



AC-magnetic susceptibility of Dy doped ZnO compounds



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ABSTRACT

Dy doped ZnO polycrystalline diluted magnetic semiconductor compounds have been prepared by the so called solid state reaction method. We have studied the M – H and AC magnetic properties of the compounds by using a PPMS magnetometer, and explored the phases and crystal structure by using a X-ray powder diffractometer. The XRD spectra of the compounds show that the substitution of Dy^{3+} for Zn^{2+} causes almost no change in the hexagonal wurtzite structure of ZnO, and the Dy^{3+} ions are successfully substituted into the Zn^{2+} site of the ZnO matrix. The magnetic measurements, M – H and χ – T , for T in the range from 10 to 300 K, show a paramagnetic behavior, including indirect antiferromagnetic couplings between some Dy^{3+} magnetic moments. Since the Curie–Weiss temperatures, θ , are all negative but decrease in magnitude with increasing Dy concentration. On the other hand, the calculated effective magnetic moments, μ_{eff} , per Dy^{3+} ion slowly increase with increasing Dy concentration, but are all very close to the free ion value of μ_{eff} , $\sim 11.0 \mu_B$. Therefore, the trends of the magnitudes of θ s and μ_{eff} s indicate that the samples are not only paramagnetic but also have antiferromagnetic couplings due to the complex nature of the compounds. In addition, the thermal variation of average magnetic moment, $P_{\text{eff}}(T)$, per Dy^{3+} ion have been calculated, and have been found to be gradually increasing with increasing temperature and Dy concentration.

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1. Introduction

During the recent years, some of scientists all around the world have explored microelectronics which include the spin and charge of some magnetic materials; named as spintronics (spin and electronics). Such materials have both magnetic and transport properties [1]. However, in order to obtain practical spintronic devices, the materials must exhibit ferromagnetic properties at high temperature. If the materials have semiconductor properties, as well as, the ferromagnetic properties, they are termed as Diluted Magnetic Semiconductors (DMSs). These materials are prepared by entering magnetic atoms or ions into the solid solutions of some semiconductor host materials, and thus are referred as magnetic semiconductor alloys or DMSs [2]. Doped semiconductors show different magnetic behaviors, depending upon the type of magnetic ions, concentration of them, synthesis route and experimental conditions [2–6]. The ferromagnetism, required for spintronic devices, is induced by incorporation of either transition metal (TM) or rare earth (RE) ions into the substitutional sites of the chosen semiconductor. TM and RE ions have partially filled d and f shells, respectively, which give rise to unpaired electrons. The d and especially f electrons are localized. However,

the localized d or f electrons of magnetic ions can couple with the extended electronics in the semiconducting band, which play an important role in the ferromagnetic origin [4]. For this purpose the semiconducting compounds of the II–VI group, i.e. ZnO, ZnS, CdS, are very useful, because of their potential applications; such as the production of solid-state lighting devices (LEDs), photonics [7], nanoelectronics [8], optoelectronics, magnetoelectronic and spintronic devices [5,9]. In this sense, Zinc oxide (ZnO) is one of the most important compounds, having a direct and wide band gap of about 3.4 eV at room temperature [10]. Earlier experimental studies related to the TM (or RE) doped ZnO compounds have shown ferromagnetism (FM) at even room temperature (RT). A few of them are: Co [11], Cr [12], Ni [4] and Gd [13] doped ZnO compounds. FM in DMS is believed to arise from the exchange interaction between the localized d (or f) orbital's spins on the TM (or RE) ions, mediated by the spins of the free (delocalized) carriers (electrons or holes from the valence band). Hence, the presence of free carriers is a required condition for the appearance of FM [14] in DMSs. If RE ions are used in DMSs, the crystal field and exchange interactions are much weaker than the spin–orbit interactions in the rare earths, since, the $4f$ electrons are much more localized. Moreover, as their orbital moments are not quenched, the rare earths have high total magnetic moments per atom [15,16]. Therefore, RE doped DMSs are expected to be weak ferromagnets or non-ferromagnetic.

In this paper, we have experimentally investigated the

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structure and the outcomes of AC magnetic susceptibility and $M-H$ measurements on small weight percentages of Dy doped ZnO DMS compounds prepared as described in the following section. Therefore, our aim is to find out the type of magnetic interactions between the magnetic moments of the RE Dy^{3+} ions in the ZnO matrix.

2. Experimental procedure

In this work, we synthesized the polycrystalline Dy doped ZnO compounds by the solid state reaction method, and labeled them as ZD1, ZD2, ZD3, ZD4 and ZD5 for 5%, 6%, 7%, 8% and 10% of small Dy weight percentages, respectively. The raw materials were pure ZnO and Dy_2O_3 powders, which are known to be non-magnetic and magnetic, respectively. In the solid state reaction route, first appropriate amounts of ZnO and Dy_2O_3 powders were mixed and ground in an agate mortar. The resulting powders were then calcinated at 350 °C for 4 h in air, and thereafter they were furnace cooled. After cooling, the resulting materials were reground and pelletized in the form of a cylindrical die 10 mm in diameter and 2 mm thick, under a pressure of 20 bars. Finally, all the pellets were heat treated at 950 °C in air for 24 h, and were then furnace cooled to the room temperature.

In order to elaborate the structural properties of the ZnO:Dy compounds X-ray powder diffraction (XRD) (with Philips X'Pert Pro, with a wavelength of 1.54 Å in the wide angle region from 25° to 70° on 2θ scale) was used. The AC magnetic properties of the compounds were measured by using a Quantum Design–Physical Properties Measurement System (PPMS) with a closed cycle helium cryostat, in a field of 10 Oe at 1 kHz.

3. Results and discussions

The X-ray powder diffraction was used to explore the phases and crystal structures of all the compounds. The XRD patterns, in range of $25^\circ \leq 2\theta \leq 70^\circ$, are shown in Fig.1. From that figure, one can see twelve strong peaks for the different Dy doped ZnO compounds. Eight of them are also observed for the pure ZnO, and the remaining four peaks emerge after the substitution of Dy^{3+} for Zn^{2+} . In addition, a new but small diffraction peak, due to the Dy_2O_3 phase, emerges at $2\theta \approx 43.7^\circ$ in the XRD patterns. In other words, the XRD patterns are in good agreement with the standard JCPDS file for ZnO. Since, after the partial substitution of Zn by Dy,

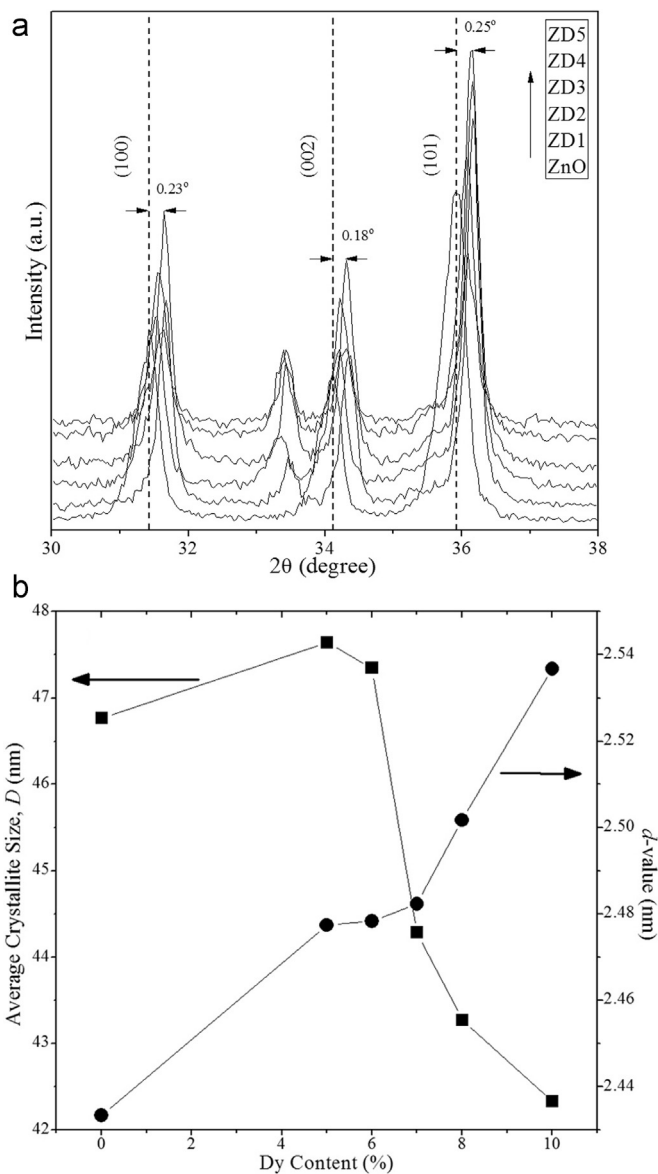


Fig. 2. (a.) The expanded XRD patterns of powdered ZnO:Dy samples in the range of $2\theta = 30^\circ - 38^\circ$, (b.) The average crystallite size and the lattice spacing, d , of the (101) plane versus Dy concentration.

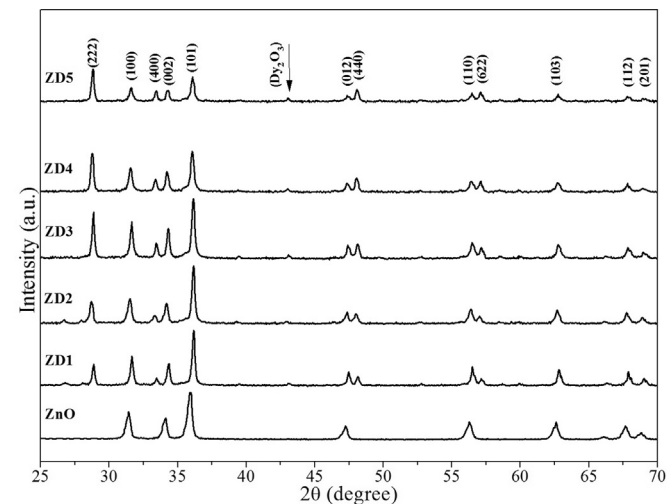


Fig. 1. XRD patterns of the powdered ZnO:Dy samples.

the diffraction patterns of all the compounds show almost similar peaks to that of ZnO, it may be pointed out that the wurtzite structure of ZnO having space group $P6_3mc$ is not much disturbed by Dy substitution. On the whole, the results clearly show that the Dy^{3+} ions have been successfully substituted for Zn^{2+} ions of the crystal lattice of ZnO. To investigate the detailed effect of substitution of the Dy^{3+} ions into the ZnO lattice, the range of $30^\circ \leq 2\theta \leq 38^\circ$ of the XRD spectra is expanded and shown in Fig.2a. The three main reflections due to the (100), (002) and (101) planes (at $2\theta \approx 31.4^\circ$, 34.1° , and 35.9°) of the samples show shifts to higher 2θ values as compared to that of the pure ZnO. The amount of shifts for the (101) planes of the samples of the compounds are tabulated in Table 1. It is obvious that the amount of shifts slightly decreases with increasing Dy concentration. This means that small variations in the lattice parameters occur as the Dy concentration increases. Previous studies related to Dy^{3+} doped ZnO [17] and Eu^{3+} doped ZnO [18] also reported similar phenomena. The d values obtained from the peaks related to the (101) planes are also given in Table 1 and plotted as a function of

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