



MAX-PLANCK-INSTITUT
FÜR QUANTENOPTIK



LUDWIG-
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UNIVERSITÄT
MÜNCHEN

Bachelor's Thesis

The role of excited atomic states in multiphoton ionization

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Supervised by:

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Prof. Ulrich Schollwöck

June 6, 2025



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Bachelorarbeit

Die Rolle angeregter atomarer Zustände bei der Mehrphotonenionisation

Johannes Porsch



FAKULTÄT FÜR PHYSIK

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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 \mathcal{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.

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

The strucutre in this chapter mainly follows [2] with some modifications.

2.1 Basic Formalism

Our goal is to come up with an expression where we can use the strong field approximation effectively. 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. For that we first need to solve the schrodinger equation for a given Hamiltonian.

We will come across some difficulties like gauge dependence.

2.1.1 Schrödinger Equation

The time evolution of a quantum system is given by the time dependent Schrödinger equation and a general hamiltonian

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

The formal solution depends on the time dependence of the hamiltonian and the physical setting. With no further assumptions about our Hamiltonian because $[\hat{\mathcal{H}}(t), \hat{\mathcal{H}}(t')] \neq 0$ we can write the formal solution to (2.1) as a Dyson series: The solution is then given by

$$|\Psi(t)\rangle = \hat{\mathcal{U}}(t) |\Psi(0)\rangle = \hat{1} + \sum_{n=1}^{\infty} (-i)^n \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \cdots \int_{t_0}^{t_{n-1}} dt_n \hat{\mathcal{H}}(t_n) \hat{\mathcal{H}}(t_{n-1}) \cdots \hat{\mathcal{H}}(t_1) |\Psi(0)\rangle. \quad (2.2)$$

2.1.2 Interaction picture and Projection operators

Solving the schrodinger equation can be cumbersome, especially when we have to deal with a time dependent Hamiltonian that doesn't commute with itself at different times. To make the calculations easier we can use projection operators with a method called feshbach method within the interaction picture.

2.1.3 Light-Matter Interaction

A light wave is defined by the Maxwell equations

$$\begin{aligned} \nabla \cdot \mathbf{E} &= \rho & \nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} \\ \nabla \cdot \mathbf{B} &= 0 & \nabla \times \mathbf{B} &= \mathbf{J} + \frac{\partial \mathbf{E}}{\partial t} \end{aligned}$$

The Maxwell equations are being solved by

$$\begin{aligned}\mathbf{E} &= -\nabla\varphi - \frac{\partial\mathbf{A}}{\partial t} \\ \mathbf{B} &= \nabla \times \mathbf{A}\end{aligned}\tag{2.3}$$

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

$$\begin{aligned}\mathbf{A} &\rightarrow \mathbf{A} + \nabla\chi \\ \varphi &\rightarrow \varphi - \frac{\partial\chi}{\partial t}\end{aligned}$$

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.

2.1.4 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 exactly one makes this assumption, first we rewrite the Maxwell equations in the dependence of the vector potential and the scalar potential as defined in (2.3). This will result in two coupled differential equations, what does not bring us any further. However we are interested 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 known. Now the Maxwell equations are uncoupled and can be written as

$$\begin{aligned}\nabla^2\varphi - \frac{\partial^2\varphi}{\partial t^2} &= \rho \\ \nabla^2\mathbf{A} - \frac{\partial^2\mathbf{A}}{\partial t^2} &= \mathbf{J}\end{aligned}$$

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

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

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

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

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

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

Even though 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 Field in the Hamiltonian. For that we need to think more about the gauge of our system.

2.1.5 Gauges

What makes calculating ionisation rates in strong field physics so difficult is the gauge, ie deciding which one you want to choose and when.

First, I will derive two basic expressions for the Hamiltonian in the so called velocity gauge and length gauge using the dipole approximation. It will be helpful to look at the semi classical Hamilton function of a free electron in an electric field¹:

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

In the dipole Approximation, this can be simplified to:

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

¹Because its interesting it will be derived in C

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 our desired gauge transformation, 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.

This is a common way to write the interaction Hamiltonian. However if we choose another gauge transformation, the equations will have a different form. look bossmann!!!

2.2 Strong Field Approximation

The difficulty with ionisation arises because we now have in some sense two Hilbert spaces, one for the states in the Hydrogen atom that deals with some distortion of the wavefunction because of the Laser field and one for the continuum states that are affected mainly by the Laser field but also by the binding potential.

We will see SFA will take care about the second Hilbertspace and make it easier. The main goal of this thesis is to see how much the Laser field has an effect on the wavefunction before ionisation happens. Previous work neglects this part so there is just one hilbertspace for the eigenstates unaffected by the laser field and one hilbertspace for the continuum states that are unaffected by the binding potential.

2.2.1 Subspaces

First we project the full timedependent Hamiltonian $\hat{\mathcal{H}}(t)$ onto subspaces using projection operators defined by:

$$\hat{X} = \sum_n |\Psi_n\rangle \langle \Psi_n| \quad \text{and} \quad \hat{Y} = \hat{1} - \hat{X}$$

With $|\Psi_n\rangle$ being the bound states of our atom. We can write:

$$\hat{\mathcal{H}}(t) = \underbrace{\hat{X}\hat{\mathcal{H}}(t)\hat{X}}_{\hat{\mathcal{H}}^X(t)=\hat{\mathcal{H}}_0(t)} + \underbrace{\hat{Y}\hat{\mathcal{H}}(t)\hat{Y} + \hat{X}\hat{\mathcal{H}}(t)\hat{Y} + \hat{Y}\hat{\mathcal{H}}(t)\hat{X}}_{\hat{\mathcal{H}}^Y(t)+\hat{\mathcal{H}}^{XY}(t)+\hat{\mathcal{H}}^{YX}(t)=\hat{\mathcal{H}}_I(t)}$$

Lets think about what these terms mean. $\hat{\mathcal{H}}^X(t)$ can be seen as just our quantum system but with a small distortion by our electric field. This part causes effects like Stark shift both in the eigenstates and eigenenergies of our atom. As long as the laser pulse is not too strong and no ionisation has happened, we can treat the pulse like a perturbation to the system. The distortion may be time dependent but the good thing is that we can easily determine the propagator with respect to $\hat{\mathcal{H}}^X(t)$ by using the interaction picture and solving a system of coupled differential equations. In other words this Hamiltonian determines the time evolution of the first hilbert space, as mentioned above.

Lets think about the other terms. For that we need to establish a setting. Say our Atom sits in the ground state and gets ionised. Based on this image we can say two from three parts are unnecessary. First $\hat{\mathcal{H}}^Y(t)$ plays no role because we project the initial state into the continuum state, and since there can be no overlap between them this part will play no role. This term actually just describes the evolution of a continuum state that remains a continuum state after the interaction so the laser pulse has happened. Second $\hat{\mathcal{H}}^{XY}(t)$ plays no role either since we project the initial (bound) state into the continuum. This term

would determine the time evolution of a continuum state that recombines with the atom after the interaction has happened. Obviously that's not what we want. The only term that remains is $\hat{\mathcal{H}}^{YX}(t)$ which describes the time evolution of a bound state that gets ionised in the continuum by the laser pulse. This is the perfect part to later start the strong field approximation with.

2.2.2 Avoiding Dyson series

For determining the time evolution we need to be careful since the whole Hamiltonian doesn't commute with itself at different times so we need to be exact. Since the full Dyson series (2.2) can be cumbersome to deal with, we choose a different way. What helps us is the fact that we can split the Hamiltonian by projecting it into subspaces in two parts of which one can be solved directly as mentioned above. Let's first write our ansatz:

$$\hat{\mathcal{U}}(t, t_0) = \hat{\mathcal{U}}_0(t, t_0) - i \int_{t_0}^t \hat{\mathcal{U}}(t, t') \hat{\mathcal{H}}_I(t') \hat{\mathcal{U}}_0(t', t_0) dt' \quad (2.6)$$

It is very easy to show that (2.6) is a solution to (2.1).

In our setting we start with the ground state of the atom so the time evolution is given by $|\Psi_n(t)\rangle = \hat{\mathcal{U}}(t, t_0) |\Psi_n(t_0)\rangle$. To make things even simpler, we project this state into a continuum state $|\Pi(t_c)\rangle$ at time t_c with t_c being sufficiently big enough for the electron to be in the continuum ($t_c \gg t'$).

Of course, there is no overlap between $|\Pi(t_c)\rangle$ and $\hat{\mathcal{U}}_0(t, t_0) |\Psi_n(t_0)\rangle$ since the electron did not get ionised yet. Furthermore, if we expand $\hat{\mathcal{H}}_I(t')$ and remind ourselves about the orthogonality of the bound states and the continuum states, we see that most of the terms of $\hat{\mathcal{H}}_I(t')$ vanish. We are being left with:

$$\langle \Pi(t_c) | \Psi_n(t) \rangle = -i \int_{t_0}^t \langle \Pi(t_c) | \hat{\mathcal{U}}(t, t') \hat{Y} \hat{\mathcal{H}}(t') \hat{X} \hat{\mathcal{U}}_0(t', t_0) | \Psi_n(t_0) \rangle dt' \quad (2.7)$$

Note: No approximations have been made, (2.7) is exact.

2.2.3 Strong Field Approximation

Before making the strong field approximation, let's think about (2.7) again. It is best to read this equation from right to left, starting with the initial state of our system and the propagation of the system in presence of a weak electric field before ionisation. At moment t' the Laser starts to interact with the system and it transitions into a virtual state. From time t' to the observed time t the system is described by the full Hamiltonian including both Laser Field and the binding potential.

In principle, SFA is the neglecting of exactly this binding 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 time evolution operator after ionisation as:

$$\hat{\mathcal{U}}(t, t') \approx \hat{\mathcal{U}}_{\text{SFA}}(t, t') = e^{-i \int_{t'}^t \hat{\mathcal{H}}_{\text{SFA}}(t'') dt''} \quad \text{and} \quad \mathcal{H}_{\text{SFA}}(t') = \hat{\mathcal{H}}(t) - \hat{V}_C$$

This is very usefull because for the eigenstates of $\hat{\mathcal{H}}_{\text{SFA}}$ we know an exact analytical solution; the Volkov states. Note that we can explicitly write $\hat{\mathcal{U}}_{\text{SFA}}(t, t')$ in (2.2.3) since after ionisation the hamiltonian commutes with itself at different times. For that lets take another look at the semi classical Hamilton (2.4). Classically, the physics driven by the momentum operator $\hat{\mathbf{P}}$ is known as the canonical momentum and given by:

$$\frac{\partial \mathcal{L}}{\partial \dot{\mathbf{x}}} = \underline{P} = m \dot{\mathbf{x}} + \frac{e}{c} \mathbf{A} \stackrel{\text{a.u.}}{=} \mathbf{p} + \mathbf{A} \quad (2.8)$$

With \mathcal{L} being the Lagrangian of the system. 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 [4]:

$$\frac{d\mathbf{p}}{dt} = \mathbf{E} + (\dot{\mathbf{x}} \times \mathbf{B}) \approx -\frac{\partial \mathbf{A}}{\partial t} = -\frac{d\mathbf{A}}{dt}$$

so $\frac{d}{dt} \underline{P} = 0$. And also the energy of the system is clear:

$$E(t) = \dot{\mathbf{x}} \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{x}}} - \mathcal{L} = \frac{\mathbf{p}^2}{2} \quad (2.9)$$

Note that the energy is not conserved because the argument we made befor does not hold for the kinetic momentum only for the canonical momentum. Now comes the interesting part. Clearly $|\Pi(t_c)\rangle$ (not $|\mathbf{p}(t_c)\rangle$!) is a solution of $e^{-i \int_{t'}^t \hat{\mathcal{H}}_{\text{SFA}}(t'') dt''}$ so combining (2.8) and (2.9) gives us:

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

\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:

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

Furthermore (how??)

$$|\Pi\rangle = |\underline{P}\rangle = |\mathbf{p} + \mathbf{A}\rangle$$

Combining all these equations give us the following expression:

$$\langle \Pi(t_c) | \Psi_n(t) \rangle = -i \int_{t_0}^t e^{-i \int_{t'}^t (P - \mathbf{A}(t''))^2 dt''} \hat{\mathcal{U}}_0(t', t_0) \langle \mathbf{p} + \mathbf{A} | \hat{Y} \hat{\mathcal{H}}(t') \hat{X} | \Psi_n(t_0) \rangle dt'$$

In this equation, $\langle \mathbf{p} + \mathbf{A} | \hat{Y} \hat{\mathcal{H}}(t') \hat{X} | \Psi_n(t_0) \rangle$ with some simplification is just the transition dipole matrix element $\mathbf{d}_n(\mathbf{p})$ between the bound state and the continuum state, generated by the laser pulse (ionisation).

Furthermore we make an ansatz for $\hat{\mathcal{U}}_0(t', t_0) | \Psi_n(t_0) \rangle = | \Psi_n(t) \rangle$ using the interaction picture. In contrast to other literature ([1], [2]) I am not neglecting transitions between different bound states before the ionisation. For instance it can happen that the laserpulse excites the electron but doesnt ionise it quite yet. But we still have a price to pay. Because with this ansatz we neglect explicit coupling to the continuum and artificially force the electron into staying inside the atom even though it may have enough energy to ionize. So we have to note for later that this ansatz is only valid for very small ionisation propabilities and even then its still an approximations.

Our expression then reads:

$$\langle \Pi(t_c) | \Psi_n(t) \rangle = -\frac{i}{2} \int_{t_0}^t e^{-i \int_{t'}^t (P - A_z(t''))^2 dt''} E_z(t') \sum_n c_n(t) e^{-i E_n t'} \langle \mathbf{p} + A_z | \hat{d}_z | \Psi_n \rangle dt'$$

Where we used the fact that the electric field is polarized along the z axis. This is the equation where most papers start with [1] known as Strong field s matrix.

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.

The interesting part is that the dynamics before ionizing the atom is exact so no approximations were made

2.3 Derivation of SFA Rate

This mainly follows [1] with some modification.

The ionization rate integrated over the time domain should yield the total ionization probability. Therefore we can write the SFA rate (2.2.3) as:

$$\begin{aligned} \lim_{t \rightarrow \infty} \langle \Psi(t) | \int d^3p |p\rangle \langle p| | \Psi(t) \rangle &= \int_{-\infty}^{\infty} \Gamma_{\text{SFA}}(t) dt \\ &= \int d^3p \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt_1 dt_2 e^{\frac{i}{2} \int_{t_1}^{t_2} (\underline{p} + A_z(t''))^2 dt''} E_z(t_1) E_z(t_2) \\ &\quad \times \left(\sum_n e^{iE_n t_1} c_n^*(t_1) d_{z,n}^*(\underline{p} + A_z(t_1)) \right) \left(\sum_n e^{-iE_n t_2} c_n(t_2) d_{z,n}(\underline{p} + A_z(t_2)) \right) \end{aligned}$$

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:

$$\begin{aligned} \Gamma_{\text{SFA}}(t) &= \int d^3p \int_{-\infty}^{\infty} dT e^{\frac{i}{2} \int_{t-T}^{t+T} (\underline{p} + A_z(t''))^2 dt''} E_z(t-T) E_z(t+T) \\ &\quad \times \left(\sum_n e^{i(t-T)E_n} c_n^*(t-T) d_{z,n}^*(\underline{p} + A_z(t-T)) \right) \left(\sum_n e^{-i(t+T)E_n} c_n(t+T) d_{z,n}(\underline{p} + A_z(t+T)) \right) \\ &= \sum_{n_1} \sum_{n_2} \int_{-\infty}^{\infty} dp p^2 \int_0^{\pi} d\theta \sin \theta \int_0^{2\pi} d\phi \int_{-\infty}^{\infty} dT e^{\frac{i}{2} \int_{t-T}^{t+T} (\underline{p} + A_z(t''))^2 dt''} e^{i(t-T)E_{n_1} - i(t+T)E_{n_2}} \\ &\quad \times E_z(t-T) E_z(t+T) c_{n_1}^*(t-T) c_{n_2}(t+T) d_{z,n_1}^*(\underline{p} + A_z(t-T)) d_{z,n_2}(\underline{p} + A_z(t+T)) \end{aligned}$$

2.4 Strong Field Ionization

Decided to do Phenomenology after theory.

Phenomenology of strong field ionization, Different types of Ionization, tunneling Ionization, multiphoton, stark effect.

2.4.1 Stark effect

2.4.2 REMPI

2.4.3 Ionization rate

What is a ionization rate and why do we want to have it?

Dilemma: numerical simulations yield good ion propabilitiees but no ionization rate, analytical calculations yield good ionization rates but incorrect propabilities (see GASFIR paper).

Furthermore, no experimental method has been developed yet to measure ionization rates, only ionization propabilities - a quantum mechanical observable.

As long as the electromagnetic field is present, projecting onto bound/unbound states yields non-physical gauge-dependent predictions [14].

Tunneling time is imaginary, source???

3. Numerical Methods

The main reason for making these ionization models is because one wants to avoid solving the Schrödinger equation numerically because it is time consuming and contains numerical difficulties. However, for comparing and verifying the models, one needs to solve it numerically.

Furthermore, numerically solving the TDSE is just good for ionization probabilities and not rates.

Especially for expanding an already existing model, it is important to compare and see if the changes of the model are going into the right direction.

3.1 tRecX

I need to check if tRecX coefficients are trustworthy, use perturbation theory first order, for $n \gg 1$ tRecX should predict different results because perturbation theory is not valid anymore.

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

In E4 it was easy because the laser had a cosine shape. Attosecond physics is not the case, more a laser pulse, cos² envelope so it doesn't make much sense to speak of Rabi oscillations.

Difference between length gauge and velocity gauge in numerics

Why is it so difficult? Solving the time dependent Schrödinger equation numerically is not that hard. But in the attosecond regime an 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 don't know how tRecX outputs the coefficients (in which picture) we need to check that.

3.1.1 irECS

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3.1.2 tSURFF

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3.1.3 Challenges

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

4. Ionization Model

For retrieving ionisation rates, there are different ways of calculating the rates.

In this chapter I will explain some concepts and methods that are used to calculate the ionisation rates and what to do with them, how to verify them and so on.

4.1 SFA Python Implementation

The implementation of formula ?? was being done in python. Most of the implementation was already done by the Authors of [1]. The only part missing in the implementation was the extension to excited states. For that, major difficulty arises from the fact that we now need the coefficients $c_n(t)$ of the state before ionization.

4.1.1 Coefficients

In theory, $|c_n(t)|^2$ or the amplitude of the coefficients is observable quantity and can be measured in experiment. It tells us how probable the system is to be in a certain state n at time t . Furthermore, and most importantly, they are gauge independent. But since SFA is gauge dependent theory, and now we are not even dealing with the amplitude instead the complex coefficients, we need to be extremely careful with the gauge.

In my thesis there are mostly two gauges used, the length gauge and the velocity gauge. In the following plot you see that the amplitudes are indeed not gauge independent. But why is that the case?

The gauge is a very fundamental problem. Not so fundamental is our ansatz, as mentioned earlier we artificially force the electron to stay in the part of the Hilbert space covered by the bound state. This restricts us to small ionization probabilities, and may also cause some numerical problems as will be discussed later.

Also the numerical issue that we need to have infinitely many bound states for covering the part of the Hilbert space completely is not possible, so we have to limit ourselves. First

this seems like a big problem, but we will see that most of the dynamics inside the electron before ionisation is in my simulations only determined by a few bound states. But one still need to be carefull with the number of used bound states, but because of numerical reasons.

For my numerica solution I used a system of ordinary differential equations (ODEs) to calculate the coefficients $c_n(t)$ using the interaction picture. In my implementation I neglected transitions to states that are forbidden via the dipole selection rules. However this is an approximation, since in reality two-photon processes can occur, effectively allowing transitions between $1s$ and $2s$ for instance. I numerically solved the schroedinger equation with tRecX and modyfied the code to print out the coefficients $c_n(t)$ allowing me to get insight in the "real" dynamics of the electron.

4.1.2 Dipole matrix Elements

The dipole matrix elements are an essential part of the formula in (2.2.3). Calculating it generall can be cumbersome, but in the case of hydrogen its even possible to do it analytically B. However, its that easy to calculate because we made a very coarse approximation, since the final state after ionisation is in realtiy a plane wave, but we approximated it with that by using SFA.

Furthermore, our problem is

4.2 GASFIR

test A general approximator for strong field ionization rates

4.3 TIPTOE

TIPTOE [5] 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.

4.3.1 Time reversal symmetry

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

5. Results and Discussion

Before discussing the results, first we have to formulate what we want to learn from this kind of generalisation. The main difference from previous literature was the use of transition to excited states before ionisation. In principle we want to investigate how this transition influences the ionisation process or in general the ionisation rate.

Previously: time reversal symmetry, what causes it?

Also we want to know what influences the ionization rate more, the Stark shift or the distortion of the ground state or the possibility for the electron to also get excited to higher states instead of direct ionization.

So let's start with the Stark effect.

5.1 Stark Shift

The Stark effect is the shift of the energy levels of an atom or molecule due to the presence of an external electric field.

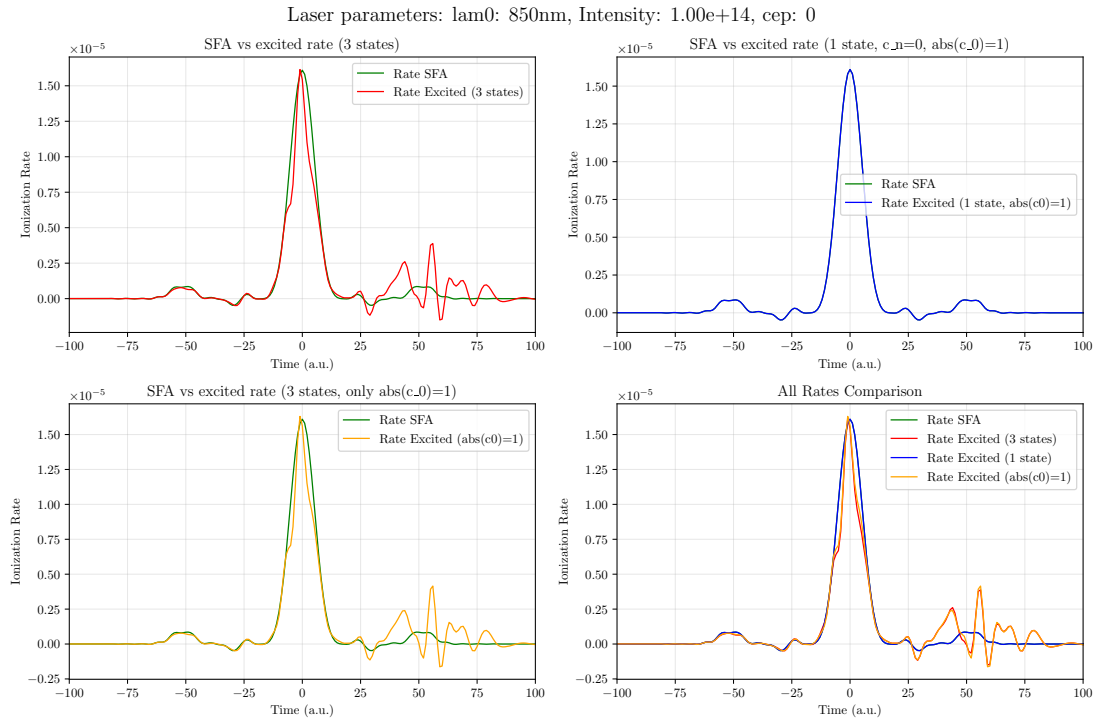


Figure 5.1: Sine function

Naiv: Stark effect changes energy in electron so its "harder" to ionise, thats why blue curve goes down (when excitedStates=1). But thats not certainly the case because of stark effect, thats why only set absc0 to 1 and phase remains. Example with oszillations with time dependent resonance frequency, and external force not at resonance but coincidence with oszillator resonance frequency so this may cause it.

5.2 Interpretation

Stark shift doesnt seem to have much contribution (sadly).

Lets investigate the influence of first coefficient, nothing more. Only the phase has a contribution, the amplitude is not important. Thats because the amplitude determines something occupation propability, but the phase is e^{-iEt} and if E is shifted by a bit you can isolate it by just using purely the phase.

Top right is the isolated stark effect

5.3 Lorem

This might be how it is supposed to be – we are in the regime where the laser field is strong enough to ionize the atom, but we artificially force the electron to stay in the part of the

Hilbert space covered by a few bound states. If the calculations are converged with respect to the time step, and you see convergence regarding the number of bound states in the weak-field regime, then it's fine if there is no convergence with respect to the number of bound states in the strong-field regime.

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. Volkov states

We will derive Volkov states in different gauges, starting with velocity gauge since its the most natural for an electron in a laser field to be described that way.

Velocity gauge

Lets write the Schroedinger equation without the Coulomb interaction part with the interaction with the laser in velocity gauge ?? using atomic units:

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = \left(\frac{\hat{\mathbf{p}}^2}{2} + \hat{\mathbf{p}} \cdot \mathbf{A}(t) + \frac{\mathbf{A}^2}{2}\right)|\Psi(t)\rangle$$

Lets use the momentum basis for the representation of the wavefunction:

$$i\frac{\partial}{\partial t}\phi(\underline{p}, t) = \left(\frac{\underline{p}^2}{2} + \underline{p} \cdot \mathbf{A}(t) + \frac{\mathbf{A}^2}{2}\right)\phi(\underline{p}, t)$$

This equation can be solved easily by maing the ansatz:

$$\phi(\underline{p}, t) = e^{-\frac{i}{2} \int_{t_0}^t (\underline{p} + \mathbf{A}(t'))^2 dt'}$$

In coordinate space this will result in:

$$\psi(\underline{x}, t) = \mathcal{F}^{-1}\{\phi(\underline{p}, t)\} = (2\pi)^{-3/2} e^{\underline{x} \cdot \underline{p} - \frac{i}{2} \int_{t_0}^t (\underline{p} + \mathbf{A}(t'))^2 dt'} \quad (\text{A.1})$$

Length gauge

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:

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

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

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

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 similar 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 [3] 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:

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

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 number 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 generalized 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 an 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 computational 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:

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

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:

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

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 slightly different form, can then be expressed as:

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

Transition Element

Now all thats left is to differentiate (B.1) with respect to \underline{p} .

new eq

$$\sum_{\iota=0}^{2l+1} \left(\frac{(-1)^\iota n \text{Ip}^{-l-3} 2^{\iota-1} (in)^l (p^2)^{l/2} \left(\frac{1}{\sqrt{p^2}} - \frac{\text{pz}^2}{(p^2)^{3/2}} \right) \Gamma(2l + \iota + 3) \binom{l+n}{-l+n-\iota-1} \sqrt{\frac{\text{Ip}^3 \Gamma(n-l)}{\Gamma(l+n+1)}} {}_2\tilde{F}_1\left(l + \frac{\iota}{2} + 2, \right.}{\dots} \right) \quad (\text{B.2})$$

$$\begin{aligned}
& \sum_{\iota=0}^{2l+1} \left(- \frac{(-1)^\iota n \text{Ip}^{-l-3} 2^{\iota-l-1} (in)^l (p^2)^{l/2} \left(\frac{1}{\sqrt{p^2}} - \frac{\text{pz}^2}{(p^2)^{3/2}} \right) \Gamma(2l + \iota + 3) \binom{l+n}{-l+n-\iota-1} \sqrt{\frac{\text{Ip}^3 \Gamma(n-l)}{\Gamma(l+n+1)}}}{\iota! \sqrt{1 - \frac{\text{pz}^2}{p^2}}} \right. \\
& \quad \times {}_2\tilde{F}_1 \left(l + \frac{\iota}{2} + 2, \frac{1}{2}(2l + \iota + 3); l + \frac{3}{2}; -\frac{n^2 p^2}{4\text{Ip}^2} \right) \left(\frac{m \text{pz} Y_l^m \left(\cos^{-1} \left(\frac{\text{pz}}{\sqrt{p^2}} \right), \tan^{-1}(\text{px}, \text{py}) \right)}{\sqrt{p^2} \sqrt{1 - \frac{\text{pz}^2}{p^2}}} \right. \\
& \quad \left. \left. + \frac{\sqrt{\Gamma(l-m+1)} \sqrt{\Gamma(l+m+2)} e^{-i \tan^{-1}(\text{px}, \text{py})} Y_l^{m+1} \left(\cos^{-1} \left(\frac{\text{pz}}{\sqrt{p^2}} \right), \tan^{-1}(\text{px}, \text{py}) \right)}{\sqrt{\Gamma(l-m)} \sqrt{\Gamma(l+m+1)}} \right) \right) \\
& \quad + \frac{(-1)^\iota l n \text{pz} \text{Ip}^{-l-3} 2^{\iota-l-1} (in)^l (p^2)^{\frac{l}{2}-1} \Gamma(2l + \iota + 3) \binom{l+n}{-l+n-\iota-1} \sqrt{\frac{\text{Ip}^3 \Gamma(n-l)}{\Gamma(l+n+1)}}}{\iota!} {}_2\tilde{F}_1 \left(l + \frac{\iota}{2} + 2, \frac{1}{2}(2l + \iota + 3); \right. \\
& \quad \left. - \frac{(-1)^\iota n^3 \text{pz} \text{Ip}^{-l-5} 2^{\iota-l-3} \left(\frac{\iota}{2} + l + 2 \right) (\iota + 2l + 3) (in)^l (p^2)^{l/2} \Gamma(2l + \iota + 3) \binom{l+n}{-l+n-\iota-1} \sqrt{\frac{\text{Ip}^3 \Gamma(n-l)}{\Gamma(l+n+1)}}}{\iota!} {}_2\tilde{F}_1 \left(l + \frac{\iota}{2} + 2, \frac{1}{2}(2l + \iota + 3); \right. \right. \\
& \quad \left. \left. \right) \right) \tag{B.3}
\end{aligned}$$

C. Semi-classical Hamiltonian

Here we will derive semi classical Hamiltonian. This part mainly follows [4].

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

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

```
1  for state in range(excitedStates):
2      for stateRange in range(excitedStates):
3          cLeft = coefficients[state, :]
4          cRight = coefficients[stateRange, :]
5          f0 = np.zeros((Tar.size, tar.size), dtype=np.cdouble)
6          phase0 = np.zeros((Tar.size, tar.size), dtype=np.cdouble)
7          for i in prange(Tar.size):
8              Ti=Ti_ar[i]
9              for j in range(tar.size):
10                 tj=N+nmin+j*n
11                 tp=tj+Ti
12                 tm=tj-Ti
13                 if tp>=0 and tp<EF.size and tm>=0 and tm<EF.size:
14                     VPt = 0 # VP[tj]
15                     T= Ti*dT
16                     DelA = (intA[tp] - intA[tm])-2*VPt*T
17                     VP_p=VP[tp]-VPt
18                     VP_m=VP[tm]-VPt
19                     counter += 1
20                     #print("counter", counter) #first state
21                     #and normal SFA are exactly 4pi apart
22                     nL, lL, mL = config[state]
23                     nR, lR, mR = config[stateRange]
24                     f_t_1= np.conjugate(transitionElementtest(nL, lL,
25                         mL, p, pz, VP_m, E_g))*transitionElementtest(
26                         nR, lR, mR, p, pz, VP_p, E_g)
27                     #f_t_1= (pz+VP_p)/(p**2+VP_p**2+2*pz*VP_p+2*E_g)
28                     #**3*(pz+VP_m)/(p**2+VP_m**2+2*pz*VP_m+2*E_g)
29                     **3
30                     G1_T_p=np.trapz(f_t_1*np.exp(1j*pz*DelA)*np.sin(
31                         theta), Theta_grid)
32                     G1_T=np.trapz(G1_T_p>window*p_grid**2*np.exp(1j*
33                         p_grid**2*T), p_grid)
34                     DelA = DelA + 2 * VPt * T
```

```

28         phase0[i, j] = (intA2[tp] - intA2[tm])/2 + T*
                VPt**2-VPt*DelA + eigenEnergy[state]*tm -
                eigenEnergy[stateRange]*tp
29         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]))
30         print("state", state, "stateRange", stateRange)
31         print("config", config[state], "configRange", config[
                stateRange])
32         plt.plot(tar, 2*np.real(IOF(Tar, f0, (phase0)*1j)))
33         plt.show()
34         plt.close()
35         rate += 2*np.real(IOF(Tar, f0, (phase0)*1j))      #*c[np.
                newaxis, :]
36     return rate

```

testa

```

1     #include <iostream>
2
3     int main() {
4         std::cout << "Hello, World!" << std::endl;
5         return 0;
6     }

```

Bibliography

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Declaration of Authorship

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

Unterschrift