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**Tools for probing on short time and length scales:  
mid-infrared high harmonic generation  
and charged-particle guiding**

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# Kurzfassung

Wir behandeln zwei Phänomene welche beide heutzutage häufig als mögliche Werkzeuge für die Manipulation von geladenen Teilchen auf kleinen Skalen und in extrem kurzen Zeitspannen diskutiert werden: Auf dem Gebiet der ultraschnellen Laser-Atom Wechselwirkung beschäftigen wir uns im Speziellen mit der Abhängigkeit der Generierung von hoch-harmonischer Strahlung von der im mittleren Infrarot liegenden Wellenlänge des treibenden Lasers. Die Dynamik von Elektronen wird während der Wechselwirkung mit dem Laser von diesem auf einer atomarer Zeit- und Längenskala kontrolliert. Die dabei erzeugte Strahlung wird unter anderem zur Synthese von hoch-frequenten, kohärenten Pulsen verwendet, welche sich durch eine Dauer von oft weniger als eine Femtosekunde und einer räumlichen Auflösung im Mikrometer-Bereich auszeichnen. Wir betrachten die Abhängigkeit der emittierten Leistung von der Laser-Wellenlänge auf großen Skalen, wobei unsere Resultate sehr gut mit jüngsten Experimenten übereinstimmen. Auf kleinen Skalen, im Bereich von nur einigen Nanometer, finden sich überraschend regelmäßige Oszillationen in der emittierten Strahlungsleistung. Diese können durch die Interferenz von Elektronen, die zu verschiedenen Zeiten ionisiert werden und danach gemeinsam zur Strahlungsleistung des Atoms beitragen, erklärt werden. Die hier beobachteten Oszillationen sind eng mit der bereits etablierten Theorie über die Schließung von Ionisationsskanälen (“channel closing effect”) verbunden, welche ursprünglich für die Wechselwirkung von Systemen kurzreichweitiger Potentiale mit Dauerstrich-Lasern entwickelt wurde. Abweichungen von der Theorie des “channel closing effect” werden von uns als Einfluss des langreichweitigen Coulomb-Potentials eines realen Atoms erklärt.

Weiters untersuchen wir geführte Transmission von geladenen Teilchen durch nichtleitende Nanokapillaren. Dieser erst vor kurzem gefundene Effekt verspricht Präparierung und Ablenkung von Teilchen-Strahlen mit einem Durchmesser im Bereich von Mikrometern, ohne dabei auf konventionelle Teilchen-Optik zurückzugreifen. Wir präsentieren eine klassische Transporttheorie zur Beschreibung der geladener Teilchen in Nanokapillaren und betrachten dabei die Wechselwirkung von langsamen, hochgeladene Ionen im Energiebereich von einigen tausend Elektronenvolt (eV) und Elektronen mit Energien von einigen hundert eV mit der Oberfläche eines Nicht-Leiters im Inneren einer Nanokapillare. Wir vergleichen unsere Rechnungen mit den Ergebnissen der jüngsten Experimente, wobei qualitative und teils auch quantitative Übereinstimmung erzielt wurde.



# Abstract

We investigate two phenomena which are currently considered as possible tools in the quest of controlling matter on short time and length scales: firstly, we discuss high harmonic generation (HHG) during the interaction of ultrashort laser pulses with an atom. In laser-atom interaction, electronic motion may be steered on atomic time and length scales. The radiation simultaneously generated may be used for the synthesis of coherent, high-frequency pulses with a duration in the sub-femtosecond range as well as with a spatial resolution of the order of micrometers. We particularly explore the influence of tunable, mid-infrared driving lasers on the HHG yield. We report on the large-scale dependence of the HHG output of single atoms on the wavelength of the driver and find our results to be in good agreement with recent experiments. Further analysis of the dependence of the HHG yield on the wavelength of the driver has additionally revealed surprisingly regular variations on a fine scale of the order of several nanometers. The origin of these oscillations is identified as the path interference of several electron quantum paths contributing to HHG. The present observations obtained for ultrashort laser pulses are related to the well-known channel closing effect initially derived for the interaction of infinitely extended laser fields with systems governed by short-ranged potentials. Discrepancies to the channel closing picture are shown to be a consequence of the long-ranged Coulomb potential present in a real atom.

Guiding of charged projectiles through insulating nanocapillaries is the second phenomenon investigated. This effect is a promising means for the formation and deflection of microbeams of charged particles without any need for electromagnetic lenses and electrical feedthroughs. We present a Classical Transport Theory for guiding of both slow highly-charged ions with keV energies as well as electrons with energies of several hundred eV. For both types of projectiles we devise interaction scenarios with the internal capillary walls and apply our theory to describe recent experiments. Good qualitative, and, to some extent, also quantitative agreement is found.



# Contents

<b>Kurzfassung</b>	<b>iii</b>
<b>Abstract</b>	<b>v</b>
<b>1 Introduction</b>	<b>1</b>
<b>2 Review of laser-atom interaction</b>	<b>7</b>
2.1 Ultrashort laser pulses . . . . .	7
2.1.1 Gauge . . . . .	7
2.1.2 Magnetic field . . . . .	8
2.1.3 Dipole approximation . . . . .	8
2.1.4 Laser electric field . . . . .	9
2.1.5 Significance of laser vector potential . . . . .	10
2.2 SAE approximation . . . . .	11
2.3 Multi-photon vs. tunneling ionization . . . . .	12
2.4 Strong field approximation . . . . .	15
2.5 Classical picture of HHG . . . . .	16
2.6 Classical picture of above-threshold ionization . . . . .	17
2.7 Quantum Strong Field Approximation . . . . .	18
2.7.1 The Lewenstein model . . . . .	18
2.7.2 Saddle-Point analysis . . . . .	20
2.7.3 Shortcomings of the SPA . . . . .	22
2.7.4 Coulomb corrections to the SFA . . . . .	22
2.8 Spatial properties of harmonic radiation . . . . .	23
2.8.1 First-order propagation equation . . . . .	24
2.8.2 Gaussian beams and diffraction . . . . .	26
<b>3 Exact solution of the TDSE</b>	<b>29</b>
3.1 Time-dependent Schrödinger equation . . . . .	29
3.2 Split-operator method . . . . .	30
3.3 Pseudo-spectral method . . . . .	32

3.4	Grid dimensions and boundaries . . . . .	34
3.5	One-electron potentials . . . . .	35
3.5.1	Hydrogen . . . . .	35
3.5.2	Argon . . . . .	36
3.6	Power spectrum . . . . .	36
3.7	Photo-electron spectrum . . . . .	38
<b>4</b>	<b>Quantum path interference in HHG</b>	<b>39</b>
4.1	Global wavelength dependence of HHG . . . . .	39
4.2	Fine scale oscillations . . . . .	43
4.2.1	Stability with pulse duration . . . . .	45
4.2.2	Stability with pulse envelope . . . . .	45
4.3	Intensity dependence and channel closings . . . . .	46
4.3.1	Intensity dependence of yield oscillations . . . . .	48
4.3.2	Channel closings . . . . .	48
4.3.3	Modulation period of HHG yield . . . . .	51
4.3.4	Comparison of different atoms with different $I_p$ . . . . .	51
4.4	Coulomb corrections . . . . .	53
4.4.1	Truncated Coulomb potential . . . . .	53
4.4.2	Effective ionization potential . . . . .	54
4.4.3	Coulomb correction along classical trajectories . . . . .	55
4.5	Oscillations in the ionization yield . . . . .	59
4.5.1	Correspondence to HHG . . . . .	59
4.5.2	Quantum path interference in the photo-electron energy spectrum . . . . .	60
4.5.3	Ultrashort pulses: $\phi_{CEP}$ effects . . . . .	63
4.6	Global wavelength dependence of ionization . . . . .	64
<b>5</b>	<b>Guiding of Highly Charged Ions</b>	<b>67</b>
5.1	Charged particle transport in a nanocapillary . . . . .	67
5.1.1	Brief description of CTT simulation . . . . .	68
5.1.2	Wall interaction at impact . . . . .	69
5.1.3	Charge transport and screening . . . . .	69
5.2	Results for HCI Guiding . . . . .	71
5.2.1	Energy-dependence of HCI guiding . . . . .	71
5.2.2	Time-dependence of HCI guiding . . . . .	72
<b>6</b>	<b>Electron transport in nanocapillaries</b>	<b>79</b>
6.1	Electron-surface interaction . . . . .	79
6.1.1	Interaction scenario with the internal capillary wall . . . . .	79
6.1.2	Electron transport through amorphous materials . . . . .	80
6.1.3	Coherent surface reflection . . . . .	85

6.2	Instantaneous electron transmission . . . . .	88
6.2.1	Planar surface scattering as test case . . . . .	88
6.2.2	PET capillaries . . . . .	89
6.2.3	Al <sub>2</sub> O <sub>3</sub> capillaries . . . . .	90
6.3	Dynamics of electron transmission during charge-up . . . . .	93
<b>7</b>	<b>Summary and outlook</b>	<b>97</b>
	<b>Appendix</b>	<b>101</b>
<b>A</b>		<b>101</b>
A.1	Fourier transform on a bounded domain . . . . .	101
A.2	Definition of radiated power . . . . .	102
A.3	Definition of pulse intensity . . . . .	102
A.4	Far-field transformation . . . . .	103
<b>B</b>		<b>105</b>
B.1	Fitting and extrapolating optical data . . . . .	105
B.2	Fitting the surface-IMFP . . . . .	106
	<b>List of Acronyms</b>	<b>109</b>
	<b>Acknowledgements</b>	<b>123</b>



# Chapter 1

## Introduction

This thesis may be decomposed into two major parts: the first one covers the generation and fine-tuning of coherent, short-wavelength radiation during ultrafast laser-matter interaction. This process has paved the way for the synthesis of high-frequency radiation with a spatial resolution of the order of micrometers and pulse durations in the attosecond range. A recent review by Krausz and Ivanov [1] may serve as an overview of the young field of “attosecond science” which could recently be established with such pulses at hand. In the second part we present a theory as well as simulations of charged particle guiding through insulating nanocapillaries (i.e. with diameters in the nanometer regime). Seemingly different, both topics can be used as tools in the quest of controlling matter on small spatial scales (the buzzword “nanotechnology” is frequently used) and, in many cases, with high temporal resolution. For example, insulating nanocapillaries represent a possibly novel means of forming microbeams of charged particles without electromagnetic lenses and electrical feedthroughs. So far, steering of charged particles almost always relies on the application of conventional lenses. Present technology already reaches a high spatial precision. Commercially-available Focused Ion Beam (FIB) systems provide a lateral resolution of only a few nanometers (nm). Temporal resolution may be introduced by fast switching of an incident, continuous beam of charged particles. Typical methods are the application of mechanical beam choppers, chopping with high-frequency electric fields, or more elaborate chopping/bunching techniques. For example, today commercial time-of-flight secondary ion mass spectroscopy (TOF-SIMS) systems reach pulse durations of 1-10 nanoseconds (ns) for ionic beams. Conventionally generated electron bunches can be as short as several picoseconds.

Even shorter bunches of charged particles directly require an ultrashort, pulsed generation process. One example for such a process is laser-particle acceleration during the interaction of ultrastrong lasers operating in the highly relativistic regime (intensity well above  $10^{20}$  W/cm<sup>2</sup>) with matter [2]. Atoms are almost immediately ionized at such intensities. Electrons and ions with energies as large as several hundred MeV may be generated as a consequence. Bunching is automatically achieved because particle emission is bound

to the interaction time with the laser which is usually of the order of several tens of femtoseconds (fs). Laser-acceleration of electrons is frequently performed in capillaries filled with pre-ionized gases [3, 4], a method which increases the electron beam pointing stability as compared to experiments performed in an unconfined gas. Beam diameters initially correspond to the capillary diameter and beam divergences as low as 0.002 rad can be reached. In contrast to laser-electron acceleration, laser-ion acceleration is typically realized during the interaction with solid targets [5] and so far leads to much larger beam divergences of about ten degrees. More details on laser-particle acceleration are given in recent reviews, see e.g. [2].

In addition to laser-particle acceleration, the development of high-power laser systems has led to a large number of attractive novel research topics in the field of nonlinear optics. Historically, the key to large laser pulse intensities has been the sustained compression of pulse energy into shorter and shorter pulse durations. Commercial laser systems yield intensities well above  $10^{14}$  W/cm<sup>2</sup> and, more importantly, today these systems reach pulse durations as short as several femtoseconds. At such intensities and pulse durations atoms are usually not fully ionized. Interaction of strong laser pulses with atoms or molecules is governed by nonlinear processes, such as nonlinear polarization, multiphoton excitation, multiphoton ionization, and tunnel ionization. The latter two processes have been extensively studied in work on above-threshold ionization (ATI) (see e.g. Ref. [6] for a recent review). The major focus in contemporary ultrafast nonlinear optics is, however, the observation of time-dependent processes which cannot be fully characterized by time-integral - i.e. spectroscopical - measurements. Timing information of ultrafast dynamics can only be obtained by probing on at least the same time scales. Chemical processes can be time-resolved as well as controlled by infrared (IR) pulses with durations of 10-100 fs [7]. Time-resolved observation and control of vibrational processes in diatomic molecules can be performed with even shorter pulse durations of the order of several femtoseconds (see e.g. Ref. [8]). Here, the optical period, which is about 2.7 fs for an infrared-range wavelength of e.g. 800 nm, becomes comparable to the pulse duration. In this limit the phase of the light wave at the maximum of the pulse envelope - the so-called carrier-envelope phase (CEP) - is crucial for the induced dynamics. The latter is governed on a sub-cycle time scale by the exact form of the laser electric field. Laser-atom interaction thus represents a unique possibility to steer the electronic motion relative to the nucleus on atomic time and length scales.

Sub-cycle dynamics is of particular importance in high-harmonic generation (HHG). In this nonlinear interaction of a driving laser pulse (usually in the IR range) with matter electrons are ionized and accelerated away from their parent atom or molecule. Eventually they may recollide with the parent core when the laser electric field reverses its sign. In case of recombination the electron emits photons that carry its excess energy. This basic process takes place inherently on sub-cycle time scales and leads to energetic radiation bursts in the attosecond (as) range. Subsequent bursts with the periodicity of

*half* the driver’s optical cycle (the sign of the laser field is irrelevant) form a comb-like radiation spectrum. In the latter “harmonic” peaks located at odd integer multiples of the driver’s frequency are observed, giving rise to the term “high-harmonic generation”. Filtering the high-frequency part of a HHG spectrum enables the syntheses of ultrashort, coherent light pulses with energies in the extreme ultraviolet (XUV) range. Because of such unique light pulses HHG nowadays represents the workhorse of ultrafast optics. XUV pulse durations and, related to that, today accessible time scales approach about hundred attoseconds (as) [9]. Production of XUV pulses may be achieved with intensities that are large enough to induce nonlinear processes [10]. State-of-the-art applications of HHG in general and XUV pulses in particular involve time-resolved spectroscopy [11], time-resolved holography [12], imaging of a molecular orbital [13], and direct imaging of an infrared laser light wave (“attosecond streaking”) [14].

HHG is usually performed in a jet of dilute gas emerging from a nozzle. Some HHG schemes, however, rely on generation inside a gas-filled capillary in which the driving laser pulse and the thereby generated harmonics jointly propagate. Such a capillary has a diameter of the order of micrometers and serves as a waveguide for the infrared laser, leading to well-defined spatial propagation modes. These modes introduce important consequences for propagation: in an empty waveguide they propagate with a phase velocity larger than the speed of light. For the combined problem of propagation through a partly-ionized atomic gas these modes modify the overall dispersion of the driving laser. It has been demonstrated that e.g. the gas pressure in the capillary may be tuned such that the driving laser and a given harmonic frequency propagate with the same phase velocity. So-called phase-matched propagation is then realized: the HHG yield grows over the whole interaction distance. With a setup like this, large enhancements in the output can be realized [15], facilitating potential applications of the XUV radiation. In both HHG in a gas jet as well as inside a capillary, the high-frequency radiation emerges spatially highly collimated with a typical divergence of below 0.1 degrees (see Sec. 2.8). This offers good lateral resolution for probing which, more importantly, comes in combination with the time structure in the sub-fs range. A well-collimated coherent beam can be e.g. used for lenseless imaging of microscopic objects with a resolution close to the wavelength employed (diffraction limited) [16].

The capillaries employed in ultrafast laser-matter interaction are usually not smaller than several hundred micrometers in diameter. The scope of guiding of charged particles, however, nowadays reaches from capillaries of micrometer size [17] down to capillaries with diameters in the nanometer range [18]. The aspect ratios of all those devices are typically similar. Moreover, guiding of slow highly charged ions (HCI) with an energy in the keV range has also been demonstrated for macroscopically large, millimeter(mm)-sized, tapered capillaries spanning in their width millimeters to micrometers from entrance to exit [19]. Such devices may facilitate the formation of HCI microbeams [19, 20] with a size of  $\gtrsim 1 \mu\text{m}$ . For ions with somewhat larger incidence energy ( $\simeq \text{MeV}$ ) an application

in the irradiation of biological material has already been realized [21].

The field of guided transmission of charged particles has been initiated by Stolterfoht *et al.* who firstly observed guiding of slow, highly charged ions through polyethylene terephthalate (PET, “Mylar”) nanocapillaries [18]. Since the first observation a variety of systems with different geometry and/or different insulating materials have been investigated, such as  $\text{SiO}_2$  [22, 23],  $\text{Al}_2\text{O}_3$  [24, 25], and glass [17]. The qualitative picture observed seems to be similar in all cases and has led to the following scenario of HCI guiding: during the charge-up phase, HCI hit the internal capillary wall and deposit their charge. These charges are transported along the surface and into the bulk of the target material. Projectiles entering the capillary subsequently are deflected due to the Coulomb field of the deposited charges and may eventually be transmitted. Guiding (stable transmission conditions) sets in as soon as a dynamical equilibrium between projectiles hitting the capillary wall (charging) and transport into the bulk or to the capillary exits (discharging) has been established. This scheme could be recently confirmed by numerical simulations [26]. An even more recent development is the observation of guided transmission of electron beams through insulating  $\text{Al}_2\text{O}_3$  nanocapillaries [27]. The observation of a strong inelastic component of transmitted projectiles [28], however, points to a clearly distinct mechanism of guided transport. In this context a scenario different to HCI guiding has been devised and successfully applied to the description of the experiment [29].

Charged particle guiding is a complex process involving the interplay of a large number of charged particles with a solid. Established in a dynamical equilibrium between incident current, discharging, and transmission the total system does not show any timing on ultrafast (atomic) time scales. Instead, the time-dependence of system observables typically reflects the rather long discharge time constants of the insulating material, usually of the order of minutes or even hours. In sharp contrast to the intrinsic temporal resolution of laser-matter interaction, any time resolution on time scales shorter than the macroscopic (dis-) charging must come from the incident beam itself. Conventional methods of how time resolution may be introduced to a continuous beam of charged particles have been mentioned above. For the shortest bunches of charged particles currently available we have referred to relativistic laser-matter interaction [2]. Although a combination of charged particle bunching and guiding might be possible in the future, today collimation and/or focusing of charged particles to microbeams can be regarded as the most promising aspect of the present guiding effect. We emphasize that for the latter no electrical feedthroughs are needed and this can make such a tool attractive for technological applications.

## Outline and focus of this work

Controlling charged particles and photons on short time and length scales is the broad theme of this thesis, out of which selected aspects will be covered in detail. In the area of ultrafast laser-atom interaction and XUV pulse generation we will focus on the physics of high-harmonic generation needed as a prerequisite for the synthesis of ultrashort pulses. We will start with a review of basic laser-atom interactions and the approximations employed in this work (chapter 2). In this context we will also introduce the framework of macroscopic propagation of the driving laser in a gaseous medium and give estimates for typical spatial properties of the laser beam and the generated XUV radiation. Chapter 3 describes the exact solution of the non-relativistic interaction of the laser with a single atom, based on a numerical integration of the time-dependent Schrödinger equation (TDSE). Approximate methods will be frequently compared to the exact results in order to facilitate their interpretation.

In the following we will particularly focus on the opportunities of an experimentally novel range of driving laser wavelengths, i.e. the mid-infrared (chapter 4). So far, the vast majority of high-power driving lasers relies on a Ti:sapphire gain medium providing broadband, ultrashort pulses around a center wavelength of  $\lambda_c \approx 800$  nm. Recent activities in laser development intend to establish driving lasers at larger wavelengths, in particular around  $2 \mu\text{m}$  (e.g. [30]). Moreover, with the application of the (non-collinear) optical parametrical amplification [31], such novel laser systems will not only explore the mid-infrared range but will also offer a much larger tunability with respect to  $\lambda_c$ . In the context of HHG the idea of employing larger wavelength stems from increasing the maximum energy being radiated, which depends quadratically on  $\lambda_c$ . Employing a mid-infrared driver, the first generation of harmonics in the water-window (above  $\simeq 300$  eV) has already been reported [32]. A different aspect is the macroscopic propagation of the driving pulse and the generated harmonics. A recent analysis assuming optimized propagation conditions suggests mid-infrared drivers to be advantageous for XUV pulse generation [33]. Furthermore, phase-matching of the driver and a particular harmonic frequency is easier to obtain and thus more effective in the mid-infrared range [34]. Along those lines, fine-tuning of HHG by variation of the driver wavelength as well as the single-atom HHG efficiency for mid-infrared drivers become topics of major interest (see Refs. [35, 36, 37, 38, 39] for some recent work). In chapter 4 we therefore address both of these issues in detail. In particular, we demonstrate the importance of quantum path interference in the HHG process (see section 4.2). Interference is capable of introducing modulations of the HHG yield on a fine wavelength scale, i.e. on a scale of the order of only several nanometers. This offers the possibility of tuning the HHG output already in the framework of single atom response. Such a scheme is presumably easier to implement than (resonant) two-color driving schemes recently discussed [40, 41] and observed [42] which lead to a large HHG enhancement as well. Aside from the advantages of increased control over HHG efficiency, an investigation of the role of quantum path interference in

HHG gives more insight in their physical relevance. In fact, considering more than one quantum path becomes of large importance when employing mid-infrared driving lasers. In addition to the HHG yield we also investigate the ionization rate averaged over the pulse duration as a function of the mid-infrared driver's wavelength (section 4.5). This observable shows oscillations as well which are surprisingly similar to the ones in the HHG yield and can also be related to path interference.

In chapter 5 we focus on a discussion of the basic physics of guiding of highly charged ions through insulating nanocapillaries. We present a Classical Transport Theory developed for the description of the guiding effect [26]. This approach has served as an important contribution to establishing the present scenario of HCI guiding as a consequence of a self-organized charge-up. The model obtained is applied to PET nanocapillaries with a diameter of 200 nm. We show essential results and put an emphasis on the time-dependence of observables.

An extension of our Classical Transport Theory to the guided transmission of electrons is introduced in chapter 6. We extensively discuss the interaction of electrons with a surface and apply the framework developed to a description of electron transmission through nanocapillaries. Our approach is able to describe key findings of the experiment for both  $\text{Al}_2\text{O}_3$  and PET nanocapillaries [29]. The most important mechanisms to establish guided transmission of electrons are found to be elastic and inelastic scattering at the internal wall surface as well as at the bulk solid. Contrary to the case of HCI guiding, charge-up and electrostatic deflection of projectiles are only two out of several ingredients needed to establish electron guiding. Based on our model we predict guided transmission of slow electrons also for metallic capillaries.

We conclude our presentation in chapter 7 with a summary and an outlook on possible extensions of the present work.

## Units and general definitions

Unless otherwise stated, atomic units (a.u.) are used throughout this thesis. In atomic units, Planck's constant, the modulus of the charge and the mass of the electron and the radius of Bohr are  $\hbar = |e| = m_e = a_0 = 1$ . The speed of light is  $c \approx 137$ .

# Chapter 2

## Review of laser-atom interaction

In this chapter we review the framework in which strong-field laser-atom interactions are commonly described. Definitions of typical laser pulse shapes are given and the validity as well as limitations of the approximations employed in this work are discussed. For facilitating the interpretation of the exact solution of the TDSE simpler models of HHG and ATI based on (semi-) classical considerations are presented.

### 2.1 Ultrashort laser pulses

A laser pulse is a coherent light wave, i.e. it can be described unambiguously by a classical, oscillating electric and magnetic field vector with a defined amplitude and phase. In the following we want to define how such a pulse is described mathematically, with emphasis on pulses with extremely short duration.

#### 2.1.1 Gauge

A laser field propagating in free space can be described by its vector potential  $\tilde{\mathbf{A}}(\mathbf{r}, t)$  having the form of wave propagating in  $z$ -direction,

$$\tilde{\mathbf{A}}(\mathbf{r}, t) = \int d\omega \tilde{\mathbf{A}}(\mathbf{r}, \omega) \exp(i\omega t - ikz) . \quad (2.1)$$

Here,  $k = \omega/c$  and  $c$  denotes the phase velocity of the propagating laser field, being the speed of light.  $\tilde{\mathbf{A}}(\omega)$  are the frequency components at frequency  $\omega$  which can be obtained by Fourier transformation (see Eq. A.1).

The electric field  $\mathbf{F}(\mathbf{r}, t)$  is given by the vector potential *plus* a scalar potential  $\Phi_C(\mathbf{r}, t)$ ,

$$\mathbf{F}(\mathbf{r}, t) = -\frac{1}{c} \frac{d\tilde{\mathbf{A}}(\mathbf{r}, t)}{dt} - \nabla\Phi_C(\mathbf{r}, t) . \quad (2.2)$$

The potentials  $\tilde{\mathbf{A}}(\mathbf{r}, t)$  and  $\Phi_C(\mathbf{r}, t)$  are not unique. In fact, a physically identical situation can be described by differential potentials introduced by a gauge transform (see [43]), which offers an additional choice for restricting the functional form of the potentials. In the frequently used Coulomb gauge one requires the relation  $\nabla \cdot \tilde{\mathbf{A}}(\mathbf{r}, t) = 0$ , which, in turn, defines  $\Phi_C$  by the charge density  $\rho_C$  only:  $\nabla^2 \Phi_C = -4\pi\rho_C$ . In absence of free charges, which will be assumed henceforth,  $\Phi_C = 0$ . Within this work we choose a definition of the vector potential that absorbs the factor  $1/c$ ,  $\mathbf{A}(\mathbf{r}, t) = \tilde{\mathbf{A}}(\mathbf{r}, t)/c$ , leading to

$$F(\mathbf{r}, t) = -\frac{dA(\mathbf{r}, t)}{dt}. \quad (2.3)$$

Here and in the following we restrict our discussion to linearly polarized fields  $\mathbf{A}(\mathbf{r}, t) = A(\mathbf{r}, t)\hat{\mathbf{z}}$ , if not otherwise stated.

### 2.1.2 Magnetic field

The magnetic field of the laser light wave is given by the vector potential as

$$\mathbf{B}(\mathbf{r}, t) = \nabla \times \tilde{\mathbf{A}}(\mathbf{r}, t). \quad (2.4)$$

With the ansatz of a plane wave (Eq. 2.1), the latter expression can give an estimate for the magnitude of the magnetic field associated with the propagating laser field which reads  $|B| = |F|$ . The amplitude of both the magnetic and electric field is thus equal. However, as magnetic fields exert a force  $\propto (\mathbf{v} \times \mathbf{B})/c$  and  $v \approx 1$  a.u.  $\ll c$  for atoms in the ground state, the magnetic force is roughly smaller by a factor of  $c$  than the electric force  $F$ .

The motion due to the magnetic field is restricted to the propagation direction of the field and introduces a displacement of the electron relative to starting point. For HHG, even small displacements are crucial. A simple estimate reveals that magnetic forces become relevant in HHG only for intensities larger than  $10^{17}$  W/cm<sup>2</sup>. The range of intensities discussed in this work is far below this value. Magnetic forces will thus be neglected within this work.

### 2.1.3 Dipole approximation

As long as the lateral dimension of the system considered is much smaller than the wavelength  $\lambda = c/\omega$  of the laser field,  $F(\mathbf{r}, t)$  does not vary much spatially and, consequently, the  $r$ -dependence can be dropped. This is referred to as the dipole approximation. In the case of interaction of an IR laser ( $\lambda \approx 800$  nm) with an atom (dimension  $\approx 0.1$  nm) this approximation is by far fulfilled. In fact, it even holds for optical fields in the ultraviolet to extreme ultraviolet range ( $\lambda \approx 100 - 10$  nm). If the atom is strongly perturbed during an interaction with a laser, electrons can be driven far away from the nucleus. As

long as the largest excursion is of the order  $r_{max} < \lambda$ , the dipole approximation will still be applicable. The laser parameters treated in this work comply with these restrictions. Therefore, we will henceforth refer to the laser electric field by  $F(t)$  only.

### 2.1.4 Laser electric field

In case of a linearly polarized laser field the electric field  $F(t)$  can be written in dipole approximation as

$$F(t) = F_0 f(t) \cos(\omega_c t + \phi_{CEP}) . \quad (2.5)$$

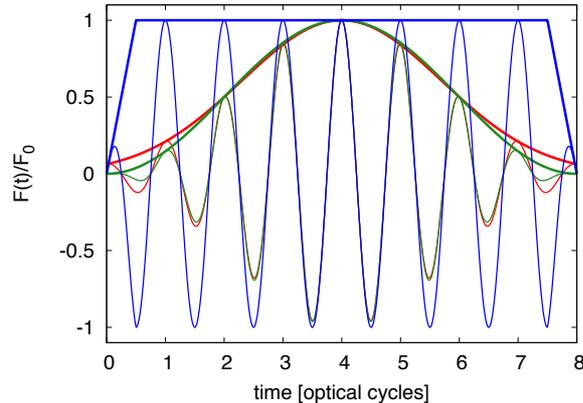
Here,  $F_0$  denotes the field amplitude,  $f(t)$  is the envelope function and  $\lambda_c = 2\pi c/\omega_c$  the central wavelength corresponding to the central frequency  $\omega_c$ . The expression above can be regarded as an ideal, infinitely long carrier wave with defined frequency  $\omega_c$ , modulated in time by the envelope  $f(t)$ .  $\phi_{CEP}$  denotes the carrier-envelope phase which becomes important only when the pulse duration is of the order of a few cycles of the field (“optical cycles”). Pulses with  $\phi_{CEP} = 0$  are frequently called cosine-like pulses while  $\phi_{CEP} = -\pi/2$  are called sine-like pulses. In this work, the field amplitude  $F_0$  is used to define the peak pulse intensity  $I_0$  by the expression  $F_0^2 = I_0$  (see appendix Sec. A.3 for more details on the definition of intensity). The conversion of intensity in SI-units ( $\text{W}/\text{cm}^2$ ) to the field amplitude in atomic units is  $F_0[a.u.] = 5.3380248 \cdot 10^{-9} \sqrt{I_0[\text{W}/\text{cm}^2]}$ . Without loss of generality we choose the polarization axis to be identical to the  $z$ -axis.

We define  $T$  to be the total length of the pulse while for practical purpose (e.g. in an experiment) the length given by the full width at half-maximum (FWHM) of the electric field or the intensity is more convenient. We choose to employ  $\tau_p$  as the FWHM of the electric field throughout this work. Commonly used pulse shapes (envelope functions) are:

- Gaussian pulse:  $f(t) = \exp(-4 \ln 2 t^2 / \tau_p^2)$
- $\sin^2$  pulse:  $f(t) = \sin^2\left(\frac{\pi t}{2\tau_p}\right)$
- Flat-top pulse with linear ramp on/off:  $f(t) = t/t_{ramp}$  for  $0 \leq t \leq t_{ramp}$ ,  $f(t) = (T - t)/t_{ramp}$  for  $T - t_{ramp} \leq t \leq T$ , and  $f(t) = 1$  otherwise.

all of which are defined for  $0 \leq t \leq T$  while  $f(t) = 0$  otherwise. Figure 2.1 shows the given pulse shapes for a FWHM of four optical cycles.

The finite duration introduced by the envelope functions leads to a change of the frequency spectrum (Fourier transform) of the pulse. Centered around  $\omega_c$  the spectrum becomes the broader the shorter the pulse duration is chosen. For a Gaussian pulse, the FWHM of the frequency components is given by  $\Delta\omega = 2.77/\tau_p$ , qualitatively similar relations hold for different envelope functions.



**Figure 2.1:** Laser electric field for different pulse shapes and a FWHM of four optical cycles. **Red:** Gaussian, **green:**  $\sin^2$ , and **blue:** Flat-top (cf. text).

In contemporary experiments, pulse durations down to the single cycle limit can already be reached for infrared (IR) pulses near a wavelength of 800 nm. Pulses can be phase-stabilized [44], and, indeed, dependence of physical observables such as e.g. the left/right information of ATI electron emission directions on the  $\phi_{CEP}$  has been measured [45].

### 2.1.5 Significance of laser vector potential

Maxwell's equations allow solutions propagating in free space only if

$$\int_{-\infty}^{+\infty} F(t)dt = 0 \quad \text{or} \quad (2.6)$$

$$\int_0^T F(t)dt = 0.$$

Equivalently, the electric field of the laser pulse can be solely described by its vector potential  $A(t)$  (Eq. 2.3). Equation 2.3 automatically fulfills the requirement of Eq. 2.6. For arbitrary ultrashort pulses with a duration of only a few (about 3 to 4) optical cycles, it is thus advantageous to define the vector potential rather than the electric field in order to avoid unphysical effects. However, for certain electric field pulse shapes such as a  $\sin^2$ -pulse with  $T$  being an entire number of optical cycles, Eq. 2.6 holds. This gives direct access to the electric field, which is the actual physical quantity. For longer pulses, Eq. 2.6 is fulfilled with sufficient accuracy in any case, therefore we usually directly fix the electric field  $F(t)$  unless otherwise stated. If  $F(t)$  is fixed, the vector potential can be calculated from

$$A(t) = - \int_t^T F(t')dt'. \quad (2.7)$$

A typical value for the vector potential is then  $A_0 = F_0/\omega_c$ . The requirement of Eq. 2.6 can be translated to  $A(0) = A(T) = 0$ . Even if  $A(0) = 0$  is not exactly guaranteed, ionized electrons experience by definition  $A(T) \equiv 0$  at the end of the pulse when physical observables are deduced. Unphysical behavior [46] is thus avoided.

From the point of view of ionization, the significance of the vector potential can be explained intuitively: If an electron is born outside a short-ranged potential at time  $t_i$  with initial momentum  $k_i$ , at the end of the pulse (at  $A(t = \infty) = 0$ ) it will acquire a momentum of  $k(t = \infty) = -A(t_i) + k_i$  according to the classical equations of motion. This (kinetic) momentum (or the energy  $E = k(t = \infty)^2/2$ ) is observable in the experiment.

For an electron initially at threshold (zero energy,  $k_i = 0$ ), the energy associated with the motion in the laser field is dubbed “ponderomotive energy”,  $U_p$ , reading

$$U_p = \frac{A_0^2}{4} = \frac{F_0^2}{4\omega_c^2}. \quad (2.8)$$

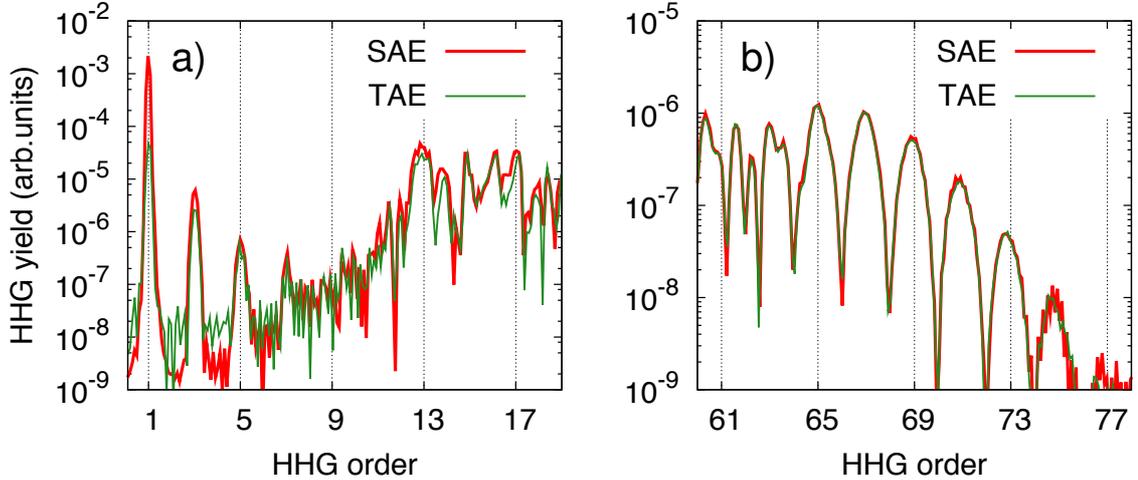
It corresponds to the averaged energy in an oscillating field, while the maximum energy is given by  $2U_p$ . A typical value for  $U_p$  at a wavelength of 800 nm and intensity  $I = 10^{14}$  W/cm<sup>2</sup> is  $U_p \approx 6$  eV.

## 2.2 SAE approximation

The most common system used to test the accuracy of calculations in theoretical atomic physics in general, as well as for laser-atom interactions in particular, is hydrogen. It has only a single electron, its wavefunction can be calculated analytically [47]. Furthermore, non-relativistic quantum theory is by far sufficient for describing the structure of the unperturbed atom. On the other hand, for practical work in experiments multi-electron atoms, i.e. rare-gas atoms like He, Ne, or Ar are usually used.

The ability to - at least qualitatively - describe laser-atom interaction of multi-electron atoms with theories based on only a single electron is explained by the energy structure of atoms. For example, the ground state ( $3p^6$ ) of argon has an energy of about -15.8 eV, while the next-lower state, the 3s-state is already bound with an energy of roughly 28.8 eV. Laser photon energies treated in this work range from 0.62 eV (wavelength 2000 nm) to 1.55 eV (1000 nm). For relatively weak laser fields, ionization of core electrons thus requires at least nine additional photons, rendering such a process highly unlikely. In strong laser fields the ionization probability even depends exponentially on the binding energy, which makes the dynamics of the weakest bound electron by far more important than the dynamics of the core electrons. A simple estimate based on the analytic quasi-static tunneling ionization rate of hydrogen (see Eq. 2.10 in section 2.3) at an intensity of  $I = 10^{14}$  W/cm<sup>2</sup> yields a rate of  $7 \cdot 10^{-5}$  for a ground state energy of -15.8 eV, while it is already as low as  $3 \cdot 10^{-13}$  for a state being bound deeper by 13 eV. Although these numbers just represent rough estimates it is clear that the dynamics is practically governed

by the outermost electron only. A more detailed comparison of the SAE approximation to exact dynamics is given by Fig. 2.2. Here, the SAE model potential employed was



**Figure 2.2:** Comparison of a HHG spectrum from helium in the SAE approximation with the exact solution obtained by a full solution of the two-electron TDSE (cf. text), courtesy of J. Feist *et al.* [48]. a) close-up near the central wavelength of the driver, b) close-up near the cut-off.

given by Tong [49] and the intensity and wavelength is  $5 \times 10^{14}$  W/cm<sup>2</sup> and  $\lambda_c = 750$  nm, respectively. The full solution of the two-electron TDSE for helium in the ground state shows excellent agreement with a corresponding simulation in the SAE approximation, in particular near the cut-off of the spectrum (Fig. 2.2 b). Amplitude, position, and shape of the harmonics at the cut-off are well reproduced within the SAE approximation. Only at low energies, the latter is somewhat inaccurate. Here, all the electrons contribute to the total polarizability of the atom. The largest discrepancy is found at the frequency of the driver itself where the linear polarizability of the multi-electron atom can not always be reproduced reliably. In the intermediate part of the HHG spectrum both, for energies larger than the ionization potential, calculations agree well with each other (not shown).

### 2.3 Multi-photon vs. tunneling ionization

When laser light hits an atom in its ground state with energy  $-I_p$  it will transfer energy, primarily to the electronic system. In the framework of the SAE-approximation (Sec. 2.2) it is sufficient to restrict the discussion to the outermost electron, the active electron. The laser can be described by its electric field (Eq. 2.5). The essential parameters are the central frequency  $\omega_c$ , which is the mean energy of the photons associated with this

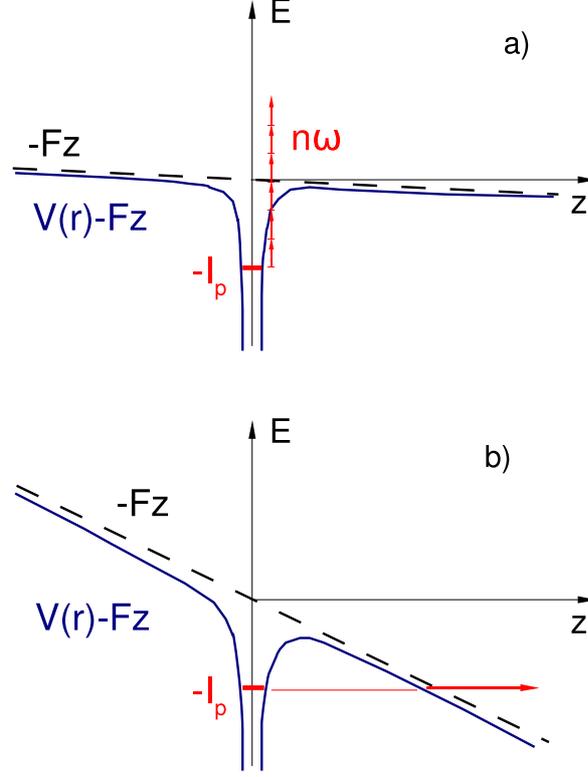
field, and the field strength  $F_0$ . In quantum mechanics the interaction with the atomic system is described by the Hamiltonian  $H(t) = H_0 + \hat{r}F(t)$ , where  $H_0$  is the Hamiltonian of the unperturbed system (without laser). If the perturbation  $\hat{r}F(t)$  is weak, it can be treated in the framework of perturbation theory [47]. One can easily show that first order perturbation theory corresponds to the absorption of a *single* photon from the field, the occupation of the target state is  $\propto F_0^2$  or  $\propto I$ . Perturbation theory of the  $n^{\text{th}}$ -order corresponds to the absorption of  $n$  photons, being  $\propto F_0^{2n}$  or  $\propto I^n$ . Processes with  $n > 1$  are not linear with respect to intensity (and with respect to the perturbation) and are usually dubbed nonlinear processes. Due to this power law, such so-called  $n$ -photon processes are only relevant for large field strengths which are yet easily accessible by modern lasers. The first nonlinear process driven by a low frequency laser was already demonstrated in 1961 by Franken *et al.* [50], today even coherent XUV radiation can be intense enough to induce nonlinear processes [10].

If absorption of one or more photons transfers enough energy to reach the ionization threshold, one speaks of *ionization*. Figure 2.3 a) sketches such a multi-photon ionization process. Ionization by one or more photons is a genuine quantum process. Classically, it can not be understood. The potential in which the electron is moving is perturbed only very weakly and on a fast time scale  $1/\omega_c \gtrsim t_k$  associated to the laser light. Here, we have introduced  $t_k$  being the (Kepler) time scale on which the classical electron revolves around the nucleus.

For field strengths that are large enough, the perturbed potential can allow the electron to either ionize “classically allowed” over-the-barrier (*barrier suppression ionization*) or semi-classically tunnel through the lowered barrier (*tunnel ionization*). A cartoon of the process of tunnel ionization is given in Fig. 2.3 b). For such a process to be viewed (semi-)classically it is moreover necessary that the perturbed potential changes only slowly on the time scale of the initial (ground) state. This condition is easy to fulfill at long wavelengths, e.g. in the infrared where the optical cycle  $T_{cyc} = 2\pi/\omega_c$  is much longer than  $t_k$ . A popular choice to set a limit for such a semi-classical, quasi-static ionization region is the Keldysh parameter  $\gamma$ , reading

$$\gamma = \sqrt{\frac{I_p}{2U_p}}. \quad (2.9)$$

$\gamma < 1$  signifies the tunneling regime. The “limit” between the two regimes is given by  $\gamma = 1$  which can be found around  $I = 1.15 \cdot 10^{14}$  W/cm<sup>2</sup> for the case of hydrogen and radiation wavelengths of about 800 nm. Note that this limit is somehow artificial. Signatures of multi-photon processes can be found for  $\gamma < 1$ , while tunneling ionization already describes the ionization process for  $\gamma > 1$  well. In fact, it can be shown that the expressions for describing a multiphoton ionization process as well as for tunneling ionization in an alternating field can be both deduced from the same starting point when taking either in the limit of  $\gamma \gg 1$  or  $\gamma \ll 1$  [51]. A popular interpretation of the Keldysh



**Figure 2.3:** Multi-photon (Fig. a) ) and tunnel ionization (Fig. b) ). Tunnel ionization takes place on a time scale much smaller than the optical period (quasi-static), the photon-character is only retained in case of multi-photon ionization.

parameter is that it compares the time the electron spends under the barrier to the optical cycle. For  $\gamma \ll 1$  tunneling is much faster than the optical period, allowing for a quasi-static treatment. On the other hand, ionization is extended in time over many optical cycles for  $\gamma \gg 1$ , underscoring a photon-like interpretation.

Quasi-static tunneling can be easily incorporated into simple, classical models once approximate expressions relating the electric field to the tunnel rate are found. Two frequently used tunneling formulas that are also employed in this work are described in the following.

For hydrogenic atoms in the ground state (effective nuclear charge of  $Z_{eff}$ ) and an ionization potential (negative binding energy) of  $I_p = Z_{eff}^2/2$ , an analytic formula for the quasi-static tunneling has been found [52]

$$\Gamma_H = \frac{4Z_{eff}^5}{|F(t)|} \exp\left(-\frac{2Z_{eff}^3}{3|F(t)|}\right) = \frac{4(2I_p)^{5/2}}{|F(t)|} \exp\left(-\frac{2(2I_p)^{3/2}}{3|F(t)|}\right). \quad (2.10)$$

A generalized theory applicable to arbitrary atoms in the general state with quantum numbers  $(n, l, m)$  has been developed by Ammosov, Delone, and Krainov [53] and is therefore dubbed ADK theory. We discuss only atoms with the magnetic quantum number  $m = 0$  because ionization for  $m \pm 1$  is much lower for laser fields linearly polarized along the axis defined by  $m = 0$ . ADK theory defines effective quantum numbers in addition to  $(n, l, m)$ . Firstly,  $n^* = Z_r / \sqrt{2I_p}$  with  $Z_r = 1$  being the charge of the atomic residue for single ionization. For example, argon ( $Z = 18$ , ground state  $3p^6$ ) has  $I_p = 0.58$ , hence  $n^* = 0.9285 \approx 1$ . Furthermore,  $l^* = n^* - 1 \approx 0$ . The ADK tunneling rate  $\Gamma_{ADK}$  is then given by

$$\Gamma_{ADK} = \frac{2^{2n^*} (2l + 1)}{n^* \Gamma(n^* + l^* + 1) \Gamma(n^* - l^*)} I_p \left( \frac{2(2I_p)^{3/2}}{|F(t)|} \right)^{2n^* - 1} \exp \left( -\frac{2(2I_p)^{3/2}}{3|F(t)|} \right). \quad (2.11)$$

Here,  $\Gamma(z) = (z - 1)!$  is the gamma function. In the limit of  $n^* \rightarrow 1$  and  $l^* \rightarrow 0$  the above expression reduces to the hydrogenic case (Eq. 2.10).

## 2.4 Strong field approximation

Let us once more review typical energies involved in laser-atom interaction: as long as the electron is still bound its motion is strongly governed by the atomic core potential with its ground state energy given by  $E_0 = -I_p$ . On the other hand, after an ionization process the dynamics of the free electron is given by the electric field, in which the average energy is the ponderomotive potential  $U_p$ , and by the long-range part of the atomic core potential. Provided that  $U_p \gtrsim I_p$ , the influence of the latter after ionization can be neglected in first approximation. Furthermore, during the free motion of the electron in the laser field the latter can be considered to be dominant if absorption of an additional photon  $\omega_c$  does not change much, hence one requires  $U_p \gtrsim I_p \gg \omega_c$ .

This simplified picture is commonly used in laser-atom interactions and referred to as *strong field approximation* (SFA). Disregarding the atomic potential after ionization makes an analytical treatment of the electronic motion possible, both in a classical and - together with some other approximations - also in a quantum mechanical approach (see Sec. 2.7 below). Hence, these assumptions greatly facilitate the discussion of the dynamics and additionally provide important guidelines near the limit of their validity ( $U_p \approx I_p$ ). For the case of an hydrogen atom subjected to a laser pulse in the IR range (near 800 nm) the intensity needs to be larger than  $2 \times 10^{14}$  W/cm<sup>2</sup> in order to fulfill  $U_p \gtrsim I_p$  and thus justify the application of the strong field approximation. The condition  $I_p \gg \omega_c$  clearly holds in the IR range.

## 2.5 Classical picture of HHG

Along the lines of the discussion in Sec. 2.4 a (semi-)classical model of HHG can be introduced. We analyze the motion of a free electron with coordinate  $z$  in a linearly polarized laser field with electric field  $F(t)$  and vector potential  $A(t)$  being released from the nucleus (assumed to be in the origin) at time  $t_i$ . This ionization process can be due to tunneling and thus represents a quantum process needed as a prerequisite of a classical theory.

The equation of motion and its solution with the initial condition  $v(t_i) = 0$  are

$$\ddot{z}(t) = -F(t) = \dot{A}(t) \text{ and } v(t_i) = 0 \quad (2.12)$$

$$\rightarrow \dot{z}(t_f) = v(t_f) = A(t_f) - A(t_i) \quad (2.13)$$

The velocity  $v(t) = \dot{z}(t)$  can also be defined via  $k + A(t) = v(t)$ , where  $k$  is the time-independent canonical momentum, being an integral of motion. Note that for the initial condition chosen  $k = -A(t_i)$ .

The amplitude of the classical motion (“quiver” amplitude  $\alpha$ ) is given by  $\alpha = A_0/\omega = F_0/\omega^2$ . For strong fields, the amplitude is much larger than the dimension of the atom in the ground state,  $\alpha \gg 1$ . On that scale, the position of an electron directly after ionization from the ground state is the position of the nucleus,  $z(t_i) \approx 0$ . Corrections from the picture of tunneling ionization (“tunnel exit”) are small.

HHG takes place when the electron recollides with the nucleus, returning to its initial state by emission of “coherent bremsstrahlung”. In the vicinity of this point recombination and the emission of photons become possible. Consequently, for the excursion  $z$  we require that

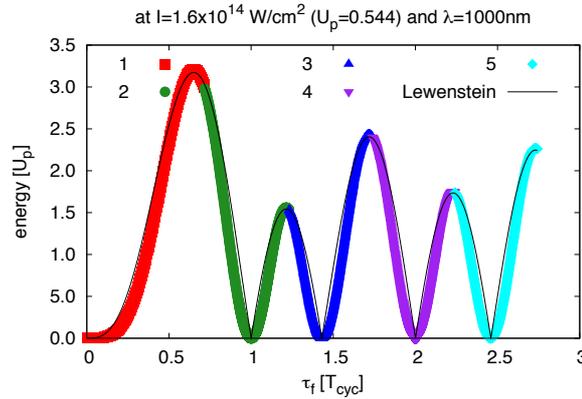
$$z(t_f) - z(t_i) = 0 = \int_{t_i}^{t_f} A(t') dt' - A(t_i) \cdot (t_f - t_i). \quad (2.14)$$

This is the recollision condition for an electron on a classical trajectory in the laser field. For a given  $t_f$  (or  $t_i$ ) it is a transcendental equation for  $t_i$  (or  $t_f$ ), which can be solved only numerically or graphically. Note that the equation may have more than one solution. The energy of the recolliding electron is given by

$$E_{\text{recoll}} = \frac{(A(t_i(t_f)) - A(t_f))^2}{2} \quad (2.15)$$

where  $t_i(t_f)$  is determined by the recollision condition Eq. 2.14. The largest energy possible can be found by maximizing Eq. 2.15 under this constraint. Its value is given by  $3.17U_p$ , which is realized by a trajectory with  $\tau_f/T_{\text{cyc}} = 0.65$  (cf. 2.4).

The basic physics can thus be summarized in the so-called three step model [55], sketched in Fig. 2.5. In the first step (red), an atom is ionized via tunneling ionization. The freed electron is subsequently accelerated in the laser field and may rescatter with the parent ionic core (green). During recombination to the ground state a high-energy



**Figure 2.4:** Recollision energy (Eq. 2.15) as function of flight time  $\tau_f = t_f - t_i$ . The color symbols show the numerical solutions based on a 8 cycle flat-top pulse (1<sup>st</sup> to 5<sup>th</sup> solution for a given recollision time), while the solid line shows the result of Lewenstein *et al.* [54] obtained for an ideal, sinusoidal field.

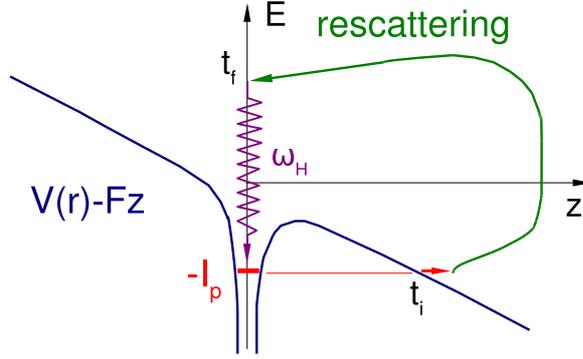
photon is emitted (blue). Due to the periodicity of the process with half the laser period, harmonic peaks with a spacing of twice the laser frequency are generated. In this model the maximum energy (“cut-off” energy) of the radiation is the recollision energy of the electron plus the ionization potential,

$$E_c = I_p + 3.17U_p. \quad (2.16)$$

For each energy  $\omega_H$  below  $E_c$ , two trajectories are possible with flight time  $\tau_f < T_{cyc}$ . In every subsequent half-cycle again two solutions of Eq. 2.14, hence two trajectories, exist. Their recollision energies are lower than those of the shortest two trajectories, the largest energy possible approaches  $2U_p$  for  $\tau_f \rightarrow \infty$ . This is illustrated by Fig. 2.4 showing the first five solutions of Eq. 2.14 (characterized by  $E_{recoll}$ ) as a function of the flight time  $\tau_f$  for the case of a flat-top laser pulse.

## 2.6 Classical picture of above-threshold ionization

The ionization process, which corresponds to the first two steps of HHG, can be investigated in the framework of classical physics as well. For this discussion we neglect the possibility of subsequent recombination at the parent ion. Monte Carlo methods based on classical trajectories launched after (non-classical) tunneling (e.g. [56]) may indeed well describe numerous aspects of strong field photo-ionization, such as photo-electron momentum distributions. However, determining classical trajectories subject to both the potential of the atom as well as the laser field requires numerical solutions of the equations



**Figure 2.5:** Cartoon of the three-step model of HHG [55]. In the first step (red), an atom is ionized via tunneling ionization. The freed electron is subsequently accelerated in the laser field and may rescatter with the parent ionic core (green). During recombination to the ground state a high-energy photon with energy  $\omega_H$  is emitted (blue).

of motion. For very strong laser fields, where in the spirit of the SFA the atomic potential can be neglected from the moment of ionization on, calculations and interpretation of the dynamics are more straightforward. As already outlined in Sec. 2.1.5 electrons will be accelerated in the laser field and, provided no rescattering at the nucleus takes place, their final momentum is then given by the vector potential at the time of ionization  $A(t_i)$  and the initial momentum  $k_i$  only,  $k(t = \infty) = -A(t_i) + k_i$ . For tunneling ionization  $k_i \approx 0$ . Hence, the largest energy acquired by the electrons values  $2U_p$ . Note that similar to HHG the periodicity of the process, here with period of  $T_{cyc}$ , introduces peaks with a spacing of  $\omega_c$  in the photo-electron spectrum. In the case of ultrashort, near-single-cycle laser pulses this periodicity is weakened and peaks will be broadened (Fourier reciprocity).

## 2.7 Quantum Strong Field Approximation

### 2.7.1 The Lewenstein model

Based on the idea of the strong field approximation (SFA) outlined in Sec. 2.4, Lewenstein *et al.* have derived quantum models for both HHG [54] and ATI [57] that extend the classical picture. In the following we will briefly review the quantum SFA theory.

We consider an hydrogen-like atom in its ground state, characterized by the ground state energy  $-I_p$  (effective nuclear charge  $\sqrt{2I_p}$ ). According to [54], three approximations are essential for a quantum SFA theory of HHG:

i) we assume that the dynamics of the bound electron is given by the ground state only

and thus neglect any excited bound states. The initial (ground) state is assumed to be unchanged by the laser field during time evolution.

ii) along the lines of the SFA we neglect the potential of nucleus in the final state (after ionization) completely, leaving only a free electron in a laser field to be described.

iii) furthermore, ground state depletion is neglected.

With these assumptions the amplitude  $b(\vec{k}, t_f)$  of the final state  $|k + A(t_f)\rangle$  (a Volkov state) can be written as

$$b(\vec{k}, t_f) = i \int_0^{t_f} dt_i \exp^{-iS_V(\vec{k}, t_i, t_f) + iI_p t_i} F(t_i) D(\vec{k} + \vec{A}(t_i)). \quad (2.17)$$

$D(\vec{k})$  is the dipole matrix element with respect to the hydrogenic ground state (Eq. 2.18). Being a matrix element between a plane wave  $\langle \vec{k} |$  and the ground state  $|0\rangle$  this expression neglects the effect of the Coulomb potential on the electron released. The dipole matrix element reads

$$D(\vec{k}) = \langle \vec{k} | z | 0 \rangle = -i \partial_{k_z} \langle \vec{k} | 0 \rangle = i \frac{2^{7/2} (2I_p)^{5/2}}{\pi} \frac{\vec{k}}{(2I_p + k^2)^3}. \quad (2.18)$$

Very frequently, in such an approach this matrix element is also employed for an arbitrary atom. Within this work we will also make use of this approximation if not otherwise stated. Finally, the phase acquired by the time evolution of a free electron in an electric field (Eq. 2.17) is given by the Volkov phase,

$$S_V(\vec{k}, t_1, t_2) = \frac{1}{2} \int_{t_1}^{t_2} (\vec{k} + \vec{A}(t'))^2 dt'. \quad (2.19)$$

High-harmonic generation results from the coherent interaction of a previously ionized continuum wavepacket with a bound state (usually the ground state) of an atom. Key quantity is the dipole moment  $d = -\langle \hat{z} \rangle$ . It can be written as [54]

$$d(t_f) = \int d^3k \langle 0 | z | k + A(t_f) \rangle b(k, t_f) + c.c. = \quad (2.20)$$

$$2\mathcal{R} \int d^3k \int_0^{t_f} dt_i e^{-iS_V(\vec{k}, t_i, t_f) - iI_p(t_f - t_i)} D^*(\vec{k} + \vec{A}(t_f)) F(t_i) D(\vec{k} + \vec{A}(t_i)). \quad (2.21)$$

To arrive at this result we have neglected dipole transitions within the continuum and, in general, we have assumed that the SFA (disregarding the atomic potential after ionization) is applicable. Ground state depletion, which has been neglected in the present derivation, can be in fact accounted for at a later stage, as we will see below.

### 2.7.2 Saddle-Point analysis

Solving the integral over  $\vec{k}$  in Eq. 2.21 in three dimensions may be cumbersome. In order to further simplify and gain more insight into the problem, the Stationary Phase Approximation (SPA) is commonly applied [54]. In this approximation it is assumed that due to the rapid oscillations of the phase  $S_V + I_p(t_f - t_i)$ , the integral over  $\vec{k}$  has its strongest contributions near its stationary points given by  $\nabla_{\vec{k}} S_V$ , around which the phase is expanded up to the second order in  $\vec{k}$ . All other terms under the integral, on the other hand, are assumed to vary only slowly with respect to  $\vec{k}$ .

Consequently, for linearly polarized fields  $\vec{A} = A\hat{z}$  the stationary phase condition selects the return momenta  $k_P(t_i, t_f)$ , which are characterized by the release and rescattering times  $(t_i, t_f)$ , i.e.

$$k_P(t_i, t_f) = -\frac{1}{t_f - t_i} \int_{t_i}^{t_f} A(t') dt'. \quad (2.22)$$

Eq. 2.22 is equivalent to the recollision condition for a classical, free electron (Eq. 2.14). Interestingly, the SPA recovers essentially classical trajectories as the most important contribution to HHG. The Volkov phase along this quantum path is denoted by  $S_P(t_i, t_f) = S_V(k_P, t_i, t_f)$ . For  $d$ , we arrive at

$$d(t_f) = 2\mathcal{R} \int_0^{t_f} dt_i e^{-iS_P(t_i, t_f) - iI_p(t_f - t_i)} D^*(\vec{k} + \vec{A}(t_f)) F(t_i) D(\vec{k} + \vec{A}(t_i)). \quad (2.23)$$

Ivanov *et al.* have shown that the integral over time can be itself tackled in the framework of the SPA, leaving for each release time  $t_i$  a finite sum over possible trajectories (or “path”  $P$ ) [58]. Furthermore, they show how to break down Eq. 2.23 explicitly into the product of three amplitudes, describing the ionization ( $a_{\text{ion}}(t_i)$ ), the free evolution ( $a_{\text{prop}}(t_i, t_f)$ ), and the recombination ( $a_{\text{rec}}(t_f)$ ), respectively. This directly reflects the classical three-step model. The expression for  $a_{\text{ion}}(t_i)$  that would follow from Eq. 2.23 is substituted by an ionization amplitude accounting for ground state depletion and the ionization rate is given by quasi-static tunneling. This is advantageous because ionization rates based on tunneling formulas like Eq. 2.10 reproduce full numerical calculations much better than SFA ionization amplitude (see Eq. 2.17). In summary, the expressions for the dipole moment  $d$  read

$$d(t_f) = \sum_{P(t_i)} \frac{1}{\sqrt{i}} a_{\text{ion}}(t_i) \cdot a_{\text{prop}}(t_i, t_f) \cdot a_{\text{rec}}(t_f) + c.c. \quad (2.24)$$

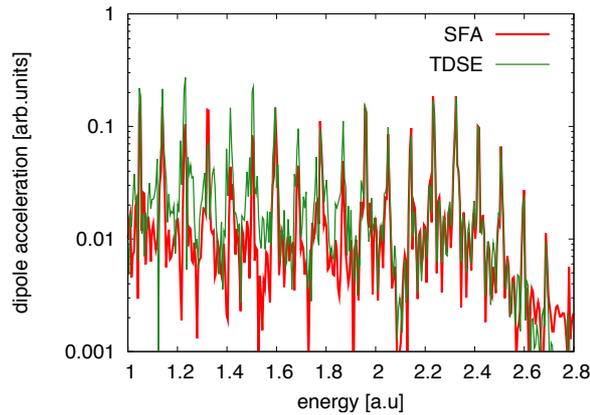
$$a_{\text{ion}}(t_i) = \sqrt{\frac{dn(t_i)}{dt}} \quad (2.25)$$

$$a_{\text{prop}}(t_i, t_f) = \left( \frac{2\pi}{t_f - t_i} \right)^{3/2} \frac{(2I_p)^{1/4}}{|F(t_i)|} \exp(-iS_P(t_i, t_f) - iI_p(t_f - t_i)) \quad (2.26)$$

$$a_{\text{rec}}(t_f) = D^*(k(t_f, t_i) + A(t_f)) \quad (2.27)$$

The electron flight time is given here by  $\tau_f = t_f - t_i$ . In Eq. 2.25  $n(t)$  represents the ground state occupation probability being calculated from the quasi-static tunneling rate  $\Gamma$  as  $n(t) = n(t = 0) \cdot (1 - \exp(-\int_0^t \Gamma(F(t'))dt')) \approx \int_0^t \Gamma(F(t'))dt'$ . This expression accounts for ground state depletion. By Fourier transforming  $d(t)$ , the HHG spectrum in the framework of SFA can be obtained.

This model works remarkably well for large parts of the HHG radiation spectrum, i.e. from the plateau region to the cut-off, as illustrated by Fig. 2.6. Position and amplitude



**Figure 2.6:** Comparison of a HHG spectrum obtained by a SFA model [58] and an *ab-initio* integration of the TDSE.  $\lambda_c$  is 1000 nm and the intensity is  $1.6 \times 10^{14}$  W/cm<sup>2</sup>. In this regime the SFA describes the high-frequency response sufficiently well. Note that the SFA data has been multiplied by a constant factor for comparison.

of the cut-off are well reproduced, and such is the overall structure of the plateau below the cut-off. The HHG spectrum of this model fails for energies lower than the ionization potential. In this region bound states govern the electron dynamics and thus the HHG spectrum. The yield based on SFA calculations, where bound states are neglected, is observed to decrease dramatically. We will consider, however, only harmonics with energies larger than  $I_p$ . The model for HHG described in Eq. 2.24 can therefore be safely applied. In the following, exactly this framework is employed whenever we are referring to an SFA calculation of HHG. We emphasize once more that the present approach relates electronic quantum paths to corresponding classical trajectories via the application of the SPA. The phase  $S_P(t_i, t_f)$  picked up along those trajectories (or paths) is equivalent to a phase introduced in semi-classics, therefore we frequently refer to the present SPA-based model as a “semi-classical” approach. Note that one important benefit of the SPA as compared to the “exact” expression in Eq. 2.21 is that the individual contributions of the respective path  $P$  to the dipole moment can be analyzed by taking only the summand  $d_j(t) = d(t)|[P = P_j]$ .

### 2.7.3 Shortcomings of the SPA

The above description suffers from the following drawback: When applying the SPA to the time and momentum integral (Eq. 2.23), the stationary points in time are given by

$$\partial_{t_i}(S_P(t_i, t_f) + I_p(t_f - t_i)) = 0 \quad (2.28)$$

$$\rightarrow \frac{1}{2}(k + A(t_i))^2 + I_p = 0. \quad (2.29)$$

The latter equation has no real solution as  $I_p > 0$ . For  $I_p = 0$ , Eq. 2.29 delivers a constraint for the velocity at release time  $k + A(t_i) = v(t_i) = 0$ . In the strong field limit  $S_V \approx U_p \gg I_p$ , this result holds approximatively. Hence, the considerable success of the SPA in the description of HHG experiments is based on the condition  $U_p \gg I_p$ .

In the case of large values of  $I_p$ , however, Eq. 2.29 can be solved allowing for complex values of  $t_i$  only. The imaginary part is usually interpreted as the tunneling time (the time the electron spends in the classically forbidden region below the potential). Generally,  $\text{Im}(t_f - t_i) \ll \text{Re}(t_f - t_i)$ , it is therefore only a small correction becoming negligible in the strong field case.

### 2.7.4 Coulomb corrections to the SFA

In the quantum SFA model for HHG as well as ionization the final expressions lack any influence of the Coulomb potential. Impact on accuracy and possible by-passes of the problem depend on which of the three steps of HHG are considered.

#### Ionization

The ionization amplitude in SFA is correct only in the exponential dependence on the field strength but inaccurate the prefactor. Ionization rates predicted are therefore too low. A remedy to the problem is to substitute  $F(t) \cdot D(k + A(t))$  by an amplitude deduced from a tunnel ionization rate  $\sqrt{\Gamma(F(t))}$  based on the full atomic potential, in our work for example Eq. 2.10. (see e.g. [59] for a discussion). In fact, we employ exactly this approach also in the model for HHG (Eq. 2.25).

#### Recombination

Recombination is governed by the matrix element at the time of recombination  $t_f$ ,  $D(k + A(t_f))$ . The correct matrix element is actually not given by a plane wave  $|k\rangle$  in the initial (i.e. pre-recombination) state but by a Coulomb function  $|El\rangle$  with energy  $E$ . This is not considered in this work. However, the approximative plane wave matrix element is accurate enough for the large energies considered in the plateau and cut-off regions of the harmonic spectra.

### Free propagation

The Coulomb potential is also acting during the free propagation of the ionized electron in the laser field. In the framework of an eikonal-approximation a correction due to the presence of the potential is accounted for only with exponential accuracy, i.e. as an additional term in the phase,

$$S_{EI}(t) = \int_0^t V_{EI}(r(t')) dt'. \quad (2.30)$$

Here it is assumed that the trajectory  $r(t)$  remains unchanged by the action of the Coulomb force. This correction will be applied in Sec. 4.4 for an improved comparison to the exact solution of the laser-atom interaction.

## 2.8 Spatial properties of harmonic radiation

Conventionally, HHG is performed in a dilute gas of atoms, typically in form of a  $\mu\text{m}$ - to mm-sized gas jet emerging from a nozzle. The medium is not opaque to the infrared driving laser which thus propagates in the gas. The laser focus can be considered to be situated in the center of the gas jet for simplicity of the discussion. Deviations from this position are, however, frequently used in the experiment to improve the phase-matched generation of an individual harmonic.

As long as ionization in the medium by the driving laser stays low propagation of the driver is only influenced by the linear refractive index and its corrections to the next order. On the other hand, with strong ionization a large density of free electrons - a plasma - is created. We introduce the electron plasma frequency  $\omega_p^2 = 4\pi n_e$ . Here,  $n_e = P_{ion} n_a$  with  $n_e$  and  $n_a$  being the number density of the free electrons and atoms, respectively. The polarization introduced by free electrons of constant density can be described by Drude's model [43] as

$$P_{el}(\omega) = \chi_{el,D}(\omega) F(\omega) = -\frac{n_e F(\omega)}{\omega^2}. \quad (2.31)$$

Light waves with frequencies below the plasma frequency  $\omega_p$  can not propagate in the plasma and are reflected. For infrared lasers, this limits the electron density to  $n_e \approx 2.6 \cdot 10^{-4} = 1.7 \cdot 10^{27} \text{ m}^{-3}$  which corresponds at a 100% level of ionization ( $n_e = n_a$ ) and room temperature to a gas pressure of 7 MPa. Naturally, experiments stay far below this value and are performed in the range of several thousand Pa. We assume  $\omega_p \ll \omega$  and thus disregard reflected waves completely.

The central question of interest is, however, the propagation properties of harmonic radiation generated. As their generation process is phase-locked with the specific electric field of the driving laser we have to consider the joint propagation of driver and harmonics.

We will first start by recalling quantities well known from linear optics. The complex refractive index  $\tilde{n}$  for a non-magnetic medium ( $\mu = 1$ ) is

$$\tilde{n}(\omega) = \sqrt{\epsilon(\omega)} = \sqrt{1 + 4\pi\chi(\omega)} \quad (2.32)$$

with

$$\chi(\omega) = \frac{P(\omega)}{F(\omega)} = \frac{P_a(\omega) + P(\omega)_{lin}}{F(\omega)} = \chi(\omega)_a + \chi(\omega)_{lin}. \quad (2.33)$$

Here, we have split the polarization of the medium  $P(\omega)$  into the sum of a part,  $P(\omega)_{lin} = \chi(\omega)_{lin}F(\omega)$  linear in the field  $F(\omega)$ , and a part resulting from the (possibly nonlinear) atomic response,  $P_a(\omega)$ . In case of laser propagation and HHG inside a capillary of “hollow waveguide” an additional term linear in  $F(\omega)$  would account for the dispersion introduced by the waveguide. We restrict our discussion to propagation in an unconfined gas and omit this term in the following. The polarization of the atoms can be rewritten in terms of their dipole moment  $P_a(\omega) = n_a d(\omega, F)$ . When employing the expansion of the square-root,  $\sqrt{1+x} \approx 1 + x/2$ , we arrive for  $\tilde{n}(\omega)$  at

$$\tilde{n}(\omega) \approx 1 + 2\pi \frac{n_a d(\omega, F)}{F(\omega)} + 2\pi\chi(\omega)_{lin}. \quad (2.34)$$

$d(\omega, F)$  is the Fourier-transform of the the dipole moment  $d(t, F)$  depending on the electric field  $F$ . Note that a number density  $n_a$  corresponds to a pressure  $p$  via  $p = n_a kT$ . Henceforth we are always assuming  $T = 300K$  when giving pressures instead of particle densities.

The term  $d(\omega, F)$  incorporates the - possibly - nonlinear response of the neutral atoms. In  $\chi(\omega)_{lin}$ , on the other hand, all linear propagation effects defined by  $P(\omega) = \chi(\omega)F(\omega)$  can be included. For example, neutral atoms contribute to  $\chi(\omega)_{lin} \propto n_a(1 - P_{ion})$ . In particular, effects introduced by free electrons may be incorporated in  $\chi(\omega)_{lin}$  (see Eq. 2.31).

### 2.8.1 First-order propagation equation

The propagation of high-intensity pulses in matter is treated by Maxwell’s equations in combination with a model for the polarization response of the medium  $\mathbf{P}(\mathbf{x}, t')$ .

As long as the electric field  $\mathbf{F}(\mathbf{x}, t')$  is linearly polarized we can assume that for an atomic gas the polarization  $\mathbf{P}(\mathbf{x}, t')$  will also be linearly polarized. Having  $\mathbf{F}(\mathbf{x}, t) = F(x_3, t)\mathbf{e}_\perp$  and  $\mathbf{P}(\mathbf{x}, t) = P(x_3, t)\mathbf{e}_\perp$  we can formulate a scalar wave equation [60] which reads after a Fourier transform to frequency space

$$\left( \partial_{x_3}^2 + \Delta_\perp + \frac{\omega^2}{c^2} \right) F(\mathbf{x}, \omega) = -\frac{4\pi\omega^2}{c^2} P(\mathbf{x}, \omega) \quad (2.35)$$

where  $\partial_{x_3}$  refers to the partial derivatives with respect to  $x_3$  and  $\Delta_{\perp}$  stands for the transverse components of the Laplace operator.

We now substitute the ansatz [61]

$$F(x_3, \omega) = U(x_3, \omega)e^{-ix_3\omega/v} \quad (2.36)$$

into Eq. 2.35, where the phase velocity  $v$  values  $c/n_m$ . The ansatz of Eq. 2.36 splits up wave-like solutions into a fast-varying part and a *slowly-varying* complex envelope. Neglecting the second-order spatial derivative of  $U(x_3, t)$  and back-substitution gives

$$\partial_{x_3}F(\mathbf{x}, \omega) = -i\frac{v}{2}\left(\frac{\omega}{c^2} + \frac{\omega}{v^2}\right)F(\mathbf{x}, \omega) - i\frac{v}{2\omega}\Delta_{\perp}F(\mathbf{x}, \omega) - i\frac{2\pi\omega v}{c^2}P(\mathbf{x}, \omega). \quad (2.37)$$

This is a first-order propagation equation which can be solved numerically with much less effort than the full scalar wave equation (Eq. 2.35) that is of second order in  $x_3$ .

The above approximation is applicable as long as changes of the electric field induced by the polarization of the medium over a distance comparable to the central wavelength  $\lambda_c$  are small. It is equivalent to the elimination of backward propagating wave solutions [60] because the ansatz of Eq. 2.36 with a *slowly* varying envelope  $U(\mathbf{x}, \omega)$  only allows for propagation in one direction. Using a first-order propagation equation is in particular valid for under-dense plasmas [62], where  $\omega_p^2 \ll \omega^2$ . This is easy to understand as electromagnetic waves with frequencies  $\omega < \omega_p$  can not propagate in a plasma with an electron density larger than  $n_e$ . In such a case the wave will be reflected or damped inside the plasma.

Coming back to the propagation equation, Eq. 2.37, we now make a coordinate transformation to a coordinate frame moving with velocity  $v = c/n_m$ ,  $\tau = t - x_3/v$ ,  $x_3 = x_3$ . This results in frequency space in the substitution  $f(\omega) \rightarrow f(\omega)e^{-i\omega/vx_3}$ . Hence,

$$\partial_{x_3}F(\mathbf{x}, \omega) = -i\frac{v}{2}\left(\frac{\omega}{c^2} - \frac{\omega}{v^2}\right)F(\mathbf{x}, \omega) - i\frac{v}{2\omega}\Delta_{\perp}F(\mathbf{x}, \omega) - i\frac{2\pi\omega v}{c^2}P(\mathbf{x}, \omega). \quad (2.38)$$

Choosing  $v = c$  as velocity of the moving coordinate system we arrive from Eq. 2.38 at

$$\partial_{x_3}F(\mathbf{x}, \omega) = -i\frac{c}{2\omega}\Delta_{\perp}F(\mathbf{x}, \omega) - i\frac{2\pi\omega}{c}(\chi(\omega)_{lin} \cdot F(\mathbf{x}, \omega) + P_a(\mathbf{x}, \omega)). \quad (2.39)$$

### Solving the propagation equation

In order to solve the propagation equation (Eq. 2.39) we employ the split-step method [63], which is a standard method for solving nonlinear, partial differential equations. At each frequency  $\omega$ , we decompose the equation into two simpler ones:

$$\partial_{x_3}F(x_3, x_{\perp}) = (\hat{D} + \hat{N})F(x_3, x_{\perp}) \rightarrow \quad (2.40)$$

$$\partial_{x_3}F'(x_3, x_{\perp}) = \hat{N}F'(x_3, x_{\perp}) \quad (2.41)$$

$$\partial_{x_3}F''(x_3, x_{\perp}) = \hat{D}F''(x_3, x_{\perp}) \quad (2.42)$$

and solve both independently for a stepsize  $\Delta x_3$ . If Eq. 2.41 is solved the solution serves as initial values for Eq. 2.42.

Under the assumption that  $P_a(x_3, x_\perp) = P_a(F(x_3, x_\perp)) \approx P_a(0, x_\perp)$  within a small step  $\Delta x_3$  the “non-linear” Eq. 2.41 is solved within the interval  $[0, x_3]$  by

$$F'(\Delta x_3, x_\perp, \omega) = \left( F(0, x_\perp, \omega) + \frac{P_a(0, x_\perp, \omega)}{\chi(\omega)_{lin}} \right) e^{-i2\pi\chi_{lin}\frac{\omega}{c}\Delta x_3} - \frac{P_a(0, x_\perp, \omega)}{\chi(\omega)_{lin}}. \quad (2.43)$$

On the other hand, Eq. 2.42 reads in full

$$\partial_{x_3} F''(x_3, x_\perp, \omega) = -i\frac{c}{2\omega}\Delta_\perp F''(x_3, x_\perp, \omega) \quad (2.44)$$

The initial values of the partial differential equation in Eq. 2.44 are the solutions  $F'(\Delta x_3, x_\perp, \omega)$  from Eq. 2.43. Various methods for solving for  $F''(x_3, x_\perp, \omega)$  exist in literature. We employ the implicit Crank-Nicolson scheme [63] that is stable for all choices of step sizes  $\Delta x_3$  and  $\Delta x_\perp$ .

## 2.8.2 Gaussian beams and diffraction

In case of propagation in vacuum ( $P_a = 0$  and  $\chi_{lin} = 0$ ) one solution of the propagation equation are the so-called Gaussian beams. They can be written as

$$F(x_3, x_\perp, t) = F(0, 0, t) \times \frac{w_0}{w(x_3)} \exp\left(-\frac{x_\perp^2}{w^2(x_3)}\right) \times \exp\left(-ikx_3 - i\frac{kx_\perp^2}{2R(x_3)} + i\Phi_{guoy}\right) \quad (2.45)$$

with  $k = \omega/c$  and the so-called confocal parameter  $z_0 = \pi w_0^2/\lambda_c$ . We have also defined  $w^2(x_3) = w_0^2(1 + x_3^2/z_0^2)$  as well as the Guoy phase

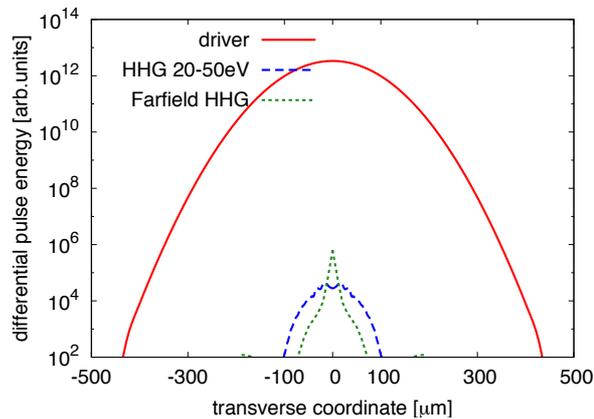
$$\Phi_{guoy} = \tan^{-1}(x_3/z_0) \quad (2.46)$$

which introduces an overall phase shift of  $\pi$  between the incident and the outgoing phase-front of the beam.

Interpretation of Eq. 2.45 is straightforward: Apart from a curved wave front with curvature  $R(x_3) = (x_3^2 + z_0^2)/x_3$  the beam resembles a plane wave and is radially of Gaussian shape with a width  $w(x_3)$  varying over the propagation coordinate. The width is zero in the origin (the focus). The beam spread depends on the wavelength and is controlled by the parameter  $z_0 = \pi w_0^2/\lambda$ . This represents the diffraction of the light wave. The angular divergence due to diffraction can be written as

$$\tan \theta_{diff} = \frac{w_0}{z_0} = \frac{\lambda}{\pi w_0}. \quad (2.47)$$

$\lambda$  is smaller for the high harmonics generated than for the driving laser leading to a much smaller diffraction. Additionally, the harmonics are created in a region  $w_{0,H} \ll w_0$  due to the strong dependence of the generation process on the local intensity  $\propto \int dt F(r, z, t)^2$ . For HHG in a loose-focusing geometry (see Fig. 2.7) this leads to an angular divergence for the harmonics as small as  $\theta_{diff} \approx 0.01^\circ - 0.03^\circ$  in the far-field (see Sec. A.4). When



**Figure 2.7:** Spatial distribution of pulse energy for the driver as well as the generated harmonics (20-50 eV) after propagation through 1 mm of hydrogen in a loose-focusing geometry ( $w_0 = 120 \mu\text{m}$ ). For the harmonics the far-field (Eq. A.13) was computed in a distance of  $d = 10$  mm from the medium. Laser parameters are  $\lambda_c = 1000$  nm and  $I_0 = 1.6 \times 10^{14}$  W/cm<sup>2</sup>, the dipole moment was computed in the framework of the SFA.

applying the HHG beam to a target in a distance of  $d = 10$  mm from the generation medium we therefore have a spatial resolution being of the order of several tens of  $\mu\text{m}$ . Moreover, this fine spatial resolution has to be viewed in combination with the ultrafast timing on a femtosecond scale.



# Chapter 3

## Exact solution of the TDSE

### 3.1 Time-dependent Schrödinger equation

In chapter 2 we have introduced the concept of the dipole approximation and have discussed that only the laser electric field is relevant for laser-matter interactions at the intensities considered in this work. Furthermore, in Sec. 2.2 it was shown that for processes like HHG and ATI, single-active electron (SAE) calculations are sufficiently accurate. This allows us to develop a fully quantum mechanical description by employing a Hamiltonian that is rather simple but includes all relevant effects.

One has the freedom of choice in which gauge the Hamiltonian and thus the time-dependent Schrödinger equation (TDSE) is formulated. According to a discussion in [64], the velocity gauge generally fits the dynamical problem of an atom in a laser field best and converges better at high laser intensities. In velocity gauge, the TDSE is written in spatial coordinates of the electron  $\mathbf{r}$  and the coordinates of the center of the atom  $\mathbf{x}$  as

$$i \frac{\partial}{\partial t'} \Psi_V(\mathbf{r}, \mathbf{x}, t') = \left[ \frac{1}{2} (-i \nabla_r + \mathbf{A}(\mathbf{x}, t'))^2 + V_{eff}(r) \right] \Psi_V(\mathbf{r}, \mathbf{x}, t') \quad (3.1)$$

Note that the dependence on  $\mathbf{x}$  comes only on a macroscopic scale; on the scale of an atom  $\mathbf{A}(t')$  depends only on time (dipole approximation). For  $V_{eff}(r) = 0$  stationary states of the system are the Volkov states which are spatially identical to plane waves (see Sec. 2.4). Such free (ionized) electrons are well described by their canonical momentum  $\mathbf{k}$  which does not change during the interaction with the laser field. Time-evolution from  $t_i$  to  $t_f$  introduces a time-dependent phase only, i.e. the Volkov phase (Eq. 2.19). Only the velocity  $v(t) = k + A(t)$  reflects the time-dependence of the field.

In length gauge, however, the TDSE reads

$$i \frac{\partial}{\partial t'} \Psi(\mathbf{r}, \mathbf{x}, t') = \left[ -\frac{1}{2} \Delta_r + V_{eff}(r) + \mathbf{r} \cdot \mathbf{F}(\mathbf{x}, t') \right] \Psi(\mathbf{r}, \mathbf{x}, t') \quad (3.2)$$

Note that the wave function in velocity gauge  $\Psi_V(\mathbf{r})$  is in general different from the wave function in length gauge, while physical observables (expectation values) are naturally independent of gauge. The wave functions can be transformed into each other by the unitary gauge transform  $\Psi_V(\mathbf{r}) = \exp(-i\mathbf{A}(\mathbf{0}, \mathbf{t}) \cdot \mathbf{r})\Psi(\mathbf{r})$  [64]. Contrary to the case of free electrons, the length gauge is the appropriate choice for describing bound states in the field. This is obvious by observing that the gauge transform given above would introduce a time-dependent oscillatory phase factor usually not present for bound states.

In the range of laser parameters ( $I_0 < 5 \cdot 10^{14}$  W/cm<sup>2</sup>) we are interested in, where at least the atomic ground state somehow retains its unperturbed character, the time-dependent Schrödinger equation in length gauge can be solved with sufficient accuracy. We employ an algorithm developed originally by X.-M. Tong and S.-I. Chu, which will be described in the following.

To begin with, we change to a coordinate frame moving with velocity  $c$ ,  $t = t' - x_3/c$ . We rewrite Eq. 3.2:

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}, x_3, t) = \left[ -\frac{1}{2} \Delta_r + V_{eff}(r) + r \cos \theta \cdot F(x_3, t) \right] \Psi(\mathbf{r}, x_3, t) \quad (3.3)$$

The coordinate system of the electron is chosen in such a way that the  $z$ -axis of the coordinate system of the electron (coordinate  $z = r \cos \theta$ ) is aligned with the linearly polarized electric field. The  $x_3$ -coordinate (atomic position) enters this equation only via the input field  $F(x_3, t)$ . On the spatial scale of an atom, this  $x_3$ -dependence is dropped (dipole approximation, see Sec. 2.1.3). It is important only when describing the propagation of the electric field within a macroscopically extended medium, hence when changing the atomic positions on a length scale of several wavelengths. In the following derivations, the  $x_3$ -dependence is dropped in order to improve readability.

## 3.2 Split-operator method

For calculating the time evolution of the wavefunction, the Hamilton operator is split into a static term and a term dependent on  $t$ :

$$\hat{H}(t) = -\frac{1}{2} \Delta_r + V_{eff}(r) + r \cos \theta \cdot F(t) = \hat{H}_0 + F(t)r \cos \theta \quad (3.4)$$

In order to solve the TDSE one needs to evaluate

$$\Psi(\mathbf{r}, t) = \hat{U}(t)\psi(\mathbf{r}, 0) = \hat{T} \exp \left( -i \int_0^t \left( \hat{H}_0 + F(t')r \cos \theta \right) dt' \right) \Psi(\mathbf{r}, 0) \quad (3.5)$$

with a time-ordered exponential  $\hat{T} \exp(\dots)$ . A (conceptionally) straightforward way to solve the TDSE is to expand the above exponential into a perturbation theory series of

interdependent integrals (Dyson series)

$$\hat{U}(t) = 1 - i \int_0^t dt' \hat{H}(t') - \frac{1}{2} \int_0^t dt' \int_0^{t'} dt'' \hat{H}(t') \hat{H}(t'') + \dots \quad (3.6)$$

If the time-dependent field perturbing the system is weak, the first order perturbation may be enough to describe the response of the system, as it is the case for single-photon processes. For strong fields, however, the convergence of the above time evolution operator  $\hat{U}(t)$  may be slow. Direct numeric evaluation of the  $m$ -th term with  $n_t$  time steps results in a numerical effort proportional to  $(n_t/2)^m$ .

A more efficient method for the time propagation of the wavefunction is the split-operator method. This method has already been introduced by Feit and Fleck in 1988 [65]. Time propagation of the wavefunction is achieved by evaluating Eq. 3.5 repeatedly for time steps  $\Delta t$  small enough for the integrand  $\hat{H}(t)$  to be approximated as being constant. This is the case when  $\Delta t < t_k = 2\pi n^3$  where  $t_k$  is the (Kepler) orbital period of a Rydberg electron in the state with energy  $E_{nl}$ . Of course,  $\Delta t$  must additionally be small enough to resolve possible small-scale variations of the driving field  $F(t)$ .

Not only is the time evolution  $U(t)$  divided into many small time steps  $\Delta t$  with  $U(t) \approx U(t_f, \Delta t) \cdots U(t_1, t)U(t_0, \Delta t)$  which needs to be done in any discretization scheme. In particular, one time step  $U(t_i, \Delta t)$  is split into [66]

$$U(t_i, \Delta t) \approx U_0(\Delta t/2) B(t_i, \Delta t) U_0(\Delta t/2), \quad (3.7)$$

where

$$U_0(\Delta t/2) = e^{-i\hat{H}_0\Delta t/2} \quad (3.8)$$

describes the unperturbed electron propagation, i.e. without the laser-atom interaction. The effect of the laser-atom coupling is taken into account by the boost operator

$$B(t_i, \Delta t) = e^{i\Delta p(t_i, \Delta t)\hat{z}} \quad \Delta p(t_i, \Delta t) = \int_{t_i - \Delta t/2}^{t_i + \Delta t/2} F(t') dt' \simeq \Delta t F(t_i), \quad (3.9)$$

which transfers the momentum  $\Delta p$  (a kick) to the electron.

The separation in Eq. 3.7 allows to calculate the first step of the time evolution  $U_0(\Delta t/2)$  in energy representation because the time evolution of the basis functions  $\Phi_{nl}(\mathbf{r})$  with defined energy  $E_{nl}$ ,

$$\hat{H}_0\Phi_{nl} = E_{nl}\Phi_{nl}, \quad (3.10)$$

is then a simple multiplication of a phase factor  $\exp(-iE_n\Delta t/2)$ . We denote the time-independent basis by  $\Phi_{nl}(\mathbf{r})$  while the time-dependent coefficient of the total wavefunction is  $\Psi_{nl}(t)$ . Consequently,  $\Psi(\mathbf{r}, t) = \sum_{n,l} \Psi_{nl}(t)\Phi_{nl}(\mathbf{r})$ .

On the other hand, the boost operator  $B(t_i, \Delta t)$  is efficiently evaluated in coordinate-space, where the representation of the wavefunction  $\Psi(\mathbf{r}, t)$  is diagonal. Again, the evaluation of the operator involves only a multiplication by a phase factor.

In summary, the total time evolution for a time step  $\Delta t$  is performed by propagating the wavefunction  $\Psi_{nl}(t)$  for a half time step  $\Delta t/2$  in energy-space:

$$\Psi_{nl}(t + \Delta t/2) = e^{-i\hat{H}_0\Delta t/2} \Psi_{nl}(t) \quad (3.11)$$

Then the wavefunction  $\Psi_{nl}(t + \Delta t/2)$  is transformed to coordinate-space  $\Psi^-(\mathbf{r}, t + \Delta t/2)$  and propagated for a time step  $\Delta t$  under the influence of the atom-field coupling alone:

$$\Psi^+(\mathbf{r}, t + \Delta t/2) = e^{i\Delta p(t+\Delta t/2, \Delta t)\hat{z}} \Psi^-(\mathbf{r}, t + \Delta t/2) \quad (3.12)$$

Finally, the wavefunction after the application of the boost operator  $\Psi^+(\mathbf{r}, t + \Delta t/2)$  is transformed back again to energy-space and propagated for another half time step  $\Delta t/2$  equivalent to Eq. 3.11,

$$\Psi_{nl}(t + \Delta t) = e^{-i\hat{H}_0\Delta t/2} \Psi_{nl}^+(t + \Delta t/2). \quad (3.13)$$

The total expression of the form

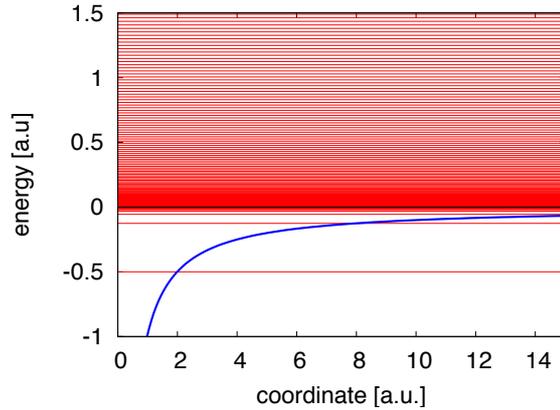
$$\Psi_{nl}(t + \Delta t) \approx U_0(\Delta t/2) B(\Delta t) U_0(\Delta t/2) \Psi_{nl} + O(\Delta t^3) \quad (3.14)$$

gives rise to an error in the third power of  $\Delta t$  per time step [65]. More sophisticated split-operator methods as discussed e.g. in Ref. [67] show an error of  $O(\Delta t^4)$ , but we found this third-order method to be sufficient as the step size in time is limited by requiring a good resolution in  $F(t)$  as well.

A sufficiently small time step for typical laser parameters ( $\lambda_c \approx 1000$  nm,  $I = 10^{14}$  W/cm<sup>2</sup>) is  $\Delta t \approx 0.25$  a.u.. Such a time step is almost one order of magnitude larger than the time step necessary for simpler numerical time evolution schemes with a different choice of  $U(t_i, \Delta t)$ . The ‘‘leap frog’’ algorithm which needs time steps of  $\approx 0.01$  a.u. may serve as one example. The split-operator method is thus very efficient for a long time propagation of a wavefunction, provided the problem to be solved is linear as in the present case of the atomic TDSE. To calculate HHG by a femtosecond laser pulse then leads to several thousands time steps ( $n_t \approx 10^3 - 10^4$ ). The numerical effort involved scales then as  $n_t n_{max}^2 \cdot l_{max}$  where  $n_{max}$ ,  $l_{max}$  are the largest quantum numbers  $n$ ,  $l$  taken into account.

### 3.3 Pseudo-spectral method

The indices  $nl$  of the wavefunction in energy representation  $\Psi_{nl}(t)$  (Eq. 3.10) can only take discrete values corresponding to the discrete energy-eigenvalues  $E_{nl}$  for  $E < 0$ . In order to describe the continuum in energy above  $E = 0$ , the latter is approximated by dense lying discrete states  $\Psi_{nl}$ . This approach is called the pseudo-spectral method for representing a continuum. The discrete states emerge when an infinite potential (hard



**Figure 3.1:** Potential (blue) and energy levels (red) for one-dimensional hydrogen with a box-size (hard-wall boundary) of  $r_{max} = 200$  a.u. The continuum is decomposed into a finite number of discrete states, being sufficiently dense for low energies.

wall) is introduced at  $r_{max}$  as indicated in Fig. 3.1. For large energies where the influence of the potential  $V_{eff}(r)$  is low, energy-eigenvalues can be approximated by a hard-walled well of infinite height and width  $r_{max}$ ,

$$E_{nl} = \frac{1}{2} \left( \frac{\pi n}{r_{max}} \right)^2 \quad (3.15)$$

$$\Delta E_{nl} = n \left( \frac{\pi}{r_{max}} \right)^2 \quad (3.16)$$

Hence, the density of states of the discretized “continuum” states is controlled by the parameter  $r_{max}$ . How dense the states need to be in order to represent the continuum depends on the laser field strength and frequency.

Employing no a-priori assumptions on  $V_{eff}(r)$  and  $r_{max}$ , the energies  $E_{nl}$  must be computed numerically. Symmetries of the Hamiltonian facilitate the calculation: The eigenfunctions  $\Phi_{nl} = \Phi_{nl}(r, \cos \theta, \phi) = \Phi_{nl}(r, \cos \theta)$  are independent of  $\phi$  for the Hamiltonian chosen (Eq. 3.3), due to the dipole approximation and the restriction to linearly polarized electric fields. Employing standard procedures [47], the  $\theta$ -dependence can be separated with the aid of the spherical harmonics  $Y_l(\cos \theta)$ , reading  $\Phi_{nl}(r, \cos \theta) = \phi_{nl}(r)/r Y_l(\cos \theta)$ . The radial eigenfunctions  $\phi_{nl}(r)$  together with their eigenvalues  $E_{nl}$  are calculated numerically on an appropriate radial mesh  $r_i \in [0, r_{max}]$ . For an efficient numerical treatment of a Coulomb-like potential  $V_{eff}(r)$ , which is diverging for  $r \rightarrow 0$ , a nonlinear mesh-point distribution is advantageous. Far away from the nucleus, a sparse mesh is still sufficient while a denser mesh is needed at the core where large momentum transfers (represented by spatially rapidly oscillating wavefunctions) can occur. We typically use a mesh where the radial spacing is larger by a factor of 20 for  $r \rightarrow 0$  when compared to  $r \approx r_{max}$ .

For a detailed description of technical issues of the numerical method as symmetrization and discretization of the radial Schrödinger equation, as well as the quadrature algorithm see the original publications by Chu *et al.* [66, 68, 69, 70, 71].

### 3.4 Grid dimensions and boundaries

Discretization of coordinate and energy space for numerical calculations is unavoidably linked to some finite spacing  $\Delta E = E_{n+1} - E_n$  and  $\Delta r$  of the grid, as well as to some maximal values  $E_{max}$  and  $r_{max}$  (“basis size”) that can be represented. Of course, these parameters have to be chosen such that all relevant physical processes can be represented on the finite numerical grid. For large intensities and long wavelengths, numerical evaluation of the Schrödinger equation is quite demanding even on modern computer systems, despite of the sophisticated technique used. It is therefore necessary to “economize” computing time and find the smallest basis that returns results of acceptable accuracy.

For a simple estimate of the basis size, the parameters defining the discretization of the radial Schrödinger equation, i.e. the number of radial grid points  $N$  and energy levels  $n_s \approx N/2$  [66, 68], the maximum in energy  $E_{max}$ , and the maximum in radius  $r_{max}$  can be related to the case of a rectangular hard wall potential (see Eqs . 3.16 and 3.16 in Sec. 3.3).  $N$  and  $r_{max}$  have to be chosen such that the largest energy expected during the time evolution is smaller than  $E_{max}$ . Quite naturally, it is at the same time required that the spacing of the states is smaller than the energy of one laser photon of the driving laser,  $\Delta E(E_{max}) < \omega_c$ . The number of angular momenta  $l_{max}$  needs to be also related to the number of photons taking part in the process, as each photon is able to supply an angular momentum of  $|l| = 1$ . Hence,  $l_{max} \propto E_{max}/\omega_c$  which can reach large numbers for the case of long-wavelength (e.g. infrared) driving lasers.

For calculating HHG, which is related to acceleration of an electron near the core, the considerations given above are already sufficient to obtain an estimate for the basis size needed. The largest energy to be described is given by the cut-off energy,  $E_{max} = E_c = I_p + 3.17U_p$ . For an intensity of  $10^{14}$  W/cm<sup>2</sup> and  $\lambda_c = 1000$  nm ( $U_p = 0.34$  a.u.), values needed are at least  $E_{max} > 1.6$  a.u.,  $l_{max} > 36$ ,  $N > 150$ , and  $r_{max} > 70$  a.u.. Numerically, using values twice as larger is typically necessary to obtain well converged results.

At the grid boundary  $r_{max}$  outgoing (ionized) wavepackets are reflected. This unphysical behavior (the grid boundary is artificial!) can be circumvented by applying an absorbing boundary described by the function  $f_d(r)$  at each time evolution step which prevents the wavepacket of reaching the grid boundary. For that purpose we use a  $\cos^2$ -function that evaluates to 1 at  $r = r_{cut}$  and goes to 0 at  $r = r_{max}$ , reading

$$f_d(r) = \cos^2 \left( \frac{\pi}{2} \frac{(r - r_{cut})}{(r_{max} - r_{cut})} \right). \quad (3.17)$$

Such a function is smooth to the  $2^{nd}$  order. The drawback is of course the loss of norm

(probability) associated with the damping. If the value of the damping integrated over time is recorded during time evolution, however, the ionization probability can be given reliably.

On the other hand, if the (doubly-differential) photo-electron spectrum shall be computed the grid dimensions need to be large enough to hold the relevant portions of the wavefunctions over the whole time evolution. Hence,  $r_{max} = \sqrt{2E_{max}} \cdot T$  and  $E_{max} > 5U_p$ . For an intensity of  $10^{14}$  W/cm<sup>2</sup>,  $\lambda_c = 1000$  nm and a pulse duration of four optical cycles ( $T \approx 550$  a.u.), values needed are at least  $E_{max} > 1.8$  a.u.,  $l_{max} > 40$ ,  $N > 1500$ , and  $r_{max} > 1100$  a.u.. Such a problem is therefore numerically much more costly than calculating a HHG spectrum.

## 3.5 One-electron potentials

A useful property of the pseudo-spectral method is that any rotationally symmetric potential  $V_{eff}(r)$  can be used. Therefore not only hydrogen-like atoms and ions can be computed by our algorithm but also all those atoms that can be treated by the “Single Active Electron” approximation (SAE) [57] with sufficient accuracy.

In the SAE approximation we assume that all tightly bound electrons of the lower-lying shells of the atom do not take part in the interaction with the laser field. Only the outermost - the “active” - electron is assumed to be relevant and its dynamics is calculated by the time-dependent Schrödinger equation using an effective radial potential  $V_{eff}(r)$  that accounts for the influence of the other electrons. This influence consists in particular in the screening of the nuclear charge  $Z$ .

Note that such effective potentials can not account for all effects in a multi-electron atom. Such potentials are often optimized to reproduce one observable of interest. In case of HHG it is important to reproduce the energy spectrum and in particular the ionization potential correctly (see chapter 2).

### 3.5.1 Hydrogen

For hydrogen, the potential  $V_{eff}(r)$  is exactly known, it is just the bare Coulomb potential  $-1/r$ . The ground state and the first excited states can be expressed numerically with an accuracy of more than 12 digits for a standard basis size.  $l$  is degenerate to the same precision. Higher states show a transition to the rectangular potential eigenvalues and have eventually positive eigenenergies. Figure 3.1 shows one-dimensional hydrogen (only  $l = 1$  states) with  $r_{max} = 200$  a.u. as an example.

### 3.5.2 Argon

Argon (Ar) has in total 18 electrons, the ground state with ionization potential  $I_p = 15.76$  eV is a  $3p^6$  state. It can be treated quite reliably in the Single-Active-Electron approximation. A much higher energy is required to excite the second electron ( $I_p = 27.62$  eV for  $\text{Ar}^+$ ). If one electron is excited and the second one remains in the ground state, the potential experienced by the single active electron can be modeled.

We employ the following one-electron potential [72]

$$V_{\text{ar}}(r) = -\frac{1}{r} [1 + A \cdot e^{-Cr} + (17 - A) \cdot e^{-Br}] \quad (3.18)$$

with the parameters  $A = 5.4$ ,  $B = 3.682$ , and  $C = 1$ . Employing this potential, the eigenenergies of the states are given by Table 3.1. Deeply bound states (1s, 2s, 2p, and

State	numerical	experimental
3p	-0.58137	-0.5792
4s	-0.15423	-0.1548
4p	-0.09711	-0.0982

**Table 3.1:** Ionization potentials out of the lowest lying states of argon in the Single-Active-Electron approximation with the potential given by Eq. 3.18. Experimental data is taken from the NIST database [73].

3s) are supported by this potential, but are occupied in the real atom. Consequently, they should not contribute to the dynamics in the framework of the SAE approximation. During the time-evolution we avoid occupation of these states by assigning to them a random phase.

Argon is frequently employed in experiments due to its easy handling. The value of the ionization potential  $I_P$  is close to the one of hydrogen, which is the only atom for which also (semi-)analytic approaches to laser-atom interaction exist.

## 3.6 Power spectrum

Once the time-dependent wavefunction is determined, the power spectrum - the most important quantity for HHG in the single-atom picture - can be deduced. In this context we consider a single atom showing a dipole moment of  $d(t) = \langle \Psi(t) | \hat{z} | \Psi(t) \rangle$  and assume sinusoidal variation of  $d(t)$ . Classically, a unit charge with dipole acceleration  $a_z = \ddot{d}$  pointing only in  $z$ -direction then emits radiation with a power of [43]

$$P(\omega) = \frac{\omega^4}{3c^3} d_{0,\omega}^2 = \frac{1}{3c^3} a_{0,\omega}^2. \quad (3.19)$$

For a short pulse with duration  $T$  one needs to use the Fourier components  $\tilde{a}(\omega)$ ,

$$P(\omega) = \frac{2\omega^4}{3c^3T} \tilde{d}(\omega)^2 = \frac{2}{3c^3T} \tilde{a}(\omega)^2. \quad (3.20)$$

A detailed discussion on the definition of radiated power can be found in the appendix A.2.

The property of the Fourier transform  $\mathcal{F}\{\}$

$$\mathcal{F}\left\{\frac{\partial^2 f}{\partial t^2}\right\} = -\omega^2 \mathcal{F}\{f\} \quad (3.21)$$

allows to calculate the power spectrum either by employing  $\langle \Psi(t) | \hat{z} | \Psi(t) \rangle$  or  $\langle \Psi(t) | \hat{a}_z | \Psi(t) \rangle$ . This is usually referred to as length (L) or acceleration (A) form of the power spectrum. For an Hamilton operator reading  $\hat{H} = \frac{\hat{\mathbf{p}}^2}{2} + V(r) + F(t)\hat{z}$  with  $\hat{z} = r \cos \theta$  the two forms can be written in the Heisenberg picture as

- Length Form (L):

$$d(t) = \langle \Psi | \hat{z} | \Psi \rangle = \langle \Psi | r \cos \theta | \Psi \rangle \quad (3.22)$$

- Acceleration Form (A):

$$a_z(t) = \langle \Psi | \frac{\partial^2 \hat{z}}{\partial t^2} | \Psi \rangle = -\langle \Psi | [\hat{H}, [\hat{H}, \hat{z}]] | \Psi \rangle = \langle \Psi | -\cos \theta V'(r) + F(t) | \Psi \rangle \quad (3.23)$$

with:

$$[\hat{H}, \hat{z}] = -i\hat{k}_z \quad [\hat{H}, [\hat{H}, \hat{z}]] = (\cos \theta V'(r) - F(t)) \quad (3.24)$$

Both forms should lead to the same result. This invariance is an excellent test for numerical convergence because the acceleration form are calculated from the wavefunction close to the origin, whereas the length form takes more remote coordinates into account. The power spectrum of the length form contains the Fourier transform of (with constant velocity) escaping electron wave packets. For ionizing laser intensities and large  $r_{max}$  (see Sec. 3.4) this contribution becomes dominant in the Fourier transform. It may even mask the response of the atom in the power spectrum that originates from electron wave packets localized (bound) at the atom.

Therefore, the acceleration form of the power spectrum should be used for highly ionizing pulses, especially for analyzing the cut-off region. On the other hand, the acceleration form tends to be less accurate for the lowest harmonics (1st-3rd harmonic) because of the loss of norm enters in its calculation. We are interested only in the plateau and cut-off region of the HHG spectra, thus we present power spectra in the acceleration form if not otherwise stated.

### 3.7 Photo-electron spectrum

After the time-integration of the TDSE up to the end of the pulse when  $F(T)$  equals to zero one arrives at the final wavefunction  $\Psi_{end} = \sum_{n,l} \Psi_{nl}(T)\Phi_{nl}(\mathbf{r})$ , given either in momentum space with  $k_{nl} = \sqrt{2E_{nl}}$  or in coordinate space. If only insignificant portions of the wavefunction have been absorbed by at the grid boundaries the total ionization probability can be written as  $P = \sum_{n,l} |\Psi_{nl}(T)|^2 \Theta(E_{nl})$ . Otherwise, the probability can also be deduced from the bound state population  $P = 1 - \sum_{n,l} |\Psi_{nl}(T)|^2 \Theta(-E_{nl})$ . However, for obtaining (doubly-)differential information on energy and/or angular distribution of photo-ionized electrons it is, of course, necessary to resolve as much of the wavefunction of the photo-electrons as possible on the grid.

The photo-electron energy spectrum is the most straight-forward differential quantity to be obtained. The pseudo-spectral method provides us directly with the spectrum (at discrete values), given by

$$\frac{dF}{dE}(E_{nl}) = \frac{|\Psi_{nl}(T)|^2}{E_{nl}}. \quad (3.25)$$

As soon as the electric field is switched off, the energy becomes an integral of motion for the Hamiltonian considered and  $(n, l)$  as well as  $E_{nl}$  are “good” quantum numbers. The photo-electron energy spectrum can be computed immediately.

# Chapter 4

## Quantum path interference in HHG

### 4.1 Global wavelength dependence of HHG

The dependence of the HHG yield on  $\lambda_c$  has become an issue of major interest [35, 36, 38, 39, 74, 75]. It has long been believed that the spreading of the returning wavepacket would result in a  $\lambda^{-3}$  dependence of the HHG efficiency [54] as long as ground state depletion can be neglected [76]; experimental findings [77] provided partial support. Recently, however, Tate *et al.* [35] have reported a different wavelength-scaling of HHG between 800 nm and 2  $\mu\text{m}$  calculated with the time-dependent Schrödinger equation (TDSE) for Ar and a strong-field approximation (SFA) for He. They found the yield to be described by a power law  $\propto \lambda^{-x}$  with  $5 \leq x \leq 6$ .

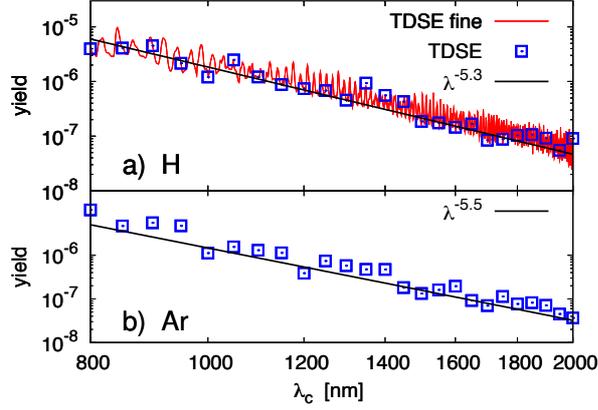
We analyze the very same problem within the framework of the single-active electron approximation. We define the HHG yield  $Y$  as described in Sec. A.2 (Eq. A.9), i.e. by integrating the modulus-squared of the dipole acceleration in an energy range  $[\omega_{lo}, \omega_{up}]$ , divided by the pulse duration. This definition has the dimension of radiated energy per unit time (power) and thus represents a useful quantity to compare the yield also from pulses with different duration. For a simple estimate - in analogy to the work of Tate *et al.* - we use a power law fit

$$Y \propto \lambda_c^{-x} \tag{4.1}$$

to describe the large-scale behavior of the HHG yield with the wavelength.

For a convenient comparison to previous work [35] we use a sine-like laser pulse (Eq. 2.5 with  $\phi_{CEP} = \pi/2$ ) and a flat-top pulse envelope with half an optical cycles ramp-on and ramp-off if not otherwise stated (see Sec. 2.1.4 for a definition of the envelope function  $f(t)$ ). When increasing  $\lambda_c$  we keep the number of optical cycles constant, hence, the total pulse duration increases as well.

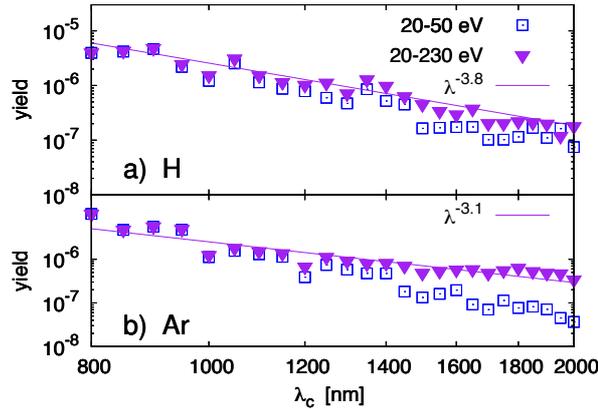
Figure 4.1 shows the HHG yield between 20 and 50 eV for a pulse of 8 cycles duration [36]. It is observed that the HHG yield decreases stronger with the driver's wavelength than a simple estimate might suggest. In agreement with previous work [35] we find



**Figure 4.1:** HHG yield as a function of  $\lambda_c$  in the energy range from 20 to 50 eV for a pulse with a duration of 8 cycles and  $I = 1.6 \times 10^{14} \text{ W/cm}^2$ . a) hydrogen, b) argon. Solid black lines represent a power law fit to the data (Eq. 4.1). The thin red line shows the fine-scale variation of the HHG yield for hydrogen.

that for argon the yield can be described on a large scale by a power law (Eq. 4.1) with  $x \approx 5.5 \pm 0.5$ . For H, the exponent is slightly lower and yields  $x \approx 5.3 \pm 0.5$ . A striking property of the wavelength dependence of  $Y$  is that it - quite unexpected - strongly fluctuates. These fluctuations are, in fact, surprisingly regular, the oscillation period is of the order of several nanometers near  $\lambda_c = 1000 \text{ nm}$  and decreases with increasing  $\lambda_c$ . Although fine-scale variations of the HHG yield are here presented only for hydrogen, they can be observed for other atomic species, in particular argon, as well. Such a behavior calls the applicability of a simple power law into question, at least when one has a fine-tuning of the HHG yield in mind. On large scales, for example when estimating the effect of switching from  $\lambda_c = 1000 \text{ nm}$  to  $\lambda_c = 1500 \text{ nm}$ , a power law remains valid.

An extension of the integration limits in the definition of  $Y$  (Eq. A.9) sheds some light on the question why  $Y$  decreases so strongly with  $\lambda_c$ . When defining the yield in the range from 20 to 230 eV, it essentially covers the whole harmonic spectrum being generated, as the classical cut-off energy is around 200 eV for the largest wavelength considered. Note also, on the other hand, that for low wavelengths of about 800 nm the range 20-50 eV covers the whole spectrum as well due to the much lower cut-off energy. Solid lines in Fig. 4.2 reveal that this re-defined, so to speak “total” yield (i.e. integrated over all energies  $\omega$ ) can be described again by a power law, but now with  $x \approx 3.1$  (Ar) and  $x \approx 3.8$  (H). This observation is now much closer to a  $\lambda^{-3}$  dependence originating from simple estimates of wavepacket spreading [54]. Consequently, the HHG yield is obviously partly determined by a “distribution” effect when observed within a given, constant range. The expansion of the whole spectrum to larger cut-off energies ( $E_c \propto \lambda^2$ ) re-distributed the HHG yield and leads to a stronger decrease in amplitude than what wavepacket spreading only ( $\lambda^{-3}$ )

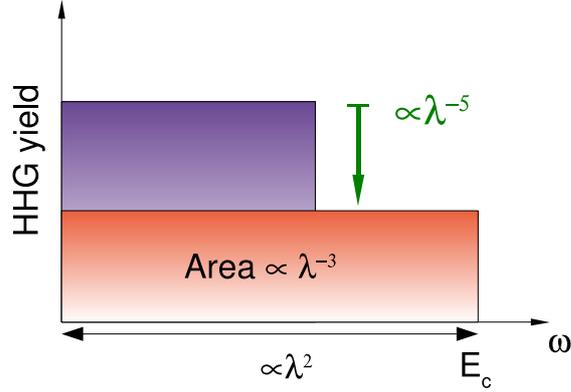


**Figure 4.2:** HHG yield as a function of  $\lambda_c$  in the energy range from 20 to 230 eV (covering the largest cut-off energy considered) compared to the range 20-50 eV, all parameters as in Fig. 4.1. a) hydrogen, b) argon. Solid lines represent a power law fit to the data (Eq. 4.1).

would imply. A cartoon visualizing these findings is presented in Fig. 4.3.

The present results have been very recently received support by experiments with Xe, where an analysis uncovering the single atom response in an energy range of 16 to 32 eV has revealed an exponent of  $x \approx 6.3 \pm 1.1$  [75]. This value is close to our result for Ar,  $x \approx 5.5 \pm 0.5$ , which is obtained for an energy range of 20 to 50 eV. At the same time, theoretical work [78] suggests a quite different picture. Here, an analytical, single-electron approach to HHG valid for short-ranged potentials [79] is combined with the true photorecombination cross sections of the multi-electron atoms employed. Focusing on the HHG yield near the cut-off only, Ref. [78] finds that the yield decreases more weakly for rare gases ( $x \approx 4.6$  for He and  $x \approx 3.9$  for Ne) as compared to hydrogenic atoms. Moreover, Ref. [78] reports that the wavelength dependence of the HHG yield may actually be insignificant for Ar or even show a large-scale, peak-like enhancement near  $1.2 \mu\text{m}$  for Xe. These surprising results are attributed to multi-electron effects such as additional resonances in the photorecombination cross section which are not present in a SAE approach. Although *ab-initio* two-electron calculations of HHG for helium have provided no evidence for a significant deviation from the SAE picture (cf. Fig. 2.2), further investigation of atoms with more than two electrons will be necessary in this context.

For comparison to the present TDSE results for hydrogen, Fig. 4.4 a) shows the large-scale wavelength dependence of HHG yield obtained in the framework of the SFA. The SFA compares nicely to the trend  $\propto \lambda^{-5.3}$  which was previously found in the TDSE analysis. With the SFA model at hand we can now investigate the relative importance of individual trajectories, in particular at long wavelengths. We define the relative weight



**Figure 4.3:** Illustration of the distribution of HHG yield with increasing  $\lambda_c$ . While the total yield (i.e. integrated over all energies  $\omega$ ) scales roughly like  $\lambda^{-3}$ , the higher cut-off energy  $E_c \propto \lambda^2$  leads to a drop of the spectral amplitude at a given energy by about  $\lambda^{-5}$ .

$r_j$  of the  $j^{\text{th}}$  trajectory by,

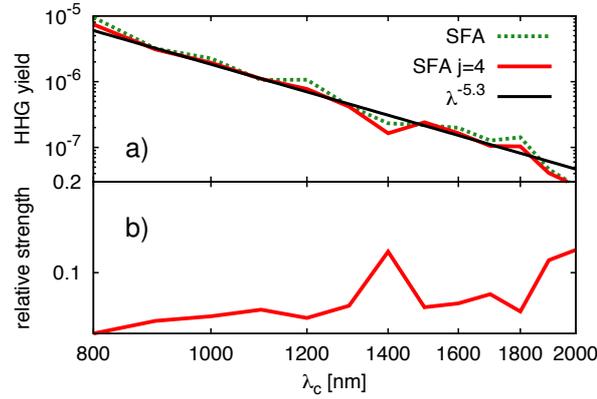
$$r_j = \frac{\mathcal{F}(d_j(t))}{\mathcal{F}(\sum_{i=1}^j d_i(t))}. \quad (4.2)$$

In words, we compare the yield of the  $j^{\text{th}}$  trajectory to the total yield obtained by summation of all trajectories of equal or lower order  $j$ . Interestingly, the contribution of long trajectories (an example is shown in Fig. 4.4 b) strongly increases with  $\lambda_c$ . This finding has been pointed out already by Tate *et al.* [35], however, the origin of this phenomenon was yet unclear.

It can be understood by a fairly simple picture: Consider a (recolliding) electronic wavepacket of Gaussian shape with spatial width  $\sigma = \sigma_k \cdot \tau_f$ . Here,  $\sigma_k$  is the initial momentum spread after (quasi-static) tunneling, it is related to the tunneling time and is independent of the wavelength [59]. In case of motion in an electric field the wavepacket will be centered around a classical trajectory characterized by the quiver amplitude  $\alpha = F_0/\omega_c^2$ . The spread relative to the position is given by

$$\frac{\Delta x}{\langle x \rangle} = \frac{\sigma_k \cdot \tau_f}{\alpha} \quad (4.3)$$

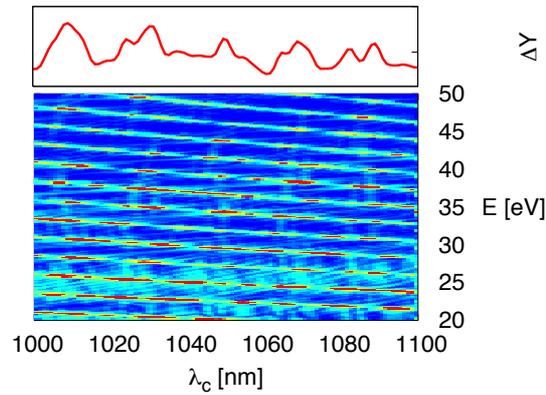
after time evolution by the flight time  $\tau_f$ . Using  $\tau_f \propto \lambda$  one arrives at a relative spread  $\propto 1/(F_0\lambda)$ . If the relative importance of wavepacket spreading decreases with  $\lambda_c$  contributions of trajectories with long excursion times that are usually suppressed become more prominent.



**Figure 4.4:** Large scale behavior of the HHG yield in SFA. a) shows the HHG yield including only four (red solid), and all (green dotted) quantum paths, the solid black line represents the trend  $\lambda^{-5.3}$  (see Fig. 4.2 a) ). b) shows the relative strength  $r_j$  (Eq. 4.2) of a particular quantum path,  $j = 4$ , which increases with  $\lambda_c$ .

## 4.2 Fine scale oscillations

Zooming into the fine-scale fluctuations in Fig. 4.1, surprisingly regular oscillations are observed. Interestingly, the regularity manifests strongly only in energy-integrated data, while a color plot as a function of both  $\lambda_c$  and energy of the radiation (Fig. 4.5) shows only weakly pronounced ridges along lines of constant  $\lambda_c$ .

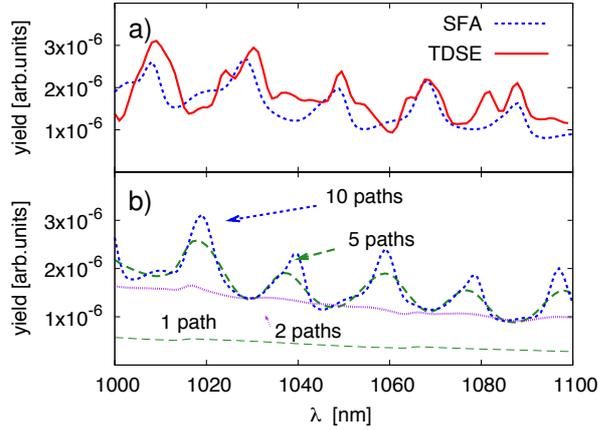


**Figure 4.5:** HHG yield as a function of both  $\lambda_c$  and radiation, same as Fig. 4.1. The upper panel show the quantity after integration over the given energy range.

Recently, we have been able to explain these oscillations in the integrated yield  $Y$  by quantum path interference of the electronic trajectories [36] involved in HHG (see Sec. 2.5

for a review of the role of “classical” trajectories in HHG). For this purpose we employ an SFA model for the time-dependent dipole moment  $d(t)$  of the radiating atom given initially by Ivanov *et al.* [58]. It relies in its core on classical electronic trajectories in the laser field which are given by the classical recollision condition (Eq. 2.14). The dipole moment  $d(t)$  is expressed in terms of three amplitudes reflecting the basic 3-step model [55] of HHG, summarized in Fig. 2.5. The exact expressions are given in Eqs. 2.24, 2.25, 2.26, and 2.27. The model employed in the following deviates from previous work only in the introduction of an “effective” ionization potential  $\tilde{I}_p$  which replaces  $I_p$  *only* in the phase factor accumulated during free propagation of the electron (cf. Eq. 2.26). This substitution leaves the structure of the HHG power spectrum largely unchanged.

The appealing advantage of employing a trajectory-based SFA model lies in the possibility of including or excluding a particular set of trajectories, which gives insight into the underlying processes and the importance of a given electronic trajectory. Figure 4.6 a) shows a comparison between the SFA model and the results obtained by numerical integration of the TDSE. The agreement in terms of modulation period and modulation



**Figure 4.6:** HHG yield as a function of  $\lambda_c$ , same as Fig. 4.1. a) comparison TDSE and SFA, b) SFA showing build-up of small-scale interference structures with increasing number of quantum orbits. Note that in a) an effective ionization potential of 13.04 eV was used for better comparison (to be discussed in Sec. 4.4), introducing an horizontal shift to the SFA curve.

amplitude of the HHG yield is astonishingly good. In Fig. 4.6 b) the SFA result is given for different total numbers of contributing electronic trajectories or “quantum paths”. While a single quantum path does not lead to any structures in the wavelength-dependent HHG yield, including more than five quantum paths gives practically converged results. Note that an effective ionization potential of 13.04 eV is used in Fig. 4.6 a). Using this “ad-hoc” value for  $I_p$  leads to an improved agreement with the TDSE result as compared to the agreement obtained with the true  $I_p$ . Employing the latter, which corresponds to

standard SFA, yields practically the same modulation period and modulation amplitude of the HHG yield. When introducing small corrections to  $I_p$ , only a horizontal shift with respect to the TDSE result is visible and essentially the same conclusions can be drawn. Employing an effective ionization potential in the context of quantum path interference has been previously suggested by various authors (see, e.g. Refs. [80, 81, 38]) as such horizontal shifts appear when comparing to SFA results.

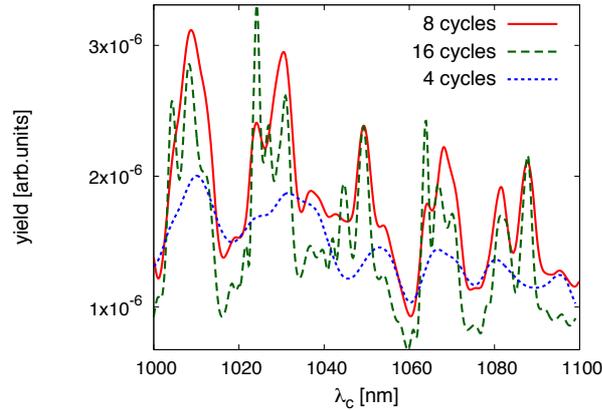
The interference structures in Fig. 4.6 appear on a scale of roughly 20 nm, which is much smaller than the Fourier width of the ultrashort pulses employed. Recall that a pulse duration of eight optical cycles corresponds to a Fourier width  $\Delta\lambda$  of about 100 nm for  $\lambda_c=1000$  nm ( $\Delta\lambda/\lambda_c \approx 0.1$ ). This - at a first glance surprising - fact reflects the origin of the oscillations being an interference structure bound to the well-defined time structure of the ultrashort laser electric field and the corresponding electronic trajectories. A similar example of observing regular structures after averaging over a large energy interval has been discussed extensively in literature in the context of “periodic orbit spectra”, see e.g. [82].

#### 4.2.1 Stability with pulse duration

We have recently analyzed the dependence of interference structures on the pulse duration [74]. Two simple effects can be expected: with decreasing pulse duration the number of possible trajectories decreases as well, hence the oscillations will eventually become less pronounced. On the other hand, the peaks of the oscillation may become sharper with increasing pulse duration due to an increasing number of trajectories contributing. This is counterbalanced by, firstly, wavepacket spreading, rendering trajectories with a long excursion time negligible and, secondly, by ground state depletion. For long pulses the ground state can be sufficiently depleted to “switch-off” HHG on the trailing edge of the pulse. Figure 4.7 presents HHG yield for hydrogen and flat-top pulses with durations of 4, 8, and 16 optical cycles, obtained by the full solution of the TDSE. The results are well in agreement with the above considerations. For a 16-cycles pulse at the present intensity ( $1.6 \times 10^{14}$  W/cm<sup>2</sup>) ionization reaches already 63%. Only minor changes are to be expected for an even longer pulse due to the large ground state depletion. Such a behavior could be confirmed by SFA calculations as well.

#### 4.2.2 Stability with pulse envelope

An important issue, especially with respect to experimental observability, is the robustness of the present results with changes of the pulse shape (pulse envelope). The exact pulse shape is experimentally hard to control. Moreover, the flat-top envelope function employed so far is - although convenient for semi-analytical analysis - very uncommon in the experiment. We have therefore investigated the stability of our present results with respect to different pulse shapes employing the full solution of the TDSE [74]. It turns



**Figure 4.7:** HHG yield as a function of  $\lambda_c$ , calculated by solving the TDSE, same as Fig. 4.1 but for pulse durations of 4, 8, and 16 optical cycles. The oscillatory structure gets more and more pronounced with increasing pulse duration.

out that whenever the effective pulse duration, namely the FWHM of the electric field envelope  $\tau_p$ , is comparable interference patterns are very similar (see Fig. 4.8).

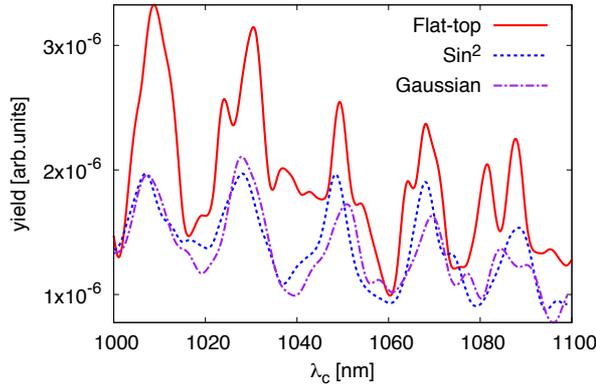
### 4.3 Intensity dependence and channel closings

To date a parameter relatively easy to control experimentally is the intensity of the driving pulse. At the same time, changing the driver's wavelength conventionally involves modifications of the laser resonator and/or the gain medium which represent a major change of the experimental setup. Along those lines, most experiments as well as theoretical investigations of HHG have so far focused on the intensity dependence. Indeed, oscillation of the HHG yield similar to those described in Sec. 4.2 have previously been reported in terms of the dependence on the intensity of the driving laser  $I_0 \propto F_0^2$ , both experimentally [83, 84] and theoretically [85, 86]. Borca *et al.* [85] and Milošević and Becker [86] have shown that HHG is enhanced at channel closings (CC), i.e., if

$$R = \frac{I_p + U_p}{\omega}, \quad (4.4)$$

is an integer. Channel closing in this context refers to the threshold for multiphoton ionization in a laser field. Note that the pondermotive potential  $U_p$  can be a function of  $\lambda_c$  and/or the intensity  $I_0$ ,  $U_p = U_p(\lambda_c, I_0)$ . Hence, enhancements can be induced either due to variation of  $\lambda_c$ , of  $I_0$ , or both parameters simultaneously.

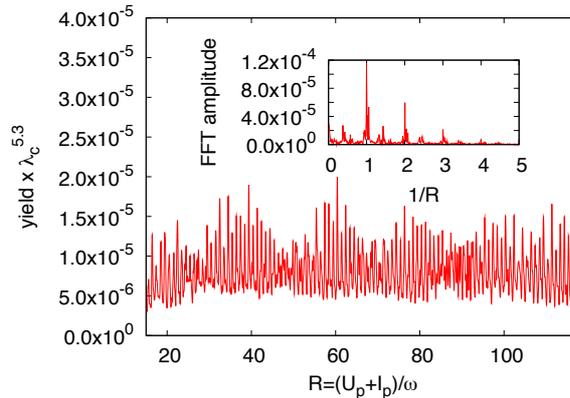
Most of these theoretical studies have employed zero-range potentials or the SFA which both neglect the influence of the long-ranged potential on the ionized electron. Furthermore, previous analysis has often made use of infinitely long pulses and neglected ground



**Figure 4.8:** HHG yield as a function of  $\lambda_c$ , calculated by solving the TDSE, same as Fig. 4.1 but for pulse envelopes corresponding to a flat-top,  $\text{Sin}^2$ , and Gaussian pulse shape. In contrast to all other data presented in this section, data was normalized here to the effective pulse duration  $\tau_p$ , which represents the FWHM of the electric field envelope, instead of the total duration  $T$ . Here,  $\tau_p = 7$  optical cycles for all pulse shapes.

state depletion. The latter is likely to introduce artefacts not being present in contemporary ultrashort (i.e. few-cycle) driving pulses. Nevertheless, the present oscillations and enhancements due to channel closings seem to be related, especially when we recall the source of quantum path interference being the semi-classical phase  $S_P$ . In fact, the expression  $I_p + U_p$  in Eq. 4.4 is identical to an order-of-magnitude estimate of  $S_P$  for large intensities and quantum paths with low energy.

In order to investigate the relation of our present results with the CC theory we replot Fig. 4.1 a) as a function of  $R$  rather than  $\lambda_c$ . Additionally, we have multiplied the data by  $\lambda^{5.3}$  in order to remove the “large-scale” behavior of  $Y$  and facilitate the analysis of the fine-scale oscillations. We observe that - as a function of the channel closing parameter  $R$  - the oscillations are now regular not only in a narrow range of  $\lambda_c$  but over the whole data range presented. In particular, the modulation period is clearly given by the spacing of  $\delta R = 1$ . This observation is underlined by a Fourier transform of the function  $Y(R) \times \lambda^{5.3}$ . The dominant frequency component of the modulation is equal to unity with an accuracy of at least three digits. The channel closing condition Eq. 4.4 predicts essentially the same modulation period  $\delta R = 1$  for variations in either  $\lambda_c$  or  $I_0$ , or both at the same time. An additional, beat-like structure with a frequency of  $\approx 20$  remains to be investigated in future work.



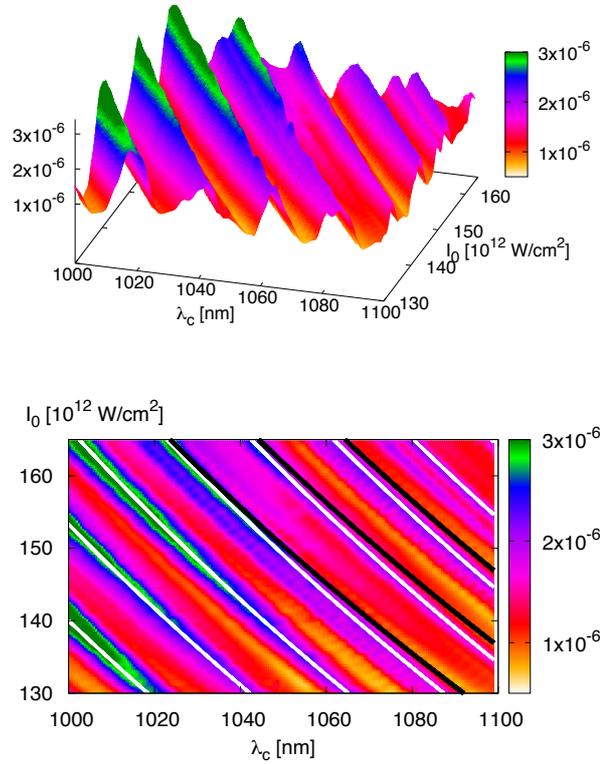
**Figure 4.9:** HHG yield for hydrogen as a function of  $R$ , same as Fig. 4.1 a). The inset represents a Fourier transform of the data, revealing  $R^{-1} = 1$  as the by far dominating contribution to the oscillatory structure.

### 4.3.1 Intensity dependence of yield oscillations

Due to the dependence of the ponderomotive potential  $U_p(\lambda_c, I_0)$  on both  $\lambda_c$  and  $I_0$  the investigation of HHG yield enhancements can be extended to the two-dimensional parameter plane  $(\lambda_c, I_0)$ , in principle. Figure 4.10 presents as an example the HHG yield for hydrogen in a narrow interval of wavelength ( $1 \mu\text{m} \leq \lambda_c \leq 1.1 \mu\text{m}$ ) and intensity ( $1.3 \times 10^{14} \text{ W/cm}^2 \leq I_0 \leq 1.6 \times 10^{14} \text{ W/cm}^2$ ) [37]. It displays regularly shaped ridges each of which can be mapped onto a fixed channel closing number  $R$ . This regularity is also reflected in the cuts through this two-dimensional data for different fixed intensities for both hydrogen (see Fig. 4.11 a) ) and argon (Fig. 4.11 b) ). In both cases, enhancements are spaced by a change  $\delta R = 1$ . In contrast to a presentation in terms of  $\lambda_c$ , one-dimensional cuts through the data of Fig. 4.10 at different  $I_0$  are almost identical when given in terms of  $R$ . In both Fig. 4.11 a) and b) curves for the HHG yield slightly shift in vertical direction with increasing intensity. This effect can be intuitively explained by an increasing ionization rate (and thus increased HHG yield) at larger intensities.

### 4.3.2 Channel closings

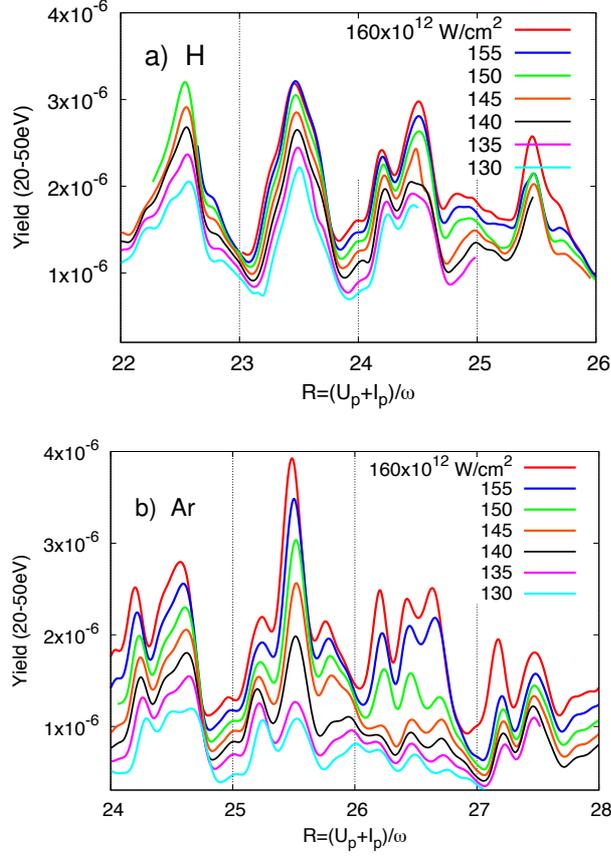
Although the modulation period of  $Y(R(\lambda_c))$  (Fig. 4.9) as well  $Y(R(\lambda_c, I_0))$  (Fig. 4.10) can be simply interpreted by a channel closing approach (i.e. the period being determined by a change of  $\delta R = 1$ ), there is one major drawback. The peaks/enhancements obtained by TDSE calculations are observed not to coincide with integer values of  $R$ . Such a disagreement between SFA-based theories and TDSE data has been already discussed in previous work [80, 81]. Attempts were made to resolve this discrepancy by introducing an “effective” ionization potential  $\tilde{I}_p$  replacing  $I_p$  in the semi-classical phase, an approach



**Figure 4.10:** TDSE-calculated integrated harmonic yield between 20 and 50 eV for H (8-cycle flat-top pulse) in the  $(\lambda_c, I_0)$  plane. In the contour plot (lower panel), white lines show values of constant  $(U_p + I_p)/\omega$ , shifted from integer values by  $+0.52$ , while black lines (only three are shown for clarity) represent values of constant  $(U_p + \tilde{I}_p)/\omega$  with  $\tilde{I}_p = 10.5$  eV.

we have previously employed as well (see Sec. 4.2 and Ref. [36]). Frolov *et al.* [38] have recently analyzed the wavelength-dependence of HHG in terms of channel closings. They have calculated the harmonic yield using the time-dependent effective range theory, and shown that the peaks of the yield oscillation around  $\lambda_c = 1000$  nm coincide indeed with integer values of  $R$  if an effective ionization potential  $\tilde{I}_p$  (e.g., 10.5 eV for H) is used in place of  $I_p$  in Eq. 4.4. Note, however, that their method is strictly applicable only for short-range potentials and also neglects excited atomic states.

Superimposed on the contour plot in the lower panel of Fig. 4.10 we present lines of constant channel closing parameters  $R$ , equally spaced by  $\delta R = 1$ . While black lines represent values of constant  $(\tilde{I}_p + U_p)/\omega$  with an effective ionization potential of  $\tilde{I}_p = 10.5$  eV, white lines show values of constant  $(I_p + U_p)/\omega$  (with the *true* ionization potential), but shifted



**Figure 4.11:** Wavelength dependence of the integrated harmonic yield (20 to 50 eV) in the range of  $\lambda_c \approx 1000 - 1100$  nm, expressed in terms of  $R$ , for a) H and b) Ar, for 8-cycle flat-top pulses for different intensities indicated in the figure. In contrast to a presentation in terms of  $\lambda_c$ , the curves for different  $I_0$  are almost identical apart from a slight vertical off-set due to an increasing ionization rate at larger intensities.

from integer values by  $+0.52$ . Although the ridges of HHG enhancement are described by both iso-lines at a first glance, closer inspection shows that the curvature of iso-lines defined with  $\tilde{I}_p$  deviates from the actual ridges. Apparently, accepting a seemingly constant off-set, employing the true  $I_p$  fits the actual TDSE-calculated HHG enhancements much better. This may not come as a surprise after the Fourier analysis of the oscillations in a broad range of  $\lambda_c$  (800 - 2000 nm) has shown that  $\delta R = 1$  holds with a very high accuracy if the true ionization potential is used. Using a constant  $\tilde{I}_p$ , valuing e.g. 10.5 eV (instead the true value of 13.6 eV for hydrogen) leads to a small, but significant deviation from the  $\delta R = 1$  law by about 3%.

In conclusion, an analysis in term of the channel closing parameter  $R$  reveals that the modulation period for enhancements of the HHG yield at variations of both  $\lambda_c$  and  $I_0$  is given by the condition  $\delta R = 1$  if and only if the true ionization potential is used. The idea of employing an effective ionization potential  $\tilde{I}_p$  constant over a large interval of  $\lambda_c$ , which has been discussed in literature in order to explain enhancements at non-integer values of  $R$ , can be excluded from our present analysis. Only in a narrow interval of  $\lambda_c$ ,  $\tilde{I}_p = \text{const.}$  may be applied. An in-depth discussion on a generalization of  $\tilde{I}_p$  as well as on alternative corrections to the channel closing picture will be given below (Sec. 4.4).

### 4.3.3 Modulation period of HHG yield

The channel closing condition offers a clear way to explain the change of the modulation period  $\delta\lambda$  with the driver's wavelength  $\lambda_c$ . More accurately,  $\delta\lambda$  can be defined by the distance between to neighboring peaks  $\delta\lambda = \lambda_{n+1} - \lambda_n$  at wavelengths  $\lambda_n$  and  $\lambda_{n+1}$ .

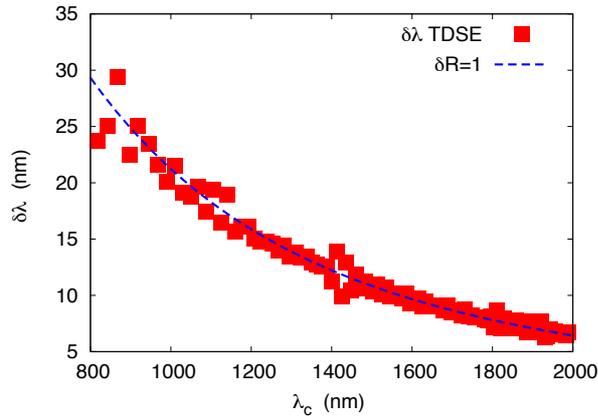
Although the general trend  $\delta\lambda \propto \lambda_c^{-2}$  can be readily explained also from estimating the largest contribution to the phase picked up by the electron during free propagation in the laser field [36], the channel closing condition  $\delta R = 1$  is more accurate and intuitive. From Eq. 4.4,  $\delta R = 1$  can be rewritten in terms of a change in wavelength  $\delta\lambda$ .

$$\begin{aligned} \delta\lambda &\propto \frac{1}{I_p + 3U_p}, \\ \rightarrow \delta\lambda &= \frac{1240}{I_p(\text{eV}) + 2.8 \times 10^{-19} I(\text{W/cm}^2) \lambda^2(\text{nm})} \text{ nm}. \end{aligned} \quad (4.5)$$

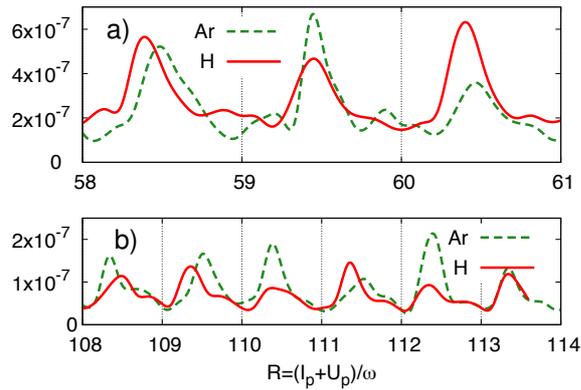
The above expression is to be interpreted as follows: Starting from a parameter set allowing for an enhancement of HHG yield (a peak), a change by  $\delta\lambda$  will again lead to peak in HHG yield. Consequently, this quantity is the modulation period to be determined. Equation 4.5 reproduces the TDSE-calculated  $\lambda$  dependence of  $\delta\lambda$  extremely well (Fig. 4.12).

### 4.3.4 Comparison of different atoms with different $I_p$

Oscillations for the HHG yield are not solely observed for hydrogen but appear to be a general phenomenon. We have analyzed different rare-gas atoms, in particular argon (Ar) and neon (Ne), due to their relevance for experimental applications. Presenting calculations in terms of the channel closing parameter  $R = (I_p + U_p)/\omega$  offers a nice way of eliminating the dependence on the ionization potential  $I_p$  of the atom under consideration. The dependence of  $I_p$  on the HHG yield via the ionization rate leads to a vertical shift only. Additionally, possible differences can then be traced to the different shape of the atomic potential. Figure 4.13 summarized results for HHG yield  $Y(R)$  as a function of  $R$  for two different atoms: hydrogen and argon. The agreement is generally surprisingly



**Figure 4.12:** Modulation period  $\delta\lambda$  as a function of the central driver wavelength  $\lambda_c$  for atomic hydrogen. ■: TDSE, dashed line:  $\delta R = 1$  (Eq. (4.5)).



**Figure 4.13:** HHG yield as a function of  $R$ , same as Fig. 4.1 but comparing H and Ar. a) show a region near  $\lambda_c = 1500$  nm while b) presents oscillations near  $\lambda_c = 2000$  nm.

good and underlines the applicability of the channel closing picture and the relevance of quantum path interference for arbitrary atoms. Discrepancies between the atoms become smaller for larger wavelengths, i.e. larger  $U_p$  (see Fig. 4.13 b) ), where the SFA-based channel closing picture is expected to be an even better description.

## 4.4 Coulomb corrections

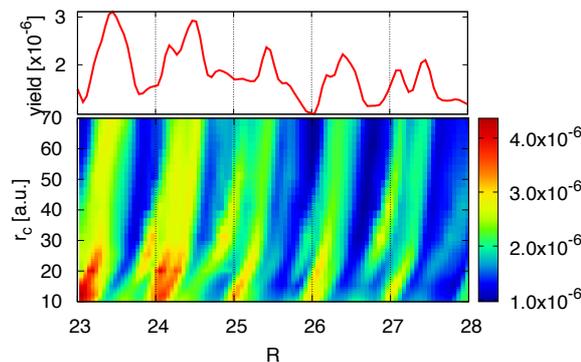
### 4.4.1 Truncated Coulomb potential

Following the discussion in Sec. 4.3.2, it is clear that employing a constant  $\tilde{I}_p$  over a wide range of parameters is problematic. On the other hand, the channel closing picture has worked extremely well for describing the modulation period of the HHG yield. If one wants to retain these findings a generalized,  $\lambda_c$ -dependent correction defined as  $\Delta\tilde{I}_p(\lambda_c) \equiv \tilde{I}_p(\lambda_c) - I_p$  can be invoked in order to bring peak positions in agreement with CC theory.

In fact, the relevance of the channel closing picture is underlined by the following numerical experiment: SFA results, which neglect the presence of any potential after the ionization by the laser, are approximately recovered by employing the full solution of the TDSE with a potential that is sufficiently short-ranged. In this context we perform calculations with a truncated Coulomb potential [37], given by

$$V_{eff}(r, r_c) = \begin{cases} -\frac{1}{r} & (r < r_c) \\ -\frac{e^{-(r-r_c)/r_d}}{r} & (r > r_c) \end{cases}, \quad (4.6)$$

where the effective range of the truncated Coulomb potential  $r_c$  is varied between  $r_c = 10$  and  $r_c = 70$  a.u. and the width of the cross-over region  $r_d$  is chosen to be  $r_d = 10$  a.u.. For these parameter values, the ionization potential and the first excitation energy remain unchanged to an accuracy of  $\approx 10^{-9}$  and  $\approx 10^{-3}$ , respectively. It should be noted that the classical electron quiver motion amplitude is  $\alpha_q \approx 32$  a.u. for  $I = 1.6 \times 10^{14}$  W/cm<sup>2</sup> and  $\lambda_c = 1000$  nm.



**Figure 4.14:** Harmonic yield as a function of both the channel closing parameter  $R$  and the effective range of a truncated Coulomb potential  $r_c$ . Enhancements shift from integer values of  $R$  to the right with increasing  $r_c$ . Convergence to the full Coulomb potential (upper panel) is reached only for  $r_c \gtrsim 60$  to 70 a.u..

Interestingly, with decreasing  $r_c$  the oscillatory pattern of the HHG yield shifts more and more to the left in an almost uniform fashion. Near  $r_c = 10$  a.u. the enhancements can be finally found at integer values of  $R$  in full agreement with the CC picture. Apparently, the potential at  $r > r_c$  is responsible for the shift observed. Note that choosing  $r_c < 8$  a.u., i.e. smaller than the tunnel exit at this intensity, would alter ionization dynamics and should thus be avoided. This approach shows not only that effects of the true (long-ranged) potential are responsible for discrepancies with an SFA-like theory, but also gives an important information about the modulus of  $\Delta\tilde{I}_p$ . Clearly, as the shift between the full and the truncated potential  $\Delta R < 1$ , it follows that  $\Delta\tilde{I}_p < \omega$ .

#### 4.4.2 Effective ionization potential

In previous literature, different lines of arguments are invoked for employing  $\tilde{I}_p$  rather than  $I_p$ . However, they all have in common that the existence of a strongly distorted, continuum-like excited state  $\varepsilon_n$  is considered responsible for an effectively *lower* ionization threshold [38, 80, 81]. In the following we explore the relevance of different choices of  $\Delta\tilde{I}_p$ :

Faria *et al.* [80] argue that  $\varepsilon_n$  should be given by the state of principal quantum number  $n$  whose radius  $r_n \approx 3n^2/2$  matches the quiver amplitude  $\alpha_q = F_0/\omega^2$ . Together with the Rydberg energy  $\varepsilon_n \approx -I_p/n^2$  this implies

$$\Delta\tilde{I}_p \approx -\frac{3\omega^2}{2F_0}I_p \propto I_0^{-1/2}\lambda_c^{-2}. \quad (4.7)$$

Accordingly, the change of the effective ionization potential,  $\Delta\tilde{I}_p$ , becomes wavelength and intensity dependent.

Employing the full solution of the TDSE provides the exact position of peaks/enhancements of HHG yield. Let us denote the position of a peak in terms of  $R$  by  $R_p$ . Assuming CC theory with a correction  $\Delta\tilde{I}_p(\lambda_c)$  to be valid, the latter can be computed. The mismatch  $\Delta R_p$

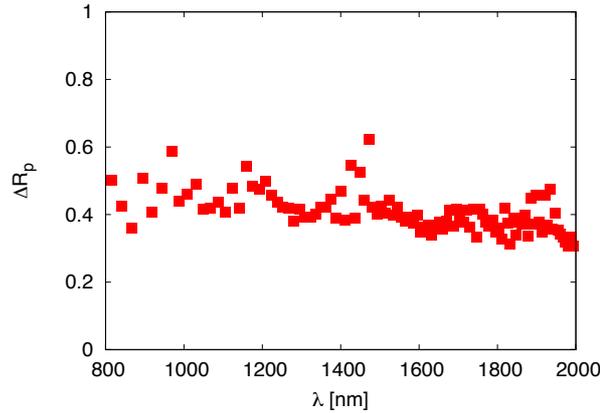
$$\Delta R_p \equiv R_p - [R_p], \quad (4.8)$$

between the actual position  $R_p$  and the nearest integer value  $[R_p]$  is related to the effective ionization potential by

$$\Delta\tilde{I}_p = -\Delta R_p \omega. \quad (4.9)$$

This mismatch is obviously almost constant in the entire range between  $\lambda_c = 800$  nm and 2000 nm (Fig. 4.15). Consequently, according to Eq. 4.9,  $\Delta\tilde{I}_p \propto \lambda^{-1}$ .

Hypothesis Eq. 4.7 is not consistent with our present results. In addition, no upper limit for  $\Delta\tilde{I}_p$  according to Eq. 4.7 was discussed in literature. This may lead to the evidently incorrect conclusion that  $\Delta\tilde{I}_p \rightarrow 3.4$  eV as soon as in a low intensity and low wavelength limit the  $n = 2$  Rydberg state (or even the ground state!) would govern the effective threshold invoked.



**Figure 4.15:** Mismatch  $\Delta R_p$  between the actual position  $R_p$  and the nearest integer value  $[R_p]$  of a peak in the HHG yield. About 100 peaks between a driver's wavelength of 800 and 2000 nm are shown, the dependence on  $\lambda_c$  is apparently weak.

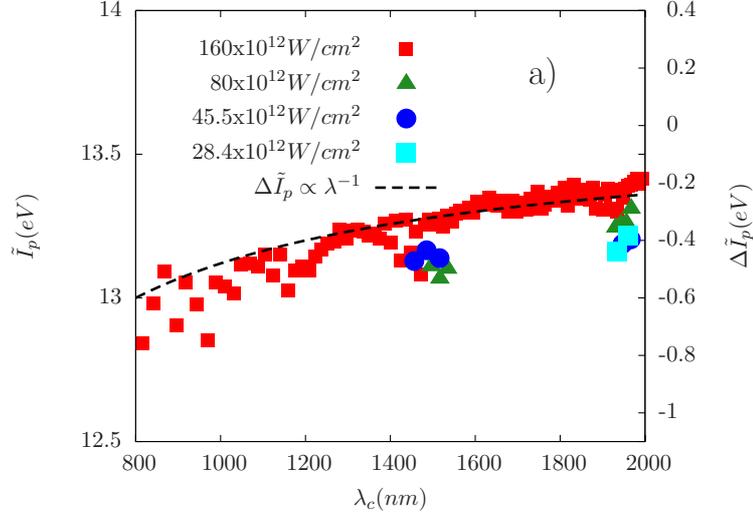
An alternative proposal put forward by Frolov *et al.* [38] relates the energy  $\varepsilon_n$  to the formation of an effective continuum by broadening of the level with principal quantum number  $n$ . Accordingly,  $\varepsilon_n$  is determined by the condition  $\Gamma_n = \Delta\varepsilon_n$ , where the width  $\Gamma_n$  (related to ionization rate) approaches the level spacing  $\Delta\varepsilon_n$ . While in the limit of quasi-static tunneling, the tunneling rate (Eq. 2.10) strongly depends on the field strength  $F_0$  but only very weakly on  $\lambda$ . The resulting value of  $\Delta\tilde{I}_p$  is estimated to be -3.1 eV for atomic hydrogen and  $I = 1.6 \times 10^{14}$  W/cm<sup>2</sup> in Ref. [38]. This is inconsistent with the observation in Sec. 4.4.1 that  $|\Delta\tilde{I}_p|$  should be smaller than the photon energy  $\omega$  ( $\lesssim 1.5$  eV in the present parameter range) of the driving laser pulse.

Extracting  $\Delta\tilde{I}_p$  from TDSE results by applying Eq. 4.9 allows to trace the dependence on both intensity and wavelength as given by Figs. 4.16 and 4.17. The intensity dependence is quite weak and can be described by a power law  $\Delta\tilde{I}_p \propto I_0^{-0.3}$ . As discussed above a proportionality of  $\Delta\tilde{I}_p \propto \lambda^{-1}$  is found.

### 4.4.3 Coulomb correction along classical trajectories

In the previous section we have seen that employing the full solution of the TDSE allows for the determination of  $\Delta\tilde{I}_p$ . Additionally, the analysis with a truncated Coulomb potential of variable effective range (Sec. 4.4.2) has underscored the importance of the long-ranged part of the potential. A more detailed investigation can be facilitated by employing simple models rather than an all-numerical treatment.

For that purpose we reexamine the role of classical trajectories - as the essential subclass of quantum paths - in HHG. According to the SFA model employed to describe HHG (Eqs. 2.24 -2.27) the electron picks up a semi-classical phase along those trajectories.



**Figure 4.16:**  $\tilde{I}_p$  (left-hand axis) and  $\Delta\tilde{I}_p$  (right-hand axis) for hydrogen as a function of  $\lambda_c$  for various intensities as indicated [37]. The dashed line presents an estimate corresponding to  $\Delta R_p = \text{const.}$  which leads to  $\Delta\tilde{I}_p \propto \lambda^{-1}$ .

Because we have identified the long-ranged Coulomb tail to influence  $\Delta\tilde{I}_p$ , we introduce the additive phase correction  $\Delta S_P(t_i, t_f)$  that builds up due to the interaction with the core potential along the path  $P$ . The interaction is assumed to be a weak correction that can be accounted for by a perturbative approach. The trajectory is assumed to be identical to the unperturbed case, i.e. the timing information  $(t_i, t_f)$  remains unchanged. With this approximation  $\Delta S$  can be written as [58]

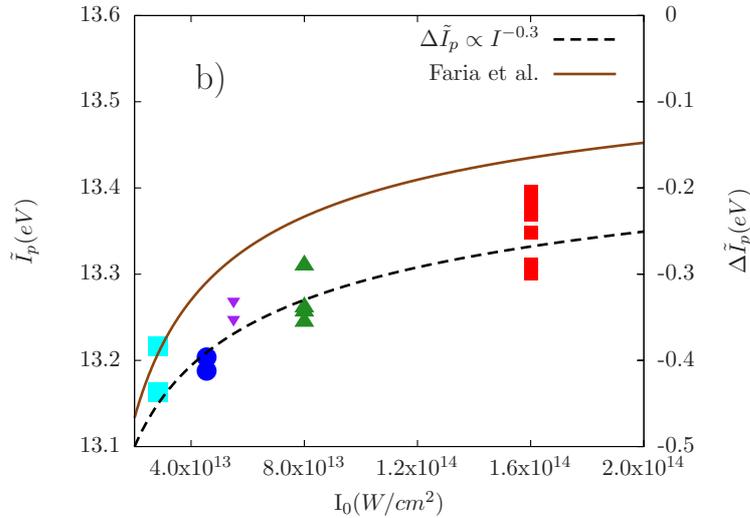
$$\Delta S_P(t_i, t_f) = \int_{t_i}^{t_f} V_{EI}(r(t')) dt'. \quad (4.10)$$

Such an approach corresponds to an eikonal approximation to a weak perturbative potential (see Sec. 2.7.4).  $V_{EI}$  denotes the potential being accounted for. Equation 4.10 is evaluated along classical trajectories in the laser electric field  $F(t)$ , confined to the direction of polarization. Trajectories start (and end) at the “tunnel exit”  $z_0 = I_p/F_0$  as suggested by the three step model.

Clearly, the eikonal approximation fails at small distances from the nucleus where the correction introduced by the potential is large. This difficulty can be bypassed using the observation (Sec. 4.4.2) that at an effective potential range  $r_c = 10$  a.u. the SFA limit of channel closings at integer values of  $R$  is reached. Consequently, we set

$$V_{EI}(r) = V_{eff}(r, r_c = \infty) - V_{eff}(r, r_c = 10) \quad (4.11)$$

when calculating the long-range phase correction.

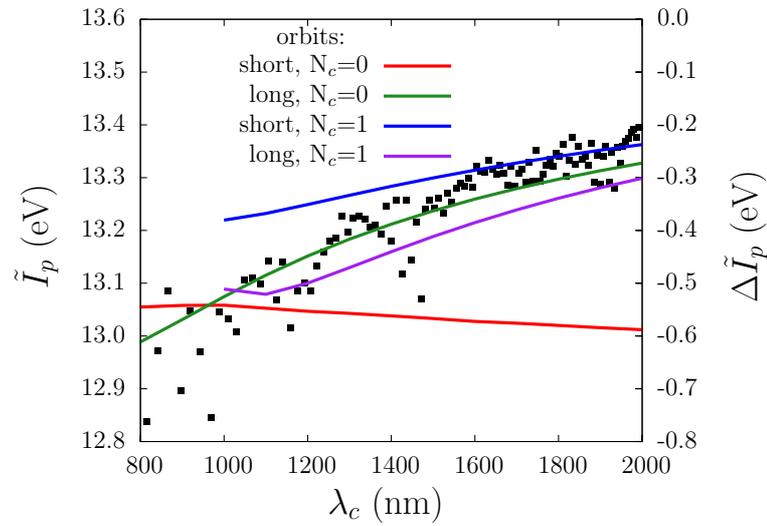


**Figure 4.17:**  $\tilde{I}_p$  (left-hand axis) and  $\Delta\tilde{I}_p$  (right-hand axis) for hydrogen as a function of driver’s peak intensity  $I_0$  extracted from peaks in the vicinity of  $\lambda_c = 1950$  nm [37]. The dashed line represents a fit to the data corresponding to  $\Delta\tilde{I}_p \propto I_0^{-0.3}$ , while the solid line compares to the prediction of Faria *et al.* (Eq. 4.7).

It is now suggestive to express this additional phase in terms of a change in the “effective ionization potential”,  $\tilde{I}_p$ . Accordingly,

$$\Delta\tilde{I}_p = \Delta S_P(t_i, t_f)/(t_f - t_i). \quad (4.12)$$

We have investigated the induced effective ionization potential stemming from the six shortest trajectories that all lead to the same recollision energy. For that energy we take  $E_c = 20$  eV, a value that corresponds to a HHG radiation energy of  $\approx 33.6$  eV, hence roughly in the center of energy range 20 to 50 eV under consideration. In this example, the electric field is taken to be perfectly sinusoidal with a constant peak intensity of  $1.6 \times 10^{14}$  W/cm<sup>2</sup> and is subject only to a variation of  $\lambda_c$ . For every given recollision energy two possible trajectories (long and short) exist with a flight time  $\tau_f < T_{cyc}$ . For all subsequent half-cycles there may again be two more trajectories each which then have revisited the parent ion  $N_c$  times (see Fig. 2.4). Extracted data is shown in Fig. 4.18 for the four shortest trajectories (orbits) that revisit the core  $N_c$  times (here: zero and one) before eventually recombining. It is remarkable that such a simple model can describe the dependence of the TDSE-calculated  $\Delta\tilde{I}_p$ . Not only is the trend well reproduced, but the prediction is also rather accurate on an absolute scale. Furthermore, trajectories with different excursion times behave surprisingly similar - save only the shortest one. This can be understood from the fact that multiple-returning trajectories quickly approach the limiting trajectory of electronic quiver motion, hence they approximately “feel” the same influence of the atomic core potential when normalized to the trajectory flight time  $\tau_f$ .



**Figure 4.18:**  $\tilde{I}_p$  (left-hand axis) and  $\Delta\tilde{I}_p$  (right-hand axis) as a function of driver wavelength [37], employing Eq. 4.12 and the potential of Eq. 4.11. Intensity is  $I = 1.6 \times 10^{14}$  W/cm<sup>2</sup>. Lines stem from the six shortest orbits recolliding with 20 eV and revisiting the core  $N_c$  (here: zero and one) times before recombination (as indicated). For each energy and each  $N_c$ , a short and a long orbit exists. ■: TDSE data (see also Fig. 4.16).

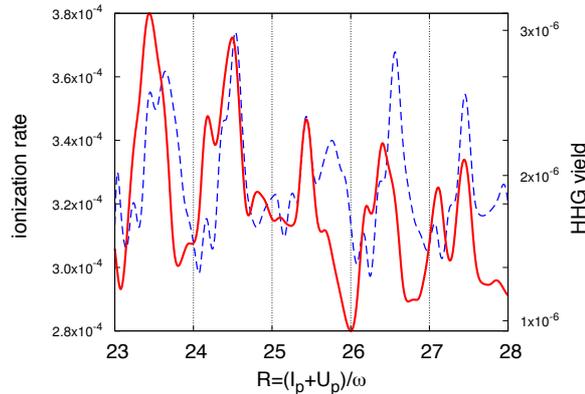
## 4.5 Oscillations in the ionization yield

Investigations in the previous sections have been devoted to the HHG response of the atom. The basic interaction, however, is the “first” step of HHG: the ionization process. Only a small fraction of ionized electrons eventually recombine with their parent ion while the largest portion will simply contribute to the photo-electron spectrum and the total ionization yield  $P_{ion}$ . In the following we will explore our results in view of the ionization rate as a function of  $\lambda_c$  and intensity of the driver and compare to HHG results.

### 4.5.1 Correspondence to HHG

In order to arrive at a quantity independent of the pulse duration  $T$  (analogously to the HHG yield) we define a cycle-averaged ionization rate as the ionization probability at the end of the pulse divided by  $T$ , hence  $\Gamma_{av} = P_{ion}/T$ . If only a narrow interval of  $\lambda_c$  or  $R$  is investigated, the pulse duration  $T$  does not change much and is only a small correction to a discussion in terms of  $P_{ion}$ .

Figure 4.19 plots  $\Gamma_{av}$  alongside the HHG yield  $Y$ . Although it is well known that ionization and HHG are unavoidably linked, their very similar dependence on  $\lambda_c$  is remarkable and is found even on a fine wavelength scale. Similar to HHG (Fig. 4.9) oscillations in



**Figure 4.19:** Ionization rate (blue dashed, left-hand axis) and HHG yield (red solid, right-hand axis) for hydrogen as a function of  $R$  compared. The pulse has a duration of eight cycles and a flat-top envelope.

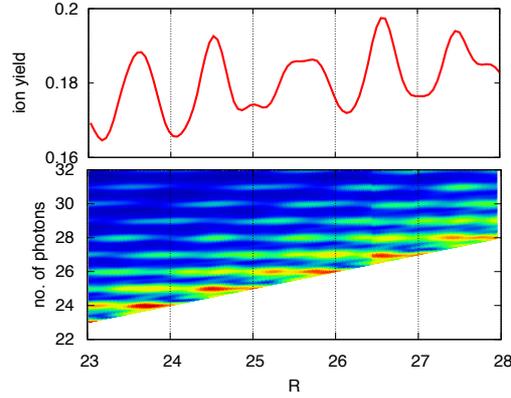
the ionization rate persist over the whole range of  $\lambda_c = 800$  to 2000 nm investigated, being permanently “locked” to the HHG yield (not shown). A Fourier analysis of  $\Gamma_{av}(R)$  has revealed a principal modulation period of  $\delta R_{ion} = 1$  as well, although the corresponding Fourier peak is slightly less prevalent as for HHG (see Fig. 4.25 b) below). A common origin of the two modulations, i.e. quantum path interference, seems evident. Although

we show here only data for our “case in point”, the flat-top pulse, we have observed a similar correspondence between HHG and ionization rate for all pulse shapes and durations investigated.

Note also that the modulation amplitude of  $\Gamma_{av}(R)$  is only of the order of ten percent in contrast to enhancements by a factor of three to six observed for HHG.

### 4.5.2 Quantum path interference in the photo-electron energy spectrum

Modulations in the ionization rate can be also observed in the energy-resolved photo-electron spectra. Employing the full solution of the TDSE photo-electron energy spectra are computed by Eq. 3.25. We present photo-electron spectra of flat-top pulses with a duration of four optical cycles (and half a cycle ramp-on/off) as a function of  $\lambda_c$  in Fig. 4.20. In the two-dimensional parameter plane ( $n = (E - I_p - U_p)/\omega_c, R$ ) oscillations are



**Figure 4.20:** Photo-electron spectra as a function of both the number of absorbed photons  $n = (E - I_p - U_p)/\omega_c$  and  $\lambda_c$ , expressed here in terms of the channel closing number  $R$ . The upper panel shows the integral over the energy, i.e. the ionization probability. The pulse has a duration of four optical cycles and a flat-top envelope,  $I = 1.6 \times 10^{14}$  W/cm<sup>2</sup>.

now observable in both coordinate directions.  $n$  is equivalent to the number of absorbed photons and is introduced here to underline the similarity of photo-electron spectra for different  $\lambda_c$ .

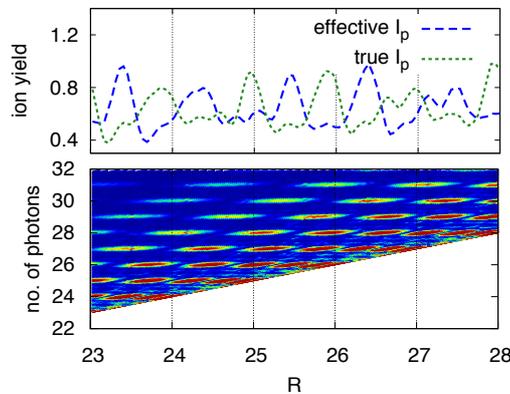
Subsequent photon peaks of the photo-electron spectrum change amplitude at fixed  $R$ , while at the same time photon peaks with  $n$  fixed oscillate as a function of  $R$  as well. The modulation period slightly depends on the photo-electron energy itself, it increases with energy. Integration over the energy (upper panel of Fig. 4.20) yields the total ionization

probability  $P_{ion}$ . After this integration the modulation period turns out to be  $\delta R = 1$  with a high accuracy.

For an interpretation of the numerical results we again consider the strong field approximation. We employ the SFA ionization amplitude, Eq. 2.17, but analogous to the approach in HHG we substitute the transition matrix element  $F(t_i) \cdot D(\vec{k} + \vec{A}(t_i))$  with a tunneling amplitude  $a_{ion} = \sqrt{\Gamma_{ADK}(F(t_i))}$ . We restrict our calculations to a one-dimensional model,  $\vec{k} \rightarrow k$ . The photo-electron energy spectrum is then obtained by

$$\frac{dF_{SFA}}{dE}(E = k^2/2) = \frac{|b(k)|^2}{k} \quad (4.13)$$

and  $P_{ion,SFA} = \int_0^\infty dF_{SFA}/dE dE = \int_0^\infty dk |b(k)|^2$  is the total ionization probability. Photo-electron spectra as a function of  $\lambda_c$  are given in Fig. 4.21. Modulations in both

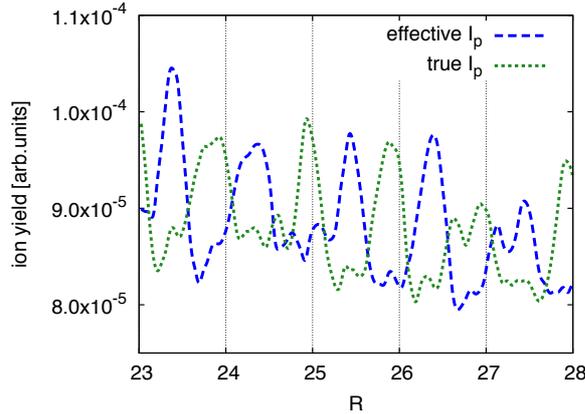


**Figure 4.21:** SFA photo-electron spectra (Eq. 4.13) as a function of both the number of absorbed photons  $n = (E - I_p - U_p)/\omega_c$  and  $\lambda_c$ , expressed here in terms of the channel closing number  $R$ . The upper panel shows the integral over the energy, i.e. the ionization probability  $P_{ion,SFA}$ . Pulse parameters are the same as in Fig. 4.20.

coordinate directions  $(n, R)$  are visible. This compares qualitatively well to the TDSE result save the two following aspects:

Firstly, oscillations are again shifted horizontally in comparison to the TDSE results. In fact, where a maximum is visible in the TDSE results, the SFA shows a minimum. In close analogy to HHG the introduction of an effective ionization potential  $I_p \rightarrow \tilde{I}_p$  in the SFA phase of Eq. 2.17 restores good comparison between the upper panels of Figs. 4.20 and 4.21. Note that we use the very same  $\tilde{I}_p$  as found in the analysis of Sec. 4.4.2. Although minor differences for distinct spatial ionization pathways can be expected, the Coulomb potential of the remaining ionic core evidently influences the action and semi-classical phase in a similar way as discussed in Sec. 4.4.3.

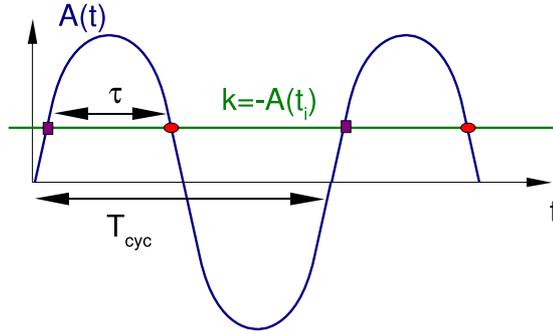
A second deviation is that the modulation amplitude of the ionization probability predicted by the SFA is too large. This is due to the over-simplification of the problem, treated only in one spatial coordinate and disregarding wavepacket spreading. As a remedy we introduce a phenomenological factor  $1/(t_f - t_i)$  under the time integral of Eq. 2.17. This factor accounts for wavepacket spreading in the two dimensions perpendicular to the polarization direction not explicitly included. The effect on the modulation of the



**Figure 4.22:** SFA ionization yield  $P_{ion,SFA}$  as a function  $\lambda_c$  expressed in terms of  $R$ , phenomenologically including wavepacket spreading. Note the much smaller oscillation amplitude (in comparison to the upper panel of Fig. 4.21) which is in good agreement with the TDSE calculations.

ionization yield is presented in Fig. 4.22. In agreement with the TDSE calculation, the amplitude of the oscillations is, in this case, about 10 % of the total yield as compared to almost a factor of two obtained without wavepacket spreading (cf. Fig. 4.21). The latter can be thus regarded as one important reason for “imperfect” interference of consecutive ionization pathways.

The present results are very similar to the ones of Lindner *et al.* [87] who observed modulations in the photo-electron energy spectra due to intra-cycle path interference. In their experiment, variations in the interference structure have been induced by varying the carrier-envelope phase  $\phi_{CEP}$ . Although in our particular case changes of the exact shape of the laser electric field are determined by changing  $\lambda_c$  instead of  $\phi_{CEP}$ , the interpretation of the underlying physics is equivalent. The phenomenon can be most easily explained by the example of a time-double slit interference, schematically explained in Fig. 4.23. For each final momentum of an electron, two different times of ionization are possible, even within the same optical cycle. Interference of these electrons depends on the difference between the phase acquired on each individual electronic trajectory. The phase difference is largely determined by the “time-double slit” spacing  $\tau$  which, depending on the exact

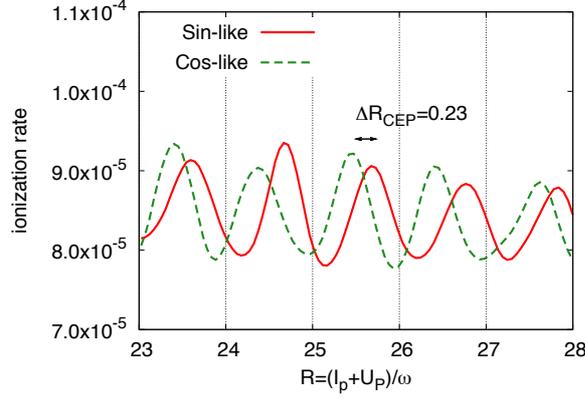


**Figure 4.23:** Path interference for photo-electron energy spectra, schematically. In the framework of SFA the final momentum acquired by an electron born at time  $t_i$  is given by  $k = -A(t_i)$ . This condition is fulfilled twice per optical cycle, contributions born at times denoted by  $\bullet$  and  $\blacksquare$  can interfere. The time-double slit spacing  $\tau$  is modified by variation of  $\lambda_c$  via  $T_{cyc}$ .

shape of the laser electric field, can be influenced by variation of either  $\lambda_c$  or - for short pulse - of the carrier-envelope phase  $\phi_{CEP}$ .

### 4.5.3 Ultrashort pulses: $\phi_{CEP}$ effects

Indeed, for short pulses ( $\sin^2$ -envelope) the  $\phi_{CEP}$  should govern the interference as it determines the exact shape of the light field and thus the spacing between two interfering paths [87]. The difference between sin- and cosine pulses can be quantified by employing an effective ponderomotive potential  $U_{p,eff} = F_m^2/4\omega^2$ , defined by the the maximum of the field  $F_m$  instead of  $F_0$ . For a sine pulse,  $F_m$  is smaller than in the case of a cosine-pulse (where  $F_m = F_0$ ), leading to a difference in  $U_{p,eff}$ . This introduces a shift in the  $R$ -coordinate reading  $\Delta R_{CEP} = \Delta U_{p,eff}/\omega$ . For the present parameters  $\Delta R_{CEP} = 0.2285$ , which is in very good agreement with the TDSE as well as SFA results (see Fig. 4.24).



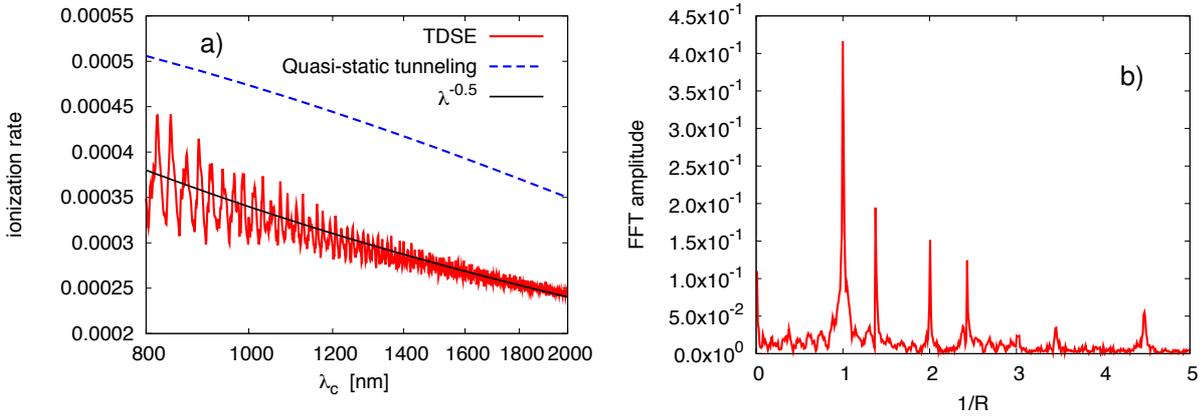
**Figure 4.24:** Cycle-averaged ionization rate calculated by solving the TDSE for a 4 cycles (FWHM)  $\sin^2$ -pulse,  $I_0 = 1.6 \times 10^{14}$  W/cm<sup>2</sup>. Sin-like and Cos-like data are shifted by roughly  $\Delta R_{CEP}$  due to a  $\phi_{CEP}$ -induced effective ponderomotive potential.

## 4.6 Global wavelength dependence of ionization

For completeness, we also present the TDSE calculated ionization rate  $\Gamma_{av}$  (averaged over the pulse duration) for hydrogen for a wide range of wavelengths, namely for  $\lambda_c = 800$  reaching up to 2000 nm. We find that oscillations in the average rate are present over the entire range investigated (Fig. 4.25). A Fourier transform of  $\Gamma_{av}(R) \times \lambda^{0.5}$  (Fig. 4.25 b)) shows that the oscillations are dominated by the period  $R^{-1} = 1$ , however, not as pronounced as in the case of HHG yield (cf. inset of Fig. 4.9).

On a large wavelength scale, the TDSE result for  $\Gamma_{av}$  is well fitted by the proportionality  $\lambda^{-x}$  with  $x = 0.5$ . When we recall our results for the HHG yield  $Y \propto \lambda^{-x}$  with  $x \approx 5.3$ , which we have contrasted with an estimate of  $Y \propto \lambda^{-5}$  based on the distribution of yield over the plateau of the spectrum (cf. Fig. 4.3), one might conclude that the decrease in the ionization rate may be responsible for an exponent  $x$  larger than five found by TDSE calculations. Further investigations in this context are needed and might reveal the contribution of each of the three steps of HHG to the overall scaling of HHG yield with  $\lambda_c$ .

In comparison to the TDSE results we also present quasi-static tunneling ionization which is frequently employed for strong field ionization. The ionization rate obtained by employing Eq. 2.10 indeed shows a very similar trend as a function of  $\lambda_c$ . Interference effects are, of course, not accounted for by tunneling rates. On an absolute scale, Eq. 2.10 overestimates the ionization by 15 to 40% as compared to present TDSE result. This accuracy is nevertheless sufficient for safely applying quasi-static tunneling within an SFA model of HHG (Eq. 2.24). In quantitative descriptions the small inaccuracy in the



**Figure 4.25:** a) Ionization rate  $\Gamma_{av}$  for hydrogen obtained by numerically integrating the TDSE (red solid) and by tunnel ionization (blue dashed, determined by employing Eq. 2.10) as a function of  $\lambda_c$  for an 8 cycle flat-top pulse,  $I = 1.6 \times 10^{14}$  W/cm<sup>2</sup>. The black solid line represents a power law fit to the TDSE data with an exponent of  $x = -0.5$ . Figure b) shows the Fourier transform of  $\Gamma_{av}(R) \times \lambda^{0.5}$ , revealing  $R^{-1} = 1$  as the dominating contribution to the oscillatory structure (cf. inset of Fig. 4.9).

absolute value of the tunneling rate is only one out of many quantities such as e.g. intensity and interaction volume which are not exactly known. Qualitatively, experimental HHG spectra are well described.



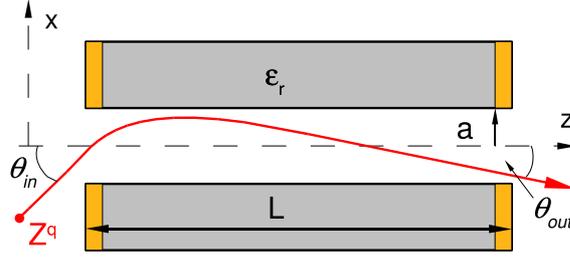
# Chapter 5

## Guiding of Highly Charged Ions

In the following chapters we investigate guided transport of charged particles through insulating nanocapillaries with a diameter of the order of several hundred nanometers. We will start by reviewing the classical transport theory (CTT) that we have recently developed for describing charged particle transport in a capillary. In the present chapter, we particularly discuss the interaction of highly charged ions (HCI) with the insulating surface of the inner walls, followed by a presentation of some outstanding results for HCI guiding that have been recently discussed extensively in literature. Here, we will once more focus on the aspect the temporal behavior of guided transmission and on the time scales involved. An extension of the discussion and the method employed to electrons as projectiles is given in the next chapter (chapter 6).

### 5.1 Charged particle transport in a nanocapillary

We consider charged projectiles with charge state  $q$  and energy  $E$  impacting onto a capillary target with an (tilt) angle  $\theta_{in}$  with respect to the surface normal and the capillary axis. A schematic view of projectile trajectories inside the nanocapillaries considered is given in Fig. 5.1. Capillaries with a length  $L$  and width  $2a$  have typically aspect ratios  $2a : L$  of 1:150 - 1:50, corresponding to geometric opening angles  $\theta_0 \approx 0.25^\circ - 1^\circ$ . The dielectric material is characterized by a static dielectric constant of  $\epsilon_r$ . Front and back sides of a capillary target are usually covered by a thin layer (several tens of nanometers) of conducting material which prevents a global charge-up of the whole target during beam exposure. The beam spot on the target covers typically  $10^4 - 10^6$  individual capillaries. The lateral inter-capillary spacing is of the order of  $\approx 10a$  such that, on an atomic level, inter-capillary interaction effects can be neglected.



**Figure 5.1:** Schematic view of charged projectile guiding through capillaries.

### 5.1.1 Brief description of CTT simulation

A theoretical description and *ab-initio* simulation of guiding of charged projectiles poses a considerable challenge in view of the widely disparate time scales simultaneously present in this problem [26], spanning about 10 orders of magnitude: while projectile-surface interaction, the microscopic charge-up, and charge transport all occur on an atomic (i.e. sub-fs) time scale, the time between two projectiles entering the same capillary is already of the order of 0.1 sec for typical experimental parameters [18]. The macroscopic discharge of the internal surfaces of the extremely well insulating materials may take hours or even days. Such multi-scale problems represent a major challenge for an *ab-initio* simulation. We have therefore developed a mean-field classical transport theory (CTT) [88] based on a microscopic classical-trajectory Monte Carlo simulation for the projectile transported. Projectile trajectories (having flight times of the order of  $10^{-10}$  sec) are calculated in the mean field of charges situated on the internal walls which move on much faster time scales. The forces governing subsequent trajectories (Eq. 5.1) are self-consistently coupled to the charge-up of and charge transport at the internal capillary walls (cf. Ref. [26]),

$$m_p \dot{\vec{v}} = \vec{F} = \vec{F}_{im} + \vec{F}_{wall} (+\vec{F}_{stoch}). \quad (5.1)$$

In Eq. 5.1,  $m_p$  and  $\vec{v}$  represent the mass and the velocity of the projectile and  $\vec{F}_{im}$  accounts for the image acceleration of the projectile by employing the classical, static limit  $\vec{F}_{im} = q^2/(4d^2) \cdot (\epsilon_r - 1)/(\epsilon_r + 1) \hat{r}$ .  $d$  and  $\hat{r}$  are the distance of the projectile to and the radial unit vector pointing towards the surface, respectively. For the HCI considered the static limit is a good approximation due to the small velocities ( $v \approx 0.1$  a.u.) involved.  $\vec{F}_{wall}$  follows from the electrostatic wall potential via  $\vec{F}_{wall} = -\vec{\nabla}V_{wall}$ . The wall potential itself is expressed by

$$V_{wall}(\vec{r}, t) = \int_{surface} da' \frac{\sigma(\vec{r}', t)}{|\vec{r} - \vec{r}'|} + \sum_{\{j\}}' \frac{q_j(t)}{|\vec{r} - \vec{r}_j|}, \quad (5.2)$$

hence by contributions from the surface charge density  $\sigma(\vec{r}, t)$  and bulk charges  $q_j(t)$ . Dielectric screening is accounted for by appropriate initial values for  $\sigma(\vec{r}, t_s)$  and  $q_j(t_s)$

at the time of deposition  $t_s$ . Recall that the time interval between two subsequent projectiles entering the same capillary is of the order of 0.1 sec. Therefore, both of the two contributions to the wall potential need to be modified by charge transport during time evolution between the subsequent trajectory calculations (see Sec. 5.1.3 for the discussion on charge transport). The stochastic force  $\vec{F}_{stoch}$  describes (non-deterministic) scattering of projectiles at the surface as a sequence of impulsive momentum transfers [89]. This only needs to be accounted for in the case of electronic projectiles (see discussion in Sec. 5.1.2 and chapter 6 below).

Employing the model adopted here, we relate the microscopic projectile-surface interaction and projectile transport to known macroscopic properties of the materials involved such as conductivity, dielectric constant, composition, and electronic structure. The aim of this approach is, despite of the complexity of the processes involved, to be predictive and to allow for a description of the projectile guiding without resorting to freely adjustable parameters or additional ad-hoc assumptions [90].

### 5.1.2 Wall interaction at impact

For the close-contact interaction between the charged projectile and a surface we distinguish two kind of projectiles: electrons and ions, in particular HCI.

Upon impact on the internal capillary wall HCI undergo a sequence of charge transfer process leading eventually to full neutralization [91]. The secondary electron emission coefficient is low for the HCI parameters (velocity, charge state) considered in this work. Moreover, as long as secondary electrons do not leave the nanocapillary, the aggregate charge deposited by the HCI impact is equal to its initial charge state  $q$ . In the simulation it is assumed that  $q$  elementary charges are Gaussian distributed on the surface with a width of distribution  $r_d \approx 1000$  a.u.. Note that after neutralization and charge deposition the trajectory of the remaining neutral atom is not followed because the focus of our work lies on the description of projectiles transported in their initial charge state.

For electrons, the interaction with a solid surface is clearly different. Electrons may scatter at the surface leading to stochastic forces  $\vec{F}_{stoch}$  which influence the overall projectile trajectory. The processes involved as well as their theoretical description will be discussed in chapter 6. However, note that once charge carriers have been deposited on the internal capillary walls, we treat the subsequent charge transport on the surface and into the bulk on equal footing for both HCI and electron transmission.

### 5.1.3 Charge transport and screening

When charge carriers are deposited on the internal capillary walls, the finite conductivity leads to charge transport at the surface and into the bulk. The materials used to manufacture nanocapillaries are extraordinarily good insulators with a bulk conductivity of e.g.  $\sigma_b \cong 10^{-16} \Omega^{-1}\text{m}^{-1}$  in the case of PET. Charge transport *along* the surface, however,

may be much faster. In the case of PET the ratio of surface and bulk conductivity is of the order of 100 [92]. Because of the supposedly large importance of surface conductivity we treat the latter explicitly by performing a two-dimensional random walk for the charge carriers which modifies the surface charge density  $\sigma(\vec{r}, t)$ . This random walk is governed by the surface diffusion constant  $D_s$  [26]. The diffusion constant is related via  $\sigma_{b,s} = \frac{ne^2}{kT} D_{b,s}$  to the experimentally determined value for the amount of charge carriers in Mylar  $n \cong 10^{18} \text{ m}^{-3}$  [93] as well as to the conductivity. Employing the order-of-magnitude values known for these quantities,  $D_b$  should be of the order of  $10^{-17} \text{ m}^2\text{s}^{-1}$  and  $D_s \approx 10^{-15} \text{ m}^2\text{s}^{-1}$ . We use in fact  $D_b = 2 \cdot 10^{-17} \text{ m}^2\text{s}^{-1}$  and  $D_s = 100 \cdot D_b$  unless otherwise stated. In addition, the charges enter the bulk material with a probability of  $D_b/D_s$ . Further charge transport is then governed by bulk charge transport which is approximated by an exponential decay of the charges (time constant  $\tau_b \approx (2a)^2/D_b$ ) where at the same time their position is kept fixed.

For  $\text{Al}_2\text{O}_3$  the density of charge carrier traps is  $n \cong 10^{23} \text{ m}^{-3}$  [94]. Together with its bulk conductivity  $\sigma_b \cong 10^{-12} \Omega^{-1}\text{m}^{-1}$  one arrives at  $D_b = 2 \cdot 10^{-18} \text{ m}^2\text{s}^{-1}$ . We use, in fact,  $D_b = 10^{-16} \text{ m}^2\text{s}^{-1}$  corresponding to  $\tau_b \approx 200 \text{ s}$ . This is motivated by similar values of  $\tau_b$  found in experiments with flat  $\text{Al}_2\text{O}_3$  surfaces [95] as well as time scales of the same order of magnitude observed in experiments with  $\text{Al}_2\text{O}_3$  capillaries [27]. The surface conductivity is given in Ref. [96] by  $\sigma_s \cong 10^{-15} \Omega^{-1}\text{m}^{-1}$  at dry conditions, and  $\sigma_b \cong 10^{-8} \Omega^{-1}\text{m}^{-1}$  at larger humidity. Experimentally, some  $\text{H}_2\text{O}$  adsorption is very likely, leading to a large surface conductivity. In a first attempt we use  $D_s = 100 \cdot D_b$  analogous to PET, and we have verified that a variation by a factor of about 5 does not significantly influence simulation results.

Within the framework of linear response, the bare charges  $q$  are screened by the dielectric medium in its vicinity [90]. Denoting the bare Coulomb interaction between a projectile and the charge deposited by  $V_C$  the effective screened interaction, denoted by  $V_{SCR}$ , depends on the location of the deposited charge  $q$ . In the limiting case that the charge has already diffused into the bulk, bulk screening would apply, i.e.  $V_{SCR} = V_C/\epsilon_r$ . For the electrostatic potential we use the static limit  $\epsilon_r = \epsilon(q \rightarrow 0, \omega \rightarrow 0)$  of the dielectric function since the characteristic time available for build-up of screening (time interval  $\tau_{rep}$  between two subsequent projectiles entering the capillary  $\approx 10^{-1} \text{ sec}$ ) is very large compared to the characteristic time for optical excitations,  $\tau_{rep} \gg 2\pi/\omega_{opt}$ . The dielectric screening of surface charges is determined by the induced polarization, i.e. the “image charge” having the weight  $\chi_s = (\epsilon_r - 1)/(\epsilon_r + 1)$ . The effective residual charge and hence the effective potential is given by  $V_{SCR} = V_C(1 - \chi_s) = 2V_C/(\epsilon_r + 1)$  [90]. Our simulation employs the latter expression, i.e. surface screening, if not otherwise stated. To illustrate the significance of dielectric screening, however, we will also show results using bulk screening instead as well as calculations for bare interactions ( $V_{SCR} = V_C$ ).

## 5.2 Results for HCI Guiding

In a number of publications [26, 90, 97, 98] we have presented various aspects of our work on HCI guiding through nanocapillaries. After developing a CTT for HCI guiding [26, 97] we have focused on the angular width of the transmitted beam [98] and have finally analyzed the effect of different levels of dielectric screening [90]. In this work we will present quintessential as well as some particularly recent results and refer to previous contributions for more details.

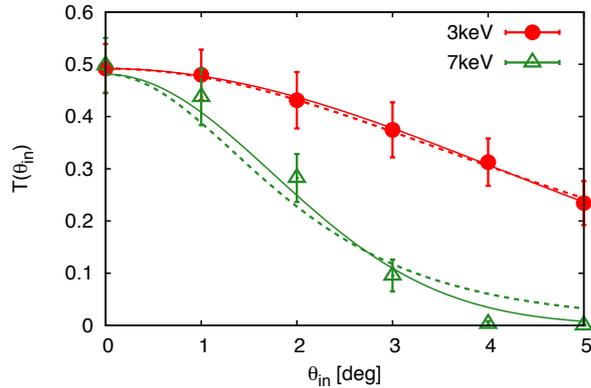
We focus on capillaries manufactured from PET ( $\epsilon_r = 3.3$ ); the length is  $L = 10 \mu\text{m}$  and diameter yields  $2a = 200 \text{ nm}$  if not otherwise stated. This system has been explored in the experiment most extensively. Starting from the first observation of HCI guiding [18], data have become available for many incident energies and charge states [99]. Additionally, characteristics of the transmitted beam have recently been measured [100, 101] with a temporal resolution that reveals the dynamical properties of the system. The time scales involved here are very distinct from the ones in ultrafast optics. The system evolves on a scale of the order of minutes due to the large macroscopic discharge times of the insulating materials employed. When observing only transmitted projectiles, time scales on which observables might change are naturally bounded from below by the time interval between two projectiles entering the same capillary,  $\approx 0.1 \text{ sec}$ .

### 5.2.1 Energy-dependence of HCI guiding

Because parameters entering our simulation can be computed from material data only with an order-of-magnitude accuracy a comparison to the experiment over a broad range of e.g. incidence energies is needed for an efficient testing of our model. For example, it has been shown that incident HCI with the same  $E/q$  ratio - typically of the order of 0.1-1 keV - lead to the same results within the experimental uncertainty [99]. This observation is in accordance with scaling properties of classical projectile motion in an electrostatic potential. Indeed, our self-consistent CTT simulation confirms this observation.

Quite naturally, the guiding effect for HCI decreases with increasing  $E/q$  ratio. The larger the energy, the stronger is the deflection and thus the charge deposition required. Large charge deposition needs more time to build up and leads to a larger discharge current, which, in turn, needs to be compensated by a diminishing transmission current. The total transmission rate  $T(\theta_{in})$  of  $\text{Ne}^{7+}$  ions through PET nanocapillaries under equilibrium conditions (Fig. 5.2) serves as one example. Transmission extends far beyond the geometrically allowed angle ( $\approx 1^\circ$ ) and the dependence on the tilt angle is similar for both 3 and 7 keV, but guiding decreases with increasing energy.

Following Hellhammer *et al.* [102] the transmission rate of HCI through nanocapillaries is fitted by a Gaussian  $T(\theta_{in}) = T(0^\circ) \exp(-\sin^2 \theta_{in} / \sin^2 \theta_g)$ . Hence, transmission can be given in terms of the guiding parameter  $b = \sin^{-2} \theta_g$  or the effective guiding angle  $\theta_g$ . At the guiding angle the transmission function has decreased to  $T(0^\circ)/e$ . The original



**Figure 5.2:** Transmission function  $T(\theta_{in})$  as obtained for  $\text{Ne}^{7+}$  ions through a PET capillary with a diameter of 200 nm for a capillary described in Fig. 5.1 for different energies (from Ref. [90]).  $\bullet$ :  $E = 3$  keV,  $\triangle$ :  $E = 7$  keV. Solid lines: Gaussian fits to the simulation [102] with  $\theta_g = 5.8^\circ \pm 0.1^\circ$  (3 keV), and  $\theta_g = 2.5^\circ \pm 0.1^\circ$  (7 keV). Dashed lines offer alternative fits to the data given by  $T(\theta_{in}) = T(0^\circ)/[1 + (\theta_{in}/\theta_l)^2]^2$  with  $\theta_l = 7.7^\circ \pm 0.1^\circ$  (3 keV) and  $\theta_l = 3.0^\circ \pm 0.3^\circ$  (7 keV).

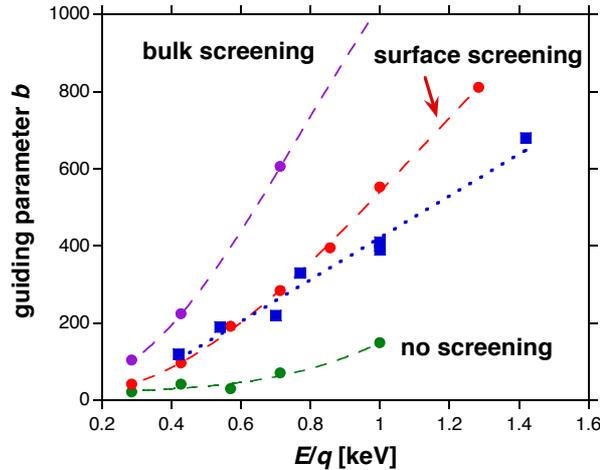
motivation for the Gaussian ansatz was a Boltzmann type “thermalization” argument for the transverse energy. Note, however, that also other choices of fitting functions of  $T(\theta_{in})$  perform similarly well. An example is shown for a square-Lorentzian in Fig. 5.2.

But how exactly does guiding change with increasing energy? By assuming guiding to be a statistical process, Refs. [99, 102] predict a linear behavior for  $b$  as a function of  $E/q$ . Fig. 5.3 shows the guiding parameter  $b$  as a function of the scaled kinetic energy  $E/q$ . Additionally, we illustrate the effect of dielectric screening (see Sec. 5.1.3) on guiding. While in all the models of dielectric screening the transmission function  $T(\theta_{in})$  behaves similar (not shown) guiding drops much faster with the energy for bulk screening than observed in the experiment [99, 102]. On the other hand, surface screening is able to reproduce experimental data surprisingly well over a broad range of  $E/q$ . Small discrepancies for  $E/q > 1$  are not yet fully understood but might be related to the very small absolute value of the transmission rate. The unfavorable statistics makes simulations increasingly difficult here.

In conclusion, comparison with the experiment has shown that surface screening dominates the dielectric shielding of deposited charges inside an insulating nanocapillary.

## 5.2.2 Time-dependence of HCI guiding

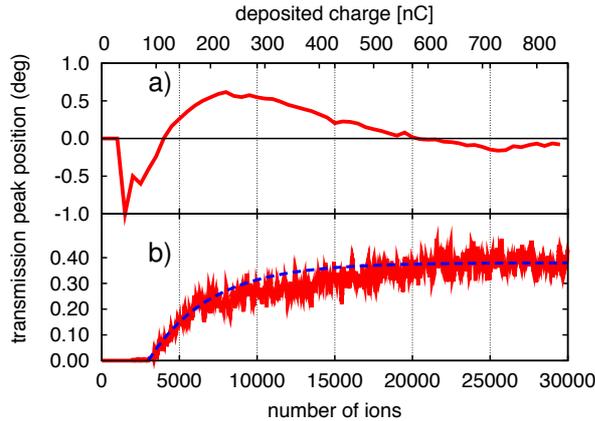
Investigation of the dynamics of HCI guiding prior to equilibrium has recently attracted increasing interest. Already in early experiments direct observation of charging effects, i.e.



**Figure 5.3:** Guiding parameter  $b$  as a function of  $E/q$  of HCI (from Ref. [90]). ■: Experimental data [99, 102]. Simulation (●) employing three models of dielectric shielding are shown (c.f. text), dashed lines serve to guide the eye. The dotted line shows a linear fit to the experimental data.

increase of the transmission rate before equilibrium has been achieved [18]. In particular, it was found that the transmission can be approximately described by  $T(\theta_{in}, t) \approx T(\theta_{in}, 0) \times [1 - \exp(-(t - \tau_s)/\tau_c)]$  for  $t > \tau_s$ .  $\tau_s$  and  $\tau_c$  are the saturation time at which guiding sets in and the charging time constant, respectively.  $T(\theta_{in}, 0)$  represents the equilibrium value for the transmission rate at an angle of incidence of  $\theta_{in}$ . Such a behavior is well reproduced by the CTT simulation for the exemplary case of 3 keV  $\text{Ne}^{7+}$  ions incident under  $\theta_{in} = 3^\circ$  on a capillary with  $2a = 200$  nm (Fig. 5.4 b). The smooth charging characteristics points to a continuous charging process that is finally balanced by beam loss. Expressed in terms of the aggregate charge,  $\int_0^t j_{in} dt'$ , where  $j_{in}$  is the incident current per capillary,  $\tau_c$  is connected to a value of  $\tau_c \approx 4000$  ions. For a beam diameter of about 1 mm this value corresponds to roughly 114 nC of charge deposited on the overall capillary target. Recent experimental work [101] has found a value of about 76 nC for parameters very similar to the ones used here ( $\theta_{in} = 2.8^\circ$  and  $E_{in} = 3.5$  keV), while other measurements (Ref. [100]) find  $\tau_c \approx 100$  nC for parameters identical to the present calculations.

In equilibrium, a *single* principal charge-patch is formed around the impact region near the entrance of the capillary (in the first 10-15% of the capillary) deflecting subsequent projectiles (see Fig. 5.5 and Fig. 5.6). Charge is accumulated near the exit only at later stages, i.e. for a charge deposition on the target  $> 200$  nC. This secondary charge patch is, in fact, rather wide spread. Its center is situated on the side of the capillary that is opposite to the principal charge patch. Figure 5.7 shows the secondary charge-patch near the capillary exit during its formation in time. Note that the color scale has been adapted

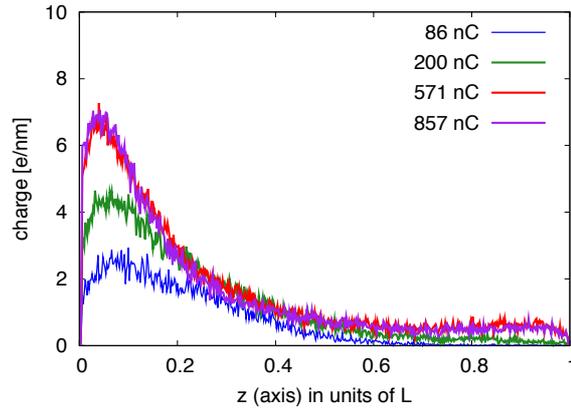


**Figure 5.4:** Time dependence of guiding through a PET capillary with a diameter of 200 nm.  $\text{Ne}^{7+}$  ions are incident at an angle of  $\theta_{in} = 3^\circ$  with  $E = 3$  keV. a) motion of the (one-dimensional) center-of-mass position of the angular distribution of transmitted projectiles. b) transmitted fraction as a function of aggregate incident charge (time). The blue dashed line represents an exponential fit to the transmission curve,  $\tau_c \approx 4000$ .

to the much lower charge deposition as compared to the principal patch (Fig. 5.6).

Early simulations of HCI guiding have already suggested the possibility of forming charge patches additionally to the main charge patch always present. Although the former are weak as compared to the principal patch, they further deflect the trajectories well inside the capillary. Analogously to the principal patch the secondary patch(es) build up in time, too, and during their formation the transmitted beam is thus moving (Fig. 5.4 a)). In equilibrium, however, the beam is collinear with the capillary axis. Additional charge patches do not always fully decay due to charge transport. In contrast, in the long run a weak secondary patch near the exit may support the guiding of projectiles and may be responsible for the beam emerging collinear around the axis.

Theoretical work [26] has very early suggested the existence of two classes of “guided” trajectories - the first class being deflected only once at the primary charge patch as well as a second class containing those that are deflected additionally near the exit. The existence of the second class has been predicted to depend on the geometry of the capillary. Although the time-dependence of transmitted beam profiles has not been explicitly discussed at that time, it is evident that the formation of the second charge patch supporting the second class of trajectories is temporally delayed relative to the build-up of the principal charge patch. Nevertheless, transient behavior of the transmitted beam prior to equilibrium has attracted considerable interest only recently. Several groups have been able to observe the angular motion of the transmitted beam, e.g. in the case of PET [100, 101] as well as for 7 keV  $\text{Ne}^{7+}$  ions guided through  $\text{SiO}_2$  capillaries [23]. Our present

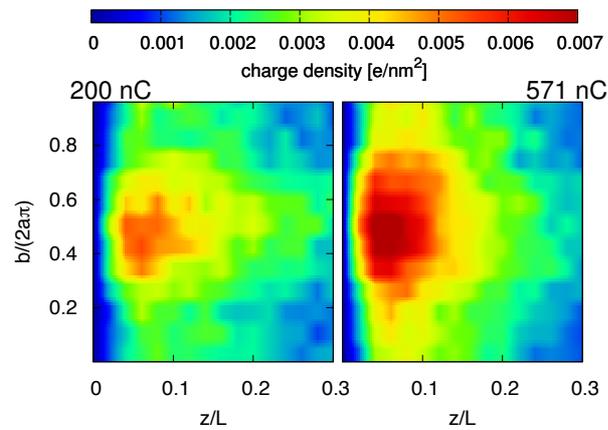


**Figure 5.5:** Charge density along the capillary axis for different amounts of charge deposition, i.e. different moments in time, as indicated in the figure. A single charge patch near the entrance is dominating, at later times some charge also accumulates near the exit.

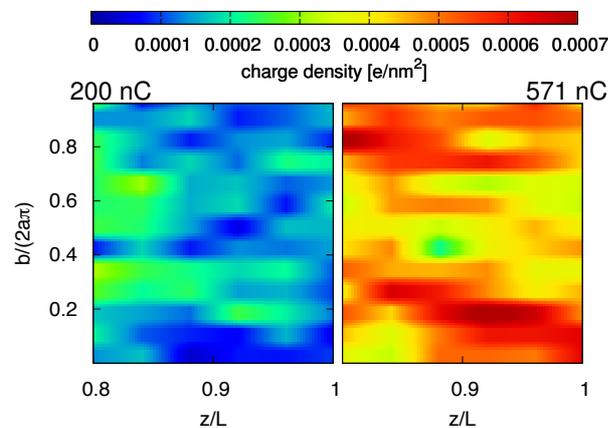
results compare indeed well to the recent experiments of Refs. [100, 101].

Once equilibrium is reached the transmitted beam is stable and centered around the capillary axis. We present two-dimensional angular distribution of transmitted ions in Fig. 5.8. While the time-integral distribution still shows some asymmetry originating from HCI transmitted prior to equilibrium, restriction of the analysis to charge depositions  $\gtrsim 550$  nC (equilibrium) leads to a symmetric transmitted beam.

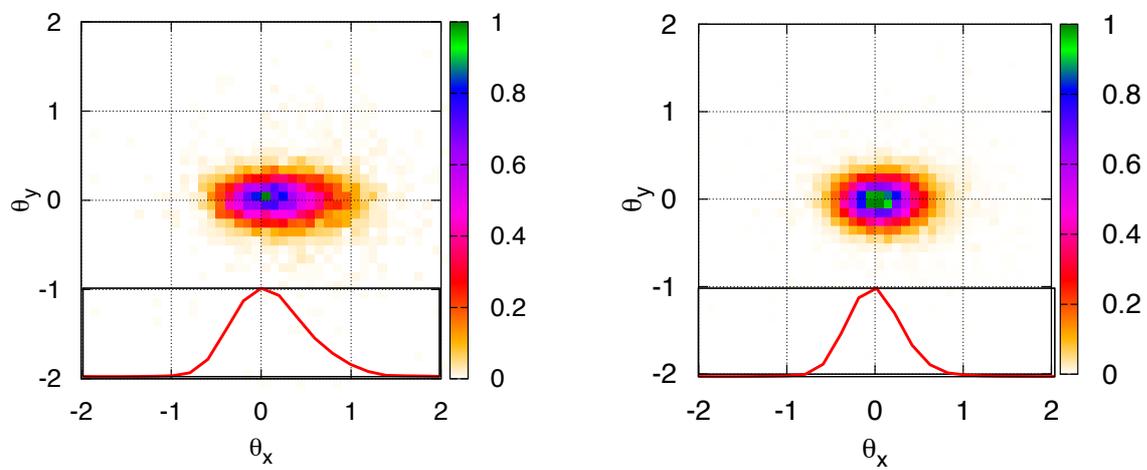
For completeness we note that angular distributions also depend on other system parameters such as e.g. the incidence energy. An extensive analysis of the dependence on energy as well as a discussion of the influence of inter-capillary fields on the observed angular width can be found in Ref. [90].



**Figure 5.6:** Close-up of the charge density on the internal wall near the capillary entrance for two different moments in time (different charge deposition on target) as indicated in the figure. Projectiles impact around a cylinder arclength of  $b/(2\pi a) = 0.5$  which is clearly reflected in the charge density.



**Figure 5.7:** Close-up of the charge density on the internal wall near the capillary exit for two different moments in time (different charge deposition on target) as indicated in the figure. Note the different color scale as compared to Fig. 5.6.



**Figure 5.8:** Two-dimensional angular distribution of transmitted ions, parameters same as in Fig. 5.4. Left panel: time-integral data including transient charge-up. Right panel: time-integral after equilibrium has been reached.



# Chapter 6

## Electron transport in nanocapillaries

Motivated by recent experiments we have extended the CTT simulation described in the previous chapter to the interaction of electron projectiles with an insulator surface. Contrary to HCI, electrons may either scatter off the planar surface potential coherently, or, otherwise, deeply penetrate the bulk material of the internal capillary wall. In the following we therefore introduce the framework of electron transport in a solid. An emphasis is put on the non-conservation of the electron number. Processes like absorption in the solid, re-emission, and secondary electron emission will be explicitly included. The method developed is finally applied to guiding of electrons through insulating nanocapillaries.

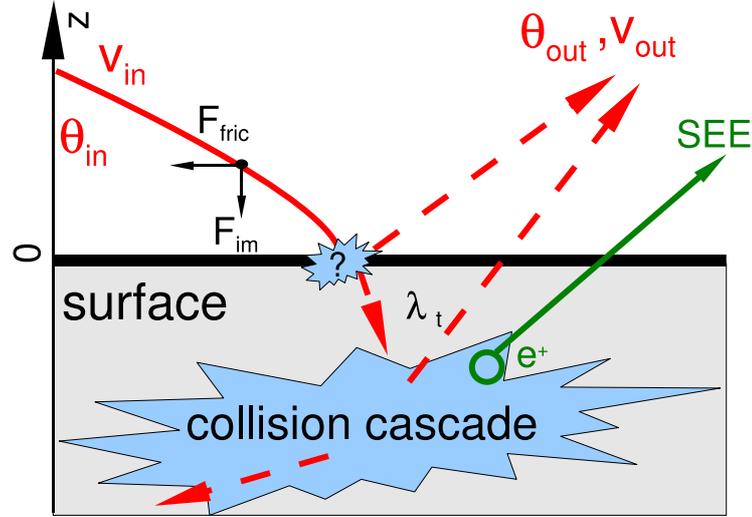
### 6.1 Electron-surface interaction

#### 6.1.1 Interaction scenario with the internal capillary wall

The following scenario (see also schematic view in Fig. 6.1) applies when an electron approaches the surface of the internal wall of a nanocapillary:

Even in the absence of a charged surface, electrons are attracted to the surface by their own image force ( $F_{im}$ ). The latter is, however, much weaker for electrons with the energy considered in this work ( $\gtrsim 200$  eV) than for slow HCI (see e.g. Refs. [103, 104, 105]). Therefore, the image acceleration is presently neglected in first approximation. Within the framework of dielectric response from which the image force is deduced, also a decelerating force parallel to the surface emerges [103, 104]. This so-called “friction force” ( $F_{fric}$ ) is related to the excitation of surface plasmons [106] and thus to the energy loss close above the surface. In contrast to the image force, above-surface energy loss increases with the velocity of the projectile and is accounted for in our approach.

At close contact with the planar surface potential electrons may be reflected coherently in specular direction. Otherwise, they are transmitted into the bulk. In such a case electron transport inside a solid needs to be described. As a result of electron transport



**Figure 6.1:** Scenario of electron-surface interaction with the internal capillary wall.

incident projectiles can be absorbed in the solid or are re-emitted into the vacuum. Both processes may be accompanied by the emission of secondary electrons (SEE) which leaves a positively charged hole behind. The framework of electron transport employed in this work will be outlined in the following section.

### 6.1.2 Electron transport through amorphous materials

Electron transport within the target material is modeled using a classical transport simulation described in detail in [89]. In brief, electrons entering the target material with initial kinetic energy  $E_{in}$  are subject to elastic scattering at constituent atomic potentials and to inelastic scattering processes at the joint electron gas of the compound material (see also Fig. 6.5). The binding potential of the electrons is approximated by a finite-depth well potential, corresponding to the jellium approximation for the electron gas considered. For metals, the electron gas is governed by the conduction band having the width  $E_F$  (Fermi energy) and the distance  $W_F$  (workfunction) to the vacuum. The depth of the potential is thus  $E_F + W_F$ . When applying this approach to insulators the valence band is considered instead of the conduction band and the workfunction needs to be replaced by the gap energy  $E_g$ .

Elastic scattering cross sections are calculated with the ELSEPA package [107]. We use muffin-tin potentials for metallic targets and bare atomic potentials for insulator constituents.  $\text{Al}_2\text{O}_3$  is described as an amorphous compound of 40% aluminum and 60% oxygen atoms, PET (“Mylar”, sum formula  $\text{C}_{10}\text{H}_8\text{O}_4$ ) as a compound of about 45.5% carbon and 18.2% oxygen. The fraction of hydrogen atoms can be safely neglected due to the small elastic cross section of hydrogen as compared to the other components. From

the energy-dependent total cross sections and the fractional densities of constituent atoms the elastic mean free path (EMFP) can be derived. In case of an elastic scattering event the atom type is randomly chosen according to the fractional densities and the scattering angle is determined by the energy-dependent differential cross sections for this atom.

The doubly-differential inelastic scattering mean free path (DD-IMFP) is derived from the momentum and energy dependent dielectric constant of the bulk material  $\varepsilon(q, \omega)$  by the relation [108]

$$\frac{d^2\lambda_{in,b}^{-1}}{dq d\omega} = \frac{1}{\pi E q} \text{Im} \left\{ -\frac{1}{\varepsilon(q, \omega)} \right\} \Theta[\omega_m(q) - \omega]. \quad (6.1)$$

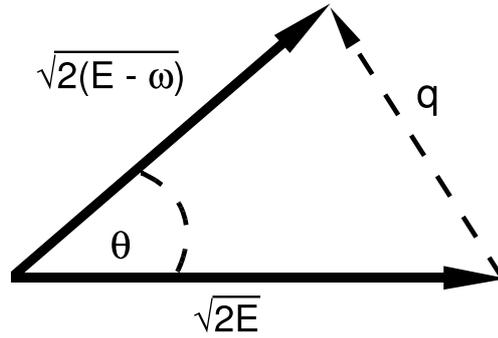
From the DD-IMFP the angular distribution of inelastically scattered electrons is computed by [109]

$$\frac{d\lambda_{in,b}^{-1}}{d\Omega} = \frac{1}{\pi^2} \int \frac{d\omega}{q^2} \sqrt{1 - \frac{\omega}{E}} \text{Im} \left\{ -\frac{1}{\varepsilon(q, \omega)} \right\} \Theta[E - E_f - \omega], \quad (6.2)$$

with  $E = v^2/2$  being the initial energy of the electron in both Eqs. 6.1 and 6.2.  $\omega$  and  $q$  are the energy and momentum transfers, respectively. The step function  $\Theta$  assures energy and momentum conservation in the scattering event with  $\omega_m(q) = \min[v^2/2 - E_F; vq - q^2/2]$ . The relation between the energy transfer  $\omega$ , the momentum transfer  $q$ , and a scattering angle  $\theta$  is established by momentum conservation in the scattering plane (see Fig. 6.2), from which the transformation of the derivative

$$\frac{d}{d\Omega} = \frac{E}{\pi q} \sqrt{1 - \frac{\omega}{E}} \frac{d}{dq} \quad (6.3)$$

can be deduced. In the simulation, differential inelastic scattering mean free paths (D-



**Figure 6.2:** Schematic drawing of inelastic scattering:  $q$  and  $\omega$  represent the momentum and energy transfers, leading to a deflection by the scattering angle  $\theta$ .

IMFP) are used for both energy loss and scattering angle distributions in order to speed up calculations. Therefore, in rare cases a combination of  $\omega$  and  $q$  that does not correspond to energy-momentum conservation for an *individual* scattering event may result. Due to their infrequency such events do not significantly influence the outcome of the simulation. A detailed comparison to a simulation directly employing the DD-IMFP of Eq. 6.1 is still needed. However, the realizations of the singly-differential IMFP are, of course, limited by energy-momentum conservation.

$\varepsilon(q, \omega)$  is constructed from an extrapolation of the optical data [ $\varepsilon_b(q = 0, \omega)$ ] for the capillary material to the  $q$ - $\omega$  plane (e.g., [110, 111]). For PET, data originates from Refs. [112, 113, 114, 115] and for  $\text{Al}_2\text{O}_3$  from Ref. [114]. For more details on method and accuracy of fitting and extrapolation see Sec. B.1. Within this framework the IMFP for PET is  $\lambda_{in,b} \approx 8.5 \text{ \AA}$  at 200 eV, which agrees well with the experimentally found value of  $\lambda_{in} = 8.8 \text{ \AA}$  [113]. For larger energies the IMFP increases and reaches  $15.8 \text{ \AA}$  at  $E = 500 \text{ eV}$ .

Additionally, energy loss due to surface excitation has been included in our simulation as in [89]. They also influence the trajectory of specularly reflected projectiles, especially when incidence angles  $\theta_{in}$  and, consequently, the penetration depth of projectile electrons are small. We use the specular-reflection model introduced by Ritchie *et al.* [116] together with the representation of the surface dielectric function  $\varepsilon_s$  [117]

$$\varepsilon_s(Q, \omega, z) = \frac{Q}{\pi} \int dq_z \frac{e^{iq_z z}}{(Q^2 + q_z^2) \varepsilon(q, \omega)} \quad (6.4)$$

where  $\vec{q} = (Q, q_z)$ . At  $z = 0$ , which is the position of the surface, we have  $\varepsilon_s(Q, \omega, z = 0) = \varepsilon(q, \omega)^{-1}$ . Using  $\varepsilon(q, \omega) \simeq \varepsilon(Q, \omega)$ , i.e. neglecting dispersion along the surface normal, we have  $\varepsilon_s = \varepsilon^{-1} \cdot \exp(-Q|z|)$ , as the integral over  $q_z$  is the Fourier representation of the function  $\exp(-Q|z|)$ . The inverse surface inelastic mean free path  $\lambda_{in,s}^{-1}$  can be then derived from [108]

$$\frac{d^2 \lambda_{in,s}^{-1}}{dQ d\omega} = \frac{e^{-2Q|z|}}{\pi \frac{v_{\parallel}^2}{2} Q \sqrt{1 - \left(\omega + \frac{Q^2}{2}\right)^2 / (Q v_{\parallel})^2}} \text{Im} \left\{ \frac{\varepsilon(Q, \omega) - 1}{\varepsilon(Q, \omega) + 1} \right\} \Theta [\omega_m(Q) - \omega] \quad (6.5)$$

where  $\omega_m(Q) = \min\{v_{\parallel}^2/2 - E_F, v_{\parallel}Q - Q^2/2\}$ . An angular D-IMFP is defined for surfaces with the aid of (cf. Eq. 6.3)

$$\frac{d}{d(\cos \theta)} = \frac{2E_{\parallel}}{Q} \sqrt{1 - \frac{\omega}{E_{\parallel}}} \frac{d}{dQ}. \quad (6.6)$$

Finally, one arrives at

$$\frac{d\lambda_{in,s}^{-1}}{d(\cos \theta)} = \int d\omega \frac{2e^{-2Q|z|} \sqrt{1 - \omega/E_{\parallel}}}{Q^2 \sqrt{1 - \left(\omega + \frac{Q^2}{2}\right)^2 / Q^2 2E_{\parallel}}} \text{Im} \left\{ \frac{\varepsilon(Q, \omega) - 1}{\varepsilon(Q, \omega) + 1} \right\} \Theta [\omega_m(Q) - \omega]. \quad (6.7)$$

Here we have used the relations  $\text{Im} \{(\varepsilon^{-1} - 1)/(\varepsilon^{-1} + 1)\} = -\text{Im} \{(\varepsilon - 1)/(\varepsilon + 1)\}$  and  $E_{\parallel} = v_{\parallel}^2/2$ .

In the implementation of the inelastic surface processes the  $z$ -dependence is explicitly calculated only for a few, usually five, values. For reasons of numerical efficiency we rather fit  $\lambda_{in,s}(E, z)/\lambda_{in,s}(E, 0)$  by the function  $f(z, E) = 1 + (a_1 - a_2 E) \cdot |z|^{(b_1 - b_2 \log E)}$  [89] and explicitly employ only  $\lambda_{in,s}(E, 0) \times f(z, E)$  with pre-determined values for the fit parameters  $a_1, a_2, b_1, b_2$  (see Sec. B.2 for more details). Note that at a given energy the  $z$ -dependence of the inverse surface IMFP is also well fitted by an exponential,  $\lambda_{in,s}^{-1}(z) = \lambda_{in,s}^{-1}(0) \exp(-z/z_s(E))$ , as long as  $z > 0$ .

### Transport and ratio of mean free paths

During transport through the solid the total mean free path of the electron transported is determined by

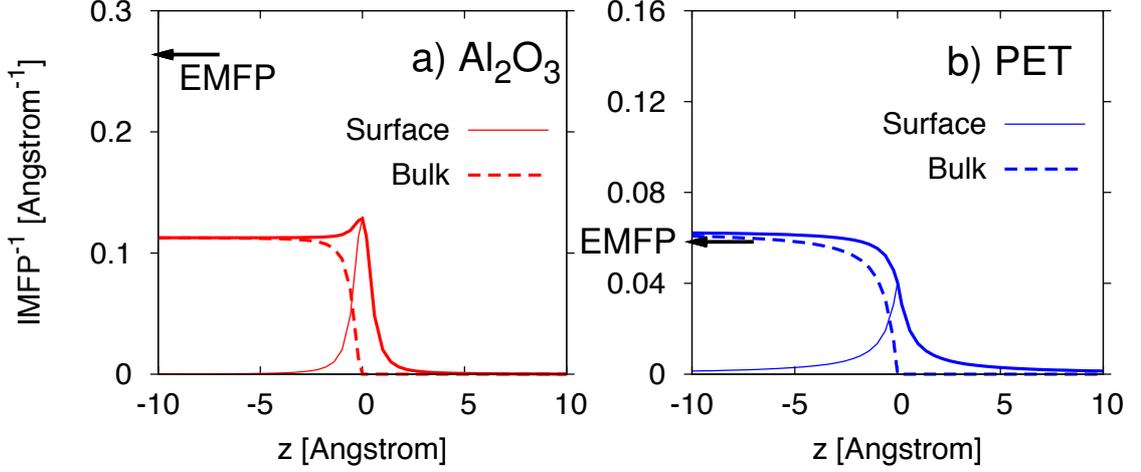
$$\frac{1}{\lambda_{tot}} = \frac{1}{\lambda_{el}} + \left( \frac{\theta(-z)}{\lambda_{in,b} \cdot (1 - f(z, E))} + \frac{1}{\lambda_{in,s}(z=0) \cdot f(z, E)} \right), \quad (6.8)$$

with the last term in brackets being the total inelastic mean free path  $\lambda_{in}(z)$ . Employing the switching function  $f(z, E)$  in Eq. 6.8 ensures the correct limit of having only bulk loss well inside the bulk [108],  $\lambda_{in}(z \rightarrow -\infty) = \lambda_{in,b}$ . At the same time, the  $\theta$ -function assures  $\lambda_{in}(z > 0) = \lambda_{in,s}$ .

The ratio  $\lambda_{tot}/\lambda_{el}$  (or  $\lambda_{tot}/\lambda_{in}$ ) represents the probability for an (in-)elastic scattering process to happen. Consequently, inspection of the ratio  $\lambda_{el}/\lambda_{in}$  teaches us about the relative importance of elastic scattering as compared to inelastic processes and can serve as a guideline of what to expect in the overall outcome of the electron transport simulation. Figure 6.3 shows the dependence of the total IMFP as well as the IMFP attributed to surface and bulk loss on the distance  $z$  to the surface. Note that for  $z \rightarrow -\infty$  the total IMFP converges to bulk loss. The extension of surface loss into the region  $z > 0$  (drop to a fraction of  $e^{-1}$ ) is about 0.7 to 1 Å. For comparison the total EMFP for both materials is given. The ratio  $\lambda_{el}/\lambda_{in}$  is evidently more than twice as large for Al<sub>2</sub>O<sub>3</sub> as compared to PET which indicates that inelastic processes are much more prevalent for PET at the energy chosen.

### Energy loss above the surface

Above the surface only surface loss is present. So far, we approximate the trajectory of the projectile near the surface by a straight-line trajectory and thus neglect image acceleration. The path of the projectile close to the surface ( $z \lesssim z_s$ ) may then be approximated by  $s \approx 2z_s/\sin\theta_{in}$ . The quantity  $z_s$  is determined by the distance at which the surface IMFP has grown by a factor of  $e$ , which implies that the inverse IMFP is described by  $\lambda_{in,s}^{-1}(z) = \lambda_{in,s}^{-1}(0) \exp(-z/z_s)$ . Along the path  $s$  we employ a suitable mean surface IMFP



**Figure 6.3:** Inverse inelastic mean free path (IMFP) for a)  $\text{Al}_2\text{O}_3$  at 250 eV and b) PET at 500 eV as a function of the distance  $z$  to the surface. Contributions of surface (solid) and bulk (dashed) loss are explicitly shown. For  $z \rightarrow -\infty$  the total IMFP (bold solid) converges to bulk loss. For comparison the total EMFP of both materials is marked in each figure by an arrow.

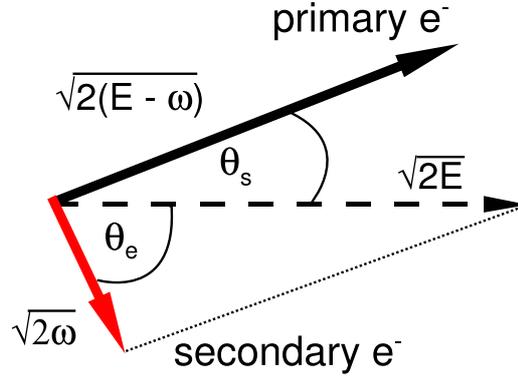
$\lambda_{in,s}(z_m)$  deduced from the relation  $\exp(-z_m/z_s) = 1/z_s \cdot \int_0^{z_s} \exp(-z/z_s) dz$ . After inelastic scattering energy and momentum are updated accordingly.

### Electron emission and absorption

In any inelastic scattering process, the lost energy  $\Delta E$  is eventually transferred to a secondary electron starting at the position of the primary electron in the case of bulk and surface loss, or with coordinate  $z = 0$  for surface loss above the surface. The angular emission pattern of secondary electrons is not trivial and may strongly depend on the exact type of the physical excitation that leads to secondary electron emission (SEE). However, the only significantly strong process to obtain large SEE energies of the order of several ten eV is a binary collision of fast projectiles with bound electrons. The angle of emission  $\theta_e$  is thus chosen from the inelastic scattering angle  $\theta_s$  according to the energy-momentum conservation in a binary collision (see schematic view in Fig. 6.4),

$$\theta_e = \text{asin}(\sqrt{(E - \Delta E)/E} \sin \theta_s). \quad (6.9)$$

This requires, however, that already  $\Delta E$  and  $\Delta\theta_s$  have been chosen accordingly.  $\Delta E$  and  $\Delta\theta_s$  are obtained from the two individual D-IMFP used to speed up calculations, i.e.  $d\lambda_{in,b}^{-1}/d(\Delta E)$  following from an integration of Eq. 6.1 over  $q$  and the expression in Eq. 6.2. These D-IMFP also represent other inelastic processes, therefore a combination of



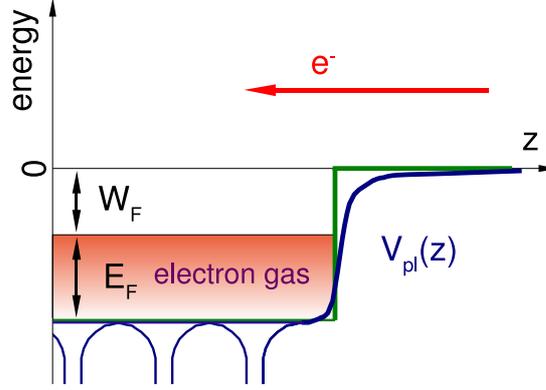
**Figure 6.4:** Schematic view of secondary electron emission after inelastic scattering.

$\Delta E$  and  $\Delta\theta_s$  incompatible with a binary collision may result. In these rare cases,  $\theta_e$  is chosen from an isotropic distribution instead.

Between subsequent scattering processes electrons are propagated along a straight-line trajectory. If an electron reaches the surface of the target it needs to have sufficient energy in the direction of the surface normal ( $E_{\perp} > W_F$ ) in order to overcome the surface potential and to be emitted into the vacuum. The electron is specularly reflected into the solid otherwise. If the total energy of the electron is too low to allow for emission into vacuum the trajectory is stopped and the electron is regarded to be absorbed by the solid. Note that trajectories of secondary electrons are followed as well and may contribute to the total spectrum of electrons emitted. Depending on the net number of emitted electrons the secondary electron emission coefficient  $\delta$  can be larger or smaller than one, resulting in a charge-up of the sample that is either positive or negative.

### 6.1.3 Coherent surface reflection

The key novel ingredient to establish guided transmission of electrons is the glancing scattering at the planar-averaged surface potential  $V_{pl}(z)$  of the capillary wall without penetrating into the bulk. This quantum reflection due to the attractive surface potential is completely absent in a truly classical simulation as it lacks coherent scattering at the surface, i.e. scattering at the *joint* potential of many atoms sitting in the surface layer(s). Such an effect can be phenomenologically included into the simulation as a stochastic process. The elastic specular reflection probability  $P_s$  and momentum transfers  $\Delta p_i = 2k_{\perp}$  are determined for  $V_{pl}(z)$  approximated alternatively by density functional theory (DFT) calculations of the target material and by a step function of the same height (i.e. of height  $V_{pl} \approx W_F + E_F$ , see also the schematic cartoon in Fig. 6.5). For DFT calculations the program package “ABINIT” [118] is employed. We find that  $P_s$  is generally larger for



**Figure 6.5:** Schematic representation of (approximated) surface potentials and energy structure of the solid surface considered: We described the electronic structure of the solid as a single band of width  $E_F$  (for insulators the width of the valence band is used). The depth of the potential well for  $z \rightarrow -\infty$  is  $V_{pl} \approx W_F + E_F$ . For insulators the workfunction  $W_F$  is replaced by the gap energy  $E_g$ .

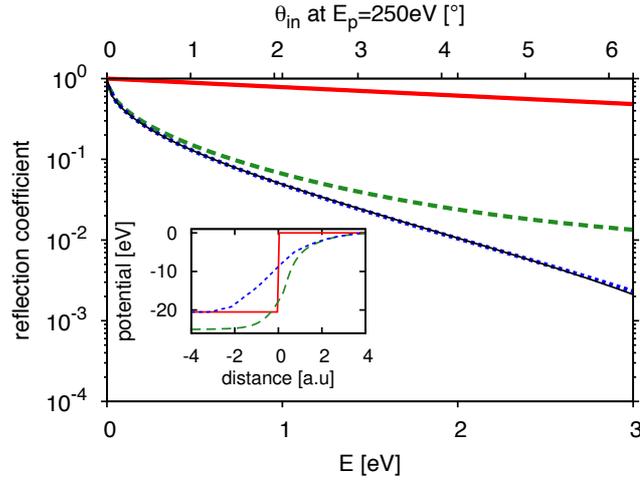
insulators than for metals as their surface potential is steeper (less electron spill-out).

For the step potential,  $P_s$  can be given analytically by

$$P_{s,step} = \left| \frac{k_1 - k_2}{k_1 + k_2} \right|^2 \quad (6.10)$$

where  $k_1 = k_{\perp} = \sqrt{2E_{\perp}}$  and  $k_2 = \sqrt{2(E_{\perp} + V_{pl})}$ . For a general,  $z$ -dependent potential,  $P_s$  is computed numerically by a standard Runge-Kutta integration of the one-dimensional stationary Schrödinger equation. As an example, calculated values for  $P_s$  are shown in Fig. 6.6 as a function of  $E_{\perp}$  and  $\theta_{in}$  (incidence angle with respect to the surface assuming  $E_{in} = 250$  eV). A crystalline structure with a  $\text{CH}_2$  group at the PET surface was assumed [119]. For aluminum oxide, a the crystal structure of corundum (“ $\alpha\text{-Al}_2\text{O}_3$ ”) was assumed [120]. To a good degree of approximation,  $P_s(E_{\perp})$  can be fitted by,  $P_s(E_{\perp}) = \exp(-b \cdot \text{asin}(\sqrt{a \cdot E}))$  with  $a = 0.005$ ,  $b = 33.3$  for PET and  $a = 0.268$ ,  $b = 5.54$  for  $\text{Al}_2\text{O}_3$ . In the limit  $E_{\perp} \rightarrow 0$ ,  $P_s$  converges to unity.

For a more realistic description of specular reflection one needs to consider the limitations of coherence in projectile-surface scattering. Small changes of the amount of momentum transferred from the surface to the projectile (due to e.g. phononic vibrations of the surface atoms or electron-electron scattering) at different positions along the surface lead to a reduction of the width of the coherent wave packet after scattering. The resulting wave packet width is then related to the average momentum transfer and to the elongation of the illuminated surface and does not significantly depend on the variations of the momentum transfer [121]. In a first attempt we account for the finite coherence of



**Figure 6.6:** Specular reflection coefficient  $R_s$  as a function of  $E_{\perp}$  and (assuming  $E_{in} = 250$  eV)  $\theta_{in}$  for different types of surface potentials (inset). Solid red line: step-potential with height 20.5 eV (adapted to the electronic structure of  $\text{Al}_2\text{O}_3$ ), dashed green line:  $V_{pl}^{(DFT)}(z)$  for PET [119], dotted blue line: same for  $\text{Al}_2\text{O}_3$  [120]. The black solid line represents a fit parameterizing the reflection coefficient (cf. text).

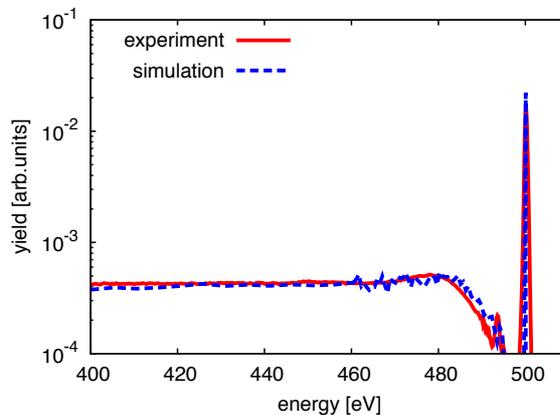
the outgoing wavepacket on a phenomenological basis. We allow for a Gaussian distribution around the specular reflection value  $\theta_{out}$  with a FWHM of  $0.1 \cdot \theta_{out}$  which corresponds to results obtained recently by a full treatment of the problem of [121].

## 6.2 Instantaneous electron transmission

Our approach to guided electron transmission through insulating nanocapillaries allows, in principle, for transmission of electrons even without time delay due to charge-up. By scattering at the surface or electron emission inside the bulk (either “re-emission” or SEE) projectiles can be transmitted for angles of incidence  $\theta_{in}$  larger than the geometric opening angle. Moreover, as for most parameters and target materials we observe a SEE coefficient of  $\delta > 1$  charge-up of the capillary internal wall is expected to be of opposite - positive - polarity. Consequently, charge-up dynamics will suppress guided transmission of projectiles rather than enhance it. The limiting case of vanishing charge-up, which represents also the limit of a “fresh” capillary ( $t \rightarrow 0$ ), is thus an interesting case in point. Furthermore, computational complexity is much reduced for such a time-independent analysis. We will therefore start the investigation of guided electron transmission by neglecting the dynamical charge-up. In a first attempt we even neglect the velocity-dependent image force in order to arrive at a simple and easy-to-solve model. As a test case scattering at a single planar surface is considered as well.

### 6.2.1 Planar surface scattering as test case

Experimental data available for electron impact on a planar PET surface [113] provide a test for the reliability of the collision kernel. Electrons with kinetic energies of 500 eV were directed on the target surface under an angle of incidence  $\theta_{in} = 40^\circ$  with respect to the surface. The measured spectrum of scattered electrons (Fig. 6.7) agrees remarkably

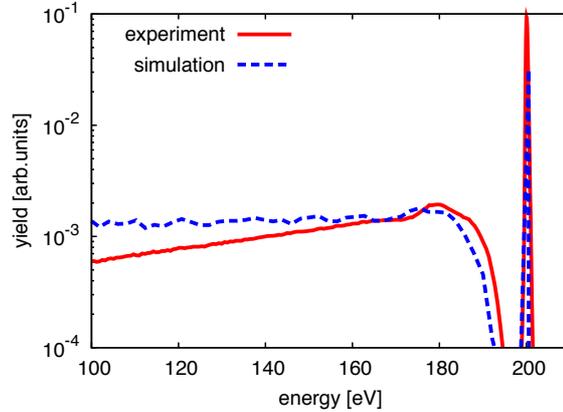


**Figure 6.7:** Energy spectrum for 500 eV electrons incident on PET under  $\theta_{in} = 40^\circ$  with respect to the planar surface. The solid line shows experimental data [113], the dashed line results from our electron transport simulation [29].

well with our simulation thus lending credence to our treatment of multiple elastic and

inelastic scattering as well as the collision cascades of penetrating trajectories.

Similar good agreement is found in case of a planar  $\text{Al}_2\text{O}_3$  surface with 200 eV electrons incident under  $40^\circ$  with respect to the surface (Fig. 6.8). These encouraging results



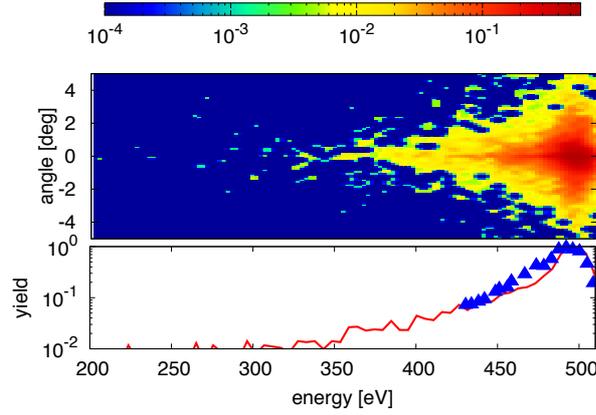
**Figure 6.8:** Energy spectrum for 200 eV electrons incident on  $\text{Al}_2\text{O}_3$  under  $\theta_{in} = 40^\circ$  with respect to the surface. The solid line shows experimental data [113], the dashed line results from our electron transport simulation.

suggest that our present method is well capable of describing electron-surface scattering at a surface of an insulator, as needed for a proper treatment of guided transmission through nanocapillaries.

### 6.2.2 PET capillaries

As a first example we investigate PET capillaries ( $\varepsilon_r = 3.3$ ) with a width  $2a = 200$  nm and length  $L = 10 \mu\text{m}$ . Electrons with an incidence energy of 500 eV (energy spread of about 20 eV) are considered. This system has recently been studied both experimentally by Das *et al.* [28] as well as theoretically [29]. In a first approach we neglect, apart from charge-up, also the image acceleration of the projectiles. These two approximations tremendously facilitate the computation as projectile trajectories between subsequent scattering events are simply straight lines ( $\vec{F} = 0$  in Eq. 5.1). As we will see below, such an approach is sufficient for describing many features of guided electron transmission, both qualitatively and, to some extent, also quantitatively. In the limit of a “fresh” capillary ( $t \rightarrow 0$ ), where charge-up can not yet be present, the approximation reduces to the neglected image force only. Because it is not yet clear whether surface excitations do indeed exist for a polymer like PET, we so far employ only bulk loss in the present example.

Within this approach, we observe that both elastic and inelastic contributions emerge near  $\theta_{out} = 0^\circ$  and are thus “guided” (see two-dimensional distribution of the exit angle and final energy of the projectiles in Fig. 6.9). However, the term “guiding” may be, in



**Figure 6.9:** Normalized energy spectrum (lower panel) as well as a distribution of exit angles and final energy ( $\theta_{out}, E$ ) (upper panel) for transmitted electrons incident with 500 eV under  $\theta_{in} = 3^\circ$  with respect to a PET capillary's axis. The energy and angular spread of the incident beam are 20 eV and  $1^\circ$  FWHM. Solid blue triangles show experimental data [28], solid lines result from our electron transport simulation without charge-up and image force.

fact, misleading as projectiles do indeed interact with the inner wall of the capillary. For  $\theta_{in} = 3^\circ$  most projectiles that are eventually transmitted are subject to an average of 2.5 scattering events upon collision with the internal wall. At least one of those scattering events (on average 1.7) is a specular reflection at the surface without penetration into the bulk material. Similar numbers (an average of about 2 collisions) apply for projectiles transmitted strictly elastically. Although small-angle atomic scattering in the bulk allows for elastic “guiding”, in principle, the transmission rate would be much weaker if elastic specular reflections at the joint surface potential are neglected. Due to its favorable angular emission pattern this type of scattering plays an important role even though such collisions represent only a fraction of the total number of collisions.

### 6.2.3 $\text{Al}_2\text{O}_3$ capillaries

Experiments with  $\text{Al}_2\text{O}_3$  ( $\epsilon_r = 9.5$ ) as target material have been the first to be carried out with electronic projectiles [27], although only quasi-elastic transmission was observed initially. Recently, inelastic guiding has also been demonstrated for  $\text{Al}_2\text{O}_3$  nanocapillaries with  $2a = 270$  nm and  $L = 15 \mu\text{m}$  [122]. The pronounced energy gap of  $E_g = 8.7$  eV [120, 123] may have been the reason for this delayed discovery. Due to the gap only energy loss larger than  $E_g$  is allowed which is reflected in the energy spectra for both planar surface scattering (Fig. 6.8) as well as for transmission through  $\text{Al}_2\text{O}_3$  capillaries (Fig. 6.10 a). Note that for  $\text{Al}_2\text{O}_3$  surface excitations are experimentally well established [124] and are

# Chapter 7

## Summary and outlook

In this thesis we have investigated selected aspects of how charged particles can be controlled on short time and length scales. A prominent example for steering of electronic motion on atomic time scales is ultrafast laser-atom interaction (“ultrafast optics”). The techniques developed in this field bring e.g. control of molecular reactions within reach.

Within the wide field of ultrafast optics, we have focused on the properties of the coherent, high-frequency radiation generated during the interaction of ultrashort, strong laser pulses with atoms, putting an emphasis on the dependence on the wavelength of the driving laser. In particular, we have investigated the mid-infrared range, which is experimentally accessible for a short time only. Employing an *ab-initio* approach, i.e. by solving the time-dependent Schrödinger equation for a single (the most weakly bound) electron, we have shown that both the high-harmonic generation (HHG) yield as well as the ionization probability feature surprisingly regular enhancements as a function of both driver intensity and wavelength of the driver. Enhancements can be as large as a factor of eight. The present work may thus serve as a basis for further optimization and fine-tuning of the HHG output. On a wavelength scale the modulation period can be considerably smaller than the Fourier width of the ultrashort pulses employed. The modulation can be related to constructive interference of electronic quantum paths with long (i.e. longer than an optical cycle of the driver) excursion times that contribute to the HHG yield [36]. Such quantum paths correspond to (semi-)classical trajectories which can be investigated more easily.

Although the length of the pulses investigated only range from a single to a few optical cycles, the present modulation is related to the well-known channel closing effect derived for infinitely extended laser fields (see e.g. [86]). Despite this similarity, we find significant differences: enhancements are shifted from positions predicted by an analysis of channel closings for zero-range potentials. This discrepancy has been discussed in previous literature and a variety of explanations has been offered. We have shown that this effect is related to the long-ranged Coulomb potential affecting the phase of the electronic wavepacket during free evolution. Numerical data obtained over a wide range

of driving pulse parameters support this findings [37].

Apart from the fine-scale oscillations of the HHG yield we have also analyzed the scaling of the yield in a fixed energy interval of 20 to 50 eV over a wide range of driving laser wavelengths  $\lambda_c$ . In agreement with previous work [35] we arrive at a scaling  $\propto \lambda_c^{-x}$  and an exponent of  $x \approx 5.5 \pm 0.5$  for the HHG yield of argon within the framework of single-atom response. Very recently, experiments with neon have reported results close to the present value [75], while theoretical work focusing on the HHG yield near the (wavelength-dependent) cut-off finds significantly lower values of  $x$  for rare gas atoms [78]. Further investigation will be necessary in this context.

Oscillations in the ionization channel being very similar to those in the HHG yield are observed as well. They, too, can be related to path interference and are found for both the total ionization probability as well as in the energy-resolved photoelectron spectrum. It is straightforward to identify semi-classical trajectories leading to such interference oscillations. However, the weight of the contribution of a particular trajectory to the total ionization yield still needs to be investigated in future work.

In ultrafast laser-atom interaction, as discussed in this work, the dynamics of the electrons (and nuclei) are governed by the external field of a coherent laser pulse with a duration in the femtosecond range. Although the time scales involved are extraordinarily short, this is conceptionally not so different from conventional particle optics where charged projectiles are steered by electromagnetic fields. By contrast, we have devoted a large portion of our research to a phenomenon that allows for mesoscopic control over highly-charged ions (HCI) without relying on external electromagnetic fields. This tool has come within reach after the first observation and subsequent further investigation of HCI guiding through insulating nanocapillaries. The guiding effect becomes manifest in HCI projectiles being transmitted through a capillary in the initial charge state without close interaction with the internal wall. For the description of this process, we have presented a Classical Transport Theory (see e.g. Ref. [26]) which uses order-of-magnitude estimates of material data and requires no additional free parameters or ad-hoc assumptions. Our self-consistent projectile trajectory simulations for PET nanocapillaries have confirmed phenomenological models of HCI guiding based on electrostatic deflection of projectiles and agree well with the experiment. The present approach works well for incident currents common in many experiments (typically 0.1-1 nA/mm<sup>2</sup>) but will result in either Coulomb blockade or insufficient charge-up as soon as the current is varied over several orders of magnitudes. The range of incident currents leading to stable projectile transmission is related to the conductivity of the material employed and may thus vary for different capillary targets.

An investigation of guided transmission of electronic projectiles through nanocapillaries has been performed as well. Here, we account for the interaction of electrons with a solid where penetration is accompanied by scattering, energy loss, absorption of the projectile and/or emission of secondary electrons [29]. Due to the clearly different inter-

action process transmission properties for electrons as projectiles are distinct from what is observed with HCI. In this context guided transmission is enabled by small-angle scattering at and below the surface of the internal wall and charge-up is found to be of minor importance. Transmitted projectiles suffer considerable energy loss. Our approach thus offers a first explanation for recent experimental results [28, 122].

Charge-up is found to be significantly different from HCI guiding. Its influence being frequently negligible for larger tilt angles, charge-up suppresses transmission through an untilted nanocapillary. This effect can be attributed to the attraction of projectiles towards the internal wall after a positively charged patch on the internal wall (caused by secondary electron emission) has developed. Above-surface loss is found to be crucial for a full description of transmission through untilted capillaries because it represents the dominant inelastic process for  $\theta_{in} \rightarrow 0^\circ$ . At larger angles of incidence this inelastic process is rather unimportant. Irrespective of the projectile, the time structure of charged particle guiding is linked to the charge-up. Therefore, observables such as the transmission rate evolve on the same time scale, being of the order of minutes. When charge-up is negligible even a time-independent approach is sufficient.

Future developments in the theoretical description of electron guiding may include an in-depth analysis of the interplay between scattering and charge-up, which will strongly depend on the material properties. For that purpose structural properties of the surface and bulk material (e.g. structure on the surface) need to be known in more detail. In this context also the influence of surface adsorbates and contamination should be considered. Both may alter the (differential) scattering probabilities for electron-surface interaction. Electrical (surface) conductivity may deviate from clean materials and thus change the characteristics of charge-up.

Furthermore, dielectric response of the surface to an approaching charged projectile needs to be considered more profoundly in order to extend present results for electron guiding that have been obtained without accounting for image forces. For fast projectiles such as electrons with the energies considered, the image acceleration towards the surface is indeed much weaker as compared to slow HCI. In first approximation, it can be neglected for the incident projectiles. For slow secondary electrons created during inelastic projectile-surface interaction, however, the image force is much stronger and approaches its static limit. Explicitly accounting for the velocity dependence of dielectric response would lead to an improved description of guided transmission. Due to the present approach of neglecting image acceleration, the length of grazing-incidence trajectories and thus the above-surface energy loss (“friction force”) may currently be overestimated. In future work the friction force exerted on a fast electron should therefore be included on the same level of approximation as the image acceleration.

At the same time, the effect of the image potential needs to be treated within the framework of coherent quantum scattering at the surface. Close to the surface, classical

theories for the dielectric response are not sufficient and lead to unphysical divergences. An *ab-initio* approach (e.g. a time-dependent density functional theory calculation) for the system consisting of a charged projectile approaching a surface may be advantageous and allow for the determination of the correct, velocity-dependent limit of the image potential when crossing the surface. This quantity is responsible for the impact angle of the projectile. Contrary to the case of HCI projectiles, where trajectories are stopped upon impact, the impact angle of electronic projectiles is crucial for determining the subsequent scattering characteristics.

# Appendix A

## A.1 Fourier transform on a bounded domain

For numerical calculations, a real-valued function  $f(t)$  is usually defined only in a bounded domain, i.e.  $t \in [0, T_m]$ ,  $T_m$  is the largest time represented on the mesh. The total pulse length is defined by  $T$ , which can be smaller than  $T_m$ . We define the Fourier transform of the function  $f(t)$  denoted by  $\mathcal{F}(f(t)) = \tilde{f}(\omega)$  as following:

$$\tilde{f}(\omega) = \frac{1}{\sqrt{2\pi}} \int_0^T f(t) \exp(-i\omega t) dt \quad (\text{A.1})$$

Note that  $\tilde{f}(\omega)$  carries the dimension of  $f(t)$  and the dimension of time. The original function can be retrieved by the inverse transform  $\mathcal{F}^{-1}(\tilde{f}(\omega)) = f(t)$  by

$$f(t) = \frac{2}{\sqrt{2\pi}} \int_0^\infty \tilde{f}(\omega) \exp(+i\omega t) d\omega \quad (\text{A.2})$$

which sums only over positive frequencies. The zero frequency component is a measure the average value of  $f(t)$  in the given domain. For brevity, sometimes the tilde may be dropped ( $\tilde{f}(\omega) = f(\omega)$ ). A frequently used property of the Fourier transform is  $\dot{f}(\omega) = -i\omega \times f(\omega)$  where  $\dot{f}$  denotes the derivative of  $f(t)$  with respect to time.

On a numerical, discrete mesh with  $N$  points and  $T_m/N = \Delta t$ , we have  $t = j\Delta t$

$$\tilde{f}(\omega_k) = \frac{T_m}{N\sqrt{2\pi}} \left[ \sum_j f(j\Delta t) \exp\left(-i\frac{2\pi}{N}j \cdot k\right) \right]. \quad (\text{A.3})$$

Here,  $\omega_k = 2\pi/T_m \cdot k$ . This integral can be efficiently computed by the Fast Fourier Transform algorithm [63]. Note that  $|\tilde{f}(\omega)|^2 d\omega$  becomes numerically

$$|\tilde{f}(\omega)|^2 d\omega = \frac{T_m^2}{N^2 2\pi T_m} \left[ \sum \right]^2 = \frac{T_m}{N^2} \left[ \sum \right]^2 \quad (\text{A.4})$$

when accounting for the dimension carried by the physical quantities. It has the dimension of a signal energy (not power!), and is linearly increasing with the signal length.

## A.2 Definition of radiated power

The radiated energy per unit time (power), averaged over one or more radiation periods, of a Hertz dipole  $d(t)$  moving sinusoidally ( $d_\omega(t) = d_{0,\omega} \sin(\omega t)$ ) is given by [43]

$$P_\omega = \frac{2}{3c^3 T} \int_0^T \ddot{d}_\omega(t)^2 dt \quad (\text{A.5})$$

$$P_\omega = \frac{\omega^4}{3c^3} d_{0,\omega}^2 = \frac{1}{3c^3} a_{0,\omega}^2 \quad (\text{A.6})$$

Because the dipole acceleration  $a(t)$  is given  $\ddot{d}_\omega(t) = a_\omega(t)$ , we have used the relation  $d_{0,\omega} = a_{0,\omega}/\omega^2$ .

For arbitrary short pulses we can now start with Eq. A.6, employ a priorly spectrally filtered acceleration  $a_f(t)$  (having Fourier components only between  $\omega_{lo}$  and  $\omega_{up}$ ) to write the yield  $Y$  of these components

$$Y = \frac{2}{3c^3 T} \int_0^T a_f(t)^2 dt = \frac{2}{3c^3 T} \int_{-\infty}^{\infty} a_f(t)^2 dt, \quad (\text{A.7})$$

because  $a_f(t) = 0$  outside  $t \in [0, T]$ . We recall also Parseval's theorem of Fourier theory

$$\int_{-\infty}^{\infty} f(t)^2 dt = \int_{-\infty}^{\infty} \tilde{f}(\omega)^2 d\omega \quad (\text{A.8})$$

which states ‘‘conservation of signal energy’’ when changing from the time to the frequency domain. Employing the above theorem we arrive at

$$Y = \frac{2}{3c^3 T} \int_{\omega_{lo}}^{\omega_{up}} |\tilde{a}(\omega)|^2 d\omega. \quad (\text{A.9})$$

This expression still has the dimension of energy per unit time, as requested.

## A.3 Definition of pulse intensity

When the function  $f(t)$  is an electric field  $F(t)$ , Parseval's theorem (Eq. A.8) has indeed the dimension of an energy (per unit area). Hence, pulse energy per unit area is

$$\frac{E_{pulse}}{A} = \int_{-\infty}^{\infty} F(t)^2 dt = \int_{-\infty}^{\infty} |\tilde{F}(\omega)|^2 d\omega = 2 \int_0^{\infty} |\tilde{F}(\omega)|^2 d\omega \quad (\text{A.10})$$

Negative and positive frequencies are equivalent. Average (over the total pulse length  $T$ ) pulse power per unit area (i.e. *intensity*) would be (compare to Eq. A.9)

$$I_{av} \equiv \frac{1}{T} \int_{-\infty}^{\infty} F(t)^2 dt = \frac{2}{T} \int_0^{\infty} |\tilde{F}(\omega)|^2 d\omega \quad (\text{A.11})$$

Peak pulse power per unit area ( $I_0$ ) assumes a sinusoidal field with constant amplitude  $F_0$ , which would lead to  $I_0 = F_0^2/2$ . However, in the system of atomic units, the intensity is *defined* disregarding the factor 1/2, reading

$$I_0 \equiv F_0^2. \quad (\text{A.12})$$

## A.4 Far-field transformation

After propagation through a medium of length  $L$  the electric field  $F(L, x_\perp, \omega)$  can be computed numerically. Integration over the transverse coordinate and Fourier transform yields the near-field  $F_{near}(L, t)$ .

The far-field at distance  $d$  under the angle of observation  $\beta$   $F_{far}(L + d, \beta)$  accounts for the propagation up to the observation point. It reads [43]

$$F_{far}(L + d, \beta, \omega) = i \frac{k}{d} \int d^2 x_\perp F''(x_3, x_\perp, \omega) e^{i \frac{x_\perp^2 k}{2d}} J_0(x_\perp \beta k) \quad (\text{A.13})$$

with  $k = \omega/c$ . Fourier transform of the above expression yields  $F_{far}(L + d, \beta, t)$ .



# Appendix B

## B.1 Fitting and extrapolating optical data

For obtaining the loss function  $\text{Im}(-1/\epsilon(q, \omega))$  we need an analytical ansatz for the extrapolation of experimental data. The most simple ansatz is one or more damped plasmon pole(s) with frequency  $\omega_p$  and width  $\gamma$ , reading  $\epsilon(\omega) = \epsilon_1 + i\epsilon_2 = 1 - \omega_p^2/(\omega^2 + i\omega\gamma)$ . Remembering  $\epsilon_s = \epsilon^{-1}$  this leads to

$$\text{Im}(g(\omega)) = \text{Im}\left(\frac{\epsilon_s - 1}{\epsilon_s + 1}\right) = -\text{Im}\left(\frac{\epsilon - 1}{\epsilon + 1}\right) = (-)\frac{2\epsilon_2}{(\epsilon_1 + 1)^2 + \epsilon_2^2} \quad (\text{B.1})$$

$$\epsilon_1 = \frac{(\omega^2 - \omega_p^2)\omega^2 + \omega^2\gamma^2}{(\omega^4 + \omega^2\gamma^2)} \quad \text{and} \quad \epsilon_2 = \frac{\omega_p^2\omega\gamma}{(\omega^4 + \omega^2\gamma^2)} \rightarrow \quad (\text{B.2})$$

$$\text{Im}(g(\omega)) = \frac{2\omega_p^2\omega\gamma}{(\omega_p^2 - 2\omega^2)^2 + 4\omega^2\gamma^2} = \frac{\omega_s^2\omega\gamma}{(\omega_s^2 - \omega^2)^2 + \omega^2\gamma^2} \quad (\text{B.3})$$

$$(\text{B.4})$$

with  $\omega_s = \omega_p/\sqrt{2}$ . The real part reads (using Eqs. B.2)

$$\text{Re}\left(\frac{\epsilon(\omega) - 1}{\epsilon(\omega) + 1}\right) = \frac{(\epsilon_1^2 - 1) + \epsilon_2^2}{(\epsilon_1 + 1)^2 + \epsilon_2^2} = \frac{\omega_s^2(\omega_s^2 - \omega^2)}{(\omega_s^2 - \omega^2)^2 + \omega^2\gamma^2} \quad (\text{B.5})$$

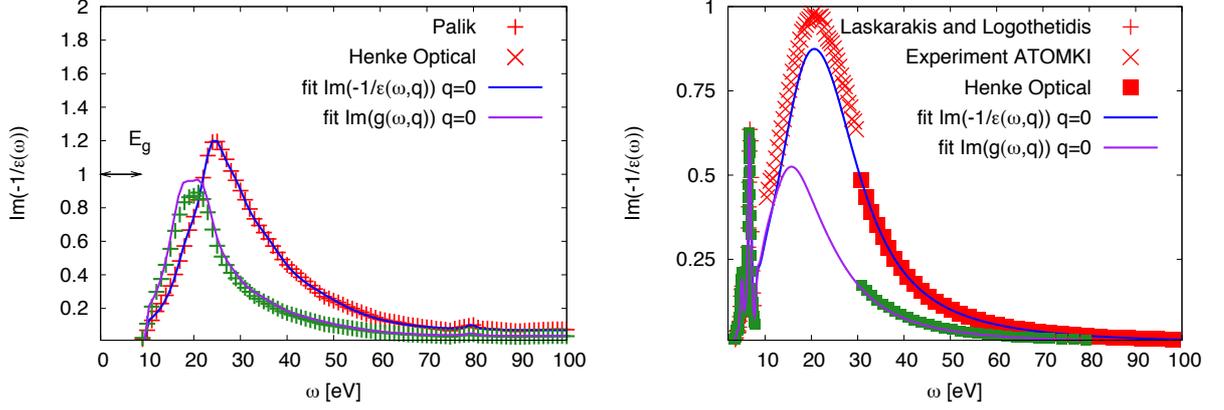
With an ansatz for the  $q$ -dependency of  $\epsilon(q, \omega) = 1 - \omega_p^2/(\omega^2 - a(q) + i\omega\gamma)$  and the abbreviation  $\omega_a^2 = \omega^2 - a(q)$ , for the bulk loss these expressions can be extended to

$$\text{Im}\left(\frac{-1}{\epsilon(q, \omega)}\right) = \frac{\epsilon_2}{\epsilon_1^2 + \epsilon_2^2} = \frac{\omega_p^2\omega\gamma}{(\omega_p^2 - \omega_a^2)^2 + \omega^2\gamma^2} \quad (\text{B.6})$$

which means that the resonance frequency is found at  $\omega_r = \sqrt{\omega_p^2 + a} = \sqrt{\omega_p^2 + sq^2 + q^4/4} \approx \omega_p + q^2/2$ . For the real part one finds

$$\text{Re}\left(\frac{1}{\epsilon(q, \omega)}\right) = \frac{\epsilon_1}{\epsilon_1^2 + \epsilon_2^2} = \frac{(\omega_a^2 - \omega_p^2)\omega_a^2 + \omega^2\gamma^2}{(\omega_p^2 - \omega_a^2)^2 + \omega^2\gamma^2}. \quad (\text{B.7})$$

Experimentally found optical data for  $\text{Im}(-1/\varepsilon(q=0, \omega))$  is now fitted by a sum of  $n$  plasmon poles with amplitude  $A_i$  and the form given in Eq. B.6. Assuming a dispersion relation  $a(q) = s^2 q^2 + q^4/4$  with  $s^2 \approx q_F/3$  being the group velocity of the plasmon, the knowledge of the  $A_i$ ,  $\omega_{p,i}$ , and  $\gamma_i$  (see Table B.1) allows for extrapolation to the  $(q, \omega)$ -plane.



**Figure B.1:** Loss function fitted to optical data given in literature. Left:  $\text{Al}_2\text{O}_3$  with  $n = 7$ . Right: PET with  $n = 12$ . For  $\text{Al}_2\text{O}_3$  the gap energy  $E_g$  is denoted by an arrow.

With the aid of the relation in Eq. B.1 knowledge of  $\text{Re}(\varepsilon^{-1})$  (Eq. B.7) and  $\text{Im}(\varepsilon^{-1})$  (Eq. B.6) enables us also to form  $\text{Im}(g(q, \omega))$ . However, for insulators  $\text{Re}(\varepsilon(q=0, \omega \rightarrow 0)) = \varepsilon_0$  approaches a constant value. The ansatz of a plasmon pole, usually valid for metals where  $\text{Re}(\varepsilon(q=0, \omega \rightarrow 0)) = \infty$ , thus fails here. An additive constant, however, can solve this inconsistency.

## B.2 Fitting the surface-IMFP

For increased numerical efficiency in the CTMC program we fit  $\lambda_{in,s}(z)/\lambda_{in,s}(0)$  by the function  $f(z, E) = 1 + (a_1 - a_2 E) \cdot |z|^{(b_1 - b_2 \log E)}$  [89] and explicitly employ only  $\lambda_{in,s}(E, 0) \times f(z, E)$  with pre-determined fit parameters  $a_1$ ,  $a_2$ ,  $b_1$ ,  $b_2$  (Table B.2). Here, also the values for  $a = a_1 - a_2 E$  and  $b = b_1 - b_2 \log E$  are given, the units are a.u. for  $z$  and eV for  $E$ .

Al <sub>2</sub> O <sub>3</sub> peak no.	$A_i$	$\gamma_i$	$\omega_{p,i}$
1	27.13	6.44	16.67
2	150.8	7.68	24.29
3	114.0	9.39	29.17
4	144.2	13.6	35.72
5	191.7	25.5	46.89
6	5.03	2.27	79.65
7	716.1	109.9	119.6
PET peak no.	$A_i$	$\gamma_i$	$\omega_{p,i}$
1	0.02	0.27	3.93
2	0.58	0.72	5.00
3	2.99	0.71	6.79
4	-1.51	1.22	7.81
5	1.18	1.23	8.24
6	-40.57	10.1	9.55
7	22.3	7.35	9.96
8	127.4	16.4	19.98
9	239.0	21.1	25.56
10	96.2	42.1	27.45
11	85.2	197.3	328.2
12	91.9	292.6	588.5

**Table B.1:** The plasmon peaks fitted to the dielectric function of Al<sub>2</sub>O<sub>3</sub> and PET.

	Al <sub>2</sub> O <sub>3</sub>	PET	Al <sub>2</sub> O <sub>3</sub> at 250 eV	PET at 500 eV
$a_1$	2.074	1.574		
$a_2$ or $a$	$3.2 \cdot 10^{-3}$	$1.2 \cdot 10^{-3}$	1.274	0.974
$b_1$	2.330	2.932		
$b_2$ or $b$	0.007	0.287	2.229	1.145

**Table B.2:** Fit parameters for Al<sub>2</sub>O<sub>3</sub> and PET employed in the function  $f(z, E)$ .



# List of Acronyms

- ATI** Above-Threshold Ionization
- CC** Channel Closing
- CEP** Carrier-Envelope Phase
- CTT** Classical Transport Theory
- EMFP** Elastic Mean Free Path
- FWHM** Full Width at Half Maximum
- HCI** Highly Charged Ion
- HHG** High-Harmonic Generation
- IMFP** Inelastic Mean Free Path
- IR** Infrared
- PET** Polyethylene-terephthalate
- SAE** Single-Active Electron
- SEE** Secondary Electron Emission
- SFA** Strong Field Approximation
- SPA** Stationary Phase Approximation
- TDSE** Time-Dependent Schrödinger Equation
- XUV** Extreme Ultraviolet



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