

Efficient output coupling of intracavity high-harmonic generation

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We demonstrate a novel technique for coupling extreme-ultraviolet (XUV) harmonic radiation out of a femtosecond enhancement cavity. We use a small-period diffraction grating etched directly into the surface of a dielectric mirror. For the fundamental light, this element acts as a high reflector. For harmonic wavelengths, it acts as a diffraction grating, coupling XUV radiation out of the cavity. Using this method, we observed the third through twenty-first odd harmonics with a dramatic increase in usable power over previous results of high-harmonic generation at high repetition rates. © 2008 Optical Society of America

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Recent developments in optical frequency combs have revolutionized optical frequency metrology [1,2]. However, these techniques have in general been limited to the visible and near IR spectral regions. Meanwhile, progress in short-wavelength light sources has been rapid, achieving unprecedented temporal resolution, spectral coverage, and brightness [3]. A powerful technique for producing extreme-ultraviolet (XUV) wavelengths is high-harmonic generation (HHG), which utilizes extreme nonlinear optical processes in atoms and molecules facilitated by amplified femtosecond pulses [4–6]. Traditional methods for generating pulses of sufficient energy for HHG employ low-repetition-rate amplifiers, leaving no comb structure in the spectrum of the harmonic radiation. Therefore, the spectral resolution of these short-wavelength sources is poor when compared with precision visible sources.

Experiments in which broadband, femtosecond pulses are coupled into passive external cavities are very promising in this respect, as the pulse energy is sufficiently enhanced to enable the HHG process without a decrease in the pulse repetition rate [7]. Already, there have been two successful efforts in intracavity HHG with explicit demonstrations of phase coherence of the third-harmonic light, resulting in high expectations that this method can push frequency comb techniques into the XUV spectral region [8,9]. Especially intriguing is the prospect of applying the recently established technique of direct frequency comb spectroscopy [10] in the XUV spectral region [11,12].

One of the outstanding technical challenges to generating XUV radiation via an enhancement cavity has been the lack of a suitable method to couple the harmonic light out of the cavity. Inside the enhancement cavity, the harmonics are generated collinearly with the fundamental light. Since there is essentially no solid material that is sufficiently transparent at XUV wavelengths, the XUV radiation cannot pass through the cavity mirrors without being absorbed. A requirement for any output coupling method is that its implementation should not significantly increase

the cavity loss, nor should it introduce significant nonlinearity that prevents efficient couplings between a train of ultrashort pulses and the cavity. Thus far, proposed XUV output coupling methods have been less than desirable in this regard. Published results have shown output coupling of harmonic radiation using a thin sapphire plate at Brewster's angle [8,9]. Since the index of the sapphire is less than unity at the harmonic wavelengths, one can achieve a large Fresnel reflection of the harmonic radiation while introducing only a small round-trip loss to the fundamental pulses. This method has a fundamental limitation, however, in that $\chi^{(3)}$ nonlinear processes within the Brewster's plate introduce prohibitively large dispersion at high intensities, severely limiting the possibility of power scaling such a system [13].

Another proposed method for coupling harmonics out of the enhancement cavity is to drill a small hole in the curved mirror after the intracavity focus [14]. Since the harmonic light will diverge less than the fundamental light, most of the harmonic light will pass through the hole, while most of the fundamental light will be reflected. While this method allows for power scaling, the small hole invariably introduces additional loss to the cavity. This intracavity loss can be somewhat mitigated by coupling a higher-order transverse mode (e.g., TEM_{01}) into the cavity at the cost of losses outside the cavity owing to mode conversion. Even so, in practice we have found it difficult to keep intracavity losses small using this method. Also, the harmonic radiation generated with a TEM_{01} fundamental beam will display a complicated transverse mode profile that is not ideal for subsequent experiments. A similar proposed technique uses a slotted mirror and two colliding pulses to produce noncollinear HHG [14,15]. This method also suffers from additional cavity loss for the fundamental light, and the implementation is very challenging technically.

In this Letter, we demonstrate a novel method for coupling harmonics out of an enhancement cavity that is far superior to the Brewster's plate method in

terms of power scalability. It also introduces substantially smaller losses than coupling the harmonic radiation out of the cavity through a small hole in a curved mirror. We add an optic directly after the intracavity focus. The element acts as a diffraction grating for all harmonic orders and yet is a near-perfect high reflector for the 1070 nm fundamental light. We find this optic produces no measurable decrease in the cavity finesse and power enhancement. It also seems to have a minimal effect on the cavity dispersion as determined from the measured transmitted spectrum of the fundamental light. To manufacture this crucial element, we began with a dielectric mirror coating, engineered for low dispersion and high reflectivity at 1070 nm and a 70° angle of incidence with *s* polarization. In the top coating layer we had a 420 nm period diffraction grating etched into the fused silica surface. In Fig. 1, we depict the cross section of this optic. The period is small enough so that the fundamental 1070 nm light has only a zeroth diffracted order. While this introduces birefringence in the top layer of the coating [16], the operation of the mirror at the fundamental wavelength is left largely unaffected. The high harmonics have higher diffracted orders, with the largest power being diffracted into the negative first order. The generation and shaping of attosecond pulses could be achieved by the use of additional gratings to recombine the harmonic orders.

The operation of this optic at XUV wavelengths relies on a simple Fresnel reflection from the surface of the grating structure, as there is strong absorption in the bulk of the material. Even so, Fresnel reflections from a vacuum/SiO₂ interface can be very large at XUV wavelengths with a grazing angle of incidence. In fact, at the ninth through nineteenth harmonics, the Fresnel reflection is $\sim 45\%$ with *s* polarization at a 70° angle of incidence. It might be interesting to increase this angle further to help improve the output coupling efficiency. However, cavity geometries under this circumstance will be more difficult to implement.

To estimate the output coupling efficiency of the intracavity grating, we used an approximation of the rigorous integral method described in [17] with the index and absorption values taken from [18]. We maximized the efficiency for the ninth through nineteenth harmonics by adjusting the period, duty cycle, and feature height of the grating with the requirement that the fundamental light diffracts only into

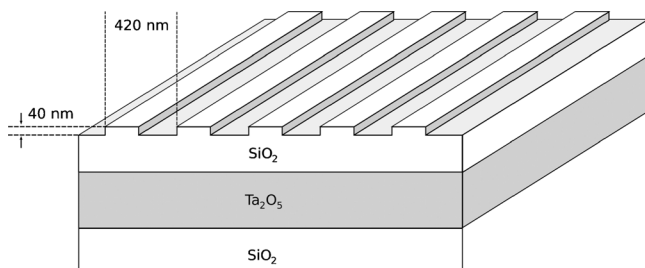


Fig. 1. Cross section of the grating used to couple harmonic radiation out of the enhancement cavity. The grating is etched directly into the surface of a dielectric coating designed for high reflectivity and low dispersion at 1070 nm. The reflectivity of the dielectric mirror is 99.988%.

the zeroth order. The final grating design had a period of 420 nm, a duty cycle of 40%, and a step height of 40 nm, as shown in Fig. 1. The calculated output coupling efficiencies are shown in Table 1. Owing to limitations in the manufacturing capability, the grating structure we used had a 47% duty cycle, which affected the performance minimally. The grating output coupling efficiencies are very competitive with the other proposed and demonstrated methods without the drawbacks mentioned previously. To increase the efficiency for a specific harmonic order using this method, one could attempt to etch an appropriate blazed structure into the surface of a high reflector.

To experimentally evaluate the performance of this output coupling method, we build on previous work where we resonantly enhanced light from an amplified mode-locked Yb-fiber laser in a passive high-finesse cavity [19]. The reconfigured cavity incorporating the grating is shown in Fig. 2. Before enhancement, the laser produces 10 W of power with ~ 75 fs pulses at a 136 MHz repetition rate. To coherently add the femtosecond pulses inside the cavity, we control both independent degrees of freedom of the frequency comb, the repetition rate f_{rep} , and the offset frequency f_{ceo} [7]. On resonance, we achieve an enhancement of 260 to obtain 2.6 kW of intracavity power and an intracavity pulse duration of ~ 100 fs. The focus in our cavity is achieved with two 10 cm radius of curvature mirrors that produce a calculated focused spot area of $960 \mu\text{m}^2$ and hence a peak intensity of $4 \times 10^{13} \text{ W/cm}^2$. We inject xenon gas at the intracavity focus using a glass nozzle with an aperture of $100 \mu\text{m}$ and a backing pressure of 700–1500 Torr. The intensity is large enough to produce high-harmonic radiation in the xenon gas, which is subsequently coupled out of the cavity by the grating. The power of the harmonics is measured with a calibrated photodiode [20] and also observed visually with a fluorescent plate, as shown in Fig. 1.

We were able to simultaneously observe all odd-harmonic orders up to the twenty-first through fluorescence on the coated glass plate. Photodiodes sensitive at XUV wavelengths with directly deposited

Table 1. Theoretical Efficiency of the Grating at Harmonic Wavelengths of the Incident 1070 nm Light and the Respective Outcoupled Powers Measured in the Experiment

Harmonic Order	$\lambda(\text{nm})$	Coupling Efficiency	Outcoupled Power (nW)
3	356.7	0.7%	-
5	214.0	1.9%	-
7	152.9	3.5%	-
9	118.9	9.6%	-
11	97.3	8.5%	-
13	82.3	9.0%	250
15	71.3	10.3%	20
17	62.9	9.6%	54
19	56.3	8.4%	38
21	51.0	6.7%	-

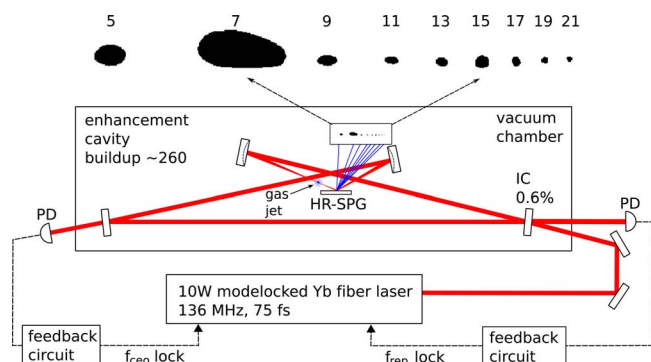


Fig. 2. (Color online) Experimental setup. IC, input coupler; PD, photodiode; HR-SPG, high reflector with small period grating on surface. The top panel shows an enlarged image of the experimentally observed high-harmonic orders fluorescing on a glass plate coated with sodium salicylate. The threshold of the image was reduced around the twenty-first harmonic to show its mode structure.

filters were used to block background fundamental light by many orders of magnitude, allowing measurement of the harmonic power. Unfortunately, we had no metallic filters available that efficiently transmit wavelengths longer than ~ 100 nm. This severely limited our ability to measure the power in the harmonics below the thirteenth. We were able to measure the power in the thirteenth harmonic with an XUV-sensitive diode coated with 200 nm of In and 20 nm of MgF_2 . The power in the fifteenth through nineteenth harmonics was measured using a photodiode with a 150 nm aluminum coating. The results of these power measurements are shown in Table 1. The cutoff of our harmonic radiation was observed to lie between the nineteenth and the twenty-first harmonics. The power level in the thirteenth harmonic shows an intracavity conversion efficiency of $\sim 10^{-9}$ (corresponding to $\sim 3 \times 10^{-7}$ for the unenhanced power). The observed conversion efficiency, cutoff wavelength, and relative power levels of the harmonic orders fit well with theoretical calculations at our intensity levels [21].

To our knowledge, this is the first demonstration of the seventeenth through twenty-first harmonic orders via intracavity HHG. It is notable to compare the measured power in the seventeenth harmonic at 63 nm with previous results at 61 nm (the thirteenth harmonic of 795 nm fundamental light), which displays an increase in the output coupled power by nearly 4 orders of magnitude [9]. We attribute this dramatic increase in usable harmonic power to the ability of using a laser with higher peak powers for the intracavity experiment without being plagued by nonlinear dispersions introduced by a Brewster's plate.

To conclude, we have demonstrated a novel method for output coupling harmonics from a femtosecond

enhancement cavity, and in the process we have shown record power levels and to our knowledge the highest harmonic orders ever produced at multi-megahertz repetition rates. The small-period intracavity grating has overcome the major difficulties of coupling high-harmonic radiation out of an optical buildup cavity. It adds minimal intracavity loss, permitting a large buildup peak power without introducing nonlinear phase shifts to the cavity resonance. The absence of nonlinearities within the enhancement cavity removes one of the major barriers to power scaling such a system.

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References

1. T. Udem, R. Holzwarth, and T. W. Hänsch, *Nature* **416**, 233 (2002).
2. S. T. Cundiff and J. Ye, *Rev. Mod. Phys.* **75**, 325 (2003).
3. T. Brabec and F. Krausz, *Rev. Mod. Phys.* **72**, 545 (2000).
4. P. B. Corkum, *Phys. Rev. Lett.* **71**, 1994 (1993).
5. M. Lewenstein, P. Balcou, M. Y. Ivanov, A. Lhuillier, and P. B. Corkum, *Phys. Rev. A* **49**, 2117 (1994).
6. C. Spielmann, N. H. Burnett, S. Sartania, R. Koppitsch, M. Schnurer, C. Kan, M. Lenzner, P. Wobrauschek, and F. Krausz, *Science* **278**, 661 (1997).
7. R. J. Jones and J. Ye, *Opt. Lett.* **29**, 2812 (2004).
8. R. J. Jones, K. D. Moll, M. J. Thorpe, and J. Ye, *Phys. Rev. Lett.* **94**, 193201 (2005).
9. C. Gohle, T. Udem, M. Hermann, J. Rauschenberger, R. Holzwarth, H. A. Schuessler, F. Krausz, and T. W. Hänsch, *Nature* **436**, 234 (2005).
10. A. Marian, M. C. Stowe, J. R. Lawall, D. Felinto, and J. Ye, *Science* **306**, 2063 (2004).
11. M. Bellini, S. Cavalieri, C. Corsi, and M. Matarazzi, *Opt. Lett.* **26**, 1010 (2001).
12. S. Witte, R. Zinkstok, W. Ubachs, W. Hogervorst, and K. Eikema, *Science* **307**, 400 (2005).
13. K. D. Moll, R. J. Jones, and J. Ye, *Opt. Express* **13**, 1672 (2005).
14. K. D. Moll, R. J. Jones, and J. Ye, *Opt. Express* **14**, 8189 (2006).
15. J. Wu and H. Zeng, *Opt. Lett.* **32**, 3315 (2007).
16. C. W. Haggans, L. Li, and R. K. Kostuk, *J. Opt. Soc. Am. A* **10**, 2217 (1993).
17. L. I. Goray, *Nucl. Instrum. Methods Phys. Res. A* **536**, 211 (2005).
18. E. D. Palik, *Handbook of Optical Constants of Solids* (Academic, 1998).
19. I. Hartl, T. R. Schibli, A. Marcinkevicius, D. C. Yost, D. D. Hudson, M. E. Fermann, and J. Ye, *Opt. Lett.* **32**, 2870 (2007).
20. <http://www.ird-inc.com/axuvabs.html>.
21. A. L'Huillier, P. Balcou, S. Candel, K. J. Schafer, and K. C. Kulander, *Phys. Rev. A* **46**, 2778 (1992).