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Effect of Swift Heavy Ion Irradiation on Dielectric Properties of Manganite Based Thin Films

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Abstract

In this communication, we report the results of the investigations on the structural, microstructural and electrical properties of $Y_{0.95}Sr_{0.05}MnO_3$ (YSMO) thin films with 100 nm thickness grown on *(100)* single crystalline $SrNb_{0.002}Ti_{0.998}O_3$ (SNTO) substrates using pulsed laser deposition (PLD) technique. YSMO films were irradiated using 100 MeV O^{+7} swift heavy ions (SHI) with different fluence using 15 UD Tandem Accelerator, IUAC, New Delhi facility. Structural investigations using $\theta - 2\theta$ X – ray diffraction (XRD) reveal that all the films crystallize in single phasic nature. With increase in ion fluence, structural strain is found to increase upon the irradiation using lower fluence of 1×10^{11} ions/cm^2 which can be ascribed to the irradiation induced creation of defects at the interface. Further increase in ion fluence up to 1×10^{12} ions/cm^2 can create more defects resulting in the degradation in lattice structure and hence strain gets enhanced. For the higher ion fluence of 1×10^{13} ions/cm^2, local annealing takes place at the interface which in turn results in the improved structural strain and recrystallization process. To study the surface morphology of the pristine and irradiated films, atomic force microscopy (AFM) measurement was performed. To understand the electrical properties of presently studied YSMO / SNTO pristine and irradiated films, frequency dependent dielectric behavior was studied. It is seen that dielectric constant increases upon irradiation using the fluence of 1×10^{11} ions/cm^2 and 1×10^{12} ions/cm^2 while gets suppressed for 1×10^{13} ions/cm^2 fluence. Variation in dielectric behavior with different ion fluence has been discussed on the basic of lattice strain and universal dielectric response (UDR) model.

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Keywords: Manganite; Thin films; SHI irradiation; Dielectric.

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1. Introduction

Discovery of colossal magnetoresistance (CMR) effect in rare earth manganite materials having a general chemical formula is $R_xA_{1-x}MnO_3$ where R is rare earth ions and A is monovalent/divalent ions, prompted intense work for their transport, magnetotransport and magnetic properties [1–11]. Rare earth manganese oxide materials have large number of applications such as various sensors (like magnetic field, temperature and electric field), p–n junction devices, electric field transistor, switching devices, etc [12–15]. Swift heavy ion (SHI) irradiation is most important tool to create defects in various oxide materials like superconductor [16], manganite [17], multiferroics [18] and ferrites [19]. SHI irradiation creates the defects at the interface between the film and substrate [17]. Many reports are available on the studies on manganite based thin film devices for the effect of irradiation on electrical [20], magnetic [21] and dielectric properties [22]. Also, few reports are available on dielectric studies pure and doped $YMnO_3$ manganites [23–27]. Still no report is available on the studies on the effect of SHI irradiation on dielectric properties of PLD grown $Y_{0.95}Sr_{0.05}MnO_3$ (YSMO) manganite based thin films. Thakur et al. [27] have studied the effect of strontium (Sr) doping in $YMnO_3$ for its dielectric, magnetic and thermodynamic properties of $Y_{1-x}Sr_xMnO_3$ (x = 0.1 & 0.2) samples, prepared by solid state reaction method. In this communication, we report the results of studies on the effect of SHI irradiation on dielectric properties of PLD grown YSMO films grown on SNTO single crystalline substrates. All the films were irradiated using 100 MeV O^{+7} SHI with different fluencies 1×10^{11} ions/cm^2, 1×10^{12} ions/cm^2 and 1×10^{13} ions/cm^2 using 15 UD Tandem Accelerator, IUAC, New Delhi facility. Effect of O^{+7} ions on dielectric constant has been discussed on the basis of lattice strain state and UDR model.

2. Experimental Details

High quality single phasic polycrystalline bulk $Y_{0.95}Sr_{0.05}MnO_3$ (YSMO) target was synthesized by conventional ceramic method [28,29]. Y_2O_3, $SrCO_3$ and MnO_2 were used as starting materials and mixed in a stoichiometric ratio thoroughly for 3 hrs followed by calcinations (on well mixed powder) and sintering (on solid pellets) processes at different temperatures between 950 – 1350 °C for different time durations between 24 – 72 hrs. YSMO films, having a thickness of ~ 100 nm, were deposited onto single crystalline n–type semiconducting SNTO *(100)* substrates by ablating the bulk $Y_{0.95}Sr_{0.05}MnO_3$ target using 248 nm KrF excimer laser with ~ 1.80 J/Cm^2 energy at 10 Hz repetition rate. Deposition parameters are given in Table. 1 All the YSMO/SNTO films, of size 10×10 mm^2, were irradiated with 100 MeV O^{+7} ions with 1×10^{11}, 1×10^{12} and 1×10^{13} ions/cm^2 fluencies, using 15 UD Tandem Accelerator facility at Inter University Accelerator Centre (IUAC), New Delhi. To understand the structure properties and its dependence upon the SHI irradiation, XRD was carried out at room temperature using Cu Kα X–ray source. To understand the role of SHI irradiation in modifying the surface of all the irradiated films, AFM was performed at room temperature. Room temperature dielectric properties of the pristine and irradiated films were investigated using Agilent E4980A impedance analyzer (Agilent Technology) with frequency range from 10 KHz to 2 MHz.

Table 1. PLD parameters employed for the fabrication of YSMO / SNTO films.

Laser used	KrF Excimer
Targets	$Y_{0.95}Sr_{0.05}MnO_3$
Single Crystalline Substrates *(100)*	$SrNb_{0.002}Ti_{0.998}O_3$ (SNTO)
Laser energy	~ 1.80 J/cm^2
Repetition rate	10 Hz
Substrate temperature	700 ºC
Oxygen partial pressure	1 x 10^{-5} Torr
Substrate to target distance	5.5 cm

3. Results and Discussion

3.1. Structural Properties

XRD patterns of YSMO films grown on single crystalline SNTO *(100)* substrates show (figure 1) single phasic nature having the orientations in (*h00*) direction parallel to the substrate orientations. The lattice mismatch between YSMO film and SNTO substrate evident from the positional difference between film and substrate peaks at ~ 47º. From XRD patterns, it is clearly seen that peak position and intensity of film change with ion fluence of 100 MeV O^{+7} ions irradiation which leads to structural modifications and hence structural strain state change in the films. Lattice strain at the interface (δ) can be quantified as: $\delta\ (\%) = [(d_{substrate} - d_{film}) / d_{substrate}] \times 100$, where d is the lattice spacing in film / substrate. Table 2 lists the value of δ suggesting the tensile strain in all the films studied. With increase in ion fluence, defect density continuously increases up to ion fluence of 1×10^{11} ions/cm^2, after which, i.e. for higher ion fluence, local heating gets generated at the interface resulting in the local annealing process and recrystallization and as a result, strain gets suppressed in this irradiated film [18].

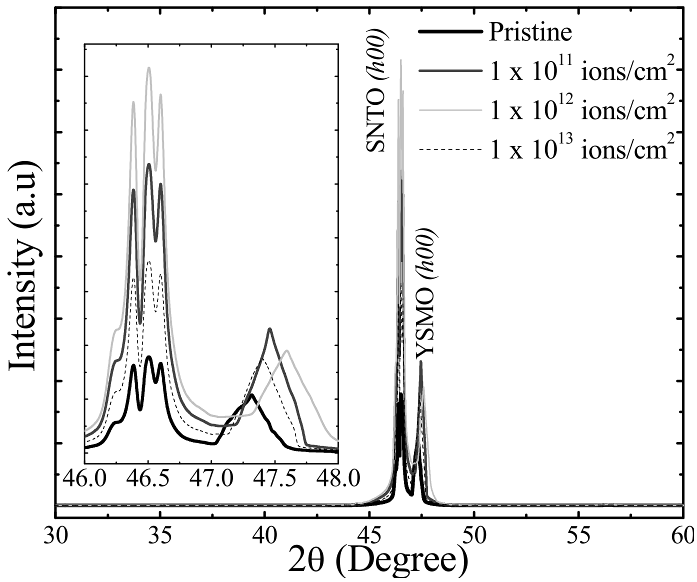

Fig. 1. XRD pattern of (a) YCMO/Si thin film; (b) an enlarged view of *(100)* peaks.

Table 2.Values of strain (δ) and rms surface roughness (RSR) for pristine and irradiated YSMO / SNTO films.

YSMO Film 100nm	Strain (δ) %	Surface roughness
Pristine	+1.53	09.41 nm
1 x 10^{11} ions/cm^2	+1.85	11.69 nm
1 x 10^{12} ions/cm^2	+2.09	12.66 nm
1 x 10^{13} ions/cm^2	+1.72	15.18 nm

3.2. Microstructural Properties

Figure 2 show that modifications in the surface morphology of pristine and irradiated films studied using AFM (2D and 3D) images. It can be seen from figure 2 that, pristine film possesses homogeneous rectangular shaped granular islands which gets modified into hillock like defects onto surface after irradiation with 1×10^{11}, 1×10^{12} and 1×10^{13} ions/cm^2 fluence.

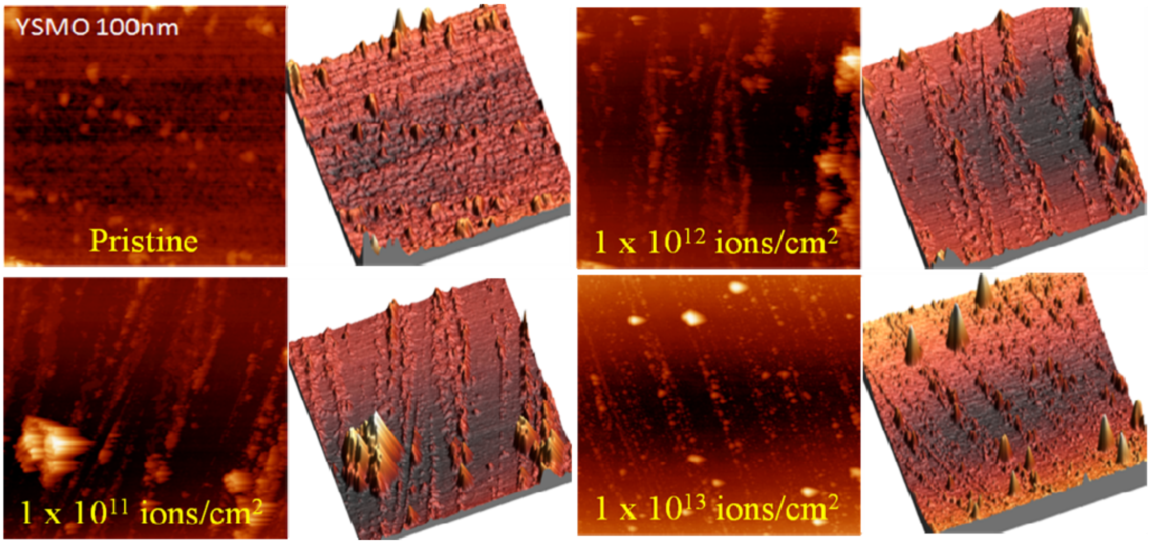

Fig. 2. Surface topography (2D and 3D images) of pristine and irradiated YSMO / SNTO films.

3.3. Dielectric Behavior

Variation in dielectric constant with frequency (in logarithmic scale) for pristine and irradiated films, obtained at room temperature, is shown in figure 3. Dielectric constant decreases with increase in frequency for pristine and irradiated films. Generally, origin of dielectric effect is due to existence of grain and grain boundary which give localized accumulation of charges resulting in interfacial polarization. After irradiation, the value of dielectric constant increases with increase in ion fluence up to 1×10^{12} ions/cm^2 over all the frequency range. This can be understood by irradiation induced creation of defects and enhanced in structural disorder and increase in lattice strain at the interface between the film and substrate. For higher ion dose i.e. 1×10^{13} ions/cm^2, dielectric constant gets suppressed may be due to the improved crystallinity, reduced structural disorder and decrease in lattice strain.

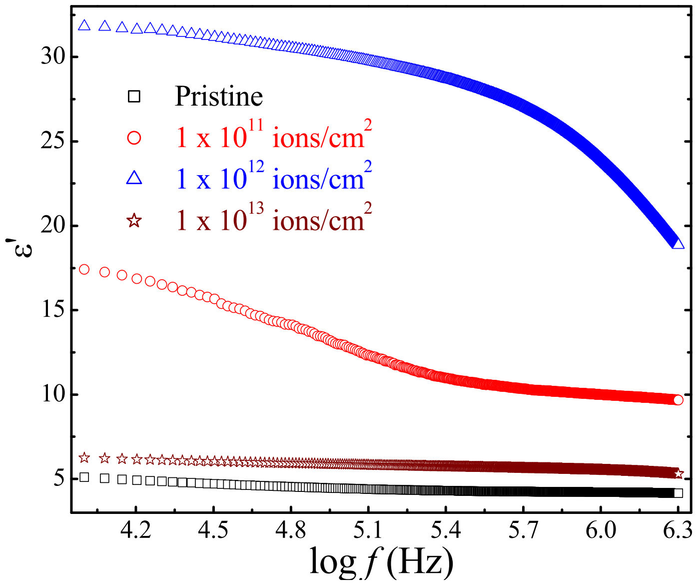

Fig. 3. Frequency dependent variation in dielectric constant (ε') of pristine and irradiated YSMO / SNTO films.

Dielectric properties of presently studied YSMO/SNTO junction have been understood by universal dielectric response (UDR) Model. Figure 4 shows the log (fε') vs. log (f) to understand the UDR model for the dielectric behavior in pristine and irradiated films. The plots show that the model fits well for pristine and irradiated films and value of slope varies non–monotonically with ion fluence suggesting a complex contribution of grains and grain boundary (including defects on the surface) to the dielectric response of the films. For YSMO/SNTO film irradiated with 1×10^{11} ions/cm^2, UDR model could not fit to the data and hence the log (fε') vs. log (f) curve of 1×10^{11} ions/cm^2 irradiated YSMO/SNTO film has been fitted in two different regions, as shown in figure 4. This can be due to the frequency induced modifications in dielectric mechanisms.

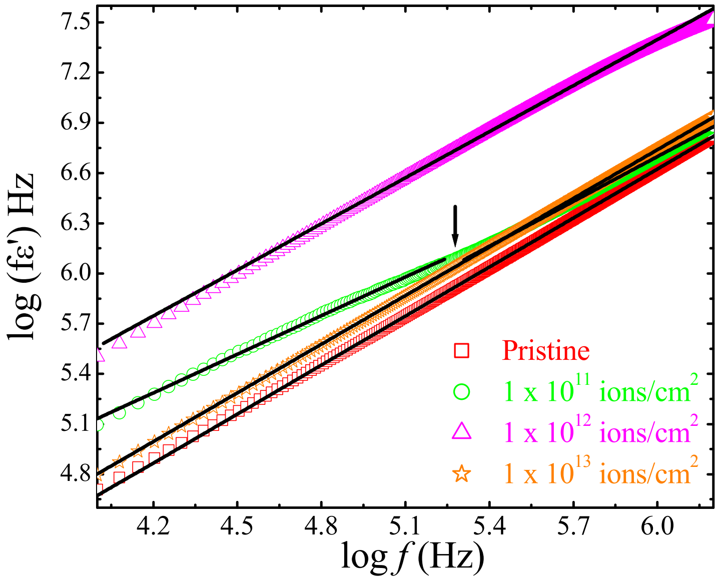

Fig. 4. Plots of log (fε') vs. log (f) with linear fits for pristine and irradiated YSMO / SNTO films.

4. Conclusion

In summary, we have successfully fabricated high quality YSMO thin film on single crystalline (*100)* SNTO substrates using pulse laser deposition (PLD) technique. XRD patterns reveal that all the films crystallize in single phasic nature having the YSMO orientations in (*h00)* direction parallel to the substrate orientations. Surface morphology has been studied by atomic force microscope (AFM). Surface roughness of the films gets increased upon irradiation due to the creation of hillocks on the surface of the irradiated films. Variation in dielectric behavior with ion fluence has been discussed in the context of lattice mismatch between the film and substrate resulting strain at the interface. The dielectric behavior has been understood on the basis of universal dielectric response (UDR) model for pristine and irradiated films.

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