UNIVERSITI PUTRA MALAYSIA

NONLINEAR OPTICAL PROPERTIES OF NANO-FLUID AND PHOSPHATE GLASS USING Z-SCAN TECHNIQUE

MASOUMEH SHOKATI MOJDEHI

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By

MASOUMEH SHOKATI MOJDEHI

Thesis Submitted to the School Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirement for the Degree of Master of Science

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This thesis is dedicated to

my beloved parents (Gholam Reza Shokati and Shamsi Shafaghi)

and

my dear brother (Mohammad Reza Shokati) who support me with their

endless love to carry out my education.

Abstract of the thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of requirement for degree of Master of Science

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October 2013

Chairman: Professor W. Mahmood Mat Yunus, PhD

Faculty: Science

The nonlinear optical properties of aluminum oxide-chromium/nickel nano-fluids and ternary zinc-magnesium-phosphate glass (obtained from other group) were investigated in this study utilizing a single-beam Z-scan method. The nonlinear refractive index, absorption coefficient, third-order nonlinear susceptibility, and figure of merit of the samples were also measured.

The nonlinear refractive indices of ethanol-based aluminum oxide-chromium nano-fluids prepared with mass fraction concentrations ranging from 0.018 to 0.146 wt%, were studied. The obtained values ranged from -0.19 $\times 10^{-9}$ to -1.99 $\times 10^{-9}$ cm^2/W, with negative nonlinearity. Thus, the samples exhibited self-defocusing phenomena. The nonlinear refractive index increases slowly with increasing concentration from 0.018 to 0.036 wt%, rapidly increasing from 0.036 to 0.073 wt%, and then from 0.073

to 0.146 wt%. The nonlinear absorption of the samples showed saturable absorption caused by the peak configuration at the focal point in order of 10^{-5}. The real and imaginary parts of third-order nonlinear susceptibility were also calculated. The value of figure of merit for each sample were calculated that satisfied the condition *FOM* <1. The nonlinear refractive index and absorption of ethylene-glycol-based aluminum oxide–chromium nano-fluid were investigated at mass fraction concentrations ranging from 0.013 to 0.104 wt%. The aluminum oxide–chromium nano-fluid revealed a good nonlinear behavior, with a negative nonlinear refractive index ranging from -0.30 $\times10^{-10}$ cm^2/W to -2.86 $\times10^{-10}$ cm^2/W. This nonlinearity increased linear gradually with an increasing concentration of sample. Furthermore, the nonlinear absorption behavior showed a saturable absorption caused by the peak configuration at the focal point. The magnitudes of figure of merit for the sample were calculated that satisfied the condition *FOM*<1. The nonlinear optical properties of ethanol-based aluminum oxide-nickel nano-fluid at different mass fraction concentrations ranging from 0.018 to 0.146 wt% were characterized. A negative nonlinear refractive index was obtained, and a self-defocusing phenomenon was observed at 405 nm from -1.36 $\times10^{-9}$ to -3.41 $\times10^{-9}$ cm^2/W. The nonlinear refractive index was increased approximate linearly (moderate trend). In case of open aperture transmittance data, no nonlinear behavior was observed in the absorption coefficient. Furthermore, the real part of third-order nonlinear susceptibility was presented in order 10^{-6} that depended on nonlinear refractive behavior of aluminum oxide- nickel nano-fluid.

The nonlinear optical properties of a phosphate glass system $[(ZnO)_x–(MgO)_{30-x}–(P_2O_5)_{70}]$, where x = 8, 10, 15, 18 and 20 mol%, with synthesized melt-quenching (obtained from other group) were investigated. In this experiment, a 405 nm CW laser was operated to determine both the sign and value of the nonlinear refractive index and absorption coefficient. The nonlinear refractive index was found to vary from 0.45 $\times 10^{-10}$ to 2.11 $\times 10^{-10}$ cm^2/W. The nonlinear refractive index value increased with increasing ZnO concentration in the glass samples. From the measurement, the positive nonlinear refractive index demonstrated self-focusing phenomenon. The nonlinear absorption behavior showed reverse saturable absorption caused by the valley configuration at the focal point. The value of figure of merit was presented to attribute the potential of the samples for utilizing in optical switching by following condition *FOM* <1. The nonlinear optical properties of phosphate glasses in $[(ZnO)_{30}-(MgO)_x-(P_2O_5)_{70-x}]$ (x = 5, 8, 13, 15 and 20 mol%) glassy system (obtained from other group) were investigated. The magnitude and the sign of nonlinear refractive index and nonlinear absorption coefficient were obtained from the closed and open aperture data, in which the magnitude of nonlinear refractive index ranged from 1.82 $\times 10^{-10}$ to 7.92 $\times 10^{-10}$ cm^2/W that the nonlinear refractive index increased. The glass samples demonstrated self-focusing phenomenon, as shown by the positive nonlinear refraction of index. The figure of merit for the sample were also calculated that satisfied the condition of *FOM*<1 to use in optical devices.

Abstrak tesis yang dikemukan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

SIFAT-SIFAT OPTIK TAKLINEAR BENDALIR-NANO DAN KACA POSFAT MENGGUNAKAN TEKNIK IMBASAN-Z

Oleh

MASOUMEH SHOKATI MOJDEHI

October 2013

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Di dalam kajian ini, ciri-ciri optik taklinear oleh cecair-nano dan kaca fosfat di selidik dengan menggunakan teknik alur tunggal imbasan-Z. Indeks biasan taklinear, pekali penyerapan, kecenderungan tahap ketiga dan gambarajah merit daripada sampel telah diukur.

Indeks biasan taklinear yang ditunjukkan oleh cecair-nano aluminium kromium-oksida yang berasaskan etanol di kaji berdasarkan kepekatan jisim pecahan di dalam lingkungan 0.018 hingga 0.146 wt%. Nilai yang diperoleh adalah dalam lingkungan -0.19×10^{-9} hingga -1.99×10^{-9} cm^2/W, berserta taklinearan negatif. Oleh itu, kesemua sampel mengalami fenomena takfokusan-sendiri. Indeks biasan taklinear menunjukkan perkembangan menaik berkadar terus dengan kepekatan jisim pecahan sampel. Kepekatan sampel cecair-nano memberi kesan kepada indeks biasan taklinear. Indeks

biasan linear meningkat secara perlahan dengan peningkatan kepekatan dari 0.018 hingga to 0.036 wt% dan kemudian meningkat dengan pesat dari 0.036 hingga 0.073 untuk renj kepekatan 0.073 hingga 0.146 wt%. Sampel yang bersifat penyerap taklinear menunjukkan penyerapan tepu disebabkan oleh konfigurasi puncak di titik tumpu pada kadar 10^{-5}. Nilai angka merit bagi setiap sampel dikira untuk keadaan yang memenuhi *FOM*<1. Indeks biasan taklinear yang ditunjukkan oleh cecair-nano aluminium kromium-oksida yang berasaskan etilena-glycol di kaji berdasarkan kepekatan jisim pecahan di dalam lingkungan 0.013 hingga 0.104 wt%. Cecair-nano aluminium oksida-kromium menunjukkan tindakbalas taklinear yang bagus dengan nilai indeks biasan di antara -0.30×10^{-10} dan -2.86×10^{-10} cm^2/W. Ini menunjukkan bahawa peningkatan kepekatan sampel akan menaikan ketaklinearan bahan. Selain itu, perilaku penyerapan taklinear menunjukkan penyerapan tepu disebabkan oleh konfigurasi puncak di titik tumpu. Ciri-ciri taklinear yang ditunjukkan oleh cecair-nano aluminium nikel-oksida dikaji dengan menggunakan kepekatan jisim pecahan yang berbeza, dalam lingkungan 0.018 hingga 0.146 wt%. Indeks biasan negatif taklinear diperolehi dan kondisi takfokusan-sendiri diperhati pada 405 nm bermula dari -1.36×10^{-3} sehingga -3.41×10^{-9} cm^2/W. Tiada perilaku taklinear dilihat di pekali penyerapan. Ciri-ciri optik taklinear pada sistem kaca fosfat $[(ZnO)_x-(MgO)_{30-x}-(P_2O_5)_{70}]$, yang mana x= 8 , 10, 15, dan 20 mol%, oleh pelindapan-lebur sintesis dikaji. Di dalam eksperimen ini, laser CW 405 nm digunakan untuk mengenalpasti simbol dan nilai yang didapati daripada indeks biasan taklinear dan pekali penyerapan. Didapati indeks biasan taklinear berubah mulai daripada 0.45×10^{-10} hinggalah 2.11×10^{-10} cm^2/W. Peningkatan nilai indeks biasan taklinear adalah berkadar terus dengan kepekatan ZnO di dalam sampel kaca. Dari pengukuran, indeks biasan positif taklinear menunjukkan fenomena ketidakfokusan-sendiri. Perilaku penyerapan taklinear menunjukkan penyerapan tepu terbalik disebabkan oleh lembah konfigurasi di titik tumpu. Angka merit untuk sampel juga dikira bahawa ia memenuhi keadaan *FOM <1* untuk digunakan dalam peranti optik. Ciri-ciri optik taklinear oleh kaca fosfat di dalam $[(ZnO)_{30}\text{-}(MgO)_x\text{-}(P_2O_5)_{70-x}]$ (x= 5, 8, 13, 15 dan 20 mol%) sistem berkaca diselidiki. Nilai bagi indeks biasan taklinear dan pekali penyerapan untuk setiap sampel dikira dengan menggunakan laser diod CW 405 nm sebagai alur pengujaan. Magnitud dan simbol indeks biasan taklinear dan pekali penyerapan diperoleh melalui data buka dan tutup bukaan, di mana magnitud indeks biasan taklinear berada di dalam lingkungan antara 1.82×10^{-10} dan 7.92×10^{-10} cm^2/W. Sampel berkaca menunjukkan fenomena pemfokusan-sendiri seperti yang ditunjukkan oleh indeks biasan positif taklinear. Nilai merit untuk sampel juga dikira dengan syarat ia memneuhi *FOM<1* untuk diguna dalam peranti optik.

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I certify that a Thesis Examination Committee has met on 21 October 2013 to conduct the final examination of Masoumeh Shokati Mojdehi on her thesis entitled "Nonlinear Optical Properties of Nano-Fluid and Phosphate Glass Using Z-Scan Technique" 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 Master of Science.

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Date: 20 November 2013

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been acceptance fulfillment of the requirement for the degree of Master of Science. The members of the supervisory Committee were as follows:

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DECLARATION

I declare that the thesis is my original work except for quotation and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

MASOUMEH SHOKATI MOJDEHI

Date: 21 October 2013

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LIST OF ABBREVIATIONS

CW	Continuous wavelength
ESA	Excited state absorption
SA	Saturable absorption
RSA	Reverse saturable absorption
NLR	Nonlinear refraction
NLA	Nonlinear absorption
SAVE	Approximation of slowly-varying amplitude
n_2	Nonlinear refractive index
β	Nonlinear absorption coefficient
$\Delta\phi$	Phase change
w_a	Beam radius at focus point
χ	Susceptibility
λ	Wavelength
FOM	Figure of merit
K	Wave number

CHAPTER 1

INTRODUCTION

1.1 Nonlinear Optics

Classical and quantum physics, thermal physics, electrodynamics, and optics, among others, are prominent fields of study in physics. The addition of nonlinearity and nonlinear optical properties to physics has created a revolution in optics and photonics. Nonlinear optics is recently emerging as a rapidly developing trend in the scientific domain. Nonlinear optics is an extensive technology and research area that encompasses physics, chemistry, engineering, medicine, and so on (Miragliotta, 1995). In recent decades, nonlinear optical materials have become technologically in demand in a broad range of applications such as in communication parts (e.g., all-optical and electro-optical applications) (Sheik-Bahae et al., 1991), telecommunications (e.g., optical storage and all-optical computing) (De Boni et al., 2004) , and in laser technology (e.g., diode laser, frequency doubling devices [laser pointers], quantum-well DFB laser, and so on).

Nonlinear optics is an affiliate of laser technology and an attractive research area that has attracted attention from the scientific optical community. Laser intensity plays a significant role in achieving nonlinear optical properties of materials (Garmire, 2011). Therefore, sufficiently high laser intensity is required to modify the nonlinear optical properties of materials.

The first study in nonlinear optics was conducted last in 1875 by Kerr et al. who discovered the effect of quadratic electro-optic in glass. Nonlinear optics was presented by Franken in 1961 after the innovation of the first laser that initiated second-harmonic generation. The second-harmonic generation facilitated the possible alteration in the laser wavelength using second order procedure and a comprehensible adjustable laser source. This phenomenon is based on the concept of nonlinearity. In terms of second-harmonic generation, the utilized nonlinear optical materials must have an asymmetrical formation and a refractive index controllable by an external electric field, which is attributed to as an electro-optic effect. Third-order nonlinear phenomenon, as an example of Kerr effect, can be applied in fiber communication. However, many of its applications are in ultrafast data processing. For the third-order nonlinear effect, properties of nonlinear optical materials are controlled using a laser light that is operated in all-optical switching devices. The second- and third-harmonic generations are related to quadratic and cubic generation of the applied optics field intensity, respectively (Harris, 1988).

A significant concept in nonlinear optics is the behavior of light interaction in a nonlinear medium. Nonlinear optical properties of media are defined when their polarization response to an optical field relates to a nonlinear mode by the electric field intensity. In a medium, the electric field of light responds to dielectric polarization, which is investigated by high light intensity. In linear optics, the electric field of the incident beam is weak, whereas in nonlinear optics, the intensity of electric field is strongly adequate. Thus, polarization and electric field have a nonlinear relationship.

Nonlinear optics is generally used to study fundamental interactions between the strength of light and material using laser technology. The majority of studies in optics cover a great variety of the nonlinear optical phenomenon. The present study is principally concerned with nonlinear refraction (NLR) and nonlinear absorption (NLA).

1.2 Nonlinear Refraction

Refraction is the change in the direction of a wave as it passes between two different media as a result of the alteration in its speed at any angle between 0° and 90°. This condition is depicted by Snell's law, in which the proportional relation between the angle of incidence in medium 1 (θ_1) and the angle of refraction in medium 2 (θ_2) is equal to the ratio of the phase velocities in the media. Snell's law is expressed as (Lyer, 1998):

$$\frac{Sin\theta_1}{Sin\theta_2} = \frac{n_2}{n_1} = \frac{v_1}{v_2} \tag{1.1}$$

where v and n represent velocity and index refraction, respectively.

Thus, refraction is used to describe how the medium can change the propagation of light rays. Under high intensity, the index of refraction is correlated with the reaction of light irradiance in nonlinear media, which is described by the following related equation (Rad et al., 2012):

$$n = n_0 + \Delta n(I) \tag{1.2}$$

where n_0 and $\Delta n(I)$ represent the linear and nonlinear refractive indices, respectively, and (I) symbolizes the optical irradiance, in which the irradiance is supplied by an electromagnetic field of laser beam. In this case, if the irradiance is sufficiently low, then Δn can be negligible and the nonlinear refraction expresses disturbance to the linear response of the material. The nonlinear refractive term plays a significant role in designing ultrafast switching devices, which are currently employed in modern optical communication systems. In the past few years, numerous studies have focused on exploring media that may possess contributions of intensity dependence to the refractive index.

1.3 Nonlinear Absorption

Nonlinear absorption is a process in which light changes the absorptive property of a material (Stryland & Hagan, 2003)**.** Nonlinear absorption relates to the transmittance change in materials as a function of intensity. In this case, a sufficiently high intensity provides the probability to absorb more than one photon before relaxing to the ground state within the material. Intense laser fields influence population redistribution, which conduct concepts, stimulate emission and absorption, and so on. Nonlinear absorption is related to the imaginary part of a third-order nonlinear susceptibility (Sheik-Bahae et al., 1991), whereas the nonlinear refraction is associated with the real part of a third-order nonlinear susceptibility (Sanghera et al., 2008). The nonlinear optical phenomenon is revealed by either saturable or reverse saturable absorption configuration (Majles Ara et al., 2010) .

1.3.1 Excited State Absorption

The condition of an atom or a molecule after absorbing energy from light, electricity, and elevated temperature may be essential in generating emission light in its excited state. In physics, the energy level in a system that is higher than the ground state is called an excited state. The system decays to the ground state and emits energy, generally in photon form (Bello, 2013). When light is absorbed by the system, each atom is taken up by an atom or molecule such that the complete energy of a quantum is transmitted to it. During energy absorption, an atom or molecule becomes excited as it is lifted from its regular ground state, which has the lowest energy, to a higher state.

An atom or molecule from ground state S_0 is excited to the first excited state S_1 by a photon that possesses sufficient energy, as shown in Figure 1.1. Given that the poor matching energy between photon A and state S_1 is inside the natural line width, the excited electrons can stay in the first excited state S_1 for approximately 10^{-3} s and 10^{-12} s . Therefore, during this period, by the arrival of photon B, the electron transition can be terminated to S_2. Hence, the process by which the atom absorbs photons that promotes it to a higher state is called excited state absorption (ESA) (Rao et al., 2011).

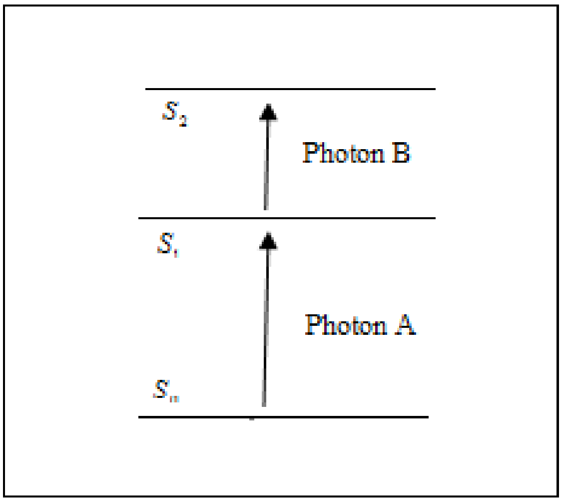

Figure 1.1: Excited state absorption (ESA) scheme diagram

1.3.2 Saturable Absorption

Saturable absorption is the property of a material in which transmittance is increased by an increase in optical intensity when a material is scanned by laser light (Venkatram et al., 2005). This property can be observed in the majority of materials. However, it occurs in high light intensity. In a sufficiently high light intensity, an atom in the ground state of a saturable absorbing material becomes excited to a higher energy level at such a rate that it cannot decay back to the ground state because of insufficient energy. Thus, and the ground state becomes empty.

According to the phenomenology of saturable absorption, the absorption cross-section (an absorption process probability measurement) from an excited state σ_1 is smaller than that from a ground state σ_0 (Majles Ara et al., 2010). The transmission is raised when the system is scanned by a high-intensity laser beam. This process is called saturable absorption (Figure 1.2).

1.3.3 Reverse Saturable Absorption

If the absorption cross-section from an excited state σ_1 is larger than that from a ground state σ_0, the transmission is decreased by increasing optical intensity when a material is scanned by a high-intensity laser beam (Venkatram et al., 2005). This procedure is called reverse saturable absorption (RSA), which occurs when an absorption cross-section σ passes that of the ground state. This condition generally results from the process of an inter-system crossing from excited states and contesting to a single ground state (Alikhani et al., 2012). This phenomenon corresponds to excited state properties such as intersystem crossing time and cross-section. The schematic diagram of this process is shown in Figure 1.2.

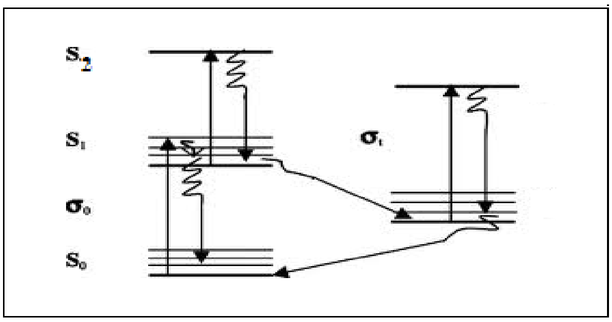

Figure 1.2: Schematic diagram of electronic transitions for SA and RSA (Ali & Palanisamy, 2006)

1.4 Single-beam Z-scan Characterization

The single-beam Z-scan method is presented as a popular and simple method for measuring nonlinearities (Natarajan et al., 2010). This method has been used to investigate the behavior of laser propagation within materials and is based on

Gaussian decomposition method. A high-intensity laser beam is applied to scan a nonlinear sample through the focal plane utilizing a tightly focused laser beam in the far field. Therefore, it can induce nonlinear refraction changes, which either causes the self-focusing or self-defocusing phenomena. The single beam Z-scan method is activated to characterize the nonlinear refractive index and the nonlinear absorption coefficient by closed and open apertures, respectively. Furthermore, this method provides the values and the sign of nonlinearity. In the Z-scan open aperture (OA) position, the beam is passed through the sample along the focal plane (z-direction), then it is focused directly into the detector by an open aperture or lens. In the closed aperture (CA) position, toward the z-direction, the sample is scanned by a focused laser beam that passes a semi-closed aperture. With the use of a semi-closed aperture, only 30% of the initial beam can be detected, whereas in an open aperture, the whole beam can be recorded. In the closed aperture, the detector can receive more or less of the amount of beam that passes through the aperture (by beam convergence or divergence, respectively). As a result of these changes, the refractive index can be observed as a pre-focal peak and post-focal valley, or vice versa. A negative change in the nonlinear refraction $(\Delta n < 0)$ shows a pre-focal peak and post-focal valley, and a positive change in refraction $(\Delta n > 0)$ presents a pre-focal valley and post-focal peak. Closed and open apertures of Z-scan method may be employed to determine the real and imaginary parts of a third-order nonlinear susceptibility, respectively (Boyd, 2003). In Z-scan measurements, the sample thicknesses are less than the beam diffraction length (z_0), and the closed and open aperture curves are normalized to linear transmittance (transmittance at large values of z).

1.5 Nonlinear Optical Properties of Materials

Various studies have focused on the nonlinear optical properties of materials to demonstrate the abilities of materials to operate in optical switches and communication (Irimpan et al., 2008). The investigations measured materials with high third-order nonlinear properties, which are designed to be applied in optical areas by virtue of their linear and nonlinear optical behavior. In this area, nanoparticles possess enormous importance in photonics and optics (Wang et al., 2010). Metal and metal oxide nanoparticles have attracted attention because of their third-order nonlinear susceptibility, which presents their potential to be employed in optics and photonics (Battaglin et al., 2004; Moslehirad et al., 2011). Metal nano-particles have been recommended as targeted thermal agents for medical applications (therapy and drug delivery) (Oneal, 2004). Furthermore, metallic nano-particles, such as Ni nano-particle, show super electromagnetic properties, which has been proven to exhibit a polarization-dependent optical absorption (Quemard et al., 2001).

Nano-fluid is a suspension of nano-particles in liquid, which displays considerable improvement in its properties at moderate nano-particles. Many studies on nano-fluids have focused on determining their potential for different applications in optics, photonics, electronics, solar collector, nuclear reactor, etc. (Kumar et al., 2008). Recent studies on nano-fluids opened up a great area for finding new nonlinear materials. Large third-order nonlinear optical properties of metal and metal oxide nano-fluids gained much attention because of their unique application in optical switch. For example, Al_2O_3 is one of the most important applied materials due to its good electrical and optical properties (Rao, 2009). Furthermore, aluminum oxide is known as a superior material for laser technology applications (Shahriari et al., 2010) .

Glasses in different shapes and sizes can be created by superior homogenity. Glasses, as macroscopic centrosymmetric matrices, play a significant role in generating third-order nonlinear optical phenomenon (Smektala & Quemard, 2000). These nonlinear optical properties are applied in telecommunication signal processing, such as in ultrafast all-optical switching. Among the different types of glasses, considerable interest has been given on the optical properties of phosphate glassy systems because of their superior transparency. Phosphate glasses possess a flexible structure that can receive several cation and/or anion exchanges, which is scientifically and technologically significant (Zhang et al., 2009). Therefore, studies on the optical and electrical properties of phosphate glass play a key role in the potential use of the glass material in optical devices. Linear optical glasses have been noticeably applied, although vitreous systems such as phosphate glasses have been approved for nonlinear optical switches.

In this study, the nonlinear optical properties of different materials such as nano-fluids and glass structure samples were investigated.

1.6 Research problem

The investigation for new materials for optical devices spreading over a wide study on nonlinearities has become a great and interesting research area. In this case, nano-particles, because of their size, and glass materials, given their unique changeable structure, are considered for nonlinear optics. Furthermore, some parameters such as concentration of materials are significant parameters that can effect on nonlinearities. Hence, studying of concentration effect on nonlinear optical properties plays a key role in determining that the material nonlinearities are as a function of concentration or not?

This study assays to solve the subsequent problems that occur in scientific research on this subject. There are several techniques which characterize the nonlinearities of materials such as eclipsing Z-scan, excite-Probe Z-scan, dual beam Z-scan, single beam Z-scan technique and so on.

Due to low simplicity and accuracy of methods to characterize nonlinear optical properties of materials, there is required to apply a simple and sensitive method in order to determine nonlinearities. Therefore, the popular method (single- beam Z-scan method) was suggested to be applied in the nano-fluid and glass samples for this study. This technique posses several advantages, some that are:

- Simplicity
- Strong sensitivity
- Concurrent determine of both nonlinear value and sign
- Feasibility of seclude the nonlinear refraction and absorption

Theory calculation is prevalently required to modify in the hope to achieve better results. Calculation of nonlinear parameter (such as figure of merit and third-order nonlinear susceptibility) is obtained to show potential of samples to apply in optical application. This study suggests that nano-fluid and glass sample exhibit a good response to nonlinear optics that can be executed in medical, commercial, industrial parts, and so on. Hence, this study plays an important role in opening a new frame to technology.

1.7 Objective of the Research

This dissertation provides a brief view of nonlinear optical properties in nanotechnology and glass technology. This thesis aims to investigate the nonlinear refractive indices and absorption coefficients of nano-fluids and phosphate glasses. The objectives of the present work are:

(i) To investigate the nonlinear optical properties of aluminum oxide–chromium/-nickel nano-fluids.

(ii) To investigate the nonlinearity of ternary zinc magnesium phosphate glasses $[(ZnO)_x\text{-}(MgO)_{30-x}\text{-}(PO)_{70}]$ and $[(ZnO)_{30}\text{-}(MgO)_x\text{-}(PO)_{70-x}]$.

(iii) To study the concentration effect on the nonlinear optical properties of aluminum oxide–chromium/-nickel nano-fluids and phosphate glass.

1.8 Outline and Scope of the Thesis

In chapter 2, the general background of the method used for the measurements, nonlinear refraction, and absorption coefficients are briefly discussed. A theoretical background of the Z-scan method is given in chapter 3. The nano-fluid samples preparation and detailed design of the Z-scan set up utilized in this study are presented in chapter 4. However, preparation of sample glasses (that were obtained from other group) is given in appendices. In chapter 5, the results and discussion of every measurement, including the UV-vis spectra, nonlinear refraction coefficient, nonlinear absorption coefficient, third-order nonlinear susceptibility, and figure of merit (*FOM*) of samples are described. The conclusion of this study and some recommendations for future scientific tasks are given in chapter 6.

REFERENCES

Acharya, A., Behera, R. and Roye, G.S. (2012). Non-linear characteristic of copper oxide (CuO) through Z-scan technique. *Latin-American Journal of Physics Educuation* 6: 402.

Ali, Q. M. and Palanisamy, P. (2006). Z-scan determination of the third-order optical nonlinearity of organic dye nile blue chloride. *Modern Physics Letters B* 20: 623-632.

Alikhani, S., Tajalli, H. and Koushki, E. (2012). Third order optical nonlinearity and diffraction pattern of Ni nanoparticles prepared by laser ablation. *Optics Communications* 286: 318-321.

Balaji, G., Rekha, R. and Ramalingam, A. (2011). Nonlinear Characterization of Safranin O Dye for Application in Optical Limiting. *Acta Physica Polonica A* 119: 359-363.

Battaglin, G., Cattaruzza, E., Gonella, F., Polloni, R., Scremin, B.F., Mattei, G., Mazzoldi, P. and Sada., C. (2004). Structural and optical properties of Cu: silica nanocomposite films prepared by co-sputtering deposition. *Applied Surface Science* 226: 52-56.

Bello, E.W.B. (2013). *Lanthanide-based dielectric nanoparticles for upconversion luminescence*. Doctoral Thesis. Univerity Rovira I Virgili.

Berthoud, T., Delorme, N. and Mauchien, P. (1985). Beam geometry optimization in dual-beam thermal lensing spectrometry. *Analytical Chemistry* 57: 1216-1219.

Bertolotti, M., Liakhout, G., Michelotti, F., Senesi F. and Sibilia, C. (1992). A beam distortion (Z-scan) technique applied to the measurements of non-linearities in Cd,S_x,Se, -x doped glasses. *Pure Applied Optics* 1: 145-156.

Boyd, R.W. (2003). Nonlinear optics. San Diego: Academic press; 2003. 640p.

Choi, S., Zhang, Z., Yu. W., Lockwood, F.E. and Grulke, E.A. (2001). Anomalous thermal conductivity enhancement in nanotube suspensions. *Applied PHysics letters* 79: 2252-2254.

Cook, R.J. and Hill, R.K. (1982). An electromagnetic mirror for neutral atoms. *Optics Communications* 43: 258-260.

Dancus, I., Vlad, V.I, Petris, A., Bazaru, T., Rau, I., Kajzar, F., Meghea. A. and Tane, A. (2013). Z-scan and I-scan methods for characterization of dna optical nonlineaities. *Romanian Reports in Physics* 65: 966-978.

De Boni, L., Andrade, A.A. Misoguti, L., Mendonca, C. and Zilio, S. (2004). Z-scan measurements using femtosecond continuum generation. *Optics Express* 12: 3921-3927.

Ganeev, R.V. (2009). Determination of nonlinear optical characteristics of transparent materials using single laser pulses. *Optics and Spectroscopy* 106: 889-894.

Garmire, E. (2011). Resource Letter NO-1: Nonlinear Optics. *American Association of Physics* 73: 245-255.

Gomes, A., Falcao Filho, E.L., De arjue, C.B., Diego, R., De arjue, R.E., Kochik, S., Mezzapesa, F.P., Carvalho, I.C.S. and Kazansky, P.G. (2007). Third-order nonlinear optical properties of bismuth-borate glasses measured by conventional and thermally managed eclipse Z-scan. *Journal of Applied Physics* 101: 033115-033115-7.

Gomes, S. L., Cuppo, F.L.S. and Figueiredo Neto, A.M. (2003). Nonlinear Optical Properties of Liquid Crystals Probed by Z-scan Technique. *Brazilian Journal Physcis* 33: 813-820.

Gun Y.K. and Kwak, Ch.H. (2009). Simple Optical Methods for Measuring Optical Nonlinearities and Rotational Viscosity in Nematic Liquid Crystals. University Campus STEP Ri Slavka Krautzeka: *Intech,* 111-126p

Harris, D. (1988). *Characterization and relaxation dynamics of the nonlinear optical properties of thiophene-based polymers.* PhD dissertation. City university of New York. July 1998.

Henari, F.Z., Moregenstern, K., Blua, W.J., Karavanskii, V.A. and Dneprovskii, V.S. (1995). Third-order optical nonlinearity and all-optical switching in porous silicon. *Applied PHysics letters* 6: 323-325.

Huang, T., Hao, Z., Gong, H., Liu, Z., Xiao, S., Li, S., Zhai, Y., You, S., Wang, Q. and Qin, J. (2008). Third-order nonlinear optical properties of a new copper coordination compound: A promising candidate for all-optical switching. *Chemical Physics Letters* 451: 213-217.

Irimpan, L., V. Nampoori, V.P.N., Radhakrishnan, B. and Deepthy, A. (2008). Size-dependent enhancement of nonlinear optical properties in nanocolloids of ZnO. *Journal of Applied Physics* 103: 033105-033105-7.

Jacobsohn, L. and Sprinkle, G. (2010). Synthesis, luminescence and scintillation of rare earth doped lanthanum fluoride nanoparticles. *Optical Materials* 33: 136-140.

Jaffar, A. F. (2012). Optical Nonlinearity of Oxazine Dye Doped PMMA Films by Z-Scan Techniques. *Journal of Al-Nahrain University* 15: 106-112.

Jia, T.J., Cheng, H., Ting-Chao, H., Yu-Ting, C. and Cui-Mei, L. (2011). Z-Scan Studies of the Nonlinear Optical Properties of Gold Aqueous Colloid Prepared by the Chemical Reaction Method. *Modern Physics Letters B* 25: 1219-1227.

Kumar, S.S.R., Venugopal Raob, S., Giribabu, L. and Narayana Rao, D., (2008). Nonlinear optical properties of alkyl phthalocyanines in the femtosecond, nanosecond, and cw excitation regimes. Proc of SPIE 6875: 1-12.

Kurum, U., Yaglioglu, H.G., Yuksek, M., Elmali, A., Ates, A., Karabulut, M., Momedov, G.M. and Gasanly, N. (2011). The effect of thickness and doping on the nonlinear absorption behaviour of IIIA-VIA group amorphous semiconductor thin films. Transparent optical networks (ICTON) 2011 13^{th} international conference. Stockholm 1-3 p

Long, M., Swofford, R. and Albert, A. (1976). Thermal lens technique: a new method of absorption spectroscopy. *Science* 191: 183-185.

Lopez-Munoz, G.A., Balderas-Lopez, J. A., Ortega-Lopez, J., Pescador-Rojas, J. and Santoy Salazar, J. (2012). Thermal diffusivity measurement for urchin-like gold nanofluids with different solvents, sizes and concentrations/shapes. *Nanoscale Research Letters*: 7 1-7.

Lyer, A. (1998). Light Measurement HandBook. *International Light, Inc*, United States of America.

Maciela, G., De arju, C., Lipovskiib, A.A. and Tagantsev, D.K. (2002). Picosecond Z-scan measurements on a glass-ceramic containing sodium niobate nanocrystals. *Optics Communications* 203: 441-444.

Majles Ara, M., Dehghani, Z., Sahraei, R. and Nabiyouni, G. (2010). Non-linear optical properties of silver nanoparticles prepared by hydrogen reduction method. *Optics Communications* 283: 1650-1653.

Mathews, S., Chaitanya Kumar, S., Giribabu, L. and Venugopal Rao, S. (2007). Large third-order optical nonlinearity and optical limiting in symmetric and unsymmetrical phthalocyanines studied using Z-scan. *Optics Communications* 280: 206-212.

Miragliotta, J.A. (1995). Analytical and Device-Related Applications of Nonlinear Optics. *Applied Research* 16: 348-357.

Moslehirad, F., Majles Ara, M.H., Torkamany, M.J. and Sabbaghzadeh, J. (2011). Nonlinear Optical Response of Titania Nanoparticles Prepared by Pulsed Laser Ablation. *Physics Procedia* 19: 152-157.

Muthukumar, V., Kiran, J.K., Reppert, J., Satyajit, R., Krishna, V., Nageshwar Rao, G., Siva Rama Krishnan, S., Siva Sankara Sai, S., Venkataramaniah, K. and Rao, A.M. (2008). Nonlinear optical transmission of surface-modified nickel sulfide nanoparticles: saturtaion of absorption and optical limiting. *Nano* 3: 161-167.

Natarajan, V., Sivanesan, T. and Pandi, S. (2010). Third order non-linear optical properties of L-arginine hydrochloride monohydrate single crystals by Z-Scan technique. *Indian Journal of Science and Technology* 3: 897-899.

Nooraldeen, A. Y., Palanichant, M. and Palanisamy, P.K. (2009). Influence of solvents polarity on NLO properties of fluorone dye. *International Journal Nonlinear Science* 7: 290-300.

Oneal, D. H., Halas, L., Payne, N. and West, J. (2004). Photo-thermal tumor ablation in mice using near infrared-absorbing nano-particles. *Cancer Letter* 209: 171-176.

Perez, J.L.J., Gutierrez-Fuentes, R., Ramirez, J.F.S., Vidal, O.U.G., Tellez-Sanchez, D.E., Pacheco, Z.N.C., Cruz Orea, A. and Garcia, J.A.F. (2013). Nonlinear Coefficient Determination of Au/Pd Bimetallic Nanoparticles Using Z-Scan. *Scientific Research* 2: 223-228.

Power, J. F. (1990). Pulsed mode thermal lens effect detection in the near field via thermally induced probe beam spatial phase modulation: a theory. *Applied Optics* 29: 52-63.

Quemard, C., Smektala, F., Couderc, V., Barthelemy, A. and Lucas, L. (2001). Chalcogenide glasses with high non linear optical properties for telecommunications. *Journal of Physics and Chemistry of Solids* 62: 1435-1440.

Quemard, C., Smektala, F., Couderc, V., Barthelemy, A. and Lucas, J. (2001). Chalcogenide glasses with high non linear optical properties for telecommunications. *Journal of Physics and Chemistry of Solids* 62: 1435-1440.

Rad, A.G., Madanipour, K., Koohian, A. and Taheri, N. (2012). Sign Identification of Nonlinear Refractive Index of Colloidal Nanoparticles by Moire Deflectometry Technique. *Journal European Optical Soc*iety 7: 1-5.

Rao, S.V. (2009). Femtosecond and continuous-wave nonlinear optical properties of (H2) 2SnPc, Sn (OH) 2Pc, Sn (Cl) 2Pc studied using Z-scan technique. SPIE LASE: Lasers and Applications in Science and Engineering, International Society for Optics and Photonics.

Rao, S.V., Anusha, P.T., Prashant, T.Sh., Sawin, D. and Tewari, S.P. (2011). Ultrafast Nonlinear Optical and Optical Limiting Properties of Phthalocyanine Thin Films Studied Using Z-Scan. *Materials Sciences and Applications* 2: 299-306.

Ren, Q., Zhang, X., Wen-Feng, G., Xiang-Bing, S., Xin-Qing, W., Hong-Liang, Y., Fue-Jun, Zh., Yi, G. and Dong, X. (2007). Third-order nonlinear optical properties of Co (dmit) 2 complexes using Z-scan technique. *Nonlinear Optics* 6839: 1-8.

Saaty, T.L. (2004). Decision making—the analytic hierarchy and network processes (AHP/ANP). *Journal of Systems Science and Systems Engineering* 13: 1-35.

Sadik, Z.S., Al-Amiedy, D.H. and Jaffar, A.F. (2012). Third Order Optical Nonlinearities of C450 Doped Polymer Thin Film Investigated by the Z-Scan *Advances in Materials Physics and Chemistry* 2: 43-49.

Sanghera, J., Florea, C., Shaw, L., Pureza, P., Nguyen, V., Bashkanski, M., Dutton, Z. and Aggarwal, I. (2008). Non-linear properties of chalcogenide glasses and fibers. *Journal of Non-Crystalline Solids* 354: 462-467.

Shahriari, E., Yunus, W.M.M. and Saion, E. (2010). Effect of particle size on nonlinear refractive index of Au nanoparticle in PVA solution. *Brazilian Journal of Physics* 40: 256-260.

Shahriari, E. and Yunus, W.M.M. (2010). Single Beam Z-Scan Measurements of Nonlinear Refraction and Nonlinear Absorption Coefficients in Silver Nano-Fluid. *American Journal of Engineering and Applied Sciences* 3: 98.

Shahriari, E., Yunus, W.M.M. (2010). Effect of Particles size on nonlinear fraction and absorption and absorption of Ag nanoparticles. *Digest Journal of Nanomaterials and Biostructures* 5: 939-946.

Shahriari, E. Yunus, W.M.M., Naghavi, K. and Talib, Z.A. (2010). Effect of concentration and particle size on nonlinearity of Au nano-fluid prepared by gamma (60Co) radiation. *Optics Communications* 283: 1929-1932.

Sheik-Bahae, M., Said, A.A., Hagan. D.J., Soileau, M.J. and Van Stryland, E.W. (1991). Nonlinear refraction and optical limiting in thick media. *Optical Engineering* 30: 1228-1235.

Sheik-Bahae, M., Said, A.A., Wei, T., Hagan, D.J. and Van Stryland E.W. (1990). Sensitive measurement of optical nonlinearities using a single beam. Journal of *Quantum Electronics* 26: 760-769.

Sheikh-Bahae, Said, A.A., Wei, T., Hagan, D.J. and Van Stryland E.W. (1989). High-sensitive, single-beam n_2 measurements. *Optics letters* 14: 955-957.

Sheldon, S., Knight, L. and Throne, J. (1982). Laser-induced thermal lens effect: a new theoretical model. *applied optics* 21: 1663-1669.

Shen, J. and Snook, R.D. (1989). Thermal lens measurement of absolute quantum yields using quenched fluorescent samples as references. *Chemical Physics Letters* 155: 583-586.

Smektala, F. and Quemard, C. (2000). Non-linear optical properties of chalcogenide glasses measured by Z-scan. *Journal of Non-Crystalline Solids* 274: 232-237.

Stryland, E. and Hagan, D. (2003). Nonlinear Absorption. *Encyclopedia of optical engineering* 2: 1475-1481.

Swofford, R. Long, L.M., Albrech, A.C. (1976). C–H vibrational states of benzene, naphthalene, and anthracene in the visible region by thermal lensing spectroscopy and the local mode model. *Journal of Chemical Physics* 65: 179.

Venkatram, N., Rao, D.N. and Akundi, M.A. (2005). Nonlinear absorption, scattering and optical limiting studies of CdS nanoparticles. *Optics Express* 13: 867-872.

Wang, J., Gu, B., Wang, H.T., Ni, X.W. (2010). Z-scan analytical theory for material with saturable absorption and two-photon absorption. *Optics Communications* 283: 3525-3528.

Wang, Y.H., Wang, Y.M., Han, J.C., Lu, J.D. and Ji, L.L. (2010). Experimental analysis of optical limiting properties of Cu nanoclusters. *Physica B: Condensed Matter* 405: 2664-2667.

Xu, M. and Tran, C.D. (1990). Thermal lens-circular dichroism detector for high-performance liquid chromatography. *Analytical Chemistry* 62: 2467-2471.

Xu, T., Chen, F., Dai, Sh., Shen, X., Wang, X., Nie, Q., Liu, Ch., Xu, K. and Heo, J. (2011). Glass formation and third-order optical nonlinear properties within TeO–Bi_2O_3–BaO pseudo-ternary system. *Journal of Non-Crystalline Solids* 357: 2219-2222.

Yang, G. Yan, L., Lu, H., Yang, G. and Chen, Zh. (2002). Z-scan determination of the large third-order optical nonlinearity of Rh:$BaTio_2$ thin films deposited on MgO substrates. *Optics Communications* 209: 445-449.

Yin, M., Li, H.P., Tang, S.H. and J, W. (2000). Determination of nonlinear absorption and refraction by single Z-scan method. *Applied Physics B* 70: 587-591.

Yu-Hua, W., Hui-Qing, L., Jian-Dau, L. and Rau-Wu. (2011). Optical Limiting Properties of Ag-Cu Metal Alloy Nanoparticles Analysis by using MATLAB. *Chinese Physics Letters* 28: 116101.

Zhang, H., Nie, Q., Dai, Shi., Shen, X., Wang, X. and Zhang, X. (2009). CsCl Effected Ultrafast Third-order Optical Nonlinearities of GeS2-Sb2S3 Chalcogenide Glasses. *Journal of materials science & technology* 25: 772.

Zhong-Yu, L., Song, X., Zi-Hui, Ch., Fu-Shi, Zh. and Kazuo, K. (2011). Third-Order Optical Nonlinearities of Squarylium Dyes with Benzothiazole Donor Groups Measured Using the Picosecond Z-Scan Technique. *Chinese Physics Letters* 28: 084208.