OPTICAL BISTABILITY IN NONLINEAR MEDIUM

NARRENDHARAN A/L ELEMARAN

UNIVERSITI SAINS MALAYSIA

2022

OPTICAL BISTABILITY IN NONLINEAR MEDIUM

By

NARRENDHARAN A/L ELEMARAN

Thesis submitted to the School of Physics, Universiti Sains Malaysia, in Fulfilment of the Requirement for the course ZCT 390/8 Pure Physics Project

JULY 2022

BISTABILITI OPTIK DALAM MEDIUM TIDAK LINEAR

ABSTRAK

Dalam kajian ini, kami telah mengkaji bistabliti optic dalam medium bukan linear dengan menggunakan kaca tellurit plumbum zink sebagai model kami. Untuk memudahkan dan menjadikan kajian berkesan, kami telah megehadkan diri kami untuk meneroka dan menyelesaikan *slowly varying envelope solution (SVEA)* dan persamaan fluks tenaga. Proses ini akan melibatkan beberapa derivasi, pengiraan dan teori. Untuk lebih spesifik, kami telah memperoleh and mangira persamaan polarasi, persamaan Maxwell, dan kebolehcurigaan linear elektrik. Selain itu, tindak balas linear dan tidak linear telah menyumbang sebagai teori utama dalam karya ini. Unutuk menujukkan dan mendapatkan hasil bistabiliti optic dalam medium tidak linear kami mengunakkan bahasa penagturcaraan Python untuk manipulasi data, menjalankan simulasi dan menjana graf. Kami telah menghasilkan semula hasil kerja J. A. Goldstone dan E.Garmire iaitu *Intrinsic Optical Bistability In Nonlinear Medium (1983).*

OPTICAL BISTABILITY IN NONLINEAR MEDIUM

ABSTRACT

In this paper, we study the optical bistability in the nonlinear medium using zinc lead tellurite glass as our model. We have restricted ourselves to exploring and solving the slowly varying envelope solution (SVES) and energy flux equation to keep things simple and effective. This process will involve several derivations, calculations, and theories. Specifically, we have derived and calculated the polarization equation, Maxwell's equations, and the electric linear susceptibility. The linear and nonlinear response has contributed to the main theory in this work. To demonstrate and obtain the results of optical bistability in a nonlinear medium, we used python programming language to manipulate data, run simulations and generate graphs. We have reproduced the work of J. A. Goldstone and E. Garmire which is Intrinsic Optical Bistability in Nonlinear Medium (1983).

ACKNOWLEDGEMENTS

First and foremost, I want to express my gratitude to Assoc. Prof. Dr Lim Siew Choo for giving me the chance to work on this project and for her support this entire year. I've had to learn many new things this year, and I couldn't have done it without her efforts. I also want to express my gratitude to the institution and the School of Physics at USM for giving me the chance to research this issue. Despite the challenges, the ZCT 390/8 Pure Physics Project course has been well-managed, allowing students to engage in real physics research while also picking up new skills that will be helpful in their future endeavours.

Additionally, I want to thank all my classmates and friends, especially my groupmate Ravin A/L Thiruchelvan. Without their support, I would not have been able to learn so many new things over this year while going through difficult and unexpected circumstances.

Last but not least, I want to convey my gratitude for my family's patience, especially my parents. Their support, which they gave me with everything they had, was crucial to my ability to be optimistic and succeed in my studies.

CONTENTS

LIST OF TABLES

LIST OF FIGURES

CHAPTER 1 INTRODUCTION

1.1 General Overview

In contrast to linear science, where we commonly have well-known tools to tackle a specific problem, nonlinear science, or the research of systems capable of creating new frequencies, is pervasive and fascinating. When nonlinearity comes into play, however, and the whole can be much more than the sum of its parts, we can rarely resort to standard techniques. It is anticipated and welcomed that qualitatively novel phenomena will emerge, with tremendous implications for basic and applied sciences [1].

Nonlinear optics (NLO), an entirely new field, was discovered after the emergence of the laser in 1960. Later, it was rapidly developed and progressed in quantum optics. This has eventually made a huge impact on cavity quantum electrodynamics as well. Pure excitement was raised by optical bistability (OB) at that time, defined as an optical system possessing two different steady-state transmissions for the same input intensity, also described as a kind of passive counterpart of the laser [2].

An atomic medium is placed in an optical cavity where a transition is resonant with one cavity mode in a laser system. Atoms are excited to cause population inversion at the appropriate levels, and stimulated emission overcomes absorption. If the subsequent amplification can balance the cavity losses, a stationary oscillation forms in the resonator, whose output is the laser field.

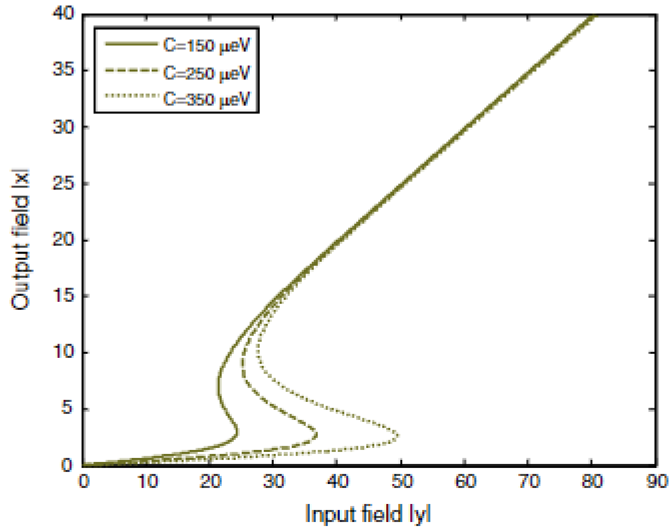

Figure 1.1: Output field $|x|$ *versus input field* $|y|$ *for different values* (Retrieved from Superlattices and Microstructures: Volume 84, August 2015, Pages 45-53)

Consider a similar system with two key differences: the atomic medium is not pumped, and a coherent external field drives the optical cavity. According to Figure 1.1, the intensity of the light transmitted from the resonator can assume two different values corresponding to the same power of the incident field in this system [3].

nonlinear optics has improved our understanding of fundamental light-matter interactions and provided us with new and powerful tools for material research [4]. On the other hand, it has a significant impact on technology since it allows for the development of coherent radiation sources that emit at new frequencies not typically available with lasers and can be tuned from ultraviolet to visible near infrared. Important applications also include optical devices that fulfil the same functions as electronic devices, such as modulators, switches, and logical circuits at radio frequencies.

1.2 Properties of Glass

In investigating the bistability of a nonlinear medium, a model is required. Hence, we have chosen glass. Most of the laser research in the early days was focused on crystals, particularly non-centrosymmetric materials like Quartz, $LiNbO_3$, $KTiOPO_4$ (KDP), and $-B_aB_2O_4$ (BBO) [5]. Second harmonic generation (SHG) or the electro-optic effect does not occur in glasses because of their isotropic nature (Pockels effect). Articles addressing the possibility of NLO characteristics in an organic glass first appeared in the scientific community in the late 1980s [6]. The creation of a unique light source capable of delivering energy in a short time and generating a high electric field in the matter is of interest. Short-pulse lasers have given researchers access to an electric field ($10^{10}\ V/m$) on order of that between the atom's outermost electrons [7]. This has developed an interest in the nonlinear optical properties of glasses. This has further inspired us to study the nonlinear properties of zinc lead tellurite glass ($[Z_nO]_x[(T_eO_2)_{0.7} - (P_bO)_{0.3}]_{1-x}$), which we also chose to make our model medium for this work.

1.3 Problem Statement

We are involved in bistability results in a nonlinear media in all macroscopic optical and material parameters. In 1983, J. A. Goldstone and E. Garmire [1] calculated the atomic polarization and susceptibility for nonlinear media. They have used a simple classical model for the complex nonlinear polarization based on the Duffing oscillator and Maxwell's equation to derive the polarization and susceptibility, whose solutions must be consistent with the relevant boundary conditions. We are focused on reproducing their results by obtaining the envelope solution and the energy flux that the authors have expressed in the mentioned paper. However, in their previous work, the derivation of the polarization of nonlinear medium at fundamental frequency into an envelope solution has not been analysed in detail. Attempting to polarize and analyse the nonlinear medium while resolving the slowly varying envelope solution (SVES) and energy flux solution is what we try to settle in this work.

1.4 Aims and Objectives

- We are keen on describing optical bistability in a nonlinear medium. We consider the nonlinear susceptibilities that govern the nonlinear medium's optical properties.
- We have aimed to reproduce and analyse in detail the work of J. A. Goldstone and E. Garmire.
- We have chosen zinc lead tellurite glass as an example to estimate the parameters required in Maxwell's equation, so we can generate graphs to analyse their behaviour.

1.5 Organisation of Thesis

In Chapter 2, related literature on bistability in nonlinear media is addressed. In Chapter 3, the theory of duffing oscillator, nonlinear response is introduced. Derivation of polarization equation and electric linear susceptibilities are presented as well. In Chapter 4, the Maxwell’s equation is outlined in detail and the slowly varying envelope solution is calculated. In Chapter 5, the results are analysed and plotted in a graphical view. Discussion on the analysis is explained in detail as well. In Chapter 6, the work done in previous chapters is summarised, and some possibilities for further study on this work are explored.

CHAPTER 2 LITERATURE REVIEW

Nonlinear optics is the science of occurrences that result from the alteration of a material system's optical characteristics by the presence of light. In 1960, Maiman demonstrated the first functional laser. Not long after, the emergence of the second-harmonic generation by Franken et al. is recognised to mark the beginning of the area of nonlinear optics in 1961 [8]. Nonlinear optical phenomena are so-called because they arise when a material system's reaction to an induced optical field relies nonlinearly on the electric field amplitude of the optical field [5].

The development in nonlinear optics has emerged with the rise of optical bistability. It has become the most researched field in the past decade due to the intriguing phenomena it encompasses. In 1969, Szöke *et al*. claimed that a Fabry-Perot optical resonator's saturable absorber could exhibit two bistable transmission states for the same input intensity [9]. The basic concept is that at the maximum power, the induced transparency permits constructive interference at resonant wavelengths due to the enormous internal field in this circumstance.

However, the studies cited had conflicting results. It wasn’t until 1974 when Gibbs *et al.* successfully established optical bistability in a passive, exciting medium of sodium (Na) vapour in a Fabry-Perot interferometer [3]. They concluded that nonlinear refraction, not saturable absorption, was what was accountable for the phenomenon. Ever since the physics underpinning optical bistability has attracted much theoretical interest, the results of that interest were shown in a wide range of materials, including the intrinsic semiconductor etalons [4,10].

According to [11], a system can be dispersive or absorptive. A system is either absorptive or dispersive based on whether the feedback happens through an intensity-dependent absorption or refractive index. According to Levin and Tang in 1979, it was the first time that bistability was seen in a laser with the hysteresis variable as the input light intensity [2]. Their technology is a hybrid dispersive system made of an Ar-laser-pumped dye laser adjusted by an electrooptical birefringent tuner.

As stated by R Bonifacio and F Casagrande [4], a polarisation, $P(x,t)$ with a volume, V in a group of N two-level atoms are induced by a traditional monochromatic radiation field, $E(x,t) = E_0 \cos{(\omega t - kx)}$ where,

$$P(x,t) = \varepsilon_0[\chi_d \cos(\omega t - kx) + \ \chi_a \sin(\omega t - kx)]E_0, \tag{2.1}$$

The response of the medium to the field is described by the complex dielectric susceptibility, $\chi = \chi_d + i\chi_a$. This shows that there are two components to the induced atomic polarization: one that is in phase (dispersive) with the field and the other that is in quadrature (absorptive) [2].

In line with J. A. Goldstone and E. Garmire [1], with one propagating wave both dispersive and absorptive optical bistability are explained by taking into account plane-wave propagation from a linear media $(z < 0)$ with index n_L into a nonlinear medium $(z \geq 0)$ in the positive z direction [1]. Hence, the differential form of equation (2.1) is developed. Leading to one dimensional Duffing potential used as a binding potential to create a stable response. Thus, the crucial component in this study is to treat the distinct parameter in the wave equation to construct a slowly varying envelope solution (SVES) and energy flux equation.

Although the authors include other reasons and theories in their publication, the discussions in those works fall outside the purview of this work. Hence, we gather the parameters and values required to move forward with this work and to show our interest in expanding it.

CHAPTER 3 THEORY

3.1 Duffing Oscillator

One of the nonlinear dynamics' prototype systems is the Duffing oscillator. In the wake of early investigations by the engineer Georg Duffing, it originally gained popularity for analysing anharmonic oscillations and later chaotic nonlinear dynamics [6]. A wide range of physical phenomena, including reinforcing springs, laser buckling, nonlinear digital components, cryogenic Josephson dynamic amplifiers, and ionisation waves in plasmas, have been effectively modelled using the system.

The following differential equation describes the Duffing oscillator

$$\ddot{x} + r\dot{x} + {\omega_0}^2 x + \beta x^3 = f\ cos\ \omega t, \tag{3.1}$$

which deviates from the forced and damped harmonic oscillator example from the introductory textbook by the nonlinear term βx^3. This significantly alters the system's dynamics. However, the harmonic oscillator

$$\ddot{x} + r\dot{x} + {\omega_0}^2 x = f\ cos\ \omega t, \tag{3.2}$$

provides a convenient way to look when discussing the nonlinear reference subsequently. Nonetheless, for $\beta = 0$, a solution is given by

$$x(t) = A\cos(\omega t - \emptyset), \tag{3.3}$$

In order to comprehend the Duffing oscillator, it is imperative to look at the potential associated with the force's time-independent component, ${\omega_0}^2 x + \beta x^3$. Be that as it may

$$V(x) = \frac{1}{2}{\omega_0}^2 x^2 + \frac{1}{4}\beta x^4, \tag{3.4}$$

this can be thought of as the first terms of a Taylor expansion of a generic (symmetric) potential. Here demonstrates the value of using the Duffing oscillator to approximate more generic interactions [3]. Consequently, we would want to emphasise that the value ${\omega_0}^2$ may also be negative, in which case ω_0 cannot be regarded as a frequency.

We want to consider a straightforward classical model for the complex nonlinear polarization based on the Duffing oscillator, which has previously been used in the analysis of bistability in four-wave mixing, to clarify the general idea and to demonstrate the range of macroscopic manifestation of intrinsic material bistability on the optical driving fields [1]. We establish that optical bistability is both absorptive and dispersive with just one propagating wave.

3.2 Nonlinear Response

In this section, we will briefly overview the nonlinear optics techniques used to determine optical bistability. The complex electric field and polarisation can be defined in various ways, as explained in [2,4].

By adopting the plane-wave propagation in the positive z direction from a linear medium ($z < 0$) with index n_L into a nonlinear medium ($z \geq 0$), we limit the analysis to low molecular densitites N, for mathematical convenience [1]. The polarization of the nonlinear medium at the fundamental frequency ω is regarded as

$$\vec{P}(z,t) = \vec{P}(z)e^{-i\omega t} = N\vec{p}(z)e^{-i\omega t}, \tag{3.5}$$

where $\vec{p}(z)$ is the amplitude of molecular polarization and the driving field is $N(z)e^{-i\omega t}$. The polarization can be written as a function of the applied field. Expanding P_i,

$$P_i = P_i^{(1)} + P_i^{(2)} + P_i^{(3)}, \tag{3.6}$$

But when the electric linear susceptibility is involved, P_i becomes

$$P_i^{(1)} = \epsilon_0 \chi_i^{(1)} E(z,t), \tag{3.7}$$

for first order linear susceptibility. The rest follows as,

$$P_i^{(2)} = \epsilon_0 \chi_i^{(2)} E(z,t)E(z,t), \tag{3.8}$$

for second order linear susceptibility, and

$$P_i^{(3)} = \epsilon_0 \chi_i^{(3)} E(z,t)E(z,t)E(z,t), \tag{3.9}$$

for third order susceptibility. Where $\chi_i = n^2 - 1$ and n is the refractive index. The cumulative convention is assumed in these equations and throughout the rest of this work. However, to be considerate only the linear susceptibility $\chi^{(1)}$ is significant for small E while the nonlinear susceptibility $\chi^{(2)}$ and $\chi^{(3)}$ must be considered for large E. High order terms are typically ignored due to being irrelevant for practical applications.

At this point, we take a monochromatic wave in the form

$$E_i = \frac{1}{2}(E_{0i}e^{-i\omega t} + E_{0i}^{*}e^{i\omega t}), \tag{3.10}$$

Equations (3.7), (3.8), and (3.9) are only said to be valid when it comes to the static field, and the expressions are more complex considering integrals are involved in a time-varying field. Hence, assuming that the polarisation responds to advancements in the field instantaneously [5], equation (3.7) is augmented into

$$P_i{}^{(1)} = \frac{1}{2}\epsilon_0\left[\chi_{il}{}^{(1)}(\omega)E_{0l}e^{-i\omega t} + \chi_{il}{}^{(1)}(\omega)E_{0l}^{*}e^{i\omega t}\right], \tag{3.11}$$

where the polarisation must be real $\chi_i^{(1)}(\omega) = \left(\chi_i^{(1)}(-\omega)\right)^*$. Correspondingly, equation (3.8) is also replaced by

$$\begin{gathered} P_i{}^{(2)} = \frac{1}{4}\epsilon_0\big[\chi_{ilm}{}^{(2)}(-2\omega;\omega,\omega)E_{0l}E_{0m}e^{-2i\omega t} + \\ \chi_{ilm}{}^{(2)}(0;\omega,-\omega)E_{0l}E_{0m}^{*} + c.c.\big], \end{gathered} \tag{3.12}$$

and similarly, equation (3.9) is replaced by

$$\begin{gathered} P_i{}^{(3)} = \frac{1}{8}\epsilon_0\big[\chi_{ilmn}{}^{(3)}(-3\omega;\omega,\omega,\omega)E_{0l}E_{0m}E_{0n}e^{-3i\omega t} + \\ \big(\chi_{ilmn}{}^{(3)}(-\omega;-\omega,\omega,\omega)E_{0l}^{*}E_{0m}E_{0n} + \\ \chi_{ilmn}{}^{(3)}(-\omega;\omega,-\omega,\omega)E_{0l}E_{0m}^{*}E_{0n} + \\ \chi_{ilmn}{}^{(3)}(-\omega;\omega,\omega,-\omega)E_{0l}E_{0m}E_{0n}^{*}\big)e^{-i\omega t} + c.c.\big], \end{gathered} \tag{3.13}$$

Since a quantum mechanical model is not what we are contemplating here, we can only explore a phenomenological model [5]. Hence, the overall permutation symmetry is preserved. Thus, we have

$$\chi_{ilm}{}^{(2)}(0;\omega,-\omega)E_{0l}E_{0m}^{*} = \chi_{ilm}{}^{(2)}(0;-\omega,\omega)E_{0l}^{*}E_{0m}, \tag{3.14}$$

$$\begin{gathered}\chi_{ilmn}{}^{(3)}(-\omega;-\omega,\omega,\omega)E_{0l}^{*}E_{0m}E_{0n} = \\ \chi_{ilmn}{}^{(3)}(-\omega;\omega,-\omega,\omega)E_{0l}E_{0m}^{*}E_{0n} = \\ \chi_{ilmn}{}^{(3)}(-\omega;\omega,\omega,-\omega)E_{0l}E_{0m}E_{0n}^{*},\end{gathered} \tag{3.15}$$

Observing from equation (3.14) and (3.15), we can obtain

$$\begin{gathered}P_i{}^{(2)} = \frac{1}{4}\epsilon_0\big[\chi_{ilm}{}^{(2)}(-2\omega;\omega,\omega)E_{0l}E_{0m}e^{-2i\omega t} + \\ 2\chi_{ilm}{}^{(2)}(0;\omega,-\omega)E_{0l}E_{0m}^{*} + \chi_{ilm}{}^{(2)}(2\omega;\omega,\omega)E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\big],\end{gathered} \tag{3.16}$$

and

$$\begin{gathered}P_i{}^{(3)} = \frac{1}{8}\epsilon_0\big[\chi_{ilmn}{}^{(3)}(-3\omega;\omega,\omega,\omega)E_{0l}E_{0m}E_{0n}e^{-3i\omega t} + \\ 3\chi_{ilmn}{}^{(3)}(-\omega;\omega,\omega,-\omega)E_{0l}E_{0m}E_{0n}^{*}e^{-i\omega t}\big] + c.c.\,,\end{gathered} \tag{3.17}$$

In the equations above, the notation c.c. stands for complex conjugate and $l, m, or\ n$ stands for orders. We will refer to equation (3.7), (3.8), and (3.9) in this section as a simplification, but it should be understood that the terms within should be revised to resemble the ones mentioned above.

3.3 Derivation of Polarization Equation

Based on the theory outlined in [1], we demonstrate and derive the polarization equation. As we discussed in section (3.2), equation (3.5) is differentiated into a form

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -\vec{\nabla} V(\vec{x}) + e\vec{E}(z,t), \tag{3.18}$$

where m is the mass of bound charges and $\gamma' \frac{\partial \vec{p}}{\partial t}$ represents a linear loss. As mentioned, in order to achieve a bistable response equation (3.4), Duffing potential is substituted into equation (3.18) and resulting in

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -\frac{1}{2}\kappa' x^2 + \frac{1}{4}\beta' x^4 + e\vec{E}(z,t), \tag{3.19}$$

where ${\omega_0}^2 = \kappa'$. Now equation (3.10) is substituted as well

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -\frac{1}{2}\kappa' x^2 + \frac{1}{4}\beta' x^4 + \frac{e}{2}(E_{0i}e^{-i\omega t} + E_{0i}^* e^{i\omega t}), \tag{3.20}$$

By simplification,

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -[\kappa' x + \beta' x^3] + \frac{e}{2}(E_{0i}e^{-i\omega t} + E_{0i}^* e^{i\omega t}), \tag{3.21}$$

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -\left[\kappa' \left(\frac{p}{e}\right) + \beta' \left(\frac{p}{e}\right)^3\right] + \frac{e}{2}(E_{0i}e^{-i\omega t} + E_{0i}^* e^{i\omega t}), \tag{3.22}$$

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -\left[\frac{\kappa'}{e} p + \frac{\beta'}{e^3} p^3\right] + \frac{e}{2}(E_{0i}e^{-i\omega t} + E_{0i}^* e^{i\omega t}), \tag{3.23}$$

Let $\kappa = \frac{\kappa'}{e}$ and $\beta = \frac{\beta'}{e^3}$. Hence,

$$\frac{m}{e}\frac{\partial^2 \vec{p}}{\partial t^2} + \gamma' \frac{\partial \vec{p}}{\partial t} = -[\kappa p + \beta p^3] + \frac{e}{2}(E_{0i}e^{-i\omega t} + E_{0i}^* e^{i\omega t}), \tag{3.24}$$

$$\frac{\partial^2 \vec{p}}{\partial t^2} + \left(\frac{e}{m}\right)\gamma' \frac{\partial \vec{p}}{\partial t} = -\left(\frac{e}{m}\right)[\kappa p + \beta p^3] + \frac{e}{2}\left(\frac{e}{m}\right)(E_{0i}e^{-i\omega t} + E_{0i}^* e^{i\omega t}), \tag{3.25}$$

Let $\gamma = \left(\frac{e}{m}\right)\gamma'$, $\bar{\kappa} = \left(\frac{e}{m}\right)\kappa$, $\bar{\beta} = \left(\frac{e}{m}\right)\beta$, and $\bar{E} = \left(\frac{e^2}{m}\right)E_{0i}$. Therefore,

$$\frac{\partial^2 p}{\partial t^2} + \gamma \frac{\partial p}{\partial t} + \bar{\kappa} p + \bar{\beta} p^3 = \frac{1}{2}\bar{E}_{0i}e^{-i\omega t} + \frac{1}{2}\bar{E}_{0i}^* e^{i\omega t}, \tag{3.26}$$

3.4 Calculation of Electric Linear and Nonlinear Susceptibility

Since the equation has been derived and simplified completely as shown above, thus the electric linear susceptibility is to be determined according to [1]. For that, we substitute equation (3.11) into equation (3.26). Hence,

$$\frac{\partial^2}{\partial t^2}\left(\frac{1}{2}\epsilon_0 \chi_{il}{}^{(1)} E_{0l}e^{-i\omega t} + \frac{1}{2}\epsilon_0 \chi_{il}{}^{(1)} E_{0l}^* e^{i\omega t}\right) + \gamma \frac{\partial}{\partial t}\left(\frac{1}{2}\epsilon_0 \chi_{il}{}^{(1)} E_{0l}e^{-i\omega t} + \frac{1}{2}\epsilon_0 \chi_{il}{}^{(1)} E_{0l}^* e^{i\omega t}\right) + \bar{\kappa}\left(\frac{1}{2}\epsilon_0 \chi_{il}{}^{(1)} E_{0l}e^{-i\omega t} + \frac{1}{2}\epsilon_0 \chi_{il}{}^{(1)} E_{0l}^* e^{i\omega t}\right) + \bar{\beta} p^3 = \frac{1}{2}\bar{E}_{0i}e^{-i\omega t} + \frac{1}{2}\bar{E}_{0i}^* e^{i\omega t}, \tag{3.27}$$

$\bar{\beta} p^3$ is ignored because it's a third order term,

$$\frac{\partial^2}{\partial t^2}\left(\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)+\gamma\frac{\partial}{\partial t}\left(\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+\right.$$

$$\left.\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)+\bar{\kappa}\left(\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)=$$

$$\frac{1}{2}\bar{E}_{0i}e^{-i\omega t}+\frac{1}{2}\bar{E}_{0i}^{*}e^{i\omega t}, \tag{3.28}$$

$$\left((-i\omega)(-i\omega)\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+(-i\omega()-i\omega)\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)+$$

$$\gamma\left((-i\omega)\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+(-i\omega)\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)+$$

$$\bar{\kappa}\left(\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)=\frac{1}{2}\bar{E}_{0i}e^{-i\omega t}+\frac{1}{2}\bar{E}_{0i}^{*}e^{i\omega t}, \tag{3.29}$$

$$-\omega^2\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}-\omega^2\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}-$$

$$\gamma i\omega\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}-\gamma i\omega\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}+\bar{\kappa}\left(\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+\right.$$

$$\left.\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}^{*}e^{i\omega t}\right)=\frac{1}{2}\bar{E}_{0i}e^{-i\omega t}+\frac{1}{2}\bar{E}_{0i}^{*}e^{i\omega t}, \tag{3.30}$$

Ignoring the complex conjugate (CC) to identify the real equation and by simplifying the equation,

$$-\omega^2\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}-\gamma i\omega\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+$$

$$\bar{\kappa}\frac{1}{2}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}=\frac{1}{2}\bar{E}_{0i}e^{-i\omega t}, \tag{3.31}$$

$$-\omega^2\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}-\gamma i\omega\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}+\bar{\kappa}\epsilon_0\chi_{il}{}^{(1)}E_{0l}e^{-i\omega t}=$$

$$\bar{E}_{0i}e^{-i\omega t}, \tag{3.32}$$

$$-\omega^2\epsilon_0\chi_{il}{}^{(1)} - \gamma i\omega\epsilon_0\chi_{il}{}^{(1)} + \bar{\kappa}\epsilon_0\chi_{il}{}^{(1)} = 1, \tag{3.33}$$

$$\epsilon_0\chi_{il}{}^{(1)}[-\omega^2 - \gamma i\omega + \bar{\kappa}] = 1, \tag{3.34}$$

Therefore, the first order linear susceptibility is,

$$\chi_{il}{}^{(1)} = \frac{1}{\epsilon_0(-\omega^2 - \gamma i\omega + \bar{\kappa})}, \tag{3.35}$$

To find the second-order nonlinear susceptibility, we substitute equation (3.12) into equation (3.26). Hence,

$$\begin{gathered}
\frac{\partial^2}{\partial t^2}\Big(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t} + \frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^* + \\
\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^*E_{0m} + \frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^*E_{0m}^*e^{2i\omega t}\Big) + \\
\gamma\frac{\partial}{\partial t}\Big(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t} + \frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^* + \\
\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^*E_{0m} + \frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^*E_{0m}^*e^{2i\omega t}\Big) + \\
\bar{\kappa}\Big(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t} + \frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^* + \frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^*E_{0m} + \\
\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^*E_{0m}^*e^{2i\omega t}\Big) + \bar{\beta}p^3 = \frac{1}{2}\bar{E}_{0i}e^{-i\omega t} + \frac{1}{2}\bar{E}_{0i}^*e^{i\omega t},
\end{gathered} \tag{3.36}$$

$\bar{\beta}p^3$ is ignored because it's a third order term and $\frac{1}{2}\bar{E}_{0i}e^{-i\omega t} + \frac{1}{2}\bar{E}_{0i}^*e^{i\omega t} = 0$ since it is a first order term. Thus,

$$\frac{\partial^2}{\partial t^2}\left(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^{*}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)+\gamma\frac{\partial}{\partial t}\left(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^{*}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)+\bar{\kappa}\left(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^{*}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)=0, \tag{3.37}$$

$$\frac{\partial}{\partial t}\left((-i\omega)\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+(-i\omega)\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)+\gamma\left((-i\omega)\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+(-i\omega)\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)+\bar{\kappa}\left(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^{*}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)=0, \tag{3.38}$$

$$-\omega^2\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}-\omega^2\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}-\gamma i\omega\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}-\gamma i\omega\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}+\bar{\kappa}\left(\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}^{*}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}+\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}\right)=0, \tag{3.39}$$

Again, by ignoring the complex conjugate (CC) to identify the real equation and by simplifying the equation,

$$-\omega^2\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t} - \gamma i\omega\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t} +$$

$$\bar{\kappa}\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}E_{0l}E_{0m}e^{-2i\omega t} = 0, \qquad (3.40)$$

$$-\omega^2\epsilon_0\chi_{ilm}{}^{(2)}e^{-2i\omega t} - \gamma i\omega\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)}e^{-2i\omega t} + \bar{\kappa}\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)}e^{-2i\omega t} = 0, \qquad (3.41)$$

$$-\omega^2\epsilon_0\chi_{ilm}{}^{(2)} - \gamma i\omega\frac{1}{2}\epsilon_0\chi_{ilm}{}^{(2)} + \bar{\kappa}\frac{1}{4}\epsilon_0\chi_{ilm}{}^{(2)} = 0, \qquad (3.42)$$

$$\epsilon_0\chi_{ilm}{}^{(2)}\left[-\omega^2 - \frac{1}{2}\gamma i\omega + \frac{1}{4}\bar{\kappa}\right] = 0, \qquad (3.43)$$

The second order nonlinear susceptibility is,

$$\chi_{ilm}{}^{(2)} = 0, \qquad (3.44)$$

To find the third-order nonlinear susceptibility, we substitute equation (3.13) into equation (3.26). Hence,

$$\frac{\partial^2}{\partial t^2}\Big(\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}E_{0n}e^{-3i\omega t} + \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}e^{-i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}^*e^{2i\omega t}E_{0n}e^{-i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}e^{-2i\omega t}E_{0n}^*e^{i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}^*e^{i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}^*e^{i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}^*E_{0n}^*e^{3i\omega t}\Big)$$

$$+ \gamma\frac{\partial}{\partial t}\Big(\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}E_{0n}e^{-3i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}e^{-i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}^*e^{2i\omega t}E_{0n}e^{-i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}e^{-2i\omega t}E_{0n}^*e^{i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}^*e^{i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}^*e^{i\omega t}$$

$$+ \frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}^*E_{0n}^*e^{3i\omega t}\Big)$$

$$+\bar{\kappa}\left(\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}E_{0n}e^{-3i\omega t}+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^{*}E_{0n}e^{-i\omega t}\right.$$

$$+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^{*}E_{0m}E_{0n}e^{-i\omega t}$$

$$+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^{*}E_{0m}^{*}e^{2i\omega t}E_{0n}e^{-i\omega t}$$

$$+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}e^{-2i\omega t}E_{0n}^{*}e^{i\omega t}$$

$$+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^{*}E_{0n}^{*}e^{i\omega t}$$

$$+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^{*}E_{0m}E_{0n}^{*}e^{i\omega t}$$

$$\left.+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^{*}E_{0m}^{*}E_{0n}^{*}e^{3i\omega t}\right)$$

$$+\bar{\beta}\left(\frac{1}{8}{\epsilon_0}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}E_{0n}e^{-3i\omega t}\right.$$

$$+\frac{1}{8}{\epsilon_0}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}E_{0n}^{*}e^{-i\omega t}$$

$$+\frac{1}{8}{\epsilon_0}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}^{*}E_{0n}e^{-i\omega t}$$

$$+\frac{1}{8}{\epsilon_0}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}^{*}E_{0n}^{*}e^{i\omega t}$$

$$+\frac{1}{8}{\epsilon_0}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}^{*}E_{0m}E_{0n}e^{-i\omega t}$$

$$+\frac{1}{8}{\epsilon_0}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}^{*}E_{0m}E_{0n}^{*}e^{i\omega t}$$

$$+\frac{1}{8}\epsilon_0{}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}^*E_{0m}^*E_{0n}e^{i\omega t}+$$

$$\frac{1}{8}\epsilon_0{}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}^*E_{0m}^*E_{0n}^*e^{3i\omega t}\Big)=\frac{1}{2}\bar{E}_{0i}e^{-i\omega t}+\frac{1}{2}\bar{E}_{0i}^*e^{i\omega t}, \tag{3.45}$$

$\frac{1}{2}\bar{E}_{0i}e^{-i\omega t}+\frac{1}{2}\bar{E}_{0i}^*e^{i\omega t}=0$ since it is a first order term. Thus, by ignoring the complex conjugate (CC) to identify the real equation and by simplifying the equation,

$$\frac{\partial^2}{\partial t^2}\Big(\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}e^{-i\omega t}$$

$$+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}E_{0n}^*e^{-i\omega t}\Big)+$$

$$\gamma\frac{\partial}{\partial t}\Big(\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}e^{-i\omega t}+$$

$$\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}E_{0n}^*e^{-i\omega t}\Big)+\bar{\kappa}\Big(\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}+$$

$$\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}^*E_{0m}E_{0n}e^{-i\omega t}+\frac{1}{8}\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}E_{0n}^*e^{-i\omega t}\Big)+$$

$$\bar{\beta}\Big(\frac{1}{8}\epsilon_0{}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}E_{0n}^*e^{-i\omega t}+$$

$$\frac{1}{8}\epsilon_0{}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}+$$

$$\frac{1}{8}\epsilon_0{}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}^*E_{0m}E_{0n}e^{-i\omega t}\Big)=0, \tag{3.46}$$

$$\frac{\partial^2}{\partial t^2}\left(\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}\right)+\ \gamma\frac{\partial}{\partial t}\left(\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}\right)+$$

$$\bar{\kappa}\left(\epsilon_0\chi_{ilmn}{}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}\right)+$$

$$\bar{\beta}\left(\epsilon_0{}^3\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}\chi_{il}{}^{(1)}E_{0l}E_{0m}E_{0n}^*e^{-i\omega t}\right)=0, \tag{3.47}$$

$$-\omega^2\epsilon_0{\chi_{ilmn}}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t} - \gamma i\omega\epsilon_0{\chi_{ilmn}}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}$$
$$+ \bar{\kappa}\epsilon_0{\chi_{ilmn}}^{(3)}E_{0l}E_{0m}^*E_{0n}e^{-i\omega t}$$
$$+ \bar{\beta}{\epsilon_0}^3{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}E_{0l}E_{0m}E_{0n}^*e^{-i\omega t} = 0, \tag{3.48}$$

$$-\omega^2{\chi_{ilmn}}^{(3)} - \gamma i\omega{\chi_{ilmn}}^{(3)} + \bar{\kappa}{\chi_{ilmn}}^{(3)} + \bar{\beta}{\epsilon_0}^2{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}{\chi_{il}}^{(1)} = 0, \tag{3.49}$$

$${\chi_{ilmn}}^{(3)}[-\omega^2 - \gamma i\omega + \bar{\kappa}] = \bar{\beta}{\epsilon_0}^2{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}, \tag{3.50}$$

Therefore, the third order linear susceptibility is,

$${\chi_{ilmn}}^{(3)} = \frac{\bar{\beta}{\epsilon_0}^2{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}{\chi_{il}}^{(1)}}{(-\omega^2-\gamma i\omega+\bar{\kappa})}, \tag{3.51}$$

Since we have calculated and achieved all the three orders of electric linear susceptibility, thus we proceed to calculate the final form of the polarization equations. By substituting equation (3.35) into equation (3.7) we obtain,

$${P_i}^{(1)} = \frac{1}{2(-\omega^2-\gamma i\omega+\bar{\kappa})}E_{0i}e^{-i\omega t}, \tag{3.52}$$

For the second order polarization, ${P_i}^{(2)} = 0$ because the second order electric linear susceptibility is ${\chi_{ilm}}^{(2)} = 0$. Hence, ${P_i}^{(2)}$ is ignored. Nevertheless, by substituting equation (3.51) into equation (3.9),

$$P_i^{(3)} = \epsilon_0 \frac{\bar{\beta}\epsilon_0{}^2 \chi_{il}{}^{(1)} \chi_{il}{}^{(1)} \chi_{il}{}^{(1)}}{(-\omega^2 - \gamma i\omega + \bar{\kappa})} \frac{1}{2} E_{0l} e^{-i\omega t} \frac{1}{2} E_{0m} e^{-i\omega t} \frac{1}{2} E_{0n} e^{-i\omega t}, \tag{3.53}$$

$$P_i^{(3)} = \frac{\bar{\beta} \frac{1}{(-\omega^2 + \gamma i\omega + \bar{\kappa})} \frac{1}{(-\omega^2 - \gamma i\omega + \bar{\kappa})} \frac{1}{(-\omega^2 - \gamma i\omega + \bar{\kappa})}}{(-\omega^2 - \gamma i\omega + \bar{\kappa})} \frac{1}{2} E_{0l}^* e^{i\omega t} \frac{1}{2} E_{0m} e^{-i\omega t} \frac{1}{2} E_{0n} e^{-i\omega t}, \tag{3.54}$$

$$P_i^{(3)} = \frac{\bar{\beta}}{(-\omega^2 + \gamma i\omega + \bar{\kappa})(-\omega^2 - \gamma i\omega + \bar{\kappa})^3} \frac{1}{2} E_{0l}^* e^{i\omega t} \frac{1}{2} E_{0m} e^{-i\omega t} \frac{1}{2} E_{0n} e^{-i\omega t}, \tag{3.55}$$

$$P_i^{(3)} = \frac{\bar{\beta}}{8(-\omega^2 + \gamma i\omega + \bar{\kappa})(-\omega^2 - \gamma i\omega + \bar{\kappa})^3} E_{0l}^* E_{0m} E_{0n} e^{-i\omega t}, \tag{3.56}$$

Now, we have obtained the final form of $P_i^{(1)}$ and $P_i^{(3)}$ which we will use for further derivations and calculations in this work.

CHAPTER 4 METHODOLOGY AND CALCULATION

4.1 Maxwell’s Equation

Maxwell's equations are named after James Clerk Maxwell, the Scottish physicist. The Scottish scientist first brought together the ideas of electricity, magnetism, and light via his ground-breaking work [12]. These equations underlie all current information and communication systems and comprehensively account for electromagnetic events. Many scientists and engineers made discoveries and advancements that contributed to the development of the theory of electromagnetism. Still, Maxwell's pivotal work in the second half of the 19th century was what allowed for the great results in electrical technology that would occur throughout the 20th century.

The term "Maxwell's Equations" designates a group of four equations that explain the characteristics and interactions of electric and magnetic fields. Which are known to be

$$\nabla \cdot D = \rho_f, \tag{4.1}$$

$$\nabla \cdot B = 0, \tag{4.2}$$

$$\nabla \times E = -\frac{\partial B}{\partial t}, \tag{4.3}$$

$$\nabla \times H = -\frac{\partial D}{\partial t} + J_f, \tag{4.4}$$

Where ρ_f is charge density and J_f is current density. Gauss' Law (4.1), Gauss' Law for magnetism (4.2), Faraday's Law (4.3) and Ampère-Maxwell Law (4.4), where E is the electric field, D is the electric displacement, H is the magnetic field and B is the magnetic flux density.

The microscopic response of a nonlinear material stimulated by an optical field can be described by one or more nonlinear constitutive laws of the form

$$f\left(\vec{E}, \vec{a}_i, b_i\right) = 0, \tag{4.5}$$

where $\vec{E}$ represents the impressed fields and $\vec{a}_i$ and b_i are vector and scalar microscopic response parameters respectively. Expanding the macroscopic parameters that $\vec{a}_i$ and b_i have determined in power series in the fields, as is done in the polarisation expansion, is the usual technique for addressing relations of this sort [1]. Then, using these macroscopic characteristics, Maxwell's equations are utilised to create nonlinear wave equations, the solutions of which must satisfy equation (4.5) and the pertinent boundary conditions. The optical fields will be represented as independent variables in this process, it is assumed. This assumption, however, is incorrect given the right circumstances, because relations (4.5) result in a multivalued response where two or more stable solutions for $\vec{a}_i$ and/or b_i may exist over a wide range of values for $\vec{E}$.

4.2 Derivation of Wave Equation

For the subsequent calculation to obtain the slowly varying envelope solution (SVES), it is critical to obtain the wave equation in terms of the electric field, E, and polarization, P. According to [1, 2], the equations need to be adapted to our example, which are straightforward dielectric medium (non-magnetic and non-ferroelectric). First of all, in dielectric, the free charge is absent. Thus $\rho_f = 0$ and equation (4.1) will become,

$$\nabla \cdot D = 0, \tag{4.6}$$

$$\nabla \cdot E = 0, \tag{4.7}$$

where $D = \epsilon E = \epsilon_0 E + P$ and ϵ is a constant which is ignored for this equation since its only valid for linear dielectric.

Moreover, in dielectric there is no free current as well. Hence $J_f = 0$, which will result equation (4.4) in,

$$\nabla \times H = -\frac{\partial D}{\partial t}, \tag{4.8}$$

and

$$H = \frac{1}{\mu_0} B, \tag{4.9}$$

By substituting equation (4.9) into equation (4.8),

$$\nabla \times B = \mu_0 (\nabla \times H), \tag{4.10}$$

Now, equation (4.3) is altered by multiplying with ∇,

$$\nabla \times (\nabla \times E) = -\frac{\partial (\nabla \times B)}{\partial t}, \tag{4.11}$$

and with the help of vector identity,

$$\nabla(\nabla \cdot E) - (\nabla \cdot \nabla)E = -\frac{\partial(\nabla \times B)}{\partial t}, \tag{4.12}$$

By substituting equation (4.7) and (4.10) into equation (4.12), we obtain,

$$\nabla(0) - (\nabla \cdot \nabla)E = -\frac{\partial\left(\mu_0(\nabla \times H)\right)}{\partial t}, \tag{4.13}$$

$$(\nabla \cdot \nabla)E = \mu_0 \frac{\partial(\nabla \times H)}{\partial t}, \tag{4.14}$$

Now, to obtain the final wave equation we substitute equation (4.8) into equation (4.14). Therefore,

$$(\nabla \cdot \nabla)E = \mu_0 \frac{\partial\left(-\frac{\partial D}{\partial t}\right)}{\partial t}, \tag{4.15}$$

$$(\nabla \cdot \nabla)E = \mu_0 \frac{\partial^2 D}{\partial t^2}, \tag{4.16}$$

$$(\nabla \cdot \nabla)E = \mu_0 \frac{\partial^2(\epsilon_0 E + P)}{\partial t^2}, \tag{4.17}$$

The final wave equation in terms of electric field and polarization as mentioned above,

$$\nabla^2 E = \mu_0 \frac{\partial^2(\epsilon_0 E)}{\partial t^2} + \mu_0 \frac{\partial^2(P)}{\partial t^2}, \tag{4.18}$$

Furthermore, by substituting equation (3.53) and (3.57) into equation (4.18), we obtain,

$$\nabla^2 E = \mu_0 \frac{\partial^2(\epsilon_0 E)}{\partial t^2} + \mu_0 \frac{\partial^2\left({P_i}^{(1)} + {P_i}^{(3)}\right)}{\partial t^2}, \tag{4.19}$$

$$\nabla^2 E = \mu_0 \frac{\partial^2(\epsilon_0 E_{0i} e^{-i\omega t})}{\partial t^2} + \mu_0 \frac{\partial^2}{\partial t^2}\Big(\frac{1}{2(-\omega^2-\gamma i\omega+\overline{\kappa})} E_{0i} e^{-i\omega t} +$$

$$\frac{\overline{\beta}}{8(-\omega^2-\gamma i\omega+\overline{\kappa})(-\omega^2-\gamma i\omega+\overline{\kappa})^3} E_{0l}^* E_{0m} E_{0n} e^{-i\omega t}\Big), \tag{4.20}$$

where this equation will be needed for derivation later in this work.

4.3 Derivation and Calculation of Slowly Varying Envelope Solution (SVES)

The slowly varying envelope approximation (SVEA) is the notion that, in comparison to a period or wavelength, the envelope of a forward-moving wave pulse changes gradually through time and space [13]. This is frequently utilised because the governing equation is often simpler to solve than the initial variables, reducing the degree of some of the highest-order partial derivatives. Still, the validity of the assumptions that are made needs to be proved.

To achieve the slowly varying envelope solution (SVEA), we need to solve the wave equation while deriving the first and second order terms of $p(z)$. Hence, the derivation continues with the complex notation for polarization and electric field,

$$\mathrm{p} = \frac{1}{2} p(z,t) + c.c, \tag{4.21}$$

$$\mathrm{p} = \frac{1}{2} p(z) e^{-i\omega t} + c.c, \tag{4.22}$$

$$\mathrm{E} = \frac{1}{2} E(z,t) + c.c, \tag{4.23}$$

$$\mathrm{E} = \frac{1}{2}E(z)e^{-i\omega t} + c.c, \tag{4.24}$$

whereas mentioned before $c.c$ indicates complex conjugate. Both equation (4.21) and (4.23) are rewritten as equation (4.22) and (4.24) respectively by putting t as explicit.

Rearranging equation (3.23) as original real field,

$$\frac{m}{e}\frac{\partial^2 p}{\partial t^2} + \gamma'\frac{\partial p}{\partial t} + \left[\frac{\kappa'}{e}p + \frac{\beta}{e^3}' p^3\right] = eE(z,t), \tag{4.25}$$

Now, we substitute the complex notations which are equation (4.21) and (4.23) into the equation (4.25) to obtain,

$$\frac{m}{e}\frac{\partial^2 p}{\partial t^2} + \gamma'\frac{\partial p}{\partial t} + \left[\frac{\kappa'}{e}p + \frac{3\beta}{4e^3}' p^3\right] = eE(z,t), \tag{4.26}$$

By simplifying the equation further,

$$\frac{\partial^2 p}{\partial t^2} + \frac{e}{m}\gamma'\frac{\partial p}{\partial t} + \frac{\kappa'}{m}p + \frac{3\beta}{4me^2}'|p|^2 p = \frac{e^2}{m}E(z,t), \tag{4.27}$$

Let $\gamma = \frac{e}{m}\gamma'$, $\omega_0^2 = \frac{\kappa'}{m}$, and $\beta = \frac{\beta'}{me^2}$,

$$\frac{\partial^2 p}{\partial t^2} + \gamma\frac{\partial p}{\partial t} + \omega_0^2 p + \frac{3}{4}\beta|p|^2 p = \frac{e^2}{m}E(z,t), \tag{4.28}$$

Let $p = p(z)e^{-i\omega t}$ and $E = E(z)e^{-i\omega t}$,

$$\frac{\partial^2}{\partial t^2}p(z)e^{-i\omega t} + \gamma\frac{\partial}{\partial t}p(z)e^{-i\omega t} + \omega_0^2 p(z)e^{-i\omega t} +$$

$$\frac{3}{4}\beta|p(z)|^2 p(z)e^{-i\omega t} = \frac{e^2}{m}E(z)e^{-i\omega t}, \tag{4.29}$$

$$((-i\omega)(-i\omega p)(z)e^{-i\omega t}) + \gamma(-i\omega p(z)e^{-i\omega t}) + \omega_0^2 p(z)e^{-i\omega t} +$$

$$\frac{3}{4}\beta|p(z)|^2 p(z)e^{-i\omega t} = \frac{e^2}{m}E(z)e^{-i\omega t}, \tag{4.30}$$

$$\omega^2 p(z)e^{-i\omega t} - \gamma i\omega p(z)e^{-i\omega t} + \omega_0^2 p(z)e^{-i\omega t} +$$

$$\frac{3}{4}\beta|p(z)|^2 p(z)e^{-i\omega t} = \frac{e^2}{m}E(z)e^{-i\omega t}, \tag{4.31}$$

Cancelling out the $e^{-i\omega t}$ and factoring out $p(z)$ to make the equation clearer,

$$(\omega^2 - \gamma i\omega)p + \omega_0^2 p + \frac{3}{4}\beta|p|^2 p(z) = \frac{e^2}{m}E(z), \tag{4.32}$$

In order to achieve the solution of order β at the fundamental frequency, let $\Delta = \omega^2 + \omega_0^2$,

$$(\Delta - \gamma i\omega)p + \frac{3}{4}\beta|p|^2 p(z) = \frac{e^2}{m}E(z), \tag{4.33}$$

From equation (4.19), some adjustments were made to the equation by adopting the wave equation. Where equation (4.22) and (4.24) are substituted into the equation and simplified into,

$$\nabla^2 \frac{e^2}{m} E(z) = \mu_0 \epsilon_0 \frac{e^2}{m} \frac{\partial^2}{\partial t^2} E(z) e^{-i\omega t} + \mu_0 \frac{e^2}{m} \frac{\partial^2}{\partial t^2} p(z) e^{-i\omega t}, \tag{4.34}$$

$$\nabla^2 \frac{e^2}{m} E(z) = -\omega^2 \mu_0 \epsilon_0 \frac{e^2}{m} E(z) e^{-i\omega t} - \omega^2 \mu_0 \frac{e^2}{m} p(z) e^{-i\omega t}, \tag{4.35}$$

By cancelling out $e^{-i\omega t}$ and substituting equation (4.33) into equation (4.35),

$$\nabla^2 \left\{ (\Delta - \gamma i\omega) p + \frac{3}{4} \beta |p|^2 p(z) \right\} = -\omega^2 \mu_0 \epsilon_0 \left\{ (\Delta - \gamma i\omega) p + \frac{3}{4} \beta |p|^2 p(z) \right\} - \omega^2 \mu_0 \frac{e^2}{m} p(z), \tag{4.36}$$

There are certain modifications made to equation (4.36). However, it is crucial to take note that these changes made won't affect the structure of the equation but will enable the addition of the speed of light term, c, to equation (4.36) where $c = \frac{1}{\sqrt{\mu_0 \epsilon_0}}$, hence $\mu_0 \epsilon_0 = \frac{1}{c^2}$. Thus, a novel equation structure is eventually discovered,

$$\nabla^2 \left\{ (\Delta - \gamma i\omega) p + \frac{3}{4} \beta |p|^2 p(z) \right\} = -\frac{\omega^2}{c^2} \left\{ (\Delta - \gamma i\omega) p + \frac{3}{4} \beta |p|^2 p(z) \right\} - \frac{\omega^2}{c^2} \frac{e^2}{\epsilon_0 m} p(z), \tag{4.37}$$

To continue the derivation we need,

$$p(z) = \Lambda(z) \mathrm{e}^{\{i[\Phi(z) + kz]\}}, \tag{4.38}$$

where $\Lambda(z)$ is a complex function and $\emptyset(z)$ is a real function.

To explain this complexity in easier terms,

$$p = \Lambda e^{i(\phi(z)+kz)}, \tag{4.39}$$

$$p^* = \Lambda e^{-i(\phi(z)+kz)}, \tag{4.40}$$

where since equation (4.39) is already a complex function, thus it will result in formation of a complex conjugate function likewise equation (4.40). Hence, both equations are substituted in equation (4.37),

$$\nabla^2 \left\{(\Delta - \gamma i\omega)p + \frac{3}{4}\beta p^* pp\right\} = -\frac{\omega^2}{c^2}\left\{(\Delta - \gamma i\omega)p + \frac{3}{4}\beta p^* pp\right\} - \frac{\omega^2}{c^2}\frac{e^2}{\epsilon_0 m}p, \tag{4.41}$$

To further demonstrate the derivation and calculations, let,

$$\nabla^2 = \frac{d^2}{dz^2} = \frac{d}{dz}\frac{d}{dz}, \tag{4.42}$$

By substituting equation (4.42) into equation (4.41),

$$(\Delta - \gamma i\omega)\frac{d^2 p}{dz^2} + \frac{3}{4}\beta\frac{d^2(p^* pp)}{dz^2} = -\frac{\omega^2}{c^2}\left\{(\Delta - \gamma i\omega)p + \frac{3}{4}\beta p^* pp\right\} - \frac{\omega^2}{c^2}\frac{e^2}{\epsilon_0 m}p, \tag{4.43}$$

$$(\Delta - \gamma i\omega)\frac{d^2 p}{dz^2} + \frac{3}{4}\beta\left\{pp\frac{d^2 p^*}{dz^2} + 4p\frac{dp}{dz}\frac{dp^*}{dz} + 2pp^*\frac{d^2 p}{dz^2} + 2p^*\frac{dp}{dz}\frac{dp}{dz}\right\} = -\frac{\omega^2}{c^2}\left\{(\Delta - \gamma i\omega)p + \frac{3}{4}\beta p^* pp\right\} - \frac{\omega^2}{c^2}\frac{e^2}{\epsilon_0 m}p, \tag{4.44}$$

It is clear that in order to continue the derivation, we need to find $\frac{dp}{dz}$, $\frac{dp^*}{dz}$, $\frac{d^2p}{dz^2}$, and $\frac{d^2p^*}{dz^2}$. Hence, using the differential quotient rule,

$$\frac{dp}{dz} = \frac{d\Lambda}{dz} e^{i(\phi+kz)} + i\Lambda\left(\frac{d\phi}{dz} + k\right) e^{i(\phi+kz)}, \tag{4.45}$$

$$\frac{d^2p}{dz^2} = \frac{d^2\Lambda}{dz^2} e^{i(\phi+kz)} + i\Lambda\left(\frac{d\phi}{dz} + k\right) e^{i(\phi+kz)} + i\,\frac{d\Lambda}{dz}\left(\frac{d\phi}{dz} + k\right) e^{i(\phi+kz)} +$$
$$i\Lambda\frac{d^2\phi}{dz^2} e^{i(\phi+kz)} - \Lambda\left(\frac{d\phi}{dz} + k\right) e^{i(\phi+kz)}, \tag{4.46}$$

$$\frac{dp^*}{dz} = -\frac{d\Lambda}{dz} e^{-i(\phi+kz)} - i\Lambda\left(\frac{d\phi}{dz} + k\right) e^{-i(\phi+kz)}, \tag{4.47}$$

$$\frac{d^2p^*}{dz^2} = -\frac{d^2\Lambda}{dz^2} e^{-i(\phi+kz)} - i\Lambda\left(\frac{d\phi}{dz} + k\right) e^{-i(\phi+kz)} - i\,\frac{d\Lambda}{dz}\left(\frac{d\phi}{dz} +\right.$$
$$\left.k\right) e^{-i(\phi+kz)} - i\Lambda\frac{d^2\phi}{dz^2} e^{-i(\phi+kz)} + \Lambda\left(\frac{d\phi}{dz} + k\right) e^{-i(\phi+kz)}, \tag{4.48}$$

Following a thorough differentiation operation, we have obtained the first and second-order derivatives including for the complex conjugates likewise in equations (4.45), (4.46), (4.47) and (4.48). To solve the slowly varying envelope approximation (SVEA), we substitute these stated equations into equation (4.44). Moreover, by removing the second-order differential terms

$$(\Delta-\gamma i\omega)\left[\frac{d^2\Lambda}{dz^2}e^{i(\phi+kz)}+i\Lambda\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}+i\frac{d\Lambda}{dz}\left(\frac{d\phi}{dz}+\right.\right.$$

$$\left.\left.k\right)e^{i(\phi+kz)}+i\Lambda\frac{d^2\phi}{dz^2}e^{i(\phi+kz)}-\Lambda\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}\right]+$$

$$\frac{3}{4}\beta\left\{pp\left[-\frac{d^2\Lambda}{dz^2}e^{-i(\phi+kz)}-i\Lambda\left(\frac{d\phi}{dz}+k\right)e^{-i(\phi+kz)}-i\frac{d\Lambda}{dz}\left(\frac{d\phi}{dz}+\right.\right.\right.$$

$$\left.\left.k\right)e^{-i(\phi+kz)}-i\Lambda\frac{d^2\phi}{dz^2}e^{-i(\phi+kz)}+\Lambda\left(\frac{d\phi}{dz}+k\right)e^{-i(\phi+kz)}\right]+$$

$$4p\left[\frac{d\Lambda}{dz}e^{i(\phi+kz)}+i\Lambda\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}\right]\left[-\frac{d\Lambda}{dz}e^{-i(\phi+kz)}-i\Lambda\left(\frac{d\emptyset}{dz}+\right.\right.$$

$$\left.\left.k\right)e^{-i(\phi+kz)}\right]+2pp^*\left[\frac{d^2\Lambda}{dz^2}e^{i(\phi+kz)}+i\Lambda\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}+\right.$$

$$i\frac{d\Lambda}{dz}\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}+i\Lambda\frac{d^2\phi}{dz^2}e^{i(\phi+kz)}-\Lambda\left(\frac{d\phi}{dz}+\right.$$

$$\left.\left.k\right)e^{i(\phi+kz)}\right]+2p^*\left[\frac{d\Lambda}{dz}e^{i(\phi+kz)}+i\Lambda\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}\right]\left[\frac{d\Lambda}{dz}e^{i(\phi+kz)}+\right.$$

$$\left.\left.i\Lambda\left(\frac{d\phi}{dz}+k\right)e^{i(\phi+kz)}\right]\right\}=-\frac{\omega^2}{c^2}\left\{(\Delta-\gamma i\omega p)+\frac{3}{4}\beta p^*pp\right\}-\frac{\omega^2}{c^2}\frac{e^2}{\epsilon_0 m}p, \tag{4.49}$$

We remove the first order differential terms and divide the real and imaginary parts. Hence, two first-order differential questions will be obtained. Thus, by integrating both sides of the equation, we emerge the slowly varying envelope solution (SVES),

$$\ln\left[\frac{\Lambda(z)}{\Lambda_0}\right]+\left\{\frac{3}{2}\beta\Delta[\Lambda^2(z)-\Lambda_0^2]+\frac{27}{64}\beta^2[\Lambda^4(z)-\Lambda_0^4]\right\}[\Delta^2+\gamma^2\omega^2]^{-1}=-\alpha z, \tag{4.50}$$

and,

$$\phi(z)=\phi_0+\left(\frac{1}{2k}\right)\left[\frac{\omega^2}{c^2}-k^2\right]z-\left(\frac{1}{\gamma\omega}\right)\left\{\Delta ln\left[\frac{\Lambda_z}{\Lambda_0}\right]+\frac{9}{8}\beta[\Lambda^2(z)-\Lambda_0^2]\right\}, \tag{4.51}$$

where for small loss,

$$k = \frac{\omega}{c}\left[1 + \frac{\omega_p^2 \Delta}{\Delta^2 + \gamma^2 \omega^2}\right]^{\frac{1}{2}} \equiv \frac{n_0 \omega}{c}, \tag{4.52}$$

and to satisfy the usual (linear oscillator) conditions,

$$\alpha = \gamma^2 \omega^3 \omega_p^2 [2kc^2(\Delta^2 + \gamma^2 \omega^2)], \tag{4.53}$$

Consequently, Λ_0 and ϕ_0 represent the amplitude and phase of $p(z)$ at $z = 0$, while $\omega_p \left[\frac{4\pi N e^2}{m}\right]^{\frac{1}{2}}$ represents the material's plasma frequency. Equations (4.50) and (4.51) establish $p(z)$, and equation (4.34) subsequently produces $E(z)$, from which it is able to quantify the energy flux in the nonlinear medium likewise,

$$\bar{S} = \left(\frac{cm^2\Lambda^2}{8\pi e^2}\right)\left\{\left[n_0 + \left(\frac{c\phi'}{\omega}\right)\right]\left[\left(\Delta + \frac{3}{4}\beta\Lambda^2\right)^2 + \gamma^2\omega^2\right] + \frac{3}{2}\beta\gamma c\Lambda\Lambda'\right\}, \tag{4.54}$$

where $\Lambda' = \frac{d\Lambda}{dz}$ and $\phi' = \frac{d\phi}{dz}$ which can be obtained from the differentiation of equations (4.50) and (4.51) respectively.

CHAPTER 5 ANALYSIS AND DISCUSSION

In this section, we generate graphs my manipulating the equations (4.51), (4.51) and (4.55). In order to solve the stated equations, we need to substitute the values of the parameters of the equations. The values for the parameters are based on the optical properties of zinc lead tellurite glass obtained from [13]. Using python programming language and Google Collaboratory, we find the roots of the equations to generate a bistable graph and describe the theory of optical bistabilty.in nonlinear medium.

5.1 Manipulation of Parameters

Observing equation (4.51) and (4.52), we can see that, β, Δ, γ^2, ω^2, α, and k are considered constants while $\Lambda(z)$, Λ_0, $\phi(z)$, and ϕ_0 are variables. In order to solve the equations and find $p(z)$, we need a specific value for the constants. Hence, we have manipulated the parameters and assumed that the parameters are optical properties of zinc lead tellurite glass. To be precise, we have recorded the parameters and their respective values in a table.

Parameters	**Values**
β	7.20×10^{11}
γ	7.20×10^{10}
Δ	1.579
ω_0	7.20×10^{12}Hz
ω_p	5.64×10^{8}Hz

Table 5.1: Manipulation of parameters based on optical properties of zinc lead tellurite glass

5.2 Finding Roots

Using the manipulated parameters stated in table (5.1), we have solved equation (4.52) and (4.53) to obtain,

$$k = 60000cm^{-1}, \tag{5.1}$$

$$\alpha = 9.26 \times 10^{-21} \tag{5.2}$$

where $c = 3 \times 10^{8} ms^{-1}$. Therefore, equation (4.50) will become,

$$2.687 \times 10^{47} \ln\Lambda(z) + 2.187 \times 10^{23}\Lambda^{4}(z) + 1.706\Lambda^{2}(z) + 2.488 \times 10^{68} = 0, \tag{5.3}$$

where $z = 0.5m$. To obtain the roots of the equation (5.3), we used Google Collaboratory which involves Phyton programming language since the equation is too complex to be solved by scientific calculators and highly time-consuming. Thus, by letting $\Lambda_0 = 0.1$, we have obtained four roots for the stated equation. However, it must be taken into note that we have obtained the value using try and error method.

Λ_0	$\Lambda(z)\ (Cm^{-2})$
0.1	$\Lambda(z)_1 = 1.4919 \times 10^{-6}$
	$\Lambda(z)_2 = 5.7918 \times 10^{-3}$
	$\Lambda(z)_3 = 13.5586 \times 10^{-3}$
	$\Lambda(z)_4 = 12.6821 \times 10^{-3}$

Table 5.2: The values of $\Lambda(z)$ obtained

Now, since we have the values of $\Lambda(z)$, we proceed to solve the equation (4.51). By substituting the parameters again, the equation becomes,

$$\phi(z) = \phi_0 + \left(\frac{1}{2(60000)}\right)\left[\frac{(7.20\times10^{12})^2}{(3\times10^8)^2} - (60000)^2\right]0.5 - \left(\frac{1}{(7.20\times10^{10})(7.20\times10^{12})}\right)\left\{(1.579)ln\left[\frac{\Lambda_z}{(0.1)}\right] + \frac{9}{8}(7.20\times10^{11})[\Lambda^2(z) - (0.1)^2]\right\}, \tag{5.4}$$

$$\phi(z) = \phi_0 - 9.60\times10^{-19} - 1.929\times10^{-24}\left\{(1.579)ln\left[\frac{\Lambda_z}{(0.1)}\right] + 8.10\times10^{11}[\Lambda^2(z) - 0.01]\right\}, \tag{5.5}$$

By assuming the value based on try and error method, we let $\phi_0 = 0.5$ and substituting every Λ_z from table (5.2) one by one, we obtain,

ϕ_0	$\phi(z)$
0.5	$\phi(z)_1 = 6.69$
	$\phi(z)_2 = 3.01$
	$\phi(z)_3 = 2.38$
	$\phi(z)_4 = 2.50$

Table 5.3: The values of $\phi(z)$ obtained

Detailed calculations of this section and Python programming notebook are shown in appendix A.

5.3 Polarization and Energy Flux

Since we have solved both equations and obtained the necessary variables, we proceed to find the $p(z)$. Bringing back equation (4.38), and substituting the constants into the equation,

$$p(z) = \Lambda(z)\mathrm{e}^{\{i[\phi(z)+2.4568\times10^{-6}]\}} \tag{5.6}$$

Hence by inputting the variables from tables (5.2) and (5.3), we obtain,

$\Lambda(z)\ (Cm^{-2})$	$\phi(z)$	$p(z)(Cm^{-2})$
$\Lambda(z)_1 = 1.4919\times10^{-6}$	$\phi(z)_1 = 6.69$	$p(z)_1 = 0.0012$
$\Lambda(z)_2 = 5.7918\times10^{-3}$	$\phi(z)_2 = 3.01$	$p(z)_2 = 0.1175$
$\Lambda(z)_3 = 13.5586\times10^{-3}$	$\phi(z)_3 = 2.38$	$p(z)_3 = 0.1465$
$\Lambda(z)_4 = 12.6821\times10^{-3}$	$\phi(z)_4 = 2.50$	$p(z)_4 = 0.1545$

Table 5.4: The values of $p(z)$ obtained

We have obtained the values of $p(z)$ and now we have to solve the energy flux equation as in equation (4.54) to generate the optical bistability graph. However, we still in need of few parameters which are $\Lambda(z)'$ and $\phi(z)'$. They can be gained by differentiating equations (5.3) and (5.5), Hence, we substitute the derivatives into equation (4.55) along with other parameters,

$$\bar{S} = \left(\frac{8.075\times10^{9}}{8\pi e^{2}}\right)\left\{\left[1.58 + \left(\frac{(3\times10^{8})\phi'}{(7.20\times10^{12})}\right)\right][(1.579 + 5.4\times10^{10}\Lambda^{2})^{2} + 2.69\times10^{47}] + 2.33\times10^{31}\Lambda - \Lambda'\right\}, \tag{5.7}$$

where $n_0 = 1.58$ and $m = 164.06\ g/mol$. $\Lambda(z)'$ is ignored since the value is too large to evaluate or infinite. Therefore, using the variables we have obtained before, energy flux,

$\Lambda(z)\ (Cm^{-2})$	$\phi(z)$	$\bar{S}\ (Wb)$
$\Lambda(z)_1 = 1.4919 \times 10^{-6}$	$\phi(z)_1 = 6.69$	$\bar{S}_1 = 2.49 \times 10^{10}$
$\Lambda(z)_2 = 5.7918 \times 10^{-3}$	$\phi(z)_2 = 3.01$	$\bar{S}_2 = 1.07 \times 10^{16}$
$\Lambda(z)_3 = 13.5586 \times 10^{-3}$	$\phi(z)_3 = 2.38$	$\bar{S}_3 = 2.79 \times 10^{14}$
$\Lambda(z)_4 = 12.6821 \times 10^{-3}$	$\phi(z)_4 = 2.50$	$\bar{S}_4 = 1.87 \times 10^{16}$

Table 5.5: The values of $\bar{S}$ obtained

Further and detailed calculations are demonstrated in appendix B.

5.4 Graph of Optical Bistability in Nonlinear Medium

We have successfully solved the slowly varying envelope solution (SVES) and energy flux equation. Hence, we have obtained the required variable which are $p(z)$ and $\bar{S}$ to plot the graph to demonstrate the behaviour of optical bistability in nonlinear medium. Therefore, using the $p(z)$ from table (5.4) and $\bar{S}$ from table (5.5),

$p(z)(Cm^{-2})$	$\bar{S}\ (Wb)$
$p(z)_1 = 0.0012$	$\bar{S}_1 = 2.49 \times 10^{10}$
$p(z)_2 = 0.1175$	$\bar{S}_2 = 1.07 \times 10^{16}$
$p(z)_3 = 0.1465$	$\bar{S}_3 = 2.79 \times 10^{14}$
$p(z)_4 = 0.1545$	$\bar{S}_4 = 1.87 \times 10^{16}$

Table 5.6: Parameters to plot $p(z)$ versus $\bar{S}$ graph

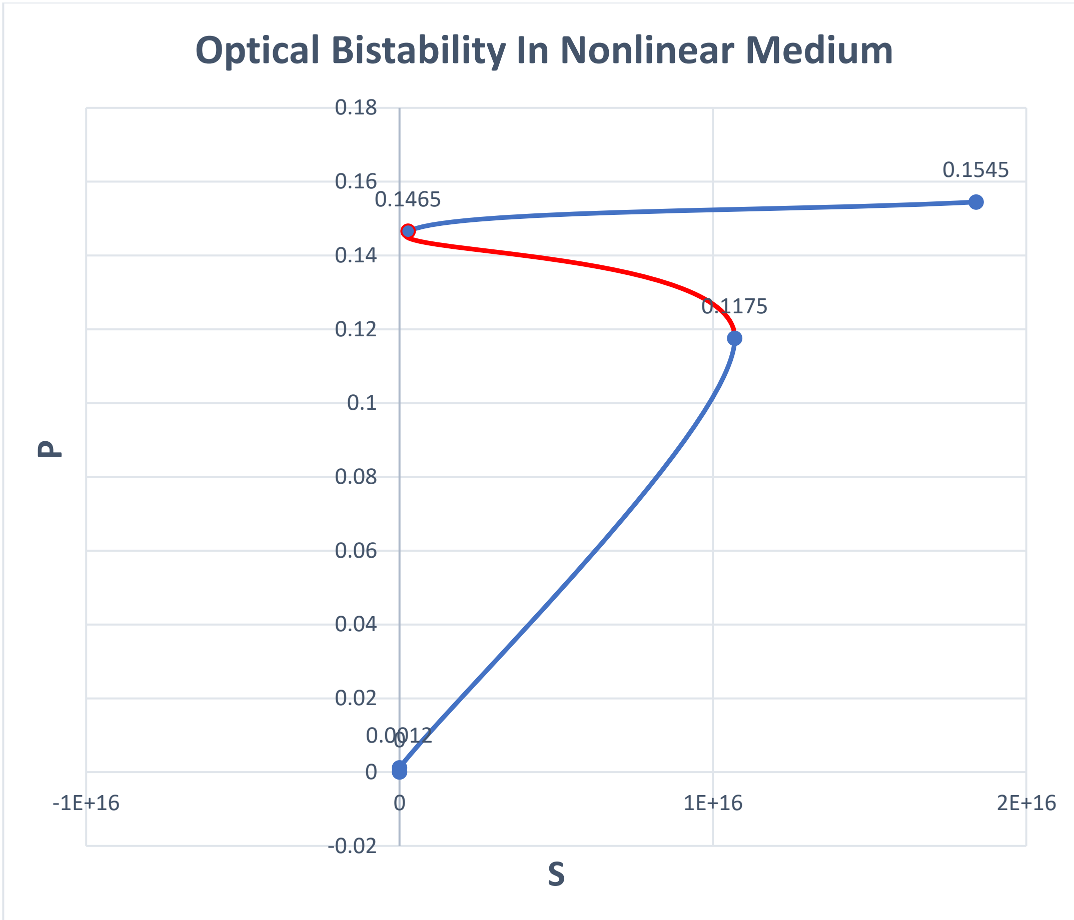

Figure 5.1: Optical Bistability in Nonlinear Medium Graph

From this graph, we can see that the curve (red line) experienced bistability from $P = 0.1175$ to $P = 0.1465$ and became stable after that point. Hence, it is clear that optical bistability exists in zinc lead tellurite glass. However, it is important to take note that the data and method used to generate the graph may not be accurate, but the fundamental material frequency response always has its distinctive "S shape".

CHAPTER 6 CONCLUSION

6.1 Conclusion

This paper has replicated and analysed the research of J. A. Goldstone and E. Garmire [1] in detail. We have derived and calculated the polarization and electric linear susceptibility, Maxwell's equation, slowly varying envelope solution (SVES), and energy flux equation that governs the nonlinear medium's optical properties. However, this work is limited since it is out of the scope of our work. We found the roots of the equations using python programming language. Moreover, we used the optical properties of zinc lead tellurite glass $([Z_nO]_x[(T_eO_2)_{0.7} - (P_bO)_{0.3}]_{1-x})$ as our medium to project our results based on the research conducted by the authors in [1]. Wherever possible, we used experimental data that was readily available to create the graphs to give a realistic picture of the behaviour of zinc lead tellurite glass.

According to [1], it should be emphasised that this form of bistability cannot be described using the typical nonlinear optics techniques that lead to power series expansions in terms of the electric fields. Similar techniques must be used to handle the potentially major class of bistable interactions that occur. Finding adequate nonlinear constitutive relations becomes the only remaining issue.

6.2 Further Avenues of Research

As mentioned in this paper, we have expressed our interest in continuing this research whenever possible. According to [10], even though optical bistability is well known and researched among physicists and mathematicians, we strongly believe there is plenty to study in this subject. Furthermore, we are limited to research until slowly varying envelope solution (SVEA) and energy flux in nonlinear medium but bistable loops has always been an option, but due to time constraints, we have not explored this.

This method employing a phenomenological model is quite complex but unique. Its uniqueness can inspire many to explore this fascinating field and theory more. This approach could prove to be highly beneficial in the growth of this discipline, given the steadily increasing interest and the emerging applications.

REFERENCES

[1] Goldstone, J. A., & Garmire, E. (1984). Intrinsic Optical Bistability in Nonlinear Media. *Physical Review Letters*, *53*(9), 910–913. https://doi.org/10.1103/physrevlett.53.910

[2] Bassani, G. (2005). *Encyclopedia of Condensed Matter Physics (6 Vol set))* (1st ed.). Academic Press.

[3] Asquini, M. L., & Casagrande, F. (1981). Optical bistability in a bidirectional ring cavity. *Zeitschrift Fur Physik B Condensed Matter*, *44*(3), 233–239. https://doi.org/10.1007/bf01297180

[4] Angelis, D. C. (2021). *Nonlinear Optics*. Frontiers. https://www.frontiersin.org/articles/10.3389/fphot.2020.628215/full#B4

[5] Boyd, R. W., & Masters, B. R. (2009). Nonlinear Optics, Third Edition. *Journal of Biomedical Optics*, *14*(2), 029902. https://doi.org/10.1117/1.3115345

[6] Chaos: a program collection for the PC. (2008). *Choice Reviews Online*, *45*(11), 45–6121. https://doi.org/10.5860/choice.45-6121

[7] Doulcier, G. (2019, March 18). *Numerical bifurcation diagrams and bistable systems*. Modelling. https://www.normalesup.org/%7Edoulcier/teaching/modeling/bistable_systems.html

[8] Franken, P. A., Hill, A. E., Peters, C. W., & Weinreich, G. (1961a). Generation of Optical Harmonics. *Physical Review Letters*, *7*(4), 118–119. https://doi.org/10.1103/physrevlett.7.118

[9] Franken, P. A., Hill, A. E., Peters, C. W., & Weinreich, G. (1961b). Generation of Optical Harmonics. *Physical Review Letters*, *7*(4), 118–119. https://doi.org/10.1103/physrevlett.7.118

[10] Gardner, T. S., Cantor, C. R., & Collins, J. J. (2000). Construction of a genetic toggle switch in Escherichia coli. *Nature*, *403*(6767), 339–342. https://doi.org/10.1038/35002131

[11] McCall, S. (1987). Controlling light with light. *Journal of Luminescence*, *37*(2), 115. https://doi.org/10.1016/0022-2313(87)90173-6

[12] Greene, W. P., Gibbs, H. M., Passner, A., McCall, S. L., & Venkatesan, T. N. C. (1980). Optical Bistability: An Undergraduate Experiment. *Optics News*, *6*(2), 16. https://doi.org/10.1364/on.6.2.000016

[13] An introduction to optically bistable devices and photonic logic. (1984). *Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences*, *313*(1525), 195–204. https://doi.org/10.1098/rsta.1984.0096

[14] ITOH, T. (2000). Fundamentals of Laser Spectroscopy. II. Luminescence Spectroscopy. *The Review of Laser Engineering*, *28*(1), 54–59. https://doi.org/10.2184/lsj.28.54

[15] Li, C. (2018). *Nonlinear Optics: Principles and Applications* (Softcover reprint of the original 1st ed. 2017 ed.). Springer.

[16] Alazoumi, S. H., Aziz, S. A., El-Mallawany, R., Aliyu, U. S., Kamari, H. M., Zaid, M. H. M. M., Matori, K. A., & Ushah, A. (2018). Optical properties of zinc lead tellurite glasses. *Results in Physics*, *9,* 1371–1376. https://doi.org/10.1016/j.rinp.2018.04.041

[17] Musgraves, D. J., Hu, J., & Calvez, L. (2019). *Springer Handbook of Glass (Springer Handbooks)* (1st ed. 2019 ed.). Springer.

[18] Ozbudak, E. M., Thattai, M., Lim, H. N., Shraiman, B. I., & van Oudenaarden, A. (2004). Multistability in the lactose utilization network of Escherichia coli. *Nature*, *427*(6976), 737–740. https://doi.org/10.1038/nature02298

[19] Wang, Z., & Yu, B. (2015). Optical bistability in triple quantum dot molecules in weak tunneling regime. *Superlattices and Microstructures*, *84*, 45–53. https://doi.org/10.1016/j.spmi.2015.04.033

[20] Wikipedia contributors. (2022, June 27). *Slowly varying envelope approximation*. Wikipedia. https://en.wikipedia.org/wiki/Slowly_varying_envelope_approximation#cite_note-1

[21] Zhong, L., Shu, S., Wang, J., & Xu, J. (2012). Two-grid methods for time-harmonic Maxwell equations. *Numerical Linear Algebra with Applications*, *20*(1), 93–111. https://doi.org/10.1002/nla.1827

APPENDICES

Appendix A Finding Root

Google Collaboratory Notebook:

Importing necessary library to begin with:

```
from scipy.optimize import fsolve
```

Defining the equation and manipulating the parameters into a single parameter to find the roots:

```
f = lambda x: 3.029*x + 2.7183**18.6564*(x**4) + 2.71828**2.5287*(x**2) + 1.497**17
```

```
fsolve(f, [2,80,160,140])
```

```
/usr/local/lib/python3.7/dist-packages/scipy/optimize/minpack.py:175: RuntimeWarning: Tl
  improvement from the last ten iterations.
  warnings.warn(msg, RuntimeWarning)
array([0.00012317, 0.00677934, 0.01355867, 0.01186384])
```

Calculation of $\phi(z)$:

$$\phi(z) = \phi_0 + \left(\frac{1}{2(60000)}\right)\left[\frac{(7.20 \times 10^{12})^2}{(3 \times 10^8)^2} - (60000)^2\right]0.5$$

$$- \left(\frac{1}{(7.20 \times 10^{10})(7.20 \times 10^{12})}\right)\left\{(1.579)ln\left[\frac{\Lambda_z}{(0.1)}\right]\right.$$

$$\left. + \frac{9}{8}(7.20 \times 10^{11})[\Lambda^2(z) - (0.1)^2]\right\}$$

$$\phi(z) = \phi_0 - 9.60 \times 10^{-19}$$

$$- 1.929$$

$$\times 10^{-24}\left\{(1.579)ln\left[\frac{\Lambda_z}{(0.1)}\right]\right.$$

$$\left. + 8.10 \times 10^{11}[\Lambda^2(z) - 0.01]\right\}$$

$$\phi(z)_1 = 0.5 - 9.60 \times 10^{-19} - 1.929 \times 10^{-24}\left\{(1.579)ln\left[\frac{1.4919 \times 10^{-6}}{(0.1)}\right] + 8.10 \times 10^{11}[(1.4919 \times 10^{-6})^2 - 0.01]\right\}$$

$$\phi(z)_1 = 6.69$$

$$\phi(z)_2 = 0.5 - 9.60 \times 10^{-19} - 1.929 \times 10^{-24}\left\{(1.579)ln\left[\frac{5.7918 \times 10^{-3}}{(0.1)}\right] + 8.10 \times 10^{11}[(5.7918 \times 10^{-3})^2 - 0.01]\right\}$$

$$\phi(z)_2 = 3.01$$

$$\phi(z)_3 = 0.5 - 9.60 \times 10^{-19} - 1.929 \times 10^{-24}\left\{(1.579)ln\left[\frac{13.5586 \times 10^{-3}}{(0.1)}\right] + 8.10 \times 10^{11}[(13.5586 \times 10^{-3})^2 - 0.01]\right\}$$

$$\phi(z)_3 = 2.38$$

$$\phi(z)_4 = 0.5 - 9.60 \times 10^{-19} - 1.929 \times 10^{-24}\left\{(1.579)ln\left[\frac{12.6821 \times 10^{-3}}{(0.1)}\right] + 8.10 \times 10^{11}[(13.5586 \times 10^{-3})^2 - 0.01]\right\}$$

$$\phi(z)_4 = 2.50$$

Appendix B Polarization and Energy Flux

Calculation of $p(z)$:

$$p(z) = \Lambda(z)\mathrm{e}^{\{i[\phi(z)+60000]\}}$$

$$p(z)_1 = \Lambda(z)_1\mathrm{e}^{\{i[\phi(z)_1+60000]\}}$$

$$p(z)_1 = (1.4919 \times 10^{-6})\mathrm{e}^{\{i[6.69+60000]\}}$$

$$p(z)_1 = 0.0012(Cm^{-2})$$

$$p(z)_2 = \Lambda(z)_2\mathrm{e}^{\{i[\phi(z)_2+60000]\}}$$

$$p(z)_2 = (5.7918 \times 10^{-3})\mathrm{e}^{\{i[3.01+60000]\}}$$

$$p(z)_2 = 0.1175(Cm^{-2})$$

$p(z)_3 = \Lambda(z)_3 \mathrm{e}^{\{i[\phi(z)_3+60000]\}}$

$p(z)_3 = (13.5586 \times 10^{-3})\mathrm{e}^{\{i[2.38+60000]\}}$

$p(z)_3 = 0.1465(Cm^{-2})$

$p(z)_4 = \Lambda(z)_4 \mathrm{e}^{\{i[\phi(z)_4+60000]\}}$

$p(z)_4 = (12.6821 \times 10^{-3})\mathrm{e}^{\{i[2.50+60000]\}}$

$p(z)_4 = 0.1545(Cm^{-2})$

Calculation of $\bar{S}$:

$$\bar{S} = \left(\frac{8.075\times10^9}{8\pi e^2}\right)\left\{\left[1.58 + \left(\frac{(3\times10^8)\phi'}{(7.20\times10^{12})}\right)\right][(1.579 + 5.4\times10^{10}\Lambda^2)^2 + 2.69\times10^{47}] + 2.33\times10^{31}\Lambda - \Lambda'\right\},$$

$$\bar{S}_1 = \left(\frac{8.075\times10^9}{8\pi e^2}\right)\{[1.58 + 11952 \times 10^8][(1.579 + 5.4\times10^{10}(1.4919\times10^{-6})^2)^2 + 2.69\times10^{47}] + 2.33\times10^{31}(1.4919\times10^{-6})\}$$

$\bar{S}_1 = 2.49 \times 10^{10} Wb$

$$\bar{S}_2 = \left(\frac{8.075 \times 10^9}{8\pi e^2}\right)\{[1.58 + 11952 \times 10^8][(1.579 + 5.4 \times 10^{10}(5.7918 \times 10^{-3})^2)^2 + 2.69 \times 10^{47}] + 2.33 \times 10^{31}(5.7918 \times 10^{-3})\}$$

$$\bar{S}_2 = 1.07 \times 10^{16} Wb$$

$$\bar{S}_3 = \left(\frac{8.075 \times 10^9}{8\pi e^2}\right)\{[1.58 + 11952 \times 10^8][(1.579 + 5.4 \times 10^{10}(13.5586 \times 10^{-3})^2)^2 + 2.69 \times 10^{47}] + 2.33 \times 10^{31}(13.5586 \times 10^{-3})\}$$

$$\bar{S}_3 = 2.79 \times 10^{14} Wb$$

$$\bar{S}_4 = \left(\frac{8.075 \times 10^9}{8\pi e^2}\right)\{[1.58 + 11952 \times 10^8][(1.579 + 5.4 \times 10^{10}(12.6821 \times 10^{-3})^2)^2 + 2.69 \times 10^{47}] + 2.33 \times 10^{31}(12.6821 \times 10^{-3})\}$$

$$\bar{S}_4 = 1.87 \times 10^{16} Wb$$