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Chirped-pulse amplification: the technology and its applications

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1. Introduction

Ultrashort pulses of light generated with lasers have enabled a wide range of applications during the past decades. In the visible to near-infrared spectral region, these electromagnetic wavepackets have durations in the picosecond to femtosecond range. Besides their short duration, which enables time-resolved measurements of some of the fastest physical and chemical processes, many applications are based on the fact that very high peak power and intensity can be reached with moderate amounts of energy per pulse. A typical ultrafast laser oscillator emits pulses with an energy of several nJ and peak power approaching a MW. State-of-the-art values exceed these typical numbers by orders of magnitude [1, 2]. By sending the beam through additional optical amplifier stages, pulse energies can be raised to many J, with peak power and intensity scaling accordingly. A MW-peak-power pulse focused to a spot size of 10 μm^2 yields a peak electric-field strength approaching 10 GV/m. The easily accessible high light intensities give rise to a multitude of nonlinear optical interactions that were out of reach with traditional light sources. These light pulses can break chemical bonds, which is a property that is used for precise, non-thermal machining of a wide range of materials, including transparent or heat-sensitive compounds. However, it is exactly these attractive properties that was hindering progress in developing sources of intense ultrashort laser pulses for a long time. While nonlinear optical interaction or the processing of materials is desired in many applications, it was a major challenge to prevent these effects from occurring where they were entirely unwanted: within the laser system itself.

One half of the 2018 Nobel Prize in Physics was awarded to Donna Strickland and Gérard Mourou for the invention of the chirped-pulse amplification (CPA) technique [3, 4]. CPA was a technological breakthrough that addressed the above problem and enabled new generations of ultrashort pulse amplifiers that produce laser pulses with many orders of magnitude higher peak power and intensity than what was previously considered possible.

2. Ultrashort pulse propagation

To understand the operation principle of CPA and the motivation behind it, we first need to understand how ultrashort optical pulses propagate through materials. We will limit the discussion to media with negligible absorption in the optical frequency range covered by the pulse, which is a reasonable assumption for the typical optical components being used inside a laser system.

The Fourier-relationship between time and frequency implies that a short pulse in time domain must exhibit a broad spectral power distribution in frequency domain. In this Fourier picture, an ultrashort optical pulse is a superposition of monochromatic (i.e., single-frequency) waves covering a wide range of different frequencies (Fig. 1a). If all of these monochromatic waves add up in phase, the resulting pulse is the shortest for a given spectral power distribution.

The propagation of monochromatic waves is governed by the refractive index of a material. In any material other than vacuum the refractive index is a function of frequency. The presence of such dispersion immediately implies that if the constituent frequencies were in phase at the input of a block of material, then this will in general not be the case anymore at its output (Fig. 1b). The resulting output pulse will thus be longer than the initial pulse. The shorter the initial pulse, the broader its frequency spectrum, the more the refractive index will vary across the pulse spectrum for a given material. Shorter pulses are therefore affected more strongly by dispersive pulse broadening than the more narrowband longer ones. This is illustrated for propagation through a typical optical material in Figure 1c.

A more detailed analysis of how dispersion affects ultrashort pulses shows that the curvature – i.e., the second deriva-

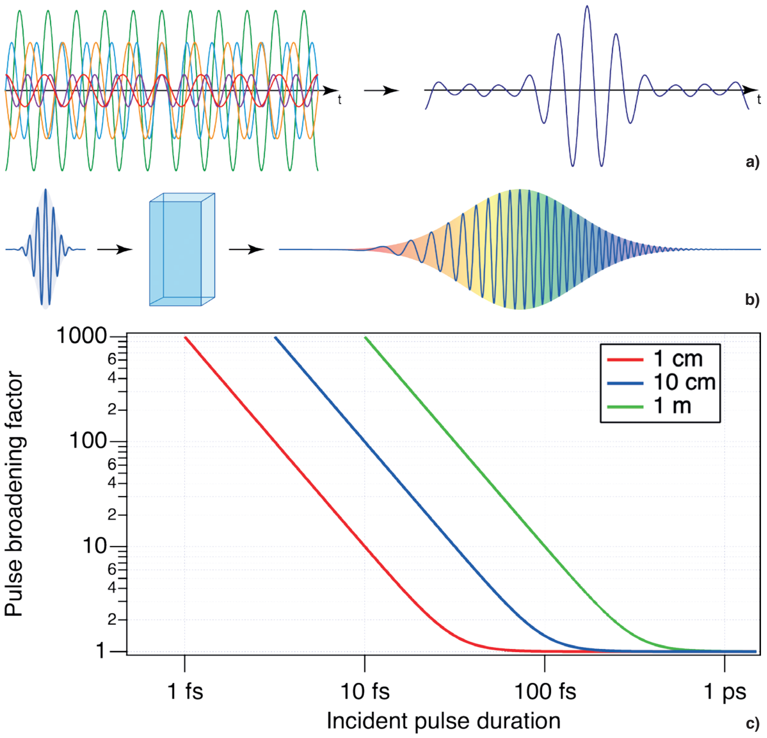


Figure 1: Ultrashort optical pulses. a) A superposition of five monochromatic waves with different frequencies (left) leads to short wavepacket of light (right). Superimposing a broad continuum of frequencies leads to very short isolated pulse. b) An initially short pulse gets stretched in time after passing through a slab of material. This is a result of the dispersion of the refractive index with frequency that is found in any real material. c) Pulse broadening experienced by an initially short Gaussian pulse with 800 nm center wavelength after propagating through fused quartz. The curves indicate the pulse broadening factor as a function of the incident pulse duration for three different thicknesses of fused quartz. An initially 10 fs long pulse will thus be 1 ps long after only 10 cm of fused quartz, whereas a 1 ps pulse will not have noticeably broadened after the same amount of material.

tive – of the refractive index as a function of frequency is the lowest order that broadens the pulse. If this curvature is positive for a material at a given frequency, this material is said to exhibit positive dispersion and negative dispersion for negative curvature. In a positive dispersion material, lower frequencies propagate faster than higher frequencies. As a result, a pulse where all superimposed frequency components perfectly line up at the input to a material will display a frequency ramp from low to high frequencies at the output (Fig. 1b). Based on the sound a corresponding acoustic wave would make, this variation of the instantaneous frequency across the pulse is referred to as chirp. In the visible to near-infrared spectral region, where most ultrashort pulse lasers operate, essentially all optical materials exhibit positive dispersion and thus lead to a chirp from low to high frequencies (called a positive chirp).

The compensation of dispersive pulse broadening or chirp is one of the main considerations when working with ultrashort optical pulses. After all, one usually aims for having the shortest pulses available at the position where the pulse interacts in an experiment or application. Since for the most common laser frequencies all materials impose the same sign of dispersion, compensation can typically only be achieved with artificial structures, which will be briefly discussed below.

As stated in the introduction, a short pulse can reach considerable intensities with moderate amounts of energy. As a consequence, the refractive index of a material will not only depend on frequency, but may also depend on intensity. The lowest order intensity-dependence of the refractive index that can be found in all materials is mediated by the optical Kerr effect. The resulting refractive index can be written as $n = n_0 + n_2 I$, with n_0 representing the normal, linear optical refractive index found at low intensity, n_2 the nonlinear refractive index coefficient, which is a material property, and I the light intensity, which can be a function of time and spatial coordinates. From this simple relation follows that an ultrashort optical pulse, that by definition has an intensity that rapidly changes with time, will induce a rapid refractive index modulation if it has sufficient peak intensity. This rapid modulation acts back on the pulse itself (therefore referred to as self-phase modulation) and results in the generation of new optical frequency components. In conjunction with dispersion, self-phase modulation can lead to unwanted pulse distortions.

Considering a laser beam with a typically Gaussian transverse intensity profile, the optical Kerr effect will impose a Gaussian refractive index profile in the spatial domain through the above relation. Similar to the self-phase modulation, this refractive index profile will act back on the beam itself. A Gaussian refractive index profile (or, in general, any approximately parabolic profile) will have a focusing effect on the beam. This self-focusing leads to a further concentration of the light, which in turn leads to an increased intensity and thus stronger self-focusing [5]. It is clear that such a positive feedback loop can lead to damage in optical components if the propagation length through the material is sufficiently long. The higher the initial intensity, the shorter the propagation distance after which damage through self-focusing can occur. Even if the intensity for damage in one component is not reached, the self-focusing action can cause the beam to shrink further downstream in an optical setup and cause the beam to exceed the damage threshold at a remote piece of optics (Fig. 2).

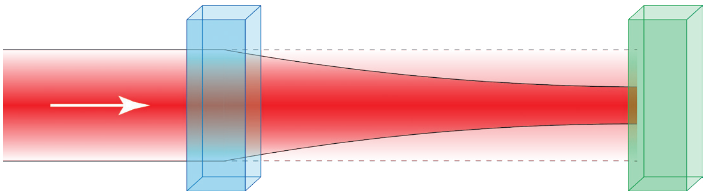


Figure 2: Self-focusing of a laser beam through the optical Kerr effect. While a low-intensity beam passes the blue material without changing its divergence (indicated by dashed lines), a high-intensity beam experiences self-focusing due to the intensity-dependent refractive index. Depending on the propagation length in the material and the intensity of the beam, laser-induced damage may occur within the focusing material directly (blue slab) or further down the optical path on a remote optical surface (here, the green slab).

The nonlinear effects described above with their resulting beam distortions and material damage were for a long time hindering the development of intense sources of ultrashort optical pulses. This roadblock could only be overcome with the invention of CPA.

3. Chirped-pulse amplification

The basic idea of CPA is to prevent the unwanted nonlinear effects and ultimately damage of optical components by stretching the pulse considerably before its amplification. In a typical femtosecond amplifier system, the pulses would be stretched to many nanoseconds prior to amplification. Such a stretching factor of about 10^5 to 10^6 leads to a corresponding reduction in peak power and intensity at a given pulse energy. As a pulse stretcher, we can use propagation in a dispersive medium as described above. Indeed, propagation through a glass fiber was used in the first demonstration of CPA by Strickland and Mourou [4]. However, in the application or experiment, one would still like to have a short pulse with the associated high peak power and intensity. After amplification, the pulses are therefore recompressed by imposing dispersion with the opposite sign to that of the stretcher. How can such a pulse compressor be implemented given that at the typical visible to near-infrared operation wavelengths of the most common amplifier systems materials only exhibit positive dispersion?

Effective negative dispersion can be obtained even in this wavelength region with artificial structures exploiting frequency-dependent diffraction, refraction or interference. The most common pulse compressor type for high-peak-power amplifiers is depicted in Fig. 3a) [6]. It exploits the angular dispersion of the polychromatic ultrashort pulse upon diffraction off a grating. A second identical grating compensates the angular dispersion of the first one, leading to all frequencies forming parallel beams at its output. A second pass in opposite direction through this arrangement assures that all frequency components are collinear again. A detailed geometrical analysis of this setup shows, that low frequencies (indicated in red in Fig. 3a) always have to travel longer pathways than the higher frequency components of the pulse (blue). Therefore, at the output of the compressor, the high frequencies are advanced with respect to the low frequencies, which can compensate a positive chirp that was previously imposed by a pulse stretcher. The amount of negative dispersion added by the compressor can be tuned by varying the distance between the two gratings.

The solution used in the original paper by Strickland and Mourou with a pulse stretcher using material dispersion and a grating compressor is not ideal. It is found that the compressor can compensate the chirp imposed by the stretcher only to lowest order. Uncompensated higher dispersion orders lead to unwanted pulse distortions, effectively reducing the maximum achievable peak intensities by not being able to concentrate all the available pulse energy into a single light burst of minimum duration. Martinez et al. [7] demonstrated that by adding two identical lenses with focal length *f* between the gratings of the pulse compressor, the resulting arrangement produces positive dispersion if the gratings are placed at a distance $L < f$ from their respective closest lens (Fig. 3b). Even better, the dispersion, including higher orders, will correspond to that of a grating compressor with a grating separation of $L - f$. This means, the addition of the lenses effectively produces a grating compressor with a negative grating separation. Since the magnitude of all dispersion orders is proportional to the grating separation, their sign is now flipped compared to a compressor with the same, but positive distance between the gratings. As a result, a grating compressor can fully compensate the dispersion imposed by a grating compressor to all orders. In practice, since also the amplifier material and other components along the beam path will add positive dispersion to the pulse, dispersion compensation will never be absolutely perfect across all orders. The system is then optimized for minimal dispersion in the dominating lowest orders.

In a standard laboratory sized amplifier system, the ultrashort pulses at the beginning of the CPA chain have a duration of tens of femtoseconds with nJ pulse energy. These pulses are stretched to nanoseconds duration and then amplified to many mJ energy. After recompression, the pulses are roughly back to their initial duration, but now with tens of GW peak power. In larger systems, final energies exceed several J and the peak power can reach up to 10 PW. It becomes clear that with such high peak powers and the associated intensities, care has to be taken with respect to the materials and components along the beam path following the compressor. As a result, for the highest-peak-power systems, the compressor and all of the subsequent beam path are placed in vacuum as otherwise even air would induce substantial self-focusing.

While the idea behind CPA appears to be simple, it represented a major breakthrough for intense laser science and associated fields. It enabled a broad range of applications and research directions that would otherwise be out of reach. It is for this tremendous impact that Donna Strickland and Gérard Mourou have been awarded with the 2018 Nobel Prize in Physics.

4. Applications of chirped-pulse amplification

CPA has enabled the construction of comparably compact high-peak-power and high-intensity laser systems that achieve their parameters by compressing moderate amounts of energy into very short – typically femtosecond – pulses. A relatively standard laboratory-sized system can now produce pulses with a peak power exceeding a TW.

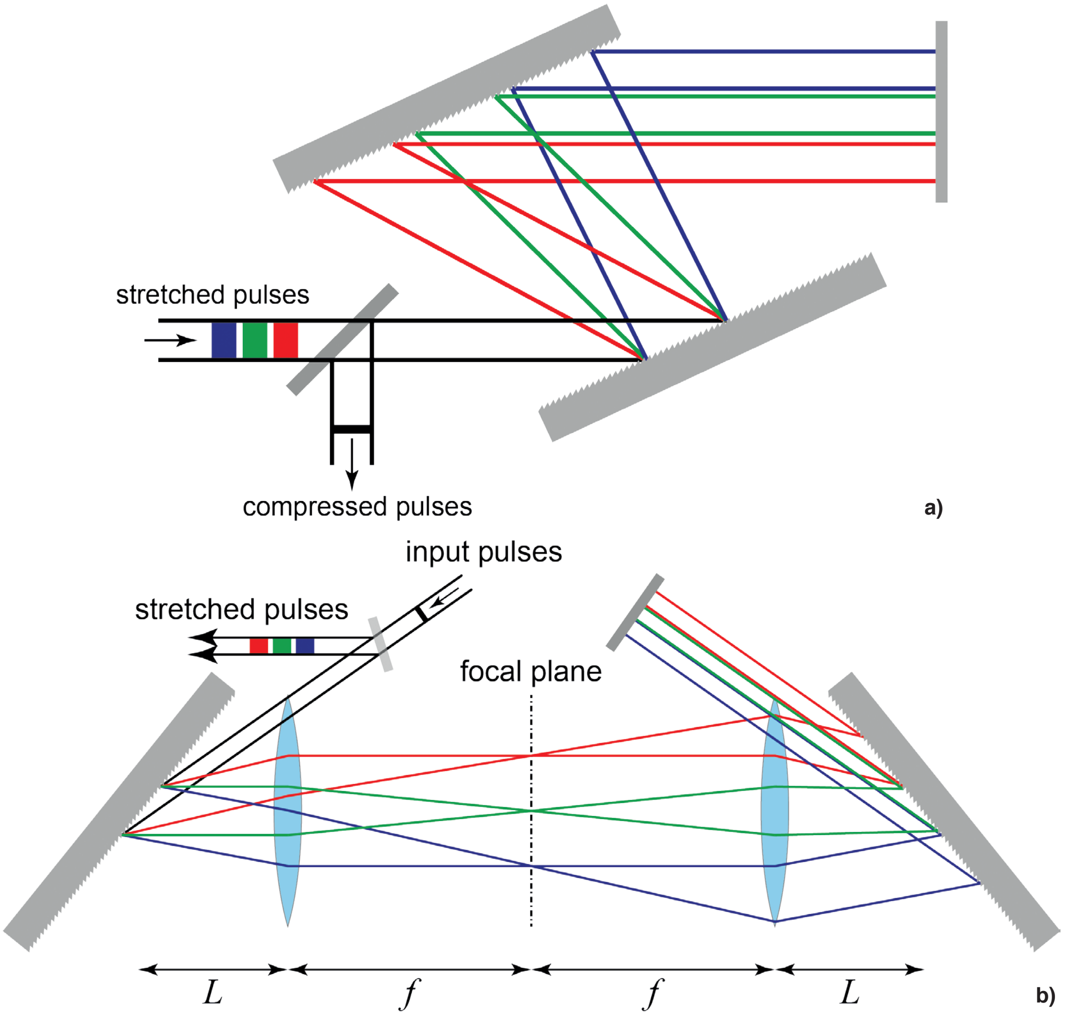
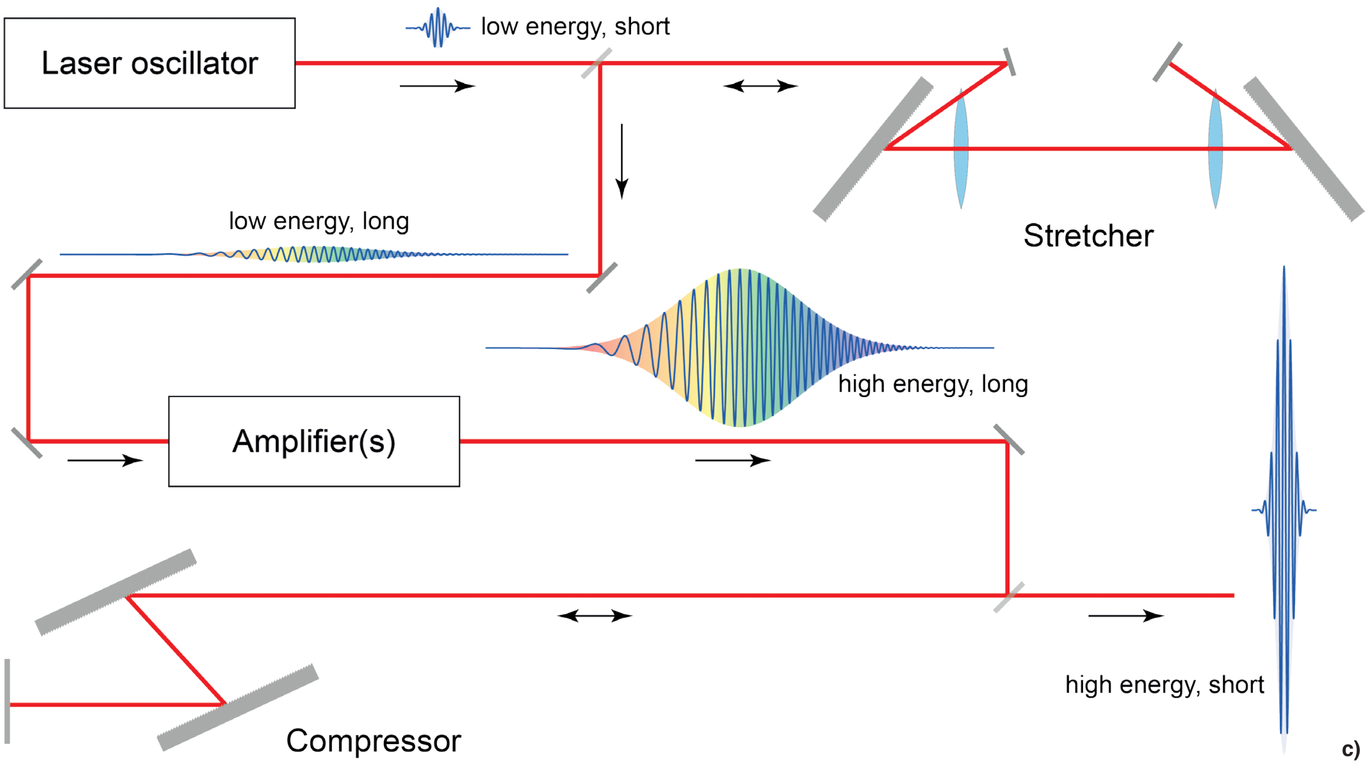


Figure 3: Chirped-pulse amplification using a grating-based stretcher and compressor. a) An arrangement of two diffraction gratings in double-pass imposes negative dispersion on an ultrashort optical pulse. Thereby it can compensate for a positive chirp of the initial pulse and recompress the wavepacket to short duration. b) By appropriately adding two identical lenses between the two gratings, the sign of the imposed dispersion can be flipped. Such an arrangement is typically used to stretch the pulse before amplification. c) A complete CPA system consists of a laser oscillator producing short, but low-energy pulses. After a stretcher, these pulses are amplified to substantial energies. To regain the short pulse duration and at the same time achieve a high peak power, the pulses are finally sent through a pulse compressor.
(Illustrations inspired by https://en.wikipedia.org/wiki/Chirped_pulse_amplification)



Larger laser systems, such as those being in their final phase of construction for the European Extreme-Light Infrastructure (ELI) will even reach 10 PW [8]. What compact means becomes apparent when comparing CPA systems with the highest-energy lasers that are still based on pre-CPA technology [9, 10]. The National Ignition Facility (NIF) at Lawrence-Livermore National Laboratory, for example, fills a large building (see [9] for a visualization) and fires approximately one laser shot per day. While the 10 PW laser of ELI Beamlines near Prague still fills a large room [11], it produces 200 times higher peak power and fires once every minute. It reaches this peak power and pulse repetition rate by compressing three orders of magnitude less energy into a pulse of only 130 fs instead of the several-nanosecond bursts used at NIF. In the following, a few examples of scientific applications made possible by CPA lasers are given.

At laser peak intensities on the order of 10^{14} W/cm^2, the electric field in the optical pulse becomes comparable to the inner-atomic binding forces and field-ionization may occur. Given the oscillating nature of the optical field, liberated electrons are further accelerated and may eventually return to their parent ion. Upon recollision with the parent ion, the electron may recombine with the ion and release excess energy in the form of a high-energy photon. With initial laser pulses in the near-infrared, the generated photons reach into the extreme ultraviolet spectral range. This laser-like, coherent extreme ultraviolet radiation is intrinsically concentrated into sub-femtosecond bursts [12, 13]. The described process lays the foundation of attosecond science. While the motion of atomic nuclei is frozen on sub-femtosecond scales, electrons are light enough to exhibit attosecond dynamics. Attosecond science has extensively studied the dynamics of ionization processes in atoms, molecules and from solids [14-17]. Recently, the field expanded towards studying electronic processes in condensed matter systems [see the authors article on p. 11 in this issue]. Attosecond science yields new insights on fundamental physical processes on atomic time and length scales that were previously inaccessible. Processes that were treated as instantaneous in the past have to be reconsidered. This entire research direction became possible only thanks to CPA, that enabled tabletop laser systems with tens of femtosecond pulses and the required focused intensities.

At the intensities presently used in attosecond science, the electrons still move non-relativistically in the focused laser beam. State-of-the-art laser systems, however, reach to almost ten orders of magnitude higher peak-intensity, which gives access to much more extreme regimes of laser-matter interaction. Systems with TW-level peak power are used for laser-driven particle acceleration. Given the extremely high electric-field gradients that can be reached with lasers, electrons can be accelerated to energies of many GeV over centimeter distances. At laser intensities on the order of 10^{18} W/cm^2 the oscillation energy of an electron in the electric field of a laser pulse becomes comparable with its rest mass, which defines the onset of relativistic interactions of free electrons with light. PW-class lasers open up access to the ultra-relativistic regime with intensities up to approximately 10^{23} W/cm^2. Such lasers enable research in extreme regimes of laser-matter interaction that are relevant for laboratory-based studies of astrophysical processes or for driving nuclear reactions. A holy grail of high-intensity laser physics is to reach the energy densities required for particle production from vacuum (i.e., separating a virtual positron-electron pair into real particles).

While these intense lasers allow studying fundamental physics directly, they can also be used to drive a wide range of secondary sources beyond the particle acceleration already mentioned above. Such lasers produce extreme ultraviolet radiation through high-harmonic generation, x-rays by heating a plasma or driving electrons through an undulator, and even intense gamma-rays beyond 10 MeV photon energy using mechanisms such as Compton back scattering. These sources are of immediate interest for a multitude of applications even if ultrafast time resolution is not needed.

5. Conclusion

As the examples above illustrate, CPA has allowed ultrafast lasers to reach into domains of physics well beyond ultrafast science itself. In addition to basic research, CPA had an impact for medical and industrial applications. High-power CPA fiber laser systems are now in use for precision micromachining in industry, but also for eye surgery in medicine. At the same time, high-intensity lasers offer the perspective for a wealth of future medical applications through their ability to produce beams of particles and high-energy photons as well as medically relevant radionuclides through laser-driven nuclear reactions.

The beauty of CPA lies in its simplicity and broad applicability. It has enabled the scaling of peak power and intensity by many orders of magnitude, which has given rise to entire new fields of research. The CPA method first demonstrated by Strickland and Mourou had a tremendous impact on science and beyond by virtually eliminating the significant technological constraints of the past. It is this impact that has been recognized by awarding one half of the 2018 Nobel Prize in Physics to Donna Strickland and Gérard Mourou. Even more than 30 years after it has been reported for the first time, CPA technology keeps pushing laser parameters towards new extremes, yielding access to unexplored regimes of light-matter interaction.

References

[1] C. J. Saraceno, F. Emaury, C. Schriber, M. Hoffmann, M. Golling, T. Südmeyer, and U. Keller, “Ultrafast thin disk laser with 80 μJ pulse energy and 242 W of average power,” Opt. Lett. **39**, 9-12 (2014).

[2] C. J. Saraceno, “Mode-locked thin-disk lasers and their potential application for high-power terahertz generation,” J. Opt. **20**, 044010 (2018).

[3] “The Nobel Prize in Physics 2018,” (NobelPrize.org, Nobel Media AB 2018), *https://www.nobelprize.org/prizes/physics/2018/summary/*.

[4] D. Strickland, and G. Mourou, “Compression of Amplified Chirped Optical Pulses,” Opt. Commun. **56**, 219 (1985).

[5] J. H. Marburger, “Self-focusing: theory,” Prog. Quant. Electr. **4**, 35-110 (1975).

[6] E. B. Treacy, “Optical Pulse Compression With Diffraction Gratings,” IEEE J. Quantum. Electron. **QE-5**, 454-458 (1969).

[7] O. E. Martinez, J. P. Gordon, and R. L. Fork, “Negative group-velocity dispersion using refraction,” J. Opt. Soc. Am. A **1**, 1003-1006 (1984).

[8] “Extreme Light Infrastructure, The ELI Project,” (ELI Delivery Consortium), *https://eli-laser.eu/the-eli-project/*.

[9] “National Ignition Facility - What is NIF?,” (Lawrence Livermore National Laboratory), https://lasers.llnl.gov/about/what-is-nif.

[10] “Le Laser Mégajoule,” (CEA), *http://www-lmj.cea.fr/en/lmj/index.htm*.

[11] “Laser 4 Aton: 10 PW, 2 kJ,” (ELI Beamlines), *https://www.eli-beams.eu/en/facility/lasers/laser-4-10-pw-2-kj/*.

[12] L. Gallmann, C. Cirelli, and U. Keller, “Attosecond Science: Recent Highlights and Future Trends,” Annu. Rev. Phys. Chem. **63**, 449-469 (2012).

[13] T. Gaumnitz, A. Jain, Y. Pertot, M. Huppert, I. Jordan, F. Ardana-Lamas, and H. J. Wörner, “Streaking of 43-attosecond soft-X-ray pulses generated by a passively CEP-stable mid-infrared driver,” Opt. Express **25**, 27506-27518 (2017).

[14] M. Nisoli, P. Decleva, F. Calegari, A. Palacios, and F. Martín, “Attosecond Electron Dynamics in Molecules,” Chem. Rev. **117**, 10760-10825 (2017).

[15] A. S. Landsman, M. Weger, J. Maurer, R. Boge, A. Ludwig, S. Heuser, C. Cirelli, L. Gallmann, and U. Keller, “Ultrafast resolution of tunneling delay time,” Optica **1**, 343-349 (2014).

[16] J. Vos, L. Cattaneo, S. Patchkovskii, T. Zimmermann, C. Cirelli, M. Lucchini, A. Kheifets, A. S. Landsman, and U. Keller, “Orientation-dependent stereo Wigner time delay and electron localization in a small molecule,” Science **360**, 1326-1330 (2018).

[17] L. Kasmi, M. Lucchini, L. Castiglioni, P. Kliuiev, J. Osterwalder, M. Hengsberger, L. Gallmann, P. Krüger, and U. Keller, “Effective mass effect in attosecond electron transport,” Optica **4**, 1492-1497 (2017).