

Chapter 2

Theoretical Tools

2.1 Introduction

My focus in this thesis is high-order harmonic generation (HHG) by an intense ultrafast laser field. In considering the interaction of an atom or a molecule with radiation, there are three basic processes [1, 2]: spontaneous emission in which an atom is treated as a classical oscillating charge that can make a spontaneous transition from an excited state to a state of lower energy, emitting a photon; absorption in which an atom absorbs a photon from an external radiation field, making a transition from a state of lower to a state of higher energy; and stimulated emission in which an atom can also emit a photon under the influence of an external radiation field. To theoretically describe the interaction of an atom or a molecule with an intense laser field, there are generally three approaches: classical theory, semiclassical theory and theory of quantum electrodynamics. In the classical theory, atoms or molecules can be modeled as a group of classical harmonic oscillators, and the laser field is simply treated as a classical electromagnetic field. In the semiclassical theory, the intense laser field is still treated classically by using Maxwell's wave equations, while the atomic or molecular system is described by using the quantum mechanics. In the theory of quantum electrodynamics, both intense laser field and atoms are treated quantum mechanically, and it can describe all phenomena involving charged particles interaction by means of exchange of photons.

As discussed in Chap. 1, HHG process involves the collective effect of many atoms or molecules in the medium interacting with a laser field. This means that a full description of the observed HHG spectra requires not only the treatment of the microscopic nonlinear laser-atom or -molecule interaction, but also the macroscopic propagation of harmonic radiation in the medium. Experimentally high harmonics are usually measured far away from the generating medium, harmonic radiation from the exit of the medium needs to propagate and diverge further in the vacuum. The individual time-dependent dipole induced by the strong oscillating laser field can be obtained by the semiclassical theory, and the fundamental laser field and high harmonic field are treated classically as the electromagnetic fields propagating in

the medium (or in the vacuum).¹ A typical HHG study thus consists of three parts: first, the calculation of microscopic single-atom or -molecule response by solving the time-dependent Schrödinger equation; second, the macroscopic propagation of fundamental and high-harmonic fields by solving the three-dimensional Maxwell's wave equations; third, the further propagation of high harmonics in the vacuum (probably involving the complicated optical system) by constructing an optical $ABCD$ matrix. Each part will be discussed in detail in the following.

2.2 Time-Dependent Schrödinger Equation

In this section, I am concerned about constructing the time-dependent Schrödinger equation (TDSE) of an atom in the laser field. Two approximate approaches, strong-field approximation (SFA) and quantitative rescattering (QRS) theory, are applied to solve it. The formulation of the TDSE for a molecular target is beyond the scope of this thesis, only two approximated solutions of TDSE based on the SFA and the QRS theory are presented. In this thesis the laser field is limited to a linearly polarized light.

2.2.1 Semiclassical Theory

The classical electromagnetic field is described by the Maxwell's wave equation. The vector potential of a monochromatic electromagnetic field (with a linear polarization) is

$$\vec{A}(\vec{r}, t) = \vec{\epsilon} A_0 \sin(\vec{k} \cdot \vec{r} - \omega_0 t), \quad (2.1)$$

where $\vec{k}$ is the wave vector. And angular frequency ω_0 and wave number k (the magnitude of $\vec{k}$) are related by

$$\omega_0 = kc, \quad (2.2)$$

where c is the speed of light. Vector potential $\vec{A}$ in the direction specified by the unit vector $\vec{\epsilon}$ (polarization vector) has an amplitude $|A_0|$. Laser wavelength is much larger than the size of an atom, characterized by the Bohr radius, a_0 , thus $k \cdot a_0 \ll 1$, and then a dipole approximation is applied here. Equation (2.1) can be written as

$$\vec{A}(t) = \vec{\epsilon} A_0 \sin(\omega_0 t). \quad (2.3)$$

Using single-active electron (SAE) approximation [3, 4], i.e., all electrons in an atom are bound except the valence electron, the time-dependent Schrödinger equation of a valence electron in a laser field can be written as

¹ The influence of atoms or molecules on the external field is also included in this treatment.

$$\begin{aligned}
i\hbar \frac{\partial}{\partial t} \psi(\vec{r}, t) &= \left[\frac{1}{2m_e} (\vec{p} - \frac{e}{c} \vec{A})^2 + V(\vec{r}) \right] \psi(\vec{r}, t) \\
&= \left[\frac{1}{2m_e} (p^2 - \frac{e}{c} \vec{p} \cdot \vec{A} - \frac{e}{c} \vec{A} \cdot \vec{p} + \frac{e^2}{c^2} A^2) + V(\vec{r}) \right] \psi(\vec{r}, t), \quad (2.4)
\end{aligned}$$

where $V(\vec{r})$ is the atomic potential. To obtain Eq. (2.4), the Coulomb gauge has been adopted, which is defined in the following:

$$\nabla \cdot \vec{A}(t) = 0, \quad (2.5)$$

and the scalar potential has been taken as $\phi = 0$. The momentum operator $\vec{p}$ is

$$\vec{p} = -i\hbar \nabla. \quad (2.6)$$

Using the commutation relation between $\vec{p}$ and $\vec{A}$:

$$\vec{p} \cdot \vec{A} - \vec{A} \cdot \vec{p} = -i\hbar \nabla \cdot \vec{A}, \quad (2.7)$$

Eq. (2.4) can be written as

$$i\hbar \frac{\partial}{\partial t} \psi(\vec{r}, t) = \left[-\frac{\hbar^2}{2m_e} \nabla^2 - \frac{e}{cm_e} \vec{A} \cdot \vec{p} + \frac{e^2}{2m_e c^2} A^2 + V(\vec{r}) \right] \psi(\vec{r}, t). \quad (2.8)$$

Under the dipole approximation, $\vec{A}(t)$ only depends on time, so the term with A^2 also depends on time only, which could be eliminated from Eq. (2.8) in terms of the unitary transformation:

$$\Psi_V(\vec{r}, t) = \exp \left(\frac{e^2}{2i\hbar m_e c^2} \int_0^t A^2(t') dt' \right) \psi(\vec{r}, t). \quad (2.9)$$

And then Eq. (2.8) can be simplified as

$$i\hbar \frac{\partial}{\partial t} \Psi_V(\vec{r}, t) = \left[-\frac{\hbar^2}{2m_e} \nabla^2 - \frac{e}{cm_e} \vec{A} \cdot \vec{p} + V(\vec{r}) \right] \Psi_V(\vec{r}, t). \quad (2.10)$$

Equation (2.10) is the time-dependent Schrödinger equation of an atom in the velocity gauge.

Actually the TDSE in the length gauge is much preferred. One can introduce a unitary transformation

$$\Psi_V(\vec{r}, t) = \exp \left(\frac{ie}{c\hbar} \vec{A} \cdot \vec{r} \right) \Psi_L(\vec{r}, t), \quad (2.11)$$

Eq. (2.10) can be expressed as

$$i\hbar \frac{\partial}{\partial t} \Psi_L(\vec{r}, t) = \left[-\frac{\hbar^2}{2m_e} \nabla^2 + \frac{e^2}{2m_e c^2} A^2 + \frac{e}{c} \frac{\partial \vec{A}}{\partial t} \cdot \vec{r} + V(\vec{r}) \right] \Psi_L(\vec{r}, t). \quad (2.12)$$

Using the transformation in Eq. (2.9), the term including A^2 could be eliminated. Because the effect of an electron with a magnetic field is much weaker compared to an electric field, the effect of the magnetic field can be neglected. The strength of electric field $\vec{E}(t)$ is related to the vector potential $\vec{A}(t)$ by

$$\vec{E}(t) = -\frac{1}{c} \frac{\partial \vec{A}(t)}{\partial t}. \quad (2.13)$$

Equation (2.12) can be further simplified as

$$i\hbar \frac{\partial}{\partial t} \Psi_L(\vec{r}, t) = \left[-\frac{\hbar^2}{2m_e} \nabla^2 + V(\vec{r}) - e\vec{r} \cdot \vec{E}(t) \right] \Psi_L(\vec{r}, t). \quad (2.14)$$

Equation (2.14) is the time-dependent Schrödinger equation of an atom in the length gauge. This is the equation used to study the microscopic nonlinear laser-atom interaction for harmonic radiation, and I use “ $\Psi(\vec{r}, t)$ ” without “ L ” to note the time-dependent wave function in the length gauge for simplicity.

Equation (2.14) can be solved by expanding the $\Psi(\vec{r}, t)$ in terms of eigenfunctions, $R_{nl}(r)Y_{lm}(\hat{r})$, of the laser-free Hamiltonian, within the box of $r \in [0, r_{max}]$,

$$\Psi(\vec{r}, t) = \sum_{nl} c_{nl}(t) R_{nl}(r) Y_{lm}(\hat{r}), \quad (2.15)$$

where radial functions $R_{nl}(r)$ are expanded in a discrete variable representation (DVR) basis set [5] associated with Legendre polynomials, while c_{nl} are calculated using the split-operator method [6].

2.2.2 Strong-Field Approximation

Based on strong-field approximation (SFA), Lewenstein et al. [7] proposed an analytic form to solve the time-dependent Schrödinger equation of an atom in a low-frequency laser field in 1994. Their theory recovers the semiclassical interpretation of HHG by Krause et al. [3] and Corkum [8] as discussed in Sect. 1.2.1, and also takes into account of quantum effects such as tunneling ionization, wave packet spreading (or quantum diffusion) and interferences. This approach will be called either “Lewenstein model” or “SFA” interchangeably in this thesis. SFA has also been applied to study the characteristics of HHG from molecular targets [9–16]. In the following I will derive the Lewenstein model for an atomic target in detail, and then it is extended for a molecular target straightforwardly.

There are two main assumptions in this model: (1) all bound states in an atom are neglected excepted for the ground state; (2) the electron in the continuum state is treated as a free particle without the influence of the Coulomb potential. The depletion of the ground state was neglected initially, and later on added by Antoine et al. [17].

Considering an atom in the SAE approximation under the influence of a laser field $E(t)$ with the linear polarization in the x direction, which satisfies Eq. (2.14), the time-dependent wave function can be expanded as (atomic units are used):²

$$|\Psi(t)\rangle = e^{iI_p t} \left[a(t)|0\rangle + \int d^3\vec{v} b(\vec{v}, t)|\vec{v}\rangle \right], \quad (2.16)$$

where $a(t) \approx 1$ is the ground-state amplitude, $|0\rangle$ denotes the wave function of ground state, and $b(\vec{v}, t)$ are the amplitudes of the corresponding continuum states. The equation for $b(\vec{v}, t)$ can be written as

$$\dot{b}(\vec{v}, t) = -i \left(\frac{\vec{v}^2}{2} + I_p \right) b(\vec{v}, t) + E(t) \frac{\partial b(\vec{v}, t)}{\partial v_x} - i E(t) d_x(\vec{v}). \quad (2.17)$$

Here $\vec{d}(\vec{v}) = \langle \vec{v} | \vec{x} | 0 \rangle$ is the transition dipole matrix element from the bound to the free state, and $d_x(\vec{v})$ is the component parallel to the polarization axis. Equation (2.17) can be solved exactly and $b(\vec{v}, t)$ can be written in the closed form,

$$b(\vec{v}, t) = -i \int_0^t dt' E(t') d_x(\vec{v} - \vec{A}(t) + \vec{A}(t')) \times \exp \left\{ -i \int_{t'}^t dt'' \left[(\vec{v} - \vec{A}(t) + \vec{A}(t''))^2 / 2 + I_p \right] \right\}, \quad (2.18)$$

where $\vec{A}(t)$ is the vector potential of laser field defined in Eq. (2.13).

To calculate the parallel component (with respect to the laser polarization) of the time-dependent dipole moment, one needs to evaluate $D(t) = \langle \Psi(t) | x | \Psi(t) \rangle$. Using Eqs. (2.16) and (2.18), one can obtain

$$D(t) = \int d^3\vec{v} d_x^*(\vec{v}) b(\vec{v}, t) + c.c.. \quad (2.19)$$

In the above formula, only the transition back to the ground state is considered, and the contribution from continuum to continuum part is neglected. The velocity operator is defined as follows³

² In the derivation, $e = -1$ in the atomic unit. This convention is different from what has been used by Lewenstein et al. [7], and this results in a sign change in front of $E(t)$ and $A(t)$.

³ The velocity operator in Lewenstein et al. [7] is $\vec{v} = \vec{p} - \vec{A}(t)$. The discrepancy is due to the convention of e in the atomic unit.

$$\vec{v} = \vec{p} + \vec{A}(t), \quad (2.20)$$

the final expression of Eq. (2.19) is

$$D(t) = -i \int_0^t dt' \int d^3 \vec{p} d_x^*(\vec{p} + \vec{A}(t)) \exp[-i S(\vec{p}, t, t')] E(t') d_x(\vec{p} + \vec{A}(t')) + c.c., \quad (2.21)$$

where

$$S(\vec{p}, t, t') = \int_{t'}^t dt'' \{ [\vec{p} + \vec{A}(t'')]^2 / 2 + I_p \}. \quad (2.22)$$

Equation (2.21) has a clear physical interpretation, and it is actually a sum of probability amplitudes for the following processes: $E(t') d_x(\vec{p} + \vec{A}(t'))$ is the probability for an electron to make a transition to the continuum at time t' with the canonical momentum $\vec{p}$; $\exp[-i S(\vec{p}, t, t')]$ is a phase factor when the electronic wave function is propagated from time t' until t , where $S(\vec{p}, t, t')$ is the quasi-classical action; $d_x^*(\vec{p} + \vec{A}(t))$ is a transition amplitude when the electron recombines to the parent ion at time t .

The major contribution to the integral over $\vec{p}$ in Eq. (2.21) is from the stationary points of the classical action,

$$\nabla_{\vec{p}} S(\vec{p}, t, t') = 0, \quad (2.23)$$

so the integral over $\vec{p}$ could be performed using a saddle-point method. Defining the electron return (or excursion) time $\tau = t - t'$, Eq. (2.21) is written as

$$D(t) = -i \int_0^\infty d\tau \left[\frac{\pi}{\epsilon + i\tau/2} \right]^{3/2} d_x^*(\vec{p}_{st} + \vec{A}(t)) \exp[-i S(\vec{p}_{st}, t, \tau)] \\ \times E(t - \tau) d_x(\vec{p}_{st} + \vec{A}(t - \tau)) + c.c., \quad (2.24)$$

where

$$\vec{p}_{st} = - \int_{t-\tau}^t dt'' \vec{A}(t'') / \tau, \quad (2.25)$$

$$S(\vec{p}_{st}, t, \tau) = I_p \tau - \frac{1}{2} \vec{p}_{st}^2 \tau + \frac{1}{2} \int_{t-\tau}^t \vec{A}^2(t'') dt''. \quad (2.26)$$

Here the small positive constant of ϵ comes from the regularized Gaussian integration over $\vec{p}$ around the saddle-point.

It is straightforward to put the depletion of ground state in Eq. (2.24), it becomes

$$D(t) = -i \int_0^\infty d\tau \left[\frac{\pi}{\epsilon + i\tau/2} \right]^{3/2} d_x^*(\vec{p}_{st} + \vec{A}(t)) a^*(t) \exp[-i S(\vec{p}_{st}, t, \tau)] \\ \times E(t - \tau) d_x(\vec{p}_{st} + \vec{A}(t - \tau)) a(t - \tau) + c.c.. \quad (2.27)$$

The ground-state amplitude $a(t)$ can be expressed as [9, 17]

$$a(t) = \exp \left(- \int_0^t \frac{\gamma(t')}{2} dt' \right). \quad (2.28)$$

Here $\gamma(t')$ is the ionization rate.

Equation (2.27) is the generally used formula of Lewenstein model to calculate the single-atom harmonic radiation induced by an intense laser field. For single-molecule HHG, I assume that linear molecules are aligned along the x axis, while laser field $E(t)$ is linearly polarized on the x - y plane with an angle θ with respect to the molecular axis. The parallel component of time-dependent dipole moment can be expressed as [9, 10]

$$\begin{aligned} D_{\parallel}(t) = & -i \int_0^{\infty} d\tau \left[\frac{\pi}{\epsilon + i\tau/2} \right]^{3/2} [\cos \theta d_x^*(t) + \sin \theta d_y^*(t)] a^*(t) \\ & \times [\cos \theta d_x(t - \tau) + \sin \theta d_y(t - \tau)] a(t - \tau) E(t - \tau) \\ & \times \exp[-iS(\vec{p}_{st}, t, \tau) + c.c.], \end{aligned} \quad (2.29)$$

where $\vec{d}(t) = \vec{d}[\vec{p}_{st}(t, \tau) + \vec{A}(t)]$ and $\vec{d}(t - \tau) = \vec{d}[\vec{p}_{st}(t, \tau) + \vec{A}(t - \tau)]$ are transition dipole matrix elements. The perpendicular component $D_{\perp}(t)$ can be given in a similar formula with $[\cos \theta d_x^*(t) + \sin \theta d_y^*(t)]$ replaced by $[\sin \theta d_x^*(t) - \cos \theta d_y^*(t)]$ in Eq. (2.29).

Equation (2.29) is the extended formula of Lewenstein model to calculate the single-molecule HHG induced by an intense laser field. In the SFA, the transition dipole moment $\vec{d}(\vec{p})$ is given by $\langle \vec{p} | \vec{r} | 0 \rangle$ with the continuum state approximated by a plane wave $|\vec{p}\rangle$. For hydrogenlike atoms, the dipole matrix element for transition from the ground state to a continuum state is given in the form

$$d(p) = i \frac{2^{7/2} (2I_p)^{5/4}}{\pi} \frac{p}{(p^2 + 2I_p)^3}. \quad (2.30)$$

For other atoms and molecules, $\vec{d}(\vec{p})$ is calculated numerically with the known wave function of ground state.⁴

2.2.3 Quantitative Rescattering Theory

Lewenstein model is well-known to give good results for the harmonics with high-photon energies, especially for the cutoff harmonics, but it fails to predict harmonics in the lower plateau since the effect of the Coulomb potential is not included. To

⁴ In the calculation, the ground-state electronic wave function of an atom or a molecule can be obtained by using quantum chemistry codes such as GAMESS or GAUSSIAN [18].

improve the SFA, a quantitative rescattering (QRS) theory has been developed in Prof. Lin's group [19–21]. The QRS theory states that single-atom or -molecule HHG can be expressed as a product of a returning electron wave packet and a photorecombination cross section (PRCS) of laser-free continuum electron back to the initial bound state. Based on the Lewenstein model, each step in the three-step model can be quantified, see Eq. (2.21). The last step, i.e., recombination, is not described precisely due to a plane-wave approximation, but the motion of an electron after tunneling ionization governed mostly by the laser field has been well treated in the SFA. One can extract an accurate returning electron wave packet by using the SFA. The PRCS as the other integral part in the QRS theory is only determined by the structure of an atomic or molecular target, and it can be accurately calculated by solving the stationary Schrödinger equation. Equations (2.27) and (2.29) for calculating the HHG are expressed in time domain, while the QRS theory deals with the induced dipole moment in frequency domain. A detailed discussion of the QRS theory for HHG is given in [20]. In the following I will briefly describe the QRS theory for atomic and molecular targets separately.

1. Atomic Target

According to the QRS theory, the induced dipole moment $D(\omega)$ of an atomic target can be written as [22]

$$D(\omega) = W(\omega)d(\omega), \quad (2.31)$$

where $d(\omega)$ is the complex photorecombination (PR) transition dipole matrix element, and $W(\omega)$ is the complex microscopic electron wave packet. $|W(\omega)|^2$ describes the flux of returning electrons and it is the property of laser only. The QRS replaces the plane wave used in the SFA by an accurate scattering wave in the calculation of PR transition dipole matrix elements, while the microscopic returning electron wave packet is the same as that in the SFA. The harmonic frequency ω is related to the electron momentum p by

$$\hbar\omega = \frac{p^2}{2m_e} + I_p. \quad (2.32)$$

In practical applications, the QRS obtains the induced dipole moment by

$$D^{\text{QRS}}(\omega) = D^{\text{SFA}}(\omega) \frac{d^{\text{QRS}}(\omega)}{d^{\text{SFA}}(\omega)}, \quad (2.33)$$

where both $D^{\text{SFA}}(\omega)$ and $d^{\text{QRS}}(\omega)$ are complex numbers, while $d^{\text{SFA}}(\omega)$ is either a pure real or pure imaginary number. Within the SAE approximation, $d^{\text{QRS}}(\omega)$ is calculated by using “exact” numerical wave functions for bound and continuum states. For Ar, the model potential is given by Müller [23],

$$V(r) = -[1 + Ae^{-Br} + (17 - A)e^{-Cr}]/r, \quad (2.34)$$

with $A = 5.4$, $B = 1$ and $C = 3.682$. In this model, the spin-orbit interaction is neglected. Parameters have been chosen such that the minimum in the photoionization (or photorecombination) cross section is reproduced correctly. Tong and Lin [24] proposed a model potential for rare atoms,

$$V(r) = -\frac{Z_c + a_1 e^{-a_2 r} + a_3 r e^{-a_4 r} + a_5 e^{-a_6 r}}{r}, \quad (2.35)$$

where Z_c is the charge seen by the active electron asymptotically, and $a_1, \dots, a_6$ are parameters obtained by fitting $V(r)$ to the numerical potential from the self-interaction free density functional theory. Note that in principle the parameters in Eq. (2.33) can be generalized to many-electron wave functions if needed.

2. Molecular Target

Within the QRS theory, the induced dipole moment $D(\omega, \theta)$ of a fixed-in-space molecule is given explicitly by⁵

$$D(\omega, \theta) = N(\theta)^{1/2} W(\omega) d(\omega, \theta), \quad (2.36)$$

where $N(\theta)$ is the alignment-dependent ionization probability, $W(\omega)$ is the microscopic electron wave packet, and $d(\omega, \theta)$ is the alignment-dependent transition dipole (complex in general). Here θ is the angle between the molecular axis with respect to the laser's polarization. Only the linear molecules are investigated, and the parallel component of HHG with respect to the laser polarization is considered in this thesis. Thus only the parallel component of transition dipole $d(\omega, \theta)$ is needed in the calculation. Note that $W(\omega)$ does not depend on the alignment angle θ .

The wave packet $W(\omega)$ can be calculated in two ways. First, it can be calculated formally as

$$W(\omega) = \frac{D(\omega, \theta)}{N(\theta)^{1/2} d(\omega, \theta)}. \quad (2.37)$$

Here $D(\omega, \theta)$ for a fixed alignment angle θ can be calculated by using Eq. (2.29). And then $N(\theta)$ and $d(\omega, \theta)$ are also calculated in the frame of the SFA, where the continuum waves are replaced by the plane waves. Since the wave packet $W(\omega)$ is independent of the alignment angle θ , it could be calculated only once for a given angle θ . The second approach of obtaining the wave packet is to use a reference atom with a similar ionization potential. For the reference atom, one can perform either TDSE or SFA to calculate the induced dipole moment $D(\omega)$, and then following Eq. (2.31) the wave packet can be expressed as

$$W(\omega) = \frac{D^{ref}(\omega)}{d^{ref}(\omega)}. \quad (2.38)$$

⁵ The induced dipole moment is actually expressed in the molecular (or body-fixed) frame.

Note that the ionization probability of an atom is incorporated into $W(\omega)$. In the QRS theory, single-molecule induced dipole moment is then obtained from Eq. (2.36) by combining the electron wave packet with the accurate $d(\omega, \theta)$ obtained from the quantum chemistry code [25, 26] and the tunneling ionization rate $N(\theta)$ obtained from the MO-ADK theory [27].

It has been well documented that QRS results are nearly as accurate as those obtained from the TDSE whenever the accurate results from the latter can be obtained, i.e., including the atom in the SAE approximation [22] and the H_2^+ molecular ion [28]. Applications of the QRS for HHG from single molecules have been investigated in [20, 29, 30].

2.3 Maxwell's Wave Equation

2.3.1 Fundamental Laser Field in an Atomic Target

In a dense and ionizing gaseous medium, the macroscopic propagation of a driving laser pulse is affected by refraction, nonlinear self-focusing, ionization and plasma defocusing.⁶ The pulse evolution in such an atomic medium is usually described by a three-dimensional (3-D) Maxwell's wave equation [31–33]:

$$\nabla^2 E_1(r, z, t) - \frac{1}{c^2} \frac{\partial^2 E_1(r, z, t)}{\partial t^2} = \mu_0 \frac{\partial J_{\text{abs}}(r, z, t)}{\partial t} + \frac{\omega_0^2}{c^2} (1 - \eta_{\text{eff}}^2) E_1(r, z, t), \quad (2.39)$$

where $E_1(r, z, t)$ is the transverse electric field of fundamental laser pulse with the angular frequency ω_0 . In cylindrical coordinates, $\nabla^2 = \nabla_{\perp}^2 + \partial^2/\partial z^2$, where z is the axial propagation direction. The effective refractive index η_{eff} of gas medium is written as

$$\eta_{\text{eff}}(r, z, t) = \eta_0(r, z, t) + \eta_2 I(r, z, t) - \frac{\omega_p^2(r, z, t)}{2\omega_0^2}. \quad (2.40)$$

The first term $\eta_0 = 1 + \delta_1 - i\beta_1$ takes refraction (δ_1) and absorption (β_1) effects of neutral atoms into account, the second term accounts for the optical Kerr nonlinearity depending on the laser intensity $I(t)$, and the third term is from the free electrons, which contains the plasma frequency $\omega_p = [e^2 n_e(t)/(\epsilon_0 m_e)]^{1/2}$, where m_e and e are mass and charge of an electron, respectively, and $n_e(t)$ is the free electron density. The absorption term $J_{\text{abs}}(t)$ due to ionization is expressed as [34, 35]

$$J_{\text{abs}}(t) = \frac{\gamma(t) n_e(t) I_p E_1(t)}{|E_1(t)|^2}, \quad (2.41)$$

⁶ The propagation of laser pulse in an isotropic molecular medium is similar to that in an atomic medium. In general, the optical properties are anisotropic, so the laser evolution in a partially aligned molecular medium should also take the alignment effects into account.

where $\gamma(t)$ is the ionization rate, and I_p is the ionization potential. This term is usually small under the conditions for high-harmonic generation [34, 35].

The absorption effect (β_1) on the fundamental laser pulse caused by neutral atoms is in general small, so it is neglected. Only real terms are kept in the refractive index η_{eff} , and Eq. (2.39) can be written as

$$\begin{aligned} \nabla^2 E_1(r, z, t) - \frac{1}{c^2} \frac{\partial^2 E_1(r, z, t)}{\partial t^2} &= \mu_0 \frac{\partial J_{\text{abs}}(r, z, t)}{\partial t} \\ &+ \frac{\omega_p^2}{c^2} E_1(r, z, t) - 2 \frac{\omega_0^2}{c^2} (\delta_1 + \eta_2 I) E_1(r, z, t). \end{aligned} \quad (2.42)$$

By going to a moving coordinate frame ($z' = z$ and $t' = t - z/c$) and neglecting $\partial^2 E_1 / \partial z'^2$ since the z' dependence of electric field is very slow, i.e., the slowly varying envelope approximation is applied, one can obtain [36]

$$\begin{aligned} \nabla_{\perp}^2 E_1(r, z', t') - \frac{2}{c} \frac{\partial^2 E_1(r, z', t')}{\partial z' \partial t'} &= \mu_0 \frac{\partial J_{\text{abs}}(r, z', t')}{\partial t'} \\ &+ \frac{\omega_p^2}{c^2} E_1(r, z', t') - 2 \frac{\omega_0^2}{c^2} (\delta_1 + \eta_2 I) E_1(r, z', t'). \end{aligned} \quad (2.43)$$

The temporal derivative in Eq. (2.43) can be eliminated by a Fourier transform, yielding the equation

$$\nabla_{\perp}^2 \tilde{E}_1(r, z', \omega) - \frac{2i\omega}{c} \frac{\partial \tilde{E}_1(r, z', \omega)}{\partial z'} = \tilde{G}(r, z', \omega), \quad (2.44)$$

where

$$\tilde{E}_1(r, z', \omega) = \hat{F}[E_1(r, z', t')], \quad (2.45)$$

and

$$\begin{aligned} \tilde{G}(r, z', \omega) &= \hat{F} \left\{ \mu_0 \frac{\partial J_{\text{abs}}(r, z', t')}{\partial t'} + \frac{\omega_p^2}{c^2} E_1(r, z', t') \right. \\ &\quad \left. - 2 \frac{\omega_0^2}{c^2} [\delta_1 + \eta_2 I(r, z', t')] E_1(r, z', t') \right\}, \end{aligned} \quad (2.46)$$

where $\hat{F}$ is the Fourier transform operator acting on temporal coordinate.

The plasma frequency $\omega_p(r, z', t')$ is determined by free-electron density $n_e(t')$, which can be calculated as following

$$n_e(r, z', t') = n_0 \{ 1 - \exp[-\int_0^{t'} \gamma(r, z', \tau) d\tau] \}, \quad (2.47)$$

where n_0 is the neutral atom density, and $\gamma(r, z', \tau)$ is the ionization rate calculated from Ammosov-Delone-Krainov (ADK) theory [24, 27, 37]. The refraction coefficient δ_1 , depending on the pressure and the temperature of gas medium, is obtained from the Sellmeier equation [38, 39]. The nonlinear refractive index η_2 , also depending on the pressure of gas medium, can be calculated through the third-order susceptibility $\chi^{(3)}$, which can be measured experimentally [40, 41]. Note that the relationship between η_2 and $\chi^{(3)}$ in Koga et al. [42] differs from that in Boyd [43] since the latter is derived by using the time-averaged intensity of optical field.

At the entrance of a gas jet ($z' = z_{\text{in}}$), the fundamental laser field is assumed to be either Gaussian or truncated Bessel in space, and in time it has a Gaussian or cosine-squared envelop.⁷ These will be specified whenever the calculated results are present. The pressure is assumed constant within the gas jet in this thesis. It can be also assumed that the atomic density distribution follows a Lorentzian [44] or Gaussian [45] profile along the propagation direction.

2.3.2 High-Harmonic Field of an Atomic Target

The 3-D propagation equation of high-harmonic field in an atomic medium is described by [36, 34, 46]

$$\nabla^2 E_h(r, z, t) - \frac{1}{c^2} \frac{\partial^2 E_h(r, z, t)}{\partial t^2} = \mu_0 \frac{\partial^2 P(r, z, t)}{\partial t^2}, \quad (2.48)$$

where $P(r, z, t)$ is the polarization depending upon the applied optical field $E_1(r, z, t)$. In this equation, the free-electron dispersion is neglected because the frequencies of high harmonics are much higher than the plasma frequency. Again going to a moving coordinate frame and neglecting $\partial^2 E_h / \partial z'^2$ (applying the slowly varying envelope approximation), Eq. (2.48) becomes

$$\nabla_{\perp}^2 E_h(r, z', t') - \frac{2}{c} \frac{\partial^2 E_h(r, z', t')}{\partial z' \partial t'} = \mu_0 \frac{\partial^2 P(r, z', t')}{\partial t'^2}. \quad (2.49)$$

One can eliminate the temporal derivative by a Fourier transform, obtaining the equation

$$\nabla_{\perp}^2 \tilde{E}_h(r, z', \omega) - \frac{2i\omega}{c} \frac{\partial \tilde{E}_h(r, z', \omega)}{\partial z'} = -\omega^2 \mu_0 \tilde{P}(r, z', \omega), \quad (2.50)$$

where

$$\tilde{E}_h(r, z', \omega) = \hat{F}[E_h(r, z', t')], \quad (2.51)$$

⁷ See Appendix D for details.

and

$$\tilde{P}(r, z', \omega) = \hat{F}[P(r, z', t')]. \quad (2.52)$$

The source term on the right-hand side of Eq. (2.50) describes the response of medium to the laser field and includes both linear and nonlinear terms. It is convenient to separate the polarization into linear and nonlinear components as: $\tilde{P}(r, z', \omega) = \chi^{(1)}(\omega)\tilde{E}_h(r, z', \omega) + \tilde{P}_{nl}(r, z', \omega)$, where the linear susceptibility $\chi^{(1)}(\omega)$ includes both linear dispersion and absorption through its real and imaginary parts, respectively. The nonlinear polarization term $\tilde{P}_{nl}(r, z', \omega)$ can be expressed as

$$\tilde{P}_{nl}(r, z', \omega) = \hat{F}\{[n_0 - n_e(r, z', t')]D(r, z', t')\}, \quad (2.53)$$

where the free electron density $n_e(r, z', t')$ is calculated from Eq. (2.47), and $D(r, z', t')$ is the single-atom induced dipole moment caused by the fundamental driving laser field.

The refractive index $n(\omega) = \sqrt{1 + \chi^{(1)}(\omega)/\epsilon_0}$ [43] (which is valid only off resonance or for the small absorption) is related to atomic scattering factors by

$$n(\omega) = 1 - \delta_h(\omega) - i\beta_h(\omega) = 1 - \frac{1}{2\pi}n_0r_0\lambda^2(f_1 + if_2), \quad (2.54)$$

where r_0 is the classical electron radius, λ is the wavelength of the harmonic, n_0 is again the neutral atom density, and f_1 and f_2 are atomic scattering factors obtained from [47, 48]. Note that $\delta_h(\omega)$ and $\beta_h(\omega)$ account for dispersion and absorption of the medium on high harmonics, respectively. Finally Eq. (2.50) can be written as

$$\begin{aligned} \nabla_{\perp}^2 \tilde{E}_h(r, z', \omega) - \frac{2i\omega}{c} \frac{\partial \tilde{E}_h(r, z', \omega)}{\partial z'} - \frac{2\omega^2}{c^2} (\delta_h + i\beta_h) \tilde{E}_h(r, z', \omega) \\ = -\omega^2 \mu_0 \tilde{P}_{nl}(r, z', \omega), \end{aligned} \quad (2.55)$$

where the nonlinear polarization as the source of high-harmonic field is explicitly given. After the propagation in the medium, one can obtain near-field harmonics at the exit face of gas jet ($z' = z_{\text{out}}$).

2.3.3 High-Harmonic Field of Aligned Molecules

In general both the fundamental laser field and the high-harmonic field are modified when they copropagate through a macroscopic medium. If both pressure and laser intensity are low, the effects of dispersion, Kerr nonlinearity and plasma defocusing on the fundamental laser field can be neglected.⁸ In other words, the source term in Eq. (2.39) can be taken as zero, and the fundamental field is not modified through the

⁸ This is also true for laser propagation in a partially aligned molecular medium.

medium. Under these conditions the profile of fundamental laser field in space (in the vacuum) can be expressed in an analytical form.⁹ For the harmonic field, dispersion and absorption effects of the medium, explicitly expressed as a dispersion-absorption term in Eq. (2.55) are not included when the gas pressure is low. These effects would become important for the high gas pressure. For molecular targets, I will only focus on the experiments carried out under the conditions of low laser intensity and low gas pressure, and only include induced dipoles for the generated harmonic field.

The 3-D Maxwell's wave equation for the high harmonics in a partially aligned molecular gas medium is [49, 50]

$$\nabla^2 E_h^\parallel(r, z, t, \alpha) - \frac{1}{c} \frac{\partial^2 E_h^\parallel(r, z, t, \alpha)}{\partial^2 t} = \mu_0 \frac{\partial^2 P_{nl}^\parallel(r, z, t, \alpha)}{\partial^2 t}. \quad (2.56)$$

Here $E_h^\parallel(r, z, t, \alpha)$ and $P_{nl}^\parallel(r, z, t, \alpha)$ are the parallel components (with respect to the polarization direction of the generating laser) of harmonic electric field and the nonlinear polarization caused by fundamental laser, respectively. α is the pump-probe angle, i.e., the angle between aligning (pump) and generating (probe) laser polarizations.

The nonlinear polarization term can be expressed as

$$P_{nl}^\parallel(r, z, t, \alpha) = [n_0 - n_e(r, z, t, \alpha)] D^{\parallel, \text{tot}}(r, z, t, \alpha), \quad (2.57)$$

where $n_0 - n_e(r, z, t, \alpha)$ gives the density of remaining neutral molecules, and $D^{\parallel, \text{tot}}(r, z, t, \alpha)$ is the parallel component of induced single-molecule dipole over a number of active electrons (including the effects from outermost and inner molecular orbitals).

Note that $\theta(\theta')$ and $\phi(\phi')$ are polar and azimuthal angles of the molecular axis in the frame attached to the pump (probe) laser field [20]. These angles are related by

$$\cos \theta = \cos \theta' \cos \alpha + \sin \theta' \sin \alpha \cos \phi'. \quad (2.58)$$

The alignment distribution in the "probe" frame at the fixed time delay between pump and probe lasers is

$$\rho(\theta', \phi', \alpha) = \rho[\theta(\theta'), \phi(\phi'), \alpha]. \quad (2.59)$$

The free-electron density $n_e(t, \alpha)$ in Eq. (2.57) can be calculated as follows

$$n_e(t, \alpha) = \int_0^{2\pi} \int_0^\pi n_e(t, \theta') \rho(\theta', \phi', \alpha) \sin \theta' d\theta' d\phi'. \quad (2.60)$$

Here $n_e(t, \theta')$ is the alignment-dependent free-electron density, obtained from

⁹ If laser beam can be considered as a Gaussian beam, its spatial and temporal dependence is given approximately in Appendix D.3.

$$n_e(t, \theta') = n_0 \{1 - \exp[-\int_0^t \gamma(\tau, \theta') d\tau]\}, \quad (2.61)$$

where $\gamma(\tau, \theta')$ is the alignment-dependent ionization rate calculated by the MO-ADK theory [27] for each individual molecular orbital.

By going to a moving coordinate frame ($z' = z$ and $t' = t - z/c$) again, Eq. (2.56) can be written in the frequency domain as

$$\nabla_{\perp}^2 \tilde{E}_h^{\parallel}(r, z', \omega, \alpha) - \frac{2i\omega}{c} \frac{\partial \tilde{E}_h^{\parallel}(r, z', \omega, \alpha)}{\partial z'} = -\omega^2 \mu_0 \tilde{P}_{nl}^{\parallel}(r, z', \omega, \alpha), \quad (2.62)$$

where

$$\tilde{E}_h^{\parallel}(r, z', \omega, \alpha) = \hat{F}[E_h^{\parallel}(r, z', t', \alpha)], \quad (2.63)$$

and

$$\tilde{P}_{nl}^{\parallel}(r, z', \omega, \alpha) = \hat{F}[P_{nl}^{\parallel}(r, z', t', \alpha)]. \quad (2.64)$$

After the propagation in the medium, one can obtain the parallel component of near-field harmonics on the exit face of gas jet ($z' = z_{\text{out}}$). For isotropically distributed molecules and partially aligned molecules with $\alpha = 0^\circ$ or 90° , by symmetry there is only the parallel harmonic component. For partially aligned molecules with other angles, the perpendicular component, which is usually much smaller, would appear, and the high harmonics would be elliptically polarized in general [51]. Generalization of Eq. (2.62) to the perpendicular component is straightforward. I focus on the parallel component in this thesis only.

Note that Eqs. (2.44), (2.55) and (2.62) are solved using a Crank-Nicholson routine for each value of ω . Typical parameters used in the calculations are 200 ~ 400 grid points along the radial direction r and 400 grid points along the longitudinal direction z for a 1-mm wide gas jet.

In the calculation, induced dipole moments included in the nonlinear polarizations of Eqs. (2.53) and (2.57) are mostly obtained by the QRS theory in this thesis. For an atomic target, one can use Eq. (2.33) to calculate the single-atom induced dipole in the frequency domain, and then transfer it back to the time domain by a Fourier transform. For a linear molecular target, it can only be partially aligned if it is placed in a short laser field (or a pump laser). The intensity of aligning laser is usually weak and not tightly focused such that it can be assumed to be a constant within the gas medium. In other words, the degree of molecular alignment is not varied in the medium at one fixed pump-probe time delay. The averaged induced dipole from partially aligned molecules at each point in the gas medium is then obtained by coherently averaging the induced dipole moment of a fixed-in-space molecule in Eq. (2.36) over a molecular angular distribution, i.e.,

$$D^{\parallel, \text{avg}}(\omega, \alpha) = \int_0^{2\pi} \int_0^\pi D^{\parallel}(\omega, \theta') \rho(\theta', \phi', \alpha) \sin \theta' d\theta' d\phi'. \quad (2.65)$$

Equation (2.65) is only for one particular molecular orbital.¹⁰

The total laser induced dipole over a number of active electrons is written as [52, 53]

$$D^{\parallel, \text{tot}}(\omega, \alpha) = \sum_{j,n} D_{j,n}^{\parallel, \text{avg}}(\omega, \alpha), \quad (2.66)$$

where index j refers to the different molecular orbital, and n is an index to account for the degeneracy in each molecular orbital. In the end, Eq. (2.66) is transferred into the time domain, and put back into Eq. (2.57).

2.4 Far-Field Harmonic Emission

Once high harmonics are emitted at the exit plane of an atomic or molecular (probably aligned) gas medium (called near-field harmonics), as shown in Fig. 2.1, they could propagate further in the vacuum until they are detected by the spectrometer. In this process, high harmonics may go through a slit, an iris or a pinhole, or be reflected by a mirror or more complicated optical system before they reach the detector (called far-field harmonics). In an axial-symmetric optical system, the complex electric field on the initial plane (near field) is related to the final plane (far field) by an $ABCD$ ray matrix, and $AD-BC=1$ for a lossless system. Here I only consider the simplest configuration shown in Fig. 2.1 without any additional optics (or within the free space propagation) between near and far fields, and $A=1$, $B=1$, $C=0$ and $D=1$ in the $ABCD$ matrix. According to the diffraction theory in the paraxial approximation, far-field harmonics can be obtained by using near-field harmonics through a Hankel

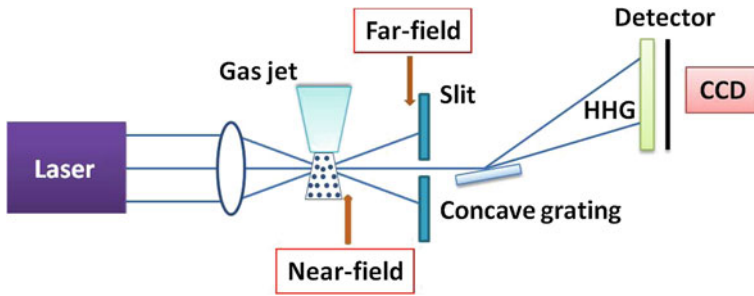


Fig. 2.1 Typical configuration for measuring the HHG in the far field. Adapted from [54]. © (2011) by IOP Publishing. Reproduced by permission of IOP Publishing. All rights reserved

¹⁰ In [49, 50], the integral over ϕ' in Eqs. (2.60) and (2.65) is incorporated into $\rho(\theta', \alpha)$.

transformation [44, 55, 56]^{11, 12}

$$E_h^f(r_f, z_f, \omega) = -ik \int \frac{\tilde{E}_h(r, z', \omega)}{z_f - z'} J_0\left(\frac{krr_f}{z_f - z'}\right) \exp\left[\frac{ik(r^2 + r_f^2)}{2(z_f - z')}\right] r dr, \quad (2.67)$$

where J_0 is the zero-order Bessel function, z_f is the far-field position from the laser focus, r_f is the transverse coordinate in the far field, and the wave vector k is given by $k = \omega/c$. For a molecular target, $\tilde{E}_h(r, z', \omega)$ is replaced by the electric field in Eq. (2.62), which also depends on the pump-probe angle α , and is parallel or perpendicular to the polarization of harmonic generating laser.

Assume that the high harmonics in the far field are collected from an extended area. By integrating the harmonic yield over this area, the power spectrum of macroscopic harmonics is obtained:

$$S_h(\omega) \propto \int \int |E_h^f(x_f, y_f, z_f, \omega)|^2 dx_f dy_f, \quad (2.68)$$

where x_f and y_f are Cartesian coordinates on the plane perpendicular to the propagation direction, and $r_f = \sqrt{x_f^2 + y_f^2}$.

Note that in Eq. (2.68), the detailed information on the experimental setup is involved. To simulate experimental HHG spectra quantitatively, besides general used laser parameters, such as intensity, duration, wavelength, spot size, and so on, one needs more parameters about the experiment, for example, the size and location of a slit.

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¹¹ $\tilde{E}_h(r, z', \omega)$ is expressed in the frequency domain, and its phase is also involved. Due to the different convention of Fourier transformation, this phase may need to change its sign before entering into the Eq. (2.67).

¹² Note that the elements of an $ABCD$ matrix are not expressed explicitly in Eq. (2.67), but the explicit expression can be found in Appendix D.2.

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