Bachelor's Thesis

The role of excited atomic states in multiphoton ionization

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Bachelorarbeit

Die Rolle angeregter atomarer Zustände bei der Mehrphotonenionisation

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Abstract

Multiphoton ionization of atoms in strong laser fields is a fundamental process in attosecond physics. In this work, we extend the strong-field approximation (SFA) by incorporating the influence of excited atomic states on ionization rates. Standard SFA formulations neglect these excited states, assuming that the laser field has no effect on the atom before ionization. However, in intense few-cycle laser pulses, the Stark shift and transient population of excited states can significantly modify ionization dynamics. We numerically solve the time-dependent Schrödinger equation (TDSE) using the tRecX code to extract time-dependent probability amplitudes for hydrogen's ground and excited states. These amplitudes are then integrated into the SFA formalism to evaluate their impact on ionization rates.

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1. Introduction

What motivates this thesis? Background: developement of SFA and GASFIR rates that doesnt have to numerically solve Schroedinger equation. Comparison of ion rates from tRecX, SFA and GASFIR. When the laser pulse is an even function in time, the SFA rate is that as well. But numerical simulations from tRecX tell us thats not the case and the time symmetry is broken. Idea: because of the neglected excitedt states in SFA. This brings up more questions: What role play excited states in ionization? Does the stark effect play an important role? Why is this so complicated? First, $[\hat{\mathcal{H}}(t), \hat{\mathcal{H}}(t')] \neq 0$ because $\hat{\mathcal{H}}_0$ and $\hat{V}$ dont share same eigenbasis, -> the electron is free. Also the thing with all these gauges.

2. Theory

Convention:

Ψ wavefunction for the whole system

$\psi(\underline{x})$ for a wavefunction in position space without choosing explicit coordinates,

$\phi(\underline{p})$ for a wavefunction in momentum space,

$\mathbf{A}$ for abstract vector as element in vector space,

$\underline{x}$ for vector in $\mathbb{R}^n$

$|\Psi\rangle$ an abstract element in Hilbert space $\mathscr{H}$,

$|\Phi\rangle$ for the abstract Eigenstates of the whole Hamiltonian,

$Y_{l,m}(\theta, \phi) = \langle \theta, \phi | l, m \rangle$ definition of spherical harmonics,

$\psi_{n,l,m}(r, \theta, \phi)$ for the wavefunction of hydrogen in spherical coordinates, with $\underline{x} = (r, \theta, \phi)$

I use underlined vectors when they are the coordinates and bold vectors when they are abstract elements in a vector space.

The canonical momentum $\underline{P}$ parametrises the phase space but the kinetic momentum does not so kinetic momentum $\hat{=}\mathbf{p}$.

The position and momentum operator are in boldface $\hat{\mathbf{x}}$ because they do not choose any kind of basis not even a representation in which they are displayed.

When i use $|\mathbf{k}\rangle$ I mean a plane wave solution so "special" a continuum state.

A $\cdot$ denotes a scalar product between two vectors, $\times$ is just normal multiplication.

The strucutre in this chapter mainly follows [**?**] with some modifications.

2.1 Basic Formalism

Our goal is to come up with a expression were we can

2.1.1 Schrödinger Equation

Basic Definitions of schröfinger qe, light dyson series, and strong field s matrix

We want the time evolution of a quantum system in the presence of an external time dependent field in order to describe the strong field ionization later on. The time evolution of a quantum system is given by the time dependent Schrödinger equation and a general hamiltonian

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = \hat{\mathcal{H}}(t)|\Psi(t)\rangle. \tag{2.1}$$

The formal solution depends on the time dependence of the hamiltonian and the physical

setting. In the following we assume ^{1} (IMPORTANT) that $[\hat{\mathcal{H}}(t), \hat{\mathcal{H}}(t')] = 0$ so we assume some sort of quasi static approximation to the Hamiltons time evolution. The solution is then given by

$$|\Psi(t)\rangle = e^{-i\int_{t_0}^{t} \hat{\mathcal{H}}(t')dt'} |\Psi(t=0)\rangle = \hat{\mathcal{U}}(t) |\Psi(0)\rangle \tag{2.2}$$

Now its time so establish a physical setting. We have Hydrogen Atom with nucleus and electron described by time indepentent Hamilton $\hat{\mathcal{H}}_0$. The external laser Field is described by an time dependent part $\hat{V}_I(t)$. To describe the interaction of the atom with the laser field we use in the following the dipole approximation.

There are two ways of representing $|\Psi(t)\rangle$, schroedinger and interaction picture. Coefficients.

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^{1}How? Later. No physical setting bzw no approximations yet. Its better to juistify it later but have a working formalism instead of the other way around.

2.1.2 Light-Matter Interaction

A light wave is defined by the Maxwell equations

$$\nabla \cdot \mathbf{E} = \rho \qquad \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$

$$\nabla \cdot \mathbf{B} = 0 \qquad \nabla \times \mathbf{B} = \mathbf{J} + \frac{\partial \mathbf{E}}{\partial t}$$

The Maxwell equations are being solved by

$$\mathbf{E} = -\nabla \varphi - \frac{\partial \mathbf{A}}{\partial t}$$

$$\mathbf{B} = \nabla \times \mathbf{A}$$

(2.3)

For these solutions we introduced the vector potential $\mathbf{A}(\underline{x}, t)$ and the scalar potential $\varphi(\underline{x}, t)$. These are not unique such that different choices can result in the same physical setting. In general

$$\mathbf{A} \to \mathbf{A} + \nabla \chi$$

$$\varphi \to \varphi - \frac{\partial \chi}{\partial t}$$

also fulfill the Maxwell equations while $\chi(t)$ is an arbitrary smooth scalar function. The arbitrariness of χ is known as gauge freedom and a direct consequence of the Maxwell equations. Choosing a gauge (i.e., a specific χ) is a matter of convenience and can be used to simplify the calculations as presented in the following.

2.1.3 Dipole Approximation

Very important approximation. The dipole approximation is valid when the wavelength of the optical field is much larger than both the size of the relevant bound electron states and the maximum displacement of a free electron during the light-matter interaction. Additionally, it assumes that the magnetic field of the light has a negligible effect on the electron's motion, meaning the velocities of the charged particles must be nonrelativistic.

To see where exaclty one makes this assumption, first we rewrite the Maxwell equations in the dependence of the vector potential and the scalar potential as defined in (**??**). This will result in two coupled differential equations, what does not bring us any further. However we are interestet in making a simple expression for the vector potential $\mathbf{A}$. We achieve this by choosing a certain gauge, the so called Lorentz gauge

$$\partial_\mu \mathbf{A}^\mu = 0 \quad \text{or} \quad \nabla \cdot \mathbf{A} + \frac{\partial \varphi}{\partial t} = 0$$

This can be achieved by solving the inhomogenous wave equation for χ that comes up when doing this calculation explicitly and is possible when $\mathbf{A}$ and φ are know. Now the Maxwell equations are uncoupled and can be written as

$$\nabla^2 \varphi - \frac{\partial^2 \varphi}{\partial t^2} = \rho$$

$$\nabla^2 \mathbf{A} - \frac{\partial^2 \mathbf{A}}{\partial t^2} = \mathbf{J}$$

We are mainly interested in the second equation. The equation is known as the wave equation therefore $\mathbf{A}$ describes plane waves

$$\mathbf{A}(\underline{x}, t) = \mathbf{A}_0 \mathrm{e}^{\pm i(\underline{k}\cdot\underline{x}-\omega t)}$$

The dipole approximation is mathemaically speaking just the leading term in Taylor expansion of $\mathrm{e}^{i\underline{k}\cdot\underline{x}}$. The vector potential is therefore independent of the spatial coordinates and can be written as

$$\mathbf{A}(\underline{x}, t) = \mathbf{A}_0 \mathrm{e}^{\mp i\omega t} \exp\left\{\pm 2\pi i \frac{|\underline{x}|}{\lambda} \underline{e}_k \cdot \underline{e}_x\right\} \approx \mathbf{A}_0 \mathrm{e}^{\mp i\omega t}\left(1 + \mathcal{O}\left(\frac{|\underline{x}|}{\lambda}\right)\right) = \mathbf{A}(t)$$

As long as the Wavelength is big enough this approximation is valid. It follows:

$$\mathbf{B} = \nabla \times \mathbf{A} \approx 0$$

Even tough we will later choose another gauge, the physics in our system remains the same. The dipole Approximation is not gauge dependent, so in another gauge $\mathbf{B}$ remains approximately zero. Choosing the lorentz gauge here is just a matter of convenience, because just expanding the vector potential to the linear term is very intuitive.

This was the essence of the dipole approximation but we also want a intuitive expression for our Laser Fild in the Hamiltonian. It will be helpfull to look at the semi classical Hamilton function of a free electron in an electric field[2]:

$$\hat{\mathcal{H}}(\underline{x}, t) = \frac{1}{2m}(\hat{\mathbf{P}} - e\mathbf{A}(\underline{x}, t))^2 + e\varphi(\underline{x}, t) \tag{2.4}$$

In the dipole Approximation, this can be simplified to:

$$\hat{\mathcal{H}}(\underline{x}, t) = \frac{\hat{\mathbf{P}}^2}{2m} + \frac{e}{m}\hat{\mathbf{P}} \cdot \mathbf{A}(t) + \frac{e^2}{2m}\mathbf{A}^2(t) - e\varphi(\underline{x}, t)$$

Note that we could set φ to zero because the source of the em wave are outside of our region of interest but the dipole approximation can be made without this assumption. We will however set φ to zero later. Another general assumption one made when working with semi classical Hamiltonians is that only the vectorpotential causes the electron to change its state but not vice versa (Bosßmann). This is reasonable approximation because in our case the intensity of the Laser is sufficiently high, so we dont have to worry about that(is it really??). Now we perform another gauge transformation to the so called length gauge via

$$\chi = -\mathbf{A}(t) \cdot \underline{x}$$

This gauge sets $\mathbf{A}$ to zero, and φ will have the following form:

$$\nabla\varphi \rightarrow \nabla \cdot (\varphi + \mathbf{x} \cdot \frac{\partial \mathbf{A}}{\partial t}) = \nabla\varphi + \frac{\partial \mathbf{A}}{\partial t} = -\mathbf{E}$$

Integrating this equation from the origin to $\mathbf{x}$ gives us the electric potential in the length gauge. Furthermore, $\mathbf{r}$ is now quantized and our Hamilton therefore reads:

$$\hat{\mathcal{H}}(\underline{x}, t) = \frac{\hat{\mathbf{P}}^2}{2m} - e\hat{\mathbf{x}} \cdot \mathbf{E}$$

We can rewirte the time dependent part $\hat{V}$ of our quantum mechanical Hamiltonian as

$$\hat{V}_I(t) = -\hat{\mathbf{d}} \cdot \mathbf{E}(t) \tag{2.5}$$

where $\hat{\mathbf{d}} = e\hat{\mathbf{x}}$ is the dipole operator and $\mathbf{E}(t)$ is the electric field.

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[2]Because its interesting it will be derived in **??**

2.2 Strong Field Approximation

For making the strong field approximation we first have to obtain a point where is its good to use. When we treat $\hat{V}_I(t)$ as the interaction term, we can write an exakt solution to (**??**):

$$|\Psi(t)\rangle = -i \int_{t_0}^{t} \mathrm{d}t' e^{-i\int_{t'}^{t} \hat{\mathcal{H}}(t'')\mathrm{d}t''} \hat{V}_I(t') e^{-i\int_{t_0}^{t'} \hat{\mathcal{H}}_0(t'')\mathrm{d}t''} \sum_n c_n(t') |\Psi_n\rangle + e^{-i\int_{t_0}^{t} \hat{\mathcal{H}}_0(t')\mathrm{d}t'} \sum_n c_n(t') |\Psi_n\rangle$$

(2.6)

as can be checked by inserting the solution into the Schrödinger equation.

The key difference in this formula compared to other literature is the fact that instead of choosing the ground state as the initial state, we use a timdependent superposition of all states $\sum_n c_n(t') |\Psi_n\rangle$. Why this solution even works and for more details, see **??**.

To make this expression more appealing we can project it into an eigenstate in the continuum ie a electron state characterized by its velocity. As can be seen in the following, the last term in (**??**) is now gone because there can be no overlap between a continuum state and the initial state.

$$\langle \Pi(t_c)|\Psi(t)\rangle = -i \int_{t_0}^{t} \mathrm{d}t' \langle \Pi(t=t_c)| e^{-i\int_{t'}^{t} \hat{\mathcal{H}}(t'')\mathrm{d}t''} \hat{V}_I(t') e^{-i\int_{t_0}^{t'} \hat{\mathcal{H}}_0 \mathrm{d}t''} \sum_n c_n(t') |\Psi_n\rangle \qquad (2.7)$$

With $|\Pi(t_c)\rangle$ being a continuum state with momentum $\underline{p}$ at time t_c with t_c being sufficiently big enough for the electron to be in the continuum ($t_c >> t'$).

Equation (**??**) is known as the strong field S-matrix amplitude. It is best to read this equation from right to left, starting with the initial state of our system and the field free time propagation of the system. At moment t' the Laser starts to interact with the system and it transisions into a virtual state instantaniously. From time t' to the observed time t the system is described by the full Hamiltonian including both Laser Field and the binding potential.

Now we have a good place to start with the strong field approximation. In principle, SFA is the neglecting of the Coulomb potential once the electron is in the continuum because the Laser Field is now the dominant force acting on the electron. We can therefore write the term with the full Hamiltonian as

$$e^{-i\int_{t'}^{t} \hat{\mathcal{H}}(t'')\mathrm{d}t''} \approx e^{-i\int_{t'}^{t} \hat{\mathcal{H}}_{SFA}(t'')\mathrm{d}t''} \quad \text{with} \quad \hat{\mathcal{H}}_{SFA}(t) = \hat{\mathcal{H}}(t) - \hat{V}_C$$

This is very usefull because for $\hat{\mathcal{H}}_{SFA}$ we know an exact analytical solution. For that lets take another look at the semi classical Hamilton (**??**). Classically, the physics driven by the momentum operator $\hat{\mathbf{P}}$ is known as the canonical momentum and given by:

$$\frac{\partial \mathcal{L}}{\partial \underline{\dot{x}}} = \underline{P} = m\underline{\dot{x}} + \frac{e}{c}\mathbf{A} \stackrel{\text{a.u.}}{=} \mathbf{p} + \mathbf{A}$$

In our case the canonical momentum is conserved. To see this, lets finally set $\varphi = 0$ so we have $\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}$ as justified above and recall the equation of motion for a charged particle in an electromagnetic field [**?**]:

$$\frac{\mathrm{d}\mathbf{p}}{\mathrm{d}t} = \mathbf{E} + [\underline{\dot{x}} \times \mathbf{B}] \approx -\frac{\partial \mathbf{A}}{\partial t} = -\frac{\mathrm{d}\mathbf{A}}{\mathrm{d}t}$$

And also the energy of the system is clear:

$$E(t) = \underline{\dot{x}}\frac{\partial \mathcal{L}}{\partial \underline{\dot{x}}} - \mathcal{L} = \frac{\mathbf{p}^2}{2}$$

Note that the energy is not conserved because the argument we made bevor does not hold for the kinetic momentum only for the canonical momentum. Now comes the interesting part. Clearly $|\Pi(t_c)\rangle$ is a solution of $e^{-i\int_{t'}^{t}\hat{\mathcal{H}}_{SFA}(t'')\mathrm{d}t''}$ so that gives us:

$$e^{-i\int_{t'}^{t}\hat{\mathcal{H}}_{SFA}(t'')\mathrm{d}t''}|\Pi(t_c)\rangle = e^{-i\int_{t'}^{t}(\underline{P}-\mathbf{A}(t''))^2\mathrm{d}t''}|\Pi(t_c)\rangle$$

$\underline{P}$ is of course independent of time, but $\mathbf{A}$ is not. Since dealing with canonical momentum in numerical simulations is not very convenient, we use the fact that its conserved and calculate the momentum at other times. In particular we are interested in times where the laser field is long gone:

$$\underline{P} = \mathbf{p}(t'') + \mathbf{A}(t'') = \mathbf{p}(t \to \infty) + \mathbf{A}(t \to \infty) = \mathbf{p}$$

Furthermore (how??)

$$|\Pi\rangle = |p + A\rangle$$

$$\langle\Pi(t_c)|\Psi(t)\rangle = -i\int_{t_0}^{t}\mathrm{d}t' e^{-\frac{i}{2}\int_{t'}^{\infty}(\mathbf{p}+\mathbf{A}(t''))^2\mathrm{d}t''}e^{-i\hat{\mathcal{H}}_0(t')}\sum_n c_n(t')\langle\mathbf{p}+\mathbf{A}(t')|\hat{\mathbf{d}}\cdot\mathbf{E}(t')|\Psi_n\rangle \quad (2.8)$$

$$= -i\int_{0}^{t}\mathrm{d}t' e^{-\frac{i}{2}\int_{t'}^{\infty}(\mathbf{p}+\mathbf{A}(t''))^2\mathrm{d}t''}\sum_n e^{-iE_n(t')}c_n(t')\mathbf{E}(t')\cdot\mathbf{d}_n(\mathbf{p}+\mathbf{A}(t'))$$

With $\mathbf{d}_n(\mathbf{p}) = \langle\mathbf{p}|\hat{\mathbf{d}}|\Psi_n\rangle$ This is the equation where most papers start with [?].

Note that SFA is not about strong laser pulses since here we are also dealing with small ionisation proabilities (<0.01) so SFA states that when ionisation does happen (regardless if its unlikely) the laser pulse will then be the dominant force.

I need to derive in paper from manoram 2023 the same thing as app A just instead of $\hat{p}$ i use $\hat{1}$ and use instead of $|\Psi\rangle_0$ i use the expansion in eigenstates from my project plan

2.3 Derivation of SFA Rate

This mainly follows [**?**] with some modification.
The ground state is now a superosition $\sum_n c_n(t) |\Psi_n\rangle$.
We can write the SFA rate as:

$$\langle\Psi(t)|\hat{\mathbf{1}}|\Psi(t)\rangle = \int_{-\infty}^{\infty} \Gamma(t)\mathrm{d}t$$

$$= \int \mathrm{d}^3 p \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathrm{d}t_1\mathrm{d}t_2\, e^{\frac{i}{2}\int_{t_1}^{t_2}(\mathbf{p}+\mathbf{A}(t''))^2\mathrm{d}t''} \mathbf{E}(t_1)\cdot\mathbf{E}(t_2)$$

$$\times \left(\sum_n e^{iE_n t_1} c_n^*(t_1)\mathbf{d}_n^*(\mathbf{p}+\mathbf{A}(t_1))\right)\cdot\left(\sum_n e^{-iE_n t_2} c_n(t_2)\mathbf{d}_n(\mathbf{p}+\mathbf{A}(t_2))\right)$$

Changing variables to $t = \frac{t_2+t_1}{2}$ and $T = \frac{t_2-t_1}{2}$ and using the fact that our Laser pulse is polarized along the z Axis gives us:

$$\Gamma(t) = \int \mathrm{d}^3 p \int_{-\infty}^{\infty} \mathrm{d}T\, e^{2iI_p T+\frac{i}{2}\int_{t-T}^{t+T}(\mathbf{p}+\mathbf{A}(t''))^2\mathrm{d}t''} E_z(t-T)\cdot E_z(t+T)$$

$$\times \left(\sum_n e^{iE_n(t-T)} c_n^*(t-T)d_{z,n}^*(\mathbf{p}+\mathbf{A}(t-T))\right)\cdot\left(\sum_n e^{-iE_n(t+T)} c_n(t+T)d_{z,n}(\mathbf{p}+\mathbf{A}(t+T))\right)$$

$$\Gamma(t) = \sum_{n_1}\sum_{n_2} \int \mathrm{d}^3 p \int_{-\infty}^{\infty} \mathrm{d}T\, e^{\frac{i}{2}\int_{t-T}^{t+T}(\mathbf{p}+\mathbf{A}(t''))^2\mathrm{d}t''} e^{iE_{n_1}(t-T)-iE_{n_1}(t+T)}$$

$$\times E_z(t-T)E_z(t+T)c_{n_1}^*(t-T)c_{n_2}(t+T)d_{z,n_1}^*(\mathbf{p}+\mathbf{A}(t-T))d_{z,n_2}(\mathbf{p}+\mathbf{A}(t+T))$$

2.4   Strong Field Ionization

Decied to do Phenomenology after theory.
Phenomenology of strong field ionization, Different types of Ionization, tunneling Ionization, multiphoton, stark effect.

3. Ionization Model

3.1 GASFIR

test

3.2 Python Implementation

A general approximator for strong field ionization rates

3.3 TIPTOE

TIPTOE [?] is a sampling method used for sub femtosecond processes. It is relevant for this thesis because it was used to verify the results from the Ionization model. TIPTOE is great because its fundamentals are very simple but it can tell you a lot about the dynamics in attosecond regime.

3.3.1 Time reversal symmetry

TIPTOE -> we found out time reversal symmetry is violated. Normally TRS comes from ...

4. Numerical Methods

4.1 tRecX

I need to check if trecx coeff are trustable, use pertubation theory first order, for n»1 trecx should predict different results because pert theory is not valid anymore

I also need to check how often I have to write the coefficients to the expec file. That depends on the characterisitc time of the state and on the frequency of the laser of course.

in E4 it was easy because laser had cosine shape. attosecond physics not the case, more a lase pulse, cos8 envelope so it doesnt make much sense to speak of rabi oszillations.

Difference between length gauge and velocity gauge in numerics

Why is it so diffficult? Solving time dependent Schroedinger euation numerically is not that hard. But in attosecond regime electron in Hydrogen likes length gauge because everything can be defined by giving the position of the electron. But a free electron in a strong laser pulse really likes velocity gauge because everything can be described by the kinetic energy of the electron.

Since we dont know how trecx outputs the coefficients (in which pciture) we need to check that.

4.1.1 irECS

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4.1.2 tSURFF

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4.1.3 Challenges

dangling pointer: interesting problem actually, how to solve it, how to find it, etc

4.2   Python Implementation of Ionization Model

it uses a kernel, quasistatic

5. Results and Discussion

5.1 Laser Fields

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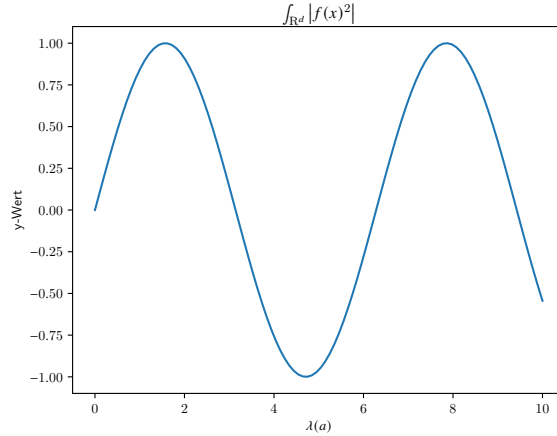$$\partial_t u = \mathcal{H}(t)\lambda \tag{5.1}$$

Figure 5.1: Sine function

$$\partial \mathbf{A} = \mathfrak{B}$$

$$\int_{\mathbb{R}^d} |f(x)|^2 \mathrm{d}x = \int_{\mathbb{R}^d} |\mathcal{F}f(\xi)|^2 \mathrm{d}\xi \tag{5.2}$$

$$\mathrm{i}\partial_t u = \mathcal{H}(t) \ket{a} \lambda \tag{5.3}$$

6. Conclusion and Outlook

Could be good for GASFIR because GASFIR learns from exact SFA rate.
What is more important, stark shift or else?

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A. Hydrogen Atom

Solving TDSE for Hydrogen atom because it will be important later. also note the structure of wavefunction:

$$\psi_{nlm}(\underline{x}) = \psi_{nlm}(r, \theta, \phi) = R_{nl}(r) Y_{lm}(\theta, \phi)$$

$E_n = \frac{Z^2}{2n^2}$

B. Dipole transition matrix elements

We want to derive the general transition dipole matrix elements into the continuum for an hydrogen-like atom. The general matrix element in our case is given by:

$$\underline{d}(\underline{p}) = \langle \Pi | \hat{\mathbf{d}} | \Psi_{nlm} \rangle \overset{\text{a.u.}}{=} \langle p | \hat{\mathbf{r}} | \Psi_{nlm} \rangle$$

With $|p\rangle$ being a plane wave. By partitioning the $\hat{\mathbf{1}}$, and using the fact that $\hat{\mathbf{r}} \to i\nabla_{\underline{p}}$ in momentum representation we find a generel formula for the transition:

$$\underline{d}(\underline{p}) = i\nabla_{\underline{p}} \int \mathrm{d}^3\underline{x}\,\psi_{nlm}(\underline{x})e^{-i\underline{p}\cdot\underline{x}} = i\nabla_{\underline{p}}\phi_{nlm}(\underline{p})$$

In principle, this integral or more precise the Fouriertransformation of the wavefunction is all we need to do. Because of the structure of ψ_{nlm} we can expect a result simular to (eqref psi=RY). A posteriori we will see that:

$$\mathcal{F}\{\psi_{nlm}(\underline{x})\} = \phi_{nlm}(\underline{p}) = F_{nl}(p)Y_{lm}(\theta_p, \phi_p)$$

With $F_{nl}(p)$ being the Fouriertransform of the radial part of the wavefunction and $Y_{lm}(\theta_p, \phi_p)$ being the spherical harmonics in momentum space similar to the hydrogen atom in position space.

Momentum space

We start with the so called plane wave expansion [**?**] of the exponential part of the integral:

$$e^{i\underline{p}\cdot\underline{x}} = \sum_{l'=0}^{\infty}(2l'+1)i^{l'}j_{l'}(pr)P_{l'}(\underline{p}\cdot\underline{x}) = 4\pi\sum_{l'=0}^{\infty}\sum_{m'=-l'}^{l'}i^{l'}j_{l'}(pr)Y_{l'm'}(\theta_p,\phi_p)Y_{l'm'}^*(\theta_x,\phi_x)$$

With $j_l(pr)$ being the spherical bessel functions. Also note that we are integrating over spherical kordinates now. At first it looks messy but we can use the orthogonality of the spherical harmonics we can reduce the integral to:

$$\phi_{nlm}(\underline{p}) = 4\pi\sum_{m=-l}^{l}Y_{lm}(\theta_p,\phi_p)i^l\underbrace{\int_0^{\infty}\mathrm{d}r\,r^2 j_l(pr)R_{nl}(r)}_{\tilde{R}_{nl}(p)}$$

This is the structure we were hoping for. Lets focus on the radial part $\tilde{R}_{nl}(p)$ of the integral. The term $R_{nl}(r)$ represents the radial function of the hydrogen atom in position space and is

independent of the magnetic bumber m. An exponential term dependent of r, a polynomial term dependent of r, the generalized Laguerre polynomials and the normalization constant. It would be convenient to have a closed expression for the generalzied Laguerre polynomials. I choose to represent them as following:

$$L_n^l(r) = \sum_{\iota=0}^{n} \frac{(-1)^\iota}{\iota!} \binom{n+l}{n-\iota} r^\iota$$

The Laguerre polynomials are therefore only dependent on and exponential term and finitely many polynomial terms. $\tilde{R}_{nl}(p)$ can be expressed (without prefactors and summation over ι) as:

$$\int_0^\infty dr \, r^{2+l+\iota} e^{-\frac{Zr}{n}} j_l(pr)$$

Before we can solve the Integral using caomputational methods, we need to transform the spherical bessel function into the ordinary ones:

$$j_l(pr) = \sqrt{\frac{\pi}{2pr}} J_{l+\frac{1}{2}}(pr)$$

Now it is a good time to write all the prefactors and summations in one expression and look at the integral as a whole:

$$\phi_{nlm}(\underline{p}) = \frac{\pi^{3/2}}{\sqrt{2p}} \sqrt{\left(\frac{2}{n}\right)^3 \frac{(n-l-1)!}{n(n+1)!}}$$
$$\times \sum_{m=-l}^{l} \sum_{\iota=0}^{n-l-1} i^l \frac{(-1)^\iota}{\iota!} \left(\frac{2}{n}\right)^{l+\iota} \binom{n+l}{n-l-1} \underbrace{\int_0^\infty dr \, r^{l+\iota+\frac{3}{2}} e^{-\frac{Zr}{n}} J_{l+\frac{1}{2}}(pr)}_{(*)} Y_{lm}(\theta_p, \phi_p)$$

To calculate the remaining Integral, I used mathematica, so I can not give a detailed explanation of that. Interestingly, there is an analytical solution for that. The result for $(*)$ is:

$$(*) = {}_2\tilde{F}_1\left(2+l+\frac{\iota}{2}, \frac{1}{2}(5+2l+\iota); \frac{3}{2}+l; -\frac{n^2p^2}{Z^2}\right)$$

With ${}_2\tilde{F}_1$ being the regularized hypergeometric function defined by:

$$_2\tilde{F}_1(a,b;c;z) = \frac{_2F_1(a,b;c;z)}{\Gamma(c)} = \frac{1}{\Gamma(a)\Gamma(b)} \sum_{n=0}^{\infty} \frac{\Gamma(a+n)\Gamma(b+n)}{\Gamma(c+n)} \frac{z^n}{n!}$$

The final formula $\phi_{nlm}(\underline{p})$ that can be also found in [atoms and molekulse] in slightely different form, can then be expressed as:

$$\phi_{nlm}(\underline{p}) = \sum_{\iota=0}^{2l+1} \frac{(-1)^\iota \, 2^{\iota+\frac{1}{2}} \, n \, (in)^l \, (p^2)^{l/2} \, Z^{-l-3} \, \Gamma(2l+\iota+3)}{\iota!}$$
$$\times \binom{l+n}{-l+n-\iota-1} \sqrt{\frac{Z^3\Gamma(n-l)}{\Gamma(l+n+1)}}$$
$$\times Y_l^m(\theta_p, \phi_p) \, _2\tilde{F}_1\left(l+\frac{\iota}{2}+2, \frac{1}{2}(2l+\iota+3); l+\frac{3}{2}; -\frac{n^2p^2}{Z^2}\right) \qquad \text{(B.1)}$$

Transition Element

Now all thats left is to differentiate (**??**) with respect to $\underline{p}$.

new eq

$$\sum_{\iota=0}^{2l+1} \left(-\frac{(-1)^{\iota}n\mathrm{Ip}^{-l-3}2^{\iota-l-1}(in)^{l}\left(p^{2}\right)^{l/2}\left(\frac{1}{\sqrt{p^{2}}}-\frac{\mathrm{pz}^{2}}{(p^{2})^{3/2}}\right)\Gamma(2l+\iota+3)\binom{l+n}{-l+n-\iota-1}\sqrt{\frac{\mathrm{Ip}^{3}\Gamma(n-l)}{\Gamma(l+n+1)}}\,_{2}\tilde{F}_{1}\left(l+\frac{\iota}{2}+2,\frac{1}{2}(2\right.}{}\right.$$

(B.2)

$$\sum_{\iota=0}^{2l+1} \left(-\frac{(-1)^{\iota}n\,\mathrm{Ip}^{-l-3}2^{\iota-l-1}(in)^{l}(p^{2})^{l/2}\left(\frac{1}{\sqrt{p^{2}}}-\frac{\mathrm{pz}^{2}}{(p^{2})^{3/2}}\right)\Gamma(2l+\iota+3)\binom{l+n}{-l+n-\iota-1}\sqrt{\frac{\mathrm{Ip}^{3}\Gamma(n-l)}{\Gamma(l+n+1)}}}{\iota!\sqrt{1-\frac{\mathrm{pz}^{2}}{p^{2}}}}\right.$$

$$\times\,_{2}\tilde{F}_{1}\left(l+\frac{\iota}{2}+2,\frac{1}{2}(2l+\iota+3);l+\frac{3}{2};-\frac{n^{2}p^{2}}{4\mathrm{Ip}^{2}}\right)\left(\frac{m\,\mathrm{pz}\,Y_{l}^{m}\left(\cos^{-1}\left(\frac{\mathrm{pz}}{\sqrt{p^{2}}}\right),\tan^{-1}(\mathrm{px},\mathrm{py})\right)}{\sqrt{p^{2}}\sqrt{1-\frac{\mathrm{pz}^{2}}{p^{2}}}}\right.$$

$$\left.+\frac{\sqrt{\Gamma(l-m+1)}\sqrt{\Gamma(l+m+2)}e^{-i\tan^{-1}(\mathrm{px},\mathrm{py})}Y_{l}^{m+1}\left(\cos^{-1}\left(\frac{\mathrm{pz}}{\sqrt{p^{2}}}\right),\tan^{-1}(\mathrm{px},\mathrm{py})\right)}{\sqrt{\Gamma(l-m)}\sqrt{\Gamma(l+m+1)}}\right)$$

$$+\frac{(-1)^{\iota}ln\,\mathrm{pz}\,\mathrm{Ip}^{-l-3}2^{\iota-l-1}(in)^{l}(p^{2})^{\frac{l}{2}-1}\Gamma(2l+\iota+3)\binom{l+n}{-l+n-\iota-1}\sqrt{\frac{\mathrm{Ip}^{3}\Gamma(n-l)}{\Gamma(l+n+1)}}}{\iota!}\,_{2}\tilde{F}_{1}\left(l+\frac{\iota}{2}+2,\frac{1}{2}(2l+\iota+3);\right.$$

$$-\frac{(-1)^{\iota}n^{3}\mathrm{pzIp}^{-l-5}2^{\iota-l-3}\left(\frac{\iota}{2}+l+2\right)(\iota+2l+3)(in)^{l}(p^{2})^{l/2}\Gamma(2l+\iota+3)\binom{l+n}{-l+n-\iota-1}\sqrt{\frac{\mathrm{Ip}^{3}\Gamma(n-l)}{\Gamma(l+n+1)}}}{\iota!}\,_{2}\tilde{F}_{1}\left(l-\right.$$

(B.3)

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C. Semi-classical Hamiltonian

Here we will derive semi classical Hamiltonian. This part mainly follows [**?**].

$$\hat{\mathcal{H}}(\underline{x}, t) = \frac{1}{2m}(\hat{\mathbf{P}} - e\mathbf{A}(\underline{x}, t))^2 + e\varphi(\underline{x}, t) \tag{C.1}$$

D. TDSE Solution

In this section we will solve the TDSE according to the method mentioned above. With parameter trick using lebesgue measure theory elstroed page 162.

E. Code

```python
for state in range(excitedStates):
    for stateRange in range(excitedStates):
        cLeft = coefficients[state, :]
        cRight = coefficients[stateRange, :]
        f0 = np.zeros((Tar.size, tar.size), dtype=np.cdouble)
        phase0 = np.zeros((Tar.size, tar.size), dtype=np.cdouble)
        for i in prange(Tar.size):
            Ti=Ti_ar[i]
            for j in range(tar.size):
                tj=N+nmin+j*n
                tp=tj+Ti
                tm=tj-Ti
                if tp>=0 and tp<EF.size and tm>=0 and tm<EF.size:
                    VPt = 0 # VP[tj]
                    T= Ti*dT
                    DelA = (intA[tp] - intA[tm])-2*VPt*T
                    VP_p=VP[tp]-VPt
                    VP_m=VP[tm]-VPt
                    counter += 1
                    #print("counter", counter)        #first state and
                        normal SFA are exactly 4pi apart
                    nL, lL, mL = config[state]
                    nR, lR, mR = config[stateRange]
                    f_t_1= np.conjugate(transitionElementtest(nL, lL,
                        mL, p, pz, VP_m, E_g))*transitionElementtest(nR,
                        lR, mR, p, pz, VP_p, E_g)
                    #f_t_1= (pz+VP_p)/(p**2+VP_p**2+2*pz*VP_p+2*E_g)
                        **3*(pz+VP_m)/(p**2+VP_m**2+2*pz*VP_m+2*E_g)**3
                    G1_T_p=np.trapz(f_t_1*np.exp(1j*pz*DelA)*np.sin(
                        theta), Theta_grid)
                    G1_T=np.trapz(G1_T_p*window*p_grid**2*np.exp(1j*
                        p_grid**2*T), p_grid)
                    DelA = DelA + 2 * VPt * T
                    phase0[i, j]  = (intA2[tp] - intA2[tm])/2  + T*VPt
                        **2-VPt*DelA + eigenEnergy[state]*tm -
                        eigenEnergy[stateRange]*tp
                    f0[i, j] = EF[tp]*EF[tm]*G1_T*np.conjugate(cLeft[tm
                        ])*cRight[tp]#(np.real(c[tp])*np.real(c[tm])+np.
                        imag(c[tp])*np.imag(c[tm]))
        print("state", state, "stateRange", stateRange)
        print("config", config[state], "configRange", config[stateRange
            ])
        plt.plot(tar, 2*np.real(IOF(Tar, f0, (phase0)*1j)))
        plt.show()
```

```
34          plt.close()
35          rate += 2*np.real(IOF(Tar, f0, (phase0)*1j))    #*c[np.newaxis,
               :]
36     return rate
```

testa

```cpp
#include <iostream>

int main() {
    std::cout << "Hello, World!" << std::endl;
    return 0;
}
```

Bibliography

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Hiermit erkläre ich, die vorliegende Arbeit selbständig verfasst zu haben und keine anderen als die in der Arbeit angegebenen Quellen und Hilfsmittel benutzt zu haben.

München, den 20.6.2025

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Unterschrift