XUV SOURCES

13-nm XUV pulses from a cavity

Cavity-enhanced high-harmonic generation has been extended to the ~10 nm wavelength range by using a pierced cavity mirror for outcoupling. This light source has the potential to realize further advances in precision extreme-ultraviolet spectroscopy and attosecond physics.

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Lasers have been steadily improving towards shorter pulses, shorter wavelengths and higher powers. The development of high-harmonic generation (HHG) resulted in a sudden leap from femtosecond, visible pulses to attosecond, extreme-ultraviolet (XUV) pulses, which has led to many discoveries regarding the behaviour of atoms and molecules at the attosecond timescale[1]. However, attosecond pulses can currently be generated only at low repetition rates, presenting problems in terms of the signal acquisition speed and precision of measurements with attosecond resolution.

Now, writing in *Nature Photonics*, Ioachim Pupeza and colleagues describe a new laser configuration that has the potential to change this situation[2]. By combining nonlinearly compressed pulses from a high-power femtosecond fibre laser with cavity-enhanced HHG, they generated XUV light with wavelengths down to 11.5 nm for the first time. They further demonstrated a new technique for cavity outcoupling that efficiently extracts such high-energy XUV pulses from the cavity.

Pupeza *et al.* found that the problem of plasma-induced intracavity phase distortions was reduced when they used relatively short (51 fs) pulses from an ytterbium fibre amplifier rather than longer pulses, which can cause undesired ionization for significant durations before and after the pulse peak. They generated such short pulses by overcoming the bandwidth limitations of fibre amplifiers through employing external pulse compression. This allowed them to achieve intracavity pulsewidths as short as 57 fs at average incident laser powers of tens of watts. Using this configuration, the researchers demonstrated an intracavity peak focused intensity of 8×10^{13} W cm^{-2}. By using improved pulse compression techniques, ytterbium fibre lasers delivering 25 fs pulses with an average power of 250 W have been demonstrated[3]; such sources should allow further intracavity intensity scaling with low-dispersion cavity mirrors.

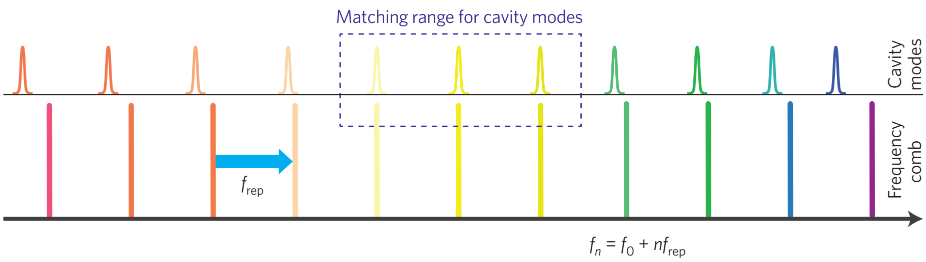

Figure 1 | A frequency comb in relation to a typical resonance spectrum of an enhancement cavity. The frequency of the comb spectrum, f_n, follows the relationship $f_n = f_0 + nf_{rep}$, where n is an integer, f_0 is the frequency of the input pulse and f_{rep} is the repetition rate of the mode-locked pulse train; the ideal comb spacing is thus determined by f_{rep}. Cavity dispersion causes the cavity comb to overlap with the frequency comb over only a limited spectral matching range.

HHG is a three-step process: (1) an intense (~10^{13} W cm^{-2}) electric field ionizes atoms in a gas jet; (2) the freed electrons are accelerated by the laser field; and (3) the varying phase of the laser field accelerates the electrons away from and then back to the parent ion, where they can recombine to the ground state and any excess energy is re-emitted as high-harmonic radiation. This process is most effective with few-cycle femtosecond pulses such as highly amplified 800 nm pulses from a Ti:sapphire laser operating at a repetition rate of about 1 kHz.

The high laser intensities required to initiate this process together with the very low efficiency of HHG has so far limited the application of this concept to the scientific realm. The use of cavity enhancement techniques has been proposed to increase the HHG efficiency. These techniques were used in the early days of nonlinear optics to improve the efficiency of second-harmonic generation with continuous-wave lasers[4]. Frequency comb technology can be utilized to adapt cavity enhancement to short optical pulses. When the individual comb teeth of a mode-locked pulse train are matched to the resonances of an enhancement cavity, subsequent pulses can add up constructively inside the cavity, leading to a large enhancement of the intracavity peak power[5,6]. One limitation is residual dispersion in the cavity, which prevents perfect matching of the comb teeth and hence restricts cavity resonance to a limited spectral range (Fig. 1). However, with present mirror technology, cavity enhancement can be demonstrated for pulses with widths of a few tens of femtoseconds. Even using cavity enhancement techniques, reaching the peak power levels required for ionization at repetition rates in the 100 MHz range requires high average pump powers of the order of tens of watts.

To enhance the convenience and power of HHG, researchers have been considering the most convenient and powerful laser sources — fibre lasers. Advances in fibre technology over the past few years have enabled fibre frequency comb lasers to be constructed that have average powers of 100 W (ref. 7). However, the potential of these high-power sources in HHG could not be fully realized because of plasma-related peak-power clamping in the cavity and the limited bandwidth of fibre amplifiers, which prevents amplification of pulses shorter than ~100 fs. Peak-power clamping occurs because HHG involves an ionization step, which inevitably means that some ions will be formed. Plasma formation in an optical cavity is problematic, but plasma formation inside the pulse is worse as it contributes intensity-dependent frequency shifts that are very difficult to control. Pupeza *et al.*

found that intracavity phase distortions can be alleviated by using shorter pulses. Thus, shorter pulses are better and allow higher intracavity peak intensities to be achieved.

Pupeza *et al.* also implemented a new solution to the long-standing problem of outcoupling from the enhancement cavity. This problem arises because substrate absorption prevents extraction of harmonics through the cavity mirrors. The harmonics must therefore be extracted from the cavity by an alternative means that maintains cavity resonance. The simplest method is to add a thin window that is oriented at the Brewster angle for the circulating light but not for the harmonics. About 10% of the harmonics can be outcoupled this way. Another method uses a diffraction grating for XUV wavelengths imprinted on one of the cavity mirrors. This avoids the need to add an extra element to the sensitive cavity, but the outcoupling efficiency is still only about 20% (ref. 8).

Pupeza *et al.* demonstrated a third way — outcoupling by drilling a tiny hole in one of the cavity mirrors. This operational mode is achievable because the divergence scales inversely with wavelength so that the divergence of the XUV radiation was much smaller than that of the pump light. The researchers showed that with a moderate hole diameter of 80 μm they could still obtain a cavity enhancement of 250 and that the output coupling efficiency could reach 90% for a wavelength of about 10 nm (the output coupling efficiency increases with decreasing wavelength); this is illustrated in Fig. 2. The achieved intracavity intensities were similar to those obtained with other fibre laser systems[8], but HHG extended to much shorter wavelengths. The researchers demonstrated a useful output down to the 91st harmonic (wavelength ~11.5 nm), which is roughly three times the harmonic order obtained with previous fibre systems.

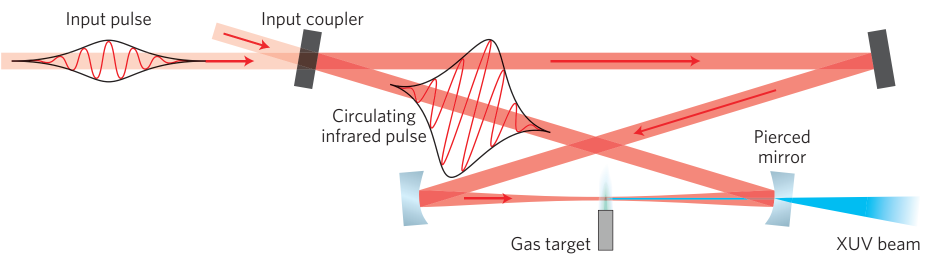

Figure 2 | Schematic showing the output coupling of high-harmonic light through a small aperture in one of the cavity mirrors. The pump light is only weakly perturbed by the central hole and is reflected with high efficiency, whereas the collinear low-divergence high-harmonic light escapes through the hole.

In the wavelength range 28–50 nm, average output powers in the range 1–10 μW per harmonic were obtained.

The overall performance of the system is still very far from the tens or hundreds of watts of incoherent XUV used in photolithography for fabricating integrated circuits, but the present results represent a significant step towards the development of a tabletop coherent XUV light source for photoelectron spectroscopy and imaging. XUV imaging will become particularly attractive if the emission wavelength can be extended to the water window near 4 nm, which now seems quite possible. Synchrotrons also produce coherent XUV light, but at a much greater cost and without the frequency comb precision of a cavity system[8]. Indeed, the coherence and comb structure of the present technique are key advantages for many other applications. For example, further advances in XUV spectroscopy can be envisaged, potentially enabling nonlinear helium spectroscopy and tests of many-body bound-state quantum electrodynamics. These new sources will also assist the search for variability in the fine-structure constant. The present results are getting close to point where the generation of isolated attosecond pulses at the repetition rate of a mode-locked laser becomes feasible, which would introduce a significant array of new capabilities to attosecond physics. Cavity-enhanced HHG pumped by simple fibre lasers holds great promise to evolve into the long-sought-after convenient source of coherent XUV light. ❐

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References

1. Corkum, P. B. & Krausz, F. *Nature Phys.* **3,** 381–387 (2007).
2. Pupeza, I. *et al. Nature Photon.* doi:10.1038/nphoton.2013.156 (7 July 2013).
3. Jocher, C., Eidam, T., Hädrich, S., Limpert, J. & Tünnermann, A. *Proc. SPIE* **8601,** 86011F (2013).
4. Ashkin, A., Boyd, G. D. & Dziedzic, J. *IEEE J. Quantum Electron.* **2,** 109–124 (1966).
5. Gohle, C. *et al. Nature* **436,** 234–237 (2005).
6. Jones, R. J., Moll, K. D., Thorpe, M. J. & Ye, J. *Phys. Rev. Lett.* **94,** 193201 (2005).
7. Ruehl, A., Marcinkevicius, A., Fermann, M. E. & Hartl, I. *Opt. Lett.* **35,** 3015–3017 (2010).
8. Cingöz, A. *et al. Nature* **482,** 68–71 (2012).

Competing financial interests

Both authors are employed by IMRA America, Inc.

Published online: 7 July 2013