Nonlinear Optical Properties of $LiNbo_3$ Thin Film Using Z-Scan Technique

Dr.Khawla jemeal tahir
Karbala university,college of science,physics department

Abstract

$LiNbO_3$ thin films were prepared on quartz substrates by sol-gel process using Lithium carbonate (Li_2CO_3) and niobium oxide. Several testing were done to study the characteristics of the sample,including X-ray diffraction,as well as transmission and digital holographic microscope(DHM).We have employed Z-Scan technique to study the nonlinear optical properties, represented by the nonlinear refractive index and nonlinear absorption coefficients of a thin film. Z-Scan experiment was performed using a CW Nd:YAG and Nd:YVO_4 lasers in two parts ; the first part has been done using a close aperture at two wavelengths(532 nm and 1064 nm) and the second part was carried out using an open aperture at two wavelengths(532 nm and 1064 nm).

الخلاصة

ان غشاء اليثيوم نيوبايت الذي تم تحضيرة على الكوارتز بطريقة sol-gel باستخدام كاربونات الليثيوم واوكسيد النيوبايت .ولغرض التعرف على خصائص العينة تم أجراء عدة فحوصات مختبرية مثل الفحص بالاشعة السينية بالاضافة الى النفاذية والمجهر الرقمي المجسم (DHM)، وتم استخدام تقنية المسح على المحور الثالث لدراسة الخصائص البصرية اللاخطية متمثلة معامل الانكسار اللاخطي ومعامل الامتصاص اللاخطي للفلم . اجرت تجربة المسح على المحور الثالث باستخدام ليزر النيديميوم-ياك المستمر ونيديميوم ياك مضاعف التردد وعلى جزئين. الجزء الاول تم بوضع ثقب ضيق وبطولين موجيين عند (1064) نانومتر و (532) نانومتر. والجزء الثاني بتكبير فتحة الثقب وبطولين موجيين (1064) نانومتر و (532) نانومتر

Introduction

Lithium niobate (LN) - stoichiometric formula LiNbO3 - is a compound of niobium, lithium and oxygen. It is a dielectric material, insoluble in water, and does not exist in nature (is a human-made compound). Its melting point is 1257°C and its density is 4.65 g/cm³.and it is now one of the most widely used nonlinear materials. In fact, it is characterized by large pyroelectric, piezoelectric, nonlinear and electro-optic (EO) coefficients and has also useful acoustic and acousto-optic properties[1]. This richness of large magnitude physical effects has caused LN to become widely used in applications such as acoustic wave devices, optical amplitude modulators, Q switching devices for lasers, second-harmonic generators, beam deflectors,memory elements, holographic data processing devices and others [2] LN has a trigonal crystal system belonging to the 3m (C3v) crystallographic point group (Fig.1), and lacks inversion symmetry. It consists of planar sheets of oxygen atoms in a distorted hexagonal close-packed configuration [3,4].

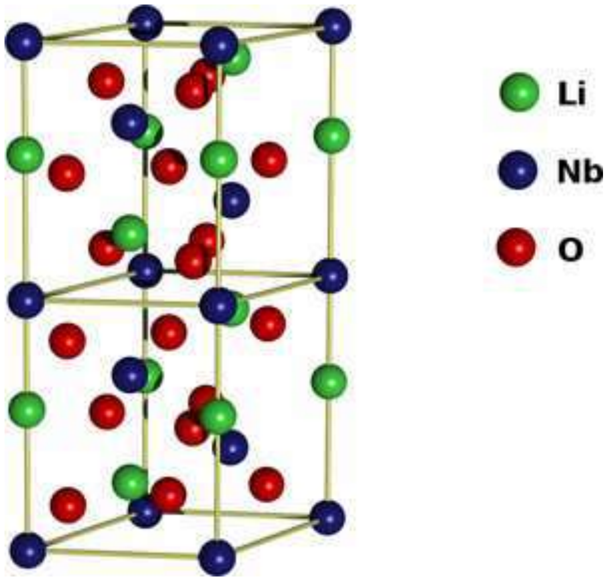


Figure (1**)** Geometrical structure of lithium niobate.[5]

Above the Curie temperature *Tc* (around 1210 °C) the phase is par electric (no spontaneous polarization) while in the ferroelectric phase below *Tc* LN exhibits spontaneous polarization. In the par electric phase Li atoms lie in an oxygen layer that is *c*/4 away from the Nb atom, while Nb atoms are centred between oxygen layers[6]

Lithium niobate is the promising material to create the memory units, electrooptical devices, and waveguides. To develop the element of integral optics,the thin LiNbo3 films with predetermined properties are required. Two of the effective methods of thin film growing with no changes in their elemental composition are RF magnetron sputtering and ion beam sputtering[7] .Those methods can be used to create a wide range of integrated elements such as optical and acoustic wave conductors and light emitting diodes . despite the thin films of $LiNbo_3$ being important elements because of relatively wide scope of their possible practical application ,some crucial properties of the film and interface film substrate playing an important role in the devices work still have not been studied thoroughly, [8]

LiNbo3 thin films have been prepared by several techniques such as ,pulsed laser deposition, sputtering and chemical vapor deposition; however most of these methods are expensive and require complex equipment .sol-gel processing ,a solution –based method is interesting due to easier production of low cost, large area thin films. However ,commonly used alkoxide precursors to produce linbo3 films by sol-gel method are highly reactive and require careful control of hydrolysis condensation reaction[9,10]

Sol–gel and co-precipitation methods have become popular for producing ceramic materials with improved compositional homogeneity at lower sintering temperature [11]. The sol–gel method utilizes expensive precursors, such as metal alkoxides, and depends on a critical drying process which requires control of atmosphere, while the co-precipitation[12]method is limited by cation solutions with similar solubility constants. On the other hand[13]

Experimental

1. Preparation of Thin film

LiNbO3 precursor powder was prepared by the sol-gel method using lithium carbonate (Li_2CO_3) and niobium oxide (Nb_2O_5) The precursor solutions of lithium and niobium were prepared by adding the raw materials into ethylene glycol and

citric acid with heating and stirring at 90 ^{0}C . After complete dissolution, Li_2CO_3 and ethylene glycol were added. To obtain homogeneous and crack-free films of $LiNbO_3$, the precursor was deposited by spin coating on the glass substrates at a spinning speed of 3000 rpm for 30 **s** .The heating rate was 5 ^{0}C/min up to 300 ^{0}C .Subsequent thermal processing is summed up In addition to sample prepared from precursors decomposed in certain atmosphere,blind sample was prepared under standard conditions (heat treatment in ambient atmosphere at 150 °C, 250 °C and 400 °C, 2 hours each step). Phase analysis of the film was performed at room temperature by X-ray diffraction using a Bragg diffractometer. Micro structural characterization of the films was carried out using digital holographic microscopy (DHM)

2. **Z-scan measurement**

The Z-scan is a simple and popular experimental technique to measure the intensity and a direct method to characterize both the nonlinear refraction index and nonlinear absorption .The schematic diagram of the z-scan technique is shown in figures (2) and (3)

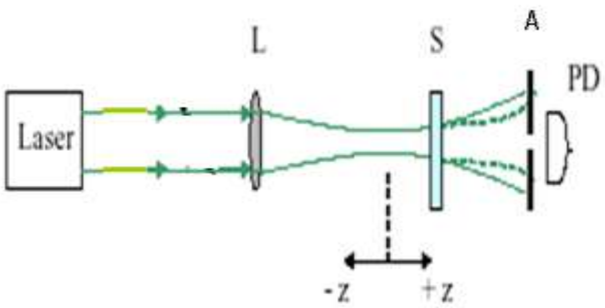
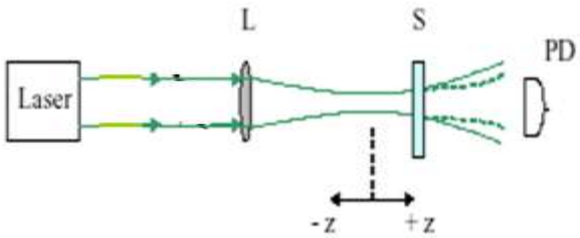


Figure (2) Closed-aperture Z-Scan[14]



Figure(3) 0pen-aperture Z-Scan[14]

It is based on a single beam method and refers to the process of inserting a sample in a focused Gaussian beam and translating it along beam axis through a focal region. The far field intensity is measured as a function of the sample position by properly monitoring the transmittance change through a small aperture at the far field position (closed aperture) .by moving the sample through the focus and without placing an aperture at the detector(open aperture)

Results and Discussion

1- X-ray diffraction analysis

The structural quality of the film $LiNbO_3$ was investigated by using X-ray diffraction. Figure 1 shows the omega – two theta X-ray diffraction spectrum for a film lithium niobate . The dominant peak is located at 2θ of (34.56 degrees). The XRD results of thin film LiNbo3 prepared on glass substrate at room temperature as shown in Figure (4)

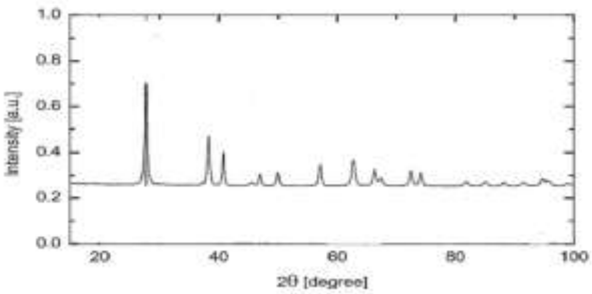


Figure (4)X-ray diffraction pattern of the thin film $LiNbO_3$

The pattern indicates four very weak peaks at approximately 2θ of (46^o), (48.8^o),(63^0), and (75^0) While, the maximum peak has occurred at2θ of (30^o),

3. Digital holographic microscope (DHM)

The digital holographic microscope records a hologram,resulting from the in-line interference between a reference wave and an object wave transmitted through a thin film, and magnified by an Microscope Object .Even if it is possible to record a digital hologram of the sharply focused object in the plane of the CCD sensor. From a single hologram, the numerical wavefront in the hologram plane is reconstructed of holograms were performed by MatLab[15]

by using in-line digital holographic microscopic DHM it is shown that film topography can be measured with an accuracy, as show in figure(5)

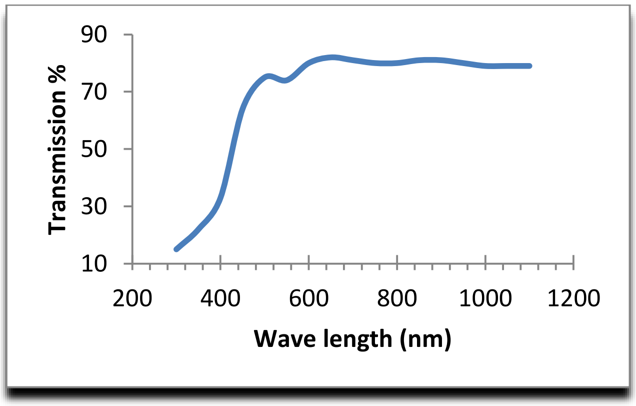
(a) (b) (c)

Figure (5) Thin film $LiNbO_3$ (a) Recording DHM (b) reconstruction DHM (c) topography of the film

Observation from Figure 5 – (C) shown topography of the film ,It is uniformity across the entire micro lens array,

4. Linear Optical properties

For study the linear optical properties. The transmission spectrum of thin film $LiNbo_3$ was analyzed using UV-VIS spectrophotometer in the range as shown in figure(6)



figure(6) the transmission spectrum of the $LiNbo_3$ thin film

The linear absorption coefficient α_0 was determined for two wavelengths 1.06 μm and 532 nm by using formula

$$\alpha_0 = \frac{1}{t}\ln\left(\frac{1}{T}\right) \ldots\ldots\ldots(1)$$

where (t) is the thickness of sample and T is the transmittance.

The refractive index -n_o-can be found from transmittance spectrum of the film according to the following equation

$$n_o = \frac{1}{T} + \left[\left(\frac{1}{T^2} - 1\right)\right]^{1/2} \quad \ldots\ldots\ldots\ldots(2)$$

The linear absorption coefficient and refractive index are shown in table 1

Table (1) linear absorption coefficient and refractive index versus wavelength

λ nm	Thickness (μm)	$\alpha_o(\mu m)^{-1}$	n_0
532	2	0.1505	2.259
1064	2	0.1178	2.041

5. Nonlinear Optical Properties

The nonlinear refractive index and nonlinear absorption coefficient of the $LiNbO_3$ film were measured by the Z-scan technique. The measurements were done at 1064 nm and 532 nm

Nonlinear Refractive Index

The nonlinear refractive index, there are two cases were chosen at 1064 nm and at 532 nm.

Figure (7) shows the closed-aperture Z-scan results at 1064 nm

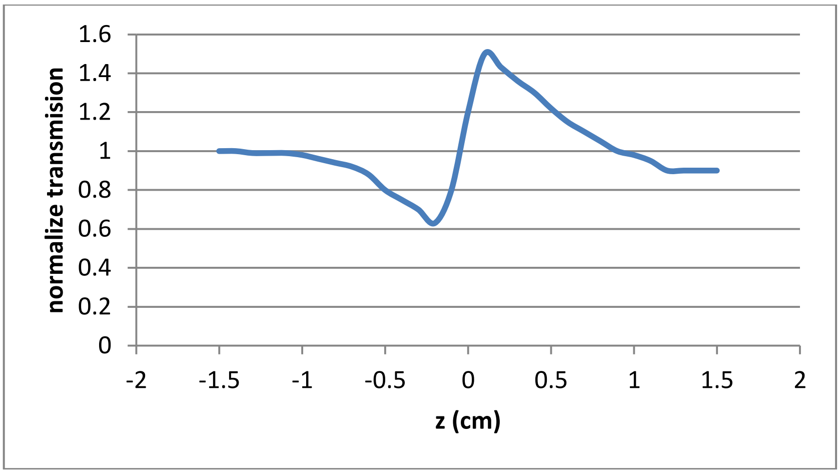


Figure (7) Closed-aperture Z-Scan at 1064 nm

At case II, Figure (8) shows the closed-aperture Z-scan at 532 nm

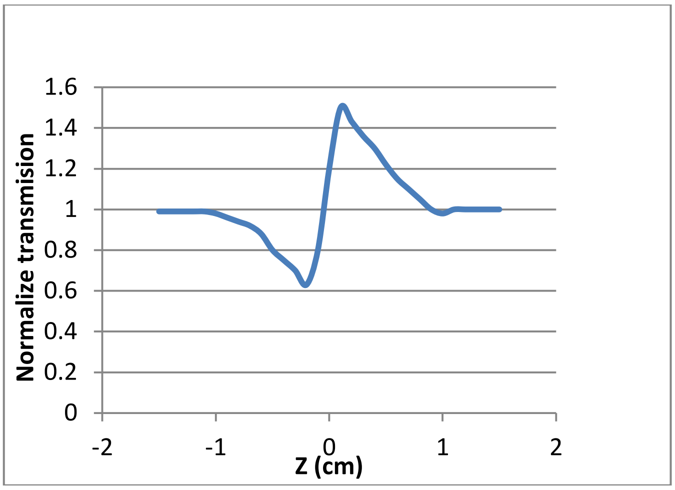


Figure (8) Closed-aperture Z-Scan at 532 nm

As shown in figure(7,8), the transmittance started with a linear behavior at different distances from the far field of the sample position (-Z) with respect to the focal plane at Z=0 mm. At the near field the transmittance begins to decrease until it reaches the minimum value (T_{Valley}) at approximately Z=-0.1cm. Afterward, the transmittance begins to increase until it reaches the maximum value (T_{Peak}) at approximately Z=0.1cm. Again, the transmittance begins to decrease toward the linear behavior at the far field of the sample position (+Z). For the calculation of the nonlinear refractive index(n_2) the following standard relations were utilized

$$\Delta T_{p-v} = 0.406(1-s)^{0.25}\left|\Delta\Phi_0\right| \text{............(3)}$$

$$n_2 = \frac{\Delta\Phi\lambda}{2\pi I_o L_{eff}} \text{.................(4)}$$

Where, ΔT_{p-v} is the peak-valley transmittance difference from the closed –aperture,$\left|\Delta\phi_0\right|$ is the on axis nonlinear phase –shift and (s) is the linear aperture transmittance given by $s = 1-\exp(-2r_a^2/w_a^2)$ where r_a is the aperture radius and w_a is the beam radius at the aperture in linear , I_o is the intensity at the focal spot given by [44]:- $I_o = 2P_{peak}/\pi\omega_o^2$, the effective length of the sample, can be determined from the following formula [15]:- $L_{eff} = (1-e^{-\alpha oL})/\alpha_o$.

At the close – aperture case, the z-scan curves were determined at 1064 nm at different powers of the laser source as shown in figure (9).

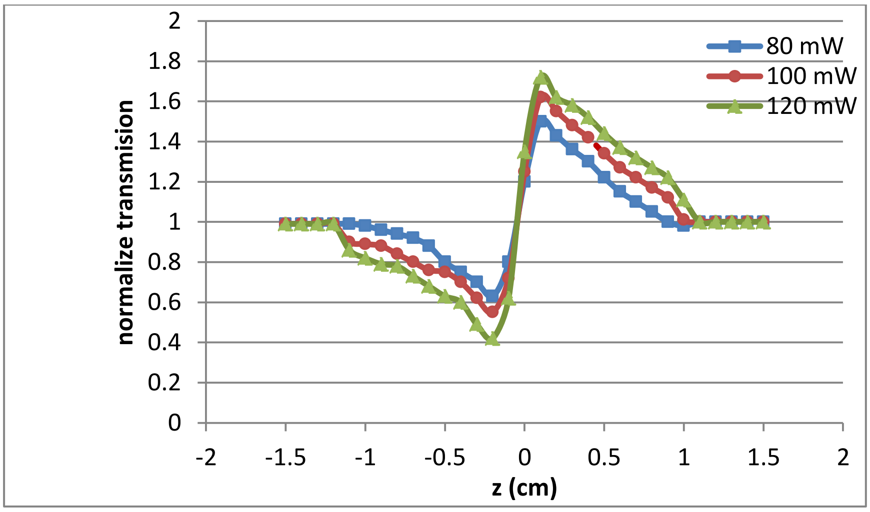


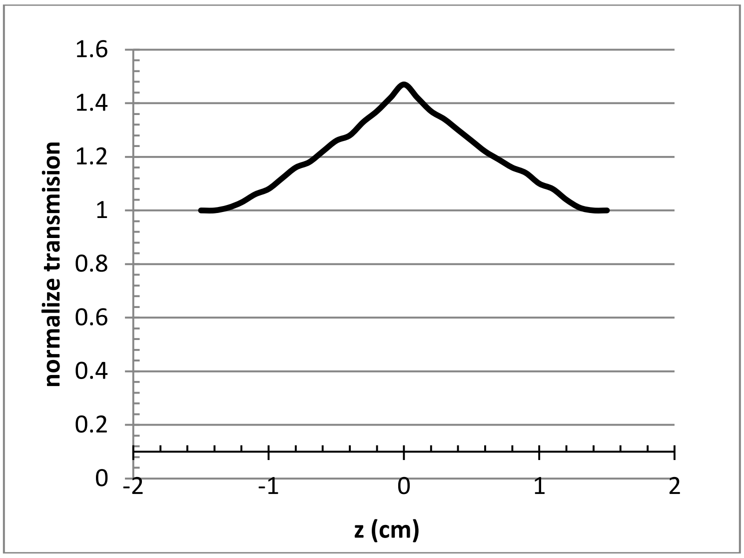
Figure (9) Closed-aperture Z-Scan at 1064 nm which used to determine the nonlinear phase shift ΔΦ and the nonlinear refractive index. This can be shown in table (2).

Table (2) Nonlinear refractive index and nonlinear phase shift

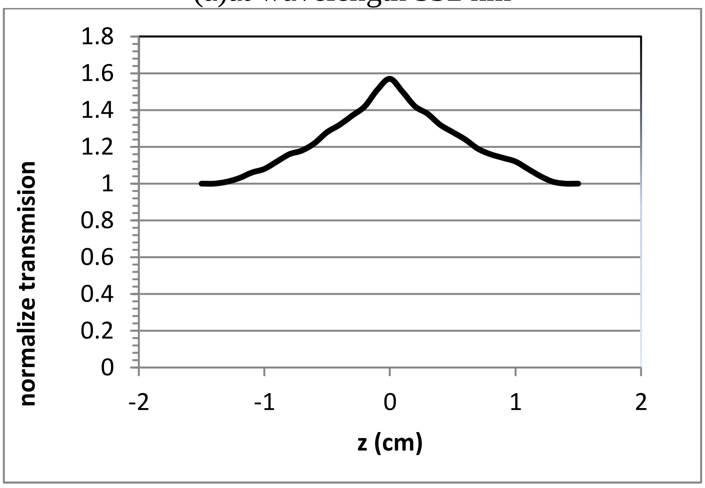
λnm	P mW	n_0	I_0 mW/cm^2	ΔΦrad	n_2 cm^2/mW
532	50	2.259	98.25×10^3	1.97	1.05×10^{-6}
1064	80		157.2×10^3	2.016	1.22×10^{-6}
	100		196.5×10^3	2.561	1.24×10^{-6}
	120	2.041	235.8×10^3	3.103	1.252×10^{-6}

Nonlinear absorption coefficient

The nonlinear absorption coefficient β, of the thin film $LiNbO_3$ was determined by performing the open aperture z-scan for wavelengths 532 nm and 1064 nm .Figure (10) represents the normalized transmission as function of position



(a)at wavelength 532 nm



(b)wavelength 1064 nm

Figure (10) open aperture z-scan

In figure(10) the curve starts linear for different distances from the far field of the transmittance when the sample is at position (-Z). At the near field, the transmittance curve begins to decrease until it reaches a minimum value at the focal point, where Z=0 cm. Afterwards, the transmittance begins to increase towards the linear behavior at the far field of the sample position (+Z).The non-linear absorption coefficient ,β, as calculated from the relation [16]

$$\beta = \frac{2\sqrt{2}}{IL_{eef}} \Delta T \text{ ....(5)}$$

Where ΔT is the one peak value at the open aperture Z-scan curve,the magnitude of β depends on the wavelength and on the input intensity, but this dependence is not very strong at low intensities. The nonlinear absorption coefficient is found to be β =0.2437 cm/mW at wavelength 532 nm and β=0.1580 .When wavelength 1064 nm, When you increase the intensity of the laser(I=196.5×10^3mW/cm^2) we note a decrease in the value β=0.126 cm/mW.The nonlinear absorption coefficient is inversely proportional to the input intensity. the nonlinear behavior of the transmission curves in good agreement with the result reported by Hebling[16]

Conclusion

Nonlinear optical properties of thin film LiNbO3 was investigated by a single-beam Z-scan technique using a cw laser beam at 532 nm and 1064nm wavelengths. Thin films of LiNbO3 were fabricated by the sol-gel method using Lithium carbonate (Li_2CO_3) and niobium oxide (Nb_2O_5) as precursor materials. The nonlinear refractive index of the thin films n_2 versus input powers and wavelengths ,which are equal n2 avarag=1.23×10^{-6} cm^2/mW at 1064 nm wavelength and n_2= 1.05×10^{-6} cm^2/mW for 532 nm wavelength The nonlinear absorption coefficient is found to be β =0.2437 cm/mW at wavelength 532 nm as well β=0.1580 When you wavelength 1064 nm, When you increase the intensity of the laser decrease in the value β.

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