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Lutz Fechner

High-Resolution Experiments on Strong-Field Ionization of Atoms and Molecules

Test of Tunneling Theory, the Role of Doubly Excited States, and Channel-Selective Electron Spectra

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Test of Tunneling Theory, the Role of Doubly Excited States, and Channel-Selective Electron Spectra

Doctoral Thesis accepted by
the Ruperto Carola University of Heidelberg, Germany

Springer

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Supervisor's Foreword

The interaction of strong laser pulses with atoms and molecules has been subject of scientific interest for decades. A large manifold of interesting phenomena arise from the quantum nature of the systems, their strong modification by the laser electric field and—after ionization—from the acceleration of the freed electrons in the laser field. Beside computationally highly demanding quantum calculations, various theoretical models have been developed that are able to approximately describe the dynamics in many cases.

In order to gain insight into the details of the processes and to refine and adapt the existing models accordingly, precise measurements have to be performed and suitable experimental setups are requested. The detection of all charged particles created in a certain process and the precise determination of their three-dimensional momentum vectors became possible with the invention of the Reaction Microscope. These spectrometers provide a very high resolution and are capable of performing true multi-particle coincidence measurements on a shot-to-shot basis with very low background. Data selection according to, e.g., the masses and charge states of the created ions or the kinetic energy released in a process can yield channel-selective data and thus helps to answer the open questions in the field.

An atom exposed to a very strong laser field typically experiences a large deformation of the binding Coulomb potential. As a consequence, a potential barrier is formed where electrons may tunnel out. Thus, a very fundamental quantum process is triggered by the laser pulse. In order to relate the experimental observable—the final momentum of the electron—to the process and its dynamics, the further interaction of the freed electron with the laser field has to be taken into account: In the vicinity of its parent ion, the electron is driven back and forth by the laser. Depending on the laser electric field at the moment of ionization and the initial momentum the electron had right after tunneling, a certain trajectory and final momentum results. Theoretical models suggest that the initial momentum distribution of the freed electrons depends on the shape of their bound state wave function prior to the tunneling process. Although used in many theoretical and

experimental works, this dependence has never been shown directly in any experiment before.

In this thesis, ionization processes of rare gas atoms and hydrogen molecules are experimentally investigated. Using few-cycle laser pulses in a pump-probe scheme and argon as target, the dependence of the final electron momentum distribution on the bound state wave function is directly proven. Highly resolved electron momentum distributions are obtained for rare gas atoms and molecular hydrogen along the transition from the multiphoton to the tunneling regime, using laser pulses over a large range of wavelengths. Unique fingerprints of autoionizing Rydberg states in argon and krypton are identified. For hydrogen, different reaction channels in terms of ionization and dissociation of the molecule are isolated using the unique features of the Reaction Microscope. This provides a set of channel-resolved benchmark data for future theoretical calculations and models.

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Abstract

In this thesis, the ionization of atoms and small molecules in strong laser fields is experimentally studied by utilization of a reaction microscope.

The fundamental process of tunnel ionization in strong laser fields is subject of investigation in a pump-probe experiment with ultrashort laser pulses. A coherent superposition of electronic states in singly charged argon ions is created within the first, and subsequently tunnel-ionized with the second pulse. This gives access to state-selective information about the tunneling process and allows to test common models.

Moreover, the ionization of krypton and argon at different wavelengths is studied, from the multiphoton to the tunneling regime. The population of autoionizing doubly excited states in the laser fields is proven and a possible connection to the well-known dielectronic recombination processes is discussed. The wavelength-dependent investigations are furthermore extended to molecular hydrogen. In addition to ionization, this system might undergo different dissociative processes. Channel-selective electron momentum distributions are presented and compared to each other.

Contents

Nomenclature

ADC	Analog-to-digital converter
ADK	Ammosov-Delone-Krainov
ARS	Autoionizing Rydberg state
ATI	Above-threshold ionization
BBO	Barium borate
CE	Coulomb explosion
CEP	Carrier-envelope phase
COLTRIMS	Cold target recoil ion momentum spectroscopy
CPA	Chirped pulse amplification
CVA	Coulomb-Volkov approximation
cw	Continuous-wave
DEC	Doubly excited complex
DES	Doubly excited state
DFG	Difference frequency generation
DR	Dielectronic recombination
EBIT	Electron beam ion trap
FEL	Free-electron laser
FSR	Free spectral range
FTI	Frustrated tunneling ionization
FWHM	Full width at half maximum
GVD	Group velocity dispersion
HeNe	Laser Helium-neon laser
HHG	High harmonic generation
KER	Kinetic energy release
KLM	Kerr lens mode-locking
laser	Light amplification by stimulated emission of radiation
MCP	Micro-channel plate
MPI	Multiphoton ionization
NSDI	Non-sequential double ionization
OPA	Optical parametric amplifier
REMI	Reaction microscope

RESI	Recollision excitation with subsequent ionization
SAE	Single-active-electron
SESAM	Semiconductor saturable absorber mirror
SFA	Strong-field approximation
SFG	Sum frequency generation
SHG	Second harmonic generation
SOWP	Spin-orbit wave packet
SPM	Self-phase modulation
TEM	Mode transverse electromagnetic mode
Ti:sapphire	Titanium-doped sapphire ($Ti:Al_2O_3$)
TOD	Third-order dispersion
VMI	Velocity-map imaging

Chapter 1
Introduction

The wave-character of light and the particle-character of matter seemed disparate until the last century, when physics proved the opposite: Light and matter may likewise be described as particles and waves, a fact commonly known as *wave-particle duality* and also valid for larger, massive objects [11]. Despite this common ground, in everyday life as well as in most experiments, light and matter act completely different and can be distinguished well by their diverse properties. This was also emphasized in the 2012 Nobel Prize in Physics press release: "David Wineland traps electrically charged atoms, or ions, controlling and measuring them with light, or photons. Serge Haroche takes the opposite approach: he controls and measures trapped photons, or particles of light, by sending atoms through a trap. Both Laureates work in the field of quantum optics studying the fundamental interaction between light and matter, a field which has seen considerable progress since the mid-1980s." [16].

Indeed, nowadays, the interaction of light and matter is the subject of investigation in a large variety of experiments. Decisive for the development of the field was the invention of the laser[1] [9]. The unique properties of laser light, for example its spectral narrowness, are in fact mandatory for many experiments and exploited in a manifold of different techniques from laser cooling of atoms [15] to high resolution spectroscopy [13].

Despite the scientific potential comprised in the interaction processes, their mathematical treatment is—at first glance—fairly simple and often relies on quantum mechanical perturbation theory. In fact, the interaction of monochromatic light with an atom is one of the basic examples of such approach in undergraduate textbooks. The approach uses the fact that the electric field associated with the light is small compared to the electric field that keeps the electrons bound.

In laboratories, this condition is often, but not always fulfilled. Remarkably soon after the invention of the laser itself, techniques were invented allowing the temporal confinement of the emitted laser radiation and thus the creation of "giant" light pulses [10]. Additional spatial confinement by focussing these pulses onto a target

[1] Light amplification by stimulated emission of radiation.

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soon provided intensities with corresponding electric fields that were equal to or even exceeded the atomic fields. It is no surprise that a perturbative treatment of the interaction between the laser field and an atom breaks down in this situation, and the seemingly simple task to describe the problem becomes tremendously complicated.

Although quantum mechanics provides the appropriate equation to describe the problem, the time-dependent Schrödinger equation, finding its full numerical solution is—even with today's fastest computers—only possible for very specific, fairly simple problems. In addition, if such a solution is found, the calculation usually only returns a final result rather than an intuitive picture of the underlying processes. However, those simple pictures are desirable since they often help to predict and understand important aspects of the light-matter interactions.

In 1965, Keldysh investigated the ionization process in strong electromagnetic fields using an alternative approach of treating the laser field classically [7]. In this work, the area of strong-field physics was divided into two regimes, a separation which still persists. One side, commonly known as the *multiphoton regime*, is associated with rather low intensities and/or short wavelengths. Here, the binding energy of the electron is large compared to the *ponderomotive energy*, the cycle-averaged kinetic energy of a free electron in the laser field. The opposite is the case for high intensities and/or long wavelengths in the *tunneling regime*. As suggested by the names, the characteristics of the ionization process can be described most appropriately either by the absorption of several photons or a tunneling process of the electron, respectively.

Since then, much effort has been spent to refine and extend the classical treatment of the interaction. In the tunneling regime, the interaction between the laser and the electron after the tunneling process turned out to be extremely important for understanding the strongly enhanced rates for double and multiple ionization obtained in moderately intense laser fields [8]. Usually, the full treatment still involves quantum mechanical tunneling as a first step and a subsequent classical treatment of the freed electron which resembles a fairly good approximation. Due to this combination of quantum mechanics and classical physics, the approaches are commonly referred to as "semi-classical models". Important milestones are the ADK theory, e.g. [1, 12, 14], and the extension to three steps in the *three-step model* [3] that also considers the possible recollision of the electron with its parent ion. It is this recollision which gives rise to the enhanced rates for double and multiple ionization as well as to other interesting phenomena, such as the generation of highly energetic radiation and the formation of extremely short light pulses with durations of down to 100 as[2] and below [17].

Despite the progress, many questions remain. One of them is related to the region "in between" the two regimes, in which neither the tunneling nor the multiphoton picture seems to be valid and where, in fact, most of the experiments are carried out. When coming from the tunneling regime and approaching shorter wavelengths and thus faster and faster oscillation of the field, one may ask, up to which point tunneling

[2] One attosecond (as) equals 10^{-18} s.

can be treated adiabatically, hence neglecting the temporal variation of the field during the process. This question is still controversially discussed [2, 6]. Another question is related to the momentum distribution of the electrons right after the tunneling process, before being accelerated by the laser field. Although different models predict certain momentum distributions, their explicit experimental validation is pending.

However, promising new experimental techniques such as the *attoclock* technique [5], provide an astonishing level of detail on information about the process of tunnel ionization. For example, few tens of attoseconds were experimentally found as an upper limit for the *tunneling time*—the time the electron needs to tunnel out—in helium [4]. Similar to conventional streaking techniques, the temporal information is mapped onto spatial coordinates. For each time of ionization, associated coordinates can be calculated and the experimental data are interpreted with the reversed relation. These calculations require detailed knowledge about the initial momenta of the electrons which is—as described above—currently only available in terms of theoretical predictions from models. One of the aims of this work is to experimentally test these models.

In this thesis, the ionization of atoms and small molecules in strong laser fields is experimentally investigated by utilization of a highly advanced momentum spectrometer: the reaction microscope. Chapter 2 gives a brief overview about the mathematical description of ultrashort, strong laser pulses and their technical creation. Furthermore, the specific laser system and the pulse manipulation techniques utilized throughout this work are described. Following this, Chap. 3 focusses on the interaction of intense laser pulses with matter, in particular on the description of ionization processes in atoms and molecules. The reaction microscope and its working principle is treated in Chap. 4.

Afterwards, the experimental results are presented, divided into three parts. In the first, Chap. 5, common theoretical models about the electron momentum distribution directly after the tunneling process are explicitly tested. This is done by using ultrashort pulses in a pump-probe scheme and a coherent superposition of electronic states in singly ionized argon as initial state for further tunnel ionization.

The subject of the second experimental part, presented in Chap. 6, is the interaction of strong laser pulses of different wavelengths with noble gas atoms, namely krypton and argon. Tuning the wavelength by utilizing an optical parametric amplifier and nonlinear crystals for frequency mixing enables the observation of characteristic changes in the electron momentum distributions and the ionization process while approaching the tunneling regime. Furthermore, the excitation of doubly-excited states of the atoms is proven and investigated.

In the last part, presented in Chap. 7, a similar investigation is performed for the most simple molecule existing, H_2. In addition to pure ionization, this system features different dissociation channels. The reaction microscope enables their separation and thus offers channel-selected electron momentum distributions at different wavelengths. The results of all experiments presented in this thesis are summarized in Chap. 8, where future perspectives are also discussed.