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Four-wave mixing: from broadband wavelength
conversion in optical fibers to free space pump and
probe experiments.

by

Jing Su

A Dissertation
Presented to the Graduate Committee
of Lehigh University
in Candidacy for the Degree of
Doctor of Philosophy
in
Physics

Lehigh University
Jan 2022

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Jing Su

Approved and recommended for acceptance as a dissertation in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

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Four-wave mixing: from broadband wavelength conversion in optical fibers to free space pump and probe experiments.

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Abstract

We show that appropriately designed dual core fibers can be used to enable coupling-length phase matching (CLPM, [1]) and high efficiency nonlinear optical frequency-downconversion by solely using the third-order nonlinearity of the glass. The dual core configuration provides enhanced phase-matching possibilities to extend the generated wavelength further into infrared. We analyze the effects of phase-modulation with an intensity-dependent correction term. Then, we show that one particular phase-matching choice can theoretically lead to 100% photon-conversion efficiency, which is unaffected by self- and cross-phase modulation and able to maximize the efficiency of the process. Fiber-based frequency converters can be designed for different pump wavelengths, to generate longer wavelength radiation based on a quasi-degenerate four-wave-mixing scheme where energy is transferred from a pump wave at a frequency ω_2 to two other waves with large frequencies $\omega_1 < \omega_2$ and $\omega_3 > \omega_2$. The increased interaction length allowed by a phase-matched interaction naturally leads to the ability of using meter-long fibers to compensate for low third-order susceptibilities. We apply our analysis to the example of silica and fluoride fibers, and wavelength conversion schemes from pump wavelengths of 1.55 or 1.3 μm to further infrared wavelengths in the 2-5 μm range.

We report an experimental study and modeling of instantaneous and temporal response of nonlinear optical organic molecules using pump-and-probe Degenerate Four-wave Mixing (DFWM) method. A theoretical model is developed to accurately model the mechanism and formation of transient population gratings. The relationship between instantaneous nonlinear optical response and the response of population gratings is also discussed. This theoretical modeling can help measure

and calculate the lifetimes of short-lived population gratings from experimental data. The lifetime of nonlinear optical organic molecules that we study is measured to be a few picosecond for this work.

Chapter 1

Introduction to multi-wave mixing

In general, any nonlinear optical effect can be modeled by solving the wave equation under specific assumptions and boundary conditions. The general form of the nonlinear optical (NLO) wave equation [2–4] is written as

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \bar{\epsilon} \cdot \mathbf{E} = -\frac{1}{c^2} \frac{\partial^2}{\partial t^2} \frac{1}{\epsilon_0} \mathbf{P}^{(NL)} \quad (1.1)$$

where $\mathbf{P}^{(NL)}$ is the nonlinear polarization within the material due to all nonlinear optical interactions and $\bar{\epsilon}$ is the dielectric tensor. The right hand side of the equation can be thought of as a source term that couples waves of different frequencies and wavevectors. The nonlinear polarization can be expanded in a Taylor series as

$$\mathbf{P}^{(NL)} = \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \dots + \mathbf{P}^{(N)} + \dots \quad (1.2)$$

The third-order nonlinear polarization can be written in terms of powers of the electric field as

$$\mathbf{P}^{(3)} = \epsilon_0 \chi^{(3)} \mathbf{E} \cdot \mathbf{E} \cdot \mathbf{E} \quad (1.3)$$

where $\chi^{(3)}$ is the third-order nonlinear susceptibility.

The propagation of light in a nonlinear medium is governed by wave equation, which was derived from Maxwell's equations (1.1). Four-wave mixing is a third-order nonlinear interaction to a fourth signal beam that occurs in the presence of three optical beams in a medium [5–9]. The fourth signal beam is radiated by the

third-order nonlinear polarization $\mathbf{P}^{(3)}$ (1.3). In this work, we study the coupled-wave equations of nonlinear third-order frequency downconversion in the process of $\omega_1 + \omega_2 - \omega_3 \rightarrow \omega_4$. The coupled-wave equations are as follows:

$$\begin{aligned} \frac{\partial}{\partial z} A_1 = & i \chi [A_1(A_1 A_1^* + 2A_2 A_2^* + 2A_3 A_3^* + 2A_4 A_4^*) \\ & + 2A_2^* A_3 A_4 e^{-i\vec{\Delta}k \cdot \vec{z}}], \end{aligned} \quad (1.4)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_2 = & i \chi [A_2(A_2 A_2^* + 2A_1 A_1^* + 2A_3 A_3^* + 2A_4 A_4^*) \\ & + 2A_1^* A_3 A_4 e^{-i\vec{\Delta}k \cdot \vec{z}}], \end{aligned} \quad (1.5)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_3 = & i \chi [A_3(A_3 A_3^* + 2A_1 A_1^* + 2A_2 A_2^* + 2A_4 A_4^*) \\ & + 2A_1 A_2 A_4^* e^{i\vec{\Delta}k \cdot \vec{z}}], \end{aligned} \quad (1.6)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_4 = & i \chi [A_4(A_4 A_4^* + 2A_1 A_1^* + 2A_2 A_2^* + 2A_3 A_3^*) \\ & + 2A_1 A_2 A_3^* e^{i\vec{\Delta}k \cdot \vec{z}}], \end{aligned} \quad (1.7)$$

where $\vec{\Delta}k = \vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4$ is the spatial wave-vector mismatch. (See Chapter 3 for an explanation of the symbols χ and A_i .)

In free space, we investigate degenerate four-wave mixing (DFWM) with $\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega$ in the process of $2\omega - \omega \rightarrow \omega$ and waves travel in different directions [10–16]. The idea comes from the fact that the nonlinear optical coupled-wave equations are the same for both optical fibers and for space-DFWM. We will report an experimental study and modeling of instantaneous and temporal response of nonlinear optical organic molecules using pump-and-probe method. The results of DFWM experiment and the theoretical model are presented in Chapter 5.

For the four-wave mixing in optical fibers [17–21], we consider the propagation direction of multi-wave with different frequencies is the same. In Chapter 2, we reviews basics and fundamentals of beam propagation in a single cylindrical dielectric waveguide, as well as two parallel cylindrical dielectric waveguides. Chapter 3 presents nonlinear third-order frequency downconversion in a single-mode fiber, as a simpler system to discuss first. The effect of phase modulation on the phase matching condition and the conversion efficiency are also presented. This chapter serves

as a preparation for the work on dual-core configuration. We show the limitations of using single-mode fiber and the flexibility with dual-core fiber, which is discussed extensively in Chapter 4.

The coupling between the cores in dual core optical glass fibers can compensate for the natural bulk dispersion of the material and lead to flexible phase matching opportunities for third-order nonlinear optical interaction [22]. The availability of appropriate phase-matching schemes allows the design of fiber-based frequency converters where a relatively weak third-order susceptibility is compensated by a long interaction length, an attractive opportunity for the implementation of new fiber-based frequency conversion schemes [23–26]. Even though various types of glasses do not have a specially large third-order nonlinear optical susceptibility, the very low loss that can be achieved in optical fibers would in principle compensate for the low nonlinearity by enabling very large interaction lengths, if not for refractive index dispersion. Dispersion and the corresponding phase-mismatch limits the interaction length between the interacting waves.

We consider third-order nonlinear optical interaction in a fiber for the generation of longer wavelengths, as an example to bridge gaps in the availability of laser sources in farther infrared, for example in coincidence with atmospheric transmission windows, where lasers could be employed for infrared backscatter imaging spectroscopy [27] or other investigations. We will focus on the third-order nonlinear optical process that leads to the generation of a longer wavelength thanks to the interaction of three waves at frequencies $\omega_3 > \omega_2 > \omega_1$. This three-wave mixing scheme can be seen as a third-order difference frequency generation process described by $2\omega_2 - \omega_3 \rightarrow \omega_1$, or a third-order parametric down conversion process $2\omega_2 \rightarrow \omega_1 + \omega_3$. It is the third-order equivalent of the second-order interactions used in standard Optical Parametric Generation (OPG), Optical Parametric Oscillation (OPO), Parametric Fluorescence, or Parametric Down-conversion.

While three- and four-wave mixing in fibers has been treated extensively in the literature, that work concentrated on the case of small frequency differences. An equivalent discussion targeted towards the generation of new wavelengths far away from the pump wavelengths has been discussed much more rarely. Among

the various known phase matching possibilities for the third-order process we are interested in, we discount quasi-phase matching by spatial structuring the material because it would be inefficient, and difficult to control over long fiber lengths. For particular materials that have specific bulk refractive index dispersion that already allows for phase-matching for certain third-order nonlinear optical interactions, it is possible to translate such phase-matching conditions for a specific set of wavelengths to single-core fibers [28] and to slightly tune the phase-matched wavelengths by modifying the radius of the fiber core. But this method is still not flexible enough, in particularly limiting the choice of pump wavelengths. Coupling-length phase matching (CLPM), on the other hand, is a flexible way to achieve the required phase matching conditions by arranging two parallel guiding cores in an optical fiber in such a way as to obtain sufficient coupling of the optical waves between the cores [1]. In this situation the dual core structure gives each interacting waves two nearby eigenmodes with similar propagation constants that can be tuned by the distance between the cores, and therefore multiplies the possibilities to achieve phase matching.

In Chapter 6, we present that numerical simulations of guided-wave propagation, conducted from COMSOL [29]. We verify the results of single-mode fiber in chapter 3 and dual-core fiber in chapter 4, then discuss the deviations from the theoretical method. Finally, we show an experimental attempt to demonstrate the predicted results from previous chapters with dual-core silica fibers from collaborations. Details will be presented in chapter 7.

Chapter 2

Review of cylindrical dielectric waveguide

In this chapter, we review the basic concepts and equations of electromagnetic wave theory for wave propagation in cylindrical waveguides [30]. An optical fiber is a cylindrical dielectric waveguide made of a low-loss material, such as silica glass. Step-index fibers with constant refractive indexes in the core and cladding are discussed in Section 2.1. When the core diameter is small, only a single mode is supported and the fiber is said to be a single-mode fiber. The wave propagation in two parallel, coupled cylindrical dielectric waveguides follow in Section 2.2. A brief introduction of coupled mode theory is also provided.

2.1 Review of propagation in a cylindrical dielectric waveguide

2.1.1 Step-index fiber

A step-index fiber is a cylindrical dielectric waveguide specified by the refractive indexes of its core and cladding, n_1 and n_0 , respectively. The radius of the core is a in Figure 2.1. The relative refractive-index difference between n_1 and n_0 is defined

as

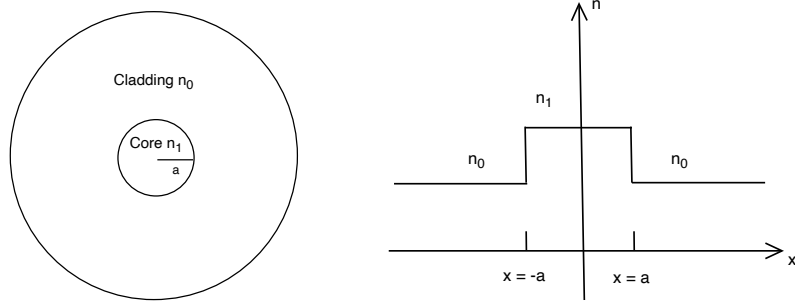


Figure 2.1: Basic structure and refractive-index profile of a step-index optical fiber.

$$\Delta = \frac{n_1^2 - n_0^2}{2n_1^2}. \quad (2.1)$$

Δ is commonly expressed as a percentage. We generally consider a constant, small refractive index contrast $\Delta n = n_1 - n_0$ that gives $\Delta \ll 1$. The numerical aperture (NA), which is also known as the maximum light acceptance angle of the waveguide, is related to the relative refractive-index difference Δ by

$$NA = n_1 \sqrt{2\Delta}. \quad (2.2)$$

And we also introduce the normalized frequency, and it is expressed by

$$V = k_0 n_1 a \sqrt{2\Delta}, \quad (2.3)$$

where $k_0 = \omega/c = 2\pi/\lambda$, with λ the vacuum wavelength of the electromagnetic wave.

We consider an electromagnetic wave having angular frequency ω and propagating in the z direction with propagation constant β . The electric and magnetic fields in optical fiber can be expressed in cylindrical coordinates as

$$\mathbf{E} = E(r, \theta) e^{j(\omega t - \beta z)}, \quad (2.4a)$$

$$\mathbf{H} = H(r, \theta) e^{j(\omega t - \beta z)}, \quad (2.4b)$$

where r denotes the position in the plane transverse to the z -axis. Each of the components of the monochromatic electric and magnetic fields obeys the Helmholtz equation [31, 32], $\nabla^2 U + n^2 k_0^2 U = 0$. In cylindrical coordinate system the Helmholtz equation is written as

$$\frac{\partial^2 U}{\partial r^2} + \frac{1}{r} \frac{\partial U}{\partial r} + \frac{1}{r^2} \frac{\partial^2 U}{\partial \theta^2} + \frac{\partial U}{\partial z} + k_0^2 n^2 U = 0, \quad (2.5)$$

where $U = U(r, \theta, z)$. Substituting

$$U(r, \theta, z) = R(r) e^{-jl\theta} e^{-j\beta z}, \quad l = 0, \pm 1, \pm 2, \dots \quad (2.6)$$

into Equation (2.5) leads to an ordinary differential equation for the radial profile $R(r)$:

$$\frac{\partial^2 R}{\partial r^2} + \frac{1}{r} \frac{\partial R}{\partial r} + (k_0^2 n^2 - \beta^2 - \frac{l^2}{r^2}) R = 0. \quad (2.7)$$

In axially symmetric optical fibers, the refractive-index distribution is only dependent of r . Here, we only discuss in detail about electromagnetic fields, dispersion equations, and propagation characteristics for step-index fibers as shown in Figure 2.1, which has a uniform refractive index in both core and cladding. The wave is guided (or bounded) if the propagation constant is smaller than the wavenumber in the core and greater than the wavenumber in the cladding. Then the following condition should be satisfied:

$$n_0 \leq \beta/k_0 \leq n_1, \quad (2.8)$$

β/k_0 is a dimensionless value and is a refractive index itself for the plane wave. Therefore it is called the effective index and is usually expressed as

$$n_{eff} = \frac{\beta}{k_0}. \quad (2.9)$$

Here we introduce the normalized propagation constant

$$b = \frac{n_{eff}^2 - n_0^2}{n_1^2 - n_0^2}. \quad (2.10)$$

Then the conditions for the guided modes are expressed, from Equation (2.8), by

$$0 \leq b \leq 1. \quad (2.11)$$

It is convenient to define

$$\alpha = \sqrt{k_0^2 n_1^2 - \beta^2}, \quad (2.12a)$$

$$\sigma = \sqrt{\beta^2 - k_0^2 n_0^2}, \quad (2.12b)$$

so that for guided waves α and σ are real. Equation (2.7) may be written in the core and cladding separately:

$$\frac{\partial^2 R}{\partial r^2} + \frac{1}{r} \frac{\partial R}{\partial r} + (\alpha^2 - \frac{l^2}{r^2})R = 0, \quad r < a \text{ (core)}, \quad (2.13a)$$

$$\frac{\partial^2 R}{\partial r^2} + \frac{1}{r} \frac{\partial R}{\partial r} + (\sigma^2 - \frac{l^2}{r^2})R = 0, \quad r > a \text{ (cladding)}. \quad (2.13b)$$

The solutions are the family of Bessel functions. We can obtain the bounded solutions:

$$R(r) \propto \begin{cases} J_l(u), & r < a \text{ (core)}, \\ K_l(w), & r > a \text{ (cladding)}, \end{cases} \quad (2.14)$$

where

$$\begin{cases} u = \alpha a, \\ w = \sigma a. \end{cases} \quad (2.15)$$

$J_l(x)$ is the Bessel function of the first kind and order l , and $K_l(x)$ is the modified Bessel function of the second kind and order l . The normalized transverse wavenumbers u and w are related by the following equations:

$$u^2 + w^2 = V^2. \quad (2.16)$$

2.1.2 Modes

We now consider the boundary conditions. At the core-cladding boundary $r = a$ establishes a relationship between the coefficients of proportionality in Equation (2.14). The hybrid modes in optical fiber are under the condition that the axial electromagnetic field components E_z and H_z are not zero. Solutions for Equation (2.5) are given by the product of n th-order Bessel functions. This would make the derivations more

complicated, so here we only present the results of dispersion equation [30, 33, 34]

$$\left[\frac{J'_n(u)}{uJ_n(u)} + \frac{K'_n(w)}{wK_n(w)}\right]\left[\frac{J'_n(u)}{uJ_n(u)} + \left(\frac{n_0}{n_1}\right)^2 \frac{K'_n(w)}{wK_n(w)}\right] = n^2\left(\frac{1}{u^2} + \frac{1}{w^2}\right)\left[\frac{1}{u^2} + \left(\frac{n_0}{n_1}\right)^2 \frac{1}{w^2}\right] \quad (2.17)$$

The propagation constant of the hybrid modes is calculated by solving Equation (2.17). Most fibers are weakly guiding ($\Delta \ll 1\%$). The linear polarized (l, m) mode is usually denoted as the LP_{lm} mode. The dispersion equations of the LP modes can be expressed in the unified form [30, 33, 34]

$$\frac{J_m(u)}{uJ_{m-1}(u)} = -\frac{K_m(w)}{wK_{m-1}(w)}. \quad (2.18)$$

Since they are not the strict modes, the eigenvalues are slightly different in the rigorous dispersion equations. The characteristic equation may be solved graphically by plotting its right- and left- hand sides from Equation (2.18) and finding the intersections. Each intersection point corresponds to a fiber mode with a distinct value of u . If we insert the definition of u and w in (2.15), together with the definition of V in (2.3) and b in (2.10), then the equation should become an equation that only depends on b and V . $b(V)$ plot does not depend on the refractive index or the core radius of the fiber, which is universal.

The dependence of the normalized propagation constant b on the V -parameter for different modes is shown in Figure 2.2. As introduced earlier, the normalized propagation constant b involves the relationship between the effective refractive index in the mode and the refractive index of core and cladding. The effective refractive index is also denoted as the eigen solution for each mode by solving (2.18). The fundamental mode LP_{01} always exist in optical fiber so that the normalized propagation constant b starts from zero in the plot.

As the V -parameter increases, i.e., the frequency increases or the fiber diameter increases, the effective refractive index n_{eff} increases from n_0 to n_1 . This is expected since the mode is more confined in the core at shorter wavelength. The buildup for the second higher mode LP_{11} is when $V > 2.405$. This is also the cut off condition for the existence of the second higher mode. When $V < 2.405$, all modes with the exception of the fundamental mode LP_{01} (or HE_{11}) are cut off. The fiber then

operates as a single-mode waveguide. The condition for single-mode operation is therefore $V < 2.405$. Since V is proportional to the optical frequency, the cut off condition for the fundamental mode yields a corresponding cut off frequency f_c . When $f < f_c$, the second higher mode LP_{11} disappears, which leaves only the fundamental mode.

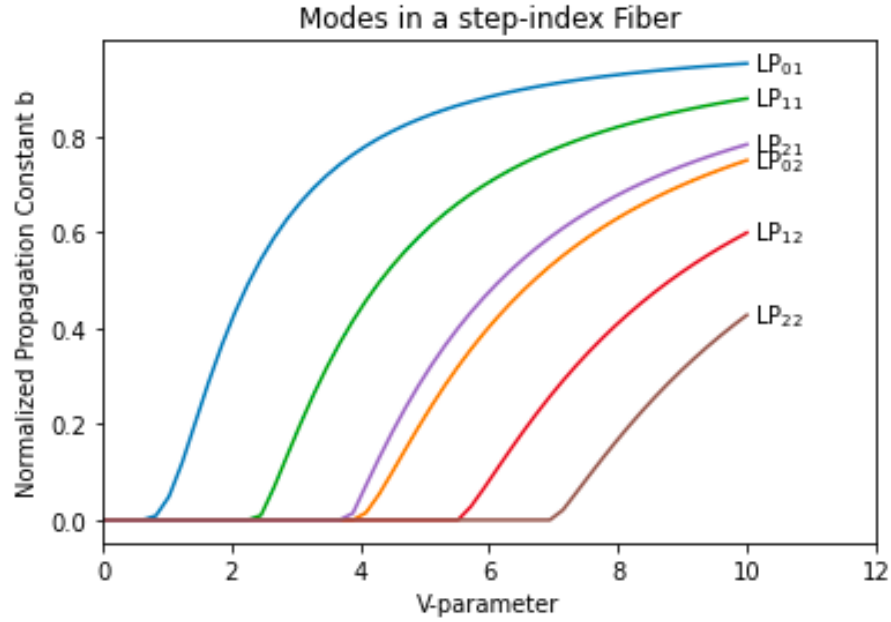


Figure 2.2: Normalized propagation constant b as a function of the V -parameter for several LP_{lm} modes.

2.2 Review of propagation in two parallel cylindrical dielectric waveguides

If two parallel waveguides are sufficiently close such that their fields overlap, light can be coupled from one to another. Optical power can then be transferred between two waveguides. The basic principle of waveguide coupling is presented in this

section.

2.2.1 Dual-core step-index fiber

We consider two parallel waveguides made of two cores with radius r , separation distance D (center to center) in a step-index optical fiber, as shown in Figure 2.3(a). Each of the waveguides is assumed to be single-mode. The separation between the waveguides is such that the optical field outside the core (in the absence of the other) overlaps slightly with the other core. Figure 2.3(b) shows that light is mostly in core 1 at $z = 0$ and at $z = L$ it is mostly in core 2. Coupling can be regarded as a scattering effect. The field of core 1 is scattered from core 2, creating a source of light that changes the amplitude of the field in core 2. The field of core 2 has a similar effect on core 1. When the electromagnetic field distributions after mode coupling do not differ substantially from those before coupling, the propagation characteristics of the coupled waveguides can be analyzed by the perturbation method [35, 36].

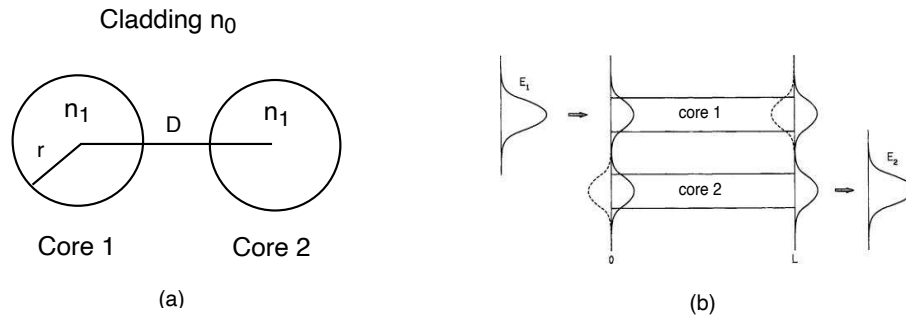


Figure 2.3: (a). Dual core optical fiber with refractive index n_1 in the core and n_0 in the cladding. The radius is r for each core and the separation distance is D from center to center. (b). Coupling between two parallel waveguides. Even mode (solid line) and odd mode (dashed line) are presented.

When we neglect the higher-order modes and only consider the fundamental mode, the electric field can be approximated by the summation of the even mode and the odd mode:

$$E(x, z) = E_e(x)e^{-j\beta_e z} + E_o(x)e^{-j\beta_o z}, \quad (2.19)$$

where $E_e(x)$ and β_e denote electric field and propagation constant of the even mode and $E_o(x)$ and β_o denote those of the odd mode, respectively. The incident electric field, which is coupled to core 1 at $z = 0$, is expressed by

$$|E(x, 0)| = |E_e(x) + E_o(x)| = E_1(x). \quad (2.20)$$

Here $E_1(x)$ denotes the eigen mode of core 1. When the two cores are not so close, this expression holds with good accuracy. The electric field amplitude at z is given as

$$|E(x, z)| = |E_e(x) + E_o(x)e^{j(\beta_e - \beta_o)z}|. \quad (2.21)$$

Electric field distribution at $z = \pi/(\beta_e - \beta_o)$ is then given by

$$|E(x, z)| = |E_e(x) - E_o(x)| = E_2(x), \quad (2.22)$$

where $E_2(x)$ denotes the eigen mode of core 2. The coupling constants are related to the single-mode propagation constant β and the propagation constants of the supermodes in the dual-core configuration by

$$\beta = \frac{\beta^{(e)} + \beta^{(o)}}{2}, \quad \kappa = \frac{\beta^{(e)} - \beta^{(o)}}{2} \quad (2.23)$$

$$\beta^{(e)} = \beta + \kappa, \quad \beta^{(o)} = \beta - \kappa \quad (2.24)$$

where $\beta^{(e)} = n^{(e)}\omega/c$ and $\beta^{(o)} = n^{(o)}\omega/c$ are the propagation constants for the even and odd mode in the two parallel waveguides. $n^{(e)}$ and $n^{(o)}$ are the corresponding effective refractive indices. The coupling length is

$$L_c = \frac{\pi}{2\kappa} = \frac{\pi}{\beta^{(e)} - \beta^{(o)}}. \quad (2.25)$$

2.2.2 Coupling constant calculated from single-mode fiber

The mode-coupling effect in two cores can be analyzed by the interference phenomena between the even and odd modes, as shown in Figure 2.3. When the center

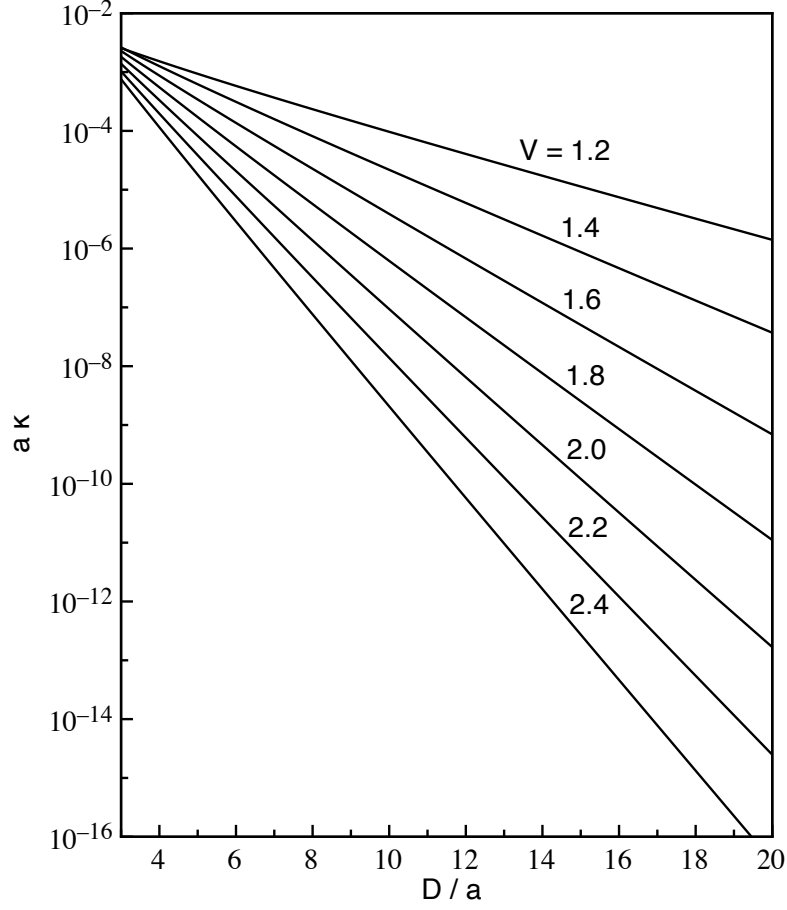


Figure 2.4: Dependencies of the normalized coupling coefficient κa on the relative core center separation D/a for various V -values.

separation D approaches infinity ($D \rightarrow \infty$), the system is reduced to a single cylindrical waveguide. If we use the eigenvalue equation of the LP_{01} mode Equation (2.18) and the formula of the modified Bessel function, then the mode-coupling coefficient of the dual core step-index optical fiber is given by [37]

$$\kappa = \frac{\sqrt{2\Delta}}{a} \frac{u^2}{V^3} \frac{K_0(\frac{wD}{a})}{K_1^2(w)}, \quad (2.26)$$

where a is the radius of each core and D is the distance between two cores, center

to center. This expression will help us calculate the coupling constant κ in dual core optical fiber, since other parameters can be obtained from the single-mode fiber. We will rely on this approximate expression for the coupling coefficient between the fundamental modes in a dual core step-index optical fiber. This expression is valid for larger values of wD/a ($wD/a > 1$). Figure 2.4 shows the dependencies of the normalized coupling coefficient on the relative core center separation D/a for various V -values, similarly compared to the results from [30]. As the value of D becomes larger, as well as two cores getting further and further apart, the coupling constant decreases exponentially.

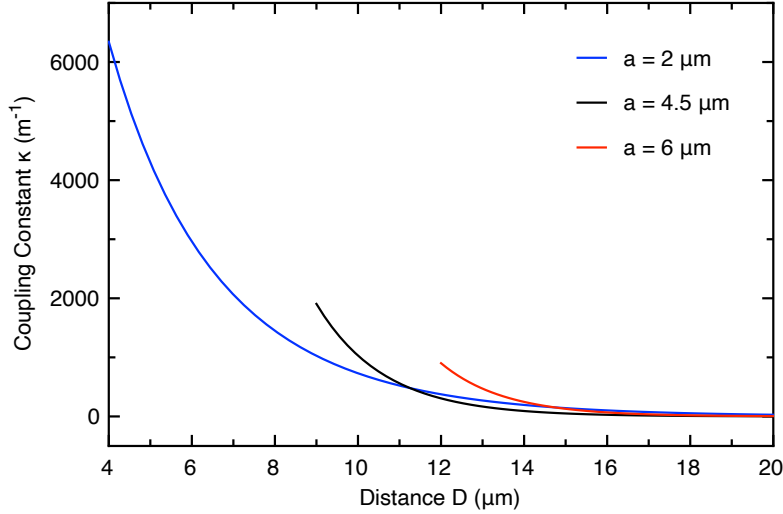


Figure 2.5: Coupling constant κ as a function of distance D at $1.55\mu m$, calculated from Equation (2.26), with different radius. And $\Delta n = 0.01$ for silica fiber.

From Equation (2.26), we calculate coupling constant κ as a function of distance D at $1.55\mu m$, with different radius for a dual core step-index silica fiber. The dispersion data is for 7979 Corning glass and we assume $\Delta n = 0.01$. The coupling becomes weaker and weaker towards zero when increasing D , which is valid for the expression (2.26). As D is smaller, we notice a dramatic increasing of coupling constant, then the expression of κ is not accurate (See Ref. [37], Chapter 18).

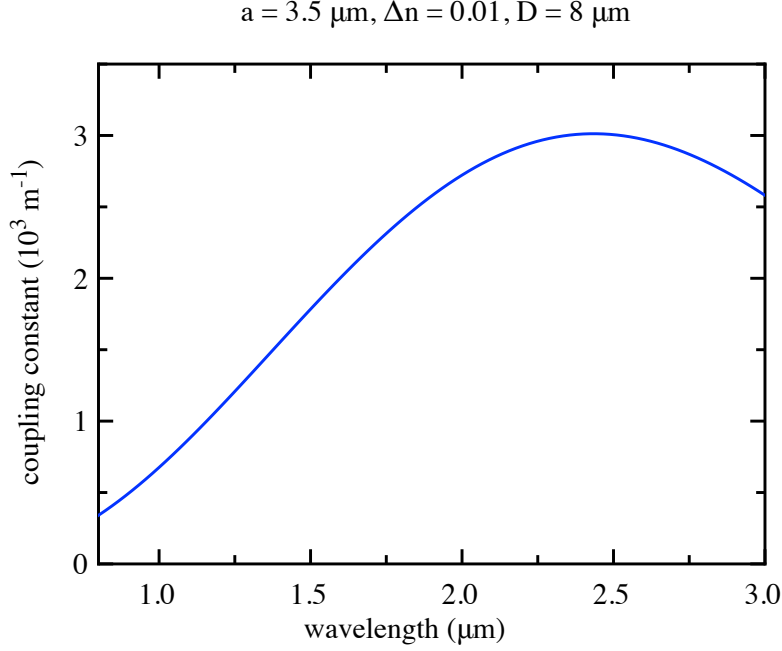


Figure 2.6: The curve of the coupling constant as a function of wavelength for a silica dual-core step-index fiber with a refractive index contrast of $\Delta n = 0.01$ and core radius of $3.5 \mu\text{m}$, obtained from Equations (2.26) and (2.24).

The plot of the coupling constant as a function of wavelength, calculated from Equations (2.26) and (2.24), is shown in Figure 2.6. As the wavelength becomes longer, the value of the coupling constant approaches to its peak, and then starts to decrease. This curve is not correct in the way that the calculation is done via the approximation in Equation (2.26). The results will be compared with the data obtained from COMSOL Multiphysics software [29] in chapter 6.

We can then calculate the propagation constants $\beta^{(e)}$ and $\beta^{(o)}$ of even and mode supermodes from Equations (2.23). In Figure 2.7, a plot of the normalized propagation constants of the even and odd supermodes in a step-index silica fiber with a core radius of $2 \mu\text{m}$, a distance $D = 7 \mu\text{m}$ apart, and $\Delta n = 0.01$, is presented. Compared with Figure 2.2, we can observe the relationship of b with frequency and wavelength mode directly. When the frequency is less than 150 THz (or the wavelength is

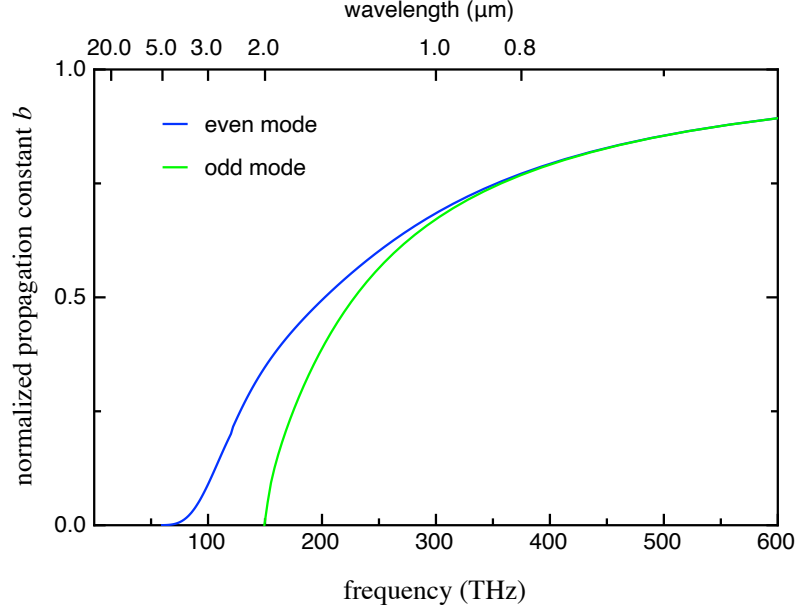


Figure 2.7: Dispersion of the even (blue curve) and odd (green curve) supermodes in a step-index silica fiber with a core radius of $2 \mu\text{m}$, a distance of $D = 7 \mu\text{m}$ apart, and $\Delta n = 0.01$. The curves are obtained from Equations (2.26) and (2.23).

greater than $2 \mu\text{m}$), the odd mode is cut off. The analysis of the disappearance of the odd mode is similar when we discuss the condition of single-mode in Section 2.1. As we see, the only propagation constant that remains is $\beta^{(e)}$ in Equations (2.23) when the odd mode disappears. Therefore, the expression of κ in (2.26) can not be used anymore. The cut off of the odd mode is of huge importance to a dual-core optical fiber, and we must work within appropriate wavelength. Also, The results will be compared with the data obtained from COMSOL Multiphysics software [29] in chapter 6.

Chapter 3

Quasi-degenerate four-wave mixing and frequency down-conversion in a single-core waveguide

This chapter presents nonlinear third-order frequency downconversion in a single-core fiber. It is based on a quasi-degenerate four-wave mixing scheme where energy is transferred from a pump wave at frequency ω_2 to two other waves with frequencies $\omega_1 < \omega_2$ and $\omega_3 > \omega_2$. We focus on the phase-matching possibilities for this process when $\omega_2 - \omega_1$ is significant and allows generation of far-infrared light from near-infrared source. We also study the limitations of phase-matching in a single-core fiber with discussions of the influence of different geometrical parameters. The basics and fundamentals about nonlinear third-order frequency downconversion in a single-core fiber are discussed in Section 3.1. Phase matching conditions and discussion of main results for frequency down-conversion in silica and fluoride fibers are shown in Section 3.2. The discussion in this chapter serves as the preparation for the work on dual-core fiber in chapter 4.

3.1 Frequency down-conversion in a single-mode fiber

3.1.1 Third-order susceptibility and down-conversion mechanism

The third-order nonlinear optical susceptibility that underlies the frequency conversion process we are interested in is $\chi_{ijkl}^{(3)}(-(2\omega_2 - \omega_3), \omega_2, \omega_2, -\omega_3)$. The third-order nonlinear optical polarization that radiates a wave at frequency ω_1 from the interaction of two waves at frequencies $\omega_3 > \omega_2 > \omega_1$ is

$$P_{NL}^{(\omega_1)} = \frac{3}{4}\epsilon_0\chi_{\text{eff}}^{(3)}(-\omega_1, \omega_2, \omega_2, -\omega_3)[E^{(\omega_2)}]^2 E^{(\omega_3)*} \quad (3.1)$$

where the $E^{(\omega_i)}$ are the amplitudes of the interacting optical electric fields, the superscript asterisk stands for complex conjugation, and $\chi_{\text{eff}}^{(3)}$ is the coefficient of the third-order susceptibility tensor that in general depends on the polarization of the interacting waves. In silica glass, $\chi_{\text{eff}}^{(3)} = \chi_{1111}^{(3)} \sim 2 \times 10^{-22} \text{V}^2 \text{m}^{-2}$ for interacting waves polarized parallel to each other [38]. We will use S.I. units throughout.

For the following analysis, we will use the normalized amplitudes

$$A_i = \sqrt{\frac{\epsilon_0 c n_i}{\hbar \omega_i}} E^{(\omega_i)} \quad (3.2)$$

where n_i is the effective refractive index for the wave with frequency ω_i , ϵ_0 is the permittivity of vacuum, c is the speed of light, and $\hbar$ is the Plank constant. The A_i have units of $(\text{m}^{-1} \text{s}^{-1/2})$ and their square represents a photon flux, which will become useful when considering the efficiency of this interaction later.

We also introduce an effective nonlinear optical coefficient

$$\chi = \frac{3}{8}\chi_{\text{eff}}^{(3)} \frac{\hbar}{\epsilon_0 c^2} \sqrt{\frac{\omega_1 \omega_2^2 \omega_3}{n_1 n_2^2 n_3}} S, \quad (3.3)$$

whose units are the product of length and time (m s). Here, the quantity $S = 1$ for plane wave interaction, and represents the overlap integral between the transverse mode-profiles of the interacting waves for propagation in a waveguide. Since

the present work focuses on third-order nonlinear optical interactions of the lowest frequency fundamental modes in weakly guided step-index fibers, the third-order interaction happens over the whole mode profile both in the core and in the cladding region, and we can assume that $S = 1$.

3.1.2 Coupled wave equations

In optical fiber, we consider the propagation direction of all beams is the same and use propagation constant β_i to represent each wave. By inserting the three waves propagating in the fiber and the induced nonlinear optical polarization (3.1) into the nonlinear optical (NLO) wave equation (1.1) allows to obtain the following set of coupled-wave equations for the amplitudes (3.2). Compared with Equations (1.4-1.7), two equations of them get merged considering the process of quasi-degenerate four-wave mixing $2\omega_2 - \omega_3 \rightarrow \omega_1$, and then a degeneracy factor of 2 comes in:

$$\begin{aligned} \frac{\partial}{\partial z} A_1 &= i \chi [A_1(2A_3A_3^* + 2A_2A_2^* + A_1A_1^*) \\ &\quad + [A_2]^2 A_3^* e^{i\Delta\beta z}], \end{aligned} \quad (3.4)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_2 &= i \chi [A_2(2A_3A_3^* + A_2A_2^* + 2A_1A_1^*) \\ &\quad + 2A_1A_2^* A_3 e^{-i\Delta\beta z}], \end{aligned} \quad (3.5)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_3 &= i \chi [A_3(A_3A_3^* + 2A_2A_2^* + 2A_1A_1^*) \\ &\quad + [A_2]^2 A_1^* e^{i\Delta\beta z}], \end{aligned} \quad (3.6)$$

where $\Delta\beta = 2\beta_2 - \beta_1 - \beta_3$. This wavevector mismatch is positive when $\omega_3/\omega_1 > (n_2 - n_1)/(n_3 - n_2)$, but can be negative for a large difference between the frequencies. For normal refractive index dispersion when $\omega_3/\omega_1 > (n_2 - n_1)/(n_3 - n_2)$, there will always exist a choice of frequency where $\Delta\beta > 0$ and phase-matching can be achieved. In each of the above three equations, the terms related to self- and cross-phase modulation are in the first line, and the terms related to the third-order interaction between the three waves are in the second line.

3.1.3 General solutions for maximum conversion efficiency

When the phase-modulation terms are neglected, one finds that the phase-matching condition $\Delta\beta = 0$ will lead to a complete transfer of the energy of the pump wave at frequency ω_2 to the “sidebands” at frequencies ω_3 and ω_1 . But at higher intensities the phase-modulation terms can lead to unwelcome and significant dephasing of the waves with propagation, even when $\Delta\beta = 0$. It is therefore important to consider the full solution of these equations in the presence of phase modulation. Here we follow the approach introduced by Cappellini *et al.* [39].

We start by pointing out in this case the following quantities must be independent from z :

$$A_{tot}^2 = A_1(z)^2 + A_2(z)^2 + A_3(z)^2 \quad (3.7)$$

$$\Delta A^2 = A_3(z)^2 - A_1(z)^2 \quad (3.8)$$

The first condition reflects energy conservation. The second one is a characteristic for the nonlinear optical interaction we are studying, a process in which two photons are taken from wave 2 while one photon is added to each of the two other waves.

To examine the effect of pump depletion and phase mismatch, we write the complex amplitudes $A_j = A_j e^{i\phi_j}$ in terms of their magnitudes A_j and phases ϕ_j . Substituting into Equations (3.4 - 3.6) and equating the real and imaginary parts of each equation leads to the following set of nonlinear equations in real variables:

$$\frac{\partial A_3}{\partial z} = -\chi A_2^2 A_1 \sin[\Delta\phi(z)] \quad (3.9)$$

$$\frac{\partial A_2}{\partial z} = 2\chi A_3 A_2 A_1 \sin[\Delta\phi(z)] \quad (3.10)$$

$$\frac{\partial A_1}{\partial z} = -\chi A_2^2 A_3 \sin[\Delta\phi(z)] \quad (3.11)$$

$$\begin{aligned} \frac{\partial \Delta\phi}{\partial z} = & \Delta\beta - \chi(2A_2^2 - A_3^2 - A_1^2) \\ & + \chi[4A_1 A_3 - A_2^2(\frac{A_1}{A_3} + \frac{A_3}{A_1})] \cos[\Delta\phi(z)] \end{aligned} \quad (3.12)$$

where $\Delta\phi(z) = \Delta\beta z + 2\phi_2(z) - \phi_1(z) - \phi_3(z)$.

It is also convenient to deal with normalized dimensionless amplitude variables. We introduce the normalized pump power $\eta(z) = [A_2^2(z)]/A_{tot}^2$, the normalized sideband amplitudes $a_{1,3}(z) = A_{1,3}(z)/A_{tot}$, and the normalized mismatch $\tau = \Delta\beta/(\chi A_{tot}^2)$. The two invariants that we introduced earlier, Equation (3.7) and (3.8) then become

$$\alpha = a_3^2 - a_1^2, \quad (3.13)$$

$$1 = \eta(z) + a_1^2 + a_3^2. \quad (3.14)$$

Now we can rewrite the real equations from (3.9 - 3.12):

$$\frac{\partial a_3}{\partial z} = -\chi A_{tot}^2 \eta(z) a_1 \sin[\Delta\phi(z)] \quad (3.15)$$

$$\frac{\partial a_1}{\partial z} = -\chi A_{tot}^2 \eta(z) a_3 \sin[\Delta\phi(z)] \quad (3.16)$$

$$\frac{\partial \eta}{\partial z} = 4\chi A_{tot}^2 \eta(z) a_3 a_1 \sin[\Delta\phi(z)] \quad (3.17)$$

$$\begin{aligned} \frac{\partial \Delta\phi}{\partial z} = & \Delta\beta - \chi A_{tot}^2 [2\eta(z) - a_3^2 - a_1^2] \\ & + \chi A_{tot}^2 [4a_1 a_3 - \eta(z) \left(\frac{a_1}{a_3} + \frac{a_3}{a_1}\right)] \cos[\Delta\phi(z)] \end{aligned} \quad (3.18)$$

Then, Cappellini *et al.* identified an additional z -independent quantity (Eq. 11 in Ref. [39]):

$$H(z) = -2\eta(z)[(1 - \eta(z))^2 - \alpha^2]^{1/2} \cos[\Delta\phi(z)] - (\tau + 1)\eta(z) + \frac{3}{2}\eta^2(z). \quad (3.19)$$

Since the mismatch τ is given from the dispersion of the coupled waves and α is also an invariant, $H(z)$ becomes a system that involves the variables $(\eta, \Delta\phi)$. With the normalized propagation length $\xi = \chi^{(3)} A_{tot}^2 z$, it is convenient to express a one-degree-of-freedom system:

$$\frac{\partial \eta}{\partial \xi} = \frac{\partial H}{\partial \Delta\phi} = 2\eta[(1 - \eta)^2 - \alpha^2]^{1/2} \sin \Delta\phi \quad (3.20)$$

$$\frac{\partial \Delta\phi}{\partial \xi} = -\frac{\partial H}{\partial \eta} = (\tau + 1) - 3\eta + 2\frac{1 + 2\eta^2 - 3\eta - \alpha^2}{[(1 - \eta)^2 - \alpha^2]^{1/2}} \cos \Delta\phi \quad (3.21)$$

Now it is a system of two self-consistent coupled equations for the variables $(\eta, \Delta\phi)$, which turns out to be conjugate through the one-dimensional conservative $H = H(\eta, \Delta\phi)$.

The advantage of $H(\eta, \Delta\phi)$ is that it contains the properties of the solutions, which are related to the topology of the phase space. This can be done simply by drawing the level curves (defined by $H = \text{constant}$) in the phase space $(\eta, \Delta\phi)$, as shown in Figure 3.1. Each closed trajectory line is the locus of points that represent the values of η and $\Delta\phi$ along the waveguide.

We find that the phase difference $\Delta\phi$ changes periodically, in response to the process in which two photons are created while one photon of wave 2 is depleted, and two photons are taken to generate one photon of wave 2. During the propagation, $\frac{\partial\Delta\phi}{\partial\xi}$ is related to both η and $\Delta\phi$. As the value of $\Delta\phi$ flips from positive to negative, $\frac{\partial\eta}{\partial\xi}$ is also reversed for the reason that it is proportional to $\sin\Delta\phi$. Simultaneously, the process of three-wave mixing is reversed to two photons are taken to generate one photon of wave 2, and η starts to increase.

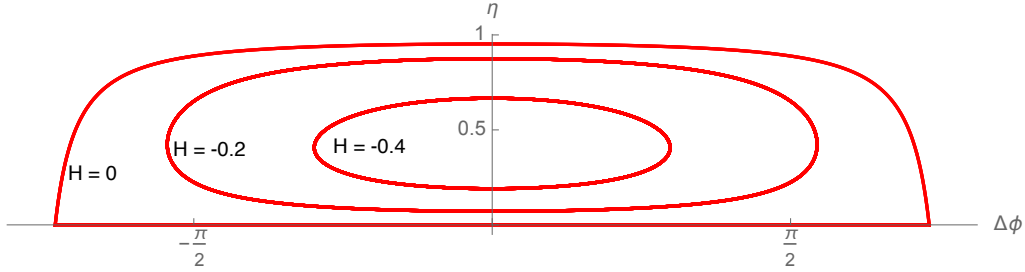


Figure 3.1: Phase space portrait with different values of $H = 0, -0.2, -0.4$.

If the level curves are under the condition $H \neq 0$ in the phase space $(\eta, \Delta\phi)$, η cannot be depleted to zero and the energy flow is reversed. Therefore, the lowest value of η can be calculated with Equation (3.19) and $\Delta\phi = 0$. The fact that remnant photons of wave 2 are left results in an incomplete process of photon conversion, which means the maximum photon conversion efficiency cannot be reached.

However, the pump power η can be totally depleted toward zero in the case of $H = 0$. The photon flux of the pump wave at ω_2 decreases from its initial value $\eta(z = 0)$ to a final value as close as zero, which implies $H = 0$. Therefore, $H = 0$

implies to the condition that maximize conversion efficiency can be achieved during the wave propagation. In other words, we need to fulfill $H(z = 0) = 0$ initially, which is

$$-2\eta(z = 0)[(1 - \eta(z = 0))^2 - \alpha^2]^{1/2} \cos[\Delta\phi(z = 0)] - (\tau + 1)\eta(z = 0) + \frac{3}{2}\eta^2(z = 0) = 0. \quad (3.22)$$

3.1.4 Strategy to maximize conversion efficiency

We study the the generated wave at frequency ω_1 always starts at zero amplitude, which implies that $A_1(0) = 0$. Equation (3.4) then implies that $\phi_1(0) = \pi/2$ and therefore $\Delta\phi(0) = 2\phi_2(0) - \phi_1(0) - \phi_3(0) = -\pi/2$. This value of the initial phase difference $\Delta\phi(0)$ causes the energy to flow from the pump wave to the two other waves, and in the absence of phase modulation this situation is maintained at all distances when $\Delta\beta = 0$.

On the other hand, the presence of the self- and cross-phase modulation terms in Equations (3.4-3.6) will always cause $\Delta\phi(z)$ to drift away from this initial value, even when $\Delta\beta = 0$, and phase-matching can never be maintained. We plot the spatial evolution of the amplitudes of each waves under $\Delta\beta = 0$ in Figure 3.2. The oscillations are getting intensified as the waves propagate. However, the pump wave won't be depleted too much under this condition, which leaves the photon conversion efficiency less than 20%. On account of the phase modulations, even though the phase-matching condition can be achieved, the photon conversion efficiency is limited to be low.

But it is still possible to obtain a large conversion efficiency for a specific value of $\Delta\beta \neq 0$. The condition for maximum conversion efficiency corresponds to the situation when the pump wave has been completely depleted at some distance z . $A_2(z) = 0$ then implies that under conditions of maximum conversion efficiency the invariant (3.19) vanishes for all values of z , and in particular also for $z = 0$, where $\cos\Delta\phi(0) = 0$. From this one finds that maximum conversion efficiency can be obtained when $\Delta\beta = \chi [(3/2)A_2^2(0) - A_{tot}^2]$, or

$$\Delta\beta = G = \chi \left[\frac{1}{2}A_2^2(0) - A_3^2(0) - A_1^2(0) \right], \quad (3.23)$$

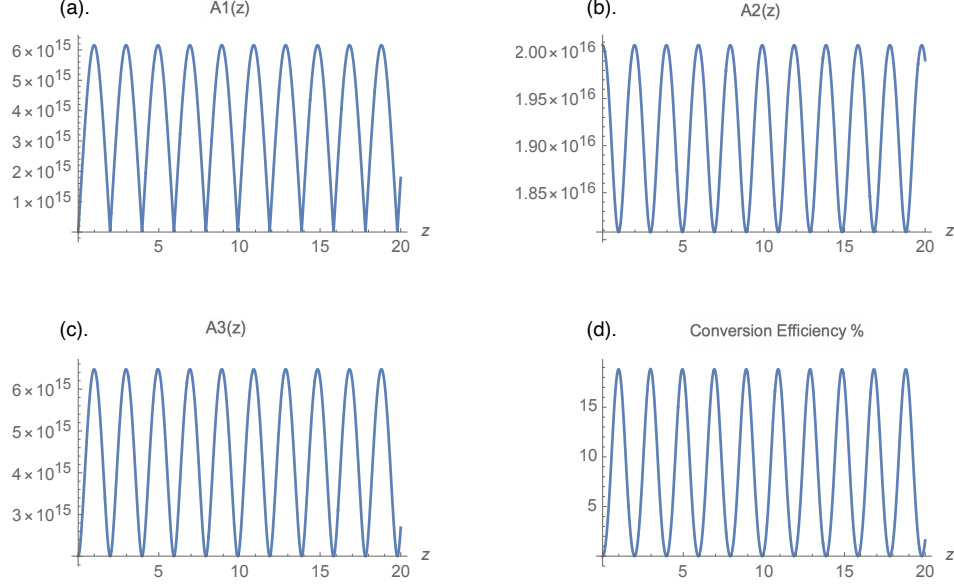


Figure 3.2: Spatial evolution of the amplitudes and periodic behavior of the interacting waves for third-order frequency down-conversion, $\omega_1 = 2\omega_2 - \omega_3$. Panels (a), (b), and (c) give the amplitudes of three waves at frequency ω_1 , ω_2 , and ω_3 , respectively. Panel (d) presents the dependence of photon-conversion efficiency on propagation length. The parameters are, $\chi = 1.069 \times 10^{-32} \text{ m}$, $A_2(0) = 2.007 \times 10^{16} \text{ m}^{-1}$, $\Delta\phi(0) = -\frac{\pi}{2}$, and $\Delta\beta = 0$.

where we have defined an intensity-dependent phase modulation correction term G . The above condition corresponds to the conventional phase matching condition for low intensities, but it is otherwise a condition for maximum conversion efficiency that depends only on the initial intensity of the pump wave at ω_2 (and any seed wave, at ω_3).

We note that inserting Equations (3.2) and (3.3) into Equation (3.23), together with the intensity $I = \epsilon_0 n c E^2 / 2$ and the nonlinear refractive index for self-phase modulation, $n_2^{NL} = 3\chi_{\text{eff}}^{(3)} / (4\epsilon_0 c n^2)$, leads, in the limit when all frequencies are similar

and the pump intensity is dominant, to

$$G = \gamma P \quad (3.24)$$

where $\gamma = 2\pi n_2^{NL}/(\lambda A_{\text{eff}})$ is the nonlinear coefficient of a fiber, with A_{eff} the effective mode area, and P is the average pump power in the guided mode. This result essentially matches earlier approximations in the limit of almost degenerate frequencies [40–45], confirming the validity of Equation (3.23).

For instance, a 10 ns duration laser pulse with an energy of 1 mJ and a beam waist of 4.2 μm has a peak intensity of 3.35 GW cm^{-2} , which is less than the damage threshold of fused silica [46]. At such an intensity for the pump wave and no seed wave, the factor G in Equation (3.23) reaches a value of 2.109 m^{-1} (using $\chi_{1111}^{(3)} = 2 \times 10^{-22} \text{ m}^2\text{V}^{-2}$ for fused silica). As a comparison, spatial evolution of the amplitudes of three interacting waves are shown in Figure 3.3. The photon-conversion efficiency reaches essentially 100% when $\Delta\beta = G = 2.109 \text{ m}^{-1}$.

As propagation continues, the pump wave at ω_2 is recreated again in full, depleting two sideband waves in reversal. We notice the spatial evolution of both $\eta(z)$ and $\Delta\phi(z)$ are also periodic, in response to the process in which two photons are taken from wave 2, and two photons are created while one photon of wave 2 is depleted. A small remnant of increase of $\eta(z)$ can help flip the direction of energy flow in this process, recreating the pump wave with almost 100%, then the whole cycle repeats. The amplitudes of three interacting waves evolve in the same way as $\eta(z)$ and $\Delta\phi(z)$ we discussed earlier. A_2 is depleted to zero while A_1 and A_3 increase to their maximum and 100% photon-conversion efficiency is reached. Similarly, a small remnant of increase of A_1 and A_3 help flip the direction of energy flow in this process, periodically.

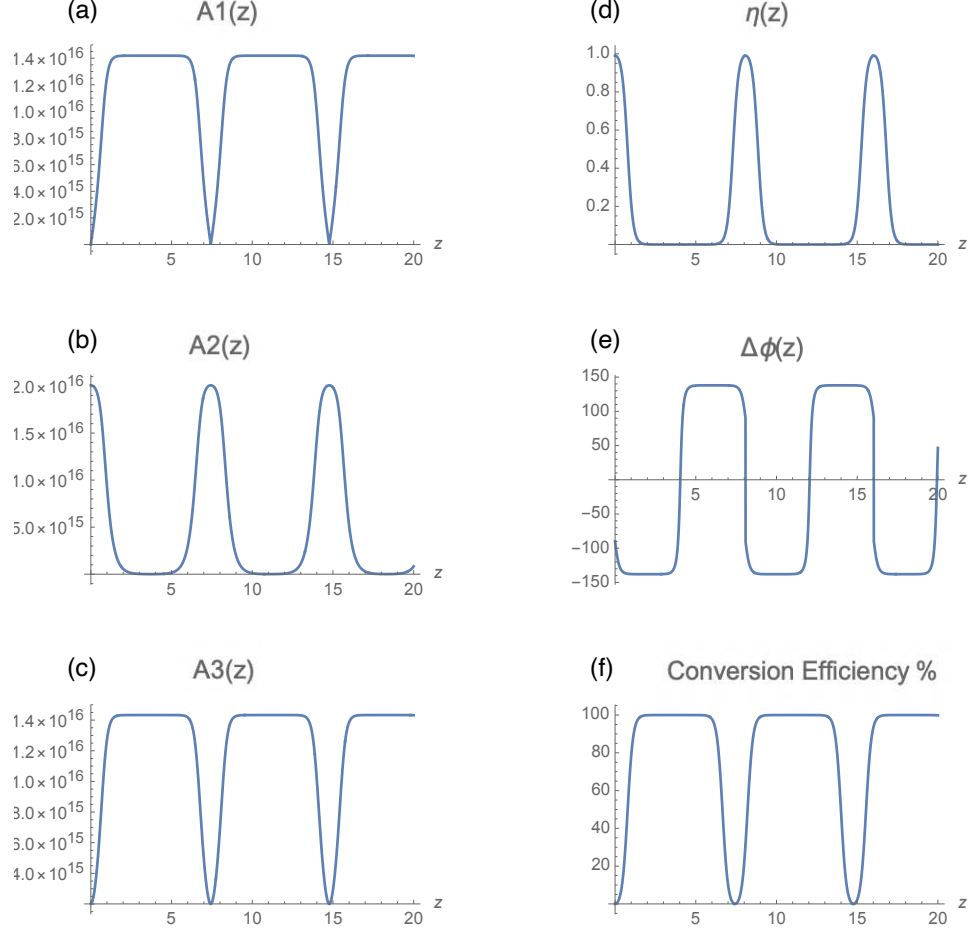


Figure 3.3: Spatial evolution of the amplitudes and periodic behavior of the interacting waves for third-order frequency down-conversion, $\omega_1 = 2\omega_2 - \omega_3$. Panels (a), (b), and (c) give the amplitudes of three waves at frequency ω_1 , ω_2 , and ω_3 , respectively. Panels (d) and (e) show the propagation distance dependence of η and $\Delta\phi$. Panel (f) presents the dependence of photon-conversion efficiency on propagation length. The parameters are, $\chi = 1.069 \times 10^{-32} \text{ m}$, $A_2(0) = 2.007 \times 10^{16} \text{ m}^{-1}$, $\Delta\phi(0) = -\frac{\pi}{2}$, and $\Delta\beta = 2.109 \text{ m}^{-1}$.

3.2 Phase-matching conditions for frequency down-conversion in silica and fluoride fibers

In this section, we consider both silica and fluoride fibers as examples. Their dispersion data was obtained from the standard Sellmeier expression $n^2 - 1 =$

Table 3.1: Sellmeier parameters for SiO₂, ZrF₄, and InF₃ glasses that we used in this work. The SiO₂ data is for 8655 and 7979 Corning glass. The fluoride data is for the cladding of Thorlabs single mode fluoride patch cables, published on their website.

	SiO ₂	ZrF ₄	InF ₃
a_1	2.623483282E-2	0.705674	0.68462594
a_2	7.306029048E-1	0.515736	0.4952746
a_3	3.475321572E-1	2.204519	1.4841315
a_4	9.216052441E-1	0.0	0.0
b_1	-5.783959035E-3	0.087503	0.0680833
b_2	5.600103210E-3	0.087505	0.11054856
b_3	1.389808930E-2	23.80739	24.4391868
b_4	1.006578079E+2	0.0	0.0

$\sum_{i=1}^4 a_i/(1 - b_i^2/\lambda^2)$ with the wavelength λ in μm and the parameters a_i and b_i given in Table 3.1.

3.2.1 Bulk dispersion of silica and fluoride fibers

Since the coupled wave equations (3.4-3.6) are valid both for a single mode fiber as well as for plane-wave propagation in the bulk material, it is useful to first consider the behavior of the phase-mismatch factor $\Delta\beta$ as determined by the refractive index dispersion in typical glasses. In contrast to simple second-order nonlinear optical interactions, the fact that the refractive index monotonically decreases does not forbid phase matching for the $2\omega_2 - \omega_3 \rightarrow \omega_1$ process. In fact, one finds that in bulk silica glass this process can generate wavelengths between 2.5 and 2.0 μm when using pump wavelengths between ~ 1.16 and ~ 1.22 μm . And in bulk InF₃ (ZrF₄) glass the same process can generate wavelengths between 5 and 2.5 μm when using pump wavelengths between ~ 1.5 and ~ 1.8 (~ 1.3 and ~ 1.55) μm , respectively [47, 48]. When using the same glass in optical fibers, waveguide dispersion has a slight effect on $\Delta\beta(\lambda_1)$ but the main characteristics of the phase-matching condition remain similar. The possibility of obtaining $\Delta\beta = 0$ for three-wave mixing in single-mode fibers was recognized early [28], but we are not aware of any experimental

demonstration of this effect.

A plot of $\Delta\beta(\lambda_1)$ for different glasses and pump wavelengths λ_2 is shown in Figure 3.4(a). The phase-matching conditions, given by the wavelengths at which the curves cross the $\Delta\beta = 0$ line, change when changing pump wavelength or, in fibers, also when changing refractive index contrast or core radius. To get a better overall view of the phase-matching possibilities, we prefer to plot the phase-matching wavelengths for different geometrical parameters in the plots that give the generated wavelengths as a function of the pump wavelength. This is done for the same three glasses in Figure 3.4(b). In order not to overload these examples, we only included the curves for a single-mode ZrF_4 fiber.

In these plots, each phase-matching curve reaches a cusp at a maximum pump wavelength above which no phase matching can be obtained. The intersection between the “degeneracy line” for the case of three equal frequencies and the cusp of the general phase-matching curves corresponds to the zero group velocity dispersion wavelength [49]. This means that the group velocity dispersion (GVD), which is described by the term $d^2k/d\omega^2$, vanishes when phase matching is fulfilled at the cusp of the curves. For wavelengths below this maximum value, the two branches of the curves give the wavelength of the generated waves, which move further and further apart as the pump wavelength decreases.

Figure 3.5 shows how the phase-matching curves change into “maximum conversion efficiency” curves when taking into account the effect of self- and cross-phase modulation. This plot gives the wavelength choices for maximum conversion efficiency, as obtained from Equation (3.23), again for a silica fiber with a refractive index contrast $\Delta n = 0.01$ and a core radius $a = 4.5 \mu\text{m}$.

For low intensities ($G \rightarrow 0$) all curves in Figure 3.5 tend to the standard phase-matching curve, with a zero dispersion wavelength and maximum pump wavelength of $\lambda_2^{max} = 1.248 \mu\text{m}$. For larger values of G , the generated wavelength λ_1 increases, an effect that is stronger near the degeneracy point at $\lambda_2^{max} = 1.248 \mu\text{m}$. While this also leads to the possibility of achieving efficient interactions for pump wavelengths above λ_2^{max} , the generated wavelengths are limited to a small interval close to the

pump wavelength. Similar effects will also happen when we will next consider third-order frequency down-conversion in dual-core fibers, but the modifications when compared to the zero-intensity phase-matching conditions are relatively small when the distance between the wavelengths is larger, which is the region we are interested in in this work.

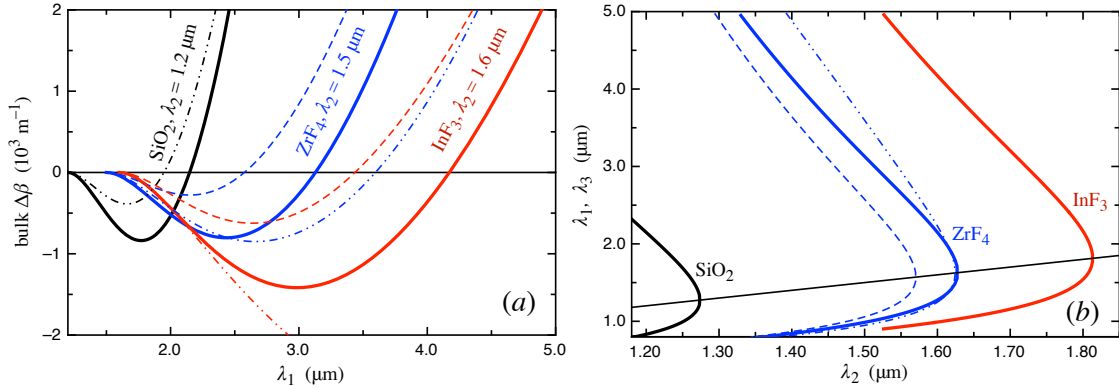


Figure 3.4: Frequency conversion for both plane-wave interaction and single-mode fibers. Top: Phase mismatch factor $\Delta\beta$ as a function of the generated wavelength λ_1 for different pump wavelength and glass types. The solid curves with different colors are for plane waves, the dashed (dashed-dotted) curves of the same color are for single-mode step-index fibers with a radius of $9\text{ }\mu\text{m}$ ($4.5\text{ }\mu\text{m}$). Bottom: Phase-matching curves representing the generated wavelengths as a function of the pump wavelength for the three glasses. Thick solid curves are for plane-wave interaction. The solid line is for the degenerate case with all frequencies equal to each other. The ZrF_4 example includes curves for a $4.5\text{ }\mu\text{m}$ core fiber (dash-dotted line) and a $9\text{ }\mu\text{m}$ core fiber (dashed line).

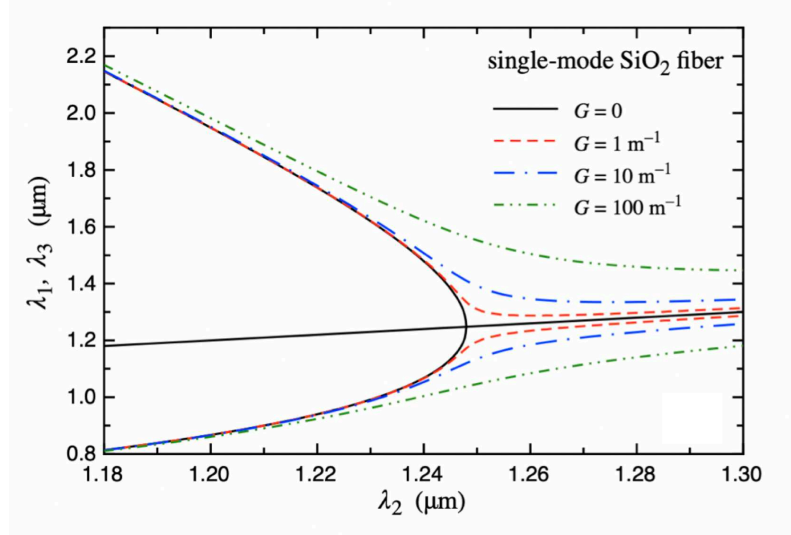


Figure 3.5: Wavelengths for maximum single-mode frequency downconversion efficiency in silica glass when neglecting self- and cross-phase modulation ($G=0$), and for various initial pump intensities expressed in terms of the G factor introduced in Equation (3.23). The solid line is for the degenerate case.

3.2.2 Dependence on core radius and index contrast

The geometrical parameters involved in order to achieve the phase-matching condition $\Delta\beta = 0$ are the radius of the core and the refractive index contrast. In this section, we consider examples for a step-index silica fiber with different geometrical parameters, focusing on how the phase-matching condition can be achieved by different pump wavelengths to produce a generated wave above $2 \mu m$.

First of all, we plot phase-matching curves for different refractive index contrast Δn in Figure 3.6 that give the generated wavelength as a function of the pump wavelength for different refractive index contrast Δn . As we increase the refractive index contrast Δn , we can see the zero group velocity dispersion wavelength λ_2^{max} becomes smaller, which limits the choice of the pump wavelength to achieve phase-matching. When the core radius is $4 \mu m$, the pump wavelength is required to be less than $1.3 \mu m$. For a larger radius, the generated wavelength can be maintained above $2 \mu m$ by going to smaller pump wavelengths λ_2 .

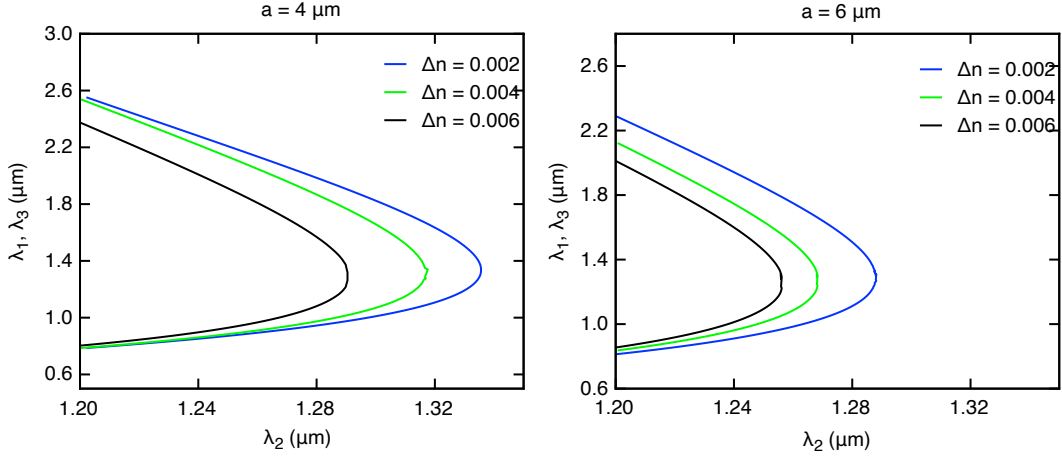


Figure 3.6: Phase-matching curves representing the generated wavelength as a function of the pump wavelength for different refractive index contrast Δn . Left: For a core radius of $4 \mu m$. Right: For a core radius of $6 \mu m$.

Figure 3.7 shows how the phase-matching condition varies with core radius and pump wavelength for different choice of generated wavelengths. For $\Delta n = 0.002$, the pump wavelengths are limited to $1.3 \mu m$ in order to produce a generated wave above $2 \mu m$. In this situation, two values of the core radius can be chosen with the same pump wavelength. As Δn becomes larger, the choice of the pump wavelengths are extended further toward $1.5 \mu m$ whereas reducing the core radius. In order to create a longer generated wave, the pump wavelength moves to smaller value while also reducing the core radius. If the core radius is limited below $3 \mu m$, then it is not suitable anymore for the fiber application.

Additionally, to investigate the relationship between the core radius and the refractive index contrast for the phase-matching condition that leads to the generated wavelength above $2 \mu m$, Figure 3.8 is presented. By maintaining the refractive index contrast, we must lower the value of core radius to generate longer wavelength. When the pump wavelength is at $1.2 \mu m$, the core radius would be extended into a wide range from 3 to $8 \mu m$ with appropriate choice of index contrast. As the pump wavelength increases to $1.35 \mu m$, although the refractive index contrast also needs to be larger, the core radius is already less than $3 \mu m$.

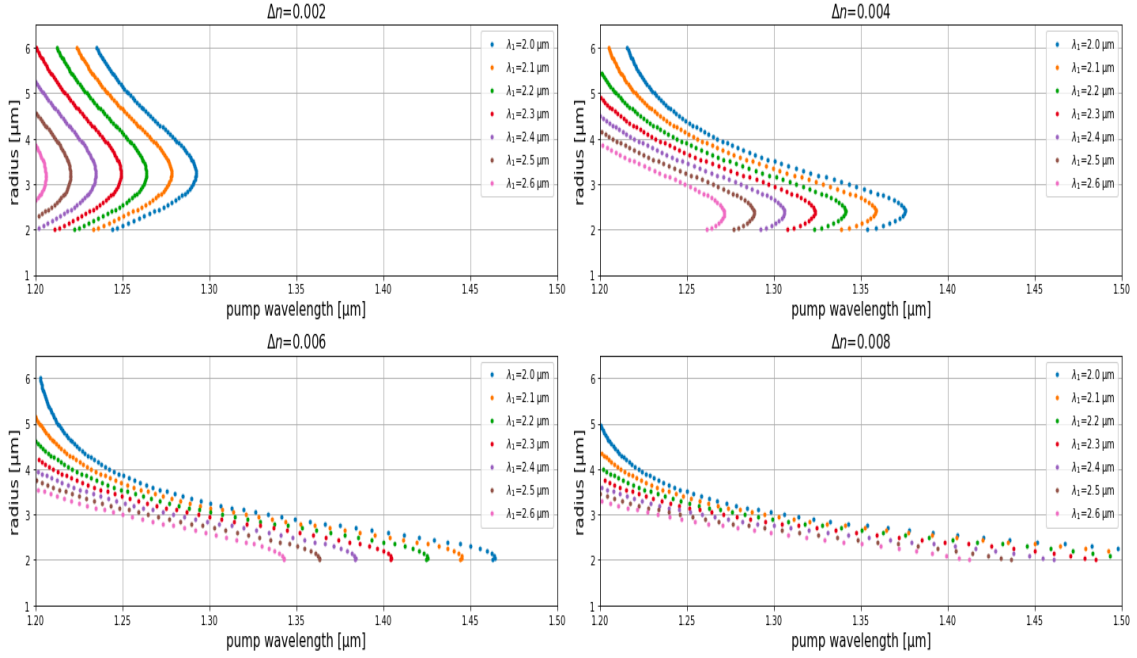


Figure 3.7: Dependence on radius and pump wavelength for different choice of generated wavelengths under the phase-matching condition in a single-core step-index silica fiber with different refractive index contrast Δn .

In summary, we discussed how geometrical parameters influence the choice of the pump wavelength for the phase-matching condition and generate a wave above 2 μm . The most important issue is the fact that we can achieve phase-matching in single-mode silica fibers is very much connected to the fact that we can achieve phase-matching in bulk silica. The phase-matching condition is enabled due to how the dispersion of bulk silica is. And it is not really a property of the optical fiber. The choice of the pump wave is also limited and it really only works for this case because of bulk dispersion. This is not a general method that allows to tune the generated wavelength.

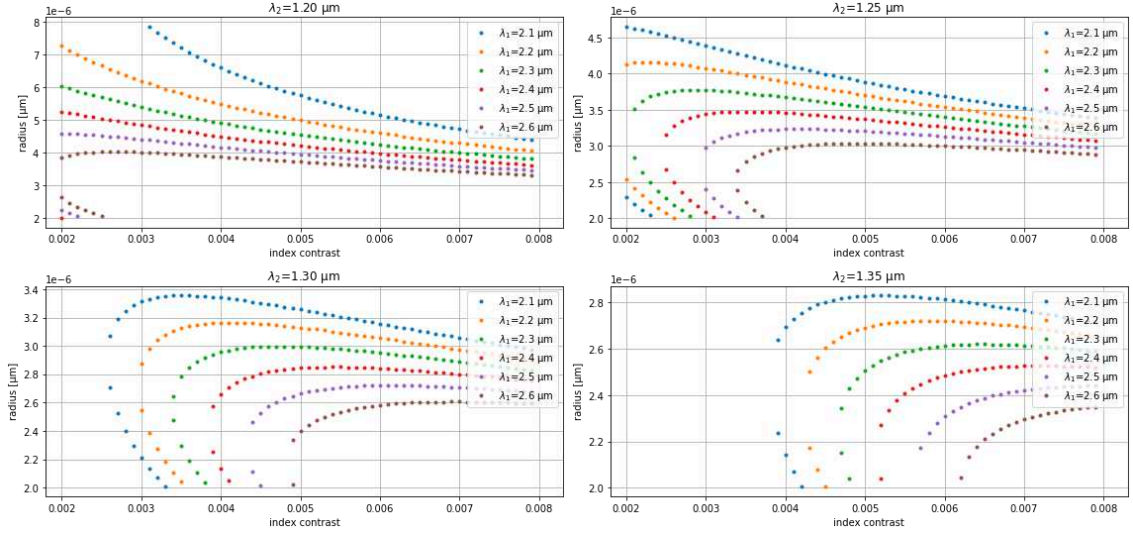


Figure 3.8: Dependence on radius and refractive index contrast Δn for different choice of generated wavelengths under the phase-matching condition in a single-core step-index silica fiber with different pump wavelength.

3.2.3 The effect of phase modulation on conversion efficiency

Similarly as we plot in Figure 3.3, to further illustrate the effect of phase-modulation on the conversion efficiency, Figure 3.9 shows a numerical solution of the coupled wave equations (3.4-3.6) for intensities such that $G = 2.15 \text{ m}^{-1}$ and for two choices of wavelengths: one that satisfies $\Delta\beta = 0$ and one that satisfies $\Delta\beta = G$. $\Delta\beta = 0$ only guarantee 100% conversion efficiency when neglecting the effects of phase modulation (dashed curves), but delivers only little frequency conversion otherwise (left graph). However, 100% conversion efficiency is recovered when the wavelengths of the interacting waves are tuned in such a way that $\Delta\beta = G$ (right graph).

The initial conditions chosen for Figure 3.9 are $A_1(0) = 0$ and $A_2(0) = 100A_3(0)$. This effectively approximates, using the coupled-wave equations (3.4-3.6), a situation where the fiber is only injected with the pump wave at frequency ω_2 , with the other two waves arising from noise. In such a situation, the distance-dependence of the amplitudes when 100% conversion is reached deviates from the familiar linear

growth of $A_1(z)$ that then saturates towards maximum conversion. In addition, it is worth mentioning that in the case when the fiber is only pumped by a single wave, the system will spontaneously choose the generated wavelengths that fulfill $\Delta\beta = G$, which implies a slight increase of the generated wavelength λ_1 as the intensity of the pump wave increases.

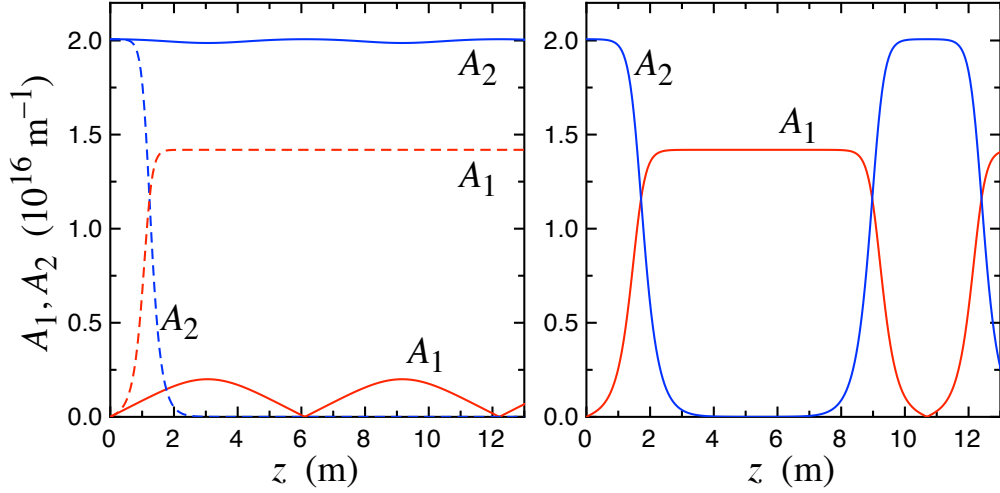


Figure 3.9: Solution of the coupled wave equations in a single mode silica fiber ($\Delta n = 0.01$, $a = 4.5 \mu\text{m}$), similarly as we plot in Figure 3.3. Left: $\Delta\beta = 0$ without taking into account phase modulation. Center: $\Delta\beta = 0$. Right: $\Delta\beta = G$. For this example, $A_2(0) = 2 \times 10^{16} \text{ m}^{-1}$, $G = 2.15 \text{ m}^{-1}$, and $\chi = 1.07 \times 10^{-32} \text{ m}$ (as obtained using (3.3) and the third-order susceptibility of fused silica of $2 \times 10^{-22} \text{ m}^2\text{V}^{-2}$). The pump wavelength is kept at $\lambda_2 = 1.2 \mu\text{m}$. The generated wavelength corresponding to the $\Delta\beta = 0$ and $\Delta\beta = G$ conditions is $\lambda_1 = 1.9480 \mu\text{m}$ and $\lambda_1 = 1.9493 \mu\text{m}$, respectively.

3.3 Conclusions

In this chapter, we show the mechanism of third-order optical nonlinear susceptibility and frequency downconversion in a single-core fiber. And we derive the general solutions for maximum conversion efficiency from nonlinear coupled wave equations. We define an intensity correction factor G , which is important to achieve a condition

for maximum conversion efficiency. Theoretical analysis of how we make strategies to maximize conversion efficiency are presented in details.

As examples, we discuss the phase-matching conditions for frequency downconversion in silica and fluoride fibers. We find that the dispersion of silica will enable phase-matching for third-order four-wave mixing in a single-core step-index fiber. We can achieve phase-matching in single core silica fibers is similar to the fact that we can achieve phase-matching in bulk silica. We also study how the phase-matching condition varies with core radius and pump wavelength for different choice of generated wavelengths. The choice of pump wave and core radius are limited and they only work for a step-index silica fiber because of bulk dispersion. But this is not a general method that allows to tune the generated wavelength.

The value of G is determined by initial values of the amplitudes of three interacting waves, and it will help achieve maximum conversion efficiency during propagation. We present a specific example in a single-mode silica fiber with $a = 4.5 \mu m$, and $\Delta n = 0.01$ to see the evolution of amplitudes of the interacting waves during propagation. The intensity-dependent condition $\Delta\beta = G$ can maximize conversion efficiency when phase modulation is taken into account in the coupled wave equations.

The discussion in this chapter serves as the preparation for the work on dual-core fiber in chapter 4. As we first study a simple system in single-mode fiber, then we use the same concepts to investigate frequency downconversion in a much complicated dual-core fiber. The dual-core system can help overcome the limits that we discovered in a single-mode fiber. And the details will be shown in chapter 4.

Chapter 4

Frequency down-conversion in dual-core fibers

In dual-core optical glass fibers, the coupling between the cores can compensate for the natural bulk dispersion of the material and lead to flexible phase-matching opportunities for third-order nonlinear optical interaction. Figure 4.1 shows that the interacting waves travel along parallel waveguides that allow the light to couple from one to the other [1]. The coupling lengths between the optical modes are responsible for an effective coupling-length phase-matching (CLPM) process [1]. The coupling constants between the waveguides play the role of "coupling wave vectors" that can compensate any mismatch between the sum of the wave vectors of the fundamental waves and that of the generated wave. In this work, we focus on three-wave mixing to generate a longer wavelength via the $2\omega_2 - \omega_3 \rightarrow \omega_1$ process.

For particular materials, like silica and fluoride, the bulk dispersion already allows for phase-matching under the frequency downconversion nonlinear optical interaction. By modifying the radius of the fiber core or the refractive index contrast, such phase-matching conditions for a specific set of wavelengths to single-core fibers can be achieved, as we discussed in chapter 3. However, this method is still not flexible enough, particularly limiting the choice of the pump wavelength. We will present that coupling-length phase-matching (CLPM) is a more flexible way

to achieve the required phase-matching conditions in this chapter. Section 3.1 introduces the concept of CLPM without accounting for phase modulation terms. The theoretical results of CLPM conditions with different relevant parameters will be compared with single-mode fiber. Section 3.2 discusses the influence of phase modulation terms and the modified CLPM rules for optimum conversion efficiency.

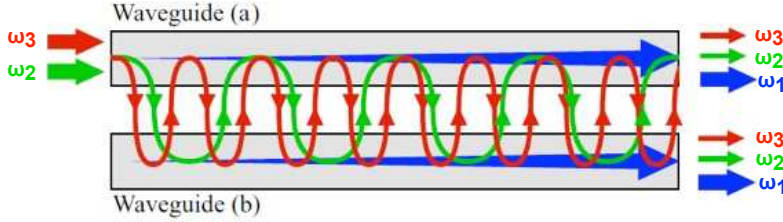


Figure 4.1: Schematic diagram of coupling in two identical waveguides a and b [1]. The pump beams (at frequency ω_2 and ω_3) are injected in waveguide a . The evanescent coupling between the two waveguides causes the power of the pump waves to oscillate between the two waveguides. The generated wave ω_1 is created by nonlinear optical frequency downconversion ($\omega_1 = 2\omega_2 - \omega_3$).

4.1 The concept of coupling-length phase-matching

We describe the optical modes with complex amplitudes $E^{(a,\omega_i)}(z)$ and $E^{(b,\omega_i)}(z)$ that propagate in waveguide a and waveguide b , and use the normalized amplitudes $A_i^{(a,b)}(z)$, similarly defined in Equation (3.2). Their square represents a photon flux in waveguide a and b , respectively. This is useful to directly visualize and evaluate conversion efficiency in two waveguides as the interacting waves propagate based on photon numbers. And such photon conversion efficiencies are independent of the specific frequencies ω_1 , ω_2 and ω_3 of the three interacting waves.

4.1.1 Coupled wave equations and CLPM conditions

For two parallel guiding cores that are coupled to each other the wave interaction process is described by six coupled-wave equations for the amplitudes of the waves in the two cores (a) and (b). The three equations for core (a) are the same as those for a single-core, but with the addition of the coupling terms to the other core:

$$\frac{\partial}{\partial z} A_3^{(a)} = i \chi [A_2^{(a)}]^2 A_1^{(a)*} e^{i\Delta k z} + i \kappa_3 A_3^{(b)}, \quad (4.1)$$

$$\frac{\partial}{\partial z} A_2^{(a)} = 2i \chi A_1^{(a)} A_2^{(a)*} A_3^{(a)} e^{-i\Delta k z} + i \kappa_2 A_2^{(b)}, \quad (4.2)$$

$$\frac{\partial}{\partial z} A_1^{(a)} = i \chi [A_2^{(a)}]^2 A_3^{(a)*} e^{i\Delta k z} + i \kappa_1 A_1^{(b)}, \quad (4.3)$$

with a symmetric set of three equations describing the waves in core (b). Here, the $\kappa_i = \kappa(\omega_i)$ are the coupling constants of each wave between the two cores. A wave of frequency ω_i launched in one waveguide will fully move to the other waveguide after a coupling length $L_c = \pi/(2\kappa_i)$.

The coupled wave equations can be solved to show that CLPM phase-matching is achieved when any of the following expressions vanish [1],

$$\Delta K_\alpha = \Delta k - (\kappa_1 - \kappa_3) = 0, \quad (4.4)$$

$$\Delta K_\beta = \Delta k - (\kappa_1 - 2\kappa_2 + \kappa_3) = 0, \quad (4.5)$$

$$\Delta K_\gamma = \Delta k - (\kappa_1 + 2\kappa_2 + \kappa_3) = 0, \quad (4.6)$$

$$\Delta K'_\alpha = \Delta k + (\kappa_1 - \kappa_3) = 0, \quad (4.7)$$

$$\Delta K'_\beta = \Delta k + (\kappa_1 - 2\kappa_2 + \kappa_3) = 0, \quad (4.8)$$

$$\Delta K'_\gamma = \Delta k + (\kappa_1 + 2\kappa_2 + \kappa_3) = 0. \quad (4.9)$$

These equations replace the simple $\Delta k = 0$ condition for the single-mode case. Here, the quantities $\kappa_i = \kappa(\omega_i)$ essentially play the roles of wavevectors that compensate for the mismatch described by Δk . In general one can expect the coupling constants to be larger for larger wavelengths ($\kappa_1 > \kappa_2 > \kappa_3$). The coupling constants are related to the single-mode propagation constant and the propagation constants of

the supermodes in the dual-core configuration by (2.23) and (2.24) as derived in chapter 2.

Substituting the relationships (2.23) into (4.4-4.9), delivers the CLPM conditions in terms of the supermode propagation constants alone:

$$\Delta K_\alpha = \beta_2^{(e)} + \beta_2^{(o)} - \beta_1^{(e)} - \beta_3^{(o)} = 0 \quad (4.10)$$

$$\Delta K_\beta = 2\beta_2^{(e)} - \beta_1^{(e)} - \beta_3^{(e)} = 0 \quad (4.11)$$

$$\Delta K_\gamma = 2\beta_2^{(o)} - \beta_1^{(e)} - \beta_3^{(e)} = 0 \quad (4.12)$$

$$\Delta K'_\alpha = \beta_2^{(e)} + \beta_2^{(o)} - \beta_1^{(o)} - \beta_3^{(e)} = 0 \quad (4.13)$$

$$\Delta K'_\beta = 2\beta_2^{(o)} - \beta_1^{(o)} - \beta_3^{(o)} = 0 \quad (4.14)$$

$$\Delta K'_\gamma = 2\beta_2^{(e)} - \beta_1^{(o)} - \beta_3^{(o)} = 0 \quad (4.15)$$

where the unprimed and primed expression are obtained by using the upper sign or the upper sign in Equations (4.4-4.9), respectively.

CLPM phase matching, which was initially expressed in terms of the coupling constants alone, is thus clearly revealed to correspond to phase-matched mode-coupling between the supermodes of the dual-core structure.

4.1.2 Relationship to single-core phase matching

By moving to a dual-core configuration it becomes possible to choose between six different phase matching conditions instead of only one for a single-mode fiber. In addition to having a limited tunability of the phase matching condition by varying core-radius and refractive index contrast, we can now also tune the phase-matching condition by tuning the distance between the two cores. This added flexibility of CLPM in dual core fibers makes it easier and more flexible to obtain phase matching for third-order nonlinear optical interaction in different materials or for different guided structures. In the following we will discuss examples for the case of simple step-index fibers with a difference between cladding and core refractive index of 0.01, focusing on how the CLPM conditions can be tuned by the wavelengths participating in the interaction, and by geometrical parameters, in particular the distance between the cores.

CLPM conditions vs. generated wavelength

The six phase-matching conditions for frequency down-conversion in dual-core fibers, expressed in terms of the single-mode propagation constant in each core and the corresponding coupling constants in Equations (4.4-4.9), or in terms of the two lowest-frequency fundamental modes in the two-core system in Equations (4.10-4.15), are shown for a dual-core step-index silica fiber in Figure 4.2 as a function of the generated wavelength λ_1 and the distance between the cores for different choices of core radius and pump wavelengths λ_2 .

The dashed curves in the figure belong to $\Delta K'_\alpha$, $\Delta K'_\beta$, and $\Delta K'_\gamma$, for which the linear combinations of coupling constants is added to Δk , and they are generally larger than zero. On the other hand the solid curves belonging to ΔK_α , ΔK_β , and ΔK_γ , where the coupling constants are subtracted from the single-mode Δk , generally cross zero at wavelengths larger than the phase-matching wavelength for a single-core fiber.

Another very important difference between these two families of CLPM conditions is the fact that the ones corresponding to solid lines in Figure 3.4 cause the generated wavelength to appear in the even supermode of the dual-core structure, as can be clearly seen from Equations. (4.10-4.12), while the other three let the generated wavelength λ_1 appear in the odd supermode (see Equations. 4.13-4.15). The latter case would make it more difficult to extract the generated wave from the fiber, but the fact that this odd mode is required for phase matching also eliminates the possibility of using the corresponding phase matching conditions when the generated frequency is below the cut-off frequency for the odd supermode. This is shown quantitatively in the standard mode-dispersion plot of Figure 6.3.

We can see that $\Delta K_\gamma = 0$ is easier to be achieved from the plots, and it demonstrates the extension of the choice of the pump wave further when applying to the dual-core fiber. Additionally, with the same distance between the cores, smaller radius of the core and shorter pump wavelength would help $\Delta K_\alpha = 0$ to generate a wave at $1.65 \mu m$. This adds more flexibility in the dual-core fibers to achieve phase-matching conditions with both $\Delta K_\alpha = 0$ and $\Delta K_\gamma = 0$.

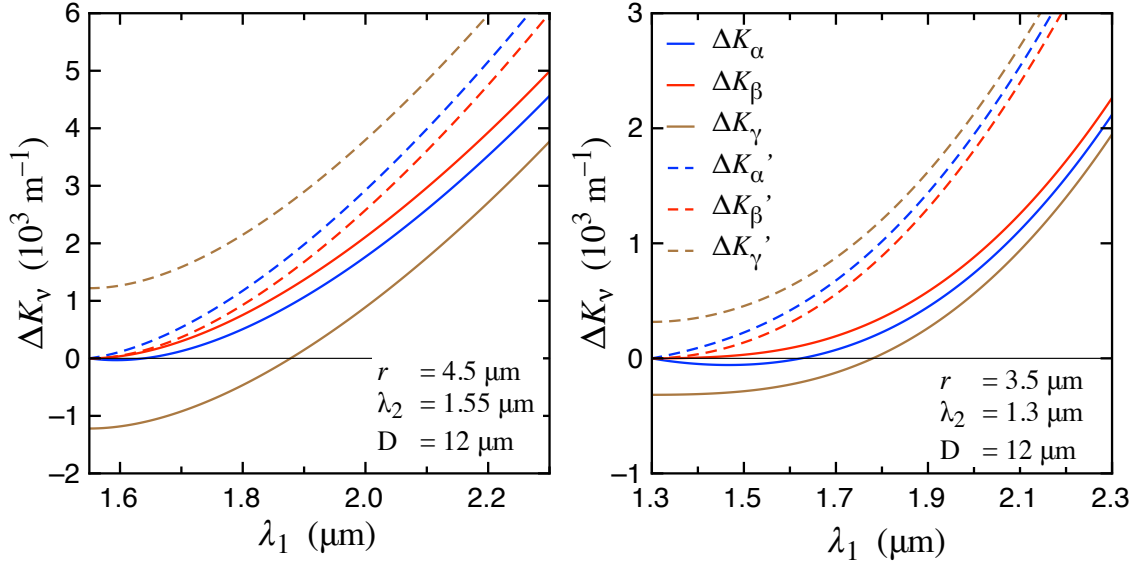


Figure 4.2: Behavior of the phase mis-match terms ΔK_ν for a dual core step-index silica fiber with refractive index contrast of 0.01. The plot on the left is for a fiber with a core radius of $4.5 \mu\text{m}$ and a pump wavelength of $1.55 \mu\text{m}$, on the right is for a core radius of $3.5 \mu\text{m}$ and a pump wavelength of $1.3 \mu\text{m}$. Both plots show the dependence of the ΔK_ν from the generated IR wavelength. The dashed curves correspond to the CLPM mismatches that involve the odd supermode for the generated wavelength (See Equations 4.13-4.15), and they are generally larger than zero. The order of the curves, from top to bottom corresponds to $\Delta K'_\gamma$, $\Delta K'_\alpha$, $\Delta K'_\beta$, ΔK_β , ΔK_α , ΔK_γ .

We should also note that the quantity ΔK_β of Equation (4.11) directly corresponds to the standard phase-mismatch for the lowest-frequency even supermode, and it is essentially the equivalent of the quantity plotted in Figure 3.4, with a weak dependence from the distance between the cores. The CLPM condition $\Delta K_\beta = 0$ applies when all interacting waves propagate in the lowest-frequency fundamental mode (or supermode), and its possible realization depends on characteristics of the bulk refractive index dispersion [28], as shown in the previous chapter.

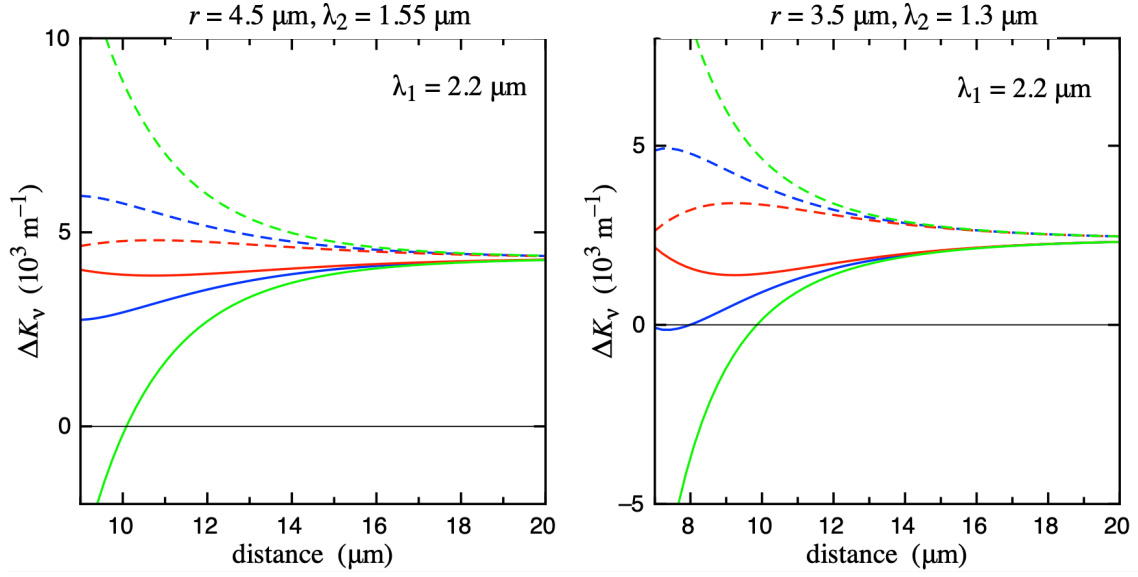


Figure 4.3: Behavior of the phase mis-match terms ΔK_ν for a dual core step-index silica fiber with refractive index contrast of 0.01. The plots on the left are for a fiber with a core radius of $4.5 \mu\text{m}$ and a pump wavelength of $1.55 \mu\text{m}$, those on the right are for a core radius of $3.5 \mu\text{m}$ and a pump wavelength of $1.3 \mu\text{m}$. Both plots show their dependence on the distance between the cores. The dashed curves correspond to the CLPM mismatches that involve the odd supermode for the generated wavelength (See Equations 4.13-4.15), and they are generally larger than zero. The order of the curves, from top to bottom corresponds to $\Delta K'_\gamma$, $\Delta K'_\alpha$, $\Delta K'_\beta$, ΔK_β , ΔK_α , ΔK_γ .

CLPM conditions vs. distance

The six phase-matching conditions for frequency down-conversion in dual-core fibers can also be presented as a function of the distance between the cores for two choices of core radius and two pump wavelengths λ_2 in Figure 4.3.

As the distance becomes larger and larger, all six phase-matching conditions converge to the same quantity of ΔK_ν due to weak coupling. It is essentially the equivalent of the quantity of Δk in the single-mode fiber. When two cores are closer, the quantities of ΔK_ν for six phase-matching conditions spread out increasingly, especially for ΔK_γ . To generate a wave at $2.2 \mu\text{m}$, we can structure the dual-core fiber with the distance of $10 \mu\text{m}$ and radius of $4.5 \mu\text{m}$ when pump at $1.55 \mu\text{m}$,

which makes $\Delta K_\gamma = 0$. Alternatively, we can also structure the dual-core fiber with the distance of $8 \mu m$ and radius of $3.5 \mu m$ when pump at $1.3 \mu m$, which makes $\Delta K_\alpha = 0$. This demonstrates more flexibility in the dual-core fibers to achieve phase-matching conditions as well.

4.1.3 CLPM phase matching diagrams

The design parameters that come into question in order to realize the CLPM conditions are the radius of each core, the distance of the cores, and the refractive index contrast. In the following we consider examples for simple step-index silica and fluoride fiber with a difference between cladding and core refractive index of 0.01, focusing on the requirement of core radius and distance between two cores when $\Delta K_\alpha = 0$ and $\Delta K_\gamma = 0$ CLPM conditions are achieved.

CLPM conditions in a (wavelength, wavelength) diagram

Figure 4.4 shows phase-matching curves similar to those plotted in Figure 3.4(b) for different center-to-center distance between the two cores. The phase matching curve in the limit of large distances between the cores corresponds to that plotted in Figure 3.4(b). But as the distance between the cores decreases, the coupling between the cores extends the phase matching possibilities to a much wider range of pump wavelengths. This is because the effect of the coupling as the distance between the cores decreases adds a distance-dependent offset to the condition $\Delta k = 0$, as seen in Equations (4.4-4.9), which changes the shape of the wavelength CLPM curves in a slightly different way for the two conditions ($\Delta K_\alpha = 0$ and $\Delta K_\gamma = 0$) that we are considering here. The way this happens also depends on the radius of the core, which influences the coupling. For $\Delta K_\alpha = 0$, the generated wavelength λ_1 moves to smaller values as the pump wavelength increases, but it can be maintained above $2 \mu m$ by going to smaller pump wavelengths λ_2 while also reducing the core-radius. Because of this, we chose to plot the CLPM curves for this condition using a core radius of $3.5 \mu m$.

The same kind of analysis that we used in the previous section can also be applied

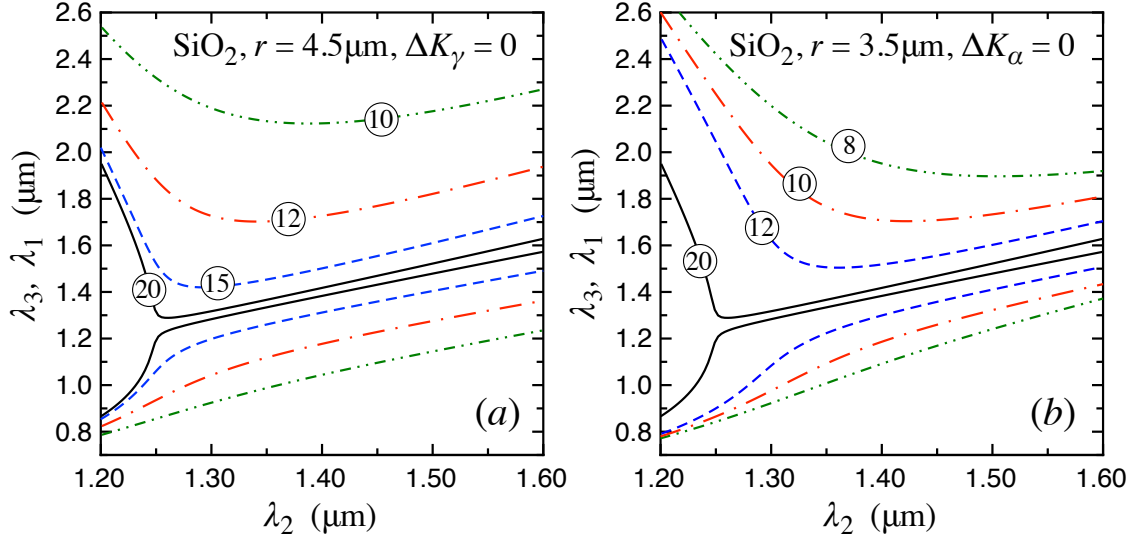


Figure 4.4: Wavelength CLPM curves for different distances between the cores in a dual-core step-index fiber with a refractive index contrast of $\Delta n = 0.01$. Left: $\Delta K_\gamma = 0$ for a core radius of $4.5 \mu\text{m}$. Right: $\Delta K_\alpha = 0$ for a core radius of $3.5 \mu\text{m}$. The CLPM curves are labeled with the center-to-center distance between the cores in micrometers.

to fibers that transmit further into the infrared such as those made from ZrF_4 or InF_3 glasses. For simplicity, and for better comparison with the case of silica fibers discussed above, we keep the same index-contrast and core-radius of $4.5 \mu\text{m}$ that we used for most of the silica examples.

The main difference between these fluoride glasses and silica is well represented by the maximum pump wavelength that can be used to achieve phase matching in the single-core configuration. It increases from $\lambda_2^{\text{max}} = 1.248 \mu\text{m}$ for silica to $\lambda_2^{\text{max}} = 1.63 \mu\text{m}$ for ZrF_4 to $\lambda_2^{\text{max}} = 1.91 \mu\text{m}$ for our InF_3 example. This then has a clear effect on the wavelength CLPM conditions, which we plotted for silica in Figure 4.4.

The wavelength CLPM curves for ΔK_α and ΔK_γ in a ZrF_4 fiber are shown in Figure 4.5. The core radius of $4.5 \mu\text{m}$ increases the coupling and creates larger deviations from the single-core limit as the two cores move closer to each other. For these configuration, the down-converted wave with wavelength λ_1 is always created

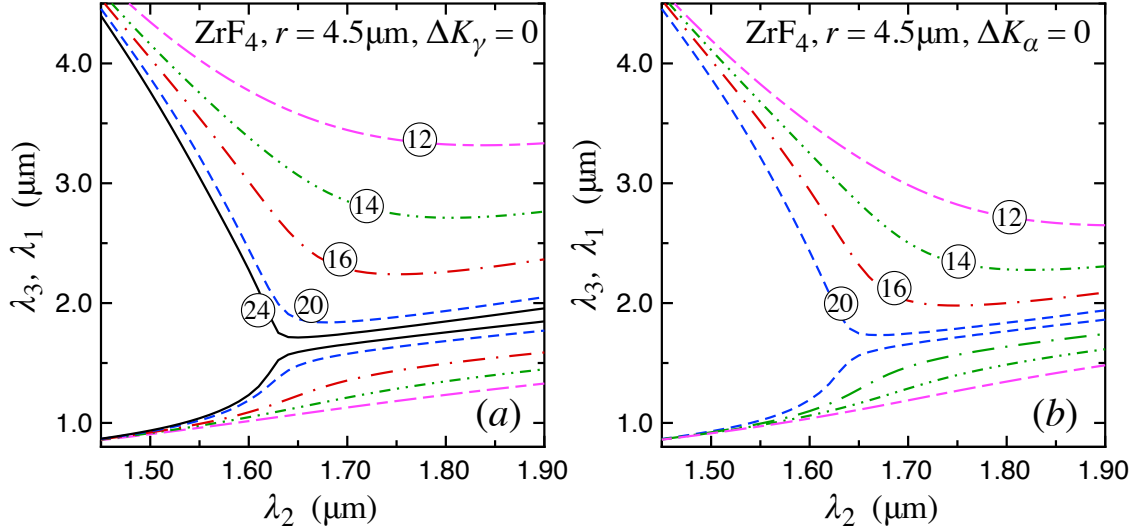


Figure 4.5: Wavelength CLPM curves for different distances between the cores in a ZrF_4 dual core step-index fiber with a refractive index contrast of $\Delta n = 0.01$ and core radius of $4.5 \mu\text{m}$. Left: $\Delta K_\gamma = 0$. Right: $\Delta K_\alpha = 0$. The CLPM curves are labeled with the center-to-center distance between the cores in micrometers.

in the even supermode, which will always exist. In this case, the small radius of $4.5 \mu\text{m}$ makes it possible to still use a pump wavelength of $1.55 \mu\text{m}$ to generate infrared light with wavelengths between $\lambda_1 \sim 3 \mu\text{m}$ and $\lambda_2 \sim 4 \mu\text{m}$ in the ZrF_4 fiber when varying the distance between the cores between 20 and $12 \mu\text{m}$.

The situation in InF_3 fibers is qualitatively the same, but the maximum pump wavelength for single-mode interaction in this material is significantly larger, and it is not possible anymore to use either ΔK_α or ΔK_γ to generate waves that are still in the transparency range of the material when using a pump wavelength of $1.55 \mu\text{m}$. In addition, even if they were not absorbed, the wavelengths of the generated waves would also be quite insensitive to changing the distance between the cores.

However, the large $\lambda_2^{\text{max}} = 1.91 \mu\text{m}$ for InF_3 also means that pump wavelengths around $1.55 \mu\text{m}$ will be in a region where $\Delta k < 0$ and then it becomes possible to obtain phase matching using $\Delta K'_\alpha$ and $\Delta K'_\gamma$. We plot these curves here, even if they are of limited use, because they are qualitatively different from what we have

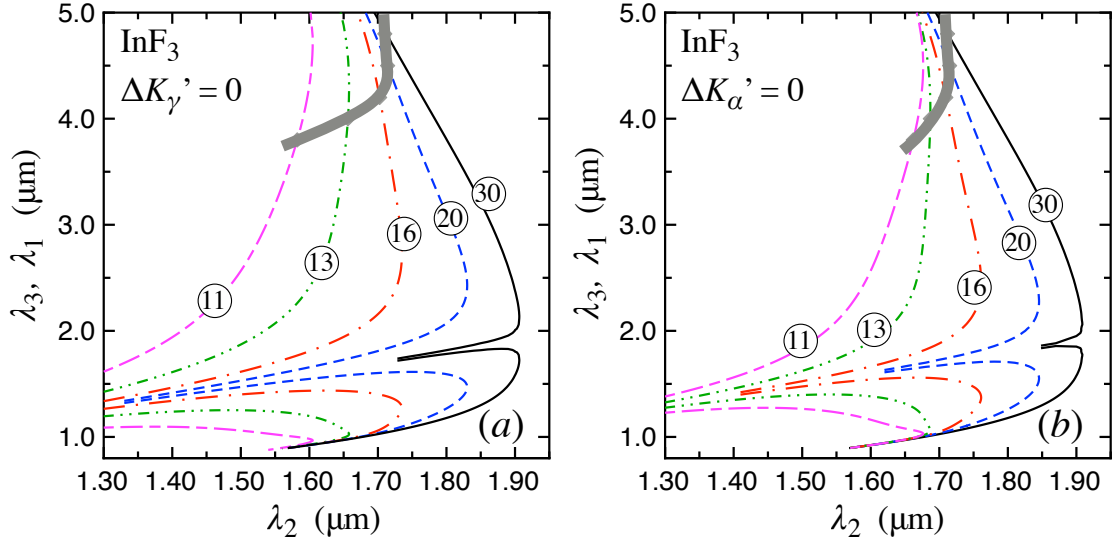


Figure 4.6: Wavelength CLPM curves for different distances between the cores in a InF_3 dual core step-index fiber with a refractive index contrast of $\Delta n = 0.01$ and core radius of $4.5 \mu\text{m}$. Left: $\Delta K'_\gamma = 0$. Right: $\Delta K'_\alpha = 0$. The CLPM curves are labeled with the center-to-center distance between the cores in micrometers. For these two CLPM conditions the longest wavelength generated wave is created in the odd supermode; the thick dotted lines towards the top of the graphs are the boundary above which the odd mode for the λ_1 disappears for the given distance between the cores.

seen up to now, with the generated wavelength λ_1 decreasing for decreasing distance between the cores. This is shown in Figure 4.6. We note the limited tunability range between $\lambda_1 \approx 2 \mu\text{m}$ $\lambda_1 \approx 3 \mu\text{m}$ and that can be achieved in this way in the InF_3 fiber. This, coupled with the fact that the generated wave is created in an odd supermode, makes InF_3 an inferior choice when compared to ZrF_4 .

CLPM conditions in a (radius, distance) diagram

Figure 4.7 shows how the same two CLPM conditions vary with core radius and distance for a fixed choice of refractive index contrast of $\Delta n = 0.01$. Top row presents $\Delta K_\alpha = 0$ CLPM condition is achieved for pump wavelengths varying from $1.2 \mu\text{m}$ to $1.5 \mu\text{m}$. The core radius is mostly confined within the range between

3 μm to 5 μm when pumped at 1.2 μm . With increasing the pump wavelength towards 1.5 μm , the core radius is already below 4 μm and keeps shorter so that the generated wavelength is still above 2 μm .

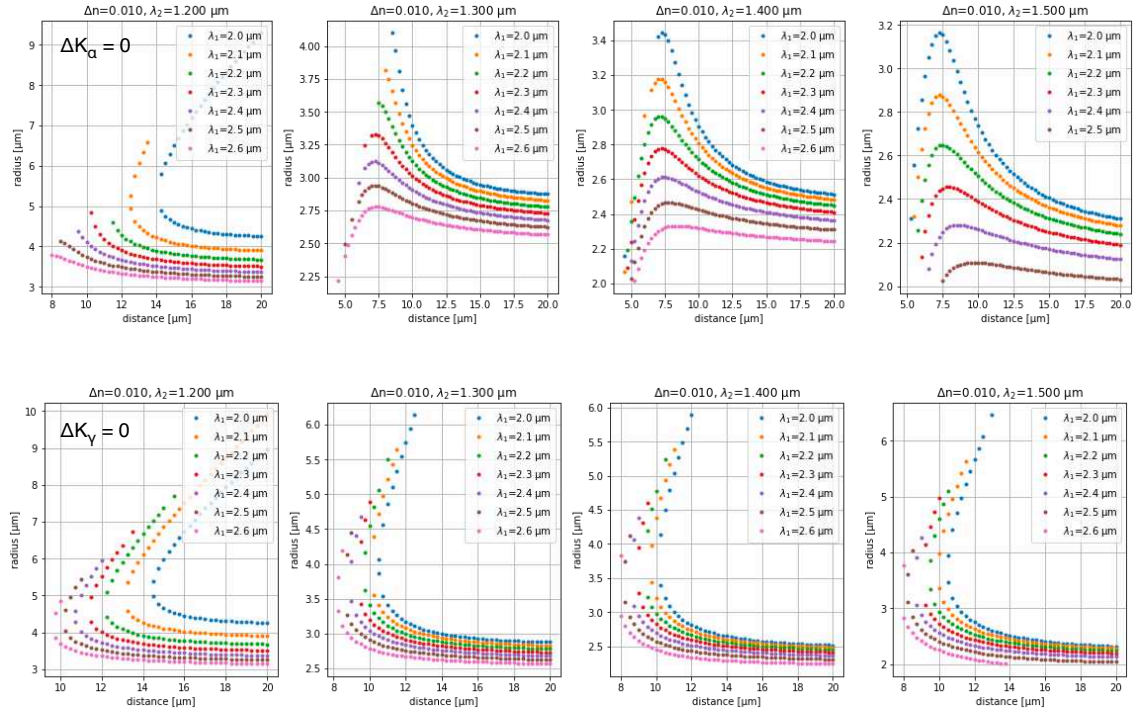


Figure 4.7: Geometry CLPM curves for different down-converted wavelengths in dual-core step-index fiber with a refractive index contrast of $\Delta n = 0.01$. Top row: $\Delta K_\alpha = 0$ for pump wavelengths varying from 1.2 μm to 1.5 μm . Bottom row: $\Delta K_\gamma = 0$ for pump wavelengths varying from 1.2 μm to 1.5 μm .

The plot for $\Delta K_\gamma = 0$ is quite different from $\Delta K_\alpha = 0$, we find two choices of the values of core radius can be selected to a fixed distance. The larger core corresponds to the stronger coupling between two cores. As the pump wave moves further, it is still possible to either reduce the core radius or make the distance closer between two cores to achieve CLPM rules. This also adds the tunability of choosing the specific design of the structure in dual-core fibers when generating the wavelength of interest.

Figure 4.8 shows the influence of the refractive index contrast to the geometric

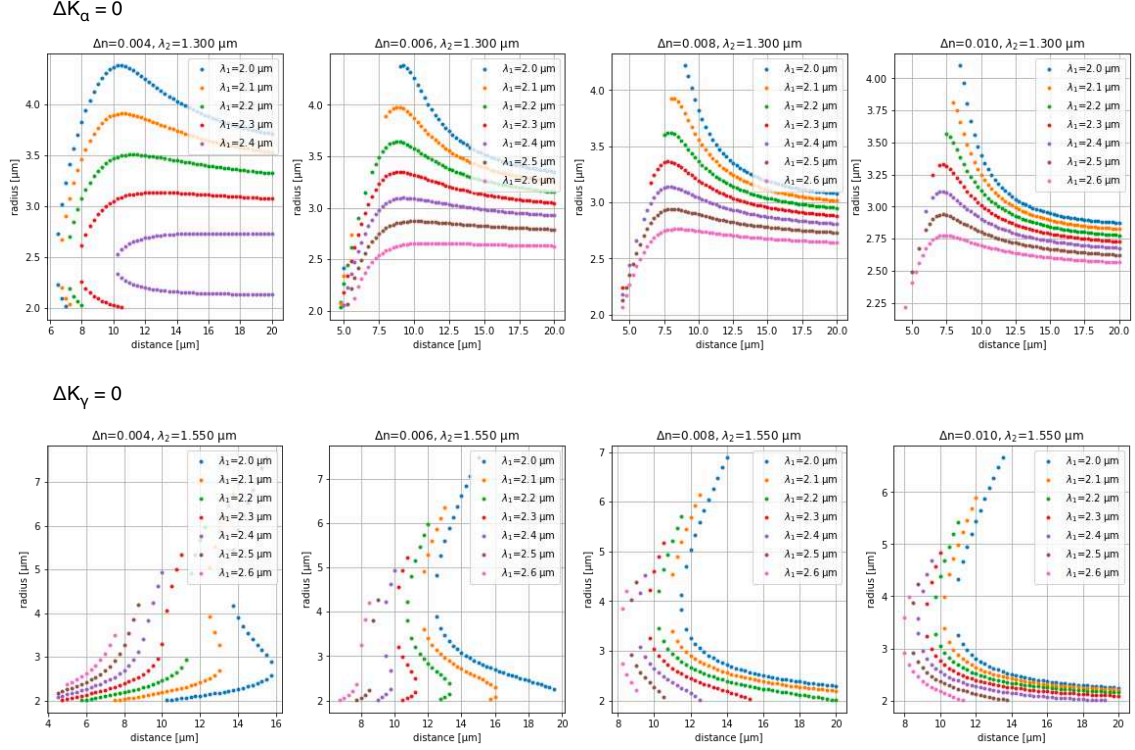


Figure 4.8: Geometry CLPM curves for different down-converted wavelengths in dual-core step-index fiber with different refractive index contrast. Top row: $\Delta K_\alpha = 0$ for a pump wavelength $1.3 \mu m$. Bottom row: $\Delta K_\gamma = 0$ for a pump wavelength $1.55 \mu m$.

parameter for both $\Delta K_\alpha = 0$ and $\Delta K_\gamma = 0$. For $\Delta K_\alpha = 0$ a pump wavelength of $1.3 \mu m$ is selected, while we chose a pump wavelength of $1.55 \mu m$ for $\Delta K_\gamma = 0$, and in both cases we considered the same generated wavelengths between $\lambda_1 = 2.0 \mu m$ and $\lambda_1 = 2.6 \mu m$. The situation that the refractive index contrast of $\Delta n = 0.004$ is different compared others. Two choices of the values of core radius can be selected to produce a wave at $2.4 \mu m$ when $\Delta K_\alpha = 0$. The choice of core radius is no longer limited, since we can always achieve CLPM by tuning the distance between two cores. $\Delta K_\gamma = 0$ also provides a wide range of the choices of both core radius and distance by varying the refractive index contrast. Especially, we are able to select three values of core radius to a fixed distance with the refractive index contrast of

$\Delta n = 0.004$ if we want to produce a wave at $2.1 \mu m$, for instance. This also adds flexibility of tuning different geometric parameters.

4.1.4 Discussion

The CLPM conditions (4.10) and (4.13) can only lead to maximum conversion efficiency when the pump wave is launched into the even and odd fundamental supermode, respectively. When the pump wave is launched in only one of the two cores, then the conversion efficiency is limited to a maximum of 50%, because only one of the supermodes benefits from phase-matched coupling to the generated wave, and therefore only half of the injected pump-wave photons can be used to create the generated wave [1].

In the following section we will concentrate on the case where the pump wave at ω_2 is injected in one core only, which then leads to its intensity oscillating between the two cores, and on the use of the CLPM conditions $\Delta K_\alpha = 0$ and $\Delta K_\gamma = 0$ (upper sign in the first two equations of 4.4 and 4.6, or Equations 4.10 and 4.12).

The first of these two CLPM conditions is the best one to achieve maximum conversion efficiency when the pump wave is injected in just one core because it requires the presence of both the odd and even supermodes for the pump wave. Its other advantages is that it does not depend on the coupling constant κ_2 and that (as we will see later) the corresponding nonlinear interaction is not affected by the refractive index changes caused by self- and cross-phase modulation.

The second CLPM condition, $\Delta K_\gamma = 0$, is also attractive (4.12), despite the fact that its maximum conversion efficiency is limited by the fact that it will only allow half of the pump wave photons (those in the odd supermode), because it can extend the range of pump wavelengths over which CLPM can be obtained.

4.2 Accounting for self- and cross-phase modulation in CLPM

When the phase modulation terms are considered, we can re-write six coupled-wave equations (4.1 - 4.3) for the amplitudes of the waves in two cores (a) and (b).

$$\begin{aligned} \frac{\partial}{\partial z} A_3^{(a)} &= i \chi [A_3^{(a)} (A_3^{(a)} A_3^{(a)*} + 2A_2^{(a)} A_2^{(a)*} + 2A_1^{(a)} A_1^{(a)*}) \\ &\quad + [A_2^{(a)}]^2 A_1^{(a)*} e^{i\Delta k z}] + i \kappa_3 A_3^{(b)}, \end{aligned} \quad (4.16)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_2^{(a)} &= i \chi [A_2^{(a)} (2A_3^{(a)} A_3^{(a)*} + A_2^{(a)} A_2^{(a)*} + 2A_1^{(a)} A_1^{(a)*}) \\ &\quad + 2A_1^{(a)} A_2^{(a)*} A_3^{(a)} e^{-i\Delta k z}] + i \kappa_2 A_2^{(b)}, \end{aligned} \quad (4.17)$$

$$\begin{aligned} \frac{\partial}{\partial z} A_1^{(a)} &= i \chi [A_1^{(a)} (2A_3^{(a)} A_3^{(a)*} + 2A_2^{(a)} A_2^{(a)*} + A_1^{(a)} A_1^{(a)*}) \\ &\quad + [A_2^{(a)}]^2 A_3^{(a)*} e^{i\Delta k z}] + i \kappa_1 A_1^{(b)}, \end{aligned} \quad (4.18)$$

with a symmetric set of three equations describing the waves in core (b).

In the above coupled-wave equations, the cross- and self-phase modulation terms are always phase matched, lacking the multiplication with the exponential containing the wavevector mismatch Δk . However, nonlinear phase-modulation can give rise to the accumulation of an additional phase shift between the interacting waves during propagation. While this effect can in principle disrupt the energy transfer process between the waves, limiting conversion efficiency, here we discuss how this disruption is intensity-dependent, and how it can be compensated by adding an appropriate intensity-dependent term to the CLPM conditions (4.4-4.9).

4.2.1 Difference from single-mode case

To discuss the influence self- and cross-phase modulation on the ability to convert a large portion of the power of the pump wave into the generated waves, we use an analytical approach similar to what we have used for a single-mode fiber. It is too complicated where the result would also depend on how the pump wave is injected into the dual-core system. But a numerical solution of the coupled-wave equations (4.16-4.18) can be used to obtain in general the dependence of the wave amplitudes

as a function of propagation length using different pump intensity values. In this way we could derive the expressions that must be used to account for the effect of phase-modulation and still obtain the maximum conversion efficiency. While for a single-mode fiber, the expression that corresponds to maximum conversion efficiency is Equation 3.23, for the dual-core configuration the equivalent expressions are

$$\Delta K_\alpha = 0 \quad (4.19)$$

$$\Delta K_\gamma = \frac{G}{4} + \eta G^2, \quad (4.20)$$

which are valid when the pump wave at wavelength λ_2 is injected in one core. When these conditions are fulfilled, the solutions of the coupled wave equations can always evolve, after a given intensity-dependent interaction length, towards the maximum theoretical conversion efficiency.

The most important result here is that the $\Delta K_\alpha = 0$ CLPM configuration is insensitive to the phase modulation, therefore intensity-independent. In other words, when this condition is fulfilled, the coupled-wave equations for the evolution of the wave amplitudes can always lead to the complete depletion of the pump wave. This insensitivity to phase modulation is a peculiar advantage of this configuration, in addition to its 100% photon conversion efficiency.

For the $\Delta K_\gamma = 0$ CLPM configuration, on the other hand, we found that phase modulation does have an effect, but that can be taken into account in a similar way as for the single-mode fiber, this time using Equation (4.20), where η is a small correction ($\eta G \ll 1$) that is essentially intensity-independent but can change slightly depending on the coupling between the cores. We confirmed the validity of Equation (4.20) up to the highest practical G -values of 100 m^{-1} . Still, the sensitivity to phase modulation is significantly attenuated also in this configuration when compared to the single-core configuration, where the G -dependent correction is four-times larger (this factor of four may be related to the fact that in this dual-core configuration the pump wave only occupies one of the cores for half the interaction length).

In practice, the plots in Figure 4.4 are essentially not changed when substituting Equation (4.20) for $\Delta K_\gamma = 0$. Even for $G = 100 \text{ m}^{-1}$, only the curves for a

large distance between the cores are slightly modified (by a small displacement corresponding to a change in distance by 10%). In fact, using Equation (4.20) for Figure 4.8 does not lead to any important change in the curves plotted there, even for this relatively large value of G . As an example, the core-distance required to down-convert to $\lambda_1 = 2.2 \mu\text{m}$ using the $\Delta K_\gamma = 0$ CLPM configuration and a core-radius of $4.5 \mu\text{m}$ changes by less than 1% when taking into account phase modulation.

CLPM in dual-core fibers is characterized by an intrinsic robustness against self- and cross-phase modulation, which have no effect at all on the conditions derived using the $\Delta K_\alpha = 0$ CLPM configuration, and only a small effect on those derived for the $\Delta K_\gamma = 0$ CLPM configuration. In addition, we note that in a practical situation where the dual-core scheme is used to down-convert a single pump-wave, the effect of phase-matching is just to slightly shift the frequencies of the generated waves away from those calculated for a certain fiber geometry. For the example plotted in Figure 4.4, the choice of $\lambda_2 = 1.55 \mu\text{m}$ and a distance between the cores of $10 \mu\text{m}$ leads to downconversion to $\lambda_1 = 2.2 \mu\text{m}$, and phase-modulation only shifts this value by much less than 1%.

4.2.2 Numerical solutions corresponding to the CLPM conditions

The numerical solutions of the evolution of the interacting waves with propagation distance, as predicted by the coupled-wave equations (4.16-4.18), is depicted in Figure 4.9. To better show the difference between these two configurations for third-order wavelength down-conversion in a dual core fiber we show in Fig.4.9 the evolution of the normalized wave amplitudes A_i in one of the two cores as a function of propagation length. We do that for a core radius of 3.5μ and a pump wavelength of $1.3 \mu\text{m}$ for $\Delta K_\alpha = 0$, and for a core radius of $4.5 \mu\text{m}$ and a pump wavelength of $1.55 \mu\text{m}$ for $\Delta K_\gamma = 0$ and its modification of Equation (4.20). In both cases we kept the generated wavelength at $\lambda_1 = 2.2 \mu\text{m}$ and adapted the distance between the cores to vary the values of ΔK_α and ΔK_γ .

Figure 4.9(a) and (b) describes spatial evolution of the amplitudes of the interacting waves when CLPM condition $\Delta K_\alpha = 0$ is fulfilled and is not fulfilled. We confirm that self- and cross-phase modulation is insensitive, which has no effect at all on the conditions derived using the $\Delta K_\alpha = 0$ CLPM configuration. When $\Delta K_\alpha = 20 \text{ m}^{-1}$, a remarkable decrease of A_1 at the conversion length while A_2 would not be depleted towards zero. As discussed earlier, $\Delta K_\alpha = 0$ is the best one to achieve 100% photon conversion efficiency because it requires the presence of both the odd and even supermodes for the pump wave. This is much useful if we can achieve 100% photon conversion efficiency at around one meter in fiber applications.

As for $\Delta K_\gamma = 0$, the influence of self- and cross- phase modulation seems to be more serious, which results in a smaller growth of the generated wave. However, with the correction of ΔK_γ using (4.20) we can still achieve the maximum photon conversion efficiency when the propagation length is at 1.5 m. Despite the fact that its maximum conversion efficiency is limited to 50% by the fact that it will only require half of the pump wave photons (those in the odd supermode). We verify that the correction is tested to be $G/4$ (where $\eta \ll 1$) so that half of the pump wave photons can be converted. η may be ignored when only half maximum photon efficiency is considered to be achieved for fiber applications, which makes CLPM condition, $\Delta K_\gamma = G/4$, attractive. Then, we only need to worry about the slight shift of the frequencies of the generated waves away from those calculated for a certain fiber geometry.

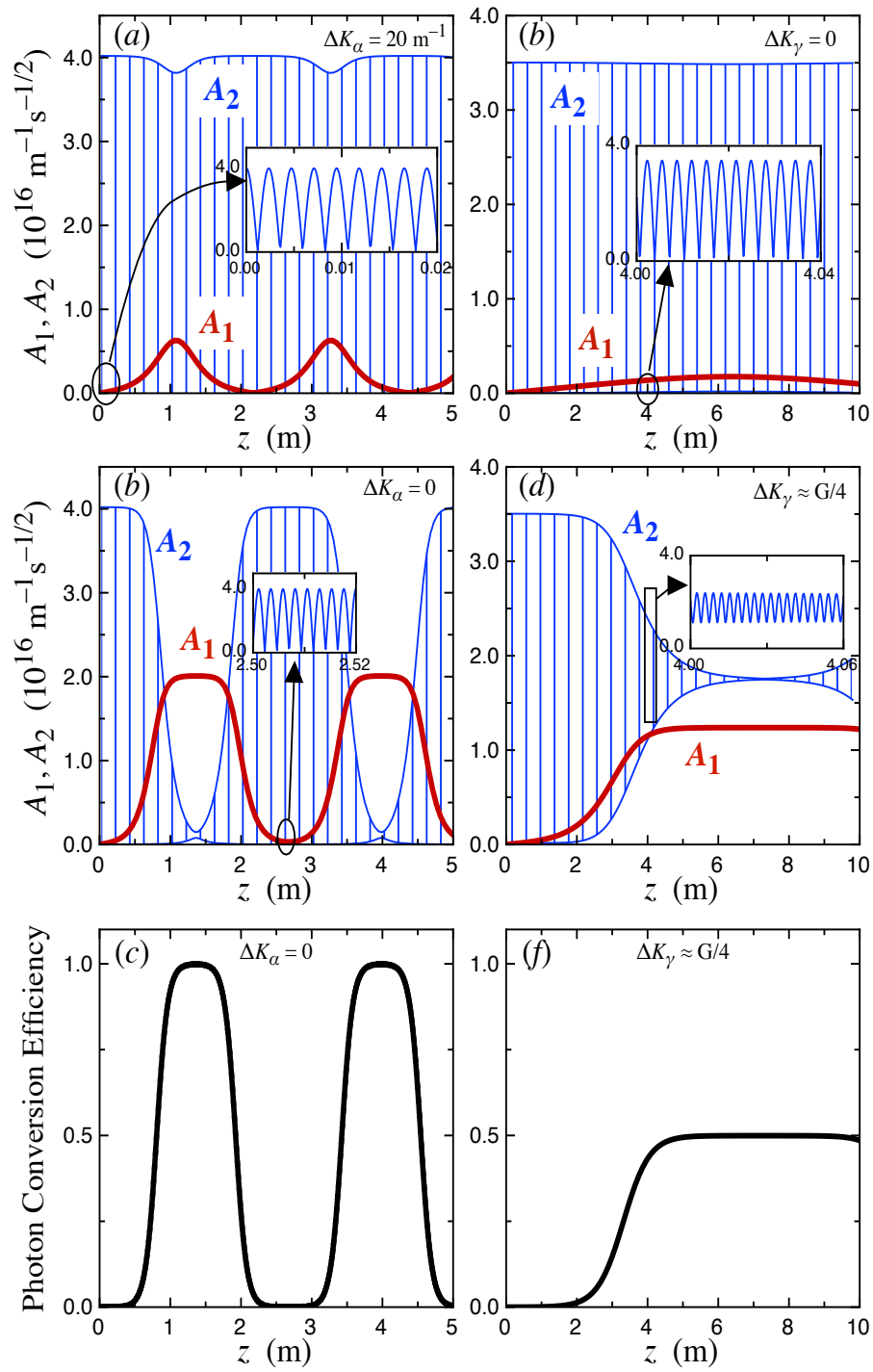


Figure 4.9: (Caption next page.)

Figure 4.9: Amplitudes of the interacting waves and photon conversion efficiency vs. propagation distance for a dual-core silica fiber with refractive index contrast of 0.01 in CLPM configurations that lead to frequency downconversion to a generated wave with $\lambda_1 = 2.2 \mu\text{m}$. While the curves for A_1 are the full z -dependence, showing that the generated wave does not oscillate between the fiber cores, the curves for A_2 are the *envelope* of oscillations that occur over coupling lengths of the order of millimeters, the vertical stripes are drawn between the minimum and the maximum of the oscillations. The calculations assume a 10 ns long pump-wave pulse with an energy of $25 \mu\text{J}$ with optimum coupling into one single core (peak intensity $\sim 10^{14} \text{ W/m}^2$). Graphs in the left column (a-c): ΔK_α CLPM configuration. Pump wave at $\lambda_2 = 1.3 \mu\text{m}$, core radius $r = 3.5 \mu\text{m}$, center-to-center core distance $d = 7.99 \mu\text{m}$, $G=7.4 \text{ m}^{-1}$. CLPM condition $\Delta K_\alpha = 0$ is not fulfilled for (a), and is fulfilled for (b) and (c). Graphs in the right column (d-e): ΔK_γ CLPM configuration. Pump wave at $\lambda_2 = 1.55 \mu\text{m}$, core radius $r = 4.5 \mu\text{m}$, center-to-center core distance $d = 10.09 \mu\text{m}$, $G=4.1 \text{ m}^{-1}$. CLPM condition $\Delta K_\gamma = 0$ is fulfilled for (d) but without the phase-modulation correction, while for panel (e) $\Delta K_\gamma = G/4 + \eta G^2$. Panels (c) and (f) show the photon conversion efficiency for the two optimum settings in both CLPM configurations.

Chapter 5

Experimental study and modeling of instantaneous and delayed response in DFWM

In chapter 2-4, we studied the four-wave mixing in optical fibers, and considered the propagation direction of multi-waves with different frequencies is the same. Additionally, We also studied and modeled the instantaneous and temporal response of nonlinear optical organic molecules using pump-and-probe Degenerate Four-wave Mixing (DFWM) method. For the free space DFWM, we investigate multi-waves with the same frequency that travel in different directions.

Dynamics of third-order nonlinear optical processes is of considerable theoretical and experimental interest because of their potential applications in all-optical signal processing [50, 51]. Time-resolved DFWM is a convenient method to probe the dynamics of resonant third-order nonlinear optical response. From the theoretical and experimental results that a resonant DFWM actually involves two dynamic process, ie., a coherent nonlinear response involving four-photon mixing and an incoherent nonlinear response due to diffraction from the population grating. The contribution of each process to the DFWM signal and the dynamic characteristics of each process are discussed. In this chapter, we present both theoretical derivation and numerical

simulation for the resonant DFWM, and the response due to diffraction from the population grating.

The organic molecules studied in this work are presented in the following, which possess a large third-order nonlinear optical susceptibility $\chi^{(3)}$. We make several liquid samples with different concentrations, and present the DFWM measurements of the response time of $\chi^{(3)}$ at different wavelengths. From this experiment, the lifetime of the lowest excited-state of the molecules can be measured. Our experimental results are satisfactorily explained by the theoretical derivation and simulation presented here.

5.1 Theoretical modeling of instantaneous DFWM and population gratings

5.1.1 Theoretical derivation of DFWM and population gratings

Polarization $\vec{P}$ to an applied optical field is given as follows:

$$\vec{P} = \epsilon_0 \chi^{(1)} \vec{E} + \epsilon_0 \chi^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \quad (5.1)$$

We assume that each molecule in the medium is a resonant two-level energy system. $|a\rangle$ is the ground state and $|b\rangle$ is the first-excited state [52].

$$\vec{P}_a = \epsilon_0 \chi_a^{(1)} \vec{E} + \epsilon_0 \chi_a^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \quad (5.2)$$

$$\vec{P}_b = \epsilon_0 \chi_b^{(1)} \vec{E} + \epsilon_0 \chi_b^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \quad (5.3)$$

Where

$$\chi_a^{(1)} = N_a f \alpha_a \quad (5.4)$$

$$\chi_b^{(1)} = N_b f \alpha_b \quad (5.5)$$

$$\chi_a^{(3)} = N_a f^4 \gamma_a \quad (5.6)$$

$$\chi_b^{(3)} = N_b f^4 \gamma_b \quad (5.7)$$

N_a : density of ground state molecules.

N_b : density of excited state molecules.

$f = \frac{n^2 + 2}{3}$: local field factor.

α_a : first order polarizability of ground state.

α_b : first order polarizability of excited state.

γ_a : third order polarizability of ground state.

γ_b : third order polarizability of excited state.

In this model, the transition is far from saturation and diffusion effect is neglected. Therefore, the molecular density in the ground state is approximately equal to the total density of molecules($N_a \approx N$), and the intensity in the medium $I \rightarrow 0$.

$$\frac{dN_b(\vec{r}, t)}{dt} = \delta(N_a - N_b)I(\vec{r}, t) - \frac{N_b(\vec{r}, t)}{T} \quad (5.8)$$

We discuss $N_a \approx N$ and $N_b \ll N_a$ approximation. So $N_a - N_b \approx N$. The excited-state population is followed by the kinetic equation:

$$\frac{dN_b(\vec{r}, t)}{dt} = \delta N I(\vec{r}, t) - \frac{N_b(\vec{r}, t)}{T} \quad (5.9)$$

In the equation above,

$$N = N_a + N_b$$

δ : absorption cross section related to absorption coefficient at given density.

T : excited state lifetime of the molecule.

$I(\vec{r}, t)$: intensity in the medium.

Then the kinetic equation can be solved as

$$N_b(\vec{r}, t) = e^{-\frac{t}{T}} \int_{-\infty}^t \delta NI(\vec{r}, t') e^{\frac{t'}{T}} dt' \quad (5.10)$$

From Equations (5.2-5.7), we can obtain the polarization derived from the ground and excited states as

$$\begin{aligned} \vec{P}_a(\vec{r}, t) &= \epsilon_0 \chi_a^{(1)} \vec{E} + \epsilon_0 \chi_a^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \\ &= \epsilon_0 (N - N_b) f \alpha_a \cdot \vec{E} + \epsilon_0 (N - N_b) f^4 \gamma_a \cdot \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \\ &= \epsilon_0 N f \alpha_a \cdot \vec{E} + \epsilon_0 N f^4 \gamma_a \cdot \vec{E} \cdot \vec{E} \cdot \vec{E} - \epsilon_0 N_b f \alpha_a \cdot \vec{E} + \dots \\ &= \epsilon_0 N f \alpha_a \cdot \vec{E} + \epsilon_0 N f^4 \gamma_a \cdot \vec{E} \cdot \vec{E} \cdot \vec{E} - \epsilon_0 f \alpha_a \cdot \vec{E} e^{-\frac{t}{T}} \int_{-\infty}^t \delta NI(\vec{r}, t') e^{\frac{t'}{T}} dt' + \dots \\ &= \epsilon_0 \chi^{(1)} \vec{E} + \epsilon_0 \chi^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} - \epsilon_0 f \alpha_a \cdot \vec{E} e^{-\frac{t}{T}} \int_{-\infty}^t \delta NI(\vec{r}, t') e^{\frac{t'}{T}} dt' + \dots \end{aligned} \quad (5.11)$$

$$\begin{aligned} \vec{P}_b(\vec{r}, t) &= \epsilon_0 \chi_b^{(1)} \vec{E} + \epsilon_0 \chi_b^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \\ &= \epsilon_0 N_b f \alpha_b \cdot \vec{E} + \epsilon_0 N_b f^4 \gamma_b \cdot \vec{E} \cdot \vec{E} \cdot \vec{E} + \dots \\ &= \epsilon_0 N_b (f \alpha_b \cdot \vec{E} + f^4 \gamma_b \cdot \vec{E} \cdot \vec{E} \cdot \vec{E}) + \dots \\ &= \epsilon_0 (f \alpha_b \cdot \vec{E} + f^4 \gamma_b \cdot \vec{E} \cdot \vec{E} \cdot \vec{E}) \cdot e^{-\frac{t}{T}} \int_{-\infty}^t \delta NI(\vec{r}, t') e^{\frac{t'}{T}} dt' + \dots \end{aligned} \quad (5.12)$$

We ignore the terms $f^4 \gamma_b \cdot \vec{E} \cdot \vec{E} \cdot \vec{E}$ in the last line. Then the total polarization from both the ground-state and the excited-state is

$$\begin{aligned} \vec{P}(\vec{r}, t) &= \vec{P}_a(\vec{r}, t) + \vec{P}_b(\vec{r}, t) \\ &= \epsilon_0 \chi^{(1)} \vec{E} + \epsilon_0 \chi^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} + \epsilon_0 f (\alpha_b - \alpha_a) \cdot \vec{E} e^{-\frac{t}{T}} \int_{-\infty}^t \delta NI(\vec{r}, t') e^{\frac{t'}{T}} dt' + \dots \\ &= P^{(1)}(\vec{r}, t) + [P^{(3)}(\vec{r}, t) + \vec{P}_n(\vec{r}, t)] + \dots \end{aligned} \quad (5.13)$$

The first and second terms in the equation above are respectively, the linear polarization and the third-order polarization, and others are higher orders that can be ignored. We only look at the third-order polarization

$$\vec{P}(\vec{r}, t) = P^{(3)}(\vec{r}, t) + \vec{P}_n(\vec{r}, t) \quad (5.14)$$

Now we can separate the polarization into two contributions: the first term describes a four-wave mixing process that is instantaneous. The second term describes the population grating contribution. The formation of a population grating is induced by two incident beams due to linear absorption, following by the Bragg diffraction of the third beam to generate the DFWM signal. The second term is not instantaneous, which depends on the excited-state lifetime of the molecule when the population grating decays [53].

$$P^{(3)}(\vec{r}, t) = \epsilon_0 \chi^{(3)} \vec{E} \cdot \vec{E} \cdot \vec{E} \quad (5.15)$$

$$\vec{P}_n(\vec{r}, t) = \epsilon_0 f(\alpha_b - \alpha_a) \cdot \vec{E} e^{-\frac{t}{T}} \int_{-\infty}^t \delta N I(\vec{r}, t') e^{\frac{t'}{T}} dt' \quad (5.16)$$

5.1.2 Analysis of instantaneous DFWM

For DFWM signal, the total electric field is

$$\vec{E} = \sum_{j=1}^4 \frac{1}{2} E_j \cdot e^{i(\vec{k}_j \cdot \vec{z} - \omega t)} + c.c \quad (5.17)$$

The electric field amplitude of each beam is

$$\varepsilon_j = \frac{1}{2} E_j(t) \cdot e^{i(\vec{k}_j \cdot \vec{z} - \omega t)} + c.c \quad (5.18)$$

We assume each beam is Gaussian pulse with the pulse width τ , then we can express the normalized electric field amplitude

$$E_j(t) = \frac{1}{\sqrt{2\pi}\tau} e^{-\frac{t^2}{2\tau^2}} \quad (5.19)$$

The complex amplitude of polarization density at the same frequency ω and wavevector $\vec{k}_4 = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$ is given by

$$P^{(3)}(\omega, \vec{k}_4) = \frac{3}{2} \varepsilon_0 \chi_{ijkl}^{(3)}(-\omega, -\omega, \omega, \omega, -\vec{k}_4, -\vec{k}_3, \vec{k}_2, \vec{k}_1) \varepsilon_3^*(-\omega, -\vec{k}_3) \varepsilon_2(\omega, \vec{k}_2) \varepsilon_1(\omega, \vec{k}_1) \quad (5.20)$$

Now the complex amplitude of instantaneous $\chi^{(3)}$ response is

$$P^{(3)}(\vec{k}_4, t) = \frac{3}{2} \varepsilon_0 \chi_{ijkl}^{(3)}(-\vec{k}_4, -\vec{k}_3, \vec{k}_2, \vec{k}_1) \varepsilon_3^*(-\vec{k}_3, t) \varepsilon_2(\vec{k}_2, t) \varepsilon_1(\vec{k}_1, t) \quad (5.21)$$

With a low-efficiency limit, where the generated wave always remains much less intense than the other waves, and therefore it is always in the non-depleted pump limit, and the coupled-wave equations (1.4-1.7) become just one equation. We can find the amplitude of the instantaneous DFWM signal A_4 is proportional to the polarization $|P^{(3)}|$. If beam 1 is delayed, we add the delay parameter t_d , which makes $\varepsilon_1(\vec{k}_1, t - t_d)$. Then we integrate it over time and get the energy of the instantaneous signal beam:

$$\begin{aligned} Energy &= \int_{-\infty}^{\infty} I_4(t) dt \propto \int_{-\infty}^{\infty} |P^{(3)}(\vec{r}, t)|^2 dt \\ &= \int_{-\infty}^{\infty} \left| \frac{3}{2} \varepsilon_0 \chi^{(3)} \varepsilon_3^*(-\vec{k}_3, t) \varepsilon_2(\vec{k}_2, t) \varepsilon_1(\vec{k}_1, t - t_d) \right|^2 dt \end{aligned} \quad (5.22)$$

As we assume Gaussian profile for each beam and we can analytically solve for the instantaneous signal:

```
DFWMSigAmp[tI_, tau_] = Integrate[PulseAmp[t, tau]^2 * NormPulseAmp[t - tI, tau], {t, -Infinity, Infinity}, Assumptions -> {tau > 0}]
```

$$\frac{e^{-\frac{tI^2}{3\tau^2}}}{\sqrt{3}}$$

```
DFWMSig[t_, tau_] = DFWMSigAmp[t, tau]^2
```

$$\frac{1}{3} e^{-\frac{2t^2}{3\tau^2}}$$

If we define a new parameter $A = \frac{3}{2} \varepsilon_0 \chi^{(3)}$ and replace it in Equation (5.1), we can numerically solve for the energy of the instantaneous signal beam, shown in Figure 5.1. Zero delay time presents that three pulses arrive to the nonlinear medium at the same time, corresponding to the peak of the instantaneous DFWM signal.

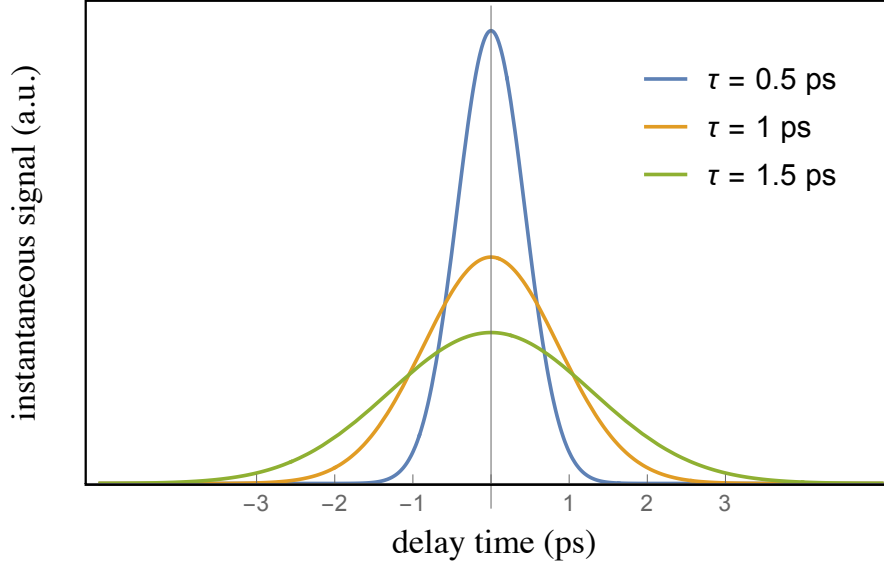


Figure 5.1: Instantaneous signal as a function of delay time for different pulse width τ with the same value of A, obtained by solving Equation (5.1) numerically.

5.1.3 Analysis of population gratings

The normalized intensity of a Gaussian pulse can be expressed as

$$I = \frac{e^{-\frac{t^2}{\tau^2}}}{\sqrt{\pi\tau}} \quad (5.23)$$

By plugging into Equation (5.2), we can solve for the amplitude of the transient grating as a function of time numerically in Figure 5.2. The intensity creates the excited states, which forms the population grating. As time goes on, the population grating decays exponentially.

For population grating formed by DFWM signal, there are several gratings that make contributions. Let's first look at the grating created by beam 2 and beam 3. In Figure 5.3, the boxcars geometry shows the population grating is formed by beam $\vec{k}_2$ and $\vec{k}_3$, then beam $\vec{k}_1$ is diffracted by the grating to generate the signal

```
s = DSolve[{y'[x] == R Normpulse[x, tau] - y[x]/T}, y[x], x]
```

$$\left\{ \left\{ y[x] \rightarrow e^{-\frac{x}{T}} C[1] - \frac{1}{2} e^{\frac{\tau^2}{4T^2} - \frac{x}{T}} R \operatorname{Erf}\left[\frac{\tau}{2T} - \frac{x}{\tau}\right] \right\} \right\}$$

```
gratingAmplitude[x_, tau_, T_, R_] = Simplify[y[x] /. s[[1]] /. C[1] -> \frac{1}{2} e^{\frac{\tau^2}{4T^2}} R]
```

$$-\frac{1}{2} e^{\frac{\tau^2 - 4Tx}{4T^2}} R \left(-1 + \operatorname{Erf}\left[\frac{\tau}{2T} - \frac{x}{\tau}\right] \right)$$

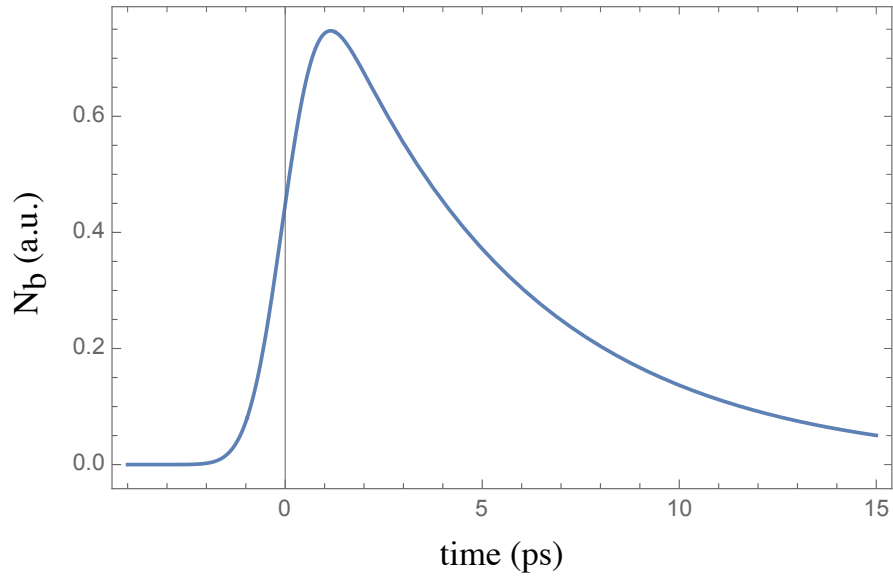


Figure 5.2: The amplitude of population grating as a function of time with the pulse width $\tau = 1$ ps, the lifetime $T = 5$ ps, obtained by solving Equation (5.9) numerically.

beam $\vec{k}_4$. The electric field amplitude of each beam is from Equation (5.18). Then,

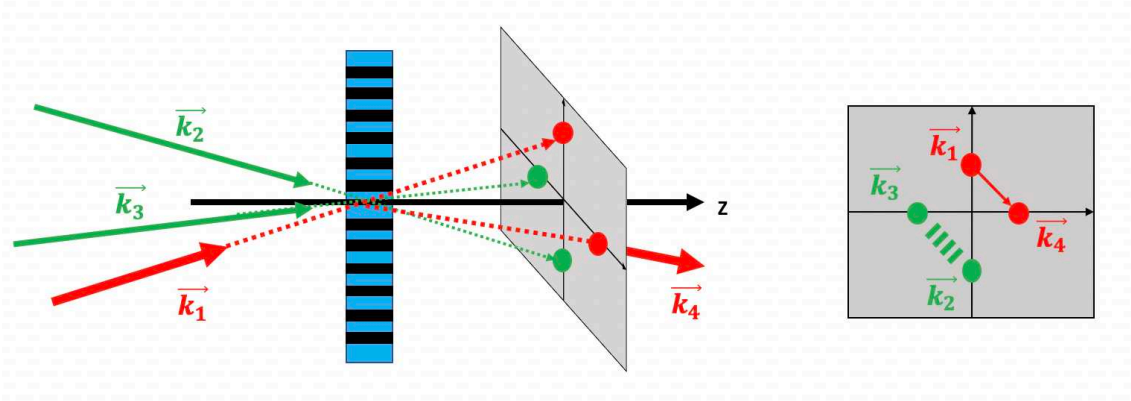


Figure 5.3: Population grating configuration. The incident beams 2 and 3 creates a grating while the arrival of beam 1 is Bragg diffracted signal. The green dashes show the diffraction grating and the arrow shows the diffraction of $\vec{k}_1$ to generate the signal beam $\vec{k}_4$.

we can derive $I(\vec{r}, t)$ from Equation (5.16)

$$\begin{aligned} I(\vec{r}, t) &= |\varepsilon_2 + \varepsilon_3|^2 \\ &= |E_2|^2 + |E_3|^2 + \frac{1}{2} E_2 E_3^* e^{i(\vec{k}_2 - \vec{k}_3) \cdot \vec{z}} + c.c \end{aligned} \quad (5.24)$$

Here, we only keep the term with $E_2 E_3^*$, which is the population grating that makes contribution to DFWM signal. We can re-write Equation (5.16):

$$\begin{aligned} \vec{P}_{n1}(t) &= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \cdot \vec{E} e^{-\frac{t}{T}} \int_{-\infty}^t \delta N I(\vec{r}, t') e^{\frac{t'}{T}} dt' \\ &= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T}} e^{i\vec{k}_1 \cdot \vec{z}} E_1 \cdot \int_{-\infty}^t E_2 E_3^* e^{i(\vec{k}_2 - \vec{k}_3) \cdot \vec{z}} e^{\frac{t'}{T}} dt' \\ &= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i(\vec{k}_1 + \vec{k}_2 - \vec{k}_3) \cdot \vec{z}} E_1 \cdot \int_{-\infty}^t E_2 E_3^* e^{\frac{t'}{T}} dt' \end{aligned} \quad (5.25)$$

Replacing $E_2(t)$ and $E_3^*(t)$ with Gaussian pulses and $E_1(t)$ with delayed Gaussian pulse, we can solve for the amplitude of the population grating 1, which is proportional to the polarization $|\vec{P}_{n1}(t)|$. The phase-matching condition is also achieved in this situation, where $\vec{k}_4 = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$. And the energy of the DFWM signal,

$$\begin{aligned}
\text{sl} &= \text{DSolve}[\{y1'[x] == R \text{Normpulse1}[x, \text{tau}] - y1[x]/T\}, y1[x], x] \\
&\left\{ \left\{ y1[x] \rightarrow e^{-\frac{x}{T}} C[1] - \frac{1}{2} e^{\frac{\text{tau}^2}{4T^2} - \frac{x}{T}} R \text{Erf}\left[\frac{\text{tau}}{2T} - \frac{x}{\text{tau}}\right] \right\} \right\} \\
\text{gratingAmplitute1}[x_, \text{tau}_-, T_-, R_-] &= \text{Simplify}[y1[x] /. \text{sl}[[1]] /. C[1] \rightarrow \frac{1}{2} e^{\frac{\text{tau}^2}{4T^2}} R] \\
&= \frac{1}{2} e^{\frac{\text{tau}^2 - 4Tx}{4T^2}} R \left(-1 + \text{Erf}\left[\frac{\text{tau}}{2T} - \frac{x}{\text{tau}}\right] \right)
\end{aligned}$$

contributed to the population grating 1, can be expressed in a similar way:

$$\begin{aligned}
\text{Energy} &= \int_{-\infty}^{\infty} |P^{(3)}(\vec{r}, t)|^2 dt \\
&= \int_{-\infty}^{\infty} \left| \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i\vec{k}_4 \cdot \vec{z}} E_1(t - t_d) \cdot \int_{-\infty}^t E_2(t') E_3^*(t') e^{\frac{t'}{T}} dt' \right|^2 dt
\end{aligned} \tag{5.26}$$

Figure 5.4 shows the DFWM signal formed by the population grating 1 as a function of delay time. We can see at 2 ps, the DFWM signal reaches its climax, representing the population grating is fully constructed. Then it follows an exponential decay as the transient grating disappears.

Similarly, we do the exact same calculation for $P_{n2}^{\vec{r}}(t)$, where the population grating 2 is created by beam 1 and beam 3. In Figure 5.5, the boxcars geometry shows the population grating is formed by beam $\vec{k}_1$ and $\vec{k}_3$, then beam $\vec{k}_2$ is diffracted by the grating to generate the signal beam $\vec{k}_4$. The difference between the population grating 1 and 2 is that the delayed beam is involved in forming the transient grating for the latter.

The intensity is derived in the same way,

$$\begin{aligned}
I(\vec{r}, t) &= |\varepsilon_1 + \varepsilon_3|^2 \\
&= |E_1|^2 + |E_3|^2 + \frac{1}{2} E_1 E_3^* e^{i(\vec{k}_1 - \vec{k}_3) \cdot \vec{z}} + c.c
\end{aligned} \tag{5.27}$$

Here, we only keep the term with $E_1 E_3^*$, which is the population grating that makes

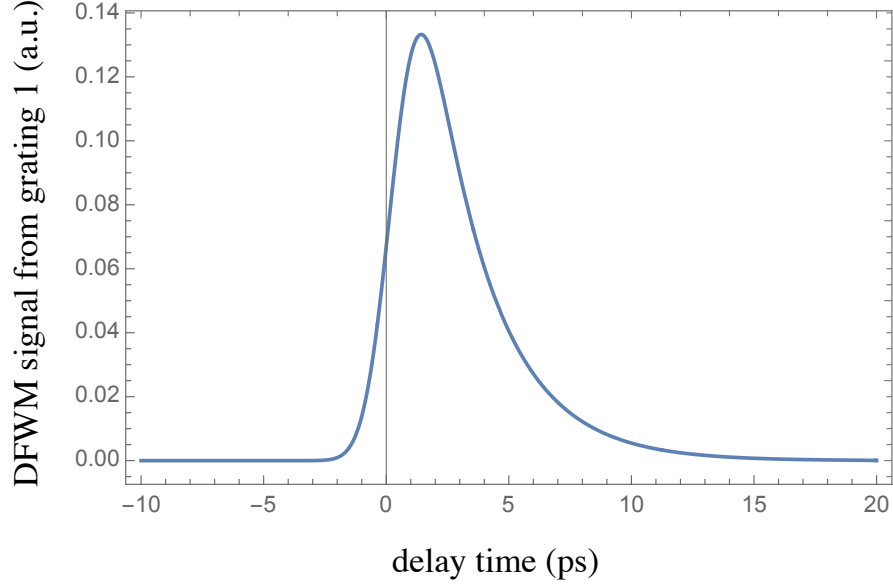


Figure 5.4: The DFWM signal formed from population grating 1 as a function of delay time with the pulse width $\tau = 1$ ps, the lifetime $T = 5$ ps, obtained by solving Equation (5.26) numerically.

contribution to DFWM signal. We can re-write the second term in Equation (5.16),

$$\begin{aligned}
\vec{P}_{n2}(t) &= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \cdot \vec{E} e^{-\frac{t}{T}} \int_{-\infty}^t \delta N I(\vec{r}, t') e^{\frac{t'}{T}} dt' \\
&= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T}} e^{i\vec{k}_2 \cdot \vec{z}} E_2 \cdot \int_{-\infty}^t E_1 E_3^* e^{i(\vec{k}_1 - \vec{k}_3) \cdot \vec{z}} e^{\frac{t'}{T}} dt' \quad (5.28) \\
&= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i(\vec{k}_1 + \vec{k}_2 - \vec{k}_3) \cdot \vec{z}} E_2 \cdot \int_{-\infty}^t E_1 E_3^* e^{\frac{t'}{T}} dt'
\end{aligned}$$

Replacing $E_2(t)$ and $E_3^*(t)$ with Gaussian pulses and $E_1(t)$ with delayed Gaussian pulse, we can solve for the amplitude of the population grating 2, which is proportional to the polarization $|\vec{P}_{n2}(t)|$. And the energy of the DFWM signal, contributed

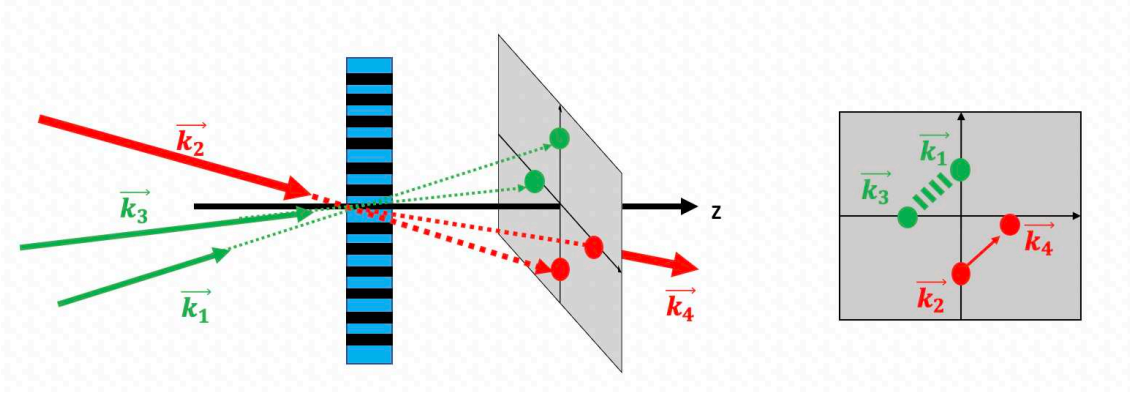


Figure 5.5: Population grating configuration. The incident beams 1 and 3 creates a grating while the arrival of beam 1 is Bragg diffracted signal. The green dashes show the diffraction grating and the arrow shows the diffraction of $\vec{k}_2$ to generate the signal beam $\vec{k}_4$.

$$\begin{aligned}
 s2 &= \text{DSolve}[\{y2'[x] = \text{RMormpulse2}[x, t1, \text{tau}] - y2[x]/T\}, y2[x], x] \\
 &\left\{ \left\{ y2[x] \rightarrow e^{-\frac{x}{T}} C[1] - \frac{e^{-\frac{T^2 t1^2 + 2 T t1 \text{tau}^2 + \text{tau}^4}{4 T^2 \text{tau}^2} - \frac{x}{T}} R \text{Erf}\left[\frac{T t1 + \text{tau}^2 - 2 T x}{2 T \text{tau}}\right]}{4 \sqrt{\pi} \text{tau}} \right\} \right\} \\
 \text{gratingAmplitude2}[x_, t1_, \text{tau}_-, T_, R_] &= \text{Simplify}\left[y2[x] /. s2[[1]] /. C[1] \rightarrow e^{\frac{x}{T}} e^{-\frac{T^2 t1^2 + 2 T t1 \text{tau}^2 + \text{tau}^4}{4 T^2 \text{tau}^2} - \frac{x}{T}} R \right] \\
 &= \frac{e^{-\frac{T^2 t1^2 + \text{tau}^4 + 2 T \text{tau}^2 (t1 - 2 x)}{4 T^2 \text{tau}^2}} R \left(-1 + \text{Erf}\left[\frac{T t1 + \text{tau}^2 - 2 T x}{2 T \text{tau}}\right] \right)}{4 \sqrt{\pi} \text{tau}}
 \end{aligned}$$

to the population grating 2, can be expressed in a similar way:

$$\begin{aligned}
 \text{Energy} &= \int_{-\infty}^{\infty} |P^{(3)}(\vec{r}, t)|^2 dt \\
 &= \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i \vec{k}_4 \cdot \vec{z}} E_2(t) \cdot \int_{-\infty}^t E_1(t' - t_d) E_3^*(t') e^{\frac{t'}{T}} dt'
 \end{aligned} \tag{5.29}$$

Figure 5.6 shows the DFWM signal formed by the population grating 2 as a function of delay time. However, it is quite different from the case of grating 1. First

of all, at -1 ps, the DFWM signal reaches its climax, representing the population grating is fully constructed. But the signal decay so fast and return to zero. The formation of the population grating 2 is still incomplete because of the influence of the delayed pulse 1. The delayed time would accelerate the destruction of the population grating 2.

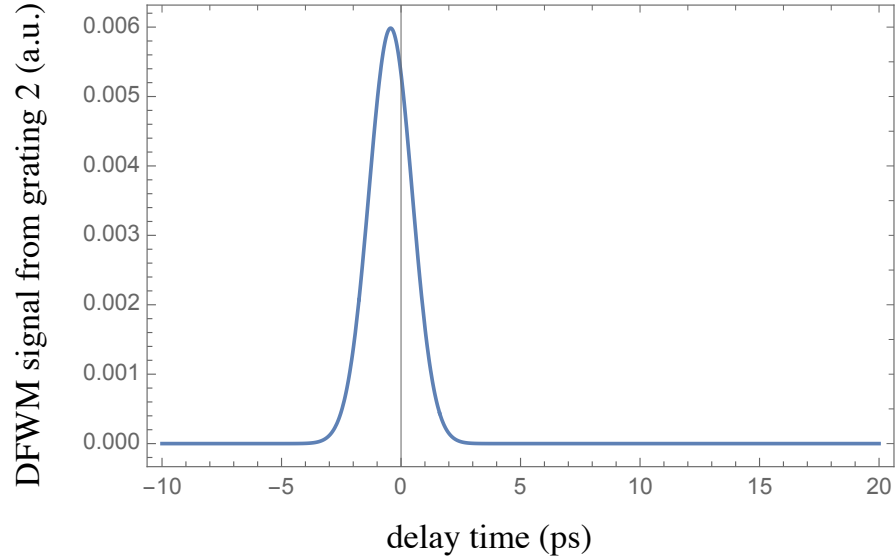


Figure 5.6: The DFWM signal formed from population grating 1 as a function of delay time with the pulse width $\tau = 1$ ps, the lifetime $T = 5$ ps, obtained by solving Equation (5.29) numerically.

Finally, we look at the population grating formed by beam 1 and beam 2. In Figure 5.7, the boxcars geometry shows the population grating is formed by beam $\vec{k}_1$ and $\vec{k}_2$, then beam $\vec{k}_3$ is diffracted by the grating to generate the signal beam $\vec{k}_4$. However, $\vec{k}_4$ isn't pointing to the original position as above. Instead, it would point downward, because it is Bragg diffracted by the new grating in this situation. Therefore, this grating won't make any contribution to the DFWM signal as we discussed above.

Now we put together two types of population gratings that contribute to the

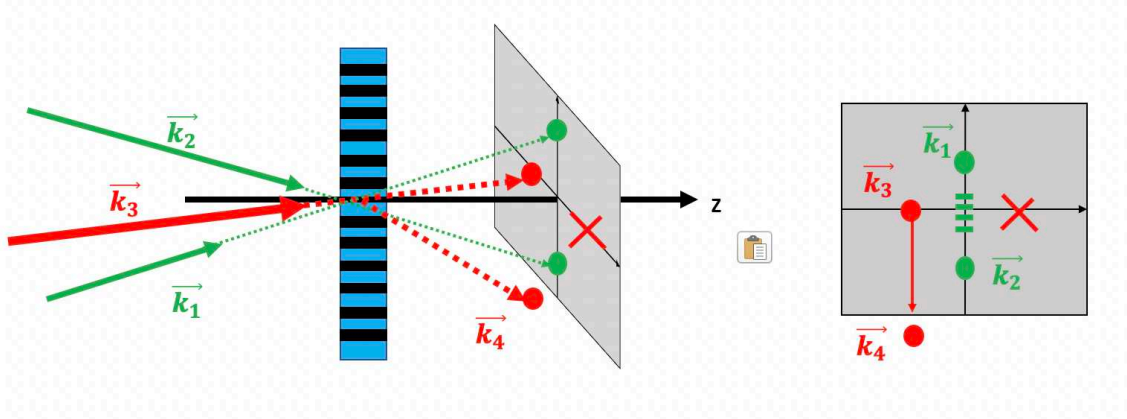


Figure 5.7: Population grating configuration. The incident beams 1 and 2 creates a grating while the arrival of beam 3 is Bragg diffracted signal. The green dashes show the diffraction grating and the arrow shows the diffraction of $\vec{k}_1$ to generate the signal beam $\vec{k}_4$ downside, which is not at the same position previously.

total polarization of the DFWM signal:

$$\begin{aligned} \vec{P}(\vec{r}, t) = & \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i\vec{k}_4 \cdot \vec{z}} E_1(t - t_d) \cdot \int_{-\infty}^t E_2(t') E_3^*(t') e^{\frac{t'}{T}} dt' \\ & + \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i\vec{k}_4 \cdot \vec{z}} E_2(t) \cdot \int_{-\infty}^t E_1(t' - t_d) E_3^*(t') e^{\frac{t'}{T}} dt' \end{aligned} \quad (5.30)$$

5.1.4 Overall analysis

The DFWM signal consists of two components: instantaneous $\chi^{(3)}$ response and population grating response. And the energy can be finally written as

$$\begin{aligned}
\int_{-\infty}^{\infty} I_4(t) dt &\propto \int_{-\infty}^{\infty} |P^{(3)}(\vec{r}, t)|^2 dt \\
&= \int_{-\infty}^{\infty} \left| \frac{3}{2} \epsilon_0 \chi^{(3)} \epsilon_3(-\vec{k}_3, t) \epsilon_2(\vec{k}_2, t) \epsilon_1(\vec{k}_1, t - t_d) \right. \\
&\quad + \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i \vec{k}_4 \cdot \vec{z}} E_1(t - t_d) \cdot \int_{-\infty}^t E_2(t') E_3^*(t') e^{\frac{t'}{T}} dt' \\
&\quad \left. + \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N e^{-\frac{t}{T} + i \vec{k}_4 \cdot \vec{z}} E_2(t) \cdot \int_{-\infty}^t E_1(t' - t_d) E_3^*(t') e^{\frac{t'}{T}} dt' \right|^2 dt
\end{aligned} \tag{5.31}$$

Here, We introduce other parameters without time-dependence:

$$B = \frac{1}{2} \epsilon_0 f(\alpha_b - \alpha_a) \delta N \tag{5.32}$$

Then we define the ratio:

$$r = \left| \frac{B}{A} \right| = \frac{|f(\alpha_b - \alpha_a) \delta N|}{3|\chi^{(3)}|} \tag{5.33}$$

where $A = \frac{3}{2} \epsilon_0 \chi^{(3)}$, and the phase difference φ which represents the interference between the instantaneous $\chi^{(3)}$ response and the non-instantaneous gratings. Then the relationship between A and B is

$$B = A \cdot r \cdot e^{i\varphi} \tag{5.34}$$

The ratio depends on both first and third order polarizability of ground and excited states, the refractive index and total density of molecules [16]. Other parameters that affect the energy of the DFWM signal are the pulse width τ and excited-state lifetime T . In the next section, we will discuss the influence of DFWM signal with various change of those parameters.

To study the dynamic characteristics of the contributions from different terms in Equation (5.31), we conducted theoretical simulations using the parameters: $\tau =$

0.5 ps, $B = 1$, $T = 10$ ps, $\varphi = 0$. With those parameters, the results obtained to represent the influence of r are shown in Figure 5.8. The blue curve represents the total DFWM signal intensity profile, the green and red curves are contributions from two population grating processes, and the yellow curve represents the coherent four-wave mixing process. As we can see, for coherent four-wave mixing process, the maximum intensity occurs at delay time $t_d = 0$. However, for the population grating contribution, their maximum intensity is shifted away from $t_d = 0$. As r becomes larger, which means population gratings contribute more, the maximum of total DFWM signal is shifted further.

Two types of population grating are formed in different situations as we mentioned previously. The main difference is that the delay time is involved in the formation of grating for the second grating, as we can see from Figure 5.5. The contribution of the first grating is always prominent, and that is because the delay time doesn't play a role in the formation of grating from beam 2 and 3, as we can see from Figure 5.3. As r becomes smaller, especially when $r < 1$, the contribution of the second grating is depressed drastically. That is to say, when $r < 1$ ($|f(\alpha_b - \alpha_a)\delta N| < 3|\chi^{(3)}|$), the second grating can be neglected.

Figure 5.9 shows the results for different lifetime T under the condition of $\tau = 0.5$ ps, $B = 1$, $r = 1$, $\varphi = 0$. When the pulse width of the incident beam (τ) is much shorter than the lifetime of the molecular excited-state (T), the population grating decays approximately $e^{1/T}$. In addition, the total signal intensity decays at a fast speed for a short delay time, but at a slow speed for long delay time. Therefore, the contributions from coherent four-wave mixing and population grating process must be separated, because the decay time of coherent DFWM signal is independent of the lifetime T . Our approximation can help measure the lifetime of the excited state of the molecules more precisely.

Figure 5.10 shows the results for different values of φ under the condition of $\tau = 0.5$ ps, $T = 10$ ps, $B = 1$, $r = 0.2$. The phase difference φ represents the interference effect between coherent four-wave mixing and population grating process. The interference varies from constructively ($\varphi = 0$) to destructively ($\varphi = \pi$). We observe that the total DFWM signal is fully influenced without phase difference and

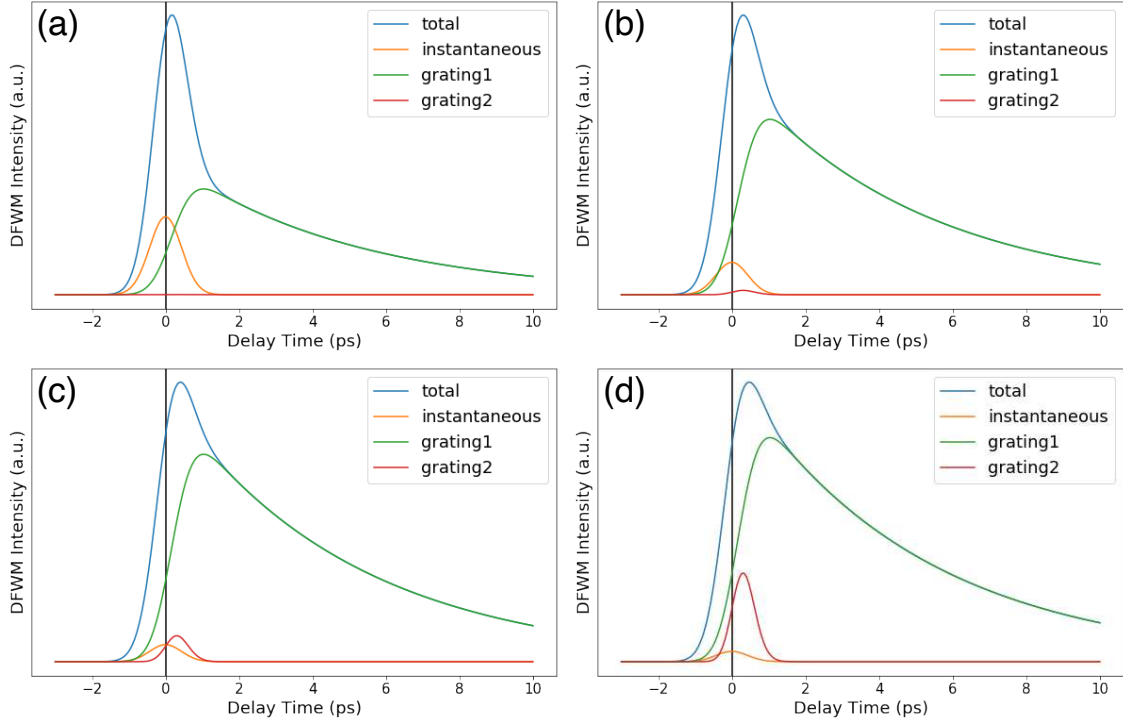


Figure 5.8: Calculated DFWM decay patterns for $\tau = 0.5$ ps, $T = 10$ ps, and $\varphi = 0$. The blue curve represents the total DFWM signal, the green and red curves are contributions from two population grating processes, and the yellow curve represents the coherent four-wave mixing process. Different values for r are simulated for each graph, respectively, (a). $r = 0.5$, (b). $r = 1$, (c). $r = 1.5$ and (d). $r = 2$.

$\pi/2$ phase difference from Figure 5.10 (a) and (b). What's worse, the contribution of coherent four-wave mixing is remarkably suppressed by destructive interference with population gratings, as we can see an obvious dip appears at 1 ps delay time in Figure 5.10 (c). Therefore, the phase difference φ may affect the maximum intensity of DFWM signal. Besides, the influence of interference should also be considered into calculating the lifetime of molecular excited-state using the method of DFWM.

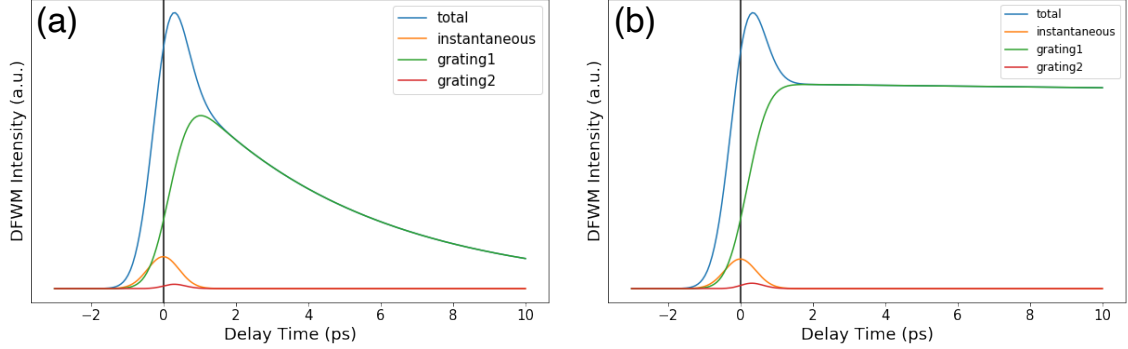


Figure 5.9: Calculated DFWM decay patterns based on (a). $T = 10$ ps and (b). $T = 1000$ ps. Each curve is defined the same as for Figure 5.8.

5.2 Experimental Setup

Figure 5.11 shows a schematic of Degenerate Four-wave Mixing (DFWM) experimental setup. The method of data collection in this experiment was obtained by a previous student. A Clark-MXR CPA series 2101 laser at a repetition rate of 1 kHz produces 1-ps pulses at 773 nm. The pulse is sent into a Light Conversion TOPAS (Traveling-Wave Optical Parametric Amplifier System) device. The TOPAS emits wavelength tunable pulses by nonlinear optical parametric generation to output a chosen wavelength. The TOPAS device allowed for the variation of the wavelength of the incident light beams from 400 nm to 1800 nm. Due to the possibility of several nonlinear optical process, the proper wavelength and filters must be used for each experiment.

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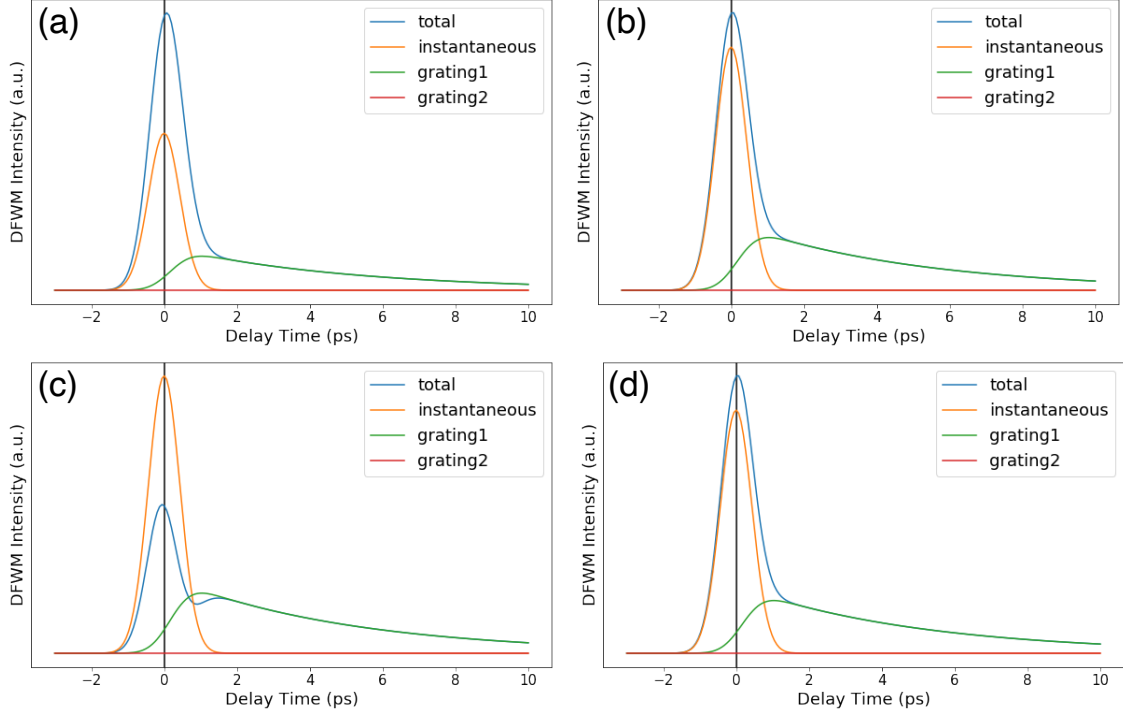


Figure 5.10: Calculated DFWM decay patterns for different values of (a). $\varphi = 0$, (b). $\varphi = \pi/2$, (c). $\varphi = \pi$, and (d). $\varphi = 3\pi/2$. Each curve is defined the same as for Figure 5.8.

of the incident light beams from 400 nm to 1800 nm. Due to the possibility of several nonlinear optical process, the proper wavelength and filters must be used for each experiment.

The beam out out TOPAS is then split into three beams, and two of them are sent into retroreflectors which are mounted on two delay tracks. Delay Track 1 delays the probe beam, relative to the two pump beams, while Delay Track 2 is used to adjust the arrival time of the two pump beams. Both delay tracks are model TLSR300B from Zaber Technologies, controlled by translation stages with a python program. All three beams with identical frequency are recombined simultaneously into the sample, which spatial configuration of pump-and-probe was shown in Figure

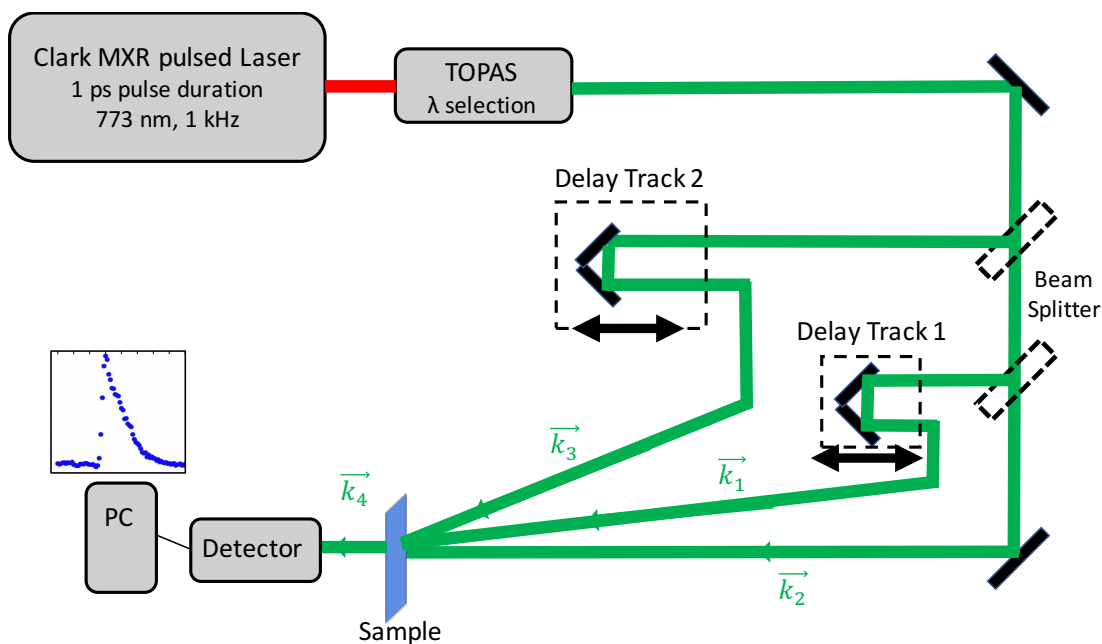


Figure 5.11: A schematic of DFWM experiment: a pump laser beam was passed through a TOPAS light-conversion system to tune the frequency of the incident beams. The initial beam was then split into three beams, two of which were delayed to adjust the arrival time, and then all three beams were recombined simultaneously into the sample. All beams are of identical frequency. The generated DFWM signal was collected by a photodetector after the sample.

5.3. When beam 1 is delayed with respect to the others, the DFWM signal can be collected by a photodetector after the sample, which can be used to observe the dynamics and calculate the lifetime of excited states.

5.3 Nonlinear Optical Organic Molecules

The majority of this work centers on the study of a group of cyanoethynylethene molecules both in the molecular and supramolecular state. It is part of a larger family of 1,1,4,4-tetracyanobuta-1,3-diene (TCBD) based molecules [54, 55] known as donor-acceptor substituted cyanoethynylethene molecules. These molecules were synthesized and their chemical and structural properties studied by the Diederich group at ETH Zurich, the details of which are covered in [56]. Here we present the molecular properties of these molecules relevant to this work.

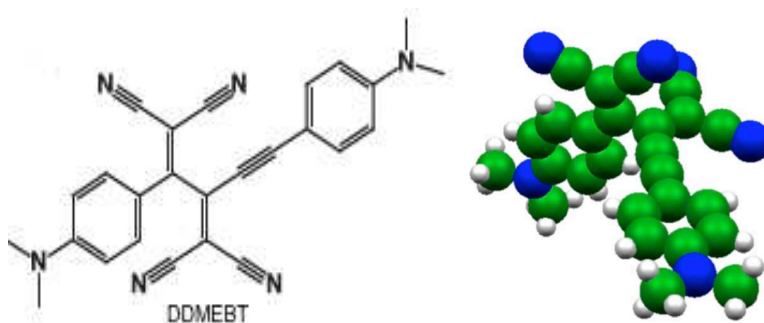


Figure 5.12: Diagram of the DDMEBT molecule.

The molecule 2-[4(dimethylamino)phenyl]- 3-([4(dimethylamino)phenyl]-ethynyl)buta-1,3-diene-1,1,4,4-tetracarbonitrile), informally known as DDMEBT, which is shown in Figure 5.12. DDMEBT has been shown to have a very large off-resonant third-order nonlinear polarizability relative to its size [57, 58]. In addition to its important molecular properties, it has also been shown that DDMEBT produces supramolecular thin films of very high optical quality [59]. The non-planar structure is actually key to reducing intermolecular interactions and producing a high-optical quality homogenous film.

DDMEBT under this study were dissolved in dichloromethane (DCM) as solvent inside 1-mm thick fused silica spectroscopy cells at concentrations varying between 0% and 2 % mass ratio. From the absorption spectrum of DDMEBT in Figure 5.13, the first absorption peak is centered at 527 nm [60]. In the DFWM experiment, we investigated the DFWM signal with the wavelength from resonant (at 530 nm) to off-resonant (at 700 nm). More details will be presented in the following section.

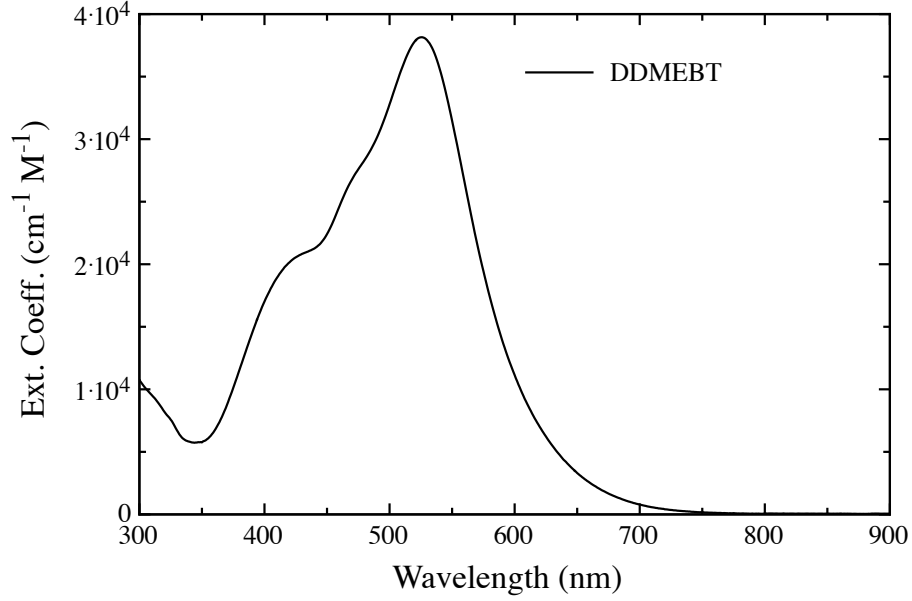


Figure 5.13: Spectrum of the linear molar extinction coefficient, in units of $\text{cm}^{-1} \text{M}^{-1}$, of DDMEBT molecules.

5.4 Experimental study of short-lived population gratings

The DFWM measurements studied in this work were performed by a previous student. We set the wavelength of the laser pulses via computer control of the TOPAS without any further adjustment of the experimental setup. The data collection of the DFWM signal were conducted by the scans of Delay Track 1 (in Figure 5.11) over a chosen time intervals with a python program in the computer. In this experiment, we input a scan over a -3 ps to 9 ps time interval, moving in 0.1 ps step. The DFWM signal is detected by a photodiode. The dependence of the DFWM signal as a function of delay time was also studied at both the signal peak and within the pure population grating contribution region. All three beams arrived at the same time when the delay time is zero, and the DFWM signal was at maximum due to

the instantaneous $\chi^{(3)}$ effect.

As discussed in the theoretical section, a resonant DFWM process involve two dynamic processes, coherent four-wave mixing and population grating. The curve-fitting method were used via the analytic calculation described by Equation 5.31. The ratio r between B and A can determine which process is dominant. Since both parameters A and B are complex, the phase information, as well as the interference, must be considered in order to precisely fit the data. Here, We replaced the pulse width τ with Full Width at Half Maximum for the pump and probe pulses using the equation $FWHM = 2\tau\sqrt{\ln 2}$. With experimental results, we can tell how well the theoretical model fits the actual dynamics. Figure 5.14 shows the temporal

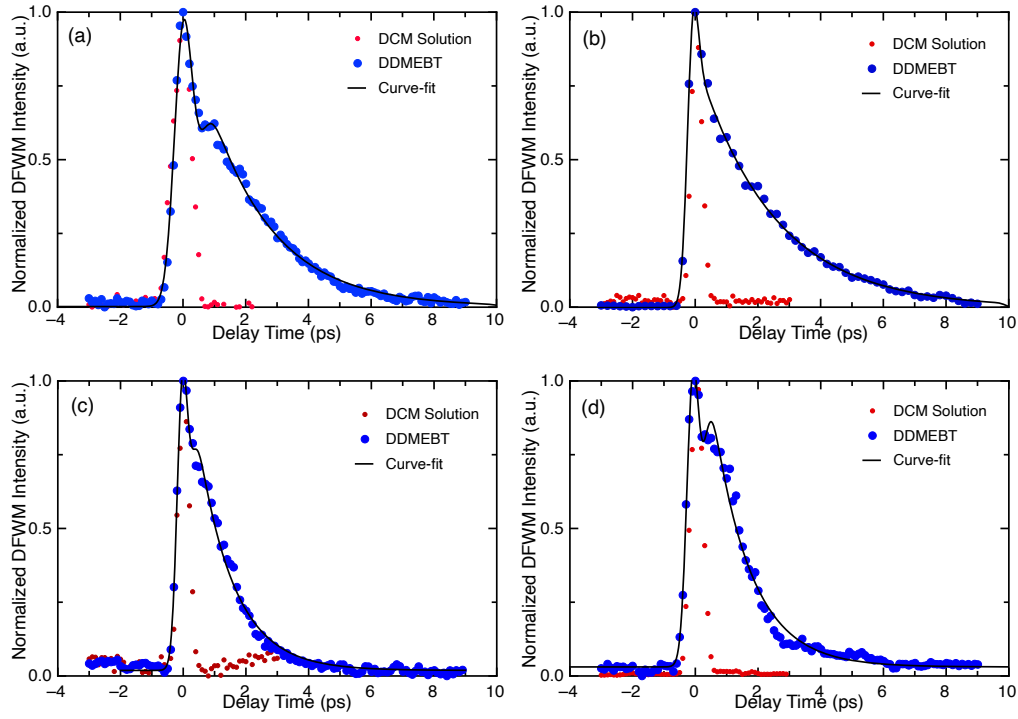


Figure 5.14: Decay of normalized DFWM signal from DCM solution (red scattered dots), DDMEBT solution (blue scattered dots), and the theoretical simulation curve (solid curve) at different wavelengths: (a). $\lambda = 530 \text{ nm}$, (b). $\lambda = 600 \text{ nm}$, (c). $\lambda = 690 \text{ nm}$ and (d). $\lambda = 720 \text{ nm}$.

Wavelength (nm)	FWHM (ps)	T (ps)	r	φ
530	0.57 ± 0.02	4.1 ± 0.5	0.28 ± 0.02	$149^\circ \pm 5^\circ$
600	0.33 ± 0.01	4.9 ± 0.4	0.46 ± 0.03	$247^\circ \pm 4^\circ$
690	0.33 ± 0.01	2.3 ± 0.5	0.30 ± 0.02	$146^\circ \pm 4^\circ$
720	0.38 ± 0.03	2.4 ± 0.6	0.29 ± 0.02	$160^\circ \pm 5^\circ$

Table 5.1: The results of each parameter from Figure 5.14 by curve-fitting based on theoretical simulation.

behaviors of DCM (solvent) and DDMEBT solution under such conditions. The solid curve is the theoretical fit obtained using Equation 5.31. It can be seen that the experimentally obtained curve of DDMEBT consists of three regions. A fast decay region is the contribution from the coherent four-wave mixing process. The slow decay region is from population grating diffraction. Additionally, there is a third region that is in between due to the existence of phase information φ . We can see a clear dip within 1 ps for each graph.

The fitting results for each graph are listed in Table 5.1 below. FWHM is measured from the DFWM signal of DCM solution. To fit other parameters, we keep the same FWHM as obtained from the DFWM signal of DCM solution. When the wavelength is near one-photon absorption peak of DDMEBT, the lifetime of excited-state is measured of 4.1 ± 0.5 ps. Since r is smaller than 1, as discussed previously, the contribution from the second grating formed by beam 1 and 3 is negligible. Both FWHM and lifetimes decrease as the wavelength moves away from resonant. Therefore, the population density of the excited-state molecules may affect the results.

5.5 Summary and conclusions

We have theoretically derived an expression for the third-order polarization in a resonant medium for the case of degenerate four-wave mixing. The theoretical analysis

explicitly consider that DFWM in a resonant medium actually involves two mechanisms. One is the coherent four-wave mixing mechanism which is a third-order nonlinear $\chi^{(3)}$ process. The other is a population grating mechanism that involves the decay lifetime of the lowest excited state of the molecules. The measured DFWM signal as a function of decay time includes contributions from both processes.

The theoretical results also indicate that the dynamic behavior of coherent four-wave mixing is different from that of the population grating contribution. When one beam is delayed, the contribution of coherent four-wave mixing reaches a peak value at the zero point of delay time. The peak value for the population grating contribution is shifted to a longer delay time because of the time integration character of the population grating.

It is obvious that the information on the lifetime of the molecular excited state is included only in the decay region of population grating contribution to the total DFWM signal. We have experimentally investigated the dynamic characteristic of DDMEBT in DCM solution in DFWM and studied the decay regions. The coherent four-wave mixing and the population grating processes are suggested to be responsible for the fast decay and the slow decay regions, respectively. Our experimental results indicate that the excited-state molecules of DDMEBT in DCM solution decay predominantly by the transient gratings. The lifetimes of the lowest excited state of DDMEBT are measured 2-5 ps in this experiment, curve-fitted by the theoretical model. In summary, Our experimental results are satisfactorily explained by the theoretical derivation and simulation presented in this chapter.

Chapter 6

Numerical simulation of guided-wave propagation

Previously we provides examples and theoretical calculations that apply to step-index cylindrical waveguides, including single-mode fiber and dual-core fiber. We obtained the propagation constants in the lowest-frequency fundamental modes. we also rely on an approximate expression for the coupling constants between the supermodes in two parallel cores, as already discussed in chapter 2. On account of the approximations of this method, we need to confirm our results by using numerical simulations of guided-wave propagation. In this work, we choose to use a simulation software called COMSOL Multiphysics [29]. Section 5.1 introduces the basics and brief strategies of how to use COMSOL Multiphysics software .

Then, we produce supplemented results obtained in this way by obtaining the propagation constants of modes using COMSOL Multiphysics software. We verify the results of single-mode fiber in chapter 3 and dual-core fiber in chapter 4, then discuss the deviations from the theoretical method. We will also show the conditions and requirements when the results of the coupling constants are in agreement between the approximate expression and COMSOL. Furthermore, we will discuss the influence of the results of the CLPM rules from those two different methods. Plots and graphs will be presented to help clarify the deviations.

6.1 COMSOL Multiphysics method

COMSOL Multiphysics is a cross-platform finite element analysis, solver and multiphysics simulation software. It allows conventional physics-based user interfaces and coupled systems of partial differential equations (PDEs). We can create a cylindrical waveguide with inner core and cladding to simulate beam propagation in a step-index optical fiber. After we build the structure of the cylindrical waveguide, we can assign the refractive index of cladding with Sellmeier equation and specify the refractive index contrast for the core. Light that propagates through a dielectric waveguide has most of the power concentrated within the central core of the waveguide. Outside the waveguide core, in the cladding, the electric field decays exponentially with the distance from the core. Mode analysis can help us obtain the propagation constant of the fundamental mode at a specific wavelength, and we can observe the mode profile at the same time.

To simulate beam propagation in a dual-core fiber, we need to create two cylindrical waveguides side by side. If two waveguide cores are close to each other, the second waveguide perturbs the mode of the first waveguide (and vice versa). Thus, instead of having two modes with the same effective index, one localized in the first waveguide and the second mode in the second waveguide, the modes and their respective effective indices split and you get a symmetric supermode with an effective index that is slightly larger than the effective index of the unperturbed waveguide mode, and an antisymmetric supermode, with an effective index that is slightly lower than the effective index of the unperturbed waveguide mode. Mode analysis can also help us obtain the propagation constants of the lowest symmetric supermode (or even mode) and antisymmetric supermode (or odd mode).

Once we obtain the values of the propagation constants from COMSOL Multiphysics software, we can do the same calculations to achieve CLPM conditions as we showed in chapter 3 and chapter 4. Since the propagation constants are depending on the structure of the fiber (core radius and distance center to center), the working wavelength and materials of core and cladding, those parameters can be tuned arbitrarily inside COMSOL.

6.2 COMSOL simulation of single-core fiber

For the simulation of the single-core fiber, we use the dispersion relationship of silica and fluoride glasses as cladding, obtained from the standard Sellmeier expression in Table 3.1. And we fix the refractive index contrast $\Delta n = 0.01$. The core of the radius depends on the materials we use, which is the same as shown in Figure 3.4. Then we run the mode analysis in COMSOL, and obtain the propagation constant of the fundamental mode at each wavelength. Phase mismatch Δk is calculated for the process third-order frequency downconversion, similarly in chapter 3.

In figure 6.1, the data points represent the coupling constants obtained in COMSOL. We can see most data points are mostly well-aligned with the theoretical values derived in chapter 3. The only deviation is the point exactly after the degenerate position, where all wavelengths are equal at the beginning, which makes $\Delta k = 0$ automatically. For silica fiber ($r = 4.5 \mu\text{m}$), Δk is slightly larger than zero whereas the theoretical value is negative. And that is the same case for ZrF_4 fiber ($r = 9 \mu\text{m}$). Obviously, the numerical errors come up around the degenerate position in those cases. But it doesn't occur to the case for InF_3 fiber ($r = 9 \mu\text{m}$). Except that, we can regard the results from two methods are in agreement with each other.

We stop to confirm the results from COMSOL before the material gets too much absorbed. For instance, InF_3 glass would get too much absorbed if the wavelength is beyond $4 \mu\text{m}$, so we stop measuring Δk at that wavelength. Taking into consideration of absorption, we conclude that data points from COMSOL are matched with the theoretical values derived in chapter 3, especially at the positions when $\Delta k = 0$ for those materials. In another words, the generated wavelength corresponding to $\Delta k = 0$ can be extracted either way.

6.3 COMSOL simulation of dual-core fiber

For the simulation of the dual-core fiber, we use the dispersion relationship of silica and fluoride glasses as cladding, obtained from the standard Sellmeier expression in Table 3.1. And we fix the refractive index contrast $n = 0.01$ as well. In COMSOL, we

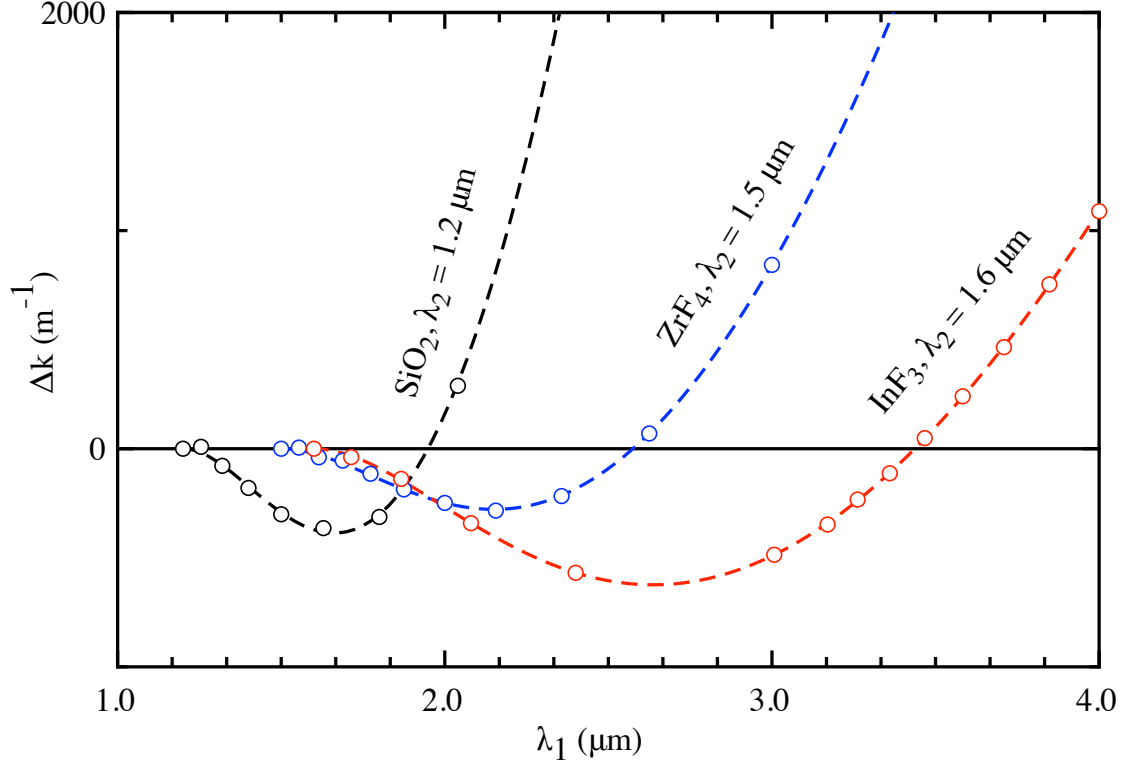


Figure 6.1: Phase mismatch factor Δk as a function of the generated wavelength λ_1 for different pump wavelength and glass types. The dashed curves are for single-mode step-index fiber with a radius of $4.5 \mu\text{m}$ (silica) and $9 \mu\text{m}$ (fluorides). The normalized propagation constants are obtained from Equations (2.26) and (2.24). The data points close to the curves represent the corresponding data obtained in COMSOL.

can tune the values of the core radius and the distance between the cores, then run the mode analysis simulation and obtain the propagation constants of two lowest-frequency supermodes. Similarly in chapter 4, we add the corresponding results obtained from COMSOL and access the accuracy of these calculations in Figure 4.2 4.3. Then we discuss the deviations from the results in chapter 4.

6.3.1 Confirmation of the results in chapter 4

The six expressions for the phase-mismatch terms ΔK_ν for frequency down-conversion in dual core fibers, expressed in terms of the single-mode propagation constant in each core and the corresponding coupling constants in Equations (4.4), or in terms of the two lowest-frequency fundamental modes in the two-core system in Equations (4.10-4.15), are shown for a dual-core step-index silica fiber in Figure 6.2 as a function of the generated wavelength λ_1 and the distance between the cores for different choices of core radius and pump wavelengths λ_2 .

The data points on top of some curves were obtained from COMSOL and Equations (4.10-4.15), to assess the accuracy of these calculations. We can see that most data points are well-matched with the curves when the distance between two cores are large enough. Since the coupling constants are obtained by two different methods, we verify that the results are accurate with each other when expression (2.26) is valid for larger values of wD/a (See Ref. [37], Chapter 18). Otherwise, the deviations come up and become larger for smaller distance. For instance, when the core radius is $3.5 \mu\text{m}$, the results are accurate with two methods when distance is larger than $10 \mu\text{m}$. Otherwise, expression (2.26) is not valid anymore, so we have to use the results obtained from COMSOL. In the following section, we will discuss other deviations since the coupling constants are obtained by two different methods.

6.3.2 Deviations from the results in chapter 4

To investigate the deviations, let's compare the corresponding results obtained from COMSOL with those in Figure 2.7. The insets shows the profiles of the two super-modes in the dual-core structure of this example, at a wavelength of $1.55 \mu\text{m}$. When the generated frequency is approaching towards the cut-off frequency for the odd mode, we find the deviations of the results of the normalized propagating constant from two methods are getting larger. In this example ($r = 2 \mu\text{m}$, $D = 7 \mu\text{m}$), the odd mode disappears when the cut-off wavelength is greater than $2 \mu\text{m}$, which leaves the even mode alone. The normalized propagation constants of the even mode, obtained from COMSOL, are also getting worse for smaller frequencies, compared with the

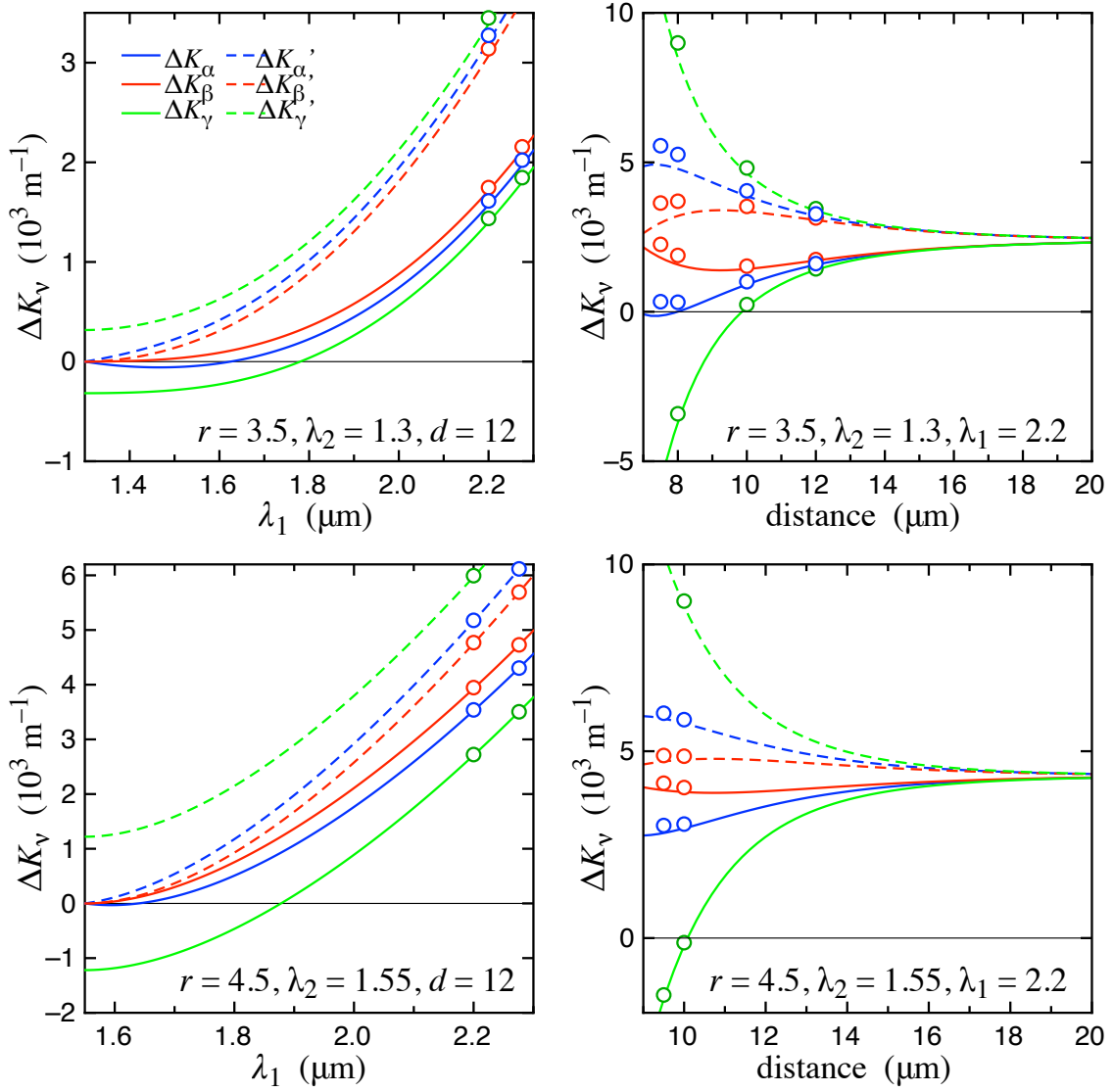


Figure 6.2: Similarly in Figure 4.2 and 4.3, phase mis-match terms ΔK_ν for a dual core step-index silica fiber with refractive index contrast of 0.01. The order of the curves, from top to bottom corresponds to $\Delta K'_\gamma$, $\Delta K'_\alpha$, $\Delta K'_\beta$, ΔK_β , ΔK_α , ΔK_γ . The data points on top of some curves were obtained from COMSOL and Equations (4.10-4.15), to assess the accuracy of these calculations.

curve obtained from Equation (2.26) in Figure 6.3. The odd mode disappears below a frequency of 150 THz where $wD/a = 0.1$, which means that there is no meaning

to the coupling constant anymore. Therefore, the generated frequency is limited to be larger than the value of the cut-off frequency for the odd mode. In CLPM configuration, the fact that the odd mode is required for phase matching also limits the generated wavelength to get too large for a specific designed structure of the dual-core fiber.

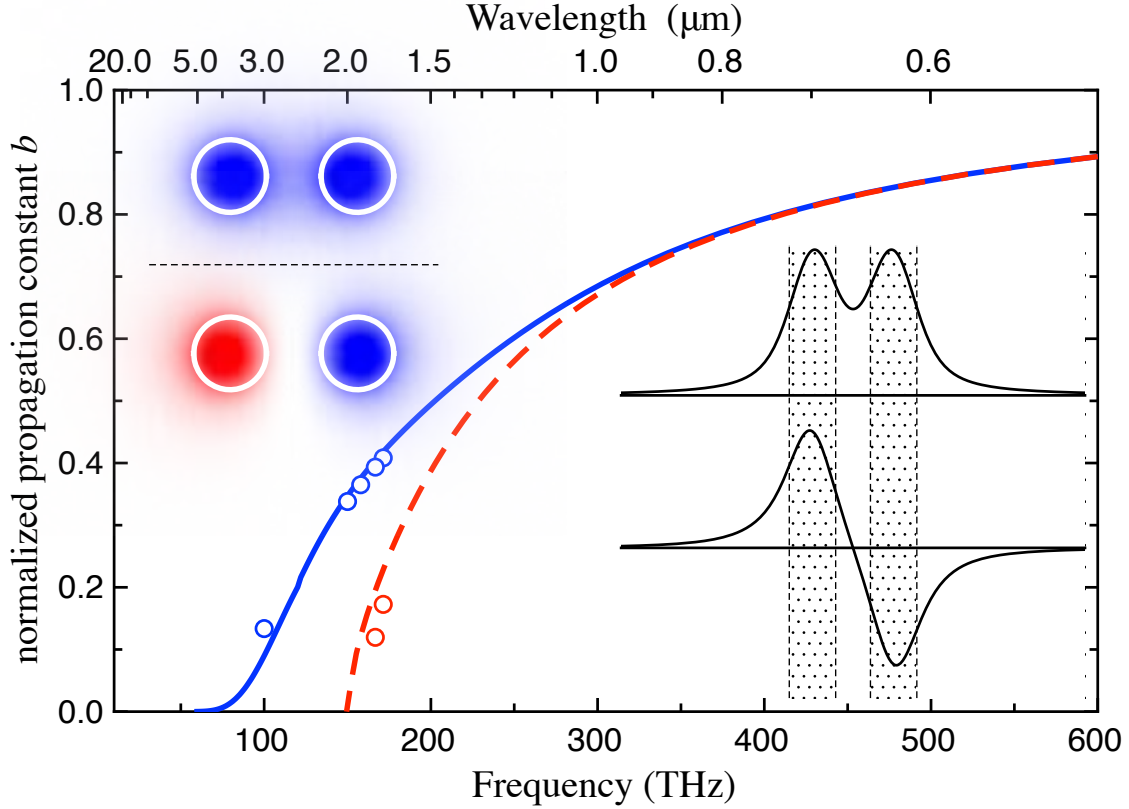


Figure 6.3: Dispersion of the even (solid curve) and odd (dashed curve) supermodes in a step-index silica fiber with a core radius of $2 \mu\text{m}$ a distance $D = 7 \mu\text{m}$ apart, and $\Delta n = 0.01$. The curves for the normalized propagation constants are obtained from Equations (2.26) and (2.24). The data points close to the curves represent the corresponding data obtained in COMSOL. The insets show the amplitude profiles of the two supermodes in the dual core structure of this example, at a wavelength of $1.55 \mu\text{m}$, as plots of the electric field amplitude along a line connecting the cores in the bottom-right corner, and as color plots in the top-left corner (where the boundary between each cylindrical core and the cladding is given by the thick white line).

Additionally, we plot the coupling constant obtained from two methods for different dual-core step-index fiber in Figure 6.4. The red bars in both graphs denote the boundary which the odd mode disappears for the given structure, tested in COMSOL. As we can see from both graphs, the deviations of the coupling constant obtained from two methods have already appeared before the wavelength approaches the cut-off. This is consistent with the analysis in Figure 6.3. Even though the odd mode disappears at the cut-off wavelength, the coupling constants can still be calculated from Equations (2.26) and (2.24). However, the way of obtaining the coupling constant from COMSOL shows the coupling constants are not matched with two methods when increasing the wavelength. We calculate that the values of wD/a are around 2 where the percent difference between the curve and the COMSOL-calculated dots is above 5%. It is satisfied with the way of calculating the coupling constant with Equation (2.26) from [37].

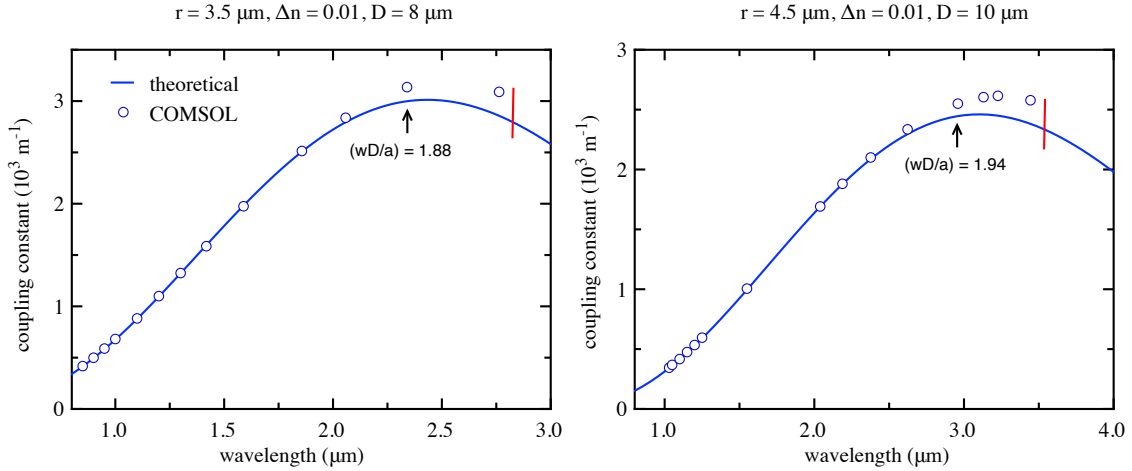


Figure 6.4: The curves for the coupling constants are obtained from Equations (2.26) and (2.24). The data points close to the curves for the coupling constants are obtained from COMSOL and Equation (2.23). The red bars in both graphs denote the boundary which the odd mode disappears for the given structure. Left: silica dual-core step-index fiber with a refractive index contrast of $\Delta n = 0.01$ and core radius of $3.5 \mu\text{m}$. Right: InF_3 dual-core step-index fiber with a refractive index contrast of $\Delta n = 0.01$ and core radius of $4.5 \mu\text{m}$.

6.4 Discussion

In this chapter, we introduced the way of getting the mode profile and effective index for each mode for cylindrical waveguides in COMSOL. As for the single-core step-index fiber, we accessed the accuracy of the results from chapter 3. We can see that most data points are well-aligned with the theoretical values derived in chapter 3. We also took into account of absorption of different materials. Before the materials get absorbed, the values of Δk are matched with two methods.

We also investigated the limitations of using COMSOL Multiphysics software. However, the cladding size and the mesh setting would affect the results obtained from COMSOL. When the mesh setting doesn't change and keep increasing the size of cladding, we found that the effective index of each mode would not become accurate anymore. We tested and found an appropriate range of the cladding so that the results of the effective index were stable. We took care of the limitations in COMSOL and obtained the most valid results as possible.

Then, we we accessed the accuracy of the results for the dual-core step-index fiber from chapter 4. Since the coupling constants are obtained by two different methods, we verified that the results are accurate with each other when expression (2.26) is valid for larger values of wD/a (See Ref. [37]. And we discussed the deviations when the distance between two cores are closer, which leads to expression (2.26) not valid anymore for small value of wD/a . Additionally, we investigated the deviations of the normalized propagation constant and the coupling constant when the generated frequency is approaching towards the cut-off frequency for the odd mode. To avoid the deviations and the cut-off for the odd mode, we have to work within the limit of the generated wavelength in CLPM configuration.

Overall, we can always use COMSOL Multiphysics software to help access the accuracy of the results obtained from expression (2.26) in CLPM configuration. We must consider the deviations and limitations from two methods when we design the structure of the fiber configuration and the generated wavelength to achieve CLPM in this work.

Chapter 7

Experimental investigation of dual-core fibers

Dual-core fiber contains a couple of parallel cores. The coupling effect between two cores which caused by light evanescent field can realize multi-selection function on light power, wavelength, and so on. In conventional fiber couplers, into which standard fibers are processed, the coupling of the evanescent-mode fields is achieved locally over a relatively short interaction length [61]. Compared with the conventional fiber, dual-core fiber has broader application in fiber communication [62–65]. From the previous discussion of investigating designs where CLPM can be achieved under dual-core fiber configuration, we seek to realize such designs experimentally, and use our laser source to experimentally demonstrate the predicted results.

In this chapter, we show some dual-core silica fibers from our collaborations. Experimental setup to determine the damage threshold power and the coupling length will also be presented. We will explain the issues and difficulties of measuring the coupling length and evaluate the dual-core silica fiber we conducted. Based on our current experimental setup, we will discuss the research plan of seeking for optimum dual-core fibers and achieving CLPM experimentally.

7.1 Description of the fibers used

We borrowed several different types of dual-core silica fiber from The Key Lab of Special Fiber Optics, led by Prof. Shi from Shanghai University. All experiments discussed in this section are involved with the dual-core silica fibers they provided. The geometric structures are respectively presented in Figure 7.1 for the dual-circular-core fiber (a), the dual-egg-shaped fiber (b) and the dual-elliptical-core fiber (c). The radius of circular core is $4.00\ \mu m$, and the distance between the centers of two cores is $12\ \mu m$. The half long axis of elliptical core is $a = 5.66\ \mu m$, and the half short axis is $b = 2.83\ \mu m$. In order to keep the same area and pitch among the three kinds of fiber, we take the egg-shaped core as in Figure 7.2. The structure of egg-shaped core is composed of two tangent circles with different radius. The radius of the big circle is $3.0\ \mu m$ and the radius of the small circle is $2.3\ \mu m$, O_1 is the center of the big circle and O_2 is the center of the small circle. The midpoint O' of O_1O_2 is supposed as the center of egg-shaped core.

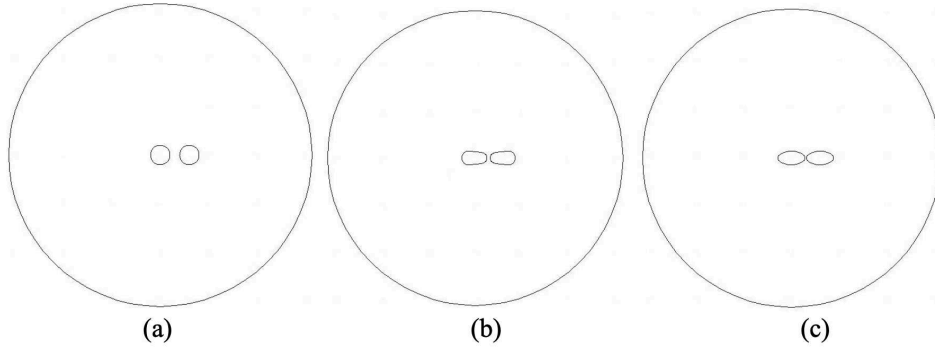


Figure 7.1: Geometric structure of (a) dual circular-core fiber, (b) dual egg-shaped-core fiber and (c) dual elliptical-core fiber.

Our collaborations provided us the end-face photos of different kinds of dual-core fiber under the microscope. Limited by manufacture condition, the cores shape and pitch in dual-core fiber often have some deviation as shown in Figure 7.2, which may affect the coupling characteristics between the two cores. We took pictures about

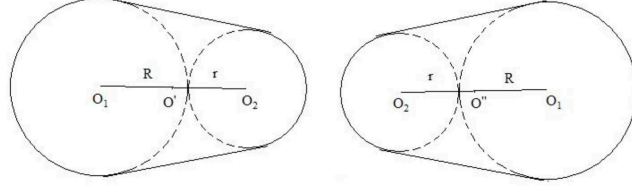


Figure 7.2: Detailed structure of the dual-egg-shaped-core.

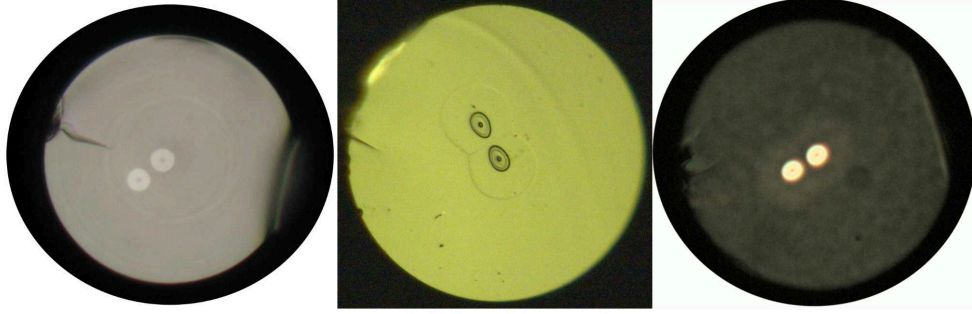


Figure 7.3: Microscopic end-face photos, obtained by Shanghai University, of three kinds of dual-core fiber (Fiber (a), (b) and (c), from left to right).

the same facet of the fiber using the microscope in our lab. Figure 7.4 shows the only two kinds of dual-core fiber (b) and (c) we borrowed from Shanghai University. Although the resolution of the camera is kind of low, we are still able to identify the shapes of those two kinds of dual-core fiber. Compared to the dual-circular-core fiber, the required coupling length between two cores for those two types may become longer. The experiments we conduct are based on those two kinds of silica dual-core fiber.

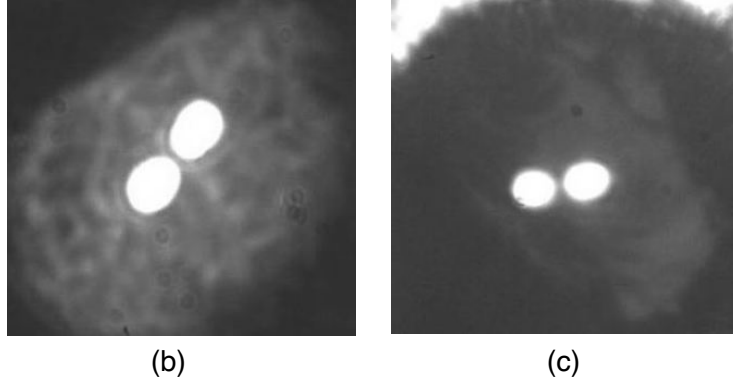


Figure 7.4: Microscopic end-face photos, obtained by ourselves, of two kinds of dual-core fiber (b) and (c) from Shanghai University.

7.2 Experimental setup

In order to study the light propagation in dual-core fiber, first of all, we built an optical system to illuminate the cross-section of dual-core fiber. Figure 7.5 shows a schematic diagram of the experimental setup. The laser system is a Clark-MXR CPA series 2101 at a repetition rate of 30 MHz which produces at 0.1-ps pulses at 1550 nm. The laser beam was guided through mirrors and focused into the front-end of the fiber by an optical correction microscope objective with its magnification 20X and focal length $f = 10\text{ mm}$. After that, one piece of 10-cm-long dual-core fiber was mounted. At the opposite end of the fiber, another microscope objective with magnification 16X was placed a focal distance away from the fiber tip. The cross-section of the fiber can be illuminated by white light on both sides and projected on a CCD camera. Thus, proper beam launching conditions were accurately monitored so that we could see the laser spot was on top of two cores on the screen of camera in Figure 7.5.

With the help of the illumination system, we can not only determine the injection of each core and measure the respective power out of each core, but also examine how good the fiber was cleaved. We should be able to observe almost equal brightness

out of two cores when the cross-section is illuminated from the opposite end of the fiber. If not, the fiber is badly cleaved or there is a damage on the core itself, which is convenient for us to identify in this way. It is of huge importance when measuring the coupling-length of dual-core fiber for later.

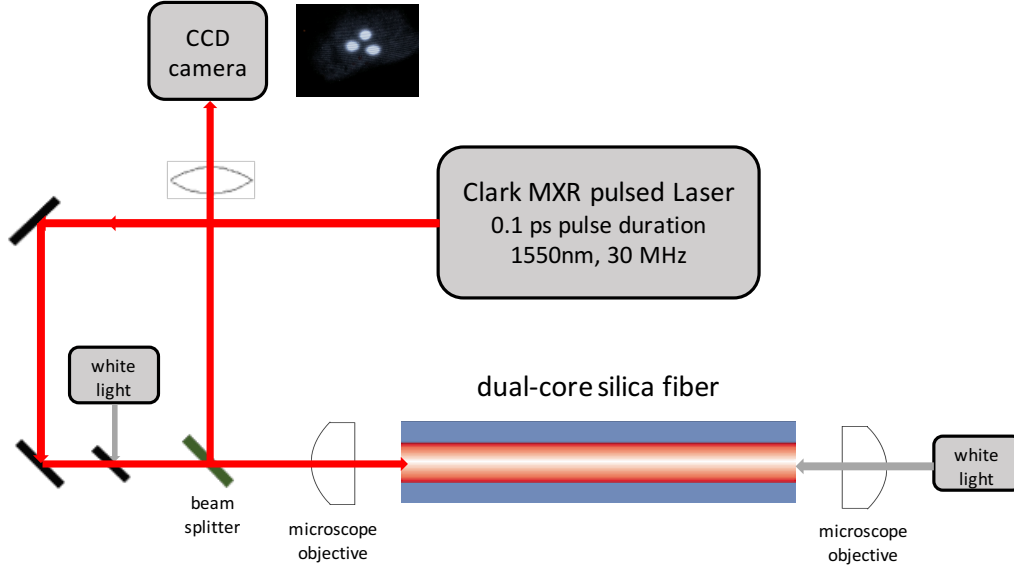


Figure 7.5: Experimental setup for the illumination of the cross-section of dual-core fiber.

7.3 Determination of damage thresholds

To find out the threshold power that causes the damage of silica dual-core fiber, we improved the previous experimental setup as displayed in Figure 7.6. The laser system is different: a Clark-MXR CPA series 2101 laser at a repetition rate of 1 kHz produces 1-ps pulses at 773 nm. The pulse is sent into a Light Conversion TOPAS (Traveling-Wave Optical Parametric Amplifier System) device. The TOPAS emits wavelength tunable pulses by nonlinear optical parametric generation to output a chosen wavelength. In this experiment, the wavelength was set at 1500 nm. Different

neutral density filters were used to control the incident optical power guided into the fiber. An optical power detector placed after the beam splitter was to collect the reflective beam as input. At the opposite end of the fiber, another optical power detector as output was placed right after the second microscope objective, which would collect the total power out of the fiber.

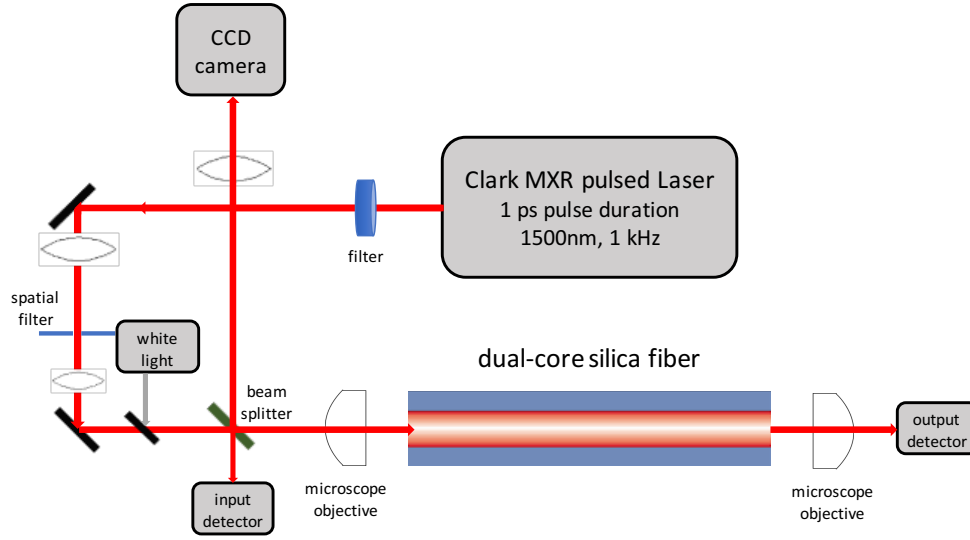


Figure 7.6: Experimental setup for threshold power that causes the damage of silica dual-core fiber.

In order to obtain a clean Gaussian beam, or a better spatial intensity variation in the beam, we utilized a spatial filter system that included a focusing lens, a pinhole, and a collimating lens. The correct optics and pinhole for the experiment depend on the wavelength of the input beam, the diameter of the source beam and the desired exit beam. The laser beam was first focused through a pinhole by an aspheric focusing lens with a longer focal length $f_1 = 100mm$. The pinhole is larger enough to accommodate almost the entire diameter of the laser source. After the pinhole, the beam passed through a planar convex lens with a shorter focal length $f_2 = 50mm$. Assuming that the beam on the input and the output sides of the spatial filter make the same angle with the main optical axis. Then, the beam was

compressed by a factor of $\frac{f_2}{f_1} = 0.5$. In this way, we are able to launch a clean Gaussian beam into one single core of the fiber. For optical performance, we chose the pinhole and lenses such that they fitted the operating wavelength range, high beam power, and had least amount of light reflected from each surface.

The peak intensity of the optical pulses can be calculated from the pulse duration, the pulse energy and effective area. To probe the linear coupling characteristics of the fiber, we switched to the laser system at a repetition rate of 30 MHz which produces 0.1-ps pulses, for the reason of lower peak intensity. The effective area of the fiber is assumed about $50\mu m^2$.

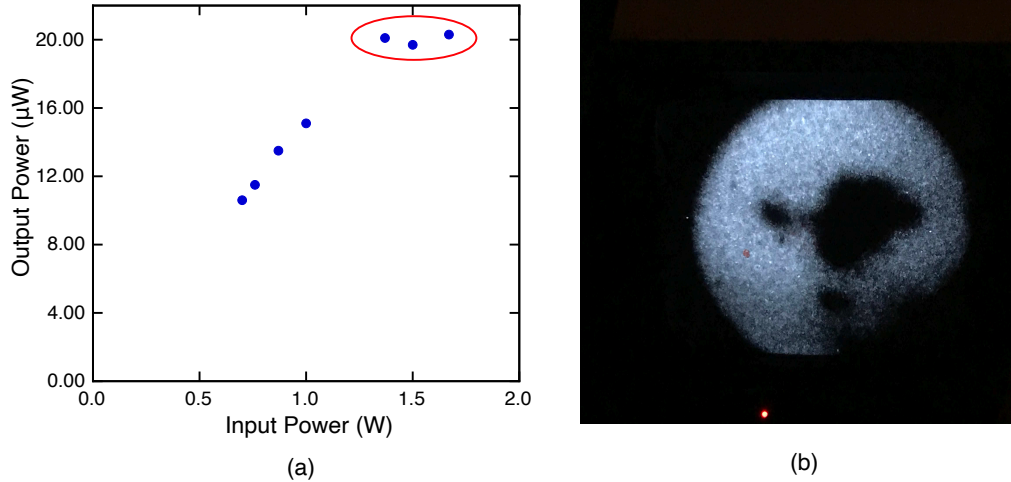


Figure 7.7: (a) Experimental results of output vs. input power, as measured in Figure 7.6. (b) The damage on dual-core fiber, when the input peak intensity was measured around $50 \text{ kW}/\mu m^2$.

To measure the threshold power that causes the damage of silica dual-core fiber, different neutral density filters were used to control the incident optical power guided into the fiber. We switched to the laser system at a repetition rate of 1 kHz which produces 1-ps pulses due to high peak intensity. Figure 7.7 (a) shows the experimental results of output vs. input power. The output grows linearly with the increase value of the input, which indicates a good coupling characteristic when the input

power of the pulses is relatively low. Then, the output power becomes saturated, indicating the fiber has already been damaged. Figure 7.7 (b) presents the damage on dual-core fiber, when the input peak intensity was calculated about $50 \text{ kW}/\mu\text{m}^2$. For pulses much longer than 50 ps, damage occurs at a nearly constant irradiance of $4.75 \text{ kW}/\mu\text{m}^2$ [46, 66, 67]. For shorter pulses, the avalanche evolves more slowly than the pulse envelope, so the threshold fluence lies above the line from long pulses.

7.4 Determination of coupling length

In order to check the optical switching for different wavelength, we improved the previous experimental setup as shown in Figure 7.8. The laser system is Clark-MXR CPA series 2101 laser at a repetition rate of 1 kHz produces 1-ps pulses at 773 nm. The pulse is sent into a Light Conversion TOPAS (Traveling-Wave Optical Parametric Amplifier System) device. The TOPAS emits wavelength tunable pulses by nonlinear optical parametric generation to output a chosen wavelength. In this experiment, the wavelength can be tuned from 1300 nm to 1700 nm. The neutral density filters were used to control the incident optical power below the threshold damage. The beam was guided into the fiber in the same way. With the help of illumination system, we focused the pump beam using a microscope objective and launched it into one fiber core. And then we fixed the power and the position of dual-core fiber.

For the output part, we first focused the beam coming out of the other end of the fiber with another microscope objective, and guided it to the position of the pinhole. The pinhole was used to separate the beam coming out of each core. Then, a two-lens system was supplemented to help us capture the image of the cross-section of the other end of the fiber on the CCD camera, as shown in Figure 7.8. In this way, we can measure different optical power of both cores using a power meter placed after a beam splitter. When the pinhole was open, we should observe two beams out of dual-core fiber with different brightness. After we controlled the pinhole and only focused on one core, the optical power can be measured. When this process

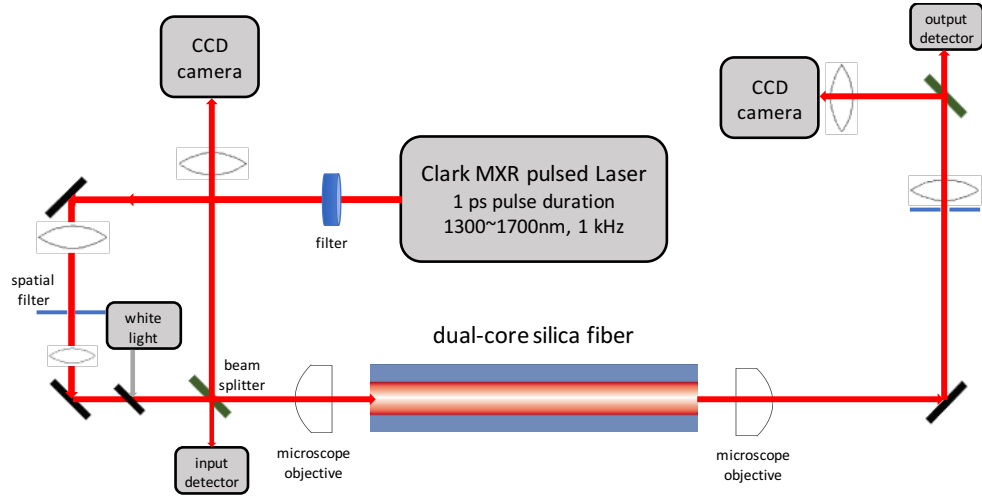


Figure 7.8: Experimental setup for the measurement of coupling-length for different wavelengths.

was complete, we tuned the wavelength from 1300 nm to 1700 nm, and recorded the output power, respectively.

We notice that the optical power coming out of each core at the output end is not changing periodically with the wavelength from 1300 nm to 1700 nm. It turns out that the coupling length is too long for us to verify the results from the analysis of achieving CLPM. If the coupling length had been a few millimeter, as we needed for CLPM, we should have seen the oscillations of the power from each core. It is likely that the index contrast in those fibers was much larger than we would have wanted. The current dual-core silica fibers are not suitable for us. With the current experimental setup, we showed the availability of achieving CLPM experimentally. So we can seek for other dual-core fibers and achieve CLPM experimentally in the future.

Chapter 8

Conclusions

By studying the mechanism of third-order optical nonlinear susceptibility and frequency downconversion in a single-core fiber, we derived the general solutions for maximum conversion efficiency from nonlinear coupled-wave equations. We defined an intensity correction factor G , which is important to achieve a condition for maximum conversion efficiency. Theoretical analysis of how we make strategies to maximize conversion efficiency are presented. We proved coupling-length phase-matching (CLPM) is a more flexible way to achieve the required phase-matching conditions by arranging two parallel waveguides in such a way as to obtain sufficient coupling of the optical waves between the cores. So other configurations are possible with other materials and different geometries.

Phase modulation is not a problem for CLPM configuration. Similarly as we discussed an intensity correction factor G for maximum conversion efficiency in a single-mode fiber, we corrected the G -dependent to the $\Delta K_\gamma = 0$ CLPM configuration. Examples are presented to demonstrate the maximum photon conversion efficiency can still be reached when taking into account the phase modulation in dual-core fiber. The most important result is that the $\Delta K_\alpha = 0$ CLPM configuration is insensitive to the phase modulation, therefore intensity-independent. When this condition is fulfilled, the coupled-wave equations for the evolution of the wave amplitudes can always lead to the completion depletion of the pump wave. This

insensitivity to phase modulation is a peculiar advantage, in addition to 100% conversion efficiency.

In practice, this idea could be used for all-fiber frequency conversion of fiber lasers, and in this work we analyzed the possibility of pumping with an Erbium fiber laser pulse with an energy of 0.5 mJ, 10 ns at $1.55\ \mu\text{m}$ or with a praseodymium fiber laser pulse with an energy of 2.3 nJ, 239 ns duration working at $1.3\ \mu\text{m}$. We presented some examples of achieving several CLPM conditions, where the intensity used in the plots is below the optical damage threshold [46]. Another important issue is that the pulse length is required to avoid pulse walk-off from three-wave interaction. Typically, the pulse lengths of Q-switched fiber lasers, nanosecond duration would be optimum. Additionally, the conversion lengths for such pulses would be around several meters, compared with absorption length in Silica and in Fluoride. We demonstrated both availability and feasibility to achieve CLPM with several possible fiber lasers.

We theoretically developed a model for the third-order polarization in a resonant medium for the case of degenerate four-wave mixing. The theoretical analysis explicitly consider that DFWM in a resonant medium actually involves two mechanisms. The measured DFWM signal as a function of decay time includes contributions from both processes. The theoretical results also indicate that the dynamic behavior of coherent four-wave mixing is different from that of the population grating contribution. When one beam is delayed, the contribution of coherent four-wave mixing reaches a peak value at the zero point of delay time. The peak value for the population grating contribution is shifted to a longer delay time because of the time integration character of the population grating. Our experimental results indicated that the excited-state molecules of DDMEBT in DCM solution decay predominantly by the transient gratings. The lifetimes of the lowest excited state of DDMEBT were measured 2-5 ps in this experiment, curve-fitted by the theoretical model.

We managed to realize designs where CLPM can be achieved under dual-core fiber configuration experimentally, and use our laser source to experimentally demonstrate the predicted results. We presented dual-core silica fibers from our collaborations. Experimental setup to determine the damage threshold power and the

coupling length was also shown. We explained the issues and difficulties of measuring the coupling length and evaluate the dual-core silica fiber. Based on our current experimental setup, we can seek for other dual-core fibers and achieving CLPM experimentally in the future.

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Vita

Jing Su graduated with bachelor degrees in Engineering, Optoelectronics Information in Huazhong University of Science and Technology, China. He began graduate school at Lehigh University where he earned his Masters degree in Photonics in May 2016. Then he earned his Ph.D. in Physics in Jan 2022. His research is mainly about nonlinear optical frequency conversion:

- Studies of four-wave mixing in free space and in fibers using coupled-wave theory.
- Investigation of a new phase matching method (Coupling Length Phase Matching, CLPM) in parallel waveguides.
- Beam propagation simulations using COMSOL and Lumerical Mode.
- Theoretical modeling of CLPM conditions in parallel waveguides in the presence of cross- and self-phase modulation.
- Investigation of the effect of population gratings on pulsed degenerate four-wave mixing.