Experience with the Time-Dependent Dirac Equation

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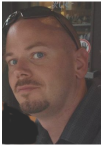
Frontiers of Intense Laser Physics, KITP Aug. 28 2014

Marcus Dahlström, Stockholm-Hamburg:
Attosecond delay in photoionization



Jimmy Vinbladh

XUV pump IR Probe simulation involving resonances - helium and towards many-elecron atoms

Luca Argenti, Madrid

Thomas Carette

Sølve Selstø, Oslo

Tor Kjellsson

Discussion here based on: The Time-Dependent Dirac Equation
Selstø, E. L., Bengtsson PRA79, 043418
and Vanne & Saenz PRA85, 033411

Why the Dirac Equation?

- Intense fields drive the electron to relativistic velocities
- High nuclear charges drive the electron to relativistic velocities
- High nuclear charges (ions or heavy elements) requires relativistic structure. E.g. attosecond delays in high-Z elements.

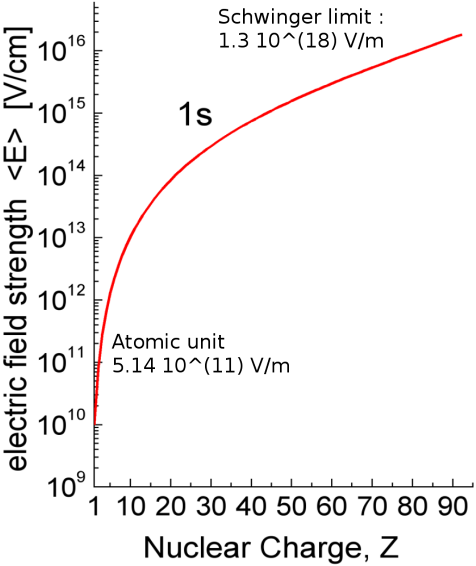
Magnetic Effects?

- Is there any point in solutions of the Dirac equation in the dipole-approximation?

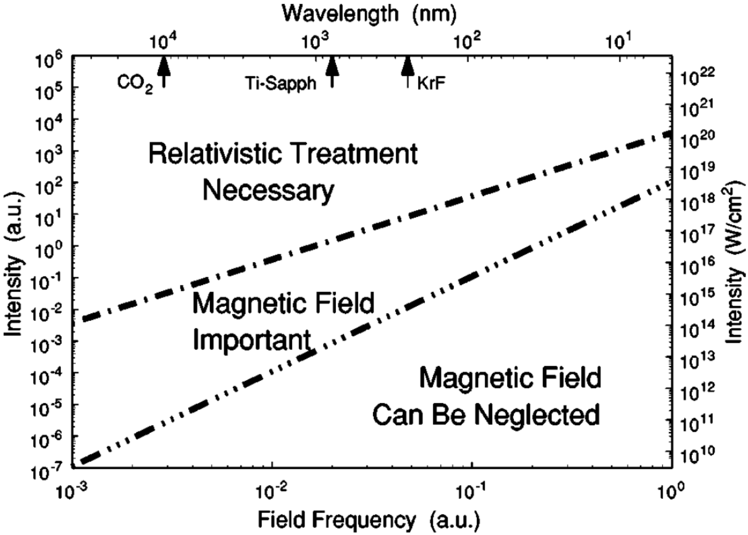
More QED?

- $e^{-}e^{+}$ Pair-production for extreme fields - should be a window where Schrödinger is insufficient and pair-production can be neglected.

Field & Nuclear charge?

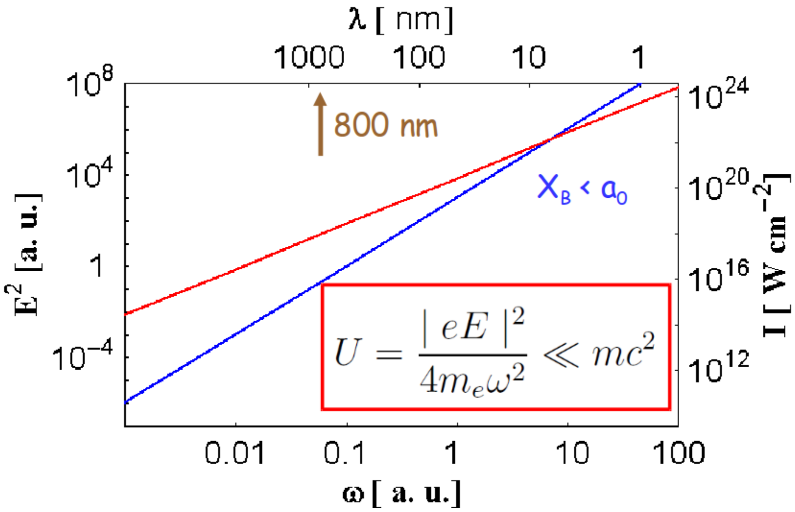


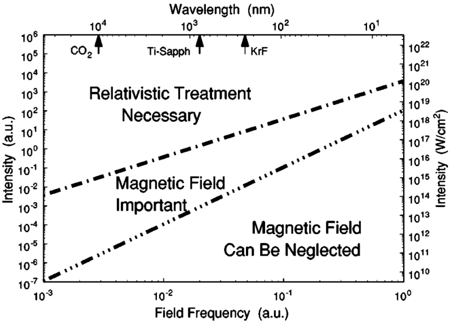
Fields & Photon energies?



From Reiss PRA63, 013409

Fields & Photon energies?

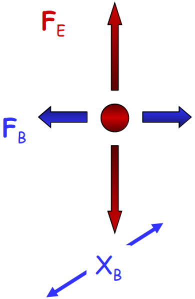




Relativistic

$$U = \frac{|\, e\mathbf{E_0} \,|^2}{4 m_e \omega^2} \sim mc^2$$

Should be included if
$U_p \approx mc^2/10$



Magnetic part cannot be neglected if the magnetic drift per cycle $\sim a_0$.

$$x_B \sim \frac{|\, e\mathbf{E_0} \,|^2}{m^2} \frac{1}{c\omega^3} \sim a_0$$

The time-dependent Dirac Equation

$$i\hbar\frac{\partial}{\partial t}\Psi = \left\{c\boldsymbol{\alpha}\cdot\left(\mathbf{p}+e\mathbf{A}\left(\omega t-\mathbf{k}\cdot\mathbf{r}\right)\right)+V(Z)+\beta mc^2\right\}\Psi$$

E.g. electric field in z- direction and magnetic field i y- direction:

$$\mathbf{A} = A(\omega t - kx)\hat{z}$$

Solutions with $A = 0$ & spherically symmetric V

$$\Psi_{n,\kappa,j,m}(\mathbf{r}) = \begin{pmatrix} F_{n,\kappa,j,m}(\mathbf{r}) \\ iG_{n,\kappa,j,m}(\mathbf{r}) \end{pmatrix} = \frac{1}{r}\begin{pmatrix} P(r)\,\chi_{\kappa,j,m} \\ iQ(r)\,\chi_{-\kappa,j,m} \end{pmatrix}$$

with continuum energies from $-\infty$ to $-mc^2$, and from mc^2 to ∞

$$F_{LARGE} \sim \Psi_{nrel},\ G_{SMALL} \sim \frac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}F_{LARGE},\ \text{positive energy states}$$

vice versa for negative energy states.

Problems

- The form of the interaction
- The negative energy states
- computer time & space

The form of the interaction

For example a simple pulse in the dipole approximation

$$A_0 sin^2\left(\frac{\pi t}{T}\right) cos\left(\omega t\right) \underset{\text{Lorentz inv.}}{\Longrightarrow} A_0 sin^2\left(\frac{\pi\left(\omega t - kx\right)}{\omega T}\right) cos\left(\omega t - kx\right)$$

with spatial dependence in both carrier and envelope

Standard procedure: expand spatial part of **A** in plane waves

$$e^{\pm i\mathbf{k}\cdot\cdot\mathbf{r}} \sim \sum_{\lambda\mu} \left(\pm i\right)^k j_\lambda\left(kr\right) Y^*_{\lambda\mu}\left(\hat{k}\right) C^\lambda_\mu\left(\theta,\phi\right)$$

$\boldsymbol{\alpha}\cdot\mathbf{A}$ leads to matrix elements of:

$$C^\lambda_\mu \alpha_q = \sum_{L=\lambda-1}^{\lambda+1} \ldots \left\{\boldsymbol{\alpha}\cdot\mathbf{C}^\lambda\right\}^L_{M=q+\mu}$$

$\lambda = 0, L = 1$ electric dipole, $\lambda = 1, L = 1$ magnetic dipole, $\lambda = 1, L = 2$ electric quadrupole ...

The form of the interaction

Straightforward:

$$cos\left(\omega t - kx\right) \rightarrow \sum_{\lambda,\mu} \sum_{L=\lambda-1}^{\lambda+1} \ldots j_\lambda\left(kr\right)\left\{\boldsymbol{\alpha}\cdot\mathbf{C}^\lambda\right\}^L_{M=q+\mu} \times sin\left(\omega t\right)/cos\left(\omega t\right)$$

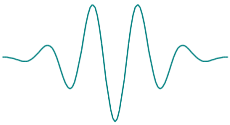
- truncate the sum over λ, L?
- expand $j_\lambda\left(kr\right)$ - ok for $kr \ll 1$?
- $\Delta m_j = M$ - the number of couplings increases very fast.

BUT, what to do with the envelope?

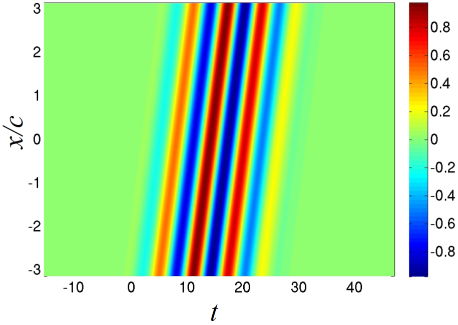
- Just skip the spatial part in the envelope? Should be OK for long pulses.
- Fourier expansion? - Note! gives trains of pulses. Problem for long propagations times.

THE SPATIAL PART OF THE ENVELOPE?

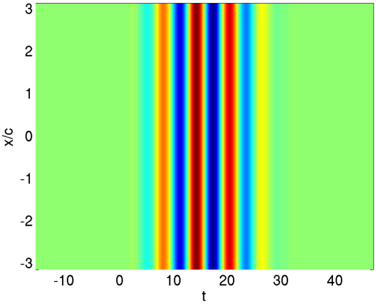
$$A_0 sin^2\left(\frac{\pi\left(\omega t - kx\right)}{\omega T}\right) cos\left(\omega t - kx\right)$$



$\omega = 2a.u.$

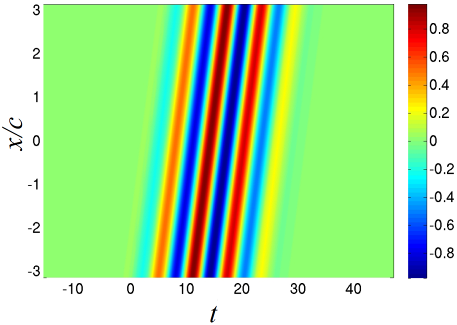


The actual field. Atomic units.

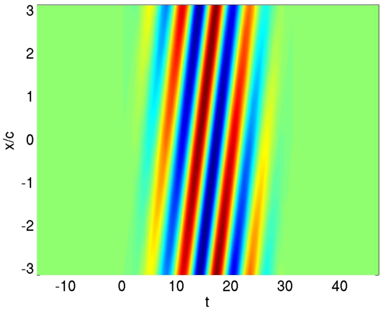


The dipole field. Atomic units

The spatial part of the envelope?

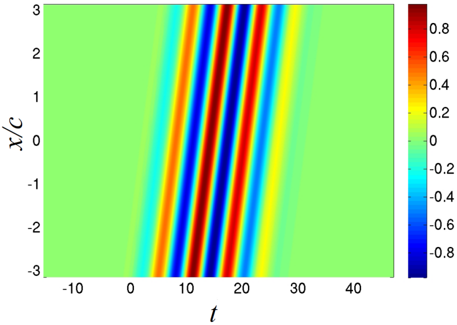


The actual field. Atomic units.

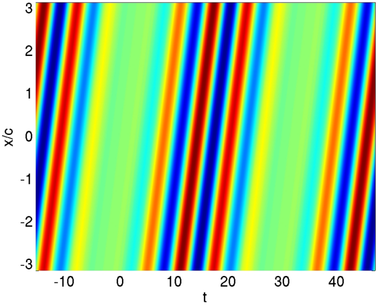


Envelope purely time-dependent.

The spatial part of the envelope?



The actual field. Atomic units.



Fourier expansion

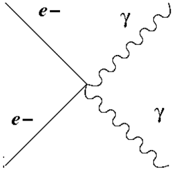
Propagation from $t = 0 \rightarrow T$? Expanding atom $\rightarrow$ interaction time $> T$ risk for interaction with the next (unphysical) pulse. Keep more Fourier components...

Neg. energy states/(Virtual) Pair Prod.

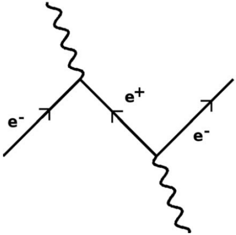
Atomic structure

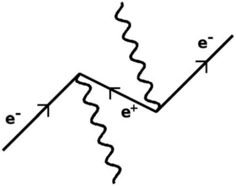
$$E_{nrel} \approx Z^2 a.u. \quad \Delta E_{Dirac} \approx \alpha^2 Z^4 a.u. \quad \Delta E_{virtualpairs} \approx \alpha^3 Z^4 a.u.$$

Thomson scattering: relativistic versus non-relativistic frame work



$$\frac{e}{m}\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m^2}\mathbf{A}^2$$





$$ec\boldsymbol{\alpha}\cdot\mathbf{A}$$

Low photon energy limit: The whole contribution arises from negative energy state. (Dirac 1930)

Neg. energy states/(Virtual) Pair Prod.

Dirac equation:

$$\left(c\boldsymbol{\alpha}\cdot(\mathbf{p}+e\mathbf{A})+V(Z)+(\beta-1)mc^2\right)\Psi=\left(E-mc^2\right)\Psi$$

In two-component form:

$$\begin{aligned} VF+c\sigma\cdot(e\mathbf{A}+\mathbf{p})\;G&=\varepsilon F\\ c\sigma\cdot(e\mathbf{A}+\mathbf{p})\,F+\left(V-2mc^2\right)\;G&=\varepsilon G\end{aligned}$$

Foldy-Wouthuysen type expansion (PR78, 29, 1950):

$$F_{LARGE}\approx\Psi_{nrel},\;\;G_{SMALL}\approx\frac{1}{2mc}\,(e\mathbf{A}+\mathbf{p})\,F_{LARGE}$$

gives the Schrödinger equation back

$$V\,F_{LARGE}+\frac{1}{2m}\,(e\mathbf{A}+\mathbf{p})^2\;F_{LARGE}=\varepsilon F_{LARGE},$$

A^2 term reappearing - through the small-component contribution

Neg. energy states/(Virtual) Pair Prod.

$$\frac{\langle \psi_{e-} \mid c\boldsymbol{\alpha} \cdot \mathbf{A} \mid \psi'_{e-} \rangle \langle \psi'_{e-} \mid c\boldsymbol{\alpha} \cdot \mathbf{A} \mid \psi_{e-} \rangle}{\Delta E} \approx$$

$$\frac{\langle F_{e-} \mid c\boldsymbol{\sigma} \cdot \mathbf{A} \mid G'_{e-} \rangle \langle G'_{e-} \mid c\boldsymbol{\sigma} \cdot \mathbf{A} \mid F_{e-} \rangle}{\Delta E \approx 1} + \ldots \approx \mid A_0 \mid^2$$

$$\frac{\langle \psi_{e-} \mid c\boldsymbol{\alpha} \cdot \mathbf{A} \mid \psi'_{e+} \rangle \langle \psi'_{e+} \mid c\boldsymbol{\alpha} \cdot \mathbf{A} \mid \psi_{e-} \rangle}{\Delta E} \approx$$

$$\frac{\langle F_{e-} \mid c\boldsymbol{\sigma} \cdot \mathbf{A} \mid G'_{e+} \rangle \langle G'_{e+} \mid c\boldsymbol{\sigma} \cdot \mathbf{A} \mid F_{e-} \rangle}{\Delta E \approx 2mc^2} + \ldots \approx \mid A_0 \mid^2$$

Neg. energy states/(Virtual) Pair Prod.

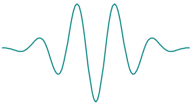
Example:

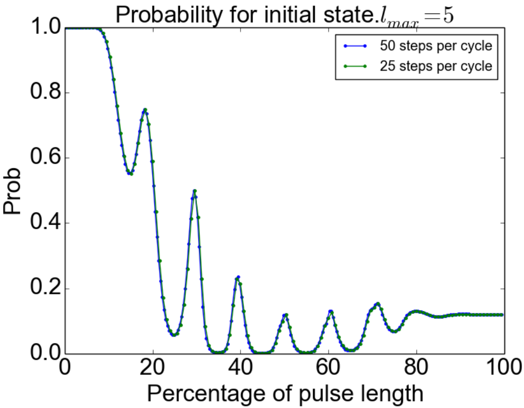
Survival probability in hydrogen ground state

$I = 3 \times 10^{19}$ W/cm^2

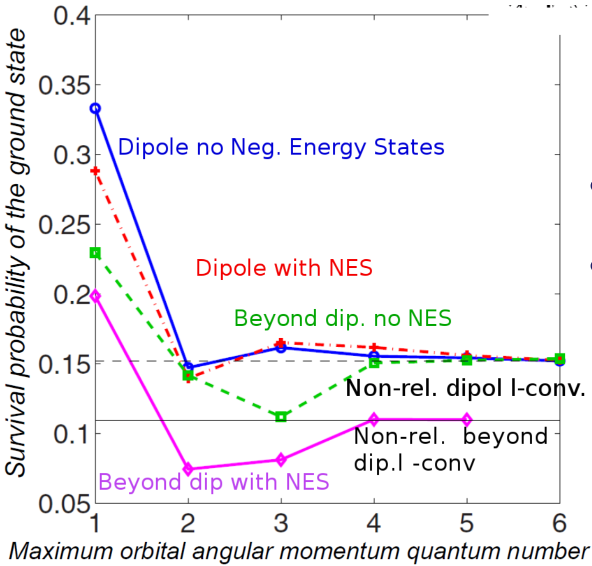
5 cycle pulse ($\sim$ 380as),

$\hbar\omega = 2$ a.u.



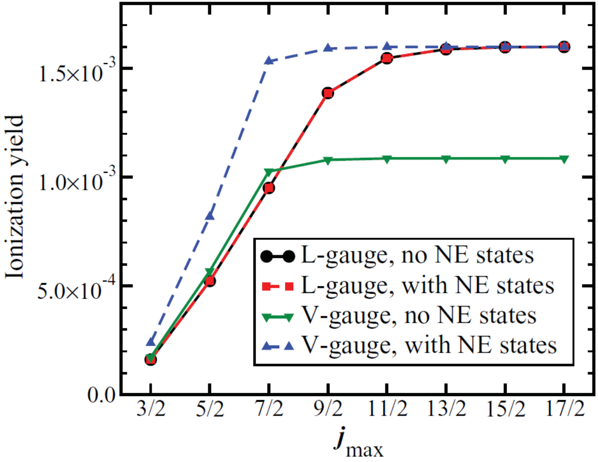


Neg. energy states/(Virtual) Pair Prod.



- Negative Energy States as defined by $A = 0$
- Spatial dependence in both carrier and envelope.

Neg. energy states/(Virtual) Pair Prod.



- Ionization yield
  $Z = 50$
  $(E_{IP} = 1250 a.u.)$
- 20-cycle cos^2 -pulse
- $I = 5 \times 10^{23}$ W/cm^2
- $\hbar\omega = 500$ a.u.
- Dipole approximation.

Vanne & Saenz PRA85 033411

Gauges

and the importance of negative energy states

- Substantial matrix elements between positive and negative energy states for operators mixing upper & lower wave function components.
- Gauge transformations?

$$ec\boldsymbol{\alpha}\cdot\mathbf{A},\ \text{negative energy states in leading order}$$

$$UH_DU^\dagger + i\hbar\frac{\partial U}{\partial t}U^\dagger,$$

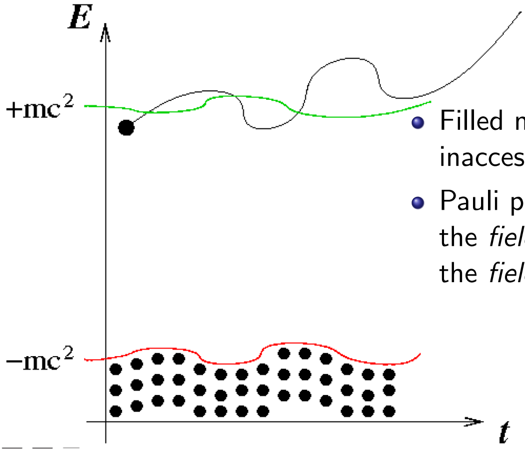
Negative energy states in relative order α^2? Use:

$$U = e^{ie\mathbf{A}\cdot\mathbf{r}/\hbar} \rightarrow e\mathbf{r}\cdot\mathbf{E} + ec\left(\boldsymbol{\alpha}\cdot\mathbf{k}\right)\left(\mathbf{r}\cdot\frac{\partial\mathbf{A}}{\partial\eta}\right),\ \eta = \omega t - kx$$

Negative energy states in relative order α^4? Use:

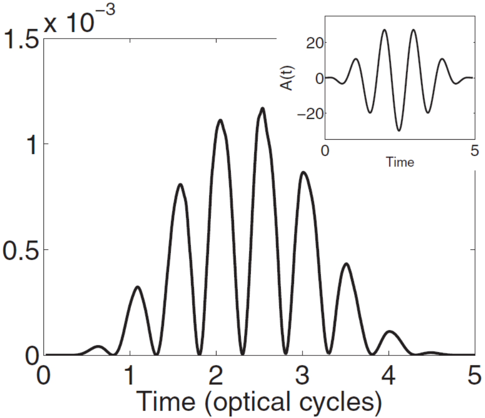
$$U = e^{e\beta\boldsymbol{\alpha}\cdot\mathbf{A}/2mc} \rightarrow \frac{e}{m}\beta\mathbf{A}\cdot\mathbf{p} + \frac{e^2}{2m}\beta\mathbf{A}^2 + \frac{e\hbar}{2m}\beta\boldsymbol{\sigma}\cdot\mathbf{B} + O\left(\beta\frac{ec\boldsymbol{\alpha}\cdot\mathbf{A}}{mc^2}\right)$$

NEG. ENERGY STATES/(VIRTUAL) PAIR PROD.



- Filled negative energy state - compare inaccessible core!
- Pauli principle excludes admixture of the *field dressed states* ($\mathbf{A} \neq 0$) - not of the *field free states* ($\mathbf{A} = 0$).

NEG. ENERGY STATES/(VIRTUAL) PAIR PROD.



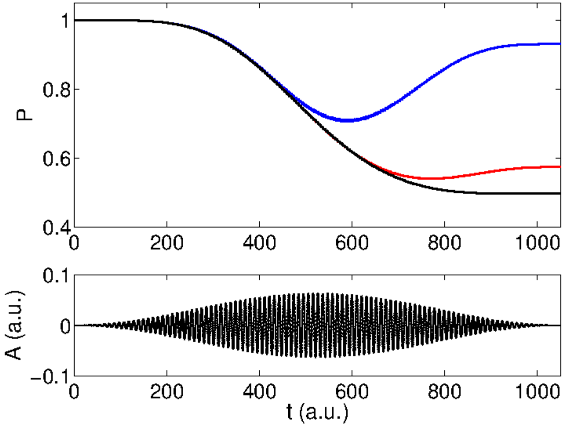
- Filled negative energy state - compare inaccessible core!
- Pauli principle excludes admixture of the *field dressed states* - not of the *field free states*

Population of negative energy states to (defined with $A = 0$) during the pulse.

Population of negative energy states to H(t) zero to machine accuracy!

Practical Considerations

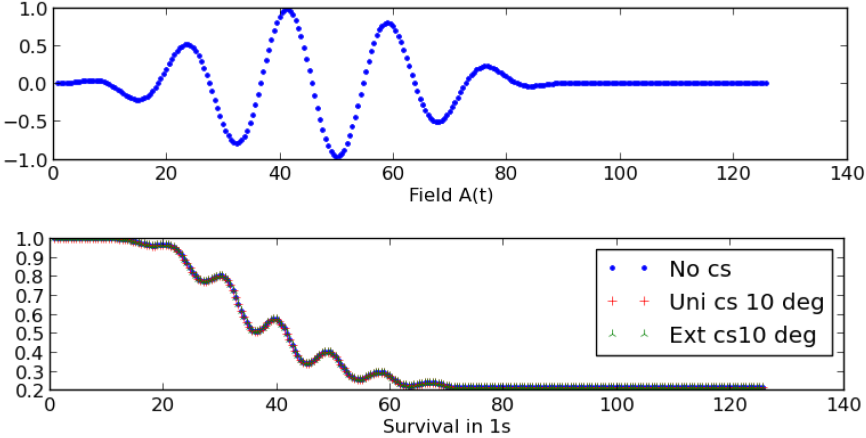
- Finite basis set: Finite difference or B-splines representation (avoid spurious states as Froese-Fisher & Zatsarinny CPC. 180, 879)
- Complex rotation (uniform CS for survival rate & ionization rate) (Complex rotation & Dirac, e.g. Zong, E.L., et al. PRA56, 386,-97)



1s survival rate:
50 basis function per ℓ
100 basis func. per ℓ
Complex scaling:
20 basis function per ℓ

Practical Considerations

- Complex rotation (uniform CS for survival rate & ionization rate)



Practical Considerations

Problems with Negative Energy States!

- States with large energy differences important $\rightarrow$ many time steps $\Delta t < \hbar/2mc^2$?
- Time propagation with complex scaling diverges ...

How can NES be avoided?

Expansion in eigenstates to $H(t)$

$$\Psi(t+\Delta t) \approx e^{-iH(t)\Delta t}\,\Psi(t) \approx \sum_j e^{-iE_j\Delta t}\mid \phi_j^t\rangle\langle\phi_j^t\mid \Psi(t)\rangle$$

i.e. diagonalization at every time-step?

- No NES needed until real pair-production sets in
- only rather sparse time-grid required

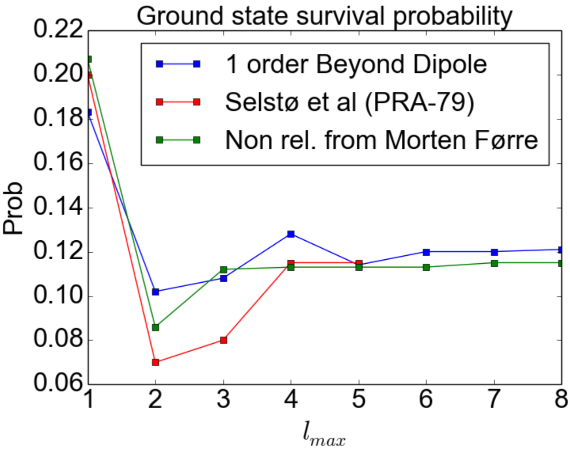
Room for improvements

- Fits well with Krylov (exclude energies $< -mc^2$.) Promising test runs.
- or parallelization: several time steps diagonalized simultaneously (working)

More to study

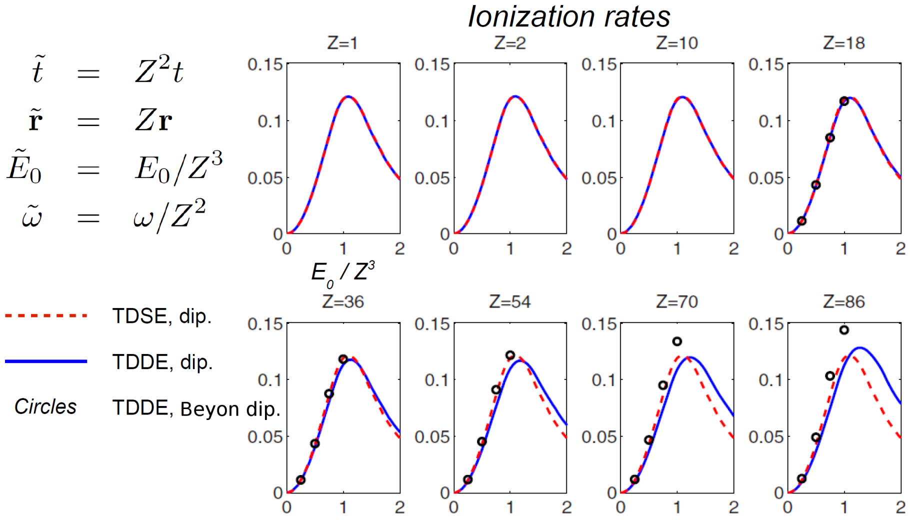
Sensitivity to the full interaction?

$$\sum_{\lambda,\mu,L,M} \ldots j_\lambda(kr)\left\{\boldsymbol{\alpha}\cdot\mathbf{C}^\lambda\right\}^L_M \ldots ? \quad \text{Keep } \lambda = 0-1 \text{ expand } j_\lambda(kr)?$$



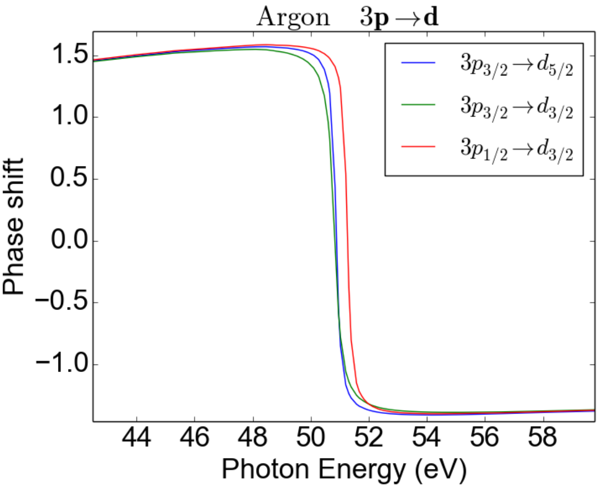
More to do

$$\begin{aligned} \tilde{t} &= Z^2 t \\ \tilde{\mathbf{r}} &= Z\mathbf{r} \\ \tilde{E}_0 &= E_0/Z^3 \\ \tilde{\omega} &= \omega/Z^2 \end{aligned}$$



Ionization rates by monochromatic light calculated with TDSE

Atomic structure?



Phase shift of the Argon photoelectron close to the Cooper minimum (RPA-caclulation based on the Dirac-Equation).

Conclusions

- Negative Energy states are important, but can be handled
- The main problem is the size of the problem!
- More to do!

SPURIOUS STATES?

A problem we know how to handle: For example with B-splines(Froese-Fisher & Zatsarinny CPC. 180, 879)

n	Exact	$k = 4$ & $\tilde{k} = 5$	$k = 5$ & $\tilde{k} = 5$
2	-0.1250020802	-0.1250020802	-0.5000066566
3	-0.5555629517	-0.5555629518	-0.1250020802
4	-0.3125033803	-0.3125033802	-0.5555629518
5	-0.2000018106	-0.2000016849	-0.3125033803

Table: The four lowest electron eigenvalues for $p_{1/2}$