COMMENT

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The development of XFELs

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X-ray free-electron lasers (XFELs) have rapidly developed into unique tools for probing diverse systems of interest to different scientific disciplines with angstrom–femtosecond resolution. Claudio Pellegrini provides an overview of the milestones in the development of XFELs and their unique capabilities.

The first ruby laser was built 60 years ago by Theodore Maiman[1]. Since then, developing an X-ray laser has been a major goal in laser physics and in science. A laser that generates high-intensity, coherent X-ray pulses at angstrom wavelengths and femtosecond pulse durations — the characteristic length and timescales for atomic and molecular phenomena — allows imaging of periodic and non-periodic systems as well as non-crystalline states, and studies of dynamic processes in systems far from equilibrium, nonlinear interactions and X-ray quantum optics. Such techniques therefore open a window into electronic, atomic and molecular phenomena of interest to biology, chemistry, materials science and physics.

The origin of XFELs

The atom-based, population inversion, traditional approach to lasing becomes challenging when pushing towards X-ray wavelengths. Excited atomic-core quantum energy levels have a short lifetime, about one femtosecond times the square of the wavelength in angstroms[2]. The short lifetime and large energy needed to excite inner-shell atomic levels, 1–10 keV compared with ~1 eV for visible lasers, require an extremely high pumping power to attain population inversion, making an X-ray laser in the 10 keV region impractical as a scientific tool.

A new paradigm was needed to solve the problem: the use of radiation from relativistic electron beams moving through a periodic magnetic array, an undulator magnet. The radiation from relativistic electrons propagating in an undulator, a free-electron laser (FEL), was used by John Madey in the 1970s to build an amplifier and an oscillator in the infrared region[3]. The extension of the system to short wavelengths in a single-pass configuration starting from noise was made possible by the high-gain, wavelength-invariant, collective instability theory developed in the 1980s[4] and verified experimentally in the late 1990s[5]. This combination led to my initial proposal in 1992 to build an X-ray FEL (XFEL) at SLAC[6], now called LCLS, operating in the nanometre to angstrom spectral region. The proposal and work done at SLAC stimulated the construction of soft X-ray FELs that operate in the wavelength region of a few nanometres or longer at DESY and, later, Trieste. LCLS started to operate successfully in 2009 and was followed by the construction of other hard XFELs in Japan (SACLA), Korea, Switzerland and Germany (European XFEL), as well as a soft XFEL in China[7].

Hard XFELs, near-angstrom wavelengths, are large and expensive instruments, kilometre-scale in size and billion-dollar-scale in cost. Soft XFELs are cheaper to build and operate, but still cost a few hundred million dollars and are hundreds of metres in length. The construction and operation of such expensive machines can only be justified by the unique possibilities they offer for the exploration of matter at the atomic and molecular levels and by offering access to a large number of users. LCLS successfully demonstrated the unique experiments that can be performed at XFELs, providing insight into complex biological, chemical and physical processes[8]. From the start, LCLS received a large number of experimental proposals, many more than could be accommodated.

XFELs today

Today, typical single X-ray pulse characteristics in the angstrom region include a peak power of 10–50 GW, a pulse duration of a few to hundreds of femtoseconds, a relative linewidth of $\sim 10^{-3}$, good transverse coherence, limited longitudinal coherence and a pulse repetition rate of 50–120 Hz. The European XFEL in Hamburg is driven by a superconducting linac, which delivers up to 27,000 pulses per second, delivered in 10 separate bunches, at wavelengths from nanometres to below 1 Å. A new project based on a superconducting linac at SLAC, LCLS-II, is under construction and will be completed by 2022. It will cover the soft and hard X-ray region and will operate at pulse repetition rates of up to 1 MHz (REF.[7]).

An important characteristic of XFELs is flexibility. The capability to continuously tune the wavelength, vary the pulse length and adjust the intensity, beam size, polarization and bandwidth ensures that the X-ray pulse can be optimized for any given experiment, making XFELs a highly desirable source. As the gain medium is an electron bunch propagating in an undulator magnet, it is possible to change the X-ray pulse characteristics

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by manipulating the electron bunch, something that the accelerator physics community has been doing successfully over the past 10 years using lasers and the electromagnetic field generated by the bunch itself[9]. Bunch manipulation has been used by several groups to generate two X-ray pulses of different colours and variable time separation, from a few to tens of femtoseconds, from the same bunch. Two-colour imaging and pump–probe experiments have taken advantage of this capability. The manipulation of the electron bunch with lasers and magnetic chicanes has also been used to generate X-ray pulses that are a few hundred attoseconds long, initially in the soft X-ray region, but now being extended into the hard X-ray region to explore electron dynamics and other ultrafast phenomena. The longitudinal coherence has been improved and the linewidth reduced by an order of magnitude through seeding, by which the lasing process is started with an external laser instead of noise, at soft X-ray wavelengths. Similar results have been obtained at hard X-ray wavelengths with self-seeding, in which the undulator magnet is split into two parts and the signal generated by the first part is filtered with a monochromator to seed that in the second.

The results obtained with XFELs are numerous and include chemical dynamics (molecular movies) on the femtosecond and attosecond timescales, protein imaging, single-particle imaging and studies in high-energy-density physics and materials science. One striking example is the emerging atomic-level understanding of water splitting and oxygen evolution in the protein complex photosystem II[10]. Using a combination of synchrotron radiation sources and XFELs in Japan and the USA, researchers observed various steps of this light-driven, oxygen-generating, catalytic cycle at room temperature. Very recently, LCLS and SACLA were used to untangle the sequence of events in the formation of the oxygen-evolving complex for the final step of water splitting, including a key step in which a water molecule acts to bridge manganese and calcium atoms. Crystallography and X-ray spectroscopy were simultaneously employed to probe the atomic structure and reveal information about the chemical processes during the reaction. The ultimate goal is to create an atomic movie using many snapshots made throughout the entire cycle, including the elusive moments at which two oxygen atoms bond to form an oxygen molecule. These studies are providing fundamental understanding of how nature uses sunlight to create molecular oxygen and will guide our approaches to artificial photosynthesis and its application.

The future of XFELs

Despite the demonstrated success, there remains considerable scope for further technological progress. Future developments for improving the capabilities of hard XFELs include increasing the peak power to the terawatt level using superconducting undulators; using optical cavities based on diamond crystals to improve the efficiency, raise the peak and average power, and reduce the linewidth with a recirculated amplifier; and a FEL oscillator based on a superconducting linac to provide higher order coherence and very narrow linewidths.

It is remarkable that several experiments have demonstrated that XFELs, because of their very high peak power in a limited bandwidth at the right wavelength, can generate population inversion and high gain in a classical atom-based approach to an X-ray laser. Population inversion-based lasing has been demonstrated in amplified spontaneous emission mode or seeded mode in Cu, Mn and He. These results, combined with progress in crystal-based optical cavities, might lead to an X-ray laser oscillator and applications in areas such as X-ray quantum optics and interferometry.

In the short time that they have been operating, XFELs have already proved to be an important instrument for the study of the properties of matter in diverse applications. They are complex instruments that are pushing science into previously inaccessible regions of observation. Until very recently, the limited access to the few available instruments has severely restricted our ability to learn how to make the best use of these remarkable machines. This situation is now changing, as new XFELs are coming online and providing access for a larger number of scientists. Together with further improvements in X-ray optics, detectors, data storage and analysis, this increase in capacity will certainly lead to new experimental techniques and ideas, making it possible to fully exploit the potential of XFELs for discovery.

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Competing interests

The authors declare no competing interests.