ATTOSECOND LIGHT PULSES

BY PAUL B. CORKUM AND ANDRÉ STAUDTE

With the invention of the laser, science gained easy access to nonlinear optical processes. Together, lasers and nonlinear optics revolutionized how to make and measure short flashes of light. By 1986 a time resolution of a few femtoseconds was reached — sufficient to access the time-scale of phonons in solids or vibrations of molecules, but too short for most electronic dynamics. While it was quickly realized that the nonlinear interactions of femtosecond pulses with matter had the potential for generating even shorter pulses [1], further progress required a new approach. The conceptual foundation for pushing pulses below the femtosecond barrier was laid out in 1993 [2] but it took until 2001 that this barrier was overcome by experiment [3,4].

Figure 1 shows a timeline of the duration of the shortest light pulses. They determine the fastest processes we are able to systematically measure. The figure shows that we have just passed an important milestone — we can now make laser pulses with durations that are measured in attoseconds. The attosecond time scale is of fundamental interest for physics, chemistry and biology because it is the scale on which electrons move: An electron in a hydrogen atom completes its Bohr orbit in 150 attoseconds. Pulses shorter than this will allow us to freeze valence electron motion and image the bond.

Any type of electromagnetic radiation, from radio waves to hard gamma rays, has its origin in accelerated electric charges. Therefore, the generation of attosecond light pulses requires the control of electron wave packets with attosecond precision. Hence, the part of extreme nonlinear optics that is concerned with the optical control of attosecond electron wave packets has become widely known as Attosecond Science. In the following we will present the essence of how to make an attosecond light pulse. The beauty of attosecond pulse generation lies in its simple concepts: it builds only on fundamental ideas taught in first year quantum mechanics, electrodynamics and optics courses.

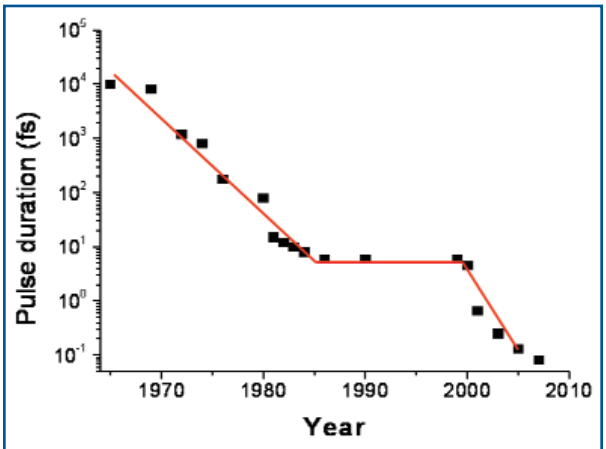

Fig. 1 A time line of the shortest optical pulses that could be produced. The pulse duration followed a Moore's Law-like behaviour — decreasing almost a factor of two each year until 1985. For comparison: Bohr orbit times for valence electrons in atoms and molecules is on the order of 100 attoseconds. Pulses shorter than ~ 100 attoseconds allow us to "freeze" valence electron motion.

SUMMARY

When Theodore Maiman invented the first laser in 1960, he could not have possibly foreseen the full extent of the transformational impact that his development would have on our lives. Since then, the laser has become an everyday commodity and a key technology with a tremendous economical impact that cannot be overestimated. Both, scientist and layman are familiar with its main properties and many of its applications. Considering our visual predisposition it does not surprise that the laser has even entered the arts. Hence, it is probably safe to say that the laser is one of the most important technological developments of the second half of the 20th century.

FEMTOSECOND LASERS – THE ENABLING TECHNOLOGY

Today several different avenues to generate attosecond pulses have been successfully explored. However, all have one key ingredient in common: a state-of-the-art femtosecond laser system. Femtosecond lasers, a success story of their own, are the heart and soul of nonlinear optics. With the invention of femtosecond lasers based on titanium doped sapphire (Ti:Sa) crystals in 1991 by Spence *et al.* [5], femtosecond pulse generation became much more robust. This started a revolution in non-linear optics. Femtosecond pulses had overnight become accessible to all fields of science.

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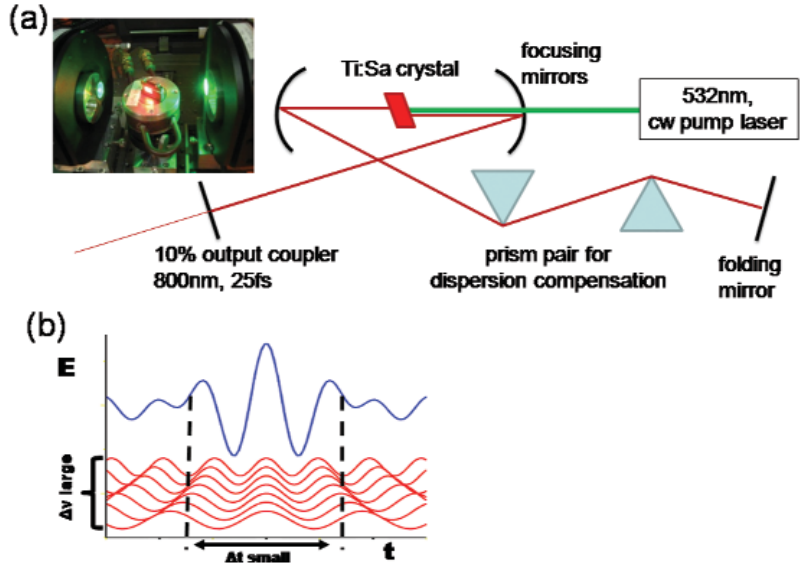

Fig. 2 a) A femtosecond laser cavity based on titanium doped sapphire, the key technical component of nonlinear optics in general and technological basis for attosecond pulse generation. Ti:Sa converts green (532 nm) pump light into fluorescence over a large bandwidth in the near infrared, centered around 800 nm. The cavity is designed that millions of different frequencies are synchronized to yield a single femtosecond pulse that circulates in the cavity.

In Figure 2(a) a sketch of a Ti:Sa femtosecond laser is shown. Remarkably, to arrive at pulses with 25 fs duration no active switching components are required. Instead, the crystal is pumped by a green, narrow band cw-laser! To generate a short pulse the laser gain medium needs to be able emit at a large range of frequencies and couple them coherently as shown in Figure 2(b). The material Ti:Sa provides a large gain bandwidth of 200nm centered around 800 nm, the near infrared. However, stimulated emission forces the medium to lase at a single frequency. Only the intensity dependent, nonlinear part of the index of refraction of the Sapphire crystal enables a cavity design that locks all the frequencies over the full gain bandwidth of the Ti:Sa medium to yield a single femtosecond pulse that roundtrips in the cavity.

Every time the femtosecond pulse has completed a roundtrip. the output coupler transmits a fraction of the pulse. The transmitted pulse can then be amplified in energy by more than a million times and be compressed down to a few femtoseconds in another nonlinear process. As a result femtosecond pulses are obtained that can be focused to intensities of well above 10^{20}W/cm^2, far into the relativistic regime.

IT ALL STARTS WITH TUNNELLING

For attosecond pulse generation these femtosecond pulses need only be focused to moderate intensities (<10^{15} W/cm^2) into a dilute gas target. Now, imagine an intense femtosecond pulse illuminating a single atom or molecule. Figure 3 (b) shows the potential that an atom's electron would experience while immersed in the light wave, near the crest of the field. If the light field is strong enough and the field changes slowly on the time scale of the bound electron, quantum mechanics dictates that the electron will tunnel. For 800 nm and longer wavelength light interacting with many atoms and molecules tunnelling is an excellent approximation [6].

Tunnelling is one of the most fundamental processes in quantum mechanics. It is one of the first of the uniquely quantum mechanical processes that were understood in the 1930's. When tunnelling occurs, quantum mechanics tells us that some probability leaks from the bound state orbital to the continuum. Thereby tunnelling splits the wave function in two (or more) parts. Some of the bound state remains behind — some escapes, becoming a wave packet in the continuum. Hence, in the language of optics, tunnelling acts as a beam splitter for the electron wave function. The continuum part of the wave function is a filtered projection of the bound wave function. We can image the continuum part of the electron wave function, or more accurately, its probability amplitude, by guiding the continuum wave function with electric fields onto a detector [7] (see figure 4). If this procedure is repeated for different orientations of the electron orbital with respect to the tunnelling field a tomographic reconstruction of the bound wave function becomes possible. Hence, tunnelling provides a simple and fundamental a view of the orbital, complementing what we can learn from single photon electron spectroscopy.

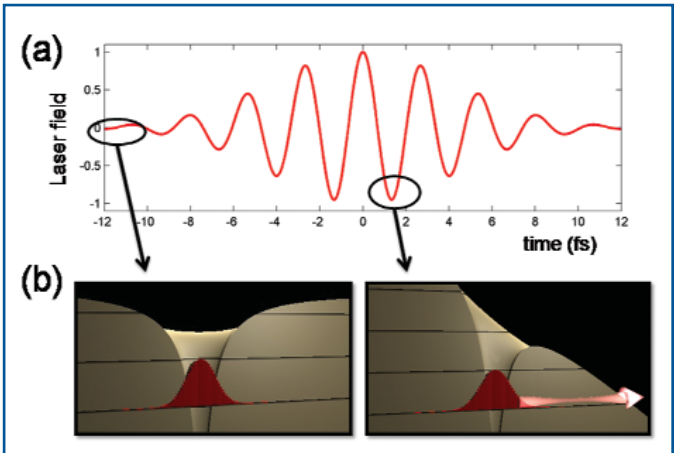

Fig. 3 (a) Electric field of a 10 fs (FWHM), IR (800 nm) laser pulse. (b) Sketch of electron wave function in an atomic coulomb potential for the field free case (left) and when subjected to an intense DC field (right). A part of the wave function can tunnel ionize through the suppressed barrier for a few hundred attoseconds at the field crest during each half cycle.

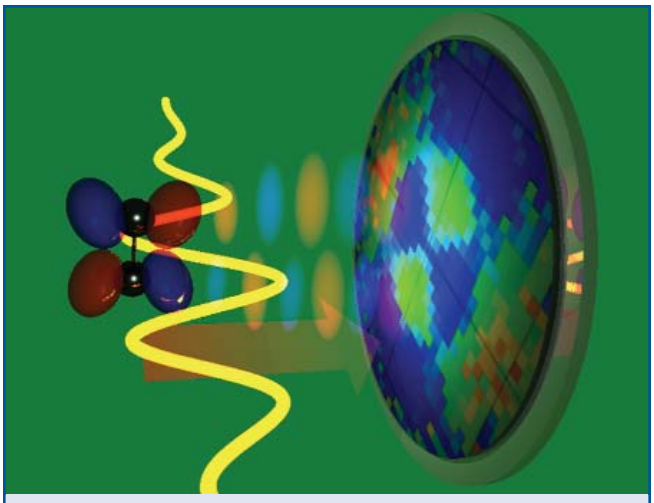

Fig. 4 The tunnelled wave function is a projection of the bound wave function, or the electron orbital, here the highest occupied molecular orbital of the oxygen molecule. If we guide the electron wave onto a detector we can image the orbital [7].

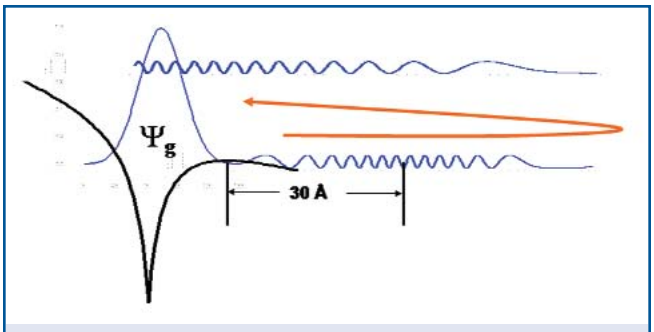

Fig. 5 Tunnelling serves as a beamsplitter. Portions of the initial wave function remain with the atom, but in addition, a wave packet is created in the continuum. These are two parts of the total wave function. Since the electron tunnels in an oscillating field, first it is pulled from the atom but soon the field reverses. Now portions of the wave packet are driven back to the ion. There the wave packet overlaps with the initial wave function. In other words, we have created an electron interferometer.

CONSTRUCTING AN ELECTRON INTERFEROMETER

The attosecond time scale first arises with the initial tunnelling step – the tunnelling rate is only appreciable during a short time near each field crest of the light field [6]. At a wavelength of 800 nm the time window for tunnelling near each field maximum is ~ 300 attoseconds. Once the electron wave function is split, the attraction of the negative electron to the positive ion rapidly decreases while the force that the light exerts on the electron does not depend on how close the electron is to the ion. Therefore, the force exerted by the light wave controls the electron wave packet motion. At first the light pulls the wave packet away from the ion, but soon the field wave reverses direction and the electron wave packet is pushed back [2]. Parts of the wave packet return and pass over their first point of origin, completing the interferometer (see figure 5). In figure 5, the motion of the electron is represented by the red arrow. The figure shows the wave packet sweeping over the ion from which it just departed. This process is often termed a "re-collision". The term "re-collision" emphasizes the particle-like nature of electrons while the electron interfering with itself [8] emphasizes the wave-like nature of the electron.

Before we continue, we would like to emphasize the importance of interferometers in optics. We measure the electromagnetic spectrum with an interferometer – a Fourier Transform Interferometer. We measure the pulse duration with an interferometer – in an autocorrelator. We perform time-resolved measurements with an interferometer – in pump-probe experiments. We can measure the spatial structure of a beam with an interferometer – a sheared interferometer.

Just as an optical interferometer is a critical component in such fundamental tasks as controlling and characterizing light, an electron interferometer is equally valuable for controlling and characterizing electron wave functions. However, in our electron interferometer we cannot directly observe the interference between the continuum and the bound parts of the electron wave function. What we do observe is the radiation emitted as a consequence of the interference. This radiation has been shown to determine the orbital structure of a molecule [9], the duration of the re-collision electron wave packet [10], and resolve bound state electronic wave packets generated by tunnelling [11]. The interferometry of attosecond electron wave packets is a powerful concept that shapes one important direction of attosecond science.

AN OSCILLATING DIPOLE EMITS LIGHT

Figure 6 (a, b) concentrates on the region where the re-collision electron and the initial wave function overlap. This is where the interference actually occurs. The plane wave in the top row images represents the re-colliding electron wave packet. At any given instant of time the electron wave packet is dispersed so much, that it can be well approximated by plane wave passing over the bound wave function. The left and right images represent two close instants in time. Between figures 6 (a) and 6 (b) the re-collision wave packet has advanced by only ½ of its wavelength between the two images. This interference leads to an oscillating dipole (shown in the bottom images at two instants of time). It turns out that the dipole oscillates with a high enough frequency to radiate light well into the XUV range. In this case the frequency of the radiation is equal to the re-collision electron energy plus the ionization potential of the neutral atom. By using only first year quantum mechanics to determine the field required to reach a tunnel ionization rate on the order of 1/T where T is the pulse duration and then Newton's equations from introductory physics to predict the classical trajectory of the electron, one can see that the kinetic energy of the electron can reach as high as 100 eV (or more). In fact, with an infrared driver pulse

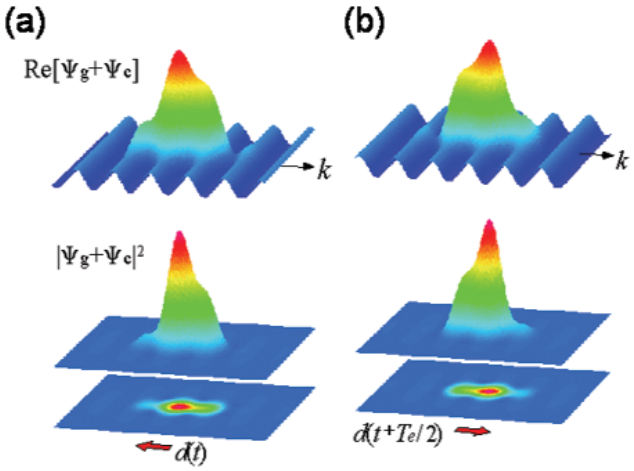

Fig. 6 Two snapshots of the interference are shown. Between them the wave packet has moved about ½ of a wavelength. In the upper figures we see the total wave function – the sum of the two components. In the center row we see the wave function squared. The charge resides on one side of the atom in (a) and in the other in (b). The bottom row figures show the dipole. An oscillating dipole radiates light at the frequency of oscillation. If tunnelling and re-collision are restricted to a single ½ period of the fundamental field, a single attosecond XUV pulse is created.

it can reach as high as 1 keV. The method is more fully described in [12].

Another beautiful aspect of this interferometer is that the transition moment that describes single photon absorption is identical to the oscillating dipole created by the interference between the re-collision wave packet and the bound wave function. In other words, when we measure the XUV radiation we measure the transition moment from the continuum electron to the state from which the electron tunnelled [13]. Therefore, the spectroscopy of the emitted XUV radiation has been called "photoelectron spectroscopy in reverse".

Of course, the signal from single atoms is very small. However, the recombination radiation from each atom coherently combines with the emission from many other atoms creating a strong signal. The experimental challenge is to make the target gas as dense as possible to have as many emitters as possible without affecting the coherence between the XUV frequencies by introducing a significant dispersion from dense media. Thereby we can create a table top source of coherent soft X-ray light – sometimes called a "table-top synchrotron".

THE KEY TO ISOLATED ATTOSECOND PULSES: A SINGLE RE-COLLISION

The three steps of tunnelling, propagation in the continuum and re-collision is repeated every half cycle of the oscillating laser field and therefore any multicycle driver pulse that is intense enough will produce a train of attosecond pulses just by itself [3]! To make an isolated attosecond pulse, all we need is to shape the electric field of the laser pulse to allow only for a single re-collision per pulse. Two approaches to shaping the field have demonstrated successfully the generation of isolated attosecond pulses, illustrated in figure 7. Conceptually simple but experimentally challenging is the approach to compress the driving IR pulse so that only a single field crest would produce significant tunnelling (figure 7(a)). As the bottom panel in figure 7(a) illustrates even a 4fs pulse can produce two re-collisions and hence a "train" of two attosecond pulses. Hence, for an actual experiment the field oscillation under the pulse envelope must be stabilized to the configuration in the upper panel and be exactly reproduced for every laser shot. As of today this approach holds the record for the fastest reproducible flashes of coherent light with 80 attoseconds [14]. The other approach shown in figure 7(b) has less stringent requirements on the pulse duration and is experimentally much simpler to implement. The so-called polarization gating [15] is based on two circularly polarized pulses of opposite helicity that are overlapping for a few oscillations of the electric field. In the overlap

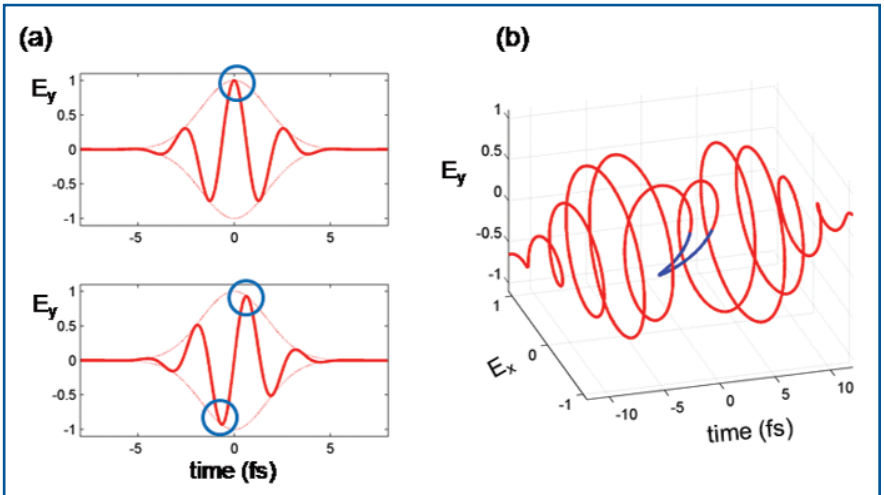

Fig. 7 Basic femtosecond pulse shapes used for generation of isolated attosecond pulses. (a) Linearly polarized, 4fs (FWHM) IR pulses. The fundamental frequency needs to be phase stabilized relative to the envelope in order to avoid the ambiguity between two equally long pulses but a different number of field crests that can generate attosecond pulses (indicated by blue circles). (b) Polarization gated re-collision with two overlapping 10fs pulses that are circularly polarized with opposite helicity. In the overlap region the circular polarizations of both pulses cancel to yield a single half cycle of linearly polarized light (blue).

region the opposite circular polarization cancels and the optical field becomes linearly polarized. As linear polarization is a prerequisite for re-collision, it is critical for generating XUV radiation via recombination. By adjusting the delay between the two pulses the effective field can be tailored such that re-collision will be possible only during a single half cycle, and hence the generation of an isolated attosecond pulse. Currently, this method has been shown to yield pulses as short as 130 attoseconds [16].

CONCLUSION

Attosecond science is founded upon extreme nonlinear optics. Extremely intense light permits us to create and steer free electron wave packets on the time scale of the oscillating electric field - a natural attosecond time scale. Re-collision produces the shortest duration light pulses (by a factor of ~20) currently available. This is a general property of multiphoton interactions – re-collision is just the first of many new routes to attosecond and sub-wavelength science. The achievable pulse duration is only limited by our ingenuity to invent new ways to coherently couple ever more frequencies of the electromagnetic spectrum. The current technology encourages speculations that we may see the fall of pulse durations below a single attosecond in as little as a few years from now.

Along the way to racing for ever new records in the shortest pulse durations, attosecond science has been cross fertilizing other fields: from laser development, universal molecular alignment, laser induced particle acceleration, to the development of new, much improved atomic clocks. Finally, if high order multiphoton processes allow us to operate below the light period, it must also allow us to operate below the light wavelength – space instead of time. Therefore, attosecond science also opens a route to Ångstrom-scale molecular imaging [17].

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