Contributions to the Theory of Atoms and Molecules in Strong Electromagnetic Fields

Defence of Dissertation for the Degree of PhD.

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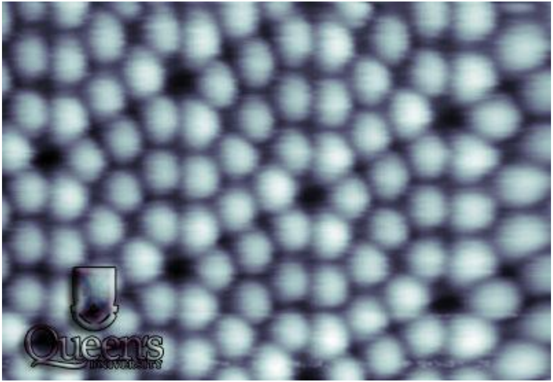
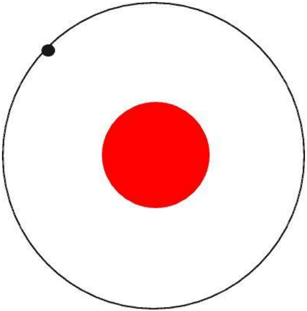
- The atom
- Light
- The interaction between atoms and light
- Hydrogen atoms in strong, short laser pulses
- The hydrogen molecular ion in laser pulses
- The Landau-Zener model and single electron transport in a double quantum dot
- Summary

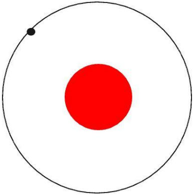
Atoms

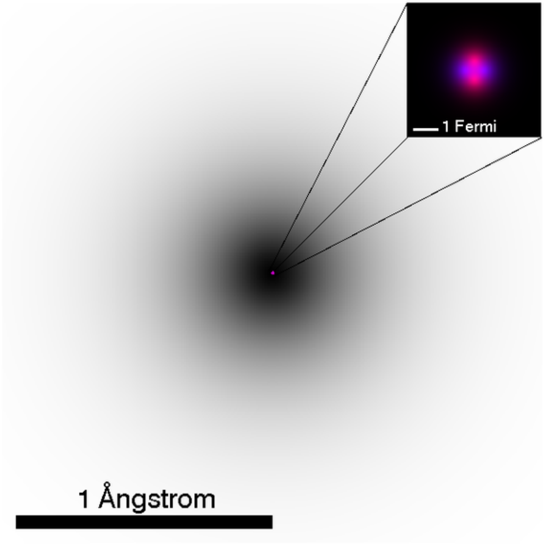


http://www.physics.queensu.ca/~nanophys/stm.html

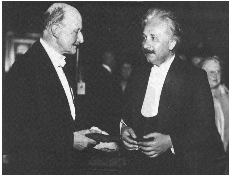
9 nm $\times$ 6.3 nm






1 Fermi
1 Ångstrom

Some "highlights" in the evolution of quantum mechanics



- Planck and Einstein, early 1900-s: Light sometime behaves as particles, not just waves
- Bohr-model, 1913
- DeBroglie, 1923: Matter sometimes behave as waves, not just particles
- Schrödinger, 1932: Wave description of matter

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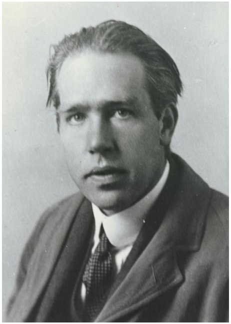

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The time independent Schrödinger Equation:

$$\begin{aligned} H\Psi(\mathbf{r}) &= E\,\Psi(\mathbf{r}) \\ H &= -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r}) \end{aligned}$$

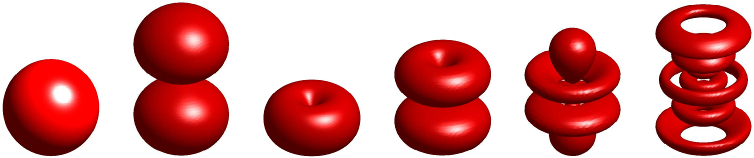
$|\Psi(\mathbf{r})|^2$ gives the probability of finding the particle near position $\mathbf{r}$

Hydrogen:

Quantum numbers n, l and m, energy $E_n = -\frac{B_0}{n^2}$

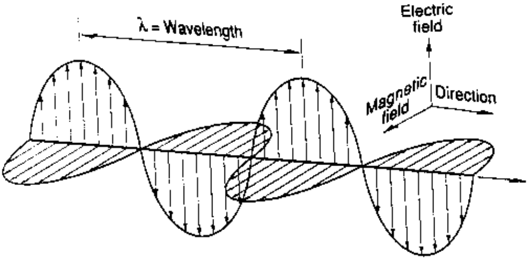
$\{n, l, m\} =$

$\{1,0,0\}$ $\{2,1,0\}$ $\{2,1,1\}$ $\{3,2,1\}$ $\{4,3,0\}$ $\{7,3,1\}$



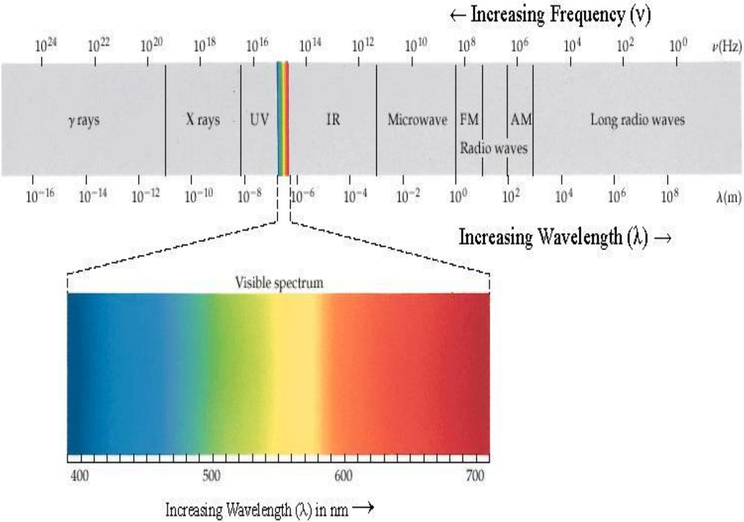
Light

-Consists of various colours/ wavelengths



Traveling electromagnetic wave

Specter

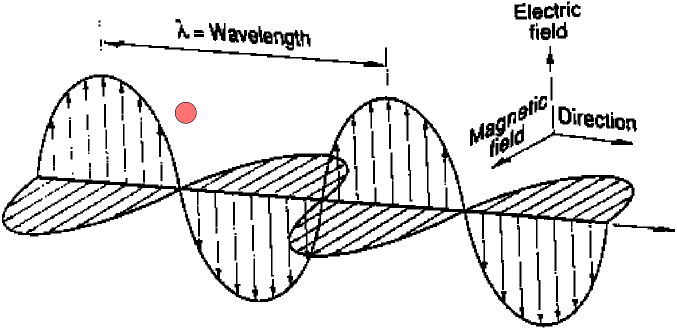

← Increasing Frequency (ν)
10^{24} 10^{22} 10^{20} 10^{18} 10^{16} 10^{14} 10^{12} 10^{10} 10^{8} 10^{6} 10^{4} 10^{2} 10^{0} ν(Hz)
γ rays
X rays
UV
IR
Microwave
FM
AM
Radio waves
Long radio waves
10^{-16} 10^{-14} 10^{-12} 10^{-10} 10^{-8} 10^{-6} 10^{-4} 10^{-2} 10^{0} 10^{2} 10^{4} 10^{6} 10^{8} λ(m)
Increasing Wavelength (λ) →
Visible spectrum
400
500
600
700
Increasing Wavelength (λ) in nm →

LASER

- Produced through stimulated emission
- One single colour
- Very intense, coherent light

- Treat the field classically

- The Dipole Approximation



$$\mathbf{E}(\mathbf{r}, t) \rightarrow \mathbf{E}(t)$$

The interaction between matter and light

The time dependent Schrödinger equation:

$$H\Psi = i\hbar \frac{\partial}{\partial t}\Psi$$

Hamiltonian: $H = H_{\mathrm{atom}} + H_{\mathrm{int}}$
Different descriptions of the interaction:

Length gauge	Velocity gauge	KH frame
$H_{\mathrm{int}} = -q\mathbf{r}\cdot\mathbf{E}(t)$	$H_{\mathrm{int}} = -\frac{q}{m}\mathbf{p}\cdot\mathbf{A}(t)$	$\mathbf{r} \to \mathbf{r} - \boldsymbol{\alpha}(t)$

- Classical trajectory of a free particle in the electric field $\mathbf{E}(t)$:

$$\boldsymbol{\alpha}(t) = -\frac{q}{m}\int_{t_0}^{t}\int_{t_0}^{t'} \mathbf{E}(t'')\,dt''\,dt'$$

The Kramers Henneberger frame

$$H_{\mathrm{KH}} = -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r} - \boldsymbol{\alpha}(t)) + \left(\frac{q^2}{2m}\mathbf{A}(t)\right)$$

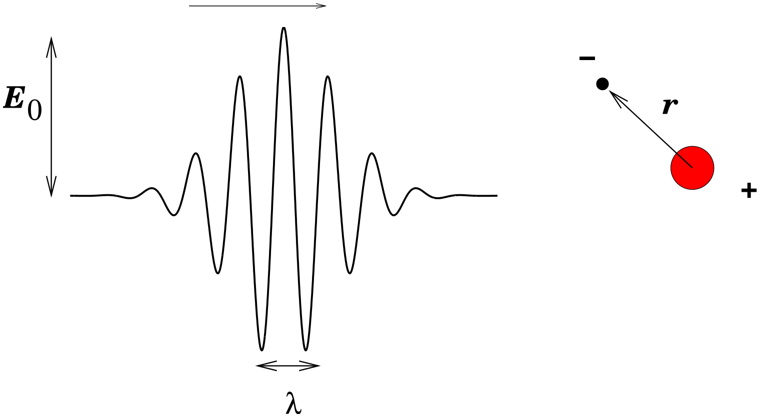
The particle "sees" a moving potential

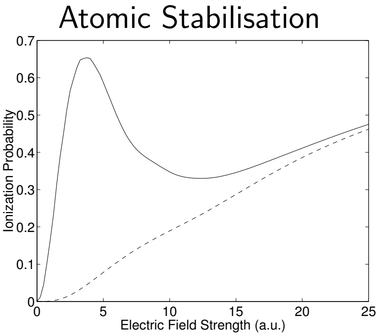
The transition from velocity gauge to KH frame can be done without the dipole approximation;

$$H_{\mathrm{KH}}^{\mathrm{ND}} \to -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r} - \boldsymbol{\alpha}(\mathbf{r},t)) + \frac{q^2}{2m}\left(\mathbf{A}(\mathbf{r},t)\right)^2$$

corresponding to a non-homogeneous field $\mathbf{E}(\mathbf{r},t)$

Hyrogen atom exposed to short, intense laser pulse

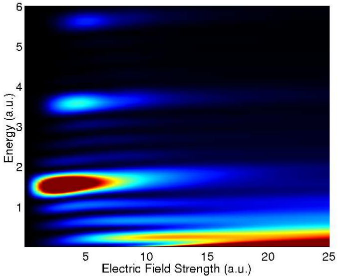




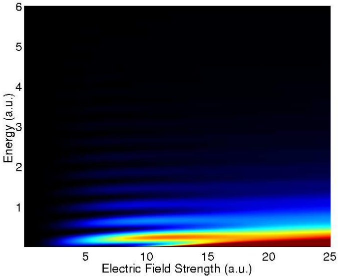
- High frequency
- Probability of ionisation *decreases* with increasing intensity/ frequency
- Claim: Artifact of the dipole approximation

- Found that the phenomena sustains inclusion of the spatial dependence of the field
- The stabilistaion mechanism understood at a consequence of the multi-photon channel "closing down"
- Dynamics described by the time averaged KH potential:
$V_{\mathrm{KH}}(\mathbf{r},t) \sim -\frac{1}{|\mathbf{r}-\boldsymbol{\alpha}(t)|} \rightarrow V^0_{\mathrm{KH}}(\mathbf{r}) = -\frac{1}{T}\int_T \frac{1}{|\mathbf{r}-\boldsymbol{\alpha}(t)|}\, dt$

$V_{\mathrm{KH}}(\mathbf{r},t)$

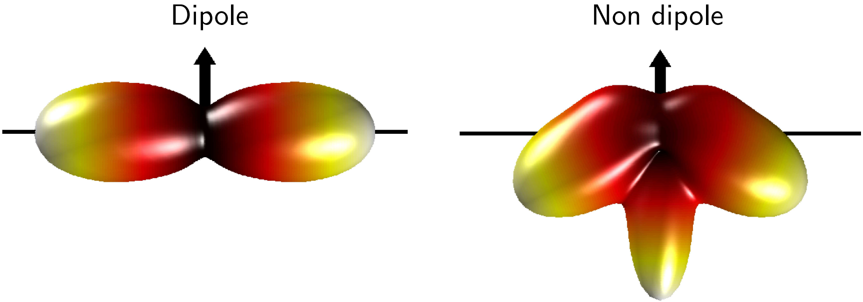


$V^0_{\mathrm{KH}}(\mathbf{r})$



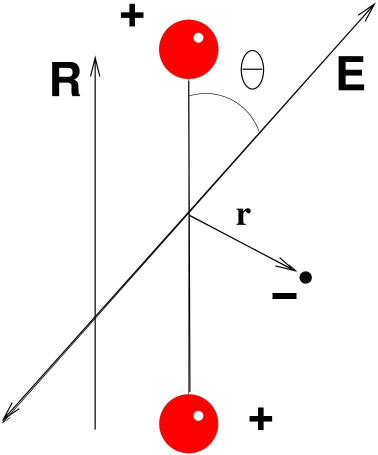
For strong fields of high frequency, the dipole approximation breaks down

Angular distribution of the photo electron after ionization of atomic hydrogen:



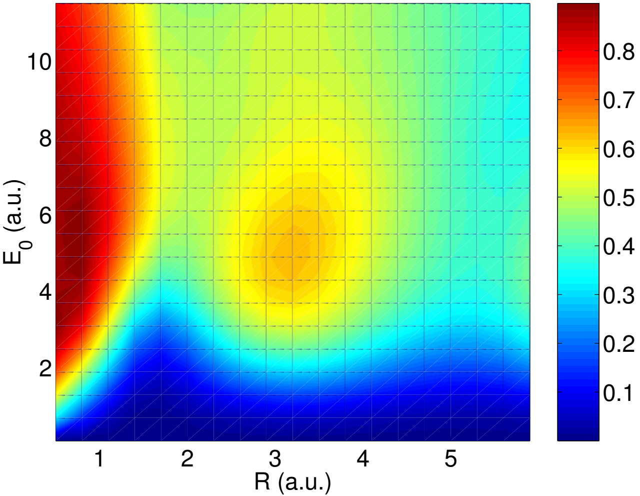
The interplay between the Coulomb, electric and magnetic field leads to the appearance of a third lobe in the angular distribution

Photo-ionisation of H_2^+

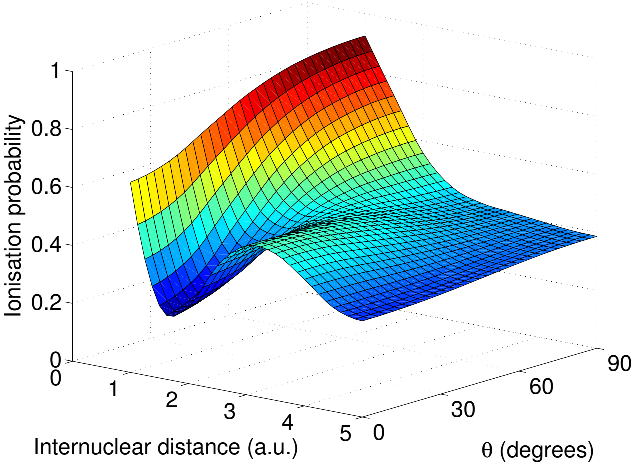


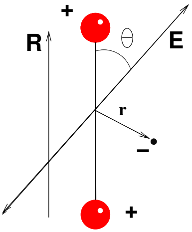
Fixed nuclei approximation

$P_I(R, E_0)$ for $\theta = 0$



$P_I(R, \theta)$ for $E_0 = 3.0$ a.u.

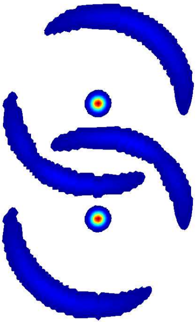




Model:

Interfering waves traveling from each of the protons:

$$\psi_{\text{out}} = f(\Omega_1)\frac{\exp(ikr_1)}{r_1} + f(\Omega_2)\frac{\exp(ikr_2)}{r_2}$$

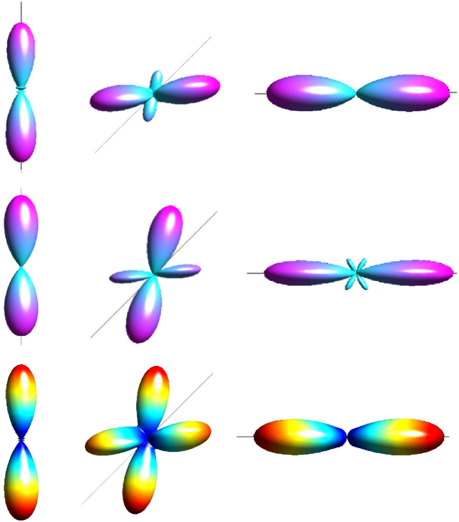


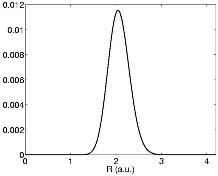
Angular Distributions

θ=0°   θ=45°   θ=90°

R=2 a.u.

R=3 a.u.



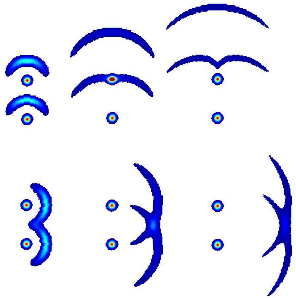


Model: $\psi_{\rm out} \rightarrow f(\Omega)\cos(1/2\, k\hat{\mathbf{r}}\cdot\mathbf{R})e^{ikr}/r$

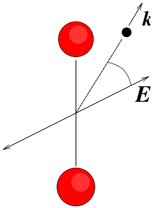
$\Rightarrow \frac{dP}{d\Omega_k} \propto |f(\Omega)|^2\cos(1/2\,\mathbf{k}\cdot\mathbf{R})$

Does not give very good agreement for parallel polarisation

Reason: Coulomb scattering

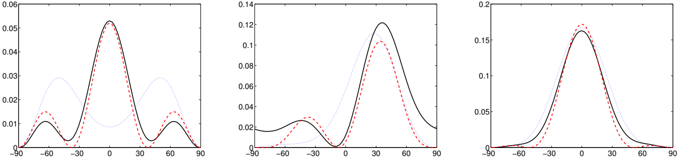


Accounted for by the eikonal (WKB) approximation

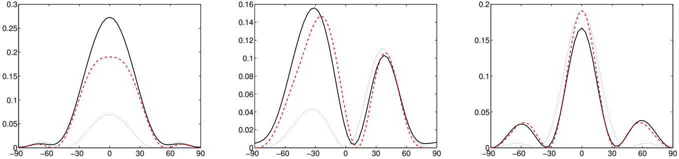


$\theta = 0^\circ$ $\theta = 45^\circ$ $\theta = 90^\circ$

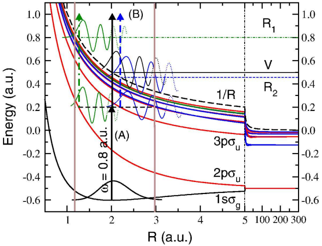
$R = 2a.u.$



$R = 3a.u.$



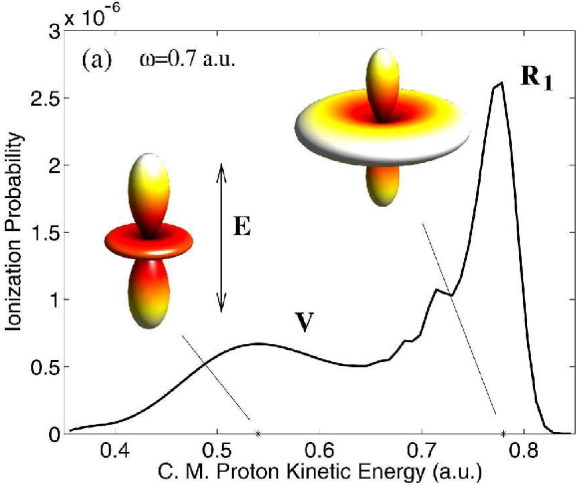
Two photon ionisation with moving nuceli



Born-Oppenheimer approximation
Correct asymptotic behaviour in the final electronic states

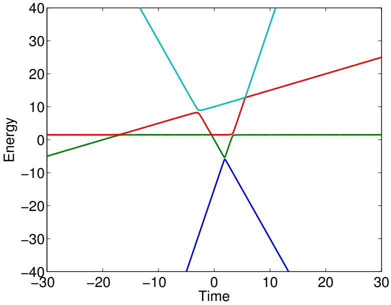
$$\Psi(R, \mathbf{r}, t) = \sum_n \sum\!\!\!\!\!\!\int_{\nu_n} c_{n,\nu_n}(t)\chi_{\nu_n}(R)\psi_n^{\mathrm{el}}(R;\mathbf{r}) + \sum_l \int d\varepsilon \sum\!\!\!\!\!\!\int_{\nu_\varepsilon} c^l_{\varepsilon,\nu_\varepsilon}(t)\chi_{\nu_\varepsilon}(R)\psi^{\mathrm{el}}_{l,\varepsilon}(R;\mathbf{r})$$

$T=10fs,\ I=10^{12}\ W/cm^2$

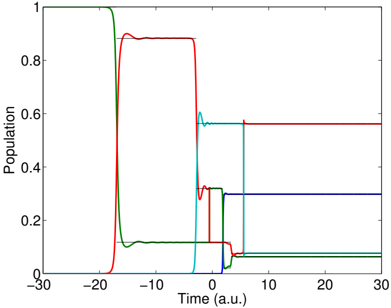


The Landau-Zener model

Energies (adiabatic)

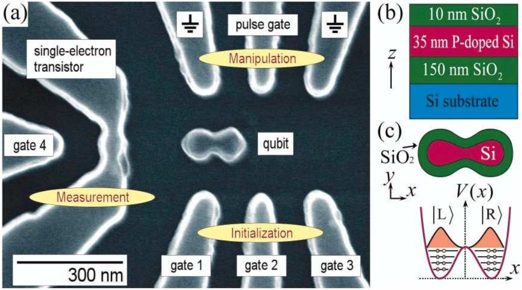


Populations



- The transition probabilities at each avoided crossing may be found analytically
- The dynamcs are given by trivial phase evolution between avioded crossings

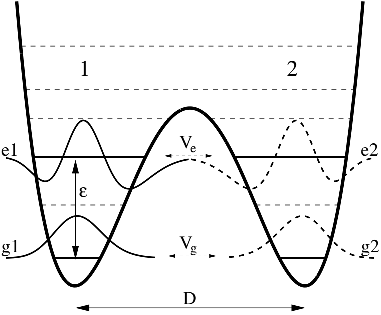
These ideas have been used to describe electron transport in a coupled quantum dot

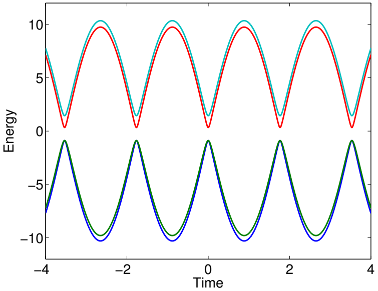


Gorman et al. Phys. Rev. Lett. **95**, *090502 (2005)*

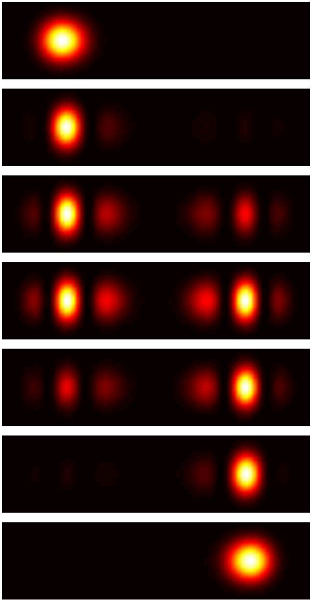
-*Artificial molecule*

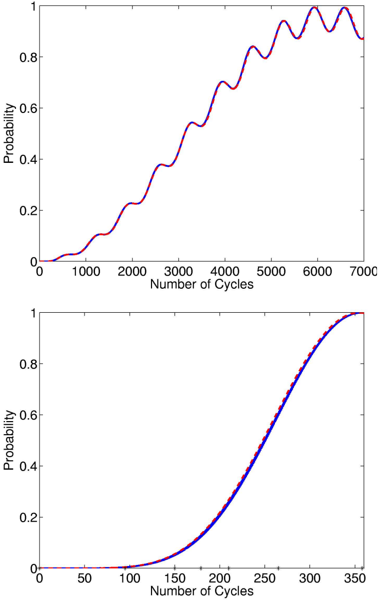
4-state model:





$$E(t) = E_0 \sin(\omega t)$$



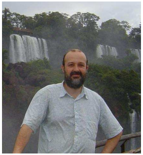

Probability
Number of Cycles
0
0.2
0.4
0.6
0.8
1
0
1000
2000
3000
4000
5000
6000
7000
Probability
Number of Cycles
0
0.2
0.4
0.6
0.8
1
0
50
100
150
200
250
300
350

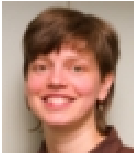
In summary

- Studied ionization dynamics of hydrogen
  - Shown that atomic stabilsation is *not* an artifact of the dipole approximation
  - Describe the phenomena accounting for dynamic stabilisation
  - Found signature of non-dipole interaction in photo-ionisation in the strong field regime
- Investigated the importance of geometrical factors in photo-ionisation of H_2^+ and explain its origin in terms of interference and refraction.
  - Describe angular distribution of the photoelctron including internuclear vibration.
- Application of analytical models to describe few-level systems
  - Feasible method of coherent single electron transport between couple quantum dots

In alphabetical order


A. Palacios


F. Martín


I. Sundvor


J. P. Hansen

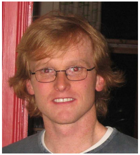

J. Fernández


J. McCann


L. Kocbach


L. B. Madsen


M. Volkov


M. Førre


T. Birkeland


V. Ostrovsky

The Keldysh parameter

$$\gamma = \frac{\omega\sqrt{2mI_p}}{|q|E_0}$$