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**THE DESIGN, CONSTRUCTION,
CHARACTERIZATION AND APPLICATION OF 9 μ m
OPTICAL PARAMETRIC CHIRPED-PULSE
AMPLIFIER**

QU SHIZHEN

School of Electrical & Electronic Engineering

A thesis submitted to the Nanyang Technological University

in partial fulfillment of the requirement for the degree of

Doctor of Philosophy

2020

Statement of Originality

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

1 AUG 2019
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Supervisor Declaration Statement

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

1 Aug 2018

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Authorship Attribution Statement

This thesis contains material from 4 paper(s) published in the following peer-reviewed journals in which I am listed as an author.

Chapter 2, section 2.4 is published as Shizhen Qu, Xiao Zou, Houkun Liang, Qijie Wang and Ying Zhang. A generalized analytical model of gain bandwidth for design of optical parametric amplifiers. Optics Communications, 449, 1-7 (2019). DOI: 10.1016/j.optcom.2019.05.031.

The contributions of the co-authors are as follows:

- Dr. Ying Zhang provided the initial project direction and edited the manuscript drafts.
- I derived the model, performed the simulation and prepared the manuscript drafts
- Dr. Ying Zhang, Dr. Xiao Zou, Dr. Houkun Liang and Prof. Qijie Wang provided valuable suggestions for the simulation, discussed the results.
- Dr. Ying Zhang, Prof. Qijie Wang and Dr. Houkun Liang reviewed the manuscript.

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- Dr. Ying Zhang provided the initial project direction and edited the manuscript drafts.
- Dr. Xiao Zou, Mr. Kun Liu and I participated in the discussion for possible solutions.

- Dr. Xiao Zou designed and conducted the simulation and prepared the manuscript drafts.
- Dr. Cheng Liu assisted the diagram plot.
- Dr. Houkun Liang, Dr. Cheng Liu, Mr. Kun Liu and I discussed the results.
- Dr. Houkun Liang, Prof. Qi Jie Wang and Dr. Ying Zhang reviewed the manuscript.

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- I designed and conducted the simulation and experiment, analysed the results and prepared the manuscript.
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Table of Contents

<u>Summary</u>	1
<u>List of Figures</u>	4
<u>List of Tables</u>	11
<u>List of Abbreviation</u>	12
<u>Chapter 1. Introduction</u>	1
1.1 Motivation and Background.....	1
1.2 Objectives.....	9
1.3 Major contributions of the thesis.....	10
1.4 Organization of the thesis.....	12
<u>Chapter 2. Theory and Models of OPCPA</u>	14
2.1 Introduction	14
2.2 Basic Principle of OPCPA	15
2.3 Challenges in development of OPCPA in LWIR regime.....	23
2.4 Gain bandwidth model for non-collinear OPCPA	31
2.5 Design non-collinear OPCPA by using N th order flatness of phase mismatch principle.....	44

2.6 Angular dispersive OPA (AD-OPA).....	47
2.7 Conclusions	51
<u>Chapter 3. Design and Implementation of a 9μm OPCPA .....</u>	<u>53</u>
3.1 Introduction	53
3.2 OPCPA system and its key challenge	54
3.3 Design of 9 μ m OPCPA system.....	61
3.4 Implementation of 9 μ m OPCPA	75
3.5 Conclusions	83
<u>Chapter 4. LWIR Pulse Self-compression in Solid .....</u>	<u>85</u>
4.1 Introduction	85
4.2 Literature review of single-cycle pulse compression.....	85
4.3 Pulse self-compression driven by the developed 9 μ m OPCPA	87
4.4 Experiment and simulation results	94
4.5 Conclusions	110
<u>Chapter 5. 9μm OPCPA driven HHG in solid .....</u>	<u>112</u>
5.1 Introduction	112
5.2 High harmonic generation.....	112

5.3 HHG in the ZnSe.....	114
5.4 Conclusions	122
<u>Chapter 6. Conclusion and Recommendation.....</u>	123
6.1 Conclusions	123
6.2 Recommendations for future research	124
<u>Author's Publications .....</u>	127
<u>Bibliography .....</u>	129
<u>Appendix I. Interferometric autocorrelator.....</u>	151

1 **Summary**

2 The high energy, high average power, ultrafast long-wavelength-infrared (LWIR, 8-
3 15 μ m) laser sources [1] have become an emerging research domain in recent years due
4 to their extensive applications, for instance the advanced molecular spectroscopy [2-7],
5 and the strong field experiment [8-10] (soft and hard X-ray generation and light
6 induced electron acceleration, and the isolated attosecond pulse generation, etc.).
7 Among all the high energy, ultrafast laser techniques, the optical parametric chirped
8 pulse amplification (OPCPA) attracts a lot of interests thanks to its high energy
9 scalability, broad gain bandwidth, low thermal load and wide wavelength tunability.
10 Nowadays, the operating wavelength range of the high energy ultrafast OPCPA system
11 has been pushed towards the $>8\ \mu\text{m}$ longer wavelength mid-IR regime.

12 However, the challenges of developing the ultrafast, longer wavelength mid-IR
13 OPCPA lies in the scaling-up of its output energy, average power and bandwidth. In
14 this dissertation, the gain bandwidth limitation of a longer wavelength mid-IR OPCPA
15 is demonstrated. This limitation is introduced by the significant dispersion in the NIR
16 regime which approaches to the band gap of the nonlinear medium. The substantially
17 increased dispersion causes the stronger group velocity mismatch between the NIR
18 pump/signal and the LWIR idler which inhibits a large gain bandwidth.

19 In order to overcome the limitation, one way is to introduce a non-collinear phase
20 matching geometry in the parametric amplification. A general analytical model of the
21 gain bandwidth which could be applied to the non-collinear OPA/OPCPA is
22 demonstrated. Compared with the traditional gain bandwidth model, the proposed
23 model breaks through the constrain about the direction of the idler wave vector and
24 could enhance 5% accuracy of the gain bandwidth evaluation, especially in the longer
25 wavelength mid-IR regime. By applying the proposed model to the OPCPA, the
26 relations between the gain bandwidth and the key design parameters such as the non-

1 collinear angle, pump intensity and crystal length are revealed. Based on the theoretical
2 analysis, an angular-dispersive OPA method is proposed to enhance the gain bandwidth
3 of the parametric amplification.

4 Using a large band gap material as the nonlinear medium is another way to enhance the
5 gain bandwidth of OPCPA. Based on the large band gap LiGaS₂ crystal, we designed
6 and implemented, to the best of our knowledge, the first LWIR OPCPA at 9μm
7 wavelength driven by the 1μm Yb:YAG pump laser. The OPCPA could provide 9μm
8 pulses with 145fs pulse duration and 14μJ pulse energy and 140mW average power.
9 Sophisticated dispersion management is conducted with considering both higher order
10 dispersion and the optical parametric phase which cannot be ignored in the LWIR
11 regime.

12 In order to further compress the LWIR pulse to single-cycle regime, we study the
13 supercontinuum (SC) generation and self-compression in the anomalous dispersive
14 materials including the KrS-5, ZnSe and GaAs. It is the first, to the best of our
15 knowledge, experimental investigation on the LWIR pulse self-compression in solids
16 with the occurrence of filamentation. The LWIR could be utmost compressed to 45fs,
17 1.5 optical cycle in the 5mm KrS-5. An over 5-octave spanning SC spectrum is
18 measured in the 9mm ZnSe crystal. The experiment results also indicate the crucial
19 roles of the filamentation and dispersion in assisting the spectrum broadening and pulse
20 self-compression.

21 At last, the LWIR OPCPA system is applied to produce the high harmonic generation
22 (HHG) in solid ZnSe material. The generated high harmonic spectrum includes the up-
23 to 24th order harmonic peaks. The cutoff energy is measured as 3.31eV which is 0.61eV
24 higher than previously reported HHG in ZnSe driven by the MWIR laser [11]. The
25 measurement result is proved to be reliable and repeatable. The experiment result
26 shows that the HHG process is a non-perturbative light matter interaction. It also shows

- 1 that the linear dependence of the cutoff energy with respect to the driving wavelength
- 2 is still fulfilled in the LWIR region.

1 List of Figures

2	Fig.1.1. (a) Development of the OPCPA systems towards the longer wavelength mid-	
3	IR. (b) Achievable few-cycle pulse durations against central wavelengths of the	
4	OPCPA in which the nonlinear pulse compression is not included. The historical	
5	position of this work is represented by the red dot.....	6
6	Fig.2.1. Comparison between the (a) Stimulated emission and (b) Optical parametric	
7	amplification.....	17
8	Fig.2.2. Definitions of (a) the non-collinear angle ρ_l, θ , pump-surface normal angle	
9	α , coordinate $\mathbf{r} = \mathbf{x}, \mathbf{y}, \mathbf{z}$ and (b) coordinate $\mathbf{r}_l = \mathbf{x}_l, \mathbf{y}_l, \mathbf{z}_l$. It should be noted that	
10	$\mathbf{z}$ points to the direction of $\Delta\mathbf{k}$ and $\mathbf{z}_l$ points to the direction of $\mathbf{k}_l$. Both $\mathbf{y}$ and $\mathbf{y}_l$	
11	are located in the surface formed by the wave vectors.....	18
12	Fig.2.3. The variation of the parametric gain in three different regions [47].	22
13	Fig.2.4. Sensitivity of phase mismatch. Here λ_{bg} is the wavelength that corresponds	
14	to band gap of GaSe crystal.....	25
15	Fig.2.5. (a) Gain bandwidth of the $9\mu\text{m}$ GaSe based OPCPA under different pump	
16	wavelength changing from $1\mu\text{m}$ to $3\mu\text{m}$. (b) Dispersion curve of the GaSe.....	26
17	Fig.2.6. The plot of (a) the OPP evolution with the propagation distance L and the	
18	comparison between OPP and material dispersion at (b) $L=2\text{mm}$, (c) $L=4\text{mm}$, (d)	
19	$L=6\text{mm}$, (d) $L=8\text{mm}$	28
20	Fig.2.7. OPP introduced GDD in (a) the BBO crystal (b) the LGS crystal. (c) The	
21	comparison between OPP introduced GDD and the material dispersion added	
22	GDD for $9\mu\text{m}$ OPCPA in the LGS crystal is also included.....	30

1	Fig.2.8. Diagram of the (a)Collinear geometry; (b)Non-collinear geometry;	
2	(c)Angular dispersive geometry	32
3	Fig.2.9. (a) Comparison between two different constraints on the direction of the idler	
4	wave vector (Eq.2.11 and Eq.2.14); (b) Gain bandwidth with different pump-	
5	surface normal angle α ranging from $\alpha = -20^\circ$ to 20° . The comparison has	
6	been made between three cases where, case 1 (blue dashed line): central	
7	wavelength of the pump $\lambda_{p0} = 2.06\mu\text{m}$ and signal $\lambda_{s0} = 3.1\mu\text{m}$; case 2 (red	
8	dash-dotted line): $\lambda_{p0} = 2.06\mu\text{m}$ and signal $\lambda_{s0} = 2.6\mu\text{m}$; case 3 (black	
9	line): $\lambda_{p0} = 2.06\mu\text{m}$ and signal $\lambda_{s0} = 2.2\mu\text{m}$;	35
10	Fig.2.10. Diagrams on the determination of idler wave vector and the wave vector	
11	mismatch, with considering (a) idler is an ordinary wave; (b) idler is an	
12	extraordinary wave.	36
13	Fig.2.11. Comparison between the evaluated gain bandwidth based on the model	
14	(Eq.2.19, black line; Eq.2.20, blue dashed line) and numerically solution of the	
15	coupled wave equation, red dotted line). (a) $\alpha = -20^\circ$, (b) $\alpha = 0^\circ$, (c) $\alpha = 20^\circ$.	
16	.....	40
17	Fig.2.12. Gain bandwidth with (a) non-collinear angle ρ_s , (b) pump intensity and	
18	crystal length, (c) pump-surface normal angle α , and the Phase matching	
19	geometry when condition $\alpha = -\theta$ is fulfilled.....	43
20	Fig.2.13. (a) Phase mismatch curve and (b) Conversion efficiency under 0^{th} (black	
21	line), 1^{st} (blue line) and 2^{nd} (red line) order flatness.....	46
22	Fig.2.14. (a) Schematic of the AD-OPA; (b) Non-collinear angle of signal for the	
23	optimized grating design (red line) and the polynomial based grating design	
24	(blue line); (c) Comparison of the flatness of the phase matching curve between	

1	collinear OPA and the AD-OPA; (d) Calculated amplified spectrum and	
2	parametric gain curve.	48
3	Fig.2.15. (a) Comparison of the phase mismatch curves between the AD-OPA system	
4	and NOPA system. The phase mismatch of the AD-OPA is shown in the red line	
5	and the phase mismatch of the NOPA is represented by the yellow line. The	
6	designed non-collinear angle of the AD-OPA system is also demonstrated with	
7	blue line. (b) Comparison of the amplified signal spectra between the AD-OPA	
8	system and NOPA system. The spectrum of the AD-OPA output is shown in the	
9	red line and the spectrum of the NOPA is represented by the yellow line.....	50
10	Fig.3.1 Illustration of the general schematic of an OPCPA system.	54
11	Fig.3.2 Illustration of (a) a Gaussian pulse and (b) the instantaneous frequency of the	
12	pulse in SPM process.....	56
13	Fig.3.3. Information of the LGS crystal, including (a) Transparency curve in 1-12 μ m	
14	spectral range; This image is provided by Mrs. Yulia Smirnova from the ASCUT	
15	Ltd. & Co. KG; (b) Transmission at 1.03 μ m under different pump energy; (c)	
16	Normalized conversion efficiency with different idler wavelength and azimuthal	
17	angle; (d) Diagram of the cutting of the LGS crystal.	63
18	Fig.3.4. (a) Schematic of the SC experiment; L1,L2: CaF2 lenses with f=100mm and	
19	f=50mm, respectively; HR: high reflective mirror. (b) The transmittance curve of	
20	the HR mirror.	66
21	Fig.3.5. Plot of the (a) SC spectra in the 4mm (black dot line), 8mm (red dash-dot	
22	line), 12mm (blue dashed line) and 16mm (orange line) YAG crystals, and the	
23	images of the conical emission, filamentation, and NIR beam profile measured at	
24	the back surface of the (b) 4mm, (c) 8mm, (d) 12mm, (e) 16mm YAG crystals.	67

1	Fig.3.6. Simulation results for the parametric gain in (a) single LGS crystal with 3mm	
2	thickness and (b) two cascaded LGS crystals with 3mm and 2mm thickness	
3	respectively, and the support idler spectrum in (c) single LGS crystal and (d) two	
4	cascaded LGS crystals, under different pump-signal energy ratio U_p/U_s . Here U_p	
5	and U_s represent for the input pump and signal pulse energy, respectively.	70
6	Fig.3.7. Comparison between a 130fs transform limited pulse at 9 μ m (blue line) and	
7	the stretched pulse with 383539 fs ³ residual TOD (red line).	75
8	Fig.3.8. Schematic of the LWIR OPCPA system.	76
9	Fig.3.9. The measured and retrieved Frog trace of (a) the seed pulses after the 1 st	
10	stretcher and (c) the seed pulses after the 2 nd stretcher, and the retrieved temporal	
11	profiles of (b) the seed pulses after the 1 st stretcher and (d) the seed pulses after	
12	the 2 nd stretcher.	77
13	Fig.3.10. Simulation results for the parametric gain in the pre-amplification stage	
14	(Pre-amp, first crystal: black line, second crystal: blue line) and the main-	
15	amplification stage (Main-amp, first crystal: red line, second crystal: yellow	
16	line).	78
17	Fig.3.11. The spectrum of signal pulses measured at the output of (a)the seed	
18	generator; (b) the pre-amplification stage, and (c) the main-amplification stage.	
19	.....	79
20	Fig.3.12. Properties of the output LWIR pulses with considering: (a) Energy long-	
21	term stability measured in 1 hour (the beam profile is demonstrated at the insect);	
22	(b) Energy scalability; (c) Spectrum; (d) Measured (black dashed line) and	
23	retrieved (red line) IAC trace; (e) Retrieved temporal profile.	81
24	Fig.4.1. Illustration of the pulse self-compression process.	86

1	Fig.4.2. The schematic of the pulse self-compression experiment based on a 9 μm	
2	OPCPA, and the spectral and temporal characterization. L: Lens; BS:	
3	Germanium beam splitter; M: Silver mirror; P1, P2: Off-axis parabolic mirrors;	
4	X: Nonlinear crystal.	88
5	Fig.4.3. Measured SC spectra in (a) 2mm BaF2 and (b) 5mm CaF2 with different	
6	distances between the front surface and the focal point.	90
7	Fig.4.4. The material dispersion of the KrS-5 (Black line), ZnSe (Blue dashed line)	
8	and the GaAs (Red dotted line).	91
9	Fig.4.5. Self-compression experiment in the ZnSe which include, (a) the SC spectra	
10	variation with different surface-focal point distance, (b) the output energy	
11	variation with different surface-focal point distance, and the IAC traces of the	
12	self-compressed pulses at (c) L=9mm, (d) L=8mm, (e) L=7mm, (f) L=6mm, (g)	
13	L=5mm and (h) L=4mm.	95
14	Fig.4.6. Images of (a) the filamentation in the ZnSe and (b) the output beam profile.	
15	.....	97
16	Fig.4.7. Spectra of the SCG in ZnSe. (a) Measured and simulated spectra at >-30db	
17	level. (b) Full spectrum of the SCG. (c) 1 st to 7 th order harmonics generated in	
18	the 0.6mm ZnSe.....	98
19	Fig.4.8. Measured and simulated temporal profile of the self-compressed pulses. (a)	
20	Measured and retrieved IAC trace; (b) Retrieved temporal profile; (c) Simulated	
21	spatial temporal profile at the output surface of the ZnSe; (d) Simulated spatial	
22	averaged temporal profile.....	99
23	Fig.4.9. Self-compression experiment in the KrS-5 which include, (a) the SC spectra	
24	variation with different surface-focal point distance, (b) the output energy	

1	variation with different surface-focal point distance, and the IAC traces of the	
2	self-compressed pulses at (c) $L=6\text{mm}$, (d) $L=5\text{mm}$, (e) $L=4\text{mm}$, (f) $L=3\text{mm}$, (g)	
3	$L=4\text{mm}$	101
4	Fig.4.10. Images of (a) the filamentation in the KrS-5 and (b) the output beam profile.	
5	.....	101
6	Fig.4.11. Measured and simulated (a) spectra, and temporal profiles which include,	
7	(b) Measured and retrieved IAC trace; (c) Retrieved temporal profile; (d)	
8	Simulated spatial temporal profile at the output surface of the KrS-5; (e)	
9	Simulated spatial averaged temporal profile.	102
10	Fig.4.12. Self-compression experiment in the GaAs which include, (a) the SC spectra	
11	variation with different surface-focal point distance, (b) the output energy	
12	variation with different surface-focal point distance. The output beam profiles	
13	measured at $L=-1.5\text{mm}$, 1.5mm and 7.5mm are also included in the insets. ..	104
14	Fig.4.13. Measured and simulated (a) spectra, and temporal profiles which include,	
15	(b) Measured and retrieved IAC trace; (c) Retrieved temporal profile; (d)	
16	Simulated spatial temporal profile at the output surface of the GaAs; (e)	
17	Simulated spatial averaged temporal profile. The photo of the output beam	
18	profile is shown in the inset of figure (a).	105
19	Fig.4.14. Simulated plasma density at different positions of (a) KrS-5 and (b) ZnSe.	
20	.....	106
21	Fig.4.15. Spatio-temporal intensity profile are shown for KrS-5 at (a) $z = 0\text{ cm}$, (b) z	
22	$= 0.2\text{ cm}$, (c) $z = 0.3\text{ cm}$, and (d) $z = 0.5\text{ cm}$. The color bars show intensity in	
23	W/cm^2	107

1	Fig.4.16. Spatio-temporal intensity profile are shown for ZnSe at (a) $z = 0$ cm, (b) $z =$	
2	0.3 cm, (c) $z = 0.5$ cm, and (d) $z = 0.9$ cm. The color bars show intensity in	
3	W/cm^2 . The simulated spatio-temporal intensity profiles look different with the	
4	ones in Fig.4.7(c) in the manuscript since the $7.3 \mu\text{m}$ long-pass filter is not added	
5	in this simulation.	108
6	Fig.5.1. The mechanism of the solid-based HHG [154].	113
7	Fig.5.2. Full HHG spectra measured in (a) 0.4mm ZnSe, (b) 2mm (blue line) and	
8	9mm (red line) ZnSe crystals. The cutoff of the HHG in 0.4mm ZnSe is detailed	
9	displayed in the inset of (a).	116
10	Fig.5.3. Intensity of the Nth order harmonic peaks as a function of pump intensity for	
11	the (a) $N=4$, (b) $N=5$, (c) $N=6$, (d) $N=7$; (e) $N=10$ to 24. The dashed lines in (a) –	
12	(d) show the I^N dependence of the harmonic peak intensity. The dashed line in	
13	(e) show the linear dependence of the harmonic peak intensity with respect to I .	
14	.....	119
15	Fig.5.4. Linear dependence of the cutoff energy with respect to the wavelength of	
16	driving laser.	120
17	Fig.5.5. Comparison between the HHGs in the 0.4mm ZnSe and (a) in the 0.5mm	
18	GaAs and (b) in the 0.5mm GaSe.	121
19	Fig.A1.1 Schematic of the interferometric autocorrelator.	151
20		

1   **List of Tables**

2	Table 3.1. Summary of the 1 μ m and 2 μ m pump sources. Here, the ‘W’, ‘H’ and ‘L’	
3	represent for the width, height and length.	59
4	Table 3.2. Summary of nonlinear materials used in the long wavelength mid-IR	
5	OPA/OPCPA.	60
6	Table 3.3. Summary of the large band gap nonlinear media.	62
7	Table 3.4. Summary of the nonlinear medium for the SCG at 1.03 μ m [97].	65
8	Table 3.5. Summary of the bulk material for the pulse stretcher.	69
9	Table 3.6. Dispersion calculation of the LWIR OPCPA system.	73
10	Table 4.1. Summary of anomalous dispersive materials at 9 μ m. The zero-dispersion	
11	wavelength (ZDW) is calculated by the Sellmeier equation [134]. The critical	
12	power P _{cr} is calculated by Eq.4.1 [133].	90
13	Table 4.2. Simulation parameters for KrS-5, ZnSe and GaAs.	93
14	Table 4.3. Comparison between self-compression experiment in gas and solid	
15	material driven by MWIR and LWIR lasers.	110
16	Table 5.1. Mean value and standard deviation (Std) of the wavelengths of the Nth	
17	order harmonic peaks.	117

18

1 **List of Abbreviation**

2	AD-OPA	Angular dispersed optical parametric amplification
3	AGS	Silver Gallium Sulfide
4	AGGS	AgGaGeS ₄ crystal
5	AOI	Angle of incidence
6	AOPDF	Acousto-optic programmable dispersive filter
7	CEP	Carrier-envelope phase
8	CPA	Chirped pulse amplification
9	DFG	Difference frequency generation
10	DOPA	Degenerate optical parametric amplification
11	EPRIAC	Evolutionary phase retrieval from interferometric autocorrelation
12	FROG	Frequency-resolved optical gating
13	FWM	Four wave mixing
14	GDD	Group delay dispersion
15	HGS	Mercury Thiogallate
16	HHG	High harmonic generation
17	HR	High reflective mirror
18	HWP	Half wave plate
19	IAC	Interferometric autocorrelation
20	IR	Infrared
21	LASER	Light amplification by stimulated emission of radiation
22	LGS	Lanthanum Gallium Silicate
23	LGSe	Lithium Gallium Selenide

1	LIS	Lithium Thioindate
2	LiSe	Lithium Selenoindate
3	LPF	Long pass filter
4	LWIR	Long-wavelength infrared
5	MWIR	Mid-wavelength infrared
6	NIR	Near infrared
7	NOPA	Non-collinear optical parametric amplification
8	OPA	Optical parametric amplification
9	OPCPA	Optical parametric chirped pulse amplification
10	OPP	Optical parametric phase
11	QCL	Quantum cascade laser
12	QPM	Quasi-phase match
13	RQPM	Random quasi phase matching
14	SC	Supercontinuum
15	SCG	Supercontinuum generation
16	SPM	Self-phase modulation
17	SWIR	Short-wavelength infrared
18	TFP	Thin film polarizer
19	TOD	Third order dispersion
20	TW	Terawatt
21	UPPE	Unidirectional pulse propagation equation
22	YAG	Yttrium aluminium garnet
23	ZDW	Zero-dispersion wavelength
24	ZGP	Zinc Germanium Phosphide

Chapter 1. Introduction

1.1 Motivation and Background

1.1.1 Motivations

In past decades, the development of ultrafast lasers in long wavelength mid-infrared (mid-IR) range is driven by both scientific research and industry applications in the area of optical spectroscopy [1-7], material processing [12-15] and remote sensing [16-19]. In the area of optical spectroscopy for material characterization, lasers of longer mid-IR wavelengths are demanded, as more and more materials such as acetone, methane, methanol, and ammonia are used in industry and they have fingerprints in the mid-IR range with wavelengths $>7\mu\text{m}$. Furthermore, not only the spectral information but also time resolution is required by the ultrafast chemistry or even attosecond chemistry [20-22] which demands the ultrafast lasers with a pulse width less than femtoseconds. Therefore, the electron dynamics which trigger or mediate chemical process could be captured. In the area of material processing, the lasers with few-cycle pulse duration are discovered to be more efficient and effective than longer laser pulses, since they introduce less heat accumulation during the processing of the polymers and macromolecular materials. For example, an ultrafast laser help to realize the nanoscale texture of material surface. Ultrafast lasers are also found capable of fabricating subsurface layers in polymer parts without damaging them.

The development of ultrafast lasers in a long wavelength mid-IR range is also an integral part of laser research as it challenges the frontier of laser technology. Nowadays, the trend of laser development is moving towards obtaining coherent radiations in two extreme regimes with either as short as possible wavelengths such as the X-ray or as long as possible wavelengths such as the THz. The challenge of developing direct X-ray and THz lasers comes from the difficulties to establish the

1 population inversion as the band gaps of concern are either too close or too apart from
2 each other in the extreme regimes. However, bright coherent sources in these regimes
3 are highly demanded for both scientific basic research and industry applications. For
4 example, coherent X-ray lasers are attractive for enabling ones to have better
5 understanding atomic structures and light-atom interaction. Traditionally, the coherent
6 light is provided by synchrotron and the free electron laser (FEL) which are both
7 expensive and space consuming. In recent years, the high harmonic generation (HHG)
8 principle has been successfully exploited to develop table-top coherent sources in the
9 X-ray regimes but in a compact size [8-10]. The frontier of the research on the table-
10 top HHG X-ray sources is to obtain as high as possible photon energy of coherent hard
11 X-ray lights. The success largely depends upon the availability of an ultrafast driving
12 laser in the longer mid-IR regime, which is used to ionize atoms to obtain HHG from
13 gases or solids or plasmas. The requirement on the ultrafast laser pulses ensures the
14 high intensity of the driving laser for atom ionization, and the demand on the as long
15 as possible mid-IR wavelength ensures the high cutoff frequency of the HHG according
16 to the scaling law of the pondermotive force in function of the driving wavelength.

17 The development of ultrafast lasers in a long wavelength mid-IR range moves towards
18 obtaining the ultrafast laser pulses of about a single cycle with a longer central
19 wavelength. The Q-switching technique pioneers the innovations in producing optical
20 pulses with high peak power and nanosecond level pulse duration. Followed is the
21 development of the mode-locking technique which initiates a new epoch in ultrafast
22 pulse generation. At present, few-cycle pulses with femtosecond level pulse duration
23 can be generated via the Kerr lens mode-locking of the Ti:sapphire lasers. With the
24 help of the chirped pulse amplification (CPA), the laser power and the pulse duration
25 can even be pushed to new highs. This ground-breaking invention on CPA has been
26 recognized by the Nobel prize in the last year. However, it is still a tough task to extend
27 the central wavelength of the traditional mode-lock lasers above 4-5 μ m. This is mainly
28 because of the serious non-radiative effects of gain medium, for instance the auger

1 effect. In contrast, a longer wavelength mid-IR radiation could be obtained by using
2 either the CO₂ lasers or the quantum cascade lasers (QCLs). The CO₂ lasers are
3 favoured for producing ~ 10 μm pulses with high power and high efficiency. The QCL
4 technique based on the cascaded inter-subband transition can push the laser central
5 wavelength to longer wavelengths towards even the THz range. However, the
6 ultrashort pulses with less than few-picosecond level pulse duration are still not
7 achievable in both the techniques due to the limited line-width of the materials. Hence
8 the optical parametric chirped pulse amplifiers (OPCPA) that are based on the
9 nonlinear frequency conversion principle have drawn more intensive attention now in
10 order to obtain both ultrafast and long wavelength mid-IR pulses with high energy. The
11 OPCPA technique borrows the idea of the CPA, where an ultrafast signal is first
12 stretched and then amplified in several OPA stages and finally re-compressed into a
13 short pulse. There is no fundamental limitation on the central wavelength of an OPCPA,
14 except on the transmission range of the nonlinear material used in the OPCPA. Since
15 the phase matching condition could be fulfilled simultaneously at various frequencies,
16 an OPCPA normally has the large gain bandwidth. Nowadays, the OPCPA systems
17 could operate at the utmost $7\mu\text{m}$ mid-IR wavelength [23]. Ultrafast, few-cycle pulses
18 could also be generated at a wide wavelength range from the near-infrared (NIR) to the
19 MWIR, for instance, the pulses with 1.5 cycles (10.5 fs) at $2.1\mu\text{m}$ [24], 3.6 cycles (38
20 fs) at $3.2\mu\text{m}$ [25], 4.5 cycles (75fs) at $5.1\mu\text{m}$ [26] and 8.1 cycles (188 fs) at $7\mu\text{m}$ [23].
21 However, it is still a research gap to extend the central wavelength of an OPCPA above
22 $7\mu\text{m}$. The generation of near-single cycle pulses through long wavelength mid-IR
23 OPCPA is of great interest particular to the development of the table-top HHG sources,
24 but it adds more challenges. The great value and potential of OPCPA inspires further
25 the development of high energy ultrafast OPCPAs in longer mid-IR wavelength ranges.

1   **1.1.2 State of the art**

2   Ever since the first demonstration of the OPCPA technique by Dubietis et al.[27] in
3   1992, the development of OPCPAs moves with a trend towards obtaining single cycle
4   pulses in longer mid-IR wavelength ranges. The development can be summarised as
5   going through three phases including the visible or NIR OPCPAs, the short wavelength
6   mid-IR ($<4\mu\text{m}$) OPCPAs, and the developing long wavelength mid-IR ($>4\mu\text{m}$)
7   OPCPAs, as shown in Fig.1.1.

8   In the first decade after 1992, a rapid progress has been made in developing the intense
9   few-cycle OPCPAs in the visible or NIR range. With the mature of the 800nm
10   Ti:sapphire laser technique and the available crystals such as the BBO and LBO
11   crystals, several pioneer OPCPA systems have been developed, for instance, the one
12   with sub-5-cycle pulses at $\sim 600\text{nm}$ wavelength were demonstrated by Cerrulo et al.
13   [28] and Shirakawa et al. [29] in 1998. In this period, several key techniques were also
14   proposed and applied to develop the OPCPA systems. The techniques include the
15   supercontinuum generation (SCG) based broadband signal generation technique and
16   the non-collinear optical parametric amplification (NOPA) technique. These
17   techniques significantly enhance the amplification bandwidth of the whole OPCPA
18   system and therefore give birth to the ultrafast lasers capable of producing two-cycle
19   pulses. The remarkable works include the system of Shirakawa et al.[30] in 1999,
20   demonstrating 4.7fs and 2.2 cycles pulses, and the system of Baltuska et al. [31] in
21   2002 which demonstrated 3.9fs and 1.9 cycles pulses.

22   In the 10 to 15 years afterwards, the ultrafast OPCPAs in the $<4\mu\text{m}$ mid-IR ranges have
23   been developed vigorously. In this period, the techniques of developing the Yb or Nd
24   doped $1\mu\text{m}$ lasers were made more mature. New materials such as the KTA, KTP, and
25   several periodically poled nonlinear crystals including the PPLN, PPLST, etc. have
26   been developed and demonstrated with large nonlinear coefficients and high

1 transmission in the mid-IR range. Supported by the intense pump lasers and the new
2 materials, ultrashort pulses have been made into 3.4 optic cycles at 2.1 μm [32], 4.2
3 optic cycles at 3.2 μm [33], and 6.4 optic cycles at 3.9 μm [34]. By using the aperiodic
4 quasi-phase matching (QPM) method, the gain bandwidth of an OPCPA can be further
5 enhanced. With the fine chirp-compensation through the acousto-optical
6 programmable dispersive filter (AOPDF), the amplified pulses in an OPCPA could be
7 further compressed to shorter pulse duration. For instance, Deng et al. [24] in 2012
8 demonstrated the 2.1 μm OPCPA that used the aperiodically poled lithium niobite
9 (APPLN) materials and the AOPDF. The 1.5 cycle pulses with 10.5fs pulse duration is
10 generated, which corresponds to 1.05times transform limited pulse width. In 2014,
11 Mayer et al. [35] reported the OPCPA capable of producing the 44.2fs, 3.9-cycle pulses
12 at 3.4 μm by using the APPLN crystal.

13 In recent few years, the research frontier of developing the ultrafast OPCPA has been
14 pushed towards the $>4\mu\text{m}$ wavelength range, as shown in the Fig.1.1(a). The rapid
15 growth of the long wavelength OPCPAs is attribute to the availability of the high
16 energy pump laser at 2 μm and the materials with broad transparency range, such as the
17 ZGP and GaSe crystals. Some pioneer works are summarized as follows. In 2018,
18 Wang et al. [36] demonstrated the 4 μm OPCPA system which can deliver 105fs pulses
19 with 2.6mJ pulse energy. Grafenstein et al [26] demonstrated the ZGP-based OPCPA
20 system operating at 5.1 μm . The system provides 4.5-cycle pulse duration with
21 dispersion management through the spatial light modulator. In 2016, Wandel et al. [37]
22 demonstrated the similar $\sim 5\mu\text{m}$ OPCPAs with 370fs pulse duration and in 2017 Kanai
23 et al. [38] developed the one with 100fs pulse width. The Biegert's group demonstrated
24 an OPCPA system with the longest central wavelength at 7 μm [23,39]. This OPCPA
25 system could now deliver 188fs, 8-cycle pulses with 0.75mJ pulse energy [23]. It set a
26 record of the longest central wavelength achieved before the OPCPA system reported
27 in this dissertation and the Ref.[40]. But the pulse duration of the OPCPA system is
28 still around or above 5 optical cycles. Further reduction of its pulse duration to a single

1 cycle is still a challenge unless using additional nonlinear pulse compression. By using
2 the SCG in gas and the chirp compensation in bulk material, Wang et al. [36]
3 demonstrated the 4 μm , 105fs pulse which can be compressed to 21.5fs with 1.6 optical
4 cycles.

5
6 Fig.1.1. (a) Development of the OPCPA systems towards the longer wavelength mid-IR. (b)
7 Achievable few-cycle pulse durations against central wavelengths of the OPCPA in which
8 the nonlinear pulse compression is not included. The historical position of this work is
9 represented by the red dot.

1.1.3 Challenges and limitations

The state-of-the-art is summarized in Fig.1.1 which shows graphically a clear trend of the OPCPA development going towards the longer mid-IR and single-cycle regimes. As shown in Fig.1.1(a), a research gap lies with the development of an OPCPA with the central wavelength above $7\mu\text{m}$. It is also shown in Fig.1.1(b) that it becomes even harder to approach the single cycle regime with the central wavelength of the OPCPA increases. The key challenges of achieving both single-cycle pulse duration and longer central wavelength can be seen from several aspects including the basic principle of OPCPA, the limitations of the key components, and the practical implementation of an OPCPA system.

The first challenge comes from the basic theory and models of OPCPA. At present, the mathematical analysis of OPCPA is still based on the traditional coupled wave equations which consider the interaction between monochromatic plane waves. More suitable models of OPCPA are under exploration to factor the pulse properties into the wave coupling processes of OPCPA. Several numerical methods, such as the well-known SISYFOS code [43], has now been widely used in the simulation of OPCPA process. However, there is still a lack of simple and explicit analytic models which relate the key performances such as the parametric gain and gain bandwidth of an OPCPA to the design parameters such as the pulse duration, the chirp rate of signal and the non-collinear angle etc. of the OPCPA. This causes the inconvenience of choosing the right parameters in the OPCPA system design. Moreover, the OPCPA system normally consists of several consecutive amplification stages. It is highly desired to have a systematic theory of modelling and optimizing a composite OPCPA system.

The second set of challenges are with the not ideal nonlinear materials and the pump lasers. On the side of the nonlinear materials, the band gaps of the currently used crystals, for instance the ZGP and GaSe crystals, are limited below 2.4eV. As a result,

1 the problems regarding the energy loss, the thermal load and the crystal damage are
2 introduced due to the multiphoton absorption. It also restricts the applied pump energy
3 and power, and in turn limits the parametric gain and the gain bandwidth. Moreover,
4 in these small-band gap materials, large material dispersion is introduced when the
5 traditional short wavelength pump sources are used in an OPCPA. In this situation, the
6 discrepancy between the group velocities of pump wave and the long wavelength mid-
7 IR signal becomes large. As a result, both the gain bandwidth and the conversion
8 efficiency of the OPCPA are limited. Regarding the pump laser, the commonly used
9 $\sim 2\mu\text{m}$ pump lasers have limited pulse energy and power. This drawback therefore
10 becomes the main handicap to enhance the energy and power scalability of the current
11 long wavelength OPCPAs. The advanced pump lasers at $0.8\mu\text{m}$ and $1\mu\text{m}$ with several-
12 order times higher energy and power, however, are still unable to be applied in the
13 current OPCPA systems due to the large multiphoton absorption and the limited
14 damage threshold.

15 Third, the practical implementation of an ultrafast OPCPA apparatus in a longer mid-
16 IR wavelength range is also a challenging job due to involving tedious alignment of
17 many reflective optics including the curved mirrors and the parabolic mirrors. In the
18 long wavelength mid-IR range, materials that are normally used in the optical lenses
19 have large dispersion. For example, the CaF_2 material has a $-5282.6\text{fs}^2/\text{mm}$ group
20 velocity dispersion at $9\mu\text{m}$. Therefore, building the beam-shaper with parabolic mirrors
21 is a better choice in this wavelength range. However, the optics alignment in the
22 parabolic mirror-based beam shaping is much tougher than the alignment of optic
23 lenses. Besides, the difference between the pump wavelength and the long wavelength
24 mid-IR signal is large while the discrepancy between the pump and idler wavelength
25 is small in the OPCPA with the longer mid-IR central wavelength. Therefore, the
26 efficient beam combination and the separation of both the signal and idler from the
27 pump are also tough to realize due to the lack of suitable dielectric mirrors. Moreover,
28 the propagation and the amplification of the long wavelength mid-IR pulses in the

1 optics could introduce large high order dispersion. It thus introduces serious difficulties
2 for fully dispersion compensation and the single cycle pulse generation.

3 Furthermore, although the near single-cycle pulses have been reported in a long
4 wavelength mid-IR range by adding the nonlinear compression stage after the OPCPA,
5 the challenges are still with the compression process. These challenges come from the
6 fundamental principle and the experimental implementation of the nonlinear
7 compression, especially when the compression is done in the solid material and in
8 the $>7\mu\text{m}$ wavelength range. The primary reason is that the principle of the solid based
9 nonlinear compression in the $>7\mu\text{m}$ wavelength range has not been fully understood.
10 Although several related theoretical works are reported in recent years [44,45], plenty
11 of questions are still waiting for answers. For instance, what is the optimal pulse
12 duration of the nonlinear compression in the $>7\mu\text{m}$ wavelength range? How does the
13 tunneling ionization and collisional ionization affect the spatial, spectral and temporal
14 profiles of the self-compressed pulses respectively? How does the plasma affect the
15 group velocity dispersion and the phase matching conditions of the coexisting
16 nonlinear process, such as the FWM and HHG, and so on? Secondly, although the
17 single cycle pulse at the $>7\mu\text{m}$ wavelength range was proved to be achievable in theory,
18 experimental verification has not been done yet. Last but not the least, researchers are
19 still exploring the suitable nonlinear materials which would support the single-cycle-
20 level pulse self-compression at the $>7\mu\text{m}$ wavelength range.

21 **1.2 Objectives**

22 The thesis addresses the above challenges and develops a high energy, near-single-
23 cycle laser source in the $>7\mu\text{m}$ longer wavelength mid-IR range. The developed laser
24 adopts the OPCPA technique with a solid-based pulse self-compression method. In this
25 thesis, the principle, design, implementation and application of the high energy,

1 ultrafast, longer wavelength mid-IR OPCPA system are systematically investigated
2 from the following aspects.

3 (1) To address the challenges with the principle of OPCPA, the traditional coupled
4 wave equation and the gain bandwidth model are revisited to make it suitable for the
5 design and analysis of the ultrafast, longer wavelength mid-IR OPCPA system.

6 (2) To mitigate the gap of a high energy laser in a longer wavelength mid-IR range, a
7 high energy OPCPA system with the central wavelength of 9 μ m is built and
8 demonstrated enhanced energy and power scalability by using the large band gap
9 nonlinear material and the advanced 1 μ m pump laser.

10 (3) To meet the trend of an ultrafast laser capable of producing single cycle pulses in
11 the long wavelength mid-IR range, a solid-based nonlinear self-compression technique
12 is adopted to further compress the output pulses of the developed OPCPA system into
13 sub-two-cycle. The basic effects of the plasma, dispersion and nonlinearity in the self-
14 compression process at the long wavelength mid-IR range are also investigated and
15 verified.

16 (4) To demonstrate the use of the developed single-cycle OPCPA system in the long
17 wavelength mid-IR range, the built laser is used to drive the high harmonic generation
18 (HHG) in solid material to prove its availability and suitability in strong field
19 applications.

20 **1.3 Major contributions of the thesis**

21 The major contributions of this thesis are summarized as follows.

22 (1) We developed a new analytical model of gain bandwidth which could be applied in
23 the non-collinear OPA/OPCPA. The new model abandons the traditional assumption
24 in which the transverse component of the wave-vector mismatch shall vanish. This

1 model is proven to be able to provide less than 15% calculation error in the calculation
2 of the gain bandwidth of the OPCPAs in the long wavelength mid-IR range. As the
3 model is not dependent upon the constraints on the wave-vector mismatch, it is more
4 general to provide more freedoms to design the performance of OPCPA. Based on the
5 theoretical model, the gain bandwidth of an OPCPA is proposed to be enhanced by
6 using an angular-dispersive optical parametric amplification technique (AD-OPA).

7 (2) We demonstrated the first of its kind high energy ultrafast OPCPA system at 9 μm
8 wavelength in the LWIR regime. The OPCPA uses the high energy 1 μm Yb:YAG laser
9 to drive the whole system and the large band gap LiGaS₂ (LGS) materials as the
10 frequency conversion media. This system is demonstrated capable of providing the
11 LWIR pulses with 14 μJ pulse energy, 140mW average power and 145fs pulse duration
12 which corresponds to sub-five optical cycles. Moreover, in the developed OPCPA, the
13 dispersion management for up to third order dispersion is also successfully
14 demonstrated by considering the OPP effect.

15 (3) We investigated the first LWIR pulse self-compression experiment through the
16 filamentation in anomalous dispersive solids such as the KrS-5, the ZnSe and the GaAs
17 crystals to compress the high energy pulses. A 1.5-cycle, 45fs self-compressed pulse is
18 generated in the KrS-5 and an over 5-octave spanning supercontinuum (SC) spectrum
19 is demonstrated in the ZnSe crystal. The effects of the filamentation, material
20 dispersion and nonlinearity in the pulse self-compression and the SC generation are
21 studied.

22 (4) We demonstrated for the first time an up-to 24th order high harmonic spectrum in
23 the ZnSe crystal driven by the developed 9 μm OPCPA system. The measurement
24 result of the high harmonic generation is proven to be repeatable and reliable. The
25 experiment also confirms the advantage of the developed long wavelength source in

1 providing higher cutoff energy and it shows the practicality of the proposed OPCPA
2 system.

3 **1.4 Organization of the thesis**

4 In this thesis, the fundamental theory, design and implementation, performance
5 enhancement and application of the long wavelength mid-IR OPCPA are
6 comprehensively illustrated. The thesis is organized in six chapters with two
7 appendixes.

8 In chapter 2, the main challenges in development of OPCPA in LWIR regime are
9 addressed by numerical studies. The numerical results show that the fundamental
10 challenge lies in the large group velocity mismatch between the pump/signal and the
11 idler waves which limits the gain bandwidth of an OPCPA. In order to enhance the
12 gain bandwidth, this chapter addresses the design of non-collinear OPCPA three
13 aspects. First of all, a general analytic model of gain bandwidth model is developed for
14 analysing non-collinear LWIR OPCPA. Based on the gain bandwidth model, the ‘Nth
15 order flatness’ design rule is introduced as a guideline to design a broadband non-
16 collinear OPCPA system. Following this rule, an AD-OPA system is numerically
17 proposed by using a designed chirped grating. The gain bandwidth provided by this
18 new method is shown to be larger than the traditionally used NOPA system.

19 Chapter 3 illustrates the development of the high energy ultrafast LWIR OPCPA
20 system. Since the angular dispersive OPA method is still hard to be applied in
21 experiment, in this chapter, we use a large band gap LiGaS₂ material to enhance the
22 gain bandwidth of the LWIR OPCPA. Based on the LiGaS₂ crystal, we design and
23 build the LWIR OPCPA with considering the material selection, the reuse of the pump
24 energy and the dispersion management in the development of the OPCPA system.
25 Comprehensive measurement in spatial, spectral and temporal domain are presented to
26 demonstrate the performance of the developed OPCPA system.

1 In chapter 4, we report the pulse compression experiment of exploiting filamentation
2 in anomalous dispersive solid materials to further reduce the $9\mu\text{m}$ pulse duration to
3 near-single-cycle regime. The chapter starts with describing the generation and
4 optimizations of both the SC spectrum and the self-compressed pulse compression in
5 the KrS-5, ZnSe and GaAs crystals. In the following, it presents the measurement
6 results and discusses the effects of the high-density plasma on the pulse self-
7 compression.

8 Chapter 5 demonstrates one application of the $9\mu\text{m}$ OPCPA in the high harmonic
9 generation. The high harmonic spectrum is generated in the polycrystalline ZnSe
10 crystal driven by the developed $9\mu\text{m}$ OPCPA. The HHG spectrum as well as the
11 repeatability of each HH peak are measured. The cutoff energy of the generated HH
12 spectrum is shown to be higher than the MWIR laser driven HHG in the ZnSe. The
13 linear dependence of the cutoff energy with respect to the driving wavelength is
14 verified to be fulfilled at $9\mu\text{m}$. The experiment confirms the advantage of the longer
15 wavelength driving lasers in the HHG application.

16 Finally, in chapter 6, we summarize the preceding chapters and draw the conclusions
17 for the dissertation. We give several recommendations for future works in developing
18 fundamental OPCPA models, enhancing the performance of the developed system, and
19 pursuing even higher average power and peak power OPCPAs in a long wavelength
20 mid-IR range.

21 Moreover, this thesis also includes one appendix. The Appendix I illustrates the
22 implementation of an interferometric autocorrelator which is used for few-cycle pulse
23 duration measurement. The phase-retrieve algorithm is also included.

Chapter 2. Theory and Models of OPCPA

2.1 Introduction

In the last two decades, the optical parametric chirped pulse amplification (OPCPA) gradually become one of the most favourable methods in generating high energy, high power, ultrafast pulses. As mentioned in Chapter 1, developing OPCPA systems in the long wavelength infrared (LWIR, 8-15 μ m) wavelength regime has attracted increasing attention. However, since this regime is still not well charted for OPCPA design particularly for generating high power and ultrafast pulses, the challenges and limitations would be better explored before we drill down into the experimental implementation. This chapter addresses the theoretical analysis and solution of the challenges in design of OPCPA in the LWIR regime. The major concerns are about the factors that limit the key performance of LWIR OPCPA, suitable analytic model of the key performance and new method that can be used to overcome the limitation. The studies on these aspects provide design guidelines and triggers solutions in the experimental development of LWIR OPCPA systems in this thesis.

The studies in this chapter are carried out by combining analytical modelling and scenario numerical simulations. First of all, numerical studies based on general wave coupling equations are carried out to address the limit of the gain bandwidth of an OPCPA in the LWIR regime. The studies disclose that a non-collinear phase matching geometry of OPCPA shall be adopted in the LWIR OPCPA. But the challenge still lies in finding an analytical way to minimize the phase mismatch in a non-collinear configuration. A new analytic gain bandwidth model for non-collinear OPCPA design is thus proposed first. Based on the model, the feasible non-collinear methods are further explored. An approach of ‘N-th order flatness of phase mismatch’ is proposed to address the challenges to obtain maximum gain bandwidth. As a result, an angular dispersive OPA (AD-OPA) method with is proposed. This new method could achieve

1 larger order number of flatness and higher gain bandwidth compared to the current
2 methods.

3 This chapter is arranged as follows. In section 2.2, we summarise the basic principle of
4 OPCPA, the wave coupling equations used for simulation studies, as well as the
5 common terminologies used in the rest parts of the thesis. Section 2.3 reports the
6 numerical studies which address the main challenges and the possible ways to
7 overcome the challenges in development of a LWIR OPCPA. In section 2.4, a new gain
8 bandwidth model is presented for qualitative analysis of the phase mismatches in non-
9 collinear geometry configuration. Section 2.5 presents the developed “Nth order
10 flatness of phase matching” design principle. Finally, the proposed AD-OPA method
11 is presented in Section 2.6, followed by a brief summary of this chapter. The materials
12 in Section 2.2 is mainly from Ref. [46], [47] and [49], and the rest are from the author’s
13 contributions.

14 **2.2 Basic Principle of OPCPA**

15 The optical parametric chirped-pulse amplification (OPCPA) is first proposed in 1992
16 [27]. Its idea was borrowed from merging the chirp pulse amplification (CPA) with the
17 optical parametric amplification (OPA). As in the CPA process the signal pulse in an
18 OPCPA is first stretched to match the pulse duration of the pump pulse, and then
19 amplified and compressed to generate a high energy, high peak power, ultrafast pulse.
20 OPCPA inherits the advantages of CPA and is favoured for broad gain bandwidth and
21 good thermal performance. It could also operate at various central wavelengths with
22 high wavelength tunability. Compared with OPA, OPCPA has larger scalability in both
23 energy and power. Nowadays the OPCPA systems have been demonstrated capable of
24 generating pulses with several tens of millijoule level pulse energy, more than ten-watt
25 level average power and few-cycle pulse duration. OPCPA has become a powerful

1 technique to build laser sources in applications such as the molecular spectroscopy [1-
2 7] and the strong field physics studies [8-10].

3 **2.2.1 Optical parametric amplification**

4 In an OPCPA system, the signal is amplified through the well-known optical
5 parametric amplification (OPA) process as illustrated as follows. The light wave to be
6 amplified is sent to propagate in the transparency medium and its electric field
7 $e(r, t)$ excites the polarization or an atomic dipole in the medium represented by $p(r, t)$.
8 By using the Taylor expansion, the polarization $p(r, t)$ could be written as [46],

$$9 \quad p(r, t) = \epsilon_0 \chi^{(1)} e(r, t) + \epsilon_0 \chi^{(2)} e^2(r, t) + \epsilon_0 \chi^{(3)} e^3(r, t) + \dots + \epsilon_0 \chi^{(N)} e^N(r, t) + \dots \quad (2.1)$$

10 where ϵ_0 is the permittivity in the vacuum and $\chi^{(N)}$ is the Nth order nonlinear
11 susceptibility. The OPA is achieved through the second order term of the polarization
12 which couples the energy among a high energy pump wave, a low energy signal wave
13 and an idler wave. Although the principle of OPA is different from the stimulate
14 emission (as shown in Fig.2.1(a)), and the energy conversion process involved in OPA
15 can still be described by a virtual energy-level diagram as shown in Fig.2.1(b) where
16 light waves are seen as their particle duals. In this process, the atom first absorbs a
17 pump photon $\hbar\omega_p$ and jumps to a high virtual energy level (Fig.2.1(b)), then it is
18 stimulated by a signal photon $\hbar\omega_s$, and the decay of the excited atom to its ground state
19 simultaneously generates two photons respectively with energy $\hbar\omega_s$ and $\hbar\omega_i =$
20 $\hbar\omega_p - \hbar\omega_s$. The energy of the pump photon $\hbar\omega_p$ is thus converted to the signal photon
21 $\hbar\omega_s$ and the idler photon $\hbar\omega_i$. The energy conservation law spells out the following
22 relationship between the frequencies of the pump, signal and idler waves,

$$23 \quad \omega_p - \omega_s - \omega_i = 0 \quad (2.2)$$

1 where ω_l ($l = p, s, i$) are the frequencies of the pump, signal and idler waves.

2
3 Fig.2.1. Comparison between the (a) Stimulated emission and (b) Optical parametric
4 amplification

5 In addition to the energy conservation law, the kinetic conservation law should also be
6 fulfilled in the OPA process. The kinetic conservation law is written as [46,47],

$$7 \quad \vec{k}_p(\omega_p) - \vec{k}_s(\omega_s) - \vec{k}_i(\omega_i) = \vec{0} \quad (2.3)$$

8 where $\vec{k}_l(\omega_l)$, ($l = p, s, i$) are the wavevectors of the pump, signal and idler waves.
9 This condition is also called the phase matching condition [46]. In an OPA/OPCPA,
10 the three interacting waves are normally non-monochromatic waves. The phase
11 matching condition could only be fulfilled for one set of frequency components
12 $(\omega_{p0}, \omega_{s0}, \omega_{i0})$ due to material dispersion, but for other frequencies $(\omega_p, \omega_s, \omega_i)$, the
13 phase matching condition may not be met and a phase mismatch is defined as [46,47],

$$14 \quad \Delta\vec{k} = \vec{k}_p(\omega_p) - \vec{k}_s(\omega_s) - \vec{k}_i(\omega_i) \quad (2.4)$$

15 This is also referred to as the wave vector mismatch. By using the direction of the
16 wavevector of the pump wave $\vec{k}_p$ as a reference, the phase mismatch could be
17 decomposed into two components on the directions which are parallel and

1 perpendicular to $\vec{k}_p$ respectively. The two components of the phase mismatch can be
 2 written as [47,48],

$$\begin{aligned} \Delta k_{\parallel} &= k_p(\omega_p) - k_s(\omega_s) \cos \rho_s - k_i(\omega_i) \cos \rho_i \\ \Delta k_{\perp} &= k_s(\omega_s) \sin \rho_s + k_i(\omega_i) \sin \rho_i \end{aligned} \quad (2.5)$$

5 where ρ_s and ρ_i are the angles between the wavevector $\vec{k}_p$ and $\vec{k}_s$, $\vec{k}_i$ respectively as
 6 shown in the Fig.2.2.

7
 8 Fig.2.2. Definitions of (a) the non-collinear angle ρ_i , θ , pump-surface normal angle α ,
 9 coordinate $r = (x, y, z)$ and (b) coordinate $r_l = (x_l, y_l, z_l)$. It should be noted that z points
 10 to the direction of $\Delta\vec{k}$ and z_l points to the direction of $\vec{k}_l$. Both y and y_l are located in the
 11 surface formed by the wave vectors.

12 2.2.2 Coupled wave equations

13 In an OPA process, new signal and idler photons could be generated by the atomic
 14 dipoles located at different positions along the light propagation trajectory in the
 15 nonlinear medium. These dipoles act as a number of light sources which radiate signal
 16 or idler waves in the form of the dipole radiation pattern. The signal or idler waves
 17 emitted at various positions are constructively interfered at the forward direction of
 18 propagation when the phase relation between three interacting waves is correct. As a

1 result, the energy of the signal or the idler wave is converted from the pump wave and
 2 scaled up along propagation. In general, this process can be described in general by the
 3 coupled wave equations. Assuming that the pump, signal and idler waves are non-
 4 monochromatic waves, the coupled wave equations are written as [46],

$$\begin{aligned}
 & \frac{\partial}{\partial z} A_i(r, \rho_i, \theta, \omega_{iw}) \\
 &= \sum_u \sum_v \frac{i\pi^2 \omega_{iw}^2}{k_i(\omega_{iw})c^2 \cos(\rho_i - \theta)} d_{\text{eff}} A_p(r, \rho_p, \theta, \omega_{pu}) A_s^*(r, \rho_s, \theta, \omega_{sv}) e^{-i\Delta k z} \\
 & \frac{\partial}{\partial z} A_s(r, \rho_s, \theta, \omega_{sv}) \\
 &= \sum_u \sum_w \frac{i\pi^2 \omega_{sv}^2}{k_s(\omega_{sv})c^2 \cos(\rho_s - \theta)} d_{\text{eff}} A_p(r, \rho_p, \theta, \omega_{pu}) A_i^*(r, \rho_i, \theta, \omega_{iw}) e^{-i\Delta k z} \\
 & \frac{\partial}{\partial z} A_p(r, \rho_p, \theta, \omega_{pu}) \\
 &= \sum_v \sum_w \frac{i\pi^2 \omega_{pu}^2}{k_p(\omega_{pu})c^2 \cos(\rho_p - \theta)} d_{\text{eff}} A_s(r, \rho_s, \theta, \omega_{sv}) A_i(r, \rho_i, \theta, \omega_{iw}) e^{i\Delta k z}
 \end{aligned} \tag{2.6}$$

12 where $A_l(r, \rho_l, \theta, \omega_l)$, $l = p, s, i$ denotes the amplitude of each frequency component of
 13 the pump, signal and idler waves that should fulfil the slowly varying amplitude
 14 approximation [46],

$$\left| \frac{\partial^2}{\partial z^2} A_l(r, \omega_l) \right| \ll \left| k_l(\omega_l) \frac{\partial}{\partial z} A_l(r, \omega_l) \right| \tag{2.7}$$

16 r is the coordinate of positions in space. Index u, v, w represent the longitudinal mode
 17 number of the pump, signal and idler waves, respectively. d_{eff} is the effective nonlinear
 18 coefficient. Δk is the module of the wave vector mismatch. ρ_l is the angle of the
 19 wavevectors of three waves with respect to the pump wavevector, and θ is the angle

1 between the $\vec{\Delta k}$ and the pump wavevector. The definitions of these two angles are
 2 illustrated in Fig.2.2. Here, these two angles are considered so that the coupled wave
 3 equations can be applied when OPA/OPCPA adopts non-collinear geometry
 4 configurations.

5 A further assumption on the un-depleted energy of the pump wave helps solve Eq.2.6
 6 with the following solution [46,47],

$$\begin{aligned}
 & A_s(r, \rho_s, \theta, \omega_{sv}) \\
 & = A_{s0} \left\{ \cosh[g(z - y \tan(\theta + \alpha))] \right. \\
 & \quad \left. + i \frac{\Delta k}{2g} \sinh[g(z - y \tan(\theta + \alpha))] \right\} e^{-i \frac{\Delta k}{2}(z - y \tan(\theta + \alpha))} \\
 & A_i(r, \rho_i, \theta, \omega_{iw}) = \frac{\Gamma_i}{g} A_{s0}^* \sinh[g(z - y \tan(\theta + \alpha))] e^{-i \frac{\Delta k}{2}(z - y \tan(\theta + \alpha))} \quad (2.8)
 \end{aligned}$$

11 where α is the angle between the pump wave vector and the normal of the surface
 12 where the light is incident on the nonlinear medium. Based on the solutions, the
 13 intensity evolution of the signal and idler waves along with the propagation in the
 14 OPCPA can be analysed, and the performance of the OPCPA can also be modelled and
 15 studied.

16 2.2.3 Key performance indicators

17 The principle of OPCPA reveals several key performance indicators of an OPCPA
 18 system including the parametric gain, the conversion efficiency and the gain bandwidth.
 19 These indicators evaluate how much energy could be converted from the pump and
 20 how many signal components could be amplified in the OPCPA of concern. They are
 21 also used as the guidelines to determine the design parameters such as the crystal length
 22 and angle, the intensity, polarization and beam size of the input light beams, etc.

1 The parametric gain of each signal frequency component is defined as its output-input
 2 intensity ratio. Considering a nonlinear medium with length L , the parametric gain G
 3 can be derived from Eq.2.8 and given by,

$$4 \quad G = 1 + \left(1 + \frac{\Delta k^2}{4g^2}\right) \sinh^2 \left(\frac{gL}{\cos(\theta + \alpha)}\right), \quad (2.9)$$

5 Here, g is defined as the small signal gain [46,47] given by,

$$6 \quad g = \sqrt{\Gamma^2 - (\Delta k/2)^2} \quad (2.10)$$

8 where Γ^2 is proportional to the square of the nonlinear coefficient d_{eff} and the pump
 9 intensity I_p . When the small signal gain is large so that $\frac{gL}{\cos(\theta + \alpha)} \gg 1$, the parametric
 10 gain approximately equals to:

$$11 \quad G \approx \frac{1}{4} e^{\frac{2gL}{\cos(\theta + \alpha)}} \quad (2.11)$$

12 According to Eq.2.10 and Eq.2.11, the parametric gain is determined by the following
 13 parameters: the phase mismatch Δk , the nonlinear coefficient d_{eff} , the crystal length L
 14 and the pump intensity I_p . The parametric gain exponentially increases with the
 15 nonlinear coefficient, the crystal length and the pump intensity, and it decreases with
 16 the phase mismatch.

17 However, in a general case where the pump beam is depleted during propagation, the
 18 variation of the parametric gain against the propagation distance is rather complicated.
 19 It is known that the variation of the parametric gain can be roughly divided into three
 20 regions [47], including the linear region, the exponential region and the saturation

1 region. As shown in Fig.2.3, the signal energy increases approximately linearly with
2 the length L in the linear region. In this region, since the photon flux of the signal is far
3 smaller than the pump photon flux, the energy conversion is slow and less efficient. In
4 the exponential region, the parametric gain exponentially increases with the length L .
5 Energy conversion rapidly occurs between massive pump and signal photon until
6 saturation is reached. As last, in the saturation region, the energy level of the signal is
7 comparable to the level of the pump, and energy start flowing back from most of the
8 signal frequency components to the pump. Therefore, the total parametric gain
9 decreases.

Fig.2.3. The variation of the parametric gain in three different regions [47].

12 The conversion efficiency of OPCPA is defined as the ratio between the amplified
13 signal energy and the total input energy. It indicates how many percentages of pump
14 energy could be converted to the signal pulses. Similar to the parametric gain, the
15 conversion efficiency is known increasing with the pump intensity, the crystal length
16 and the nonlinear coefficient but decreasing with the phase mismatch [47]. It is also
17 shown by previous research that the conversion efficiency can be increased with the
18 input signal-to-pump energy ratio [47]. When the signal-to-pump energy ratio is higher,
19 the increasing number of signal photons can also increase the consumption of the pump
20 photons. Therefore, the energy extraction from the pump beam becomes more efficient.

1 The gain bandwidth is another key performance indicator of an OPCPA system. It
2 characterizes how much signal frequency components are involved in the efficient
3 amplification process. As shown in Eq.2.10 and Eq.2.11, the parametric gain of the
4 signal is determined by the wave vector mismatch. For the central frequency of signal
5 where the wave vector mismatch vanishes, the parametric gain reaches its maximum
6 value. However, the parametric gain decreases for other signal frequencies which
7 deviate from its central value due to the wave vector mismatch. Therefore, the
8 parametric gain can be considered as the function of the signal frequency, denoted by
9 $G(\omega_s)$. The FWHM of the parametric gain is defined as the gain bandwidth, denoted
10 by $\Delta\Omega$. Since the gain bandwidth originates from the parametric gain, it is also related
11 to the parameters such as the phase mismatch, the nonlinear coefficient, the crystal
12 length L and the pump intensity I_p . Detailed studies of these relation will be introduced
13 in section 2.4.

14 **2.3 Challenges in development of OPCPA in LWIR regime**

15 In the previous section, a general theory of OPCPA are demonstrated. The theory
16 shows that the LWIR OPCPA is achievable if the energy and kinetic conservation laws
17 are both fulfilled. However, there are still several challenges in the development of an
18 OPCPA in the LWIR regime to achieve high energy, high efficiency and broad gain
19 bandwidth. In this section, the challenges are addressed by numerical studies that are
20 based on directly on the general theory of OPCPA, and the studies also reveal some
21 possible solutions to overcome the challenges.

22 **2.3.1 Limitation in gain bandwidth**

23 According to the general theory introduced in previous section, efficient frequency
24 conversion from pump to signal could be achieved only when the energy and kinetic
25 conservation laws are fulfilled. The kinetic conservation law, which is also called the
26 phase matching condition, is normally fulfilled for one signal frequency component

1 due to the material dispersion. For other frequency components of signal, the phase
 2 mismatch is written as Eq.2.4. By expanding the wave numbers $k_l(\omega_l)$ ($l = p, s, i$) into
 3 their first order of Taylor series, the phase mismatch modulus is obtained as [47],

$$\begin{aligned}
 \Delta k &= k_p(\omega_p) - k_s(\omega_s) - k_i(\omega_i) \\
 &= \left(\frac{1}{v_{gp}} - \frac{1}{v_{gi}} \right) \Delta\omega_p + \left(\frac{1}{v_{gi}} - \frac{1}{v_{gs}} \right) \Delta\omega_s
 \end{aligned} \tag{2.12}$$

5 where v_{gl} , ($l = p, s, i$) are the group velocity of the pump, signal and idler waves, and
 6 they are obtained by [49],

$$v_{gl}(\omega) = \frac{c}{n_l(\omega) + \omega_l \frac{\partial n_l}{\partial \omega_l}} \tag{2.13}$$

8 Seen from Eq. 2.13, if the $\frac{\partial n_p}{\partial \omega_p}$ or $\frac{\partial n_s}{\partial \omega_s}$ term is much higher than $\frac{\partial n_i}{\partial \omega_i}$ of idler, one of the
 9 group velocity mismatch terms $\frac{1}{v_{gp}} - \frac{1}{v_{gi}}$ and $\frac{1}{v_{gs}} - \frac{1}{v_{gi}}$ is large. As a result, the phase
 10 mismatch is more sensitive to the frequency disturbance $\Delta\omega_p$ and $\Delta\omega_s$. This situation
 11 occurs when the wavelength of pump or signal approaches the wavelength
 12 corresponding to the band gap. The situation could be verified by the simulation shown
 13 in Fig.2.4. In this simulation, we calculate the sensitivity of the phase mismatch $\frac{d\Delta k}{d\Delta\omega_s}$
 14 under a small disturbance on the signal frequency. Here we take the GaSe crystal as
 15 the nonlinear medium of OPCPA. The wavelength of the idler is fixed in LWIR regime
 16 as 9 μ m. Several sensitivity values are calculated with 22 different combinations of the
 17 pump and signal wavelengths. The central wavelength of the pump varies from 900nm
 18 to 3000nm. Correspondingly, the central wavelength of signal changes from 1000nm
 19 to 4500nm. The calculated sensitivities are plotted into a curve as in Fig.2.4. The figure
 20 shows that the sensitivity increases dramatically when the difference between the pump

1 wavelength and the wavelength corresponding to the band gap of GaSe is smaller than
 2 500nm. This result is in accordance with our expectation.

3
 4 Fig.2.4. Sensitivity of phase mismatch. Here λ_{bg} is the wavelength that corresponds to band
 5 gap of GaSe crystal.

6 Actually, the situation mentioned above is exactly the case for design of an OPCPA in
 7 LWIR regime. In order to develop a LWIR OPCPA with high energy and power, a
 8 powerful pump laser is always required. Nowadays, this role could be served by the
 9 technically advanced $\sim 1\mu\text{m}$ and $\sim 0.8\mu\text{m}$ laser sources. However, these wavelengths of
 10 pump lasers are very close to those corresponding to the band gap of the normally used
 11 nonlinear medium materials, for example, 2.1eV band gap of the GaSe and 2.45eV
 12 band gap of ZGP. The difference between the pump wavelength and the wavelength of
 13 band gap is smaller than 500nm. Therefore, the material dispersion $\frac{\partial n_p}{\partial \omega_p}$ is much larger
 14 than $\frac{\partial n_i}{\partial \omega_i}$ of the idler if the signal is targeted in LWIR regime, as shown in the Fig.2.5(b).
 15 Moreover, since the wavelength of the LWIR idler is small, the signal wavelength is
 16 also closed to the pump wavelength. The dispersion $\frac{\partial n_s}{\partial \omega_s}$ of signal is also larger than the
 17 $\frac{\partial n_i}{\partial \omega_i}$ of idler. Therefore, the phase mismatch is more sensitive to the frequency
 18 disturbance $\Delta\omega_p$ and $\Delta\omega_s$ in case of LWIR OPCPA. Compared to the case of OPCPA
 19 in shorter wavelength regimes, fewer signal frequency components can be efficiently

1 amplified in LWIR regime due to the large phase mismatch, this results in a smaller
2 gain bandwidth of LWIR OPCPA.

3 This situation is verified by another simulation shown in Fig.2.5 (a). In this simulation,
4 we numerically compare the gain bandwidths of the LWIR OPCPAs driven by different
5 pump sources. We use a 1mm thick GaSe crystal with Type I (ooe) phase match and
6 the collinear geometry in the calculation. The wavelength of the idler is set to be 9 μ m.
7 The pump wavelength is decreased to approach the band gap wavelength from 3 μ m to
8 1 μ m with 0.5 μ m in each step. In each case, the gain bandwidths are calculated with
9 pump intensity varying from 0 to 5GW/cm². The simulation results are summarized in
10 Fig.2.5 (a).

11

12 Fig.2.5. (a) Gain bandwidth of the 9 μ m GaSe based OPCPA under different pump
13 wavelength changing from 1 μ m to 3 μ m. (b) Dispersion curve of the GaSe.

14 The simulation results show that the gain bandwidth of the LWIR OPCPA drops when
15 the pump wavelength approaches the wavelength of the band gap (590nm). Therefore,
16 the large GVM between pump/signal and idler pulse causes the limitation of the gain
17 bandwidth of the LWIR OPCPA.

18 In order to enhance the gain bandwidth, one possible solution is to use longer
19 wavelength pump lasers. For example, the Ho/Tm doped laser at ~2 μ m, the Cr:ZnS or
20 Cr:ZnSe operating at 2.4 μ m and the Fe doped laser at 4 μ m. These wavelengths are far

away from the wavelength of the band gap. Another way to overcome the gain bandwidth limitation is using the large band gap material as the nonlinear medium. This kind of material should have large transparency window covering both NIR and LWIR regime. It should also support the frequency down-conversion from the short wavelength pump to the LWIR output. It is also noticed that the GVM between these two pair of waves could be mitigated by introducing non-collinear angle between pump and idler as well as signal and idler. The gain bandwidth of the OPCPA could also be enhanced.

2.3.2 Parametric phase distortion: the OPP

In the case of LWIR OPCPA, another challenge that should also be taken into consideration lies with increasing phase distortion introduced by the optical parametric phase. According to the solution of coupled wave equation (Eq. 2.8), the total phase of the output signal wave can be calculated by,

$$\begin{aligned} \phi_s = \phi_{s0} - k_s(\omega_s) \frac{L}{\cos(\theta + \alpha)} - \frac{\Delta k}{2} \frac{L}{\cos(\theta + \alpha)} \\ + \tan^{-1} \left[\frac{\Delta k}{2g} \tanh \left(\frac{gL}{\cos(\theta + \alpha)} \right) \right] \end{aligned} \quad (2.14)$$

The total phase ϕ_s includes three different terms, the ϕ_{s0} term is the initial phase term, the $k_s(\omega_s) \frac{L}{\cos(\theta + \alpha)}$ term is the phase generated via wave propagation, the rest of the total phase is the so called optical parametric phase (OPP). As shown in Eq.2.14, the OPP is determined by the phase mismatch. When the phase matching condition is fulfilled, the OPP term will be vanished. This spectral phase distortion is first identified by Ross et al [50]. Later on, the features OPP and its compensation are further studied in theory [51] and experiment [52].

Fig.2.6. The plot of (a) the OPP evolution with the propagation distance L and the comparison between OPP and material dispersion at (b) $L=2\text{mm}$, (c) $L=4\text{mm}$, (d) $L=6\text{mm}$, (e) $L=8\text{mm}$.

In order to demonstrate the effect of the OPP, we perform the simulation in the NIR OPCPA as shown in Fig.2.6. Here, we consider an OPCPA process in a BBO crystal. The thickness of the BBO crystal is chosen to be 1cm so that the saturation region of the OPCPA is also included in the simulation. The central wavelengths of the pump and the signal waves are 532nm and 800nm, respectively. Both the pump and signal pulses have the 2nd order super-Gaussian pulse shape. The pulse duration of the pump pulse is set as 1ps and the signal pulse width is also stretched to 1ps. The phase matching type is the Type I, ooe with the 22.1° phase matching angle. The pump intensity and the signal intensity are chosen as 15 GW/cm² and 4.5kW/cm² respectively.

1 In the simulation, only group delay dispersion (GDD) of the material is considered.
2 The variation of the optical parametric phase of each signal frequency component along
3 the propagation distance is shown in Fig.2.6 (a). The spectral shape of signal, total
4 spectral phase, propagation induced phase and OPP are calculated at 2mm, 4mm, 6mm
5 and 8mm positions, as exhibited in Fig.2.6 (b)-(e).

6 It is seen from the simulation result shown in Fig.2.6 (a) that the OPP is kept to be zero
7 at the central-frequency of the signal since the phase matching condition is fulfilled.
8 For the frequency components located at both wings of the spectrum, the absolute OPP
9 value of these components increases with propagation distance due to the phase
10 mismatch. Detailed comparisons between the propagation induced phase and the OPP
11 are shown in Fig.2.6 (b)-(e). The curve of the propagation induced phase obtained
12 before the position at 6mm has the perfect parabola shape which results from the GDD
13 of the material dispersion. The OPP curve also has a parabola shape but introduces a
14 different sign of GDD compared with the propagation induced phase. Compared with
15 the GDD of the material, the GDD introduced by the OPP is much smaller. As a result,
16 the total GDD is slightly smaller than the GDD of the material. However, when the
17 propagation distance of the signal is larger than 6mm, the OPP as well as the total phase
18 curve start to be severely distorted. The distortion mainly comes from the complex
19 phase shift in the saturation region of the OPCPA process. It can be shown that the gain
20 saturation of the signal is reached with the energy reduction occurred at the center of
21 the signal spectrum. In short, the simulation results show that the OPP should be taken
22 into consideration in dispersion compensation of OPCPA, especially when the OPCPA
23 is operated in the saturation regime.

24 Moreover, we found that the GDD introduced by the OPP becomes more significant
25 compared with the material dispersion in the long wavelength mid-IR OPCPA, when
26 the wavelengths of the pump and signal are closed to each other. Fig.2.7 (a) and (b)
27 show the results of another simulation study comparing the OPP of an OPCPA in short

IR wavelength regime to its counter partner in long wavelength mid-IR regime. In this simulation, we calculate the extra GDD introduced by the OPP in 5mm thick BBO crystal and 5mm thick LGS crystal. The center wavelength of the pump pulse is fixed as $1.03\mu\text{m}$ and $15\text{GW}/\text{cm}^2$ pump intensity and 1ps pulse duration are considered in both cases. The input signal pulse is stretched to 1ps with additional 20000fs^2 initial GDD. For the BBO case, the signal wavelength varies from $1.5\mu\text{m}$ to $1.9\mu\text{m}$, which corresponds to an idler wavelength decreases from $3.3\mu\text{m}$ to $2.2\mu\text{m}$. For the LGS case, the signal wavelength varies from $1.148\mu\text{m}$ to $1.244\mu\text{m}$, which corresponds to an idler wavelength decreases from $10\mu\text{m}$ to $6\mu\text{m}$.

Fig.2.7. OPP introduced GDD in (a) the BBO crystal (b) the LGS crystal. (c) The comparison between OPP introduced GDD and the material dispersion added GDD for $9\mu\text{m}$ OPCPA in the LGS crystal is also included.

In both cases, the GDD by factoring into OPP increases with both propagation distance and idler wavelength. For near IR (NIR) OPCPA or short wavelength IR (SWIR) OPCPA, the OPP induced GDD is much smaller than the initial chirp. This phenomenon is also observed in an 1090nm OPCPA [52] and 2108nm OPCPA [53] in which the OPP-induced GDDs equal to 30fs^2 and 243fs^2 GDD respectively. However, the amount of the GDD generated in the LGS OPCPA at longer wavelength is over 10 times of the GDD introduced in the BBO case. For a particular situation among all LGS cases where the idler wavelength is chosen as $9\mu\text{m}$ (Fig.2.7(c)), the proportion of the OPP induced GDD in the total amount is as large as 30%, about 5 times larger than the

1 GDD of the material. Therefore, the OPP is an important factor limiting the
2 performance of OPCPA in the LWIR regime, and it cannot be ignored in the dispersion
3 management of the long wavelength mid-IR OPCPA. If the OPP induced GDD is not
4 considered in the chirp compensation, the $>10000\text{fs}^2$ residual GDD is large enough to
5 destroy a few-cycle pulse and decreases the peak intensity of the pulse to less than half
6 of its maximum reached value.

7 **2.4 Gain bandwidth model for non-collinear OPCPA**

8 As shown in section 2.3, the challenge to develop an OPCPA in LWIR regime lies with
9 its limited gain bandwidth. Three solutions include the use of longer wavelength pump
10 laser, the use of larger bang gap nonlinear medium and the use of non-collinear
11 geometry. Among these solutions, the use of the non-collinear geometry is the most
12 promising way as it leverages currently available high-power pump sources and
13 available nonlinear mediums. In this section, we focus on studying the non-collinear
14 method to develop OPCPA in LWIR regime and leave other two possible solutions to
15 be discussed in the next chapters. Three new development are obtained in the thesis,
16 respectively regarding the gain bandwidth model of non-collinear OPCPA, the N-th
17 order flatness concept proposed for optimizing the gain bandwidth, and the angular
18 dispersion non-collinear OPA technique to achieve broad gain bandwidth.

19 **2.4.1 Non-collinear OPA/OPCPA**

20 The non-collinear OPA/OPCPA gets its name because of the non-collinear angle
21 introduced between the pump, signal and idler. By introducing the non-collinear angle,
22 the projected group velocity of signal and idler pulses could be identical along the
23 signal direction, as shown in Fig.2.8 (b). The temporal profiles overlap between the
24 signal and idler pulses will always be kept during the interaction. The GVM is thus
25 mitigated and the gain bandwidth could be enhanced.

Fig.2.8. Diagram of the (a)Collinear geometry; (b)Non-collinear geometry; (c)Angular dispersive geometry

The pioneering contribution of the non-collinear OPA (NOPA) is the work reported by Gale et al. [54] in 1995 which generates nanojoule-level, 13fs pulses. Following the Gale's work, several limitations of the NOPA, such as the pulse-front mismatch between pump and signal [55], and the angular dispersion of the idler wave [56] are broken through. The NOPA system can now provide ~1-octave amplification bandwidth concept which is enough to generate sub-two-cycle pulses.

Based on the NOPA, the gain bandwidth could be further enhanced by introducing an angular-dispersive signal beam as shown in Fig.2.8(c). Compared with the NOPA (Fig.2.8(b)), the phase matching condition now fulfilled not only for the central frequency of signal but also for the frequency components within a small range around it. Therefore, the phase matching is achieved in broader wavelength range. The gain bandwidth thus could be enhanced. The way of using angular dispersive signal in NOPA has been demonstrated in both theory [57,58] and experiment [59,60].

However, although the non-collinear OPAs have been demonstrated for many years, there is still not a simple and direct gain bandwidth model which could connect the non-collinear angle with the gain bandwidth. It thus hinders the qualitatively and quantitatively analysis the performance of the non-collinear OPA/OPCPA. Also, from design point of view, the model is required for conveniently choosing design

1 parameters of non-collinear OPA/OPCPA to obtain a desired gain bandwidth.
 2 Therefore, a new gain bandwidth model which is suitable for the non-collinear
 3 OPA/OPCPA should be proposed.

4 **2.4.2 Principle of analytic model for gain bandwidth**

5 As shown in Eq.2.10 and Eq.2.11, the parametric gain of the signal is determined by
 6 the wave vector mismatch. For the central frequency of signal where the wave vector
 7 mismatch vanishes, the parametric gain reaches its maximum value. However, the
 8 parametric gain decreases for other signal frequencies which deviate from its central
 9 value due to the wave vector mismatch. Therefore, the parametric gain could be
 10 considered as the function of the signal frequency, denoted by $G(\omega_s)$. The FWHM of
 11 the parametric gain is defined as the **gain bandwidth**, denoted by $\Delta\Omega$.

12 In the non-collinear OPA/OPCPA, the phase mismatch should be considered as a
 13 vector function. The gain bandwidth is tightly related with the vectoral phase mismatch
 14 $\Delta\vec{k}$. Therefore, the first and foremost thing for deriving a gain bandwidth model is
 15 calculating both the module and the direction of the $\Delta\vec{k}$. The challenge lies with
 16 determining the wave vector $\vec{k}_i$ of the generated idler wave. This needs to solve the
 17 vectoral-form coupled wave equations, and results in a complicate model. Traditional
 18 solution is to consider a vanishing transverse component of $\Delta\vec{k}$, which is demonstrated
 19 in the previous models [48]. Therefore, the complex vectoral equations will be
 20 simplified to a scalar one for deriving the gain bandwidth model. Fig.2.2 suggests this
 21 condition could be written as

$$22 \quad \Delta k_{\perp} = k_s \sin \rho_s + k_i \sin \rho_i = 0 \quad (2.15)$$

23 However, we find that this condition loses its validity when numerically solving the
 24 coupled pulsed-wave equations with boundary condition specified in the Ref.[61],

1 where the component of the $\vec{\Delta k}$ at the input surface which is transversal to the surface
 2 normal is considered to be zero. This condition is written as,

$$3 \quad k_p - k_s \sin(\rho_s + \alpha) - k_i \sin(\rho_i + \alpha) = 0 \quad (2.16)$$

4 where α is the angle between $\vec{k}_i$ and the surface normal as shown in Fig.2.2. It deviates
 5 from the previous condition Eq.2.15 when $\alpha \neq 0$. The phase matching geometries
 6 under these two conditions are compared in Fig.2.9(a). According to condition in
 7 Eq.2.15, the wave vector mismatch $\vec{\Delta k}_1$ is parallel to the $\vec{k}_p$. On the contrary, $\vec{\Delta k}_2$
 8 which corresponds to the condition in Eq.2.16 is parallel to the surface normal. It is
 9 also obvious that the module of $\vec{\Delta k}_2$ is smaller than $\vec{\Delta k}_1$. Therefore, we can expect that
 10 the gain bandwidths calculated under both conditions are also different.

11 To see this clearly, a simulation is done. In this simulation, we consider an OPA in
 12 10mm thick GaSe crystal with 2060 nm pump wavelength, 10GW/cm² pump intensity
 13 and 4° non-collinear angle ρ_s of the signal. The gain bandwidths calculated under
 14 condition Eq.2.11 and Eq.2.14 are denoted as $\Delta\Omega_1$ and $\Delta\Omega_2$, respectively. Here, $\Delta\Omega_1$ is
 15 calculated via using the model in Ref.[48] and $\Delta\Omega_2$ is obtained by solving numerically
 16 the coupled wave equations with boundary condition in Ref.[61]. In the numerical
 17 calculation of $\Delta\Omega_2$, the pump wave is assumed to be a monochromatic wave and its
 18 pulse energy is assumed un-depleted for a fair comparison with $\Delta\Omega_1$. We consider three
 19 different signal central wavelength, 3.1 μm , 2.6 μm and 2.2 μm , which correspond to
 20 the 6 μm , 10 μm and 15 μm idler output respectively. In each case, the values of angle
 21 α which change from -20° to 20° and the discrepancy between $\Delta\Omega_1$ and $\Delta\Omega_2$ is
 22 evaluated simultaneously. Fig.2.9(b) shows that the difference between $\Delta\Omega_1$ and $\Delta\Omega_2$
 23 increases with the α value. Also, for the long wavelength mid-IR cases where idler
 24 wavelength equals to 10 μm and 15 μm , the discrepancy could be larger than 10% when
 25 α equals to $\pm 20^\circ$. Therefore, for long wavelength OPA/OPCPA design, the traditional

gain bandwidth model is unreliable. A new, more general model is expected without applying the condition in the Eq.2.11.

Fig.2.9. (a) Comparison between two different constraints on the direction of the idler wave vector (Eq.2.11 and Eq.2.14); (b) Gain bandwidth with different pump-surface normal angle α ranging from $\alpha = -20^\circ$ to 20° . The comparison has been made between three cases where, case 1 (blue dashed line): central wavelength of the pump $\lambda_{p0} = 2.06\mu\text{m}$ and signal $\lambda_{s0} = 3.1\mu\text{m}$; case 2 (red dash-dotted line): $\lambda_{p0} = 2.06\mu\text{m}$ and signal $\lambda_{s0} = 2.6\mu\text{m}$; case 3 (black line): $\lambda_{p0} = 2.06\mu\text{m}$ and signal $\lambda_{s0} = 2.2\mu\text{m}$;

On the contrary to the previous constrain, we propose that the direction of the wave vector $\vec{k}_i$ of the idler wave could be determined in another way. Since the directions of wave vectors of pump and signal are fixed, the direction of $\vec{k}_i$ and the direction of the wave vector mismatch $\Delta \vec{k}$ have a one-to-one correspondence. Here, we assume that the direction of $\Delta \vec{k}$ falls in the direction where its modulus Δk is minimized. Under this consideration, the direction of $\vec{k}_i$ is also determined so that the parametric gain could also be calculated via Eq.2.11 as a function of the wave vector mismatch. A gain bandwidth model, which could provide at least upper-limit gain bandwidth evaluation could thus be derived according to its definition.

1 The principle of determining the direction of the wave vector $\vec{k}_i$ is illustrated in
 2 Fig.2.10(a) and (b) where the idler wave is considered as an ordinary wave and extra-
 3 ordinary wave, respectively. For an ordinary idler wave, before we determine its wave
 4 vector direction, the $\vec{k}_i$ could falls on any position of the circle centred at O_i with radius
 5 k_i . The minimum Δk is obtained when the circle centred at O_p with a radius of Δk is
 6 tangent with the circle O_i . In this case, the $\vec{k}_i$ is determined to be parallel with the $O_i O_p$
 7 direction. For an extraordinary idler wave, the $\vec{k}_i$ could falls on any position of the
 8 elliptical centered at O_i . With the same procedure, the minimum Δk could be achieved
 9 when the circle centered at O_p with a radius of Δk is tangent with the elliptical O_i . The
 10 obtained minimum Δk implies that the parametric gain of each signal frequency
 11 component could reach its largest possible value after propagating the same length.
 12 Therefore, the maximum reachable gain bandwidth can be predicted.

14 Fig.2.10. Diagrams on the determination of idler wave vector and the wave vector mismatch,
 15 with considering (a) idler is an ordinary wave; (b) idler is an extraordinary wave.

16 **2.4.3 Proposed gain bandwidth model**

17 According to the introduced principle, the direction of the idler wave vector $\vec{k}_i$ which
 18 is characterized by its non-collinear angle ρ_i could be determined by the following
 19 equations,

$$\rho_i = \begin{cases} \tan^{-1} \left(-\frac{k_s \sin \rho_s}{k_p - k_s \cos \rho_s} \right) & \text{if idler is an ordinary wave} \\ \rho_{i0} - \frac{f(\rho_{i0})}{\left. \frac{\partial f}{\partial \rho_i} \right|_{\rho_{i0}}} & \text{if idler is an extraordinary wave} \end{cases} \quad (2.17)$$

where ρ_{i0} , $f(\rho_{i0})$ and $\left. \frac{\partial f}{\partial \rho_i} \right|_{\rho_{i0}}$ can be calculated by,

$$\begin{aligned} \rho_{i0} &= \tan^{-1} \left(-\frac{k_s(\omega_{s0}) \sin \rho_s}{k_p - k_s(\omega_{s0}) \cos \rho_s} \right) \\ f(\rho_{i0}) &= k_p \left(k_i(\rho_{i0}) \sin \rho_{i0} - \left. \frac{\partial k_i}{\partial \rho_i} \right|_{\rho_{i0}} \cos \rho_{i0} \right) \\ &\quad - k_s \left(k_i(\rho_{i0}) \sin(\rho_{i0} - \rho_s) - \left. \frac{\partial k_i}{\partial \rho_i} \right|_{\rho_{i0}} k_s \cos(\rho_{i0} - \rho_s) \right) \\ &\quad + k_i(\rho_{i0}) \left. \frac{\partial k_i}{\partial \rho_{i0}} \right|_{\rho_{i0}} \\ \left. \frac{\partial f}{\partial \rho_i} \right|_{\rho_{i0}} &= \left(\left. \frac{\partial k_i}{\partial \rho_i} \right|_{\rho_{i0}} \right)^2 + k_i(\rho_{i0}) \left. \frac{\partial^2 k_i}{\partial \rho_i^2} \right|_{\rho_{i0}} \\ &\quad + k_p \left[\left(k_i(\rho_{i0}) - \left. \frac{\partial^2 k_i}{\partial \rho_i^2} \right|_{\rho_{i0}} \right) \cos \rho_{i0} + 2 \left. \frac{\partial k_i}{\partial \rho_i} \right|_{\rho_{i0}} \sin \rho_{i0} \right] \\ &\quad - \left[\left(k_i(\rho_{i0}) - \left. \frac{\partial^2 k_i}{\partial \rho_i^2} \right|_{\rho_{i0}} \right) k_s \cos(\rho_{i0} - \rho_s) - 2 \left. \frac{\partial k_i}{\partial \rho_i} \right|_{\rho_{i0}} k_s \sin(\rho_{i0} - \rho_s) \right] \end{aligned} \quad (2.18)$$

where detailed calculation of the non-collinear angle ρ_i could refer to the Ref.[62]. After the ρ_i is determined, the wave vector mismatch and its components $\Delta k_{\parallel}$ and $\Delta k_{\perp}$ are determined by Eq.2.5. By expanding both components into Taylor series, the gain bandwidth is solved through calculating the FWHM of the parametric gain via Eq.2.11.

1 When the 1st order term in the Taylor series is dominating, the gain bandwidth is
 2 calculated by,

$$3 \quad \Delta\Omega = 3.33 \left[\frac{|\Gamma|}{L} \frac{\cos(\theta + \alpha)}{(\Delta k_{\parallel}^{(1)})^2 + (\Delta k_{\perp}^{(1)})^2} \right]^{\frac{1}{2}} \quad (2.19)$$

4 where $\Delta k_{\parallel}^{(1)}$ and $\Delta k_{\perp}^{(1)}$ are the 1st order coefficients of the phase mismatch components.
 5 As shown in the model Eq.2.19, the gain bandwidth is inverse proportional to the
 6 square root of the crystal length and proportional to the fourth root of pump intensity.

7 When the $(\Delta k_{\parallel}^{(1)})^2 + (\Delta k_{\perp}^{(1)})^2$ approaches to zero, the gain bandwidth increases to
 8 infinity. This situation describes a group velocity matching between signal and idler
 9 pulses, in another word, the 1st order flatness of the phase mismatch curve is reached.

10 Under this situation, the first order terms $\Delta k_{\parallel}^{(1)}$ and $\Delta k_{\perp}^{(1)}$ vanish and the second order
 11 terms dominate. The gain bandwidth is calculated by,

$$12 \quad \Delta\Omega = 2.58 \left[\frac{|\Gamma|}{L_c} \frac{\cos(\theta + \alpha)}{(\Delta k_{\parallel}^{(2)})^2 + (\Delta k_{\perp}^{(2)})^2} \right]^{\frac{1}{4}} \quad (2.20)$$

13 where $\Delta k_{\parallel}^{(2)}$ and $\Delta k_{\perp}^{(2)}$ are the 2nd order coefficients of the phase mismatch components.
 14 As mentioned previously , the principle of this model is based on pursuing the
 15 minimum value of the modulus Δk for each signal frequency component. Considering
 16 the case where the 1st order term in the Taylor series is dominating, the Δk can be
 17 calculated as,

$$18 \quad \Delta k = \sqrt{(\Delta k_{\parallel}^{(1)})^2 + (\Delta k_{\perp}^{(1)})^2} |\omega_s - \omega_{s0}| \quad (2.21)$$

1 Therefore, the $\left(\Delta k_{\parallel}^{(1)}\right)^2 + \left(\Delta k_{\perp}^{(1)}\right)^2$ term also takes its minimum value. It is seen from
2 the proposed model Eq.2.19 that the expected gain bandwidth $\Delta\Omega$ is an upper-limit of
3 all possible reached value. Also, the proposed models consist of a new relation between
4 the gain bandwidth and the new design parameters such as the pump-surface normal
5 angle α . It should be noted that, when the perpendicular component $\Delta k_{\perp}$ and angle α
6 both equal to zero, the proposed models reduce to the previous model demonstrated in
7 paper [48]. Therefore, the proposed model could be regarded as the more general gain
8 bandwidth model.

9 **2.4.4 Feasibility studies of the proposed gain bandwidth model**

10 The performance of the proposed gain bandwidth model is evaluated by simulation
11 studies. It focuses on answering ‘in what case’ and ‘how well’ the proposed model can
12 predict the gain bandwidth value of the OPA/OPCPA. In the simulation, we consider a
13 non-collinear, 5mm thick GaSe based OPCPA process with Type I (ooe) phase match.
14 The OPCPA is driven by the 2060nm pump pulses with 10ps pulse duration and 1
15 GW/cm² pump intensity. The non-collinear angle is set to be 1.5°. In the simulation,
16 the centre wavelength λ_{s0} of the signal wave varies from 2500nm to 3400nm under
17 three cases where the pump-surface normal angle α taking the value -20° , 0° , and 20° ,
18 respectively. The simulation results are calculated respectively by using the gain
19 bandwidth model Eq.2.19, Eq.2.20 and by solving numerically the coupled pulsed-
20 wave equations, for a comparison purpose. The simulation results are shown in the
21 Fig.2.11.

22 In all cases shown in Fig.2.11, it is shown that the gain bandwidth estimated by the
23 proposed model is the upper bound of the true value. The feasibility of the proposed
24 model is confirmed within both small bandwidth regime ($\lambda_{s0} < 3000$ nm) with $< 20\%$
25 relative bandwidth and the large bandwidth regime ($\lambda_{s0} > 3000$ nm) with $> 20\%$
26 relative bandwidth). The error of the gain bandwidth evaluation is less than 15% when

1 $\lambda_{s0} < 3000$ nm. However, the evaluation error grows bigger in the large gain bandwidth
2 regime. In the large gain bandwidth regime, especially for the λ_{s0} around 3300nm, the
3 gain bandwidth calculated via Eq.2.19 increases dramatically towards infinity. This
4 phenomenon is introduced by the matching of the group velocities of the signal and
5 idler pulses. In this case, the model Eq.2.19 loss its feasibility and the model Eq.2.20
6 becomes effective in this region, as shown by the blue dashed lines. In the region
7 between 3000nm and 3300nm, both models cannot provide an accurate estimation for
8 the gain bandwidth. This is because the 1st and 2nd order terms of the Taylor series have
9 the approximately equal value so that none of them could dominate the value of the Δk .
10 Therefore, both models are invalid in this region.

12 Fig.2.11. Comparison between the evaluated gain bandwidth based on the model (Eq.2.19,
13 black line; Eq.2.20, blue dashed line) and numerically solution of the coupled wave equation,
14 red dotted line). (a) $\alpha = -20^\circ$, (b) $\alpha = 0^\circ$, (c) $\alpha = 20^\circ$.

15 The feasibility of the proposed model is further evaluated to see its capability of
16 revealing the relation between the key performance indicator, the gain bandwidth, and
17 the design parameters. Here, we mainly concern about the following design parameters:
18 the non-collinear angle ρ_s , the crystal length L , the pump intensity I_p and the pump-

1 surface normal angle α . The simulations based on the GaAs OPA driven by the 2060nm
2 pump pulse are conducted to reveal these relationships.

3 **1) Gain bandwidth and the non-collinear angle ρ_s**

4 In this simulation, the crystal length is set to be 5mm and the pump-surface normal
5 angle α is chosen to be zero. The pump pulse has 1 GW/cm² intensity. The centre
6 wavelength of the signal wave is 2774nm which corresponds to an 8 μ m idler output.
7 We change the non-collinear angle ρ_s from 0° to 3° with every 0.1° in steps while
8 evaluating the gain bandwidth at the same time. The simulation results are summarized
9 in Fig.2.12(a) which demonstrates clearly that a gain bandwidth increases first and then
10 decreases. Its maximum value is reached at a specific angle, where ρ_s equals to ~1.1°.
11 The same results can also be observed in another case with 3137 nm signal input and 6
12 μ m idler output. In this case, the critical non-collinear angle equals to 1.527°.

13 **2) Gain bandwidth and the pump intensity I_p**

14 In this simulation, the pump-surface normal angle α is zero and the non-collinear angle
15 ρ_s is chosen to be its critical value. Therefore, model Eq.2.20 is used to evaluate the
16 gain bandwidth. The centre wavelength of the signal waves is 2595 nm respectively.
17 By varying the pump intensity from 0 to 1GW/cm² with 0.1GW/cm² in each step, the
18 gain bandwidth is calculated as shown in Fig.2.12(b). It shows that the gain bandwidth
19 increases when using higher pump intensity with a decreasing changing rate.

20 **3) Gain bandwidth and the crystal length L**

21 In this case, the simulation environment is the same as in the previous case. We take
22 three crystal lengths, 5 mm, 10 mm, and 15 mm respectively in each simulation. As
23 shown in Fig.2.12(b), the gain bandwidth decreases with the longer crystal length.
24 Therefore, together with the previous simulation, the results show that the

1 OPA/OPCPA with higher pump intensity and thinner nonlinear crystal would be
2 favoured to produce broadband output.

3 **4) Gain bandwidth and the pump-surface normal angle α**

4 In this simulation, three design parameters, the non-collinear angle ρ_s , the pump
5 intensity I_p and the crystal length L are chosen as 3° , $1\text{GW}/\text{cm}^2$ and 5mm , respectively.
6 The centre wavelength of the signal wave is set at 2595 nm , the same as in the previous
7 cases. The gain bandwidth is calculated with angle α tuned from -15° to 15° in steps
8 of 1° , as illustrated in Fig.2.12(c). The curve shows a peak value of gain bandwidth
9 which occurs at $\alpha = 11.8^\circ$, which also fulfils $\alpha = -\theta$. Here, θ is the angle between the
10 wavevector mismatch $\vec{\Delta k}$ and the pump wavevector $\vec{k}_p$. The existence of this critical
11 angle α is attribute to the minimum phase mismatch accumulation in the parametric
12 amplification.

13 In the parametric amplification, the phase mismatch accumulated along the direction
14 of $\vec{\Delta k}$ which could be characterized by angle $\theta + \alpha$ between $\vec{\Delta k}$ and the surface normal,
15 as shown in Fig.2.2. Along this direction, the phase mismatch accumulates from 0 to
16 its upper-limit $\frac{\Delta k L}{\cos(\theta + \alpha)}$. In this process, the signal waves generated at each position
17 destructively interfere with each other. The destructive interference is minimized only
18 when $\frac{\Delta k L}{\cos(\theta + \alpha)}$ also takes its minimum value. As demonstrated previously, the phase
19 mismatch is determined by the choice of the direction ρ_i of the idler wave vector.
20 Therefore, for a minimum phase accumulation, the direction ρ_i should follow the
21 below constrain,

$$22 \quad \frac{\partial}{\partial \rho_i} \left(\frac{\Delta k L_c}{\cos(\theta + \alpha)} \right) = 0 \quad (2.22)$$

1 Take the ordinary idler wave as an example, Eq.2.22 has the solution $\rho_i = 2\theta + \alpha$.
2 Since we also require the Δk be its minimum, therefore the idler wave vector should
3 have the same direction with the $\vec{\Delta k}$. Therefore, ρ_i also equal to θ , which immediately
4 repeat the simulation result $\alpha = -\theta$. Under this critical angle, both the wave vector
5 mismatch Δk and the phase accumulation in the whole process reach their minimum
6 value. As a result, all signal frequency components are most efficiently amplified, and
7 this means a maximum gain bandwidth. The phase matching geometry in this case is
8 shown in Fig.2.12(d).

9

10 Fig.2.12. Gain bandwidth with (a) non-collinear angle ρ_s , (b) pump intensity and crystal
11 length, (c) pump-surface normal angle α , and the Phase matching geometry when condition
12 $\alpha = -\theta$ is fulfilled.

13

2.5 Design non-collinear OPCPA by using N^{th} order flatness of phase mismatch principle

According to Eq.2.19 and Eq.2.20, the gain bandwidth could be significantly enhanced by making not only the first but also the second order terms of phase mismatch equal to zero. This way actually can be extended to the N^{th} order terms of phase mismatch. By vanishing higher order terms of phase mismatch, the value of phase mismatch can be made zero within a broader signal wavelength range. Therefore, more signal frequency components can be efficiently amplified and the gain bandwidth will be enhanced. The idea can be seen easily from the spectrum curve relating the phase mismatch and the signal frequency, the higher order flatness of the spectrum means a larger gain bandwidth. This session discloses in details the principle of using the N^{th} -order-flatness of the spectrum of phase mismatch as a design guideline for the design of non-collinear OPA/OPCPA..

As shown in the Eq.2.23, the phase mismatch could be decoupled into two components $\Delta k_{\parallel}$ and $\Delta k_{\perp}$. For simplicity, let us assume that the pump wave is a monochromatic wave. Since the spectral bandwidth of the pump wave is much smaller than the signal bandwidth, therefore this assumption could be safely made. In this case, both $\Delta k_{\parallel}$ and $\Delta k_{\perp}$ can be considered as the function of signal frequency ω_s . By expanding Eq.2.23 into its Taylor series over a certain signal frequency ω_{s0} , we have,

$$\begin{aligned}\Delta k_{\parallel}(\omega_s) &= \Delta k_{\parallel}^{(0)}(\omega_{s0}) + \Delta k_{\parallel}^{(1)}(\omega_{s0})(\omega_s - \omega_{s0}) + \Delta k_{\parallel}^{(2)}(\omega_{s0})(\omega_s - \omega_{s0})^2 + \dots \\ \Delta k_{\perp}(\omega_s) &= \Delta k_{\perp}^{(0)}(\omega_{s0}) + \Delta k_{\perp}^{(1)}(\omega_{s0})(\omega_s - \omega_{s0}) + \Delta k_{\perp}^{(2)}(\omega_{s0})(\omega_s - \omega_{s0})^2 + \dots\end{aligned}\quad (2.23)$$

In order to achieve the broadest gain bandwidth, we expected that the phase mismatch components $\Delta k_{\parallel}(\omega_s)$ and $\Delta k_{\perp}(\omega_s)$ vanish for all ω_s . However, due to the material dispersion, this expectation can only be fulfilled for only one of the signal frequency components ω_{s0} . But we still expect that the phase mismatch of the frequency

1 components ω_s around the ω_{s0} do not vary too much and hopefully as small as
2 possible. If the phase mismatch is plotted as a curve with respect to ω_s , the curve
3 should be as flat as possible at the point ω_{s0} . For easy reference, the curve is also
4 referred as the phase mismatch spectrum. This gives rise to the N^{th} order flatness for
5 the phase mismatch spectrum, which is detailed as follows,

6 **Definition of the N^{th} order flatness**

7 For $\forall n \leq N$, $\Delta k_{\parallel}^{(n)} = \Delta k_{\perp}^{(n)} = 0$ and $\Delta k_{\parallel}^{(N+1)} \neq 0$, $\Delta k_{\perp}^{(N+1)} \neq 0$, we call the phase
8 mismatch curves $\Delta k_{\parallel}(\omega_s)$ and $\Delta k_{\perp}(\omega_s)$ have the N^{th} order flatness.

9 A simulation is conducted here to illustrate how the **N^{th} order flatness** affects the phase
10 mismatch curve and the conversion efficiency improvement. It simulates an OPA
11 process in a 1mm BBO crystal with 532nm pump wavelength and 1000nm signal
12 wavelength. Here, we take the Type I, ooe phase matching type with 22.8° phase
13 matching angle and a collinear phase matching geometry. In order to realize the N^{th}
14 order flatness in a simple way, we manually cancel out the first $N-1^{\text{th}}$ order term of the
15 phase mismatch. The conversion efficiency is evaluated against the frequency by using
16 the traditional sinc^2 function, which is detailed illustrated in the chapter 2 of Ref.[46].
17 The FWHM value also implies the gain bandwidth of the system. The simulation
18 results are shown in the Fig.2.13.

19 Fig.2.13(a) shows clearly that the phase matching condition is fulfilled only at the
20 center frequency of signal. When the order of flatness is increased from 0^{th} to the 2^{nd} ,
21 the phase mismatch curve becomes flatter around the central frequency. Within the
22 400THz spectral range, the upper-limit of the phase mismatch decreases from 120 rad
23 to 3.14 rad. As a result, larger amount of the signal frequency components would have
24 phase mismatch $|\Delta kL|$ less than π in the higher order flatness case. This means
25 alternatively that the gain bandwidth is larger for higher order flatness, as shown in

1 Fig.2.13 (b). In a conclusion, the flatness of the phase mismatch spectrum gives a direct
 2 guidance for designing non-collinear OPA/OPCPA with broader gain bandwidth.

3
 4 Fig.2.13. (a) Phase mismatch curve and (b) Conversion efficiency under 0th (black line), 1st
 5 (blue line) and 2nd (red line) order flatness.

6 Nowadays, the 0th, 1st and 2nd order flatness of phase mismatch spectrum have been
 7 experimentally achieved. For the 0th-order flatness, the phase matching condition could
 8 only be fulfilled at one specific signal frequency. This type of flatness could be
 9 achieved by using material birefringence [46] and the quasi-phase-matching (QPM)
 10 technique [63-68]. When the 1st-order flatness is reached, the following equations
 11 should be both fulfilled,

$$\begin{aligned}
 \Delta k_{\parallel}^{(0)}(\omega_{s0}) &= \Delta k_{\perp}^{(0)}(\omega_{s0}) = 0 \\
 \Delta k_{\parallel}^{(1)}(\omega_{s0}) &= \Delta k_{\perp}^{(1)}(\omega_{s0}) = 0
 \end{aligned}
 \tag{2.24}$$

14 These two conditions are simultaneously satisfied in the degenerate OPA (DOPA) with
 15 collinear geometry [69] and the non-collinear OPA (NOPA) [54-56] with no angular
 16 dispersion of signal. For the 2nd-order flatness, an extra condition should be fulfilled
 17 compared with Eq.2.24 which is,

$$\Delta k_{\parallel}^{(2)}(\omega_{s0}) = \Delta k_{\perp}^{(2)}(\omega_{s0}) = 0 \quad (2.25)$$

Therefore, a new free variable should be introduced to make Eq.2.25 valid. A straightforward way is to choose such a variable as the angular dispersion rate $\left. \frac{\partial \rho_s}{\partial \omega_s} \right|_{\omega_{s0}}$ of signal.

In order to further increase the order of flatness, more new free variables such as the higher order terms $\left. \frac{\partial^2 \rho_s}{\partial \omega_s^2} \right|_{\omega_{s0}}, \left. \frac{\partial^3 \rho_s}{\partial \omega_s^3} \right|_{\omega_{s0}}$ of the angular dispersion can be chosen. However, the choices of the variables are not arbitrary as done in previous angular-dispersive NOPA. This is because the combination of gratings or prisms and achromatic lenses could only generate monotonic angular dispersion. Currently, the gain bandwidth of the angular-dispersive NOPA is still below one octave. New design of gratings is expected in order to further enhance the gain bandwidth.

2.6 Angular dispersive OPA (AD-OPA)

Following the idea in realizing the 2nd order flatness, we can naturally deduce that if N free variables, $\rho_s(\omega_{s0}), \left. \frac{\partial \rho_s}{\partial \omega_s} \right|_{\omega_{s0}}, \dots, \left. \frac{\partial^{N-1} \rho_s}{\partial \omega_s^{N-1}} \right|_{\omega_{s0}}$, are introduced, the N^{th} order flatness could be achieved. Therefore, when phase matching condition is fulfilled for all signal frequency components within a spectral range, there must be one specific angular dispersion geometry with the non-collinear angle $\rho_s(\omega_s)$ to be decided. This angle $\rho_s(\omega_s)$ can be derived by forcing the right-hand side of Eq.2.4 equal to zero, which is given by,

$$\rho_s(\omega_s) = \cos^{-1} \left[\frac{k_p^2 + k_s^2 - k_i^2}{2k_p k_s} \right] \quad (2.26)$$

1 This non-collinear angle of the signal wave can be designed by using a combination of
2 a prism and a chirped grating with particularly designed grating constant [70]. The
3 OPA system based on this design is called the angularly dispersed OPA (AD-OPA). Its
4 schematic is illustrated in Fig.2.14 (a).

5
6 Fig.2.14. (a) Schematic of the AD-OPA; (b) Non-collinear angle of signal for the optimized
7 grating design (red line) and the polynomial based grating design (blue line); (c) Comparison
8 of the flatness of the phase matching curve between collinear OPA and the AD-OPA; (d)
9 Calculated amplified spectrum and parametric gain curve.

10 The designed system runs as follows. It is pumped by a 1030nm laser generated in a
11 Yb:YAG CPA system. The pumped beam is assumed to have a flat-top profile
12 generated by using a commercialized beam shaper. The pulse energy and the pulse
13 duration of pump are 10mJ and 20ps, respectively. The broadband signal pulse with 2
14 μm central wavelength is generated through the intra-pulse DFG (IPDFG) process
15 driven by another Yb:YAG CPA system. The signal pulse is assumed to be a Gaussian

1 shape pulse with 6fs pulse duration. Its pulse energy is set as 10nJ. After the IPDFG, a
2 prism and a dedicatedly designed chirped grating are used to generate the angular-
3 dispersive signal. The non-collinear angle $\rho_s(\omega_s)$ of signal is designed to follow
4 Eq.2.26. In the amplification stage, the signal and pump are combined by an input
5 coupler (D1 in Fig.2.14 (a)). The nonlinear material used in the designed system is the
6 MgO-doped lithium niobate (MgO:LiNbO₃) crystal. Another pair of prism and chirped
7 grating is designed after amplification stage to compensate the angular dispersion of
8 the output signal. This pair of prism and chirped grating are both identical with the one
9 used before the nonlinear material. A 4:1 imaging system is also applied to magnify
10 the beam diameter of the signal and reduce its peak intensity. Therefore, the amplified
11 high peak power signal will not damage the prism II shown in Fig.2.14 (a).

12 Due to the limitation of the chirped grating fabrication, the non-collinear angle
13 generated through the prism and grating pair can only be calculated by a high-order
14 polynomial fitting method. In this design, we consider a tenth-order polynomial fitting
15 of the target function $\rho_s(\omega_s)$ in 1.3 to 4 μm spectral range. According to Fig.2.14(b),
16 the polynomial curve fits well with the target curve $\rho_s(\omega_s)$. The phase mismatch
17 calculated under the polynomial grating design is also close to the value that is
18 calculated under ideal grating design. The amplified spectrum and the gain curve are
19 calculated by the coupled wave equations in Eq.2.6. As shown in Fig.2.14(d), the
20 resultant gain bandwidth of the designed AD-OPA system is as large as 1.3-octave at
21 its FWHM. This value is larger than the gain bandwidth of the NOPA with 2th order
22 flatness [59,60].

23 Simulation studies are also done to compare the gain bandwidth of the AD-OPA system
24 to the one of the NOPA system using a Barium metaborate (BBO) crystal. In this
25 calculation, both systems are pumped by a 515nm laser and seeded by a broadband
26 signal with spectrum spanning from 600nm to 1500nm. The phase matching angle is
27 set as 24° for both systems. The non-collinear angle of the NOPA system is set as 2.6°

to reach the broadest gain bandwidth. In the AD-OPA system, the non-collinear angle follows the Eq.2.26. Fig.2.15 (a) show that the phase mismatch spectra of both systems. It is clearly seen that the phase mismatch of the AD-OPA is dramatically reduced over the whole signal spectral range. The calculated spectra of the amplified signal in both systems are shown in Fig.2.15 (b). The bandwidth of the AD-OPA is shown to be more than two times larger than the bandwidth of NOPA. The simulation result shows the advantage of the AD-OPA system in enhancing the gain bandwidth.

Fig.2.15. (a) Comparison of the phase mismatch curves between the AD-OPA system and NOPA system. The phase mismatch of the AD-OPA is shown in the red line and the phase mismatch of the NOPA is represented by the yellow line. The designed non-collinear angle of the AD-OPA system is also demonstrated with blue line. (b) Comparison of the amplified signal spectra between the AD-OPA system and NOPA system. The spectrum of the AD-OPA output is shown in the red line and the spectrum of the NOPA is represented by the yellow line.

Another point worthy to note is that the designed AD-OPA system is not limited by the signal wavelength and the nonlinear material. Hence we expect that the proposed AD-OPA method can be applied in the LWIR OPCPA system to relax the gain bandwidth limitation of the LWIR OPCPA system.

1   **2.7 Conclusions**

2   In this chapter, we reviewed first the fundamental theory of the OPCPA process, which
3   involves two basic conservation laws, the energy and kinetic conservation laws as well
4   as the coupled pulsed-wave equations [46,47]. These basic theory serves as the first
5   principles used to characterize the behaviours of OPCPA process and to calculate
6   ground truths for benchmarking the performance of the OPCPA models and methods
7   developed in the thesis.

8   Based on the first principles of OPCPA, the main challenges in development of OPCPA
9   in LWIR regime are addressed by simulation studies. The numerical results show that
10   the fundamental challenge lies in the large group velocity mismatch between the
11   pump/signal and the idler waves which is introduced by the limited band gap of the
12   nonlinear medium material used in OPCPA. It is also found that the OPP in a LWIR
13   OPCPA introduces extra GDD and even higher order dispersion into the signal and
14   idler pulses compared to the OPCPA systems operated at NIR/SWIR regimes.
15   Therefore, the effect of the OPP must be considered in the chirp compensation of the
16   LWIR OPCPA.

17   In order to dealing with the basic challenges of the LWIR OPCPA, we proposed three
18   possible solutions. One of the possible solutions of the gain bandwidth limitation is
19   discussed in this chapter. In order to enhance the gain bandwidth, the non-collinear
20   phase match geometry could be introduced in the LWIR OPCPA. At first, a general
21   gain bandwidth model which is suitable for the non-collinear OPCPA is proposed. This
22   model is based on the coupled pulsed-wave equation and the minimum wave vector
23   mismatch assumption. The model is proved to be able to evaluate the upper-limit of the
24   gain bandwidth within 15% calculation error in the long wavelength mid-IR regime.
25   This value is 5% higher than the calculation error previous model [48] as shown Fig.2.7.
26   Based on the gain bandwidth model, the ‘Nth order flatness’ design rule is introduced

1 as a guideline to design a broadband non-collinear OPA system. Following this rule,
2 we propose an AD-OPA system by using a designed chirped grating. The bandwidth
3 of the AD-OPA system is shown to be larger than the traditionally used NOPA system.
4 Therefore, the proposed AD-OPA system could be considered as a good solution of the
5 gain bandwidth limitation. Other two solutions of the gain bandwidth limitation will be
6 detailed studied in the next chapter.

7

8

1   **Chapter 3. Design and Implementation of a 9 μ m OPCPA**

2   **3.1 Introduction**

3   In previous chapter, developing an OPCPA at LWIR regime was theoretically shown
4   to be achievable. The numerical studies also provide design guidelines and solutions of
5   challenge for the experimental development of LWIR OPCPA systems. Based on these
6   studies, this chapter addresses the design and implementation of LWIR OPCPA with high
7   energy and large gain bandwidth. The major concerns are about the suitable pump sources and
8   nonlinear materials that can help to achieve large gain bandwidth and high output energy, the
9   careful dispersion management for generating transform-limited pulse, numerical design and
10   experimental develop of all components of the OPCPA system.

11   In this chapter, the LWIR OPCPA is developed by using literature review, numerical analysis
12   and experimental verification. First of all, the key challenge of developing a LWIR OPCPA
13   and its solutions are revisit through the review of the pump sources and nonlinear
14   materials. The studies show that combining the 1 μ m pump laser and the nonlinear
15   crystal with large band gap ($>3\text{eV}$) is a better solution to reach both large gain
16   bandwidth and high output energy. Based on this combination, we numerically design
17   each component of the LWIR OPCPA system. A large band gap nonlinear crystal
18   called LiGaS₂ is used to support the LWIR pulse generation. Fine dispersion
19   management of the LWIR pulse with considering the optical parametric phase (OPP)
20   is also conducted, as suggested in previous chapter. Following the numerical design,
21   we experimentally develop an OPCPA system at 9 μ m. The developed 9 μ m OPCPA is
22   the first, to the best of our knowledge, OPCPA system that operates at LWIR region.
23   The gain bandwidth of the 9 μ m OPCPA is shown to be larger than the reported 5 μ m
24   [26] and 7 μ m [23] OPCPAs.

The contents of this chapter are arranged as follow. In section 3.2, the general schematic of an OPCPA is first demonstrated. The key challenge of developing a LWIR OPCPA system and its solution are revisited by literature review of the pump source and nonlinear medium. Section 3.3 introduces the design of each component of the LWIR OPCPA. Detailed chirp management is also achieved by considering the optical parametric phase into the calculation. Based on the design, section 3.4 addresses the implementation of the LWIR OPCPA apparatus. The performances of the LWIR OPCPA is also examined through the measurement of the spectrum, pulse shape, beam profile of the output.

3.2 OPCPA system and its key challenge

3.2.1 General schematic of an OPCPA

Traditionally, the real OPCPA systems share the similar architecture [75] as shown in Fig.3.1. An OPCPA consists of five fundamental components. A high energy, high power pump laser drives the whole OPCPA system. The seed generator provides broadband pulses to be further amplified. The seed pulse is then stretched in a pulse stretcher, amplified in an OPA stage and compressed to reach high intensity in a pulse compressor.

Fig.3.1 Illustration of the general schematic of an OPCPA system.

1 The seed generator is located at the front end of the OPCPA. Traditionally, in the NIR
 2 regime, the broadband seed could be directly provided by the an octave-spanning laser
 3 oscillator, for example an Ti:sapphire oscillator [107]. In the mid-IR region, the intra-
 4 pulse DFG is another way to generate seed pulses driven by an oscillator [108]. This
 5 technique generates various seed wavelengths through nonlinear wave mixing of any
 6 two different components of the broadband oscillator output. It could also ensure a
 7 passive CEP stability of the generated seed pulse. However, these methods are
 8 normally based on a broadband oscillator which is independent to the high energy
 9 pump laser. The synchronization between the pump laser and the oscillator introduces
 10 the complexity and the time jitter problem into the OPCPA system.

11 The broadband seed could now be generated directly from the narrow-band pump laser
 12 via the supercontinuum generation (SCG) in gas [109] and solid [110]. The broadening
 13 of the spectrum during the SCG process is due to the self-phase modulation (SPM)
 14 effect. The SPM effect is a nonlinear effect of the light-matter interaction. In this
 15 process, the refractive index varies with the pulse intensity due to the optical Kerr effect,
 16 which will introduce a phase change for the input pulse which varies with time. Here,
 17 the instantaneous phase of the input pulse could be written as [111],

$$\begin{aligned}
 \Phi(t) &= \omega_0 t - k_0 L \\
 &= \omega_0 \left(t - \frac{n(I)}{c} L \right)
 \end{aligned}
 \tag{3.1}$$

20 Therefore, the instantaneous angular frequency could be calculated from Eq.3.1 as,

$$\omega(t) = \frac{d\Phi(t)}{dt} = \omega_0 - \frac{\omega_0 L}{c} \frac{dn(I)}{dI} \frac{dI}{dt}
 \tag{3.2}$$

22 Eq.3.2 illustrates a Gaussian pulse and its instantaneous frequency which changes with
 23 time in a SPM process. At the leading edge of the pulse (Fig.3.2), the pulse intensity
 24 increases with time so that the frequency shift to a smaller value. On the contrary, at

1 the trailing edge (Fig.3.2), the higher instantaneous frequencies are generated. It is
 2 obvious that new frequencies are generated in a SPM process and the spectrum could
 3 be broadened.

4
 5 Fig.3.2 Illustration of (a) a Gaussian pulse and (b) the instantaneous frequency of the pulse in
 6 SPM process.

7 Nowadays, SCG assisted by the filamentation in solid material is widely used to
 8 generate the seed pulse. According to Eq.3.2, the frequency shift $\omega(t) - \omega_0$ is
 9 proportional to the $\frac{dn(I)}{dI}$ term which equals to the nonlinear refractive index n_2 .
 10 Normally, the solid materials could provide large n_2 which is normally orders of
 11 magnitude larger than the n_2 of gas. Therefore, a broader spectrum could be obtained
 12 in the solid compared with the SCG gas with similar pump intensity and interaction
 13 length. Moreover, the interaction length of the SCG in solid could be significantly
 14 enhanced with the help of the filamentation so that the supercontinuum spectrum could
 15 be even broader.

16 The dispersion management of the seed pulse is achieved through carefully design of
 17 pulse stretcher and compressor. Both the stretcher and compressor can be built with the
 18 combination of bulk material, grating pairs, prism pairs and the acousto-optical

programmable dispersive filter (AOPDF). The bulk material is used to control the group velocity dispersion (GDD) of the seed pulse. If compensation of the third order dispersion (TOD) is also required, the grating pairs or prism pairs should be applied in the dispersion management. Nowadays, the AOPDF is commercially available for even higher order dispersion management and phase fine-adjustment. With the help of the AOPDF, complete dispersion compensation can be achieved in >300 nm bandwidth in visible range which could support a sub-10-fs pulse [75].

3.2.2 Key challenges and solutions

As discussed in chapter 2, the key challenge of developing an OPCPA system lies in its limited gain bandwidth, especially in the LWIR regime. Besides, the energy scale-up is also a challenge due to the limited pump energy and the damage threshold of the nonlinear medium. Previous chapter addresses three solutions of the key challenge, including using non-collinear phase matching geometry, applying longer wavelength pump laser and larger band gap nonlinear crystals in the OPCPA system. However, the proposed non-collinear AD-OPA method is still hard to be implemented due to the difficulty of manufacturing the chirped grating. Therefore, in order to experimentally develop an OPCPA system, we need to focus on another two solutions and discuss about the available pump source and nonlinear medium. In this section, we will see that by using the large band gap ($>2.4\text{eV}$) material, we can overcome the gain bandwidth limitation and ensure the energy boost-up at the same time.

1) Pump source

According to the study of Chapter 2, the limited gain bandwidth could be relieved by using the $>2\text{ }\mu\text{m}$ pump sources in a long wavelength mid-IR OPA/OPCPA. Thanks to the development of the Ho/Tm doped high energy laser sources operating at $\sim 2\text{ }\mu\text{m}$, MWIR and LWIR pulses could now be generated through DFG [71,72] and intra-pulse DFG [73,74] with high average power. Besides, driven by the $2\text{ }\mu\text{m}$ pump laser,

OPCPAs at $>4\mu\text{m}$ MWIR wavelength have also been demonstrated with around millijoule level pulse energy. Among all the demonstrations, the most striking works are the ZnGeP₂ crystal based OPCPAs which provide $5\mu\text{m}$ [26] and $7\mu\text{m}$ [23,39] pulses with 1.3mJ and 750 μJ pulse energy, as well as the 75fs and 188fs pulse duration, respectively. Apart from the Ho/Tm doped lasers, commercialized $2\text{-}\mu\text{m}$ OPCPA is another choice as the pump source. In recent years, long wavelength mid-IR pulses with few micro-joule energy and few-cycle pulse duration have also been produced in the OPA [76] and DFG [77] driven by the $2\text{-}\mu\text{m}$ OPCPA.

However, compared with the more advanced $1\text{-}\mu\text{m}$ pump sources, the pulse energy and power of the $2\text{-}\mu\text{m}$ pump lasers nowadays are still limited which will also confine the energy boost-up of the OPCPA system. For example, the performances of several representative $1\text{-}\mu\text{m}$ and $2\text{-}\mu\text{m}$ pump sources are summarized in the Table 3.1. The available manufacturable sizes of the $2\text{-}\mu\text{m}$ gain media, such as the Ho:YLF and Cr:ZnSe crystals, reach $\sim 5\text{cm}$ which is similar as the $1\text{-}\mu\text{m}$ gain media [78]. However, the $2\text{-}\mu\text{m}$ pump sources nowadays could only provide hundreds of millijoule level pulse energy, several tens of Gigawatt-level peak power and Watt-level average power. For instance, the Ho:YLF based pump laser developed by the Elu et al. [23] could provide pulses with 260mJ pulse energy, 16.25GW high peak power and 26W average power. On the contrary, the Nd-based and Yb-based $1\mu\text{m}$ CPA systems could now provide much higher peak power and average power for driving the OPCPA systems. The peak power and average power of the $1\mu\text{m}$ pump sources could be as high as hundreds of gigawatt and kilowatt level respectively, which are one order or several orders higher than their $2\mu\text{m}$ counterparts. With the help of the advanced thin-disk technique, the $1\mu\text{m}$ Yb:YAG laser demonstrated by Nubbemeyer et al. [82] provides 181.8GW peak power and 1kW average power. Single pulse energy of the $1\mu\text{m}$ pump sources could reach Joule-level for picosecond lasers or even hundreds-joule-level for nanosecond lasers [90].

Table 3.1. Summary of the 1 μ m and 2 μ m pump sources. Here, the ‘W’, ‘H’ and ‘L’ represent for the width, height and length.

	Energy (mJ)	Pulse width (ps)	Peak Power (GW)	Repetition rate (kHz)	Average Power (W)	Amplifier Material	Crystal Size (W*H*L, mm)	Citation
1μm pump lasers	1500	5	300	0.5	750	Yb:YAG	30*30*2	[79]
	130	0.45	290	0.01	1.3	Yb:YAG	-	[80]
	200	1	200	1	200	Yb:YAG	-	[81]
	200	1.1	181.8	5	1000	Yb:YAG	14*14*0.2	[82]
	4.7	6.5	0.72	300	1410	Yb:YAG	17*17*0.13	[83]
	44.5	0.82	54.2	100	4450	Yb:YAG	17*17*0.36	[84]
	1000	100	10	0.02	20	Nd:YAG	-	[85]
2μm pump lasers	330	350000	0.0009	0.05	16.5	Ho:YLF	50*2*10	[86]
	260	16	16.25	0.1	26	Ho:YLF	50mm length	[23]
	39	10	3.9	0.1	3.9	Ho:YLF	35mm length	[87]
	25	4.3	5.8	1	25	Ho:YLF	50mm length	[88]
	2.3	0.088	26.1	1	2.3	Cr:ZnSe	40mm length	[89]

In a conclusion, the comparison between the 1 μ m and 2 μ m pump lasers shows that the 2 μ m pump laser is not suitable for energy boost-up although it can help to overcome the gain bandwidth limitation. However, if we could find a suitable nonlinear crystal which has large band gap and supports frequency down-conversion from the advanced 1 μ m pump pulse to the LWIR pulse, both the high output energy and large gain bandwidth can still be achieved.

2) Nonlinear medium

Nowadays, generating LWIR photons in the OPA/OPCPAs is supported by several non-oxide nonlinear crystals such as the GaSe [91-93], AgGaS₂ [71,94], AgGaSe₂

[95,96], CdSiP2 [76] and ZnGeP2 [23,26,38,39]. The band gaps of these materials are around or even far less than 2.4eV (Table 3.2), which is twice of the photon energy of the traditional 1 μm pump lasers. The gain bandwidth of OPCPA would also be limited due to the large group velocity mismatch between the pump/signal and idler pulses, as discussed in the previous chapter. By using these materials, the applied pump energy is also restricted due to the severe multiphoton absorption.

Table 3.2. Summary of nonlinear materials used in the long wavelength mid-IR OPA/OPCPA.

	GaSe	AgGaS2	AgGaSe2	CdSiP2	ZnGeP2
Band gap ^[97] (eV)	2.1	2.73	1.8	2.45	2.45
Transparency range ^[98] (μm)	0.65-18	0.5-13	0.71-18	0.66-6.5	0.74-12

In order to enhance the gain bandwidth, materials with larger band gap ($>2.4\text{eV}$) should be studied. Fortunately, several nonlinear materials such as the LiGaS₂ (LGS) and LiGaSe₂ (LGSe) are found to have large band gap and broad transparency range. The band gaps of these materials are normally larger than 3eV so that the group velocity mismatch problem will be mitigated. Even more exciting, these materials also support the directly frequency down-conversion from 1 μm to LWIR wavelength. The advanced 1 μm laser thus can be used in the OPCPA system with less multi-photon absorption. In recent years, several pioneering LGS based lasers, such as the OPAs [74,99], DFG [91] and the intra-pulse DFGs [73,100,101] driven by 1 μm pump sources, have been reported. These systems could generate LWIR pulses with only nano-joule-level pulse energy. For example, a recent reported OPA [74] could provide 8.5 μm output with 34.7 nJ pulse energy and 1.3 W average power. Although these studies are still in their preliminary stage, they still pave the way to generate high energy, ultrafast LWIR pulses in an OPCPA system.

1 In a conclusion, using a large band gap material in the LWIR OPCPA is a better choice
2 since it kills two birds with one stone. It can not only overcome the limited gain
3 bandwidth by mitigating the group velocity mismatch, but also support the advanced
4 $1\mu\text{m}$ pump laser and ensure the energy boost-up of the OPCPA.

5 **3.3 Design of $9\mu\text{m}$ OPCPA system**

6 The studies in previous section show that the combination of $1\mu\text{m}$ pump laser and large
7 band gap nonlinear crystal is required to achieve large gain bandwidth and high output
8 energy in a LWIR OPCPA. Following this conclusion, we design the LWIR OPCPA
9 based on the $1.03\mu\text{m}$ Yb:YAG laser and currently available large band gap nonlinear
10 crystal in this section. First of all, we discuss the best choice of the nonlinear medium.
11 Other fundamental components including the seed generator, the pulse stretcher, the
12 amplification stages and the pulse compressor are then theoretically and experimentally
13 designed.

14 **3.3.1 Nonlinear medium**

15 In Table 3.3, we list the properties of seven eligible large band gap nonlinear media
16 which could be used in the LWIR OPCPA. The demonstrated crystals include the
17 AgGaGeS₄ (AGGS), AgGaS₂ (AGS), HgGa₂S₄ (HGS), LiInS₂ (LIS), LiInSe (LISe),
18 LiGaS₂ (LGS) and LiGaSe (LGSe). In this table, the group velocity mismatch between
19 signal and idler is calculated by considering the signal and idler wavelengths equal to
20 $1.163\mu\text{m}$ and $9\mu\text{m}$ respectively. As shown in Table 3.3, all crystals have broad
21 transparency range which spans from the visible range to LWIR wavelength. Among
22 all materials, the band gaps of the AGS, HGS and LISe are small. At the same time,
23 the group velocity mismatches of these materials are large which means a small gain
24 bandwidth according to the proposed model in chapter 2. Among all other crystals, the
25 LGS has the largest band gap 3.75eV , largest damage threshold $240\text{MW}/\text{cm}^2$ at
26 1064nm , 14ns [106], and the smallest group velocity mismatch 43.3 ps/m . Its small

group velocity mismatch guarantees a large gain bandwidth of the parametric amplification. Although its nonlinear coefficient is relatively small, we can still achieve high energy output by increasing the pump energy and the crystal length. Therefore, a lot of evidences show that the LGS is the most suitable nonlinear crystal in developing LWIR OPCPA.

Table 3.3. Summary of the large band gap nonlinear media.

	Transparency range ^[98] (μm)	Band gap ^[102] (eV)	d_{eff} ^[102] (pm/V)	Damage threshold ^[102-105] (MW/cm ² , @1064nm, 14ns)	Group velocity mismatch ^[98] (ps/m)
AGGS	0.7-13	3.0	3.32 (XZ, oo-e) 5.43 (XY,oo-e)	103	160 (XZ, oo-e) 160 (XY,oo-e)
AGS	0.5-13	2.7	8.86 (oo-e) 13.65 (eo-e)	20	233 (oo-e) 306 (eo-e)
HGS	0.5-13	2.79	15.57 (oo-e) 21.18 (eo-e)	315	210 (oo-e) 280 (eo-e)
LIS	0.4-12	3.57	4.65 (XZ, oo-e) 6.77 (XY, eo-e)	40	46.7 (XZ, oo-e) 120 (XY, eo-e)
LiSe	0.5-12	2.86	7.26 (XZ, oo-e) 10.57 (XY, eo-e)	36	296.7 (XZ, oo-e) 373 (XY, eo-e)
LGS	0.32-11.6	3.76	4.47 (XZ,oo-e) 5.60 (XY,eo-e)	240	106.7 (XZ,oo-e) 43.3 (XY,eo-e)
LGSe	0.37-13.2	3.65	7.82 (XZ,oo-e) 9.31 (XY,eo-e)	<240	140 (XZ,oo-e) 210 (XY,eo-e)

The LGS crystal is a biaxial material with the orthorhombic structure (mm2). It has large transparency range from 0.32-11.6 μm . According to the data provided by the manufacturer, the ASCUT Ltd. & Co. KG, the LGS crystal with 3mm thickness has the >75% transmission at 1-8.5 μm and the >50% transmission below 10 μm wavelength as shown in Fig.3.3(a). The transmission of the 1 μm pump wave is verified as 80% which is in accordance with the data. The measurement is done by increasing the pulse energy of a 1.03 μm Yb:YAG laser from 0 to 800 μJ . According to the Fig.3.3(b), the transmission curve shows a linearly increasing of the output energy and

- 1 a constant transmission rate. It also implies that there is no obvious two photon
- 2 absorption occurs even under high pump intensity.

Fig.3.3. Information of the LGS crystal, including (a) Transparency curve in 1-12μm spectral range; This image is provided by Mrs. Yulia Smirnova from the ASCUT Ltd. & Co. KG; (b) Transmission at 1.03μm under different pump energy; (c) Normalized conversion efficiency with different idler wavelength and azimuthal angle; (d) Diagram of the cutting of the LGS crystal.

There are two different types of phase matching for an LGS crystal. According to [Table 3.3](#), Type I (ooe) phase matching could be achieved with 4.47 pm/V nonlinear coefficient when all waves are propagating in the XZ plane ($\Phi = 0$). Type II (eo-e) phase matching is achieved with a larger 5.60 pm/V nonlinear coefficient at the XY plane ($\theta = \frac{\pi}{2}$). Compared with the Type I phase matching, the group velocity mismatch of the Type II phase matching (43ps/m) is smaller which implies a larger gain

1 bandwidth. Therefore, using the Type II phase matching is the best choice for achieving
2 both higher gain and broader gain bandwidth.

3 The conversion efficiencies of idler for the Type II phase matching is characterized by
4 the equation $\text{sinc}^2(\Delta kL/2)$ which is a function of the azimuthal angle Φ and the idler
5 wavelengths [46]. The phase matching curve is shown in Fig.3.3 (c). According to
6 Fig.3.3 (c), the most widely gain bandwidth would be achieved at the 38.4° azimuthal
7 angle. In this case, efficient amplification could be achieved in the $8.2\ \mu\text{m}$ to $11\ \mu\text{m}$
8 spectral range. At 38.4° angle, the central wavelength of the idler wave is located at $9\ \mu\text{m}$
9 as shown by the white line in Fig.3.3(c).

10 In a conclusion, we will use the LGS crystal with 3.76eV band gap, Type II (ooe) phase
11 matching in the XY plane and the $\Phi = 38.4^\circ$ phase matching angle [98] in the LWIR
12 OPCPA. The designed directions of the crystal surface normal, the principle axes of
13 the crystal and the polarizations of the pump, signal and idler waves are all shown in
14 Fig.3.3(d).

15 **3.3.2 Seed generator**

16 According to section 3.2, the seed generator based on the SCG effect in solid material
17 is favoured for its compact design, large output bandwidth and elimination of the pump-
18 seed synchronization problem. Therefore, we generate the seed pulse based on the
19 solid-based SCG process. In the LWIR OPCPA, we hope that the bandwidth of the
20 seed could be at least the same as the gain bandwidth of the nonlinear material. In
21 section 3.3.1, the gain bandwidth of the LGS crystal was calculated to span from $8.2\ \mu\text{m}$
22 to $11\ \mu\text{m}$, which corresponds to a $1.13\text{-}1.18\ \mu\text{m}$ signal spectrum. Therefore, we
23 target at the seed generator should broaden the spectrum of the $1.03\ \mu\text{m}$ wave to at least
24 $1.18\ \mu\text{m}$.

Several solid materials such as the Sapphire (Al_2O_3), Quartz and YAG crystal could support the ps-level, $1.03\mu\text{m}$ pulses driven SCG (Table 3.4). Among all these crystals, YAG crystal has the highest nonlinear refractive index n_2 and the highest damage threshold [112] according to Table 3.4. The reported SCG could extend its bandwidth to octave-spanning, which is far wider than the target. Therefore, we use the YAG crystal in the seed generator.

Table 3.4. Summary of the nonlinear medium for the SCG at $1.03\mu\text{m}$ [97].

	n_0	n_2 (10^{-16} cm^2/W)	P_{cr} (MW)	Damage threshold (J/cm^2 , 1030nm , 500fs) [112]
Quartz	1.4500	3.06	3.5866	4.85
Sapphire	1.7551	2.9	3.1266	5.36
YAG	1.8153	7.3	1.2009	7.5

In order to determine the optimized thickness of the YAG crystal, an experiment is conducted with the schematic shown in the Fig.3.4. We use 1ps, $80\mu\text{J}$ pump pulse with p-polarization to drive the seed generator. The pump beam pass through an iris which is used to control its Rayleigh range and intensity. The pulse energy behind the iris is optimized to be $14\mu\text{J}$ in order to generate a stable filamentation in the solid. The pump beam is then focused by an $f=100\text{mm}$ lens located right after the iris. The YAG crystal is put behind the focal spot to avoid crystal damage and the generation of the unstable multi-filamentations. After passing through the crystal, the SC output is recollimated via a $f=50\text{mm}$ lens. The required frequency components with p-polarization are then extracted from the intensive fundamental wave by using a high reflective mirror at $1.03\mu\text{m}$. Here, the transmission curve of the high reflective mirror is shown in the Fig.3.4(b). It has extreme low transmittance for the $1.03\mu\text{m}$ fundamental beam but $>50\%$ transmittance for the $>1.1\mu\text{m}$ frequency components. Therefore, it could be used for extracting the SC pulses apart from the intensive fundamental pulses.

Fig.3.4. (a) Schematic of the SC experiment; L1, L2: CaF2 lenses with $f=100\text{mm}$ and $f=50\text{mm}$, respectively; HR: high reflective mirror. (b) The transmittance curve of the HR mirror.

According to Eq.3.2, the thickness of the YAG crystal L should support enough self-phase modulation to expand the maximal wavelength at least to $1.18\text{ }\mu\text{m}$. In this experiment, we measure the spectra and the beam profiles of the SCG in the 4mm, 8mm, 12mm and 16mm YAG materials, respectively. The measured spectra of the SCGs are shown in the Fig.3.5(a). From the Fig.3.5(a), we can obviously see that the spectral bandwidth of the SCG increases with the material thickness. The spectrum could be broadened to above $1.18\text{ }\mu\text{m}$ at -10dB level when the crystal length is larger than 8mm. Also, we measure the output beam profiles in each case as shown in the third images in Fig.3.5(b)-(e). The beam profiles of the SCG at $>1.1\text{ }\mu\text{m}$ spectral range are measured via the NIR camera (Xeva-Lin-1.7, Xenics). We could also observe that when the crystal length is as large as 16mm, the beam profile shows an obvious ring-shape. As reported in Ref.[113], this ring-shape beam profile could be explained by result of the plasma absorption. At the leading edge of the pulse, intensive electric field excited plasma with high density. Since the beam has a gaussian shape intensity profile, therefore the intensity is higher on the optical axis. As a result, when the central and trailing part of the pulse pass through the high-density plasma, serious on-axis absorption occurs so that the intensity profile in transverse will look like a ring-shape. The poor quality of the beam profile will affect both the alignment of the signal and

the efficiency of power amplification. Therefore, by considering both SC spectrum and the beam profile, using 12mm YAG crystal is a promising choice for generating the SCG. It could provide a broadband seed with enough spectral bandwidth, good beam quality and high beam stability which could be verified by the photos of the stable conical emission profile and filamentation shown in Fig.3.5(b)-(e).

Fig.3.5. Plot of the (a) SC spectra in the 4mm (black dot line), 8mm (red dash-dot line), 12mm (blue dashed line) and 16mm (orange line) YAG crystals, and the images of the conical emission, filamentation, and NIR beam profile measured at the back surface of the (b) 4mm, (c) 8mm, (d) 12mm, (e) 16mm YAG crystals.

3.3.3 Pulse stretcher

In an OPCPA, the broadband seed pulse should be stretched to match its pulse duration with the pump pulse. Normally, the pulse stretching could be realized via bulk materials, prism or grating pairs and the more AOPDF. In the LWIR OPCPA, the bulk material

1 is used to stretch the pulse through material dispersion due to its simplicity and high
2 transmittance.

3 The optimized seed pulse duration under the maximum efficiency-bandwidth-product
4 condition has been studied in Ref. [114]. According to the Ref. [114], the optimized
5 pulse duration is determined by the input pump-signal energy ratio. For the LWIR
6 OPCPA, the input pump-signal energy ratio is $\sim 10^6$ at the pre-amplification stage and
7 $\sim 10^3$ at the main-amplification stage. Therefore, the optimized seed pulse duration is
8 calculated as 600fs in the pre-amplification stage and 900fs in the main amplification
9 stage. In order to achieve this goal, $\sim 15000 \text{ fs}^2$ GDD should be added to seed pulses
10 before the first stage and then extra $\sim 5000 \text{ fs}^2$ should be provided before the second
11 stage.

12 Apart from the GDD, the third order dispersion (TOD) is also need to be compensated
13 by bulk material-based pulse stretcher. However, the TOD is always positive in the
14 bulk material-based stretcher and other crystals in OPCPA so that it cannot be
15 compensated. In order to suppress the effect of TOD, the pulse stretcher should be able
16 to provide as small TOD as possible.

17 In the NIR regime, the commonly used bulk material stretcher includes the sapphire,
18 the calcium fluoride (CaF_2), the barium fluoride (BaF_2), the Quartz (SiO_2) and the zinc
19 selenide (ZnSe). The properties of these materials are summarized in Table 3.5. Among
20 all the materials, the ZnSe has the smallest TOD-to-GDD ratio. As a result, when the
21 target GDD is achieved, the ZnSe based pulse stretcher could provide the smallest TOD.
22 Moreover, the GDD provided by the ZnSe per millimetre is over ten times larger than
23 other materials. Therefore, the required thickness of ZnSe is the shortest among all
24 candidates which is favoured for a compact design.

25 In order to provide $\sim 15000 \text{ fs}^2$ GDD before the pre-amplification stage and $\sim 5000 \text{ fs}^2$
26 GDD before the second stage, we use a 30mm thick ZnSe crystal and a 10mm thick

1 ZnSe crystal in the first and the second pulse stretcher respectively. In order to reduce
 2 the Fresnel loss, both ZnSe crystals are fabricated with 1-1.3 μm anti-reflective coating.

3 Table 3.5. Summary of the bulk material for the pulse stretcher.

	Refractive index (@1.163μm)	GDD (fs²/mm, @1.163μm)	TOD (fs³/mm @1.163μm)	TOD/GDD ratio
Sapphire	1.752	17.764	80.169	4.513004
CaF2	1.428	13.725	24.667	1.797231
Quartz	1.448	8.975	56.115	6.252368
ZnSe	2.474	556.439	475.202	0.854006
BaF2	1.468	22.756	23.232	1.020918

4 3.3.4 Amplification stage

6 In the OPCPA system, the energy of the seed is boost-up in the amplification stage.
 7 However, the parametric gain is limited by the damage threshold, aperture size and
 8 length of the nonlinear medium. In this experiment, the damage threshold of the LGS
 9 crystal is measured to be 50GW/cm² using 1ps pulses at 1.03 μm . Therefore, the
 10 maximum pump energy that could be safely applied on each crystal is ~1.5 millijoule.
 11 The apertures of the LGS crystals are limited by 5mm $\times$ 5mm. The thicknesses of the
 12 available LGS crystals are all less than 3mm.

13 Under these limitations, the maximal parametric gain in a single crystal could be
 14 estimated via simulation. The simulation is applied by numerically solving the coupled
 15 pulsed-wave equations demonstrated in chapter 2. In the simulation, the thickness of
 16 the LGS crystal is set to be 3mm. The input pump pulses have 50GW/cm² intensity,
 17 1.5mJ pulse energy and 1ps pulse duration. The pulse width of the 1.163 μm signal
 18 pulse is chirped to match the 1ps duration of the pump pulse with 30000fs² GDD. Here,

we consider two cases, the weak signal amplification with 10^6 level pump-signal energy ratio at the input, and the large signal amplification with 10^3 level input pump-signal energy ratio. The simulated parametric gain and spectra of the LWIR idler in both cases are shown in the Fig.3.6(a) and (c), respectively.

Fig.3.6. Simulation results for the parametric gain in (a) single LGS crystal with 3mm thickness and (b) two cascaded LGS crystals with 3mm and 2mm thickness respectively, and the support idler spectrum in (c) single LGS crystal and (d) two cascaded LGS crystals, under different pump-signal energy ratio U_p/U_s . Here U_p and U_s represent for the input pump and signal pulse energy, respectively.

According to the simulation results, the parametric amplification in both cases are far away from saturation regime. The parametric gain in the weak signal amplification case is 712. In the large signal case, the signal pulse energy has been amplified by only 33 times. Therefore, even if a two-stage parametric amplification is implemented, the

1 pulse energy of the amplified signal is still limited at $23\mu\text{J}$, which corresponds to a few
2 micro-joule level LWIR pulse energy.

3 In order to enhance the parametric gain, we design the amplification stage with two
4 LGS crystals placed in tandem. The residual pump energy after passing through the
5 first crystal could thus be re-used in the second one. The only limitation of the
6 parametric gain in the second crystal is the temporal walk-off between the pump pulses
7 and the signal/idler pulses. However, the group velocity mismatches between the pump
8 and signal, and between pump and idler are calculated as 6.67fs/mm and 50fs/mm
9 respectively, which could be ignored compared with the picosecond-level pulse
10 duration. For illustrating the advantage of this design, we conduct another simulation
11 with the similar settings as the previous case. In this simulation, another 2mm thick
12 LGS crystal is added after the 3mm thick one. The $\sim 30\%$ loss of the pump energy due
13 to the Fresnel reflection is also considered. The simulated parametric gain and spectra
14 of the LWIR idler are shown in the Fig.3.6 (b) and (d), respectively.

15 The simulation results show that, by sequentially placing two crystals, the gain factor
16 is increased to 20400 and 68 in the 10^6 and 10^3 ratio cases, respectively. Under this
17 situation, it is expected to obtain the 1.3mJ signal pulse energy which corresponds to a
18 $100\mu\text{J}$ level LWIR output. Compared with the single crystal case (Fig.3.6(c)), the
19 spectrum of the LWIR pulses could still span from $8\mu\text{m}$ to $10\mu\text{m}$. It should be noticed
20 that, for the 10^3 pump-signal energy ratios (red dashed line in Fig.3.6(d)), the
21 bandwidth of the LWIR spectrum is even broader due to the gain saturation. As a result,
22 we can conclude that the enhancement of the parametric gain via re-using the pump
23 energy is not only achievable but also advantageous.

24 **3.3.5 Compressor**

25 During the OPCPA process, the chirped signal pulses are amplified by the un-chirped
26 pump pulses. At the same time, the chirped idler pulses are generated. Take the positive

1 chirped signal pulses as an example, the longer wavelength components are located at
 2 the leading edge of the pulse while the shorter wavelength components located at the
 3 trailing edge. According to the energy conservation law, shorter wavelength
 4 components will be generated at the front of the LWIR idler pulse and longer
 5 wavelength components are located at the back. Therefore, the LWIR idler is regarded
 6 as a negative chirped pulse. According to the solution of the coupled pulsed-wave
 7 equations Eq.2.14, the spectral phase of the LWIR pulse could be written as,

$$11 \quad \phi_i = \frac{\pi}{2} - \phi_{s0} - k_i(\omega_i)L - \frac{\Delta k}{2}L \quad (3.3)$$

8 Here, we assume a collinear phase matching geometry and a zero pump-surface normal
 9 angle α which are in accordance with the experiment. Therefore, the GDD and TOD
 10 of the idler pulse could be calculated as,

$$12 \quad \text{GDD}_i = \frac{\partial^2 \phi_i}{\partial \omega_i^2} = -\text{GDD}_{s0} + \text{GDD}_{ip} + \text{GDD}_{OPP}$$

$$13 \quad \text{TOD}_i = \frac{\partial^3 \phi_i}{\partial \omega_i^3} = \text{TOD}_{s0} + \text{TOD}_{ip} + \text{TOD}_{OPP} \quad (3.4)$$

14 where the subscripts s0, ip and OPP represents for the GDD or TOD introduced by the
 15 input signal pulse, the propagation of idler pulse and the optical parametric phase (OPP).
 16 The third term on the right-hand side of Eq.3.4 could be written as,

$$17 \quad \text{GDD}_{OPP} = \frac{1}{2} \left(\frac{\partial^2 k_i}{\partial \omega_i^2} + \frac{\partial^2 k_s}{\partial \omega_s^2} \right) L$$

$$18 \quad \text{TOD}_{OPP} = \frac{1}{2} \left(\frac{\partial^3 k_i}{\partial \omega_i^3} - \frac{\partial^3 k_s}{\partial \omega_s^3} \right) L \quad (3.5)$$

19 This set of equations is different from the calculation used in NIR and SWIR OPCPAs
 20 [38] due to the emergence of the OPP. According to Eq.3.4, the GDD and TOD of the

output idler pulses could be calculated to be -33383fs^2 and 186387fs^3 . Each part of the contribution of the GDD and TOD are shown in Table 3.6. For a four-cycle, 130fs pulse at $9\mu\text{m}$ supported by the gain bandwidth, the residual GDD and TOD could stretch its pulse duration to $\sim 780\text{fs}$, 26 optical cycles. However, by only considering the residual 186387fs^3 TOD, the $9\mu\text{m}$ pulse can still maintain its pulse duration to less than 140fs, sub-five optical cycles. Therefore, for the pulse compressor, we mainly target at compensating the residual GDD of the LWIR pulses.

Table 3.6. Dispersion calculation of the LWIR OPCPA system.

		GDD (fs^2)	TOD (fs^3)
GDD_{s0} and TOD_{s0} at $1.163\mu\text{m}$	40mm ZnSe stretcher	22228	18954
	Propagation in pre-amp stage LGS	701	518
	OPP in pre-amp stage LGS	4862	93824
	Other materials	1298	4186
GDD_{ip} and TOD_{ip} at $9\mu\text{m}$	Main-amp stage LGS	-8455	132080
	Ge beam splitter	423	2503
GDD_{opp} and TOD_{opp} at $9\mu\text{m}$	Main-amp stage LGS	3737	-65678
Total dispersion GDD_i and TOD_i (eq.3.5)		-33383	186387

Normally, the dispersion compensation could be reached through the bulk material, the grating and prism pairs and the acousto-optic programmable dispersive filter (AOPDF).

1 Among all the candidates, the AOPDF methods are favoured for compensating both
2 GDD and TOD. This technique could provide either positive or negative GDD and TOD
3 within a wide range. However, due to the low damage threshold of the acoustic-optic
4 crystal, this device could only be located before the amplification stage for pre-
5 compensation of dispersion. The TOD added by the AOPDF could affect the
6 amplification efficiency in the amplification stages. Moreover, this device has <15%
7 efficiency which limit the energy boost-up of the LWIR OPCPA system. The
8 simultaneously compensation of the GDD and TOD could also be reached by designing
9 a double-pass, dual-prism compressor. However, in order to compensate 33383fs^2 at
10 $9\mu\text{m}$, the distance between two prisms could be as large as several meters. For example,
11 by using two Germanium prisms with 28.037° apex angle and 75.982° angle of
12 incidence (AOI), the prism separation L could be as large as $\sim 2.5\text{m}$ for providing
13 33383fs^2 GDD and -116093fs^3 TOD. The beam spot size at the surface of the second
14 prism will be $\sim 70\text{mm}$ for 8 to $10\mu\text{m}$ spectral range so that a prism with extreme large
15 aperture is required. As a result, this design is not only incompact but also hard to be
16 implemented.

17 Considering about all the situations above, we use the bulk material compressor instead
18 due to its simplicity and compactness although the TOD cannot be compensated. The
19 suitable candidates of the bulk material compressor should have at least large
20 transmittance in the $<11\mu\text{m}$ LWIR spectral range. Also, it should provide positive GDD
21 in order to compensate the negative GDD_i . Among all the normally used bulk materials,
22 the germanium crystal fulfils all the requirements. By using the 78.75mm thickness,
23 Brewster-angle cut germanium window, the GDD_i could be fully compensated. The
24 extra TOD introduced by the germanium is $1.97 \times 10^5\text{fs}^3$. Together with the residual
25 $1.86 \times 10^5\text{fs}^3$ TOD of the OPCPA process, the total TOD is calculated as $3.83 \times 10^5\text{fs}^3$
26 which could stretch a 130fs transform limited pulse at $9\mu\text{m}$ to 145fs, as shown in
27 Fig.3.7. As a result, although the TOD is not compensated, we can still expect to
28 achieve a sub-five-cycle LWIR pulse from the OPCPA.

Fig.3.7. Comparison between a 130fs transform limited pulse at 9 μ m (blue line) and the stretched pulse with 383539 fs³ residual TOD (red line).

3.4 Implementation of 9 μ m OPCPA

3.4.1 9 μ m OPCPA setup

Based on the design of the LWIR OPCPA, we develop the OPCPA system as shown in the Fig.3.8. The whole system, including the seed generator and two amplification stages, is driven by a commercialized 1.03 μ m Yb:YAG laser system (AMPHOS 100). The pump source could provide at most 13mJ energy with 10kHz repetition rate, 130W average power and 1ps pulse duration. Due to the limited damage threshold ($\sim 50\text{GW}/\text{cm}^2$, at 1.03 μ m, 1ps [74]) and the finite crystal aperture (7mm $\times$ 7mm), only $\sim 2.5\text{mJ}$ pump energy is used driving the OPCPA. The total energy of the pump laser is distributed into three stages via the combination of the half wave plate (HWP) and the thin film polarizer (TFP).

Fig.3.8. Schematic of the LWIR OPCPA system.

The broadband seed pulses at $1.163\ \mu\text{m}$ are generated through the supercontinuum generation in a 12mm YAG crystal. The generated seed pulses have $\sim 2\text{nJ}$ pulse energy and a broadband spectrum spanning from $1.1\ \mu\text{m}$ to $>1.3\ \mu\text{m}$, which is enough to support the gain bandwidth. Due to the optimization of the crystal thickness, the seed beam also have good Gaussian beam profile and high beam stability.

The $1.163\ \mu\text{m}$ seed beam is first stretched in a 30mm ZnSe-based pulse stretcher and then amplified in the pre-amplification stage. After that, the amplified seed pulses are further stretched in a 10mm thick ZnSe material and boost-up its energy in the main-amplification stage. The pulse durations of the seed pulses in both stretchers are measured by a commercialized frequency-resolved optical gating (FROG) apparatus. The retrieved FROG traces and the temporal profiles of the stretched seed pulses are shown Fig.3.9. The seed pulse durations are measured to be 560.6fs and 859fs after the first and second stretcher respectively, which are basically fulfil the requirement. Both pulse stretchers have high transmittance which is larger than 90%.

Fig.3.9. The measured and retrieved Frog trace of (a) the seed pulses after the 1st stretcher and (c) the seed pulses after the 2nd stretcher, and the retrieved temporal profiles of (b) the seed pulses after the 1st stretcher and (d) the seed pulses after the 2nd stretcher.

The stretched seed pulses are amplified consecutively in the pre-amplification stage and the main-amplification stage. In both stages, we sequentially placed two LGS crystals to reuse the residual pump energy. In the pre-amplification stage, two LGS crystals with 3mm and 2mm thickness are used. The s-polarized input signal pulses with 2nJ combine with the p-polarized, 1.2mJ, 43.6GW/cm² peak intensity pump pulses via the thin film polarizer (TFP). The signal pulse energy then is amplified to 240nJ in the first 3mm thick crystal and then further amplified to 10 μ J through another 2mm thick LGS crystal. It should be noted that, due to the Fresnel loss, the pump energy that reach the second crystal is only 0.95mJ. The parametric gain in the first amplification stage could reach ~5000 times. The amplified signal pulses are then separated from the intensive pump pulses through the TFP. After the second stretcher,

Fig.3.10. Simulation results for the parametric gain in the pre-amplification stage (Pre-amp, first crystal: black line, second crystal: blue line) and the main-amplification stage (Main-amp, first crystal: red line, second crystal: yellow line).

the signal pulses enter the main amplification stage for further energy enhancement. The main amplification stage is driven by 1.3mJ pump pulses with $47.2\text{GW}/\text{cm}^2$ peak intensity. By passing through a 2mm thick LGS and a 1.5mm thick LGS consecutively, the signal pulse energy finally could be amplified to 155 μJ with 15.5 times parametric gain. The parametric gain is smaller than the calculated one demonstrated in section 3.3.4 which could be explained by two possible reasons. First, there is ~30% energy loss of both signal and idler pulses in each amplification stage which is not taken into consideration in the simulation. The energy loss is introduced by both Fresnel reflection and scattering due to the material impurity. Second, two thinner LGS crystals (2mm+1.5mm) are adopted in the main amplification stage compared with the crystals (3mm+2mm) we used in the simulation. Therefore, we simulate the OPCPA process again with considering the actual crystal length and the energy loss. The simulation result is shown in Fig.3.10. The amplification in the two crystals of the pre-amplification stage is shown by the black and blue line. The amplification in the two crystals of the main-amplification stage is demonstrated by the red and yellow line. The sudden change of the parametric gain is introduced by energy loss of the signal

1 due to the Fresnel reflection of the second crystal. The calculated gain in the pre-
2 amplification and main-amplification stages are 6555 and 19.4 times respectively,
3 which are similar with the experiment.

4 The spectra of the input signal and the amplified signal in both stages are also measured
5 as shown in Fig.3.11. The measurement results show that the amplified spectra in both
6 stages span from 1.14-1.18 μm , corresponding to 8.1-10.6 μm wavelength range of idler.
7 It agrees well with the calculated gain bandwidth which covers 1.13-1.18 μm spectral
8 range. No significant gain narrowing is found according to the measured spectra.

9
10 Fig.3.11. The spectrum of signal pulses measured at the output of (a)the seed generator; (b)
11 the pre-amplification stage, and (c) the main-amplification stage.

12 Accompanied with the signal amplification, the p-polarized idler pulses are also
13 generated in the main amplification stage. By using a 1mm thick, Brewster-angle-cut
14 Germanium (Ge) window as the beam splitter, the idler pulses could be split with both
15 pump and signal pulses. It should be noted that, half of the pulse energy of the pump
16 and signal is absorbed by the Ge beam splitter. Therefore, the accumulated heat should
17 be removed to avoid damage the beam splitter. Here, we apply a dry air-cooling system
18 at the back surface of the Ge window in order to dissipate the accumulated heat. There
19 is no crystal damage found in the beam splitter.

1 At the output of the OPCPA, the idler pulses are compressed in the 78.75mm Ge
2 window to 145fs pulse duration with the GDD total compensated. The transmittance
3 of the pulse compressor is over 90%. The final output energy of the LWIR OPCPA is
4 14μJ. The total amplification bandwidth of the idler could be as large as 2000nm which
5 could support ~4 optical cycle pulse width at 9μm. The total conversion efficiency of
6 the OPCPA system is ~0.6% which is still need to be improved.

7 **3.4.2 Performance of 9μm OPCPA**

8 The performance of the LWIR OPCPA is experimentally characterized after the pulse
9 compressor. The properties include the pulse energy and its long-term stability, the
10 beam profile, the spectrum and the temporal profile. All measurement results are
11 demonstrated in Fig.3.12. Here, the pulse energy of the LWIR pulse is measured by a
12 digital power meter from Thorlabs with a thermal sensor. The spectrum at LWIR
13 regime is measured by using a scanning-grating monochromator with a liquid-nitrogen-
14 cooled MCT detector. For the temporal profile measurement, traditional frequency-
15 resolved optical gating (FROG) technique could not be applied here due to the lack of
16 LWIR spectrometer with small acquisition time. As a result, a home-made
17 interferometric autocorrelator (IAC) is used. In the IAC apparatus, the interferometric
18 LWIR signal excites the second harmonic (SH) pulse in a 0.5mm GaAs crystal. The
19 second harmonic output is measured by an InAsSb amplified photodetector (Thorlabs
20 PDA10PT) which has high responsivity within the spectral range of 1.0-5.8 μm. Details
21 of the basic theory and setup of the IAC is demonstrated in the Appendix II. The electric
22 field of the measured pulse is numerically reconstructed through a genetic algorithm
23 based ‘evolutionary phase retrieval from interferometric autocorrelation’ (EPRIAC)
24 algorithm [115]. The electric field is retrieved by using the measured IAC trace and the
25 spectrum.

26 **1) Stability and scalability**

1 The output energy of the LWIR pulses is measured to be 14 μJ , corresponding to the
2 140mW average power. The long-term power stability of the LWIR pulse is also
3 measured for 1 hour, as shown in Fig.3.12 (a). The power stability is characterized by
4 the ratio between the standard variation value and the averaged value of the output
5 power. The measurement result shows a 1.6% rms instability within 1 hour. Also, a
6 stable, Gaussian shaped beam profile is also measured as shown in the inset of Fig.3.12
7 (a). Both results show the stability of the amplification in the OPCPA.

9 Fig.3.12. Properties of the output LWIR pulses with considering: (a) Energy long-term
10 stability measured in 1-hour (the beam profile is demonstrated at the inset); (b) Energy
11 scalability; (c) Spectrum; (d) Measured (black dashed line) and retrieved (red line) IAC trace;
12 (e) Retrieved temporal profile.

13 The variation of the idler pulse energy with different pump intensity is also measured
14 at the main amplification stage, which is shown in Fig.3.12 (b). Obviously, the input-
15 output energy curve shows a linear growth of the idler pulse energy. It implies that the
16 OPCPA system is still operating at the non-saturation region so that the idler pulse

1 energy can still be boost up. In the current OPCPA, the energy scalability is mainly
2 limited by the LGS thickness (3mm) and the energy loss of the pump pulses. The anti-
3 reflective coated LGS crystal with 8mm thickness is now available to further enhance
4 the output energy of the OPCPA.

5 **2) Spectrum**

6 The spectrum of the LWIR pulse is shown in Fig.3.12 (c). It shows an 8 to 10 μm
7 spectrum with $\sim 2000\text{nm}$ bandwidth. The measured bandwidth is closed to the
8 calculated 2800nm gain bandwidth with spectrum covering $8.2\text{ }\mu\text{m}$ to $11\text{ }\mu\text{m}$. The
9 bandwidth narrowing is mainly due to the low parametric gain of the wavelength
10 components located at the wings of the chirped signal pulses [114]. The measured
11 LWIR spectrum could support a 130fs transform limited pulse duration which
12 corresponds to sub-five optical cycles. The LWIR spectrum is also verified by the
13 simulation where the coupled pulsed-wave equation is numerically solved. In the
14 simulation, the energy, peak intensity, pulse duration and the dispersion of the input
15 pump and signal pulses at each stage are all the same with the real experiment. The
16 Fresnel loss for the pump energy in each stage is also taken into consideration. As
17 shown in the Fig.3.12 (c), the simulated spectrum matches well with the measured one.

18 **3) Temporal profile**

19 The IAC trace of the LWIR pulses is plot in Fig.3.12(d). The 1:8 ratio between the
20 background and the maximal of the IAC trace proves the reliability of the measurement
21 result. For the comparison, the retrieved trace is also shown with the red line. The error
22 indicator of the retrieved IAC trace equals to 0.1697 according to the Eq.2 in Ref.[113]. The
23 discrepancy between the retrieved trace and the measured one mainly comes from the intensity
24 noise of the LWIR pulses, the fluctuation of the beam pointing and the unperfect alignment of
25 the IAC apparatus. The retrieved pulse shows a 145fs , 4.8 optical cycles pulse width (Fig.3.12
26 (e)). The retrieved pulse width corresponds to 1.08 times transform limited pulse duration. The

1 side lobe of the pulse is introduced by the residual $\sim 32.6 \times 10^4 \text{ fs}^3$ TOD according to the phase
2 retrieval. This value is closed to the calculated $35.3 \times 10^4 \text{ fs}^3$ value as demonstrated in section
3 3.3.5. It mainly comes from the dispersion of all bulk material in the route of signal and idler
4 propagation. The low retrieve error shows that the GDD of the idler pulses in the OPCPA
5 has been fully compensated.

6 **3.5 Conclusions**

7 In this chapter, we demonstrate the design and implementation of the LWIR which
8 operates at $9\mu\text{m}$ wavelength. The key challenges of developing LWIR OPCPA are
9 revisited. We found that, the problems of the limited gain bandwidth can be solved by
10 using the large band gap LGS crystal in the amplification stages. At the same time, we
11 can still ensure the energy boost-up of the OPCPA since the energy down-conversion
12 from the advanced $1\mu\text{m}$ pump laser to the LWIR wavelength is still supported by using
13 this crystal.

14 Based on the LGS and $1\mu\text{m}$ pump laser, we numerically design each component of the
15 LWIR OPCPA system. The broadband signal pulses are generated through SCG in
16 YAG crystal. The thickness of the YAG is optimized. In the amplification stage, we
17 designed a double-crystal structure to reuse the pump energy and enhance the
18 parametric gain. For the chirp compensation, the GDD of the significant OPP is taken
19 into consideration and fully compensated. Based on the design, a LWIR OPCPA is
20 experimentally implemented. The OPCPA provides $9\mu\text{m}$ pulses with $\sim 2000\text{nm}$
21 amplification bandwidth, $14\mu\text{J}$ pulse energy and 140mW average power. Both the
22 parametric gain and the bandwidth match the simulation very well. Thanks to the careful
23 dispersion management, we fully compensate the group delay dispersion (GDD) and
24 minimized the effect of third order dispersion (TOD) of the LWIR pulse. The compressed
25 pulse duration could reach 145fs , sub-five optical cycles. The cycle number is 1.67
26 times shorter than the reported $7\mu\text{m}$ [23] OPCPA which shows the ability of the large band

1 gap material in achieving large gain bandwidth. The output LWIR beam also has good
2 energy stability and beam quality.

3 Without a doubt, we have to admit that the efficiency of the OPCPA system is still
4 limited at 0.6%. However, since the current OPCPA system is still operating at the
5 linear amplification region with large amount of energy loss, further efficiency scale-
6 up is still straightforward by using thicker crystal with AR-coating at the input surface.
7 Therefore, we [can](#) still expect the LWIR OPCPA to get higher energy and power in the
8 near future. Besides, the pulse duration of the LWIR pulse is still limited at 5 optical
9 cycle due to the gain bandwidth of the LGS crystal. In order to generate a single cycle
10 pulse at LWIR wavelength, an extra compression stage should be added. This pulse
11 compression stage will be demonstrated in the next chapter.

12

Chapter 4. LWIR Pulse Self-compression in Solid

4.1 Introduction

In Chapter 3, the 9 μ m pulses with microjoule level pulse energy are generated through the OPCPA system. However, the pulse duration of the LWIR pulse is still limited at five optical cycles which limit their applications such as isolated attosecond pulse generation and time-resolved molecular spectroscopy. In these applications, near-single-cycle pulses are required. Therefore, we focus on further compressing the LWIR pulse to single-cycle regime in this chapter.

We successfully generate the near-single-cycle LWIR pulses with multi-octave-spanning spectrum through pulse self-compression in solid materials. The plasma effect in the self-compression is also explored through experiment and simulation. This work can be considered as an experimental complement to the current theoretical results [45,116,117] on LWIR pulse self-compression in solid materials.

The contents of this chapter are arranged as follows. In section 4.2, a short review about single-cycle pulse compression and self-compression is given. Section 4.3 summarizes the experiment and simulation conditions of our works on LWIR self-compression. In section 4.4, a systematic study of the supercontinuum generation (SCG) is presented where the pulse self-compression is demonstrated in three different anomalous dispersive materials such as KrS-5, ZnSe and GaAs. Section 4.5 concludes the chapter by addressing the effect of the filamentation in the self-compression process.

4.2 Literature review of single-cycle pulse compression

The most common method of shortening the pulses to the near-single-cycle regime is generating a broadband spectrum followed by pulse compression. Intensive efforts have been made along this routine. A broadband spectrum can be generated by the four-

1 wave-mixing through filamentation in gases [118], coherent pulse synthesis [76] and
2 the intra-pulse DFG [119]. Since the spectral bandwidth is broadened, the pulse
3 duration can be further reduced according to the uncertainty law. Further pulse
4 compression is achieved through material dispersion.

5 In contrast to this method, the spectral broadening and the pulse compression can be
6 both reached within one step. This simpler method is called the pulse self-compression
7 in the anomalous dispersive medium. The pulse self-compression is realized by joint
8 action of self-phase modulation (SPM) and anomalous dispersion. The SPM process is
9 responsible for spectrum broadening and generation of new wavelengths. In the SPM,
10 long wavelength components are generated at the leading edge of a pulse and move
11 towards the trailing edge of the pulse due to anomalous dispersion. On the contrary,
12 the short wavelength components that are generated at the trailing edge swept to the
13 front when propagating (Fig.4.1). As a result, the pulse is compressed in time,
14 meanwhile its spectrum is broadened. This simple and direct technique has been
15 realized in both gases [42,120,121] and solids [122-126] driven by the SWIR/MWIR
16 laser sources. For example, the pulse self-compression is achieved in CaF₂, BaF₂
17 crystals by using the 2.1 μm laser as a pump [122]. A high efficient self-compression
18 method is also demonstrated in the YAG crystal [127] driven by the 3.9 μm pump
19 source. This highly efficient self-compression method provide sub-three-cycle pulses
20 with >93% efficiency and 19.7mJ pulse energy [127].

Fig.4.1. Illustration of the pulse self-compression process.

Generation of sub-two-cycle self-compressed pulses at $\sim 3\mu\text{m}$ in the YAG crystals are reported by Zou Xiao et al [128] and Faming Lu et al. [124]. With the cascaded OPA, DFG and the ionization-assisted self-compression process in the near-zero dispersion wavelength regime, single cycle pulses at $7.9\mu\text{m}$ have been generated [71]. However, the research of the pulse self-compression in LWIR regime is still in its infancy. The soliton-based self-compression and filamentation in air, driven by the picosecond CO₂ laser, have been theoretically [129] and experimentally [130] studied. SCG and self-compression of picosecond CO₂ laser in anomalous dispersive solid materials were experimentally studied without considering the filamentation effect. ([131,132].) Recently, several theoretical works [45,116,117] have predicted that more efficient self-compression is possible by plasma-assisted nonlinear propagation. For example in [45], a theoretical study of single-cycle self-compression of LWIR pulses in solids was reported by leveraging the combination effects of plasma defocusing, nonlinear absorption, and self-steepening. However, there are still no experimental studies and results reported on the plasma-assisted self-compression in solid material driven by LWIR pulses.

4.3 Pulse self-compression driven by the developed $9\mu\text{m}$ OPCPA

4.3.1 Experiment testbed for studying pulse self-compression in different materials

A general experimental testbed is set up in order to carry out experiment study of LWIR SCG and self-compression in solids of different materials. The setup is based on the $9\mu\text{m}$ OPCPA system developed in chapter 3, as shown in Fig.4.2. The $9\mu\text{m}$ pulses with $8\mu\text{J}$ pulse energy and 145fs pulse duration are focused by an $f=38.1\text{mm}$ off-axis gold parabolic mirror. The focused beam passes through the anomalous dispersive material, represented by X, and generates ultra-broadband supercontinuum and self-compressed

1 pulses. The output beam is then re-collimated by another $f=20.8\text{mm}$ off-axis parabolic
 2 mirror for spectral and temporal measurement. Since the generated supercontinuum

3
 4 Fig.4.2. The schematic of the pulse self-compression experiment based on a $9\text{ }\mu\text{m}$ OPCPA,
 5 and the spectral and temporal characterization. L: Lens; BS: Germanium beam splitter; M:
 6 Silver mirror; P1, P2: Off-axis parabolic mirrors; X: Nonlinear crystal.

7 may cover multi-octaves, several types of lens, fibers, filters and spectrometers are used
 8 for different spectral range. For the $3.4\text{-}14\text{ }\mu\text{m}$ range, an $f=76.5\text{mm}$ parabolic mirror and
 9 a hollow core fiber are used for beam coupling. The spectrum is measured by an
 10 infrared scanning-grating monochromator with a liquid-nitrogen-cooled HgCdTe
 11 (MCT) detector as well as the $2.4\text{ }\mu\text{m}$, $4.5\text{ }\mu\text{m}$ and $7.3\text{ }\mu\text{m}$ long pass filters. For the 2-
 12 $3.4\text{ }\mu\text{m}$ range, we use an $f=76.5\text{mm}$ parabolic mirror for beam focusing, an InF_3 fiber
 13 for beam coupling, and an Optical Spectrum Analyzer for measuring the spectrum. For
 14 lower wavelength range, we use free space coupling with the $f=40\text{mm}$ CaF_2 lens in
 15 order to have higher coupling efficiency. The spectrum is measured by the NirQuest
 16 spectrometer for $1\text{-}2\text{ }\mu\text{m}$ range, the HR2000+ spectrometer for $0.43\text{-}1\text{ }\mu\text{m}$ range. The
 17 temporal profiles of the self-compressed pulses are measured by the same
 18 interferometric autocorrelator (IAC) as we used in chapter 3. A 5mm Germanium

1 window is used for pre-compensation of the material dispersion in the IAC. It is
 2 noteworthy that the generated spectra might cover the same frequency components as
 3 the SHG signal excited in the IAC. These frequency components will also be responded
 4 by the photodetector and thus distort the IAC trace. Therefore, they are removed by an
 5 BaF2 infrared polarizer added after the nonlinear crystal in the IAC.

6 **4.3.2 Materials considered in the experimental studies**

7 The experiment testbed is general for studying different materials by just simply
 8 putting a solid of the material at the position of the crystal X and replacing the relevant
 9 optics. In the study, the crystal X is chosen based on the following criterions: First, the
 10 crystal should have broad transmission range spanning from NIR to LWIR so that the
 11 whole supercontinuum spectrum could pass through it. Second, the material should
 12 also have suitable zero-dispersion wavelength (ZDW) which is smaller than 9 μ m. Third,
 13 large third-order nonlinear refractive index (n_2) is also required for strong self-phase
 14 modulation. Moreover, the critical power P_{cr} of the self-focusing is also need to
 15 considered, where the critical power [133] is calculated by,

$$16 \quad P_{cr} = \frac{3.77\lambda_0^2}{8\pi n_0 n_2} \quad (4.1)$$

17 where λ_0 is the central wavelength of the driving laser and n_0 is the refractive index at
 18 λ_0 . In order to generate the filamentation, the input power should surpass the critical
 19 power. The filamentation will further assist the spectral broadening in the self-phase
 20 modulation process as we will see in the next sessions. As a result, the critical power
 21 threshold should be lower than 55.2MW (10 μ J, 145fs), which is the peak power of the
 22 9 μ m laser.

23 The commonly used anomalous dispersive materials in the LWIR regime are
 24 summarized in the Table 4.1. Among all the materials, CaF2 and BaF2 have large

critical powers which exceed the peak power of the LWIR pulses. Both crystals also have small nonlinear refractive index n_2 . Therefore, they are unsuitable for the supercontinuum generation and pulse self-compression. There is also no sign of both filamentation generation and spectral broadening in these two materials according to the experimental measurements (Fig.4.3). In this experiment, we use a 2mm BaF2 and a 5mm CaF2. Various spectra are measured when changing the input intensity by tuning the distance between the focal plane and the front surface of the crystal. There is not spectral broadening observed no matter how intensity change, which proves that both materials are not suitable for this experiment.

Table 4.1. Summary of anomalous dispersive materials at 9 μ m. The zero-dispersion wavelength (ZDW) is calculated by the Sellmeier equation [134]. The critical power P_{cr} is calculated by Eq.4.1 [133].

	n_0 [98]	n_2 (10^{-16} cm ² /W)	ZDW (μ m)	GDD [98] (@9 μ m, fs ² /mm)	P_{cr} (MW)
ZnSe	2.4122	85 [135]	4.8	-818.73	5.9259
GaAs	3.4881	500 [97]	6.6	-619.16	0.697
KrS-5	2.3727	105 [136]	6.5	-182.25	4.877
BaF2	1.4145	5 [97]	1.9	-2155.7	171.8
CaF2	1.3268	3 [97]	1.6	-5230.6	305.3

Fig.4.3. Measured SC spectra in (a) 2mm BaF2 and (b) 5mm CaF2 with different distances between the front surface and the focal point.

On the contrary, Thallium Bromo-Iodide (KrS-5), Zinc-selenide (ZnSe) and Gallium Arsenide (GaAs), fulfil all the above-mentioned requirements. KrS-5 and ZnSe have similar nonlinear refractive indices but with different ZDW. As shown in Fig.4.4, ZnSe has a shorter ZDW at 4.8 μm . KrS-5 has a smaller GDD at 9 μm and a flatter dispersion curve in the whole spectral range from 4 to 14 μm range. GaAs has a similar ZDW as KrS-5 but a few times larger nonlinear refractive index.

All three materials are right candidates for the generation of filamentation, the SCG and the self-compression of the 9 μm pulses. Among all the materials, the critical power of KrS-5 and ZnSe is larger by 1/10 times than the input power, and the generation of single filamentation is ensured [133]. However, the critical power of GaAs is about 1/80 of the input power so that multi-filament is expected to be generated in GaAs, but this is unfavored. Therefore, we mainly focus on the SCG and self-compression in KrS-5 and ZnSe and consider GaAs as an opposite example in this chapter.

14

15

16 Fig.4.4. The material dispersion of the KrS-5 (Black line), ZnSe (Blue dashed line) and the

17

GaAs (Red dotted line).

1   4.3.3 Simulation testbed for studying pulse self-compression

2   In addition to the experimental studies, a simulation testbed is also set up in software
3   to simulate the spatio-temporal characteristics of the generated SC and the self-
4   compressed pulses. The simulation helps explanation of the experiment results. The
5   numerical simulations of pulse propagation are performed by solving a carrier-resolved
6   radially symmetric unidirectional pulse propagation equation (UPPE), which is given
7   by [45,137].

$$8 \quad \frac{\partial \tilde{E}}{\partial z} = \frac{i}{2k(\omega)} \nabla_{\perp}^2 \tilde{E} + iD\tilde{E} + \frac{i}{2k(\omega)} \frac{\omega^2}{c^2} \frac{\tilde{P}_{NL}}{\epsilon_0} - \frac{1}{2k(\omega)} \frac{\omega}{c} \frac{\tilde{J}(\rho)}{\epsilon_0} - \frac{\tilde{\alpha}_{NL}}{2} \quad (4.2)$$

9   The terms on the right-hand side of Eq.4.2 represent diffraction, dispersion, nonlinear
10   polarization, plasma effect and nonlinear absorption respectively. Here, $\tilde{E}$ is the electric
11   field in the frequency domain, the constant ϵ_0 is the permittivity of free space and the
12   c is the speed of light. The wavenumber is written as $k(\omega)$ and the dispersion term is
13   presented as $D = k(\omega) - \omega/v_g$, where v_g is the group velocity. $\tilde{P}_{NL}$ represents the
14   nonlinear polarization in the frequency domain. In this simulation, we only consider
15   the 2nd and 3rd order nonlinearities and ignore all other higher order nonlinear effects.
16   In particular, we describe the 2nd-order nonlinearity of ZnSe by using the same way as
17   Eq. 1 in Ref. [138] and randomly changing the effective nonlinear coefficient from 0 to
18   30 pm/V for every 70 μm distance. The free electron current in frequency domain is
19   denoted by $\tilde{J}(\rho)$ which is determined by the free electron density ρ . The evolution of
20   the electron density is described by [111],

$$21 \quad \frac{\partial \rho}{\partial t} = W(I)(\rho_0 - \rho) + \frac{\sigma_B I}{U} \rho - \frac{\rho}{\tau_r} \quad (4.3)$$

22   where $W(I)$ is the photoionization rate, ρ_0 is the neutral atomic density, σ_B is the
23   inverse bremsstrahlung cross section, U is the band gap of the material and τ_r is the

plasma recombination time. In this simulation, Eq.4.2 is solved by using the split-step method. The simulation parameters we used are summarized in Table 4.2. Here, the electron collision time in plasma is not a well-known parameter and thus researchers have used various values of electron collision time in condensed media [139]. We used 10 fs electron collision time for both media to match the experimental results with our simulations. The neutral atomic density ρ_0 is calculated by the following equation,

$$\rho_0 = \frac{\text{Density}}{\text{Molar Mass}} \times 6.023 \times 10^{23} \quad (4.4)$$

The density of the KrS-5, ZnSe and GaAs are 7.4 g/cm³ [144], 5.27 g/cm³ [145] and 5.32 g/cm³ [146], respectively. The molar mass of the KrS-5, ZnSe and GaAs are 615.58 gmol⁻¹ [144], 144.35 g/cm³ [145] and 144.645 g/cm³ [146] respectively.

Table 4.2. Simulation parameters for KrS-5, ZnSe and GaAs.

	KrS-5	ZnSe	GaAs
Input energy (μJ)	7	7.3	7
Input radius (1/e², μm)	122	188	401
Input pulse duration (fs)	145	145	145
2nd order nonlinear susceptibility^[98] (pm/V)	0	30	70
Grain size (μm)	-	70	-
U (eV)	2.5 ^[140]	2.82 ^[141]	1.42 ^[142]
ρ₀ (cm⁻³)	0.76×10 ²²	2.2×10 ²²	2.21×10 ²²
σ_B^[143] (cm²)	1.97×10 ⁻²²	1.97×10 ⁻²²	1.97×10 ⁻²²
τ_r (fs)	150	150	150
Electron collision time (fs)	10	10	10

1   **4.4 Experiment and simulation results**

2   **4.4.1 Pulse self-compression in ZnSe**

3   The supercontinuum generation and pulse self-compression are experimentally studied
4   in a 9mm, polycrystalline ZnSe crystal with 8μJ input energy. In the experiment, we
5   gradually increase the input peak intensity and observe the output spectrum and pulse
6   duration. We increase the input peak intensity by pushing the ZnSe crystal towards the
7   focal point step by step while keeping the input energy the same. The distance between
8   the front surface of the ZnSe crystal and the focal point is characterized by L , which
9   has a positive value when the ZnSe crystal is located in front of the focal point.

10   The experiment results are shown in Figure 4.5. In the experiment, the filamentation is
11   observed in ZnSe. At A and B positions ($|L|=6\text{mm}$) shown by the arrows in Figure
12   4.5(a), single filamentation start to be observed. Accompanied with the filament, the
13   output pulse energy is decreased as shown in Figure 4.5(b). The energy loss is
14   introduced by both the Fresnel loss (30%) from both surfaces and the plasma absorption
15   (25%-30%). When the crystal is moved to position C, the multi-filamentation
16   phenomenon occurs. Irregular intensity distribution is observed at the back end of the
17   ZnSe, as shown in the inset of Figure 4.5(b). The multi-filamentation phenomenon is
18   grown from the local noise in the beam intensity profile when high enough peak
19   intensity is applied [133].

Fig.4.5. Self-compression experiment in the ZnSe which include, (a) the SC spectra variation with different surface-focal point distance, (b) the output energy variation with different surface-focal point distance, and the IAC traces of the self-compressed pulses at (c) $L=9\text{mm}$, (d) $L=8\text{mm}$, (e) $L=7\text{mm}$, (f) $L=6\text{mm}$, (g) $L=5\text{mm}$ and (h) $L=4\text{mm}$.

As shown in Figure 4.5(a), we also observe significant spectrum broadening in the $4\text{mm} < |L| < 6\text{mm}$ region where single filament is generated. The bandwidth of the spectrum reaches its maximum when L is around $\pm 4\text{mm}$. For the $|L| < 4\text{mm}$ region,

1 reduction of the SC spectral bandwidth is also observed. This is due to the occurrence
2 of the multi-filamentation.

3 The temporal profiles of the output pulses are measured as the intensity increasing. It
4 should be noted that the strong second harmonic of the LWIR pulse generated in the
5 ZnSe could significantly destroy the IAC trace, since it covers the same spectral range
6 and polarization as the IAC signal. Therefore, a 7.3 μm LPF is added before the IAC
7 in order to suppress the strong SHG. The measured IAC traces are shown in Figure
8 4.5(c)-(h), corresponding to L decreasing from 9mm to 4mm. For the $L > 6\text{mm}$ region
9 where no filament is generated, the reduction of pulse duration is observed but varies
10 slowly. The reduction is only the result of the co-action of the self-phase modulation
11 and the anomalous dispersion. The compressed pulse duration is still maintained at 3.5-
12 4 optical cycles level (Figure 4.5(c)-(e)). However, for the $L < 6\text{mm}$ region where single
13 filament is generated, the pulse self-compression is significantly enhanced. We could
14 generate 3 cycle pulse at $L=6\text{mm}$, 2.5 cycle pulse at $L=5\text{mm}$ and 2 cycle pulse at
15 $L=4\text{mm}$.

16 As shown in Fig.4.5, the optimized spectrum broadening and pulse self-compression is
17 achieved at $L=4$. At that position, stable, several millimeters length filamentation is
18 clearly formed in the ZnSe crystal as shown in the photo Fig.4.6 (a). The complete
19 beam profile measured at the back surface of the dielectric is also demonstrated. It has
20 a stable Gaussian beam profile in the center part with no secondary beams observed,
21 which verifies the single filamentation formed in the ZnSe. Compared to the previously
22 reported self-compression experiment in the MWIR wavelength, for example the 2.1
23 μm [122], and the 3.1 μm [123] cases, although the Rayleigh range of the focused 9
24 μm beam is only 1/5 that of the 2.1 μm and 3.1 μm lasers, robust single filament with
25 multi-millimeter lengths could still be preserved.

Fig.4.6. Images of (a) the filamentation in the ZnSe and (b) the output beam profile.

The spectrum at $>-30\text{db}$ level and the full spectrum of the optimized SC in the ZnSe are shown in the Fig.4.7 (a) and (b), respectively. As demonstrated in Fig.4.7 (a), the spectrum could cover the spectral range from $1.2\text{ }\mu\text{m}$ to $14\text{ }\mu\text{m}$ at the -30db level and from $0.46\text{ }\mu\text{m}$ to $14\text{ }\mu\text{m}$ for the full spectrum, which corresponds to over five-octave spanning. The spectrum with the wavelength longer than $14\text{ }\mu\text{m}$ is not measured due to the lack of the long-pass filter with the longer cutoff wavelength. In the Fig.4.7 (a), a distinct dip at $4.25\text{ }\mu\text{m}$ could be observed in the spectrum which is attribute to the absorption of the CO_2 in the atmosphere. Moreover, the second harmonic peak is significant as shown in the spectrum. Owing to the $\bar{4}3\text{m}$ symmetry, the non-centrosymmetric semi-conductor ZnSe could possess second order nonlinearity. Although it is an isotropic material, the phase mismatch of the SHG could still be compensated through the random quasi phase matching (RQPM) [147] effect. This effect originates from the randomly oriented crystallites in the polycrystalline ZnSe we used. The RQPM effect also contributes to the efficient generation of the 4th and 5th order harmonics as revealed in the measured spectrum through the cascaded three-wave mixing. In order to verify the effect of the RQPM, we also measured the 2nd to 7th order harmonics in a 0.6mm polycrystalline ZnSe crystal with $6\mu\text{J}$ input energy, and a 0.5mm monocrystalline GaAs with the same zinc-blende structure for fair comparison. The clear image of the harmonic peaks is observed from the ZnSe as shown in Fig.4.7(c). On the contrary, in the GaAs, only up to 5th order harmonics peaks are measured with much lower intensity. By considering the GaAs has the higher second and third order

Fig.4.7. Spectra of the SCG in ZnSe. (a) Measured and simulated spectra at $>30\text{db}$ level. (b) Full spectrum of the SCG. (c) 1st to 7th order harmonics generated in the 0.6mm ZnSe.

nonlinearity [97], we can conclude that the RQPM could significantly enhance the harmonic generation and the supercontinuum generation in ZnSe. This effect is not considered in the previous theoretical study of LWIR self-compression in the ZnSe crystal [45].

The temporal profile of the self-compressed pulse is also measured through the IAC apparatus. The measured IAC trace and retrieved temporal profile are presented in Fig.4.8 (a) and (b) respectively, revealing that the pulse is compressed to 69 fs, 2.3-cycle pulse width at 9 μm central wavelength. The measured trace fit well with the retrieved one with 0.15 fitness error. The fitting error mainly comes from the intensity noise and the imperfect beam alignment. The retrieved pulse duration is 1.5 times the transform limited (TL) one calculated using the measured spectrum. This discrepancy is introduced by $\sim 30000\text{fs}^3$ TOD introduced by the germanium dispersion compensator

1 and dispersive optics in the IAC. Despite this, the measurement result still implies the
 2 good balance between the self-phase modulation induced chirp and the anomalous
 3 dispersion.

4
 5 Fig.4.8. Measured and simulated temporal profile of the self-compressed pulses. (a)
 6 Measured and retrieved IAC trace; (b) Retrieved temporal profile; (c) Simulated spatial
 7 temporal profile at the output surface of the ZnSe; (d) Simulated spatial averaged temporal
 8 profile.

9 The SC spectrum and the temporal profile are also confirmed by the simulation. The
 10 simulated SC spectrum agrees with the measured spectrum except that it predicts
 11 weaker SHG and 3rd order harmonic components at $\sim 4.5 \mu\text{m}$ and $3 \mu\text{m}$, respectively.
 12 This is because the nonlinear coefficient of ZnSe used in the simulation, which varies
 13 with the grain size and orientation of the polycrystalline ZnSe, could not be accurately
 14 acquired. Since we consider both RQPM and the cascaded three-wave mixing in the
 15 simulation, strong 4th and 5th order harmonics from ZnSe are also reproduced. The
 16 simulated spatial-temporal profile and the spatial averaged pulse shape of the self-
 17 compressed LWIR pulse are also illustrated in Fig.4.8 (c) and (d). A $7.3 \mu\text{m}$ LPF is
 18 applied for ZnSe to mimic the measurement condition. The simulation shows a 75fs
 19 pulse duration of the main pulse. This value approaches to the measured 69 fs pulse

1 duration. Effects such as the pulse self-compression, self-steepening, and pulse
2 splitting in the anomalous dispersion regime are also revealed in the spatial-temporal
3 profile.

4 **4.4.2 Pulse self-compression in KrS-5**

5 Supercontinuum generation and pulse self-compression are also observed in the 5mm
6 thick KrS-5 crystal. As we can see from Fig.4.9, the LWIR spectrum is broadened when
7 the input intensity is gradually increased. In the $|L| < 4\text{mm}$ regime where single
8 filamentation is formed, significant supercontinuum and pulse compression is observed.
9 The multi-filamentation is only observed at around $L=0\text{mm}$. At this position, the
10 spectrum shrink is also found. These experiment results show similar behavior
11 compared with the ZnSe case. However, the spectrum in KrS-5 extends more to the
12 longer wavelength compared with the one in ZnSe. This is due to a flatter dispersion
13 curve and lower dispersion value of the KrS-5 crystal within the $4\text{-}14\mu\text{m}$ region. In this
14 case, efficient energy conversion between the $9\mu\text{m}$ pump and the longer wavelength
15 components could be reached due to the small temporal walk-off. This effect is also in
16 accordance with the measurement of the MWIR SCG and pulse self-compression [122]
17 in the solid material. Due to a broader supercontinuum is obtained, shorter pulse
18 duration of the self-compressed pulse is also obtained. The minimum pulse duration
19 equals to 1.5 optical cycles.

20 At the $L=2\text{mm}$ position, we get the maximum bandwidth of the SCG and the smallest
21 compressed pulse duration. In this situation, a single and stable filamentation with
22 $\sim 5\text{mm}$ length is generated. The existence of the single filamentation could be verified
23 through the photo of filamentation and the Gaussian beam profile with no secondary
24 beam observed, as shown in the Fig.4.10.

1

2 Fig.4.9. Self-compression experiment in the KrS-5 which include, (a) the SC spectra
 3 variation with different surface-focal point distance, (b) the output energy variation with
 4 different surface-focal point distance, and the IAC traces of the self-compressed pulses at (c)
 5 L=6mm, (d) L=5mm, (e) L=4mm, (f) L=3mm, (g) L=2mm.

6

7 Fig.4.10. Images of (a) the filamentation in the KrS-5 and (b) the output beam profile.

Fig.4.11. Measured and simulated (a) spectra, and temporal profiles which include, (b) Measured and retrieved IAC trace; (c) Retrieved temporal profile; (d) Simulated spatial temporal profile at the output surface of the KrS-5; (e) Simulated spatial averaged temporal profile.

The spectrum of the SC generated in KrS-5 is illustrated in Fig.4.11(a). The measured SC spectrum spans from 2 μm to beyond 14 μm at -30 dB intensity level. There is no SHG peak measured in KrS-5, due to its centrosymmetric cubic structure and the vanished 2nd-order nonlinearity. Compared with the SC spectrum generated by 3.1 μm laser in the normal dispersion region of the KrS-5 [136], this spectrum extends more towards the higher frequency with 4.5 times expansion in frequency. The temporal profile of a 45fs, 1.5 optical cycle pulse is also measured. The retrieved IAC trace fit

1 the measurement well at the main peaks in the centre with 0.16 fitness error. The
2 retrieved temporal profile shows a self-steepened pulse shape with several side lobes,
3 which could be explained by the pulse splitting effect [45].

4 The rationality of the measurement results is also verified by the numerical calculation.
5 As shown in Fig.4.11(a), the calculated spectrum spans from the wavelength $<2\mu\text{m}$
6 to $>14\mu\text{m}$ which fits the measured spectrum well. The 3rd order harmonic peak could
7 be observed in both experiment and simulation result. The discrepancy of the cut-off
8 wavelength in the NIR region is due to the inaccurate settings of the input parameters,
9 for instance the beam radius curvature. The asymmetric spectral shape is also
10 confirmed by the simulation. This could be explained by the self-steepening effect
11 assisted by the plasma which attenuates and sharpens the trailing edge of the pulse. The
12 self-compression effect also reinforced this phenomenon by compressing the pulse and
13 increasing the rate of intensity variation. As a result, the phase shift introduced by the
14 SPM becomes more significant at the back of the pulse. More higher frequency
15 components are thus generated. In the simulation, the pulse width of the self-
16 compressed pulse is also calculated as 41fs which approaches the measured 45fs. A
17 more significant pulse splitting phenomenon is also observed in the simulation
18 compared with the measurement result. This is because the simulation parameters, such
19 as the plasma recombination time and electron collision time, may not be accurate
20 acquire.

21 **4.4.3 Pulse self-compression in GaAs**

22 According to the Table 4.1, the critical power of GaAs is about 10 times smaller than
23 the values of the ZnSe and the KrS-5. As a result, the peak power of the input pulses
24 could reach ~ 80 times the critical power of GaAs, which means the multi-filamentation
25 phenomenon could happen. Even when we use a parabolic mirror with 3 times longer
26 focal length, secondary beams could still be easily observed at the back surface of the

1 GaAs. In the multi-filamentation region, we measure the variation of the SC spectrum
 2 with the distance L as shown in Fig.4.12(a).

3
 4 Fig.4.12. Self-compression experiment in the GaAs which include, (a) the SC spectra
 5 variation with different surface-focal point distance, (b) the output energy variation with
 6 different surface-focal point distance. The output beam profiles measured at $L = -1.5$ mm,
 7 1.5 mm and 7.5 mm are also included in the insets.

8 In Fig.4.12, the SC spectrum in GaAs shows a different character compared with the
 9 one measured in ZnSe or KrS-5. When the front surface of the crystal is located before
 10 the focal plane, the multi-filamentation could be observed even at a far position
 11 $L = 12.5$ mm according to the occurrence of the secondary beam. In the multi-
 12 filamentation region, the spectrum shows both less red and blue shift. Strong plasma
 13 absorption ($\sim 40\%$) of the fundamental wave at $9\mu\text{m}$ is also measured as shown in the
 14 output energy curve (Fig.4.12(b)). Only when the crystal is moved to position $L > -$
 15 1.5 mm, the secondary beams vanishes. The SCG could still be generated through SPM
 16 without the help of filamentation.

17 At the $L = -1.5$ mm, the photo of the output beam profile is taken as shown in the inset
 18 of Fig.4.13 (a). The secondary beams could be barely observed which proves the
 19 disappearance of the multi-filamentation effect.

1

2 Fig.4.13. Measured and simulated (a) spectra, and temporal profiles which include, (b)
 3 Measured and retrieved IAC trace; (c) Retrieved temporal profile; (d) Simulated spatial
 4 temporal profile at the output surface of the GaAs; (e) Simulated spatial averaged temporal
 5 profile. The photo of the output beam profile is shown in the inset of figure (a).

6 The spectrum of the SC from the GaAs is illustrated in the Fig.4.13(a). The spectrum
 7 of the LWIR pulse could be broadened to the 3-14 μ m wavelength range. The measured
 8 spectrum shows a similar profile as the spectrum of the MWIR driven SCG in 5mm
 9 GaAs [71]. Compared with the SC of the KrS-5 which has the similar ZDW, the SC
 10 of the GaAs extends more to the shorter wavelength especially in the 3-6 μ m range.
 11 This could be attributed to the higher nonlinearity of the GaAs and stronger self-phase

modulation effect. Due to the self-compression effect, the pulse duration of the LWIR pulse is reduced to 68fs, 2.3 optical cycles in 5mm GaAs, according to the measured and retrieved IAC trace (Fig.4.13(b) and (c)). The retrieve temporal profile of the self-compressed pulse shows a strong pulse splitting effect.

The simulated SC spectrum could well repeat the measurement result, as shown by the red line in Fig.4.13(a). The wavelength cutoff at 3 μ m is also predicted by the numerical calculation. For the spatial-temporal profile of the self-compressed pulse, strong lobe peaks could also be observed as shown in Fig.4.13(d) which verifies the measured pulse splitting. The main peak of the spatial averaged temporal profile has a 70fs duration at the FWHM which is in accordance with the measured value.

4.4.4 Further discussions

According to the simulation, the plasma densities in ZnSe and KrS-5 could reach $9.42 \times 10^{18} \text{ cm}^{-3}$ and $3.5 \times 10^{18} \text{ cm}^{-3}$ respectively, as shown in Fig.4.14. This value at LWIR is smaller but still be at the similar level as the filament generated by the MWIR laser [45]. Also, this value is still much more significant than the plasma density (10^{14} - 10^{16} cm^{-3}) generated in gas [117]. Therefore, the plasma effects are still significant in both ZnSe and KrS-5 crystals.

Fig.4.14. Simulated plasma density at different positions of (a) KrS-5 and (b) ZnSe.

1 The plasma effect together with the self-steepening effect contribute to the generation
 2 of high frequencies as shown in the measured asymmetrical spectra in Fig.4.7(a) and
 3 Fig.4.11(a). The plasma effect also contributes to the strong reshaping of the spatio-
 4 temporal profile in ZnSe and KrS-5. The simulated spatio-temporal intensity profiles
 5 at four different propagation distances are shown in Fig.4.15 (a-d) and Fig.4.16 (a-d)
 6 for KrS-5 and ZnSe, respectively. As shown in both figures, the trailing part of the self-
 7 compressed pulse is attenuated during propagation. The pulse shape at zero radius also
 8 becomes steep. These effects are introduced by various nonlinear effects including the
 9 self-steepening and plasma effects. The self-steepening effect makes the trailing part
 10 of the pulse steep and the plasma absorption further enhances the pulse steepening.
 11 During pulse propagation, the plasma density is higher at central axis where the radius
 12 equals to zero. The plasma attenuates the trailing edge of the pulse mainly at central
 13 axis rather than the off-axis position. That explains the formation of the U-shape
 14 spatial-temporal profile in both crystals.

15
 16 Fig.4.15. Spatio-temporal intensity profile are shown for KrS-5 at (a) $z = 0$ cm, (b) $z = 0.2$
 17 cm, (c) $z = 0.3$ cm, and (d) $z = 0.5$ cm. The color bars show intensity in W/cm^2 .

Fig.4.16. Spatio-temporal intensity profile are shown for ZnSe at (a) $z = 0$ cm, (b) $z = 0.3$ cm, (c) $z = 0.5$ cm, and (d) $z = 0.9$ cm. The color bars show intensity in W/cm^2 . The simulated spatio-temporal intensity profiles look different with the ones in Fig.4.7(c) in the manuscript since the $7.3 \mu\text{m}$ long-pass filter is not added in this simulation.

However, in the GaAs crystal, there is no filamentation generated in the pulse self-compression process. Without the plasma effect, abrupt intensity spikes could be observed in both measured temporal shape (Fig.4.13(c)) and the calculated spatial temporal profile (Fig.4.13(d)). This spatial-temporal profile shows the importance of plasma in arresting the beam collapse. Without the plasma effect, the self-steepening effect could be also observed. However, the pulse abruptly splits into several portions with lower peak power which prevent the beam collapse. In the KrS-5 crystal, the peak power of the self-compressed pulse is maintained by the plasma absorption so that the beam collapse can still be observed. Due to the beam collapse is arrested, shorter self-compressed pulse with clear temporal shape could be generated in KrS-5, although its nonlinear refractive index is much smaller than that of GaAs.

In a conclusion, the experiment and simulation results show similar relation between self-compression and plasma effect as previous theoretical works [45,133]. In the self-compression process, four plasma effects should be taken into consideration [133]. First, in the LWIR filamentation in solids, the plasma defocusing effect compensates

1 the pulse self-focusing effect. Therefore, it prevents the pulse to complete collapse and
2 thus help maintain a stable filamentation, which enhance the effective length of the
3 SPM. Second, the buildup of the plasma density also changes the refractive index. The
4 variance of the plasma density in time domain leads to the phase modulation of the
5 pulse. Therefore, similar as SPM effect, the time-dependent plasma density also
6 contributes to the generation of new frequencies. Third, when a pulse passes through
7 the solid material, its leading edge excites high density plasma. When the trailing edge
8 of the pulse goes through the plasma region, the plasma defocusing effect will
9 significantly reduce its intensity and thus lead to an asymmetry pulse shape with a sharp
10 rear edge. According to the principle of the SPM, more high frequency components
11 will be generated due to the sharp rear edge of the pulse. Four, it is proved that the
12 plasma density changes the refractive index and the ZDW could be shifted to a shorter
13 wavelength when the plasma density is high. Therefore, more frequency components
14 will be located in the anomalous dispersion region and the self-compression regime of
15 the pulse will be expanded.

16 Compared with the previous MWIR and LWIR pulse self-compression experiment, the
17 experiment result also shows the advantage of having filamentation effect. As shown
18 in Table 4.3, compared with the self-compression in gas [42,120], although the output
19 pulse energy is much lower, the cycle number of the compressed pulse in this work is
20 ~62.5% of the cited works. This is because the plasma density in solid material is much
21 stronger than its value in gas so that the self-compression is enhanced. Similar as this
22 work, the MWIR pulse driven self-compression in CaF₂ [122] shows a sub-two-cycle
23 compressed pulse which is also shorter than the gas-based self-compression. This result
24 also shows that efficient pulse self-compression with sub-two-cycle pulse duration
25 could be achieved in both MWIR and LWIR experiments, since the plasma density at
26 LWIR wavelength is smaller but still at the same level of the density value at MWIR
27 wavelength. Compared with the solid-based LWIR self-compression without
28 filamentation [131], this work shows an even more shorter pulse duration and better

1 pulse compression. In short, these comparison between this work and previous research
 2 also shows the advantage of generating filamentation in the solid-based self-
 3 compression experiment.

4 Table 4.3. Comparison between self-compression experiment in gas and solid material driven
 5 by MWIR and LWIR lasers.

	Wavelength	Material	Filamentation or not	Pulse duration (cycle number)	Pulse energy	Reference
1	3.9	N2	Yes	31.5fs (2.4 cycle)	13mJ	[42]
2	2	Xe	Yes	17.9fs (2.7 cycle)	0.27mJ	[120]
3	2.1	CaF2	Yes	13.5fs (1.93 cycle)	9uJ	[122]
4	3.1	YAG	Yes	32fs (2.9 cycle)	3uJ	[123]
5	2.1	BBO	No	22fs (3.1 cycle)	73uJ	[125]
6	10	GaAs	No	2ps (66.7 cycle)	-	[131]
7	9	KrS-5	Yes	45fs (1.5 cycle)	3.5uJ	This work

7 4.5 Conclusions

8 In this chapter, the supercontinuum generation and pulse self-compression are studied
 9 in ZnSe, KrS-5 and GaAs. In all the materials, the spectra of the LWIR driving laser
 10 could be broadened to above 2-octave spanning. The broadest SC spectrum which is
 11 achieved in the 9mm ZnSe crystal spans from 0.47 μ m to 12 μ m and covers over 5
 12 octaves. At the same time, pulse duration of the driving laser is compressed from 5
 13 optical cycles to less than 2 optical cycles. The shortest pulse duration which is
 14 obtained in 5mm KrS-5 crystal equals to 45fs, 1.5-cycle.

1 The experiment results show that the self-compression in anomalous dispersive solid
2 is introduced by the joint action of the material dispersion, nonlinearity and plasma
3 effects. Among all the effects, the plasma effect is experimentally verified as the vital
4 role in spectrum broadening and pulse self-compression. High-density plasma is
5 generated in both ZnSe and KrS-5 crystals. It is shown that the plasma absorption in
6 both crystals enhances the pulse self-steepening effect. Due to the self-steepening
7 effect, more high frequency components are generated and the resultant SC spectrum
8 exhibits an asymmetric profile. Furthermore, by comparing the spatial-temporal
9 profiles generated in KrS-5 and GaAs, we can see that the plasma effect would also
10 help arrest the beam collapse and reject the pulse splitting. As a result, shorter self-
11 compressed pulses with clear pulse shape are generated in KrS-5 crystal. The
12 experiment results reported in this chapter can be considered as a complement to the
13 existing theoretical works on LWIR self-compression in solid materials.

14

15

Chapter 5. 9 μ m OPCPA driven HHG in solid

5.1 Introduction

Motivated by the applications such as the field-resolved spectroscopy [5,148], the petahertz electronics [149], and the strong-field experiments [150,151], the development of high energy, ultrafast, LWIR laser sources has become a research frontier in recent years. The development of LWIR lasers, in turn, trigger deeper exploration in new areas. In this chapter, we use the developed LWIR OPCPA system to generate the high harmonic generation (HHG) in solids.

We successfully demonstrate a high harmonic generation in a polycrystalline ZnSe. The linear dependence of the cutoff energy with respect to the driving wavelength is checked and compared with the previous works on ZnSe-based HHG [23,152]. The properties of the high harmonic spectrum in the non-perturbative region are also analysed.

This chapter is organized as follows. In section 5.2, an introduction about the high harmonic generation and its mechanism is given. Section 5.3 reports our experimental results on HHG, and discusses on the properties of the high harmonic spectrum.

5.2 High harmonic generation

The high harmonic generation is a typical strong-field light-matter interaction process. In this progress, the spectrum of the emitted light contains frequency peaks which are integer multiples of the driving frequency. The HHG is first achieved in the atomic gases [153]. Since then, the HHG process in solid material has attracted huge attentions. In order to observe the HHG in solid, it is required to have driving laser with high field strength and non-resonant frequency. The first HHG experiment was realized in ZnO [16] crystal. As the development of strong mid-infrared sources in recent years, first

1 HHG experiment has been realized in ZnO [8] crystal. This experiment triggers a series
 2 of experimental and theoretical studies on HHG in semiconductors and dielectrics.

Fig.5.1. The mechanism of the solid-based HHG [154].

5 The mechanism of the solid-based HHG is shown in Fig.5.1. In the HHG process, an
 6 electron in the valence band is first excited to the conduction band through tunneling
 7 ionization. Then, the electron could move under the electric field of the driving laser
 8 and acquires kinetic energy. Finally, in one case, the electron could return to the parent
 9 ion. During this recombination, the photons could be released as high harmonics since
 10 this process repeat over many optic cycles. In another case, the mobile electrons form
 11 the intra-band current. The intra-band current source could also contribute to the high
 12 harmonics through Bloch oscillation process [155,156]. At low frequency region, the
 13 intra-band current dominates the generation of odd order harmonics and the inter-band
 14 transition determines the generation of even order harmonics. At high frequency region
 15 ($>10^{\text{th}}$ order harmonics), the generation of harmonic peaks is dominated by the inter-
 16 band transition.

17 Compared with the atomic HHG in gases, the solid-based HHG shows several unique
 18 properties. In the atomic gases, even order harmonics could not be generated due to its

1 inversion symmetric structure. However, the spectrum of solid-based HHG could
2 exhibit both odd and even order harmonic peaks since the spatial inversion symmetric
3 may be broken in solid material, such as the ZnO [8] and GaSe [10] crystals.
4 Additionally, the high harmonic generation in gas and solid material show a different
5 behavior of the cutoff energy with respect to excitation. The cutoff energy is
6 proportional to the maximum kinetic energy an electron can obtain when it is
7 accelerated by the electric field. In atomic HHG, the cutoff energy quadratically scales
8 with the wavelength and field strength of the driving laser. On the contrary, in the HHG
9 in solid materials, a linear dependence is found in both experiment [8,10,157] and
10 theory [158,159].

11 Nowadays, the HHG experiment have been demonstrated in multiple solid materials
12 such as the Si [157,160], Sapphire [161], MgO [162], ZnO [8,163], MoS2 [164], GaSe
13 [10], ZnSe [23,165], graphene [166] and solid Ar and Kr [167], driven by different
14 wavelengths varying from NIR to LWIR. The HHGs in solids nowadays could be used
15 in probing the crystal band structure [163] and imaging the charge dynamics in solids
16 [168]. The current experimental and theoretical studies of the HHG in solid materials
17 have been summarized in Ref. [154] and [169], respectively.

18 **5.3 HHG in the ZnSe**

19 **5.3.1 Experiment conditions**

20 In this experiment, we studied the HHG in ZnSe driven by the 9 μ m LWIR OPCPA
21 system. The 9 μ m LWIR pulses with 14 μ J pulse energy and ~ 0.35 TW/cm² peak
22 intensity are focused into the ZnSe crystal through an f=38.1mm parabolic mirror. The
23 thickness of the ZnSe crystal varies from 0.4mm, 2mm to 9mm. The emitted HHG is
24 collected by an f=40mm CaF₂ lens and then focused for further measurement. Here,
25 we use the NIRQuest spectrometer for measuring the 0.9-2.5 μ m wavelengths and two
26 customized HR2000+ spectrometers for the measurement of the 0.75-0.9 μ m region

1 and 0.25-0.75 μm region. In order to enhance the coupling efficiency, the HHG
2 emission is coupled to a spectrometer in free space.

3 **5.3.2 HHG spectrum in ZnSe**

4 The measured full spectra of the HHGs in 0.4mm, 2mm and 9mm ZnSe crystals are
5 shown in the Fig.5.2. In Fig.5.2(a), clear picture of the HHG spectrum with up to 24th
6 order harmonic peak is measured after a 0.4mm ZnSe. In the measured spectrum, both
7 even and odd high harmonic peaks could be observed. This result is in accordance with
8 the previous measured spectra of HHG in ZnSe [23,152]. In this HHG spectrum, a
9 plateau which spans from the 13th to 17th order harmonics is also observed. The plateau
10 is followed by a strong fluorescence peak located at around 470nm which is introduced
11 by the electron-hole recombination. The intensity of the fluorescence peak is shown
12 several orders higher than the harmonic peaks around it. Due to the fluorescence, the
13 spectra of the 18th and 21st order harmonic peaks are distorted. The broadband
14 fluorescence also covers the similar spectral range with the 19th and 20th harmonic
15 peaks so that these two peaks are indistinguishable in the spectrum. The cutoff of the
16 HHG and the spectra of the 21st to 24th order harmonics are shown in the inset of
17 Fig.5.2(a). The 21st to 24th order harmonics are all beyond the band gap (2.7eV, 470nm)
18 of ZnSe. Following the 24th order harmonics, the HHG spectrum is cutoff at 374nm,
19 corresponding to 3.31eV photon energy. For the spectra measured in the 2mm and 9mm
20 ZnSe crystals (Fig.5.1(b)), distinct peaks are replaced by the supercontinuum due to
21 the more dominant self-phase modulation effect over the HHG effect. Both spectra are
22 cut off at the 470nm which corresponds to the band gap energy of ZnSe crystal. The
23 harmonic peaks beyond the band gap are too weak to be observed in both materials.

Fig.5.2. Full HHG spectra measured in (a) 0.4mm ZnSe, (b) 2mm (blue line) and 9mm (red line) ZnSe crystals. The cutoff of the HHG in 0.4mm ZnSe is detailed displayed in the inset of (a).

The repeatability and the reliability of the measured spectrum in 0.4mm ZnSe have been examined with the help of mathematical statistics. Under the same experimental condition, we repeat the measurement of the HHG spectrum for 5 times. The mean value and the standard variation of the wavelength of each high harmonic peak are calculated, as shown in the Table 5.1. Since the 19th and 20th harmonic peaks are covered by the strong fluorescence, the statistics could not be calculated for these two

1 harmonics. According to Table 5.1, the mean wavelength value of each harmonic peak
2 approaches the expect value with measurement error less than 12nm. The reason why
3 this measurement error is high at the 21st harmonic peak may be the spectral distortion
4 introduced by the strong fluorescence. The ratio between the Std. and the mean value
5 of all harmonic peaks are equal or less than 1% rms, which proves the measurement is
6 repeatable and reliable.

7 Table 5.1. Mean value and standard deviation (Std) of the wavelengths of the Nth order
8 harmonic peaks.

	4 (2250nm)	5 (1800nm)	6 (1500nm)	7 (1286nm)	8 (1125nm)	9 (1000nm)	10 (900nm)	11 (818nm)
Mean (nm)	2263.8	1810.9	1501.8	1286.1	1124.7	994.1	895.1	810
Std (nm)	4.91	8.84	9.58	9.79	10.56	7.54	9.27	3.91
Ratio	0.21%	0.49%	0.64%	0.76%	0.94%	0.76%	1.03%	0.48%
	12 (750nm)	13 (692nm)	14 (643nm)	15 (600nm)	16 (562nm)	17 (529nm)	18 (500nm)	19 (473nm)
Mean (nm)	749.4	699.2	652.8	606.8	566.4	528.9	504.5	-
Std (nm)	3.92	4.08	6.64	3.86	2.55	2.85	4.26	-
Ratio	0.52%	0.58%	1.01%	0.64%	0.45%	0.53%	0.84%	-
	20 (450nm)	21 (428nm)	22 (409nm)	23 (391nm)	24 (375nm)			
Mean (nm)	-	440.4	417	397.6	374.1			
Std (nm)	-	2.49	0.59	1.15	1.41			
Ratio	-	0.57%	0.14%	0.29%	0.37%			

9

1   5.3.3 Discussions

2   In the measured high harmonic spectrum, we observe the generation of both odd and
3   even order harmonics, a plateau and the harmonics that are beyond the band gap.
4   According to Ref.[169], the intra-band current source dominates the generation of odd
5   harmonics in the low frequency region ($<10^{\text{th}}$ order harmonics). The inter-band
6   polarization determines the generation of even order harmonics and the harmonics
7   beyond the band gap. Therefore, the HHG in ZnSe is generated through the collective
8   effect of the intra-band current and inter-band polarization.

9   In this HHG spectrum, the existence of plateau is a hallmark of a non-perturbative light
10   matter interaction [169]. The non-perturbative behaviour could also be confirmed by
11   verifying whether the intensity of the N^{th} harmonic peak equals to I^N . Here, I is the
12   intensity of driving laser. For the perturbative interaction, the intensity of the N^{th} order
13   harmonics should be proportional to the I^N [8,152,169] according to the traditional
14   coupled wave equation. However, this relation should no longer be fulfilled in the non-
15   perturbative region. In this experiment, we measured the intensities of the 4 to 7th order
16   HH peaks with respect to different pump intensity I . For the 4th and 5th order harmonics,
17   the intensity I varies from 0.05TW/cm² to 0.35TW/cm². For the 6th and 7th order
18   harmonics, the variations of the intensity I start from 0.1 TW/cm² and 0.2TW/cm²,
19   respectively. For lower pump intensity, the HH peaks are below the noise level. We
20   compare the measured HH intensities with the calculated one based on the I^N relation.
21   The comparisons are shown in Fig.5.3 (a)-(d).

Fig.5.3. Intensity of the N th order harmonic peaks as a function of pump intensity for the (a) $N=4$, (b) $N=5$, (c) $N=6$, (d) $N=7$; (e) $N=10$ to 24. The dashed lines in (a) – (d) show the I^N dependence of the harmonic peak intensity. The dashed line in (e) show the linear dependence of the harmonic peak intensity with respect to I .

According to the Fig.5.3, the I^N -scaling is basically fulfilled when $I < 0.1 \sim 0.2 \text{ TW/cm}^2$. However, the I^N relation is observed to be deviated at $I > 0.1 \sim 0.2 \text{ TW/cm}^2$, which

1 indicates the occurrence of the non-perturbative light-matter interactions. Similar
 2 deviation of the I^N -scaling relation could also be observed in higher order harmonics,
 3 as shown in Fig.5.3(e). In Fig.5.3(e), we can also observe that the dependencies of the
 4 harmonic intensities with respect to pump intensity converge to a linear dependence
 5 for all harmonics when $I > 0.35\text{TW}/\text{cm}^2$. This regular pattern is also observed in the
 6 HHG experiment in ZnO crystal [8].

7 In this experiment, we also compare the measured cutoff energy with the previous
 8 reported one [23,152]. The HHGs in ZnSe reported in Ref. [23] and [152] are driven
 9 by $4.6\mu\text{m}$ and $6.7\mu\text{m}$ MWIR laser sources, respectively. The $4.6\mu\text{m}$ and $6.7\mu\text{m}$ lasers
 10 driven HHG could generate harmonic peaks up to the 8th and 15th order, with cutoff
 11 energy equal to 2.1eV (575nm) and 2.7eV (459nm), respectively. Compared with these
 12 works, we obtain a higher cutoff energy at 3.31eV (374nm) which is 1.23 times higher
 13 than Ref. [152]. We also find that the linear dependence of the cutoff energy with
 14 respect to the driving wavelength predicted by theoretical works [152,165] is still
 15 fulfilled at $9\mu\text{m}$, as shown in Fig.5.4. The measured cutoff energy (3.31eV) at $9\mu\text{m}$ is
 16 shown closed to the expected 3.35eV photon energy, which is calculated based on the
 17 previously mentioned works and the linear dependent law. This result shows the
 18 advantage of a LWIR laser in scaling-up the cutoff energy of the HHG process.

19

20 Fig.5.4. Linear dependence of the cutoff energy with respect to the wavelength of driving
 21 laser.

Fig.5.5. Comparison between the HHGs in the 0.4mm ZnSe and (a) in the 0.5mm GaAs and (b) in the 0.5mm GaSe.

We also find that the high harmonic spectrum could also be affected by the random quasi-phase matching (RQPM) which is a basic property of the polycrystalline ZnSe material [133]. In the HHG, harmonic peaks could also be generated through cascaded three wave mixing process. For example, a 4th order harmonics 4ω could be generated through two cascaded processes, $3\omega = \omega + 2\omega$ and $4\omega = \omega + 3\omega$. This process could be assisted by the RQPM to reduce its phase mismatch. In order to reveal the RQPM effect, we compare the spectrum generated in 0.4mm ZnSe with other HHG spectra generated in a 0.5mm GaAs and a 0.5mm GaSe. Here, GaAs was also adopted in several HHG experiments [23,170]. The GaAs crystal we used is a single crystal with the same zinc-blende structure as ZnSe and a 1.43 eV band gap energy. As shown

1 in Fig.5.5 (a), the HH spectrum in GaAs has only up to the 5th order harmonic peaks
2 with others below the noise level. We can also observe that the 4th and 5th order
3 harmonics measured in the GaAs are both ~ 1 to 2 orders weaker than the corresponding
4 peaks of the ZnSe case. The GaSe crystal is another commonly used solid material in
5 HHG experiment [10]. The HHG spectrum of GaSe, as shown in Figure 5.5 (b) only
6 extends to the 4th order harmonics with similar intensity compared to the corresponding
7 peak of the GaAs case. This comparison shows the role of the RQPM in enhancing the
8 HHG conversion efficiency.

9 **5.4 Conclusions**

10 In this chapter, the HHG up to 24th order harmonics is generated in the polycrystalline
11 ZnSe material driven by the 9 μ m LWIR OPCPA. The HHG spectrum includes an
12 observable plateau at 13th to 18th harmonics, followed by a strong fluorescence peak,
13 several harmonic peaks beyond the band gap and a cutoff at 3.31eV (374nm). The
14 measurement result is proved to be either repeatable or trustworthy.

15 The HHG is shown to be a non-perturbative light matter interaction. The traditional I^N
16 dependence of the harmonic intensity with respect to the driving intensity is replaced
17 by a linear dependence due to the non-perturbative process. The measured cutoff
18 energy of HHG in ZnSe is 1.23 times higher than the previous record [152]. The
19 experiment results also show that the cutoff energy follows a linear dependence with
20 respect to the driving wavelength. This result shows the advantage of a high energy
21 LWIR laser in enhancing the cutoff energy of the high harmonic spectrum. The random
22 quasi-phase matching effect is also shown to be effective in enhancing the efficiency
23 of HHG.

24 In a conclusion, the experiment indicates that the reported 9 μ m LWIR OPCPA system
25 could be a promising tool in the strong-field applications and research.

1 **Chapter 6. Conclusion and Recommendation**

2 **6.1 Conclusions**

3 In this dissertation, we conduct a systematical study of the basic theory, design,
4 implementation and application of the LWIR OPCPA system. The proposed OPCPA
5 system is proved to be a reliable and advanced laser source. It is potentially useful for
6 various applications such as the strong-field light matter interaction and ultrafast mid-
7 IR spectroscopy. The main contributions of this work are summarized as below:

8 First, the key challenge of developing the LWIR OPCPA is shown to be the limited
9 gain bandwidth. The non-collinear method is proposed to overcome this limitation. In
10 order to design a non-collinear OPCPA system, the general mathematical model of the
11 gain bandwidth of the non-collinear OPA/OPCPA is modified. By proposing the
12 minimal wave-vector mismatch condition instead of the previous constrains on the
13 wave vector of the idler beam, the new model can evaluate the upper-limit of the gain
14 bandwidth within 15% calculation error in the LWIR regime. Based on the gain
15 bandwidth model, the 'Nth order flatness' design rule is introduced to guide the design
16 of a non-collinear OPCPA. Following this rule, we proposed an AD-OPA system by
17 using a designed chirped grating. This new method is shown to achieve higher order
18 flatness and provide larger gain bandwidth than the traditionally used NOPA system.

19 Second, a micro-joule level, few-cycle LWIR OPCPA is designed and implemented.
20 Since the angular-dispersive OPA method is hard to applied in experiment, the
21 limitation of the gain bandwidth is solved by using a large band gap LGS crystal in the
22 OPCPA system. The OPCPA system could produce 9 μ m LWIR pulses with 14 μ J pulse
23 energy, 140mW average power and 145fs pulse duration. The output pulses are
24 compressed with a meticulous dispersion management for the 2nd and 3rd order
25 dispersions, and the optical parametric phase, which is significant in the LWIR regime.

1 The OPCPA system is proved to be long-term stable. It is also the first, to the best of
2 our knowledge, OPCPA system operating at the LWIR regime.

3 Third, in order to compress the LWIR output of OPCPA to single cycle regime, the
4 SCG and pulse self-compression in the anomalous dispersive solids are systematically
5 studied. The experiment is conducted in the KrS-5, ZnSe and GaAs crystals. The 1.5-
6 cycle self-compressed pulse with micro-joule pulse energy is demonstrated in KrS-5
7 crystal and a supercontinuum spectrum with over 5-octave spectral spanning is
8 generated in the ZnSe crystal. The measured LWIR temporal and spectral profiles fit
9 well with the simulation results. Moreover, the experiment results show the crucial
10 roles of the filamentation, GDD and the nonlinear refractive index in the pulse self-
11 compression, which provides the solid evidences for the theoretical prediction.

12 Finally, the LWIR OPCPA is applied in generating high harmonics in the solid material.
13 The HHG spectrum with the up-to 24th order harmonic peak is observed. The
14 measurement result is verified to be reliable and repeatable. The experiment result
15 shows that the LWIR laser driven HHG could generate 0.61eV higher cutoff energy
16 compared with the MWIR laser driven HHG. The linear dependence of the cutoff
17 energy with respect to the driving wavelength is also proved by the experiment. In a
18 conclusion, it proves that the high energy, high average power, few-cycle LWIR
19 OPCPA system is a useful tool in strong field applications.

20 **6.2 Recommendations for future research**

21 In this section, brief outlooks are conducted for the potential improvements of both the
22 basic OPCPA theory and the LWIR OPCPA implementation. For the fundamental
23 theories, the recommendations aim to lead the way for in-depth research on the OPCPA
24 performance analysis. For the implementation of the OPCPA, possible solutions for
25 further enhancing its performances are introduced.

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1 domain from each segment separated by the window, which just shares the similar idea
2 with the temporal mapping. Based on the Gabor transform or the wavelet transform,
3 energy conversion could not only be analyzed in frequency domain but also modeled
4 and studied in each time intervals.

5 **6.2.2 Flat-top LWIR OPCPA**

6 As shown in chapter 3, the current efficiency of the LWIR OPCPA is still limited to
7 less than 1%. One of the most important reasons is that the spatial and temporal profiles
8 of the pump beam are all Gaussian-like. At the wings of these profiles, the pump
9 intensity as well as the small signal gain is low. Therefore, signal components located
10 at these places could not be sufficiently amplified.

11 One promising solution is to use the pump beam with flat-top shaped spatial and
12 temporal profiles. By conducting a top hat shaping for the pump beam profile, the
13 parametric gain could be enhanced due to a uniform gain distribution in space. Also,
14 under the damage threshold limitation, more energy could be stored in single pump
15 pulse which is also favored for higher gain. The gain bandwidth could also be
16 broadened when using a flat-top pump pulse since more frequency components located
17 at the wings of the chirped signal pulse could also be efficiently amplified.

18 Nowadays, the flat-top shaping has been investigated in both spatial [171,172] and
19 temporal domain [173,174]. Moreover, specific phase plate for top-hap beam shaping
20 could now applied with high efficiency. Therefore, using the flat-top method to
21 improve the performance of the LWIR OPCPA will be realized soon.

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Author's Publications

A. Journal Paper

- [1] **Shizhen Qu**, Houkun Liang, Kun Liu, Xiao Zou, Wenkai Li, Qi Jie Wang, Ying Zhang, "9 μm few-cycle optical parametric chirped-pulse amplifier based on LiGaS₂", Optics Letters 44.10 (2019): 2422-2425. <https://doi.org/10.1364/OL.44.002422>
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5 **B. Conference Paper**

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7 Zhang, “High-energy 9 μ m LiGaS₂-based Optical Parametric Chirped-Pulse Amplifier.”
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3

Appendix I. Interferometric autocorrelator

In this appendix, the basic theory and the setup of the home-build interferometric autocorrelator (IAC) are introduced. Due to the lack of high-speed spectrometer in the 3-5 μm regime, traditional SHG Frequency resolved optical gating (FROG) technique could not be applied in the LWIR pulse measurement. Therefore, in this dissertation, the interferometric autocorrelator is used for all the temporal profile measurements of the few-cycle, LWIR pulses.

The schematic of the IAC apparatus is shown in Fig.A1.1. The driving laser is fairly divided into two arms, the R1 and R2, by a 2mm ZnSe beam splitter. In R1, a piezo-stage with feedback control loop and a 300 μm travel range is used for control the time delay. After being reflected by the retro-reflectors in both arms, two parts of the driving laser combine together to form the interferometric signal. This signal is focused into a 0.5mm GaSe crystal by using a $f=150\text{mm}$ parabolic mirror. The interferometric signal excites SHG in the GaSe crystal. Finally, the SHG is detected by the photodetector which has high response in the 1-6 μm spectral range.

Fig.A1.1 Schematic of the interferometric autocorrelator.

1 Here, we let $E_1(t)$ and $E_2(t - \tau)$ represent for two pulses from the arms R1 and R2,
 2 respectively, where τ represents for the time delay between them. The electric field of
 3 the SHG is thus proportional to the square of the electric field of the interferometric
 4 signal, where

$$5 \quad E_{\text{SHG}}(t, \tau) \sim [E_1(t) + E_2(t - \tau)]^2 \quad (\text{A1.1})$$

6 When the SHG signal arrives at the photodetector, its time integrated information is
 7 recorded since the speed of the photodetector is much lower than the carrier frequency
 8 of the SHG signal. As a result, the IAC signal obtained by the photodetector can be
 9 written as,

$$10 \quad I_{\text{IAC}}(\tau) = \int_{-\infty}^{\infty} |[E_1(t) + E_2(t - \tau)]|^2 dt \quad (\text{A1.2})$$

11 The equation could be expanded as,

$$12 \quad \begin{aligned} I_{\text{IAC}}(\tau) = & \int_{-\infty}^{\infty} [I_1^2(t) + I_2^2(t - \tau)] dt \\ & + 4 \int_{-\infty}^{\infty} I_1(t) I_2(t - \tau) dt \\ & + 4 \int_{-\infty}^{\infty} [I_1(t) + I_2(t - \tau)] \text{Re}[E_1(t) E_2^*(t - \tau)] dt \\ & + 2 \int_{-\infty}^{\infty} \text{Re}[E_1^2(t) E_2^{*2}(t - \tau)] dt \end{aligned} \quad (\text{A1.3})$$

13 Here the first term represents for the constant background, the second term is the
 14 intensity autocorrelation, the third term is the modified interferogram of the electric

1 field and the fourth term is the interferogram of the SHG electric field. Since the pulses
 2 $E_1(t)$ and $E_2(t - \tau)$ are equally split from one signal source, as a result, we have

$$\begin{aligned} 3 \quad E_1(t) &= E(t) \\ 4 \quad E_2(t - \tau) &= E(t - \tau) \end{aligned} \quad (A1.4)$$

5 As a result, the SHG could be re-write as,

$$\begin{aligned} 6 \quad I_{IAC}(\tau) &= \int_{-\infty}^{\infty} [I^2(t) + I^2(t - \tau)] dt \\ 7 \quad &+ 4 \int_{-\infty}^{\infty} I(t)I(t - \tau) dt \\ 8 \quad &+ 4 \int_{-\infty}^{\infty} [I(t) + I(t - \tau)] \text{Re}[E(t)E^*(t - \tau)] dt \\ 9 \quad &+ 2 \int_{-\infty}^{\infty} \text{Re}[E^2(t)E^{*2}(t - \tau)] dt \end{aligned} \quad (A1.5)$$

10 and the intensity ratio of these four terms is 1:2:4:1 which shows a 1:8 ratio between
 11 the background and the maximum SHG intensity. However, for the situations where
 12 the input beam is unequally split or the imperfect beam alignment, this ratio is not
 13 fulfilled any more. As a result, this 1:8 ratio could be used as the proof of perfect
 14 alignment of the IAC.

15