Thin Solid Films 589 (2015) 817–821

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Electrical and magnetic transport properties of undoped and Ni doped ZnO thin films

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ARTICLE INFO

Article history:
Received 17 November 2014
Received in revised form 15 July 2015
Accepted 15 July 2015
Available online 17 July 2015

Keywords:
Zinc oxide
Semiconductors
Magnetoresistance
Magnetic scattering
Magnetic polaron

ABSTRACT

We report the transport and longitudinal magnetotransport studies of undoped and nickel (5 at.% Ni) zinc oxide thin films prepared by pulsed laser deposition technique. Low temperature (3 K) negative magnetoresistance observed in pure and Ni doped films exhibit nearly same character and is ascribed to localized magnetic moment, while at high temperatures, three times larger magnetoresistance in Ni doped ZnO has been observed and has been assigned to the magnetic scattering due to transition metal ion impurity. Enhanced magnetic ordering in NiZnO thin film up to room temperature is also observed from its magnetization data.

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1. Introduction

II–VI semiconductors have always been the source of potential research in the field of optoelectronics [1,2], spintronics [3], photovoltaics [4] etc. Doping these semiconductors with a small amount of magnetic ions forms a class of semiconductors called as diluted magnetic semiconductors [5]. Such materials provide an opportunity to combine the semiconducting and magnetic properties together, which can be explored for spintronic applications.

Among II–VI semiconductors, ZnO is one of the largely studied compounds which has profound applications in spintronics [6,7]. Properties like large band gap (3.4 eV), long spin coherence (10.7 nm at 10 K) [8] and large exciton binding energy (60 meV) make it highly attractive for these applications. Efficient spin injection, transport and polarization are the key factors for the spintronic applications [9].

Transition metal ion doping induces the localized magnetic moments in the ZnO matrix which undergo indirect exchange interaction mediated via charge carriers when subjected to external magnetic field. The external field aligns the moments and results in the exchange interaction between band (s/p) electrons of ZnO and the d-electrons of the impurity ions leading to the spin polarized transport [10]. Spin polarization is the direct consequence of this sp–d exchange interaction. Magnetoresistance (MR) measurement is one of the important tools to probe the presence of spin polarized transport in magnetic semiconductors. Apart from this, in semiconductors, the density of magnetic impurity atoms strongly affects its conductivity as well as magnetotransport phenomena.

Doping with Ni in ZnO not only imparts the magnetism [11] in ZnO but also results in the red shift of the band gap [12]. These properties combiningly make it an important source for the use in monolithic optical integrated circuit application with higher threshold [13]. The large driving force for phase segregation into NiO and ZnO in Ni doped ZnO makes it very challenging to dope Ni at higher concentrations typically greater than 5% [14]. Only few studies related to the magnetic and transport properties of Ni doped ZnO have been reported so far. Also the nature of magnetotransport phenomena is not clearly known till now. Recently, Chickodoize et al. have observed positive MR at 300 K and negative MR at 80 K in 1% Ni doped ZnO thin films [15]. They have attributed the origin of positive MR to electron–magnetic impurity interaction and intergrains tunneling, where as negative MR is attributed to the electron hopping mechanism. Since the measurements were performed in transverse geometry so the effect of Lorentz force could be another contribution to their findings. In our opinion the absolute contributions of electron–magnetic impurity interaction and intergrains tunneling to the MR can be judged in a parallel geometry where Lorentz force contributions are negligible. In our previous magnetotransport measurements on pulsed laser deposited (PLD) Ni doped ZnO thin films, we found the change in sign of MR from negative to positive at 2 K temperature and 6 T magnetic field while only negative MR was observed at room temperature [12].

The understanding of the transport properties under such circumstances requires the knowledge of the modifications in the electronic

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http://dx.doi.org/10.1016/j.tsf.2015.07.028
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structure of the host ZnO matrix due to doping of magnetic impurities in it.

In the present manuscript, we have studied the valence band spectroscopy (VBS) of undoped and Ni doped ZnO thin films grown by PLD. We have also studied the role of magnetic field (H), temperature (T) and impurity dependence on MR and magnetism in doped ZnO.

2. Experiment

For the present investigations, pure and Ni doped ZnO (5 at.% Ni) thin films of ≈ 200 nm thickness were grown on Silicon (100) substrate by PLD technique using excimer (KrF) laser (λ = 248 nm). The targets in the form of pellets of 25 mm diameter for the film deposition were made by standard solid state reaction route. These targets were initially calcinated at 900 °C for 24 h and then sintered at 1400 °C for 12 h [12]. The substrate was initially washed with methanol followed by acetone and then ultrasonicated. The substrate was kept at a distance of 5 cm from the target and at 400 °C temperature. Laser energy was set at 220 mJ with repetition rate of 10 Hz. The deposition was carried out in the chamber evacuated up to 2.5×10^{-3} Pa and the oxygen partial pressure during the deposition was maintained at 1.3×10^{-2} Pa. The films were characterized by X-ray diffraction (XRD) with Bruker D8 advance powder diffractometer using Cu-Kα radiation source (λ = 1.541 Å) for structural analysis. To get an insight of electronic/ionic state of Ni in the ZnO matrix, we have performed X-ray photoelectron spectroscopy (XPS) measurements using Al-Kα lab source and at a vacuum better than 1.3×10^{-7} Pa at photoelectron beamline of RRCAT, Indore, India. The contamination present on the sample surface due to the exposure to open atmosphere was reduced by bombardment of 500 V Ar^{+} ions for about 5 min. All the data was recorded by an Omicron hemispherical analyzer. After subtracting the Tougard background, all the XPS core-level (CL) data were fitted with mixed Lorentzian–Gaussian profile. To explore the modifications of valence band due to doping in ZnO, we performed ultraviolet photoelectron spectroscopy (UPS) (also called as VBS) measurements under the same conditions present during XPS measurements. The samples were excited at photon energy ≈ 67 eV using AIPES beamline facilities of INDUS-1, RRCAT Indore, India. The conductivity variation with temperature was examined by the standard linear four probe technique in the temperature range between 5–300 K on rectangular shaped samples. Change in resistance due to the application of magnetic field at constant temperatures (3 K and 320 K) was also studied. The magnetic field is applied parallel to the direction of the applied current giving rise to the longitudinal MR. Magnetization as a function of temperature was also recorded using Quantum design SQUID-VSM at 0.05 T magnetic field in the same geometry as that of MR.

3. Results and discussion

3.1. XRD results

The XRD patterns of pure ZnO and NiZnO films are presented in Fig. 1. From the figure, it can be concluded that within the XRD detection limits, prepared films are single phase, preferentially textured along c-direction, retaining the hexagonal wurtzite structure of ZnO. The c-axis lattice parameter of the samples calculated from the (002) reflection plane are found to be almost equal i.e. 5.194 Å and 5.197 Å for ZnO and NiZnO films, respectively.

3.2. XPS and VBS results

In Fig. 2(a), we have given the detailed narrower scan of Ni 2p core-levels (CLs) which clearly shows that the peak position of Ni $2p_{3/2}$ is 853.6 eV and that of $2p_{1/2}$ is 870.5 eV which lie very close to NiO [16]. This observation suggests that Ni is existing in +2 oxidation state ruling out the presence of metallic Ni. It is a well known fact that the transport properties of ZnO are greatly affected by the presence of defects like

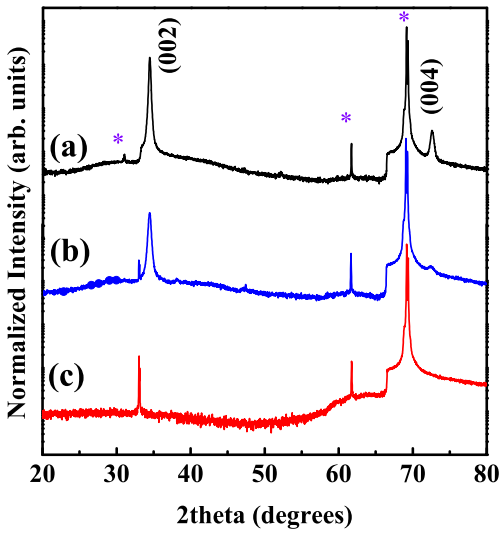

Fig. 1. X-ray diffraction patterns of the films; (a), (b) and (c) correspond to the diffraction patterns of pure ZnO, NiZnO films and Si(100) substrate, respectively. '*' represents the diffraction peaks of the substrate.

oxygen vacancies (V_O) [17]. Thus in order to investigate the presence of V_O, we have also recorded the O-1s XPS CLs in both the films as shown in Fig. 2(b) and (c). The asymmetric line shape of the O 1 s CL

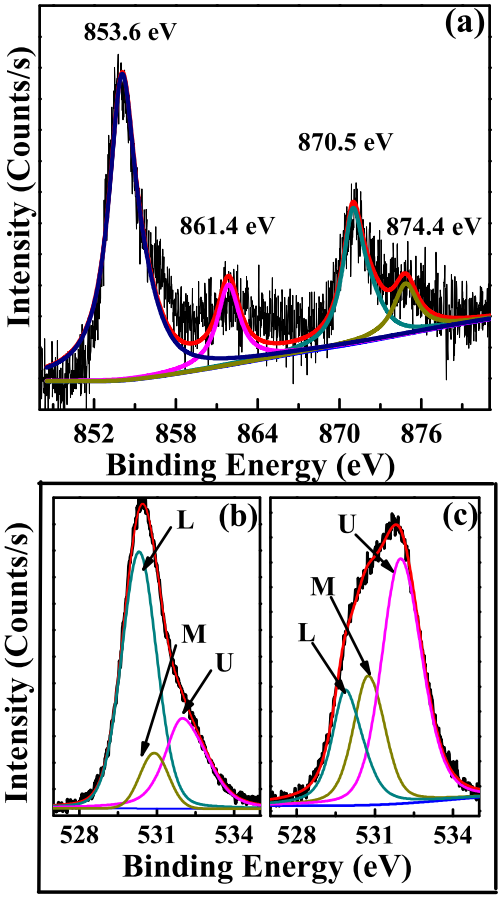

Fig. 2. Detailed scan of XPS core levels; (a) represents the Ni 2p XPS CL in NiZnO film, (b) and (c) shows the O 1 s CL in pure and NiZnO films, respectively. The peaks 'L', 'M' and 'H' being the lower, middle and higher binding energy peaks appearing at ≈ 530, 531 and 532 eV that corresponds to O^{2-} ions in ZnO matrix, oxygen vacancies and chemisorbed oxygen species, respectively.

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