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loachim Pupeza

Passive Optical Resonators for Next-Generation Attosecond Metrology



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To my family

Preface

The unparalleled control over broadband light fields afforded by laser architectures based on visible/near-infrared (VIS/NIR) phase-stabilized mode-locked oscillators has steadily spawned new means for expanding our understanding of basic processes in nature and the universe. On the one hand, the temporal confinement of light to bursts with durations reaching down to merely a few oscillations of the optical carrier wave, with exquisite repeatability and with field strengths rivaling those of the atomic Coulomb fields, has enabled real-time measurements (and control) of ultrafast processes with ever-improving temporal resolution. Nowadays, *attosecond metrology* grants experimental access to the fastest events outside the atomic core, namely transitions between quantum electronic states that determine the physical and chemical properties of atoms, molecules, and condensed matter and mediate chemical reactions or biological processes.

On the other hand, the temporal periodicity of the pulse train emitted by modelocked oscillators results in a comb-like spectrum, consisting of equidistant laser lines separated by the pulse repetition frequency. *Frequency combs* thus conveniently link the domains of optical frequencies (few-to-100s of THz) and of radio frequencies (~100 MHz) accessible to contemporary electronics. This enables the most precise measurements of time with optical clocks permitting, e.g., the search for miniscule drifts of natural constants.

The availability of radiation sources providing broadband coverage of the vacuum-/extreme-ultraviolet spectral ranges (henceforth, for simplicity referred to as XUV) with multi-MHz pulse repetition frequencies and coherence properties akin to those of mode-locked oscillators promises a substantial impact on precision frequency metrology, photoelectron spectroscopy (PES) and attosecond science. For instance, a particularly severe shortcoming of laser systems employed in attosecond-resolution PES arises from their relatively low pulse repetition frequencies, usually in the lower kHz range, in the context of space charge effects limiting the admissible number of photoelectrons ejected during each laser pulse from a solid. At typical nanosecond-range travel times of photoelectrons inside the detector, the detection duty cycle in state-of-the-art measurements amounts to a fraction of a percent, resulting in unpractically or even prohibitively long measurement times. In

addition, applications like precision spectroscopy of hydrogen-like ions for tests of bound-state quantum electrodynamics, or the realization of robust, multi-PHz optical clocks tuned to nuclear transitions have constituted another important impetus for the development of XUV frequency combs.

This book reviews the research activities conducted by the author and his team over the course of five years toward such a source and its experimental validation. Primarily motivated by the development of a new generation of high-repetition frequency instruments for multi-dimensional attosecond PES, the source, based on cavity-enhanced high-harmonic generation (HHG), operates at a repetition frequency of 18.4 MHz, emitting 5×10^5 photons per pulse in the 25–60 eV range. The emission is scalable to higher photon energies (> 100 eV) at the cost of the photon rate. This book summarizes and arranges the developments leading to this source, as well as proof-of-principle ultrafast attosecond PES measurements, published in 18 original publications, and is structured as follows.

As a first prerequisite for efficient intracavity HHG, power scaling of femtosecond enhancement cavities is thoroughly investigated. Advanced resonator designs are derived and verified, featuring large illuminated spots on all mirrors, mitigating both intensity and thermally induced enhancement limitations. With suitable mirror-substrate and dielectric-coating materials, at a repetition frequency of 250 MHz, record average powers for ultrafast laser technology of 670, 400, and 20 kW are demonstrated for 10-ps, 250-fs, and 30-fs pulses, respectively, circulating in a passive optical resonator.

Secondly, the dynamics of a high-finesse, passive resonator in the presence of a highly nonlinear optical process, such as HHG, are quantitatively investigated in theory and experiment. These investigations were instrumental in allowing for a holistic optimization of the XUV source reported here, which for the first time reached intracavity HHG conversion efficiencies similar to those achieved in single-pass setups with similar gas targets (~ 10^{-7} in an argon gas target).

Thirdly, we extensively studied geometric XUV output coupling from the enhancement cavity, where the high-photon energy beam exits the cavity through an opening in the mirror following the HHG focus. Among various output coupling methods, geometric output coupling exhibits the advantages of robustness, low distortion to the participating pulses, and photon-energy scalability. Several implementations are discussed in this book, using both the fundamental transverse Gaussian resonator mode and tailored, higher-order modes. The latter offers the prospect of broadband output coupling efficiencies approaching unity, as well as of novel spatio-temporal gating methods for the generation of isolated XUV attosecond pulses.

Finally, we report proof-of-principle attosecond angle-resolved PES experiments carried out at 18.4 MHz, with attosecond pulse trains emerging via HHG driven by sub-40-fs, near-infrared pulses circulating in a femtosecond enhancement cavity. In our experiment, 1×10^{10} photoelectrons were released per second from a 10-µm-diameter spot on tungsten, at space charge distortions of only a few tens of meV. Broadband, time-of-flight photoelectron detection with nearly 100% temporal duty cycle evidences a count rate improvement between two and three