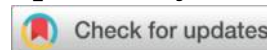




Characterization of Aluminum-Doped Zinc Oxide Thin Films Deposited by the GLAD Technique (Glancing Angle Deposition)



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Abstract

Aluminum-doped zinc oxide (ZnO:Al) has attracted significant attention for its electrical and optical performance, high transparency, and low fabrication cost. ZnO is a wide band gap (~3.3 eV) semiconductor; aluminum doping increases electrical conductivity while preserving optical transmittance. In this study, thin films of ZnO with 2% Al were deposited by glancing-angle deposition (GLAD) and their structural, optical, and electrical properties were examined. The films show a wurtzite crystal structure, high visible-range transparency, and a marked conductivity enhancement attributable to Al incorporation. These results indicate that ZnO:Al thin films are promising for optoelectronic applications, especially as transparent conducting electrodes in devices such as solar cells and displays.

Keywords: Aluminum-doped ZnO; thin films; GLAD; structural characterization; optical properties.

Introduction

Zinc oxide (ZnO) has become one of the most extensively studied semiconductor materials due to its physical, chemical, and optical properties [8–10]. It is non-toxic, chemically stable, transparent in the visible region, and has a wide band gap, which makes it suitable for numerous technological applications. Owing to its wurtzite crystal structure and intrinsic stability, ZnO has been employed in photovoltaic cells, gas sensors, light-emitting diodes (LEDs), field-effect transistors, ultraviolet (UV) lasers, and piezoelectric devices [4,7,8,11–17].

However, a key drawback of pure ZnO is its low intrinsic electrical conductivity, which can limit its direct use in devices requiring efficient charge transport [7,8]. To address this limitation, various doping strategies have been explored to introduce controlled impurities into the ZnO lattice. Among the most effective dopants, aluminum (Al) is particularly attractive [1,2,4,7,8]. Aluminum-doped zinc oxide (AZO) combines good electrical conductivity with high optical transparency, making it a promising alternative to indium tin oxide (ITO), which is widely used but costly and scarce [2,4,8].

Aluminum doping modifies the electronic structure of ZnO by increasing the carrier concentration without significantly affecting transparency in the visible range [1,4,7]. This balance between transparency and conductivity makes ZnO:Al an excellent candidate for transparent conducting oxide (TCO) thin films, which are essential in solar cells, flat-panel displays, and other optoelectronic devices [2,4,8,9]. Nevertheless, composition control remains crucial: too little Al only partially enhances conductivity, whereas excessive doping can degrade crystallinity and reduce optical transmittance [1,7].

Optically, ZnO exhibits a direct band gap of approximately 3.37 eV at room temperature, corresponding to near-UV emission [5,6,8]. Its high exciton binding energy (~60 meV) enables efficient excitonic luminescence even at room temperature, unlike some other semiconductors such as gallium nitride (GaN) [5,6,11]. ZnO can be synthesized in multiple morphologies—nanowires, nanoparticles, thin films, and multilayers—offering broad opportunities for nanotechnology [3,4,12,20].

Chemically, ZnO is stable and can be etched in acidic or basic solutions at low temperatures, which facilitates microfabrication [20–22]. It also exhibits high thermal conductivity for heat dissipation, along with radiation tolerance and mechanical robustness suitable for harsh environments, including space applications [6,9,10].

Electronically, ZnO is typically n-type, with free electrons originating from oxygen vacancies or interstitial zinc [8,9]. Electron mobility in ZnO thin films commonly ranges from 20 to 30 cm²/V·s and can be improved through optimized doping and controlled deposition conditions [7,8]. In particular, Al incorporation increases electron concentration and thus conductivity while largely preserving transparency [1,4,7,8].

In this context, the present work focuses on the fabrication of aluminum-doped ZnO thin films using an appropriate deposition technique and on the assessment of how doping influences their structural, electrical, and optical properties. The aim is to balance electrical conductivity and optical transparency, and to compare the experimental results with prior reports to evaluate performance [1–4,7–9]. In summary, ZnO and its doped derivatives represent a strategic materials family for modern microelectronics and optoelectronics, combining chemical stability, a wide band gap, tunable conductivity, high transparency, and compatibility with diverse substrates [2,4,8–10]. This study contributes to understanding ZnO:Al as a next-generation transparent conducting material [1–4,7–9].

Experimental Methods

I.1. Objective

The main objective of the present work is to characterize the electrical and optical properties of a wide band-gap semiconductor, namely aluminum-doped zinc oxide (ZnO:Al).

Substrate Preparation

The substrates used for the deposition of the studied material were prepared by cutting soda-lime glass slides using a dicing-cutting saw.

I.2. Substrate Cleaning

Before depositing aluminum-doped ZnO thin films, the substrates were cleaned according to the following procedure:

- Cleaning with a detergent in an ultrasonic bath;
- Rinsing with deionized water (DI water);
- Cleaning with acetone in an ultrasonic bath;
- Cleaning with isopropyl alcohol (IPA) in an ultrasonic bath;
- Rinsing again with DI water;
- Drying under a nitrogen (N₂) jet.

Deposition of ZnO:Al Thin Films

The deposition of aluminum-doped ZnO thin films was carried out using a sputtering system,.

Experimental Conditions

- Base pressure: 4×10^{-7} mbar
- Deposition pressure: 2.5×10^{-6} mbar
- Power: **90 W**
- Deposition rate: **1 Å/s**
- Film thickness: **200 nm**
- Material: **Aluminum-doped ZnO (ZnO:Al)**

I.3. Physicochemical Characterization

After depositing the thin film, the phases present in the deposited layer were identified using an **Equinox 3000 diffractometer**.

The diffractogram obtained **before annealing** (Figure.1) shows only a broad hump between $2\theta = 20^\circ$ and $2\theta = 40^\circ$, which corresponds to the amorphous structure of the glass substrate. This indicates that the deposited layer also exhibits an amorphous structure.

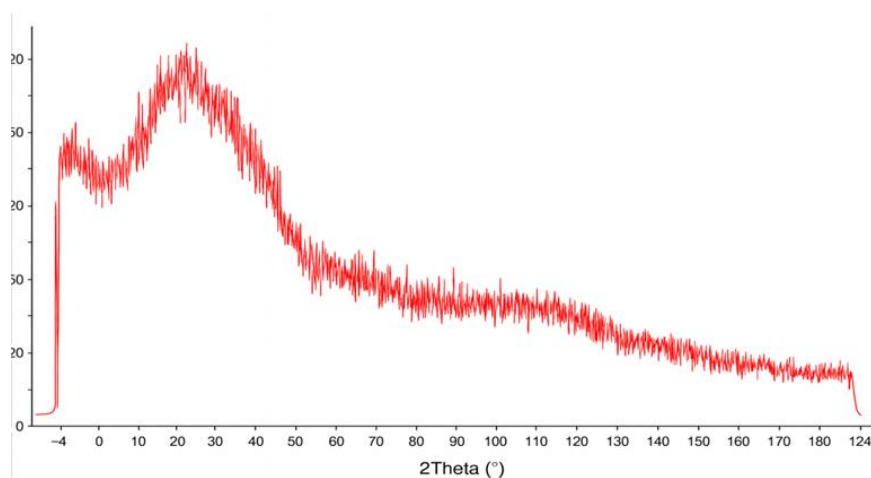


Figure 1 : Diffractometer of the aluminum-doped ZnO layer before annealing

The diffractogram obtained **after annealing at 300°C for 45 minutes** **Figure 2** reveals a diffraction peak around **34.5°**, indicating that the aluminum-doped ZnO layer has crystallized along the **(002) orientation**. No diffraction peak related to aluminum was observed, which can be attributed to the low doping concentration (2 wt%).

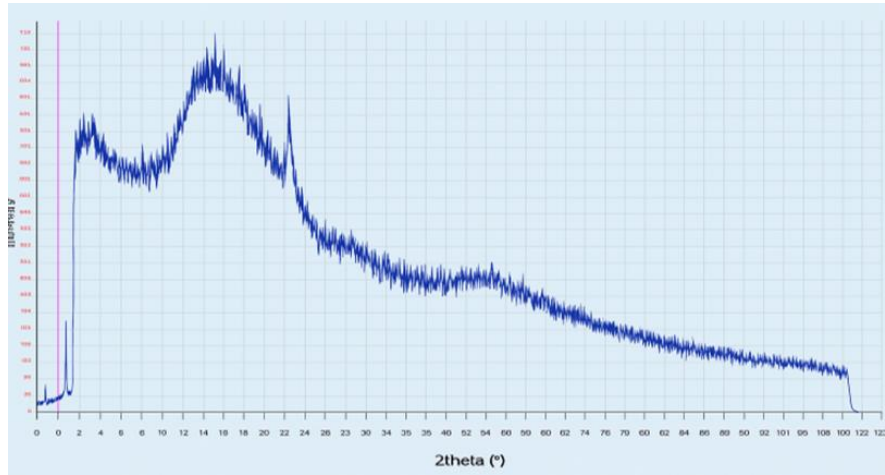


Figure 2 : Diffractometer of the aluminum-doped ZnO layer after annealing

I.4. Optical Characterization

The transmittance, reflectance, and absorbance spectra were recorded using an **F10-RT-UV spectrophotometer**.

I I.1. Transmittance Spectra

The curves in **Figure 3** show the variation of transmittance (%) as a function of wavelength (nm). It can be observed that the transmittance is very low for short wavelengths (below approximately **350 nm**) and then increases rapidly to reach a plateau around **400 nm**, where it stabilizes at about **80%**. This behavior suggests that the studied material is nearly opaque in the ultraviolet region but becomes highly transparent in the visible and near-infrared regions.

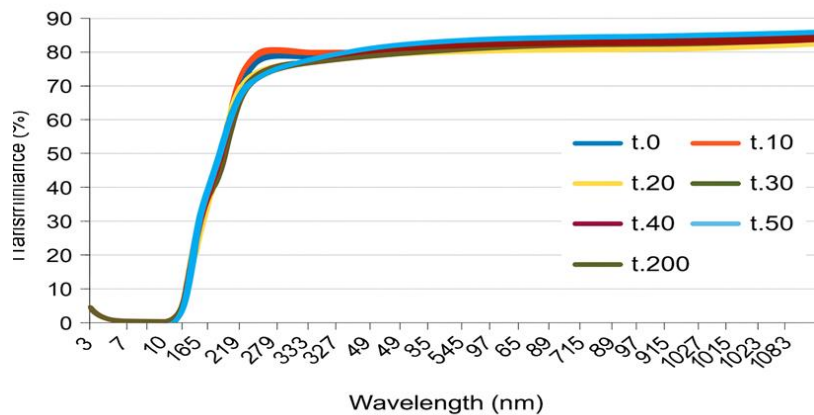


Figure 3: Transmittance spectrum of ZnO/Al layers

Effect of Inclination on Transmittance

At short wavelengths, the transmittance remains very low for all incidence angles. Increasing the inclination shifts the onset of the rapid transmittance rise toward longer wavelengths, corresponding to a “red shift” of the absorption edge. At longer wavelengths (visible/IR range), the maximum transmittance slightly decreases with increasing inclination.

Interpretation:

This behavior may be attributed to the fact that, as the inclination increases, the layer becomes more diffuse, leading to a reduction in overall transmittance. The shift of the absorption edge indicates a modification in the structure or the optical density of the film.

I I.2. Reflectance Spectra

The curves in **Figure 4** show the variation of reflectance (%) as a function of wavelength (nm). It can be observed that the reflectance increases rapidly to reach a maximum of approximately 27% around 390 nm, then gradually decreases with increasing wavelength, reaching about 10% at 1100 nm. This behavior indicates that the material exhibits higher reflectivity in the ultraviolet and the early visible regions, and becomes less reflective in the infrared range.

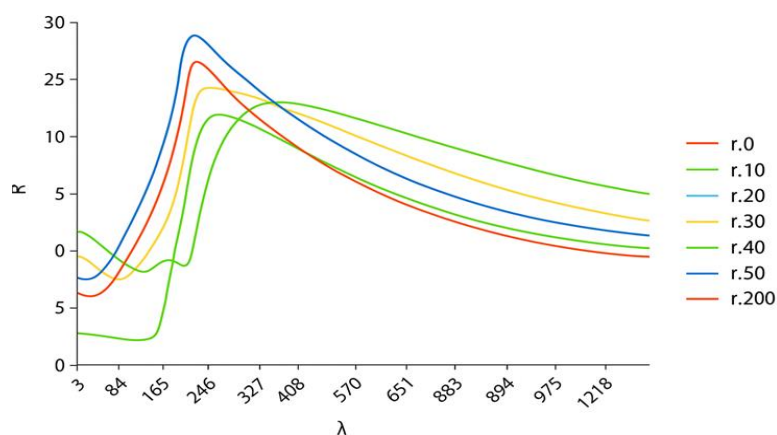


Figure 4 : Reflectance spectra of ZnO/Al

Effect of the inclination on the reflectance

At short wavelengths (UV, < 400 nm), the reflectance exhibits a pronounced peak for all inclination angles. As the inclination increases, the maximum reflectance value slightly increases, and the peak shifts toward longer wavelengths. For longer wavelengths (> 400 nm), the reflectance gradually decreases but remains higher for the films deposited at larger inclination angles.

Interpretation:

The increase in inclination during GLAD deposition makes the film more porous and rough, which enhances light reflection, especially in the UV region. This also alters the effective refractive index, explaining the broadening and shift of the reflectance peak.

II.3. Absorbance spectra:

The curves in Fig.5 show the absorbance (%) as a function of wavelength (nm). A high absorbance (approximately 90%) is observed at short wavelengths, followed by a sharp decrease around 400 nm. Beyond this region, the absorbance remains very low (below 10%) and relatively constant up to 1100 nm. This behavior suggests that the sample primarily absorbs in the ultraviolet region and transmits most of the light in the visible and near-infrared ranges.

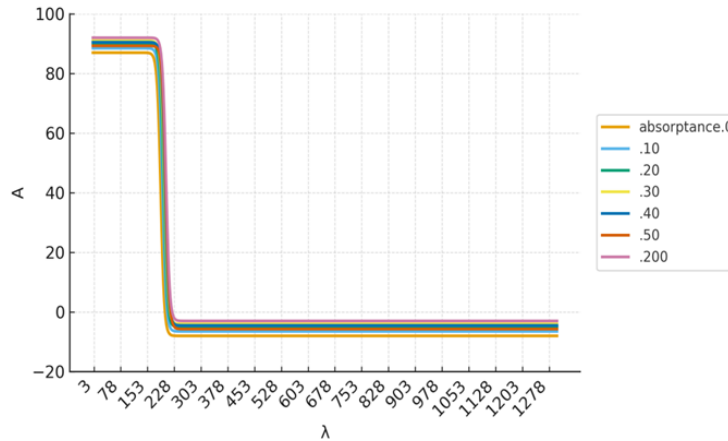


Figure 5 : Absorbance spectra of ZnO/Al layers

Effect of inclination on absorbance

At short wavelengths (UV, < 400 nm), the absorbance is high for all inclination angles, followed by a sharp drop. As the inclination increases (darker curves), the maximum absorbance slightly increases, and the curve broadens somewhat toward longer wavelengths. For longer wavelengths (> 400 nm), the absorbance remains very low regardless of the inclination angle.

Interpretation:

The increase in the inclination angle during GLAD deposition leads to a slight enhancement in absorbance, particularly in the UV region. This behavior can be attributed to an increase in porosity, surface roughness, or effective optical thickness, which promotes light absorption.

II.4. Global analysis of absorbance, reflectance, and transmittance spectra:

The graph Figure 6 illustrates the evolution of absorbance, reflectance, and transmittance as a function of wavelength. It is observed that, at short wavelengths (below approximately 350 nm), the absorbance is very high while the transmittance is nearly zero, indicating that the material strongly absorbs in the ultraviolet region. Starting from around 400 nm, the transmittance increases sharply and reaches a plateau around 80%, showing that the material becomes highly transparent in the visible region and up to 1100 nm. The reflectance remains low and relatively stable throughout the investigated spectral range. This behavior demonstrates that the material is opaque in the ultraviolet but highly transparent in the visible and near-infrared regions.

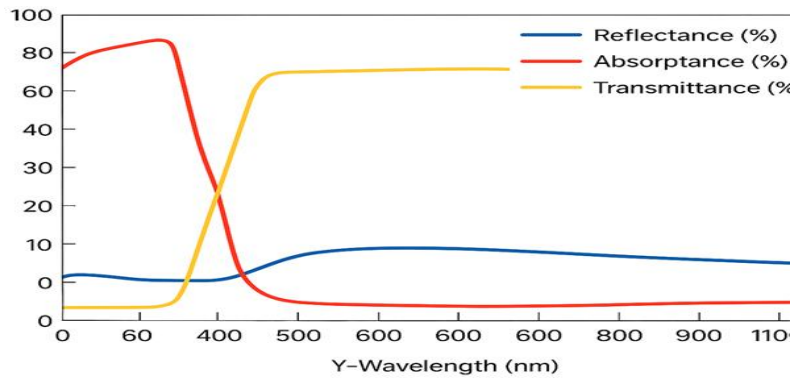


Figure 6 : Absorbance, reflectance, and transmittance spectra

The overall analysis of the absorbance, reflectance, and transmittance spectra allows the following conclusions to be drawn:

- **Low inclination:** The deposited films are denser and more homogeneous, resulting in higher transmittance and lower absorbance.
- **High inclination (GLAD):** The deposited films are more porous and rough, with inclined nanostructures that enhance light scattering and absorption, thereby reducing transmittance.
- **Application:** From these observations, it can be inferred that controlling the inclination angle allows tuning of the optical properties of thin films for applications in optoelectronics, sensors, and UV filters.

II.5. Electrical characterization

The prepared structure used for electrical characterization consists of a thin ZnO:Al film with a thickness of 200 nm deposited on a glass substrate. The aluminum doping concentration is 2 wt%.

The selected technique for electrical characterization is the **Van der Pauw method**, an electrical measurement technique used to determine the resistivity and Hall coefficient of thin samples with arbitrary shape but uniform thickness. This method is particularly suitable for two-dimensional materials and thin films.

In our case **Figure 7a and 7b** we focused on the variation of resistivity (ρ) and conductivity (σ) as a function of the deposition angle (θ). A clear inverse relationship between these two parameters was observed: when the resistivity reaches its minimum, the conductivity simultaneously reaches its maximum at the same angle. Conversely, for other angles, higher resistivity corresponds to lower conductivity, illustrating the physical relationship $\sigma = 1/\rho$. The variations are non-linear, indicating a pronounced angular dependence of the electrical properties of the studied material.

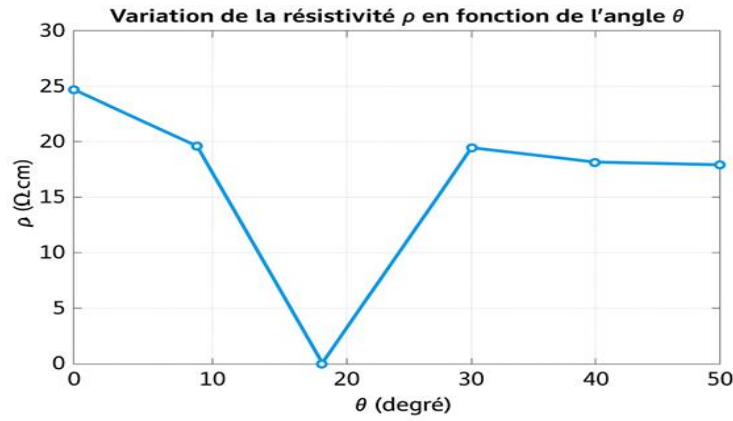


Figure 7 a : Variation of the resistivity and conductivity with respect to the angle.

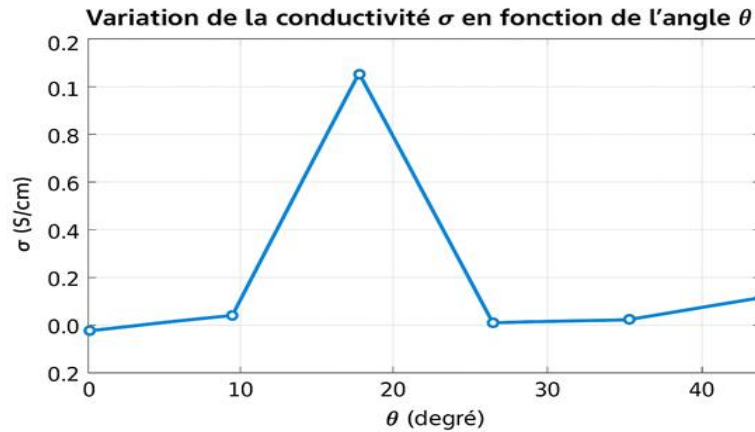


Figure 7 b : Variation of the resistivity and conductivity with respect to the angle.

Conclusion

In this work, we studied and characterized aluminum-doped zinc oxide (AZO) thin films prepared using the **GLAD (Glancing Angle Deposition)** technique. Structural, optical, and electrical analyses of these films revealed the influence of doping and deposition conditions on the final properties of the material.

The results show that the GLAD technique enables the fabrication of thin films with nanostructured morphology and good uniformity. Optical measurements reveal strong absorbance in the ultraviolet region, while the transmittance reaches high values (above 80%) in the visible and near-infrared regions, confirming the excellent transparency of the AZO layers. The reflectance remains low over the entire investigated spectrum, which is advantageous for optoelectronic applications.

Thus, aluminum-doped ZnO thin films obtained by GLAD exhibit particularly promising properties for applications such as **transparent electrodes, solar cells, and advanced optical devices**. This study highlights the potential of the GLAD technique for producing high-performance functional materials and opens the way for future optimization tailored to the specific requirements of targeted applications.

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