UNIVERSITI PUTRA MALAYSIA

Z-SCAN TECHNIQUE WITH HIGH REPETITION RATE FEMTOSECOND LASER FOR NONLINEAR OPTICAL PROPERTIES MEASUREMENT

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By

MOHD SHAHNAN BIN ZAINAL ABIDIN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Doctor of Philosophy

October 2014

To my beloved wife, kids and parents.

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

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MOHD SHAHNAN BIN ZAINAL ABIDIN

October 2014

Chair : Ahmad Shukri Muhammad Noor, PhD
Faculty : Engineering

Nonlinear optical properties are crucial in determining the behavior of the optical material under the intense laser beam. Z-scan method is a simple technique to measure nonlinear refraction and absorption with high degree of sensitivity and non-complex arrangement setup. Ultrafast laser technology benefits the Z-scan method by providing high nonlinearity condition within the material. Nevertheless, most of today's ultrafast lasers are designed with high repetition rate (HRR) pulse trains which is known able to trigger thermal lensing effect.

The lensing formation in Z-scan measurement corrupts the pure nonlinear response thus exhibits invalid results. Thermal lensing effect by the HRR laser is originated by the accumulated heat across the pulse trains due to the significantly long thermal diffusion time of the material in comparison to the very short HRR pulse separation. Optical chopper deployment in Z-scan measurement limits the material time exposure to the HRR beam and eliminates the lensing effect with the optimized chopper frequency. However, the optimization comes with a fixed chopper duty cycle, typically equal opening and blanking period. Therefore, applying duty cycle variation on top of the chopper frequency would reveal a new working range for the Z-scan to obtain accurate measurements without the thermal lensing influence. This research reports the Z-scan measurement with 780 nm HRR femtosecond beam on the nonlinear material AC-39.

The experiment is performed with the adoption of the chopper frequency and duty cycle variation to minimize the thermal lensing effect by the precisely control the exposure time on AC-39. The modulated HRR beam Z-scan is carried out over the modulation frequency and duty cycle variation by evaluating the change of the peak and valley transmittance. For a 50% fixed duty cycle, 500 Hz of optimized minimum chopper frequency is achieved. The minimum chopper frequency is reduced further by adopting 10 and 25% duty cycle. It is deduced that this optimization is obtained by keeping the HRR beam time exposure on AC-39 material well below its thermal diffusivity time, thus seizes the thermal lensing build up formation before the next duty cycle. The minimum chopper frequency is correctly predicted by associating the chopper duty cycle factor, F with the material thermal diffusivity time, t_c. Additional frequency optimization is achieved by considering the stable peak-valley transmittance difference over the frequency variations. The technique of finding the optimized minimum chopper frequency leads to the thermal diffusivity determination of the sample material for the unknown thermal diffusivity time. The optimization opens up for a potential of low operational frequency and offers a simple and straightforward solution and implementation in Z-scan technique of nonlinear optical properties measurement with high repetition rate femtosecond laser.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

TEKNIK Z-SCAN DENGAN LASER FEMTOSAAT BERKADAR PENGULANGAN TINGGI UNTUK PENGUKURAN SIFAT TIDAK LINEAR OPTIK

Oleh

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Sifat tidak linear optik adalah penting dalam menentukan kelakuan bahan optik di bawah ketumpuan sinaran laser. Kaedah Z-scan adalah teknik yang mudah untuk mengukur penyerakan dan penyerapan tidak linear dengan tahap kesensitifan yang tinggi dan susun atur eksperimen yang tidak kompleks. Teknologi laser ultra pantas memberikan faedah kepada kaedah Z-scan dengan menyediakan keadaan tidak linear yang tinggi di dalam sesuatu bahan. Walaubagaimanapun, kebanyakan laser ultra pantas pada masa kini direka dengan rentetan denyutan berkadar pengulangan tinggi (*high repetition rate* – HRR) yang diketahui mampu mencetuskan kesan kanta terma.

Pembentukan kanta terma di dalam pengukuran Z-scan memusnahkan tindak balas tidak linear yang tulen oleh yang demikian mempamerkan keputusan yang tidak sah. Kesan kanta terma oleh laser HRR bermula daripada pengumpulan haba di sepanjang rentetan denyutan disebabkan oleh masa penyerakan haba yang ketara panjang berbanding dengan masa pemisah antara denyutan HRR yang amat pendek. Penggunaan pencincang optik di dalam pengukuran Z-scan menghadkan masa pendedahan bahan kepada sinaran laser HRR dan menghilangkan kesan kanta dengan frekuensi pencincang yang optimum. Namun, pengoptimuman ini hadir dengan kitaran tugas pencincang yang tetap, kebiasaannya dengan tempoh bukaan dan halangan yang sama. Oleh itu, dengan meletakkan kitaran tugas yang pelbagai disamping frekuensi pencincang boleh mendedahkan lingkup kerja baru untuk Z-scan dalam mendapatkan pengukuran yang tepat tanpa pengaruh kanta terma. Kajian ini melaporkan pengukuran Z-scan dengan sinaran laser femtosaat 780 nm HRR ke atas bahan tidak linear AC-39.

Eksperimen ini dilakukan dengan penggunaan frekuensi dan kitar tugas pencincang yang pelbagai untuk mengurangkan kesan kanta terma dengan mengawal dengan jitu masa pendedahan ke atas AC-39. Z-scan dengan sinaran laser termodulat HRR dijalankan pada kepelbagaian modulasi frekuensi dan kitar tugas dengan menilai perubahan puncak dan lembah kehantaran. Bagi kitar tugas tetap 50%, frekuensi pencincang teroptimum paling rendah 500 Hz dicapai. Frekuensi tersebut direndahkan lagi dengan penggunaan kitar tugas 10 dan 25%. Disimpulkan bahawa pengoptimuman ini diperolehi dengan memastikan masa pendedahan sinaran laser

HRR ke atas bahan AC-39 berada dibawah masa penyerakan haba, oleh yang demikian kenaikan dalam pembentukan kanta haba dihentikan sebelum kitar tugas seterusnya. Frekuensi pencincang yang minimum di ramalkan dengan betul dengan mengaitkan faktor kitar tugas, F dan masa penyerakan haba bahan, t_c. Frekuensi optimum tambahan dicapai dengan mengambil kira beza kehantaran puncak dan lembah disepanjang kepelbagaian frekuensi. Teknik pencarian frekuensi minimum pencincang teroptimum membuka jalan kepada penentuan penyerakan haba bahan sample bagi masa penyerakan yang tidak diketahui. Pengoptimuman frekuensi ini membuka ruang kepada potensi frekuensi operasi yang rendah dan menawarkan penyelesaian dan pelaksanaan teknik Z-scan yang mudah dalam pengukuran sifat tidak linear optic dengan laser femtosaat berkadar pengulangan tinggi.

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I certify that a Thesis Examination Committee has met on 16 October 2014 to conduct the final examination of Mohd Shahnan Bin Zainal Abidin on his thesis entitled “Z-Scan Technique with High Repetition Rate Femtosecond Laser for Nonlinear Optical Properties Measurement” in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctor of Philosophy.

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This is to confirm that:

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

BS	Beam Splitter
CA	Closed Aperture
CCD	Charge-coupled Device
CW	Continuous Wave
DCM	Dicholoromethane
DFG	Difference Frequency Generation
DFWM	Degenerated Four Wave Mixing
EDFL	Erbium Doped Fiber Laser
ESU	Electrostatic Unit
FWHM	Full Wave Half Maximum
FWM	Full Wave Maximum
HRR	High Repetition Rate
ISO	International Organization for Standardization
KDP	Kalium Dihydrogen Phosphate
KTP	Kalium Titanyl Phosphate
LRR	Low Repetition Rate
MKS	Meter-Kilogram-Second
NDF	Neutral Density Filter
NDFWM	Nearly Degenerated Four Wave Mixing
NLRI	Nonlinear Refractive Index
OA	Open Aperture
OPA	Optical Parametric Amplification
OPO	Optical Parametric Oscillation
PD	Photodetector

PPLN	Periodically Poled Lithium Niobate
SESAM	Semiconductor Saturable Absorber Mirrors
SFG	Sum Frequency Generation
SHG	Second Harmonic Generation
SNR	Signal to Noise Ratio
THG	Third Harmonic Generation
TL	Thermal Lensing
TPA	Two-photon Absorption
TPACS	Two-photon Absorption Cross Section
TPE	Two-photon Excitation
XPM	Cross Phase Modulation

CHAPTER 1

INTRODUCTION

1.1 Overview

Since the laser discovery by G. Gould in 1959 [1] and later a functional laser by T. Maiman [2], laser related applications have enormously multifold covering technology fields such as spectroscopy [3], sensors [4], computing storage [5] and telecommunication [6]. In its early stage, the laser produced fairly low output intensity [2, 7] with a limited operational wavelength. As the laser technology advances, multi-kilowatt output power has been reached [8] with vast wavelength selections, thanks to the maturity of laser gain material [9–11] and enhanced free space and optical fiber coupling techniques [12]. These improvement allows better coupling efficiency for laser amplification and wide spectrum tuning ability [13]. To date, remarkably high power output comes with high repetition rate (HRR) femtosecond laser pulse in which delivers applications improvement such as real time spectroscopy observation [14] and 3D optical storage [15]. The HRR laser is advantageous due to its pulse stability for constant power response over time and good signal-noise ratio (SNR) for excellent differentiation between pulse and noise level.

The laser invention and development concurrently opens up for nonlinear optics as a new research field. Nonlinearity in a material becomes significant due to intense laser beam incidence on the sample within the small area of interaction thus creating the material response such as wavelength shift, density and absorption properties change. The properties changes are beneficial for applications such as micro-machining, laser ablation [16] and nonlinear spectroscopy [17] as been shown by recent works in high power fiber laser whereas a 0.1mJ of laser energy at 10 MHz pulse train was produced with cavity-enhanced high harmonic generation [18].

In nonlinear optics research field, the nonlinear material properties characterization is essential parameter to be determined. The characterization is performed to classify the material suitability in the related applications. For example, a semiconductor saturable absorber mirrors (SESAMs) is known for its properties with low linear and nonlinear absorption acted as nonlinear mirror for the generation of ultrashort semiconductor laser [19]. For frequency doubling conversion laser, the nonlinear crystal material with known effective nonlinear coefficient is important for the phase-matching configuration to convert the pump energy to the generated second harmonic wave [20].

Numerous measurement techniques were adopted to determine the material nonlinear properties. Interferometric [21], degenerated and nearly degenerated four wave mixing method [22] are sensitive to the measurement phase changes but its complex experimental setup is discouraging. Beam distortion measurement method is insensitive yet require detailed wave propagation analysis [23]. Z-scan technique is a preferred method over previously mentioned techniques to determine nonlinear optical properties, thanks to its high sensitivity and simple experimental setup. Discovered by Sheik-Bahae and co-workers [24], the Z-scan technique measures nonlinear refractive index (NLRI), n_2 and two-photon absorption cross section (TPACS), σ_2 by translating the material along the intense focusing beam vicinity and monitoring its self-focusing and defocusing behavior. The technique reveals both the sign and magnitude of the nonlinear properties by evaluating the transmittance level through an aperture at far field.

The advancement of ultrafast laser technology has benefited the Z-scan measurement technique by providing high peak power to induce the desirable nonlinearity condition in the material. Previously, low repetition rates Q-switch and Gain-switch pulse lasers have been extensively adopted in the Z-scan measurement from a few Hz up to tens of kHz [25–27]. However most of today's high repetition rate (HRR) lasers have been known to trigger thermal lensing effect within the Z-scan measurement due to heat accumulation across the pulse trains via the skeletal libration and vibrations process [28].

Thermal lensing effect is realized as the incident laser beam energy is converted into the heat within the material contact area of the incidence beam. With the maximum heat is at the beam center, it is radially distributed outward and consequently changes the material density thus the refractive index of the material. The index change effectively forms a thin lens that alters the divergence and convergence manner of the next incidence beam pulses. The lensing effect corrupts the Z-scan measurement by overwhelming the purely electronic nonlinear Kerr response therefore leading to an erroneous result.

The thermal lensing effect is formed due to the very short HRR laser pulse separation time in comparison to the considerably long thermal diffusivity decay time [28]. The generated heat by each of the individual pulse is not properly diffused out from the laser incident area before the next consecutive pulse. This simply builds up the heat across the pulse trains until the thermal lensing dominates. For MHz repetition rate laser, the condition can be easily met under the high beam intensity. On top of the short pulse separation, the heat accumulation may also be contributed by thermo-acoustic effect as the generated heat travels at a material sound speed within the incidence beam area, typically for nanosecond pulse [29, 30]. If the time taken for the heat to travel out of the incidence beam area is longer that the pulse separation time, the accumulated heat allows the thermal lensing to set in. On the other hand, HRR pulse duration may give rise to the thermal effect via the excited non-radiative skeletal motion [31], profoundly for nanosecond pulse [30–32], whilst pico- and femtosecond pulse is less influencing.

The adoption of low repetition rate laser (LRR) is a direct solution to avoid thermal lensing effect by the HRR laser within the Z-scan measurement. There were several researchers incorporating a pulse picker device that selects some pulses to be omitted thus obtaining the low repetition rate condition [33, 34]. However the pulse picker implementation has added up for the experimental complexity as the modification of the ultrafast laser must be done electrically. Falconieri *et. al* and several other researchers have suggested a solution of time modulating the HRR laser pulse train with an optical chopper [35–38]. This is the desired technique as the chopper can be inserted independently from the ultrafast laser configuration. The chopper introduces a periodic opening and blanking time of the HRR beam depending on the chopper applied frequency. The chopper opening width determines the material exposure to the HRR beam, giving a minimized thermal lensing effect frequency range thus allowing for the nonlinear properties measurement recovery [35, 39]. A modified Z-scan technique known as time-resolved Z-scan measures the transmittance at various temporal points during the opening time allowing for separation of the slow thermal component from the fast nonlinearity response [40, 41].

1.2 Problem Statement

Despite these extensive works to optimize the chopper frequency for precise Z-scan measurement with HRR femtosecond laser, they rely on a fixed chopper wheel duty cycle only [36, 38, 39]. As a matter of fact, the opening time duration varies with the chopper wheel frequency and duty cycle. Therefore the optimized frequency with the duty cycle variation would opens up for the improved frequency range without thermal lensing influence. In this thesis, we investigate a closed and open aperture Z-scan with the modulated HRR femtosecond laser to minimize the thermal cumulative effect in the sample material. In order to avoid the influence of the thermal effect within the Z-scan measurement, the chopper opening and blanking time optimization is determined to reveal the chopper optimized operational frequency range. The analysis of the Z-scan peak and valley transmittance level throughout the frequency and duty cycle variations is used to analytically predict the minimum operational chopper frequency.

1.3 Research objectives

The objectives of this research work are as the following:

a. To investigate the nonlinear optical properties of a material with the influence of thermal lensing effect by the HRR femtosecond laser.
b. To optimize the chopper frequency and duty cycle in minimizing the thermal lensing effect in Z-scan measurement with HRR femtosecond laser
c. To determine the minimum obtainable chopper operational frequency with the minimized thermal lensing effect.

1.4 Research scope

Figure 1.1 shows the scope of the conducted research. The nonlinear optical properties measurement is specifically directed into the 3rd order nonlinearity measurement and characterization. The incorporation of Z-scan technique particularly with ultrafast femtosecond laser is desirable over other nonlinear characterization technique for the laser ability to provide high beam intensity at lower average power output. Single beam Z-scan technique with HRR femtosecond laser source is preferable over other Z-scan technique variants for the simplicity purpose of the research work and the fact on its ability to generate thermal lensing effect in the material. The research emphasization is given to the thermal accumulation within the sample material by the HRR laser, the laser pulse duration and the material thermal diffusivity relationship with the optical chopper frequency and duty cycle variation.

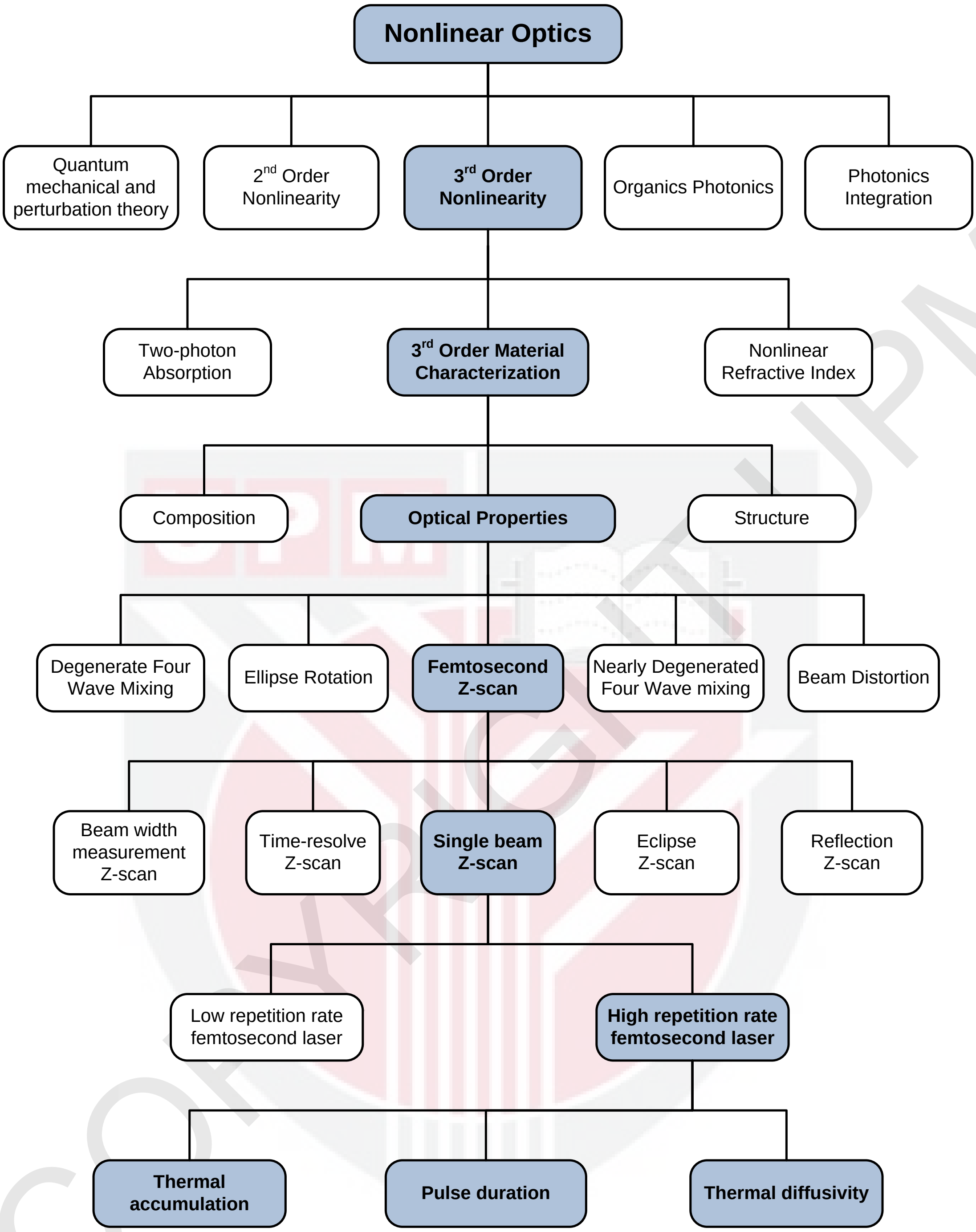

Figure 1.1. Research scope chart

1.5 Methodology flow chart

Figure 1.2 summarizes the brief methodology to achieve the objective of the the thesis. In this research work, the thermal lensing effect in the Z-scan nonlinear optical properties measurement is formed in the sample material without modulating the HRR femtosecond laser beam. Next, an optical chopper is deployed to time-modulate the beam in two optical chopper conditions. Firstly by a variable chopper frequency and fixed duty cycle, and secondly by a variable chopper frequency and duty cycle. Subsequently, the thermal lensing minimization achieved by both conditions is analyzed based on the optimization of the chopper frequency range, minimum obtainable chopper frequency and the prediction of thermal diffusivity time of the sample material.

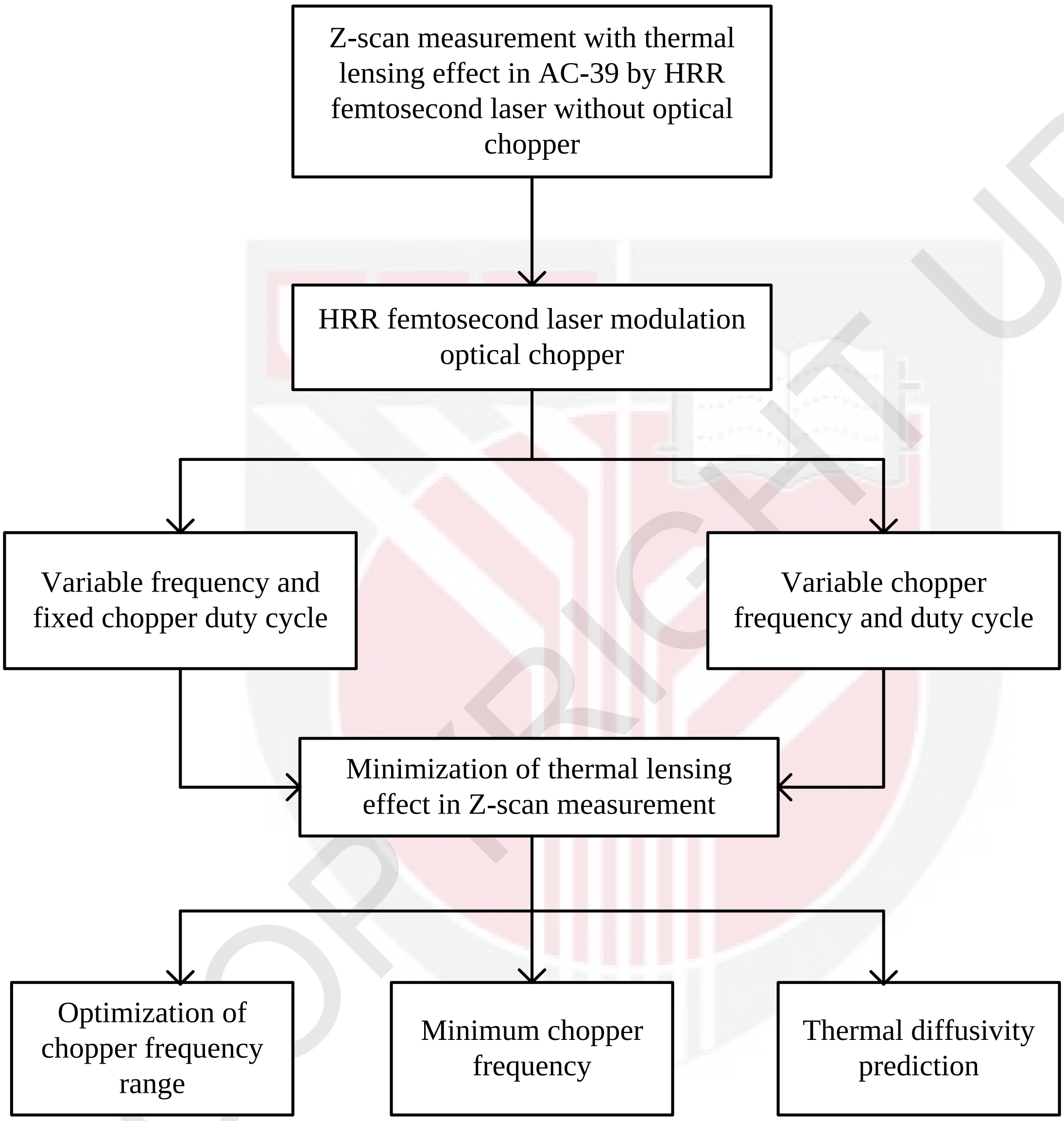

Figure 1.2. Methodology flow chart

1.6 Thesis organization

Generally, the thesis is divided into seven main chapters. Chapter 1 gives an overview of the research topics under study, problem statement, research objectives and scope. Chapter 2 critically discusses the nonlinear optics fundamentals towards the understanding of the main topics as well as the current available techniques of nonlinear optical properties measurement. Chapter 3 gives a concentration on synthesization and characterization of the nonlinear material and Z-scan experimental arrangement. The component and device characterization is also elaborated to obtain precise Z-scan measurement. Chapter 4 specifically emphasizes on the thermal lensing formation within the Z-scan measurement. Chapter 5 reveals the applied thermal management by an optical chopper that leads to the measurement optimization. Chapter 6 looks into further optimization provided by the duty cycle variations. Finally, Chapter 7 summarizes and concludes all findings of the research and suggests related future extension works.

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