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Synthesis and evaluation of the structural, microstructural, optical and magnetic properties of $Zn_{1-x}Co_xO$ thin films grown onto glass substrate by ultrasonic spray pyrolysis

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Abstract

Ultrasound pyrolysis spray technique is used to prepare single-phase thin films of $Zn_{1-x}Co_xO$ ($x = 0$–22 at.%). The hexagonal wurtzite structure of the films is confirmed by X-ray diffraction with an average crystallite size estimated in the range of 18–30 nm. The compound structure and stoichiometry of the films are further characterized by energy-dispersive spectroscopy (EDAX). The spectrum analysis agreement great chords between the expected and measured Co atomic content in the films indicating an effective doping. The results also reveal a high solubility of Co into ZnO solid solution at about 14 at.%. For the optical proprieties, the bandgap energy decreases due to the presence of high concentrations of localized states in the thin films. The photoluminescence spectra of all the samples exhibited a broad emission in the visible range. In addition, the magnetic properties of $Zn_{1-x}Co_xO$ thin films are found to be strongly influenced by Co doping.

1 Introduction

Zinc oxide semiconductors have demonstrated extraordinary properties in recent years due to their extensive applications particularly in the field of spintronics. ZnO is a semiconductor with a large bandgap ($E_g = 3.31$ eV) and large exciton binding energy ~ 60 meV at room temperature [1]. In a hand, these properties make them more interesting for optoelectronic applications [2]. On the other hand, the nonstoichiometric structure of ZnO and the intrinsic structural defects such as oxygen vacancies (V_O) and zinc interstitial (Zn_i) improves the transparent conductive properties [3, 4]

Recently, transition metal-doped semiconductors have been at the core of various research reviews due to their exceptional optical properties and the promising potential applications in the optoelectronic medium [5-8]. The doping of ZnO by transition metals, in particular by cobalt, draws the attention of many researches. Cobalt (Co) doping is greater because of similar ionic radii (0.058 nm) to that of Zn (0.060 nm), and its powered magnetic moment compared to other 3*d* metals ($\mu_{Co} = 1.8\ \mu_B$) [9]. This type of doping is known as diluted magnetic semiconductor (DMS) because ZnO provides ferromagnetic properties at room temperature [10].

Thin layers of Co-doped ZnO were synthesized by the use of different evaporation techniques [11-14]. In thin film, doping ZnO with Co^{2+} improves optical absorption and decreases the band gap which is assigned to the *sp*–*d* spin exchange interaction [15-17]. Note that the defect environment can be distort when Co dopant atom substitutes a Zn atom. Along these lines, it is interesting to study the influence of this doping on the optical properties of Co-doped ZnO.

Many studies listed in the literature, on the elaboration of thin layers of ZnO, $Zn_{1-x}Co_xO$ with different techniques under different experimental conditions, produce in the formation of single-phase films, with different properties.

Ivill et al. [18] observed the plane, i.e., (0 0 2) of ZnO along with the secondary phase of (0 0 4) plane of Co ions. They observed paramagnetic behavior for 0–15 at.% of cobalt, for higher (>30 at.%) cobalt, they noticed ferromagnetic behavior, which was due to intrinsic defects and a large amount of Co^{+2} within the ZnO lattice. Tarwal et al. [19] prepared Co-doped ZnO films with 0–20 at.% doping, confirmed the polycrystalline nature of the films with (0 0 2)

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