Multiphoton and Strong-Field Processes

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Abstract

The excitation of atoms by intense laser pulses can be divided into two broad regimes: the first regime involves relatively weak optical laser fields of long duration, and the second involves strong fields of short duration. In the first case, the intensity is presumed to be high enough for multiphoton transitions to occur. The resulting spectroscopy is not limited by the single-photon selection rules for radiative transitions. However, the intensity is still low enough for a theoretical description based on perturbations of field-free atomic states to be valid, and the time dependence of the field amplitude does not play an essential role. In the second case, the field intensities are too large to be treated by perturbation theory, and the time dependence of the pulse must be taken into account. In addition to a detailed picture of the two subjects described above, we include a discussion on the generation of attosecond pulses and applications of both high-order harmonic generation (HHG) and above-threshold ionization (ATI). Furthermore, we incorporate a brief summary about the incipient field of atto-nano physics.

Keywords

strong laser fields · Rydberg atoms · multiphoton ionization · multiphoton processes · attosecond physics

78.1 Introduction

The excitation of atoms by intense laser pulses can be divided into two broad regimes determined by the characteristics of the laser pulse relative to the atomic response. The first regime involves relatively weak optical laser fields of long

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duration (> 1 ns), and the second involves strong fields of short duration (< 10 ps). These will be referred to as the weak-long (WL) and strong-short (SS) cases, respectively.

In the case of atomic excitation by WL pulses, the intensity is presumed to be high enough for multiphoton transitions to occur. The resulting spectroscopy of absorption to excited states is potentially much richer than single-photon excitation because it is not limited by the single-photon selection rules for radiative transitions. However, the intensity is still low enough for a theoretical description based on perturbations of field-free atomic states to be valid, and the time dependence of the field amplitude does not play an essential role.

The SS case is fundamentally different in that the atomic electrons are strongly driven by fields too large to be treated by perturbation theory, and the time dependence of the pulse as it switches on and off must be taken into account. Atoms may absorb hundreds of photons, leading to the emission of one or more electrons, as well as photons of both lower and higher energy. Because the flux of incident photons is high, a classical description of the laser field is adequate, but the time-dependent Schrödinger equation (TDSE) must be solved directly to obtain an accurate representation of the atom–field interaction.

For SS pulses of optical wavelength, it is sufficient in most cases to consider only the electric dipole (E1) interaction term defined in Chap. 72. The atom–field interaction can then be expressed in either the length gauge or the velocity gauge [1] (Chap. 22). In the length gauge, the TDSE is

$$\mathrm{i}\hbar\frac{\partial\Psi(\boldsymbol{r},t)}{\partial t} = \left[H_0 + e\boldsymbol{r}\cdot\boldsymbol{E}(t)\right]\Psi(\boldsymbol{r},t)\;, \tag{78.1}$$

where H_0 is the field-free atomic Hamiltonian, $\boldsymbol{r}$ the collective coordinate of the electrons, and $\boldsymbol{E}(t)$ the electric field of the laser given by

$$\boldsymbol{E}(t) = \mathcal{E}(t)\left[\hat{\boldsymbol{x}}\cos(\omega t+\varphi) + \epsilon\hat{\boldsymbol{y}}\sin(\omega t+\varphi)\right]\;. \tag{78.2}$$

Here, $\mathcal{E}(t)$ defines the envelope, φ is the so-called carrier envelope phase (CEP), and ϵ characterizes the polarization: linear if $\epsilon = 0$ and circular if $|\epsilon| = 1$. In the velocity gauge, the TDSE is

$$\mathrm{i}\hbar\frac{\partial\psi(\boldsymbol{r},t)}{\partial t} = \left[H_0 - \frac{\mathrm{i}e\hbar}{mc}\boldsymbol{A}(t)\cdot\boldsymbol{\nabla} + \frac{e^2}{2mc^2}A^2(t)\right]\psi(\boldsymbol{r},t)\;. \tag{78.3}$$

Here, $\boldsymbol{A}(t) = -c\int^t \boldsymbol{E}(t')\mathrm{d}t'$ is the vector potential of the laser field. The solutions of Eqs. (78.1) and (78.3) are related by the phase transformation

$$\Psi(\boldsymbol{r},t) = \exp\left[\frac{\mathrm{i}e}{\hbar c}\boldsymbol{r}\cdot\boldsymbol{A}(t)\right]\psi(\boldsymbol{r},t)\;. \tag{78.4}$$

Since lasers must usually be focused to reach the strong-field regime, measured electron and ion yields include contributions from a distribution of field strengths. The photoemission spectrum, on the other hand, contains a coherent component due to the macroscopic polarization of all the atoms and is, therefore, sensitive also to the laser phase variations within the focal volume. In this chapter, methods for solving Eqs. (78.1) and (78.3) are discussed along with details of the atomic emission processes.

Three relevant books provide excellent introductions to this subject [1–3]. Further developments are well described in the proceedings of the International Conferences on Multiphoton Physics [4–7] and the NATO workshop on Super-Intense Laser-Atom Physics [8, 9].

78.2 Weak-Field Multiphoton Processes

78.2.1 Perturbation Theory

Since atomic ionization energies are generally $\gtrapprox 10$ eV, while optical photons have energies of only a few eV, several photons must be absorbed to produce ionization, or even electronic excitation in the case of the noble gases. For WL pulses, the electronic states are only weakly perturbed by the electromagnetic field. The rate of an n-photon transition can then be calculated using the n-th order perturbation theory for the atom–field interaction. For an incident photon number flux ϕ of frequency ω, the rate is

$$W^{(n)}_{i\to f} = 2\pi\left(\frac{2\pi\alpha\phi\omega}{e^2}\right)^n \left|T^{(n)}_{i\to f}\right|^2 \delta(\omega_i + n\omega - \omega_f)\;, \tag{78.5}$$

where

$$\begin{aligned} T^{(n)}_{i\to f} = \big\langle f\big|d\,G[\omega_i+(n-1)\omega] \\ \times d\,G[\omega_i+(n-2)\omega] \\ \cdots d\,G[\omega_i+\omega]\,d\big|i\big\rangle\;, \end{aligned} \tag{78.6}$$

$|i\rangle$ is the i-th eigenstate of the field-free atomic Hamiltonian, $d = e\hat{\boldsymbol{\epsilon}}\cdot r$, with $\hat{\boldsymbol{\epsilon}}$ the polarization direction, and

$$G(\omega) = \sum_j \frac{|j\rangle\langle j|}{(\omega-\omega_j+\mathrm{i}\Gamma_j/2)}\;. \tag{78.7}$$

The sum over j includes an integration over the continuum for all sequences of E1 transitions allowed by angular momentum and parity selection rules. Methods for calculating cross sections and rates in the weak-field regime are described in [1, 10] and in Chap. 25.

78.2.2 Resonant-Enhanced Multiphoton Ionization

For multiphoton ionization, ω can be continuously varied because the final state in Eq. (78.5) lies in the continuum. If ω is tuned so that $\omega_i + m\omega \cong \omega_j$ for some contributing intermediate state $|j\rangle$ in Eq. (78.7), then in that state lies an integer m photons above the initial state, and the corresponding denominator vanishes (to within the level width Γ_j), producing a strongly peaked resonance. Since it takes $k = n - m$ additional photons for ionization, the process is called m, k resonant-enhanced multiphoton ionization (REMPI). Measurements of the photoelectron angular distribution are useful in characterizing the resonant intermediate state.

Calculations using the semi-empirical multichannel quantum defect theory to provide the needed matrix elements have been very successful in describing experimental results. This technique is discussed in more detail in Chap. 49.

The perturbation equation Eq. (78.5) indicates that the rate for nonresonant multiphoton ionization scales as ϕ^n for an n-photon process [11]. However, this is not the case for REMPI since the resonant transition saturates, and Eq. (78.5) no longer applies. Then the rate can be controlled either by the m-photon resonant excitation step or by the number of photons k needed for the ionization step.

78.2.3 Multielectron Effects

Multiply excited states can play a role in multiphoton excitation dynamics. These states are particularly important if their energies are below or not too far above the first ionization potential. Configuration expansions including these states have been used successfully in studies, for example, of the alkaline earth atoms, which have many low-lying doubly excited states. The presence of these states can also enhance the direct double ionization of an atom [12].

78.2.4 Autoionization

The configuration interaction between a bound state and an adjacent continuum leads to an absorption profile in the single photon ionization spectrum with a Fano lineshape. The actual lineshape reflects the interference between the two pathways to the continuum. Autoionizing states can also be probed via multiphoton excitation [13, 14]. Because, in the strong-field regime, coupling strengths and phases change with intensity, the lineshapes can be strongly distorted by changing the incident intensity. At particular intensities, the phases of the excited levels can be manipulated to prevent autoionization completely. Then a trapped population with energy above the ionization limit can be created [15].

78.2.5 Coherence and Statistics

Real laser fields exhibit various kinds of fluctuations and so are never perfectly coherent. The effects of such fluctuations on the complex electric field amplitude

$$E(t) = \mathcal{E}(t) \exp\big[-\mathrm{i}\omega t + \varphi(t)\big] \tag{78.8}$$

can be modeled by a variety of stochastic processes [16], depending on the conditions [10, 17–20], as follows.

For continuum wave (CW) lasers, a phase diffusion model (PDM) is often used, for which $\mathcal{E}(t) = \mathcal{E} = \text{const.}$ and

$$\dot{\varphi}(t) = \sqrt{2b}F(t) \,, \tag{78.9}$$

where $F(t)$ describes white noise by a real Gaussian function [16] characterized by the averaged values $\langle F(t)\rangle = 0$, $\langle F(t)F(t')\rangle = 2b\delta(t - t')$. The stochastic electric field then has an exponential autocorrelation function

$$\big\langle E(t)E^*(t')\big\rangle = \mathcal{E}^2 \exp\big[-b\big|t - t'\big| - \mathrm{i}\omega(t - t')\big] \,, \tag{78.10}$$

and a Lorentzian spectrum of width b. Far-off resonance, such a Lorentzian spectrum often gives unrealistic results, and the model Eq. (78.9) is then replaced by an Ornstein–Uhlenbeck process,

$$\ddot{\varphi}(t) = -\beta\dot{\varphi}(t) + \sqrt{2b\beta}F(t) \,, \tag{78.11}$$

where the parameter β for $\beta \ll b$ plays the role of a cutoff of the Lorentzian spectrum.

A multimode laser with a large number M of independent modes has a field of the form $E(t) = \sum_{j=1}^{M} \mathcal{E}_j \exp[-\mathrm{i}\omega_j t + \mathrm{i}\varphi_j(t)]$, and according to the central limit theorem [16], can be described for large M as a complex Gaussian process defined to be a chaotic field,

$$\dot{E}(t) = -(b + \mathrm{i}\omega)E(t) + \sqrt{2b\big\langle|E(t)|^2\big\rangle}F(t) \,, \tag{78.12}$$

where $F(t)$ is now a complex white noise, and ω is the central frequency of the field. The field, Eq. (78.12) has an exponential autocorrelation function and a Lorentzian spectrum of width b.

Various other stochastic models have been discussed in the literature. These include Gaussian fluctuations of the real amplitude of the field $\mathcal{E}(t)$; Gaussian chaotic fields with non-Lorentzian spectra; non-Gaussian, nonlinear diffusion processes (that describe for instance a laser close to threshold [16]); multiplicative stochastic processes (that describe a laser with pump fluctuations [18]) and jump-like Markov processes [21–23]. Statistical properties of laser fields can sometimes be controlled experimentally to a great extent [19, 20].

78.2.6 Effects of Field Fluctuations

Since the response of systems undergoing multiphoton processes is, in general, a nonlinear function of the field intensity (and, in particular, of the field amplitude), it depends in a complex manner on the statistics of the field. The enhancement of the nonresonant multiphoton ionization rate illustrates the point. According to the perturbation equation Eq. (78.5), the rate of an n-photon process is proportional to ϕ^n; i.e., to I^n, where I is the field intensity. For fluctuating fields, the average response is thus

$$W_{i\to f}^{(n)} \propto \left\langle I^n \right\rangle \propto \left\langle \left| E(t) \right|^{2n} \right\rangle . \tag{78.13}$$

Phase fluctuations (as described by PDM) do not affect the average. On the other hand, for complex chaotic fields, the average is

$$\langle I^n \rangle \approxeq n! \langle I \rangle^n , \tag{78.14}$$

i.e., there exists a significant enhancement of the rate for $n > 1$.

Field fluctuations lead to more complex effects in resonant processes. Two well-studied examples are the enhancement of the AC Stark shift in resonant multiphoton ionization [24], and the spectrum of double optical resonance – a process in which the AC Stark splitting of the resonant line is probed by a slightly detuned fluctuating laser field [18]. Double optical resonance is very sensitive not only to the bandwidth of the probing field but also to the shape of its frequency spectrum.

78.2.7 Excitation with Multiple Laser Fields

The simultaneous application of more than one laser field produces interesting and novel effects. If a laser and its second (2ω) or third (3ω) harmonic are combined and the relative phase between the fields controlled, product state distributions and yields can be altered dramatically. The effects include reducing the excitation or ionization rates in the $\omega - 3\omega$ case [25] or altering the photoelectron angular distributions and the harmonic emission parity selection rules using a $\omega - 2\omega$ laser source [26].

A laser field can dress or strongly mix the field-free excited states, including the continuum, of an atom. This can produce a number of effects depending on how the dressed system is probed. By coupling a bound, excited state with the continuum, ionization strengths and dynamics are altered, resulting in new resonance-like structures where none existed before. This effect is called laser-induced continuum structure, or LICS [15, 27]. This general idea has been exploited to design schemes for lasers without inversion [28] in which the dressed atom can have an inverted population, allowing gain, even though in terms of the undressed states, the lower level has the largest population. A laser can produce dramatic changes in the index of refraction of an atomic medium [29], creating, at specific frequencies, laser-induced transparency for a second, probe laser field. Multistep ionization, where each step is driven by a laser at its resonant frequency, has resulted in two useful applications. These are: efficient atomic isotope separation [23]; and the detection of small numbers of atoms in a sample, called single-atom detection. This technique is extremely sensitive because the use of exact resonance for each step yields very large cross sections for ionization, and the efficiency of collecting ions is high [30].

78.2.8 Waveforms

One step beyond would be to drive the atomic or molecular system with a so-called multicolor laser field, i.e., with a laser source created starting from different laser frequencies, not necessarily a multiple of each other (these sources are also known as waveforms or light-field transients). Waveform-controlled light transients with a bandwidth spanning almost two octaves have been demonstrated at microjoule energy and gigawatt peak power levels. These particular sources allow temporal confinement of optical radiation to less than 1 fs in subcycle waveforms. With their power substantially enhanced, these extreme waveforms may open up a new stage in nonlinear optics and attoscience. This is mainly due to, among other things, the feasibility of suppressing ionization up to unprecedented peak intensities and instantaneous ionization rates approaching optical frequencies, respectively. A prototypical three-color few-cycle system is already available, and it offers a conceptually simple route to scaling multioctave optical waveform synthesis to the multiterawatt regime (e.g., [31]). To this end, with the three channels delivering few-cycle pulses in the visible (VIS), $0.45 \approx 0.65\,\mu$m, near-infrared (NIR), $0.7 \approx 1.3\,\mu$m, and mid-infrared (MIR), $1.6 \approx 2.7\,\mu$m, spectral ranges are recombined using a set of dichroic chirped mirrors to yield one single beam. Additionally, the feasibility of superoctave optical waveform synthesis was recently demonstrated in the NIR-VIS-ultraviolet (UV) spectral range by seeding a three-channel and, more recently, a four-channel, synthesizer consisting of broadband chirped mirrors with a continuum originating from a Ti:sapphire-laser-driven hollow-fiber/chirped-mirror compressor. Further degrees of freedom for waveform sculpting can be introduced by shaping the amplitude and phase of the spectra of the individual channels, e.g., via an acousto-optic pulse shaper and/or a spatial light modulator (for a recent review see, e.g., [32]).

78.3 Strong-Field Multiphoton Processes

Recently developed laser systems can produce very short pulses, some as short as a few to tens of femtoseconds, while at the same time maintaining the pulse energy so that the peak power becomes very high. In this regard, e.g., focused intensities up to $10^{19}\,\mathrm{W/cm^2}$ have been achieved. Because the pulses are short, atoms survive to much higher intensities before ionizing, making possible studies of laser–atom interactions in an entirely new regime [33]. A discussion of the status of short-pulse laser development is given in Chap. 75 and in [34].

With increasing intensity, higher-order corrections to Eq. (78.6) contribute to the transition rate. The next order correction comes from transitions involving two additional photons, one absorbed and one emitted, leading to the same final state. One effect of these terms is to shift the energies of the excited states in response to the oscillating field. This is called the dynamic or AC Stark shift. The AC Stark shift of the ground state tends to be small because of the large detuning from the excited states for long wavelength photons. On the other hand, in strong fields, the shift of the higher states and the continuum can become appreciable. Electrons in highly excited states respond to the oscillating field in the same manner as a free electron. Their energies shift with the continuum by the amount

$$U_\mathrm{p} = \frac{(1+\epsilon)e^2\mathcal{E}^2}{4m\omega^2}\,, \tag{78.15}$$

where U_p is the cycle-averaged kinetic energy of a free electron in the field, and ϵ defines the polarization of the field Eq. (78.2); U_p is called the ponderomotive or quiver energy of the electron. For strong laser fields, U_p can be several eV or more, meaning that during a pulse, many states shift through resonance as their energies change by an amount larger than the incident photon energy. The resulting intensity-induced resonances can dominate the ionization dynamics.

Electrons promoted into the continuum acquire the ponderomotive energy, oscillating in phase with the field. In a linearly polarized field, the amplitude of the quiver motion of a free electron, given by $e\mathcal{E}/m\omega^2$, can become many times larger than the bound-state orbitals. If the initial velocity of an electron is small after ionization, it can be accelerated by the laser electric field and back into the vicinity of the ion core, when the field reverses its direction. The subsequent rescattering process, amongst other effects, changes the photoelectron energy and angular distributions, and allows the emission of high-energy photons [35, 36]. This simple dynamical picture forms the basis of the current understanding of many strong-field multiphoton processes.

78.3.1 Nonperturbative Multiphoton Ionization

The breakdown of perturbation theory for nth-order multiphoton processes occurs when the higher-order correction terms become comparable to the n-th-order term. Assuming that the dipole strength is $\propto ea_0$, where a_0 is the Bohr radius, and the detuning is $\delta \propto \omega$, the ratio of an $(n+2)$-order contribution to the n-th-order term from Eq. (78.6) is [1]

$$R_{n+2,n} \approxeq \left(\frac{2\pi\alpha\phi\omega}{e^2}\right)\left(\frac{ea_0}{\omega}\right)^2 = \left(\frac{I}{I_\gamma}\right)\left(\frac{\omega_\mathrm{a}}{2\omega}\right)^2\,, \tag{78.16}$$

where $\hbar\omega_\mathrm{a} \approxeq 27.2114\,\mathrm{eV}$ is the atomic unit of energy e^2/a_0, and $I_\gamma \approxeq 3.50945\times10^{16}\,\mathrm{W/cm^2}$ is the intensity corresponding to an atomic unit of field strength, given by $E_\mathrm{a} = \alpha c\left(m/a_0^3\right)^{1/2} \approxeq 5.1422\times10^9\,\mathrm{V/cm}$. The atomic unit of intensity itself is defined by

$$I_\mathrm{a} = \phi_\mathrm{a}\hbar\omega_\mathrm{a} = \alpha c E_\mathrm{a}^2\,, \tag{78.17}$$

which is $6.436414(4)\times10^15\,\mathrm{W/cm^2}$. Thus, $I_\gamma = I_\mathrm{a}/(8\pi\alpha)$. For photon energies of 1 eV, $R_{n+2,n}$ becomes unity for $I \approx 10^{14}\,\mathrm{W/cm^2}$. Because of the large number of $(n+2)$-order terms, perturbation theory actually fails for $I > 10^{13}\,\mathrm{W/cm^2}$. Above this critical intensity, nonresonant n-photon ionization ceases to scale with the ϕ^n dependence predicted by perturbation theory.

78.3.2 Tunneling Ionization

At sufficiently high intensity and low frequency, a tunneling mechanism changes the character of the ionization process. For lasers in the infrared (IR) or optical range, a strongly bound electron can respond to the instantaneous laser field since the oscillating electric field varies slowly on the timescale of the electron. The Coulomb attraction of the ion core combines with the laser electric field to form an oscillating barrier through which the electron can escape by tunneling, if the amplitude of the laser field is large enough. The DC rate for this process is $e\mathcal{E}/\sqrt{2mI_\mathrm{P}}$, where I_P is the ionization potential of the electron. When this rate is comparable to the laser frequency, tunneling becomes the most probable ionization mechanism [37–39]. The ratio of the incident laser frequency to the tunneling rate is called the Keldysh parameter and is given by

$$\gamma = \sqrt{I_\mathrm{P}/2U_\mathrm{p}}\,, \tag{78.18}$$

which is less than unity ($\gamma \ll 1$) when tunneling dominates and larger than unity ($\gamma \gg 1$) when multiphoton ionization

governs the laser–matter process. Interestingly, many experiments are carried out in an intermediate or *crossover* region, defined by $\gamma \approx 1$. Another way to interpret γ is to note that we can write $\gamma = \tau_T/\tau_L$, where τ_T is the Keldysh time (defined as $\tau_T = \frac{\sqrt{2I_P}}{\mathcal{E}}$) and $\tau_L = 2\pi/\omega$ is the laser period. Hence, γ serves as a measure of nonadiabaticity by comparing the response time of the electron wavefunction to the period of the laser field.

78.3.3 Multiple Ionization

Excitation and ionization dynamics are dominated by single-electron transitions in the strong-field regime. Although atoms can lose several electrons during a single pulse, the electrons are released sequentially. There is no convincing evidence of significant collective excitation in atoms in strong fields, even though it has been extensively sought. Once one electron is excited in an atom, the remaining electrons have much higher binding energies. As a result, the laser field is unable to affect them significantly until it reaches a much higher intensity. By that time, the first electron has already been emitted.

Simultaneous ejection of two electrons occurs as a minor channel ($< 1\%$) in strong-field multiple ionization. Although it is possible that doubly excited states of atoms could assist in the double ionization, in the helium and neon cases studied, these states are unlikely to be contributors [40].

Correlated multielectron ionization became a very hot subject over the past 20 years. Various mechanisms were proposed and demonstrated theoretically and experimentally. Perhaps the most important is the recollision mechanism, in which one electron tunnels out and is driven back to the nucleus by the laser field, where it recollides and releases a second atomic or molecular electron [41].

78.3.4 Above Threshold Ionization

In strong optical and IR laser fields, electrons can gain more than the minimum amount of energy required for ionization. Rather than forming a single peak, the emitted electron energy spectrum contains a series of peaks separated by the photon energy. This is called above-threshold-ionization, or ATI [42–45]. The peaks appear at the energies

$$E_s = (n + s)\hbar\omega - I_P \,, \tag{78.19}$$

where n is the minimum number of photons needed to exceed I_P, and $s = 0, 1, \ldots$ is called the number of excess photons or above threshold photons carried by the electron. Calculations in the perturbative regime for ATI are given for hydrogen in [11].

Peak Shifting

As the intensity approaches the nonperturbative regime, the AC Stark shift of the atomic states begins to play a significant role in the structure of the ATI spectrum. The first effect is a shift of the ionization potential, given roughly by the ponderomotive energy U_p. Additional photons may then be required in order to free the electron from the atom, i.e., enough to exceed $I_P + U_p$. If the emitted electron escapes from the focal volume while the laser is still on, it is accelerated by the gradient of the field. The quiver motion is converted into radial motion, increasing the kinetic energy by U_p and exactly canceling the shift of the continuum. In this situation, the electron energies are still given by Eq. (78.19). However, when U_p exceeds the photon energy, the lowest ATI peaks disappear from the spectrum. In this long pulse limit, no electron is observed with energy less than U_p. This is called peak shifting and names the situation where the dominant peak in the ATI spectrum moves to higher order as the intensity increases.

ATI Resonance Substructure

If the laser pulse is short enough (< 1 ps for the typical laser focus), the field turns off before the electron can escape from the focal volume. Then the quiver energy is returned to the field, and the ATI spectrum becomes much more complicated. The observed electron energy can now be computed by

$$E_s \text{ (short pulse)} = (n + s)\hbar\omega - (I_P + U_p) \,, \tag{78.20}$$

which results shifted an amount U_p with respect to the long pulses case Eq. (78.19). Electrons from different regions of the focal volume are thus emitted with different ponderomotive shifts, introducing a substructure in the spectrum, which can be directly associated with AC Stark-shifted resonances [42, 43].

ATI in Circular Polarization

The above discussion is particularly appropriate for the case of linear polarization, where the excited states of the atom can play a significant role in the excitation. On the contrary, in a circularly polarized field, the orbital angular momentum L must increase one unit with each photon absorbed, so that multiphoton ionization is allowed only to states that have high L, and, hence, a large centrifugal barrier. The lower energy scattering states thus cannot penetrate into the vicinity of the initial state. Thus, the ATI spectrum in circular polarization peaks at high energy and is very small near threshold.

ATI Applications

In the few-cycle regime, a precise knowledge of the CEP is instrumental to adequately characterize the subsequent nonlinear laser–matter phenomena. Then, the CEP plays

a major role in, for instance, the production and control of single attosecond pulses and the investigation of nonsequential double-electron ionization [46–48], amongst others. Recently, experimental observations of ATI driven by few-cycle IR-laser pulses have demonstrated that using a so-called "stereo" technique measurement of electrons emitted on the left and on the right with respect to the linear laser polarization, it is possible to extract the CEP of the driving laser [49].

In addition, another important application is to use energetic rescattered ATI electrons to estimate the bond distance in molecules. For instance, recent measurements in acetylene (C_2H_2) [50] have proved that is entirely possible to extract structural information of the chemical internuclear distance between the carbon atoms in the C–C pair and the carbon and hydrogen atoms in the C–H one, both with unprecedented high accuracy. This singular technique is named laser-induced electron diffraction (LIED) and promises to lead the time-resolved studies in complex molecules [51].

The two above-mentioned examples, namely the few-cycle CEP characterization and the LIED technique, show the capabilities of ATI to interrogate nature and answer fundamental questions about both the electron dynamics and molecular structure, just to name a few. Hence, ATI provides a powerful tool to not only give light about time-dependent atomic and molecular processes but also to configure an invaluable ally for extracting information of the driving laser itself [49].

78.3.5 High-Order Harmonic Generation

High-order harmonic generation (HHG) in noble gases is a rapidly developing field of laser physics [33, 52–54]. When an SS pulse interacts with an atomic gas, the atoms respond in a nonlinear way, emitting coherent radiation at frequencies that are integral multiples of the laser frequency. Due to the inversion symmetry of the atom, only odd harmonics of the fundamental are emitted. In the high-intensity ($> 10^{13}\,\mathrm{W/cm^2}$), low-frequency regime, the harmonic strengths fall off for the first few orders, followed by a broad plateau of nearly constant conversion efficiency and then a rather sharp cutoff [52, 53]. The plateau extends to well beyond the hundredth order of the 800–1000 nm incident wavelengths, using the light noble gases as the active medium. Moreover, there has also been experimental evidence of HHG from ions. Considering the laser-induced electron dynamics behind HHG natively develops on a sub-fs timescale, HHG provides a source of very bright, short-pulse, coherent XUV radiation, which can have several advantages over the other known sources, such as the synchrotron.

Plateau and Cutoff

The well-known three-step model [35, 36], which in 2018 turned 25 years old and combines quantum and classical aspects of laser-atom physics, accounts for many strong-field phenomena. In this model, the electron first tunnels out [55] from the ground state of the atom through the barrier formed by the Coulomb potential and the laser field. Its subsequent motion can be treated classically, and primarily consists of oscillatory motion in phase with the laser field. If the electron returns to the vicinity of the nucleus with kinetic energy $\mathcal{T}$, it may recombine into the ground state with the emission of a photon of energy $(2n+1)\hbar\omega \leq \mathcal{T} + I_\mathrm{P}$, where n is an integer. (The relation of this recombination process to atomic photoionization is discussed briefly in Chap. 25.) The maximum kinetic energy of the returning electron turns out to be $\mathcal{T} \approxeq 3.2\,U_\mathrm{p}$, resulting in a cutoff in the harmonic spectrum at the harmonic of order

$$N_\mathrm{max} \approxeq (I_\mathrm{P} + 3.2U_\mathrm{p})/\hbar\omega\ . \tag{78.21}$$

Theoretical Methods

Calculation of harmonic strengths requires the evaluation of the time-dependent dipole moment of the atom,

$$\boldsymbol{d}(t) = \langle\Psi(t)|e\boldsymbol{r}|\Psi(t)\rangle\ . \tag{78.22}$$

The strength of harmonics emitted by a single atom are then related to the Fourier components of $\boldsymbol{d}(t)$, or more precisely, its second time derivative, $\ddot{\boldsymbol{d}}(t)$.

The induced dipole moment $\boldsymbol{d}(t)$ can be directly evaluated from the numerical [56] or Floquet [26] solutions of the TDSE. Good agreement with numerical and experimental data is also obtained using the strong-field approximation discussed below and a Landau–Dyhne formula. This latter approach can be considered to be a quantum mechanical implementation of the three-step model [57].

Propagation and Phase-Matching Effects

A single atom response is not sufficient to determine the macroscopic response of the atomic medium. Because different atoms interact with different parts of the focused laser beam, they feel different peak field intensities and phases (which actually undergo a rapid π shift close to the focus, due to geometrical effects). The total harmonic signal results from coherently adding contributions from single atoms, accounting for propagation and interference effects. The latter effects can wipe out the signal completely if a constructive phase matching does not take place.

The propagation and phase-matching effects in the strong-field regime [58] can be studied by solving Maxwell's equations for a given harmonic component of the electric

field $\mathcal{E}_M(\boldsymbol{r})$ Eq. (72.23),

$$\nabla^2 \mathcal{E}_{\mathrm{M}}(\boldsymbol{r}) + n_{\mathrm{M}}(\boldsymbol{r})\mathcal{E}_{\mathrm{M}}(\boldsymbol{r}) = -\frac{1}{\epsilon_0}\left(\frac{\mathrm{M}\omega}{c}\right)^2 \mathcal{P}_{\mathrm{M}}(\boldsymbol{r}) \,, \tag{78.23}$$

where $n_{\mathrm{M}}(\boldsymbol{r})$ is the refractive index of the medium (which depends on atomic, electronic, and ionic dipole polarizabilities), while $\mathcal{P}_{\mathrm{M}}(\boldsymbol{r})$ is the polarization induced by the fundamental field only. The latter can be expressed as

$$\mathcal{P}_{\mathrm{M}}(\boldsymbol{r}) \propto N(\boldsymbol{r}) d_{\mathrm{M}}\mathrm{M}(\boldsymbol{r}) \exp\left[-i\mathrm{M}\Delta(\boldsymbol{r})\right] , \tag{78.24}$$

where $N(\boldsymbol{r})$ is the atomic density, $d_{\mathrm{M}}(\boldsymbol{r})$ is the M-th Fourier component of the induced dipole moment, and $\Delta(\boldsymbol{r})$ is a phase shift coming from the phase dependence of the fundamental beam due to focusing. All of these quantities may have a slow time dependence, reflecting the temporal envelope of the laser pulse.

Typically, phase matching is the most efficient in the forward direction. In general, the strength and spatial properties of a harmonic depend in a very complex way on the focal parameters, the medium length, and the coherence length of a given harmonic. Propagation and phase-matching effects can lead to a shift of the location of the cutoff in the harmonic spectrum [59].

Harmonic Generation by Elliptically Polarized Fields

The three-step model implies that for harmonic emission it is necessary that the tunneling electrons return to the nucleus and recombine into their initial state. According to classical mechanics, there are many trajectories in a linearly polarized field that involve one or more returns to the origin. However, there are practically no such trajectories in elliptically polarized fields. As a result, the three-step model predicts a strong decrease of the harmonic strengths as a function of the laser ellipticity. This prediction has been already confirmed experimentally [60].

The Generation of Attosecond XUV Pulses

Manipulation of generated harmonics by allowing the temporal beating of superposed high-order harmonics can produce a train of intense and very short spikes, on the order of ≈ 100 as and shorter, where $1\,\mathrm{as} = 10^{-18}$ s [61–63]. The structural characteristics of the generated pulse trains depend on the relative phases of the harmonics combined. Employing driving pulses that were themselves only a few femtoseconds long, experimental groups in Vienna [64] and Paris [65] reported the first observations and measurements of such subfemtosecond UV/XUV light pulse trains. Very recently, a new world record was set in Florida, where a single 53-attosecond X-ray pulse, with a photon energy enough to reach the carbon K-edge, was measured [66]. The scientific importance of breaking the femtosecond barrier is obvious: the timescale necessary for probing the motion of an electron in a typical bound, the valence state, is measured in attoseconds (atomic unit of time $\equiv 24$ as). Attosecond pulses will allow the study of the time-dependent dynamics of correlated electron systems by freezing the electronic motion, in essence exploring the structure with ultrafast snapshots. A crucial aspect for all attosecond pulse generation is the control of spectral phases. Measurements of the timing of the attosecond peaks relative to the absolute phase of the IR driving field have been accomplished [67]. This provides insight into the recollision, the key step in the harmonic generation process. Also, the control of the group velocity phase relative to the envelope of the few-cycle driving pulses allows the production of reproducible pulse trains [68]. Thus, the highly nonperturbative, nonlinear multiphoton interactions of very short IR or visible light pulses with atoms or molecules is becoming a novel, powerful, and unique source for studies of the fastest quantum electronic processes known to date.

HHG Applications

One of the singular aspects of HHG is the possibility to produce and control both trains of and single attosecond pulses. These ultrashort light sources are extremely important for the investigation of the ultrafast process in atomic and molecular systems. The Auger decay, the delay in the photoemission absorption via an XUV source and double-electron ionization are examples of phenomena that occur on the subfemtosecond time domain. HHG allows not only to get knowledge about the ultrafast electron dynamics inside atoms and molecules, but it also has also been demonstrated that it configures an indispensable tool to extract atomic and molecular structural information [69, 70].

The fascinating technological advances of pump-probe experiments, where attosecond pulses and delayed IR lasers are being employed to firstly trigger the system and later interrogate its dynamical time evolution, have allowed us to address unexplored questions on the above-mentioned processes. The extension of these techniques to more complex systems, such as solid structures and biological materials, is already in the pipeline.

HHG in Bulk Matter

So far, the cornerstone of attosecond science applications has been the HHG phenomenon in gases. However, there still exists a clear disadvantage of HHG due to its poor conversion efficiency—the ratio between the outgoing XUV and incoming IR photon fluxes. Recent advances in light sources within the mid-infrared (MIR) domain, $1.7 \approx 7\,\mu\mathrm{m}$, have opened a new avenue in the investigation of HHG using condensed matter materials [71]. In particular, the control of such MIR sources allows us to overcome the low-efficiency problem and substantially increase the photon flux per produced at-

tosecond pulse. This characteristic could lead to unexplored physical processes in that condensed matter phase.

As is known, in an insulator, there exists the concept of band energy dispersion for electrons in its valence or conduction bands. For instance, the HHG in semiconductors (dielectrics) is governed by electron or holes bound wavefunctions that are not spatially (or energetically) localized. Holes, in addition, also have the possibility to move on their own valance band while, on the other hand, the electron dynamics takes place entirely in the conduction band. This leads to two main contributions to the HHG process, namely, one dictated by an intraband oscillation and other by an interband electron–hole current [72, 73].

This emerging field is attracting the attention of both the attosecond science and condensed matter communities. Join efforts are being carried out to address fundamental questions such as how particle collisions can be mimicked in a solid through HHG processes [74] or how Bloch oscillations might be observed in real time [75]. Additionally, questions related to topological insulators and how their features might affect the harmonic emissions were recently brought up [76, 77].

78.3.6 Stabilization of Atoms in Intense Laser Fields

It has been argued [78] that in very intense laser fields of high frequency, atoms undergo dynamical stabilization and do not ionize. The stabilization effect can be explained by gauge transforming the TDSE Eq. (78.1) to the Kramers–Henneberger (K–H) frame; i.e., a noninertial oscillating frame that follows the motion of the free electron in the laser field. The K–H transformation consists of replacing $\boldsymbol{r} \to \boldsymbol{r} + \boldsymbol{\alpha}(t)$, where, for the linearly polarized monochromatic laser field, $\boldsymbol{\alpha}(t) = \hat{\boldsymbol{x}}\alpha_0 \cos(\omega t - \varphi)$; $\alpha_0 = e\mathcal{E}/\left(m\omega^2\right)$ is the excursion amplitude of a free electron, and $\hat{\boldsymbol{x}}$ is the polarization direction. The TDSE in the K–H frame is

$$\mathrm{i}\hbar\frac{\partial\Psi(\boldsymbol{r},t)}{\partial t} = \left\{-\frac{\hbar^2\nabla^2}{2m} + V\left[\boldsymbol{r} + \boldsymbol{\alpha}(t)\right]\right\}\Psi(\boldsymbol{r},t)\ , \quad (78.25)$$

i.e., it describes the motion of the electron in an oscillatory potential. In the high-frequency limit, this potential may be replaced by its time average

$$V_{\mathrm{K\text{–}H}}(\boldsymbol{r}) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} \mathrm{d}t\ V\left[\boldsymbol{r} + \boldsymbol{\alpha}(t)\right]\ , \quad (78.26)$$

and the remaining Fourier components of $V[\boldsymbol{r} + \boldsymbol{\alpha}(t)]$ treated as a perturbation. When α_0 is large, the effective potential Eq. (78.26) has two minima close to $\boldsymbol{r} = \pm\hat{\boldsymbol{x}}\alpha_0$. The corresponding wave functions of the bound states are centered near these minima, thus exhibiting a dichotomy. The ionization rates from the K–H bound states are induced by the higher Fourier components of $V(\boldsymbol{r} + \boldsymbol{\alpha}(t))$. For large enough α_0, the rates decrease, if either the laser intensity increases, or the frequency decreases.

Numerical solutions of the TDSE [79, 80] show that stabilization, indeed, occurs for laser field strengths and frequencies of the order of one atomic unit. More importantly, stabilization is possible even when the laser excitation is not monochromatic but, rather, is produced by a short laser pulse. Physically, free electrons in a monochromatic laser field cannot absorb photons due to the constraints imposed by energy and momentum conservation. Absorption is possible only in the vicinity of a potential, such as the Coulombic attraction of the nucleus. In the case of a strong excitation, i.e., when α_0 is much larger than the Bohr radius, the electron spends most of the time very far from the nucleus, and, therefore, does not absorb energy from the laser beam. Therefore, stabilization, as viewed from the K–H frame, has a classical analog. Other mechanisms of stabilization based on the quantum mechanical effects of destructive interference between various ionization paths have also been proposed [81].

Due to classical scaling (Sect. 78.3.8) stabilization is predicted to occur for much lower laser frequencies, if the atoms are initially prepared in highly excited states. If additionally, the initial state has a large orbital angular momentum corresponding to classical trajectories that do not approach the nucleus, stabilization is even more easily accomplished. For instance, the stabilization of a $5g$ Rydberg state of neon has been reported [82].

Although recently interest in stabilization in intense laser fields became very limited, the K–H transformation and the stabilization effect have been discussed in a completely different situation. In [83, 84], the stabilization of atoms in a shaken trap Bose–Einstein condensate was proposed. In fact, recently, a K–H trick was used to develop an experimental simulator of ultrafast processes in strong laser fields using ultracold trapped atoms [85].

78.3.7 Molecules in Intense Laser Fields

Molecular systems are much more complex than atoms because of the additional degrees of freedom resulting from nuclear motion. Even in the presence of a laser field, the electron and nuclear degrees of freedom can be separated by the Born–Oppenheimer approximation, and the dynamics of the system can be described in terms of motions on potential energy surfaces. In strong fields, the Born–Oppenheimer states become dressed, or mixed by the field, creating new molecular potentials. Because of avoided crossings between the dressed molecular states, the field induces new potential wells in which the molecules become trapped.

These states, known as laser-induced bound states, are stable against dissociation, but exist only while the laser field is present [86]. Their existence affects the spectra of photoelectrons, photons, and the fragmentation dynamics. If the field is strong enough, many electrons can be ejected from a molecule before dissociation, producing highly charged, energetic fragments [87]. Such experiments are similar to beam–foil Coulomb explosion studies of molecular structure. However, because of changes from the field-free equilibrium geometries in laser dissociation, the energies of the fragments lie systematically below the corresponding values from Coulomb explosion studies. Additionally to the latter, a similar phenomenon was intensively studied with atomic clusters [88].

78.3.8 Microwave Ionization of Rydberg Atoms

Similar phenomena appear in the ionization of highly excited hydrogen-like (Rydberg) atoms by microwave fields [89, 90], but the dynamical range of the parameters involved is different from the case of tightly bound electrons. Recent developments have greatly extended techniques for the preparation and detection of Rydberg states. Since, according to the equivalence principle, highly excited Rydberg states exhibit many classical properties, a classical perspective of ionization yields useful insights (Sect. 78.4.5).

Classical Scaling

The classical equations of motion for an electron in both a Coulomb field and a monochromatic laser field polarized along the z-axis are invariant with respect to the following scaling transformations:

$$\begin{aligned} \boldsymbol{p} &\propto n_0^{-1}\tilde{\boldsymbol{p}}\,, \quad & \boldsymbol{r} &\propto n_0^{2}\tilde{\boldsymbol{r}}\,, \\ t &\propto n_0^{3}\tilde{t}\,, \quad & \varphi &\propto \tilde{\varphi}\,, \\ \omega &\propto n_0^{-3}\tilde{\omega}\,, \quad & \mathcal{E} &\propto n_0^{-4}\tilde{\mathcal{E}}\,. \end{aligned} \tag{78.27}$$

In the scaled units, the Hamiltonian $\tilde{H} = n_0^{-2}H$ of the system becomes

$$\tilde{H} = \frac{\tilde{p}^2}{2m} - \frac{1}{\tilde{r}} + \tilde{z}e\tilde{\mathcal{E}}\cos\left(\tilde{\omega}\tilde{t} + \tilde{\varphi}\right)\,, \tag{78.28}$$

i.e., it depends only on $\tilde{\omega}$ and $\tilde{\mathcal{E}}$. In experiments, the principal quantum number n_0 of the prepared initial state typically ranges from 1 to 100.

Classical scaling extends to the fields of other polarizations and to pulsed excitations, provided that the number of cycles in the rise, top, and fall of the pulse is kept fixed. This scaling does not hold for a quantum Hamiltonian, unless one also rescales Planck's constant, $\tilde{\hbar} = \hbar/n_0$. In practice, increasing n_0, keeping $\tilde{\mathcal{E}}$ and $\tilde{\omega}$ constant, corresponds to a decrease in the effective $\hbar$ toward the classical limit. In view of this classical scaling, experimental and theoretical results are usually analyzed in terms of the scaled variables. Since the classical dynamics generated by the Hamiltonian Eq. (78.28) exhibits chaotic behavior in some regimes, the dynamics of the corresponding quantum system is frequently referred to as an example of quantum chaos [91–93].

Regimes of Response

By varying the initial n_0, several regimes of the scaled parameters can be covered. The experimentally measured response of Rydberg atoms in microwave fields can be divided into six categories:

The tunneling regime

For $\tilde{\omega} \leq 0.07$, the response of the system is accurately represented as tunneling through the slowly oscillating potential barrier composed of the Coulomb and microwave potentials.

The low-frequency regime

For $0.05 \leq \tilde{\omega} \leq 0.3$, the ionization probability exhibits distinct structures (bumps, steps, changes of slope) as a function of the field strength. The quantum probability curves might be lower or higher than the corresponding classical counterparts, calculated with the aid of the phase averaging method (Sect. 78.4.5).

The semiclassical regime

For $0.1 \leq \tilde{\omega} \leq 1.2$, the ionization probabilities agree well for most frequencies with the results obtained from the classical theory. In particular, the onset of ionization and appearance intensities (i.e., the intensities at which a given degree of ionization is achieved) coincide with the onset of chaos in the classical dynamics. In the ionization probabilities, resonances appear that correspond to the classical trapping resonances [91–94].

The transition region

For $1 \leq \tilde{\omega} \leq 2$, the differences between the quantum and classical results are visible. Quantum ionization probabilities are frequently lower and appearance intensities higher than their classical counterparts.

The high-frequency regime

For $\tilde{\omega} \geq 2$, quantum results for ionization probabilities are systematically lower and appearance intensities higher than their classical counterparts. This apparent stability of the quantum system has been attributed to three kinds of effects: quantum localization [94], quantum scars [95], and perhaps the stabilization of atoms in intense laser fields (Sect. 78.3.6).

The photoeffect regime
When the scaled frequency becomes greater than the single-photon ionization threshold, the system undergoes single-photon ionization (the photoeffect).

Quantum Localization
The classical dynamics changes as the field increases. Chaotic trajectories start to fill the phase space and, as the KAM tori (describing periodic orbits) [91–93] break down, the motion becomes stochastic, resembling a random walk. This process, in which the mean energy grows linearly in time, is termed diffusive ionization. In quantum theory, diffusion corresponds to a random walk over a ladder of suitably chosen quantum levels. However, both diagonal and off-diagonal elements of the evolution operator, which describe quantum mechanical amplitudes for transitions between the levels, depend in a quasi-periodic manner on the quantum numbers of the levels involved. Such quasiperiodic behavior is quite analogous to a random one. Electronic wave packets, which initially spread in accordance with the classical laws, tend to remain localized for longer times due to destructive quantum-interference effects. Quantum localization is an analog of the Anderson localization of electronic wave functions propagating in random media [94].

Quantum Scars
Even in the fully chaotic regime, classical phase space contains periodic, although unstable, orbits. Nevertheless, quantum mechanical wave-function amplitudes can become localized around these unstable orbits, resulting in what are called quantum scars. The increased stability of the hydrogen atom at $\tilde{\omega} \approxeq 1.3$ has, in fact, been attributed [95] to the effects of quantum scars. These effects are very sensitive to fluctuations in the driving laser field. Control of the laser noise, therefore, provides a powerful spectroscopic tool to study such quantal phenomena [96]. Using this tool, it has become possible to demonstrate the effects of quantum scars in the intermediate regime of scaled frequencies, i.e., for values less than but close to 1.

78.4 Strong-Field Calculational Techniques

The SS pulse regime requires a nonperturbative solution of the TDSE. We describe here two of the most used approaches: the explicit numerical solution of the TDSE and the Floquet expansion technique. In addition to these, several approximate methods have been proposed.

78.4.1 Floquet Theory

The excitation and ionization dynamics of an atom in a strong laser field can be determined by turning the problem into a time-independent eigenvalue problem [26, 97]. From Floquet's theorem, the eigenfunctions for a perfectly periodic Hamiltonian of the form

$$H = H_0 + \sum_{N\neq 0} H_N \, \mathrm{e}^{-\mathrm{i}N\omega t} \tag{78.29}$$

can be expressed as

$$\Psi(t) = \mathrm{e}^{-\mathrm{i}Xt/\hbar} \sum_N \mathrm{e}^{-\mathrm{i}N\omega t} \psi_N \ . \tag{78.30}$$

Putting this into the time-dependent Schrödinger equation results in an infinite set of coupled Floquet equations for the harmonic components ψ_N. In the velocity gauge, the Floquet equations are

$$(X + N\hbar\omega - H_0)\psi_N = V_+\psi_{N-1} + V_-\psi_{N+1} \ , \tag{78.31}$$

where, for a vector potential of amplitude $\boldsymbol{\mathcal{A}}$,

$$V_+ = -\frac{e}{2mc}\boldsymbol{\mathcal{A}} \cdot \boldsymbol{p} \ , \tag{78.32}$$

and $V_- = V_+^\dagger$. The Eq. (78.31) has been solved, after truncation to a manageable number of terms, using many techniques to provide what are called the quasi-energy states of the laser-atom system. The eigenvalues X of these equations are complex, with $\mathrm{Im}(X)$ giving the decay or ionization rate for the system. The generated rates are found to be very accurate as long as the pulse length of the laser field is not too short, at least hundreds of cycles. The eigenfunctions provide the amplitudes for the photoelectron energy spectra, and the time-dependent dipole of the state can be related to the photoemission spectrum of the system. Yields for slowly varying pulses can be constructed by combining the results from the individual, fixed-intensity calculations [26].

The Floquet method can be applied for any periodic Hamiltonian. In strong enough fields of high frequency, the Floquet equations can be truncated to a very small set in the K–H frame [78].

78.4.2 Direct Integration of the TDSE

Methods for the direct solution of the time-dependent Schrödinger equation (TDSE) are described in general in Chap. 8 and in [98, 99] for multiphoton processes. The wave functions are defined on spatial grids or in terms of an expansion in basis functions. The time evolution is obtained by either explicit or implicit time propagators. All these methods are capable of generating numerically exact results for an atom with a single electron in a short pulsed field for a wide range of pulse shapes, wavelengths, and intensities. The solutions are time-dependent wave functions for the electrons,

which can be analyzed to obtain excitation and ionization rates, photoelectron energies, angular distributions, and photoemission yields. The ability to generate an explicit solution of the TDSE allows the study of arbitrary pulse shapes and provides insight into the excitation dynamics.

For multielectron atoms, one generally has to limit the calculations to that for a single electron in effective potentials, which represent, as well as possible, the influences of the remaining atomic electrons. This approach is called the single-active electron (SAE) approximation, and it gives generally accurate results for systems with no low-lying doubly excited states, for example, the noble gases [98]. In these cases, the excitation dynamics is dominated by the sequential promotion of a single electron at a time.

78.4.3 Volkov States

A laser interacting with a free electron superimposes an oscillatory motion on its drift motion in response to the field. The wave function for an electron with drift velocity $\boldsymbol{v} = \hbar\boldsymbol{k}/m$ is given by

$$\Psi_{\mathrm{V}}(\boldsymbol{r},t) = \exp\left(-\frac{\mathrm{i}}{2m\hbar}\int^{t}\left[\hbar\boldsymbol{k}-\frac{e}{c}\boldsymbol{A}(t')\right]^2\mathrm{d}t'\right)\mathrm{e}^{\mathrm{i}[\boldsymbol{k}-e\boldsymbol{A}(t)/\hbar c]\cdot\boldsymbol{r}}\,, \tag{78.33}$$

where $\boldsymbol{A}(t) = -c\int^{t}\boldsymbol{E}(t')\mathrm{d}t'$ is the vector potential of the field; Ψ_{V} is called a Volkov state. In a linearly polarized field, the electron oscillates along the direction of polarization with an amplitude $\alpha_0 = e\hbar\mathcal{A}/(mc\omega)$. In the strong-field regime, this amplitude can greatly exceed the size of a bound-state orbital. Volkov states provide a useful tool that can be applied in various strong-field approximations discussed in Sect. 78.4.4.

78.4.4 Strong-Field Approximations

There have been several attempts to solve the TDSE in the strong-field limit using approximate but analytic methods. Such strong-field approximations (SFA) typically neglect all the bound states of the atom except for the initial state. In the tunneling regime ($\gamma < 1$), and a quasi-static limit ($\omega \to 0$), one can use a theory [55] in which the ionization occurs due to the tunneling through the Coulomb barrier distorted by the electric field of the laser. The wave function is constructed as a combination of a bare initial wave function of the electron (close to the nucleus) and a wave function describing a motion of the electron in a quasi-static electric field (far from the nucleus). In a second approach [37–39], the elements of the scattering matrix $\hat{S}$ are calculated assuming that initially the electronic wave function corresponds to a bare bound state. On the other hand, the final, continuum states of the electron are described by dressed wave functions that account for the free motion of the electron in the laser field. In the simplest case, such dressed states are Volkov states Eq. (78.33). Alternatively, the time-reversed $\hat{S}$-matrix is obtained by dressing the initial state and using field-free scattering states for the final state.

Yet another method consists of expanding the electronic continuum–continuum dipole matrix elements in terms corresponding to matrix elements for free electrons plus corrections due to the potential [57]. In the latter version of the SFA, the amplitude of the electronic wave function $b(\boldsymbol{p})$, corresponding to an outgoing momentum $\boldsymbol{p}$, is given by

$$b(\boldsymbol{p}) = \mathrm{i}\int_0^{t_{\mathrm{F}}}\mathrm{d}t\,\boldsymbol{d}\left[\boldsymbol{p}-e\boldsymbol{A}(t)/c\right]\cdot\boldsymbol{E}(t)\exp\left[-\mathrm{i}S(t_{\mathrm{F}},t)/\hbar\right]. \tag{78.34}$$

Here, $\boldsymbol{d}[\boldsymbol{p}-e\boldsymbol{A}(t)/c]$ denotes the dipole matrix element for the transition from the initial bound state to the continuum state in which the electron has the kinetic momentum $\boldsymbol{p}-e\boldsymbol{A}(t)/c$, t_{F} is the switch-off time of the laser pulse, and

$$S(t_{\mathrm{F}},t) = \int_t^{t_{\mathrm{F}}}\mathrm{d}t'\left[\frac{\left[\boldsymbol{p}-e\boldsymbol{A}(t')/c\right]^2}{2m}+I_{\mathrm{P}}\right] \tag{78.35}$$

is a quasi-classical action for an electron that is born in the continuum at t and propagates freely in the laser field. The form of the expression Eq. (78.34) is generic to the SFA.

78.4.5 phase-space Averaging Method

The methods of classical mechanics are particularly useful in describing the microwave excitation of highly excited (Rydberg) atoms [89, 90] (Sect. 78.3.8), but have also been applied to describe HHG, stabilization of atoms in super intense fields, and two-electron ionization [100–102].

The classical phase-space averaging method [103] solves Newton's equations of motion

$$\dot{\boldsymbol{r}} = \frac{\boldsymbol{p}}{m}\,, \tag{78.36}$$

$$\dot{\boldsymbol{p}} = -\nabla V(\boldsymbol{r}) - e\boldsymbol{E}(t)\,, \tag{78.37}$$

for an electron interacting with an ion core and an external laser field. A distribution of initial conditions in phase space is chosen to mimic the initial quantum mechanical state of the system, and a sample of classical trajectories is generated. Quantum mechanical averages of physical observables are then identified with ensemble averages of those observables

over the initial distribution. Since the dynamics of multiphoton processes is very complex, the neglected phases in this approach generally cause negligible errors, and the results can be in quite good agreement with quantum calculations. Additionally, an examination of the trajectories provides details of the excitation dynamics, which are often difficult to extract from a complex time-dependent wave function.

78.5 Atto-Nano Physics

The interaction of ultrashort strong laser pulses with larger systems has recently received much attention; it has led to a consequent advance in our understanding of the attosecond to few-femtosecond electronic and nuclear dynamics. For instance, the interaction of atomic clusters with strong ultrafast laser fields has long been known to lead to the formation of nanoplasmas in which there is, on the one hand, a high degree of charge localization and ultrafast dynamics and, on the other hand, the emission of both very energetic (multiple keV) electrons and highly charged ions (in the MeV energy range). Interestingly, the most recent utilization of ultrashort few-cycle pulses ($\approx 10\,\mathrm{fs}$) to trigger the laser–matter processes has succeeded in isolating the electron dynamics from the longer ion dynamics timescale (which can essentially be considered as frozen) revealing a higher degree of fragmentation anisotropy in both electrons and ions compared to the isotropic distributions found from much longer (multicycle) pulses (100 fs).

Additionally, interactions of intense lasers with nanoparticles, such as micron and submicron scale liquid droplets, lead to hot plasma formation. An important role is found for enhanced local fields on the surface of these droplets, the so-called hot spots. Furthermore, studies of driving bound and free charges in larger molecules, e.g., collective electron dynamics in fullerenes and in graphene-like structures, proton migration in hydrocarbon molecules, charge migration in proteins and biomolecules, amongst others, could be included in this category. In addition, biological applications of atto-nano physics could be envisaged as well, e.g., to explain the DNA-protein interactions in solutions of living cells, study the induced covalent crosslink between aromatic amino acids and peptides, and characterize the protein–protein interactions in living cells. In turn, laser-driven broadband electron wavepackets have been used for static and dynamic diffraction imaging of molecules, obtaining structural information with subnanometer spatial and subfemtosecond temporal resolution (Sect. 78.3.4).

Tailored ultrashort and intense fields have also been used to drive electron dynamics and electron or photon emission from (nanostructured) solids (for a recent compilation: [104]). The extraordinary progress seen recently has been largely driven by advances in both experimental, e.g., laser technology, and engineering, e.g., nanofabrication, techniques. Amongst the remarkable achievements in just the most recent years are the demonstration of driving electron currents and switching the conductivity of dielectrics with ultrashort pulses, controlling the light-induced electron emission from nanoparticles and nanotips, and the subcycle-driven photon emission from solids. Furthermore, the intrinsic electron propagation and photoemission processes have been investigated on their natural, attosecond timescales.

A key feature of light-nanostructure interactions is the enhancement, by several orders of magnitude, of the electric near-field and its local confinement on a subwavelength scale. From a theoretical viewpoint, this field localization presents a unique challenge: we have at our disposal strong fields that change on a comparable spatial scale of the oscillatory electron dynamics that are initiated by those same fields. This peculiar property entails profound consequences in the underlying physics of the conventional strong-field phenomena. In particular, it violates one of the main assumptions that the modeling of strong-field interactions is based upon: the spatial homogeneity of laser fields in the volume of the electronic dynamics under scrutiny (this hypothesis was *per se* considered in all the previous sections of this chapter).

Interestingly, an exponential growing attraction in strong-field phenomena induced by plasmonic-enhanced fields was sparked by the controversial work of *Kim* et al. [105]. These authors claimed having observed efficient HHG from noble-gas atoms driven by the plasmonic-enhanced field generated by bow-tie metallic nanostructures. Although later on the interpretation of the outcomes was demonstrated to be incorrect [106], *Kim*'s paper definitively stimulated a steadily constant interest in the plasmonic-enhanced HHG and ATI. We should mention, however, a very recent result of the same group, which clearly seems to be well justified and, as such, opens new perspectives and ways toward efficient HHG using nanostructures [107]. In this recent contribution, the authors demonstrate experimentally plasmonic-driven HHG by devising a metal-sapphire nanostructure that provides a solid tip as the HHG emitter instead of gaseous atoms. Measured EUV spectra show odd-order harmonics up to 60-nm wavelengths, without the plasma atomic lines typically seen when using gaseous atoms as the HHG-driven media. This experimental data confirm that the plasmonic HHG approach is a promising way to make real coherent EUV sources for nanoscale near-field applications in spectroscopy, microscopy, lithography, and attosecond physics (for another recent related experiment, see, e.g., [108]).

Within the conventional assumption, both the laser electric field, $\boldsymbol{E}(\boldsymbol{r},t)$, and the corresponding vector potential, $\boldsymbol{A}(\boldsymbol{r},t)$, are spatially homogeneous in the region where the electron moves and only their time dependence is considered, i.e., $\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}(t)$ in Eq. (78.2) and $\boldsymbol{A}(\boldsymbol{r},t) = \boldsymbol{A}(t)$ in Eq. (78.4). This is a genuine assumption considering the usual electron excursion α_0 is bounded roughly by a few nanometers in the NIR, for typical laser intensities, and sev-

eral tens of nanometers for MIR sources (note that $\alpha_0 \propto \mathcal{E}\lambda^2$, where λ is the wavelength of the driving laser and $\mathcal{E} = \sqrt{I}$, where I is the laser intensity). Hence, electron excursions are very small relative to the spatial variation of the field in the absence of local (or nanoplasmonic) field enhancement. On the contrary, the fields generated using surface plasmons are spatially dependent on a nanometric region. As a consequence, all the standard theoretical tools in the strong-field ionization toolbox described previously, ranging from purely classical to frequently used semiclassical and complete quantum mechanical descriptions, have to be reexamined. For a comprehensive review about atto-nano physics see, e.g., [109]

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