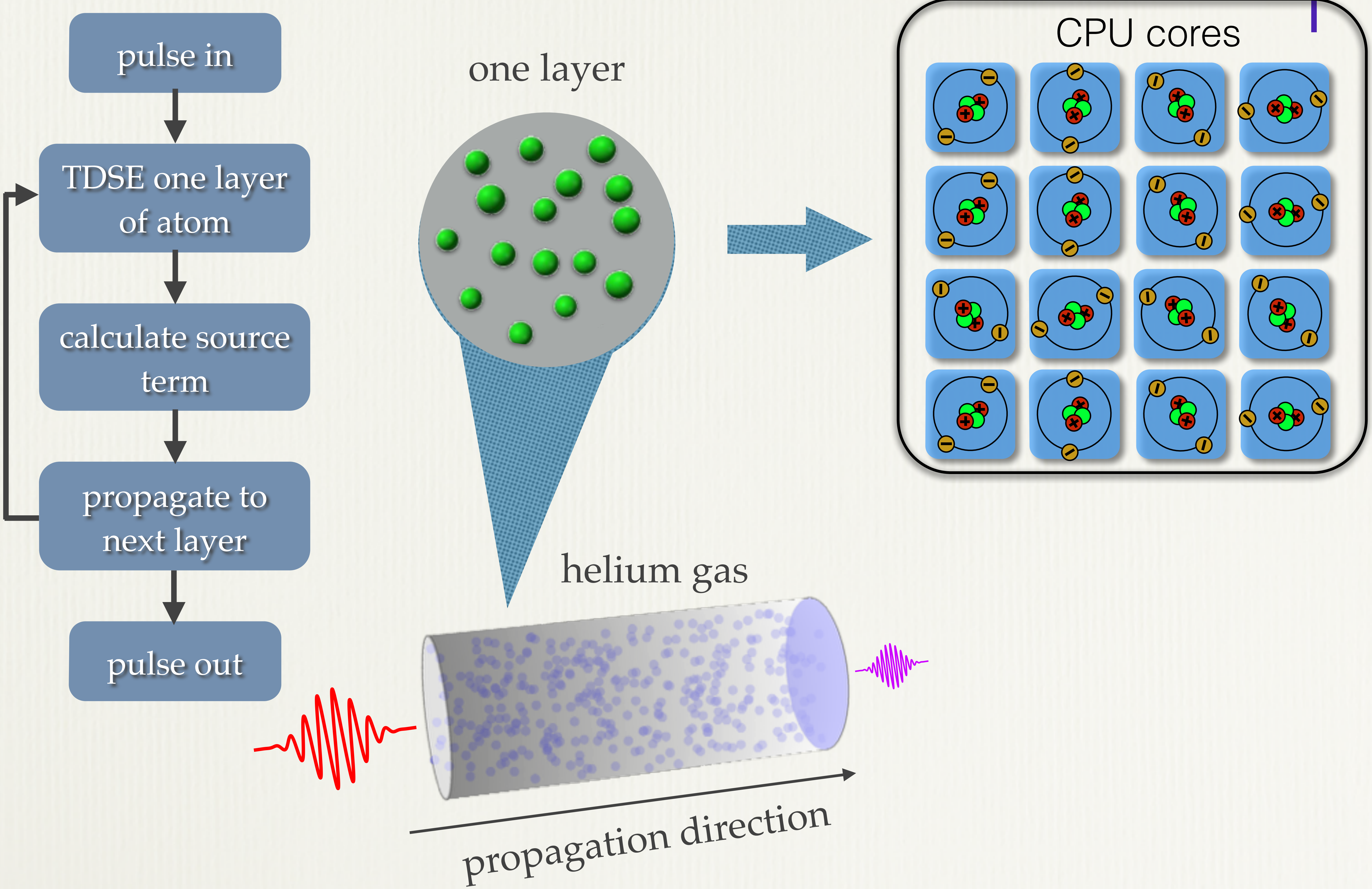

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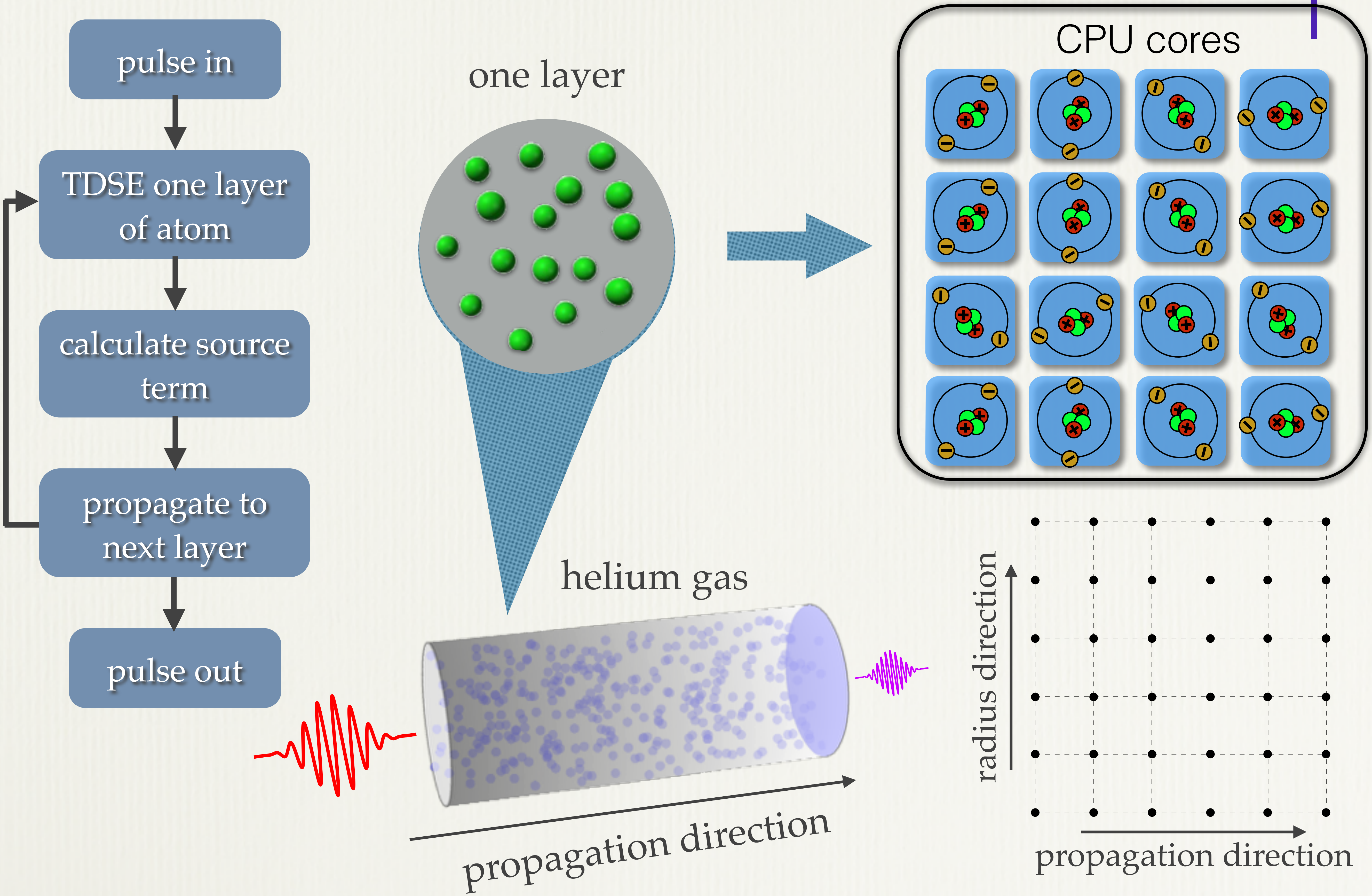

Methods, details

TDSE: Often we care only about transitions/overlap with ground state

$$d(t) = -\langle \psi(\vec{r}, t) | \psi_0 \rangle \langle \psi_0 | z | \psi(\vec{r}, t) \rangle + c.c.$$

Example, Argon has 3p ground state

$$d_s(t) = -e \langle \psi (t) | \phi_g \rangle \langle \phi_g | z | \psi_s(t) \rangle + \text{c.c.}$$

$$d_d(t) = -e \langle \psi (t) | \phi_g \rangle \langle \phi_g | z | \psi_d(t) \rangle + \text{c.c.}$$

$$\bar{d}_z(t) = d_s(t) + d_d(t)$$

MWE: Sometimes self-consistent solution is not stable/desired

Separate propagation of fundamental and generated fields

$$\nabla_\perp^2 \tilde{\mathcal{E}}_1 + \frac{2i\omega}{c} \frac{\partial \tilde{\mathcal{E}}_1}{\partial z} = \frac{1}{\epsilon_0 c^2} \tilde{FT}[\frac{\partial J(t)}{\partial t}]$$

$$\nabla_\perp^2 \tilde{\mathcal{E}}_X + \frac{2i\omega}{c} \frac{\partial \tilde{\mathcal{E}}_X}{\partial z} = \frac{1}{\epsilon_0 c^2} \tilde{FT}[\frac{\partial^2 (P_L(t) + P_{NL}(t))}{\partial t^2}]$$

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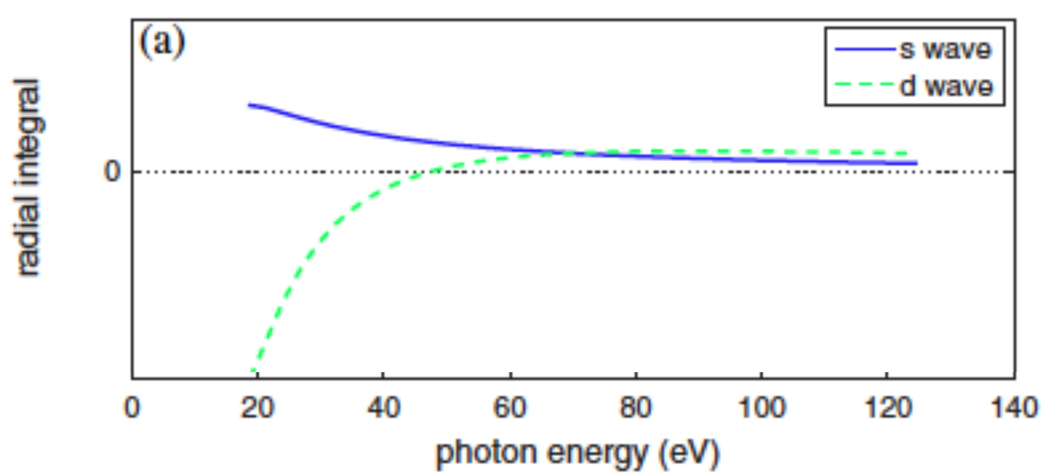
Example I: Cooper minimum in HHG

Schoun et al, PRL 2014



Worner et al, PRL 2009 Cooper, Phys Rev (1962)

Min in photoionization cross section due to zero in one angular momentum channel

Argon $\langle \phi_{3p} | z | \phi_{d,E} \rangle = 0$, at ≈ 47 eV

How does this manifest in HHG?
Three step model says:

$$D(\omega) = \sqrt{P_{ion}} \mathrm{e}^{i\phi_c(\omega)} p_{rdm}(\omega)$$

Recombination dipole moment:

$$p_{rdm}(\omega) = A_s(\omega)\mathrm{e}^{i\phi_s(\omega)} + A_d(\omega)\mathrm{e}^{i\phi_d(\omega)}$$

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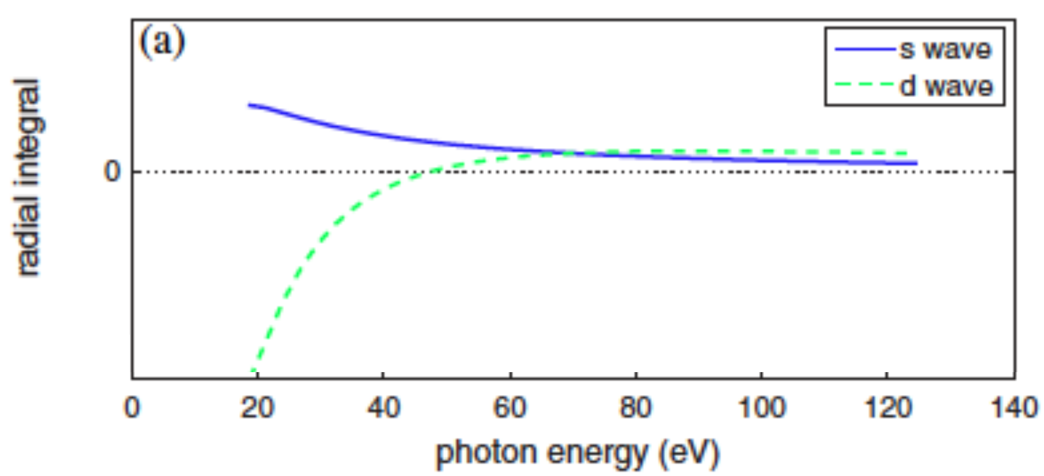

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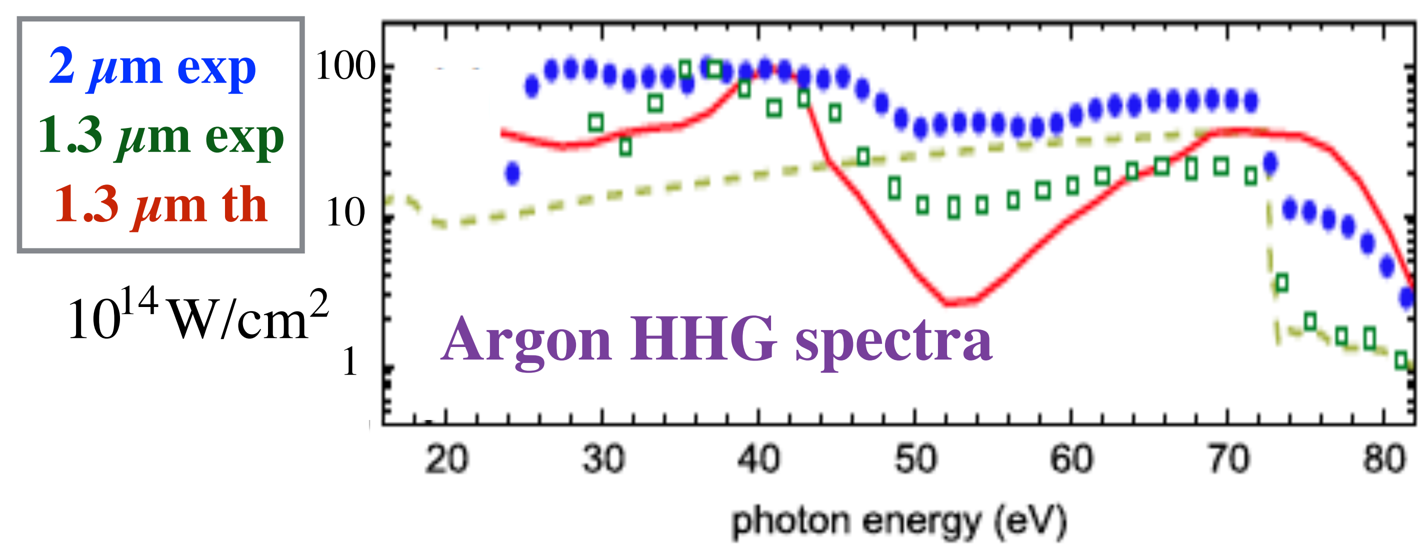
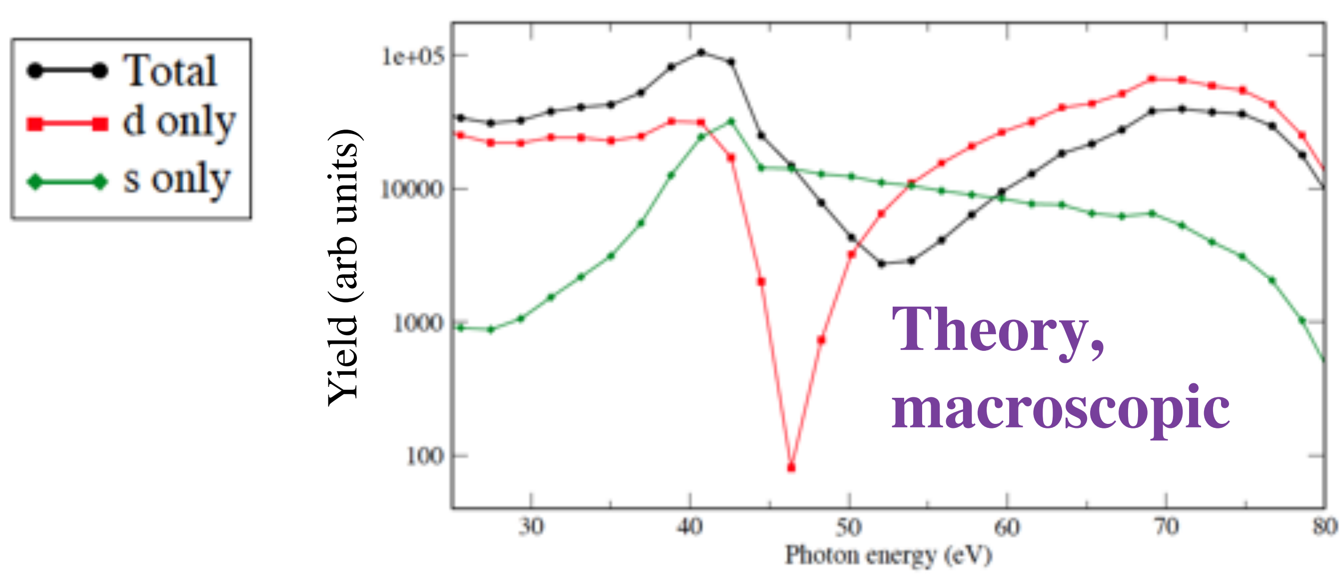
Coherent sum, HHG minimum is at higher energy than Cooper min





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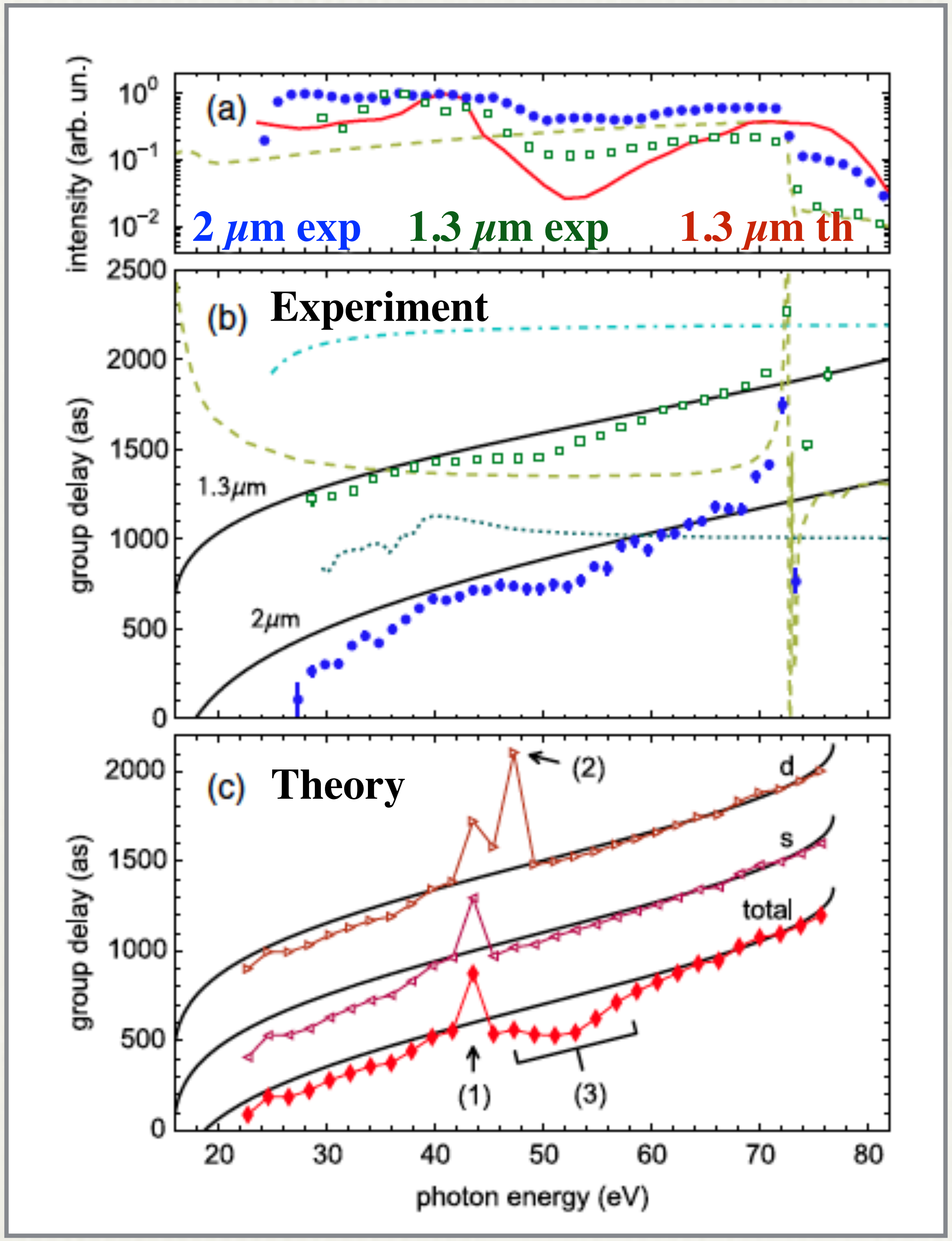
Example I: What's the point? Phase of RDM

Schoun et al, PRL 2014



Measure group delay $\frac{d\phi(\omega)}{d\omega}$ with RABITT

MIR advantage: higher cutoff and better resolution

Two wavelengths (1.3 μm, 2.0 μm), approx. 10^{14} W/cm^2

Optimized for short trajectory, spatially filtered

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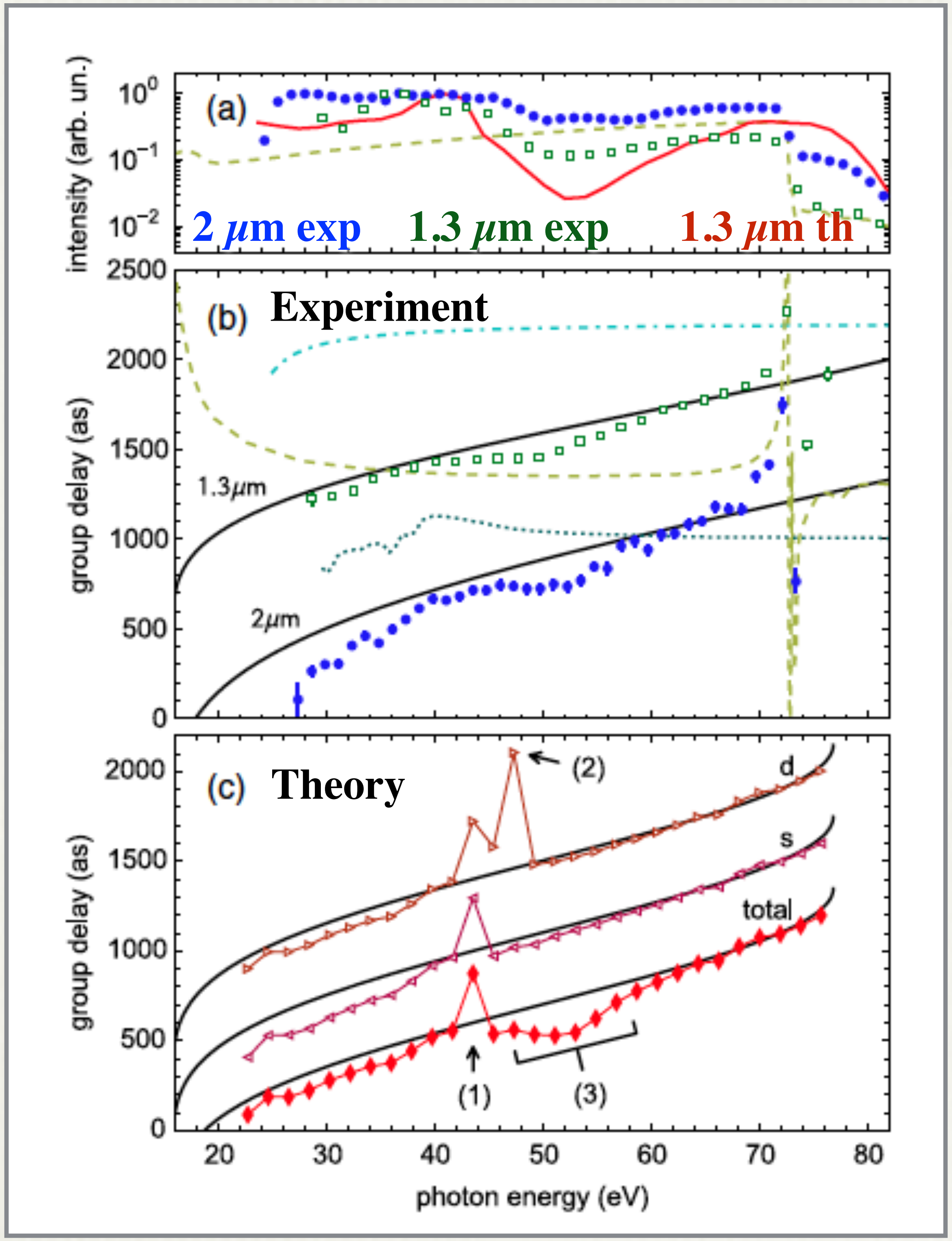

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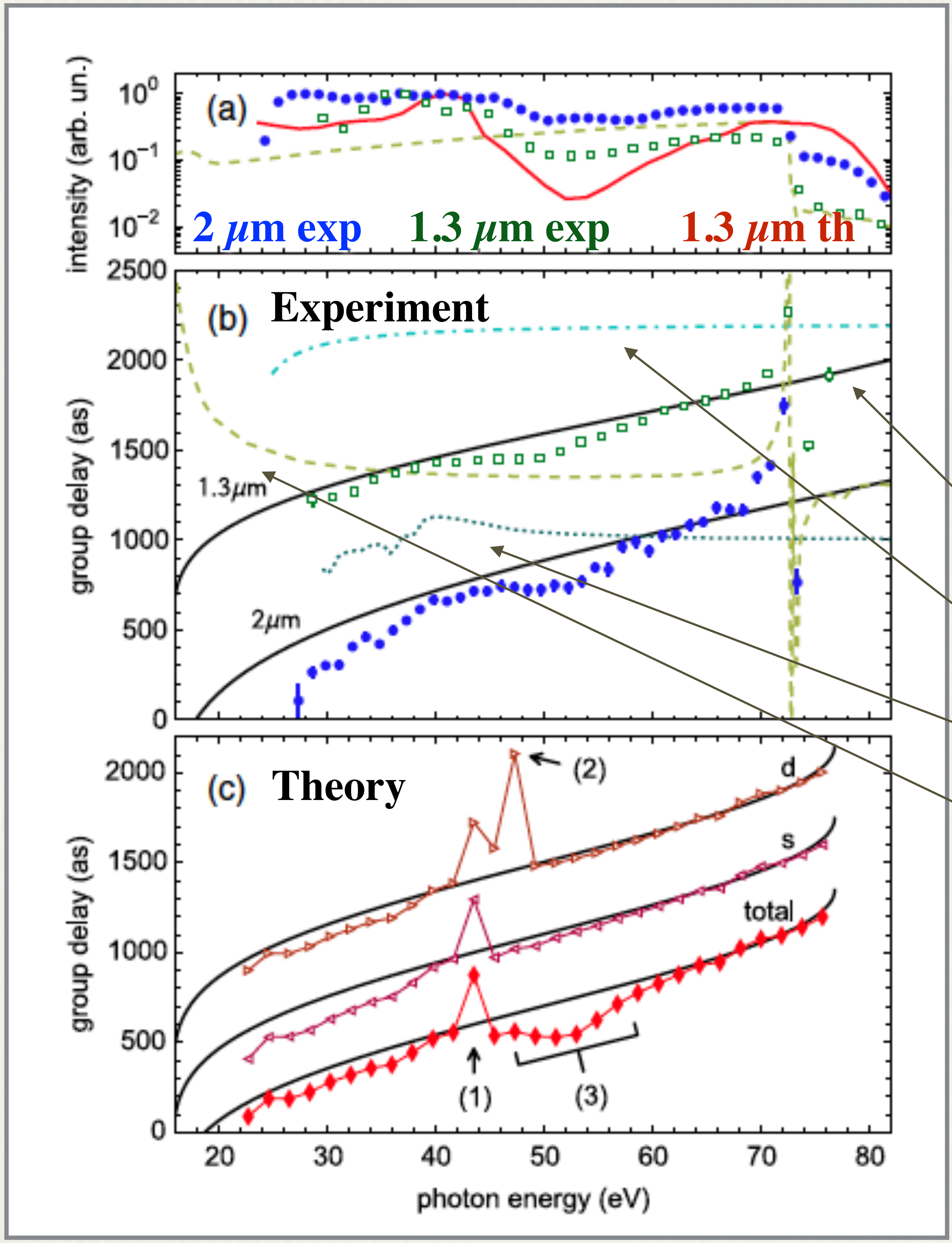

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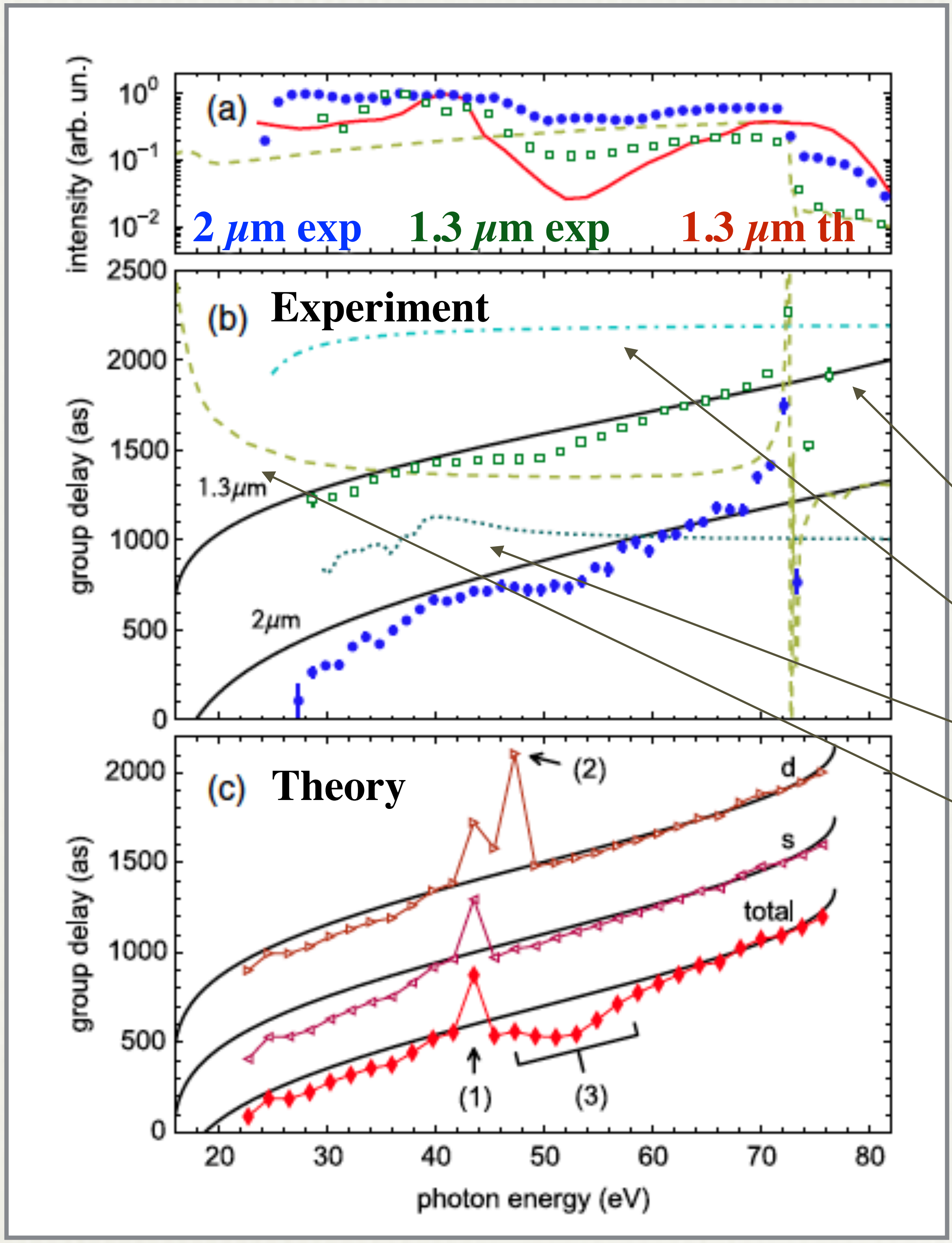

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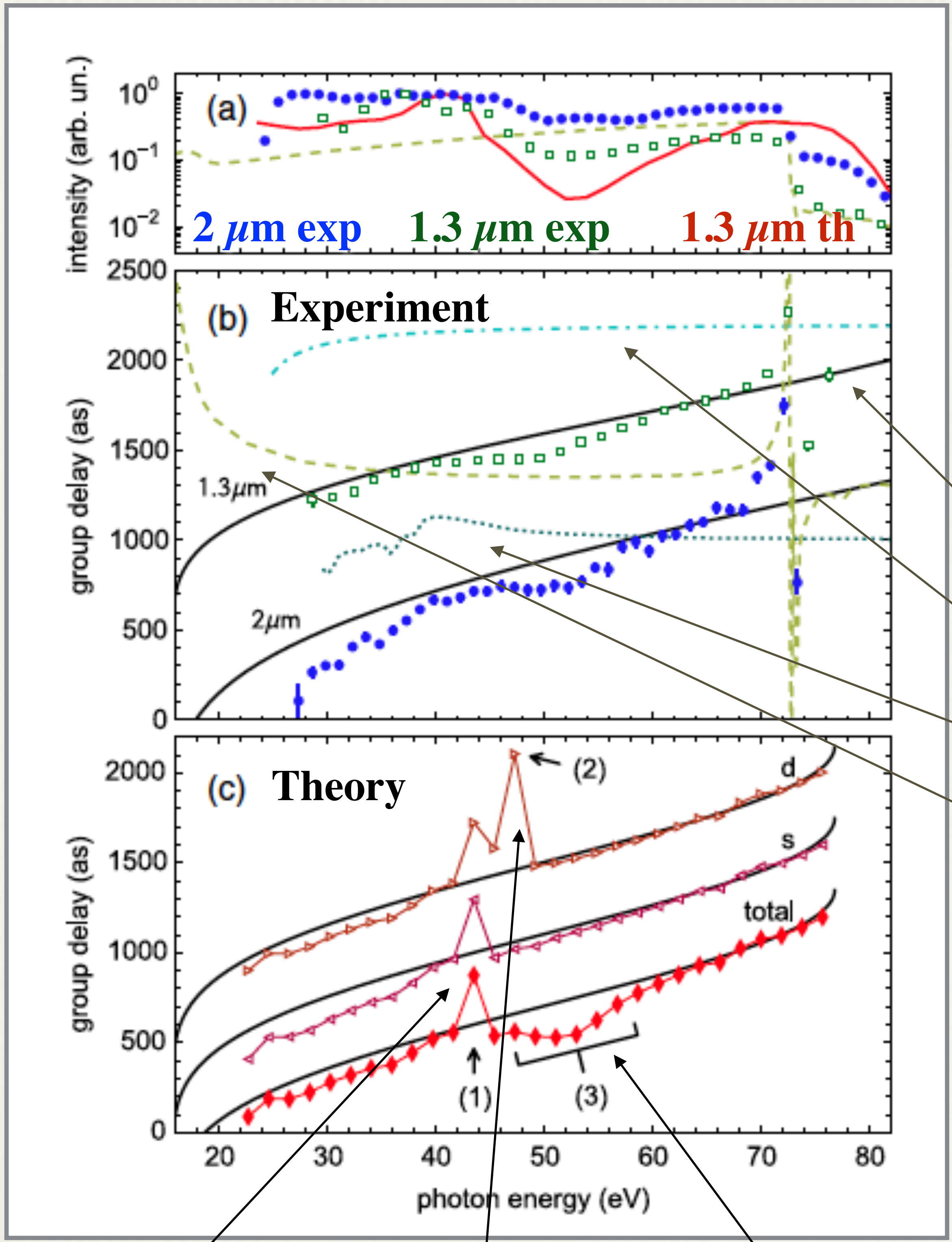

Ar dispersion

d phase jump

RDM phase

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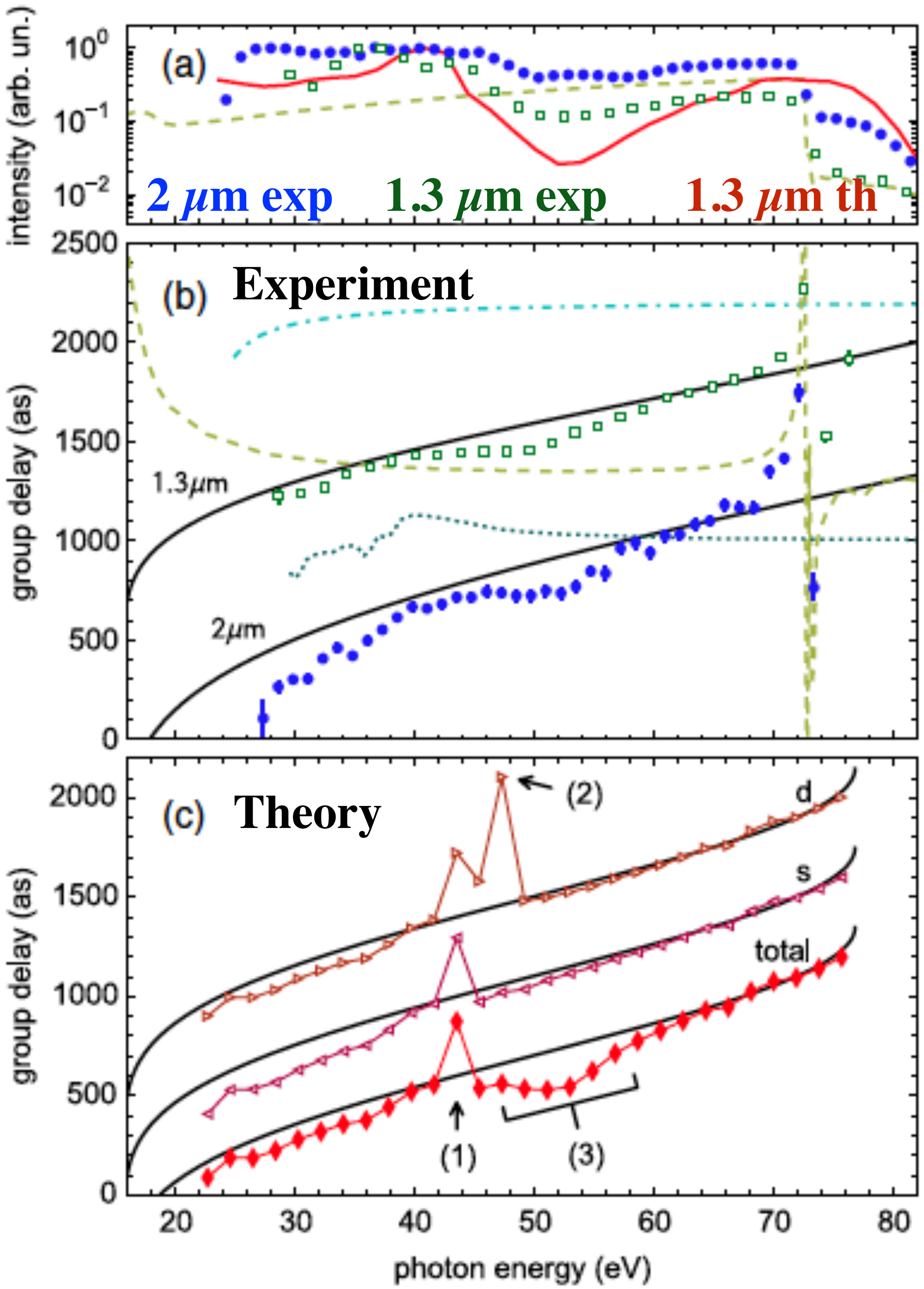

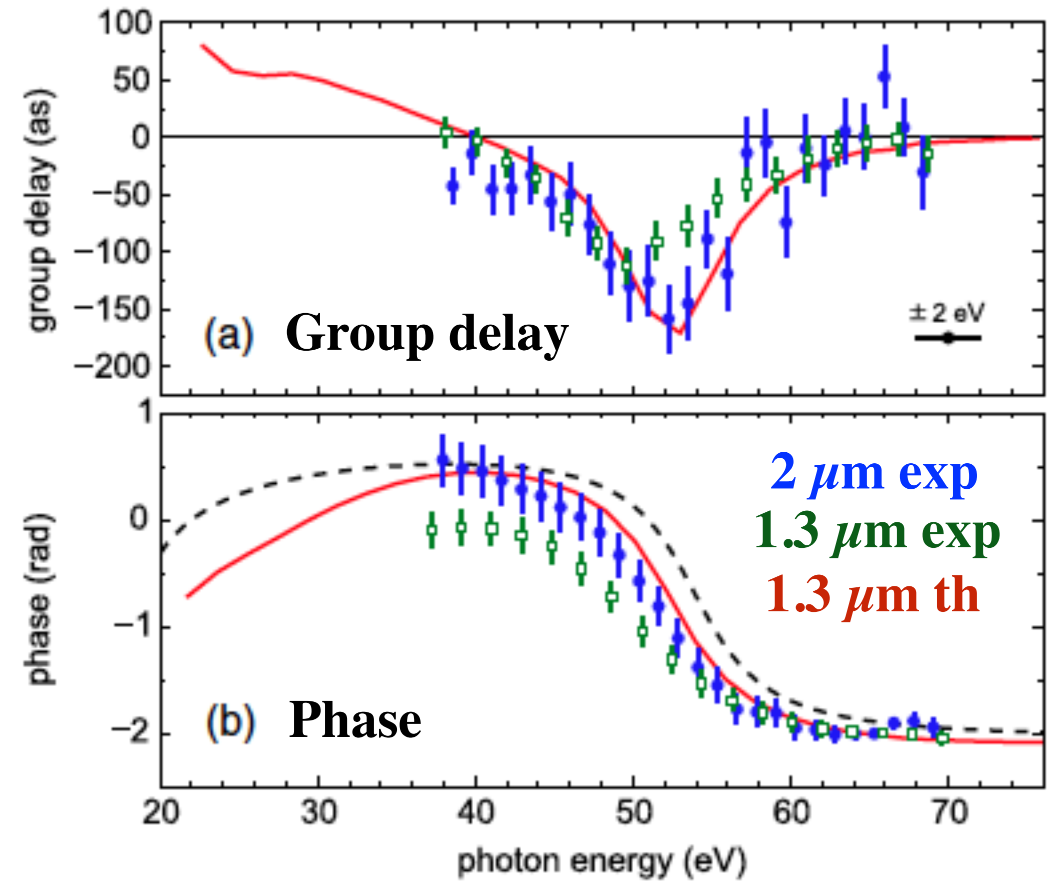

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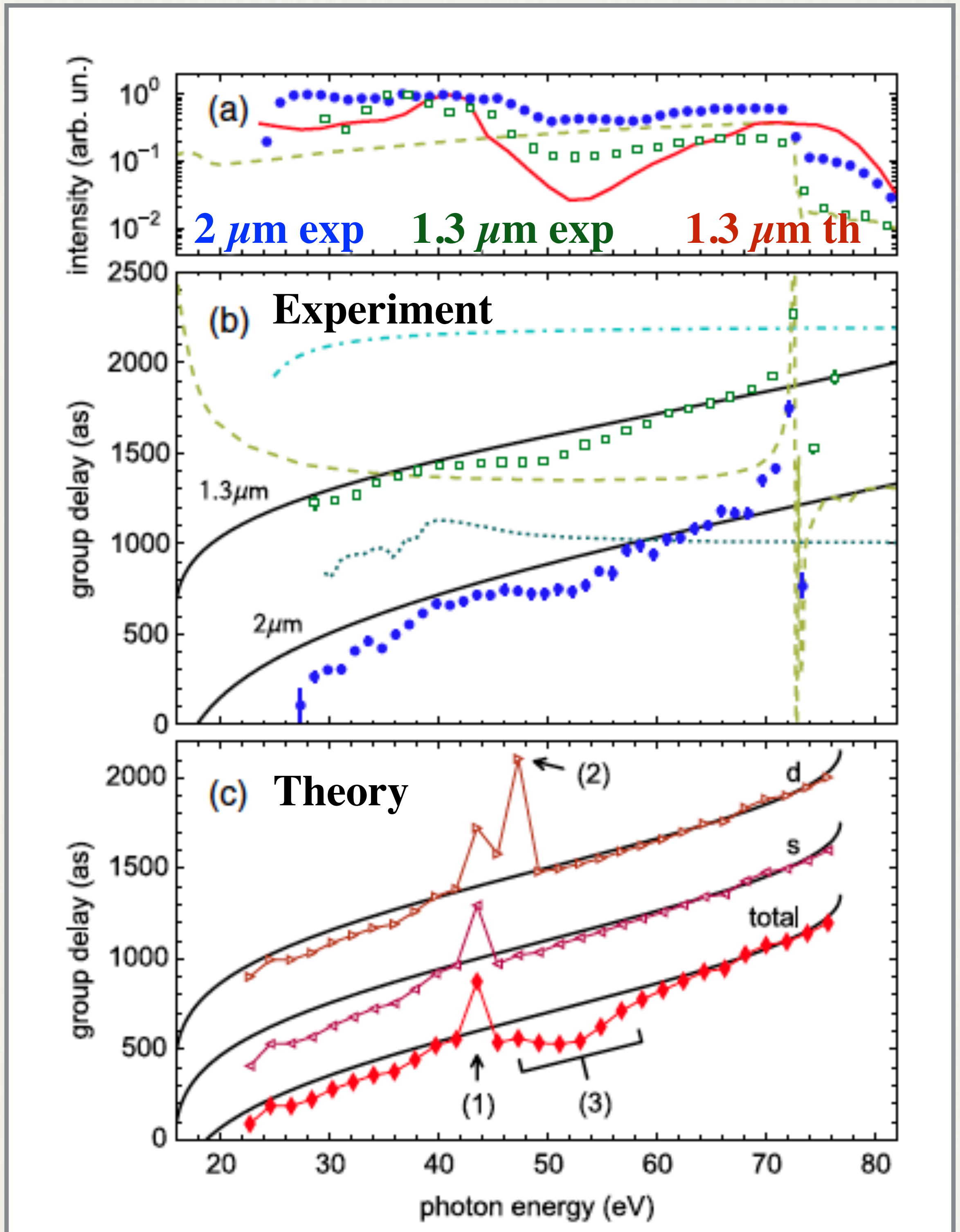

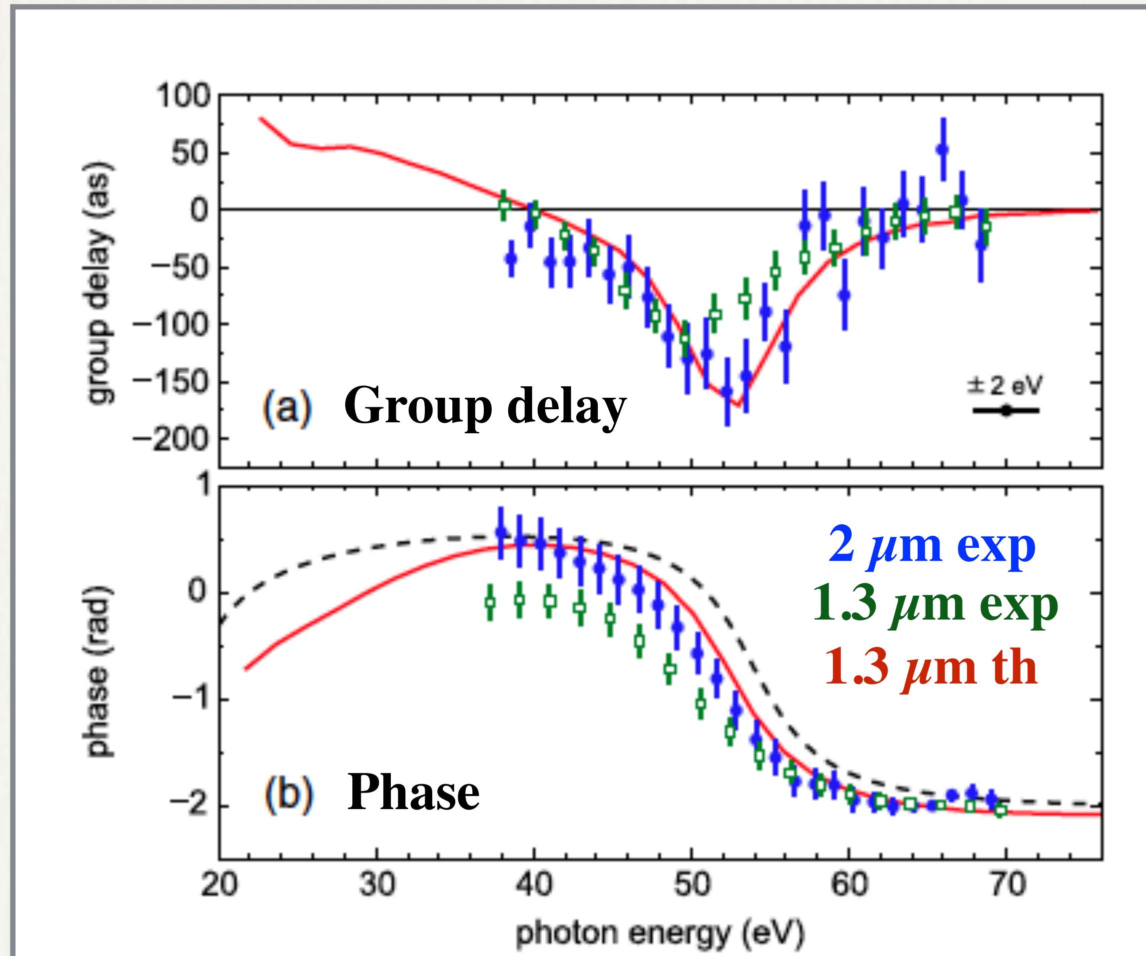

- Total RDM phase decreases by 2-2.5 rad over 20 eV

Le et al, PRA 2008

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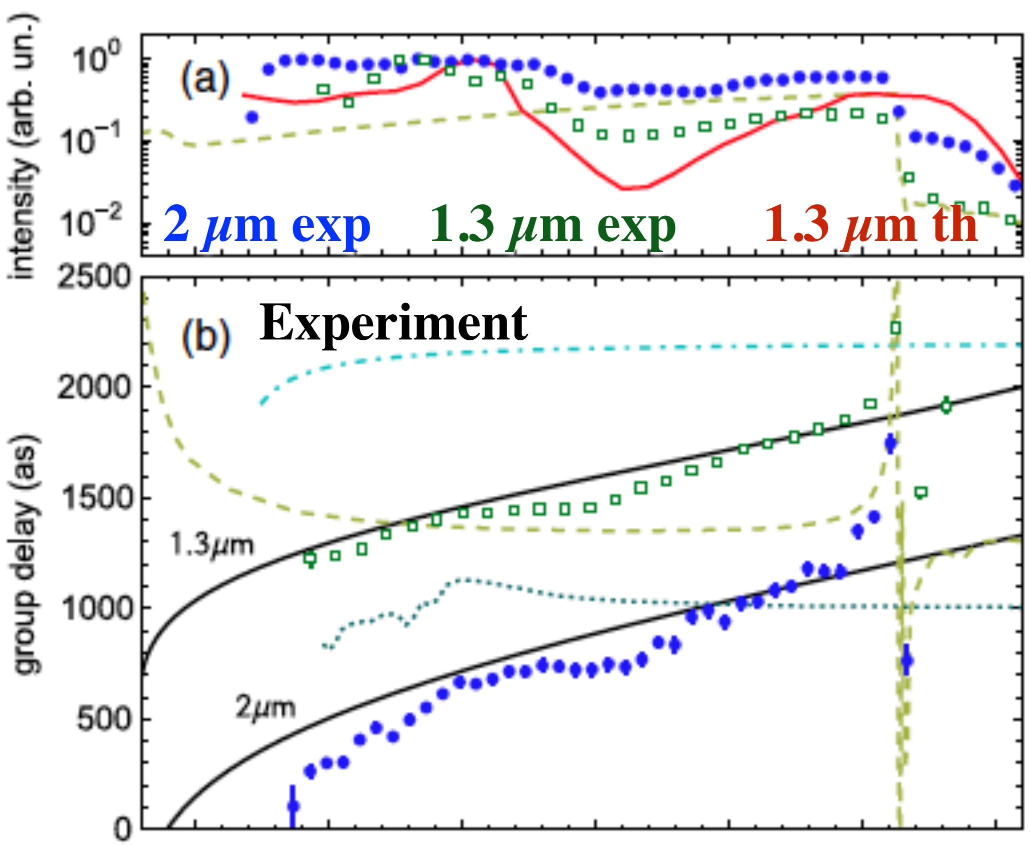

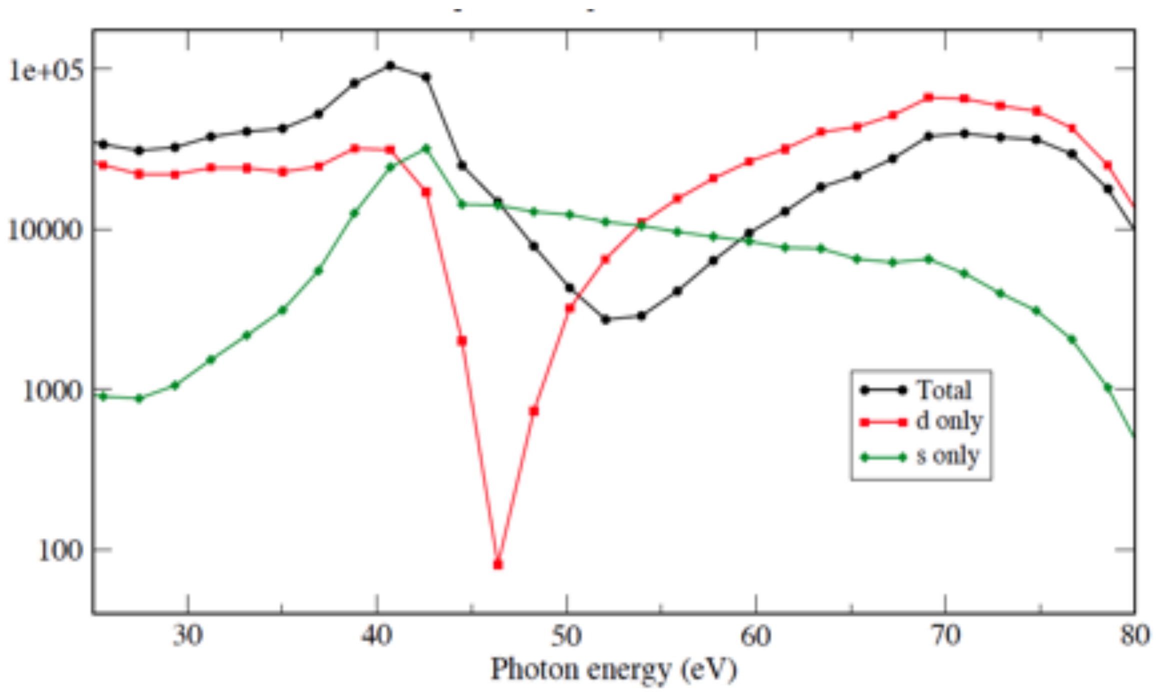

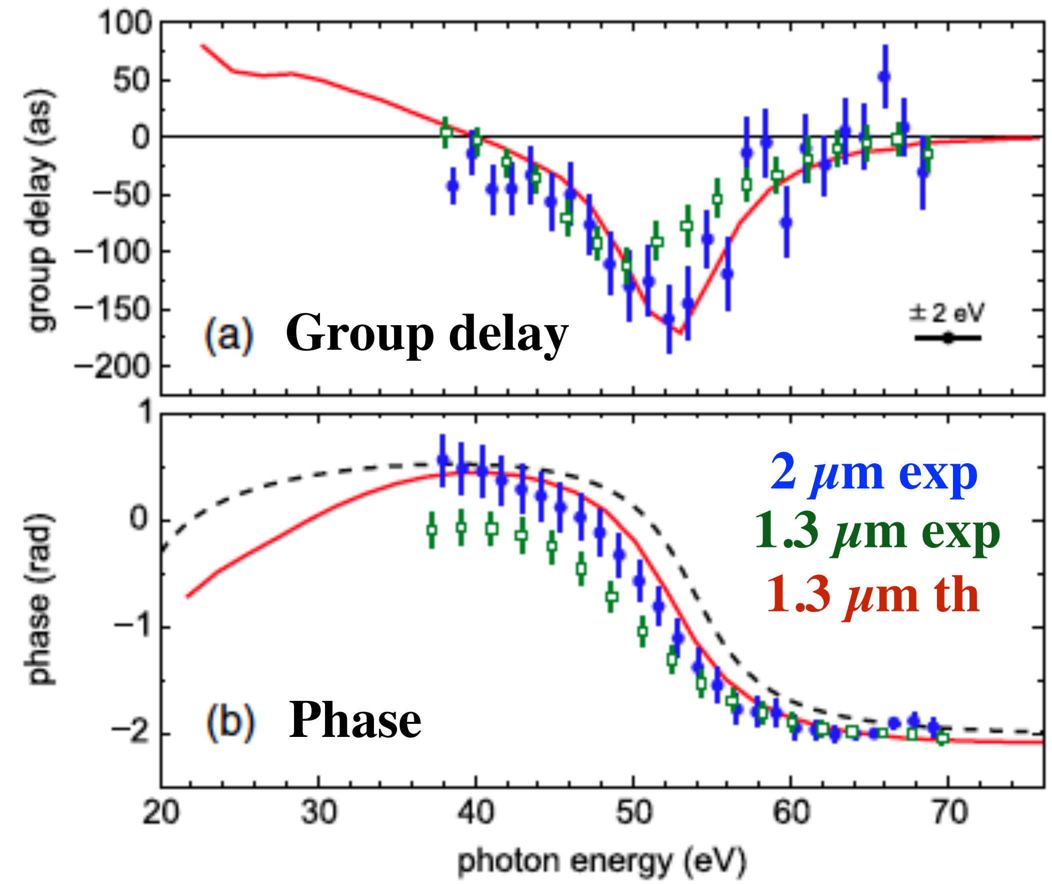

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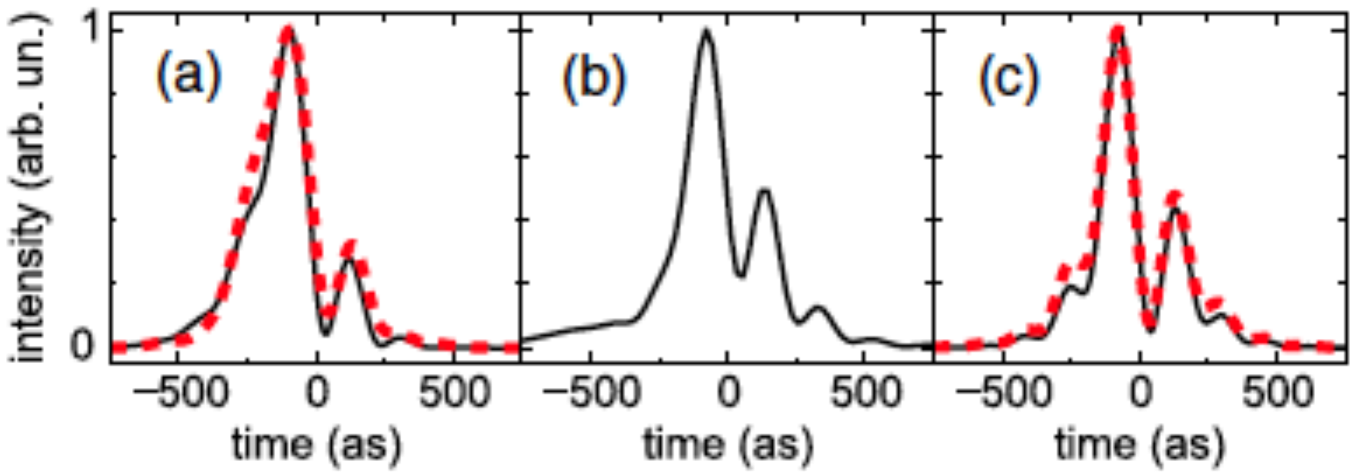
- Sharp π phase jump in d-channel moderated by "constant" s-channel

Consequences for XUV attosecond time profile

Schoun et al, PRL 2014

Sub-cycle time profile of plateau harmonics (24 eV FWHM)



1.3 μm exp
2 μm exp

1.3 μm TDSE

Simple model

- Returning EWP acquires spectral phase over large range
- Leads to reshaping of emitted attosecond pulses - **two pulses**
- In this case **group delay should not be interpreted as time delay**

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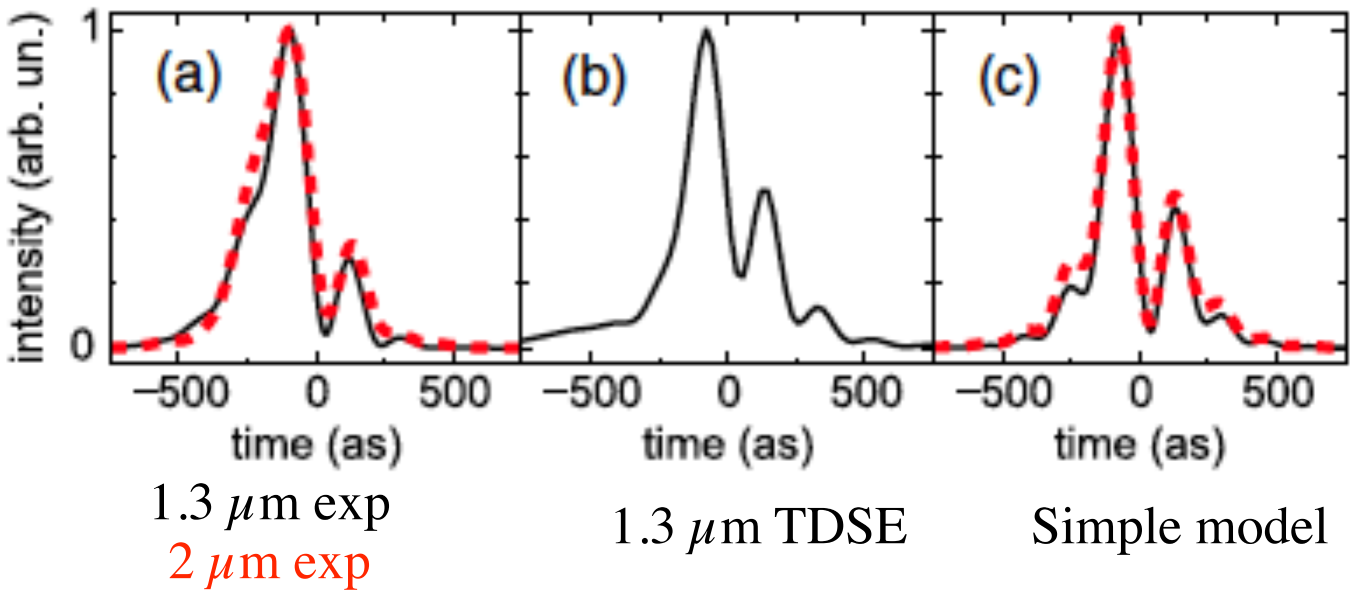

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$$p(\omega) = s(\omega)e^{i\eta_0(\omega)}[1 + \chi(\omega)e^{i\xi(\omega)}],$$

$$\chi(\omega) = \frac{d(\omega)}{s(\omega)}, \qquad \xi(\omega) = \eta_2(\omega) - \eta_0(\omega).$$

s(w) constant, d(w) linear: $\chi(\omega) = -\frac{\omega - \omega_C}{\Delta\omega}$

$\omega_C = 48.5$ eV

$\Delta\omega$ range of d(w) sign change

$\xi(\omega)$ scattering phases

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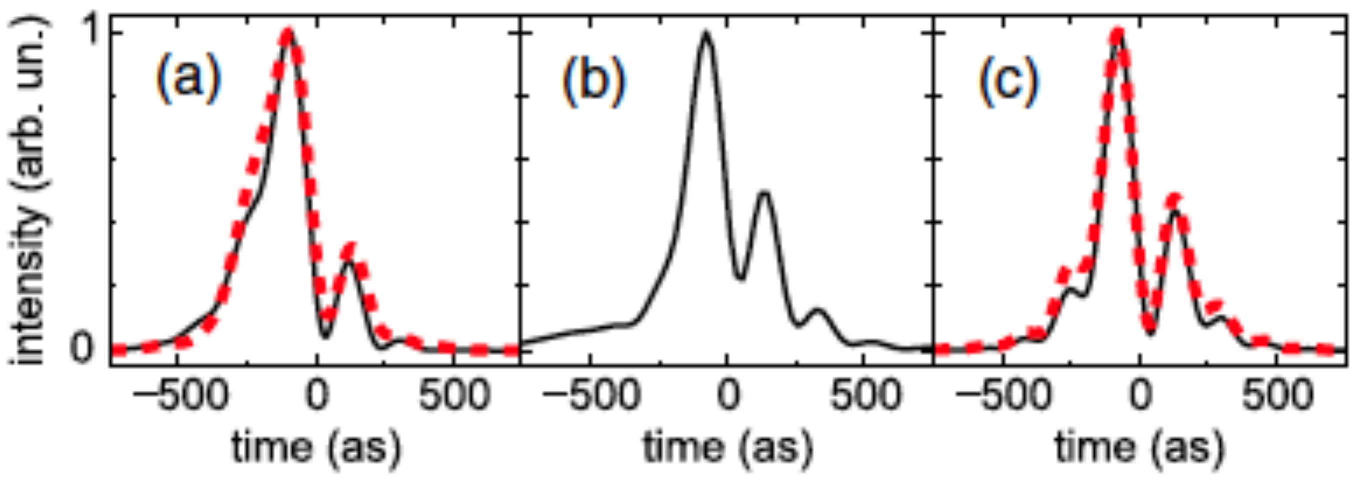

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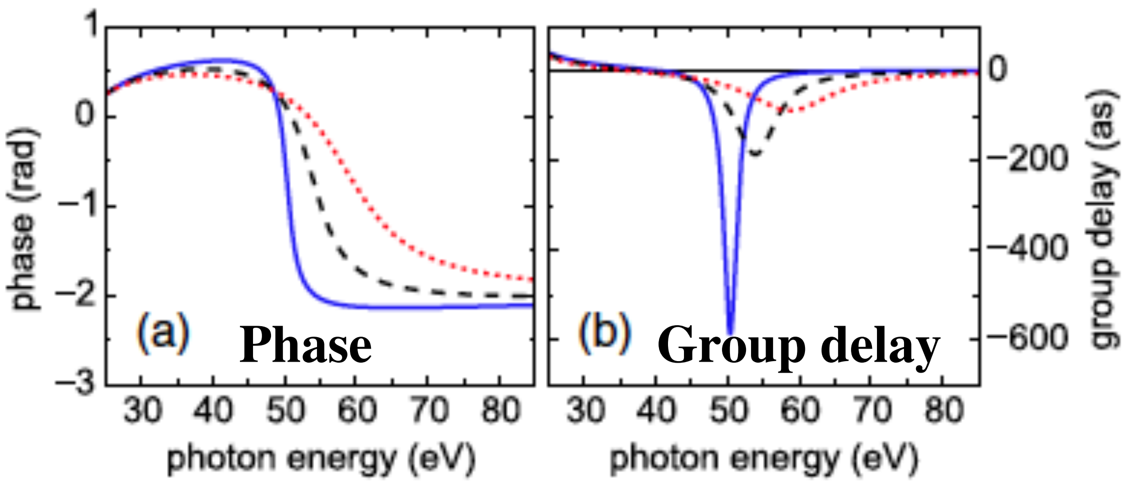
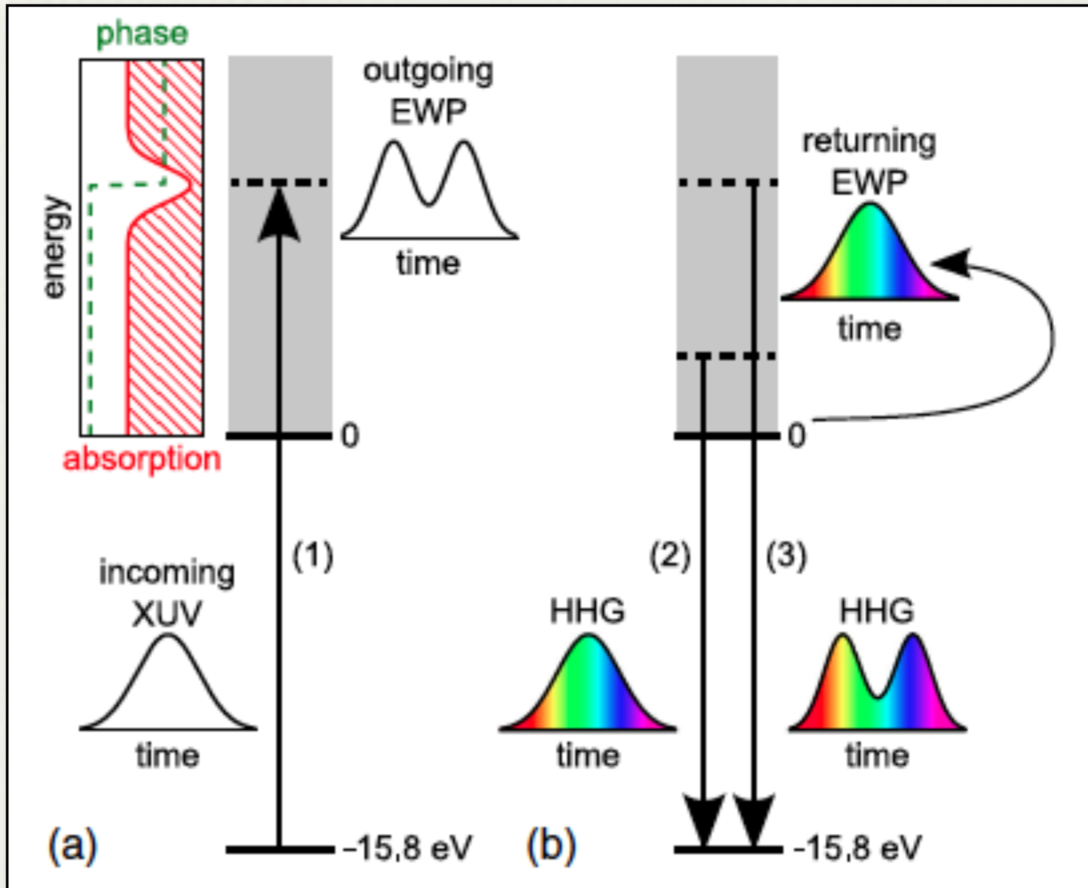
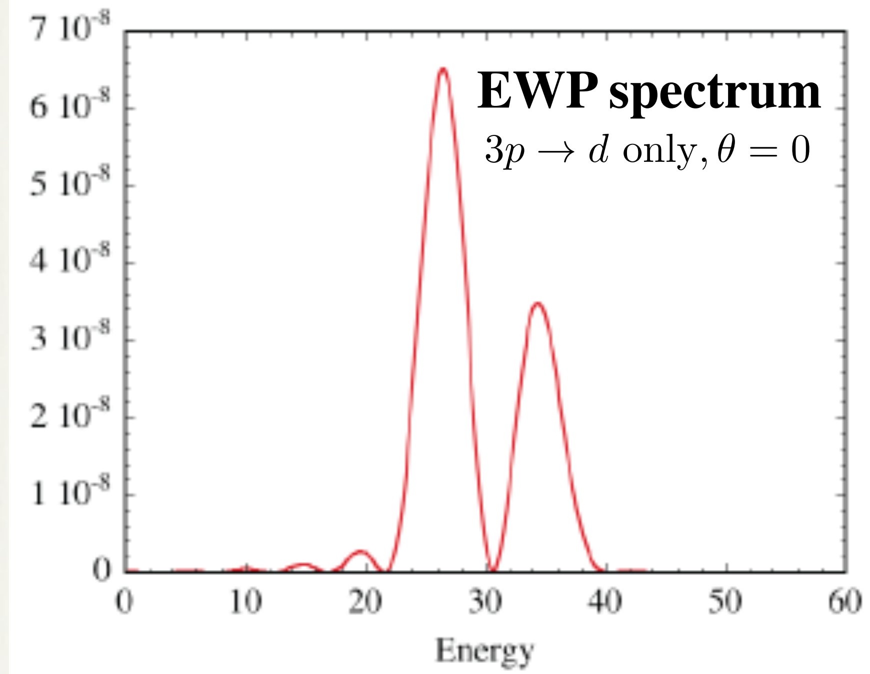


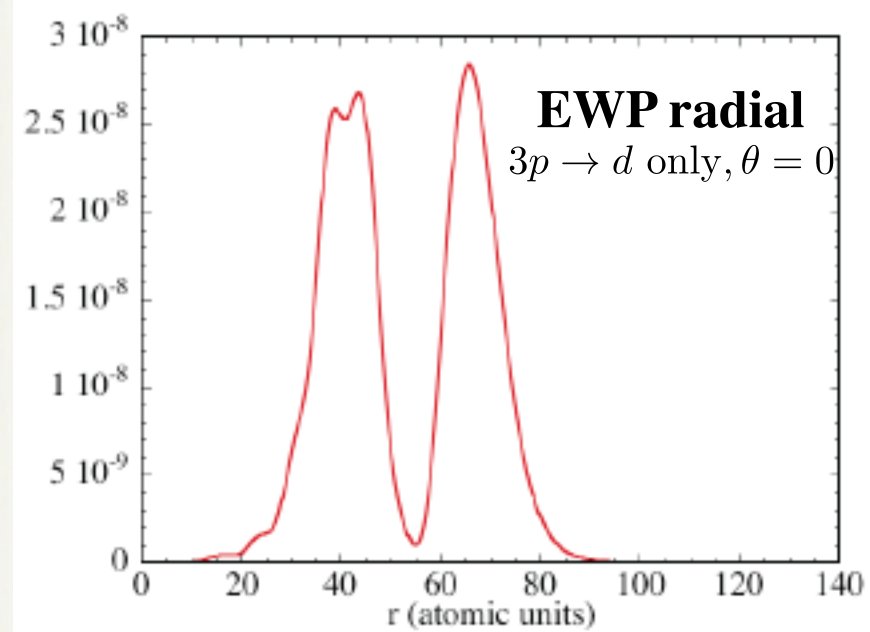
Rapid variation in d-channel can make group delay arbitrarily large - **not meaningful as a time delay**

Reciprocal nature of photoionization and recombination





The spectrum of electrons along theta=0 from a 47 eV pulse on argon where only the 3p -> Ed channel is open.



The electron wave packet along theta=0 about 1 fs after the center of a 300 as FWHM pulse. Only the 3p->Ed channel is open.

Hole in EWP created by photo ionization first discussed by Noordam et al. Measurement of EWP amplitude and phase challenging though

Hoogenraad et al, PRA 1998 Yakovlev et al, PRL 2010

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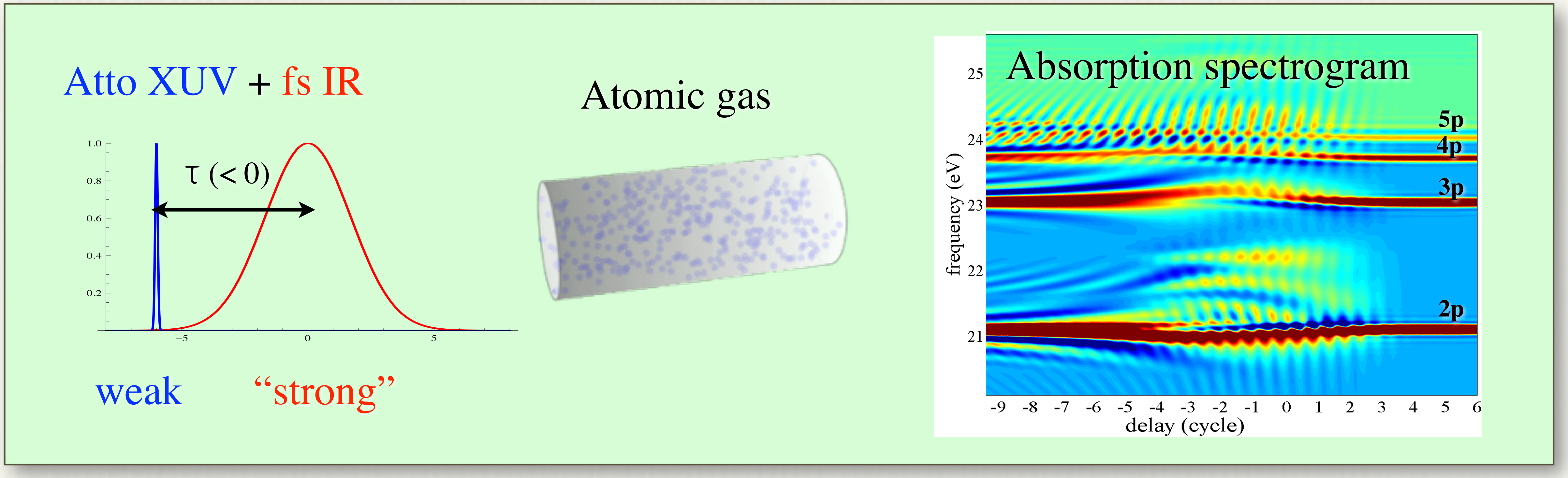
Theme: what happens to the XUV light as a result of it's interaction with the atomic system (here: structure)

- Introduce theoretical methods, coupled TDSE-MWE
- Example, HHG: Cooper minimum in argon reshapes asec pulses

  Collaboration Ohio State group
- **Example, transient absorption: Macroscopic propagation can also change absorption line shape in presence of IR**

  Collaboration Arizona group

Example II: Introduction to transient absorption



- Study electron dynamics from what happens to the XUV light

  From delay-dependence, absorption is time-integrated

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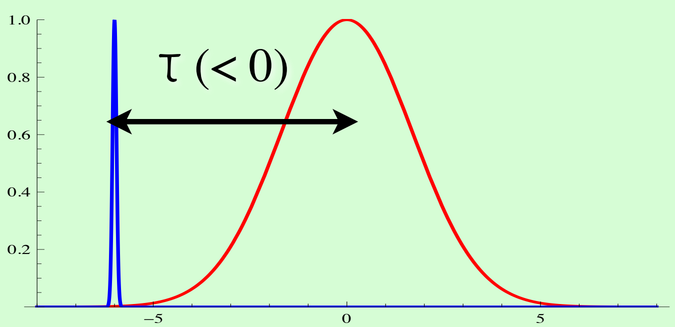
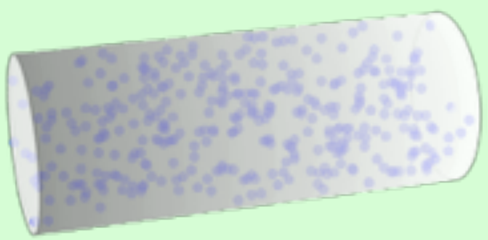
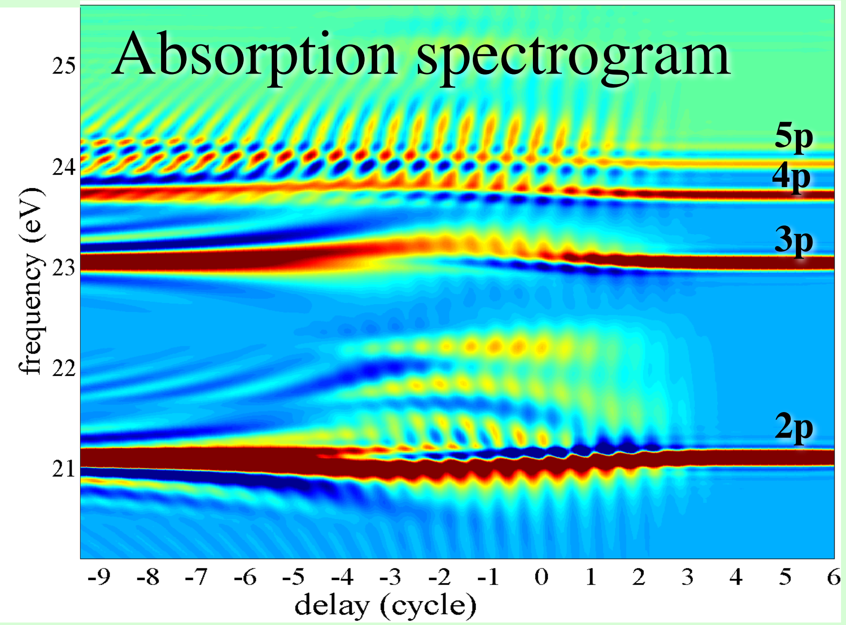
Atto XUV + fs IR



weak "strong"

Atomic gas





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Single atom response function $S(\omega) = 2\ \mathrm{Im}[d(\omega)E^*(\omega)]$ find d(t) from TDSE

absorption probability per frequency, depends on light field

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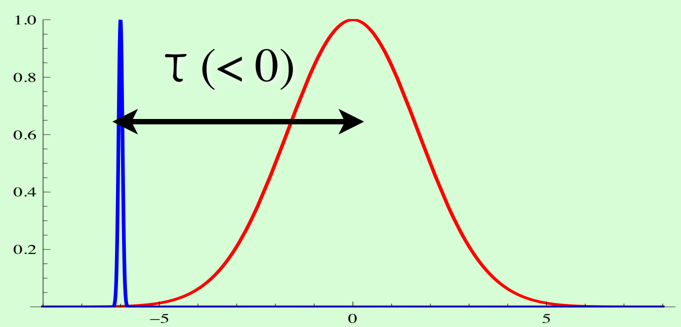

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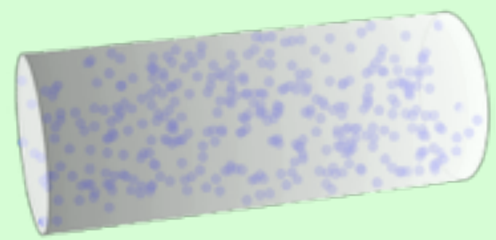

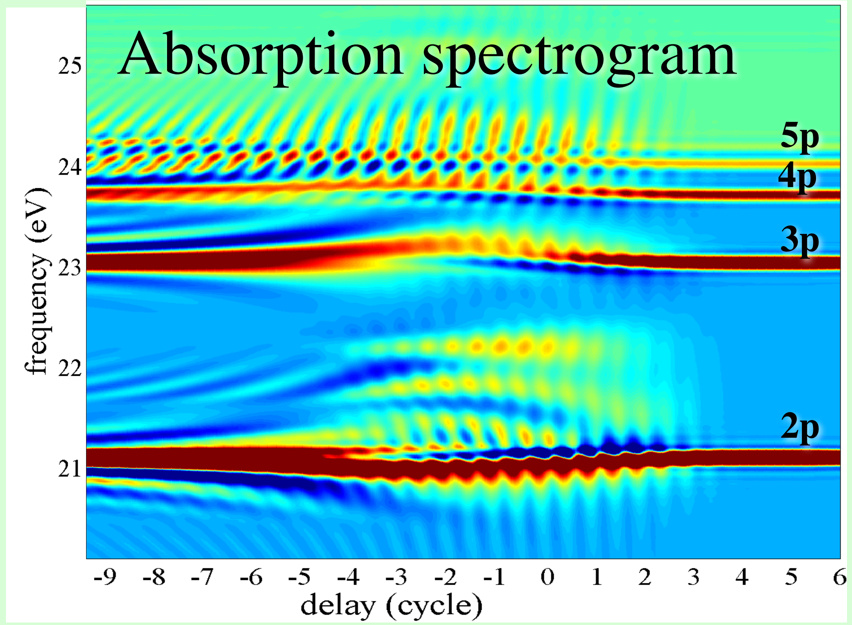

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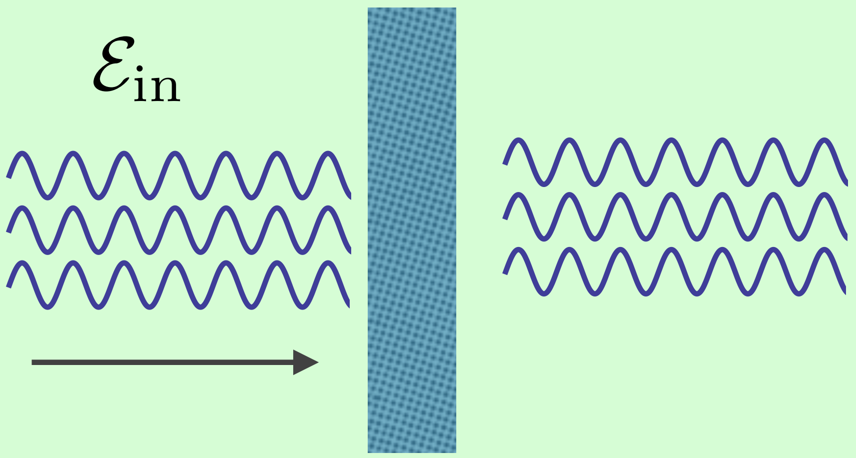
absorption probability per frequency, depends on light field

Macroscopic optical density $\mathrm{OD} = -\log[I_{out}/I_{in}]$ find E(t,r,z) by solving coupled, self-consistent TDSE and MWE

note: if $I(z) = I_0 e^{-\alpha z}$ then OD prop. to cross section

Example II: Absorption in the time domain

What happens to E-field in medium



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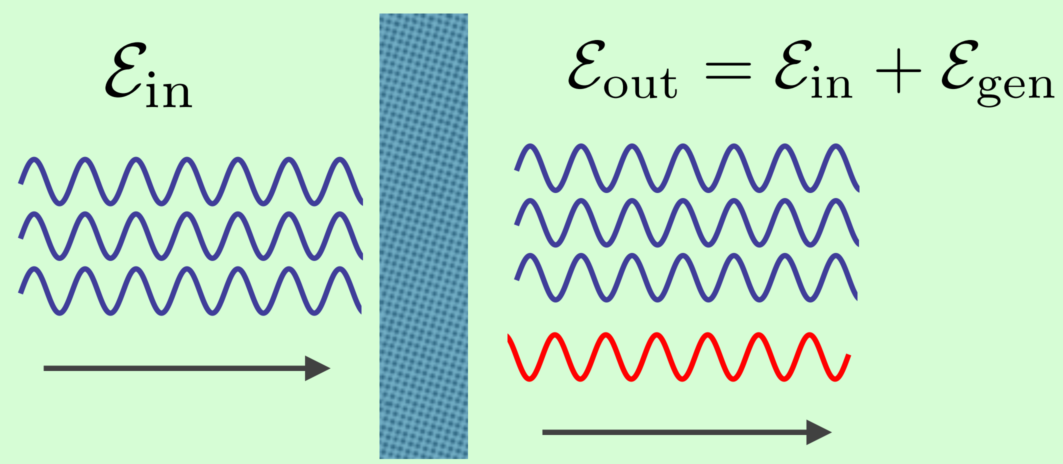

- Absorption happens when generated E-field is π out of phase with driving E-field

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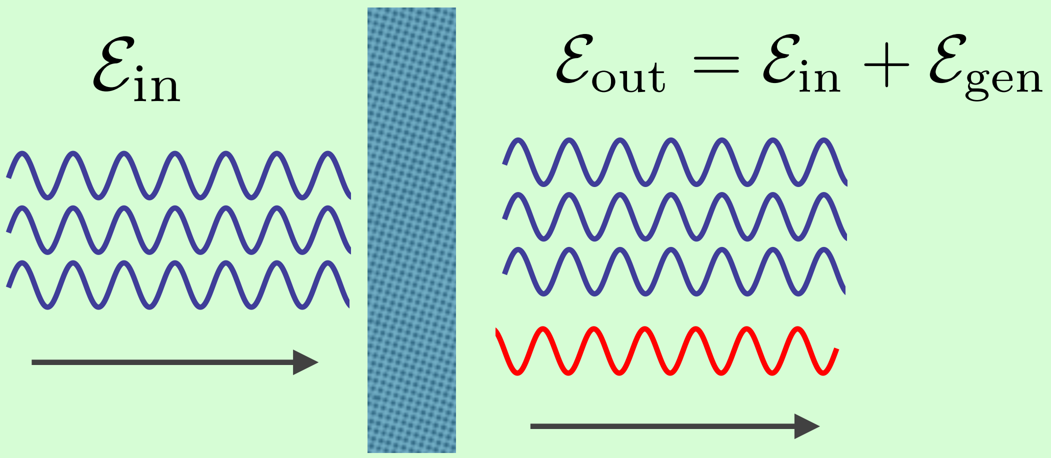

- Absorption happens when generated E-field is π out of phase with driving E-field
- This means that time-dependent dipole moment is π/2 out of phase with driving field

$$\frac{2i\omega}{c}\frac{\partial E}{\partial z} = -\frac{\omega^2}{\epsilon_0 c^2}P \qquad \sigma(\omega) = 2\mathrm{Im}[\frac{d(\omega)}{E(\omega)}]$$

- Response function $S(\omega) = 2\ \mathrm{Im}[d(\omega)E^*(\omega)]$

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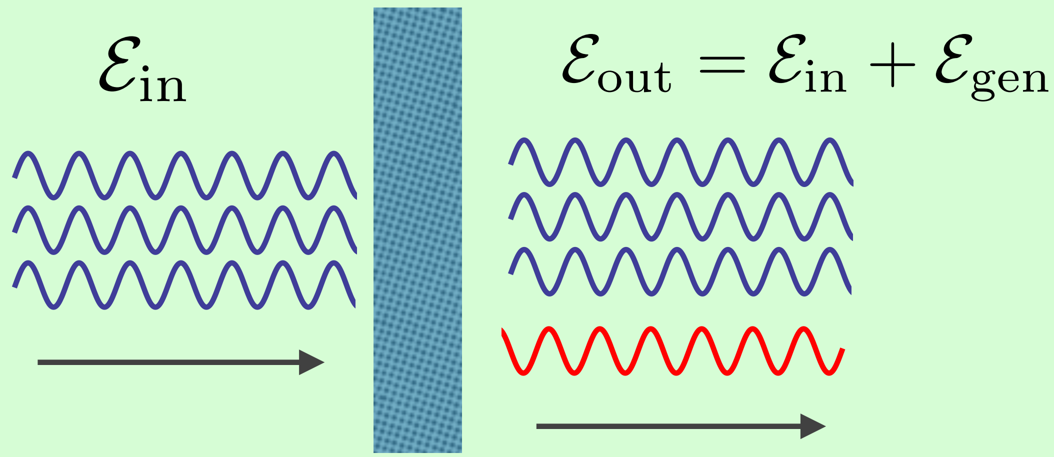

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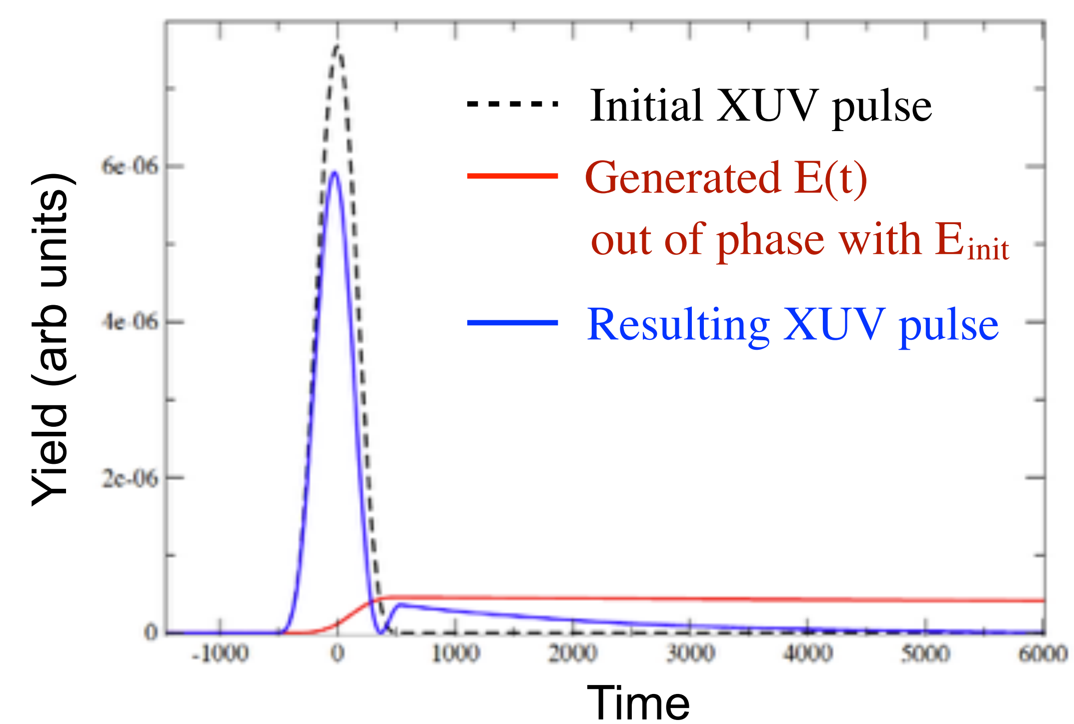

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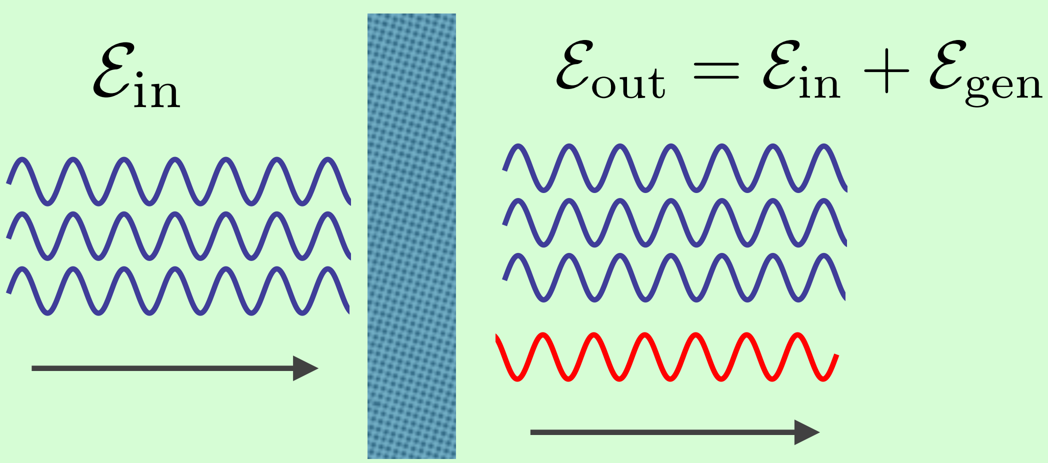

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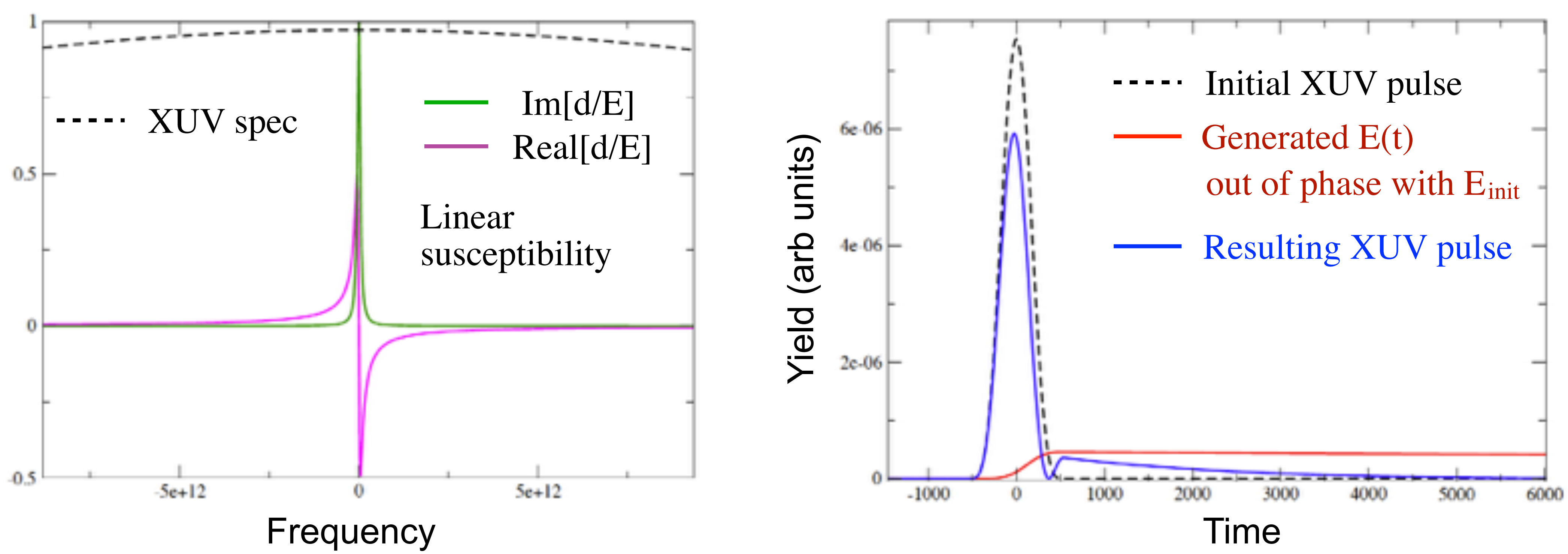
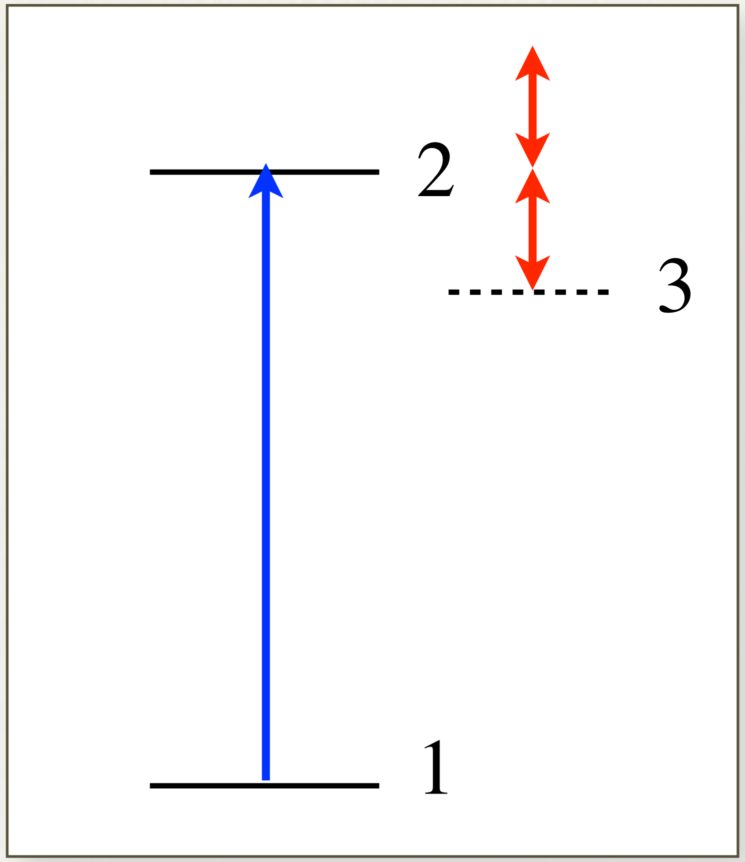


Example II: Line shape changes

Chen et al., PRA 2013

- One photon excitation: absorption
- Access excitation more than once.
  This adds a **phase shift** to d(t)



Autoionization: Heidelberg (Ott/Pfeifer) CFEL (Pabst/Santra) Madrid (Argenti/Martin) Kansas (Chu/Lin)

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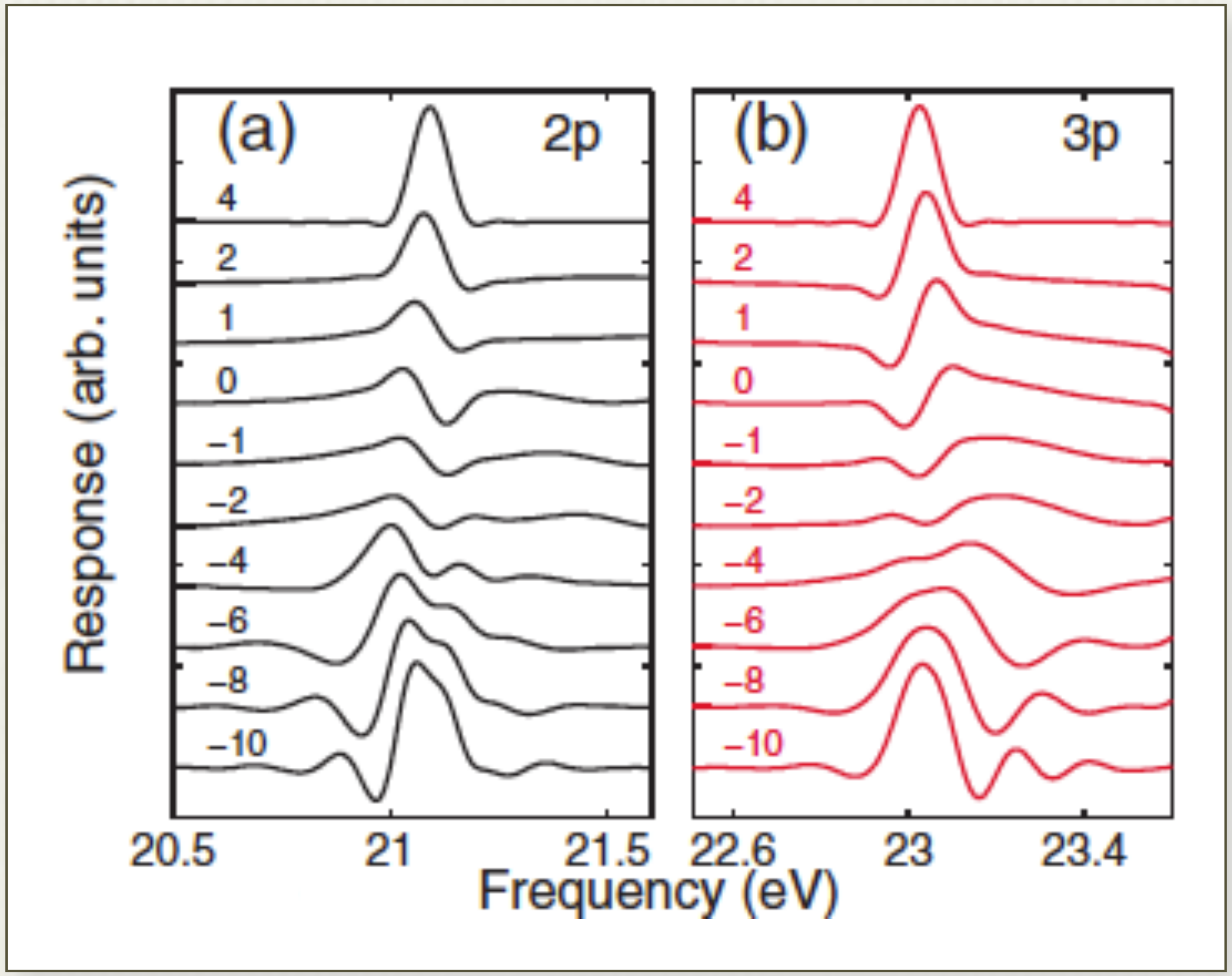

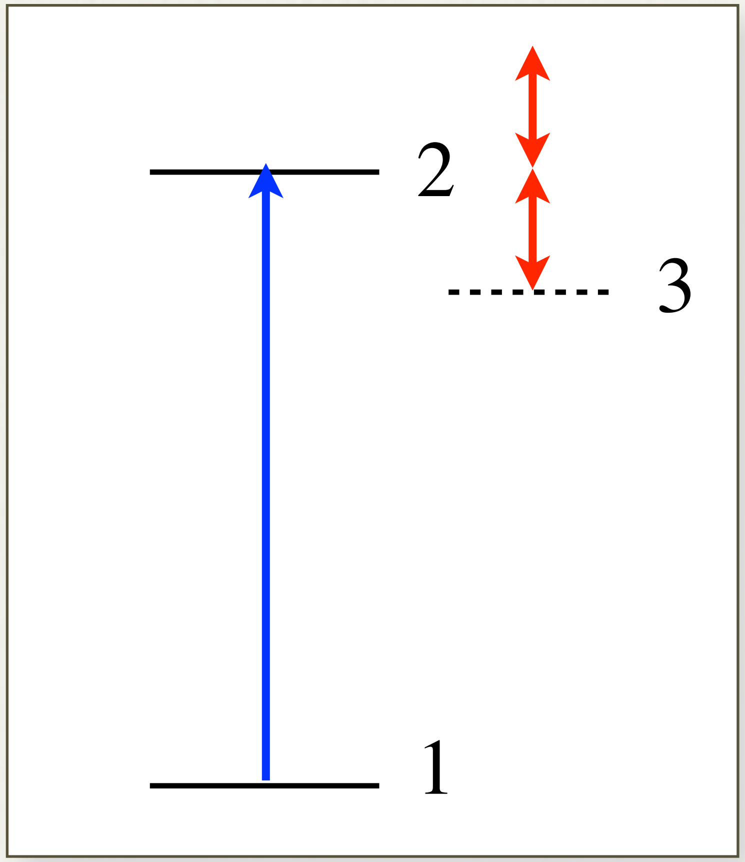

Stark effect

$$\phi(t,\tau) = \int_\tau^t \left[\delta E_{np}(t') - \delta E_{1s}(t')\right] dt'$$

$$\tilde{S}(\omega,\tau) \approx b \cdot Re\left[e^{i\phi_0(\tau)} \int_\tau^{\tau+T_2} e^{-i(\omega-\omega_{21})t} e^{-t/T_2} dt\right]$$

$$\approx \mathcal{L}(\omega)\left[\cos(\phi_0) + (\omega-\omega_{21})T_2 \sin(\phi_0)\right]$$

Autoionization: Heidelberg (Ott/Pfeifer) CFEL (Pabst/Santra) Madrid (Argenti/Martin) Kansas (Chu/Lin)

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Example II: Macroscopic reshaping of absorption line

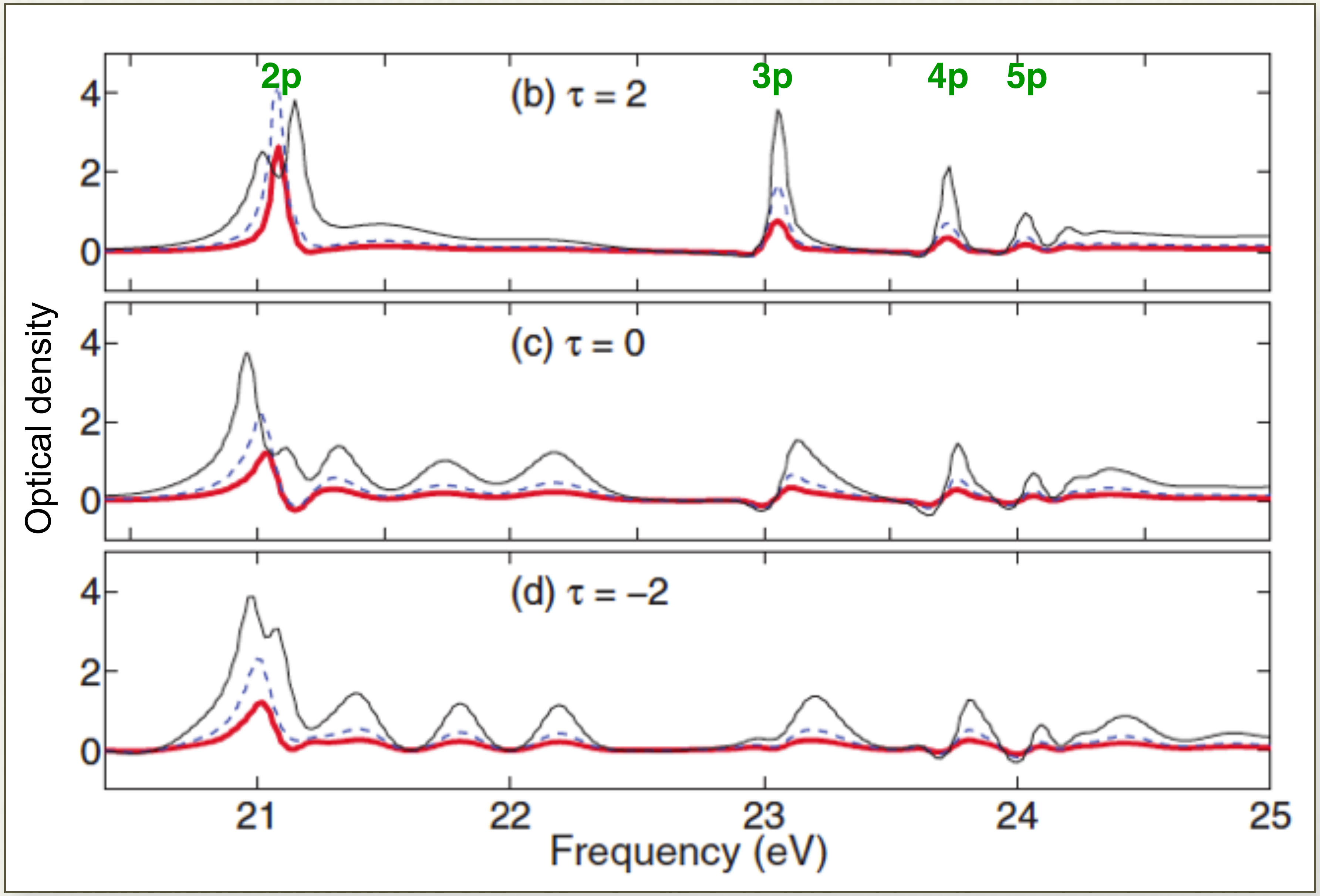
Chen et al., PRA 2013

Low dens (1e17)

High dens (5e17)

XUV 400 as, 25 eV

IR 800 nm, 1e12 W/cm2



- Broadening, shifts, new spectral features

  Also observed experimentally (Sandhu & Liao)

LSU (Gaarde et al, PRA 2011, WU et al PRA 2013) Kansas (Chu & Lin, PRA 2012) Berkeley (Pfeiffer et al PRA 2013)

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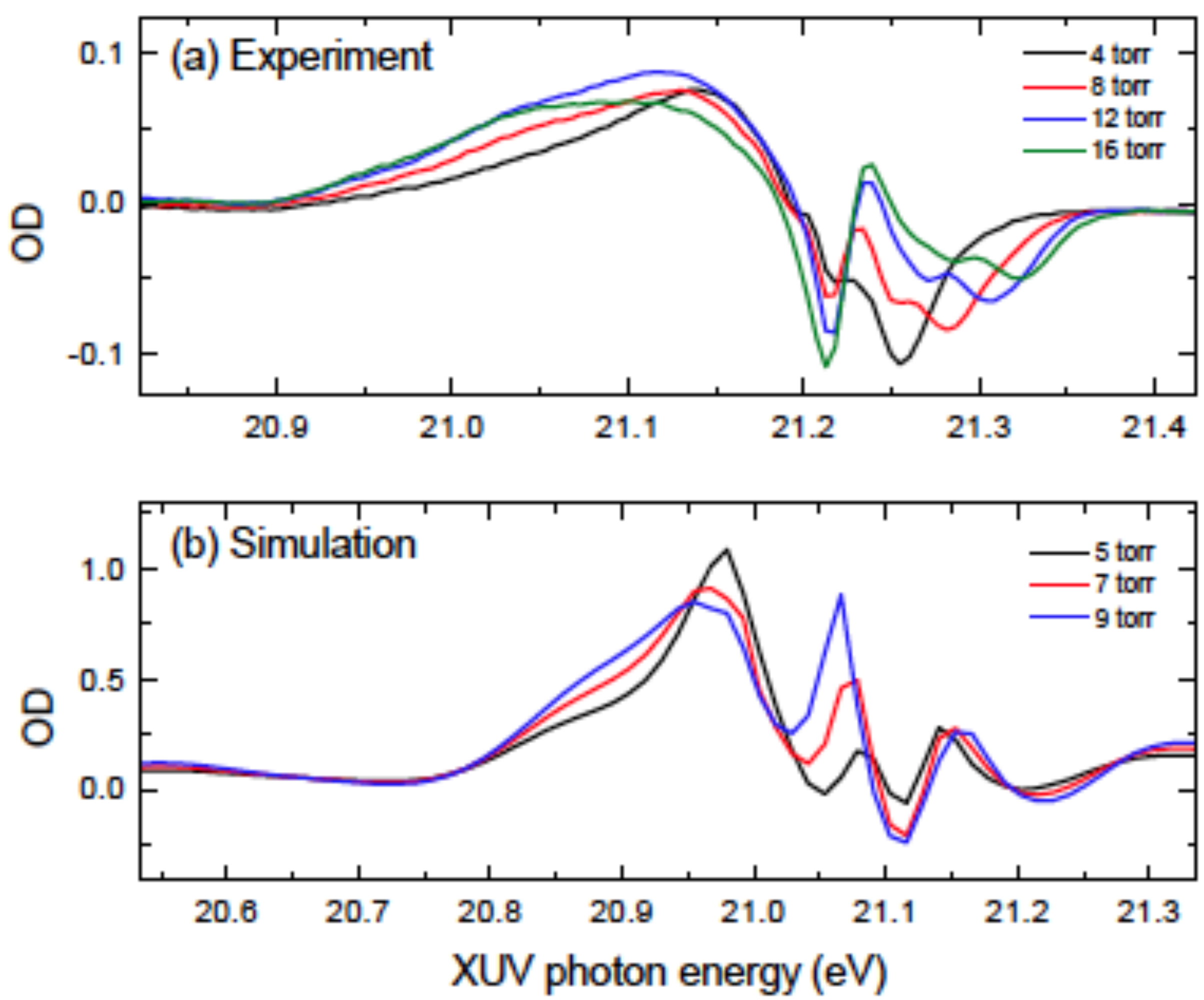
Liao et al, in prep.

Evolution of 2p line shape with pressure



Experiment and theory

XUV APT, H13 - H17 (440 as pulses)

40 fs IR 800 nm, 3e12 W/cm2

Long medium (1 cm)

Theory: fully coupled, self-consistent TDSE-MWE

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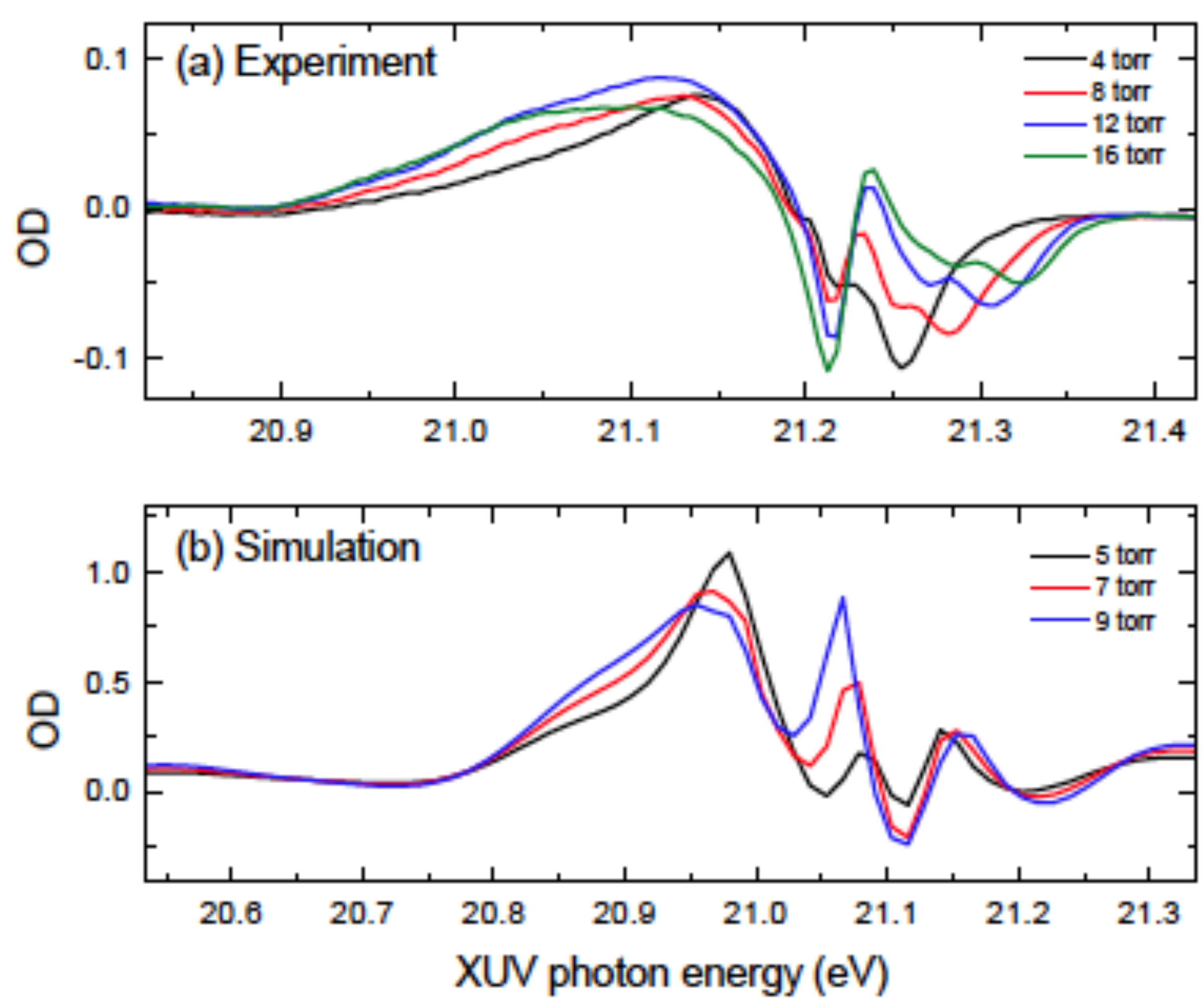

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- Appearance of new, narrow feature at center of resonance

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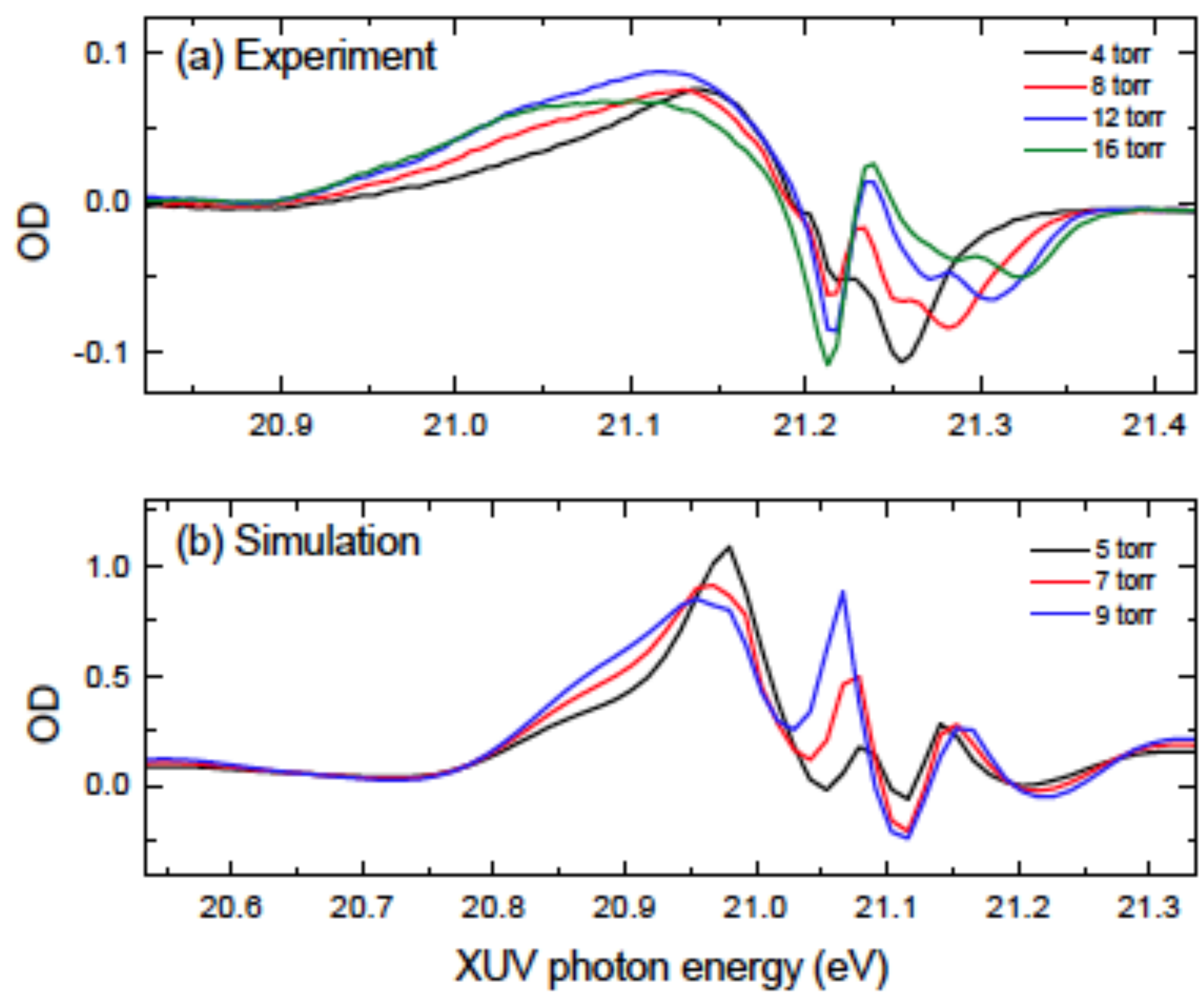

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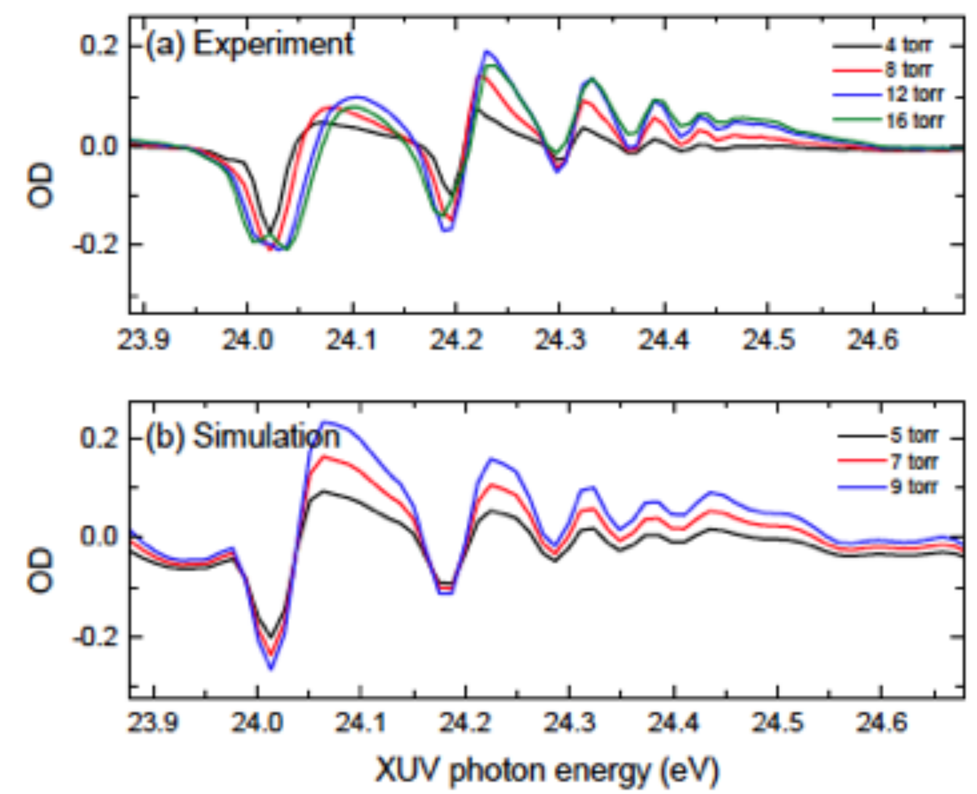
Long medium (1 cm)

Theory: fully coupled, self-consistent TDSE-MWE

np lines similar, but less extreme



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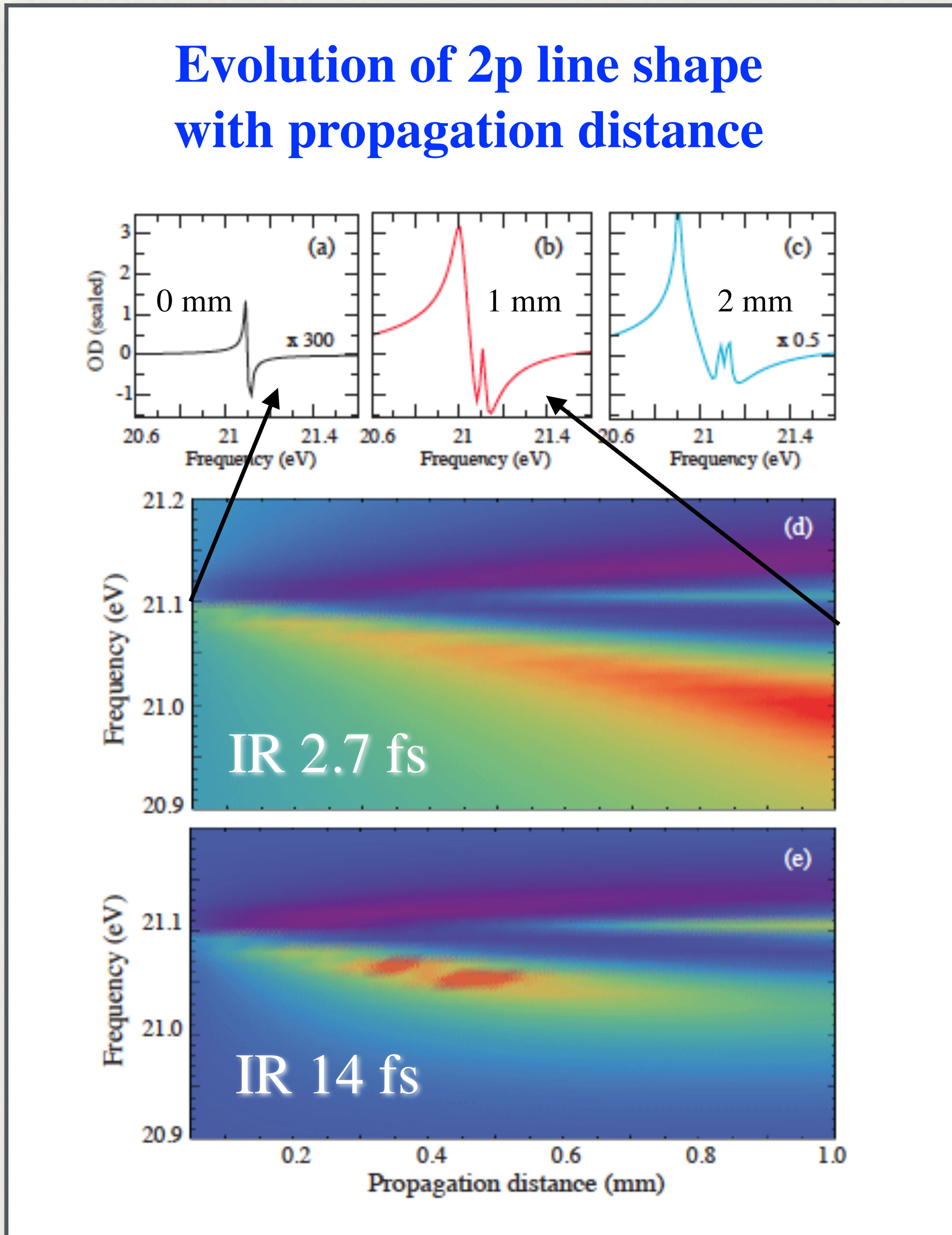
Example II: Simple model for perturbed dipole response

Liao et al, in prep.



- Time-dependent dipole moment calculated from model atom: two-level system, 1s-2p in He, resonant with XUV SAP
- IR perturbation is time-dependent phase, proportional to IR intensity
- Solve coupled TDSE-MWE with model dipole moment

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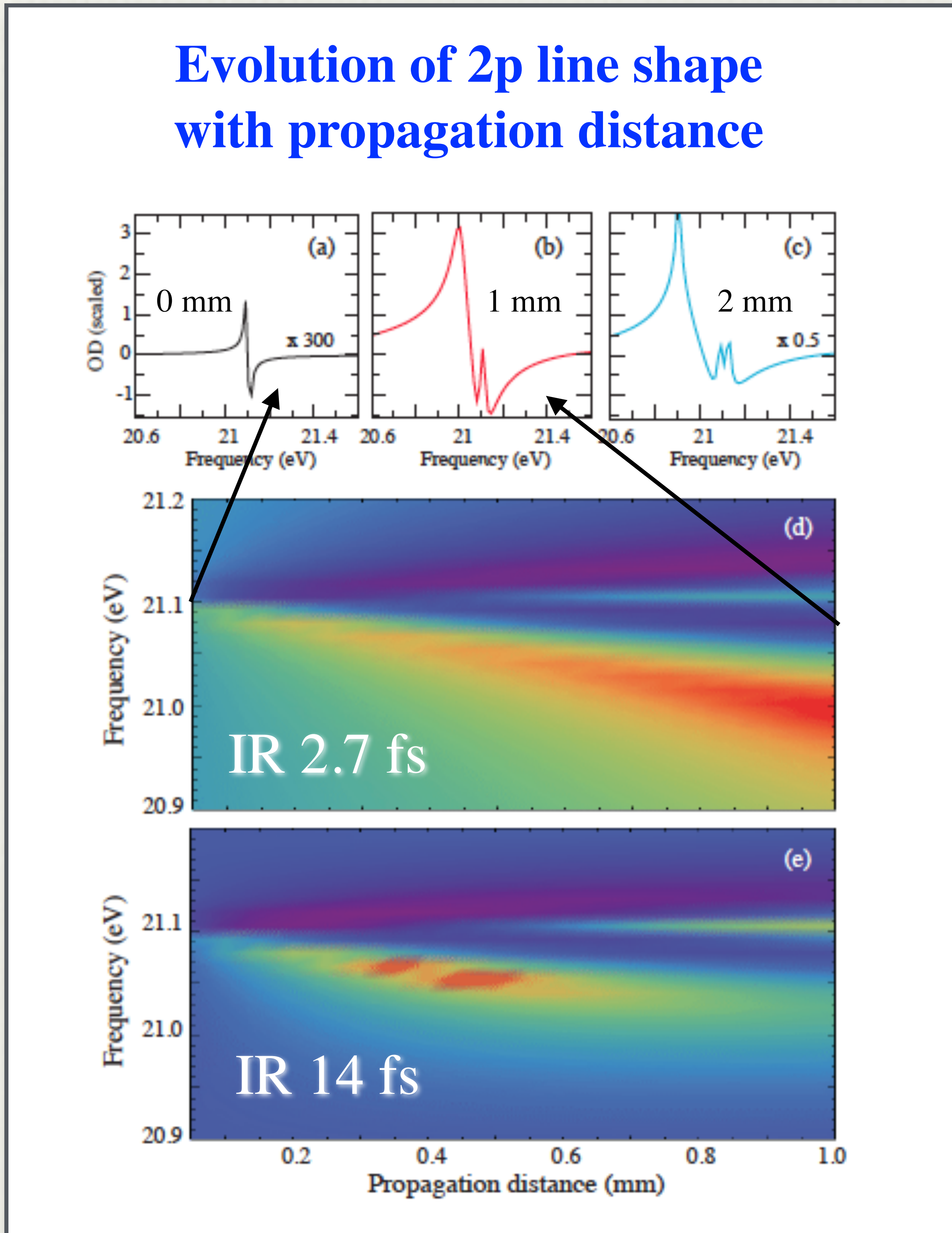

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Observations, short IR case

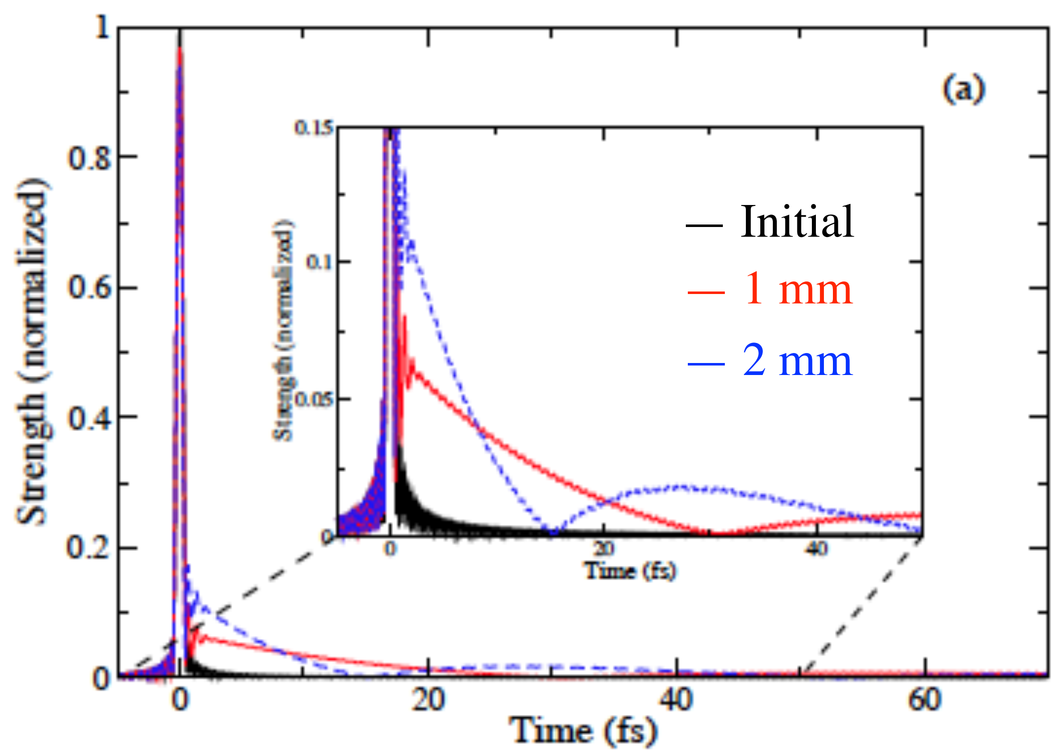
- Broadening, linear in z
- New features appear, broaden
- Shape of outer feature preserved

Example II: back to the time domain

Liao et al, in prep.

XUV time profile, end of medium (no IR)

Note: time profiles with IR almost identical



- Extra peaks in time caused by alternating sign of E-field due to absorption (well known) Crisp, PRA 1, 1970
- With longer propagation: move closer to main peak, grow in size and number

IR 2.7 fs

IR 14 fs

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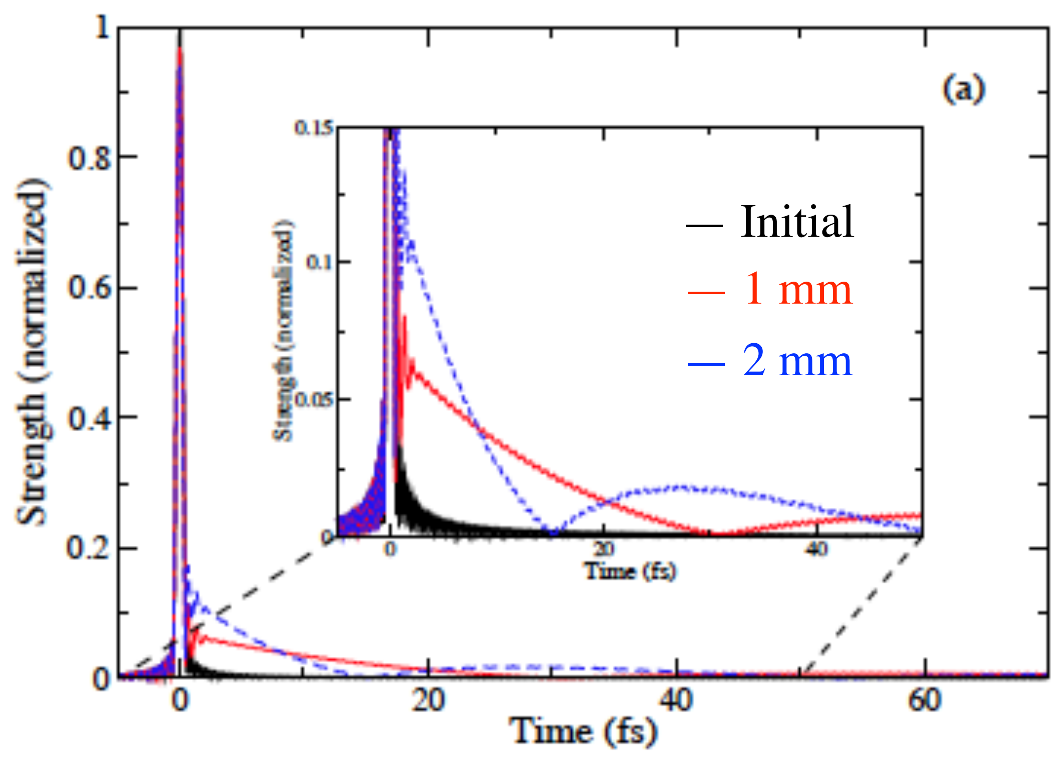
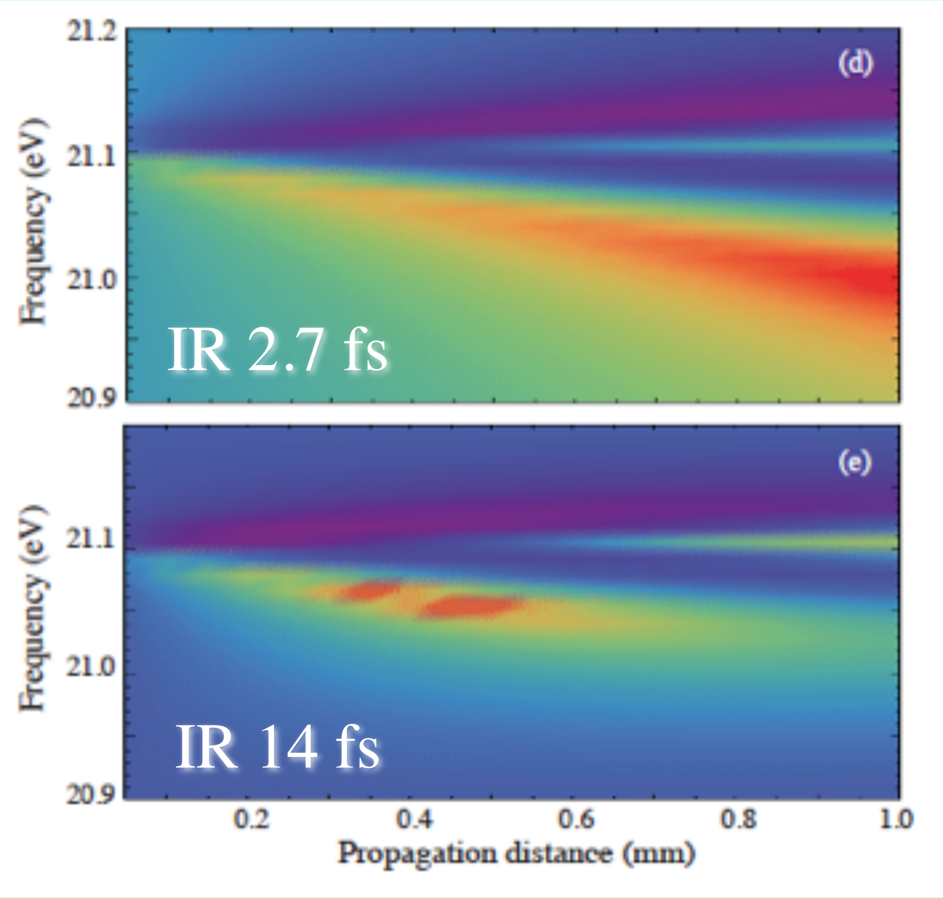


- Short IR: first extra burst is new "lifetime" over which perturbed dipole oscillates (bandwidth of outer structure)



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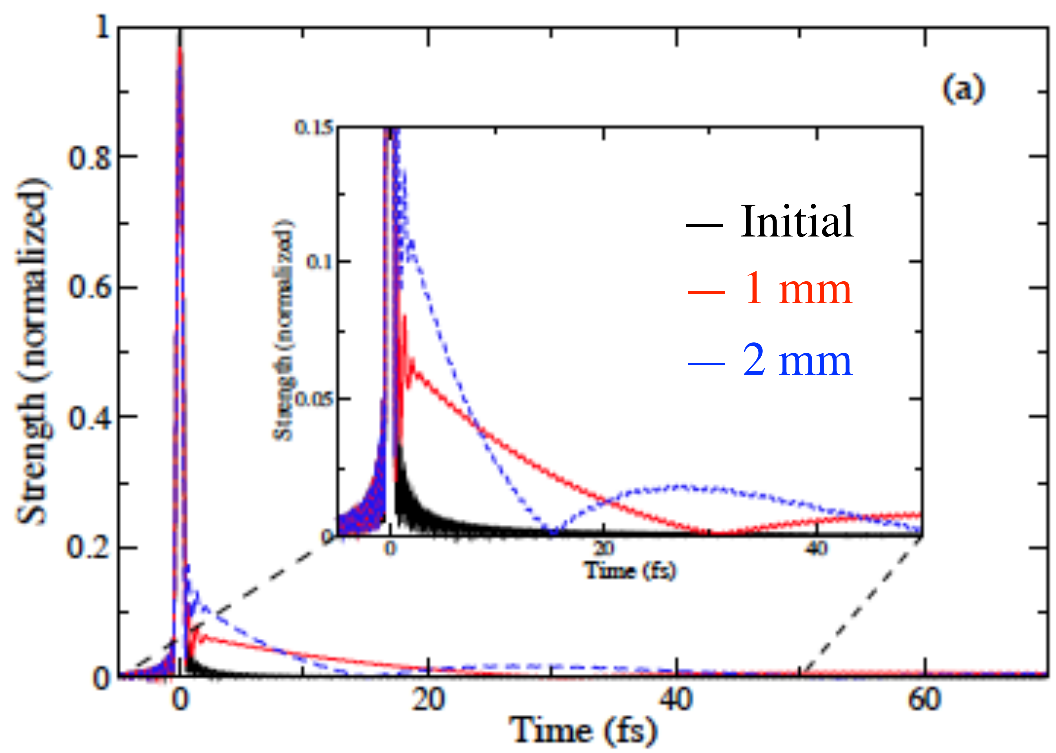

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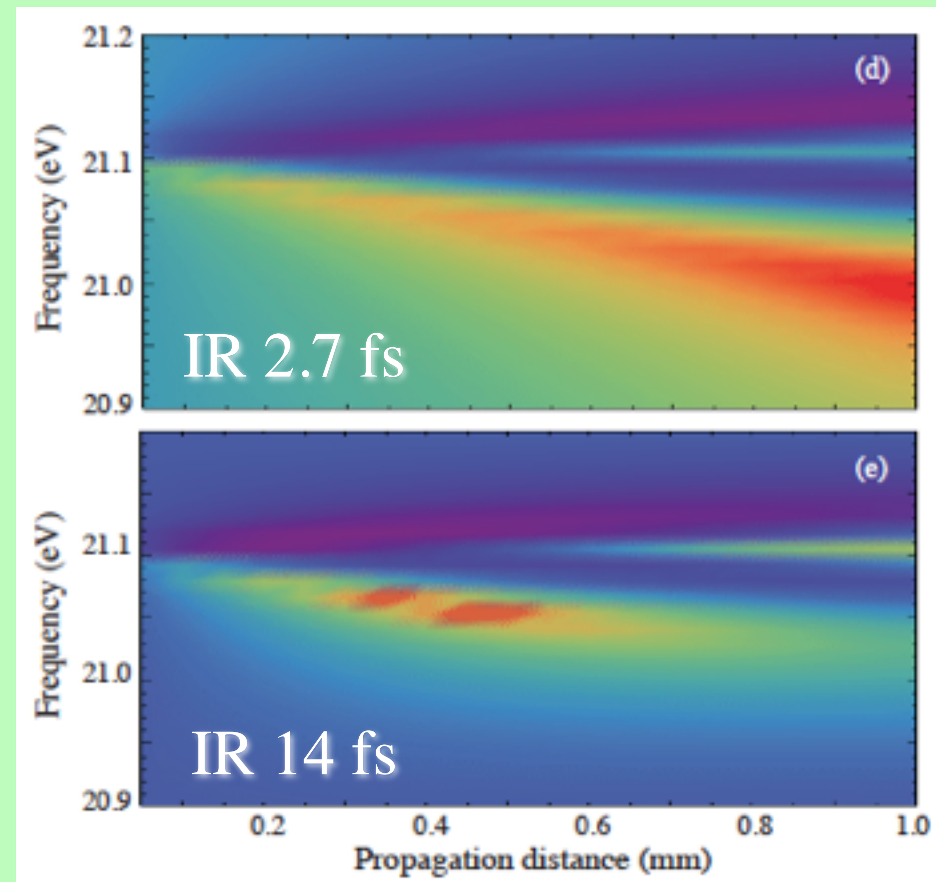
- Inner structure(s) come from additional bursts in time



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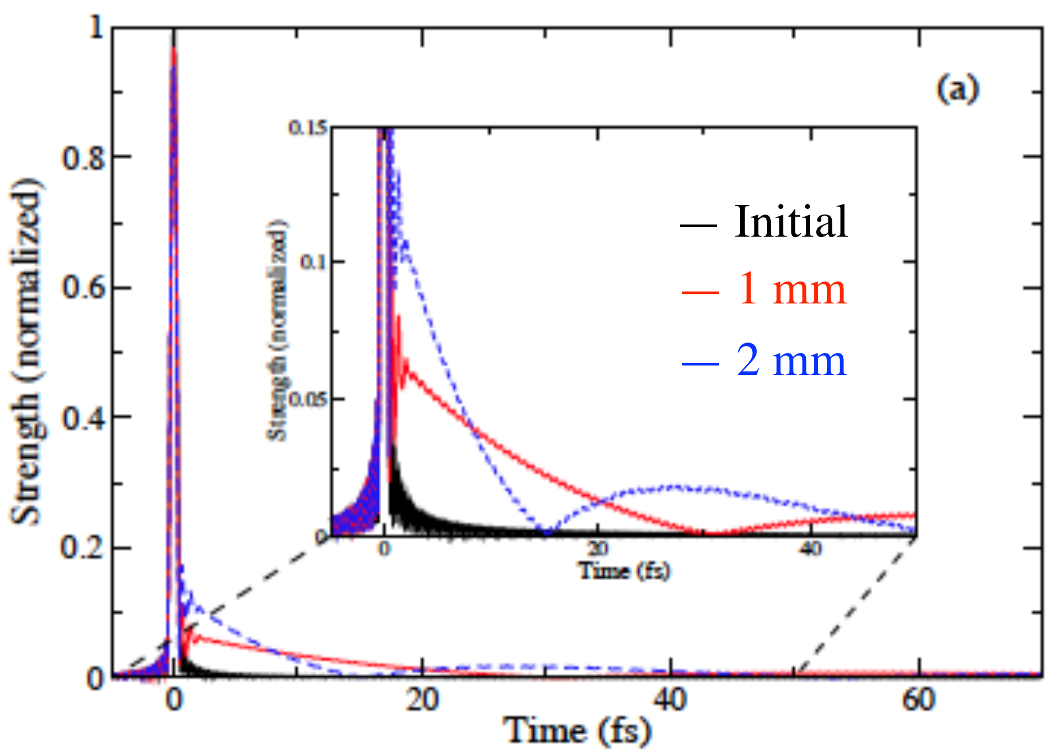
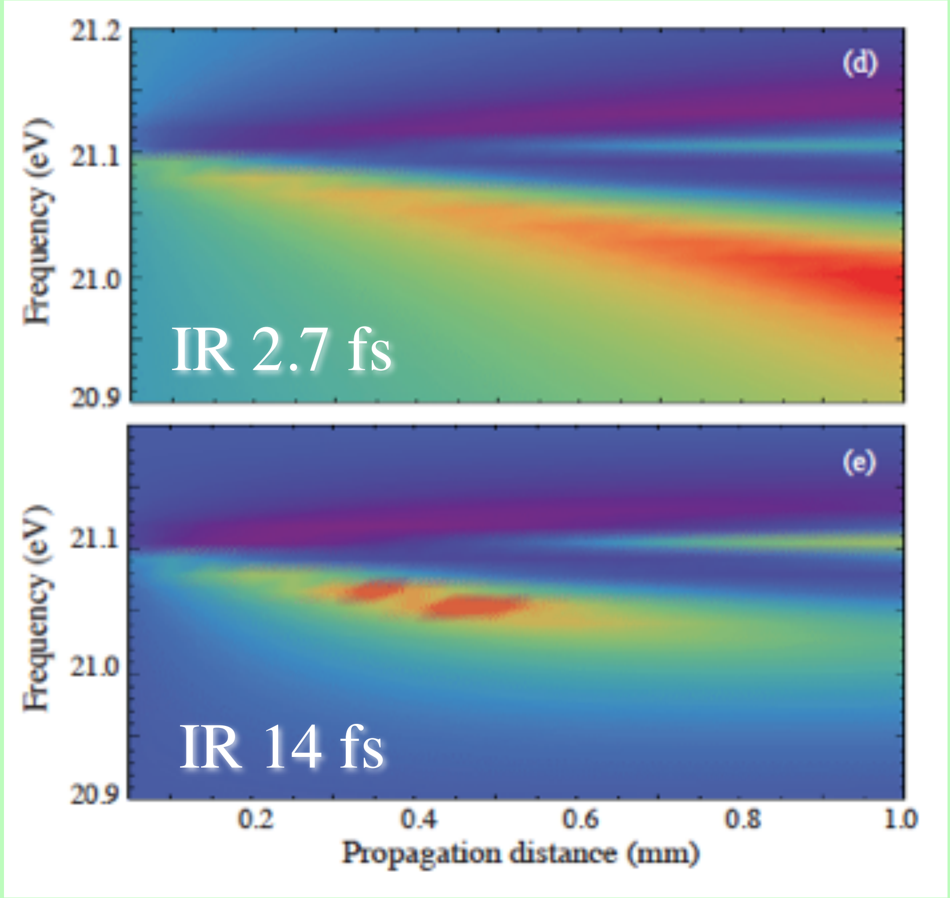


Note: more complicated when first burst becomes comparable to duration of IR perturbation:

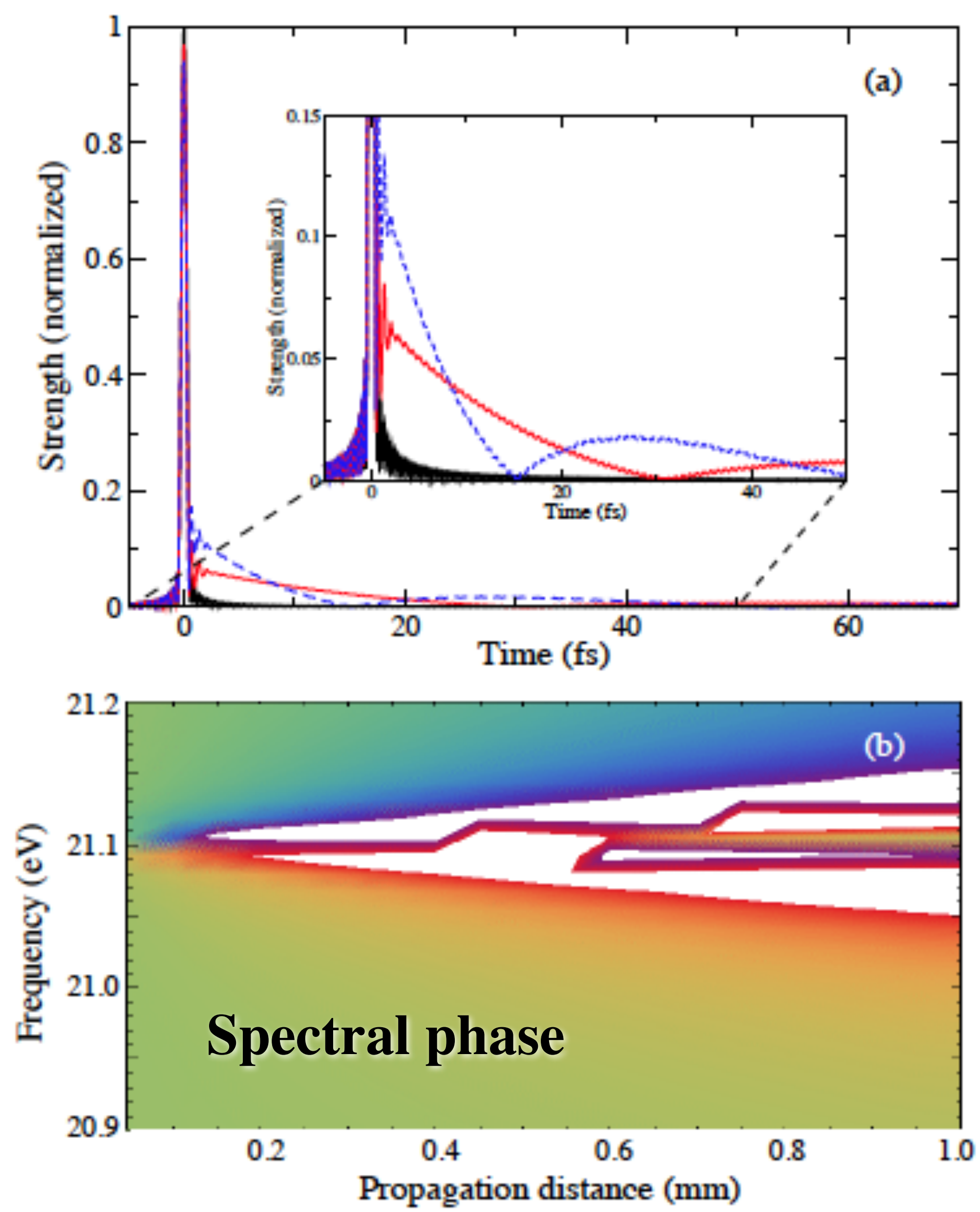
- outer lineshape not preserved
- increase in width not linear in z

- Short IR: first extra burst is new “lifetime” over which perturbed dipole oscillates (bandwidth of outer structure)
- Inner structure(s) come from additional bursts in time

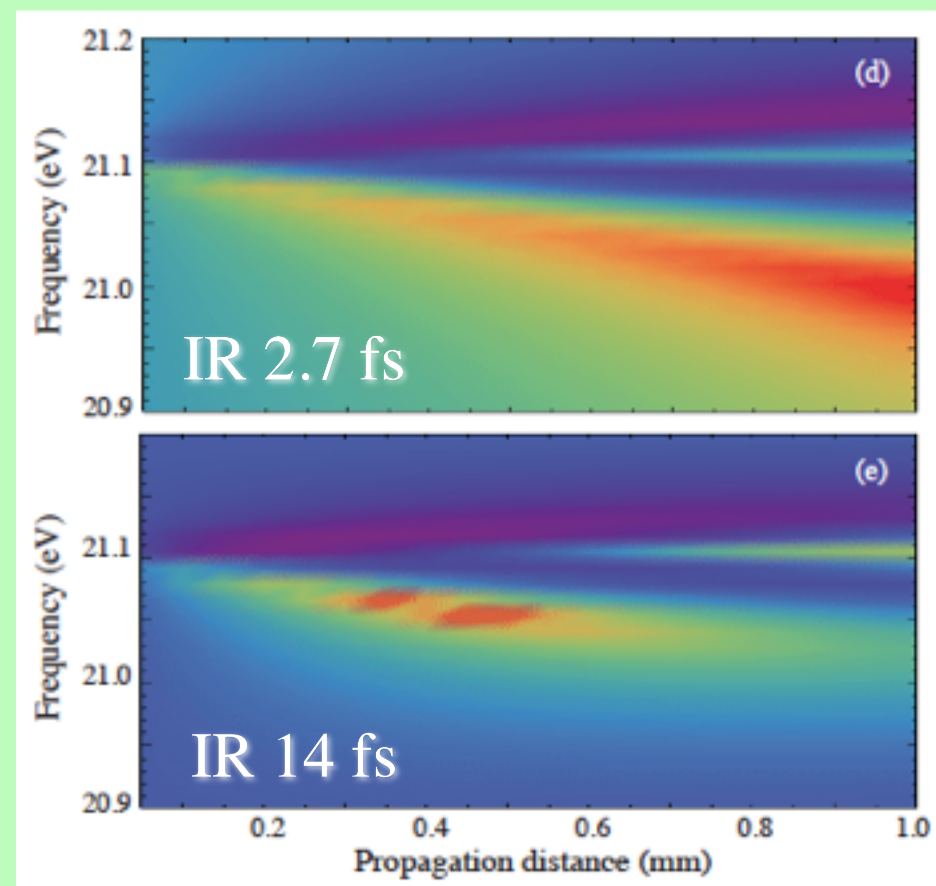


Example II: frequency domain thoughts

Spectral phase accumulated due to dispersion



- Bandwidth of ±π/2 controls duration of first extra peak - causes sign change of E-field
- In absence of IR only broadening through saturation



Summary

- TDSE-MWE, self-consistent solution allows to study reshaping of light by medium
- Argon Cooper min induces reshaping of attosecond pulses even at single atom level
- Reshaping - effects of attosecond pulse propagation in dense medium with narrow absorption lines, dressed by IR pulse

LSU Attosecond th. group
Ken Schafer
Ken Lopata (Chemistry)
Mengxi Wu
Seth Camp
Paul Abanador
Xiaoxu Guan
Renate Pazourek
Shaohao Chen (now CCT)

Ohio State exp. group
Lou DiMauro
Pierre Agostini
Stephen Schoun
Razvan Chirla
Jonathan Wheeler
Christoph Roedig

U Arizona exp. group
Arvinder Sandhu
Chen-Ting Liao

Funding
NSF, DOE BES, LONI (computing time)

Two post doc positions at LSU, starting immediately

- One postdoc in physics (Schafer/Gaarde)
- One postdoc in chemistry (Lopata)

Project: Charge migration using HHS?
Collaboration physics/chemistry, exps at OSU/UVA