1
Introduction

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The generation of attosecond ($1\,\text{as} = 10^{-18}\,\text{s}$) laser pulses in 2001 [1, 2] gave birth to attosecond physics, a field that continues to see rapid development [3]. The field was initially dominated by studies of electron/nuclear dynamics in atoms, molecules, and solids; however, the field has matured to include studies of nanomaterials. Ultrashort, intense light pulses with a well controlled electric-field waveform have enabled the generation of isolated attosecond light pulses [4]. Interaction of such fields with solids and nanomaterials leads to ultrafast nonlinear phenomena and dynamics and is an important research direction in attosecond nanophysics; a variety of such nonlinear and ultrafast phenomena are discussed in this book. In this chapter, we outline common photonic tools and the principle phenomena that can be used to study them. We also indicate where they are discussed in the text.

1.1
Attosecond Tools

The attosecond physics community has developed a few photonic tools to control and trace electron dynamics in matter. Two of the most important photonic tools have been applied in studies in this book and are briefly introduced. These tools are light pulses with a controlled waveform and attosecond light pulses.

1.1.1
Strong Field Control Using Laser Pulses with Well-Defined Waveforms

There are many degrees of control over laser pulses – frequency, wavelength, pulse duration, and intensity are straightforward and readily accessible

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parameters – but access to the carrier envelope phase (CEP) provides another (extremely precise) degree of control. The CEP is the offset of the maximum of the carrier wave relative to the maximum of the pulse envelope (Figure 1.1) and impacts many areas of ultrafast physics. In particular, when individual laser pulses last only a few optical cycles, the CEP becomes extremely relevant. In these cases, a variation of the waveform of the laser pulse (e.g., by changing the CEP) may significantly alter the outcome of an experiment.

Additionally, control over the CEP (φ) allows for the sculpting of optical waveforms: Fourier synthesis of waves with certain phases over a broad range of frequencies may result in non-sinusoidal electric-field waveforms, such as sawtooth or square waveforms. Such waveforms are well known in conventional electronics; however, in conventional electronics, these correspond to gigahertz frequencies, while optical light fields reach the petahertz (PHz) domain. Ultimately, such sculpted fields – similar to conventional electronics – will permit the control of electrons with the highest possible speed. Such control of electronic phenomena in nanomaterials on attosecond timescales would correspond to electronics operating at petahertz frequencies. The rapid development of this control is exemplified in several chapters in this book.

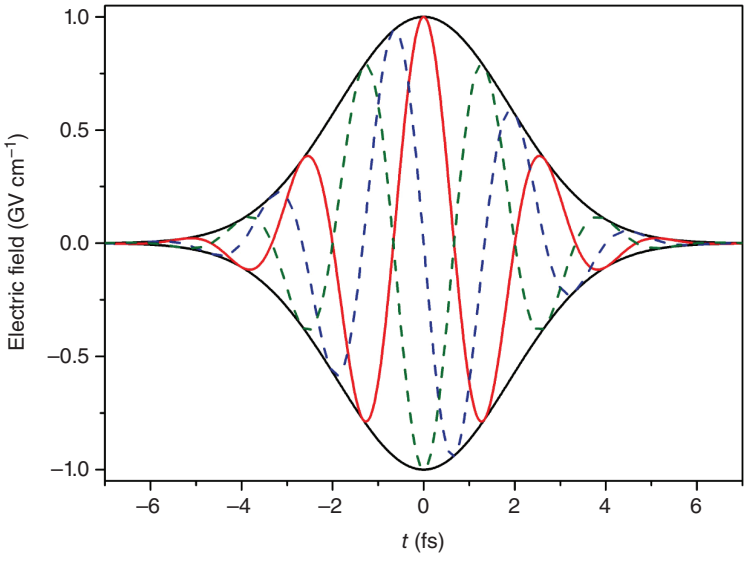


Figure 1.1 Few-cycle light fields with a controlled waveform. A few-cycle pulse (pulse duration 2.7 fs) at 800 nm with three different CEPs (red: $\varphi = 0$, green: $\varphi = \pi$, and blue: $\varphi = \pi/2$). The pulse envelope is shown as a black line.

1.1.2

Attosecond Light Pulses: Tracing Electron Dynamics

Attosecond light pulses in the extreme ultraviolet (XUV) spectral range can be generated via high-harmonic generation (HHG). HHG is commonly described as a three-step process [5, 6] (Figure 1.2) where a strong laser field first tunnel ionizes an atom or molecule (step 1), then the emitted electron is accelerated in the laser field (step 2) and finally, an XUV photon is created upon the recollision and recombination of the electron with the ion (step 3). HHG was first observed with atomic neon in 1992 [7] and has facilitated tabletop sources of coherent XUV and soft X-ray radiation [3, 8]. These novel sources find applications in time-resolved studies of electron and nuclear dynamics in atoms, molecules, nanostructures, and solids [3, 9–13].

While HHG is a coherent process that can lead to the generation of attosecond light pulses, incoherent XUV light emission might occur through fluorescence. In conventional attosecond pulse generation, a dense target and suitable phase-matching conditions can render incoherent processes negligible; however, typically a high-power laser system is required to drive the coherent process. As Chapter 2 by Pfullmann *et al*. describes, the generation of sufficiently strong fields for the generation of XUV light can also involve nanoscopic field enhancement in the near-fields of (coupled) nanostructures.

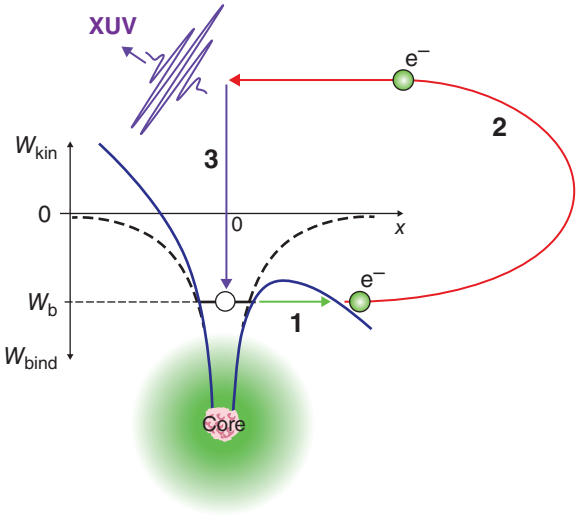


Figure 1.2 Illustration of the three-step high-harmonic generation process. Step 1, as demarcated by the black number, shows a strong laser field tunnel-ionizing an atom or molecule. Step 2 shows the electron being accelerated in the strong-laser field, and step 3 shows the higher-energy electron recombining with the core. This step causes the emission of an XUV photon.

1.2 Solids in Strong Fields

The picture of the driven electron (wavefunction) recolliding with the parent atom has been broadly used to explain the underlying physics of a plethora of gas-phase experiments with atoms and molecules, as well as for semi-infinite boundaries such as those of nanospheres and nanostructures. However, inside solids another picture has to be developed to reflect the very different environment that the driven electronic matter (wave) experiences, (i.e., compared to a vacuum in the former case). First experiments on the generation of HHG *inside* solids have indicated a different laser intensity scaling behavior, which underlines the need for a new physical picture [14]. We note in passing that even inside solids the recolliding electron picture has been successfully employed to explain high-order sideband generation and has elucidated exciton dynamics of electron–hole pairs in semiconductor quantum wells driven with terahertz fields [15].

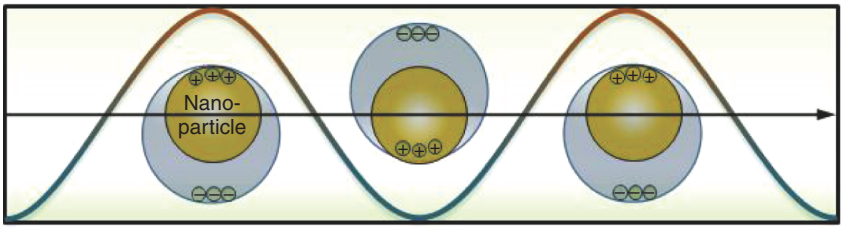
Discussions of these topics can be found in Chapter 7. Apalkov and Stockman discuss what happens to solids when they are exposed to strong laser fields. Wannier-Stark localization [16, 17] at optical field strengths can take place, which can dramatically alter the nature of the material. For example, a metal can be changed into a semiconductor, or even into a dielectric, depending on the field strength. Here, the opposite can also hold true: a dielectric can be turned into a conductor or semiconductor. In Chapter 8, initial experimental results along these lines are discussed. Schiffrin, Paasch-Colberg, and Schultze show that the resistance inside a transparent dielectric structure can be altered to an extent that its resistance indicates semiconducting behavior. It is fascinating to consider that this only holds while the optical field is large, that is, the time scale is much shorter than that of the optical period.

1.3 Attosecond Physics in Isolated Nanosystems

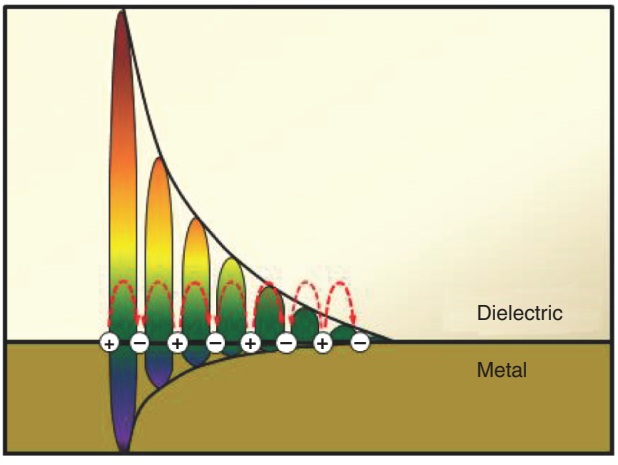
Isolated nanosystems, such as clusters, nanoparticles, and nanotips are ideal model systems for attosecond studies on the nanoscale; complex multi-electron physics can be explored under well defined conditions. In all cases, the intense field can excite collective electron dynamics. Plasmons are example of collective excitations in nanosystems at metal surfaces where conduction electrons excited by the incident light's electromagnetic field (typically ultraviolet to the visible range) oscillate collectively [18]. Plasmons are currently being explored in many directions: to increase sensitivity of optical probes [19], as single photon emission sources [20], for use in nanophotonic devices with smaller-than-conventional optical circuits, [21] and even in medical applications [22]. The plasmonic response of materials can change drastically with only small changes to the metal

nanoparticle or surrounding dielectric. This high sensitivity is largely responsible for many applications of plasmonic nanomaterials in sensing and spectroscopy.

Figure 1.3a depicts how the laser wave excites a plasmon in a nanoparticle – here a gold nanosphere. When the laser field is applied to the nanoparticle, it drives the conduction electrons collectively. This creates a strong, oscillating dipole: the plasmon. In isolated nanostructures these plasmons are *localized* surface plasmons (LSPs). Their eigenfrequencies depend on the composition, size, and shape of the nanostructure [18] as well as the surrounding dielectric. Plasmons can also propagate along metal-dielectric interfaces as *surface plasmon polaritons* (SPPs). SPPs decay exponentially into the dielectric and form an evanescent field (Figure 1.3b). The excitation of SPPs requires the matching of the light and the SPP's $\boldsymbol{k}$-vectors, which can be achieved (e.g., by gratings that are carved into the metal surface) as illustrated in Chapter 9 for metal nanotips by Lienau, Raschke, and Ropers. Surface plasmons are being explored for their



(a)



(b)

Figure 1.3 Representation of plasmonic excitation at metal-dielectric interfaces. (a) The laser's oscillatory electric field causes an oscillatory motion of the conduction electrons (localized surface plasmon) in a nanoparticle. (b) On extended surfaces, surface plasmon polaritons are formed that can propagate along the metal-dielectric interface.

potential in subwavelength optics, data storage, light generation, microscopy, and bio-photonics [19]. Propagating plasmons are of particular interest to the development of ultrafast electronics since they reach speeds close to the vacuum speed of light and can transfer information on length scales well below the diffraction limit.

Süßmann *et al.* describe the CEP-controlled electron emission from metallic nanotips and dielectric nanoparticles in intense, few-cycle laser fields in Chapter 6. The emitted electrons experience the enhanced near-fields of the nanostructures and are accelerated to energies exceeding the atomic cutoff for backscattered electrons. The measured cutoff can in turn serve as a measure of the field-enhancement. At longer, mid-infrared wavelengths the electron can leave the near-field of the nanosystem in a fraction of the laser-driven oscillations – this interesting regime is discussed in Chapter 9. Finally, Chapter 6 shows that dielectric nanostructures can be used to drive laser acceleration of electrons to an entirely new level, approaching acceleration gradients in the giga-electron volt per meter range and opening the door to optical electron accelerators – on a chip.

At intensities where multiple ionization occurs within an intense laser pulse, charge interaction becomes important. This is theoretically discussed in Chapter 4 by Saalmann and Rost for cases where the light interaction is so strong that the nanomatter is converted into a nanoplasma. The chapter shows that the complex dynamics of such transient nanoplasmas can be resolved with attosecond tools. When the diameter of a nanoparticle approaches the excitation wavelength, propagation of the light inside the particle has to be taken into account. Varin *et al*. introduce a new theoretical approach in Chapter 5, the microscopic particle-in-cell (MicPIC) simulations, that can treat the interaction of light with large nanosystems and at high intensities with the accuracy of a microscopic model. The simulations can be applied in strong near-IR or extreme-ultraviolet photoemission of clusters and large nanoparticles and offer new opportunities for modeling time-dependent diffraction studies using free-electron lasers.

1.4 Attosecond Physics on Nanostructured Surfaces

Nanostructured surfaces offer increased complexity and ultimately the ability to build nanophotonic devices with new functionalities. Of particular interest are plasmonic nanosystems. SPPs can travel short distances along a metal nanofilm, be coupled into nanotips (Chapter 9), and propagate through a nanowire [20] or along nanostructured surfaces (Chapter 10). Importantly, using the principle of adiabatic focusing [23] SPPs can be focused much below the diffraction limit, leading to extreme enhancement of fields locally. Because the dispersion of the propagating plasmon can be measured and controlled, it is even feasible to maintain an ultrashort pulse duration at the apex of a tip. This enables researchers to expose localized nanostructures on surfaces to femtosecond light pulses, certainly also with CEP control, in a nano-localized volume. The electron photoemission and acceleration and its CEP control from plasmonic nanostructured surfaces is

discussed in Chapter 3 by Dombi and Elezzabi. High-energy electrons can be generated due to the field enhancement of coupled nanostructures.

Similarly, a corrugated surface leads to localized, large field-enhancement factors (hot spots). If the local fields are large enough, electrons are emitted from the hot spot. Chapter 10 by Chew *et al*. discusses attosecond photoemission electron microscopy (ATTO-PEEM) [11] as a new metrology that enables the measurement of the plasmonic fields of hot spots and of nanostructures on surfaces with both attosecond time and nanometer spatial resolution. The chapter describes the principal methodology and progress toward its implementation. The realization of the ATTO-PEEM would comprise a wholly new surface science technique that would be broadly applicable – from understanding plasmon behavior on its natural time scale to the understanding of molecular surface bond formation, for example.

The large field-enhancement factor of nanostructured surfaces is also at the center of attempts to generate XUV light at megahertz repetition rates. As already mentioned in Chapter 2, Pfullmann *et al*. report on the status of this enabling field. The idea is to fill the surface of a transparent material with structures that locally enhance the IR laser field such that XUV generation takes place even though the laser pulse energy is much smaller than in conventional schemes. The small interaction volume at the nanostructures is mitigated by the arrangement of as many nanostructures as possible, which is an easy task with today's nanofabrication capabilities.

1.5 Perspectives

Attosecond nanophysics is a rapidly developing field. The interaction of intense fields with (nanostructured) solids offers access to nonlinear phenomena, which enables ultrafast circuitry and ultimately petahertz electronics. Progress is fueled by the application of ultrashort pulses (of just a few cycles or even less), where damage to the nanostructures can be avoided even at high intensities. The studies in this book show that the resulting collective electron dynamics are highly controllable by the waveform of the optical field interacting with the nanostructures. If we recognize that field-driven electron motion is at the very basis of microelectronics, the potential of light-field driven electron motion becomes obvious. But the impact that understanding the processes inside nanostructures on their inherent time scales could have is not limited to the topics mentioned earlier. Even sunlight harvesting through optical-antenna-enhanced solar cells might be implemented. We have already mentioned new time-resolved nanoscale imaging techniques and the in-depth understanding of plasma processes. With the help of dielectric nanostructures, new electron acceleration schemes come into reach that may one day enable the construction of small, laser-driven particle smashers. Today these machines are large and rely on particle accelerator development taken

to extreme limits over many decades, so they are likely not replaced fast. However, close to 10 000 accelerators, each about a meter in size, are operational in hospitals in oncology departments – chip-scale photonics-based dielectric counterparts may take over at some point.

While already many real-world applications of attosecond nanophysics come to mind, this book focuses on the fundamental physics behind the various directions of this nascent field. The editors hope that the book will introduce unfamiliar readers to this new and fascinating area of physics and give an overview of the various research directions. Researchers in the field may obtain an overview of ongoing activities and potentially discover new links to related fields. We thank all authors for their excellent contributions and look forward to jointly discovering where attosecond nanophysics will take us.

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