Attoscience goes OPCPA

New Laser Developments in High Harmonic Generation

Time Resolution

Time resolution is key to the study of dynamic processes. When the appropriate resolution is lacking, indirect evidence is helpful but nothing is as convincing as to actually see what's happening. The often cited dispute about the horse galloping illustrates this fact: does the horse lift all feet off the ground for one moment? The definite answer is yes - given by the first slow motion movie ever made [1]. Today's cameras are much faster taking 1000 pictures per second. All of us know the beautiful slow motions of flying birds or bursting air balloons. With the advent of pulsed lasers, the race for ever faster captures of moments in time began – leaving the twinkling of an eye far behind. This is the story of the Titanium:Sapphire laser which became widespread in our laser labs in the 1990's and is nowadays the work horse in ultrafast science. For almost one decade it dictated the shortest possible snapshots. Only a few femtoseconds (fs) long are the laser pulses of elaborate versions but the standard pulse duration amounts 100 fs. To imagine this tiny fraction of time we have to rely on the fastest velocity that we know, the velocity of light: light travels in roughly one second from earth to the moon but can hardly cross a human hair in just 100 fs.

Femtoseconds are Prominent in Chemistry

In 1999 the Nobel prize was awarded to Ahmed Zewail for his contribution to spectroscopy with femtosecond time resolution [2]. It may come as a surprise that the category was chemistry and not physics although most ultrafast laser scientists were physicists at that time. When looking at the background however, it becomes clear: the femtosecond time scale belongs to the realm of chemical reactions like bond breaking and forming, dynamical energy distribution inside molecules or energy transfer between molecules. The underlying physics traces the breaking of a chemical bond back to the vibrations of the two nuclei of that bond. The nuclei are surrounded by shared electrons that actually form the chemical bond. Put in simple words: when the vibration becomes too strong, the bond breaks because the electrons can't be shared over the increasing distance but are tightened to one or the other nucleus. For a better understanding, it is important to know how the motion of the electrons is connected to the motion of the vibrating nuclei. According to the well-established Franck-Condon-principle, the electrons immediately follow the motion of the nuclei. But what means "immediately"? For chemistry this means that it is much faster than one femtosecond. For the underlying physics it means that we require attosecond time resolution - that is 1000 times faster than one femtosecond. Therefore, the question is answered in more detail by attosecond physics also known as "Attoscience" which looks fast enough to follow electrons inside atoms.

Generating Attosecond Light Pulses in Three Steps

The Ti:Sa laser can't surpass the limit of a few femtoseconds which was the universal limit for time resolution until 2001. Then P.M. Paul as well as H.M. Hentschel achieved the breakthrough by the generation of attosecond light flashes [3,4]. One funny side note worth mentioning is that the group led by Ferenc Krausz got an entry in the famous Book of Guinness World Records in 2008 for the shortest flash of light that lasts only 80 attoseconds [5]. The theoretical background was published 15 years earlier and explains the attosecond generation process by a semiclassical three-step model: 1. Tunneling 2. Acceleration 3. Recombination [6,7].

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The process involves an extremely powerful pulsed infrared laser focused on an ensemble of noble gas atoms (see Figure 1). Step 1: the very strong electric field of the laser bends the coulomb potential of the atom in such a way that one of its electrons – described by a quantum-mechanical wave packet – can tunnel through the finite potential barrier. Step 2: the electric force of the oscillating laser field accelerates the freed electron wave packet and finally drives it back towards the ionized atom. Step 3: When it collides with the parent ion again, they recombine generating an attosecond flash of extreme ultraviolet (XUV) light. Such very weak flashes are generated in millions of atoms synchronously and in such a manner that they build up coherently into an intense laser-like beam. The three steps require three half-cycles of the laser field oscillations. But each step can happen at the same time in different atoms: while one electron wave packet tunnels, another is accelerated and a third wave packet crashes into its parent ion during the same half-cycle. The half-cycles last only about 3 fs and so an entire pulse train of many

XUV pulses is generated within one infrared laser pulse. Another consequence of this half-cycle periodicity in conjunction with the coherent build up is the fact that the XUV light is considered as a high harmonic of the fundamental infrared laser. Therefore this parametric process is also called High Harmonic Generation (HHG).

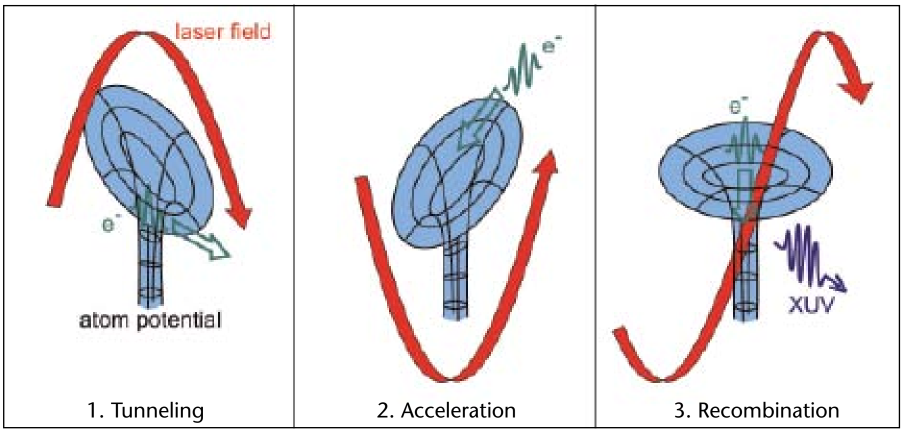

FIG. 1: **Visualization of the three steps in attosecond pulse generation.**

New Trends in High Harmonic Generation

The first HHG setups were based on powerful Ti:Sa lasers: a complete laser chain consists of an oscillator, a regenerative amplifier and several multi-pass amplifiers – all of them equipped with strong green lasers for pumping the gain crystals. Although the complete system fits into a laser lab it occupies the entire room and is rather complex. For example, the heat management of the gain crystals is not an easy task but crucial to avoid "thermal lensing" which destroys the beam quality tremendously. But also other unwanted effects become more prominent and decrease the laser qualities when scaling the Ti:Sa laser to high powers. Therefore laser scientists looked for alternative approaches. The most promising concept in power scaling is called optical parametric chirped pulse amplification – OPCPA in short (see Figure 2). For ultrafast pulse amplification, the extremely high peak powers are the first limit to overcome. By chirping the pulses they become stretched in time and their peak power drops efficiently. After the amplification of the stretched pulses they are recompressed to nearly their Fourier-transform limit resulting in extremely high peak powers. Because the compression is done in free space, no gain or other optical material needs to withstand the destructive peak power. The three steps – stretcher, amplifier and compressor – are the heart of the chirped pulsed amplification (CPA) scheme [8]. CPA is implemented not only in OPCPA setups but also in most regenerative amplifiers built today. In contrast to the amplification with laser gain media, optical parametric amplification is a direct interaction of light fields mediated by the nonlinear polarization they induce in certain crystals. These so-called "nonlinear crystals" can be viewed as catalysts promoting the interaction but not exchanging energy with the light fields. Therefore the process is attributed "optical parametric" and the energy and momentum conservation (=phase-matching) must be fulfilled by the interacting light fields themselves. These restraints can lead to a directed energy flow from one strong light field referred as "pump" to a weak light field referred as "signal". As further consequence an additional third light field called "idler" is generated.

Advantages of OPCPA

Similar to Ti:Sa lasers where energy flows from the green pump laser to the red Ti:Sa output, the energy of the pump field is converted into the red-shifted signal field. On the other hand, many differences account for the many advantages of OPCPA over Ti:Sa-CPA. First, there's only very reduced thermal load to the crystal because absorption is avoided as much as possible whereas in gain media this is the fundamental ingredient. In lasers, the so-called quantum defect meaning the energy difference of a pump photon and the laser photon is transferred to the laser crystal resulting in heat that needs to be removed by active cooling. In high power lasers this becomes an issue as mentioned above. In OPCPA on the other hand, the energy difference is converted into idler photons leaving the crystal without hindrances. Without the urgent need for cooling, the diameter of the crystals and consequently the diameter of the light fields can be increased for scaling purposes: the power impact per area is effectively reduced to stay well below the damage threshold of the material. Secondly, because of this scaling technique and the high gain, the nonlinear crystals can be made very short along the propagation axis. This has very positive effects on the beam quality because the nonlinear phase distortions (=B-integral) accumulated by the laser pulse while passing through material is reduced to moderate values. Thirdly, the gain bandwidth of OPA can be made much broader than that of lasers.

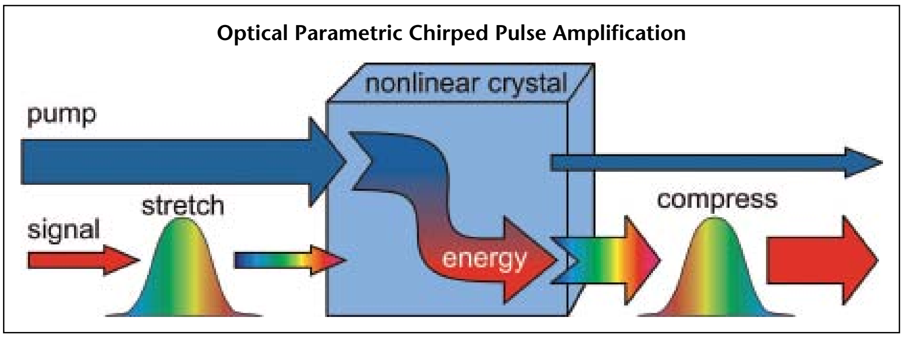

FIG. 2: **Schematic Drawing of Optical Parametric Chirped Pulse Amplification (OPCPA).**

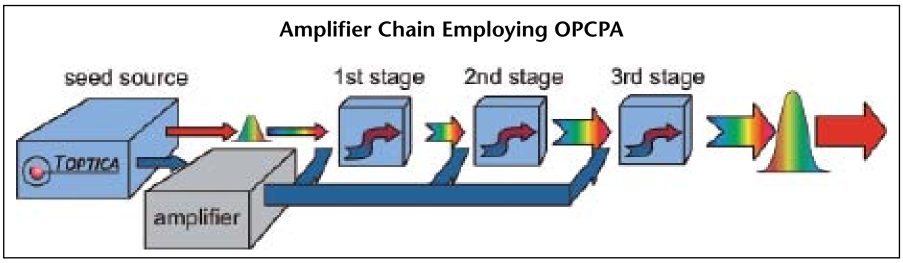

FIG. 3: **Example of an OPCPA setup employing TOPTICA Photonics's fiber seed source.**

This requires some tricks like non-collinear geometries [9] or operating the OPA near its degeneracy point [10]. The ultrabroad bandwidth supports the shortest possible laser pulses which only last a few if not one single optical cycle. The list of advantages can be further extended, e.g. surpressing spontaneous amplification.

Seed Source for OPCPA

The major drawback of OPCPA is that two laser pulses with different wavelengths are necessary – the pump and the signal. They must be synchronized and have complementary requirements: the pump can be narrow in bandwidth but needs to be powerful whereas the signal is preferably ultrabroad but can be very weak. Therefore the signal is usually generated out of the pump by focusing the latter into a sapphire plate: highly nonlinear processes take place resulting in a broad supercontinuum. These processes are very sensitive to intensity fluctuations of the pump and are one major source for phase and intensity noise of the amplified signal. Therefore it is desirable to generate the signal in a different way, e.g. by fiber technology. Here, supercontinuum generation doesn't need high powers because the necessary high intensities are maintained over long distances due to the single mode waveguide. Also amplifying a weak supercontinuum to the moderate signal power is straightforward with rare earth doped gain fibers.

TOPTICA Photonics developed the ideal seed source for OPCPA based on fiber technology (see Figure 3). The ultrabroad signal is generated by an Er-doped fiber laser that is broadened in a nonlinear fiber. The broad bandwidth supports laser pulses as short as 25 fs with moderate average powers of 180 mW. The optically synchronized pump is generated by shifting a portion of the laser output in another nonlinear fiber from 1.5 µm to 1.03 µm. Filtering and amplifying the shifted wavelength leads to the perfect seed pulses for a third party Yb-doped solid state amplifier that delivers the narrow but strong pump pulses. Not only the perfect timing of both laser outputs is ensured but also the phase relations of both outputs are preserved. They share the same phase-slip of the carrier in respect to the envelope of the laser pulses which is always present in mode-locked lasers. The OPCPA process removes by its nature this unwanted phase-slip in the generated idler pulses. This is known as passive Carrier-Envelope-Phase-Slip (CEP) stabilization and is an important feature in few-cycle laser pulses. With the passive CEP stabilization it is easily possible to maximize the electric field amplitude of every laser pulse. And this is what matters most in attosecond pulse generation as described above by the theory of the three-step model.

THE COMPANY

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TOPTICA develops and manufactures high-end lasers and laser systems for scientific and industrial applications in the three following technology fields: diode and fiber lasers as well as Terahertz system design. The lasers provide exceptional specifications for a clientele as diverse as life sciences, semiconductor industry or quality assurance. With this, TOPTICA Photonics AG has become one of the leading laser photonics companies in Europe.

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