Towards coherent X-ray free-electron lasers

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Increased bandwidth and fluctuations are hurdles on the path towards generating intense fully coherent X-ray free-electron laser output. A recent experiment at FERMI Trieste demonstrated that these difficulties can be overcome by an approach called echo-enabled harmonic generation.

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The advent of a fully coherent X-ray free-electron laser (FEL) may have broad applications in many areas such as biology, chemistry and materials science. Coherent output from an FEL using an approach called echo-enabled harmonic generation (EEHG) has so far been demonstrated only at relatively long wavelengths. In particular, to achieve high-intensity output from an X-ray FEL, after the harmonic generation, the short-wavelength radiation must be amplified by many orders of magnitude, a process called exponential growth. So far, EEHG with exponential growth[1] has been demonstrated in the 300–400 nm region.

Now, writing in *Nature Photonics*, Primož Rebernik Ribič and colleagues have demonstrated coherent emission followed by, what is essential, exponential growth in the soft X-ray region using an EEHG configuration at FERMI, Trieste[2]. Owing to the much shorter wavelengths the recent experiment at FERMI is an important milestone in the development of a fully coherent soft X-ray FEL.

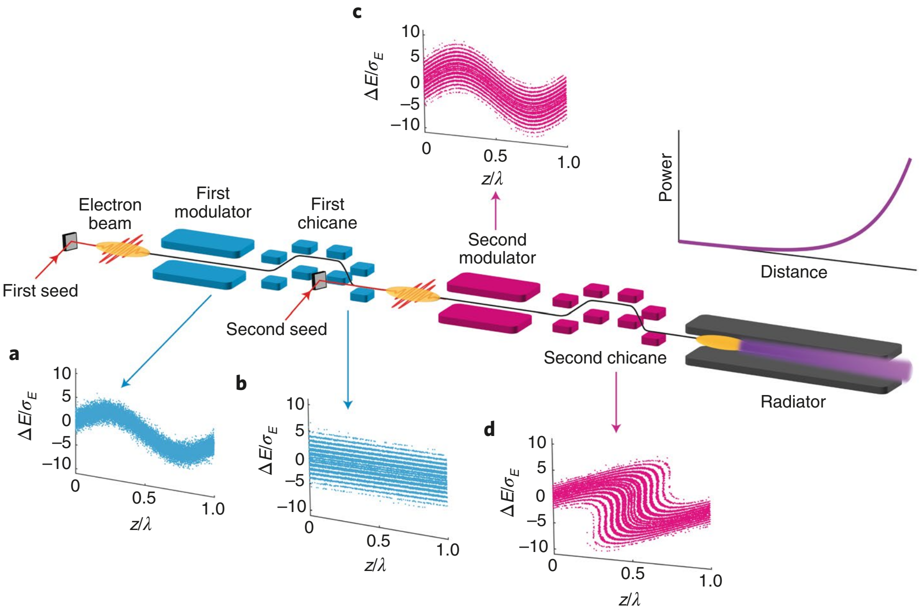

Fig. 1 | Illustration of EEHG followed by exponential growth. a–d, The evolution of the electron energy distribution ΔE expressed in terms of its ratio over the energy spread σ_E along the system length z, expressed in the units of laser wavelength λ. Panels **a–d** refer to the first modulator, the first chicane, the second modulator and the second chicane, respectively. The power–distance graph (right) shows that the radiation is exponentially amplified as it travels through the radiator. Middle: experiment set-up. Figure adapted from ref. [2], Springer Nature Limited.

One of the main features of the FERMI EEHG experiment, as compared with previous FEL studies, is that the output is less sensitive to the initial bunch imperfections even at such a short wavelength as 5.9 nm, the exponentially amplified 45th harmonic of the seed laser at 264 nm. This result is important because the sensitivity is closely related to the coherence and stability of the output laser pulses and may have deep impact on the future development of fully coherent X-ray FELs.

One of the most important characteristics of any laser is its temporal coherence, that is, the phases of the head and the tail of the laser pulse are correlated, resulting in a very narrow bandwidth determined by the Fourier transform limit. This is essential for a wide range of applications in laser physics, for example in spectroscopy and holography. However, conventional lasers cannot reach into the soft X-ray regime.

The advent of high-gain X-ray FELs extended the wavelength down to the hard X-ray region[3–5]. In an FEL, an electron bunch passes through a device with an alternating magnetic field called an undulator. The electrons in the bunch follow a sinusoidal trajectory and emit spontaneous radiation. The individual electrons are randomly distributed. Hence, the radiation starts from random noise. When a certain set of conditions on the electron-beam quality is satisfied, the radiation from an electron can catch up with some of the electrons in front of it and be amplified. As a result, collectively, the spontaneous radiation from the whole bunch can be amplified exponentially. This mechanism is known as self-amplified spontaneous emission (SASE), which lacks temporal coherence because it starts from noise.

For an X-ray FEL the lack of coherence is a substantial drawback. It is obvious that a method is needed to establish phase correlation between the head and the tail of the bunch. Many techniques have been developed to create this correlation at the start of the amplification process. One of them is self-seeding[6], realized by sending the SASE output from an undulator through a monochromator to select the part of the radiation within a narrow bandwidth as a seed to be amplified in another undulator. The random nature of the SASE output induces large fluctuations in the seed, leading to both large intensity and bandwidth fluctuations intrinsic to the SASE FEL process.

Another approach, proposed to enable fully coherent X-ray FEL output, is called high-gain harmonic generation (HGHG)[7]. In this method, an external laser is used to generate an energy modulation in the electron beam in an undulator (or 'modulator'). Then, the energy modulation is converted to a density modulation in a chicane. The density modulation is called microbunching because the electrons are grouped with the period equal to the laser wavelength. The microbunching can be arranged so that the beam is bunched into peaks much narrower than the laser wavelength so that there is rich harmonic content in the beam. This highly microbunched beam is sent into another undulator (or 'radiator'), which is resonant to higher harmonics of the seed wavelength to generate radiation of much shorter wavelength, to be exponentially amplified until saturation is reached. Clearly, in this process, the phase correlation between the head and the tail is provided by the external laser.

The highest harmonic number generated is determined by the ratio of the energy modulation over the intrinsic energy spread. On the other hand, in a high-gain FEL there is a parameter called the FEL parameter that limits the allowed magnitude of the energy modulation. If the energy spread due to this modulation is larger than the FEL parameter, the exponential growth of intensity will be largely reduced, and the saturation power drops rapidly. Thus, the maximum harmonic number is limited by this ratio, which constitutes the main principle in the design of an HGHG experiment. As pointed out by Ribič and colleagues, one of the HGHG experiments, also carried out at FERMI, has shown the typical limit for the highest harmonic number at which the exponential growth can be maintained is about $h = 15$. This leads to the search for different methods to generate smaller intrinsic energy spread in order to achieve higher harmonic number, thus reaching shorter FEL wavelengths.

One of these methods is EEHG[8]. As shown in Fig. 1, there are two modulators and two chicanes in the EEHG set-up. In the first modulator, the electron-beam energy is modulated, as shown in Fig. 1a, within one laser wavelength (a period). In the first chicane, electrons with higher energy go through a path shorter than that of the electrons with lower energy, so they advance further to the right in the upper part of Fig. 1b, while the electrons with lower energy fall behind and move to the far left in the lower part of Fig. 1b. With sufficiently large path-length difference, after passing through the first chicane, the phase space shows that the beam is sheared into many very thin stripes; each stripe has a distinct energy different from the others. The important point is that each of these stripes behaves like a beamlet with very small energy spread (very thin slice). In comparison, the initial energy spread in Fig. 1a is much larger (thicker). In the second modulator and second chicane, the process is then very similar to HGHG, the beam is modulated in energy again (Fig. 1c) and when it passes through the second chicane, the energy modulation is converted to microbunching (Fig. 1d). However, the phase space now has much higher harmonic content than conventional HGHG, mainly because the existence of the many very thin stripes (Fig. 1b). As shown in the EEHG experiment by Ribič and colleagues, starting from a seed at 264 nm, the harmonic number reached $h = 45$ at 5.9 nm with exponential growth, and at the end of the last radiator section showing some indication of saturation. Also, coherent emission at 2.6 nm reached the harmonic number $h = 101$.

Another method to overcome the limit set by the ratio of the FEL parameter over the intrinsic energy spread, in the HGHG process, is called cascaded HGHG. This method has also been demonstrated experimentally at FERMI[9] by arranging two stages of HGHG in succession, where the first HGHG stage generated the seed for the second stage. The second stage used a 'fresh bunch', that is, another part of the bunch, which was not affected by the FEL interaction in the first stage. In this part of the bunch, the energy spread is not increased by the energy modulation generated in the first stage, hence the exponential growth is continued into the second stage. The final harmonic number is the product of the harmonic numbers of both stages, thus this FEL can reach the very-short-wavelength range.

Cascaded HGHG is sensitive to the imperfection of the energy distribution in the electron bunch. In the modulator these imperfections are converted into fluctuations of the central wavelength and intensity. However, as demonstrated by the work of Ribič and colleagues at FERMI, the very large shearing process in the first chicane erases the noise generated by the imperfections in the electron bunch. As a result, the EEHG process is less sensitive to the beam quality and generates very stable output both in intensity and central wavelength. As mentioned earlier, this is one of the distinct features of the experiment at FERMI[2], in spite of the very high harmonic number and short wavelength.

Ribič and colleagues also provide a comparison between EEHG and two-stage HGHG, showing that EEHG provides both narrower and cleaner spectra with significantly less shot-to-shot jitter in the central wavelength, and also point out that in the regime studied two-stage HGHG outperforms EEHG in terms of energy per pulse. Thus, this indicates the possibility of a cascade employing both EEHG and HGHG methods, using the advantages inherent to both schemes.

In conclusion, the two main obstacles towards a fully coherent X-ray FEL are the difficulty of reaching high harmonic numbers and noise due to the limited quality of the electron beam. But now, Ribič and colleagues provide convincing evidence that overcoming these difficulties is possible. Consequently, this opens the door to a fully coherent soft X-ray FEL, or even a fully coherent hard X-ray FEL, as suggested in a recent proposal[10] of a cascaded EEHG scheme using two bunches instead of the 'fresh' part of one bunch, or a combination of EEHG and conventional HGHG. Thus, the work of Ribič and colleagues perhaps has already given us a glimpse into a very bright future of hard X-ray holography of protein molecules. This would be our dream. ❐

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