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The Effect of Heat Treatment on Electrical Properties of $[ZnO/C]_{25}$ Multilayer Structure

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Abstract. The effect of heat treatment on magnetoresistance of $[ZnO/C]_{25}$ multilayered thin films prepared by ion-beam sputtering has been investigated. A decrease in the magnetoresistance with an increase in the annealing temperature was detected, which is associated with a change in the mechanism of electrical transport.

INTRODUCTION

Undoped zinc oxide ZnO is a wide-gap semiconductor belongs to the group of $A_{II}B_{VI}$ compounds with electronic type of conductivity. Interest in this compound is caused by a wide range of properties. Particularly ZnO doped with various chemical elements and especially transition metals demonstrates the ferromagnetism at room temperature, which makes it a promising material as a diluted magnetic semiconductor with a high Curie temperature for use in spintronics devices. Recently, magnetic ordering was also discovered in ZnO doped with non-magnetic carbon C. Despite the fact that there is currently no reliable explanation of the physical nature of magnetic ordering in the ZnO-C system, and the experimental results presented in the publications are contradictory, most researchers are inclined to that defects of the crystalline lattice are responsible for ferromagnetism of such systems. However, there is no consensus on questions what these defects are, how they interact with each other, and why this interaction leads to the appearance of ferromagnetism at room temperature. Obviously, heat treatment allows controlling the degree of defectiveness of semiconductors and to some extent control their properties. Therefore, the investigation of the heat treatment on the electrical properties of the ZnO/C multilayer system, particularly magnetoresistance, is actual both as fundamental and practical points of view.

EXPERIMENTAL DETAILS

Multilayered $[ZnO/C]_{25}$ thin films were observed by ion beam layer-by-layer sputtering of ZnO and C targets according to the method described in [1]. Deposition was carried out in argon atmosphere (Ar purity of 99.998 %) at a pressure of 7×10^{-4} Torr on ceramic ST-50 (sitall), glass and silicon substrates. The $[ZnO/C]_{25}$ sample thickness (where 25 is the number of ZnO/C bilayers) changed from 180 to 240 nm. The bilayer thickness h_{bl} changed from 7,5 to 9,5 nm with the thickness of the C layer being constant ~ 3.5 nm and the thickness of the ZnO layer changed from 4 to 6 nm.

The structure was investigated by X-ray diffraction methods (XRD) on a Bruker D2 Phaser diffractometer ($\lambda_{CuK\alpha1}$ = 1,54 Å) using DIFFRAC.EVA 3.0 software with the ICDD PDF Release 2012 database. Transmission electron microscopy (TEM) images of the cross section and electron diffraction were obtained on a Hitachi HT7700 TEM (acceleration voltage 100 kV, W source). The cross-sectional samples on the sitall substrates for TEM studies were prepared by focused ion beam (single - beam FIB, Hitachi FB2100) at 40 kV to a thickness of about 40-50 nm.

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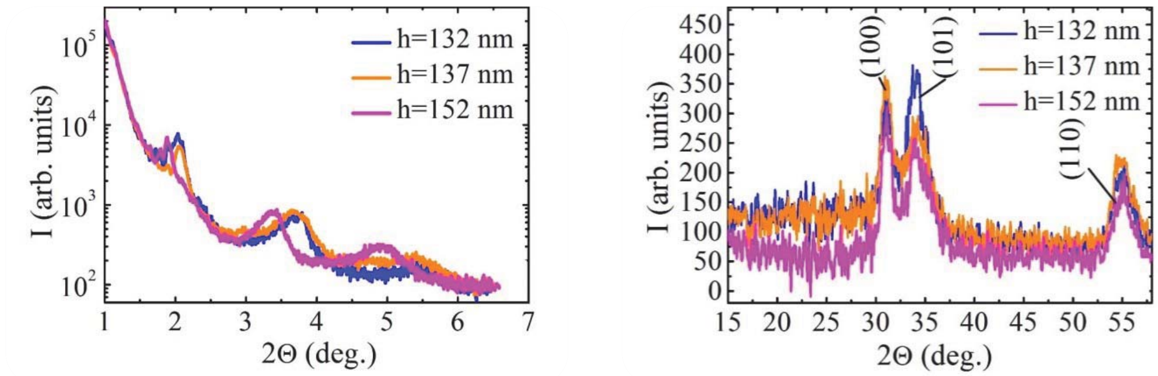
In order to protect the surface from milling by the Ga+ ion beam during sample preparation, a Ge layer was deposited onto $[ZnO/C]_{25}$ film before cross-sectional sample preparation by FIB. Thickness of the samples was measured using the MII - 4 optical interferometer.

The dependence of the resistivity versus magnetic field was taken by DC two point probe technique by means of a V7 - 78/1 multipurpose digital voltmeter. The relative measurement error for the resistivity was no greater than 2 %. Heat treatment was carried out in vacuum at a residual pressure of 10^{-2} Torr, within 30 minutes. The field dependence of magnetization was measured using a SQUID magnetometer (Cryogenic S700X) in the range of 5 T at a temperature of 10 K. The samples were located vertically in the magnetometer, with their long axis oriented to the field.

RESULTS AND DISCUSSION

Analysis of X-ray diffraction patterns (Fig. 1b) of obtained thin films showed that the carbon interlayers have an amorphous structure and the ZnO interlayers have a hexagonal crystal structure. Diffractograms of ZnO/C heterostructures in the region of small angles (1–7 deg.) shows maxima (Fig. 1a), which indicates the formation of a layered structure.



(a) (b)

FIGURE 1. Diffraction patterns of $[ZnO/C]_{25}$ thin films

An analysis of TEM micrographs confirmed the earlier conclusions about the obtained thin films (Fig. 2). Using electron diffraction, phases of crystalline ZnO were also detected, which manifests itself in the form of ring reflections at the electron diffraction patterns (insert in Fig. 2) and amorphous carbon (wide halo).

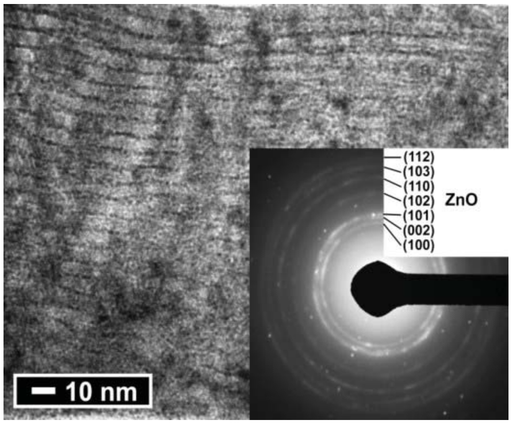


FIGURE 2.TEM micrograph of the cross section of a $[ZnO/C]_{25}$ thin film (h ~ 135 nm). The insets show electron diffraction patterns

We found that in the initial state, multilayer thin films $[ZnO/C]_{25}$ exhibit magnetoresistive properties (Fig. 3). With increasing field strength, the electrical resistance decreases both at room temperature and at the T = 77 K.

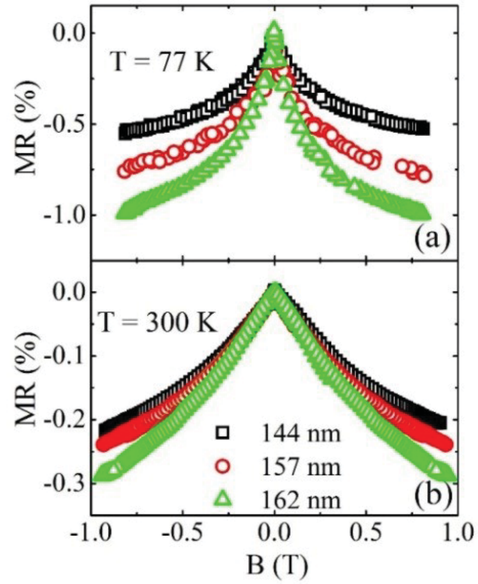


FIGURE 3. The electrical resistivity as a function of the magnetic field strength for the $(ZnO/C)_{25}$ multilayer structures with different thickness measured at T=77 K (a) and T=300 K (b)

Usually, negative magnetoresistance is considered a consequence and a sign of ferromagnetic ordering. However, measurements of magnetization (Fig. 4) did not reveal signs of ferromagnetic ordering even at 10 K, since no traces of hysteresis were detected. This obviously excludes the presence of ferromagnetism in the studied samples at room temperature.

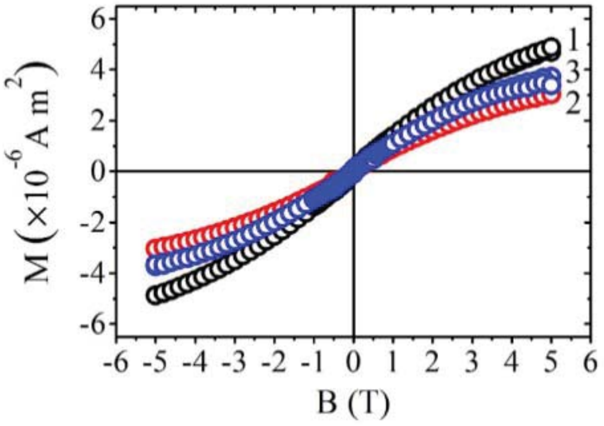


FIGURE 4.The magnetization as a function of the magnetic field for multilayer $(ZnO/C)_{25}$ structures with various thickness: 1–132 nm, 2–136 nm, 3–145 nm and measured at T=10 K

After heat treatment in vacuum no worse than 10^{-2} Torr and a holding time of 30 minutes. the magnetoresistance of the studied samples decreases with increasing annealing temperature (Fig. 5), and heat treatment at 400 ° C leads to the complete disappearance of the negative magnetoresistance even at 77K.

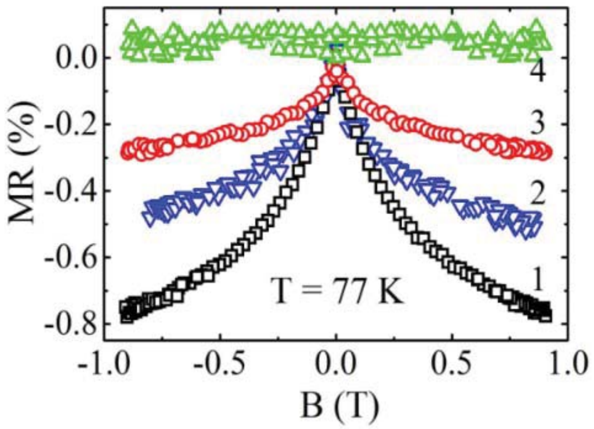


FIGURE 5.The electrical resistivity as a function of the magnetic field strength for the $(ZnO/C)_{25}$ multilayer structure with h = 165 nm measured at T=77 K. 1 – initial state; 2 – annealing at 200 °C; 3 – annealing at 300 °C; 4 – annealing at 400 °C

Let us discuss the obtained results. There are quantum interference and non-interference mechanisms of negative magnetoresistance. The implementation of a non-interference mechanism is unlikely in our case, since a distinctive feature of these mechanisms is a large absolute value (estimated to be - 15%), a weak temperature dependence and a linear dependence on the magnetic field [2]. Interference mechanisms are possible both in the case of hopping conductivity with a variable hop length [3, 4] and in the case of weak localization [5]. An analysis of the temperature dependences of the electrical resistivity measured in the temperature range 77–300 K showed that they satisfactorily straighten in the coordinates ρ ~ lnT (Fig. 6) for samples in the initial state.

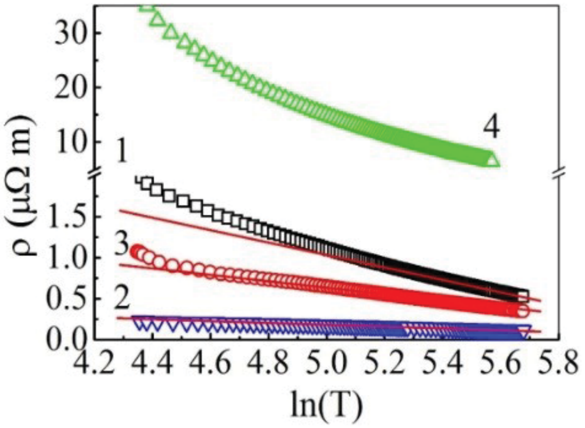


FIGURE 6.Temperature dependence of resistivity for the $(ZnO/C)_{25}$ multilayer structure with h = 165 nm measured at 77–300 K in the ρ-ln(T) coordinates: 1 – initial state; 2 – annealing at 200 °C; 3 – annealing at 300 °C; 4 – annealing at 400 °C

Such a dependence is characteristic of 2D systems in which conditions of weak electron localization are realized and the ZnO–carbon 2D boundaries are apparently the electron transport channels. The thermal treatment of $[ZnO/C]_{25}$ thin films leads to a deviation from the linear temperature dependences of the resistivity at 77–300 K shown in the coordinates ρ ~ lnT (Fig. 6), which is associated with a change in the conductivity mechanism and a transition from weak to strong localization charge carriers. However, this transition is completely carried out only after heat treatment at 400°C, and is probably associated with the blurring of the ZnO–carbon 2D boundaries. Further more detailed studies will help clarify the mechanism of this effect.

CONCLUSIONS

ZnO/C thin films with 25 bilayers and a total film thickness h = 150–170 nm were obtained by layer-by-layer deposition of ZnO and C use by ion beam sputtering technic. The results of X-ray diffraction analysis indicates that the obtained samples of $[ZnO/C]_{25}$ thin films are multilayers with crystalline ZnO and amorphous C layers. A negative magnetoresistance does not related to ferromagnetic properties was detected in multilayer thin-film $[ZnO/C]_{25}$ structures in the temperature range from 77 to 300 K and magnetic field strength less than 1T. Heat treatment of $[ZnO/C]_{25}$ thin-films leads to reduction of magnetoresistance and deviation from linear the temperature dependences of resistivity at 77–300K showed in ρ - ln(T) coordinates. It is attributes to the transition from the weak to the strong localization which is characteristic for 2D systems.

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