Transparent Oxide MgO Thin Films Prepanred By Reactive Pused Laser Deposition

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Abstract

Transparent dielectric thin films of MgO have been deposited on glass substrates at different oxygen pressure between (50-300) mbar using a pulsed laser deposition technique to ablation of Mg target in the presence of oxygen as reactive atmosphere. Structural, and optical, properties of these films have been investigated. The films crystallize in a cubic structure and X-ray diffraction measurements have shown that the polycrystalline MgO films prepared at oxygen pressure (200) mbar and substrate temperature (150°C) with (111) and (002) orientations. The films deposited at oxygen pressure between (150-300) mbar and substrate temperature (150°C) exhibited highest optical transmittivity (>80%) and the direct band gap energy was found to be 5.01 eV at oxygen pressure (200) mbar. The measured of the resistivity of the film prepared at oxygen pressure (200) mbar and substrate temperature (150°C) was $1.45x10^{7}\Omega$ cm.

Keywords: laser deposition, Thin films, Magnesium Oxide.

تحضير ودراسة خصائص غشاء اوكسيد المغنيسيوم باستخدام تقنية الترسيب بالليزر النبضي

الخلاصة

أغشيه اوكسيد المغنيسيوم الشفافة العازلة المرسبة على قواعد من الزجاج عند ضغوط أوكسجين مختلفة تتراوح بين (50-300) ملى بار باستخدام تقنية الترسيب بالليزر النبضي الذي يعمل على تشظية أهداف معدن المغنيسيوم المتمثلة باستخدام الأوكسجين كوسط ترسيب. تضمن البحث دراسة وتحليل للخصائص البصرية ،, والتركيبية للأغشية المحضرة بظروف مختلفة. أظهرت نتائج الخصائص التركيبية للأغشية المنماة من خلال دراسة حيود الأشعة السينية تبين بان الحبيبات لغشاء اوكسيد المغنيسيوم قد تمحورت باتجاه (111) و(002) عند افضل ضروف للترسيب التي وجدت لتكون (200mbar) ضغط اوكسجين و($150C^{o}$) درجة حرارة قاعدة . أظهرت نتائج الخصائص البصرية لأغشية اوكسيد المغنيسيوم إن فجوة الطاقة عند أفضل الشروط هي(5.01eV). كما إن نسبة النفاذية لأغشية اوكسيد المغنيسيوم كانت عالية وتصل إلى (80%) عند ضغط أوكسجين تتراوح بين (150-300mbar) ملي بار. أما نتائج قياس المقاوميه لهذه الأغشية تدل على أنها ذات مقاوميه عالية تصل الى($1.45*10^{7}\Omega cm$) عند نفس ضروف الترسيب السابق.

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Introduction

MgO is a highly ionic crystalline solid, which crystallizes into a rock salt structure. It has fcc Mg+ and O- sub lattices, and low energy neutral (100) cleavage planes. The lattice constant of MgO is 4.212A° and its refractive index and dielectric constant are 1.72 and 9.83 respectively. Magnesium oxide seems to be a good candidate regarding its bulk properties: large band gap (7.8 eV), high thermal conductivity and stability it foung to be an alternative dielectric to Silicon dioxide (SiO_2) to reduce the electric field in capacitive networks [1]. MgO, is widely used as a substrate for high-temperature superconductor films deposition, has attracted much attention due to its low dielectric constant, low dielectric loss, and low mismatch with YBCO films [2]. With these properties, MgO shows a wide application in microwave devices. And due to its low refractive index, MgO is especially a suitable buffer for expitaxial optical waveguide films [3]. Ceramic or metal/alloy oxide films are of scientific and technological importance due to their applications in optical and electronic devices, in catalytic reactions, as protective coatings on metals, as single tunnel barriers and in gas sensors. They are mostly preferred due to their wide band gap and inertness against many chemical reactions [4].Interestingly, ceramic oxide thin film substrates are used as support for different metallic films or multi-layers. These applications need the growth of expitaxial insulating films with single crystalline nature and surface smoothness of few nanometer levels. Fundamentally also, MgO with its simple cubic rock salt structure is an attractive model system to investigate oxide surface chemistry. Since it is a prototype ionic insulator with a wide band gap. motivation is high to grow MgO films on a conductive substrate useful for charged-particle measurement, in addition to the applications mentioned above. Moreover, the usefulness of MgO films as buffer layers for the growth of high Tc superconducting [5], and ferroelectric [6,7] films has led to an increased interest in the growth of this material.MgO films can be prepared by a variety of deposition techniques including ion-beam-assisted deposition [8], metal organic chemical vapor deposition [9],electron beam evaporation [10] and sputter deposition [11].Pulsed laser deposition (PLD) has been also successfully applied due to its flexible characteristics. Fork et al.[12] reported the epitaxial growth of (1 0 0)-oriented MgO on Si(1 0 0) using a Mg metal target. In the present work, we report study of the structural, optical, and electrical properties of MgO film deposited by reactive PLD on glass substrate as a function of oxygen deposition pressure.

Experiments

The MgO thin films were deposited on cleaned glass substrates by using Q-switched Nd:YAG laser having $\lambda = 1.064\mu m$, 7ns full width at half maximum. The laser beam was focused using a 4.8 cm lens onto a rotating Mg target with purity >

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99.9% (Aldrich chemical company) located at 45° angle of incidence. Figure (1) shows the schematic diagram of the PLD system used in this work. The laser fluence at the target was kept constant at 0.7J/cm^2 each film was made using15 laser shots. The substrates were cleaned with standard method. Films were deposited at substrate temperature of 150 °C (optimum temperature) [13] in an oxygen ambient at pressure ranging from (50-300) mbar. A stylus profile meter was then used to measure the film thickness. The electrical resistance of the grown films was investigated using a two point probe method, and hence the resistivity r could be obtained using the standard equation $R = \rho \frac{l}{A}$ and the sheet then equal to $R_{sh} = \frac{\rho}{t}$. The transmittance of the films was examined in the spectral range of (200–1100 nm) using UV-VIS Shimatzu double beam spectrophotometer-ray.

Diffractometer (Philips) using CuKα radiation (k = 0.1548 nm) was used to study the crystal structure of the grown film, the scan speed was 3°/min. The morphology of the films was investigated using an optical microscope.

Results and discussion

Figure (2) shows the MgO film thickness versus background oxygen pressure. The film thickness decreases with the increasing oxygen pressure. This behavior is attributed primarily to the increased collision of the ablated MgO particles with the ionized gas plasma during deposition [14, 15].

The XRD spectra of the MgO films deposited at various oxygen pressures are demonstrated in Fig. (3). the deposited film at pressure 50 mbar exhibited polycrystalline structure and the reflected Peaks, Located at 32.2^o, 34.5^o, 37.9, and 42.9^{o}are corresponding to a small amount of unioxidized Mg grains mixed with the oxide. for oxygen pressure at 100 mbar and 150mbar, a high quality MgO films with (111) and (002) orientation planes were obtained. Further increase in the oxygen Pressure (200 mbar) resulted in the production of highly oriented grains in the direction of (111) and (002) with FWHM 2 and 1.3 respectively. This result is close to obtained by Zhu et al [12].

Figure (4) displays the optical transmittance as a function of wavelength for MgO films prepared at different Oxygen pressures. The films demonstrate low optical transmittance at low oxygen pressure and were black in color as shown in Fig. (5a). the coloration of the film is attributed to excessive Mg ions existing at interstitial sites.

Excellent optical properties with high transmittance (~ 80%) in the visible region was noticed for MgO films deposited at 200 mbar, this film shows good Uniformity and it was approximately free from any Mg droplets as shown in Fig. (5b). The improvement in the transmittance can be also ascribed to an increase in the grain size of the films (smaller FWHM) as noticed in the XRD spectra.The optical band gap of film was determined from αhυ versus photon energy plots (α is the absorption Coefficient and hυ photon energy) as presented in Fig. (6). the band gap energy (Eg) of film at optimum condition (oxygen pressure 200 mbar, and substrate temperature 150°C) was 5.01eV. Generally, it can be stated that this reduced band gap energy of MgO may be due to varied extent of non-stoichometry of the deposited layers. But, interestingly these observed band gap energies of MgO film is lower than the band gap value of bulk MgO (7.8 eV) which may be due to the Various lattice associated atomic interaction phenomena come into play from its ionic crystalline nature [16]. As far as ionic crystal lattice is concerned, the band gap separation or variation is entirely different in thin films compared to bulk material [17]. Here the energy gap between the conduction Band and the valence band is perturbed which is mainly depends on the following electronic structure characteristics [18]: (i) the Made lung energy due to charge-charge interactions in the system, (ii) the delocalization energy due to electron sharing between atoms, (iii) the internal energy due to the filling of atomic orbital and intra-atomic electron-electron Interactions and (iv) the short-range repulsion energy between atoms that prohibits the atoms not to come close to each other. In addition, the presence of oxygen vacancies also induces changes in the electronic structure of surfaces. On planar surfaces, the Madelung potential is weakened leading to a reduction of the ionic gap, and in turn, its total band gap becomes smaller compared to bulk.

The electrical resistivity of MgO films strongly depend on their structure (grain size and shape, defect etc) purity (concentration of impurities like) and absorbed gases (moisture, etc) and on the preparation conditions. From the current-voltage measurement of the MgO film at optimum condition, it appears that the film has a resistance of ($6*10^{11}$ Ω) and resistivity of about (1.45 $*10^{7}$Ωcm). These results are similar to that obtained by other workers [19].

Conclusions

Highly transparent and dielectric MgO films have been prepared on glass substrates by reactive PLD. Structural, and optical, characteristics of the films as a function of oxygen pressure had been investigated .The XRD data confirmed the formation of highly oriented MgO films in the direction of (111)and (002)planes. The films deposited at oxygen pressure of 200 mbar have the height resistivity with an average transmittance of 80% in the visible region and an optical band gap of 5.01eV.

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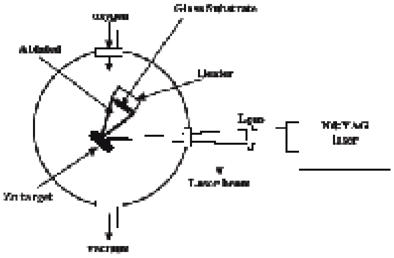

Figure(1)Schemati diagram of PLD system

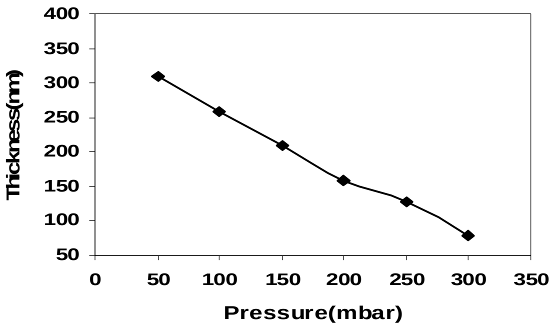

Figure (2) Film thickness vs. O_2 pressure Mg target

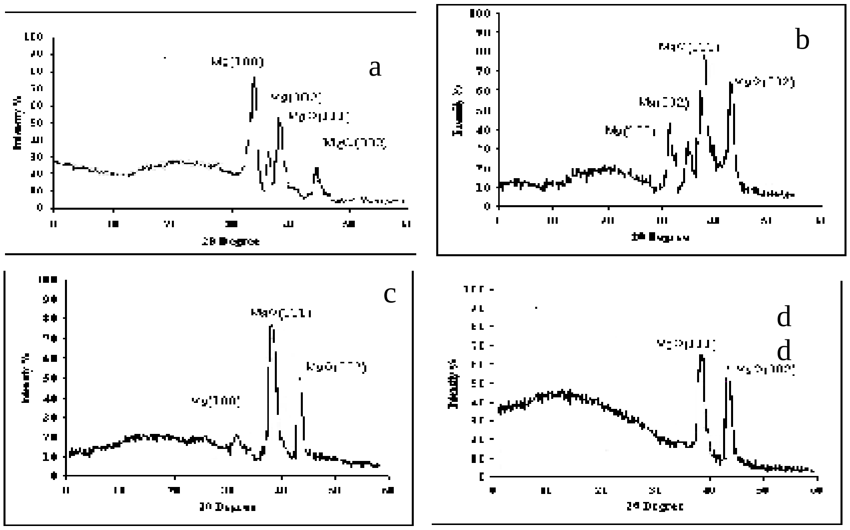

Figure (3) XRD spectra of MgO films at(a)p=50mbar,(b)p=100mbar , (c) p=150mbar and(d) p=200 mbar.

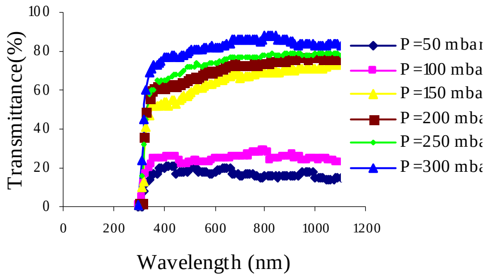

Figure (4) Transmittance as a function of oxygen pressure

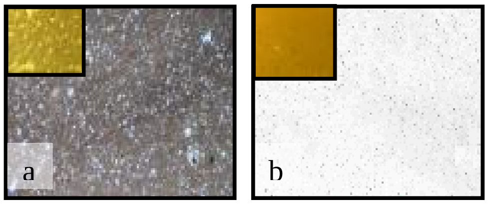

Figure (5) Optical micrograph of MgO film (a) 50mbar (b) 200 mbar.

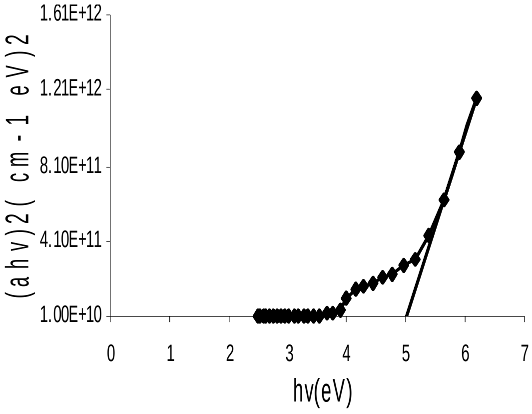

Figure (6) Energy band gap of the MgO at pressure (200 mbar) and T_{sub} (150°C).