Nonlinear propagation in a highly dispersive medium

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Introduction

- Highly absorptive, thus dispersive, medium: two close resonances
- Extreme optical non-linearity
- 2-3 photon ionisation (more effective than in air)
- Very long lifetimes (broadening mechanisms are negligible on the pulse width scale)
- Several meter-long propagation distance
  - To find a practical and effective modelling strategy is crucial
  - Conventional strategies are problematic

Modelling propagation in optics

- Expanding the dielectric susceptibility in a Taylor series in the electric field
  - This assumes the response is instantaneous and stationary
- In ultra-fast optics (fs pulses in the IR), we work far from resonances (normally in the UV or far-IR), so this approximation is sound
- Where experts are going: time-dependent Schrödinger equation (TDSE)
  - Mostly to justify further approximations, e.g. comparing to Maxwell-Bloch equations (MBE and their generalizations)
  - Exploit and understand new effects beyond PPT (conventional ionization theory): e.g. Kramers-Henneberger, high-order corrections to nonlinearities

Nonlinear wave equation

- Nonlinear polarisation $$P(E) = \epsilon_0(\hat{\chi}^{(1)}E + \hat{\chi}^{(2)}EE + \hat{\chi}^{(3)}EEE + \cdots)$$
- Nonlinear wave equation $$\partial_z^2 E - \frac{1}{c^2}\partial_t^2(E + \hat{\chi}^{(1)}E + \hat{\chi}^{(3)}EEE) = 0$$
- Conventional constant Kerr $$(\hat{\chi}^{(3)}EEE)_{\text{Kerr}} = \chi E^3, \quad \chi = \text{const.}$$
- Bidirectional wave equation: linear dispersion and four-wave mixing $$\partial_z^2 E_\omega + \beta^2(\omega)E_\omega + \frac{\omega^2}{c^2}\sum_{123|\omega}\chi^{(3)}_{123\omega}E_{\omega_1}E_{\omega_2}E_{\omega_3} = 0$$

$$\omega_1 + \omega_2 + \omega_3 = \omega$$

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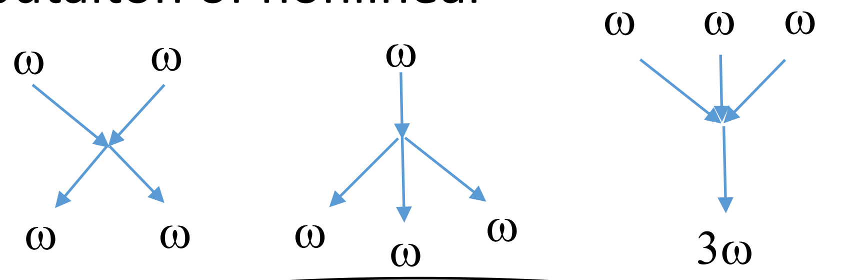
UPPE: unidirectional pulse propagation equation

$$i\,\partial_z \mathcal{E}_\omega + |\beta|\mathcal{E}_\omega + \frac{\omega^2}{c^2|\beta|}\sum_{123|\omega} \chi^{(3)}_{123\omega}\breve{\mathcal{E}}_{\omega_1}\breve{\mathcal{E}}_{\omega_2}\breve{\mathcal{E}}_{\omega_3} = 0$$

- Use of analytic signal $\mathscr{E}_\omega = \dfrac{E_\omega + E^*_{-\omega}}{2}$
  - Only positive frequencies ($\omega > 0$)
  - Discard negative frequencies at each computaiton of nonlinear response
- Conservation of energy
  - Either Kerr only $|\mathscr{E}|^2\mathscr{E}$
  - …or three terms: Kerr effect,
  - Conjugate Kerr effect,
  - and Third-order generation



$$|\mathscr{E}|^2\mathscr{E} \qquad (\mathscr{E}^*)^2\mathscr{E} \qquad \frac{1}{3}\mathscr{E}^3$$

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Dispersive non-linearities?

- Suppose we can obtain an effective nonlinear coefficient $n_2(\omega)$ (e.g. from “nonlinear Kramers-Kronig relations”)
- Is it an effective non-linearity legit?

$$n_{2,eff} = \frac{\int_0^\infty n_2(\omega) I(\omega) \mathrm{d}\omega}{\int_0^\infty I(\omega) \mathrm{d}\omega}$$

- Miller rule: far from resonances relationship between linear and nonlinear susceptibilities

$$\chi^{(3)}_{123\omega} = \text{const} \times \chi^{(1)}(\omega_1)\chi^{(1)}(\omega_2)\chi^{(1)}(\omega_3)\chi^{(1)}(\omega)$$

- What about losses?
- Each resonance has to be treated separately
- Does it give the same answer of steady-state response of Maxwell-Bloch or semi-classical approaches? No

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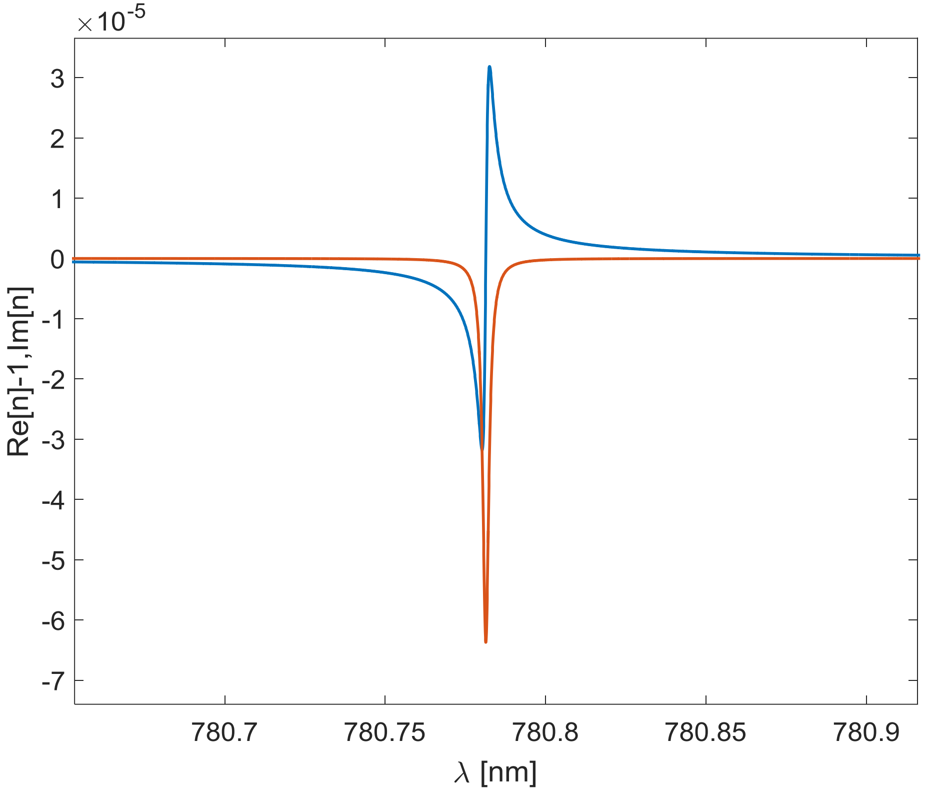
Linear and nonlinear susceptibilities



- Refractive index: real part odd, imaginary part even
- Nonlinear susceptibility (MBE, 2-level approximation)

$$\chi^{(3)} = \frac{\alpha_0(0)}{3\omega_{ba}/c}\left[\frac{\Delta T_2 - i}{(1+\Delta^2 T_2^2)^2}\right]\frac{2\epsilon_0 c}{I_s^0}$$

  - Real part odd
  - Imaginary part even

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New normalisation

$$\mathscr{A}_\omega = \chi^{(1)}(\omega)\mathscr{E}_\omega$$

Positive part of the spectrum

$$i\partial_z\mathscr{A} + |\beta|\mathscr{A} + \sum_{\text{resonances}} \frac{\omega^2\chi^{(1)}}{c^2|\beta|}[\mathscr{A}^3]_+ = 0$$

- Numerical technicalities
  - Reduce spectrum sampling by solving in a frame moving at group velocity at laser frequency
  - Split-step/pseudo-spectral
  - Adaptive

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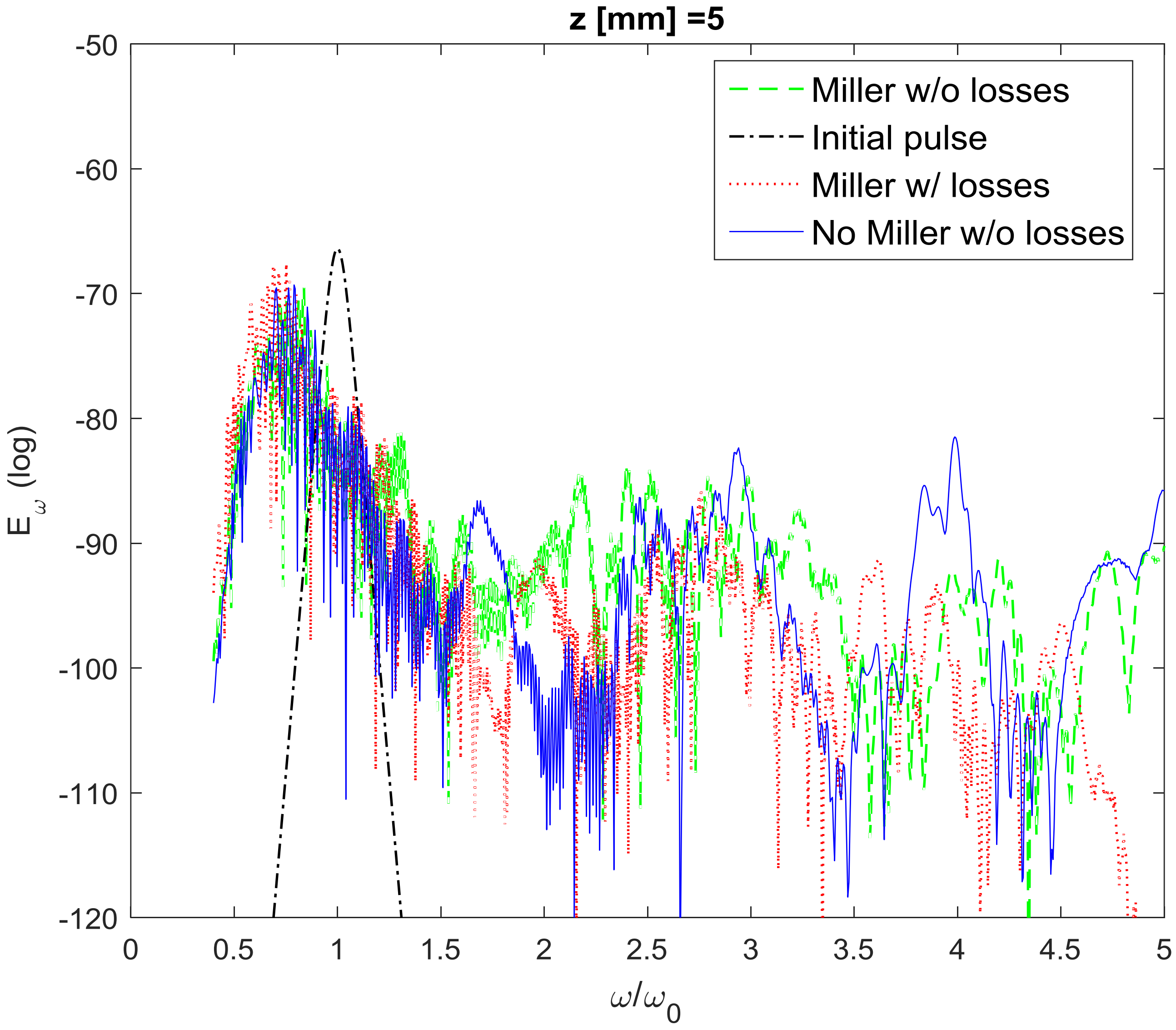
Example: Short pulse in Silica

- T0 = 15 fs
- Ipeak = 10 TW/cm^2
- lambda0 = 2mu
- L = 5 mm
- n2 = 2.7 x 10^-20 m^2/W
- Sellmeier 3 resonances (68 nm,116 nm, 10 μm)
- Nt = 2^15
- Nz = 2x10^4
- Complex interplay of resonant radiation, self-steepening and 3rd harmonic generation
  - Tiny changes in Super-continuum features



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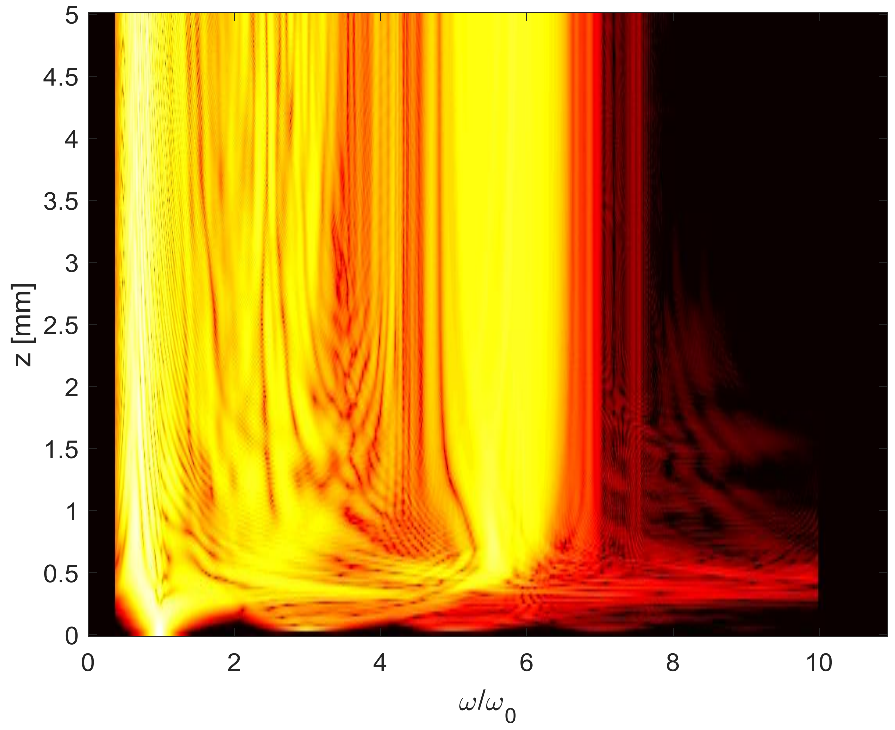
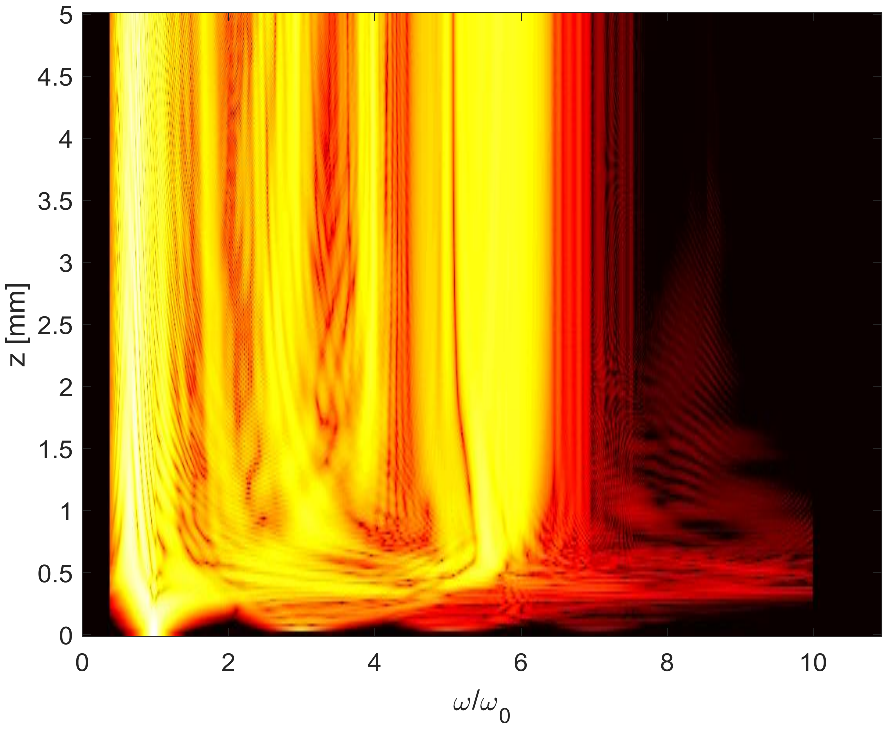
Example: Short pulses in Silica

Miller



No Miller



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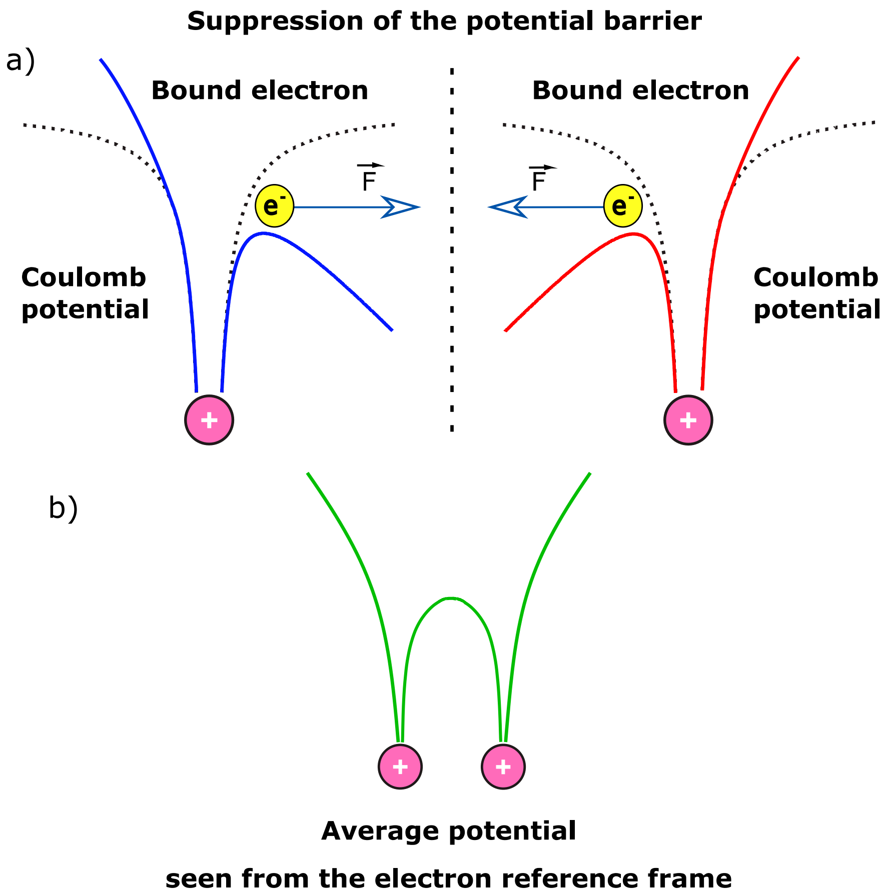
Pros&Cons

- Pros:
  - Easy to include ionisation and spatial effects
  - Lesser numerical effort than MBE

- Cons:
  - Narrow resonances mean extreme non-linearity
    - Losses at the linear level, non-linear part is more difficult
    - Stability of the numerical solver
  - Still Miller rule is debated, other solutions are not factorisable in the same way

The problem with ionization

- Limits of PPT (see Gabor Demeter's talk)
- Stabilization of atoms in strong electric fields: KH atom



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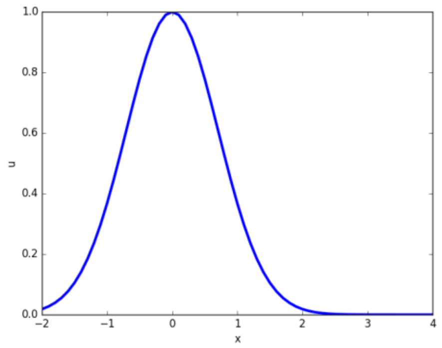
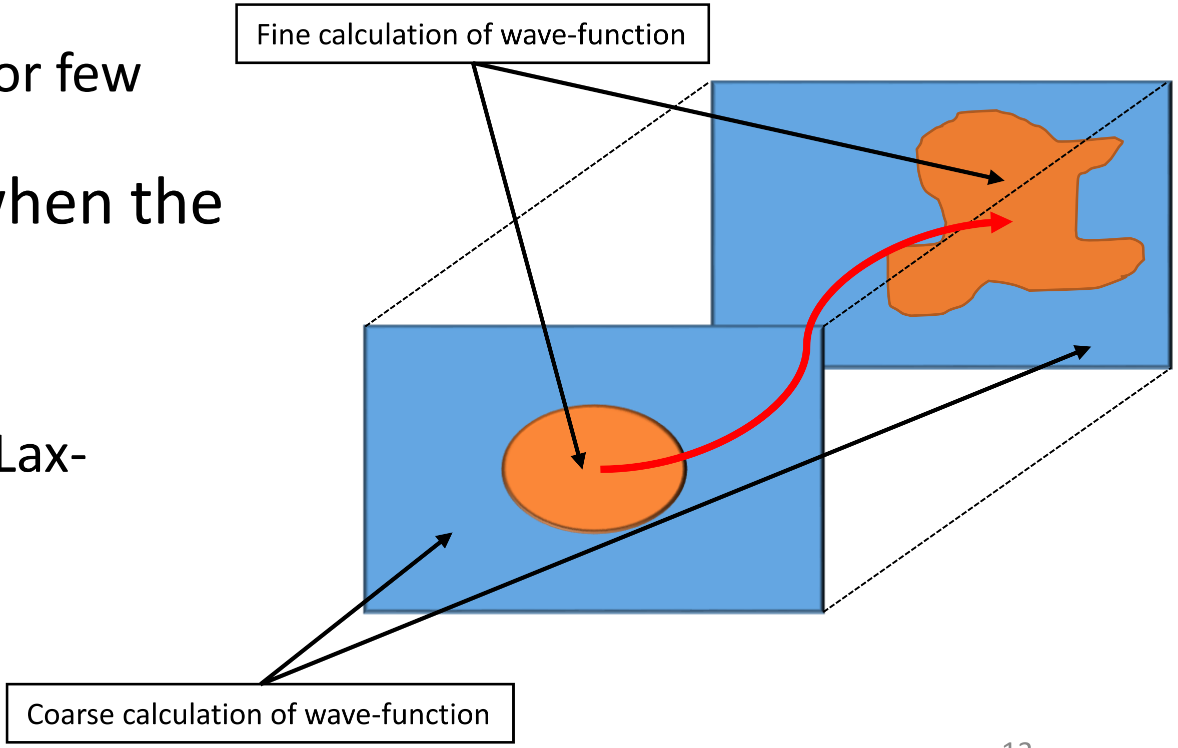
TDSE

- Back to ab-initio
  - In collaboration with prof. E. Cormier (Bordeaux): solve directly the full *electronic polarisation* from the Time-Dependent Schrödinger equation (density of dipoles)

$$\mathscr{P}(t) = N_{\text{at}} q_e \langle \Psi(t) | \hat{x} | \Psi(t) \rangle$$

  - Different exact and effective potentials available (H, He, Ar…)
  - 1D (+1D for time) propagation takes months for few centimetres length
- Possible improvements: solve TDSE only when the field is strong
  - Adapt TDSE to different field amplitudes
  - Multi-thread computation of different points
  - Follow the propagation of the wave: upwind, Lax-Wendroff…





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Is it really a filament

- If it were a filament, we would have an homogeneous intensity and stable propagation across the cell.
- A filament requires high intensity to be kept over several centimetres or metres and losses to be not too strong
- Here we have strong linear and non-linear absorption and non-negligible ionization probability (2-3 photons instead of >5)
- In alkali atoms, it seems clear that the nonlinearity switches from focusing to defocusing across the resonance.

Different strategies

- For filamentation in gases, particularly at higher intensity regimes (e.g. multifilament), the effect of the background dispersion is an almost negligible cause of catastrophic self-focusing effect
- It becomes important in the interplay with plasma near the focusing point
- Ionisation wipes coherent effects away (is it true?)
- Ionisation is much more efficient in Rb and several paths are in competition
- Looking for a simplified approach: what if only plasma mattered and the populations and coherences are fast saturated to a predictable value?
- If focusing occurs, what are the nonlinear mechanisms?