

Generation of Coherent sub-20 nm XUV Radiation at 78 MHz via Cavity-Based HHG

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Abstract. We present two major advances of enhancement-cavity-based high-order harmonic generation (HHG). First, the generated extreme ultraviolet (XUV) radiation is coupled out collinearly through an on-axis opening in the mirror following the HHG focus. This minimizes the interaction of both the fundamental and the intracavity generated radiation with the output coupler while simultaneously enabling a large enhancement and an output coupling efficiency that increases with the harmonic order. Second, we use the nonlinearly compressed pulses of an Yb-based laser to drive intracavity HHG allowing for a unique power regime combining short pulses with high average powers. Together, these advances overcome fundamental limitations of current enhancement cavity setups and extend intracavity HHG towards higher photon energies. In a proof-of-principle experiment we use a 3-kW and 78-MHz train of 54-fs to generate and couple out coherent sub-20 nm radiation.

1 Introduction

Over the past two decades, extreme ultraviolet (XUV) optical pulses in the spectral region 1 nm - 100 nm obtained via laser-driven high-order harmonic generation (HHG) in a gas have emerged as a key experimental tool enabling the study and control of time-resolved electron motion on the atomic and molecular scale [1,2]. In ultrashort-pulse laser systems the high peak intensities (exceeding 10^{13} W/cm²) necessary to drive HHG are usually obtained at the cost of the pulse repetition rate, which typically lies well below 1 MHz. For many pump-probe and spectroscopy techniques this constitutes a major shortcoming. For instance, the scheme proposed in [3] combining XUV photoelectron emission microscopy with attosecond streaking spectroscopy promises “filming” the collective dynamics of electrons induced by an optical pulse in metal nanostructures with nanometer-scale spatial resolution and subfemtosecond temporal resolution. However, space charge severely limits the usable intensity of the XUV probe, implying the necessity of a large number of probe pulses with a moderate intensity [4]. Another example is precision spectroscopy of atomic and molecular transitions with frequency combs. As the comb line spacing in the frequency domain equals the pulse repetition rate, resolving individual comb lines requires a large repetition rate. So far, frequency combs generated via HHG reaching down to 40 nm have been demonstrated [5]. For these applications and many others,

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the availability of a table-top coherent XUV radiation source combining high photon energies with high repetition rates and short pulse durations is highly desirable.

The technique of coherently overlapping the (amplified) ultrashort pulses of a mode-locked laser at the original repetition rate in a passive enhancement cavity (EC) housing the HHG process seems to be one of the most promising approaches to reach this goal. Femtosecond ECs, seeded by Ti:Sa-based lasers were first demonstrated in 2005 [6,7]. More recently, advances in Yb-based fiber laser technology enabled reaching higher circulating powers in ECs [5,8], resulting in higher photon energies and higher XUV average powers. However, due to the narrower gain bandwidth of Yb-doped active materials, these systems operate with pulse durations significantly longer than 100 fs. Moreover, so far in all HHG-ECs the spatial separation of the generated harmonic radiation from the fundamental beam and, thus, coupling out of the XUV relies on reflection or diffraction, precluding a performance scaling of these concepts towards significantly higher photon energies. In this contribution we present two major advances in the femtosecond EC technology. First, to couple out the harmonic radiation directly and collinearly we use a mirror with a laser-machined small opening aligned with the resonator optical axis. Second, we seed our cavity with the pulses of an Yb-based laser, spectrally broadened in a nonlinear fiber and subsequently compressed using chirped mirrors. This is facilitated by the fact that our output coupler (OC) introduces neither additional dispersion nor nonlinearities. For the first time, sub-100 fs pulses are enhanced to several kW of average power and the generation of sub-20-nm coherent radiation with the Gaussian fundamental transverse mode of a resonator is demonstrated. This concept is simultaneously scalable towards higher generated photon energy and photon flux and towards shorter pulses.

2 Experimental Setup and Results

The experimental setup (Fig.1a) is an extension of the one described in [8]. The seeding Yb fiber-based CPA laser delivers 200-fs pulses centered at $1.04 \mu\text{m}$ with a repetition rate of 78 MHz and an average output power of up to 50 W. The pulses can be nonlinearly compressed down to 50 fs using a piece of 3-cm long $40\text{-}\mu\text{m}$ core PCF and several bounces on a pair of chirped mirrors with 75% power throughput. The laser is locked to a passive ring resonator placed in vacuum using the Pound-Drever-Hall technique. The mirror following the cavity focus has an on-axis opening, manufactured by inverse laser drilling [9] into a standard substrate before the coating process. The hole has a $\sim 40\text{-}\mu\text{m}$ inner and a $\sim 70\text{-}\mu\text{m}$ outer radius, which determine the output coupling efficiency for the XUV and the losses for the fundamental radiation, respectively. As the divergence of the harmonic beam decreases with increasing order, the XUV output coupling efficiency increases with the photon energy (Fig. 1c).

To achieve a high power enhancement (> 150) with the on-axis hole in place, we use an asymmetric focusing geometry (ROC=100 mm, 200 mm) leading to a beam radius of 2 mm on the output coupler (OC) and choose a nearly impedance-matched input coupler (R=99.5%). Up to 3 kW of circulating power and stable operation in this regime are obtained. On timescales on the order of several seconds, we find no significant difference in the lock stability compared to the enhancement of uncompressed pulses. Gas is introduced at the focus through a tapered glass nozzle with a $100\text{-}\mu\text{m}$ -diameter opening. The output coupled XUV light is steered by mirrors, anti-reflection coated for the fundamental radiation, to a grazing incidence spectrometer and a CCD camera. Injecting xenon with 0.5 bar backing pressure limits the intracavity power to 1.9 kW allowing for the generation of harmonics up to the 37th order. At this power level, ionization leads to a spectral blue-shift of the intracavity spectrum resulting in a decrease of the spectral overlap with the incident spectrum. Increasing the gas pressure typically increases the harmonic yield in the plateau at the cost of peak intensity, due to increasing plasma-induced distortions (spatially as well as spectrally) [5]. With argon, higher average powers can be reached in the cavity (2.2 kW). The achievable peak intensities on the order of 10^{14} W/cm^2 and the higher ionization potential of Ar lead to a boost of the harmonic order (53th) to record levels for multi-MHz HHG (Fig.1b). Saturation of the intracavity power is again observed due to nonlinear interaction of the laser pulse with the plasma. With neon, even higher intensities are reached, leading to even higher harmonics (59th, 17.6 nm).

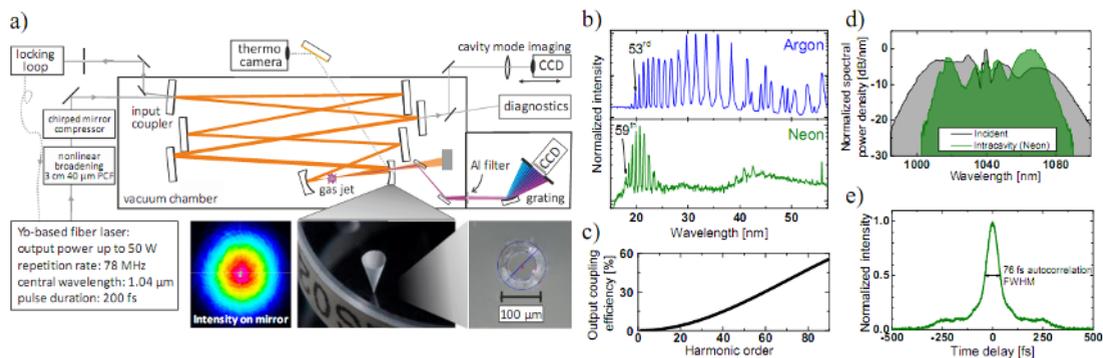


Fig. 1. a) Experimental setup. The curved mirror following the cavity focus has an on-axis opening through which the XUV radiation is coupled out of the cavity. b) HHG spectra obtained with argon and neon behind a 300-nm thin Al filter. c) Calculation of the XUV output coupling efficiency using the strong field model described in [10] for the given geometry. d) Spectra of the nonlinearly broadened fundamental field (incident and intracavity). e) Intracavity autocorrelation trace (76 fs FWHM) during HHG operation with neon at 2.8 kW of average power.

3 Conclusions and Outlook

We have demonstrated experimentally two improvements in EC-based HHG. First, XUV output coupling through an on-axis opening in the mirror following the focus minimizes the interaction of both the fundamental and the generated radiation with the OC while simultaneously allowing for a large enhancement and output coupling efficiency. Second, we enhance nonlinearly compressed pulses in a cavity for the first time, enabling a reduction of the circulating pulse duration in combination with high average powers typical for Yb-based sources. In a proof-of-concept, we generate (and couple out) sub-20-nm coherent radiation for the first time at multi-10-MHz repetition rates.

The main significance of our experiment is, however, the demonstration of the unprecedented versatility of this EC concept. The hole-OC allows for the first time efficient photon-energy-scalable collinear XUV output coupling without introducing additional dispersion, nonlinearities or polarization discrimination for the fundamental radiation. This opens the door to a new power regime for EC-based HHG and, in particular, will allow the investigation and optimization of ionization-related effects, which are expected to limit further scaling [5], over a large parameter range. Moreover, due to the lack of intracavity polarization discrimination, this concept promises to be suitable for the implementation of a polarization-gating technique [2] for the generation of isolated attosecond pulses with multi-10-MHz repetition rates.

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