Photoionisation of H_2^+ by short, intense laser pulses

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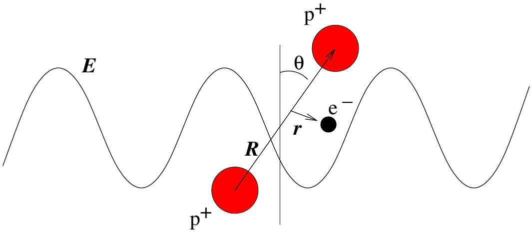


Method

- Solve TDSE by split step scheme on a spherical grid
  Hamiltonian (atomic units):
  $H = \frac{1}{2}\nabla^2 - \frac{1}{|\mathbf{r}-\mathbf{R}/2|} - \frac{1}{|\mathbf{r}+\mathbf{R}/2|} + \mathbf{E}\cdot\mathbf{r}$
  Dipole approximation:
  $\mathbf{E} \approx \mathbf{E}(t)$
- Neglect vibration and rotation
  Pulse duration on sub femtosecond time scale
- Analyse the photo electron spectrum by Fourier transform of the outgoing wave

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
$H_{\mathrm{int}} = -q\mathbf{r}\cdot\mathbf{E}(t)$	$H_{\mathrm{int}} = -\frac{q}{m}\mathbf{p}\cdot\mathbf{A}(t)$	$\mathbf{r} \to \mathbf{r} - \alpha(t)$

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

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

The Kramers Henneberger frame

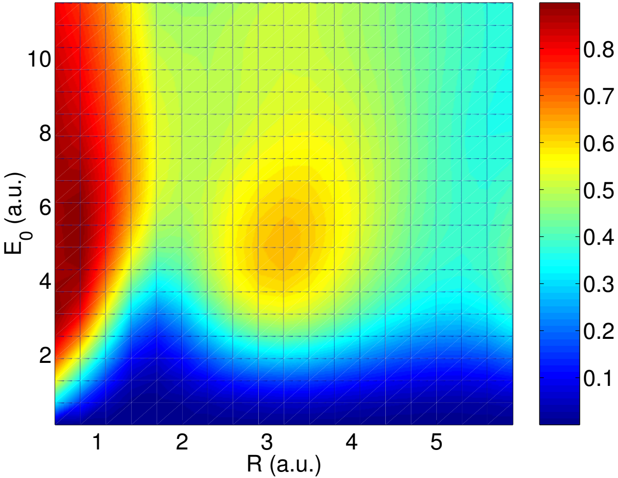
$$H_{\mathrm{KH}} = -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r} - \boldsymbol{\alpha}(t)) + \left(\frac{q^2}{2m}\mathbf{A}(t)\right)$$

The particle "sees" a moving potential
The transition from velocity gauge to KH frame can be done without the dipole approximation;

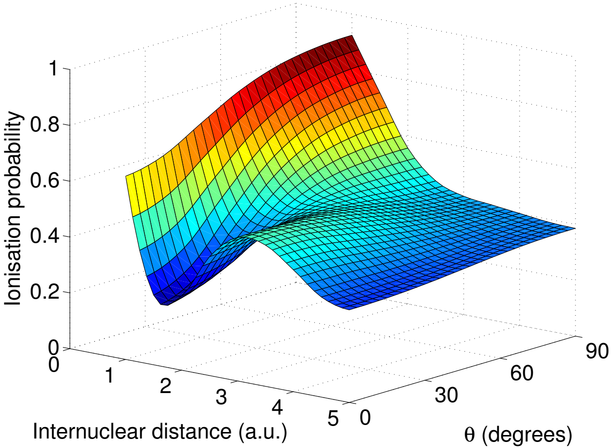
$$H_{\mathrm{KH}}^{\mathrm{ND}} \to -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r} - \boldsymbol{\alpha}(\mathbf{r}, t)) + \frac{q^2}{2m}\left(\mathbf{A}(\mathbf{r}, t)\right)^2$$

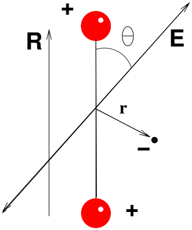
corresponding to a non-homogeneous field $\mathbf{E}(\mathbf{r}, t)$

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



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





Energetic and angular spectrum of the photo electron

Wave function in position space:

$$\Psi(r, \Omega, t_{\text{final}}) = \sum_{l,m} f_{l,m}(r, t_{\text{final}}) Y_{l,m}(\Omega)$$

Fourier transform of the outgoing wave:

$$\tilde{\Psi}_{\text{out}}(k, \Omega_k) = \mathcal{F}\{\Psi_{\text{out}}(r, \Omega, t_{\text{final}})\} = \sum_{lm} g_{l,m}(k) Y_{l,m}(\Omega_k)$$

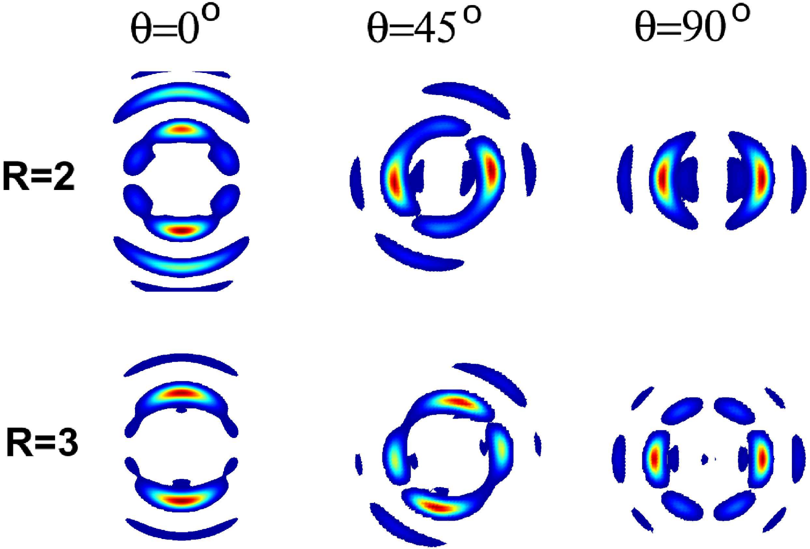
with

$$g_{l,m} = \sqrt{\frac{2}{\pi}}(-i)^l \int_a^\infty j_l(kr) f_{lm}(r, t_{\text{final}})\, r^2 dr$$

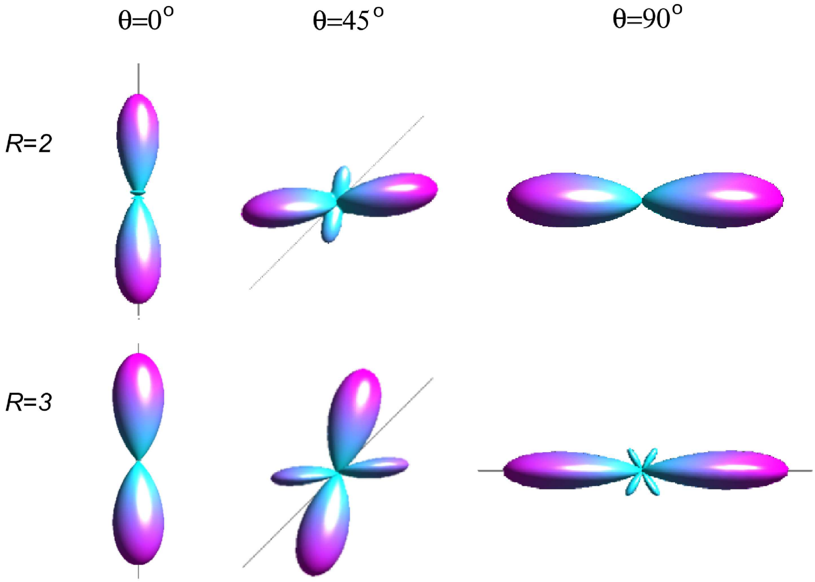
a: Separates outgoing wavepacket from bound wavepacket
Spectra:

$$\begin{aligned} \frac{dP_I}{dk} &= \int_{4\pi} |\tilde{\Psi}_{\text{out}}|^2 k^2 d\Omega_k \\ \frac{dP_I}{d\Omega_k} &= \int_0^\infty |\tilde{\Psi}_{\text{out}}|^2 k^2 dk \end{aligned}$$

Momentum wave functions



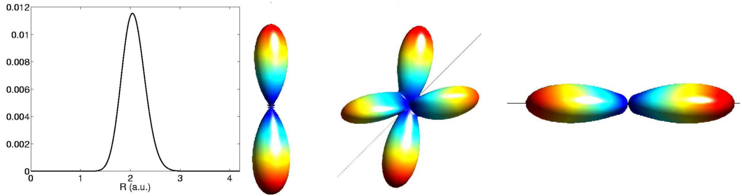
Angular distributions



But R is not sharply defined...

Use wavefunction averaged over various R,

$$\bar{\Psi}(t_{\mathrm{final}}) = \frac{1}{R_{\mathrm{max}} - R_{\mathrm{min}}} \int_{R_{\mathrm{min}}}^{R_{\mathrm{min}}} F(R)\Psi_R(t_{\mathrm{final}})dR$$



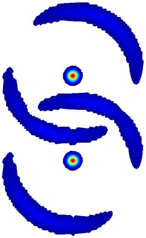
Model:

Interfering waves traveling from each of the protons:

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

For large r:

$$\psi_{\text{out}} \to 2f(\Omega)\cos(1/2\,k\mathbf{R}\cdot\hat{\mathbf{r}})\frac{e^{ikr}}{r}$$

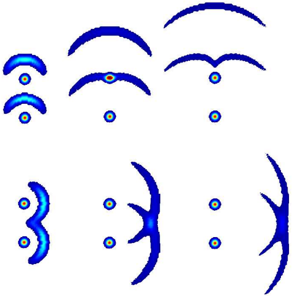


Model

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

- Explains oscillations for $\theta = 0^\circ$ and their absence for $\theta = 90^\circ$
- Does not explain the angular distribution for $\theta = 0^\circ$

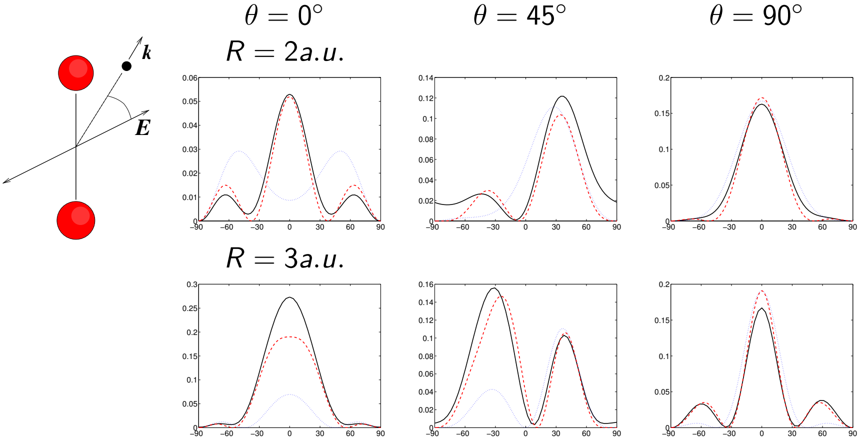
Reason: Coulomb scattering



Accounted for by the eikonal (WKB) approximation;

$$\exp(ikr_j) \to \exp\left(i\int_0^{r_j}\sqrt{E - V(\mathbf{r}'_j)}\hat{\mathbf{k}}\cdot d\mathbf{r}'_j\right), \quad j = 1, 2$$

For one photon ionization



Black: From TDSE

Blue: Simple interference model

Red: Including refraction

Non-dipole effects

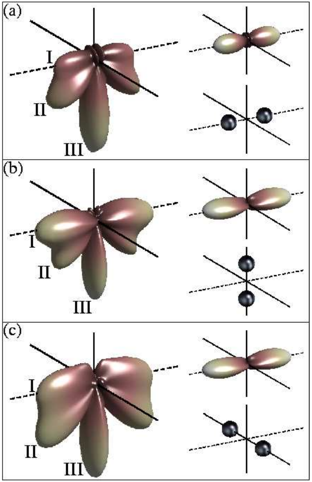
- For high photon energy $\omega\hbar$ and/or very strong fields: Dipole approximation breaks down.
- $\Rightarrow$ The influence of the magnetic field becomes significant.
- Treated by ND-version of the Kramers Henneberger frame:

$$H \approx \frac{p^2}{2m} + V(\mathbf{r} + \alpha(\mathbf{r}, t)) + \frac{1}{2m} \left(A(\mathbf{r}, t)\right)^2$$

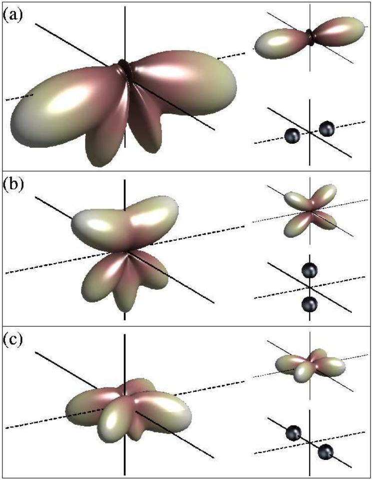
- Manifested in a "new lobe" in the angular distribution in the direction oposite to the propagation of the pulse.

Linear polarisation

Gerade initial state

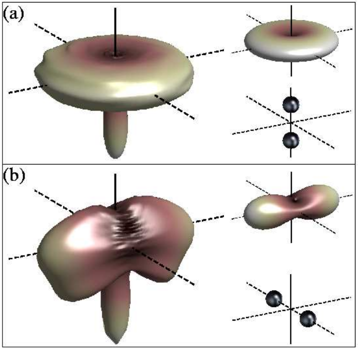


Ungerade initial state

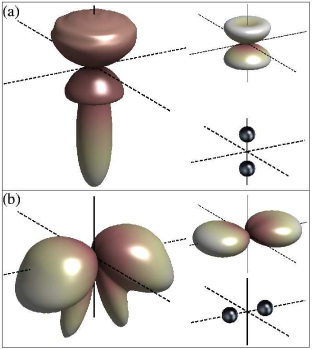


Circular polarisation

Gerade initial state



Ungerade initial state



Outlook:

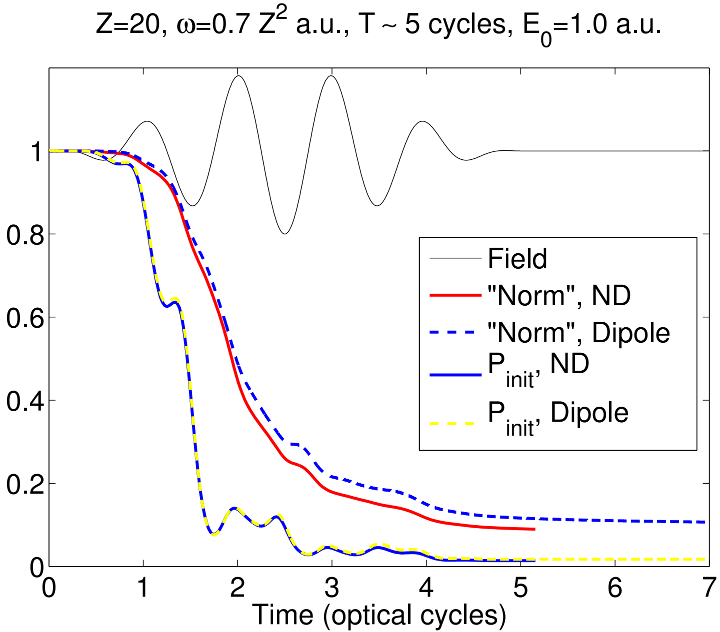
ND-treatment of relativistic, H-like systems in strong fields

Basis set of stationary solutions of the Dirac equation,

$$psi_{n,l,j,m} = \begin{pmatrix} r^{-1}F_{n,l}(r)\mathcal{Y}_{j,m,l}(\Omega) \\ ir^{-1}G_{n,l}(r)\mathcal{Y}_{j,m,l\pm1}(\Omega) \end{pmatrix}.$$

- Field: $A(\mathbf{r},t) \approx f(t)\sin(\omega t - \mathbf{k}\cdot\mathbf{r}) = 2\pi\mathbf{f(t)}\left(\mathbf{e^{i\omega t}}\sum_{\lambda,\mu}\mathbf{i}^{\lambda}\mathbf{j}_{\lambda}(\mathbf{kr})\mathbf{Y}^{*}_{\lambda,\mu}(\hat{\mathbf{k}})\mathbf{Y}_{\lambda,\mu}(\hat{\mathbf{r}}) + \mathbf{c.c.}\right).$
- Interaction: $c\boldsymbol{\alpha}\cdot\mathbf{A}(\mathbf{r},t)$.
- Couplings: $\langle\psi_{n',\kappa',j',m'}|H_I|\psi_{n,\kappa,j,m}\rangle \sim cf(t)\int G_{n',l'}j_{\lambda}(kr)F_{n,l}dr\,\langle l'\pm1,s,j',m'|\sigma_q Y_{\lambda,\mu}|l,s,j,m\rangle$.

$q \sim$ polarisation.



Z=20, ω=0.7 Z^2 a.u., T ~ 5 cycles, E$_0$=1.0 a.u.
Field
"Norm", ND
"Norm", Dipole
P$_{init}$, ND
P$_{init}$, Dipole
0
0.2
0.4
0.6
0.8
1
0
1
2
3
4
5
6
7
Time (optical cycles)