Strong field ionization

$$H = H_0 + V(t),$$

the evolution operator can be written as:

$$\boxed{\mathcal{U}(t_f, t_i) = \mathcal{U}_0(t_f, t_i) - \frac{i}{\hbar} \int_{t_i}^{t_f} \mathcal{U}(t_f, \tau) V(\tau) \mathcal{U}_0(\tau, t_i) d\tau}$$

We are interested to describe transition of a system from an initial state $|\Psi(t_i)\rangle$ (ground state of the hydrogen atom) to the continuum state $|\Psi(t_f)\rangle$. (~~state~~ the electron moving ~~to the detector~~ around the atom.)

$$|\Psi(t_f)\rangle = \mathcal{U}(t_f, t_i) |\Psi(t_i)\rangle$$ – solution of the TDSE.

The time dependent WF $|\Psi(t)\rangle$ is the superposition of all possible states (solutions) of the SE with the Hamiltonian H:

$$\cancel{|\Psi(t)\rangle = \sum\!\!\!\!\int c} \quad |\Psi(t)\rangle = \underbrace{\sum_i c_i(t)\varphi_i}_{\text{bound states}} + \underbrace{\int d\vec{k}\, c(\vec{k})\phi(\vec{k})}_{\text{continuum states}} \qquad (1)$$

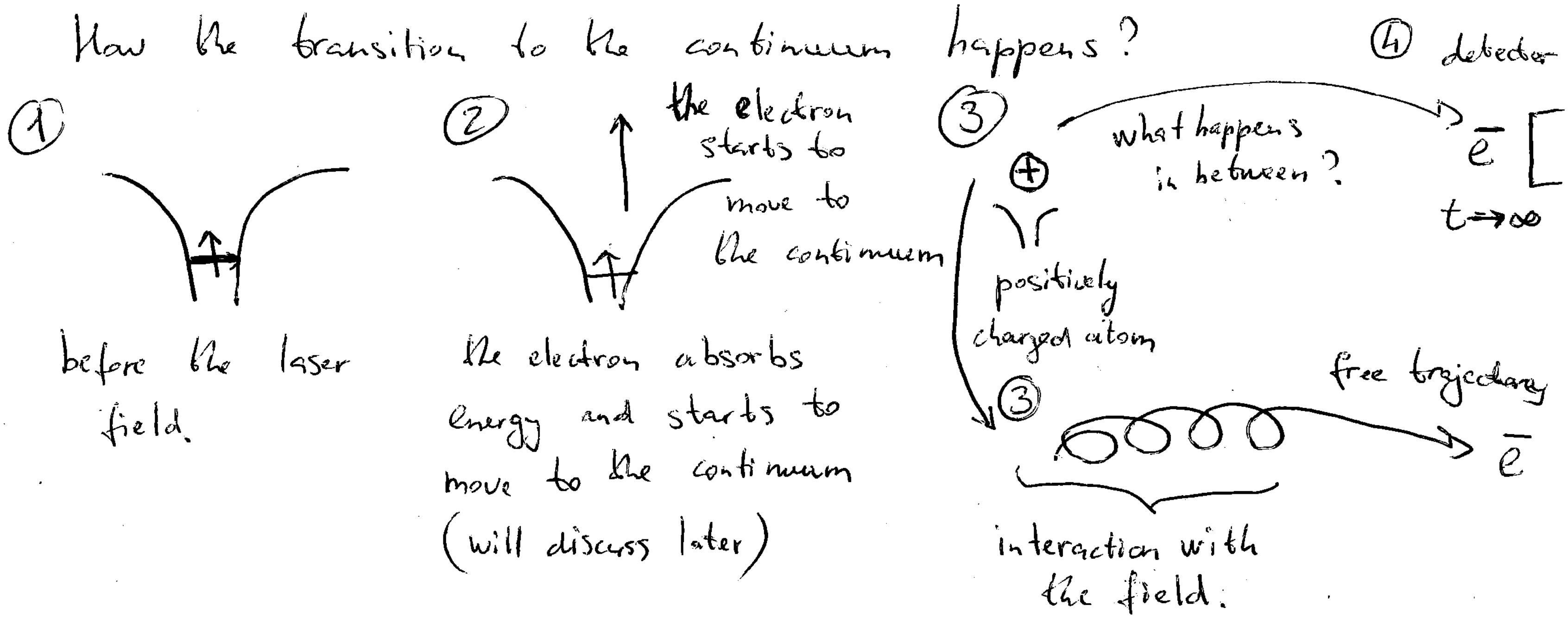
Our goal is to understand how the system evolves from $|\Psi(t=t_i)\rangle = \varphi_0$ – ground state of the H atom, to something which looks like (1). This transition is driven by laser field.

Eventually, we want to understand what are the expansion coefficients

$$c_i \left(\text{or } c(\vec{k})\right) = \langle \varphi_i \left(\text{or } \phi(\vec{k})\right) | \Psi(t)\rangle,$$ they can be used to compute the spectra.

In the case of ionization, the states of interest are the continuum states $\phi(\vec{k})$.

How the transition to the continuum happens?



We know the solution of the SE for situations ① and ④, bound states and plane-wave states, respectively.

What happens in steps ② and ③?

Let us consider step ③: electron is in the continuum but <u>still experiences the action of the field</u>

Classical picture:

$$\frac{d^2\vec{x}(t)}{dt^2} = -\vec{E}_o \sin(\omega t)$$

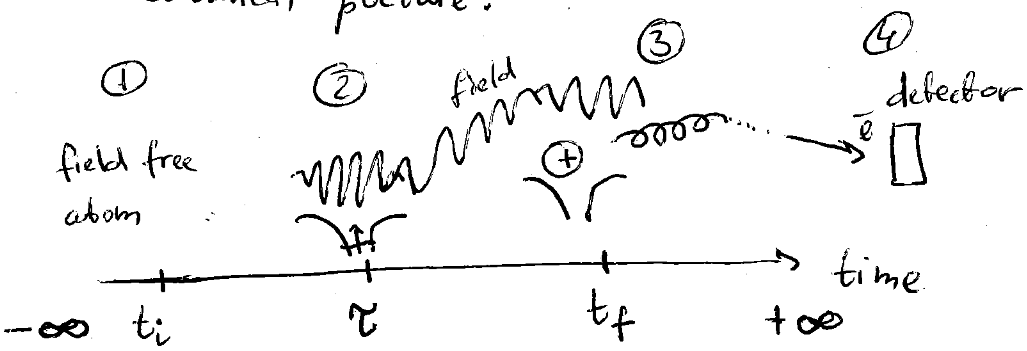
velocity: $\vec{x}'(t) = \vec{V}_{drift} + \frac{\vec{E}_o}{\omega}\cos(\omega t)$

position: $\vec{x}(t) = \vec{x}_o + \vec{V}_{drift} \cdot t + \frac{\vec{E}_o}{\omega^2}\sin(\omega t)$

KE: $T = \frac{(\vec{x}'(t))^2}{2} = \frac{V_{drift}^2}{2} + \vec{V}_{drift} \cdot \frac{\vec{E}_o}{\omega}\cos(\omega t) + \frac{E_o^2}{2\omega^2}\cos^2(\omega t)$

Average KE: $\langle T \rangle = \frac{V_{drift}^2}{2} + \boxed{\frac{E_o}{4\omega^2}}$ – ponderomotive potential

Quantum mechanical picture:



interaction of an atom with the field

To get analytical solution for 3 we neglect the interaction of the electron and the remaining ion:

$$\cancel{H = -\frac{1}{2}\nabla^2 + V(t)}$$

dipole approximation:

$$\hat{H} = \frac{1}{2}\left(-i\hbar\nabla - \frac{e}{c}\vec{A}\right)^2 \cancel{H = \frac{1}{2}\left(p - \frac{e}{c}A\right)^2}, \quad \text{where} \quad \vec{A} = \vec{A}_0 \cos \omega t$$

$$\vec{E} = -\frac{1}{c}\frac{\partial \vec{A}(t)}{\partial t} = \frac{\omega \vec{A}_0}{c}\sin(\omega t) = \vec{E}_0 \sin\omega t$$

Then solving $i\hbar\frac{\partial}{\partial t}|\phi_K^V(t)\rangle = \hat{H}(t)\,|\phi_K^V(t)\rangle$, we find the Volkov states:

$$|\phi_K^V(t)\rangle = \exp\left[i(\vec{K} + \vec{A}(t))\cdot\vec{x} - i S(t)\right],$$

and the classical action

$$S(t) = \frac{1}{2}\int\left[\vec{K} + \vec{A}(t')\right]^2 dt', \quad \text{using}$$

we can write the action explicitely:

$$\boxed{S(t) = \left(\frac{K^2}{2} + U_p\right)t + \left(\frac{\vec{K}\cdot\vec{E}_0}{\omega^2}\cos(\omega t) - \frac{U_p}{2\omega}\sin(2\omega t)\right)}$$

We go back to the problem of propagation

$$|\psi(t_i)\rangle \xrightarrow[\text{evolution}]{} |\psi(t_f)\rangle,$$

let us find the probability to find a system in a final state with the momentum $\vec{K}$ (plane wave with momentum $\vec{k}$):

$$\langle\phi(\vec{k})|\psi(t_f)\rangle = \langle\phi(\vec{k})|\hat{\mathcal{U}}(t_f,t_i)|\psi(t_i)\rangle =$$

$$= \underbrace{\langle\phi(\vec{k})|\hat{\mathcal{U}}_0|\psi(t_i)\rangle}_{\substack{= 0 \text{ if} \\ \text{both } \phi(\vec{k}) \text{ and } \psi(t_i) \\ \text{are eigenstates of } H_0}} - \frac{i}{\hbar}\int_{t_i}^{t_f}\langle\phi(\vec{k})|\hat{\mathcal{U}}(t_f,\tau)\,V(\tau)\,\underbrace{\mathcal{U}_0(\tau,t_i)|\psi(t_i)\rangle}_{e^{-\frac{i}{\hbar}E_0 t}|\psi(t_i)\rangle}\,d\tau$$

assuming that $\bar{e}$ doesn't interact with the ion, we get $\langle\phi_K^V(t)|$ Volkov states,

$E_0 = -I_p$

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so eventually we get:

$$\boxed{\langle\phi(\vec{k})|\psi(t_f)\rangle = -\frac{i}{\hbar}\int_{t_i}^{t_f}\langle e^{i(\vec{k}+\vec{A}(t))\vec{x}}|\vec{x}\cdot\vec{E}(t)|\psi(t_i)\rangle e^{i(S(t)+I_p t)}}$$

amplitudes of the probability to find an electron with momentum $\vec{k}$.

Assuming $t_i \to -\infty$, $t_f \to +\infty$

The integral can be evaluated analytically as:

$$\langle\phi(\vec{k})|\psi(t_f)\rangle = -2\pi i\sum_{n\geq n_0}^{\infty} L_n(\vec{k})\,\delta\left(\frac{k^2}{2} + U_p + I_p - n\omega\right),$$

with the coefficients (Fourier series)

$$L_n(\vec{k}) = \int_{-\infty}^{+\infty} dt\, e^{in\omega t}\left\{\exp\left(i\left(\frac{\vec{k}\cdot\vec{E}_0}{\omega^2}\cos\omega t - \frac{U_p}{2\omega}\sin(2\omega t)\right)\right)\langle e^{i(\vec{k}+\vec{A})}|x\cdot E|\psi\rangle\right.$$

and $n_0\omega = U_p + I_p$ – n_0 is the minimum number of photons required to overcome the barrier $U_p + I_p$.

Approximations for the integral $\langle \phi(\vec{k}) | \Psi(t_f) \rangle$:

$$\langle \phi(\vec{k}) | \Psi(t_f) \rangle \simeq -\frac{i}{\hbar} \int_{t_i}^{t_f} f(t)\, e^{i\tilde{S}(t)}\, dt$$

$$f(t) = \langle \exp(i(\vec{k}+\vec{A})\cdot\vec{x}) | \vec{x}\cdot\vec{E}(t) | \Psi(t_i) \rangle$$ – slowly varying function of time

and

$$\tilde{S}(t) = S(t) + I_p t =$$

$$= \left(\frac{k^2}{2} + U_p + I_p\right) t + \left(\frac{\vec{k}\cdot\vec{E}_0}{\omega^2} \cos(\omega t) - \frac{U_p}{2\omega} \sin(2\omega t)\right)$$

~~fast oscillating function~~

$e^{i\tilde{S}(t)}$ – fast oscillating function of time.

A way to simpify the integral $\int_{t_i}^{t_f} f(t)\, e^{i\tilde{S}(t)} dt$ is to compute saddle points of $\tilde{S}(t)$.

• Keldysh derived two limiting cases:

(he used the probability of the transition $P_{i\to f}(\vec{k}) = |\langle \phi(\vec{k}) | \Psi(t_f) \rangle|^2$,

and the total ionization rate $\boxed{P = \int \frac{d^3\vec{k}}{(2\pi)^3} P_{i\to f}(\vec{k})}$

$$\omega_k = \frac{\sqrt{3E_0\pi}}{4} \exp\left[-\frac{2}{3E_0}\left(1 - \frac{\gamma^2}{10}\right)\right]$$

$\gamma = \frac{\omega}{\omega_t}$ – is the Keldysh parameter, where

$$\omega_t = \sqrt{\frac{I_p}{2U_p}}$$

Tunneling limit

$$\Gamma = \frac{\sqrt{6\pi}}{2} \sqrt{\frac{I_0 \omega}{\gamma}} \exp\left(-\frac{4}{3} \frac{I_0 \gamma}{\omega}\left[1 - \frac{\gamma^2}{10}\right]\right)$$

Multi-photon limit

$$\Gamma = 2\sqrt{2 I_0 \omega} \left(\frac{1}{4\gamma^2}\right)^{\langle I_0/(\omega+1)\rangle} \exp\left(2\left\langle \frac{I_0}{\omega} + 1\right\rangle - \frac{I_0}{\omega}\right) \times$$

$$\times \Phi\left\{\left[2\left\langle \frac{I_0}{\omega} + 1\right\rangle - 2\frac{I_0}{\omega}\right]^{1/2}\right\},$$

where $\langle\,\rangle$ denotes the integer part of a real number

$\Phi(z) = \int_0^z e^{y^2 - z^2}\, dy$ is the Dawson function.

Pondermotive energy

Classical equations of motion for $\vec{e}$ in monochromatic laser field

$$\vec{x}''(t) = -\vec{E}_0 \sin(\omega t)$$

velocity $$\vec{x}'(t) = \vec{V}_{drift} + \frac{\vec{E}_0}{\omega}\cos(\omega t)$$

position $$\vec{x}(t) = \vec{x}_0 + \vec{V}_{drift}t + \frac{\vec{E}_0}{\omega^2}\sin(\omega t)$$

Kinetic energy:

$$T = \frac{(\vec{x}')^2}{2} = \frac{V_{drift}^2}{2} + \vec{V}_{drift}\cdot\frac{\vec{E}_0}{\omega}\cos\omega t + \frac{E_0^2}{2\omega^2}\cos^2(\omega t)$$

Average KE:

$$\langle T\rangle = \frac{V_{drift}^2}{2} + \boxed{\frac{E_0^2}{4\omega^2}}$$

$U_p = \frac{E_0^2}{4\omega^2}$ is pondermotive potential.