Electrons towards the speed of light: Photoionization in the strong field region

(Solving the Dirac equation)

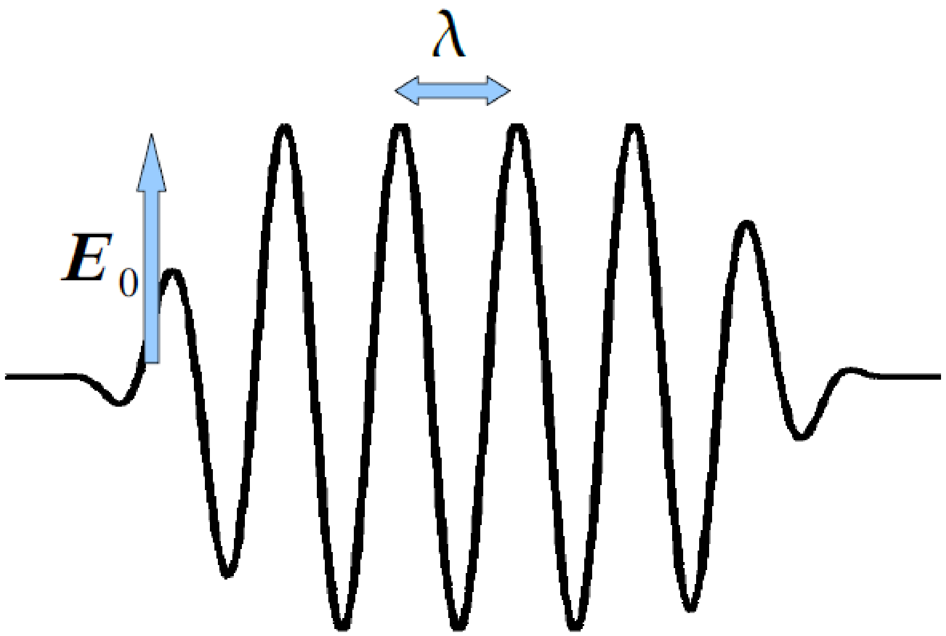
LPHYS’18, Nottingham, July 17th 2018

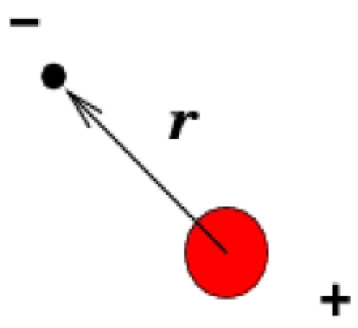
OSLOMET

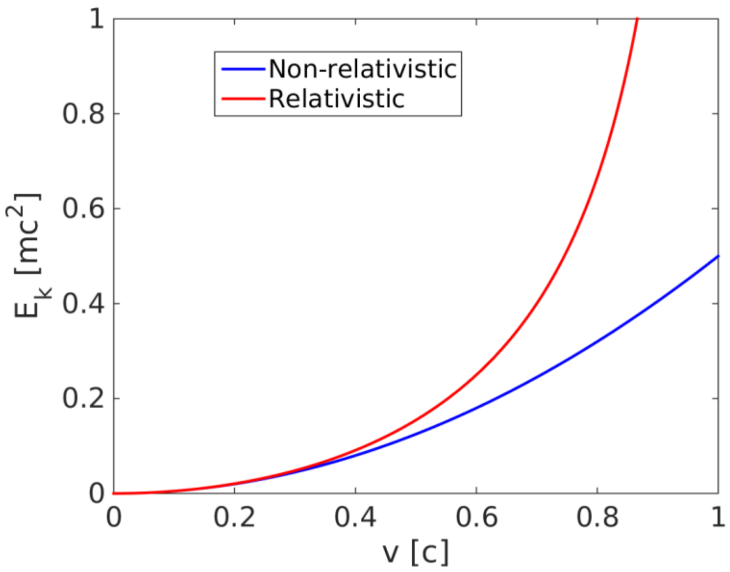
Aim:

Describe photoionization with laser fields strong enough to accelerate electrons towards the speed of light

XUV-region







Aim:
Describe photoionization with laser fields strong enough to accelerate electrons towards the speed of light
XUV-region


Paul Dirac

In other words:
We want so solve the time-dependent Dirac equation:

$$i\hbar\frac{d}{dt}\Psi = H\Psi$$

$$H = c\boldsymbol{\alpha}\cdot(\mathbf{p}+e\mathbf{A}) + V\mathbb{1}_4 + \beta mc^2$$

Aim:
Describe photoionization with laser fields strong enough to accelerate electrons towards the speed of light
XUV-region

In other words:
We want so solve the time-dependent Dirac equation:

Paul Dirac

$$i\hbar \frac{d}{dt}\Psi = H\Psi \qquad \Psi \in \mathbb{C}^4$$

$$H = c\boldsymbol{\alpha} \cdot (\mathbf{p} + e\mathbf{A}) + V\mathbb{1}_4 + \beta mc^2$$

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix} \qquad \beta = \begin{pmatrix} \mathbb{1}_2 & 0 \\ 0 & -\mathbb{1}_2 \end{pmatrix}$$

Aim:
Describe photoionization with laser fields strong enough to accelerate electrons towards the speed of light
XUV-region

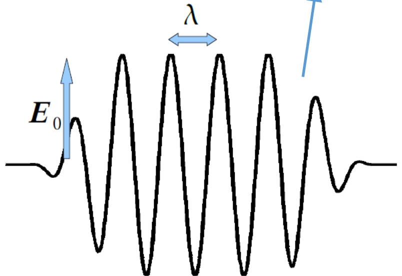
In other words:
We want so solve the time-dependent Dirac equation:

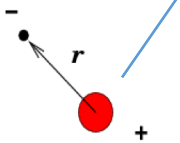
$$i\hbar \frac{d}{dt}\Psi = H\Psi \qquad \Psi \in \mathbb{C}^4$$


Paul Dirac

$$H = c\boldsymbol{\alpha}\cdot(\mathbf{p} + e\mathbf{A}) + V\mathbb{1}_4 + \beta mc^2$$

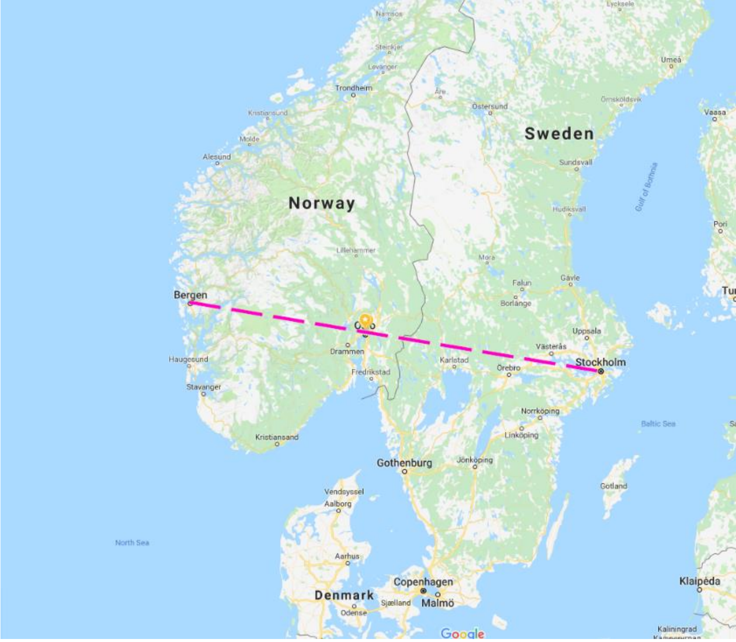
$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix}$$





$$\beta = \begin{pmatrix} \mathbb{1}_2 & 0 \\ 0 & -\mathbb{1}_2 \end{pmatrix}$$

The Team



Tor Kjellsson Lindblom, PhD

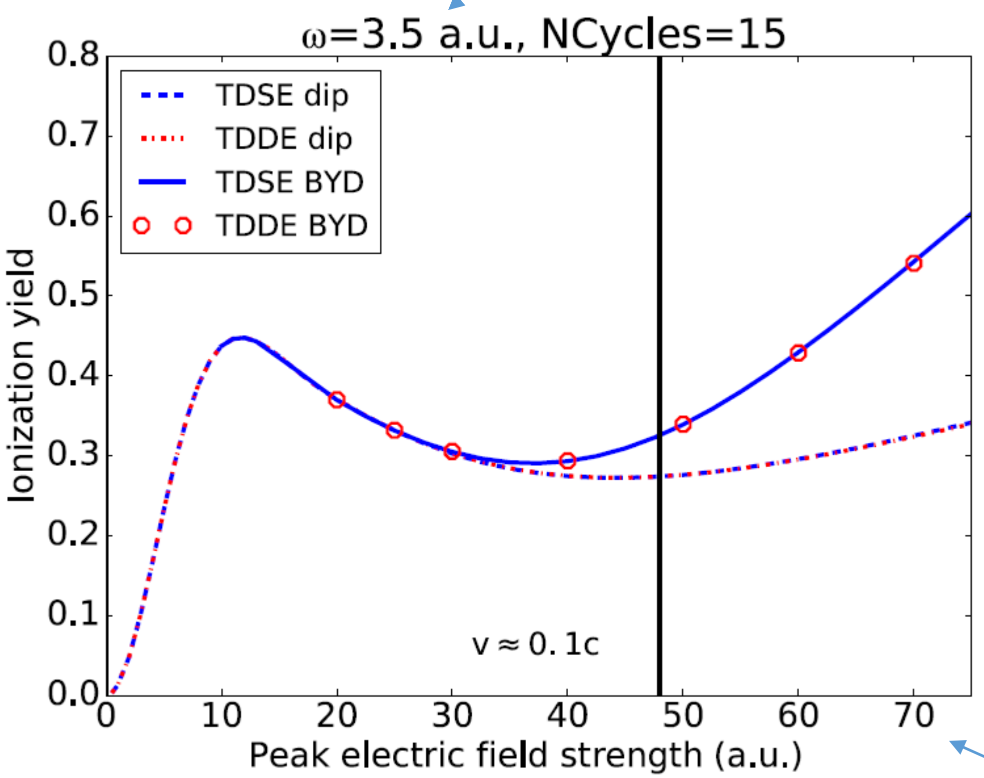
Prof. Eva Lindroth

Prof. Morten Førre

Prof. Sølve Selstø

Results

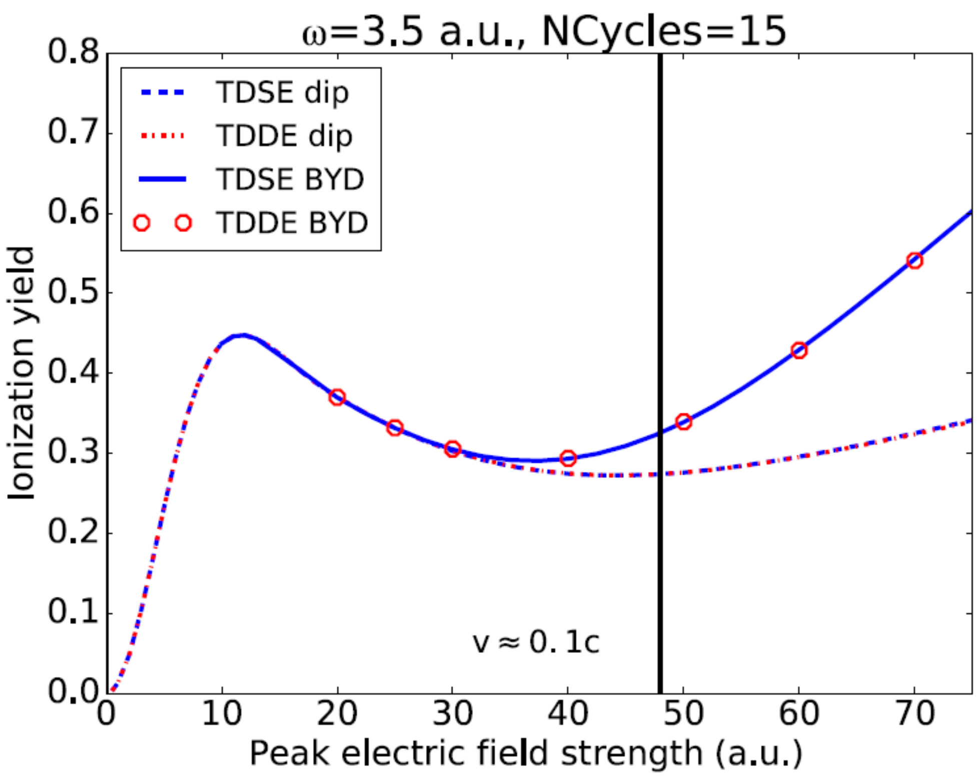
~ 100 eV

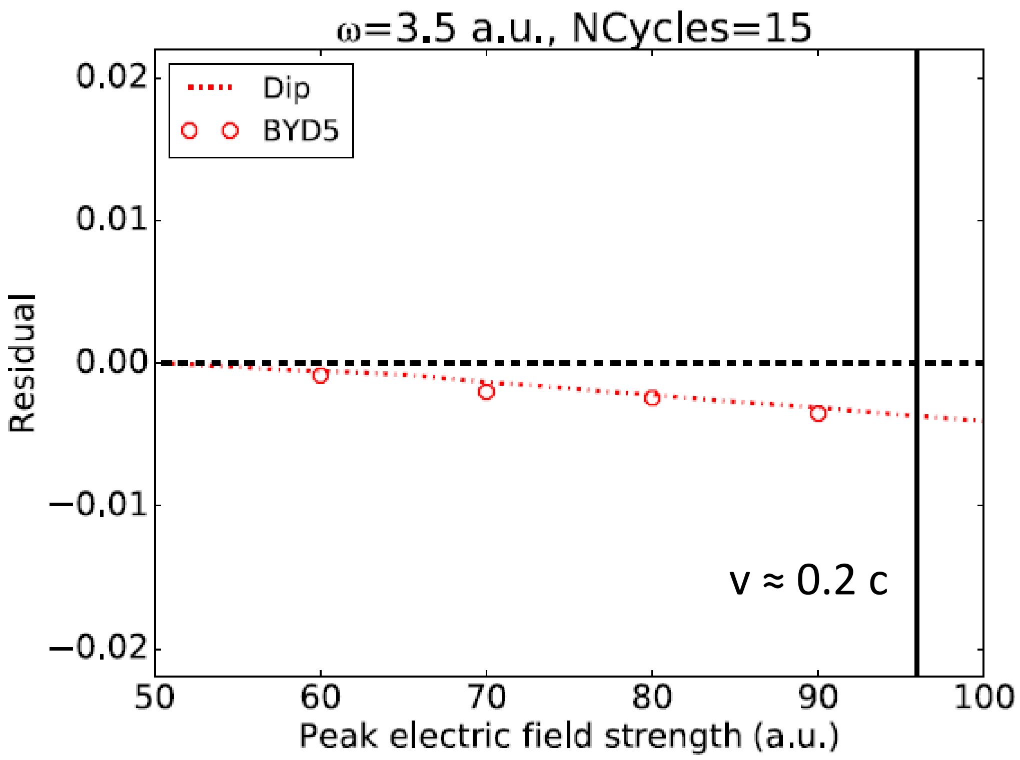


~ $1.7 \cdot 10^{20}$ W/cm^2

Kjellsson, Selstø, Lindroth, Phys. Rev. A **95**, 043403 (2017)

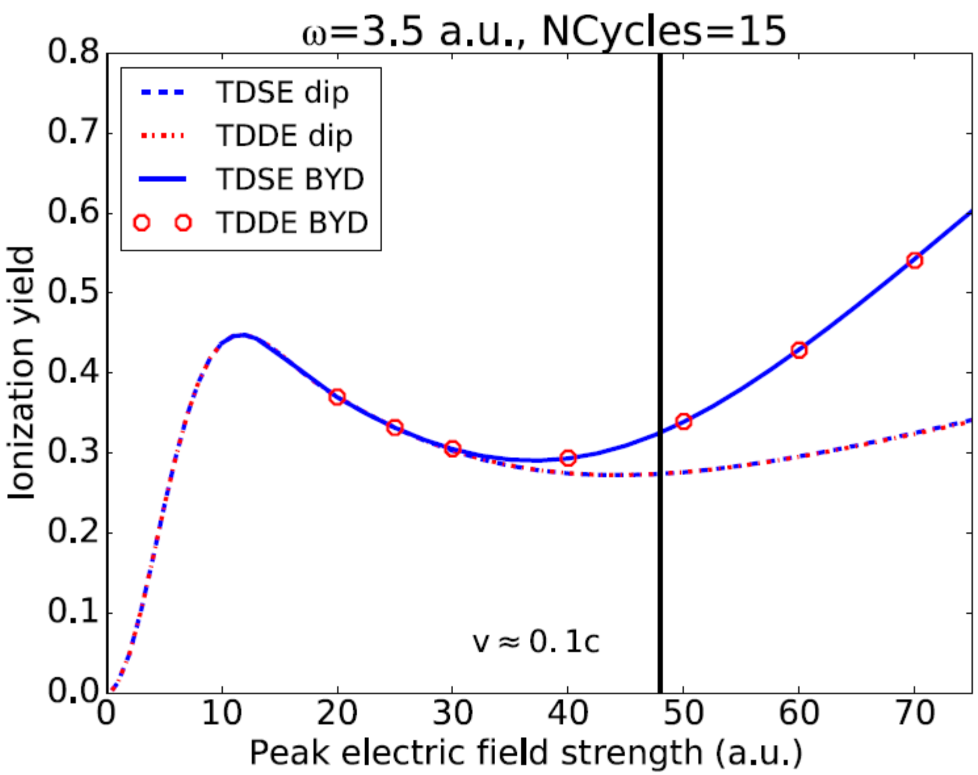
Results

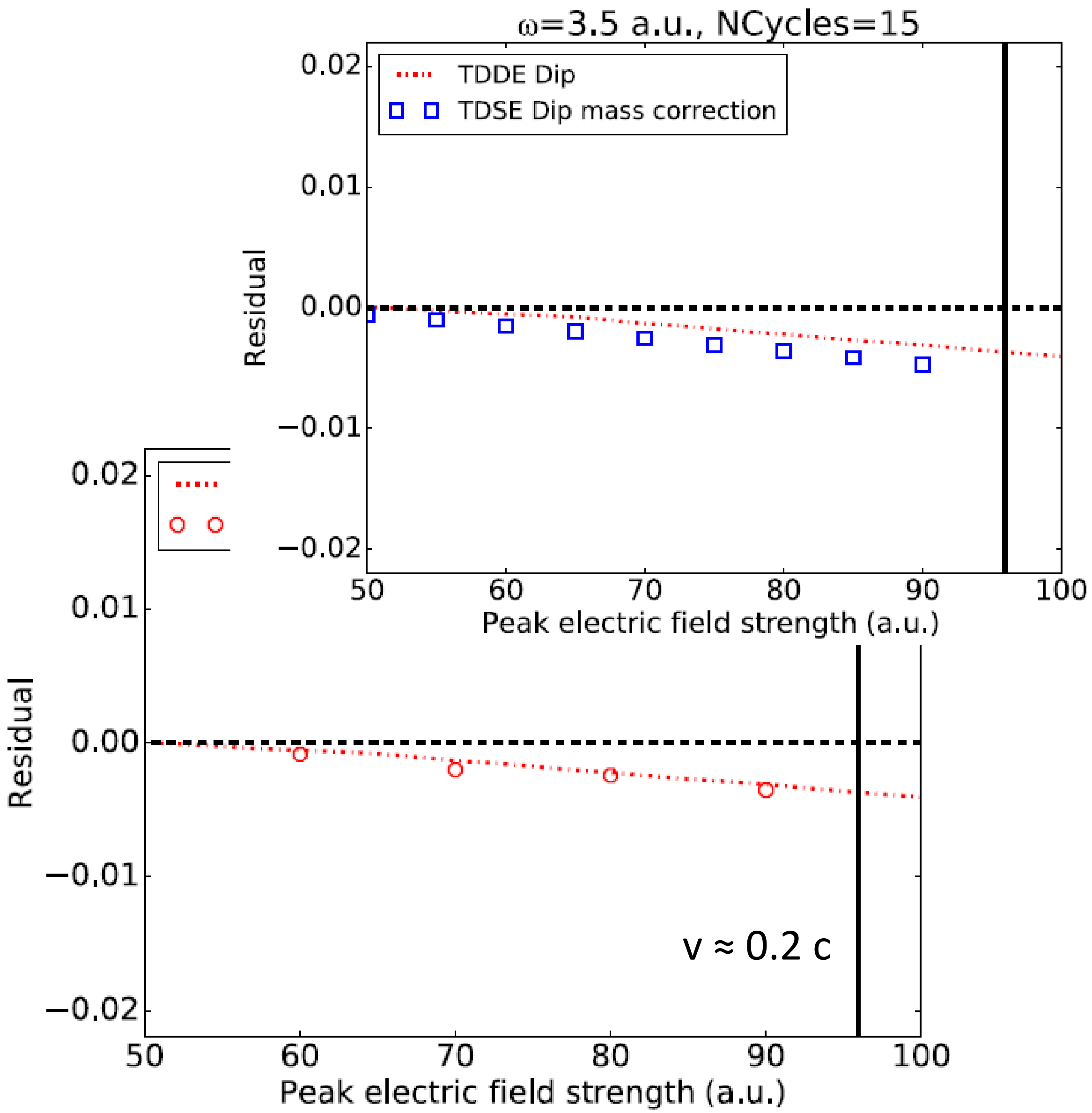




Kjellsson, Selstø, Lindroth, Phys. Rev. A **95**, 043403 (2017)

Results





Kjellsson, Selstø, Lindroth, Phys. Rev. A **95**, 043403 (2017)

Approach:

$$i\hbar\frac{d}{dt}\Psi = [H_0 + H'(t)]\Psi$$

1) Construct spectral basis by solving the time independent Dirac equation

$$H_0\varphi_n = \varepsilon_n\varphi_n, \qquad H_0 = c\boldsymbol{\alpha}\cdot\mathbf{p} + V + mc^2\beta$$

2) Express interaction in terms of this basis:

$$H'_{kl} = \langle\varphi_k|c\boldsymbol{\alpha}\cdot e\mathbf{A}|\varphi_l\rangle$$

3) Solve the resulting ordinary differential equation (ODE):

$$i\hbar\frac{d}{dt}\mathbf{c} = [\mathrm{Diag}(\varepsilon_1, \varepsilon_2, \ldots) + H']\,\mathbf{c}, \qquad \Psi = \sum_n c_n\varphi_n$$

Approach:

$$i\hbar\frac{d}{dt}\Psi = [H_0 + H'(t)]\Psi$$

1) Construct spectral basis by solving the time independent Dirac equation

$$H_0\varphi_n = \varepsilon_n\varphi_n, \qquad H_0 = c\boldsymbol{\alpha}\cdot\mathbf{p} + V + mc^2\beta$$

2) Express interaction in terms of this basis:

How hard can it be?

$$H'_{kl} = \langle\varphi_k|c\boldsymbol{\alpha}\cdot e\mathbf{A}|\varphi_l\rangle$$

3) Solve the resulting ordinary differential equation (ODE):

$$i\hbar\frac{d}{dt}\mathbf{c} = [\mathrm{Diag}(\varepsilon_1, \varepsilon_2, \ldots) + H']\,\mathbf{c}, \qquad \Psi = \sum_n c_n\varphi_n$$

Approach:

$$i\hbar\frac{d}{dt}\Psi = [H_0 + H'(t)]\Psi$$

1) Construct spectral basis by solving the time independent Dirac equation

$$H_0\varphi_n = \varepsilon_n\varphi_n, \qquad H_0 = c\boldsymbol{\alpha}\cdot\mathbf{p} + V + mc^2\beta$$

2) Express interaction in terms of this basis:

$$H'_{kl} = \langle\varphi_k|c\boldsymbol{\alpha}\cdot e\mathbf{A}|\varphi_l\rangle$$

How hard can it be?

Actually, there are a number of problems

3) Solve the resulting ordinary differential equation (ODE):

$$i\hbar\frac{d}{dt}\mathbf{c} = [\mathrm{Diag}(\varepsilon_1,\varepsilon_2,\ldots) + H']\,\mathbf{c}, \qquad \Psi = \sum_n c_n\varphi_n$$

Problem 1: Stiffness

$$\Psi(t+\Delta t) = U(t+\Delta t, t)\Psi(t) + \mathcal{O}(\Psi^{(n)}(t)\Delta t^n)$$

$$\Psi^{(n)}\Delta t^n \sim H^n \Psi(t) \Delta t^n$$

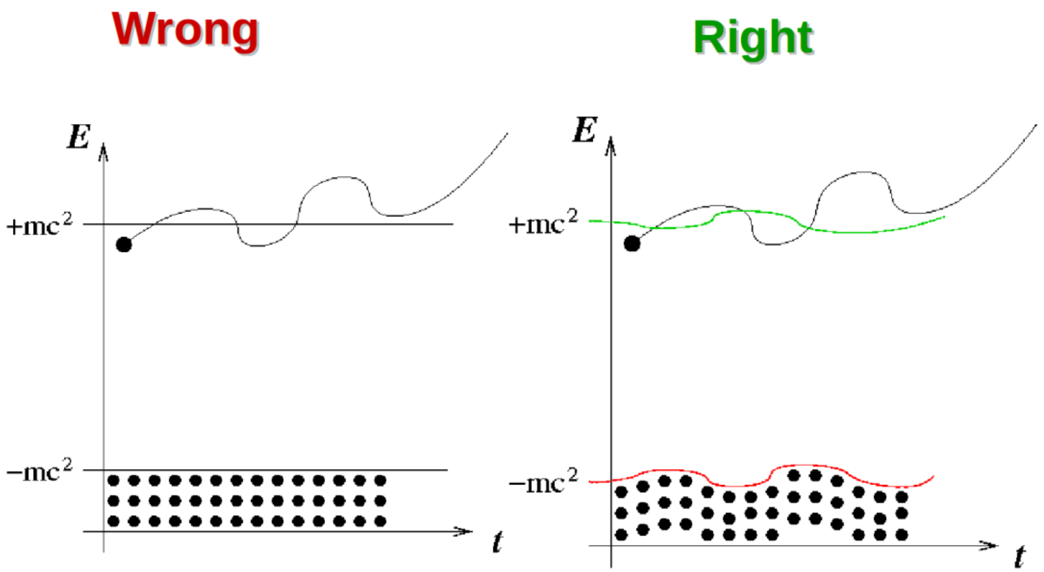
$$\beta mc^2$$

Problem 1: Stiffness

$$\Psi(t+\Delta t) = U(t+\Delta t, t)\Psi(t) + \mathcal{O}(\Psi^{(n)}(t)\Delta t^n)$$

$$\Psi^{(n)}\Delta t^n \sim H^n \Psi(t)\Delta t^n$$

$$\Delta t \ll \frac{1}{2mc^2} \quad (\sim 10^{-5}\ \text{a.u.})$$



Selstø, Lindroth, Bengtsson, Phys. Rev. A **79**, 043418 (2009)

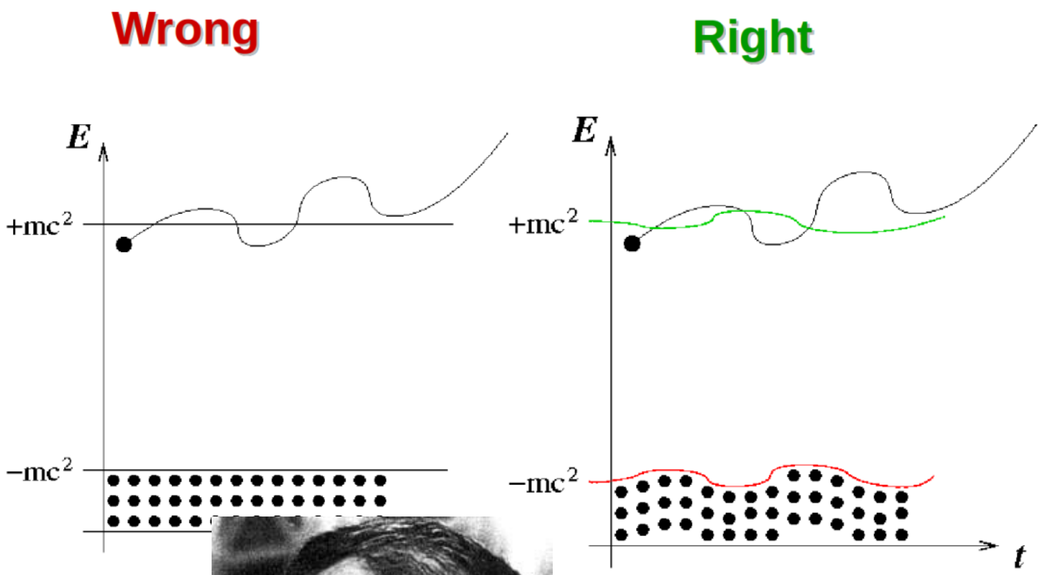
Vanne, Saenz, Phys. Rev. A **85**, 033411 (2012)

Problem 1: Stiffness

$$\Psi(t+\Delta t) = U(t+\Delta t, t)\Psi(t) + \mathcal{O}(\Psi^{(n)}(t)\Delta t^n)$$

$$\Psi^{(n)}\Delta t^n \sim H^n \Psi(t) \Delta t^n$$

$$\Delta t \ll \frac{1}{2mc^2} \quad (\sim 10^{-5}\ \text{a.u.})$$



Solution: Magnus propagator

$$U(t+\Delta t, t) = e^{-i\bar{H}\Delta t}$$


Wilhelm Magnus

Hochbruck, Lubich, SIAM J. Numer. Anal. **41**, 945 (2003)

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad\ \ \text{otherwise} \end{cases}$$

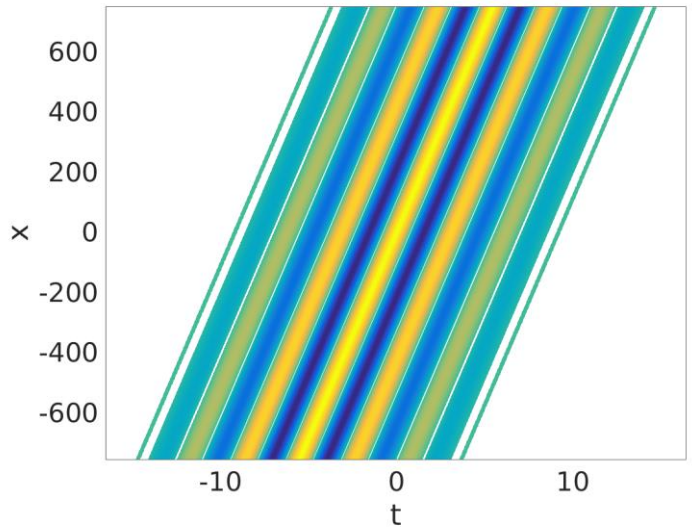
$$\eta = \omega t - kx, \qquad k = \omega / c$$

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$



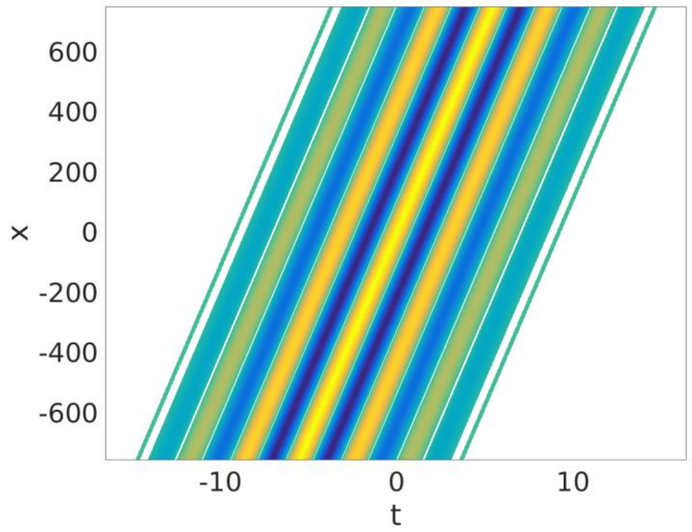
Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

Calculate couplings at each time step?



Problem 2: Including the spatial dependence of the field

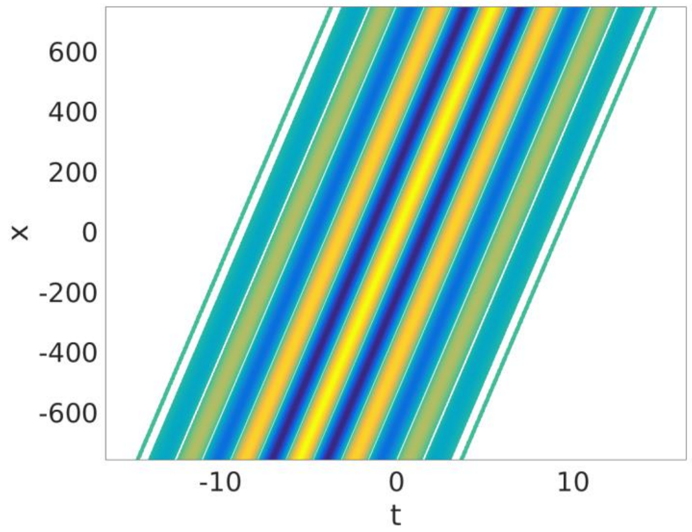
$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

Calculate couplings at each time step?

-Too time consuming



Has been implemented, though:
Ivanov, Phys. Rev. A **91**, 043410 (2015)

Problem 2: Including the spatial dependence of the field

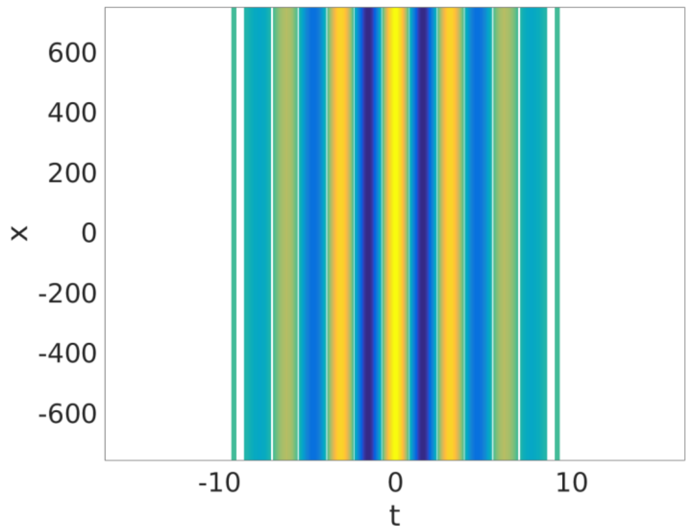
$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

Simply disregard the spatial dependence?

$$k \approx 0, \qquad \eta \approx \omega t, \qquad \mathbf{B} = \nabla \times \mathbf{A} = \mathbf{0}$$

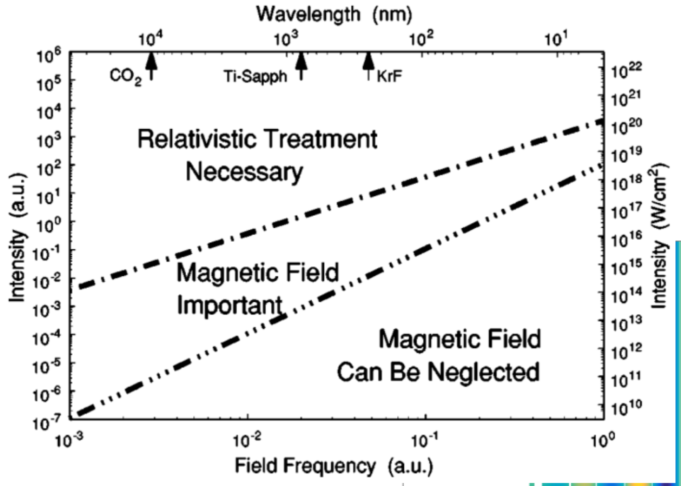


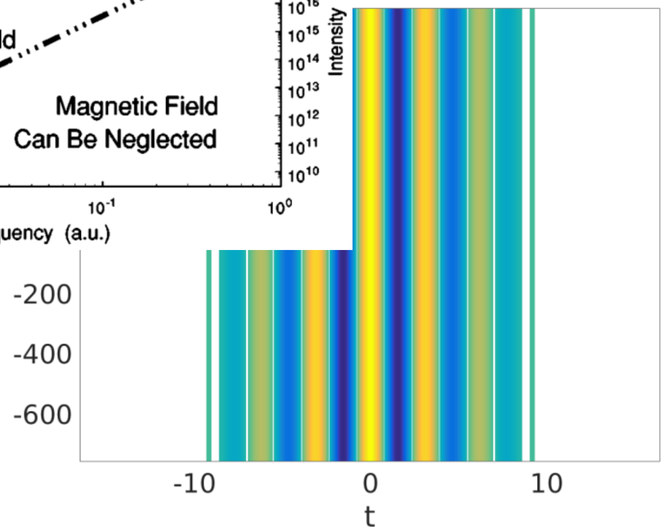
Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$





Simply disregard the spatial dependence?

$$k \approx 0, \qquad \eta \approx \omega t, \qquad \mathbf{B} = \nabla \times \mathbf{A} = \mathbf{0}$$

-Dipole approximation. Not valid.

Reiss, Phys. Rev. A **63**, 013409 (2000).

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

Separate time and space somehow

$$A(t, x) = \sum_n a_n T_n(t) X_n(x)$$

$$\langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle = \sum_n a_n T_n(t) \langle \varphi_k | X_n(x) | \varphi_l \rangle$$

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

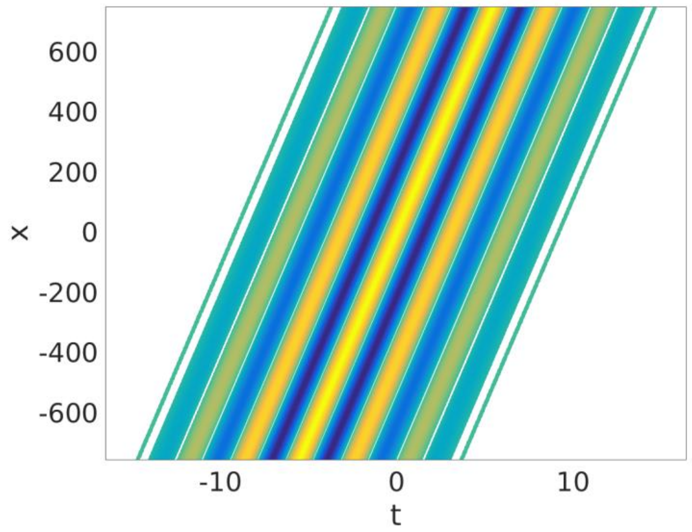
$$\eta = \omega t - kx, \qquad k = \omega / c$$

Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n(x)$$

Fourier:

$$A(t,x) = \sum_n a_n e^{i\frac{2\pi}{P}\eta} = \sum_n a_n e^{i\frac{2\pi}{P}\omega t} \cdot e^{-i\frac{2\pi}{P}kx}$$



Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \leq \eta \leq \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

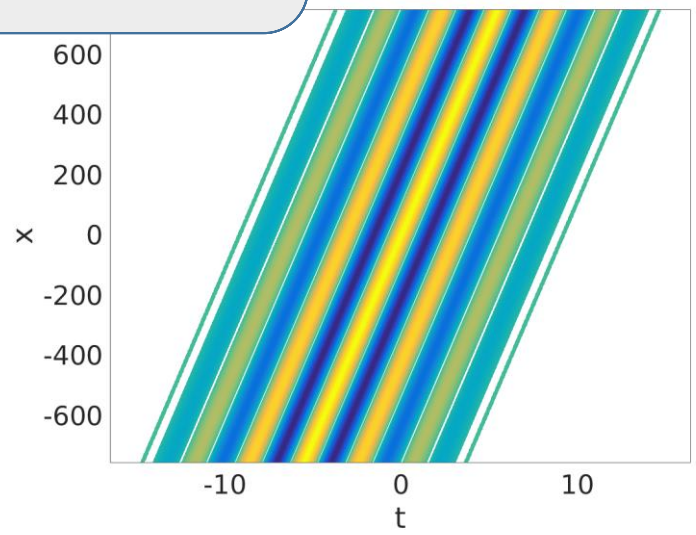
Period *T*: 6 terms

Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n(x)$$

Fourier:

$$A(t,x) = \sum_n a_n e^{i\frac{2\pi}{P}\eta} = \sum_n a_n e^{i\frac{2\pi}{P}\omega t} \cdot e^{-i\frac{2\pi}{P}kx}$$



Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) \;, \quad \text{everywhere/always}$$

$$\eta = \omega t - kx, \quad k = \omega / c$$

Period T: 6 terms

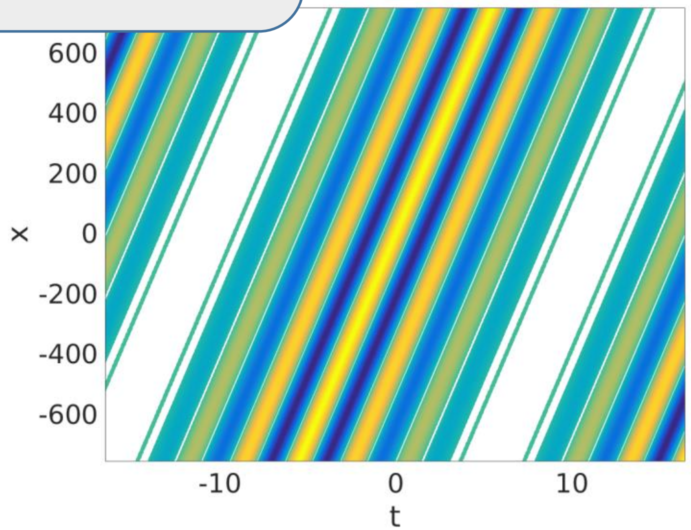
Maybe ok for longer pulses

Separate time and space somehow

$$A(t, x) = \sum_n a_n T_n(t) X_n(x)$$

Fourier:

$$A(t, x) = \sum_n a_n e^{i\frac{2\pi}{P}\eta} = \sum_n a_n e^{i\frac{2\pi}{P}\omega t} \cdot e^{-i\frac{2\pi}{P}kx}$$



Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A \approx \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{T} t\right) \sin(\eta + \varphi) & , \quad 0 \leq t \leq T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

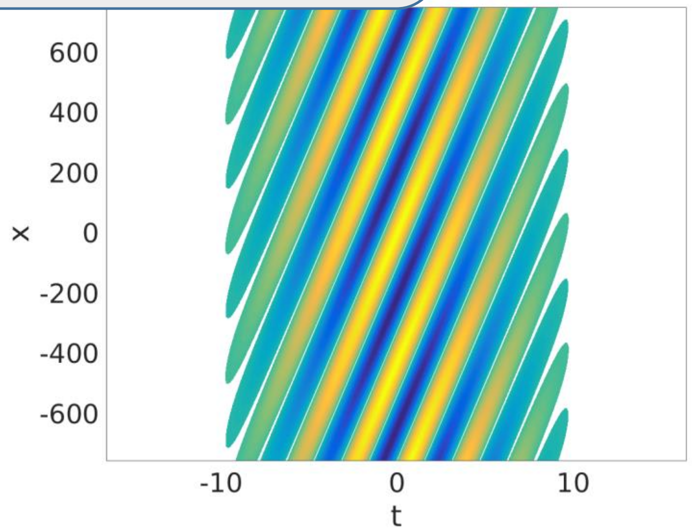
Neglect *x* in envelope

Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n(x)$$

Fourier:

$$A(t,x) = \sum_n a_n e^{i\frac{2\pi}{P}\eta} = \sum_n a_n e^{i\frac{2\pi}{P}\omega t} \cdot e^{-i\frac{2\pi}{P}kx}$$



Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A \approx \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{T} t\right) \sin(\eta + \varphi) & , \quad 0 \leq t \leq T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

Neglect *x* in envelope

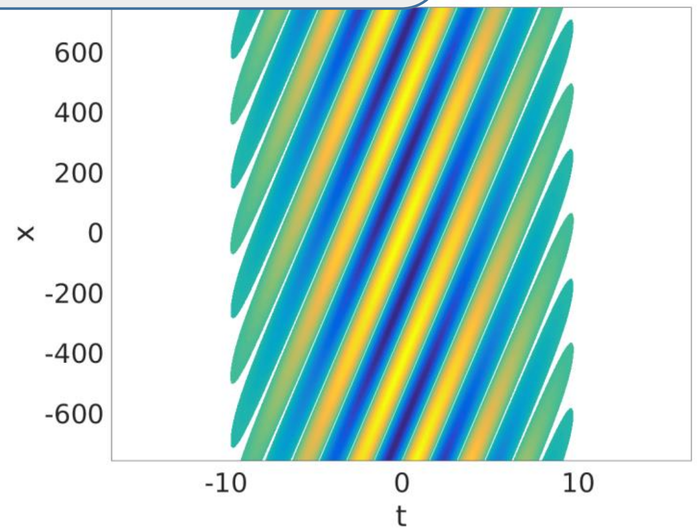
Actually, it's the contrary

Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n(x)$$

Fourier:

$$A(t,x) = \sum_n a_n e^{i\frac{2\pi}{P}\eta} = \sum_n a_n e^{i\frac{2\pi}{P}\omega t} \cdot e^{-i\frac{2\pi}{P}kx}$$



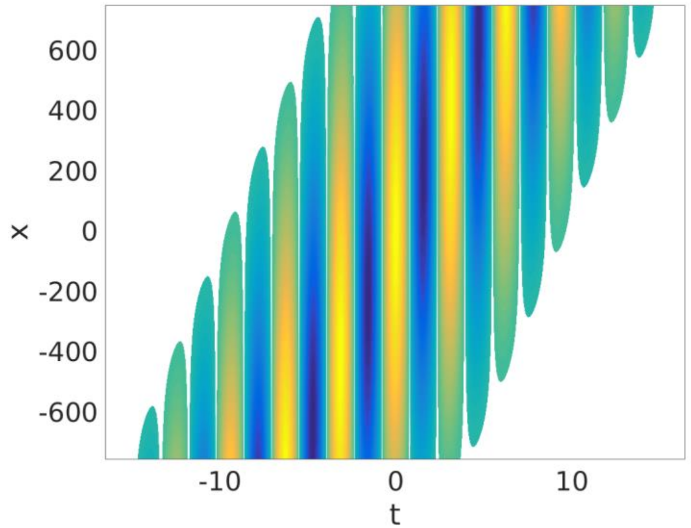
Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A \approx \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\omega t + \varphi) & , \quad 0 \leq \eta \leq \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

The envelope approximation

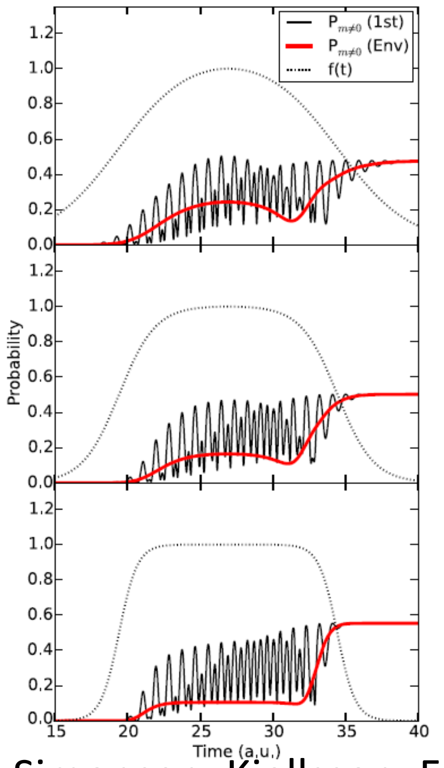


Problem 2: Including the spatial dependence of the field

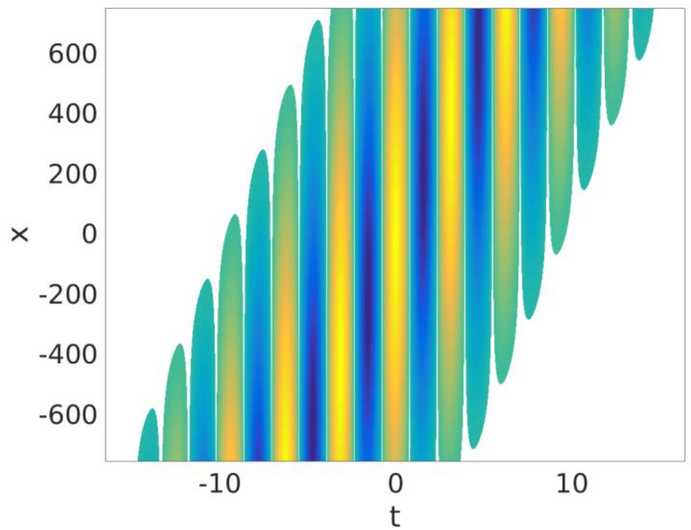
$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A \approx \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\omega t + \varphi) & , \quad 0 \leq \eta \leq \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

The envelope approximation



Population in $m \neq 0$-states within and without the envelope approximation

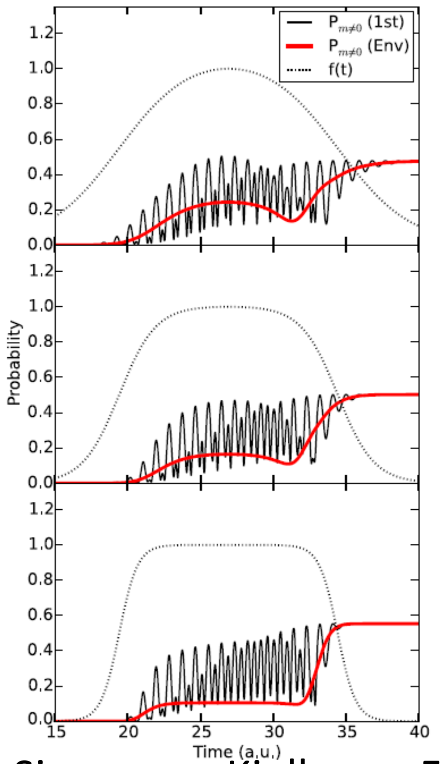


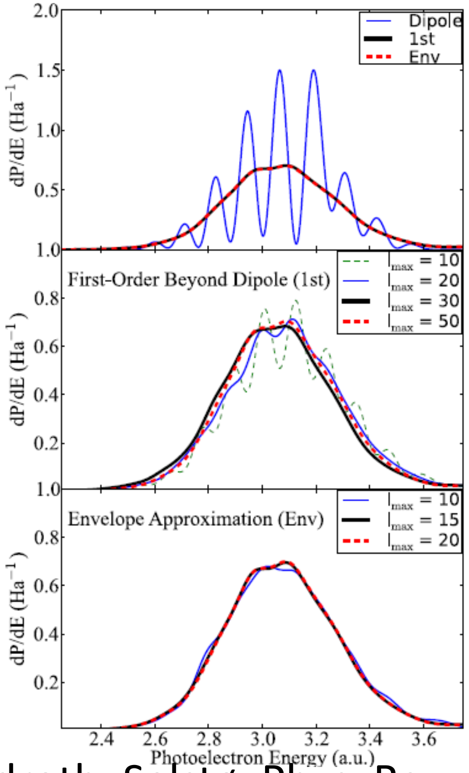
Simonsen, Kjellsson, Førre, Lindroth, Selstø, Phys. Rev. A **93**, 053411 (2016)

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

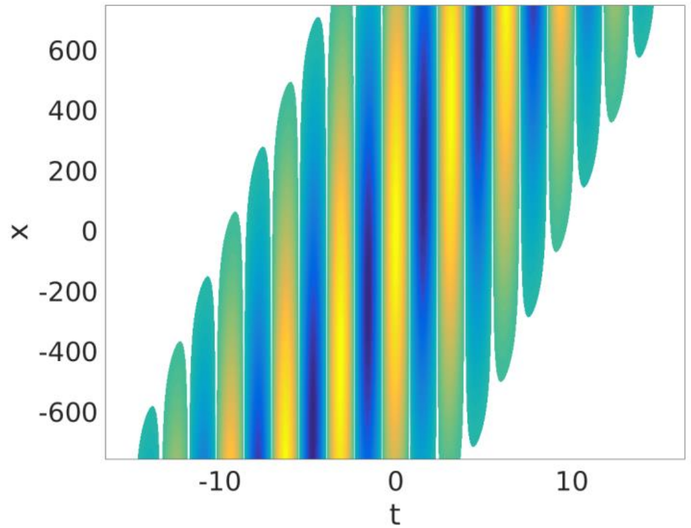
$$A \approx \begin{cases} \dfrac{E_0}{\omega} \sin^2\left(\dfrac{\pi}{\omega T}\eta\right) \sin(\omega t + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \text{otherwise} \end{cases}$$





The envelope approximation

Photoelectron spectrum



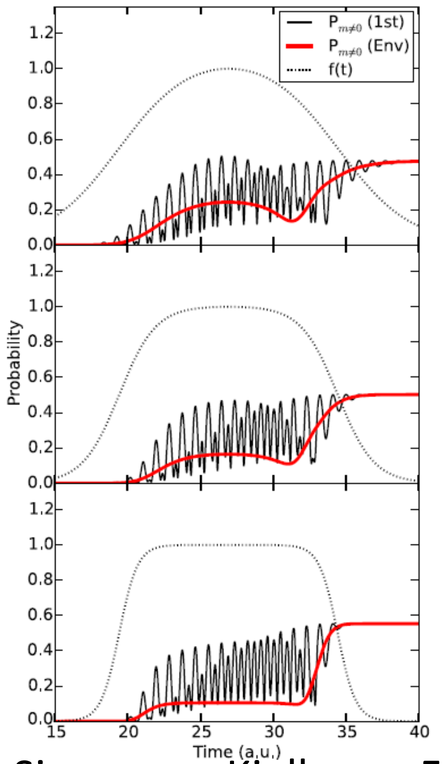
Simonsen, Kjellsson, Førre, Lindroth, Selstø, Phys. Rev. A **93**, 053411 (2016)

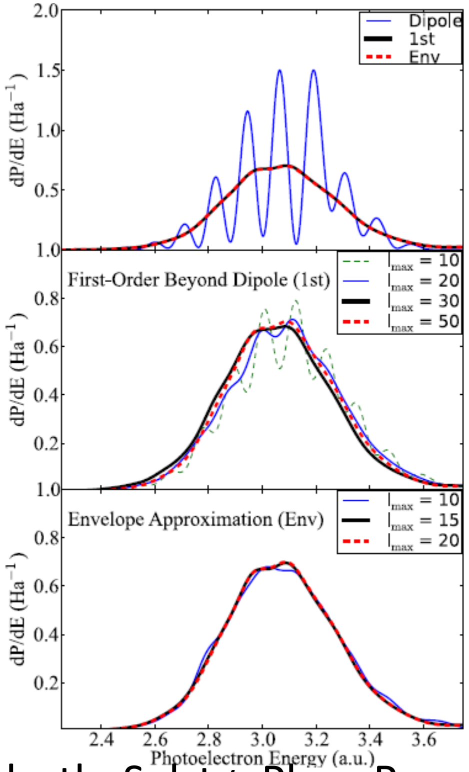
Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A \approx \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\omega t + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

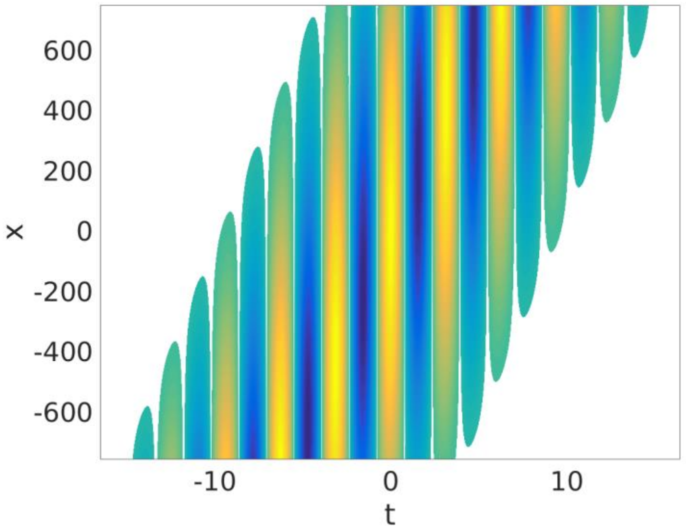
The envelope approximation





Photoelectron spectrum

Results from the Schrödinger equation

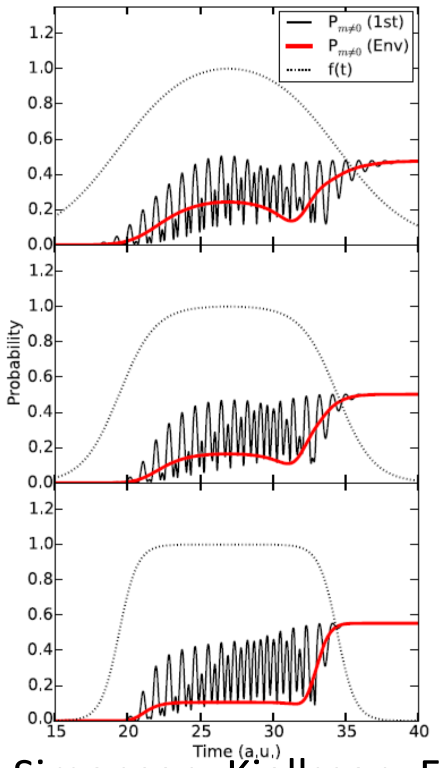


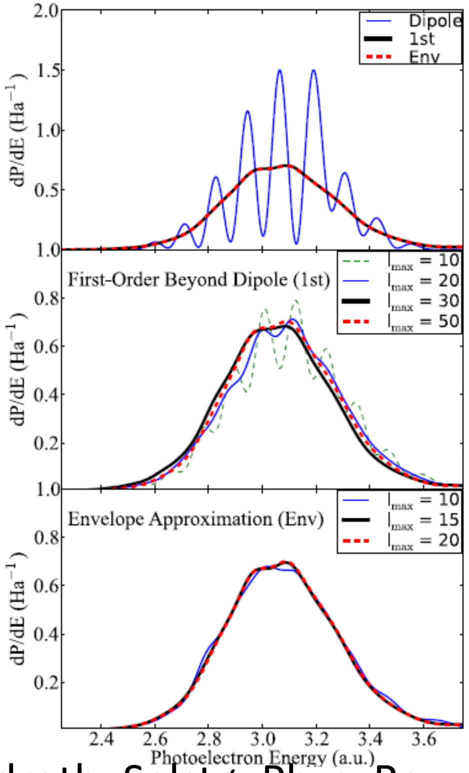
Simonsen, Kjellsson, Førre, Lindroth, Selstø, Phys. Rev. A **93**, 053411 (2016)

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A \approx \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\omega t + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \text{otherwise} \end{cases}$$



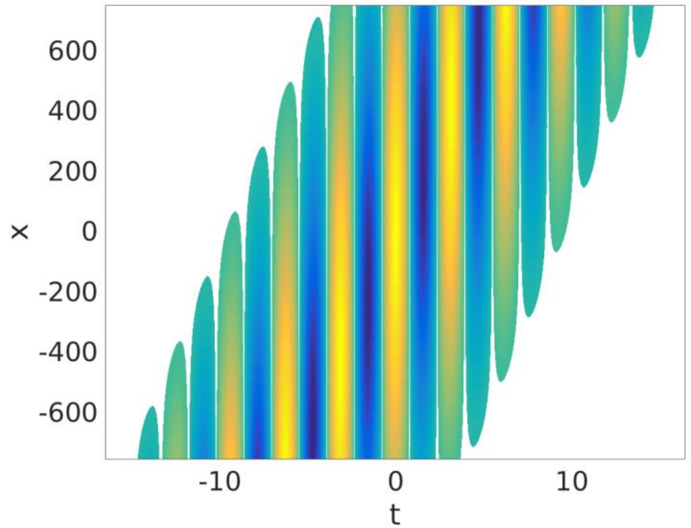


The envelope approximation

Alas: Dubious in the relativistic regime

Photoelectron spectrum

Results from the Schrödinger equation



Simonsen, Kjellsson, Førre, Lindroth, Selstø, Phys. Rev. A **93**, 053411 (2016)

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \leq \eta \leq \omega T \\ 0 & \text{otherwise} \end{cases}$$

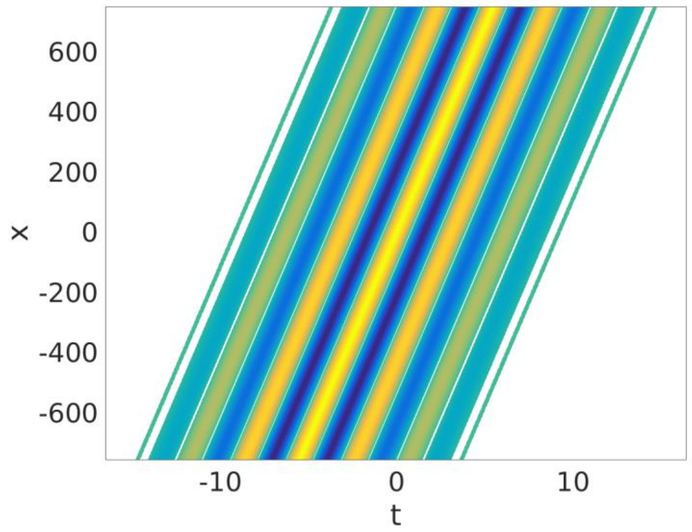
$$\eta = \omega t - kx, \qquad k = \omega / c$$

Separate time and space somehow

$$A(t, x) = \sum_n a_n T_n(t) X_n(x)$$

Taylor:

$$A(\eta) = \sum_n \frac{1}{n!} A^{(n)}(\eta)\Big|_{x=0} x^n$$



Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \leq \eta \leq \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

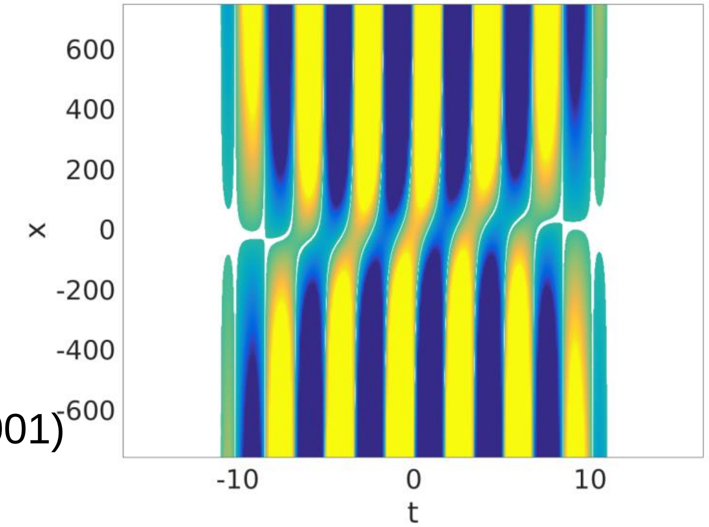
Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n(x)$$

Taylor:

$$A \approx A(t) - A'(t) x / c$$

First order



Vázquez de Aldana, Kylstra, Roso, Knight, Patel, Worthington, Phys. Rev. A **64**, 013411 (2001)
Førre, Simonsen, Phys. Rev. A **90**, 053411 (2014)

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \text{otherwise} \end{cases}$$

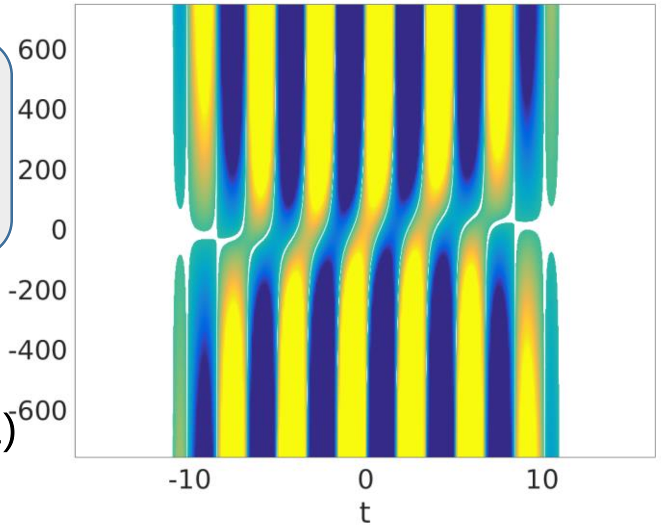
$$\eta = \omega t - kx, \qquad k = \omega / c$$

Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n($$

Sufficient for the Schrödinger equation

First order



Taylor:

$$A \approx A(t) - A'(t)x/c$$

Vázquez de Aldana, Kylstra, Roso, Knight, Patel, Worthington, Phys. Rev. A **64**, 013411 (2001)

Førre, Simonsen, Phys. Rev. A **90**, 053411 (2014)

Problem 2: Including the spatial dependence of the field

$$H'_{kl} = \langle \varphi_k | c\boldsymbol{\alpha} \cdot e\mathbf{A} | \varphi_l \rangle$$

$$A = \begin{cases} \frac{E_0}{\omega} \sin^2\left(\frac{\pi}{\omega T}\eta\right) \sin(\eta + \varphi) & , \quad 0 \le \eta \le \omega T \\ 0 & \quad \text{otherwise} \end{cases}$$

$$\eta = \omega t - kx, \qquad k = \omega / c$$

Separate time and space somehow

$$A(t,x) = \sum_n a_n T_n(t) X_n($$

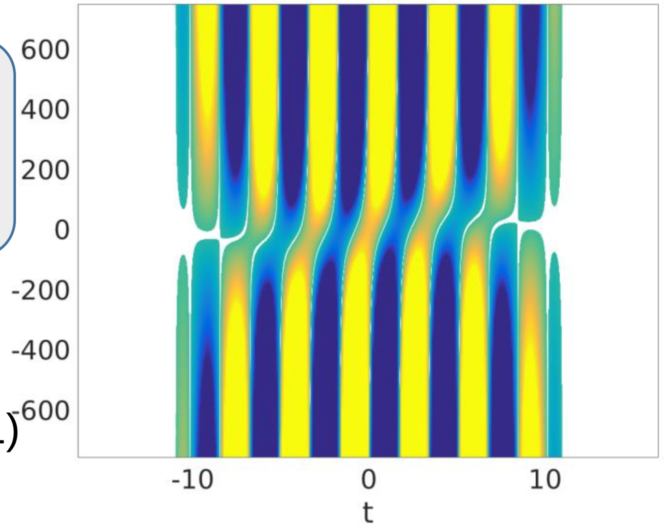
Taylor:

$$A \approx A(t) - A'(t) x / c$$

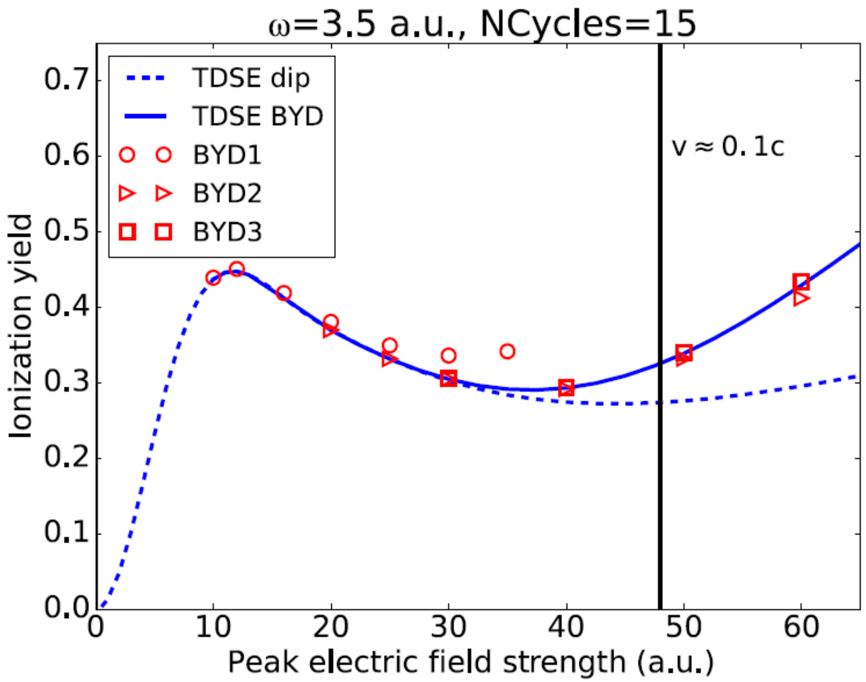
Sufficient for the Schrödinger equation

Completely wrong for the Dirac equation!

First order

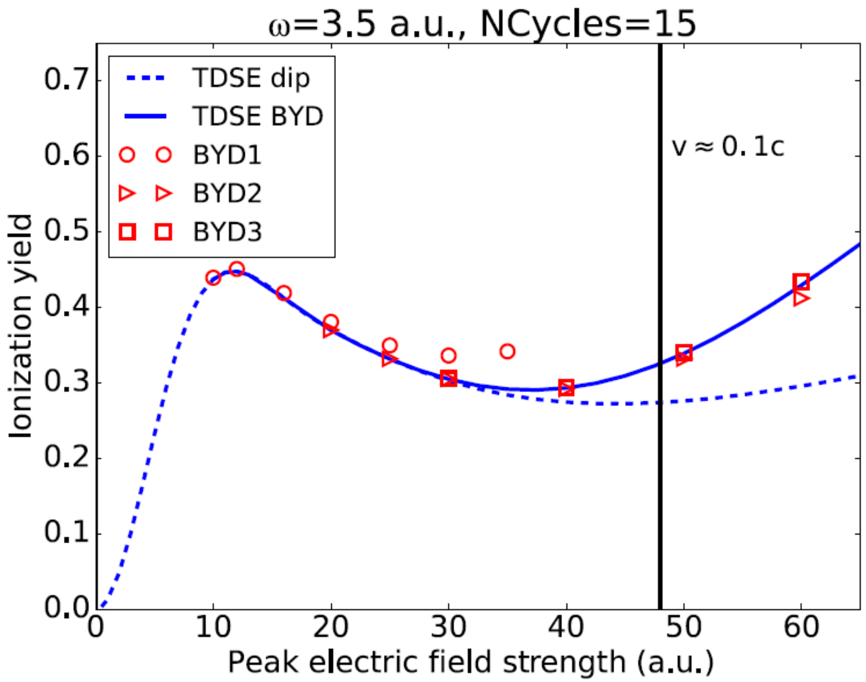


Vázquez de Aldana, Kylstra, Roso, Knight, Patel, Worthington, Phys. Rev. A **64**, 013411 (2001)
Førre, Simonsen, Phys. Rev. A **90**, 053411 (2014)



1st order: Actually worse than the dipole approximation!

Why?



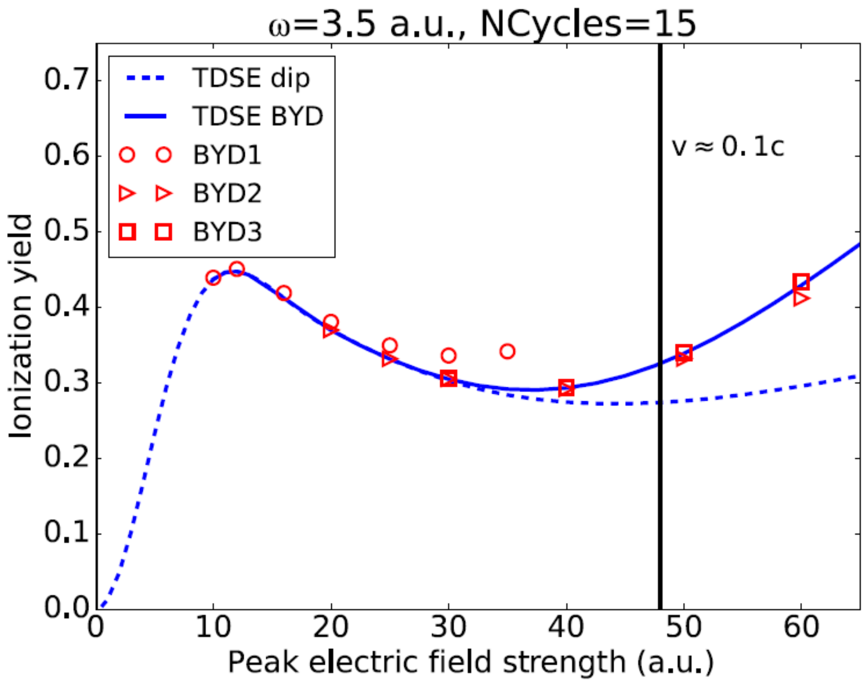
1st order: Actually worse than the dipole approximation!

Why?

Large component

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\ X\end{pmatrix}=\begin{pmatrix}V+mc^2 & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & V-mc^2\end{pmatrix}\begin{pmatrix}\Phi\\ X\end{pmatrix}$$

Small component

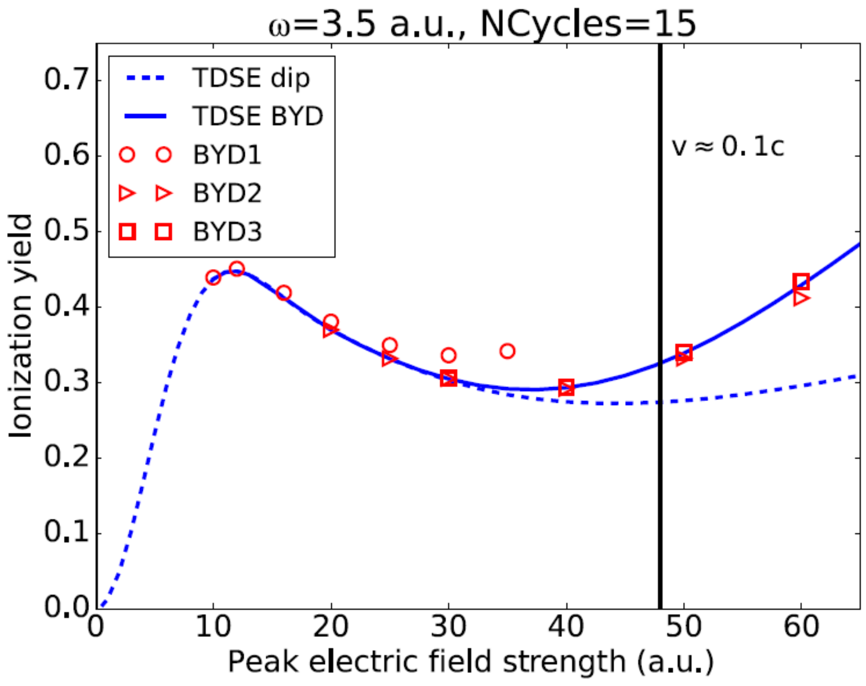


1st order: Actually worse than the dipole approximation!

Why?

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\X\end{pmatrix}=\begin{pmatrix}V+mc^2 & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & V-mc^2\end{pmatrix}\begin{pmatrix}\Phi\\X\end{pmatrix}$$

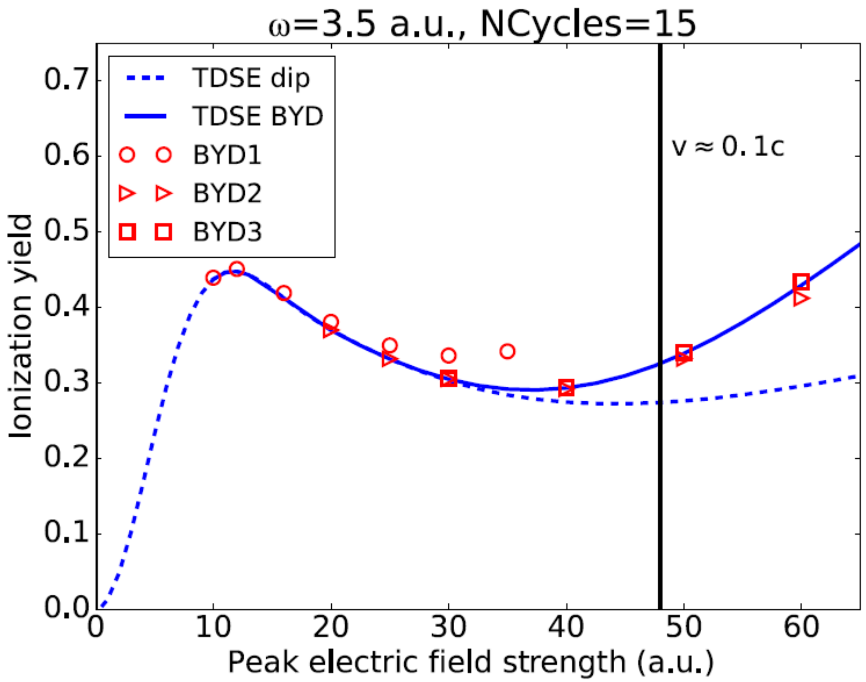
$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\X\end{pmatrix}=\begin{pmatrix}V & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & V-2mc^2\end{pmatrix}\begin{pmatrix}\Phi\\X\end{pmatrix}$$



1st order: Actually worse than the dipole approximation!

Why?

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\X\end{pmatrix}=\begin{pmatrix}V & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & V-2mc^2\end{pmatrix}\begin{pmatrix}\Phi\\X\end{pmatrix}$$

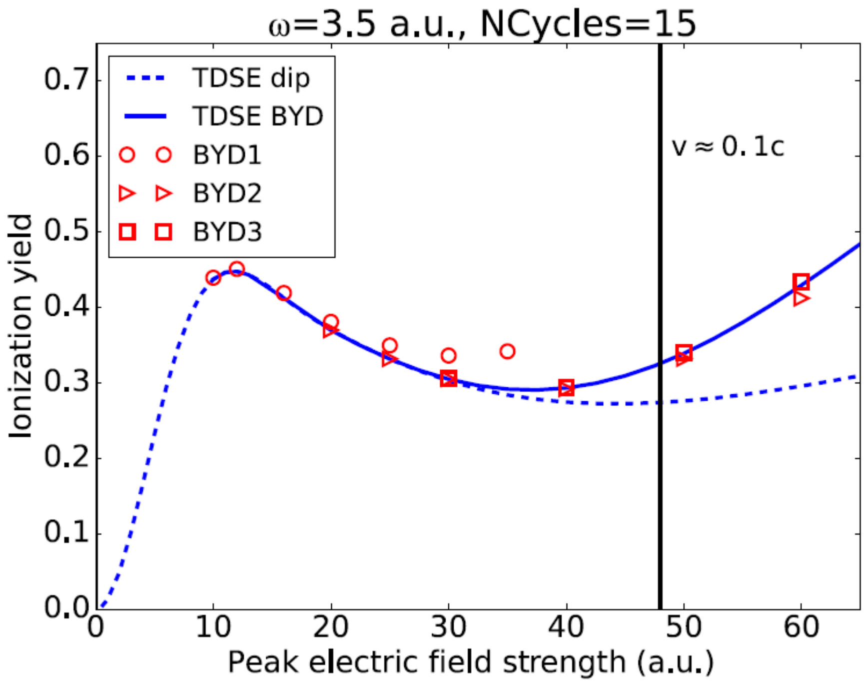


1st order: Actually worse than the dipole approximation!

Why?

$$i\hbar\frac{d}{dt}\begin{pmatrix} \Phi \\ X \end{pmatrix} = \begin{pmatrix} V & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) \\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & V-2mc^2 \end{pmatrix}\begin{pmatrix} \Phi \\ X \end{pmatrix}$$

$$i\hbar\frac{d}{dt}\begin{pmatrix} \Phi \\ 0 \end{pmatrix} \approx \begin{pmatrix} V & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) \\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & -2mc^2 \end{pmatrix}\begin{pmatrix} \Phi \\ X \end{pmatrix}$$

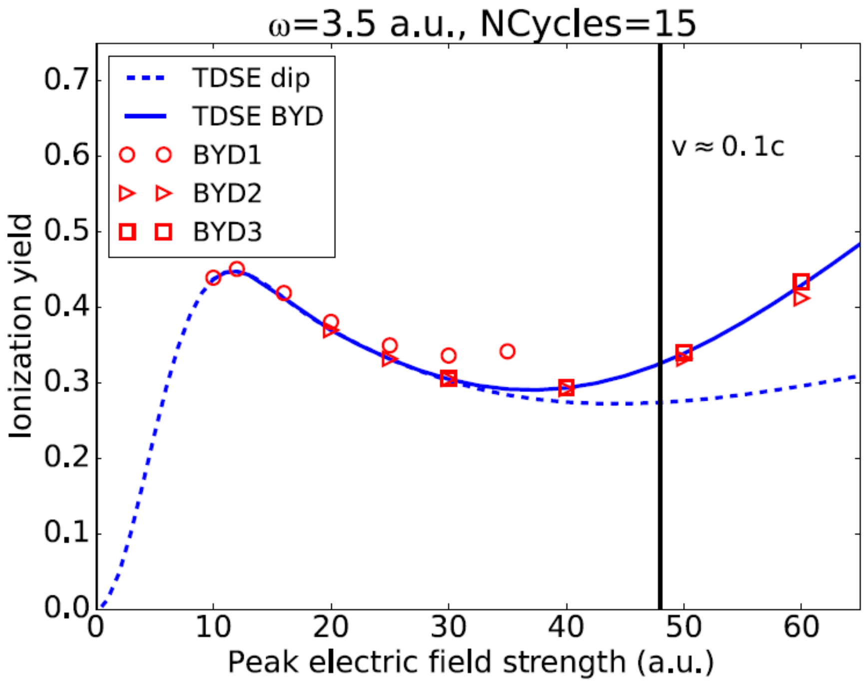


1st order: Actually worse than the dipole approximation!

Why?

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\0\end{pmatrix}\approx\begin{pmatrix}V & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & -2mc^2\end{pmatrix}\begin{pmatrix}\Phi\\X\end{pmatrix}$$

$$X\approx\frac{1}{2mc^2}c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\Phi$$



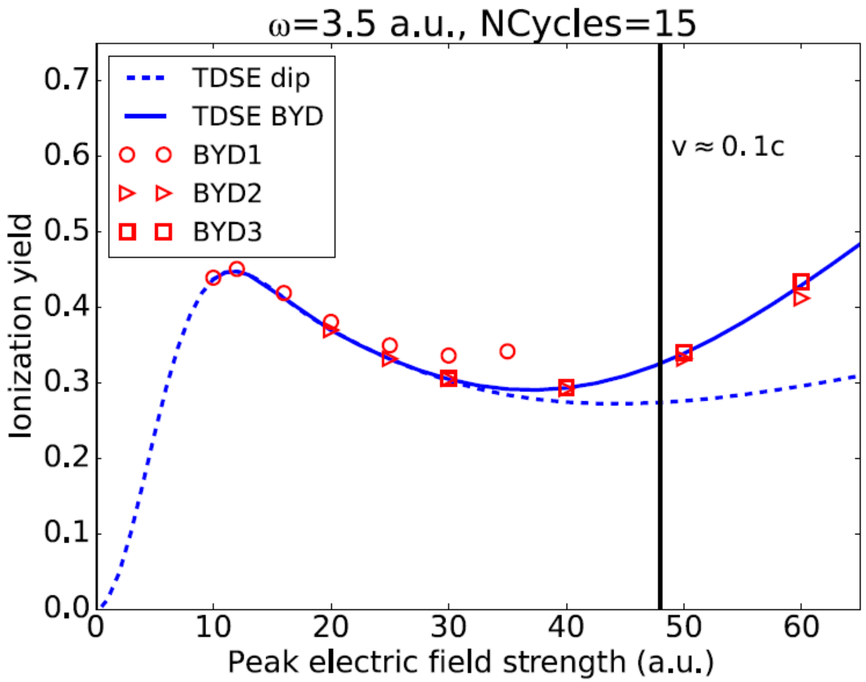
1st order: Actually worse than the dipole approximation!

Why?

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi\\0\end{pmatrix}\approx\begin{pmatrix}V & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & -2mc^2\end{pmatrix}\begin{pmatrix}\Phi\\X\end{pmatrix}$$

$$X\approx\frac{1}{2mc^2}c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\Phi$$

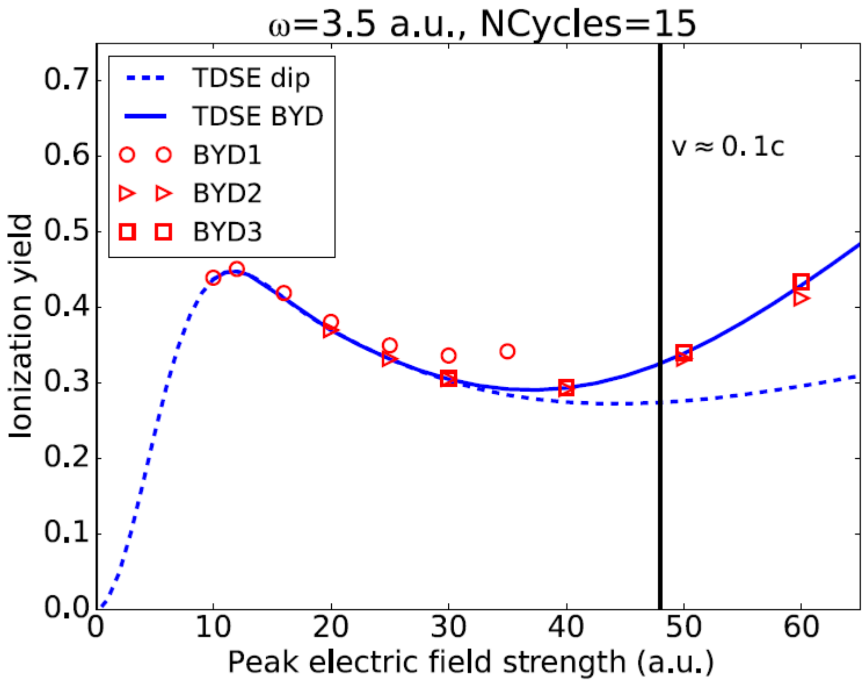
$$i\hbar\frac{d}{dt}\Phi\approx V\Phi+\frac{1}{2mc^2}\left(c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A})\right)^2\Phi$$



1st order: Actually worse than the dipole approximation!

Why?

$$i\hbar\frac{d}{dt}\Phi = \left(\frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m}A^2 + \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)\Phi$$



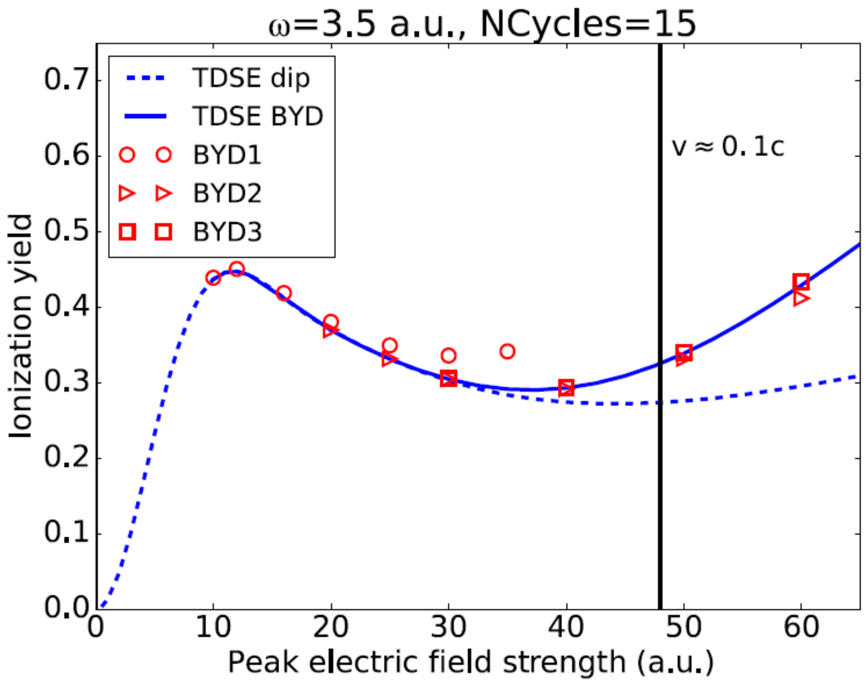
1st order: Actually worse than the dipole approximation!

Why?

$$A \approx A(t) - A'(t)x/c$$

$$i\hbar\frac{d}{dt}\begin{pmatrix}\Phi \\ X\end{pmatrix} = \begin{pmatrix} V + mc^2 & c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) \\ c\boldsymbol{\sigma}\cdot(\mathbf{p}+e\mathbf{A}) & V - mc^2 \end{pmatrix}\begin{pmatrix}\Phi \\ X\end{pmatrix}$$

$$i\hbar\frac{d}{dt}\Phi = \left(\frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m}A^2 + \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)\Phi$$

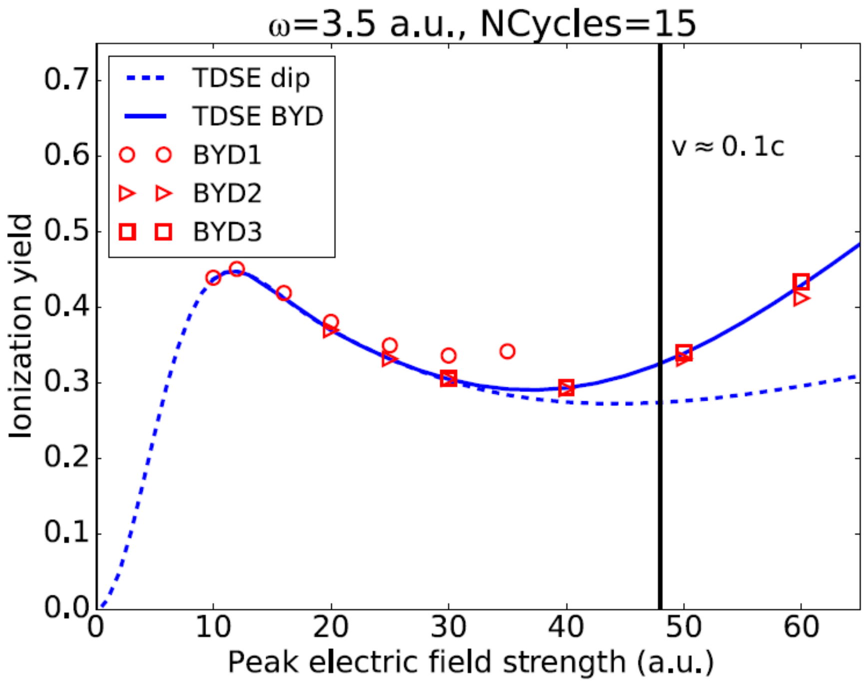


1st order: Actually worse than the dipole approximation!

Why?

$$A^2 \approx (A(t) + A'(t)\,x/c)^2 = A^2 + 2AA'\,x/c + (A')^2\,x^2/c^2$$

$$i\hbar\frac{d}{dt}\Phi = \left(\frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m}A^2 + \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)\Phi$$



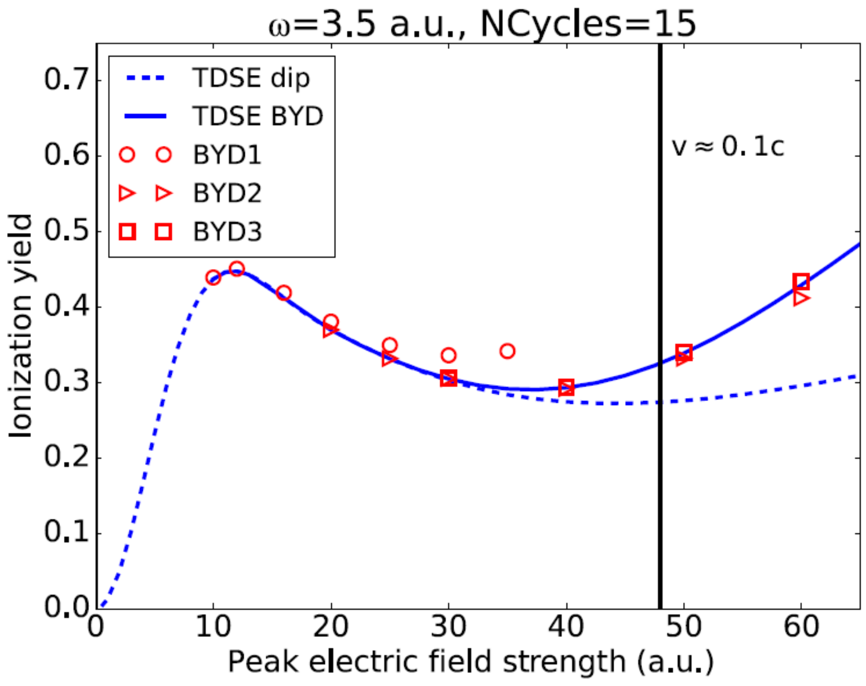
1st order: Actually worse than the dipole approximation!

Why?

$$A^2 \approx (A(t) + A'(t)\, x/c)^2 = A^2 + 2AA'\, x/c + (A')^2\, x^2/c^2$$

$$A^2 \approx A(t) + 2AA'\, x/c + \frac{1}{2}\, 2((A')^2 + AA'')\, x^2/c^2$$

$$i\hbar \frac{d}{dt}\Phi = \left(\frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m}A^2 + \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B} \right)\Phi$$



1st order: Actually worse than the dipole approximation!

Why?

$$A^2 \approx (A(t) + A'(t)\,x/c)^2 = A^2 + 2AA'\,x/c + (A')^2\,x^2/c^2$$

$$A^2 \approx A(t) + 2AA'\,x/c + \frac{1}{2}2((A')^2 + AA'')\,x^2/c^2$$

$$i\hbar\frac{d}{dt}\Phi = \left(\frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m}A^2 + \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)\Phi$$

«Solution»: Add x^n-terms in A in the Dirac equation until it works

Problem 3: How do we deal with such large basis sets?

For convergence:
l_{max}= 30 (# partial waves)

Spectral basis: 500 positive and 500 negative energies per spin-angular symmetry (filter out the highest ones)

In total: ~ 2 million states
With up to x^5: ~ $4 \cdot 10^{11}$ non-zero matrix elements (3 TB)

Problem 3: How do we deal with such large basis sets?

Subproblem 3a: How can we exponentiate such a huge matrix?

Subproblem 3b: How can we fit such a huge matrix in the memory?

Problem 3: How do we deal with such large basis sets?

Subproblem 3a: How can we exponentiate such a huge matrix?

$$\Psi(t + \Delta t) \approx e^{-i\bar{H}\Delta t}\Psi(t)$$

Problem 3: How do we deal with such large basis sets?

Subproblem 3a: How can we exponentiate such a huge matrix?

$$\Psi(t+\Delta t) \approx e^{-i\bar{H}\Delta t}\Psi(t)$$

Solution: Krylov subspaces

$$\mathcal{K}_n(t) = \mathrm{Span}\{\Psi, \bar{H}\Psi, \bar{H}^2\Psi, \ldots, \bar{H}^n\Psi\}$$

-We exponentiate within this space, and then transform back

Aleksej Krylov

Arnoldi-method

Problem 3: How do we deal with such large basis sets?

Subproblem 3b: How can we fit such a huge matrix in the memory?

Problem 3: How do we deal with such large basis sets?

Subproblem 3b: How can fit such a huge matrix in the memory?

Solution: The Wigner-Eckhart theorem

Keep only radial part in memory, calculate spin-angular part of the coupling on the fly using the Wigner-Eckhart theorem.

Carl Eckhart

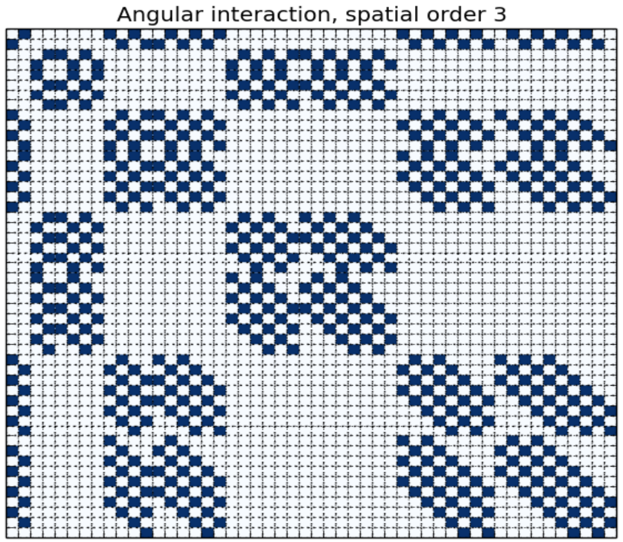
Eugene Wigner

Problem 3: How do we deal with such large basis sets?

Subproblem 3b: How can fit such a huge matrix in the memory?

Solution: The Wigner-Eckhart theorem

Keep only radial part in memory, calculate spin-angular part of the coupling on the fly using the Wigner-Eckhart theorem.



Carl Eckhart

Eugene Wigner

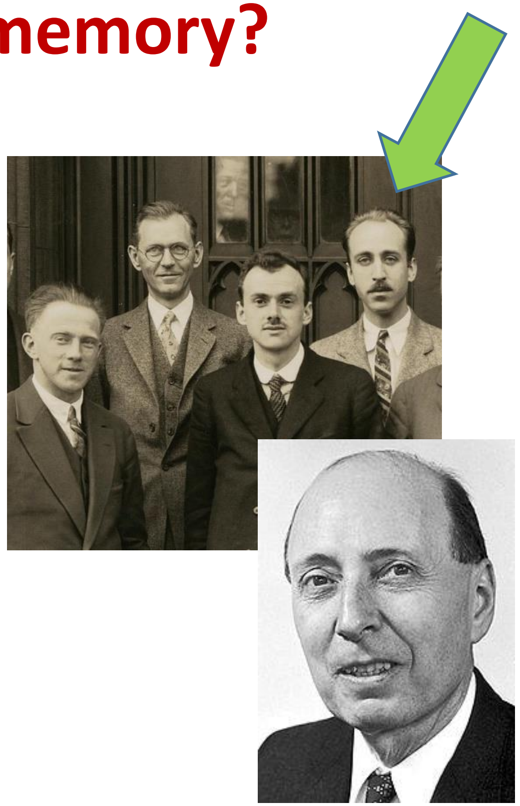
Problem 3: How do we deal with such large basis sets?

Subproblem 3b: How can fit such a huge matrix in the memory?

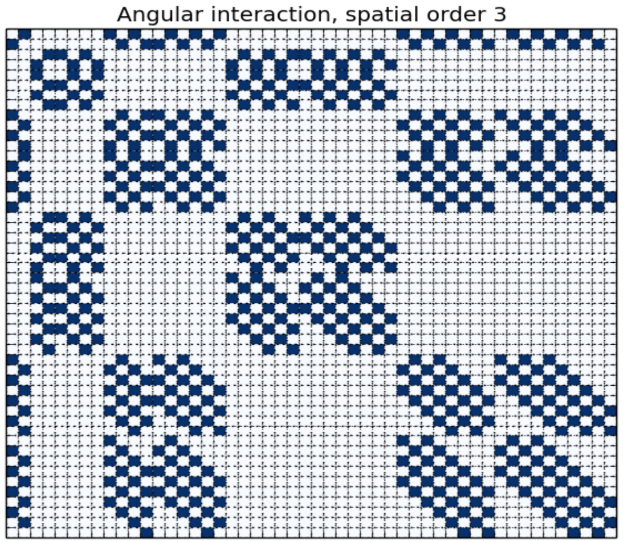
Solution: The Wigner-Eckhart theorem

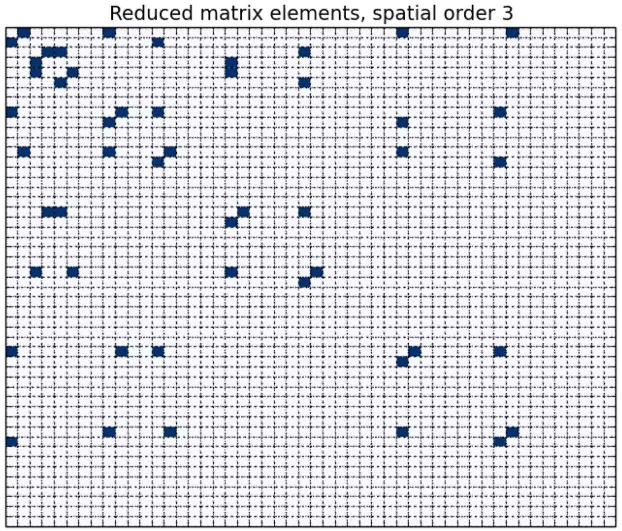
Keep only radial part in memory, calculate spin-angular part of the coupling on the fly using the Wigner-Eckhart theorem.

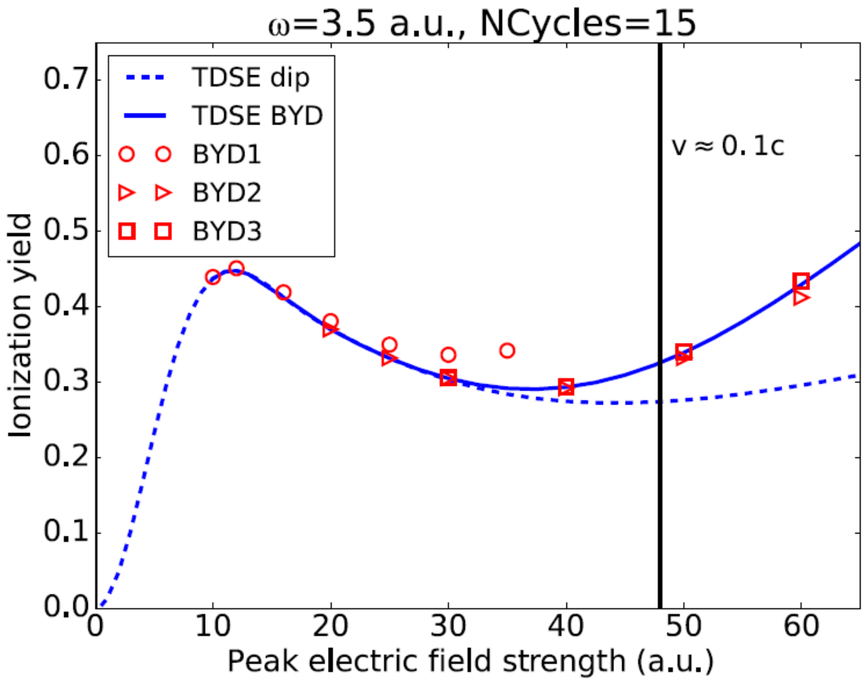
Carl Eckhart



Eugene Wigner

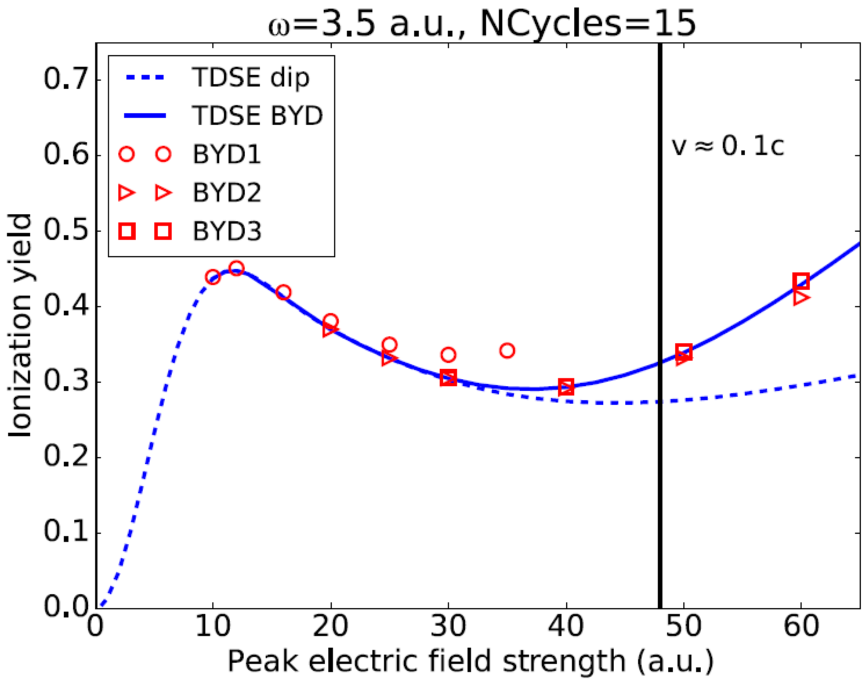




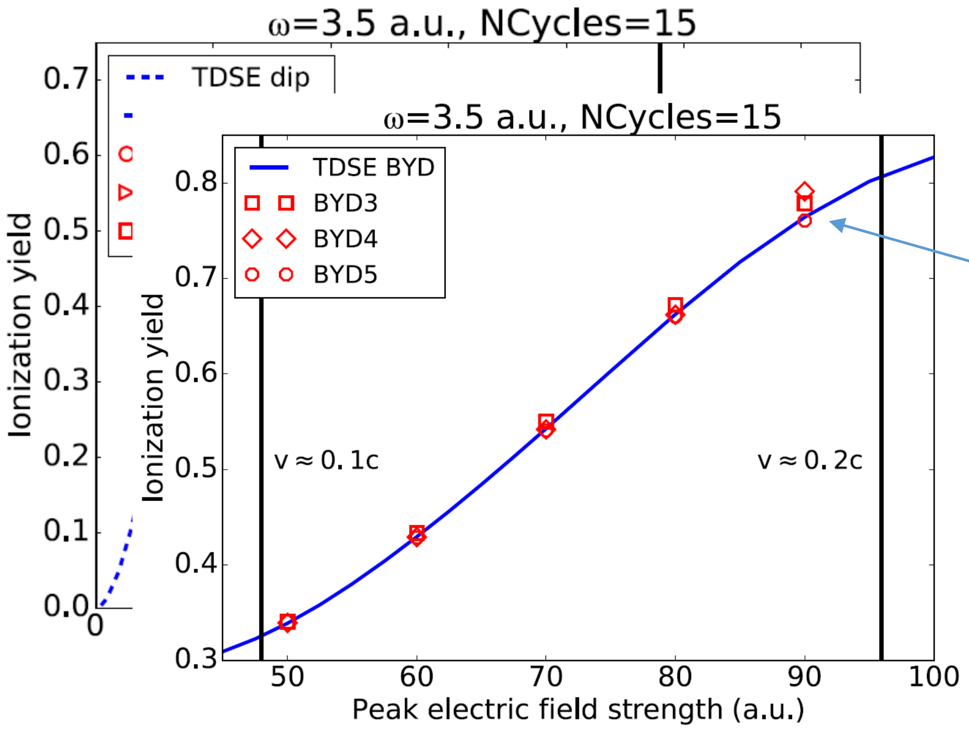


ω=3.5 a.u., NCycles=15
TDSE dip
TDSE BYD
BYD1
BYD2
BYD3
v ≈ 0.1c
Ionization yield
Peak electric field strength (a.u.)
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0
10
20
30
40
50
60

Problem 4: Weak convergence in x^n

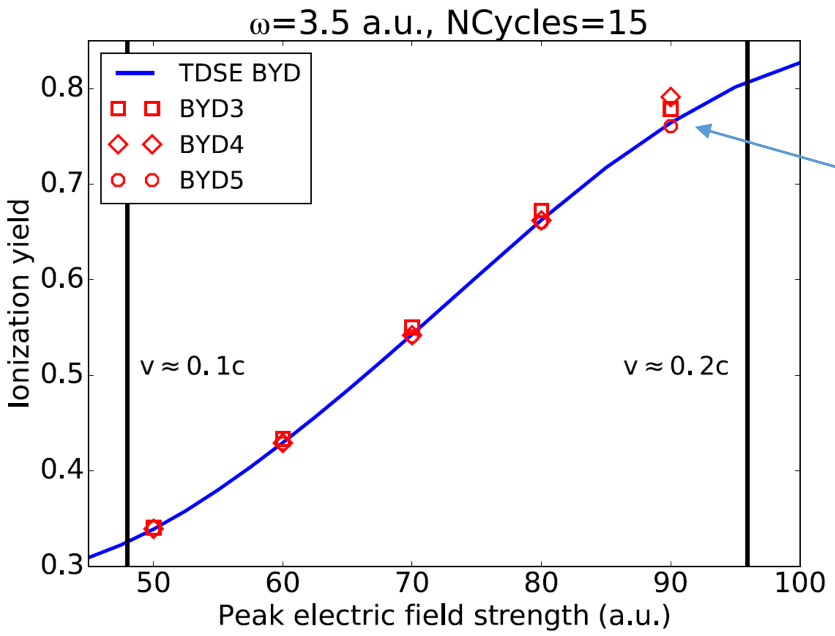


Problem 4: Weak convergence in x^n



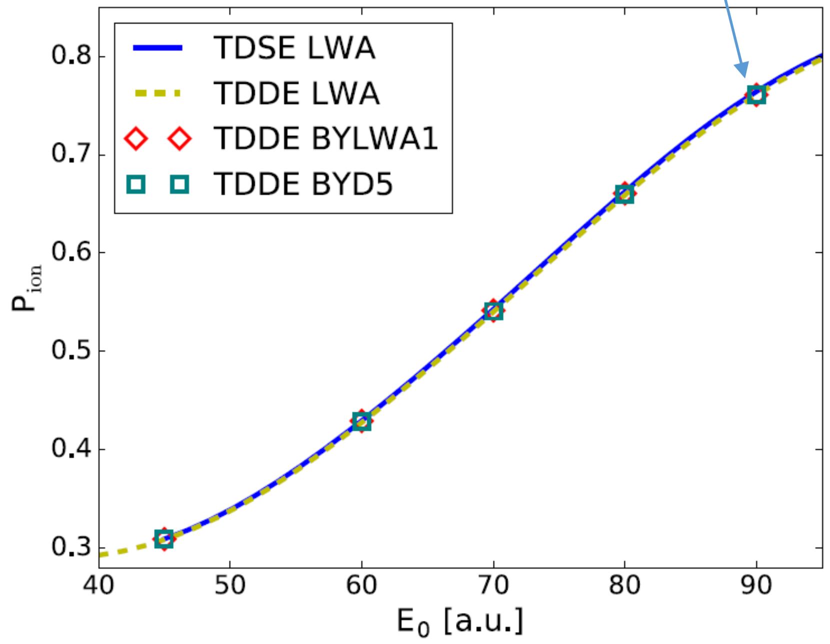
Up to x^5 in the interaction

Problem 4: Weak convergence in x^n

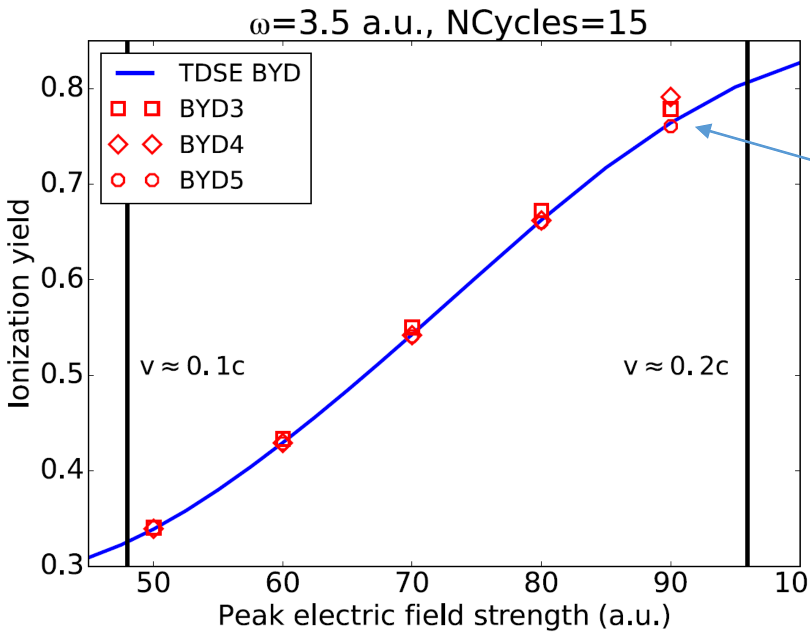


Up to x^5 in the interaction

Up to x^0 in the interaction

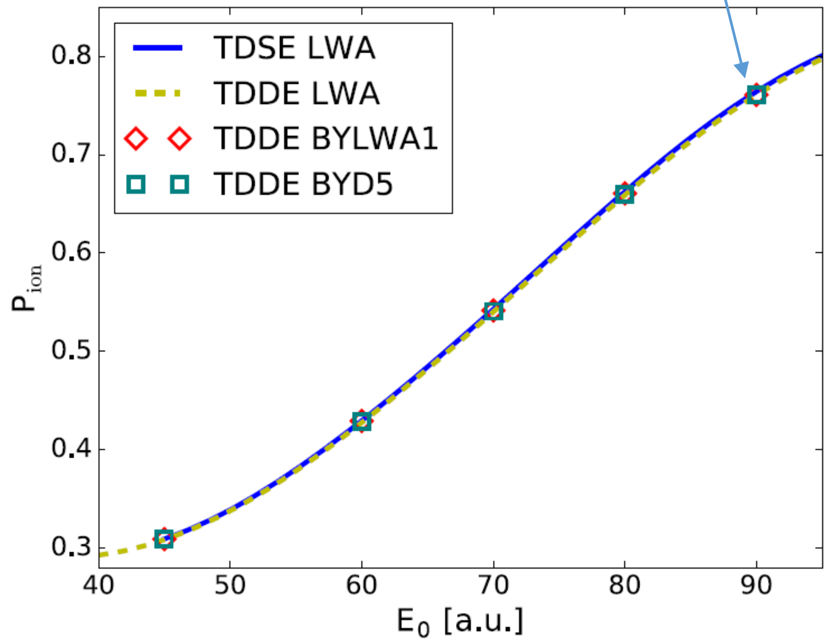


Problem 4: Weak convergence in x^n



Up to x^5 in the interaction

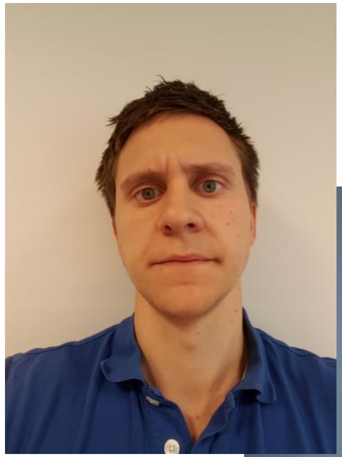
Up to x^0 in the interaction



Solution: The *propagation gauge*

Kjellsson, Førre, Simonsen, Selstø, Lindroth, Phys. Rev. A **96**, 023426 (2017)

The propagation gauge



Minimal coupling:

$$\mathbf{p} \to \mathbf{p} + e\mathbf{A}$$

Alternatively:

$$\mathbf{p} \to \mathbf{p} + e\mathbf{A} + \frac{e^2}{2mc} A^2 \hat{\mathbf{k}}$$

Vázquez de Aldana et al., Phys. Rev. A **64**, 013411 (2001)
Førre, Simonsen, Phys. Rev. A **93**, 013423 (2016)
Simonsen, Førre, Phys. Rev. A **93**, 063425 (2016)

Schrödinger equation

The propagation gauge Hamiltonian

Schrödinger Hamiltonian:

$$H = \frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{c}{2}\left\{1 - \sqrt{1 - \left(\frac{eA}{mc}\right)^2}, \hat{\mathbf{k}}\cdot\mathbf{p}\right\}$$

Førre, Simonsen, Phys. Rev. A **93**, 013423 (2016)

Dirac Hamiltonian:

$$H = c\boldsymbol{\alpha}\cdot\left(\mathbf{p} + e\mathbf{A} + \frac{e^2A^2}{2mc}\hat{\mathbf{k}}\right) + V\mathbb{1}_4 + \beta mc^2 - \frac{e^2A^2}{2m}\mathbb{1}_4$$

Kjellsson, Førre, Simonsen, Selstø, Lindroth, Phys. Rev. A **96**, 023426 (2017)

The propagation gauge Hamiltonian

Schrödinger Hamiltonian:

$$H = \frac{p^2}{2m} + V + \frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{c}{2}\left\{1 - \sqrt{1 - \left(\frac{eA}{mc}\right)^2}, \hat{\mathbf{k}}\cdot\mathbf{p}\right\}$$

Førre, Simonsen, Phys. Rev. A **93**, 013423 (2016)

Push in propagation direction, induced by the magnetic field

Dirac Hamiltonian:

$$H = c\boldsymbol{\alpha}\cdot\left(\mathbf{p} + e\mathbf{A} + \frac{e^2A^2}{2mc}\hat{\mathbf{k}}\right) + V\mathbb{1}_4 + \beta mc^2 - \frac{e^2A^2}{2m}\mathbb{1}_4$$

Kjellsson, Førre, Simonsen, Selstø, Lindroth, Phys. Rev. A **96**, 023426 (2017)

The propagation gauge Hamiltonian

Cancels the $\boldsymbol{A}^2$-term in the non-relativistic limit

Dirac Hamiltonian:

$$H = c\boldsymbol{\alpha} \cdot \left(\mathbf{p} + e\mathbf{A} + \frac{e^2 A^2}{2mc} \hat{\mathbf{k}} \right) + V \mathbb{1}_4 + \beta mc^2 - \frac{e^2 A^2}{2m} \mathbb{1}_4$$

Kjellsson, Førre, Simonsen, Selstø, Lindroth, Phys. Rev. A **96**, 023426 (2017)

The propagation gauge Hamiltonian

Now:
Makes sense to truncate at zeroth order and let *A* be purely time-dependent

-Far less restrictive than the dipole approximation

Cancels the *A*^{2}-term in the non-relativistic limit

Dirac Hamiltonian:

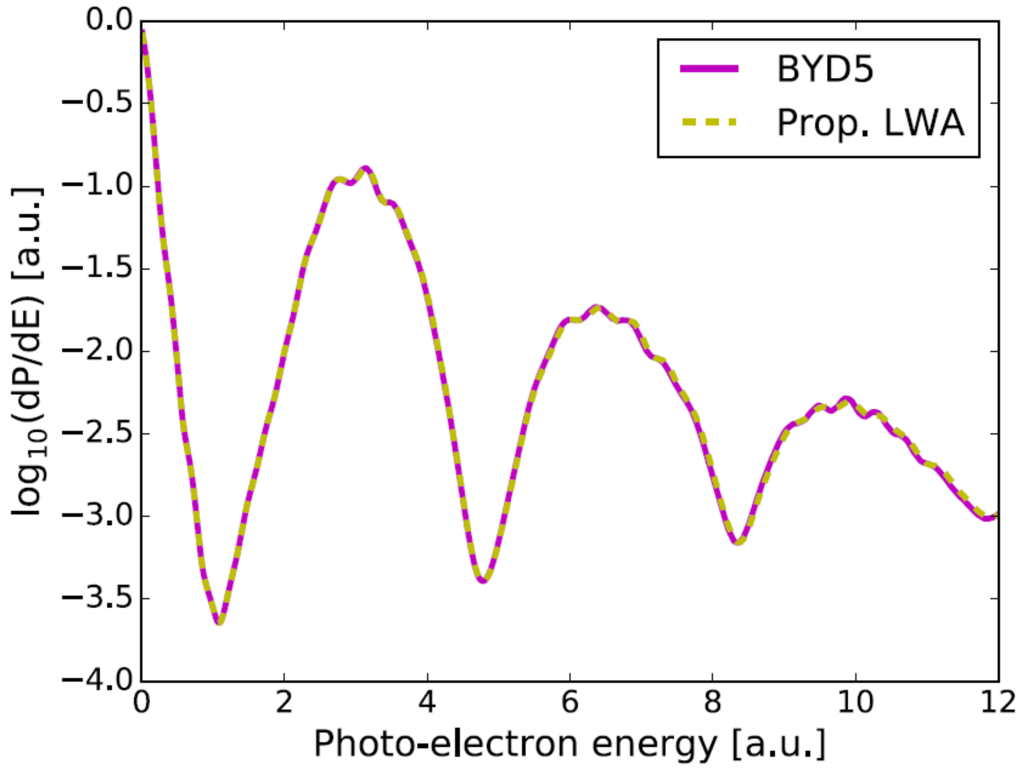
$$H = c\boldsymbol{\alpha} \cdot \left(\mathbf{p} + e\mathbf{A} + \frac{e^2 A^2}{2mc}\hat{\mathbf{k}}\right) + V \mathbb{1}_4 + \beta mc^2 - \frac{e^2 A^2}{2m}\mathbb{1}_4$$

Kjellsson, Førre, Simonsen, Selstø, Lindroth, Phys. Rev. A **96**, 023426 (2017)

The propagation gauge Hamiltonian

Now:
Makes sense to truncate at zeroth order and let *A* be purely time-dependent

-Far less restrictive than the dipole approximation



Dirac Hamiltonian:

$$H = c\boldsymbol{\alpha}\cdot\left(\mathbf{p} + e\mathbf{A} + \frac{e^2A^2}{2mc}\hat{\mathbf{k}}\right) + V\mathbb{1}_4 + \beta mc^2 - \frac{e^2A^2}{2m}\mathbb{1}_4$$

Kjellsson, Førre, Simonsen, Selstø, Lindroth, Phys. Rev. A **96**, 023426 (2017)

Remaining questions we would like to try and answer

- Can we push the numerics deeper into the relativistic region?
- What is the range of validity for using a homogeneous ***A*** (within the *propagation gauge*)?
- How about circular polarization?
- Can we see any relativistic corrections in high harmonic generation or in the spin dynamics?
- How strong are the relativistic corrections in the x-ray and the optical regions?
- Other things we should think about?

Remaining questions we would like to try and answer

- Can we push the numerics deeper into the relativistic region?
- What is the range of validity for using a homogeneous ***A*** (within the *propagation gauge*)?
- How about circular polarization?
- Can we see any relativistic corrections in high harmonic generation or in the spin dynamics?
- How strong are the relativistic corrections in the x-ray and the optical regions?
- Other things we should think about? **Suggestions?**