Understanding the pulse propagation dynamics in gas-filled hollow-core optical waveguides

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**UNDERSTANDING THE PULSE PROPAGATION
DYNAMICS IN GAS-FILLED HOLLOW-CORE
OPTICAL WAVEGUIDES**

WAN YING

School of Electrical & Electronic Engineering

A thesis submitted to the Nanyang Technological University
in partial fulfilment of the requirement for the degree of

Doctor of Philosophy

2021

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research, is free of plagiarised materials, and has not been submitted for a higher degree to any other University or Institution.

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Wan Ying

Supervisor Declaration Statement

I have reviewed the content and presentation style of this thesis and declare it is free of plagiarism and of sufficient grammatical clarity to be examined. To the best of my knowledge, the research and writing are those of the candidate except as acknowledged in the Author Attribution Statement. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

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Asst Prof Chang Wonkeun

Authorship Attribution Statement

This thesis contains material from 4 papers published in the following peer-reviewed journals and from papers accepted at conferences in which I am listed as an author.

Chapter 3 is published as Y. Wan, J. Luo, and W. Chang, "Tuning the soliton-effect compression via stretching of the pump," *Laser Physics*, vol. **30**, no. 10, p. 105401 (2020). DOI: 10.1088/1555-6611/abad42.

The contributions of the co-authors are as follows:

- Asst Prof Wonkeun Chang edited the manuscript drafts, verified the correctness of my simulation results and analyzed the results.
- I proposed the manuscript topic, performed the simulation and analysis. I also prepared the manuscript drafts.
- Dr. Jiaqi Luo discussed the results and provided valuable suggestions.

Chapter 5 is published as Y. Wan and W. Chang, "Effect of decreasing pressure on soliton self-compression in higher-order modes of a gas-filled capillary," *Opt. Express*, vol. **29**, no. 5, pp. 7070-7083 (2021). DOI: 10.1364/OE.418217.

The contributions of the co-authors are as follows:

- Asst Prof Wonkeun Chang provided the initial research direction and edited the manuscript drafts.
- I proposed the manuscript topic, performed the simulation and analyzed the simulation results. I also wrote the drafts of the manuscript.

The contributions of the co-authors are as follows:

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Summary

Gas-filled hollow-core optical waveguides offer high damage threshold, wide transmission window and tunable optical properties, providing an ideal platform for studying ultrafast nonlinear dynamics and light-matter interactions. In this thesis, we numerically investigate pulse propagation dynamics in two types of gas-filled hollow-core optical waveguides, namely, gas-filled capillaries and anti-resonant hollow-core fibers.

The pulse propagation dynamics inside a gas-filled hollow-core waveguide can be tuned by altering the parameters of pump pulse, medium or waveguide. One commonly used tuning technique is to introduce linear frequency chirp on the pump pulse. We investigated numerically the effect of initial chirp on pulse compression dynamics. By stretching the pulse temporally using a dispersive optical component, the chirp can be introduced without affecting the spectrum. Our study shows that the soliton-effect pulse compression can be enhanced by both positive and negative initial chirp through different mechanisms. Moreover, the compression length can be tuned over a wide range by varying the amount of chirp. Interestingly, the initial chirp that leads to the best compression always shifts the compression length to 1.23 times that of corresponding unchirped pulse.

Alternatively, the parameters of the propagation medium can also be tuned to improve the pulse compression. One advantage of gas-filled hollow waveguides is that the optical property can be conveniently tailored by changing the gas pressure. We numerically demonstrated soliton-effect compression and supercontinuum generation by pumping a 100 fs 5 uJ pulse centered at 2460 nm into the fundamental mode of a xenon-filled anti-resonant hollow-core fiber with three types of pressurization configurations, yield decreasing pressure from 41 to 0 bar, constant pressure of 27.3 bar and increasing pressure from 0 to 41 bar. We demonstrated that, as compared to a constant pressure, applying decreasing pressure scales up the peak power of the compressed pulse by 1.63 times and enhances the blue edge of spectrum with emission of

multiple dispersive waves. The enhancement of soliton-effect compression in the mid-infrared region by decreasing pressure is not observed before. On the contrary, increasing pressure results in the lowest peak power and the least spectral broadening. We also showed that a decreasing pressure system is highly resistant to ionization.

We further extended investigation on impact of decreasing pressure to HE_{12} , HE_{13} and HE_{14} mode of a gas-filled capillary with core diameter of $300\ \mu m$. When applying decreasing pressure from 1.8 to 0 bar, a 30 fs 100 μJ pulse centered at 800 nm can be compressed down to 1.85 fs in the HE_{12} mode, with a peak power of 7.96 GW. The compression ratio is enhanced by four times as compared to the equivalent constant pressure of 1.2 bar. This is similar to the results in previous section, where we pump in the fundamental mode of an anti-resonant hollow-core fiber. There is no enhancement of compression with decreasing pressure in HE_{13} and HE_{14} mode. Furthermore, a decreasing pressure effectively suppresses intermodal nonlinear coupling near the end of the fiber and produce output pulse with high mode purity for all the three modes.

We also investigated another effect that depends on pulse and fiber parameters and is neglected in most cases—the mode-area dispersion in anti-resonant hollow-core fiber. It directly links to strong wavelength dependency of nonlinear coefficient near resonance region. We find this effect is nonnegligible when using a small-core fiber and pumping near the resonance. In this work, we demonstrated the effect of mode-area dispersion on pulse propagation dynamics by comparing the results obtained with two models, one with inclusion of this effect and the other one without this effect. Differences between the two results are analyzed and explained. We also carried out simulation for different input energies and observe that mode area dispersion become important as pump energy increases.

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List of Abbreviations

AR-HCF	anti-resonant hollow-core fiber
ARROW	anti-resonant reflecting optical waveguide
DW	dispersive wave
DUV	deep ultraviolet
DFWM	degenerate four-wave mixing
FEM	finite element method
FWM	four-wave mixing
FROG	frequency-resolved optical gating
GNLSE	generalized nonlinear Schrodinger equation
GVD	group-velocity dispersion
HOM	higher-order mode
IR	infrared
MIR	mid-infrared
MM-GENLSE	multimode generalized nonlinear Schrodinger equation
MPA	massively parallel algorithm
NLSE	nonlinear Schrodinger equation
SPM	self-phase modulation
UPPE	unidirectional pulse propagation equation
VUV	vacuum ultraviolet
XPM	cross-phase modulation
ZDW	zero-dispersion wavelength

Chapter 1: Introduction

1.1 Background

In recent years, increasing attention has been paid to gas-filled hollow waveguides and they have revolutionized the field of ultrafast nonlinear optics [1, 2]. As compared to solid-core waveguides, one major difference for gas-filled hollow-core waveguides being that the refractive index of the core is smaller than that of the cladding. Light can be guided in hollow-core waveguides by various mechanisms, such as partial reflection (for capillaries), anti-resonant reflection [3] and inhibited-coupling [4, 5] (for anti-resonant hollow-core fibers), or photonic bandgap effect [6] (for photonic bandgap fibers).

Gas-filled hollow waveguides possess unique advantages for ultrafast optics. They can tolerate high intensity that is otherwise damaging to solid materials. They can also access unconventional wavelength which suffers from high absorption loss in silica. They also provide platform for extreme nonlinear dynamics such as plasma effect [7], which is not attainable in conventional solid-core fibers. Importantly, the dispersion and nonlinearity in gas-filled hollow-core waveguides can be conveniently tuned by simply adjusting the gas pressure or filling with different types of gas [1, 2], thus providing extra degree of freedom on engineering the optical properties.

Two types of gas-filled hollow-core optical waveguides are covered in this thesis, namely hollow capillaries and anti-resonant hollow-core fibers. Hollow capillaries have excellent power handling ability and weak waveguide dispersion due to the large core size. They have been utilized for nonlinear-optics based applications since 1970 [8, 9], with major focus on spectral broadening in post-compression system [10, 11]. They are also used for self-focusing [12] and frequency conversion via various nonlinear processes such as high-harmonic generation and four-wave mixing [13, 14]. Ever since 2019, capillaries have shown great

success in energetic soliton-effect compression down to sub-cycle regime, with generation of dispersive wave from vacuum ultraviolet (VUV) to infrared (IR) region [15, 16], paving the way to the development of high-energy ultrafast optics. On the other hand, negative-curvature anti-resonant hollow-core fibers emerge in 2011 [17, 18]. It has attractive features such as low light-silica overlap [19] with wide transmission window from VUV to MIR. Therefore, they are widely applied in multioctave-spanning supercontinuum generation and wavelength-tunable dispersive wave generation [20-25]. Besides, it has one more feature that is absent in the capillary fiber—the resonance band, which has been a hot topic in recent years [26-28]. There are different types of anti-resonant hollow-core fibers with various cladding structures in the form of single-ring [17], double-ring [29], ice-cream-cone [18] as well as nested-element [30]. Among these, single-ring anti-resonant hollow-core fibers have the simplest cladding structure and can provide low-loss guidance especially in the MIR region [19]. The confinement loss of anti-resonant hollow-core fiber is much lower than that of the capillary fiber with equivalent size. Moreover, they can be scaled down to much smaller core size and significantly reduce the required input energy [31].

Ultrafast light sources generated by gas-filled hollow-core optical waveguides target at wide range of applications such as spectroscopy [32], microscopy [33], metrology [34] and optical coherence tomography [35] across the field of physics, chemistry, biology and materials science. In this thesis, we focus on techniques for enhancing soliton-effect compression in gas-filled hollow-core waveguides. We also carry out in-depth analysis on factors that affect the pulse propagation dynamics as well as the mechanisms behind.

1.2 Motivation

To fully exploit the advantages such as high-damage threshold, wide transmission window and pressure-tunability of gas-filled hollow-core optical waveguides, there are active research in

three general directions. These are (i) achieving large pulse compression factor [36-38], (ii) reaching high peak power [39, 40] and (iii) generating exotic wavelength [15, 23], for a wide range of ultrafast pump sources and media.

In gas-filled hollow-core optical waveguides, high-intensity ultrashort pulses with large compression factor can be obtained by two types of pulse compression techniques, yield post-compression and soliton-effect compression. In the post-compression method, a high-intensity pulse propagates in a hollow-core fiber to increase the pulse bandwidth via self-phase modulation, resulting in a positive chirp across the central part of the pulse. This is compressed using anomalously dispersive optical components placed at the output of the fibre to temporally compress the pulse down to few-cycles. In the soliton-effect compression, it is the interplay of anomalous dispersion and self-phase modulation that leads to a pulse self-compression in a single stage. As compared to post-compression technique, soliton-effect compression enables pulse to be compressed down to few-cycle and even sub-cycle in a single-stage system and no chirp compensation is required [41-43]. In some circumstance, the compressed pulse requires further optimization in terms of pulse duration or pulse quality. Pulse compression length may also need to be adjusted to ensure the pulse exits the fiber with shortest possible duration. However, due to the complex experimental set-up for gas-filled hollow waveguides, the fiber length cannot be freely adjusted. One way to optimize the compression and control the compression length is by stretching the pulse and introducing a linear chirp. However, questions like how much chirp to apply, what is the optimal chirp to achieve best compression as well as how does the compression length changes with chirp remains unclear.

As the pressure of gas chambers attached at both ends of fiber can be independently controlled, it is easy to achieve pressure gradient in gas-filled hollow waveguides. Applying pressure gradients for near-infrared soliton-effect compression in gas-filled hollow-core waveguides has been discussed in literatures [44, 45]. In [44], a decreasing pressure is shown to degrade the

compression performance. However, we are not sure whether the observation is generally applicable when we use a different pulse, gas or waveguide. Moreover, when there is pressure in the gas chamber attached at the output end of fiber, the ultrashort pulse could be distorted by dispersion and nonlinearity of the gas, which is undesired. Hence a decreasing pressure with evacuated end is more preferred for practical purposes.

A decreasing pressure could be more beneficial for soliton-effect compression in the higher-order modes of gas-filled capillary. High intensity pulse propagates in the higher-order modes of gas-filled capillary could suffer from strong intermodal nonlinear coupling, which degrades the mode purity. For those applications that require specific unconventional intensity profile, such as microscopy [46], micromachining [47] etc., a high mode purity is important. The characteristic of decreasing pressure shows its ability to suppress intermodal coupling raised by strong nonlinearity in the late stage of compression. By taking advantages of this special type of pressure distribution, gas-filled capillary could provide an ideal platform to obtain energetic ultrashort pulse with high mode purity in the higher-order modes, which otherwise cannot be achieved in conventional solid-core fibers due to much lower power damage threshold and stronger intermodal nonlinear coupling induced by high nonlinear coefficient of solid-core material, such as silica.

Besides initial chirp, pressure distributions, one more factor that could affect the pulse propagation dynamics and is often neglected is the mode-area dispersion. In recent years, there is a growing interest on investigating nonlinear dynamics near the resonance [26, 48], where the effect of mode area dispersion is relatively strong. Hence it is important to consider such effect in these cases. When dealing with numerical studies on nonlinear dynamics near resonance region, the accuracy of simulation results could be affected if the effect of mode-area dispersion is not considered.

1.3 Objective

This thesis aims to numerically investigate some most-overlooked parameters of pulse, medium and waveguide that affect the pulse propagation dynamics in gas-filled hollow-core waveguides and explore solutions to improve the soliton-effect compression by tuning some of these parameters, it includes:

- Investigating the impact of stretching-induced chirp on soliton-effect compression to provide additional control on the compression length, pulse duration, peak power as well as pulse quality in a set-up for gas-filled hollow-core waveguide by just stretching the pulse.
- Exploiting the pressure-tunability of gas-filled hollow-core waveguides and utilizing the advantage of decreasing pressure to scale up the peak power for soliton-effect compression of supercontinuum in the MIR region.
- Taking advantage of decreasing pressure to enhance the soliton-effect compression in the higher-order modes of gas-filled capillaries and suppress intermodal coupling to produce energetic ultrashort pulse with high mode purity.
- Investigating the impact of resonance-induced mode-area dispersion on nonlinear dynamics in AR-HCFs and demonstrating how the nonlinear dynamics are affected differently with different pump wavelength and input pulse energy.

1.4 Major contribution

1. We numerically investigated the effect of stretching-induced linear chirp on soliton-effect pulse compression in a dispersive nonlinear medium using the Nonlinear Schrödinger Equation (NLSE). Unlike in previous studies where only few chirp values are selected for investigation, we systematically scanning through a series of chirp values spanning from negative to positive for higher-order soliton with $N= 2$ to 8 and calculated the four important parameters: shortest pulse duration, highest peak power,

compression length and pulse quality for all of these chirp values. We found that soliton-effect compression can be enhanced by both positive and negative chirp through different mechanisms. In particular, enhancement of soliton-effect compression by negative initial chirp is never observed in the past. We also found the compression length can be widely adjusted by fine-tune the initial chirp. It is interesting to note that: within a certain range of positive chirp, the compression length can be reduced to below that of the transform-limited pulse. Furthermore, we found the compression length for pulse with optimal positive initial chirp is always 1.23 times that of the unchirped pulse. Our results reveal some hidden characteristics that is undiscovered before and complement the previous studies.

2. We exploited the pressure-tunability and numerically studied the effect of different pressure distributions on the soliton-effect compression of MIR supercontinuum in a xenon-filled AR-HCF. We found that a decreasing pressure from 41 bar to 0 bar can achieve a 1.63 times higher compression ratio as compared to the equivalent constant pressure at 27.3 bar while increasing pressure degrades the compression performance. By comparing the three types of configurations, we deduced that the enhancement of soliton-effect compression is a result of strengthened group-velocity dispersion and suppression of higher-order dispersion with decreasing pressure.
3. We developed a numerical tool for simulating pulse propagation in gas-filled capillary based on Multimode Generalized Nonlinear Schrodinger Equation (MM-GNLSE) and further extended our study to higher-order modes. We numerically studied the effect of decreasing pressure on soliton-effect compression in higher-order modes of gas-filled capillary for the first time. By launching a 30 fs 100 μJ pulse centred at 800 nm into HE_{12} , HE_{13} and HE_{14} mode of a capillary fiber with core diameter of 300 μm , we observed that: a decreasing pressure from 1.8 bar to 0 bar improves the compression

ratio in HE_{12} mode by 4 times as compared to the equivalent constant pressure at 1.2 bar. Like our previous studies in Chapter 4, the enhancement in HE_{12} mode is due to suppression of higher order dispersion and thus weaken of perturbation imposed on soliton by decreasing pressure. Moreover, we found decreasing pressure brings an additional benefit—it results in longitudinally diminishing nonlinear coefficient and this can effectively attenuate the strength of intermodal nonlinear coupling, and thus improve the mode purity at fiber output. These findings are of great importance to the field of ultrafast optics.

4. Mode-area dispersion is previously neglected in modelling pulse propagation in gas-filled AR-HCF. We implemented a modified GNLSE model to take account of the mode-area dispersion and compared the numerical results when including and excluding this effect in simulation. We found this effect cannot be neglected when we use a small-core AR-HCF with diameter of 20 μm and pumping close to the resonance wavelengths. We identified that the mode area dispersion affects the pulse propagation by inducing strong wavelength dependency of nonlinear coefficient, which affects the soliton dynamics and conversion efficiency of dispersive wave generation and four-wave mixing process. Besides fiber dimension and pump wavelength, we observed this effect is also subject to input pulse energy and it generally become more prominent with increasing pulse energy. The detailed investigation on effect of mode-area dispersion in gas-filled AR-HCF is not reported before.

1.5 Organization of the Thesis

Chapter 1 briefly introduces the background on gas-filled hollow-core waveguides as well as the motivation and objectives of the thesis topic. The major contribution is highlighted and the organization of the thesis content is outlined.

In Chapter 2, different linear and nonlinear responses in gas-filled hollow-core waveguides and some widely observed nonlinear phenomenon are introduced. Four main physical models that are used for modelling pulse propagation are described. Hollow capillary fiber and anti-resonant hollow-core fiber are introduced.

Chapter 3 investigates how the linear chirp induced by stretching affect the soliton-effect compression. Key parameters related to soliton-effect compression with different initial chirps are calculated. The mechanisms that explain the enhancement of compression in different regimes are analyzed.

In chapter 4, simulation on soliton-effect compression and MIR-supercontinuum generation in gas-filled anti-resonant hollow-core fiber with three types of pressure distributions are demonstrated. The reasons for improved compression and enhanced blue edge of spectrum with decreasing pressure are explained.

In chapter 5, impact of decreasing pressure on soliton-effect compression in HE_{12} , HE_{13} and HE_{14} modes of gas-filled capillary is investigated. The compression performance with decreasing pressure and equivalent constant pressure are compared and the differences are analyzed. The suppression of strength of intermodal nonlinear coupling with decreasing pressure is presented.

The effect of mode-area dispersion on nonlinear dynamics in a small-core AR-HCF is investigated in Chapter 6. Simulation is carried out at two different pump wavelengths near the resonance region, each one is simulated with two models, namely with and without the effect of mode-area dispersion. The results are compared and theoretical analysis about the differences caused by the two models is presented.

Chapter 2: Basis theories on pulse propagation dynamics in gas-filled hollow-core optical waveguides

2.1 Electromagnetic wave propagation equation

The propagation of electromagnetic wave can be well described by the Maxwell's equations [49]:

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad (2.1)$$

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J}, \quad (2.2)$$

$$\nabla \cdot \mathbf{D} = \rho_f, \quad (2.3)$$

$$\nabla \cdot \mathbf{B} = 0, \quad (2.4)$$

where:

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}, \quad (2.5)$$

$$\mathbf{B} = \mu_0 \mathbf{H} + \mathbf{M}, \quad (2.6)$$

$\mathbf{E}$ and $\mathbf{H}$ are the electrical and magnetic field, respectively. $\mathbf{D}$ is the electric displacement. It represents material's response to the applied electric field. $\mathbf{B}$ is the magnetic flux density. ρ_f and $\mathbf{J}$ represent the free charge and current density. $\mathbf{P}$ and $\mathbf{M}$ are the induced electric and magnetic polarization. $\mathbf{P} = \mathbf{P}_L + \mathbf{P}_{NL}$ where $\mathbf{P}_L$ and $\mathbf{P}_{NL}$ is the linear and nonlinear polarization. In dielectric and non-magnetic waveguide without free charges, $\rho_f = \mathbf{J} = \mathbf{M} = 0$. ε_0 and μ_0 are the permittivity and permeability of free space, respectively.

By taking the curl operation on Eq. (2.1), we get:

$$\nabla \times \nabla \times \mathbf{E} = -\frac{\partial}{\partial t}(\nabla \times \mathbf{B}). \quad (2.7)$$

From Eq. (2.2), (2.5) and (2.6), we can obtain:

$$\nabla \times \mathbf{B} = \mu_0(\nabla \times \mathbf{H}) = \mu_0 \frac{\partial \mathbf{D}}{\partial t} = \varepsilon_0 \mu_0 \frac{\partial \mathbf{E}}{\partial t} + \mu_0 \frac{\partial \mathbf{P}}{\partial t}, \quad (2.8)$$

Substitute Eq. (2.8) into Eq. (2.7) and make use of the identity $\nabla \times \nabla \times \mathbf{E} \equiv \nabla \cdot (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$ as well as the fact that $\nabla \cdot \mathbf{E} = 0$, we have:

$$\nabla^2 \mathbf{E} = \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2} = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int_{-\infty}^t \varepsilon_r(t-t') \mathbf{E}(t') dt + \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}, \quad (2.9)$$

ε_r is the relative permittivity. Taking the Fourier transform of Eq. (2.9) and using the relation $\frac{\partial^2 \mathbf{E}(t)}{\partial t^2} \leftrightarrow (i\omega)^2 \mathbf{E}(\omega)$, we get [50]:

$$\nabla^2 \mathbf{E}(\omega) + k_0^2 n_l^2(\omega) \mathbf{E}(\omega) + \mu_0 \omega^2 \mathbf{P}_{NL}(\omega) = 0, \quad (2.10)$$

where $k_0 = \omega/c$ is the wave vector in free space. n_l is the linear refractive index.

2.2 Linear effects

Electric polarization consists of both linear and nonlinear parts given as:

$$\mathbf{P} = \mathbf{P}_l + \mathbf{P}_{nl} = \varepsilon_0 (\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} |\mathbf{E}(t)| \mathbf{E}(t) + \chi^{(3)} |\mathbf{E}(t)|^2 \mathbf{E}(t) + \dots), \quad (2.11)$$

$\mathbf{P}_l = \varepsilon_0 \chi^{(1)} \mathbf{E}(t)$ is the linear polarization, which responses linearly with the electrical field $\mathbf{E}(t)$. Linear effects are related to $\beta(\omega) = \frac{\omega \sqrt{\varepsilon}}{c}$, where $\varepsilon = \chi^{(1)} + 1$ is the linear permeability and $\chi^{(1)}$ is the first order susceptibility in the complex form: $\chi^{(1)} = \chi' - i\chi''$ [49]. χ' is responsible for dispersion while $i\chi''$ is related to attenuation. Dispersion and attenuation make up the two types of linear effects for pulse propagation in optical waveguides. They affect only the phase of propagating pulse without generating new frequency components. Dispersion can be expanded into a Taylor series around carrier frequency ω_0 :

$$\beta(\omega) = \beta_0 + \beta_1(\omega - \omega_0) + \frac{1}{2} \beta_2(\omega - \omega_0)^2 + \dots, \quad (2.12)$$

where $\beta_k = \frac{d^k \beta}{d\omega^k} |(\omega = \omega_0)$ is known as the dispersion coefficient that evaluated at central frequency ω_0 . Among these, β_1 is the inverse of group velocity. β_2 is commonly known as the group velocity dispersion, which refers to the frequency dependence of group velocity. When $\beta_2 > 0$, long wavelength component in the pulse travels faster than the short wavelength components and this is known as normal dispersion regime. When $\beta_2 < 0$, it is the vice versa

and this is known as anomalous dispersion regime.

Generally speaking, attenuation is also a wavelength dependent parameter that can be expanded into Taylor series given as:

$$\alpha(\omega) = \alpha_0 + \alpha_1(\omega - \omega_0) + \frac{1}{2}\alpha_2(\omega - \omega_0)^2 + \dots, \quad (2.13)$$

An input pulse with peak power of P_i propagates in waveguide with loss of α will lead to an output pulse with peak power P_o that satisfies the relation:

$$P_o = P_i \exp(-\alpha(\omega)z), \quad (2.14)$$

where $\alpha(\omega)$ is in the unit of dB/m.

2.3 Nonlinear effects

New frequency components can only be generated by nonlinear effects through nonlinear polarization expressed as:

$$\mathbf{P}_{nl} = \varepsilon_0(\chi^{(2)}|\mathbf{E}(t)|\mathbf{E}(t) + \chi^{(3)}|\mathbf{E}(t)|^2\mathbf{E}(t) + \dots). \quad (2.15)$$

We only consider homogenous and isotropic media such as noble gas, the even orders of susceptibility are neglected due to symmetry. $\chi^{(3)}$ will be the lowest order of susceptibility for nonlinear response and it dominates over the rest of higher orders terms. Table 2.1 below lists the $\chi^{(3)}$ values of noble gas that are commonly used in our studies [51]. The heavier the gas atom, the higher the $\chi^{(3)}$ value.

Table 2. 1 $\chi^{(3)}$ values of major types of noble gas [51]

Gas species	$\chi^{(3)}$ ($10^{-28} \text{ m}^2/\text{V}^2$)
Helium	3.43
Neon	6.17
Argon	80.61
Krypton	219.52
Xenon	645.53

The nonlinear polarization due to third order susceptibility is a process involving interaction among four waves, which leads to various nonlinear phenomenon such as Kerr effect or third harmonic generation. In our studies, we mainly focus on Kerr effect induced nonlinearity. Kerr effect refers to the intensity dependence of effective refractive index. For linearly polarized light, the Kerr effect induced nonlinear polarization is expressed as [52]:

$$\mathbf{P}_{nl}(t) = \frac{3}{4}\epsilon_0\chi^{(3)}|\mathbf{E}(t)|^2\mathbf{E}(t). \quad (2.16)$$

This term alters the refractive index of the media by introducing an intensity-dependent part given as:

$$n = n_0 + n_2 I, \quad (2.17)$$

where n_0 and n_2 are the linear and nonlinear refractive index, respectively. $n_2 = \frac{3\chi^{(3)}}{4c\epsilon_0 n^2}$. $I = \frac{1}{2}c n \epsilon_0 |\mathbf{E}(t)|^2$ is the intensity of the pulse. c is the velocity of light in vacuum defined as: $c = \left(\frac{1}{\epsilon_0 \mu_0}\right)^{\frac{1}{2}}$. In the next sections we list some main nonlinear effects induced by Kerr nonlinearity.

2.3.1 Self-phase modulation (SPM)

It refers to the process where the phase of electrical field is modified due to change of refractive index by Kerr nonlinearity. Under the effect of SPM, the pulse propagation can be described by a simple envelope equation [53]:

$$\frac{\partial A(z,t)}{\partial z} = i \left(\frac{2\pi n_2 I(t)}{\lambda} \right) A(z,t), \quad (2.18)$$

where $A(z,t)$ is the envelope of the pulse, ω is the angular frequency, $I(t)$ is the time-dependent intensity of the pulse. By solving Eq. (2.18) we have:

$$A(z,t) = A(0,t) e^{i\phi_{nl}(z,t)}, \quad (2.19)$$

where $\phi_{nl}(z,t) = \frac{2\pi n_2 I(t)z}{\lambda}$ is the nonlinear phase shift induced by SPM. Eq. (2.19) shows that SPM only changes the phase of the electrical field while keeps the envelope of the pulse invariant along the propagation. The amount of nonlinear phase shift is proportional to both

nonlinearity as well as the distance z . The nonlinear phase shift gives rise to instantaneous frequency shift of the pulse:

$$\Delta\omega(t) = \frac{\partial\phi_{nl}(z,t)}{\partial t} = -\frac{2\pi}{\lambda} n_2 z \frac{dI(t)}{dt}. \quad (2.20)$$

For a pulse with typical Gaussian profile, the instantaneous frequency shift is given as:

$$\Delta\omega(t) = \frac{4\pi I_0 n_2 z t}{\lambda T^2} \exp\left(-\frac{t^2}{T^2}\right). \quad (2.21)$$

Hence there is a downshift of frequency on the leading edge ($t < 0$) of the pulse and upshift on the trailing edge ($t > 0$). The spectral broadening towards both high frequency and low frequency is symmetrical. A positive chirp (the second derivative of the nonlinear phase shift is a positive value), is induced by SPM near the peak of the pulse.

2.3.2 Cross-phase modulation (XPM)

When there are two pulses of different wavelengths co-propagating in a waveguide, each pulse will affect the phase of the other one through Kerr nonlinearity. This is known as cross-phase modulation. The nonlinear phase shift for each pulse is written as:

$$\phi_{nl1}(z) = \frac{\omega n_2 I_1(t) z}{c} + \frac{2\omega n_2 I_2(t) z}{c}, \quad (2.22)$$

$$\phi_{nl2}(z) = \frac{\omega n_2 I_2(t) z}{c} + \frac{2\omega n_2 I_1(t) z}{c}. \quad (2.23)$$

In both Eq. (2.22) and Eq. (2.23), the first term represents the phase shift induced by SPM and the second term represents that induced by XPM. The coefficient 2 for the XPM term indicates the nonlinear phase shift induced by XPM is two times that of the SPM, leading to asymmetrical spectral broadening.

2.3.3 Self-steepening

When considering the frequency dependence of the refractive index for n_2 , the pulse propagation under nonlinear response is given as [54]:

$$\frac{\partial A(t,z)}{\partial z} = i \frac{\omega n_2 I}{c} \left(1 + \tau_{shock} \frac{\partial}{\partial t}\right) |A(t,z)|^2 A(t,z), \quad (2.24)$$

where $\tau_{shock} = 1/\omega_0$. Self-steepening is a higher-order perturbation that relates to the intensity

dependence of group velocity. As a short pulse propagates in medium with positive nonlinear refractive index n_2 , higher intensity peak experiences lower group velocity as compared to the pulse wings. Subsequently, the peak will be caught up by the trailing edge of the pulse. Self-steepening sharpens the slope at trailing edge and enhances frequency blueshift, making the spectrum asymmetric towards the blue side. The extreme case is the formation of an infinitely large slope known as shock wave, which is followed by breaking down of the pulse.

2.3.4 Optical solitons

Optical solitons are formed by the interplay between nonlinearity and dispersion. A pulse propagating in anomalous dispersion regime will experience positive chirp induced by SPM and negative chirp induced by anomalous dispersion. The relative strength of dispersion and nonlinearity is governed by soliton order N defined as:

$$N = \sqrt{\frac{L_d}{L_{nl}}} = \sqrt{\frac{t_0^2 \gamma P_0}{|\beta_2|}}, \quad (2.25)$$

where $L_d = \frac{t_0^2}{|\beta_2|}$ is the dispersion length, t_0 is the initial duration of the pulse at $1/e^2$ width. L_d is the characteristic length for group velocity dispersion to be important. $L_{nl} = \frac{1}{\gamma P_0}$ is the nonlinear length where P_0 is the initial peak power and γ is the nonlinear coefficient (unit: W^{-1}/m) given as [50]:

$$\gamma = n_2 \omega_0 / c A_{eff}, \quad (2.26)$$

where A_{eff} is effective mode area written as:

$$A_{eff} = \frac{\left(\iint_{-\infty}^{\infty} |F(x,y)|^2 dx dy \right)^2}{\iint_{-\infty}^{\infty} |F(x,y)|^4 dx dy}, \quad (2.27)$$

$F(x,y)$ is the transverse mode profile. L_{nl} defines the length required for a nonlinear phase shift of 1 rad.

When the nonlinearity-induced positive chirp and dispersion-induced negative chirp compensate each other, the pulse will propagate as a time-invariant wave packet (when $N =$

1) or evolves periodically (when $N > 1$). The first form of pulse is known as fundamental soliton and the latter form is referred to as higher-order soliton.

2.3.5 Dispersive wave (DW)

When a broadband pulse propagates in a medium with weak anomalous dispersion, it is susceptible to strong perturbation by higher order dispersion such as β_3 , which results in soliton fission [2]. The soliton will shed part of its energy into the normal dispersion regime and forming a dispersive wave. The wavelength of dispersive wave is determined by the phase matching condition [50, 55]:

$$\Delta\beta_{DW}(\omega) = \beta(\omega) - \left[\beta(\omega_{sol}) + \beta_1(\omega_{sol})(\omega - \omega_{sol}) + \frac{\gamma P_{sol}}{2} \right], \quad (2.28)$$

where ω_{sol} is the angular frequency of soliton and P_{sol} is the peak power of soliton. When the phase of a soliton in anomalous dispersion region matches with that of a non-solitonic (dispersive) wave in the normal dispersion region, energy will be transferred from the soliton to that dispersive wave, provided the soliton spectrum sufficiently overlaps with the phase matching point.

2.3.6 Four-wave mixing (FWM)

Four-wave mixing is a Kerr-induced nonlinear effect where two photons with frequency ω_1 and ω_2 convert to another pair of photons with frequency ω_3 and ω_4 . When $\omega_1 = \omega_2$, it is known as degenerate four-wave mixing. FWM process only occurs when energy and momentum conservation are satisfied simultaneously [50]. For the case of degenerate four-wave mixing it is written as:

$$2\omega_p = \omega_i + \omega_s, \quad (2.29)$$

$$2\beta(\omega_p) = \beta(\omega_i) + \beta(\omega_s) + \Gamma_{NL}, \quad (2.30)$$

where ω_p , ω_i and ω_s are the angular frequency for the pump, idle and signal. $\Gamma_{NL} = 2\gamma P$ is the Kerr-induced nonlinear correction term and P is the peak power of the pump.

2.4 Different pulse propagation models

2.4.1 Nonlinear Schrödinger Equation (NLSE)

If we assume the pulse is linearly polarized along the z axis and rescale the variables in Eq. (2.10), after simplification [56], we can get the Nonlinear Schrödinger Equation (NLSE) given as:

$$\frac{\partial A(z,t)}{\partial z} = -i \frac{\beta_2}{2} \frac{\partial^2 A(z,t)}{\partial t^2} + i\gamma |A(z,t)|^2 A(z,t), \quad (2.31)$$

where $A(z, t)$ is the complex envelope of the pulse moving in a retarded time frame $t = T - \beta_1 z$, where T is the absolute time frame (non-moving time frame) and β_1 is the reciprocal of group velocity v_g . The first term on the right hand of Eq. (2.31) is responsible for group velocity dispersion and the second term describes the self-phase modulation induced by Kerr nonlinearity.

NLSE is developed based on the following assumptions:

1. The pulse envelope is slowly varying on the time scale of a single cycle of the carrier.
2. The electric field is assumed to be quasi-monochromatic with central angular frequency ω_0 .
3. Nonlinear polarization is treated as weak perturbation.
4. Electric field keep the same polarization orientation along the propagation.

Since it does not include loss, higher-order linear and nonlinear effects such as higher-order dispersion, self-steepening, Raman effect etc., it is only applicable to energy-conserved pulse propagation without involving high intensity. NLSE has shown excellent agreement with experimental results related to generation of optical soliton in solid-core fibers [50, 57] and picosecond pulse compression in gas-filled hollow-core fibers [58].

2.4.2 Generalized Nonlinear Schrödinger Equation (GNLSE)

Given the limitation by NLSE, a more complete description of pulses propagation in a

nonlinear dispersive medium is made by the Generalized Nonlinear Schrodinger Equation. It takes account of the loss, higher-order linear and nonlinear effects and can accurately model the propagation of high intensity ultrashort pulses down to few cycle. It has shown great success in predicting novel phenomenon, with one representative example of supercontinuum generation, in different types of optical waveguides including solid-core photonic crystal fibers, gas-filled hollow-core fibers and fiber tapers [19, 55, 59].

More details on the derivation are described in [50] and the commonly adopted form of GNLSE is given as:

$$\begin{aligned} \frac{\partial A(z,t)}{\partial z} = & i \sum_{n \geq 2} \frac{\beta_n}{n!} \left(i \frac{\partial}{\partial t} \right)^n A(z,t) - \frac{\alpha A(z,t)}{2} + \\ & i\gamma \left(1 + \frac{i}{\omega_0} \frac{\partial}{\partial t} \right) ((1 - f_R)|A(z,t)|^2 A(z,t) + f_R g(z,t, A(z,t))), \end{aligned} \quad (2.32)$$

where $A(z,t)$ is the complex envelope of the pulse at distance z moving in a reference time frame travels at group velocity $v_g = 1/\beta_1$. β_n is the n th order dispersion coefficient obtained by expanding the propagation constant into Taylor-series around carrier angular frequency ω_0 .

Inclusion of the higher-order-dispersion terms $i \sum_{n \geq 3} \frac{\beta_n}{n!} \left(i \frac{\partial}{\partial t} \right)^n A(z,t)$ is necessary when dealing with wide spectrum bandwidth and when pulse propagates near zero-dispersion wavelength. α is the fiber loss. For most cases, the nonlinear coefficient γ is evaluated at pump frequency ω_0 , unless in some special cases as we shall see in Chapter 6. $\frac{i}{\omega_0} \frac{\partial}{\partial t}$ is the shock derivative term that is responsible for self-steepening effect. f_R is the fraction of Raman contribution. The term $(1 - f_R)|A(z,t)|^2 A(z,t)$ represents the self-phase modulation induced by instantaneous Kerr effect and $f_R g(z,t, A(z,t))$ corresponds to the delayed Raman effect.

2.4.3 Multimode Generalized Nonlinear Schrödinger Equation (MMGNLSE)

GNLSE can be further extended to incorporate intermodal linear and nonlinear effects that

commonly and inevitably exist in large core fibers supporting multimode guidance. Derivation of a general form of MMGNLSE is explicitly elucidated in Ref [60] and later a simplified version is presented in [61] given as:

$$\begin{aligned} \frac{\partial A_p}{\partial z} = & i(\beta_0^{(p)} - \beta_0^{(1)})A_p + (\beta_1^{(p)} - \beta_1^{(1)})\frac{\partial A_p}{\partial t} + i\sum_{n \geq 2} \frac{\beta_n^{(p)}}{n!} \left(i\frac{\partial}{\partial t}\right)^n A_p \\ & + i\frac{n_2\omega_0}{c} \left(1 + \frac{i}{\omega_0}\frac{\partial}{\partial t}\right) \sum_{l,m,n} \{ (1 - f_R) S_{plmn}^K A_l A_m A_n^* + f_R S_{plmn}^R A_l [h * A_m A_n^*] \}, \end{aligned} \quad (2.33)$$

where A_p is the complex envelope in mode p , $\beta_n^{(p)}$ is the n th order dispersion coefficient at mode p . The second line includes all the intra-modal and intermodal nonlinear terms induced by Kerr and Raman effect. h is the Raman response function. The convolutional term can be written as:

$$[h * A_m A_n^*](z, t) = \int h(t') A_m(z, t - t') A_n^*(z, t - t') dt'. \quad (2.34)$$

Note that the nonlinear coefficient γ in Eq. (2.32) is replaced by $\frac{n_2\omega_0}{c}$ with association of overlap integral terms denoted by S_{plmn}^K and S_{plmn}^R , which describe the overlap factor that is related to Kerr and Raman effect, respectively. For modes with arbitrary polarization, S_{plmn}^K and S_{plmn}^R are given as:

$$\begin{aligned} S_{plmn}^K = & \frac{2 \int dxdy [F_p^* \cdot F_l] [F_m \cdot F_n^*]}{3 \left[\int dxdy |F_p|^2 \int dxdy |F_l|^2 \int dxdy |F_m|^2 \int dxdy |F_n|^2 \right]^{\frac{1}{2}}} \\ & + \frac{\int dxdy [F_p^* \cdot F_n^*] [F_m \cdot F_l]}{3 \left[\int dxdy |F_p|^2 \int dxdy |F_l|^2 \int dxdy |F_m|^2 \int dxdy |F_n|^2 \right]^{\frac{1}{2}}}, \end{aligned} \quad (2.35)$$

$$S_{plmn}^R = \frac{\int dxdy [F_p^* \cdot F_l] [F_m \cdot F_n^*]}{\left[\int dxdy |F_p|^2 \int dxdy |F_l|^2 \int dxdy |F_m|^2 \int dxdy |F_n|^2 \right]^{\frac{1}{2}}}, \quad (2.36)$$

where $F_q (q = p, l, m, n)$ is the mode function of corresponding mode q in vector form. For linearly polarized modes, Eq. (2.35) and Eq. (2.36) can be further simplified into a scalar form:

$$S_{plmn}^K = S_{plmn}^R = \frac{\int dxdy F_p F_l F_m F_n}{\left[\int dxdy F_p^2 \int dxdy F_l^2 \int dxdy F_m^2 \int dxdy F_n^2 \right]^{\frac{1}{2}}}. \quad (2.37)$$

2.4.4 Unidirectional Pulse Propagation Equation (UPPE)

UPPE is the carrier-resolved field propagation equation by assuming pulse propagates in the forward direction only. Derivation detail of UPPE model is represented in Ref. [62, 63]. It is more efficient than GNLSE when third harmonic term is included. Besides, field-dependent photoionization effect can be accurately incorporated into UPPE model. It has shown great success in modeling ultrafast pulse propagation dynamics in gas-filled hollow-core waveguides [24, 31, 64]. One of the commonly adopted form of UPPE is given as:

$$\frac{\partial E(z, \omega)}{\partial z} = i \left(\beta(\omega) - \frac{\omega}{v} + \frac{i\alpha}{2} \right) E(z, \omega) + i \frac{\omega^2 \mu_0}{2\beta(\omega)} \mathcal{F} \{ \epsilon_0 \chi^{(3)} E(z, t)^3 \} - \frac{\omega \mu_0}{2\beta(\omega)} \mathcal{F} \left\{ \frac{\partial \rho(z, t)}{\partial t} \frac{I_P}{E(z, t)} + \frac{e^2}{m_e} \int_{-\infty}^t \rho(z, t') E(z, t') dt' \right\}, \quad (2.38)$$

where $E(z, \omega)$ is the carrier-resolved optical field in the frequency domain. $\beta(\omega)$ denotes the full linear dispersion and it is contributed by both waveguide and filling gas. The last term represents plasma effect where I_P is the first ionization energy, e is the electron charge and m_e is the electron mass. $\rho(z, t)$ is the local free-electron density calculated based on the Ammosov, Delone, and Krainov (ADK) model [65]. Raman effect can be neglected when considering noble gases.

2.5 Gas-filled hollow-core waveguides

In this thesis, we mainly focus on two different types of hollow-core waveguides: hollow capillaries and anti-resonant hollow-core fibers (AR-HCFs).

2.5.1 Capillaries

Ever since 1970s, capillaries have been commonly used for nonlinear-optics based applications [8, 9]. They are the simplest hollow-core waveguides consisting of only a thick tube as shown in Fig. 2.1:

Figure 2. 1 Cross-section of Capillary

Light is guided in capillaries based on partial internal reflection and hence the loss is very high. The capillary loss $\alpha \propto \lambda^2/a^3$, where λ is pulse wavelength and a is the core radius. For efficient delivery of pulses, a large core diameter greater than $100 \mu m$ is always considered in the applications. Consequently, large core diameter results in two constraints: weak nonlinearity and weak anomalous dispersion. The weak nonlinearity can be counteracted by pumping at high pulse energy. Stronger anomalous dispersion can be achieved by pumping in the higher-order modes or at longer wavelength [16, 66, 67]. Capillaries are also sensitive to bending-induced loss. This can be eliminated by stretching a capillary to keep it straight, however the difficulty increases with the length of capillary [15].

Capillaries are intrinsic multimode waveguides. Due to the lossy guidance, only leaky modes are supported. The azimuthal and radial components of the transverse mode profile expressed in cylindrical coordinates are given as [68]:

$$E_{nm}^{\theta}(r, \theta) = J_{n-1}\left(\frac{\mu_{nm}r}{a}\right) \cos(n\theta), \quad (2.39)$$

$$E_{nm}^r(r, \theta) = J_{n-1}\left(\frac{\mu_{nm}r}{a}\right) \sin(n\theta). \quad (2.40)$$

2.5.2 Anti-resonant hollow-core fibers (AR-HCFs)

Negative curvature anti-resonant hollow-core fibers (AR-HCF) have gained growing popularity ever since 2011 [17, 18] and it is widely applied in the field of ultrafast optics [23, 69, 70]. As compared to capillaries, anti-resonant hollow-core fibers can be scaled down to

much smaller dimension and yet keep a moderate loss that is far below that of the capillary with equivalent size [71]. One of the commonly used AR-HCFs in recent years is the single-ring negative curvature gas-filled hollow-core fiber as shown in Fig 2.2(a). Other types of design such as Kagome-type fiber [72], nested-element fiber [73] and ice-cream-cone fiber [18] are shown in Fig. 2.2(b), (c) and (d) respectively.

Figure 2. 2 Anti-resonant hollow-core fibers with different structures. Courtesy of Wonkeun Chang.

The guiding mechanism of AR-HCFs can be described by the anti-resonant reflecting optical waveguide (ARROW) model [3] where the high-index glass web of cladding tubes can be treated as a Fabry-Perot resonator with cavity length equal to the thickness of the cladding wall. According to ARROW model, light will be leaked out when its' wavelength falls within the resonance bands. The locations of different resonance wavelengths are given by:

$$\lambda_r = \frac{2d}{m} \sqrt{n_{mat}^2 - 1}, \quad (2.41)$$

where d is the cladding wall thickness, $m = 1, 2, 3 \dots$ represents the resonance order. n_{mat} is the refractive index of cladding material, in our case is fused silica. Wavelengths that differ from resonance wavelengths λ_r can be tightly confined in the core area through strong reflection. Those fall within the resonance bands will be leaked out, resulting in sharp transmission dips at discrete resonance bands, as illustrated in Fig. 2.3 (a) and 2.3 (b). Alternatively, the guidance of AR-HCFs can also be described by inhibited-coupling theory [4, 5]. High loss is induced at wavelengths where the effective index of the core mode and the cladding mode are matched with sufficient mode overlap, as shown in Fig. 2.3 (c).

Figure 2. 3 Illustration of guiding mechanism in AR-HCFs. Courtesy of Wonkeun Chang

Figure 2.4 shows a typical experimental setup for a gas-filled hollow-core fiber. Gas chamber is connected on each end of fiber and gas will flow from one end to the other end until fiber is fully filled with pure injecting gas. Pressure can be flexibly adjusted outside gas chambers. A gas-filled hollow-core waveguide is a nonlinear dispersive medium that supports various of linear and nonlinear optical effects subject to different parameters of pulse, filling gas and waveguide.

Figure 2. 4 Experimental setup for a gas-filling hollow-core fiber. Courtesy of Wonkeun Chang

Chapter 3: Tuning the soliton-effect compression via stretching of the pump

3.1 Introduction

Generation of ultrafast light sources has been a hot topic in recent years. They can be widely used in different applications. For example, they can be used as probes for investigating ultrafast phenomena [74], as data bits in optical communication systems [75, 76] and in high-precision metrology [77]. The energy in an ultrashort pulse is confined within a short time interval, making it possible to reach extremely high intensities. This attractive feature can be exploited in micromachining [78], precision surgery [79] as well as other industrial and medical applications.

Ultrashort pulses can be obtained by various compression methods. One commonly used technique to compress the pulse below its transform-limit duration is the soliton-effect compression [42, 43]. When the pulse is launched in the anomalously dispersive nonlinear medium with soliton order greater than 1, it can be highly compressed after a predetermined propagation distance. Pulse compression can be achieved in a single stage with this approach,, which is advantageous over the two-stage compression technique, where separate stages for the spectral broadening and chirp compensation [41] are required.

Compression factor and compression quality are important parameters to measure the performance of soliton-effect compression. One option to improve these parameters is by modifying the pulse input. In fact, the exact pump pulse amplitude and phase profiles that give rise to the shortest possible pulse for given dispersion and nonlinear parameters have been worked out analytically in the early 90s [80]. However, the proposed initial conditions are difficult to achieve in experiments. A more practical and simpler approach is to linearly stretch the pulse in the time domain using dispersive elements. In fact, there are number of past studies

about effect of chirp on pulse compression [81-87]. Results in these studies show that only positive chirp can improve the pulse compression while negative chirp is always detrimental to the compression performance.

In this chapter, we numerically investigate the effect of stretching-induced initial chirp on the soliton-effect pulse compression. We find the compression can be improved by both the positive and negative initial chirps through different mechanisms. We show, for the first time, that even the negative initial chirp can lead to enhanced compression. As we shall see, different mechanisms are responsible for the enhancement in the two cases. With the positive initial chirp, the amount of stretching that yields the best achievable compression decreases with increasing soliton order while for the optimal negative chirp it is just the other way around. By varying the amount of stretch-induced chirp in the pump, it is possible to tune the compression length over a wide range, which can even fall below that of the transform-limited pulse within a small range of positive initial chirp. Interestingly, we find the stretching factor that gives the shortest output duration extends the compression length by 1.23 times for any given soliton number. Our study shows that the soliton-effect compression can be readily controlled and optimised by placing a variable stretching element beforehand. In particular, it is beneficial for experimental setups where the length of the compressor cannot be readily modified, such as for gas-filled hollow-core waveguides setups.

3.2 Modelling the propagation of higher-order soliton with initial chirp

For the numerical study, we use the focusing nonlinear Schrödinger equation (NLSE) given by Eq.(2.3.1), which models the optical pulse propagation in anomalously dispersive nonlinear medium. In its normalized and dimensionless form, the equation is given by [50]:

$$\frac{\partial \psi}{\partial z} = i \frac{1}{2} \frac{\partial^2 \psi}{\partial t^2} + i |\psi|^2 \psi, \quad (3.1)$$

where $\psi(z, t)$ is the slowly varying envelope of the optical field, z is the propagation variable and t is the transverse variable. On the right-hand side, the first term describes the anomalous

dispersion, and the last term is responsible for the self-phase modulation (SPM).

Eq. (3.1) has a well-known soliton solution, i.e., $\psi_0 = N \text{sech}(t)$, Where $N = 1, 2, 3, \dots$ is the soliton number. A higher-order soliton ($N > 1$) undergoes a periodic evolution. As exemplified in Figs. 3.2(a) and 3.2(b) for $N = 2$, it first undergoes a temporal compression and spectral broadening until it reaches the maximum compression point. The degree of the compression depends largely on N with a higher N leading to a stronger compression [88].

We now look at the case where the chirp is applied to the pump through the pulse stretching. Mathematically, this can be implemented in the soliton's initial condition in the following manner:

$$\psi_0 = \mathcal{F}^{-1} \left\{ N \sqrt{\frac{\pi}{2}} \text{sech} \left(\frac{\pi}{2} \Delta\omega \right) \exp \left(\frac{i}{2} C \Delta\omega^2 \right) \right\}, \quad (3.2)$$

where C is the stretching factor that introduces the frequency chirp. Experimentally, this is the group-delay dispersion of the stretching element. $\Delta\omega$ is the angular frequency detuning from the carrier, and $\mathcal{F}^{-1}$ denotes the inverse Fourier transformation. Eq. (3.2) introduces the frequency chirp without affecting the spectral profile.

For better visualization of the stretching-induced chirp, we use the frequency-resolved optical gating (FROG) spectrograms calculated based on [89, 90] :

$$S(t, \omega) = \left| \int_{-\infty}^{+\infty} dt' A_{gate}(t' - t) A(t') e^{-i\omega t} \right|^2, \quad (3.3)$$

where A_{gate} and A are the amplitude of the gate pulse and propagating pulse, respectively.

Fig 3.1(a) and 3.1(b) shows the spectrogram of an $N = 2$ soliton at initial position with stretching factor of $C = 0$ and $C = 1.43$ respectively. We can see that applying stretching factor redistributes the spectral components in temporal domain and introduces a linear chirp across the pulse, where the slope is determined by the sign and magnitude of the stretching factor.

Figure 3. 1 Spectrogram of $N = 2$ soliton with (a) $C = 0$ and (b) $C = 1.43$ at $z = 0$

This can be realized experimentally by employing linearly dispersive optical elements such as chirped mirrors or a grating pair on an un-chirped pulse. Alternatively, we can deliberately under- or over-compensate the chirp by tuning the pulse compression stage that is often in the final stage of high-power ultrafast laser systems.

3.3 Numerical implementation

Since we need to carry out intensive simulations by scanning through a wide range of chirp parameters induced by linear stretching, it is important to solve the NLSE in an efficient manner with high accuracy. In this study, we implement the Fourth-Order Runge–Kutta in the Interaction Picture Method (RK4IP) [91] to numerically solve the NLSE. For convenience purpose, we can rewrite Eq. (3.1) in the following form:

$$\frac{\partial \psi}{\partial z} = (\hat{D} + \hat{N})\psi, \quad (3.4)$$

where $\hat{D} = i\frac{1}{2}\frac{\partial^2}{\partial t^2}$ and $\hat{N} = i|\psi|^2$ are the dispersion and nonlinear operator, respectively.

We transfer Eq. (3.4) into an interaction picture expressed as:

$$\psi_i = \exp\left((z' - z)\hat{D}\right)\psi, \quad (3.5)$$

where z' is the length difference between the interaction picture and normal picture. By taking derivative of Eq. (3.5), we get:

$$\frac{\partial \psi_i}{\partial z} = \hat{N}_i \psi_i, \quad (3.6)$$

and

$$\hat{N}_i = \exp((z' - z)\hat{D}) \hat{N} \exp((z - z')\hat{D}). \quad (3.7)$$

We choose z' to be at midpoint of the propagation step h , namely: $z' = z + 0.5h$, by doing so we can effectively reduce the number of Fast Fourier Transform operations involved in evaluation of nonlinear and linear operators. The solution of $\psi(z, T)$ moving forward with one step size to $\psi(z + h, T)$ can be obtained with the algorithm given below [92]:

$$\psi_i = \exp(0.5h\hat{D}) \psi(z, T), \quad (3.8)$$

$$k_1 = \exp(0.5h\hat{D}) [h\hat{N}\psi(z, T)] \psi(z, T), \quad (3.9)$$

$$k_2 = h\hat{N}(\psi_i + 0.5k_1) [\psi_i + 0.5k_1], \quad (3.10)$$

$$k_3 = h\hat{N}(\psi_i + 0.5k_2) [\psi_i + 0.5k_2], \quad (3.11)$$

$$k_4 = h\hat{N} \left(\exp(0.5h\hat{D})(\psi_i + k_3) \right) \exp(0.5h\hat{D}) [\psi_i + k_3], \quad (3.12)$$

$$\psi(z + h, T) = \exp(0.5h\hat{D}) \left[\psi_i + \frac{1}{6}k_1 + \frac{1}{3}k_2 + \frac{1}{3}k_3 \right] + \frac{1}{6}k_4. \quad (3.13)$$

The RK4IP method only requires 8 Fast Fourier Transform (FFT) operations per steps with error in the order of $O(h^5)$. It significantly improves the calculation efficiency as compared to the conventional fourth-order Runge-Kutta method, which requires 16 FFT operations for each step.

3.4 Simulation results and discussion

Fig. 3.2 (a) and 3.2 (b) shows the temporal and spectral evolutions of a higher-order soliton ($N = 2$) in the focusing nonlinear Schrödinger equation. Fig. 3.2 (c) and 3.2 (d) show those with a time-stretched pump where $C = 1.43$. The maximum compression points, L_0 for 3.2 (a) and 3.2 (b), and L for 3.2 (c) and 3.2 (d), are marked with black horizontal-dashed lines. Fig. 3.2

(e) shows the comparison between the maximally compressed pulses achieved with the un-chirped (blue-solid line) and time-stretched (red-solid line) initial conditions. The respective input pulses are shown in dashed lines. Fig. 3.2 (f) shows the corresponding pulse spectra at the maximum compression points obtained using the un-chirped (blue-solid line) and time-stretched (red-solid line) initial conditions. The spectral profiles of the two input pulses are identical (black-dashed line). Fig. 3.2 (e) and 3.2 (f) indicate that a positive chirp increases the initial pulse duration and reduces the peak intensity without altering the spectrum.

Figure 3. 2 (a) Temporal and (b) spectral evolutions of a higher-order soliton ($N = 2$) in the focusing NLSE. (c) and (d) show those with a time-stretched pump where $C = 1.43$. Black dashed lines indicate the maximum compression point. (e) maximally compressed pulses achieved with the un-chirped (blue-solid line) and time-stretched (orange-solid line) initial conditions. Dashed lines indicate the input pulse. (f) pulse spectra at the maximum compression points obtained using the un-chirped (blue-solid line) and time-stretched (orange-solid line) initial conditions. Black-dashed line shows the input spectrum for both cases.

While the time-stretched initial condition appears to undergo roughly a similar propagation dynamic as its un-chirped pump counterpart, the details such as the compression length, as well as the duration, peak power and bandwidth of the pulse at the maximum compression point are different. Even though the input pulse is temporally stretched for the case of $C = 1.43$, as seen in Fig. 3.2 (e) dashed lines, it is still able to achieve improved compression as compared to the unchirped counterpart.

Let us take a step further and study the influence of stretching the pump on the soliton-effect compression over a wide range of C spanning both positive and negative values. Our numerical results for N from 2 to 8 are presented in Fig. 3.3. Namely, Fig. 3.3 (a) plots the relative compressed pulse width, τ/τ_0 , as a function of C , where τ and τ_0 are the full-width at half-maximum (FWHM) durations of the maximally compressed pulses that are obtained for a given C and $C = 0$, respectively. This ratio quantifies the impact of the pump stretching on the soliton-effect compression in a way that a value less than 1 means improvement and that greater than 1 signifies otherwise. Similarly, the relative compressed peak power, p/p_0 , presented in Fig. 3.3 (b) is another measure of the effectiveness of the linear stretching in the soliton-effect compression. Here, p is the peak power of the maximally compressed pulse for a given C , i.e., the maximum value of $|\psi|^2$ at the compression length L , and p_0 is that for the transform-limited initial condition. Hence, in this case, p/p_0 greater than 1 implies a better compression and vice versa. As for the effect of the pump stretching on the compression length, the relative compression length, L/L_0 , is plotted as a function of C in Fig. 3.3 (c), where L is the compression length for the input for a given C and L_0 is that for the un-chirped input. It expresses the fractional increase or decrease in the compression length caused by the stretching. The arrows show the direction of increasing N from 2 to 8. The horizontal black-dashed lines are drawn at 1 to indicate the reference points for the respective ratios. Each of the vertical black-dotted line marks the optimal positive chirp, C_p , for the corresponding soliton order N .

Note that for all N , the relative compression length is ~ 1.23 at C_p . Table 3.1 lists the compression length L_0 , shortest pulse duration τ_0 and maximum peak power p_0 after compression for unchirped soliton with $N = 2$ to 10.

Figure 3. 3 (a) Relative compressed pulse width, (b) relative compressed peak power, (c) relative compression length, and (d) relative compressed pulse quality factor as functions of C . The arrows show the direction of increasing N from 2 to 8. The horizontal black-dashed lines are drawn at 1 to indicate the reference points for the respective ratios. Each of the vertical black-dotted line marks C_p for the corresponding N .

Table 3. 1 Compression parameters of unchirped higher order solitons

N	L_0	τ_0	p_0
2	0.785	0.398	4
3	0.372	0.211	6.194
4	0.235	0.141	7.755
5	0.169	0.105	8.916
6	0.131	0.084	9.815
7	0.106	0.069	10.534
8	0.089	0.059	11.122
9	0.077	0.051	11.614
10	0.067	0.045	12.032

One distinct but unwelcome characteristic of the soliton-effect compression is the appearance of pedestals as observed in Fig. 3.2 (e). For instance, while a higher N leads to a stronger compression, it also results in having a bigger fraction of the pulse energy residing in the pedestals, degrading the overall quality of the compressed pulse. We also examine the effect of the pump stretching on the quality of the output by plotting the relative compressed pulse quality factor, η/η_0 , in Fig. 3.3(d). η is the fractional energy within the main peak of the compressed pulse as illustrated with the blue-shaded region in the example pulse in the inset of Fig. 3.3(d) for a given C , and η_0 is that of the un-stretched pump. Therefore, when η/η_0 is greater than 1, the quality of the compressed pulse improves with the stretching, while the output pulse quality becomes poorer with the stretching if it is less than 1.

We pick up several interesting observations in Fig. 3.3. Firstly, for each N , there exists a fixed positive stretching factor, C_p , that leads to the best-achievable compression as indicated by the vertical-dotted lines in Fig. 2.3. Its magnitude reduces with increasing N . This is in line with

findings reported in previous studies [84, 86]. The degree of the improvement lessens as N increases with the best possible scenario in Fig. 3.3 occurring at $N = 2$ and $C = 1.33$. In this case, the compressed pulse is expected to have 10% shorter duration and 12% higher peak power compared with when the transform-limited input pulse is used.

Secondly, we reveal that an initial negative chirp can also give rise to a better soliton-effect compression. This is an important new discovery that has been overlooked in the past. In both Figs. 3.3 (a) and 3.3 (b), there exists another local extremum in $C < 0$, such that there is a specific value, C_n , that can bring about the best possible outcome in the negative range of C . As N is increased, C_n increases in its magnitude and the compression at C_n becomes better, which are in stark contrast to C_p , indicating that the mechanisms responsible for the compression enhancement are different at C_p and C_n . We also observe from Fig. 3.3(d) that these stronger compressions are realized in conjunction with the improvements in the compressed pulse quality factor by a small margin.

To analyze these phenomena, let us first remind ourselves that the soliton number, N , characterizes the relative strength between the effect of the anomalous dispersion and SPM, i.e. the nonlinear effect becomes comparably stronger as N is increased, and vice versa. Therefore, the diminishing value and effect of C_p for increasing N means that the dispersion is acting dominantly in concert with the positive initial chirp to facilitate the improved compression. On the other hand, the better compression with the negative initial chirp is due to its combined effect with the SPM.

We first examine the enhanced compression in the positive range of C by comparing the evolutions of $N = 2$ pumps with $C = 0$ and two other positive stretching factors, one at $C = 0.37$ where the compression length is the shortest, and the other at $C = C_p = 1.33$. Namely, Figs. 3.4(a), 3.4(b) and 3.4(c) present the spectral width (root-mean-squared), peak power and frequency chirp at the pulse center, respectively, along z for the three different initial

conditions. The vertical gray-dotted lines indicate the maximum compression points for the three cases. The spectral width translates to the strength of the negative dispersion, dictating the degree of the down-chirp induced, while the peak power relates to the extent of SPM, affecting the size of the up-chirp that develops in the pulse center. Therefore, the change in frequency chirp as illustrated in Fig. 3.4(c) is determined by the relative strength between the effect of the dispersion and SPM.

Figure 3. 4 Evolutions of the (a) root-mean-squared (RMS) spectral width, (b) peak power and (c) frequency chirp at the pulse center along z for $N = 2$, $C = 0, 0.37$ and 1.33 .

In the early stage of the soliton-effect compression, the effect of SPM dominates that of the

dispersion. This induces a net positive frequency chirp across the pulse center with its magnitude increasing along z as shown in Fig. 3.4(c). As the spectrum becomes broader further along, the effect of negative dispersion gets stronger, overtaking that of SPM in time and decreasing the chirp. The maximum compression is achieved when the accumulated frequency chirp is fully compensated, crossing the zero, i.e., at vertical gray-dotted lines in Fig. 3.4. For $N = 2$ and $C = 0$, this happens at $z = \pi / 4$.

In the case of $C = 1.33$, the accumulation of the up-chirp at the beginning is slower due mainly to the reduced peak power of the time-stretched pump. Moreover, the peak power increases also at a slower rate in the early phase. This is because the effect of anomalous dispersion—the main driver that temporally compresses the SPM-broadened pulse—is weakened by the presence of the initial positive chirp. Despite the setback, the pulse continues to compress temporally and broaden spectrally, and this process eventually accelerates later in the evolution as the peak power increases and the anomalous dispersion counteracts the initial chirp. The maximum compression point, nonetheless, is delayed due to the slow pace of the evolution in the initial stage, increasing the interaction length to accumulate larger total nonlinear phase shift. This leads to a wider spectral width that can support a shorter pulse. The optimum positive stretching factor C_p is therefore one that gives the biggest increase in the total accumulated nonlinear phase shift as a result of delaying the compression length. Figure 3.3 (c) shows that this always happens when the compression length is extended by approximately 1.23 times the compression length of the un-chirped pump. From the practical point of view, it means we can always optimize the compression to its full capacity by using this extended compression length and tuning the pump pulse stretcher to obtain optimal chirp. Figure 3.5 shows how the positive optimal chirp and negative optimal chirp change with soliton order.

Figure 3. 5 Optimal positive and negative chirp at different soliton order N

We fit the curves with polynomial functions up to 5th order and get the expression of optimal positive chirp C_{op} and optimal negative chirp C_{on} , given as:

$$C_{op} = -0.0003N^5 + 0.0111N^4 - 0.1526N^3 + 1.0336N^2 - 3.5126N + 5.3667, \quad (3.14)$$

$$C_{on} = -0.0002N^5 + 0.0052N^4 - 0.0689N^3 + 0.4486N^2 - 1.4354N + 1.5387. \quad (3.15)$$

Based on the above approximations, we can estimate the optimal positive chirp and negative chirp for any given soliton order N ranges from 2 to 10.

Another interesting finding in Fig. 3.3 (c) is that within a certain range of positive C , e.g., $0 < C < 0.79$ when $N = 2$, the compression length shortens while the enhancement is still realised. At a relatively small stretching factor, e.g., at $C = 0.37$, the impact of the decreased initial peak power on SPM is marginal. Actually, it is evident from Fig. 3.4 (a) that the broadening of the spectrum when $C = 0.37$ at the beginning is faster than when $C = 0$. This is because the positive stretching factor places the lower frequency components in the leading half and higher frequency components in the trailing half of the pump, which causes the nonlinear phase shift

to induce new frequencies that are further out from the carrier, ultimately leading to more rapid spectral broadening and temporal compression.

The improved compression shown in the negative C range in Figs. 3.3 (a) and 3.3 (b) is being reported for the first time here. At higher N where the enhancement due to the negative initial chirp is stronger, the effect of SPM largely outweighs that of the dispersion. The dominant SPM induces a positive chirp across the central part of the pulse, which flattens quickly the initial negative chirp. In fact, this is related to a well-known SPM-driven spectral narrowing process in the presence of initial negative chirp [93-95]. As an example, we look closely at the case when $N = 8$ and $C = -0.2$.

Figure 3.6(a) shows the instantaneous frequency across the pulse at $z=0$ (blue-solid line) and 0.0045 (red-solid line) for the pump with $N = 8$ and $C = -0.28$. The pump profile is included as a reference (grey-dotted line). We observe that the negative chirp seen at $z = 0$ is mostly compensated at $z = 0.0045$ by the strong effect of SPM. Note that the temporal profile of the chirp-flattened pulse is almost identical to the pump which, we stress again, is the linearly stretched $N = 8$ soliton. Once the initial chirp is flattened, we can consider the propagation from this point onwards as a transform-limited higher-order soliton having a new effective soliton number, N_{eff} , that corresponds to the duration and peak power of the stretched pump. In our example shown in Fig. 3.6(a), the stretched pump has FWHM duration of 1.92 and peak power of 60.3, which gives us N_{eff} of 8.46 that is higher than the original N . The enhanced compression in this case is due to having a higher soliton number that generally leads to a higher compression factor.

Figure 3. 6 (a) Instantaneous frequency across the pulse at $z = 0$ (blue-solid line) and 0.0045 (red-solid line) for the pump with $N = 8$ and $C = -0.28$. The pump profile is included as a reference (grey-dotted line). (b) FWHM duration (solid lines) and peak power (dashed lines) versus z for pump with $N = 8$ and $C = -0.28$ (blue), $N_{\text{eff}} = 8.46$ and $C = 0$ (red), $N = 8$ and $C = 0$ (grey).

Fig 3.6 (b) shows the evolutions of the FWHM duration (solid lines) and peak power (dashed lines) along propagation distance z for negatively-chirped pump (blue) and a chirp-free soliton with the same FWHM duration and peak power (red). The latter is shifted in z by 0.0045 (marked with the vertical-dotted line) which is the amount required to flatten the initial chirp. The grey curves present the evolutions of the two quantities for $C = 0$. While the reduction in pulse duration (3%) is not easily observable in the vertical scale given in Fig. 3.6 (b), we can clearly see the enhanced peak power ($\sim 4\%$) and increased compression length (30%) when compared with those at $C = 0$. Moreover, the same two quantities are plotted in Fig. 3.6 (b) for a transform-limited soliton with the same duration and peak power as the time-stretched pump.

Note that the plot is shifted in z by 0.0045—the amount required to flatten the initial chirp. It closely follows the evolution curves of the time stretched pump. Here, the compression length of the stretched pump is given by the sum of the propagation amount required for flattening the initial negative chirp and the compression length for the transform-limited N_{eff} soliton. We attribute the discrepancy in the compressed duration and peak power to the fact that the chirp is not fully compensated at the spectral compression point, hence its evolution cannot be totally identical to that of the zero-chirp $N = 8.46$ soliton.

While stretching the pump linearly in time brings some degree of improvement to the soliton-effect compression, a more compelling reason to incorporate a linear pulse stretcher before the compression stage, from the experimental standpoint, is its ability to tune the compression length as demonstrated in Fig. 3.3 (c). It implies that the choice of the propagation length is no longer a critical parameter in the soliton-effect compression if the time stretching of the pump can be tuned. This is very useful especially in setups where the length of the compressor cannot be readily modified, e.g., in gas-filled hollow-core fiber compressors [96].

Here, we focus our investigation only on the simplest model that supports the soliton-effect compression, as to gain a clear insight into the effect of stretching without the interference from the higher-order terms. In fact, this simple model still serves as a good guideline in many circumstances, e.g., in picosecond pulse compressors [42], and in atomic-gas-filled hollow-core fiber compressors [2]. The effect of the higher-order terms, such as the higher-order dispersions, self-steepening and Raman scattering on the soliton-effect compression has been thoroughly investigated in many past studies [83, 84, 97]. In Ref. [86], it notes that the inclusion of the higher-order terms has negligible effect on the soliton-effect compression with the time-stretched pump. A detailed study on this topic is left for further investigation.

3.5 Conclusion

We studied the effect of temporally stretching the pump using a linearly dispersive medium on

soliton-effect compression. We reveal, for the first time, that both the positive and negative initial chirps due to the stretching can give rise to improved compression, with two distinct optimum stretching factors on each side. The enhancement achieved with the positive initial chirp is attributed to the extended interaction length that permits accumulation of larger total nonlinear phase shift, while it is the increased effective soliton number that leads to the better compression in the case of the negative initial chirp. In practice, such method is suitable for enhancing pulse compression in the pico-second regime, where the enhancement can be more pronounced than shorter pulse in the femto-second regime. We also show that it is possible to tune the compression length over a wider range by introducing a variable stretching element before the soliton-effect compression, and a small range of positive initial chirp can even lead to a compression length shorter than that of the transform-limited pump. Furthermore, we report that the compression length of the higher-order soliton with optimized positive chirp is always 1.23 times that of the un-chirped case. These findings can effectively relax the strict length requirement of the soliton-effect compression and provide the extra degree of freedom in experiments.

Chapter 4: Soliton-effect compression of MIR supercontinuum in anti-resonant hollow-core fibers filled with pressure gradients

In the previous chapter, we investigate the effect of chirp on soliton-effect compression with consideration of only group velocity dispersion and self-phase modulation. In fact, other effects such as higher-order dispersion also plays an important role in the performance of soliton-effect pulse compression. A key factor that affects the dispersion in gas-filled hollow-core fiber will be gas pressure, which can be flexibly adjusted by controlling the gas chamber at each end of hollow-core fiber. In this chapter, we numerically investigate pulse propagation dynamics with three types of pressurization configurations inside a gas-filled AR-HCF, namely decreasing pressure, constant pressure and increasing pressure.

4.1 Introduction

Recently, there is growing attention on generation of supercontinuum in MIR region, where strong absorption for majority of molecules is located [98]. It has great potential in various applications such as time-resolved molecular spectroscopy [99], remote sensing [100], thermal imaging [101], optical coherence tomography [102] etc. One standard method to generate the supercontinuum is by pumping near the zero-dispersion wavelength in the anomalous dispersion regime and making use of the soliton dynamics for spectral broadening.

On the other hand, effect of pressure gradients in gas-filled hollow waveguides has been studied since 2005 [103]. A gas-filled hollow core fiber is connected by gas chambers at each end, where there is independent control of pressure. This facilitates the controlling of different types of pressure distribution inside the fiber. Ref [103] shows an increasing pressure can be utilized to scale up the peak power for post-compression in normal dispersion regime. In Ref. [45], it demonstrates applying decreasing pressure eliminates the stretching of UV dispersive wave.

Another work in Ref. [44] shows that applying decreasing pressure downgrades the compression performance in the near infrared region when compared with the equivalent constant pressure and increasing pressure. Nevertheless, the effect of pressure gradients has not been investigated for soliton dynamics in the MIR region, which could lead to different results.

We numerically demonstrate the effect of pressure gradients on soliton-effect compression of octave-spanning supercontinuum generated in the MIR region. We find that: applying decreasing pressure in the fundamental mode of an AR-HCF can achieve a higher pulse peak power and enhance the blue edge of the MIR supercontinuum as compared to the equivalent constant pressure and increasing pressure.

4.2 Fiber Parameters

We use a 6-tube single-ring anti-resonant hollow-core fiber as presented in [19]. It has a core diameter $D = 80 \mu\text{m}$ surrounded by 6 cladding tubes, each has a diameter $d = 60 \mu\text{m}$ and wall thickness $t = 0.350 \mu\text{m}$. The gap between two neighbouring tubes is $g = 10 \mu\text{m}$. It has been verified through finite element modeling that this fiber exhibits excellent guidance in the MIR region with low confinement loss and light-silica overlap [19]. With limited input energy, pulse experiences low intensity and thus low nonlinearity due to the large effective mode area. This can be counteracted by filling the fiber with heavy gas that has high nonlinear susceptibility, for instance, xenon. Figure. 4.1 shows the transverse structure of the fiber.

Figure 4. 1 Cross-section of the AR-HCF. $D = 80 \mu\text{m}$ and $d = 60 \mu\text{m}$ denote the core and cladding diameter. $g = 10 \mu\text{m}$ is the gap between the adjacent tubes. $t = 0.35 \mu\text{m}$ is the cladding thickness.

4.3 Modeling of soliton-effect compression and MIR supercontinuum generation in AR-HCF

The pulse propagation is modeled using the single mode unidirectional pulse propagation equation (UPPE) given in Eq. (2.38). Due to the relatively short fiber length, loss is neglected in our simulation. Raman effect induced by both gas and silica are neglected, since we use noble gas and the light-silica overlap is very small. Moreover, the effect of mode-area dispersion [104] can also be safely omitted since we use a large core diameter and pumping far away from resonance bands.

We use an input field with gaussian profile expressed as:

$$E(0, \omega) = \mathcal{F} \left\{ E_0 \exp \left(-\frac{t^2}{2t_0^2} \right) \exp(-i\omega_0 t) \right\}, \quad (4.1)$$

where E_0 is the initial amplitude of electric field, $t_0 = \frac{t_{fwhm}}{2\sqrt{\ln 2}}$ and t_{fwhm} is the duration at full width half maximum of the pulse and ω_0 is the carrier angular frequency.

The linear propagation constant $\beta(\omega)$ for the fundamental core mode is expressed as:

$$\beta(\omega) = \frac{\omega}{c} \left(n_{gas} - \frac{1}{2} \left(\frac{\mu_{01} c}{n_{gas} \omega r_{eff}} \right)^2 \right). \quad (4.2)$$

The first term represents the dispersion from gas. To accurately model the dispersion of xenon from 0 bar to 41 bar, we adopted the wavelength-dependent dispersion profile of Xenon from Ref. [105] and get the xenon density for a given temperature and pressure, using the data from NIST [106]. The second term is responsible for the waveguide contribution obtained with empirical formula [107], where R_{eff} is the effective radius of the fiber, given as:

$$r_{eff} = f_1 r \left[1 - \frac{4f_2 \pi^2 c^2}{\omega^2 r t} \right], \quad (4.3)$$

r is the core radius of the AR-HCF, t is the cladding wall thickness, f_1 and f_2 are the two fitting parameters given as:

$$f_1 = 1.095 \exp \left[\frac{0.097041g}{r} \right], \quad (4.4)$$

$$f_2 = 0.007584n_{tube} \exp\left(\frac{0.76246g}{r}\right) - 0.002n_{tube} + 0.012, \quad (4.5)$$

$g = 2(r + r_{clad}) \sin\left(\frac{\pi}{n_{tube}}\right) - 2r_{clad}$ is the gap between neighbouring tubes and r_{clad} is the radius of cladding tube.

Implementation of the empirical formula improves the accuracy of dispersion especially at long wavelength in the MIR region as compared to the capillary model [107]. It also significantly reduces the time required for calculating dispersion using finite element method (FEM).

4.4 Pressure distribution in gas-filled hollow-core waveguides

Different pressurization configurations can be achieved by controlling the pressure of gas chamber positioned at each end of fiber. To quantitatively determine the gas pressure at any position inside fiber, we assume an invariant volumetric gas flow $q_{pV} = C(p_i - p_o)$ along the fibre length L , where p_i and p_o are the pressure at input and output of fibre, C is the fluidic conductance [108] and $C \propto (p_i + p_o)$. Therefore, the overall volumetric gas flow inside a fiber with length L is equal to that at any given position z , yield [44]:

$$\frac{(p_i - p_o)(p_i + p_o)}{L} = \frac{(p_i - p(z))(p_i + p(z))}{z}. \quad (4.6)$$

By solving Eq. (4.6) we obtained the pressure at any given position z as [21]:

$$p(z) = \sqrt{p_i^2 + \frac{z(p_i^2 - p_o^2)}{L}}. \quad (4.7)$$

We simulate the pulse propagation in the fundamental mode of AR-HCF by pumping at 2460 nm in the weak anomalous dispersion regime, with FWHM duration of 100 fs and input energy of 5 μJ . To ensure sufficient nonlinearity for significant spectral broadening in such a large-core fiber, we fill the fiber with xenon. Three different types of pressure distributions are applied in the simulation, namely increasing pressure from 0 bar to 41 bar, constant pressure of 27.3 bar as well as decreasing pressure from 41 bar to 0 bar at temperature $T = 293 K$. This is to ensure the nonlinear phase shift are approximately matched for all the three cases in a

nonlinearity-dominated medium [45, 103], which is applicable for the parameters given in this chapter.

4.5 Group-velocity dispersion and nonlinear coefficient at different pressure

Figure 4.2(a) and (b) show the group-velocity dispersion (GVD) and nonlinear coefficient γ at different wavelengths. When pressure decreases, both curves shift down and zero-dispersion wavelength (ZDW) shifts towards shorter wavelength.

Figure 4. 2 (a) Group velocity and (b) nonlinear coefficient of xenon-filled AR-HCF at different wavelength and pressures

4.6 Simulation results and discussion

We consider a 100 fs pulse with input energy of 5 μJ and centered at 2460 nm. This pump source can be obtained by a tunable optical parametric amplifier [23]. Figure 4.3 shows the temporal and spectral evolution for pulse propagation under the three types of pressure distribution. Zero-dispersion wavelength (ZDW) is indicated by the black dashed lines. For the case of decreasing pressure, ZDW shifts slowly at beginning and accelerates as it approaches

the output, where the pressure is the minimum. This is due to the nature of pressure distribution inside the gas-filled hollow core fibers. It is the opposite way for the case of increasing pressure. The maximum compression occurs near the end of fiber for all the three cases, namely, $L_{comp} = 12.95 \text{ cm}$ for the case of decreasing pressure and increasing pressure and $L_{comp} = 13 \text{ cm}$ for the case of constant pressure.

Figure 4. 3 Temporal and spectral evolution of $5 \mu\text{J}$, 100 fs pulse centered at 2460 nm for decreasing pressure (first row), constant pressure (second row) and increasing pressure (last row).

Figure 4.4 (a) compares the output spectra for the three cases. The widest spectrum is obtained with constant pressure of 27.3 bar and it spans from 568 nm to 4.82 μm at -40 dB level. Followed by decreasing pressure, with slightly narrower and blue shifted spectra bandwidth from 363 nm to 4.23 μm and enhanced blue edge from 363 nm to 963 nm, as compared to the constant pressure. The worst spectral broadening is obtained with increasing pressure. Due to the low nonlinearity at initial stage, the compression rate is the slowest. The noisy oscillation occurs at around 820 nm in all the three spectra is due to third harmonic of the pump pulse.

Figure 4. 4 (a) Output spectral profiles and (b) dephasing curves at maximum compression point with decreasing pressure (blue curve), constant pressure (red curve) and increasing pressure (yellow curve). Black dashed lines mark the phase-matching wavelengths for DW waves.

The wavelength of soliton-induced dispersive wave (DW) is determined by the phase-matching condition [50]. To accurately predict the position of phase-matched dispersive wave induced by soliton-effect compression in the MIR region, a more precise dephasing rate expression is

required, given as:

$$\beta_{sol} = \beta_0 + \beta_1(\omega - \omega_0) + \Gamma_{NL}, \quad (4.8)$$

$$\Gamma_{NL} = \frac{3}{16} \frac{\varepsilon_0 \mu_0 \omega_0 \omega}{\beta_0} \chi^{(3)} |E_c|^2, \quad (4.9)$$

where Γ_{NL} is the nonlinear correction term, taking into account the dispersion of nonlinear coefficient. $|E_c|^2$ is the field strength of the self-compressed soliton at maximum compression point. For the case of constant pressure at 27.3 bar, we calculated that $\beta_0 = 2.6 \times 10^6 \text{ m}^{-1}$, $\beta_1 = 3.4 \times 10^{-9} \text{ sm}^{-1}$, $\chi^{(3)} = 7.7 \times 10^{-24}$, $|E_c|^2 = 4.18 \times 10^{19} \text{ V/m}$. In the case of decreasing and increasing pressure with longitudinally varying nonlinearity, the nonlinear correction term cannot be explicitly determined at every position. We assume the same nonlinear correction term Γ_{NL} for all the three cases, this is a valid assumption since similar amount of nonlinear phase shift are accumulated approaching the end of the fiber for all the three cases. The linear part is calculated based on the dispersion at corresponding maximum compression point. Therefore, we get the dephasing curves for decreasing pressure, constant pressure and increasing pressures as shown in Fig. 4.4 (b).

The zero-mismatch wavelengths for DW located at 682 nm, 930 nm and 1186 nm, respectively, which agree well with the spectra peaks, as shown in Fig. 4.4 (a) and (b), for the case of decreasing pressure, constant pressure and increasing pressure, respectively. This deduces that one reason causes the enhancement on the blue edge of spectrum for the case of decreasing pressure--with other parameters kept the same, is because decreasing the pressure shifts the dispersive wave to shorter wavelength. The differences of phase-matching mainly come from the linear dispersion with different pressure, which is determined by the position. Figure 4.5 shows the comparison of the temporal profiles for the three cases. The field amplitude is converted to peak power through:

$$P = \frac{1}{2} A_{eff} |E|^2 c \varepsilon_0 n_{eff}, \quad (4.10)$$

where A_{eff} is the effective mode area, E is the output field strength in our case, ε_0 is the

vacuum permittivity, n_{eff} is the linear effective refractive index obtained at the fiber output. Note that the output pressure, and therefore the n_{eff} for the case of decreasing pressure, constant pressure and increasing pressure are different. The highest output peak power is obtained with decreasing pressure (blue curve), reaching 292.3 MW, which is 6.2 times higher than the initial peak power of 46.97 MW. It is 1.63 times higher than constant pressure case (red curve) with a peak power of 178.8 MW. The lowest output peak power is obtained when applying increasing pressure (yellow curve) with a value of 96.7 MW only. The pulse is compressed down to 14.36 fs, 14.5 fs and 36 fs for the case of decreasing pressure, constant pressure and increasing pressure, respectively. As pulse is highly compressed, a shock front can be observed for the case of decreasing pressure and constant pressure under the effect of self-steepening, which also contributes to enhancement on the blue edge of the spectrum. Pulse with increasing pressure is perturbed by higher-order dispersion, as reflected by the jagged line on the yellow curve. As pulse experiences soliton-effect compression in anomalous dispersion regime, the pulse spectrum broadens and duration reduces. Increased spectral bandwidth strengthen the dispersion while reduced duration increases the pulse intensity and thus the nonlinearity. This in turn accelerates the spectral broadening. This process keeps going on until the two effects balance with each other. If the pulse is propagating in a constant pressure gas medium. the spectrum experiences the strongest perturbation by higher-order dispersion at the maximum compression point. Subsequently, the higher-order soliton breaks up into multiple constituents followed by significant amount of spectral broadening with emission of dispersive wave [109]. It is the key mechanism for supercontinuum generation in the anomalous dispersion regime.

Figure 4. 5 Output temporal profiles (converted to peak power) with decreasing pressure (blue curve), constant pressure (red curve) and increasing pressure (yellow curve). Grey dotted line shows the input pulse.

Applying decreasing (or increasing) pressure, however, changes the dispersion and nonlinearity and thus, the soliton dynamics. Figure 4.6 shows the evolution of GVD with propagation distance for different types of pressure distribution.

Figure 4. 6 Group-velocity dispersion at different positions for decreasing pressure (blue curve), constant pressure (red curve) and increasing pressure (yellow curve).

For the case of decreasing pressure, GVD increases in magnitude and further away from the

zero-dispersion as pulse propagates, and it is the vice versa for increasing pressure. At the end of fiber, GVD is the largest for the case of decreasing pressure, indicating that the impact of higher-order dispersion on the pump is weakest. As a result, the soliton propagates with decreasing pressure can achieve the highest peak power, even though it has a slightly narrower spectral bandwidth as compared to the case of constant pressure. For increasing pressure case, the strength of anomalous dispersion reduces as distance increases and experiences increasing amount of perturbation by higher-order dispersion, this together with the slow compression rate due to initially low nonlinearity, lead to worst compression for increasing pressure as compared to the rest.

We look into the spectrogram for more detail. It is evident that there is emission of multiple dispersive waves for the case of decreasing pressure, as shown in Fig. 4.7 (a). The high frequency part is significantly enhanced as compared to the case of constant pressure (Fig. 4.7 (b)) and increasing pressure (Fig. 4.7 (c)). This is due to blueshift of phase-matched wavelength caused by rapidly reducing pressure in longitudinal direction near the output of fiber. The two distinguishable dispersive waves are also observed in the spectral profile in Fig. 4.3 (a). At $L = 11$ cm, where the pressure is 16.1 bar, a strong dispersive wave emits at around 830 nm. At $L = 12$ cm, the pressure drops to 11.3 bar with emission of dispersive wave at 664 nm. As pulse propagates further towards the end of the fiber, the dispersive wave forms at shorter wavelength with weaker intensity, since the spectral overlap between the soliton and dispersive wave become weaker.

Figure 4. 7 Output Spectrogram for (a) decreasing pressure (b) constant pressure and (c) increasing pressure at $z=13$ cm

We also repeat the simulation for the case of decreasing pressure but exclude the ionization terms and compare it with the full model obtained previously, as shown in Fig. 4.8. The indistinguishable temporal and spectral profiles for both elucidates that photoionization effect is negligible and therefore does not contribute to the enhancement at blue edge of the spectrum [110]. This result also shows that such system is highly resistant to ionization.

Figure 4. 8 Comparison of (a) output temporal profiles and (b) output spectral profiles for the case of decreasing pressure, obtained with full model (blue curve) and without ionization (red curve)

4.7 Conclusion

We numerically investigate the impact of different pressurization configurations on soliton-effect compression of supercontinuum in MIR region. We find decreasing pressure can increase the output peak power of MIR supercontinuum induced by soliton-effect compression by 1.63 times and 3 times as compared to the equivalent constant pressure and increasing pressure,

respectively, for the set of parameters given in our simulation. Decreasing pressure introduces longitudinally increasing group-velocity dispersion, which is accompanied by suppression of higher-order dispersion. As a result, the soliton suffers from less perturbation from higher-order dispersion and can compress to higher peak power. It is worth noting that enhancement of pulse compression in the MIR region with decreasing pressure is not observed before. Besides, the blue edge of the spectrum is significant enhanced with decreasing pressure and multiple dispersive waves are generated.

In fact, applying decreasing pressure is more desired in practical situation for dispersion-free delivery of pulse [45]. Just that the pulse and fiber parameters should be properly selected to ensure the maximum compression point is near the output of the fiber. Alternatively, the compression length can be flexibly adjusted by linearly stretching the pulse [111]. The results presented in this chapter is not optimized yet. For better compression performance, further investigation on the scaling rules for high quality soliton-effect compression in gas-filled hollow waveguides with decreasing pressure is required.

Chapter 5: Effect of decreasing pressure on soliton self-compression in higher-order modes of a gas-filled capillary

In Chapter 4, we demonstrate enhancement of soliton-effect compression with decreasing pressure in the fundamental core mode of an AR-HCF. In this chapter, we further extend our study to the higher-order modes of a gas-filled capillary.

5.1 Introduction

With the development of gas-based nonlinear optics, there is a growing interest in generating high-energy ultrashort pulses in gas-filled capillaries using the soliton self-compression [15, 16]. Gas-filled capillaries have many advantages, such as high power-damage threshold, adjustable dispersion and nonlinearity, as well as widely accessible transmission window from vacuum UV to infrared. As an inherently multimode waveguide, capillary supports various linearly polarized leaky modes denoted as HE_{1p} modes [68]. One interesting direction that calls for further investigations is the soliton self-compression in the higher-order modes of gas-filled capillaries. There are several attractive aspects about staging the soliton self-compression in the higher-order modes of gas-filled capillaries. Firstly, in a capillary, HE_{1p} mode (with $p \geq 2$) has a smaller spatial dimension than the fundamental mode, leading to stronger nonlinearity. Secondly, the contribution of negative waveguide dispersion increases with the mode order. This provides wider selection of gas species and pressure range that facilitate the anomalous dispersion regime for the soliton self-compression. Thirdly, the larger anomalous dispersion also means the self-compression can be done within a shorter length [15], which is critical for large-core capillary compressors, since their bending-induced loss is one of the major limiting factors.

The soliton self-compression in higher-order modes of gas-filled capillaries has emerged as a

topic of interest in the last couple of years. Numerical investigations showed that compressed pulses approaching the single-cycle regime can be obtained in the higher-order modes [66, 67]. These studies assumed constant gas pressure along the length of the capillary. On the other hand, there are some major benefits of keeping the exiting end evacuated in the gas-filled hollow waveguide-based compressors [45]. Doing so can avoid the dispersion and nonlinearity induced distortion of the compressed pulse when it passes through the gas chamber attached at the output end. Furthermore, by applying a negative pressure gradient, we can minimize the nonlinear mixing between the modes and improve the mode purity of the output beam.

In this work, we apply negative pressure gradient along the longitudinal direction in the argon-filled capillary pulse compressor by evacuating its output end. We numerically investigate the soliton self-compression by launching the pump in different higher-order modes. We show that, in addition to the distortion-free pulse delivery at the output, applying the decreasing pressure enhances the compression in HE_{12} mode by a factor of four compared to the case where an equivalent constant pressure is used. The outcome is a highly compressed pulse having a sub-cycle duration of 1.85 fs and the peak power of 7.96 GW achieved from a single-stage compressor. Additionally, high mode purities can be realized by suppressing the intermodal coupling through the decreasing pressure.

5.2 Modeling of soliton-effect compression in the higher-order modes of gas-filled capillary

The model is based on the multimode generalized nonlinear Schrödinger equation (MM-GNLSE) [60]. Starting from Eq. (2.33), after adopting the full wavelength-dependent dispersion, dropping the Raman term and taking the Fourier transformation, we get the mode-resolved equation in the frequency domain, given as:

$$\begin{aligned} \partial_z A_p(z, \omega) = & i \left(\beta^{(p)} - \beta_0^{(1)} - \beta_1^{(1)}(\omega - \omega_0) + i \frac{1}{2} \alpha^{(p)} \right) A_p(z, \omega) \\ & + i \frac{\omega}{c} \mathcal{F} \left\{ n_2 \sum_{l,m,n} s_{plmn} A_l(z, t) A_m(z, t) A_n^*(z, t) \right\}, \end{aligned}$$

(5.1)

where $A_p(z, \omega)$ is the complex envelope of the pulse in HE_{1p} mode in the frequency domain at position z along the length of the fibre and ω is the angular frequency. t is the time frame moving at the group-velocity of the pump pulse in HE_{11} mode, $1/\beta_1^{(1)}$.

The first term on the right-hand side of Eq. (5.1) describes the linear effects in HE_{1p} mode. From the analytical model derived by Marcatili and Schmeltzer for a dielectric capillary, the wave vector, $\beta^{(p)}$, for HE_{1p} mode is given by [68]:

$$\beta^{(p)} = \frac{\omega}{c} \left(n_g - \frac{1}{2} \frac{u_p^2 c^2}{\omega^2 a^2} \right). \quad (5.2)$$

Here, n_g is the pressure dependent refractive index of the gas obtained by taking the Sellmeier coefficients reported in [112] and rewriting Equation (14) in Ref [112] as an function of angular frequency ω :

$$n_g = \sqrt{1 + \frac{P}{P_{ref}} \cdot \frac{T_{ref}}{T} \cdot \left[\frac{A_1^2 B_1}{A_1^2 - \omega^2} + \frac{A_2^2 B_2}{A_2^2 - \omega^2} \right]}, \quad (5.3)$$

where $P_{ref} = 1 \text{ bar}$ and $T_{ref} = 273 \text{ K}$ is the standard pressure and temperature, respectively.

$$B_1 = 20332.29 \times 10^{-8}, B_2 = 304458.31 \times 10^{-8}, A_1 = \frac{2\pi c \times 10^6}{\sqrt{206.12 \times 10^{-6}}}, A_2 = \frac{2\pi c \times 10^6}{\sqrt{8.066 \times 10^{-3}}}.$$

u_p is the p -th zero of the first-order Bessel function, c is the speed of light in vacuum, and a is the bore radius of the dielectric capillary. $\beta_0^{(1)}$ and $\beta_1^{(1)}$ are the first two Taylor series expansion coefficients of $\beta^{(1)}$ at the pump frequency, ω_0 , and are related to the phase and group velocities of the pump in HE_{11} mode. $\alpha^{(p)}$ is responsible for the power loss in HE_{1p} mode, and is given by [68]:

$$\alpha^{(p)} = \frac{u_p^2 c^2}{\omega^2 a^3} \frac{n_d^2 + 1}{\sqrt{n_d^2 - 1}}, \quad (5.4)$$

where n_d is the refractive index of the dielectric material comprising the hollow waveguide, i.e. silica. The gas contribution to the loss can be neglected at wavelength far from its atomic

resonances.

The second term on the right-hand side of Eq. (5.1) is the nonlinear term. n_2 is the pressure dependent nonlinear index of the gas which we obtain from [113]. The summation denotes the intra- and inter-modal optical Kerr effects, where $A_l(z, t)$, $A_m(z, t)$ and $A_n(z, t)$ refer to the complex envelopes in the interacting modes l , m and n . Namely, $l = m = n = p$ represents the self-phase modulation, while any other combinations of l , m and n imply intermodal cross-phase modulations and four-wave mixing processes [114]. s_{plmn}^K is the Kerr induced overlap integral between HE_{1p} and the other interacting modes, as provided in Eq. (2.37). It is given as the inverse of the effective mode area for the nonlinear coupling. $\mathbf{F}_p$ represents transverse field profile of HE_{1p} mode and its effective mode area, $A_{\text{eff}}^{(p)}$, is given by $1/s_{pppp}^K$. Additional nonlinear contributions such as the Raman effect and photoionization may need to be included when using molecular gases or involving ionizing intensities. Here, we neglect the contribution from the former with the use of only the atomic gases in the study. Furthermore, we confine our study only to non-ionizing regime. We have numerically confirmed the accuracy of our model for the set of parameters considered in the current study by comparing some of the results against a full field-based model that includes the effect of ionization. The details shall be presented later in Section 4.

The filling gas pressure influences two parameters in Eq. (5.1), i.e., $\beta^{(p)}$ and n_2 , through the change of atomic density. We create a pressure gradient along the waveguide of length L by setting different pressures at the input and output ends of the capillary, P_0 and P_L , respectively. The pressure at position z is then given by [21]:

$$P(z) = \sqrt{P_0^2 + \frac{z}{L}(P_L^2 - P_0^2)}. \quad (5.5)$$

In such a case, the pulse propagation dynamics is subject to the fiber length, L , as it determines the pressure change, and therefore the changes in the system's dispersion and nonlinearity

along the propagation.

In our simulations, we include the first ten HE_{1p} modes that are coupled through the nonlinear term in Eq. (5.1). We only consider the HE_{1p} modes in the calculations as a particular input mode results only in output modes of the same azimuthal symmetry [115]. In each numerical simulation, we excite and launch into only a single higher-order mode, i.e., HE_{12} , HE_{13} , or HE_{14} . The predominant excitation of a selected higher-order mode can be realized experimentally by, for example, using a prism-coupling method [116]. As compared to the fundamental HE_{11} mode, pulse propagation in higher-order modes of gas-filled capillary can easily couple with other spatial modes through nonlinear interaction, shedding some of its energy into these modes. We have verified that spatial modes with order higher than ten have negligible impact on our results. This choice provides a good compromise between the computational accuracy and resource requirement. Table 5.1 below lists the intensity profiles of the first ten spatial modes involved in the calculation.

Table 5. 1 Intensity profiles for the first ten HE_{1p} modes

HE_{11}	HE_{12}	HE_{13}	HE_{14}	HE_{15}

HE_{16}	HE_{17}	HE_{18}	HE_{19}	HE_{110}

5.3 Numerical implementation

Eq. (5.1) can be numerically implemented using the conventional Split-Step Fourier Method.

For an optical pulse propagating across p different modes, the iteration required using split step

Fourier method will be $n_{total_ss} = L_{fibre}/\Delta z$. Each iteration involves calculation of p^4 inter-modal nonlinear coupling coefficients, which requires extensive computational resources especially for large mode numbers. To reduce the computation time and improve the calculation efficiency, we implement a Massively Parallel Algorithm (MPA) for simulating pulse propagation in multimode fibers [117-119].

With MPA method, a large step size $\Delta L = M\Delta z$ is used. There are $M + 1$ nodes with equal separation distance of Δz in each ΔL and these nodes are denoted as z_q where $q = 0, 1, 2, \dots, M$. Δz , ΔL should be properly selected such that it is much shorter than the nonlinear length $L_{NL} = \frac{1}{P_0 \gamma}$. This is to ensure the envelope is slowly varying along fiber length L [120, 121].

Like the Split-Step Fourier Method, we decouple the effect of dispersion and nonlinearity, calculate the linear term in frequency domain and nonlinear terms in time domain. Assuming the pulse envelope of mode p at position z in frequency domain is denoted as $\varphi_p(z, \omega)$, after considering effect of linear dispersion, it will become:

$$\hat{A}_p(z, \omega) = \hat{\varphi}_p(z, \omega) e^{D_\omega^{(p)}(z)}, \quad (5.6)$$

where

$$D_\omega^{(p)} = i[\beta^{(p)}(\omega) - \beta_0^{(1)} - \beta_1^{(1)}(\omega - \omega_0)], \quad (5.7)$$

$$\tilde{D}_\omega^{(p)}(z) = \int_{z_0}^z D_\omega^{(p)}(z') dz', \quad (5.8)$$

We refer z to any position satisfying $z_0 \leq z \leq z_M$. The purpose of Eq. (5.6) is to perform a change of variable which facilitates the integration process later. We use the initial condition $\hat{\varphi}_p^{(0)}(\omega, z) = \hat{\varphi}_p(\omega, z_0) = A_p(\omega, z_0)$ at every z_q along z . By substituting Eq. (5.6) into equation Eq. (5.1), applying the Fourier Transform and taking the integration on both side of Eq. (5.1), we get [122]:

$$\begin{aligned}\hat{\phi}_p(z, \omega) = & \hat{\phi}_p(z_0, \omega) + i \frac{n_2 \omega_0}{c} (1 \\ & + \frac{\omega}{\omega_0}) \times \sum_{l,m,n} \int_{z_0}^z \mathcal{F}\{s_{plmn}^K A_l(z', t) A_m(z', t) A_n^*(z', t)\} e^{-\tilde{D}_\omega^{(p)}(z)} dz'.\end{aligned}\quad (5.9)$$

Eq. (5.9) can be numerically implemented as following steps:

1. Assign the same initial envelope of $\hat{\phi}_p(z_0, \omega)$ to $z_0, z_1 \dots z_M$.
2. Calculate $\hat{A}_p(z, \omega) = \hat{\phi}_p(z, \omega) e^{\tilde{D}_\omega^{(p)}(z)}$ at each position of $z_0, z_1 \dots z_M$.
3. Take the Fourier Transform of $\hat{A}_p(z, \omega)$ at each position of $z_0, z_1 \dots z_M$ to get $A(z, t)$.
4. Calculate $U_p(z, t) = \sum_{l,m,n} s_{plmn}^K A_l(z, t) A_m(z, t) A_n^*(z, t)$.
5. Take Fourier Transform of $U_p(z, t)$ to get $\hat{U}_p(z, \omega)$ at each position of $z_0, z_1 \dots z_M$ for all the p modes.
6. Integrate $\hat{R}_j = i \frac{n_2 \omega_0}{c} (1 + \frac{\omega}{\omega_0}) \hat{U}_p(z, \omega) e^{-\tilde{D}_\omega^{(p)}(z)}$ along z and keep the intermediate results at each position of $z_0, z_1 \dots z_M$. ($j = 0, 1, \dots M$).
7. Repeat step 2 to 6 for the next iteration until meet the required accuracy.
8. Obtain $\hat{A}_p(z_M, \omega)$ at the end of large step ΔL .

In this way we can simultaneously process M calculations from step 2 to step 5 and then integrate $\hat{R}_j$ over z at step 6. All the intermediate results at each node from $z_1 \dots z_M$ are calculated using trapezoidal rule and stored for use in the next iteration:

$$\hat{\phi}_{p,q}^{(n+1)}(\omega) = \hat{\phi}_{p,0}^{(n)}(\omega) + \frac{\hat{R}_0^{(n)}}{2} + \sum_{j=1}^{M-1} \hat{R}_j^{(n)} + \frac{\hat{R}_M^{(n)}}{2}. \quad (5.10)$$

(n is the iteration number and $q = 1, 2, \dots M$)

After each iteration, the $\hat{\phi}_{p,q}$ at each node q will be updated and approach to the exact solution at that particular node z_q . The iteration stops when the estimated error falls below the tolerance.

The value at last iteration, $\hat{\phi}_{pM}^{(n)}$ for the node z_M at the end of ΔL will be used as the starting

point for the next ΔL , where the above procedures repeat again until the pulse reaches the end of fibre. If we assume the iteration number for every ΔL are the same, then the total number of loops requires to complete the propagation in fibre with length L_{fibre} will be:

$$n_{total_mpa} = \frac{nL_{fibre}}{\Delta L} = \frac{n}{M} \cdot \frac{L_{fibre}}{\Delta z} = \frac{n}{M} \cdot n_{total_ss}. \quad (5.11)$$

When the iteration number n for each ΔL is much smaller than the number of nodes M , implementing MPA method can substantially reduce the computation time as compared to the Split-Step Fourier Method. The schematic diagram that describes all the above procedures is shown in Fig. 5.1.

Figure 5. 1 Schematic diagram of MPA

5.4 Loss, effective mode area, group-velocity dispersion and nonlinear coefficient

In our numerical study, we use an 800nm, 100 μ J transform-limited Gaussian pulse with 35 fs full-width at the half-maximum (FWHM) duration. Such pulses can be obtained from a commercial mode-locked laser system such as Ti:Sapphire lasers. Figure. 5.2 (a) shows the loss and effective area versus the core diameter $2a$, evaluated at 800 nm for HE_{11} , HE_{12} , HE_{13} , and HE_{14} modes in a silica capillary. Left-hand side panels show the intensity profiles of the

four modes with the color of each surrounding box indicating the line color for the corresponding mode in 5.2 (a) and 5.2 (b). According to Fig. 5.2 (a), the loss depends strongly on the core diameter in all modes. A large core gives much lower loss but at the expense of increased effective mode area that leads to reduced nonlinearity. We choose the core diameter of 300 μm , which strike a good balance between the loss and mode size. At 300 μm core diameter, the losses at 800 nm are 0.36 $\text{dB}\cdot\text{m}^{-1}$, 1.88 $\text{dB}\cdot\text{m}^{-1}$, 4.61 $\text{dB}\cdot\text{m}^{-1}$, and 8.56 $\text{dB}\cdot\text{m}^{-1}$ in HE_{11} , HE_{12} , HE_{13} , and HE_{14} modes, respectively, as shown in Fig. 5.2 (a). These values are tolerable for experiments requiring a relatively short fiber, e.g., <2 m. Likewise, for the pump pulse considered in our study, the effective areas of these modes are small enough to induce sufficient nonlinear phase shifts within the short length at moderate argon pressure.

Figure 5. 2 (a) Loss (solid lines) and effective mode area ($A_{\text{eff}}^{(p)}$, dashed lines) versus bore diameter evaluated at 800 nm for HE_{11} , HE_{12} , HE_{13} , and HE_{14} modes. (b) GVD parameter ($\beta_2^{(p)}$, solid lines) and nonlinear coefficient ($\gamma^{(p)}$, dashed lines) at 800 nm versus argon pressure for HE_{11} , HE_{12} , HE_{13} , and HE_{14} modes.

Figure 5.2 (b) shows the group-velocity dispersion (GVD) parameter, $\beta_2^{(p)}$, and nonlinear coefficient, $\gamma^{(p)}$, defined as $n_2\omega_0/cA_{\text{eff}}^{(p)}$, at 800 nm versus argon pressure for HE_{11} , HE_{12} , HE_{13} , and HE_{14} modes in a capillary fiber with the core diameter of 300 μm . We note that for the given set of system parameters, the pump lies in the normal dispersion regime only in HE_{11} mode, while all the higher-order modes place it in the anomalous dispersion regime. This opens the opportunity to excite soliton-driven nonlinear effects, including the soliton self-compression, in the higher-order modes of the capillary fiber. From Fig. 5.2 (b), we note that the magnitude of the anomalous dispersion increases as the mode number is increased, while

it decreases with pressure in each higher-order mode.

5.5 Simulation results and discussion

The calculations are carried out for decreasing argon pressure along the length of the capillary fiber with 1.8 bar at the input end and evacuated at the other end. We assume the nonlinear effect initially dominates that of the dispersion, which is a valid assumption for launching a higher-order soliton. Moreover, we consider the intermodal interaction is weak such that only a small fraction, i.e., <10%, of the total energy launched end up being coupled into other modes.

The total nonlinear phase shift, $\phi_{NL}^{(p)}$ (also known as the B-integral) experienced by the pulse launched into HE_{1p} mode in decreasing gas pressure from P_0 at the input to $P_L = 0$ at the output can be approximated by [103]:

$$\phi_{NL}^{(p)} = I_0 \int_0^L \gamma^{(p)}(z) dz = I_0 \gamma_0^{(p)} \int_0^L \sqrt{1 - \frac{z}{L}} dz = \frac{2}{3} I_0 L \gamma_0^{(p)}, \quad (5.12)$$

where I_0 is the peak power of the pump, and $\gamma_0^{(p)}$ is the nonlinear parameter at the fiber input which is directly proportional to the input pressure P_0 for an ideal gas. Eq. (5.12) implies that an equivalent amount of total nonlinear phase shift from a system of the same length can also be achieved with a constant pressure $P_{const} = 2/3 P_0$.

In our simulation, different fiber length L is used for each higher order modes. It is chosen to be the pulse compression length at the corresponding mode with constant gas pressure of $2/3 P_0 = 1.2$ bar. This choice allows us to highlight the effect of the decreasing pressure on the soliton self-compression in each higher-order modes against the case involving the constant pressure. For the set of fiber and input pulse parameters that are considered in our numerical study, this amounts to $L = 1.96$ m, 1.03 m, and 0.69 m for HE_{12} , HE_{13} , and HE_{14} modes, respectively. The higher the mode order, the shorter the length becomes, since $|\beta_2^{(p)}|$ increases with p leading to a shorter compression length. In fact, this works in favor of the higher-order modes, as the shorter propagation length can compensate for larger attenuation experienced in

the higher-order modes. We stress that the parameters used in the numerical study are well within what is realizable in the laboratory.

Figures 5.3 (a), 5.3 (b), and 5.3 (c) show the evolutions of the FWHM durations when the pump is launched into HE_{12} , HE_{13} , and HE_{14} modes, respectively, for the decreasing pressure (solid lines) and constant pressure (dotted lines). The black dash-dotted lines in Fig. 5.3 are evolutions of the compression rate figure-of-merit, FOM_{comp} , values which we introduce to estimate the compression dynamics as we shall discuss later. The pulse, in each case shown, undergoes soliton self-compression due to the interplay between anomalous dispersion and nonlinear effect. In HE_{11} mode, which is not presented here, is in the normal dispersion regime and therefore no soliton self-compression takes place.

Figure 5. 3 Evolutions of the FWHM durations in (a) HE_{12} , (b) HE_{13} , and (c) HE_{14} modes in the argon-filled capillary with the decreasing pressure (solid lines). The evolutions under the equivalent constant pressure system (dotted lines) are plotted together for reference. The changes of the FOM_{comp} values along the propagation are shown in black dash-dotted lines.

We notice substantial enhancement for pulse compression in HE_{12} mode with decreasing pressure. Namely, the compressed pulse at the capillary exit has a FWHM duration of 1.85 fs, reaching the sub-cycle regime under the decreasing pressure. This corresponds to the compression ratio, $F_c = 19$, which is four times that achievable in the constant pressure system where the output duration is 7.48 fs amounting to $F_c = 4.68$. On the contrary, the decreasing pressure deteriorates the compression in HE_{13} and HE_{14} modes, resulting in the output pulse duration of 14.85 fs and 5.7 fs, respectively, as opposed to 2 fs and 4.3 fs when the equivalent

constant pressure is applied.

In the initial stage of soliton self-compression, the effect of nonlinearity dominates that of the anomalous dispersion. The relative strength between nonlinearity and dispersion is governed by the soliton order. The self-phase modulation broadens the spectrum and induces positive chirp, which gets compensated by the negative chirp from the anomalous dispersion. This leads to pulse compression that in turn reinforces the nonlinearity. The process repeats until the pulse spectrum broadens to the extent that the effects of the dispersion and nonlinearity are at a balance.

The wide spectral bandwidth increases the importance of the higher-order dispersion, which is particularly strong when the pulse is in the vicinity of zero dispersion [123]. The strong perturbation by the higher-order dispersion causes pulse to break, limiting the compression performance. This is what happens in HE_{12} mode when the constant pressure is applied, because the anomalous dispersion in this case is very weak at 1.2 bar as shown in Fig. 5.2 (b). However, when the decreasing pressure is used, the anomalous dispersion that starts near zero increases in its magnitude along the length, and surpasses that of the constant pressure at $z = 0.65$ m. The increased anomalous dispersion effectively mitigates the perturbation caused by the higher-order dispersion in the later stage of the compression. Here, the spectral bandwidth increases as the pulse compresses down the capillary. Considering that the higher-order dispersion has more severe impact when the spectrum is broader, having larger anomalous dispersion towards the output end where the spectrum is broader works in favor of mitigating the effect. This substantially improves the compression, reducing the output pulse duration down to a quarter of that achievable under the constant pressure. As we shall see in Fig. 5.4(a), this improvement is achieved even though there is similar amount of spectral broadening for the case of decreasing and constant pressure. In HE_{13} and HE_{14} modes, the waveguide dispersion becomes stronger, shifting the overall dispersion further away from the zero

dispersion. This effectively weakens the influence of the higher-order dispersion. Hence, the suppression of the higher-order dispersion effect introduced by the decreasing pressure does not play a prominent role, and no enhancement is seen in HE_{13} and HE_{14} modes.

The second important factor that determines the output pulse duration in a decreasing pressure is the mismatch between the maximum compression point and the fiber end. The compression length in the soliton self-compression, L_{comp} , is related approximately to γ and $|\beta_2|$ as below [15]:

$$L_{comp} \propto \sqrt{\frac{1}{\gamma|\beta_2|}}. \quad (5.13)$$

A higher $\gamma|\beta_2|$ value leads to a shorter compression length and a faster compression process. In decreasing pressure, γ is continuously decreasing while $|\beta_2|$ is increasing. To qualitatively describe the impact of these two parameters on the self-compression dynamics, we use the compression rate figure-of-merit, FOM_{comp} , denoted as $\sqrt{(\gamma(z)|\beta_2(z)|)/(\gamma'|\beta_2'|)}$, where $\gamma(z)$ and $|\beta_2(z)|$ are the z dependent nonlinear and GVD parameters for the decreasing pressure at the given mode, while γ' and $|\beta_2'|$ are those at the constant pressure of 1.2 bar. We note that in the regions where FOM_{comp} is greater than 1 (as indicated by the shaded area with gray color in Fig. 5.3), the pulse duration reduces more rapidly when the decreasing pressure is applied. This agrees well with Eq. (5.13) where a larger value of $\gamma|\beta_2|$ shortens the compression length and accelerates the compression rate. It shows that the combined effect of γ and $|\beta_2|$ has significant impact on the self-compression dynamics. FOM_{comp} can be used to estimate how the compression rate in the decreasing pressure changes with respect to the case involving the equivalent constant pressure.

As the compression rate changes accordingly with FOM_{comp} in the decreasing pressure, the compression length is continuously adjusted. For HE_{12} mode, the compression length in the decreasing pressure happens to roughly coincide with that of the constant pressure in our

numerical example, yielding the maximally compressed pulse at the evacuated fiber end and significant gain in the compression factor. In HE_{13} and HE_{14} modes, on the other hand, the compression points under the decreasing pressure are brought forward. In HE_{13} mode, the pulse reaches the maximum compression at around $z = 0.9$ m. This is followed by a soliton fission as reflected in the red-solid line in Fig. 5.3 (b) with a dramatic increase in the pulse duration. In HE_{14} mode, the pulse compresses to the shortest duration at $z = 0.59$ m, after which the pulse broadens slowly as indicated by the orange-solid line in Fig. 5.3 (c). This is because further compression is prohibited due to relatively high loss, as well as the diminishing nonlinearity. In both cases, the pulses temporally broaden and peak powers decay thereafter before reaching the fiber end, and results in weaker compressions.

Figure 5.4 shows the temporal and spectral profiles of the pulses at the output end of the fibers for the three higher-order mode launches, i.e., (a) HE_{12} , (b) HE_{13} , and (c) HE_{14} modes. We observe that by introducing the decreasing pressure, peak power of the compressed pulse in HE_{12} mode is enhanced by 2.7 times, from 2.9 GW to 7.96 GW. On the other hand, peak powers in the case of the decreasing pressure are reduced for HE_{13} and HE_{14} modes. It is evident in Fig. 5.4 that the change in the pressure profile does not influence much the bandwidth of the output spectrum. This is expected as the equivalent constant pressure is determined based on the assumption that the two systems accumulate approximately the same amount of total nonlinear phase shift by matching the B-integrals. The oscillations seen in the trailing edge of the compressed pulses in Fig. 5.4 (a) is due to the higher-order dispersion. In HE_{12} mode, GVD parameter is closer to zero, and hence the effect of the higher-order dispersion is more prominent and the oscillation is relatively larger than the rest of higher-order modes. The oscillation in HE_{12} mode under the constant pressure is even stronger than that of the decreasing pressure and the pulse profile is severely perturbed, resulting in poor compression performance.

Figure 5. 4 Temporal (left) and spectral (right) profiles of the compressed pulses in (a) HE_{12} , (b) HE_{13} , and (c) HE_{14} modes. Red-solid lines are for the decreasing pressure systems while blue-solid lines are for the equivalent constant pressure cases. The initial profiles are plotted in gray-dashed lines for reference.

Another notable feature in Fig. 5.4 (a) is the delay of the pulse peak with respect to the pedestal in the leading edge. This is caused by the extreme self-steepening which occurs when the pulse self-compresses to sub-cycle duration [16]. In the self-compression process, the pulse peak experiences the strongest compression while the tails stay relatively unchanged. As compression continues, the pulse peak lags due to the intensity dependence of the group velocity, resulting in the high-intensity peak to trail behind the low-intensity pedestal. In Fig. 5.4 (b), we can see the solitonic structure in HE_{13} mode is distorted at the output end due to

soliton fission. In HE_{14} mode, the self-compression process ceases too early due to the relatively high loss, as well as the reducing nonlinearity. Although the solitonic structure is still maintained, the pulse is temporally dispersed and the peak power decays by 17% as it reaches the output end, as shown in Fig. 5.4 (c).

An additional feature to note in the spectra presented in Fig. 5.4 is the dispersive wave generation [124]. The DW is emitted as the higher-order soliton undergoes fission after reaching the maximum compression point. The location of the dispersive wave can be estimated using the phase-matching condition below [50, 55]:

$$\Delta\beta^{(p)} = \beta^{(p)} - \beta_0^{(p)} - \beta_1^{(p)}(\omega - \omega_0) - \frac{I_0\gamma^{(p)}}{2}. \quad (5.14)$$

The dispersive wave starts to appear close to the output end where the spectrum is broad enough to seed the energy conversion to the phase-matching point. This means Eq. (5.14) needs to be solved for a lower pressure value in the decreasing pressure case which can have markedly different phase-matching point from that of the equivalent constant pressure system. To illustrate this, we show the dephasing diagram in Fig. 5.5 for HE_{12} , HE_{13} , and HE_{14} modes at two different pressures, i.e. 1.2 bar for the equivalent constant pressure case and 0.6 bar to exemplify a lower pressure value near the output end in the decreasing pressure capillary. At 1.2 bar, the phase-matching occurs at around 460, 320, and 240 nm in HE_{12} , HE_{13} , and HE_{14} modes, respectively, with the phase-matching wavelength decreases with increasing mode order. This is consistent with the results shown in blue lines in Figs. 5.4 (a) and 5.4 (b), where spectral features are prominent at these wavelengths in the corresponding constant pressure cases. Due to high loss in HE_{14} mode, spectral broadening is not sufficient to warrant dispersive wave radiation. At 0.6 bar, the phase-matching points blue-shift, which is also the case as shown with red lines in Figs. 5.4 (a) and 5.4 (b) where the short-wavelength bands are shifted towards shorter wavelength.

Figure 5.5 Dispersive wave dephasing diagram for HE₁₂ (blue), HE₁₃ (red), and HE₁₄ (orange) modes. Solid lines are for 1.2 bar and dashed lines are for 0.6 bar to illustrate the equivalent constant pressure and decreasing pressure cases, respectively. Vertical-dotted lines mark the phase-matching wavelengths.

At this point, we check the accuracy of our results and corroborate MM-GNLSE for the HE₁₂ mode launch cases considered above. For the validation, we solve the uni-directional pulse propagation equation (UPPE) for the same set of parameters with and without ionization term [125]. Note that this field-based model considers only a single mode propagation in the launch mode, and therefore does not include intermodal effects. Figure 5.6 (a) shows evolutions of the FWHM durations obtained from three different models, i.e. MM-GNLSE, UPPE with ionization term, and UPPE without ionization term, for the identical cases studied in Fig. 5.3(a). Qualitatively the same evolutions are observed in all three models, except for intermodal coupling-mediated small oscillations that appear in MM-GNLSE but not in the single-mode models. In particular, the significant enhancement of compression in HE₁₂ mode for the case of decreasing pressure is reproduced with UPPE, which indicates that this improvement is not contributed by intermodal effects and the single-mode configuration is sufficient to describe it. We also show that results from the models with and without photoionization effect are practically indistinguishable. This is despite the appearance of higher peak powers in the UPPE model. Therefore, we can safely omit ionization in our study. In fact, when decreasing pressure is used, the gas density diminishes as the pulse compresses down the capillary, effectively

eliminating the impact of ionization. Figure 5.6 (b) is the comparison of the output pulse profiles for the same decreasing pressure case computed using the three models. Likewise, no qualitatively significant differences are observed for the UPPE models with and without ionization terms.

Figure 5. 6 (a) Evolutions of the FWHM durations for the decreasing pressure (solid lines) and equivalent constant pressure (dashed lines) cases obtained using MM-GNLSE (blue), UPPE with ionization term (orange), and UPPE without ionization term (green). Note the orange and green lines overlap indicating that the effect of ionization is negligible. (b) Output profiles for the decreasing pressure case calculated using the three models.

When the pulse is highly compressed, it can induce strong intermodal coupling and significantly degrade the spatial beam quality, if sufficient nonlinearity is present in the system. One additional benefit of using the decreasing pressure is that: it ensures small nonlinear parameter near the output end of the capillary where the pulse has high peak power, minimizing the nonlinear interaction between modes. Hence high degree of mode purity and excellent beam quality can be achieved. Figure 5.7 (a), (b) and (c) shows the evolutions of the percentage energy weightages in the first ten HE_{1p} modes when the pump is launched into HE_{12} , HE_{13} , and HE_{14} mode, respectively. The energy weightage defines the ratio of energy contains in the predominant mode to the total energy. When there is no intermodal coupling, 100% of the energy should remain in the launched mode. At capillary output, the energy weightage in HE_{12} , HE_{13} , and HE_{14} mode is 94.9%, 97.8%, and 98.8% respectively, indicating high mode purity.

We attribute the oscillations in the energy weightages observed at the beginning of each propagation due to nonlinear coupling between different spatial modes. Similar mode coupling-induced oscillations are reflected in the FWHM durations evolution in Fig. 5.3. One of the reasons that oscillations appear only at the beginning is because, in the case of decreasing pressure, the nonlinearity and therefore the intermodal interaction is strong near the input of the capillary. It also explains why the oscillations are weaker for the equivalent constant pressure systems as seen in Fig. 5.3, where the nonlinearity remains below that required to stage noticeable intermodal interaction. There are other factors that govern the strength and extent of the mode coupling which we shall discuss in detail in the later part of this section. We notice in Fig. 5.7 (a), the energy weightage of HE_{12} mode decreases slowly towards the end of the fiber while that in HE_{11} mode gradually rises. This is mainly due to the loss in HE_{12} mode being higher than that in HE_{11} mode, and hence the energy in HE_{12} mode decays faster compared to HE_{11} mode. The near-field intensity profiles of the output beams are presented in the insets in Fig. 5.7 for the three launch cases. Their modal intensity patterns are well maintained, indicating good spatial mode qualities.

Figure 5. 7 Evolutions of the energy weightages in the first ten HE_{1p} modes for the pump launched into (a) HE_{12} , (b) HE_{13} , and (c) HE_{14} modes in the system with the decreasing pressure. Insets show the near-field intensity profiles of the corresponding output beams.

Among the three main nonlinear effects that appear in the system, self-phase modulation, cross-phase modulation, and four-wave mixing, only the intermodal four-wave mixing can induce energy exchange between the modes [114]. This is apparent when expanding the summation

in Eq. (3.1). The nonlinear terms responsible for self-phase modulation, i.e. $S_{pppp}|A_p|^2 A_p$, and cross-phase modulation, i.e. $S_{pnpn}|A_n|^2 A_p$ and $S_{ppnn}|A_n|^2 A_p$, have purely real nonlinear wave vectors, which introduce modulations on the phase alone and not any gain or loss in HE_{1p} mode. Only four-wave mixing terms carry imaginary parts that give rise to energy exchange between HE_{1p} and other modes. Out of these, terms that contain $|A_p|^2$ play major role in the mode coupling where HE_{1p} is the dominant mode. The intermodal coupling is then proportional to $n_2|A_p|^2 = n_2 I$ where n_2 and I are the distance dependent nonlinear index and peak power in HE_{1p} mode. Here we adopt the four-wave mixing figure-of-merit (FOM_{fwm}) for determining the strength of the intermodal interaction, which is given by $(n_2(z)I(z))/(n_2(z=0)I(z=0))$. FOM_{fwm} of greater than 1 means increase in the intermodal coupling strength as compared to that at the input, while that less than 1 suggests otherwise. Figure 5.8 shows n_2 (black-dotted lines), I (red-dotted lines), as well as FOM_{fwm} (solid-blue lines) along the propagation when the pump is launched into HE_{12} , HE_{13} , and HE_{14} modes. The peak power always slightly lags the local maxima of FOM_{fwm} values for all three cases presented. For the case where the pump is launched into HE_{12} mode, FOM_{fwm} is kept well below 1 even though the peak power increases by almost three times through the compression. It implies that the intermodal interaction is effectively suppressed by adopting the decreasing pressure. For HE_{13} mode, FOM_{fwm} first decreases but later rises beyond 1. This directly results in the increased energy transfer from HE_{13} mode to the other modes (e.g., HE_{12} mode) at $z = 0.8$ m as shown in Fig. 5.7 (b). This clearly demonstrates that FOM_{fwm} provides an excellent indication of the intermodal coupling strength in the system.

Figure 5.8 Evolutions of n_2 (black-dotted lines), I (red-dotted lines), and FOM_{fwm} (blue-solid lines) in the case of the decreasing pressure systems when the pump is launched into (a) HE₁₂, (b) HE₁₃, and (c) HE₁₄ modes.

Another important factor that affects the strength and extent of the mode coupling is temporal overlap between fields in the interacting modes, which is governed by the pulse duration and group-velocity mismatch. Namely, a shorter pulse and larger group-velocity mismatch results in weaker and shorter-lived interaction. This is yet another reason why the mode coupling-induced oscillations are seen only in the early part in Figs. 5.3 and 5.7. The pulse continues to compress as it propagates which, together with the walk-off due to modal dispersion, reduces substantially temporal mode overlap. This effect also explains the much weaker mode coupling observed when the pump is launched into HE₁₃ or HE₁₄ mode as shown in Figs. 5.3 (b) and 5.3 (c) as well as 5.7 (b) and 5.7 (c). We observe from Fig. 5.3 that the pulse initially compresses more rapidly in HE₁₃ or HE₁₄ mode than in HE₁₂ mode, which leads to smaller temporal mode overlap in the early section where the nonlinearity is high. The group-velocity mismatch

(GVM) dictates for how long the temporal overlap is sustained, it is given as:

$$GVM = \left| \frac{1}{v_g^{(p)}} - \frac{1}{v_g^{(q)}} \right| = |\beta_1^{(p)} - \beta_1^{(q)}|. \quad (5.15)$$

Its effect on extent of the mode coupling is illustrated in Fig. 5.9 (a). It shows temporal evolution of the total field when the system is pumped in HE_{12} mode. Here, the time frame is moving at the group velocity of HE_{12} mode that is changing with the pressure along the length. At the very beginning, some of the launch energy couples into other modes, notably HE_{11} and HE_{13} modes. The interaction shows up as interference fringe on top, which lasts until HE_{11} and HE_{13} modes walk off due to group-velocity mismatch. It agrees with the positions in Figs. 5.3 (a) and 5.7 (a) where the oscillations diminish. The group velocities of the first ten HE_{1p} modes are plotted in Fig. 5.9 (b). The velocity decreases as the pressure and mode order increases with the difference becoming larger as the mode order gets higher. These characteristics are reflected in Fig. 5.9 (a). As they walk off, HE_{11} mode advances and HE_{13} mode lags with respect to HE_{12} mode. Moreover, HE_{13} mode decouples earlier than HE_{11} mode due to bigger group-velocity mismatch. This also explains why the mode interaction is shorter lived for the case where HE_{13} or HE_{14} mode is excited. The non-dominant modes eject soon after the launch because of the increased group-velocity mismatch.

Figure 5. 9 (a) Temporal evolution of the total field when the decreasing pressure system is pumped in HE_{12} . The time frame is moving at the group velocity of HE_{12} mode that is changing with pressure along the length. (b) Group velocities normalized to the speed of light versus pressure for the first ten HE_{1p} modes.

We observed similar effect in the simulations under different decreasing pressure gradients and pulse energies when the same capillary core diameter of $300\text{ }\mu\text{m}$ is used. That is, when HE_{12} mode remains close to zero dispersion at the capillary input, we yield enhanced compression in HE_{12} mode while degradations are seen in HE_{13} and HE_{14} modes with respect to the equivalent constant pressure systems. When the pressure at the input is raised to 4.2 bar such that HE_{13} mode is in the vicinity of zero dispersion at the input, the self-compression enhancement compared to its equivalent constant 2.8 bar is observed only in HE_{13} mode. The same happens upon further increase in the input pressure that places HE_{14} mode just next to zero dispersion. In other words, we can selectively improve the compression in different higher-order modes simply by tuning the pressure at the capillary input which, from an application point of view, is very useful. This suggests that our findings are not restricted only to the specifics of the parameters considered here, but they can be applied to broader systems. Moreover, the enhancement is expected to be more pronounced when the compression approaches the ionizing intensity, allowing further power scaling of the compression. The additional loss and defocusing effects caused by the photoionization [7, 126] can be largely alleviated in decreasing pressure since there is only weak presence of the gas medium where the pulse is highly compressed.

5.6 Conclusion

In this work, we numerically investigate the effect of decreasing pressure on soliton self-compression dynamics in higher-order modes of a gas-filled capillary. In HE_{12} mode, we observe four times improvement in the compression ratio under the decreasing pressure with the output pulse in the sub-cycle duration of 1.85 fs, achieving $F_c = 19$ as compared to $F_c = 4.68$ when the equivalent constant pressure is applied. The notable enhancement in the decreasing pressure is realized through the effective suppression of the higher-order dispersion in the later stage where the anomalous dispersion increases with the pressure drop. In HE_{13} and

HE_{14} modes, the waveguide contribution to the dispersion is stronger, making the higher-order dispersion less important. Hence the decreasing pressure does not make any significant impact. Moreover, the maximum compression points under the decreasing pressure do not match exactly the compression lengths in the equivalent constant pressure systems due to different compression dynamics governed by the compression rate figure-of-merits. Namely, the compression points in HE_{13} and HE_{14} modes are brought forward in the decreasing pressure, after which the pulse either undergoes a soliton fission (HE_{13} mode) or decays and disperses (HE_{14} mode) due to attenuation and diminishing nonlinearity. Both results in degraded compressions. One more advantage of the negative pressure gradient is that it can effectively mitigate the intermodal nonlinear coupling in the compression process and therefore preserve the spatial beam quality with high mode purity. Additionally, with the fiber evacuated at its output end, we can minimize the pulse distortion in the gas chamber. This allows the delivery of the compressed pulse free of dispersion and nonlinear effects. We believe these findings will benefit applications that require high-intensity ultrashort pulses in non-conventional spatial modes, such as in nonlinear frequency conversion, micromachining, microscopy as well as particle manipulation.

Chapter 6: Impact of mode-area dispersion on pulse propagation dynamics in gas-filled anti-resonant hollow-core fibers

6.1 Introduction

Light guidance in AR-HCF can be described by the ARROW model [3], where light is tightly confined within the central core area by strong reflection at the core-cladding boundary and only small portion of light will be leaked out through resonance. Due to the unique guiding mechanism, its group velocity dispersion, loss as well as effective mode area (A_{eff}) changes dramatically in the resonance regions [3]. In fact, cladding resonances in AR-HCF have been exploited in recent years for multi-octave supercontinuum generation [26, 48] and frequency conversion [27]. The rapid variation of dispersion in the resonance region provides rich phase matching conditions for soliton-DW emission and four-wave mixing (FWM) process [28]. More importantly, the position of resonance is solely determined by the cladding wall thickness, which means that the phase matching wavelengths can be flexibly tailored to the desired value by changing the thickness of cladding wall. Comparing to the dispersion profile, less attention has been paid to the mode effective area around resonance regions. The variation of effective mode area with wavelength is also known as mode-area dispersion. Theoretical investigation of mode-area dispersion is firstly reported in [127] and later it has been considered in photonic bandgap fibers [128, 129] and multimode solid-core fibers [130]. This effect has previously been neglected when modeling pulse propagation in AR-HCFs and constant effective mode area evaluated at pump wavelength is assumed. However, such assumption may be less accurate when using small-core fiber and pumping close to the resonance where there is strong mode-area dispersion. In this paper, we present our numerical

study on the effect of mode-area dispersion in a small-core anti-resonant hollow-core fiber. Our results show that: the impact of mode-area dispersion is nonnegligible when pumping the pulse near the resonance wavelengths at sufficient input energy.

6.2 Modeling of pulse propagation in AR-HCFs with mode-area dispersion

To accurately model the pulse propagation near resonance regions of AR-HCFs, we use a modified Generalized Nonlinear Schrödinger equation (GNLSE), which considers the full-wavelength dependence of the effective mode area. The modified GNLSE is derived in spectral domain by rewriting the nonlinear coefficient γ and the pulse envelope $\tilde{A}(z, \omega)$ in a form with inclusion of wavelength dependent effective mode area, given as [127, 128, 131]:

$$\frac{\partial \tilde{C}}{\partial z} - \hat{L}(\omega) \tilde{C}(z, \omega) = i\gamma(\omega) \left[1 + \frac{\omega - \omega_0}{\omega_0} \right] \mathcal{F}\{C(z, t) \int_{-\infty}^{\infty} R(T') |C(z, T - T')|^2 dT'\}, \quad (6.1)$$

where:

$$\gamma(\omega) = \frac{n_2 n_0 \omega_0}{c n_{\text{eff}}(\omega) \sqrt{A_{\text{eff}}(\omega) A_{\text{eff}}(\omega_0)}}, \quad (6.2)$$

$$\hat{L}(\omega) = i[\beta(\omega) - \beta_0 - \beta_1(\omega - \omega_0)] - \frac{\alpha(\omega)}{2}, \quad (6.3)$$

$$\mathcal{F}\{C(z, t)\} = \tilde{C}(z, \omega) = \left[\frac{A_{\text{eff}}(\omega)}{A_{\text{eff}}(\omega_0)} \right]^{-\frac{1}{4}} \tilde{A}(z, \omega). \quad (6.4)$$

ω is the angular frequency. $\gamma(\omega)$ is the wavelength dependent nonlinear coefficient that includes the dispersions of the effective index $n_{\text{eff}}(\omega)$ and the effective mode area $A_{\text{eff}}(\omega)$. n_0 and n_2 are the linear and nonlinear refractive index, respectively. $\tilde{C}(z, \omega)$ is the normalized form of the frequency domain amplitude $\tilde{A}(z, \omega)$, to ensure that the unit is consistent within the equation [131]. ω_0 is the carrier angular frequency. $T = t - \beta_1 z$ is the time moving in a reference frame travels at group velocity of the pulse. $\hat{L}(\omega)$ represents the linear operator including the full dispersion $\beta(\omega)$ and attenuation $\alpha(\omega)$. β_0 and β_1 are the zero and first order dispersion coefficient, which are related to the phase velocity and group velocity at carrier

frequency, respectively. To describe the envelope evolution moving in the reference frame, these two terms are removed from the equation. $R(T')$ represents the Raman response function, which is neglected for hollow-core waveguides filled with noble gases. For simplicity, we also neglect the photoionization effect, which is a valid assumption for the set of parameters used in our simulation.

6.3 Fiber parameters, Loss, group-velocity dispersion and nonlinear coefficient

We consider an AR-HCF with central core diameter $D = 20 \text{ } \mu\text{m}$ surrounded by 8 cladding rings, each with inner diameter $d = 7 \text{ } \mu\text{m}$ and wall thickness $t = 570 \text{ nm}$. An ideal cross-section of the structure is presented in inset of Fig 6.1(b). Optical properties such as effective refractive index, effective mode area and confinement loss for the fundamental core mode are obtained with finite element method (FEM) using COMSOL. We fill the AR-HCF with 20 bar xenon, which allows us to observe clear impact of mode area dispersion with moderate pulse energy below $1 \text{ } \mu\text{J}$. We have also carried out simulation with different gas species such as argon, however, significant effect of mode-area dispersion only appears at the filling gas pressure as high as 85 bar, due to much lower third order susceptibility of argon when compared with Xenon [104]. Such a high pressure is difficult to achieve in the laboratory and requires an elaborate pressurization system. Figure 6.1(a) blue curve shows group-velocity dispersion (GVD) of the fundamental mode when it is filled with 20 bar xenon. The first to fourth order resonance bands are located between 900 to 1400 nm, 550 to 630 nm, 390 to 420 nm as well as 300 to 320 nm respectively with decreasing width, as highlighted in yellow. The red curve represents the GVD profile for the equivalent capillary model, which fits well with the FEM result especially at short wavelengths outside resonance regions. The two curves diverge from 1650 nm onwards due to deviation from capillary model to the FEM model at long wavelength. The grey shaded area shows the fiber confinement loss in dB/m. Figure 6.1(b) shows the

effective mode area at different wavelengths. The effective mode area curve is smooth at wavelengths far away from the resonance and oscillates strongly with increment by orders-of-magnitude in the resonance regions. Abruptly enlarged mode area in the resonance bands leads to substantially reduced nonlinearity since they are inversely proportional to each other. In general, the effective mode area increases with wavelength. Interestingly, it is the opposite way within each transmission band, yield: the effective mode area decreases with wavelength, for example, between 630 to 900 *nm*. As we shall see later, by pumping close to the resonance bands as much as possible and yet avoiding the high loss resonance region, more prominent effect of the mode-area dispersion can be observed.

Figure 6. 1 (a) Fundamental mode GVD curve obtained with FEM (blue curve) and capillary model (red curve) when filled with 20 bar xenon, and fiber confinement loss (grey shaded area) obtained with FEM. (b) effective mode area obtained with FEM. Inset shows the cross-section of the 8 tubes AR-HCF with the core diameter $D = 20 \mu\text{m}$, cladding tube diameter $d = 7 \mu\text{m}$ and cladding ring thickness $t = 570 \text{ nm}$.

6.4 Simulation results and discussion

6.4.1 Pulse propagation at 1430 nm

We firstly investigate for the case with pump wavelength of 1430 nm, which is located near the first resonance band and within the first transmission window. We launch a 30 fs 0.8 μ J pulse centered at 1430 nm into a 5 cm long AR-HCF and simulate the pulse propagation using two different numerical models. One considers the wavelength-dependent area (WDA model) and the other one assumes a wavelength-independent area (WIA model). The simulated spectrums obtained with the two models at the end of fiber are shown in Fig. 6.2. Both are normalized with reference to the maximum peak power obtained by WDA model. The spectrum obtained with WDA model is flatter with a spectral peak of DW at 330 nm, which is absent with the WIA model.

Figure 6. 2 Output spectrum for wavelength dependent area (WDA) model (black curve) and wavelength independent area (WIA) model pumped at 1430 nm.

To analyze the nonlinear dynamics for the two cases, we further compare the spectral evolution for WDA and WIA model, as shown in Fig. 6.3 (a) and (d) respectively. The pump experiences weak normal dispersion at 1430 nm. Due to strong nonlinearity, both undergo rapid spectral broadening induced by self-phase modulation and majority of the spectrum tunnels into anomalous dispersion regime, which is located at wavelength longer than 1450 nm. Since the

pump is close to the first resonance band, further spectral broadening to shorter wavelength side is inhibited by the resonance band, due to high loss and low nonlinearity in this region. Similar phenomenon is also observed in [128] and it is known as frequency locking effect. In this case, the spectrum broadens asymmetrically towards longer wavelength, forming a soliton in the anomalous dispersion regime. For both cases, the maximum spectral broadening is achieved at 0.75 cm , where the soliton is compressed to shortest duration. After which, soliton fission occurs due to perturbation of higher-order dispersion [109]. This process is usually accompanied by emission of dispersive wave (DW), provided the phase-matching condition is satisfied and soliton spectrum is sufficiently overlapped with the phase-matching point [123, 132, 133]. Nevertheless, as shown in Fig. 6.3(a) and 6.3(d), the dispersive wave is only observed for the case of WDA model at 330 nm while it is absent for the case of WIA model. To further examine the source of differences, we extract the soliton spectral components for both cases as highlighted in Fig. 6.3 (b) and 6.3 (e). The insets show the corresponding temporal profiles for the highlighted solitons. For WDA model, the soliton has a central wavelength (defined as the wavelength with highest intensity) of 1591 nm . When transfer to time domain, this corresponds to a $N = 2.8$ soliton with pulse duration of 21.7 fs and peak power of 1.32 GW . For WIA model, the soliton is centered at 1559 nm , corresponding to a 23.5 fs pulse with slightly higher peak power (1.36 GW) and soliton number ($N = 3.4$). Fig. 6.3(c) and 6.3(f) shows the dephasing curves for DW and degenerate four-wave mixing (DFWM) process obtained with [50]:

$$\Delta\beta_{DW}(\omega) = \beta(\omega) - \left[\beta(\omega_{sol}) + \beta_1(\omega_{sol})(\omega - \omega_{sol}) + \frac{\gamma P_{sol}}{2} \right], \quad (6.5)$$

$$\Delta\beta_{DFWM}(\omega) = \beta(\omega_s) + \beta(\omega_i) - 2\beta(\omega_p) + 2\gamma(\omega)P_p. \quad (6.6)$$

In Eq. (6.5), ω_{sol} is the angular frequency of soliton and P_{sol} is the peak power of soliton at maximum compression point. In Eq. (6.6), ω_p , ω_i and ω_s are the angular frequency for pump, idle and signal. P_p is the peak power of the pump soliton.

Figure 6. 3 First row: spectral evolution for 1430 nm pulse propagates in a 5 cm AR-HCF filled with 20 bar xenon obtained with (a) WDA model and (d) WIA model. Second row: spectrum in linear scale at $z = 0.75$ cm for (b) WDA model and (e) WIA model. The soliton is highlighted in green. Last row: DW (blue) and DFWM (red) dephasing curve of the highlighted soliton for (c) WDA model and (f) WIA model.

In Fig. 6.3 (c), the DW emission at 330 nm for WDA model is accurately predicted. Other phase-matching points at $1.06\text{ }\mu\text{m}$, $1.18\text{ }\mu\text{m}$ as well as $1.23\text{ }\mu\text{m}$ on dephasing plot also agree well with the radiation appear on spectral evolution in Fig. 6.3 (a). Similar phase matching positions are also observed for the WIA model as seen from Fig. 6.3 (f) and zero mismatch for DW is also satisfied at 330 nm , but no spectral peak of DW is observed on the spectral evolution, as shown in Fig. 6.3 (d). We attribute this to the weaker spectral overlap between soliton and DW phase-matching point. Note that differences between the dephasing curves for both models are small, this is because inclusion of mode-area dispersion only slightly affects the nonlinear correction term, which has small contribution on the dephasing curves for both DW emission and DFWM process.

Figure 6.4 (a) and 6.4 (b) shows the spectrogram at different positions for WDA and WIA model. The pulse initially undergoes monotonic SPM-induced spectral broadening. Soon after propagating for a short distance, the blue-edge of the spectrum enters the highly dispersive resonance region, where pulse experiences rapid temporal broadening. In the meanwhile, further spectral broadening towards shorter wavelength slows down and eventually ceases, making the spectrum highly asymmetric. When blue edge of the spectrum reaches the resonance region, the dramatically increased dispersion causes the pulse to disperse quickly in time, significantly reducing the pulse intensity. On the other hand, the loss and effective mode area also increases substantially in resonance region. The combination of these effects prohibits spectrum's further extension towards shorter wavelength due to weak nonlinearity.

At 0.75 cm , the pulse reaches the maximum compression point, as indicated by the black dashed line in Fig. 6.3 (a) and 6.3 (d). For the case of WDA model, the spectral intensity of soliton is strong enough to pass through the high loss resonance region (highlighted in yellow) and is sufficiently overlapped with the DW phase-matching point at 330 nm . For the case of WIA model, the spectrum has weaker broadening and is not able to reach the phase-matching

point at 330 nm, thus no spectral peak of DW is observed. Note that the wider spectral broadening for WDA model is achieved despite the fact that the soliton has lower soliton order ($N=2.8$) as compared to that of the WIA model ($N=3.4$).

Figure 6. 4 Spectrogram evolution at different positions for (a) WDA model and (b) WIA model.

This discrepancy can be explained by the wavelength-dependent nonlinear coefficient $\gamma(\omega)$ as shown in Fig. 6.5 and it is what we implement in the WDA model. Since the nonlinear coefficient is inversely proportional to effective mode area $A_{eff}(\omega)$, its profile is a mirror image to that of $A_{eff}(\omega)$ with respect to wavelength axis. The vertical dashed line marks the pump wavelength of 1430 nm. The horizontal red dashed line marks the corresponding nonlinear coefficient at the pump wavelength. In WIA model, the nonlinear coefficient is kept as a constant at this value. As soliton propagates in the red shaded area, the nonlinear coefficient experienced by the soliton in WIA model is always underestimated as compared to that of the WDA model. The higher nonlinear parameter allows soliton in WDA model shift further

towards longer wavelength (central wavelength of 1591 nm at 0.75 cm) as compared to the corresponding soliton in WIA model (central wavelength of 1559 nm at 0.75 cm) due to larger nonlinear phase shift induced by higher nonlinearity. As seen from the GVD profile in Fig. 6.1 (a), further apart from the zero-dispersion wavelength (1450 nm) increases the importance of group-velocity dispersion and thus alleviate the impact of higher-order dispersion [134], resulting in shorter compressed duration for the soliton in WDA model (21.67 fs , as compared to 23.51 fs for WIA model), which supports wider spectral broadening. Moreover, higher nonlinear coefficient also enables more efficient energy transfer by DFWM process [135] to the intermediate wavelength at $1.06 \mu\text{m}$ for WDA model, as compared from Fig. 6.3 (a) and 6.3 (d), which facilitates spectral broadening further towards 330 nm , as shown in the middle panels of Fig. 6.4 (a) and 6.4 (b).

Figure 6. 5 Nonlinear coefficient profile at different wavelengths. Vertical red and black dashed lines indicate position of pump wavelength (1430 nm) and soliton (for WDA model), respectively. Horizontal red dashed line indicates the nonlinear coefficient value evaluated at 1430 nm . Red shaded area is the region of interest with nonlinear coefficient in WDA model higher than that of WIA model.

Figure 6.6 (a) and (b) show the output spectrum of the pulse after propagating in the 5 cm long AR-HCF with different input energies for WDA and WIA model, respectively. The two models show indistinguishable results at low energy level (below 350 nJ), indicating that effect of mode-area dispersion is negligible in this regime. As the input energy increases, notable differences appear for the two cases, implying growing importance of mode-area dispersion.

We also notice the threshold energy required for spectral peak of DW to appear with WIA model (850 nJ) is higher than that of WDA model (740 nJ).

Figure 6. 6 Output spectrum for 1430 nm pump at different input energies for (a) WDA model and (b) WIA model. Both plots are normalized based on maximum output spectrum intensity for WDA model.

6.4.2 Pulse propagation at 800 nm

We further generalize our study by pumping at 800 nm in the second transmission window. This allows us to see how the mode-area dispersion in different regions affect the pulse propagation dynamics in a different way. The other parameters are kept same as before. Figure 6.7 shows the spectrum outputs obtained with the two models. Enhanced emissions at around 1300 nm , 1430 nm , 1620 nm as well as 1840 nm are observed when using WDA model.

Figure 6. 7 Output spectrum for WDA model (black curve) and WIA model pumped at 800 *nm*.

Unlike the previous case, where there is a wide anomalous dispersion region that facilitates soliton propagation. In this case, the pump pulse is bound by resonance bands on both sides. Soliton can only be formed in a narrow region from 800 to 900 *nm*, within the anomalous dispersion regime of the second transmission window. Strong dispersion and high loss are presented outside this range, severely perturbing the pulse and suppressing the intensity. A weak soliton is observed with central wavelength of 889 *nm* and 895 *nm* at distance $z = 1.65$ cm for WDA model and WIA model respectively, as indicated by the arrow in Fig. 6.8 (a) and 6.8 (d). Figure 6.8 (b) and 6.8 (e) show the spectrum in linear scale at $z = 1.65$ cm for WDA and WIA model with the soliton highlighted in green. In time domain, the highlighted part is converted into a higher-order soliton with $N = 4.64$ (peak power is 0.17 GW, pulse duration is 49.6 fs) and $N = 3.54$ (peak power is 0.16 GW, pulse duration is 40.5 fs) for WDA and WIA model, respectively. Figure 6.8 (c) and (f) show the dephasing profiles for DW emission and DFWM process induced by the soliton highlighted in Fig. 6.8 (b) and (e). Multiple zero-mismatched wavelengths are achieved with both models. Some of them can be identified on the spectral evolution profiles in Fig. 6.8 (a) and 6.8 (d), such as the closely distributed radiation bands in the first resonance region as well as the radiation at 1.68 μm . Their positions match well with those predicted on the dephasing plots in Fig. 6.8 (c) and 6.8 (f). Other radiations

such as the $2\ \mu\text{m}$ band are induced through four-wave mixing processes as we shall see later.

Figure 6. 8 First row: spectral evolution for 800 nm pulse propagates in a 5 cm AR-HCF filled with 20 bar xenon obtained with (a) WDA model and (d) WIA model. Second row: spectrum in linear scale at $z = 1.65\ \text{cm}$ for (b) WDA model and (e) WIA model. The soliton is highlighted in green. Last row: DW (blue) and DFWM (red) dephasing curve of the highlighted soliton for (c) WDA model and (f) WIA model.

We also plot the spectrogram for the case of WDA and WAI model, as shown in Fig. 6.9 (a) and (b), respectively. For both WDA and WIA model, the pulse initially experiences self-phase modulation induced spectral broadening. At $z = 0.85 \text{ cm}$, the red edge of spectrum reaches around $1.17 \mu\text{m}$, which is within the first resonance band (the wide yellow area) while the blue edge of spectrum continues to shift and pass through the second resonance band (the narrow yellow area). At the same time, weak radiation emerges at $2 \mu\text{m}$ for both cases. When the pulse propagates to $z = 1.05 \text{ cm}$, the red edge of spectrum is pushing further into the first resonance band and more energy is transferred to around 1.9 to $2.2 \mu\text{m}$, resulting in a stronger radiation band centered at around $2 \mu\text{m}$ in the anomalous dispersion regime. This radiation is possibly caused by four-wave mixing process induced by pump source in the first resonance band at around $1.17 \mu\text{m}$ but still need further verification. As pulse reaches $z = 1.65 \text{ cm}$, the soliton (as indicated by arrows) generates a new wavelength component centered at $1.68 \mu\text{m}$ through DFWM process. Intuitively, the newly generated band at $1.68 \mu\text{m}$ is temporally overlapped with the pump soliton for both models, as shown in the last panels on Fig. 6.9 (a) and (b). Similarly, the $2 \mu\text{m}$ radiation is also temporally overlapped with the pump source located in the first resonance band for both models.

Figure 6.9 spectrogram evolution at different positions for (a) WDA model and (b) WIA model

Due to the complex pulse dynamics under severe perturbation by resonance bands, the phase-matching conditions for the multiple radiation bands in the first transmission window, such as the $2\ \mu\text{m}$ radiation, cannot be explicitly determined. To verify our previous assumption that these radiation bands are generated by DFWM processes originate from the first resonance region at around $1.17\ \mu\text{m}$, we locate the pump wavelength to be between $1170\ \text{nm}$ to $1180\ \text{nm}$ and calculate the DFWM-induced phase-matched wavelengths by scanning through a wide range of peak power from $0.2\ \text{MW}$ to $15\ \text{MW}$. Since there could be multiple phase-matched wavelengths with resonance structures, we only select the longest phase-matched idle wavelength. The result is shown in Fig. 6.10.

Figure 6.10 DFWM-induced phase-matched idle wavelength at different peak power ranges from 0.2 to 15 MW. The pump wavelength is shown beside the corresponding curve.

As we can see, the phase-matched idle wavelength increases with decreasing pump source wavelength ω_p and increasing peak power P_p in this region. Since the pump source is in the first resonance region, even 1 nm difference of pump source wavelength could result in large shift of the idle wavelength. As the pump source goes deeper into the first resonance region with increasing wavelength, the phase-matched wavelength changes more abruptly. According to Fig. 6.10, we deduce the pump source that is responsible for the radiation band centered at 2 μm (spans from 1.9 μm to 2.2 μm) has wavelength between 1173 to 1177 nm (within the pink region) with an average wavelength of 1175 nm, as expected. We also check the phase-matching conditions with other ω_p values and therefore confirm that it is not possible to generate 2 μm radiation band induced by DFWM outside this wavelength range. There are also some weaker radiations locate between 1.4 to 1.6 μm from both models, as shown in Fig. 6.8 (a) and (d). We attribute these radiations caused by DFWM process too, with slightly longer ω_p at 1177 to 1179 nm (within the cyan region). As pulse propagates, it continues to push the red edge of spectrum deeper into the first resonance band and ω_p further increases a bit and as

a result, the idle wavelength shifts towards shorter wavelength. However, due to exponentially increasing loss and substantially reduced nonlinearity, further spectral broadening is largely impeded and conversion efficiency of DFWM reduces. This also explains why the $2\ \mu\text{m}$ radiation band appears first and radiation bands with shorter wavelength appear later with weaker intensity.

By comparing Fig. 6.9 (a) and (b), we observe that the $2\ \mu\text{m}$ radiation at $z = 1.05\ \text{cm}$ for WDA model is weaker than that of the WIA model. On the contrary, the $1.68\ \mu\text{m}$ radiation at $z=1.65\ \text{cm}$ for WDA model is stronger than that of the WIA model. To investigate the source of difference, Figure 6.11 shows the same nonlinear coefficient profile with the reference line taken from the new pump wavelength at $800\ \text{nm}$.

Figure 6. 11 Nonlinear coefficient profile at different wavelengths. Vertical red and black dashed lines indicate position of pump wavelength ($800\ \text{nm}$) and ω_p , respectively. Grey dashed line marks the separation of section 1 and section 2. Horizontal red dashed line indicates the nonlinear coefficient value evaluated at $800\ \text{nm}$. Red (green) shaded area is the region of interest with nonlinear coefficient in WDA model higher (lower) than that of WIA model.

The green shaded area indicates the region in the first resonance band with nonlinear coefficient in WDA model lower than that of the WIA model. The highlighted soliton in Fig 6.8 (b) and (e) locates in the red region where the nonlinear coefficient in WDA model is higher than that of the WIA model. Since the nonlinear coefficient at $1175\ \text{nm}$ in the WDA model is well below that of the WIA model, as labeled on Fig. 6.11, there is less conversion efficiency for DFWM process and thus, weaker idle radiation at $2\ \mu\text{m}$ as compared to WIA model. The nonlinear

coefficient at soliton central wavelength of 889 nm in WDA model is higher than that experienced by the soliton in WIA model, hence more energy is transferred to $1.68 \mu\text{m}$ in the WDA model through DFWM process by the soliton, due to higher conversion efficiency.

We also separate the nonlinear coefficient profile into three sections as indicated in Fig. 6.11. The third and fourth resonance band are included in section1. The second and first resonance band are included in section 2 and section 3, respectively. Figure 6.12 (a), (b) and (c) show the simulation results when include only section 1, section 2 and section 3 of the wavelength-dependent nonlinear coefficient profile respectively while replace the rest part with constant value evaluated at 800 nm , as indicated by the red horizontal dashed line. This is to individually examine the impact of mode-area dispersion at each resonance region. We found that when we only consider the mode-area dispersion in section 1, the spectral evolution is almost identical to the WIA model, as seen from Fig. 6.8 (d) and Fig. 6.12 (a). We can then deduce that the mode-area dispersion near third and fourth resonance band have negligible impact on the pulse propagation dynamics. This is because the third and fourth resonance bands are further away from the initial pump at 800 nm as well as their resonance is narrower and weaker. Inclusion of mode-area dispersion in the first or second resonance band affect both the nonlinear dynamics in the first transmission window (1400 to 2450 nm) and second transmission window (550 to 900 nm), as compared Fig. 6.12 (b) (c) to 6.12 (a).

Figure 6. 12 (a) (b) and (c) shows the spectral evolution obtained when include section 1,2 and 3 of nonlinear coefficient curve in Fig. 6.11 and replace the rest parts with value indicated by horizontal red dashed line.

Figure 6.13 (a) and (b) show the spectrum output of the 800 *nm* pump pulse after propagating for 5 cm with different input energies for WDA and WIA model, respectively. Like the case of 1430 *nm* pump, the differences between WDA and WIA models are small at low energy level (below 400 *nJ*). As input energy increases, the deviation become more pronounced. The radiations in the first transmission window induced by DFWM process are completely different for the WDA and WIA models at all energy levels, indicating that they are highly sensitive to the mode-area dispersion. This makes sense since they are induced by pump sources in the first resonance region, where the mode-area changes dramatically.

Figure 6. 13 Output spectrum for 800 *nm* pump at different input energies for (a) WDA model and (b) WIA model, both plots are normalized based on maximum output spectrum intensity for WDA model.

6.5 Conclusion

In this chapter, we numerically investigate the effect of mode-area dispersion on pulse propagation dynamics in a small-core anti-resonant hollow-core fiber. The pulse is launched at two different pump wavelengths--1430 *nm* and 800 *nm* near resonance bands and propagate in different transmission windows. We compare the results obtained with two different models,

yield the wavelength-dependent area (WDA) model and wavelength-independent area (WIA) model. The former one includes the effect of mode-area dispersion while a constant mode area is assumed for the latter one. When pumping the pulse at 1430 nm , a dispersive wave is emitted at 330 nm for the WDA model while it is absent in the WIA model under the same pulse parameters. We identified that the differences come from the nonlinear parameter experienced by the soliton that induces the dispersive wave generation. In WDA model, the soliton experiences a smaller effective mode area and thus higher nonlinearity at its central wavelength, which causes larger nonlinear phases shift and moves the soliton to longer wavelength, suppressing the perturbation by higher-order dispersion and resulting in shorter pulse duration that supports wider spectrum. The higher nonlinearity also leads to higher frequency conversion efficiency to the intermediate wavelength at $1.06\text{ }\mu\text{m}$ through DFWM, as compared to the case of WIA model, facilitating the spectral broadening further towards 330 nm . The combined effects of both enable the emission of DW spectral peak with an input energy lower than the threshold energy required in a WIA model. When pumping the pulse at 800 nm , the WDA and WIA model result in significantly different radiation spectrums in the first transmission window. Among these, we have identified that the radiation at $1.68\text{ }\mu\text{m}$ appears in both cases is generated through DFWM process by a pump soliton (centered at 891 nm for WDA model and 895 nm for WIA model) while the rest of radiation bands are induced by DFWM process with pump source located in the first resonance band. The strength of these radiations is related to the nonlinear coefficient at the wavelength of the corresponding pump source, which influences the conversion efficiency of the DFWM process. In WIA model, the nonlinear coefficient for the pump soliton is underestimated while that for the pump source in the first resonance band is overestimated as compared to the actual values in WDA model. Therefore, in the WIA model we observe a lower intensity for the soliton induced radiation at $1.68\text{ }\mu\text{m}$ and stronger radiation at $2\text{ }\mu\text{m}$ as compared to the counterparts in WDA model. Our

results suggest that inclusion of mode-area dispersion is important especially for investigating ultrafast dynamics near resonance bands and at high energy levels, such as when investigating resonance-enhanced supercontinuum generation [26, 48].

Chapter 7: Conclusion and Recommendations

7.1 Conclusion

Detailed studies on pulse propagation dynamics in gas-filled anti-resonant hollow-core fibers and gas-filled capillaries are carried out.

Firstly, the effect of stretching-induced initial chirp on soliton-effect compression is systematically studied with a simplified NLSE model. The key compression parameters such as shortest pulse duration, highest pulse peak power, compression length, as well as pulse quality across a series of chirp values are obtained. Enhancement of compression can be found at both positive and negative chirp. The optimal positive and negative chirp values are located for different soliton order. The amount of optimal positive chirp decreases with increasing soliton order while it is opposite way for optimal negative chirp. Wide tunability of compression length with initial chirp is observed. The ratio between the compression length for pulses with optimal positive chirp and zero chirp is a fixed value of 1.23.

Secondly, soliton dynamics and MIR supercontinuum with different pressurization configurations in the fundamental core mode of a xenon-filled anti-resonant hollow-core fiber is investigated numerically using the UPPE model. As compared to constant pressure of 27.3 bar, a decreasing pressure from 41 to 0 bar increases the soliton-effect compression by 1.63 times with enhanced blue edge of the spectrum while an increasing pressure from 0 to 41 bar degrades the compression performance. The enhancement of soliton-effect compression in the MIR region with decreasing pressure is not reported before and its mechanism is analysed. Such system is highly resistant to ionization effect.

Thirdly, investigation on effect of decreasing pressure is extended to higher-order modes of a gas-filled capillary. Pulses is predominantly launched into the HE_{12} , HE_{13} and HE_{14} mode and pulse propagation is modelled using the MM-GNLSE. As compared to the case of constant

pressure, four times enhancement on soliton-effect compression is achieved in HE_{12} mode when a decreasing pressure is applied, where the pulse reaches sub-cycle duration of 1.85 fs. The accuracy of the results is verified against that obtained with the UPPE model. A decreasing pressure also introduces axially reducing nonlinear parameter and largely alleviates the intermodal nonlinear coupling as pulse compresses, which is an additional benefit for pulses propagates in the higher-order modes.

Lastly, the effect of mode-area dispersion on pulse propagation dynamics in a small-core AR-HCF is numerically investigated at two different pump wavelengths, namely 800 nm and 1430 nm. The pulse propagation is modelled using a modified GNLSE. The simulation results obtained with and without effect of mode-area dispersion are compared. The differences are analysed in detail, with the origin comes from the strong wavelength dependence of nonlinear parameters. Impact of mode-area dispersion become prominent when pumping near the resonance regions and its importance grows with higher input energy.

7.2 Recommendations for further research

7.2.1 Soliton-effect compression in second-order vector mode of gas-filled anti-resonant hollow-core fibers

Although higher-order modes (HOM) in gas-filled capillaries have been widely investigated, studies on soliton dynamics in gas-filled AR-HCFs is only limited to the fundamental core mode. In fact, ultrafast pulse propagating in HOM of gas-filled AR-HCF is favoured for many applications. Vector modes that belong to the LP_{11} group in AR-HCFs is one of the examples. Since intensity profiles in these modes have doughnut shape with unique polarization distribution, they are very attractive for applications such as microscopy [46], plasmonic focusing [136], particle trapping and manipulation [137] as well as laser processing [47]. There are four vector modes in the LP_{11} group, namely TE_{01} , TM_{01} and a degenerate pair-- HE_{12} (even)

and HE_{12} (odd), as shown in Fig. 7.1.

Figure 7. 1 (a) TE_{01} (b) TM_{01} (c) HE_{21} (even) and (d) HE_{21} (odd). The red arrow indicates the electric vector distribution.

By properly designing the fiber parameters, fundamental core mode can be largely suppressed with high distinction ratio between the second-order vector modes and fundamental core mode [138]. The propagation constant can be determined by FEM.

7.2.2 Scaling rule for high quality soliton-effect pulse compression in gas-filled anti-resonant hollow-core fibers filled with decreasing pressure

Anti-resonant hollow-core fibers enable generation of few-fs pulses across wide range of pulse energies (sub μJ to few mJ [69, 139, 140]) and wavelengths (spanning from UV to MIR). Such great degree of freedom is attributed to an important property known as scale-invariance [2]. Ref [2] shows that similar pulse propagation dynamics can be achieved with significantly different input parameters (such as pump wavelength/energy, gas pressure/species or fiber core-

size) under a proper scaling rule. Moreover, as pulse compresses in gas-filled hollow waveguides, the shortest pulse duration is ultimately limited by several factors such as higher-order dispersion, plasma effect, fiber loss, modulational instability as well as self-focusing. For decreasing pressure, we also need to consider additional factors, such as the compression length. We need to ensure it is close to fiber output end so that the pulse can exist the fiber when it is compressed to the shortest duration.

To control the soliton dynamics, improve the pulse compression ratio and pulse quality of soliton-effect compression with decreasing pressure, it is important to determine the scaling rule with consideration of these factors for selection of optimized pulse and fiber parameters. Current available scaling rule is only applicable for pulse propagating in the fundamental core mode of AR-HCFs filled with constant pressure [141]. There is lack of study on scaling rules for AR-HCFs filled with decreasing pressure.

List of Publications

Journal article

Y. Wan, M. I. Hasan and W. Chang, "Impact of mode-area dispersion on nonlinear pulse propagation in gas-filled anti-resonant hollow-core fiber," *Photonics*, vol. **9**, no. 1, 2022, DOI: 10.3390/photonics9010025.

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