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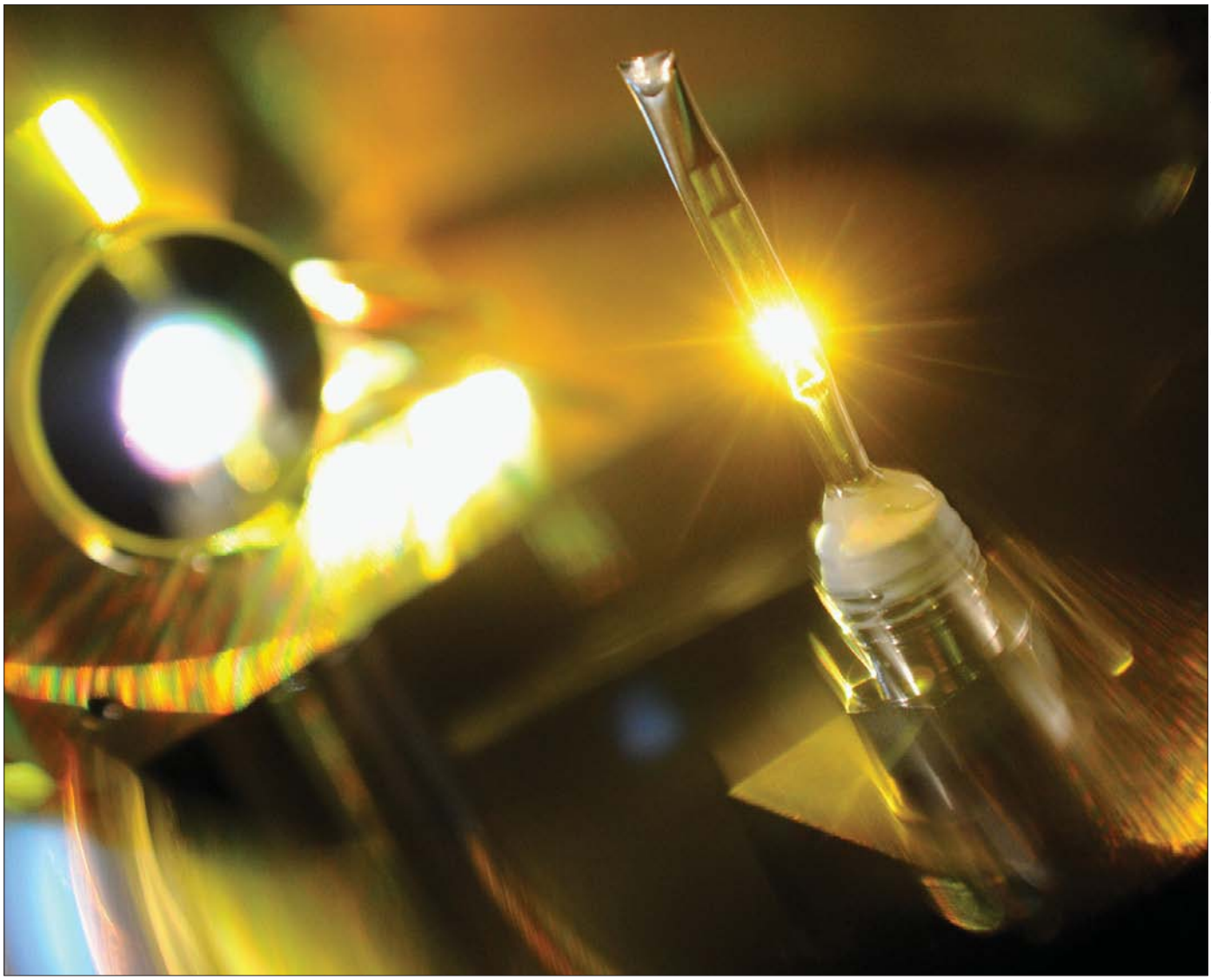
THE LASER AT 50

Beyond ultrafast

As pulsed lasers are developed to resolve dynamics occurring on ever smaller time and length scales, **Adrian Cavalieri** reviews the laser technology that has enabled us to directly observe incredibly fast processes, in fields ranging from atomic physics to molecular biology



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Since the first days of the ruby laser 50 years ago, laser systems have steadily improved thanks to advances in laser gain media and mirror technology. On the one hand, incredibly powerful lasers have been developed for fusion research that are able to deliver huge pulses of energy to their targets, resulting in local environments similar to the interior of the Sun; on the other, we now have lasers that can precisely deliver ultrashort pulses of energy to atomic, molecular and condensed-matter systems to trigger various physical processes and to measure their instantaneous characteristics.

The term "ultrafast" was originally coined in 1982 to describe dynamical processes observed with lasers that occur on the sub-picosecond (10^{-12} s) or femtosecond (10^{-15} s) timescale. However, the phrase has now become essentially outdated, as processes on the attosecond (10^{-18} s or 1 as) timescale – 1000 times shorter than a femtosecond – are now accessible (see box on page 51). Indeed, attosecond spectroscopy could lead to the ability to directly observe charge transfer in photovoltaic cells and transistors, for example, allowing researchers to understand precisely how the electrons move around such devices. This knowledge could help us to create photovoltaic cells that are more efficient, and transistors that switch faster, both of which would impact on our daily lives.

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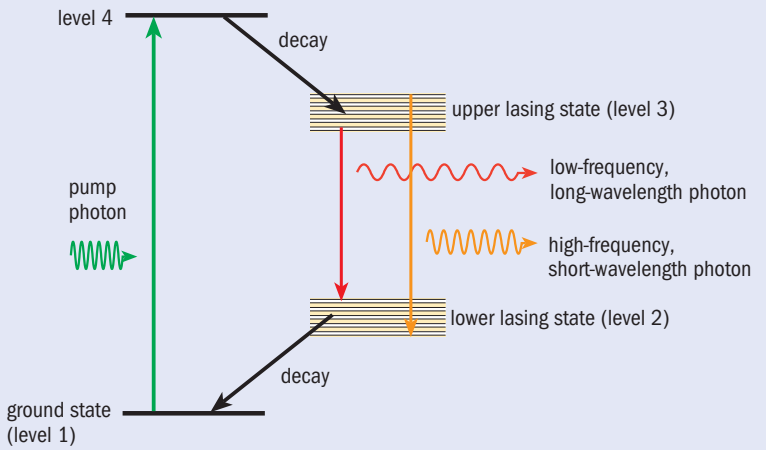
1 Four-level picture



The lasing media used for pulsed lasers allow lasing transitions with a large range of energies, which thus emit photons with a large range of frequencies. Here, the laser gain material – a doped crystal – is "pumped" by photons from an excitation source, such as a very bright white flashed lamp or another laser, specifically tuned to the pump transition energy. Absorbed "pump" photons transfer their energy to electrons in the ground state (level 1), exciting them to a higher energy (level 4). Next, through a decay process such as a vibration, the electrons relax into the upper lasing state (level 3). When the electron population in level 3 is greater than that in level 2, the lower lasing state, a "population inversion" is said to exist and the conditions are set for stimulated emission. When an electron decays from level 3 to level 2, a photon is emitted, which, if a population inversion exists, can "stimulate" another electron in level 3 to decay to level 2, emitting a second photon, and so on. The stimulated photons are emitted in the same direction and in phase with the first, resulting in the emission of coherent laser radiation.

Pump it up

While some lasers operate in a continuous-wave mode, emitting radiation with nearly a single monochromatic frequency, pulsed lasers emit radiation over a broad range of frequencies – billions in fact. These frequency components are timed exactly so that their electric fields nearly cancel each other out, except for during one tiny period of time when they combine constructively in one intense pulse. So when we refer to a pulsed laser, there is no mechanical shutter controlling when the light is emitted; rather, the pulse is created by coherent radiation with many different frequencies interfering.

The frequencies of light that exist in a laser's resonant cavity are determined by two factors. First, they depend on the lasing transitions that occur in the laser gain medium. In pulsed lasers, a gain material is chosen that has many different lasing transitions (figure 1). Second, the cavity dimensions only allow light frequencies for which the electric field has nodes at the cavity's end mirrors. With the right material and cavity it is possible for billions of frequencies to lase simultaneously and generate a short pulse (figure 2).

Because they alternate between a short burst of emission known as the "pulse" and a much longer downtime in between, pulsed lasers can produce high peak powers: since power is energy per unit time, compressing a fixed amount of energy into a shorter time interval results in a higher peak power. Using the 10^3–10^4 W of power from a standard wall socket, for example, lasers can easily produce pulses with terawatt (10^{12} W) peak powers – but only for a few millionths of a billionth of a second.

As in stroboscopic photography, where the fastest motion that can be captured is defined by the camera shutter speed or the duration of the flash, ultrafast time-resolved studies on the femtosecond and now attosecond timescale are generally bound by the duration of the laser pulse. In ultrafast, time-resolved "pump–probe" experiments, a first pulse, called the "pump pulse", is used to trigger a dynamic process, while a second subsequent pulse, called the "probe pulse", is used to observe the system a short period of time later. Today, we can access the attosecond regime, with the production of laser pulses as short as 80 as.

Hand in hand with pulsed lasers is the field of nonlinear optics. Typically, when light passes through a transparent material, the polarization of the material – the distribution of positive ions and electrons – is affected proportionally (or almost) to the intensity of the incident light, i.e. the optics are linear. However, any deviation from a truly proportional response results in nonlinear effects that can become significant if the light propagates through a very thick material or through a long fibre-optic cable, for example. But rather than using low-intensity light and very thick or long materials, nonlinear effects can also be studied using thinner materials and a very intense, coherent laser beam. Thus the field of nonlinear optics began shortly after the invention of the laser in 1960.

Indeed, only a year later, researchers at the University of Michigan showed that laser light could be doubled in frequency when fired into a normally transparent quartz crystal. Since then, we have taken advantage of our understanding of this "second-harmonic generation", and other nonlinear processes, and fed-back the knowledge into the design of pulsed lasers. Coming full circle, nonlinear effects are now inherent in nearly every step of creating a laser pulse, and present a promising route to the next generation of laser technology.

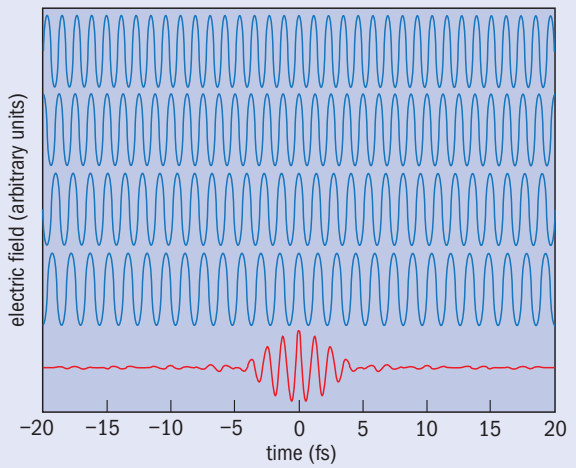
Sapphire to start

One of the first steps towards attosecond pulsed lasers occurred in 1981 with the demonstration of titanium-doped sapphire, or Ti:sapphire, as a suitable broadband laser gain crystal by Peter Moulton at the Massachusetts Institute of Technology. Ti:sapphire is unique in that when it is pumped by a continuous-wave laser, a huge range of lasing transitions can occur. While the laser that pumps the Ti:sapphire is narrowband, i.e. nearly monochromatic, Ti:sapphire emits over a range of frequencies and is broadband. From Heisenberg's energy–time uncertainly principle, $\Delta E \Delta t \geq \hbar/2$, a broad bandwidth of laser energies, or frequencies, is required to produce a short pulse of laser light: the greater the bandwidth, the shorter the pulse.

Pumped by a laser with a wavelength of approximately 530 nm, Ti:sapphire emits light with wavelengths from about 600–900 nm (orange to infrared). But while a broad range of colours, or frequencies, are produced, a pulse is not created without some further intervention. What is needed is for the laser frequencies to be in phase at some point in the cavity.

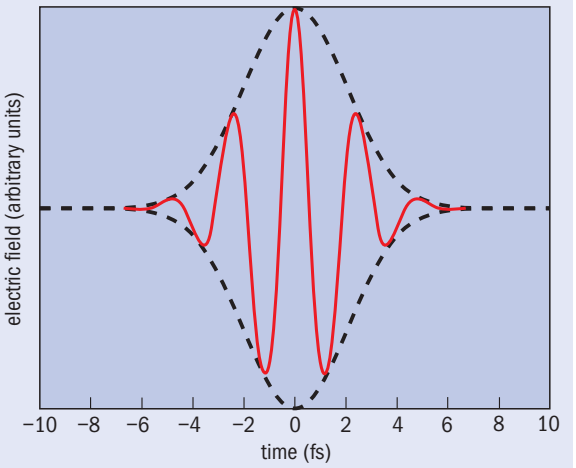
Forcing the various frequencies into phase is achieved through a process known as "mode locking". The laser pulse then exists at the dynamic point in space at which the many frequencies are in phase with each other. This "point of coincidence" moves back and forth in the cavity at the speed of light, as the phase

2 Constructive interference



Frequencies that are in phase sum to produce a pulse. The bottom curve (red) is a sum over the four representative curves shown in blue, as well as about 500 others with frequencies in between. The top-most blue sine wave shows the highest frequency component and the bottom-most wave shows the lowest. The electric fields of the different light beams are in phase and interfere constructively at one moment in time, before falling off quickly and just about cancelling out until the next in-phase moment a relatively long time later.

3 Near-single-cycle optical pulse



This figure shows a simulated near-single-cycle laser pulse, which is nearly the shortest optical pulse that can be generated. The "drive pulse" envelope (black) simply characterizes the pulse and is an artificial construct. The light's carrier electric field (red) drives physical phenomena. The real pulse results from constructive interference of a broad band of frequency components. This optical pulse needs to be converted to extreme-ultraviolet radiation to create an attosecond pulse.

velocity and group velocity of light are nearly identical in air. Each time the pulse passes the "output coupler" where the laser light leaves the cavity, which can be thought of as an imperfect mirror, a portion of the pulse is emitted. The emitted light appears as pulses separated by the cavity round-trip time. Today's most advanced Ti:sapphire oscillators emit optical pulses lasting about 5 fs – barely a few cycles of the electric field and near the fundamental limit. Maxwell's equations of electrodynamics tell us that the shortest pulse of light that can ever be generated is equal to a single cycle of the carrier electric field (figure 3).

Chirping and harmony

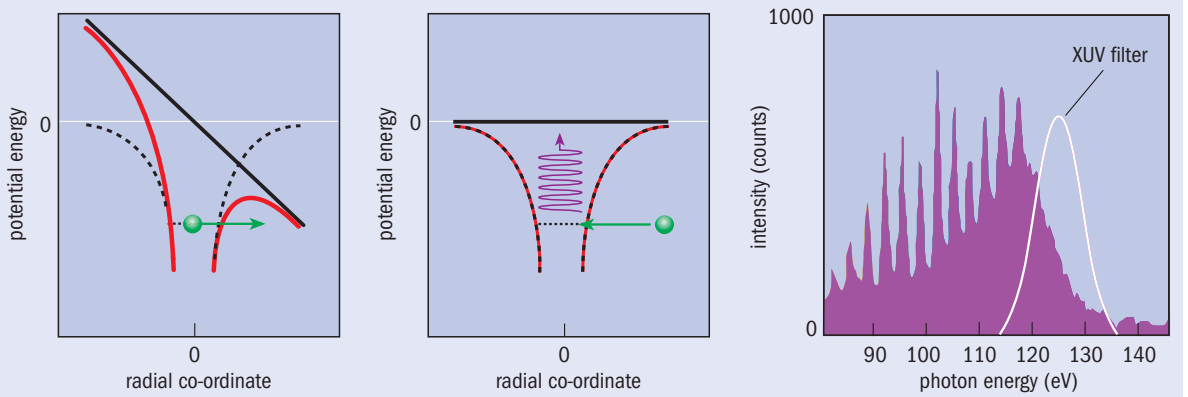
To generate shorter, attosecond pulses, the laser light must be shifted to shorter wavelengths. But before this conversion can be achieved, the pulses need to be much more energetic and so are amplified in energy from several nanojoules to the millijoule level. So as not to damage the crystal used to amplify the energy, "chirped-pulse amplification" – first demonstrated by Gerard Mourou at the University of Rochester in 1985 – is employed, where the pulses are stretched in time to as long as a nanosecond by "de-phasing" their frequency components to reduce the peak intensity of the pulse. Following amplification, the pulses are recompressed in time by putting the frequency components back in phase. Next, the amplified pulse is shortened using a spectral broadening and pulse-time compression stage, which uses nonlinear effects in a noble gas and dispersive mirrors that correct for additional de-phasing in the broadening process. Currently, the most advanced Ti:sapphire amplifier systems utilizing all of these components can produce optical pulses as short as 3.3 fs with up to half a millijoule of energy. So now we have laser pulses that have much more energy than when we started but are still not yet in the attosecond regime.

Because light pulses cannot be shorter than one oscillation of the carrier electric field, or the light's wavelength, the shortest pulse that Ti:sapphire could emit for light pulses centred on 750 nm, or near-infrared (NIR), is 2.5 fs. Evidently, attosecond laser pulses must be composed of shorter wavelengths, equivalent to using photons with higher energies. For pulses shorter than 100 as, this requires the light to be in the extreme ultraviolet (XUV) range, where typical photons have a wavelength and energy of about 12 nm and 100 eV, respectively. Converting the Ti:sapphire NIR photons (at about 1.6 eV) to XUV photons (at 100 eV) requires a significant energy boost. The NIR pulses are converted into attosecond XUV pulses through a process known as "high-order harmonic generation" (HHG) (figure 4). Today, using this method, the shortest pulses are 80 as in duration.

There can be up to 10^8 photons in the attosecond pulse; but while this sounds like a lot, it is not enough to be split into the two pulses required for standard pump–probe experiments and also obtain a good enough signal-to-noise ratio to measure anything. If there were enough photons to do such experiments, the time resolution would be limited only by the attosecond pulse duration, as is the case in more standard

Today's most advanced Ti:sapphire oscillators emit optical pulses lasting about five femtoseconds – barely a few cycles of the electric field and near the fundamental limit

4 From infrared to extreme ultraviolet



Creating attosecond pulses requires a process known as "high-order harmonic generation", in which near-infrared laser pulses are shone into an inert gas jet. The electrons in the gas atoms, originally in an unperturbed atomic well (dotted black line), see a different potential (red) when the laser electric field (solid black line) perturbs the potential. What happens is that the laser electric field folds down the potential of the atomic well (left), increasing the probability that an electron will escape by tunnelling through the barrier and be accelerated away from its parent ion by the same electric field. When the oscillating electric field changes its sign (which happens every half-cycle), the electron's direction of travel is reversed and it is accelerated back towards its parent atom. The electron then recombines with its parent atom (middle) and releases the kinetic energy that it gained while being accelerated in the form of a high-energy, extreme-ultraviolet (XUV) photon. Many electrons take part in this process on each half-cycle of the driving laser field and the result is a broad distribution of kinetic energies at recombination and a corresponding broad band of XUV emission. In the shortest drive laser pulses there are only a few half-cycles of the carrier electric field. At lower XUV photon energies, these photons interfere with each other, resulting in a spiked structure in the measured spectrum (right). In contrast, the highest energy photons are emitted in a single burst, by the electric-field oscillation with the largest amplitude, resulting in an interference-free, smooth drop-off at the highest photon energies. A filter is used to let through only those photons with high energies and a smooth spectrum, resulting in an isolated attosecond XUV pulse.

femtosecond pump–probe experiments. Currently, in attosecond spectroscopy only a single attosecond pulse and the NIR drive pulse are available. Nevertheless, attosecond resolution can still be achieved using a measurement technique now called the "attosecond transient recorder". The attosecond pulse is used to trigger the dynamics, while a much weaker replica of the drive-pulse electric field is used as a probe. The attosecond transient recorder was first demonstrated by Ferenc Krausz and co-workers at the Technical University of Vienna, Austria, in 2004 and has since been the basis for many measurements made in attosecond spectroscopy.

What we gain

Ultrafast lasers have been widely used in industry for micromachining parts, as no material can withstand the intensity of a femtosecond laser pulse. Since only a small amount of energy is used to reach these high intensities, only a tiny amount of material is removed with each pulse, allowing for high-precision cutting. This same property is also exploited in laser eye surgery. Originally discovered through an unfortunate laser eye injury, ultrafast laser pulses can deliver the precise amount of energy required to break protein bonds in the eye without affecting the surrounding tissue.

Attosecond laser pulses, which are not only short but are also composed of high-energy photons, can be used to study inner-shell atomic processes including relaxation following ionization by energetic photons. The study of these processes may be highly relevant to our understanding of how human cells become malignant and cancers develop.

In 2007 attosecond pulses were used by Krausz and colleagues to probe one of the fastest distinct events yet recorded in the time domain. The researchers were able to use attosecond spectroscopy to observe the individual elementary steps of photoemission – the process by which electrons are emitted from a material by light – namely excitation, transport and emission. In fact, electrons from the delocalized conduction-band states were found to be emitted approximately 100 as before the electrons from localized, deeply bound core states. It is not yet precisely clear why such a delayed emission exists, but as photoemission is one of the most fundamental examples of quantum mechanics, experiments will continue until full understanding has been gained.

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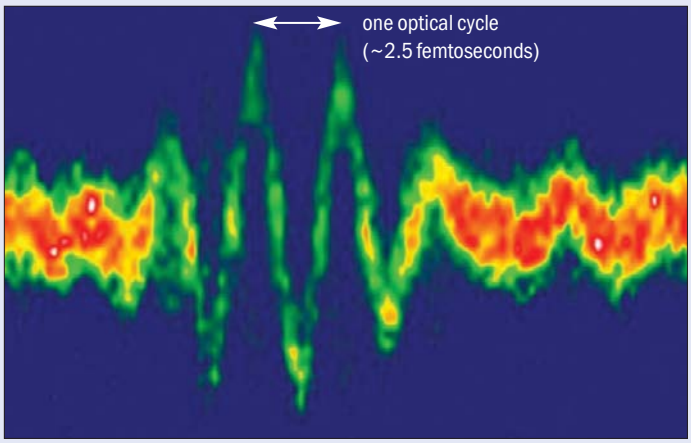
Fast forward

There is a lot we can do with attosecond laser technology, but to make progress, such systems need to be used more widely. With their extremely short but relatively weak pulses, attosecond laser systems complement large-scale "hard" (high photon energy) laser facilities, such as the Linac Coherent Light Source at

Grasping timescales



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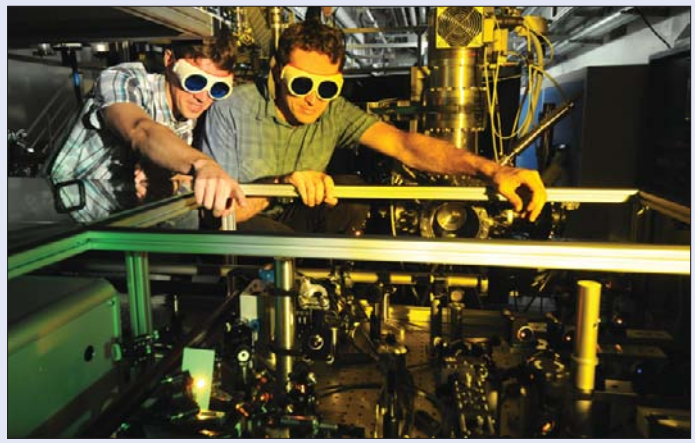

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As laser performance improves, the quantities that define it sound ever more impressive, with each successive prefix more exotic than the last. For power, the numbers become increasingly large – gigawatt (10^9 W), terawatt (10^{12} W) and petawatt (10^{15} W). Pulses, however, are getting ever shorter – picosecond (10^{-12} s), femtosecond (10^{-15} s) and attosecond (10^{-18} s). It is these short pulses from ultrafast lasers that are now letting us explore processes on these timescales in real time.

But these quantities can remain abstract, as it is difficult to relate them to anything in our daily lives. One second is a quantity with palpable meaning, as it is roughly the time between beats of the human heart and is an interval that we can easily perceive. A picosecond, in contrast, is the characteristic time taken for molecules to move back and forth. We are actually able to sense these motions without any fancy instruments, as they are responsible for the temperature of the air and everyday objects.

Getting shorter, the femtosecond regime is the characteristic timescale on which chemical reactions take place, or the time required for bonds between atoms and molecules to be broken and formed. Observing this in real time has been called the "holy grail" of chemistry, letting us study everything from the storage and release of energy in batteries to the fundamental process of photosynthesis.

And finally to the timescale that is the most recent barrier broken by pulsed lasers: the attosecond. On the attosecond timescale, even the making and breaking of chemical bonds appear to occur slowly, and would be akin to watching a slow-motion nature video of shoots unfurling their leaves. In this regime, we are able to resolve charge dynamics – the movement of electrons between the energy levels of an atom, or of electrons or holes (electron counterparts) through an insulating interface such as a p–n junction in a transistor.

the SLAC National Accelerator Laboratory in the US and the European XFEL currently under construction in Germany.

As attosecond laser systems improve, they will let us access ever more complex systems and dynamics. In most experiments, a system can be prepared, its dynamics triggered and subsequently observed every few microseconds on a fresh sample to build up measurement statistics. Therefore, ideally, attosecond lasers would pulse much faster at repetition rates higher than those that are currently possible. However, increasing the repetition rate of current attosecond laser systems is impossible owing to the heating that this causes in conventional laser gain media.

In the future, it will be possible to use nonlinear optics to move beyond the limits of standard laser gain media in attosecond lasers. In a process called parametric amplification, the nonlinear response of certain transparent crystals can be used to couple energy from one frequency of light to another. For femtosecond pulses, parametric amplification can be used to transfer light from a single frequency into a broad band of other frequencies, as is the case in Ti:sapphire. However, in contrast to traditional laser systems, during parametric amplification light is not strongly absorbed, so heating is no longer a problem, allowing higher pulse repetition rates and average powers. The average power output of optical parametric amplifier (OPA) systems could be increased to kilowatt levels. Furthermore, it is possible to achieve even higher gain bandwidths in parametric amplifiers that allow direct amplification of quasi-few-cycle optical light pulses, which is not possible in today's chirped-pulse amplification systems.

OPAs can also be used to create HHG drive pulses with different carrier wavelengths. The maximum photon energy that can result from the HHG process depends on the amount of kinetic energy that the ionized electron can accumulate while being accelerated in the carrier laser field. The longer the electron travels in the electric field, the more kinetic energy it has time to amass, and the greater the emitted photon energy. This interaction time can be increased by using longer-wavelength drive pulses. By moving to longer wavelength, ultrashort drive pulses, the photon energy of isolated attosecond XUV pulses can be increased, thus allowing the efficient generation of pulses deeper in the XUV or even in the "soft" (or low energy) X-ray regime. This is critical, as carbon absorbs XUV radiation at 284 eV making it visible, while water remains transparent, allowing researchers to probe deep inside organic materials with attosecond resolution on atomic length scales.

Looking further ahead, the development of a technique called "quasi-phase-matching" may lead to attosecond light sources producing hard X-ray photon energies and orders of magnitude more photons per pulse, which would allow these sources to compete with large-scale facilities. It has been predicted that the bandwidth of emission will also increase substantially, allowing for the generation of sub-attosecond, that is zeptosecond (10^{-21} s), pulses, at which point it would be safe to say that we have gone beyond ultrafast. ■