The Supercontinuum Laser Source
Second Edition
Robert R. Alfano
Editor

The Supercontinuum Laser Source
Fundamentals with Updated References
Second Edition

With 259 Illustrations

Springer
To my father, Alfonso L. Alfano
and my father-in-law, Samuel J. Resnick
whose advice I deeply miss.
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 \( \sim 150 \text{ as} \); reorientation time is \( \sim 1 \text{ ps} \); rocking and libration response about the field is \( \sim 1 \text{ ps} \); vibrational dephasing is \( \sim 0.1 \text{ ps} \); and molecular motion is \( \sim 1 \text{ ps} \). 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 (\( 10^{-12} \text{ s} \)) and femtosecond (\( 10^{-15} \text{ s} \)) 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 inter-valleys 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 (\( 10^{-18} \text{ s} \)) 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 $\sim 6000 \text{ cm}^{-1}$ 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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Part I
Fundamentals
1. Introduction

Self-phase modulation refers to the phenomenon in which a laser beam propagating in a medium interacts with the medium and imposes a phase modulation on itself. It is one of those very fascinating effects discovered in the early days of nonlinear optics (Bloembergen and Lallemand, 1966; Brewer, 1967; Cheung et al., 1968; Lallemand, 1966; Jones and Stoicheff, 1964; Shimizu, 1967; Stoicheff, 1963). The physical origin of the phenomenon lies in the fact that the strong field of a laser beam is capable of inducing an appreciable intensity-dependent refractive index change in the medium. The medium then reacts back and inflicts a phase change on the incoming wave, resulting in self-phase modulation (SPM). Since a laser beam has a finite cross section, and hence a transverse intensity profile, SPM on the beam should have a transverse spatial dependence, equivalent to a distortion of the wave front. Consequently, the beam will appear to have self-diffracted. Such a self-diffraction action, resulting from SPM in space, is responsible for the well-known nonlinear optical phenomena of self-focusing and self-defocusing (Marburger, 1975; Shen, 1975). It can give rise to a multiple ring structure in the diffracted beam if the SPM is sufficiently strong (Durbin et al., 1981; Santamato and Shen, 1984). In the case of a pulsed laser input, the temporal variation of the laser intensity leads to an SPM in time. Since the time derivative of the phase of a wave is simply the angular frequency of the wave, SPM also appears as a frequency modulation. Thus, the output beam appears with a self-induced spectral broadening (Cheung et al., 1968; Gustafson et al., 1969; Shimizu, 1967).

In this chapter we are concerned mainly with SPM that leads to spectral broadening (Bloembergen and Lallemand, 1966; Brewer, 1967; Cheung et al., 1968; Lallemand, 1966; Jones and Stoicheff, 1964; Shimizu, 1967; Stoicheff, 1963). For large spectral broadening, we need a strong SPM in time (i.e., a large time derivative in the phase change). This obviously favors the use of short laser pulses. Consider, for example, a phase change of $6\pi$ occurring in $10^{-12}$ s. Such a phase modulation would yield a spectral broadening of
In practice, with sufficiently intense femtosecond laser pulses, a spectral broadening of 20,000 cm$^{-1}$ is readily achievable by SPM in a condensed medium, which is essentially a white continuum (Alfano and Shapiro, 1970). The pulse duration of any frequency component (uncertainty limited) in the continuum is not very different from that of the input pulse (Topp and Rentzepis, 1971). This spectrally super-broadened output from SPM therefore provides a much needed light source in ultrafast spectroscopic studies—tunable femtosecond light pulses (Busch et al., 1973; Alfano and Shapiro, 1971). If the SPM and hence the frequency sweep in time on a laser pulse are known, then it is possible to send the pulse through a properly designed dispersive delay system to compensate the phase modulation and generate a compressed pulse with little phase modulation (Treacy, 1968, 1969). Such a scheme has been employed to produce the shortest light pulses ever known (Fork et al., 1987; Ippen and Shank, 1975; Nakatsuka and Grischkowsky, 1981; Nakatsuka et al., 1981; Nikolaus and Grischkowsky, 1983a, 1983b).

Self-phase modulation was first proposed by Shimizu (1967) to explain the observed spectrally broadened output from self-focusing of a $Q$-switched laser pulse in liquids with large optical Kerr constants (Bloembergen and Lallemand, 1966; Brewer, 1967; Cheung et al., 1968; Jones and Stoicheff, 1964; Lallemand, 1966; Shimizu, 1967; Stoicheff, 1963). In this case, the spectral broadening is generally of the order of a hundred reciprocal centimeters. Alfano and Shapiro (1970) showed that with picosecond laser pulses, it is possible to generate by SPM a spectrally broadened output extending over 10,000 cm$^{-1}$ in almost any transparent condensed medium. Self-focusing is believed to have played an important role in the SPM process in the latter case. In order to study the pure SPM process, one would like to keep the beam cross section constant over the entire propagation distance in the medium. This can be achieved in an optical fiber since the beam cross section of a guided wave should be constant and the self-focusing effect is often negligible. Stolin and Lin (1978) found that indeed the observed spectral broadening of a laser pulse propagating through a long fiber can be well explained by the simple SPM theory. Utilizing a well-defined SPM from an optical fiber, Grischkowsky and co-workers were then able to design a pulse compression system that could compress a laser pulse to a few hundredths of its original width (Nakatsuka and Grischkowsky, 1981; Nakatsuka et al., 1981; Nikolaus and Grischkowsky, 1983a, 1983b). With femtosecond laser pulses, a strong SPM on the pulses could be generated by simply passing the pulses through a thin film. In this case, the beam cross section is practically unchanged throughout the film, and one could again expect a pure SPM process. Fork et al. (1983) observed the generation of a white continuum by focusing an 80-fs pulse to an intensity of $\sim 10^{14}$ W/cm$^2$ on a 500-μm ethylene glycol film. Their results can be understood by SPM along with the self-steepening effect (Manassah et al., 1985, 1986; Yang and Shen, 1984).
Among other experiments, Corkum et al. (1985) demonstrated that SPM and spectral broadening can also occur in a medium with infrared laser pulses. More recently, Corkum et al. (1986) and Glownia et al. (1986) have independently shown that with femtosecond pulses it is even possible to generate a white continuum in gas media.

The phase modulation induced by one laser pulse can also be transferred to another pulse at a different wavelength via the induced refractive index change in a medium. A number of such experiments have been carried out by Alfano and co-workers (1986, 1987). Quantitative experiments on spectral superbroadening are generally difficult. Self-focusing often complicates the observation. Even without self-focusing, quantitative measurements of a spectrum that is generated via a nonlinear effect by a high-power laser pulse and extends from infrared to ultraviolet are not easy. Laser fluctuations could lead to large variations in the output.

The simple theory of SPM considering only the lower-order effect is quite straightforward (Gustafson et al., 1969; Shimizu, 1967). Even the more rigorous theory including the higher-order contribution is not difficult to grasp as long as the dispersive effect can be neglected (Manassah et al., 1985, 1986; Yang and Shen, 1984). Dispersion in the material response, however, could be important in SPM, and resonances in the medium would introduce pronounced resonant structure in the broadened spectrum. The SPM theory with dispersion is generally very complex; one often needs to resort to a numerical solution (Fisch and Bischel, 1975; Fisher et al., 1983). It is possible to describe the spectral broadening phenomenon as resulting from a parametric wave mixing process (in the pump depletion limit) (Bloembergen and Lallemand, 1966; Lallemand, 1966; Penzkofer, 1974; Penzkofer et al., 1973, 1975). In fact, in the studies of spectral broadening with femtosecond pulses, four-wave parametric generation of new frequency components in the phase-matched directions away from the main beam can be observed together with the spectrally broadened main beam. Unfortunately, a quantitative estimate of spectral broadening due to the parametric process is not easy. In the presence of self-focusing, more complication arises. Intermixing of SPM in space and SPM in time makes even numerical solution very difficult to manage, especially since a complete quantitative description of self-focusing is not yet available. No such attempt has ever been reported. Therefore, at present, we can only be satisfied with a qualitative, or at most a semiquantitative, description of the phenomenon (Marburger, 1975; Shen, 1975).

This chapter reviews the theory of SPM and associated spectral broadening. In the following section, we first discuss briefly the various physical mechanisms that can give rise to laser-induced refractive index changes responsible for SPM. Then in Section 3 we present the simple physical picture and theory of SPM and the associated spectral broadening. SPM in space is considered only briefly. Section 4 deals with a more rigorous theory of SPM that takes into account the higher-order effects of the induced refractive index change. Finally, in Section 5, a qualitative picture of how self-focusing can influence
2. Optical-Field-Induced Refractive Indices

The material response to an applied laser field is often nonlinear. An explicit expression for the response is not readily available in general. Unless specified otherwise, we consider here only the case where the perturbative expansion in terms of the applied field is valid and the nonlocal response can be neglected. We can then express the induced polarization in a medium as (Shen, 1984)

\[ \mathbf{P}(t) = \mathbf{P}^{(1)}(t) + \mathbf{P}^{(2)}(t) + \mathbf{P}^{(3)}(t) + \ldots, \]

\[ \mathbf{P}^{(1)}(t) = \int \chi^{(1)}(t-t') \cdot \mathbf{E}(t')dt', \]

\[ = \int \chi^{(1)}(\omega) \cdot \mathbf{E}(\omega)d\omega, \]

\[ \mathbf{P}^{(n)}(t) = \int \chi^{(n)}(t-t_1, \ldots, t-t_n) : \mathbf{E}(t_1) \ldots \mathbf{E}(t_n)dt_1 \ldots dt_n \]

\[ = \int \chi^{(n)}(\omega = \omega_1 + \omega_2 + \ldots + \omega_n) : \mathbf{E}(\omega_1) \ldots \mathbf{E}(\omega_n)d\omega_1 \ldots d\omega_n, \]  

(1)

where the applied field is

\[ \mathbf{E}(t) = \int \mathbf{E}(\omega)d\omega \quad \text{with} \quad \mathbf{E}(\omega) \propto \exp(-i\omega t) \]  

(2)

and the \( n \)th-order susceptibility is

\[ \chi^{(n)}(t-t_1, \ldots, t-t_n) = \int \chi^{(n)}(\omega = \omega_1 + \ldots + \omega_n) \exp[i\omega_1(t-t_1) + \ldots + i\omega_n(t-t_n)]d\omega_1 \ldots d\omega_n. \]  

(3)

We note that, strictly speaking, only for a set of monochromatic applied fields can we write

\[ \mathbf{P}^{(n)} = \chi^{(n)}(\omega = \omega_1 + \ldots + \omega_n) : \mathbf{E}(\omega_1) \ldots E(\omega_n). \]  

(4)

In the case of instantaneous response (corresponding to a dispersionless medium), we have

\[ \mathbf{P}^{(n)} = \chi^{(n)} : [E(t)]^n. \]  

(5)

Here, we are interested in the third-order nonlinearity that gives rise to the induced refractive index change. We consider only the self-induced refractive index change; extension to the cross-field-induced change should be straightforward. Thus we assume a pulsed quasi-monochromatic field \( \mathbf{E}_\text{in}(t) = 4 \  \text{Y.R. Shen and G.-Z. Yang} \)
The third-order nonlinear polarization in a medium, in general, takes the form

$$\mathbf{P}_{\omega}^{(3)}(t) = \int \Delta \chi(t - t') \cdot \mathbf{E}_{\omega}(t') dt'$$

with $$\Delta \chi(t - t') = \chi^{(3)}(t - t', t - t'', t - t''') : \mathbf{E}_{\omega}(t') \cdot \mathbf{E}_{\omega}(t'') dt'' dt'''$$. If the optical field is sufficiently far from resonances that the transverse excitations are all virtual and can be considered as instantaneous, we can write

$$\mathbf{P}_{\omega}^{(3)}(t) = \Delta \chi(t) \cdot \mathbf{E}_{\omega}(t),$$

$$\Delta \chi(t) = \int \chi^{(3)}(t - t') : |\mathbf{E}_{\omega}(t')|^2 dt'.$$

In the dispersionless limit, the latter becomes

$$\Delta \chi(t) = \chi^{(3)} : |\mathbf{E}_{\omega}(t)|^2.$$

Equation (8) is a good approximation when the dispersion of $$\Delta \chi$$ is negligible within the bandwidth of the field. The optical-field-induced refractive index can be defined as

$$\Delta n = \left(2\pi/n_0\right) \Delta \chi,$$

where $$n_0$$ is the average linear refractive index of the medium. With $$\Delta n \equiv n_2 |\mathbf{E}_{\omega}|^2$$, we have $$n_2 = (2\pi n_0) \chi^{(3)}$$.

A number of physical mechanisms can give rise to $$\Delta \chi$$ or $$\Delta n$$ (Shen, 1966). They have very different response times and different degrees of importance in different media. We discuss them separately in the following.

2.1 Electronic Mechanism

Classically, one can imagine that an applied optical field can distort the electronic distribution in a medium and hence induce a refractive index change. Quantum mechanically, the field can mix the electronic wave functions, shift the energy levels, and redistribute the population; all of these can contribute to the induced refractive index change. For a typical transparent liquid or solid, $$n_2$$ falls in the range between $$10^{-13}$$ and $$10^{-15}$$ esu. For gases at 1 atm pressure, $$n_2 \sim 10^{-16}$$ to $$10^{-18}$$ esu far away from resonances. The response time is of the order of the inverse bandwidth of the major absorption band ($$\sim 10^{-14}$$ to $$10^{-15}$$ s in condensed media) except for the population redistribution part. As the optical frequency approaches an absorption band, $$n_2$$ is resonantly enhanced. In particular, when the population redistribution due to resonant excitation is significant, the enhancement of $$n_2$$ can be very large, but the time response will then be dominated by the relaxation of the population redistribution. In a strong laser field, saturation in population redistribution and multiphoton resonant excitations can become important. The perturbative expansion in Eq. (1) may then cease to be valid. For our discussion of SPM

.. [[[20d689aa669f4c939680ad8851e4d354] [1. Theory of Self-Phase Modulation and Spectral Broadening]]]
in this chapter, we shall assume that the laser beam is deep in the transparent region and therefore all these electronic resonance effects on the induced refractive index are negligible.

2.2 Vibrational Contribution

The optical field can also mix the vibrational wave functions, shift the vibrational levels, and redistribute the populations in the vibrational levels. The corresponding induced refractive index change $\Delta n$ is, however, many orders ($\sim 5$) of magnitude smaller than that from the electronic contribution because of the much weaker vibrational transitions. Therefore, the vibrational contribution to $\Delta n$ is important only for infrared laser beams close to vibrational resonances. For our discussion of SPM, we shall not consider such cases.

If the laser pulse is very short (10 fs corresponding to a bandwidth of 500 cm$^{-1}$), the vibrational contribution to $\Delta n$ can also come in via Raman excitations of modes in the few hundred cm$^{-1}$ range. The Raman transitions are also much weaker than the two-photon electronic transitions, so their contributions to the self-induced $\Delta n$ are usually not important for the discussion of SPM unless femtosecond pulses are used.

2.3 Rotation, Libration, and Reorientation of Molecules

Raman excitations of molecular rotations can, however, contribute effectively to $\Delta n$. This is because the rotational frequencies of molecules are usually in the few cm$^{-1}$ region except for the smaller molecules. Thus, even with a monochromatic field, one can visualize a Raman process (in which absorption and emission are at the same frequency $\omega$) that is nearly resonant. (The difference frequency of absorption and emission is zero, but it is only a few cm$^{-1}$ away from the rotational frequencies.) In condensed media, the rotational motion of molecules is, however, strongly impeded by the presence of neighboring molecules. Instead of simple rotations, the molecules may now librate in a potential well set up by the neighboring molecules. The librational frequencies determined by the potential well are often in the range of a few tens of cm$^{-1}$. The modes are usually heavily damped. Like the rotational modes, they can also contribute effectively to $\Delta n$ via the Raman process.

Molecules can also be reoriented by an optical field against rotational diffusion. This can be treated as an overdamped librational motion driven by the optical field. More explicitly, molecular reorientation arises because the field induces a dipole on each molecule and the molecules must then reorient themselves to minimize the energy of the system in the new environment.

All the above mechanisms involving rotations of molecules can contribute appreciably to $\Delta n$ if the molecules are highly anisotropic. Typically, in liquids, $n_2$ from such mechanisms falls in the range between $10^{-13}$ and $10^{-11}$ esu, with
a response time around $10^{-11}$ s for molecular reorientation and $\sim 10^{-13}$ s for libration. In liquid crystals, because of the correlated molecular motion, $n_2$ can be much larger, approaching 0.1 to 1, but the response time is much longer, of the order of 1 s. The rotational motion is usually frozen in solids, and therefore its contribution to $\Delta n$ in solids can be neglected.

2.4 Electrostriction, Molecular Redistribution, and Molecular Collisions

It is well known that the application of a dc or optical field to a local region in a medium will increase the density of the medium in that region. This is because the molecules in the medium must squeeze closer together to minimize the free energy of the system in the new environment. The effect is known as electrostriction. The induced density variation $\Delta \rho$ obeys the driven acoustic wave equation, and from $\Delta n = (\partial n/\partial \rho)\Delta \rho$ the induced refractive index change can be deduced. For liquids, we normally have $n_2 \sim 10^{-11}$ esu with a response time of the order of 100 ns across a transverse beam dimension of $\sim 1$ mm.

Molecules will also locally rearrange themselves in a field to minimize the energy of induced dipole–induced dipole interaction between molecules in the system. Whereas electrostriction yields an isotropic $\Delta n$, this molecular redistribution mechanism will lead to an anisotropic $\Delta n$. Molecular correlation and collisions could also affect molecular redistribution. A rigorous theory of molecular redistribution is therefore extremely difficult (Hellwarth, 1970). Experimentally, molecular redistribution is responsible for the anisotropic $\Delta n$ observed in liquids composed of nearly spherical molecules or atoms in cases where the electronic, electrostrictive, and rotational contributions should all be negligible. It yields an $n_2$ of the order of $10^{-13}$ esu with a response time in the subpicosecond range. In solids, the molecular motion is more or less frozen, so the contribution of molecular redistribution to $\Delta n$ is not significant.

2.5 Other Mechanisms

A number of other possible mechanisms can contribute to $\Delta n$. We have, for instance, laser heating, which increases the temperature of a medium and hence its refractive index; photorefraction, which comes from excitation and redistribution of charged carriers in a medium; and induced concentration variation in a mixture.

We conclude this section by noting that there is an intimate connection between third-order nonlinearity and light scattering (Hellwarth, 1977): each physical mechanism that contributes to $\Delta n$ (except the electronic mechanism) is also responsible for a certain type of light scattering. The third-order susceptibility from a given mechanism is directly proportional to the
scattering cross section related to the same mechanism, and the response time is inversely proportional to the linewidth of the scattering mode. Thus from the low-frequency light scattering spectrum, one can predict the value of $n_2$ for the induced refractive index. For example, in most liquids, light scattering shows a Rayleigh wing spectrum with a broad background extending to a few tens of cm$^{-1}$. This broad background is believed to arise from molecular libration, redistribution, and collisions (Febellinski, 1967), but the details have not yet been resolved. For our semiquantitative prediction of $n_2$ and the response time, however, we do not really need to know the details if the Rayleigh wing spectrum of the medium is available. A broad and strong Rayleigh wing spectrum is expected to yield a large $n_2$ with a fast response.

In Table 1.1 we summarize the results of our discussion of the various physical mechanisms contributing to $\Delta n$. It is seen that in nonabsorbing liquid, where all the mechanisms could operate, electrostriction and molecular reorientation may dominate if the laser pulses are longer than 100 ns; molecular reorientation, redistribution, and libration may dominate for pulses shorter than 100 ns and longer than 1 ps; molecular redistribution and libration and electronic contribution may dominate for femtosecond pulses. In transparent solids, usually only electrostriction and electronic contribution are important. Then for short pulses the latter is the only mechanism contributing to $\Delta n$.

### Table 1.1.

<table>
<thead>
<tr>
<th>Physical mechanism</th>
<th>$n_2$ (esu)</th>
<th>Response time $\tau$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electronic contribution</td>
<td>$10^{-15}$–$10^{-13}$</td>
<td>$10^{-14}$–$10^{-15}$</td>
</tr>
<tr>
<td>Molecular reorientation</td>
<td>$10^{-13}$–$10^{-11}$</td>
<td>$10^{-11}$</td>
</tr>
<tr>
<td>Molecular libration and redistribution</td>
<td>$\sim10^{-13}$</td>
<td>$10^{-13}$</td>
</tr>
<tr>
<td>Electrostriction</td>
<td>$\sim10^{-11}$</td>
<td>$\sim10^{-16}$</td>
</tr>
</tbody>
</table>

* For a beam radius of $\sim$1 mm.

3. Simple Theory of Self-Phase Modulation and Spectral Broadening

For our discussion of SPM of light, let us first consider the case where the propagation of a laser pulse in an isotropic medium can be described by the wave equation of a plane wave:

$$\left( \frac{\partial^2}{\partial z^2} - \frac{n_0^2}{c^2} \frac{\partial^2}{\partial t^2} \right) E = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P^{(3)},$$

where
and \( n_0 \) is the linear refractive index of the medium. In the simple theory of SPM (Cheung et al., 1968; Gustafson et al., 1969; Shimizu, 1967), we use the usual slowly varying amplitude approximation by neglecting the \( \frac{\partial^2 \psi}{\partial t^2} \) term on the left and keeping only the \( (4\pi/c^2)\chi^{(3)}|\psi|^2 \) term on the right of Eq. (10), which then becomes

\[
\left( \frac{\partial}{\partial z} + \frac{n_0}{c} \frac{\partial}{\partial t} \right) \psi = \frac{4\pi\omega_0^2}{i2k_0c^2} \chi^{(3)}|\psi|^2 \psi. \tag{11}
\]

The approximation here also assumes an instantaneous response of \( \chi^{(3)} \). Letting \( z' \equiv z + ct/n_0 \) and \( \psi = |\psi| \exp(i\phi) \), we obtain from the above equation

\[
\frac{\partial |\psi|}{\partial z'} = 0,
\]

\[
\frac{\partial \phi}{\partial z'} = \frac{2\pi\omega_0^2}{c^2k_0} \chi^{(3)}|\psi|^2. \tag{12}
\]

They yield immediately the solution

\[
|\psi| = |\psi(t)|, \tag{13a}
\]

\[
\phi(z,t) = \phi_0 + \frac{2\pi\omega_0^2}{c^2k_0} \chi^{(3)}|\psi(t)|^2 z. \tag{13b}
\]

Equation (13a) implies that the laser pulse propagates in the medium without any distortion of the pulse shape, while Eq. (13b) shows that the induced phase change \( \Delta \phi(t) = \phi(z,t) - \phi_0 \) is simply the additional phase shift experienced by the wave in its propagation from 0 to \( z \) due to the presence of the induced refractive index \( \Delta n = (2\pi/n_0)\chi^{(3)}|\psi|^2 \), namely \( \Delta \phi = (\omega/c)\int_0^z \Delta n dz \). Since the frequency of the wave is \( \omega = \omega_0(\partial \phi/\partial t) \), the phase modulation \( \Delta \phi(t) \) leads to a frequency modulation

\[
\Delta \omega(t) = -\frac{2\pi\omega_0^2}{c^2k_0} \chi^{(3)} \frac{\partial |\psi|^2}{\partial t} z. \tag{14}
\]

The spectrum of the self-phase-modulated field is, therefore, expected to be broadened. It can be calculated from the Fourier transformation

\[
|E(\omega)|^2 = \frac{1}{2\pi} \left| \int_{-\infty}^{\infty} \psi(t) e^{-i\omega t + i\omega_0 t} dt \right|^2. \tag{15}
\]

An example is shown in Figure 1.1. We assume here a 4.5-ps full width at half-maximum (FWHM) Gaussian laser pulse propagating in a nonlinear medium that yields an SPM output with a maximum phase modulation of
Δφ_{max} \approx 72\pi \text{ rad}. The spectrum of the output shows a broadening of several hundred cm\(^{-1}\) with a quasi-periodic oscillation. It is symmetric with respect to the incoming laser frequency because the SPM pulse is symmetric. The leading half of the Δφ pulse is responsible for the Stokes broadening and the lagging half for the anti-Stokes broadening. The structure of the spectrum can be understood roughly as follows. As shown in Figure 1.1, the Δφ curve following the laser pulse takes on a bell shape. For each point on such a curve, one can always find another point with the same slope, except, of course, the inflection points. Since \(\partial \phi / \partial t = -\omega\), these two points describe radiated waves of the same frequency but different phases. These two waves will interfere with each other. They interfere constructively if the phase difference Δφ_{12} is an integer of 2\(\pi\) and destructively if Δφ_{12} is an odd integer of \(\pi\). Such interference then gives rise to the peaks and valleys in the spectrum. The inflection points that have the largest slope on the curve naturally lead to the two outermost peaks with \(|\omega_{\max}| \sim |\partial \phi / \partial t|_{\max}\). To find how many peaks we should expect in the spectrum, we need only to know \(\phi_{\max}\) as the number of pairs of constructive and destructive interferences is simply \(N \sim \phi_{\max}/2\pi\) on each side of the spectrum. The broadened spectrum has Stokes–anti-Stokes symmetry because Δφ\(\cdot\)t is directly proportional to \(|E(t)|^2\) and is a symmetric pulse.

Figure 1.1. Theoretical power spectrum obtained by assuming an instantaneous response of Δ\(\eta\) to the intensity variation \(|E(t)|^2\), so that the phase modulation Δφ\(\cdot\)t is proportional to \(|E(t)|^2\). (a) Δφ versus \(t\) and (b) power spectrum of the phase-modulated pulse.
With the above qualitative picture in mind, we can now generalize our discussion of SPM somewhat. The response of the medium to the laser pulse is generally not instantaneous. One therefore expects

\[ |E(t)|^2 \text{ is symmetric, } Df(t) \text{ is asymmetric and is no longer proportional to } |E(t)|^2. \]

The consequence is a Stokes–anti-Stokes asymmetry. An example is given in Figure 1.2. Because of the finite response time of the medium, the leading part of the \( Df(t) \) curve always sees a larger portion of the intensity pulse \( |E(t)|^2 \), and therefore the Stokes side of the spectrum is always stronger. This Stokes–anti-Stokes asymmetry can be drastic if the response time becomes comparable to or smaller than the laser pulse width.

In the more rigorous theory, one should also expect a distortion of the pulse shape as the pulse propagates on in the nonlinear medium. Self-steepening of the pulse, for example, is possible and may also affect the spectral broadening (DeMartini et al., 1967; Gustafson et al., 1969; see Chapter 6). However, the above qualitative discussion still applies since the \( D\phi(t) \) curve should still take on an asymmetric bell shape in general.
The experimental situation is usually not as ideal as the simple theory describes. The laser beam has a finite cross section and will diffract. The transverse intensity variation also leads to a $\Delta n(r)$ that varies in the transverse directions. This causes self-focusing of the beam and complicates the simultaneously occurring SPM of the beam (Shen, 1975; Marburger, 1975). Moreover, stimulated light scattering could also occur simultaneously in the medium, in most cases initiated by self-focusing (Shen, 1975; Marburger, 1975). All these make the analysis of SPM extremely difficult.

One experimental case is, however, close to ideal, namely SPM of a laser pulse in an optical fiber. The transverse beam profile of a guided wave remains unchanged along the fiber. As long as the laser intensity is not too strong, self-focusing and stimulated scattering of light in the fiber can be neglected. For a sufficiently short pulse, the nonlinearity of the fiber is dominated by the electronic contribution and therefore has a nearly instantaneous response. Then if the pulse is not too short and the spectral broadening is not excessive, the slowly varying amplitude approximation is valid and $\partial^2 P(3)/\partial t^2$ in the wave equation can be well approximated by $-\omega_0^2 P(3)$. The only modification of the simple theory of SPM we have discussed is to take into account the fact that we now have a wave in a waveguide with a confined transverse dimension instead of an infinite plane wave in an open space. Thus the quantitative analysis can easily be worked out. Indeed, Stolen and Lin (1978) found excellent agreement between theory and experiment.

The above discussion of SPM in time can also be used to describe SPM in space. As we already mentioned, the transverse intensity variation of a laser beam can induce a spatial variation of $\Delta n$ in the transverse directions. Let us consider here, for simplicity, a continuous-wave (cw) laser beam with a Gaussian transverse profile. The phase increment $\Delta \phi(r, z)$ varying with the transverse coordinate $r$ is given by

$$\Delta \phi(r, z) = (\omega/c) \Delta n(r) z. \quad (17)$$

This leads to a distortion of the wave front. Since the beam energy should propagate along the ray path perpendicular to the wave front, this distortion of the wave front would cause the beam to self-focus. If the propagation length is sufficiently long, the beam will actually self-focus and drastically modify the beam cross section. However, if the length of the medium is much shorter than the self-focusing distance, then the self-focusing effect in the medium can be neglected and we are left with only the SPM effect on the beam. The results of Figure 1.1 can describe the spatial SPM equally well if we simply replace $t$ by $r$ and $\omega$ by $k_\perp$, where $k_\perp$ is the transverse component of the wave vector of the beam. We realize that $k_\perp$ defines the deflection angle $\theta$ of a beam by the relation $k_\perp = (\omega n/c) \sin \theta$. Therefore, the quasi-periodic spectrum in the $k_\perp$ space actually corresponds to a diffraction pattern with multiple bright and dark rings. This has indeed been observed experimentally (Durbin et al., 1981; Santamato and Shen, 1984). An example is shown in Figure 1.3. Self-focusing or diffraction in the medium can modify the spatial
Figure 1.3. Diffraction ring pattern arising from spatial self-phase modulation of a CW Ar\(^+\) laser beam passing through a 300-\(\mu\)m nematic liquid crystal film. (After Durbin et al., 1981; Santamato and Shen, 1984.)