at low supersaturation because there are many step sites available for attachment. Crystal growth of 100 μm over 20 days was obtained at the exposed face, yielding an overall increase of 30% in the size of the crystal. The laser-treated crystals were analysed at the SPring-8 synchrotron facility in Harima, Japan. The quality of the resulting crystals was as high as would be expected from crystals grown using conventional methods optimized through laborious trials.

In contrast to crystal growth, several previous studies have explored the use of laser light to nucleate crystals from solution or melt. The mechanisms proposed for nucleation include putative alignment of solute species by the optical electric field[4] (optical Kerr effect), or formation of intermediate bubbles in the nucleation process[5]. Aggregation of solute and nucleation can be stimulated near surfaces by a form of optical trapping[6]. Photochemical reactions may also stimulate nucleation, of liquids from vapours[7] as well as of crystals from liquids[8], although damage to the solute is generally an unwanted side-effect. Lasers have also been used to fragment the surfaces of crystals, at much higher pulse energies compared with those used in the current work, causing daughter crystallites to be formed[9].

The work of Tominaga and colleagues establishes an exciting new direction for optical control of crystallization, enabling the use of laser light to both initiate nucleation and stimulate crystal growth. The spiral mechanism of growth is common to many crystalline materials, and therefore the method should be widely applicable. By stimulating a specific crystal face, we open up the potential for optical engineering of crystals, whereby morphology and therefore local properties may be dictated by site-specific growth. At present, the method relies on obtaining a viable crystal to work with, and the laser pulse energies must be just above the threshold for ablation, which requires testing. However, by offering unprecedented temporal and spatial control, the technique promises unique opportunities for studying mechanisms of crystal growth, for example, using laser pump–probe methods. ❐

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X-RAY LASERS

Multicolour emission

The X-ray free-electron laser at the SLAC National Accelerator Laboratory in the US can now generate multicolour X-ray pulses with unprecedented brightness using the fresh-slice technique. The development opens the way to new forms of spectroscopy.

Chao Feng and Haixiao Deng

Many forms of time-resolved spectroscopy, such as ultrafast pump–probe techniques, or nonlinear optics experiments, such as four-wave mixing, require a sample to interact with multiple ultrashort pulses of coherent radiation of different wavelengths (colours). While this is routinely possible with long-wavelength photon sources in the infrared, accomplishing it in the extreme-ultraviolet or X-ray region has been beyond the capabilities of most short-wavelength light sources based on high-harmonic generation or synchrotron radiation. Now, writing in *Nature Photonics*, Lutman *et al.* illustrate an experiment[1] at the Linac Coherent Light Source (LCLS) at Stanford, USA that delivers multicolour X-ray pulses with unprecedented brightness, via a technique called fresh-slice.

The LCLS is a free-electron laser (FEL) — a coherent source of electromagnetic radiation that utilizes accelerated relativistic electrons as a gain medium. The advent of high-intensity and ultrashort pulse X-ray FELs around the globe has opened the door for scientists to observe and control ultrafast phenomena that occur on the atomic timescale of femtoseconds. The unequivocal success of X-ray FELs is now being followed by efforts to give the sources expanded capabilities, such as shorter pulse duration and spectral control. The innovation of Lutman *et al.* is to use a carefully tailored electron beam to obtain two-colour FEL pulses with saturated powers of tens of gigawatts, durations of a few to tens of femtoseconds and a variable time delay of up to the picosecond level.

An FEL generates coherent radiation by passing relativistic electron bunches through a spatially alternating magnetic field, usually created by a periodic magnetic structure called an undulator. The lasing wavelength of an FEL is determined by the period and magnetic field of the undulator, and the kinetic energy of the electrons.

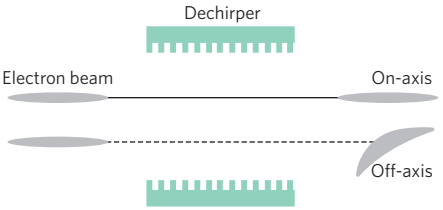

Figure 1 | Electron-beam manipulation with a dechirper. When the electron beam travels on-axis, the transverse distribution will not change. When the electron beam travels off-axis, it will experience a strong transverse head-to-tail kick that results in a transverse tilt.

In the past few years, several techniques for multicolour operation and spectral control of X-ray FELs have been developed worldwide. The key to achieving multicolour pulse generation can be easily understood from the basic principles of FEL operation. For example, the first two-colour

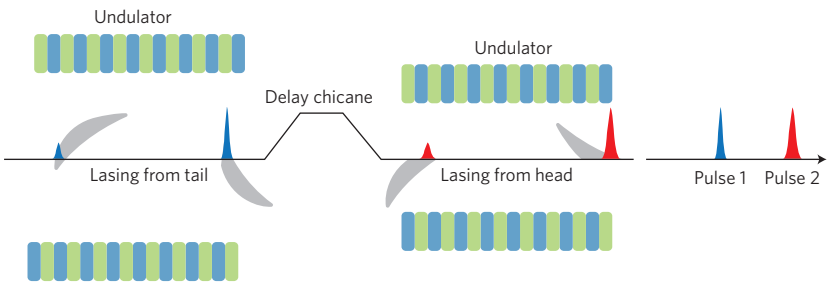

Figure 2 | Schematic layout for two-colour X-ray generation with the fresh-slice technique. The undulator beamline consists of two undulator sections with different magnetic field and a delay chicane. The designed electron bunch orbits are different in the two undulator sections to choose different parts of the electron bunch to lase. The central wavelength of each pulse can be independently tuned by changing the magnetic field of the undulator, and the temporal separation between two pulses can be continually tuned by changing the strength of the delay chicane.

experiment at the LCLS employed an ultrashort electron beam successively passing through two undulator sections with different magnetic fields[2]. The central wavelengths of the two FEL pulses could be independently tuned by simply changing the gaps of the two undulators. While this method is very simple and has already been successfully demonstrated at the LCLS and the SACLA facility in Japan[3], it does have some limitations. The most important issue is that the power of the radiation leaving the first undulator needs to be far from saturation, otherwise the electron beam will be deeply disturbed and unable to lase again in the second undulator. To obtain two saturated FEL pulses of different colours, researchers have tried to make two ultrashort electron bunches with different kinetic energies to lase in a single long undulator[4]. However, this set-up prevents the independent and flexible tuning due to the strong correlation between the kinetic energy and longitudinal position of the two electron bunches, and has the drawback of a limited range for spectral control.

The fresh-slice technique proposed and demonstrated by Lutman *et al.* provides a new approach to multicolour pulse generation. It uses a pair of planar aluminium corrugated structures, called a dechirper[5], and installed upstream of the undulator, as shown in Fig. 1, to manipulate the time and transverse position of the electron bunch.

It is interesting to note that the dechirper at the LCLS was originally designed to narrow the bandwidth of the X-ray FEL pulses by controlling the time–energy correlations of the electron bunch[6], as demonstrated at the SDUV-FEL test facility in China[7].

In this latest experiment, the dechirper is used to introduce a time-correlated transverse kick on an electron bunch with an off-axis propagation path. After a suitable drift, this transverse kick results in a transverse tilt of the electron bunch. The lasing undulator consists of two undulator sections designed to resonate at different X-ray wavelengths. A magnetic chicane to delay the electron bunch with respect to the light is also included as shown in Fig. 2 to control and tune the time gap between the different coloured pulses as desired.

The idea is that the electron beam is carefully steered so that the tail of the bunch will travel straight, on-axis in the first undulator section and generate the first saturated FEL pulse, while the head of the bunch experiences an oscillating trajectory that results in a totally suppressed FEL gain and thus no lasing. Then, in the second undulator section, the transverse positions of the bunch head and tail are exchanged and the gain from the bunch head increases and generates another FEL pulse but at a different wavelength.

Employing different slices of the electron beam to lase in different undulator sections gives the fresh-slice technique several benefits. In particular, this set-up avoids the destruction of the whole electron beam in each undulator section and thus holds the potential to fully amplify X-ray pulses with different colours separately. Based on the present layout of the LCLS, researchers have performed experiments for both two-colour and three-colour pulse generation. The two-colour experiments were carried out at photon energies around 707 eV. Two FEL pulses with pulse durations of approximately 5 fs (from tail) and 17 fs (from head), pulse energies of around 248 μJ (from tail) and 484 μJ (from head) and colour separation corresponding to a photon energy difference of about 15 eV have been achieved.

The time delay between these two radiation pulses can be easily tuned from 0 ps to approximately 1 ps by using the delay chicane. Potentially one can even control the polarization of the X-ray pulses with the inclusion of an additional variable polarization undulator and thus enable polarization-related pump–probe experiments. The fresh-slice technique was also successfully employed to generate triple-colour X-ray pulses around 780 eV photon energy, which has not been achieved before.

The fresh-slice technique looks set to take multicolour X-ray FEL generation to a new era. X-ray pulses with high peak power, flexible delay, large colour separation and variable polarization are now routinely delivered for user experiments at the LCLS. Such light source significantly improves the temporal resolution and flexibility of X-ray pump–probe experiments and can even be used to perform a broad variety of four-wave-mixing spectroscopies.

In the near future, the output photon energy range will be further extended to the hard X-ray region by using an electron beam with higher energy and longer undulator sections. More importantly, the achievements of Lutman *et al.* have opened the door for the application of a novel scheme to accurately manipulate electron beams with a passive device. The authors also point out that the scheme could be applied to self-seeding operation in pursuit of achieving longitudinal coherence[8].

Besides, the fresh-slice technique also holds promises for terawatt–attosecond[9] and broad-bandwidth[10] FEL pulse generation. For example, one could first imprint a small vertical tilt on the electron bunch with the dechirper and then directly send the electron bunch to a weak-focused long planar undulator. Since the planar undulator has a symmetric magnetic field gradient in the vertical direction, the electrons with different vertical positions will naturally emit X-ray photons with different energies. Such experiments performed at the existing and the under-construction X-ray FEL facilities will immediately open up new frontiers for X-ray sciences. ❐

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