## Chapter 1

## Introduction

The invention of **photonic crystal fibers** (PCF) in 1996 [1] attracted much interest because of their interesting linear and nonlinear properties and their large potential for applications. We first define the object of our study. The term photonic crystal is used to denote a material that is periodically patterned in one, two, or three dimensions, the characteristic length or period being on the order of the wavelength. To obtain a photonic crystal fiber, one has to introduce a defect in the regular structure of a two-dimensional photonic crystal. An example of such a structure is given in Fig. 1.1. Here, the defect manifests itself as a missing cylindrical hole in an array, thus forming a guiding core surrounded by cladding which consists of photonic crystal. It was this type of PCF's which was first manufactured and studied in 1996. A typical method to manufacture a PCF [1] is to stretch a hexagonal preform at high temperature, cut it, stack the cut pieces and repeat this until the desired size and geometry is achieved. A lot of properties specific to PCF's has been discovered and studied since then. The peculiar combination of a solid microstructure core and the periodic photonic crystal around it gives rise to a

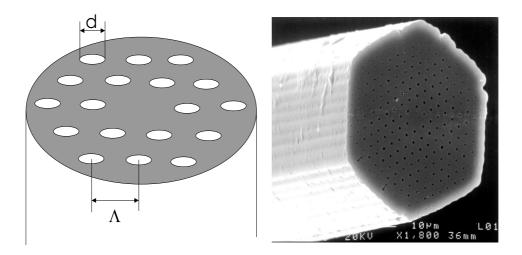


Figure 1.1: Schematic representation of a PCF and a cross-section photo of the first PCF (from [1]).  $\Lambda$  is the pitch, d is the hole diameter.

large number of exciting effects in waveguiding and dispersion. Among them, infinitely-single-mode guiding [2, 3]: due to the redistribution of the field in the photonic crystal cladding only one mode can propagate in PCF for any frequency, in contrast to standard fibers which become multimode at higher frequencies. Another interesting effect is photonic band gap guiding [4, 5], which occurs when states with certain frequencies cannot exist in photonic crystal cladding and are therefore guided in the core.

However, the most promising property from the point of view of applications is a **the shift of the zero dispersion wavelength** [6, 7, 8]. It was quickly recognized that PCF's allow the repositioning of the zero dispersion wavelength and to shift it far from its usual position at 1.3  $\mu$ m. It is also possible to design PCF's which have dispersion significantly lower than that in usual fibers over a wide spectral range [9]. The reason for this effect is a large index step between the core and the cladding combined with the  $\mu$ m-scale of the core. In this respect PCF's are very similar to so-called tapered

fibers, which are made by stretching a standard fiber to form a  $\mu$ m-diameter strand of silica surrounded by air [10]. Indeed many PCF's with thin dividers between the holes have a geometry similar to that of tapered fibers (see e.g. Ref. [7],[11]). It will be shown in this work that another important peculiarity, namely generation of spectrally ultrabroad radiation also called <u>supercontinuum</u> (SC) after low-intensity pulse propagation in PCF's, is caused by this shift. **Low-intensity supercontinuum generation** has been observed in PCF's in 2000 [8] and also in tapered fibers [10].

As such, spectral broadening by self-phase modulation was discovered in fibers decades ago [12]. It is based on the amplitude dependence of the refractive index which induces a time-dependent phase and yields new spectral components. Spectra covering several octaves have been observed for high-intensity input pulses in various media, as e.g. in Ref. [13].

However, until recently no explanation existed for the mechanism of such broadening in PCF's, because the intensities necessary to obtain two-octave-broad spectra by the effect of self-phase modulation (SPM) are more than two orders of magnitude higher [14]. To resolve this problem and to clarify the mechanism of SC generation is thus interesting and an important task, which is undertaken by this work. It will be shown that SC in PCF is generated not by SPM but by a novel mechanism involving higher-order solitons which arise after the pulse enters the fiber in the anomalous dispersion regime. These solitons split due to the higher-order dispersive effects and the constituent fundamental solitons shift to the IR while simultaneously emitting non-solitonic radiation which covers the wide spectral range. Numerical as well as experimental evidence for this mechanism is presented, and the soliton dynamics is studied in detail.

There is another important nonlinear effect in the PCF's: **four-wave-mixing**. While it also contributes to spectral broadening, it specifically manifests itself in the spectrum as two clearly separated sidebands far from the pump frequency [15, 16]. Peculiar four-wave-mixing is also caused by the specific dispersion in PCF's. The positions of sidebands are determined by the phase-matching condition, and for the PCF's this condition shows that radiation can be generated in the IR and around 600 nm by the pump in the visible region at 800 nm – effects which would never be possible for standard fibers. We systematically study the factors which affect the effectivity of the 4-wave-mixing such as walkoff, show the possibility to control the position of sidebands in a wide range by the appropriate pulse and fiber parameters, and establish the contribution of 4-wave-mixing to SC generation.

The interest for such radiation is caused by a large number of applications for supercontinuum radiation (for an overview see [13]). Recently, a very fascinating method to use SC radiation in PCF's for frequency metrology was proposed [17, 18]. Known complicated methods to measure the optical frequency with high accuracy can be replaced by a novel approach which requires significantly less efforts. The radiation of the stabilized laser is known to be constituted from separate spectral lines known as a "frequency comb". This comb could serve as a "ruler" to measure the unknown frequency by letting it interfere with the comb and measuring the beat frequency. However, the unknown phase shift between the envelope and the carrier causes a drift of the comb and constitutes the main problem of this method. This is where supercontinuum generation comes into play: by comparing the frequencies of octave-separated peaks of the comb the phase shift can be measured and fixed [18]. Other important applications include, for

example, time-resolved absorption spectroscopy in which ultrafast relaxation processes are studied by sending the SC pulse after the intense pump pulse which excites the medium and examining the dependence of the induced transient optical density on the delay [19]. Various processes in the solid state [20], biological tissue (see e.g. Ref. [21]), and chemistry [22] have been studied by this method. Several ultrafast excitation spectroscopy techniques using the continuum were developed; they include coherent anti-Stokes Raman scattering [23], Raman-induced phase conjugation [24], and others. However, one of the most important applications for SC is the generation of ultrashort pulses.

A lot of effort in the recent decades has been dedicated to the production and operating of ultrashort pulses, with the current record in the visible and near-UV being 3.7 fs obtained by compensating Raman-induced broad spectra [25]. The reasons for this are the various applications which such pulses find in time-domain study of atoms, molecules, solids, and processes of biological interest (for overviews see e.g. [26, 13, 27]). Femtosecond pulses allow the study of ultrafast transients in atoms, for example by exciting a radial Rydberg wave packet [28], which is a superposition of many highlyexcited electronic states with classical properties. Examples of the research of ultrafast properties in molecules are the studies of the dissociation dynamics. This is done by a delayed probing of the dissociating molecule to determine the decay of the excited electronic state and the potential surface [29]. In solids, the intraband relaxation properties can be studied with fs pulses by creating states above the gap, which has important applications for miniaturization [30]. Femtosecond pulses can provide, for example, an ultrashort gating function for the 3D imaging. In this way it is possible to

achieve the fs temporal resolution [31] for the signal, which is necessary to obtain detailed depth image of a target. Femtosecond pulses are also important in high-harmonic generation [32]. Early efforts in producing ultrashort pulses utilized modelocking of laser radiation. Modelocking techniques can be divided into two categories: active and passive modelocking. Active modelocking implies external modulation of one of the resonator elements with a period equal to the round-trip time thus amplifying a certain part of the radiation (see e.g. [33] and other references in [27]), and can be implemented as gain modulation (switching the pump on and off), loss modulation (periodical decrease of loss in some element), and cavity dumping (accumulating energy in the resonator cavity and releasing it in a short burst by misaligning one mirror). Passive modelocking implements lower losses (or higher gain) for the more intense fractions of the radiation. These lower losses are induced by such fractions themselves, examples being saturable absorption modelocking [34] and Kerr lens modelocking [35], in which more intense radiation achieves favorable transverse profile by selffocusing. However, most of the recent success in the generation of short pulses is based on the compensation of the phase of the SC. Large spectral width does not automatically mean that the corresponding pulse is short. The shortest pulse (for the given spectrum) forms in the case when the spectral phase is a linear function of the frequency. To obtain this, it is necessary to introduce an element which will compensate the phase modulation of the broad spectrum by introducing its own contribution. Several devices were proposed for this purpose. For example, a 4.5 fs pulse was obtained in 1997 by passing a SC, which emerges after propagation of an intense pulse through an Ar-filled fiber, through a pair of chirped mirrors, as reported in Ref. [36] (see also [37]). A chirped

mirror consists of several layers of alternating materials with different refractive index. The interfaces between the layers serve to reflect light, and for a certain combination of the wavelength and interlayer distance the interference between the waves reflected from interfaces causes full reflection [27]. The layer thickness can be varied through the depth of the mirror, so that waves with different wavelength traverse different paths through the mirror before being reflected. This allows the creation of linear elements with designable negative group-velocity dispersion (GVD) which can compensate for normal GVD of other optical elements as well as dispersion in higher orders. However, such mirrors are not controllable and compensate only a limited number of phase modulation orders. Spatial liquid crystal light modulators do not have these disadvantages. The working principle is generally simple: the input pulse is decomposed into the spectral components by a grating, so that the transverse displacement is given to each spectral component of the beam [38]. After that, they pass through a liquid crystal, which is composed of numerous (>100) channels. Each channel is controlled by an external voltage which leads to a change of its refractive index due to reorientation of molecules of the liquid crystal. In this way, a controllable phase shift is introduced for each of the spectral components, which are finally recombined into the single pulse. Currently available modulators can have up to 400 channels and operate in the wavelength range from 400 to 1800 nm, albeit there exists no single device with such a broad transmission window. One can hope that with further development of the technology it will be possible to control the phase shift of radiation from 400 to 1800 nm, and, with sufficient number of channels, to achieve an output pulse duration around 1 fs.

The main reason why gas-filled hollow fibers can not be used as a

source of still shorter pulses with duration around and below 1 fs is due to the dispersion in them. Phase modulation of the final pulse imposes practical limitations on the shortening by phase compensation. However, a phase modulation during the propagation also affects the effectivity of self-phase modulation. During the spectral broadening, the new spectral components have a large difference of group velocities and therefore the peak amplitude of the pulse drops rapidly with propagation. One of the possible solutions of this problem is to combine the dispersive properties of the hollow fiber and bulk noble gas to maintain the high intensity of a pulse during propagation. This could lead to an increase of the effective nonlinear length and therefore could allow achievement of broader spectra. One of the aims of this work is to study the interaction of nonlinear and dispersive effects in hollow fibers and to try to optimize the system design to obtain broadest possible pulses.

Standard theoretical methods of nonlinear optics are not applicable to simulate the processes with SC, and novel theoretical approaches are necessary. The reason for this is that usual methods rely on the slowly varying envelope approximation (SVEA), in which the temporal dependence of the field is represented as a product of a carrier (plane wave with fixed frequency) and an envelope. The crucial requirement in the standard methods is that the envelope changes much slower with time than the carrier, thus simplifying theoretical analysis. However, this implies that the spectral width of the radiation is much smaller than the wavelength. This is clearly not the case for SC. In principle, it is possible to represent a field which contains several waves as a sum of waves each satisfying the SVEA. But even this does not help for SC, because the necessary number of waves will be extremely large and because it is impossible to predict which frequen-

cies will be excited before the numerical study. Another standard method is the representation of the linear dispersion by a Taylor expansion of the wavenumber with only a few lowest orders of dispersion taken into account. This is not enough for a supercontinuum because of the large spectral width of the radiation, which makes a Taylor expansion of dispersion around one frequency inapplicable. Both of these problems are overcome by the novel propagation equation developed and proposed in this work. It does not rely on the envelope methods, because it resolves the carrier oscillations, and thus can be applied to radiation with arbitrarily broad spectra. Moreover, it uses a global approach for dispersion, i.e. accounts for dispersion to all orders, which is especially important for the problems considered here.