

Chapter 1

Introduction

Ultrafast laser technology makes it possible to generate extremely high field intensities above 10^{18} W/cm², or alternatively, to generate pulses with extremely short time durations corresponding to only a few femtoseconds (10^{-15} s).[8, 14] One of the most prominent applications of very high-power, ultrashort-pulse lasers has been the study of non-perturbative light-matter interactions. At intensities above 10^{14} W/cm², the magnitude of the electric field of the laser radiation is comparable to the field binding an electron to an atom. In this regime, the laser field can easily ionize atoms. Once the atom is ionized, the subsequent motion of the free electron is controlled by the oscillating laser field, and the electron can easily reach kinetic energies many times that of the original electron binding energy. One significant consequence of this intense light-matter interaction is the process of high-order harmonic generation (HHG).[51, 28] In HHG, the free electron recollides with its parent ion and recombines with it, releasing a high energy photon. Since all atoms experience the same coherent laser field, the harmonic radiation is also highly coherent. Temporally, high harmonic radiation is a series of attosecond-duration bursts occurring every half-cycle of the driving laser. This unique property has allowed measurements of electronic processes in atoms occurring on timescales previously inaccessible.[40, 24] The emission from HHG can extend from the ultraviolet up to the soft x-ray region of the spectrum (to ~ 950 eV). This spectral region is interesting for studies of chemical and material processes because these photon energies can access valence and inner shell electrons of atoms and molecules.[11, 33, 55] Also, HHG is an extremely useful light source for applications such as

plasma interferometry[23, 60] or extreme-ultraviolet (EUV) lithography, a technology that will be needed to implement the next generation of integrated circuits. Thus, high harmonic generation has both fundamental and practical applications. However, one major problem is that the efficiency of HHG is extremely low, typically on order of 10^{-5} to 10^{-10} conversion of laser energy into each harmonic order. As with any harmonic generation process, the efficiency can be greatly enhanced by phase matching. Traditional optical phase matching techniques have been applied successfully to high harmonic generation for low harmonic orders (< 90 eV).[62, 26] However, before now there were no techniques for enhancement of HHG at higher photon energies.

In this thesis, I will present two major breakthroughs in the field of high harmonic generation. First, I will discuss work on quasi-phase matching of high harmonic generation, which has allowed increased conversion efficiency of high harmonic light up to the water window region of the soft X-ray spectrum (~ 300 eV) for the first time.[31] This spectral region is significant because at these photon energies, water is transparent while carbon strongly absorbs, making it a useful light source for very high resolution contrast microscopy on biological samples. Since the resolution is on order of the wavelength of the light (~ 4 nm for 300 eV), detailed structures of cells and DNA can be viewed. A table-top source of light in the water window soft X-ray region would greatly benefit biological and medical research. Second, I will present work on the generation of very high harmonic orders from ions. This work is the first to show that harmonic emission from ions is of comparable efficiency to emission from neutral atoms thereby showing that high harmonic emission is not limited by the saturation intensity, or the intensity at which the medium is fully ionized, but can extend to much higher photon energies.[30] Both results were obtained by using a waveguide geometry for HHG, allowing manipulation of the phase matching conditions and reducing the detrimental effects of ionization. The ideas from this work are expected to increase the number of applications of high harmonic generation as a light source by increasing the efficiency of the process and opening up the possibility of generating multi-keV photon energies.

1.1 High-order Harmonic Generation

High-order harmonic generation occurs when an intense linearly polarized laser field interacts with a gas or material. An intuitive model of HHG at the atomic level was developed by Corkum, Kulander, and others,[22, 44] and is sometimes referred to as the three-step model. In the first step, the strong electric field of the laser suppresses the Coulomb barrier binding an electron to an atom, freeing the valence electron either by tunnelling or by over-the-barrier ionization. The freed electron is then accelerated by the field. Since the laser field is oscillating, the electron can with some probability return to its parent ion and recombine, emitting a high-energy photon. This process occurs for many atoms driven coherently over several laser cycles, resulting in emission of higher-order odd harmonics of the fundamental in a coherent, low-divergence beam. The three-step model accurately predicts that the highest photon energy that can result from HHG occurs when an electron is ionized at a phase corresponding to 18 degrees after the peak of the laser cycle. This cutoff photon energy is predicted to be:

$$E_{max} = I_p + 3.2U_p, \quad (1.1)$$

where I_p is the ionization potential of the atom and U_p is the ponderomotive energy given by:

$$U_p = e^2 E^2 / 4m\omega^2 = 9.33 \times 10^{-14} I \lambda^2 eV, \quad (1.2)$$

where e , E , m , ω , I , and λ are the electron charge, field amplitude, electron mass, fundamental laser frequency, intensity in W/cm², and wavelength in microns respectively. The highest photon energy therefore scales linearly with the laser intensity.

The recollision energies associated with the intensities required to field-ionize atoms can be as high as hundreds of electron volts.[15, 70, 67] Thus, the emitted high-harmonic photons can correspond to the combined energy of several hundred incident photons. The linear relationship between the cutoff and the incident intensity presents a very attractive scaling as compared to, for example, EUV laser schemes where the power requirements scale as the photon energy to the 3rd-5th power.[39] However, to take advantage of this favorable scaling, several

challenges must be overcome. The HHG process necessarily ionizes the medium, generating a free-electron plasma. The dispersion of the plasma creates a mismatch in the phase velocities of the fundamental and harmonic light, significantly reducing the amount of harmonic signal produced. Also, the generated plasma can defocus the laser beam, decreasing the peak intensity and thereby limiting the maximum harmonic energy.[48] To date, the highest energy HHG emission has been achieved using short-duration laser pulses and noble gases with high ionization potentials such as helium, which can reduce the amount of ionization for a given peak intensity. However, helium has a small cross-section giving it a lower efficiency for HHG than larger, multi-electron atoms. In Ch. 3, I will discuss a method of generating high harmonics even in a fully ionized gas medium, allowing HHG in argon up to photon energies of 250 eV, previously achieved only with He gas, and demonstrating that HHG from ions is feasible. In combination with quasi-phase matching techniques, this method should extend the efficient energy range of HHG even further.

The simple semi-classical picture of HHG as given by the three-step model accurately predicts the range of photon energies that can be obtained for a given incident laser intensity. However, a quantum mechanical treatment is needed for a more complete picture of the HHG process. In the quantum picture, as the laser field strength increases, portions of the electron wave function tunnel through the suppressed atomic barrier and then propagate large distances - relative to the atomic radius - away from the atom. When the laser field reverses, this extended electron wavefunction re-encounters the part of the wavefunction still in the ground state of the atom, leading to quantum interferences. As a result, rapid oscillations in the electronic probability distribution and therefore the transient dipole moment of the atom lead to the emission of very high-order harmonics of the fundamental laser.

A fully quantum, analytical theory of HHG, derived by Lewenstein et al.[46], has been extremely successful in describing both the general characteristics of HHG such as the photon energy cutoff, as well as more specific characteristics such as the divergence properties of the generated beam[9] and the specific spectral characteristics of the emission. Qualitatively, the

spectral characteristics of HHG emission are a result of the fact that the freed electron has an associated deBroglie wavelength corresponding to the kinetic energy acquired in the laser field, $\lambda = h/p$, where h is Planck's constant and p is the electron momentum. The emission also has a phase related to the total phase the electron accumulates during its free trajectory, given by:[9]

$$\Phi = q\omega t_f - S(p_{st}, t_i, t_f)/\hbar, \quad (1.3)$$

where q is the harmonic order, ω is the laser frequency, t_i is the time the electron is released in the laser field, t_f is the recollision time, and $S(p_{st}, t_i, t_f)$ is the semi-classical action over the electron's trajectory. The phase of the emitted harmonic light is therefore not simply related to the phase of the driving laser, but also includes an intrinsic phase component that can vary rapidly with laser intensity. The intrinsic phase has consequences for both the spatial and spectral emission characteristics. Spatially, the intrinsic phase can result in a complex spatial profile of the harmonic emission when generated using a beam that is converging toward a focus. This is in contrast to the gaussian spatial profile for HHG generated by a beam diverging from a focus[9, 63, 58] or the full spatial coherence of light generated using a waveguide configuration.[49] In the spectral domain, this time-varying intrinsic phase results in large frequency shifts and spectral broadening or narrowing of the harmonic emission.[16] The intrinsic phase also plays a role in quasi-phase matching of HHG as will be discussed in the next chapter.