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Structural, Optical and Electrical properties of Al doped ZnO thin films for transparent conducting oxide applications

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Abstract
Aluminum-doped zinc oxide (AZO) thin films were successfully deposited on amorphous fused silica substrates via pulsed laser deposition (PLD) technique. This study systematically investigates the influence of key deposition parameters, including substrate temperature, oxygen partial pressure, laser fluence, with aluminum doping concentration of 1 wt% on the structural, electrical, and optical properties of the resulting AZO films. The electrical resistivity of the films was characterized using a four-point probe method, while their optical transmittance was assessed across the UV-Visible-NIR spectrum. Notably, the films with 1 wt% Al exhibited a favorable balance of properties, achieving a high visible transmittance of approximately 79% and a low resistivity of 7.01 × 10^{-3} Ω·cm. Optimal film performance was observed for samples deposited at elevated substrate temperatures (up to 500°C) and under reduced chamber pressures (10^{-2} – 10^{-3} Pa). Additionally, a clear trend of improved electrical conductivity and enhanced transparency was identified with increasing aluminum content. The results underscore the potential of carefully tailored AZO thin films as promising candidates for transparent conducting oxide (TCO) applications.
Keywords: *Al-doped ZnO, Pulsed Laser Deposition, Transparent Conducting Oxide, Optical Properties, Electrical Properties.*

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I. Introduction

Transparent conducting oxides (TCOs) are an essential class of materials that exhibit the rare combination of high optical transparency in the visible spectrum and excellent electrical conductivity. Their unique properties have made them indispensable in a broad range of optoelectronic applications, including flat panel displays, touch screens, photovoltaic cells, organic light-emitting diodes (OLEDs), low-emissivity windows, and smart windows [1]. Among the various TCO materials, indium tin oxide (ITO) has traditionally been the most widely used due to its superior electrical and optical performance. However, the limited availability, high cost, and toxicity of indium have driven the scientific community to explore alternative TCO materials that are more sustainable, cost-effective, and environmentally benign [2].

Zinc oxide (ZnO), a II-VI semiconductor with a direct wide band gap (~3.37 eV at room temperature), has emerged as a promising candidate to replace conventional TCOs. ZnO inherently exhibits high transmittance in the visible range, excellent chemical stability, and compatibility with a variety of substrates, including flexible and amorphous materials [3]. Furthermore, its earth abundance, non-toxicity, and ease of fabrication make ZnO-based materials an attractive alternative to ITO [4]. However, pristine ZnO suffers from relatively low electrical conductivity due to its moderate carrier concentration. Therefore, controlled doping is essential to enhance its electrical performance while preserving its optical transparency.

Among various dopants investigated, aluminum (Al) has proven to be highly effective in improving the electrical conductivity of ZnO by substituting Zn^{2+} ions with Al^{3+}, introducing additional free electrons into the conduction band. Aluminum-doped zinc oxide (AZO) films have demonstrated remarkable potential as TCOs due to their enhanced carrier concentration, stable chemical properties, and relatively low processing costs [4]. Recent studies have highlighted that the electrical and optical properties of AZO films can be finely tuned by optimizing parameters such as Al doping concentration, deposition method, substrate type, and post-deposition annealing [5]. These improvements have positioned AZO as a viable, scalable, and environmentally friendly candidate for next-generation optoelectronic devices.

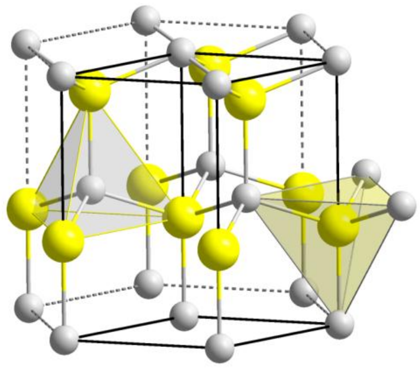

Fig. 1: Hexagonal Wurtzite structure of ZnO

Furthermore, the choice of substrate plays a critical role in determining the crystallinity, microstructure, and subsequent optoelectronic properties of AZO films. While AZO films have been successfully deposited on single crystalline substrates like sapphire, much attention has recently shifted toward amorphous substrates such as glass and polymeric materials, driven by the growing demand for flexible and lightweight electronics [6]. Understanding the interplay between substrate characteristics, doping concentration, and deposition parameters is crucial for tailoring the functional properties of AZO films to meet specific application requirements.

In this study, we systematically investigate the structural, optical, and electrical properties of 1 wt% doped Al-doped ZnO thin films deposited using pulsed laser deposition on amorphous fused silica substrates. Employing a controlled doping strategy and optimized deposition parameters, we aim to achieve high transparency and conductivity suitable for transparent electrode applications. The present work contributes to the research on sustainable TCO alternatives by providing insights into the influence of Al doping on the performance of ZnO films, paving the way for their practical integration into a wide spectrum of optoelectronic devices.

Experimental details

Aluminum-doped zinc oxide (AZO) thin films were deposited with a fixed aluminum doping concentration of 1 wt%. The films were synthesized using the Pulsed Laser Deposition (PLD) technique. To systematically investigate the influence of deposition parameters on the structural, optical, and electrical properties of the AZO films, key parameters such as laser fluence, substrate temperature, and working oxygen pressure were deliberately varied across different deposition runs. This approach enabled the identification of optimal conditions for achieving desired film characteristics.

The structural characterization is recorded using GI-XRD technique. The optical transmittance spectra of the deposited films were measured in the wavelength range of 190 to 2500 nm using a UV-VIS-NIR spectrophotometer (Jasco V-570). These measurements provided insights into the transparency and band gap behavior of the films under different deposition conditions. Electrical characterization was performed by recording the current-voltage (I-V) characteristics of each film using a standard four-point probe method integrated with a semiconductor device analyzer (Agilent Technologies B1500A). From the linear I-V curves, the sheet resistance of each film was determined, and corresponding resistivity values were calculated. Additionally, the thickness of the films was measured using a stylus profilometer to ensure uniformity and assess its correlation with optical and electrical properties.

The detailed deposition parameters corresponding to each sample, including laser fluence, substrate temperature, and working pressure, are summarized in Table 1 for reference.

Table 1: Deposition conditions of Al doped ZnO thin films using Pulsed laser deposition technique

Sample	Laser Fluence (J/cm^2)	Number of Counts	Repetition Rate (Hz)	Working Pressure (mbar)	Substrate Temperature (°C)
ZnO-01	8.93	10000	10	0.015	500
ZnO-02	10.12	4000	10	0.005	500
ZnO-03	10.12	6000	10	0.001	500
ZnO-04	10.12	6000	10	0.001	500
ZnO-05	10.12	10000	10	0.1	500

II. Results and discussion

Phase Identification and Crystalline Structure:

The X-ray diffraction (XRD) patterns of the Al-doped ZnO (AZO) thin films deposited under different pulsed laser deposition conditions reveal the polycrystalline nature of the films with a hexagonal wurtzite structure, which is typical for ZnO-based materials. The diffraction peaks correspond predominantly to ZnO phases, with no observable secondary phases related to aluminum or its oxides, indicating successful incorporation of Al into the ZnO lattice without altering the primary phase. The prominent diffraction peaks around 2θ ≈ 34.5° and 63° correspond to the (002) and (103) planes of ZnO, respectively, confirming the wurtzite structure (JCPDS card no. 36-1451) [7].

A detailed comparison of the films shows variations in preferential orientation depending on deposition conditions. The ZnO-03, 04 and 05 sample exhibits a distinct (002) peak, signifying strong c-axis-oriented growth perpendicular to the substrate surface, which is highly favorable for transparent conducting oxide (TCO) applications due to enhanced carrier mobility along this direction. Conversely, other samples such as ZnO-01 and ZnO-02 show a dominant (103) peak, suggesting growth in an off-axis direction, likely influenced by the varied substrate temperatures, oxygen pressures, and laser fluences used during deposition. The emergence of different growth orientations indicates the critical role of process parameters in tailoring film crystallinity and texture, impacting their electrical and optical properties.

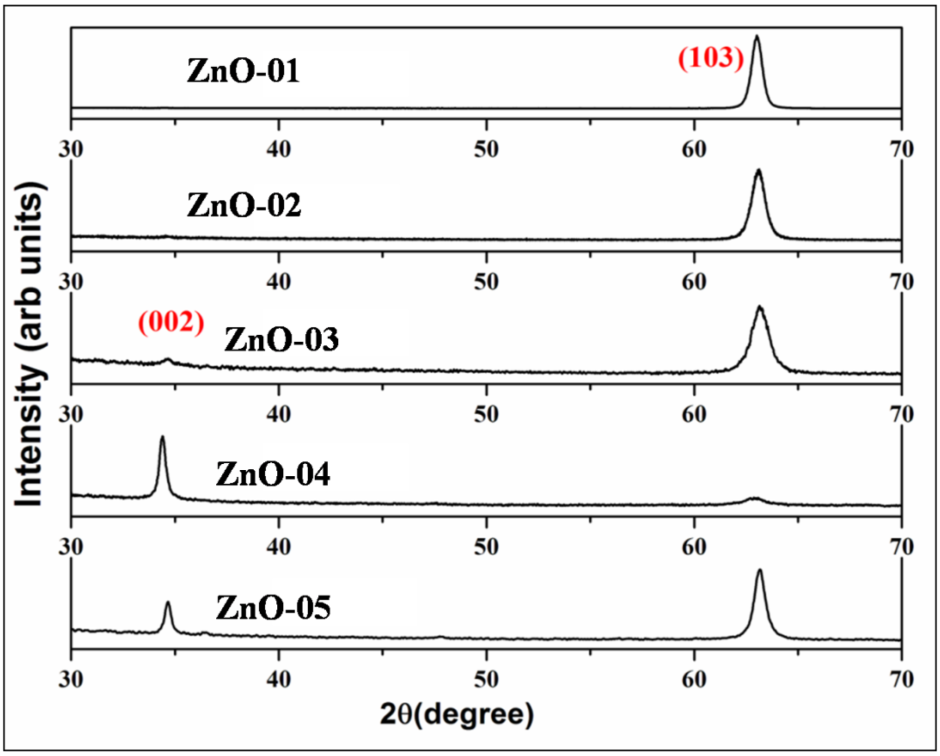

Fig. 2: X-ray diffraction spectra of Al (1 wt%) doped ZnO thin films at different deposition conditions as mentioned in table 1.

The sharp and intense peaks, especially in ZnO-03 and 04, reflect improved crystallinity with larger crystallite sizes. The crystallite size can be estimated using the Debye-Scherrer formula, and preliminary inspection suggests that ZnO-03 (with (002) peak prominence) likely possesses relatively larger crystallites compared to others. Slight peak shifts (though not explicitly visible in the image) and the absence of secondary phases imply successful Al incorporation, which may lead to minor changes in lattice parameters due to the substitution of Zn^{2+} (ionic radius ~0.74 Å) with smaller Al^{3+} ions (ionic radius ~0.53 Å). This substitution typically results in a slight contraction of the lattice, enhancing the electrical conductivity. Overall, the XRD analysis confirms the high phase purity, preferred orientation, and structural integrity of the deposited AZO films, essential for optimizing their performance in TCO applications.

Microstructure (FESEM):

The scanning electron microscopy (SEM) images of the Al-doped ZnO (AZO) thin films reveal a compact, granular microstructure characteristic of nanocrystalline thin films. The low-magnification image (left) demonstrates uniform surface coverage with densely packed grains, while the high-magnification image (right) highlights the presence of well-defined nanostructured grains, including some rod-like features and a few larger agglomerated clusters. Such microstructural features are typical in AZO films grown via pulsed laser deposition (PLD) and are known to influence both the electrical and optical performance of the films. The fine granular morphology is beneficial for achieving high optical transparency due to minimal light scattering, whereas the compact nature aids in providing continuous conductive pathways, reducing resistivity, as reported in similar studies [8]. Additionally, the observed nanoscale grains and occasional larger clusters might be attributed to localized variations in adatom mobility or growth kinetics during deposition. These microstructural characteristics underscore the importance of optimizing deposition parameters to tailor the film's morphology for enhanced transparent conducting oxide (TCO) applications.

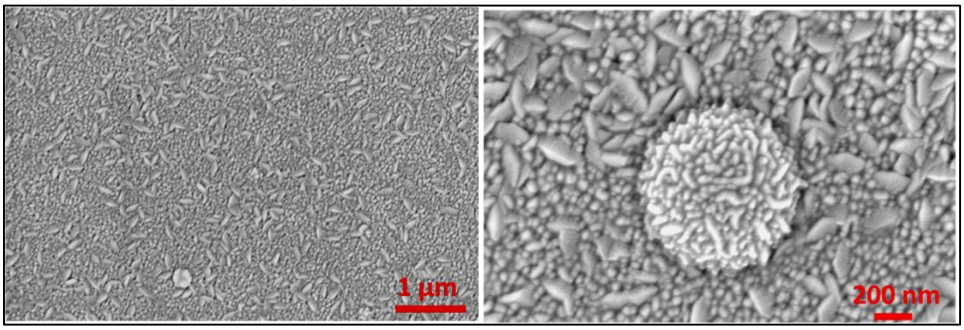

Fig. 3: Microstructure images of Al (1 wt%) doped ZnO thin films deposited at annealing temperature of 500ºC.

Optical Properties (Transmittance spectra):

The UV-Vis-NIR transmittance spectra presented in the figure reveal the optical behavior of AZO thin films deposited under five different PLD conditions, labeled as in Table-1. Across all samples, a sharp absorption edge is observed around ~370 nm, which corresponds to the intrinsic band gap absorption of ZnO, indicating the preservation of the wide band gap (~3.3-3.4 eV) characteristic of ZnO. Beyond this edge, all films exhibit high transmittance in the visible range (400–800 nm), which is crucial for transparent conducting oxide (TCO) applications. Among the samples, ZnO-03 (blue curve) displays the highest transmittance, consistently above 85% in the visible range, suggesting optimal growth conditions for achieving superior transparency.

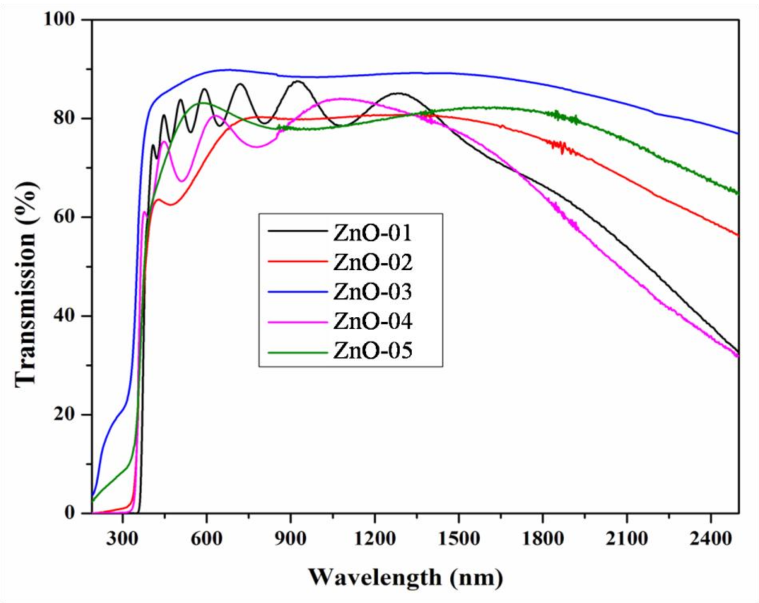

Fig. 4: Transmission spectra for ZnO:Al (1 wt%) deposited at different conditions (deposition conditions are tabulated in table 1)

In comparison, the other films, especially ZnO-02 (red curve) and ZnO-05 (green curve), show relatively lower average transmittance, likely due to increased defect density or suboptimal Al doping levels. The reduced transparency in ZnO-05 could be attributed to excessive oxygen vacancies or Al doping beyond the solubility limit, leading to increased scattering centers or secondary phase formation. ZnO-01 and ZnO-04 also display a moderate decline in transmittance, particularly noticeable in the near-infrared region (1000–2400 nm), which may be linked to increased free carrier absorption caused by higher carrier concentration due to excess Al incorporation, as described in literature [9].

Moreover, the presence of interference fringes in the spectra, particularly prominent in ZnO-01 and ZnO-03, indicates smooth and uniform film surfaces, corroborating high-quality crystalline growth. The damping of these fringes in ZnO-04 and ZnO-05 could suggest increased surface roughness or thickness variations. The observed trends align well with previous findings where optimized Al doping concentration and precise control of PLD parameters (such as laser fluence, oxygen partial pressure, and substrate temperature) were critical in balancing electrical conductivity and optical transparency [10]. Therefore, ZnO-03 appears to be the most promising candidate, offering a favorable combination of high visible transmittance and minimal free carrier absorption, essential for TCO applications.

Electrical (Resistivity) Properties of AZO Thin Films:

The optical transmittance and electrical properties of the Al-doped ZnO thin films deposited via Pulsed Laser Deposition (PLD) reveal a complex interplay between film thickness, resistivity, and transparency. As shown in the transmittance spectra and summarized in the provided table, all samples exhibit high optical transmittance in the visible region, with values ranging from ~79% to ~90%, confirming their potential as transparent conducting oxides (TCOs). Specifically, Sample ZnO-03 stands out, showing the highest average transmittance of 89.9%, which correlates well with its relatively moderate thickness (130.8 nm). This observation is consistent with prior studies, where thin AZO films with controlled thickness demonstrated higher transparency due to minimized light scattering and absorption losses [11].

From the I-V characteristics obtained, the average resistance Rs was obtained for each sample and the corresponding resistivity ρ was calculated using the formula

$$\rho = 4.53 \times R_s \times t$$

where, “t” is the thickness of the film.

However, when examining the electrical properties, a contrasting trend is evident. Despite ZnO-03 having the highest transparency, its sheet resistance is measured at 346.1 Ω, resulting in a resistivity of 2.05×10^{-2} Ω·cm—higher than ZnO-04 (81.7 Ω, 1.03×10^{-2} Ω·cm) and ZnO-02 (270.3 Ω, 7.01×10^{-3} Ω·cm). Notably, ZnO-02, though thinner (57.25 nm), demonstrates a relatively low resistivity but suffers from lower transmittance (~78.9%). This trade-off between conductivity and transparency is a common feature of TCOs, as highlighted in literature, where increased carrier concentration improves conductivity but also raises free carrier absorption, reducing transmittance [12].

Table 2: Resistance and Resistivity parameters of deposited Al doped ZnO thin films

Sample	Thickness (nm)	Current Range (Amperes)	Resistance (Ohms)	Resistivity (Ohm.cm)	%T
ZnO-01	331.7	$-5x10^{-4}$ to $5x10^{-4}$	132.5	$1.99x10^{-2}$	79.6
ZnO-02	57.25	$-5x10^{-4}$ to $5x10^{-4}$	270.3	$7.01x10^{-3}$	78.9
ZnO-03	130.8	$-5x10^{-4}$ to $5x10^{-4}$	346.1	$2.05x10^{-2}$	89.9
ZnO-04	279.0	$-5x10^{-4}$ to $5x10^{-4}$	81.7	$1.03x10^{-2}$	80.6
ZnO-05	363.7	$-5x10^{-4}$ to $5x10^{-4}$	317.9	$5.23x10^{-2}$	83

Sample ZnO-04 exhibits a desirable balance, with a low resistivity (1.03×10^{-2} Ω·cm) and reasonable transmittance (~80.6%), suggesting optimal Al doping and film density under its specific deposition conditions. Conversely, ZnO-05, despite having the highest thickness (363.7 nm) and fair transmittance (~83%), shows a significantly higher resistivity (5.23×10^{-2} Ω·cm), likely due to over-doping or increased defect density, which deteriorates carrier mobility. ZnO-01, with a thickness of 331.7 nm, also displays moderately low transmittance (79.6%) and a resistivity of 1.99×10^{-2} Ω·cm.

In conclusion, comparing these results to previous works it is evident that achieving an optimal balance between film thickness, doping concentration, and deposition parameters is crucial for tailoring the optoelectronic performance of AZO films. In this study, ZnO-03 demonstrates superior optical transparency, while ZnO-04 shows the best electrical performance, making both samples strong candidates depending on the specific TCO application requirements.

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