

LUDWIG-MAXIMILIANS-UNIVERSITÄT
MÜNCHEN

Bachelors Thesis

The role of excited atomic states in multiphoton ionization

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Supervisor:
apl. Prof. Vladislav Yakovlev
Prof. Ulrich Schollwöck

April 23, 2025

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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.

Contents

Abstract	1
List of Figures	2
1 Introduction	3
2 Theory	4
2.1 Basic Formalism	4
2.1.1 Schrödinger Equation	4
2.1.2 Light-Matter Interaction	6
2.1.3 Dipole Approximation	6
2.2 Strong Field Approximation	8
2.3 Strong Field Ionization	9
2.4 Multiphoton Ionization	10
2.5 Transition matrix elements	11
3 Ionization Model	12
3.1 GASFIR	12
3.2 Python Implementation	12
3.3 TIPTOE	12
3.3.1 Time reversal symmetry	12
4 Numerical Methods	13
4.1 tRecX	13
4.1.1 irECS	13
4.1.2 tSURFF	13
4.1.3 Challenges	13
4.2 Python Implementation of Ionization Model	14
5 Results and Discussion	15
5.1 Laser Fields	15
6 Conclusion and Outlook	16
A Appendix A	17
Bibliography	18

List of Figures

5.1 Sine function	15
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1. Introduction

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2. Theory

Convention:

Ψ wavefunction for the whole system

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

$\psi(\underline{p})$ 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 $\hat{=}\mathbf{p}$.

This chapter mainly follows [2] 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 schröfingfer 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

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

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

¹How? 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.

given by

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

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.

2.1.2 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.

2.1.3 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 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 inhomogeneous 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. It will be helpful to look at the semi classical Hamilton function of a free electron in an electric field [3]:

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

In the dipole Approximation, this can be simplified to:

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

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. Now we perform another gauge transformation to the so called length gauge via

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

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 rewrite the time dependent part \hat{V} of our quantum mechanical Hamiltonian as

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

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

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 (2.1):

$$|\Psi(t)\rangle = -i \int_{t_0}^t dt' e^{-i \int_{t'}^t \hat{H}(t'') dt''} \hat{V}_I(t') e^{-i \int_{t_0}^{t'} \hat{H}_0(t'') dt''} |\Psi(0)\rangle + e^{-i \int_{t_0}^t \hat{H}_0(t'') dt''} |\Psi(0)\rangle \quad (2.6)$$

as can be checked by inserting the solution into the Schrödinger equation using the parameter Integral trick. 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 (2.6) is now gone because there can be no overlap between a continuum state and the initial state.

$$\langle \Xi(t_c) | \Psi(t) \rangle = -i \int_{t_0}^t dt' \langle \Xi(t = t_c) | e^{-i \int_{t'}^t \hat{H}(t'') dt''} \hat{V}_I(t') e^{-i \int_{t_0}^{t'} \hat{H}_0(t'') dt''} | \Psi(0) \rangle \quad (2.7)$$

With $|\Xi(t_c)\rangle$ 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 ($t_c \gg t'$). Equation (2.6) 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 instantaneously. Note that $|\Psi(0)\rangle$ is not the ground state of the Hydrogen atom, just the initial state of the system at $t = 0$. 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

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

Now lets take another look at the semi classical Hamilton (2.4). Classically, the momentum operator $\hat{\mathbf{P}}$ is known as the canonical momentum and is 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 [3]:

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

Now we use this result to calculate the momentum at other times:

$$\mathbf{p}(t) = \mathbf{p}(t') + \mathbf{A}(t') - \mathbf{A}(t)$$

Now

$$\langle \Xi(t_c) | \Psi(t) \rangle = -i \int_{t_0}^t dt' e^{-i \int_{t'}^t \hat{H}(t'') dt''} e^{-i \int_{t_0}^{t'} \hat{H}_0(t'') dt''} \langle \Xi(t = t_c) | |\hat{\mathbf{d}} \cdot \mathbf{E}(t')| | \Psi(0) \rangle \quad (2.8)$$

This is the equation where most papers start with [1].

2.3 Strong Field Ionization

Phenomenology of strong field ionization.

2.4 Multiphoton Ionization

2.5 Transition matrix elements

The transition dipole matrix element is given by

$$\begin{aligned}
\mathbf{d}(\mathbf{p}) &= \langle \mathbf{p} | \hat{\mathbf{d}} | \Psi_{nlm} \rangle = \nabla_{\mathbf{p}} \Psi_{nlm}(\mathbf{p}) \\
\langle \mathbf{p} | 100 \rangle &= \frac{8\sqrt{\pi}}{\sqrt{\frac{1}{a^3}} (a^2 p^2 + 1)^2} \text{if } \Re\left(\frac{1}{a}\right) > 0 \\
\langle \mathbf{p} | 200 \rangle &= \frac{32\sqrt{2\pi} (4a^2 p^2 - 1)}{\sqrt{\frac{1}{a^3}} (4a^2 p^2 + 1)^3} \text{if } \Re\left(\frac{1}{a}\right) > 0 \\
\langle \mathbf{p} | 210 \rangle &= -\frac{128i\sqrt{2\pi}\sqrt{\frac{1}{a^3}} a^4 p}{(4a^2 p^2 + 1)^3} \text{if } \Re\left(\frac{1}{a}\right) > 0 \\
\langle \mathbf{p} | 300 \rangle &= \frac{72\sqrt{3\pi} (81a^4 p^4 - 30a^2 p^2 + 1)}{\sqrt{\frac{1}{a^3}} (9a^2 p^2 + 1)^4} \text{if } \Re\left(\frac{1}{a}\right) > 0 \\
\langle \mathbf{p} | 310 \rangle &= -\frac{864i\sqrt{2\pi}\sqrt{\frac{1}{a^3}} a^4 p (9a^2 p^2 - 1)}{(9a^2 p^2 + 1)^4} \\
\langle \mathbf{p} | 320 \rangle &= -\frac{1728\sqrt{6\pi}\sqrt{\frac{1}{a^3}} a^5 p^2}{(9a^2 p^2 + 1)^4} \\
\langle \mathbf{p} | 400 \rangle &= \frac{256\sqrt{\pi} (4096a^6 p^6 - 1792a^4 p^4 + 112a^2 p^2 - 1)}{\sqrt{\frac{1}{a^3}} (16a^2 p^2 + 1)^5} \text{if } \Re\left(\frac{1}{a}\right) > 0 \\
\langle \mathbf{p} | 410 \rangle &= -\frac{2048i\sqrt{\frac{\pi}{5}} \left(\frac{1}{a^3}\right)^{3/2} a^7 p (32a^2 p^2 (40a^2 p^2 - 7) + 5)}{(16a^2 p^2 + 1)^5} \text{if } \Re\left(\frac{1}{a}\right) > 0 \\
\langle \mathbf{p} | 420 \rangle &= -\frac{32768\sqrt{\pi}\sqrt{\frac{1}{a^3}} a^5 p^2 (16a^2 p^2 - 1)}{(16a^2 p^2 + 1)^5} \\
\langle \mathbf{p} | 430 \rangle &= \frac{262144i\sqrt{\frac{\pi}{5}}\sqrt{\frac{1}{a^3}} a^6 p^3}{(16a^2 p^2 + 1)^5} \\
\langle \mathbf{p} | 900 \rangle &= \frac{1944\sqrt{\pi} (27a^2 p^2 - 1) (243a^2 p^2 - 1) (243a^2 p^2 (6561a^4 p^4 - 729a^2 p^2 + 11) - 1) (729 (243a^6 p^6 - 99a^4 p^4 + 135a^2 p^2 - 1))}{\sqrt{\frac{1}{a^3}} (81a^2 p^2 + 1)^{10}} \\
\langle \mathbf{p} | 510 \rangle &= -\frac{4000i\sqrt{10\pi}\sqrt{\frac{1}{a^3}} a^4 p (15625a^6 p^6 - 3375a^4 p^4 + 135a^2 p^2 - 1)}{(25a^2 p^2 + 1)^6} \text{if } \Re\left(\frac{1}{a}\right) > 0
\end{aligned}$$

Different types of Ionization, tunneling Ionization, multiphoton

3. Ionization Model

3.1 GASFIR

test

3.2 Python Implementation

A general approximator for strong field ionization rates

3.3 TIPTOE

TIPTOE [4] 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

Difference between length gauge and velocity gauge in numerics

Why is it so difficult? Solving time dependent Schroedinger equation 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.

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

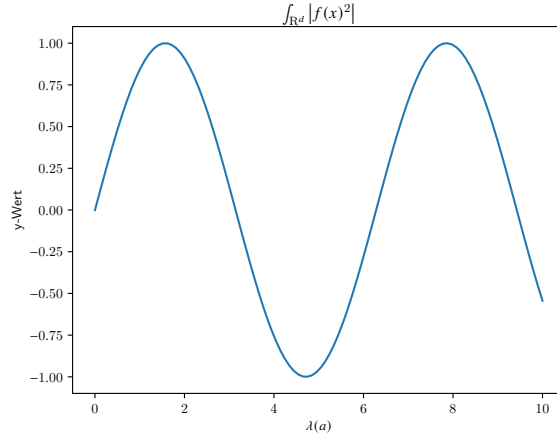


Figure 5.1: Sine function

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

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

$$\mathrm{i}\partial_t u = \mathcal{H}(t) \left| a \right\rangle \lambda \quad (5.3)$$

6. Conclusion and Outlook

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A. Appendix A

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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

Unterschrift