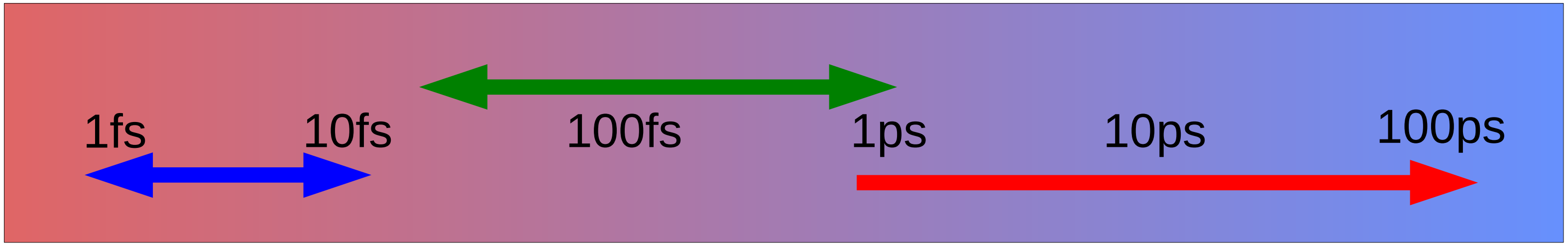
Light-matter interactions at ultra-fast time-scales and high intensities: modeling and simulation perspective

OPTI-583 Lecture Notes

Light-matter interaction models in optical filamentation

Models based on experience
from longer time-scales:

“traditional” filament modeling



first few fs

plasma formation, THz, ...

Need to return to first principles,
Ideally:
Maxwell + Schrodinger

From free electrons to collective effects...

- How exactly does this happen?
- What about many-bod effects?

Optical pulse propagation

$$\partial_z E_{k_x,k_y}(\omega,z) = ik_z E_{k_x,k_y}(\omega,z) + \frac{i\omega^2}{2\epsilon_0 c^2 k_z} P_{k_x,k_y}(\omega,z) - \frac{\omega}{2\epsilon_0 c^2 k_z} j_{k_x,k_y}(\omega,z)$$

$$k_z = \sqrt{\omega^2 \epsilon(\omega)/c^2 - k_x^2 - k_y^2}$$

Not computable, but available...
You want this separated from the rest of medium properties

Can we obtain this from QM,
without doing (much of) QM?

The “standard” model for light-matter interactions in extreme nonlinear optics

Optical pulse propagation

$$\partial_z E_{k_x,k_y}(\omega, z) = i k_z E_{k_x,k_y}(\omega, z) + \frac{i\omega^2}{2\epsilon_0 c^2 k_z} P_{k_x,k_y}(\omega, z) - \frac{\omega}{2\epsilon_0 c^2 k_z} j_{k_x,k_y}(\omega, z)$$

$$k_z = \sqrt{\omega^2 \epsilon(\omega)/c^2 - k_x^2 - k_y^2}$$

Medium response:
(short time)

$$\vec{P}(\vec{r}, \vec{E}(\vec{r}, t)) \quad \text{and/or} \quad \vec{J}(\vec{r}, \vec{E}(\vec{r}, t))$$

$$\vec{P} = \epsilon_0 \bar{n}_2 E^2 \vec{E}$$

Kerr and third harmonic generation

$$\frac{\partial \rho}{\partial t} = W(|\mathcal{E}|^2)(\rho_{at} - \rho)$$

ionization

$$\frac{\partial \mathbf{J}}{\partial t} + \frac{\mathbf{J}}{\tau_c} = \frac{q_e^2}{m_e} \rho \mathbf{E}$$

freed-electron current evolution

$$\frac{1}{2} \mathbf{J} \cdot \mathbf{E} = W(|\mathcal{E}|^2) K \hbar \omega_0 \rho_{nt}$$

additional current to account for loss

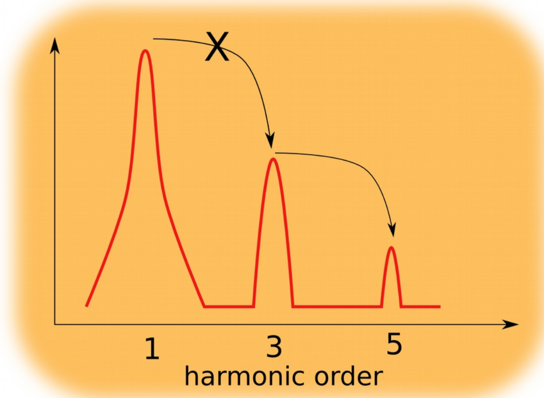
The "standard" model for light-matter interactions in extreme nonlinear optics

Medium response:
(beyond standard?)

Higher-Order Kerr Effect (HOKE):

$$\Delta n(I) = n_2 I + n_4 I^2 + n_6 I^3 + n_8 I^4 + n_{10} I^5$$

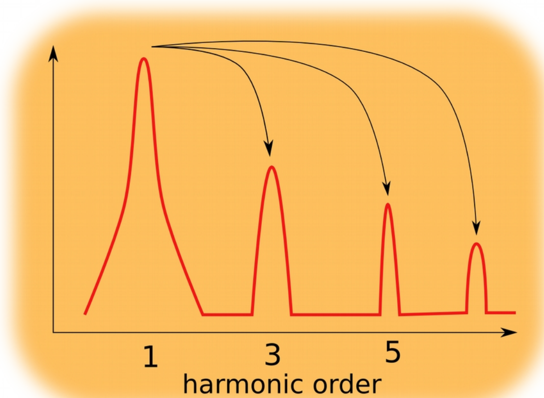
... no third harmonic



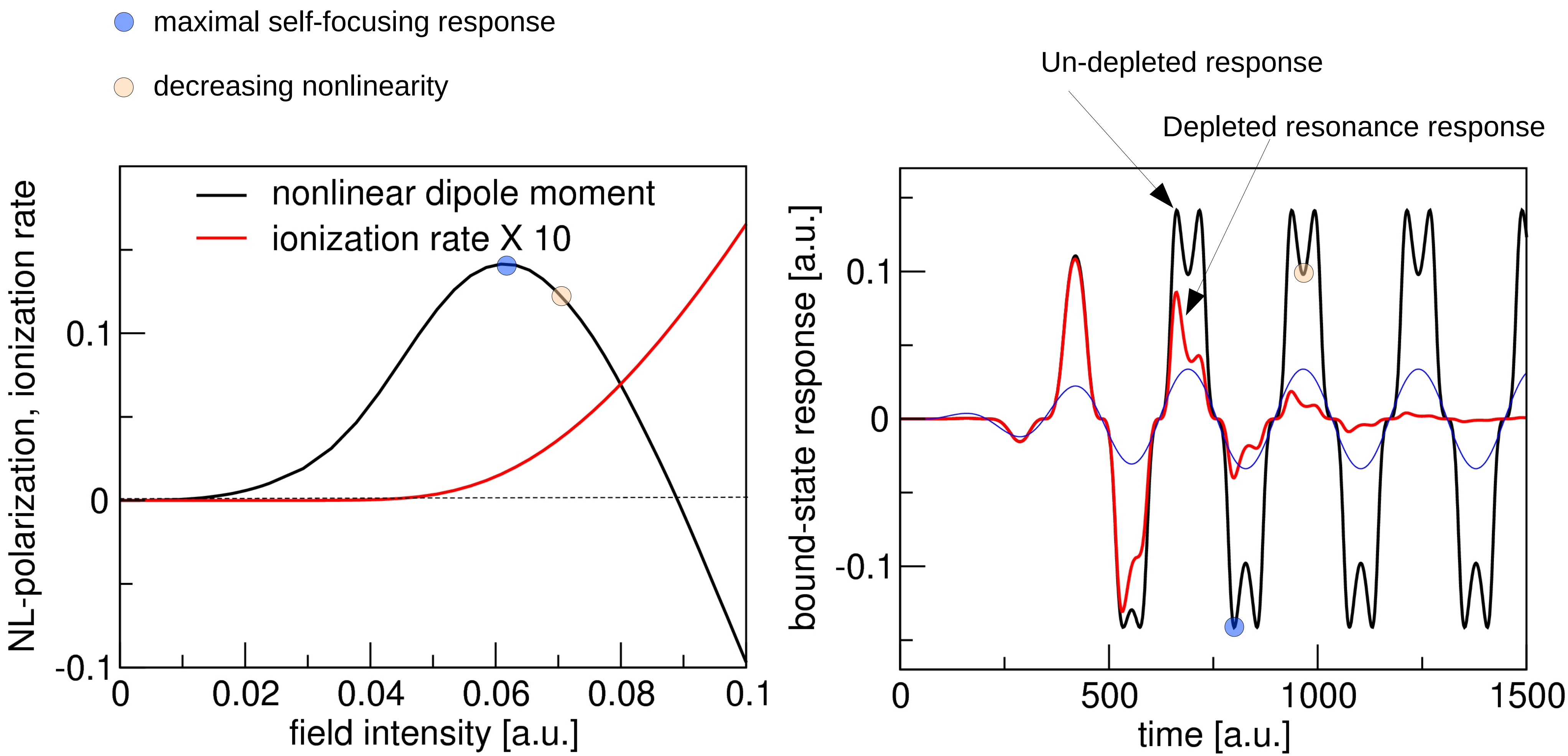
.. or perhaps this:

$$\Delta P(t) \approx \Delta n(E^2)E = \left[\tilde{n}_2(E^2) + \tilde{n}_4(E^2)^2 + \tilde{n}_6(E^2)^3 + \tilde{n}_8(E^2)^4 + \tilde{n}_{10}(E^2)^5\right] E(t)$$

... too many harmonics?



Since clearly neither of the two shown here is correct, what is the proper description of electronic nonlinearity?



Ground-state depletion effectively suppresses higher-order nonlinear signatures

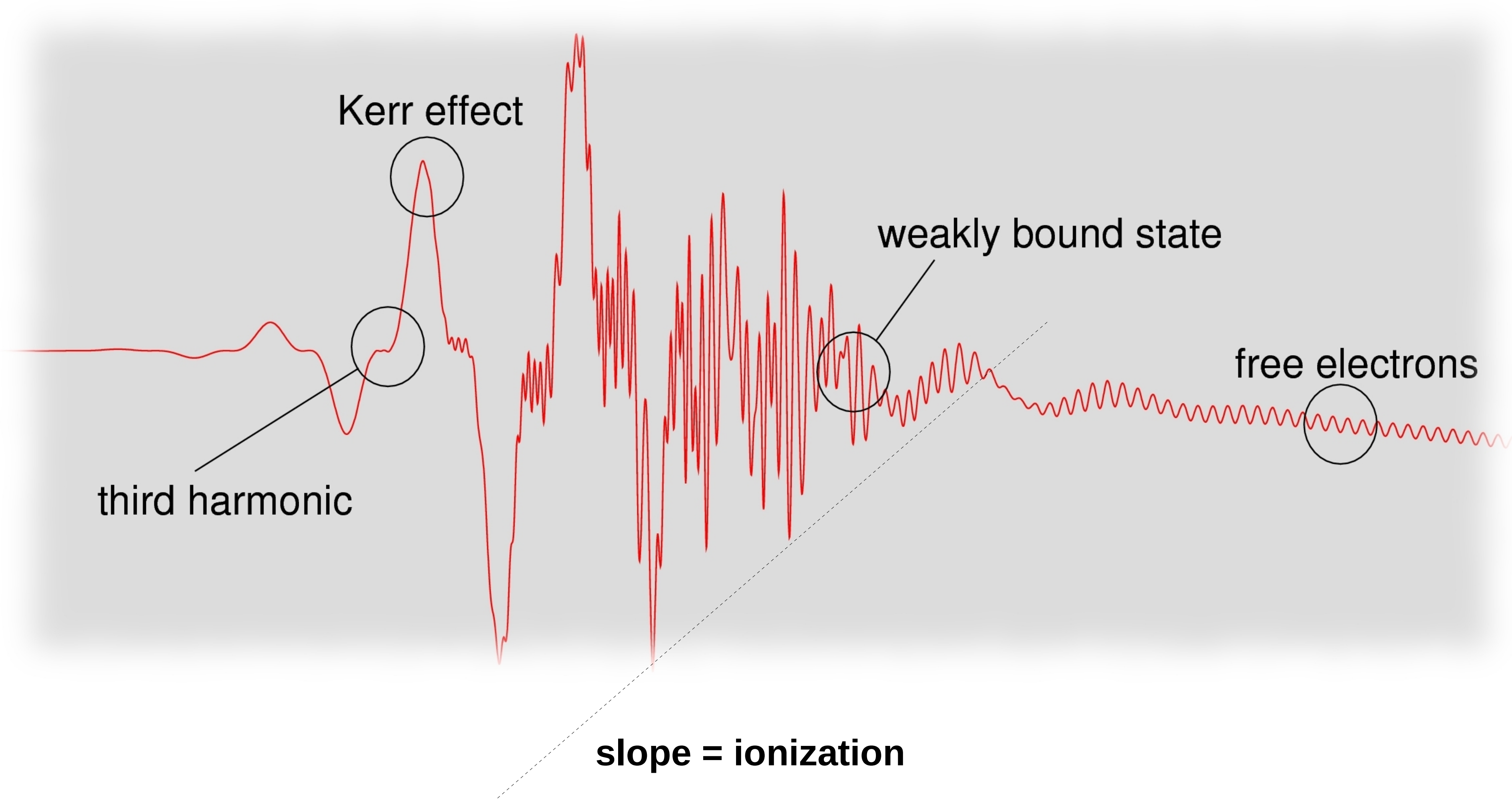
The standard model: Conceptual deficiencies, problems

- It is a phenomenological sum of uncorrelated parts
- electrons either bound (Kerr) or free (Drude plasma)
- weakly bound states ignored
- excited states are ignored

... which are important for intensities characteristic of many self-organized regimes

- free-electrons:
  linear or nonlinear? Isotropic?
  what is the meaning of 'collision time'?
- ionization:
  expressed as ionization *rate* ... which means there is no dependence on history!
  single frequency, long pulse regime
  causes no losses!
  causes no phase change!
- higher-order nonlinearity: not clear if susceptibility language is appropriate

“Reading” non-linear response: Effect signatures



Examples of regimes where simple models do not work

- Memory effects in ultrafast-excited ionization
- Exotic pulses, with extreme chirp and intensity spikes
- Anisotropic response to probing during and after excitation with strong pulses
- Multi-color and carrier-enginered filamentation

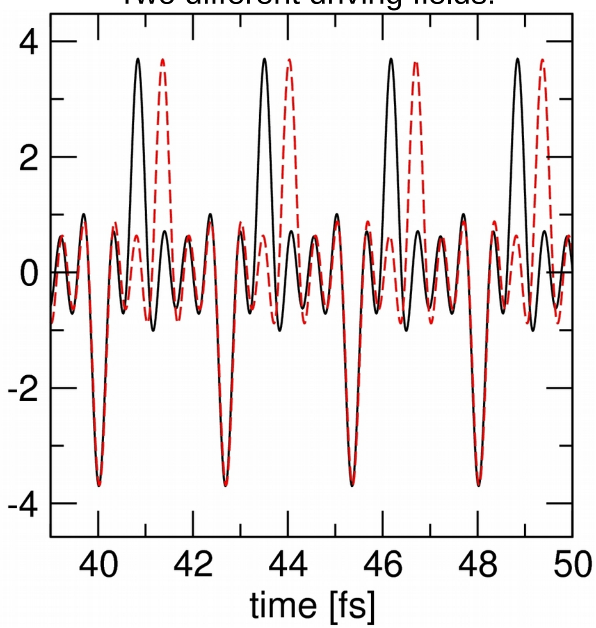
EX 1: Memory effects in strong-field ionization: When the yield depends on timing

Q: Modeling Ionization in term of rate implies no memory. Where is the limit?

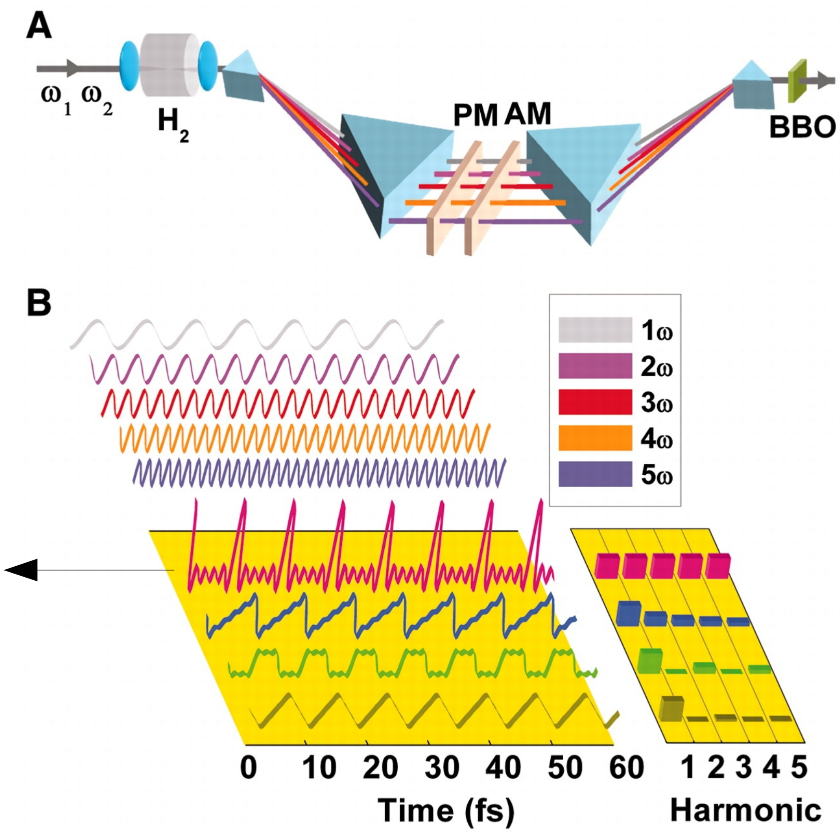
A: Synthesized Pulse Trains

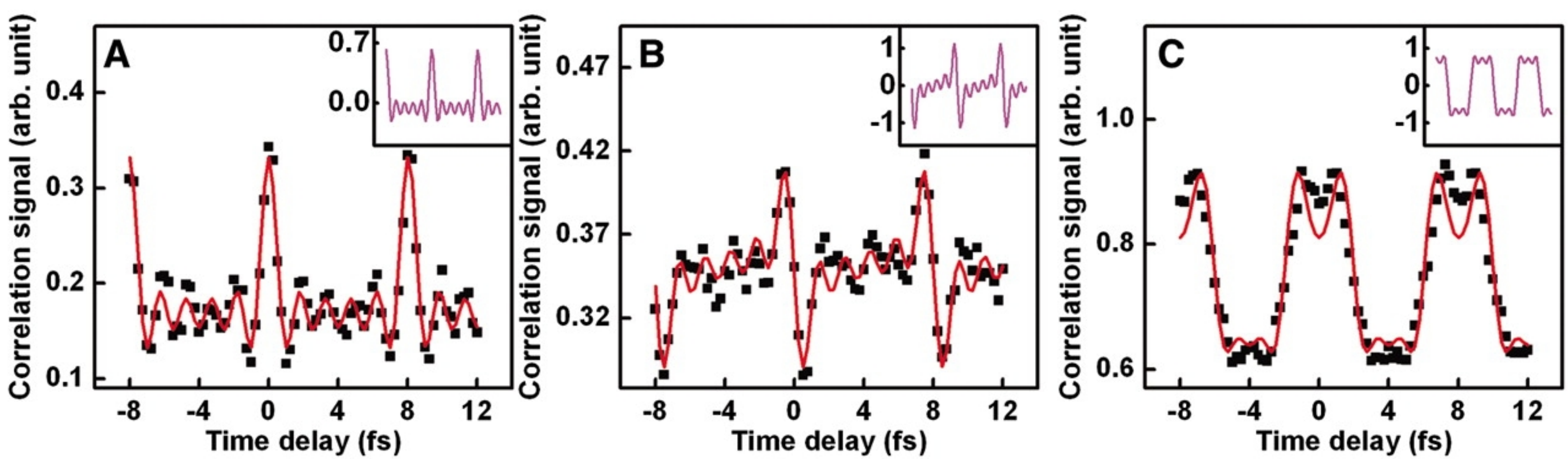
Han-Sung Chan et al., Science 331, 1165 (2011):

Two different driving fields:



flip+delay

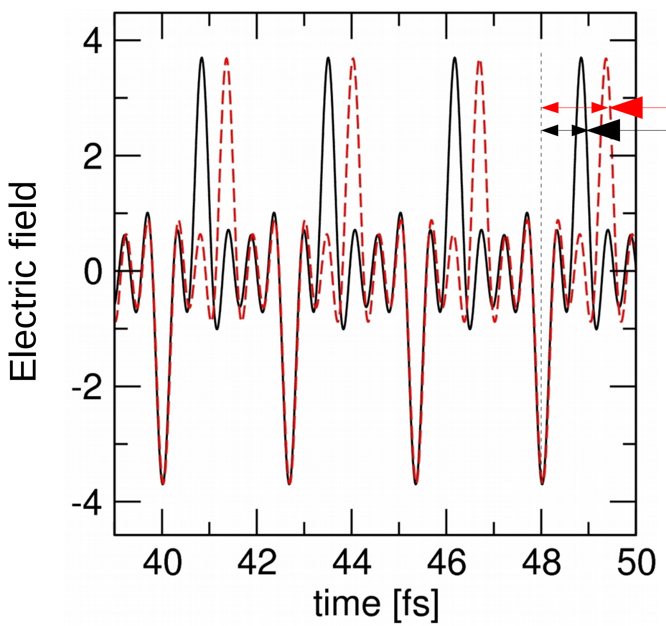




EX 1: Memory effects in strong-field ionization: When yield depends on timing

EXCITATION

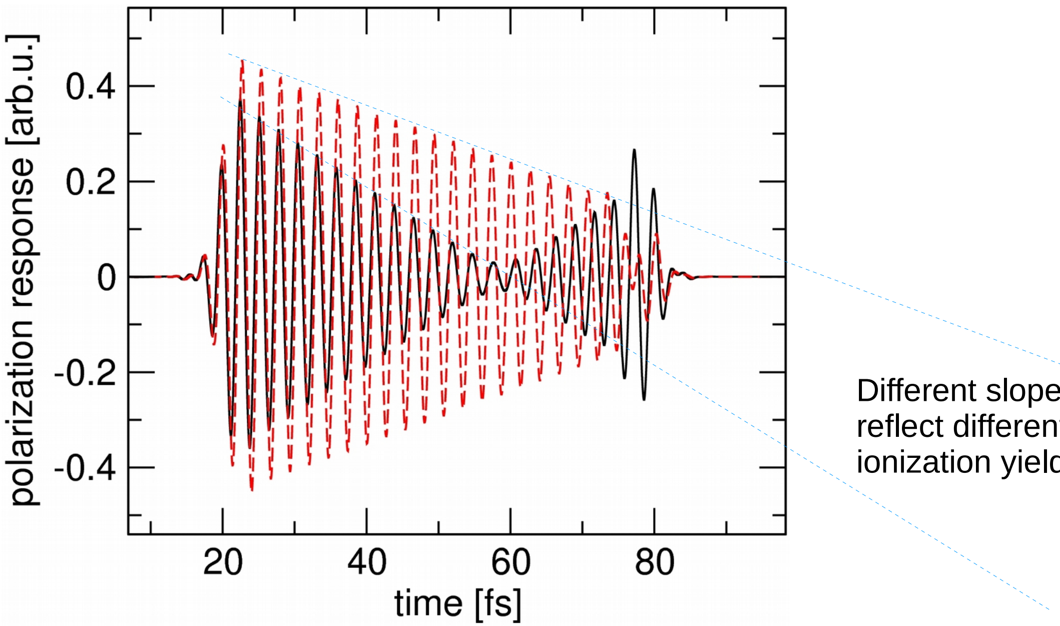
5 harmonics were mixed, to synthesize two pulse trains:
different time separation between peaks of different polarity



Current model says:
These two electric field waveforms are equivalent -
Ionization does not depend on time delay between positive and negative pulses

NONLINEAR RESPONSE

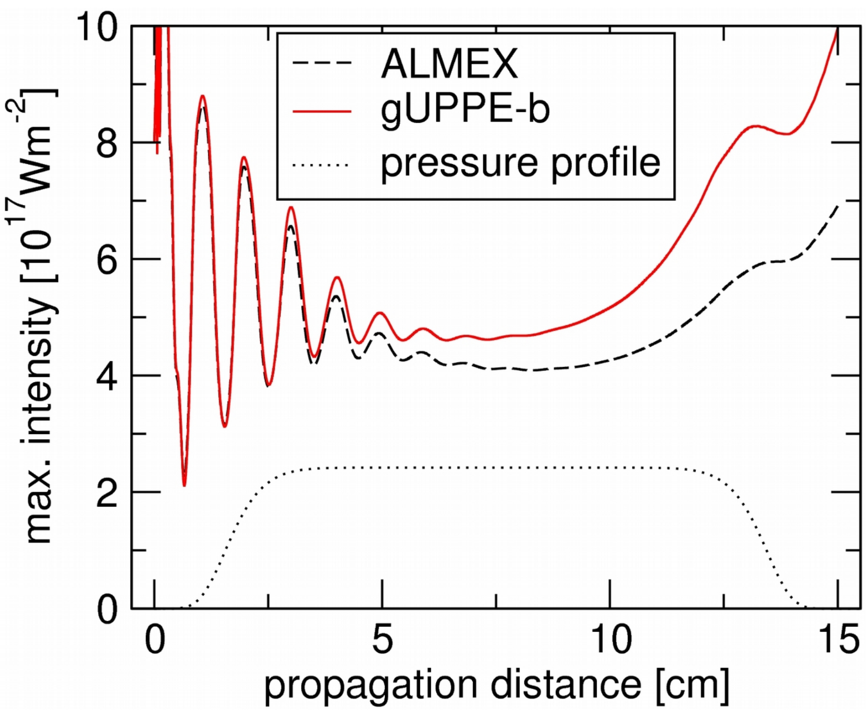
Exact QM solution:
Different nonlinear responses in the two pulse trains

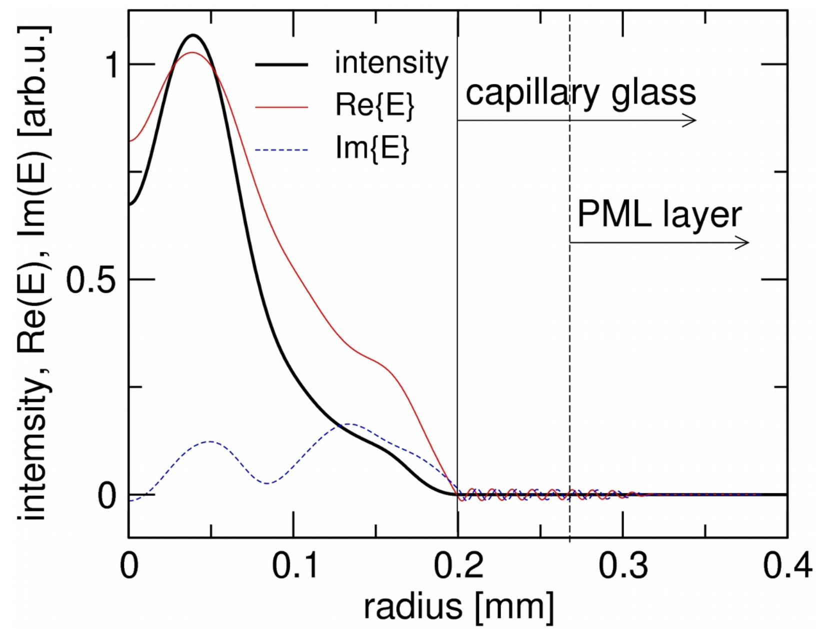


Mechanism???
Coherent excitation of
Higher-energy resonance states

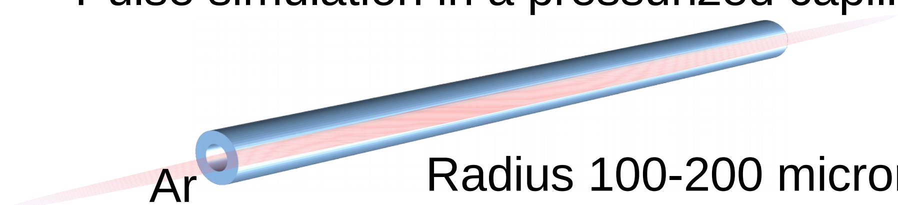
First-principle informed, quantum solution:
reveals that history of the system driven by a strong field matters:
Ionization is stronger if subsequent pulses “hit” fast

EX 2: Extreme waveforms in mid-infrared pulses in leaky waveguides

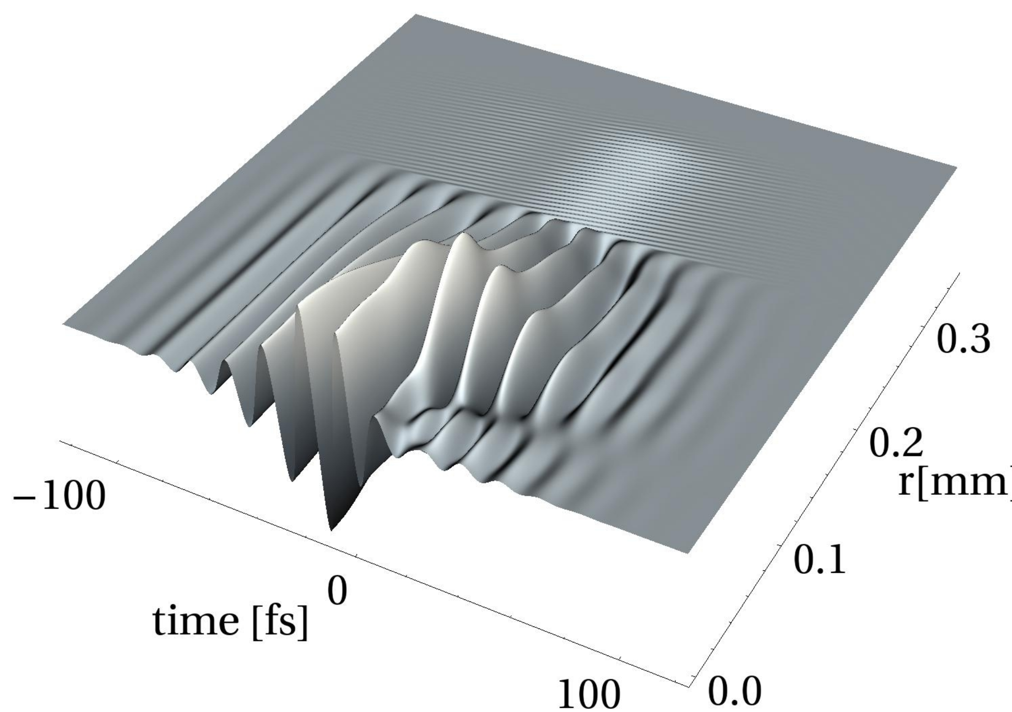




Pulse simulation in a pressurized capillary

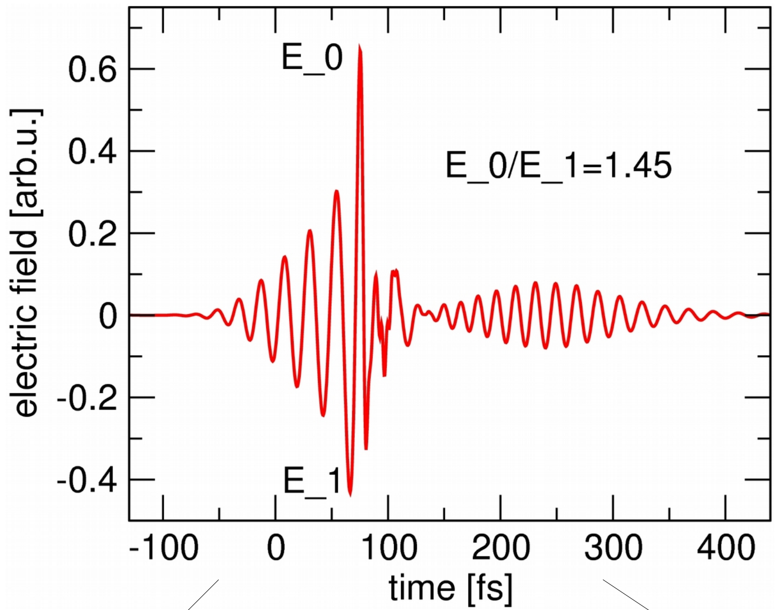


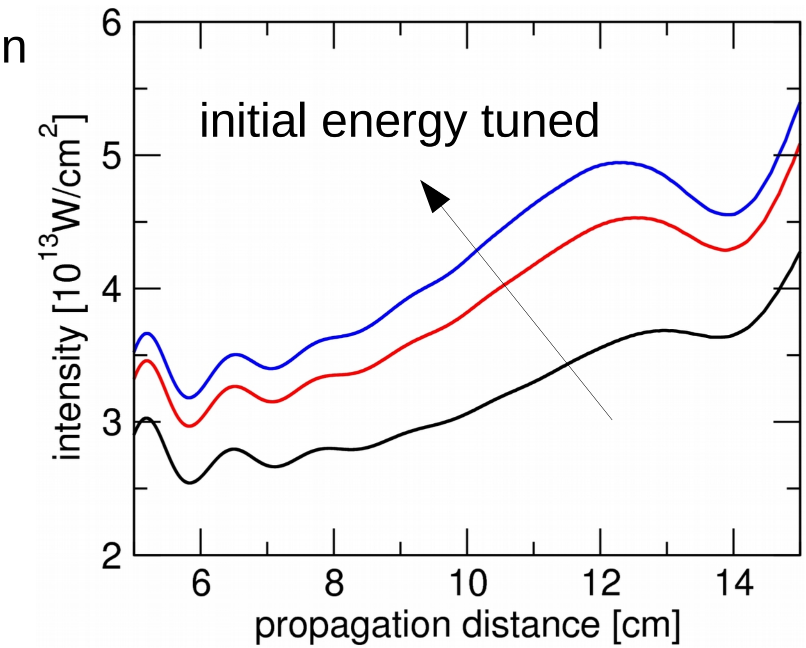
Radius 100-200 micron
Pressure 10-20 atm
Wavelengths 4-8 micron

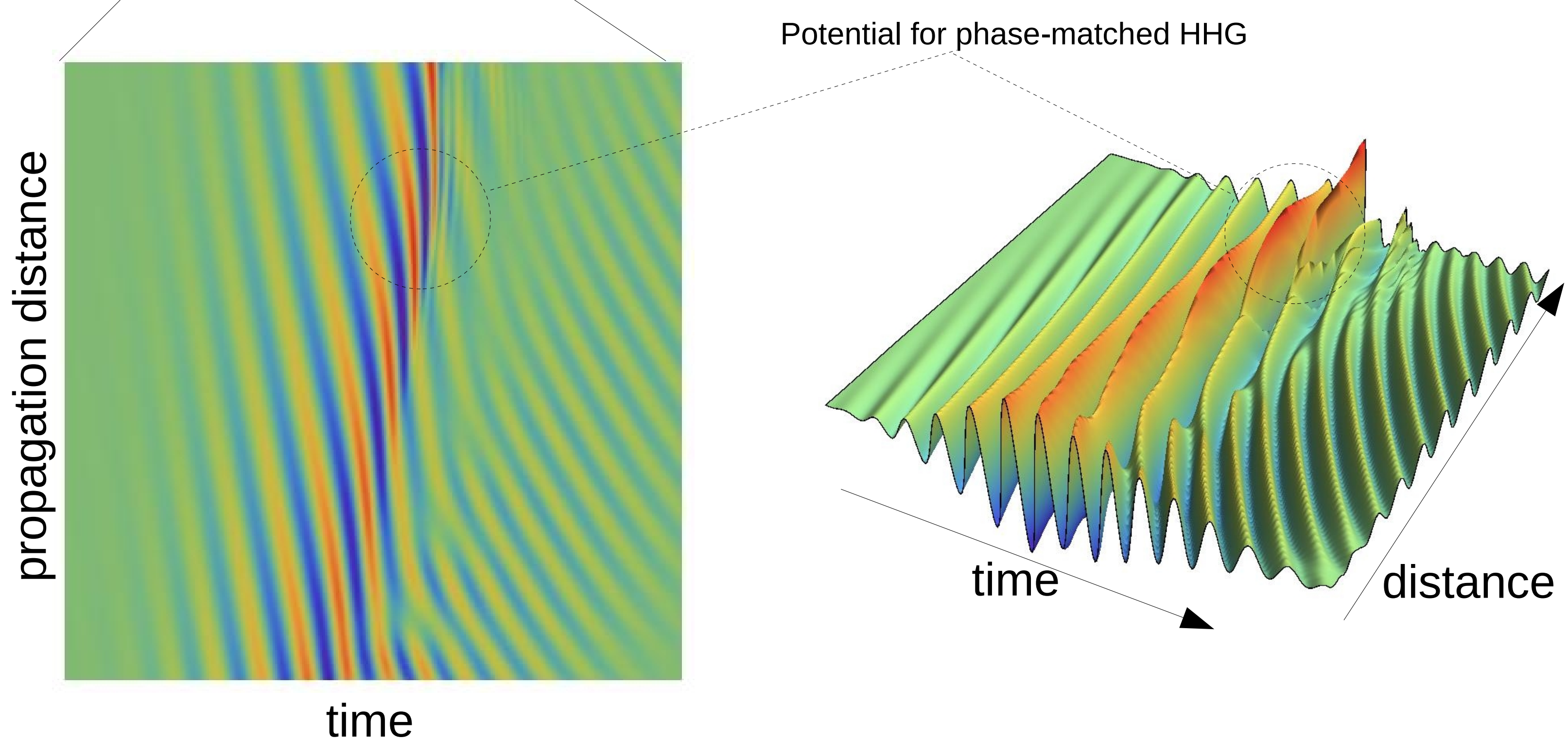


EX 2: Few-cycle mid-infrared waveforms generated in pressurized capillaries

“half-cycle” waveform from a 100fs pulse at 6 micron

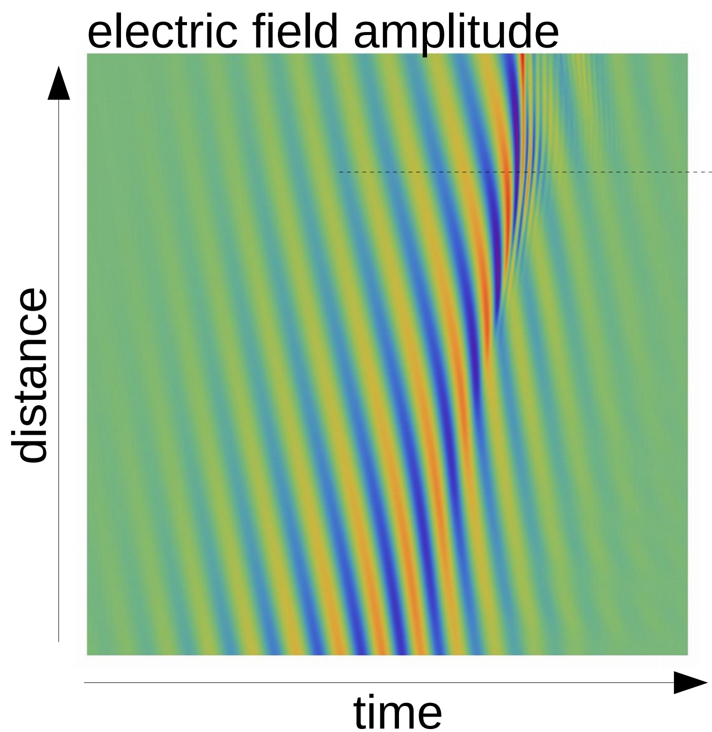


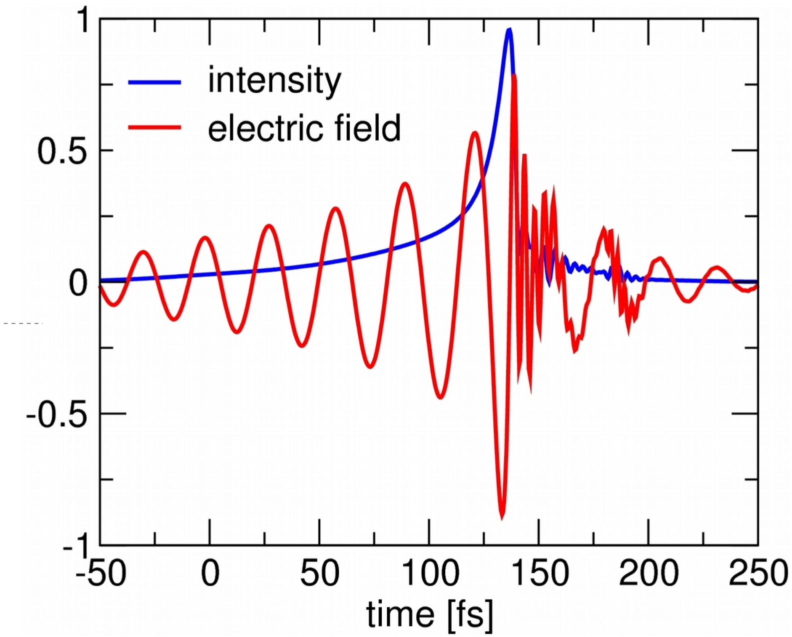


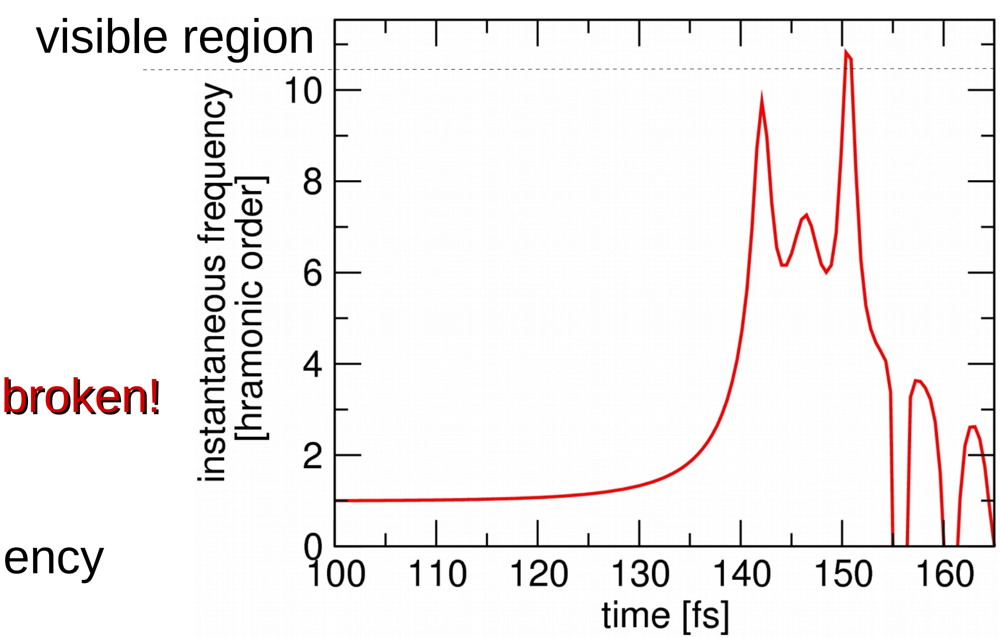


EX 2: Few-cycle mid-infrared waveforms generated in pressurized capillaries

Extreme chirp from a 100fs pulse at 8 micron



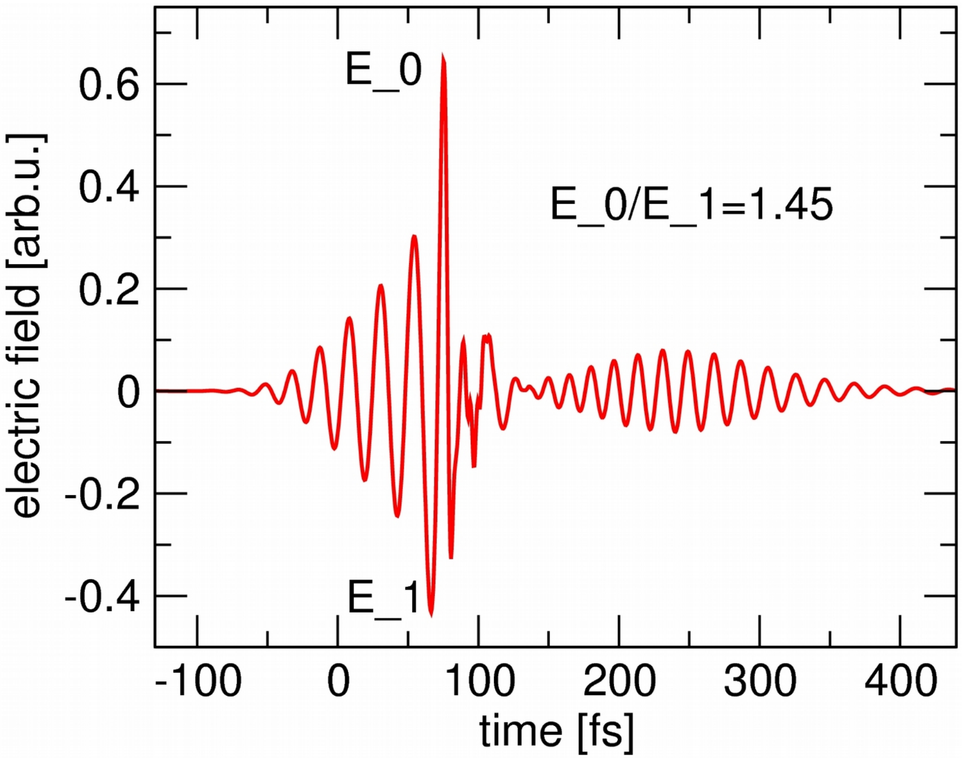




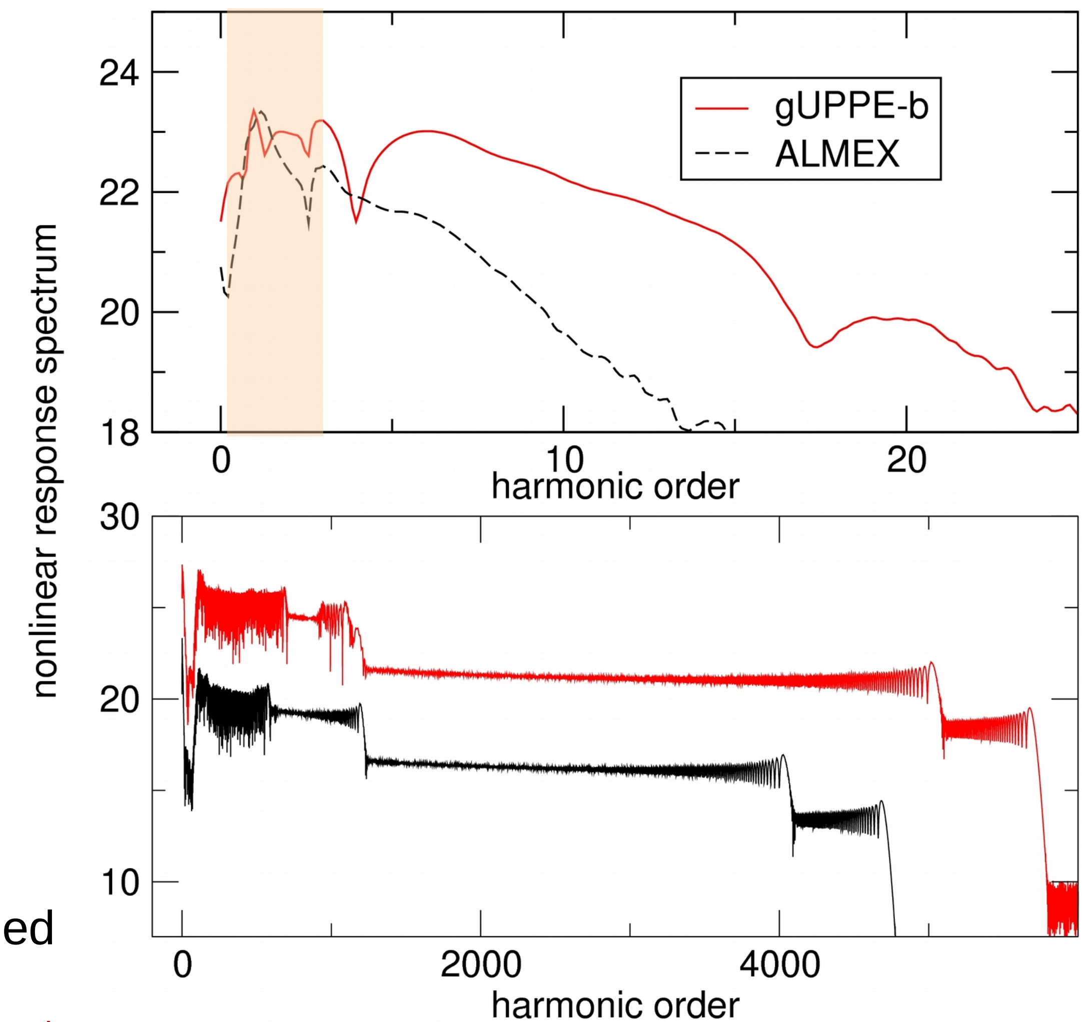
“Every single” (model-underlying) assumption broken!

- Non-dispersive Kerr nonlinearity
- Ionization rate calculated for a specific frequency

Self-consistent (all-in-one) models have their own problems



Only a tiny portion of the response spectrum affects propagation (or feeds back into) of the driving pulse:



Nonlinear response spectra calculated for an exactly solvable model

Lesson learned:

- Self-consistent model alone is not all we need
- Ability to calculate only the low-frequency part of its nonlinear response is important

First-principle based modeling of light-matter interaction

- MASP – integrated Maxwell Schroedinger system simulation
- Extraction of nonlinear susceptibilities from TDSE simulations
- Analytic approaches (toy models an approximations)
- From ionization to plasma formation: Maxwell-Bloch equations

Maxwell–Schrödinger–Plasma (MASP) model for laser–molecule interactions: Towards an understanding of filamentation with intense ultrashort pulses

E. Lorin[a,c,*], S. Chelkowski[b], E. Zaoui[a], A. Bandrauk[a,b]

Physica D 241 (2012) 1059-1071

Full Maxwel system,
Driven by microscopic polarization and current

$$\begin{aligned}
&\partial_t \mathbf{B}(\mathbf{r},t) = -c\nabla \times \mathbf{E}(\mathbf{r},t),\\
&\partial_t \mathbf{E}(\mathbf{r},t) = c\nabla \times \mathbf{B}(\mathbf{r},t) - 4\pi\big(\partial_t \mathbf{P}(\mathbf{r},t) + \mathbf{J}(\mathbf{r},t)\big),\\
&\nabla \cdot \mathbf{B}(\mathbf{r},t) = 0,\\
&\nabla \cdot \big(\mathbf{E}(\mathbf{r},t) + 4\pi \mathbf{P}(\mathbf{r},t)\big) = e(\rho_I - \rho_e).
\end{aligned}$$

Maxwell–Schrödinger–Plasma (MASP) model for laser–molecule interactions: Towards an understanding of filamentation with intense ultrashort pulses

E. Lorin[a,c,*], S. Chelkowski[b], E. Zaoui[a], A. Bandrauk[a,b]

Physica D 241 (2012) 1059-1071

Full Maxwel system,
Driven by microscopic polarization and current

$$
\begin{aligned}
&\partial_t \mathbf{B}(\mathbf{r},t) = -c\nabla \times \mathbf{E}(\mathbf{r},t),\\
&\partial_t \mathbf{E}(\mathbf{r},t) = c\nabla \times \mathbf{B}(\mathbf{r},t) - 4\pi\big(\partial_t \mathbf{P}(\mathbf{r},t) + \mathbf{J}(\mathbf{r},t)\big),\\
&\nabla \cdot \mathbf{B}(\mathbf{r},t) = 0,\\
&\nabla \cdot \big(\mathbf{E}(\mathbf{r},t) + 4\pi \mathbf{P}(\mathbf{r},t)\big) = e(\rho_I - \rho_e).
\end{aligned}
$$

$$
\begin{aligned}
\mathbf{P}(\mathbf{r},t) &= \rho(\mathbf{r})\sum_{i=1}^{m}\mathbf{P}_i(\mathbf{r},t) = \rho(\mathbf{r})\sum_{i=1}^{m}\chi_{\Omega_i}(\mathbf{r})\\
&\times \int_{\mathbb{R}^3\times\mathbb{R}_+}\psi_i(\mathbf{r}',t)\mathbf{r}'\psi_i^*(\mathbf{r}',t)d\mathbf{r}',\\
\mathtt{i}\partial_t\psi_i(\mathbf{r}',t) &= -\frac{\triangle'_{\mathbf{r}}}{2}\psi_i(\mathbf{r}',t) + \mathbf{E}_{\mathbf{r}_i}\cdot\mathbf{r}'\psi_i(\mathbf{r}',t) + V_c(\mathbf{r}')\psi_i(\mathbf{r}',t)
\end{aligned}
$$

Polarization obtained from TDSE.
TDSE also provides “ionization rate”,
and number density depletion

Maxwell–Schrödinger–Plasma (MASP) model for laser–molecule interactions: Towards an understanding of filamentation with intense ultrashort pulses

E. Lorin[a,c,*], S. Chelkowski[b], E. Zaoui[a], A. Bandrauk[a,b]

Physica D 241 (2012) 1059-1071

Full Maxwel system,
Driven by microscopic polarization and current

$$\partial_t \mathbf{B}(\mathbf{r},t) = -c\nabla \times \mathbf{E}(\mathbf{r},t),$$
$$\partial_t \mathbf{E}(\mathbf{r},t) = c\nabla \times \mathbf{B}(\mathbf{r},t) - 4\pi\left(\partial_t \mathbf{P}(\mathbf{r},t) + \mathbf{J}(\mathbf{r},t)\right),$$
$$\nabla \cdot \mathbf{B}(\mathbf{r},t) = 0,$$
$$\nabla \cdot \left(\mathbf{E}(\mathbf{r},t) + 4\pi \mathbf{P}(\mathbf{r},t)\right) = e(\rho_I - \rho_e).$$

$$\mathbf{P}(\mathbf{r},t) = \rho(\mathbf{r})\sum_{i=1}^{m}\mathbf{P}_i(\mathbf{r},t) = \rho(\mathbf{r})\sum_{i=1}^{m}\chi_{\Omega_i}(\mathbf{r}) \times \int_{\mathbb{R}^3\times\mathbb{R}_+}\psi_i(\mathbf{r}',t)\mathbf{r}'\psi_i^*(\mathbf{r}',t)d\mathbf{r}',$$
$$\mathrm{i}\partial_t\psi_i(\mathbf{r}',t) = -\frac{\triangle_{\mathbf{r}}'}{2}\psi_i(\mathbf{r}',t) + \mathbf{E}_{\mathbf{r}_i}\cdot\mathbf{r}'\psi_i(\mathbf{r}',t) + V_c(\mathbf{r}')\psi_i(\mathbf{r}',t)$$

Polarization obtained from TDSE.
TDSE also provides “ionization rate”,
and number density depletion

TDSE “splits off” electrons deemed to be free of interaction with the atom, and these contribute to the classical current.

$$\partial_t\rho_e(\mathbf{r},t) = -\rho(\mathbf{r})\sum_{i=1}^{m}\chi_{\omega_i}(\mathbf{r})\partial_t\|\psi_i(t)\|_{L^2(\omega_i)}$$

$$\partial_t\mathbf{J} + \frac{1}{\tau_c}\mathbf{J} = \frac{e^2}{m_e}\rho_e\mathbf{E}.$$

Maxwell–Schrödinger–Plasma (MASP) model for laser–molecule interactions: Towards an understanding of filamentation with intense ultrashort pulses

E. Lorin[a,c,*], S. Chelkowski[b], E. Zaoui[a], A. Bandrauk[a,b]

Physica D 241 (2012) 1059-1071

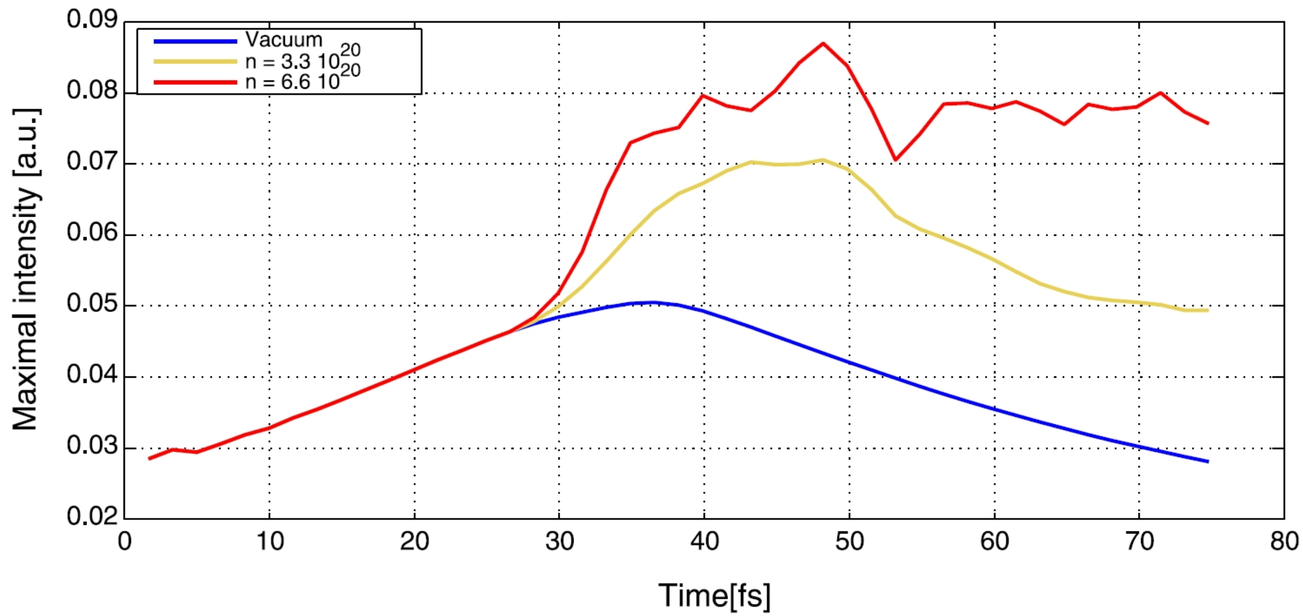


Fig. 14. Pulse amplitude maximum evolution for $I = 10^{14}$ W cm^{-2} as a function of time (fs).

Basic filament properties reproduced, BUT: it requires <u>extreme</u> computational effort

Maxwell–Schrödinger–Plasma (MASP) model for laser–molecule interactions: Towards an understanding of filamentation with intense ultrashort pulses

E. Lorin [a,c,*], S. Chelkowski [b], E. Zaoui [a], A. Bandrauk [a,b]

Physica D 241 (2012) 1059-1071

Appreciate the large computational scale

- The laser pulse possesses 5–6 cycles, with a pulse duration of $\sim$ 15 fs.
- The initial pulse wavelength is $\sim$ 800 nm.
- The total physical time duration of the propagation (vacuum/gas/vacuum) is typically $\sim$ 150 fs.
- The propagation length in the gas is $\sim$ 0.02 mm.
- The total length of the domain in z is $\sim$ 0.05 mm.
- The transversal window size is $\sim$10 μm $\times$ 10 μm.
- The chosen medium is a H_2^+-molecule gas.

...in what is rather small interaction volume

As of now, approach not practical for "whole experiment modeling"...
... but will be central to numerical experiments to elucidate new physics

First-principle based modeling: Extracting NL susceptibility from TDSE

PHYSICAL REVIEW A **87**, 043811 (2013)

Saturation of the nonlinear refractive index in atomic gases

Christian Köhler,[1] Roland Guichard,[2] Emmanuel Lorin,[3] Szczepan Chelkowski,[4] André D. Bandrauk,[4]
Luc Bergé,[1] and Stefan Skupin[5,6]

Linear system to extract susceptibility parameters:

$$\hat{P}_{\rm NL}(\omega_0, I_1) = \epsilon_0 {\rm FT}\{\chi^{(3)} E_1^3(t) + \chi^{(5)} E_1^5(t) + \cdots\}\big|_{\omega_0},$$
$$\hat{P}_{\rm NL}(\omega_0, I_2) = \epsilon_0 {\rm FT}\{\chi^{(3)} E_2^3(t) + \chi^{(5)} E_2^5(t) + \cdots\}\big|_{\omega_0},$$
$$\cdots = \cdots,$$

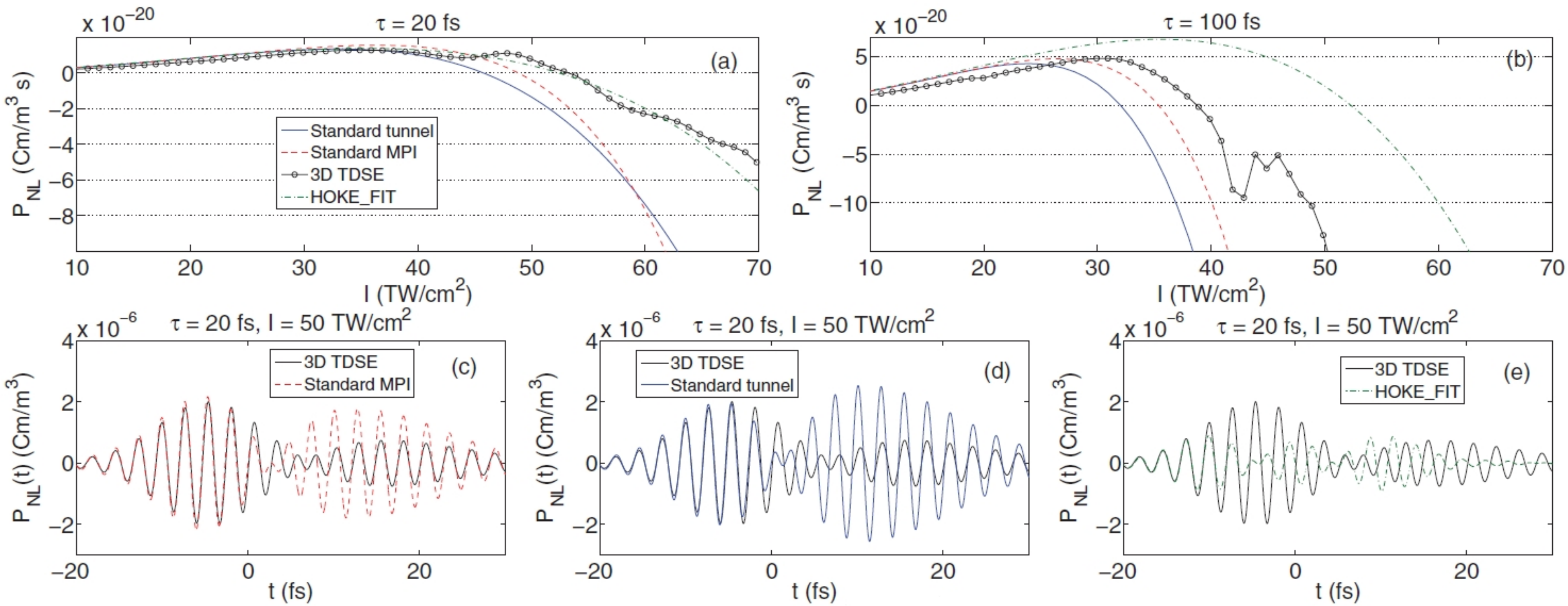
This system is over-determined,
Optimal solution would suggest existence (physical meaning to) susceptibilities

First-principle based modeling: Extracting NL susceptibility from TDSE

PHYSICAL REVIEW A **87**, 043811 (2013)

Saturation of the nonlinear refractive index in atomic gases

Christian Köhler,[1] Roland Guichard,[2] Emmanuel Lorin,[3] Szczepan Chelkowski,[4] André D. Bandrauk,[4]
Luc Bergé,[1] and Stefan Skupin[5,6]



BAD NEWS:
Single set of susceptibility parameters not applicable across different regimes/pulses

First-principle based modeling: Extracting NL susceptibility from TDSE

PRL 110, 043902 (2013) PHYSICAL REVIEW LETTERS week ending 25 JANUARY 2013

High-Field Quantum Calculation Reveals Time-Dependent Negative Kerr Contribution

P. Béjot,[1,*] E. Cormier,[2] E. Hertz,[1] B. Lavorel,[1] J. Kasparian,[3] J.-P. Wolf,[3] and O. Faucher[1]

$$P_{\mathrm{NL}}(t) = P(t) - \mathcal{N} \lim_{I_0 \mapsto 0} \alpha(I_0) E(t),$$

Implies linearity! $$\alpha(\omega_0) = \frac{p(\omega_0)}{E(\omega_0)}$$

$$\Delta\alpha(I_0) = \alpha(I_0) - \lim_{I_0 \mapsto 0} \alpha(I_0),$$ Breaks causality!

where I_0 is the peak intensity of the pulse.

First-principle based modeling: Extracting NL susceptibility from TDSE

PRL **110**, 043902 (2013) PHYSICAL REVIEW LETTERS week ending 25 JANUARY 2013

High-Field Quantum Calculation Reveals Time-Dependent Negative Kerr Contribution

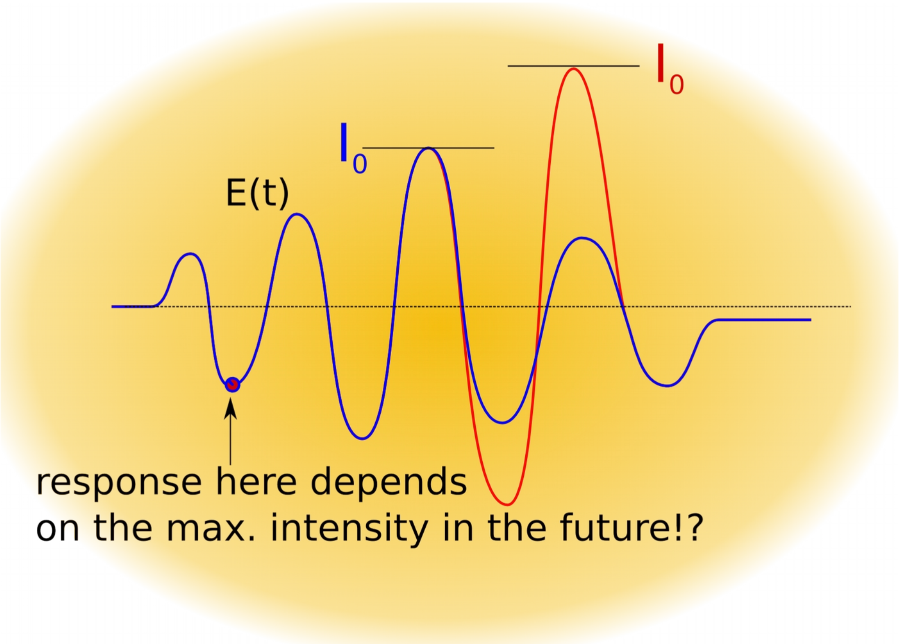
P. Béjot,[1,*] E. Cormier,[2] E. Hertz,[1] B. Lavorel,[1] J. Kasparian,[3] J.-P. Wolf,[3] and O. Faucher[1]

$$\alpha(\omega_0) = \frac{p(\omega_0)}{E(\omega_0)}$$

$$P_{\mathrm{NL}}(t) = P(t) - \mathcal{N} \lim_{I_0 \mapsto 0} \alpha(I_0) E(t),$$

$$\Delta\alpha(I_0) = \alpha(I_0) - \lim_{I_0 \mapsto 0} \alpha(I_0),$$

where I_0 is the peak intensity of the pulse.

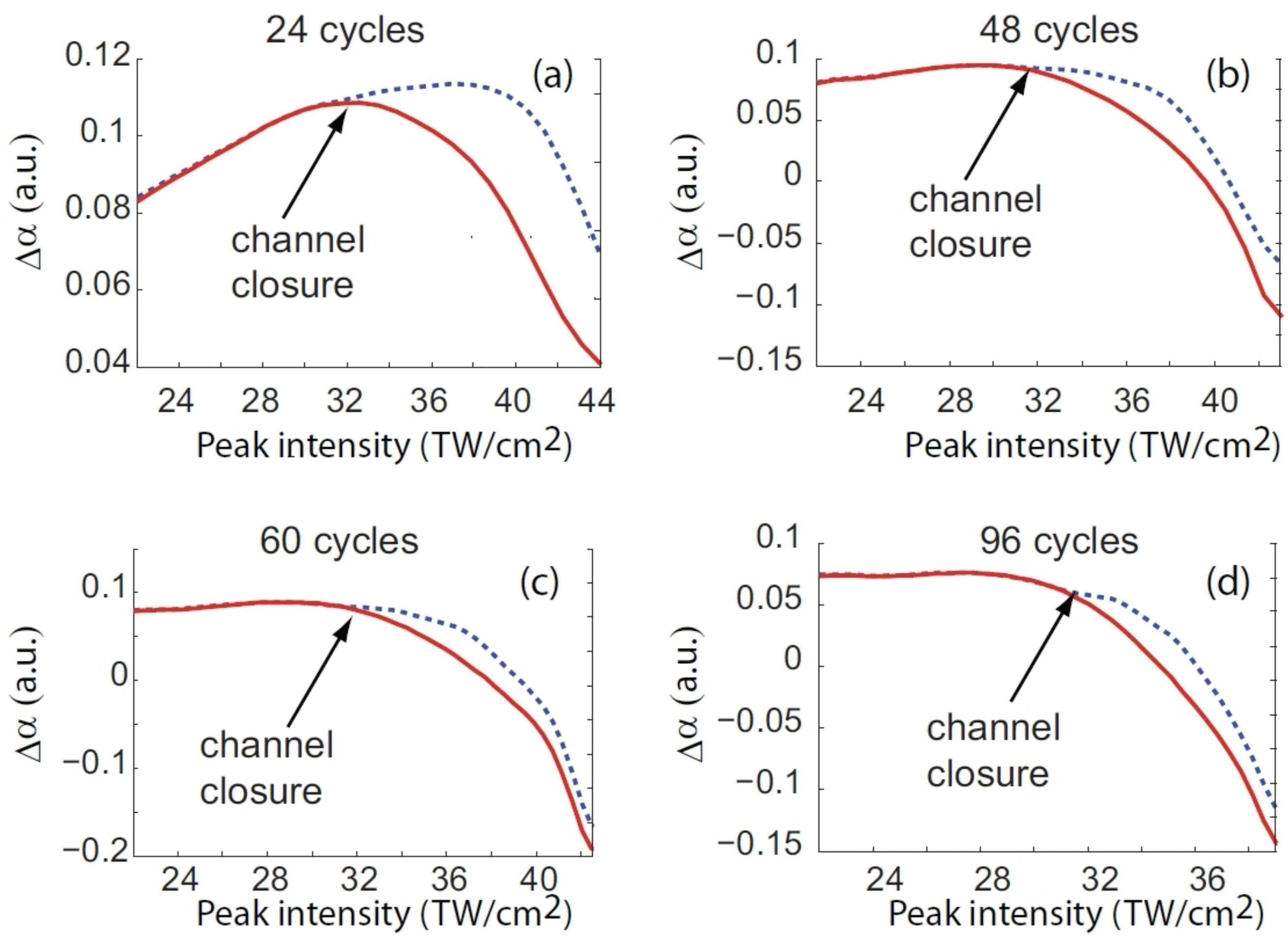


First-principle based modeling: Extracting NL susceptibility from TDSE

PRL 110, 043902 (2013) PHYSICAL REVIEW LETTERS week ending 25 JANUARY 2013

High-Field Quantum Calculation Reveals Time-Dependent Negative Kerr Contribution

P. Béjot,[1,*] E. Cormier,[2] E. Hertz,[1] B. Lavorel,[1] J. Kasparian,[3] J.-P. Wolf,[3] and O. Faucher[1]



TDSE results compared to KD (Kerr and Drude) model

The same approach, yet very different conclusion... -> perturbation approach not suitable

First-principle based modeling: Exactly solvable ionization model

PHYSICAL REVIEW A **83**, 013401 (2011)

Ionization of atoms by strong infrared fields: Solution of the time-dependent Schrödinger equation in momentum space for a model based on separable potentials

H. M. Tetchou Nganso,[1,2,*] Yu. V. Popov,[3] B. Piraux,[1,†] J. Madroñero,[4] and M. G. Kwato Njock[2]

$$\left[i\frac{\partial}{\partial t} - \frac{p^2}{2} - \frac{1}{c}A(t)(\vec{e}\,\vec{p})\right]\Phi(\vec{p},t) - \int \frac{d^3p\,'}{(2\pi)^3}V(\vec{p},\vec{p}\,')\Phi(\vec{p}\,',t) = 0; \quad \Phi(\vec{p},0) = \frac{8\sqrt{\pi}}{(p^2+1)^2}$$

$$V(\vec{p},\vec{p}\,') = -\frac{4\pi}{|\vec{p}-\vec{p}\,'|^2}. \qquad V(\vec{p},\vec{p}\,') \rightarrow -\frac{16\pi}{(p^2+1)(p'^2+1)}$$

Coulomb potential

Separable potential,
the same ground-state

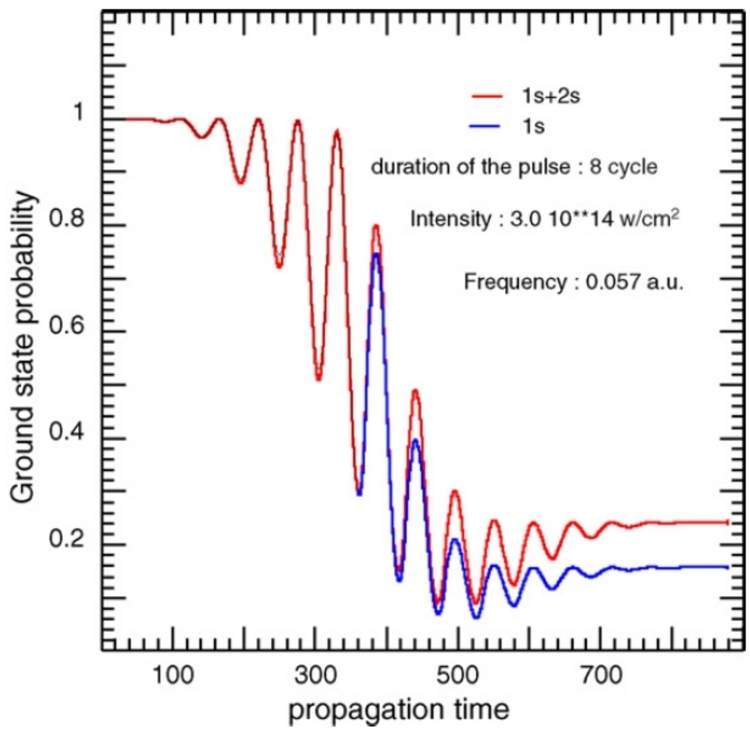
Exactly solvable model family. Ideal test-bed for approximate methods.

First-principle based modeling: Exactly solvable ionization model

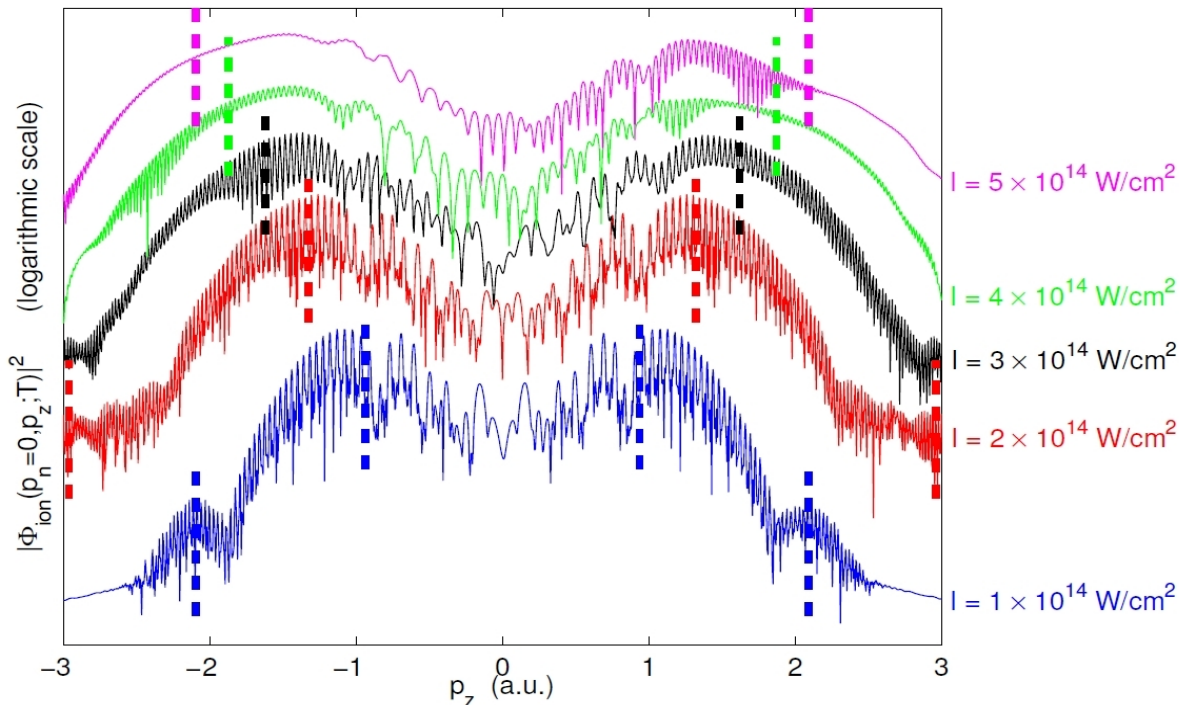
PHYSICAL REVIEW A **83**, 013401 (2011)

Ionization of atoms by strong infrared fields: Solution of the time-dependent Schrödinger equation in momentum space for a model based on separable potentials

H. M. Tetchou Nganso,[1,2,*] Yu. V. Popov,[3] B. Piraux,[1,†] J. Madroñero,[4] and M. G. Kwato Njock[2]



Adiabatic field following
in the ground-state survival



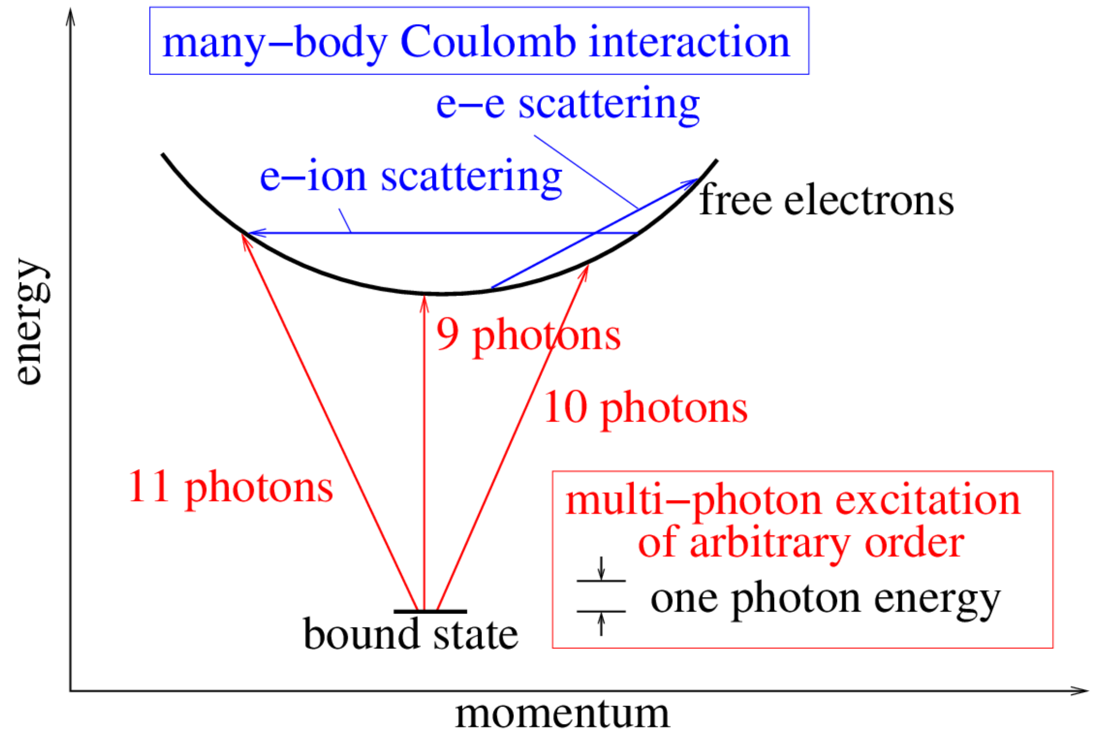
Ionized electrons momentum spectrum

Exactly solvable model family. Ideal test-bed for approximate methods.

Many-Body Theory of Short-Pulse Ionization

K. Schuh, J.Hader, J.V. Moloney, and S.W. Koch

- ionizing electrons experience Coulomb interactions with all other ionized electrons and ions
- dynamic modifications of electron distribution already during pulse
- N atoms, arbitrary detuning of electromagnetic field



- simplest example: one bound state and ionization continuum

Many-Atom Bloch Equations

optical polarization between ground state and continuum

$$i\hbar\frac{\partial}{\partial t}P_{sk} = (\epsilon_s - \epsilon_k)P_{sk} + \Omega^*_{sk} f_k - \Omega_{ks} f_s$$

Rabi energy $\Omega_{\alpha\beta} = \mathbf{d}_{\alpha\beta}\mathbf{E}(\mathbf{t})$ transition dipole $\mathbf{d}_{\alpha\beta} =< \alpha| - e\mathbf{r}|\beta >$

ground-state population

$$i\hbar\frac{\partial}{\partial t}f_s = \sum_k [\Omega^*_{sk}P_{ks} - \Omega_{sk}P^*_{ks}]$$

continuum-state population

$$i\hbar\frac{\partial}{\partial t}f_k = \Omega^*_{ks}P_{sk} - \Omega_{ks}P^*_{sk} + i\hbar\frac{\partial}{\partial t}f_k\Big|_{\mathrm{Coul}}$$

where $\frac{\partial k}{\partial t} = -\frac{eE}{\hbar}$ field acceleration of ionized carriers

Coulomb Scattering

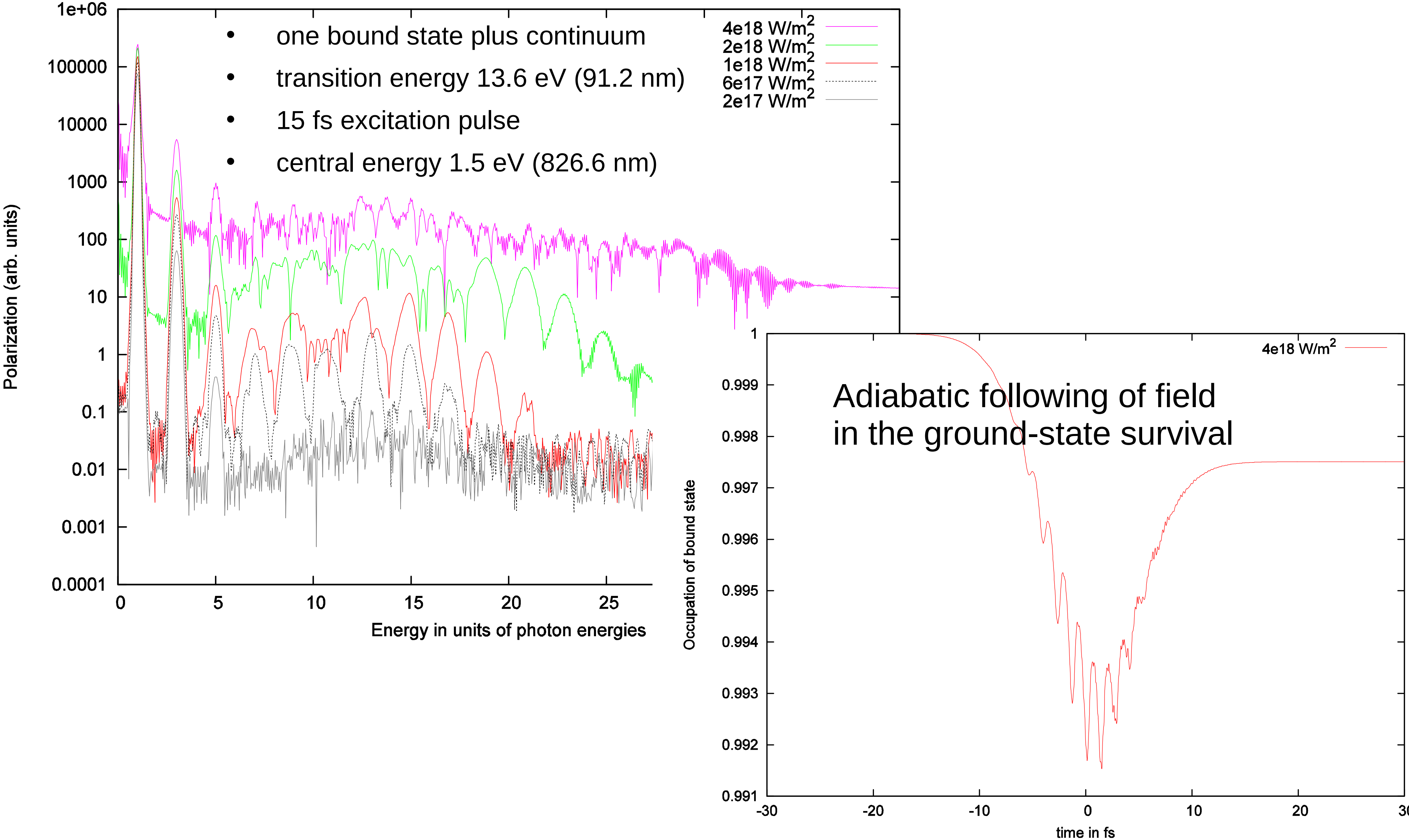
scattering between ionized electrons (low density limit)

$$\frac{\partial}{\partial t} f_\alpha \Big|_{\mathrm{Coul}}^{el-el} = \frac{1}{(i\hbar)^2} \frac{m}{\hbar\pi} \sum_{\beta\gamma\delta} \left[f_\alpha f_\delta - f_\beta f_\gamma \right] V_C(k_\alpha - k_\beta) V_C(k_\gamma - k_\delta) \delta(k_\alpha^2 + k_\delta^2 - (k_\beta^2 + k_\gamma^2))$$

elastic scattering with static ions

$$\frac{\partial}{\partial t} f_k \Big|_{\mathrm{Coul}}^{el-ion} = \frac{\hbar}{(i\hbar)^2} \frac{1}{2\pi} \sum_{k'} \delta(\epsilon_k - \epsilon_{k'}) V_C^2(k' - k) \left[f_k - f_{k'} \right]$$

First-principle based modeling: From ionization to equilibrated plasma



Three disparate time-scales treated in one model:
Quantum dynamics, optical pulse, and plasma equilibration

Exactly solvable "1D atom" in a homogeneous electric field

Hamiltonian = continuum + Dirac delta + external field

$$H\psi(x) = \frac{-\hbar^2}{2m}\frac{d^2\psi(x)}{dx^2} - eFx\psi(x).$$

Attractive delta-function potential realized through "boundary" conditions:

$$\left.\frac{d\psi(x)}{dx}\right|_{x=0+} - \left.\frac{d\psi(x)}{dx}\right|_{x=0-} = -A\psi(0)$$

Zero-field eigenstates: ground, continuum (even,odd)

$$< x|g >= \sqrt{k_0}e^{-k_0|x|} \quad , \quad E_g = -\frac{\hbar^2 k_0^2}{2m}$$

$$< x|a(k) >= \frac{1}{\sqrt{\pi}}\sin(kx) \quad , \quad E_k = \frac{\hbar^2 k^2}{2m}$$

$$< x|s(k) >= \frac{1}{\sqrt{\pi}}\cos(kx + \mathrm{sgn}(x)\phi(k)) \quad , \quad \tan(\phi(k)) = \frac{k_0}{k}$$

Exactly solvable “1D atom” in a homogeneous electric field

Hamiltonian resolvent
(tells us about the spectrum of the system)

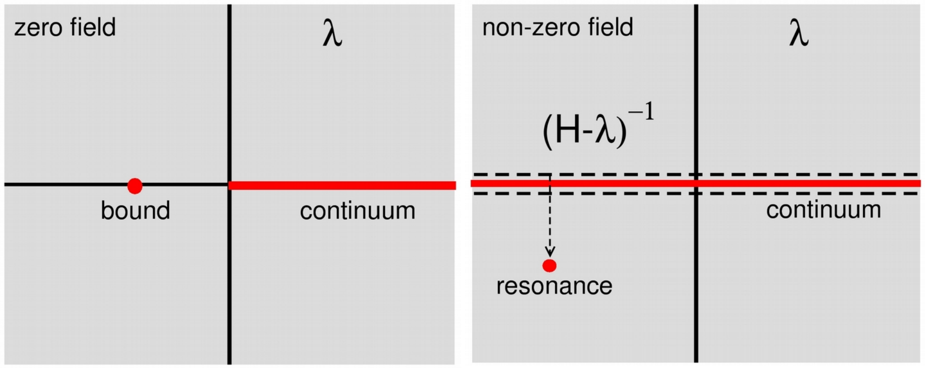
$$G(x,y,\lambda) = G_0(x,y,\lambda) + \frac{G_0(x,0,\lambda)G_0(0,y,\lambda)}{1/A - G_0(0,0,\lambda)}$$

$$G_0(x,y,\lambda) = \begin{array}{lll} -\psi_L(x)\psi_R(y)/W(\psi_R,\psi_R) & , & x<y \\ -\psi_R(x)\psi_L(y)/W(\psi_L,\psi_R) & , & x>y \end{array}$$

$$\psi_L(x,\lambda) = \mathrm{Ai}(-\xi) \quad \psi_R(x,\lambda) = \mathrm{Bi}(-\xi) + i\mathrm{Ai}(-\xi)$$

$$\xi = F^{1/3}(x+\lambda/F) \quad \text{and} \quad W = -F^{1/3}/\pi$$

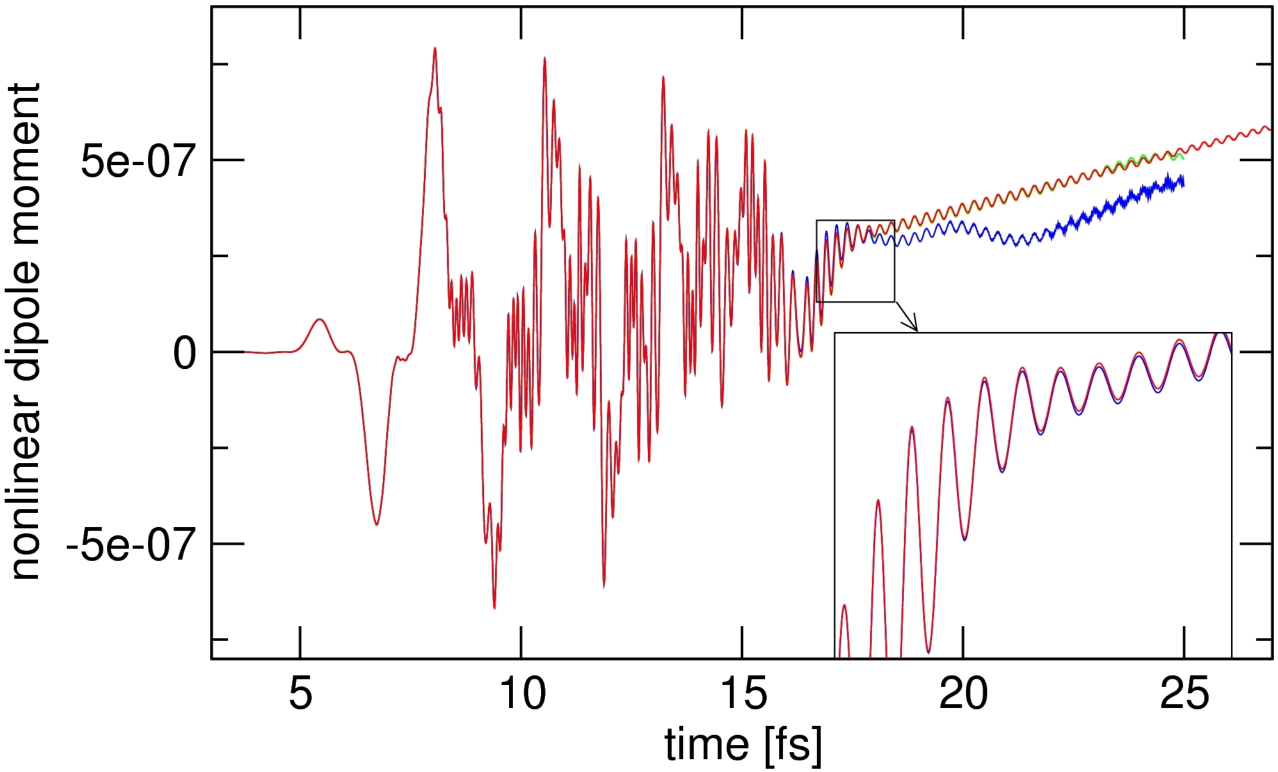
Spectrum in a static field



Homogeneous electric field
can never be a weak perturbation:

Spectrum immediately changes
its “topology”

Exact solution for arbitrary time-dependent field



Exactly solvable “1D atom” in a homogeneous electric field

Step 1: evaluate classical electron trajectory for the given time-dependent field

Exactly solvable "1D atom" in a homogeneous electric field

Step 1: evaluate classical electron trajectory for the given time-dependent field
Step 2: solve the integral equation

$$A(t) = \psi_R(-x_{cl}(t), t) + \frac{iB}{\sqrt{2\pi i}} \int_0^t dt' \frac{e^{+i\frac{B^2}{2}(t'-t)}}{\sqrt{t-t'}} \exp\left[\frac{i(x_{cl}(t) - x_{cl}(t'))^2}{2(t-t')}\right] A(t') \; ,$$

$$\psi_R(x,t) \equiv \frac{e^{+Bx}}{2} \mathrm{erfc}\left(\frac{iBt + x}{\sqrt{2it}}\right) + \frac{e^{-Bx}}{2} \mathrm{erfc}\left(\frac{iBt - x}{\sqrt{2it}}\right) \, .$$

Exactly solvable "1D atom" in a homogeneous electric field

Step 1: evaluate classical electron trajectory for the given time-dependent field
Step 2: solve the integral equation

$$A(t) = \psi_R(-x_{cl}(t), t) + \frac{iB}{\sqrt{2\pi i}} \int_0^t dt' \frac{e^{+i\frac{B^2}{2}(t'-t)}}{\sqrt{t-t'}} \exp\left[\frac{i(x_{cl}(t) - x_{cl}(t'))^2}{2(t-t')}\right] A(t') \, ,$$

$$\psi_R(x,t) \equiv \frac{e^{+Bx}}{2} \mathrm{erfc}\left(\frac{iBt + x}{\sqrt{2it}}\right) + \frac{e^{-Bx}}{2} \mathrm{erfc}\left(\frac{iBt - x}{\sqrt{2it}}\right) .$$

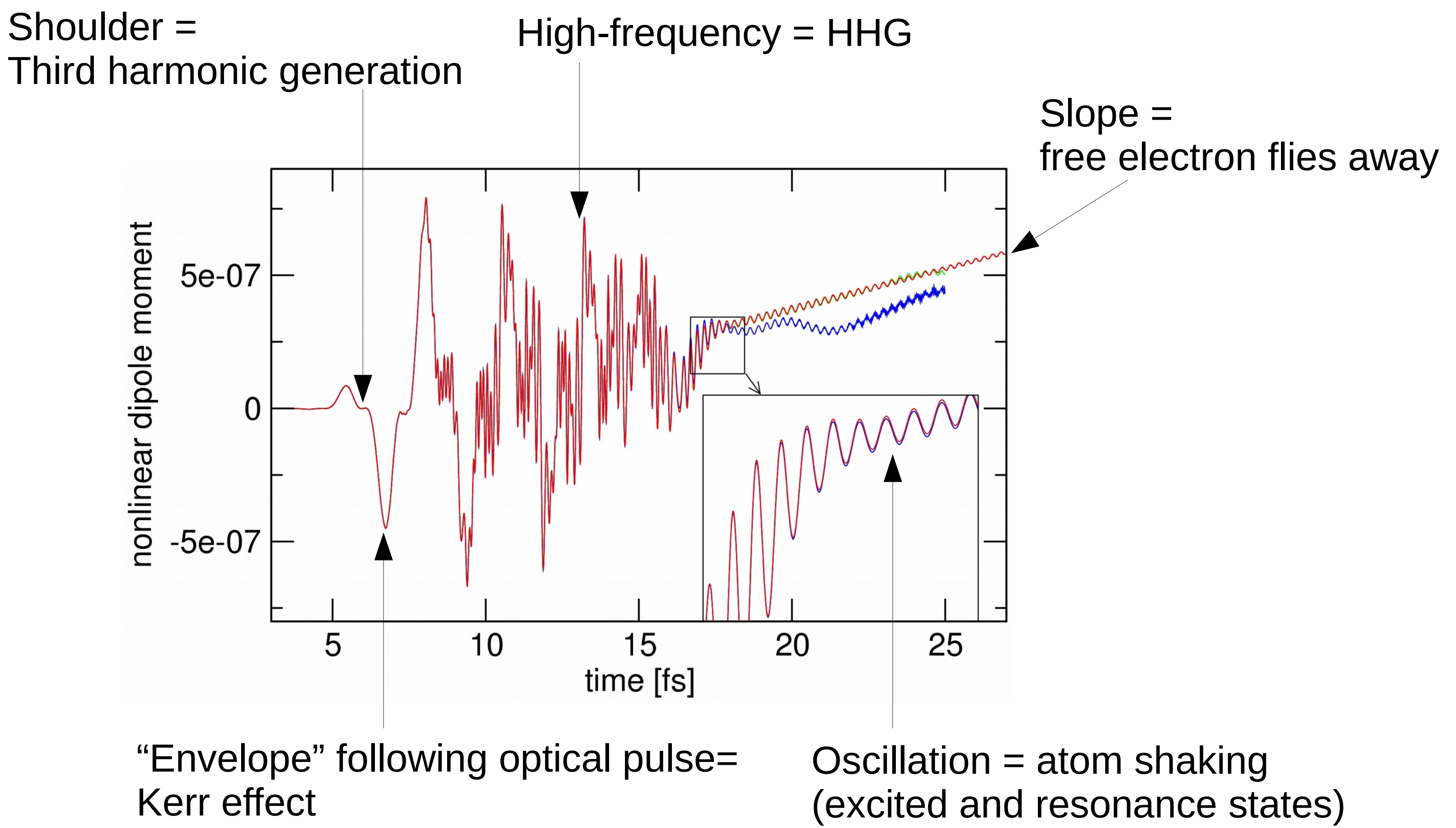
Step 3: evaluate non-linear component of the induced current

$$J_{SS}^{(nl)} = 2\mathrm{Im}\left\{\int_0^t dt_1 \int_0^{t_1} dt_2 \frac{(-i)^{\frac{3}{2}} B^3 W(t_1,t_2)}{\sqrt{2\pi}} \left[e^{\frac{i[x_{cl}(t_1)-x_{cl}(t_2)]^2}{2(t_1-t_2)}} A^*(t_1) A(t_2) - 1\right] \frac{x_{cl}(t_1) - x_{cl}(t_2)}{t_1 - t_2}\right\}$$

$$J_{FS} = \mathrm{Im}\left\{iB^3 \int_0^t dt_1 A^*(t_1) \left[e^{+Bx_{cl}(t_1)} \mathrm{erfc}\left(\frac{(1+i)(Bt_1 - ix_{cl}(t_1))}{2\sqrt{t_1}}\right) - e^{-Bx_{cl}(t_1)} \mathrm{erfc}\left(\frac{(1+i)(Bt_1 + ix_{cl}(t_1))}{2\sqrt{t_1}}\right)\right]\right\}$$

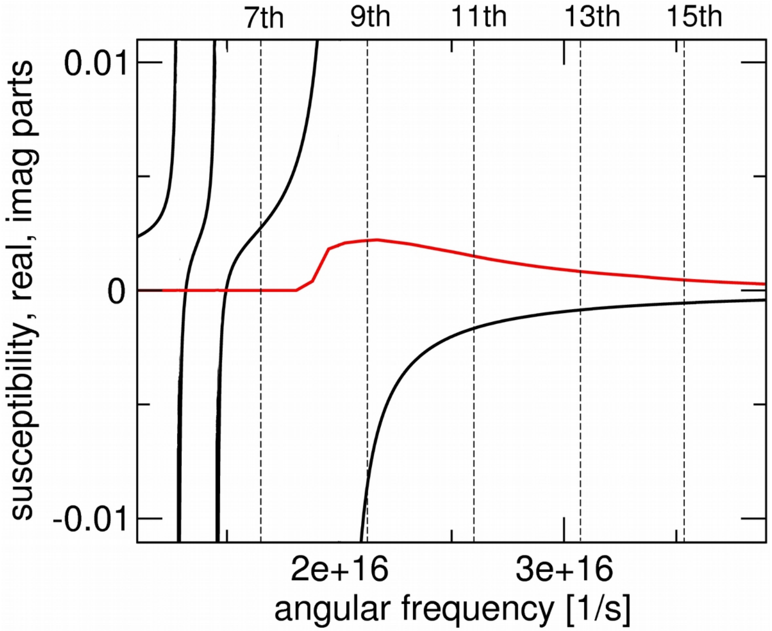
$$J_{FS}^{(nl)} = J_{FS} - \mathrm{Im}\left\{2B^3 \int_0^t dt_1 x_{cl}(t_1) \left(iB \, \mathrm{erfc}\left(\frac{(1+i)B\sqrt{t_1}}{2}\right) - \frac{1+i}{\sqrt{\pi t_1}} e^{-i\frac{B^2}{2}t_1}\right)\right\}$$

Step 4: optionally, integrate in time to obtain nonlinear polarization



This is all-in-one, microscopically calculated nonlinear response

Integration of Quantum-Mechanical and Pulse Propagation Solvers



Model consists of:

1. Linear chromatic dispersion and absorption :

2. Instantaneous Kerr, models contribution from bound electronic states

3. one-D quantum system, ground + continuum states, exact solution:

$$\left[i\partial_t + \frac{1}{2}\partial_x^2 + B\delta(x) - xF(t) \right] \psi(x,t) = 0$$

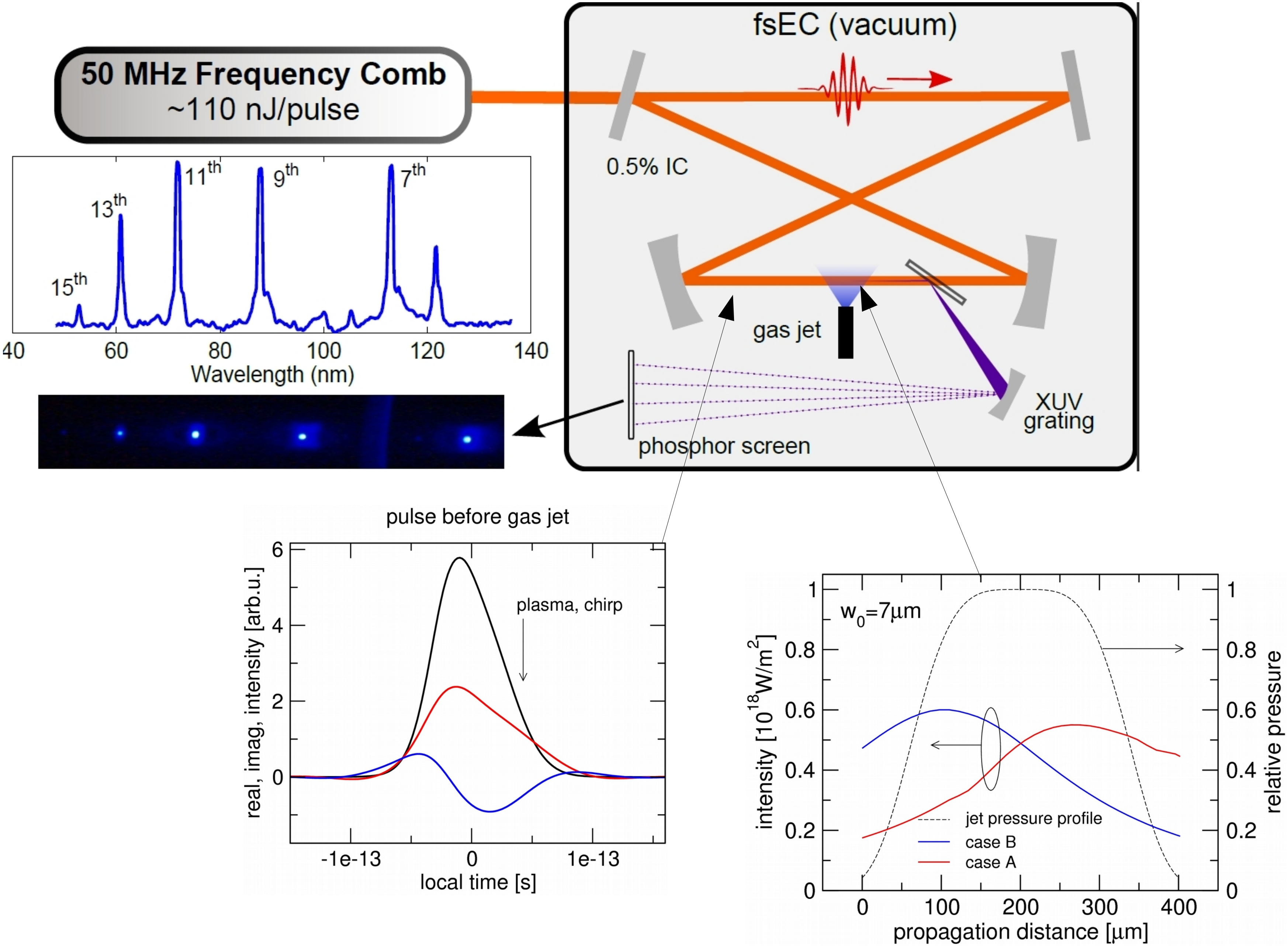
1+2+3 are fed into a UPPE simulator, and ***all*** frequency components are simulated as a single field

What is "missing" from the model:

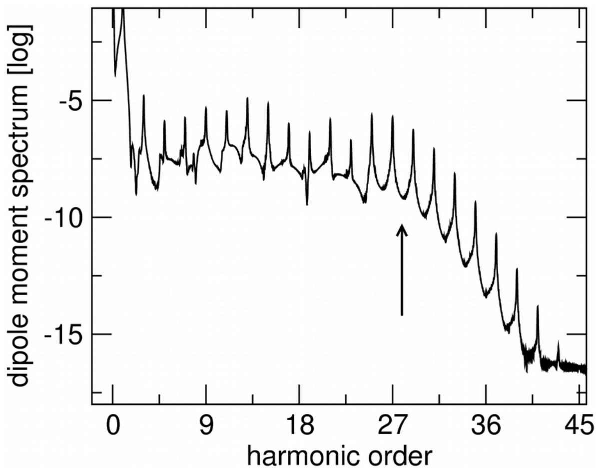
No ionization rate
No Drude plasma
No splitting into infra-red + high-harmonics
No splitting into bound + free electrons

... = most self-consistent approach yet

Fully resolved, self-consistent model for HH generation in a femtosecond enhancement cavity

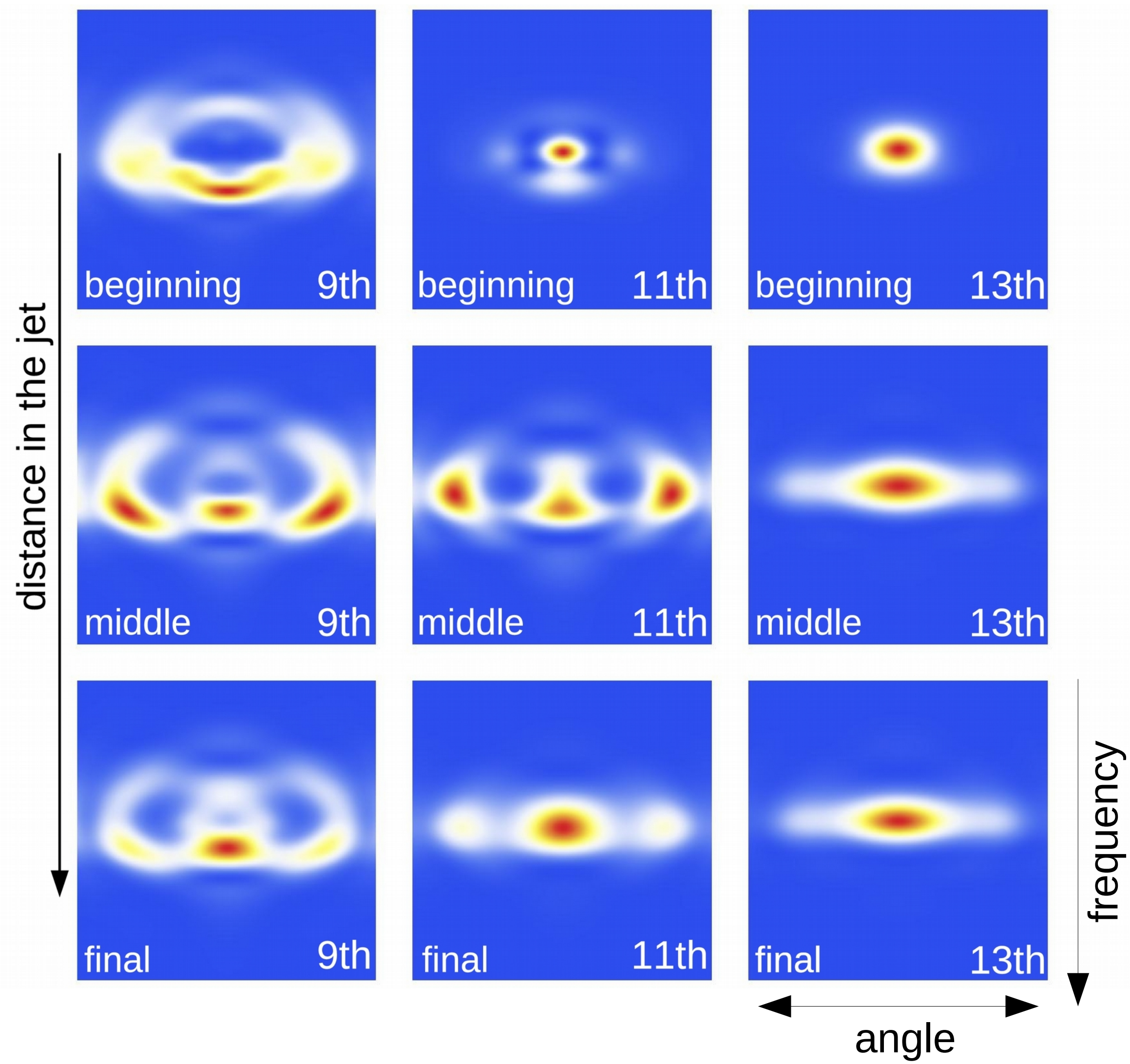


Fully resolved, self-consistent model for HH generation in a femtosecond enhancement cavity

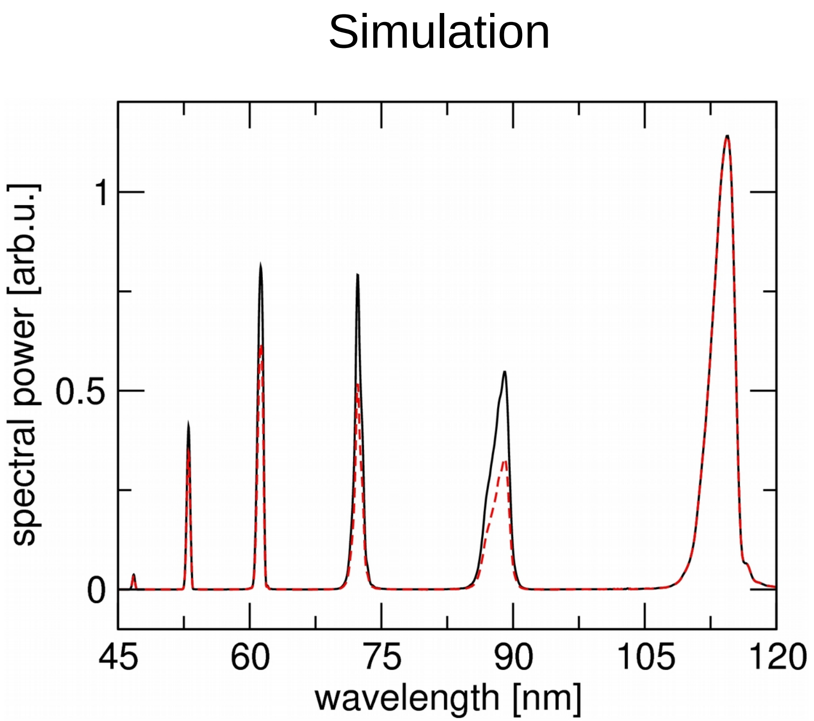


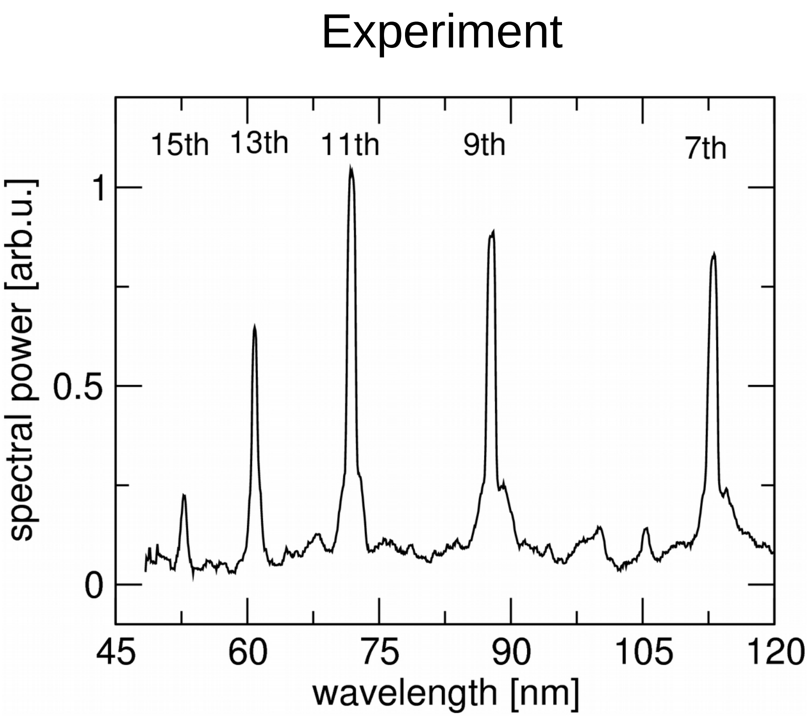
HHG physics similar to SFA

Angularly resolved HHG spectra



Fully resolved, self-consistent model for HH generation in a femtosecond enhancement cavity





Despite the simplicity of the atom model, agreement is good...

Lesson from using simple models:
Capturing qualitative physics more important than “fitting the result”

Way forward:
1) Characterize the atom/molecule by QM means
2) Tabulate and store “all important” responses
3) Use the latter in a full-blown Maxwell+Schrodinger simulation