



# Detecting spin polarization of nano-crystalline manganese doped zinc oxide thin film using circular polarized light



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## ABSTRACT

The presence of spin polarization in Mn-doped ZnO thin film is very important for spintronic applications. Spin polarization was detected using simple method. This method depends on measuring the optical transmittance using circular polarized light in visible and near infra-red region. It was found that, there is a difference in the optical energy gap of the film for circular left and circular polarized light. For temperatures  $>310$  K the difference in energy gap is vanished. This result is confirmed by measuring the magnetic hysteresis of the film. This work introduces a promising method for measuring the ferromagnetism in diluted magnetic semiconductors.

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## 1. Introduction

Mn-doped ZnO is one of the promising diluted magnetic semiconductors (DMSs) for fabricating spintronic devices. The essential criterion to develop DMSs is the presence of ferromagnetism at room. Theoretical calculations [1] based on Zener's p-d exchange model has predicted that wide-gap semiconductors such as Mn-doped ZnO are promising candidates for room temperature ferromagnetic DMSs. In fact, some experimental results [2,3,4] reported the observation of ferromagnetism while others [5,6,7] reported the absence of ferromagnetism in these materials. Moreover, the first-principles calculations [8], expected that, ferromagnetism could be created due to hole doping, e.g., through N substitution for O. These reports imply that hole doping is necessary for ferromagnetism in Mn-doped ZnO (ZnO:Mn). Experimental studies reported the presence of ferromagnetic component for N-doped ZnO:Mn (ZnO:Mn,N) [9,10]. Also, ferromagnetism has also been observed for n-type ZnO:Mn [11]. The discrepancy between the experimental results may be attributed to the preparation method and the change of the microstructure of the prepared samples. Many theoretical studies showed that, the electronic structure, density of states and energy gap of ferromagnetic material depend on its spin polarization [12,13,14]. The detection of the spin polarization is the most important evidence about the presence of the ferromagnetism in DMSs. One of the most important methods for detecting the magnetic properties of DMSs is Faraday Effect (circular birefringence) [15]. This method depends on measuring the rotation of plane polarized light during the propagation in the DMSs materials [16,17]. It is found that, the rotation

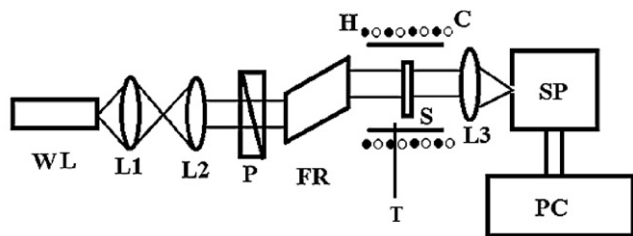
angle has a strong correlation with the magnetic interactions between the magnetic moments in DMSs [18,19]. F. Oliveira et al. studied spectral response of Faraday rotation of Mn-doped ZnO thin film [20]. They found that, Verdet constant has strong dependence on both of the Mn concentration and the photon energy.

The coupling between the circular polarized light and the magnetic properties of any magnetic oxides (not only DMS) is arising from the circular dichroism (Faraday Effect). If there is difference between the number of parallel and antiparallel magnetic moments (spins), in the presence of a magnetic field (external or internal), there will be a change of the optical properties of the sample, refractive index and energy gap, with changing the state of the circular light (left or right rotation). The previous works were measuring the rotation angle in the plane of polarization with the magnetic field. It is known that, the plane polarized light could be considered as sum of two circularly polarized light (Left and Right). Therefore, the variations in the optical properties of the DMSs between the two components may give an information about the magnetic interactions in these materials. In this work, the detection of the spin polarization in (ZnO:Mn,N) using simple optical method is introduced. This method depends on measuring the circular birefringence of the optical transmission spectra of (ZnO:Mn,N) thin film, in Vis–NIR region using circularly polarized light.

### 1.1. Experimental details

Nitrogen-doped  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}_4$  thin film was deposited on properly cleaned soda lime glass substrates by chemical spray pyrolysis technique. The precursor solution was made by dissolving high purity Zn-acetates and Mn-acetates (Merk, india) with the proper concentration is distilled water and ethanol solution with ratio 1 : 3. To obtain good

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WL: White light source L1: Condensing lens L2: Collimating lens P: Polarizer

FR: Fresnel Rhomb ( $\lambda/4$ ) H: Heating wires C: Cooling Pipes T: Thermo couple

S: Sample L3: Projecting lens SP: Grating Spectrometer PC: Computer

Fig. 1. Optical setup for measuring the optical transmittance of  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$  thin film at different temperatures.

quality ZnO thin film, the optimized value of substrate temperature was found to be  $400 \pm 10^\circ\text{C}$ . The carrier gas was nitrogen. After spraying, the samples were annealed at  $450^\circ\text{C}$  for 3 h in nitrogen atmosphere.

Surface morphology of the film was studied in a field emission scanning electron microscope (FE-SEM: FBI company Quanta FEG 250).

Fig. 1 shows the experimental setup of the optical transmission measurement in both left and right circular polarized light. The absorption coefficient  $\alpha$  was estimated from the formula  $\alpha = (1/d) \ln(1/T)$ , where  $d$  is the film thickness and  $T$  is the optical transmittance. Film thickness was determined by interference method. The film thickness was found to be 100 nm. The magnetic hysteresis of the prepared sample were measured using vibrating sample magnetometer model 7410 Lakeshore USA.

## 2. Results and discussion

### 2.1. X-ray and morphology

Fig. 2 shows XRD pattern of the  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$  thin film. Comparing the  $d$  values of the XRD peaks with those reported in JCPDS, it is found that the sample has hexagonal wurtzite single phase structure. The lattice constants are found to be  $a = 3.254\text{\AA}$  and  $b = 5.226\text{\AA}$  which are in good agreement with those reported in literatures [21,22]. Furthermore, the XRD pattern consists of a (0 0 2) main peak, which means that the crystallites are highly oriented with their  $c$ -axis perpendicular to the plane of the substrate. From the FE-SEM image of the sample shown in the inset of Fig. 2, the grain size is found to be about 45 nm. In spite

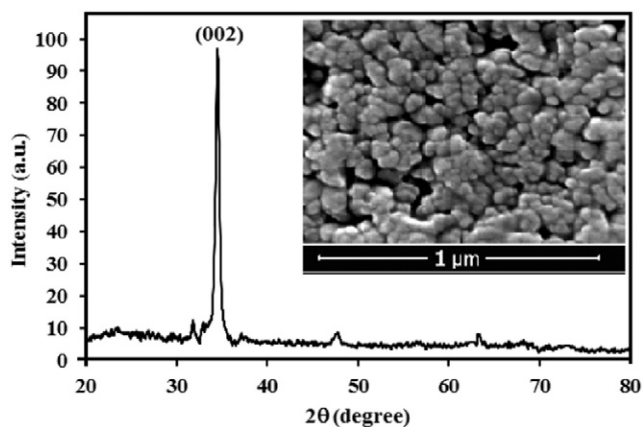


Fig. 2. X-ray diffraction pattern of  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$  thin film. The inset shows FE-SEM image of the sample.

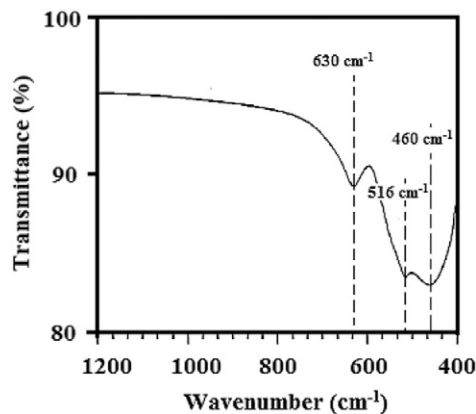


Fig. 3. FTIR spectrum of  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$  thin film.

of annealing of the sample, the grain size is found to be in the nano scale. This could be attributed to the presence of clusters of MnO at the grain boundaries. Fig. 3 shows FTIR spectrum of  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$ . It is obvious that there are two peaks at  $460\text{ cm}^{-1}$  and  $516\text{ cm}^{-1}$  which belong to Zn–O vibrational modes [23]. The peak at  $630\text{ cm}^{-1}$  is a characteristic vibrational frequency of Mn–O [24] which confirms the presence of MnO clusters.

The quantitative analysis of the films was carried out by using energy dispersive X-ray (EDX) analysis to study the stoichiometry of the nanostructured thin films. Fig. 4 represents EDX spectra for the prepared  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$  thin film. The results exhibit the presence of Zn, Mn and O peaks. Also, there is small peak at 0.524 keV which belongs to  $K_{\alpha}$  line of nitrogen which confirms the doping of the prepared sample with nitrogen. The Zn/Mn/O/N weight percentage are found to be (55.8/6.5/35/2.7) respectively.

### 2.2. Optical measurements

Fig. 5 shows the transmission spectra of  $\text{Mn}_{0.1}\text{Zn}_{0.9}\text{O}$  thin film for both circular left ( $T_L$ ) and circular right ( $T_R$ ) polarized light at different temperatures. It is noticed that, the sample has circular birefringence and by increasing the temperature  $T_L$  curve approaches  $T_R$  curve at the absorption region.

Fig. 6 shows Tauc plot for which the optical band gap of the sample has been calculated for both circularly left ( $E_{gL}$ ) and circularly right ( $E_{gR}$ ) polarized light [25]. Table 1 shows the variations of  $E_{gL}$  and  $E_{gR}$  with temperature. One can observe that, there are variations between the two energy gaps. Furthermore, for Temperatures  $\leq 310\text{ K}$ ,  $E_{gR}$  values

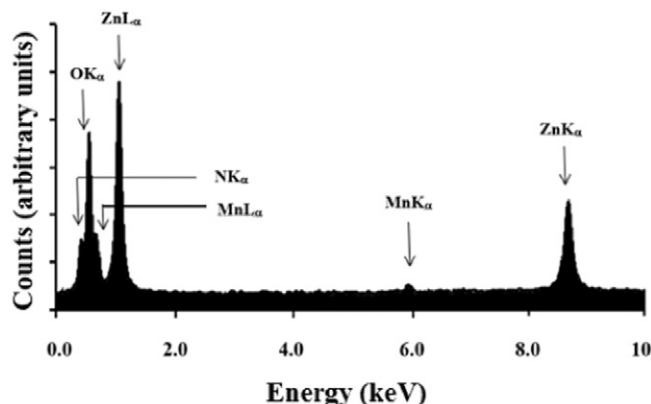


Fig. 4. Energy dispersive X-ray (EDX) spectra of  $\text{Mn}_{0.05}\text{Zn}_{0.95}\text{O}$  thin film.

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