Cover illustration: Shows the supercontinuum generation of intensity versus wavelength for 1 mm of carbon tetrachloride liquid excited by a 120-fs, 625-nm laser pulse. Photo by Robert R. Alfano, A. Katz, and P.P. Ho.

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To my father, Alfonso L. Alfano
and my father-in-law, Samuel J. Resnick
whose advice I deeply miss.
Preface to the Second Edition

The “supercontinuum” (SC) has become one of the hottest topics to study in optical and photonic sciences since the first book on the supercontinuum was published, entitled *The Supercontinuum Laser Source*, by Springer in 1989. That book, now becoming Part I in this second edition, reviewed the progress achieved on the experimental and theoretical understanding of the ultrafast nonlinear and linear processes responsible for the supercontinuum generation and related applications occurring over 20 years since its discovery by Robert R. Alfano and Stanley Shapiro in 1969.

There is a great need for a sequel part covering the recent worldwide surge of research activity on the supercontinuum phenomena and the numerous technological applications that have occurred over the past 15 years. This void will partly be covered in this new rejuvenated second edition, called Part II, by an overview of the recent advances with an updated compendium of references on the various breakthroughs to understand the supercontinuum and its new diverse applications.

The supercontinuum is the generation of intense ultrafast broadband “white-light” pulses spanning the ultraviolet to the near infrared that arises from the nonlinear interaction and propagation of ultrafast pulses focused into a transparent material. The supercontinuum can be generated in different states of matter—condensed media (liquids and solids) and gases. The supercontinuum is one of the most dramatic and elegant effects in optical physics. The conversion of one color to white-light is a startling result. This is multicolored light with many of the same desirable properties as conventional laser light: intense, collimated, and coherent. The supercontinuum has a beam divergence as good as that of the input pump laser pulse. Moreover, the coherence length of the supercontinuum is comparable with that of an incoherent white-light source from a light bulb. The interference pattern measured for the supercontinuum from a pair of filaments in water shows a constant phase relationship between the supercontinuum produced by each filament. There is a constant phase relationship between the pump laser pulse and its supercontinuum. The white-light supercontinuum is an ideal tunable ultrafast white-light laser source. Supercontinuum has overtaken the study of
other nonlinear optical effects such as second harmonic generation (SHG) and two-photon absorption for usefulness in a number of diverse applications. The supercontinuum field is still active after 36 years, and is today finding new and novel uses.

Various processes are involved in the supercontinuum generation. Whenever an intense ultrashort laser pulse propagates through a medium, it changes the refractive index from the distortion of the atomic and molecular configuration, which in turn changes the phase, amplitude, and frequency of the incident pulse. The phase change and amplitude change can cause a frequency sweep of the carrier wave within the pulse envelope and can alter the envelope and spatial distribution (self-focusing). There are various mechanisms responsible for the index of refraction change in material with intensity. The frequency broadening mechanisms are electronic cloud distortion, reorientational, librations, vibrational, and molecular redistribution, to name the major ones. The operation of these mechanisms depends on its relaxation time relevant to the laser pulse duration. The relaxation times associated with electronic distribution is of the order of Bohr orbit time $\sim 150$ as; reorientation time is $\sim 1$ ps; rocking and libration response about the field is $\sim 1$ ps; vibrational dephasing is $\sim 0.1$ ps; and molecular motion is $\sim 1$ ps. Most of these mechanisms are involved in the supercontinuum generation with 100 fs to ps laser pulses.

Soon after the supercontinuum discovery in 1969, it initially found applications in time-resolved pump-supercontinuum probe absorption and excitation spectroscopy to study the fundamental picosecond ($10^{-12}$ s) and femtosecond ($10^{-15}$ s) processes that occur in biology, chemistry, and solid-state physics. Briefly, in biology, the primary events in photosynthesis and vision were explored; in chemistry, a better understanding of the basic chemical dynamical steps in reactions and nonradiative processes in photoexcited chemicals was achieved; and in solid-state physics, the underlying kinetics of how elementary excitations behave and relax, such as optical phonons, polaritons, excitons, and carriers (electrons and holes) dynamics among the intervals and intravalley of semiconductors, were unraveled.

With the advent of microstructure fibers, there has been a rebirth of the supercontinuum field in the type of applications in which the supercontinuum can play a decisive role. These applications include frequency clocks, phase stabilization and control, timing, optical coherence tomography (OCT), ultrashort pulse compression, optical communication, broad spectrum LIDAR, atmospheric science, lighting control, attosecond ($10^{-18}$ s) pulse generation, and coherence control.

Over the past several years, supercontinuum generation in microstructure photonic crystal fibers by ultrashort pulse propagation has become a subject of great interest worldwide. The main reasons are the low pulse energies required to generate the supercontinuum; its coherences and high brightness makes the continuum an ideal white-light source for diverse applications; and the effects of zero dispersion and anomalous dispersion regions has resulted
in higher-order solutions generation, pulse compression, and an ultrabroadband continuum exceeding 1000 nm, extending from the ultraviolet to the infrared spectral regions.

In microstructural fibers, when pump wavelength lies in an anomalous dispersion region, it is the solitons that initiate the formation of the continuum. In a normal dispersion region, self-phase modulation is the process that initiates the continuum generation. The combination of four-wave mixing and Raman processes extends the spectral width of the continuum. In that regard, the pulse duration of an ultrafast laser determines the operational mechanisms—for 10 fs to 1000 fs laser pulses, self-phase modulation and soliton generation dominates; and for pulses >30 ps, stimulated Raman and four-wave mixing play a major role in extending the spectra. Of course, the pump wavelength location, relative to the zero dispersion wavelength and the anomalous dispersion region, plays a role in the active mechanism and coherence region of the supercontinuum. The supercontinuum spectra can span more than a two-optical octave bandwidth spread from 380 nm to 1600 nm using 200 fs pulses with energy in the tens of nanojoules. The span over an octave (i.e., 450 nm to 900 nm) is important in controlling the phase of the carrier wave inside the pulse envelope of a mode-locked pulse train. Using the $f$ and $2f$ waves in the supercontinuum, the carrier-envelope offset (CEO) phase can be detected using heterodyne beating between the high-frequency end of the supercontinuum with the doubled low end frequency of the supercontinuum in an interferometer. These phase-controlling effects are important for maintaining the accuracy of frequency combs for clocking and timing in metrology, high-intensity atomic studies, and attosecond pulse generation.

The increasing worldwide demand for large-capacity optical communication systems needs to incorporate both the wavelength and time. The ultrabroad bandwidth and ultrashort pulses of the supercontinuum may be the enabling technology to produce a cost-effective superdense wavelength division multiplexing (>1000 λ) and time multiplexing for the future Terabits/s to Pentabits/s communication systems and networks. The supercontinuum is an effective way to obtain numerous wavelength channels because it easily generates more than 1000 optical longitudinal modes while maintaining their coherency.

The propagation of ultrahigh power femtosecond pulses ~100 GW (10 mJ at 100 fs) in “air” creates the supercontinuum from the collapse of the beam by self-focusing into self-guided small-size filaments. These filament tracks in air are more or less stable over long distances of a few kilometers due to the balance between self-focusing by the nonlinear index of refraction ($n_2$) and the defocusing by the ionized plasma formation via multiphoton ionization. The supercontinuum in air can be used to monitor the amount of trace gases and biological agents in aerosols in the backscattering detection geometry for LIDAR applications. Furthermore, remote air ionization in the atmosphere by the intense femtosecond pulses in the filaments plasma (uses the supercontinuum as the onset marker) has the potential to trigger, control, and
guide lightning from one point to another and possibly even induce condensation by seeding clouds to make rain. This approach may be able to secure and protect airports and power stations from lightning and may be used to collect and store energy from lightning. Moreover, creating an ionized filament track in a desirable region may be used to confuse and redirect the pathway of incoming missiles for defense.

This new second edition will consist of two parts. The major portion (Part I) of the new book will be the reprinting of Chapters 1 to 10 from the first edition. These chapters lay down the understanding and foundation of the birth of the supercontinuum field. They go over the salient experimental and theoretical concepts in the research works produced up to 1989. The second part of this new second edition includes a new chapter (Chapter 11) highlighting the supercontinuum coherence and 10 additional chapters (Chapters 12 to 21) listing updated references of papers on the recent advances made in our understanding and applications of supercontinuum. These papers will be referenced and arranged within a topical group where a brief overview of the key features of these papers within a topic will be presented.

The following are the selected topics to be highlighted in the new Chapters 12 to 21 of updated references:

- Supercontinuum generation in materials (solids, liquids, gases, air).
- Supercontinuum generation in microstructure fibers.
- Supercontinuum in wavelength division multiplex telecommunication.
- Femtosecond pump—supercontinuum probe for applications in semiconductors, biology, and chemistry.
- Supercontinuum in optical coherence tomography.
- Supercontinuum in femtosecond carrier-envelope phase stabilization.
- Supercontinuum in ultrafast pulse compression.
- Supercontinuum in time and frequency metrology.
- Supercontinuum in atmospheric science.
- Coherence of the supercontinuum.

Special thanks to Ms. Lauren Gohara and Dr. Kestutis Sutkus for their assistance in the production of the second edition.

New York, New York

ROBERT R. ALFANO
Preface to the First Edition

This book deals with both ultrafast laser and nonlinear optics technologies. Over the past two decades, we have seen dramatic advances in the generation of ultrafast laser pulses and their applications to the study of phenomena in a variety of fields. It is now commonplace to produce picosecond ($10^{-12}$ s) pulses. New developments have extended this technology into the femtosecond ($10^{-15}$ s) time region. Soon pulses consisting of just a single cycle will be produced (i.e., 2 fs at 600 nm). These ultrafast pulses permit novel investigations to study phenomena in many disciplines. Sophisticated techniques based on these laser pulses have given rise to instruments with extremely high temporal resolution. Ultrafast laser technology offers the possibility of studying and discovering key processes unresolved in the past. A new era of time-resolved spectroscopy has emerged, with pulses so fast that one can now study the nonequilibrium states of matter, test quantum and light models, and explore new frontiers in science and technology. Ultrashort light pulses are a potential signal source in future high-bit-rate optical fiber communication systems. The shorter the pulses, the more can be packed into a given time interval and the higher is the data transmission rate for the tremendous bandwidth capacity of optical fiber transmission.

Nonlinear optics is an important field of science and engineering because it can generate, transmit, and control the spectrum of laser pulses in solids, liquids, gases, and fibers. One of the most important ultrafast nonlinear optical processes is the supercontinuum generation—the production of intense ultrafast broadband “white-light” pulses—that is the subject of this book.

The first study on the mechanism and generation of ultrafast supercontinuum dates back over 19 years to 1969, when Alfano and Shapiro observed the first “white” picosecond pulse continuum in liquids and solids. Spectra extended over $\sim 6000$ cm$^{-1}$ in the visible and infrared wavelength region. They attributed the large spectral broadening of ultrafast pulses to self-phase modulation (SPM) arising from an electronic mechanism and laid down the formulation of the supercontinuum generation model. Over the years, the improvement of mode-locked lasers led to the production of wider super-
continua in the visible, ultraviolet, and infrared wavelength regions using various materials.

The supercontinuum arises from the propagation of intense picosecond or shorter laser pulses through condensed or gaseous media. Various processes are responsible for continuum generation. These are called self-, induced-, and cross-phase modulations and four-photon parametric generation. Whenever an intense laser pulse propagates through a medium, it changes the refractive index, which in turn changes the phase, amplitude, and frequency of the pulse. However, when two laser pulses of different wavelengths propagate simultaneously in a condensed medium, coupled interactions (cross-phase modulation and gain) occur through the nonlinear susceptibility coefficients. These coupled interactions of two different wavelengths can introduce phase modulation, amplitude modulation, and spectral broadening in each pulse due to the other pulse using cross-effects.

An alternative coherent light source to the free electron laser, the supercontinuum laser source, can be wavelength selected and coded simultaneously over wide spectral ranges (up to 10,000 cm$^{-1}$) in the ultraviolet, visible, and infrared regions at high repetition rates, gigawatt output peak powers, and femtosecond pulse durations.

Ultrafast supercontinuum pulses have been used for time-resolved absorption spectroscopy and material characterization. Supercontinuum generation is a key step for the pulse compression technique, which is used to produce the shortest optical pulses. Future applications include signal processing, three-dimensional imaging, ranging, atmospheric remote sensing, and medical diagnosis.

Thus far, a great deal of information on supercontinuum technology has been obtained and has enhanced our understanding of how intense optical pulses propagate in materials. These developments are most often found in original research contributions and in review articles scattered in journals. Textbooks do not cover these subjects in great detail. There is a need for a book that covers the various aspects of ultrafast supercontinuum phenomena and technology.

This book reviews present and past progress on the experimental and theoretical understanding of ultrafast nonlinear processes responsible for supercontinuum generation and related effects such as pulse compression and ultrashort pulse generation on a picosecond and femtosecond time scale. The content of the chapters in the book is a mixture of both theoretical and experimental material. Overviews of the important breakthroughs and developments in the understanding of supercontinuum during the past 20 years are presented. The book is organized into 10 chapters.

Summarizing the highlights of the 10 chapters of the book:

In Chapter 1, Shen and Yang focus on the theoretical models and mechanisms behind supercontinuum generation arising mainly from self-phase modulation.
In Chapter 2, Wang, Ho, and Alfano review the experiments leading to the supercontinuum generation in condensed matter over the past 20 years. In Chapter 3, Agrawal discusses the effects of dispersion on ultrafast light pulse propagation and supercontinuum generation in fibers. In Chapter 4, Baldeck, Ho, and Alfano cover the latest experimental observations and applications of the cross-interactions in the frequency, time, and space domains of strong pulses on weak pulses. In Chapter 5, Manassah reviews the theoretical models giving rise to many phenomena from self-phase and induced modulations. In Chapter 6, Suydam highlights the effect of self-steepening of pulse profile on continuum generation. In Chapter 7, Corkum and Rolland review the work on supercontinuum and self-focusing in gaseous media. In Chapter 8, Glownia, Misewich, and Sorokin utilize the supercontinuum produced in gases for ultrafast spectroscopy in chemistry. In Chapter 9, Dorsinville, Ho, Manassah, and Alfano cover the present and speculate on the possible future applications of the supercontinuum in various fields. In Chapter 10, Johnson and Shank discuss pulse compression from the picosecond to femtosecond time domain using the continuum and optical dispersive effects of gratings, prisms, and materials.

The reader will find that these chapters review the basic principles, contain surveys of research results, and present the current thinking of experts in the supercontinuum field. The volume should be a useful source book and give young and seasoned scientists, engineers, and graduate students an opportunity to find the most necessary and relevant material on supercontinuum technology in one location.

I hope these efforts will stimulate future research on understanding the physics behind supercontinuum technology and exploring new applications.

I wish to thank all the expert contributors for their cooperation in this endeavor. Most thought it would not be completed. Special thanks goes to Mrs. Megan Gibbs for her administrative and secretarial assistance. I gratefully acknowledge T. Hiruma for his continued support. I pay particular tribute to my friend Stan Shapiro, who missed seeing the outgrowth of our first work in this field 20 years ago.

New York, New York

ROBERT R. ALFANO
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