Ionization of atomic systems
Lecture 10

Beamer

Overview

time dependence, ω and/or γ dependence!

left over from last time:

- exit radius
- orbital, magnetic quantum number dependence m_ℓ

Consistency check

Adiabatic limit of a non-adiabatic theory

PPT: $\Gamma_{PPT} \propto \exp\left(-\frac{2 I_p}{\omega} f(\gamma, \varepsilon)\right)$

with:

$$f(\gamma, \varepsilon) = \left(1 + \frac{1+\varepsilon^2}{2\gamma^2}\right) \operatorname{arcsinh} \sqrt{\frac{\gamma^2 + s^2}{1-s^2}}$$

$$- \frac{1+\varepsilon^2 - 2\varepsilon s}{2\gamma^2 (1-s^2)} \sqrt{(1+\gamma^2)(s^2+\gamma^2)}$$

where $s \in [0, \varepsilon]$ parameter to fit the system (from transition point t_0)

quasistatic limit → consider every thing as linear $\Rightarrow \varepsilon = 0$

$$f(\gamma, 0) = \left(1 + \frac{1}{2\gamma^2}\right) \operatorname{arcsinh}(\gamma) - \frac{\sqrt{1+\gamma^2}}{2\gamma}$$

→ γ very small $\Rightarrow$ Taylor expansion up to second order

$$\operatorname{arcsinh}(\gamma) \approx \gamma - \frac{\gamma^3}{6} + \mathcal{O}(\gamma^5)$$

$$\sqrt{1+\gamma^2} \approx 1 + \frac{\gamma^2}{2} - \frac{\gamma^4}{8} + \mathcal{O}(\gamma^6)$$

(because of divisions)

$$\Rightarrow f(\gamma, 0) \approx \left(1 + \frac{1}{2\gamma^2}\right)\left(\gamma - \frac{\gamma^3}{6}\right) - \frac{1 + \frac{\gamma^2}{2} - \frac{\gamma^4}{8}}{2\gamma}$$

$$\approx \cancel{\frac{1}{2\gamma}} + \frac{11\gamma}{12} - \cancel{\frac{\gamma^3}{6}} - \cancel{\frac{1}{2\gamma}} - \frac{\gamma}{4} + \cancel{\frac{\gamma^3}{16}}$$

$$\approx \gamma\left(\frac{11}{12} - \frac{1}{4}\right) = \frac{2}{3}\gamma$$

$$\Rightarrow \Gamma_{PPT} \underset{\sim}{\propto} \exp\left(-\frac{2 I_p}{\omega} \cdot \frac{2}{3}\gamma\right)$$

$$\underset{\sim}{\propto} \exp\left(-\frac{2 \cdot 2 I_p}{\omega\, 3} \cdot \frac{\sqrt{2 I_p}}{F}\omega\right)$$

$$\underset{\sim}{\propto} \exp\left(-\frac{2 (2 I_p)^{3/2}}{3F}\right)$$

= Keldysh exponent !

q.e.d.

---

exit Radius

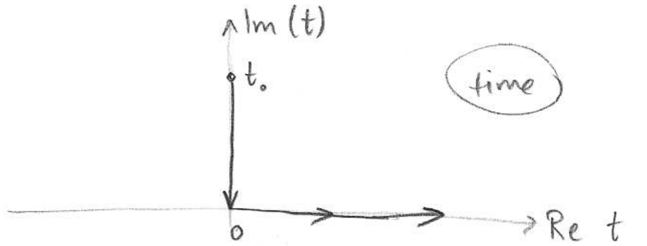
what we already have: t_0 complex time, "start of Ionisation process)

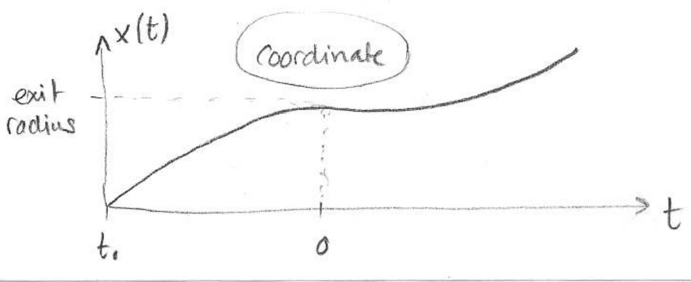
$Re(t_0)$ electron exits the barrier, enters the continuum.

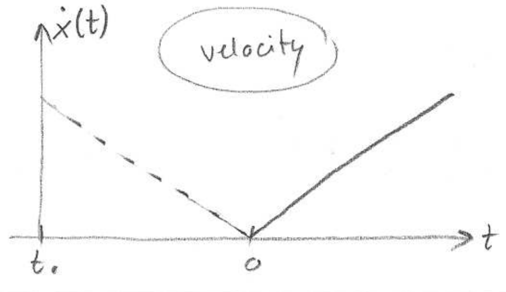
mixture between PPT, Bondar

to simplify: look at trajectory starting @ the peak of the field $\rightarrow$ $Re(t_0) = 0$

comic picture:







classical trajectories in elliptical laser field:

$$F(t) = F_{max} \begin{pmatrix} \cos(\omega t) \\ \varepsilon \sin(\omega t) \\ 0 \end{pmatrix}$$

are given by

$$\dot{x}(t) = \frac{F_{max}}{\omega} \sin(\omega t)$$

$$\dot{y}(t) = \pm p_0 - \frac{\varepsilon F_{max}}{\omega} \cos(\omega t)$$

$$\dot{z}(t) = 0$$

assuming $\dot{x}(0) = 0$. Using $x(t_0) = y(t_0) = 0$:

$$x(t) = \frac{F_{max}}{\omega^2} \left(\cos(\omega t_0) - \cos(\omega t) \right)$$

$$y(t) = \frac{\varepsilon F_{max}}{\omega^2} \left(\sin(\omega t_0) - \sin(\omega t) \right) \pm p_0 (t - t_0)$$

so, exit radius @ $t = 0$:

$$x_e = \frac{F_{max}}{\omega^2} \left(\cos(\omega t_0) - 1 \right)$$

$$y_e = \frac{\varepsilon F_{max}}{\omega^2} \sin(\omega t_0) \mp p_0 \cdot t_0 \overset{!}{=} 0$$

for $y(t)$ to be real @ $t \to \infty$

$$\boxed{\Rightarrow p_0(\gamma, \varepsilon)}$$

what is t_0?

remember the energy transition point

$$E_i(t_0) = E_f(t)$$

$$-I_p = \frac{1}{2} \left(\dot{x}(t_0)^2 + \dot{y}(t_0)^2 \right)$$

$$\Rightarrow \sin(\omega t_0) = -i \sqrt{\frac{s^2 + \gamma^2}{1 - s^2}}$$ again, $s \in [0, \varepsilon]$

use $\cos(\omega t_0) = \sqrt{1 - \sin^2(\omega t_0)}$

$$\Rightarrow x_e = \frac{F_{max}}{\omega^2} \left(\sqrt{\frac{1 + \gamma^2}{1 - s^2}} - 1 \right) = \frac{2 I_p}{F \gamma^2} \left(\sqrt{\frac{1 + \gamma^2}{1 - s^2}} - 1 \right)$$

Beamer

Orbital effects

Start from an experiment:

Herath et. al, PRL 2012

Beamer

Argon ionised by 2 pulses

$$Ar \rightarrow Ar^{+} \rightarrow Ar^{2+}$$

$\Rightarrow$ Ar^{2+} yield with $\boxed{LR}$ rotating pulses almost 4 times!

let's calculate what happens:

→ apriori unknown ionisation rates

- $W_{m\,L/R}$ from pump
- $W'_{m\,L/R}$ from probe
- symmetry: $\begin{cases} W_{-L} = W_{+R} \\ W_{0L} = W_{0R} \\ W_{+L} = W_{-R} \end{cases}$

→ always ionise two electrons from different m_e

then:
$$\begin{aligned} I_{SDI-LR} &= (W_{-L} + W_{+L}) W'_{0R} + (W_{-L} + W_{0L}) W'_{+R} \\ &\quad + (W_{0L} + W_{+L}) W'_{-R} \\ &= (W_{+R} + W_{-R}) W'_{0R} + (W_{+R} + W_{0R}) W'_{+R} \\ &\quad + (W_{0R} + W_{-R}) W'_{-R} \end{aligned}$$

$$\begin{aligned} I_{SDI-RR} &= (W_{-R} + W_{+R}) W'_{0R} + (W_{-R} + W_{0R}) W'_{+R} \\ &\quad + (W_{0R} + W_{+R}) W'_{-R} \end{aligned}$$

devide by $(W_{+R} W'_{+R})$

define $\alpha = \frac{W_{-R}}{W_{+R}}$ $\beta = \frac{W_{0R}}{W_{+R}}$

then the yield ratio is given by

$$\frac{I_{SDI-LR}}{I_{SDI-RR}} = \frac{(1+\alpha)\beta' + (1+\beta) + (\beta+\alpha)\alpha'}{(\alpha+1)\beta' + (\alpha+\beta) + (\beta+1)\alpha'} \overset{!}{=} 3.63$$

measurement

$\beta \ll \alpha$ (calculations: factor ~ 20)

$$\approx \frac{1+\alpha\alpha'}{\alpha+\alpha'} \overset{!}{=} 3.63$$

Solve for α' $\Rightarrow$ $\alpha' = \frac{1 - 3.63\alpha}{3.63-\alpha}$

Beamer

remember: $\alpha = \frac{W_{-R}}{W_{+R}}$

even if $\varkappa = \alpha^{-1}$ is defined, the equation for $\varkappa$ and $\varkappa'$ would be equivalent

Simplified picture

calculations show ratios of

$$W_- : W_+ : W_0 \approx 1 : \frac{1}{3} : \frac{1}{20}$$

for righthand rotation, favouring counter rotating orbitals.

Idea: rotating field creates a doorway for the electron to tunnel through.
counter-rotating electrons see this doorway more often.

strategy for calculating m - dependence

back in SFA / Keldysh theory derivation:

$$\langle \vec{v} \,|\, \Phi(t) \rangle = \Phi(\vec{v}, t)$$ transition amplitude

with $$\Phi(\vec{v},t) \propto \int_0^t dt' \, \langle \vec{v} \,|\, \underbrace{\hat{H}(t,0,t')}\,|\, \Phi_i \rangle$$

final state @ detector

initial state

- brings Φ_i to time t',
- interaction with field @ t'
- propagation from t' to t

similarly:

$$A_{\vec{p}n} \propto \int_0^T \Psi^*_{\vec{p}}(\vec{r},t) \; V_F(t) \, \Psi_0(\vec{r},t) \, dt$$

wave function in the continuum, $E_{\vec{p}} = \frac{1}{2}\vec{p}^{\,2} + \frac{F_{max}^2 e^2}{4\omega^2}$

Ponderomotive Energy in $F_{max}\cos(\omega t)$ field

field interaction

$= e^{-iE_0 t}\, \Phi_i(\vec{r})$

Coulomb field neglected

Volkov Wave function

plane wave

propagator

$$\Psi_{\vec{p}}(\vec{r},t) = \exp\left(i(\vec{p} + \vec{k}_t)\cdot\vec{r} - \frac{i}{2}\int^t (\vec{p} + \vec{k}_{t'})^2 \, dt' \right)$$

with $\vec{k}_t = e \cdot \int^t \vec{F}(t')\, dt'$ momentum due to the field

$$\Phi_i(\vec{r}) = A \cdot R(r) \cdot Y_{\ell m}(\vartheta, \varphi)$$

$\Rightarrow$ m dependence is in the prefactor we previously neglected!

$$Y_{\ell m}(\vartheta,\varphi) = \frac{1}{\sqrt{2\pi}} e^{im\varphi} (-1)^{\frac{m+|m|}{2}} \sqrt{\frac{2\ell+1}{2}\frac{(\ell-|m|)!}{(\ell+|m|)!}} \cdot P_\ell^{|m|}(\cos\vartheta)$$

Legendre polynomial

then, $|A_{\vec{p}n}|^2$ is the probability of ionising an atom such that the electron carries a final momentum $\vec{p}$.

$\Rightarrow$ only $|e^{im\varphi}|^2$ depends on $\text{sgn}(m)$, $\rightarrow$ non-adiabatic

the rest depends on $|m|$. $\rightarrow$ adiabatic

because: non-adiabatic: φ is a generalised, complex angle

from $\cos\varphi = \frac{(\vec{p}_{\parallel} - \vec{A}(t_0))\cdot\hat{x}}{|\vec{p}_{\parallel} - A(t_0)|}$

where t_0 is the transition point (complex)

adiabatic limit, $(p_{\parallel} - A(t_0))\hat{x} \rightarrow 0$

$\Rightarrow \varphi = \pm \pi \quad \Rightarrow |e^{im\varphi}|^2$ independent of $\text{sign}(m)$