Passive Optical Resonators for Next-Generation Attosecond Metrology

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The outstanding control of broadband, coherent light fields enabled by visible/infrared femtosecond modelocked laser technology, has steadily contributed to the advancement of our understanding of fundamental processes in nature. For instance, the temporal confinement of light to pulses with durations reaching down to merely a few oscillations of the optical carrier wave, with exquisite repeatability and with field strengths rivalling those of the atomic Coulomb fields, has enabled real-time measurements (and control) of ultrafast processes with ever-improving temporal resolution. Nowadays, *attosecond metrology* experimentally accesses 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.

Among the tools for attosecond metrology, photoelectron spectroscopy (PES) that harnesses high-order harmonic generation (HHG) provides particularly direct access to electron dynamics in matter. HHG in gases driven by visible/infrared pulses combines two features. Firstly, the emitted eV-to-keV photon energies grant access to an ultrabroad range of electronic transitions – including even those from tightly-bound core levels – via single-photon absorption. Secondly, this radiation emerges in ultrashort, typically sub-femtosecond bursts that are rigidly locked to the oscillations of the electric field driving HHG, permitting attosecond-precision timing in experiments employing both fields. However, space-charge-induced measurement distortions limit the number of ejected photoelectrons affordable per pulse. At nanosecond travel times of the photoelectrons in the detector, for state-of-the-art kHz-repetition-rate laser systems employed in multi-dimensional attosecond-PES, the detection duty cycle amounts to a fraction of a percent, implying unpractical (or even prohibitive) measurement times of several days.

In *passive optical resonators* (enhancement cavities), femtosecond pulses can be coherently stacked, enabling efficient HHG at repetition rates of tens of MHz. These radiation sources uniquely combine broadband vacuum- and extreme-ultraviolet spectral coverage with high pulse repetition rates and coherence properties akin to those of modelocked lasers. In the talk, a review of the research conducted by the author and his team over the course of five years will be presented, focused on the development of a laser system for attosecond-PES at multi-MHz pulse repetition rates. Cavity-enhanced HHG results in 5×10^5 photons emitted per pulse in the 25-to-60-eV range, with the emission being scalable to photon energies > 100 eV, at a pulse repetition rate of 18.4 MHz. Broadband, time-of-flight photoelectron detection with a nearly 100% temporal duty cycle provides a count rate improvement between two and three orders of magnitude over state-of-the-art attosecond-PES experiments under identical space charge conditions and, thus, a reduction of the measurement time from days to minutes.

This research will be presented in the broader context of controlling coherent light waves in passive optical resonators, with applications including the generation of high-power temporal dissipative solitons, the prospect of generating isolated attosecond pulses by intracavity spatiotemporal tailoring of light fields, and that of generating ultrabroadband frequency combs.