

dc and ac transport properties of Mn-doped ZnO thin films grown by pulsed laser ablation

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Abstract

Mn_xZn_{1-x}O ($x=0.20$) thin films were deposited on Pt coated Si substrates using pulsed laser ablation technique. The structural characteristics of the films were investigated by X-ray diffraction (XRD), while the dielectric response of the films was studied as a function of frequency and ambient temperature by employing impedance spectroscopy. It was found that all the films deposited on Pt coated Si substrates had c-axis preferred orientation perpendicular to the substrate, with full width at half maximum (FWHM) of the (002) X-ray reflection line being less than 0.5°. The dc and ac electrical conductivity of Mn-doped ZnO films were investigated as a function of temperature. The ac conductivity, $\sigma_{ac}(\omega)$, varies as $\sigma_{ac}(\omega) = A\omega^s$ with s in the range 0.4–0.9. The complex impedance plot showed data points lying on a single semicircle, implying the response originated from a single capacitive element corresponding to the bulk grains. The value of the activation energy computed from the Arrhenius plot of both dc and ac conductivities with $1000/T$ were 0.2 eV suggesting hopping conduction mechanism. The optical properties of Zn_{0.8}Mn_{0.2}O thin films were studied in the wavelength range 300–900 nm. The data were analyzed in the light of the existing theories and reflected a Burstein–Moss shift in these films. The films show magnetic properties, which are best described by a Curie–Weiss type behavior.

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1. Introduction

In recent years, spintronics [1,2] has been studied extensively by many of the researchers due to its spin-dependent phenomena, which can be applied to modern electronic devices. Diluted magnetic semiconductors (DMS) are considered ideal systems for spintronics, in which transition metal ions replace cations of host semiconductor materials. The replaced transition metal ions couple with the electrons in the semiconducting band. Such a spin–spin exchange interaction between the d electrons of magnetic ions and band carriers results in large Zeeman splitting of band states and giant Faraday rotation. Such a coupling leads to various interesting properties such as magneto-optical and magneto-electrical effects [3]. ZnO can be used as a host material for the growth of diluted magnetic semiconductors in which Mn will substitute the Zn cations. Fukumura et al. [4] have studied epitaxial thin films of Mn-doped ZnO by pulsed laser ablation

with Mn content as high as 35 at.%. Because ZnO is an attractive optical material with large optical band gap (~ 3.3 eV) [5] and high exciton binding energy (60 meV) [6], magneto-optical properties of DMS based on ZnO would be useful for a variety of short wavelength optical applications. Though intensive efforts are evident on the growth of Mn-doped ZnO films by pulsed laser deposition method, their electrical properties were not studied in detail.

Impedance spectroscopy has been used as a powerful technique to determine the conductivity of the ionic conductors [7]. Recently, power law analysis of the ac conductivity data has been found to be a tool for studying the ion dynamics of such systems. In most of the doped semiconductors [8] transport mechanism occurs through hopping. As suggested by Pollak and Geballe [9], the transport occurs by the electrons hopping between states, which are essentially localized around the acceptor or donor impurities. For such a hopping to take place, it is necessary for some of the localized states to be vacant and hence compensation of the majority impurity becomes essential feature of impurity conduction. In this paper, we report the growth of Mn-doped ZnO thin films by pulsed laser ablation. Hopping conduction in

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Mn-doped ZnO were studied and verified by analyzing the dc and ac conductivity data.

2. Experimental details

Pulsed laser ablation has been used to deposit thin films of variety of materials including ZnO, due to several advantages such as good stoichiometry control, faster growth rate, etc. A single phase dense $\text{Zn}_{1-x}\text{Mn}_x\text{O}$ ($x=0.2$) target was prepared via a conventional solid-state reaction by mixing and calcining the multicomponent oxides at 900°C for 4 h and then the powders were pressed into a pellet. The pressed pellet was sintered at 1000°C for 4 h to form $\text{Zn}_{1-x}\text{Mn}_x\text{O}$ target. The target was used for the ablation and the thin films were deposited on different substrates such as Pt coated Si and corning glass by KrF (248 nm) pulsed excimer laser. The oxygen pressure in the deposition chamber was kept at 50 mTorr. Substrate temperature was varied from 500 to 650°C . The deposition was carried out for 20 min. Structural phase determination was carried out using X-ray diffractometer and the grain morphology was examined by scanning electron microscope. The thickness of the film was measured using Dektak thickness profilometer. The measured thickness of the films was about $0.2\ \mu\text{m}$. Nickel dots of $1.96 \times 10^{-3}\ \text{cm}^2$ area were deposited by thermal evaporation onto $\text{Zn}_{0.8}\text{Mn}_{0.2}\text{O}$ films as top electrodes in order to study the electrical properties of the films. The electrodes were annealed at 200°C for 30 min in order to minimize the contact resistance and to enhance the adhesion of the deposited metal film. While the dc conductivity measurements were carried out using Van der Pauw technique, the ac conductivity measurements were done using impedance spectroscopy. Capacitance–voltage characteristics were carried out using computer aided LCZ meter. Electrical properties in frequency domain (ac electrical properties) were studied using Keithley 3330 LCZ meter with an oscillation level of 0.1 V over a temperature range of 30 – 100°C . The range of frequencies selected was from 100 Hz to 100 kHz. Magnetic properties were examined using superconducting quantum interference device magnetometer (SQUID).

3. Results and discussion

3.1. Structural characterization

The structure details of Mn-doped ZnO thin films were determined by X-ray diffraction technique. Fig. 1 shows the XRD pattern of the films deposited at 50 mTorr at various substrate temperatures on Pt coated Si substrates. For the whole range of deposition temperatures employed in this study (500 – 650°C), all the films were found to be *c*-axis oriented, exhibiting (002) XRD reflection lines. Fig. 1 (inset) shows the FWHM of Mn-doped ZnO (002) XRD peak at different substrate temperatures. FWHM was found to decrease with increase in substrate temperature, which was attributed to the increase in grain size. The grain size of the films calculated from the Scherrer formula was found to lie in the range of 30–40 nm. From the compositional analysis (obtained by EDAX) of the films, it was found that 20 at.% is substituted in the ZnO lattice by Mn.

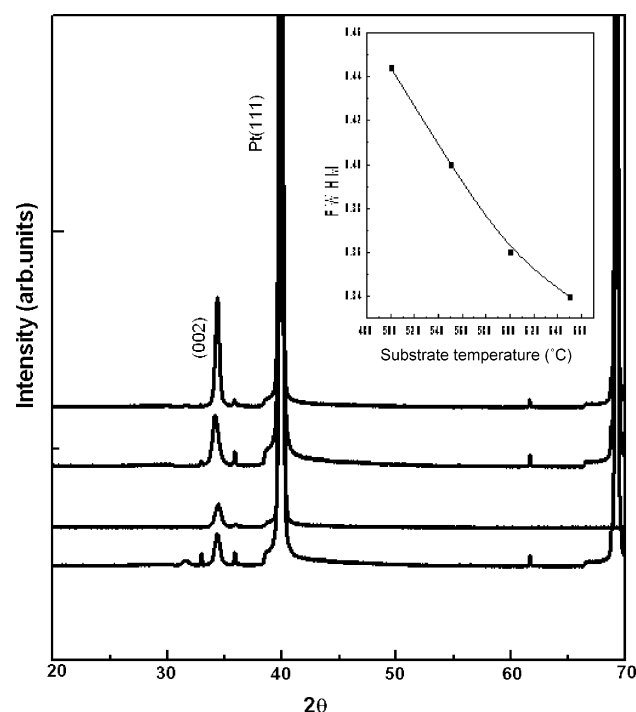


Fig. 1. XRD pattern of $\text{Zn}_{0.8}\text{Mn}_{0.2}\text{O}$ films at different growth temperatures.

3.2. dc conductivity

Fig. 2 shows the room-temperature electrical conductivity as a function of substrate temperature (T_s). As calculated from the XRD spectrum by Scherrer formula, the grain size increases with increase in T_s . This would make the mobility to increase, which in turn increases the conductivity. In order to investigate the nature of conductivity in these films temperature-dependent studies were carried out. Hopping conduction in doped semicon-

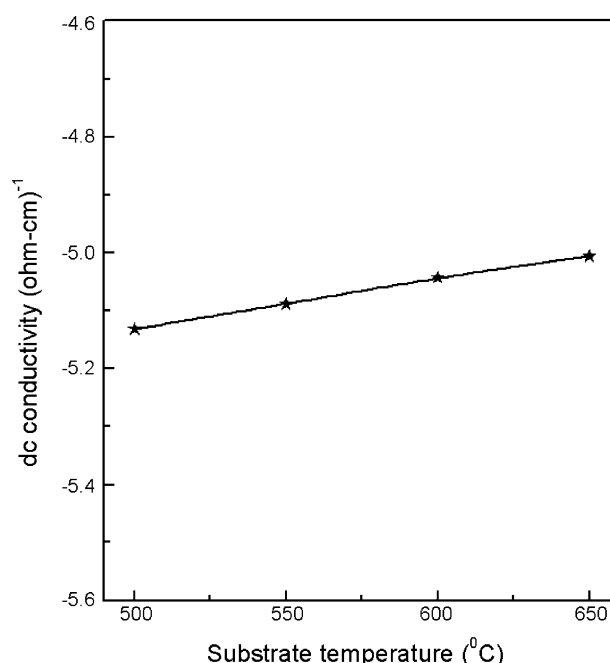


Fig. 2. Dependence of dc conductivity on substrate temperature (T_s).

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