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Low-Resistivity Sputtered Films of Transparent Conducting Ta-Doped In_2O_3 Oxide

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Low-resistivity Ta-doped In_2O_3 (InTaO) films were grown on Corning # 1737 glass substrates from a ceramic target of $(In_{0.95}Ta_{0.05})_2O_3$ by radio-frequency magnetron sputtering. The electrical and the optical properties of these films were investigated by varying the oxygen partial pressure pO_2 (0 Torr $\leq pO_2 \leq 1.0 \times 10^{-4}$ Torr) and the deposition temperature T_S (25 °C $\leq T_S \leq$ 350 °C) during the deposition. The film grown at 350 °C and $pO_2 = 0$ Torr showed a resistivity as low as 0.28 mΩcm with a carrier density of 7.4×10^{20} cm^{-3}, a Hall mobility of 30.1 cm^{2}V^{-1}s^{-1}, an optical band gap of 4.04 eV, and an average transmittance above 85 % for wavelengths between 400 and 700 nm. These values are comparable to those of optimized Sn-doped In_2O_3 (ITO).

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I. INTRODUCTION

Because of their mutually exclusive properties of low electrical resistivity and high optical transparency in the visible range of the spectrum, transparent conducting oxides (TCOs) have been used as transparent contacts in numerous opto-electronic devices, including thin film transistor liquid crystal displays (TFT-LCDs) and photovoltaic cells [1, 2]. Among the three primary TCOs, In_2O_3, SnO_2, and ZnO, In_2O_3 doped with Sn (ITO) has become the TCO of choice. ITO has a low resistivity due to its high carrier electron concentration created by oxygen vacancies and substitutional tin dopants during the film growth. ITO also exhibits high transmission due to its wide band gap ($\sim$ 3.7 eV) in the visible and near-IR regions of the spectrum [3,4]. The typical resistivity, optical transparency, and carrier density of commercial ITO films grown on Corning glass are $\sim$ 0.2 mΩcm, $\sim$ 90 %, and $\sim 1 \times 10^{20}$ cm^{-3}, respectively. As the display size of TFT-LCDs increases, the demand is expected to grow for new TCO materials with lower resistivities while retaining good optical properties.

In search of improved TCO materials, we have studied the transport and the optical properties of Ta-doped In_2O_3 (InTaO) films. In this study, the film properties, such as the resistivity (ρ), the carrier density (n_H), the mobility (μ_H), and the optical transparency, have been investigated under different sputtering conditions, such as various oxygen partial pressures pO_2 (0 Torr $\leq pO_2 \leq 1.0 \times 10^{-4}$ Torr) and deposition temperatures T_S (25 °C $\leq T_S \leq$ 350 °C). In this paper, we report the structural, electrical, and optical properties of InTaO films prepared by radio-frequency (rf) magnetron sputtering.

II. EXPERIMENTAL DETAILS

Ta-doped In_2O_3 (InTaO) films were deposited on Corning # 1737 glass substrates by rf magnetron sputtering from a sintered $(In_{0.95}Ta_{0.05})_2O_3$ ceramic target. The sputtering chamber was pumped down to 2×10^{-6} Torr by using a turbo molecular pump. The depositions were performed with mixed Ar gas and O_2 gas at a total pressure of 5 mTorr. The Ar flow rate was fixed to 50 sccm, and the O_2 flow rate was varied from 0 to 1 sccm (thus, during the deposition, the oxygen partial

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pressure, pO_2, varied from 0 Torr to 1×10^{-4} Torr). During the film deposition, the substrate temperature was fixed at a desired temperature in the range from 20 to 350 °C. The film thickness was measured with a profilometer (AlphaStep 500 Surface Profiler, KLA-Tencor). The substrates were cleaned in an ultrasonic cleaner with acetone, ethanol, methanol, and de-ionized water sequentially and were finally dried with high-purity nitrogen gas (99.999 % purity).

The resistivity (ρ), the carrier density (n_H), and the Hall mobility (μ_H) were measured with the van der Pauw method at room temperature in a magnetic field of 3 kOe [5]. The temperature dependence of the resistivity was measured by using a standard four point probe technique. The crystalline structure of the films was analyzed by using X-ray diffraction (XRD) measurements with Cu-Kα radiation. The optical transmission was measured using a spectrometer equipped with an optical multi-channel analyzer.

III. RESULTS AND DISCUSSION

The electrical properties of Ta-doped In_2O_3 (InTaO) films were found to depend on the oxygen partial pressure pO_2 and the deposition temperature T_S. The carrier type of the InTaO films was found to be n-type. Fig. 1 shows the dependences of the electrical resistivity (ρ), the carrier density (n_H), and the Hall mobility (μ_H) of the InTaO films grown at T_S = 350 °C with pO_2 being varied from 0 Torr to 1.0×10^{-4} Torr. The resistivity increased from 0.28 mΩcm to ∼ 72 mΩcm as the pO_2 was increased from 0 Torr to 4×10^{-5} Torr and then remained nearly constant for pO_2 up to 1.0×10^{-4} Torr. Both n_H and μ_H decreased rapidly from 7.4×10^{20} cm^{-3} and 30.1 cm^{2}V^{-1}s^{-1} to 1.2×10^{19} cm^{-3} and 7.2 cm^{2}V^{-1}s^{-1}, respectively, as pO_2 was increased from

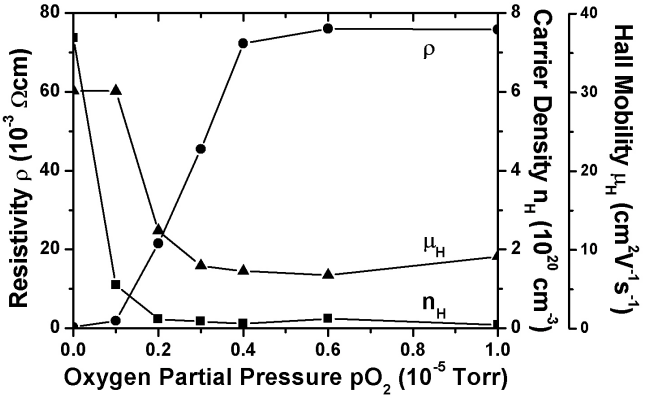

Fig. 1. Dependences of the electrical resistivity ρ(●), the carrier density n_H(■) and the Hall mobility μ_H(▲) of Ta-doped In_2O_3 (InTaO) films grown at a deposition temperature (T_S) of 350 °C on the oxygen partial pressures (pO_2).

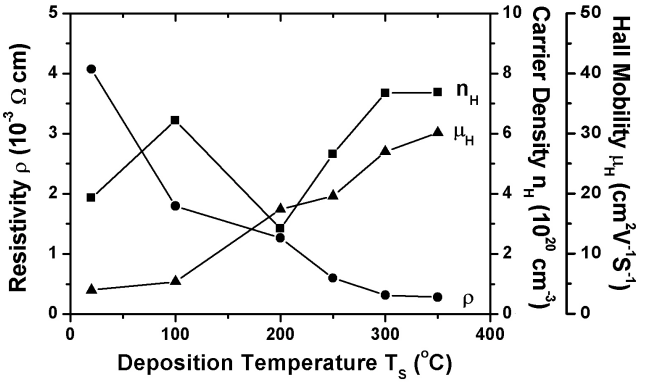

Fig. 2. Dependences of the electrical resistivity ρ(●), the carrier density n_H(■) and the Hall mobility μ_H(▲) of Ta-doped In_2O_3 (InTaO) films on the deposition temperature (T_S) for temperatures from 20 °C to 350 °C under an oxygen partial pressure of 0 Torr.

0 Torr to 4×10^{-5} Torr; then, they remained nearly constant for pO_2 up to 1.0×10^{-4} Torr. The minimum resistivity of 0.28 mΩcm with n_H of 7.4×10^{20} cm^{-3} and μ_H of 30.1 cm^{2}V^{-1}s^{-1} was observed for the InTaO films grown at pO_2 = 0 Torr. The high resistivity of the InTaO films grown at a high pO_2 may be explained by the extinction of the carrier density due to a decrease in oxygen vacancies [6].

Fig. 2 shows the dependence of the electrical resistivity (ρ), the carrier density (n_H), and the Hall mobility (μ_H) of InTaO films on T_S for temperatures from 20 °C to 350 °C at pO_2 = 0 Torr. The pO_2 = 0 Torr was chosen because the InTaO film grown at T_S = 350 °C with pO_2 = 0 Torr had the minimum resistivity. When T_S was increased from 20 °C to 350 °C, the resistivity of the InTaO films decreased from 4.1 mΩcm to 0.28 mΩcm, and the mobility increased from 4.0 cm^{2}V^{-1}s^{-1} to 30.1 cm^{2}V^{-1}s^{-1}. The increase in mobility with increasing T_S may be due to enhanced crystallinity. The decrease in ρ with increasing T_S was mainly due to enhanced μ_H. The carrier density was in the range of 3.9×10^{20} cm^{-3} ∼ 7.4×10^{20} cm^{-3} and showed an anomalous dependence on T_S with two maximum values, one at T_S = 100 °C and the other at 300 °C. The carrier electrons in InTaO films are generated from oxygen vacancies and Ta atoms that substituted for In atoms in the crystal lattice (activated Ta atoms). With increasing T_S, the carrier electrons from the oxygen vacancies are expected to decrease and the carrier electrons from activated Ta atoms to increase. The observed anomalous dependence of the carrier density on the deposition temperature, therefore, is attributed to the different deposition temperature dependences of the oxygen vacancies and the activated Ta atoms.

Since the minimum value (0.28 mΩcm) of the room-temperature resistivity of the InTaO films was observed in the InTaO film grown at 350 °C and pO_2 = 0 Torr, we have studied the transport and the optical properties

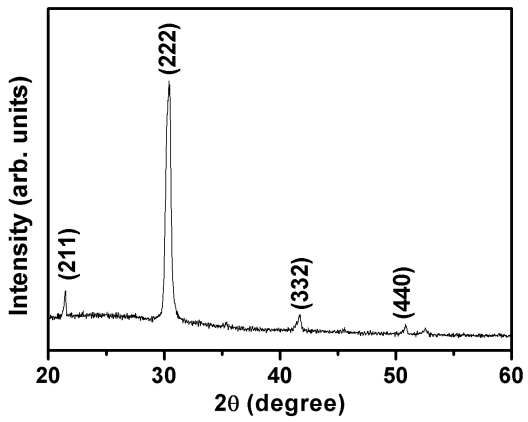

Fig. 3. Temperature dependence of the resistivity of the InTaO film grown at 350 °C with $pO_2 = 0$ Torr. The inset shows $\rho - \rho_0$ versus T^2 for the InTaO film.

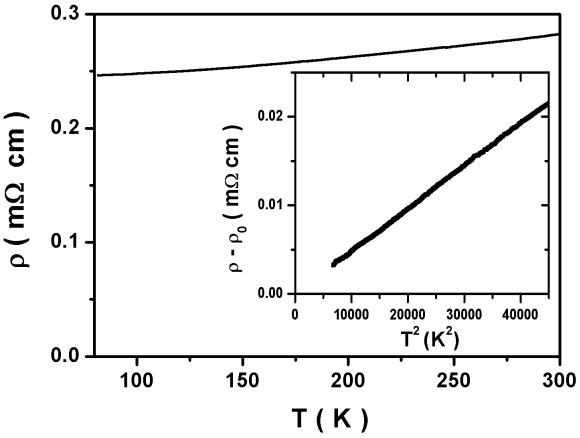

Fig. 4. X-ray diffraction pattern of the InTaO film films grown at 350 °C with $pO_2 = 0 \times 10^{-5}$ Torr.

of that InTaO film. Fig. 3 shows the XRD pattern of the InTaO film grown at 350 °C with $pO_2 = 0$ Torr. The XRD pattern of the InTaO film shows the characteristic peaks of a cubic bixbyite structure. A lattice parameter of 1.015 nm was obtained using a least-squares analysis and the measured peak positions of the (211), (222), (400), and (440) peaks. The values of the lattice parameters for the InTaO film were similar to those (1.01 – 1.02 nm) of 10 % Sn-doped In_2O_3 (ITO) films [7]. The average grain size in the InTaO film, calculated by using Scherrer's equation with the XRD line broadening method, was about 15 nm [8]. To investigate whether the scattering mechanism at the grain boundary or in the grain plays an important role in ρ of InTaO film, we calculated the mean free path (l) of carrier electrons in the InTaO film. The l in a highly degenerate electron gas at the Fermi surface is given by the equation $(3\pi^2)^{1/2}(\hbar/e^2)\rho^{-1}n^{-2/3}$ [9]. The calculated l of the carrier electrons in the InTaO film grown at $T_S = 350$ °C and $pO_2 = 0$ Torr was 5.5 nm, which was smaller than the diameter ($\sim$ 15 nm) of the polycrystalline grains in the InTaO film. Since the l value calculated above is significantly smaller than the diameter of the grain, the grain boundary scattering mechanism is not likely to play an important role.

Fig. 4 shows the temperature dependence of the resistivity of the InTaO film grown at $T_S = 350$ °C and $pO_2 = 0$ Torr. The InTaO film shows a metallic behavior with relatively a weak temperature dependence. The ρ(80 K) and the ρ(295 K) for the InTaO film were 0.25 mΩcm and 0.28 mΩcm, respectively, where ρ(80 K) and ρ(295 K) were the resistivities at 80 K and 295 K. The relative resistivity ρ(80 K)/ρ(295 K) of the film was 0.88. For temperatures below 200 K, as shown in the inset of Fig. 4, ρ fits the equation $\rho(T) = \rho_0 + AT^2$ with $\rho_0 = 0.243$ mΩcm and $B = 4.83 \times 10^{-7}$ mΩcm/K^2. Here, ρ_0 is the residual resistivity at zero temperature, and the T^2 term is possibly due to electron-electron scattering [10]. The

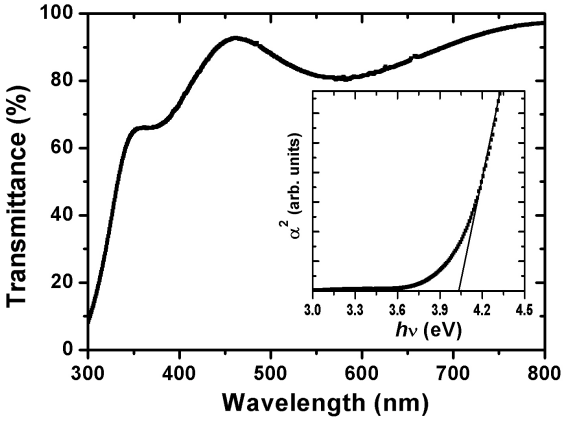

Fig. 5. Optical transmittance of the InTaO film grown at 350 °C with $pO_2 = 0$ Torr. The optical bandgap was estimated to be 4.04 eV from the x-intercept of the inset.

large value of ρ_0 was possibly due to the temperature-independent scattering mobility of neutral impurity scattering, ionized impurity scattering, and grain boundary scattering [11]. The analysis shows that grain boundary scattering contributes little to ρ_0. ρ_0 is, therefore, believed to come from neutral impurity scattering and ionized impurity scattering.

Fig. 5 shows the optical transmittance of the InTaO film grown at 350 °C with $pO_2 = 0$ Torr. The average optical transmittance for wavelengths between 400 and 700 nm of the film was above 85 %. The presence of a minimum at a wavelength of $\sim$ 580 nm in the transmittance was related to the interference of the incident light in the InTaO film. The transmittance fell sharply in the UV region due to the onset of fundamental absorption. By plotting α^2 (α : absorption coefficient) versus h (photon energy), we obtained the value of the bandgap as 4.04 eV [12].

IV. CONCLUSIONS

Highly conductive and transparent Ta-doped In_2O_3 (InTaO) films on Corning # 1737 glass substrates were deposited from a sintered $(In_{0.95}Ta_{0.05})_2O_3$ ceramic target by rf magnetron sputtering. The resistivity, the carrier density (n_H), and the Hall mobility (μ_H), of the InTaO films were in the ranges of 0.28 $\sim$ 76 mΩcm, 0.1 $\sim$ 7.4 $\times$ 10^{20} cm^{-3}, and 6.7 $\sim$ 30.1 $cm^2V^{-1}s^{-1}$, respectively. The minimum resistivity of the InTaO films was as low as 2.8 $\times$ 10^{-4} Ωcm with a n_H of 7.4 $\times$ 10^{20} cm^{-3}, a μ_H of 30.1 $cm^2V^{-1}s^{-1}$, a mean free path of 5.5 nm, and an average transmittance above 85 % for wavelengths between 400 and 700 nm. Applications of InTaO films to TFT-LCDs and photovoltaic cell are expected in further research.

ACKNOWLEDGMENTS

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