Audrius Dubietis Arnaud Couairon

# Ultrafast Supercontinuum Generation in Transparent Solid-State Media



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# Ultrafast Supercontinuum Generation in Transparent Solid-State Media



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#### Preface

Nonlinear propagation of intense femtosecond laser pulses in bulk transparent media produces a wealth of physical effects, whose combined action leads to a specific propagation regime, termed femtosecond filamentation, which in turn produces dramatic broadening of the pulse spectrum, termed supercontinuum generation. Due to low cost, compactness, efficiency, robustness, and virtual alignment insensitivity, femtosecond supercontinuum represents a unique and versatile source of coherent ultrabroadband radiation, whose wavelength range spans a considerable part of the optical spectrum. Altogether, these outstanding properties make femtosecond supercontinuum highly suitable for diverse applications in time-resolved spectroscopy, photonics, femtosecond technology, and lightwave engineering. During recent years, this research field has reached a high level of maturity, both in understanding of the underlying physics and in achievement of exciting practical results. In this book, we present the underlying physical picture and overview the state of the art of femtosecond supercontinuum generation in various transparent solid-state media, ranging from wide bandgap dielectrics to semiconductor materials and in various parts of the optical spectrum, from the ultraviolet to the mid-infrared. A particular emphasis is given to the most recent experimental developments: multioctave supercontinuum generation with pumping in the mid-infrared spectral range, spectral control, power and energy scaling of broadband radiation, and the development of simple, flexible, and robust pulse compression techniques, which deliver few optical cycle pulses and which could be readily implemented in a variety of modern ultrafast laser systems. The expected audience includes graduate students, professionals, and scientists working in business and academia, in the field of laser-matter interactions and ultrafast nonlinear optics.

Vilnius, Lithuania Paris, France January 2019 Audrius Dubietis Arnaud Couairon

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#### Chapter 1 Introduction



Supercontinuum (SC) generation is one of the most spectacular and visually perceptible effects produced by the nonlinear propagation of intense ultrashort laser pulse in a transparent medium. The discovery of supercontinuum generation in a bulk solid-state medium dates back to the early years of nonlinear optics, when Alfano and Shapiro reported on white light generation, produced by self-focusing of powerful picosecond pulses in a borosilicate glass sample [1]. The discovery was immediately followed by observations of spectral broadening in various crystals and glasses, confirming the universal nature of the phenomenon [2]. (See also [3] for a complete historical account on the early developments of SC generation in various optical media.)

SC generation in bulk media constitutes a compact, efficient, low cost, highly robust, and virtually alignment-insensitive technique for the generation of coherent ultrabroadband radiation at various parts of the optical spectrum [4]. The physical picture of SC generation in transparent bulk media is unveiled in the framework of femtosecond filamentation, which provides a universal scenario of nonlinear propagation and spectral broadening of intense femtosecond laser pulses in bulk solids, liquids, and gases [5-9]. SC generation in bulk media appears to be a complex process that involves an intricate coupling between spatial and temporal effects: diffraction, group velocity dispersion, self-focusing, self-phase modulation, and multiphoton absorption or ionization. In the space domain, the interplay of these effects leads to the formation of a narrow light channel, termed a "light filament" that is able to propagate over extended distances much larger than the typical diffraction length and which leaves a narrow luminous plasma trail in its wake. In the time domain, the pulse undergoes dramatic transformations: pulse splitting or compression, pulsefront steepening, and generation of optical shocks. These transformations altogether produce a broadband, spatially, and temporally coherent emission with a low angular divergence (supercontinuum), which is accompanied by the generation of colored conical emission that is emitted at different angles with respect to the propagation

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axis, forming a beautiful array of concentric colored rings. Therefore, SC generation in bulk is markedly different from SC generation in optical fibers, where the propagation dynamics of the optical pulse is essentially one-dimensional and spectral broadening arises from soliton generation and fission due to the interplay between self-phase modulation and material dispersion [10].

SC generation with femtosecond laser pulses was first reported in 1983 [11], long before the phenomenon of femtosecond filamentation was discovered [12]. In that pioneering experiment, Fork and co-authors observed spectral broadening from the deep ultraviolet to the near infrared by focusing intense 80-fs pulses at 627 nm from the dye laser into an ethylene glycol jet [11]. Apart from large-scale spectral broadening, the authors underlined an improvement of pulse-to-pulse reproducibility and spatial uniformity of the beam, which resulted from the short duration of the input pulse; see also [13] and references therein for an account of SC generation in various solid-state and liquid media using femtosecond dye lasers.

A major breakthrough in femtosecond solid-state laser technology was inspired by the groundbreaking invention of chirped pulse amplification (CPA) technique by D. Strickland and G. Mourou [14], which was awarded the Nobel Prize in Physics in 2018. The CPA concept solved the long-standing problem of safe and efficient amplification of ultrashort optical pulses without the onset of optical damage of the amplifier material and other optical components, enabling a tremendous leap in the peak power and intensity of laser pulses. Shortly after that, the discovery of Kerr lens mode locking has led to the invention of femtosecond Ti:sapphire laser oscillator in 1991 [15]. Demonstration of the CPA technique-based regenerative amplification of the Ti:sapphire oscillator pulses, constituted a significant breakthrough in solidstate laser technology and marked a new era in femtosecond SC generation [16]. The amplified Ti:sapphire lasers outperformed then widely spread femtosecond dye lasers in all essential parameters of operation, setting a new standard for the entire femtosecond solid-state laser technology [17].

The commercial availability of novel femtosecond laser sources as combined with a growing practical knowledge of femtosecond SC generation in transparent condensed media [18–20], boosted the development of femtosecond optical parametric amplifiers (OPAs). In that regard, the SC radiation was recognized as an indispensable seeding source for these devices, which produced femtosecond pulses with unprecedented wavelength tunability well exceeding the tuning range afforded by conventional laser sources [21]. Broad spectral bandwidth and high temporal coherence of the SC radiation allowed compressibility of the pulses down to the Fourier transform limit, contributing to the invention of ultrabroadband noncollinear optical parametric amplifiers, the so-called NOPAs [22], which currently deliver few optical cycle pulses at various parts of the optical spectrum, ranging from the visible to the mid-infrared [23].

The advances in the optical parametric amplification techniques fostered exciting developments in the optical parametric chirped amplification (OPCPA). The general idea of the OPCPA was to replace the laser amplifier by the OPA and was originally proposed as an alternative to existing laser amplifiers in 1992 [24]. At present, OPCPA is deservedly regarded as an important offspring of the CPA technique, since

as compared to the laser amplifier, the OPA offers the advantages of very high gain, broad amplification bandwidth, great wavelength flexibility, low thermal effects and superior intensity contrast of the amplified pulses, offering unique possibilities for the amplification of ultrashort laser pulses [25–27]. Interestingly, the first demonstrations of both, CPA and OPCPA, used an optical fiber to broaden the pulse spectrum before the amplification and compression stages. In that regard, SC generation in condensed bulk media offered a number of advantages due to its robustness and compactness, enabling to elaborate novel and compact architectures of tabletop SCseeded OPCPA systems [28]. In particular, a considerable effort is currently directed to the development of the SC-seeded OPCPA systems that deliver intense few optical cycle pulses in the mid-infrared spectral region, see, e.g., [29], which is out of the grasp for existing mid-infrared solid-state lasers and laser amplifiers, see, e.g., [30].

These developments in turn facilitated experimental studies of SC generation in the region of anomalous group velocity dispersion (GVD) of dielectric solid-state media, yielding ultrabroadband, multioctave SC with unprecedented wavelength coverage, see, e.g., [31–33]. Moreover, using long-wavelength ultrashort pulses, SC generation was made possible in various highly nonlinear materials, such as narrow bandgap dielectric crystals, soft glasses, and semiconductors to produce octave-spanning SC spectra extending into far infrared. From a future perspective, SC generation represents one of the fundamental building blocks of the emerging third-generation femtosecond technology, which foresees boosting the peak and average powers of few optical cycle pulses simultaneously to the multiterawatt and hundreds of watts range, respectively, thereby paving the way for the generation of powerful sub-cycle pulses with full control over the generated light waves [34]. Finally, within the past decade, the term "supercontinuum generation" has been extended well beyond the optical range, to include high-order and nonperturbative nonlinear optical processes, such as high harmonic generation in the vacuum ultraviolet and X-ray ranges [35].

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