Femtosecond laser additive manufacturing of micro-3D optical devices and their applications

Li, Shufan

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DEVICES
AND THEIR APPLICATIONS**

LI SHUFAN

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ENGINEERING**

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SCHOOL OF MECHANICAL AND AEROSPACE
ENGINEERING

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

2021

Statement of Originality

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

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

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Prof Su Pei-Chen

Authorship Attribution Statement

This thesis contains material from 5 paper(s) published in the following peer-reviewed journal(s) / from papers accepted at conferences in which I am listed as an author.

Chapter 3 (Sec. 3.1 and 3.2) is published as Li, S., Jiao, J., & Kim, Y. J. (2018). 3D printing of polymeric optical components by two-photon polymerization. *Proceedings of the International Conference on Progress in Additive Manufacturing*, 2018-May, 190–195. <https://doi.org/10.25341/D4R59R>, &

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The contributions of the co-authors are as follows:

- Prof Young-Jin Kim provided the initial project direction and edited the manuscript drafts.
- I and Dr Jiao performed all the laboratory work with the help of Prof Kim at the School of Mechanical and Aerospace engineering at NTU.
- Dr. Mishra provided the chemicals and the material preparation.
- I mainly prepared the manuscript drafts.
- Prof Young-Jin Kim helped to revise the manuscript and provided some suggestion on the data analysis.

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The contributions of the co-authors are as follows:

- Prof Young-Jin Kim provided the initial project direction and edited the manuscript drafts.
- I and Dr Jiao set up the fabrication system.
- I performed the laboratory work with the help of Dr. Sandeep at the School of Mechanical and Aerospace engineering at NTU.
- I also analyzed the data with Dr. Sandeep.
- I mainly prepared the manuscript drafts.
- Dr Jiao, Dr. Sandeep, and Prof Su Pei-Chen helped to revise the manuscript and provided some suggestion on the data analysis.
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- Prof Young-Jin Kim provided the initial idea and edited the manuscript drafts.
- I and Dr Jiao set up the FsLDW system and fabricate the samples.
- Dr Low set up the FsLDS system and fabricate the samples.
- Dr Jiao and Dr Gao Yi set up the observation system.
- Dr Jiao and Dr Sandeep analyzed the experimental data and prepared the manuscript draft.
- I and all authors reviewed the results and the manuscript writing.

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The contributions of the co-authors are as follows:

- Prof Young-Jin Kim and Dr Low provided the initial idea and edited the manuscript drafts.
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- I fabricate the samples and set up the observation system.
- I also analyzed the data with Dr. Sandeep.
- I mainly prepared the manuscript drafts.
- Dr. Sandeep, and Prof Su Pei-Chen helped to revise the manuscript and provided some suggestion on the data analysis.

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Abstract

Femtosecond laser direct writing (FsLDW) technology is a novel additive manufacturing (AM) method that is newly developed in recent years. AM technology addresses the shortcoming of conventional manufacturing methods in fabricating complex geometric micro-optical structures. Commercial optical lenses are fabricated by subtractive manufacturing, such as grinding and etching, and formative manufacturing, such as moulding. However, they are not suitable for manufacturing micro and nano-sized optical elements. Due to AM's flexibility in the free-shape fabrication and the high resolution into the micro-/nano-dimension, it has drawn attention in the optical fields. Optical device miniaturization with complex structures and specific applications is actively sought out. There have been developments in micro-fabrications by stereolithography (SLA), digital light processing (DLP), or chemical vapor deposition (CVD) via electron beam lithography (EBL). Still, these methods encounter one or some drawbacks in terms of time and energy-consuming, material option, and resolution. I seek a fast and precise fabrication method that can produce customized optical elements and devices with miniature thickness and dimension. The femtosecond (fs) laser machining and fabrication technology have shown the potentials in the research field in this context.

This research work aims to design and fabricate the micro-optical components by FsLDW technology on customized substrates for specific applications. The first proposed optical application is the light sheet fluorescent microscopy using FsLDW fabricated micro-axicon which generates the Bessel beam. It is capable of generating light sheets for illumination and imaging system that can potentially be used in the intracorporal environment. The second application achieved in this work is the collimated beam tilt angle measurement using the FsLDW diffractive axicon, which produces the diffractive pattern evolution directly correlated to the tilt angle.

Two types of laser direct writing systems were established and used in this research. The system Femtosecond Laser Direct Writing (FsLDW) was built with a stationary laser

source and motorized sample stage, consisting of fs laser source and precision instruments, beam-guiding mirrors, a nano-positioning stage, and a real-time monitoring camera. The High-Speed Femtosecond Laser Direct Scanning system (FsLDS) with the scanning laser source and stationary sample stage was built with the laser source, beam-guiding mirrors, and Galvano scanner coupled with an f-theta lens. This system is extensively used for thin-film structure fabrication. In this study, two- and three-dimensional structures with photoresists, including SU-8, etc, as well as reduced graphene oxide (rGO), were successfully fabricated. Parametric studies were carried out to optimize the device geometry and surface quality to meet the requirements of the micro-optical elements. The micro-optics were designed to address specific applications and their optical performances were simulated using ZEMAX OpticStudio®. The physical performances of fabricated elements were compared with theoretical and simulation results and demonstrated their practical applications.

In this thesis, specialized 3D and thin-film (2D) optical components were designed and fabricated, and the following applications with the optical components were demonstrated:

a) The light sheet fluorescence microscopy (LSFM) with the fabricated micro-axicon created Bessel beam. The non-diffractive Bessel beam created by the micro-axicon is used for the illumination and detection of micro-objects and live cells. The proposed scheme for LSFM using this micro-axicon improves the flexibility of the system, as it replaces the bulky illumination objective and greatly improves the axial resolution of imaging compared to Gaussian beam-based imaging. It offers the possibility to reconstruct the high-quality 3D image of biological creatures non-invasively.

b) Beam tilt angle measurement using the fabricated rGO thin-film diffractive axicon. The FsLDS fabricated rGO thin film diffractive axicon transforms a Gaussian beam to a Bessel beam, and hence serves as a good substitute to the commercial bulky refractive optics. The Quasi-Bessel beam generated is very sensitive to the tilt angle between the incident beam and its normal axis. As this thin-film axicon can be easily attached to any surface under consideration, it provides the flexibility to precisely detect the tilt angle between the incident beams and object surfaces.

Table of Contents

Abstract	i
Table of Contents	iii
Acknowledgements	v
List of Figures.....	vi
List of Tables	xi
Chapter 1 Introduction.....	1
1.1 Research Background.....	1
1.2 Research Objectives	7
1.3 Contribution and Scope	10
1.4 Report Organization	11
Chapter 2 Literature Review	13
2.1 Preamble.....	13
2.2 Introduction of Additive Manufacturing	13
2.3 Ultra-Fast Laser Assisted Additive Manufacturing Technology	17
2.3.1 Femtosecond Laser	19
2.3.2 Two-Photon Polymerization Mechanism	26
2.3.3 Photoresist Material.....	28
2.4 Bessel Beam's Generation and Characterization	34
2.4.1 Mathematical Explanation of Ideal Bessel Beam.....	34
2.4.2 Characterization of Experimentally Generated Quasi-Bessel Beam..	37
2.5 Light Sheet Fluorescence Microscopy	43
2.5.1 Principle of Light Sheet Fluorescence Microscopy.....	43
2.5.2 Types and Feature Comparison of Light Sheet Fluorescence Microscopy	47
2.6 Diffractive Lens's Transformation on Beam	52
2.6.1 Design of Fresnel Zone Plate and Fresnel Axicon	52
2.6.2 Interferometric Sensing of Tilted Angle	54
Chapter 3 Establishment of Femtosecond Laser Additive Manufacturing System and Fabrication of Micro-Optics	57
3.1 Overview of Femtosecond Laser Additive Manufacturing System	57

1	3.2 Fabrication of SU-8 3D Micro-Structures by Femtosecond Laser Direct	
2	Writing System.....	58
3	3.2.1 Setup of Femtosecond laser direct writing system	58
4	3.2.2 Parametric Condition and Optimization of SU-8 Micro-Structure	
5	Fabrication	66
6	3.3 Fabrication of Reduced Graphene-Oxide Thin Film Structures by High-Speed	
7	Femtosecond Laser Scanning System	72
8	3.3.1 Setup of High-Speed Femtosecond Laser Scanning System.....	73
9	3.3.2 Parametric Condition of Reduced Graphene-Oxide Thin Film	
10	Structures	77
11	Chapter 4 Light Sheet Microscopy with Micro-Axicon Created Bessel Beam	80
12	4.1 Optical Layout of Light Sheet Fluorescence Microscopy.....	80
13	4.2 Transformation of Gaussian Beam to Bessel-Gauss Beam at a Micro-Axicon .	
14		86
15	4.3 Self-Reconstructing Bessel Beam: Simulation of Propagation Characteristics	
16		89
17	4.4 Fluorescence Microscopy with the Self-Reconstructive Bessel-Gauss Beam	
18	from the Micro-Axicon	94
19	Chapter 5 Collimated Beam Tilt Angle Measurement with Diffractive Axicon	101
20	5.1 Optical Layout of Diffractive Axicon Based Beam Tilt Angle Measurement	
21		101
22	5.2 Transformation of Gaussian Beam to Bessel-Gauss Beam at Diffractive	
23	Axicon	108
24	5.3 Beam Tilt Angle Measurement by the Diamond Pattern	113
25	5.3.1 Transformation by Diffractive Axicon Tilted to Different Angles ...	117
26	5.3.2 Transformation by certain tilted diffractive axicon along the beam axis	
27		119
28	Chapter 6 Conclusion and Recommendation for Future Work.....	132
29	6.1 Conclusion.....	132
30	6.2 Recommendation for Future Work	136
31	6.2.1 Development of Femtosecond Laser Direct Writing System	136
32	6.2.2 Development of Light Sheet Fluorescence Microscopy with Micro-	
33	Axicon	137
34	6.2.3 Development of Beam Tilt Angle Measurement with Diffractive	
35	Axicon	138

1	Publication	141
2	Reference	142
3	Appendix	156
4		

List of Figures

Figure 1	Comparison between different manufacturing methods.....	14
Figure 2	Laser-material interaction for different lasers. (a) femtosecond laser induces minimised HAZ (image credit: https://www.imra.com/applications/ablation-and-micromachining); (b-d) Examples of micro-machining with different pulse widths from nanosecond, picosecond, to femtosecond regimes ang their HAZs [84].....	23
Figure 3	Comparison of single- and two-photon absorption and polymerization. (Top) the excited electron states under single- and two-photon absorption; (Bottom) the TPP voxel is much smaller than the 1PP voxel	27
Figure 4	(left) the CW laser excites the entire column of sample and (right) the ultra-fast pulse laser excites only a small spot of sample	28
Figure 5	Schematic of the SU-8 molecule [86]	30
Figure 6	collection of different geometries of SU-8 high aspect-ratio structures produced by lithography [100], [101].....	31
Figure 7	Optical characteristics of SU-8: (a) Absorption spectrum of SU-8, plotted on a logarithmic scale. (b) Fluorescence photon number versus excitation intensity [102].....	32
Figure 8	SU-8 Transmittance after 300°C Hard Bake for different durations (image credit: MicroChem datasheet for SU-8 3000 Permanent Epoxy Negative Photoresist) ...	33
Figure 9	the intensity distribution f Bessel beam along z-axis	36
Figure 10	Comparison of radial field strengths of a Gaussian beam and the Bessel beams of 0 th - 4 th orders.....	37
Figure 11	(a) Experimental arrangement for the approximate 0 th order Bessel beam with a circular slit and a focusing lens; (b) the profiles of generated J_0 beam and the comparable Gaussian beam with same spot size at beam waist.	38
Figure 12	Schematic of an axicon with base angle α transforming the collimated Gaussian beam of size 2ω to a Bessel-Gauss beam. The Bessel-Gauss beam can be considered as formed by wave vectors lying on the cone surface with an apex angle 2θ	40
Figure 13	Comparison of ray trace diagrams between the collimated Gaussian beam and Bessel beam. The beam rays come from left to right. Illuminated objects are shown in green, while blocked ones are in blue.	42
Figure 14	The principle of light sheet microscopy. The illumination and the detection objectives are oriented orthogonally. The sample is placed at the intersection of their focal planes. A single slice of the sample is illuminated with a thin sheet of laser light.	44
Figure 15	Two types of light sheet generation. Static light sheet is directly generated by a cylindrical lens, and scanned light sheet is generated by rapid beam scanning. ...	46
Figure 16	Schematic diagram of optical components assembles for OPFOS [27].....	48
Figure 17	Schematic diagram of mSPIM [124]. (a) Parallel illumination of the sample in SPIM results in shadows and attenuation across the FOV. (c) In mSPIM, these	

1	artifacts are reduced by illuminating the sample from two sides with a thinner	
2	pivoting light sheet. IL: illumination, DL: detection lens, S: sample.....	49
3	Figure 18 Schematic diagram of the HROPFOS setup [125].	50
4	Figure 19 Design of Fresnel lens. (a) Original refractive lens. (b) Division of the Fresnel	
5	zone by thickness t . (c) Fresnel lens. (d, e) Binary FZP. (f, g) Multi-level FZP.	
6	The red lines in (d,f) represent the geometry of the Fresnel lens, and black lines	
7	represent the profile of phase FZP. The different colors in amplitude FZP in (e, g)	
8	represent different material or dense giving different refractive indexes.....	53
9	Figure 20 Schematics of sensing the object M's tilted angle. (a) Basic spatial displacement	
10	scheme "beam deflection" technique. (b) Mach-Zehnder Interferometric technique	
11	in which the object is placed inside the interferometer that is constructed with a	
12	polarizing beam splitter (PBS). (c) Sagnac interferometric method, in which is	
13	constructed with a PBS. The counterpropagating horizontal and vertical	
14	polarization components are displaced in opposite transverse directions.	
15	Information about θ can be obtained via spatial measurements, polarization	
16	measurements, or both.	55
17	Figure 21 Schematic diagram of 3D printing system configuration.....	58
18	Figure 22 Schematic diagram of 3D printing microstructure by the femtosecond laser spot	
19	scanning the photoresist with pre-designed routine.....	60
20	Figure 23 FemtoFiber pro NIR laser and its parameters. (a) laser source appearance; (b)	
21	internal setup; (c) laser pulse characteristics centered at 780 nm, the	
22	autocorrelation pulse width is less than 100 fs; (d) linear emission spectrum.	61
23	Figure 24 E-545 PI-nano® Piezo Controller and Multi-Axis Positioner.	63
24	Figure 25 The LabVIEW control panel	64
25	Figure 26 Monitoring panel in ThorCam during the printing process. The circle in the	
26	center is the printed cylinder pattern.	66
27	Figure 27 Flow chart of the SU-8 micro-structure fabrication	67
28	Figure 28 Schematic figure for the FsLDS system based on a Galvano scanner	74
29	Figure 29 Appearance of Yb-doped Femtosecond laser Satsuma HP	75
30	Figure 30 (left) Sino Galvano scanner; (right) f-theta lens. The two devices are coupled to	
31	direct the laser beam focus over a plane at distance of 160 mm to the f-theta lens	76
32	Figure 31 Femtosecond laser direct scanning (FsLDS) of the ultra-thin rGO FZPs by	
33	photoreduction in three regimes. (a) The three regimes are fabricated by the	
34	femtosecond laser tuned at different repetition rates and pulse energy levels. (b-d)	
35	Transmission microscope images of the patterned ultra-thin rGO FZPs under the	
36	three regimes respectively, LTPR, TPR, and ABL.	78
37	Figure 32 Light sheet fluorescence microscopy scheme with Bessel-Gauss beam created by	
38	the two-photon-polymerized micro-axicon. (a) Configuration of conventional	
39	LSFM consisting of illumination and detection lenses in orthogonal axes showing	
40	the physical restrictions of the scheme. (b-d) LSFM with micro-axicon: (b)	
41	Configuration of the LSFM utilizing Bessel-Gauss beam created by the micro-	
42	axicon from fiber-carried Gaussian input; (c) schematic of the micro-axicon based	
43	Bessel-Gauss beam scanning for optical sectioning of the cell; (d) optical	

1	microscope and SEM images of the fabricated two-photon polymerized micro-	
2	axicon.....	81
3	Figure 33 (top) The schematic representation of the axicon transforming a Gaussian beam	
4	into a Bessel-Gauss beam. (bottom) Illustration of the self-reconstruction property	
5	of the micro-axicon based Bessel-Gauss beam	83
6	Figure 34 Micro-axicon transforms the collimated Gaussian beam into the Bessel-Gauss	
7	beam. (a) Lateral profile images of the micro-axicon based Bessel-Gauss beam at	
8	$z = 230, 280, 330, 380,$ and $430 \mu\text{m}$. (b) Lateral profile of the Bessel-Gauss beam	
9	collected at the peak position, $z = 380 \mu\text{m}$, matching with the 0 th -order Bessel	
10	beam of first kind. (c) Colormap of the Bessel-Gauss beam in y-z plane. (d)	
11	Intensity propagation of the Bessel-Gauss beam along the z axis. (e) Bessel-Gauss	
12	beam's energy concentration and lateral FWHM of the central bright spot along	
13	the beam axis.	87
14	Figure 35 Simulations of the self-reconstruction capability. (a) Gaussian beam	
15	encountering a bead of diameter $20 \mu\text{m}$, with $n = 1.37$, in the beam axis. (b-d)	
16	Bessel-Gauss beam encountering beads of diameters $10, 20,$ and $30 \mu\text{m}$, with $n =$	
17	1.37 . (e, f) Bessel-Gauss beam encountering a bead of diameter $20 \mu\text{m}$, with	
18	refractive indices $n = 1.46$ and 1.76 . (g, h) Bessel-Gauss beam encountering a	
19	bead of diameter $20 \mu\text{m}$, which is deviated $10 \mu\text{m}$ and $20 \mu\text{m}$ from the axis. (i)	
20	Self-reconstruction distance with respect to the bead diameter. (j) Self-	
21	reconstruction distance with respect to bead's refractive index. (k) Self-	
22	reconstruction distance with respect to the bead's deviation distance from the	
23	beam axis.	90
24	Figure 36 Image comparison of different microscopic methods. (a) and (b) conventional	
25	optical microscope images of the beads of different sizes. (c) and (d) show the	
26	microscopic images of the beads using a Bessel-Gauss beam generated	
27	fluorescence filtered with a band pass filter (FB600). (e) and (f) show microscopic	
28	images of the beads using Gaussian beam generated fluorescence filtered with a	
29	band pass filter (FB600). (g) Comparison of the intensity profiles of the Bessel-	
30	Gauss beam and Gaussian beam before encountering the beads. (h) Comparison of	
31	the Bessel-Gauss beam and Gaussian beam after encountering the beads,	
32	illustrating the non-diffractive nature of the Bessel-Gauss beam. Scale bars: 100	
33	μm	95
34	Figure 37 Microscopic images of HeLa cells with different imaging methods. (a)	
35	Conventional optical microscope image of the cells. (b) Microscopic images of	
36	cells with Gaussian beam generated fluorescence. (c) Microscopic images of the	
37	HeLa cells with Bessel-Gauss beam generated fluorescence. (d) The intensity	
38	profile of the Bessel-Gauss illumination along the beam axis, where the shaded	
39	regions bounded by high contrast indicate the presence of HeLa cells. (e) The	
40	Bessel-Gauss beam profiles before and after the cells, indicating the Bessel-Gauss	
41	beam self-reconstruction. Scale bars: $100 \mu\text{m}$	98
42	Figure 38 Geometry of an axicon forming a Bessel beam: (a) Thorlabs axicon and (b)	
43	diffractive axicon DOE [132].	102

1	Figure 39 Optical microscopic images of the diffractive axicon with a ring width of 33.1	
2	μm that can work as the axicon AX251-A	108
3	Figure 40 Axicons' transformation performance. (a) The Thorlabs Axicon AX251-A	
4	transforms the Gaussian beam into smooth Bessel-Gauss beam; (b) the diffractive	
5	axicon transforms the Gaussian beam into Quasi-Bessel beam with some intensity	
6	fluctuation. (c) The lateral profile captured at $z = 50.5$ mm at the dashed line in	
7	(a). (d) The lateral profile captured at $z = 48$ mm at the dashed line in (b).....	109
8	Figure 41 Normalized lateral intensity profiles of Quasi-Bessel beams created by printed	
9	diffractive axicon and Axicon AX251-A, and comparison with the standard 0 th -	
10	order Bessel function of the first kind.	111
11	Figure 42 FWHM propagation of the resulted Bessel beams transformed by (red)	
12	diffractive axicon and (green) Thorlabs axicon AX251-A. Dashed lines represent	
13	the mean of FWHM along the propagation distance.	112
14	Figure 43 Schematic diagram of the setups of axicons transforming the incident beams. (a)	
15	the Thorlabs Axicon transforms the perpendicularly incident beam. (b) the	
16	Thorlabs axicon is tilted for an angle β with respect to beam axis. (c) the printed	
17	diffractive axicon tilted for the same β transforms the incident beam.	115
18	Figure 44 Resulted beam pattern variation regarding the diffractive axicon's tilted angle to	
19	the incident beam. (a) Schematic diagram of tilted diffractive axicon transforming	
20	the incident beam. (b-g) the lateral profile of resulted beam patterns transformed	
21	by diffractive angle at different tilted angles β . (h) X-cross-sectional profiles	
22	captured by CCD camera at $z = 48$ mm, with tilted angle interval of 1° . Each	
23	figure is normalized individually.....	118
24	Figure 45 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 5^\circ$. (a)	
25	Schematic diagram of 5° -tilted diffractive axicon transforming the incident beam.	
26	(b-i) the lateral profile of resulted beam patterns transformed by the tilted	
27	diffractive angle captured at different distance from the lens. (j) X-cross-sectional	
28	profiles captured by CCD camera from $z = 28$ to 77 mm, with interval of 0.5 mm.	
29	Each figure is normalized individually.....	121
30	Figure 46 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 10^\circ$. (a)	
31	Schematic diagram of 10° -tilted diffractive axicon transforming the incident	
32	beam. (b-i) the lateral profile of resulted beam patterns transformed by the tilted	
33	diffractive angle captured at different distance from the lens. (j) X-cross-sectional	
34	profiles captured by CCD camera from $z = 29$ to 76 mm, with interval of 0.5 mm.	
35	Each figure is normalized individually.....	122
36	Figure 47 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 15^\circ$. (a)	
37	Schematic diagram of 15° -tilted diffractive axicon transforming the incident	
38	beam. (b-i) the lateral profile of resulted beam patterns transformed by the tilted	
39	diffractive angle captured at different distance from the lens. (j) X-cross-sectional	
40	profiles captured by CCD camera from $z = 29$ to 76 mm, with interval of 0.5 mm.	
41	Each figure is normalized individually.....	124
42	Figure 48 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 20^\circ$. (a)	
43	Schematic diagram of 20° -tilted diffractive axicon transforming the incident	
44	beam. (b-i) the lateral profile of resulted beam patterns transformed by the tilted	

1	diffraction angle captured at different distance from the lens. (j) X-cross-sectional	
2	profiles captured by CCD camera from $z = 29$ to 76 mm, with interval of 0.5 mm.	
3	Each figure is normalized individually.....	126
4	Figure 49 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 25^\circ$. (a)	
5	Schematic diagram of 25° -tilted diffractive axicon transforming the incident	
6	beam. (b-i) the lateral profile of resulted beam patterns transformed by the tilted	
7	diffraction angle captured at different distance from the lens. (j) X-cross-sectional	
8	profiles captured by CCD camera from $z = 31$ to 76 mm, with interval of 0.5 mm.	
9	Each figure is normalized individually.....	128
10	Figure 50 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 30^\circ$. (a)	
11	Schematic diagram of 30° -tilted diffractive axicon transforming the incident	
12	beam. (b-i) the lateral profile of resulted beam patterns transformed by the tilted	
13	diffraction angle captured at different distance from the lens. (j) X-cross-sectional	
14	profiles captured by CCD camera from $z = 31$ to 76 mm, with interval of 0.5 mm.	
15	Each figure is normalized individually.....	130
16		
17		

1 **List of Tables**

2	Table 1 SU-8 Specification.....	33
3	Table 2 Types and features of LSFM	51
4	Table 3 FemtoFiber pro NIR specifications.....	62
5	Table 4 Femtosecond laser Satsuma HP specifications.....	75

6

7

Chapter 1

Introduction

1.1 Research Background

Electromagnetic and optical engineerings are fast-developing research and industrial topics that are driven by the increasing need for simple rapid and customized devices s in a broad range of applications. One of the development trends is to extend the concept of the lab-on-a-chip (LOC) to produce the high-precision optical instruments to be portable, miniaturized, and flexible and can be applied in the specific environment under investigation. This trend triggers a great revolution in the design and fabrication of optical devices. Scientists and engineers are seeking a new frame of work that covers designing, fast fabrication, and performance testing of specific optical applications.

It is challenging to produce the optical devices in sub-millimeter, micro-, or nano-meter sizes using traditional fabrication methods, due to the machining limitation on the accuracy, precision, and resolution, and the difficulties to produce non-symmetric and non-spherical structures. Additive manufacturing (AM, also known as 3D printing) is a good choice for producing such complex geometric devices. Femtosecond laser direct writing (FsLDW) is the new promising AM technology to produce customized optical components for its high resolution and flexibility and address the aforementioned questions. A recent review article [1], by M. Malinauskas, et al., has summarized the developments

1 and combination applications for ultrafast laser processing AM technologies in
2 precision engineering, including the direct laser writing method.

3 Two-photon polymerization, realized by exposing a femtosecond laser
4 beam inside photoresists, is a new class of high-precision AM technique for
5 micro- and nano-scale objects [2]–[5]. The femtosecond laser induces the two-
6 photon polymerization (TPP) mechanism to produce the polymeric
7 microstructures, and thus it is a good option to fabricate the highly transparent
8 and precise optical components. Compared to the conventional fabrication and
9 other AM methods, FsLDW shows the distinct advantages of fabricating
10 arbitrarily designed structures, mask-free patterning, high-precision in micro- or
11 nano-scale, and low cost. Since the optical devices are designed and used for
12 specified research purpose and applications, the capability to fabricate the free-
13 geometric structures with proper optical properties are crucial. The technology
14 has been employed to fabricate the customized optical structures [6], including
15 micro-lenses [7]–[9], and waveguide structures [8], [10], cell biology [11], [12],
16 scaffolds for tissue engineering [13], etc. Therefore, the FsLDW is a good option
17 for the proposed fabrication of the micro-axicon used in light sheet fluorescence
18 microscopy and the diffractive Fresnel lens used in beam tilt angle detector,
19 which are the two proposed high-precision customized optics discussed later in
20 this thesis.

21 Since the first introduction of fluorescent optical sectioning technique to

1 3D biological imaging [14] and further developments to various light sheet
2 microscopies [15], high-resolution 3D imaging of micro-organisms in compact
3 imaging devices has been of great interest in the fields of developmental biology
4 [16], quantitative biology [17], [18], bio-sensing [19], and medical research [20].
5 The optical sectioning capability, without the need for any physical sectioning, is
6 the key enabler for this 3D imaging technique. Traditional optical microscopes
7 have the same optical axis for illumination and detection objectives, which result
8 in poor axial resolutions [16]. In contrast, the light-sheet fluorescence microscopy
9 (LSFM) realizes the optical sectioning techniques for tomographic 3D imaging
10 by separating the excited fluorescence plane (formed by the illumination
11 objective) from the detection focal plane (formed by the detection objective)
12 against the out-of-focus background signals. In LSFM, the illumination objective
13 is used to convert an input Gaussian beam into a sheet-like planar beam with the
14 aid of a cylindrical lens [14], [21]–[23] or by the lateral scanning of a Gaussian
15 beam [19], [23]. The detection objective is placed orthogonal to the illumination
16 axis, which captures the sectioned plane images illuminated by the thin light sheet
17 to achieve high axial resolution. It has been reported that selective plane
18 illumination microscopy (SPIM, one of the LSFM technologies) successfully
19 imaged sub-millimeter-size embryos and smaller creatures with a cylindrical lens,
20 with high-speed, wide field-of-view, low phototoxicity, low photobleaching, and
21 good penetration depth [21]. However, these light-sheet microscopes based on
22 the Gaussian beam have suffered from the trade-off between the sectioning

1 resolution (determined by the sheet-beam thickness) and the field-of-view
2 (determined by the illumination beam's depth-of-focus) [24]. Moreover, a bulky
3 lens with a high numerical aperture (NA) is required for generating thin light
4 sheets, which greatly restricts the imaging sample volume [15]. Therefore, for
5 more versatile 3D imaging of living cells and micro-organisms, there have been
6 strong demands for simple and compact illumination optics that can provide non-
7 diffractive light sheets. FsLDW technology provides the possibility of the fast
8 design and fabrication of compact optics.

9 Though the 3D-solid micro-lens has outstanding features in precise
10 optical engineering, a Fresnel lens enables further compression of the micro-lens.
11 The principle of Fresnel lens operation is straightforward given that the refractive
12 power of a lens functions only at the optical interfaces, i.e, the lens surfaces, one
13 can remove the lens's partial material as much as possible and maintain the lens
14 surface curvatures. If the thickness of every pixel in the lens plane of a refractive
15 lens is digitalized to binary, ternary, or higher levels, fabrication of such Fresnel
16 lens can be simplified. The amplitude Fresnel lenses are introduced when the
17 thickness is represented by the opaque degree at each pixel. Both the amplitude
18 and phase Fresnel lenses share the same patterns. The phase Fresnel lens has
19 zones of different thicknesses that induce the π phase shift between the
20 neighboring zones, so that generate the designed beam patterns. The amplitude
21 Fresnel lens has every other zone different transparency degree and the diffraction

1 occurs at the interfaces between the opaque-transparent zones. Such amplitude
2 Fresnel lens brings a new era to the lens design on a flexible thin sheet, serving
3 as a standalone lens or a superimposed function on an existing optical system.
4 The micro-axicon can be transferred to the two-dimensional binary amplitude
5 Fresnel lens with alternative transparent and opaque rings, that can produce
6 similar approximated Bessel beam profile by diffraction.

7 The beam alignment is one of the most important issues in optical design.
8 Both the theoretical and experimentally produced Bessel beams have non or less
9 diffractive propagation distances over a hundred times of the spot size and have
10 a large tolerance margin for the misalignment in the imaging and machining
11 applications. However, beam alignment detection and tilt angle measurement are
12 still critical processes in an optical system.

13 Currently, there are some techniques to detect the beam tilt angle or the
14 mechanical motion of an object with respect to the reference. These techniques
15 are usually based on interferometric methods. An interferometer is used to
16 generate beam patterns of constructive and destructive interference of the
17 deviated beam and the referenced coherent beam [25]. However, the destructive
18 interference contains quite less light intensity, so that it generates some loss in the
19 measurement precision, and reversely forces the source energy to be much larger
20 and even exceed the detector saturation threshold. Alternatively, some limited
21 information can be abstracted by detecting and measuring the difference between

1 the bright (constructive) and dark (destructive) zones intensity of the interference.
2 S. P. Walborn and his colleagues have creatively shown in [25] that quantum
3 metrology methods can unify the treatment for both classical and single-photon
4 fields. They presented the interferometric schemes in both Sagnac and Mach-
5 Zehnder-like configuration to estimate the tilt angle of an optical beam with
6 technical advantages in terms of precision and longitudinal position superiority.
7 Exploration of the quantum theory to classical optical science is reported to
8 resolve the two incoherent or partially coherent radiation sources. It leads to a
9 scheme for the optical ultra-resolution analysis that goes beyond the Rayleigh
10 criterion, as well as the discussion regarding the attainable precision at the
11 minimized source separation limitation. These schemes of the super-precision
12 analysis share the same issue that the system at least contains the light source,
13 beam splitter, motion stage with the reflecting mirror, the interferometer arms
14 including the guiding mirrors, and the spatial or polarization detector and
15 measurement system. In this thesis, the proposed FsLDW technique is used to
16 produce the light-weight and thin-sheet Fresnel lens that can replace the bulky
17 instruments and fulfil the same function to detect the beam tilt angle and fine
18 mechanical motion.

19 In summary, to realize the full potential of fabrication of micro- or nano-
20 optical 3D polymeric optical devices by FsLDW, the following research targets
21 were identified: design, and fabrication of optical devices for specific

1 applications, controlling the properties of FsLDW printed objects, and optical
2 performance characterization and real-world application of the fabricated optical
3 elements.

4

5 **1.2 Research Objectives**

6 This research focuses on the realization of the Femtosecond Laser Direct Writing
7 (FsLDW) technique for microstructure additive manufacturing (AM), and the
8 design, fabrication, and performance characterization of customized optical
9 components and devices. I aim to demonstrate two optical applications with the
10 customized optics that are fabricated by the femtosecond laser assisted AM. Three
11 proposed objectives are presented as following in the order: 1) the establishment
12 and parametric study of the FsLDW technique for the 3D micro-structures
13 fabrication; 2) the realization of the Light Sheet Fluorescence Microscopy using
14 the real product micro-axicon by FsLDW; and 3) the tilt angle measurement by
15 the FsLDW diffractive Fresnel axicon.

16

17 **Two-Photon Polymerization and Additive**

18 **Manufacturing by**

19 **Femtosecond Laser Direct Writing Technique**

20 The main objective of the study is to fabricate micro-optical elements and explore

1 their practical uses. The full production cycle starts from designing the optical
2 components and their simulation ZEMAX OpticStudio®, transforming the
3 optimized virtual model to the CAD model and programming the laser direct
4 writing routine, fabricating components with FsLDW, followed by performance
5 characterization of printed structures. Femtosecond Laser Direct Writing
6 (FsLDW) technique is selected to fabricate the micro-structures in this research.
7 The FsLDW system is set up and tested for its feasibility of micro-fabrication.
8 The system is established with a femtosecond laser source, beam-guiding mirrors,
9 a nano-positioning stage that fits the samples, and a real-time monitoring camera.
10 The photoresist SU-8 and the reduced graphene oxide (rGO) are selected among
11 the candidates including Polyethylene glycol diacrylate (PEGDA), Polymethyl
12 methacrylate (PMMA), and gelatin, etc, to produce the proposed optical devices.
13 The objective of this section is to fabricate the designed optical elements with
14 sufficient precision and surface quality and test the optical performance to realize
15 the proposed functions.

16

17 **Micro-Axicon Design, Fabrication, and** 18 **Its Application in Light Sheet Fluorescence Microscopy**

19 The objective is to demonstrate the potential for light sheet fluorescence
20 microscopy with the self-reconstructing Bessel-Gauss beam formed by a two-
21 photon polymerized micro-axicon. The polymeric micro-axicon will be produced

1 by polymerizing the photoresist SU-8 with the above-mentioned FsLDW process.
2 The capability of the micro-axicon to transform a Gaussian beam to a Bessel
3 beam will be investigated and compared with the results obtained using a
4 commercial axicon. The potential for light sheet fluorescence microscopy and
5 bio-imaging with the customized FsLDW-built micro-axicon will be investigated.

6

7 **Collimated Beam Tilt Angle Measurement with** 8 **Diffractive Axicon Generated Pattern**

9 In this work, a beam tilt angle detector with the regularly evolving patterns
10 formed by the graphene oxide diffractive Fresnel lens will be investigated. A
11 lightweight, thin sheet diffractive lens using reduced graphene oxide (rGO) will
12 be designed and fabricated with the femtosecond laser direct scanning system
13 developed from the FsLDW technique. The diffractive lens's transformation of
14 the incoming collimated beam will be captured at different tilt angles. By
15 analyzing the resulted pattern evolution from the diffractive lens, the beam tilt
16 angle measurement will be performed.

17 The contribution and scope of each objective in this full research are
18 summarized in the next section.

19

1.3 Contribution and Scope

1) Additive manufacturing of microstructures with Femtosecond laser direct writing technology: In the initial phase of this research, the femtosecond laser direct writing system will be established, working as a robust, cost-effective, and flexible design method to produce the customized micro-optics structures with an unlimited selection of various photo-sensitive materials and flexible substrates. The successful fabrication of the proposed optics that achieve good performance in real applications will be demonstrated.

2) Potential for light sheet fluorescence microscopy with micro-axicon created Bessel-Gauss beam: In this research, I will demonstrate the potential for light sheet fluorescence microscopy using the self-reconstructing pencil-like Bessel beam formed by a two-photon polymerized micro-axicon. The polymeric micro-axicon will be fabricated by the FsLDW process. The small spot size of the Bessel beam created by the micro-axicon is expected to support high-resolution optical sectioning, and the extended-depth-of-focus is expected to enable a wide field-of-view in 3D tomographic imaging, and the self-reconstructing beam is expected to facilitate illumination of target samples located even behind scattering micro-particles or located in multi-layered micro-structures. Microscopic imaging of Hela cells will be carried out to demonstrate these capabilities.

3) Tilt angle detection and measurement with rGO-thin film diffractive axicon: The sensing of the beam tilt angle with a thin film diffractive axicon is proposed.

1 The thin film graphene-oxide diffractive axicon will be fabricated by high-speed
2 femtosecond laser direct scanning (FsLDS) system, which is an extension of the
3 FsLDW process. The transformation of the Gaussian beam by this diffractive
4 axicon will be investigated and the beam tilt angle sensing using the diffractive
5 axicon based measurement system will be demonstrated.

6

7 **1.4 Report Organization**

8 This thesis consists of six chapters. Chapter 1 highlights the research background
9 and listed objectives following the research flows. In Chapter 2, the literature
10 review covers the overview of additive manufacturing (AM), the principle of
11 FsLDW technology, and the state-of-art of AM application in optical engineering.
12 It follows by the mathematical formulation of the Bessel beam and its generation
13 approach from the Gaussian beam, which provide the foundation of the optics
14 design and applications. Chapter 2 also introduces the different schemes of light
15 sheet fluorescence microscopy. To compare the proposed technique with the
16 previous studies, the strengths and drawbacks of the previous techniques are
17 reviewed and discussed. The last section of Chapter 2 introduces the existing
18 interferometric sensing methods of the tilt angles of the collimated beam. Chapter
19 3 covers the establishment of the femtosecond laser based additive manufacturing
20 system, and the parametric study of the micro-fabrication. It provides the optical
21 fabrication condition for further studies. Chapter 4 describes the scheme of the

1 light sheet fluorescence microscopy with the FsLDW micro-axicon. The
2 transformation properties and self-reconstructing capability are investigated and
3 are compared with the simulation results. Fluorescence microscopy is performed
4 on polystyrene micro-beads and live Hela cells. Chapter 5 describes the scheme
5 of the collimated beam tilt angle measurement with the thin film diffractive
6 axicon fabricated using FsLDS technology. It compares the performances of the
7 commercial refractive axicons and the FsLDS diffractive axicons. This chapter
8 demonstrates the beam tilt angle measurement by detecting the regular evolution
9 of diffractive axicon generated patterns from the Gaussian beam. Chapter 6
10 provides the conclusion of this research contribution and recommendations for
11 future work.

1 Chapter 2

2 Literature Review

3 2.1 Preamble

4 This chapter briefly provides a glance at the development of additive
5 manufacturing (AM) technology, and the existing AM methods widely used in
6 the research and industrial field. The proposed applications in this research
7 require the fine fabrication method to produce the optical devices, which can be
8 fulfilled with the appearance of femtosecond lasers and the deep exploration of
9 the photoresist.

10 One of the proposals central to this project is light sheet fluorescence
11 microscopy with the flexible miniaturized lens. Here introduces the conventional
12 light sheet microscopy that is well developed in the community, but yet too huge
13 to use in-vivo environments or severe scenarios. We explore the transformation
14 of the Bessel beam and the possibility to realize the light sheet fluorescence
15 microscopy with customized miniaturized lenses. The second proposal is the beam
16 tilt angle detector and measurement. Comparing the existing different methods of
17 beam tilted angle measurement in this chapter, integrating with the clear review
18 and investigation of Bessel beam generated by various lenses, we again attempt
19 to realize the beam tilt angle measurement with compact micro-optics.

2.2 Introduction of Additive Manufacturing

The Additive Manufacturing (AM) technology, also named three-dimensional printing (3DP), was developed from the rapid prototype (RP) since the 1990s. According to American Society for Testing and Materials (ASTM) 2792-12, AM is defined as the process of joining material to form objects derived from 3D model data, in a layered manner. It is a process opposed to subtractive manufacturing fabrication methodologies” [26]. It is powerful, by concept, to customize and tool the direct parts manufacturing with complex geometry. Its capability to process with a wide range of materials and fabricate any complex-geometric functional three-dimensional structures makes this technology outstanding from the conventional manufacturing methods, including formative and subtractive manufacturing.

Figure 1 Comparison between different manufacturing methods (adapted from [27])

1 Nowadays it has been widely used as an indispensable method in research
2 and industry. AM allows the three-dimensional (3D) construction of volumetric
3 objects layer by layer as shown in Figure 1, directly from a computer-aided design
4 (CAD) model [28], [29], without or minimizing the need for post-tooling and
5 machining. It dramatically reduces the material waste and the mould preparation,
6 as well as reduces the production duration from design to the final products.
7 Compared with conventional methods, including formative and subtractive
8 manufacturing, which requires finer machining tools, this method has the intrinsic
9 flexibility to produce almost any dimensions and geometries with proper
10 materials of selective mechanical, chemical or optical characteristics. It is
11 important to announce that AM technology became possible by other
12 technologies, including computer-aided design (CAD), computer-aided
13 engineering (CAE), and computer numerical control (CNC) [30], [31].

14 AM technologies nowadays have developed simple ink-jet printing [32],
15 [33], the LIGA process (stands for lithography, electroforming, and molding) [34],
16 ultraviolet lithography [35], direct laser writing [6], [36], [37], and other methods
17 reported in references [38]–[42]. However, these technologies suffer from
18 drawbacks such as limited miniaturization, inability to combine multiple
19 elements, restrictions in designing the surface geometries [33], [34], [39], [40],
20 and problems with the structure or optical beam alignments. Precision fabrication
21 of micro-sized free-form optics requires high-quality control of the surface figure,

1 surface roughness, surface waviness, transparency, and refractive index. Three
2 representative technologies satisfying these requirements are mask-based
3 photolithography, mask-less electron-beam lithography, and mask-less focused
4 ion-beam process [43], [44]. Photolithography requires pre-designed masks, and
5 thus it has low flexibility in pattern design and manufacturing; the electron-beam
6 and ion-beam processes enable flexible manufacturing with high resolution but
7 with a low throughput due to the time-consuming beam scanning process. The
8 community is seeking novel technologies to address the problems.

9 So far, AM products can be made of various polymers [31], [45]–[47],
10 metals [48]–[52], ceramics [53], and hybrid materials. This enables AM to be
11 employed in different fields of research and industry, including the automotive
12 and aerospace industry [49], furniture, architecture [54]–[56], medical, bio-
13 medical engineering [57]–[62], dental, micro-electromechanical systems
14 (MEMS), as well as ultra-precision engineering [29], in the order from large
15 macro-scale to the micro-metric. For different applications, energy sources of AM
16 techniques range from heat, laser, electron beam, etc.

17 Nowadays there is increasing demand for micro-fabrication technologies
18 and merging them into some new manufacturing platforms, due to the tendency
19 towards miniaturization of products in the fields of electronics, optics, automotive,
20 and bio-medicines. Currently, it has been developed into sub-micro-metric. Major
21 kinds of micro-fabrication are classified as micro-electromechanical systems

1 (MEMS), micro-optical electronics systems (MOES), and micro-opto-
2 electromechanical systems (MOEMS), depending on the fundamental operations
3 [28], [29], [63]. Besides, micro-fabrication has a bottleneck attracting great
4 attention that nowadays products demand the employment of the hybrid of
5 materials and complex-geometric structure, and reduce of time duration from the
6 needs for products arise until the finished products are put to use.

7 A wide range of micro-fabrication technologies has been reported with
8 different capabilities and applications. They can be classified as lithographic and
9 non-lithographic according to manufacturing methods. Since the 1970s, micro-
10 fabrication adopted the 3DP concept, and developed the technologies, such as the
11 LIGA process and soft lithography, which are limited to 2.5D objects, and multi-
12 layered photolithography, which attempts to produce toward true 3D structures.
13 Thereafter technologies including the poly-jet [64], selective laser sintering,
14 scalable AM technology, fused deposition modeling, electron beam melting,
15 laminated object manufacturing, 3D direct writing, and hybrid processes [29], are
16 considered to realize the true 3D geometric micro-fabrication.

17 **2.3 Ultra-Fast Laser Assisted**

18 **Additive Manufacturing Technology**

19 The rapid development of ultrafast laser systems with high energy density in a
20 wide spectrum has triggered the application in precise micromachining and
21 micro-fabrication. In 1994, there was pioneering work on ultrafast laser

1 micromachining [65], [66], including picosecond (ps, 10^{-12} second) and
2 femtosecond (fs, 10^{-15} second) laser machining, which was used to ablate micro-
3 sized structures on silica and silver surface. The precision and resolution of the
4 ultrafast laser micromachining have developed to nano-scale in a decade [67]–
5 [70]. Since then, the direct laser writing (DLW) technology became true.
6 Research on single-photon direct laser writing (DLW) was firstly reported in early
7 1990s.

8 The femtosecond laser was found to exercise nonlinear optical
9 phenomena, including the two-photon excitation and polymerization. The laser
10 having a pulse duration of less than 100 fs scale and repetition rate of more than
11 100MHz has been built to output with enduring stability. DLW with two or multi-
12 photon phenomena, also named nonlinear optics, was then realized with the
13 femtosecond laser, and provides the theoretical potential to achieve the resolution
14 beyond the light diffraction limit. These properties that it functions extremely
15 high power density within fs scale pulse and a rather long cooling down period,
16 are then utilized to create micro/nano-structures for replacing or complementary
17 functions to some applications, such as micro-fabrications, micro-chip machining,
18 and etching. The techniques involved with the nonlinear phenomena were
19 immediately embraced by the photonics community, providing the capability for
20 the fabrication of fully 3D nanostructures. Successful reports include
21 ultracompact multi-lens objectives [36], the micro-lens array with a different

1 curvature (MLADC) unit lens for curvature correction and real-time 3D imaging
2 [71], micro-scale 3D lithography for micro-lens, and an array of nine single
3 micro-lenses [4] that performs impressive results comparable to the reported
4 spatial light modulator [72].

5

6 **2.3.1 Femtosecond Laser**

7 The ultrafast laser refers to the lasers possessing an electromagnetic pulse whose
8 time duration is in the order of a picosecond or less, basically contains two groups:
9 femtosecond and picosecond lasers. They have four outstanding features that
10 enable their functions in precision engineering: high peak intensity, ultrashort
11 pulse duration, high pulse repetition rate, and broad spectrum [69]. This section
12 focuses the review on femtosecond lasers.

13 A femtosecond laser pulse can be produced with the mode-locking
14 mechanism, together with a saturable absorber. The word mode-locking describes
15 the locking of multiple axial modes in a laser cavity. By enforcing coherence
16 between the phases of different modes, pulsed radiation can be produced. Mode-
17 locking is a resonant phenomenon [73], [74]. The pulsed radiation can be
18 produced by enforcing the coherence between phases of different resonant modes.
19 The basis of the mode-locking technique is to induce the constant phase
20 relationship between the longitudinal modes of the laser's resonant cavity. In

1 contrast to the continuous laser which has oscillating resonant modes
2 independently and produces a random or constant output intensity, the ultrafast
3 pulse has each mode operates with a fixed phase between it and the other modes,
4 the modes of the laser periodically all constructively interfere with one another,
5 forces the intense burst or pulse of light. The mode-locking methods can be
6 classified to two groups: active and passive mode-locking.

7 The first indications of active mode locking appear in the work of Gürs
8 and Müller in German on ruby lasers [74], [75], and Statz and Tang on He–Ne
9 lasers [76]. The first papers identifying the mechanism were written in 1964 by
10 DiDomenico [77], Hargrove et al. [78], and Yariv [79]. Among them, Hargrove
11 et al. achieved mode-locking by internal loss modulation inside the resonator. An
12 optical Fabry–Pérot resonator is formed of two mirrors that has electromagnetic
13 field bounding in between with axial modes of frequencies $\Omega = 2\pi n/T_R$, where T_R
14 is the roundtrip time, and the frequency interval $\Delta\Omega = 2\pi/T_R$. Denote the
15 frequency of the central mode by ω_0 . The laser is mode-locked by an amplitude
16 modulator placed near one of the mirrors. A cosinusoidal modulation of the
17 central mode at the frequency produces sidebands at $\omega_0 \pm \Delta\Omega$. These injections
18 lock the adjacent modes, which in turn lock their neighbors. The locking can be
19 described as a coupling, and it is only dissipative because of the dissipative nature
20 of the modulation.

21 In contrast, passive mode-locking is the technique that does not require a

1 signal external to the laser to produce or modulate pulses. Rather, they use the
2 light in the cavity to cause a change in some intracavity element, which will then
3 itself produce a change in the intracavity light. A commonly used device to
4 achieve this is a saturable absorber. The passive mode-locking mainly has two
5 methods to realize: the Q-switch and saturable absorber. Mocker and Collins [80]
6 showed that the saturable dye used in ruby lasers to Q-switch the laser could also
7 be used to mode lock. Q-switching is the process in which the laser is caused to
8 emit pulses that are many roundtrips in duration. The saturable dye, as the gain
9 medium, is bleached by the radiation in the cavity. The radiation emission stops
10 when the gain medium is depleted, and the process starts all over again. Mocker
11 and Collins observed that the Q-switched pulses broke up into a train of ultra-
12 short pulses separated by the roundtrip time. However, the accuracy of the pulse
13 measurements was affected by the somehow unpredictable nature of the transient
14 mode-locking. This remains a drawback until Ippen, Shank, and Dienes [8]
15 reported generating the first CW saturable absorber mode-locking using a
16 saturable dye in a dye laser. The first laser pulses of sub-picosecond duration were
17 generated with the assistance of a passive saturable-absorber element in the laser
18 [81], [82], passive mode-locking has been one of the main streams for generating
19 shortest pulses. Ultimately, a record 6-fs pulse duration was achieved by Fork et
20 al. using pulse compression external to the cavity [83]. The saturable absorber
21 reported in 1992 [84], usually referred to as semiconductor saturable absorber
22 mirrors (SAM), to launch a self-amplitude modulation of the laser power inside

1 the resonant cavity by introducing high loss to low laser intensity while low loss
2 during the short pulse of high intensity exists. In this manner, the performances
3 of ultrafast lasers have been improved by a few orders of magnitudes in pulse
4 duration and stability.

5 Nowadays the laser pulse duration has been constrained to range from
6 order of ps down to several fs depending on the laser source material and the
7 parameters of the saturable absorber. The ultrashort pulse allows a fast temporal
8 resolution. The pulse repetition rate has been developed to reach more than 100
9 GHz. The average power goes to as high as 60 W in femtosecond pulse, much
10 smaller than the CW lasers that reach several kilowatts but has outstanding pulse
11 energy larger than 1 mJ. Therefore, the ultrafast laser can exercise the nonlinear
12 optical phenomenon, that is to exercise a single molecule with two or more
13 photons simultaneously, and thus two-photon absorption (TPA) and
14 polymerization (TPP) occur. In contrary to single-photon polymerization, the TPP
15 occurs only at the focal voxel of the laser beam within the nanometric scale.
16 Therefore, the ultrafast laser, especially femtosecond laser machining, with TPP
17 reaches higher spatial resolution than the optical diffraction limits.

18 One of the most important features of femtosecond laser machining is that
19 it can be considered as cold processing since its ultrafast pulse within sub-
20 picoseconds effectively suppresses the heat-affected zone in the laser-scanned
21 region of materials as shown in Figure 2. Energy deposition duration of

1 femtosecond machining is greatly shorter than the relaxation process timescale in
 2 the order of 10^{-7} scale. Hence, femtosecond laser micromachining is considered
 3 as a cold processing method. The femtosecond laser machining shows a better
 4 surface and edge quality than nano- and picosecond laser machining in Figure 2.

5
 6 Figure 2 Laser-material interaction for different lasers. (a) femtosecond laser induces
 7 minimized HAZ (image credit: IMRA Application Note "Pulsed Laser Deposition with the
 8 FCPA μ Jewel Femtosecond Fiber Laser", <https://www.imra.com>.) (b-d) Examples of
 9 micro-machining with different pulse widths from nanosecond, picosecond, to
 10 femtosecond regimes and their HAZs [85]

11
 12 Femtosecond laser nowadays develops several types: bulk laser, fiber
 13 laser, semiconductor laser, and dye laser. Nowadays fiber laser is widely used

1 considering its advantages: 1) higher temporal and spatial resolution and beam
2 quality, 2) high reliability with a lifetime of at least 25,000 lase hours and
3 maintenance-free, 3) high stability, and 4) cheaper and more compact than bulk
4 lasers.

5 To date, there are several published methods to fabricate the
6 microstructures, using different energy sources including laser interference
7 lithography through the micro-lens array (MLA) [86], electron-beam lithography,
8 and reactive ion etching, etc. Laser-based 3D printing technologies consist of
9 stereolithography, selective laser sintering. However, these fabrication methods
10 face the challenges that either they are unable to reach the high resolution towards
11 nanometric scale to cater for application with specific wavelengths or spectrum,
12 or they are not flexible enough to produce geometric complex structures.
13 Compared to the other technologies, femtosecond Laser Direct Writing (FsLDW)
14 technology creates a superior resolution in nano-scale, and it does not require the
15 need of preceding fabrication of a photomask, recoating process of materials, or
16 layer-by-layer fabrication. FsLDW induces the two-photon polymerization (TPP),
17 the process which is introduced in the next section, at the accurate position in
18 polymeric materials and thus additive manufacturing is expected to address the
19 problem of low resolution and complex geometries. Two-photon polymerization
20 occurs only at the center of the laser focal spot and reaches a higher resolution
21 than the optical diffraction limit. The direct laser writing (DLW) method offers

1 the convenience to produce any geometric structures without the assistance of
2 photomasks or other tools. On the other hand, the FsLDW process is much slower
3 than those two methods. More recently, direct laser writing (DLW) has developed
4 applications in more diverse fields, such as microfluidics [63], [87], biomedical
5 implants [61], tissue engineering [88], [89], and in 3D scaffolds for cell cultures
6 [72], [90].

7 TOPTICA FemtoFiber pro NIR laser (780 nm/1560 nm, 100 fs, 80 MHz)
8 is used in this study for the FsLDW. The engine of TOPTICA's fiber lasers is a
9 SAM mode-locked ring oscillator. The configuration is protected by an optical
10 isolator and is reported to be robust against back reflections. The high stability of
11 the mode-locking is attributed to the operation in the solitonic regime. The soliton
12 pulse is amplified to high peak power in a subsequent core-pumped fiber
13 amplifier. The laser system uses a Master Oscillator Power Amplifier (MOPA)
14 platform which is a polarization-maintaining all-fiber setup. By using Erbium-
15 doped fibers, the system emits the laser beam at 1560 nm wavelength. The
16 TOPTICA FemtoFiber series, using SAM mode-locking technique and
17 polarization-maintaining fiber technology, make its laser system the state-of-the-
18 art ultrafast fiber laser series and is well utilized in our research. It produces the
19 ultrafast laser pulse at 1560 nm and 780 nm with the second harmonic generation.
20 The repetition rate is standard 80 MHz while pulse duration is less than 100 fs
21 with a maximum power of 350 mW.

2.3.2 Two-Photon Polymerization Mechanism

The two-photon absorption (TPA) process was theoretically proposed by Göppert-Meyer in 1931. The two types of TPA processes are degenerate and non-degenerate processes [91]. The degenerate process refers to the simultaneous absorption of two photons of the same wavelength, while the non-degenerate one is the process of absorbing two photons with different wavelengths [3]. This work mainly focuses on the degenerate TPA process.

Taking the TOPTICA FemtoFiber pro NIR laser of 780 nm as the example, the UV-sensitive photoresist filters out the laser beam at the near-infrared (NIR) wavelength, however, the intense power density enforces the TPA process at the focal point of the laser beam, which is equivalent to the absorption of a photon in UV spectrum of 390 nm within the photoresist molecules. This induces the crosslinking of the photoresist molecules to form solid objects within a restricted voxel. Thereby, photoresist-based micro-sized free-form optical components can be manufactured with high precision within a reasonably short working time via two-photon polymerization. Negative photoresists, such as SU-8, are well-known for their high chemical stability, high mechanical stability for ultra-precision manufacturing [63], [92], and high optical quality over infrared, visible, and ultraviolet wavelengths for optical applications [2], [36], [43], [93].

Figure 3 Comparison of single- and two-photon absorption and polymerization.
 (Top) the excited electron states under single- and two-photon absorption;
 (Bottom) the TPP voxel is much smaller than the 1PP voxel

Generally, the TPA cross-section area is much smaller as compared to one-photon absorption, which is comparable to the optical diffraction limit [94] [95], as shown in Figure 3 [96]. The TPA process is only realizable with the existence of the ultrafast laser. TPA and the subsequent cross-link (two-photon polymerization TPP) occurs only at the focal voxel inside the SU-8 film, where the power intensity exceeds the threshold of the SU-8 polymerization as the green

1 region shown in Figure 4 schematic diagram, instead of occurring along the laser
 2 beam. This property enables the fabrication of precise 3D microstructure by 3D
 3 printing with SU-8 [91], [97].

4
 5 Figure 4 (left) the CW laser excites the entire column of sample and
 6 (right) the ultra-fast pulse laser excites only a small spot of the sample

8 **2.3.3 Photoresist Material**

9 The photoresist is a light-sensitive material that produces patterned coating or
 10 structures when is illuminated and thus absorbs adequate photon energy. It is
 11 commonly used in several processes, such as photolithography and
 12 photoengraving, to form complex geometries. This process is crucial in the
 13 electronic industry, shape memory alloy (SMA) applications [98], and
 14 microfluidic devices for neuroscience research [99], etc. Two categories of

1 photoresists are positive and negative tone resists. Negative refers to a photoresist
2 whereby the parts exposed to UV or two-photon IR-excitation become
3 crosslinked to solidify, while the remainder of the material remains unchanged
4 and soluble to photoresist developer (1-methoxy-2-propanol acetate). and is
5 washed away after curing in the development process. SU-8, a negative
6 photoresist, is a commonly used epoxy-based photoresist to fabricate the highly
7 precise optical components and other high-aspect-ratio 3D microstructures [93].
8 SU-8 3D structure is additively manufactured by UV beam scanning inside the
9 vat of raw liquid SU-8 along the pre-designed routine and stacking the cross-
10 linked voxels to form the volume. The other type of resists, positive photoresist,
11 functions in the counter way as reductive manufacturing, that its portion exposed
12 to light becomes soluble to the photoresist developer, while the unexposed
13 portion of the photoresist is insoluble in the developer.

14 SU-8 derives its name from the presence of 8 epoxy groups (a statistical
15 average per moiety) as shown in the structural schematic in Figure 5. These
16 epoxies are cross-linked under UV exposure to give the final structure with the
17 assist of baking and development with 1-methoxy-2-propanol acetate [87], [91],
18 [94], [100].

Figure 5 Schematic of the SU-8 molecule [87]

SU-8 3000 series are chosen out of the epoxy series in this research. The selection of materials must consider the following conditions catering to the applications:

1. Geometric aspect ratio and fabrication accuracy: the ratio of structures' longer dimensions to shorter ones, which in this fabrication, refers to the ratio of height to base dimension.
2. Surface finish and quality: the specifications describing the degree of a surface deviating from the shape of a reference surface, and the cosmetic appearance and includes such defects as scratches and pits, or bumps.
3. Transparency to the visible spectrum, especially in range of 532 – 780 μm .
4. Refractive index.
5. Laser damage threshold, i.e, the mechanical and chemical stability: Laser

1 damage threshold refers to the maximum amount of laser power intensity
2 that the optics can withstand before the damage, whose values are given
3 for both pulsed and CW lasers.

4 Figure 6 indicates that SU-8 is capable to be well fabricated high aspect-
5 ratio structures. The cured SU-8 is a mechanically and chemically stable material
6 and is transparent to a wide spectrum of visible light, which is an ideal candidate
7 to fabricate the optical devices.

8
9 Figure 6 collection of different geometries of SU-8 high aspect-ratio structures
10 produced by lithography [101], [102]

11

Figure 7 Optical characteristics of SU-8: (a) Absorption spectrum of SU-8, plotted on a logarithmic scale. (b) Fluorescence photon number versus excitation intensity [103]

Figure 7 shows the absorption spectrum of SU-8, on a logarithmic scale,

highlighting the very low absorption coefficient, $\sigma < 723 \text{ m}^{-1}$, at $\lambda > 532 \text{ nm}$.

Larger the TPA cross-section area, the more efficient the two-photon polymerization activity [104]. The polymerized SU-8 performs good

transparency to the visible to NIR spectrum as in Figure 8, hence it is a good

material candidate for the optical devices.

1

2 Figure 8 SU-8 Transmittance after 300°C Hard Bake for different durations (image
3 credit: MicroChem datasheet for SU-8 3000 Permanent Epoxy Negative Photoresist)

4 Some of the polymerized SU-8 specifications are listed in the table:

5 Table 1 SU-8 Specification

Property	Value
Mass density	1.190 g/cm ³
Refraction index (polymerized)	1.52–1.74, at $\lambda = 300\text{-}1040$ nm 1.54, at $\lambda = 633$ nm 1.58, at $\lambda = 532$ nm
Young's modulus	4.02–4.95 GPa
Tensile or fracture strength	34 Mpa
Dielectric constant (at 1GHz)	3.28
Magnetic permeability	1.08
Biocompatibility	Good
Hydrophobicity	Hydrophobic

6

7

2.4 Bessel Beam's Generation and Characterization

Bessel beam is one of the well-known non-diffractive classes of the solution to the Helmholtz Equation in free space and is widely applied in the optical industry due to its outstanding features and simple generation of the approximated beam, the so-called Quasi-Bessel beam.

2.4.1 Mathematical Explanation of Ideal Bessel Beam

B.E.A Saleh and M.C. Teich have summarized the derivation of the Bessel beam function in *Fundamentals of Photonics* [105]. A monochromatic wave can be represented by a paraxial wavefunction $u(\mathbf{r}, t)$ with harmonics time dependence:

$$u(\mathbf{r}, t) = a(\mathbf{r}) \cos[2\pi\nu t + \varphi(\mathbf{r})] \quad (2.4.1.1)$$

where $\mathbf{r} = (x, y, z)$ is the position, $a(\mathbf{r})$ = amplitude, $\varphi(\mathbf{r})$ = phase, ν = frequency (Hz), $\omega = 2\pi\nu$ = angular frequency (radians/s), and $T = 1/\nu = 2\pi/\omega$ = period (s).

$u(t)$ can be described as the real part of the complex wavefunction $U(\mathbf{r}, t)$. and $U(\mathbf{r})$ refers to the complex amplitude function:

$$U(\mathbf{r}, t) = a(\mathbf{r}) \exp[j\varphi(\mathbf{r})] \exp(j2\pi\nu t) = U(\mathbf{r}) \exp(j2\pi\nu t) \quad (2.4.1.2)$$

Substituting $U(\mathbf{r}, t) = U(\mathbf{r}) \exp(j2\pi\nu t)$ into the wave equation

$$\nabla^2 U - \frac{1}{c^2} \frac{\partial^2 U}{\partial t^2} = 0 \quad (2.4.1.3)$$

1 we obtain the Helmholtz equation

$$2 \quad \nabla^2 U + k^2 U = 0, \quad k = \frac{2\pi\nu}{c} = \frac{2\pi}{\lambda} \quad (2.4.1.4)$$

3 k is referred to as the wavenumber. There exist multiple solutions to the equation.

4 Different solutions are obtained with different boundary conditions.

5 In the exploration of the beam-like waves, consider the complex
6 wavefunction that has an axial symmetric characterization:

$$7 \quad U(\mathbf{r}) = A(x, y)e^{-j\beta z} \quad (2.4.1.5)$$

8 In order to satisfy the Helmholtz equation (2.4.1.4), $A(x, y)$ must follow the two
9 dimensional Helmholtz equation

$$10 \quad \nabla_T^2 A + k_T^2 A = 0 \quad (2.4.1.6)$$

11 where $k_T^2 + \beta^2 = k^2$ and $\nabla_T^2 = \partial^2 / \partial x^2 + \partial^2 / \partial y^2$ is the transverse Laplacian
12 operator. The amplitude $A(x, y)$ can be expressed in the polar coordinates

$$13 \quad \begin{aligned} A(x, y) &= A_m J_m(k_T \rho) e^{-j\beta\phi}, \\ x &= \rho \cos \phi, \quad y = \rho \sin \phi, \quad m = 0, \pm 1, \pm 2, \dots, \end{aligned} \quad (2.4.1.7)$$

14 J_m is the m^{th} order Bessel function of the first kind, A_m is a constant. Note that the
15 solutions that are singular at $\rho = 0$ are not included.

16 For $m = 0$, the wavefunction can be expressed as

$$17 \quad U(\mathbf{r}) = A_0 J_0(k_T \rho) e^{-j\beta z} \quad (2.4.1.8)$$

1 which has a planar wavefront. All the wave rays (wavefront normals) are parallel
 2 to the beam axis. The beams having the intensity of J_m^2 are defined as the Bessel
 3 beams. The intensity distribution of 0th order Bessel beam is

$$4 \quad I(\rho, \phi, z) = |A_0|^2 J_0^2(k_T \rho) \quad (2.4.1.9)$$

5 The beams are circularly symmetric intensity around the axis, and the intensity
 6 distribution in the transverse plane is independent of z as shown in Figure 9.

7
 8 Figure 9 the intensity distribution f Bessel beam along z-axis

9
 10 It is important to point out that the complex amplitude of the Gaussian
 11 beam is only an approximate solution of the Helmholtz equation, while the Bessel
 12 beam is an exact solution to the equation [105]. The field qualities of the beams
 13 are compared in Figure 10. Only the 0th order Bessel beam of the first kind and
 14 the Gaussian beam have non-zero intensities at the central beam, the rest of Bessel
 15 beams of higher orders have a minimum at the center.

Figure 10 Comparison of radial field strengths of a Gaussian beam and the Bessel beams of 0th - 4th orders

2.4.2 Characterization of Experimentally Generated Quasi-Bessel Beam

It has been widely proven and reported that the Bessel beam is one of the easiest generated non-diffractive beams. In contrast to the Gaussian beam which has a finite beam radius, Mathematically the ideal Bessel solution to the Helmholtz equation has infinite transverse RMS, or so-called the side rings to $\rho = \infty$, and carries infinite power over the infinite area. Thus, the ideal Bessel beam does not exist. However, one can produce the Quasi-Bessel beams that have transverse profiles analogous to Bessel functions.

There have been several different generation methods of Quasi-Bessel beams since 1987 when J. Durnin and J.J. Mceeli, Jr. [106], [107], firstly showed

1 their result to produce the approximate Bessel beam by illuminating a circular slit
2 located in the focal plane of a focusing lens, as shown in Figure 11 [106].

3
4 Figure 11 (a) Experimental arrangement for the approximate 0th order Bessel beam
5 with a circular slit and a focusing lens; (b) the profiles of generated J_0 beam and the
6 comparable Gaussian beam with same spot size at the beam waist.

7

8 The Quasi-Bessel beam was created by the setup shown in Figure 11. The
9 circular slit of mean diameter $d = 2.5$ mm and $\Delta d = 10$ μ m was placed at the focal
10 plane of the focusing lens, which has a focal length $f = 305$ mm and radius $R =$
11 3.5 mm. A collimated beam of wavelength $\lambda = 632$ nm is incident normal to the

1 slit. It can be regarded as the set of plane waves that has wave vectors lying on
 2 the cone surface. The beam produces the approximate non-diffractive Bessel
 3 beam transverse profile having a spot size (FWHM) of 70 μm as the shown solid
 4 line in Figure 11(b). The resulted beam has a central spot with

$$5 \quad \text{Wave vector } \alpha = \frac{2\pi}{\lambda} \sin \theta, \text{ beam radius } r = \alpha^{-1}, \quad (2.4.2.1)$$

6 where $\theta = \tan^{-1}(\frac{d/2}{f})$, and the propagation distance

$$7 \quad \begin{aligned} Z_{\max} &= R / \tan \theta \approx R / \sin \theta \\ &= \frac{R}{\alpha / k} = 2\pi r R / \lambda \end{aligned} \quad (2.4.2.2)$$

8 The Bessel beam profile is invariant to the z-axis. However, the Gaussian
 9 beam having the same beam waist FWHM of 70 μm exhibits a rapid diffractive
 10 spreading and decrease of peak intensity. The dashed line (i) is the Gaussian
 11 profile taken at $z = 0$, and the profiles of line (ii) at $z = 10$ cm and line(iii) at $z =$
 12 100 cm have been multiplied by a factor of 30 and 2000 respectively for visibility.

13 This method of Bessel beam generation by the circular slit dramatically
 14 blocks the beam energy and produces fluctuating axial intensity. Evolving from
 15 the concept that the beam rays forming the Bessel beam are lying on the surface
 16 of a cone, there are plenty of trials to produce the approximate Bessel beams with
 17 better performance in terms of beam spot invariance and propagation intensity,
 18 such as generation by resonator-generated category or nonlinear wave-mixing
 19 process to constructively produce synthesized Bessel beam [108], as well as a

1 simple transformation by axicons [109]–[111]. This section focuses on the
 2 axicon-created Quasi-Bessel beam. The conceptual diagram of an axicon
 3 transforming a collimated Gaussian beam into a Quasi-Bessel beam is shown in
 4 Figure 12.

5
 6 Figure 12 Schematic of an axicon with base angle α transforming the collimated
 7 Gaussian beam of size 2ω to a Bessel-Gauss beam. The Bessel-Gauss beam can be
 8 considered as formed by wave vectors lying on the cone surface with an apex angle 2θ .

9
 10 Adopting the concept described by David S. Simon [111], that the Bessel-
 11 Gauss beam is defined as the Bessel beam having an electric field E (derived from
 12 the quality $A(x,y)$ in equation (2.4.1.5) and (2.4.1.7)) of the J_n function, as the
 13 equation (2.4.2.3) multiplied by an additional Gaussian factor to decrease the
 14 amplitude away from the origin. The Quasi-Bessel beam formed as the ray trace
 15 diagram in Figure 12 is a Bessel-Gauss beam. In cylindrical coordinates, the ideal
 16 solution of the electric field for the Bessel beam is of the form [27,32]:

$$17 \quad E_n(r, \varphi, z) = \varepsilon e^{ik_z z} J_n(k_r r) e^{\pm i n \varphi}, \quad k = \sqrt{k_z^2 + k_r^2} = 2\pi/\lambda \quad (2.4.2.3)$$

18 The function J_n is the n th-order Bessel function of the first kind, k_z and k_r are the
 19 longitudinal and radial wave vectors, and r , φ and z are the radial, azimuthal and

1 longitudinal components respectively. It is obvious that the resulted Bessel beam
 2 is formed as a set of plane waves propagating on a cone with apex angle θ [109]

$$3 \quad \theta = \tan^{-1} \left(\frac{k_r}{k_z} \right) \quad (2.4.2.4)$$

4 and the central core spot size is defined as

$$5 \quad r_0 = \frac{2.405}{k_r}, \quad k_r = k \sin \theta. \quad (2.4.2.5)$$

6 The spot size, or the FWHM of the central bright lobe of the Bessel-Gauss
 7 beam is inversely proportional to wave vector k_r , which is directly related with
 8 the waves' apex angle θ and the axicon's base angle α , shown by the ray trace
 9 diagram in Figure 12. Thus, the spot size of the axicon created Bessel-Gauss beam
 10 is inversely proportional to the axicon's base angle α . The propagation distance
 11 L_B is given by

$$12 \quad L_B = \frac{\omega}{\tan \left[\sin^{-1}(n_{axicon} \times \sin \alpha) - \alpha \right]} - thickness_{axicon} \quad (2.4.2.6)$$

13 Besides the non-diffractive property that inherits from the equation
 14 derivation, the Bessel beam exhibits another outstanding feature of self-
 15 reconstruction within the propagation distance [110]–[112]. The self-
 16 reconstruction nature of the beam is the capability to recover its initial intensity
 17 profile after disrupting by an object, or a scattering obstacle that locally shifts the
 18 phase or absorbs energy from the incident beam. The self-reconstruction property
 19 has been experimentally demonstrated in various reports and is widely regarded

1 as a consequence of the conical wavefronts of the beam [27,32,34,35]. The
 2 mechanism underlying this phenomenon is that the rays refracted from the
 3 axicon's outer rings that bypass the obstruction begin to enter the axis and
 4 interfere. As they carry energy, the same profile in the beam axis can be restored
 5 as shown in Figure 13.

6
 7 Figure 13 Comparison of ray trace diagrams between the collimated Gaussian beam
 8 and Bessel beam. The beam rays come from left to right. Illuminated objects are shown
 9 in green, while blocked ones are in blue.

10
 11 The collimated Gaussian beam has no way to restore to its original profile
 12 when it is blocked by some objects or scatterers as shown in Figure 13(a). For the
 13 illuminating or other beam-material interaction purpose, the subsequent objects
 14 are no longer to participate, like the blue objects in Figure 13. The Bessel beam
 15 however, can restore to its profile after blocked by the objects and maintain its
 16 profile and interact with the objects within the propagation distance.

1 It has also been shown that Bessel-Gauss beams are able to self-
2 reconstruct even inside inhomogeneous, scattering media [34–36]. This self-
3 healing capability of the Bessel beam is highly advantageous in the high-
4 resolution imaging of thick, scattering micro-organisms or discrete clusters that
5 are difficult to image with a Gaussian illumination beam. However, there are very
6 few quantitative reports on the parameters that affect the self-reconstruction
7 properties of Bessel-Gauss beams. We herein investigate the dependence of self-
8 reconstruction distance on the scatterer’s size, refractive index, and the offset of
9 the scatterer from the beam axis.

10

11 **2.5 Light Sheet Fluorescence Microscopy**

12 This section introduces the concept and reviews the rapid development of the
13 light sheet fluorescence microscopy (LSFM), one form of the optical biological
14 imaging in the last two decades [118]. Since the concept of light sheet was
15 realized in 1993 [14], it has been served as an alternative to confocal microscopy
16 due to several superior features. I would also summarize its advantages, some of
17 the typical implementations, and successful applications.

18

19 **2.5.1 Principle of Light Sheet Fluorescence Microscopy**

20 The principle of the LSFM is that it combines two distinct optical paths, one for

1 illumination with a thin sheet of light, and one for fast wide-field detection that
 2 are orthogonal to each other [118], as shown in Figure 14. The sample is tuned to
 3 locate at the intersection of their focal planes. In contrast to the conventional
 4 optical microscopy, epifluorescence microscopy, or confocal laser scanning
 5 microscopy, LSFM has only a thin layer portion (usually a few hundred
 6 nanometers to a few micrometers) of the sample illuminated that is visible to the
 7 observation. The top view of the light sheet has a waist in the center of the field
 8 of view and overlaps perfectly with the focal plane of the detection objective.

9
 10 Figure 14 The principle of light sheet microscopy. The illumination and the detection
 11 objectives are oriented orthogonally. The sample is placed at the intersection of their
 12 focal planes. A single slice of the sample is illuminated with a thin sheet of laser light.

13

14 In conventional optical microscopy and epifluorescence microscopy, they
 15 have the overlapping optical axes for illumination and detection objectives, which

1 result in poor axial resolutions. In contrast, LSM realizes the optical sectioning
2 techniques for tomographic 3D imaging by separating the excited fluorescence
3 focal plane (formed by the illumination objective) from the detection focal plane
4 (formed by the detection objective) against the out-of-focus background noise
5 signals. As only the actual observed section is illuminated, the good optical
6 sectioning capability reduces the background noise signal and thus creates images
7 with higher contrast, comparable to confocal microscopy. The optical sectioning
8 capability, without the need for any physical sectioning, is the key enabler for this
9 non-invasive 3D imaging technique. This method also reduces the photodamage
10 and stress induced on a living sample [21].

11 The properties of light sheet are of the most crucial importance, such as
12 the thickness, uniformity, and the penetrating depth over the scattering tissue. An
13 ideally thin optical section is the sheet of light illuminated only the focal plane of
14 the detection objective along x-axis in Figure 14. However, there is a trade-off
15 between the thickness of the sheet and the dimension, i.e., the valid field of view,
16 governed by the laws of diffraction. Basically, in the community nowadays, there
17 are two classes of the light sheet generation: static light sheet and scanned light
18 sheet as in Figure 15. The static light sheets are usually produced by a cylindrical
19 lens focusing a laser beam in one dimension [21]. The scanned light sheets are
20 generated by scanning the beam throughout the illuminated layer of sample [23].

Figure 15 Two types of light sheet generation. Static light sheet is directly generated by a cylindrical lens, and scanned light sheet is generated by rapid beam scanning.

The detection system of LSFM is the same as the wide-field microscopy, that records the image across the entire plane within a single exposure time. The lateral resolution of LSFM is equal to that of a wide-field system [118]. Meanwhile, the axial resolution is primarily determined by the light sheet thickness, improved by a factor of 2 to 2.5 over the confocal or wide-field microscopies [118]. This is a great advantage comparing to the confocal laser scanning microscopy, which has overlapping illuminating and detection beam path and has the entire sample instead of a single layer illuminated. The confocal system's scanner has to rush from one pixel to another and only rest for microseconds, hence affects the signal-to-noise ratio (SNR). Also, in confocal, each pixel is multiply scanned depending on the sectioning layers. With the assistance of the fast and sensitive cameras, the LSFM captures images and

1 acquires dataset much faster than other techniques, obtaining superior SNR and
2 minimal phototoxicity and photodamage.

3

4 **2.5.2 Types and Feature Comparison of**

5 **Light Sheet Fluorescence Microscopy**

6 Although Arne H. Voie and his team had deeply researched into the optical
7 sectioning technology [14], [119]–[121], the accelerated development of light
8 sheet fluorescence microscopy has only been launched since the implementation
9 of Selective Plane Imaging Microscopy (SPIM) by Jan Huisken in 2004 [21].
10 Thereafter there have emerged multiple techniques to realize the optical
11 sectioning and imaging in the community. Each investigator or researcher has
12 developed a somewhat different technique and given them different names. This
13 section reviews some of the typical LSFM in order of publication time.

14 **Orthogonal-plane fluorescence optical sectioning OPFOS**

15 The Orthogonal-plane fluorescence optical sectioning (OPFOS) was developed
16 by Arne H. Voie et al in 1993 [14]. Voie, David Burns, and Francis Spelman
17 developed the OPFOS instrument with the side illumination to optically section
18 for the first time, adopting the concept from side illumination photomicrography
19 [122], [123]. OPFOS features all the elements that are presented in the current
20 LSFM, which are the laser source, light sheet generation lenses, orthogonal or

1 side illumination of specimen, fluorescent optical sectioning, and z-stack motion
 2 of specimen for reconstruction of 3D image. The illumination beam was focused
 3 by the cylindrical lens into a plane, which is co-planar with the depth of field of
 4 the detector objective as shown in Figure 16. The lateral and axial resolution
 5 reached $10\text{ }\mu\text{m}$ and $26\text{ }\mu\text{m}$ respectively within 1.5 mm field of view. The similar
 6 light sheet generation implementation with cylindrical lens was also
 7 demonstrated by Eran Fuchs et al., in 2002 [124], in where they termed their
 8 device as Thin laser light sheet microscope (TLSM), and used it to detect the fast-
 9 motion of living marine bacteria.

10

11 Figure 16 Schematic diagram of optical components assemblies for OPFOS [14]

12

13 **Selective plane imaging microscopy SPIM**

14 Jan Huiskens et al. have demonstrated their developed selective plane imaging
 15 microscopy (SPIM) since 2004 [21], and they have updated the system to a multi-
 16 directional SPIM (mSPIM) in 2007 [125]. Each light sheet in the two beam paths
 17 was generated by pivoting a cylindrical lens and an objective lens. The two well-

1 corrected beams from opposite directions produced the light sheet with minimal
 2 scattering and intensity attenuation by dense live tissue across the specimen. The
 3 axial resolution was increased by $\sqrt{2}$ compared to the single-directional SPIM.

4
 5 Figure 17 Schematic diagram of mSPIM [125]. (a) Parallel illumination of the sample in
 6 SPIM results in shadows and attenuation across the FOV. (c) In mSPIM, these artifacts
 7 are reduced by illuminating the sample from two sides with a thinner pivoting light sheet.
 8 IL: illumination, DL: detection lens, S: sample.

9

10 **High resolution Orthogonal plane fluorescence optical** 11 **sectioning HROPFOS**

12 Jan Buytaert and Joris Dirckx, in 2007, developed the high-resolution Orthogonal
 13 plane fluorescence optical sectioning (HROPFOS) technique [126] based on

1 PPFOS. It greatly improved the lateral resolution from tens of micron to 1 μm ,
 2 and axial resolution from 26 μm to 2 μm , and demonstrated the reconstruction of
 3 the 3D model that is comparable to SEM image. The setup is shown in the Figure
 4 18.

5

6 Figure 18 Schematic diagram of the HROPFOS setup [126].

7

8 The laser light coming along x-axis passes through a Galilean beam
 9 expander (BE) with spatial filter, a vertical field stop (FS), and a cylindrical
 10 achromat (CL) that focuses the laser along one dimension within the transparent
 11 and fluorescent object (O). A two-axis motorized object translation stage (OTS)
 12 allows scanning of the specimen. CCD camera equipped with objective lens and
 13 color filter captures the fluorescence light emitted by the object. The focusing

translation stage (FTS) is used to coincide the objective lens focal plane with the illumination plane. The HROPFOS no longer illuminates the full plane of object and captures the image at one shot. It scans the object through the line of best focus, created by a large aperture cylindrical lens, and reconstructs the full image of the object. The resolutions are therefore greatly improved to 1 and 2 μm for lateral and axial axes.

Table 2 Types and features of LSFM below summarizes the type and features of different LSFM for reference since the optical sectioning technique was published in 1993. Herein includes those techniques have not been listed explicitly in this Chapter.

Table 2 Types and features of LSFM

Name/ Acronym	Light sheet generation	Illumination direction	Lateral/axial Resolution	Reference, Year
OPFOS	Cylindrical lens	Single	10 μm /26 μm	[14], 1993
TSLM	Cylindrical lens	Single	<3 m/23 μm	[124], 2002
mSPIM	Pivoting Cylindrical lens	Dual	6 μm /6 μm	[125], 2007
HROPFOS	Cylindrical lens	Single	1 μm /2 μm	[126], 2007
OCPI	Cylindrical or gradient index lens	Single	<28 μm , depend on fast detection/ 3-5 μm	[127], 2008
DSLIM	f-theta lens	Dual	300 nm/1000 nm	[23][128], 2008

Basically, all types of the LSFM consist of two sets of beam path instruments, one for illumination and the other for detection. Alignment of the

1 beam path and configuration of the instruments greatly affect the performance
2 and the flexibility of the specimen selection and imaging.

3

4 **2.6 Diffractive Lens's Transformation on Beam**

5 This section reviews the diffractive lens, which is also called the amplitude
6 Fresnel lens development, that shows some outstanding features and serves as a
7 good substitute to the refractive lens in some applications. We deeply review the
8 interferometric sensing technologies of the tilt angle of an incident beam, and
9 explore the possibility of replacing the interferometric system with thin-film lens
10 for better and more flexible application.

11

12 **2.6.1 Design of Fresnel Zone Plate and Fresnel Axicon**

13 A Fresnel lens is a type of composite compact lens developed by the French
14 physicist Augustin-Jean Fresnel. It is also named as Fresnel Zone Plate (FZP) in
15 his honor. The Fresnel lens uses diffraction instead of refraction or reflection to
16 manipulate the light beams. It has been called "the invention that saved a million
17 ships. For the lenses made of isotropic material, the modification of light happens
18 at the interface of media instead of the interior of lens. The Fresnel design allows
19 manufacturing of lenses of large aperture and short focal length with dramatically
20 reduced material.

21 Adopting the design concept from Kenro Miyamoto's report [129], we

1 simplify the design of the Fresnel lens as shown in Figure 19. The height of a
 2 spherical or aspherical lens is divided into slices with same thickness t , which is
 3 in order of wavelength as the green lines in Figure 19(b). In each divided zone,
 4 defined as the Fresnel zone, the height of lens is compressed by integer times of
 5 t with the same surface geometry as in Figure 19(c).

6
 7 Figure 19 Design of Fresnel lens. (a) Original refractive lens. (b) Division of the Fresnel
 8 zone by thickness t . (c) Fresnel lens. (d, e) Binary FZP. (f, g) Multi-level FZP. The red
 9 lines in (d,f) represent the geometry of the Fresnel lens, and black lines represent the
 10 profile of phase FZP. The different colors in amplitude FZP in (e, g) represent different
 11 material or dense giving different refractive indexes.

12
 13 To further simplify the planar zone plate design, one can digitize the
 14 surface pattern to multi-level, usually as binary pattern, as shown in Figure 19(d-
 15 g). One type of the FZP, termed as phase FZP, is fabricated as the alternate
 16 concentric annular grooves are cut in a single piece of optics [130] as Figure 19(d,
 17 f). Alternatively, for the amplitude FZP as shown in Figure 19(e, g), different

1 materials or a material with different composite dense and refractive index can
2 be used to make up alternate concentric rings [130], [131]. Since the amplitude
3 FZP use the diffraction instead of the refraction at the lens surface to manipulate
4 the light beams, it includes much complicated calculations to produce the FZP
5 that fulfills the beam transformation requirement.

6 As discussed in Section 2.4, the axicon is well-known to generate the
7 Quasi-Bessel beam that possesses outstanding features. We are able to produce
8 the axicon with the Fresnel lens design and compress it to thin film morphology.
9 There have been reports that demonstrates the design and fabrication of the
10 Fresnel axicon [132], [133]. Y. Kizuka and M. Yamauchi refer to their optics as
11 diffractive optical elements (DOE) in their work [133] and so the specially
12 designed DOE that has alternate ring pattern with two materials/transparenties
13 and gives the analogous function to the refractive axicon is named as binary
14 diffractive axicon. We use the term “diffractive axicon” throughout our work in
15 this thesis. The diffractive axicon which is analogous to the refractive axicon has
16 a common design that it is made up of concentric rings with equal widths and
17 pitches, where the ring width and pitch depend on the incident wavelength and
18 the analogous axicon’s refractive power.

19

20 **2.6.2 Interferometric Sensing of Tilted Angle**

21 In optical field, the alignment or the precise manipulation of light beam are of

most crucial importance. It is also extended to measure the mechanical motion of an object. Thus, precise measurement of the tilted angle between beams or surface with flexible or portable devices are drawing attention in the community. To the best of my knowledge, there are several reports [25], [134], that typically use the Mach-Zehnder or Sagnac interferometric techniques that constructed with two beam paths for the tilted angle identification or generation.

S.P. Walborn et al., have shown the several schemes that measure the tilted angles of a beam or an object's mechanical motion as shown in Figure 20.

Figure 20 Schematics of sensing the object M's tilted angle. (a) Basic spatial displacement scheme "beam deflection" technique. (b) Mach-Zehnder Interferometric technique in which the object is placed inside the interferometer that is constructed with a polarizing beam splitter (PBS). (c) Sagnac interferometric method, in which is constructed with a PBS. The counterpropagating horizontal and vertical polarization components are displaced in opposite transverse directions. Information about θ can be obtained via spatial measurements, polarization measurements, or both.

1 Figure 20(a) shows the direct measurement as the “beam deflection”
2 technique. We can describe that the beam is incident on the reflecting object M
3 located at origin, and assume the deflection happens only in x -direction. It allows
4 to ignore the variation in y -direction. When M is tilted by an angle θ , the beam
5 suffers an angular deflection of 2θ . The deflection 2θ can be given by the relation
6 of the distance D between M and detector, and the beam displacement d measured
7 by detector as $d/D = \tan 2\theta$. Figure 20(b) shows the Mach-Zehnder interferometer
8 that interact with the two beam paths, which are associated with horizontal H and
9 vertical V polarization directions. Only one beam path probes the object M.
10 comparing to the Sagnac scheme in Figure 20(c), where both the beam paths of
11 polarizations probe the object M. The information of beam deflection and thus
12 motion of M are given by the polarization states of the resulted beams and the
13 variance and position of the input beam. We notice that the configuration of the
14 Sagnac interferometer is larger than that of the Mach-Zehnder system. However,
15 Sagnac interferometer obtains higher precision, since it allows two beam paths
16 probe the tilted angle θ of M, and gives larger detectable variance and ratio of
17 resulted beam parameters.

18

Chapter 3

Establishment of Femtosecond Laser Additive Manufacturing System and Fabrication of Micro-Optics

3.1 Overview of Femtosecond Laser Additive Manufacturing System

The Additive manufacturing (AM) technology was developed since the 1990s to customizes and tools the direct parts manufacturing with complex geometry. The product dimensions vary from as large as concrete buildings to as compact as sub-micron optoelectronic devices. In this research, we seek the additive manufacturing technology which can produce the customized free-shaped optical devices with the sub-micron precision and resolution. Owing to the two-photon phenomena induced by the femtosecond laser and the nano-positioning piezo-stage, the femtosecond laser additive manufacturing technology is proven to be capable of fabricating the target-oriented optical devices with transparent polymeric material, flexible graphene-oxide/reduced graphene-oxide film, and other various optical devices.

In this chapter, I set up the femtosecond laser additive manufacturing systems to fabricate the pre-designed optical devices. The femtosecond laser direct writing (FsLDW) system is used to fabricate the 3D micro-lens with the

transparent epoxy resin, such as SU-8, PDMS, and GelMA. The high-speed femtosecond laser scanning system is used to fabricate the flexible optical/optoelectronic film structures with graphene-oxide/reduced graphene-oxide.

5

3.2 Fabrication of SU-8 3D Micro-Structures by Femtosecond Laser Direct Writing System

3.2.1 Setup of Femtosecond laser direct writing system

The femtosecond laser direct writing (FsLDW) system mainly consist of the femtosecond laser, the beam path guiding mirrors and focusing objective, nano-positioning stage, and the real-time monitoring CCD camera. The system is set up as shown in the Figure 21.

13

Figure 21 Schematic diagram of 3D printing system configuration

14

1 The second harmonic from an Er-doped fiber femtosecond laser
2 (TOPTICA FemtoFiber pro-NIR, 780 nm, 100 fs, 80 MHz) was used as the light
3 source for the FsLDW process. The laser beam passes through the half-wave
4 plate (HWP), polarized beam splitter (PBS) and beam splitter (BS) that are used
5 to control the laser power, and reflects by the Broadband Dielectric Mirrors (M,
6 Thorlabs BB1-E02-Ø1") that are used to direct the beam. The laser beam is then
7 focused by 60× objective lens (O, Olympus LUCPLFLN60X, dry, NA = 0.7) into
8 the material layer applied on a coverslip and carried on the 3-axis PZT stage. The
9 nano-positioning 3-axis PZT stage (PI nano® TM series, with open-window
10 sample holder) with the sample, moves relative to the laser beam focus following
11 the designed routines. The routine of the movements is pre-designed and
12 controlled by the LabVIEW program shown in section “Nano-Positioning Stage
13 and Control program”. In this manner, the initial material in gel form is
14 polymerized and solidified at the scanned voxel and fabricate the designed
15 structures.

16 The laser beam of 1.2 mm beam waist is focused by the 60× objective
17 lens to $2.15 \pm 0.16 \mu\text{m}$ (FWHM) in the photoresist SU-8. The beam power can be
18 adjusted by the polarized beam splitter (PBS) and beam splitter (BS), which can
19 be monitored by the power monitor (PM). Beam damper (BD) is set to absorb the
20 reflected beams for safety reason. The polymerized voxel theoretically reaches
21 smaller dimension than the focused spot size, i.e, to the sub-micron scale. The 3-

1 axis PZT stage, carrying the sample, moves at the computer-aided design three-
 2 dimensional routine with respect to the laser beam focus. In this manner, the
 3 scanned part of the initial material in liquid form is polymerized and solidified at
 4 the scanned voxel, and fabricate the designed structures as shown in Figure 22.

5
 6 Figure 22 Schematic diagram of 3D printing microstructure by the femtosecond laser
 7 spot scanning the photoresist with pre-designed routine

8
 9 The printing process can be monitored by the CCD camera in real time,
 10 with the illumination of LED white light from the bottom. It is shown in the
 11 section “Real-Time Monitoring System” in the following description.

12

1 TOPTICA FemtoFiber pro NIR laser

2 The TOPTICA FemtoFiber pro NIR laser is an Erbium-doped fiber laser,
3 providing femtosecond laser pulse of switchable wavelengths of 1560 nm and
4 780 nm, and maximum power over 300 mW. In the research, wavelength 780 nm
5 is selected as it induces two-photon polymerization in polymeric materials,
6 equivalent to excite a polymeric molecule with a photon of 390 nm wavelength.
7 A plenty of photoresists, including the SU-8, are sensitive to the UV exposure, so
8 that provide a flexible option of materials for different applications. The spectral
9 bandwidth is typically 10 to 15 nm and the auto-correlation shows a pulse
10 duration less than 100 fs.

11
12 Figure 23 FemtoFiber pro NIR laser and its parameters. (a) laser source appearance;
13 (b) internal setup; (c) laser pulse characteristics centered at 780 nm, the
14 autocorrelation pulse width is less than 100 fs; (d) linear emission spectrum.

1 Relevant specializations of the TOPTICA FemtoFiber pro NIR laser with
 2 operating wavelength at 780 nm are summarized in the table below:

3 Table 3 FemtoFiber pro NIR specifications

Key Specifications	Value
Second-harmonic wavelength	780
Laser output power	> 140 mW
Pulse width	< 100 fs
Repetition rate	80 MHz standard
Linear polarization	> 95 % (horizontal)
Beam shape	TEM ₀₀ , M ² < 1.2
Beam size	Typical Ø 1.2 mm
Beam divergence	< 1 mrad

4

5 **Focusing Objective**

6 The Nikon 60× objective lens (O, Olympus LUCPLFLN60X, dry, NA =
 7 0.7) is used to focus the femtosecond laser spot for the polymerization in SU-8.
 8 As shown in the article [135], the 60× objective lens tightly focuses the fs laser
 9 beam to a lateral focal spot size (one-photon spot size) of $2.15 \pm 0.16 \mu\text{m}$
 10 (FWHM), and is capable of producing a single polymerized line width of $1.4 \pm$
 11 $0.15 \mu\text{m}$. The theoretical one-photon axial resolution of the system is calculated
 12 to be $3.18 \mu\text{m}$ in air.

13

14 **Nano-positioning Stage and Control program**

15 The nano-positioning 3-axis piezoelectric stage stage (PI nano® TM

1 series, with open-window sample holder) carrying the materials coated on the
2 coverslip, moves in three dimensions relative to the focusing point of the laser
3 which is stationary and so that forms the 3D structures. The movement of the
4 stage is controlled and monitored by the LabVIEW program.

5 Although the above-mentioned axial resolution is as large as $3.18\text{ }\mu\text{m}$, the
6 feature size of the micro-fabrication mainly depends on the motion resolution of
7 the stage. The piezoelectric scanning stage is optimized for high-resolution
8 microscopes. It features a very low profile of 20 mm for easy integration with
9 devices, a large aperture with the accessory and holder to stable the sample, and
10 a long travel ranges in three-axes of up to $200\text{ }\mu\text{m} \times 200\text{ }\mu\text{m} \times 200\text{ }\mu\text{m}$, employs
11 a step resolution of less than 1 nm with sub-nanometer closed-loop resolution,
12 that is ideal for the imaging, machining, and microscopy applications.

13
14 Figure 24 E-545 PI-nano® Piezo Controller and Multi-Axis Positioner.

1 The stage can deliver very fast and highly accurate motion with the nano-
2 scale resolution. The motion is controlled by the software provided with the
3 connecting ports and control module including the speed, acceleration and stop
4 functions. The LabVIEW program is used to design the printing routine and
5 control the stage motion, as shown in the panel in Figure 25.

Figure 25 The LabVIEW control panel

The control software made in LabVIEW mainly consists of four parts:

- 1) System default set;
- 2) Real-time monitoring system: parameters reflect the real-time position of the printing platform;
- 3) Parameter control: it basically controls the moving velocities in three dimensions and layered step size and heights in the z-direction of the printing

1 process;

2 4) Design panel: there are several designed patterns can be selected to fabricate
3 with the materials, including arbitrary 2D quadrilaterals, circles, cylinders, and
4 cones. More complex geometric patterns will be implemented into the system.
5 The pattern simulations are shown in the right panel.

6

7 **Real-Time Monitoring System**

8 The printing process can be monitored by the Thorlabs DCC1645C
9 camera in real time, with the illumination of LED light, and shown by software
10 ThorCam. Thorlabs DCC1645C camera is a CMOS color sensor that can cover
11 the exact sensitive area of $4.61\text{ mm} \times 3.69\text{ mm}$, containing a resolution of 1280
12 $\times 1024$ pixels that cover the printing region, with a square pixel size of $3.6\text{ }\mu\text{m}$,
13 and a frame rate of 24.9 fps, so that it is able to capture the real-time condition of
14 the sample during 3D printing. The real-time monitoring panel when observing
15 the printing of a cylinder with a diameter of $60\text{ }\mu\text{m}$ and height of $20\text{ }\mu\text{m}$ is shown
16 as in Figure 26. The printed cylinder can be seen clearly after the femtosecond
17 laser spot scans the two base layers.

Figure 26 Monitoring panel in ThorCam during the printing process. The circle in the center is the printed cylinder pattern.

3.2.2 Parametric Condition and Optimization of SU-8 Micro-Structure Fabrication

The process of fabricating the SU-8 structure goes through the coating of material, soft-baking, laser exposure, post-baking, and development. For different materials, the soft-baking and post-baking steps are optional depending on the composite of the photoresists, including the cross-linker, photo-initiator, and doping particles etc.

1 **Fabrication procedure**

2 The procedure of the femtosecond laser direct writing of the SU-8 micro-
3 structures is shown in the flow chart in Figure 27.

4

5 Figure 27 Flow chart of the SU-8 micro-structure fabrication

6

7 The fabrication starts with smearing a thin layer/coating over a clean and
8 oxygen plasma treated glass coverslip with thickness depending on the designed
9 structures, usually of 50 to 100 μm . For more evenly applying the material, the
10 spin-coating is required. The raw SU-8 is viscous liquid and the spin-coating
11 should be handled carefully. For the case that builds the micro-structures on
12 specified substrates, such as facet of optical fibers, spin-coating is not applicable,
13 but the drop casting of material is sufficient for the fabrication in this work. The
14 specification recommends that the sample is spun at 1500 to 3000 rpm for 30

seconds with the acceleration of 300 rpm/second to prepare the sample of the thickness of 50 to 100 μm . The sample is thereafter heated gradually to 95°C by 5°C/min from room temperature and kept soft-baking at 95°C for 30 min. The prepared sample (after naturally cooled down) is then carried by the 3-axis nano-positioning stage, moves relative to the femtosecond laser spot of 780 nm wavelength, scanning along the pre-designed routine and inducing the two-photon polymerization process to form the designed shape layer by layer. The nano-positioning platform, whose movement resolution reaches as high as nanometric, carrying the sample moves along the designed routine relatively to the laser focal point of the diameter of around 1 μm . The scanned structure is then post-baked at 65 °C for 1 min and then 95 °C, the time of which depends on the designed thickness of the patterns. The SU-8 structure is finally developed by SU-8 developer (MicroChem® 1-methoxy- 2-propanol acetate) and rinsed off the uncured rest with IPA.

The specific printing parameters such as baking time and temperature, laser exposure power and scanning speed, and development duration are of the most importance, so that controlled and tested as shown in the following section.

Parametric testing

The parametric testing for SU-8 fabrication has been carried out.

1 Fabrication with other materials has also succeeded with the femtosecond laser
2 direct writing system and can be applied to the specific fields. To fabricate the
3 ultra-precision micro-lens or other optical structures, the resolution and stability
4 of the fabricated structures are the most important factors. Therefore, the
5 parametric testing is carried out to settle the optimal printing condition. This
6 section focuses on the SU-8 micro-structures.

7 The parameters that come to our attention by intuition and during
8 experiments have major effects on the final structure of printed patterns:

9 1) Laser power intensity: the laser beam coming out initially with beam size 1.2
10 mm is focused to a beam waist of $2.15 \pm 0.16 \mu\text{m}$ by the 60×objective lens
11 (Olympus LUCPLFLN60X, dry, NA = 0.7) in the raw SU-8. The theoretical
12 axial resolution of the system is calculated to be $3.18 \mu\text{m}$ in air. The typical
13 laser power range to fabricate the microstructure in this research is
14 determined as 5 to 15 mW. The intensity at the focal spot inside the material
15 is then calculated to be 1.38×10^{-9} to $4.13 \times 10^{-9} \text{ W/m}^2$. It is capable of
16 producing a single polymerized line width of $1.4 \pm 0.15 \mu\text{m}$. The Lower the
17 laser power, the thinner the single line is fabricated by scanning, and
18 therefore, the structure with higher precision and resolution is formed.
19 However, there is a trade-off between the laser power and exposure time.
20 Lower power requires a longer time to ensure sufficient photons absorbed by
21 the material, which sometimes leads to produce a solidified background.

1 Besides, the absorption rate of the photon and induced two-photon
2 polymerization efficiency are not directly proportional to the laser power.
3 Instead, it is related with the material's inherent properties molecular
4 arrangement, including the polymer cross-section area.

5 2) Scanning speed: scanning speed is negatively related to the laser power for
6 same cross-linking rate. The proper speed at the power of 7 mW is
7 determined to be in the range of 10 to 20 $\mu\text{m/s}$.

8 3) Repeated scanning layer: the repeated scanning layer does matter to the
9 patterns' stability and dimension, especially for the single line patterns such
10 as waveguides.

11 4) SU-8 film thickness: the thickness of the manual coating of SU-8 sample is
12 around 50 μm , depending on the height of designed structures, and less than
13 100 μm . Since the designed patterns in this research are within the dimension
14 of 30 μm , coating thickness of 50 μm is sufficient for the fabrication.

15 5) Step size, the interval between scanning lines to fabricate solid structures:
16 calibration work shows that the single scanned line width with laser power
17 of 7 mW and scanning speed of 20 $\mu\text{m/s}$ is $1.4 \pm 0.15 \mu\text{m}$ in lateral resolution.
18 We control the line width by tuning the laser power. Laser power of 15 mW
19 and same scanning speed give line width up to 4.3 μm . Theoretical axial
20 resolution of the femtosecond laser spot is calculated to be 3.18 μm in air.

1 However, the axial resolution of fabrication is mainly determined by the
2 designed step size that controlled by the 3-axis nano-positioning stage, which
3 reaches a step resolution of < 1 nm. In this research, both the lateral and axial
4 step sizes are set to $1\text{ }\mu\text{m}$.

5 6) Temperature and duration of soft-baking and post-baking: soft-baking makes
6 a great effect on the sensitivity of the material to absorb the photons and form
7 cross-links. The SU-8 film with thickness of $50\text{ }\mu\text{m}$ is soft baked on hot plate
8 or oven with gradually increase of temperature to 95°C by $5^{\circ}\text{C}/\text{min}$ and kept
9 heated at 95°C for 30 min, and thereafter cooled down before laser scanning.
10 Post-baking assists to solidify the cross-links and thus the scanned geometry.
11 Longer post-baking at higher temperature is required for thick patterns or
12 structures. Structures of 20 to $50\text{ }\mu\text{m}$ heights should be post-baked at 65°C
13 for 1 min and then 95°C for at least 5 min. There is also a report that claims
14 no post bake is needed [6], depends on the application and desired
15 polymerization degree.

16 7) Development time: the polymerization is finalized by the development by
17 SU-8 developer (MicroChem®, 1-methoxy-2-propanol acetate) to reveal the
18 solid and stable patterns. We observed that a minimum 6 min is required to
19 immerse the structures in developer solution to fully develop the structure of
20 $20\text{ }\mu\text{m}$ height and dissolve the unexposed material with IPA. For certain
21 application that the unexposed SU-8 serves as the cladding of the

polymerized part, the development is not necessary.

3.3 Fabrication of Reduced Graphene-Oxide Thin Film Structures by High-Speed Femtosecond Laser Scanning System

Optical elements control and modify the propagation of light by refraction, reflection, diffraction, or resonance effect, by the refractive, reflective, diffractive optics, and hybrid optics respectively. As discussed in Chapter 3.2, the femtosecond laser direct writing on SU-8 coated layer has been proven to well realize the 3D fabrication of the refractive micro-optics. In some application, I am also seeking for the fabrication method for some rapid two-dimensional planar fabrication with various materials. The designing and manufacturing of ultra-thin diffractive binary Fresnel zone plates (FZPs) are placed great hope for the realization of next-generation hybrid optics which creatively integrate the refractive, reflective, and diffractive elements, but have remained less searched to date.

In this part, we are exploring the femtosecond laser method to fabricate the thin-film binary diffractive lenses by photoreduction of reduced graphene oxides (rGO). In recent years, rGO have attracted great attention due to that they have advantages for large-scale production with the unique characteristic of thermal, electrical, and mechanical properties similar to pristine graphene [136].

1 Recent researches have reported that rGO structures can be fabricated by
2 photoreduction or fast thermal quenching, and but still have limitation of the
3 photo-mask design and optical phase delay [131], [137]. To achieve non-mask
4 rapid fabrication with the sub-micron feature size, we set up the femtosecond
5 laser direct scanning (FsLDS) system to print the rGO diffractive lens via two-
6 photon photoreduction. It can be a promising solution candidate to fulfill all these
7 requirements as the direct laser patterning can be realized with lower cost in a
8 time-efficient manner circumventing the drawbacks such as the necessity of
9 pattern mask, vacuum environment, thermal process, and toxic chemicals.

10

11 **3.3.1 Setup of High-Speed Femtosecond Laser**

12 **Scanning System**

13 The femtosecond laser direct scanning (FsLDS) system is established to fabricate
14 thin planar micro-structures and micro-sintering with larger dimension. The
15 proposed technique enables the direct patterning of FZP with multiple materials
16 on a wide option of flexible and stretchable substrates. The setup consists of the
17 femtosecond laser, the light path guiding mirrors and the Galvano scanner, f-theta
18 lens, and sample stage. The Schematic diagram of the FsLDS setup is shown in
19 Figure 28.

Figure 28 Schematic figure for the FsLDS system based on a Galvano scanner

The second harmonic from the Yb-doped femtosecond laser was used as the light source for the FsLDS process. The laser beam pass through the beam expander, and is directed by the guiding mirrors to the Galvano scanner GM, and focused by the f-theta lens onto the sample plate installed on the steady stage.

The Yb-doped fiber femtosecond laser (Amplitude Systèmes, Satsuma HP, 515 nm, 220 fs, 500kHz) is selected as the laser source in the system. It can provide the high-power laser beam of wavelength 1030 nm, which can generate the targeted laser beam of 515 nm by second harmonic. It can provide an average power over 10W and reach the energy larger than 20 μ J per pulse.

Figure 29 Appearance of Yb-doped Femtosecond laser Satsuma HP

Table 4 Femtosecond laser Satsuma HP specifications

Key Specifications	Value
Second-harmonic wavelength	515
Average power	> 10 W
Pulse width	< 350 fs to > 10 ps
Energy per pulse	> 20 μ J
Repetition rate	< 40 MHz
Beam shape	$M^2 < 1.2$
Beam size	Typical $\varnothing$ 1.2 mm
Beam divergence	< 25 μ rad/ $^{\circ}$ C

The laser beam is directed through the digital scanning device consisting of a Galvano scanner coupled with an f-theta lens. The Sino® Galvano scanner contains two galvo-scan mirrors that deflect an incident laser beam in x- and y- axes individually. Each mirror has a motor scan angle of $\pm 7.5^{\circ}$, corresponding to a maximum optical scan range of $\pm 15^{\circ}$. It is able to rotate the beam within the displacement of 13.4 mm, having an angle resolution θ' of 12 μ rad. In this work, the valid scanning area is restricted to a $6 \times 6 \text{ mm}^2$ window.

The f-theta lens is commonly used in combination with Galvano scanners or polygon scanners in various industrial processing fields and applications from

1 laser machining, synthetic materials, surface cutting. confocal microscopy, and
2 biomedical research. The standard objective lenses focus the laser beam on a
3 spherical surface instead of an ideal flat or plane field, which makes the resolution
4 of micro-patterning good enough at the central regime and aberrated at the outer
5 area. In contrast, the use of f-theta lenses provides a plane focusing surface and
6 almost constant spot size over the entire scanning field. The position of the spot
7 on the image plane is directly proportional to the scan angle. The f-theta lens (SL-
8 100-F160-532, Suzhou Xuhong Photonics Technology Co. Ltd.,) provides the
9 focal length $f = 160$ mm along the normal axial direction at the wavelength of 532
10 nm. The scanning resolution d' is therefore can be calculated as $d' = f \times \theta' =$
11 $0.19 \mu\text{m}$.

12
13 Figure 30 (left) Sino Galvano scanner; (right) f-theta lens. The two devices are coupled
14 to direct the laser beam focus over a plane at distance of 160 mm to the f-theta lens

15
16 The MarkingMate program is a window-based laser marking software
17 developed to design the scanning routine and control the power level. It is a

1 powerful tool that helps to design the thin-film singular or multiple patterns
2 within the $6 \times 6 \text{ mm}^2$ window and control the power level at each part of the
3 patterns.

4

5 **3.3.2 Parametric Condition of Reduced Graphene-Oxide** 6 **Thin Film Structures**

7 In this part, I employ the femtosecond laser direct scanning (FsLDS) system to
8 fabricate the thin-film binary diffractive lenses with reduced graphene oxides
9 (rGO). This section summarize the in-depth study on the photothermal/
10 photoreduction effect, which include the reduction degree and the patterning
11 linewidth with various laser parameters such as laser intensity and pulse
12 repetition rate.

13 The graphene oxide suspended aqueous solution (Product number:
14 777676, Sigma Aldrich) is formulated as 0.5 mg/ml concentration for fabricating
15 thin flim of $\sim 500 \text{ nm}$ thickness. The photoreduction of rGO using the Yb-doped
16 fiber laser (Amplitude Systèmes, Satsuma HP) with a central wavelength of 515
17 nm, a pulse duration of 230 fs and tunable repetition rates up to 2 MHz. The
18 focused laser spot scans on the GO samples with a scaning speed of 100 mm/s
19 and a spot size of $14 \text{ }\mu\text{m}$ using a galvano scanner coupled with the f-theta lens.
20 Photoreduction were conducted in varies repetition rate (31.25, 62.5, 125, 250
21 and 500 kHz) and average power (15-120 mW).

Three different patterning regimes on rGO thin film are realized by controlling the repetition rate and the power level of femtosecond laser: the first is phase-changing less-thermal photoreduction (LTPR) regime, the second is transmittance-changing thermal photoreduction regime (TPR), and the third is ablation (ABL) regime as shown in Figure 31. The three regimes of rGO products give different transmittance and thickness that can be manipulated to fabricate the arbitrarily designed Fresnel zone plate, including the proposed diffractive axicon in this research.

Figure 31 Femtosecond laser direct scanning (FsLDS) of the ultra-thin rGO FZPs by photoreduction in three regimes. (a) The three regimes are fabricated by the femtosecond laser tuned at different repetition rates and pulse energy levels. (b-d) Transmission microscope images of the patterned ultra-thin rGO FZPs under the three regimes respectively, LTPR, TPR, and ABL.

The ring patterns provide different transmittance to the incident light in the optical applications, so that generate the diffraction to the transmitting light.

1 In this research using the diffractive axicon to generate the Quasi-Bessel beams
2 for tilted angle detection (Chapter 5), the alternate rings of diffractive axicon are
3 fabricated at thermal photoreduction (TPR) and ablation (ABL) regimes.
4

Chapter 4

Light Sheet Microscopy with Micro-Axicon Created Bessel Beam

In this chapter, I demonstrate the proposed design of the light sheet fluorescence microscopy using the micro-axicon created non-diffractive Bessel-Gauss Beam. This chapter presents the results in the order: schematic design of the light sheet fluorescence microscopy with micro-axicon and the design of the axicon, the fabrication of the axicon and its optical performance test, the performance comparison with the simulation results; and the realization of the light sheet fluorescence microscopy to image the polystyrene beads and live Hela cells.

4.1 Optical Layout of Light Sheet Fluorescence Microscopy

Light sheet fluorescence microscopy (LSFM) realizes the high-resolution fluorescence imaging of live biological specimens by selective layered illumination of the sample in the focal plane of the detection objective, as shown in as Figure 32(a). Conventional LSFM produces light sheet using a cylindrical lens before the illumination objective. Figure 32(a) shows the illumination and detection objectives installed in conventional LSFM, illustrating the restricted system flexibility and sample installation. In the micro-axicon based LSFM scheme as shown in Figure 32(b), the illumination objective is replaced by a two-

1 photon-polymerized micro-axicon attached onto the optical fiber's end facet. The
 2 micro-axicon readily generates a non-diffractive pencil-like Bessel-Gauss beam
 3 from the input Gaussian beam, and hence eliminates the bulky illumination
 4 objective system from Figure 32(a), simplifying the sample installation. The
 5 Bessel-Gauss beam generated by the micro-axicon excites the target objects over
 6 a large field-of-view in a fluorescent environment. By scanning the beam, 3D
 7 stacked images of the target objects can be collected (Figure 32(c)). This method
 8 reduces background signals from different depths and hence the detection system
 9 can selectively capture the optically sectioned images to reconstruct high-contrast
 10 3D images.

11
 12 Figure 32 Light sheet fluorescence microscopy scheme with Bessel-Gauss beam
 13 created by the two-photon-polymerized micro-axicon. (a) Configuration of conventional
 14 LSFM consisting of illumination and detection lenses in orthogonal axes showing the
 15 physical restrictions of the scheme. (b-d) LSFM with micro-axicon: (b) Configuration of
 16 the LSFM utilizing Bessel-Gauss beam created by the micro-axicon from fiber-carried
 17 Gaussian input; (c) schematic of the micro-axicon based Bessel-Gauss beam scanning
 18 for optical sectioning of the cell; (d) optical microscope and SEM images of the fabricated
 19 two-photon polymerized micro-axicon.

1 The micro-axicon was designed to have the base diameter of 100 μm and
2 base angle α of 20° , and was additively manufactured by FsLDW with epoxy
3 resin, SU-8. The dimensions of the micro-axicon was carefully chosen to
4 fabricate it accurately on an optical fiber's end facet. The lens diameter of 100
5 μm provides the pencil-like Bessel beam a propagation distance of $\sim 160 \mu\text{m}$,
6 while the base angle 20° results in a spot size of $\sim 2.3 \mu\text{m}$. The second-harmonic
7 from an Er-doped fiber femtosecond laser (TOPTICA FemtoFiber pro NIR, 780
8 nm, 100 fs, 80 MHz) is used for the two-photon polymerization as shown in
9 Chapter 3.2. The optical microscope and scanning electron microscope (SEM)
10 images of the fabricated micro-axicon are shown in Figure 32(d). The polished
11 optical fiber's end facet or other conventional substrates (on which uncured SU-
12 8 was coated) were 3D scanned in the pre-designed routes with the aid of nano-
13 positioning stages (PI nanoTM series) over $100 \times 100 \times 18 \mu\text{m}^3$ to fabricate the
14 micro-axicon.

15 The conceptual diagram and the collected colormap of the FsLDW
16 fabricated micro-axicon of base angle 20° transforming a Gaussian beam into a
17 Bessel-Gauss beam is shown in the Figure 33.

1

2 Figure 33 (top) The schematic representation of the axicon transforming a Gaussian
 3 beam into a Bessel-Gauss beam. (bottom) Illustration of the self-reconstruction property
 4 of the micro-axicon based Bessel-Gauss beam

5

6 The incoming Gaussian beam is refracted toward the central axis and the
 7 interference generates the Bessel-Gauss beam. The axicon transformation can be
 8 understood based on the Helmholtz equation, which provides the universal
 9 solutions to describe the propagation of electromagnetic waves. In addition to
 10 Gaussian beams and parabolic waves, the Bessel-type wave has been introduced
 11 as a class of diffraction-free solutions to the Helmholtz equation [30,31]. In
 12 cylindrical coordinates, the ideal solution of the electric field for the Bessel beam
 13 is of the form [27,32]

$$14 \quad E_n(r, \varphi, z) = \varepsilon e^{ik_z z} J_n(k_r r) e^{\pm i n \varphi}, \quad k = \sqrt{k_z^2 + k_r^2} = 2\pi/\lambda \quad (4.1.1)$$

15 The function J_n is the nth-order Bessel function of the first kind, k_z and k_r are the
 16 longitudinal and radial wave vectors, and r , φ and z are the radial, azimuthal and

1 longitudinal components respectively. The zeroth-order Bessel beam J_0 is the
 2 only solution in the Bessel class that has a non-diffracting central maximum on
 3 the axis and is circled by a sequence of circular nodes, while the high-order Bessel
 4 beams have a phase singularity on the beam axis and hence have the non-
 5 diffracting dark core. The Bessel beam can be considered as a set of plane waves
 6 propagating on a cone with apex angle [109]

$$7 \quad \theta = 2 \times \tan^{-1} \left(\frac{k_r}{k_z} \right) \quad (4.1.2)$$

8 and the central core spot size is defined as

$$9 \quad r_0 = \frac{2.405}{k_r}. \quad (4.1.3)$$

10 Mathematically the ideal Bessel solution to the Helmholtz equation
 11 carries infinite power over infinite area. One can create an approximation to a
 12 Bessel beam experimentally from a Gaussian beam using conical refractive
 13 surfaces. The corresponding beam is termed a Bessel-Gauss beam of zero order
 14 [138], which can be considered as a plane wave of the form J_0 multiplied by a
 15 Gaussian profile. FWHM of the central bright lobe of the Bessel-Gauss beam is
 16 inversely proportional to wave vector k_r , which is directly related with the waves'
 17 apex angle θ and the axicon's base angle α , shown by the ray trace diagram in
 18 Figure 33(c). Thus, the spot size of the axicon created Bessel-Gauss beam is
 19 inversely proportional to the axicon's base angle α . For the experimental
 20 verification, a collimated Gaussian beam from a diode-pumped solid-state CW

1 laser of 532 nm wavelength (GL532T8, SLOC) was made incident onto the
 2 axicons with different base angles. The resulting Bessel-Gauss beams were
 3 captured by a high-resolution CCD camera (DCU223C, Thorlabs) using a 10×
 4 magnification objective lens. The axicon based Bessel-Gauss beam maintains the
 5 central spot size along the propagation distance, L_B depending on the axicon's
 6 base angle α . The FWHM of Bessel-Gauss beam created by the axicon with base
 7 angle of 1°, 10°, and 20° (Thorlabs, AX251-A, AX2510-A, AX2520-A) are 40.0
 8 μm, 3.6 μm, and 1.9 μm respectively. The FWHM of the Bessel-Gauss beam is
 9 found to follow the relation

$$10 \quad FWHM_{Bessel} \approx \frac{38.0}{\alpha}. \quad (4.1.4)$$

11 The propagation distance L_B is given by

$$12 \quad L_B = \frac{\omega}{\tan[\sin^{-1}(n_{SU-8} \times \sin \alpha) - \alpha]} - thickness_{axicon}. \quad (4.1.5)$$

13 The micro-axicon fabricated has a base angle of 20° and a base diameter of 100
 14 μm, based on which the propagation distance L_B is calculated to be 203.5 μm. and
 15 the spot size is calculated to be 2.03 μm. The measured propagation distance from
 16 the experiments is 160 μm and the average spot size is 2.3 ± 0.25 μm, both which
 17 agrees to the theoretically calculated values.

18 The Bessel beam exhibits another outstanding feature of self-
 19 reconstruction as illustrated in the simulation colormap shown in colormap in
 20 Figure 33. The self-reconstruction nature of the beam is the capability to recover

1 its initial intensity profile after interacting with an object, or a scatterer that
2 locally shifts the phase or absorbs energy from the incident beam. The self-
3 reconstruction property has been experimentally demonstrated in various reports
4 and is widely regarded as a consequence of the conical wavefronts of the beam
5 [27,32,34,35]. The mechanism underlying this phenomenon is that the rays
6 refracted from the axicon's outer rings that bypass the obstruction begin to enter
7 the axis and interfere. As they carry energy, the same profile in the beam axis can
8 be restored. It has also been shown that Bessel-Gauss beams are able to self-
9 reconstruct even inside inhomogeneous, scattering media [34–36]. This self-
10 healing capability of the Bessel beam is highly advantageous in the high-
11 resolution imaging of thick, scattering micro-organisms or discrete clusters that
12 are difficult to image with a Gaussian illumination beam. However, there are very
13 few quantitative reports on the parameters that affect the self-reconstruction
14 properties of Bessel-Gauss beams. We herein investigate the dependence of self-
15 reconstruction distance on the scatterer's size, refractive index, and the offset of
16 the scatterer from the beam axis.

17

18 **4.2 Transformation of Gaussian Beam to Bessel-** 19 **Gauss Beam at a Micro-Axicon**

20 Figure 34 shows the transformation of the incident Gaussian beam to Bessel-
21 Gauss beam by the two-photon polymerized SU-8 micro-axicon.

Figure 34 Micro-axicon transforms the collimated Gaussian beam into the Bessel-Gauss beam. (a) Lateral profile images of the micro-axicon based Bessel-Gauss beam at $z = 230, 280, 330, 380, \text{ and } 430 \mu\text{m}$. (b) Lateral profile of the Bessel-Gauss beam collected at the peak position, $z = 380 \mu\text{m}$, matching with the 0th-order Bessel beam of first kind. (c) Colormap of the Bessel-Gauss beam in y-z plane. (d) Intensity propagation of the Bessel-Gauss beam along the z axis. (e) Bessel-Gauss beam's energy concentration and lateral FWHM of the central bright spot along the beam axis.

The lateral profiles of the propagating beam are captured by a CCD camera with $10 \mu\text{m}$ intervals along the central axis; five representative lateral profiles are presented in Figure 34 (a). The lateral beam profile at $z = 380 \mu\text{m}$ is shown in Figure 34 (b); this intensity profile matches well that of a zeroth-order

1 Bessel beam of the first kind, shown by the red dashed line. The Bessel-Gauss
2 beam shows an intense central lobe surrounded by weaker side lobes all along the
3 propagation axis. The signal-to-noise ratio, defined as the ratio of the central
4 signal intensity to first-order maximum herein, is measured to be 6.99 dB.

5 Figure 34(c) shows the axial intensity colormap in y-z plane; the Bessel-
6 Gauss beam formed by the micro-axicon maintains its spot size well along an
7 extended depth-of-focus over 500 μm from the micro-axicon's tip, which is much
8 longer than the depth-of-focus of 8.512 μm given by conventional objective lens
9 (NIKON 10 $\times$ Plan Achromat Objective, numerical aperture = 0.25) at wavelength
10 of 532 nm. Figure 34(d) shows the intensity along the propagation axis, which
11 increases after coming out from micro-axicon until 380 μm and decreases. The
12 axial-FWHM (z-FWHM) along the propagation axis is measured to 160 μm for
13 the range 280 – 440 μm from the tip of micro-axicon. In Figure 34(e), the blue
14 line shows the energy contained in the central main lobe of the Bessel-Guass
15 beam (the red shadowed portion in lateral profile in Figure 34(b)), which is
16 confirmed to carry more than 50% of total energy. The energy concentration gets
17 even higher to 71.6% of the total energy in the z-FWHM regime (between 280
18 and 440 μm distance from the micro-axicon tip). The lateral FWHM of the
19 Bessel-Gauss beam's central lobe is measured as shown to the green points in
20 Figure 34(e). It needs to be noted that the discrete nature of the green data points
21 comes from the pixel resolution (0.465 μm) of the CCD camera; the lateral

1 FWHM of the central lobe obtained by the data fitting is $2.3 \pm 0.25 \mu\text{m}$.

2

3 **4.3 Self-Reconstructing Bessel Beam: Simulation** 4 **of Propagation Characteristics**

5 The Bessel-Gauss beam can be formed by a well-designed conical surface which
6 refracts an input Gaussian beam towards the beam axis. Therefore, the rays
7 located outside of the obstruction still carry the photon energy and can form the
8 Bessel beam again even after encountering obstructions. The ray trace diagram
9 for a Gaussian beam (as described in Figure 13(a)) shows that the central
10 Gaussian beam cannot be restored after encountering obstructions, indicated as a
11 green sphere. On the contrary, the Bessel-Gauss beam can be reformed after
12 encountering each scatterer within the central Bessel region and can deliver
13 sufficient illumination over a long depth-of-focus; this self-reconstruction
14 capability makes the Bessel-Gauss beam the prerequisite to wide field-of-view
15 light-sheet beam microscopy. The self-reconstruction distance, defined as the
16 distance required to recover to the Bessel profile, is investigated by numerical
17 simulations via ZEMAX OpticStudio®. Mathematically, the self-reconstruction
18 distance (ZSR) can be defined as the distance from the object at which the lateral
19 profile recovers to the 0th-order Bessel function of the first kind with R^2 of 0.95.
20 The effects of the obstructing object's size, refractive index, and the lateral
21 deviation from the propagation axis onto the self-reconstruction are

1 systematically investigated.

2

3 Figure 35 Simulations of the self-reconstruction capability. (a) Gaussian beam
 4 encountering a bead of diameter $20 \mu\text{m}$, with $n = 1.37$, in the beam axis. (b-d) Bessel-
 5 Gauss beam encountering beads of diameters 10 , 20 , and $30 \mu\text{m}$, with $n = 1.37$. (e, f)
 6 Bessel-Gauss beam encountering a bead of diameter $20 \mu\text{m}$, with refractive indices $n =$
 7 1.46 and 1.76 . (g, h) Bessel-Gauss beam encountering a bead of diameter $20 \mu\text{m}$, which

1 is deviated 10 μm and 20 μm from the axis. (i) Self-reconstruction distance with respect
2 to the bead diameter. (j) Self-reconstruction distance with respect to bead's refractive
3 index. (k) Self-reconstruction distance with respect to the bead's deviation distance
4 from the beam axis.

5

6 The micro-axicon's geometrical shape is modeled in SOLIDWORKS®
7 and imported into the non-sequential simulation mode of ZEMAX OpticStudio®.

8 The micro-axicon has a base diameter of 100 μm , base angle of 20° , and refractive
9 index of 1.58 at the illumination wavelength of 532 nm, which is the same with
10 the experiment. A collimated 532-nm Gaussian beam with a beam diameter of
11 100 μm is set as the illumination source and 100,000,000 rays are used for the
12 simulation. This large number of rays ensures the number of rays collected by
13 each detector pixel to be more than 10,000 within the 3rd order side-lobe of the
14 Bessel-Gauss beam, so that the irradiance resolution is to be higher than 40 dB.
15 Figure 35(a) shows the Gaussian beam propagation (as the control model) when
16 the beam encounters a transparent bead of 20 μm diameter; the collimated
17 Gaussian beam of a 100 μm diameter gets diffracted after encountering the
18 transparent without reconstruction. Figure 35(b-h) show the propagation of the
19 Bessel-Gauss beams created by a micro-axicon, when it encounters transparent
20 beads of different sizes and refractive indices, located at an axial distance of 30
21 μm from the axicon tip. The bead's diameter was varied from 5 to 30 μm (See
22 Figure 35(b-d)); the bead size of 20 μm is taken as the control case analogous to
23 the HeLa cell to be investigated later. The refractive indices are set between 1.37

1 to 1.76 (See Figure 35(c, e, f)), considering typical biological organisms of red
2 blood cell, HeLa cell, cell membrane, chromosome, and nucleolus [139]. Further
3 analysis was addressed with different bead positions from 5 to 50 μm apart from
4 the propagation axis (See Figure 35 (g, h)). The resulting images were captured
5 using a rectangular detector of $100\text{ }\mu\text{m} \times 100\text{ }\mu\text{m}$ in size with 1001 pixels in each
6 direction (pixel size: $0.1\text{ }\mu\text{m}$); the lateral images were recorded from the axicon
7 tip with an interval spacing of one fifth of the bead diameter. The self-
8 reconstruction capabilities of the Gaussian and Bessel-Gauss beam are displayed
9 by colormaps, lateral profiles along the propagation axis (1) immediately after
10 the sphere, (2) at the halfway to the self-reconstruction distance, and (3) at the
11 final position where the profile recovers to the 0th-order Bessel function of the
12 first kind.

13 The self-reconstruction distance is strongly dependent on the object size
14 as shown in Figure 35(i). When the Bessel-Gauss beam encounters the
15 transparent beads with diameters of 5, 10, 20 and 30 μm (with the same refractive
16 index of 1.37), the beam recovers to the original Bessel profile at the distances of
17 $Z = 14, 30, 50$ and $71\text{ }\mu\text{m}$, respectively (Figure 35(b-d)). The self-reconstruction
18 distance is linearly proportional to the bead diameter by (in μm):

$$19 \quad Z_{SR} = 2.2 \times d_{bead} + 5.2. \quad (4.3.1)$$

20 The linear slope of this equation agrees well with the theoretical expectation of
21 2.27. Because the rays not encountering the transparent beads intersect at the

1 propagation axis and reconstruct the beam profile, the theoretical intersection
 2 distance can be estimated to:

$$3 \quad Z_{SR} = \frac{d_{bead}/2}{\sin[\sin^{-1}(n_{axicon} \times \sin \alpha) - \alpha]} = 2.27 \times d_{bead}, \quad (4.3.2)$$

4 given $n_{axicon} = n_{SU-8} = 1.58$, and axicon base angle $\alpha = 20^\circ$.

5 The effect of the refractive index onto the self-reconstruction distance of
 6 Bessel beam is shown in Figure 35(j). The different refractive indices ranging
 7 from 1.37 to 1.76 (with the same diameter of 20 μm) did not make a noticeable
 8 difference in the self-reconstruction distance. It is because the self-reconstruction
 9 distance is determined not by the beam passing through the bead but by the beam
 10 propagating out of the bead. Therefore, the self-reconstruction distance (Z_{SR}) was
 11 the same to 50 μm . Figure 35(k) shows the offset distance effect (of 20- μm sized
 12 beads) from the propagation axis onto the self-reconstruction distance. The
 13 strongest impact was observed when the bead is positioned at 10 μm from the
 14 propagation axis ($d_{bead} = 20 \mu\text{m}$, $n_{bead} = 1.37$), where the Bessel-Gauss beam recovered
 15 at $Z_{SR} = 75 \mu\text{m}$ after the bead. As the offset increases, the energy intersecting with
 16 the bead decreases, which results in a smaller the impact on the self-
 17 reconstructive capability. This impact gets negligible when the offset becomes
 18 larger than 50 μm .

1 **4.4 Fluorescence Microscopy with the** 2 **Self-Reconstructive Bessel-Gauss Beam from** 3 **the Micro-Axicon**

4 Figure 36 shows the fluorescence images of micro-beads recorded with micro-
5 axicon-based Bessel-Gauss beam illumination in comparison with the Gaussian
6 beam illumination; Rhodamine-B was used as the fluorescent dye herein. Figure
7 36(a, b) are conventional microscope images of the transparent micro-beads of
8 diameter 10 μm and 20 μm floating in Rhodamine B solution. Figure 36(c, d)
9 show the microscopic images recorded with the Gaussian beam illumination from
10 the left side. When the Gaussian beam encounters a single transparent polystyrene
11 bead of a 20 μm diameter, the beam is severely diffracted as in Figure 36(c).
12 Similar diffraction can also be seen in Figure 36(d) with two smaller 10- μm beads
13 for the Gaussian beam illumination. On the contrary, Figure 36(e, f) show the
14 microscopic images recorded with the Bessel-Gauss beam excitation from the left
15 side. When the Bessel-Gauss beam illuminates a single bead (see Figure 36(e)),
16 the beam recovers its original beam size and propagation characteristics after a
17 short distance. Compared to the conventional microscopic images, the photon
18 signals from other beads at different depths or background noise are greatly
19 reduced. In Figure 36(f), three 10 μm beads are successfully detected in series
20 along the same propagation axis, without severe diffraction issues.

1
2 Figure 36 Image comparison of different microscopic methods. (a) and (b) conventional
3 optical microscope images of the beads of different sizes. (c) and (d) show the
4 microscopic images of the beads using a Bessel-Gauss beam generated fluorescence
5 filtered with a band pass filter (FB600). (e) and (f) show microscopic images of the beads
6 using Gaussian beam generated fluorescence filtered with a band pass filter (FB600). (g)
7 Comparison of the intensity profiles of the Bessel-Gauss beam and Gaussian beam
8 before encountering the beads. (h) Comparison of the Bessel-Gauss beam and Gaussian
9 beam after encountering the beads, illustrating the non-diffractive nature of the Bessel-
10 Gauss beam. Scale bars: 100 μm

11

12 Figure 36(g) shows the intensity profiles of the Bessel-Gauss beam and
13 Gaussian beam (corresponding to line 1's in Figure 36(c, e)) before they meet the

1 beads. The two beams have similar beam profile and beam size before
2 encountering the beads. However, after obstructed by the beads (see Figure 36(h),
3 corresponding to line 2 in Figure 36(c, e)), it is evident that the Gaussian beam is
4 greatly diffracted (area shaded in red) compared to the Bessel-Gauss beam (area
5 shaded in blue). The Bessel-Gauss beam still maintains its narrow beam profile
6 and provides a steady and uniform illumination regardless of the existence of
7 prior objects or obstacles.

8 This self-reconstructive nature of the Bessel-Gauss beam makes the
9 micro-axicon to be an ideal beam shaping solution for light-sheet fluorescence
10 microscopy in biological imaging. The micro-organisms are generally cultured in
11 large clusters and are often complicated to image and analyze. Optical sectioning
12 techniques can assist in exploring the detailed structures in such clusters or tissues.
13 We used HeLa cells to explore the capabilities of the proposed light-sheet
14 fluorescence microscopy for biological imaging. The HeLa cell line is one of the
15 most commonly used immortal cell line in scientific research [140]; it can divide
16 an unlimited number of times and is a remarkably robust and durable cell, giving
17 rise to extensive uses in research. The cells grow rapidly in culture media under
18 room temperature and the cluster can spread over the petri dish in three days. In
19 the intracorporeal environment, these cancer cells survive as a cluster of tumor
20 tissues; while in the laboratory environment, the cells are adhered to the glass and
21 are overlaid with culture medium[141]. The overlaid cells create difficulties to

1 investigate the intracellular structures, so that it requires optical sectioning
2 techniques to better image the single cells and layers.

3 The microscopic images of the HeLa cells using conventional optical
4 microscopy and light-sheet fluorescence microscopy with micro-axicon are
5 shown in Figure 37.

Figure 37 Microscopic images of HeLa cells with different imaging methods. (a) Conventional optical microscope image of the cells. (b) Microscopic images of cells with Gaussian beam generated fluorescence. (c) Microscopic images of the HeLa cells with Bessel-Gauss beam generated fluorescence. (d) The intensity profile of the Bessel-Gauss illumination along the beam axis, where the shaded regions bounded by high contrast indicate the presence of HeLa cells. (e) The Bessel-Gauss beam profiles before and after the cells, indicating the Bessel-Gauss beam self-reconstruction. Scale bars: 100 μm .

The HeLa cells were cultured on a glass slide and each cell had spread to

1 an estimated diameter of 40 μm . Figure 37(a) shows the conventional optical
2 microscopic images of the HeLa cells. Figure 37(b) shows the fluorescence
3 microscope image with a Gaussian beam illumination from the left side; the
4 Gaussian beam is severely diffracted by the early cells so the subsequent sections
5 are blurred by the diffracted light. On the contrary, when the micro-axicon-based
6 Bessel-Gauss beam is incident from the left, the Bessel-Gauss beam uniformly
7 illuminates multiple target cells over a significantly longer depth-of-focus as
8 shown in Figure 37(c) and shows a higher image contrast (for example, in regime
9 (i)) compared to Figure 37(a) or (b). Because the Bessel-Gauss beam selectively
10 illuminates the cells at the specified plane, light-sheet fluorescence microscopy
11 is immune to the background noise formed at different depths of sample, as
12 highlighted in regime (ii). Figure 37(d) shows the cross-sectional intensity profile
13 along the propagation axis from Figure 37(c). The HeLa cells are distinguished
14 from the background with a high contrast over $30 \mu\text{m}^{-1}$ (See red shadow sections
15 in Figure 37(d)). The image contrast was calculated based on the Michelson
16 contrast, the intensity change per unit length along the propagation axis [142].
17 Figure 37(e) shows six cross-sectional intensity profiles at different positions in
18 Figure 37(c) as indicated with white arrows along the propagation axis. The
19 original input profile is shown at 25 μm before encountering the HeLa cells. The
20 later profiles deviated from the original profile clearly indicate the existence of
21 individual HeLa cells at each position.

1 These series of experiments and simulations confirm that the light-sheet
2 fluorescence microscopy adopting an additively manufactured micro-axicon on
3 the tip of an optical fiber can provide a selective Bessel-Gauss illumination
4 supporting uniform illumination in scattering media and improved image contrast.
5 Furthermore, this micro-axicon eliminates the need for a bulky illumination
6 objective lens for generating a non-diffractive pencil-like beam for enhanced
7 axial resolution. Depending on target samples, one can tailor an optimized
8 Bessel-Gauss beam illumination by designing the micro-axicon's conical angle
9 and base diameter appropriately, which finally determine the axial resolution and
10 field-of-view of the light-sheet fluorescence microscope. Although the lateral
11 resolution of the light-sheet fluorescence microscope is the same as the
12 conventional optical microscope's, the axial resolution can be enhanced to the
13 thickness of the light-sheet.

14

Chapter 5

Collimated Beam Tilt Angle

Measurement with Diffractive Axicon

The diffractive optical element (DOE) is defined as the optics possess with operation principles which are essentially based on the phenomenon of light diffraction. A various range of optical functions can be obtained with such devices. The diffraction functions are usually work at the order of wavelength dimension. Thus, the term diffractive optical element is most frequently used in the context of micro-/nano-optic elements. Due to its advantages in the fast design and fabrication of the reduced graphene oxide (rGO) thin film, and the flexibility of applying the thin optics attached to the various devices, it can serve as the important substitutes to some refractive optical elements in many applications.

In this chapter, I demonstrate the proposed application of the light beam tilted angle measurement by the rGO diffractive axicon transformed beam patterns, followed by the design and fabrication of the diffractive axicon, the optical performance, and the beam tilt angle measurement by the diffractive axicon generated pattern evolution.

5.1 Optical Layout of Diffractive Axicon Based Beam Tilt Angle Measurement

The two-level diffractive optical element is fabricated with reduced graphene-

oxide (rGO) thin film by the high-speed femtosecond laser direct scanning (FsLDS) technology, as introduced in the Chapter 3.3.

The rGO diffractive axicon in this work is designed and applied as the Thorlabs axicon (AX251-A, 1.0° , AR coated, $\varnothing 25.4$ mm). The axicon, precisely manufactured from high-quality UV Fused Silica, has a base angle of 1° and apex angle of 178° , and a refractive index $n_{FS} = 1.46$ at $\lambda = 532$ nm.

7

8 Figure 38 Geometry of an axicon forming a Bessel beam: (a) Thorlabs axicon and
9 (b) diffractive axicon DOE [133].

10

11 The converging angle θ_b of the refractive axicon AX251-A is calculated as

$$\sin \theta_{refractive} \times n_{air} = \sin \alpha \times n_{FS} \quad (5.1.1)$$

$$\begin{aligned} \theta_b &= \theta_{refractive} - \alpha \\ &= \arcsin(\sin \alpha \times n_{FS}) - \alpha \\ &= \arcsin(\sin 1^\circ \times 1.46) - 1^\circ = 0.46^\circ \end{aligned} \quad (5.1.2)$$

The diffraction-free propagation distance L_B is the same as in Equation 4.1.5:

$$L_B = \frac{\omega}{\tan \left[\sin^{-1}(n_{FS} \times \sin \alpha) - \alpha \right]} - thickness_{axicon} \quad (5.1.3)$$

where the ω is the radius of input beam.

$U(X, Y) = U(R)$ denotes the optical field immediately after the lenses, and the propagation of the field is expresses as $u(x, y, z) = u(r, z)$, where r is the radial coordinate in the observation field. We can ignore the thickness of DOE, then we the equation:

$$u(x, y, z) = \frac{ik}{2\pi} \iint_A U(X, Y) \frac{\exp(-ikL)}{L} dXdY \quad (5.1.4)$$

where L is the distance between the observation point and a known point in X-Y plane immediately after the lens ($z = 0$), wave number $k = 2\pi / \lambda$, and A is the incident area (either the input beam size or the aperture size, which is the smaller one. In Fresnel diffraction field, the distance L can be expanded and simplified to a near-axis approximation:

$$\begin{aligned}
L &= z \left[1 + \frac{(X-x)^2 + (Y-y)^2}{z^2} \right]^{1/2} \\
&= z + \frac{1}{2z} [(X-x)^2 + (Y-y)^2] + \frac{1}{8z^3} [(X-x)^2 + (Y-y)^2]^2 + \dots \\
&\approx z + \frac{1}{2z} [(X-x)^2 + (Y-y)^2]
\end{aligned} \tag{5.1.5}$$

Express the optical field U in the cylindrical coordinates:

$$U(R) = \begin{cases} \exp(ikL \sin \theta_b), & R \leq \omega \\ 0, & R > \omega \end{cases} \tag{5.1.6}$$

We are able to obtain the following equation if substituting the Equation 5.1.6 into 5.1.4:

$$\begin{aligned}
u(r, z) &= \frac{ik}{z} \exp \left[-ik \left(z + \frac{r^2}{2z} \right) \right] \times \int_0^\omega \exp(ikR \sin \theta_b) \\
&\times \exp \left(-ik \frac{R^2}{2z} \right) J_0(kr \sin \theta_b) R \cdot dR
\end{aligned} \tag{5.1.7}$$

Then the optical intensity along the beam axis can be easily derived as $r = 0$:

$$u(z) = \frac{ik}{z} \exp[-ikz] \int_0^\omega \exp(ikR \sin \theta_b) \times \exp \left(-ik \frac{R^2}{2z} \right) R \cdot dR \tag{5.1.8}$$

J_0 is the 0th-order Bessel beam function of the first kind. The light intensity $I(r; z)$ is calculated as the square of the optical field $u(r; z)$, is expressed using the stationary-phase method:

$$\begin{aligned}
I(r, z) &= |u(r, z)|^2 \\
&\propto z J_0^2(rk \sin \theta_b)
\end{aligned} \tag{5.1.9}$$

This equation implies that the beam is a Quasi-Bessel beam, i.e, similar results as

1 the refractive axicon near the beam axis. The radius r_0 of the resulted Bessel beam
 2 in Equation 5.1.9 is then calculated as:

$$\begin{aligned}
 r_0 &= \frac{2.405}{k \sin \theta_b} = \frac{2.405}{k_r} \\
 &= \frac{0.382}{\sin \theta_b} \lambda = 25.313 \mu\text{m}
 \end{aligned}
 \tag{5.1.10}$$

4 which is uniform with the Equation 4.1.3. The equation shows that the resulted
 5 Bessel beam spot size $2r_0$ does not change with the propagation distance. The
 6 phenomenon has been verified with the simulation and experiment of the
 7 analogous refractive axicon in Chapter 4.

8 Figure 38(b) shows the propagation and focusing diagram of diffractive
 9 axicon. Center of the diffractive axicon is defined as the coordinate origin and its
 10 normal axis aligns with the beam axis. The diffractive axicon is found to be
 11 composed of concentric rings with constant ring width and pitch on planar
 12 substrate. The thickness of the lens, which mainly refers to the thickness of the
 13 substrate is ignored.

14 The beams are diffracted to different orders at the borders of rings.
 15 Positive order diffracted beams are convergent to the beam axis, while the
 16 negative ones are divergent. Negative orders are ignored here since the diverging
 17 beams have small effect on the focusing and the resulted beam profile. The
 18 diffraction angle θ_j , which is the j 'th-order diffracted beam formed by the DOE,
 19 is given as

$$\theta_j = \arcsin\left(\frac{j\lambda}{A}\right), \quad j=1,2,3,\dots \quad (5.1.11)$$

where A is the pitch of the concentric rings. Then the corresponding propagation distance z_j is given as

$$z_j = \frac{\omega}{\tan(\theta_j)} \quad (5.1.12)$$

The radius r_j of the central spot of resulted beam, which is formed by the j 'th-order diffracted beam, is then derived from Equation 5.1.9 by directly substituting the θ_b by θ_j :

$$r_j = \frac{2.405}{k \sin \theta_j} = \frac{2.405}{(2\pi / \lambda) j \lambda} A = \frac{0.383}{j} A \quad (5.1.13)$$

It can be found that the radius r_j is independent of incident wavelength, but inversely proportional to the diffraction order j . The optical field $u_j(z)$ on the axis formed by j 'th diffracted beam is derived from Equation 5.1.8:

$$u_j(z) = \frac{ik}{z} \exp[-ikz] \int_0^\omega \exp(ikR \sin \theta_j) \times \exp\left(-ik \frac{R^2}{2z}\right) R \cdot dR \quad (5.1.14)$$

where z is in the domain of $(0, z_j)$. Therefore, the overall intensity $I(z)$ is the integrate of all the diffracted fields:

$$I(z) = \left| \sum_{j=0}^N \sqrt{\eta_j} u_j(z) \right|^2 \quad (5.1.14)$$

where N is the outermost ring participating in the diffraction, and η_j is the

1 diffraction efficiency of the corresponding diffracted beam.

2 The diffractive axicon is designed as the optical field of its 1st-order
3 diffracted beam is proportional to the refracted beam from the refracting axicon
4 from Equation 5.1.11:

$$\begin{aligned} \sin \theta_b &= \frac{j\lambda}{A}, \quad j = 1 \\ A &= \frac{\lambda}{\sin \theta_b} \end{aligned} \quad (5.1.15)$$

6 The diffractive axicon in this work is designed as the Thorlabs axicon
7 AX251-A with base angle of 1° and refractive index $n = 1.46$, in the $\lambda = 532$ nm
8 environment. Hence the diffractive axicon is calculated to have a constant pitch
9 A of 66.265 μm and a duty cycle of 50%, that is a diffractive axicon consisting
10 of a central circle of radius 33.132 μm and concentric rings with constant ring
11 width and interval of 33.132 μm , as shown in Figure 39. Corresponding
12 diffractive axicons analogous to the refractive axicons of commonly used base
13 angle 10° has calculated ring width and interval of 6.512 μm , and that of 20° has
14 3.077 μm . For the binary diffractive axicon, the alternate transparent and opaque
15 concentric rings share the identical ring width and interval, that depend on
16 incident λ and θ_b , which in turn depends on the refractive power of the comparable
17 refractive axicon. The sharper the refractive axicon is, the smaller the apex angle,
18 and the thinner rings the comparable diffractive axicon has.

Figure 39 Optical microscopic images of the diffractive axicon with a ring width of 33.1 μm that can work as the axicon AX251-A

5.2 Transformation of Gaussian Beam to Bessel-Gauss Beam at Diffractive Axicon

The designed diffractive axicon has been designed and fabricated with the graphene-oxide/reduced graphene-oxide thin film, as described in Chapter 3.3, and tested for the transformation performance. The result is compared with the Thorlabs axicon AX251-A.

Figure 40 shows the transformation colormaps of the incident Gaussian

1 beam to Quasi-Bessel beams by the Thorlabs axicon AX251-A and the printed
2 diffractive axicon.

3
4 Figure 40 Axicons' transformation performance. (a) The Thorlabs Axicon AX251-A
5 transforms the Gaussian beam into smooth Bessel-Gauss beam; (b) the diffractive
6 axicon transforms the Gaussian beam into Quasi-Bessel beam with some intensity
7 fluctuation. (c) The lateral profile captured at $z = 50.5$ mm at the dashed line in (a).
8 (d) The lateral profile captured at $z = 48$ mm at the dashed line in (b)

9
10 A collimated Gaussian beam of wavelength 532 nm and beam width of 1
11 mm is incident perpendicularly to the axicons. The lateral profiles of the
12 propagating beams are captured by a CCD camera with 0.5 mm intervals along

1 the central axis, starting from 35 mm and 27 mm from the axicons respectively.
2 The preceding profiles are not able to be captured due to the setup restriction of
3 the lens holder and tube mount in front of the CCD's sensor panel. The tip of the
4 Thorlabs axicon AX251-A and the central point of the printed diffractive axicon
5 are taken as the origins in the coordinate system.

6 Axicon AX251-A has been confirmed to transfer the perpendicularly
7 incident Gaussian beam into the approximate Bessel beam, which is termed as
8 Bessel-Gauss beam, with constant full width of half maximum (FWHM) of 40.0
9 μm . and finite propagation distance (z-FWHM, the distance having intensity
10 larger than half of the maximum) of more than 55.5 mm (35.0 to 90.5 mm from
11 the axicon tip).

12 The diffractive axicon transfers the incident Gaussian beam into Quasi-
13 Bessel beam as well. The colormap clearly shows that the resulted beam
14 possesses a constant beam size along the beam axis. The detected z-FWHM is
15 found in a range of 24.0 mm that between 29.5 to 53.5 mm from the lens, having
16 two peak regions: 29.5 to 38.5 mm and 43 to 53.5 mm. The concentric ring
17 patterns in the profile shows some extend of diverging. However, the profile
18 within the z-FWHM range shows good coincidence with the 0th-order Bessel
19 function of the first kind. The shorter propagation distance and less constant
20 intensity are reasonable since the diffractive axicon has opaque ring patterns that
21 block some of the incident energy.

1 Figure 40(c) and (d) show that the resulted Quasi-Bessel beams have the
2 similar concentric ring patterns in profiles. The ring radii and intensities are
3 further investigated in Figure 41.

4
5 Figure 41 Normalized lateral intensity profiles of Quasi-Bessel beams created by printed
6 diffractive axicon and Axicon AX251-A, and comparison with the standard 0th-order
7 Bessel function of the first kind.

8
9 The lateral profiles of the created Quasi-Bessel beams captured at the
10 peak points are shown in Figure 41. The peak of Quasi-Bessel beam created by
11 the diffractive axicon happens at $z = 48$ mm from the axicon, while the one by
12 Axicon AX251-A happens at $z = 50.5$ mm. The results are compared with
13 standard 0th-order Bessel function of the first kind. The 0th-order Bessel function
14 is fitted to the Axicon AX251-A created Bessel-Gauss beam, having a central

1 beam size of $55.80\text{ }\mu\text{m}$ and the FWHM of the central beam size of $26.27\text{ }\mu\text{m}$. The
 2 first-order side lobes occurring at $46.5\text{ }\mu\text{m}$ from the center have intensity of 15.8%
 3 of the central beam spot.

4 The lateral profile created by Axicon AX251-A matches well with the
 5 Bessel function. The intensity ratio of 1st-order side lobe to the central spot is
 6 measured to be 13.6%. In other words, signal-to-noise ratio (SNR), defined as the
 7 ratio of the central signal intensity to first-order maximum herein, is 8.66 dB. The
 8 profile created by the diffractive axicon also shows the same FWHM of central
 9 beam similar side lobes as the Bessel function and the Bessel-Gauss beam. The
 10 ratio of noise is taken as the second maximum to the central intensity at the radius
 11 of $69.75\text{ }\mu\text{m}$ from the central point, and is measured to be 9.94%. The signal-to-
 12 noise ratio (SNR) is then 10.03 dB.

13
 14 Figure 42 FWHM propagation of the resulted Bessel beams transformed by (red)
 15 diffractive axicon and (green) Thorlabs axicon AX251-A. Dashed lines represent the
 16 mean of FWHM along the propagation distance.

1 The lateral FWHM of the resulted Bessel beams' central lobes are
2 measured as shown in the data points in Figure 42. The discrete nature of the data
3 points comes from the pixel resolution ($0.465\text{ }\mu\text{m}$) of the CCD camera. The lateral
4 FWHM of the diffractive axicon resulted central lobe obtained by the data fitting
5 is $26.80 \pm 3.88\text{ }\mu\text{m}$. The effective central lobe can be further confined to $25.39 \pm$
6 $2.32\text{ }\mu\text{m}$ if the deviated points after 64.5 mm are ignored. The result is well
7 matched with the one created by Thorlabs axicon AX251-A that having a central
8 lobe of $26.48 \pm 2.14\text{ }\mu\text{m}$. Comparing to the standard 0th-order Bessel beam of
9 first kind that has a FWHM of $26.27\text{ }\mu\text{m}$, it shows that the diffractive axicon
10 presents the transformation function well matched with the theoretical design and
11 the commercialized refractive axicon.

12

13 **5.3 Beam Tilted Angle Measurement by the** 14 **Diamond Pattern**

15 The Bessel beam possesses the advantages, including an extended working
16 distance between the workpiece and lens can be used, avoiding ablated material
17 sticking to the lens, as well as the precise positioning for focusing is not required.
18 In addition to these advantages, it has also demonstrated that an axicon allows a
19 larger maximum permissible misalignment than a conventional lens.

20 In this section, we are demonstrating that the beam axis maintains

1 regardless of the tilted angle of the axicon. Although a permissible misalignment
2 does not alter the resulted beam axis with respect to the axicon's normal axis, the
3 transformed patterns are sensitive to the tilted angle between the beam axis and
4 the axicon's normal axis, so that it can work as a good device to detect the tilted
5 angle between a transparent surface and the laser beam axis. We herein
6 investigate the variation of beam pattern with respect to the tilted angle between
7 the beam axis and lens normal, as well as the distance from the lens.

Figure 43 Schematic diagram of the setups of axicons transforming the incident beams. (a) the Thorlabs Axicon transforms the perpendicularly incident beam. (b) the Thorlabs axicon is tilted for an angle β with respect to beam axis. (c) the printed diffractive axicon tilted for the same β transforms the incident beam.

The schematic diagrams of the axicons transforming the incident beams at different angles are shown in Figure 43. Figure 43(a) shows the Axicon AX251-A is installed perpendicularly to the beam axis and therefore, the resulted Bessel-Gauss beam propagates along the original beam axis that extended from the

1 incident beam. In Figure 43(b) the AX251-A is tilted for an angle β between the
2 normal of the axicon and beam axis around the origin/the tip of the axicon. It is
3 found that the resulted beam propagates parallel to the original beam axis, which
4 implies that the tilted axicon does not alter the direction of the resulted beam.
5 However, the tilted angle of the axicon has incident beam refract within the lens
6 itself, so that the resulted beam is deviated from the original beam axis for a
7 distance. It implies that the central of incident beam has to be shifted for the
8 distance d_{sh} , so that the central beam will interact with the tip of axicon. The
9 central beam passing through the tip of axicon is indicated as the red line in Figure
10 43(b). The deviation distance d_{sh} is calculated by

$$11 \quad \begin{aligned} d_{sh} &= thickness_{axicon} \times \sin(\beta - x) \\ n_{air} \sin x &= n_{axicon} \sin \beta \end{aligned} \quad (5.2.1)$$

12 where x is the angle between the refractive beam inside the axicon and its normal
13 line.

14 In practice, the laser source is installed on the linear translator and shifted
15 in x-axis to fit the tip of axicon. The DOE has an advantage in investigation of
16 the transformation performance versus tilted angle since that the thin film
17 structure enable us to ignore the refraction and deviation distance d_{sh} . It is found
18 that the propagation axis is extended from the original incident axis, and does not
19 alter with the tilted angle of DOE. The deviation distance d_{sh} is negligible since
20 the thickness of diffractive axicon is as fine as 150 μm , and can be further

1 minimized by using the finer substrates. The laser source is rarely necessary to
2 shift so that the performance recording precision is enhanced.

3 We herein investigate the variation of beam pattern with respect to the
4 tilted angle between the beam axis and lens normal, as well as the distance from
5 the diffractive lens.

6

7 **5.3.1 Transformation by Diffractive Axicon Tilted to** 8 **Different Angles**

9 The diffractive axicon is rotated from the perpendicular position to the beam axis
10 to a tilted angle β , as shown in Figure 44, with an interval of 1° , up to 30° where
11 the resulted beam intensity fades. The resulted beam patterns are captured by
12 CCD camera at $z = 48$ mm from the origin point, where is found to be the position
13 of peak intensity in the diffractive axicon straight transformation case and having
14 smooth intensity propagation in the nearby region.

1

2 Figure 44 Resulted beam pattern variation regarding the diffractive axicon's tilted angle
3 to the incident beam. (a) Schematic diagram of tilted diffractive axicon transforming the
4 incident beam. (b-g) the lateral profile of resulted beam patterns transformed by
5 diffractive angle at different tilted angles β . (h) X-cross-sectional profiles captured by
6 CCD camera at $z = 48$ mm, with tilted angle interval of 1° . Each figure is normalized
7 individually.

8

9 The resulted beam maintains the Quasi-Bessel profile until the diffractive
10 axicon is tilted to 9° . Thereafter the central beam spot shows obvious division to

1 two side spots in X-axis. The X-axis is perpendicular to the beam axis while the
2 Y-axis, rotation axis, is herein perpendicular to the paper in Figure 44(a). The
3 first order side spot is defined as a spot that occurs closest to the central position
4 and has an intensity larger or equal to 15.77% of the central maximum value,
5 which is the degree of the first order side lobe of the Bessel function. Besides,
6 there is significant intensity difference between the spots and other position on
7 the ring that has same distance to the center position. When the tilted angle β of
8 axicon is increased to 12° , the central beam spot has developed to two side X-
9 spots. When the axicon is tilted to $\beta = 18^\circ$, the pattern has two side X-spots having
10 equal intensity with the central beam, and the central spot eliminates at $\beta = 21^\circ$,
11 where the central position becomes a local minimum. The resulted pattern by
12 tilted diffractive axicon develops to a checkerboard pattern.

13

14 **5.3.2 Transformation by certain tilted diffractive axicon** 15 **along the beam axis**

16 I further investigate the beam pattern development along the beam axis that
17 transformed by tilted diffractive axicon at certain angle β . The trend of pattern
18 development shows that further the propagation distance, more evolution from
19 the Bessel profile or straight-transformed Quasi-Bessel beam profile is generated.

20 The resulted profiles created by the tilted diffractive axicon start to show

1 the full trend of pattern development along the beam axis: (1) the tilted axicon
2 transforms the beam to the Quasi-Bessel beam immediately after the lens; (2) the
3 central spot develops two side X-spots on the left and right sides to the center,
4 and the intensity ratio of side spot to central beam increases; (3) the pattern
5 develops two Y-spots above and below the central spot, and becomes
6 checkerboard pattern, and the central spot fades; (4) the checkerboard pattern
7 converts to Y-spots, and the central beam spot appears dominated again; (5) the
8 beam pattern recovers to the Quasi-Bessel profile.

9

10 **Transformation by 5°-tilted axicon**

11 The diffractive axicon transforms the incident Gaussian beam at tilted angle $\beta =$
12 5° . The CCD camera scans the resulted beam profiles along the beam axis from
13 $z = 28$ to 77 mm, with an interval $z = 0.5$ mm. The results are collected in the
14 colormaps in Figure 45.

1

2 Figure 45 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 5^\circ$.
3 (a) Schematic diagram of 5° -tilted diffractive axicon transforming the incident beam.
4 (b-i) the lateral profile of resulted beam patterns transformed by the tilted diffractive angle
5 captured at different distance from the lens. (j) X-cross-sectional profiles captured by
6 CCD camera from $z = 28$ to 77 mm, with interval of 0.5 mm. Each figure is normalized
7 individually.

8

9 The central beam aligns well with the incident beam axis regardless of the
10 tilted angle of diffractive axicon. The intensity shows the similar trend as the
11 straight transformation of the lens in Figure 40(b), that has two local peaks at $z =$
12 32.0 and 48.0 mm from the lens. The intensity along the beam axis degrades to

1 $1/e^2$ of maximum value after $z = 70.5$ mm. The beam pattern does not show
2 obvious division to the side spots or checkerboard pattern within the
3 measurement range. The FWHM of the central beam is calculated as $(26.31 \pm$
4 $5.02) \mu\text{m}$ within the propagation distance of 70.5 mm.

6 Transformation by 10° -tilted axicon

7
8 Figure 46 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 10^\circ$.
9 (a) Schematic diagram of 10° -tilted diffractive axicon transforming the incident beam.

1 (b-i) the lateral profile of resulted beam patterns transformed by the tilted diffractive angle
2 captured at different distance from the lens. (j) X-cross-sectional profiles captured by
3 CCD camera from $z = 29$ to 76 mm, with interval of 0.5 mm. Each figure is normalized
4 individually.

5

6 The general trend of transformation by the 10° -tilted diffractive axicon is
7 shown in Figure 46. The transformation is similar to the case that tilted angle $\beta =$
8 5° in Figure 45. The beam pattern does not show the total division from central
9 spot to the checkerboard pattern within the measurement range. However, in the
10 region from 54.5 to 58.0 mm from the lens, there shows the division to the side
11 X-spots on the left and right sides of the central spot. The X-spots merge to the
12 ring pattern before it develops the Y-spots. The resulted pattern recovers to the
13 dominated central spot pattern as the Quasi-Bessel beam profile at $z = 58.0$ mm.
14 The intensity along the beam axis degrades to $1/e^2$ of maximum value after the
15 distance $z = 69.0$ mm. The FWHM of the central beam within the propagation
16 distance is measured to be $(26.11 \pm 5.18) \mu\text{m}$.

17

1 Transformation by 15°-tilted axicon

2

3 Figure 47 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 15^\circ$.
 4 (a) Schematic diagram of 15°-tilted diffractive axicon transforming the incident beam.
 5 (b-i) the lateral profile of resulted beam patterns transformed by the tilted diffractive angle
 6 captured at different distance from the lens. (j) X-cross-sectional profiles captured by
 7 CCD camera from $z = 29$ to 76 mm, with interval of 0.5 mm. Each figure is normalized
 8 individually.

9

10 The transformation by the 15°-tilted diffractive axicon is shown in Figure

11 47. The propagation length (the distance where the intensity fades to less than

1 $1/e^2$ of the maximum intensity) is reduced to 53.0 mm from the lens. The FWHM
2 of the central beam is calculated as $(27.24 \pm 6.22) \mu\text{m}$ within the propagation
3 distance of 53.0 mm.

4 In this measurement of the 15° -tilted diffractive axicon transforming the
5 incoming beam, the resulted pattern has developed the side X-spots beside the
6 central spot from $z = 34.5$ mm. It then develops to the four equal-intensity side
7 spots in both X- and Y-axes forming the checkerboard pattern since $z = 38.0$ mm.
8 It converts to two Y-spots from $z = 42.0$ mm and the recover to the Quasi-Bessel
9 profile at $z = 45.5$ mm. It repeats the steps that develops the X-spots pattern again
10 from $z = 47.0$ mm

11

1 Transformation by 20°-tilted axicon

2

3 Figure 48 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 20^\circ$.
 4 (a) Schematic diagram of 20°-tilted diffractive axicon transforming the incident beam.
 5 (b-i) the lateral profile of resulted beam patterns transformed by the tilted diffractive angle
 6 captured at different distance from the lens. (j) X-cross-sectional profiles captured by
 7 CCD camera from $z = 29$ to 76 mm, with interval of 0.5 mm. Each figure is normalized
 8 individually.

9

10 The transformation performance by the 20°-tilted diffractive axicon is
 11 shown in Figure 48. The propagation length of central beam is measured to be
 12 49.0 mm. The central beam does not dominate the propagation in the

1 measurement range. Due to the detector system restriction, the propagation within
2 the range of 29.0 mm from the lens was not captured. The resulted beam pattern
3 shows that it repeats the trend of propagation thereafter. From the distance $z = 29$
4 mm, the pattern starts with a checkerboard pattern with a central spot, that has 4
5 equal-intensity side spots on the X- and Y-axes around and having intensity of
6 24.5% of the central beam. Then it has developed to have the side X-spots from
7 $z = 31.5$ mm. The central spot eliminates at $z = 35.0$ mm and the profile becomes
8 the checkerboard pattern without the central spot from $z = 37.0$ mm, and becomes
9 the Y-spot pattern from $z = 40.0$ mm. Thereafter it converts to the checkerboard
10 pattern instead of the ring pattern in a Bessel profile, and accompany with the
11 central spot from $z = 44.0$ mm. it repeats the pattern development from there on
12 until where the intensity fades to less than $1/e^2$ of the peak value at $z = 29.0$ mm.

13

1 Transformation by 25°-tilted axicon

2

3 Figure 49 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 25^\circ$.
 4 (a) Schematic diagram of 25°-tilted diffractive axicon transforming the incident beam.
 5 (b-i) the lateral profile of resulted beam patterns transformed by the tilted diffractive angle
 6 captured at different distance from the lens. (j) X-cross-sectional profiles captured by
 7 CCD camera from $z = 31$ to 76 mm, with interval of 0.5 mm. Each figure is normalized
 8 individually.

9

10 The transformation performance by the 25°-tilted diffractive axicon is
 11 shown in Figure 49. The detector system can only capture beam profiles from z
 12 $= 31.0$ mm. Basically there is no dominating central beam in the measurement

1 range. It starts with the X-spot pattern, and the central beam eliminates at $z = 32.0$
2 mm. Then it develops to become the checkerboard pattern without the central
3 beam from $z = 35.0$ mm, and the Y-spot pattern from $z = 39.0$ mm. It later turns
4 to the checkerboard pattern at $z = 43.0$ mm. However, the pattern consists of the
5 second-order side spot in both X- and Y-axes that having comparable intensities,
6 so that the beam profile becomes complicated that exceed the discussion in this
7 work.

1 Transformation by 30°-tilted axicon

2

3 Figure 50 Resulted beam pattern transformed by tilted diffractive axicon at $\beta = 30^\circ$.
 4 (a) Schematic diagram of 30°-tilted diffractive axicon transforming the incident beam.
 5 (b-i) the lateral profile of resulted beam patterns transformed by the tilted diffractive angle
 6 captured at different distance from the lens. (j) X-cross-sectional profiles captured by
 7 CCD camera from $z = 31$ to 76 mm, with interval of 0.5 mm. Each figure is normalized
 8 individually.

9

10 The transformation performance by the 30°-tilted diffractive axicon is
 11 shown in Figure 50. There is no dominating central beam in the measurement
 12 range and the resulted beam pattern diverges more than the cases before. It starts

1 with the checkerboard pattern from $z = 31.0$ mm, and the side spots spread to
2 away from the central position. Thereafter the intensity of 1st-order X-spots is
3 decreasing and that of 2nd-order X-spots is increasing until $z = 36.5$ mm. The two
4 Y-spots develop to split to four circular-symmetrical side spots in the four
5 quadrants, i.e, on both $\pm 45^\circ$ axes to the X-axis, and the 1st-order X- and Y-spots
6 are reduced at $z = 40.0$ mm. The beam profile becomes a checkerboard pattern on
7 the $\pm 45^\circ$ axes. The pattern is stretched along the X-axis and the quadrant-spots
8 have the intensity reduced and the X-spots stand out from $z = 48.0$ mm. The Y-
9 spots repeats to split to quadrant side spots from $z = 54.0$ mm. The beam intensity
10 spreads to the outer-ring pattern, so that the beam profile becomes complicated
11 that exceed the discussion in this work.

12 The transformation performance by tilted diffractive axicon at certain
13 angles show the regular evolution of the profiles. This is a good property that can
14 be used to detect the tilted angle between two incident beams or with a object
15 surface by attaching the thin film rGO diffractive axicon onto the object.

16

Chapter 6

Conclusion and Recommendation for Future Work

6.1 Conclusion

In this study, I successfully set up the femtosecond-laser-based fabrication technologies for the micro-optics, and use them for fabrications of different optics. The functional micro-optics showed their practical applications in the Light Sheet Fluorescence Microscopy and the Beam tilt angle measurement. I conclude the research results and my contributions in the following sections.

10

Establishment of Femtosecond Laser Direct Writing System

I established the femtosecond laser direct writing (FsLDW) and the high-speed femtosecond laser direct scanning (FsLDS) systems for the micro-lens fabrication.

FsLDW system is a powerful tool to fabricate the micro-optics with a dimension of less than 200 μm and the resolution of an estimated 1.4 μm . We additively manufactured the 3D micro refractive optics by relatively scanning the laser focal point inside the material. The femtosecond laser pulse of 780 nm wavelength and 7 mW power was focused to a focal point of lateral spot size 2.15 μm and axial one 3.18 μm into the photoresist SU-8, which induced the two-

1 photon absorption and polymerization within the photoresist. The single scanned
2 polymerized SU-8 line width achieved lateral resolution as fine as 1.4 μm , which
3 was better than the spot size. The axial resolution of the polymerization depended
4 on the 3D scanning nano-positioning stage, which achieved resolution as high as
5 1 nm, as well as the pre-designed step size of scanning routines.

6 This printing system enables the fabrication of the free shape micro-optics
7 with sufficient precision and accuracy. Besides, it allows a wide choice of
8 materials, including the polymers and compounds whichever are sensitive to UV
9 excitation. The progressive results are published as conference papers, Li, Mishra
10 and Kim, 2017; Li, Jiao and Kim, 2018, and a journal article, Li *et al.*, 2021.

11 FsLDS system was used to fabricate the thin film diffractive optical
12 elements (DOE) with reduced graphene oxides (rGOs). Femtosecond laser pulse
13 of 515 nm wavelength was employed to reduce the graphene oxides with three
14 regimes: (1) phase-changing less-thermal photoreduction (LTPR) regime; (2)
15 transmittance-changing thermal photoreduction regime (TPR); and (3) ablation
16 (ABL) regime. The three regimes were achieved by selecting the proper
17 femtosecond laser repetition rate and pulse energy. The fabrication resolution
18 achieved as high as 5 μm . They formed the diffractive optics with the rGO thin
19 film by giving different transmittances and diffractive effects to the incident light.

20

1 **Light Sheet Fluorescence Microscopy with** 2 **Micro-Axicon Created Bessel-Gauss Beam**

3 I fabricated a SU-8 micro-axicon of base diameter 100 μm , base angle 20° , and
4 tip apex angle of 140° with the established FsLDW system. Its performance that
5 transforms the incident collimated Gaussian beam into Bessel-Gauss beam has
6 been confirmed and compared with the commercial 1-inch-sized axicon AX2520-
7 A that has the same apex angle. Unlike the Gaussian beam that diverges after a
8 certain distance, the generated Bessel-Gauss beam possessed the non/less-
9 diffractive property as it maintained the beam size of $2.3 \pm 0.25 \mu\text{m}$ along the
10 propagation distance of 160 μm . Besides, the Bessel-Gauss beam showed its self-
11 reconstructive property when it encountered objects or disturbing scatterers. The
12 properties made the micro-axicon created Bessel-Gauss beam an ideal
13 illumination source for light sheet fluorescence microscopy.

14 I demonstrated the light sheet fluorescence microscopy (LSFM) with the
15 FsLDW micro-axicon-created Bessel-Gauss beam. The floating polystyrene
16 micro-beads and cultured Hela cells cultured on glass were detected and imaged
17 by the Bessel-like illumination, and the detection system that was identical to the
18 conventional LSFM or epifluorescence microscopy. The lateral resolution of the
19 micro-axicon based LSFM depended on the wide-field detection system and was
20 the same as the conventional LSFM or epifluorescence microscopy, while the
21 axial resolution depended on the thickness of the light sheet generated by the

1 Bessel-Gauss beam and was dramatically improved compared to the
2 epifluorescence microscopy. The most outstanding feature of our micro-axicon-
3 based LSFM eliminated the bulky objective lenses to produce the non-diffractive
4 Bessel beam and potential light sheet. It provides a significantly higher system
5 flexibility for sample installation that could find potential applications in
6 intracorporeal or hazardous environments. The results are published as the
7 journal article, Li *et al.*, 2021.

8

9 **Collimated Beam Tilted Angle Measurement with** 10 **Diffractive Axicon**

11 I fabricated a rGO thin film diffractive axicon consisting of the concentric rings
12 of equal ring width and pitches of 33.132 μm with the FsLDS system. The
13 alternate ring patterns were fabricated in the TPR and ABL regimes. This
14 diffractive axicon was designed to simulate the commercial 1-inch sized axicon
15 AX251-A with a base angle of 1° . The performance that transforms the incident
16 collimated Gaussian beam into a Quasi-Bessel beam has been confirmed to
17 maintain the less-diffractive beam size of 26.27 μm along the propagation
18 distance of 24.0 mm, and the cross-sectional profiles were conformed to the result
19 of axicon AX251-A.

20 The tilted diffractive axicon with respect to the incident Gaussian source
21 showed that it transformed the beam into the beam profiles with high-order Bessel

1 functions, that it presented a regular pattern evolution versus the two crucial
2 parameters: tilted angle and the propagation distance. In this research, I
3 particularly investigated the beam transformation of tilted diffractive axicon
4 (analogous to refractive axicon of base angle 1° and the beam profile evolution
5 versus the two parameters). The transformation and pattern evolution have been
6 recorded and analyzed for the detection of the tilt angle between the beams and/or
7 object surfaces. The manuscript of this work is under preparation and will be
8 published in an article.

9

10 **6.2 Recommendation for Future Work**

11 I am proud of the research results shown in this thesis. Still, I am keen to future
12 improve the system and promote the results to more practical uses.

13

14 **6.2.1 Development of Femtosecond Laser Direct Writing** 15 **System**

16 To date, I have successfully fabricated the 3D and thin-film microstructures with
17 both lateral and axial resolution in micro/sub-micron scale with different
18 materials. The femtosecond laser additive manufacturing system can be further
19 improved considering the following parameters:

20 a) Adjustable pulse power level and scanning speed: there is a trade-off

1 between the material's energy absorption and polymerization resolution versus
2 the fabrication time period. The fs-laser power level and scanning speed were
3 selected and set to constant and fit the reasonable resolution and fabrication time
4 cycle. I propose the adjustable laser power and the scanning speed in the future
5 fabrication, that the interior portion of the designed structures can be scanned
6 with higher pulse energy to shorten the printing duration, while the outer portion
7 and edges and corners to be scanned carefully with minimized energy and mild
8 speed to achieve higher geometric accuracy and surface quality. The automated
9 control of the scanning procedure with proper pulse energy and scanning speed
10 requires further investigation and updates.

11 b) The mass production of repeated patterns and arrays: the mass and fast
12 production of duplicated patterns and lens arrays are of great interest in the
13 community. I propose to realize such fabrication by split the focal spot of the
14 femtosecond laser into multiple spots, and thus one scanning process produces
15 duplicated patterns, forming some gratings or arrays with real applications. In this
16 manner, the fabrication time period for the devices dramatically reduces and it
17 ensures the identical properties of each of the duplicated patterns.

18

19 **6.2.2 Development of Light Sheet Fluorescence**

1 **Microscopy with Micro-Axicon**

2 There have successfully demonstrated the detection and imaging of the micro-
3 beads and Hela cells with the micro-axicon-based light sheet fluorescence
4 microscopy. Note that I performed the illumination with a pencil-like Bessel-
5 Gauss beam. In the further development of the LSM, I propose that the scanned
6 light-sheet can be achieved by installing the micro-axicon tipped fiber on high-
7 speed ultrasonic actuators, for example, V-522 High-Dynamics PIMag® linear
8 stage, which can scan the beam in the detection focal plane with 250 mm/s
9 velocities, and 10 nm resolution, across a range of 5 mm. In this manner, a
10 physical beam sheet can be produced, and thus the layered images can be
11 generated with faster speed and smooth illumination.

12

13 **6.2.3 Development of Beam Tilt Angle Measurement with** 14 **Diffraction Axicon**

15 I have successfully demonstrated the transforming power of the diffractive axicon
16 and investigated the tilted angle effect on the transformance.

17 To further detect the finer tilted angle between the beams and some object
18 surfaces, the system can be further improved with the following considerations:

19 a) Diffractive axicon with finer ring width and pitches: based on the

1 calculation shown in Chapter 5, diffractive axicon with finer ring width and
2 pitches is analogous to the refractive axicon with a larger base angle and sharper
3 apex angle, which generates Bessel beams having smaller spot size. A diffractive
4 axicon with finer ring width and pitches is more sensitive to the changing of tilted
5 angle and gives minimized detectable changes.

6 b) Property study of the GO/rGO film: the degree of reduction of the rGO
7 film and its physical properties should be investigated. X-ray photoelectron
8 spectroscopy (XPS) can identify the ratio of carbon, oxygen, hydrogen, and other
9 elements in compounds. Raman spectroscopy will be applied to identify
10 molecules, chemical bonding, and intramolecular bonding. X-ray diffraction
11 (XRD) and Fourier transform infrared spectroscopy (FTIR) etc. will be also
12 applied for structural analysis and the functional groups of the compounds. It is
13 important to analyze the relationship between fabrication conditions and the
14 degree of reduction of rGO as well as the optical and other physical properties of
15 rGO thin films and lenses.

16 c) Multi-order Fresnel zone plate: there has been shown that rGO thin film
17 can be fabricated with the three regimes: LTPR, TPR, and ABL, and therefore,
18 gives multi-ordered or continuously varying opacity or transparency. It gives
19 possibilities to produce multi-order diffractive lenses with the ring of discrete
20 transmittances and continuous Fresnel zone plates. It could improve the
21 efficiency and the propagation properties of the generated beams, in terms of the

1 beam size, propagation distance, SNR, and smoothness of beam profiles, etc.

2

Publication

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1 **Appendix**

2 **A.1 Side view profile of FsLDW fabricated structure**

3
4 Figure S.1 Side view profile of the FsLDW micro-axicon

5 The side view profile reveals that the FsLDW micro-axicon possesses a good
6 geometry as design and fine surface property for the optical usage. The top view
7 optical microscope and SEM images are provided in the Figure 32(d). The
8 transformation performance is shown in the Chapter 4.2. Transformation of
9 Gaussian beam to Bessel-Gauss beam by a micro-axicon. The FsLDW technique
10 and the fabricated micro-axicon perform well in optical applications.

11

12

A.2 Performance comparison of micro-axicon and commercial axicon

Figure S.2 Comparison of the transformations from Gaussian beam to Bessel-Gauss beam by the FsLDW micro-axicon and a commercial 1" axicon (Thorlabs, AX2520-A).

The authors tested the Bessel-Gauss beam generated by the FsLDW micro-axicon and a commercial 1-inch sized axicon (Thorlabs, AX2520-A, $n = 1.46$). Both the resulted beams perform well matched with the theoretical results in terms of the central beam radius given by Equation:

$$r_0 = 2.405 / k_r$$

and the depth-of-focus:

$$Z = \frac{\omega}{\tan[\sin^{-1}(n_{axicon} \times \sin \alpha) - \alpha]} - height_{axicon} .$$

A collimated Gaussian beam of beam size of an aperture 100 μm is transformed by the micro-axicon into a Bessel-Gauss beam of beam size $2r_0$ is calculated to be 1.85 μm and measured as $2.3 \pm 0.25 \mu\text{m}$ with an error rate of 24.4%. The depth of focus is calculated to be 202.8 μm and measured to be 160 μm with an error rate of -21.1%.

The Gaussian beam of beam size 1040 μm is transformed by the commercial 1'' axicon into a Bessel-Gauss beam of beam size calculated to be 2.36 μm and measured to be $1.54 \pm 0.23 \mu\text{m}$ (error rate = -34.6%) and a depth of focus calculated 2774 μm and measured 2200 μm (error rate = -20.7%).

A.3 Configuration of biological imaging

Figure S.3 Schematic showing the illumination arrangement for the imaging of the HeLa cells cultured on glass slide

The glass slide with HeLa cells is installed parallel to the incident Bessel beam and the Gaussian beam axis. The sample was installed such that the glass had little interaction with the incident illumination Bessel-Gauss beam. In the case of the Gaussian beam, the later parts of the beam after the focus does interact with the glass slide.