ATTOSECOND LIGHT SOURCES

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Since the invention of photography in the 19th century, scientists have been trying to freeze the motion in fast-moving events. Motion pictures of galloping horses were an early application of high speed photography, see Fig. 1. Shutter speeds of cameras are limited to about 1 millisecond, and scientists then turned to brief light flashes to illuminate the subject for microseconds. The invention of the laser in the 1960s then reduced the duration of the light flashes to nanoseconds.

Figure 2 shows the progress that has been made in reducing the pulse duration of laser sources over the past 40 years. For the first 20 years, the shortest pulse duration decreased exponentially, much like Moore’s law in the semiconductor industry. Then a floor of about 5 femtoseconds was reached. The reason for this floor is illustrated in Fig. 3. A pulse of light from a laser source is composed of two components – the carrier frequency corresponding to the optical wavelength of the laser, and the envelope that defines the pulse duration. Figure 3(a) shows a 50 fsec pulse containing many optical cycles, and Fig. 3(b) shows the corresponding Fourier transform. The spectrum is centered at the optical frequency of 375 THz corresponding to the 800 nm laser source.

The spectral width and the pulse duration go hand-in-hand. A pulse of a certain duration requires at least the spectral bandwidth of its Fourier transform. The converse is not true – a broad spectrum does not guarantee that you have a short pulse. The solar spectrum is very broad, yet it is not composed of femtosecond pulses because the different frequencies are not coherent. To work as a laser, the spectral bandwidth must be supported by the gain bandwidth of the laser source. This is the reason that titanium-doped sapphire (Ti:Sapphire) is the laser medium of choice for femtosecond lasers. It has a gain spectrum that covers the range of 700-1000 nm. This is short enough to support a 5 fsec pulse, as shown in Fig. 3(c,d).

SUMMARY

The shortest-duration laser pulses now have durations below 100 attoseconds. This is less than the time it takes an electron to go around a hydrogen atom. This article describes how these new sources work.

Fig. 1 Eadweard Muybridge was an English photographer who moved to San Francisco. In 1877 he created a series of photographs of a horse trotting for former California governor and Stanford University founder Leland Stanford. This is the earliest example of using high speed photography to study moving objects. Attosecond science is at the extreme limit of time resolution.

Fig. 2 Lasers produce the shortest man-made pulses of light. There has been steady progress in reducing the pulse duration, using Q-switched and mode-locked laser sources. A completely different approach was required to break into the attosecond frontier.
To break through the barrier of insufficient gain bandwidth to support an even shorter pulse, it was realized that a shorter wavelength carrier frequency was required. The practical limit to this was around 200 nm, the point at which most materials become absorbing. Physicists discussed coherently combining the output of several lasers to cover a wider spectrum. The lasers would operate at integer multiples of frequency. Figure 4 shows what happens if we add together the electric fields from sources at 1, 3 and 5 times a fundamental frequency. The sum of the fields is seen to be localized into shorter and shorter times.

This approach was seen to be impractical and was never seriously pursued. However, in an example of serendipitous discovery that is so important to scientific breakthroughs, a completely different approach was discovered. It was called high harmonic generation. It was first noticed [1] that an intense picosecond laser beam focused into a gas sample would produce odd harmonics of the laser frequency. This was considered a bit of a scientific curiosity without any application, until it was realized that it provided a source of short wavelength radiation that had the same duration as the laser pulse. An example of the experimental setup is seen in Fig. 5. A 30 fs Ti:Sapphire laser pulse with a wavelength of 800 nm is focused into a pulsed supersonic gas jet. In the forward direction, an xuv spectrometer disperses the spectrum onto a microchannel plate detector. The laser pulse has an intensity of over $10^{14}$ W/cm$^2$, sufficient to ionize the gas sample.

The spectrum is seen to be composed of odd harmonics of the laser frequency, in this case extending to a photon energy of 60 eV. This source is of interest for photoionization of molecules, and is fast enough to be able to time-resolve molecular dynamics. It was also found that the xuv radiation is collimated into a few milliradian beam, and was coherent.
In 1993 the mechanism behind the xuv radiation was identified [2]. It was called the three-step process, and is illustrated in Fig. 6. The time scale for these three steps is the period of a single optical cycle, about 2 femtoseconds. An electron is tunnel-ionized from an atom by a strong laser field. When the laser field reverses direction a femtosecond later, the electron can be driven back to the parent ion. The freed electron is accelerated by the laser field, first away from the ion, and then is driven back towards the ion. Most of the time the returning electron will simply miss the ion, but with a small but finite probability to recombine, and in doing so it will liberate its kinetic energy in the form of a photon.

The simple kinematics of the electron motion is described by Corkum [2]. The maximum kinetic energy of the electron as it returns is $3.17 U_p$, where the ponderomotive potential $U_p = 9.3 \times 10^{-14} I \lambda^2$ (in eV) when the laser intensity $I$ is in W/cm² and the laser wavelength $\lambda$ is in microns. In addition to the kinetic energy, the electron also acquires the ionization potential $I_p$ when it recombines. The highest energy photon produced by this mechanism is thus $E_{\text{max}} = 3.17 U_p + I_p$. By using a very intense laser pulse and helium whose ionization potential is 25 eV, high harmonic emission has been extended to kilovolt photons [3].

This semiclassical description explains the presence of high energy photons, but it does not explain the coherent nature of the emission. A quantum mechanical model does [4]. Figure 7 shows the quantum equivalent of the three-step model for a simple atom like hydrogen. The bound state wave function of the electron is held by the $-1/r$ potential of the nucleus. The oscillation of the charge produced by this interference leads to emission of electromagnetic radiation in the xuv spectral range.

The xuv electric field is shown along with the femtosecond laser field that produces it. Within each optical half-cycle, an attosecond pulse is produced. If the laser pulse has many cycles, many attosecond pulses are produced. The interference of the different pulses in the spectral domain leads to the creation of odd harmonics. If the laser pulse duration is very short, then only a single electron recollision occurs, and only a single attosecond pulse is produced. The shortest pulse currently produced is 80 attoseconds.
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strong laser field \(E(t)\) adds an additional potential \(E(t)x\). If the field is strong enough, it causes some of the bound state wave function to tunnel through the potential barrier and away from the nucleus. When the laser field changes sign a femtosecond later, some of this wave function is pushed back to the nucleus, where it interferes with the remaining bound state wave function. The interference contains a high frequency term that is the difference between the energy of the bound state \((-I_1)\) and the wave function in the continuum \((p^2/2)\). If the total wave function is written as the sum of the bound part and the continuum part, the emission is due to an electric dipole transition between the continuum and the bound parts.

This model also explains why the high harmonic spectrum contains only odd harmonics of the laser frequency. Figure 8 shows the electric field of the high harmonic emission that is produced by a laser pulse. It is seen that an xuv pulse is produced during each half-cycle of the laser electric field. This is because the ionization occurs preferentially near the peak of the laser cycle, and recollision occurs within the same laser cycle. The result is a train of xuv pulses, each with a duration of less than a femtosecond, that repeats every half-cycle of the laser field. The Fourier transform of this pulse train must have a spectral periodicity of twice the laser frequency. The fact that it is at odd multiples of the laser frequency is determined by the alternating left-right parity of the recollision.

The attosecond pulse train that is seen in Fig. 8 can be and is used in experiments. But for true attosecond experiments, a single xuv pulse must be selected. The trick is to choose a laser electric field shape that results in only a single recollision. The most direct way is to make the laser pulse as short as possible, under 5 fsec. This is seen in Fig. 8(b). There are other approaches such as controlling the polarization of the laser pulse, or adding the second harmonic, that also achieve a single attosecond pulse. Single 130 attosecond duration pulses have been measured, and the current record is 80 attoseconds.

As the field of attosecond science is still young, we are still learning about its potential. Attosecond science has been used to observe decay processes in excited atoms, and to measure the emission time of photoelectrons from a metal surface.

The process underlying attosecond xuv pulses is an electron recolliding with a parent ion. This recollision electron is itself an attosecond probe that can measure fast molecular motion. Since the recombination process is a transition between the continuum electron and the bound state wave function, the emitted spectrum contains information on the bound state wave function, and can be used to determine the shape of a molecular orbital or observe molecular vibrations on the time scale of the laser pulse duration or on the time scale of the optical cycle.

**FURTHER READING**

If you are interested in learning more about attosecond science, there are some excellent review articles. For example the production of attosecond pulses is described in; applications are described in.

**REFERENCES**