# Keldysh Theory

Cornelia Hofmann

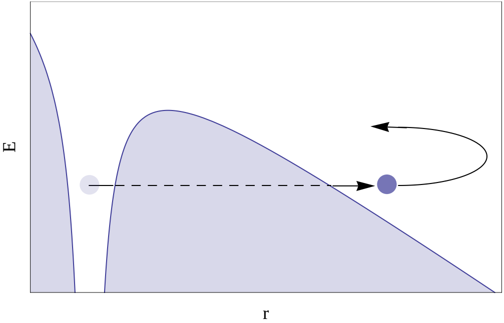
Department of Physics, Institute for Quantum Electronics,
ETH Zurich, Switzerland

# 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



- strong field → tunnelling
- ultrashort pulses
  → FWHM ≈ fs regime
- slow field compared to electron dynamics

# 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

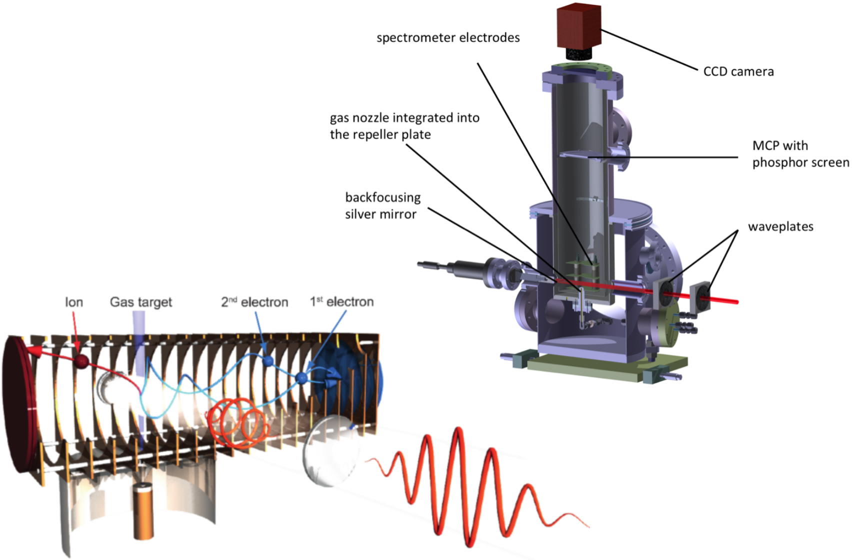
Atomic units to make calculations/formulas easier:

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

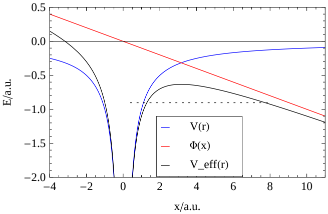
# Outline

1. Initial Situation
2. SFA, dipole and quasi-static approximation
3. Linear polarisation
4. Evaluate the probability amplitude
5. Keldysh exponent
6. End Result

# Experiments



# Laser-Atom system



- what we have to calculate:

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

field-free Hamiltonian of the atom

interaction with the laser field

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

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

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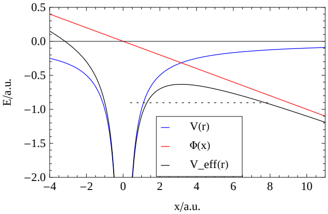

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$$\Psi = \Psi(t, \vec{r}_1, \ldots \vec{r}_n)$$

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

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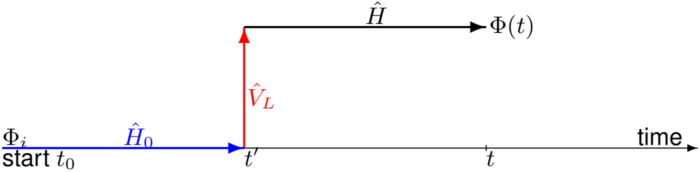
- dipole approximation: $\lambda >> r(\mathrm{atom}) \Rightarrow \vec{F}(t)$
- SFA 1: neglect laser field while in bound state
- adiabatic case (quasi-static approximation) $\omega << \omega_B$
- **exact** solution:

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



# Solution Check

Subsitute (3) into Schrödinger's Equation:

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

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quasi-static:

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

$$= -i\hat{H}\int_{t_0}^{t}\mathrm{d}t'\left[e^{-i(t-t')\hat{H}}\right]\hat{V}_L(t')\left[e^{-i(t'-t_0)\hat{H}_0}\right]|\Phi_i\rangle + e^{-i(t-t)\hat{H}}\hat{V}_L(t)e^{-i(t-t_0)\hat{H}_0}|\Phi_i\rangle \qquad + \hat{H}_0e^{-i(t-t_0)\hat{H}_0}|\Phi_i\rangle$$

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

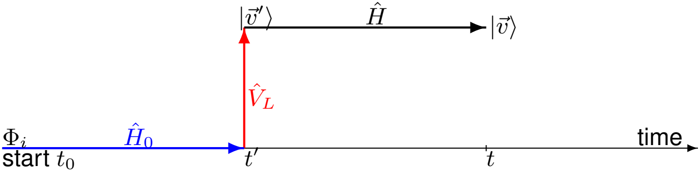
# Basis in the Continuum

- 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: $\langle\vec{v}|\Phi_i\rangle = 0$

# 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: $\langle\vec{v}|\Phi_i\rangle = 0$
- 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 $\hat{H}_F = \frac{1}{2}\left(\vec{p} - q\vec{A}\right)^2$

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

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

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

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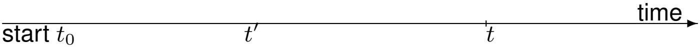
- the canonical momentum $\vec{p} = \vec{v} + q\vec{A}$ is conserved

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

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

# Intuitive picture so far

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



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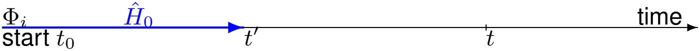
$\Phi_i$ $\hat{H}_0$

start $t_0$

$t'$

$t$

time

- waiting in groundstate

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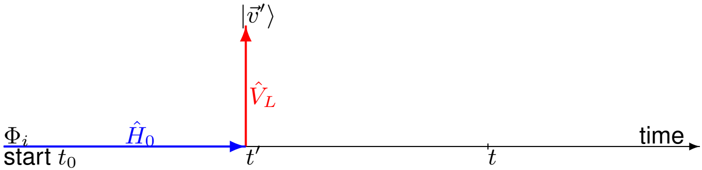
$$\Phi(\vec{v},t) = -i\int_{t_0}^{t} \mathrm{d}t' e^{-i\int_{t'}^{t}\frac{1}{2}\left(\vec{v}-\vec{A}(t)+\vec{A}(t'')\right)^2 \mathrm{d}t''} e^{i(t'-t_0)I_\mathrm{p}} \times \underbrace{\langle \vec{v}-\vec{A}(t)+\vec{A}(t')\left|\hat{V}_L(t')\right|\Phi_i\rangle}_{\text{prefactor}} \tag{7}$$



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

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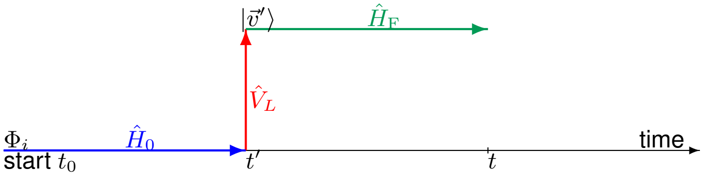

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- kick from laser field, jump up to continuum state, exit tunnel with velocity $\vec{v}'$
- oscillating in the laser field

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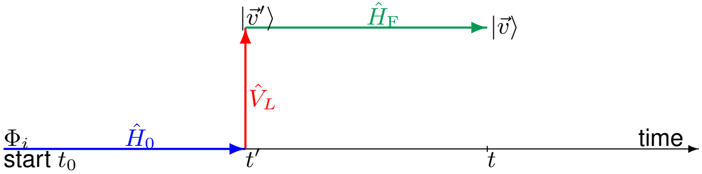
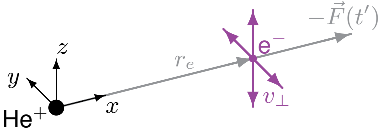


- waiting in groundstate
- kick from laser field, jump up to continuum state, exit tunnel with velocity $\vec{v}'$
- oscillating in the laser field
- recorded on detector with velocity $\vec{v}$

# Linear polarisation

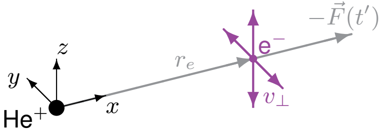


- laser field polarised along $\hat{x}$
  → vector potential:

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

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

# Linear polarisation



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- define action

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

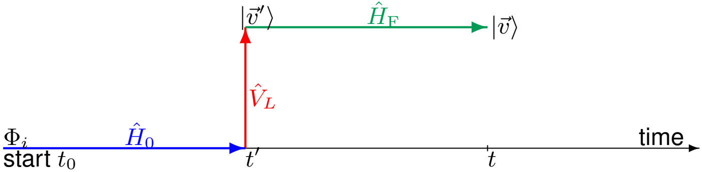
$$\Phi(\vec{v},t) \propto -i\int_{t_0}^{t}\mathrm{d}t' e^{-i\int_{t'}^{t}\frac{1}{2}\left(\vec{v}-\vec{A}(t)+\vec{A}(t'')\right)^2\mathrm{d}t''}\, e^{i(t'-t_0)I_\mathrm{p}}$$

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

# Recap

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

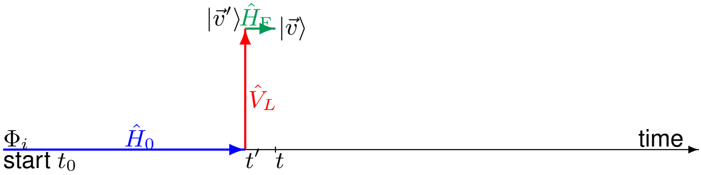
$$\Phi(\vec{v},t) \propto -i\int_{t_0}^{t} \exp\left(-iS_{\vec{v}}(t,t')\right)\,\mathrm{d}t'$$



# Outlook

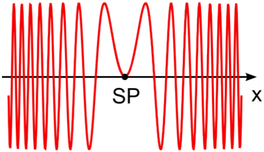
- Now comes the fun part.
- Find the **probability of ionisation** depending on
  - ionisation time $t'$
  - assumption: instantaneous tunneling time
  - and tunnel exit velocity $\vec{v}'$.

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



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

# 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)

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

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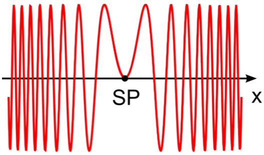

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$$-I_\mathrm{p} t'$$

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

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

⇒ 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: $\vec{v} = 0$

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

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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 $n=0$:

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

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

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

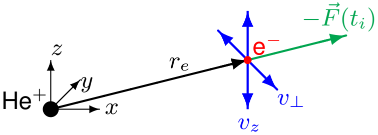
# The generalised Keldysh exponent I

- 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

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

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

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



# 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:

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

with

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