NONLINEAR OPTICAL STUDIES OF ORGANIC COMPOUNDS USING Z-SCAN TECHNIQUE AT 532 NM

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DEDICATION

The most beautiful thing in this world is to see your parents smiling and knowing that you are the reason behind that smiles

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ABSTRACT

This research reports on the third order nonlinear optical (NLO) attributes for both nonlinear refractive indices, n_2 and nonlinear absorption coefficient, β of various type of organic compounds, namely α-Mangostin (α-MG), Malachite green (MG), Light Green SF Yellowish (LGSF), Coumarin 500 (C500) and Ponceau BS (PBS). The purpose of this study are comprised by optimizations of Z-scan measurements in order to determine the nonlinear refraction (NLR) and nonlinear absorption (NLA) behaviours of each compound mentioned and also to detect the optical limiting (OL) threshold for further applications. A single-beam Z-scan technique was utilized for the nonlinearity measurement. This indicated the NLA phenomena paralleled with the laser excitations wavelength, ($\lambda = 532\ nm$) The existence of vital vibrational π-bonds for intermolecular charge transfer (ICT) of the compounds were confirmed via Fourier Transform Infrared (FTIR) Spectroscopy. The vibrational π-bonds are responsible for the appearance of remarkable responds towards nonlinearity under intense laser power. To calculate the values of nonlinearity, Thermal Lens Model (TLM) fitting curve was used due to the significant contribution of heat in continuous-wave (CW) laser setup. For instance, MG, LGSF and PBS chromophores possess both NLR and NLA from closed and open aperture signals, respectively. It was recorded that the magnitude of third order optical susceptibility, $|\chi^3|$ of the chromophores were in the order of 10^{-6} (MG), 10^{-6} (LGSF) and 10^{-5} (PBS). On the other hand, α-MG and C500 revealed closed-aperture signals only with the value of $|\chi^3|$ in the order of $10^{-11}$and 10^{-8} respectively. The power range used in the experiment varies between each sample starting from 0.10 W (min.) to 2 W (max.) depending to the positive respond shown in specific samples. As a whole, all samples exhibited NLO properties under intense laser power. The open aperture signals appeared in MG, LGSF and PBS chromophore as a reverse saturable absorption (RSA). Thus, MG, LGSF and PBS chromophores OL threshold is found at 0.567, 1.154 and 1.124 W, respectively and it can be further developed as optical limiter for optical safety purposes.

ABSTRAK

Penyelidikan ini melaporkan tentang sifat optik tak linear tertib ketiga bagi kedua-kedua indeks pembiasan tak linear, n_2 dan pekali serapan tak linear, β untuk pelbagai jenis sebatian organik seperti α-Mangostin (α-MG), Hijau Malekit (MG), Hijau Muda SF Kekuningan (LGSF), Kumarin 500 (C500) dan Ponceau BS (PBS). Tujuan kajian ini terdiri daripada pengoptimuman pengukuran imbasan-Z untuk menentukan tingkah laku pembiasan tak linear (NLR) dan penyerapan tak linear (NLA) bagi setiap sebatian yang disebutkan dan juga mengesan ambang had optik (OL) untuk aplikasi selanjutnya. Teknik alur-tunggal imbasan-Z digunakan untuk pengukuran ketaklinearan. Ini menunjukkan fenomena NLA selari dengan panjang gelombang pengujaan laser (λ = 532 nm). Kewujudan ikatan-π getaran vital bagi cas perpindahan antara molekul (ICT) sebatian tersebut disahkan melalui Spektroskopi Inframerah Transformasi Fourier (FTIR). Ikatan--π getaran adalah bertanggungjawab kepada kemunculan tindak balas yang luar biasa terhadap ketaklinearan di bawah kuasa laser yang amat tinggi. Untuk menghitung nilai ketaklinearan, penyuaian lengkung Model Kanta Terma (TLM) digunakan oleh kerana sumbangan haba yang ketara dalam penyediaan laser gelombang selanjar (CW). Contohnya, kromofor MG, LGSF dan PBS masing-masing mempunyai NLR dan NLA dari isyarat bukaan tertutup dan terbuka. Telah dicatatkan bahawa magnitud kerentanan optik tertib ketiga, $|\chi^3|$ kromofor berada dalam lingkungan 10-6 (MG), 10-6 (LGSF) dan 10-5 (PBS). Sebaliknya, α-MG dan C500 mendedahkan isyarat bukaan-tertutup sahaja dengan nilai $|\chi^3|$ masing-masing dalam lingkungan 10^{-11} dan 10-8. Julat kuasa yang digunakan dalam eksperimen ini adalah berbeza antara setiap sampel, bermula dari 0.10 W (min.) hingga 2.00 W (maks.) bergantung pada tindak balas positif yang ditunjukkan dalam sampel yang tertentu. Secara keseluruhanya, semua sampel mempamerkan sifat NLO di bawah kuasa laser yang amat tinggi. Isyarat bukaan terbuka muncul dalam kromofor MG, LGSF dan PBS sebagai penyerapan tepu songsang (RSA). Maka, ambang OL kromofor MG, LGSF dan PBS masing-masing didapati pada 0,567, 1,154 dan 1,124 W dan ianya dapat dikembangkan sebagai pengehad optik untuk tujuan keselamatan.

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LIST OF ABBREVIATIONS

NLO	-	Nonlinear Optical
NLR	-	Nonlinear Refraction
NLA	-	Nonlinear Absorption
CW	-	Continuous-Wave
NIR	-	Near-Infrared
DPSS	-	Diode-Pumped Solid-State
TLM	-	Thermal Lens Model
OL	-	Optical Limiting
α-MG	-	Alpha Mangostin
m.p.	-	Melting point
MW	-	Molecular Weight
MG	-	Malachite Green
DMSO	-	Dimethyl Sulfoxide
LGSF	-	Light Green SF Yellowish
OKE	-	Optical Kerr Effects
RSA	-	Reverse Saturable Absorption
TPA	-	Two Photon Absorption
ESA	-	Excited State Absorption
RE	-	Real
IM	-	Imaginary
DOF	-	Depth of Focus
SA	-	Saturable Absorption
P1	-	Phase One
P2	-	Phase two
P3	-	Phase Three
P4	-	Phase Four
PD	-	Photodiodes
ICT	-	Intermolecular Charge Transfer
TLE	-	Thermal Lens Effects

LIST OF SYMBOLS

$\chi^{(3)}$	-	Third order susceptibility
n_2	-	Nonlinear refractive index
β	-	Nonlinear absorption coefficient
λ	-	wavelength
$\chi^{(n)}$	-	General order of optical susceptibility
$\chi^{(1)}$	-	First order susceptibility
$\chi^{(2)}$	-	Second order susceptibility
ε_o	-	Vacuum permittivity
E	-	Electric field
n_o	-	Weak-field refractive index
I	-	Intensity of light
α_o	-	Linear absorption coefficient
σ_{exc}	-	Excited state absorption cross section
σ_g	-	Ground state absorption cross section
ω_o	-	Beam waist
Θ	-	Divergence angle
z_r	-	Rayleigh length
C	-	Concentration
r_o	-	Aperture radius
L	-	Length
v	-	Velocity
s	-	Displacement
t	-	Time
c	-	Speed of light
f	-	Focus length
L_{eff}	-	Effective path length
T_N	-	Normalized transmittance
$\hbar$	-	Reduced Planck constant

CHAPTER 1

INTRODUCTION

1.1 Introduction

Since the discovery of laser in 1960s by Theodore H. Maiman at Hughes Research Laboratories [1] formulated rigorous theoretical work by Charles Hard Townes and Arthur Leonard Schawlow, nonlinear optical (NLO) attributes of various compounds have seen to become reachable to date [2-5]. NLO is a branch of optics that explains the behaviour of intense light to interact with matter [6]. Technological advancement has created various necessities that press the demand in order to complement the needs especially related to the nonlinear phenomenon, e.g. self-focusing [7], Kerr-lens mode-locking [8], self-phase modulation [9], multiphoton absorptions [10], optical soliton [11], modulational instability [12], etc. The modern applications such as ultra-high definition (UHD) and high definition (HD) display [13, 14], great-performance transmitting optical fibres [15-17], and laser-manufactured in medicine [18-21] are seen more accessible to date. In order to find novel materials with relatively huge value of third-order susceptibility, $\chi^{(3)}$ continuous efforts globally have been made to achieve the modern lifestyles [22-26].

Materials like metal complexes [27, 28], nanocomposites [29, 30], organic compounds [31, 32], hybrid materials [33, 34], etc., are sought and utilized for the excellent outcomes. However, off those, single organic compounds were seen as potential candidates to exhibit the demand mentioned. Organic molecular framework is highly favoured over the inorganic ones [35]. Organic material provides the structural flexibility and ability to maximize NLO responses by varying the substituents and respective positions over the molecular framework [36]. As a result, a series of organic compounds were chosen in this study through its unique chemical structures systematically organize by aromatic hydrocarbons which later show prominent NLO attributes. Some of these compounds are not yet reported by any individual within NLO field as it remains hidden. Thus, this is a golden opportunity to unleash the hidden attributes as these organic compounds hold.

Furthermore, this research was done by proposing a Z-scan technique where Z-scan was first discovered in 1989 by M. Sheik-Bahae et al. at Centre for Research in Electro-Optics & Lasers, Florida [37]. It is a technique that is well known for its simplicity and accuracy to obtained both values of nonlinear refraction (NLR) and nonlinear absorption (NLA) at once [38, 39]. In fact, there are various methods to determine nonlinearities of material namely nonlinear interferometry [40], degenerate four wave-mixing [41], ellipse rotation [42] and beam distortion method [43]. However, those methods are rather complicated and insensitive as compared to Z-scan technique.

1.2 Problem Statement

In the recent past, rapid technological advancements in optoelectronic have placed a great demand on the development of nonlinear material that possesses large nonlinearities and satisfying various technological requirements for photonic device applications. Due to this reason, the NLO properties of various solid-state, inorganic, and organic materials have been, and are being, extensively investigated for such purposes. However, majority of the materials stated above suffer from poor photo-thermal stability, low dissolvability, and complicated preparation virtue. Apart from it, organic compounds are considered as promising candidates due to their large optical nonlinearities and fast response, applicability over a wide range of visible spectral region, photochemical stability, and high damage thresholds.

For any NLO materials investigated, there is a need to explore their properties through the determination of NLR, and NLA magnitudes in order to establish their potential applications in the appropriate field of interest. In particular, there are less critical appraisals on the group of organic compounds namely α-Mangostin, Malachite Green, Light green SF Yellowish, Coumarin 500 and Ponceau BS for this matter. Their NLO potential is yet to be explored in detail leaving significant insight on their respective third-order NLO behaviours. Therefore, this study was proposed with the urged to determine the sign and magnitude of its nonlinear refractive index, n_2, nonlinear absorption coefficient, β, $\chi^{(3)}$ as well as the optical limiting (OL) ability of such organic compounds mentioned under intense continuous-wave (CW) laser illumination,

1.3 Research Goals

The main objective of this study is to determine the third-order NLO response of various organic compounds at 532 nm via Z-scan technique. In order to achieve this goal, there are specific objectives need to be achieved:

1. To align and optimize the Z-scan setup with respect to its open and closed-aperture responses.
2. To determine and analyse the NLR and NLA behaviours of organic samples including its n_2, β, and magnitude of $|\chi|^{(3)}$.
3. To determine the OL threshold of such organic compounds under intense CW laser illumination subjected to the presence of NLA.

1.4 Scope of Study

This study focuses on single organic compounds and its interaction with different laser power. The laser excitation source was chosen at λ = 532 nm using CW diode-pumped solid-state (DPSS) laser. The organic compounds samples were purchased from Sigma-Aldrich (Ponceau BS, Malachite Green, Coumarin 500, Light green SF Yellowish) without further purification while α-Mangostin sample was synthesized in the laboratory. A Z-scan method was used as the primary experimental setup to operate and determine the value of NLO attributes in term of NLR and NLA for each compound. Specific values of n_2, β and $\chi^{(3)}$ were calculated based on the theoretical Thermal Lens Model (TLM) framework considering the prominent thermal effect arise from the CW laser source. The OL properties of samples were examined in the range of 0 to 2.0 W.

1.5 Significant of Study

NLO materials play a pivotal role in the future evolution of nonlinear optics and its impact in technology and industrial applications are excellent. In the field of optics, nonlinear effects became a subject of interest only after the invention of the laser. Since then, nonlinear optics had become a rapidly growing field in Physics. Nonlinearities are found everywhere in optical applications and at present, NLO properties of many optical materials with significant relevance to technological and optical applications have been found. In the present scenario, where a lot of emphasis is devoted to the growth and structural elucidation of NLO materials, this word presents a detailed investigation of the NLO properties of selected single organic compounds. Conclusively, the significances of this study mainly contribute towards the increment of our underlying knowledge on the nonlinear behaviours of α-Mangostin, Malachite green, Light green SF Yellowish, Coumarin 500 and Ponceau BS to be specific. Understanding and quantifying the physics of such process gives an insight into the field of nonlinear optics. A successful development of the proposed study leaves a direct benefit for scientific awareness of the country, and the whole research activities can be used for future references.

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