Ionization of atomic systems
Lecture 8

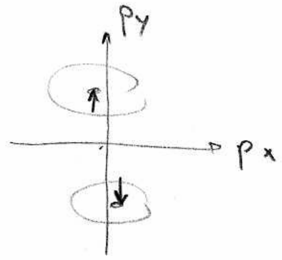
Non-adiabatic Theory

Phenomenology

collect list

- $\gamma \gtrsim 1$ non-adiabatic tunnelling
- $v_\perp$, initial $\Rightarrow$ blobs in momentum distro further apart
- wider spreads (transverse and longitudinal)

  today: reason for that



- dependence on $\mathrm{sgn}(m_e)$ of ionisation probability
- shorter exit radius

Instantaneous ionisation rate without initial momentum assumption

Bondar, PRA 78 2008

build up on Keldys theory, • assume slowly varying, but time dependent
• neglect Coulomb potential

probability of transition $\Psi_i \to \Psi_f$ (Dykhne method)

$$\Gamma = \exp(-2\,\mathrm{Im}(S))$$

$$S = \int_{t_1}^{t_0} [E_f(t) - E_i(t)]\,dt \qquad (1)$$

t_1 real

t_0 such that
- $E_i(t_0) = E_f(t_0)$ transition point
- closest to real axis
- $\mathrm{Im}(t_0) > 0$

System and situation:
- single electron ionisation
- linear polarisation $\vec{F}(t) = \vec{F} \cdot \cos(\omega \cdot t)$

  $\vec{A}(t) = \frac{-\vec{F}}{\omega} \sin(\omega \cdot t)$
- $\vec{k}$ canonical momentum measured on detector

  $\vec{k} = \vec{v} + q \cdot \vec{A}$
- $E_i(t) = -I_p$, $\quad E_f(t) = \frac{1}{2} v(t)^2 = \frac{1}{2}[\vec{k} + \vec{A}(t)]^2 \qquad (2)$

then:

$$S(\vec{k}, I_p) = \int_{t_1}^{t_0} \Big(\underbrace{\tfrac{1}{2}[k_\parallel + A(t)]^2}_{\text{one coord.}} + \underbrace{\tfrac{1}{2}k_\perp^2}_{\text{2 coord.}} + I_p \Big)\, dt \qquad (3)$$

almost exactly like the action in SFA derivation!
(slightly different definitions of times)

Find transition point t_0 such that $E_i(t_0) = E_f(t_0)$:

$$\frac{\partial}{\partial t_0} S(\vec{k}, I_p) \overset{!}{=} 0 \qquad \text{(cf. saddle point approximation)}$$

$$\tfrac{1}{2}[k_\parallel + A(t_0)]^2 + \tfrac{1}{2}k_\perp^2 + I_p = 0$$

$$\tfrac{1}{2}[k_\parallel - \tfrac{F}{\omega}\sin(\omega t_0)]^2 + \tfrac{1}{2}k_\perp^2 + I_p = 0 \qquad \Big/ \cdot \frac{2\omega^2}{F^2}$$

$$\left[\frac{k_\parallel \omega}{F} - \sin(\omega t_0)\right]^2 + \gamma^2\left(\frac{k_\perp^2}{2I_p} + 1\right) = 0 \qquad /\pm\sqrt{}$$

$$\text{since } \gamma = \frac{\sqrt{2I_p}}{F}\omega$$

no additional approximations in this!

$$\sin(\omega t_0) = \pm i\gamma\sqrt{\frac{k_\perp^2}{2I_p} + 1} + \frac{k_\parallel \omega}{F}$$

($\pm$ Im Part doesn't matter...)

$$t_0 = \frac{1}{\omega} \cdot \arcsin\left[\gamma\left(\frac{k_\parallel}{\sqrt{2I_p}} + i\sqrt{\frac{k_\perp^2}{2I_p} + 1}\right)\right] \qquad (4)$$

Handbook of mathematical functions:

$$\arcsin(x + iy) = 2K\pi + \arcsin\beta + i\log(\alpha + \sqrt{\alpha^2 - 1})$$

$$\text{with } K \in \mathbb{N}, \quad \begin{Bmatrix}\alpha \\ \beta\end{Bmatrix} = \tfrac{1}{2}\sqrt{(x+1)^2 + y^2} \pm \tfrac{1}{2}\sqrt{(x-1)^2 + y^2}$$

made sure that $\mathrm{Im}(t_0) > 0$

- solve integral (3):

$$S(\vec{k}, I_p) = \frac{k^2}{2}(t_0 - t_1) + \frac{F \cdot k_\parallel}{\omega^2}\left(\cos(t_0\omega) - \cos(t_1\omega)\right)$$

no approximation here!

$$+ \frac{F^2}{8\omega^3}\left(\sin(2t_1\omega) - \sin(2t_0\omega)\right)$$

$$+ I_p(t_0 - t_1) \qquad (5)$$

$t_1 \in \mathbb{R}$, choose $t_1 = \mathrm{Re}(t_0)$, take $\mathrm{Im}(S(\vec{k}, I_p))$

Algebra simplify

Beames: Mathematica

$\gamma = \frac{\sqrt{2I_p}}{F}\omega$

$\Rightarrow$ Bondar instantaneous ionisation probability

$k_\parallel = p_\parallel^{\text{initial}} - A(t)$ $\Rightarrow$ time dependent formulation

$$\Gamma(\gamma, p_\parallel, k_\perp, t) \propto \exp\left(-\frac{2 I_p}{\omega} f(\gamma, p_\parallel, k_\perp, t)\right) \qquad (6)$$

with

$$f(\gamma, p_\parallel, k_\perp, t) = \left(1 + \frac{1}{2\gamma^2} + \frac{k_\perp^2 + (p_\parallel - A(t))^2}{2 I_p}\right)\operatorname{arccosh}(\alpha) \qquad (7)$$

$$- \sqrt{\alpha^2 - 1}\left(\frac{\beta}{\gamma}\sqrt{\frac{2}{I_p}}\left(p_\parallel - A(t)\right) + \alpha\frac{(1 - 2\beta^2)}{2\gamma^2}\right)$$

$$\begin{Bmatrix}\alpha \\ \beta\end{Bmatrix} = \frac{\gamma}{2}\left(\sqrt{\frac{k^2}{2I_p} + \frac{2}{\gamma}\frac{k_\parallel}{\sqrt{2I_p}} + \frac{1}{\gamma^2} + 1} \pm \sqrt{\frac{k^2}{2I_p} - \frac{2}{\gamma}\frac{k_\parallel}{\sqrt{2I_p}} + \frac{1}{\gamma^2} + 1}\right) \qquad (8)$$

where $k^2 = k_\perp^2 + (p_\parallel - A(t))^2$, $k_\parallel = p_\parallel^{\text{initial}} - A(t)$

compare Bondar to PPT

$$\Gamma_{PPT} \propto \exp\left(-\frac{2I_p}{\omega} g(\gamma, \varepsilon)\right) \cdot \exp\left(\frac{v_x^2}{2\sigma_x^2}\right)$$
$$\cdot \exp\left(\frac{(v_y \pm v_0)^2}{2\sigma_y^2}\right)$$
$$\cdot \exp\left(\frac{v_z^2}{2\sigma_z^2}\right)$$

similar structure, but v_x, v_y, v_z dependence in Bondar hidden in $f(\gamma, p_\parallel, k_\perp, t)$

Mathematica: Comparison PPT vs. Bondar

Beamer

→ transverse and time dependence match

→ longitudinal momentum dependence close