Structural and optical properties of $BaTiO_3$ thin films prepared by pulsed laser deposition

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Abstract

$BaTiO_3$ thin films have been deposited on Si (111) and glass substrates by using pulsed laser deposition technique. The films were characterized by using X-ray diffraction, atomic force microscope and optical transmission spectra. The films growth on Si after annealing at 873K showed a polycrystalline nature, and exhibited tetragonal structure, while on glass substrate no growth was noticed at that temperature. UV-VIS transmittance measurements showed that the films are highly transparent in the visible wavelength region and near-infrared region for sample annealing on glass substrate. The optical gap of the film were calculated from the curve of absorption coefficient $(\alpha h\nu)^2$ vs. hν and was found tobe 3.6 eV at substrate temperature 573K, and this value increases up to 3.69 eV at annealing temperature of (673K), but the refractive index was found to decrease from 2.4 to 2.2 at that temperature.

Key words

$BaTiO_3$ thin films, Pulsed laser deposition

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الخواص التركيبية والبصرية لأغشية الباريوم تيتانيت الرقيقة المرسبة بتقنية الليزر النبضي

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الخلاصة

تم ترسيب مركب الباريوم تيتانيت على قواعد من السيليكون (111) والزجاج بأستخدام تقنية ترسيب الليزر النبضي .تم فحص الافلام بواسطة حيود الاشعة السينية ومجهر القوى الذري وطيف النفاذية البصري.وقد ظهر ان نمو الغشاء على السيلكون بعد التلدين عند درجة 873كلفن بشكل متعدد البلورات، بتركيب رباعي، بينما الاغشية المرسبة على الزجاج لم تظهر اي نمو في الغشاء عند التلدين بتلك الدرجة. كذلك تم دراسة الخصائص البصرية بواسطة قياسات مطياف النفاذية للأشعة المرئية وفوق البنفسجية وقد ظهر ان الاغشية شفافة جداً في منطقةِ اطوالِ الموجة المرئيةِ ومنطقةِ الأشعة تحت الحمراءِ القريبة بالنسبة للعيّنةِ الملدنةِ على قاعدة زجاجية. كذلك تم حساب قيمة فجوة الطاقة من خلال معرفة معامل الامتصاص وتغيره مع الطاقة حيث وجدت قيمتها 3.6الكترون فولت للغشاءالمحضر عند درجة حرارة القاعدة 573كلفن وهذه القيمة تزداد الى 3.69الكترون فولت، اما معامل الانكسار يقل عندما يلدن الغشاء لينخفض من 2.4الى2.2 عند تلدين 673 كلفن.

Introduction

Barium titanate ($BaTiO_3$) is one of the most important ferroelectric materials and has attracted much attention for its remarkable properties such as high dielectric constant, good ferroelectric properties, and large electro-optic, nonlinear optic coefficients and as piezoelectric properties resulting in broad applications in the control systems [1,6] $BaTiO_3$ thin films have been deposited by several methods like metal–organic chemical vapour deposition (MOCVD)[7], sol–gel method [8, 9], chemical solution deposition[10], radio-frequency (RF) sputtering [11] and pulsed laser deposition[12,13]. Among the various methods, pulsed laser deposition is an excellent method to produce thin films on various substrates with good stoichiometry that can easily be up scaled for industrial applications [14].

The aim of this work is to investigate $BaTiO_3$ thin films prepared by pulsed laser deposition under different annealing temperatures and study their structural and optical properties.

Experiments

The composition $BaTiO_3$ were prepared by solid state reaction of mixed oxides, BaO, TiO_2 compounds in [1:1] mole ratio as starting materials. The powders were mixed in alumina crucible. Calcined powder was pressed to form a pellet of 1.5cm diameter with a pressure of 5 ton, using uniaxial hydraulic press. The samples were then sintered at 1223K for 5hr which ready for X-ray diffraction test .Thin films of $BaTiO_3$ were deposited on the p-type Si (111) and glass substrate by pulsed laser deposition method using a Nd:YAG laser (1064/532 nm) with a pulse energy and duration of 800 mJ and 20 ns, respectively. The substrate temperature during the deposition was maintained at 573 K, and the substrate target distance at 3.5 cm. The main vacuum chamber is a 10cm diameter evacuated (10^{-3}mbar).

The thin films were characterized by x-ray diffraction (XRD; SHIMADZU 6000) radiation and atomic force microscope (AFM) (AA3000 Scanning Probe Microscope SPM, tip NSC35/AIBS).The UV–VIS absorption spectra of the films were measured using a spectrophotometer (A double–beam UV–VIS-NIR 210A Spectrophotometer).

Results and Discussion

1. X-ray diffraction

Fig (1) shows the XRD pattern of $BaTiO_3$ thin film on Si (111) and glass substrate with different annealing temperatures. No evidence of any phases present on glass substrate or Si substrate as deposited that mean the formation of $BaTiO_3$ phase amorphous. After annealing both samples at 873K for (5) hr, the x-ray diffraction detected a $BaTiO_3$ on Si (111) substrate but not for glass substrate. All diffraction peaks can be assigned to $BaTiO_3$ phase with out any indication of other crystalline phase. The films show a polycrystalline, and exhibited tetragonal structure according to the International Centre for diffraction data (JCPDS) [15]. The diffraction peaks are broad which indicates the formation of nano crystals [16]. The average crystalline size was found to be (68nm) from application of scherrer equation by taking account of the peak broadening at (111) diffraction plane.

Fig (2) shows atomic force microscopy (AFM) scans of film deposite on Si (111) at annealing temperature (873K) confirm grain size on the order of (58nm) and RMS roughness of (0.571nm). The surface morphology of the $BaTiO_3$ thin films as observed from the AFM micrographs proves that the grains are uniformly distributed within the scanning area (429nm x 429nm).

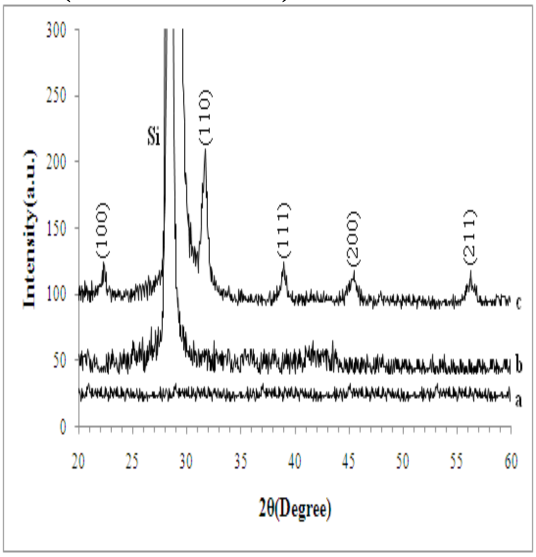

Fig (1) The X-Ray diffraction pattern of $BaTiO_3$ thin films at:(a) $BaTiO_3$/glass thin films deposited at substrate temperature (T_s=573K),(b) $BaTiO_3$/Si thin films deposited at substrate temperature (T_s= 573K), (c) $BaTiO_3$/Si at annealing temperature equal (873 K).

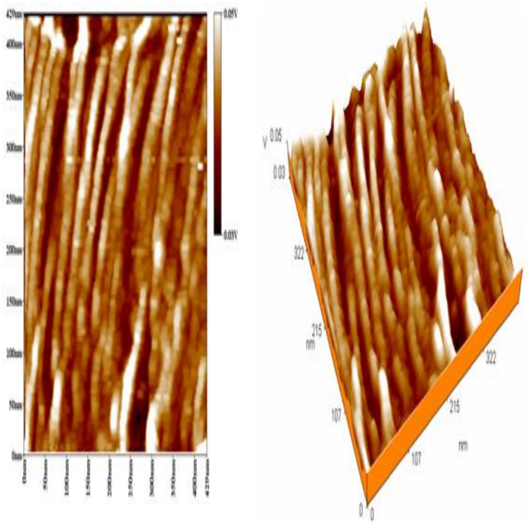

Figure (2) The AFM images of $BaTiO_3$/Si at annealing temperature equal (873 K).

2. Optical properties

Fig.(3) shows the optical transmission as a function of wavelength in the range (200 – 1100 nm) with thickness (300nm) at temperature (T_S=573K) and annealing temperature 673K.The spectrum shows strong attenuation in the short wave length region. The maximum transmittance observed for $BaTiO_3$ was almost (42%) in the near-infrared region, while after the annealing at 673K films the maximum transmittance was found to be (58%). It could be notice that transmittance increases with increasing of annealing temperature which means a decrease in the reflection and absorption. The band gap was determined using the relation for band to band transition, depending on Eq. (1) [17]:

$$\alpha h\nu = B(h\nu - E_g)^m \quad (1)$$

Where hν is the photon energy, E_g is the band gap, m is a power factor and B is a constant. A plot of $(\alpha h\nu)^2$ against the photon energy hν is presented in Figure (4) The energy band gap is obtained from intercept by extrapolated linear part of the curve with the energy axis, the direct band gap of the pure $BaTiO_3$ films increases from 3.6 -3.69 ev From the figure we can see that the annealing shifts the optical band gap from approximately 3.69 eV at 673K.

Fig (5) shows the variation in refractive index of amorphous $BaTiO_3$ film .It is observed that the refractive index, in general decreases slightly with increasing of annealing temperature. The values of the refractive index varied from 2.4-2.2 depend on annealing temperature.

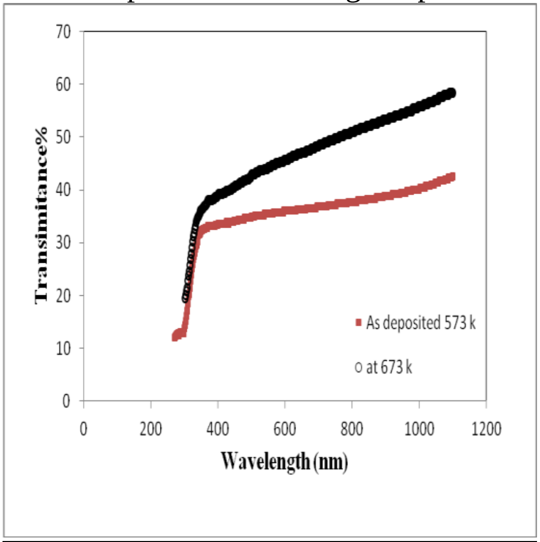

Figure (3) UV-VIS transmittance spectra of the amorphous $BaTiO_3$ films at glass substrate as deposited (573K) with annealing temperature (673K).

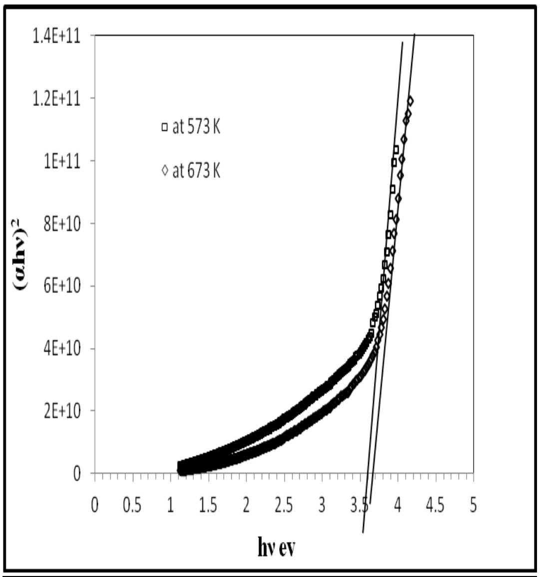

Figure (4) $(\alpha h\nu)^2$ versus photon energy (hν) of amorphous $BaTiO_3$ thin films on glass Substrate as deposited (573K) with annealing temperature (673 K).

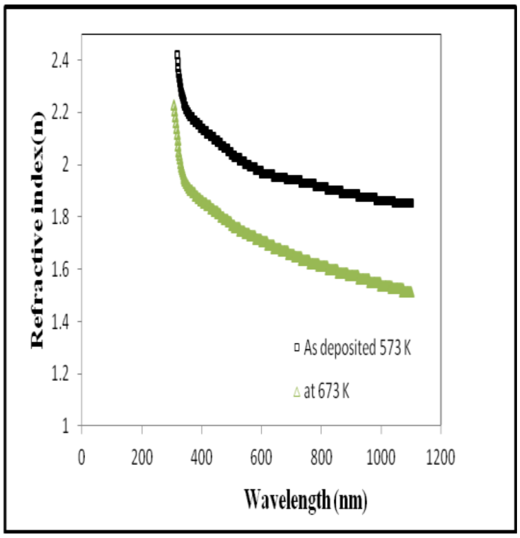

Fig (5) The variation of the refractive index (n) with photon energy for as deposited (Ts=573K) and annealed amorphous $BaTiO_3$ /glass film at (673K) in air.

Conclusions

In this work, $BaTiO_3$ thin film has been synthesized using pulsed laser deposition technique on Si (111) and glass substrate. The X-ray diffraction of both films are amorphous for as deposited. The crystalline phase was detected on Si (111) substrate after annealed at 873K. The grain size and the surface roughness as observed by AFM increase with increasing annealing temperature. The spectrum transmittance observed for amorphous $BaTiO_3$ attenuation in the short wave length region. The transmittance increases with increasing of annealing temperature. The energy band gap is direct increases with increasing annealing temperatures, while the refractive index decreases slightly.

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