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Thermally-induced formation of secondary phases in ZnO:Al films grown by reactive pulsed magnetron sputtering

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Segregation of Al_2O_3 or $ZnAl_2O_4$ in Al-doped ZnO (AZO) is often discussed as a reason for deterioration of the film electrical properties during growth at temperatures above a certain optimum value (150-300 °C). However, conclusive evidence of these phase segregation in AZO is lacking since electrical properties and crystallinity of the films deteriorate simultaneously and, the disordered structure precludes a proper analysis using conventional methods. The present work overcomes these limitations using characterization techniques based on synchrotron radiation, such as X-ray diffraction and X-ray absorption near edge structures. These studies were combined with investigations by spectroscopic ellipsometry, Hall effect measurements and elastic recoil detection analysis. The AZO films grown by reactive pulsed magnetron sputtering at substrate temperatures, T_S, ranging from RT to 550 °C, were investigated. It is found that Al-sites in an insulating metastable homologous $(ZnO)_3Al_2O_3$ phase are favored above an optimum T_S value (200-400 °C), which depends on the metal/oxygen flux ratio. Energy deposition during growth due to the elevated T_S and from the flux of energetic particles incident on the substrate causes preferential Zn desorption. Thus, increasing T_S above the optimum value leads to a higher Al concentration (c^F_{Al}) in the films, as compared with that of the sputter targets. It exceeds the solubility limit and triggers the formation of this phase, whose volume fraction scales with increasing c^F_{Al}. This impedes crystal growth, causes a significant increase of free electron scattering, and results in an increase of the film electrical resistivity. It is shown that one can grow low-resistivity AZO films in a wider range of T_S using lower metal/oxygen flux ratios during deposition. This may be suggested as an approach to minimizing the influence of such undesirable phase formation on AZO film electrical properties.

Keywords
Transparent conductive oxides
Al-doped ZnO
Reactive pulsed magnetron sputtering
X-ray absorption near edge structure
Phase composition