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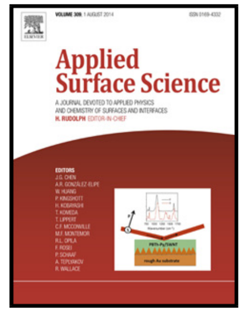

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DC-magnetron sputtering of ZnO:Al films on (00.1)Al_2O_3 substrates from slip-casting sintered ceramic targets

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Keywords Al-doped ZnO, DC-magnetron sputtering, X-ray diffraction, X-ray specular reflectivity.

High (>350°C) temperature DC-sputtering deposition of ZnO:Al thin films onto single-crystal (00.1)-oriented Al_2O_3 (sapphire) substrates is reported, using a ultrahigh-density, low-resistivity and low-cost composite ceramic target produced by slip-casting (pressureless) sintering of ZnO-Al_2O_3 (AZO) powders. The original combination of high-angle θ-2θ (Bragg-Brentano geometry) X-ray diffraction with low angle θ-2θ X-ray reflectivity (XRR) techniques allow us to define the AZO target composition and investigate the structural properties and surface/interface roughness of as-sputtered ZnO:Al films; besides, the growth dynamics of ZnO:Al is unambiguously determined. The target turned out composed of the sole wurtzite ZnO and spinel $ZnAl_2O_4$ phases. X-ray diffraction analyses revealed highly (00.1)-oriented (epitaxial) ZnO:Al films, the material mean crystallite size being in the 13–20 nm range and increasing with temperature between 350°C and 450°C, while the film growth rate (determined via XRR measurements) decreases appreciably. XRR spectra also allowed to determine rms surface roughness <1 nm for present films and showed ZnO:Al density changes by only a few percent between 350°C and 450°C. The latter result disproves the often-adopted Thornton-Model for the description of the sputtered-grown ZnO films and instead points out towards a reduction of the sticking coefficients of impinging species, as the main origin of film growth rate and grain size dependence with temperature. ZnO:Al films appear highly-transparent, with visible-NIR integrated transmittance >97%.

1 Introduction

Thin films of aluminum-doped zinc oxide (ZnO:Al) continue to gather considerable interest as transparent conductive oxide (TCO) for a variety of applications, including photovoltaic cells and transparent thin-film transistors [1–3]. In substitution of the more traditionally used indium-tin-oxide (ITO), ZnO:Al allows for the use of inexpensive, non-toxic and earth-abundant elements [4]. Several deposition methods, e.g., sputtering [5], thermal evaporation [6], chemical vapour deposition [7], sol-gel [8], and spray pyrolysis [9] have been reported in the literature for ZnO:Al films. Among them, the direct current (DC) magnetron sputtering deposition remains one of the most attractive technique, as it is a relatively simple and industrially scalable process, and it guarantees ZnO:Al films with high uniformity over large areas, low (down to ~10^{-4} Ω·cm) resistivity and high (over 90%) visible-NIR light transmission coefficients [10,11].
The properties of DC-sputtered thin films are influenced by applied DC power, sputtering pressure and atmosphere, target-to-substrate distance, substrate temperature, and film thickness. Furthermore, in the case of ZnO:Al films, the use of ceramic targets sintered from composite ZnO-Al_2O_3 powders (so-called AZO targets) generally offers an easier control and a major reproducibility of both sputtering process and film properties with respect to reactive co-sputtering from Zn and Al metallic targets [12]. Therefore, AZO ceramic targets are today the most common choice for industrial applications of DC-sputtering to ZnO:Al thin films, despite their

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