Keldysh Theory

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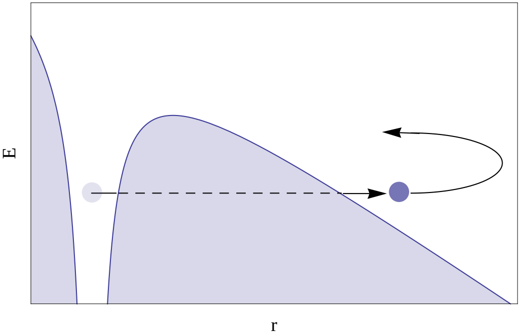
Goal

derive ionisation probability

$$P(t_0) \propto \exp\left(-\frac{2(2I_{\mathrm{p}}(t_0))^{3/2}}{3F(t_0)}\right)\exp\left(\frac{v_\perp^2}{2\sigma_\perp^2}\right)$$

- Keldysh exponent
- ADK transverse velocity probability



Resources - Units

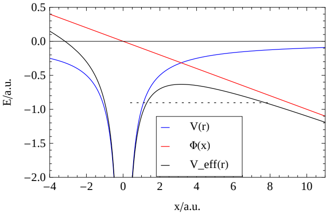
- M. Y. Ivanov, M. Spanner and O. Smirnova, *Anatomy of Strong Field Ionization*, Journal of Modern Optics, 52:2-3, 165-184
- A. S. Landsman, *Laser-Atom Interaction*, Lecture notes FS 2011

Atomic units to make calculations/formulas easier:

- $\hbar$ = 1
- $|q_e|$ = 1
- m_e = 1

Outline

Laser-Atom system



- what we have to calculate:

$$\boxed{i\frac{\partial}{\partial t}|\Psi\rangle = \hat{H}(t)|\Psi\rangle} \qquad \text{with:} \quad \hat{H}(t) = \hat{H}_0 + \hat{V}_L(t) \qquad (1)$$

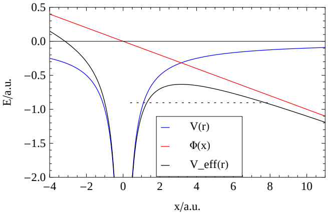
field-free Hamiltonian of the atom

interaction with the laser field

$$\Psi = \Psi(t, \vec{r}_1, \ldots \vec{r}_n)$$

- full multi-electron wave function
- explicitly time-dependant Hamiltonian (no separation ansatz)

Laser-Atom system



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field-free Hamiltonian of the atom

interaction with the laser field

$$\Psi = \Psi(t, \vec{r}_1, \ldots \vec{r}_n)$$

- full multi-electron wave function
- explicitly time-dependant Hamiltonian (no separation ansatz)

- general solution:

$$|\Psi(t)\rangle = e^{-i\int_{t_0}^{t} \mathrm{d}t'\,\hat{H}(t')}|\Phi_i\rangle \tag{2}$$

Approximations I

- single active electron

$$\Psi(t, \vec{r}_1, \ldots \vec{r}_n) \approx \Psi_{n-1}(\vec{r}_1, \ldots \vec{r}_{n-1}) \times \Phi(\vec{r}_n, t)$$

- dipole approximation: $\lambda >> r(\text{atom}) \Rightarrow \vec{F}(t)$

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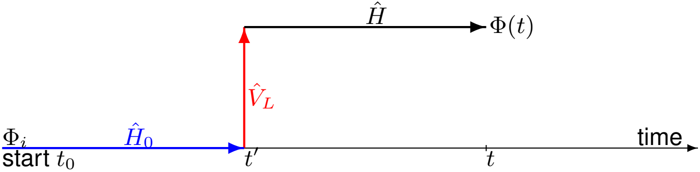
- dipole approximation: $\lambda >> r(\mathrm{atom}) \Rightarrow \vec{F}(t)$
- SFA 1: neglect laser field while in bound state
- adiabatic case (quasi-static approximation) $\omega << \omega_B$
- **exact** solution:

$$|\Phi(t)\rangle = -i\int_{t_0}^{t} \mathrm{d}t' \left[e^{-i\int_{t'}^{t}\mathrm{d}t''\hat{H}(t'')}\right] \hat{V}_L(t') \left[e^{-i\int_{t_0}^{t'}\mathrm{d}t''\hat{H}_0(t'')}\right] |\Phi_i\rangle + e^{-i\int_{t_0}^{t}\mathrm{d}t''\hat{H}_0(t'')}|\Phi_i\rangle \quad (3)$$



Solution Check

Subsitute (3) into Schrödinger's Equation:

$$i\frac{\partial|\Phi(t)\rangle}{\partial t} = i\frac{\partial}{\partial t}\left(-i\int_{t_0}^{t} \mathrm{d}t' \left[e^{-i\int_{t'}^{t} \mathrm{d}t''\hat{H}(t'')}\right] \hat{V}_L(t') \left[e^{-i\int_{t_0}^{t'} \mathrm{d}t''\hat{H}_0(t'')}\right] |\Phi_i\rangle + e^{-i\int_{t_0}^{t} \mathrm{d}t''\hat{H}_0(t'')} |\Phi_i\rangle \right)$$

$$= \frac{\partial}{\partial t}\left(\int_{t_0}^{t} \mathrm{d}t' \left[e^{-i\int_{t'}^{t} \mathrm{d}t''\hat{H}(t'')}\right] \hat{V}_L(t') \left[e^{-i\int_{t_0}^{t'} \mathrm{d}t''\hat{H}_0(t'')}\right] |\Phi_i\rangle + ie^{-i\int_{t_0}^{t} \mathrm{d}t''\hat{H}_0(t'')} |\Phi_i\rangle \right)$$

$$= -i\hat{H}(t) \int_{t_0}^{t} \mathrm{d}t' \left[e^{-i\int_{t'}^{t} \mathrm{d}t''\hat{H}(t'')}\right] \hat{V}_L(t') \left[e^{-i\int_{t_0}^{t'} \mathrm{d}t''\hat{H}_0(t'')}\right] |\Phi_i\rangle + \hat{V}_L(t) e^{-i\int_{t_0}^{t} \mathrm{d}t''\hat{H}_0(t'')} |\Phi_i\rangle \quad + \hat{H}_0(t) e^{-i\int_{t_0}^{t} \mathrm{d}t''\hat{H}_0(t'')} |\Phi_i\rangle$$

$$= \hat{H}(t)|\Phi(t)\rangle \qquad \square$$

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quasi-static:

$$= \frac{\partial}{\partial t}\Bigg(\int_{t_0}^{t} \mathrm{d}t' \left[e^{-i(t-t')\hat{H}}\right] \hat{V}_L(t') \left[e^{-i(t'-t_0)\hat{H}_0}\right]|\Phi_i\rangle + ie^{-i(t-t_0)\hat{H}_0}|\Phi_i\rangle \Bigg)$$

$$= -i\hat{H}\int_{t_0}^{t} \mathrm{d}t' \left[e^{-i(t-t')\hat{H}}\right] \hat{V}_L(t') \left[e^{-i(t'-t_0)\hat{H}_0}\right]|\Phi_i\rangle + e^{-i(t-t)\hat{H}}\hat{V}_L(t)e^{-i(t-t_0)\hat{H}_0}|\Phi_i\rangle \qquad + \hat{H}_0 e^{-i(t-t_0)\hat{H}_0}|\Phi_i\rangle$$

$$= \hat{V}_L(t)|\Phi(t)\rangle + \hat{H}_0|\Phi(t)\rangle = \hat{H}(t)|\Phi(t)\rangle \qquad \square$$

Basis in the Continuum

- velocity basis $|\vec{v}\rangle$
- eigenstates: plane waves $e^{i\vec{\kappa}\vec{r}}$ for $\vec{r}$ large
- no projection of ground state onto continuum states: $\langle\vec{v}|\Phi_i\rangle = 0$

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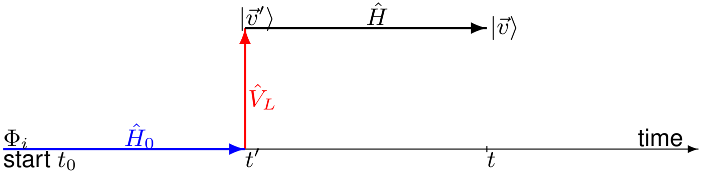
- velocity basis $|\vec{v}\rangle$
- eigenstates: plane waves $e^{i\vec{\kappa}\vec{r}}$ for $\vec{r}$ large
- no projection of ground state onto continuum states: $\langle\vec{v}|\Phi_i\rangle = 0$
- projection of the evolved wave function onto a specific velocity $\vec{v} = (v_x, v_y, v_z)$:

$$\begin{aligned}\langle\vec{v}|\Phi(t)\rangle &= \Phi(\vec{v},t)\\ &= -i\int_{t_0}^{t} \mathrm{d}t' \left\langle \vec{v}\middle| e^{-i\int_{t'}^{t} \mathrm{d}t''\hat{H}(t'')}\hat{V}_L(t')e^{-i\int_{t_0}^{t'} \mathrm{d}t''\hat{H}_0(t'')}\middle|\Phi_i\right\rangle \qquad (4)\end{aligned}$$



Approximations II

- SFA 2: neglect ion field in continuum

  → Volkov propagator with $\hat{H}_F = \frac{1}{2}\left(\vec{p} - q\vec{A}\right)^2$

$$\langle \vec{v} | e^{-i \int_{t'}^{t} \hat{H}_{\mathrm{F}}(t'')\, \mathrm{d}t''} = e^{-i \int_{t'}^{t} \frac{1}{2}\left[\vec{p} + \mathbf{A}(t'')\right]^2 \mathrm{d}t''} \langle \vec{v'} | = e^{-i \int_{t'}^{t} \frac{1}{2} \vec{v'}^2 \, \mathrm{d}t''} \langle \vec{v'} |$$

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$$\langle\vec{v}|e^{-i\int_{t'}^{t}\hat{H}_{\mathrm{F}}(t'')\,\mathrm{d}t''} = e^{-i\int_{t'}^{t}\frac{1}{2}\left[\vec{p}+\mathbf{A}(t'')\right]^2\mathrm{d}t''}\langle\vec{v'}| = e^{-i\int_{t'}^{t}\frac{1}{2}\vec{v\prime}^2\,\mathrm{d}t''}\langle\vec{v'}|$$

- ground state has energy $-I_{\mathrm{p}}$

$$\begin{aligned}\Phi(\vec{v},t) &= -i\int_{t_0}^{t}\mathrm{d}t'\left\langle\vec{v}\middle|e^{-i\int_{t'}^{t}\hat{H}_{\mathrm{F}}(t'')\,\mathrm{d}t''}\hat{V}_L(t')e^{-i\int_{t_0}^{t'}\mathrm{d}t''\hat{H}_0(t'')}\middle|\Phi_i\right\rangle \\ &= -i\int_{t_0}^{t}\mathrm{d}t'e^{-i\int_{t'}^{t}\frac{1}{2}\vec{v\prime}^2\,\mathrm{d}t''}e^{i(t'-t_0)I_{\mathrm{p}}}\left\langle\vec{v'}\middle|\hat{V}_L(t')\middle|\Phi_i\right\rangle \end{aligned} \qquad (5)$$

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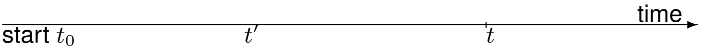
- the canonical momentum $\vec{p} = \vec{v} + q\vec{A}$ is conserved

$$\vec{v} - \vec{A}(t) = \vec{v}^* - \vec{A}(t^*) \quad \Rightarrow \quad \vec{v}^* = \vec{v} - \vec{A}(t) + \vec{A}(t^*)$$

$$\begin{aligned} \Phi(\vec{v},t) = -i\int_{t_0}^{t} \mathrm{d}t' e^{-i\int_{t'}^{t} \frac{1}{2}\left(\vec{v}-\vec{A}(t)+\vec{A}(t'')\right)^2 \mathrm{d}t''} e^{i(t'-t_0)I_\mathrm{p}} \\ \times \left\langle \vec{v} - \vec{A}(t) + \vec{A}(t') \middle| \hat{V}_L(t') \middle| \Phi_i \right\rangle \qquad (6) \end{aligned}$$

Intuitive picture so far

$$\Phi(\vec{v},t) = -i\int_{t_0}^{t} \mathrm{d}t' e^{-i\int_{t'}^{t}\frac{1}{2}\left(\vec{v}-\vec{A}(t)+\vec{A}(t'')\right)^2 \mathrm{d}t''} e^{i(t'-t_0)I_\mathrm{p}} \times \underbrace{\left\langle \vec{v}-\vec{A}(t)+\vec{A}(t') \middle| \hat{V}_L(t') \middle| \Phi_i \right\rangle}_{\text{prefactor}} \tag{7}$$



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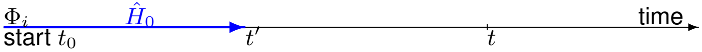

- waiting in groundstate

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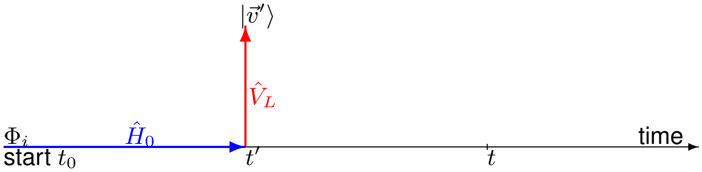

- waiting in groundstate
- kick from laser field, jump up to continuum state, exit tunnel with velocity $\vec{v}'$

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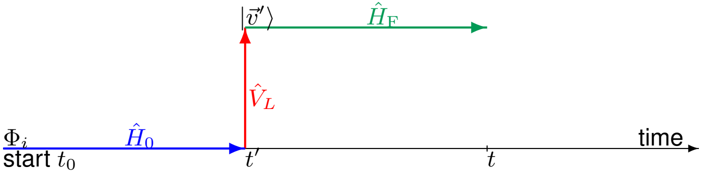

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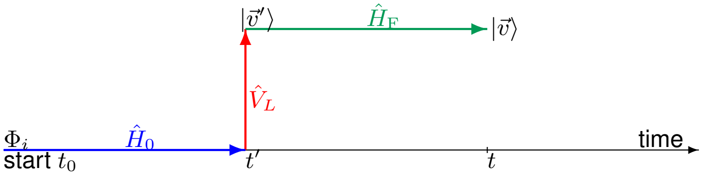
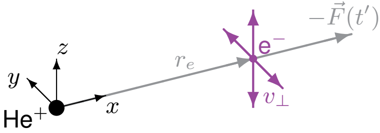


- waiting in groundstate
- kick from laser field, jump up to continuum state, exit tunnel with velocity $\vec{v}'$
- oscillating in the laser field
- recorded on detector with velocity $\vec{v}$

Linear polarisation

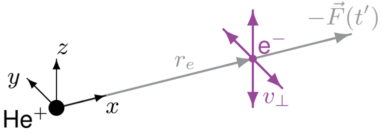


- laser field polarised along $\hat{x}$
  → vector potential:

$$\vec{A}(t) = \frac{F_0}{\omega}\sin(\omega t)\hat{x} \tag{8}$$

$$\Rightarrow \quad \left(\vec{v} - \vec{A}(t) + \vec{A}(t'')\right)^2 = \left(v_x - \frac{F_0}{\omega}\sin(\omega t) + \frac{F_0}{\omega}\sin(\omega t'')\right)^2 + v_y^2 + v_z^2 \tag{9}$$

Linear polarisation



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- define action

$$S_{\vec{v}}(t,t') := \frac{1}{2}\int_{t'}^{t}\left[v_x - \frac{F_0}{\omega}\sin(\omega t) + \frac{F_0}{\omega}\sin(\omega t'')\right]^2 \mathrm{d}t'' + \frac{v_y^2 + v_z^2}{2}(t-t') - I_\mathrm{p}(t'-t_0) \tag{10}$$

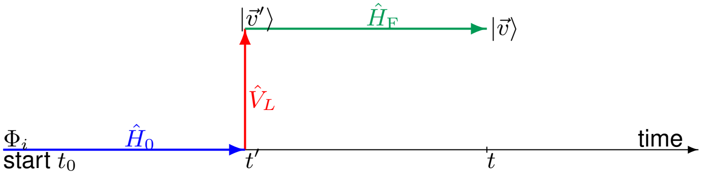
$$\Phi(\vec{v},t) \propto -i\int_{t_0}^{t} \mathrm{d}t' e^{-i\int_{t'}^{t}\frac{1}{2}\left(\vec{v}-\vec{A}(t)+\vec{A}(t'')\right)^2 \mathrm{d}t''} e^{i(t'-t_0)I_\mathrm{p}}$$

$$\Rightarrow \boxed{\Phi(\vec{v},t) \propto -i\int_{t_0}^{t}\exp\left(-iS_{\vec{v}}(t,t')\right)\,\mathrm{d}t'} \tag{11}$$

Recap

- That was the hard part. ☺
- **Probability amplitude** of finding an electron
  - with velocity $\vec{v}$ on the detector,
  - from **any** ionisation time t'.

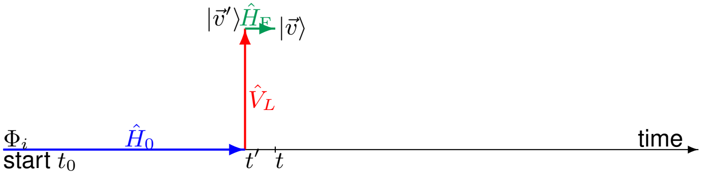
$$\Phi(\vec{v},t) \propto -i \int_{t_0}^{t} \exp\left(-iS_{\vec{v}}(t,t')\right)\,\mathrm{d}t'$$



Outlook

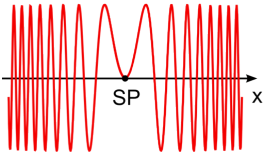
- Now comes the fun part.
- Find the **probability of ionisation** depending on
  - ionisation time t'
  - assumption: instantaneous tunneling time
  - and tunnel exit velocity $\vec{v}'$.

$$P(t', \vec{v}') = ?$$



- set $t_0 = 0$
- push t towards t' and all that towarts t_0
- we have to evaluate $\int_{t_0}^{t} \exp\left(-iS_{\vec{v}}(t, t')\right) \,\mathrm{d}t'$

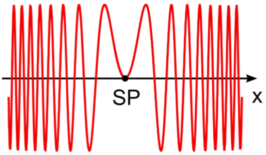
Saddle point approximation I



- $\int_{x_1}^{x_2} f(x) e^{ig(x)} \, \mathrm{d}x$
- $f(x)$ slowly varying (in our case: $f(x) \equiv 1$)
- main contribution where $g'(x) = 0$ (no fast oscillations which cancel contributions out)

$$S_{\vec{v}}(t, t') := \frac{1}{2} \int_{t'}^{t} \left[v_x - \frac{F_0}{\omega} \sin(\omega t) + \frac{F_0}{\omega} \sin(\omega t'') \right]^2 \mathrm{d}t'' + \frac{v_y^2 + v_z^2}{2} (t - t')$$
$$-I_\mathrm{p} t'$$

Saddle point approximation I



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$$S_{\vec{v}}(t,t') := \frac{1}{2}\int_{t'}^{t}\left[v_x - \frac{F_0}{\omega}\sin(\omega t) + \frac{F_0}{\omega}\sin(\omega t'')\right]^2 \mathrm{d}t'' + \frac{v_y^2+v_z^2}{2}(t-t') - I_{\mathrm{p}}t'$$

- find a particular t'_* such that $\left.\frac{\delta S_{\vec{v}}(t,t')}{\delta t'}\right|_{t'_*} = 0$

$$0 = \frac{-1}{2}\left[v_x - \frac{F_0}{\omega}\sin(\omega t) + \frac{F_0}{\omega}\sin(\omega t'_*)\right]^2 - \frac{v_y^2+v_z^2}{2} - I_{\mathrm{p}}$$

Saddle point approximation II

$$0 = \frac{1}{2}\left[v_x - \frac{F_0}{\omega}\sin(\omega t) + \frac{F_0}{\omega}\sin(\omega t'_*)\right]^2 + \frac{v_y^2 + v_z^2}{2} + I_\mathrm{p}$$

- define a varied Keldysh parameter $\left(\text{usually: } \gamma = \frac{\sqrt{2I_\mathrm{p}}}{F_0}\omega\right)$

$$\tilde{\gamma} = \frac{\sqrt{2I_\mathrm{p} + v_y^2 + v_z^2}}{F_0}\omega \qquad (12)$$

$\Rightarrow$ perpendicular velocity “adds” to the ionisation potential

- final equation to solve in order to find the saddle point:

$$0 = \left[\frac{v_x\omega}{F_0} - \sin(\omega t) + \sin(\omega t'_*)\right]^2 + \tilde{\gamma}^2 \qquad (13)$$

Special Case: $\vec{v} = 0$

$$0 = \left[\frac{v_x \omega}{F_0} - \sin(\omega t) + \sin(\omega t'_*) \right]^2 + \tilde{\gamma}^2$$

- let's find $|\Phi(\vec{v} = 0, \omega t = n\pi)|^2$

$$v_x = 0 \qquad \tilde{\gamma} = \gamma \qquad \sin(\omega t) = 0$$

$$\sin(\omega t'_*) = \pm i\gamma$$

Special Case: $\vec{v} = 0$

$$0 = \left[\frac{v_x \omega}{F_0} - \sin(\omega t) + \sin(\omega t'_*)\right]^2 + \tilde{\gamma}^2$$

- let's find $|\Phi(\vec{v} = 0, \omega t = n\pi)|^2$

$$v_x = 0 \qquad \tilde{\gamma} = \gamma \qquad \sin(\omega t) = 0$$

$$\sin(\omega t'_*) = \pm i\gamma$$

- ionisations only happen when the field is strong enough $\Rightarrow \omega t'_* << 1$

$$\omega t'_* \approx \pm i\gamma$$

$$t'_* \approx i\frac{\gamma}{\omega} = i\frac{\sqrt{2I_\mathrm{p}}}{F_0} = i\tau_\mathrm{Keldysh} \tag{14}$$

Special Case: $\vec{v} = 0$

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- dominant contribution to ionisation happens (or starts) at imaginary time! (and there are more tunnelling times which come out imaginary from the calculations . . .)

The Keldysh exponent

$$P(\vec{v}=0,t) = |\Phi(\vec{v}=0,t)|^2 \propto \left| -i \int_0^t e^{-iS_{\vec{v}}(t,t')}\,\mathrm{d}t' \right|^2 \overset{\text{SPA}}{\approx} \left| e^{-iS_{\vec{v}}(t,t'_*)} \right|^2$$

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- substituting $t'_* = i\frac{\sqrt{2I_\mathrm{p}}}{F_0}$ into $S_{\vec{v}}(t,t')$, $\omega t = n\pi$, set $n=0$:

$$\begin{aligned} S_{\vec{v}}(0,t'_*) &= \frac{1}{2}\int_{t'_*}^{0} \left[\frac{F_0}{\omega}\sin(\omega t'')\right]^2 \mathrm{d}t'' - I_\mathrm{p} t'_* \\ &\approx \frac{1}{2}\int_{t'_*}^{0} \left(\frac{F_0}{\omega}\omega t''\right)^2 \mathrm{d}t'' - I_\mathrm{p} t'_* = \frac{-1}{2} F_0^2 \frac{(t'_*)^3}{3} - I_\mathrm{p} t'_* \\ &\approx \frac{i}{2}\frac{(2I_\mathrm{p})^{3/2}}{F_0}\left(\frac{1}{3}-1\right) = \frac{-i}{2}\frac{2}{3}\frac{(2I_\mathrm{p})^{3/2}}{F_0} \end{aligned} \qquad (15)$$

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- Probability of ionisation at the peak of the field, with zero velocity, to exponential accuracy:

$$\boxed{P(\vec{v}=0,t=0) \propto \exp\left(-\frac{2(2I_\mathrm{p})^{3/2}}{3F_0)}\right)} \tag{16}$$

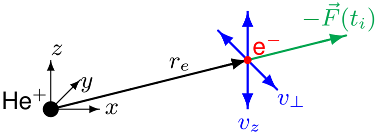
The generalised Keldysh exponent I

- quasi-static idea: ionisations at other times $t \neq 0$ means we have lower field strength $F(t) \leq F_0$
- account for Stark shift in the ionisation potential

$$I_{\mathrm{p}}(t) = I_{\mathrm{p}}(F(t)) = I_{\mathrm{p},0} + \frac{1}{2}\left(\alpha_N - \alpha_I\right) F(t)^2, \qquad (17)$$

- allow transverse velocity at exit tunnel (adding to the ionisation potential), laser propagation in z direction, elliptically polarised in $x - y$ plane

$$P(v_{||} = 0, v_\perp, v_z, t) \propto \exp\left\{\frac{-2\left(2I_{\mathrm{p}}(t) + v_\perp^2 + v_z^2\right)^{3/2}}{3F(t)}\right\} \qquad (18)$$



Final result

first order taylor:

$$\left(2I_\mathrm{p}(t) + v_\perp^2 + v_z^2\right)^{3/2} \approx \left(2I_\mathrm{p}(t)\right)^{3/2} + \frac{3}{2}\left(2I_\mathrm{p}(t)\right)^{1/2}\left(v_\perp^2 + v_z^2\right)$$

everything together:

$$P(v_{||} = 0, v_\perp, v_z, t) \propto \exp\left(\frac{-2\left(2I_\mathrm{p}(t)\right)^{3/2}}{3F(t)}\right)\exp\left(-\frac{v_\perp^2 + v_z^2}{2\sigma_\perp^2}\right) \quad (19)$$

with

$$\sigma_\perp^2 = \frac{F(t)}{2\left(2I_\mathrm{p}(t)\right)^{1/2}} = \frac{\omega}{2\gamma} \quad (20)$$