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The Structural and Photoluminescence properties of Ta: In_2O_3 Thin Films grown by Pulsed Laser Deposition Technique

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Abstract. High purity Tantalum oxide and indium oxide powders are taken as starting materials to prepare Tantalum doped Indium oxide powder by solid state reaction method to prepare robust targets of $(In_{1-x}Ta_x)_2O_3$ (x=0.06) for Pulsed Laser Deposition (PLD) to grow Ta:In_2O_3 Thin films, prepared pellet was sintered at 1000ºC for 12hours. The films are deposited on ultrasonically cleaned (100) oriented Silica substrates are maintained at a substrate temperature of 400ºC. The vacuum chamber evacuated by a turbo molecular pump to a base vacuum better than 10^{-6} Torr and oxygen partial pressure was maintained at 0.3 mTorr. The deposited film structural characterization was done by GIXRD, EDX and stylus profile meter. UV-vis spectroscopy and Photoluminescence spectroscopy studies are performed for optical properties of the deposited films. GIXRD suggest polycrystalline nature with the preferred orientation along (222) direction. TEM images of the deposited films show nanometric grained rod like morphology. CIE plots confirm that deposited films are applicable for blue LED.

INTRODUCTION

Transparent conducting oxides (TCO) are unique materials that exhibit mutually exclusive material properties such as low resistivity and high optical transparency by utilizing these unique properties results in wide range of applications. TCO thin films have received much attention because of their wide applications such as thin film solar cells [1], Flat panel display[2],defrosters [3], opto-electronics [4], anti-static coatings[5].Organic light-emitting diodes [6]. Metal oxides exhibits unacceptably high resistivity for TCO applications. Thus, various metal oxides are employed to enhance the film conductivity. It has been widely reported that doping appropriate impurities into metal oxide films can decrease the resistivity by one or two orders of magnitude [7]. Among various TCOs Sn doped Indium oxide (ITO) is most widely used for optoelectronic applications because ITO thin films have high transmittance and low resistivity [8]. However the research focuses on new elemental dopings which can achieve better properties for prescribed applications. The doped material oxides should be chosen through consideration of both the charge and radius of the atom. There are some depends such as In_2O_3:Mo[9],In_2O_3:Ti[10] and In_2O_3:Zr[11] . Ta is one of the promising dopant because of its ionic radius (Ta^{5+}) is nearly similar and smaller than that of In^{3+} ions.There are some reports on Ta doped metal oxides but relatively very less number of papers available in the literature on Ta doped metal oxides prepared by Pulsed Laser Deposition (PLD).

EXPERIMENTAL

In the deposition of In_2O_3 thin films, to achieve desirable film growth the first priority goes to cleaning of the substrate.Undoped and Ta doped In_2O_3 thin films are grown on Silica (100) substrates by PLD technique. The vacuum chamber was evacuated by the Turbomolecular pump. The source material $In_2O_3$and Ta_2O_5 of 99.999% purity is pelletized by taking the fine powder from Sigma-Aldrich Chemicals. The well-grounded powder is heated in air at 800^o C for 10 h and the prepared powder is cold pressed at 10 ton load and made the pellet of 2 cm diameter and 2 mm thickness. This pellet was sintered at 800^{o}C for 12 hours.

The undoped and Ta doped In_2O_3 thin films were deposited on Silica (100) substrates attached with a miniature heater, to maintain substrate temperature at 673 K. A KrF (248 nm) excimer laser (Lambda PhysikCOMPex) operating at a pulse repetition rate of 10 Hz and a 220 mJ of energy is used for deposition. The energy density of the laser beam is kept at 2 J/cm^2. Initially, 2x10^{-6} Torr base pressure is maintained in the vacuum chamber. To maintain an oxygen atmosphere in the chamber, a needle valve is used to admit oxygen into the chamber. The final pressure is maintained inside the vacuum chamber at oxygen ambient pressure of 0.1 m Torr.

All deposited films are characterized structurally using GIXRD (Bruker D8 Advance, USA). GIXRD is carried out using nickel-filtered Cu-Kα radiation (λ= 0.15418 nm) under 40 kV voltage and a current of 30 mA. For all the measurements, Diffraction space is spanned over a 2θ range of 15–80^o, to investigate the structural and crystallographic phases present in the films. The average size of Crystallites (D) of In_2O_3 films is estimated using Debye Sherrer formula.

The film morphology is examined using SEM (Carl ZEISS EVO 18, Germany). The elemental composition is recorded with Energy-dispersive X-ray spectroscopy attached with SEM.

Photoluminescence (PL) measurements were performed on JobinYvon Fluorolog-3 spectrofluorimeter.

RESULT AND DISCUSSION

GIXR

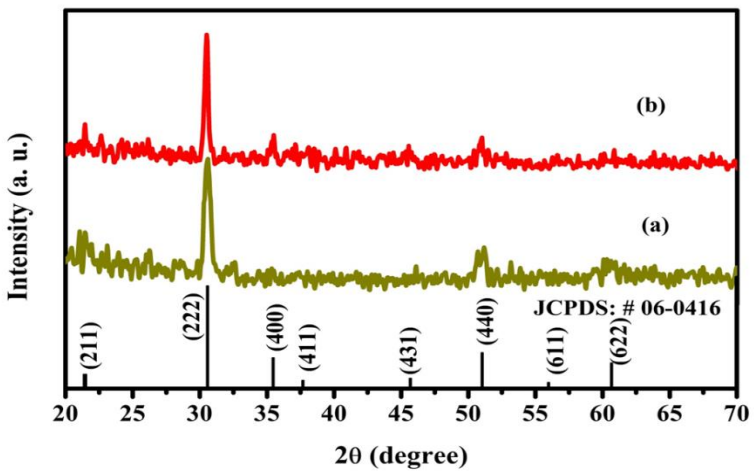

FIGURE 1. GIXRD spectra (a) Undoped In_2O_3 thin film, (b) Ta: In2O3 thin film.

The Structure and crystallinity of the un doped and Ta^{+5} doped In_2O_3 thin films are by Glancing X-Ray diffraction patterns are shown in Figure 1(a) (b). The GIXRD pattern for the films of undoped and Ta^{+5} doped In_2O_3 thin films deposited matched with JCPDS Cord No.06-0416 and find the cubic bixbyite phase of deposited thin films. No additional diffraction peaks are observed even after Ta^{+5} doped thin films.Planes (211), (222), (400), (411), (431), (440), (611), and (622) with their corresponding diffraction angles 21.49, 30.57, 35.46, 37.68, 45.68, 51.03, 55.98 and 60.67 respectively. Furthermore, peaks of Ta+5 shifts towards lower 2Θ as compared with Un doped In2O3 thin films it was indicated by slight increase of the lattice constant 'a' calculated by employing the Bragg's law and Debye Sherrer equation.Shift in lattice constant due to less ionic radius of Ta^{+5} than the In^{+3} ions. Direct substitution of Ti and Ta into the Indium oxide lattice also observed before for thin films deposited via Physical methods [12,13].

SEM and EDS

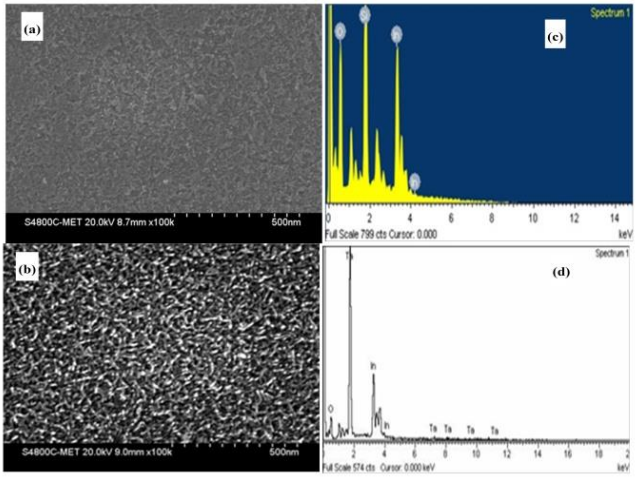

FIGURE 2.SEM and EDS image of (a) Undoped In_2O_3 thin film, (b) Ta: In_2O_3 thin film, (c) Undoped In_2O_3 thin film, (d) Ta: In_2O_3 thin film.

The Scanning electron microscopy was employed to examine the surface morphology of Undoped and Ta doped In_2O_3 thin films shown in Figure2.It shows the SEM and EDS pictures of Undoped and Ta doped the thin films deposited on Si substrate, Figure. 2 (a) Un doped In_2O_3Figure 2 (b) Shows Ta doped In_2O_3 thin film. The films deposited are showing uniformly distributed nano structures. Ta doped In_2O_3 thin films shown rod like morphology.

Figure 2(c, d) shown spectra of Undoped and Ta doped In_2O_3 thin films. In figure 2 (c) the presence of O and In in In_2O_3 sample indicates purity of the film deposited. In the figure 2 (d) the presence of Ta, O and In in Ta doped In_2O_3 sample indicates doping Ta it also confirms there is no extra peaks observed in GIXRD spectra. the stoichiometry is found to be well maintained in total film area it was confirmed by the EDS analysis was taken from different parts of the thin film.

RAMAN SPECTROSCOPY

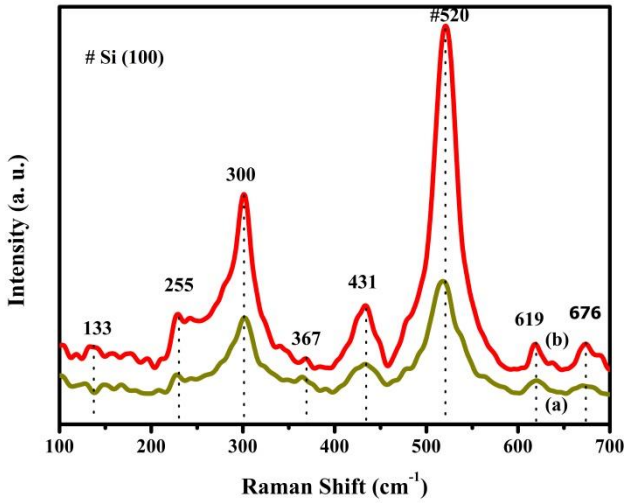

FIGURE 3. Raman spectra of (a) Undoped In_2O_3 thin film, (b) Ta: In2O3 thin film.

We performed the room temperature Raman scattering to identify the defect states of undoped and Ta doped In_2O_3 thin films. Figure 3 shows Raman shift data from 100 -700 cm^{-1}.with Body-centered cubic bixbyite structure with a space group I_{a3} (Inversion) that contains 8 formula units per Bravais unit cell.

The Raman spectra of undoped and Ta doped In_2O_3 thin films for different D_{ST} has shown in Figure 3 (a).The plots presents two sharp bands one at 300 cm^{-1} and the other at 520 cm^{-1} along with several another medium intense and weak intense bands. In comparison to the bulk material, thin films show weak Raman scattering. peak shifts and broadening are explained by the quasi-momentum conservation rules for vibrational mode relaxation that may arise due to decreasing of the crystalline correlation length in nanometric structures. The high-intensity band at ~300 cm^{-1}can be attributed to the bending vibrations of InO_6 at the Octahedron site and the band at ~619 cm^{-1} can be attributed to the stretching vibrations of InO_6 . The band at 520 cm^{-1} may be the spectral contribution from substrate. In Ta doped thin films there is an extra peak at ~676cm^{-1} is attributed to Ta+5 state. [14-16].

PHOTOLUMENISCENCE

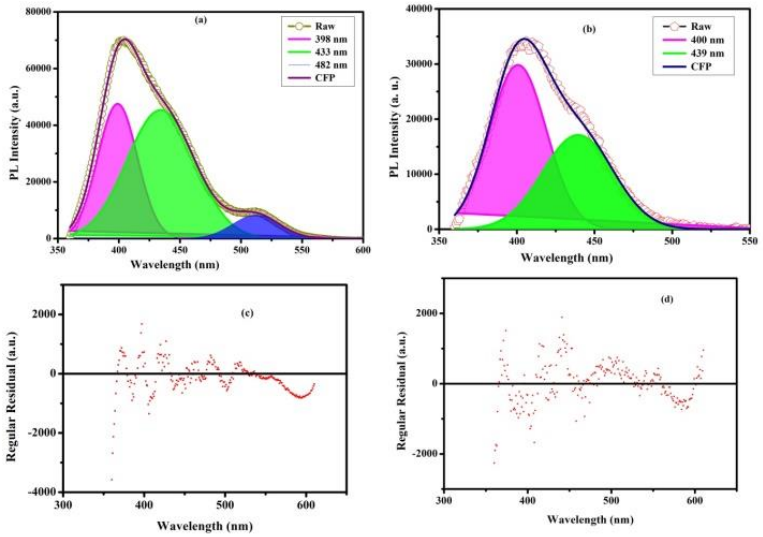

FIGURE 4.PL spectra of (a) Undoped In_2O_3 thin film, (b) Ta: In_2O_3 thin film, and corresponding residues of (c) Undoped In_2O_3 thin film, (d) Ta:In_2O_3 thin film.

PL spectra carried out with the excitation wavelength of 367 nm and plotted Pl intensity for Undoped and Ta doped In_2O_3 thin films and their corresponding general residues shown in figure 4 (a, b, c, and d). Un doped and Ta doped (Near Band Edge) NBE shown higher wavelengths region, in case of undoped thin film Oxygen interstitial sites and Indium interstitial sites intensity is high compared with Ta doped thin films. Doping of Ta into In_2O_3 the PL spectra indicates no evidence for the presence of In Interstitial sites.

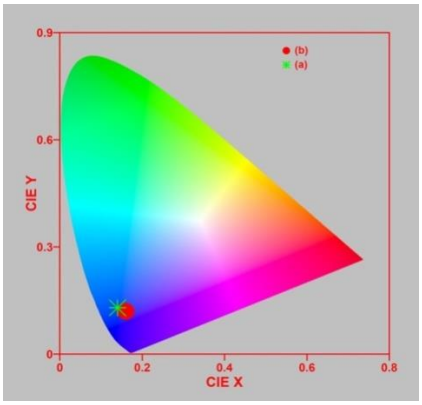

FIGURE 5. CIE (Commission International Eclairage) 1931 Chromaticity coordinates of (a) Undoped In_2O_3 thin film, (b) Ta: In_2O_3 thin film.

Figure.5.Shows CIE 1931 chromaticity diagrams for undoped and Ta doped In_2O_3 thin film. under 367 nm excitation. It can be clearly seen from figure CIE coordinates for (a) Undoped In_2O_3 thin film are (0.181,0.152), (b) (0.189, 0.150) coordinates of Ta:In_2O_3 thin films corresponding to the primary blue shade these coordinates are close to National Television Standard Committee (NTSC) standard [17].

SUMMARY

Undoped and Ta: In_2O_3 thin films were deposited using PLD. The films have shown structural, chemical, morphological consistency as verified from GIXRD, SEM, EDS and Raman Spectroscopy. XRDPatterns have revealed polycrystalline nature with cubic bixbyite structure. From Raman, the films are in phase by showing a peak of In_2O_3 at 303 $cm^{-1}$and Ta doped thin films shown 673 cm^{-1}peak belongs to Ta^{+5}. Photoluminescence confirms the variation of defect states with and without doping these films shown primary blue emission.

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