Summary

This thesis (entitled, *Table-top tunable narrow-band extreme-ultraviolet sources: from low to high-order optical harmonic generation*) is devoted to the development and application of table top sources in the vacuum-ultraviolet (VUV, 200-100 nm) and extreme-ultraviolet (XUV, 100-10 nm) regions of the electromagnetic spectrum. Specifically, nonlinear up-conversion of laser radiation in a gaseous medium is used to generate narrow-band continuously tunable radiation. With such a source, on-line accurate frequency domain spectroscopy can be performed at high-resolution, revealing features in the level structures of atoms and molecules.

In Chapter 1 an introduction on the nonlinear physical process of optical harmonic generation in gases with intense laser pulses is given. The two different regimes of low and high-order harmonic generation are presented: low-order harmonic generation (LHG) with pulses of relatively moderate intensity ($I < 10^{13}$ W/cm$^2$) is described in term of perturbation theory; the process of high-order harmonic generation (HHG), occurring at higher intensity ($I > 10^{13}$ W/cm$^2$), is described in terms a of the three-step semiclassical re-collision model. The break-down of the perturbative approach and the on-set of substantial photoionization, due to both multiphoton absorption or tunneling, is also discussed. Specific table-top VUV/XUV sources based on of both low and high-order harmonic generation and examples of their applications are presented.

In Part I (Chapter 2 and 3) VUV spectroscopic measurements of xenon and krypton performed with a table top source based on LHG are presented. Precision metrology studies enable accurate determination of the excited state level energies. Specifically, the energy level structure of krypton is determined with an unprecedented absolute accuracy of about 40 MHz. New accurate determination of the ionization energies of both xenon and krypton is also reported. Mass resolved spectroscopy enables the measurement of the isotope shift and hyperfine structure of the excited states. For both xenon and krypton the well known King plot analysis reveal a strong screening effect of the outer $p$-electron on the $s$-electron in the closed shell configuration typical of noble gases. For xenon, hyperfine constants are determined for some excited states. For krypton, investigation on the ac-Stark shift on a specific two-photon transition gives useful insight for the absolute frequency calibration of XUV radiation produced by two-photon resonant-enhanced four-wave-mixing.

Part II concerned with the development and characterization of a novel XUV source based on HHG.

In Chapter 4 an original laser system delivering continuously tunable near-infrared pulses with 300 ps time duration, 225 mJ of energy and 10 Hz repetition rate is presented. The tunable pulses are generated in a pulsed-dye-amplifier pumped by the second harmonic of a Q-switched Nd:YAG laser and seeded by a continuous Ti:Sapphire laser. The 300 ps pump pulses are obtained via a compression technique based on the stimulated-Brillouin-scattering process. The near-infrared pulses are then amplified in a Ti:Sapphire amplifier chain. The energetic near infrared pulses have a good
beam quality ($M^2 = 1.2$) and are close to the Fourier transform limit (time-frequency band-width product = 0.48).

In Chapter 5 the production and characterization of tunable narrow-band high-order harmonics is reported. Focusing the energetic near infrared pulses with a 20 cm focal length lens an intensity of $5 \times 10^{13}$ W/cm$^2$ is achieved and coherent radiation down to 40 nm ($21^{st}$ harmonic order) is generated in noble gases. The frequency spectrum of the harmonics is determined by performing linear absorption on well calibrated narrow atomic lines. Specifically, the band-width of the $9^{th}$ and $15^{th}$ harmonic, at 86 nm and 52 nm respectively, is measured demonstrating a resolving power ($\lambda/\Delta \lambda$) of $2.8 \times 10^5$. From the prospectives of band-width and resolution these results are superior to what is achieved at the dedicated high-resolution VUV/XUV beamlines at synchrotrons worldwide, and bring HHG in the realm of high-resolution VUV/XUV spectroscopy.

In Chapter 6 an accurate investigation on frequency chirp in HHG is reported. In particular the frequency shift of the harmonics with respect to the expected value (integer multiple of the fundamental frequency) is measured at high-resolution as function of the gas density. It is found that the major contribution to frequency chirp in HHG is due to self-phase modulation (SPM) of the fundamental beam induced by temporal changes of the refractive index in the laser-produced plasma. This results in a blueshift of the harmonic frequency proportional to the gas density. Other effects, like SPM due to the Kerr-effect of neutral atoms, or the nonadiabatic effect related to the re-collision nature of HHG, give a negligible contribution when 300 ps pulses are used. Of particular interest is the observation of a density-independent redshift in the harmonic frequency. This phenomenon, never observed before in HHG, may be related to plasma dynamic effects or to a strong frequency chirp present in the fundamental pulses. Possible experiments to investigate the origin of this redshift are suggested.