ELECTROPHYSICAL PROPERTIES OF ITO:Ga_2O_3 FILMS GROWN BY RF MAGNETRON SPUTTERING

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Abstract

In this paper, the electrophysical properties of ITO:Ga_2O_3 thin films grown by RF magnetron sputtering on glass and sapphire substrates are studied. Targets prepared by mechanical pressing of ITO and Ga_2O_3 powders are used as an evaporation source. The electrophysical characteristics as a function of optimumfilm growth parameters—the correlation between the argon and oxygen flows, the substrate temperature, and the discharge power of the magnetron—are studied.

Keywords: thin films, RF magnetron sputtering, current–voltage (I–V) characteristics

Rezumat

În această lucrare au fost studiate proprietățile electrofizice ale filmelor ITO:Ga_2O_3 obținute prin pulverizare magnetron RF pe substraturi de sticlă și safir. Ca sursă de evaporare au servit țintele confecționate prin presare mecanică a pulberelor de ITO și Ga_2O_3. Au fos tcercetate caracteristicile electrofizice în dependență de parametrii optimi de obținere a filmelor, cum ar fi,~~:~~coraportul dintre fluxul de argon și oxigen, temperature substratului, puterea de descărcare a magnetronului.

Cuvinte cheie: filme subțiri, pulverizare magnetron RF, caracteristica voltamperică I-V

1.

2. Introduction

Indium tin oxide (ITO), which is a wide-bandgap semiconductor exhibiting chemical and thermal stability and high optical transparency at wavelengths of $\lambda > 300$ nm, is a promising material for the development of photoreceptors and functional sensors in the near-ultraviolet region (185 nm) [1, 2]. Indium tin oxide films are commonly used in semiconductor devices [3], such as liquid crystal displays and light emitters [4]. Nanostructured ITO has many new applications due to its unique surface and quantum effects; it can be applied in photovoltaic cells [5] and gas sensors [6]. Indium tin oxide films are also commonly used in other commercial applications, although many other transparent conductive oxide films have been developed to replace ITO [7]. Since ITO films are commercially available, the research of these films is focused on the fields of their application or preparation techniques. The following methods are most commonly used to grow ITO thin films: electron beam deposition, ionic or thermal evaporation, laser evaporation, cathode or high-frequency (HF) sputtering in an electric field, and magnetron sputtering of a target. In the case of using magnetron sputtering method, the parameters of the resulting films largely depend on the deposition mode: the total gas pressure in the chamber, the partial pressure of oxygen, the substrate temperature, the electrode configuration, and the deposition rate. Adhering to the one-to-one relationship between the deposition rate and the partial oxygen pressure in the chamber, it is possible to grow ITO:Ga_2O_3 films exhibiting optimum conductivity and transparency by varying the oxygen and argon content in the working atmosphere in a fairly wide range. For example, ITO:Ga_2O_3 films were grown by RF magnetron sputtering. The bandwidth of the resulting film can achieve values in a range of 3.6 eV for ITO to 4.6 eV for β-Ga_2O_3 at electrical and optical parameters comparable to the parameters of ITO.

3. Experimental

The study was conducted on a setup comprising a magnetron placed at the middle of the working chamber and a water-cooled target. The sputtering target was made of a mechanical mixture of indium, tin, and gallium oxide powders taken in appropriate proportions: ITO (90% In_2O_3 and 10% SnO_2) and the ITO:Ga_2O_3 compound (95% ITO and 5% Ga_2O_3) in weight units. The diameter of the target made by mechanical pressing was 50 mm. A power supply operating at an RF frequency of 13.6 MHz was used as the magnetron power source. The gas flow was fed by a two-channel system based on gas flow regulators. The total pressure inside the working chamber during condensation was 7.4×10^{-3} mBar. The percentage ratio of oxygen and argon pressures was varied accordingly from 0 : 100 to 10 : 90 counted in mln/min. The film deposition rate was varied in a range of 0.5–4.0 Å/s. The substrate was arranged parallel above the target at a distance of 80 mm; it was equipped with a rotation mechanism centered relative to the target. The rotation of the substrate during condensation provides the formation of homogeneous films on the entire surface of the substrate. The sapphire and glass substrates subjected to sputtering were preventive degreased, washed in running distilled water, and then dried by centrifugation. The substrate temperature during film condensation was varied from 50 to 500°C. The thickness of the thin films during condensation was monitored using a standardized-ratio quartz sensor. Indium point ohmic contacts were deposited by high-vacuum thermal evaporation. Conductive and transparent layers were grown in the above gas flow pressure and film condensation rate ranges.

4. Results and Discussion

The percentage of indium, tin, and gallium oxide in ITO:Ga_2O_3 films has a significant—yet not decisive—effect on the conductivity and transparency of the films [8]. The homogeneity over the entire surface of the support and the deposition of the films correlate with the growth conditions, while the oxygen composition determines the physical properties. Earlier, it was reported [9–11] that, in thin layer deposition methods, the stoichiometry of oxide films is determined by the partial pressure of oxygen (P_{O2}) during growth. Upon the ignition of a plasma inside the chamber, the oxide undergoes dissociation into separate components due to the RF applied to the system with the appearance of oxygen in the atomic (O) and molecular form (O_2), which significantly affects the film growth on the substrate surface. During RF magnetron sputtering, the In, Sn, and Ga species deposited on the substrate surface arrive directly from the target, while the oxygen species arise from both the oxygen pressure on the target and the oxygen pressure in the chamber. It was found that the ratio of In, Sn, Ga, and O species fluxes onto the substrate depends on the nature of the gas during sputtering, as reported for ITO films [12].

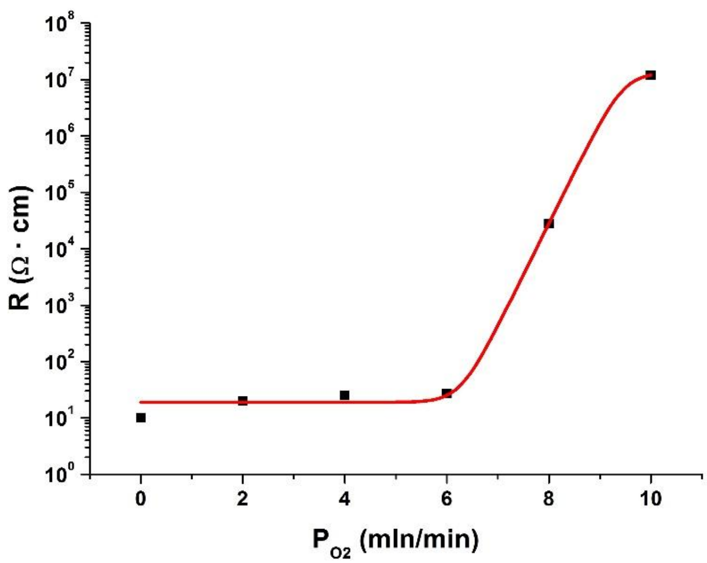

Fig. 1. Dependence of the resistivity of ITO:Ga_2O_3 films on the partial pressure of oxygen in the working chamber during film condensation.

Figure 1 shows the fundamental shape of the dependence of the resistivity of ITO:Ga_2O_3 films on the partial pressure of oxygen (P_{O2}) at a constant argon flow rate (50 mln/min), a constant deposition rate (~1 Å/s), and a constant substrate temperature (100°C). It is evident from the plot that the curve exhibits a tendency to increasing with an increase in oxygen flow rate. The minimum film resistivity corresponds to the range from the point of the total absence of an oxygen flow to a partial flow rate of about 6 mln/min. A further increase in the oxygen flow rate, in turn, leads to a substantial increase in the resistivity of the films. A low oxygen deficiency in the films can play an important role in their physical properties [13].

An increase in the oxygen flow rate leads to the formation of dense smooth stoichiometric films observed under an electron microscope. Figure 2 shows a plot of the current–voltage characteristics of ITO:Ga_2O_3 films grown at substrate temperatures of 50–450°C. The minimum temperature is limited to the growth conditions with the heating of the substrate from 20 to 50°Cduring the deposition of the layers. Other parameters, such as a growth rate of V = 0.5 Å/s, a magnetron discharge power of P = 140 W, an argon flow rate of F = 50 mln/min, an oxygen flow rate of F = 6 mln/min were maintained constant during film growth. The current–voltage characteristics are represented by straight lines symmetrical with respect to the abscissa axis. The measurements showed an ohmic behavior of the contacts, a decrease in the film resistance in a substrate temperature range of 50–150°C, and an increase in the film resistance with a further increase in the substrate temperature.

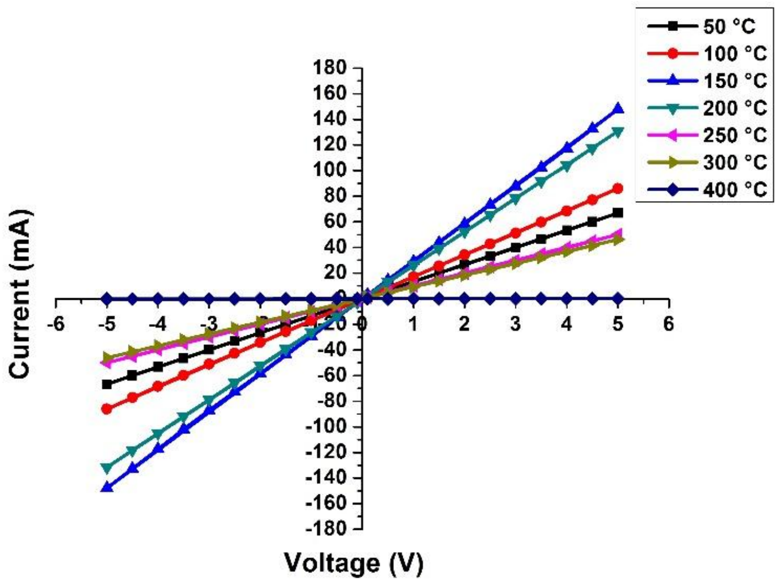

Fig. 2. Current–voltage characteristics of ITO:Ga_2O_3 films grown at different substrate temperatures.

Figure 3 shows the dependence of the resistivity of the films on the substrate temperature during condensation. It is evident that, in a temperature range of 150–200°C, the grown films have a minimum resistivity in a range of 30–40 Ω.

In a temperature range of 50–150°C, the resistivity of the films is higher; this finding isassociated with the fact that, in this temperature range, the grown films are amorphous and their conductivity decreases, as in the case of ITO films described previously [8, 14]. The significant increase in the resistivity of the films at temperatures above 200°C can be attributed to the re-evaporation of the more volatile component from the substrate surface during condensation; that is, the composition of the material condensed on the substrate changes, which, in turn, leads to a change in the conductivity of the film.

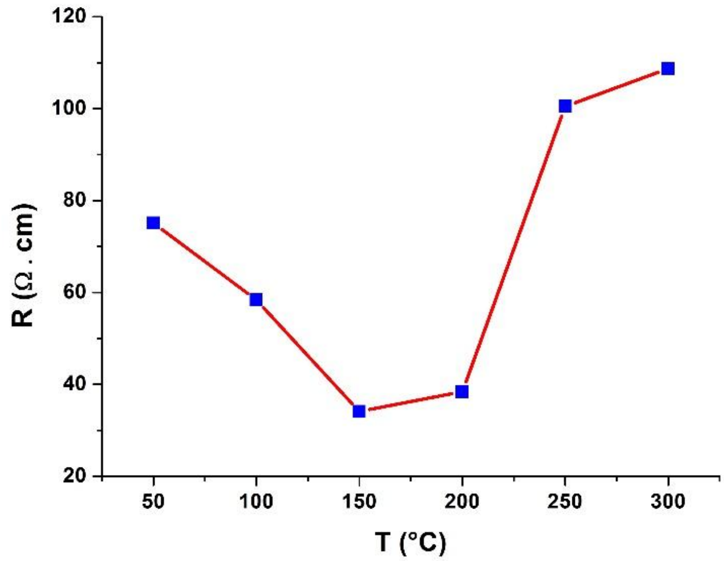

Fig. 3. Dependence of the resistivity of the films on the substrate temperature during condensation.

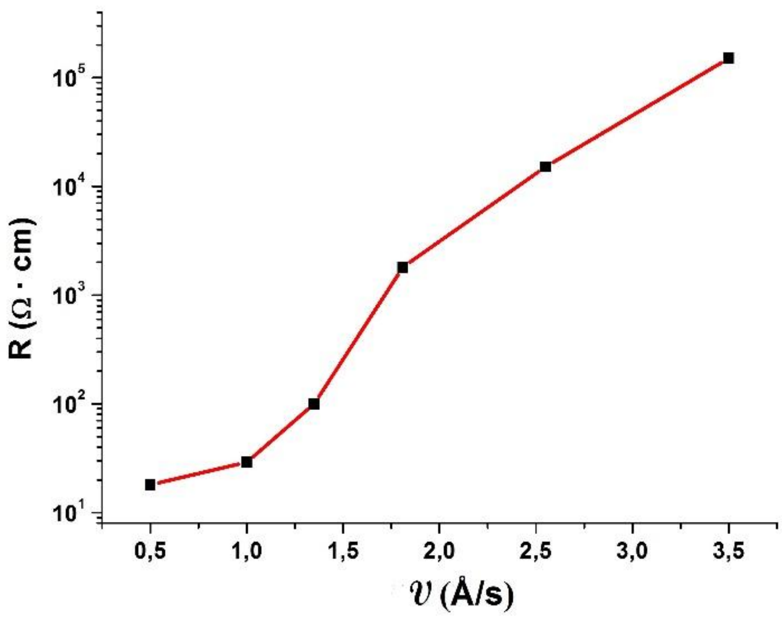

Fig. 4. Dependence of the resistivity of ITO:Ga_2O_3 films on the deposition rate at a fixed partial pressure of oxygen (6 mln/min).

Figure 4 shows the dependence of the resistivity of the ITO:Ga_2O_3 films on the deposition rate, where the discharge power of the magnetron is varied from 80 to 150 W, the substrate temperature of 150°C is maintained constant, and the rest of the above-mentioned parameters are kept constant. It is evident that, at low condensation rates of 0.5–1 Å/s, the resistivity of the films has a minimum value; with an increase in the deposition rate, the resistivity of the films increases, because the migration of atoms on the substrate surface and association in larger clusters is limited to the high condensation rate.

5. Conclusions

Since the ITO:Ga_2O_3 compound films exhibit conductivity and transparency comparable to those of ITO films and it is possible to vary the band width from 3.72 eV for ITO to 4.69 eV for β-Ga_2O_3, this compound appears attractive in terms of both theoretical aspects and practical applications. By varying the oxygen flow rate in the working chamber, it is possible to grow both conductive films and films exhibiting dielectric properties, which can be commonly used in various electrical and optoelectronic devices.

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