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Ion beam induced modifications in electron beam evaporated aluminum oxide thin films

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Abstract

Al_2O_3 thin films find wide applications in optoelectronics, sensors, tribology etc. In the present work, Al_2O_3 films prepared by electron beam evaporation technique are irradiated with 100 MeV swift Si^{7+} ions for the fluence in the range 1×10^{12} to 1×10^{13} ions cm^{-2} and the structural properties are studied by glancing angle X-ray diffraction. It shows a single diffraction peak at 38.2° which indicates the γ-phase of Al_2O_3. Further, it is observed that as the fluence increases up to 1×10^{13} ions cm^{-2} the diffraction peak intensity decreases indicating amorphization. Surface morphology studies by atomic force microscopy show mean surface roughness of 34.73 nm and it decreases with increase in ion fluence. A strong photoluminescence (PL) emission with peak at 442 nm along with shoulder at 420 nm is observed when the samples are excited with 326 nm light. The PL emission is found to increase with increase in ion fluence and the results are discussed in detail.
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1. Introduction

Most solid state technologies are based on thin films. Thin films are usually prepared using vacuum techniques such as evaporation, sputtering or molecular beam epitaxy and is often combined to form complex multilayer structures for various applications. The most important microstructural properties of thin films are grain size, crystallographic texture and surface roughness [1]. Al_2O_3 is an important material used in planar wave guides because of its relatively high refractive index ($n = 1.64$) [2]. Further, Al_2O_3 films find wide applications – microelectronic devices, resistive coatings, catalysts, optoelectronics, sensors, tribology, corrosion protective coatings etc. [3,4]. Its wide usefulness is primarily due to its hardness, high melting point and low electrical conductivity at high temperature. Swift heavy ion irradiation is a unique post deposition treatment used to modify structural, optical and optoelectronic properties of thin film samples through the intense interaction of incident ions with the target atoms. There are several reports on structural and microstructural phase transformation due to irradiation using high energy heavy ions in thin films. Ratheesh Kumar et al. [5] have studied 100 MeV Au ion irradiated In_2S_3 thin films. They reported that on irradiation, crystallinity became poor and the sample became amorphous at 1×10^{13} ions cm^{-2}. Further, at 3×10^{13} ions cm^{-2} the samples show recrystallization. Kumar et al. [6] reported swift heavy ion (SHI) induced optical modifications in LiF thin films. They studied 80 MeV Ni ion induced color

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centers in LiF thin films using optical absorption and photoluminescence (PL) spectroscopy. Optical absorption studies show a broad band with peak at 445 nm corresponding to F_2/F_3^+ color centers. PL spectra show two broad bands of F_2 and F_3^+ color centers. The intensity of both the color centers increases up to 1×10^{12} ions cm^{-2} followed by an exponential decrease.

Investigations of the radiation damage in Al_2O_3 under high energy heavy ion irradiation are currently a big problem [7]. Also, the study of radiation damage of Al_2O_3 has significance as this material is presently used in radiation dosimetry. In the present work, the effect of heavy ion irradiation on structural and optical properties of Al_2O_3 thin films has been studied and the results are reported.

2. Experimental

The Al_2O_3 films are deposited on Silicon ⟨1 0 0⟩ substrate by an electron beam evaporation method under a vacuum of 10^{-6} Torr. Aluminum oxide samples (Sigma–Aldrich) are used as target. The substrates are kept at room temperature (RT). The thickness of the films of 100 nm is obtained by monitoring a quartz crystal oscillator during deposition. After deposition, the films are annealed at 500 °C for three hours in oxygen atmosphere in a tubular furnace. One of these films is kept as pristine and the other films are irradiated with 100 MeV swift Si^{7+} ions for the fluence in the range 1×10^{12} to 1×10^{13} ions cm^{-2} using the 15 UD Pelletron at Inter University Accelerator Centre, New Delhi. The glancing angle (1°) X-ray diffraction (GAXRD) studies are carried out for structural characterization using Brucker AXS X-ray diffractrometer. Surface morphology is studied using Digital Instruments, Nanoscope IIIa, atomic force microscope (AFM) in the Tapping mode. PL studies are performed using excitation by 326 nm from He–Cd laser (KIMMON) and Mechelle900 spectrograph in 200–1100 nm wavelength region. The PL setup has a cooled CCD array-based detection system. The laser light is incident on the sample at 45° and the luminescence light is collected using a collector assembly and transmitted to the spectrograph through optical fiber for detection and analysis.

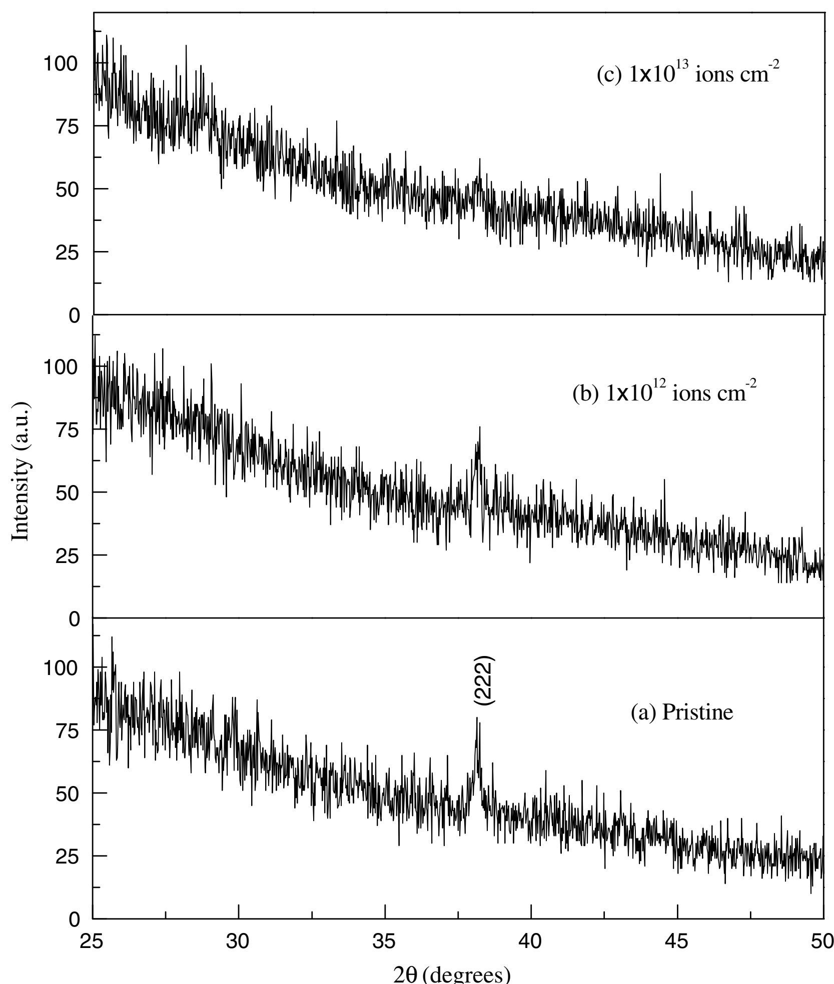

Fig. 1. Glancing angle (1°) X-ray diffraction patterns of electron beam evaporated Al_2O_3 thin films annealed at 500 °C (a) pristine and (b) and (c)100 MeV Si^{7+} ion irradiated films.

3. Results and discussions

3.1. Glancing angle X-ray diffraction

When an energetic ion passes through a matter, it loses energy by elastic scattering (nuclear energy loss) and inelastic scattering (electronic energy loss). The electron energy loss (S_e) and nuclear energy loss (S_n) of 100 MeV Si^{7+} ions in Al_2O_3 matrix is calculated using SRIM 2003 program and the values are estimated to be 4.412 keV nm^{-1} and 0.003 keV nm^{-1} respectively and the range for Si^{7+} ions is calculated to be 19.6 μm. The GAXRD patterns of electron beam evaporated, 500 °C annealed (a) pristine and (b and c) 100 MeV Si^{7+} ion irradiated aluminum oxide films are shown in Fig. 1. In the pristine film a diffraction peak at 38.2° due to (222) reflection is observed. As we know the Al_2O_3 exists in several distinct crystallographic phases such as γ, δ, θ and α-Al_2O_3. The observed diffraction peak is attributed to γ phase of aluminum oxide [8]. It is observed that the diffraction peak intensity decreases for the films irradiated with fluence 1×10^{12} ions cm^{-2}. Further, with increase in ion fluence the diffraction peak completely disappears at 1×10^{13} ions cm^{-2}. The sample in this case is amorphized as a result of cascade quenching with swift heavy ion irradiation [9]. The possible explanation for this kind of structural modification induced by SHI irradiation can be explained by total energy deposited in electronic excitations or ionizations in the films by energetic ions. The imparted energy of the incoming ions in our films at higher fluence may lead to overlapping of tracks to promote lattice disordering inside large grains [10].

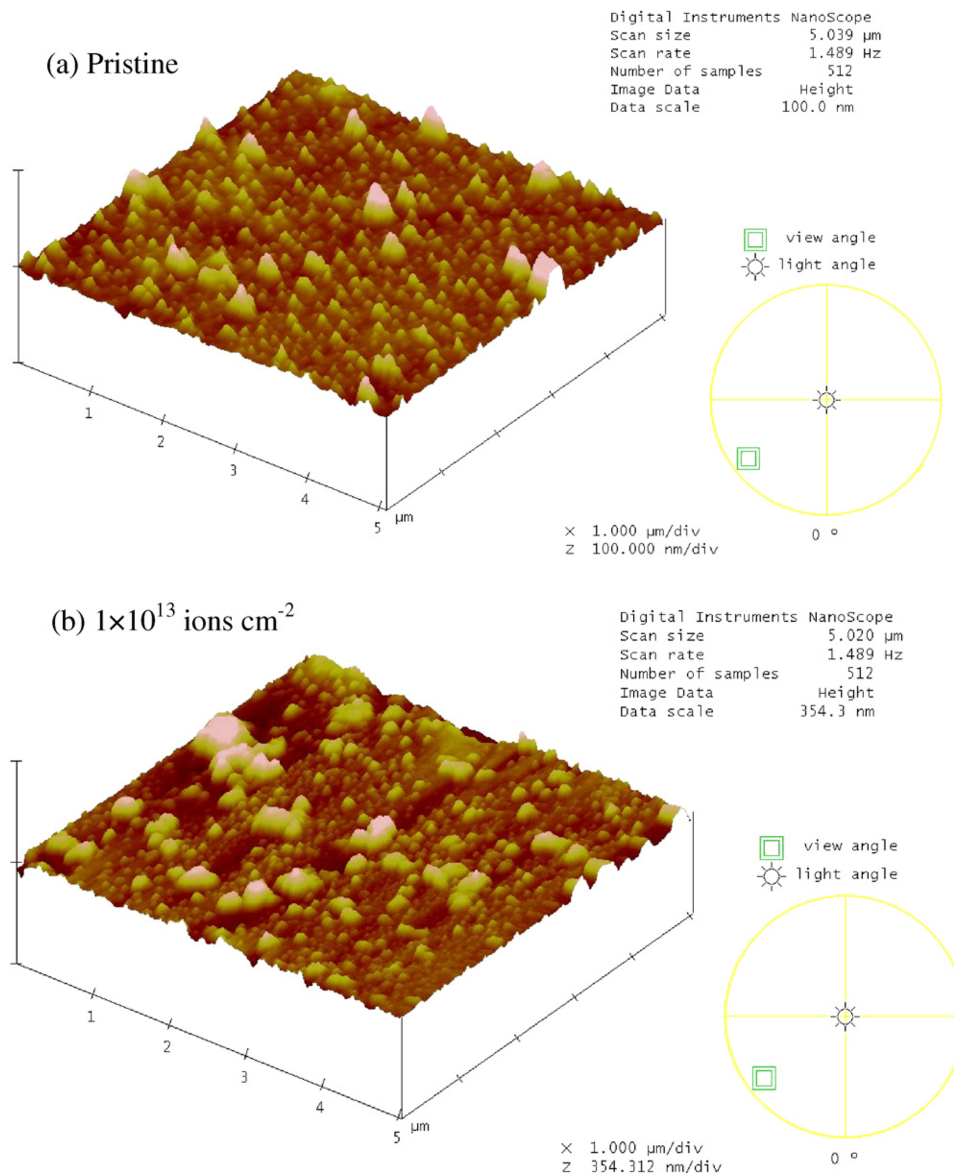

Fig. 2. AFM images of (a) pristine and (b) 1×10^{13} ions cm^{-2} irradiated Al_2O_3 thin films.

3.2. Atomic force microscopy

AFM is a very effective tool for examining surface modifications and surface structures and can be applied to understand the modification in the roughness and morphology of films upon SHI irradiation. Fig. 2 shows $5 \times 5\ \mu m^2$ 3D-images of (a) pristine and (b) 1×10^{13} Si^{7+} ions cm^{-2} irradiated aluminum oxide films. It is observed that conical shaped hillocks are uniformly distributed in pristine sample. In samples irradiated with 1×10^{13} ions cm^{-2}, the conical shaped hillocks disappear and randomly distributed granular shaped hillocks have been observed as can be seen from Fig. 2(b). Also, the surface is smoother in irradiated sample than the pristine one. The roughness of the films was estimated by the software attached with Nanoscope IIIa. The $5 \times 5\ \mu m^2$ images are utilized for measuring the surface roughness of the films. The roughness of pristine film is measured to be 34.73 nm. Further, the roughness of the samples decreases with increase in ion fluence. The roughness of 1×10^{12}, 5×10^{12} and 1×10^{13} ions cm^{-2} is found to be 29.49, 17.85 and 9.28 nm, respectively. The decrease in surface roughness value might be due to discontinuous tracks that may lead to amorphization as well as simple defects such as color centers. The XRD results revealed the amorphization of films as discussed in Section 3.1

3.3. Photoluminescence

The defects production mechanism may be understood by PL technique. PL is a very sensitive tool as compared to the optical absorption for the lower concentration of defects. Further, from PL studies one can identify the presence of defects whose optical absorption bands overlap. Fig. 3 shows the photoluminescence spectra of electron beam evaporated Al_2O_3 thin films irradiated with 100 MeV Si^{7+} ions. When these samples are excited at 326 nm light the pristine films show very weak luminescence response while

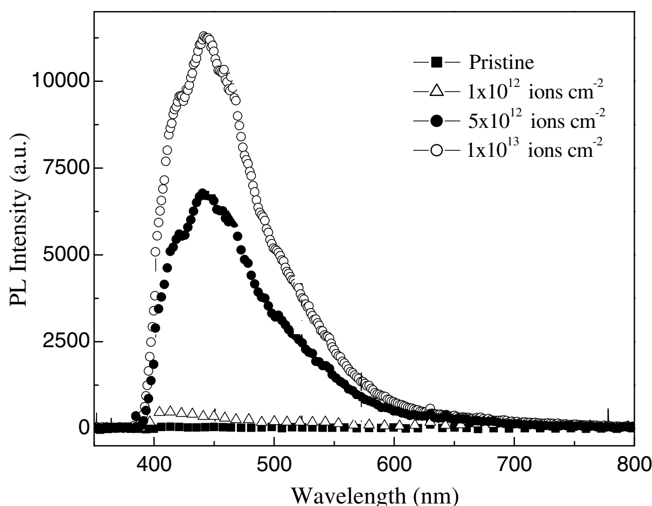

Fig. 3. Photoluminescence ($\lambda_{ex} = 326$ nm) spectra of pristine and 100 MeV Si^{7+} ion irradiated Al_2O_3 thin films.

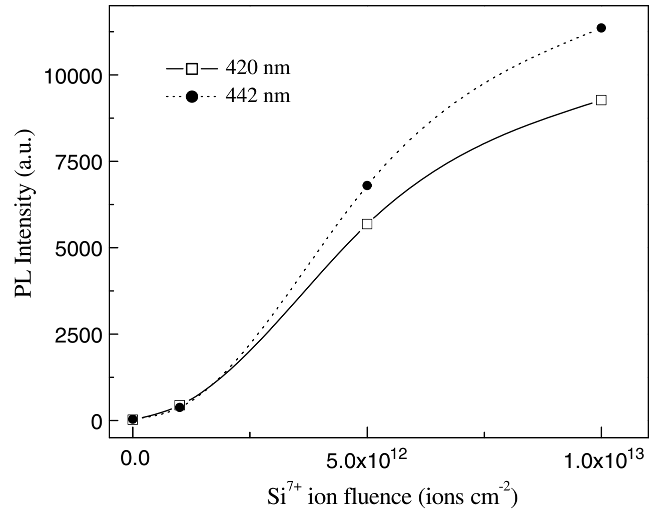

Fig. 4. Variation of PL intensity with ion fluence in 100 MeV Si^{7+} ion irradiated Al_2O_3 thin films.

SHI irradiated films shows a prominent and strong PL emission with peak at 442 nm along with shoulder at 420 nm. These emission bands arise from SHI induced color centers such as F and F_2^{2+}-centers, respectively. Skuratov and coworkers [11] studied the radiation damage under 245 MeV swift Kr ion irradiation in aluminum oxide. They reported three PL emissions with peaks at 330, 420 and 510 nm and ascribed to F^+, F and F_2-centers, respectively. Therefore, it is believed that 420 nm PL emission in the present studies may be attributed to F-centers. Further, the prominent PL emission at 442 nm might be due to F_2^{2+}-centers. Song et al. [12] have studied the photoluminescence using 350 nm light as excitation source in 230 MeV Pb ion irradiated aluminum oxide. They reported PL emission with peak at 380, 413, 450 and 516 nm in irradiated samples. They attributed the 450 nm emission to latent excitation luminescence of F_2^{2+}-centers whereas PL emission at 380, 413 and 516 nm were attributed to F_2^+, F and F_2-centers respectively. Hence, 442 nm emission in the present work may be attributed to latent excitation luminescence of F_2^{2+}-centers. However, electron spin resonance (ESR), Raman spectroscopy etc., studies may throw some light on better understanding of the defects responsible for luminescence. Fig. 4 shows the variation of PL intensity at 420 and 442 nm with ion fluence. The data point at zero fluence represents the PL intensity of unirradiated films. It is observed that PL intensity is low at 1×10^{12} ions cm^{-2} and it increases after 5×10^{12} ions cm^{-2}. This can be attributed to increase in concentration of color centers responsible for luminescence through radiative recombination [13].

4. Conclusions

Electron beam evaporated Al_2O_3 thin films annealed at 500 °C show a single XRD peak at 38.2° which indicate γ-phase of Al_2O_3. The films are amorphized when irradiated with 100 MeV Si^{7+} ions for the fluence 1×10^{13} ions cm^{-2}.

The pristine film shows roughness of 34.73 nm. The roughness of the irradiated films decreases with increase in ion fluence and is found to be 9.28 nm at 1×10^{13} ions cm^{-2}. The decrease in surface roughness is attributed to discontinuous tracks that may lead to amorphization. Further, the irradiated films show a prominent and strong PL emission with peak at 442 nm along with shoulder at 420 nm. In pristine and samples irradiated at lower fluence, the PL emission is very weak and PL intensity shoots up at and above 1×10^{12} ions cm^{-2}. This is due to increase in concentrations of color centers. The 442 nm emission is attributed to latent excitation luminescence of F_2^{2+}-center and 420 nm emission is attributed to F-centers.

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